SKI Report 02:35

Research

Encyclopaedia of Features, Events and Processes (FEPs) for the Swedish SFR and Spent Fuel Repositories

Preliminary Version

Bill Miller
Dave Savage
Tim McEwen
Matt White

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SKI perspective

Background

In performance assessments of repositories for radioactive waste disposal, a structured approach to describe the repository system and its evolution with time is to use the concept of FEPs (Features, Events and Processes). The system description consists of descriptions of each FEP and how they interact, usually organised with some graphical tool. In order to utilise compilations of FEP descriptions in different performance assessments in a consistent way, a FEP Encyclopaedia is required.

In the SKI performance assessment project SITE-94, a FEP list for a repository for spent nuclear fuel was developed. The list was then used to construct the PID (Process Influence Diagram) that shows how the FEPs are influencing each other, i.e., the description of the system. The FEP descriptions were however not sufficiently detailed to constitute a FEP Encyclopaedia.

Purpose of the project

The purpose of this project is to:

- Update and develop the FEP descriptions from SITE-94.
- Extend the list to include FEPs to be used in constructing the PID for SFR-1 (Final Repository for Radioactive Operational Waste).

The latter task is part of the preparatory work conducted for the SKI review of SKB’s renewed safety assessment for SFR-1 (the SKB SAFE project).

Results and continued work

The outcome of the project is a catalogue with descriptions of FEPs, relevant to the Swedish repository for low and intermediate level wastes (SFR-1) and the proposed Swedish repository for spent nuclear fuel.

The current version of the FEP catalogue is marked ”Preliminary Version”. It has been reviewed by both SKI staff and experts working in SKI research projects, but more extensive review will be performed in connection to further use of the FEP catalogue in future performance assessment projects.

To be a useful tool the FEP Encyclopaedia needs to be a living document that is regularly updated. For example there may be necessary to consider new scientific knowledge, changes in performance assessment methodology or changes in repository concepts. Also the required level of detail for the FEP descriptions may change during the process of developing the repository concept.

Project information

Responsible for the project at SKI has been Christina Lilja.
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Preliminary Version

Bill Miller
Dave Savage[1]
Tim McEwen[2]
Matt White[3]

QuantiSci Ltd
47 Burton Street
Melton Mowbray
Leicestershire
LE13 1AF
United Kingdom

August 2002

[1] now at Quintessa Ltd

[2] now at SAM Ltd

[3] now at Galson Sciences Ltd

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1 Introduction

This is an ‘Encyclopaedia’ providing descriptions of Features, Events and Processes (FEPs) that are relevant to the Swedish repository for low and intermediate-level wastes (the SFR) and the proposed Swedish repository for spent fuel.

Although the FEPS and their descriptions found in this encyclopaedia are specific to these two repository concepts, many of the descriptions will also be relevant to other repository designs and concepts, although they have not been written to be inclusive of the features of other repositories. As such, this encyclopaedia may be of interest to a wide range of individuals and organisations involved in repository safety assessment around the world.

The purpose of this encyclopaedia is to describe, in qualitative terms, the various FEPs which have been identified as being relevant to the two Swedish repository designs. These descriptions may be used in a variety of ways. One important role will be to support quantitative performance assessments (PAs) by describing the conceptual understanding of the various components of the repository (e.g. the barriers, the rock and the groundwater) and their evolution: this conceptual understanding is crucial because it is the foundation upon which the mathematical analysis is based.

The descriptions have been written at a level of detail appropriate for a scientifically literate reader without specialist knowledge of radioactive waste disposal technology or assessment procedures. As such, the descriptions avoid the use of specialist terms, acronyms and equations. Many of the FEP descriptions relate to issues which are the focus of ongoing research and, thus, they reflect the current state of knowledge and may require updating at regular intervals, either to include more recent technical information or procedures for treating the FEP in mathematical safety assessment.

1.1 Background

When attempting to predict the future behaviour of a repository for radioactive wastes, it is sensible to use a systematic approach to dealing with all the FEPs that could occur and which might influence repository safety. A commonly adopted procedure is known as the ‘Sandia Methodology’ (Cranwell et al., 1990) which, in simple terms, involves defining all potential FEPs that may affect the performance of the disposal system, classifying and screening them for relevance, and then combining them into scenarios by means of identifying related groups of FEPs.

In 1988, both SKI and SKB embarked on a joint exercise to try to achieve consensus on the principles for scenario selection for a Swedish repository for spent fuel. This joint exercise (Andersson, 1989) was built on the Sandia Methodology and began by constructing a list of FEPs considered relevant to a deep spent fuel repository in granitic rocks in Sweden. One of the outcomes of the joint SKB/SKI exercise, was the development of the ‘Process System’ which is an ordered assembly of those FEPs which might have to be considered, at some level of detail, in PA calculations in all scenarios.

In 1994, SKI developed further the Sandia Methodology in the SITE-94 PA (Chapman et al., 1995) and introduced the concept of the ‘Process Influence Diagram (PID)’ which is a graphical representation of the many FEPs considered relevant to the disposal system together with the potential influences that may occur between them.

The use of a PID was found to be a constructive and informative approach to thinking about the evolution of the repository. In particular, it forces consideration of how the action of one FEP may affect the behaviour of the overall disposal system, and specific components of it, through its influence on other FEPs. However, evaluation of these influences requires that the definition and scope of each individual FEP is commonly agreed. There was, therefore, a requirement for definitions to be written for each FEP. This was the original impetus for writing this encyclopaedia.

1.2 Approach

The first step in the approach to compiling the encyclopaedia was to agree on a list of FEP names to include. Originally, this encyclopaedia was planned to cover FEPs of relevance to the Swedish spent fuel repository. Consequently, the list of FEPs from the SITE-94 Reference Case and Central Scenario was used as a starting point (Appendix 1 in Chapman et al., 1995). However, after initiation of the project, it was agreed that the focus of the work would be expanded to include also the repository for low and intermediate-level radioactive wastes at Forsmark (the SFR).

The SITE-94 FEP list is not appropriate to the SFR because of the different wastes, engineered barrier materials and near-field designs of the two repositories. Consequently, a second FEP list for the SFR was drawn-up from FEPs considered in an ongoing project to develop a PID for the SFR repository (Appendix C in Stenhouse et al., 1998).

Comparison of the FEP lists for the spent fuel and SFR repositories shows that a number of FEPs are common to both, whilst others are specific to their respective repository designs. Consequently, the two FEP lists were merged and reorganised such that the final FEP list (Table 1) contains the following 3 types of FEPs:

1) GEN - those FEPs which are general and apply to both the spent fuel and SFR repositories. These mostly relate to processes that occur in the far-field rock (e.g. groundwater flow) and to those fundamental mechanisms which are independent of repository design (e.g. radioactive decay).

2) SFR - those FEPs which are specific to the SFR repository. These generally relate to issues affecting the wasteform and the cementitious materials used in the near-field (e.g. degradation of the cement matrix).

3) SFL - those FEPs which are specific to the spent fuel repository. These generally relate to issues affecting the spent fuel, the canister and the bentonite buffer and backfill (e.g. corrosion of the copper shell of the disposal canister).

1.3 Descriptions

Each FEP description is typically 3 or 4 pages long and has been written in a standard format with the following common elements:

Number: A unique number in the encyclopaedia, indicating whether it is a GEN, SFR or SFL type of FEP.

Name: Descriptive name for the FEP.

Short description: A one or two sentence long description of the FEP that gives the basic definition of the term.

Technical description: A longer description that provides information on the technical details of the FEP, its implication to repository safety, information on research into the FEP and how it may be modelled in PA.

Origin in the repository system: Short discussion of how (and when) the FEP occurs in the repository and which other FEPs have an influence over the FEP under discussion.

Impact on the repository system: Short discussion of how the FEP under discussion affects the repository system, including those FEPs on which it may have a direct impact.

Bibliographic references: Recent published references relating to the technical aspects of the FEP, its consideration in PA and modelling.

Equivalent FEPs: How the FEP under discussion relates to FEPs in other FEP lists, in particular the NEA international FEP list (NEA, 1998).

Production: A note of the authors of the FEP description and date of writing.

Table 1: The number and names of the FEPs included in the encyclopaedia.

Number	**Name**
General FEPs:	
GEN-1	Alteration and weathering along flowpaths
GEN-2	Anion exclusion
GEN-3	Cave-in
GEN-4	Colloid behaviour in the host rock
GEN-5	Creeping of the rock mass
GEN-6	Groundwater salinity changes
GEN-7	Degradation of the borehole and shaft seals
GEN-8	Degradation of the rock reinforcement and grout
GEN-9	Diffusion
GEN-10	Radionuclide dispersion
GEN-11	Distribution and release of radionuclides from the far-field
GEN-12	Earth tides
GEN-13	Electrochemical effects
GEN-14	Enhanced rock fracturing
GEN-15	Excavation effects on the near-field rock
GEN-16	External flow boundary conditions

Number	Name
GEN-17	Faulting
GEN-18	Gas flow in the far-field
GEN-19	Gas generation in the far-field
GEN-20	Gas generation in the near-field rock
GEN-21	Glaciation
GEN-22	Far-field groundwater chemistry
GEN-23	Groundwater flow
GEN-24	Interfaces between different waters
GEN-25	Matrix diffusion
GEN-26	Permafrost
GEN-27	Radionuclide precipitation and dissolution
GEN-28	Radioactive decay
GEN-29	Erosion and weathering
GEN-30	Radiolysis
GEN-31	Radionuclide reconcentration
GEN-32	Hydraulic resaturation of the near-field rock
GEN-33	Sea level changes
GEN-34	Radionuclide sorption
GEN-35	Fast transport pathways
GEN-36	Stress field
GEN-37	Surface water chemistry
GEN-38	Temperature of the far-field
GEN-39	Uplift and subsidence
FEPs specific to the SFR repository:	
SFR-1	Colloid generation in the waste package
SFR-2	Colloid generation in the shell and grout
SFR-3	Degradation of the cement mortar and the silo shell
SFR-4	Degradation of steel reinforcements in the silo shell
SFR-5	Degradation of the concrete packages and the cement matrix
SFR-6	Degradation of the bitumen matrix
SFR-7	Degradation of the inorganic waste
SFR-8	Degradation of the organic waste
SFR-9	Diffusion in the near-field
SFR-10	Colloid filtration in the near-field
SFR-11	Gas generation in the repository
SFR-12	Gas flow in the near-field
SFR-13	Mechanical impact on the engineered barriers
SFR-14	Radiation effects in the near-field
SFR-15	Radionuclide release from the waste
SFR-16	Hydraulic resaturation of the near-field
SFR-17	Temperature of near-field
SFR-18	Radionuclide release from the waste package
SFR-19	Transport and release from the silo
SFR-20	Groundwater chemistry in the near-field
SFR-21	Groundwater movement in the near-field
SFR-22	Changes in the radionuclide inventory
SFR-23	Groundwater chemistry in the near-field rock
SFR-24	Evolution of the bentonite layer in the silo

FEPs specific to the spent fuel repository:

SFL-1	Swelling of the bentonite buffer
SFL-2	Changes in the spent fuel radionuclide inventory
SFL-3	Chemical alteration of the buffer and backfill
SFL-4	Coagulation of bentonite
SFL-5	Colloid behaviour in the buffer and backfill
SFL-6	Colloids and particles in the canister
SFL-7	Corrosion of the copper shell
SFL-8	Corrosion of the metal non-fuel waste parts
SFL-9	Corrosion of the cast iron insert
SFL-10	Canister corrosion prior to wetting
SFL-11	Creeping of the metal in the canister
SFL-12	Criticality
SFL-13	Degradation of the spent fuel elements
SFL-14	Differential thermal expansion and contraction of the near-field barriers
SFL-15	Diffusion in and through the canister
SFL-16	Dilution of the buffer and backfill
SFL-17	Erosion of the buffer and backfill
SFL-18	Failure of the copper shell
SFL-19	Failure of the cast iron insert
SFL-20	Groundwater flow through the buffer and backfill
SFL-21	Spent fuel dissolution and conversion
SFL-22	Gap and grain boundary release
SFL-23	Gas escape from the canister
SFL-24	Gas flow through the buffer and backfill
SFL-25	Gas generation in the canister
SFL-26	Gas generation in the buffer and backfill
SFL-27	Radionuclide accumulation at the spent fuel surface
SFL-28	Radionuclide interaction with corrosion products
SFL-29	Internal gas pressure
SFL-30	Mechanical impact on the canister
SFL-31	Mechanical impact on the buffer and backfill
SFL-32	Microbial activity
SFL-33	Movement of the canister in the buffer
SFL-34	Preferential transport pathways in the canister
SFL-35	Radiation effects on the buffer and backfill
SFL-36	Radiation effects on the canister
SFL-37	Radiolysis inside the canister prior to wetting
SFL-38	Redox fronts
SFL-39	Reduced mechanical strength of the canister
SFL-40	Radionuclide release from the spent fuel matrix
SFL-41	Radionuclide release from the metal non-fuel parts
SFL-42	Hydraulic resaturation of the buffer and backfill
SFL-43	Sedimentation of the buffer and backfill
SFL-44	Soret effect in the buffer and backfill
SFL-45	Swelling of the tunnel backfill
SFL-46	Temperature of the near-field
SFL-47	Thermal degradation of the buffer and backfill
SFL-48	Total radionuclide release from the spent fuel elements
SFL-49	Radionuclide release and transport from the canister

SFL-50	Radionuclide release and transport from the buffer and backfill
SFL-51	Expansion of solid corrosion products
SFL-52	Evolving water chemistry in the canister
SFL-53	Evolving water chemistry in the buffer
SFL-54	Evolving water chemistry in the backfill
SFL-55	Evolving water chemistry in the near-field rock
SFL-56	Water turnover in the copper shell
SFL-57	Water turnover in the cast iron insert

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Chapman NA, Andersson J, Robinson P, Skagius K, Wene C-O, Wiborgh M and Wingefors S (1995) Systems analysis, scenario construction and consequence analysis definition for SITE-94. SKI Technical Report, 95:26.

Cranwell RM, Guzowski RV, Campbell JE and Ortiz NR (1990) Risk methodology for geologic disposal of radioactive waste: scenario selection procedure. Sandia National Laboratories, NUREG/CR-1667. [Revised from an original document published in 1982].

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Number: **GEN-1**

Name: **ALTERATION AND WEATHERING ALONG FLOW PATHS**

Short description:

Chemical (water-rock) reactions between flowing groundwater and the rock and any fracture minerals will lead to progressive changes to the solid phases along the flow path and to its hydraulic properties. These water-rock reactions can impede or enhance radionuclide transport depending on their nature.

Technical description:

Groundwater can react with the adjacent rock, and any fracture-coating and infilling minerals causing alteration and weathering of the groundwater flow paths. This can occur both in the near and far-fields. Rock-water reactions generally occur when the system is out of chemical equilibrium and this situation could arise from perturbations caused by the presence of a repository or by the rapid influx of chemically different waters from the surface.

Under natural geological conditions, alteration and weathering processes at depth are generally slow at ambient temperatures and require geological timescales for significant changes to the physical and chemical properties of the rocks to take place. However, severe and more rapid alteration to the rock might be experienced around a repository because the local groundwater chemistry will be significantly altered by reaction with the waste and the engineered barrier system materials. These altered groundwaters, when they flow out of the engineered barriers, may be far from chemical equilibrium with the host rock and will react with them. This will be particularly the case for the SFR where the large volumes of cement will cause the groundwater to become very alkaline (see SFR-23 “Groundwater chemistry in the near-field rock”). Also, in the spent fuel repository, the higher temperatures and temperature gradients occurring after repository closure will impact on the equilibrium between the groundwater and the rock, where maximum temperatures are reached a few hundred years after closure (see SFL-46 “Temperature of the near-field”).

In the upper part of the host rock (near surface), infiltrating groundwater is usually out of chemical equilibrium with the rock. Changes to the groundwater recharge system, such as variations in recharge rates, volumes, locations and depths, can be brought about by climate change. If recharge increases (e.g. as a consequence of glaciation), it is possible that fresh, oxidising waters would be able to penetrate deeper into the far-field rock than they do at the present time. These waters would then be out of chemical equilibrium with the far-field rock and would react with them. Scenarios such as these are considered in a number of performance assessments.

Many water-rock reactions will involve the replacement of thermodynamically unstable minerals by hydrous minerals stable at low temperature and pressure. This alteration may liberate radionuclides already sorbed on the existing minerals, which would be a non-conservative process. However, the newly formed minerals may themselves incorporate radionuclides in their mineral structure making them inaccessible to the flowing groundwater.

In addition, the neoformed hydrous alteration products generally are more efficient at sorbing radionuclides to their surfaces than the minerals they replace. However, they may be efficient sorbents for only a limited time as they are gradually transformed to more stable crystalline phases.

Other water-rock reactions will involve dissolution and precipitation if the solubility of fracture filling mineral phases changes due to variations in the groundwater temperature or chemistry (Eh, pH and concentrations of dissolved species). For example, an increase in temperature in the near-field of the spent fuel repository due to the early thermal peak may cause increased dissolution of silicate minerals and a subsequent precipitation of silica (potentially as colloids) downstream from the repository. Calcite, on the other hand, which has a lower solubility at higher temperatures, may precipitate closer to the repository. Likewise, the introduction and mixing of chemically distinct groundwaters from the near-surface along fast pathways (see GEN-35 “Fast transport pathways”) may cause more rapid dissolution and precipitation of minerals in the fractures.

Both mineralogical transformation and dissolution-precipitation reactions will result in changes to the hydraulic properties of both the accessible bulk rock and any fractures present. Changes in rock porosity will be linked to the groundwater flux through the rock and to volumetric changes associated with dissolution and precipitation reactions. Changes in hydraulic conductivity and fracture transmissivity will be linked to changes in porosity. Relatively minor changes in porosity may cause large changes in hydraulic conductivity. Consequently, alteration of fracture surfaces may have an effect on the fraction of rock available for flow, sorption and matrix diffusion. The impact on repository safety may thus be positive or negative depending on whether the water-rock reactions enhance or impede groundwater flow.

Rock deformation due to stress changes (e.g. caused by the thermal load or glaciation) may also affect porosity. These changes will be most marked in lithologies which contain higher proportions of minerals thermodynamically unstable at low temperatures and pressures. Neoformed minerals will be most abundant in zones of enhanced water flow. Thus alteration and weathering of rock minerals has the potential to heal existing and newly formed fractures.

Water-rock reactions will be controlled by groundwater residence time, the rate of mineral-water reaction and temperature. In some cases, groundwaters never reach chemical equilibrium due to slow reaction rates and the inherent thermodynamic instability of the mineralogy concerned. Silicate mineral dissolution reactions are generally slow (in the range 10^{-1} to 10^{-7} mol m^{-2}/yr at 25°C) and silicates are poorly soluble (of the order of 10^{-3} to 10^{-5} g/l). Some minerals may never reach equilibrium and will exist only due to their slow kinetics and limited contact with the water.

Dissolution and precipitation reactions are driven by departure from thermodynamic equilibrium. In water-rock systems, departure from chemical equilibrium is governed by groundwater flow and changes in temperature. Flow of groundwater will redistribute solutes and thus potentially perturb chemical equilibria between minerals and aqueous solutions. A thermodynamic description of a water-rock system only defines whether a solid will tend to dissolve or precipitate. The rate at which a solid reacts with a fluid may be more relevant. Rate limiting mechanisms are of two types (Berner, 1978): 'transport-controlled' kinetics where dissolution is limited by the rate of transport of solutes away from the dissolving crystal and 'surface reaction-controlled' kinetics where dissolution is limited by the rate of detachment of ions from the surface of the crystal.

The applicable rate limiting mechanism can be estimated from the solubility of the mineral concerned. Below a solubility of about 10^{-4} moles/l, surface reaction control dominates. This would include all silicates and carbonates in systems where pH > 4 (Lasaga, 1984). The dissolution behaviour of evaporite minerals such as halite, gypsum, anhydrite etc. should be transport-controlled. In contrast, the rate of dissolution of silicates should be dominated by the intrinsic dissolution rate constant and should be relatively unaffected by the groundwater flow rate. Using solubility data from Berner (1978) one volume of calcite would require approximately 10^5 volumes of freshwater for complete dissolution and one volume of feldspar would require approximately 10^7 volumes of freshwater for complete dissolution.

Alteration and weathering along groundwater flow paths can be predicted using thermodynamic equilibrium models and databases. However, these models are limited in their ability to treat kinetics and solid-solution mineral series.

Origin in the repository system:

The alteration of groundwater flowpaths will occur in the near-field rock by repository-induced processes such as the early thermal pulse causing chemical disequilibrium between the groundwater and by changes to the near-field groundwater composition due to reaction with the engineered barrier system materials. Alteration and weathering along groundwater flow paths will occur in the far-field rock by natural, time-dependent processes, such as climate change driven variations in the volume and composition of recharge groundwaters.

Impact on the repository system:

Rock-water interactions may change the radionuclide transport properties of the near-field and far-field rock by potentially changing the hydraulic properties (groundwater flow system) of the bulk rock mass and any fractures that may be present.

In addition, rock-water interactions can fix radionuclides into (or liberate radionuclides from) the mineral lattice and surface sorption sites. In general the neoformed alteration minerals have better sorptive properties than the minerals they replace.

Bibliographic references:

Berner RA (1978) Rate control of mineral dissolution under earth surface conditions. American Journal of Science, 278, 1235-1252.

Lasaga AC (1984) Chemical kinetics of water-rock interactions. Journal of Geophysical Research, 89, 4009-4025.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.2.05 (Contaminant transport path characteristics in geosphere) and 2.2.08 (Chemical/geochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEPs 4.1.7 (Thermochemical changes) and 6.6 (Weathering of flow paths) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

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Number: **GEN-2**

Name: **ANION EXCLUSION**

Short description:

Anion exclusion is an electrostatic phenomenon whereby anionic species in solution are repelled from mineral surfaces by their negative electrical charge. In fine grained materials, such as a clay-rich buffer, anion exclusion can be an effective retardation process.

Technical description:

Due to the atomic structures of many silicate minerals, their surfaces are generally negatively electrically charged. This phenomenon is particularly pronounced for the flat, ‘sheet-like’ phyllosilicate minerals such as clays and micas. As a consequence, negatively charged anionic species and negatively charged complexes in solution in ground and porewaters (and suspended colloids also with negatively charged mineral surfaces) are electrically repelled from solid mineral surfaces in rock or in mineral-based materials, such as compacted bentonite. This electrostatic repulsion phenomena is known as ‘anion exclusion’.

This repulsion force arises because cations may be released from interlayer sites in the mineral structure, resulting in a negatively charged silicate framework, surrounded by a diffuse cloud of cations. This charged surface and cloud of ions is called a double layer. The double layer consists of ions attached to the mineral surface (the ‘fixed’ or ‘Stern’ layer), whereas outside that lies the ‘diffuse’ or ‘Gouy layer’ in which ions are free to move. In smectite clays, the surface charge results from substitutions in the silicate framework, so the charge is effectively independent of pH. In a compacted clay such as bentonite, the Gouy layers on adjacent particles overlap, so that anions will be effectively excluded from the pores.

If a pressure gradient is applied to the system, water molecules are able to move through the pores, but negatively charged dissolved species cannot. The clay can thus act as a semi-permeable membrane allowing the passage of water but not dissolved anionic species. This process is also known as ‘membrane filtration’ (Hanshaw and Coplen, 1973).

Anion exclusion is thus dependent on the characteristics of the pores in the solid material (e.g. clay-rich rock or bentonite), the ionic strength of the solution and the charge of the diffusing species. Experiments examining the diffusion of anionic species through compacted bentonite have shown that the diffusion flux is dependent on the density of the bentonite, the electrolyte concentration in the porewater and the charge of the anionic species. At high bentonite densities and low electrolyte concentrations, the pore volume accessible to anionic species, the effective porosity, is smaller than at lower densities and higher electrolyte concentrations.

Anion exclusion can affect the diffusive mass transport of species in solution. When anion exclusion does occur, its likely impact will be to lower the diffusion flux (retard) anionic species, with respect to the flux of cationic species. Consequently anion exclusion should be considered when assigning diffusivity values to diffusing anionic species.

In the repository environment, anion exclusion theoretically can cause retardation of radionuclides in anionic form in solution (or bound to negatively charged colloids). However, since the electrical repulsion forces act over very small distances, anion exclusion is only likely to cause retardation in clay-rich materials with small pore spaces. The most likely parts of a repository where this may occur are (i) a clay-rich buffer or backfill, (ii) a clay-rich host sedimentary rock, or (iii) in small aperture microfractures in crystalline rock that are filled or lined with clay minerals due to alteration of the rock matrix (see GEN-1 “Alteration and weathering along flow paths”).

In the bentonite buffer or backfill of the spent fuel repository, anion exclusion may occur once it has resaturated and swelled to form a very low permeability material. If the bentonite buffer becomes disrupted so that advective flow could occur through it, then anion exclusion would no longer be a significant retardation process in the buffer.

In the cementitious near-field of the SFR repository, modelling results suggest anion exclusion could still occur, although, perhaps, to a lesser extent.

In clay-rich, sedimentary host rocks, anion exclusion could occur only if anions were able to migrate from the near-field and it would then only be a significant retardation process if transport through the clay-rich rock was dominated by diffusion. To neglect anion exclusion in clay materials in performance assessment would be a conservative assumption.

In microfractures in crystalline rock, anion exclusion could occur if there was groundwater migration from advecting fractures (e.g. matrix diffusion) and the microfractures were substantially filled with clay materials.

Anion exclusion is generally not taken into consideration in performance assessments, and was not considered in the SITE-94 performance assessment. However, two models for anion exclusion were considered by Nirex in their Nirex 97 performance assessment of a deep cementitious repository for low and intermediate-level wastes (Nirex, 1997). The two models were (i) an anion exclusion factor applied to all anions and all negatively charged organic complexes, and (ii) an anion exclusion factor applied to all anions but not to negatively charged organic complexes.

Origin in the repository system:

Anion exclusion can occur in a low permeability clay-rich media (e.g. the buffer, backfill or clay-rich far-field rock) where advection does not occur, where a chemical gradient exists to drive diffusion and where anions are in solution.

Anion exclusion is thus controlled by the physical characteristics of the transport medium (pore space sizes and mineral surface charge); the chemistry of the porewater; and the presence of anionic species releases from the wasteform.

Impact on the repository system:

If anion exclusion operates in low permeability clay-rich materials (rock or buffer), then it will result in a decrease in the diffusive transport of anionic species. Therefore it will act as a retardation process and will cause a change in the trace-element (anionic dissolved content) chemistry of the near-field pore and groundwaters and potentially cause an accumulation of retarded anions in the clay material. A change in the near-field pore and groundwater chemistry may have an impact on the corrosion and degradation of the near-field barriers.

If anion exclusion operates in clay-filled microfractures in crystalline rock, such that it reduces the inflow of anions to the rock matrix from advecting fractures, this would have the effect of limiting the potential for dispersion of anions in the crystalline host rock by matrix diffusion. That is, anion exclusion in microfractured rock can reduce the diffusivity and available porosity. Neglecting this process in performance assessment would not, necessarily, be a conservative assumption.

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Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Science Report, S/97/012. (Four volumes plus overview).

Kim H-T, Suk T-W and Park S-H (1993) Diffusivities for ions through compacted Na-bentonite with varying dry density. Waste Management, 13, 303-308.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics), 2.2.05 (Contaminant transport path characteristics in geosphere), 2.2.08 (Chemical/geochemical processes and conditions in geosphere) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

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Number: **GEN-3**

Name: **CAVE-IN**

Short description:

Cave-in or collapse of deposition holes, caverns, tunnels or access shafts in a repository could take place under certain circumstances. This, in turn, might change the properties of the surrounding rock and the engineered barriers leading to earlier or faster radionuclide release. Cave-in is, therefore, of potential significance in performance assessment.

Technical description:

Cave-in of the excavated parts of the repository is possible if the rock is not self-supporting and if the buffer, backfill and any engineering-supports are not sufficiently strong to hold-up the rock mass. It represents an extreme example of loss of mechanical strength of the rock mass and is only possible under certain circumstances, such as when fracturing of the rock mass is sufficient to allow movement of blocks of rock to take place or when this movement is made possible by some change in the properties or continuity of the near-field materials and supports.

There is a complete spectrum of possible damage to the rock mass, from minor adjustments on a single fracture to total collapse of an underground structure. Small movements of the rock can occur in the engineered damaged zone and can be caused by creeping of the rock mass (see GEN-5 "Creeping of the rock mass"). This FEP is concerned with extensive, but not necessarily total collapse, of a part of an underground opening anywhere in the repository.

Unless sufficient space is available in the repository near-field, significant cave-in could not take place, since a prerequisite for such a process to occur is the dilation of the rock mass. If space is available, cave-in will be exacerbated by an increase in the number or size of fractures, and displacements along them. This could be caused by repeated cycles of glacial loading (e.g. Hansson et al., 1995), by thermally-induced stresses, by the swelling of materials in the near-field (e.g. bentonite) or by time-dependent decreases in the strength of the rock mass due to chemical processes.

Fracturing will also tend to be increased where a high contrast exists in the value of the deformation modulus between the rock mass and the backfill. In places where rock support has been required during repository construction, e.g. when a tunnel intersects a fault zone, the support (which is likely to consist of rock bolts, mesh, shotcrete etc.) will degrade over time and this could allow movement to take place on fractures that had previously been held securely. Chemical changes to the engineered barriers and reinforcements could alter the support provided by these materials to the rock mass.

Thus, in order for a cave-in to occur, it would be necessary for substantial changes to have taken place in the support provided to the rock surrounding a part of the repository and for sufficient space to have been made available. A reduction in rock support on its own, e.g. if rock bolts were to lose their effectiveness, would be insufficient to allow a cave-in to occur. In this case, fractures could suffer some displacement and may show some dilation but insufficient additional space would not allow the rock mass to collapse.

In the spent fuel repository, the buffer within the deposition hole will, under the majority of chemical and hydrogeological conditions expected in the repository, provide considerable support to the wall of the deposition hole, and the thickness of bentonite between the rock and the canister and the strength of the canister itself will prevent the possibilities for cave-in. Unless a significant proportion of this bentonite is removed from a deposition hole, or unless the canister degrades to such an extent that its volume is sufficiently reduced, it seems unlikely that a reduction in the swelling capacity of the bentonite on its own would allow cave-in of the hole to occur.

A tunnel or an access shaft has an increased possibility of suffering a cave-in, since these parts of the repository are more likely to intersect fracture zones with substantially higher transmissivities than the deposition holes, which will be selected so as to minimise fracture intersections. There is also a chance of movement on these larger fracture zones. Thus, there is an increased likelihood of damage, either to the backfill, to the rock support systems or to the rock mass adjacent to the deposition area. Nevertheless, cave-in will, again, only occur if sufficient space is available and this seems likely to occur only when a sufficient volume of the backfill has been removed by erosion (see SFL-17 “Erosion of the buffer and backfill”) or when movement on a fault or fracture zone has resulted in considerable dilation of the rock mass (see GEN-17 “Faulting”).

In the SFR, cave-in of the silo is unlikely because the reinforced silo structure and the cementitious backfill will act to support the rock. Cave-in of the rock vaults is possible because they will be backfilled only loosely with sand and crushed rock with no swelling pressure. However, the radiological impact of cave-in of the rock vaults would be limited because the majority of the activity in the SFR is located in the silo.

The implications of a cave-in are dependent on where it takes place. The closer it is to the waste the more significant it is likely to be. The potential effects of a cave-in on the performance of a repository are an increase in the hydraulic conductivity of the rock mass in the vicinity of the cave-in, increased channelling of groundwater flow that could result in there being further damage to the backfill or, in an extreme case, damage to a waste packages, although this is considered unlikely.

Origin in the repository system:

Cave-in of all or a part of the repository excavations might occur if the rock mass is fractured and sufficient void space is available to allow movement of the individual rock blocks. Given that most void spaces will be filled by a buffer or a backfill during normal repository closure operations, this void space can only occur by extensive degradation, collapse and removal of the engineered barrier materials.

Impact on the repository system:

Extreme cave-in occurring close to the waste might impact on the mechanical stability of the engineered barriers and may lead to early failure of the waste packages, resulting in early or enhanced radionuclide release, although this is considered unlikely.

Cave-in is most likely to affect the hydraulic characteristics of the rock mass at the site of the collapse. Hydraulic conductivity and the extent of channelled groundwater flow are both likely to be enhanced by cave-in.

Bibliographic references:

Hansson U, Shen B, Stephansson O and Jing L (1995) Rock mechanics modelling for the stability and safety of a nuclear waste repository. SKI Technical Report, 95:41.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Shen B and Stephansson O (1990) Modelling of rock mass response to repository excavation, thermal loading from radioactive waste and swelling pressure of bentonite. SKI Technical Report, 90:12.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics), 2.1.07 (Mechanical processes and conditions in wastes and EBS), and 2.2.06 (Mechanical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-4**

Name: **COLLOID BEHAVIOUR IN THE HOST ROCK**

Short description:

Colloids may be generated by chemical, physical and microbial processes in the host rock in both the near and far-fields, and can be transported by advecting groundwater. Radionuclides can sorb onto colloids thus providing a mechanism to increase their transport rates.

Technical description:

Colloids are small, solid particulate materials suspended and dispersed in groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed ‘suspended particles’ and anything smaller is considered to be in solution.

In natural deep groundwaters, colloids are always present and generally have concentrations of less than 1 mg/l, and they can be of both inorganic and organic origin. Inorganic colloids can form from alteration or physical erosion of the rock and fracture minerals or by direct precipitation in suspension. The latter form are commonly associated with chemical gradients or ‘fronts’ in the groundwater leading to precipitation; for example, as a result of changes in redox potential or pH.

Common forms of inorganic colloids are silica, clay materials and Fe-oxyhydroxides. Temperature changes can also cause colloid formation due via change in chemical equilibria and reaction rates. Under extreme external conditions (e.g. glaciation or faulting) transients in inorganic colloid concentration might occur.

Organic colloids may comprise small fragments of degrading organic material (or dead individual microorganisms) or they may be organic macromolecules, such as humic and fulvic acids. Organic colloids are generally thought to have a surface origin and to be transported to depth by infiltrating groundwaters. Organic materials can form coatings on inorganic colloids which may make them more stable and potentially more mobile.

The presence of a repository is likely to increase the concentration and range of colloids above that occurring naturally due to the excavation works (repository construction), and post-closure degradation of the wasteform and the engineered barrier system materials. Colloids formed in the near-field may be transported to the host-rock by advecting groundwater where they add to the natural groundwater colloid population.

Colloids are stable in fluids of constant chemical composition, that that are not affected by particle flocculation and settling. Colloids remain in suspension due to mutual charge repulsion and Brownian motion (Stumm and Morgan, 1996) and may be transported with groundwater at any velocity. Depending on composition and physico-chemical characteristics (e.g. size distribution, surface potential etc.) colloids are transported more or less with the same velocity as the groundwater.

The significance of colloids arises from the fact that they can sorb radionuclides and are mobile. This means that colloids can increase the apparent concentration in groundwater of radionuclides with low true solubilities. In addition, because of their very small size, they provide a very large surface area and thus, volume for volume, they may sorb radionuclides more efficiently than the surrounding rock mass. The transport and retardation (and dispersion) of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution.

Within fractures, mobile colloids tend to stay in the centre of the stream where the flow is highest. In addition, the colloids are generally excluded from the rock pores because of size and anionic exclusion processes which means that the effectiveness of matrix diffusion and sorption as retardation mechanisms is reduced. Thus, if the colloids are free to move, colloid-bound radionuclides proceed at a faster average flow rate than those in solution (peak releases are earlier and higher).

Colloids may themselves be immobilised by a number of processes, causing the retardation of radionuclides sorbed onto them. If the pore spaces in the flowing porosity are very small, then colloids may be physically filtered from the advecting groundwater. Also, colloids can sorb onto exposed mineral surfaces of the rock and fracture fills.

A key question with regard to the importance of colloids for radionuclide transport relates to whether radionuclide sorption onto them is reversible or irreversible. Rapid reversible sorption will result in lesser impacts than irreversible sorption because, in the latter case, a greater proportion of the transport behaviour is defined by the properties of the colloids rather than the solute. A related question concerns the stability of the colloids. Short-lived unstable colloids potentially are of lesser importance than long-lived stable colloids for the same reason.

Due to the potential significance of colloids for affecting radionuclide transport and retardation in the near and far-field host rock, it is important to be able to model colloid behaviour. To date, there have been few successful attempts to integrate a dynamic colloid migration model with a conventional flow-transport code in performance assessment. Most performance assessments do not include a detailed, quantitative evaluation of the impact of colloids upon far-field radionuclide transport. The degree to which mechanistic models of chemical transport may be included in performance assessment is limited by our understanding of chemical processes, data constraints and representation of the heterogeneity of the natural system.

Origin in the repository system:

In the near and far-field host rock, colloid populations are controlled the water composition (e.g. ionic strength, Eh, pH), flow rate and the nature of colloid-generating materials. In particular, the presence of chemical gradients and interfaces between different water bodies can lead to direct colloid formation by precipitation and by weathering and alteration of rock and fracture minerals. Increased colloid populations may be generated in the near-field by wasteform dissolution and degradation of the engineered barrier system materials.

Impact on the repository system:

Colloids can sorb radionuclides thus increasing their apparent solubility. Once sorbed to colloids, radionuclide transport and retardation behaviour can be significantly changed from that of the same radionuclide in solution. Radionuclides bound to mobile colloids can have earlier and increased peak arrivals, while radionuclides bound to immobile colloids are effectively retarded.

Bibliographic references:

Degueldre C, Triay I, Kim J-I, Vilks P, Laaksoharju M and Miekeley N (2000) Groundwater colloid properties: a global approach. Applied Geochemistry, 15, 1043-1051.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D (1995, editor) The scientific and regulatory basis for the geological disposal of radioactive waste. John Wiley & Sons.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Stumm W and Morgan JJ (1996) Aquatic Chemistry, 2nd ed. John Wiley & Sons.

Equivalent FEPs:

Relates to FEPs 2.2.08 (Chemical/geochemical processes and conditions in geosphere), 2.2.09 (Biological/biochemical processes and conditions in geosphere) and 3.2.04 (Colloids, contaminant interactions and transport with) in the NEA International Database (NEA, 1998).

Relates to FEP 5.45 (Colloid generation and transport) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-5**

Name: **CREEPING OF THE ROCK MASS**

Short description:

Creeping of the rock mass is the slow movement of the rock in response to changes in the stress field. Creeping can occur along pre-existing discontinuities or in the rock matrix due to differential stress fields at a very slow rate. Creep of the rock mass may affect the hydraulic properties of the rock and may have a mechanical impact on the buffer and backfill.

Technical description:

Creep of the rock mass is a slow, quasi-continuous (time-dependent) deformation process that generally occurs along pre-existing discontinuities in the rock. However, creeping at a very slow rate can also take place in the rock matrix due to differential stress fields.

Creep occurs in response to a changing (or unstable) stress field in the rock mass and can occur on a variety of temporal and spatial scales. In the context of a deep repository in Sweden, the processes most likely to generate creep are related to plate tectonics, climate change, repository excavation and, in the spent fuel repository, to the heat load and the swelling pressure exerted by the bentonite buffer.

The slow deformation of rocks in Sweden occurring due to plate tectonic processes is in response to the opening of the northern Atlantic Ocean. These plate tectonic processes are very slow and long-term. They cause straining of the rock mass which results in either slow creep deformation, generally along suitably oriented fault zones, or rapid seismic (earthquake) events.

Deformation associated with climate change occurs in response to repeated glacial loading and unloading events, and eustatic sea level changes (see GEN-21 “Glaciation”). Loading and unloading of the continental crust by the advance and retreat of thick ice sheets and the sea will produce creep of the crust and the underlying upper mantle to several tens of kilometres depth. Relaxation times for this deep-seated deformation are long and the whole of Scandinavia is still rebounding from the last glaciation. True creep deformation at depth is sometimes accompanied by brittle deformation in the upper crust which results in processes such as displacements along fault zones.

Deformation associated with creep relaxation of the rock mass will also occur around underground openings created during repository excavation. Stress concentrations produced in the rock mass around the repository will have a tendency to close with time. However, much of the closure around such openings may occur more by fracture displacement than by true creep (see GEN-17 “Faulting”).

The most significant form of creep occurring in most repositories is likely to be that caused by the repository excavations because deformation by plate tectonic and climate change processes occurs extremely slowly, even when compared to the expected lifetime of a repository. In the spent fuel repository, rock creep can also be generated by the thermal loading from the waste through thermal-mechanical coupling (see SFL-46 “Temperature of the near-field”) and by the swelling pressure generated by the bentonite buffer (see SFL-1 “Swelling of the bentonite buffer”).

The extent of any creep deformation around the repository openings will depend on the in situ stress ratios in the rock mass; the creep properties of the rock mass, including the contribution to the creep provided by the deformation of the fracture network; the orientation of the underground openings with respect to the in situ stress tensor; the contrast in deformation properties between the rock mass and the backfill and buffer; and the temperature increases caused by the waste (e.g. Hansson et al., 1995).

The impact of creep will be enhanced by higher in situ stress ratios, by rock that allows creep at lower stresses, by misaligned or inappropriately shaped underground openings which will maximise the stress concentrations and by having a large contrast between the deformation properties of the rock mass and the backfill. Creep deformation is also likely to be maximised in areas of higher fracture density, since the potential sliding rates on fractures could be high and the shear stress required to initiate such movements low, in comparison with the shear stress required to initiate substantial creep in unfractured rock.

Creep deformation on the scale of the repository is likely to be concentrated in areas of highest shear stress or in areas where the resistance to deformation is lowest. For the former these are likely to be areas around underground openings and for the latter within fracture zones with large amounts of weak infill material.

Evidence from site investigations in Finland (e.g. TVO, 1992) shows that some Precambrian mica schists have considerably lower strengths that the other basement rocks. Under similar stress conditions, creep rates in this type of rock would be expected to be greater than those in the more competent components of the crystalline basement. Furthermore, creep rates are likely to be enhanced at times when thick ice loads are present over the repository.

Creep occurs along pre-existing discontinuities and is likely to change the hydraulic and transport properties of these discontinuities. In particular the fracture connectivity and channelling properties could change, such that the preferential flow paths through the rock mass could alter with time.

Creep of the rock around repository excavations will deform buffer and backfill materials. In an extreme case it could affect the waste package, although considerable creep deformation would be required for any damage to occur and this is considered very unlikely.

Despite the fact that creep is certain to occur in the repository host rock, its significance for safety is minimal. This is because creep processes operate only very slowly due to the very long time constants for creep in hard crystalline rock. Because of this, rock deformation is likely to be dominated by fracturing, rather than by true creep.

Origin in the repository system:

Creep along pre-existing discontinuities in the rock will occur due to changes in the stress field caused by plate tectonics and climate change processes. However, the impact of creep by these processes will be limited by the slow rates of these processes.

Creep is more likely to impact on the repository as the rock mass deforms in response to the excavation of the repository openings and, in the spent fuel repository, by the heat loading and swelling of bentonite. The extent of creep will be controlled in part by the geometry of the excavations and by the strength of the engineered barriers.

Impact on the repository system:

Creep of the rock mass will affect its hydraulic properties, particularly the hydraulic conductivity and channelling attributes of the fractures. Thus creep can impact on radionuclide transport in the host rock. However, rock deformation is likely to be dominated by fracturing processes and not creep. The impact of creep on the repository is, therefore, considered to be minimal.

Bibliographic references:

Hansson U, Shen B, Stephansson O and Jing L (1995) Rock mechanics modelling for the stability and safety of a nuclear waste repository. SKI Technical Report, 95:41.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

TVO (1992) Final disposal of spent nuclear fuel in the Finnish bedrock: preliminary site investigations. TVO Technical Report YJT-92-32E.

Equivalent FEPs:

Relates to FEPs 2.2.01 (Excavation disturbed zone, host rock), 2.2.06 (Mechanical processes and conditions in geosphere) and 1.2.02 (Deformation, elastic, plastic or brittle) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.9 (Creeping of rock mass) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-6**

Name: **GROUNDWATER SALINITY CHANGES**

Short description:

Significant changes in groundwater salinity may occur due to changes in groundwater flow, to the intrusion of marine recharge waters or to the intrusion of dilute glacial meltwaters. Changes in groundwater salinity will affect groundwater flow paths, the swelling properties of bentonite, the degradation of metal components in the engineered barriers, radionuclide solubility, colloid stability and sorption.

Technical description:

Groundwater is considered to be saline if it contains more than 1000 mg/l of dissolved solids. Deep saline groundwaters are referred to as brines, denoting saline waters containing Ca, Na, K, Cl and minor amounts of other elements. Brines commonly have 10 to 15% salinity. The Br/Cl ratio may often be used to distinguish marine waters from formation brines.

The groundwater in the far-field rock is likely to show natural variations in salinity with depth because deeper waters generally have had longer rock-water interaction times and, hence, their concentration of dissolved species (salinity) increases (see GEN-22 “Far-field groundwater chemistry”). This has been observed at many locations such as Äspö (Smellie and Laaksoharju, 1992). The salinity of groundwater also will be affected by the mineralogy of the rocks through which they flow, with salinity increasing with the abundance of readily soluble salt minerals in sedimentary lithologies (see GEN-1 “Alteration and weathering along flow paths”).

Groundwater salinity often increases with depth and may vary laterally according to groundwater recharge and discharge zones, and past and present saline interfaces related to sea-level. Changes in groundwater salinity due to glacial effects will influence chemical equilibria, and salinity gradients may be of importance for groundwater flow. Intrusion of saline water in significant amounts could occur during the operational and resaturation phases of a waste repository.

An increase in groundwater salinity can occur if the groundwater flow system is modified by the onset of glaciation (see GEN-21 “Glaciation”). When either a glacier or a thick permafrost horizon covers the land mass, many groundwater recharge and discharge sites may become frozen, causing the groundwater flowpaths and times to be significantly extended (see GEN-26 “Permafrost”). The subsequent increase in rock-water interaction times often means that deep subglacial and sub-permafrost groundwaters become very saline (McEwen and de Marsily, 1991). However, at sites where significant volumes of glacial meltwater penetrate into the ground, groundwaters may become very dilute and oxidising.

Lastly, the salinity of the groundwater may also be affected by present and past intrusions of saline recharge water of marine origin. In repository areas located near to coastal sites, isostatic and eustatic changes in sea-level can result in the migration of the saline water interface (the boundary between groundwater of marine origin and groundwater of fresh meteoric origin).

A change to the groundwater salinity will affect the groundwater chemistry in the far-field and in the repository. Some chemical elements form soluble complexes with chloride which may influence canister corrosion, and may change the solubility and sorption of radionuclides. High salinity may also affect the swelling capacity of clay buffer materials as well as the stability of colloids. The pH of groundwaters in crystalline rocks is considered to be buffered by reactions involving aluminosilicate minerals such as feldspars, clays and micas in conjunction with carbonates such as calcite (Hanor, 1994). Changing the salinity of groundwater in such systems will change the pH in accordance with these buffering reactions.

The formation of sharp interfaces between saline and fresh groundwaters should be considered in terms of effects on both geochemical processes and groundwater flow (Voss and Andersson, 1993; Provost et al., 1996). Saline water intrusion implies changing density gradients and changing groundwater flow geometry: the location of discharge areas may alter as a consequence.

In much of the bedrock of Sweden, saline water exists at depth, ranging in salinity from values typical of present sea water at shallower depths of a few hundred meters, to higher salinities at depths of a few kilometres. The significance of variable salinity-controlled density on groundwater flow at repository depths is uncertain. Further questions arise as to the change in the flow field during a future time in which the existing saltwater inland is gradually swept out of the bedrock by permeating freshwater.

The origin of the deeper highly saline waters may represent in-situ geochemical processes, whereas the upper saline waters, particularly in the regions below the highest post-glacial coastline, appear to derive from post-glacial seas. For repository depths of a few hundred meters, waters are likely to be of post-glacial sea origin and the salinity reflects long residence times (Voss and Andersson, 1993). However, long residence times do not automatically imply that present-day or future groundwater movement is slow because the flow field can be significantly affected by processes such as uplift or glaciation. In such cases, the presence of saline water will not indicate a slow or stagnant groundwater flow field. Consequently, significant saltwater flows may occur in fracture zones even at considerable depths. Thus, a repository situated in a saltwater zone is not necessarily in a stagnant flow field, and a safety analysis may not treat it as such.

Changing groundwater salinity is of significance to both the spent fuel repository and the SFR. In the spent fuel repository, because of the long time periods of interest in the performance assessment, saline waters may be generated in the future by both sea-level changes and the onset of glaciation. The significance of saline water on the spent fuel repository is largely due to the potential for an increase in the corrosion rate of the copper outer canister (see SFL-7 “Corrosion of the copper shell of the canister”) and the possible reduction in bentonite swelling (see SFL-1 “Swelling of the bentonite buffer”).

In the SFR, the initial groundwaters are saline. However, these waters are not thought to be the result of substantial intrusions of seawater despite the facility being sited beneath the Baltic Sea. Rather, these waters reflect long rock-water interaction times. Future uplift of the land will change the groundwaters at the SFR to a more dilute composition, as the uplift causes the saline water interface to migrate past the repository in about 1000 years time (SKI, 1994).

Origin in the repository system:

Groundwater saturates the entire repository system and the far-field. The salinity of this groundwater may vary due to flow accompanying long-term processes such as rock-water interactions and glaciation. Any change in salinity may result from upwelling of deeper water or immersion of the land by seawater.

Impact on the repository system:

Changing salinity of groundwater will affect radionuclide solubility and sorption, colloid stability, the degradation of engineered barrier system materials and the stability of bentonite. It may also impact upon groundwater flow.

Bibliographic references:

Hanor J (1994) Physical and chemical controls on the composition of waters in sedimentary basins. Marine and Petroleum Geology, 11, 31-45.

McEwen TJ and de Marsily GH (1991) The potential significance of permafrost to the behaviour of a deep radioactive waste repository. SKI Technical Report, 91:8.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Provost A, Voss C and Neuzil C (1996) Glaciation and regional groundwater flow in the Fennoscandian shield. SKI Technical Report 96:11.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Report 94:30.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Smellie JAT and Laaksoharju M (1992) The Äspö Hard Rock Laboratory: final evaluation of the hydrogeochemical pre-investigations in relation to existing geologic and hydraulic conditions. SKB Technical Report, TR 92-31.

Voss C and Andersson J (1993) Regional flow in the Baltic shield during Holocene coastal regression. Groundwater, 31, 989-1006.

Equivalent FEPs:

Relates to FEPs 2.2.07 (Hydraulic/hydrogeological processes and conditions in geosphere) and 2.2.08 (Chemical/geochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 5.1 (Saline (or fresh) groundwater intrusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-7**

Name: **DEGRADATION OF THE BOREHOLE AND SHAFT SEALS**

Short description:

Degradation of the borehole and shaft seals may occur by a combination of physical and chemical processes. This will lead to partial or total loss of their sealing function and thus may impact on groundwater flow and radionuclide transport processes.

Technical description:

Seals will be placed in exploratory boreholes and in access shafts and tunnels to prevent artificial fast pathways being introduced which could bypass the majority of the far-field barrier (see GEN-35 “Fast transport pathways”). The majority of seals proposed are based either on bentonite, both compacted and uncompacted, or cementitious materials, normally concrete or cement grout. Seals will be used in both the SFR and spent fuel repositories, thus this issue is of relevance to both repository designs.

The potential flux of groundwater along or across a shaft or borehole could be considerable if it is unsealed and connects together (short-circuits) highly transmissive features such as flowing fracture systems. This potentially could occur if the shaft or borehole seals fail, e.g. due to changes in the stress field (see Gen-36 “Stress field”). The shafts, in particular, need to be sealed in such a way that degradation of the seals is likely to be a slow process under expected future stress and climate conditions. Sealing boreholes is generally easier than shafts, and will normally be carried out using either compressed bentonite or cement grout. This process will be relatively easy, provided the boreholes are still accessible to the necessary depths, although there will be a considerably greater number of boreholes than shafts that will require sealing. In addition, highly transmissive zones intersected during repository construction will also be sealed (e.g. Borgesson and Pusch, 1989) using concrete and compacted bentonite.

The degradation of the seals will take place by a variety of physical and chemical processes and is dependent on the types of seals that are installed. Cement-based seals are more brittle than bentonite-based seals and will have greater difficulty in accommodating movements in the rock mass. Bentonite-based seals, however, will be more prone to erosion and could be ineffective if frozen. A number of degradation processes are envisaged (Savage et al., 1998; Stroes-Gascoyne and Johnson, 1998).

Both cement and clay-based seals may not be in either thermal or chemical equilibrium with the repository conditions and changes can be expected due to this. In repositories which undergo thermal transients, changes will be more rapid in the near-field than the far-field due to the higher thermal gradient expected in the near-field.

The porosity, pore-size distribution and hydraulic conductivity of cements and concretes all change with time. These changes are the result of crystallisation of amorphous phases and other reactions such as progressive hydration. On their own these processes are unlikely to affect the sealing capacity of cement-based seals, unless secondary alteration phases are formed that cause cracking of the cement by volume expansion, although this is considered unlikely.

Mineralogical alteration of bentonite to a non-expandable clay can occur by reaction with the groundwater, with a concomitant loss in swelling capacity, although at the temperatures expected in the repository such a reaction will be very slow and significant changes are not expected within the lifetime of the repository.

Reactions between bentonite and hyperalkaline pore fluids generated from adjacent cement-based materials could also affect the swelling capacity and plasticity of the bentonite (a process known as embrittlement). This is particularly an issue for the cementitious SFR. These effects could, however, be partially counteracted by a decrease in porosity of the bentonite. Cement-based seals can also partially dissolve in the groundwater and react with saline constituents, and reprecipitation may provide the possibility of self-sealing in favourable cases. Dissolution may be limited in cements with a low hydraulic conductivity.

Cement-based seals can be affected by thermally-induced stresses and by rock movements, since they are inherently more brittle than clay-based seals. A reduction in thermally induced cracking of the seals can be achieved, however, by the use of dense, low-heat cements, by pozzolanic materials and lime-based components and by the use of established concrete placement techniques. These problems are unlikely to affect thin, cement-based seals which, in many cases have been shown to be self-healing (e.g. Stroes-Gascoyne and Johnson, 1988).

Freezing of seals, both cement and clay-based, may affect their subsequent behaviour and make them less effective, although most high quality cements are resistant to freeze cracking. Freezing may take place in extreme conditions to depths of several hundred metres but not as deep as the repository itself (SITE-94 Central Scenario: King-Clayton et al., 1995).

Microbial action could enhance the degradation of seals by direct microbial attack or by catalysing other deleterious chemical reactions.

Of the many processes that can affect the seals, the chemical processes are the least significant for repository safety because they are generally slow and are similar in nature to the well understood degradation processes that will affect bentonite and cement in the near-field. In contrast, the physical processes could be more significant, especially for those seals near the surface which will be subject to the strong impacts of climate change, especially due to glaciation causing deformation to the rock mass (see GEN-21 “Glaciation”). This is most relevant for the spent fuel repository given the very long time period of interest in safety assessments for this repository.

Below the maximum depth that permafrost is thought to reach (c. 200 m), the seals should remain intact for long periods of time (see GEN-26 “Permafrost”). However, the quantitative lifetime of all seals will need to be assessed in future safety submissions.

Origin in the repository system:

Degradation of seals in exploratory boreholes and in shafts and access tunnels will occur by a combination of physical and chemical processes. However, the chemical (alteration and dissolution) processes are expected to be very slow and are not thought likely significantly to impact on safety during expected repository evolution scenarios. The most problematic seal degradation processes are thus physical and result from mechanical damage caused by deformation of the rock mass. Therefore, the longevity of seals is controlled largely by changes to the stress field in the rock and the onset of glaciation/permafrost.

Impact on the repository system:

Seal degradation will permit enhanced groundwater flow along exploratory boreholes and access shafts and tunnels, from repository depth towards the surface. Complete failure of the seals may provide short-circuit pathways resulting in a loss of the isolation capacity of the far-field rock. However, even partial failure of the seals might cause enhanced groundwater flow in the near-field of the repository which could lead to faster degradation of the engineered barriers and thus to a changed near-field groundwater chemistry.

Bibliographic references:

Borgesson L and Pusch R (1989) Rock sealing by dynamic injection. Stripa project Phase III. In: Sealing of radioactive waste repositories. Proceedings of a NEA/OECD Workshop.

King-Clayton LM, Chapman NA, Kautsky F, Svensson NO, de Marsily G and Ledoux E (1995) The central scenario for SITE-94. SKI Technical Report, 95:42.

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Savage D, McLeod R and McEwen T (1998) An evaluation of repository-induced disturbances for a KBS-3 type repository. SKI Technical Report, 98:20.

Stroes-Gascoyne S and Johnson L (1988) Clay and cement-based backfill and sealing materials designed for use in a Canadian nuclear fuel waste disposal vault. Proceedings of Joint EA/USDOE Workshop on backfill materials, Carlsbad, New Mexico, May 1998.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.05 (Seals, cavern/tunnel/shaft), 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 5.11 (Degradation of hole and shaft seals) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-8**

Name: **DEGRADATION OF THE ROCK REINFORCEMENT AND GROUT**

Short description:

Degradation of rock reinforcements in the repository may occur by a combination of physical and chemical processes. The post-closure safety of the repository is not, in general, dependent on the functioning of these materials but their degradation may impact on other parts of the engineered barriers.

Technical description:

During the repository construction phase, reinforcements may be used to provide support for the excavations, and grouts may be used to restrict the inflow of groundwater. Their purpose is to ensure a safe and dry working environment during the operational phases and no post-closure containment role is assigned to them in formal performance assessments. Particular care was given to the design of the reinforcements for the roof of the silo in the SFR because of its large diameter span and long operational period.

Grouts and reinforcements are likely to be concretes and mortars (e.g. shotcrete, which is pneumatically applied concrete), and steel (e.g. beams and rockbolts) and may be dual purpose, for example a shotcrete surface usually contains reinforcing steel wires to provide mechanical support and to limit groundwater ingress. However, additional materials may be used in smaller amounts, such as epoxy resin in conjunction with the concrete and steel.

The design lives of such support and reinforcement techniques are measured in decades or possibly hundreds of years and, thus, they can be expected to begin to degrade shortly after repository closure. This degradation can occur by a range of physical and chemical processes, and can have a variety of impacts in the near-field.

Cement-based grout and shotcrete will react with the groundwater and, depending on the ambient groundwater chemistry, a number of dissolution and precipitation reactions can occur. Calcite, brucite and ettringite are all possible and their precipitation can occur in fractures and pores in the rock, buffer or backfill, but also in the concrete material itself, causing changes to the porosity distribution in the near-field. Ettringite formation has, in some cases, been shown to cause disruptive expansion and, if this were to occur in the cement structures, this could cause enhanced physical degradation.

If large volumes of cement are present (e.g. as seals in deposition holes or tunnels), dissolution of the cement-based materials will also create alkaline pore-fluids that can affect the swelling capacity and plasticity of bentonite (a process known as embrittlement). This is likely to be the most serious potential problem resulting from degradation of cement seals in the near-field of the spent fuel repository.

If the alkaline pore-fluids migrate to the near-field rock, they are predicted to react with silicate minerals in a replacement reaction to form zeolites and calcium-silicate-hydrate minerals. Some of the phases predicted to form have a high sorption capacity and hence may retard radionuclide transport. However, they may be formed in mobile colloidal form which could enhance radionuclide transport. This is more particularly a potential problem in repository designs with large volumes of cement, such as the SFR.

Anaerobic degradation of steel material used as reinforcement will help to buffer the redox conditions in the near-field and will result in hydrogen formation and formation of solid corrosion products. The latter could potentially cause blocking of pores and fractures in the adjacent backfill material and rock.

Solid degradation products from both the cement and steel will provide efficient sorption sites for radionuclides migrating through the bentonite buffer, although they are unlikely to be very abundant. However, these solid alteration products may also increase the colloid population in the near-field porewater.

Physical degradation to the reinforcements and grouts is likely during the immediate post-closure period as the rock mass deforms in response to stress readjustments (see GEN-36 “Stress field”) and due to hydraulic resaturation (see GEN-32 “Hydraulic resaturation of the near-field rock”).

In the SFR repository an important property of grouting concrete is that it should allow gas to permeate at relatively low pressure differences. Consequently, gas permeable material is used as aggregate in the concretes. However, post-closure reactions involving the cement could cause a reduction in the cement porosity and thus to its gas permeability. The most significant reaction in this regard is the ‘pozzuolan reaction’ whereby calcium hydroxide in the cement slowly reacts with the silica in the aggregate to form swelling, gel-like secondary products that reduce porosity. Also, calcite may be formed in the pores when dissolved carbon dioxide comes into contact with the alkaline concrete. However, assessments of these reactions for the SFR (SKI, 1994) suggest that any reduction in gas permeability will not be significant.

Origin in the repository system:

Degradation of the rock reinforcement and grout is controlled primarily by the chemistry of the groundwater, although physical degradation will result from rock mass deformation (stress readjustments) and hydraulic resaturation in the immediate post-closure period.

Impact on the repository system:

Degradation of rock reinforcements and grout affects the groundwater chemistry, notably by buffering the pH (in the case of cement) and the Eh (in the case of steel). The changed groundwater may react with other components of the engineered barriers, most immediately the bentonite buffer. Anaerobic corrosion of steel reinforcements will generate hydrogen gas but in smaller volumes than generated by corrosion of the iron waste packages.

Solid alteration products and precipitates may reduce the porosity of the bentonite and the rock, and may increase the colloid concentration.

Continued degradation will impact on the physical stability of the reinforcements and the grouts, although this is of little radiological significance since they are not assigned any role in repository safety.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1994) Evaluation of SKB's In-depth Safety Assessment of SFR-1. SKI Technical Report 94:30.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.06 (Other engineered features materials and characteristics), 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.10 (Chemical effects of rock reinforcement) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-9**

Name: **DIFFUSION**

Short description:

Diffusion is the process whereby chemical species move under the influence of a chemical potential gradient (usually a concentration gradient). In the repository, diffusion of dissolved species might be significant in the buffer, the backfill and certain host rock environments where advective transport does not occur or is limited.

Technical description:

Diffusion theory is adequately defined and discussed in the scientific literature (e.g. Crank, 1975). In terms of understanding diffusion in the repository environment, a simple but adequate definition is that diffusion is the result of the thermal movements in the solution (Brownian motion) and effectively is the net flux of solutes from a zone of higher concentration to a zone of lower concentration, that does not depend on any bulk movement of the solution (Spitz and Moreno, 1996). In the repository, diffusion of dissolved species might be significant where transport by advection is limited. Diffusion in the porosity of the bulk mass of a crystalline rock is referred to as matrix diffusion (see GEN-25 “Matrix diffusion”).

Diffusion is discussed here mostly in terms of diffusion through compacted bentonite. This material is used as the buffer around canisters in the spent fuel repository and between the concrete silo and the host rock in the SFR. However, the concepts can also be applied to a bentonite-rich backfill and to a low permeability sedimentary host rock, although account must be taken of the differences in the physical characteristics (e.g. porosity and bulk density) of these materials.

Diffusion through low permeability materials in the near-field was extensively reviewed in recent literature (NEA, 1993). For performance assessment, the critical issue is one of quantifying repository-relevant diffusion coefficients for the chemical species of interest. In low permeability materials, diffusion takes place almost wholly through the interstitial water and is slower than it would be in pure water because porosity is less than unity and diffusion paths are not straight. These effects can be taken into account by the definition of an effective diffusion coefficient, D_e. However, for chemical species whose transport is significantly affected by retardation processes, it is possible to model and describe their behaviour using an ‘apparent’ diffusivity, D_a, which is related to D_e via the porosity and bulk density of the transport medium, and the distribution coefficient (K_d) for the diffusing species in the transport medium.

The diffusion coefficient is ion-specific (or particle specific for a colloid) because it is a function of the radius of the hydrated ion that diffuses. It is also temperature specific. When D_e values are determined from laboratory experiments for non-sorbing species on compacted bentonite, they fall in the range 5×10^{-13} to 5×10^{-11} m^2/s, with values for most sorbing actinide species falling into the upper part of this range. In contrast, the alkali and alkaline earth cations (e.g. Cs^+ and Sr^{2+}) behave abnormally, often showing values of about one order of magnitude higher. This phenomenon is often referred to as 'surface diffusion' (e.g. Cheung, 1990) and is postulated to occur in the electric double-layer adjacent to mineral surfaces. However, this interpretation has been challenged because, if it were correct, it would imply that measured diffusion rates would increase with compaction by enhancing connections in the electric double-layers, but laboratory evidence shows this does not happen (Conca et al., 1993). It has thus been suggested that 'surface diffusion' is only an artefact of the data fitting procedure and the use of inappropriate (overly simplistic) diffusion models (NEA, 1993). Part of the problem is that, to calculate D_e, laboratory measurements of K_d must also be undertaken using separate sorption tests and this data used to normalise the D_a values. There is some concern that this procedure is invalid.

It is clear that the fundamental conceptual understanding of diffusion in low permeability porous media has not yet been established and agreed and, thus, further investigations of this issue are warranted, and it is probable that more detailed, multi-phase models to predict diffusion in compacted bentonite are required that can take account of apparent 'surface diffusion' as well as other factors that affect diffusion such as anion exclusion.

However, regardless of the exact mechanism of surface diffusion, it is important that diffusion coefficients used in performance assessment are not non-conservative.

The diffusivity is affected not only by the characteristics of the low permeability transport medium (porosity and bulk density) but also by the ionic strength of the porewater. The ionic strength of the water affects the characteristics of the electric double layer adjacent to the mineral surfaces which in turn has implications both for anion exclusion and surface diffusion effects on the diffusive mass transport.

Diffusive mass transfer in the bentonite buffer is not, however, completely decoupled from advection in the host rock. Increasing the flow rate in the host rock will reduce the concentration of migrating species at the buffer-rock interface. This maintains a steep concentration gradient and, thus, a larger diffusional driving force through the bentonite.

Origin in the repository system:

Diffusion in the repository will be significant as a transport mechanism when advection is limited and when a chemical gradient exists to drive the diffusing species. In low permeability media (e.g. the buffer or backfill), diffusion is thus controlled by the physical characteristics of the transport medium; the chemistry of the porewater (ionic strength and chemical gradient of the migrating species); release rates for the migrating species from the wasteform; and the groundwater flow rate in the advecting part of the system.

Impact on the repository system:

Diffusion through the compacted buffer and backfills controls the porewater chemistry in the near-field and thus can have an impact on the corrosion and degradation of the near-field barriers. Diffusion through the bentonite buffer is also the primary controlling process for the bulk radionuclide release rate to the far-field and consequently on the far-field groundwater chemistry (radionuclide contents).

Diffusion in a low permeability host rock can be a dominant transport process in the far-field. This will impact on far-field chemistry and on the bulk radionuclide release rate to the biosphere.

Bibliographic references:

Cheung SCH (1990) A new interpretation of measured ionic diffusion coefficients in compacted bentonite-based materials. Engineering Geology, 28, 369-378.

Conca JL, Apted M and Arthur R (1993) Aqueous diffusion in repository and backfill environments. Scientific Basis for Nuclear Waste Management, XVI.

Crank J (1975) The mathematics of diffusion (2nd edition). Oxford University Press.

NEA (1993) The status of near-field modelling. NEA/OECD, Paris.

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Rebour V, Billiotte J, Deveughele M, Jambon A and Le Guen C (1997) Molecular diffusion in water saturated rocks: a new experimental method. Journal of Contaminant Hydrology, 28, 71-94.

Spitz K and Moreno J (1996) A practical guide to groundwater and solute transport modelling. John Wiley and Sons Ltd.

Equivalent FEPs:

Relates to FEP 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.6 (Diffusion - surface diffusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Dave Savage and Bill Miller

Number: **GEN-10**

Name: **RADIONUCLIDE DISPERSION**

Short description:

Dispersion is the collective name for the consequences of a number of processes that cause 'spreading-out' of a contaminant plume in all directions, superimposed on the bulk movement predicted by a simple advection model. It results in a spatially distributed contaminant plume.

Technical description:

The process by which solutes are transported by the bulk motion of a flowing groundwater is advection (Bear, 1972; de Marsily, 1986). Regardless of the level of detail of the model used to describe advective transport, there will always be some smaller-scale variations in the flow field that are not explicitly accounted for in the advection term. The consequences of these variations, coupled with mechanical mixing and molecular diffusion are termed 'hydrodynamic dispersion' and can result from differences in microscopic velocities along the flow path, mechanical mixing during advection and molecular diffusion resulting from the thermal-kinetic energy of solute particles.

Spreading of the solute plume can occur in the direction of advection, in which case it is known as longitudinal dispersion, or it can occur perpendicular to the direction of advection, in which case it is known as transverse dispersion. Together, or separately, longitudinal and transverse dispersion can produce variations in solute concentration throughout the contaminant plume and can transport contaminants into zones that do not participate directly in the advective plume evolution. Dispersion effects such as velocity variations and mixing between different flow paths will increase the spatial and temporal spreading of solutes. Mixing of groundwaters of different composition, may also affect the chemical and physical retardation of migrating radionuclides.

The need for a dispersion factor arises from the heterogeneity of the transport medium on scales beneath that modelled and by the limited description of the flow profiles in predictive models. Thus, dispersion is not a fundamental process but rather a mathematical representation of the net effects of small-scale variations in the flow field, in combination with molecular diffusion and mechanical mixing.

Dispersion can occur in all parts of the repository system where the contaminant plume is migrating. Thus it can occur in the near-field, where dispersion is controlled by diffusion processes in the low permeability buffer (see GEN-9 "Diffusion"), or it can occur in the far-field where bulk transport is dominated by advection.

In accordance with the different orders of heterogeneity in hydrogeological systems, dispersion can be visualised as taking place at several different scales in a fractured crystalline rock. At the small scale (heterogeneities of first order), contaminants are spread by mechanical dispersion. Diffusion within the fractures, velocity variations within the fracture apertures, mixing at fracture intersections and similar mechanisms occurring during transport within the porosity contribute to dispersive spreading at the microscale.

At the rock mass scale, larger-scale heterogeneities within the fracture network (heterogeneities of second order) contribute to the dispersive spreading. Permeability differences within the fracture network cause a flow field with velocities varying in a complex irregular manner in three dimensions. Flow in highly transmissive fractures will carry the contaminant faster than the average transport velocity, generating dispersive spreading.

At the field scale, large-scale geological structures can significantly influence contaminant transport. Whether or not individual geological structures dominate the transport behaviour depends mainly on the size of the complete flow system, in comparison to the size of the geological heterogeneities.

In cases of extreme heterogeneity, such as a fractured crystalline rock, meaningful estimates of large-scale dispersion parameters may not be obtainable. Hence, descriptions of large-scale transport involving dispersion are best handled by detailed hydrogeological models, such as discrete fracture network models or stochastic continuum models. In the SITE-94 performance assessment, both a discrete feature site model (Geier, 1996) and a stochastic continuum site model (Tsang, 1996) of the Äspö site were produced.

Quantifying dispersion for inclusion in simplified advection-dispersion transport models is problematic. In some cases, usually for porous media, dispersion is represented using a Fickian approach (assuming diffusion-like behaviour whereby the associated flux is proportional to the gradient in contaminant concentration). However, this approach is not necessarily accurate because it does not explain the observed scale dependency of dispersivities as measured in field and laboratory experiments (Neuman, 1990; Dagan, 1990; Neuman and Zhang, 1990).

When considering a fractured crystalline rock at the field scale, represented using channel or channel network models, dispersion cannot be described as a Fickian process, not even for one-dimensional flow. It has been suggested (e.g. in KBS-3) that channelling could be described with a constant Peclet number [usually between 4 and 10, determined as vL/D, with v the flow velocity, L the transport distance and D the effective dispersion coefficient] implying that the dispersivity will increase with travel distance. Large-scale dispersion may not be significant in cases of extreme channelling because transport is skewed to the fast flow paths.

An important lesson from the many laboratory, field and modelling studies that have investigated advection and dispersion processes is that, even for simple, fully characterised systems, the dispersion behaviour is not truly Fickian up to very large scales. This apparent scale dependency suggests that, unless the phenomenon of dispersion is better understood, there can be little hope of using current models to extrapolate results based on the laboratory scale to the field scale and to quantify dispersion coefficients. Hence, large-scale experiments are needed. Further, the use of dispersion to calculate dilution requires care because dispersion is more a measure of uncertainty in concentration values and the calculation of dilution depends on sampling scale relative to dispersivity.

Uncertainties regarding dispersion are greatest for the transport of weakly and non-sorbing radionuclides. For sorbing radionuclides, in contrast, uncertainties related to parameters controlling retardation (e.g. distribution coefficients) are of greater importance, although dispersion does affect the wetted surface area available for sorption.

Origin in the repository system:

Dispersion can be regarded as an artefact of the heterogeneity of the transport medium at smaller scales than that represented by the advective flow model or the representation of advection in the transport model. Thus, dispersion is controlled by the physical characteristics (including spatial heterogeneity and scale dependence) of the transport medium (e.g. buffer, backfill or host rock).

Impact on the repository system:

Dispersion impacts on transport in all parts of the repository system, although it is of most significance in the far-field where dispersion causes the contaminant plume to spread-out away from the direction of advection. It thus impacts most notably on the far-field groundwater chemistry (radionuclide distributions, concentrations and gradients) and the bulk radionuclide release rate (breakthrough curves) to the biosphere.

Bibliographic references:

Bear J (1972) Dynamics of fluids in porous media. Elsevier, New York.

Dagan G (1990) Transport in heterogeneous porous formations: spatial movements, ergodicity and effective dispersion. Water Resources Research, 26, 1281-1290.

Geier JE (1996) Discrete-feature modelling of the Äspö site. SKI Technical Report, 96:14.

Gelhar LW (1986) Stochastic subsurface hydrology from theory to applications. Water Resources Research, 22, 135-145.

Gelhar LW and Axness CL (1983) three dimensional stochastic analysis of macrodispersion in aquifers. Water Resources Research, 19, 161-180.

Impey MD and Grindrod P (1993) Channelling and Fickian dispersion in fractal simulation of porous media. Water Resources Research, 12, 4077-4089.

de Marsily, G (1986) Quantitative hydrogeology; groundwater hydrology for engineers. Academic Press, New York.

NEA (1991) Heterogeneity of groundwater flow and site characterisation. NEA/OECD.

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Neuman SP and Zhang YK (1990) A quasi-linear theory of non-Fickian and Fickian subsurface dispersion. 1: theoretical analysis with application to isotropic media. Water Resources Research, 26, 887-902.

Tsang YW (1996) Stochastic continuum hydrogeological model of Äspö. SKI Technical Report, 96:9.

Equivalent FEPs:

Relates to FEP 3.2.07 (Water mediated transport of radionuclides) in the NEA International Database (NEA, 1998).

Relates to FEP 6.4 (Dispersion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-11**

Name: **DISTRIBUTION AND RELEASE OF RADIONUCLIDES FROM THE FAR-FIELD**

Short description:

Radionuclides migrating from the near-field will become dispersed in the far-field rock and some proportion of these radionuclides could be released to the surface environment. The distribution of radionuclides in the far-field and their release mechanisms to the biosphere are dependent on the mechanisms and rates of all processes that control release, transport and retardation in both the near and far-fields.

Technical description:

Radionuclides that migrate from the near-field into the far-field rock spread-out as they travel through the rock mass by a process termed hydrodynamic dispersion which produces variations in solute concentration (dilution) throughout the contaminant plume and can transport contaminants into zones that do not participate directly in the advective plume evolution (see GEN-10 “Radionuclide dispersion”). Dispersion effects such as velocity variations and mixing between different flow paths will affect (increase) the spatial and temporal distributions of solutes.

Gas generation and transport processes in the repository can also affect the distribution and release of radionuclides which can be associated with a gas phase (e.g. ^{14}C in CO_2) in the far-field and need to be considered when estimating radionuclide distributions. Gases can be generated in both the SFR and spent fuel repositories (see SFR-11 “Gas generation in the repository” and SFL-25 “Gas generation in the canister”). Minor amounts of gas may also be generated in the far-field (see GEN-19 “Gas generation in the far-field”).

Retardation processes also act to change the radionuclide distribution in the far-field. In particular, sorption-desorption, precipitation-dissolution and isotopic exchange reactions will produce nuclide specific concentration variations throughout the far-field (see GEN-27 “Radionuclide precipitation and dissolution” and GEN-34 “Radionuclide sorption”) and these must also be considered when estimating radionuclide distributions.

The many processes that contribute to the radionuclide distribution in the far-field and release to the biosphere may occur around both the spent fuel and SFR repositories, although some processes may be more important for one facility than for the other, particularly with regards to near-field behaviour.

One of the objectives of far-field transport calculations in performance assessment is to model the time dependent contaminant distributions in the rock mass. This information is needed for estimating releases to the surface environment.

There is the potential for radionuclide reconcentration to occur at the geosphere-biosphere interface or, less likely, in the far-field (see GEN-31 “Radionuclide reconcentration”). This could occur, for example, if discharge occurred to an organic-rich sediment with a high radionuclide sorption capacity. Over time, sorption onto the sediment would cause the radionuclide concentration in the sediment to rise. A subsequent change in surface conditions (e.g. a sudden increase in erosion rates) could then cause quasi-instantaneous releases of high concentrations of radionuclides.

Release to the biosphere for radionuclides dissolved in the groundwaters will normally occur at groundwater discharge zones, and this might be in the form of springs, seeps or to rivers and lakes. The radionuclide release pattern from the far-field is thus highly spatially heterogeneous and will be time dependent.

Because release to the surface environment occurs near the end of the exposure pathway for repository-derived radionuclides, it is affected by all essential preceding release, transport and retardation processes.

Origin in the repository system:

The distribution and subsequent release of radionuclides to the surface environment is dependent on the mechanisms and rates of all processes that control release, transport and retardation in both the near and far-fields.

In particular, radionuclide distribution and release is affected by waste package failure; wasteform dissolution; near-field transport processes; groundwater flow and dispersion in the far-field rock; far-field retardation processes; radioactive decay; gas generation and transport; and colloid generation and transport in the far-field.

Impact on the repository system:

The distribution and subsequent release of radionuclides from the far-field are fundamental controls on the amount of repository-derived radionuclides in the biosphere and their distributions and, ultimately, on the radionuclide exposure to the surface environment.

