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Swedish Radiation Safety Authority

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Technical Note

2012:10

Review of the Geomicrobiological Aspects of SKB's Licence Application for a Spent Nuclear Fuel Repository in Forsmark, Sweden

Report number: 2012:10 ISSN: 2000-0456
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SSM perspektiv

Bakgrund
Strålsäkerhetsmyndigheten (SSM) granskar Svensk Kärnbränslehantering AB:s (SKB) ansökningar enligt lagen (1984:3) om kärnteknisk verksamhet om uppförande, innehav och drift av ett slutförvar för använt kärnbränsle och av en inkapslingsanläggning. Som en del i granskningen ger SSM konsulter uppdrag för att inhämta information i avgränsade frågor. I SSM:s Technical note-serie rapporteras resultaten från dessa konsultuppdrag.

Projektets syfte
Granskning av SKB:s redovisning av hur mikrobiell aktivitet på förvarsdjup påverkar produktion av sulfid. Syftet med granskningen är att bedöma och utvärdera SKB:s redovisning, om osäkerheter bedömts, bedöma kompletteringsbehov samt föreslå vad SSM bör granska inom detta område i huvudgranskningsfasen.

Författarnas/Författarens sammanfattning
Det rekommenderas att långsiktiga (år) experiment genomförs under representativa förhållanden som kan råda efter deponering av kapslar i slutförvarsanläggningen, inklusive: radioaktivt material i koppar / järn kapseln, b) förväntade temperaturvariationer och variation i buffertens vattenmättnad, i syfte att bedöma om de variationer i omgivande miljö som beskrivs ovan, och delas av andra (Stroes-Gascoyne et al., 2010), har betydande inverkan på SKB: s nuvarande bedömning av begränsad bakteriell korrosion av kopparkapslar.

Projektinformation
Kontaktperson på SSM: Jan Linder
Diarienummer avrop: SSM2011-3086
Aktivitetsnummer: 3030007-0502

SSM perspective

Background

The Swedish Radiation Safety Authority (SSM) reviews the Swedish Nuclear Fuel Company's (SKB) applications under the Act on Nuclear Activities (SFS 1984:3) for the construction and operation of a repository for spent nuclear fuel and for an encapsulation facility. As part of the review, SSM commissions consultants to carry out work in order to obtain information on specific issues. The results from the consultants' tasks are reported in SSM's Technical Note series.

Objectives of the project

To evaluate whether the research work by SKB has been adequately comprehensive to describe the influence of microbial activity for sulphide production at repository depth. To document any uncertainties with regard to predictions of rates and consequences of microbial sulphide production.

Project information

Contact person at SSM: Jan Linder

Summary by the author

It is recommended that long-term (years) experiments are conducted under conditions as close as possible to those likely to prevail in the repository, including: a) radioactive material in model copper/iron canisters, b) simulatedexpected temperature and clay saturation changes; in order to assesswhether the uncertainties described above, and shared by others (Stroes-Gascoyne et al., 2010), have significant impact on SKB's current assessmentof limited bacterial corrosion of the copper canisters.

Strål
säkerhets
myndigheten

Swedish Radiation Safety Authority

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Technical Note 1

2012:10

Review of the Geomicrobiological Aspects of SKB's Licence Application for a Spent Nuclear Fuel Repository in Forsmark, Sweden

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Content

1. Objectives

To consider whether the research work by SKB has been adequately comprehensive to describe the influence of microbial activity for sulphide production at repository depth? To document any uncertainties with regard to predictions of rates and consequences of microbial sulphide production

2. Sulphate-reducing bacterial activity in the buffer and backfill and implications for canister corrosion

2.1. General Statement of findings

1) SKB's assessment relies almost entirely on the results of the research that they commissioned from the University of Göteborg. This is not unreasonable, as there is no directly comparable research available. However, there is considerable evidence of sulphate-reducing bacterial activity over geological time scales, and enhancement of activity due to the presence of metals and corrosion, radioactivity, and elevated temperatures, that needs also to be taken into consideration when assessing potential microbially influenced corrosion of the nuclear fuel canisters.

