mkg
Miljöorganisationernas
kärnavfallsgranskning

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To: Strålsäkerhetsmyndigheten (SSM)
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Input to the SSM quality assurance review of the SKB LOT project corrosion results in the autumn of 2020

Miljöorganisationernas kärnavfallsgranskning[1], hereafter called MKG, together with Naturskyddsföreningen[2] attended a meeting on September 30, 2020, with Strålsäkerhetsmyndigheten[3], hereafter called SSM, and the company Galson Sciences. Also present at the meeting were two researchers from the Royal Institute of Technology (KTH) in Stockholm. At the meeting MKG provided input for the planning of the quality assurance review of the copper corrosion results from the LOT project experimental packages A3 and S2 that will be carried out by SSM with the support of Galson Sciences. The experimental packages were retrieved in the autumn of 2019 by the nuclear waste company Svensk Kärnbränslehantering AB, hereafter called SKB.

The MKG input as presented at the meeting are summarised in this document, where the documents referred to in the meeting presentation are referenced to with links or added as appendixes.

MKG plans to follow up with further comments to SSM after studying the SKB reporting of the copper corrosion in the experimental packages.

1. Why the interest from MKG and its member organisations in the copper corrosion results from the LOT A3 and S2 packages?

MKG would like to stress that the interest of the organisation in the copper corrosion results from the LOT A3 and S2 packages is not based on a belief that the results of a 20-year experiment can be extrapolated into the long term.

[1] The Swedish NGO Office for Nuclear Waste Review (http://www.mkg.se)
[2] The Swedish Society for Nature Conservation (http://www.naturskyddsforeningen.se)
[3] The Swedish Radiation Safety Authority

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The important question that MKG understands could be answered with the results from the LOT packages is if the unexpectedly high corrosion with pitting that appears be taking place in hard rock laboratory experiments with clay and copper is a sign that the theoretical understanding of copper's behaviour in an anoxic repository environment, that is the scientific basis for SKB's safety analysis, is flawed.

To be able to answer this question there has to be a correct and scientific description of the copper corrosion that has taken place in the 20-year LOT A3 and S2 packages.

Importantly, a qualified assessment also has to be made after what time the packages became anoxic, how much corrosion could have taken place before that time, and how much corrosion can to be attributed to corrosion under anoxic conditions.

MKG understands that the LOT experiment, despite the fault of not measuring the oxygen in the system, is likely the best available experiment – and possibly the only one done by SKB – that can answer the important question above with some confidence.

MKG acknowledges that if the question of when the LOT packages became anoxic still remains a controversy after the reporting of the LOT A3 and S2 results, and after the quality assurance review by SSM is carried out, there may be a need for further experimental work to decide the issue. It would not be very costly or difficult to emplace a number of new, but simplified, LOT packages in the Äspö Hard Rock laboratory with the same emplacement methodology as originally used and with the sole purpose of studying when the packages become anoxic and how much corrosion has taken place at that time.

In 2008 and 2009 MKG closely studied the reporting regarding copper corrosion following the retrieval of the LOT A2 package in 2006, and understood how very inadequate the reporting was. Especially as it became clear how much unexpected corrosion had taken place after 5-6 years on the central copper tube.

MKG has closely followed the development of knowledge of how fast repository systems become anoxic and understands that even large experiments normally become anoxic after only a matter of months after sealing.

MKG has noted that SKB throughout the years following the retrieval and reporting of the LOT A2 package has not wanted to retrieve the LOT A3 and S2 packages (that are actually intended as a 10-year and a 5-year package). MKG understands that an important reason for this could have been the risk of the copper corrosion results being negative for the theoretical framework that underlies the KBS method, i.e. that copper is basically immune to corrosion after the repository conditions become anoxic[4].

MKG wants it noted that the organisation with good cause is sceptical to SKB's scientific integrity on copper corrosion issues, as well as to the scientific integrity

[4] MKG understands that sulphides may cause some copper corrosion.

and independence of the contractors the company is using for the analysis of the LOT A3 and S2 results. This means that the SSM review of the work is extremely important.