Bibliographic references:

NEA (1991) Heterogeneity of groundwater flow and site characterisation. NEA/OECD.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEP 3.2 (Contaminant release/migration factors) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-12**

Name: **EARTH TIDES**

Short description:

Earth tides are small pressure variations in the groundwater flow system caused by changes to the gravitational field due to the relative movements of the Earth, the Sun and the Moon. Earth tides may have an influence on the transport and retardation of radionuclides in the far-field.

Technical description:

The relative movement of the Earth and the moon in their orbits around the Sun cause changes to the gravitational field experienced at or near the Earth's surface and are responsible for the tides observed in the seas and oceans. This gravitational effect can also affect other liquid bodies and analogous 'tides' are well known in surface lakes and rivers, and underground magma chambers, for example.

The same gravitational effect also impacts on the solid crust and the ductile mantle of the Earth. In the crust, the rock mass is deformed in so-called 'Earth tides' and any ground or porewaters contained in the rock porosity respond to this deformation, as well as being directly affected by the gravitational effect themselves. As a consequence, water levels in boreholes have been observed to change in response (e.g. Bredehoeft 1967; Millich and Neugebauer, 1993). More recently, Earth tide effects have been recorded at the Äspö Hard Rock Laboratory in Sweden and at the Grimsel Test Site in Switzerland (Kuhlmann et al., 1991). This is analogous to the 'seismic pumping' phenomenon (Muir-Wood and King, 1993) whereby groundwater levels responds to earthquakes (see GEN-17 "Faulting").

Earth tides potentially can influence solute transport in fractured rock via two separate processes. Firstly, because the effect depends on the stiffness of the rock, it is expected that a pulsating exchange of groundwater and dissolved species is induced between fractures and the rock matrix. However, once in the rock matrix, some dissolved species would be retarded due to the irreversibility of hydrodynamic dispersion (see GEN-10 "Radionuclide dispersion") and by sorption onto mineral surfaces (see GEN-34 "Radionuclide sorption"). In this way, Earth tides are expected to enhance the normal rock matrix retardation processes (see GEN-25 "Matrix diffusion"). Secondly, tidal pressure variations may occur within the plane of the fracture due to lateral variations in fracture stiffness. The corresponding pressure gradients may enhance lateral groundwater and solute dispersion.

Although the theory suggests that Earth tides could potentially have an effect on radionuclide transport in the far-field, it is not clear if the effect will be significant. Observations of radon emanations from the ground suggest that Earth tides can have a measurable impact on natural radionuclide transport through rock (Lenzen and Neugebauer, in press). Also, modelling studies (Kuhlmann et al., 1991) have concluded that Earth tides may increase the effectiveness of matrix diffusion.

It seems likely that the Earth tide effect would be of very minor importance compared to other processes (and their uncertainties) controlling groundwater flow and radionuclide transport. However, sensitivity analyses would be necessary to confirm this.

Standard performance assessments do not account for the Earth tide phenomenon and, thus, may not be a conservative modelling assumption.

Origin in the repository system:

Earth tides are induced in the solid rock by the relative movements of the Earth, the Sun and the Moon. Groundwaters contained in the rock respond to these tides and water levels can change. The groundwater response is thus partly controlled by the physical characteristics of the rock mass. The primary influence of Earth tides is thus external to the repository system.

Impact on the repository system:

Earth tides may influence the transport and retardation of radionuclides by either enhancing the matrix diffusion process or enhancing flow through fractures. Although the effect is real, it is not clear if it is a significant control on radionuclide transport in the far-field.

Bibliographic references:

Bredehoeft JD (1967) Response of well-aquifer systems to Earth tides. Journal of Geophysical Research, 72, 3075-3087.

Kuhlmann U, Vomvoris S and Hufschmied P (1991) Evaluation of Earth tide matrix diffusion. Proceedings of an NEA/SKI Symposium on Validation of Geosphere Flow and Transport Models (GEOVAL 1990), 173-181.

Lenzen M and Neugebauer HJ (in press) Strong correlation between earth tides and radon concentration in a gypsum mine in Walferdange, Luxembourg. Submitted to the Journal of Geophysical Research.

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NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 1.5.03 (Miscellaneous and FEPs of uncertain relevance) in the NEA International Database (NEA, 1998).

Production:

Version 2.0	30th June 2000	Tim McEwen and Bill Miller

Number: **GEN-13**

Name: **ELECTROCHEMICAL EFFECTS**

Short description:

Electrochemical gradients may be established in the waste package and in other parts of the engineered barrier system where two different metals occur close together in water saturated conditions or in response to natural electrical currents in the far-field rock. These gradients may affect metal corrosion and dissolution and, possibly, radionuclide transport.

Technical description:

Localised electrochemical gradients may be established in the waste package and in other parts of the engineered barrier system where two different metals occur close together in water saturated conditions (bimetallic corrosion). For example, they may occur in the spent fuel canister after failure due to the presence of both iron and copper metals in the canister structure. Electrochemical effects may also arise from different local micro-environments (e.g. Eh, pH) on the surface of the same metal barrier. The distribution of water, as well as the water chemistry, will affect the nature of the electrochemical gradients.

Localised electrochemical gradients in the near-field may be focussed on the metallic components of the engineered barriers which can provide preferential electrically conducting pathways. This mechanism may increase the general and localised (pitting and crevice) corrosion rates of the metals. For the example of iron and copper in basic solution, the standard electrochemical half reactions are as follows (from Krauskopf and Bird, 1995):

$$Fe + 2OH^- = Fe(OH)_2 + 2e^- \qquad -0.55\ V$$

$$2Cu + 2OH^- = Cu_2O + H_2O + 2e^- \qquad -0.36\ V$$

Balancing these reactions gives:

$$Fe + H_2O + Cu_2O = Fe(OH)_2 + 2Cu \qquad -0.19\ V$$

In other words, iron can reduce Cu_2O in the presence of an electrolyte (groundwater) to produce Cu, although other reactions such as the cathodic consumption of oxygen might provide a larger oxidation capacity than the Cu. Electrochemical gradients accelerating bimetallic corrosion of metals could be a significant process in the degradation of metallic components of the engineered barriers. However, this is a difficult process to quantify and, hence, is not normally explicitly accounted for in standard performance assessments.

Natural currents occurring in the ground, known as ‘telluric currents’, could also generate electrochemical gradients on a large scale. Telluric currents are principally generated in response to solar activity which creates ionospheric currents around the earth in relation to its magnetic field. Telluric currents are random, and cover a large range of frequencies. The frequencies below 1 Hz have a penetration depth which exceeds the repository depth and they therefore have potential significance for repository behaviour.

Since most rocks are poor electrical conductors, telluric currents flow mainly through water conducting features such as fractures or porous materials (such as the backfill). Theoretically, the development of an electric field in the rock (electrochemical gradient) can affect migration of radionuclides by electroosmosis and electrophoresis. Electroosmosis refers to the ability of an applied electrical field to induce fluid flow in a porous medium while electrophoresis refers to the migration of charged species in water in the presence of an electric field. Thus the large-scale telluric currents may potentially affect the transport of radionuclides in groundwater. However, this affect is likely to be minor compared to advection and, thus, may only be of significance in low permeability materials where advection does not operate. Even in this case, the majority of radionuclide migration will be dominated by normal diffusion in the chemical gradient (see GEN-9 “Diffusion”). Consequently, neither electroosmosis nor electrophoresis are routinely considered in performance assessments.

Origin in the repository system:

Localised electrochemical gradients my be established in the near-field due to the proximity of two different metals in water saturated conditions.

Large-scale electrical currents (telluric currents) could be established in the far-field in response to solar activity which creates ionospheric currents around the earth in relation to its magnetic field.

Impact on the repository system:

Localised electrochemical gradients may accelerate corrosion of the metallic components of the engineered barriers, although the effect is very difficult to quantify.

Large-scale electrical currents (telluric currents) theoretically could affect the mobility of radionuclide by the processes of electroosmosis and electrophoresis. However, these processes are unlikely ever to be significant in the repository environment.

Bibliographic references:

Krauskopf KB and Bird DK (1995) Introduction to Geochemistry. McGraw-Hill.

National Academy of Science (1986) The Earth’s Electrical Environment. US National Academy of Science.

Equivalent FEPs:

Relates to FEPs 1.5.03 (Miscellaneous and FEPs of uncertain relevance), 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.2.08 (Chemical/geochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 2.1.6.2 (Natural telluric electrochemical reactions) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-14**

Name: **ENHANCED ROCK FRACTURING**

Short description:

A hard host rock will contain many natural fractures. However new fractures will be created and some existing fractures modified due to repository construction and by later rock movements occurring in response to the stress field. This enhanced rock fracturing is more likely to occur in the near-field rock.

Technical description:

A hard (crystalline) host rock will inevitably contain a number of pre-existing fractures. However, the fracture network is likely to be somewhat altered by the creation of new fractures and by the modification of the pre-existing fractures during repository construction and by post-closure rock movements. This enhanced rock fracturing will occur around both the spent fuel repository and the SFR, and is more likely to occur in the near-field rock than in the far-field.

Construction of a repository is likely to employ blasting techniques for the shafts and tunnel boring machines (TBMs) or smooth-wall blasting for the underground tunnels. In the spent fuel repository, the deposition holes in the floors of the tunnels are likely to be drilled out. All of these construction processes will result in the development of additional fractures, although the extent to which such fractures might form depends critically on the physical characteristics of the rock, the construction technique used, how it is applied and on the design of the facility. Blasting, for example, even smooth-wall blasting is likely to result in more additional fractures than using a TBM. These newly formed fractures will extend a certain distance out into the near-field rock and define the engineered damaged zone which will affect physical and hydrogeological conditions around the repository (see GEN-15 “Excavation effects on the near-field rock”).

Experiments carried out at Äspö as part of the ZEDEX programme (SKB, 1996) provided information on the extent of the damage caused by the different methods of excavation. In addition to the damage to the walls of the excavation caused by the excavation method, the act of removing material will alter the stress conditions local to the excavation and create a region of disturbance along existing fractures. From modelling and observations of excavations, the majority of the perturbations to the stress field occur within a distance of a few tunnel radii from the excavation centre (Bell, 1992). The rock mass will respond to these changes by redistributing the stress around the opening. This movement will depend on a variety of factors, including the strength of the rock mass and the regional stress field (see GEN-17 “Faulting”). The extent to which this process will enhance the fracturing of the rock mass will depend on the magnitude of the stresses, the extent of existing fracturing and the strength of the rock mass.

After repository closure, additional stresses will be imposed on the near-field rock as the repository system moves towards physical, hydrogeological and chemical equilibria. In particular, stresses will be imposed by thermal loading of the rock mass (the thermal gradient), hydraulic resaturation of the void spaces and expansion of bentonite buffer and backfills.

Together by superposition (or individually), these stresses may cause new fractures to form in the near-field rock. However, in the spent fuel repository, it is likely that the thermal load may be the most likely cause of fracturing (see SFL-46 “Temperature of the near-field”). In the SFR, thermal effects will be less significant because of the lower temperatures generated by radioactive decay (see SFR-17 “Temperature of near-field”).

Enhanced fracturing will increase the fracture density and this is likely to increase the bulk hydraulic conductivity of the near-field rock. In the rock adjacent to horizontal tunnels, enhanced fracturing is likely to occur parallel to the tunnel axis and, possibly, preferentially below the floor of the tunnel. In the case of the vertical shafts the location of any enhanced fracturing will be controlled by the in situ stress field and the lithology, with a tendency for an increase in fracturing as stress concentrations increase with depth. There may, therefore, be only localised enhancement of fracturing around the shafts.

In the spent fuel repository, enhanced fracturing, probably on a very localised scale, will be produced around the deposition holes during their construction. This may be of minor significance and may be limited to the opening of microcracks and minor displacement of joints. Further enhancement, however, is possible during the thermal phase of the repository, when longitudinal fracturing is also possible. Under the compressional loading applied by the expanded bentonite these fractures may close. During cooling and contraction, there is a possibility that fractures may open.

Thus, in general, enhanced rock fracturing will tend to increase the groundwater fluxes in the near-field rock, close to the waste packages. However, other factors, such as pore blockage by expanding bentonite (see SFL-1 “Swelling of the bentonite buffer”), may limit the extent to which the enhanced fracturing actually does increase the transmissivity of the rock mass.

The rock and mineral surfaces freshly exposed by enhanced rock fracturing will increase the surface area available for rock-water interactions and for radionuclide sorption, and the volume of rock available for matrix diffusion. The freshly exposed minerals will increase bulk rock-water interaction rate and, if ferric minerals are present, will help to buffer the redox conditions and, thus, the chemistry of the near-field groundwaters.

The creation of newly formed fractures in the near-field rock during construction and post-closure potentially can affect the behaviour of the repository and, thus, needs to be considered in performance assessment modelling.

Other post-closure time-dependent stresses will be imposed on the rock mass from external forces, such as plate tectonics and climate change that will cause slow creep (see GEN-5 “Creeping of the rock mass”) and rapid fault movement (see GEN-17 “Faulting”). However, these processes are unlikely to cause significant generation of new fractures because the changes to the stress fields will occur over long periods of time and normally cause readjustments along pre-existing discontinuities.

Origin in the repository system:

Enhanced rock fracturing is caused largely by the excavation method employed during the construction phase, by the post-closure stresses that are imposed during the thermal peak and by the bentonite swelling, as well as by external processes. Thus the extent of enhanced fracturing is controlled by the strength of the rock (physical characteristics), the maximum temperature at the waste package outer surface (radionuclide content of waste) and the thermal conductivity of the engineered barrier materials and the rock.

Impact on the repository system:

Enhanced rock fracturing will be concentrated in the near-field rock, close to the repository excavations. The primary effect will be to increase the fracture density and the wetted surface area of the rock available for interaction with the groundwater and solutes.

The increased fracture density will tend to increase the bulk hydraulic conductivity of the near-field rock compared to the far-field rock which may influence the transport of radionuclides released from the engineered barriers.

The increased surface area will produce greater numbers of sorption sites and a larger volume or rock for matrix diffusion. The freshly exposed minerals will increase bulk rock-water interaction rate and, if ferrous iron minerals are present, will help to buffer the redox conditions and, thus, the chemistry of the near-field groundwaters.

Bibliographic references:

Bell FG (1992) Engineering in rock masses. Butterworth Heinemann.

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NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Shen B and Stephansson O (1996) Modelling of rock fracture propagation for nuclear waste disposal. SKI Technical Report 96:18.

SKB (1996) ZEDEX - A study of the zone of excavation disturbance for blasted and bored tunnels. SKB International Co-operation Report, 96-03. Three Volumes.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 1.2.02 (Deformation, elastic, plastic or brittle), 2.2.01 (Excavation disturbed zone, host rock), 2.2.06 (Mechanical processes and conditions in geosphere) and 2.2.10

(Thermal processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.8 (Enhanced rock fracturing) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Tim McEwen and Bill Miller

Number: **GEN-15**

Name: **EXCAVATION EFFECTS ON THE NEAR-FIELD ROCK**

Short description:

The rock mass around all of the underground openings in a repository excavated in hard, crystalline rock will be affected by the excavations in various ways. Changes to the affected rock mass are likely to be mechanical, hydrogeological and chemical in nature.

Technical description:

The excavation of the repository will cause fracturing of a volume of the near-field rock close to the repository openings. This volume of fractured rock is called the engineered damage zone. The impacts of the engineered damage zone on the near-field rock are likely to be time-dependent, and mechanical, hydrogeological and chemical in nature. Excavation effects will occur in the rock around both the spent fuel repository and the SFR.

The process of excavation can be considered as comprising two basic elements: breaking up of the intact rock mass and the subsequent removal of the spoil. The excavation results in the rock mass being disturbed and altered in an immediate and possibly a time-dependent manner due to the excavation process (e.g. drilling or blasting) and by the subsequent stress redistribution. The process of breaking up the intact rock can be achieved in different ways and these vary in the extent to which they might damage the surrounding rock mass. The drill and blast method is less controllable than full face boring using a tunnel boring machine (TBM), and will tend to create more peripheral damage, but can still achieve very good results with well-designed blast patterns and suitable charge densities in perimeter holes to minimise the energy input to the walls of the excavation (e.g. smooth wall blasting - Kelsall et al. 1984).

Damage to the surrounding rock and the redistribution of in situ stresses may result in the development of new fractures (see GEN-14 “Enhanced rock fracturing”), the reactivation of healed fractures or displacements on existing, open fractures, altering their apertures in the process (see GEN-17 “Faulting”). These effects are likely to modify the hydraulic conditions in the rock surrounding the underground opening. The impact of excavation damage will possibly create a zone of enhanced radial or longitudinal hydraulic conductivity. This zone could provide a preferential pathway for radionuclide migration to the prevailing groundwater flow system and must be taken into account when sealing the different parts of the repository.

The response of the groundwater system to the excavation process can be considered as the introduction of a new set of boundary conditions and as local modification to the flow properties of the rock (see GEN-23 “Groundwater flow”). The opening of the excavation will introduce an atmospheric pressure boundary condition within the rock mass and set up a head difference within the system. Groundwater flow will be altered for a considerable distance around the excavation as water flows into the void and is pumped out to the surface (during the construction and operation stages). This can be thought of as a de-pressurisation of the groundwater system around the excavation, and it will also have the effect of changing the effective stress of the rock mass and promoting consolidation, i.e. there will a tendency for certain fractures to close.

The chemical effects that are associated with the excavation of a repository relate to the increased surface area of fresh rock for rock-water interactions; the introduction of fluids (including gases) used during the construction process that will not be in equilibrium with the existing groundwater; the introduction of atmospheric gases at depth, containing oxygen, carbon dioxide and other gases that are out of equilibrium with those in the groundwater; and the introduction of construction materials such as grout, concrete, metal reinforcement and resins/bonding agents.

The groundwater chemistry in the near-field rock will thus be affected by the repository excavations. Initially, the near-field pore-waters are likely to become oxidising as they react with the atmospheric gases. The waters may also become acidic as sulphides present in the rock oxidise. However, this will be a transient phenomenon and, after closure, the abundant ferrous iron in the system will buffer the redox conditions, returning them to the natural ambient Eh and pH values.

Groundwater reactions with the repository construction materials will be longer term and the effects will depend on the nature of the materials used and their degradation rates and processes. In the SFR, these reactions will be dominated by the large volume of cement present (see SFR-20 “Groundwater chemistry in the near-field”).

Origin in the repository system:

Excavation effects on the near-field rock are induced by enhanced rock fracturing caused by the blasting and tunnelling operations to construct the underground openings. The extent of these excavation effects is controlled by the width of the engineered damage zone which, itself, is a function of the mechanical properties of the rock, the virgin rock stresses and the excavation method employed.

Impact on the repository system:

The engineered damage zone will impact on the repository system in various ways that can be divided into mechanical, hydrogeological and chemical. The mechanical effects relate to the formation, opening and closure of fractures. The hydrogeological effects relate to a general increase in the bulk hydraulic conductivity of the engineered damage zone compared to the far-field rock. This change in hydraulic properties will impact on the groundwater flow and radionuclide transport processes. The chemical effects relate to an increase in the fresh rock surfaces available for rock-water interaction, and the reactions between the groundwater and any gas phases that accumulate in the engineered damage zone during the construction and operation phase. These reactions will have the effect of creating transient changes to the chemistry of the near-field rock porewaters, initially making them oxidising and acidic.

Bibliographic references:

Kelsall PC, Case JB and Chabanns CR (1984) Evaluation of excavation-induced changes in rock permeability. International Journal of Rock Mechanics, 21, 123-135.

NAGRA (1994) Kristallin-1 Safest Assessment Report. Nagra Technical Report, NTB 93-22.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D, McLeod R and McEwen T (1998) An evaluation of repository-induced disturbances for an SKB-type repository. SKI Technical Report, 98:20.

SKB (1996) ZEDEX - A study of the zone of excavation disturbance for blasted and bored tunnels. SKB International Co-operation Report, 96-03. Three Volumes.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.2.01 (Excavation disturbed zone, host rock) and 2.2.06 (Mechanical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.2.1 (Excavation/backfilling effects on nearby rock) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-16**

Name: **EXTERNAL FLOW BOUNDARY CONDITIONS**

Short description:

The external flow boundary conditions of the hydrogeological system around a repository control the location and amount of recharge and discharge, and are a control on the geometry of the flow system. The external flow boundary conditions are, thus, important to define for modelling groundwater flow and radionuclide transport in the far-field.

Technical description:

The external flow boundary conditions of the hydrogeological system around a repository control the location and amount of recharge and discharge, and are a control on the geometry of the flow system.

The primary factors which determine the external flow boundary conditions are the topography of the land surface, sea-level and the location of the coast, amount of precipitation and evapotranspiration, and the distribution of surface permeability which controls the amount and location of infiltration. Between them, these factors will determine the location and rate of recharge and discharge.

The fluid density and temperature of the recharge or discharge from the system can also be considered but these parameters have a less significant role in controlling the groundwater flow system.

Certain of these controlling factors will be time dependent. For example, precipitation may show seasonal variations in magnitude and longer-term variations in the future due to climate change. Likewise, changes to sea-level (see GEN-33 “Sea level changes”) will result from climate change and the topography will change due to uplift and erosion (see GEN-39 “Uplift and subsidence”). Human actions may alter the flow boundary conditions by building dams, other construction work, extracting groundwater from wells, and changing agricultural practices and surface vegetation types.

The presence of a repository will have little or no impact on the external boundary conditions. The most likely possible impacts may be a delay in the formation and a limited reduction in the thickness of permafrost during the next glacial cycle due to radiogenic heating from the spent-fuel repository (see GEN-26 “Permafrost”).

In hydrogeological modelling, the appropriate external boundary conditions to consider are dependent on the nature and size of the area to be investigated; for example, the flow boundary conditions considered in the regional scale hydrogeological model will be different to those considered for the site scale model. The location of these boundaries can be selected to simplify their descriptions for the purposes of numerical modelling, but will always be a simplification of the real system.

The external flow boundary conditions provide the primary driving forces for the flow of groundwater through the hydrogeological system around the repository (at the scale of interest) and hence are an important factor in any performance assessment. The main external flow boundary conditions considered in performance assessment are topography, sea level and the amount and distribution of recharge.

External flow boundary conditions are clearly site specific and, thus, assessment specific. All performance assessments of real or generic sites must define external flow boundary conditions for the quantification of groundwater flow and radionuclide transport through the far-field. Furthermore, assessments must consider the spatial and temporal variations in flow boundary conditions due to climate change (e.g. King-Clayton et al., 1995).

Origin in the repository system:

The flow boundary conditions are controlled by parameters external to the repository system such as the surface topography and the magnitude of rainfall. The origins of the external boundary conditions are thus related to climatic and topographic factors that are time dependent.

Impact on the repository system:

The external flow boundary conditions are of primary significance in performance assessment because they control the groundwater flow geometry, recharge and discharge areas, flowpath lengths, and flow rates through the repository and the far-field rock and, thus, the transport of radionuclides to the surface.

The changes with time of the external flow boundary conditions will also be significant in determining the quantity and location of any discharges. The development of future climate scenarios allows the impact of these changes to be assessed

Bibliographic references:

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.3 (Surface environment) in the NEA International Database (NEA, 1998).

Relates to FEPs 5.27 (Human induced actions on groundwater recharge) and 5.46 (Groundwater recharge/discharge) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-17**

Name: **FAULTING**

Short description:

Faulting encompasses both the development of new faults, but also the movement on and growth of existing faults. Either of these actions could alter the groundwater flow conditions and affect the mechanical stability of the rock around the repository.

Technical description:

The response of the rock mass to changes in the stress field may either be sudden fracturing and faulting (see GEN-17 “Faulting”) or slow creeping (see GEN-5 “Creeping of the rock mass”). Fracturing refers to the rapid breaking of the rock mass and faulting refers to the movement along a fracture. While new fractures and faults can be created, it is more likely that changes to the stress field would be accommodated by movement along pre-existing faults and fractures.

Changes to the stress field can occur over a range of temporal and spatial scales, and can be compressional or extensional (see GEN-36 “Stress field”). Large spatial scale (super-regional), slow changes in the stress field can occur in response to plate tectonic processes while, on a regional scale, slow changes can result from ice loading and to the lowering of sea level, both of which have an effect on the crustal rocks and on the upper mantle. These long-term processes are of relevance only to the spent fuel repository because of the long time periods considered in performance assessments for this facility. For the SFR, relevant changes to the stress field result from continued isostatic uplift of the land and the change from submarine to subaerial conditions.

One a smaller scale, faulting in the vicinity of the repository could be initiated by changes in the local stress field (see GEN-36 “Stress field”) caused by the repository excavation, the thermal load or swelling of the engineered barriers.

Plate tectonic processes cause a relatively low level of seismic activity and associated fault movement in Sweden (Muir-Wood, 1993). The assumption of future fault movement in Sweden in relation to the siting of a repository is that such movement will occur almost exclusively on existing faults. The crystalline basement in Sweden contains a large number of such faults or fracture zones, the country is not seismically active and there is unlikely to be a rotation in the direction of maximum horizontal stress within the next million years. The assumption of fault movement is, therefore, generally valid, since the energy necessary to develop a new fault is many times that needed to cause movement on a suitably oriented existing fault. The possibility of a new fault forming through a repository due to super-regional scale tectonic processes is small.

Processes that take place on a regional scale could result in the formation of new faults or, again more probably, on the reactivation of existing faults. Loading by continental ice sheets up to 2 or 3 km thick depresses the crust and causes a compensating flow within the upper mantle (see GEN-21 “Glaciation”). It is thought that such loading may suppress the normal levels of seismic activity associated with tectonic activity and may also cause movement on existing faults, as the crustal rocks respond to their new state of stress. Several hundred metres of vertical crustal depression has taken place in past glaciations over northern parts of Sweden. Large seismic events, together with associated reverse fault movement, is thought to have taken place during rapid retreat of the ice sheets at the end of the last glaciation. Evidence of such faulting, is found, for example, in northern Sweden (Lagerbäck, 1988).

Modelling of the effect of ice loading on the rock mass (Hanson et al., 1995) has demonstrated how fracture apertures may be changed without corresponding movement along the fractures, thereby altering their aperture distributions and changing their hydraulic properties.

On a smaller spatial scale and on shorter time periods, stress changes in the host rock will be induced by the construction of the repository, the thermal gradient from the waste and swelling of the engineered barrier materials. These stress changes could cause displacement on small fractures and joints.

Excavation of the repository openings introduces a stress inhomogeneity into the rock mass (see GEN-15 “Excavation effects on the near-field rock”). Although, by itself, repository construction is very unlikely to result in anything more than local stress redistributions, which may be accompanied by microseismic activity, such at as Loviisa in Finland (Saari, 1996), future fault movement may be influenced by the presence of the repository.

Faulting, due to any cause, may alter the bulk permeability of the rock, alter or create short-circuits in the flow paths close to the repository and create new pathways through the repository. Newly formed or regenerated faults may enhance groundwater fluxes and destabilise and damage engineered barriers, thereby decreasing radionuclide transport times.

Existing fractures can act as hydraulic ‘pumps’ if movement occurs along them or their apertures change in response to seismic activity or glaciation (Muir-Wood and King, 1993). Such a pumping mechanism can introduce near-surface waters (oxidising) into the near-field or, in contrast, could pump contaminated waters away from the repository into the far-field. Groundwater pumping in either direction by such a mechanism could potentially affect radionuclide transport if the waste package has failed.

Faulting could affect the mechanical stability of the rock adjacent to the repository and, eventually, could lead to collapse of part of the repository openings. However, complete cave-in would only occur if significant void space volumes were present in the repository, which is unlikely (see GEN-3 “Cave-in”). The mechanical damage to the rock can also liberate particulates and colloids which could then be transported by moving groundwater.

Origin in the repository system:

Faulting occurs in response to changes in the host rock stress field. The extent and mode of faulting depends on the rate of changes in the stress field, the existing fracture network (number and orientation) and the mechanical strength of the bulk rock. In most geological environments, it is likely that faulting will involve movement along pre-existing faults and fractures, rather than generation of new fractures or faults/

Impact on the repository system:

Faulting will impact on the hydrogeological characteristics of the host rock. Movement along pre-existing fractures (or formation of new fractures) may create short-circuit pathways from the repository through the far-field.

Faulting can also affect the mechanical stability of the host rock and can lead to cave-in of the repository excavations.

Bibliographic references:

Hanson H, Shen B, Stephansson O and Jing L (1995) Rock mechanics modelling for the stability and safety of a nuclear waste repository. SKI Technical Report, 95:41.

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Lagerbäck O (1988) Post-glacial faulting and palaeoseismicity in the Lansjärv area, northern Sweden. SKB Technical Report, TR 88-25.

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Muir-Wood R and King GCP (1993) Hydrological signatures of earthquake strain. Journal of Geophysical Research, 98, 22035-22068.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Saari J (1996) Seismic emissions induced by excavations of the rock repository at Loviisa. Posiva Technical Report, 96-05.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 1.2.02 (Deformation, elastic, plastic or brittle), 2.2.04 (Discontinuities, large scale in geosphere) and 2.2.06 (Mechanical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.6 (Faulting) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Tim McEwen and Bill Miller

Number: **GEN-18**

Name: **GAS FLOW IN THE FAR-FIELD**

Short description:

Gases may exist in the far-field. These gases may be dissolved in the groundwater but they may also occur as discrete gas bubbles, in which case they will travel through the groundwater driven by buoyancy. These bubbles potentially may carry radionuclides and may alter the flow behaviour of the groundwater.

Technical description:

Gases formed by degradation of the waste and the engineered barriers may occur in the far-field rock. Typically, these gases would be H_2 from the anaerobic degradation of steel and radiolysis of water but other gases (such as CO_2, CH_4 and H_2) may also be present, especially around the SFR repository due to the degradation of organic materials.

Small volumes of gas may be generated directly in the far-field by microbial degradation of organic material present in the groundwaters and by radiolysis from radionuclides that have migrated from the repository (see GEN-19 “Gas generation in the far-field”).

Initially, these gases will be dissolved in the water and, thus, their transport behaviour will be the same as any other dissolved species. However, as the groundwater migrates upwards and the confining pressures decreases, the solubility of the gas may be exceeded and discrete gas bubbles may form.

The gas may contain radionuclides directly, e.g. as ^{14}C in CO_2 or as tritium in H_2. In this case, the gas bubbles will provide a rapid transport mechanism for these nuclides because the buoyancy of the bubbles will allow them to travel faster than the mean groundwater flow rate. In addition, other nuclides may be carried with the bubbles if they occur in colloidal form because colloids are sometimes preferentially attached to the gas-water interface (i.e. the bubble surface).

The formation of gas bubbles could also affect the transport of radionuclides dissolved in the groundwater by changing the groundwater flow behaviour. This could occur by a number of mechanisms. First, bubble formation and movement may change the apparent viscosity of the groundwater and induce or accelerate groundwater movement, this is most likely to occur if large volumes of free gas are formed. Second, bubbles may push groundwater in front of them along preferred directions, this is most likely to occur if a bubble is similar in size to the aperture of the advecting fractures. Third, immobile bubbles may change the direction of water flow by effectively sealing certain pathways (an air-lock), this might occur if the fracture changes direction allowing gas to accumulate in the highest part of the fracture.

In the far-field rock, gas bubbles will move along the fractures in much the same way the groundwater does, moving in response to a pressure gradient. Because the fractures may carry both groundwater and a free gas phase, two-phase transport models may be required to evaluate the significance of gas transport for repository safety, although this has not been done routinely in many performance assessments. However, gas migration away from the proposed Nirex cementitious L/ILW repository has been considered in detail (Nirex 1994, 1997).

The significance of gas bubbles for radionuclide transport and for changing groundwater flow patterns is very largely dependent on the volume of gas present. Since the SFR repository is more likely to generate large volumes of gas than the spent fuel repository, gas transport in the far-field is more relevant to the behaviour of the SFR than it is to the spent fuel repository (see SFR-11 “Gas generation in the repository”).

The chemistry of the groundwater will be altered by the exsolution of gas bubbles and this could have a subsequent effect on the solubility of radionuclides in solution. If other physico-chemical changes occur to the far-field environment, then the gas bubbles potentially could dissolve back into the groundwater.

Origin in the repository system:

The development of a free gas phase is controlled by the solubility of the gaseous species. Gas generation rates are much higher in the SFR than the spent fuel repository and, thus, far-field gas transport as bubbles is most likely to occur around the SFR.

Gas pressures and gas transport mechanisms in the far-field are dependent on the hydraulic characteristics of the fracture network in the rock mass.

Impact on the repository system:

The development of a mobile free gas phase in the far-field may have significant impacts on radionuclide transport. Radionuclides incorporated directly in the gas or attached to the bubble surface will move faster than the mean groundwater flow rate.

Gas bubbles may significantly affect groundwater flow behaviour by changing flow direction, or by inhibiting or accelerating groundwater flow.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Nirex (1994) Post-closure performance assessment: gas generation and migration. Nirex Science Report, S/94/003.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Science Report, S/97/012, Four Volumes.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.2.11 (Gas sources and effects in geosphere) and 3.2.09 (Gas-mediated transport of radionuclides) in the NEA International Database (NEA, 1998).

Relates to FEP 6.2 (Gas transport) in the in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-19**

Name: **GAS GENERATION IN THE FAR-FIELD**

Short description:

Gas may be generated in the far-field by radiolysis of porewaters and by microbial degradation of organics. This gas potentially may have a small influence on radionuclide transport and retardation in the far-field but the largest gas effect will be from gas formed in the repository that migrates into the far-field.

Technical description:

Gas generation in the far-field may occur by two processes: radiolysis of porewaters and microbial degradation of organics. These processes can occur in the far-field rock of both the SFR and spent fuel repositories.

Hydrogen gas may be generated by radiolysis of the groundwater in the far-field rock due to ionising radiation emitted from mobile radionuclides (dissolved or colloidal) or from radionuclide-bearing precipitates that have formed in the far-field fracture surfaces. Hydrogen gas production by radiolysis is likely to be most significant in the spent fuel repository, after the canister has failed, because of the higher activity of the spent fuel compared to the wastes in the SFR.

Most species produced by alpha radiolysis are extremely reactive or unstable, and quickly convert to more stable or inert species (see GEN-30 “Radiolysis”). However, the volume of gas produced by this process will be extremely small and could not occur until after the waste package has failed. Under normal conditions this would not occur until long after the repository was closed, by which time radioactive decay will have significantly reduced the levels radioactivity. Consequently, this process may only be of limited significant in early canister failure scenarios for the spent fuel repository.

Microbial decomposition of organic material in the far-field rock could also lead to gas generation. Microbial populations will occur in the repository and the host rock but their viability will be restricted by the availability of nutrients. In the far-field, organic material may be present naturally in the groundwater or could be supplied in the spent fuel repository by the small organic content (0.4 vol.%) of the bentonite and in the SFR from the large amounts of organic material (e.g. cellulose) in the waste.

This material could potentially be transported to the far-field where it may subsequently undergo microbial decomposition to generate H_2, CO_2 and CH_4. Alternative microbially mediated gas generating reactions could produce other gases, depending on the microbial species present and the availability of organic carbon and other nutrients. Because of the very large organic content of the SFR, gas production in the far-field of this repository could be more significant than for the spent fuel repository.

The gas generated in the far-field rock by the above processes will dissolve into the groundwater and thus will influence the groundwater chemistry. If gas accumulation exceeds the solubility limits, then a gas phase may be produced. Gas bubbles forming in the far-field rock could affect groundwater flow (see GEN 19 “Gas flow in the far-field”).

However, although it is theoretically possible that gases would be generated in the far-field rock by these processes, it is evident that the volumes evolved will be insignificant compared to the volume of gas produced at the waste package and the engineered barriers. This means that the majority of gas found in the far-field rock will have formed in the repository and subsequently migrated to the far-field rock, rather than being formed there. Consequently, gas generation in the far-field rock is considered to be of no significance to repository safety for either the spent fuel repository or the SFR.

Gas generation in the far-field rock is not usually considered in performance assessments for spent fuel repositories and was not considered in SITE-94.

Origin in the repository system:

Gas generation in the far-field rock by radiolytic decomposition of the porewaters is largely controlled by the lifetime of the waste package and the transport of radionuclides to the far-field. After failure, radiolytic decomposition will be dependent on the ‘age’ of the waste and thus the level of radiation emitted. Gas generation by microbial degradation of organic material is dependent on there being viable populations of microbes and a supply of organic material.

Impact on the repository system:

Gas generation in the far-field rock may potentially have an effect on the groundwater chemistry and on far-field groundwater flow. However, these impacts are expected to be very limited and eclipsed by gas generated in the repository which subsequently migrates to the far-field.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.1.11 (Gas sources and effects in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.4 (Gas generation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-20**

Name: **GAS GENERATION IN THE NEAR-FIELD ROCK**

Short description:

Gas may be generated in the near-field rock by corrosion of steel, radiolysis of porewaters and by microbial degradation of organics. In addition, for a short period after closure, air will be present in the rock that can be considered as a gas source during the resaturation phase. This gas potentially may have a small influence on radionuclide transport and retardation in the near-field but the largest gas effect will be from gas formed in the repository that migrates into the near-field.

Technical description:

Gas generation in the near-field rock may occur by three processes: anaerobic corrosion of steel reinforcements (e.g. roof supports and rock bolts), radiolysis of porewaters and microbial degradation of organics. These processes can occur in the near-field rock of both the SFR and spent fuel repositories.

It is likely that some steel reinforcements will be required to keep open the repository excavations during the operational phase. This steel will react directly with groundwater to produce hydrogen under anaerobic conditions, after the repository has resaturated and chemically-reducing conditions have been established. This reaction can occur in both the SFR and spent fuel repositories. However, in the SFR, the high pH environment caused by the cement will tend to reduce the anaerobic corrosion rate of iron.

Hydrogen gas may also be generated by radiolysis of the groundwater in the near-field rock due to ionising radiation emitted from mobile radionuclides (dissolved or colloidal) or from radionuclide-bearing precipitates that have formed on fracture surfaces (see GEN-30 “Radiolysis”). Hydrogen gas production by radiolysis will be most significant in the spent fuel repository, after the canister has failed, because of the higher activity of the spent fuel compared to the wastes in the SFR.

Most species produced by alpha radiolysis are extremely reactive or unstable, and quickly convert to more stable or inert species. However, the volume of gas produced by this process will be limited and could not occur until after the waste package has failed. Under normal conditions this would not occur until long after the repository was closed, by which time radioactive decay will have significantly reduced the levels of radioactivity. Consequently, this process may only be significant in early canister failure scenarios for the spent fuel repository.

Microbial decomposition of organic material in the near-field rock could also lead to gas generation. Microbial populations will occur in the repository but their viability will be restricted by the availability of nutrients. In the near-field, organic material may be present naturally in the groundwater or could be supplied in the spent fuel repository by the small organic content (0.4 vol.%) of the bentonite and in the SFR from the large amounts of organic material (e.g. cellulose) in the waste. This material could potentially be transported to the near-field rock where it may subsequently undergo microbial decomposition to generate H_2, CO_2 and CH_4. Alternative microbially mediated gas generating reactions could produce other gases, depending on the microbial species present and the availability of organic carbon and other nutrients. Because of the very large organic content of the SFR, gas production in this repository is more significant than for the spent fuel repository.

The gas generated in the near-field rock by the above processes will dissolve into the groundwater and thus will influence the groundwater chemistry. If gas accumulation exceeds the solubility limits, then a separate gas phase may be produced. Gas bubbles forming in the near-field rock could affect water flow at the interface between the far-field rock and the buffer and backfill. However, although it is probable that gases would be generated in the near-field rock by these processes, it is evident that the volumes evolved will be insignificant compared to the volume of gas produced in the waste package and the engineered barriers. This means that the majority of gas found in the near-field rock was formed in the repository and migrated to the near-field rock, rather than being formed there. Consequently, gas generation in the near-field rock is considered to be of minor significance to repository safety for either the spent fuel repository or the SFR.

Gas generation in the near-field rock is not usually considered in performance assessments for spent fuel repositories and was not considered in SITE-94.

An additional supply of gas will come from the air trapped during the construction phase in pores and fractures in the rock, including those in the engineered damaged zone. After repository closure, this gas will diffuse out of the near-field rock into the engineered barriers and, thus, should be considered as a gas source, at least during the resaturation phase (see GEN-32 “Hydraulic resaturation of the near-field rock”).

The impact of this air could be significant for understanding repository behaviour in the initial post-closure period because the air is likely to be oxidising and thus is a significant short-term control on the near-field chemical conditions. The presence of this air will also affect the resaturation process and rate.

Origin in the repository system:

Gas generation in the near-field rock by anaerobic corrosion of the steel reinforcements will be controlled by the chemistry of the groundwaters and the temperature. Gas generation by radiolytic decomposition of the porewaters is largely controlled by the lifetime of the waste package. After failure, radiolytic decomposition will be dependent on the ‘age’ of the waste and thus the level of radiation emitted. Gas generation by microbial degradation of organic material is dependent on there being viable populations of microbes and a supply of organic material. This is most significant for the SFR.

Trapped air released from pores, fractures and cracks after closure will be controlled by the volume of air-filled fractures in the rock that occur naturally and in the engineered damaged zone. The duration of air supply by this mechanism is controlled by the rate of resaturation which is, in turn, a function of the hydraulic properties of the host-rock.

Impact on the repository system:

Gas generation in the near-field rock will affect the groundwater chemistry and water flow at the interface between the far-field rock and the engineered barriers. However, the impacts are expected to be very limited and eclipsed by gas generation in the waste package.

Air released from pores, fractures and cracks after closure will control the chemistry (especially redox conditions) of the near-field for an initial period. This air will also affect the resaturation process and rate.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.1.11 (Gas sources and effects in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.4 (Gas generation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-21**

Name: **GLACIATION**

Short description:

Thick ice sheets are expected to cover part or the whole of Sweden on several occasions over the next million years. These ice sheets are likely to have a considerable influence on the behaviour of the repository system and the surrounding rock mass.

Technical description:

Glaciation involves many different processes (causes and effects) on the repository system but, for convenience, these processes are generally considered together. In scenario analysis in performance assessment, glaciation is usually considered as an external disturbance (EFEP).

The world's climate is expected to be characterised by periods of glaciation over the next million years, as has been the situation since the beginning of the Quaternary period, more than 2 million years ago. Evidence indicates that the timing and severity of climate changes may be determined by periodic changes in the Earth's orbit around the Sun (governed by the Milankovitch orbital parameters) which vary the amount of solar radiation reaching the Earth. The known periodicity of these parameters, together with other climate factors, such as atmospheric CO_2 levels, makes it possible to make estimates of the future climate sequence, which includes multiple future glaciations.

In one estimation, full glaciation in Sweden is anticipated 60 000 in the future (e.g. King-Clayton et al., 1995). Hence glaciation is a significant process for the spent fuel repository because this time is within the period of interest for performance assessments for this repository. In the case of the SFR, this time is beyond the period of interest for quantitative performance assessments and, consequently, glaciation is not particularly significant for the SFR.

Evidence from the last glaciation in Scandinavia indicates that during full glacial conditions the ice thickness may exceed 3 km in the central parts of the ice sheet and that the weight of the ice depresses the Earth's surface by several hundreds of metres (King-Clayton et al., 1995). The ice load affects the regional stress field (see GEN-36 “Stress field”), which in turn may induce fault movements or the creation of new fractures (see GEN-17 “Faulting”). Such movements would be associated with seismic events. The development of tensional forces or shearing on dipping fractures may increase their apertures and thereby alter their hydraulic conductivities. Glaciation will have a large impact on the magnitude of principal stresses, but only minor effects on stress orientations. The ice will also tend to close horizontal fracture zones and may have an impact on the apertures of vertical fractures, depending on the stress field.

The ice sheet itself and the accompanying frozen ground (permafrost) beneath the majority of the ice sheet (the cold-based portion) may constitute a barrier to groundwater flow and to heat loss (see GEN-26 “Permafrost”). If the basal transmissivity of the till and bedrock below the ice is low, water pressures may rise to levels equalling the ice pressure inducing the formation of major conduits in the subglacial material. The central parts of the ice sheet are likely to be warm-based and could permit groundwater recharge to take place, possible to great depths if high groundwater heads are generated at the base of the ice sheet. Discharge of groundwater is likely to take place close to and beyond the frontal parts of the ice sheet. Its location being determined, in part, by the presence, or otherwise, of permafrost and its thickness. Excessive recharge at the margin of the ice sheet could provide direct recharge of oxidising water to considerable depths in conductive fracture zones (e.g. Glynn and Voss, 1996). If the permeability at and beyond the rim of the ice is low, e.g. due to permafrost, the water pressures may again build up resulting in hydrofracturing of the ice or the rock mass. As the ice sheet advances, these induced fractures may increase their aperture and depth due to freezing of subglacial meltwater (King-Clayton et al., 1997).

The development of ice sheets will transfer large volumes of water from the sea to the land and will result in a global (eustatic) lowering of mean sea level (see GEN-33 “Sea level changes”). A Fennoscandian ice sheet extending to the Stockholm-Helsinki region would, for example, correspond to a lowering of the sea level of approximately 85 m. The maximum probable decrease in eustatic sea level is likely to be 140 m. Immediately following a deglaciation, but before isostatic uplift has had much impact on the land surface, the sea level in the Stockholm-Helsinki could be some 100 m higher than the present highest shoreline (see GEN-39 “Uplift and subsidence”).

The current topography in parts of Sweden where a repository is likely to be located is subdued, and the widespread erosional effects of any future ice sheets will probably be minimal (see GEN-29 “Erosion and weathering”). However, considerable localised erosion may take place, with the production of canyons cut into the rock mass, possibly preferentially developed along fracture zones.

Glaciation can have numerous and significant impacts on the repository system, such as the possibility for enhanced transport of radionuclides by changing groundwater flow paths, rates and chemistries; the possibility of direct mechanical impact to borehole and shaft seals; and changes to stress conditions in the near-field. There are a number of uncertainties when evaluating these impacts in performance assessment. The most obvious relates to the timing and magnitude of glacial events (i.e. time of maximum glaciation and extent of maximum advance). Other uncertainties relate to the magnitude of the impacts on the repository system caused by glaciation, such as the influence on groundwater flow in the far-field and its chemistry (related to changes in recharge).

Origin in the repository system:

Glaciation is one consequence of natural, long-term climate changes that are, themselves, caused by variations in the solar radiation reaching the Earth’s surface. As such, glaciation is caused by processes external to the repository.

Impact on the repository system:

Glaciation will result in a thick ice sheet covering the land surface in the vicinity of the repository. It may also be accompanied by a thick layer of permafrost extending down in to the ground. The consequences of the weight of the ice sheet on the host rock will be to produce high stresses at depth and the possibility of movement on fractures and, less likely, the formation of new fractures. Glaciation is also likely to increase the rate of erosion of the land surface.

The ice sheet and the permafrost both will have important influences on the groundwater flow system by modifying the locations of discharge and recharge, by altering the head distributions and by changing the hydraulic characteristics of the fracture network (via the stress field). However, the effect of ice sheets will be to limit the extent of permafrost development by acting as thermal insulators

The characteristics of groundwater bodies may also be affected. They may become pressurised due to the additional weight of the ice sheet and could be forced to considerable depths or drawn upwards due to lower pressures generated within sub-glacial ice tunnels. The chemistry of the groundwater may also be changed due to changes in the rock-water interaction times and by the variations in the recharge chemistry.

Bibliographic references:

Glynn P and Voss C (1996) Geochemical characterisation of Simpevarp groundwaters near the Äspö Hard Rock Laboratory, SKI Report 96:29.

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

King-Clayton L, Chapman N, Ericsson LO and Kautsky F (1997) Glaciation and hydrogeology - workshop proceedings. SKI Technical Report, 97:13.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 1.3.05 (Glacial and ice sheet effects, local) in the NEA International Database (NEA, 1998).

Relates to FEP 5.42 (Glaciation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-22**

Name: **FAR-FIELD GROUNDWATER CHEMISTRY**

Short description:

The initial chemistry of the host rock groundwaters reflects the natural, present-day system in the deep rock. This far-field groundwater chemistry is controlled largely by rock-water interactions and by mixing with waters from the near-field and from the surface. However, perturbations can occur due to climate change which can cause infiltration of sea-water or glacial meltwaters.

Technical description:

The rock-groundwater system in the far-field is required to provide a stable chemical environment for the repository that ensures long-term stability of the engineered barriers and to limit solubility of released radionuclides. Although all aspects of the groundwater chemistry are of interest, the properties of primary concern for these requirements are the redox and pH conditions, and the contents of species that buffer them; salinity (ionic strength); content of species aggressive to metal, bentonite and cement; content and nature of organic and inorganic complexing agents; and the colloidal populations.

The far-field groundwater chemistry initially is controlled by rock-water interactions between infiltrating groundwaters and the minerals in contact with them (see GEN-1 “Alteration and weathering along flow paths”). In general, in a crystalline or argillaceous host rock, these rock-water interactions cause waters to become chemically reducing, pH neutral to mildly alkaline and more saline with depth and time since infiltration. For groundwaters with a long residence time, rock-water interactions can generate a saline (brine) groundwater composition (see GEN-6 “Groundwater salinity changes”). However, the exact composition of deep groundwaters is strongly site specific, dependent on the mineralogical characteristics of the rock mass, the nature of the groundwater flow system and the composition of the recharge waters.

The variations in the major chemical variables that define the groundwater composition are due to a number of aspects of rock-water interactions, but several can be considered to be coupled, e.g. pH and Eh.

pH is a key variable in groundwater and water-rock systems and, in the far-field, is an important control on radionuclide solubility (see GEN-27 “Radionuclide precipitation and dissolution”), speciation and sorption (see GEN-34 “Radionuclide sorption”). In addition to affecting the overall magnitude of heterogeneous reactions, pH can also impact upon their rates through surface sorption and desorption processes. The pH of most groundwaters of interest to waste disposal is in the range 6 to 10. There are a number of factors which are important in governing the pH of groundwater, such as reactions with minerals in the rock, particularly calcite and aluminosilicates such as feldspars, micas and clays; the partial pressure of carbon dioxide (P_{CO2}), the concentration of so-called ‘conservative’ or ‘mobile’ ions such as chloride, and temperature.

Redox, like pH, is a key variable in groundwater systems and also affects solubility, speciation and sorption of redox-sensitive radionuclides. The redox state of a groundwater is generally expressed as Eh or pe. Redox levels in groundwater are controlled by the relative influx of oxidising water, the rate of groundwater flow and circulation, and the rate of consumption of oxidants (oxygen, ferric iron, sulphate etc.) by reduction reactions with reduced aqueous species or reduced chemical species in the rock. Potential reductants include: dissolved hydrogen, ferrous iron aqueous species, ferrous silicates, sulphides, oxides and carbonates; reduced sulphur aqueous species and sulphides; reduced manganese aqueous species and oxides; and organic materials such as bitumen, graphite, or aqueous organic compounds such as dissolved methane, carboxylic and humic acids etc. These reactions are often microbially mediated.

P_{CO2}, the partial pressure of carbon dioxide, represents the activity or thermodynamic concentration of carbon dioxide in the aqueous phase. It is not measured directly in groundwaters without a coexisting gas phase but can be calculated from the activities of aqueous H_2CO_3 and water. P_{CO2} has a direct effect upon the abundance of inorganic carbon in groundwaters and impacts upon the speciation and solubility of some actinides, and the stability of cementitious backfill materials.

Organic and inorganic ligands in the far-field groundwater can form strong complexes with dissolved cations, especially with trivalent radionuclides and, therefore, can have an influence on the apparent solubility and sorption of radionuclides. In saline waters, chloride complexes can be formed, e.g. with neptunium. Other important complexing agents are carbonate, phosphate and sulphate. Carbonate forms strong complexes with uranium, plutonium and americium, and sulphate may form complexes with plutonium. Dissolved organics can also affect redox and thus the concentrations of redox-sensitive elements and they can be decarboxylated into carbon dioxide and hydrocarbon gases. A range of natural organics, such as humic and fulvic acids, exist in groundwaters and are derived from degradation of biological material at the surface.

Colloids (particles with diameters less than 10 μm) have the potential to sorb radionuclides from aqueous solution and can be transported with groundwater flow. Colloids may be mineral particles, organic, or biological matter and may be hydrophobic (e.g. metal oxides) or hydrophilic (e.g. bacteria). As ionic strength increases, the tendency for colloids to flocculate (aggregate) increases.

Gas generation and transport, and microbial activity in the far-field rock are other processes that influence the groundwater chemistry.

The far-field groundwater will, after closure, resaturate the near-field (see GEN-32 “Hydraulic resaturation of the near-field rock”) and react with the engineered barrier system materials, thus becoming chemically distinct. The chemistry of the far-field groundwaters is, therefore, a primary control on the nature of the solid-groundwater reactions that may occur in the near-field.

These chemically distinct near-field groundwaters may later migrate back out to the far-field, where they can mix with and alter the far-field groundwater composition (see GEN-24 “Interfaces between different waters”). However, the large chemical buffering capacity of the far-field rock should ensure that the far-field groundwater chemistry is not grossly altered. The exception to this is in the case of cementitious repositories, such as the SFR, where a hyperalkaline contaminant plume could migrate and interact with the far-field rock and groundwaters (see SFR-23 “Groundwater chemistry in the near-field rock”).

The far-field groundwater can be perturbed by climate change which can cause infiltration of sea-water or glacial meltwaters (see GEN-21 “Glaciation”) or the uprising of deep shield brines (see GEN-6 “Groundwater salinity changes”).

Far-field groundwater interactions with the host rock, migrating near-field groundwaters and recharge waters, coupled with the complexity of the groundwater flow system, will ensure that the far-field groundwater chemistry is spatially and temporally heterogeneous. It is not always possible to know the extent of heterogeneity, so the least favourable (but reasonable) groundwater composition is often used as a conservative starting point in performance assessment calculations.

Thermodynamic models and coupled chemistry and transport models can be used partially to simulate the evolution of far-field groundwater chemistry. However, this approach is not perfect in that it does not take into account other controlling processes, such as the kinetics of reactions and mixing of different bodies of water. Furthermore, the thermodynamic databases frequently lack data for the solid solution mineral phases that are abundant in many potential host rocks.

Origin in the repository system:

The far-field groundwater chemistry is controlled by the mineralogical composition of the rock mass in contact with the groundwater. This, in turn, is controlled by the porosity and permeability structures of the rock mass, which affect the groundwater flow geometry and residence times. For groundwaters with a long residence time, rock-water interactions can generate a saline (brine) groundwater composition. Far-field groundwater chemistry is also controlled by the composition of infiltrating (recharge) waters, including marine (saline) waters at coastal and sub-seabed sites.

After closure, far-field groundwater chemistry will be further affected by mixing with chemically distinct waters flowing from the near-field that have interacted with the engineered barrier system materials.

Impact on the repository system:

The far-field groundwater composition will affect the water chemistry in the near-field rock and in the engineered barrier system. It thus controls the degradation processes affecting the buffer, the canister and the wasteform. Far-field groundwater composition is also the primary control on radionuclide solubility and speciation; sorption and desorption; and precipitation and dissolution in the far-field rock.

The far-field groundwater composition also affects alteration and weathering of the host rock in contact with the groundwater, microbial activity and the generation and stability of colloids in the water. Precipitation and dissolution reactions may affect the groundwater flow system.

Bibliographic references:

Arthur R and Apted M (1996) Modelling of near-field chemistry for SITE-94. SKI Report 96:31

Bottomley DJ, Gregoire DC and Raven KG (1994) Saline groundwaters and brines in the Canadian Shield: geochemical and isotopic evidence for a residual evaporite brine component. Geochimica et Cosmochimica Acta, 58, 1483-1498.

Glynn P and Voss C (1996) Geochemical characterisation of Simpevarp groundwaters near the Äspö Hard Rock Laboratory, SKI Report 96:29.

Lasaga AC (1984) Chemical kinetics of water-rock interactions. Journal of Geophysical Research, 89, 4009-4025.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.2.08 (Chemical/geochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 6.3 (Far-field hydrochemistry) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Dave Savage and Bill Miller

Number: **GEN-23**

Name: **GROUNDWATER FLOW**

Short description:

Refers to the magnitude, distribution and direction of the groundwater flow in the near-field and far-field rock. Groundwater flow may be altered locally around a repository both temporarily during the repository construction and operation stages, and subsequently due to changes in the engineered barriers or the near-field rock; in the far-field due to changes in the stress field and globally due to changes in the groundwater recharge and discharge. The groundwater flow may also be affected by gas generation and flow, and by density changes. Groundwater flow is of primary significance to the performance of the repository because it constitutes the major pathway for radionuclide migration back to the biosphere.

Technical description:

Most deep rocks are saturated with water and this water (groundwater) can flow through the rock carrying any dissolved (or suspended colloidal) substances with it. In the repository system, groundwater will migrate through the near-field, where it may dissolve radionuclides from the waste and transport them out to the far-field rock and, then potentially, up to the surface environment to groundwater discharge points, such as rivers and lakes (see GEN-16 “External flow boundary conditions”).

By this process, groundwater flow constitutes the primary pathway for the release of radionuclides from the repository. As a consequence, understanding the groundwater flow in and around a repository is a fundamental objective of performance assessment.

Natural groundwater flow occurs principally in response to differences in pressure and elevation (potential) within the groundwater system. This potential may be caused by a number of different mechanisms including gravity (topographic/elevation differences) and fluid density (groundwater chemistry and temperature differences). Any one (or combinations) of these mechanisms may produce a net driving force that causes the groundwater to move. However, it is differences in topographic elevation that provide the major driving force for most natural groundwater systems (see GEN-16 “External flow boundary conditions”).

The groundwater flow system can be affected by changes to recharge rates and locations. This can occur in response to climate change where the onset of permafrost and glacial conditions (see GEN-21 “Glaciation”) or sea-level changes (see GEN-33 “Sea level changes”) may radically alter recharge and the spatial extent of flow paths. The recharge rates and locations can also be affected by changes in land use.

The rate and direction of groundwater flow is also affected by certain physical properties of the rock through which it is flowing. The fundamental properties of importance are the porosity and the permeability (dictated by the nature of the connections between the porosity). Standard nomenclature refers to porosity as the total part of the rock that is void space, while effective porosity refers to that proportion of the total porosity which is interconnected and capable of allowing water to pass through. In practical terms, groundwater may flow either through the connected pore space within a porous media or through the open connected sections of fractures in a fractured system. The ability of a rock to allow groundwater to pass through it is referred to as the 'hydraulic conductivity' which can be inferred from field studies. In mathematical terms, hydraulic conductivity is the constant of proportionality relating groundwater flow rate to the hydraulic gradient in Darcy's Law.

The porosity and permeability of the rock mass through which the groundwater flows is not usually uniform and both may change depending on variations in the physical structure of the rock (e.g. lithological variations). This heterogeneity in the system means that the actual hydraulic conductivity of the rock and, hence, the velocity and direction of flow can vary widely throughout the rock mass.

This natural heterogeneity in the groundwater flow system is impossible to characterise in detail from field measurements. Therefore it is common to consider the direction and velocity of groundwater flow integrated over a volume of rock large enough to be considered to be a representative volume; hence a mean direction and velocity can be described. This known as the Darcy velocity. However, this approach cannot strictly be applied when considering the transport of radionuclides through the rock mass, because the heterogeneity, channelling and sorption properties will effect the migration of the contaminant plume within the representative volume.

In fractured crystalline rock, such as will form the host rock for repositories in Sweden, the groundwater flow will be concentrated in the network of interconnecting fractures. However, even within this system, there is a large range of conceptual uncertainty as to how flow actually occurs. Conceptual models range from the simplification of the fractures into a parallel plate model to a pipe network representing highly channelled flow within fractures.

One concept of channelling is that each fracture plane consists of open and closed parts. This concept might only be viewed as an extension of the discrete fracture approach, at least if the closed portion is not too large. There is little data on how the fracture transmissivity is distributed on the fracture plane. It is clear that the flow distribution among the different fractures will depend very much on the shape of the open parts. To complicate matters further this shape depends upon the rock stress field (see GEN-36 "Stress field"), the strength of the rock, and the asperity distribution and strength of the fractures.

There is a need to establish the geometry and statistics of such channels. Statistical analysis of fracture and hydraulic conductivity data, and resin imprints of natural fractures are possible tools to increase the understanding of the channelling effect. Detailed mapping of inflows on tunnel and shaft walls have also been suggested. However, it is questionable whether these observations are relevant for the bedrock beyond the disturbed rock zone, but simply show the perturbation of flow by the disturbed zone (see GEN-15 "Excavation effects on the near-field rock").

Another concept of channels is ‘extreme channelling’ where there are only a few paths where most of the water flows in the rock mass. These paths may be considered and modelled as physical conduits, in the rock mass. Other means of obtaining extreme channelling are through a poorly percolating fracture network model or a poorly percolating stochastic continuum model and a given set of boundary conditions. Geostatistically, channelling will emerge in a model when the correlation scale of the hydraulic conductivity is equal to the flow domain and its variance is high. A poorly percolating fracture network could be the result of a relatively low fracture density.

An important consideration when modelling groundwater flow is therefore the spatial scale of the problem under consideration. For large scale (regional or area), it may be adequate to model the groundwater flow system using assumptions of a porous media and Darcy velocities. However, on the smaller scale, such as portions of a host rock around a repository, it may be necessary to adopt a more detailed discrete fracture or channel network model.

The groundwater flow field around a repository will be different to that which occurred naturally in the virgin rock before repository construction began. The construction work creates an engineered damaged zone around the repository which will have a larger bulk hydraulic conductivity than the intact rock (see GEN-15 “Excavation effects on the near-field rock”). Consequently, groundwater flow in the region of the repository may tend to be concentrated in this engineered damage zone. The subsequent re-saturation after the emplacement of waste will also impact the groundwater flow over a time period of decades (see GEN-32 “Hydraulic resaturation of the near-field rock”). Over longer time periods, groundwater flow in the vicinity of the repository may be further altered by degradation of the barriers and alteration of the near-field rock.

Furthermore, radiogenic heat output from near-field (most significant in the case of spent fuel repository) may affect groundwater flow paths by initiating thermally driven convection currents around the repository. However, the groundwater flow also affects the temperature field because the flowing water will transport the heat through advection. Heat is also transported through conduction in both the water and the solid phase. In very low permeability media, heat conduction is the dominant heat transport mechanism. In general, the temperature effects on groundwater flow are relatively well understood. However, special attention to the problem may be required in relation to coupled thermo-hydro-mechanical effects.

The description of the groundwater flow field in the far-field after closure of the repository is the first stage in the understanding of the radionuclide transport. However, in performance assessment it is not only the velocity and direction of groundwater flow that is important for understanding radionuclide transport. Radionuclides transported by groundwater flow concentrated in one or two fast flowing channels will have less opportunity to interact with and sorb on the rock surfaces (see GEN-34 “Radionuclide sorption”) than similar concentrations of radionuclides carried in slower moving water dispersed throughout a larger fracture network or through the porous rock.

Additional information on the interaction of radionuclides and the rock mass for processes such as sorption, diffusion and dispersion are required for a complete description of the transport of contaminants.

Origin in the repository system:

The groundwater flow system around a repository on the large-scale is primarily controlled by the topography of the land surface which induces gravity-driven pressure (head) gradients that causes groundwater to flow. Other driving forces can relate to salinity and temperature variations.

On the local scale, groundwater flow velocities and directions are controlled by heterogeneities in the physical structure of the rock, such as fractures.

Impact on the repository system:

Groundwater flow is the primary mechanism for the release and transport of radionuclides through the far-field to the biosphere. It is, therefore, of critical importance in performance assessment. The nature of groundwater flow affects the geochemical conditions at the repository and, hence, the retardation potential for radionuclides in solution or associated with suspended colloids and the degradation of the engineered barriers. The distribution of the groundwater flow affects radionuclide retardation by determining the amount of rock surfaces accessible to matrix diffusion and sorption.

Groundwater flow also affects the duration of the resaturation phase, the rate of water turnover in the near-field and, hence, the near-field groundwater composition. Furthermore, groundwater flow affects the rock stresses, the temperature and gas flow and transport in the rock. The groundwater flow in the near-field rock will affect the resaturation of the bentonite buffer and the tunnel backfill and the water flow in them, and may cause erosion of these materials.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Spitz K and Moreno J (1996) A practical guide to groundwater and solute transport modelling. Wiley InterScience.

Equivalent FEPs:

Relates to FEP 2.2.07 (Hydraulic/hydrogeological processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEPs 4.2.3 (Extreme channel flow of oxidants and nuclides), 4.2.4 (Thermal buoyancy) and 4.2.5 (Changes of groundwater flow) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-24**

Name: **INTERFACES BETWEEN DIFFERENT WATERS**

Short description:

There is potential for the development of interfaces between groundwaters of different composition in the near and far-field rock. At these interfaces changes may occur in radionuclide solubilities and groundwater flow which could affect radionuclide transport and release.

Technical description:

The far-field groundwater chemistry is initially controlled by rock-water interactions between infiltrating groundwaters and the minerals in contact with them, and by mixing between waters of different origin (see GEN-22 “Far-field groundwater chemistry”). Because of the nature of these interactions, the groundwaters will inevitably exhibit some degree of spatial variability in their chemistry.

In general, in a crystalline or argillaceous host rock, the rock-water interactions cause waters to become chemically reducing, mildly alkaline and more saline with depth and time since infiltration. For deep groundwaters with a long residence time, rock-water interactions can generate a saline (brine) groundwater composition (see GEN-6 “Groundwater salinity changes”). In much of the bedrock of Sweden, saline water exists at depth, with an interface or mixing zone between these waters and the shallower, less saline waters. The exact composition of deep groundwaters is strongly site specific, dependent on the mineralogical characteristics of the rock mass, the nature of the groundwater flow system and the composition of the recharge waters. The latter includes marine waters at coastal and sub-seabed sites.

Many of the changes in chemistry will be gradual but others may quite sharp. Sharp chemical interfaces are likely to result at the contact zone between waters of different origin or due to a mineral dissolution or reaction front. Even when significant mixing does occur, certain chemical parameters may still present a sharp chemical interface. For example, in a mixing zone between a deep reducing water and an infiltrating oxidising water, although many of the chemical constituents will show a gradual change, the dissolved oxygen may show a sharp interface located within the zone of mixing, which is controlled by the availability of reducing minerals such as pyrite.

Mixing may be inhibited by a range of processes, such as density differences in the waters. An example of this case would be the very distinct saline water interface that is likely to occur in coastal regions where marine and meteoric recharge waters meet.

Mixing will also be limited between waters in fast advecting fractures and slow moving (stagnant) waters in the voids and fractures in the rock mass. This case would be typical of fractured crystalline rock in which the majority of groundwater flow is localised in a small number of hydraulically-active fractures. Such a case was found in the vicinity of the Äspö hard rock laboratory (Smellie and Laaksoharju, 1992). Waters of different composition could also develop as a result of external perturbations such as glaciation causing sea-level change (see GEN-33 “Sea level changes”) or enhanced freshwater recharge.

Finally an interface between different waters will occur when contaminated waters leaving the repository migrate out to the far-field. This will particularly be the case for the SFR repository where a chemically distinct alkaline plume, derived from cement-water interactions, will migrate from the repository (see SFR-23 “Groundwater chemistry in the near-field rock”). The spent fuel repository will also create a plume of chemically altered water, although the chemical contrast between this plume and the far-field groundwaters will be less than at the SFR repository.

Whenever an interface or mixing zone exists between waters of different chemistry, there is likely to be a change in radionuclide solubilities on either side. Consequently, radionuclide mobility either side of an interface or mixing zone could vary.

If the interface itself is moving (such as at the alkaline plume migrating away from the SFR repository), then the behaviour of radionuclides caught in the path of the interface will be significantly affected. For example, this could lead to a gross increase or decrease in solubility. The interfaces between waters is likely to be a site for preferential colloid formation.

The formation of sharp interfaces between groundwaters should also be considered in terms of potential effects on groundwater flow (Voss and Andersson, 1993; Provost et al., 1996). For example, saline water intrusion implies changing density gradients and changing groundwater flow geometry: the location of discharge areas may alter as a consequence (see GEN-23 “Groundwater flow”).

Origin in the repository system:

Sharp interfaces between groundwaters with different chemistries will occur whenever waters with variable origins show limited mixing. Different origins could be due to differences in rock-water interaction rates and processes; different recharge chemistries and locations; or external perturbations (e.g. due to glaciation).

There are likely to be interfaces in fractured crystalline bedrocks between waters in fast flowing fractures (short water-rock reaction times) and waters in the bulk of the rock matrix (long water-rock reaction times).

Other interfaces could occur as a result of the migration of contaminated waters (e.g. alkaline waters) from the repository moving into the far-field.

Impact on the repository system:

Waters with different chemistries will affect radionuclide solubilities and thus radionuclide transport and release. The interfaces between waters is likely to be a site for preferential colloid formation.

The interface between waters with different densities may also have an impact on groundwater flow systems.

Bibliographic references:

Provost A, Voss C and Neuzil C (1996) Glaciation and regional groundwater flow in the Fennoscandian shield. SKI Technical Report 96:11.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Report 94:30.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Smellie JAT and Laaksoharju M (1992) The Äspö Hard Rock Laboratory: final evaluation of the hydrogeochemical pre-investigations in relation to existing geologic and hydraulic conditions. SKB Technical Report, TR 92-31.

Voss C and Andersson J (1993) Regional flow in the Baltic shield during Holocene coastal regression. Groundwater, 31, 989-1006.

Equivalent FEPs:

Relates to FEPs 2.2.07 (Hydraulic/hydrogeological processes and conditions in geosphere) and 2.2.08 (Chemical/geochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 5.1 (Saline (or fresh) groundwater intrusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-25**

Name: **MATRIX DIFFUSION**

Short description:

Matrix diffusion is the process by which radionuclides and other species in the water flowing along fractures migrate into the non-flowing microfractures and into the microporosity of the surrounding rock mass. Matrix diffusion can provide an efficient retardation mechanism for both sorbing and non-sorbing contaminants.

Technical description:

Matrix diffusion is the diffusion of radionuclides out of the principal flow paths into the stagnant (non-advecting) microfracture network and into the microporosity of the rock mass (Neretnieks, 1980). Matrix diffusion occurs in both porous media and in fractured rock. However, the impact on performance assessment in porous media may be small whereas, in fractured rock, it may be very significant. Matrix diffusion may occur in the far-field rock of both the spent-fuel and SFR repositories.

In a fractured rock, the diffusion of contaminants (see GEN-9 “Diffusion”) from advecting fractures into the microfracture network and into the microporosity of the rock mass will be controlled by a number of factors such as the flow rate in the advecting fracture, the fracture aperture, the flow wetted surface and the nature of the fracture surface and of any fracture coating mineralogy that may exist. Anion exclusion effects may limit the role of matrix diffusion for negatively charged species (see GEN-2 “Anion exclusion”) by retarding their migration into the rock matrix . Temperature could theoretically act as a secondary control on matrix diffusion rates.

The impact of matrix diffusion on radionuclide transport in fractured crystalline rock can be highly significant. Firstly, the migration into (and subsequently out of) the stagnant water of the rock matrix may cause temporal dilution of all contaminants, including non-sorbing conservative species. Secondly, matrix diffusion substantially increases the available rock surface area for sorption, thus further retarding the sorbing radionuclides (see GEN-34 “Radionuclide sorption”).

The actual significance of matrix diffusion on repository releases is controlled by the volume of rock available for the process. In turn, this volume is restricted by the extent of the flow wetted surface in the advecting fractures and the depth of interconnected microporosity in the rock mass. The flow wetted surface is partly controlled by any alteration of the fracture surface. For example, a thick layer of secondary low permeability clay minerals on the fracture surface could effectively ‘seal’ the rock matrix from the groundwater and would limit matrix diffusion.

While it is generally accepted that some matrix diffusion effects will occur in all fractured rocks, it is very difficult to quantify it because the flow wetted surface and the depth of interconnected porosity are hard to measure and are site (rock) specific. Field studies indicate that the volume of rock available for matrix diffusion is actually only a small proportion of the total volume and possibly restricted to a volume of rock just a few centimetres thick adjacent to the advecting fractures (e.g. Alexander et al., 1988). Furthermore, in field studies, it is sometimes difficult to separate the effects of matrix diffusion from other retardation processes (e.g. precipitation or sorption).

Recently Ohlsson and Neretnieks (1995) compiled available data on matrix diffusion from crystalline rocks. They found large ranges of measured values of the diffusion coefficient for the same nuclide which they attributed to the natural variations in the characteristics of the rock samples. Measurements using different sorbing nuclides in the same rock have been undertaken only infrequently.

Due to the uncertainty in quantifying matrix diffusion, some performance assessments have not taken account of it in transport calculations, which is a conservative assumption. However, to actually include it in the transport model is not very difficult, if the flow wetted surface and depth of interconnected porosity could be quantified.

Origin in the repository system:

Matrix diffusion occurs as a result of the formation of a network of microfractures in the rock mass adjacent to advecting fractures or the existence of a connected network of micropores in the rock, at the boundary between minerals. The volume of rock available for matrix diffusion is controlled by the distribution of migrating contaminants in the fracture network and the depth of the interconnected porosity adjacent to the advecting fractures.

Impact on the repository system:

Matrix diffusion has a significant impact on the transport and retardation of radionuclides released to the far-field rock. By increasing the surface area available for rock-water interaction, matrix diffusion may increase the potential for sorption.

Excluding matrix diffusion from radionuclide transport calculations is a conservative assumption.

Bibliographic references:

Alexander WR, Scott RD, MacKenzie AB and McKinley IG (1988) A natural analogue study of radionuclide migration in a water conducting fracture in crystalline rock. Radiochimica Acta, 44/45, 283-289.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Neretnieks I (1980) Diffusion in the rock matrix: an important factor in radionuclide migration? Journal of Geophysical Research, 85, 4379-4397.

Ohlsson Y and Neretnieks I (1995) Literature survey of matrix diffusion theory and of experiments and data including natural analogues. SKB Technical Report, TR 95-12.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Related to FEP 4.1.5 (Matrix diffusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **GEN-26**

Name: **PERMAFROST**

Short description:

Permafrost is ground that remains frozen throughout the year and is associated with cold climates. If permafrost formed in the vicinity of a repository, it would have significant impacts on the groundwater flow system and, hence, on the migration of radionuclides through the far-field. Some direct physical damage to borehole and shaft seals could occur.

Technical description:

Permafrost is defined as ground, soil or rock, with temperatures which remain continuously below 0°C for two or more years; that is, surface materials where the porewaters are frozen. Permafrost development in the vicinity of a repository will affect groundwater flow and composition, and can also affect the integrity of borehole and shafts seals.

Permafrost occurs in regions where the mean annual air temperatures are below 0°C. Due to climate change, the regions experiencing permafrost can vary with time. It is probable that large parts of Sweden will experience glacial conditions within the next 100 000 years and that these glacial conditions will be accompanied by the formation of permafrost (see GEN-21 “Glaciation”). Hence permafrost development is a significant process for the spent fuel repository because this time is within the period of interest for performance assessments for this repository. In the case of the SFR, this time is beyond the period of interest for quantitative performance assessments and, consequently, permafrost development is not considered significant for the SFR.

There is abundant evidence to suggest that Sweden has gone through several cycles of extensive permafrost development during the Quaternary Period (the last 2 million years). Permafrost exists today in the far north and in the high mountains of Sweden, and more extensively in other parts of the world: for example, in the Spitzbergen area, the permafrost depth is approximately 450 m and, in Siberia, depths exceeding 1500 m have been reported.

Where permafrost occurs, it is generally overlain by an active layer, which is subject to seasonal freezing and thawing. The occurrence of permafrost is controlled by the surface heat balance which, in turn, is influenced by the properties of the ground surface, such as relief, slope aspects, vegetation, snow cover, moisture content, soil and rock type and the presence of surface water bodies, and also by the geothermal flux. The geothermal flux is an important control on the maximum depth of permafrost (see GEN-38 “Temperature of the far-field”).

Kersten (1959) has derived an empirical formula relating the depth of frost penetration to the number of days per year with air temperatures near or below 0°C. It has also been found that the limit of permafrost shows a strong correlation with the mean annual air temperature isotherm of -1 to -2°C.

Locally, moving groundwater can have a significant influence on permafrost development. Near the margins of a permafrost area, the permafrost consists of thin, isolated masses, and is more likely to be present under north facing slopes with thin winter snow cover. Towards the north, the areas in which permafrost occurs gradually become larger, and the depth to the base of the permafrost can increase to depths in excess of 500 m at high latitudes, in particular in areas which do not have a thick ice cover which acts as a thermal insulator.

Permafrost can have a major influence on the recharge and discharge of groundwater and, in the repository far-field, could be significant in localising the return of radionuclides to the surface environment. It can produce changes in groundwater chemistry by increasing rock-water interaction times and by limiting freshwater recharge (see GEN-22 “Far-field groundwater chemistry”). This can result in the formation of saline groundwaters, especially at the base of the permafrost zone, and this could be significant in radionuclide migration.

According to climate change predictions, thick permafrost is not expected to form during the main thermal phase of the repository and, therefore, this potential interaction is not significant.

If, instead, permafrost occurs after radionuclides have migrated into the far-field, then this may result in a concentration of radionuclides in the groundwater beneath, or within, the permafrost zone. When the permafrost subsequently melts, these radionuclides may be released as a concentrated pulse (see GEN-31 “Radionuclide reconcentration”).

Gases derived from deeper within the rock mass may become frozen as clathrates within the permafrost zone or may be trapped as a gaseous phase beneath the permafrost. Gas accumulation could lead to enforced outflow of groundwater from the repository, though this is thought to be of minor significance. The influence of any gas cushion on the groundwater flow field is considered possibly more important. Clathrates are not normally found in crystalline rocks and their significance is uncertain in the context of a repository for spent fuel.

In addition, depending on the depth of the permafrost, parts of the repository structure itself could be affected. Calculations (McEwen and de Marsily, 1991) demonstrate that if the glacial period is of sufficient length, permafrost could reach to depths of up to several hundred metres (although 200 m is a likely maximum depth) and this could be significant in increasing the potential for shaft seal failure (see GEN-7 “Degradation of the borehole and shaft seals”).

Origin in the repository system:

Permafrost is one consequence of natural, long-term climate changes that are, themselves, caused by variations in the solar radiation reaching the Earth’s surface. As such, permafrost is caused by processes external to the repository.

