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Quality Assurance Review of the Swedish Nuclear Fuel and Waste Management Company's LOT Experiment (Phase S2 and A3) at the Äspö Facility in Sweden

2021:06

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SSM perspective

Background

This report presents a quality assurance (QA) review of the work done by SKB to retrieve the S2 and A3 parcels from the Long term test of buffer material (the LOT experiment) at the Äspö Hard Rock Laboratory. Each LOT parcel comprises a heated copper tube surrounded by bentonite, with a number of copper coupons and various other test and monitoring instruments included in the bentonite. The S2 and A3 test parcels were recovered in 2019, after 20 years of LOT operation and SKB has analysed corrosion of the copper coupons and tubes from the parcels. The QA review has focused on SKB's copper corrosion analysis.

SKB's management of the LOT S2 and A3 project and the reports on dismantling the test parcels (TR 20 11) and analysing the corrosion of the copper coupons and copper tubes (TR-20-14) were reviewed. This provided an understanding of the reliability of the results from a QA perspective. The review found that SKB's management and QA arrangements were appropriate, meeting modern standards. SKB engaged a number of contractors to work on the project, who all have extensive experience and appropriate management systems for such work. The corrosion experts from the contractor teams worked collaboratively with SKB and co-authored the corrosion report TR 20-14

Results

It was found that some aspects of the way the LOT project was set up in the 1990s mean that there are limitations in terms of what can be learnt about copper corrosion. For example, the copper coupons, copper tubes and copper reference materials were not pre-characterised. This means that it is difficult to distinguish between defects associated with material preparation and machining and the effects of corrosion under LOT conditions. Also, redox conditions were not monitored so the time of transition from aerobic to anaerobic is uncertain, and there were no measurements of microbial populations in groundwater, so that no clear conclusions can be drawn on the relative effects of microbes and copper corrosion on oxygen consumption.

SKB argues that O2 was the main oxidant causing copper corrosion before the O2 was consumed, followed by a period in which aqueous Cu2+ may have prevailed as an intermediate oxidant. A long period of minor anaerobic corrosion may have occurred as a result of diffusion of low concentrations of sulphide from groundwater to the copper surfaces. However, uncertainty in the saturation time of the parcels and the effects of different oxygen consumption processes mean that alternative interpretations of system evolution and oxygen availability for corrosion could be made. For example, if full saturation coupled with rapid microbial consumption of oxygen had occurred before the tubes could be exposed to a significant period of increased temperature, then a temperature-dependent anaerobic process would have been responsible for corrosion before any arrival of sulphide. However, any copper corrosion by sulphide attack would far exceed the corrosion depths of penetration that have been estimated could occur by anoxic corrosion in pure water in saturated bentonite. Thus, corrosion by sulphide attack is of greater concern in safety assessments than any postulated corrosion in oxygen-free water. Also, alternative arguments do not support the observation from analysis of different LOT parcel tests conducted over different lengths of time that most corrosion appears to have occurred in the early stages of the tests when conditions are likely to have been aerobic. Thus, although it is not possible to conclude with absolute certainty that corrosion of the copper tubes and coupons occurred predominantly under aerobic conditions in the early stages of LOT, there is no evidence available from these results to suggest that SKB's interpretation of copper corrosion behaviour during LOT exposures is incorrect.

Relevance
The main objective of this project is to provide a detailed analysis and assessment of SKB's approach to quality assurance in the decommissioning of the LOT S2 and A3 parcels, focusing on the analysis of copper corrosion.

Project information
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Content

Summary

The Swedish Nuclear Fuel and Waste Management Company (SKB) began the Long term test of buffer material (the LOT experiments) at the Äspö Hard Rock Laboratory (HRL) in Sweden over 20 years ago. LOT has comprised a series of experiments involving seven 'test parcels', with each parcel comprising a heated copper tube surrounded by bentonite and placed in a vertical borehole in granite. LOT test parcels S2 and A3 were recovered in 2019, after 20 years of operation. This has enabled a study of the coupled processes that affect bentonite and copper behaviour based on collection of test data over a relatively long period. SKB has used the results of the LOT S2 and A3 study to check its model for the initial evolution of the engineered barriers used in the safety assessment of a KBS-3 spent fuel repository.

SKB submitted a licence application for a spent fuel repository based on the KBS-3 concept in 2011 and the Swedish Government's consideration of the licence application is ongoing. The Swedish Radiation Safety Authority (SSM) has a responsibility to review SKB's work relating to concept development and implementation (including the licence application). This includes checking that SKB's work is underpinned by sound project management and appropriate quality assurance (QA) procedures. This report presents a QA review, conducted on behalf of SSM, of the LOT S2 and A3 parcel tests, focusing on the analysis of copper corrosion.

SKB's management of the LOT S2 and A3 project has been reviewed in order to understand how QA procedures have been applied. In addition, SKB's reports on dismantling the S2 and A3 test parcels (TR-20-11) and on analysing the corrosion of copper coupons and copper tubes from the test parcels (TR-20-14) have been reviewed. During the course of the work, meetings were held with SKB in order to discuss questions arising from the QA review. Also, the interests of a number of Swedish environmental organisations have been considered via a meeting and document review.

SKB's management and QA arrangements for LOT were found to be appropriate, meeting modern standards. The project management system and responsibilities for activities in the LOT project have inevitably changed over the two decades that the project has run to date. The changes do not appear to have had any significant detrimental impacts on how the project has been run.

The timing of the LOT S2 and A3 parcel recovery and analysis has received some criticism from those who believe that SKB has delayed parcel retrieval in order to suppress the copper corrosion debate. SKB had originally planned to recover the LOT S2 and A3 parcels after five years, but extended this to 20 years in line with delays in repository licensing. SKB noted that internal documents from 1999 mention the possibility of running the experiment for a 20-year period. The plan to retrieve the test parcels in 2019 was stated in SKB's 2016 RD&D programme. The extended experimental period has allowed a longer exposure period for the copper.

SKB has approached openness and transparency in the LOT S2 and A3 project by ensuring that parcel retrieval was filmed, and by making bentonite samples available for analysis on request, although this depends on the objectives and competence of the interested researcher. However, as noted in the discussion of results below, certain decisions on the analyses to be undertaken and presented are unclear.

The LOT S2 and A3 project is organised according to work packages, with details of the work set out in SKB's activity plans or work plans provided by contractors against work package requirements. SKB engaged a number of contractors to work on the LOT S2 and A3 project: Clay Technology AB, Uppländska Bergborrning AB, RISE KIMAB AB and Swerim AB. These contractors all have extensive experience and appropriate management systems for such work. Even so, SKB's experts were significantly involved throughout the LOT S2 and A3 corrosion analysis and wrote report TR-20-14 collaboratively with RISE KIMAB and Swerim. SKB has been provided with all results and images from the corrosion analysis performed by its contractors. SKB stores raw data in its SICADA database management system and information such as photos, reports and memos in a document management system.

The original objective of LOT was to validate models and hypotheses about the properties of bentonite buffer material as well as microbiology, radionuclide transport, copper corrosion and gas transport processes under repository-like conditions. However, certain aspects of the way the LOT project was set up over twenty years ago mean that there are inevitable limitations in terms of what can be learnt about copper corrosion under repository conditions. In particular, the analysis of copper corrosion under LOT conditions has been hampered by the fact that the copper coupons, copper tubes and copper reference materials were not pre-characterised. This means that it is difficult to distinguish between defects of mechanical origin associated with material preparation and machining, the effects of corrosion after 20 years under LOT conditions, and the effects of any corrosion of the reference materials after 20 years of dry storage. Also, no measurements of microbial populations in groundwater were made for the LOT experiment. Therefore, no clear conclusions can be drawn about the influence of microbial populations on oxygen consumption and how it might have affected the amount of oxygen available for aerobic corrosion of copper. The time at which conditions might have transitioned from aerobic to anaerobic is uncertain; there was no monitoring of redox conditions - which would have aided such understanding - because limited technologies were available to measure redox potential in compacted bentonite in the 1990s.

Test parcel S2 was exposed to standard KBS-3 repository conditions (maximum temperature of almost 100°C) and parcel A3 was exposed to adverse conditions (maximum temperature of almost 140°C), but the parcels were not subjected to the radiation conditions that would be expected in the repository and bentonite swelling pressures were lower than would be expected because of the smaller scale of the LOT experiment. Analyses of the copper coupons and samples of the copper tubes from these tests do confirm some temperature dependence of corrosion in LOT. However, the corrosion analysis of the parts of the copper tubes that experienced the highest temperatures was limited. For instance, copper from the hottest parts of tube A3 was not examined metallographically, although corrosion in such regions was estimated based on measurements of the accumulated mass of copper corrosion products that had diffused into the bentonite next to the tube. SKB has stated that the selection of tube samples for analysis was done for the practical reason that they were in the same bentonite blocks as the corrosion coupons and that the maximum temperatures experienced by the tube samples that were examined are close to the peak temperature that any copper canister in the KBS-3 repository would be expected to experience.

Based on measurements of the accumulated mass of corrosion products in the bentonite next to the tube samples, average corrosion depths were estimated to be 0.2 to 4.8 μm for the LOT S2 copper tube and 0.2 to 13.8 μm for the LOT A3 copper tube. The largest average corrosion depths are associated with the copper tube samples that experienced the highest temperatures. Corrosion analyses for other LOT parcels (A0, S1 and A2) retrieved after 1 year and 6 years show similar temperature-dependent average depths of corrosion. Such observations suggest, but do not prove, that corrosion occurred early. It is not possible to infer how much corrosion occurred before the tube heaters were switched on and had reached their maximum temperatures; there was a period of about four months between LOT S2 and A3 parcel installation and the heaters being switched on, and it took a few months for maximum temperatures to be reached. Also, the more rapid consumption of oxygen by corrosion in the warmer parts of the tube is likely to have drawn oxygen from the cooler parts to the warmer parts, thereby enhancing total aerobic corrosion in the warmer parts and reducing it in the cooler parts. Such an axial thermal gradient would not be present along disposal canisters in a repository, because heat would be generated more uniformly along the length of the canister by radioactive decay of the spent fuel. It can be concluded that the maximum integrated corrosion rate of 0.7 μm/year for the hottest part of LOT A3 copper tube, when assuming that corrosion occurred at a uniform and linear rate with respect to time for the duration of the 20-year experiment, is not representative of, and most likely overestimates, the long-term corrosion rate for copper.

The total average accumulated corrosion depth of the LOT S2 and A3 copper coupons was estimated to be 0.7 to 1.3 μm based on gravimetric analysis. This is reasonably consistent with observation of coupon corrosion in other LOT parcels (1.5 to 4.8 μm). Differences are likely to be due to spatial variations in local conditions and the lower temperatures in the vicinity of the coupons.

SKB argues that O_2 was the main oxidant causing the corrosion of the copper coupons and tubes in LOT before the O_2 was consumed. Aqueous Cu^{2+} may have prevailed as an intermediate oxidant on a much longer timescale. However, the cathodic reactant Cu^{2+} is finite in such a closed system, assuming that it is only produced during oxic corrosion, and can be expended to produce a finite depth of attack. A long period of minor anaerobic corrosion, except for uncertain areas of non-uniform attack, may have occurred as a result of the slow diffusion of low concentrations of sulphide from groundwater to the copper surface to form insoluble Cu_2S, although evidence of the Cu-S phase on the tubes is limited. Cross-sections of corroded copper surfaces do indicate inner scale consisting of Cu_2O and outer layers of Cu_xS, which could be interpreted to support the notion that an initial short period of oxygen-induced corrosion was followed by a long period of anoxic sulphide-induced corrosion. However, observations of thicker corrosion products in pits and the lack of detailed analysis of surface anomalies versus pitting leaves open the possibility that copper pitting has occurred. Understanding the analysis of micrographic cross-sections, and any dependency there might be on temperature, is hampered by the lack of clarity in how the cross-sections relate to locations on the copper tubes.

Uncertainty in the saturation time of the LOT parcels and the effects of different oxygen consumption processes does mean that alternative interpretations of system evolution and oxygen availability for corrosion could be made. For example, if the gaps around the copper tubes had filled with water rapidly before the tubes could be exposed to a significant period of increased temperature, and rapid microbial consumption of oxygen had occurred, then a temperature-dependent anaerobic process would have been responsible for corrosion before any arrival of sulphide. However, any copper corrosion by sulphide attack would far exceed the corrosion depths of penetration that have been estimated could occur by anoxic corrosion in pure water in saturated bentonite. Thus, corrosion by sulphide attack is of greater concern in safety assessments than any postulated corrosion in oxygen-free water.

Alternative arguments, however, do not support the observation from analysis of different LOT parcel tests conducted over different lengths of time that most corrosion appears to have occurred in the early stages of the tests when conditions are likely to have been aerobic. Thus, although it is not possible to conclude with absolute certainty that corrosion of the copper tubes and coupons occurred predominantly under aerobic conditions in the early stages of LOT (noting the above observations about possible pitting), there is no evidence available from these results to suggest that SKB's interpretation of copper corrosion behaviour during LOT exposures is incorrect.

1. Introduction

1.1. Background

The Swedish Nuclear Fuel and Waste Management Company (SKB) began the Long term test of buffer material (the LOT experiments) at the Äspö Hard Rock Laboratory (HRL) in Sweden over 20 years ago. LOT has aimed to support understanding of how the bentonite buffer surrounding a copper disposal canister will behave under disposal conditions (e.g., its swelling pressure, the possibility of alteration of smectite to illite at elevated temperatures, and the possibility of salt enrichment through a cyclic condensation/evaporation process), as well as investigating related copper corrosion, microbiological, radionuclide transport, and gas transport processes [1]. The tests have focused on studying system behaviour during the period after which the buffer has saturated and the barriers are subject to the highest expected temperatures. The tests are described as 'long term' because they have been, and continue to be, conducted over periods long enough to study bentonite behaviour at full water saturation, but are not long term in the sense of the timescales of concern to the overall performance a KBS-3 spent fuel repository.

LOT has comprised a series of experiments involving seven 'test parcels', where a test parcel comprises a stack of prefabricated bentonite blocks placed around a copper tube in a vertical borehole in granite. The copper tube in each parcel contains a heater. Three test parcels (S1 to S3) have been exposed to standard KBS-3 repository conditions (maximum temperature of almost 100°C) and four parcels (A0 to A3) have been exposed to adverse conditions (maximum temperature of almost 140°C).

Tests A1 and S1 were 'pilot tests' (A1 and S1) that were run for about 1 year, being terminated in 1998 and subsequently analysed by Karnland *et al*. [2]. The central part of the A1 parcel was, however, lost during the drilling operation to extract the parcel. In response, SKB installed an additional short-term test parcel A0 to complement the A1 test; the A0 parcel was run for almost two years and was recovered in 2001, with the A0 parcel analysis reported by Karnland *et al*. in 2011 [3].

Test parcels A2, A3 and S2 were originally planned to run for 5 years [1]. Test parcel A2 was recovered in 2006 after about 6 years of operation and the A2 parcel analysis was reported by Karnland *et al*. in 2009 [4]. However, test parcels A3 and S2 were not recovered until 2019, after 20 years of operation. The dismantling of the parcels A3 and S2 has been reported by Sandén and Nilsson [5] and the results of the copper corrosion analysis have been reported by Johansson *et al*. [6]. SKB is preparing a report of the A3 and S2 parcel buffer analysis, but it is not expected to be available until 2022. The final test parcel S3 has been operating for 21 years and SKB plans to terminate and recover it in 2023 [7].

SKB submitted a licence application for a spent fuel repository based on the KBS-3 concept in 2011 and the Swedish Government's consideration of the licence application is ongoing. The Swedish Radiation Safety Authority (SSM) has a responsibility to review SKB's work relating to the licence applications. An important aspect of SSM's role is to check that SKB's work is underpinned by sound project management and appropriate quality assurance procedures. In this respect, SSM has previously undertaken quality assurance (QA) reviews of SKB's tests and experiments related to the performance of the KBS-3 barrier system.

Galson Sciences Ltd (GSL) (a UK-based consultancy) has supported these QA reviews, which has included consideration of the LOT experiment and, in particular, the S1, A0, A1 and A2 test parcel analyses [8, 9, 10, 11]. In continuation of this process, SSM has requested that GSL provides a QA review of the LOT S2 and A3 parcel tests, as reported by Sandén and Nilsson [5] and Johansson *et al*. [6], focusing on the copper corrosion analyses. In addition, Prof. John Scully of the University of Virginia, USA, has been requested to provide an independent review the reliability and traceability of SKB's findings on copper corrosion presented in the copper corrosion analysis report [6]. Prof. Scully has previously provided reviews of the treatment of copper corrosion in SKB's post-closure safety assessment for the licence application for a spent fuel repository in Sweden [12], as well as supplementary information provided by SKB to support the licence application [13]. This report presents the results of the LOT QA review.

1.2. Objective and Approach

The main objective of this project is to provide a detailed analysis and assessment of SKB's approach to quality assurance in the decommissioning of the LOT S2 and A3 parcels, focusing on the analysis of copper corrosion.

As well as reviewing the SKB reports on dismantling the S2 and A3 test parcels (Sandén and Nilsson [5]) and on analysing the corrosion of copper coupons and copper tubes from the test parcels (Johansson *et al*. [6]), the review has considered SKB's overall management of the LOT project in order to understand the framework under which QA procedures have been applied. The latter part of the work has involved reviewing information provided by SKB on its project management model and how it has been applied to the LOT project as a whole and specifically to the project to dismantle, retrieve and analyse the LOT S2 and A3 test parcels.

A series of video conference meetings involving SKB, SSM and GSL has been a key component of the work. Initially, a meeting was held on 18th September 2020 to discuss the scope and objectives of SSM's QA review and SKB's control documents and instructions relevant to the LOT tests [14, 15]. Three project review meetings followed, which involved discussion of specific aspects of QA in the LOT S2 and A3 project, as follows:

> Meeting 1: 'Management system and project management', 5th November 2020 [16].
>
> Meeting 2: 'Retrieval, sampling, handling of samples and analysis', 13th November 2020 [17].
>
> Meeting 3: 'Interpretation of results', 27th November 2020 [18].

Before each review meeting, SSM and GSL provided SKB with a set of topical questions. SKB presented written responses to these questions during the meetings and each topic was discussed and explored further as necessary so that SSM and GSL could fully understand SKB's views on the topic. The records of these meetings were provided by SKB [16, 17, 18], and they included the topic questions, SKB's written responses to the topic questions and a note of the topic discussions. SSM and GSL reviewed the meeting records for accuracy and provided comments where clarifications were required and where it was deemed that further information would be beneficial.