2. Input from the FEBEX experiment

The FEBEX experiment was operated at the Grimsel Test Site underground research laboratory between 1997 and 2016. The experiment consists of heated steel containers (100°C) surrounded by clay emplaced in a long tunnel. The experiment also included coupons of metals, including copper[5].

There was quite intensive corrosion (≈ 100 µm and pitting) on copper coupons after 18 years in the FEBEX experiment as reported in the report "FEBEX-DP Metal Corrosion and Iron-Bentonite Interaction Studies, P. Wersin & F. Kober (eds.), Arbeitsbericht NAB 16-16, Nagra, October 2017" that is enclosed as appendix 1[6]. One of the editors of report is a consultant that has done a lot of work for SKB[7].

There is less corrosion and no clear pitting on copper coupons retrieved from the FEBEX experiment after 5 years as reported in the report "FEBEX Project Post-mortem Analysis - Corrosion Study, V. Madina & I. Azkarate (INASMAT), ENRESA Technical Publication 08/2004". The report is enclosed as appendix 2[8].

In the Nagra NAB 16-16 report it is claimed that all the corrosion in FEBEX is oxic. But it appears the authors/editors has not even understood that there is normally anoxic steel corrosion from water, and therefore to claim that the steel in the experiment must have corroded with an external oxygen source has no basis.

There is a discussion in the FEBEX reporting that there may have been a two-year period where oxygen has leaked into FEBEX. There are two reports of relevance for understanding this possibility, "FEBEX: Assessment of Redox Conditions in Phase 2 before Dismantling, N. Giroud, Arbeitsbericht NAB 14-055, Nagra, July 2014" and "Gas and water sampling from the FEBEX in situ test, A.M. Fernández, Arbeitsbericht NAB 16-13, Nagra, May 2019". The reports are enclosed as appendices 3 and 4[9]. Although there has been some sampling of oxygen at times

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[5] The experiment is described at https://www.grimsel.com/gts-phase-vi/febex-dp/febex-dp-introduction.

[6] Many of the FEBEX experiment reports can be found at https://www.grimsel.com/gts-phase-vi/febex-dp/febex-dp-literature-publications. The report NAB 16-16 can also be downloaded here: https://www.grimsel.com/febex-dp-general-files/351-nab-16-016-metal-corrosion-and-iron-bentonite-interaction-studies-1.

[7] There are thirteen SKB reports with Paul Wersin as an author between 1992 and 2013.

[8] The report can also be downloaded here: http://www.mkg.se/uploads/Appendix_2_ENRESA_Technical_Publication_08-2004_FEBEX_Project_Post-mortem_Analysis_-_Corrosion_Study_V_Madina_I_Azkarate.pdf.

[9] The NAB 14-55 cannot be found on the internet but has been received by MKG directly from Nagra and can be downloaded here: http://www.mkg.se/uploads/Assessment_Redox_Conditions_Phase_2_before_Dismantling_Giroud_NAB_14-55_Nagra_July_2014.pdf. The NAB 16-13 can be downloaded here: https://www.grimsel.com/febex-dp-general-files/386-nab-16-013-gas-and-water-sampling-from-the-febex-in-situ-test-1.

after closure, the general conclusion is that the FEBEX experiment has likely been anoxic from very early on.

MKG understands that is difficult to explain how oxygen could be available for corrosion in the environment of the FEBEX metal coupons for any length of time. As there is normally anoxic groundwater in an experiment after only a few months (see section 4), it is unclear how any oxygen that leaks in from a distance many meters away could reach the metal coupons as oxygen would be consumed by bacteria and chemical processes on the way.

3. Input from the reporting of the LOT A2 Package

The LOT project (Long Term Test of Buffer Material) in the Äspö Hard Rock Laboratory has in total consisted of 7 experimental packages of which four (A0, A1, A2, A3) have been at a higher temperature (120-150°C) and three (S1, S2, S3) have been at a normal repository temperature (90°C). There is a 4-meter-high central copper tube (10 cm diameter) surrounded by bentonite clay. The lower half of the copper tube is heated. Each experimental package also includes four copper coupons in the upper part of the clay. In 2009 MKG made a summary of information about the experiment. This is enclosed as appendix 5[10].