The depth and extent of permafrost development depend not only on the mean annual air temperature but also the characteristics of the surface materials and the local geothermal gradient.

Impact on the repository system:

Permafrost development can have significant impacts on the repository system. The initial effect is to modify the groundwater flow system by localising the recharge and discharge zones. A potential consequence will be to increase the rock-water interaction times and limit fresh water recharge. Thus groundwaters beneath permafrost may become saline and, in a repository environment, may contain increased concentrations of trapped radionuclides.

Permafrost may cause physical damage to shaft and borehole seals, although it is less likely that permafrost would extend deep enough directly to impact on the repository itself.

Bibliographic references:

Brown J, Ferrians JA, Heginbottom E and Melnkow ES (1998) Digital circumarctic map of permafrost and ground-ice conditions. Circumpolar Active Layer Permafrost System (CAPS) CD-ROM v1.0. International Permafrost Association, Data and Information Working Group. National Snow and Ice Data Centre, University of Colorado.

Kersten MS (1959) Frost penetration: relationship to air temperature and other factors. US Highway Research Board Bulletin, 225, 45-80.

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

McEwen TJ and de Marsily GH (1991) The potential significance of permafrost to the behaviour of a deep radioactive waste repository. SKI Technical Report, 91:8.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 1.3.04 (Periglacial effects) in the NEA International Database (NEA, 1998).

Relates to FEP 5.17 (Permafrost) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-27**

Name: **RADIONUCLIDE PRECIPITATION AND DISSOLUTION**

Short description:

Radionuclides dissolved from the waste may be precipitated in the near or far-fields if their solubility is exceeded due to changes in chemical conditions or temperature. The precipitated solids may be re-mobilised as a result of further changes in chemical and physical conditions. Precipitation and dissolution processes are thus important for understanding the transport behaviour of radionuclides.

Technical description:

Radionuclides released from the wasteform may be dissolved in groundwaters in the near and far-fields. The solubility of these radionuclides is controlled by standard thermodynamic laws and is largely a function of Eh, pH, concentrations of complexing ligands and, to a lesser extent in the repository environment, temperature and salinity (see GEN-22 “Far-field groundwater chemistry”).

Radionuclides in solution will migrate with the groundwater out of the engineered barriers, through the near-field rock, to the far-field. Along the flow path, the physico-chemical conditions will change and, as a result, the solubility of particular chemical species may be reduced. If this occurs, and the solubility reduces to the point that the concentration of a given chemical species in the groundwater exceeds solubility limits, then that chemical species will precipitate from solution. For example, a change to a more reducing environment will cause precipitation of uranium and other redox sensitive elements which have a lower solubility under reducing conditions.

If the physico-chemical conditions change again such that the solubility of a particular species increases, then previously precipitated radionuclides could dissolve. This could occur, for example, if the redox conditions shift from reducing to oxidising as a result of the movement of a redox front associated with radiolysis (see GEN-30 “Radiolysis”).

In the spent fuel repository, redox conditions are particularly important for controlling solubility because radiolysis may create an oxidising near-field and a mobile redox front. The position and potential movement of the redox front away from the waste will control radionuclide precipitation and dissolution behaviour (see SFL-38 “Redox fronts”).

In the SFR repository, it is likely that pH conditions will be the dominant control on radionuclide solubility, with the pH conditions being buffered by the large volumes of cementitious material used in the engineered barriers (see SFR-23 “Groundwater chemistry in the near-field rock”).

Other factors that may cause precipitation and dissolution away from the waste are changes in temperature (due to radiogenic heating, for example) or in the concentration of ions and complexes which affect the speciation of the nuclides.

Radionuclides initially precipitated as microcrystalline or amorphous solid phases may recrystallise as coarser crystals, a process which is thermodynamically favourable, and leads to lower solubility of the radionuclides. Radionuclides present in trace quantities in the waste will probably be incorporated (coprecipitated) as trace components in solid solution within alteration phases, such as aluminosilicates, oxides, carbonates, sulphates, phosphates, and sulphides. In addition, radionuclides may be coprecipitated with some carrier compound, e.g. hydrous oxides of iron, and may later be either released into solution, or enter into a solid solution if the carrier substance recrystallises to another with a higher degree of crystallinity.

In performance assessment, it is essential to be able to predict radionuclide solubilities under near and far-field conditions. Generally, thermodynamic codes and their associated databases can be used to calculate the elemental solubilities and speciation, although there is not always consensus on which solubility data to use (e.g. McKinley and Savage, 1996).

A number of ‘blind predictions’ have been made whereby performance assessment codes have been used to attempt to replicate the trace element concentrations measured in natural waters (e.g. Bruno et al., 1990). These tests have highlighted the limitations in the codes and databases. In particular, these codes are unable to take into account processes such as co-precipitation and co-dissolution.

Consequently, there is uncertainty over the ability of performance assessment always to predict accurately the solubilities of certain elements. As a result, performance assessments sometimes tend to estimate conservative solubilities (rather than predicted solubilities) for some elements to avoid under-predicting actual solubilities and, thus, transport rates, for example by assessing formation of amorphous phases.

Origin in the repository system:

Precipitation and dissolution of elements released from the waste is influenced by changes in pH, redox conditions, temperature, or concentration of ions and complexes in the water which influence the speciation of the elements.

Impact on the repository system:

Precipitation and dissolution of elements released from the waste affects the transport and release of radionuclides away from the waste package and out into the near and far-field rock. In addition it may affect the water chemistry, and radiation from precipitated radionuclides might affect water (radiolysis) and barrier materials.

Bibliographic references:

Arthur R and Apted M (1996a) Radionuclide solubilities for SITE-94. SKI Report 96:30.

Arthur R and Apted M (1996b) Modelling of near-field chemistry for SITE-94. SKI Report 96:31.

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Grenthe I and Puigdomenich I (1997, editors) Modelling of aquatic chemistry. NEA/OECD.

McKinley IG and Savage D (1996) Comparison of solubility databases used for HLW performance assessment. Journal of Contaminant Hydrology, 21, 335-350.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.7 (Recrystallisation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-28**

Name: **RADIOACTIVE DECAY**

Short description:

Radioactive decay is a fundamental process that affects all radioactive (unstable) nuclides. Radioactive decay will change the inventory of radionuclides in the waste, and the heat generation will affect the temperature in the near-field and the stability of the wasteform and other materials.

Technical description:

Radioactive decay is a fundamental process that affects all radioactive (unstable) nuclides. During radioactive decay, the nuclei of unstable atoms undergo spontaneous transformations that involve the emission of particles and of radiant energy.

The radiation emitted due to radioactive decay (alpha, beta and gamma) is the primary reason why such care has to be taken for the disposal of nuclear wastes.

The rate at which radioactive decay occurs is nuclide specific, and is defined by the half-life for the particular nuclide. This half-life is independent of all other physico-chemical parameters (such as redox, temperature etc.) in the repository.

Radioactive decay will occur in all waste types in both the spent fuel and SFR repositories. Furthermore, radioactive decay will affect the nuclides regardless of their location in the repository or their chemical and physical form. Thus, radioactive decay will affect mobile soluble nuclides as well as nuclides included in solid phases or sorbed onto solid phases.

Since the decay rate of the nuclides in the waste is well known, the changing inventory of the repository can be accurately predicted over time. Thus radioactive decay is a fundamental consideration in performance assessment models.

The type of radiation generated by the decay depends on the radionuclide. The emitted radiation can cause radiolysis of water and gas around the waste package (see GEN-30 “Radiolysis”) and may also affect the stability of the wasteform, the engineered barriers and thus their degradation processes and rates. The extent of physical damage is partially controlled by the penetration depth of the radiation in the material of interest.

This is particularly an issue for spent fuel, where the spent fuel dissolution rate (see SFL-21 “Spent fuel dissolution and conversion”) is affected by the generation of reactants created by radiolysis of the water (e.g. Werme et al., 1990; Sunder et al., 1992). Molecular oxidants, radical species, H_2 and H^+ generated by radiolysis will affect the dissolution rate of the fuel and the subsequent transport of dissolved radionuclides. Because of the short-range penetration of a radiation, a radiolysis only affects the thin water film close to the UO_2 surface (Werme et al., 1990). Oxidants produced in this layer migrate to the UO_2 surface, resulting in an oxidative dissolution of the UO_2 and potentially the formation of new oxidised uranium compounds. In the bounding case, the oxidative dissolution rate of UO_2 may be assumed to be linearly related to the a dose rate (SKB, 1992).

Besides emitting radiation, radioactive decay also generates heat which will cause the near-field temperature of the repository to rise after closure. Because the spent fuel repository has a larger radioactive inventory than the SFR repository, radiogenic heat production is more critical to understand for the spent fuel repository.

Origin in the repository system:

Radioactive decay is a fundamental process whose rate is independent of other physico-chemical parameters in the repository.

Impact on the repository system:

Radioactive decay will change the inventory of radionuclides in the waste, and the heat generation will affect the temperature in the near-field and, thus, radionuclide solubilities and the degradation of engineered barriers.

Bibliographic references:

Benedict M, Pigford TH and Levi HW (1981) Nuclear Chemical Engineering. McGraw-Hill, New York.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Sunder S, Shoesmith DW, Christenson H and Miller NH (1992) Oxidation of UO_2 fuel by the products of gamma radiolysis of water. Journal of Nuclear Materials, 190, 78-86.

Werme L, Sellin P and Forsyth R (1990) Radiolytically induced oxidative dissolution of spent nuclear fuel. SKB Technical Report, TR 90-08.

Equivalent FEPs:

Relates to FEP 3.1.01 (Radioactive decay and in-growth) in the NEA International Database (NEA, 1998).

Relates to FEP 1.1.2 (Radioactive decay, heat) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-29**

Name: **EROSION AND WEATHERING**

Short description:

Erosion and weathering are processes which lead to the denudation of the land surface and a reduction in topography. In Sweden, the most significant erosion events are associated with glaciation and these potentially may be of significance to repository safety. Erosion and weathering may cause modifications to the groundwater flow system.

Technical description:

Weathering refers to the denudation of the surface rock due to the combined action of physical degradation and chemical dissolution processes acting on the rock and its component minerals. The solid products of weathering create the sediments and soils which overlie the bedrock.

Erosion is the process whereby soils and sediments are transported away from their place of formation. The main agents of erosion are water, wind, mass movement and ice. Soil particles are loosened from the parent mass by mechanical and chemical processes. Thus, erosion removes the products of weathering.

The combined actions of weathering and erosion denude the land and, over long periods of time, will remove the rock forming the far-field barrier for a repository. Erosion and weathering will occur at the sites of both the spent fuel and SFR repositories.

Weathering and erosion by wind and water are very slow processes and will have no impact on either repository during the time periods of interest to performance assessment.

The most rapid denudation rates are associated with glaciation and, thus, future glacial events potentially may have significance for repository safety (see GEN-21 “Glaciation”). Full glaciation in Sweden is anticipated 60 000 in the future (e.g. King-Clayton et al., 1995). Hence glacial erosion may be a relevant process for the spent fuel repository because this time is within the period of interest for performance assessments for this repository. In the case of the SFR, this time is beyond the period of interest for performance assessments and, consequently, glacial erosion is not considered significant for the SFR.

Sweden has experienced repeated glacial events in the geological past and calculations indicate that, in flat terrain areas that characterises most of Sweden, the cumulative erosion of the crystalline basement caused by these glaciations has been limited to only a few tens of meters. The rates of glacial erosion in the Scandinavian mountain ranges would be higher than the rates in the flat terrain areas. However, during the last glaciation in Sweden, erosion at the centre of the ice sheet in the flat areas was minimal in extent and depth.

However, field evidence indicates that localised erosion processes that occurred at the fringes of the last ice sheet may have been more extensive. Glaciofluvial canyons in Sweden have been described by Olvmo (1989) who interprets them as having been formed by glaciofluvial streams flowing subaerially, along the margins of an ice sheet, or by supraglacial streams superimposed on the ground.

This field evidence suggests that, if the margin of a future ice sheet occurred in the vicinity of the repository for extended periods of time, there may be a risk that extensive erosion could occur. Even so, a glacial canyon 100 m deep forming near the repository would not directly impact on the repository but it might have an indirect effect through changes to the stress field, the groundwater flow patterns and the chemical distribution of water around the repository.

Erosion (both glacial and non-glacial) can, however, affect groundwater recharge and discharge and thus may impact on groundwater flow and radionuclide transport. Nonetheless, the impact on the groundwater flow is minor relative to the general uncertainty in the distribution of the groundwater recharge.

Overall, erosion and weathering is not expected to present a significant risk to repository safety, although a few uncertainties remain with regards to localised processes at the fringes of ice sheets that may, under certain circumstances, cause deep excavations into the rock.

Origin in the repository system:

Erosion and weathering are caused by forces external to the repository system. Climate forces (causing glaciation) are the most significant.

Impact on the repository system:

The impact of erosion and weathering on the repository at depth will be minimal.

At the surface, erosion and weathering will cause changes to the recharge and discharge zones for the groundwater flow system and the characteristics of the biosphere. The most rapid erosion rates will be due processes that occur at the fringes of ice sheets.

Bibliographic references:

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Olvmo, M (1989) Meltwater Canyons in Sweden: A study of the canyons of the kursu-, skura- and grav-type. GUMI Rapport 27. University of Göteborg.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

SKI (1994) Evaluation of SKB's In-depth Safety Assessment of SFR-1. SKI Report, 94:30.

Equivalent FEPs:

Relates to FEP 2.3 (Surface environment) in the NEA International Database (NEA, 1998).

Relates to FEP 5.26 (Erosion of surface/sediments) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **GEN-30**

Name: **RADIOLYSIS**

Short description:

Radiation emitted during radioactive decay of unstable nuclides can cause radiolysis of the groundwater and of water-bearing solid materials. This radiolysis can lead to the formation of oxidants and free hydrogen gas which will impact on the redox conditions in the near-field, leading to a change in radionuclide solubilities.

Technical description:

Radiation emitted during radioactive decay of unstable nuclides (see GEN-28 “Radioactive decay”) will cause radiolysis if it impinges on water molecules in the groundwater or structurally bound in solid phases, such as organic materials.

All forms of radiation (alpha, beta and gamma) can cause radiolysis but it is the alpha particles that are the most efficient. Radiolytic decomposition of water molecules leads to the formation of numerous ionised species. Although equal amounts of oxidants and reductants are formed, most of these species are extremely reactive or unstable, and quickly convert to more stable or inert species.

In pure water, radiolysis leads to the formation of molecular hydrogen (gas) and oxidants such as hydrogen peroxide (H_2O_2) and molecular oxygen. The hydrogen gas is mobile and relatively unreactive under near-field conditions so will tend to diffuse out of the system. The net effect of the radiolysis of groundwater is, thus, to create a build-up of oxidants which will drive the redox conditions towards more oxidising conditions.

The actual redox conditions established in the near-field will be controlled by many factors but one of the most important is the availability of Fe^{2+} in the engineered barrier materials and in the rock to buffer the redox conditions and limit the development of oxidising conditions. Radiolysis reactions in repository groundwaters will be further complicated by the presence of dissolved ‘scavenger’ ions, as well as the presence of reactive surfaces on the engineered barrier materials.

If insufficient Fe^{2+} is present in the near-field to buffer the redox, then progressively more oxidising conditions can be established which will then slowly migrate outwards as a mobile redox front. Under oxidising conditions, the solubility of many radionuclides will increase. Consequently, the introduction of radiolytically induced oxidising conditions may increase the radionuclide release rate from the near-field.

In addition, the onset of oxidising conditions will increase the corrosion rate of the wasteform and the engineered barrier materials. This is particular an issue for spent fuel where, if it is assumed that all the oxidants react with the fuel causing dissolution, the dissolution rate occurs in proportion to the a decay rate (see SFL-21 “Spent fuel dissolution and conversion”). This, however, is unlikely since the majority of the oxidants are likely to recombine with radiolytic reductants or with more reactive Fe^{2+} materials, as indicated in the Cigar Lake analogue studies (SKI, 1996).

Radiolysis can occur in both the spent fuel and SFR repositories. In the case of the spent fuel repository, radiolysis will be limited to the less intense and less efficient gamma radiolysis during the period the canister remains intact because the alpha and beta radiations are unable to penetrate through the thickness of the metal canister wall. Once the canister is breached, however, a radiolysis can begin once water is in contact with the spent fuel. Christensen and Bjergbakke (1982) calculate that 1 mol H_2 per m^2 of fuel surface will be generated after 1 million years. The contact area between fuel and water determines the amount of water being radiolytically decomposed. Once radionuclides have migrated from the canister, radiolysis can occur due to the decay of mobile nuclides (e.g. on colloids or dissolved in the water) or nuclides sorbed or precipitated on to mineral surfaces in the near or far-field rock.

For the spent fuel repository, however, the most important potential impacts of radiolysis is the creation of an oxidising near-field which would increase radionuclide solubilities and increased spent fuel dissolution rates (see SFL-38 “Redox fronts”). Performance assessment calculations, suggest that this is not likely to be a problem because sufficient amounts of elemental and ferrous iron will be present to react with the oxidants.

In the SFR repository, a radiolysis will be able to begin soon after closure because the waste packages are not designed to isolate the waste for extended periods of time. However, the activity of the waste in the SFR is considerably less than in the spent fuel and, as a result, the potential radiolytic yield is less. In the SFR radiolysis of organic materials (e.g. cellulose and bitumen) will occur in addition to radiolysis of the groundwater. The significance of radiolysis of organic material is related more to a potential increase in material degradation rates than to a control over redox conditions.

For the SFR, the very large volumes of steel in the engineered barriers will mean that it is unlikely that the redox conditions will be significantly affected by radiolysis. Consequently, radiolysis is not considered to be a serious problem for SFR repository safety.

The radiolytically-produced hydrogen gas may also have an impact on the repository behaviour if gas generation rates are sufficient to create a discrete gas phase. In the SFR much larger volumes of gas will be generated from anaerobic corrosion of steel and, consequently, the smaller volumes of gas produced from radiolysis are only of secondary importance (see SFR-11 “Gas generation in the repository”). In the spent fuel repository, gas may be important because it has the potential to disrupt the bentonite buffer around the canisters. However, it is unlikely that radiolysis alone would be able to create a large enough gas pressure to cause mechanical disruption to the buffer (see SFL-24 “Gas flow through the buffer and backfill”).

Origin in the repository system:

Radiolysis will occur whenever radiation impinges on water molecules. The rate of radiolytic decomposition of water will thus be largely controlled by the inventory (radioactivity of the waste) and the longevity of waste packages which isolates the waste from the water. The impact of radiolysis on near-field chemistry will be controlled by the groundwater composition and particularly on the availability of Fe^{2+} to react with the radiolytically produced oxidants.

Impact on the repository system:

Radiolysis will affect the redox conditions in the near-field, particularly if insufficient volumes of Fe^{2+} are present to react with the radiolytically produced oxidants. The potential impact will be to create an oxidising near-field which will increase radionuclide solubilities.

Hydrogen gas will also be created by radiolysis and this gas potentially may affect repository behaviour. However, other gas production mechanisms will dominate gas generation, particularly the anaerobic degradation of iron in the engineered barriers.

Bibliographic references:

Christensen H and Bjergbakke E. (1982) Radiolysis of groundwater from spent fuel. SKB Technical Report, TR 82-18.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Neretnieks I and Faghihi M (1991) Some mechanisms which may reduce radiolysis. SKB Technical Report, TR 91-46.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.1.13 (Radiation effects in the wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.1 (Radiolysis) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-31**

Name: **RADIONUCLIDE RECONCENTRATION**

Short description:

Radionuclides may be reconcentrated in the near or far-field rock or at the geosphere-biosphere interface by physical or chemical processes, such as a precipitation at a redox front. Subsequent remobilisation of these radionuclides due to changed conditions may give rise to a pulsed discharge to the surface environment.

Technical description:

Mobile radionuclides released from the repository travel predominantly in the dissolved phase or in colloidal form, with minor release occurring in the gaseous state.

Mobile radionuclides travelling along a flow path potentially could be stopped at a particular location by a range of physical or chemical retardation processes. For example, colloidal particle movement could be halted by filtration in a low permeability formation. Alternatively, dissolved radionuclides could be precipitated from solution to form solid phases if the groundwater flow passes a redox or pH front causing a reduction in solubility (see GEN-27 “Radionuclide precipitation and dissolution”). Radionuclides could also be immobilised from water passing into a formation with a very high sorption capacity, such as an organic rich sediment near to the surface (see GEN-34 “Radionuclide sorption”).

Reconcentration could potentially occur around both the spent fuel and SFR repositories. In the spent fuel repository it could occur at a radiolytically-induced redox front, while at the SFR it might occur at a pH front.

If radionuclides continue to be immobilised at a particular location, over time an accumulation of radionuclides will occur, such that they become reconcentrated. Examples from nature are the roll-front ore deposits in which economic accumulations of metals can be created at a redox front. Theoretically, similar processes could occur in the far-field rock or at the geosphere-biosphere interface.

If accumulations of radionuclides do form, there is a possibility that changes to the physico-chemical environment could remobilise radionuclides, giving rise to a pulsed discharge to the environment. For example, near-surface accumulations could be physically released if the erosion rate were to increase suddenly (see GEN-29 “Erosion and weathering”). Precipitated nuclides could be chemically released if redox conditions became oxidising, such that solubilities increased.

It is probable that this mechanism would be significant only in the biosphere (and are currently considered in biosphere models) or at the geosphere-biosphere interface where rapid changes in conditions can occur. In the far-field, where physico-chemical conditions are generally more stable, it is unlikely that reconcentration followed by pulse release mechanisms would operate to any significant extent, unless initiated by the migration of a chemically distinct groundwater plume from the near-field, such as high pH groundwaters from the cementitious SFR (see SFR-23 “Groundwater chemistry in the near-field rock”). However, reconcentration could also occur close to the waste package where chemical gradients and fronts are most extreme, such as at a radiolytically generated redox front around a spent fuel canister (see SFL-38 “Redox fronts”).

Performance assessment models do not generally consider these reconcentration processes, and they were not explicitly considered in normal evolution scenarios for either the spent fuel or the SFR repositories. Nonetheless, it might be sensible to perform scoping calculations for repository sites that may be subject to rapid changes in external physico-chemical driving forces or where distinct groundwater plumes are expected to migrate from the near-field.

Origin in the repository system:

Reconcentration of radionuclides is influenced by the retardation processes that may operate in confined volumes of near and far-field rock. Reconcentration will be induced by spatial variations in the ambient chemistry (particularly redox and pH).

Impact on the repository system:

Reconcentration of radionuclides and subsequent release mechanisms may affect local groundwater chemistry and radionuclide release rates to the surface environment

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) and 3.2.03 (Sorption/desorption processes, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEP 4.1.6 (Reconcentration) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-32**

Name: **HYDRAULIC RESATURATION OF THE NEAR-FIELD ROCK**

Short description:

After closure of a repository, groundwater will flow from the saturated far-field rock into the void spaces in the near-field and will cause it to become hydraulically resaturated. This inflow of water will impact on the thermal and stress fields and, when in contact with the engineered barrier materials, will cause them to begin to corrode. During the resaturation process, the groundwater chemical condition will shift from oxidising in the near-field rock to increasingly reducing conditions.

Technical description:

During operation of a repository, groundwater flowing into the excavations will be pumped out. This will desaturate the surrounding rock mass and lead to the development of oxidising conditions within the near-field rock. Atmospheric pressure will prevail in the repository and fractures in the nearby rock will be partially drained. At repository closure, pumping is stopped and resaturation of the repository will begin. Local flow directions will gradually change from radially converging flow towards the underground openings to flow directions more aligned with the original (regional) flow field. However, the successive closure and sealing of sections of the repository as they are filled will lead to resaturation of parts of the repository before final closure.

Resaturation will be driven by hydraulic pressure (head) gradients between the unsaturated near-field and the saturated far-field rock. As long as atmospheric pressure prevails in the repository, the hydraulic head is determined by the location and depth of the repository. Therefore, drawdown effects will cause a reduction in head throughout the host rock.

Initially, groundwater will flow along fractures from the far-field into the engineered damaged zone of the near-field rock and this will begin to resaturate (see GEN-15 “Excavation effects on the near-field rock”). As the groundwater continues to flow it will pass from the near-field rock into the excavations themselves and will begin to resaturate the void spaces there.

Flow into the repository will be restricted by any shotcrete coating (if used) on the walls of the excavations. However, this shotcrete is not intended to be a post-closure hydraulic barrier and will only delay resaturation for a short period of time.

Because groundwater flow into the near-field will occur along hydraulically-active fractures, resaturation will tend to occur via discrete line or point sources depending on the degree of channelling within the fractures (see GEN-23 “Groundwater flow”). The presence of air and unsaturated conditions in the near-field rock will cause two-phase flow conditions with lower effective permeability and also trapped air may locally reduce the permeability until all the air is dissolved into the groundwater.

The head gradients caused by desaturation of the repository are likely to be significantly larger than regional flow gradients. Therefore these local gradients will dominate and the resaturation time depends on the permeability of the host rock and the dimension and permeability of the repository. An important consideration therefore is the state of desaturation of the repository and surrounding rock at closure. As mentioned above, the repository is likely to be unsaturated to various degrees. In the Nirex-97 performance assessment (Nirex, 1997) the resaturation of the repository was calculated to take only a few decades and did not, therefore, play a significant role in repository release and transport.

Resaturation of the near-field rock may be impacted by gas generation as well as by the remaining air trapped when the repository is closed. In particular, gas migrating from the repository will increase the time taken for resaturation. Sealing of a repository could delay resaturation and the subsequent escape of gas. This will have an impact on the radiological consequences of tritium (Nirex, 97).

It is likely that two-phase flow conditions initially exist in the rock zone disturbed by the excavation of the repository. Unsaturation and invasion of gas bubbles may temporarily change the hydraulic characteristics of the engineered damage zone. During resaturation, the possible two-phase flow effects in the disturbed rock zone and in the nearby rock will dissipate, to finally disappear at fully developed saturation.

During the resaturation process, the groundwater chemical conditions will shift from oxidising in the nearby rock to progressively increasing reducing conditions. It is likely that oxygen will be trapped during resaturation: this oxygen will be consumed by iron in the engineered barrier materials and the rock.

Resaturation of the near-field rock will lead to saturation and swelling of the bentonite buffer in the spent fuel repository and in the silo structure of the SFR repository. Manufacturing and engineering constraints require that the bentonite buffer is emplaced in the repository in a compacted state. The process of bentonite saturation is controlled by the suction and this therefore has the potential to be also a driving force for resaturation of the near-field rock. The rates of saturation of the bentonite decrease with increasing saturation of bentonite. Uneven resaturation of the near-field rock, due to, for example, concentrated flow along a few fractures, could lead to uneven swelling of the bentonite buffer, enhanced swelling pressures and, in the spent fuel repository, an unevenly distributed load on the canister (see SFL-1 “Swelling of the bentonite buffer”).

Origin in the repository system:

Resaturation of the near-field rock is influenced by the groundwater flow rate and by the geometry and hydraulic characteristics of the fracture network and by the two-phase (air-water) state of the near-field rock and the bentonite buffer. Resaturation rate may also be affected by processes in the engineered barriers, such as swelling of compacted bentonite and the production of gas and a gas overpressure.

Impact on the repository system:

Resaturation of the near-field rock will affect the groundwater flow in the near-field rock and the rate of resaturation of the excavations and any buffer and backfill materials in them. It may also affect the water chemistry in the near-field rock and the displacement of entrapped air.

Bibliographic references:

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Science Report, S/97/012. (Four volumes plus overview).

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.2.07 (Hydraulic/hydrogeological processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 5.1.4 (Resaturation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **GEN-33**

Name: **SEA LEVEL CHANGES**

Short description:

Changes in sea level are related to natural climate change processes over the long-term and to anthropogenic factors (global warming) over the short-term. Changes to the sea level will impact on a repository at a site that may change from being subaerial to submarine, or visa versa, largely by altering the groundwater flow system and chemistry.

Technical description:

On the short-term (up to 10^3 years), sea levels will rise around the world as the result of global warming brought about by anthropogenic changes in the greenhouse gas composition of the atmosphere. Sea level rises due to this process are expected to be limited to a few metres because an increase in temperature will cause the polar ice caps to begin to melt.

However, in Sweden, continued isostatic uplift of the land (resulting from the last glaciation) will mean that the relative sea-levels will drop in the short term (see GEN-39 "Uplift and subsidence"). Consequently, at the SFR which is presently beneath the Baltic Sea, isostatic uplift will mean that conditions will change from submarine to subaerial.

Over longer time periods, these processes will be overtaken by variations to the sea level that reflect natural climate change processes. Evidence indicates that the timing and severity of natural climate changes are determined by periodic changes in the Earth's orbit around the Sun (governed by the Milankovitch orbital parameters) which vary the amount of solar radiation reaching the Earth. The known periodicity of these parameters, together with other climate factors, such as atmospheric CO_2 levels, makes it possible to make estimates of the future climate sequence. For Sweden, some predictions suggest that glacial climates will occur approximately 60 000 in the future (e.g. King-Clayton et al., 1995).

Sea levels respond to climate change in a world-wide (eustatic) manner and, more locally, isostatically to tectonic influences and to ice-loading (see GEN-21 "Glaciation"). Sea levels during the Quaternary Period (the last 2 million years) have normally been substantially below present levels, since we are now experiencing a climatic optimum, and during the most of the Quaternary the climate has been colder than that of the present day. As the world's climate cools (from 5 000 years in the future, onwards) the eustatic sea level will drop, since water is taken out of the sea and is transferred to land-based ice sheets. The estimated average eustatic sea level over the next million years lies in the range -30 to -50 m, with periods when significantly lower sea levels, to as low as -140 m, will be experienced during times of glacial maxima.

The sea level changes local to Sweden are made complex by the presence of thick ice sheets, which develop initially on the land and which can cause large vertical displacements of the land surface of up to almost 1 km (see GEN-21 "Glaciation"). The phase lags associated with the changes in eustatic sea level and the isostatic response of the land to the removal and addition of ice sheets means that Sweden is expected to suffer from repeated and complex sequences of rises and falls in sea level over the next 1 million years. In addition, there may also be periods when large, ice-dammed fresh water lakes could form, at times of lowered sea level, so that the land is inundated not by saline, but by fresh water.

In addition, the prediction of sea level is made more complex by the uncertainties over the thermal expansion and contraction of the oceans due to temperature changes.

These long-term sea level changes induced by natural climate change can have a number of significant impacts on the repository.

Hydraulic gradients are likely to change in magnitude and direction, they may increase or decrease to zero, depending on whether the sea level rises or falls, to what extent this takes place and on the topography, both onshore and offshore. Hydraulic boundary conditions can also change (see GEN-16 “External flow boundary conditions”). Tilting of the land, which commonly takes place during glacial advances and retreats, will also change hydraulic gradients and will be accompanied by local changes in sea level. An increase in sea level would tend to reduce the topographically induced hydraulic heads controlling groundwater flow, while a decrease in sea level would tend to increase the hydraulic heads.

Variations in groundwater chemistry will also result from sea level changes because the chemistry of the recharge waters may alter from fresh to marine in origin (see GEN-6 “Groundwater salinity changes”). Fluctuations in the sea level can mean that a saline-fresh water interface will occur in the far-field rock in the vicinity of the repository which can have significant impacts on the groundwater flow system (density contrasts) and on the behaviour of released radionuclides.

Given that the average sea level over the next 1 million years will be substantially below the current level, this will mean that there will be a reduction in the potential diluting influence of the marine biosphere on radionuclide releases.

The change in sea level will also mean a change to the characteristics of the biosphere above the repository. A lowering in sea level may mean that the discharge zone changes from being in the marine environment to being in lakes or rivers.

Origin in the repository system:

Short-term sea level changes (increases) are anthropogenic in origin and relate to an increase in the concentration of greenhouse gases in the atmosphere.

Long-term sea level changes are natural and are caused by climate change which, itself, is due to variations in the solar radiation reaching the Earth’s surface. As such, sea level changes are caused by processes external to the repository.

Impact on the repository system:

Sea level changes can have a significant impact on the repository system by affecting groundwater flow systems and groundwater chemistry.

The groundwater flow system will change at a site if it moves from being subaerial to submarine (or coastal). This change will impact on the hydraulic gradient and direction and, thus, the direction and rate of groundwater flow. The groundwater chemistry will also change because the recharge waters can vary from fresh to marine in origin.

Both a change in groundwater flow direction and chemistry will have significant impacts on the transport of radionuclides in the far-field.

Bibliographic references:

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 1.3.03 (Sea level change) in the NEA International Database (NEA, 1998).

Relates to FEP 5.31 (Change in sea level) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-34**

Name: **RADIONUCLIDE SORPTION**

Short description:

Sorption is the collective term for the uptake of particles (molecules, ions, colloids) on the external and internal surfaces of materials. Sorption of radionuclides from the waste can occur on the engineered barrier system materials and their degradation products, as well as on the exposed mineral surfaces in the near and far-field rock. Sorption of radionuclides will retard the migration of those species.

Technical description:

Sorption is the collective term for uptake of particles (molecules, ions, colloids) on the surfaces of solids. The forces responsible for sorption range from 'physical' interactions (van der Waals' forces) to the formation of 'chemical' bonds. Sorption retards the transient diffusion of radionuclides in the near-field and the advective transport in the nearby rock and the far-field.

Sorption is element specific and depends both on radionuclide speciation (valency state, hydrolysis, complexation and groundwater composition) and the solid phase composition and surface characteristics. At true thermodynamic equilibrium these two sets of conditions are linked together.

Sorption encompasses a range of mechanisms which result in the uptake of solute onto solid surfaces, such as ion-exchange and physical adsorption. Generally, these are fast, reversible reactions and consequently sorption is generally modelled as a reversible process whereby sorbed radionuclides may be released to solution if the solution concentrations or composition change. Desorption kinetics are generally slower than sorption kinetics and there may be instances where sorption is effectively irreversible.

Precipitation and co-precipitation are not sorption processes but it can be difficult to discriminate between these mechanisms in both field and laboratory studies (see GEN-27 “Radionuclide precipitation and dissolution”). Generally, sorption would be expected to be the dominant process at low solute concentrations while precipitation can only occur at high concentrations (exceeding saturation). Although hard to distinguish between the two in some practical studies, it is important to be able to discriminate between them. In performance assessment, sorption and precipitation are modelled in different ways.

In general, there are two methods of treating adsorption-desorption reactions at mineral surfaces; an empirical approach and a conceptual model for surface complexation. With respect to the empirical approach, in the equation for radionuclide transport, sorption is represented by the parameter K_d, or (mass) distribution coefficient. This parameter may be defined as the equilibrium partitioning of an element between solid and liquid phases and is the ratio of the total concentration (per unit mass) of elemental species sorbed on the solid to the total concentration (per unit volume) of elemental species in solution.

Surface complexation models are based on a thermodynamic treatment of the electrical double layer at the solid-water interface (Davis and Hayes, 1986). The double layer is created by ions, originally free in solution, being subsequently bound to the interface. Complexes can form at the interface in a similar way to in a bulk solution. Thus surface reactions are modelled for sorbing species, and thermodynamic parameters are used to determine sorption at the interface. Necessary input data (intrinsic equilibrium constants, surface site density etc.) are not always easy to obtain experimentally. Despite this drawback, surface complexation modelling has been used successfully to model pH dependent sorption in simple geochemical systems (e.g. Langmuir, 1997).

K_d has its origins as a thermodynamic parameter but, due to the complexity of natural systems, is now considered as an empirical parameter which can vary by several orders of magnitude depending on chemical conditions at the solid-liquid interface.

Sorption may also be represented by a surface-based sorption coefficient, K_a, which may be defined as the ratio of the total concentration (per unit area) of elemental species sorbed on the solid to the total concentration (per unit volume) of elemental species in solution.

The argument for using K_a rather than K_d is that, since sorption is a surface process, all of the individual grains in a mass of bulk rock may not be available for sorption. However, measurement of K_a depends on being able to measure the surface area of the solid which is not easy experimentally and, consequently, such sorption measurements are limited.

The implicit assumption in using the parameter K_d is that the system being studied behaves ideally and in particular, that sorption is reversible and independent of solute concentration or of other species present. For some elements, however, notably caesium, this is not the case and is described better using the Freundlich isotherm.

Since sorption occurs at the liquid-solid interface, the solid phase has a major influence on the extent of sorption. For example, most clay minerals have large surface areas and a correspondingly large number of surface sites available for sorption. Consequently, measured K_d values for those elements which sorb by ion-exchange are higher for clay minerals than for low-capacity minerals such as quartz. The cation-exchange capacity (CEC) of clay minerals can be measured experimentally and is a good measure for predicting the extent of sorption by ion-exchange.

In a repository, sorption sites are provided by the engineered barrier system materials (e.g. the bentonite buffer) and their degradation products (e.g. iron oxyhydroxides), as well as on the exposed mineral surfaces in the near and far-field rock (e.g. fracture coating minerals).

The kinetics of sorption on these materials is clearly important to long-term predictions of radionuclide behaviour. Irreversible sorption is obviously beneficial in safety assessment terms but is difficult to demonstrate, and the most robust performance assessment models make the conservative assumption that all chemical retardation processes are instantaneously reversible.

The kinetics of sorption processes are generally fast enough to allow their exclusion from performance assessment models. In an essentially very sluggish groundwater flux environment such as the near-field of some repository concepts, the system is often modelled as a chemical mixing tank, where the presence of sorbing surfaces is a key part of the model generating steady-state solution concentrations of radionuclides. In this environment, the kinetics of sorption processes may become important if any mechanisms are identified which can lead to gross chemical inhomogeneities in the system or rapid groundwater transit pathways through the system.

In the far-field, sorption kinetics are not considered in safety assessment studies, although precipitation and mineralisation kinetics would clearly be relevant if these processes were to be included in an assessment. Where groundwater fluxes are relatively rapid (as may occur, for example, in a major fracture zone), then retardation mechanisms become of diminishing significance in affecting the rates of release of radionuclides, and, in some assessments, are given no credit.

Origin in the repository system:

Sorption is a radionuclide retardation process that can occur on the engineered barrier system materials and their degradation products, as well as on the exposed mineral surfaces in the near and far-field rock.

Sorption is element specific and depends both on radionuclide speciation (valency state, hydrolysis, complexation) and the solid phase composition and surface characteristics. At true thermodynamic equilibrium these two sets of conditions are linked together.

In an ideal system, sorption is instantly reversible while being independent of solute concentration and of the presence of other solutes. However, in real systems, sorption behaviour is more complex.

Impact on the repository system:

Sorption of radionuclides on engineered barrier system materials and their degradation products, and mineral surfaces will retard those radionuclides. It is thus one of the primary chemical processes responsible for the safety of the repository.

Sorption will affect the distribution of radionuclides in the near and far-fields, as well as their concentration in the groundwaters.

Bibliographic references:

Arthur RC (1997) Adaptation of sorption models for performance assessment calculations. SKI Report (in publication).

Davies JA and Hayes KF (1986, editors) Geochemical processes at mineral surfaces. American Chemical Society Symposium Series, 323.

Langmuir D (1997) The use of laboratory adsorption data and models to predict radionuclide releases from a geological repository: a brief history. Material Research Society Symposium Proceedings, 465, 769-780.

McKinley IG and Hadermann J (1985) Radionuclide sorption database for Swiss safety assessments. Nagra Technical Report, NTB 84-40.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 3.2.03 (Sorption/desorption processes, contaminant) in the NEA International Database (NEA, 1988).

Relates to FEP 4.1.4 (Sorption) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-35**

Name: **FAST TRANSPORT PATHWAYS**

Short description:

Fast pathways for water and radionuclide transport from depth towards the surface can occur in the form of highly transmissive natural features (such as large fractures) or as poorly sealed shafts and boreholes. The presence of such fast pathways could potentially short-circuit large regions of the far-field rock and lead to early and large releases to the surface.

Technical description:

The far-field rock will contain numerous natural features that may act as pathways for water flow (see GEN-23 “Groundwater flow”). However, the site characterisation programme should ensure that the repository is located remote from any particularly large or highly transmissive features. Nonetheless, there remains a possibility that some fast pathway features may go unnoticed in the far-field or could form after repository closure.

Fast pathways may occur in the far-fields of both the spent fuel and SFR repositories and, thus, may be significant to the safety of both repository designs.

Pre-existing highly transmissive features could be in the form of large fractures or fracture zones; unconformities between two lithological units; depositional structures in sedimentary rocks; or the contacts between igneous intrusions and the host rock. However, features such as these are most likely to have been identified in the site characterisation programme.

Highly transmissive features could be formed during repository construction (see GEN-15 “Excavation effects on the near-field rock”), in the engineered damaged zone, or form in the far-field after repository closure due to the reactivation of existing faults or formation of new faults as a consequence of changes in the stress field (see GEN-17 “Faulting”). This may be brought about by glaciation, radiogenic heat output or tectonic forces.

A third type of fast pathway could result from the failure of seals in shafts and boreholes (or from unsealed boreholes) at some point after repository closure (see GEN-7 “Degradation of the borehole and shaft seals”).

Regardless of the nature and mode of formation of fast pathways, if they occur in the far-field rock they present a mechanism for rapid radionuclide transport from depth towards the surface, provided the radionuclides are capable of migrating from the near-field to connect with the fast pathway at depth.

Fast pathways offer limited opportunity for radionuclide retardation because they can have a relatively small wetted surface area for sorption (see GEN-34 “Radionuclide sorption”) and less opportunity for diffusion into the rock matrix (see GEN-25 “Matrix diffusion”), relative to the many small fractures required to carry a similar volume of groundwater. Thus, if fast pathways do exist in the far-field rock, they can result in very early and large radionuclide releases to the surface.

If it can be demonstrated that the site characterisation methodology is effective at identifying potential fast pathways that might be active under post-closure hydrogeological conditions, and all boreholes and shafts are well sealed, then fast pathways should not be of concern in normal repository evolution assessments. They become of interest when considering other scenarios.

Origin in the repository system:

Fast pathways may occur in the far-field rock if they went unnoticed during site characterisation, if the rock fractures during the post-closure phase, or if the seals fail in boreholes and shafts. Fast pathways formation is thus largely dependent on the stress field and on the degradation processes affecting the seals.

Impact on the repository system:

If fast pathways occur in the far-field rock, they can act as short-circuit routes for radionuclide transport to by-pass large sections of the far-field rock. They potentially may result in very early and large radionuclide releases to the surface.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.2.06 (Mechanical processes and conditions in geosphere) and 3.2.07 (Water mediated transport of contaminants) in the NEA International Database (NEA, 1988).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **GEN-36**

Name: **STRESS FIELD**

Short description:

The rock mass around a repository will be subject to a regional applied stress which is a combination of gravitational loading and active tectonic stress. Local changes in the stress field are caused by effects such as the presence of the underground openings, thermal loading and by the expansion of the bentonite buffer and backfill.

Technical description:

The stress at any point in a rock mass will be determined by a variety of processes and effects. The stress can be defined as total stress or effective stress, where the effective stress is less than the total stress by the magnitude of the hydrostatic pressure, at least for those parts of the rock mass that are hydraulically connected, i.e. where the law of effective stress is applicable. The stress state can be represented by a stress tensor or stress ellipsoid and can be described in terms of three principal stresses, s1>s2>s3. In areas on low relief and where tectonic activity is subdued, such as in Sweden, s1 is often horizontal or sub-horizontal, s2 vertical and s3 also horizontal.

Distant from the repository, the in situ stress tensor will be determined by the gravitational loading at that point, which is determined by the depth and the density of the overlying rocks, and by the applied stress, which is determined by tectonic factors. There will be local changes in the orientation and magnitude of the stresses due to factors such as the local topography, although in an area of subdued relief, such is likely to be the situation in the area of a repository, this effect will be minimal.

Changes in tectonic activity will cause changes in stress magnitude, but such changes are also thought likely to be insignificant in Sweden over the next 1 million years. Changes will also take place due to seismic activity, with stress build up on faults being dissipated during seismic events (see GEN-17 “Faulting”). Similarly, this is not considered important in the Swedish context, though stress cycling will take place which may affect groundwater systems by altering fracture apertures and fracture connectivity. An extreme example of this would be seismic pumping, where groundwater is expelled and/or taken back into the rock mass due to elastic rebound following seismic events. The significance of this process in the Swedish context is not thought to be great.

During times of glacial advances (see GEN-21 “Glaciation”), the stresses applied to the rock mass will vary greatly due to the unloading effect of the sea water as eustatic sea levels decrease and, more importantly, due to the loading by ice sheets. The stresses applied to the rock mass by ice sheets will depend on the thickness of the ice sheet; the shape of the top surface of the ice sheet but only where surface ice gradients are significant near the margins of the ice sheet; and the hydrostatic pressures developed in the rock mass.

The greater the hydrostatic pressure the lower will be the effective stress acting on the rock mass. The hydrostatic pressure will itself be determined by the hydraulic head within the ice and the extent of drainage of the water from the base of the ice sheet and within the rock mass. This, in turn, will be determined by the form of the ice sheet, i.e. whether it is cold or warm based and on the hydraulic conductivity of the underlying rock mass, the most important factor here being whether or not it is frozen.

In the area around the spent fuel repository, the regional stress field will be modified due to bentonite swelling (see SFL-1 “Swelling of the bentonite buffer”), the thermal gradient imposed by the waste (see SFL-46 “Temperature of the near-field”) and stress concentrations around repository excavations. Stress concentrations will be present around all underground openings, with maximum stresses being up to approximately 3 times the regional stresses. The orientation and shape of the underground openings will influence the magnitude, location and form of the stress concentrations.

Determining the in situ stresses can be carried out using standard equipment and is now a routine procedure in crystalline rock to the depths of interest for a repository in Sweden. Calculating what the stress changes are likely to be due to construction activities and thermal loading can be done using finite element techniques. What is more difficult, and is the subject of current research as part of the DECOVALEX programme (e.g. Stephansson et al., 1994), is thermo-hydro-mechanical (THM) modelling, which examines the coupling between the stress changes and changes in groundwater flow.

Stress changes in the near-field rock will be determined by the strength of the intact rock, the fracture pattern and the rock’s thermal properties. Stress changes may result in deformation of the rock mass by faulting and by creep of the rock mass. This may cause damage to the backfill and the buffer and could, ultimately affect the waste canister itself. Sufficiently large changes in stress could result in collapse of the repository openings. It is more likely, however, that stress changes will affect the hydraulic properties of the fracture network and create enhanced rock fracturing (see GEN-14 “Enhanced rock fracturing”).

The largest anticipated changes in stress within the near-field will be due to loading by ice sheets. Ice thicknesses of up to 3 km could be expected on several occasions over the next 1 million years. Modelling of the deformation caused by this loading shows how fracture displacements could take place, with a concomitant changes in their hydraulic properties. However, the stresses due to the ice sheets are only relevant to the behaviour of the spent fuel repository because glaciation will not occur until beyond the period of interest to performance assessments of the SFR.

Origin in the repository system:

The stress field around the repository is the combination of the weight of the overlying rock, the natural regional tectonic processes and localised features cause by the repository excavations and the thermal loading. This stress field may then be modified by other processes, such as glaciation and rock movements.

It is only where stresses become excessive or where large changes in stress occur that stresses themselves become significant for understanding repository evolution

Impact on the repository system:

Changes to the stress field can be important for repository evolution. They can cause movement to the rock mass by faulting, creep and cave-in, and can alter the hydrogeological system by changing the hydraulic characteristics of the fractures.

Bibliographic references:

Hanson H, Shen B, Stephansson O and Jing L (1995) Rock mechanics modelling for the stability and safety of a nuclear waste repository. SKI Technical Report, 95:41.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Shen B and Stephansson O (1996) Modelling of rock fracture propagation for nuclear waste disposal. SKI Technical Report 96:18.

Stephansson O, Jing L, Tsang CF and Kautsky F (1994) Development of coupled models and their validation against experiments - DECOVALEX project. Proceedings from the NEA/SKI GEOVAL Symposium, 1994.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.2.06 (Mechanical processes and conditions in the geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.7 (Thermo-hydro-mechanical effects) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **GEN-37**

Name: **SURFACE WATER CHEMISTRY**

Short description:

Surface waters are the rivers, lakes, ponds etc. that cover part of the continental land masses. These water bodies can act as recharge zones for groundwater and, as such, can be a control on the far-field groundwater chemistry.

Technical description:

Surface waters are the rivers, lakes, ponds etc. that occur on the continental land masses. Some of these features occur for long periods of time and can be considered permanent, while others are transient features that may exist for only short periods of time relative to the lifetime of a repository.

Generally, surface water bodies contain 'fresh' water, as opposed to the seas and oceans which are saline. These surface water bodies can act as recharge and discharge zones for groundwater and, as such, they are a control on the groundwater chemistry in the upper part of the rock mass.

As water from the surface infiltrates downwards through the rock, it will interact with the exposed mineral surfaces (see GEN-1 "Alteration and weathering along flow paths") and will mix with the existing groundwater bodies at depth. Due to these processes, the chemistry of the water will tend to change with depth. In general, in a crystalline or argillaceous host rock, the rock-water interactions cause waters to become chemically reducing, pH neutral to mildly alkaline and more saline with depth and time since infiltration. However, the exact composition of deep groundwaters is strongly site specific, dependent on the mineralogical characteristics of the rock mass, the nature of the groundwater flow system and the composition of the recharge waters (see GEN-22 "Far-field groundwater chemistry").

Where there is a slow, gradual change in groundwater chemistry with depth, it is normal to consider the surface and near-surface waters (which are generally oxidising) separately from the deeper far-field waters (which are generally reducing).

Under certain circumstances, however, the surface waters may infiltrate more rapidly to depth so that their chemistry is not appreciably changed by slow rock-water interactions. This might happen along highly permeable fracture zones which might dilate during processes such as uplift or tectonic activity. The rapid intrusion of surface waters may also be a result of glaciation or may be caused by human activities e.g. drilling of boreholes or excavation of shafts in the vicinity of the repository. If this were to occur, oxidising waters at depth may intrude into the existing reducing waters, forming sharp interfaces between waters of different chemistry (see GEN-24 "Interfaces between different waters").

The chemistry of the surface water bodies is generally dependent on the erosion and weathering of the surface rocks and sediments which, in turn, is controlled by the climate state (see GEN-29 “Erosion and weathering”). However, variations to the surface water chemistry can be caused by local (time dependent) surface process. For example, changes in infiltration areas and rates will change the chemistry of near-surface waters, as will human activities. However, climate change will be the main natural control on the surface water chemistry and on the recharge; in particular, climate change induced glaciation (see GEN-21 “Glaciation”), permafrost (see GEN-26 “Permafrost”) and sea level changes (see GEN-33 “Sea level changes”) can have severe impacts on the chemistry, volume and location of surface and recharge waters.

Analysis of these rapid surface water penetration processes suggest that they potentially could reach repository depths over timescales relevant to performance assessment. Arthur (1996) estimated migration velocities of redox fronts that could be generated in granitic rocks by the downward flow of oxygenated glacial meltwaters. This study suggested that redox front velocities would be attenuated by redox reactions involving ferrous silicate, oxide and sulphide minerals, and that the extent of attenuation was proportional to the rates of oxygen consumption and inversely proportional to the product of buffer intensity factors (stoichiometric number of moles of oxidant consumed) and capacity factors (modal abundances of Fe^{II} minerals). The results suggest that oxidising glacial meltwaters could migrate to repository levels within 10 000 to 20 000 years in the case of slow kinetics for oxygen consumption, which is comparable to periods of sustained infiltration of meltwaters during glacial-interglacial events in Sweden over the next 10^5 years.

Even faster potential travel times were indicated in a further analysis by Glynn and Voss (1996). In one calculation, these authors applied a similar approach to that adopted by Arthur (1996) but considered the case of a single ‘relatively clean’ fracture and a more realistic capacity factor. In this case, a travel time of less than 1000 years was estimated. In further calculations using kinetic models, the travel time was calculated on the basis of the results of biotite and chlorite oxidation experiments (Malström et al., 1996). In these calculations, travel times of much less than 1000 years and possibly as low as 60 years were estimated.

These results indicate that considerable uncertainty exists with regard the actual impact of infiltrating glacial waters. Nonetheless, in the absence of glaciation, it is very unlikely that oxidising surface waters could impact directly on the repository near-field.

Origin in the repository system:

The surface water chemistry is controlled by the erosion and dissolution of surface materials. During recharge, surface waters will undergo rock-water interactions with the exposed mineral surfaces and will be modified.

The depth to which chemically distinct surface waters may penetrate is controlled by the recharge rate and the porosity and permeability of the flowing zones.

Impact on the repository system:

The surface water chemistry will affect directly the chemistry of the recharge and is a primary control on the chemistry of the far-field waters.

Under certain circumstances, chemically distinct surface waters may penetrate to depth in the rock where they can mix with the ambient groundwater changing the far-field groundwater chemistry. If this occurs, the surface water chemistry may affect radionuclide transport and retardation processes in the far-field rock.

Bibliographic references:

Arthur RC (1996) Estimated rates of redox-front migration in granitic rocks (SITE-94). SKI Technical Report, 96-35.

Glynn P and Voss C (1996) Geochemical characterisation of Simpevarp groundwaters near the Äspö Hard Rock Laboratory (SITE-94). SKI Technical Report 96-29.

Malström M, Banwart S, Lewenhagen J, Duro L and Bruno J (1996) The dissolution of chlorite and biotite at 25°C in the near-neutral pH region. Journal of Contaminant Hydrology, 21, 201-213.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.3 (Surface environment) in the NEA International Database (NEA, 1998).

Relates to FEP 7.8 (Altered surface water chemistry by humans) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **GEN-38**

Name: **TEMPERATURE OF THE FAR-FIELD**

Short description:

The temperature of the far-field is largely controlled by the natural geothermal gradient, although it may be influenced by heat output from the repository and by changing climate at the surface. The temperature in the far-field will be a control on the rates of chemical and microbiological processes, and can influence the stress field, groundwater flow, diffusion rates and, hence, radionuclide transport.

Technical description:

The temperature of the far-field is largely controlled by the natural geothermal gradient, although there will be some influence from repository heat output and climate change.

The ambient temperature of the far-field rock is controlled by the natural geothermal gradient (approximately 10 to 20°C/km for Sweden) and the mean surface temperature (approximately 5 to 10°C for Sweden).

Heat will be generated in the repository by radioactive decay (see GEN-28 “Radioactive decay”) and by curing of any cement. Thus, for the spent fuel repository, radiogenic heat production is most significant and, for the SFR, exothermic cement curing is most significant. Heat generation by both processes is time dependent and will decrease with time.

The heat generated in the repository will be dissipated by conduction through the rock mass and, to a much lesser extent, by advection in the moving groundwaters. Consequently, the thermal impact of the repository will reduce with distance from the near-field. As a result, the far-field rock temperature will be raised above that predicted by the geothermal gradient by an amount dependent on the distance from the near-field and the time after repository closure.

Performance assessment calculations indicate that the temperature rise in the far-field rock as a consequence of the repository is quite limited. For example, around a spent fuel repository, the temperature rise in the far-field due to radiogenic heat is around 10°C at a distance of 200 m, at a time 5000 years after closure (Claesson and Probert, 1996; Probert and Claesson, 1997). However, the more important process is the thermal stresses imposed on the rock by the thermal gradient which can cause displacement on local faults and joints (see GEN-17 “Faulting”).

The far-field rock temperature may also be affected to a small degree by the onset of glacial conditions at the land surface which may reduce the annual mean temperature from around 10°C at present to below zero (see GEN-21 “Glaciation”). Nonetheless, the thermal impact of glacial conditions is unlikely to be significant below a depth of approximately 500 m, which is the maximum predicted depth that permafrost (see GEN-26 “Permafrost”) could reach during a prolonged period of cooling (McEwen and de Marsily, 1991).

The far-field temperature is, thus, largely controlled by the natural geothermal gradient, with minor and time dependent fluctuations caused by heat output from the repository and climate change.

The far-field temperature will be a control on the rates of chemical and microbiological processes that occur there. However, few geochemical and microbiological processes relevant to repository safety (other than sorption) will occur remote from the near-field.

Due to the thermal gradient arising from the repository, rock displacement could occur in the far-field as a consequence of thermal-mechanical coupling, causing opening and closing of fractures. This could affect the groundwater flow field through changes in the permeability of the rock, and causing temporary over or underpressures, although the latter impact will be much less than that due to pressure (head) differences. However, the effects will be less than in the near-field rock because the maximum temperature and thermal gradient will be lower in the far-field rock than in the near-field rock.

Due to the lower impacts on the far-field rock, the thermal modelling undertaken in some performance assessments is focussed only on the near-field and the adjacent rock, although this is not necessarily a conservative modelling approach.

Origin in the repository system:

The temperature in the far-field rock is largely controlled by the natural geothermal gradient, with time dependent fluctuations caused by heat output from the repository and climate change.

Impact on the repository system:

The temperature in the far-field rock is a control over far-field geochemical and microbiological processes, the stress field and groundwater flow. However, other processes (e.g. head differentials) are generally more significant.

Bibliographic references:

Claesson J and Probert T (1996) Temperature field due to time-dependent heat sources in a large rectangular grid - derivation of analytical solution. SKB Technical Report, TR 96-12.

Come B (1995) A review of thermal, mechanical and hydrogeological properties of hard, fractured rocks (granite) for deep geological disposal of radioactive wastes. Proceedings of a workshop held in Brussels, 12-13 January 1995. EC Report, EUR 16219 EN.

Haijtink B (1995) Testing and modelling of thermal, mechanical and hydrogeological properties of host rocks for deep geological disposal of radioactive waste. Proceedings of a workshop held in Brussels, 12-13 January 1995. EC Report, EUR 16219 EN.

Gobet A and de Marsily G (2000) Evaluation of the thermal effects in a KBS-3 type repository. SKI Technical Report, in press.

McEwen TJ and de Marsily GH (1991) The potential significance of permafrost to the behaviour of a deep radioactive waste repository. SKI Technical Report, 91:8.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Probert T and Claesson J (1997) Temperature field due to time-dependent heat sources in a large rectangular grid - application for the KBS-3 repository. SKB Technical Report, TR 97-27.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Tarandi T (1983) Calculated temperature field in and around a repository for spent fuel. SKB Technical Report, TR 83-22.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **GEN-39**

Name: **UPLIFT AND SUBSIDENCE**

Short description:

Uplift and subsidence are caused by tectonic and glacial process, the most significant of which is isostatic rebound in response to deglaciation. Land uplift around a repository may lead to a change in the groundwater chemistry, increased groundwater flow through the repository, increased erosion rates and a change in the biosphere characteristics.

Technical description:

Vertical movements of the land surface can involve uplift or subsidence. Uplift of the land surface may occur as a result of compressional tectonics, igneous processes or isostatic rebound (stress relief due to deglaciation or erosion). Subsidence may be the result of extensional tectonics, subsidence or glaciation.

The most important process leading to vertical movements in Sweden, over the time periods of interest to performance assessment, is past and future glaciation (see GEN-21 “Glaciation”). In the future, the formation of ice sheets will cause depression of the land surface beneath it. Given densities of approximately 0.9 g/cm^3 for the ice sheet and 2.7 g/cm^3 for the underlying bedrock, a continental ice sheet 3 km thick could, in theory, depress the land surface by about 1 km.

According to one estimation, glaciation is anticipated to occur again in Sweden approximately 60 000 in the future (e.g. King-Clayton et al., 1995). Hence subsidence due to glaciation is a significant process for the spent fuel repository because this time is within the period of interest for performance assessments for this repository. In the case of the SFR, this time is beyond the period of interest for performance assessments and, consequently, glacially induced subsidence is not considered significant for the SFR.

When an ice sheet is removed, the land rises upwards to restore its previous elevation at equilibrium. This is known as isostatic rebound. There is a continuous ongoing isostatic rebound in Sweden following the last glacial event. The maximum rate of uplift in northern Sweden is 9 mm/yr, in Stockholm it is 5 mm/yr and in some parts uplift has effectively stopped. Field studies indicate that the greatest rate of uplift occurred shortly after the retreat of the ice and has slowed since then. The total cumulative land uplift since deglaciation is estimated at about 850 m. The remaining future uplift is estimated to lie between 20 m and 200 m. Because this uplift is ongoing, it is relevant to both the spent-fuel and SFR repositories. Consequently, at the SFR which is presently beneath the Baltic Sea, isostatic uplift will mean that conditions will change from submarine to subaerial.

The ongoing land uplift is mainly due to compression of the rockmass under the ice load during the last glaciation. This is a similar movement that the bedrock has undergone repeatedly in connection with previous glaciations.

It is unclear whether disturbances in the state of equilibrium of the crust due to ice loading and unloading would affect a repository at depth. In the KBS-3 performance assessment it was argued that it would not affect a spent fuel repository at 500 m depth. Rosengren and Stephansson (1990), modelled the impact of the load of an ice sheet on the Finnsjön site in central Sweden. They concluded that most of the vertical movements associated with isostatic rebound will take place in the major fracture zones, which the repository will avoid.

At the surface, uplift will have greater impacts. It will cause an increase in the rate of erosion because erosion rates generally increase with elevation, although it is not expected that the increase in erosion rate will be significant for repository safety (see GEN-29 “Erosion and weathering”).

Uplift will also cause a change in relative sea-level (see GEN-33 “Sea level changes”) and this may have a significant impact on the groundwater flow system in the far-field, particularly for coastal sites. The geometry of the groundwater flow system may be significantly altered, as may be the geochemistry of recharge waters (see GEN-37 “Surface water chemistry”). For example, relative sea-level fall may cause dissolution of sulphate minerals due to the ingress of meteoric waters and the resulting increase in average rock porosity, fracture aperture and hence hydraulic conductivity (for a saline base case). The hydrochemistry of the near-field may be affected by an influx of fresher, more oxidising meteoric waters, as anticipated for the SFR.

Land uplift will have a particularly significant impact on the SFR repository. The current rate of uplift at Forsmark is approximately 6 mm/yr. Calculations show that around 30 m of uplift remain from the last glacial event. The southern part of the site is estimated to be above sea-level after 600 to 800 years and the other parts of the site will be above sea level after 1000 years. The major consequences of the land uplift is to change the groundwater chemistry from a saline regime to fresh water and to increase the flux of groundwater through the repository.

One further effect of uplift will be a change in the biosphere characteristics. During the salt water period, consumption of fish is the main biosphere pathway. During the inland period the critical group is a farming population subsisting exclusively on its own produce, eating fish caught in the sea, drinking water from ground abstraction and using lake water for irrigation.

Origin in the repository system:

Uplift and subsidence is caused by forces external to the repository system. Climate forces (causing glaciation and deglaciation) are most significant, although tectonic forces will also cause uplift over longer time periods.

Impact on the repository system:

The impact of uplift and subsidence on the repository at depth will be minimal and limited to causing reactivation of pre-existing faults and fractures in the far-field rock.

At the surface, uplift and subsidence will cause changes to the recharge and discharge zones for the groundwater flow system, changes to surface erosion rates and the characteristics of the biosphere. Most significant changes will be due to relative changes in sea-level caused by isostatic uplift.

Bibliographic references:

King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Rosengren L and Stephansson O (1990) Distinct element modelling of the rock mass response to glaciation at Finnsjön, central Sweden. SKB Technical Report, TR 90-40.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

SKI (1994) Evaluation of SKB's In-depth Safety Assessment of SFR-1. SKI Report, 94:30.

Equivalent FEPs:

Relates to FEP 1.3.03 (Sea level change) and 2.3 (Surface environment) in the NEA International Database (NEA, 1998).

Relates to FEP 5.16 (Uplift and subsidence) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-1**

Name: **COLLOID GENERATION IN THE WASTE PACKAGE**

Short description:

Colloids may be generated by chemical, physical and microbiological processes in the waste package. Radionuclides can sorb onto these colloids which may affect their subsequent transport out of the near-field.

Technical description:

Colloids are small, solid particulate materials suspended and dispersed in the groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed 'suspended particles' and anything smaller is considered to be in solution.

The significance of colloids arises from the fact that they can sorb radionuclides and can be mobile in flowing groundwaters. This means that colloids can increase the apparent concentration in groundwater of radionuclides with low true solubilities. In addition, because of their very small size, they provide a very large sorptive area. The transport and retardation of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution.

In the SFR repository, a number of different waste packages will be used in the silo and in the vaults. The waste package comprises the wasteform (metal, organic waste and inorganic waste), the immobilisation matrix (cement or bitumen), the container (steel or cement) and any overpack (cement).

Most of these materials can form colloids when they degrade by chemical, physical and microbiological processes. Radionuclides could be associated with the colloids by sorption onto the colloid surfaces or they may be directly included in the colloidal material if the colloid is a small fragment of the radionuclide-bearing wasteform itself. In addition, colloids may be formed by direct precipitation within the waste package as a result of the thermal and chemical gradients that may occur there.

The stability of colloids formed within the waste package will be dependent on pH and ionic strength in such a manner that high pH and high cation concentrations tend to destabilise colloids.

Of the various materials in the waste packages, the most significant for the production of colloids would be the cement. This is partly because cement is the most abundant material in the near-field (by volume) but also because cement will generate colloids as it matures and degrades. Furthermore, colloids are likely to be generated at the pH 'front' that is created at the leading edge of the alkaline plume that will migrate out of the near-field into the host rock (see SFR-23 "Groundwater chemistry in the near-field rock"). This processes will occur because certain species (e.g. silica) display significantly different solubilities in alkaline and pH-neutral conditions, and precipitate out in colloidal form at the pH front. Any radionuclides moving with, or located at the pH front, may become associated with the colloids when they form.

Colloids formed in the waste packages within the silo are unlikely to increase radionuclide transport rates to the near-field rock because the layer of bentonite between the silo and the host rock will act as a colloid filter for as long as the barrier remains intact (e.g. Kurosawa et al., 1997). In the rock vaults, however, where there is no bentonite, no significant colloid filtration will occur (see SFR-10 “Colloid filtration in the near-field”).

Nonetheless, colloidal transport of radionuclides from the SFR repository is not considered to be a significant process controlling releases. Other processes such as gas generation and transport, and the formation of complexing agents from the degradation of organic materials are more important processes. As such, the formation and behaviour of colloids has not been addressed in the performance assessments for the SFR.

Colloids were considered in the Nirex-97 performance assessment of the proposed UK cementitious ILW repository (Nirex, 1997). The calculations in this assessment also indicated that colloids are less important than complexing agents for enhancing radionuclide transport. The Nirex-97 performance assessment concluded that colloids only become significant in a scenario where no organic complexants occur in the groundwaters.

Origin in the repository system:

Colloid generation in the waste packages of SFR is controlled by the groundwater chemistry and temperature which control the degradation of the materials that comprise the waste package. In particular, colloids will be created by the degradation of cement and at the pH front at the leading edge of the alkaline plume.

Impact on the repository system:

Colloids generated in the waste packages may enhance radionuclide transport out to the near-field rock. However, the significance of this processes is not as great as that due to complexing agents formed by the degradation of organic materials in the repository.

Bibliographic references:

Kurosawa S, Yui M and Yoshikawa H (1997) Experimental study of colloid filtration by compacted bentonite. Scientific Basis for Nuclear Waste Management, 465, 963-970.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Report No S/97/012, Four Volumes.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1994) evaluation of SKB’s in depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS), 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) and 3.2.04 (Colloids, contaminant interactions and transport with) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-2**

Name: **COLLOID GENERATION AND TRANSPORT IN THE SHELL AND GROUT**

Short description:

Colloids may be generated by chemical, physical and microbiological processes in the shell and grout of the silo. Radionuclides can sorb onto these colloids which may affect their subsequent transport out of the near-field.

Technical description:

Colloids are small, solid particulate materials suspended and dispersed in the groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed 'suspended particles' and anything smaller is considered to be in solution.

The significance of colloids arises from the fact that they can sorb radionuclides and can be mobile in flowing groundwaters. This means that colloids can increase the apparent concentration in groundwater of radionuclides with low true solubilities. In addition, because of their very small size, they provide a very large sorptive area. The transport and retardation of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution.

In the silo of the SFR repository, the shell of the silo is formed from concrete with steel reinforcing rods while, within the silo, all spaces between waste packages will be backfilled with a porous, cementitious grout.

The cement in the shell and grout could produce colloids as it matures and degrades (see SFR-3 "Degradation of the cement mortar and the silo shell"). Iron oxyhydroxide colloids may also be created from the degradation of the steel reinforcement rods in the shell. Radionuclides could be associated with any of these colloids by sorption onto the colloid surfaces.

In addition, colloids may be formed by direct precipitation within the silo as a result of the thermal and chemical gradients (especially pH) that will occur there. This processes will take place because certain species (e.g. silica) display significantly different solubilities in different chemical conditions and precipitate out in colloidal form across a chemical gradient. Any radionuclides released to the grout may become associated with the colloids when they form.

A third source of colloids in the silo are colloids generated in the waste package.

The stability of colloids formed within the silo will be dependent on pH and ionic strength in such a manner that high pH and high cation concentrations tend to destabilise colloids.

Colloids formed in the silo shell or in the grout are unlikely to increase radionuclide transport rates to the near-field rock because the layer of bentonite between the silo and the host rock will act as a colloid filter (see SFR-10 "Colloid filtration in the near-field").

As such, colloidal transport of radionuclides from the silo is not considered to be a significant process controlling releases (see SFR-19 “Transport and release from the silo”). Other processes such as gas generation and transport, and the formation of complexing agents from the degradation of organic materials are more important processes. Consequently, the formation and behaviour of colloids has not been addressed in the performance assessments for the SFR.

Colloids were considered in the Nirex-97 performance assessment of the proposed UK cementitious ILW repository (Nirex, 1997). The calculations in this assessment also indicated that colloids are less important than complexing agents for enhancing radionuclide transport. The Nirex-97 performance assessment concluded that colloids only become significant in a scenario where no organic complexants occur in the groundwaters.

Origin in the repository system:

Colloid generation in the silo is controlled by the groundwater chemistry and temperature which controls the degradation of the cement in the shell and in the grout. In addition, colloids may be created across any pH gradient formed in the silo.

Impact on the repository system:

Colloids generated in shell and the grout within the silo may enhance radionuclide transport out to the near-field rock. However, colloids are likely to be filtered by the bentonite layer between the shell and the rock. The significance of colloidal transport is not considered to be as great as that due to complexing agents formed by the degradation of organic materials in the repository.

Bibliographic references:

Kurosawa S, Yui M and Yoshikawa H (1997) Experimental study of colloid filtration by compacted bentonite. Scientific Basis for Nuclear Waste Management, 465, 963-970.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Report No S/97/012, Four Volumes.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1994) evaluation of SKB’s in depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS), 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) and 3.2.04 (Colloids, contaminant interactions and transport with) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-3**

Name: **DEGRADATION OF THE CEMENT MORTAR AND THE SILO SHELL**

Short description:

The cementitious backfills and the concrete shell of the silo will be affected by physical and chemical degradation processes. These processes will affect the porewater chemistry, the solubility and sorption of radionuclides, and the mechanical stability of the silo shell. Together, these effects will impact on the radionuclide transport processes and rates.

Technical description:

Cementitious materials are used extensively in the silo structure of the SFR repository. The shell of the silo (and its dividing walls) are made from steel reinforced concrete while the spaces between the waste packages are backfilled with a cementitious grout.

The concrete wall of the silo acts to reinforce the flow resistance of the bentonite buffer through its low hydraulic conductivity and to provide mechanical support to the buffer and to the contained waste. The cement will also act as a chemical barrier by creating high pH porewaters in which many radionuclides are poorly soluble. The cement minerals also will sorb nuclides from the aqueous phase and thereby retard their outward transport from the repository's near-field.

The concrete used in the shell walls consists of cement and aggregate. The aggregate is normally sand or gravel. The main constituent of the cement is $Ca(OH)_2$ (portlandite), calcium-silicate-hydrate gels after cement hydration and lesser amounts of KOH and NaOH.

Groundwater resaturating the silo will react with the large volumes of cement causing its pH to rise from natural neutral values to around 12 or 13 by dissolution of the small amounts of NaOH and KOH hydroxides present in the cement. As these early high-pH waters are displaced by incoming groundwaters, the pH will be controlled by dissolution of the $Ca(OH)_2$ which will buffer the pH at around 12.5. The rate of this process is controlled by the water turnover. For the silo, turnover is very slow because of the very low hydraulic conductivity of the concrete shell and bentonite layer. Consequently, hyperalkaline conditions are likely to be maintained for thousands of years.

These conditions will strongly affect the solubility of radionuclides released from the waste (see SFR-15 "Radionuclide release from the waste"). Most nuclides (though not all) are poorly soluble under these chemical conditions. The high pH will also affect sorption of Cs and Sr during the early period when K and Na are released during dissolution of their hydroxides, although other nuclides will not be affected. The importance of the high pH conditions for controlling radionuclide solubility is critical for determining the safety of the repository. Consequently, performance assessments for the SFR pay particular attention to modelling radionuclide solubilities under these chemical conditions using thermodynamic solubility and speciation codes, and their associated databases. However, it is difficult to rely on modelling because the characteristics of the solid phases formed are poorly known and, thus, empirical solubility data are sometimes used.

In addition to buffering the porewater pH, other chemical reactions will affect the cementitious materials. The most significant processes are sulphate attack, carbonation and the pozzuolan reaction.

Sulphate attack on concrete will occur when sulphate is present in the near-field. This sulphate could be introduced by the groundwater or may be generated through chemical and radiolytic degradation of sulphonate-based ion-exchange resins. Sulphate (gypsum) is also generally included in the cement to control the setting time. Sulphate will react with the cement to form solid reaction products such as ettringite and mono-sulphate.

Carbonation occurs when dissolved CO_2 reacts with carbonate. The CO_2 will be released by microbial degradation of organic materials while the carbonate is present as impurities in the bentonite layer around the shell and may be introduced by the groundwater. Carbonation results in the precipitation of calcite.

The pozzuolan (alkali-silica) reaction occurs where the $Ca(OH)_2$ present in the cement slowly reacts with the silica in the aggregate to form calcium-silicate-hydrate secondary products. These are initially gel like but, over long periods of time, they transform into more stable crystalline forms. Brucite will also precipitate as a hydrated phase because small amounts of MgO always occur in cement and some dissolved Mg is found in the groundwaters.

Together, the formation of ettringite, gypsum, calcite and the calcium-silicate-hydrate gels in the backfill mortar will cause a reduction in its hydraulic and gas permeability, and in its mechanical strength. More significantly, ettringite formation results in expansion as the ettringite binds water into the solid phase. If large volumes of ettringite form by this process, the swelling pressure may adversely affect the mechanical integrity of the concrete shell (see SFR-13 “Mechanical impact on the engineered barriers”).

During the saline water infiltration phase, consideration should be given to the impact of chloride on the cement degradation. Chloride generally is not corrosive to concrete but can be incorporated into the calcium-silicate-hydrate gels. It is possible that a hydrated chloride compound (known as Friedel’s salt) can form. The formation of Friedel’s salt liberates sulphates (e.g. from ettringite) which may affect the porosity and physical stability of the material. The detailed behaviour of cement-chloride reactions is not well understood (Lagerblad and Trägårdh, 1994) but Friedel’s salt has been the subject of recent studies to define its stability. In addition, chloride will diffuse through the concrete and cause an increase in the corrosion rate of the steel reinforcing rods in the structural concrete parts of the repository.

The formation of solid alteration products in the pores spaces of the concrete shell may cause it to crack causing mechanical damage to the concrete, forming cracks and spallation of the surface. The consequences of this would be to increase the hydraulic conductivity (allowing greater water flow) and a reduction in mechanical strength of the shell.

The same processes may occur in the shotcrete (concrete) surfaces in the rock vaults. However, because the activity content of the rock vaults is much less than in the silo, the impact on repository safety is less significant. Furthermore, the shotcrete is not afforded any flow barrier function in the performance assessments.

Origin in the repository system:

The continued degradation of cementitious materials (cements and concretes) in the SFR repository will be dependent on the supply of groundwater into the near-field, the porewater chemistry and the degradation of the wastes (e.g. release of SO_4 from ion-exchange resins). The supply of groundwater is controlled by the hydraulic conductivity of the near-field rock and the engineered barrier materials. The porewater chemistry is largely controlled by the chemistry of the natural groundwaters and the degradation of the engineered barriers (cement and steel).

Other controls are the near-field temperature, the resaturation state of the near-field and internal and external stresses that may cause mechanical damage.

Impact on the repository system:

Chemical degradation of the cementitious materials is desirable in the sense that their sacrificial dissolution of creates high pH porewaters that reduces radionuclide solubility. This may decrease radionuclide release rates from the near-field.

However, continued chemical degradation and mechanical impacts can reduce the physical strength and increase the hydraulic conductivity of the concrete shell, thus increasing the flow of water through the near-field. This may increase radionuclide release rates.

Bibliographic references:

Lagerblad L and Trägårdh J (1994) Conceptual model for concrete long time degradation in a deep nuclear waste repository. SKB Technical Report, TR 95-21.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D, Bateman K, Hill P, Hughes C, Milodowski A, Pearce J, Rae E and Rochelle C (1992) Rate and mechanism of the reaction of silicates with cement pore fluids. Applied Clay Science, 7, 33-45.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0	30th June 2000	Matt White and Bill Miller

Number: **SFR-4**

Name: **DEGRADATION OF STEEL REINFORCEMENTS IN THE SILO SHELL**

Short description:

The shell of the silo is made of reinforced concrete. Degradation of the reinforcing rods may occur by a combination of physical and chemical processes leading to gas generation, colloid generation and possible impacts on the structural integrity of the silo.

Technical description:

The silo in the SFR is constructed from a reinforced concrete shell, 90 cm thick, with a layer of bentonite, approximately 1.3 m thick, filling the void space between the shell and the rock. The reinforced concrete shell provides the primary strength and stability for the silo complex. The reinforcement rods are made of steel.

The very large volumes of cement used in the construction of the shell and in the grout used to backfill spaces between waste packages in the silo will buffer the pH values in the SFR to around 12.5 within decades of closure and, over thousands of years, will gradually decrease to around 10 as the $Ca(OH)_2$ is leached away.