Given that SKB has employed a number of sub-contractor organisations (Clay Technology AB, Uppländska Bergborrning AB, RISE KIMAB AB and Swerim AB) to support the LOT parcel retrieval operations and sample analyses, the approaches to project management and QA followed by these organisations have also been reviewed under this LOT QA project. This part of the review included a visit to the shared RISE KIMAB and Swerim facilities in Kista, Stockholm, where the LOT S2 and A3 project copper corrosion analyses were undertaken, to discuss the quality system used at the facilities [19]. SSM and SKB participated in the meeting with RISE KIMAB; GSL staff were unable to participate owing to travel restriction associated with the COVID-19 pandemic.

The interests of certain environmental organisations and corrosion scientists have been considered as part of the LOT QA review project. In particular, on 30th September 2020, SSM and GSL held a video conference meeting with:

- Johan Swahn and Joachim Stormvall of the Environmental Organisations' Nuclear Waste Review (MKG);
- Oscar Alarik of the Swedish Society for Nature Conservation (Naturskyddsföreningen); and
- Peter Szakálos and Christofer Leygraf of Szakálos Materials Science AB/Royal Institute of Technology (KTH).

This meeting was held in order to understand the groups' views on what is important for the QA review of the LOT S2 and A3 project [20]. Subsequently, MKG provided a number of documents and reports that relate to concerns about the recovery, analysis and interpretation of the LOT S2 and A3 test parcels. In summary, the principal concerns raised are as follows:

- Microbial activity may have been responsible for consuming a substantial amount of the oxygen present in the initial phase of the LOT experiment, which would mean that there was less oxygen available than assumed by SKB for copper corrosion under aerobic conditions. In this case, some of the observed corrosion would have occurred under anaerobic conditions.
- The hottest and therefore most corroded parts of the copper tubes and the copper bottom plates have not been studied sufficiently; surface images and metallographic cross-section images and analysis of these regions are not available. The differences in corrosion behaviour between the copper tubes and the bottom plates is not explained.
- The locations of samples for which metallographic cross-sections are presented in the corrosion analysis report [6] are not clear, and descriptions of the surfaces are not sufficiently detailed to allow full understanding of the relevance of the results.
- It is not clear how the gaps in the LOT parcels were filled with groundwater via titanium tubes at the start of the experiment and whether the gaps could have been filled before the parcels were sealed and the heaters turned on. Potentially, only oxygen trapped in bentonite would have been available for corrosion and this may have been affected by microbial consumption of oxygen.
- The amount of corrosion that could have occurred in the four-month period before groundwater was injected and the heaters were switched on is not clear.
- Other repository experiments indicate the rapid development of anoxic conditions. For example, the REX experiment at the Äspö HRL found that oxygen in rock fractures was consumed in a few days, largely by microbes but also by mineral reactions [21], the MiniCan experiment at Äspö showed

that oxygen in compacted bentonite was consumed in a period of months [22], the Full-Scale Emplacement (FE) experiment at the Mont Terri rock laboratory in Switzerland (Opalinus Clay) showed anoxic conditions developing along a bentonite backfilled tunnel after a few months [23], and conditions in the FEBEX experiment in Switzerland may have become anoxic early in the experiment [20].

- The corrosion products remaining on the copper surfaces have not been included in the estimates of the amount of corrosion based on the mass of corrosion products found in the bentonite.
- Discussion is required on whether any oxygen could have leaked into the LOT parcels and affected copper corrosion.
- The limited corrosion on the inside of the copper tubes that have been exposed to air for 20 years and how it differs from corrosion on the outside of the tubes is not sufficiently well explained.
- Differences in copper corrosion in the A2 and A3 LOT parcels are not sufficiently well explained.
- Publication of research results is controlled by SKB, which could mean that any results found that do not favour SKB's model of corrosion are excluded.

SSM and GSL considered these concerns when preparing questions for the above-noted project review meetings with SKB and during review of SKB's documents.

The findings of the QA review are presented in this report. The conclusions drawn are based on the evidence available to explain how the quality of the LOT S2 and A3 test parcel copper corrosion analyses has been assured in the context of the overall LOT test carried out over the last 20 years. This includes findings relating to how uncertainties and any credible alternative interpretations of the LOT S2 and A3 test parcels have been considered based on the test measurements and observations made.

1.3. Report Structure

A summary description of the LOT S2 and A3 test parcels is provided in Section 2. The review of SKB's overall project management and QA framework for the LOT S2 and A3 tests is discussed in Section 3, and the review of the copper corrosion analysis and interpretation of results is discussed in Section 4. Overall conclusions of the QA review are presented in Section 5.

2. LOT Test Parcels S2 and A3

Details of the composition of the LOT S2 and A3 test parcels and their instrumentation are provided in SKB's report TR-20-11 on parcel installation, monitoring, dismantling and analyses [5]. In summary:

- The copper tubes used in the test parcels were manufactured from SS 5015-04 grade copper; the tubes were 4,700 mm long, with an inner diameter of 100 mm and a wall thickness of 4 mm. A copper plate and four copper reinforcement parts were welded to the bottom end of each tube. The upper part of the copper tube was open to the Äspö tunnel so that the interior of the tube was filled with air during the entire exposure period. The impenetrability of each copper tube was tested after soldering by use of a helium source inside each tube and an external detector.
- Wyoming bentonite (MX-80) was used to manufacture the bentonite blocks and plugs. After installation of the test parcels, an air-filled annular gap remained between the bentonite blocks and rock surface (approximately 10 mm wide) and between the bentonite blocks and the central copper tube (approximately 1 mm wide). These gaps were gradually filled with water in conjunction with the test start. The borehole diameter was 300 mm so that after saturation the buffer was 96 mm thick.
- The test parcels rested on about 100 mm of sand, and sand-filled pilot holes (76 mm diameter) beneath the parcels.
- To mitigate concerns that the inflow rates from fractures in the boreholes would be too low to saturate the bentonite in an acceptable time, external groundwater was added to the test holes during the test period via a system of titanium tubes connected to a water-bearing fracture that had been intersected by drilling into the tunnel wall nearby.
- A number of the bentonite blocks included instruments to measure total pressure, pore pressure, relative humidity and temperature, as well as copper coupons (to be used to quantify total corrosion) and ^{60}Co tracer doped plugs (to study radionuclide migration in compacted bentonite). The locations of the instrumentation, copper coupons and tracer plugs in the S2 and A3 test parcels are shown in Figure 1 and Figure 2, respectively.

The test is smaller than the reference design for KBS-3, where the canister has a diameter of 1,050 mm and a length of 4,835 mm, and the buffer has a nominal thickness of 350 mm, with 500 mm of buffer below the canister and 1,500 mm of buffer above it [24]. The smaller scale shortens the time for full water saturation to be reached [5], although with a lower swelling pressure than would be expected in the repository.

The LOT A3 and S2 test parcels were installed at the Äspö HRL in September to October 1999. For parcel S2, the heater was switched on and the water injection was started 133 days (almost 4.5 months) after installation. For parcel A3, heating and water injection began 112 days (a little under 4 months) after installation. The titanium tubes were open in the period between parcel installation and test start. A titanium tube in the sand below each bottom plate was used to fill the voids with water when the tests started, although there may have been some water inflow from the rock before then. Tubes at the periphery of block 32 in each parcel were used for de-airing during water filling and thereafter to inject water.

During the 20-year test period, parcel A3 was maintained under ‘adverse’ conditions, where the maximum temperature near the copper tube reached about 140°C after one year, after which time it was reduced to about 120°C. Parcel S2 was maintained under ‘standard’ conditions where the maximum temperature near the copper tube was about 90°C throughout the test period. Temperature distributions for each parcel based on monitoring data for the day before the heaters were switched off are shown in Figure 3.

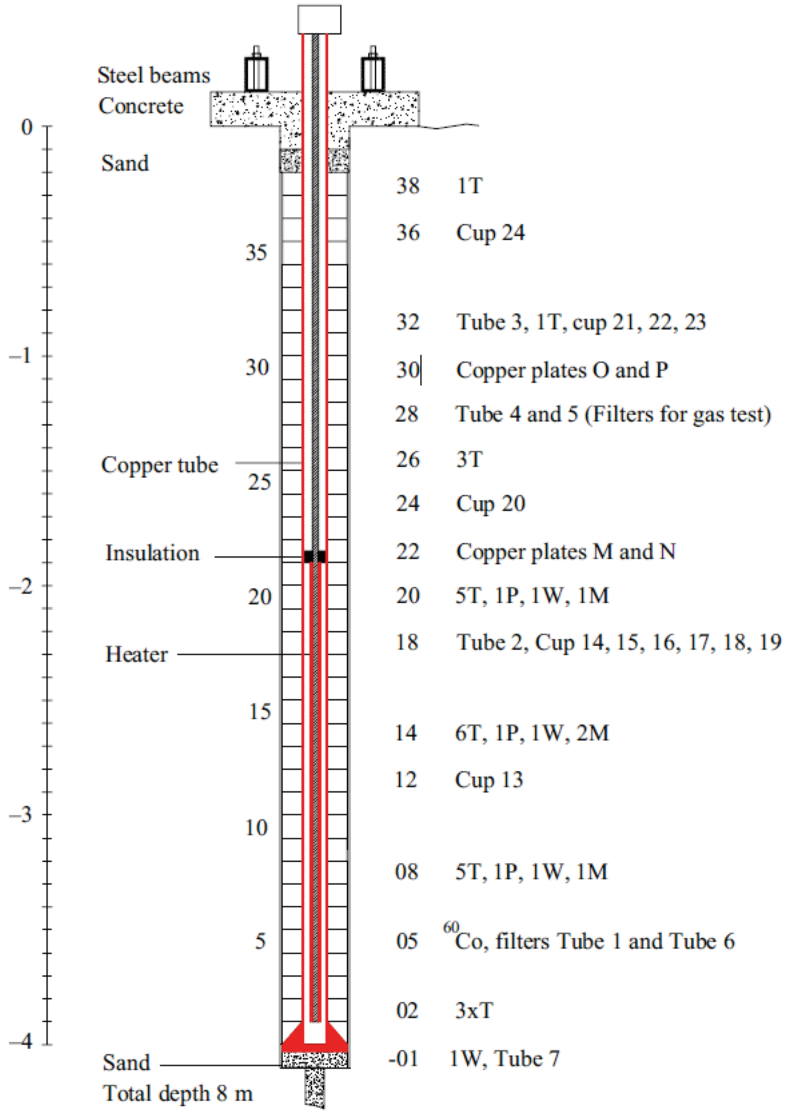

Figure 1 LOT S2 Test Parcel with the instruments, bentonite block numbers and depth indicated (T = thermocouple, P = total pressure sensor, W = water pressure gauge, M = relative humidity gauge, Cup = water sampling cup, Tube = titanium tube with filter). Figure from SKB report TR-20-11 [5].

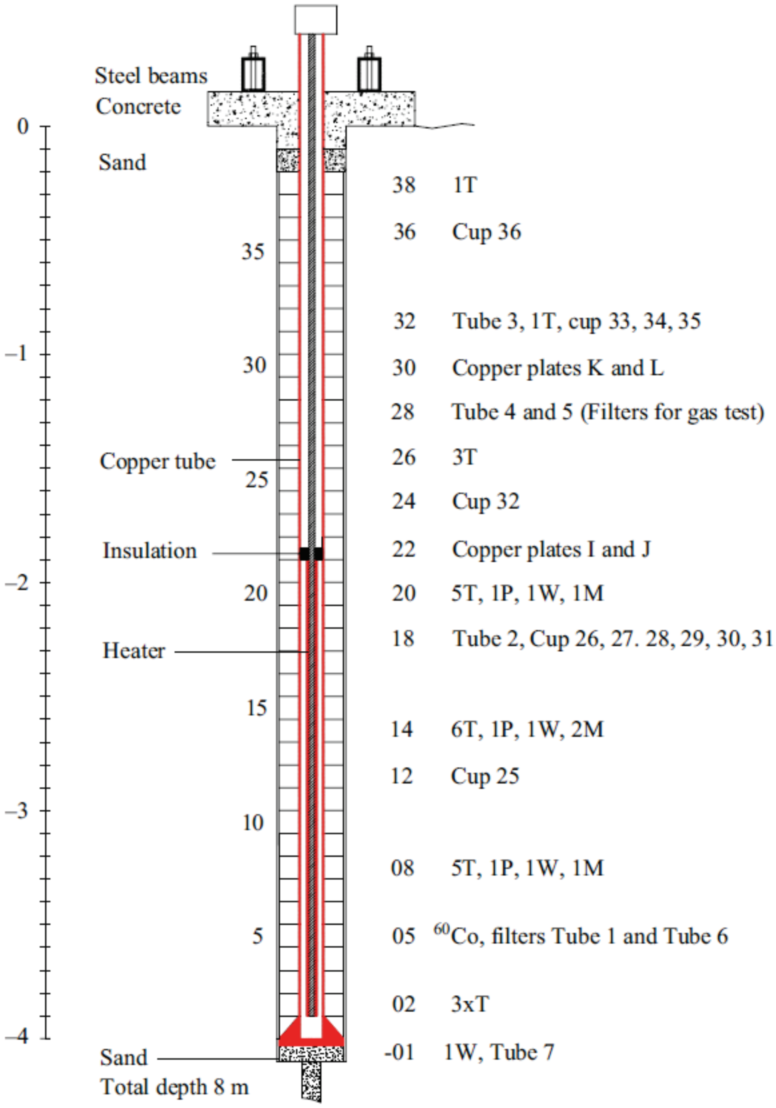

Figure 2 LOT A3 Test Parcel with the instruments, bentonite block numbers and depth indicated (T = thermocouple, P = total pressure sensor, W = water pressure gauge, M = relative humidity gauge, Cup = water sampling cup, Tube = titanium tube with filter). Figure from SKB report TR-20-11 [5].

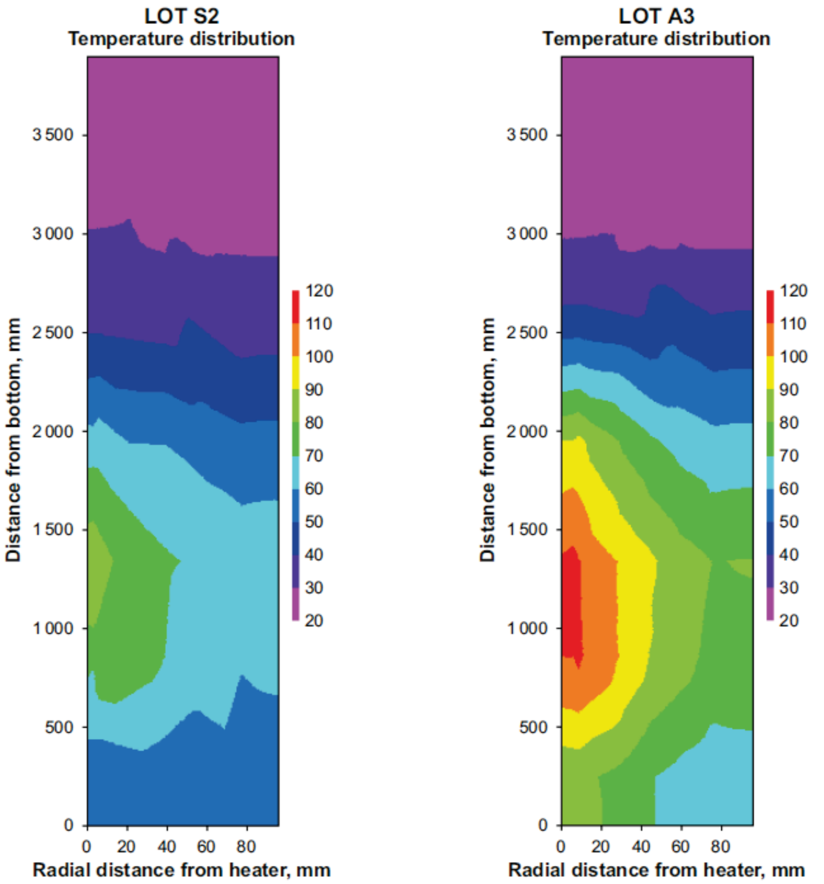

Figure 3 Contour plots of temperature (°C) in test parcels S2 (left) and A3 (right) on the day before the heaters were switched off. Figures from SKB report TR-20-11 [5].

The measured temperatures in the hottest part of LOT parcel S2 (block 14) as a function of time (starting from 1st September 1999 - just before the time of parcel installation) and radial distance from the tube are shown in Figure 4. It took a little over four years for full saturation to be reached in the bentonite near to the hottest part of parcel S2 (near the centre of block 14) according to the time it took for pressure to reach a steady state (see Figure 5) and for the relative humidity to reach 100% (see Figure 6). SKB [5] noted that, in such tests, the relative humidity sensors usually become contaminated with saline water once the bentonite has become fully saturated, which leads to erroneous results, as seen in Figure 6. Note that swelling pressures are expected to be in the range 4.5 to 13 MPa in the repository [24].

The measured temperatures in the hottest part of LOT parcel A3 (block 14) as a function of time and radial distance from the tube are shown in Figure 7. It appeared to take about four years for pressure to reach a steady state (see Figure 8) near to the hottest part of the parcel (near the centre of block 14), but the sensor stopped working after four years. It took only 0.5 to 2.5 years for saturation to be completed (according to data from relative humidity sensors - Figure 9), suggesting an ongoing bentonite swelling process after saturation.

The LOT A3 and S2 test parcels were dismantled in September 2019, which involved extracting the parcels from their boreholes by overlapping drilling, and transporting the parcels to a laboratory to be divided for analysis [5]. Details of the copper corrosion analysis are provided in SKB report TR-20-14 [6].

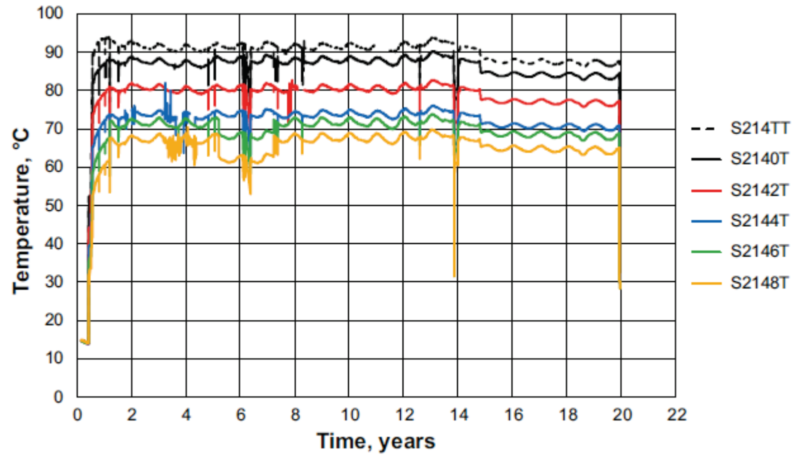

Figure 4 Temperature as a function of time and radial distance from the copper tube in block 14 of test parcel S2 [5]. The small drop in temperature after about 14 years is because of an exchange of power regulators.