The LOT A2 package was retrieved in the beginning of 2006. A draft report was ready already in September 2008 but it took more than a year more to publish the final report as “Long term test of buffer material at the Äspö Hard Rock Laboratory, LOT project: Final report on the A2 test parcel, O. Karnland et al., TR-09-29, SKB, November 2009”. The report is enclosed as appendix 6[11].

There is no reporting of the unexpectedly large copper corrosion from the central tube in report. The copper coupons at 70°C are reported as damaged and there are no pictures or analysis of them in the report.

There are only general photographs and microscopic pictures of surfaces copper coupons at 30°C in the reporting of these in appendix 3 of the report. No microscopic cross sections are shown and there is just a statement that there is no obvious sign of pitting corrosion. The general and microscopic pictures do show quite large corrosion and a micro hardness indentation mark of unknown depth had disappeared.

In the autumn 2008 MKG was sent a draft of the LOT A2 report from SKB as the final report was taking time to be finalise. The report and appendix 3 of the report are enclosed as appendix 7 and 8[12]. The final report differs from the draft report in

[10] The summary can also be downloaded here: http://www.mkg.se/sites/default/files/old/uploads/Summary_of_experimental_packages_in_the_SKB_LOT_experiment_-_Johan_Swahn_MKG_-_May_2009.pdf.

[11] The report SKB TR-09-29 can also be downloaded here: https://www.skb.se/publikation/1961944.

[12] The TR-09-29 draft report with all the appendixes can also be downloaded here: http://www.mkg.se/mkg-lagger-ut-rapporter-fran-forsoken-i-skbs-berglaboratorium-i-aspo. For direct download of the draft report: http://www.mkg.se/sites/default/files/old/uploads/LOT_A2_Draft_main_report_080930.pdf. For direct download of appendix 3:

that there is an attempt to explain the corrosion in the package by oxygen trapped in the system.

There is also a separate SKB report, written by the same consultant that co-edited the FEBEX NAB 16-16 corrosion report (Paul Wersin), that tries to estimate the corrosion on the central tube by using measurements of copper in the clay. It is called “A2 test parcel, Compilation of copper data in the LOT A2 test parcel, P. Wersin, TR-13-17, SKB, December 2013”. The report is enclosed as appendix 9[13].

The author of the report comes up with a case where the corrosion can almost be explained if all possible oxygen inside the package causes copper corrosion. This explanation misses that 1) there is an extensive crust of corroded copper still in the central tube that involves much more corrosion and that can therefore not be explained by trapped oxygen and 2) that it is impossible that all the oxygen from remote parts of the package reaches the hottest part of the heater. In fact, most of the trapped oxygen in the package is likely consumed by bacteria and chemical processes and never takes part in any copper corrosion.

In addition, the LOT packages are filled with groundwater from a pipe into the bedrock as part of the emplacement at the time the heaters were started. This water is very likely anoxic as it enters the package. This water has filled the space between the clay and the bedrock and the space between the clay and the copper tube. This groundwater will remain anoxic and any oxygen that reaches the water will be consumed immediately and never reach the copper tube.

So, with high likelihood most – if not all – corrosion of the central metal tube of the experimental package A2 has been due to anoxic corrosion.

In May-June of 2008 here was a correspondence between Peter Szakálos (KTH) and Stephan Kaufhold (BGR in Germany), the latter who had done similar calculations of corrosion using clay measurements as were done in SKB TR-13-17. The main results of the calculations of Stephan Kaufold are enclosed as appendix 10[14]. The correspondence between Peter Szakálos and Stephan Kaufhold is enclosed as appendix 11[15]. The calculated corrosion rate is 2-4 µm per year for a position representing clay zone 15 and to this number an amount (up to times 4?) should be added to correspond to the corrosion products left on the copper surface.

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http://www.mkg.se/sites/default/files/old/uploads/Appendix_3_with_appendices_to_LOT_A2_Main_draft_report_-_BR_080930.pdf.

[13] The report SKB TR-13-17 can also be downloaded here: https://www.skb.se/publikation/2682520.