2) Sulphate reducing bacterial activity is often associated with pitting,rather than uniform corrosion, hence, if even the limited microbial corrosion rates considered by SKB (~3 mm in 10^6 years) were concentrated in pits, then the potential for perforation of the canister would be increased considerably.

3) Due to these and other considerations, it is recommended that new, long-term (years) experiments are conducted, under conditions as close as possible to those likely to occur in the repository. These would need to include radioactive material in model copper/iron canisters, simulated expected temperature and clay saturation changes, in order to assess whether the uncertainties described in the report, and shared by others (e.g. Stroes-Gascoyne et al., 2010), have significant impact on SKB's current assessment of limited bacterial corrosion of the copper canisters during long-term storage in the subsurface repository.

3. Review Method

SKB's documentation and referenced published research was considered and then a review of other relevant literature was conducted. Relevant publications were obtained from the Web of Science^{SM}, a personal reference data base and more general Web searches for relevant scientific information.

3.1. Documents Reviewed

Documents reviewed include SKB 2011 TR-11-01 Volume 1, Long-term safety for the final repository for spent nuclear fuel at Forsmark, Main report of the SR-Site project Volume I to page 259 for detailed background information and Volume II Section 10.3.13 Canister evolution, particularly “Sulphate reducing bacteria in buffer and backfill, and TR-10-46 Fuel and canister process report for the safety assessment SR-Site section 3.5.4 “Corrosion of copper canister”.

4. SKB’s Position

SKB states that “Although there are uncertainties as to the fraction of organic matter that could be used by sulphate reducing bacteria, the corrosion due to sulphide produced by microbial activity with organic matter in the repository or anaerobic corrosion of steel as energy source is considered to have negligible impact on the copper thickness, even in a 10^6 year perspective (TR-11-01 Volume 2, page 421) and summarized elsewhere in the reports”.

5. Evaluation and analyses of SKB’s Position

SKB’s assessment relies almost entirely on the results of the research that they commissioned from the University of Göteborg (Pedersen, 2010, Masurat et al., 2010a, b). This is not an unreasonable position, as there is no directly comparable research available. However, there is considerable literature demonstrating continued activity of sulphate-reducing bacteria over geological time scales and enhanced activity of sulphate-reducing bacteria in interaction with metals and corrosion, that should also be taken into consideration when assessing potential microbially influenced corrosion (Beech and Sunner, 2007) of the copper canister.

1 There is clear evidence that sulphate-reducing bacteria are active on geological time scales and in subsurface consolidated rocks. Active bacterial sulphate reduction has been demonstrated in Cretaceous (~145 to ~65 Ma) rocks at ~170 to 300 m depth, Cerro Negro, New Mexico (Krumholz et al.,1997). Although activity was greatest at sandstone-shale interfaces, therewas measurable activity in the Clay Mesa Shale interval. This is consistent with detection of viable sulphate reducing bacteria in the