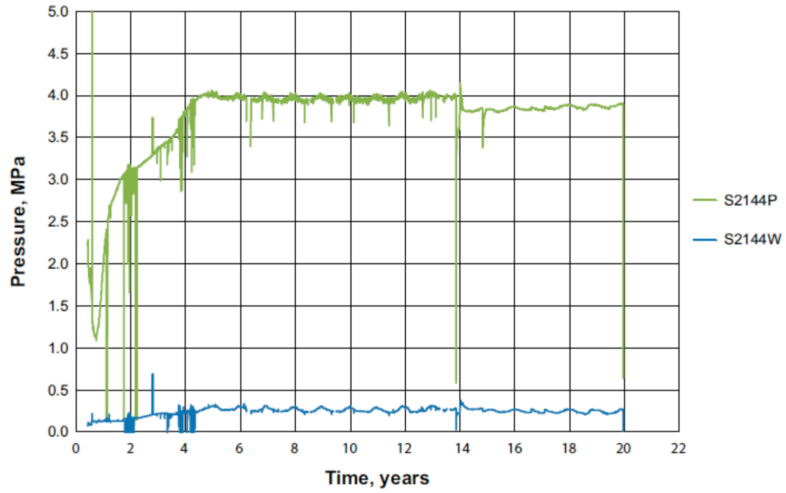

Figure 5 Total pressure, S2144P, and water pressure, S2144W, as a function of time near the centre of block 14 in test parcel S2 [5].

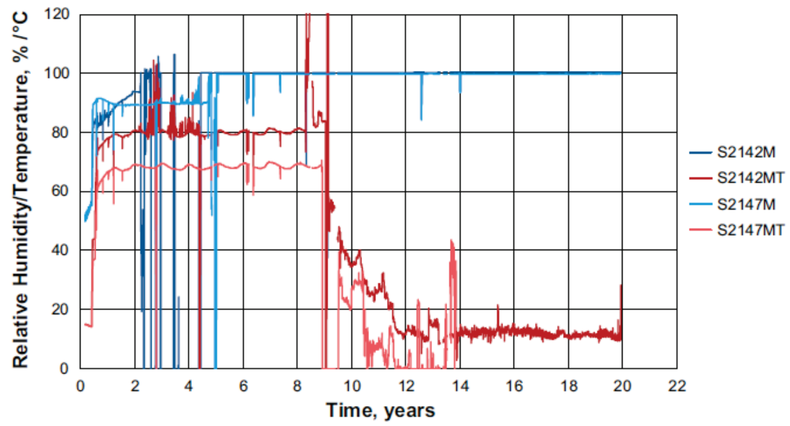

Figure 6 Relative humidity (blue) and temperature (red) as a function of time in block 14 of test parcel S2 [5]. Full saturation appears to have occurred after about four years, after which time results are not reliable.

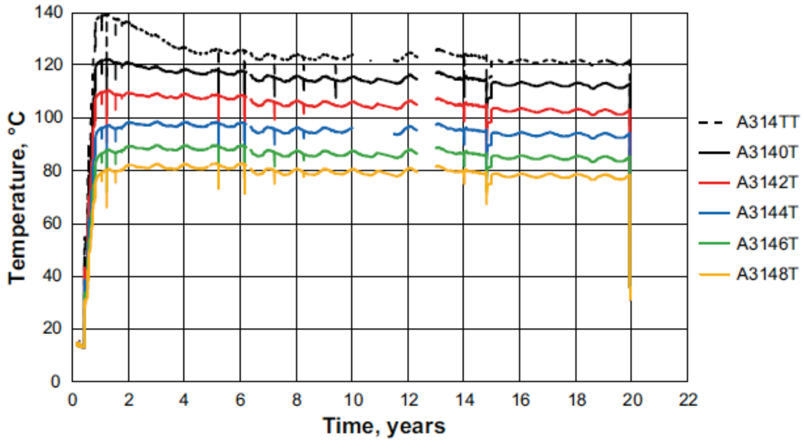

Figure 7 Temperature as a function of time and radial distance from the copper tube in block 14 of test parcel A3 [5]. Data gaps are a result of problems with data loggers.

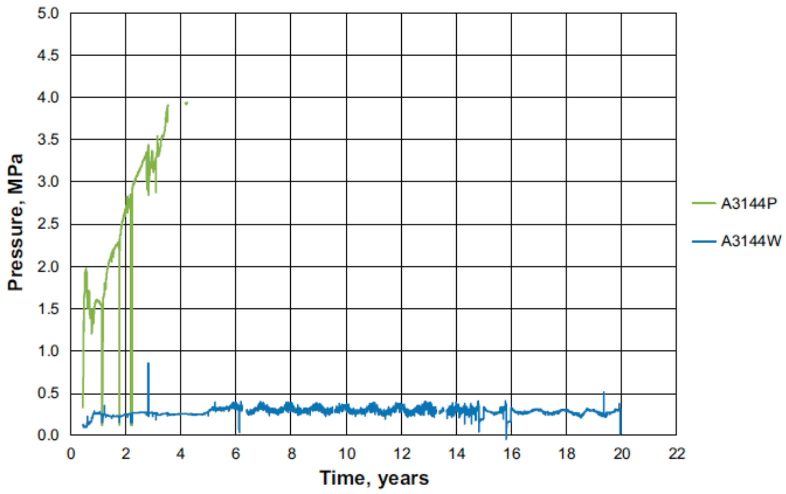

Figure 8 Total pressure, A3144P, and water pressure, A3144W, as a function of time near the centre of block 14 in test parcel A3 [5]. The pressure sensor stopped working after about four years.

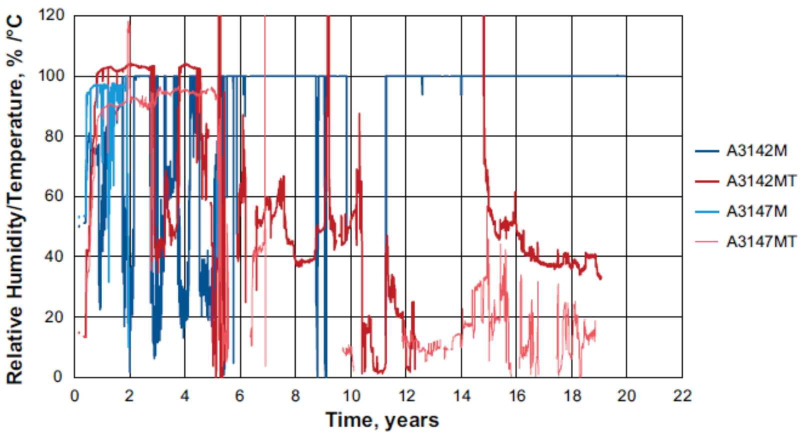

Figure 9 Relative humidity (blue) and temperature (red) as a function of time in block 14 of test parcel A3 [5]. Full saturation appears to have occurred after about two to three years, after which time results are not reliable.

3. SKB's Project Management and Quality Assurance for LOT

This section presents a review of SKB's management of the LOT test, and the steps taken by SKB to assure that the work undertaken by its staff and contractors in the preparation, running, dismantling, analysis and reporting of the LOT test (specifically relating to test parcels S2 and A3) is of a suitably high quality. First, SKB's overall project management approach is discussed, and then QA procedures relating to measurement methods, working with external suppliers, data and records management, and reporting for the LOT S2 and A3 test parcels are reviewed. Findings are based on review of project management documents provided by SKB and discussions at the project review meetings, the first of which focused on topics related to LOT project management.

3.1. Project Management Approach

3.1.1. Project Planning

SKB manages its activities within a company-wide project management model [25], which aims to establish a framework for how projects are initiated, implemented and completed. This approach is consistent with the project management approach defined by the energy company Vattenfall AB [26], which is one of the four organisations that own SKB - the other four owners are Forsmarks Kraftgrupp AB, OKG Aktiebolag and Sydkraft Nuclear Power AB. The project management model requires a staged decision-making process, which includes tollgates (or decision points) at specific points in the process, as well as development of a Project Initiation Note (PIN), Project Charter, Project Management Plan (PMP) and Project Assurance Review (PAR) plan.

The LOT project as a whole was, however, established before SKB's current management system was implemented. Plans for LOT were documented via test plans developed in the 1990s, such as set out in SKB's International Progress Report IPR-99-01 [1], but dismantling activities for the LOT parcels are now managed via dedicated projects according to SKB's project management model. This has meant that responsibilities for managing different aspects of the LOT project have changed over the years. For example, during the first QA review meeting, SKB noted that data from LOT monitoring and test parcel analysis are transferred to SKB's relational database management system SICADA, and that deliveries from LOT to SICADA were managed by Clay Technology AB up to 2012, after which time the experiment was transferred to SKB's administration for the Äspö HRL, with activities such as data transfer to SICADA managed according to activity plans [16].

The timing of the LOT S2 and A3 parcel recovery and analysis has received some criticism from those who believe that SKB has delayed parcel retrieval in order to suppress the copper corrosion debate. At the first project QA review meeting [16], the reason for delaying the recovery of the parcels was discussed; LOT S2 and A3 were originally planned to run for five years, consistent with the then planned schedule for construction of the spent fuel repository. The idea was to dismantle the experiments before the first real canister was deposited, and when the planning for LOT started, repository operations were expected to start in 2008. However, SKB noted that there are internal documents from 1999 mentioning the possibility of running the experiment for a 20-year period that became a reality due to delays in constructing the repository. SKB also commented that, in general, a longer duration of a field scale test is positive from a scientific point of view because of the opportunity to acquire more data on system behaviour, although sensors may start to fail. However, for LOT, although the exposure time is longer, the size of the data set on copper corrosion does not change.

Under SKB's project management model, dismantling, retrieval and analysis of the LOT S2 and A3 test parcels is being managed as project '*KBP1019, Dismantling and evaluation of LOT S2 and A3*', which was initiated in 2018. Copies of the PIN [27], Project Charter [7] and PMP [28] for project KBP1019 were supplied by SKB for the QA review.

The objective of a PIN is to identify the project goals, budget, schedule, decision points (tollgates), responsibilities, and project review and quality assurance requirements, and to document any planned deviations or additions to the standard project model instruction. The PIN [27] for project KBP1019 was created at the start of the project; it is a brief document that meets the requirements of a PIN, although it refers to the Project Charter [7] for information on project goals and activities, staff responsibilities, and the types of project review required at each project stage.

The SKB project management model requires that the following project tollgates, as illustrated in Figure 10, are always used [25, §4; 26, §5.3]:

- TG0 = Decision to start a project.
- TG1 = Decision on which alternative concept solution to select.
- TG2 = Decision on requirements and scope.
- TG3 = Decision on realising the project result.
- TG4 = Decision on start of hand-over to receiving organisation.
- TG5 = Decision on accepted hand-over and start conclusion of the project.

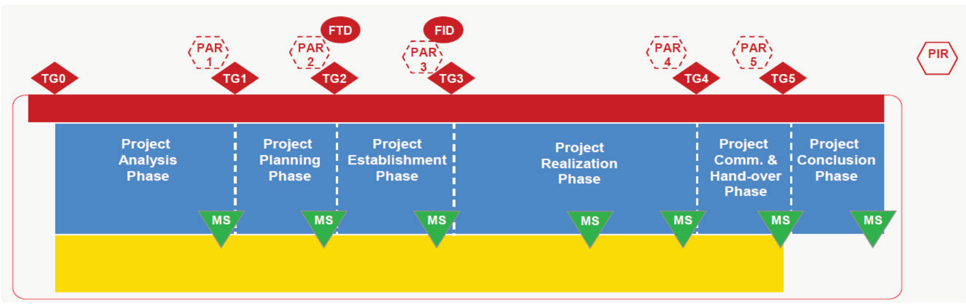

Figure 10 Project management lifecycle [26, §5.3].

SKB [25, §4] states that each tollgate decision is preceded by a mandatory milestone where the project manager together with the project team conducts an evaluation of the documentation to be considered in the decision, and ensures that all criteria for proceeding to the next phase are met and that the project is considered ready for the tollgate decision. The tollgate decision is documented in a milestone report. In addition to the required milestones and tollgates, the SKB project manager can add their own milestones to facilitate the project's structure, progress and follow-up [25, §4], and, if there are more specific needs to be accomplished within a project phase, the tollgate criteria can be refined to reflect these [26, §5.3].

For the LOT S2 and A3 project, the PIN [27, §6] notes that tollgates TG2 and TG3 were effectively merged, with the TG2 and TG3 decisions planned to be made at the same time in June 2019 (i.e., the decision to begin parcel retrieval, dismantling and analysis). At the first QA review meeting [16], SKB stated that no additional milestones were added for the project, although two changes were made to the plans set out in the original Project Charter:

- The PIN [27, §2] states that the LOT S2 and A3 project was originally due to be completed in December 2021. However, the project schedule has been updated, with TGs 4, 5 and 6 delayed by two years owing to unavailability of internal resources to undertake the bentonite analyses. SKB judged that the bentonite analyses are, in general, not time critical and so deemed the delay to be acceptable. The exception is the bentonite analysis required to support the assessment of copper corrosion, and so this analysis was prioritised and undertaken according to the original schedule, as reported by Johansson *et al*. [6].
- Dedicated studies to measure microbial activity were included in the first version of the Project Charter. However, the activity and survival of microbes was studied for the LOT A2 parcel and the results gave no new information on microbes compared to other tests. Also, microbial activity in the form of sulphate reduction cannot be studied in the LOT setup. Therefore, during project planning, SKB concluded that questions relating to the effects of microbes would be better addressed in other dedicated experiments so that microbial activity studies were removed from the project, as reflected in the revised Project Charter [7].

The tollgate decisions are recorded in protocols (minutes) from LOT steering group meetings. These minutes were reviewed by SSM during their visit to the RISE/Swerim facility on 26th November 2020 [19] and records of the above-noted decisions to change the project schedule and scope were identified.

The Project Charter [7, §4.2.1] requires that review comments from SSM on the repository licence application that are relevant to issues addressed by the project must be accounted for in project planning. Three relevant SSM comments were identified in the PMP [28, §4.5], along with commentary on how the project will support SKB's response to these comments:

- SSM has commented that SKB needs to improve its data on the copper phases that would form in bentonite under repository conditions. The PMP states that the total copper content of bentonite samples from the LOT test parcels will be measured using X-ray fluorescence (XRF), and scanning electron microscopy (SEM) and X-ray diffraction (XRD) will provide information regarding the type of copper phases present. If optical microscopy identifies any green/blue parts of the bentonite, or if for some other reason corrosion products are suspected of being present, further analyses will be performed to try to distinguish which minerals are present.

- SSM commented that some buffer conversions can be caused by elevated temperatures and high pH, and these processes need to be elucidated. The PMP states that LOT was designed to study buffer conversion at high temperature. The project will investigate the bentonite from many positions in the warmer parts of the experiment using XRD, and XRF will be used to study the clay to see if its elemental composition has changed, and in particular if illitisation has occurred. However, consideration of the effects of high pH on the buffer is outside the scope of the LOT S2 and A3 project. Cement plugs were only included in the bentonite of the LOT parcels A0 and A2 to generate high pH conditions [1].
- SSM commented that a more detailed understanding is needed of how the slow build-up of swelling pressure can impact the deformation of the copper casing, microbial sulphate reduction and other degradation processes such as copper corrosion processes associated with the presence of gas. The PMP states that deformations of the copper casing will not be studied in LOT because the tube geometry is different to that of the KBS-3 copper canister. Any microbial sulphate reduction will be measured indirectly through analysis of the chemical and elemental composition of the corrosion film. The PMP states that local corrosion is not expected under unsaturated conditions in the presence of gas, but the topography of the copper surfaces will anyway be investigated.

3.1.2. Risk Management and Lessons Learnt

The PMP [28, §9] states that project risks are identified in a Risk List. SSM examined the Risk List in the visit to the RISE/Swerim facilities [19]. At the first QA review meeting [16], SKB explained that the project risks and risk mitigation plan were initially identified through the Project Manager's review of documentation (activity plans and reports) from previous LOT parcel dismantling activities. The initial list of risks was then further developed by the project group, which included several personnel with experience from earlier LOT dismantling activities as well as from other Äspö installation and dismantling projects. SKB considered that it was valuable that members of staff from Clay Technology who had been involved in the LOT experiment from its installation were able to contribute to the development of the Risk List. This is because, even though such experience is recorded in reports, and risks are noted in activity plans from previous test parcel retrievals, hands-on practical experience is more difficult to document and transfer to others. Thus, it is beneficial to use the same team for test parcel retrieval as previously. SKB noted that risks are also considered in the risk assessment included in each activity plan, with activity plan authors taking account of the main Risk List during production of the activity plans [16; 29, §4.2, §9].

The Risk List is treated as a living document, and risks have been reviewed and added at working meetings and project group meetings [16]. The most significant risks are also recorded in Antura (an SKB management system) for monthly reporting to the SKB project client.

The PIN for the LOT S2 and A3 test parcel recovery and analysis project [27, §5] states that a Project Assurance Review (PAR) is required at TG3; project assurance is defined in the project management model as an external quality and value assurance method that is carried out at a minimum in connection with tollgate decisions to support control and decision-making [25, §5.2.3]. A Project Health

Check (PHC) review in connection with the steering group meetings is required at other project stages. SKB [16] stated that four potentially significant risks were highlighted as part of the PAR in the TG3 decision meeting relating to:

- availability of internal resources;
- schedule;
- cost; and
- potential damage of packages by water during dismantling.

The schedule was updated (as noted in Section 3.1.1) and the budget was increased, as approved at TG3, thereby mitigating risks associated with resource availability, schedule and cost. Mitigation plans associated with potential damage by water involved suction of water during dismantling, and use of alarms [16].