Anaerobic steel degradation in alkaline environments proceeds at a much slower rate than in pH neutral conditions (Grauer, 1988). This is because the iron (steel) corrosion product (composition similar to magnetite) is very stable at high pH and, consequently, forms a passivating layer on the metal surface. However, embedded in the cement, the formation of magnetite on the reinforcing rods requires a supply of oxygen through the cement matrix. Consequently, the formation of this passivating layer is partly controlled by the permeability of the cement that forms the silo shell. In the absence of a magnetite passivating layer, the rate at which the steel will corrode depends on a number of factors including the pH of the pore solution, the availability of water, the electrical resistivity of the concrete, the ability of the corrosion products to migrate away from the steel, and the partial pressure of H_2.

Anaerobic degradation of steel material used as reinforcement will help to buffer the redox conditions in the near-field and will result in hydrogen generation and formation of solid corrosion products. The latter could potentially cause blocking of pores and fractures in the cement and backfill materials adjacent to the steel.

Rapid gas production (see SFR-11 “Gas generation in the repository”) or a volume increase of the reinforcing rods caused by the solid alteration products could cause mechanical damage to the concrete, forming cracks and spallation of the surface. The consequences of this would be to increase the hydraulic conductivity (allowing greater water flow) and a reduction in mechanical strength of the shell (see SFR-13 “Mechanical impact on the engineered barriers”).

Solid degradation products from both the cement and steel will provide efficient sorption sites for any radionuclides migrating through the bentonite buffer, although they are unlikely to be very abundant. However, these solid alteration products may also increase the colloid population in the near-field porewater.

Physical damage to the silo is also likely to be caused by stresses internal and external to the silo. Internal stresses could be caused by a gas overpressure (arising if the gas vents in the top of the silo become blocked) or, less likely, from swelling of the solid waste materials, such as dehydrated ion-exchange resins immobilised in bitumen. Short-term external stresses on the silo shell are most likely to arise during the immediate post-closure period as the rock mass deforms in response to stress readjustments (see GEN-36 “Stress field”) and due to hydraulic resaturation (see SFR-16 “Hydraulic resaturation of the near-field”). Over the longer term, external stresses will result from isostatic uplift of the land mass (see GEN-39 “Uplift and subsidence”).

Both the internal and external stresses, together with chemical degradation of the silo concrete, will eventually result in failure of the silo structure. This will, in turn, result in mechanical damage to the reinforcing rods. However, SKI estimates of the silo behaviour (SKI, 1994), are that structural integrity should be maintained for at least 1000 years after closure, with the chemical buffering role lasting considerably longer than that.

Origin in the repository system:

Degradation (corrosion) of the steel reinforcing rods in the concrete shell of the silo will occur under anaerobic, highly alkaline conditions. The rate of corrosion will largely be affected by the mechanical properties (strength, water and gas permeabilities) of the silo concrete, the resaturation state of the repository, the pore water chemistry and the temperature.

Physical damage to the reinforcing rods will occur as the concrete silo shell is subject to internal and external stresses. The most significant impact is likely to come from gas overpressures within the silo resulting from hydrogen gas evolved during anaerobic corrosion of the steel waste packages.

Impact on the repository system:

Degradation (corrosion) of the steel reinforcing rods will impact directly on the physical strength and integrity of the silo shell, and may cause fracturing and spallation of the concrete. If the silo fractures through this process, this will impact on the water and gas permeabilities of the shell and, thus, the gas pressure within the silo.

Corrosion of the reinforcing rods will generate hydrogen gas and will also impact on the pore-water chemistry, buffering the local redox conditions. However, these impacts will be eclipsed by the corrosion of the much larger volumes of steel in the waste packages.

Bibliographic references:

Grauer (1988) The corrosion behaviour of carbon steel in Portland cement. Nagra Technical Report, NTB 88-02.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-5**

Name: DEGRADATION OF THE CONCRETE PACKAGES AND THE CEMENT MATRIX

Short description:

The concrete waste packages and the cementitious immobilisation matrix will be affected by physical and chemical degradation processes. These processes will affect the porewater chemistry, the solubility and sorption of radionuclides, and the mechanical stability of the packages. Together, these effects will impact on the radionuclide transport processes and rates.

Technical description:

In the SFR repository, concrete will be used to manufacture certain waste packages and overpacks placed in the silo, the BMA rock vault as well as the concrete tanks in the BTF rock vault. In addition, cement will be used as an immobilisation matrix for several of the waste types (such as ion-exchange resins and sludges).

The mechanical stability of the concrete waste packages is only of marginal importance for the long-term safety of the repository. In most cases, no special requirements are made for post-closure longevity or water-tightness of the packages, thus resistance against corrosion is not an essential design property. In performance assessments for the SFR, no credit is assigned for any mechanical lifetime of the waste packages (SKI, 1994).

However, the chemical buffering capacity provided by the cementitious materials in the near-field (waste packages, cement immobilisation matrix together with the grout in the silo) is of primary importance to repository safety. This is because the cement will act as a chemical barrier by creating high pH porewaters in which many radionuclides are poorly soluble. The cement minerals also will sorb nuclides from the aqueous phase and thereby retard their outward transport from the repository's near-field (see SFR-18 “Radionuclide release from the waste package”).

The most significant constituent of the cement is $Ca(OH)_2$ and the calcium-silicate-hydrate gels formed after cement hydration, together with lesser amounts of KOH and NaOH. Groundwater contacting the waste packages will react with the cement and will begin to dissolve the Na, K and Ca hydroxides, causing the pH of the porewater to rise. In the silo, the greatest pH buffering effect will be from the very large volumes of porous, cementitious grout used to backfill between the waste packages, not from the packages themselves. However, in the rock vaults, where no cement backfill will be used, the pH buffering capacity is largely controlled by dissolution of the concrete waste packages and the immobilisation matrix.

Because the silo has much larger volumes of cement and a slower water turnover than the rock vaults, the silo porewater will reach a higher pH and maintain it for longer. These conditions will strongly affect the solubility of radionuclides released from the waste. The high pH will also affect sorption of Cs and S during the early period when K and Na are released during dissolution of their hydroxides, although other nuclides will not be affected. Consequently, the pH control on radionuclide solubility and sorption is most important than in the silo where the highest activity waste is located.

Performance assessments for the SFR thus pay particular attention to modelling radionuclide solubilities under these chemical conditions using thermodynamic solubility and speciation codes and their associated databases.

In addition to buffering the porewater pH, other chemical reactions will affect the cement waste package and immobilisation matrix. The most significant processes are sulphate attack, carbonation and the pozzuolanic reaction.

Sulphate attack on concrete will occur when sulphate is present in the near-field. This sulphate could be introduced by the groundwater or may be generated through chemical and radiolytic degradation of sulphonate-based ion-exchange resins. Sulphate (gypsum) is also generally included in the cement to control the setting time. Sulphate will react with the cement to form solid reaction products such as ettringite.

Carbonation occurs when dissolved CO_2 reacts with $Ca(OH)_2$ or other hydrated phases of cement. The CO_2 will be released by microbial degradation of organic materials while the carbonate is present as impurities in the bentonite layer around the shell and may be introduced by the groundwater. Carbonation results in the precipitation of calcite.

The pozzuolan (alkali-silica) reaction occurs where the $Ca(OH)_2$ present in the cement slowly reacts with the silica in the aggregate to form calcium-silicate-hydrate secondary products. These are initially gel like but, over long periods of time, they transform into more stable crystalline forms. Brucite will also precipitate as a hydrated phase because small amounts of MgO always occur in cement and some dissolved Mg is found in the groundwaters.

Together, the formation of ettringite, gypsum, calcite and the calcium-silicate-hydrate gels in the cement waste package and immobilisation matrix may infill pore spaces in the cement. More significantly, ettringite formation results in expansion as the ettringite binds water into the solid phase. If large volumes of ettringite form by this process, the swelling pressure may causing mechanical damage to the concrete. The consequences of this would be to increase the water flow through the waste package and to increase the contact area between the water and the waste. This may result in more rapid degradation and an increase in the rate of formation of gaseous reaction products.

Origin in the repository system:

The degradation of concrete waste packages and cementitious immobilisation matrixes in the SFR repository will be dependent on the supply of groundwater into the near-field and the porewater chemistry. The supply of groundwater is controlled by the hydraulic conductivity of the near-field rock and the engineered barrier materials. The porewater chemistry is largely controlled by the chemistry of the natural groundwaters and the degradation of the engineered barriers (cement and steel).

Other controls are the near-field temperature and the resaturation state of the near-field.

Impact on the repository system:

Chemical degradation of the waste packages and immobilisation matrixes will help to buffer the pH of the near-field porewaters. In the silo, this buffering capacity will be less significant than that provided by the larger volumes of cementitious grout. However, in the rock vaults, the degradation of the waste packages and immobilisation matrixes provides the dominant pH buffering mechanism.

The development of high pH porewaters will reduce radionuclide solubility and thus may decrease radionuclide release rates from the near-field.

Continued degradation of the waste packages can increase the flow of water through them. This will increase the waste degradation rate and thus the rate of gas production.

Bibliographic references:

Lagerblad L and Trägårdh J (1994) Conceptual model for concrete long time degradation in a deep nuclear waste repository. SKB Technical Report, TR 95-21.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-6**

Name: **DEGRADATION OF THE BITUMEN MATRIX**

Short description:

Bitumen is used as an immobilisation matrix for certain waste types. The bitumen will be subject to degradation by physical, chemical and microbiological processes. Organic and colloidal degradation products of bitumen may impact on radionuclide transport and retardation processes in the near-field.

Technical description:

Bitumen will be used as an immobilisation matrix for certain wastes types. Some dehydrated and powdered ion-exchange resins have been immobilised in bitumen. Other reactor wastes, such as filter aids from reactor clean-up systems, are also immobilised in bitumen. The bitumenised wasteforms are contained in steel drums or boxes and emplaced either in the silo or the BMA rock vault sections of the SFR repository.

The technological bitumen used in the SFR is derived from natural organic materials. It consists of mixtures of mainly aliphatic and aromatic hydrocarbons of high molecular weight obtained from the heaviest petroleum fractions.

In general, bitumen is chemically unreactive with natural groundwaters, however it can be degraded by sulphuric acid which attacks its aromatic constituents. There is, thus, a small possibility that bitumenised waste could be attacked by acidic solutions formed, for example, by the dissolution of sulphide minerals in the rock by oxidising waters shortly after closure of the repository. However, the large pH buffering capacity offered by the cement in the repository (see SFR-20 “Groundwater chemistry in the near-field”) will rapidly neutralise any acids formed this way and, consequently, this degradation process is of little concern.

Under the highly alkaline environment characteristic of the SFR repository, the bitumen is generally stable, although it could be subject to microbially-mediated degradation (Zobell and Molecke, 1978). Bitumen, in common with most organic compounds, is susceptible to many micro-organisms which have the ability to utilise hydrocarbons as sources of energy and nutrients. It is generally accepted, however, that the rate of biodegradation of bitumen is highly dependent on the chemical nature of the hydrocarbons present, on the microbial community and on environmental factors that influence microbial activities.

Bitumen degradation can be enhanced in the presence of a high radiation field. However, given that the majority of the bitumen is used to encapsulate low activity wastes, this is not of great concern. The exception to this could be some older packages of dried ion-exchange resins which were immobilised in bitumen and are already in the silo. The radiation field in the silo is much greater than in the BMA and, consequently, the potential for radiation enhanced bitumen degradation is also much higher. However, the actual increase in degradation rate by this process is considered still to be negligible.

The degradation products of bitumen might comprise organic complexants which have the potential for enhancing radionuclide solubility, particularly of the polyvalent cations (plutonium, americium, technetium, niobium). However, the formation of complexing agents by the biodegradation of the cellulose (and, to a lesser extent, bakelite, PVC and polyethylene) in the waste is considerably more significant (SKI, 1994). Because there is a much greater amount of organic material in the BMA rock vault than in the silo, the importance of complexing agents is also more significant for radionuclide transport in the BMA.

Degradation of bitumen may also generate colloids (see SFR-1 “Colloid generation in the waste package”). The significance of this colloidal formation is uncertain but not thought to be large.

Microbially-mediated degradation of bitumen will also generate CO_2, H_2 and CH_4 gases (see SFR-11 “Gas generation in the repository”). However, the volume of gases generated by microbial degradation will be significantly less than the volumes of gas generated by anaerobic corrosion of steel and microbially-mediated generation of cellulose waste.

Given that neither the production of complexing agents or gas is a primary cause for concern, the chemical degradation of bitumen has not been a priority issue in performance assessments for the SFR.

Physical degradation of the bitumen is a more significant process. The older packages of dried ion-exchange resins which were immobilised in bitumen have the potential to swell as the powdered resin absorbs water. Initial designs for the SFR called for large numbers of this type of waste package to be placed in the silo and, consequently, there was some concern that their swelling would cause cracking of the silo walls. However, the adoption of new immobilisation methods means that the volume of ion-exchange resins placed in bitumen is considerably less than originally planned. While this bitumen swelling issue is not now as significant as it once was, the post-closure behaviour of the bitumen packages already manufactured needs to be considered carefully in performance assessment.

Origin in the repository system:

Degradation of most of the bitumenised waste will occur under anaerobic, highly alkaline conditions. The degradation rate of corrosion will largely be affected by the resaturation state of the repository, the pore water chemistry, microbiological populations, the radiation field and the temperature.

Physical damage to the bitumen wasteform may occur as dried ion-exchange resins mixed into the bitumen absorb water and swell.

Impact on the repository system:

Physical and chemical degradation of the bitumen wasteform will impact directly on the water flow in the waste package and, consequently, on the transport and release of nuclides from waste packages. Degradation of the bitumen will also impact on the pore-water chemistry, gas generation and pressure and colloid populations. The most significant effect will be the production of organic complexants which may increase radionuclide solubility. However, these impacts will be eclipsed by the degradation of the much larger volumes of cellulose in the waste.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Zobell CE and Molecke MA (1978) Survey of microbial degradation of asphalts with notes on relationships to nuclear waste management. Sandia Laboratories Technical Report, SAND-78-1371.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS), 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-7**

Name: **DEGRADATION OF THE INORGANIC WASTE**

Short description:

The SFR repository will house many diverse waste types, both inorganic and organic. The inorganic wastes may degrade by a combination of physical, chemical and microbial processes. This degradation may have a limited impact on the release and transport of radionuclides from the near-field.

Technical description:

The SFR repository will contain numerous types of inorganic wastes which are generated mostly from reactor operations but with contributions from research, industry and health care activities.

The nature of this waste is variable. Solid inorganic wastes include, among other things, insulating material (mineral wool); steel and other scrap metals from reactor refuelling and maintenance operations; spent radioisotope sources; spent air filters and cleaning materials.

Some of this material (such as metallic objects) is directly encapsulated in cement in the waste packages. However, the low activity combustible material is incinerated and the resulting ash is immobilised in cement to reduce waste volumes. Depending on the activity of the inorganic waste, it is emplaced either in the silo (higher activity waste) or in one of the rock vaults (lower activity waste).

In the repository these inorganic materials will be generally unreactive and will degrade at a slow rate. Initially, all of the material immobilised in cement will largely be protected against degradation by the immobilisation matrix, with only limited surface area available for reaction. However, as the cement matrix degrades, larger surface areas may be exposed.

The metal components of the inorganic waste will undergo anaerobic corrosion, releasing H_2. However, this processes will proceed slowly because the metal oxide corrosion products are generally stable at high pH and, consequently, will form a passivating layer on the metal surface. The consequence of this gas production will be minimal because much greater volumes of gas will be generated by corrosion of the steel waste packages in the silo or by microbially-mediated degradation of organic materials in the BLA and BMA rock vaults (see SFR-11 “Gas generation in the repository”).

Any non-metallic inorganic materials (e.g. ceramics) will be present in the waste in only small amounts and will be essentially inert, although they may be subject to slow microbially-mediated degradation products. In this case, small volumes of CO_2, CH_4 and H_2 gases could be generated but they would be much smaller than the equivalent gas production from degradation of organic materials, such as cellulose.

In addition to gas production, the degradation of inorganic materials is likely to generate colloids (see SFR-1 “Colloid generation in the waste package”). In some cases, radionuclides could be associated with the colloids by sorption onto the colloid surfaces or they may be directly included in the colloidal material if the colloid is a small fragment of the radionuclide-bearing inorganic wasteform itself. However, much larger populations of colloids are likely to be generated from the massive volumes of cement in the repository.

Although inorganic materials in the waste can degrade, they are likely to do so slowly and with minimal impacts on the transport and release of radionuclides from the near-field. Consequently, degradation of inorganic waste has not been addressed in detail in the performance assessments for the SFR.

Origin in the repository system:

Degradation of inorganic materials in the waste will occur under anaerobic, highly alkaline conditions. The rate of degradation will largely be affected by the mechanical properties of the cement immobilisation matrix, the resaturation state of the repository, the pore water chemistry, populations of microorganisms and the temperature. However, degradation rates are expected to be slow.

Impact on the repository system:

Degradation of inorganic materials in the waste will impact directly on the transport and release of radionuclides, although the impact will be small in relation to the effects of other processes. Degradation of these materials will generate gas and colloids and this may impact on the pore-water chemistry. However, gas and colloid generation rates from inorganic wastes will be eclipsed by the degradation of the much larger volumes of steel forming the waste packages and the organic wastes.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEP 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-8**

Name: **DEGRADATION OF THE ORGANIC WASTE**

Short description:

The SFR repository will house many diverse waste types, both inorganic and organic. The organic wastes may degrade by a combination of physical, chemical and microbial processes. This degradation is likely to create large volumes of gas and a variety of organic compounds which may influence radionuclide solubilities. Consequently, degradation of organic wastes may impact on the release and transport of radionuclides from the near-field.

Technical description:

The SFR repository will contain numerous types of organic wastes which are generated mostly from reactor operations but with contributions from research, industry and health care activities.

The nature of this waste is variable. Organic wastes include, among other things, ion-exchange resins and sludges; filter pulps; halogenated and non-halogenated rubbers; halogenated and non-halogenated plastics; disposal clothing and cleaning materials. Some of this material is directly encapsulated in cement in the waste packages. However, the low activity combustible material is incinerated and the resulting ash is immobilised in cement to reduce waste volumes. Incineration will destroy most, but not all, of the organic constituents in the material. Depending on the activity of the inorganic waste and ash, it is emplaced either in the silo (higher activity waste) or in one of the rock vaults (lower activity waste).

The degradation of this organic material has two significant impacts on the performance of the SFR repository. Firstly, their degradation can generate large volumes of gas either by microbially-mediated alteration reactions or by direct volatilisation (see SFR-11 "Gas generation in the repository"). Secondly, their degradation may produce colloids and organic complexants that may act to increase radionuclide solubility (see SFR-1 "Colloid generation in the waste package").

Concerning gas production, the material of greatest concern is cellulose (a polysaccharide) which is readily biodegradable. Cellulose may occur in the waste in the form of wood and paper etc., although materials such as these are likely to have been incinerated prior to emplacement in the SFR. Microbial degradation of cellulose creates CO_2, CH_4 and H_2 gases and the production of these gases is likely to be greatest in the BMA and BLA rock vaults which contain the greatest content of cellulose. This rate of gas generation may be rapid and could result in the formation of a separate gas phase if gas production exceeds the rate at which gas can escape to the near-field and gas concentrations eventually exceed solubility limits (see SFR-12 "Gas flow in the near-field").

The silo contains negligible quantities of cellulose in the waste. However, it is noted that cellulose may be used as an additive (to prevent cement-water separation) in the grout used to backfill between the waste packages in the silo. In the silo, assuming negligible cellulose contents, the dominant gas production process is anaerobic corrosion of steel generating H_2.

Gas production from other organic materials is not considered to be as significant as from cellulose. The majority of the organic ion-exchange resins will be immobilised in cement, limiting the surface area for microbial attack. Furthermore, ion-exchange resins are generally resistant to chemical degradation, designed to be inert in laboratory and industrial usage.

Gas would also be generated by the microbial degradation of bitumen. However, the rate of biodegradation of bitumen is much slower than for cellulose and the volume of bitumen is much less, meaning that gas generation from bitumen is less significant.

The production of gas through the degradation of organic matter is of concern because the formation of a gas overpressure could cause physical damage to the engineered barriers or to the near-field rock (see SFR-13 “Mechanical impact on the engineered barriers”). Furthermore, a mobile gas phase leaving the near-field, may also impact on radionuclide transport by expelling contaminated water.

A second concern of gas production is that the gas itself could be radioactive if it comprises ^{3}H or ^{14}C derived directly from the waste material. Rapid transport of radioactive gases generated by degradation of organic material is one of the primary issues for the safety of the SFR repository (see SFR-15 “Radionuclide release from the waste”).

Concerning the production of organic complexants via the degradation of organic materials, again it is cellulose that is of primary concern due to its biodegradability. Indeed, analysis of the impact of complexing agents (SKI, 1994; Brownsword et al., 1992) suggest that it is only cellulose which, through its degradation into complexing agents, appears to affect the rate at which radionuclides can be transported out of the near-field. Again it should be noted that cellulose may be used as an additive in cements and grouts in the SFR.

Other organic substances in the waste which can generate complexants are bakelite, PVC and polyethylene (Brownsword et al., 1992). However, the biodegradability and volumes of these materials are significantly less than for cellulose, making them less efficient producers of complexing agents.

Complexants formed by degradation of organic wastes can potentially have a significant impact on the solubility of radionuclides. For example, cellulose degradation can create substances capable of increasing the solubility of plutonium by up to 6 orders of magnitude, even at high pH levels. The actual complexing agents formed will depend significantly on the nature of the wasteform and the chemistry of the porewaters but, generally, they tend to be polyhydroycarboxyl acids.

The impact in performance assessment of including assumptions for the creation and behaviour of degradation products of cellulose is very significant and can result in the estimates of individual doses to be increased by a factor of ten as opposed to scenarios without complex formation (SKI, 1994).

Because the significance of gas and complexant formation from organic material degradation is potentially so great, performance assessments for the SFR have paid a great deal of attention to these processes.

Origin in the repository system:

Degradation of organic wastes will occur under anaerobic, highly alkaline conditions. Most degradation will be microbially-mediated and the rate of this degradation will largely be affected by the available surface area, populations of microorganisms, availability of nutrients, the temperature and, above all, the degradability of the material. Cellulose is the material most affected.

Impact on the repository system:

Physical and chemical degradation of organic wastes will impact directly on the transport and release of nuclides from waste packages by increasing radionuclide solubilities via the formation of complexing agents.

In addition, microbially-mediated degradation of organic materials will generate large volumes of gas that may form a separate gas phase. This free gas can affect transport of radionuclides in solution by expelling contaminated water. The nuclides ^{3}H and ^{14}C may be released directly from the waste in gaseous form.

Bibliographic references:

Brownsword M, Gaudies SC, Greenfield BF, Robertson GP, Spindler MW and Woodwark DR (1992) The effect of organic degradation products on the sorption of actinides on cement matrices. UK Nirex Safety Studies Report, NSS/R203.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-9**

Name: **DIFFUSION IN THE NEAR-FIELD**

Short description:

Diffusion in the near-field may occur through water-filled cracks and voids in the mass of the immobilisation matrices and through the compacted bentonite layer surrounding the silo structure. Diffusion will be an important process controlling porewater chemistry in the near-field and the radionuclide release rate to the far-field.

Technical description:

Diffusion theory is adequately defined and discussed in the scientific literature (e.g. Crank, 1975). In terms of understanding diffusion in the repository environment, a simple but adequate definition is that diffusion is the net flux of solutes from a zone of higher concentration to a zone of lower concentration, that does not depend on any bulk movement of the solution (Spitz and Moreno, 1996).

In the SFR repository, diffusion may be an important process for the release of radionuclides in two cases.

In the first case, radionuclides may escape from the waste by diffusing through cracks and void spaces in the bulk mass of the immobilisation matrix, which is either cement or bitumen. It is possible that the immobilisation matrices will suffer post-emplacement cracking in response to interactions with the porewaters and because of differential thermal expansion due to heat generated by exothermic cement curing (see SFR-17 “Temperature of near-field”). Porewaters will fill these cracks and voids but is unlikely to be mobile. Consequently, radionuclides released to this porewater will travel through the water-filled cracks by diffusion, in a manner analogous to matrix diffusion in crystalline rock (see GEN-25 “Matrix diffusion”). Diffusion of this nature may take place in the waste emplaced in both the silo and the rock vaults.

In the second case, diffusion could take place through the bentonite layer surrounding the reinforced concrete silo structure. The bentonite layer at the sides of the silo is compacted and approximately 1.3 m thick. At the top and bottom of the silo, a sand/bentonite mixture (90% sand) is used as the buffer material to provide greater bearing strength (at the bottom) and gas permeability (at the top).

Due to the high degree of compaction of this bentonite layer, no water will flow through it. Consequently, dissolved radionuclides can exit through the silo walls only by diffusion.

However, the bentonite will slowly degrade by a variety of chemical and physical processes (see SFR-24 “Evolution of the bentonite layer in the silo”), such as reaction with the hyperalkaline leachates derived from the cementitious materials and from internal and external stresses. These processes will act with time to increase the hydraulic conductivity of the bentonite layer, most likely by forming a number of fast pathways through the bentonite.

Eventually, continued chemical and physical degradation of the bentonite will cause it to fail entirely as a barrier to advective flow. At this point, the diffusive barrier function will also be lost. However, SKI estimates of the silo behaviour (SKI, 1994), are that structural integrity should be maintained for at least 1000 years after closure.

Transport and release from the silo is a primary control on the safety of the SFR repository (see SFR-19 “Transport and release from the silo”). Therefore, performance assessments for the SFR have paid detailed attention to diffusion through the silo walls and the bentonite layer.

Origin in the repository system:

The release of radionuclides from the wasteform can occur by diffusion in the water filled cracks and voids in the immobilisation matrix. Cracking of the immobilisation matrix is controlled by the thermal field and the chemistry of the porewater.

Radionuclides dissolved in the silo porewaters can diffuse through the silo walls at a rate controlled by the physical characteristics of the bentonite layer; the chemistry of the porewater (ionic strength and chemical gradient of the migrating species); release rates for the migrating species from the wasteform; and the groundwater flow rate in the advecting part of the system. As the silo degrades, the permeability of the bentonite layer will increase, eventually allowing direct flow to occur across the walls.

Impact on the repository system:

Transport and release from the silo is a primary control on the safety of the SFR repository. The diffusion of radionuclides through the silo wall affects the porewater compositions inside and outside the silo and, thus, sorption-desorption and precipitation-dissolution reactions.

Bibliographic references:

Crank J (1975) The mathematics of diffusion (2nd edition). Oxford University Press.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Spitz K and Moreno J (1996) A practical guide to groundwater and solute transport modelling. John Wiley and Sons Ltd.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.6 (Diffusion - surface diffusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-10**

Name: **COLLOID FILTRATION IN THE NEAR-FIELD**

Short description:

Colloids formed in the silo of the SFR will be filtered by the bentonite layer around the silo structure for as long as the bentonite provides a barrier to advective groundwater flow. This will cause a reduction in the release of radionuclides to the near-field rock. No other material in the SFR provides a significant colloid filtration role.

Technical description:

Some of the radionuclides released from the waste packages are likely to be associated with colloids. Colloids are small, solid particulate materials suspended and dispersed in the groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed ‘suspended particles’ and anything smaller is considered to be in solution.

The colloids will be generated by the degradation of the wasteform and the materials used in the engineered barriers (see SFR-1 “Colloid generation in the waste package” and SFR-2 “Colloid generation in the shell and grout”). Radionuclides could be associated with the colloids by sorption onto the colloid surfaces or they may be directly included in the colloidal material if the colloid is a small fragment of the radionuclide-bearing wasteform itself. In addition, colloids may be formed by direct precipitation within the waste package as a result of the thermal and chemical gradients that may occur there.

Of the various materials in the waste packages, the most significant for the production of colloids would be the cement. This is partly because cement is the most abundant material in the near-field (by volume) but also because cement will generate colloids as it matures and degrades.

The transport and retardation of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution. In particular, the colloids in the repository may have their transport to the near-field rock retarded by filtration processes in low permeability buffer and backfill materials with respect to nuclides in solution.

In the silo, the reinforced concrete shell is surrounded by a layer of bentonite between the shell and the rock. The bentonite layer at the sides of the silo is compacted and approximately 1.3 m thick. At the top and bottom of the silo, a sand/bentonite mixture (90% sand) is used as the buffer material to provide greater bearing strength (at the bottom) and gas permeability (at the top).

Due to the high degree of compaction of this bentonite layer, no water will flow through it. Consequently, colloids formed within the silo will be essentially immobilised by filtration processes and any radionuclides sorbed onto them will be retarded provided the bentonite layer remains intact (Kurosawa et al., 1997).

However, the bentonite will slowly degrade by a variety of chemical and physical processes (see SFR-24 "Evolution of the bentonite layer in the silo"), such as reaction with the hyperalkaline leachates derived from the cementitious materials and from internal and external stresses. These processes will act with time to increase the hydraulic conductivity of the bentonite layer, most likely by forming a number of fast pathways through the bentonite. Eventually, continued chemical and physical degradation of the bentonite will cause it to fail entirely as a barrier to advective flow. At this point, the colloidal filtration capacity will also be lost. However, SKI estimates of the silo behaviour (SKI, 1994), are that structural integrity should be maintained for at least 1000 years after closure.

The bentonite layer in the silo provides the most significant colloid filter in the SFR repository. In the rock vaults, where there is no bentonite layer, no significant colloid filtration will occur, although the shotcrete coated walls will provide a short-lived barrier to groundwater and colloid movement.

However, colloids are not considered important for the safety of the SFR repository and colloid transport and colloid filtration were not specifically addressed in the performance assessments for the SFR. Other mechanisms such as increased solubilities provided by organic complexing agents and the barrier to groundwater flow provided by the bentonite layer in the silo are considered more critical.

However, colloids were considered in the Nirex-97 performance assessment of the proposed UK cementitious ILW repository (Nirex, 1997). The calculations in this assessment also indicated that colloids are less important than complexing agents for enhancing radionuclide transport. The Nirex-97 performance assessment concluded that colloids only become significant in a scenario where no organic complexants occur in the groundwaters.

Origin in the repository system:

Filtration of colloids in the near-field of the SFR repository is provided by the bentonite later around the silo structure. The ability of this layer to filter colloids is limited by the physical properties of the bentonite, particularly its permeability which is controlled by the degree of compaction.

The filtration capacity of the bentonite layer will reduce over time as chemical and physical processes degrade the bentonite and, eventually, will allow water to flow through it.

No other materials in the SFR act as a significant colloid filter.

Impact on the repository system:

The colloid filtration capacity offered by the bentonite layer around the silo will cause a reduction in the release of radionuclides to the near-field rock. This will have an impact on the porewater compositions inside and outside the silo. However, the filtration of colloids is not as significant for controlling radioactive releases as other processes. Notably, the barrier to groundwater flow offered by the bentonite layer. Consequently, colloid filtration is not considered in performance assessments for the SFR.

Bibliographic references:

Kurosawa S, Yui M and Yoshikawa H (1997) Experimental study of colloid filtration by compacted bentonite. Scientific Basis for Nuclear Waste Management, 465, 963-970.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Report No S/97/012, Four Volumes.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) evaluation of SKB's in depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.4 (Colloid generation - source) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-11**

Name: **GAS GENERATION IN THE REPOSITORY**

Short description:

Gases can be generated in the repository by three processes: corrosion of steel, radiolysis and microbial activity. The gas generated may form a free gas phase and this could impact on the transport and release of radionuclides. In some cases, radionuclides may be directly associated with the gas molecules whilst, in other cases, the gas phase will impact on the movement of groundwaters containing dissolved radionuclides.

Technical description:

Gases may be generated in the SFR repository, in the silo and in the rock vaults, shortly after closure, when conditions remain oxidising, and may continue to be generated in the longer term when conditions are chemically reducing.

Shortly after closure of the SFR repository, when free oxygen is available (oxidising environment), this free oxygen will be consumed through the aerobic corrosion of steel and aerobic microbial degradation of organic materials. The latter process will generate CO_2, equivalent in volume to the volume of oxygen consumed, together with some inert gases, mainly H_2 and methane (CH_4). Aerobic microbial activity will occur predominantly in the rock vaults because these contain higher contents of organic material in the waste, compared to the silo.

This microbial action, together with aerobic steel corrosion, will affect the chemical environment in the repository. The generation of CO_2 could acidify the groundwaters, while the aerobic steel corrosion will rapidly consume any free oxygen. Anaerobic conditions will then be established and should be maintained due to the low oxygen content of the groundwaters.

After anaerobic conditions have been established, gases will be produced by three primary processes: microbially-induced anaerobic degradation of organic materials, anaerobic corrosion of steel and radiolysis of water and organic material.

Microbially-induced anaerobic degradation of organic materials is one of the most significant gas generating mechanisms for the SFR because large volumes of organic material will be emplaced in the rock vaults and, to a lesser extent, in the silo. The volume of gas generated by this process is controlled by the accessibility of the organic material, availability of nutrients, the temperature and, most importantly, the biodegradability of the material.

The most easily biodegradable material in the SFR is cellulose which may occur in the waste in the form of wood and paper etc., although materials such as these are likely to have been incinerated prior to emplacement in the SFR. Microbial degradation of cellulose creates CO_2, CH_4 and H_2 gases and the production of these gases is therefore likely to be greatest in the BMA and BLA rock vaults, as these contain the greatest content of cellulose. The rate of gas generation may be rapid and could result in the formation of a gas phase if gas production exceeds the rate at which gas can escape to the near-field and if gas concentrations exceed solubility limits (see SFR-12 “Gas flow in the near-field”).

The silo contains negligible quantities of cellulose in the waste and hence microbially generated gas production rates will be low. However, it is noted that cellulose may be used as an additive to prevent cement-water separation in the grout used to backfill between the waste packages in the silo.

Gas production from other organic materials is not considered to be as significant as from cellulose. The majority of the organic ion-exchange resins will be immobilised in cement, limiting the surface area for microbial attack. Furthermore, ion-exchange resins are generally resistant to chemical degradation. They are designed to be inert in laboratory and industrial usage.

Gas would also be generated by the microbial degradation of bitumen used as an immobilisation matrix (see SFR-6 “Degradation of the bitumen matrix”). However, the rate of biodegradation of bitumen is much slower than for cellulose and the volume of bitumen is much less. Gas generation from bitumen is therefore expected to be less significant.

Anaerobic corrosion of steel will affect the steel waste packages (drums etc.), steel reinforcing rods, and steel (and other ferrous metals) in the waste itself (e.g. scrap metals from reactor refuelling and maintenance operations). The steel will react directly with water to produce hydrogen:

$3Fe^{\circ} + 4H_2O\ (l) = Fe_3O_4\ (s) + 4H_2\ (aq)$

The rate of this reaction will be controlled by the accessibility of metal surfaces and the chemical conditions. Steel waste packages will corrode first and most rapidly because the steel in the waste and in concrete reinforcing rods will be partly shielded from the porewaters. However, over time as the cement immobilisation matrix and the cementitious reinforcements (and the concrete shell of the silo) degrades, these steel components will increasingly become available for reaction.

Anaerobic steel corrosion in alkaline environments proceeds at a much slower rate than in pH neutral conditions (Grauer, 1988). This is because the iron (steel) corrosion product (magnetite) is very stable at high pH and, consequently, forms a passivating layer on the metal surface. SKB estimates the steel corrosion rate in the SFR to be between 1 and 10 µm/yr, with 3 µm/yr taken as the reference value (SKI, 1988). This is probably a conservative estimate and corrosion in the silo will therefore probably be slower than corrosion of steel in the rock vaults.

However, anaerobic corrosion of iron is catalysed by carbonic acid (H_2CO_3) which also means that the hydrogen gas production rate increases with the partial pressure of carbon dioxide. Since the anaerobic degradation of organic material generates CO_2, there remains the possibility that corrosion of the steel in the repository could be accelerated. However, the actual significance of the linkage between organic material degradation and the corrosion of steel remains uncertain because the CO_2 may be consumed by other processes (e.g. by the presence of hydroxide in the system) or because the carbonic acid concentration will be minimised by the high pH of the system.

Hydrogen gas production by anaerobic steel corrosion will be the dominant gas production mechanism in the silo where there is limited quantities of organic material to generate gas by microbial action.

Gas will also be produced in the silo in significant quantities by radiolysis (see GEN-30 “Radiolysis”). Ionising radiation from the higher activity wastes will interact with water molecules and organic materials in the waste (e.g. cellulose) to produce radicals and molecular products such as H_2O_2, H_2 and O_2. Radiolysis can begin as soon as ionising radiation can impact the water molecules and, thus, may take place inside a waste package before emplacement if water is present in pore spaces in the cement immobilisation matrix. Depending on the porewater composition, this could lead to the formation of corrosive gases such as nitrous and nitric acids and ammonia which may accelerate degradation of the cement.

After emplacement, radiolysis of the groundwater outside the package potentially can occur if ionising radiation can penetrate through the entire thickness of the package. However, as long as the waste package remains intact, the amount of radiolysis by this process will be very small because only gamma radiation has significant penetration capability and the abundance of gamma emitting wastes in the SFR will be small.

The most significant radiolytic processes will be alpha radiolysis and this can only occur after the waste package has been breached and groundwater can contact the waste materials directly. Most species produced by alpha radiolysis are extremely reactive or unstable, and quickly convert to more stable or inert species. Radiolysis leads to the production of equal amounts of oxidising and reducing species. Hydrogen, being least reactive, will accumulate over time.

Gas production by any process will be significant if a free gas phase is produced. This could occur because the gas production rate may be high and could exceed the rate at which gas can escape to the near-field. The formation of a free gas phase would be of concern for repository safety because the formation of a gas overpressure could cause physical damage to the engineered barriers or to the near-field rock (see SFR-13 “Mechanical impact on the engineered barriers”). Furthermore, a mobile gas phase leaving the near-field, may also impact on radionuclide transport by expelling contaminated water (see SFR-19 “Transport and release from the silo”).

A second concern of gas production is that the gas itself could be radioactive if it comprises ^{3}H or ^{14}C derived directly from the waste material. Rapid transport of radioactive gases generated by degradation of organic material is one of the primary issues for the safety of the SFR repository.

Origin in the repository system:

Gas generation in the near-field could occur by microbial action, anaerobic steel degradation and by radiolysis. The rate of gas generation by these processes will largely be affected by the available surface area of the materials, the degradability of the materials, the chemistry of the porewaters, the temperature and (for radiolysis) the activity of the waste.

Impact on the repository system:

Gas generation in the near-field will impact directly on the transport and release of nuclides from waste packages by expelling groundwaters if a free gas phase is created. A gas overpressure could be created that may cause physical damage to the engineered barriers and the near-field rock.

The nuclides ^{3}H and ^{14}C may be released directly from the waste in gaseous form.

Bibliographic references:

Grauer (1988) The corrosion behaviour of carbon steel in Portland cement. Nagra Technical Report, NTB 88-02.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS), 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) and 2.1.13 (Radiation effects in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-12**

Name: **GAS FLOW IN THE NEAR-FIELD**

Short description:

Gases can be generated in the near-field by three processes: corrosion of steel, radiolysis and microbial activity. The gas generated may form a free gas phase which could be mobile. The flow of gas from the near-field could have a significant impact on the transport and release of radionuclides and the structural integrity of the engineered barriers.

Technical description:

Gas may be generated in the SFR repository by three processes: microbially-mediated degradation of organic materials; anaerobic degradation of steel; and radiolysis (see SFR-11 “Gas generation in the repository”). Of these, the anaerobic degradation of steel is likely to be the dominant gas production mechanism for all parts of the repository. Gas generation from the degradation of organic material is likely to be significant only for the decomposition of cellulose and, consequently, an important process only in the rock vaults because limited quantities of cellulose will be placed in the silo. Radiolysis will produce only small quantities of gas and is therefore not of primary significance when considering gas issues.

The gases produced (mostly H_2, CO_2 and CH_4) will form a free gas phase if their rate of production exceeds their rate of removal from the near-field, causing concentrations to exceed solubility limits. Analysis of gas behaviour in the repository for performance assessment (SKI, 1988) suggests that this outcome is probable and, consequently, the behaviour of a free gas phase is an important consideration.

Once a free gas phase has been produced, it will tend to accumulate in void spaces in the excavations as a ‘bubble’ in the upper regions of the silo and the rock vaults. The formation of a bubble will mean that groundwater must be expelled from the excavations to make room for the gas. This groundwater potentially may be contaminated with radionuclides in dissolved or colloidal form.

The free gas itself may also be mobile and might migrate away from the repository excavations driven by density contrasts with the surrounding water and pressure gradients. If, however, the gas transport is restricted (e.g. by low gas permeabilities of the rock or surrounding engineered barriers) then a gas overpressure could develop. This overpressure has the potential to cause mechanical damage to the surrounding materials (see SFR-13 “Mechanical impact on the engineered barriers”).

This scenario is most significant for the silo. The concrete shell and the bentonite layer of the silo are designed to have low permeability to avoid lateral water flow through the structure. Gas produced in the silo should normally escape through the permeable sand/bentonite mixture used as the backfill at the top of the silo. However, in some situations, this gas pathway could become blocked allowing gas overpressure to develop. The highest possible overpressure in the silo under normal operating conditions is determined by the opening pressure for the overlying sand/bentonite mixture, which is 50 kPa. If this gas pressure did develop, this would result in a lowering of the water level in the silo by 5 m (SKI, 1994).

If the overlying sand/bentonite mixture became completely blocked (e.g. cemented) and gas pressures continued to increase in the silo, then the walls of silo will be capable of withstanding a pressure of 280 kPa at the top, without cracking, which would result in a 28 m drop in the water level. If the silo is blocked at several levels, all mobile water can, theoretically, be displaced from the silo. Although the blockage of gas vents is not considered probable, it cannot be completely excluded that such a blockage would be able to occur during the inland period (SKI, 1994).

Because the silo contains the highest activity waste in the repository, expulsion of contaminated water or mechanical damage to the silo structure due to gas overpressure could be significant for repository safety.

The situation is less likely to happen in the rock vaults. Although significant volumes of gas may be produced in the vaults due to microbially-mediated degradation of organic material, there is no low-permeability engineered barrier, with the exception of the shotcrete wall reinforcement. As a consequence, gas should be able readily to migrate from the vaults. Analysis suggests (SKI, 1988) that only a small gas bubble will form in the rock vaults (equivalent to less than 1 % of repository volume) and that gas will migrate away before significant gas overpressure will result. The consequence of this and the potential for the expulsion of contaminated groundwater is considered to be negligible.

In the near-field rock, gas will flow along the fractures in much the same way the groundwater does, moving in response to a pressure gradient. Because the fractures may carry both groundwater and a free gas phase, two-phase transport models may be required to evaluate the significance of gas transport for repository safety, although this has not been done routinely in performance assessments for the SFR. However, gas migration away from the proposed Nirex cementitious L/ILW repository has been considered in detail (Nirex 1994, 1997).

Gas bubbles could affect the transport of radionuclides in solution or colloidal form in the fractured rock mass by a number of mechanisms: 1) bubble formation may change the apparent viscosity of the groundwater and induce or accelerate movement, 2) bubbles may push groundwater in front of them along preferred directions, 3) immobile bubbles may change the direction of water flow by effectively sealing certain pathways, 4) bubbles may scavenge radionuclides and colloids from the groundwater which then attach to the gas/water interface and move with the bubble, and 5) gas dissolution/exsolution along the flowpath may affect the groundwater chemistry.

Origin in the repository system:

The development of a free gas phase is controlled by the balance between the gas generation and the release rates. If a free gas phase develops, it may accumulate in the repository excavation or may disperse to the rock, depending on the gas permeability of the near-field rock mass and the engineered barrier structures. Gas behaviour in the near-field is thus largely controlled by the degradation rate of steel and organic materials.

Gas pressures and gas transport mechanisms away from the excavations are dependent on the hydraulic characteristics of the fracture network in the near-field rock mass.

Impact on the repository system:

The development of a mobile free gas phase in the repository may have significant impacts on the structural integrity of the engineered barriers. Gas overpressure could cause these barriers to fail.

The generation of a free gas phase can expel contaminated groundwaters from the repository excavations and, in the fractured rock mass, may perturb groundwater pathways and flow rates. Gas generation thus has a potentially significant impact on the transport and release of radionuclides from the repository.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Nirex (1994) Post-closure performance assessment: gas generation and migration. Nirex Science Report, S/94/003.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Science Report, S/97/012, Four Volumes.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.12 (Gas sources and effects in wastes and EBS), 2.2.11 (Gas sources and effects in geosphere) and 3.2.09 (Gas-mediated transport of radionuclides) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-13**

Name: **MECHANICAL IMPACT ON THE ENGINEERED BARRIERS**

Short description:

Mechanical impacts on the near-field can arise due to a range of internal and external stresses. These stresses will act on the repository excavations and support structures and can be partially responsible for failure of the engineered barriers. Mechanical impacts thus can affect radionuclide releases rates and times to the far-field.

Technical description:

The near-field of the SFR repository, comprising the waste packages, backfills and the reinforced concrete structure of the silo, will be subjected to a range of mechanical impacts over the operating and post-closure phases of the repository lifetime.

These mechanical impacts will act (together with chemical processes) to degrade the engineered barrier materials and ultimately will cause them to fail. The repository is designed, however, to remain intact for a period sufficiently long that the radiological hazard posed by the radionuclide content after failure will be very low.

The most significant mechanical impacts for the SFR will be those that act on the silo because this is where the largest activity content is located. These mechanical impacts will have both internal and external origins. Similar impacts will arise in the rock vaults, however their lower activity waste content and the lack of an engineered support structure means that mechanical failure of the vaults is less critical to repository safety.

The most important internal stresses will be due to a substantial pressure rise in the silo. This pressure will result from gas generation (see SFR-11 “Gas generation in the repository”) and, to a lesser extent, from swelling of the bentonite layer around the silo (see SFR-24 “Evolution of the bentonite layer in the silo”) and swelling of any dehydrated ion-exchange resins. The concrete shell and the bentonite layer of the silo are designed to have low permeability to avoid lateral water flow through the structure. Gas produced in the silo should normally escape through the permeable sand/bentonite mixture used as the backfill at the top of the silo (see SFR-12 “Gas flow in the near-field”). However, in some situations, this gas pathway could become blocked allowing gas overpressure to develop. The highest possible overpressure in the silo under normal operating conditions is determined by the opening pressure for the overlying sand/bentonite mixture, which is 50 kPa. If the overlying sand/bentonite mixture became completely blocked (e.g. cemented) and gas pressures continued to increase in the silo, then the walls of silo will be capable of withstanding a pressure of 280 kPa at the top, without cracking (SKI, 1988).

Other internal stresses will result from the increase in temperature (see SFR-17 “Temperature of near-field”). The most important heat generation process will be exothermic curing (hydration) of the large volumes of cement in the repository. Cement curing generates significant amounts of heat over short periods (a few years) and, in the centre of the large concrete waste packages, the temperature could rise by up to about 100°C (SKI, 1984). This temperature rise will cause differential thermal expansion of the various engineered barrier materials (e.g. concrete and reinforcing rods in the silo structure) and the near-field rock, which will induce changes to the mechanical stresses in the repository.

External stresses on the repository will arise from a number of processes. In the short-term, stresses will develop during the immediate post-closure period as the rock mass deforms in response to stress readjustments and due to hydraulic resaturation (see SFR-16 “Hydraulic resaturation of the near-field”). Over the longer-term, the repository will be subjected to the changing stress field in the rock (see GEN-36 “Stress field”) which results from tectonic forces, continued isostatic uplift of the land mass and groundwater flow through the rock.

Origin in the repository system:

Mechanical impacts on the repository near-field will arise from both internal and external stresses. Internal stresses will be due to a pressure rise (from gas generation and swelling of solid materials) and an increase in temperature due to cement curing.

External stresses will be due to hydraulic resaturation, stress readjustments in the rock mass, groundwater flow and isostatic uplift of the land.

Impact on the repository system:

The mechanical impacts on the near-field of the repository will affect the stability of the excavations and the reinforced concrete silo structure. These impacts could cause mechanical failure of reinforcements and partial or total collapse (cave-in) of the excavations. This would cause mechanical impacts to be transmitted to the backfill materials and to the waste packages.

Ultimately, these mechanical impacts could result in early or enhanced release of radionuclides to the far-field.

Collapse of the repository excavations will have an effect on the surrounding rock mass and could cause further stress readjustments in the rock and fast pathways (fractures and faults) could develop through the far-field (see GEN-35 “Fast transport pathways”).

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1984) Licensing of final repository for reactor waste: SFR-1. SKI Technical Report, 84:2.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-14**

Name: **RADIATION EFFECTS IN THE NEAR-FIELD**

Short description:

Radiation emitted from the waste will impinge on all materials in the near-field and potentially radiation effects could accelerate material degradation and dissolution. However, at the relatively low activity levels in the SFR these effects are mostly insignificant for repository safety. The only potentially important process will be radiolysis of the groundwaters and organic materials.

Technical description:

Radiation from the radioactive nuclides will impinge on the wasteform and other materials in the engineered barriers of the near-field. Potentially this radiation could enhance degradation of the materials and dissolution of the wasteform. Because the activity content of the silo is much greater than in the rock vaults, radiation effects on the materials in the silo are of greatest interest.

Radiation will be emitted not only from radionuclides within the wasteform but also from radionuclides released from the waste that occur in dissolved and colloidal forms, or which have been precipitated or sorbed onto the surfaces of near-field materials (see SFR-1 “Colloid generation in the waste package”).

The effect of radiation on cement and clay (bentonite) minerals has been investigated in a number of studies (e.g. Allen and Rawson, 1986). Theoretically, radiation could affect the mineralogical structure of these minerals and, ultimately, could initiate mineralogical transformations. If this occurred, it may cause modifications to the characteristics of the minerals, such as the swelling and sorption capacities. The conclusion from experimental studies is that there is very little evidence of crystallographic change to the mineralogical structures due to radiation, even at the high radiation levels experienced in HLW repositories. Consequently, it is unlikely that radiation will have any significant impact upon cement and clay minerals in the SFR repository.

The effect of radiation on metals has been investigated from the perspective of metal canisters for HLW disposal (e.g. Shoesmith et al., 1992) and evidence suggests corrosion processes are specific to individual metals. However, radiation on metals potentially could accelerate corrosion by damaging passivating layers and initiating localised corrosion. Furthermore, the production of radiolytic oxidants and reductants could directly affect the kinetics of anodic and cathodic reactions. Nonetheless, the results from laboratory studies suggest that the effect of radiation on metals will be insignificant at the activity levels in the SFR (see SFR-4 “Degradation of steel reinforcements in the silo shell”).

Bitumen degradation can be enhanced in the presence of a high radiation field (see SFR-6 “Degradation of the bitumen matrix”). However, given that the majority of the bitumen is used to encapsulate low activity wastes, this is not of great concern to the SFR. The exception to this is could be some early packages of dried ion-exchange resins which were immobilised in bitumen and are placed in the silo. The radiation field in the silo is much greater than in the BMA rock vault and, consequently, the potential for radiation enhanced bitumen degradation is also higher. However, the actual increase in bitumen degradation rate by this process is considered still to be negligible.

Radiation from radioactive nuclides will impinge on all solid materials in the near-field and potentially this could enhance degradation and dissolution rates. However, at the activity levels in the SFR (even at the higher levels in the silo), the radiation damage will be extremely limited and not of any concern to repository safety. Consequently, radiation effects have not been investigated in detail in any ILW disposal programme and are not specifically addressed in performance assessments for ILW repositories.

The only significant radiation effect might be radiolysis (see GEN-30 “Radiolysis”). Ionising radiation from the higher activity wastes will impact on water molecules and organic materials in the waste (e.g. cellulose) to produce radicals and molecular products such as H_2O_2, H_2 and O_2. Radiolysis of the groundwater will add to the total gas production rate in the repository, although the volume of gas produced by radiolysis will be insignificant compared to the gas formed by anaerobic degradation of steel and by microbial activity (see SFR-11 “Gas generation in the repository”). Radiolysis could also lead to the formation of corrosive species such as nitrous and nitric acids and ammonia which may accelerate degradation of the cement.

Origin in the repository system:

The level of radiation exposure of the materials in the near-field is time dependent and is controlled by the longevity of the waste package and by radioactive decay.

Radiation emitted from radionuclides dissolved in the groundwater, in colloidal form or precipitated and sorbed onto near-field material surfaces will also act as sources of radiation. This radiation exposure is also time dependent and is controlled by the time taken for the waste packages to fail, radioactive decay as well as the porosity distribution in the near-field and the groundwater chemistry which controls radionuclide solubility.

Impact on the repository system:

Radiation effects from radioactive nuclides will impinge on all material in the near-field and potentially this could enhance degradation and dissolution rates. However, at the activity levels in the SFR (even at the higher levels in the silo), the radiation damage will be extremely limited and not of any concern to repository safety.

The only potentially significant process is radiolysis which contributes to the total gas production in the repository and which might create corrosive species such as nitrous and nitric acids and ammonia which may accelerate degradation of the cement.

Bibliographic references:

Allen CC and Rawson SA (1986) Effects of irradiation and dry heating on bentonite: a transmission electron microscopy and X-ray diffraction study. Microbeam Analysis, 1986, 135-137.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Shoesmith DW, Ikeda BM and King F (1992) Effect of radiation on the corrosion of candidate materials for nuclear waste containers. Material Research Society, Scientific Basis for Nuclear Waste Management, XV, 407-414.

SKI (1984) Licensing of final repository for reactor waste: SFR-1. SKI Technical Report, 84:2.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.13 (Radiation effects in wastes and EBS) and 2.1.04 (Buffer/backfill materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.13 (Radiation effects on bentonite) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-15**

Name: **RADIONUCLIDE RELEASE FROM THE WASTE**

Short description:

Radionuclides will be released from the wasteforms by a variety of mechanisms either to solution, to secondary solid alteration phases, as colloids or in the gaseous phase. The nature and abundance of the released species will depend on the chemistry of the near-field pore-waters and the degradation rate of the wasteform. Radionuclide release from the wasteform is a fundamental control on the safety of the repository.

Technical description:

Various types of inorganic and organic wastes will be present in the SFR repository, immobilised in cement and bitumen.

Inorganic wastes include, among other things, insulating material (mineral wool); steel and other scrap metals from reactor refuelling and maintenance operations; spent radioisotope sources; spent air filters and cleaning materials (see SFR-7 “Degradation of the inorganic waste”).

Organic wastes include, among other things, ion-exchange resins and sludges; filter pulps; halogenated and non-halogenated rubbers; halogenated and non-halogenated plastics; disposal clothing and cleaning materials (see SFR-8 “Degradation of the organic waste”).

Some of these materials (such as metallic objects) are directly encapsulated in cement in the waste packages, and dehydrated and powdered ion-exchange resins are immobilised in bitumen and cement. Some low activity combustible material is incinerated to reduce waste volumes and the resulting ash is then immobilised in cement.

Depending on the activity of the final wasteform, it is emplaced either in the silo (high activity waste) or in one of the rock vaults (lower activity waste). Release from the wastes in the silo is most important because the silo houses the greatest proportion of the activity in the repository.

Radionuclides will be released from the various wastes and immobilisation matrices by a number of mechanisms, such as microbial action and hydrolysis. If the wasteform is soluble, radionuclides could be released directly into solution, or else they could be incorporated into secondary (solid) alteration phases.

As the wasteform undergoes degradation, then small particles of the waste can be liberated as colloids and radionuclides could be directly incorporated in them (see SFR-1 “Colloid generation in the waste package”).

Finally, if gas is generated by degradation of the wasteform, then radionuclides could be associated directly with the gas (see SFR-11 “Gas generation in the repository”). The radionuclides most likely to be associated in a gas phase are ^{14}C and ^{3}H in gases such as

H_2, CO_2 and CH_4. Gas generation may also occur due to anaerobic degradation of steel and by microbially-mediated degradation of organic materials.

The rate of release is strongly dependent on the evolving radionuclide inventory (see SFR-22 “Changes in the radionuclide inventory”) and porewater chemistry which controls the gross degradation rate of the wasteforms and immobilisation matrices. The groundwater chemistry also is a control on the nature of the released species because the cement buffered, high pH of the near-field porewaters will act to decrease the solubilities of most radionuclides. The nature of secondary alteration phases is, in part, dependent on porewater chemistry (see SFR-20 “Groundwater chemistry in the near-field”).

A further control on the release rate is the distribution of radionuclides within the wasteform, since there will be some degree of heterogeneity in the mixing of wastes within the immobilisation matrices. There will also be heterogeneity in the distribution of radionuclides within certain wastes, such as some decommissioning components.

The most significant release processes are direct uptake in solution or in the gas phase because the water and gas are mobile and capable of transport out of the repository. Radionuclides released to solid secondary alteration phases remain immobile, while releases to colloids are essentially immobilised in the silo because of filtration in the bentonite layer around the silo but not in other rock vaults

The release of radionuclides from the wasteform is a principal control on the safety of the repository. Consequently, performance assessments for the SFR pay attention to the degradation processes affecting the wasteforms and the immobilisation matrices.

Origin in the repository system:

The release of radionuclides from the wasteform is dependent on the evolving radionuclide inventory; the degradation rate and processes affecting the wasteforms and, thus, the porewater chemistry.

Impact on the repository system:

The release of radionuclides from the wasteforms will control the wasteform inventory; the porewater chemistry in the near-field and the rate of nuclide transport from the near-field.

The release of radionuclides from the wasteform is a principal control on the safety of the repository.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0	30th June 2000	Bill Miller

Number: **SFR-16**

Name: **HYDRAULIC RESATURATION OF THE NEAR-FIELD**

Short description:

After closure of the repository, groundwater will flow from the saturated far-field rock into the void spaces in the near-field and will cause it to become hydraulically resaturated. This inflow of water will impact on the thermal and stress fields and, when in contact with the engineered barrier materials, will cause them to begin to degrade. During the resaturation process, the groundwater chemical condition will shift from oxidising in the near-field rock to increasingly reducing conditions.

Technical description:

During operation of the SFR repository, the excavations will be constantly pumped to keep them dry. After closure and sealing of the repository, however, all pumping will cease and, consequently, groundwater will begin to flow from the saturated far-field rock into the near-field causing it to resaturate. Resaturation will be driven by hydraulic pressure gradients between the unsaturated near-field and the saturated far-field rock.

Initially, groundwater will flow along fractures from the far-field into the engineered damaged zone (see GEN-15 “Excavation effects on the near-field rock”) of the near-field rock and this will begin to resaturate. As the groundwater continues to flow it will pass from the near-field rock into the excavations themselves and will begin to resaturate the void spaces there.

Flow into the excavations will be restricted by the shotcrete coating (if used) on the walls of the silo, the rock vaults and connection tunnels. However, this shotcrete is not intended to be a post-closure hydraulic barrier and will only delay resaturation for a short period of time.

Because groundwater flow into the near-field will occur along hydraulically-active fractures (see GEN-23 “Groundwater flow”), resaturation will tend to occur via discrete line or point sources depending on the degree of channelling within the fractures.

Resaturation of the silo will be a relatively slow process because the bentonite layer around the concrete shell acts as a barrier to advective flow. In addition, uneven uptake of water by the bentonite produces uneven swelling of the material and this will tend to reduce the flow of groundwater into the silo at that location (see SFR-24 “Evolution of the bentonite layer in the silo”). However, because the buffer material used at the top and bottom of the silo is a more permeable mixture of sand and bentonite, it is likely that resaturation of the silo will occur via water travelling in the vertical direction. The presence of air and unsaturated conditions in the near-field will cause two-phase flow conditions with lower effective permeability and also trapped air may locally reduce the permeability until all the air is dissolved into the groundwater.

Once water has entered the silo, it is relatively free to flow around the waste packages because of the porous nature of the cementitious grout used to backfill between the waste packages. Water movement within the silo will only be restricted by the walls that segment the internal structure of the silo.

Resaturation of the rock vaults will be more rapid than in the silo because the crushed rock backfill offers no resistance to groundwater flow. In the absence of the flow barrier, the rate of resaturation of the rock vaults will be dependent on the groundwater flow rate in the surrounding rock which will vary with time as the hydraulic gradients changes in response to progressive resaturation.

The duration of resaturation is likely to be measured in years and, consequently, is only a short time in relation to the overall time periods considered in performance assessments. As a result, no specific quantitative assessments of resaturation are considered in performance assessments, other than assuring structural integrity of the excavations and engineered barriers.

Resaturation may be delayed or even reversed by gas production (see SFR-11 “Gas generation in the repository”). Large volumes of gas will be evolved by degradation of steel and organic material. It is possible, though not likely, that large enough volumes of gas will accumulate in the excavations to generate a gas pressure sufficient to slow down significantly the resaturation rate.

As the silo and the rock vaults resaturate, water will also begin to saturate those waste packages manufactured from concrete and filled with cementitious immobilisation matrix, because the cement is permeable to a certain extent. Resaturation of waste in the drums cannot commence until corrosion perforates the steel drum. However, in performance assessments for the SFR, no credit is assigned for any actual lifetime of the waste packages (SKI, 1994).

The composition of the first waters to enter the excavations will change rapidly as they interact with the free air and the engineered barrier materials (see SFR-20 “Groundwater chemistry in the near-field”). Dissolution of the free air will cause the porewaters to be oxidising initially. However, early cement degradation will rapidly buffer the pH of the porewaters, driving them to alkaline values.

Resaturation will also affect the thermal and stress field in the near-field, as mobile water begins to transport away the heat energy generated from exothermic cement degradation reactions. In addition, during the resaturation phase itself, heterogeneous resaturation will induce complex stress fields in the near-field rock and the engineered barriers (see GEN-36 “Stress field”).

Once resaturation is complete and after the initial thermal excursion, the hydraulic, thermal and stress fields will rapidly approach equilibrium. The exception to this would be if a gas overpressure developed that could cause expulsion of porewaters from the near-field.

Origin in the repository system:

Resaturation of the near-field will commence as soon as the repository is sealed and pumping stops. Resaturation will be driven by hydraulic pressure gradients between the unsaturated near-field and the saturated far-field rock.

The rate and duration of resaturation will be controlled by the groundwater flow field (in the fracture network), the hydraulic conductivity of the engineered barriers (particularly the bentonite layer around the silo), the temperature and the gas pressure in the excavations.

Impact on the repository system:

Resaturation of the near-field will have a significant effect on the repository because most chemical degradation processes and radionuclide transport processes cannot commence until the near-field is saturated. Therefore, resaturation will affect the timing of the onset of degradation of the engineered barriers and the wasteforms.

Resaturation will also affect the thermal and stress fields in the engineered barriers and the near-field rock.

Ultimately, because groundwater provides the primary vector for the transport of radionuclides, resaturation impacts on the radionuclide release rates and processes.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-17**

Name: **TEMPERATURE OF THE NEAR-FIELD**

Short description:

The temperature of the near-field is controlled by heat generation in the waste and engineered barriers, superimposed on natural geothermal conditions. Elevated temperatures in the near-field can affect the rates of chemical and microbiological processes, and can influence groundwater flow, diffusion coefficients and, hence, radionuclide transport.

Technical description:

The ambient temperature of the rock at repository depth in the vicinity of the SFR is approximately 15°C (see GEN-38 "Temperature of the far-field"). This temperature is controlled by the natural geothermal gradient (approximately 10 to 20°C /km for Sweden) and the mean surface temperature (approximately 5 to 10°C for Sweden).

The temperature of the SFR near-field will be controlled by heat generating processes superimposed on the ambient temperature. Three heat generating processes will operate in the SFR near-field, the most important being the curing of cement. Other heat generating processes are radioactive decay and microbially-induced degradation of organic material.

Heat is evolved during the cement curing (hydration) process. Thus heat will be generated in the concrete shell and other reinforcements, in the cementitious grout used between waste packages in the silo, and in the cemented waste packages themselves. Cement curing generates significant amounts of heat over short periods (a few years) and, in the centre of the large concrete waste packages, the temperature could rise by up to about 100°C (SKI, 1984).

The second most important heat generating mechanism is radioactive decay (see GEN-28 "Radioactive decay"). The highest activity waste will be placed in the silo and, consequently, radiogenic heat generation is potentially of significance only to this part of the repository. The most important (highest activity) nuclides for heat generation in the silo are the activation product ^{60}Co and the fission product ^{137}Cs. However, the content of heat-producing waste in the silo is still quite low, compared to spent fuel and, as a result, this process will only be of secondary importance. Radiogenic heat production was not considered in detail in performance assessments for the SFR but it has been investigated quantitatively in the performance assessments for the Nirex cementitious L/ILW repository which was planned to contain higher activity waste than the SFR (Nirex, 1997).

Microbial degradation of organic material is also a potential heat generating mechanism. However, this process is likely to be only of tertiary significance to repository temperatures and, consequently, this process has not been considered in any performance assessment for a deep repository.

Heat generation rates will be greatest for the silo because it contains large volumes of cement and the highest activity waste. In addition, heat loss from the silo will occur predominantly by conduction because the concrete shell and bentonite buffer will stop heated groundwaters from leaving by convection/advection mechanisms. The balance between heat generation and heat loss will mean that, in the short period after closure when cement curing is active, temperatures can rise significantly. For perspective, calculated thermal evolution of the Nirex L/ILW repository showed that cement curing would raise the temperature of the near-field to approximately 80°C with 10 years of closure, dropping to 60°C after 30 years. The temperature would be maintained at 60°C by radiogenic heat production (Nirex, 1995). Temperatures in the silo are not expected to be this high because the SFR is not as deep (lower ambient temperature) and contains lower activity waste than the Nirex repository. However, the temperature in the silo could still be expected to rise by a few tens of degrees due to cement curing.

Maximum temperatures in the rock vaults will be less than in the silo because the heat generation rates will be lower and because groundwaters can flow through the vaults.

Elevated temperatures in the repository will impact on chemical and microbiological processes. The chemical degradation of organic wastes and the corrosion of metals are likely to be accelerated, for instance. Furthermore, the solubility of many chemical species is increased at elevated temperatures.

Thermal gradients across the near-field will also induce thermal convection of groundwaters, thermal diffusion (see SFL-44 “Soret effect in the buffer and backfill”) and thermal stresses in the rock mass and the engineered barriers. These processes may combine to enhance radionuclide transport out of the near-field. Nonetheless, the overall safety of the repository is less sensitive to temperature increases than to other processes, notably the production of organic complexants that act to increase radionuclide solubilities.

Origin in the repository system:

Heat will be generated in the near-field by exothermic cement curing (hydration) processes and, to a lesser extent, by radioactive decay and microbially-induced degradation of organic material. The rate of heat generation will be controlled by the mass and composition of the cement, the supply of water to the cement, the activity content of the waste and the availability of readily biodegradable material.

The rise in near-field temperatures will be controlled by the balance between heat generation and heat loss (conduction and convection) which is largely controlled by the hydraulic conductivity of the near-field rock and engineered barriers. Temperatures in the silo potential might rise by a few tens of degrees.

Impact on the repository system:

A rise in the near-field temperature potentially can impact on all chemical and microbiological processes such as material degradation and radionuclide solubility. Material degradation rates are likely to be accelerated. Temperature gradients across the near-field can impact on the stress field, groundwater convection and thermal diffusion. These processes can combine to enhance radionuclide transport.

Because expected maximum temperatures are not excessive and the thermal pulse is not of a long duration, the impact of heat generation on the safety of the repository is not considered to be of primary significance.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Nirex (1995) Post-closure performance assessment: near-field evolution. Nirex Science Report, S/95/009.

Nirex (1997) An assessment of the post-closure performance of a deep waste repository at Sellafield. Nirex Science Report, S/97/012, Four Volumes.

SKI (1984) Licensing of final repository for reactor waste: SFR-1. SKI Technical Report, 84:2.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Matt White and Bill Miller

Number: **SFR-18**

Name: **RADIONUCLIDE RELEASE FROM THE WASTE PACKAGE**

Short description:

Radionuclides released from the waste will initially be located inside the waste packages. The packages themselves are not designed with any specific post-closure containment role and will degrade relatively quickly after repository closure. Consequently, radionuclide release from the waste packages will occur quickly after release from the wasteform. The release process from the waste packages does not, therefore, afford any particular role in repository safety.

Technical description:

A variety of waste packages will be used in the SFR repository to hold the waste and they will all be fabricated either from steel or from concrete. None of these waste packages is designed for any particular long-term, post-closure containment role.

After repository closure and resaturation, these containers will not, therefore, isolate the wastes from groundwaters for any significant period of time. Consequently, the wasteforms can begin to degrade and radionuclides can begin to be released from the waste soon after closure. Radionuclides released from the waste will initially be located inside the waste packages.

Some radionuclides will be released from the waste in mobile form, either dissolved in the groundwater or in colloidal (see SFR-1 “Colloid generation in the waste package”) or gaseous phases (see SFR-11 “Gas generation in the repository”). Other radionuclides will be incorporated into solid secondary alteration phases and will, thus, remain immobile and cannot be released from the waste package.

A primary control on the amount of radionuclides released from the waste package will be the groundwater composition which controls the solubility of radionuclides, particularly in terms of the cement buffered, high pH of the near-field porewaters which will act to decrease solubilities of most radionuclides (see SFR-20 “Groundwater chemistry in the near-field”).

The mobile radionuclides will, however, be able to be released from the waste packages easily because their containment ability will be limited. Their release from the waste package will be controlled by the groundwater and gas flow rates through the waste package.

Because no containment role is attributed to the waste packages, the performance assessments of the SFR do not pay any specific attention to the lifetime or behaviour of the waste packages. The more important process is the release of radionuclides from the wasteform itself.

Origin in the repository system:

The release of radionuclides from the waste package is dependent on the degradation of the waste package material (steel and concrete); the radionuclide release rate from the wasteform and the groundwater and gas flow rates through the waste package.

The porewater chemistry controls the degradation of the waste package material and the wasteform itself.

Impact on the repository system:

The release of radionuclides from the waste package affects the porewater chemistry in the near-field. However, the waste package is not assigned any containment role, consequently the release from the waste package is not, itself, considered in performance assessment.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-19**

Name: **TRANSPORT AND RELEASE FROM THE SILO**

Short description:

Radionuclides may be transported out of the silo in groundwater or in gaseous form. While the bentonite layer around the silo remains intact, dissolved radionuclides can be released only by diffusion through the bentonite layer. Gases can permeate through the bentonite/sand layer at the top of the silo. Radionuclide release from the silo is a primary control on the safety of the repository because of the high activity content of the silo.

Technical description:

The predicted total activity for the SFR is $9.9\text{x}10^6$ GBq based on the 1987 waste forecast, most of which will be located in the silo. Consequently, the release of radionuclide from the silo is a primary control on repository safety. Transport and release of radionuclides from the silo will occur via groundwater (for radionuclides in dissolved and colloidal form) and in the gas phase.

The silo is constructed from a reinforced concrete shell, 90 cm thick, with a layer of bentonite filling the void space between the shell and the rock. The reinforced concrete shell provides the primary strength and stability for the silo complex. The bentonite layer at the sides of the silo is compacted and approximately 1.3 m thick. At the top and bottom of the silo, a sand/bentonite mixture (90% sand) is used as the buffer material to provide greater bearing strength (at the bottom) and gas permeability (at the top).

Due to the high degree of compaction of the bentonite layer, no water will flow through it, provided it remains intact. Consequently, radionuclides in solution and in colloidal form within the silo will be retarded. Dissolved radionuclides may only exit the silo through the walls by diffusion (see SFR-9 “Diffusion in the near-field”) and radionuclides in colloidal form will be essentially immobilised by filtration in the bentonite (see SFR-10 “Colloid filtration in the near-field”).

The bentonite layer will slowly degrade by a variety of chemical and physical processes, such as reaction with the hyperalkaline leachates derived from the cementitious materials and from internal and external stresses (see SFR-24 “Evolution of the bentonite layer in the silo”). These processes will act with time to increase the hydraulic conductivity of the bentonite layer, most likely by forming a number of fast pathways through the bentonite. Eventually, continued chemical and physical degradation of the bentonite will cause it to fail entirely as a barrier to advective flow. At this point, the groundwater, colloids and gas could all pass through the silo walls. However, SKI estimates of the silo behaviour (SKI, 1994), are that structural integrity should be maintained for at least 1000 years after closure.

A primary control on the amount of radionuclides released in groundwater will be the groundwater composition which controls the solubility of radionuclides, particularly in terms of the availability of organic complexants which can significantly increase the solubility of certain species (see SFR-20 “Groundwater chemistry in the near-field”). On the other hand, the high pH of the silo porewaters will act to decrease solubilities of most radionuclides.

Radionuclides in solution will have their transport retarded by a number of mechanisms in addition to the diffusive barrier provided by the bentonite layer. In particular, the sorption capacity of the near-field will be very high because of the large surface area of cementitious materials and iron-oxyhydroxides (from steel corrosion).

Gas produced in the silo should normally escape through the permeable sand/bentonite mixture used as the backfill at the top of the silo (see SFR-12 “Gas flow in the near-field”). However, in some situations, this gas pathway could become blocked allowing gas overpressure to develop. The highest possible overpressure in the silo under normal operating conditions is determined by the opening pressure for the overlying sand/bentonite mixture, which is approximately 50 kPa. The gas released from the silo may be associated with radionuclides (e.g. as ^{14}C in CO_2) and, consequently, gas release from the silo will affect total radionuclide release.

A primary control on the amount of radionuclides released in gaseous form will be the gas production rate which will define whether a free gas phase will occur. If a free gas phase does occur, then the gas production rate will be a partial control on the gas pressure and, therefore, the flow rate of gas out of the silo.

Transport and release from the silo is a primary control on the safety of the SFR repository. Therefore, performance assessments for the SFR pay attention to these processes, particularly to the longevity of the diffusive barrier provided by the silo walls and the bentonite layer.

Origin in the repository system:

Transport and release of radionuclides from the silo will occur via groundwater (for radionuclides in dissolved and colloidal form) and in the gas phase.

The release of radionuclides in groundwater will be controlled by the groundwater chemistry (radionuclide solubilities); the hydraulic characteristics of the silo walls and the bentonite layer, particularly its permeability; the hydraulic characteristics of the far-field and the groundwater flow rate. As the silo degrades, the permeability of the bentonite layer will increase, eventually allowing direct flow to occur across the walls.

The release of radionuclides in association with the gas phase will be largely controlled by the gas production rate and the gas permeability of the bentonite/sand mixture at the top of the silo.

Impact on the repository system:

Transport and release from the silo is a primary control on the safety of the SFR repository. The release rates of dissolved radionuclides from the silo affects the porewater compositions inside and outside the silo and, thus, sorption-desorption and precipitation-dissolution reactions.

The release rates of gases from the silo affects the gas pressure in the silo and thus the mechanical impact on the system.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) evaluation of SKB’s in depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS), 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) and 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-20**

Name: **GROUNDWATER CHEMISTRY IN THE NEAR-FIELD**

Short description:

The groundwater chemistry in the near-field will be substantially altered by changes to recharge chemistry and by direct reaction with the waste and with the materials used in the engineered barriers, particularly cement and steel. Alterations to the groundwater chemistry will affect radionuclide solubility and sorption, may affect barrier performance and, consequently, will impact on radionuclide release.

Technical description:

The natural groundwater in the rocks at repository depth at the SFR is controlled by the chemistry of the recharge waters (see GEN-37 “Surface water chemistry”) and their reaction with the rock (see GEN-1 “Alteration and weathering along flow paths”). At the present time, the groundwater is saline but, as the land mass continues isostatic uplift, the groundwater will become more dilute as the saline water interface passes the repository. The change to ‘fresh’ water is expected to occur in approximately 1000 years time (SKI, 1994).

The groundwater will be substantially altered as it passes into the repository near-field. The groundwater will react with the engineered barriers (predominantly cement and steel) causing its chemistry to be substantially altered. The groundwater will react with the large volumes of cement causing its pH to rise from natural near-neutral values to at least pH 12.5, conditioned by dissolution of $Ca(OH)_2$ from the cement. During the early stages, the pH may exceed this owing to dissolution of NaOH and KOH from the cement. However, this period is likely to be relatively short owing to the small amounts of these hydroxides contained in the cement. Thus, the main control of pH is $Ca(OH)_2$ which buffers pH to • 12. The rate of $Ca(OH)_2$ dissolution is controlled by the water turnover in the near-field. For the silo, turnover is very slow because of the very low hydraulic conductivity of the concrete shell and bentonite layer. In the rock vaults, however, where there is no barrier to advection, groundwater turnover will be faster. Consequently, hyperalkaline conditions will be maintained for longer in the silo than in the rock vaults.

The groundwater will also react with the steel containers and reinforcements, causing them to corrode. Initially corrosion will occur under oxidising conditions. However, as the free oxygen is consumed in the process, the groundwater will become increasingly chemically reducing and slower anaerobic corrosion of the steel. The rate of this buffering reaction, and the final redox potential reached, will depend on the availability of steel for reaction. Steel will be present in all parts of the repository, although smaller volumes will be present in vaults housing concrete tanks than elsewhere. Consequently, redox conditions in these vaults may remain oxidising for longer than elsewhere in the repository.

The evolution of high pH, chemically reducing conditions will tend to limit the rates of further chemical corrosion. More importantly, these conditions will strongly affect the solubility of radionuclides released from the waste. Most nuclides (though not all) are poorly soluble under these chemical conditions. The high pH will also affect sorption of Cs and Sr during the early period when K and Na are released during dissolution of their hydroxides, although other nuclides will not be affected.

The importance of the pH, Eh conditions for controlling radionuclide solubility is a critical for determining the safety of the repository. Consequently, performance assessments for the SFR pay particular attention to modelling radionuclide solubilities under these chemical conditions using thermodynamic solubility and speciation codes and their associated databases.

The groundwater chemistry will also be affected by the presence of organic complexing agents. These occur naturally in low concentrations in most groundwaters but much larger concentrations may be generated in the repository near-field by the degradation of organic materials in the waste (see SFR-8 “Degradation of the organic waste”) and from substances used in connection with repository construction, especially additives to the concrete.

The effect of organic complexing agents can be to increase the solubility of some otherwise poorly soluble species. The generation of colloids capable of sorbing radionuclides by the degradation of solid materials will also act to increase the mobility of radionuclides (see SFR-1 “Colloid generation in the waste package”). Thus, together, an increase in the concentration of organic complexing agents and population of colloids could enhance the potential for the release and transport of radionuclides. The significance of this process will depend on potential for radionuclides to form complexes with the organic species and to be sorbed to the colloids, and the flow of water through the near-field causing mobilisation of the complexes and colloids. In the silo, groundwater flow will be slower than in the rock vaults due to the flow barrier presented by the silo structure. Consequently, radionuclides associated with complexes and colloids in the silo are not likely to be released in significant amounts to the near-field rock until the concrete shell and bentonite layer are mechanical disrupted. Radionuclides associated with complexes and colloids in the rock vaults will more easily be transported by flowing groundwater but the activity content of the wastes in the rock vault is much lower than the wastes in the silo, making the impact on radioactive releases minimal.