[14] The document can also be downloaded here: http://www.mkg.se/uploads/Appendix_10_Kaufhold_-_Corrosion_rate_LOT_A2_calculations.pdf. An Excel file with all the data and all the calculations can be downloaded here: http://www.mkg.se/uploads/Appendix_10_data_Kaufhold_-_Corrosion_rate_LOT_A2.xls.

[15] The correspondence can also be downloaded here: http://www.mkg.se/uploads/Appendix_11_Correspondence_between_Peter_Szakalos_KTH_and_Stephan_Kaufhold_BGR_090602-090603.pdf.

4. What do we know about oxygen consumption?

In the REX experiments carried out by SKB around the year 2000 it was shown that water in bedrock becomes anoxic in a matter of days with a substantial contribution by bacterial activity. The report is called "O_2 depletion in granitic media: The REX project, I. Puigdomenech et al., TR-01-05, SKB, February 2001" and is enclosed as appendix 12[16].

In the MiniCan project, also an SKB experiment in the Äspö Hard Rock Laboratory it was shown by electrochemical potential measurements that anoxic conditions in compacted bentonite are reached in 2-3 months. The results are in the report "Miniature canister corrosion experiment – results of operations to May 2008, N.R. Smart & A.P. Rance, TR-09-20, SKB, July 2009". The report is enclosed as appendix 13[17].

Measurements during the emplacement of the FE experiment at the Mont Terri hard rock laboratory in Switzerland shows that an over 20 m long tunnel section filled with clay became anoxic after a few months. More information about this can be found in the articles "Implementation of the full-scale emplacement (FE) experiment at the Mont Terri rock laboratory, H.R. Müller et al., Swiss J Geosci, vol. 110, pp. 287–306", 2017" (see page 296 and figure 11) and "The corrosion of radioactive waste disposal canisters based on in situ tests, N. Diomidis & F. King, pp 371-389 in Nuclear Corrosion Research, Progress and Challenges European Federation of Corrosion (EFC) Series, 2020". The first article is available as "open access" and the second article been purchased by MKG and they are both enclosed as appendices 14 and 15[18].

The second article is co-authored by Frasier King who is a corrosion scientist that SKB has relied upon historically to argue for the company's theoretical model for copper corrosion in the KBS method. In the article it is stated:

"It thus seems as if not only is the aerobic phase much shorter than previously believed, but also that the initially trapped O_2 may be consumed by processes other than canister corrosion. If true, this observation has potentially profound implications for the performance of HLW/SF canisters since it is only during the initial aerobic phase that rapid localized forms of corrosion are believed to be possible for a number of candidate canister materials."

From the above studies MKG finds it very likely that all the copper corrosion in the FEBEX and LOT experiments have been under anoxic conditions already from the start, especially the LOT experiment where the experimental packages have been filled with anoxic granite ground water at the time the heating has started. If there

[16] The report SKB TR-01-05 can also be downloaded here: https://www.skb.se/publikation/18352.
[17] The report SKB TR-09-20 can also be downloaded here: https://www.skb.se/publikation/1964603.
[18] Müller et al. is available here: https://link.springer.com/article/10.1007/s00015-016-0251-2 and also here: http://www.mkg.se/uploads/Appendix_14_Implementation_of_the_full-scale_emplacement_(FE)_experiment_at_the_Mont_Terri_rock_laboratory_Muller_Swiss_J_Geosci_(2017).pdf. Diomidis & King is available here: https://www.sciencedirect.com/science/article/pii/B978012823719900010X, but also here: http://www.mkg.se/uploads/Appendix_15_The_corrosion_of_radioactive_waste_disposal_canisters_based_on_in_situ_tests_Diomidis_&_King_EFC_Series_2020.pdf.

is anoxic water between the clay and the central copper tube, no oxygen can reach the copper surface at it will instead be consumed by bacteria in the anoxic water “barrier”.

In addition, even if the SKB claim that the copper corrosion in the LOT A2 package was only due to all the trapped oxygen is accepted, there is no more oxygen available for any further corrosion in the A3 and S2 packages. Any further corrosion compared to the A2 package cannot therefore be explained as oxic.