Two risks were realised during the LOT S2 and A3 test parcel recovery and analysis process [16]:

- It was intended that the test parcels would be extracted by overlapping percussion drilling in the surrounding rock, partly because percussion drilling does not require cooling water (which could influence conditions in the bentonite). However, problems with the accuracy of the drilling for the first parcel (A3) meant that the boreholes did not overlap each other completely and the risk handling plan to drill additional larger core holes had to be implemented to remove the remaining rock. This caused some delays, but the cooling water required for core drilling was pumped away according to the mitigation plan and there were no implications for the parcel. Problems with the percussion drilling equipment were resolved and the issue was avoided during retrieval of the second parcel (S2).
- A risk relating to ordering delays was realised, with the Mössbauer analysis used to measure the oxidation state of iron in the bentonite having to be delayed, although this is not relevant to the copper corrosion analysis. The bentonite samples were stored in vacuum-sealed bags in order to keep them stable until the analysis could be performed.

At the first QA review meeting [16], SKB stated that lessons learnt will be reported in an experience report at the end of the project. No formal notes are kept while the project is running, although the Project Manager has recorded key findings, such as the challenges with the percussion drilling of the first parcel and the tight fit of the crane when lifting the parcels (which will be an even tighter for the final parcel S3 and will likely require a modified lifting procedure).

3.1.3. Stakeholder Communication

The PMP [28, §3.3] acknowledges that those who disagree with SKB’s findings on copper corrosion processes have expressed the view that SKB has deliberately delayed LOT S2 and A3 parcel retrieval and analysis in order to suppress the debate on copper corrosion. Thus, SKB recognises that recovery and analysis of the copper coupons and heaters from the S2 and A3 parcels has significant value in supporting SKB's credibility and the viability of the KBS-3 concept. The PMP concluded that there may be a benefit in project management, engineers and technicians being able to take part in communication activities relating to retrieval and break-up of the S2 and A3 parcels. A physical copy of the project communication plan for LOT S2 and A3 parcel retrieval and analysis was reviewed by SSM during the visit to the RISE/Swerim facilities [19].

The Project Charter [7, §2.2] identified Posiva as an external stakeholder with particular interest in the LOT S2 and A3 project, to be informed of the project and to be consulted regarding collaboration opportunities before TG1 (see Figure 10). The Project Charter also noted that a number of external stakeholders wanted to observe the project and that this needed to be taken in to account when planning the project. SKB stated at the first QA review meeting that the retrieval project was discussed with Posiva and bentonite samples were provided; additional samples will be supplied upon request [16]. SKB did consider allowing impartial observers to be present during S2 and A3 parcel recovery, with different alternatives to enable this discussed by the project steering group [16]. Normally, SKB does not invite impartial observers to the retrieval of long-term experiments, but exceptions have been made in the past. Different options were discussed, but access to Äspö and safety limitations during the retrievals, as well as challenges in identifying an appropriate observer, led SKB to decide that filming the retrieval of the test parcels was the best option and so impartial observers were not invited. Furthermore, SKB stated that the LOT experiment is not considered to be unique and chose to handle it according to normal procedures [16]. Note that the plan for Work Package 1 (parcel retrieval) [29, §4.2.4] states that the retrieval work will be followed by an independent 'auditor' from SP Swedish Technical Research Institute; it is assumed that this section of the document was not updated following the decision not to include impartial observers.

SKB did film the LOT test parcel retrieval process to a large extent, including the percussion and core drilling, lifting and dismantling of the test parcels, cutting of the copper tubes, and extraction of the copper coupons. The SKB webpage on the LOT S2 and A3 parcel retrieval project (https://www.skb.se/nyheter/langtidsforsok-lyft-efter-20-ar/) provides access to part of the available film, showing retrieval of the S2 parcel and the efforts required to locate the copper coupon. Public access to this footage is helpful and supports demonstrations of openness and transparency. The film also shows the difficulties and uncertainties associated with obtaining measurements. SKB has saved all of the film material, which runs to days of footage [16].

During the first QA review meeting [16] the transparency to stakeholders of SKB's retrieval plan for LOT was discussed, because some stakeholders have stated that the timing of retrieval was not publicised. SKB noted that the plan to retrieve the test parcels during 2019 was stated in its 2016 RD&D programme [30, §10.3.1], but agreed that the plans could have been presented in a more transparent way. According to current plans, SKB intends to retrieve the last LOT test parcel S3 in 2023.

Although SKB has not publicised the availability of samples from the LOT S2 and A3 parcels for independent study, the possibility of distributing samples for analysis is discussed in SKB's PMP [28, §4.7; 16]. There are too few copper coupons in the S2 and A3 parcels to share with others. However, bentonite samples are available for analysis on request provided the objectives of the study are sound and clear, the researcher in question has the competence to perform the study, and the results of the analysis are shared with SKB [16]. Indeed, organisations involved in SKB's Alternative Buffer Material (ABM) project had expressed interest in analysing samples from the S2 and A3 parcels, and a meeting with ABM organisations was planned for summer 2020 at the Äspö HRL [16]. However, due to Covid-19 restrictions on travel, the meeting had to be cancelled. The PMP notes also that it may be possible to distribute parts of the copper tube and attached bentonite for analysis on a case-by-case basis [28, §4.7]. All material from the retrieved S2 and A3 test parcels has been stored [16].

3.1.4. Project Structure and Specification

The goals of the project to recover and analyse the LOT S2 and A3 parcels are noted in the Project Charter [7, §1.2] and the PMP [28, §1.2]; two principal objectives relating to bentonite behaviour and copper corrosion are highlighted. First, the project will lead to increased knowledge of mineralogical changes in bentonite under repository conditions, providing results that can be used to increase the reliability of future safety analyses for the nuclear fuel repository. Second, SKB recognised that the recovery and analysis of the copper coupons and tubes from the S2 and A3 parcels will help to address questions about SKB's credibility associated with the long delay in the project, as discussed in Section 3.1.3.

The PMP sets out a strategy for achieving the project goals and objectives [28, §1.3]. Regarding copper corrosion, the strategy includes production of data on the corrosion depth of metallic copper to enable an assessment of how the copper has been affected after 20 years of heating under LOT conditions, and evaluation of whether the measured corrosion depth is consistent with results from previous experiments and with model calculations.

As described in the PMP, the project is divided into three work packages (WPs) covering retrieval and dismantling of the S2 and A3 parcels (WP1) [29], the copper corrosion analysis (WP2) [31] and the buffer analysis[1] (WP3). The WP plans include descriptions of the specific goals of the work package, the scope and cost of activities to be undertaken, organisational responsibilities, reporting, schedule, and risks.

WP1: parcel retrieval and dismantling

Details of the planned work to retrieve and dismantle the S2 and A3 test parcels are provided in WP1 [29], which includes preparation and implementation of activity plans to:

- Drill and extract the two test parcels and transport them to the bentonite laboratory.
- Divide and package samples of buffer material.
- Carry out initial analysis of the water content and density of the bentonite.

The data to be delivered to SICADA are identified [29, §4.2.6], and include:

- The daily log, which describes the different activities and what has been achieved.
- Photographs taken from all activities.
- Results from the initial analyses of buffer water content and density.

The work package is described as contributing to the overall project objectives by safely dismantling the test packages, delivering samples of buffer material, the copper coupons and the central copper tubes for analysis in the subsequent two work packages, and delivering a report of the work [29].

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[1] The buffer analysis plan WP3 has not been reviewed because this QA review project has focused on retrieval and analysis of the copper in the LOT S2 and A3 test parcels.

WP2: copper corrosion analysis

The work package for the copper corrosion analysis (WP2) [31, §2.2.2] sets out plans to:

- Quantify the average corrosion depth of the copper by mass loss and measurement of the thickness of the oxide layer.
- Determine the composition of copper corrosion products in terms of both chemical form and elemental composition.
- Analyse corrosion morphology, in cross-section as well as by analysis of larger areas.

The work package plan includes a description of SKB's expectations regarding corrosion of the copper coupons and tubes on the basis of known corrosion processes associated with residual oxygen and possibly sulphide from the groundwater [31, §3]. Expectations include average corrosion depths of about 1-10 μm, corrosion products in the form of Cu_2O and possibly the Cu (II) compound paratacamite, and any sulphur to be present on the copper surfaces as Cu_2S. The corrosion morphology and specifically the topography of the underlying copper surface was expected to be relatively evenly corroded (noting that pits and defects of about 1-10 μm are often present in the copper surface after manufacture, and significantly deeper defects may be present due to wear and mechanical impact). It was also noted that the hydrogen content may be slightly elevated in superficial corrosion products and superficially in the metal, but is not expected to be elevated within the body of the metal.

The WP2 plan [31, §4.2.1] includes activities to prepare copper coupons and tube samples for analysis, to undertake the analysis, to report results, and to store the data in SICADA.

The work package is described as contributing to the overall project objectives by contributing new data to the analysis of copper corrosion in field trials and supporting the corrosion modelling used in SKB's safety analysis of the nuclear fuel repository. The work package is also described as contributing results that can be used to increase the reliability of future safety analyses for the repository or the repository's design [31].

3.2. Measurement Methods, Techniques and Procedures

3.2.1. Activities under WP1: Parcel Retrieval and Dismantling

Activities at the Äspö HRL are covered by SKB's general rules for work at the Äspö HRL and for work underground [28, §10.2]. Activities at the Äspö HRL must also be undertaken in accordance with SKB's activity management procedure [32], which covers all work associated with planning, implementation and completion of the activity. More generally, SKB requires that operation and maintenance activities at the Äspö HRL are carried out according to an Äspö HRL maintenance instruction [33].

Activity plan for parcel extraction

The activity plan for extraction of the S2 and A3 test parcels covers the following steps [34, §1]:

1. Preparatory work in the tunnel.
2. Preparation of test packages (disconnection of heaters and instruments).
3. Disassembly of the control cabinet.
4. Preparations for drilling.
5. Drilling around the test packages.
6. Wire sawing under the test packages.
7. Lifting and transport of the test packages to the test hall.

The activity plan identifies approaches and techniques to constrain and/or mitigate risks, with a key risk during parcel retrieval being the need to avoid water contact with the bentonite, which can lead to the bentonite swelling and making certain analyses impossible [34, §2]. The activity plan acknowledges that the work requires close collaboration between the drilling contractor and activity managers, and specifies risk management requirements as follows:

- Drilling plans must be prepared that account for the geologist's data; if there is a risk of intersecting water-bearing cracks, these areas should be drilled as late as possible [34, §3.5].
- The drilling contractor must immediately notify the activity leader or co-ordinator if water is found [34, §4.6.3]. Slots are to be drilled 0.3 m under the package to provide a buffer volume for water if leakage occurs.
- If the measurement of the borehole shows that the hole has gone off course, the contractor's work preparation must show a proposal for a mitigating action or action plan [34, §4.6.2].
- When drilling is completed for the day, two water level alarms must be installed 0.3 m above the slot bottom and suction equipment must be in operation [34, §4.6.3].

The drilling activity plan [34, §3.5, §4.6.2, §5.1] also specifies safety and quality requirements. Requirements include the need to measure the distance between the crane and the roof, the need for electrical current for the electrical cabinets and backup generator connections, and the need for exhaust systems, water-level and fire alarms. Also, a minimum width of the slot created between the solid rock and the test parcel column and the diameter of the drilled pillar are specified. The activity plan [34, §5.2] also requires that the supplier develop a quality plan, which must describe how to check that the activities are carried out in accordance with the quality requirements defined in the plan.

During the first QA review meeting, SKB was asked whether any problems were encountered or deviations made from the activity plans for retrieval of the parcels [16]. SKB explained that the preferred way to dismantle the LOT packages is percussion drilling as it is a dry drilling method (core drilling requires the addition of some water for cooling). However, percussion drilling has commercially been replaced by wire sawing so percussion drilling tools are not readily available and old tools had to be used. It was found that there was some play in the re-used tools, which led to the holes not being perfectly straight, and thus edges of rock were left between the holes. These edges had to be removed using core drilling (as set out in the risk handling plan described in Section 3.1.2). The water used was managed appropriately and dry conditions were maintained around the parcel. There were no implications other than a time delay and increased costs; the equipment was updated for drilling of the second parcel.

Two minor incidents were reported during parcel extraction [16], one relating to a contractor's helmet lacking straps and one relating to an individual being unsure of how to operate the elevator. There was also an incident reported relating to a door not being locked after the parcels had been removed, but before the area was formally confirmed to be free from radioactive contamination, although all of the ^{60}Co tracer doped plugs had been removed with the parcels. The noted incidents were documented and reported according to procedures, and addressed directly when discovered [16]. The incidents did not affect the experiment or the results of the experiment.

Activity plan for dividing and packaging samples

The activity plan for dividing and packaging the LOT test parcels describes the coarse division of the test parcels following their transport to the Test Hall, sampling of the bentonite for the initial analyses of water content and density distribution, and material handling and packaging [35, §1]. The activity plan defines responsibilities of SKB and the supplier (Clay Technology AB) in terms of activities and materials and equipment to be provided [35, §3]. The division of the two test packages comprises several activities, including activities to identify and disassemble the two blocks that contain copper coupons (blocks 22 and 30) and to divide the copper tube and take samples of bentonite from the tube surface for analysis [35, §4.2.1].

The need to mitigate the potential for contamination of the bentonite by copper shavings when sawing the central copper tube is identified in the activity plan [35, §4.2.5]. Requirements for carefully locating the copper coupons in blocks 22 and 30 before division are also specified [35, §4.2.7], with a metal detector used to check the position of the copper coupons, and a margin of about half a block height in both directions added so that the copper coupons are not damaged. During the QA review meetings, SKB stated that the original LOT drawings were used to identify the coupon positions and the metal detector was used to verify the positions within a few centimetres, which minimised the risk of scratching the coupons [16; 17]. In addition, hand tools (a rubber hammer and a wooden wedge) were used to remove the surrounding bentonite clay and extract the coupons. The leader of the copper corrosion analysis work package (WP2) participated in order to oversee the safe extraction of the coupons. Coupon retrieval was successful and SKB noted that any scratches or damage would have been noticed in the gravimetrical analysis and/or in the microscopic examination [16].

The activity plan also covers the planned filming of the retrieval process, including requirements for the activity leader to call in film technicians at key points, such as for the work with metal detectors to identify the copper coupons, the sawing of the copper tubes, and on occasions when the copper tube is exposed [35, §4.2.1].

The activity plan requires that all retrieved bentonite is vacuum packed and stored, and all parts of the centre copper tube and instruments are packed and saved [35, §4.2.3]. The samples were directly placed in vacuum bags and transported to the laboratory, where they were immediately placed in a plastic tent purged with nitrogen gas [16]. SKB has confirmed that all material and samples from the S2 and A3 test parcels have been stored, and estimates that the total exposure of samples to dry air was less than one hour [16]. Rock pieces that arise from dismantling of the test parcels are handled as waste [35, §4.2.3].

Activity plan for initial bentonite analysis

The third activity plan for WP1 describes the initial bentonite sampling for determination of water ratio and density distribution, but the plan has not been considered in this review. However, it is noted that the XRF analysis to measure copper in samples of bentonite that were next to the copper tube was done according to an SKB method description [36]. SKB uses the Omnian method for XRF, as developed by the supplier of the equipment (Panalytical), and the supplier provided training, which formed the basis of the method description. The equipment has an internal calibration system [17].

3.2.2. Activities under WP2: Copper Corrosion Analysis

The WP2 plan [31, §4.2.1] sets out the different activities required for the copper corrosion analysis. The initial activity is described as involving work package preparations, including developing activity plans and arranging quotes for the corrosion analysis. The copper corrosion analyses are prescribed as follows:

- Prepare copper coupons: weighed copper coupons (four per package) are to be removed as far as possible embedded in bentonite and vacuum packed in a plastic bag so that the sample does not come into contact with the aluminium bag. These samples remain vacuum-packed until the external lab is ready to begin mass loss and other analyses:
    - SEM-EDS (scanning electron microscopy - energy-dispersive x-ray spectroscopy) of at least one sample per package, more if there are differences in surface morphology;
    - removal of residual bentonite;
    - pickling/removal of corrosion products and gravimetric analysis (mass loss) with photographs taken during this process; and
    - optical microscopy of the surface of at least one sample per package.
- Prepare copper tube samples: cut discs from the central copper tube, if possible with bentonite remaining, at four positions per package, probably a few centimetres thick, for analysis:
    - surface chemical analysis of the composition of the corrosion products (XRD, Raman) is to be performed after removal of the bentonite;
    - SEM-EDS on metallographic cross-sections, will be performed partly to measure oxide layers and partly to determine the elemental composition of the film; and
    - measurements of the elementary profile are to focus on H, as well as measurement of H in bulk material.
- Analyse the copper coupons:
    - SEM-EDS normal analysis and XRD (four per package);
    - determining the pickling method of pre-oxidised samples and reference samples;
    - mass loss (four per package); and
    - microscopy of surfaces (four per package).
- Analyse the copper tubes:
    - metallographic cross-sections (SEM-EDS) for measuring oxide layers and elemental composition (two positions with six cross-sections per position to give 12 cross-sections per package); and

- o hydrogen measurements (total content and surface profile), one disc per package, five measurements of total content, five measurements of surface profile.

Other activities in the WP2 plan involve presentation of results, final report preparation, and completion of the work package, including storage of data in SICADA.

Although listed as a delivery item for WP2 [31, Table 4.1], a detailed activity plan and method description for the copper analysis was not supplied by SKB for the QA review, because an external contractor was responsible for undertaking the analysis. During the first QA review meeting, SKB explained that, when seeking suppliers, the LOT project asked for offers including what should be measured [16]. This was done in an iterative way through discussions between SKB and the supplier. The resulting orders then refer to the offers regarding what should be done and delivered. No contracts or work orders were reviewed under this QA review, but it is assumed that the analyses listed in the WP plan formed the scope of the contract with the suppliers and this is evident from the work that has been reported [6].

RISE KIMAB AB and Swerim AB were contracted to undertake the copper corrosion analyses. These organisations were formed in 2018 following a division of Swerea KIMAB and are located at the same premises at Kista in Stockholm; they share laboratories and other facilities [16]. RISE KIMAB focuses on corrosion research and Swerim focuses on metal research. SKB [16] explained that the RISE KIMAB project manager coordinated the work performed by both RISE KIMAB and Swerim. RISE KIMAB uses documents similar to SKB's activity plans to describe in detail the work to be done and the methods to be used. SKB was involved in preparing these documents, but SKB does not approve them because they are handled according to the supplier's management system. SKB worked closely with RISE KIMAB to develop the plan for the copper corrosion analysis, because it was important that the analyses were done in the right order given that there were many different measurements to be made from only a few samples. It was also important to ensure that changes could be made to the plan as necessary in response to the ongoing findings of the analyses. For example, SKB [16] was involved in decisions regarding the steps to be taken in the pickling analysis for the copper coupons (see discussion in Section 4.2.1). In addition, if the supplier noticed a problem or suspected that something was wrong, they contacted SKB and the issue was discussed and how to proceed was decided.