The impact of organic complexants on the radionuclide solubility is a key issue for performance assessment but is one that is less easily quantified than the impact of Eh and pH because predictive models for the nature and concentrations of complexants formed from microbial degradation of organic materials are not yet well established. Particular uncertainties relate to microbial populations expected to exist under hyperalkaline conditions. Performance assessments for the Nirex cementitious L/ILW repository suggest that microbial populations may be limited by the hyperalkaline conditions (Nirex, 1995).

Near-field groundwater chemistry will further be affected by gases (e.g. H_2, CO_2 and CH_4) released from corrosion of steel and organic materials and, to a lesser extent, from radiolysis (see SFR-11 “Gas generation in the repository”). These gases will initially be dissolved in the groundwater but may form a free gas phase if they exceed solubility limits. The generation of these gases will also act to control the pH and redox potential, together with the large buffering capacity provided by the volumes of cement and steel. The presence of naturally occurring dissolved gases in the groundwaters will also be a control on these processes.

The increased temperature of the near-field for a short period after repository closure (see SFR-17 “Temperature of near-field”), due to exothermic cement curing (hydration) and, to a lesser extent, from radiogenic heat output, might accelerate chemical and microbiological processes, and may affect the solubility of some radionuclides. However, this thermal excursion will be minor and relatively short-lived and, consequently, the changing temperature is only a secondary control on the groundwater chemistry.

Origin in the repository system:

The groundwater chemistry in the near-field will be determined by the composition of the natural groundwaters and their interactions with the engineered barrier materials. The natural water composition is controlled by the composition of recharge waters, their interactions with the rock and the rate of isostatic uplift, and is expected to change from saline to dilute in the next 1000 years.

The most important engineered barrier materials that affect the groundwater chemistry are cement (buffering pH), steel (buffering Eh) and organic material (controlling concentration of organic complexants and Eh). The groundwater composition in the near-field is thus controlled by the availability of these materials for reaction and the rate of their degradation.

Maintenance of the chemical conditions is affected, in part, by the rate of groundwater movement through the near-field. Hence the hydraulic gradient, and the hydraulic conductivity of the near-field rock and the engineered barrier materials are important controlling factors.

Impact on the repository system:

Changing groundwater compositions in the near-field will affect the processes and rates of further material degradation. The evolution of high pH, chemically reducing conditions will tend to limit the rates of further chemical corrosion and will reduce microbial populations, slowing microbial degradation of organic materials.

The solubility of most radionuclides will be significantly reduced by the development of hyperalkaline conditions which will limit the radionuclide releases.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Nirex (1995) Post-closure performance assessment: near-field evolution. Nirex Science Report, S/95/009.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS), 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0	30th June 2000	Matt White and Bill Miller

Number: **SFR-21**

Name: **GROUNDWATER MOVEMENT IN THE NEAR-FIELD**

Short description:

Post-closure water movement in the SFR repository will depend on location. In the silo, water movement across the silo walls will occur by diffusion because of the compacted bentonite layer. However, in the rock vaults, water may flow freely between the vaults and the near-field rock, driven by head gradients in the rock mass.

Technical description:

The initial water movement in the SFR repository will be associated with post-closure resaturation when, after pumping, groundwater will begin to flow from the saturated far-field rock into the near-field. Resaturation will be driven by hydraulic pressure gradients between the unsaturated near-field and the saturated far-field rock (see SFR-16 “Hydraulic resaturation of the near-field”). After resaturation, the nature of groundwater movement in the repository will depend on location.

In the silo structure, there will be no groundwater flow across the bentonite layer that surrounds the reinforced concrete structure, as long as it remains intact, because of the very low permeability of the compacted bentonite layer. Consequently, water movement inside the silo will be disconnected from any flow in the near-field rock. The water inside the silo will move through the porous cementitious backfill emplaced around the waste packages. This water movement may be driven by convection cells, if a thermal gradient occurs across the silo due to heat generated by exothermic cement curing. Alternatively, the groundwater movement may be driven by upward, buoyant transport of a free gas phase derived by the anaerobic corrosion of steel and degradation of organic materials. However, as mentioned above, although the water is free to move within the silo, it cannot flow across the bentonite layer while it is intact. Instead, individual water molecules and contaminant may only move between the silo and the near-field rock, through the bentonite, by diffusion (see SFR-9 “Diffusion in the near-field”).

However, the bentonite will slowly degrade by a variety of chemical and physical processes, such as reaction with the hyperalkaline leachates derived from the cementitious materials and from internal and external stresses (see SFR-24 “Evolution of the bentonite layer in the silo”). These processes will act with time to increase the hydraulic conductivity of the bentonite layer, most likely by forming a number of fast pathways through the bentonite. Eventually, continued chemical and physical degradation of the bentonite will cause it to fail entirely as a barrier to advective flow. At this point, water will be free to flow directly between the silo and the near-field rock. However, SKI estimates of the silo behaviour (SKI, 1994), are that structural integrity should be maintained for at least 1000 years after closure.

The situation is different for the rock vaults because no specific low permeability barrier is used in their construction. Consequently, immediately after closure and resaturation, groundwater will be free to flow into the vaults from the near-field, with flow being restricted only by the shotcrete coating (if used) on the walls of the rock vaults and connection tunnels. However, this shotcrete is not intended to be a post-closure hydraulic barrier and will only restrict flow for a short period of time. Water flow in the vaults will be through the permeable crushed rock, or crushed rock and sand, backfill.

Within the vaults, water flow will be connected directly to water flow in the near-field rock and will be driven by hydraulic pressure (head) gradients in the surrounding system (see GEN-23 “Groundwater flow”). The head gradient in the surrounding rock will be time dependent and will change as the position of the coast moves as the land rises and surface conditions change from submarine to subaerial.

Water movement in the near-field rock will be concentrated in the hydraulically-active fractures. The fracture density immediately adjacent to the excavations will be higher than in the rock mass further away because of the engineered damage zone that will extend into the rock for a short distance from the walls of the excavation (see GEN-15 “Excavation effects on the near-field rock”).

Water movement in the near-field will be responsible for the transport of radionuclides in solution and in colloidal form. If two phase flow occurs, then water movement will also have an impact on the transport of radionuclides associated with the gas phase. Consequently, water movement through the near-field is important for determining overall repository safety.

Origin in the repository system:

Water movement in the near-field is controlled by the pressure (head) gradients in the surrounding rock mass, by gas generation and transport, by the temperature field, and by the hydraulic characteristics of the engineered barriers and the near-field rock.

In the silo, the bentonite layer will restrict water movement between the silo and the near-field rock to diffusive transport only. No diffusive barrier exists in the rock vaults.

Impact on the repository system:

Water movement in the near-field is a control on the transport of radionuclides in solution and in colloidal form. If two phase flow occurs, then water movement will also have an impact on the transport of radionuclides associated with the gas phase.

Water flow leaving the repository will affect the chemistry of the near-field porewaters and thus the continued processes of degradation of the engineered barrier materials.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEP 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-22**

Name: **CHANGES IN THE RADIONUCLIDE INVENTORY**

Short description:

The initial radionuclide inventory defines the abundance of every radionuclide species in the waste at the time of emplacement. After emplacement, the initial radionuclide inventory will change due to radioactive decay and migration of radionuclides out of the near-field.

Technical description:

The initial radionuclide inventory defines the abundance of every radionuclide species in the waste at the time of emplacement. The majority of the waste to be emplaced in the SFR is reactor waste (also known as operating waste) from the operation of nuclear power plants and from the central interim facility for spent fuel (CLAB). In addition, the SFR will house smaller quantities of radioactive materials generated in research, industry and medical activities.

The nature of this waste is variable. Solid inorganic wastes include, among other things, insulating material (mineral wool); steel and other scrap metals from reactor refuelling and maintenance operations; spent radioisotope sources; spent air filters and cleaning materials. Organic wastes include ion-exchange resins and sludges; filter pulps; halogenated and non-halogenated rubbers; halogenated and non-halogenated plastics; disposal clothing and cleaning materials.

The predicted total activity for the SFR is 9.9×10^6 GBq based on the 1987 waste forecast, most of which will be located in the silo. Initially, the activity of the waste will be dominated by ^{60}Co and ^{137}Cs, both of which have short half-lives of 5.2 and 30.2 years respectively. The ^{60}Co is an activation product in structural materials and hence its final inventory is relatively simple to predict while the ^{137}Cs is a fission product whose final inventory will depend on any fuel failures which will arise.

Other important high activity nuclides are activation products ^{55}Fe (2.7 years) and ^{63}Ni (100 years), and fission products ^{90}Sr (28.8 years) and ^{134}Cs (2.3 years).

The quantities of transuranic nuclides in the operating waste will be very low. The most important transuranic nuclides will be ^{239}Pu and ^{240}Pu because of their very long half-lives; 24400 and 6600 years respectively. Calculations of individual dose rates from performance assessments for the SFR (SKI, 1988) suggest these nuclides will dominate doses after 10000 years post-closure, but at extremely low absolute values.

The inventory waste forecast is only an estimate and is based on assumptions for operating life-times for reactors and fuel batches, fuel manufacturing and processing procedures and the nature of radioactive sources from non-power related operations. The inventory forecast is, however, a primary requirement for the performance assessment calculations.

After emplacement, the initial radionuclide inventory will change due to radioactive decay (see GEN-28 "Radioactive decay") of the radionuclides present in the waste. Radioactive decay will result in a gradual decrease in the total radioactivity of the waste, over time after emplacement, and in a decrease in the activity of most individual radionuclides. However, the activity of some daughter nuclides in the decay chains (e.g. ^{241}Am) will increase during well-defined periods after emplacement as parent-daughter equilibria are established. Radioactive decay will occur under all circumstances in the repository.

The radionuclide inventory of the waste will also be subjected to change as radionuclides are released and transported out of the near-field. Transport will preferentially remove from the waste the most mobile nuclides: these will be those that can occur in gaseous form (e.g. ^{14}C in CO_2 and ^{3}H in H_2) and those that are more soluble under repository physico-chemical conditions. Transport process do not, however, change the total radionuclide inventory of the repository system only the inventory of that component of the waste remaining within the waste package.

Origin in the repository system:

The initial radionuclide inventory of the waste is defined by the activity of materials produced during normal reactor operations and, to a lesser extent, from industry, research and medicine.

After emplacement, the radionuclide inventory of the repository will change (net decrease) due to radioactive decay. The radionuclide inventory of the waste remaining in the near-field will be further changed (net decrease) by release and transport processes which are, themselves, affected by physical, chemical and microbiological processes operating in the near-field. However, the release and transport processes cannot by themselves change the total radionuclide inventory of the repository system, only the inventory remaining within the waste packages.

Impact on the repository system:

Changes to the radionuclide inventory act as a primary control on the release rates for radionuclides from the wasteform and, thus, on calculated dose rates.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 3.1.01 (Radioactive decay and in-growth) and 2.1.01 (Inventory, radionuclide and other material) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-23**

Name: **GROUNDWATER CHEMISTRY IN THE NEAR-FIELD ROCK**

Short description:

The water chemistry in the near-field rock is controlled by the composition of the ambient natural groundwater, the composition of waters leaving the repository excavations having interacted with the engineered barriers and the degree of mixing between these two waters. The water chemistry of the near-field rock is a control on the transport and retardation processes affecting radionuclide released from the engineered barriers.

Technical description:

The water chemistry in the near-field rock around the SFR repository will be spatially and temporally variable. In simple terms, two potential end-member groundwaters can be considered at occur at any time after closure and resaturation of the repository.

The first end-member groundwater will be the natural, ambient groundwater occurring in the rock at repository depth. Considering a simple, linear groundwater flow pattern with groundwater flowing in, through and out of the engineered barriers, the natural groundwater end-member represents the composition of groundwater before it comes into contact with the engineered barrier system materials (see GEN-22 “Far-field groundwater chemistry”).

This natural groundwater composition will be time dependent, controlled by the rate of uplift of the land mass (see GEN-39 “Uplift and subsidence”). At the present time, the groundwater is saline but, as the land mass continues isostatic uplift, the groundwater will become more dilute as the saline water interface passes the repository. The change to ‘fresh’ water is expected to occur in approximately 1000 years time (SKI, 1994).

The second end-member groundwater will be the water leaving the repository excavations after having interacted with the engineered barrier system materials, particularly the cement and steel (see SFR-20 “Groundwater chemistry in the near-field”). The composition of this water will be significantly changed compared to the natural groundwater. Dissolution of the Na, K and Ca hydroxides in the cement will cause the pH of the water to become highly alkaline, while redox buffering by iron in the system will buffer the Eh to strongly chemically reducing conditions. In addition, these waters may also contain significant concentrations of organic complexants from the degradation of organic waste materials, such as cellulose and its soluble degradation products; dissolved gases (e.g. H_2, CO_2 and CH_4) from the anaerobic degradation of steel and the microbially-induced degradation of organic materials; and dissolved and colloidal radionuclides released from the various wasteforms.

The composition of the waters leaving the engineered barriers will be time dependent, controlled by the extent of degradation of the near-field materials. In particular, the pH will gradually reduce as the system is buffered first to around 12 pH units by the rapid dissolution of the small amounts of NaOH and KOH in the cement (Lagerblad and Trägårdh, 1994) and then to around 12.5 pH units by the slower dissolution of the $Ca(OH)_2$. Also the concentration of released radionuclides will be controlled by the time taken for the waste packages and the silo structure to degrade, and by radioactive decay (see GEN-28 “Radioactive decay”).

The waters leaving the repository are thus alkaline and contaminant-rich. They will be out of chemical equilibrium with the rock and the natural groundwaters, and represent a ‘contaminant plume’ spreading away from the repository.

In simple terms, then, the composition of the waters in the near-field rock up-stream of the repository will be natural while the composition of the waters in the near-field rock down-stream of the repository will be alkaline and contaminant-rich.

Travelling away from the repository, the contaminant plume will spread-out by dispersion mechanisms and mix with the natural groundwaters (see GEN-10 “Radionuclide dispersion”). The degree of mixing will increase with distance from the repository, consequently the composition of the waters in the rock down-stream of the repository will decrease in alkalinity and contain lower concentrations of contaminants.

The contaminant plume will also react with the minerals in the rock and fracture coating surfaces because of the enhanced solubility and rate of reaction of most silicates and aluminosilicates at high pH (Savage et al., 1992). The region of chemical reaction represents the chemically disturbed zone. Because of the dissolution of silicates, the chemically disturbed zone may be characterised by increased porosity. However, zeolites and calcium-silicate-hydrate minerals are likely to form as the feldspars and micas react with the alkaline pore waters (alkali-silica reaction). This reaction may lead to the filling of porosity and sealing of groundwater flow paths. This in turn may lead to the preferential channelling of groundwaters and gas releases. The impact of the chemically disturbed zone on the rock porosity thus is controlled by the balance between the dissolution and precipitation reactions. The size of the chemically disturbed zone will be controlled by the geometry and velocity of groundwater flow and the rates of mineral reaction.

The behaviour of radionuclides released from the engineered barriers to the near-field rock will be controlled by the composition of the waters in the chemically disturbed zone and by the dissolution-precipitation reactions occurring there. Potentially, the formation of zeolites and calcium-silicate-hydrate minerals will act to ‘fix’ in the mineral lattices those radionuclides that can be included in the mineral structure, while other radionuclides can be immobilised by sorption to the mineral surfaces (see GEN-34 “Radionuclide sorption”). Furthermore, the elevated pH conditions will ensure low radionuclide solubility (see GEN-27 “Radionuclide precipitation and dissolution”). However, blocking or porosity and channels will limit the opportunity for matrix diffusion and reduce the flow wetted surface (see GEN-25 “Matrix diffusion”). Consequently, the net effect of the chemically disturbed zone may be to increase or decrease the transport of radionuclides through the near-field rock.

Performance assessments for the SFR (SKI, 1994) conservatively neglect radionuclide retardation in the far-field and therefore do not need to pay particular attention to the chemically disturbed zone and its impact on retardation. This is largely because the high pH conditions within the engineered barrier systems creates a chemical barrier where radionuclide retention is more effective. Consequently, the opportunity for radionuclides to reach the near-field rock in solution is minimal, although some radionuclides may still reach the far-field at a slow rate.

Origin in the repository system:

The chemistry of the groundwaters in the near-field rock is governed by mixing between the natural groundwater and waters leaving the engineered barrier system.

The composition of the natural waters is controlled by the rate of land uplift and the chemistry of the recharge waters, while the chemistry of the waters leaving the engineered barriers is controlled by the extent of degradation of the barrier materials and the wasteforms. The chemistry of the near-field groundwaters is thus time dependent.

The size of the chemically disturbed zone will be controlled by the geometry and velocity of groundwater flow and the rates of mineral reaction.

Impact on the repository system:

The chemistry of the near-field groundwaters impacts on radionuclide solubility and sorption and, thus, on transport and release processes in the near-field rock.

The near-field rock porewater chemistry also affects the distribution of porosity and permeability in the near-field via mineral dissolution-precipitation reactions. Consequently the groundwater flow geometry in the near-field will be affected.

Bibliographic references:

Lagerblad L and Trägårdh J (1994) Conceptual model for concrete long time degradation in a deep nuclear waste repository. SKB Technical Report, TR 95-21.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D, Bateman K, Hill P, Hughes C, Milodowski A, Pearce J, Rae E and Rochelle C (1992) Rate and mechanism of the reaction of silicates with cement pore fluids. Applied Clay Science, 7, 33-45.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB's in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 3.2.01 (Dissolution, precipitation and crystallisation, contaminant), 3.2.03 (Sorption/desorption processes, contaminant) and 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFR-24**

Name: **EVOLUTION OF THE BENTONITE LAYER IN THE SILO**

Short description:

The bentonite layer of the silo provides a barrier to groundwater flow. It also acts as a mechanical barrier to protect the concrete shell of the silo. The bentonite will degrade over time by physical and chemical processes and thus its barrier functions will diminish. This will have an impact on radionuclide release processes and rates from the silo.

Technical description:

The silo structure in the SFR repository comprises a reinforced concrete shell surrounded by a layer of bentonite between the shell and the rock. The bentonite layer at the sides of the silo is approximately 1.3 m thick and is compacted. At the top and bottom of the silo, a sand/bentonite mixture (90% sand) is used as the buffer material to provide greater bearing strength (at the bottom) and gas permeability (at the top). In all cases, the bentonite that is used is Na-bentonite (montmorillonite).

The purpose of the bentonite layer is to provide a barrier to groundwater flow and to serve as a mechanical buffer between the rock and the concrete shell. Consequently, for as long as the silo remains intact, transport of groundwater and radionuclides out of the silo will predominantly occur by slow diffusion rather than by faster advection (see SFR-9 “Diffusion in the near-field”).

Bentonite can react with dissolved species in groundwater and undergo mineralogical transformations with resulting changes to its physical properties (e.g. its swelling capacity). The principal mechanism by which these transformations will occur is ion-exchange, although other reactions are possible. In the presence of natural groundwaters, the ion-exchange process may be restricted because their content of exchange cations is generally quite limited. However, in the SFR environment, the supply of exchange cations (and other reactants) is considerably greater than in natural groundwaters because of the presence of large volumes of cement. In addition, the initial thermal excursion (caused by exothermic cement hydration) can accelerate reactions (see SFR-17 “Temperature of near-field”). The groundwater chemistry will also evolve as a result of these interactions, however it is unclear if the bentonite would ever attain thermodynamic equilibrium at near-field temperatures (Aja and Rosenberg, 1992).

The groundwater will react with the large volumes of cement causing its pH to rise from neutral values to around 12 or 13 by dissolution of the NaOH, KOH and $Ca(OH)_2$ present in the cement. The dissolved Na^+, K^+, Ca^{2+} and OH^- ions from the cement will diffuse through the concrete shell into the bentonite layer and each of these can interact with the bentonite minerals.

The potassium and sodium will be involved with ion-exchange reactions. The substitution of calcium for sodium in the bentonite will result in a reduction in plasticity and in swelling capacity, leading to an increase in the hydraulic conductivity. The substitution of potassium for sodium can result in mineralogical transformation from smectite to illite. Illitisation would result in an almost complete loss of swelling capacity, which would have a considerably more serious impact on the hydraulic characteristics of the buffer.

In the high pH environment, the Na can react with the bentonite to form Na-rich zeolites, such as analcime, and the hydroxide ions can also lead to changes in the aluminium silicate structure of the bentonite to form zeolites, calcium-silicate-hydrate minerals or gels.

Other mineralogical transformations of the bentonite can result from reaction with chemical species released from other materials in the silo (such as metals) and from impurities in the cement and in the bentonite itself.

Reactions may also occur between the bentonite and dissolved iron from the metallic components of the engineered barrier system, resulting in the formation of non-swelling phases such as chamosite, greenalite and nontronite (Grauer, 1990). Conversion of smectite to these minerals is poorly understood and it is uncertain whether such processes could be important over repository lifetimes.

The bentonite also contains small quantities of sulphate and carbonate. Precipitation of gypsum and calcite can occur when the dissolved calcium is provided by the degrading cement. Brucite can also precipitate because magnesium occurs naturally in the groundwater, although this may be unlikely. These and other precipitation reactions can cause cementation of the bentonite, affecting its physical and hydraulic properties.

Although many different mineralogical transformations are theoretically (thermodynamically) possible, it is formation of Ca-bentonite by ion-exchange that is considered the most important because $Ca(OH)_2$ (portlandite) is volumetrically the most significant component of the cement. Consequently, it is this reaction that is considered most extensively in performance assessments for the SFR repository (SKI, 1988; 1994). However, the rate of this reaction is relatively slow and will be limited by the rate of diffusion of the reactants through the compacted bentonite. Consequently, the progression of the reaction front will be more rapid in the higher permeability sand/bentonite layers at the top and bottom of the silo than through the compacted bentonite layer at the sides of the silo.

Mineralogical transformation of the bentonite may also affect (reduce) it sorption capacity for radionuclides. However, it is difficult to quantify this affect and, consequently, performance assessments for the SFR do not consider changing values for the distribution coefficients over time to reflect alteration of the bentonite. This is an issue which could be further investigated.

Physical damage to the bentonite layer of the silo may also be caused by stresses internal and external to the silo. Internal stresses could result from a gas overpressure (arising if the gas vents in the top of the silo become blocked) or, less likely, from swelling of the solid waste materials, such as dehydrated ion-exchange resins immobilised in bitumen. However, physical damage to the bentonite layer resulting from internal stresses is only likely to occur after mechanical failure of the concrete shell (see SFR-13 “Mechanical impact on the engineered barriers”).

External stress will impact via the rock directly onto the bentonite layer. One important role of the bentonite layer is to protect the concrete shell of the silo from external stresses (see GEN-36 “Stress field”). Short-term external stresses on the bentonite are most likely to arise during the immediate post-closure period as the rock mass deforms in response to stress readjustments and due to hydraulic resaturation (see SFR-16 “Hydraulic resaturation of the near-field”). Over the longer term, external stresses will result from isostatic uplift of the land mass (see GEN-39 “Uplift and subsidence”).

Both the internal and external stresses, together with degradation of the concrete shell, will eventually result in mechanical damage to the bentonite layer. The most significant impact of this mechanical damage will be to increase the hydraulic conductivity of the bentonite layer, most likely by forming a number of fast pathways through the bentonite. Eventually, continued chemical and physical degradation of the bentonite will cause it to fail entirely as a barrier to advective flow. SKI estimates of the silo behaviour (SKI, 1994) are that structural integrity should be maintained for at least 1000 years after closure.

Origin in the repository system:

Evolution of the bentonite layer in the silo will occur under anaerobic, highly alkaline conditions. The most significant chemical impact on the bentonite will result from alkaline leachates from cement degradation with the most important reaction being exchange of calcium (from cement leachate) for sodium in the bentonite. Other, less important, mineralogical transformations are also likely to occur.

The rate of alteration will be affected by the rate of degradation of the cement and then the rate of diffusion of the reactants through the bentonite. Thus alteration of the bentonite is controlled by the water permeability of the silo concrete, the resaturation state of the repository, the pore water chemistry and the temperature.

Physical damage to the bentonite will occur as the silo is subject to internal and external stresses. The most significant impact is likely to come from gas overpressures within the silo resulting from hydrogen gas evolved during anaerobic corrosion of the steel waste packages.

Impact on the repository system:

The degradation of the bentonite layer will impact on the ability of the silo to act as a barrier to advective flow. Consequently, the evolution of the bentonite controls the period during which radionuclides can leave the silo only by diffusional processes.

Degradation of the bentonite layer will also impact on the pore water chemistry, the sorption capacity of the bentonite and on the ability of the bentonite to act as a mechanical buffer protecting the concrete shell from stresses in the rock.

Bibliographic references:

Aja SU and Rosenberg PE (1992) The thermodynamic status of compositionally-variable clay minerals: a discussion. Clays and Clay Minerals, 40, 292-299.

Grauer R (1990) The chemical behaviour of montmorillonite in a repository backfill - selected aspects. Nagra Technical Report, NTB 88-24E.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1988) Review of final repository for reactor waste SFR-1. SKI Technical Report, 88:2.

SKI (1994) Evaluation of SKB’s in-depth safety assessment of SFR-1. SKI Technical Report, 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-1**

Name: **SWELLING OF THE BENTONITE BUFFER**

Short description:

Compacted, dry bentonite blocks used for the buffer will take-up water during the resaturation phase and swell as the clay minerals adsorb water into their lattice structure. Swelling of the bentonite buffer will affect properties which are important for water and gas transport through the buffer and for radionuclide transport and release.

Technical description:

The bentonite buffer will be emplaced as highly compacted bentonite blocks. Bentonite is dominantly composed of smectite clay. The smectites are noted for their ability to take-up water or organic liquids between their structural layers, and they all show marked cation-exchange properties. The swelling property means that, during the resaturation, the tunnel backfill will absorb water and swell, causing a significant decrease in the porosity and hydraulic conductivity of the material.

The tunnel backfill will be similar to the buffer in that it is a mixture of bentonite and an inert filler material, such as quartz sand, and will also swell (see SFL-45 “Swelling of the tunnel backfill”).

Smectites have 2:1 layer structures, so-called because of the proportion of ‘tetrahedral’ layers of Si^{4+}, O^{2-} and OH^{-} ions and ‘octahedral’ layers of Al^{3+}, O^{2-} and OH^{-} ions. It is the interlayer cation that is exchangeable, such that in a solution of NaCl, the interlayer cations will be mainly Na^{+}. If the interlayer cation is divalent, two layers of water molecules may be present in the interlayer space, unless the humidity is very low. When the interlayer cation is Na^{+}, the amount of interlayer water can increase almost indefinitely as the relative humidity approaches 100 %, causing swelling. However, increasing ionic strength and divalent cation content of saturating groundwaters will tend to reduce the swelling capacity of smectites.

In the repository situation, bentonite may be used as the buffer material and will be emplaced as machine-cut blocks of compacted dry bentonite. As the repository resaturates after closure, the bentonite will uptake groundwater causing it to swell and fill void spaces between the canister and the near-field rock. The swelling bentonite may also fill any cracks intersecting the disposal holes.

Swelling into the void spaces should reduce groundwater flow in the near-field and thus is a positive feature. However, in most performance assessment models, it is assumed that the interface between the bentonite and the rock is perfect. If, in fact, some voids do remain due to bentonite swelling failing to fill all spaces caused by the irregularity of the rock surface and cracks in the rock, then this would be a non-conservative assumption. The final swelling pressure and hydraulic properties of the water-saturated bentonite buffer and backfill will depend on the amount of void space to be filled during swelling. Consequently, the greater the number of cracks and the larger the void spaces in the disposal hole and in any tunnels connected to disposal holes, the lower will be the swelling pressure and the higher the final hydraulic conductivity.

During resaturation, a transient stage of heterogeneous swelling of the bentonite is likely to occur as infiltrating groundwater will be focussed on certain high-flow zones intersecting the disposal hole (see SFL-42 “Hydraulic resaturation of the buffer and backfill”). In extreme cases, uneven swelling of the bentonite could lead to permanent preferential diffusive pathways through the buffer (or even zones of flow). Uneven swelling of the bentonite may also lead to heterogeneous stresses on the canister (shearing forces) and the impact of this on the canister may need to be investigated (see SFL-30 “Mechanical impact on the canister”). Furthermore, during the resaturation phase, the inflow of groundwater will be partially opposed by the radiogenic heat output from the waste which will cause some drying of the initially wetted bentonite. The balance between the groundwater influx and drying processes will affect the rate of resaturation.

The swelling pressure will be transmitted to the canister and to the near-field rock. The canister should be designed initially to resist this pressure, which is likely to be approximately 10 MPa in addition to the hydrostatic pressures of around 5 MPa, according to the SITE-94 analysis. However, as the canister begins to degrade, any build-up of corrosion products which are more voluminous than the canister material will result in a compression of the bentonite buffer and an increase in swelling pressure. When the canister eventually fails (see SFL-18 “Failure of the copper shell”), the bentonite will swell into the canister, resulting in a small drop in swelling pressure and a small increase in hydraulic conductivity in the buffer.

The impact of the swelling pressure on the rock should not result in any gross mechanical disruption of the rock mass. Indeed, the swelling pressure may act like a rock support. However, swelling of bentonite into cracks intersecting the disposal holes potentially could lead to their widening. Mathematical analysis of this process for the SITE-94 performance assessment indicated that this is only likely to occur for cracks shorter than one disposal hole diameter. The swelling capability of the bentonite is strongly controlled by its mineralogy. Mineralogical changes due to interactions with the groundwater or adjacent barrier system materials, may result in the formation of a new clay mineral form with reduced swelling capacity.

Considerable amounts of cement and concrete will be used as grouting and structural materials in the repository. Cement pore fluids are hyperalkaline (pH 12.5-13.5) and highly reactive with carbonate, silicate, and aluminosilicate minerals. Migration of these pore fluids into the clay buffer material has the potential to exchange calcium ions from the cement for sodium ions on smectite mineral surfaces (see SFL-3 “Chemical alteration of the buffer and backfill”). This may potentially lower the swelling capacity of the clay, and dissolve the framework of the buffer clay, precipitating a variety of calcium-silicate-hydrate minerals and zeolites, thus decreasing the porosity, plasticity, and swelling capacity of the clay (Savage, 1997). However, because of the amounts of cement and concrete envisaged, alkaline alteration of montmorillonite can affect only a small part of the buffer. In addition, these interactions are likely to lead to positive solids volume changes. Although there may be detrimental effects to plasticity and clay swelling properties where this alteration is most pronounced, this may be counteracted by decreased porosity.

Origin in the repository system:

Swelling of the bentonite in the buffer results from post-closure resaturation of the compacted bentonite by infiltrating groundwaters. The rate of resaturation and the final swelling pressures are controlled, in part, by the hydrogeological characteristics of the far-field and near-field rock. However, the proportion of void space in the near-field to be filled by swelling is also a significant control on the final swelling pressure and on the final hydraulic and mechanical characteristics of the buffer.

Impact on the repository system:

Swelling of the bentonite in the buffer will result in gross changes to the hydraulic properties of the near-field, such that transport in the buffer should occur by diffusion only. This will have significant impact on the transport of water, gas and released radionuclides. Swelling pressures will impact on the rock but the effect should be minimal. Swelling pressures will also impact on the canister but the canister will resist this pressure until corrosion has significantly reduced its load bearing capacity.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Pusch R and Börgesson L (1992) Performance assessment of bentonite clay barrier in three repository concepts: VDH, KBS-3 and VLH. SKB Technical Report, TR 92-40.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.1.1 (Swelling of bentonite into tunnels and cracks) and 3.2.1.2 (Uneven swelling of bentonite) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-2**

Name: **CHANGES IN THE SPENT FUEL RADIONUCLIDE INVENTORY**

Short description:

The initial radionuclide inventory defines the abundance of every radionuclide species in the spent fuel at the time of waste emplacement. After emplacement, the initial radionuclide inventory will change due to radioactive decay and any nuclear reactions that result from criticality events. The inventory of the spent fuel will also change as radionuclides are released and migrate out of the canister.

Technical description:

The initial radionuclide inventory describes the abundance of every radionuclide species in the spent fuel at the time of emplacement. This includes radionuclides in the spent fuel itself and any radioactive activation products in the non-fuel components of the waste; e.g. radionuclides formed by neutron capture on the zirconium alloy (Zircaloy) cladding tubes.

This initial radionuclide inventory is controlled by the composition of the original nuclear fuel (before being loaded into a nuclear power reactor); the burn-up conditions, position and times in the reactor; the nature of any (re-)processing operations on the spent fuel; and the duration of storage between removal from the reactor and emplacement in the repository.

Of these factors, the burn-up conditions can be the most variable. Average reactor burn-up rates differ for different reactor designs and for individual reactors. Burn-up rates have increased over the last decade as improved fuel quality has allowed the fuel to be used for longer periods in the reactor. Thus, spent fuel can be generated with very different burn-up histories. These burn-up rates are important because the inventory of radionuclides generated during reactor operation is related directly to them.

The inventory used in performance assessment calculations is not measured in the spent fuel but rather is calculated on the basis of known or predicted burn-up rates for individual batches of fuel from each reactor. The initial radionuclide inventory of the spent fuel will comprise several ‘types’ of radionuclide:

- the remaining long-lived radionuclides originally present in the nuclear fuel;
- radioactive fission products formed by nuclear fission during reactor operation; and
- radioactive isotopes of the transuranic elements (actinides) formed by activation (neutron capture) in uranium (and in any heavier elements already formed) during reactor operation.

The majority of these radionuclides will be incorporated into the spent fuel matrix. However, small but significant fractions (up to a few percent) of certain fission and activation products (such as I, Cs, Tc, Sb, Mo, Sr, Cl, Pd, Sn, Se, Sm) are likely to be present in cracks in the UO_2 matrix and in the gaps between fuel and cladding as well as in grain boundaries (see

SFL-22 “Gap and grain boundary release”). Smaller releases (around 0.5%) of other elements (such as Cm, Am, Pu, Pa, Th, U and Ra) can also occur by this process.

In addition, there will be a fourth ‘type’ of radionuclide, the radioactive activation products (C, Ni, Nb and possibly Zr) formed by neutron capture on metallic non-fuel components of the waste.

After emplacement, the initial radionuclide inventory will change due to radioactive decay (see GEN-28 “Radioactive decay”) of the radionuclides present in the spent fuel and in the non-fuel components of the waste. Radioactive decay will result in a gradual decrease in the total radioactivity of the waste, over time after emplacement, and in a decrease in the activity of most individual radionuclides. However, the activity of some daughter nuclides in the decay chains (e.g. ^{241}Am) will increase during well-defined periods after emplacement as parent-daughter equilibria are established. Radioactive decay will occur under all circumstances in the repository.

The post-emplacement radionuclide inventory may also change if nuclear reactions are initiated as a result of a criticality event (see SFL-12 “Criticality”). If criticality occurs in the repository it will initiate chain reactions that will generate new fission products and, possibly, transuranic elements if neutron capture occurs. As a consequence, the inventory and activity of fission products and transuranic elements, and the total activity of the waste will increase during the period of the criticality event, as occurred during criticality in the natural fission reactors at Oklo, Gabon. The increase in the fission product inventory of a repository due to criticality would be system specific but analyses suggest that it would be low in most cases (around a 1% increase: Rechard et al., 1997). A criticality event could only occur in the repository if fissile material were redistributed in the near-field by an appropriate sequence of release, transport and deposition processes, such that a critical mass could form. Criticality is considered to be a very low probability event in the Swedish reference spent fuel repository design (Oversby, 1996).

The radionuclide inventory of the spent fuel will also be subjected to change as radionuclides are released and transported out of the waste package by groundwater. This process does not, however, change the total radionuclide inventory of the repository system only the inventory of that component of the waste remaining within the canister.

Origin in the repository system:

The initial radionuclide inventory of the spent fuel is defined by fuel production, reactor operation and any reprocessing operations. After emplacement, the radionuclide inventory of the repository will change (net decrease) due to radioactive decay. The radionuclide inventory of the waste remaining in the canister will be further changed (net decrease) by release and transport processes which are, themselves, affected by canister integrity, the chemistry of the groundwater and the physico-chemical stability of the wasteform. However, the release and transport processes cannot by themselves change the total radionuclide inventory of the repository system, only the inventory remaining within the waste package.

The total radionuclide inventory of the repository system could, however, be changed (net increase) if the release and transport processes initiated a criticality event. Criticality would be dependent on the physico-chemical conditions in the near-field that control transport and deposition (reconcentration) of fissile material and on the geometry of the near-field such that a suitable mass and shape of fissile material could form.

Impact on the repository system:

Changes to the radionuclide inventory (including abundances and distributions in the fuel, grain boundaries, metallic non-fuel components and gap spaces) act as a primary control on the release rates for radionuclides from the wasteform and, thus, on the chemistry of the near-field groundwaters.

Bibliographic references:

Liljenzin JO (1990) Some estimates of the total nuclide inventory in the year 2100 from Swedish nuclear power production. SKI Technical Report, 90:18.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Oversby VM (1996) Criticality in a high level waste repository - a review of some of important factors and an assessment of the lessons that can be learned from the Oklo reactors. SKB Technical Report, TR 96-07.

Rechard RP, Tierny MS, Sanchez LC and Martell MA (1997) Bounding estimates for criticality events when directly disposing of highly enriched spent nuclear fuel in unsaturated tuff. Risk Analysis, 17.

Equivalent FEPs:

Relates to FEPs 3.1.01 (Radioactive decay and in-growth) and 2.1.01 (Inventory, radionuclide and other material) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.1.2 (Radioactive decay) and 1.1.1 (Criticality) in the SKI/SKB Scenario Development Project (SKI, 1989)

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-3**

Name: **CHEMICAL ALTERATION OF THE BUFFER AND BACKFILL**

Short description:

The bentonite buffer and bentonite-rich backfill may be chemically altered due to interaction with the groundwater, including its dissolved components, adjacent solid materials and by the thermal load. This will affect the physico-chemical characteristics of the buffer and backfill.

Technical description:

The buffer and the backfill will be chemically altered due to interactions with the natural groundwater; the dissolved species from engineered barrier system materials; any adjacent solid barrier materials and their degradation products; and the adjacent near-field rock. However, it is unclear if clays would ever attain thermodynamic equilibrium in low-temperature systems (Aja and Rosenberg, 1992). Similar reactions may occur involving the bentonite in both the buffer and backfill which will result in an increase in hydraulic conductivity. A limited increase in hydraulic conductivity may be less significant in the buffer considering that the bentonite would be emplaced in a heavily compacted form. The thermal load will also affect alteration rates.

The principal mechanism by which the bentonite (smectite) will interact with the groundwater and its dissolved species is ion-exchange. This will have a significant impact on the porewater chemistry (see SFL-53 “Evolving water chemistry in the buffer” and SFL-54 “Evolving water chemistry in the backfill”). The most likely exchange reaction is between sodium in the bentonite and calcium in the groundwater, although other exchanges are possible depending on the composition of the groundwater. The substitution of sodium for calcium in the bentonite will result in a reduction in plasticity and in swelling capacity, leading to an increase in the hydraulic conductivity, although the significance of this process is thought to be minimal.

If potassium is abundantly present in the groundwaters, then exchange of potassium for sodium can result in mineralogical transformation from smectite to illite. Illitisation would result in an almost complete loss of swelling capacity, which would have a considerably more serious impact on the hydraulic characteristics of the buffer and backfill. However, under expected repository conditions, illite transformation reactions will be limited and should not impact on repository behaviour, even after 10^5 years, because of the low concentration of potassium in many groundwaters (although this is a site specific characteristic) and because the temperatures will be too low after the initial thermal peak for illitisation reactions to proceed (Huang et al., 1993; Pusch, 1995).

Reaction may also occur between the bentonite and dissolved iron and copper derived from the metallic components of the engineered barrier system, resulting in the formation of non-swelling phases such as chamosite, greenalite and nontronite (Grauer, 1990). Conversion of smectite to these minerals is poorly understood and it is uncertain whether such processes could be important over repository lifetimes. It is expected that they might be limited to bentonite in the buffer immediately adjacent to the waste package (Amcoff, 1998).

The reduction in swelling capacity would occur for bentonite in the backfill as well as the buffer. In the backfill the main impact of this would be on the groundwater fluxes in the vicinity of deposition holes due to the increase in transmissivity of shafts and drifts, predominantly at the interface between the backfill and the rock. This potentially may allow a 'short-circuit' between shallow waters and waters at repository depth.

The chemistry of the buffer and the backfill may also be affected by dissolution-precipitation reactions. A possible reaction in a calcium-rich groundwater would be precipitation of calcite, involving the carbonate component of the buffer (which is not pure bentonite). Calcite precipitates may result in a reduction in the porosity (blocking of pores) and also in a reduction in plasticity; a cementation reaction. This process is most likely during the initial high temperature phase because calcite solubility is lower at high temperature. Other precipitates could occur later in the form of iron and copper solid phases, and in precipitates involving radionuclides leached from the wasteform.

Potentially more serious chemical alterations could affect the bentonite in the buffer and the backfill in the presence of hyperalkaline fluids derived from the degradation of any cement present in the repository. The rate and products of bentonite-hyperalkaline leachate reactions are uncertain, and will be system dependent, but hydroxide ions in the water are likely to react with smectite to form zeolites, calcium-silicate-hydrate minerals or gels. Such alteration will impact clay swelling behaviour, transport properties (porosity, permeability) and sorption properties. The extent of such reactions will be partially controlled by the supply of hyperalkaline fluids and by the high pH-buffering capacity of smectite. Similar reactions would occur when cement is in direct physical contact with the buffer or backfill. Experimental and analogue evidence is required to examine these issues.

Chemical degradation of the buffer and the backfill could also be caused by deficiencies in the materials used or by unexpected chemical compositions of the groundwater. However, the ion-exchange properties of the bentonite may then be beneficial and have a positive impact in cases where the groundwater chemistry deviates from the standard composition.

Origin in the repository system:

Chemical alteration of the buffer and backfill will result from interaction (ion-exchange and precipitation reactions) between the bentonite and the groundwater and dissolved species derived from degradation of the engineered barrier system components. Chemical alteration of the buffer and backfill is thus affected by resaturation of the buffer after repository closure; the composition of the water in the near-field rock, the tunnel backfill and in the canister; canister and wasteform corrosion; microbial activity; radiolytic decomposition of water in the buffer; temperature in the buffer; groundwater flow in the buffer; diffusion of species in the buffer; generation and transport; thermal diffusion of dissolved species and Soret effects.

Impact on the repository system:

The result of chemical alteration of the buffer and backfill could be a reduction of swelling capacity and a reduction in plasticity. The effect of these processes would be to change the hydraulic characteristics of the near-field by increasing the hydraulic conductivity of the buffer and backfill and, possibly, leading to preferential transport pathways and even the formation of flow paths. Precipitation of solid phases in the pore spaces and other cementation reactions could result in a reduction of the permeability of the material and channelling.

Chemical reactions involving the bentonite and its impurities such as sulphides, sulphates, organic compounds, carbonates, and Fe(II) will influence the chemical composition of the groundwater which will have a consequent effect on the degradation of other engineered barrier system components.

Bibliographic references:

Aja SU and Rosenberg PE (1992) The thermodynamic status of compositionally-variable clay minerals: a discussion. Clays and Clay Minerals, 40, 292-299.

Amcoff O (1998) Mineral formation on metallic copper in a ‘future repository site environment’ textural considerations based on natural analogues. SKI Technical Report, 98:7.

Grauer R (1990) The chemical behaviour of montmorillonite in a repository backfill - selected aspects. Nagra Technical Report, NTB 88-24E.

Huang W, Longo J and Pevear D (1993) An experimental kinetic model for smectite to illite conversion and its use as a geothermometer. Clays and Clay Minerals, 41, 162-177.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1994) Evaluation of SKB’s In-depth Safety Assessment of SFR-1. SKI Technical Report 94:30.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA,1998).

Relates to FEPs 3.1.1 (Degradation of bentonite by chemical reactions), 3.1.7 (Reactions with cement porewaters) and 3.1.10 (Interactions with corrosion products and waste) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-4**

Name: **COAGULATION OF BENTONITE**

Short description:

In a ground or porewater containing suspended bentonite clay particles, there is sometimes a tendency for the bentonite particles to coagulate and form larger aggregates. This process is promoted by an increase in the salinity of the solution. Newly formed aggregates may sink due to gravity and settle to form a bentonite-rich sediment.

Technical description:

In a repository, it is possible that a dilute suspension of bentonite clay particles in groundwater may form in void spaces in the near-field. Void spaces could arise if the bentonite buffer or bentonite-rich backfill is removed by flowing groundwater in the near-field rock adjacent to disposal holes or tunnels (see SFL-17 “Erosion of the buffer and backfill”). If a void space did form by this or a similar process, the remaining bentonite buffer would expand, causing a reduction in its density and some of the remaining bentonite could form a suspension with the groundwater.

Coagulation can not occur if the density and swelling capacity of the bentonite are both sufficiently high to avoid a water-filled void forming.

The suspended bentonite particles initially may remain in solution and become evenly distributed throughout the water-filled space due to the electrostatic repulsion between their negative electrically charged surfaces (electrical double layers). However, this electrical repulsion force may be overcome by two closely related processes causing particles to aggregate and precipitate out of the suspension (see SFL-43 “Sedimentation of the buffer and backfill”).

The first process is “coagulation” and results from a reduction of this electrostatic repulsion, such that suspended particles of bentonite may aggregate. This coagulation process can be readily promoted by an increase in salinity: because of the double layer of electrical charge surrounding a charged particle, this aggregation mechanism is sometimes called ‘double-layer compression’.

The second process is “flocculation” and results from the simultaneous adsorption of an organic or inorganic complex on two separate clay particles, thereby acting as a bridging compound to bind the particles together. Introduction of cationic complexes, particularly large, hydrophobic molecules, into a clay-rich suspension can stimulate flocculation. As the bentonite particles bind together they form larger particles which are commonly referred to as ‘floc’. The flocs increase in size until they become heavy enough to fall out of the suspension. Flocculation can also remove from suspension populations of microorganisms that may occur in the near-field groundwater.

In practice, however, both coagulation and flocculation may occur in a suspension and, consequently, it is common for both the terms to be used interchangeably.

Both of these processes require that a sufficiently high concentration of particles occur in the suspension such that particles are close enough together to bind either by coagulation or flocculation processes. This phenomenon is represented by the concept of a critical coagulation concentration.

In the repository environment, coagulation of bentonite can only occur if a suspension forms in a water-filled void space with a concentration that exceeds the critical coagulation concentration. Such a requirement implies that significant disruption of the engineered barrier system (e.g. erosion of the buffer/backfill) must occur before coagulation of bentonite is a possibility. If such a situation did occur, coagulation could limit further erosion of the buffer by removing from suspension bentonite particles that would otherwise be transported with the flowing groundwater. Likewise, coagulation limits the transport of radionuclides that might be sorbed to suspended bentonite particles.

Origin in the repository system:

The origin of coagulation in the repository near-field relates to the formation of water-filled void spaces in which a bentonite particle suspension can occur. Void spaces may be formed by the removal of buffer and backfill material by flowing groundwater (“erosion”) or by the movement of the plastic buffer into fractures apertures and spaces in perforated canisters to such an extent that the swelling capacity of the bentonite becomes insufficient completely to fill all voids. The bentonite suspension will coagulate if the groundwater is saline or contains sufficient ligand concentrations to bind together the bentonite particles. Coagulation of bentonite is thus determined by the near-field groundwater flow regime, the degradation of engineered barrier system components and the near-field groundwater chemistry.

Impact on the repository system:

If a suspension of bentonite can form in a water-filled void in the near-field, and coagulation subsequently occurs, then this will have an impact on the near-field groundwater chemistry. Coagulation will limit further erosion of the buffer/backfill by removing from suspension particles that could be carried away in the flowing groundwater. It thus also limits transport of any radionuclides that may be sorbed onto those bentonite particles. However, the greater impact comes about due to an increase in the hydraulic conductivity of the remaining bentonite resulting from a decrease in its density and compaction. In other words, the significance of coagulation for repository safety is minimal compared to the impacts of other processes (buffer erosion and density reduction) that bring about the formation of a bentonite suspension from which coagulation can take place.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.5 (Coagulation of bentonite) in SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-5**

Name: **COLLOID BEHAVIOUR IN THE BUFFER AND BACKFILL**

Short description:

Colloids may be generated by chemical, physical and microbiological processes in the bentonite buffer and bentonite-rich backfill. Radionuclides can sorb onto these colloids which may affect their subsequent transport and retardation in the buffer, backfill and beyond.

Technical description:

Colloids are small, solid particulate materials suspended and dispersed in the groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed 'suspended particles' and anything smaller is considered to be in solution.

The significance of colloids arises from the fact that they can sorb radionuclides and can be mobile in flowing groundwaters. This means that colloids can increase the apparent concentration in groundwater of radionuclides with low true solubilities. In addition, because of their very small size, they provide a very large sorptive area. The transport and retardation of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution.

The bentonite used in the buffer and backfill is comprised of small clay particles. Physical erosion of this material by advecting groundwater at the interface between the buffer or backfill and the near-field rock can liberate individual clay minerals that become suspended in the water as colloids (see SFL-17 "Erosion of the buffer and backfill"). These colloids will then be additional to the colloid population in the host rock and may act as carriers for radionuclides and thereby affect the radionuclide transport from the repository.

Colloids may also be formed by direct precipitation within the mass of the buffer and backfill as a result of the thermal and chemical gradients and 'fronts' (e.g. a redox front) that may occur within them. These colloids could, for example, be formed of carbonate due to the lower solubility of calcite at higher temperatures.

In addition, microbes in the buffer/backfill or substances produced by microbes may form colloidal aggregates. However, due to the very high compaction of the buffer and backfill under normal conditions, no water will flow through them. Consequently, colloids formed within the buffer and backfill will be immobilised (filtration) and any radionuclides sorbed onto them will be retarded.

Likewise, colloids formed within the waste package (wasteform and canister) will not be able to pass through the compacted bentonite buffer. Therefore, any radionuclides sorbed onto these colloids will be effectively retarded. Because of the ability of the compacted bentonite buffer to immobilise colloids (filtration), the transport of radionuclides by colloids is generally considered to be of significance only in the far-field rock (NEA, 1993).

However, if erosion of the buffer or backfill is excessive, then the bentonite will become less compact in which case it may be possible for water to flow to occur through the bentonite. In this case, colloids formed within the buffer and backfill or the waste package may be able to migrate out to the near-field rock. However, this is considered to be a very low probability event, although it could be more likely in the bentonite-rich backfill because this has a lower initial compaction than the buffer.

Origin in the repository system:

In the bentonite buffer and bentonite-rich backfill, colloid populations are controlled by physical erosion by flowing groundwater at the interface with the near-field rock. In addition, colloids may be formed within the mass of the buffer and backfill due to the presence of chemical and thermal gradients and interfaces, and microbiological activity.

Impact on the repository system:

Colloids can sorb radionuclides thus increasing their apparent solubility. Once sorbed to colloids, radionuclide transport and retardation behaviour can be significantly changed from that of the same radionuclide in solution.

Radionuclides bound to colloids formed by erosion of the buffer and backfill at the interface with the near-field rock may be transported by the flowing groundwater. However, radionuclides bound to colloids formed within the mass of the buffer and backfill will be effectively retarded because the high degree of compaction will make the colloids immobile.

Bibliographic references:

Kurosawa S, Yui M and Yoshikawa H (1997) Experimental study of colloid filtration by compacted bentonite. Scientific Basis for Nuclear Waste Management, 465, 963-970.

NEA (1993) The status of near-field modelling. NEA/OECD, Paris.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D (1995, editor) The scientific and regulatory basis for the geological disposal of radioactive waste. John Wiley & Sons.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in geosphere), 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) and 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.4 (Colloid generation - source) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-6**

Name: **COLLOIDS AND PARTICLES IN THE CANISTER**

Short description:

Colloids may be generated by chemical, physical and microbiological processes in the canister. Radionuclides can sorb onto these colloids which may affect their subsequent transport out of the near-field.

Technical description:

Colloids are small, solid particulate materials suspended and dispersed in the groundwater. Usually, colloids are defined as suspended solids between 1 nm and 1 mm in diameter, larger solids are termed 'suspended particles' and anything smaller is considered to be in solution.

The significance of colloids arises from the fact that they can sorb radionuclides and can be mobile in flowing groundwaters. This means that colloids can increase the apparent concentration in groundwater of radionuclides with low true solubilities. In addition, because of their very small size, they provide a very large sorptive area. The transport and retardation of colloid-bound radionuclides is dependent on the behaviour of the host colloid population and may be very different from that of the same radionuclide in solution.

In the spent fuel repository, colloids can be formed by degradation of the spent fuel, metal non-fuel waste parts and the canister itself. Radionuclides could be associated with the colloids by sorption onto the colloid surfaces or they may be directly included in the colloidal material if the colloid is a small fragment of the spent fuel or metal non-fuel waste part containing radionuclide activation products.

In addition, colloids may be formed by direct precipitation within the canister as a result of the thermal and chemical gradients and 'fronts' (e.g. a redox front) that may occur there. These colloids could, for example, be formed of carbonate due to the lower solubility of calcite at higher temperatures.

If the colloids are suspended in the water and are small enough to be able to escape through the failed canister they may enhance radionuclide release (see SFL-49 "Radionuclide release and transport from the canister"). However, they are unlikely to be able to migrate through the bentonite buffer because of its high degree of compaction which filters them out (see SFL-5 "Colloid behaviour in the buffer and backfill").

Alternatively, the colloids may be sorbed to surfaces inside the canister or be trapped in the canister by some other mechanism. Either way, it is considered unlikely that colloids formed in the waste package could travel to the near-field rock and, therefore, radionuclides sorbed onto colloids within the waste package will be retarded with respect to the same radionuclides in solution. Because of this, colloid generation and behaviour in the canister is not usually treated explicitly in performance assessments, and was not included in the SITE-94 analysis.

The stability of colloids formed within the waste package will be dependent on pH and ionic strength in such a manner that high pH and high cation concentrations tend to destabilise colloids.

Origin in the repository system:

In the canister, colloid populations are controlled by degradation of the wasteform, metal non-fuel waste parts and the canister itself. In addition, colloids may be formed within the canister due to the presence of chemical and thermal gradients and interfaces, and microbiological activity.

Impact on the repository system:

Colloids can sorb radionuclides thus increasing their apparent solubility. Once sorbed to colloids, radionuclide transport and retardation behaviour can be significantly changed from that of the same radionuclide in solution. Radionuclides bound to colloids formed within the canister are unlikely to be able to travel out to the near-field rock because of the high compaction of the bentonite buffer (filtration). Thus radionuclides bound to colloids within the waste package will be effectively retarded with respect to the same radionuclides in solution.

Bibliographic references:

Kurosawa S, Yui M and Yoshikawa H (1997) Experimental study of colloid filtration by compacted bentonite. Scientific Basis for Nuclear Waste Management, 465, 963-970.

NEA (1993) The status of near-field modelling. NEA/OECD, Paris.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D (1995, editor) The scientific and regulatory basis for the geological disposal of radioactive waste. John Wiley & Sons.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS), 2.1.10 (Biological/biochemical processes and conditions in wastes and EBS) and 3.2.04 (Colloids, contaminant interactions and transport with) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.4 (Colloid generation - source) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-7**

Name: **CORROSION OF THE COPPER SHELL**

Short description:

The canister consists of two parts, a cast iron insert and a corrosion resistant copper shell. The corrosion of the outer copper shell may occur by a number of processes, such as general, localised (pitting), crevice, galvanic and stress corrosion.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

The copper will be of high purity, oxygen-free with Cu contents in excess of 99 wt.% and is chosen because of its generally low reactivity in most copper-water systems and, in pure water, is thermodynamically stable (Möller, 1995). However, in natural groundwaters, copper will corrode by reactions and at rates controlled largely by the nature of dissolved species in the water and the redox potential (Eh). Copper is usually defined in engineering terms as a 'corrosion-allowance' material.

In contact with an oxidising, neutral pH groundwater, copper will be oxidised to form solid copper(II)oxide and any chloride, carbonate, sulphate and hydroxide present may also be incorporated into the oxide phases. These phases will coat the copper surface, forming a passivating layer which will inhibit further corrosion. However, if chloride is also present in high concentrations (e.g. a saline groundwater), then the oxidised copper may form soluble chloride complexes and no passivating layer may form. Thus a saline groundwater may allow for more rapid general surface (uniform) corrosion on the copper shell than would be the case for a dilute (non-saline) groundwater. The corrosion rate then generally increases with the concentration of oxidising species. Oxidising conditions are likely to exist only for a short time after repository closure until free oxygen has been consumed in redox reactions, after which conditions in the near-field will be chemically reducing.

Changes to groundwaters in the near-field may be brought about by climate change (glaciation) and, thus, it is important to evaluate the behaviour of the canister under potential future geochemical conditions (see GEN-22 "Far-field groundwater chemistry"). The SITE-94 performance assessment took account of two different groundwaters in the Central Scenario (i) highly saline, reducing water, and (ii) low salinity, weakly oxidising water.

In a chemically reducing environment with dissolved (reduced) sulphur species present in the groundwater, corrosion would be dominated by sulphide attack. By this mechanism copper sulphide solid phases may form on the canister surface; however, these solids might not form a passivating layer. In this environment, copper sulphide growths ('whiskers' or crystals) could grow from the canister into the buffer because the mobility of copper in the solid copper sulphides is higher than the mobility (diffusion) of dissolved sulphide in the porewater. These growths lead to irregular corrosion of the canister surface. However, the concentration of dissolved sulphides is low in most deep groundwaters and, thus, the corrosion rate is largely controlled by the supply of sulphide diffusing through the bentonite buffer. Under these conditions, the copper shell would be expected to remain intact for at least 100 000 years (Werme, 1998).

Corrosion of copper in the presence of sulphur species can be microbially induced by sulphate reducing bacteria but microbes are not considered to be viable in the bentonite buffer after it is resaturated (see SFL-32 “Microbial activity”).

There is experimental evidence to suggest that copper corrosion rates may be enhanced by the presence of gamma radiation. However, the relatively thick canister should limit the gamma dose rate at the outer canister wall (see SFL-36 “Radiation effects on the canister”). Thus, in the repository, gamma radiation enhanced copper corrosion is not thought to be significant.

If a passivating layer does form on the copper, then any subsequent disruption to that layer may initiate localised corrosion (pitting) which could result in a more rapid perforation of the copper shell than would be predicted by the general corrosion rate. Pitting could also be promoted by micro-scale chemical heterogeneities in the porewater, arising from irregularities in the composition and structure of the bentonite buffer. Appropriate pitting factors (the ratio between pit depths and mean corrosion rates) are difficult to assign and will be strongly dependent on the chemistry of the porewaters. A pitting factor of 25 was adopted in the KBS-3 performance assessment which is thought to be conservative; a lower pitting factor of 5 was used in the SITE-94 assessment which is considered more realistic by comparison with natural analogue observations.

Corrosion of the copper shell may also progress by processes other than general (uniform) or localised (pitting) corrosion. During the manufacturing process, it is possible that the copper metal will develop pores, fissures and irregular grain boundaries of different sizes and shapes that may promote crevice corrosion. Crevice corrosion is a process very similar to pitting and, once initiated, will progress in a similar manner to pit development. Localised corrosion may also be initiated in the vicinity of the weld if corrodants accumulate in the larger grain boundaries that might be associated with the weld.

Copper can also be susceptible to stress corrosion cracking, which is the result of the combined action of corrosion and tensile stress. This stress could be applied by the swelling pressure of the buffer. Copper can corrode by either intergranular or transgranular stress corrosion cracking in the presence of oxygenated ammonia or ammonium containing solutions. In the absence of these species occurring naturally in deep groundwaters, nitrite remaining in the near-field as a consequence of blasting operations during construction may be the most likely cause of stress corrosion cracking of the copper shell.

Galvanic corrosion cells may be initiated by any temperature differences on the surfaces of a copper shell. If this occurs, heterogeneous corrosion rates will be established across the metal surface. In addition, bimetallic (galvanic) corrosion could occur once the copper shell has been perforated and the cast iron insert is exposed to the groundwater. The significance of galvanic corrosion for canister degradation has not yet been resolved. Some studies (e.g. Blackwood and Naish, 1994) suggest that in a reducing environment, galvanic corrosion may not be significant for canister corrosion because, although the process can potentially accelerate corrosion, the formation of the magnetite layer passivates further attack.

Once the copper shell has been perforated, groundwaters will begin to corrode the iron insert and, once that fails, the wasteform (spent fuel). Continued corrosion of the copper, after the wasteform is in contact with groundwater, may be enhanced by corrodants released from the waste (e.g. I, Br, Se and Te). Continued corrosion of the canister will eventually increase the contact area between the waste and the groundwater and thus increase the bulk degradation rate of the waste.

Origin in the repository system:

Corrosion of the copper shell is primarily controlled by the chemistry of the bentonite porewaters, notably the concentrations of dissolved oxygen, chloride and sulphur species. The corrosion rate is limited by the supply of these corrodants and, thus, by the physical properties of the compacted bentonite that limit migration of these species to diffusion; and by the presence of any of these corrodants in the bentonite mineralogy.

Corrosion rates and processes may be further (minimally) modified by gamma radiation fields and microbial populations.

Impact on the repository system:

Corrosion of the copper shell eventually will lead to exposure of the cast iron insert to groundwater and, ultimately, failure of the waste package. Thus corrosion of the copper shell is a primary control on the duration of radionuclide containment.

Corrosion of the copper shell will change the geochemistry of the bentonite porewaters and, consequently, degradation rates and processes for the other engineered barrier system materials.

If large volumes of solid copper alteration products are formed due to corrosion of the canister, then this may cause physical compaction and movement of the bentonite buffer adjacent to the canister. These solid copper alteration products may also react with the bentonite.

Bibliographic references:

AECL (1994) The disposal of Canada’s nuclear fuel waste: engineered barriers alternatives. AECL-10718, COG-93-8.

Blackwood DJ and Naish CC (1994) The effect of galvanic coupling between the copper outer canister and the carbon steel inner canister on the corrosion resistance of the Advanced Cold Process canister. SKB Report, PR 95-04.

Hoch AR and Sharland SM (1994) Assessment study of the stresses induced by corrosion in the Advanced Cold Process Canister. SKB Technical Report, TR 94-13.

Hermansson HP (1995) Some properties of copper and selected heavy metal sulphides: a limited literature review. SKI Technical Report, 95:29.

Möller K (1995) Kopparkorrosion i rent syrefritt vatten. SKI Technical Report, 95:72.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Wersin P, Spahiu K and Bruno J (1994) Kinetic modelling of bentonite-canister interaction: long-term predictions of copper canister corrosion under oxic and anoxic conditions. SKB Technical Report, TR 94-25.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions (in wastes and EBS)) in the NEA International Database (NEA, 1998).

Relates to FEPs 2.1.1 (Chemical reactions: copper corrosion) and 2.1.7 (Pitting) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-8**

Name: **CORROSION OF THE METAL NON-FUEL WASTE PARTS**

Short description:

Complete fuel elements will be placed in the canister and these comprise metallic structural parts in addition to the UO_2 fuel. In contact with groundwater, after canister failure, these metal non-fuel parts will corrode, enhancing groundwater contact with the spent fuel and causing release of the radioactive activation products they contain.

Technical description:

The reference canister will be loaded with complete fuel elements: twelve fuel elements from a boiling water reactor (BWR) or four fuel elements from a pressurised water reactor (PWR). The construction of the BWR and PWR fuel elements are different to each other in detail but similar in basic form (Werme, 1998). A fuel element comprises many (50 to 300) individual fuel rods, each stacked with UO_2 fuel pellets. The fuel rods are made of Zircaloy (zirconium alloy with 98 wt.% zirconium) with welded Zircaloy end plugs to form hermetically sealed units. The fuel rods are fixed together to form single fuel elements by stainless steel and Inconel (nickel alloy with 73 wt.% nickel) spacer grids and upper and lower tie-plates. Because complete fuel elements are loaded in the canister, it means that significant masses of metallic non-fuel components are present in the waste. Average masses (TVO, 1992) of metal in BWR fuel per tonne of uranium fuel are Zircaloy-2, 275 kg/tU; Zircaloy-4, 5 kg/tU; Inconel X-750, 6 kg/tU; and stainless steel, 20 kg/tU.

Corrosion of the metal non-fuel waste parts may begin as soon as the canister is sealed, even if they are constructed and filled without faults. If air or water are trapped in the void spaces within the canister, water, oxygen and nitrogen can cause corrosion of the metal parts by redox reactions (see SFL-10 “Canister corrosion prior to wetting”). These reactions could be limited by purging the canister with inert gas prior to sealing. However, even if this were not done, it is unlikely that significant corrosion could occur prior to canister failure.

Once the outer copper shell and, subsequently, the iron insert have been perforated, groundwater can infiltrate into the canister and contact the fuel elements. Modelling results suggest that the Zircaloy components corrode only very slowly in the chemically reducing environment expected in the near-field at the time of canister failure (Fraker and Harris, 1990). Corrosion of Zircaloy results in the formation of a passivating layer of zirconium oxide alteration products on the metal surface which limits further corrosion. The uniform rate of corrosion is estimated to be 0.01 mm/yr (Garisto and Garisto, 1991) which implies that it would take 10^5 years to breach the thickness of the Zircaloy cladding tube, allowing groundwater to contact the wasteform (spent fuel). However, performance assessments do not take credit for the isolation capacity of the Zircaloy.

Corrosion of the Zircaloy will liberate hydrogen which could cause hydriding of the Zircaloy, resulting in a lowering of its mechanical strength. Some of the Zircaloy fuel rods will already be affected by hydriding when removed from the reactor and, thus, before emplacement in the repository. A build-up of hydrogen pressure within the canister may further act to limit subsequent uniform corrosion but could enhance stress corrosion cracking.

Continued corrosion of the Zircaloy fuel rods will enhance the contact area between the groundwater and the spent fuel, increasing the bulk corrosion rate of the fuel within each container.

Other structural components of the fuel element (spacer grids and tie-plates) made of stainless steel and Inconel are expected to corrode more rapidly than the Zircaloy. However, these components do not form a barrier between the groundwater and the spent fuel. Nonetheless, this process could contribute to an increase in the mechanical stresses on the fuel rods leading to their potential early failure.

The corrosion rate and the nature of the corrosion products formed are affected by the temperature inside the canister (see SFL-46 “Temperature of the near-field”) and the water composition (see SFL-52 “Evolving water chemistry in the canister”), in particular by the presence of carbonate, chloride, sulphate and nitrate. In addition, release of ‘prompt iodine’ from the spent fuel (that proportion of iodine concentrated in gaps and cracks in the fuel pellets which escapes rapidly from the fuel) can induced stress corrosion cracking of cladding and crevice corrosion of spacers/cladding interface.

Once the groundwater is in contact with different metals within the canister, bimetallic (galvanic) corrosion can occur, which may increase the corrosion rate of the less stable metals.

Corrosion of the metal non-fuel waste parts will release the radioactive activation products they contain. At the time of canister failure (taken to be 10^4 years), the activation products calculated to be present (TVO, 1992) in Zircaloy are ^{14}C, ^{36}Cl and ^{93}Zr and, in the other metal structural parts, ^{14}C, ^{59}Ni and ^{94}Nb. In addition, corrosion of the metal non-fuel waste parts will release any radionuclides precipitated or sorbed onto the metal surfaces.

Origin in the repository system:

Corrosion of the metal non-fuel waste parts is primarily controlled by the lifetime of the canister and, when that fails, by the temperature and chemistry of the near-field porewaters, notably the concentrations of dissolved oxygen, carbonate, chloride and sulphur species. The corrosion rate of the metal parts will be enhanced by bimetallic (galvanic) corrosion due to the proximity of the several metal components inside the canister.

Corrosion rates and processes may be further modified (reduced) by the build-up of hydrogen pressure caused by anaerobic corrosion of the metal and by the formation of a passivating layer of solid alteration products on the Zircaloy fuel rods.

Impact on the repository system:

Corrosion of the Zircaloy fuel rods will expose the spent fuel to the groundwater. In addition, corrosion of the Zircaloy fuel rods and the other metal non-fuel waste parts will release the radioactive activation products they contain and any radionuclides precipitated or sorbed onto the metal surfaces. Corrosion of the metal non-fuel waste parts is thus a control on the release rate of radionuclides from the canister.

Corrosion of the metal non-fuel waste parts will affect the chemistry of the near-field groundwaters but not as significantly as that caused by corrosion of the canisters.

Bibliographic references:

AECL (1994) The disposal of Canada's nuclear fuel waste: engineered barriers alternatives. AECL-10718, COG-93-8.

Fraker AC and Harris JS (1990) Corrosion behaviour of zirconium alloy nuclear fuel cladding. Material Research Society, Scientific Basis for Nuclear Waste Management, XIII, 549-556.

Garisto F and Garisto NC (1991) Source term models for the release of radionuclides from used nuclear fuel. Proceedings of near-field workshop. SKB Technical Report, TR 91-59.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

TVO (1992) TVO-92 safety analysis of spent fuel disposal. YJT Technical Report, YJT-92-33-E.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-9**

Name: **CORROSION OF THE CAST IRON INSERT**

Short description:

The canister consists of two parts, a cast iron insert and a corrosion resistant copper shell. Corrosion of the cast iron insert will occur after the copper shell has been perforated and may proceed by a number of corrosion processes, such as general, localised (pitting and crevice), galvanic and stress corrosion.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). When in contact with natural groundwaters, iron is thermodynamically unstable and will corrode by reactions and at rates controlled largely by the nature of dissolved species in the water and the redox potential (Eh).

Corrosion of the iron insert might begin immediately on the inside wall of the canister (inside the canister) and in the annular gap between the insert and the copper shell, even if the canister is constructed and filled without faults (see SFL-10 "Canister corrosion prior to wetting"). If air or water are trapped in the void spaces within the canister, water, oxygen and nitrogen can cause corrosion of the iron by redox reactions. These reactions could be limited by purging the canister with inert gas prior to sealing. However, even if this were not done, modelling results suggest that the volume of entrapped air or water would be too small to cause significant damage to the integrity of the canister. Thus internal corrosion of a sealed iron insert is not considered to compromise repository safety.

Significant corrosion of the outside wall of the cast iron insert can not occur until the copper shell is perforated and groundwater can penetrate. In the absence of manufacturing defects, perforation of the copper shell will occur a long time after closure because of the very slow copper corrosion rates (see SFL-7 "Corrosion of the copper shell"). By the time the copper shell has been perforated, chemical conditions in the near-field will be reducing. Thus it is unlikely that the iron insert will corrode in an oxic environment. Iron corrosion in a chemically reducing environment will be anaerobic and hydrogen will be evolved according to the reaction:

$$3Fe^{\circ} + 4H_2O_{(l)} = Fe_3O_{4\,(s)} + 4H_{2\,(aq)}$$

Experiments suggest that the most probable solid phase will be magnetite (Fe_3O_4) at repository temperatures although, if additional dissolved species are present in the groundwater, other solid alteration products may be formed, e.g. pyrite, siderite or Fe-aluminosilicates. The magnetite will form a passivating layer on the surface of the iron insert, reducing further corrosion to a low, steady-state value until all the iron has been consumed. The magnetite may subsequently react with the near-field porewaters to form a number of possible Fe(III) solid species. Most likely, the first solid would be an amorphous ferric-oxyhydroxide ($Fe(OH)_3$) and, over long periods of time, this may transform to more stable solids such as goethite ($FeOOH$) and haematite (Fe_2O_3).

Potentially very large volumes of hydrogen could be produced (see SFL-25 "Gas generation in the canister") and, because the bentonite is designed with low permeability, this hydrogen cannot easily escape (see SFL-24 "Gas flow through the buffer and backfill"). Some may escape dissolved in the groundwater but it is also possible that hydrogen gas will form in the near-field. A build-up of hydrogen pressure may act further to suppress the corrosion rate. The hydrogen may also lead to embrittlement of the iron which may reduce the structural integrity of the iron insert, although this is not thought to be a serious problem.

If the repository evolution deviates from expected behaviour, it might be possible that the iron insert would corrode in an oxic environment if, for example, the copper shell failed unexpectedly early or there was an ingress of oxygenated water from the surface. In these circumstances, the iron corrosion rate would increase with oxygen concentration, although an Fe-oxide passivating layer would still form.

If a passivating layer does form on the iron, then any subsequent disruption to that layer may initiate localised corrosion (pitting and crevice) which could result in a more rapid perforation of the iron insert than would be predicted by the general corrosion rate. Pitting could also be promoted by micro-scale chemical heterogeneities in the porewater. Appropriate pitting factors (the ratio between pit depths and mean corrosion rates) are difficult to assign and will be strongly dependent on the chemistry of the porewaters.

Dissolved species that may significantly affect corrosion of the iron insert are carbonate, chloride, sulphate, sulphide and nitrate. Carbonates may have a catalytic effect on iron corrosion by acting as proton-donators. In oxidising environments, chloride can act as an initiator for localised corrosion and, in anaerobic systems, passivating oxide layers are less stable in the presence of chloride. Sulphate may act as an oxidising agent in anaerobic environments but is not readily reduced. However, sulphate may be reduced to sulphide by microbial activity, provided that a carbon source is available, causing an oxidation and dissolution of iron. Similarly, nitrate may be reduced by carbon steel in anaerobic solutions.

Galvanic corrosion cells may be initiated by any temperature differences on the surfaces of an iron insert. If this occurs, heterogeneous corrosion rates will be established across the metal surface. Once the copper shell has been perforated and the iron insert is exposed to the groundwater, bimetallic (galvanic) corrosion might occur. A coupling of roughly equal areas of the two metals would lead to a doubling of the iron corrosion rate.

Corrosion of the iron will eventually lead to failure of the insert and of the entire canister. The accumulation of solid alteration products may induce early failure because they will have a larger volume and will thus cause an increase in compressive stresses on the remaining canister components (see SFL-51 "Expansion of solid corrosion products").

Once the copper shell has been perforated, groundwaters will begin to corrode the cast iron insert and, once that fails, the spent fuel. Continued corrosion of the iron insert, after the spent fuel is in contact with groundwater, may be enhanced by corrodants released from the waste (e.g. I, Br, Se and Te). Continued corrosion of the canister will eventually increase the contact area between the waste and the groundwater and thus increase the bulk degradation rate of the spent fuel.

Origin in the repository system:

Corrosion of the cast iron insert is primarily controlled by the lifetime of the copper shell of the canister and, when that fails, by the chemistry of the near-field porewaters, notably the concentrations of dissolved oxygen, carbonate, chloride and sulphur species. The corrosion rate of the iron insert will be enhanced by bimetallic (galvanic) corrosion due to the proximity of the two metal components of the canister.

Corrosion rates and processes may be further modified (reduced) by the build-up of hydrogen pressure caused by anaerobic corrosion of the iron.

Impact on the repository system:

Corrosion of the cast iron insert will lead to the failure of the waste package and contact between the groundwater and the spent fuel. Thus corrosion of the cast iron insert(after perforation of the copper shell) is a primary control on the duration of radionuclide containment.

Corrosion of the cast iron insert will change the geochemistry of the near-field porewaters and, consequently, degradation rates and processes for the other engineered barrier system materials.

If large volumes of solid alteration products are formed due to corrosion of the iron insert, then this may cause differential compressive stresses on the remainder of the canister and accelerate complete canister failure.

Bibliographic references:

AECL (1994) The disposal of Canada’s nuclear fuel waste: engineered barriers alternatives. AECL-10718, COG-93-8.

Arthur R and Apted M (1996) Radionuclide solubilities for SITE-94. SKI Technical Report, 96-30.

Arthur R and Apted M (1996) Modelling of the near-field chemistry for SITE-94. SKI Technical Report, 96-31.

Blackwood DJ, Hoch AR, Naish CC, Rance A and Sharland SM (1994) Research on corrosion aspects of the Advanced Cold Process Canister. SKB Technical Report, TR 94-12.

Hoch AR and Sharland SM (1993) Assessment study of the stresses induced by corrosion in the Advanced Cold Process Canister. SKB Technical Report, TR 94-13.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-10**

Name: **CANISTER CORROSION PRIOR TO WETTING**

Short description:

The void spaces inside the cast iron insert, and between the insert and the copper shell may contain small amounts of air, water or vapour when the canister is initially sealed. As a result, these species may cause some corrosion of the iron insert and the metallic components of the fuel assemblies before the copper shell is perforated and groundwater can infiltrate the canister. The extent of this corrosion prior to wetting is not considered to be significant for repository safety.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

It is likely that the void spaces within the iron insert and in the annular gap between the insert and the copper shell will contain some air, water or vapour when the canister is originally sealed, even in the absence of manufacturing faults. The water is most likely to be present in the fuel pins. Although these will be dried prior to emplacement in the canister, pessimistic assumptions are that one 'wet' fuel pin will be placed in a canister (SKB, 1999).

The air and any small amounts of water vapour contained in the central void spaces can initiate corrosion of the iron insert and the external surfaces of the metallic components of the fuel assemblies before the copper shell is perforated and groundwater can infiltrate the canister.

The water contained in the fuel pins may initially be trapped by the intact Zircaloy cladding tubes. This water cannot be involved in the corrosion of the iron insert until the cladding tubes are breached.

Initially, after canister sealing, the temperature will be high and, thus, water will occur as water vapour (see SFL-46 “Temperature of the near-field”). Thus, in the early stages, oxygen from the trapped air will mean that the atmosphere inside the canister is humid and oxidising. Consequently, the first reactions are likely to be rapid oxidation of the iron, resulting in the formation of iron oxide on the metal surfaces.

Assuming one wet fuel pin and a breached Zircaloy cladding tube, the amount of water inside a canister would be approximately 50 g. If all this water reacted on the inside wall of the iron insert to form FeO, a maximum mass of 150 g of oxide would be created: equivalent to a corrosion depth of 1 μm if distributed equally over the iron insert surface (Werme, 1990). However, under high temperature conditions, localised corrosion 'pitting' is probable. Nonetheless, the amount of water and free oxygen in the void spaces will be quite small and, thus, this reaction will be short lived.