MKG would like to strongly stress that the way SKB continually keeps stressing that a number of copper corrosion products can only occur if the environment has been oxic is fundamentally flawed. This would be true only if the theoretical framework that the company builds its safety case on is true. If there is a reaction between copper and water molecules, that of course contain oxygen, then all the same copper corrosion products would be the result.

MKG would also like to point out that the question of when the LOT experiment and other experiments operated by SKB have become anoxic has been of interest for previous quality assessments done by SSM of the copper corrosion work carried out by SKB. As an example, minutes from a meeting between SSM, Galson Sciences and SKB in August 2014 includes such discussions and are enclosed as appendix 16[19]. MKG is of the understanding that it should be clear by now that SKB has no interest in improving the scientific understanding of this issue as it allows the company to claim that all copper corrosion is oxic, while in fact it is much more likely that very little of the copper corrosion in experiments carried out in hard rock laboratories is anoxic.

5. What do we know about the LOT S2 and A3 packages?

Information on approximate temperatures in different parts of the packages can be found in the SKB report “Installation, monitoring, dismantling and initial analyses of material from LOT test parcel S2 and A3: Results from field test, T. Sandén & U. Nilsson, TR-20-11, SKB, June 2020”. The report is enclosed as appendix 17[20].

In the report there are details in sections 2.2.2 (Water inflow) and 3.2.2 (Water supply) of how the gaps between the copper and clay, and clay and rock, were filled with granitic anoxic water at the time the heating was started, and that a feed of anoxic water under pressure from a borehole in the rock was kept attached to the bottom of the experimental packages for the whole operation.

The copper in all LOT copper tubes is DHP copper (SS 5015-04), i.e., deoxidised acid-free 99,9% copper with 0,04% phosphorus. This copper is quite comparable to the OFP copper with 50 ppm phosphorus to be used in the copper canisters in the KBS method.

[19] The minutes can also be downloaded here: http://www.mkg.se/uploads/Appendix_16_SSM_Minutes_from_the_meeting_with_SKB_regarding_QA_in_SKBs_Copper_Corrosion_Experiments_29_August_2014.pdf.
[20] The report SKB TR-20-11 can also be downloaded here: https://www.skb.se/publikation/2495225.

At the bottom end of each tube, a copper plate and 4 copper reinforcement parts were attached by use of soldering silver. The composition of the copper plate is not described in the report.

The upper part of the copper tube was open to the Äspö tunnel and the interior of the tube has thus been filled with air during the entire exposure period. At the bottom end of each tube the copper plate provides an air-tight endpiece. Since no leakage of water/steam or air between the Äspö tunnel and the buffer material in LOT could be accepted the impenetrability was tested after the soldering of the bottom plate by use of a helium source inside the copper tube and an external detector.

All eight copper coupons from the S2 and A3 packages were retrieved. In the SKB TR-09-29 report, appendix 3, the author (Bo Rosborg) mentions that he saved a reference coupon for later to allow a comparison during cleaning procedures.

6. What do we know about Taxén at Swerea/Kimab and Clay Technology?

Clay Technology Sweden AB (started by Lennart Börgesson and owning 70%), co-owned by Harald Hökmark (15%) and Ola Karnland (15%)) owns the daughter company Clay Technology Lund AB, where all work is done. Since the companies were started in the mid-1990s basically only work for SKB has been done and much of the work has been with the experiments at the Äspö Hard Rock Laboratory. Scientifically the company Clay Technology Lund AB cannot be separated from SKB. The company Clay Technology Sweden AB has been making millions of SEK in profits yearly that have been made the owners multi-millionaires with most of the funding coming from SKB[21].

Claes Taxén at Swerea/Kimab has a long history of working for SKB on projects involving copper corrosion. He has been an author on sixteen SKB reports between 2000-2019.

He has carried out at least two projects showing problematic results for SKB where the results have not been published.

1. Claes Taxén is the author of a 2009 Swerea/Kimab report called “Exposure of Copper in Äspö Groundwaters” that discovered by SSM in the autumn of 2014 as a part of the then ongoing quality review of SKB’s copper corrosion work. The report is registered in SSM Dnr SSM2011-2306 and is enclosed as appendix 18[22]. SKB has claimed that the project was discontinued for technical reasons, but the report shows possible anoxic corrosion of copper as the water used is clearly anoxic water from the bedrock.