SSM and SKB staff visited the RISE KIMAB and Swerim laboratories in November 2020 as part of the QA review project [19]. The RISE KIMAB Group Manager for the Infrastructure and Energy Unit explained that seven employees from RISE KIMAB and Swerim participated in the LOT S2 and A3 analyses. RISE KIMAB staff carried out visual inspections of the samples on arrival, as well as the gravimetry measurements and LOM (light optical microscopy), while Swerim staff performed the XRD, SEM-EDS, FIB (Focused Ion Beam)/TEM (transmission electron microscopy), and GDOES (glow-discharge optical emission spectroscopy) analysis. The samples for TEM analysis were prepared by Swerim's staff using FIB machining at KTH in Stockholm. It was emphasised that the types of analysis techniques performed are undertaken on a daily basis at the facilities and the staff have extensive experience in their application.

During the lab visit, it was noted that analytical methods have not been accredited at the laboratories since 2016 [19]. The old system used by KIMAB (the predecessor to Swerea KIMAB, as discussed in Section 3.3) had instrument descriptions and

Swerim has instrument descriptions, but no methodological descriptions of its own, because analytical methods generally have to be adapted to customer needs. However, standards are applied where possible and these standards generally include methodological descriptions. For example, standards SS-EN ISO 8407:2014 and SS-EN ISO 7407:2014 E.3.1 were applied; these describe methods for the pickling of samples and they were applied before carrying out the mass loss measurements in the LOT project. For the pickling method, preparatory tests were carried out on pre-oxidised samples prior to the LOT analyses in order to identify an appropriate procedure. However, the corrosion products on the pre-oxidised coupons had a different character and were very easily dissolved. Therefore, RISE and SKB chose to apply the standards referred to in SKB report TR-20-14 [6].

The issue of potential deviations in expected outcomes was discussed. It was explained that all at RISE KIMAB use the same management system within the company intranet [19]. The system is relatively new and is applied when dealing with systematic errors and minor deviations. If deviations of a more specific nature are identified, for example linked to a specific analytical instrument, this is first reported to the instrument manager. SKB [17] reported that there were no major deviations from the original plan for the corrosion analysis. As noted above, the pickling method was tested and adjusted, but this was not unexpected and is not regarded by SKB as a deviation.

Instrument calibration was discussed during the QA review. During the initial meeting with SKB and during the lab visit, it was explained that annual instrument calibration is carried out, but that rapid calibrations are performed by the operator before an experiment [14; 19]. For example, test weights are initially applied to quickly calibrate the scale for gravimetry measurements. During the second QA review meeting, it was explained that the following calibration activities were carried out [17]:

- Analytical balance (RISE KIMAB): Calibrated annually.
- XRD (Swerim): Calibration of the instrument is done on a regular basis by measuring a Corundum NIST standard sample. The peak position identification and offset calculation is then done using an EVA Diffraction program available from Bruker.
- SEM-EDS (Swerim): Typically, the calibrations for magnification and energy positions in the EDS spectra are checked every year during maintenance. Normally, there is no need to correct them since they vary very little over time. For EDS, it is obvious if the calibration is wrong, since all peak positions will be off, and an experienced operator would notice if anything is wrong.
- TEM (Swerim): Magnification calibration (i.e., the scale bar for imaging and also for diffraction in the TEM) may not be more accurate than +/-5%. For diffraction, calibration/normalisation against a known phase is typically performed before extracting the data.

During the lab visit, SSM was able to inspect the LOT A3 and S2 copper coupons and parts of the copper tubes that had been analysed. This included studying two coupons with a microscope.

3.3. Working with External Suppliers

SKB's Project Management and Quality Assurance system includes a procurement procedure '*SP5 Carry out procurement, supplier assessment and follow-up*'. As explained by SKB [14], the purpose of SP5 is to ensure that SKB has access to goods and services that meet all requirements at the right quality and price, and that follow-up, supplier assessment and experience feedback take place in accordance with current legislation and routines. These requirements are set out in SKB's purchasing instructions [37]. As defined in the instruction [37, §5.1.10], the qualification requirements for suppliers may relate to legal and financial status, competence and experience, resources, management system, and safety, quality and environmental management. Once an assignment has begun, any changes in personnel must be agreed with SKB [18].

As part of ensuring that suppliers have appropriate QA processes in place that are at least equivalent to SKB's, SKB [16; 18] prefers ISO-certified suppliers and performs a supplier evaluation before contracting a new supplier. Supplier evaluations are renewed regularly to assure that the company still meets SKB's expectations. The evaluation criteria are described in the purchase instruction [37] and focus on finances, QA and environmental impact. After delivery of contracted work, an evaluation of a new company is done by the SKB client according to a template provided by the procurement unit at SKB. For previously evaluated companies, additional evaluations should be made if called for based on new experience. These procedures are in place to ensure that past experience is taken into account when considering a company for additional assignments. Minor companies and sole proprietorships do not necessarily have ISO-certifications and they are typically chosen for their unique expertise [18]. The supplier evaluation is then based on CVs, level of education, publications in the scientific literature, etc.

All contracts signed with suppliers allow for SKB to undertake audits. Audits are done according to ISO 9001, with SKB reviewing the overall management system and supplier competence [16].

Four key suppliers have been used in the LOT S2 and A3 project [14]: Clay Technology AB; Uppländska Bergborrning AB; RISE KIMAB AB; and Swerim AB. SKB [16] noted that there are few suppliers available for these types of assignments and, in the case of the LOT A3 and S2 project, SKB considered it valuable to use suppliers that had the experience of being involved in previous recovery and analyses of LOT samples.

3.3.1. Clay Technology AB

Clay Technology AB was formed in 1988 and SKB has been using the company from its inception [18]. Clay Technology was formed from parts of SGAB (Sveriges Geologiska AB, a state-owned company formed in 1982) with which SKB had previously collaborated.

Within the LOT S2 and A3 project, Clay Technology has worked on three different work orders [14]:

1. KBP1019-19-1 Work package management – this involved development of the WP1 plan for retrieval and dismantling of the LOT S2 and A3 test parcels [29] and managing WP1.
2. KBP1019-19-2 Dismantling, sampling and reporting – this involved production of the three activity plans for drilling [34], sampling [35] and

analysis of the retrieved test parcels. The drilling itself was undertaken by Uppländska Bergborrning.

3. KBP1019-19-3 Hydromechanical analyses of LOT materials – this order includes development of a test plan and undertaking the hydromechanical analysis of the bentonite.

The WP1 manager and activity leader was a member of staff from Clay Technology who had been involved in the LOT experiment since its installation. While the Äspö HRL facility working instruction does state that competent external staff can be authorised to be work managers [38, §5.4], the purchasing instruction [37, §4.6] requires that contacts with tenderers by such staff are limited and only refer to technical issues; the external staff member is not allowed to independently make and communicate decisions that directly influence the choice of supplier.

The role and responsibilities of the work package manager are described further in the WP1 plan [29, §5.1]. This includes the requirement that internal resources should check that invoices linked to the work package are correct. SKB clarified that internal resources refers to SKB staff and not contractor staff, and emphasised that the latter are not permitted to check invoices [16]. Also, according to the WP1 plan, the work package manager prepares tender documents, evaluates quotes and writes technical specifications for the work orders [29, §14.1]. At the QA review meeting [16], SKB reiterated that, with respect to contracts, a consultant may not be the technical administrator or contact person and cannot be the deciding party when selecting a supplier. However, they may provide technical expertise when writing invitations to tender and assessing offers. For WP1, the consultant was involved with the SKB purchase department in compiling technical specifications in preparation for invitations to tender for the drilling work and when technically evaluating drilling companies who bid for the work, but this is not a business for which Clay Technology competes, so that there is no conflict with the purchase instruction.

Clay Technology AB works according to an operating system based on ISO 9001:2015 and was most recently certified on 29 October 2019 [14]. The business system includes management of the company, requirements for documentation, project management, administration and work quality. External and internal audits are carried out according to a set schedule.

SKB undertook a supplier evaluation of Clay Technology in 2017 [16]. SKB [14] explained that, as part of the business system, there is a lab handbook that describes the company's routines and activities in Clay Technology AB laboratories. The handbook describes requirements for staff, premises and equipment. Additional documents describe general routines such as those for calibration, laboratory work, handling of chemicals, a checklist for experiments, and a checklist for non-standard methods. Clay Technology has also documented specific method descriptions, including for cation exchange capacity (CEC), density, hydraulic conductivity, pressure, water content, and granule size distribution. Deviations from these method descriptions must be reported. In addition, Byggforskningsrådets (Building Research Council) geotechnical instructions or other described methods can be used, but their use must be reported.

3.3.2. Uppländska Bergborrning AB

Uppländska Bergborrning AB was contracted to drill and retrieve the LOT S2 and A3 test parcels under WP1. SKB [14] stated that Uppländska Bergborrning is ISO-certified according to ISO 9001:2015, Environment 14001:2015, and Working environment 45001:2018, and that the work was carried out according to:

- Work order 23078 for drilling and retrieval of LOT parcels S2 and A3.
- The drilling activity plan [34].
- Uppländska Bergborrning AB's environment and quality policy.

3.3.3. RISE KIMAB AB and Swerim AB

RISE KIMAB AB and Swerim AB were contracted by SKB to undertake the copper corrosion analyses. SKB [14] stated that RISE KIMAB and Swerim are ISO-certified according to ISO 9001:2015, and that the work was carried out according to:

- Order 22932 - Analysis of corrosion samples from field trials (LOT).
- Order 23867 - FIB and TEM on copper samples.

SKB work orders and supplier internal QA documents were not provided for this QA review.

SKB (and its predecessor the KBS-project) has collaborated with the Swedish Corrosion Research Institute (Korrosionsforskningsinstitutet) and the Swedish Institute for Metals Research (Institutet för Metallforskning) at the Royal Institute of Technology (KTH) since 1977 [18]. Corrosion studies were performed from 1977 and creep studies started in 1984. These two institutes then joined and formed KIMAB, later Swerea KIMAB, before the company was divided into RISE KIMAB and Swerim in 2018, as noted in Section 3.2.2.

The RISE Group consists of approximately 3,000 employees, while RISE KIMAB's corrosion department has about 40 employees and Swerim about 100. Swerim is owned mainly by Swedish industry, with the state as a minority shareholder, and RISE KIMAB is wholly state-owned. Approximately 50% of RISE KIMAB's corrosion activities consist of assignments from industry and about 50% is research. The corrosion analysis carried out for SKB forms only a small part of the department's overall corrosion activities

Until 2018, the corrosion and metal research departments had a common quality system, but after RISE KIMAB took over the corrosion department, the RISE Group quality system has been applied whilst Swerim applies a separate quality system [19]. The entire RISE Group holds ISO-9001 certification. Other parts of RISE, except the corrosion department, also hold ISO-17025 certification relating to general competence requirements for testing and calibration laboratories. RISE KIMAB and Swerim have yearly independent audits against ISO-9001 [16].

RISE KIMAB ensures suitably qualified and experienced staff undertake the research, with specific researchers responsible for certain instruments and a specific person who is responsible for the entire laboratory [19]. Most laboratories and instruments require the operator to hold a so-called 'driving licence' for a particular instrument; an employee receives the driving licence for a particular analytical instrument after passing a test based on theoretical and practical knowledge.

The RISE management system requires impartiality, and has procedures to support this [19]. Employees have a responsibility to alert their superiors to possible conflicts of interest. The RISE Group's management system includes whistle-blower functions and there is also a group-wide quality group.

SKB carried out an audit of Swerea KIMAB AB in 2017. A physical copy of the audit report was reviewed by SSM during the facility visit in November 2020 [19]. SKB's audit report concluded that the supplier can be considered to have a sufficiently good management system for the work it performs for SKB. Although the company has since been divided into RISE KIMAB AB and Swerim AB, this is not considered by SKB to challenge the audit report findings.

Regarding the independence of contractors from SKB, SKB [16] stated that all contractors are regarded as capable companies, with the major ones in the LOT project all being ISO certified. SKB does not affect the results obtained by the suppliers.

In some cases, contracting companies provide internal company reports to SKB that are then editorially post-processed within SKB (involving typesetting and printing of the report) to produce SKB TR- or R-reports [18]. However, this depends on the degree of involvement of SKB's experts in the actual work done; when SKB experts are involved in the work, they are normally involved at an early stage of experimental planning, analysis and reporting. In such cases the SKB expert is usually a co-author of the report. This is the case for the TR-20-14 copper corrosion analysis report [6], which was written collaboratively by SKB, RISE KIMAB and Swerim. The order of the authors listed on the report corresponds to the extent of their involvement in the work. Most of the bentonite analyses related to corrosion were performed by SKB staff using scientific equipment available at SKB, and SKB's experts were responsible for the report's conclusions [6].

During the first QA review meeting [16] there was some discussion regarding the extent to which SKB reviews and fully understands the analyses done by suppliers. Typically, SKB staff would aim to ensure that they have a thorough understanding of the analyses. However, the TEM and diffraction work done by Swerim and included in the TR-20-14 report [6] was noted as an example of analysis that SKB hadn't understood in detail; the conclusions of the work were incorporated into the report as provided by the experts at Swerim.

3.4. Data and Records Management

According to the Project Charter [7, §4.2.2], the project manager is responsible for handling information (documents and data) in accordance with the following SKB management system documents [14; 7, §4.2.2]:

- Principles for Information Management [39], which has been developed to implement the standard *SS-ISO 30300 Information and documentation - Management system for business information - Principles and terminology (ISO 30300:2011)*.
- Joint information management plan [40].
- Information management plan for department R (Research and Development) [41].
- Data management of primary data [42].
- Data delivery to, as well as data ordering from, GIS or SICADA [43].

The Project Charter [7, §4.2.2] and PMP [28, §12.1] require that all data generated during the project's implementation and which form the basis for the project's results, must be traceable and stored in SKB's databases.

For work carried out at the Äspö HRL, the activity tables in the WP1 plans list the separate activities, the deliverables/data they generate and how they are to be stored. In order to track work and submissions to the databases, SKB [16] stated that the relevant activity table is completed when the work is done, when data are delivered and, finally, when deliverables/data are stored and approved.

In addition, the drilling activity plan for WP1 sets out the requirements for data management, which include [34, §6]:

- Submitting the signed original Daily Log to the responsible activity leader immediately after completing the assignment in the field; the activity leader is responsible for ensuring that the delivered material is quality assured.
- Raw data must be submitted continuously immediately after the activity leader has quality-approved the data. The activity manager is responsible for receiving and checking the raw data from any external laboratories/suppliers.
- Processed and calculated data must be delivered so that it can be stored without change in SKB's database (SICADA or GIS). The material must be correctly linked back to previously submitted activities in terms of activity type, ID codes and times.
- Deadlines for delivery of documentation by the supplier are clearly specified in the activity plan.

The data management requirements of the activity plan for dividing and packaging samples include [35, §5]:

- Field data: Submitting the signed original Daily Log to the responsible activity leader immediately after completing the assignment in the field; the activity leader is responsible for ensuring that the delivered material is quality assured.
- Raw data: Delivery to SICADA in the Excel template of (1) protocols showing how each individual block is divided and how the pieces are marked; (2) protocols with results from the post-control of instruments.
- Other documentation: Storage in SKBdoc of reports, memos, photos, accounts that are not to be stored in SICADA or GIS, which includes photos and minutes from the work to divide the test package.
- SKB's activity leader checks that the agreed requirements for the specified deliveries are met, with any deficiencies to be rectified by the supplier.

The copper corrosion work package (WP2) plan defines delivery of data to SICADA as one of the deliverables [31, Table 4.1], but an activity plan setting out the data requirements in more detail was not produced because SKB [16] stated that the work required is defined in supplier work offers/orders. Work orders specify delivery of data to SICADA and this is checked by the WP leader [16]. For this review, SKB provided a copy of the SICADA data delivery form for recording the results of the gravimetric analysis of the copper corrosion coupons, and the results of a query to SICADA to show the data submitted. As part of this review, the data were spot-checked to confirm that they reflect the information that underpins the gravimetric analysis presented in SKB report TR-20-14 [6, Appendix D].

All of the analytical instruments at RISE KIMAB and Swerim are linked to laboratory computers by which all raw data are obtained and stored digitally, including photographs taken from exposed samples [19]. Data are then transferred to a project folder that collects all of the information associated with a specific project, and are saved to a server. Exposed samples are archived and stored for three years, or are sent back to the customer.

SKB [18] explained that data are sent (by e-mail or via ftp) by contractors and stored in SICADA; an SKB staff member responsible for the work approves/releases the data to SICADA. How the data are handled depends on the data type. For example, temperature values are stored as raw data, while other data are stored both as a raw data measurement files and calculated results in a template. For reports, calculated results are generally plotted or presented in tables. A general principle is that no data should be omitted. If it is obvious that, for example, a sensor is malfunctioning (e.g., showing unrealistic or unphysical values), it should be documented that data from this sensor have been removed and why.

The general approach at RISE KIMAB and Swerim to internal review of documentation and measurement results is for a senior researcher to review the material. Typically, RISE KIMAB and Swerim would provide a summary report of the results to the customer, but this project involved much closer collaboration with SKB and co-authorship of the SKB report; for this reason, no separate RISE KIMAB or Swerim reports have been produced for the LOT S2 and A3 project [19]. All employees from RISE KIMAB and Swerim who worked on the LOT project are co-authors of the SKB LOT corrosion analysis report SKB TR-20-14 [6]. RISE KIMAB and Swerim reported that all of the results obtained from the project have been delivered to SKB, and more results and images than would usually be provided have been included in the report appendices [19].

3.5. Report Review Requirements

The project documentation defines the public reports that are to be produced in accordance with the following high-level SKB procedures [14]:

- Report review [44].
- Checklist for review and quality control of public reports in department RS (Research and Post-closure Safety) [45].

Overall, these procedures require that a fact-checking review of public reports is undertaken, with the scope of the review defined in a review plan and use of appropriately qualified reviewers. For the report of the copper corrosion analysis (SKB TR-20-14 [6]), a review plan/instruction was produced [46], which suitably specifies the requirements and criteria for the review, and identifies the reviewers, their competence and their review scope. The resulting comments from the three reviewers have been appropriately recorded, along with the report author's response to the review comments [47; 48; 49].