Similarly, air or water in the annular gap between the insert and the copper shell will mean that oxidation of the outer wall of the iron insert will occur. However, the amount of water or air in this space will be much less than that contained in the fuel pin and the significance is correspondingly low.

The water vapour inside the canister will be subjected to a radiation field from the fuel (see SFL-37 “Radiolysis inside the canister prior to wetting”). Radiolysis of the water vapour will generate nitrous and nitric acids (Henshaw, 1994). These species are corrosive to the iron and, thus, the corrosion rate may be accelerated. However, the rapid consumption of water in the iron corrosion processes will limit the possible nitric acid generation rate (Werme, 1990).

After all the free oxygen is consumed by reaction, a high humidity, chemically reducing atmosphere may be present inside the canister and anaerobic corrosion of the iron surfaces by water vapour can commence, which will generate hydrogen. Continued radiolysis in an atmosphere containing hydrogen and nitrogen compounds can generate ammonia. The build-up of corrosive ammonia and nitric acids may lead to stress corrosion cracking of the iron insert locally at points on the canister subjected to tensile stresses (generally the canister is subjected to compressive stresses).

Corrosion prior to wetting of the walls of the iron insert by any of these reactants is not considered to be significant considering the small amount of reactants which may be present (water vapour) or may form (nitric acid) in the void spaces, compared to the very large mass of iron in the canister.

Corrosion prior to wetting of the metallic components of the fuel assemblies may be more significant and it is possible that the fuel cladding may be breached before the entire canister is perforated. However, these processes have not been investigated in any detail by laboratory or modelling studies, and this possibility cannot be quantified. If the fuel cladding was perforated by these processes, it would mean that the cladding would not provide an additional containment barrier after the canister is breached. However, this would not affect calculated releases because the cladding is not assigned any safety role in performance assessments.

All of these reactions could be limited to some extent by more efficient drying of the fuel and by purging the canister with inert gas (e.g. Ar) prior to sealing.

Overall, corrosion prior to wetting will change the local chemical environment inside the canister before it is perforated. Corrosion prior to wetting will also slightly reduce the lifetime of the canister and may possibly result in faster releases from the canister after the canister has been breached (see SFL-49 “Radionuclide release and transport from the canister”). However, all scoping calculations indicate that the volume of entrapped air or water would be too small to cause significant damage to the integrity of the canister. Thus corrosion prior to wetting is not considered to compromise repository safety.

Origin in the repository system:

Corrosion prior to wetting can occur inside a sealed canister. This process is dependent on the introduction of water and air with the fuel assemblies, prior to canister sealing. The amount of species introduced can be limited by drying the fuel and purging the void spaces with an inert gas (e.g. Ar). Procedures for fuel and canister handling are, thus, important controls on this process.

If water and air are present inside the canister, the important controls on the progress of corrosion prior to wetting are the respective amounts of these species, the radiation field inside the canister and the temperature which control the generation of radiolytically-generated corrodants and their state (liquid or vapour).

Impact on the repository system:

Corrosion prior to wetting inside a sealed canister can have minor significance for repository evolution. Corrosion of the inside and outside surfaces of the iron insert may mean that the lifetime of the canister is very slightly reduced.

Corrosion prior to wetting of the metallic components of the fuel assemblies may mean that the cladding becomes perforated, allowing prompt releases (e.g. I) to migrate into the canister cavity before the canister fails. These radionuclides will thus be released immediately the canister is breached.

Bibliographic references:

Henshaw J (1994) Modelling of nitric acid production in the advanced cold process canister due to irradiation of moist air. SKB Technical Report, TR 94-15.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1990) Near-field performance of the advanced cold process canister. SKB Technical Report 90-31.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics), 2.1.03 (Container materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.1 (Stress corrosion cracking) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-11**

Name: **CREEPING OF THE METAL IN THE CANISTER**

Short description:

As a result of their exposure to high temperatures and pressures for long periods of time, the metal components of the canister may experience ‘creep’, which is a slow but continuous plastic deformation. Creep will result in the deformation of the canister and could, in extreme cases, lead to canister failure, although this is extremely unlikely.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

Once emplaced in the repository, the canister will be subjected to high external pressures, of around 14 MPa, from the swelling bentonite buffer and hydrostatic pressures (see SFL-1 “Swelling of the bentonite buffer”). However, maximum pressures could be higher under some circumstances, such as a glacial event (see GEN-21 “Glaciation”). In the first few thousand years after emplacement, the canister will also be subjected to elevated temperatures, with a maximum surface temperature of around 90 °C and an internal temperature in the region of 300 °C, depending on the burn-up and pre-emplacement cooling histories (see SFL-46 “Temperature of the near-field”).

Due to the pressure increase imposed on the canister by the swelling bentonite, it may deform elastically and, at high pressures, plastically. The magnitude of any deformation will be controlled by the load bearing iron insert (see SFL-30 “Mechanical impact on the canister”).

The canister metals may also exhibit creep if the stresses imposed on them are below their yield strength (the point at which significant plastic strain may occur). Creep is a continuous deformation with time in response to the imposition of an external stress. In general, metals only begin to creep at temperatures of around one third of their melting temperature (measured in Kelvin) and, then, the creep behaviour is dependent on the physical properties of the metal concerned, particularly its composition and grain structure (SKB, 1999). The likelihood of creep occurring in the canister metals will be greatest during the period soon after repository closure, during the thermal peak.

Copper is considerably softer (lower yield strength) than iron and, hence, creep will be more apparent in the copper shell of the canister than in the iron insert. Creep of the copper shell will lead to its deformation which will probably result in the migration of the metal into the annular gap between the iron insert and the copper shell of the canister. Ultimately, the annular gap may be totally eliminated (SKI, 1994). This creep may relieve tension stresses imposed on the copper shell during manufacture (Bowyer, 1995).

If the stresses imposed on the canister are not uniform (e.g. due to uneven swelling pressures) then further creep of the copper shell may occur. This would result in a thinning of the copper at the points of highest local stress, and thickening at points with lowest stress.

The welding zones on the copper shell are most sensitive to creep, as a result of the large temperature changes experienced during the manufacture of the canister. Potential creep failure at the welds is thus a possibility if they are subjected to tensile stresses. Furthermore, creep failure is a possibility in canisters with manufacturing defects.

The consequences of creep are thus, localised stress readjustments in the near-field, deformation of the canister and potentially accelerated failure of the copper shell of the canister (see SFL-18 “Failure of the copper shell”).

Origin in the repository system:

Metal creep will be most significant for the copper shell of the canister because copper has a relatively low yield strength. The extent of deformation caused by creep of the copper shell is dependent on the maximum pressures imposed on the canister, the presence of any heterogeneous stress field, the geometry of the annular gap in the canister, the grain structure of the copper (consequence of manufacturing), presence of manufacturing defects in the canister and the temperature.

Impact on the repository system:

Creep of the copper shell will result in localised stress readjustments in the near-field, deformation of the canister and potentially accelerated failure of the copper shell of the canister. However, creep probably is not a major control on final canister failure distributions.

Bibliographic references:

Bowyer WH (1995) Design basis for the copper canister: stage one. SKI Technical report 95:73.

Henderson PJ (1998) Low temperature creep ductility of OFHC copper. Material Science and Engineering, A246, 143.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.07 (Mechanical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 2.2 (Creeping of copper) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-12**

Name: **CRITICALITY**

Short description:

Criticality is the initiation of sustained nuclear (fission) chain reactions in the spent fuel repository. It may occur if a sufficient mass and density of fissile material can accumulate in one place. Criticality, if it were to occur, would affect the radionuclide inventory and the thermal output and, in extreme cases, may damage the integrity of the engineered barriers and the rock mass.

Technical description:

Spent fuel contains a number of fissile radionuclides, the two most important for criticality considerations are ^{239}Pu and ^{235}U. Although significant masses of these nuclides are present in the repository inventory, careful design of the repository geometry will ensure that these nuclides are distributed throughout the near-field in subcritical masses. Criticality is, thus, impossible under normal repository evolution while the canisters remain intact.

Criticality would theoretically be possible if a leak occurred in the canister after disposal and water filled the void space. The water would act as a neutron moderator, reducing the energies of neutrons emitted during fission of ^{239}Pu and ^{235}U from their original high values to thermal energies before they escape from the canister, thus promoting criticality. The Swedish disposal concept includes the requirement on canister design that there is no risk of criticality in the event of water entering the canister.

The key factors that determine whether criticality will occur are the original enrichment of the fuel, the burn-up of the fuel, the time after emplacement which controls the concentration of fissile nuclides in the waste, the presence of a burnable poison (such as gadolinium), the presence of enough fuel to create a critical mass, and the geometric configuration of the fuel to provide sufficient void space.

Whether or not a system is critical is generally expressed in terms of an effective multiplication factor, k_{eff}. A system is subcritical if $k_{eff} < 1$ but criticality analyses include a safety margin, typically requiring $k_{eff} < 0.95$ (Hicks and Green, 1999). Calculations have shown that criticality cannot occur in the reference canister design, even after canister failure, provided the fuel has experienced a normal burn-up history (Hicks and Prescott, 2000).

Plutonium criticality within the canister would call for selective dissolution and transport of uranium out of the canister before most of the plutonium has decayed to ^{235}U, i.e. within approximately 10^5 years after disposal, which is considered very unlikely to occur.

Due to the theoretical possibility of criticality within a canister, the reference canister design for the spent fuel repository is configured to reduce the internal void space and, thus, the potential for criticality. This reference design comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). The cast iron insert is formed with individual channels for the fuel elements which keep the fuel elements separated. Different internal configurations for the canister are possible to house PWR and BWR fuel elements. The internal canister design avoids the potential for the accumulation of fuel pellets in the bottom of the canister after the Zircaloy fuel element cladding is degraded. Calculations suggest criticality cannot occur in the reference canister design, even after canister failure, provided the fuel has experienced a normal burn-up history (Werme, 1998).

Criticality could potentially occur outside of the canister if dissolution, transport and precipitation processes act to remove fissile material (uranium or plutonium) from the canister and reconcentrate it elsewhere (see GEN-31 “Radionuclide reconcentration”), such as in the buffer material or in fractures in the near-field rock. In-growth from the decay of ^{239}Pu will increase the fraction of ^{235}U to a level where criticality is theoretically possible but uranium from several canisters would need to accumulate in one location for a critical mass to form. However, this event is considered extremely unlikely in a normal repository evolution scenario due to the very low solubilities of uranium and plutonium in the near-field chemical environment, and the fact that transport and reconcentration processes would have to take place by slow diffusional processes.

If criticality did occur in the repository near-field, it would initiate chain reactions which would generate new fission products and, possibly, transuranic elements if neutron capture occurs. As a consequence, the inventory and activity of fission products and transuranic elements, and the total activity of the waste will increase during the period of a criticality event (see SFL-2 “Changes in the spent fuel radionuclide inventory”). The increase in the fission product inventory of a repository due to criticality would be system specific but analyses suggest that it would be low in most cases. The criticality event would be moderated by the groundwater and may be self-sustaining until the supply of fissile fuel was reduced to below the mass required to sustain reaction. Natural analogues to a repository criticality event are the natural fission reactors at Oklo, Gabon (Blanc, 1996).

The heat emitted by the fission reactions would change the thermal output from the repository and, in extreme cases, this might damage the integrity of the engineered barriers by impacting on the stresses in the rock mass (see GEN-36 “Stress field”).

Origin in the repository system:

Criticality theoretically could occur in the repository but consideration of the necessary sequence of events required to initiate criticality make it an extremely unlikely event in a normal repository evolution scenario.

Criticality depends on the concentration of fissile radionuclides, the amount and geometry of the fissile material and the availability of a moderator (groundwater). Criticality could only occur, therefore, after the canister has failed and, due to radioactive decay reducing the concentration of fissile radionuclides, the expected long life of the canister makes the likelihood of criticality very remote.

Because criticality is considered very unlikely, it is not considered in the main repository evolution scenarios in performance assessments for spent fuel repositories (e.g. the SITE-94 performance assessment).

Impact on the repository system:

If criticality did occur in the repository, it would impact on the radionuclide inventory because the chain reactions would generate new fission products and, possibly, transuranic elements if neutron capture occurs.

The heat emitted by the fission reactions would change the thermal output and, in extreme cases, this may damage the integrity of the engineered barriers and the rock mass.

Bibliographic references:

Blanc PL (1996) Oklo - natural analogue for a radioactive waste repository (Phase 1). EC Nuclear Science and Technology Report, EUR 16857/1.

Hicks T and Green TH (1999) A review of the treatment of criticality in post-closure safety assessment for radioactive waste disposal. UK Environment Agency, R&D Technical Report, P222.

Hicks T and Prescott A (2000) A study of criticality in a spent fuel repository based on current canister designs. SKI Technical Report, 00:13.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Oversby VM (1996) Criticality in a high level waste repository - a review of some of important factors and an assessment of the lessons that can be learned from the Oklo reactors. SKB Technical Report, TR 96-07.

Rechard RP, Tierny MS, Sanchez LC and Martell MA (1997) Bounding estimates for criticality events when directly disposing of highly enriched spent nuclear fuel in unsaturated tuff. Risk Analysis, 17, 19-35.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 2.1.14 (Nuclear criticality) in the NEA International Database (NEA, 1998).

Relates to FEP 1.1.1 (Criticality) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-13**

Name: **DEGRADATION OF THE SPENT FUEL ELEMENTS**

Short description:

The spent fuel pellets will be subjected to a variety of physical and chemical processes which will cause their degradation over time. This degradation may increase the surface area exposed to groundwater and thus may increase the bulk dissolution and radionuclide release rates. Depending on the geometry of the fuel assemblies and the canister design, fragments or whole of fuel pellets may come loose and accumulate in the canister void spaces.

Technical description:

The spent fuel pellets will not be intact when they are loaded into the canister. The thermal history experienced in the nuclear reactors will have caused cracking and fragmentation of the pellets, and this is controlled by the burn-up history.

In the repository after emplacement, when the canister is intact, dissolution and degradation of the spent fuel UO_2 matrix will be limited to reaction with the small amounts of air or water trapped in the void spaces within the canister (see SFL-10 “Canister corrosion prior to wetting”). These reactions could be minimised by purging the canister with inert gas prior to sealing. However, even if this were not done, the very small amounts of water which are likely to be present in a sealed canister will mean that degradation prior to canister failure will be negligible.

After failure of the canister and the Zircaloy cladding, groundwater will come into direct contact with the surface of the spent fuel and will cause the fuel to degrade by dissolution and conversion reactions (see SFL-21 “Spent fuel dissolution and conversion”). Destruction of the metal components of the fuel assembly, especially the Zircaloy cladding tubes, will expose large spent fuel surface areas to groundwater and this can increase the bulk dissolution and radionuclide release rates, and the extent of radiolysis (see GEN-30 “Radiolysis”). Cracking and fragmentation of the fuel will also create additional paths through which I and Cs could migrate from the fuel matrix to the surface (see SFL-27 “Radionuclide accumulation at the spent fuel surface”).

The fuel pellets could also could be altered and degraded by high-energy alpha particles and by decay processes. Local pressure build-up of helium from radioactive decay and hydrogen from radiolysis could cause creep and cracking of the Zircaloy cladding, and fragmentation of the fuel. Furthermore, the integrity of the cladding could be compromised by corrosion and by gamma and neutron flux from decaying radionuclides in the fuel.

Continued degradation of the spent fuel, particularly along the lines of pre-existing cracks and fractures will result in extensive mechanical damage to the fuel pellets. At the same time, continued destruction of the Zircaloy cladding tubes may mean that fragments of the fuel pellets may fall free from the assemblies and accumulate in the void spaces inside the canister. If this were to happen to any extent, then there could be the risk of a criticality event inside the canister (see SFL-12 “Criticality”). To limit the potential for this happening, the canister is designed with individual channels for the fuel elements to keep them separated and to reduce the internal void space and, thus, the potential for criticality (Werme, 1998).

Origin in the repository system:

Degradation of the fuel pellets will occur as a consequence of the dissolution and conversion of the fuel by groundwater, by radioactive decay and by high energy particles. The extent of degradation will be controlled by the number and geometry of pre-existing cracks in the pellets and by the rates of the spent fuel degradation.

Impact on the repository system:

Degradation of the fuel pellets can increase the surface area available for dissolution, conversion and radionuclide release and, thus, the total release rate from the fuel can be enhanced.

Mechanical damage can fragment the fuel pellets and they may fall free and accumulate in the canister void spaces. However, the reference canister is design in such a way as to avoid a criticality event from this mechanism.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in the wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 1.5 (Release of radionuclides from the failed canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-14**

Name: **DIFFERENTIAL THERMAL EXPANSION AND CONTRACTION OF THE NEAR-FIELD BARRIERS**

Short description:

The variety of materials in the near-field barriers will have different thermal expansion coefficients. Thus, when the temperature of the near-field changes due to the radiogenic heat output, the barriers may expand or contract at different rates, causing changes to the stresses acting on them. Spatial variation in temperature in the near-field will also cause differential expansion. These processes may cause minor physical effects on some barriers but their significance is thought to be small.

Technical description:

The primary material used in the near-field barriers are iron and copper in the canister, and bentonite in the buffer and backfill (mixed with sand or crushed rock in the backfill). In addition, groundwater and some gas will be present in all the void spaces in the near-field.

These different materials all posses different thermal expansion coefficients. Thus, as the near-field is heated by the radiogenic heat output from the spent fuel (see SFL-46 “Temperature of the near-field”), these materials will expand by different amounts. The most significant process will be the thermal expansion of the groundwater in the bentonite buffer porespaces. The thermal expansion of water is about 100 times larger than the thermal expansion of bentonite mineral particles, and dominates this process (SKB, 1999). Likewise, the thermal expansion of any separate gas phase may also contribute to the process.

The impact of the thermal expansion of the water on the near-field is dependent on whether the near-field is hydraulically saturated and the bentonite has swelled (see SFL-1 “Swelling of the bentonite buffer”). Before complete water saturation, thermal expansion of water is easily taken up by expansion into the air filled void spaces. However, after saturation, continued thermal expansion of the bentonite porewater must result in an increase in the porewater pressure as the volume is essentially constrained. This pressure rise causes the buffer to expand further upwards pushing into the tunnel backfill and for some of the porewater to be expelled into the rock. Although the porosity and water content of the backfill is larger than that of the buffer, the proximity of the buffer to the heat from the canister means that the thermal expansion is greatest in the buffer.

Thermal expansion of the canister metals will also occur causing the canister to expand outwards, providing further pressures on the bentonite buffer. However, this is not expected to have a significant effect and is much smaller than the effect of the expansion of the bentonite porewater.

Thermal expansion and differential thermal expansions will change the stresses acting on the engineered barriers and the rock. This may cause minor physical effects on some barriers but the significance of these processes is thought to be small (see GEN-36 “Stress field”).

The increased stresses will change the hydraulic conductivity of the buffer and the backfill. However, the extra pressures caused by thermal expansion will be small in comparison to the swelling pressure generated by the adsorption of water into the bentonite mineral (see SFL-1 “Swelling of the bentonite buffer”).

As the thermal pulse subsides, the near-field will cool allowing the various materials in the engineered barriers to contract. Thermal contraction will result in volume reduction (shrinkage) and a reduction in the near-field stresses. This will impact on the hydraulic conductivity of the rock, the buffer and the backfill (see SFL-20 “Groundwater flow through the buffer and backfill”).

The rate of thermal expansion and contraction will be controlled by the thermal properties of the different materials, the thermal pulse from the spent fuel and the natural geothermal gradient which combine to control the temperature in the near-field (see SFL-46 “Temperature of the near-field”).

Origin in the repository system:

Differential thermal expansion of the near-field materials will occur due to differences in the thermal expansion coefficients of these materials and spatial variations in the temperature field. The extent of thermal expansion is controlled by the temperature rise in the near-field which is generated by radiogenic heat from the spent fuel. As the radiogenic heat output from the waste diminishes, the near-field barriers may contract. This contraction will reduce pressures and stresses in the near-field, and will affect the groundwater flow field.

Impact on the repository system:

Thermal expansion of the near-field materials will be greatest for the groundwater in the bentonite pore spaces. Once the buffer is saturated, expansion of the groundwater will cause the porewater pressure to rise, forcing the buffer upwards in to the tunnel backfill and expelling some of the water to the rock. Other materials have much lower thermal expansion coefficients and, thus, have little effect on the near-field.

Thermal expansion and contraction of the near-field materials will affect the stresses and pressures in the near-field. The hydraulic conductivity of the buffer could be affected.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.11 (Thermal processes and conditions in wastes and the EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 4.2.7 (Thermo-hydro-mechanical effects) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-15**

Name: **DIFFUSION IN AND THROUGH THE CANISTER**

Short description:

Radionuclides released from the spent fuel can migrate from inside the canister to the bentonite buffer, after the canister has been breached, by diffusion. In addition, dissolved species in the groundwaters outside the canister can be transported into the canister by diffusion. Both inward and outward diffusion of species will affect the groundwater chemistry (radionuclide content) inside the canister and in the buffer, and the release from the fuel of solubility controlled species.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

No inward or outward diffusion from the canister can occur until after the copper shell and, subsequently, the iron insert are perforated and a continuous transport path is established. Once the canister is breached, groundwater can enter the central void spaces of the canister and radionuclides can begin to be released by dissolution and conversion of the spent fuel (see SFL-40 “Radionuclide release from spent fuel matrix”) and from the metallic non-fuel components (see SFL-41 “Radionuclide release from metal non-fuel parts”). Radionuclides released from the fuel and metal parts may go directly to solution, if they are soluble, or may be incorporated into newly formed separate mineral phases (e.g. uranyl silicates) or co-precipitates, or sorbed onto canister corrosion products (see GEN-34 “Radionuclide sorption”).

The soluble radionuclides may migrate out of the canister to the bentonite buffer by diffusional processes. At the same time, inward diffusion to the canister of dissolved species in the bentonite porewaters can occur. Both the inward and outward diffusional processes will be driven by the respective concentration gradients across the void spaces inside the canister and in the buffer (see GEN-9 “Diffusion”). The rate of these diffusional processes will thus be controlled by the supply of dissolved species to form the concentration gradient (e.g. by the radionuclide release rate from the spent fuel). In addition, the rate of diffusion will be temperature dependent and, thus, will slow with time as the radiogenic heat output decreases (see SFL-46 “Temperature of the near-field”). Diffusion inside the canister will also be partly controlled by the action of the Soret effect which drives diffusion of chemical species under the influence of a thermal gradient, and couples the concentration transport to the temperature gradient (see SFL-44 “Soret effect in the buffer and backfill”). The actual mass transport rate of dissolved species by diffusion in and out of the canister will also be dependent on the number, size and geometry of perforations in the canister walls.

Transport of dissolved species may also occur in the canister void spaces by water turnover driven by thermal convection (see SFL-56 “Water turnover in the copper shell” and SFL-57

“Water turnover in the cast iron insert”). The diffusional processes inside the canister will be partly coupled to this water turnover because water turnover will perturb the concentration gradients inside the canister and, thus, will affect the diffusional driving forces.

Diffusional transport through the canister will affect the groundwater composition (radionuclide content) inside the canister and in the bentonite buffer, and the total release rate from the canister (see SFL-49 “Radionuclide release and transport from the canister”).

The removal of dissolved uranium and other radionuclides from the canister waters by diffusion can enhance further dissolution of the spent fuel because the release of uranium from the spent fuel matrix is solubility limited.

Origin in the repository system:

Diffusion of dissolved species into and out of the failed canister is largely driven by the concentration gradient (and hence by the supply of dissolved species, e.g. by the radionuclide release rate from the spent fuel) but is also partly controlled by the temperature gradient.

Diffusion will also be affected by the size of perforations in the canister wall and the turnover of water in the canister caused by thermal convection.

Impact on the repository system:

Diffusion of dissolved species into and out of the failed canister will affect the groundwater chemistry inside the canister and in the buffer, and will affect the total radionuclide release rate from the canister.

Diffusion of radionuclides will also affect the spent fuel dissolution rate, because this dissolution rate is controlled by the solubility limited behaviour of uranium in the groundwater adjacent to the fuel.

Bibliographic references:

Crank J (1975) The mathematics of diffusion (2nd edition). Oxford University Press.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 3.2.07 (Water-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.6 (Diffusion - surface diffusion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-16**

Name: **DILUTION OF THE BUFFER AND BACKFILL**

Short description:

Extensive expansion of the buffer or backfill will reduce its swelling pressure and, hence, increase its hydraulic conductivity. This could occur if the buffer or backfill swells into large voids in a failed canister or in the near-field rock, or if it is eroded. If dilution were to occur, the potential for groundwater or gas flow to occur through the buffer and backfill would increase, and radionuclide transport would be enhanced.

Technical description:

The diffusional barrier provided by the buffer and backfill materials is directly dependent on the maintenance of a high swelling pressure, creating low hydraulic conductivities. Initially, after the buffer and backfill have first hydraulically resaturated, the bentonite they contain will have swollen to fill all void spaces in the disposal holes and tunnels, and any intersecting fractures in the near-field rock, creating very high swelling pressures (see SFL-1 “Swelling of the bentonite buffer”). Over time, a number of processes may occur which would cause the buffer and backfill materials to become diluted, which would cause their swelling pressures to reduce.

The most likely process causing dilution of the bentonite buffer is failure of the canisters (see SFL-18 “Failure of the copper shell” and SFL-19 “Failure of the cast iron insert”). The reference canister design for the spent fuel repository contains a number of voids: the annular gap between the copper shell and the iron insert, and the central void spaces inside the iron insert (Werme, 1998). When the copper shell and, subsequently, the iron insert are breached, the very high swelling pressures in the bentonite (around 14 MPa) may cause the bentonite to intrude into the canister voids. The likelihood of this process occurring depends, in part, on the size of the perforations in the canister. Takase et al. (1998) suggest that bentonite intrusion would occur for perforations wider than 0.1 mm. The amount of bentonite which would intrude would be dependent on the iron corrosion processes forming an opposing gas pressure and expanding iron corrosion products inside the canister (see SFL-51 “Expansion of solid corrosion products”). The maximum volume of bentonite which could intrude is approximately 1 m^3, which is the void space inside the iron insert of the canister (Werme, 1998): actual intruded volumes would be expected to be less than this.

The initial swelling pressures in the backfill will be less than those in the buffer (lower bentonite content in the backfill). This means that the buffer will swell and expand upwards into the tunnel backfill, until a point when the pressures have equilibrated. This process will result in a net dilution of the buffer and a net concentration of the backfill. The extent of the dilution of the buffer by this process is controlled by the relative compositions (bentonite contents) of the buffer and backfill, and any retarding forces caused by friction between the disposal hole walls and the buffer.

Dilution of both the buffer and the backfill could occur if they expand into large fractures intersecting the disposal holes or tunnels. During repository operation, any large fractures should be sealed and, hence, excessive expansion into fractures would occur only after inadequate repository operation procedures or, alternatively, if large fractures open after the repository is closed (e.g. due to changes in the near-field rock stress). In any event, it is unlikely that very large voids could be created by these mechanisms.

Similarly, dilution of both the buffer and the backfill could occur due to inadequate repository operation procedures, if they resulted in incomplete emplacement of buffer or backfill materials, leaving behind large, unfilled voids.

Dilution of both the buffer and the backfill may also occur by erosion of these materials by fast groundwater flow at the interface with the near-field rock or due to a reduction in groundwater salinity which will tend to encourage the bentonite to form a suspension in the groundwater (see SFL-17 “Erosion of the buffer and backfill”). However, both of these process are likely only to be significant during unusual, transient events and not under the normal conditions expected in stable rock at repository depth.

Dilution of the buffer or backfill, by any process, will result in a reduction of the material swelling pressure and a corresponding increase in the hydraulic conductivity, although these affects may be localised. This would have a number of impacts on the performance of the repository. Most significantly, the potential for groundwater and gas flow to take place through the buffer and backfill would be increased (see SFL-20 “Groundwater flow through the buffer and backfill” and SFL-24 “Gas flow through the buffer and backfill”). This might then increase the rate of canister corrosion and, subsequently, radionuclide transport. The reduction in the density of the buffer would also limit this materials ability to provide mechanical protection to the canister.

Origin in the repository system:

Dilution of the buffer and backfill will depend on the potential for these materials to expand into void spaces created either by failure of the canister, expansion of fractures in the near-field rock or removal (erosion) of these materials by groundwater. As a consequence, dilution of the buffer and backfill is dependent on the changing groundwater flow conditions, changing groundwater chemistry and changing near-field stresses.

Impact on the repository system:

Dilution of the buffer and backfill will cause changes to the physical and hydraulic properties of these materials. In particular, their swelling pressures will be reduced and their hydraulic conductivities will be increased. This might increase the potential for gas and water flow (and radionuclide transport) to occur through the buffer and backfill.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Takase H, Benbow S and Grindrod P (1998) Mechanical failure of SKB spent fuel disposal canisters: mathematical modelling and scoping calculations. SKB Technical Report, TR 99-34.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.1.1 (Swelling of bentonite into tunnels and cracks) and 3.2.4 (Erosion of buffer/backfill) in SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-17**

Name: **EROSION OF THE BUFFER AND BACKFILL**

Short description:

Particulates in the buffer and the backfill could be removed by flowing groundwaters at the interfaces with the near-field rock, adjacent to disposal holes or tunnels, or due to a reduction in groundwater salinity. This would create void spaces which would be filled as the remaining buffer or backfill materials expand into them, resulting in a decrease in their swelling pressures. Eroded particulates may transport sorbed radionuclides or may clog hydraulically-active fractures, altering groundwater flow paths in the near-field rock.

Technical description:

After the buffer and backfill have hydraulically resaturated, the bentonite they contain will have swollen to fill all void spaces in the disposal holes and tunnels, and any intersecting fractures in the near-field rock (see SFL-1 “Swelling of the bentonite buffer”). Under certain unusual situations, solid particulates from the outer parts of the buffer and backfill materials, adjacent to the near-field rock, may be eroded by mobilisation and transport processes involving the groundwater.

This could occur if the groundwater flow rates increase through the near-field rock, causing the buffer and backfill materials to be washed away. This is not considered to be a significant process under the normal low-flow conditions expected in deep, stable crystalline rocks. However, due to certain transient disturbances (e.g. seismicity, or the advance or retreat of a glacier), groundwater flow rates and patterns through the far and near-field rocks could be significantly altered (see GEN-23 “Groundwater flow”). In these cases, it is possible that the erosion of the buffer and backfill may become significant. Mechanical erosion would initially be localised to the bentonite which has extruded furthest into the fractures, where it is least confined, or to bentonite at the interface with the near-field rock adjacent to hydraulically-active fractures (Le Bell, 1978).

An alternative erosion mechanism involves significant changes to the groundwater chemistry. Normally, groundwaters in deep, stable crystalline rocks would be expected to have an elevated ionic strength or salinity (see GEN-22 “Far-field groundwater chemistry”). However, in certain situations (e.g. due to deep flushing of groundwaters beneath a retreating glacier), the groundwater in the near-field rock may have a very low ionic strength. In low ionic strength waters, bentonite tends more easily to form a suspension. If this were to occur in the repository near-field, erosion of the buffer and backfill materials adjacent to the near-field rock could be initiated. Individual suspended particulates may be sufficiently small to diffuse away or to be transported by flowing groundwater.

The mechanisms which might cause a change to the groundwater chemistry in the near-field rock (e.g. glaciation) are also the mechanisms which might cause an increase in the groundwater flow rate. In this case, erosion of the buffer and backfill may be doubly affected.

Erosion of the buffer and backfill would initially be localised to the material which has extruded furthest into the fractures or occurs at the intersections of hydraulically-active fractures with the disposal holes or tunnels, where it is least confined. Erosion of the backfill may be more likely than erosion of the buffer because of the lower swelling pressure and higher hydraulic conductivity of the former. In particular, the backfill near the top of the tunnels, where the density is lowest, is most prone to erosion (SKB, 1999).

The potential for erosion of the buffer and backfill materials may be altered by any preceding physical or chemical processes. For example, uneven swelling during hydraulic resaturation may lead to localised regions in the buffer or backfill with lower swelling pressures which may favour groundwater access and erosion (see SFL-1 “Swelling of the bentonite buffer”). In contrast, cementation of the buffer and backfill materials may inhibit erosion by more strongly binding together the constituent components (see SFL-3 “Chemical alteration of the buffer and backfill”).

Significant erosion of the buffer and backfill materials would cause a reduction in the swelling pressure of the remaining material, although the affect may be localised. This would also affect the potential for coagulation of the bentonite (see SFL-4 “Coagulation of bentonite”). The overall effect might be to change the potential for groundwater and gas transport through the buffer and backfill (see SFL-16 “Dilution of the buffer and backfill”).

Eroded particulates would act as colloids and, thus, radionuclide transport through the far-field rock could be enhanced by the mobilisation of bentonite particulates onto which radionuclides may be sorbed (see GEN-4 “Colloid behaviour in the host rock”). However, eroded particulates may clog hydraulically-active fractures, reducing groundwater flow through them and modifying the groundwater flow patterns through the near-field rock. Whether such processes would increase or decrease the potential for radionuclide transport is uncertain.

Origin in the repository system:

Erosion of the buffer and backfill materials is not a significant process during normal repository conditions. However, during certain unusual transient events, erosion may occur. Mechanical erosion is controlled predominantly by the groundwater flow rate through the near-field rock, and is localised at sites where the flowing water is in contact with the furthest reaches of the buffer or backfill, which are least confined.

Erosion can be initiated by a change in groundwater chemistry to low salinity conditions, under which the bentonite may readily form a suspension. This will be controlled by the composition of the groundwater in the far-field rock.

Impact on the repository system:

Erosion of the buffer and backfill materials may affect the physical conditions in the remaining materials, lowering the swelling pressure and raising the hydraulic conductivity. Eroded particulates may act as colloids and, potentially, can increase radionuclide transport through the near-field rock. Mobile particulates may clog fractures and modify groundwater flow patterns in the near-field rock.

Bibliographic references:

Le Bell JC (1978) Colloid aspects of the confined bentonite concept. KBS Technical Report, 97.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.4 (Erosion of buffer/backfill) in SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-18**

Name: **FAILURE OF THE COPPER SHELL**

Short description:

The outer copper shell of the canister provides long canister lifetimes because of the very slow corrosion rate of copper in the repository environment. However, corrosion will eventually cause the canister to fail, although early failure could be caused by manufacturing defects, mechanical impacts or creep of the copper. Failure of the copper shell exposes the cast iron canister insert to groundwaters and, thus, initiates and enhances corrosion of the iron insert.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

The copper will be of high purity, oxygen-free with Cu contents in excess of 99 wt.% and is chosen because of its generally low reactivity in most copper-water systems and, in pure water, is thermodynamically stable (Möller, 1995). However, in natural groundwaters, copper will corrode by reactions and at rates controlled largely by the nature of dissolved species in the water and the redox potential (Eh). The most important corrosion mechanism would be sulphide attack from dissolved (reduced) sulphur species present in the groundwater (see SFL-7 “Corrosion of the copper shell”). Dissolved sulphides corrode the copper forming copper sulphide and hydrogen. However, the concentration of dissolved sulphides is low in most deep groundwaters and, thus, the corrosion rate is largely controlled by the supply of sulphide diffusing through the bentonite buffer. Under these conditions, the copper shell would be expected to remain intact for at least 100 000 years (Werme, 1998).

Premature failure of the copper shell could be caused by mechanical impacts due to the numerous combined external forces acting on the canister (see SFL-30 “Mechanical impact on the canister”). External impacts on the copper shell are due to the combined hydrostatic and swelling pressure imposed by the bentonite buffer, which can total 14 MPa. The pressure may be uneven around the canister due to uneven swelling which may result in tensile stresses acting on some parts of the canister surface. Additional external forces may arise due to rock mass movement by faulting (see GEN-17 “Faulting”), creep of the rock mass (see GEN-5 “Creeping of the rock mass”) and collapse of the repository openings (see GEN-3 “Cave-in”). However, the buffer will tend to protect the canister from these events to some extent.

The copper shell will also experience metal creep at the near-field temperatures (see SFL-11 “Creeping of the metal in the canister”). Creep of the copper shell will lead to its deformation which will probably result in the migration of the metal into the annular gap between the iron insert and the copper shell of the canister. Ultimately, the annular gap may be totally eliminated (SKI, 1994). If the stresses imposed on the canister are not uniform (e.g. due to uneven swelling pressures) then further creep of the copper shell may occur which could result in a thinning of the copper at the points of highest local stress which could locally reduce the metal strength (see SFL-39 “Reduced mechanical strength of the canister”).

Creep or mechanical impacts (e.g. due to uneven swelling pressures) may cause localised tensile stresses on the copper shell. In the presence of the sulphide containing groundwater, this would initiate stress corrosion cracking of the copper (see SFL-7 “Corrosion of the copper shell”).

The mechanism and timing of failure of the copper shell will be controlled thus by the chemistry of the groundwater adjacent to the copper, the temperature and the local stress field. Radiation and microbial activity could also have minor impacts on failure. Manufacturing defects may also lead to accelerated failure.

The geometry of perforations on the canister will be dependent on the mode of failure and the stress field acting on the canister. When the canister fails, the size and location of these perforations may induce further mechanical failure, and will affect the ingress of groundwater, the initiation of corrosion of the iron insert, the outflow of gas and the transport of radionuclides to the bentonite buffer.

Origin in the repository system:

Failure of the copper shell of the canister is most likely to occur as a result of sulphide attack on the copper. Thus, the timing of failure is dependent on the groundwater chemistry and the supply of sulphide through the bentonite buffer.

Early failure of the copper shell could be caused by mechanical impact on the canister due to uneven swelling pressures, movement of the rock or creep of the copper. If these resulted in localised tensile stresses on the canister, then stress corrosion cracking could be initiated.

Impact on the repository system:

Failure of the copper shell of the canister will expose the iron insert to groundwaters and, thus, will initiate corrosion of the iron. The mode of failure will control the geometry of perforations and, in turn, the inflow and outflow of water and gas, and the release of radionuclides to the bentonite buffer.

Bibliographic references:

Möller K (1995) Kopparkorrosion i rent syrefritt vatten. SKI Technical Report, 95:72.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.07 (Mechanical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 2.1.1 (Chemical reactions - copper corrosion) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-19**

Name: **FAILURE OF THE CAST IRON INSERT**

Short description:

The cast iron insert of the canister provides mechanical strength. It is protected by the long-lived copper shell but, eventually, corrosion of the copper and iron components will eventually cause the entire canister to fail. Early failure of the iron insert could be caused by manufacturing defects or mechanical impacts on the canister. Failure of the iron insert exposes the fuel elements to groundwater, and initiates release of radionuclides from the canister.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

Significant corrosion of the cast iron insert cannot occur until the copper shell is perforated, allowing groundwater to penetrate into the annular gap between the insert and the copper shell (see SFL-18 "Failure of the copper shell"). However, some minor corrosion can occur before complete canister failure, caused by any residual water contained in the fuel pins (see SFL-10 "Canister corrosion prior to wetting").

Once the copper shell is perforated, anaerobic corrosion of the outer wall of the iron insert will begin, creating iron oxides and hydrogen (see SFL-9 "Corrosion of the cast iron insert"). The continued corrosion mechanism is complicated by a number of coupled processes, such as the build-up of hydrogen and iron oxides in the annular gap, which will limit further water access to the iron, and the possible intrusion of bentonite under pressure into the gap. These processes are also partly controlled by the geometry and size of the perforations in the copper shell. Eventually, however, progressive corrosion will cause the iron insert to fail, leading to failure of the entire canister.

Premature failure of the iron insert could be caused by mechanical impacts due to the numerous combined external and internal forces acting on it (see SFL-30 "Mechanical impact on the canister"). Creep of the copper shell may cause total elimination of the annular gap (SKI, 1994), leading to direct transfer of external forces from the copper shell to the iron insert (see SFL-11 "Creeping of the metal in the canister"). External impacts are due to the combined hydrostatic and swelling pressure imposed by the bentonite buffer, which can total 14 MPa. The pressure may be uneven around the canister due to uneven swelling which may result in tensile stresses acting on some parts of the iron insert. The accumulation of solid alteration products may also induce early canister failure because they will have a larger volume and will thus cause an increase in compressive stresses on the iron insert and tensile stresses on the copper outer canister (see SFL-51 "Expansion of solid corrosion products").

Additional external forces may arise due to rock mass movement by faulting (see GEN-17 “Faulting”), creep of the rock mass (see GEN-5 “Creeping of the rock mass”) and collapse of the repository openings (see GEN-3 “Cave-in”). However, the buffer will tend to protect the canister from these events to some extent. The most significant internal force on the canister will be the internal (gas) pressure which is largely due to hydrogen production by anaerobic iron corrosion (see SFL-25 “Gas generation in the canister”). This internal pressure is controlled by the balance of the rates of gas production and gas loss from the canister.

The mechanism and timing of failure of the iron insert will be controlled initially by the lifetime of the copper shell and then by the chemistry of the groundwater penetrating the copper shell, the temperature and the local stress field. Radiation and microbial activity could also have minor impacts on failure. Manufacturing defects may also lead to accelerated failure.

The geometry of perforations on the canister will be dependent on the mode of failure and the stress field acting on the canister. When the canister fails, the size and location of these perforations may induce further mechanical failure, and will affect the ingress of groundwater, the initiation of corrosion of the spent fuel, the outflow of gas and the transport of radionuclides to the bentonite buffer.

Origin in the repository system:

Significant corrosion of the iron insert cannot commence until the copper shell has been perforated. Failure of the insert is then most likely to occur as a result of anaerobic corrosion of the iron.

Early failure of the iron insert could be caused by mechanical impact on the canister due to uneven swelling pressures or movement of the rock. Manufacturing defects may also lead to accelerated failure.

Impact on the repository system:

Failure of the iron insert of the canister will expose the spent fuel to groundwaters and, thus, will initiate release of radionuclides. The mode of failure will control the geometry of perforations and, in turn, the inflow and outflow of water and gas, and the release of radionuclides to the bentonite buffer.

Bibliographic references:

Blackwood DJ, Hoch AR, Naish CC, Rance A and Sharland SM (1994) Research on corrosion aspects of the Advanced Cold Process Canister. SKB Technical Report, TR 94-12.

Hoch AR and Sharland SM (1993) Assessment study of the stresses induced by corrosion in the Advanced Cold Process Canister. SKB Technical Report, TR 94-13.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.07 (Mechanical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0	30th June 2000	Bill Miller

Number: **SFL-20**

Name: **GROUNDWATER FLOW THROUGH THE BUFFER AND BACKFILL**

Short description:

Groundwater flow through the buffer and the backfill will occur if the hydraulic characteristics of these materials are altered to such an extent that the transport of contaminants through them becomes dominated by advective processes rather than by diffusional processes. This might arise due either to physical or chemical degradation processes, or due to an initial defect in the emplacement of these materials.

Technical description:

As a consequence of the ideal post-closure hydraulic resaturation of the buffer and backfill, the bentonite clay component of these materials will expand to such an extent that their hydraulic conductivities will be extremely low (see SFL-42 "Hydraulic resaturation of the buffer and backfill").

The performance of the buffer, when resaturated, is designed to control the release of radionuclides by restricting migration to the rates of molecular diffusion (see GEN-9 "Diffusion"). This is achieved by the very high final swelling pressure in the bentonite buffer material creating an extremely low hydraulic conductivity, in the order of 10^{-13} to 10^{-14} m/s (SKB, 1999).

The performance of the backfill, when resaturated, is also designed to restrict groundwater flow but the final hydraulic conductivity in the bentonite-sand backfill material will be lower than that in the buffer, in the order of 10^{-9} to 10^{-10} m/s (SKB, 1999). This means that, when resaturated, advective flow through the backfill will still be theoretically possible, although the expected flow rates will be so slow that, from the perspective of the performance of the engineered barrier, this flow will be insignificant.

Over the lifetime of the repository, various physical and chemical processes will act to degrade the buffer and backfill materials, and these processes may result in an increase in their hydraulic conductivities. If the hydraulic conductivities were increased significantly, then it could lead to an increase in the groundwater flux through the bulk of these materials due to advective flow.

This could occur, for example, due to the removal (washing away) of material by groundwater at the buffer/backfill contact with the near-field rock (see SFL-17 "Erosion of the buffer and backfill") or due to illitisation of the bentonite, causing a loss of swelling pressure (see SFL-3 "Chemical alteration of the buffer and backfill"). However, it is unlikely that any process could result in advective groundwater flow becoming the dominant transport mechanism through the bulk mass of the buffer or backfill in any normal evolution scenario.

Also, over the lifetime of the repository, various processes may cause localised disruption to the buffer or backfill, causing the opening of preferential pathways through these materials, along which groundwater may flow. This could occur as a consequence of the cementation and subsequent fracturing of the buffer and backfill materials (see SFL-3 “Chemical alteration of the buffer and backfill”) or due to the movement of a separate gas phase (bubbles) through these materials (see SFL-24 “Gas flow through the buffer and backfill”).

Similarly, thermal expansion of the water adjacent to the canister could cause mechanical disruption to the buffer, although this would not be expected to propagate throughout the entire buffer thickness. However, due to the very high swelling capability of the bentonite containing buffer and backfill materials, they are likely to maintain a considerable ability to ‘self-heal’ any small localised pathways, provided their rheological properties have not been degraded.

If a preferential flow path was initiated, it is feasible that positive feedback will act to enhance the flow channel. The increased volume of flow through the buffer or backfill will change the chemical conditions more rapidly and potentially accelerate the degradation process. An example of this type of positive feedback is illustrated by the failure of an earth dam due to piping where, in a short space of time, a small leak becomes a large breach as the increasing flow rates erode material in increasing large quantities. This type of failure mechanism would be more likely to occur in the backfill mix of bentonite and sand, than in the bentonite of the buffer, due to possible inhomogeneous mixing of the material and the particle size distribution of the sand.

Preferential flow paths possibly may be initiated during the near-field hydraulic resaturation event (see SFL-42 “Hydraulic resaturation of the buffer and backfill”), when local head gradients could develop which are significantly greater than the equilibrium conditions within the regional groundwater flow system. More rapid water movement through the buffer and backfill in response to the head gradients, before swelling is completed, may result in disruption to the material distribution. This would be exacerbated if resaturation was localised at individual fast-flowing fractures in the near-field rock which were inadequately sealed.

Groundwater flow through the buffer and backfill would enhance the exchange of dissolved species in the groundwater between the near-field rock and the engineered barriers and, thus, would affect the near-field groundwater chemistry (see SFL-53 “Evolving water chemistry in the buffer” and SFL-54 “Evolving water chemistry in the backfill”). This may cause an increase to the chemical degradation rate of the buffer and backfill materials themselves. Preferential pathways may also expose the canister to an increased supply of corrosive agents and accelerate localised copper corrosion (see SFL-7 “Corrosion of the copper shell”).

After failure of the canister and the exposure of the spent fuel to groundwater, flow through the buffer would enhance the radionuclide release rate from the near-field while, at the same time, limiting the opportunity for dispersion and sorption on the buffer materials. The impact on radionuclide releases due to groundwater flow through the backfill is not so clearly defined but would still be a non-conservative process (see SFL-50 “Radionuclide release and transport from the buffer and backfill”).

Ideally, the changing chemical, physical and hydraulic properties of the buffer and backfill materials should be described in models as functions of time, although this rarely attempted in performance assessments because of the complexity of the system. Likewise, laboratory studies of flow in buffer materials are poorly representative of the repository systems and generally have been restricted to model systems with large hydraulic gradients in order to accelerate migration processes to measurable rates (e.g. Börgesson et al., 1995).

Origin in the repository system:

Flow through the buffer or backfill could occur in the repository if the physical and chemical conditions cause the hydraulic properties of bentonite to be severely degraded or there is an initial defect in the materials.

The potential for groundwater flow to be initiated in the buffer and backfill is dependent on the characteristics of the hydraulic resaturation event, the hydraulic properties of the material after resaturation, and the evolving temperature, hydraulic and gas pressure gradients across the buffer and backfill materials during the lifetime of the repository.

Impact on the repository system:

Groundwater flow through the buffer and backfill will allow the more rapid circulation of groundwater across the entire near-field. This will promote changes to the near-field groundwater composition and degradation of the engineered barriers, including localised corrosion of the copper outer canister.

Once radionuclides are released from spent fuel, groundwater flow through the buffer would cause an increase in the radionuclide release rate to the near-field rock. The impact on radionuclide releases due to groundwater flow through the backfill is not so clearly defined. The development of fast pathways in the backfill could concentrate any radionuclides thus causing the benefits of dispersion and retardation processes in the backfill material to be lost.

Bibliographic references:

Börgesson L, Johannesson L-E, Sandén T and Herelind J (1995) Modelling of the physical behaviour of water-saturated clay barriers: laboratory tests, material models and finite element application. SKB Technical Report, TR 95-20.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and the EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.8 (Preferential pathways in the buffer/backfill) and 3.2.9 (Flow through buffer/backfill) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-21**

Name: **SPENT FUEL DISSOLUTION AND CONVERSION**

Short description:

Dissolution and conversion of the UO_2 spent fuel matrix will occur after the canister fails and groundwater comes in contact with the fuel surface. As a consequence, the fuel matrix will begin to dissolve and radionuclides incorporated in the matrix will be liberated.

Technical description:

After emplacement, when the canister is intact, dissolution and conversion of the spent fuel UO_2 matrix will be limited to reaction with the small amounts of air or water trapped in the void spaces within the canister (see SFL-10 “Canister corrosion prior to wetting”). These reactions could be minimised by purging the canister with inert gas prior to sealing. However, even if this were not done, the very small amounts of water which are likely to be present in a sealed canister will mean that dissolution and conversion reactions prior to canister failure will be negligible.

After failure of the canister and the Zircaloy cladding, groundwater will come into direct contact with the surface of the spent fuel. If this groundwater is chemically-reducing (free of dissolved oxidants) then the spent fuel will begin to dissolve at a rate controlled by the solubility of the UO_2 matrix in the water adjacent to the fuel surface and the rate of uranium transport out of the canister. Radionuclides would then be released from the spent fuel matrix at a rate determined by the uranium dissolution rate. Since the solubility of uranium under these conditions is low and the rate of water turnover in the canister is also low, this radionuclide release rate will be very slow.

Radionuclides contained in the fuel-cladding gap will be released rapidly to solution when the Zircaloy cladding is perforated at a release rate which is controlled by the solubility of the segregated materials and is independent of the dissolution rates of the spent fuel UO_2 matrix (see SFL-22 “Gap and grain boundary release”).

Radiation (particularly alpha radiation) emitted from the spent fuel matrix is likely to cause radiolysis of the water adjacent to the fuel surface (see GEN-30 “Radiolysis”). Radiolytic decomposition of water molecules leads to the formation of numerous ionised species. Although equal amounts of oxidants and reductants are formed, these species are extremely reactive or unstable, and quickly convert to more stable or inert species. This leads to the formation of molecular hydrogen (gas) and oxidants such as hydrogen peroxide (H_2O_2) and molecular oxygen. The hydrogen gas is relatively unreactive under near-field temperatures (<100°C) so will tend not to impact on redox reactions. The net effect of the radiolysis of groundwater is, thus, to make the groundwater in contact with the fuel surface oxidising.

These radiolytic processes create an intrinsic redox disequilibrium at the spent fuel surface which may result in the oxidative conversion of the UO_2 spent fuel matrix and an increased radionuclide release rate. How far this oxidation proceeds depends on the supply of oxidants to the fuel surface. A proportion of the radiolytically-generated oxidants will be consumed in oxidation reactions involving the inner iron insert. Thus the supply of oxidants to the spent fuel will be restricted.

An oxidation of the spent fuel to U_4O_9 or U_3O_7 can take place without changing the crystal structure of the fuel matrix. Under such conditions, the radionuclide release rate remains controlled by the rate of spent fuel dissolution (still determined by the solubility of the uranium oxide matrix and the transport of uranium out of the canister). Conversion to higher oxidation states, to U_3O_7 or UO_3 or some other U(VI) compound, will alter the crystal structure of the fuel. The solubility of U(VI) compounds is considerably higher than the less oxidised U(IV) species. Furthermore, the lattice conversion is likely to cause the direct expulsion of some proportion of the radionuclides from the uranium oxide matrix. Thus, for the higher states of oxidative conversion, the radionuclide release rate will be controlled directly by the rate of fuel conversion rather than the rate of uranium dissolution.

As the uranium oxide matrix is converted, the released radionuclides may go directly to solution, if they are readily soluble, and be transported from the canister. Otherwise, these radionuclides can be incorporated into newly formed separate uranium-bearing mineral phases (e.g. uranyl silicates) or co-precipitates, or sorbed onto canister corrosion products.

The main factors controlling the long-term dissolution and conversion of the original UO_2 spent fuel matrix, and the radionuclide release rates are, thus, the radiation dose rates to the groundwater in the canister, the rate of oxidant generation, the competitive oxidant uptake by the fuel and other materials (principally iron), the rate of water turnover in the canister and the oxidation state reached by the spent fuel.

Other dissolved species in the groundwater and the changing near-field temperatures may complicate these processes, although salinity is generally not a problem for the matrix stability of spent fuel unless the saline groundwaters which might contact the fuel are also of low pH.

Some proportion of the uranium released from the spent fuel is likely to sorb onto canister corrosion products and filling materials inside the failed canister (see SFL-28 “Radionuclide interaction with corrosion products”). This will affect the concentration of uranium close to the fuel surface and thus also the continued fuel dissolution and conversion rates. Increased amounts of canister corrosion products inside the failed canister will also restrict further groundwater access to the spent fuel surface and may act to slow the bulk spent fuel dissolution rate.

Natural analogues have provided considerable insight into the long-term processes of UO_2 (uraninite) dissolution and conversion. Studies at the Cigar Lake uranium orebody in Canada particularly have examined the impact of radiolysis. Mineralogical data from Cigar Lake show that the stoichiometry of the ore ranges from U_4O_9 to U_3O_7, suggesting it may have been oxidised as a consequence of the radiolysis to a limited extent. Geochemical modelling studies indicate that the U_3O_7 is the main uranium solubility limiting phase (Janeczek and Ewing, 1992; Cramer and Smellie, 1994).

Predictions of the dissolution of spent uranium fuel can be described by a surface-reaction control mechanism and explicitly accounting for formation of alteration/secondary phases, and feedback from the evolving composition of the coexisting solution. This approach permits the integration of fuel dissolution with other near-field processes (e.g. diffusional transport, sorption) to model early, transient dissolution behaviour. However, the long-term, steady-state dissolution rate is constrained to be derived from an empirical fit of shorter-term laboratory data. Accordingly, the predictive capability of such detailed, kinetic models for long-term fuel dissolution analysis is problematical. However, the approach clarifies the importance of formation of alteration phases at the surface of the fuel. These phases control the subsequent concentration of most radionuclides released from the fuel.

Origin in the repository system:

The dissolution and conversion of spent fuel may progress only after the canister and the Zircaloy fuel cladding have been breached, and groundwater can contact the fuel. The dissolution and conversion processes are strongly controlled by the chemical environment in the canister and, most importantly, by reaction with radiolytically generated oxidants. The significance of these processes is thus dependent largely on the lifetime of the canister (defining the radiolytic dose to the groundwater and thus the rate of oxidant production) and then balance between oxidant production and competitive reaction between the spent fuel and the inner iron canister.

If oxidation of the spent fuel does not progress beyond the U_3O_7 stage, the radionuclide releases from the fuel is controlled by the rate of uranium dissolution. If oxidation of the fuel passes the U_3O_8 stage, then radionuclide releases are controlled by the rate of conversion.

Impact on the repository system:

The processes of dissolution and conversion of spent fuel are first order controls on repository safety and define the primary radionuclide source term, excluding gap releases which occur independently of spent fuel matrix dissolution or conversion.

Spent fuel dissolution and conversion controls the water chemistry (radionuclide content) in the canister and the near-field. Release of radionuclides from the fuel by these processes will change the inventory of the waste.

Bibliographic references:

Bruno J, Cera E, Grivé M, Eklund U-B and Eriksen T (1999) Experimental determination and chemical modelling of radiolytic processes at the spent fuel/water interface. SKB Technical Report, TR 99-26.

Cramer JJ and Smellie JAT (1994) Final report of the AECL/SKB Cigar Lake analog study. AECL Technical Report, AECL-10851; SKB Technical Report, TR 94-04.

Eriksen TE, Eklund U-B, Werme LO and Bruno J (1995) Dissolution of irradiated fuel: a radiolytic mass balance study. Journal of Nuclear Materials, 227, 76-82.

Forsyth RS (1995) Spent nuclear fuel: a review of properties of possible relevance to corrosion processes. SKB Technical Report, TR 95-23.

Forsyth RS and Werme LO (1992) Spent fuel corrosion and dissolution. Journal of Nuclear Materials, 190, 3-19.

Grambow B (1990) Status in understanding and modelling radionuclide release from high level waste glass and spent fuel. Proceedings of Symposium on Safety Assessments of Radioactive Waste Repositories, Paris 1989. NEA/OECD.

Janeczek J and Ewing RC (1992) Dissolution and alteration of uraninite under reducing conditions. Journal of Nuclear Materials, 190, 157-173.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform and characteristics), 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.2.6 (Solubility within the fuel matrix) and 1.2.9 (Dissolution chemistry) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-22**

Name: **GAP AND GRAIN BOUNDARY RELEASE**

Short description:

Some elements accumulate at the fuel surface and in the pores, grain boundaries and in the gap between the fuel pellets and the Zircaloy cladding of the fuel rod. When the canister is perforated and groundwater contacts the spent fuel, these elements can be preferentially released in solution.

Technical description:

During irradiation in the nuclear fuel reactor a number of gaseous fission products are formed, such as xenon (Xe). A fraction of these gaseous species migrate through the fractures and pores in the fuel pellets towards the fuel surface.

Certain other elements may be associated with the gaseous phases and migrate with them, to some extent, to become enriched in the fuel pores, the grain boundaries and in the gap between the fuel pellets and the Zircaloy cladding of the fuel rod. Laboratory studies indicate that the elements most likely to display this behaviour are iodine (I) and caesium (Cs). However, this is not the case for all fission products. The solubility of fission products in the UO_2 matrix and the reactor burn-up temperatures are important parameters for controlling the inventory of the gap accumulation and release.

The chemical form of these elements in the grain boundary and gap fractions is not well established. However, evidence suggests that Cs and I exist as CsI, although some I may occur in elemental form. Actual gap inventories are found often to be significantly less than the percentage of volatile fission gases. Most studies suggest that the proportion is under 10 % for I and Cs.

Some related metals (e.g. Tc, Mo, Ru, Rh and Pa) can form metallic precipitates, known as ‘white inclusions’, mainly at the grain boundaries but sometimes also in connection with the pores. The quantity of these inclusions is also related to the burn-up history and the oxygen-metal ration in the fuel.

The Cs and I inventories are highly soluble and upon contact with even a small volume of groundwater undergo rapid and complete dissolution in a time period which is short in comparison with those normally associated with performance assessment calculations. This event is represented as an instant release of those radionuclides to an ‘interior’ volume of groundwater. The amount of radionuclides released divided by this volume represents an initial concentration and, if these radionuclides are not supplemented by dissolution of other wasteform sources, then a single pulse release will occur.

Under diffusive conditions, these radionuclides will migrate away from the waste source in response to their concentration gradients. Because such instant releases can contain up to several percent of ^{129}I and ^{135}Cs, modelling their subsequent transport is of concern in repository safety analyses.

In the conservative bounding case, the abundance of volatile radionuclides in this instant release source can be equated with the measured percentage of fission gas released to this same gap region. An estimated range for instant release is < 0.1 % to approximately 20 % for volatile fission products from high temperature CANDU fuel, with values below 2 % more representative of LWR fuel. A systematic approach has been developed for CANDU fuel (Stroes-Gascoyne and Sellinger, 1986; Garisto et al., 1989) to derive empirical relations between fission gas releases and actual gap inventories of the less volatile fission products, and similar relationships could be applied to other fuels.

Release from a grain boundary phase or phases may be modelled as a separate source, having a characteristic release rate intermediate between the gap/instant release and the dissolution of the UO_2 matrix. Although modelling radionuclide release from a discrete grain boundary source is feasible, the efficacy of such a model is severely compromised by several factors. The chemical and physical characterisation of such a grain boundary phase is poor, and there are irreducible uncertainties in the overall percentage of released volatile fission products.

Furthermore, controlled oxidation and analysis of fuel grains show negligible quantities of important fission products along grain boundaries (Gray et al., 1992). However, the grain boundary inventories are unlikely to be constant during the period of fuel storage and during the long period after emplacement before the canister is perforated, because irradiation assisted diffusion may cause accumulations of fission products to occur over time at the grain boundaries.

The importance of gap and grain boundary release is partly dependent on the number of cracks in the fuel pellets. Cracking occurs in all fuel pellets because of the high temperatures in the reactor but enhanced cracking can occur if successive heating/cooling occurs due to frequent reactor shutdowns or changes in burn-up rate. If this takes place, this will affect the distribution of the volatile species within the pellets.

Cracking and embrittlement of the fuel pellets can also occur after canister emplacement, and enhanced temperature inside the canister may influence the potential migration in the fuel matrix. Long-term degradation of the fuel pellet in contact with the near-field groundwaters may also affect the release of fission products (see SFL-13 “Degradation of the spent fuel elements”).

Origin in the repository system:

Radionuclide accumulation at the fuel surface and in the pores, grain boundaries and the gap between the fuel pellets and the Zircaloy cladding of the fuel rod is determined by the operating temperature and the burn-up history of the fuel.

Accumulations of I and Cs in the gap and grain boundaries will occur which are very soluble and, thus, there is an instantaneous release of these elements when the canister is breached. The time of release depends on the lifetime of the canister.

Impact on the repository system:

The prompt release of the I and Cs in the gap and grain boundaries will have a significant impact on the timing and rate of radionuclide release from the near-field. Because these elements, particularly I, can exhibit conservative behaviour they can migrate quickly through the far-field and thus impact on the calculated dose and risk to humans in the surface environment.

The prompt release of these elements will impact on the chemistry of the near-field groundwater.

Bibliographic references:

Garisto NC, Johnson LH and Hocking WH (1989) Proceedings of the Second International Conference on CANDU fuel. October 1989, Chalk River, Canada.

Gray W, Leider H and Seward S (1992) Parametric study of LWR spent fuel dissolution kinetics. Journal of Nuclear Materials, 190, 46-52.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Stroes-Gascoyne S and Sellinger DM (1986) Proceedings of the First International Conference on CANDU fuel. October 1986, Chalk River, Canada.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.5 (I, Cs migration to the fuel surface) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-23**

Name: **GAS ESCAPE FROM CANISTER**

Short description:

Gases will be generated inside the canister by a number of processes: the most important will be hydrogen production from anaerobic iron corrosion. At low gas pressures, these gases may dissolve in the groundwaters and escape from the canister by diffusion or water turnover. At high gas pressures, gas flow from the canister may occur. Gas escape from the canister could affect total radionuclide releases from the canister, groundwater inflows to the canister and, thus, continued corrosion of the canister and the spent fuel.

Technical description:

A variety of gases can be generated inside the canister by a number of different processes (see SFL-25 “Gas generation in the canister”). Prior to canister failure, some hydrogen gas may be generated inside the canister by anaerobic corrosion of the iron insert by residual water contained in the fuel pins (see SFL-10 “Canister corrosion prior to wetting”).

After perforation of the copper shell of the canister, groundwater can penetrate into the annular gap between the shell and the iron insert initiating anaerobic corrosion of the outer wall of the iron insert and hydrogen production. Subsequently, when the iron insert fails, groundwater can penetrate into the central void spaces of the canister under the action of the hydraulic gradient between the external groundwater pressure and the interior pressure. At this point, anaerobic corrosion of the inner wall of the iron insert and the stainless steel components of the fuel assembly can commence, generating additional volumes of hydrogen.

Gases will also be produced in significant quantities by radiolysis of the groundwater (see GEN-30 “Radiolysis”), although in much smaller volumes than the hydrogen produced by anaerobic corrosion of iron. Ionising radiation from the spent fuel will produce radicals and the molecular products H_2O_2, H_2 and O_2.

Lastly, helium gas will also be produced within the waste package by alpha decay of radionuclides in the fuel and some gases (e.g. CO_2, H_2 and CH_4) may be produced by microbial decomposition of organic materials introduced into the canister with the groundwater. However, the volumes of gas produced by these additional processes will be very small.

These gases (mostly hydrogen) will initially dissolve into the groundwater inside the canister and will be transported through and out of the canister by diffusion or by turnover of the water (see SFL-49 “Radionuclide release and transport from the canister”). However, if the gas production rate exceeds the gas transport rate, then a separate gas phase can form if the increasing hydrogen partial pressure exceeds the local hydrostatic pressure. Modelling work on the spent fuel repository near-field suggests that this is likely to occur (Wikramaratna et al., 1993; Bond et al., 1997).

The behaviour of the separate gas phase (bubble) depends on the geometry of the perforations in the canister. If the perforations are at the top of the canister, then the gas bubbles will rise upwards out of the canister and accumulate at the interface between the canister and the bentonite buffer. If the perforations are in the sides or bottom of the canister, then the gas bubble will be trapped inside the canister. Water remaining in the canister will then be consumed in the process of steel corrosion producing more hydrogen gas (SKB, 1999). As further gas is generated, the gas bubble will grow and will expel groundwater from the canister, through the perforations. This process will continue until the gas bubble grows sufficiently large to extend down to the perforation in the side of the canister when it may begin to flow out to the interface between the canister and the bentonite buffer.

Gas pressures will continue to rise inside the canister and outside the canister at the canister-buffer interface if the loss of gas through the buffer by diffusion is slower than the gas generation rate. In this case, the gas pressure will rise until it exceeds a critical point when a gas flow pathway is opened through the bentonite (see SFL-24 “Gas flow through the buffer and backfill”). Laboratory experiments suggest this critical point is equivalent to the sum of the hydrostatic pressure and the bentonite swelling pressure (Horseman and Harrington, 1997). Further gas loss through the buffer could occur only by diffusion until gas pressures once again reach a critical point.

The opening of a gas flow pathway will allow the excess gas to escape from the canister and the gas pressure to drop until it falls below a shut-in pressure, which is dependent on the bentonite swelling pressure (see SFL-1 “Swelling of the bentonite buffer”). As the gas pressures drop, gas will flow from inside to the outside of the canister, through the perforations, to equilibrate gas pressures across the canister. When the gas pressure inside the canister drops to below the hydrostatic pressure, groundwater will be forced through the perforations back into the canister.

Gas flow from the canister is, thus, controlled by the rate of gas generation in the canister (mostly hydrogen production due to anaerobic iron corrosion), the gas diffusion rate through the buffer, and the hydrostatic and bentonite swelling pressures.

Gas escape from the canister will affect the total radionuclide release rate from the canister (see SFL-49 “Radionuclide release and transport from the canister”) because some radionuclides can be associated with the gas molecules (e.g. tritium). Gas escape will also affect the inflows and outflows of water to the canister (see SFL-56 “Water turnover in the copper shell” and SFL-57 “Water turnover in the cast iron insert”), if the geometry of the perforations allows a gas bubble to accumulate in the canister. In turn, this would affect the rates of corrosion of the spent fuel and the inner surfaces of the canister. A change in gas pressure would also impact on the concentration of gas dissolved in the groundwater and, thus, on the groundwater chemistry adjacent to the canister. Transport of gas from the canister to the near-field rock will impact on the groundwater chemistry in the rock.

Origin in the repository system:

Gas escape from the canister would only be possible after perforation of the copper shell. Gas generation and escape will be more rapid after the iron insert is also perforated because this allows for greater surface areas for anaerobic iron corrosion. Escape of gas dissolved in groundwater is dependent on the rate of water turnover or diffusion through the perforations in the canister. Escape of a separate gas phase by flow is dependent on the balance between the rates of gas generation at the canister and gas diffusion through the buffer, and the hydrostatic and bentonite swelling pressures.

Impact on the repository system:

Gas escape from the canister will affect the radionuclide release rate from the canister, water turnover in the canister, the rates of corrosion of the spent fuel and the inner surfaces of the canister, and the groundwater chemistry in the canister and in the bentonite buffer.

Bibliographic references:

Bond AE, Hoch AR, Jones GD, Tomczyk AJ, Wiggin RW and Worraker WJ (1997) Assessment of a spent fuel canister: assessment studies for a copper canister with a cast steel inner component. SKB Technical Report, TR 97-19.

Horseman ST and Harrington JF (1997) Study of gas migration in MX80 buffer bentonite. BGS Technical Report, WE/97/7.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Wikramaratna RS, Goodfield M, Rodwell WR, Nash PJ and Agg PJ (1993) A preliminary assessment of gas migration from the copper/steel canister. SKB Technical Report, TR 93-31.

Equivalent FEPs:

Relates to FEPs 2.1.12 (Gas sources and effects in wastes and the EBS) and 3.2.09 (Gas mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.1.4 (Gas generation: He production) and 3.2.12 (Gas transport in bentonite) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-24**

Name: **GAS FLOW THROUGH THE BUFFER AND BACKFILL**

Short description:

Some gas will be trapped in the buffer when the repository is first closed. After resaturation, hydrogen gas will be generated by anaerobic corrosion of the iron insert. Smaller volumes of gas may be generated by radiolysis, radioactive decay and microbial activity. A separate gas phase may form and, if the gas pressure rises beyond a critical point, may open pathways in the bentonite buffer and flow through them.

Technical description:

When the repository is first closed, some pore spaces in the bentonite and around the canister will be filled with air. As the near-field hydraulically resaturates, this air will be dissolved in the infiltrating groundwaters and removed by diffusion or consumed in reactions with the near-field barriers (see SFL-42 “Hydraulic resaturation of the buffer and backfill”).

After the hydraulic resaturation event, gases will be generated at the canister by a number of processes (see SFL-25 “Gas generation at the canister”). The most significant will be anaerobic corrosion of the iron insert but this can only occur after the copper shell has been perforated by corrosion (or other mechanical failure) and water can penetrate the void between the insert and the shell. When the insert has also been perforated, some gases may be generated by the radiolytic decomposition of water (see GEN-30 “Radiolysis”), by radioactive decay (see GEN-28 “Radioactive decay”) or by microbial activity (see SFL-32 “Microbial activity”).

These gases (mostly hydrogen) will initially dissolve in the groundwaters adjacent to the canister and be removed with the groundwater by diffusional processes or water turnover. However, if the gas production rate exceeds the diffusional transport rate, then a separate gas phase can form if the increasing hydrogen partial pressure exceeds the local hydrostatic pressure. Modelling work on the spent fuel repository near-field suggests that this is likely to occur (Wikramaratna et al., 1993).

The separate gas phase (bubble) will be immobile at the canister/bentonite interface and may disturb the buffer locally to create room (see SFL-31 “Mechanical impact on the buffer and backfill”). If the gas pressure continues to increase, then a critical point may be reached allowing the gas to breakthrough the bentonite and flow. Laboratory experiments suggest this critical point (opening pressure) is equivalent to the sum of the hydrostatic pressure and the bentonite swelling pressure, which is around 12 MPa (Horseman and Harrington, 1997; Tanai et al., 1997). When the opening pressure is exceeded a gas flow pathway is opened through the bentonite and the build-up of gas adjacent to the canister is released. As the gas pressure drops, the pathway will close when the pressure falls below a “shut-in pressure”, which is dependent on the bentonite swelling pressure and around 9 MPa. At all pressures below the shut-in pressure, further gas transport can only occur by diffusion.

Continued hydrogen production may result in a subsequent rise in gas pressure and the gas flow pathway may be opened again. It is possible that the opening pressure required for a second gas flow event will be lower than that required for the first (Nash et al., 1998). The geometry and characteristics of the gas flow pathways are uncertain, and it is also not clear if they will be fully healed by the bentonite after closure. To date no experimental studies have fully replicated the gas volumes and pressures which will be associated with gas flow through bentonite in a repository environment.

Over the lifetime of the repository, various physical and chemical processes will act to degrade the buffer, and these processes may result in a decrease in its swelling pressure. This could occur, for example, due to the removal (washing away) of material by groundwater at the buffer contact with the near-field rock (see SFL-17 “Erosion of the buffer and backfill”) or due to illitisation of the bentonite (see SFL-3 “Chemical alteration of the buffer and backfill”). Also, over the lifetime of the repository, various processes may cause localised disruption to the buffer which might provide localised gas flow pathways. This could occur as a consequence of the cementation and subsequent fracturing of the buffer materials. If any of these processes did occur, then the opening pressure required to allow gas flow to occur through the bentonite would decrease.

Gas flow events through the buffer would cause a rapid decrease in the gas pressure and volume adjacent to and inside the canister. This would in turn affect the water turnover in the canister and the rate of further metal and spent fuel dissolution, if a gas bubble had been expelling water from the canister void spaces (see SFL-57 “Water turnover in the cast iron insert”). A change in gas pressure would also impact on the concentration of gas dissolved in the groundwater and, thus, on the groundwater chemistry adjacent to the canister. Transport of gas from the canister to the near-field rock will impact on the groundwater chemistry in the rock.

Gas flow through the buffer may affect the bulk physical characteristics of the buffer and could locally reduce the hydraulic conductivity if the buffer if it does not fully self-heal after the closure of a gas flow pathway. This could affect continued radionuclide transport in the near-field by diffusion (see GEN-9 “Diffusion“) or groundwater flow (see SFL-20 “Groundwater flow through the buffer and backfill”).

Origin in the repository system:

Gas flow through the buffer may occur as a direct consequence of the generation of gases at the canister. The most significant gas is hydrogen which is generated by anaerobic corrosion of the iron insert. Gas flow through the buffer (as opposed to diffusion) will take place if the gas generation rate exceeds the gas diffusion rate, allowing the gas pressure to increase and eventually to exceed the ‘opening pressure’ required to form a gas flow pathway through the bentonite buffer. This opening pressure is dependent on both the hydrostatic pressure and the bentonite swelling pressure.

Impact on the repository system:

Gas flow through the buffer will impact on gas pressure in the canister and, thus, the rate of further corrosion of the canister and the spent fuel if a gas bubble inside the canister had shielded these materials from the groundwater. Changing gas pressure will affect the groundwater chemistry in the vicinity of the canister and in the near-field rock.

Gas flow through the buffer may affect the bulk physical characteristics of the buffer, including its hydraulic conductivity, and thus could affect continued radionuclide transport in the near-field.

Bibliographic references:

Horseman ST and Harrington JF (1997) Study of gas migration in MX80 buffer bentonite. BGS Technical Report, WE/97/7.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Tanai K, Kanno T and Gallé C (1997) Experimental study of gas permeabilities and breakthrough pressures in clays. Scientific Basis for Nuclear Waste Management, XX, 1003-1010.

Wikramaratna RS, Goodfield M, Rodwell WR, Nash PJ and Agg PJ (1993) A preliminary assessment of gas migration from the copper/steel canister. SKB Technical Report, TR 93-31.

Equivalent FEPs:

Relates to FEPs 2.1.12 (Gas sources and effects in wastes and the EBS) and 3.2.09 (Gas mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.12 (Gas transport in bentonite) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-25**

Name: **GAS GENERATION IN THE CANISTER**

Short description:

Gas may be generated inside and on the outside surfaces of the canister by a number of processes such as anaerobic metal corrosion, decay-derived helium generation and radiolysis. The gas production will impact on further corrosion of the canister and transport of water in the canister and the buffer.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). Gas will be produced inside and on the outside surfaces of the canister by a number of different processes.

In terms of gas volume produced, the most significant process is likely to be corrosion of the inner iron insert, and the Zircaloy and stainless steel non-fuel waste parts. These components can begin to corrode and evolve gas immediately after the canister is sealed by reaction with aqueous and gaseous corrodants trapped inside the canister (see SFL-10 “Canister corrosion prior to wetting”). However, significant corrosion will not commence until after the copper shell is perforated and groundwaters can contact the inner iron insert.

The iron will react directly with water to produce hydrogen under anaerobic conditions (the corrosion of the Zircaloy proceeds by similar hydrogen evolving reactions):

$3Fe^{\circ} + 4H_2O_{(l)} = Fe_3O_{4\,(s)} + 4H_{2\,(aq)}$

Hydrogen gas production has been considered in the some performance assessments for HLW repositories (e.g. Werme, 1990; TVO, 1992; Wikramaratna et al., 1993) and these suggest that corrosion produces more hydrogen than can be dissolved in the groundwater present in each deposition hole in a KBS-3 type repository. Although these calculations assumed a steel container rather than an iron one, the conclusion remains the same. Consequently, a separate gas phase is likely to form and hydrogen must be transported away from the waste package as a gas.

Gas will also be produced in significant quantities by radiolysis (see GEN-30 “Radiolysis”). Ionising radiation from the spent fuel will produce radicals and the molecular products H_2O_2, H_2 and O_2. If there is water present in the canister when it is initially sealed, radiolysis will begin immediately on encapsulation. Radiolysis of moist air in the canister can take place prior to water intrusion in the canister. This would lead to the formation of corrosive gases such as nitrous and nitric acids and ammonia (see SFL-10 “Canister corrosion prior to wetting”).

After emplacement, radiolysis of the groundwater can occur. However, as long as the canister remains intact, the amount of radiolysis will be very small because only gamma radiation can penetrate out through the canister and this will be considerably attenuated.

The most significant radiolytic processes will be alpha radiolysis and this can only occur after the canister has been perforated and groundwater can contact the spent fuel. What is important for this process is the time of canister failure since the alpha radiation (thus gas production) clearly decreases with time because of radioactive decay (see GEN-28 “Radioactive decay”). Most species produced by alpha radiolysis are extremely reactive or unstable, and quickly convert to more stable or inert species. Radiolysis leads to the production of equal amounts of oxidising and reducing species. Hydrogen being least reactive will accumulate over time.

Helium gas will also be produced within the waste package by alpha decay of radionuclides in the fuel and some gases (CO_2, H_2 and CH_4) could be produced by microbial decomposition of organic material. However, the volumes of gas produced by these additional processes will be secondary in comparison to hydrogen production from iron corrosion.

