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

On October 8, 2020, Miljöorganisationernas kärnavfallsgranskning[1], hereafter called MKG, made a first input to the quality assurance review that is being carried out by Strålsäkerhetsmyndigheten[2], hereafter called SSM, of the copper corrosion results from the LOT project experimental packages A3 and S2. The 20-year experimental packages were retrieved in the autumn of 2019 by the nuclear waste company Svensk Kärnbränslehantering AB, hereafter called SKB.

The first input was put together based on the observations MKG wanted to share with SSM before analysing the report that SKB has published with the copper corrosion results. The input included the following points[3]:

- Why the interest from MKG and its member organisations in the copper corrosion results from the LOT A3 and S2 packages?
- Input from the FEBEX experiment
- Input from the reporting of the LOT A2 Package
- What do we know about oxygen consumption?
- What do we know about the LOT S2 and A3 packages?
- What do we know about Taxén at Swerea/Kimab and Clay Technology?
- What do we know about SKB's contracts with consultants?
- What do we need to see from the reporting of LOT S2 and A3?

On October 8 SSM asked a number of questions to MKG regarding the input that were answered on Nov 3. With the answers MKG sent a corrected version of the first input to SSM[4].

[1] The Swedish NGO Office for Nuclear Waste Review (http://www.mkg.se)
[2] The Swedish Radiation Safety Authority
[3] The input with 20 appendices can be found here: http://www.mkg.se/mkg-bidrar-med-synpunkter-till-myndighetens-lot-granskning .
[4] The questions/answers with three attached documents and the corrected first input can be found here: http://www.mkg.se/mkg-svarar-p-uppf-ljande-fr-gor-fr-n-ssm-i-kvalitetsgranskningen-av-lot .

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In the first input to SSM, MKG stated that the reporting from SKB on the copper corrosion in the LOT packages needed to include:

- 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.

The SKB report with copper corrosion results from the LOT project experimental packages A3 and S2 was published on October 1 as "SKB TR-20-14, Corrosion of copper after 20 years exposure in the bentonite field tests LOT S2 and A3, Adam Johannes Johansson et al., September 2020"[5][6].

MKG has also studied the report and also the documents that SKB has provided to SSM on October 1 (Äspö facility and corrosion project steering documents and October 2 (SKB review documents of TR-20-14) as part of the authority's quality assurance work[7].

Below is the second MKG input to the SSM quality assurance review project.

1. The is a need to see the supporting consultancy reports that SKB has used to write the report TR-20-14, including better quality images

The report TR-20-14 is authored by SKB as well as by consultants from the entities Rise KIMAB and Swerim. As the consultants must have worked on contracts there should be supporting reports from these two research groups with results that have been sent to SKB. These reports should be made available to SSM in versions that have not been "quality controlled" by SKB. SKB's "quality control" of research results possibly includes a component of not releasing results that are not favourable to the company.

It is not stated in the report what work the consultants from Swerim have done. It is possible that their work has been the XRF studies, but this needs to be verified.

There may also be other supporting reporting of results and information made available to SKB that has been used to make the report TR-20-14. This should also be made available to SSM, as well as all the contacts for supporting work that SKB have had for the analysis and review of the LOT S2 and A3 packages.

In the original report TR-20-14 many of the images had a poor resolution which made it difficult to see the corrosion, especially in metallographic cross sections.

[5] The report can be found here: https://www.skb.se/publikation/2496000 .
[6] In this input the SKB reports that can be found on the SKB web site are not added as appendixes. However, they can also be found here on the MKG web site: http://www.mkg.se/mkg-i-det-andra-ssm-bidraget-om-lot-f-rs-ket-behov-av-mer-korrosionsstudier .
[7] These documents (points 5 and 7) can be found on the MKG web site where all documents registered by SSM in the quality assurance project can be found: http://www.mkg.se/ssm2020-5740-kvalitetsgranskning-av-skbs-lot-experiment .

MKG contacted SKB early to ask for a version of the report with higher resolution. There is now a higher resolution version of TR-20-14 on the SKB web site[8].

Any supporting reports that are made available must also have high enough resolution images to be able to be analysed.

The reason that MKG has understood that it is important to have the supporting consultancy reports is to show that SKB is accurately representing all important results in the report TR-20-14. MKG would also like SSM to contact the consultants directly to ensure that they regard the representation of their work in TR-20-14 as being correct.

The reason why it is important for SSM to have access to the consultancy contracts is to know who is responsible for the supporting work, if there are limits to what the consultants can communicate with other parties than SKB regarding the results, and if the consultants have control of how the results are presented by SKB in the report TR-20-14.