[21] MKG has studied the annual reports of the companies from 1998 to 2015. Between 2005 and 2015 the owners of Clay Technology Sweden AB received over SEK 50 million in dividends.
[22] The report is also available here: http://www.mkg.se/uploads/Appendix_18_Exposure_of_copper_coupons_in_Aspo_groundwaters_Taxen_Swerea-KIMAB_2009.pdf.

2. Claes Taxén is the co-author of an internal 2007 SKB report on corrosion from earth currents in Forsmark. The report is enclosed as appendix 19[23]. The report is a result of a project created when corrosion damage was found in measuring equipment (see SKB P-05-265, not enclosed[24]) and contains information of relevance for the safety of underground activities in Forsmark. MKG received the report anonymously and informed SSM about it in October 2012. The report is registered as part of Dnr SSM2011-3937-8. Claes Taxén has later in 2014 written an SKB report (SKB TR-14-15, not enclosed[25]) together a staff member from the company where he helps with the argument that earth currents are not a threat to the safety case of the spent fuel repository.

In recent years Claes Taxén has worked for SKB with projects on stress corrosion cracking of copper. Two reports are SKB TR-17-16 and TR-19-13 and are not enclosed[26]. In the first report the use of metallographic cross-sections is traceable to corroded surfaces. In the second the cross-sections are in certain cases only presented as “representative”.

7. What do we know about SKB:s contracts with consultants?

MKG has through a freedom of information request to Uppsala University obtained a 2011 research contract between SKB and researchers at a university department[27]. The contract includes a part with general conditions for an SKB research contract. These are enclosed as appendix 20[28].

One clause regarding “Ownership” states that “Alla resultat som framkommer i samband med Uppdraget, inklusive datorprogram med tillhörande källkod och dokumentation, ska utgöra SKB:s egendom. Resultatet av Uppdraget får fritt disponeras, ändras och bearbetas av SKB”.

Freely translated this means that “All results that transpire from the Assignment, including computer programmes with associated source code and documentation, are to be owned by SKB. The results from the Assignment may freely be used by, changed by and processed by SKB”.

MKG is of the understanding that this means that SKB can edit all results in the final published SKB report so that only results that are to the benefit of the company are presented. This is especially true if SKB staff are co-authors of a report.

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[23] The report can also be downloaded here: http://www.mkg.se/uploads/Appendix_19_SKB_Korrosionsutredning_slutrapport_-_Utredning_korrosionsproblem_jordstrommar_borrhal_Forsmark_Oskarshamn_R_Sandstrom_et_al_071214.pdf.
[24] The report SKB P-05-265 can be downloaded here: https://www.skb.se/publikation/1093421.
[25] The report SKB TR-14-15 can be downloaded here: https://www.skb.se/publikation/2478347.
[26] The report SKB TR-17-16 can be downloaded here: https://www.skb.se/publikation/2491754. The report SKB TR-19-13 can be downloaded here: https://www.skb.se/publikation/2492745.
[27] See http://www.mkg.se/skbs-forskningsavtal-ger-aganderatt-till-resultat.
[28] The document can also be downloaded here: http://www.mkg.se/uploads/Appendix_19_SKBs_Inkopsvillkor_forskningsuppdrag_2011.pdf.

8. What do we need to see from the reporting of LOT S2 and A3?

MKG expects to see the following in the SKB reporting of the copper corrosion in the LOT A3 and S2 packages:

- Overall corrosion pictures of all the copper coupons and the central copper tubes, especially of the heated bottom parts.
- Clearly traceable pictures, microscopic pictures and microscopic cross sections of the most corroded parts.
- Comparable measurements of copper in clay to compare to the measurements from LOT A2. Also, an estimate of copper in corrosion products left on the surface of the copper tube when clay is removed.
- Corrosion rate results from the copper coupons and the central copper tubes (best estimate)
- An evaluation if there been more corrosion in LOT A3 than in LOT A2 and the difference in corrosion with LOT S2 at repository temperature.

MKG plans to follow up with further comments to SSM after studying the SKB reporting of the copper corrosion in the experimental packages.

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