Wyoming bentonite MX-80 (which conforms to required specification for use in the repository) documented in SKB report and quoted references (Masurat et al., 2010a, Masurat et al., 2010b, Pedersen, 2010); plus in Callovo-Oxfordian clay rock studied as a potential host for a nuclear waste repository in France (El Hajj et al., 2010). Referring to the paper of Masurat (Masurat et al., 2010a), the SKB report states (TR-11-01 Volume 2, page 421) that "In the experiments by /Mazurat et al. 2010/ it can thus not be excluded that some of the formed copper sulphide stemmed from sulphide diffusing in from the circulating groundwater, and not only from sulphide produced by microbial activity in the bentonite. The values in Figure 10-85 ((TR-11-01 Volume 2, page 421) are thus overestimates of the sulphide production in the bentonite for several reasons". This interpretation is not completely correct, as the total sulphide produced was calculated from the ^{35}S-sulphide produced during the experiment, and as there is no exchange between the added ^{35}S-sulphate and preformed sulphide (Fossing and Jorgensen, 1990) all the reported sulphide was newly formed. In addition, subsequent experiments by Pedersen (Pedersen, 2010) using similar experimental setup were isolated from the borehole during incubation, confirming that all the ^{35}S-sulphide was produced within the experimental system and was not from perfusing groundwater. Hence, sulphide production rates were not overestimates due to sulphide from groundwater. It is correct that in the experiments of Masurat (Masurat et al., 2010a), sulphate reduction activity was stimulated by the addition of lactate, which would have artificially increased sulphide formation, and thus the extrapolations up to 10^6 years. Within the experiments by Pedersen (Pedersen, 2010), however, a control was used without any substrate addition and still measurable Cu_XS was produced, and at all densities of clay used. Further, the reduced concentrations of ^{35}S in the clay compared to the circulating water could indicate active sulphate reduction in the clay, and increasing overall rates of sulphate reduction corresponded with decreasing ^{35}S in the clay (Table 1, Pedersen, 2010). Again this result is consistent with viable sulphate reducing bacteria being present in MX-80 bentonite clay (Masurat et al., 2010b) and active sulphide production in clay exposed to onlysterile filtered groundwater (Masurat et al., 2010a).

2 As both copper in the canister (Nies, 1999) and the radioactivity being generated by the enclosed spent nuclear fuel, are detrimental to many microbes, it might be considered that microbial corrosion would be very limited, even over 10^6 year timescales, which is consistent with the view of SKB. However, the formation of CuS by the activity of sulphate reducing bacteria protects them against copper toxicity (Temple and Le Roux, 1964), and radioactivity may have some beneficial aspects. For example, at 2.8 km depth in the South African, Mponeng Gold Mine, a thermophilic (50-60°C) sulphate-reducing bacterial species is sustained over geological time scales by radiolytic decomposition of pyrite to sulphate and hydrogen (Lin et al., 2006). Pyrite is present in the clays considered by SKB (0.4 wt %, TR-11-01 page 180, Table 5-10). Hydrogen can also be formed from radiolysis of water (TR-10-46 page 110). Oxidants (e.g. O_2, oxygen, H_2O_2, hydrogen peroxide) are also produced by this process that can be used by microbes to oxidise the H_2 and provide energy, hence, removing potential feedback inhibition and allowing further formation of radiolytic products. This positive feedback was not considered in the SKB report (TR-10-46 page 110), and if on-going radiolytic H_2 consumption was coupled to sulphate reduction, higher rates of sulphide induced copper corrosion could occur, in a similar way to the stimulation of Cu_XS formation (by a factor of 7) by external H_2 addition in the experiments of Pedersen (Pedersen, 2010). SKB comment that it will take about 317 years (TR-10-46 page 110) until irradiation has substantially decreased, which is a long time to potentially have stimulated microbially activity close to the canister. Corrosion itself can also produce H_2 (Beech and Sunner, 2007), and this H_2 can stimulate microbial activity, including sulphate-reducing bacteria (Dinh et al., 2004, including possibly even more effectively by direct removal of electrons from iron). In fact H_2 consumption by sulphate reducing bacteria is considered one mechanism of microbial enhancement of corrosion, via cathodic depolarization (Beech and Sunner, 2007). Sulphate reducing bacteria are often associated with pitting, rather than uniform corrosion, hence, if even the limited microbial corrosion rates considered by SKB (~3 mm in 10^6 years TR-11-01 Volume 2, page 421) were concentrated in pits rather than uniform corrosion, then the potential for perforation of the canister would be increased. If the overall

estimated 3 mm corrosion depth over 10^6 years was concentrated by a factor of ~17 in corrosion pits, this would lead to canister perforation. Perforation might further enhance microbial activity and corrosion due to greater access to radiolytic products, and the iron insert etc. Figure 1 of Pedersen (Pedersen, 2010), clearly shows that build up of Cu_xS corrosion products is heterogeneous, which could lead to pitting corrosion.