4. Review of QA in SKB's Corrosion Analysis and its Interpretation

This section presents a QA review of SKB's copper corrosion analysis for the LOT S2 and A3 test parcels and SKB's interpretation of results. The QA review focuses on the reliability of the evidence that underpins SKB's conclusions about copper corrosion processes and corrosion rates and the extent to which the results support SKB's copper corrosion model. Findings are based on a review of SKB's report on the LOT S2 and A3 copper corrosion analysis (TR-20-14) [6] and discussions between SKB, SSM and GSL at the three project review meetings, although principally the second meeting that focused on the copper corrosion analysis and the third meeting that focused on the interpretation of results.

4.1. Pre-characterisation of Copper

There are certain aspects of the way the LOT project was set up over twenty years ago that inevitably mean that there are limitations in what can be learnt about copper corrosion under repository conditions from analyses of the LOT S2 and A3 test parcels. SKB does acknowledge these issues in the LOT S2 and A3 copper corrosion report (TR-20-14) [6, §1.5], stating in particular regarding the copper coupons that '*…their surface topography was not pre-characterised, meaning that the examination of corrosion morphology is inherently uncertain*.' Also, the report states that '*[t]he surface of the copper pipe was not characterised regarding surface topography or deposits before the start of the experiment, meaning that it is not strictly possible to conclude how the surface topography has changed during the experiment and how it has been affected by corrosion*.'

Why the copper surfaces were not characterised and what uncertainties this introduces to the corrosion analyses were discussed at the second QA review meeting [17]. SKB considered that the importance of pre-characterisation of the copper at the microscopic level was probably not realised at the time of initiation of the LOT experiment. In the 1990s, SKB's assessment of localised corrosion was mainly based on literature studies of pitting of copper tubes, archaeological artefacts, etc. SKB noted that the resulting uncertainty in the corrosion analysis is difficult to quantify [17].

4.2. Analysis of Copper Corrosion Coupons

4.2.1. Gravimetric and topographic analysis

Observations of the properties of reference coupons do not greatly reduce the uncertainties associated with the lack of pre-characterisation of the corrosion coupons. Two copper reference coupons had been stored under dry indoor conditions since the start of the LOT experiments and these were examined as part of the LOT S2 and A3 parcel retrieval and analyses, although with the additional complication that the reference coupons were also not pre-characterised. Differences in the condition of the reference coupons and corrosion coupons are highlighted by the results of topographic analysis (optical microscopy) [6, Tables 3-3 and 3-4] and mass loss analysis [6, Table 3-2], which are reproduced in Table 4-1:

- The reference coupons Ref L and Ref K were found to have 2 and 4 pits/cm^2 respectively - where a pit is counted if it is deeper than 6 µm (although no clearer characterisation criteria are provided for identification of a pit). Maximum pit depths were 22 and 25 µm before pickling to remove surface deposits. The two coupons were found to have 36 and 40 pits/cm^2 with maximum depths of 25 and 28 µm after pickling. Based on the mass loss measurements, the average corrosion depths of the reference coupons are 0.072 and 0.156 µm.
- The corrosion coupons were found to have 2 to 14 pits/cm^2 (with a pit depth of greater than 6 µm) and a maximum depth of 17 µm before pickling, and about 10 to 28 pits/cm^2 with a maximum depth of 57 µm after pickling, although the majority of observed pits are shallower than those found in the reference coupons. Coupons A3/L and S2/P show fewer and some shallower pits after pickling than before pickling, although they both show mass loss after pickling. Possibly, the analysis of the surface topography of the corrosion coupons is affected by the use of the corroded surface as the reference point, because the reference point changes after pickling [6, §3.3.3]. Based on the mass loss measurements, the average corrosion depths of the corrosion coupons range from 0.666 to 1.322 µm.

Thus, the reference coupons show a greater density of pits and generally deeper pits than the corrosion coupons after pickling, but the reference coupons have a much smaller mass loss than the corrosion coupons after pickling, with the average corrosion depth of the reference coupons being only about 10% of that of the corrosion coupons [6, §3.3.3]. SKB suggested that the topography observed on the reference coupons and the LOT corrosion coupons may be due to initially occurring defects of mechanical origin associated with coupon preparation and machining that have later been affected by different corrosion processes, although it is difficult to distinguish between pits formed by corrosion and those that are mechanically-induced, especially with the analysis method utilised [17].

Table 4-1. Deepest pits and density of pits deeper than 6 µm for coupon areas of 0.5 cm^2 before and after pickling [6, Tables 3-3 and 3-4].

Coupon	Before or after pickling	1	2	3	4	5	No. of pits	Pits /cm^2
Ref K	Before	25	19				2	4
	After	28	28	25	24	23	20	40
Ref L	Before	22	18	11			1	2
	After	25	23	23	22	22	18	36
A3/I	Before	8					1	2
	After	16	12	11	10	10	5	10
A3/J	Before	13	12	11	10	9	7	14
	After	39	22	13	10	10	11	22
A3/L	Before	17	9	8	8	7	6	12
	After	9	8	8	7	7	5	10
S2/M	Before	12	8	6			2	4
	After	57	26	15	13	12	14	28
S2/O	Before	16	14	13	13	12	7	14
	After	20	16	14	14	12	12	24
S2/P	Before	14	13	13	10	9	6	12
	After	39	14	12	8	8	5	10

For the reference coupons, the pickling process removed small amounts of superficial deposits associated with corrosion to reveal surfaces affected by a combination of mechanical wear and pitting corrosion during the twenty years of storage [6]. For the corrosion coupons, the pickling process removed bentonite deposits as well as corrosion products to reveal surfaces affected by mechanical defects from coupon preparation and corrosion under LOT conditions. The mass loss associated with removal of bentonite deposits and removal of corrosion products cannot be distinguished. One possible explanation for the corrosion coupons showing greater mass loss but fewer deep pits than the reference coupons is that uneven rather than pitting corrosion occurred across the entire surface of the coupons, resulting in few pits much deeper than 6 μm.

Reliance on pit depth, and pit area density unfortunately has led to a situation where such numerical analysis sheds little light in terms of quantifiable metrics. However, examination of the micrographs from individual pits is more definitive. Although the traceability of surfaces is not available and milled surfaces were used, possibly confounding the true depths of attack, these reviewers observe that there is some information to be gained by discarding numerical analysis and considering the corrosion products in pits - in some cases filling them - as a potential indication that some form of local corrosion may be taking place. The conditions for and possibility of local corrosion are discussed further in Section 4.7.4.

At the second QA review meeting [17] there was some discussion as to whether there would be any benefit in examining the surface of newly prepared copper samples in order to understand surface defects on manufacture. However, SKB considered that this may not be possible for the LOT experiment because the LOT coupons were prepared at Studsvik more than 20 years ago and the method of their preparation is not well documented.

There was also discussion about why the copper coupons had been prepared with a milled side and a polished side. SKB was unsure of the reasons for this, but considered that the polished side was probably intended for the evaluation of localised corrosion [17]. SKB used LOM to examine the milled side because it was judged to be potentially more reactive than the polished side and because it more closely resembles the rough KBS-3 canister surface.

In conclusion, the lack of pre-characterisation of the reference and corrosion coupons means that there is limited value in comparing the condition of the two types of coupon. It is conservative to assume that all mass loss from cleaning the corrosion coupons is due to corrosion, but little is learnt about how corrosion has affected the topography of the coupons.

4.2.2. Analysis of corrosion processes

The corrosion coupons were not in direct contact with the gas-phase during LOT, and SKB [6] suggests that corrosion may have been limited by diffusion of Cu^+ away from the copper surfaces and/or O_2 diffusion through the bentonite clay towards the coupons. However, there is no detailed analysis of the Cu^+ flux and the copper corrosion rate to support this argument. In addition, some corrosion occurred as a result of the slow diffusion of low concentrations of sulphide to the coupons through the compact bentonite clay from the groundwater.

The XRD analysis of the corrosion coupons prior to cleaning provided some insights into the corrosion processes that occurred. Certainly, evidence for Cu_2O is clear [6,

Figure 3-9], but the XRD evidence for Cu_2S claimed by SKB is not obvious. However, the SEM-EDS analysis shows evidence of sulphur, which SKB reasonably concludes may be in the form Cu_xS or $CaSO_4$. Observations of the relative atom percentages of Ca and S in the samples supported conclusions about the form of S present; in some cases, the S content was found to be about 6 to 9 atom percent and it was judged that this was too high to be explained by $CaSO_4$ precipitation alone, because the Ca was present at a smaller atom percent, indicating that a Cu_xS phase (i.e., a corrosion product) had formed. The SEM-EDS analysis also provided further evidence of the presence of Cu-O phase [6, Sections 3.3.1 and C1.3]. A Cu_xS phase was not detected at the surfaces of all of the coupons and, at the second QA review meeting [17], it was noted that sulphide would have been transported to the coupons from the groundwater, which may not have been evenly distributed in the test parcels. The SEM-EDS analysis also gives clear evidence of the presence of bentonite on the coupons, as indicated by observations of Si, Al, Fe, Mg, Ca, Na, and K.

At the second QA review meeting [17], the possibility of identifying a bentonite composition or 'fingerprint' that would enable the bentonite component to be removed from the EDS results to give clearer focus on the corrosion product composition was discussed. SKB argued that the quantification method is not accurate enough to do so when there are several phases in the same position. Also, the bentonite is not homogeneously distributed; there are different compositions in different positions. Thus, SKB considers that the EDS results should mainly be used in a qualitative way.

Cross-sections from two coupons were examined using SEM-EDS [6, Section 3.3.1]. The pits and surface defects were found to be less than 10 µm deep, which is consistent with the view that corrosion was non-uniform or uneven across the surface of the corrosion coupons, but did not result in deep pits. The EDS analysis indicates the presence of Cu_2S and Cu_2O. In some cases, there is a Cu_2O layer between the Cu and a thicker Cu_2S layer [6, Section C1.4.3] and there is a tendency for the level of sulphur to decrease towards the copper surface [6, §4.1].

The TEM and diffraction analysis on FIB-cut lamellae from the two coupons also provide evidence for Cu_2S and Cu_2O. However, the distances from the centre of the diffraction patterns to the reflection that indicate phase matches are not provided so the analysis cannot be fully appreciated. At the second QA review meeting [17], SKB noted that it is not possible to unambiguously determine which phase(s) exist in the samples because there are so many small particles close to each other and each diffraction pattern obtained usually contains information from several different phases.

Two minor issues were identified regarding sources of contamination in the analyses:

- The EDS results for the reference coupons indicate traces of Zn, which SKB has been unable to explain, and reference coupon Ref K shows contamination with Si, Al, Na, Zn, K, Cl and traces of other elements, the presence and cause of which is not discussed by SKB [6, Section C1.3.9]. At the second QA review meeting, SKB [17] reiterated that the source of Zn on the reference coupons is unclear and nothing in the handling of the samples at RISE KIMAB should have caused any contamination. SKB suggested that there may have been an unknown source of Zn in the laboratory at Clay Technology where the reference specimens were stored for over 20 years.

- The EDS results also indicate C contamination [6, Figure C-46]. SKB [17] stated that the contamination is typically a few monolayers (sometimes more) on the surface, covering the whole sample surface. In SEM and TEM, the electron beam attracts adsorbed carbon and hydrocarbons, so that carbon migrates to the beam where it is cracked and a layer of carbon builds up in the area that the beam scans. During an EDS analysis, the beam scans the same area for a long time, so a lot of carbon can build up. In SEM, the vacuum is lower, so even more carbon can end up on the sample from parts inside the chamber.

4.3. Analysis of Copper Tubes

4.3.1. Copper grade

SKB [6, §1.5] noted that the copper tubes used in the LOT experiment were made of a standard de-oxygenated copper (SS 5015-04) rather than oxygen-free phosphorous-doped copper (Cu-OFP) that is the reference material for the KBS-3 canister. During the second QA review meeting [17], there was some discussion about whether this difference in copper grade could have any impact on corrosion processes. SKB considered that the difference is probably of low significance, noting that the oxygen content of de-oxygenated copper was likely to be too low for the copper to be susceptible to hydrogen embrittlement involving hydrogen reactions with internally dispersed Cu_2O.

4.3.2. Selection of tube samples

The intention of examining 'interesting parts' of the copper tubes was noted in the original planning for LOT [1, §4.2.6]. SKB selected sections of the copper tubes from the LOT A3 and S2 parcels for corrosion analyses, but they did not include the parts of the tubes that were exposed to the highest temperatures. The selection of tube samples was discussed at the third QA review meeting [18]. SKB argued that the maximum temperatures experienced by the tube samples that were examined metallographically were 70 to 80°C for parcel A3, which is close to the peak temperature of 95°C that any copper canister in the KBS-3 repository would be expected to experience. Moreover, the maximum temperature in parcel A3 was 120°C, which is higher than is relevant to the KBS-3 concept. SKB also noted that the particular tube sections in blocks 21-23 were selected for practical reasons; that is, they were in the same bentonite blocks as the corrosion coupons [17, 18]. Note that detailed analysis of corrosion of the tube in the LOT A2 parcel had not been attempted [4, 10], so that analysis of the tubes from the LOT A3 and S2 parcels represents a beneficial progression. Also, the copper concentration profiles in bentonite samples that were next to the hottest parts of the copper tubes were examined, as discussed in Section 4.4.

SKB selected representative type areas of the tube samples for SEM-EDS analysis [6, §2.3]. SKB elaborated on the rationale for sample selection at the second QA review meeting [17]. 'Type-areas' were chosen based on their visual appearance: light (Cu coloured); dark/black; and with grey deposits that could be bentonite and/or gypsum. Several cross-sections were examined for each area using SEM. SKB noted that the darkest parts are not necessarily the most corroded, because corrosion products may in some cases adhere to the bentonite surface in contact with the copper tube.

At the third QA review meeting, SKB commented that tube material from the LOT S2 and A3 parcels has been stored with the potential for possible further metallographic examination [18]. The tube that was exposed to the highest temperature could contain the deepest pits, and analysis of samples from different parts of the tube would facilitate a better understanding of thermal dependence of corrosion and the structure of Cu_xS layers. However, SKB considers it unlikely that further examination of tube samples would change any conclusions regarding corrosion under the initially oxidising conditions of the repository [18].

4.3.3. Topographic analysis and corrosion processes

SEM analysis of the copper tube surfaces and cross-sections identified pits up to 25 μm deep, but the pits were frequent and generally wide and shallow [6, §3.4.1, Appendix F]. EDS analysis identified Cu_2O, bentonite (indicated for example by high Si content) and low levels of S, but Cu-S phases could not be distinguished [6, Appendix G]. Some enhanced level of S occurred at locations with enhanced levels of Ca, indicating that the deposit was $CaSO_4$ from the bentonite.

A set of SEM and EDS images and data are provided in the appendices. However, it is difficult to fully understand the relationship between the different SEM cross-section figures for the same sample [6, Appendix F]. In some cases, the same area is being shown at different magnifications, but in most cases the location on the tube of the cross-section shown for a sample is not apparent. It is also not explained how a 'defect' has been identified and counted [6, Table F-1]. For example, sample A3-2 is shown as having just two defects, but it is not clear from the images of sample A3-2 what is considered to be a defect. There is no detailed discussion of the figures.

4.3.4. Analysis of inner surface of tubes

SEM-EDS analysis of inner tube surfaces revealed some corrosion as indicated by Cu-O and some surface contamination, which perhaps is bentonite (Mg, Al, Si, S, Cl, K, Ca as well as Cu and O) [6, Figure G-20]. Pits of a few μm depth were detected [6, §3.1], but any corrosion processes that occurred over the 20-year period over which the inner surface was exposed to air and humidity are not discussed in detail, presumably because the corrosion did not occur under repository-like conditions.

4.3.5. Analysis of hydrogen in tube samples

The hydrogen content of copper tube samples was measured using a melt extraction process, although the reason for the analysis is not explained [6, §3.4.2.1]. SKB confirmed at the second QA review meeting that the purpose was to investigate whether there was any uptake of hydrogen in the copper, which could lead to embrittlement effects [17]. The analysis found hydrogen to be present only at or near to the surface of the copper, which is where corrosion products and bentonite deposits are present; there is no detected enhancement of hydrogen in the copper.

At the third QA review meeting, SKB argued that the only process that could lead to hydrogen uptake by copper under LOT conditions is corrosion by sulphide, but the extent of such corrosion in LOT was very limited [18]. As evidence of the reliability of the measurement technique, SKB [18] referred to the results of another study to measure hydrogen in uncorroded copper specimens by similar methods, which gave similar results, with the main hydrogen content being situated near the surfaces of the samples [50].

If oxygen was depleted quickly by bacteria, then some other argument for anoxic corrosion before sulphide arrival would be of interest. A previous review of SKB's corrosion analysis concluded that any copper corrosion by sulphide attack would far exceed the corrosion depths of penetration that have been estimated could occur by anoxic corrosion in pure water in saturated bentonite [13]. Even if that were the case, SKB [6, §3.4.2.1] does not discuss what the hydrogen concentration 'should' be and what absorbed hydrogen profile 'would then' exist in copper, nor whether it would be detectable, in the event of any anoxic corrosion by water and hydronium ion reduction or during sulphide attack in LOT.

4.4. Analysis of Copper Concentrations in Bentonite

4.4.1. Corrosion depth

SKB [6, §3.5] estimated the average corrosion depth of the copper tubes by analysing the copper concentration profiles in the bentonite close to the copper surface using XRF. For this analysis, bentonite samples next to the hottest parts of the copper tubes were examined (e.g., samples from LOT S2 and A3 blocks 9 and 11).

The measurement method was tested on clay that had been in contact with the copper coupons, for which the extent of corrosion could be confirmed by standardised gravimetric methods. This enabled a suitable bentonite sample interval to be determined for the analysis (5 mm); an interval of 10 mm was found to be too large because the copper concentration was too small to be measured reliably [6, §3.5.1]. SKB accounted for uncertainties in the analysis and concluded that the estimated average corrosion depth of 0.5 to 1.2 μm is consistent with the results of the gravimetric analysis (0.7 to 1.3 μm, as discussed in Section 4.2.1).

SKB [6, §3.5.2] used the method to determine copper concentration profiles in the bentonite near the copper tubes. Six samples of thickness 0 to 2 mm, 2 to 10 mm, 10 to 20 mm, 20 to 50 mm, and 70 to 100 mm were cut. Given that the analysis for the copper coupons found that the sample interval should be 5 mm in order to give reliable results (although the copper was at cooler temperatures where corrosion would be less than for the tubes), it is not clear why larger and increasing intervals were used for the bentonite samples after the first interval. In effect, any copper concentrations in the samples 10 mm or more from the copper surface would most likely have been too small to be measurable, especially for the samples that experienced the coolest temperatures. However, in some cases, copper presence in samples in the 10 to 20 mm range are detected. This does limit the number of data points that could be used to determine copper concentration profiles.