Origin in the repository system:

Gas production at the waste package is largely controlled by the lifetime of the canister. Prior to canister failure, gas production will be insignificant. After canister failure, the most important process for producing gas at the waste package will be hydrogen producing anaerobic corrosion of the cast iron insert, and the Zircaloy and stainless steel non-fuel waste parts. The corrosion process is dependent on the groundwater composition and the temperature. Other processes will produce lesser volumes of gas; these are radiolytic decomposition of the groundwater, decay related helium generation and microbial decomposition of organic material.

Impact on the repository system:

Gas generation at the waste package will impact on several near-field processes. A build-up of hydrogen pressure will act to reduce further metal corrosion rates, although this might not be significant at the gas pressures expected. The formation of a separate gas phase in the near-field will also impact on the stability of the spent fuel, radionuclide release from the canister and transport through the compacted bentonite buffer. Corrosive gases produced by radiolysis may exacerbate the corrosion of the spent fuel and the canister. Some gas will dissolve in the groundwater and influence the groundwater chemistry. For example, a build-up of hydrogen could buffer the redox conditions, although this is not usually accounted for in performance assessments.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1991) Project-90. SKI Technical Report, 91:23.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

TVO (1992) TVO-92 safety analysis of spent fuel disposal. YJT Technical Report, YJT-92-33-E.

Werme L (1990) Near-field performance of the advanced cold process canister. SKB Technical Report, TR 90-31.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Wikramaratna RS, Goodfield M, Rodwell WR, Nash PJ and Agg PJ (1993) A preliminary assessment of gas migration from the copper/steel canister. SKB Technical Report, TR 93-31.

Equivalent FEPs:

Relates to FEP 2.1.12 (Gas sources and effects in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.1.4 (Gas generation: He production), 1.2.4 (Gas generation) and 2.3.8 (Internal pressure) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-26**

Name: **GAS GENERATION IN THE BUFFER AND BACKFILL**

Short description:

Gas may be generated in the buffer and backfill by radiolysis of porewaters and by microbial degradation of organics. The gas may form a separate gas phase and may impact on the physical structure of the bentonite and buffer, and on the near-field chemistry.

Technical description:

Gas generation in the backfill and buffer of the spent fuel repository may occur by two processes, radiolysis and microbial degradation of organics.

Radiolysis of the porewaters may occur due to ionising radiation emitted from the canister or from radionuclide-bearing precipitates that have formed in the buffer or backfill (see GEN-30 “Radiolysis”). Most species produced by alpha radiolysis are extremely reactive or unstable, and quickly convert to more stable or inert species. Radiolysis leads to the production of equal amounts of oxidising and reducing species. Hydrogen being least reactive will accumulate over time.

The amount of radiolysis due to radiation from the canister will be very small because only gamma radiation can penetrate out through the canister and this will be considerably attenuated by the thickness of the canister walls.

Radiolysis of porewater due radionuclide-bearing precipitates in the buffer is also likely to be limited. Precipitates could not form until after the canister has failed and the wasteform has begun to dissolve. Under normal conditions this would not occur until long after the repository was closed, by which time radioactive decay will have significantly reduced the levels radioactivity (see GEN-28 “Radioactive decay”). Consequently, this process may only be significant in early canister failure scenarios.

Microbial decomposition of organic material could also lead to gas generation. Microbial populations will occur in the repository but their viability will be restricted by the availability of nutrients (see SFL-32 “Microbial activity”). In the buffer and backfill, there may be small amounts of organic carbon present in the bentonite: for example, Wyoming MX-80 bentonite contains 0.4 vol.% of organic carbon. Microbial decomposition of this organic carbon could lead to the generation of H_2, CO_2 and CH_4.

Alternative microbially mediated gas generating reactions could produce other gases, depending on the microbial species present and the availability of organic carbon and other nutrients.

The gas generated in the buffer and backfill by radiolysis and microbial action will dissolved into the groundwater and thus will influence the groundwater chemistry. An increase in the concentration of dissolved H_2 will further buffer the reducing capacity of the near-field, although this hydrogen may diffuse rapidly from the system.

If gas accumulation exceeds the solubility limits, then a separate gas phase may be produced. Gas bubbles forming in the bentonite buffer or bentonite-rich backfill will affect the structure of the materials which could impact on their hydraulic properties (see SFL-31 “Mechanical impact on the buffer and backfill”).

However, although it is probable that gases would be generated in the buffer and backfill by these processes, it is evident that the volumes evolved and their consequences will always be insignificant compared to the volume of gas produced at the waste package. Consequently, gas generation in the buffer and backfill is considered to be of little significance to repository safety.

Gas generation in the buffer and backfill of HLW and spent fuel repositories is not usually considered in performance assessments and was not considered in SITE-94.

Origin in the repository system:

Gas generation in the buffer and backfill by radiolytic decomposition of the porewaters is largely controlled by the lifetime of the canister. After canister failure, the composition of the porewater will control wasteform dissolution, and radionuclide transport/retardation processes, that may lead to precipitation in the bentonite. If this occurs, subsequent radiolytic decomposition will be dependent on the ‘age’ of the waste and thus the level of radiation emitted.

Gas generation in the buffer and backfill by microbial degradation of organic material is dependent on there being viable populations of microbes and a supply of organic material. This in turn is controlled by the supply of nutrients, e.g. the composition of the near-field porewaters and the buffer and backfill materials.

Impact on the repository system:

Gas generation in the backfill and buffer will affect the groundwater chemistry and the structure of the compacted bentonite material. However, the impacts are expected to be very limited and eclipsed by gas generation at the waste package

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 2.1.12 (Gas sources and effects in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-27**

Name: **RADIONUCLIDE ACCUMULATION AT THE SPENT FUEL SURFACE**

Short description:

Certain elements in the spent fuel have a tendency to accumulate at the fuel surface, in cracks and pores in the fuel pellets, and in the gap between the fuel pellets and the Zircaloy cladding of the fuel rod. When the canister is perforated and groundwater contacts the spent fuel, these elements can be preferentially released.

Technical description:

During irradiation in the nuclear fuel reactor, the individual fuel pellets become heavily fractured due to the high temperatures (up to 1300°C) and the associated thermal stresses. At high temperatures, this process can lead to the formation of an open porosity in the fuel pellets. The extent of fracturing and open porosity is related directly to the operating temperature and the burn-up history of the fuel. A number of the fission products formed during irradiation are gaseous (volatile), such as xenon (Xe). These gaseous species migrate through the fractures and pores in the fuel pellets towards the fuel surface.

Certain other elements may be associated with the gaseous phases and migrate with them, to some extent, to become enriched in the fuel pores, the grain boundaries and in the gap between the fuel pellets and the Zircaloy cladding of the fuel rod (see SFL-22 “Gap and grain boundary release”). The elements most likely to display this behaviour are iodine (I) and caesium (Cs). This effect is well known from laboratory studies which show the proportion of these elements present in the grain boundaries and gaps depends on the burn-up history of the fuel.

The chemical form of these elements in the grain boundary and gap fractions is not well established. However, evidence suggests that Cs and I exist as CsI, although some I may occur in elemental form. Actual gap inventories are found to be significantly less than the percentage of volatile fission gases. Most studies suggest that the proportion is under 10 % for I and Cs.

Some related metals (e.g. Tc, Mo, Ru, Rh and Pa) can form metallic precipitates, known as ‘white inclusions’, mainly at the grain boundaries but sometimes also in connection with the pores. The quantity of these inclusions is also related to the burn-up history and the oxygen-metal ration in the fuel. The actinides occur in solid solution in the UO_2 matrix with an enrichment towards the surface of the fuel pellet. This is because the actinides are formed for the most part by absorption of epithermal neutrons and because the flux of these neutrons is greatest at the surface of the fuel pellet. The highest concentration of actinides is therefore present in the narrow zone at the edge of the pellet.

When the groundwater penetrates the canister and comes into contact with the spent fuel, radionuclides preferentially accumulated at the fuel surface and in the grain boundaries and gap are available for immediate release.

Origin in the repository system:

Radionuclide accumulation at the fuel surface and in the pores, grain boundaries and the gap between the fuel pellets and the Zircaloy cladding of the fuel rod is determined by the operating temperature and the burn-up history of the fuel.

Impact on the repository system:

The accumulation of elements at the fuel surface and in the pores, grain boundaries and the gap makes them preferentially available for release in the groundwaters, after canister failure, provided they occur in soluble forms.

Bibliographic references:

Garisto NC, Johnson LH and Hocking WH (1989) Proceedings of the Second International Conference on CANDU fuel. October 1989, Chalk River, Canada.

Gray W, Leider H and Seward S (1992) Parametric study of LWR spent fuel dissolution kinetics. Journal of Nuclear Materials, 190, 46-52.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Stroes-Gascoyne S and Sellinger DM (1986) Proceedings of the First International Conference on CANDU fuel. October 1986, Chalk River, Canada.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.5 (I, Cs migration to fuel surface) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-28**

Name: **RADIONUCLIDE INTERACTION WITH CORROSION PRODUCTS**

Short description:

Corrosion of the canister metals will generate a range of possible solid secondary alteration products which depend on the groundwater chemistry. Radionuclides released from the spent fuel may interact with these alteration products by a range of processes such as sorption/desorption or (co-)precipitation/dissolution reactions. These interactions have the potential for significantly controlling radionuclide release rates from the near-field.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

Corrosion of the copper shell by deep groundwater is likely to be dominated by sulphide attack to form copper sulphide growths (‘whiskers’ or crystals) which grow from the canister into the buffer because the mobility of copper in the solid copper sulphides is higher than the mobility (diffusion) of dissolved sulphide in the porewater (see SFL-7 “Corrosion of the copper shell”).

After the copper shell has been perforated, groundwaters can begin corrosion of the cast iron insert. Anaerobic corrosion of iron by deep groundwaters is likely to form amorphous iron oxyhydroxides which will crystallise over time to form solid magnetite (Fe_3O_4) at repository temperatures although, if additional dissolved species are present in the groundwater, other solid alteration products may be formed, e.g. pyrite, siderite or Fe-aluminosilicates (see SFL-9 “Corrosion of the cast iron insert”).

Radionuclides released from the fuel assemblies may interact with the copper and iron corrosion products by sorption/desorption or (co-)precipitation/dissolution reactions. Most evaluations of radionuclide-metal corrosion product interactions focus on iron because a greater volume and mass of iron oxyhydroxides will form than copper corrosion products, and because the iron oxyhydroxides have the greatest affinity for radionuclide sorption.

The strong affinity for radionuclides with iron oxyhydroxides means that radionuclides can become associated by sorption or co-precipitation with these phases as they form. If the iron oxyhydroxides precipitate directly on the metal surface, then the radionuclides are removed from the groundwater and their transport is retarded. The very large mass of iron and, subsequently, the very large mass of iron oxyhydroxides which may form provide a potentially large radionuclide retardation capacity within the failed canister. However, over time, the iron oxyhydroxide phases may convert to more crystalline phases such as the magnetite. During this solid state recrystallisation process, it is possible that adsorbed radionuclides will be expelled or, alternatively, that they will become directly incorporated in the new phase by mineralisation. The nature of this process will be largely dependent on the groundwater chemistry inside the canister (controlling radionuclide solubilities and the potential solid phases which may form) and the rate at which the recrystallisation takes place.

The association of radionuclides with secondary iron phases has been investigated in natural analogue studies, such as at the Osamu Utsumi uranium mine, Poços de Caldas in Brazil (Waber et al., 1992). At this site, enrichment of radionuclides by a factor of up to 300 was observed with iron oxyhydroxides on the oxidising side and with pyrite on the reducing side of a natural redox front, proving the association occurs in natural groundwater conditions. Applying these observations to performance assessment, McKinley et al. (1992) considered the inventory of some radionuclides in vitrified HLW and the iron content which will be present in the canister corrosion products. They concluded that radionuclide associations with iron canister corrosion products would be quite capable of immobilising the whole inventory of most safety relevant nuclides.

Colloidal iron oxyhydroxides may also precipitate from a solution of ferrous iron in the canister and radionuclides can be associated with these. Depending on the mobility of the colloidal phases, this process may or may not retard radionuclide migration. However, since colloids cannot migrate through the bentonite buffer (see SFL-5 "Colloid behaviour in the buffer and backfill"), the association of radionuclides with iron oxyhydroxide colloid precipitates should only be of concern if it were to occur in the far-field rock, which is not considered to be a significant process.

The interaction of radionuclides with canister corrosion products will be largely dependent on the groundwater chemistry inside the canister because this controls the nature of the solid phases which may form and radionuclide solubilities in the system (see SFL-52 "Evolving water chemistry in the canister"). The rate of metal corrosion is also important because sufficient mass of alteration phases needs to be produced to coincide with the release of radionuclides from the spent fuel. This will largely be controlled by the groundwater chemistry but is also affected by the temperature.

The association of uranium species with metal corrosion products can reduce the radionuclide concentration in solution. In turn, this can accelerate the dissolution of the spent fuel because this is restricted by the solubility limited behaviour of uranium in the canister groundwater (see SFL-21 "Spent fuel dissolution and conversion"). Radionuclide release rates from the canister can be reduced by their association with immobile solid alteration phases. Association with mobile colloids may cause their transport out of the canister but they will not be able to migrate through the bentonite buffer.

Metal corrosion products have a larger volume than the original metal and thus expansion will occur. This process can provide an increase in the surface area available for sorption but can impact on other near-field barriers (see SFL-51 "Expansion of solid corrosion products"). Radionuclides may subsequently be released from solid metal corrosion products by desorption or dissolution reactions. This might occur if groundwater conditions inside the chemistry change (become more oxidising) due either to an increase in the radiolytic oxidant generation rate or the ingress of oxidising waters from the far-field rock.

Origin in the repository system:

Radionuclides may interact with canister metal corrosion products by sorption/desorption or precipitation/dissolution reactions. The most important interaction will be with iron oxyhydroxides formed by the anaerobic corrosion of the iron insert. The interaction will be largely controlled by the groundwater chemistry and the rate of metal corrosion.

Impact on the repository system:

The interaction of radionuclides with canister metal corrosion products can reduce the radionuclide content in the groundwater and the radionuclide release rate from the canister. However, association of uranium with corrosion products can increase the spent fuel dissolution rate because this reaction is controlled by the solubility limited behaviour of uranium in the groundwater in the canister.

Bibliographic references:

McKinley I, Alexander R, McCombie C and Zuidema P (1992) Application of results from the Poços de Caldas Project in the Kristallin-1 HLW performance assessment. Proceedings of the Second International High-Level radioactive Waste Management Conference, Las Vegas, Nevada.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Waber N, Schorscher HD and Peters T (1992) Hydrothermal and supergene uranium mineralisation at the Osamu Utsumi mine Poços de Caldas, Minas Gerais, Brazil. Journal of Geochemical Exploration, 45, 53-112.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 3.2.01 (Dissolution, precipitation and crystallisation of contaminant) and 3.2.03 (Sorption/desorption processes of contaminant) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-29**

Name: **INTERNAL GAS PRESSURE**

Short description:

Internal pressure is the gas pressure inside the canister prior to and after its failure. This gas pressure can affect corrosion of the canister and of the wasteform, and the migration of water and radionuclides in to and out of the canister.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). Internal pressure is the gas pressure inside this canister before and after failure, and it includes the gas pressure in the void between the cast iron insert and the outer copper shell, as well as the local pressure in cracks in the fuel and in the void between fuel and the cladding.

Prior to the failure of the iron insert, generation of helium (due to alpha decay of radionuclides in the fuel) may lead to a pressure increase in cracks in the fuel and in the gap between fuel and the cladding which may cause further cracking (see SFL-10 "Canister corrosion prior to wetting"). Helium generation and possibly hydrogen formation (due to anaerobic corrosion of the iron) inside the iron insert prior to failure will lead to a pressure increase with time which is dependent on the gas generation rate and the void space inside the iron insert.

Prior to failure of the copper shell, gas formation by radiolysis and corrosion due to the presence of air with a high relative humidity in the void between the cast iron insert and the outer copper shell may lead to a pressure increase. After failure of the copper shell, hydrogen formation may lead to a pressure increase in the void if gas accumulation can take place. A pressure increase will affect the inflow of water to the void and may also displace water from the void if the pressure increases above the surrounding hydrostatic pressure.

After failure of the entire canister, generation of hydrogen from corrosion of the iron insert may continue to lead to an increased pressure inside the package if gas accumulation takes place (Bond et al., 1997). This will occur if the gas cannot escape from the canister or if the gas generation rate is faster than the rate of escape. This will, itself, be controlled by the nature of the perforations in the canister and the gas permeability of the bentonite buffer. Displacement of water from the interior of the canister may take place if the gas pressure increases above the surrounding hydrostatic pressure.

Fluctuations in internal pressure will probably take place, with a pressure increase during periods of gas accumulation, and a decrease in pressure as a result of gas escape or a decrease in the gas generation rate. These pressure fluctuations will affect the movement (displacement) of water inside the canister (see SFL-56 "Water turnover in the copper shell" and SFL-57 "Water turnover in the cast iron insert").

In addition, the internal gas pressure will impact on the corrosion of the remaining canister metal, the spent fuel and the metal non-fuel parts. This will occur because any free gas phase (bubble) forming inside the canister will restrict contact between the water and the solid materials and will also affect the porewater chemistry as some of the gas is dissolved. The gas pressure will thus also affect the radionuclide release and transport rates from the canister.

The internal pressure will impact on the mechanical stability of the waste package and, after the package has suffered significant degradation, may lead to its collapse. A build-up of gas pressure in the vicinity of the waste package could also damage the bentonite buffer.

Origin in the repository system:

The internal pressure in the canister is directly controlled by the gas generation rates (predominantly from metal corrosion with lesser gas generated from radiolysis) and the rate of gas escape from the canister.

Corrosion (gas generation) rates are related to the chemistry of the porewaters and temperature, while the gas escape rate is controlled by the nature of perforations in the canister and by gas permeability of the bentonite buffer.

Impact on the repository system:

Depending on the balance between gas generation and escape, the internal pressure may increase or decrease. The formation of a separate gas phase will limit further aqueous corrosion of the canister metal, the fuel and the metal non-fuel parts. Thus internal pressure can control the rate of radionuclide escape from the canister.

The internal pressure may also impact on the mechanical stability of the waste package and of the bentonite buffer.

Bibliographic references:

Bond AE, Hoch AR, Jones GD, Tomczyk AJ, Wiggin RM and Worraker WJ (1997) Assessment of a spent fuel disposal canister - assessment studies for a copper canister with cast steel inner component. SKB Technical Report, TR 97-19.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1990) Near-field performance of the advanced cold process canister, SKB Technical Report, TR 90-31.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Wikramaratna RS, Goodfield M, Rodwell WR, Nash PJ and Agg PJ (1993) A preliminary assessment of gas migration from the copper/steel canister. SKB Technical Report, TR 93-31.

Equivalent FEPs:

Relates to FEP 2.1.12 (Gas sources and effects in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 2.3.8 (Internal pressure) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **SFL-30**

Name: **MECHANICAL IMPACT ON THE CANISTER**

Short description:

The mechanical impact is the effect of the numerous combined internal and external forces acting on the canister. Depending on the nature and extent of these forces, they could lead to early failure of the canister.

Technical description:

There are several different forces which could have a mechanical impact on a waste canister. These forces can be both internal and external to the canister and generally involve processes that produce increased loads and stresses. These can be caused by chemical reactions which produce gas or expanded solid reaction products. In addition, bentonite swelling pressures and processes that occur in the rock mass, such as fracture movement, will also have an effect.

The most significant internal force on the canister will be the internal gas pressure (see SFL-29 “Internal gas pressure”) which is largely due to hydrogen production by anaerobic iron corrosion (see SFL-25 “Gas generation in the canister”). This internal pressure is controlled by the balance of the rates of gas production and gas loss from the canister.

The most significant external force on the canister under normal repository evolution is the combined pressure from lithostatic and bentonite swelling forces. This external pressure will be controlled by the choice of the bentonite and its degree of compaction on emplacement. This swelling pressure may be uneven during the hydraulic resaturation stage because infiltrating groundwater will be focussed on certain high-flow zones intersecting the disposal hole (see SFL-1 “Swelling of the bentonite buffer”).

Other external forces may also act on the canister due to rock mass movement by faulting, creep of the rock mass and collapse of the repository openings. However, the mechanical properties of the bentonite will limit, to some extent, the impact of rock mass movement on the canister because the bentonite will tend to even out any very localised loads that might otherwise be transferred to the canister by the surrounding rock.

The magnitude of all of these internal and external forces will be time-dependent to some degree and, therefore, the consequent mechanical impact on the canister is likely to vary with time, due either to a change in the forces being applied to the canister or due to initial damage allowing other processes to take place that would otherwise have been prevented. An example of the former might be an increase in the heterogeneous swelling of bentonite, and of the latter mechanical damage to the canister initiating and accelerating the process of stress corrosion cracking.

The effect of mechanical impact on the canister will be to reduce the strength of the canister (see SFL-39 “Reduced mechanical strength of the canister”). This reduction in strength may, in turn, affect the rate of canister corrosion by stress corrosion cracking, affect the creep deformation of the canister, affect the position of the canister within the bentonite buffer and, ultimately may allow the canister to rupture. These same processes may also affect the shape of an intact canister and determine the shape of a failed canister.

Thus, mechanical impact could affect the lifetime of the canister and the characteristics of the failed canister with time, both of which are important controls on the release of radionuclides from the near-field (see SFL-49 “Radionuclide release and transport from the canister”). It should be noted, however, that the canisters are designed to withstand an evenly distributed external pressure of approximately 14 MPa from the surrounding bentonite and rock and, therefore, it is only unexpectedly high or heterogeneously applied forces that are likely to cause early or rapid canister failure.

Origin in the repository system:

Mechanical impact on the canister is the combined effect of internal and external pressures. Thus, the mechanical impact is largely controlled by the internal pressure (gas) and the combination of external lithostatic and bentonite swelling pressures. Additional external forces may be applied to the canister due to rock mass movements.

Impact on the repository system:

The consequence of the mechanical impact on the canister may be to reduced the strength of the canister and, therefore, the duration of containment. Early failure of the canister is most likely if the mechanical impact is unexpectedly high or heterogeneously applied. Thus mechanical impact on the canister has the potential to cause early release of radionuclides.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.07 (Mechanical processes and conditions) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **SFL-31**

Name: **MECHANICAL IMPACT ON THE BUFFER AND BACKFILL**

Short description:

The mechanical impact is the effect of the numerous combined forces acting on the buffer and backfill. Depending on the nature and extent of these forces, they could lead to an increase in hydraulic conductivity and radionuclide transport rates.

Technical description:

There are several different forces which could have a mechanical impact on the buffer and the backfill. These forces generally relate to mass movements of the rock mass but forces could also be generated from degradation of the canister and the waste.

Mass movements in the host rock can occur by faulting, creep of the rock mass and collapse of the repository openings. These mass movements would then exert a force on the buffer and the backfill materials which could be disturbed or damaged to a degree dependent on their mechanical resistance or plasticity (see SFL-31 “Mechanical impact on the buffer and backfill”). However, the greatest mechanical impact is likely to be due to movement along fractures that intersect the disposal holes and the repository tunnels.

Under normal repository evolution, the bentonite buffer and the bentonite-rich backfill will be plastic and should be able to even out any very localised loads that might otherwise be transferred to the canister by the surrounding rock. However, physical and chemical alterations to the buffer and backfill may reduce their ability to withstand mechanical impact over the long term. Such alterations may include cementation and mineralogical alterations.

The extent of disturbance of the buffer and backfill is also determined by the type, rate and magnitude of the mechanical impact, i.e. whether slow creep deformation takes place or whether there is a sudden and significant displacement on one fracture.

The impact will also be different on the buffer and on the backfill because of the different physical characteristics of these two materials. The inclusion of sand in the backfill may give it greater strength than the buffer but less plasticity.

Mechanical impact to the buffer could also occur from degradation of the canister and the waste. Corrosion of the canister metal will create solid alteration products that could have greater volume than the metal they replace (see SFL-51 “Expansion of solid corrosion products”). This corrosion will then generate a mechanical impact on the buffer by compression. In addition, the gas generated by the anaerobic corrosion of the iron insert may form a separate gas phase which might disrupt the buffer as it migrates towards the near-field rock (see SFL-24 “Gas flow through the buffer and backfill”).

Mechanical impact on the buffer and backfill will directly affect their properties; most significantly, it could increase their hydraulic conductivity, especially if they were partially cemented. Initial damage to the buffer and backfill may localise the effects of later processes, which may result in increased levels of heterogeneity in buffer and backfill properties.

Mechanical impacts on the buffer and backfill potentially can reduce radionuclide transport times to the near-field rock by increasing transport rates (increase hydraulic conductivity) or reducing transport distances (thinning of the buffer).

Origin in the repository system:

Mechanical impact on the buffer and backfill is the combined effect of forces arising from movements of the rock mass and degradation of the canister and the wasteform. The extent of the impact is controlled by the magnitude of these forces and the mechanical properties of the buffer and backfill at the time of impact.

Impact on the repository system:

Mechanical impact on the buffer and backfill will directly affect their properties, such as the hydraulic conductivity. Initial failure may localise the effects of later processes, which may result in increased levels of heterogeneity in buffer and backfill properties.

Mechanical impacts on the buffer and backfill potentially can reduce radionuclide transport times to the near-field rock by increasing transport rates or reducing transport distances (thinning of the buffer).

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.04 (Buffer/backfill materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.3 (Mechanical failure of buffer/backfill) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **SFL-32**

Name: **MICROBIAL ACTIVITY**

Short description:

Microbes will exist naturally in the far-field rock and in the groundwater. Other microbes will be introduced on construction materials and in the air during repository excavation, operation and closure procedures. These microbes may affect chemical conditions in the repository and can affect the rates of some reactions. They may also directly affect radionuclide transport by acting as organic colloids.

Technical description:

Microbes exist naturally in most deep rocks and groundwaters (Pedersen and Karlsson, 1995) and these are likely to migrate into the repository excavations. In addition to these, a range of microbes will be introduced to the repository on construction and barrier materials, and in the ventilated air during repository excavation, operation and closure procedures.

Once the repository is closed, these microbes may potentially affect chemical conditions and catalyse certain reactions (for example, metal corrosion). However, in order for microbial populations to persist in the near-field of a closed repository, they must be viable in its the thermal, chemical and radiation environment. In general terms, many of the introduced microbes are adapted for life in aerobic environments and, thus, may not be viable in the near-field after conditions become chemically reducing. It follows that the anaerobic species, which exist naturally in deep rock systems, would be the most likely species to prove viable in the near and far-fields: these include species such as methanogenic bacteria and sulphate reducing bacteria (Stroes-Gascoyne et al., 1997).

In addition to suitable redox conditions, a viable microbial population must also be supported by a supply of water, nutrient elements and energy. In the near-field, microbes will have to compete with the bentonite for water, since the bentonite adsorbs water when it swells (Pedersen et al., 1995; Motamedi et al., 1996). Microbes in the buffer may be killed-off by dehydration as their water is adsorbed into the bentonite. In addition, the supply of certain nutrient elements (e.g. P) and suitable energy sources will be in short supply in the near-field. The radiation field is not expected to be a major limit to viability because many microbes are very radiation resistant. Combined, these features suggest that microbes will not be able to enter a saturated confined bentonite buffer nor will they be able to survive in such an environment (Pusch, 1999).

Microbial populations will be larger in the far-field but it is unlikely that these could migrate into the bentonite buffer after it has saturated, and increase populations there, because the size of most microbes is larger (0.2 to 600 μm) than the porespaces in the buffer (0.002 μm) with high swelling pressures (SKB, 1999). Likewise, for the same reason, microbes close to the canister will be unable to influence the transport of radionuclides which might sorb to them. This situation might change if extensive chemical degradation or alteration to the buffer occurred which substantially reduced the swelling pressure (see SFL-3 “Chemical alteration of the buffer and backfill"). However, the potential significance of this process is thought to be minimal.

The microbes which do occur in the near-field after resaturation may exert some influence on the chemical environment. Microbial processes involving anaerobic species can give rise both to gas evolution, sulphide formation and the generation of organic acids. Organic acids may also act as complexing agents for many metal ions, but they are usually only intermediate products which are further decomposed to carbon dioxide, hydrogen and methane. The sulphide formation is important because sulphide is a primary corrosive agent to the copper shell of the canister (see SFL-7 "Corrosion of the copper shell").

Microbes and the organic acids they generate in the near and far-field rock may influence radionuclide transport because they can take up radionuclides by sorption or complex formation. These aggregates may act as mobile species of radionuclides which would otherwise have a low solubility or a strong tendency to sorb on the mineral surfaces. Microbially-catalysed geochemical reactions can also (at least in principle) cause generation of colloids, e.g. Fe-hydroxide particles by oxidation of iron.

Some microbes can survive sorbed on to mineral surfaces. This may add to the uptake of radionuclides on mineral surfaces. This also introduces an uncertainty in relation to laboratory sorption measurements and the fact that dead bacteria or decomposition products of them may become released to the water phase.

Origin in the repository system:

Microbial activity at the canister and in the buffer will be restricted to the anaerobic populations which can infiltrate the buffer before it resaturates and remain viable there. After saturation, microbes will be too large to move through the bentonite buffer pore spaces. Continued viability is restricted by the limited supply of nutrients, water and useable energy sources in the buffer.

Microbial activity in the rock may be greater because water supply and nutrient in the flowing groundwater may be larger than in the buffer.

Impact on the repository system:

Microbial activity may lead to the formation of corrosive agents, complexing agents and gas. By taking part in redox reactions they will influence redox conditions. Microbes may also take up radionuclides and thereby act as colloids if suspended in groundwater in the rock and as a trap for radionuclides if attached to mineral surfaces.

Bibliographic references:

Motamedi M, Karnland O and Pedersen K (1996) Survival of sulphate reducing bacteria at different water activities in compacted bentonite. Microbiological Letters, 141, 83-87.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Pedersen K and Karlsson F (1995) Investigation of subterranean microorganisms: their importance for performance assessment of radioactive waste disposal. SKB Technical Report, TR 95-10.

Pedersen K, Motamedi M and Karnland O (1995) Survival of bacteria in nuclear waste buffer materials: the influence of nutrients, temperature and water activity. SKB Technical Report, TR 95-27.

Pusch R (1999) Mobility and survival of sulphate reducing bacteria in compacted and fully water saturated bentonite - microstructural aspects. SKB Technical Report, TR 99-30.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Stroes-Gascoyne S, Pedersen K, Haveman SA, Dekeyser K, Arlinger J, Daumas S, Ekendahl S, Hallbeck L, Harmon CJ, Jahromi N and Delaney TL (1997) Occurrence and identification of microorganisms in compacted clay-based buffer material designed for use in a nuclear fuel waste disposal vault. Canadian Journal of Microbiology, 43, 1133-1146.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Biological/biochemical processes and conditions in wastes and EBS) and 2.2.09 (Biological/biochemical processes and conditions in geosphere) in the NEA International Database (NEA, 1998).

Relates to FEP 2.1.10 (Microbes) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-33**

Name: **MOVEMENT OF THE CANISTER IN THE BUFFER**

Short description:

The bentonite buffer provides a stable physical and chemical environment for the canister, and isolates the canister from the near-field rock and flowing groundwaters. These functions may be diminished if the canister moves within the buffer to touch or come close to the near-field rock. This could occur as a result of sinking through the buffer, uneven swelling of the buffer, expansion of canister corrosion products or movement of the near-field rock.

Technical description:

The canister will be located vertically and concentrically within the disposal hole and all spaces around the canister will be filled with bentonite buffer. The nominal thickness of bentonite in the annular gap between the canister and the rock walls of the disposal hole is 350 mm, and the nominal thickness of bentonite between the base of the canister and the floor of the disposal hole is 500 mm (SKB, 1999).

Any movement of the canister within the bentonite buffer will bring some part of the canister closer to the near-field rock and, thus, will reduce the thickness of the bentonite barrier. Extreme movement could, potentially, bring the canister into direct contact with the rock which would entirely negate the buffer's barrier functions. Canister movement could occur for a number of reasons.

The buffer is likely to be emplaced as machined blocks of dry, compacted bentonite. After repository closure, groundwater will infiltrate into the near-field, causing the bentonite to adsorb water and swell (see SFL-42 “Hydraulic resaturation of the buffer and backfill”). Initially, infiltrating groundwater will be focussed on water conducting features intersecting the disposal holes and this could potentially cause localised and uneven swelling of the buffer (see SFL-1 “Swelling of the bentonite buffer”).

Uneven swelling would cause uneven pressures on the canister which might result in its movement in the disposal hole. Such a process would be exacerbated by poor quality control on the buffer material or during the emplacement of the bentonite blocks, especially if large void spaces were left between bentonite blocks. Swelling of the bentonite beneath the canister may cause the canister to initially rise upwards in the disposal hole (to ‘heave’) as the swelling pressures and density of the bentonite are lowest at the top of the disposal hole where the bentonite expands into the tunnel backfill (SKB, 1999).

After the bentonite buffer has completely resaturated, there is a possibility that the very heavy and dense canister might then slowly sink through the bentonite. Some degree of sinking is inevitable as the bentonite beneath the canister is consolidated after hydraulic resaturation. However, the canister may continue to sink through the water-saturated buffer if the buffer exhibits rheological and viscous properties which allow it to flow slowly. However, investigations of this issue as part of the Swiss Kristallin-I performance assessment indicate that it is extremely unlikely that a canister could sink through the full thickness of a bentonite buffer (Nagra, 1994).

Movement of the canister is also possible once the canister metal begins to corrode. The secondary corrosion products all have higher molar volumes than the original metal and thus will expand, creating increasing mechanical stresses on both the canister and the buffer. If uneven stresses built-up around the canister these may be relieved by movement of the canister sideways (see SFL-51 “Expansion of solid corrosion products”). However, since the copper shell will corrode very slowly, this is unlikely to be a significant problem (Werme, 1998).

Once the copper shell has been perforated, anaerobic corrosion of the iron insert will generate hydrogen gas which may form a free gas phase (see SFL-25 “Gas generation at the canister”). The expansion of the gas bubble and the increase in the gas pressure will also generate increasing mechanical stresses on both the canister and the buffer. It is possible that the gas pressure could be uneven around the canister but this too is unlikely to be a significant problem.

External forces might also bring about movement of the canister. For example, rock mass movement by faulting, creep of the rock mass and collapse of the repository openings could potentially cause disruption to the buffer and movement of the canister within the buffer (see SFL-30 “Mechanical impact on the canister”).

The likelihood of canister movement is largely controlled by the physical properties of the bentonite buffer (swelling pressure, strength, viscosity etc). These properties could be altered by chemical alteration and mineralogical changes to the bentonite (see SFL-3 “Chemical alteration of the buffer and backfill”) which might cause a loss of swelling capability or cementation.

Canister movement (especially sinking) would also be exacerbated if the swelling pressure and density of the buffer were to drop due to dilution of the buffer (see SFL-16 “Dilution of the buffer and backfill”). This could occur as the bentonite expands into fractures in the near-field rock, the void spaces in the canister (after canister failure) or due to erosion of the buffer by flowing groundwater.

Movement of the canister in the buffer would potentially reduce the capability of the buffer to act as a barrier to groundwater flow and colloid transport. Radionuclide releases to the near-field rock could be accelerated due to reducing the distance across which diffusion only transport would occur in the buffer and by limiting the opportunity for radionuclide sorption to occur on the buffer minerals.

Origin in the repository system:

Movement of the canister in the bentonite buffer might occur due to uneven pressures acting on the canister as a result of swelling of the bentonite; expansion of solid canister corrosion products and gas; gravity - sinking of the canister; and external mechanical impacts from the rock.

The extent and likelihood of canister movement is controlled by the physical properties of the bentonite, most importantly its swelling pressure and viscosity. These physical properties may alter over time due to mineralogical changes (affected by groundwater chemistry) and dilution of the buffer.

Impact on the repository system:

Movement of the canister in the bentonite buffer will bring some part of the canister closer to the near-field rock and, thus, will reduce the thickness of the bentonite barrier. Extreme movement could, potentially, bring the canister into direct contact with the rock which would entirely negate the buffer functions. Movement of the canister in the bentonite buffer could potentially thus increase the radionuclide release times to the near-field rock.

Bibliographic references:

Nagra (1994) Kristallin-I: safety assessment report. Nagra Technical Report, NTB 93-22.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.07 (Mechanical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.2 (Movement of canister in buffer/backfill) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-34**

Name: **PREFERENTIAL TRANSPORT PATHWAYS IN THE CANISTER**

Short description:

Once the canister has failed, groundwater, solutes, colloids and gases will migrate through the canister and eventually radionuclides may be released to the bentonite buffer. The transport pathways through the canister will evolve over time as void spaces become filled with iron corrosion products, intruding bentonite, gas bubbles etc., and new perforations in the canister create additional inflow and outflow points. As such, preferential transport pathways in the canister will be generated but these may change with time.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

Preferred pathways in the canister cannot form until after the canister has failed. Thus the primary control on the formation of preferential pathways in the canister is the lifetime of the canister, controlled primarily by the corrosion rate of the copper shell.

Once both the outer shell and the iron insert have failed, groundwater can access the central void spaces and contact the spent fuel. After initial canister failure, the pathway for groundwater access to the spent fuel will be via the hole(s) in the copper shell, through the annular gap between the shell and the iron insert, through the hole(s) in the iron insert and then through hole(s) in the Zircaloy cladding tube. The first radionuclides released from the fuel assemblies will be transported out of the canister along the reversed route, these nuclides are likely to be the prompt releases from the gap between the cladding and the spent fuel (see SFL-22 “Gap and grain boundary release”).

Over time, a number of processes will occur which will change the pathways through the canister. As the groundwater reacts with the iron insert, large volumes of solid iron corrosion products (see SFL-51 “Expansion of solid corrosion products”) and hydrogen gas will be generated (see SFL-25 “Gas generation in the canister”). In addition, bentonite might extrude into the canister as a consequence of its high swelling pressure (see SFL-16 “Dilution of the buffer and backfill”). Together, the increasing volumes of iron corrosion products, gas and bentonite will gradually fill the void spaces in the canister and the annular gap between the shell and the iron insert. In addition, the annular gap could be reduced and even closed due to creep of the copper (see SFL-11 “Creeping of the metal in the canister”) and the Zircaloy cladding will continue to corrode.

The consequence of all these processes will be to restrict the free movement of groundwater, colloids and gas flow through the canister. The overall impact may be to slow down radionuclide releases and continued degradation of the spent fuel (by restricting water access to the fuel). As the void spaces are reduced, water and gas movement will be limited to particular preferred pathways through the corrosion products and the intruding bentonite. Gas bubbles formed inside the canister will rise upwards until they become trapped, further restricting water movement through the canister (see SFL-23 “Gas escape from canister”).

As the canister continues to corrode, additional perforations may occur in the canister wall. These will provide additional inflow and outflow points for water and gas turnover in the canister (see SFL-56 “Water turnover in the copper shell” and SFL-57 “Water turnover in the cast iron insert”). The geometry of these additional perforations in relation to the location of void spaces in the canister may cause existing preferred pathways to be modified or new preferred pathways to be created.

The net effect of these processes over time is thus initially to create preferred pathways through the canister and then to cause the geometry of these pathways to become time dependent, in response to ongoing corrosion of the canister and intrusion of the bentonite buffer. Thus the geometry of these preferred pathways will be dependent on the corrosion rate of the iron insert (controlled in part by the composition of the groundwater) and the bentonite swelling pressure.

The creation and alteration of preferred pathways through the canister will change the rate of radionuclide release from the canister due to solute and colloid movement in groundwater, and as a gas phase. The creation of new pathways will increase radionuclide release but, if the pathways later become restricted (e.g. filled by corrosion products), then the radionuclide release rate may fall. In addition, preferred pathways will tend to restrict groundwater access to the spent fuel which will limit continued spent fuel leaching and radionuclide release.

Origin in the repository system:

Preferred pathways in the canister cannot form until after the canister has failed. Thus the primary control on the formation of preferential pathways is the lifetime of the canister, controlled primarily by the corrosion rate of the copper shell. Once the canister has failed, preferential pathways are generated as a consequence of the corrosion of the iron insert (generating expanding volumes of solid corrosion products and hydrogen gas) and the intrusion of bentonite due to its high swelling pressure.

As these processes continue, the geometry of the preferred pathways will change at a rate controlled by the rate of iron corrosion and the intrusion of bentonite.

Impact on the repository system:

The creation and alteration of preferred pathways through the canister will change the rate of radionuclide release from the canister due to solute and colloid movement in groundwater, and as a gas phase. The creation of new pathways will increase radionuclide release but, if the pathways later become restricted (e.g. filled by corrosion products), then the radionuclide release rate may fall. In addition, preferred pathways will tend to restrict groundwater access to the spent fuel which will limit continued spent fuel leaching and radionuclide release.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.02 (Wasteform materials and characteristics) and 2.1.03 (Container materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 1.5 (Release of radionuclides from the failed canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-35**

Name: **RADIATION EFFECTS ON THE BUFFER AND BACKFILL**

Short description:

The buffer and the backfill may be exposed to radiation from the spent fuel that penetrates the canister. This radiation potentially can affect the mineralogical structure of the bentonite minerals causing modifications to the characteristics of the bentonite material, such as the swelling capacity and sorption capacity.

Technical description:

The bentonite buffer may be exposed to radiation from the spent fuel that penetrates the canister. In addition, both the bentonite buffer and the bentonite-rich backfill may be exposed to radiation from dissolved radionuclides passing or precipitating in the pores of the buffer, the backfill and the near-field rock. The radiation will be time-dependent and controlled by radioactive decay (see GEN-28 “Radioactive decay”).

Theoretically, this radiation could affect the mineralogical structure of the bentonite minerals and, ultimately, could initiate mineralogical transformations (see SFL-3 “Chemical alteration of the buffer and backfill”). If this occurred, it may cause modifications to the characteristics of the bentonite material, such as the swelling capacity and sorption capacity. Thus, a number of experiments have been performed to investigate the potential significance of this process to repository safety.

Krumhansl (1985) exposed clay derived from bentonite to high radiation fields at ambient temperature and reported no crystallographic changes to the clay material. Allen and Rawson (1986) exposed bentonite consisting of sodium montmorillonite (85 %), quartz, feldspar, cristobalite, illite calcite and gypsum to high radiation fields at elevated temperatures (300 °C) for two weeks. The total absorbed dose was estimated to be more than one order of magnitude more than would be experienced during a 1000 year period in a repository. Investigation of the clay structure after irradiation using transmission electron microscopy (TEM) showed no mineralogical or crystallographic changes. However, X-ray diffraction (XRD) analysis detected a decrease in the smectite clay’s <001> basal spacing, indicating a loss of loosely bound water from the clay structure. This loss of interlayer water was reversible upon rehydration with water. Other diffraction peaks showed no shifts which argued against loss of significant amounts of structural water from the clay. The swelling capacity of the clay was not affected.

Haire and Beall (1979) exposed bentonite to high doses of alpha radiation. Total absorbed doses ranged from 4.8×10^9 to 4.8×10^{11} rad. Some loss of clay crystallinity was noted in the experiments with the highest radiation doses. These every high dose rates may not be relevant to a repository environment.

The conclusion from these experimental studies is that bentonite shows little evidence of crystallographic change to the clay structure due to radiation. Consequently, it is unlikely that this process will have any significant impact upon repository safety.

Radiation damage to the bentonite buffer or backfill is not considered in standard performance assessment calculations.

Origin in the repository system:

Radiation emitted from the spent fuel in the canister will impact on the bentonite buffer. The level of radiation exposure is time dependent and is controlled by the thickness of the canister and by radioactive decay.

Radiation emitted from radionuclide-bearing precipitates in the buffer, backfill or near-field rock will also impact on bentonite in the buffer and the backfill. This radiation exposure is also time dependent and is controlled by the time taken for canister perforation to occur, radioactive decay as well as the porosity distribution in the near-field and the groundwater chemistry which controls radionuclide solubility.

Impact on the repository system:

Radiation exposure of the buffer and backfill may affect the mineralogy and structure of the bentonite. This potentially could influence the characteristics of the bentonite material, such as the swelling capacity and sorption capacity. However, laboratory evidence suggests that the process is insignificant with regards to repository safety.

Bibliographic references:

Allen CC and Rawson SA (1986) Effects of irradiation and dry heating on bentonite: a transmission electron microscopy and X-ray diffraction study. Microbeam Analysis, 86, 135-137.

Haire RG and Beall GW (1979) Consequences of radiation from sorbed transplutonium elements on clays selected for waste isolation. American Chemical Society Symposium Series 100, 291-295.

Krumhansl JL (1985) Observations regarding the stability of bentonite backfill in a high-level waste repository in rock salt. Sandia National Laboratories Report SAND83-1293.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.13 (Radiation effects in wastes and EBS) and 2.1.04 (Buffer/backfill materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.13 (Radiation effects on bentonite) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Dave Savage and Bill Miller

Number: **SFL-36**

Name: **RADIATION EFFECTS ON THE CANISTER**

Short description:

The canister may be exposed to radiation from the spent fuel or from radionuclide-bearing precipitates in the buffer. This radiation potentially can affect the canister metals causing modifications to their corrosion rate and their mechanical properties.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

The canister metals will be exposed to radiation from the spent fuel and from dissolved radionuclides passing or precipitating in the pores of the buffer. The radiation will be time-dependent and controlled by radioactive decay (see GEN-28 “Radioactive decay”).

Theoretically, this radiation could affect the metals of the canister. If this occurred, it may cause modifications to their corrosion rate (see SFL-7 “Corrosion of the copper shell” and SFL-9 “Corrosion of the cast iron insert”) and their mechanical properties (see SFL-39 “Reduced mechanical strength of the canister”). Thus, a number of experiments have been performed to investigate the potential significance of this process to repository safety.

The effects of radiation on the performance of waste canisters have been reviewed by Shoesmith et al. (1992) who noted two effects. Firstly, photo-radiation effects within passive, semiconducting layers could cause electron-hole separation processes which may change the properties of the layer, thereby affecting the initiation of localised corrosion on passive corrosion-resistant materials. Secondly, the production of radiolytic oxidants and reductants could directly affect the kinetics of anodic and cathodic reactions.

These authors concluded that, for copper, the effect of radiation on the general corrosion rate is only significant at dose rates • 5 Gy/h. At lower dose rates there is some evidence that radiation reduces general corrosion rates, either by inducing the formation of more compact films of corrosion products, or by affecting the chemistry within the backfill .

The effect of radiation on localised corrosion processes appears to be specific to individual materials. For stainless steel, there is evidence for the enhancement of the protective properties of the passive oxide film which inhibits the initiation of localised corrosion (Marsh et al., 1986). However, during initial oxic conditions in the disposal vaults, there may be a susceptibility to stress corrosion cracking.

The results from laboratory studies suggest that the effect of radiation on the canister is minimal. No direct damage to the canister is caused by the radiation, although the radiation in combination with chemical attack can modify general and localised corrosion rates. However, the effect is considered to be too small to be significant.

Radiation impacts on the canister are generally not considered in standard performance assessment calculations.

Origin in the repository system:

Radiation emitted from the spent fuel will impact on the canister. The level of radiation exposure is time dependent and is controlled by radioactive decay and thickness of the canister. In particular, the effect on the outer copper shell is controlled by the thickness of the inner iron insert.

Radiation emitted from radionuclide-bearing precipitates in the buffer will also impact on the canister. This radiation exposure is also time dependent and is controlled by the time taken for canister perforation to occur, radioactive decay as well as radionuclide migration (required for precipitates to form outside the canister), the porosity distribution in the near-field and the groundwater chemistry which controls radionuclide solubility.

Impact on the repository system:

Radiation potentially can affect the canister metals causing modifications to their corrosion rate and their mechanical properties. However, laboratory evidence suggests that the radiation causes very little impact on the metals directly but can cause a change to the corrosion rate in combination with chemical attack. Nonetheless, this process is considered to be insignificant with regards to repository safety.

Bibliographic references:

Marsh GP, Taylor KJ, Bryan G and Worthington SE (1986) Corrosion Science, 26, 971.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Shoesmith DW, Ikeda BM and King F (1992) Effect of radiation on the corrosion of candidate materials for nuclear waste containers. Material Research Society, Scientific Basis for Nuclear Waste Management, XV, 407-414.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.13 (Radiation effects in wastes and EBS) and 2.1.03 (Container materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 2.3.5 (Radiation on canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-37**

Name: **RADIOLYSIS INSIDE THE CANISTER PRIOR TO WETTING**

Short description:

The void spaces inside the iron insert of the canister may contain small amounts of water or vapour when the canister is initially sealed. The intense radiation field inside the canister may cause radiolytic decomposition of the water to generate nitrous and nitric acids. These acids may accelerate corrosion of the iron insert and the metallic components of the fuel assemblies, before the copper shell is perforated and groundwater can infiltrate the canister.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

The void spaces within the iron insert will contain some water or vapour when the canister is originally sealed, even in the absence of manufacturing faults. The water is most likely to be present in the fuel pins. Although these will be dried prior to emplacement in the canister, pessimistic assumptions are that one 'wet' fuel pin will be placed in a canister (SKB, 1999). The water contained in the fuel pins may initially be trapped by the intact Zircaloy cladding tubes. This water cannot migrate into the central void spaces of the canister until the cladding tubes are breached.

The high initial temperature (see SFL-46 "Temperature of the near-field") will mean that the water within the iron insert occurs as water vapour. This vapour will be subjected to the intense radiation field inside the canister from the spent fuel, causing the radiolytic generation of nitrous and nitric acids (Henshaw, 1994). In addition, anaerobic iron corrosion of the iron insert by the water vapour will also generate hydrogen. Continued radiolysis in an atmosphere containing hydrogen and nitrogen compounds can generate ammonia.

The build-up of corrosive ammonia and nitric acids may lead to accelerated general corrosion and localised stress corrosion cracking of the iron insert at points on the canister subjected to tensile stresses (see SFL-10 "Canister corrosion prior to wetting"). However, the nitric acid generation rate will be minimised because of the rapid consumption of water by the iron corrosion processes (Werme, 1990). As such, the impact of nitric acid generation is expected to be minimal.

All of these reactions could be limited to some extent by more efficient drying of the fuel and by purging the canister with inert gas (e.g. Ar) prior to sealing. This reduces the fraction of the radiation absorbed by nitrogen, oxygen and water and, thereby, also the rate of conversion to nitric acid.

Overall, corrosion prior to wetting will change the local chemical environment inside the canister before it is perforated and can lead to slightly earlier canister failure times.

Origin in the repository system:

Radiolysis prior to wetting can occur inside a sealed canister. This process is dependent on the introduction of water with the fuel assemblies, prior to canister sealing. The amount of water introduced can be limited by drying the fuel and purging the void spaces with an inert gas (e.g. Ar). Procedures for fuel and canister handling are, thus, important controls on this process.

If water is present inside the canister, the important controls on radiolysis prior to wetting are the amount of water (vapour) and the radiation field inside the canister (which relates to burn-up and storage times before emplacement in the canister).

Impact on the repository system:

Radiolysis prior to wetting affects local chemical environment inside the canister before it is perforated. In particular, it controls the formation of corrosive agents such as nitrous and nitric acids, and ammonia which will affect corrosion of the canister prior to water intrusion and, can lead to slightly earlier canister failure times.

Bibliographic references:

Henshaw J (1994) Modelling of nitric acid production in the advanced cold process canister due to irradiation of moist air. SKB Technical Report, TR 94-15.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme, L. (1990) Near-field performance of the advanced cold process canister. SKB Technical Report 90-31.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 2.1.13 (Radiation effects in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 1.2.1 (Radiolysis) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-38**

Name: **REDOX FRONTS**

Short description:

Redox fronts may be generated in both the near and far-fields, at the interfaces between groundwaters with different redox potentials. The location of a redox front may change with time if one body of water is moving relative to the other; for example, the redox front generated by the downward infiltration of oxic recharging groundwaters. The generation and propagation of a redox front will affect radionuclide transport because radionuclide solubilities could be substantially different on either side of the front. This geochemical instability may also result in the generation of colloids.

Technical description:

A redox front is the chemically defined interface between two bodies of water with different redox potentials. In the repository system and the far-field, redox fronts may be generated at a number of locations.

Redox fronts may occur in the far-field rock, close to the surface where downward moving (recharging) oxidising groundwaters flow into deeper and older chemically reducing groundwaters. This redox front is entirely natural and occurs independently of the presence of the repository (see GEN-24 “Interfaces between different waters”)

Redox fronts may also occur in the near-field rock, close to the repository excavations during the transient period after repository closure, when chemically reducing groundwaters hydraulically resaturate the near-field and flow into the rocks which had been made oxidising by contact with ventilated air through the open excavations (see GEN-32 “Hydraulic resaturation of the near-field rock”).

Lastly, redox fronts may arise in the engineered barriers by the generation of oxidants due to radiolysis of the groundwater by the spent fuel (see GEN-30 “Radiolysis”).

The first redox front, close to the surface, will migrate downwards as a continuous supply of oxidising groundwaters gradually consumes the redox buffering capacity of the shallow rock mass. The redox front will also be driven downwards by continued erosion of the land surface (see GEN-29 “Erosion and weathering”). The rate of redox front propagation will generally be slow but, during periods of glaciation, much larger volumes of oxidising waters at the front of a glacier may flush through the rock, causing the redox front to penetrate to considerable depth along fast flowing fractures (Arthur, 1996) and, in extreme conditions, may approach the repository near-field (see GEN-21 “Glaciation”).

The second redox front, caused by repository excavation, will be short-lived (a few hundred years) because the oxidised rock and porewaters will quickly be converted by redox reactions involving the ambient chemically reducing groundwaters, from the rock, which will resaturate the near-field (see GEN-22 “Far-field groundwater chemistry”). This process should have no significance for repository performance because this transient feature will have ceased long before canister failure.

The third, radiolytically generated, redox front will be controlled by the balance between the rate of radiolytic oxidant generation, and the rate of consumption of these oxidants by the iron canister insert, the spent fuel and dissolved species in the groundwater. Potentially, if the rate of oxidant generation is fast, the redox front may grow and migrate out of the canister into the bentonite buffer. However, this is considered to be an unlikely event because the very long lifetime of the canister means that the radiation field will be quite weak by the time the canister fails and water can contact the fuel, and because the radiolytic oxidants should be rapidly consumed within the canister (SKB, 1999).

The generation of a redox front and its propagation is dependent on a supply of oxygenated water or radiolytically generated oxidants, and the rate at which these oxidised waters may be consumed in redox reactions involving the very large volumes of iron in the rock and the engineered barriers.

Current understanding of oxidant production and consumption processes in the near-field suggests that, although radiolysis will occur, oxidant consumption will be sufficiently rapid to stop the near-field environment from becoming permanently oxidising. Instead, the oxidising zone (redox front) will remain localised at the spent fuel surface, where the majority of radiolytic oxidants will be consumed by reaction with the spent fuel (see SFL-21 “Spent fuel dissolution and conversion”) or by molecular hydrogen (SKB, 1999). This has been supported by investigations of radiolytic processes and UO_2 oxidation in the Cigar Lake natural analogue study (Smellie and Karlsson, 1996).

If extensive oxidising conditions can be maintained inside the canister, and a redox front around the canister is established, this will have a significant effect on repository performance. Corrosion of the spent fuel and the canister would be accelerated because these degradation processes are faster in an oxidising environment. Radionuclide releases from the canister would also be enhanced because radionuclide solubilities are generally higher in oxidising environments (see SFL-49 “Radionuclide release and transport from the canister”).

However, mobile radionuclides (and other redox sensitive species) would be precipitated once they cross the redox front and encounter the chemically reducing waters on the other side (Carnahan, 1988). This process may cause a reconcentration of radionuclides on the reducing side of a redox front. Such phenomena have been studied at the Osamu Utsumi mine in Brazil as part of the Poços de Caldas natural analogue study (MacKenzie et al., 1990). Precipitated species on the reducing side of the front may form colloidal particles and thus increase the colloid population and the potential for colloid mediated radionuclide transport.

Origin in the repository system:

Redox fronts are a natural feature of the near-surface environment where oxidised groundwaters infiltrate downwards into the deeper rock mass. The depth and propagation of this front is controlled by the recharge water volume, the redox buffering capacity of the rock and the erosion rate.

Redox fronts will also occur in the near-field. One is caused by oxidation of the rock walls in the tunnels during the operational phase. This is a transient feature which will be rapidly buffered by the resaturating groundwater. A third redox front may be generated in the failed canister by radiolysis of the groundwater. The propagation of this front will be dependent on the balance between radiolytic oxidant generation and their consumption in redox reactions on the spent fuel, the iron canister insert and dissolved species in the groundwater.

Impact on the repository system:

The establishment and propagation of a redox front in the near-field will affect the rates and mechanisms of corrosion of the spent fuel and the canister. Radionuclide solubility, speciation and transport will be affected by the development of oxidising conditions, although mobilised nuclides will be precipitated on the reducing side of the front if they diffuse across the boundary. Colloids may be formed at the redox front, and the location of the redox front may then affect the location of colloid formation.

Bibliographic references:

Arthur RC (1996) Estimated rates of redox-front migration in granitic rocks (SITE-94). SKI Technical Report, 96-35.

Carnahan CL (1988) Simulation of effects of redox and precipitation on diffusion of uranium solution species in backfill. Materials Research Society, Scientific Basis for Nuclear Waste Management XI, 293-302.

MacKenzie AB, Scott RD, Linsalata P, Miekeley N, Osmond JK and Curtis DB (1990) Natural radionuclide and stable element studies of rock samples from the Osamu Utsumi mine and Morro do Ferro analogue study sites, Poços de Caldas, Brazil. SKB Technical Report, TR 90-16.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Smellie JAT and Karlsson F (1996, editors) The Cigar Lake analogue project: a reappraisal of some key issues and their relevance to repository performance assessment. SKB Technical Report, TR 96-08, SKB, Stockholm, Sweden.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.2.08 (Chemical/geochemical processes and conditions (in geosphere)) in the NEA International Database (NEA, 1998).

Relates to FEP 3.1.11 (Redox front) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Bill Miller

Number: **SFL-39**

Name: **REDUCED MECHANICAL STRENGTH OF THE CANISTER**

Short description:

The mechanical strength of the canister is largely provided by the cast iron insert. A reduction of the mechanical strength can occur due to a number of physical and chemical processes. The effect will be to limit the canister's resistance to failure by mechanical impact.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). During normal repository evolution, the canister will be subject to a number of physical and chemical processes that will slowly reduce the canister's resistance to failure by mechanical impact.

The most significant process will be anaerobic corrosion of the canister metal by the groundwater. Initially, the copper shell will corrode which will cause some reduction in mechanical strength (see SFL-7 "Corrosion of the copper shell"). However, this process is not significant because the copper provides only a small part of the total strength of the canister and this small component is disregarded in performance assessment calculations. Once the copper shell is perforated, the iron insert will begin to corrode by contact with the groundwater (see SFL-9 "Corrosion of the cast iron insert") and, because this provides the canister's support, the canister's mechanical strength will be more rapidly reduced.

Radiation from the waste may also act to reduce the mechanical strength of the canister metal through a process of embrittlement, although this is of secondary significance (see SFL-36 "Radiation effects on the canister").

In addition to chemical attack, the canister will be subject to mechanical impact. The largest mechanical impact will be due to the combined lithostatic and bentonite swelling pressures which, together, can amount to approximately 15 MPa under normal climatic conditions and will be even higher during glaciation (see SFL-1 "Swelling of the bentonite buffer"). Other mechanical impacts can occur on the canister due to mass movement of the host rock by faulting, creep and cave-in.

Together, the combined chemical and physical processes acting on the canister will reduce its resistance to failure by mechanical impact (see SFL-30 "Mechanical impact on the canister"). After its strength has been reduced to the point where it can no longer resist the external pressures acting on it, the canister will collapse if any void space exists within it. If the canister loses strength unevenly over its surface or, if the external forces are heterogeneously distributed, the reduced mechanical strength will cause the canister to deform and fail in the deposition hole.

If either of these processes happened, failure of the canister would occur earlier than would be predicted solely on the basis of metal corrosion rates. Failure of the canister by loss of mechanical strength would allow the groundwater to contact the wasteform and begin to corrode.

Deformation or collapse of the canister would also cause a mechanical impact on the bentonite buffer (see SFL-31 “Mechanical impact on the buffer and backfill”) and could, potentially, lead to the opening of preferential pathways through the bentonite, especially if the bentonite had been partially cemented.

Origin in the repository system:

A reduction in the mechanical strength of the canister will come about largely due to chemical (metal corrosion) and physical (external pressures) processes acting on it. The rate of loss of strength will be controlled by the rates of these processes and, thus, are linked to the chemistry and temperature of the groundwater (controlling metal corrosion) and the stress field in the rock mass and characteristics of the bentonite (controlling the external pressures acting on the canister).

Impact on the repository system:

A reduction in the canister’s mechanical strength will make the canister more susceptible to mechanical impact and can cause a reduction in the lifetime of the canister, leading to the early release of radionuclides.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 2.1.03 (Container materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 2.3.5 (Radiation effects on canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Tim McEwen and Bill Miller

Number: **SFL-40**

Name: **RADIONUCLIDE RELEASE FROM THE SPENT FUEL MATRIX**

Short description:

The majority of radionuclides in the fuel assemblies are contained within the spent fuel matrix. After canister failure, when groundwater comes into contact with the spent fuel, these radionuclides can be released by dissolution and oxidative conversion of the fuel. The rate of radionuclide release depends on the groundwater chemistry in contact with the fuel, the radiation field which controls radiolysis of the groundwater, and the water turnover in the canister. Radionuclide release from the spent fuel matrix is the primary control on the radionuclide content of the groundwater in the canister.

Technical description:

The majority of radionuclides in the fuel assemblies are contained within the spent fuel matrix. The remainder are contained in the gap between the fuel pellets and the Zircaloy cladding and in grain boundaries (see SFL-22 “Gap and grain boundary release”), and as activation products in the metallic components of the fuel assemblies (see SFL-41 “Radionuclide release from metal non-fuel parts”).

After emplacement, when the canister is intact, radionuclide release from the spent fuel UO_2 matrix will be limited to reaction with the small amounts of air or water trapped in the void spaces within the canister. These reactions could be minimised by purging the canister with inert gas prior to sealing. However, even if this were not done, the very small amounts of water which are likely to be present in a sealed canister will mean that dissolution and conversion reactions prior to canister failure will be negligible.

After failure of the canister and the Zircaloy cladding, groundwater will come into direct contact with the surface of the spent fuel and radionuclide releases from the fuel can be initiated. The first stage of radionuclide release involves dissolution of the UO_2 matrix (see SFL-21 “Spent fuel dissolution and conversion”). The rate of the dissolution reaction is controlled by the solubility of uranium in the groundwaters adjacent to the spent fuel and the rate of uranium transport out of the canister. Radionuclides would be released from the spent fuel matrix at a rate determined by the uranium dissolution rate. Since the solubility of uranium under these conditions is low and the rate of water turnover in the canister is also low, this radionuclide release rate will be very slow.

Many investigations have been made about whether radionuclide release mechanism is congruent, that is, components are released into solution in the same atomic proportion as they occur in the dissolving solid. The irreversible dissolution of unstable phases does occur congruently. Apparent incongruent dissolution reactions can arise from selective precipitation/adsorption of released components, which is commonly observed in wasteform dissolution studies. Congruency also may be masked by the dissolution of two or more different solids (e.g. UO_2 and gap phases) that contain one or more of the same components (e.g. ^{137}Cs, ^{90}Sr).

Radiation emitted from the spent fuel matrix will cause radiolysis of the water adjacent to the fuel surface (see GEN-30 “Radiolysis”). These radiolytically generated oxidants can cause oxidative conversion of the UO_2 spent fuel matrix, which might be associated with an increased dissolution rate. Oxidative conversion to U_3O_7 or UO_3 or some other U(VI) compound, will alter the crystal structure of the fuel, causing the direct expulsion of some proportion of the radionuclides from the uranium oxide matrix. Thus, for these higher states, the radionuclide release rate would be controlled directly by the rate of fuel conversion rather than the rate of uranium dissolution (see SFL-21 “Spent fuel dissolution and conversion”).

The total radionuclide release from the spent fuel matrix is therefore dependent largely on rate of spent fuel dissolution and conversion which, itself, is controlled by the composition of the groundwater in contact with the fuel and the intensity of the radiation field causing groundwater radiolysis. The lifetime of the canister will be a primary control on the radiation field at the time groundwaters can contact the spent fuel. Radioactive decay will also change the inventory available for release.

The total radionuclide release from the spent fuel matrix will change the groundwater composition in the canister and in the bentonite buffer. Transport of dissolved species away from the spent fuel by diffusion (see SFL-15 “Diffusion in and through the canister”) or water turnover (see SFL-56 “Water turnover in the copper shell” and SFL-57 “Water turnover in the cast iron insert”) will reduce induce further dissolution of solubility limited species. Therefore, mass transport of dissolved matrix components away from the waste-form surface will result in a corresponding net dissolution of a stable waste matrix. For expected near-field conditions with an impermeable backfill, transport will be largely diffusion limited. The release rate from the wasteform will thus be controlled by the solubility-limited, diffusive mass-transfer rate of the components through the canister and buffer (e.g. Chambré et al., 1988).

Origin in the repository system:

The total radionuclide release from the spent fuel matrix is initially dependent on the lifetime of the canister. No significant releases can occur until after the canister has failed and groundwater contacts the spent fuel.

After canister failure, the total release from the spent fuel is dependent on rate of spent fuel dissolution and conversion, which is controlled by the groundwater chemistry inside the canister, the intensity of the radiation field which causes radiolysis of the groundwater and the rate of water turnover in the canister (which controls the release of solubility limited species).

Impact on the repository system:

The total radionuclide release from the spent fuel matrix controls the groundwater composition (radionuclide content) in the canister and in the bentonite buffer and is, therefore, a primary control on the total releases to the near-field rock.

Bibliographic references:

Chambré PL, Kang C.H, Lee W and Pigford TH (1988) The role of chemical reaction in wasteform performance. Scientific Basis for Nuclear Waste Management, XI, 285-291.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) and 3.2.02 (Speciation and solubility, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.2.6 (Solubility within the fuel matrix) and 1.2.9 (Dissolution chemistry) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-41**

Name: **RADIONUCLIDE RELEASE FROM THE METAL NON-FUEL PARTS**

Short description:

Complete fuel elements will be placed in the canister and these comprise metallic structural parts in addition to the UO_2 fuel. The metallic components contain various activation products due to neutron irradiation in the reactor. When these metallic components begin to corrode, after canister failure, they will release the activation products to the groundwater.

Technical description:

The reference spent fuel canister will be loaded with complete fuel elements: twelve fuel elements from a boiling water reactor (BWR) or four fuel elements from a pressurised water reactor (PWR). The construction of the BWR and PWR fuel elements are different to each other in detail but similar in basic form (Werme, 1998). A fuel element comprises many (50 to 300) individual fuel rods, each stacked with UO_2 fuel pellets. The fuel rods are made of Zircaloy (zirconium alloy) with welded Zircaloy end plugs to form hermetically sealed units. The fuel rods are fixed together to form single fuel elements by stainless steel, Inconel and Incoloy (nickel alloys) spacer grids and upper and lower tie-plates etc. Because complete fuel elements are loaded in the canister, it means that significant masses of metallic non-fuel components are present in the waste.

Corrosion of the metal non-fuel waste parts may begin as soon as the canister is sealed, even if they are constructed and filled without faults (see SFL-8 “Corrosion of the metal non-fuel waste parts”). If air or water are trapped in the void spaces within the canister, water, oxygen and nitrogen can cause corrosion of the metal parts by redox reactions. These reactions could be limited by purging the canister with inert gas prior to sealing. However, even if this were not done, if is unlikely that significant corrosion could occur prior to canister failure.

Once the outer copper shell and, subsequently, the iron insert have been perforated, groundwater can infiltrate into the canister and contact the fuel elements. Corrosion of the metal non-fuel waste parts will release the radioactive activation products they contain. At the time of canister failure (taken to be 10^4 years), the activation products calculated to be present (TVO, 1992) in Zircaloy are ^{14}C, ^{36}Cl and ^{93}Zr and, in the other metal structural parts, ^{14}C, ^{59}Ni, ^{60}Co and ^{94}Nb. In addition, corrosion of the metal non-fuel waste parts will release any radionuclides precipitated or sorbed onto the metal surfaces.

Radionuclides may be released congruently from these metallic parts at rates that depend on the corrosion rate of the different metals. Zircaloy has been studied in most detail because it is used to manufacture the fuel cladding and, thus, controls the access of groundwater to the fuel after the canister has been breached. The corrosion of Zircaloy is passivated by a zirconium dioxide film which forms and is tightly bound to the metal surface. This ZrO_2 is extremely insoluble (Bruno et al., 1997) and, thus, the radionuclide release rate from Zircaloy will be very low. Experimental and modelling studies suggest that the Zircaloy cladding might have a life of around 100 000 years (Mattsson and Olefjord, 1990).

The corrosion rate of the other metals and, therefore, their associated radionuclide release rates have not been investigated in as much detail. However, on the basis of experimental data (e.g. Gdowski and Bullen, 1988) the corrosion rates of stainless steel, Inconel and Incoloy will also be slow but generally not as slow as for Zircaloy.

The release of soluble species such as ^{14}C will affect the groundwater chemistry (radionuclide content) and, thus, the total radionuclide release rate from the canister to the buffer. The release of insoluble species will result in their incorporation in other secondary phases or sorption onto the surfaces of metals or metal corrosion products (see SFL-28 “Radionuclide interaction with corrosion products”).

Origin in the repository system:

Radionuclide releases from the metallic non-fuel components occurs congruently in response to corrosion of the metals. The radionuclide release rates is, thus, dependent on the corrosion rate which is, itself, controlled by the chemistry of the groundwater within the canister, the temperature and the solubility of any oxide layers forming on the metal surface.

Impact on the repository system:

Radionuclide releases from the metallic non-fuel components can affect the radionuclide content of the groundwaters (groundwater chemistry) for the soluble species and the releases from the canister to the buffer.

Bibliographic references:

Bruno J, Cera E, de Pablo J, Duro L, Jordana S and Savage D (1997) Determination of radionuclide solubility limits to be used in SR97: uncertainties associated to calculated solubilities. SKB Technical Report, TR 97-33.

Gdowski GE and Bullen DB (1988) Survey of degradation modes of candidate materials for high-level radioactive waste disposal containers: oxidation and corrosion. Lawrence Livermore National Laboratory Technical Report, UCID-21362, Volume 2.

Mattsson H and Olefjord I (1990) Analysis of oxide formed on titanium during exposure in bentonite clay - I: the oxide growth. Werkstoffe und Korrosion, 41, 383-390.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

TVO (1992) TVO-92 safety analysis of spent fuel disposal. YJT Technical Report, YJT-92-33-E.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 3.2.01 (Dissolution, precipitation and crystallisation of contaminant) in the NEA International Database (NEA, 1998).

Relates to FEP 1.5 (Release of radionuclides from failed canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-42**

Name: **HYDRAULIC RESATURATION OF THE BUFFER AND BACKFILL**

Short description:

After closure of the repository, groundwater will flow from the resaturating near-field rock into the buffer and backfill causing them to hydraulically resaturate. Groundwater will be taken up by the bentonite in the buffer and backfill causing these materials to expand, resulting in a general homogenisation of physical and chemical characteristics. This resaturation will impact on the thermal, hydraulic and stress fields in the near-field, and the near-field groundwater chemistry.

Technical description:

During operation of a repository, groundwater flowing into the excavations will be pumped out. This will desaturate the surrounding rock mass and lead to the development of oxidising conditions within the near-field rock. At repository closure, pumping is stopped and hydraulic resaturation of the repository will begin. Initially, the near-field rock will be resaturated (see GEN-32 “Hydraulic resaturation of the near-field rock”).

Groundwaters will then flow from the near-field rock into the buffer and backfill, initiating the hydraulic resaturation of these materials. As groundwater moves into the buffer and backfill, the bentonite in these materials will take up the groundwater causing it to swell and expand to fill any void spaces and cracks in the near-field rock (see SFL-1 “Swelling of the bentonite buffer” and SFL-45 “Swelling of the tunnel backfill”). The flow of groundwater from the near-field rock into the buffer and backfill will occur via the fracture network, hence entering as discrete line or point sources depending on the degree of channelling within the fractures. This may result in uneven uptake of groundwater and swelling.

During the resaturation event, large thermal and hydraulic gradients will occur across the buffer and, to a lesser extent, across the backfill. Groundwater moving toward the canister will be subject to increasing temperatures and the water at the resaturation ‘front’ may begin to evaporate. This would result in the precipitation of any dissolved species from the groundwater, such as carbonates, and their precipitation could cause the cementation of the buffer minerals, leading to a reduction in swelling capacity and hydraulic conductivity.

Evaporation may also cause a change in the ion concentration gradient in the infiltrating groundwater, leading to osmotic forces acting on water penetration through the buffer and backfill. The principal driving force for the liquid phase water is the porewater pressure gradient across the buffer and backfill. The creation of a vapour phase alters the resaturation mechanism and vapour transport is largely driven by the thermal gradient (see SFL-46 “Temperature of the near-field”).

The rate of propagation of the resaturation ‘fronts’ through the buffer and backfill (and thus the duration of the resaturation event) will be controlled by numerous features and mechanisms in the near-field, including the initial moisture content of the unsaturated buffer and backfill materials; the thermal gradient causing evaporation; the water transport rate through the partially resaturated material (porewater pressure gradient); and the groundwater flow-rate through the near-field rock.

The rate of resaturation may also be affected by the chemistry of the infiltrating groundwater. High salinity groundwaters limit the swelling rate of the bentonite and can initially leave open void spaces for the passage of water. This has the potential advantage that the resaturation of the buffer and backfill can occur more rapidly than for low salinity waters. It is unclear if the final swelling pressure is affected (SKB, 1999).

Groundwater supply to the buffer and backfill will be restricted by any shotcrete coating (if used) on the walls of the excavations. However, this shotcrete is not intended to be a post-closure hydraulic barrier and will only delay resaturation for a short period of time.

Once the buffer and backfill are completely resaturated there will tend to be a general homogenisation of their physical and chemical properties. Due to the lower bentonite content in the backfill, its final swelling pressure will be lower, and its hydraulic conductivity higher, than that in the buffer.

Resaturation of the buffer and backfill will impact on the thermal, stress and hydraulic fields across the near-field. During the resaturation process, chemical conditions in the buffer and backfill will shift from oxidising (due to trapped air in the void spaces) to progressively increasing reducing conditions as this air is dissolved into the groundwater and consumed by oxidation reaction involving iron in the engineered barrier materials and the rock (see SFL-38 “Redox fronts”), and some microbial oxygen consumption. When resaturation of the buffer is completed, groundwaters will contact the copper shell of the canister, initiating aqueous corrosion processes (see SFL-7 “Corrosion of the copper shell”).

Origin in the repository system:

Resaturation of the buffer and backfill will be controlled by the flow of groundwater through the near-field rock, the initial moisture contents of the buffer and backfill materials, the thermal gradient and the changing water transport rate through the initially partially resaturated materials. The principal driving force for the liquid phase water is the porewater pressure gradient across the buffer and backfill. The principal driving force for the vapour phase water is the temperature gradient.

Impact on the repository system:

Resaturation of the buffer and backfill is required for the repository system to function in accordance with its designed performance. Resaturation will impact on most aspects of the near-field, including the thermal gradient, the near-field stress field, the hydraulic gradient and flow patterns and the near-field groundwater chemistry.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 4.2.5 (Changes of groundwater flow) and 5.14 (Resaturation) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-43**

Name: **SEDIMENTATION OF THE BUFFER AND BACKFILL**

Short description:

Sedimentation of the buffer and backfill is possible if these materials become sufficiently diluted that a free suspension in the groundwater could form. Sedimentation from the suspension would create a bentonite-rich sediment overlain by ponded groundwater. This process would cause a significant reduction in the barrier function of the buffer and backfill but is very unlikely to occur in a normal repository evolution scenario.

Technical description:

Dilution of the buffer and backfill is possible if large masses of these materials are removed (eroded) by fast groundwater flow at the interface with the near-field rock or due to a reduction in groundwater salinity which will tend to encourage the bentonite to form a suspension in the groundwater (see SFL-17 “Erosion of the buffer and backfill by flowing groundwater”). Alternatively, dilution of the buffer and backfill could occur if these materials expanded into large void spaces formed by canister failure or the opening of fractures. Likewise, dilution could also occur due to inadequate repository operation procedures, if they resulted in incomplete emplacement of buffer or backfill materials, leaving behind large, unfilled voids (see SFL-16 “Dilution of the buffer and backfill”). However, it is improbable that any of these mechanisms could cause the removal of so much buffer or backfill that further swelling of these materials could not expand to occupy the void spaces created. Nonetheless, if this were to occur, then the remaining void spaces would fill with groundwater and this water might initially create a suspension with the remnant bentonite and buffer material. A bentonite suspension would be most likely to form in low salinity waters.

Sedimentation from the suspension could occur whereby bentonite particles coagulate and then settle out due to gravitational forces to create a layer of bentonite-rich sediment overlain by particle-free ponded groundwater (see SFL-4 “Coagulation of bentonite in the buffer or backfill”). Coagulation and sedimentation will partly depend on the groundwater chemistry, with coagulation being inhibited in low salinity waters. The relative volumes of the sediment and the groundwater layers would depend on the amount of buffer or backfill material removed.

If this unlikely sequence of events were to occur in the disposal holes or the tunnels, the barrier function of the buffer and backfill would be substantially compromised. At the extreme, the buffer could be grossly removed and free water could fill the gap between the canister and the near-field rock. The likelihood of this occurring is dependent on processes taking place which could cause substantial erosion and dilution of these materials. This would presuppose some dramatic change to the groundwater flow system in the near-field rock which could only arise due to some external mechanism (e.g. large magnitude seismic, denudation or climate change events). Sedimentation might increase the radionuclide release rate but, overall, the effects of sedimentation in the disposal holes or the tunnels would, in this case, be negligible compared to the consequences of the event causing the dilution. However, small scale sedimentation events may occur in fractures in the near-field rock whereby small volumes of bentonite eroded from the buffer or backfill settle out to clog the fracture system. This would cause a redistribution of the groundwater flow system (see GEN-23 “Groundwater flow”). Since the fractures are not large, the volume of bentonite needed to clog the fractures would not be large in comparison to the volumes of the buffer and backfill.