2. There is a general need for more detailed information of where images of metallographic cross sections on different surfaces in report TR- 20-14 originate from

MKG has observed that it is generally difficult to understand exactly where the metallographic cross section images presented in TR-20-14 originate from on different surfaces. There is a need to describe in more detail exactly how the surface looks like at the exact point where the cross-section images are taken. It is also important to have images of the whole surfaces that have been examined made available in order to be able to assure that the areas that are examined with metallographic cross section images are the most scientifically relevant to study.

The reason that MKG has understood that this is important is to make sure that the most scientifically interesting areas for the understanding of the extent and type of copper corrosion on the copper tubes and coupons are studied and reported in the report TR-20-14. Only by doing this can the report be seen as scientifically relevant for answering the question of whether copper is suitable as a canister material.

3. There is a need for a detailed description of how groundwater was filled into the LOT packages as the heaters were turned on

In the first input to the SSM quality assurance project MKG discussed the way that SKB filled the experimental packages through titanium tubes to the bottom of the packages. The packages were filled with anoxic groundwater with a slight overpressure that was kept after the packages were sealed.

MKG understands that there is a need to have a more detailed description of how this was done in order to understand if the spaces between both the rock and clay and copper and clay were filled with anoxic groundwater before the packages were sealed and the heaters turned on.

[8] There is a low-resolution and a higher-resolution version of SKB TR-20-14 here: https://www.skb.se/publikation/2496000 .

The reason for filling the packages with groundwater was that there was not enough water available from the bedrock surrounding the deposition holes. The water was needed for the bentonite clay to swell as fast and evenly as possible.

To MKG it would seem reasonable that the experimental packages were filled so that both gaps contained groundwater and that they were then sealed. If both gaps were not filled the swelling of the clay would not be as uniform as possible. If the results of the one-year LOT packages are studied the swelling pressure reaches an equilibrium after a year. It is possible that SKB can provide more information if there are measurements that show that the swelling has mostly taken place due to water from the gap between the rock and the clay showing that there was less water, and thereby more enclosed air, in the gap between the copper tube and the clay.

With the slight overpressure in the external groundwater supply, the seals at the top of the experimental holes had to be tight so that water would not leak out onto the tunnel floor.

MKG has studied how the installation is described in the reports for the results of the LOT packages S1 and A1, A0 and A2. The packages S1 and A1 were installed separately and earlier and did not have the system for controlled filling with groundwater from a drilled hole in the tunnel wall. The packages A0, A2, A3, S2 and S3 were installed later, and at the same time, and with the system for controlled filling.

MKG has found it difficult to understand how the system of titanium tubes in each experiment worked. In the report with the results from the one-year package A0 that was retrieved in 2001, with the report published in 2011, it is described as a titanium tubing system[9]. It is possible there are three tubes to the bottom with three filtered holes on the way down. This is to let water enter at different depths of the hole in order to simulate water entering the hole from three cracks. In, addition, in the report SKB TR-20-11, with the installation information for packages S2 and A3, on page 27 it is stated that the pore water pressure was measured via these titanium tubes[10].

SSM needs to get more information on the titanium tubing system and how it was used to fill the holes. In the report with the results from the package A0 there is a reference to an installation report with the number AP TD F62-99-114[11]. This report could provide more information to start with.

4. There is a need for a detailed description of how, if any, oxygen can have leaked into the packages

MKG sees a need for a qualified explanation from SKB of how, if any, oxygen has been able to leak into the LOT packages to be able to contribute to corrosion. Of

[9] Page 29 in ”SKB TR-09-31, Long term test of buffer material at the Äspö HRL, LOT project Final report on the A0 test parcel. O. Karnland et al., February 2011”. The report can be downloaded here: https://www.skb.se/publikation/2224202 .

[10] The report ”SKB TR-20-11, Installation, monitoring, dismantling and initial analyses of material from LOT test parcel S2 and A3, Results from field test, T. Sandén & U. Nilsson, June 2020” can be downloaded here: https://www.skb.se/publikation/2495225 .

[11] Page 27 in ”SKB TR-09-31, Long term test of buffer material at the Äspö HRL, LOT project Final report on the A0 test parcel. O. Karnland et al., February 2011”. The report can be downloaded here: https://www.skb.se/publikation/2224202 .

special interest is knowing how oxygen from the tunnel can have reached the copper bottom plate by passing through five metres of groundwater or through the pressurised tube feeding groundwater from the hole drilled into the bedrock. MKG has understood that at no time was oxygen measured in the water in the titanium tube system feeding in this groundwater to the bottom of the packages where the bottom plate is.