Nevertheless, it is clear that the copper concentrations are highest in the bentonite next to the copper tube samples that experienced the highest temperatures [6,

Figure 3-33 and Figure 3-34]. SKB interpreted this finding as showing how the corrosion reaction rate increases as a function of temperature; corrosion of copper in the hotter parts consumed more of the oxygen in the LOT parcels, potentially also affected by the hotter parts being desaturated and exposed to air for longer. Also, the discretisation of the bentonite samples is sufficiently fine for the mass of copper oxide that has diffused into the bentonite and the average corrosion depth to be estimated (0.2 to 13.8 µm for LOT A3 and 0.2 to 4.8 µm for LOT S2), which again reflects the temperature dependence of the copper corrosion reaction.

4.4.2. Effects of corrosion products on tube surfaces

SKB [6, §3.5.2.3] did not take account of the corrosion compounds adhering to the surface of the copper tubes when estimating average corrosion depths based on the mass of copper found in bentonite. This issue was discussed at the second QA review meeting [17] and SKB emphasised that the adherent layer of corrosion products was very thin so that it was considered unnecessary to apply a correction factor to the corrosion depth to account for this. The topic was discussed at greater length at the third QA review meeting [18], where SKB reported that the oxide films on both the tube samples and coupons were typically around 1 µm. This finding is consistent across samples, despite the temperature differences and the fact that both lighter and darker areas were examined for each test parcel.

Although the thickness of the corrosion product layer on the copper tubes is not discussed in detail in the SKB corrosion report, the SEM-EDS analysis of one of the copper coupons revealed a 360-nm-thick corrosion product layer [6, §3.3.1], which corresponds to 250 nm of corrosion assuming Cu_2O at a typical density [6, §3.5.1.3]. The extent of corrosion of the copper coupons based on measurements of mass loss into the bentonite was estimated to be greater at 0.5 to 1.2 µm. As noted in Section 4.4.1, the amount of corrosion of the copper tubes based on measurements of mass loss into the bentonite was estimated to be 0.2 to 13.8 µm. Thus, by comparison, the amount of corrosion associated with the corrosion product layer on the copper tubes is less than that associated with mass loss into the bentonite, especially for the parts of the tubes that were exposed to the highest temperatures.

SKB also noted that the gravimetric analysis of the corrosion coupons effectively takes account of the Cu in the adherent corrosion products, and, as discussed in Section 4.4.1, there was good agreement between the results of the bentonite analysis (0.5 to 1.2 µm corrosion depth) and the gravimetric analysis (0.7 to 1.3 µm corrosion depth) for the copper coupons, showing that the effects of the thin film of corrosion products are small [18]. SKB argued that situations in which a multiplication factor (up to a value of 19) has been found to be necessary to match gravimetric results involve experiments where there is a 50:50 sand-bentonite mixture and a larger initial O_2 inventory than in LOT. For experiments similar to LOT, the multiplication factor has been found to be close to 1.

4.4.3. Corrosion products

EDS mapping [6, §3.5.1.4] for the bentonite next to the copper coupons showed that the corrosion product is most likely to be a copper sulphide, and spot analysis indicated the possible presence of Cu_2S. However, XRD analysis [6, §3.5.1.5] could not identify the presence of Cu_2S, or any other identifiable Cu-S phase. XRF analysis for the bentonite next to the copper tubes [6, §3.5.2.2] identified copper and sulphur in the bentonite samples; through XRD analysis, SKB concluded that sulphur is associated with gypsum enrichment [6, §3.5.2.4], presumably rather than any Cu-S phase. SKB noted that the steep copper profile in the bentonite clay may be indicative of Cu^{2+} being adsorbed [18]. Ion exchange between copper and Na and Ca in the bentonite affects the form of copper in the bentonite.

4.5. Analysis of Corrosion on the Copper Bottom Plates

During the third QA review meeting [18] there was some discussion of the differences in the corrosion of copper bottom plates next to the sand and corrosion of the copper tubes next to bentonite. SKB noted that this has not been investigated in detail, because sand is not a material that will be used in the KBS-3 repository.

The corrosion analysis report does note that blue-green deposits formed under the copper bottom plates in both test parcels during LOT [6, §3.1]. A sample of these deposits from the A3 tube was analysed using SEM-EDS and powder XRD and various compounds of copper were found including $Cu_2(OH)_3Cl$ (a Cu^{2+} corrosion product) and possibly Cu-S compounds [6, Appendix I]. SKB argued that the adherence of $Cu_2(OH)_3Cl$ (paratacamite) to the bottom plate suggests exposure to aerobic conditions for a relatively long period of time, with the paratacamite remaining kinetically stable under subsequent reducing conditions [18]. SKB further argued that the presence of sand rather than bentonite at this part of the copper surface meant that there was no potential for ion exchange between Cu^{2+} and Na and Ca [6, §4.2.2]. However, there is little information available on how long it would have taken the volume of sand around the bottom plate to saturate and oxygen to be consumed after the initial four-month period prior to water injection, which occurred close to the bottom plate.

4.6. Analysis of the Effects of Microbes

Two microbial processes are of potential significance to copper corrosion under repository conditions: those that involve reduction of sulphate to sulphide and those that involve reduction of oxygen to water [51, §2.3.1]. The generation of sulphide is a concern because of its corrosive effect on copper, but oxygen reduction is beneficial in that it reduces the potential for oxygen-induced copper corrosion.

With regard to sulphate reduction by microbes, the copper corrosion analysis report [6] indicates that two of the coupons in LOT parcel S2 were immersed in a bacterial growth medium before being installed in the test parcel, although this is not mentioned in the original planning report [1]. Information about this process has been lost, but SKB [6, §2.1] considers that the growth medium would most likely have contained sulphate reducing bacteria (SRB). During the second QA review meeting [17] SKB elaborated that, by mistake, data concerning the bacteria placed on the copper coupons were not stored in SICADA. Based on observations of the surfaces of the copper coupons after extraction, SKB [6, §4.1] concluded that immersion in a bacterial solution had no discernible impact on corrosion.

With regard to oxygen reduction by microbes, the LOT project originally had an objective to gather information on the survival, activity and migration of bacteria in the buffer, as discussed in SKB report IPR-99-01 [1]. This was to involve analysing microbial populations in groundwater before emplacement of the LOT parcels and at the end of the experiment, and to examine bentonite samples for microbial populations. However, SKB confirmed at the second QA review meeting that no specific measurements of microbial populations in groundwater were made for the LOT experiment [17] (as discussed in Section 3.1.1). At the third QA review meeting [18], SKB emphasised that the conditions in the LOT experiment were not favourable for microbes due to the initially unsaturated (low water content) and cold conditions, and later high temperature and high bentonite density, so the oxygen consumption from bacteria would have been low; the O_2 would have been consumed by corrosion before any microbial consumption could begin. At the second meeting, there was some discussion as to whether bacteria in the sand beneath the test parcels could have consumed O_2, but this was not studied [17]. Instead, SKB considered that sufficient data on microbes would be available from other experiments at the Äspö HRL, although there are large variations in the data. Therefore, no clear conclusions can be drawn about the influence of microbial populations on oxygen depletion in the LOT A3 and S2 parcel analysis and the extent to which this might have affected copper corrosion processes.

4.7. Evolution of Conditions and Corrosion Reactions in LOT

Section 4 of SKB's corrosion analysis report [6] provides a summary discussion of copper corrosion during the LOT A3 and S2 parcel tests based on the analyses of corrosion products, the depth of corrosion on the copper coupons and tubes, and corrosion morphology. SKB's understanding of how conditions evolved and corrosion occurred in different regions of the LOT parcels was further summarised at the third QA review meeting [18].

4.7.1. Corrosion of the copper coupons

The corrosion coupons were embedded in bentonite with no air gaps, but SKB considers that most of the corrosion of the coupons occurred during the initially oxygenated conditions [6, §4.1; 18]. Within less than a year of the heaters being switched on, the coupons were at temperatures of 25 to 55°C depending on location and parcel [6, §4.2.1], and the system is considered to have become saturated after no more than a few years (see Section 2). There is evidence of Cu_2O on the copper surfaces, and SKB suggests that corrosion may have been limited by diffusion of Cu^+ away from the copper surfaces and/or O_2 diffusion through the bentonite clay towards the coupons. However, there is no detailed analysis of the Cu^+ flux and the copper corrosion rate to support this argument. Long-term corrosion would have been controlled by the slow diffusion of low concentrations of sulphide from the groundwater through the bentonite clay to the copper surface.

As noted in Section 4.2.1, the total average corrosion depth of the coupons was estimated to be 0.7 to 1.3 μm. Differences in corrosion depths observed in copper coupons from the 1 year LOT parcel A0 and S1 tests (3.7 to 4.8 μm), the 6 year LOT parcel A2 test (1.5 to 2.5 μm) and the 5 year Äspö ABM test (2.3 to 5.0 μm) [52] - a field test similar to LOT - were discussed at the third QA review meeting [18]. SKB considered that the differences in the depths of corrosion in these experiments were minor, and factors other than time and temperature are likely to have controlled corrosion (e.g., spatial variations in saturation rates and O_2 transport).

SKB [6] also made comparisons with the results of the large-scale Febex experiment in Switzerland, where copper coupons embedded in bentonite clay were heated to about 100 °C for nearly 18 years. The mass loss of one coupon corresponded to an average corrosion depth of about 9 μm and the deepest pit was about 100 μm. These values are larger than the corresponding values for the coupons in LOT, and SKB [18] argued that this may be due to a combination of higher temperature, a larger clay volume (with more O_2) and an extended oxic period in FEBEX compared to LOT, possibly with oxygen leakage from a tunnel in the FEBEX experiment.

4.7.2. Corrosion of the copper tubes

SKB [6, §4.1] concluded that O_2 was the main oxidant causing the corrosion of the copper tubes as well as the coupons in LOT. The O_2 was supplied to the copper tubes by transport within and between air-filled gaps, which was more rapid than O_2 diffusion through bentonite. The Ti-tubes used to speed up the water saturation process were open to the Äspö tunnel for about four months after installing the test parcels and before water injection began beneath the bottom plate, so it is possible that some of the corrosion on the copper tubes occurred during this initial period in an aerated and humid environment [6, §4.2.2]. Some of the air initially present in the gaps or in the sand porosity may have remained after the initial four-month period and some of the O_2 may have dissolved in the groundwater injected to fill the voids [6, §4.2.2].

At the third QA review meeting, SKB noted that reaching full saturation and swelling pressure of the bentonite clay took several years after water injection started [18], as discussed in Section 2. SKB [6, §4.1] argued that corrosion under aerobic conditions probably ceased many years before retrieval of the LOT parcels due to O_2 consumption by corrosion and other chemical and/or microbial processes. Subsequently, aqueous Cu^{2+} may have prevailed as an intermediate oxidant on a much longer timescale to react with metallic copper to form Cu^{+} [18]. The corrosion rate was controlled by temperature-dependent corrosion reactions as shown in Figure 11 [6]. If this corrosion largely occurred under aerobic conditions, it requires that O_2 was still present for a period after the heaters had been switched on.

SKB suggested that the slightly deeper corrosion (13.8 μm) on the warmest parts of tube A3 than seen on the previously examined LOT tube A2 (9.6 μm) (recovered after 6 years) could have been due to the longer period of exposure of tube A3 to Cu^{2+}, although differences may otherwise be due to differences in the amounts of O_2 in the two parcels [18]. The only other oxidant available in the groundwater environment is sulphide, for which transport limitations control the long-term corrosion rate. Although sulphur was detected on the tube samples and in the bentonite, the Cu-S phase associated with the corrosion process is not clear [6, §4.1].

At the second QA review meeting [17], there was some discussion as to whether the axial thermal gradient in the experiment could have further affected the corrosion depths and rates. That is, the more rapid consumption of oxygen by corrosion in the warmer parts of the tube could draw oxygen from the cooler parts to the warmer parts, thereby enhancing corrosion in the warmer parts and reducing it in the cooler parts. It was noted that such an axial thermal gradient would not be present in the repository, where the temperature increase is caused by more uniformly emitted spent fuel residual heat rather than an electric heater that provides a more localised heat source.

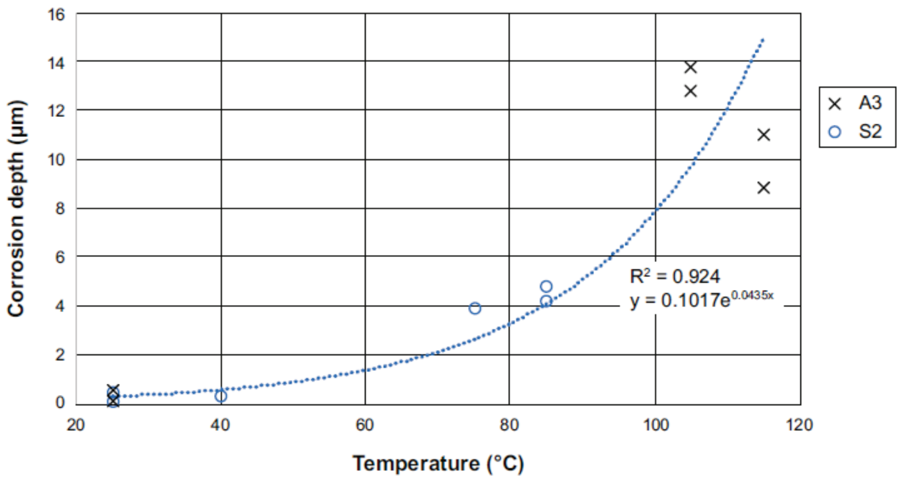

Figure 11 Corrosion depth as a function of temperature for the S2 and A3 copper pipe samples [6, Figure 4-1].

SKB estimated the mass of O_2 in the bentonite and sand pore volumes, in the gaps between the rock and bentonite blocks, and in the gaps between the bentonite and the copper tube. This O_2 mass was found to correspond to an average uniform corrosion depth of 13.2 µm for the tube surface. Taking account of the temperature distribution along the tubes, SKB estimated the average corrosion depth to be about 1.5 µm for tube S2 and about 5.1 µm for tube A3 [6, §4.2.2], which is not inconsistent with the corrosion determined through the XRF analysis.

SKB [18] also noted that the view that most corrosion of copper under disposal conditions occurs rapidly under initial aerobic conditions is supported by the results of Canadian studies [53], although the Canadian experiments did not have air-filled gaps next to the copper at the start of the experiment and so may be more reflective of the condition for the LOT coupons.

At the third QA review meeting [18], the possibility of leakage of O_2 into the test parcels via the cables around the parcels was discussed. SKB stated that such leakage cannot be ruled out. This could lead to a slow rate of corrosion limited by the supply of O_2, although other reactions could also consume the O_2.

At the second QA review meeting [17], there was some discussion as to why measurements of the corrosion potential of the copper (as proposed in the original planning report [1, §4.2.6]) or redox potential were not attempted during LOT. Monitoring of redox conditions would have reduced uncertainty in the time of transition from aerobic to anaerobic conditions. SKB noted that in general, the main focus of the LOT series was not copper corrosion. Also, limited technologies were available to measure redox potential in compacted bentonite in the 1990s, especially in a field test where it is difficult to install and maintain a reference electrode on long time-scales.

The uncertainty in the time taken for the gaps around the tubes to fill with water does mean that alternative interpretations of system evolution and oxygen availability for corrosion could be made. For example, if the gaps filled with water before any significant temperature increase occurred and there was limited O_2 dissolution in the groundwater, then it could be argued that a temperature-dependent anaerobic process was responsible for corrosion. Conversely, SKB [18] commented that the gap between the copper and bentonite clay may have remained open for a longer time where the temperature is highest because conditions are dryer and resaturation is slower. SKB concluded that this, as well as the temperature-dependent corrosion rate, could have influenced the distribution of corrosion over the copper tube surfaces in LOT.

At the third QA review meeting [18], SKB argued that the corrosion of copper in pure, O_2-free water is negligible in the LOT context (where O_2 is initially present and the groundwater is not pure water). If water does corrode copper under anoxic conditions, then hydrogen produced as a cathodic half-cell reaction product would be detected after it absorbed into the copper. As discussed in Section 4.3.5, SKB's hydrogen measurements showed no detectable enhancement of hydrogen in the copper, although there is no discussion of what the hydrogen concentration in copper associated with this process would be or whether it would be less than detectable.

4.7.3. Oxygen consumption

A key area of uncertainty relates to the role of competing mechanisms for oxygen consumption during LOT. Potential mechanisms for consuming oxygen other than copper corrosion were discussed at the third QA review meeting [18]:

- **Microbial consumption**. Although no specific measurements of microbial populations in groundwater were made for the LOT experiment, as discussed in Section 4.6, SKB considered that the conditions in the LOT experiment were not favourable for microbes so that any oxygen consumption by bacteria would have been minor.
- **Reactions in bentonite**. SKB cited studies (on bentonite pellets) that showed how O_2 consumption by inorganic reactions in clay is slow under unsaturated conditions at low temperatures [54].
- **Pyrite oxidation**. SKB commented that oxidation of pyrite to form sulphate occurs at 55°C, but the occurrence of pyrite oxidation would be difficult to confirm. It was noted that the sulphate content or the reduction in pyrite could be measured, but this would be difficult because the pyrite content is initially low and the bentonite also includes gypsum, from which sulphate may move towards the heater.
- **Fe(II) oxidation**. SKB argued that the Fe(II) concentration of the groundwater that is supplied to LOT is low (around 70 ppb) and the exchange of groundwater with the LOT-packages is limited, so that oxidation of Fe(II) to Fe(III) can be assumed to have had a negligible effect on oxygen consumption.

The lack of direct evidence of the influence of microbial populations on oxygen depletion in the LOT A3 and S2 parcel tests means that alternative interpretations of how conditions transitioned to anaerobic are not readily dismissed, which leaves uncertainty in understanding the extent to which copper corrosion occurred under aerobic conditions.