Origin in the repository system:

Sedimentation of the buffer or backfill is possible only if substantial dilution of these materials has occurred due either to erosion by groundwater or swelling into voids created by failure of the canister and expansion of fractures in the near-field rock. Sedimentation is then dependent on the water/bentonite ratio in the repository excavations being sufficiently large that a suspension may form. Sedimentation rates from the suspension are controlled partly by the groundwater chemistry.

Impact on the repository system:

Sedimentation of the buffer and backfill in the disposal holes or tunnels will create a free water volume in the repository excavations. This will increase the possibility for radionuclide release but this effect will be negligible compared to the consequences of the event causing the dilution. Sedimentation of bentonite which has been carried by the groundwater into fractures may cause clogging which could modify groundwater flow patterns in the near-field rock.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.1.6 (Sedimentation of bentonite) in SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0   30th June 2000   Bill Miller

Number: **SFL-44**

Name: **SORET EFFECT IN THE BUFFER AND BACKFILL**

Short description:

The Soret effect is the diffusion of chemical species in response to the imposition of a thermal gradient. Theoretically, it has the potential to influence the distribution of chemical species in the buffer and the backfill during the early thermal stage after repository closure.

Technical description:

The Soret effect is diffusion of chemical species under the influence of a thermal gradient and couples the concentration transport to the temperature gradient (Fargue et al., 1998).

The consequence of the Soret effect operating in a solution having both light and heavy solutes will be to concentrate the heavier molecules in the cold region.

In the repository, radioactive decay (see GEN-28 “Radioactive decay”) will generate heat and, thus, a temperature gradient across the bentonite buffer and in the bentonite-rich backfill (see SFL-46 “Temperature of the near-field”). Theoretically, therefore, it is possible that the Soret effect would operate and could influence the radionuclide concentrations (distributions) in the bentonite porewaters.

However, there is little or no experimentally obtained information about Soret effects in bentonite-water systems (Soler, 1999). While it is possible that it will occur, it is likely that the Soret effect will have only minor significance for the behaviour of radionuclides in the near-field.

The Soret effect has not been considered in the primary calculations undertaken in any performance assessment.

Origin in the repository system:

The Soret effect is controlled by the thermal gradient. Its operation is, thus, time dependent and related to the falling heat output of the waste and the consequent decreasing thermal gradient across the buffer and backfill.

Impact on the repository system:

The Soret effect is likely to have only a very small influence on the distribution of radionuclides within the porewaters in the buffer and the backfill. Its effect is likely to be dominated by other transport and retardation processes.

Bibliographic references:

Fargue D, Jamet P and Costesèque P (1998) Dispersion phenomena in thermal diffusion and modelling of thermogravitational experiments in porous media. Transport in Porous Media, 30, 323-344.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Soler JM (1999) Coupled transport phenomena in the Opalinus Clay: implications for radionuclide transport. Nagra Technical Report, NTB 99-09.

Equivalent FEPs:

Relates to FEPs 2.1.11 (Thermal processes and conditions in wastes and EBS) and 3.2.07 (Water mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.10 (Soret effect) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-45**

Name: **SWELLING OF THE TUNNEL BACKFILL**

Short description:

The backfill will be a mixture of partially dried bentonite and an inert filler material. The bentonite will take-up water during the resaturation phase and swell as the clay minerals adsorb water into their lattice structure. Swelling of the bentonite will affect properties which are important for water and gas transport through the backfill and for radionuclide transport and release.

Technical description:

The tunnel backfill in the spent fuel repository will be a mixture of partially dried bentonite and an inert filler material (such as quartz sand). Bentonite is dominantly composed of smectite clay. The smectites are noted for their ability to take-up water or organic liquids between their structural layers, and they all show marked cation-exchange properties. The swelling property means that, during the resaturation, the tunnel backfill will absorb water and swell, causing a significant decrease in the porosity and hydraulic conductivity of the material (see SFL-42 “Hydraulic resaturation of the buffer and backfill”).

The extent of bentonite swelling in the backfill will influence its final hydraulic properties and the swelling pressure. The aim is to obtain a backfill with a hydraulic resistance which is as least as high as the host rock. Any additional swelling into cracks and fractures intersecting the tunnels may result in higher hydraulic conductivity and lower swelling pressure in the vicinity of the cracks and fractures.

On the other hand, excessive swelling of the bentonite buffer in the deposition holes into the tunnels may decrease the hydraulic conductivity of the backfill just above the deposition holes. Uneven swelling can occur, but probably represents a transient state (resaturation phase). Deficiencies in material structure, cementing etc. could cause steady-state imperfections. Uneven swelling could cause preferential pathways or even flow instead of diffusion. The mineralogy and density of the bentonite/sand backfill and volume available for expansion will affect the extent of swelling once resaturation of the backfill is initiated.

The swelling pressure will also affect the stress field in the nearby rock. Swelling into fractures in the nearby rock will affect the apertures of these fractures and their hydraulic properties.

The inert filler material mixed with the bentonite in the backfill will provide mechanical stability and help restore the regional groundwater flow patterns after resaturation occurs. Various inert materials have been considered, including quartz sand grains and crushed rock. The nature and proportion of the filler material will be a primary control on the final mechanical and hydraulic properties of the backfill. Pusch (1995) has evaluated the consequences of using crushed rock instead of quartz particles in a repository backfill and concluded that the crushed rock backfill should perform as well as that containing quartz, albeit with slightly greater technical requirements for its emplacement. The hydraulic conductivity of the rock-clay backfill is anticipated to be less than 10^{-9} m/s, which is of the same order as low hydraulic conductivity rock (Pusch, 1995).

In addition to the normal backfill used in the tunnels, there will be additional use of highly compacted bentonite used to seals any zones of high hydraulic conductivity intersected by the repository tunnels. The use of bentonite as a sealing material will also be accompanied by the use of concrete bulk heads as deposition tunnels are filled and sealed. This is necessary for bentonite which, although able to produce a low hydraulic conductivity seal, has insufficient strength without the use of additional confinement. A similar combination of backfill and bulk heads is proposed for the final sealing of the repository. The degradation of these tunnel and shaft seals and the impact on the swelling of the backfill must be considered (see GEN-7 “Degradation of the borehole and shaft seals”).

Backfill swelling has a number of effects on the mechanical environment, as it provides support to the excavation, which changes as the backfill material resaturates and expands (Savage et al., 1998). In addition to the changes in the stress conditions due to the material placed in the excavation, the recovery of the groundwater pressure within the cavern will mean that the effective normal stress on discontinuities will change as the water takes up some of the load. The support provided by the backfill material and groundwater pressure will reduce the likelihood of a collapse of the roof as rock bolts and grout deteriorate with time.

Backfill swelling has a number of hydrogeological impacts. Once the repository has been sealed, a new transient state will exist as the local groundwater regime re-equilibrates with the regional groundwater flow. The possibility of the presence of large head differences within the excavation will need to be considered when defining the strategy for the backfilling and sealing. The material will resaturate differentially and a thermal gradient will exist across the deposition tunnels with distance from the wastes. The dominant groundwater flow into the deposition tunnels will be via fractures that will set up complex head gradients and flow patterns and it will be necessary to avoid the development of preferential flow paths due to high head gradients that could subsequently act as fast paths for the release of radionuclides (see GEN-35 “Fast transport pathways”).

Considerable amounts of cement and concrete will be used as grouting and structural materials in the repository. Cement pore fluids are hyperalkaline (pH 12.5 to 13.5) and highly reactive with carbonate, silicate, and aluminosilicate minerals. Migration of these pore fluids into the clay backfill material has the potential to exchange calcium ions from the cement for sodium ions on smectite mineral surfaces, potentially lowering the swelling capacity of the clay, and dissolve the framework of the backfill clay, precipitating a variety of calcium-silicate-hydrate minerals and zeolites, thus decreasing the porosity, plasticity, and swelling capacity of the clay (Savage, 1997).

However, because of the small amounts of cement and concrete envisaged in the spent fuel repository, alkaline alteration of montmorillonite can affect only a small part of the repository backfill. In addition, these interactions are likely to lead to positive solids volume changes. Although there may be detrimental effects to plasticity and clay swelling properties where this alteration is most pronounced, this may be counteracted by decreased porosity (see SFL-3 “Chemical alteration of the buffer and backfill”).

Origin in the repository system:

Swelling of the bentonite in the backfill results from post-closure resaturation of the bentonite-inert filler mixture by infiltrating groundwaters. The rate of resaturation and the final swelling pressures are controlled, in part, by the hydrogeological characteristics of the far-field and near-field rock. However, the proportion of void space in the tunnels to be filled by swelling is also a significant control on the final swelling pressure and on the final hydraulic and mechanical characteristics of the backfill.

Impact on the repository system:

Swelling of the bentonite in the backfill will result in gross changes to the hydraulic properties of the near-field, such that the hydraulic conductivity of the backfill should be similar or lower than the conductivity of the near-field rock. This will have significant impact on the transport of water, gas and released radionuclides. Swelling pressures will impact on the rock but the effect should be minimal.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Pusch R (1995) Consequences of using crushed crystalline rock as ballast in KBS-3 tunnels instead of rounded quartz particles. SKB Technical Report, TR 95-14.

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Savage D (1997) Review of the potential effects of alkaline plume migration from a cementitious repository for radioactive waste. Implications for performance assessment. UK Environment Agency Report P60.

Savage D, McLeod R and McEwen T (1998) An evaluation of repository-induced disturbances for a KBS-3 type repository. SKI Technical Report, 98:20.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics) and 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.1.1 (Swelling of bentonite into tunnels and cracks) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-46**

Name: **TEMPERATURE OF THE NEAR-FIELD**

Short description:

The temperature of the near-field is controlled by heat generated by radioactive decay in the spent fuel, superimposed on the natural geothermal conditions, as well as by the thermal properties of the engineered barriers and the rock, and the geometry of canister in the repository. The temperature will initially increase to a peak value and then decay away over a long period of time. Elevated temperatures in the near-field can affect the rock mass stability, rates of chemical processes, groundwater flow and, hence, radionuclide transport.

Technical description:

The ambient temperature of rock at repository depths (see GEN-38 “Temperature of the far-field“) will be controlled by the natural geothermal gradient (approximately 10 to 20°C /km for Sweden) and the mean surface temperature (approximately 5 to 10°C for Sweden). Thus the ambient temperature is partly controlled by repository depth and climatic conditions (King-Clayton et al., 1995).

During repository construction and operation, the rock around the repository openings will be cooled, largely by ventilation of the tunnels.

After repository closure, the rock around the repository openings will be heated by radiogenic heat from the spent fuel (see GEN-28 “Radioactive decay”). The radiogenic heat generation per canister is dependent on the fuel loading, its burn up history and the surface storage time before emplacement in the repository. These parameters will be controlled to ensure that the maximum temperature achieved on the outside wall of the canister will be approximately 90°C (Werme, 1998).

The maximum temperature achieved in the engineered barriers and the thermal distribution throughout the barriers and the near-field rock will be a function of the number of canisters emplaced in the repository, the geometry of the emplaced canisters, and the thermal properties (particularly thermal conductivity) of the various near-field materials (canister metal, bentonite buffer and the rock).

Temperatures in the near-field will initially increase to a peak value and then decay away over a long period of time, controlled by the slow reduction in the radiogenic heat output from the waste and the thermal conductivity of the near-field materials (Claesson and Probert, 1996). Heat is lost from the engineered barriers to the near-field rock almost exclusively by conduction because no groundwater flow (advection) is possible through the bentonite buffer. However, heat can be dissipated through the rock both by conduction and by advection of groundwater through the hydraulically-active fractures.

The rate of heat transfer through the bentonite buffer (and backfill in the tunnels) and the near-field rock will change as the near-field undergoes hydraulic resaturation (see GEN-32

“Hydraulic resaturation of the near-field rock” and SFL-42 “Hydraulic resaturation of the buffer and backfill”). Heat transfer will increase after resaturation because the water in the pore spaces will have a higher thermal conductivity than the air it replaces (see SFL-14 “Differential thermal expansion and contraction of the near-field barriers”). However, vapour flow can also contribute to heat transport before resaturation is complete (SKB, 1999).

If a separate gas phase develops around a failed canister due to anaerobic corrosion of the iron insert (see SFL-25 “Gas generation in the canister”), then the heat transfer to the bentonite buffer may be locally reduced.

The evolving temperature in the near-field can impact on the temperature and stress fields in the near and far-field rock masses, which may result in a change in their hydraulic conductivities. This might affect groundwater flow, as will the initiation of groundwater convection cells driven by the radiogenic heat output. The diffusion coefficient is also affected and thermal diffusion may occur (see SFL-44 “Soret effect in the buffer and backfill”).

The evolving temperature in the near-field will also affect the groundwater chemistry because elemental solubilities are temperature sensitive. A reduction in solubility may lead to precipitation of mineral phases in porespaces in the buffer, backfill or near-field rock which may, in turn cause cementation and changes in hydraulic conductivities.

The evolving temperature in the near-field might also influence corrosion of the canister and the spent fuel; mineralogical alteration of the buffer and backfill, and fracture surfaces exposed to groundwater; microbial activity; and diffusion rates, including matrix diffusion in the rock.

Origin in the repository system:

The temperature in the near-field is largely controlled by the radiogenic heat output from the spent fuel (dependent on burn up and storage times before emplacement), the repository geometry and the thermal conductivities of the near-field barrier materials.

Impact on the repository system:

Most chemical processes and many mechanical/hydraulic processes are affected by changes in the near-field temperature. Thus, the evolving temperature in the near-field will affect dissolution of the spent fuel; corrosion of the canister; mineralogical changes to the buffer, backfill and near-field rock; groundwater flow patterns; and, thus, radionuclide release rates to the far-field rock.

Bibliographic references:

Claesson J and Probert T (1996) Temperature field due to time-dependent heat sources in a large rectangular grid - derivation of analytical solution. SKB Technical Report, TR 96-12.

Come B (1995) A review of thermal, mechanical and hydrogeological properties of hard, fractured rocks (granite) for deep geological disposal of radioactive wastes. Proceedings of a workshop held in Brussels, 12-13 January 1995. EC Report, EUR 16219 EN.

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King-Clayton L, Chapman N, Kautsky F, Svensson N, de Marsily G and Ledoux E (1995) The central scenario for SITE-94: a climate change scenario. SKI Technical Report, 95:42.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.2.10 (Thermal processes and conditions of geosphere) and (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 1.1.2 (Radioactive decay: heat) in SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-47**

Name: **THERMAL DEGRADATION OF THE BUFFER AND BACKFILL**

Short description:

Elevated temperature, temperature gradients and changes in the temperature with time may cause physical and chemical degradation of the buffer and backfill materials. This may impact on the chemical, hydrological and mechanical properties of the buffer and backfill and, thus, on their ability to retard radionuclide migration.

Technical description:

In the spent fuel repository, the buffer is comprised of highly compacted bentonite while the backfill is similar except that it is comprised of bentonite mixed with an inert filler material (such as quartz sand) and is less compacted than the buffer material.

Thermal degradation of both the buffer and the backfill relates mainly to the chemical aspects of bentonite alteration and degradation that are initiated or accelerated by the heat produced from the waste, although some physical effects are also possible. All thermal impacts on the bentonite in the backfill can be expected to be less than those experienced by the bentonite in the buffer. This is mainly due to the fact that the thermal gradient extending from the waste canisters implies that the maximum temperatures in the backfill will be significantly lower than that in the buffer (see SFL-46 “Temperature of the near-field”).

Changes to the ambient temperature may cause precipitation and dissolution reactions to occur (see GEN-27 “Radionuclide precipitation and dissolution”). At elevated temperatures, congruent dissolution of smectite will occur at a rate determined by the temperature and the concentration of dissolved silica in the porewater (see SFL-3 “Chemical alteration of the buffer and backfill”). On exceeding its solubility, silica will precipitate either as cristobalite or amorphous silica.

Similarly, calcite may precipitate as a result of the dissolution of carbonates in the bentonite and the concentration of dissolved calcium in the porewater. The possibility of this process is highest during the high temperature period since the solubility of calcite decreases with increasing temperature.

Precipitation of silica or calcite may lead to cementation of the bentonite material, causing blockage of pores in the buffer and a reduction in swelling properties. The greater porosity in the backfill than the buffer will also mean that cementation potentially will have the greatest effect in the buffer.

It is possible that other mineral and mineraloids will precipitate depending on the abundance of chemical species and the T, Eh and pH conditions. Radionuclides released from the waste package could be incorporated in the precipitating phases by solid-solution or may be sorbed to the mineral surfaces.

In the initial high temperature resaturation stage, evaporation of the infiltrating water could lead to precipitation of naturally-occurring dissolved components. This is only likely to occur in the buffer (higher temperatures) and not the backfill. In addition, steam generation during the buffer resaturation period can induce a significant volume reduction of the clay. Experimental results show that this can occur even at relatively short times of contact with steam at 110°C (Couture, 1985; Oscarson and Dixon, 1989). The possibility of steam generation in a repository must be evaluated extremely carefully, since significant contraction of the bentonite backfill would severely impact upon repository performance.

A set of hydrothermal field tests on the resaturation of compacted clay has been conducted, in part to test this concept and impact of cementation. The tests were conducted for up to 4 years at a maximum 180°C and a gradient of 13°C /cm (Pusch et al., 1992). They found that there was considerable cementation of the clay, causing brittleness and loss of expansion within a few centimetres of the steel surface of the heat source. Clay samples from further away showed slight to negligible changes in properties. There was no evidence of increased iron content in the clay near the steel heater, indicating that iron compounds were not the cause of the cementation. In the hottest region, anhydrite and probably hexa-hydrate were precipitated, probably attributable to their retrograde solubility (i.e., these phases are less soluble at higher temperatures). Slightly further away from this hottest zone, new amorphous silica-aluminium phases formed in this slightly cooler region. The mobilisation and deposition of these phases may be attributable to their solubility in a thermal gradient or to cyclic evaporation/condensation that takes places in a resaturating clay under a thermal gradient. Gas was also found to have filled interior voids of the backfill, probably water vapour dominantly, although some hydrogen gas may have been evolved from corrosion of the steel heater.

Most attention to bentonite alteration has been paid to the conversion of smectite to illite (e.g. Pusch and Karnland, 1988). This transformation is potentially of significance to repository performance (especially of the buffer) because it is accompanied by changes in density, permeability and chemistry. The smectite to illite transition requires the addition of K^+, and to a lesser extent, Al^{3+}, either from interstitial water derived from diffusional transport from the surrounding host rock or from available exchange positions in the smectite. Inter-layer water is expelled during the transition and the phase becomes less ordered and less dense. Silica is released by exchange for aluminium (beidellitisation) and may be precipitated on the clay or in pores, leading to cementation and loss of plasticity. Bulk permeability, however, may be higher for the original compacted bentonite mass.

Natural analogue studies have examined this transformation in some detail (e.g. Pusch and Karnland, 1988). These analogue studies indicate the dependence of the smectite to illite transition on temperature, starting materials, access of water, and duration. They show that the bentonite is expected to remain largely unaltered for more than 10^6 years under repository conditions. Furthermore, if illitisation were to occur, the resulting backfill would still be extremely non-conductive and its effective diffusion coefficient may not change significantly (Pusch and Karnland, 1990). Indeed, studies show that the properties of natural clays, similar in composition to possible alteration assemblages of smectite, give acceptable performance with respect to physical properties could still be expected (Pusch and Hökmark, 1987).

The retardation properties of illite, however, may be significantly different than the starting smectite. The importance and effectiveness of sorption, however, in affecting radionuclide release to the far-field may be somewhat overemphasised except for cases involving relatively thick (> 1 metre) backfills or highly specific sorption of primary components of the waste matrix. Illitisation would result in only negligible change to the initial porosity of compacted smectite clay and should, therefore, have a negligible effect on long-term releases.

Origin in the repository system:

Thermal degradation of the buffer and backfill is controlled by the heat generated by the waste and by the thermal conductivity of the rock and the engineered barrier system materials. Since the radiogenic heat will decrease with time, thermal degradation processes are most important during the early post-closure stages. Dissolution, precipitation and mineralogical transformations are also dependent on the chemistry of the bentonite porewaters.

The maximum temperatures and the thermal gradient in the backfill will be less than in the buffer. Thus the extent of thermally induced bentonite should be less in the backfill than in the buffer.

Impact on the repository system:

Thermal degradation of the buffer and the backfill may affect the mineralogical, hydrological, mechanical and gas transport properties of the materials. Consequently, this process has potential significance for the retardation of radionuclides. However, most analyses suggest that the extent of thermal degradation is small.

Bibliographic references:

Couture RA (1985) Steam reduces the swelling capacity of bentonite. Nature, 318, 50-52.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Oscarson DW and Dixon DA (1989) The effect of steam on montmorillonite. Applied Clay Science, 4, 279-292.

Pusch R and Hökmark H (1987) Outline of models of water and gas flow through smectite clay buffers. SKB Technical Report, TR 87-10.

Pusch R and Karnland O (1988) Hydrothermal effects on montmorillonite: a preliminary study. SKB Technical Report, TR 88-15.

Pusch R and Karnland O (1990) Preliminary report on longevity of montmorillonite clay under repository related conditions. SKB Technical Report, TR 90-44.

Pusch R and Börgesson L (1992) Performance assessment of bentonite clay barrier in three repository concepts: VDH, KBS-3 and VLH. SKB Technical Report, TR 92-40.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.04 (Buffer/backfill materials and characteristics), 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.11 (Thermal processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEP 3.2.5 (Thermal effects on the buffer material) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Dave Savage and Bill Miller

Number: **SFL-48**

Name: **TOTAL RELEASE FROM THE SPENT FUEL ELEMENTS**

Short description:

The total release from the fuel elements involves the release of radionuclides from the UO_2 fuel matrix, from the gap between the fuel pellets and the Zircaloy cladding tubes and from cracks in the fuel pellets, and from the metal non-fuel components of the fuel assemblies. These various mechanisms are a primary control on the total radionuclide releases possible from the canister to the bentonite buffer.

Technical description:

Radionuclides are variously distributed throughout entire fuel assemblies and can be released by a number of different mechanisms. The majority of nuclides (fission products, actinides and their daughters) are contained in the UO_2 fuel matrix. These cannot be released until after the canister has failed and groundwater contacts the spent fuel. When this occurs, radionuclides can be released from the spent fuel as it begins to dissolve at a rate controlled by the solubility of the UO_2 matrix in the water adjacent to the fuel surface (see SFL-21 "Spent fuel dissolution and conversion"). In addition, radiolytically generated oxidants can cause oxidative conversion of the UO_2 spent fuel matrix, which might be associated with an increased dissolution rate.

Oxidative conversion to U_3O_7 or UO_3 or some other U(VI) compound, will alter the crystal structure of the fuel, causing the direct expulsion of some proportion of the radionuclides from the uranium oxide matrix. Thus, for these higher states, the radionuclide release rate would be controlled directly by the rate of fuel conversion rather than the rate of uranium dissolution (see SFL-21 "Spent fuel dissolution and conversion").

Some radionuclides preferentially accumulate at the fuel surface and in the pores, grain boundaries and in the gap between the fuel pellets and the Zircaloy cladding of the fuel rod. When the canister is perforated and groundwater contacts the fuel assembly, these radionuclides can be rapidly released as soon as the cladding tubes are perforated (see SFL-22 "Gap and grain boundary release").

Other radionuclides (activation products) occur within the metallic components of the fuel assemblies. These radionuclides will be released as soon as the canister is perforated and groundwater begins the corrode the stainless steel, Zircaloy and Inconel components (see SFL-41 "Radionuclide release from metal non-fuel parts"). The rate of release of these activation products will be controlled by the corrosion rate of the metals.

The total release of radionuclides from the fuel by all these mechanisms must be modelled in performance assessment in order to calculate correctly the source term (SKI, 1996).

Radionuclides released from the spent fuel matrix, gap and grain boundaries, and the metallic components of the fuel assemblies may go directly to solution, if they are soluble.

The most mobile phases will be the gaseous (e.g. Xe) and highly soluble (e.g. Cs and I) species in the gap and grain boundaries. Soluble species may be released from the canister by diffusion or by water turnover.

Radionuclides which do not go into solution can be directly incorporated into newly formed solid phases which form by the alteration of the spent fuel or the engineered barrier materials, or from direct precipitation from solution. Alternatively, radionuclides may be sorbed onto surfaces, such as the iron oxyhydroxides formed by corrosion of the iron insert of the canister (see SFL-28 “Radionuclide interaction with corrosion products”).

The total release of radionuclides from the fuel by all these mechanisms is controlled largely by the ingress of water to the canister (and thus by the canister lifetime). After canister failure, water access to the fuel assembly and water turnover (and thus radionuclide release) is controlled in part by the accumulation of a gas phase and iron corrosion products in the void spaces in the iron insert. Temperature and microbial activity could also have minor impacts on releases. Radioactive decay changes the radionuclide inventory available for release from the fuel elements (see GEN-28 “Radioactive decay”).

The release of radionuclides from the fuel is a primary control on the release from the canister to the bentonite buffer (see SFL-49 “Radionuclide release and transport from the canister”). Radionuclide release from the fuel controls the groundwater chemistry in the canister and the remaining radionuclide inventory.

Origin in the repository system:

Total release from the fuel is the combined releases due to dissolution and conversion of the spent fuel matrix, release from the gap and grain boundaries, and release of activation products due to corrosion of the metal components of the fuel assemblies.

No significant releases are possible until the canister is failed and, thus, the lifetime of the canister is a primary control on the total release from the fuel, after that, the composition of the groundwater inside the canister is the dominant control on the release rates and mechanisms.

Impact on the repository system:

Total release from the fuel controls the radionuclide content in the groundwater inside the canister and, thus, the radionuclide releases from the canister to the bentonite buffer. The remaining inventory in the waste is controlled by the total release from the fuel.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEP 3.2.01 (Dissolution, precipitation and crystallisation, contaminant) in the NEA International Database (NEA, 1998).

Relates to FEPs 1.2.9 (Dissolution chemistry) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-49**

Name: **RADIONUCLIDE RELEASE AND TRANSPORT FROM THE CANISTER**

Short description:

Radionuclides released from the spent fuel can migrate from inside the canister to the bentonite buffer, after the canister has been breached. Radionuclides in the form of solutes or colloids can migrate with the groundwater by diffusion or by water exchange between the canister and the buffer. Radionuclides can also migrate in the gas phase. Release and transport of radionuclides and other solute species from the canister will affect the groundwater chemistry (radionuclide content) inside the canister and in the buffer.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). No radionuclide releases from the canister can occur until after the copper shell and, subsequently, the iron insert are perforated (see SFL-7 “Corrosion of the copper shell of the canister” and SFL-9 “Corrosion of the cast iron insert”).

Once the canister is breached, groundwater can enter the central void spaces of the canister. At this stage, the spent fuel may be isolated from the groundwater by intact Zircaloy cladding tubes. However, these cladding tubes may have been breached by corrosion with water contained in the fuel pins before the canister failed (see SFL-10 “Canister corrosion prior to wetting”).

Progressive corrosion of the metallic components of the fuel assemblies will cause congruent release of activation products contained (or precipitated or sorbed onto the surfaces) in the stainless steel, Zircaloy, Inconel and Incoloy metal parts (see SFL-41 “Radionuclide release from metal non-fuel parts”).

Once the Zircaloy cladding tubes are perforated, groundwater can come into direct contact with the spent fuel. At this point, those mobile radioelements which accumulate in the gap between the cladding and the fuel pellets, and in the grain boundaries within the fuel pellets may be rapidly released from the fuel (see SFL-22 “Gap and grain boundary release”).

Continued groundwater contact with the spent fuel will initiate dissolution and oxidative conversion of the UO_2 spent fuel, causing radionuclide releases from the spent fuel matrix (see SFL-21 “Spent fuel dissolution and conversion”).

These various release processes will liberate radionuclides from their respective materials and locations within the canister. The subsequent behaviour of these radionuclides depends largely on their chemical behaviour in the changing chemical conditions inside the canister. Certain chemical parameters (e.g. salinity and pH) will be largely dependent on groundwater interactions with the near-field rock and the bentonite buffer but other parameters (e.g. Eh, temperature) will be controlled by processes occurring in the canister. These latter processes are related to the time dependent thermal and radiation fields generated by the spent fuel which control temperature and radiolytic processes (see SFL-52 “Evolving water chemistry in the canister”).

Some proportion of the liberated radionuclides will go into solution, depending on their solubilities in the changing chemical conditions inside the canister, and so will alter the radionuclide content of the groundwater in the canister. However, most liberated radionuclides will be poorly soluble and are expected to be incorporated directly in newly formed secondary alteration mineral phases or co-precipitates, or sorbed onto the canister corrosion products (see SFL-28 “Radionuclide interaction with corrosion products”). Some of the latter radionuclides may become associated with colloids, either by attaching themselves directly to existing colloids or when solid materials physically degrade (see SFL-6 “Colloids and particles in the canister”). Lastly, some radionuclides may become associated with a gas phase, either as part of the gas molecules themselves (e.g. $^{14}CO_2$) or by association with the water-gas (bubble) interface (see SFL-25 “Gas generation in the canister”).

The potentially mobile radionuclides in the groundwater (solutes and colloids) may be transported through the canister largely by diffusion, if the water inside the canister void spaces is not turning over (see SFL-15 “Diffusion in and through the canister”), or by advection (see SFL-57 “Water turnover in the iron insert of the canister”). Along the transport path, retardation processes may reduce the amount of mobile radionuclide as a consequence of precipitation, sorption or filtration (of colloids). The transport of these species is thus controlled by release rates from the spent fuel, water turnover (outflow) and diffusion rates, retardation processes, colloid generation and filtration, and radioactive decay.

The potentially mobile radionuclides in the gas phase may be transported through the canister by gas flow, depending on the balance of gas pressures inside the canister and hydrostatic pressures outside the canister, and the geometry of perforations in the canister walls (see SFL-23 “Gas escape from the canister”).

The transport of radionuclides through and out of the canister may be partially controlled by the thermal and electrochemical gradients, if water turnover (outflow) rates are small.

The transport pathways for solutes, colloids and gas phase radionuclides will be controlled in the canister by the original geometry of the canister and the spent fuel, perforations in the canister and the Zircaloy cladding, and the growth of any iron corrosion products.

The transport of radionuclides and other solute species out of the canister will change the remaining groundwater composition inside the canister and the groundwater composition in the bentonite buffer. The removal of dissolved uranium and other radionuclides from the canister waters can enhance further dissolution of the spent fuel because the release of these species from the spent fuel matrix is solubility limited. Transport from the canister will also affect radionuclide distribution in the buffer (see SFL-50 “Radionuclide release and transport from the buffer and backfill”).

Origin in the repository system:

Radionuclide releases from the canister are controlled largely by the lifetime of the canister, since no releases are possible before the canister fails.

After canister failure, radionuclide releases from the canister are controlled by the rate of radionuclide release from the spent fuel and activation products from the metallic fuel components; sorption on corrosion products; the groundwater chemistry inside the canister; radioactive decay; gas pressures; and the water outflow and diffusion rates.

Impact on the repository system:

Radionuclide releases from the canister will control the groundwater chemistry (radionuclide content) inside the canister and in the bentonite buffer. The removal of radionuclides from groundwaters adjacent to the spent fuel will initiate further releases of uranium and other solubility controlled species from the spent fuel matrix.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 3.2 (Contaminant release and migration factors), 3.2.01 (Dissolution, precipitation and crystallisation of contaminant), 3.2.03 (Sorption/desorption processes of contaminant), 3.2.04 (Colloids and contaminant interactions and transport), 3.2.07 (Water-mediated transport of contaminants) and 3.2.09 (Gas-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEP 1.5 (Release of radionuclides from the failed canister) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-50**

Name: **RADIONUCLIDE RELEASE AND TRANSPORT FROM THE BUFFER AND BACKFILL**

Short description:

Radionuclides released from the canister can be transported through the buffer and backfill in solution or as a gas, and possibly also in colloidal form. The mechanisms and rate of radionuclide transport through the buffer and backfill are determined largely by the physical properties of the buffer and backfill materials, mostly the swelling pressure. Transport through the buffer and backfill controls the release rate to the near-field rock.

Technical description:

No radionuclides can be transported through the buffer or backfill until after the copper shell and, subsequently, the iron insert of the canister have failed (see SFL-7 “Corrosion of the copper shell of the canister” and SFL-9 “Corrosion of the cast iron insert”). In the absence of manufacturing defects, the canister will not fail during the early hydraulic resaturation event and, thus, when radionuclides are released from the canister they will come into contact with the highly compressed, water saturated bentonite buffer (see SFL-1 “Swelling of the bentonite buffer”).

Radionuclide transport through the buffer and backfill is initiated by radionuclide releases from the failed canister (see SFL-49 “Radionuclide release and transport from the canister”). Potential transport mechanisms through the resaturated buffer and backfill include diffusion of dissolved species in the groundwater, advection of dissolved species and colloids in groundwater flow, and gas flow. In the normal evolution scenario, diffusion and gas flow will be the dominant mechanisms.

Under normal water saturated conditions, the bentonite buffer will have a high swelling pressure and a very low hydraulic conductivity (10^{-13} to 10^{-14} m/s), such that groundwater flow is effectively zero. Radionuclides in solution can, thus, only be transported through the bentonite buffer by the very slow rates of molecular diffusion (see SFL-20 “Groundwater flow through the buffer and backfill”). Diffusion rates through the buffer and backfill will be influenced by the concentration gradient of released contaminants (see GEN-9 “Diffusion”) and by the temperature gradient due to radiogenic heat output from the spent fuel (see SFL-44 “Soret effect in the buffer and backfill”).

Colloids generated at the canister or in the buffer will also be effectively immobilised (filtered) by the very fine pore spaces in the bentonite buffer and any radionuclides associated with the colloids will be retarded (see SFL-5 “Colloid behaviour in the buffer and backfill”).

Radionuclides associated with a gas (e.g. tritium) may initially dissolve into the groundwater and be transported through the buffer by molecular diffusion. However, the gas pressure is likely to exceed the confining pressure inside the buffer and a separate gas phase (bubble) may then form. This gas will remain immobile until the gas pressure exceeds a critical level (opening pressure), allowing a gas flow pathway to be opened through the bentonite (see SFL-24 “Gas flow through the buffer and backfill”). Transport of gas through the buffer and backfill is thus dependent on the rate of gas generation at the canister.

Under normal conditions, the tunnel backfill will have a swelling pressure which is lower than that in the bentonite buffer and a hydraulic conductivity which is higher (10^{-9} to 10^{-10} m/s). Under these conditions, groundwater flow may occur through the backfill but at a very slow rate, thus colloids will also remain largely immobile. Gas flow through the backfill will be possible at lower opening pressures compared to the buffer, because of the lower bentonite swelling pressure in the backfill.

The mechanism for transport through the buffer and backfill (diffusion or flow) and the rate of transport is dependent on the physical characteristics of the materials, in particular, the swelling pressure. Over time, the transport characteristics of the buffer and backfill materials may change due to physical and chemical degradation processes, such as mineralogical alteration (see SFL-3 “Chemical alteration of the buffer and backfill”) or dilution (see SFL-16 “Dilution of the buffer and backfill”). The effect of these processes may be to reduce the swelling pressures (thus increasing the hydraulic conductivity) or to open permanent preferential pathways through which groundwater, gas or colloids may flow.

Radionuclide transport through the buffer and backfill is also controlled by the release rate from the failed canister, radioactive decay and the retardation processes occurring in the buffer and backfill, such as sorption and precipitation which will limit releases to the near-field rock.

Radionuclide transport through the buffer and backfill will change the groundwater chemistry within the canister (reduce the inventory of dissolved species), which will initiate further dissolution from the spent fuel of solubility controlled species. Transport through the buffer will also affect distribution and release to the far-field (see GEN-11 “Distribution and release of radionuclides from the far-field”).

Origin in the repository system:

Radionuclide transport through the buffer and backfill is initiated by radionuclide releases from the failed canister. After canister failure, the transport mechanisms (flow or diffusion) through the buffer and backfill are controlled largely by the swelling pressure of the bentonite, which creates a very low hydraulic conductivity and an efficient colloid filter.

The rate of transport is dependent on the release rate from the canister, the evolving properties of the buffer and backfill materials, gas generation rate at the canister, retardation processes in the buffer and backfill materials, and the temperature and chemical gradients.

Impact on the repository system:

Radionuclide transport through the buffer and backfill controls the groundwater chemistry in the engineered barriers, the rate of release of solubility controlled species, and the release rate to the near-field rock.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Equivalent FEPs:

Relates to FEPs 3.2 (Contaminant release and migration factors), 3.2.01 (Dissolution, precipitation and crystallisation of contaminant), 3.2.03 (Sorption/desorption processes of contaminant), 3.2.04 (Colloids and contaminant interactions and transport), 3.2.07 (Water-mediated transport of contaminants) and 3.2.09 (Gas-mediated transport of contaminants) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.6 (Diffusion - surface diffusion) and 3.2.9 (Flow through buffer/backfill) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-51**

Name: **EXPANSION OF SOLID CORROSION PRODUCTS**

Short description:

The solid alteration products generated by the corrosion of the metal canister have lower densities than the original metal. As a consequence, metal corrosion is accompanied by volume expansion. On the outside of the canister, expanding corrosion products will compress the bentonite buffer and increase the stresses on the canister. On the inside of the canister, expanding corrosion products may restrict further water contact with the metal and spent fuel surfaces.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

Corrosion of the copper shell will proceed by processes dependent on the chemistry of the groundwater adjacent to the canister (see SFL-7 “Corrosion of the outer copper shell of the canister”). In the near-field, it is probable that corrosion of the canister will take place in a chemically reducing environment with dissolved (reduced) sulphur species present in the groundwater. In this system, copper corrosion is dominated by sulphide attack and solid copper sulphide phases will form on the canister surface.

However, in the presence of groundwaters with different chemistry, other copper corrosion products are possible. For example, in the unlikely event that oxidising waters could penetrate the near-field (e.g. associated with glaciation), then solid copper oxide phases will form on the canister surface.

Regardless of the chemical form of the copper corrosion products which will form, they will all have a higher molar volume than the original copper metal, even in their most dense state. Thus, the continued corrosion of the copper shell will generate increasing volumes of copper corrosion products at the interface between the canister and the buffer. This in turn will lead to increased mechanical stresses on both the canister (see SFL-30 “Mechanical impact on the canister”) and the buffer (see SFL-31 “Mechanical impact on the buffer and backfill”).

An increase in the mechanical stresses acting on the canister may accelerate continued degradation or cause mechanical damage to the canister, especially after it has been weakened by substantial corrosion (see SFL-39 “Reduced mechanical strength of the canister”). At the extreme, the canister lifetime could be reduced by this process, although this is considered extremely unlikely because of the very slow copper corrosion rate in reducing conditions (Werme, 1998).

An increase in the mechanical stresses acting on the buffer will cause it to compress slightly. This could change the bentonite swelling pressure in the vicinity of the canister. In extreme cases, mechanical impacts to the buffer could result in local changes to the hydraulic conductivity of the buffer, which might increase the potential for groundwater or gas flow to occur through the buffer (see SFL-20 “Groundwater flow through the buffer and backfill” and SFL-24 “Gas flow through the buffer and backfill”). This could also cause movement of the canister within the disposal hole (see SFL-33 “Movement of the canister in the buffer”). However, this is also considered extremely unlikely because of the very slow copper corrosion rate in reducing conditions (Werme, 1998).

After the copper shell has been perforated, anaerobic corrosion of the iron insert will be initiated and will generate hydrogen and solid iron oxide (see SFL-9 “Corrosion of the cast iron insert”). The most probable solid phase will be magnetite (Fe_3O_4) at repository temperatures although, if additional dissolved species are present in the groundwater, other solid alteration products may be formed, e.g. pyrite, siderite or Fe-aluminosilicates.

Over time, the iron oxides may subsequently react with the near-field porewaters to form a number of possible Fe(III) solid species. Most likely, the first solid would be an amorphous ferric-oxyhydroxide ($Fe(OH)_3$) and, over long periods of time, this may transform to more stable solids such as goethite (FeOOH) and haematite (Fe_2O_3).

As with the copper corrosion products, all solid iron corrosion products (regardless of chemical form) will have a higher molar volume than the original iron metal, even in their most dense state. Thus, the continued corrosion of the iron insert will generate increasing volumes of iron oxides inside the canister. Initially, these iron oxides will expand into the void spaces in the canister (around 1 m^3). This may restrict continued access of water to the metal and spent fuel surfaces within the canister, limiting further corrosion, and also may decrease the transport and release of radionuclides from the canister.

Corrosion of the outside walls of the iron insert in the void between it and the copper shell will lead to a growth of iron oxides in the void. This will lead, in turn, to an increase in the mechanical stresses acting on the inside of the copper shell (e.g. Hoch and Sharland, 1993). This could result in a widening of cracks and perforations in the copper shell. Modelling results suggest that it would take at least 100 000 years before extensive damage to the canister could result from this process (Bond et al., 1997).

If iron corrosion continues and all internal void spaces are filled, further expansion of the iron oxides may result in their extrusion out of the canister through perforations in the copper shell leading to increased mechanical stresses on the canister and the buffer. However, the extent to which this process might occur is uncertain. Some modelling approaches assume iron corrosion ceases when the internal void spaces are filled (e.g. Blackwood et al., 1994).

Continued generation of hydrogen will lead to an increase in the gas pressure which may result in gas flow through the buffer (see SFL-24 “Gas flow through the buffer and backfill”).

Origin in the repository system:

An expansion in the volume of metal corrosion products is a direct and unavoidable consequence of the corrosion of the copper shell and, subsequently, the iron insert of the canister. The rate of expansion and the final expanded volume are dependent on the metal corrosion rates which are, themselves, dependent on the diffusional transport of corrodants and corrosion products through the buffer, the ambient groundwater chemistry and the temperature.

Impact on the repository system:

An expansion in the volume of metal corrosion products on the outside of the canister will impact on the mechanical stresses acting on the canister and the bentonite buffer. In extreme cases, this could potentially lead to earlier failure of the canister and a localised decrease in the hydraulic conductivity of the buffer.

An expansion in the volume of metal corrosion products on the inside of the canister will lead to a reduction in the volume of the void spaces, a restriction in the access of water to metal and spent fuel surfaces and, thus, a reduction in the rates of further corrosion and radionuclide releases. The expanding corrosion products in the gap will create tensile stresses on the outer copper shell, which may lead to its accelerated failure.

Bibliographic references:

Blackwood DJ, Hoch AR, Naish CC, Rance A and Sharland SM (1994) Research on corrosion aspects of the Advanced Cold Process Canister. SKB Technical Report, TR 94-12.

Bond A, Hoch A, Jones G, Tomczyk A, Wiggin R and Worraker W (1997) Assessment of a spent fuel disposal canister: assessment studies for a copper canister with cast steel inner component. SKB Technical Report, TR 97-19.

Hoch AR and Sharland SM (1993) Assessment study of the stresses induced by corrosion in the Advanced Cold Process Canister. SKB Technical Report, TR 94-13.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics), 2.1.07 (Mechanical processes and conditions in wastes and EBS) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Relates to FEPs 3.2.7 (Swelling of corrosion products) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

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Number: **SFL-52**

Name: **EVOLVING WATER CHEMISTRY IN THE CANISTER**

Short description:

Failure of the copper shell and subsequently the iron insert components of the canister will lead to intrusion and turnover of groundwater in the canister void spaces. This water will react with the various metals and the spent fuel causing considerable changes to the chemistry of the intruding water. When radionuclides are released from the spent fuel, this will result in further changes to the water chemistry. When this water migrates out of the canister it will mix with the porewaters in the buffer.

Technical description:

Groundwaters in the bentonite buffer will corrode the copper shell of the canister (see SFL-7 "Corrosion of the copper shell"), although this is a very slow process and the copper shell is expected to remain intact for at least 100 000 years (SKB, 1999). After perforation of the copper shell, the groundwater will penetrate the gap between the shell and the insert and initiate anaerobic corrosion of the iron (see SFL-56 "Water turnover in the copper shell"). Eventually, progressive corrosion of the iron will perforate the insert and the groundwaters will be able to infiltrate into the central void spaces in the canister (see SFL-57 "Water turnover in the cast iron insert").

The water which infiltrates the centre of the canister will have considerably altered chemistry compared to the groundwaters in the near-field rock because it will have evolved over time by interaction with the major and minor mineral components of the bentonite buffer (montmorillonite clay, pyrite, quartz, feldspar, calcite, sulphates etc.) and by corrosion of the copper and iron metals of the canister. The groundwater at this point will be chemically-reducing and mildly alkaline. The water may also contain variable concentrations of other species such as organic complexes, nitrates etc. which may be derived from the natural groundwater recharge and evolution in the far-field rock or from minor components in the near-field materials.

Once inside the canister, this modified groundwater will be in contact with the walls of the iron insert, the fuel assemblies and, eventually, the spent fuel after the Zircaloy cladding surrounding it is corroded. Reactions with these materials will further modify the water chemistry by a series of different processes.

Corrosion of the metallic (stainless steel, Zircaloy and Inconel) components of the fuel assemblies will release neutron activation products, such as ^{14}C, ^{93}Zr and ^{60}Co (see SFL-41 "Radionuclide release from metal non-fuel parts"). Some proportion of these activation nuclides will go into solution, depending on their solubilities in the ambient groundwater chemistry.

Continued corrosion will eventually perforate the Zircaloy cladding surrounding the spent fuel pellets. At this time, species segregated into the gap between the fuel pellets and the cladding, and in grain boundaries in the fuel, such as ^{129}I and ^{135}Cs, will be released (see SFL-22 “Gap and grain boundary release”). These nuclides are readily soluble and so will further alter the radionuclide content of the groundwater in the canister.

Once the Zircaloy cladding is perforated and water can contact the spent fuel, radiation (particularly alpha radiation) emitted from the spent fuel matrix will cause radiolysis of the water immediately adjacent to the fuel surface (see GEN-30 “Radiolysis”). Some radiolysis would have been possible prior to this but the rate of radiolysis will be greatly increased by direct contact between the spent fuel and the water. Radiolytic decomposition of water leads to the formation of equal amounts of oxidants and reductants, which quickly convert to more stable or inert species. This leads to the formation of H_2 (gas) and oxidants such H_2O_2 and O_2. The hydrogen gas is relatively unreactive under near-field temperatures (<100°C) and can migrate from the system. The net effect of the radiolysis of groundwater is, thus, to make the groundwater in contact with the fuel surface oxidising.

This oxidising water will begin to dissolve the spent fuel UO_2 matrix, releasing uranium, actinides and fission products. The oxidising conditions close to the spent fuel matrix will also cause oxidative conversion of the spent fuel to U_4O_9 or U_3O_7 and, possibly, to the higher oxidation states of U_3O_7, UO_3 or some other U(VI) compound. If the higher oxidation states are reached, lattice conversion will occur causing the direct expulsion of radionuclides from the uranium oxide matrix (see SFL-21 “Spent fuel dissolution and conversion”). Some proportion of the nuclides released from the spent fuel will go into solution, depending on their solubilities in the ambient groundwater chemistry and so will further alter the radionuclide content of the groundwater in the canister. However, most of these species are poorly soluble and will be incorporated into newly formed separate uranium-bearing mineral phases (e.g. uranyl silicates) or co-precipitates, or sorbed onto canister corrosion products.

At the same time, the water inside the canister will also cause corrosion of the walls of the iron insert and the stainless steel components of the fuel assembly, generating hydrogen gas and solid iron-oxyhydroxide alteration products. As iron corrosion proceeds, the partial pressure of the hydrogen may eventually exceed the local hydrostatic pressure and a separate gas phase can evolve (see SFL-25 “Gas generation in the canister”). Depending on the geometry of the perforations in the canister, this gas phase may be trapped inside the canister void spaces and, as it grows in volume, may expel groundwater. If this were to occur, the gas bubble would limit further aqueous corrosion of the canister, the metallic components of the fuel assemblies and the spent fuel. Equilibrium between the hydrogen gas and the groundwater will be one factor in defining the overall redox conditions for the groundwater inside the canister.

The formation of solid iron-oxyhydroxide alteration products is associated with a volume increase and this expansion process will reduce the void space in the canister and may also restrict groundwater access to the spent fuel and metal surfaces, further limiting aqueous corrosion (see SFL-51 “Expansion of solid corrosion products”). The iron-oxyhydroxide has a large sorption capacity and a significant proportion of the less soluble radionuclides released from the spent fuel are likely to sorb directly onto its surface or to be incorporated directly into co-precipitate phases.

Overall, the bulk chemical composition of the groundwater in the canister will be controlled by these processes. Redox conditions will be defined by the competitive reactions of radiolytic oxidant production and oxidation of the iron insert and the spent fuel. The balance will depend on the lifetime of the canister. Since the copper shell is expected to last for at least 100 000 years, the radiation dose to the water after canister failure, to drive radiolysis, will be lessened. This, combined with the very large mass of iron probably means that conditions will largely remain chemically reducing, except perhaps very locally to the spent fuel surface. The pH will largely be controlled by reaction with minerals in the buffer (calcite, feldspar etc.) because there is very little pH buffering material within the canister, and so will be mildly alkaline.

The radionuclide content of the water will depend in the first instance on the rate of radionuclide release from the spent fuel and then on the pH and Eh balance in the canister water since these parameters are major controls on radionuclide solubilities. With progressive corrosion of the spent fuel and the iron, the radionuclide distribution between solid and liquid phases may change. The radionuclide content in the groundwater will also change as a consequence of radioactive decay and ingrowth.

Lastly, temperature changes in the canister have a small impact on the groundwater chemistry because the reaction rates and solubility limits are partly dependent on temperature. However, the copper shell will remain intact during the early thermal peak so that, by the time water can enter the canister, the radiogenic heat production will have diminished such that temperatures in the near-field will be controlled largely by the local geothermal gradient.

It is probable that some other processes may have minor impacts on the groundwater chemistry in the canister, such as microbial activity (see SFL-32 “Microbial activity”) or electrochemical gradients (see GEN-13 “Electrochemical effects”) but, their significance will be minor.

Changes in the groundwater chemistry in the canister will impact on the evolving groundwater chemistry outside the canister in the buffer, backfill and near-field rock as water turnover results in the chemically-altered water leaving the canister and migrating back into the other near-field barriers.

Origin in the repository system:

The groundwater chemistry in the canister is controlled by a complex interaction between a number of other time-dependent processes and mechanisms. The starting point is the natural groundwater composition in the near-field rock. This is then sequentially altered by reaction with major and mineralogical components in the buffer and backfill, and then the copper shell and iron insert of the canister. These reactions mean that the fluid which initially reaches the spent fuel will be chemically reducing and mildly alkaline.

Depending on the time of canister failure (controlling the radiation dose and radionuclide content of the waste), the water in the canister may be made oxidising due to radiolytic decomposition and will dissolve from the waste a radionuclide content largely dependent on the Eh, pH, and time, and to a lesser extent, temperature.

Impact on the repository system:

The evolving water chemistry in the canister will affect the continued degradation of the spent fuel and, thus the rate of radionuclide release and radionuclide content in the water. As this evolving water migrates out of the canister into the other near-field barriers, it will mix with the existing porewaters changing their chemistries. As a consequence, the evolving water chemistry in the canister will affect the degradation of the near-field barriers and the transport of radionuclides through the far-field barriers.

Bibliographic references:

Arthur R and Apted M (1996) Radionuclide solubilities for SITE-94. SKI Technical Report, 96-30.

Arthur R and Apted M (1996) Modelling of the near-field chemistry for SITE-94. SKI Technical Report, 96-31.

Grenthe I, Fuger J, Lemire RJ, Muller AB, Nguyen-Trung C and Wanner H (1992) Chemical Thermodynamics of Uranium. Chemical Thermodynamics Series Volume 1, North-Holland, New York.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Equivalent FEPs:

Relates to FEPs 2.1.03 (Container materials and characteristics) and 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-53**

Name: **EVOLVING WATER CHEMISTRY IN THE BUFFER**

Short description:

The chemistry of the porewater in the bentonite buffer is defined in terms of the major and trace element concentrations, including repository-derived radionuclides, and the major variables (T, Eh and pH) and any chemical gradients in the bentonite. It is largely controlled by groundwater-bentonite chemical interactions.

Technical description:

The buffer around the waste canisters in the spent fuel repository will comprise highly compacted bentonite. It is emplaced in a partially dried state (MX-80 bentonite contains 5 to 15 wt.% moisture and hydrate water). During resaturation, groundwater with ambient far-field composition (see GEN-22 “Far-field groundwater chemistry”) will infiltrate into the bentonite buffer and other parts of the engineered barrier system, and will approach equilibrium first with the bentonite and then with the canister and its corrosion products, and later with the exposed fuel (Arthur and Apted, 1996).

For a short period after resaturation (see SFL-42 “Hydraulic resaturation of the buffer and backfill”), the chemistry of the buffer porewaters may be oxidising due to the dissolution of the free air in the near-field void spaces. Oxidative dissolution of sulphide minerals in the bentonite and the rock may then cause the initial porewaters to become acid. However, conditions will rapidly become chemically reducing and mildly alkaline due to buffering by the fresh rock and the bentonite.

Over longer time periods, the chemistry of the buffer porewater is largely a function of the composition of the saturating ambient host rock groundwater, temperature, interactions with the major and minor mineralogical components of the bentonite, interactions with the waste canister and, after canister failure, interactions with the wasteform.

Most predictive models of bentonite porewater evolution take account of ion-exchange reactions between the bentonite (smectite) and the infiltrating groundwater. The dominant ion-exchange reaction is likely to be between sodium in the bentonite and calcium in the groundwater, although other exchanges are possible depending on the composition of the groundwater. This will cause chemical alteration of the buffer and backfill (see SFL-3 “Chemical alteration of the buffer and backfill”). Modelling results (Wanner, 1986; Wieland et al., 1994) indicate that the effect of ion-exchange reactions is not generic and thus site-specific groundwater compositions must be used in the calculations.

In the buffer, ion-exchange reactions will be limited by the inflow of groundwater. When groundwater flow is restricted, modelling results (Liu and Neretnieks, 1997) indicate that no sharp reaction fronts will form, with ion-exchange reactions taking place uniformly throughout the buffer.

Ion exchange reaction is not the only model proposed for predicting long-term bentonite pore water chemistry. Bentonite (smectite) will be involved in slow dissolution-precipitation processes so that alternative models reflecting these interactions have been promulgated (Arthur, 1994).

Bentonite porewater chemistry will also be influenced by reaction between the infiltrating groundwater and the other mineralogical constituents of the buffer (which is not pure bentonite) notably carbonates and pyrite, but also any organic compounds present. Dissolution of trace amounts of pyrite within the bentonite will help maintain reducing conditions in the near-field, but may generate acidity, according to ambient geochemical conditions. Carbonates, such as calcite or dolomite/ankerite, can be important in reactions buffering pH (Sasaki et al., 1995) because of their rate of reaction. A consideration in this regard is the ambient P_{CO2} of the host rock groundwater which may govern the calculated pH of a bentonite porewater, if carbonate dissolution is assumed to be important.

The chemistry of the bentonite porewaters can further be effected by processes occurring at the canister, such as interactions with the canister, the wasteform and their corrosion products; radiolytic decomposition of the porewater closest to the canister leading to formation of oxygen, hydrogen peroxide and hydrogen; and gas generation and transport. The porewater chemistry will also be impacted by the release transport of radionuclides from the waste package into the buffer (see SFL-49 “Radionuclide release and transport from the canister”). Depending on the relative rates of these reactions and of diffusion through the bentonite, and any thermal gradient (Soret effect), chemical gradients (oxidants/reductants and canister alteration products) could be established in the buffer that might affect radionuclide migration.

The bentonite porewater chemistry (and changes over time) is an important control over the release of radionuclides from the near to the far-field because many radionuclides are poorly soluble in equilibrium bentonite porewater chemistries. Performance assessments often evaluate radionuclide solubilities on the basis of these bentonite pore-water chemistries because they are considered to be better defined and more time-invariant than are waters in contact with other engineered barrier system components (McKinley and Savage, 1996).

Origin in the repository system:

The water chemistry in the bentonite buffer is influenced by many different processes, including the resaturation of the buffer after repository closure; the composition of the water in the near-field rock, the tunnel backfill and in the canister; canister and wasteform corrosion; chemical alteration of the bentonite buffer; microbial activity; radiolytic decomposition of water in the buffer; temperature in the buffer; groundwater flow in the buffer; diffusion of species in the buffer; generation and transport; thermal diffusion of dissolved species and Soret effects.

Impact on the repository system:

The evolving chemistry of the bentonite pore waters is an important control on the behaviour of the repository because it impacts most importantly upon the long-term stability of the engineered barrier system components and on the near-field solubilities of released radionuclides. It also affects other processes, including microbial activity in the buffer; radiolytic decomposition of water in the buffer; thermal degradation of the backfill; coagulation of bentonite particles in the backfill; sorption and desorption, precipitation and dissolution of radionuclides in the backfill; colloid generation and stability in the backfill; anion exclusion; the water chemistry in the canister, the tunnel backfill and in the near-field rock.

Bibliographic references:

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McKinley IG and Savage D (1996) Comparison of solubility databases used for HLW performance assessment. Journal of Contaminant Hydrology, 21, 331-350.

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Sasaki Y, Shibata M, Yui M and Ishikawa H (1995) Experimental studies on the interaction of groundwater with bentonite. Scientific Basis for Nuclear Waste Management, XVIII, 337-344.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Wanner H (1986) Modelling interaction of deep groundwaters with bentonite and radionuclide speciation. Nagra Technical Report, NTB 86-21.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.04 (Buffer/backfill materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 4.1.8 (Change of groundwater chemistry in nearby rock) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Dave Savage and Bill Miller

Number: **SFL-54**

Name: **EVOLVING WATER CHEMISTRY IN THE BACKFILL**

Short description:

The chemistry of the porewater in the bentonite-rich backfill is defined in terms of the major and trace element concentrations, including repository-derived radionuclides, and the major variables (T, Eh and pH) and any chemical gradients in the bentonite. It is largely controlled by groundwater-bentonite chemical interactions.

Technical description:

The bentonite-rich backfill used to fill tunnels in the spent fuel repository is similar to the bentonite buffer around the canisters except that the backfill is comprised of bentonite mixed with an inert filler material (such as quartz sand) and is less compacted than the buffer material. Some designs call for a lower quality bentonite than used in the buffer. Like the porewater in the buffer, the chemistry of the porewater in the backfill is largely controlled by post-closure groundwater-bentonite chemical interactions.

The bentonite-rich backfill will be emplaced in the partially-dry state. During resaturation (see SFL-42 "Hydraulic resaturation of the buffer and backfill"), groundwater with ambient far-field composition will infiltrate into the backfill and other parts of the engineered barrier system, and will approach equilibrium first with the bentonite and then with the canister and its corrosion products, and later with the exposed fuel (Arthur and Apted, 1996).

For a short period after resaturation, the chemistry of the backfill porewaters may be oxidising due to the dissolution of the free air in the near-field void spaces. However, conditions will rapidly become chemically reducing due to buffering by the fresh rock and the bentonite.

Over longer time periods, the chemistry of the backfill porewater is largely a function of the composition of the saturating ambient host rock groundwater, temperature, interactions with the major and minor mineralogical components of the bentonite.

Most predictive models of bentonite porewater evolution take account of ion-exchange reactions between the bentonite (smectite) and the infiltrating groundwater. The dominant ion-exchange reaction is likely to be between sodium in the bentonite and calcium in the groundwater, although other exchanges are possible depending on the composition of the groundwater. This will cause chemical alteration of the backfill. Modelling results (Wanner, 1986; Wieland et al., 1994) indicate that the effect of ion-exchange reactions is not generic and thus site-specific groundwater compositions must be used in the calculations.

In the repository, ion-exchange reactions will be limited by the inflow of groundwater to the backfill. The groundwater flow in the backfill is likely to be faster than in the buffer because of the engineered damaged zone surrounding the tunnels and the higher hydraulic conductivity of the bentonite-sand backfill mix than the compacted bentonite buffer material.

Ion exchange reaction is not the only model proposed for predicting long-term bentonite pore water chemistry. Bentonite (smectite) will be involved in slow dissolution-precipitation processes so that alternative models reflecting these interactions have been promulgated (Arthur, 1994). There is considerable debate in the geological literature concerning the thermodynamic stability of smectite clays and some authors view these materials as 'metastable micas'.

Bentonite porewater chemistry will also be influenced by reaction between the infiltrating groundwater and the other mineralogical constituents of the backfill, notably carbonates and pyrite, but also any organic compounds present. The filler material in the backfill composition (quartz sand) can be considered inert and will not be involved in any chemical interactions with the porewater to any significant extent.

Dissolution of trace amounts of pyrite within the bentonite will help maintain reducing conditions in the near-field, but may generate acidity, according to ambient geochemical conditions. Carbonates, such as calcite or dolomite/ankerite, can be important in reactions buffering pH (Sasaki et al., 1995) because of their rate of reaction. A consideration in this regard is the ambient P_{CO2} of the host rock groundwater which may govern the calculated pH of a bentonite porewater, if carbonate dissolution is assumed to be important.

The bentonite porewater chemistry (and changes over time) is an important control over the release of radionuclides from the near to the far-field because many radionuclides are poorly soluble in equilibrium bentonite porewater chemistries. This is important in the case of the backfill, because the tunnels may act as a fast pathway for groundwater flow in the vicinity of the near-field. This will be particularly the case if the backfill is eroded to leave a water filled void space in the tunnels.

Performance assessments often evaluate radionuclide solubilities on the basis of these bentonite pore-water chemistries because they are considered to be better defined and more time-invariant than are waters in contact with other engineered barrier system components (McKinley and Savage, 1996).

Origin in the repository system:

The water chemistry in the bentonite-rich backfill is influenced by many different processes, including the resaturation of the backfill after repository closure, the composition of the water in the near-field rock, groundwater flow through the backfill and, most importantly, groundwater-bentonite chemical interactions in the backfill.

Impact on the repository system:

The evolving chemistry of the backfill porewaters is an important control on the behaviour of the repository because it impacts on the near-field solubilities of released radionuclides. It also affects other processes, including the composition of the water in the buffer and in the near-field rock.

Bibliographic references:

Arthur RC (1994) Equilibria in saturated bentonite. Scientific Basis for Nuclear Waste Management, XVII, 955-960.

Arthur R and Apted M (1996) Modelling of near-field chemistry for SITE-94. SKI Technical Report, 96:31.

McKinley IG and Savage D (1996) Comparison of solubility databases used for HLW performance assessment. Journal of Contaminant Hydrology, 21, 331-350.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Sasaki Y, Shibata M, Yui M and Ishikawa H (1995) Experimental studies on the interaction of groundwater with bentonite. Scientific Basis for Nuclear Waste Management, XVIII, 337-344.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Wanner H (1986) Modelling interaction of deep groundwaters with bentonite and radionuclide speciation. Nagra Technical Report, NTB 86-21.

Equivalent FEPs:

Relates to FEPs 2.1.09 (Chemical/geochemical processes and conditions in wastes and EBS) and 2.1.04 (Buffer/backfill materials and characteristics) in the NEA International Database (NEA, 1998).

Relates to FEP 4.1.8 (Change of groundwater chemistry in nearby rock) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0 30th June 2000 Dave Savage and Bill Miller

Number: **SFL-55**

Name: **EVOLVING WATER CHEMISTRY IN THE NEAR-FIELD ROCK**

Short description:

The chemistry of the porewater in the near-field rock is defined in terms of the major and trace element concentrations, including repository-derived radionuclides, and the major variables (T, Eh and pH) and any chemical gradients. It is largely controlled by rock-water interactions and by mixing with waters that have interacted with bentonite in the buffer and the backfill. It could be significantly altered if chemically different recharge waters penetrated to depth.

Technical description:

During the operational phase of the spent fuel repository, the repository will be pumped to keep it dry. Consequently, some of the void spaces (pores and fracture apertures) close to the walls of the excavations will be unsaturated. Some of the near-field rock close to the walls will also be oxidised due to chemical interactions with the air and moisture.

After closure, the near-field will resaturate as groundwaters flow in from the far-field to fill all the air-filled void spaces (see SFL-42 “Hydraulic resaturation of the buffer and backfill”). The far-field groundwater initially will be out of equilibrium with chemical conditions in the engineered barriers and in the oxidised zone of rock close to the excavation walls. This will lead to the development of oxidising porewaters due to the dissolution of the free air in the near-field void spaces (see SFL-38 “Redox fronts”).

However, conditions will become chemically reducing over time due to the very large Eh buffering capacity provided by the near-field rock. In general, in a crystalline or argillaceous near-field host rock, rock-water interactions will cause the porewaters to become chemically reducing and pH neutral to mildly alkaline and more saline. However, the exact composition of the near-field rock porewaters will be strongly site specific, dependent on the mineralogical characteristics of the rock mass, the nature of the groundwater flow system and the composition of the far-field waters (see GEN-22 “Far-field groundwater chemistry”).

After passing through the near-field rock, the groundwater will migrate into the engineered barrier system. Thus, the chemistry of the water in the near-field rock may be time dependent and could be affected by the porewaters flowing out of the engineered barriers which have interacted with the bentonite buffer and the canister metals, and any concrete or steel tunnel reinforcements (Savage et al., 1998). The chemistry of the near-field rock porewaters could also change in response to variations in the chemistry of the recharge waters.

Changes to the chemical composition of groundwater in the near-field may lead to precipitation or dissolution of fracture minerals (see GEN-27 “Radionuclide precipitation and dissolution”) thereby influencing both sorption (see GEN-34 “Radionuclide sorption”) and hydraulic properties of the rock.

The composition of the porewaters in the near-field rock will have a significant impact on the longevity of the engineered barrier system. It will affect the degradation rates and processes of the engineered barrier system materials, rock reinforcements, grout and the rock and fracture minerals themselves.

Later, when the canister has failed, the concentration of radionuclides in the near-field rock porewaters may increase if radionuclides are capable of migrating through all of the engineered barrier systems. Likewise, the near-field rock porewaters will also change as soluble products of the degradation of the engineered barriers migrate outwards. At this time, the water chemistry in the near-field rock will control the transport and retardation of radionuclides migrating out to the far-field (see SFL-50 “Radionuclide release and transport from the buffer and backfill”).

Significant changes to the groundwater chemistry in the near-field could occur if chemically distinct recharge waters were to penetrate to depth. This might occur as a consequence of climate change which could lead to fresh, oxidising waters infiltrating under a glacier or, alternatively, the infiltration of saline water when sea levels rise. Both of these scenarios are possible for a coastal site and they were investigated as part of the SITE-94 performance assessment.

Origin in the repository system:

Initially, the near-field rock groundwater chemistry is controlled by the mineralogical composition of the rock mass in contact with the groundwater. This, in turn, is controlled by the porosity and permeability structures of the rock mass, which affect the groundwater flow geometry and residence times. Subsequently, the near-field rock groundwater is modified by waters leaving the engineered barrier system that have chemically interacted with the bentonite and the canister metals.

Near-field groundwater chemistry could be significantly altered if chemically distinct recharge waters penetrated to depth as a consequence of glaciation or sea-level change.

Impact on the repository system:

The water chemistry in the near-field rock may affect the chemistry of the waters in the engineered barrier system. It will also affect the degradation rates and processes of the engineered barrier system materials, rock reinforcements, grout and the rock and fracture minerals themselves.

When the canister has failed, the water chemistry in the near-field rock will control the transport and retardation of radionuclides migrating out of the engineered barrier system.

Bibliographic references:

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

Savage D, McLeod R and McEwen T (1998) An evaluation of repository-induced disturbances for a KBS-3 type repository. SKI Technical Report, 98:20.

SKI (1989) The joint SKI/SKB scenario development project. SKI Technical Report, 89:14.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Wanner H (1986) Modelling interaction of deep groundwaters with bentonite and radionuclide speciation. Nagra Technical Report, NTB 86-21.

Equivalent FEPs:

Relates to FEPs 2.2.08 (Chemical/geochemical processes and conditions in geosphere) and 2.2.01 (Excavation disturbed zone in host rock) in the NEA International Database (NEA, 1998).

Relates to FEP 4.1.8 (Change of groundwater chemistry in nearby rock) in the SKI/SKB Scenario Development Project (SKI, 1989).

Production:

Version 2.0	30th June 2000	Bill Miller

Number: **SFL-56**

Name: **WATER TURNOVER IN THE COPPER SHELL**

Short description:

Water turnover in the outer copper shell of the canister relates to the initial intrusion of groundwater after it has been breached, and the subsequent inflows and outflows of water. The rate of water turnover will be largely controlled by the balance between the groundwater pressures outside the canister and the opposing internal pressures due to gas evolution.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998).

For some long period of time after emplacement, in the absence of manufacturing defects, the canister will remain intact due to the inert behaviour of the copper shell (see SFL-7 “Corrosion of the copper outer canister”). In normal evolution scenarios the canister is expected to remain intact for at least 100 000 years (SKB, 1999). During this period no water turnover will occur in the canister.

Eventually, due to corrosion of the copper outer canister, failure of welds or other structural failures, the copper outer canister will be breached (see SFL-18 “Failure of the copper shell”). At this time, water can penetrate into the void space between the iron inner canister and the copper outer canister as liquid or water vapour, at a rate partly controlled by the hydraulic conductivity of the bentonite buffer. The initial gap between the two components of the reference canister design is 3.5 mm (Werme, 1998), although this will change over time due to creep of the copper (see SFL 11 “Creeping of the metal in the canister”).

Boiling and condensation processes may occur in the void depending on the local temperature (related to the time of failure of the copper outer canister and radiogenic heat production) creating a water vapour phase in the void. The presence of water and water vapour in the void will initiate anaerobic corrosion of the iron inner canister which will, in turn, generate hydrogen (see SFL-9 “Corrosion of the cast iron insert”). As iron corrosion proceeds, the partial pressure of hydrogen may eventually exceed the local hydrostatic pressure and a separate gas phase can evolve. This gas phase may be trapped in the void and, as it grows in volume, may expel groundwater from the canister, limiting further corrosion of the iron inner canister (see SFL-25 “Gas generation in the canister”).

In the case of the KBS-3 style repository where canisters are positioned vertically with the welded lid at the top of the deposition hole and the bentonite buffer controlling the flow of groundwater around the canister, the time for water to enter the void space is estimated to be a few hundred years after perforation. Hydrogen production will then occur until sufficient pressure has built-up to force the water out again. It is estimated to take over a thousand years to expel the water, not consumed by the corrosion process, from a canister with a circumferential breach at the base.

The actual amount of water expelled from the void by the gas will depend on the number, location and size of perforations in the copper outer canister; for example a single perforation half way up the canister would only allow a maximum of half the void space to be cleared of water by the gas pressure. Water remaining in the canister will then be consumed by further corrosion of the iron inner canister.

Progressive iron corrosion will generate increasing volumes of corrosion products and these will gradually fill the void space, restricting further contact between the groundwater and the iron inner canister (see SFL-51 “Expansion of solid corrosion products”).

Continued inflows and outflows of groundwater to the void will be controlled by the changing properties of the failed copper outer canister (e.g. number, location and size of perforations), the changing void volume due to iron corrosion products, and the balance between the groundwater pressures outside the canister and the opposing internal gas (hydrogen, water vapour) pressures (Bond et al., 1997). Ingress of water will continue to be partly controlled by the hydraulic conductivity of the bentonite buffer.

Progressive corrosion of the iron inner canister will affect the chemistry of the water in the void and, as this water is transported out of the copper outer canister, will, in turn, affect the chemistry of the water in the bentonite buffer.

Origin in the repository system:

Water turnover in the copper outer canister occurs after this shell has been breached by corrosion or structural failure. Once the copper outer canister is perforated, the turnover of water in the void between the inner and outer canister components will be controlled by the action of a number of competing processes, including the diffusion (supply) of groundwater through the bentonite buffer; the generation of hydrogen gas and water vapour in the void, and the balance between the hydrostatic and gas pressures; the geometry of the perforations and the volume increase of iron corrosion products in the void.

Impact on the repository system:

Groundwater turnover in the copper outer canister will directly affect the timing and rate of corrosion of the iron insert and, thus, the operational lifetime of the canister as a whole.

Groundwater turnover in the copper outer canister will also affect the chemistry of the groundwater in the void between the inner and outer canister components and, as this water is transported out of the canister, the chemistry of the water in the bentonite buffer.

Bibliographic references:

Bond AE, Hoch AR, Jones GD, Tomczyk AJ, Wiggin RW and Worraker WJ (1997) Assessment of a spent fuel canister: assessment studies for a copper canister with a cast steel inner component. SKB Technical Report, TR 97-19.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and the EBS) in the NEA International Database (NEA, 1998).

Production:

Version 2.0 30th June 2000 Bill Miller

Number: **SFL-57**

Name: **WATER TURNOVER IN THE CAST IRON INSERT**

Short description:

Water turnover in the iron insert of the canister relates to the initial intrusion of groundwater after it has been breached, and the subsequent inflows and outflows of water. The rate of water turnover will be largely controlled by the balance between the groundwater pressures outside the canister and the opposing internal pressures due to gas evolution. Water outflows will be responsible for the transport of dissolved radionuclides away from the canister.

Technical description:

The reference canister design for the spent fuel repository comprises an outer copper shell to assure long-term containment and a cast iron insert to provide mechanical support (Werme, 1998). For some long period of time after emplacement, the iron inner canister will be protected from corrosion by the inert copper shell. After the copper shell eventually has been perforated (see SFL-18 “Failure of the copper shell”), water can penetrate the gap between the iron insert and the copper shell, at a rate partly controlled by the hydraulic conductivity of the bentonite buffer. The gap between the two is 3.5 mm in the reference canister design (Werme, 1998), although this will change over time due to creep of the copper (see SFL 11 “Creeping of the metal in the canister”).

The presence of water in the void between the two canister components will initiate anaerobic corrosion of the iron insert which will generate hydrogen (see SFL-9 “Corrosion of the cast iron insert”). Due to progressive corrosion of the iron canister, failure of welds or other structural failures, the iron insert will also eventually be breached (see SFL-19 “Failure of the cast iron insert”). Until this occurs, no water turnover will take place in the iron insert.

When the iron insert is breached, this will allow ingress of groundwater into the centre of the canister and contact between this water and the inner walls of the iron insert, the fuel assemblies and, eventually, the spent fuel after the Zircaloy cladding becomes perforated. At this time, soluble species segregated into the gap between the fuel pellets and the cladding, and in grain boundaries will be readily mobilised (see SFL-22 “Gap and grain boundary release”).

Contact between the groundwater and the spent fuel will initiate dissolution and conversion of the fuel matrix (see SFL-21 “Spent fuel dissolution and conversion”) and accelerate radiolytic decomposition of the groundwater (see GEN-30 “Radiolysis”).

Boiling and condensation processes may occur within the iron inner canister depending on the local temperature (related to the time of failure of the copper outer canister and radiogenic heat production) creating a water vapour phase in the centre of the canister. The presence of water and water vapour will initiate anaerobic corrosion of the iron inner canister and the metallic components of the fuel assemblies which will, in turn, generate hydrogen (see SFL-9 “Corrosion of the cast iron insert”).

As iron corrosion proceeds, the partial pressure of hydrogen may eventually exceed the local hydrostatic pressure and a separate gas phase can evolve. This gas phase may be trapped in the iron canister and, as it grows in volume, may expel groundwater from the canister, limiting further corrosion of the canister, the metallic components of the fuel assemblies and the spent fuel. The actual amount of water expelled from the canister by the gas will depend on the number, location and size of the perforations.

Corrosion of the iron inner canister will also generate increasing volumes of corrosion products and these will gradually fill the void space in the canister, restricting further contact between the groundwater and the iron inner canister.

Continued inflows and outflows of groundwater to the iron inner canister will be controlled by the changing properties of the failed copper and iron canister components (e.g. number, location and size of perforations), the changing void volume due to iron corrosion products, and the balance between the groundwater pressures outside the canister and the opposing internal gas (hydrogen, water vapour) pressures (Bond et al., 1997). Ingress of water will continue to be partly controlled by the hydraulic conductivity of the bentonite buffer.

In addition, the inflows and outflows of water may be partially driven by any temperature gradient across the whole canister. The extent of this temperature gradient will be related to the time of failure of the canister and continued radiogenic heat production. However, considering that the copper outer canister is expected to remain intact for at least 100 000 years (SKB, 1999), the temperature gradient across the canister by the time water turnover within the iron inner canister can occur is expected to be small.

Water outflow from the iron inner canister is the primary mechanism for transporting radionuclides released from the spent fuel from the canister to the bentonite buffer. Water turnover in the inner canister will thus affect the chemistry of the water in the canister and, in turn, the chemistry of the water in the bentonite buffer.

Origin in the repository system:

Water turnover in the iron inner canister occurs after the copper outer canister and, subsequently, the iron inner canisters have both been breached by corrosion or structural failure.

Once the iron inner canister is perforated, the turnover of water in this canister will be controlled by the action of a number of competing processes, including the diffusion (supply) of groundwater through the bentonite buffer; the generation of hydrogen gas and water vapour in the canister void space, and the balance between the hydrostatic and gas pressures; the geometry of the canister perforations and the volume increase of iron corrosion products in the void.

Impact on the repository system:

Groundwater turnover in the iron inner canister will directly affect the timing and rate of corrosion of the spent fuel. Furthermore, water turnover in the iron inner canister will be responsible for the transport of radionuclides out of the canister to the bentonite buffer. As a consequence, water turnover in the iron inner canister is a primary control on repository performance. Groundwater turnover in the iron inner canister will also affect the chemistry of the groundwater in the canister components and, as this water is transported out of the canister, the chemistry of the water in the bentonite buffer.

Bibliographic references:

Bond AE, Hoch AR, Jones GD, Tomczyk AJ, Wiggin RW and Worraker WJ (1997) Assessment of a spent fuel canister: assessment studies for a copper canister with a cast steel inner component. SKB Technical Report, TR 97-19.

NEA (1998) An international database of Features, Events and Processes. NEA/OECD.

SKB (1999) SR-97: processes in the repository evolution. SKB Technical Report, TR 99-07.

SKI (1996) SITE-94: deep repository performance assessment project. SKI Technical Report, 96:36, Two Volumes.

Werme L (1998) Design premises for canister for spent nuclear fuel. SKB Technical Report, TR 98-08.

Equivalent FEPs:

Relates to FEP 2.1.08 (Hydraulic/hydrogeological processes and conditions in wastes and the EBS) in the NEA International Database (NEA, 1998).

Production:

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