MKG has noted by reading the factual reviews of the report TR-20-14 by the long-time consultants Frasier King and Paul Wersin that SKB seems to be of the understanding that there has been corrosion by trapped, but not leaked-in, oxygen gas in the LOT packages for all the 20 years of operation. The reviews are appended as appendixes 1 and 2[12].

It appears that Frasier King may be somewhat sceptical of the explanation that trapped oxygen has contributed to corrosion for an extended period of time. This is understandable as he has recently published an article where he shows that he has understood that experimental packages in repository-like environments very quickly become anoxic[13].

Regarding the review of Paul Wersin, he appears to try and support the view that oxygen gas has been available for corrosion in the LOT packages for an extended period of time and have also leaked into the experimental packages. He refers to a report from the FEBEX experiment where he says that oxygen gas has leaked in and was measured all the way to the end in the 18-year experiment. He calls the report “Fernandez & Giroud, 2017, Nagra, NAB 16-003”. MKG has searched for this report and it does not exist.

After contact with the Swiss waste management company Nagra we can inform SSM that the report Paul Wersin cites eventually ended up as a single authored report by A.M. Fernandez with the report number NAB 16-13. MKG has discussed this report in the first input to SSM on October 8 and it is included as appendix 4 to that input[14]. MKG has looked at the report again and it is true that oxygen is measured in sampling throughout the experiment. However, measured redox conditions at the same time show strongly reducing conditions in the bentonite. It is suggested that any measured oxygen values are either measurement artefacts or due to leakages. Indeed, the figures 71 and 72 of oxygen measurements on page 75 show very erratic results and cannot be used to reliably say that oxygen has been present in the experiment to influence corrosion results on metals in contact with clay and normally anoxic groundwater.

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[12] The reviews can also be downloaded on the MKG web site: http://www.mkg.se/mkg-i-det-andra-ssm-bidraget-om-lot-f-rs-ket-behov-av-mer-korrosionsstudier

[13] See appendix 15 of the first MKG input to SSM, ”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” That can be downloaded 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 .

[14] The appendix 4 of the first MKG input can also be downloaded here: http://www.mkg.se/uploads/Appendix_4_Gas_and_water_sampling_from_the_FEBEX_in_situ_test_AM_Fernández_Arbeitsbericht_NAB_16-13_Nagra_May_2019.pdf .

5. There is a need for detailed corrosion studies of the surfaces of the hottest areas of the central copper tubes of both packages

MKG has understood that the maximum corrosion on the copper tubes in the LOT packages is on the areas that have been heated to the highest temperatures. These areas are in the middle of the bottom half of the tubes as the heater is situated in the bottom half of the copper tubes. MKG finds it scientifically remarkable that SKB has not chosen to publish findings with images and metallographic cross section images of this part of the tube. Without such images it is not possible to accurately estimate the extent of corrosion or the amount and extent of pitting corrosion on the most corroded parts of the copper tubes.

Instead of properly scientifically examining the surfaces of the hottest parts of the copper tubes, SKB has only tried to estimate the corrosion rate by evaluating measurements of copper content in the clay at different parts of the tube.

In order for the report TR-20-14 to be used as a scientific basis for understanding the corrosion that has taken on the copper tubes SKB must deliver images of surfaces, metallographic cross sections and analysis of the corrosion that has taken place on the hottest surfaces of the copper tubes in the LOT packages S2 and A3. It is especially important to understand the possible extent of pitting corrosion.

6. There is a need for detailed corrosion studies of the surface of the copper bottom plate of both packages.

MKG finds it interesting that there appears to be quite significant amounts of corrosion on the copper bottom plates of the two LOT packages. The copper bottom plate of the A3 package has been at a temperature of about 80°C that is a good representation of the temperature of the copper cannisters in the planned repository.

MKG understands that the corrosion of the copper bottom plates has taken place in likely anoxic groundwater from the time the heaters were turned on, and with free access to water as the plate has been standing on sand.

In order for the report TR-20-14 to be used as a scientific basis for understanding the corrosion that has taken on the bottom plate SKB must deliver images of surfaces, metallographic cross sections and analysis of the corrosion that has taken place at least on the copper bottom plate of the LOT packages S2 and A3. It is especially important to understand the possible extent of pitting corrosion.