4.7.4. Corrosion products

SKB claims that observations of the composition of corrosion products support the view that corrosion of the copper tubes and coupons occurred predominantly under aerobic conditions in the early stages of LOT. The main solid corrosion product adherent to the copper surface is Cu_2O. SKB [18] argued that, based on thermodynamic data, the Cu^+ corrosion product Cu_2O is formed under a positive redox potential, and higher potentials are required to form Cu^{2+} solid phases such as $Cu_2(OH)_3Cl$ (paratacamite) and $Cu_2(OH)_2CO_3$ (malachite). Further, SKB [18] argued that, under LOT experiment conditions, when all O_2 is consumed the redox potential would most likely be determined by iron compounds in groundwater, and redox potentials are too negative for the formation of Cu_2O, $Cu_2(OH)_3Cl$ or $Cu_2(OH)_2CO_3$.

The observation of Cu_2O and $Cu_2(OH)_3Cl$ (on the bottom plates) in LOT is consistent with thermodynamic data for the initial conditions when O_2 is present (high Eh). The paratacamite has not dissolved for the duration of LOT. Where bentonite is present, the Cu^{2+} tends to be absorbed in the clay by ion exchange between copper and Na and Ca rather than forming a solid precipitate.

The solid compounds once formed may remain kinetically stable or may dissolve only slowly after reducing conditions are established. SKB [18] noted that there are signs of blue-green Cu^{2+} corrosion products on the coupons in the FEBEX *in situ* experiment, which may be due to the longer period of oxic conditions and possibly oxygen leakage from the FEBEX tunnel. A report on corrosion processes in the FEBEX experiment [55] found that the estimated corrosion depths on the steel components were 10 to 20 times larger than would be expected based on the amount of O_2 thought to be available, suggesting that there was an external O_2 source contributing to steel corrosion.

Anaerobic corrosion may have occurred as a result of diffusion of sulphide to the copper surfaces to form Cu_2S. Accepted tenets include the notion that Cu_2S is not protective (i.e., it is not akin to a passive film) owing to, for example, its porosity, and it does not provide a basis for local sites of attack that corrode much faster. Also, fast attack at selected sites under anoxic environments such as to produce a 'pit factor' is incompatible with slow sulphide production and mass transport limitations both inside and outside the pit. In effect, pit growth would be 'cathodically controlled' by the slow mass transport of S^{2-} (HS^-) and subsequent coupled rate of water or proton reduction at the nearby cathode area.

In a pit model that accounts for faster anodic dissolution at discrete sites, the cathodic site must be capable of supporting an anode site, and there must be a physical basis for local attack, such as a weak area in the Cu-S film with greater porosity or some other kind of defective property that enables anodic dissolution. There would also have to be a separation of anode and cathode, which seems improbable since the pit site itself cannot support faster transport of HS^- or S^{2-}.

In spite of these arguments, local sites of shallow pitting are observed in cross sections [6, Section 3] and they appear to contain more corrosion products than the surfaces that experienced more uniform corrosion, which contain relatively uniform micrometer-thick scale films. It is unclear if conditions are suitable for compact non-porous Cu_2S formation, which might occur at some condition involving a combination of temperature, sulphate and Cl^- [13].

Evidence of corrosion products in pits may be considered as evidence that some form of local corrosion may be taking place. This defies the notion of S mass transport as S_2^- or HS^- as controlling copper corrosion, which leaves no room for fast anodes, as discussed above. However, sulphate induced pits in copper and Cu^{2+} reduction as a cathodic reaction could support pit dissolution at high rates, albeit temporarily. Depletion of the finite amounts of Cu^{2+} might stifle pits because the high rates cannot be sustained over long times once Cu^{2+} is depleted.

If corrosion was taken to be entirely uniform and also occurred in a region with a surface irregularity due to mechanical effects (not due to pitting), then the corrosion product would be no thicker than that seen on the outer surfaces of the copper, and maybe even less thick inside due to mass transport limitations. The situation is complicated by the fact that the ‘indications’ of shallow pitting are no deeper than other typical non-corroding surface indications seen in copper with typical commercial surface preparations, such as milled surfaces. It is therefore hard to draw conclusions. A plot of mean as well as 90 and 99 percentile pit depths or shape factors as a function of temperature might be informative if a trend is obtained. If a trend is not obtained, the evidence for mechanical indications would be strengthened, because the pits would be constant with time, produce the same corrosion products, and would not be affected by exposure temperature. In other words, there would be no correlation.

4.7.5. Groundwater composition

SKB observed that the formation water supplied to the boreholes became more saline and alkaline over the duration of LOT [6, §1.3]. The reasons for these changes and their potential impacts on the experiment were discussed at the second QA review meeting [17]. SKB commented that ‘upconing’, where more saline, deeper water rises towards the tunnel system, is frequently observed at Äspö and is the most likely explanation for the change in groundwater composition. SKB considered that this would have only a marginal effect on bentonite properties and noted that there was very little Cl^- identified on the copper surfaces.

4.7.6. Further experiments

SKB [18] reported that two new experimental studies have been started with the aim of improving the detailed description of corrosion under unsaturated conditions in the early stages after disposal: a small-scale laboratory test being conducted at a lab in the UK to measure corrosion at different stages of O_2 depletion; and a medium-scale test being conducted at the Äspö HRL to monitor the development of the gas-phase composition in an experimental setup with copper and unsaturated bentonite clay.

Based on assessment of the corrosion analysis carried out by SKB in the LOT S2 and A3 project, the reviewers have identified opportunities for improving the quality of the analysis and approach that could be considered in SKB’s plans for recovery and analysis of the final LOT test parcel S3:

- The mean or 90 and 99 percentile pit depths or shape factors could be plotted as a function of temperature for copper samples. This would be informative if a trend is obtained, or it might otherwise strengthen the evidence for mechanical indications rather than temperature-dependent pitting [56, 57].
- It would be useful to correlate other corrosion metrics (mass loss, depth of penetration, etc.) with position and thus temperature to a greater extent than presented in TR-20-14 [6]; only one figure was presented with a few data points. These figures are potentially useful given that the period of oxic corrosion is unknown. If oxygen depletion had largely occurred before heating of the parcel, then a temperature dependency with respect to depth of corrosion attack or mass loss would imply that corrosion occurred mainly during the anoxic period. Consideration of the expected

temperature dependency of copper corrosion in groundwater under well-controlled and constant conditions, coupled with analysis of the oxygen budget and the rate of oxygen transfer from cooler regions to hotter regions (where corrosion and thus oxygen depletion would have been most rapid) would improve understanding of the spatial distribution of corrosion over the copper tube in LOT. It would be useful to compare the results of such analysis with the dependency on temperature seen by the correlation shown in the TR-20-14 report [6]. This analysis would support understanding of the amount of corrosion that occurred during the aerobic stage of LOT before the planned temperature was reached.

- Machine learning methods could be used to correlate the set of corrosion metrics and exposure data related to copper corrosion with various environmental, temporal and physical parameters [58, 59, 60]. This could only be done assuming that sufficient data can be acquired to train the model while leaving enough data to verify such a model. Then the model could be exercised under various conditions with various parameters as hidden layers. This would not provide any new science but would perhaps enable correlations suggestive of cause and effect to be discovered and correlated with causative factors more clearly. A good example is the effect of temperature on pitting or uneven attack where a relationship might emerge.
- Electrochemical pitting versus local mechanical damage produced during preparation of as-received materials could be better understood – perhaps by comparing three-dimensional reconstructions to distinguish pit shapes from the inherent shapes of mechanical anomalies. This method (i.e., three-dimensional quantitative reconstruction of corrosion damage morphology) is becoming more widely accessible if not routine today [61, 62, 63]. Three-dimensional pit reconstruction could be used to distinguish real pits from grinding-induced mechanical defects based on a more rigorous quantified process.
- It would be useful to ascertain whether LOT-type exposures can produce compact copper-sulfide films. The retrieved LOT specimens could be tested using linear sweep voltammetry (LSV) or by potential step repassivation or other means to assess whether thin film is protective and passivating. Additional Cu-S growth under rotating disk electrode tests could clarify whether Cu_2S formation on LOT coupons is mass transport controlled [64, 65, 66].
- An analysis of proposed mechanisms for hydrogen production by corrosion coupled to a hydrogen uptake law in copper and hydrogen diffusion could be used to estimate the depth profile of hydrogen in copper for up to 20 years. A method such as GDOES could be used to measure the hydrogen concentration in copper (if above its detection limit). Such an analysis may provide further understanding of the corrosion processes that occurred.

5. Conclusions

This report presents the results of an assessment of SKB's approach to quality assurance in the decommissioning of the LOT S2 and A3 parcels, focusing on the analysis of copper corrosion. The work has involved review of SKB's management of the LOT project in order to understand how QA procedures have been applied, as well as review of SKB's reports on dismantling the S2 and A3 test parcels [5] and on analysing the corrosion of copper coupons and copper tubes from the test parcels [6]. A series of meetings has held with SKB in order to discuss specific aspects of QA in SKB's LOT S2 and A3 project. Also, the interests of a number of Swedish environmental organisations and corrosion scientists have been considered via a meeting and document review in order to understand their expectations from the QA review of the LOT S2 and A3 project. Conclusions are presented in this section in terms of reviews of QA in the management of the LOT S2 and A3 parcel project and of QA in the copper corrosion analyses.

5.1. Project Management

SKB manages its activities within a company-wide project management model, but the LOT project was established before the current management system was implemented. The fact that the project management system and responsibilities for activities in the LOT project have changed over the two decades that the project has run to date is not surprising. The changes do not appear to have had any significant detrimental impacts on how the project has been run.

The timing of the LOT S2 and A3 parcel recovery and analysis has received some criticism from those who believe that SKB has delayed parcel retrieval in order to suppress the debate on copper corrosion. SKB had originally planned to recover the LOT S2 and A3 parcels after five years, but extended this to 20 years in line with delays in repository licensing. SKB noted that internal documents from 1999 mention the possibility of running the experiment for a 20-year period. The plan to retrieve the test parcels in 2019 was stated in SKB's 2016 RD&D programme. The extended experimental period has allowed a longer exposure period for the copper.

SKB is running LOT S2 and A3 parcel recovery and analysis as a project that has a clear management and decision-making structure covering the project lifecycle. Notable aspects of the planning and conduct of the project are:

- Project planning included consideration of comments from SSM relating to the repository licence application that are relevant to the project.
- Risks and risk mitigations were considered, with SKB noting in particular that it was valuable to involve contractors who had substantial previous experience of LOT since its installation, because learning from the previous work was directly available.
- Openness and transparency in the LOT S2 and A3 project have been addressed by, for example, ensuring that parcel retrieval was filmed and making samples from the parcels available for analysis on request, although this depends on the objectives and competence of interested researchers. However, as noted in Section 5.2, certain decisions on the analyses to be undertaken and presented are unclear.

Work on the LOT S2 and A3 project is organised according to work packages, with details of the work set out in SKB's activity plans or work plans provided by contractors against work package requirements. SKB has engaged a number of contractors to work on the LOT S2 and A3 project: Clay Technology AB, Uppländska Bergborrning AB, RISE KIMAB AB and Swerim AB. These contractors all have extensive experience and appropriate management systems for such work. Although such contractors can be authorised to be work managers on SKB projects, they are not permitted to be involved in decision-making relating to selection of suppliers where there may be conflicts of interest. Also, the corrosion specialists RISE KIMAB AB and Swerim AB are large organisations; work for SKB forms only a small part of their overall corrosion research activities, so there is no sense of dependency on SKB.

Contracting companies working on research projects may provide internal company reports to SKB that are then editorially post-processed within SKB (involving typesetting and printing of the report). However, SKB's experts were significantly involved throughout the LOT S2 and A3 corrosion analysis and so wrote report TR-20-14 collaboratively with RISE KIMAB and Swerim. No separate RISE KIMAB or Swerim reports have been produced for the LOT S2 and A3 project, but SKB has been provided with all results and images from the corrosion analysis. SKB stores raw data in its SICADA database management system and information such as photos, reports and memos in a document management system.

5.2. Copper Corrosion Analysis

The original objective of LOT was to validate models and hypotheses about the properties of bentonite buffer material as well as microbiology, radionuclide transport, copper corrosion and gas transport processes under repository-like conditions. However, certain aspects of the way the LOT project was set up over twenty years ago mean that there are inevitable limitations in terms of what can be learnt about copper corrosion under repository conditions from analyses of the LOT S2 and A3 test parcels. Specifically:

- The copper coupons and copper tubes were not characterised before installation. Also, the copper reference materials selected as controls were not characterised prior to their twenty-year period of storage. Thus, it is difficult to distinguish between defects of mechanical origin associated with material preparation and machining, the effects of corrosion under LOT conditions, and the effects of corrosion during dry storage of the reference materials.
- No measurements of microbial populations in groundwater were made for the LOT experiment. Thus, no clear conclusions can be drawn about the influence of microbial populations on oxygen consumption and how it might have affected the amount of oxygen available for aerobic corrosion of copper, although SKB argues that conditions were not favourable for microbes.
- There was no monitoring of redox conditions, because limited technologies were available to measure redox potential in compacted bentonite in the 1990s.
- Radiation effects are not accounted for, although they are not expected to be significant [13].

The LOT A3 and S2 copper corrosion analyses do confirm a temperature dependence of corrosion, although the form of the temperature dependence is uncertain. The corrosion analysis of the parts of the copper tubes that experienced the highest temperatures was limited. For instance, copper from the hottest parts of tube A3 was not examined metallographically, although corrosion in such regions was estimated based on measurements of the accumulated mass of copper corrosion products that had diffused into the bentonite next to the tube sample. SKB has stated that the selection of tube samples for analysis was done for the practical reason that they were in the same bentonite blocks as the corrosion coupons and that the maximum temperatures experienced by the tube samples that were examined are close to the peak temperature that any copper canister in the KBS-3 repository would be expected to experience.

Based on measurements of the accumulated mass of corrosion products in the bentonite next to the tube samples, average corrosion depths were estimated to be 0.2 to 4.8 μm for the LOT S2 copper tube and 0.2 to 13.8 μm for the LOT A3 copper tube. The largest average corrosion depths are associated with the copper tube samples that experienced the highest temperatures. Also, corrosion analyses for other LOT parcels retrieved after 1 year and 6 years show similar corrosion behaviour. Such observations suggest, but do not prove, that corrosion occurred early. It is not possible to infer how much of the corrosion occurred before the tube heaters were switched on and had reached their maximum temperatures; there was a period of about 4 months between LOT S2 and A3 parcel installation and the heaters being switched on, and it took a few months for maximum temperatures to be reached. Also, the more rapid consumption of oxygen by corrosion in the warmer parts of the tube is likely to have drawn oxygen from the cooler parts to the warmer parts, thereby enhancing total aerobic corrosion in the warmer parts and reducing it in the cooler parts. Such an axial thermal gradient would not be present along disposal canisters in a repository, because heat would be generated more uniformly along the length of the canister by radioactive decay of the spent fuel. It can be concluded that the maximum integrated corrosion rate of 0.7 μm/year for the hottest part of LOT A3 copper tube, when assuming that corrosion occurred at a uniform and linear rate with respect to time for the duration of the 20-year experiment, is not representative of, and most likely overestimates, the long-term corrosion rate for copper.

The total average accumulated corrosion depth of the LOT S2 and A3 copper coupons was estimated to be 0.7 to 1.3 μm based on gravimetric analysis. This is reasonably consistent with observation of coupon corrosion in other LOT parcels (1.5 to 4.8 μm). Differences are likely to be due to spatial variations in local conditions and the lower temperatures in the vicinity of the coupons.

SKB argues that O_2 was the main oxidant causing the corrosion of the copper coupons and tubes in LOT. Some corrosion of the tubes may have occurred in the period before the heaters were switched on and groundwater injection began. Temperature dependent aerobic corrosion requires that O_2 was still present for a period after this commencement of the tests. Some O_2 may have dissolved in the groundwater injected to fill the voids, which could have affected corrosion if it diffused to the copper surfaces. Aqueous Cu^{2+} may have prevailed as an intermediate oxidant on a much longer timescale to react with metallic copper to form Cu^+. However, the cathodic reactant Cu^{2+} is finite in such a closed system, assuming that it is only produced during oxic corrosion, and can be expended to produce a finite depth of attack. A long period of minor anaerobic corrosion, except for uncertain areas of non-uniform attack, may have occurred as a result of the slow diffusion of low concentrations of sulphide (S^{2-}) from groundwater and possibly sulphate-reducing bacteria through the bentonite to the copper surface to form insoluble Cu_2S, although evidence of the Cu-S phase on the tubes is limited. Cross-sections of corroded copper surfaces do indicate inner scale consisting of Cu_2O and outer layers of Cu_xS, which could be interpreted to support the notion that an initial short period of oxygen-induced corrosion was followed by a long period of anoxic sulphide-induced corrosion. However, observations of thicker corrosion products in pits and the lack of detailed analysis of surface anomalies versus pitting leaves open the possibility that copper pitting has occurred. Understanding the analysis of micrographic cross-sections, and any dependency there might be on temperature, is hampered by the lack of clarity in how the cross-sections relate to locations on the copper tubes.

A mass balance was attempted where the moles of O_2 available in the LOT parcel volumes were related to the amount of copper corrosion, although this exercise was complicated by the fact that oxidised copper exists in Cu cations in corrosion products and in bentonite as it diffuses away from the copper tubes. Also, the mass balance did not consider potential oxygen consumption by other means, such as bacterial interactions with O_2 within the groundwater.

The uncertainty in the saturation time of the LOT parcels and the effects of different oxygen consumption processes does mean that alternative interpretations of system evolution and oxygen availability for corrosion could be made. For example, if the gaps around the copper tubes had filled with water rapidly before the tubes could be exposed to a significant period of increased temperature, and rapid microbial consumption of oxygen had occurred, then a temperature-dependent anaerobic process would have been responsible for corrosion before any arrival of sulphide. However, a previous review of SKB's corrosion analysis concluded that any copper corrosion by sulphide attack would far exceed the corrosion depths of penetration that have been estimated could occur by anoxic corrosion in pure water in saturated bentonite [13]. Thus, corrosion by sulphide attack is of greater concern in safety assessments than any postulated corrosion in oxygen-free water. Even so, SKB does not present an understanding of what the hydrogen concentration 'should' be and what absorbed hydrogen profile should exist in copper, or whether it would be detectable, if anoxic corrosion by water and hydronium ion reduction occurred in LOT.

Alternative arguments do not support the observation from analysis of different LOT parcel tests conducted over different lengths of time that most corrosion appears to have occurred in the early stages of the tests when conditions are likely to have been aerobic. Thus, although it is not possible to conclude with absolute certainty that corrosion of the copper tubes and coupons occurred predominantly under aerobic conditions in the early stages of LOT (noting the above observations about possible pitting), there is no evidence available from these results to suggest that SKB's interpretation of copper corrosion behaviour during LOT exposures is incorrect.

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