3 Radioactive decay of the spent nuclear fuel will generate heat with the highest temperatures and the greatest heat flows occurring within tens of years after deposition and repository closure. However, surface temperature of the outer copper shell will be below 100°C TR-10-46, page 80) and with heat generation being halved every 30 years. Considering the upper temperature limit for prokaryotes is 122°C (Takai et al., 2008) and for sulphate-reducing bacteria 110°C (Jorgensen et al., 1992), temperatures in the repository will consistently enable the presence of active bacteria. In addition, higher initial temperatures may enhance bacterial activity, as many bacteria have optimum activities above ~20°C and higher. In addition, a period of higher, then decreasing temperatures may stimulate activation of bacterial spores (Masurat et al., 2010a, Stroes-Gascoyne et al., 2010), which could utilize organic compounds from vegetative bacterial cells killed by the initial higher temperatures (note organic carbon released would be limited due to low prokaryotic biomass in bentonite clay, (Stroes-Gascoyne et al., 2010)). There are a number of known spore forming sulphate-reducing bacteria and some of these are present in subsurface environments (Lin et al., 2006). The average organic carbon content of the two clays considered in SKB's reports (TR-11-01, Volume 1 page 180, Table 5-10) is 0.2 wt %. However, this will probably be recalcitrant material which is closely attached to the clay and, hence, not readily degradable/hydrolysed by bacterial activity (TR-11-01, Volume II page 420). It needs to be considered, however, that this would mean that this organic matter will be very slowly degradable over geological time scales, allowing continuing very low levels of bacterial activity (Fry et al., 2009). In addition, local heating near the canisters would likely increase the degradability of organic matter within the clays (Parkes et al., 2007), and thus stimulate bacterial activity; especially in the presence of radiolytically generated oxidants (e.g. oxygen, hydrogen peroxide, sulphate) . The experiments to simulate bacterial activity

and corrosion in the repository, which are quoted in the SKB reports did not consider the potential impacts ofelevated temperatures; experiments being conducted at ~22°C (Pedersen, 2010).

6. Recommendation

It is recommended that long-term (years) experiments are conducted under conditions as close as possible to those likely to prevail in the repository, including: a) radioactive material in model copper/iron canisters, b) simulated expected temperature and clay saturation changes; in order to assess whether the uncertainties described above, and shared by others (Stroes- Gascoyne et al., 2010), have significant impact on SKB's current assessment of limited bacterial corrosion of the copper canisters.

7. References

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APPENDIX 1

Coverage of SKB reports

Table 1:

Reviewed report	Reviewed sections	Comments
TR-11-01 Volume 1	[*insert reviewed sections*]	[*insert comments, if any*]
TR-11-01 Volume 2		
TR-10-67		

2012:10

The Swedish Radiation Safety Authority has a comprehensive responsibility to ensure that society is safe from the effects of radiation. The Authority works to achieve radiation safety in a number of areas: nuclear power, medical care as well as commercial products and services. The Authority also works to achieve protection from natural radiation and to increase the level of radiation safety internationally.

The Swedish Radiation Safety Authority works proactively and preventively to protect people and the environment from the harmful effects of radiation, now and in the future. The Authority issues regulations and supervises compliance, while also supporting research, providing training and information, and issuing advice. Often, activities involving radiation require licences issued by the Authority. The Swedish Radiation Safety Authority maintains emergency preparedness around the clock with the aim of limiting the aftermath of radiation accidents and the unintentional spreading of radioactive substances. The Authority participates in international co-operation in order to promote radiation safety and finances projects aiming to raise the level of radiation safety in certain Eastern European countries.

The Authority reports to the Ministry of the Environment and has around 270 employees with competencies in the fields of engineering, natural and behavioural sciences, law, economics and communications. We have received quality, environmental and working environment certification.

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