7. There is a need to complement the corrosion estimates based on the amounts of copper in the clay with an added estimate of corrosion due to the copper in the aggregated corrosion products still remaining on the tube surface

In the report TR-20-14 SKB estimates the corrosion along the copper tube by measuring the amount of copper in the clay. Depending on how badly the tube has corroded there remains an aggregate of corrosion products on the metal surface. The corrosion that has resulted in these corrosion products has to be added to the corrosion that is calculated by measuring the copper in the clay. This is not done by SKB in the report TR-20-14.

It is difficult to estimate how much larger the measured corrosion becomes if both values are added. SSM and MKG had a discussion about this issue when SSM asked questions about the first MKG input on Sept 28 that MKG answered on November 3[15]. It is certainly possible that the corrosion in the clay should be multiplied by at least a factor of 2.

In order to find out what this factor actually is on the hottest parts of copper tubes of the LOT packages S2 and A3, SKB needs to provide information, including images of how much copper remained aggregated on the copper tube when separated from the clay. The task of providing this information would be part of the process to answer to the need for detailed corrosion studies of the surfaces of the hottest areas of the central copper tubes of both packages (point 5).

8. There is a need to try and understand why there may possibly be less corrosion when copper is in contact with a clay/water interface than with a sand/water interface

In the report TR-20-14 there are no images of the corrosion on the hottest part of the copper tubes. When such images, as well as metallographic cross sections, are made available they can be compared to the corrosion that has taken place on the copper plates (see point 6).

As the copper bottom plate in the LOT package A3 has been at an approximate temperature of 80°C the corrosion on the central tubes with this temperature can be compared to the corrosion on the plate. When looking at the corrosion on the A3 bottom plate (see Figure 1 below) MKG gets the impression that the corrosion is quite significant.

Figure 1. Image of the copper bottom plate of the LOT A3 package (Source: Figure 3-1 in SKB report TR-20-14 on page 20)

Even though it is possible that the corrosion at the same temperature on the copper tubes is of the same magnitude, the corrosion on the tubes as measured

[15] See http://www.mkg.se/mkg-svarar-p-uppf-ljande-fr-gor-fr-n-ssm-i-kvalitetsgranskningen-av-lot .

by SKB – even corrected with the corrosion of the aggregated corrosion products (see point 7) – is perhaps not be as large.

If this is the case MKG is of the understanding that there is a need to understand why this is so. A possible explanation is the free availability of anoxic groundwater in sand at the surface of the copper plate. It is possible that such an interface can allow an easier transport of copper from the surface than if the interface is with clay.

9. There is a need to understand if there is a relation between the available water in a clay/copper interface and the amount of corrosion on the copper tubes

MKG understands that if there is corrosion process involving water molecules in anoxic ground water, the availability of water to the copper surface from the clay may influence the corrosion rate. It is therefore of interest that an evaluation is made of the corrosion on the copper tubes and the copper coupons and the availability of water at the copper surfaces in the clay.

MKG understands it might be useful if SSM tries to get information on the amounts of water in the clay in different parts of the LOT S2 and A3 packages, especially at the coper tube surface and the surface at the copper coupons.

10. There is a need to explain why, if all corrosion is due to oxygen early in the experiment, there is more corrosion in the 20-year A3 package compared to the 5-year A2 package

Even though it is uncertain how high the corrosion is on the warmest part of the copper tubes has actually been (see point 7), SKB in the report TR-20-14 arrived a maximum corrosion depth of 13.8 µm on the 20-year A3 LOT package by measuring the copper in clay. SKB reports a maximum corrosion depth of 9.6 µm on the hottest part of the copper tube in 5-year A2 LOT package retrieved in 2006.

MKG understands that SKB claims that all corrosion on the copper tubes is due to trapped oxygen and early in the experiment. SKB needs to explain why there is more corrosion after 20 years than after 5 years if all the corrosion is due to approximately the same amount of trapped oxygen early in the experiment.

11. There is a need to evaluate if the SKB reasoning that all the corrosion is due to oxygen initially trapped in the packages is correct

In table 4-1 on page 67 in TR-20-14 there is an attempt by SKB to make a mass balance of the corrosion in the LOT packages due to initially entrapped oxygen. There is a need for SSM to evaluate if this explanation to all the corrosion that has taken place in the LOT test packages is credible.

MKG has understood that the sand and the rock-bentonite gap, and likely also the bentonite-copper gap, were filled with anoxic water as the heating was started. This means that in reality only the oxygen in the clay itself would be available for corrosion on the copper tube. A considerable amount of this oxygen is likely consumed by bacterial and chemical processes early in the experiment, and certainly when anoxic groundwater migrates into the clay, and cannot move towards the copper tube. Finally, if there is anoxic groundwater in the gap between the copper tube and the clay, it is not possible for oxygen from the clay to reach

the copper surface as the oxygen is consumed almost immediately as it meets the bacteria in the groundwater.

MKG has been wondering how it is possible for bacteria in ground water at depth in the bedrock to be able to consume oxygen. After all, if there is no oxygen available there should be no such bacteria there. The answer is actually quite simple. Degradation of organic matter provides the best energy exchange if the bacteria are breathing oxygen. However, if there is no oxygen most bacteria can switch to other electron acceptors with a slightly lower exchange of energy. Oxygen respiration is constitutive in many bacteria, located in the genes even if it is not needed due to lack of oxygen. This group of bacteria can be called optional anaerobes and will consume oxygen when it is available, even if the normal energy source is another compound.

12. There is a need to understand how much corrosion could have taken place in the up to 4 months before the packages were flooded and heating started

SKB appears to claim in the report TR-20-14 that considerable corrosion on the copper tube could have taken place in the up to 4 months before the packages were flooded with anoxic groundwater and the heating was started. During this time the environment around and inside the copper tubes was humid and between 10-15°C. An indication of the amount of oxic corrosion that took place is the interior of the copper tubes as shown in the images of the copper tubes on pages 81-82 in the report TR-20-14, i.e. a negligible corrosion. The bottom plate was at this time in contact with sand and groundwater that very likely was anoxic or became anoxic a short time after emplacement. Also in this case the corrosion due to dissolved oxygen would therefore be negligible.

SSM needs to investigate this claim by SKB.

13. There is a need to further understand how oxygen in water molecules take part in reactions with a copper surface even when oxygen is available

MKG is of the understanding that it is often not understood that even when there is oxygen available in water the oxygen in the water molecules take part in the corrosion process on certain metal surfaces, including copper. It is necessary to study corrosion with isotopically marked oxygen in the water to see that the oxygen in the corrosion products comes also from the water. There is an experiment by Hultquist et al (1994) that shows this, albeit at higher temperatures than in a repository environment. The article is enclosed as appendix 3[16]. This means that in reality corrosion in an oxic environment is more severe when water is also available.

MKG understands that research using isotopically marked water is quite expensive but encourages SSM to examine this issue further.

[16] ”The analysis of gas consumption in the reaction of Fe and Cu”, G. Hultquist et al., Corrosion Science, vol 36, no 9, pp. 1459-1471, 1994. The article is also available here: http://ww.mkg.se/uploads/Appendix_3_The_analysis_of_gas_consumption_in_the_reaction_of_Fe_and_Cu_Hultquist_et_al_Corrosion_Science_vol_36_no_9_pp_1459-1471_1994.pdf

14. There is a need to have more details of the oxic corrosion inside the copper tubes and how it compares to the anoxic corrosion of the copper surface outside

MKG finds it interesting that the corrosion on the inside of the copper tubes in the central part that SKB has examined in TR-20-14 appear to be less corroded that the outer part that has been exposed to anoxic water. After all, the inside of the tubes have been exposed to air for 20 years. See the images of the copper tubes on pages 81-82 in the report TR-20-14.

MKG understands that a possible explanation for this is that the heaters in the bottom part of the tube has caused the air to be very dry. Without the availability of water molecules, the corrosion of copper in air is relatively slow.

MKG finds it interesting to see that the corrosion inside the tubes is higher at the bottom of the tubes. See the images on page 23 in the report TR-20-14. A possible explanation for this is that there has been some water at the bottom of the tubes from the beginning and the corrosion has taken place before the heater has been able to dry out the tube completely.

MKG thinks it would be useful if SSM follows up on this issue.

15. Some final general remarks

This input has been written entirely with expertise from within MKG. In the writing of the input, there has been no interaction with or input from researchers at the Royal Institute of Technology (KTH) in Stockholm. When the report TR-20-14 was published MKG discussed the contents with Peter Szakálos at KTH, but only with the purpose of understanding the report better. MKG wants to ensure that SSM can continue to use the researchers Christofer Leygraf and Peter Szakálos as experts for the authority and encourages SSM to do so.

MKG as an organisation has no possibility of influencing the nuclear waste company SKB to provide any of the information that is described above. SSM is the only actor in Sweden that can do this, and MKG therefore has to rely on the authority to act. It is of utmost importance that all uncertainties are removed regarding the risks for the long-term integrity of the copper canisters in a repository environment before the Swedish government can make a decision on the permissibility of the licence application for repository for spent nuclear fuel in Forsmark.

Best regards,

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