

The most important comments to the SKB LOT-report TR-20-14

There are two major problems with the SKB report TR-20-14 [1].

i) SKB has excluded scientific facts concerning microbial activity in the ground water and used flawed thermodynamics when they claim that the partially severe copper corrosion in the LOT-project can be explained by oxygen gas from the air.

ii) SKB has omitted to study the most corroded parts of the central copper tubes and the bottom plates.

i. Consumption of dissolved O₂ in anoxic and anaerobic groundwater

All groundwater is anoxic, i.e. without dissolved oxygen gas and it always contains microbial activity that keeps the environment anaerobic. There are detailed studies performed in Äspö hard rock laboratory, for instance the REX-project [2] that shows that added or trapped oxygen is consumed within days by microbial activity or as soon as the studied system or experimental set-up is filled with natural ground water. A full-sized repository tunnel reaches an anoxic state within a few month [3]. Any added oxygen represents fast energy for the microbes in the groundwater, as explained by prof. K. Pedersen.

SKB filled the quite small LOT A3 and S3 test volumes directly with pressurized groundwater via titanium tubes. Therefore, it can be concluded that an anoxic state occurred instantly, especially when considering the 20-year time frame of the LOT-project. It is impossible that the injected natural groundwater via the titanium tubes could have contained dissolved oxygen as SKB implies on page 63. The statement on page 71 has as also no scientific support and is obviously wrong with today's knowledge: *“Estimates of the initially available volume of air in macroscopic gaps and porous materials in the test parcels, were consistent with the interpretation that entrapped O₂ is the main oxidant behind the corrosion.”*

SKB uses the identified corrosion products as another evidence for oxid corrosion, from page 9: *“The EDS analysis indicated high levels of chlorine and the XRD showed good agreement with the diffraction pattern of Cu₂(OH)₃Cl, an expected corrosion product of copper in the presence of O₂ and chloride (Karnland et al. 2009).”* This is based on flawed thermodynamics since both copper oxides [4] and Cu₂(OH)₃Cl, paratacamite [5] are thermodynamically

stable in both anoxic and oxic environments and thus proves nothing in this case. In the potential-pH diagram in part II on page 35 in ref. [4] it is shown that both Cu_2O and CuO are stable in anoxic water. Paratacamite is for instance found naturally in anoxic groundwater/brine bearing sandstone and quarts [6].

To summarize, the detected corrosion in LOT A3 and S3 can only be explained with anoxic copper corrosion by natural groundwater.

ii. The most important metallographic evaluations are omitted

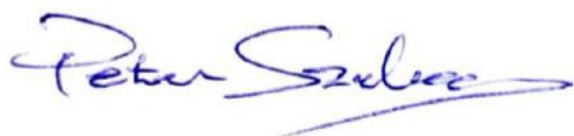
The hottest part of the central copper tubes, which are expected to corrode the most, corresponds to bentonite blocks 8-15, see Fig. 2-2 in the report. SKB has omitted to present any metallographic cross-section from the hottest part (block 8-15). Only the copper content in the bentonite, which is recalculated to some non-relevant and underestimated corrosion rates are included in the report, see Table 3-19. The highest corrosion rate from Table 3-19, only based on copper content in bentonite, corresponds to 13,8 μm during 20 years for the A3-parcel. However, this must be complemented with several metallographic-cross-sections of the central copper tube in the hot part (block 8-15). The pitting corrosion might be up to 100 μm as observed in a similar repository study with approximately the same exposure time, see the FEBEX-report [7].

The most corroded part of the copper structures are the bottom plates which were exposed directly to the anoxic groundwater. Based on the appearance of the bottom plate from parcel A3, see Figure 3-1 to the left and compare it with the bottom plate from the S3-parcel to the right, it can be estimated that the deepest pits may be in the range of 0.5-1 mm on the A3-parcel which were heated to around 80°C, see Fig. 1-3 to the right. It is of paramount importance to make several metallographic cross-sections of the bottom plates and measure the variation in total thickness. No cleaning of the corrosion products should be done since SKB argues in the report that the removal of the corrosion products might create up to 50-60 μm deep pits, see page 68. This is obviously a wrong statement if a correct cleaning process has been performed.

Considering the depth of the pits observed it should be remembered that the current LOT-experiments were performed without any radiation. In view of previous experience [8] one can expect that the extent of localized corrosion is accelerated further in the presence of γ -radiation. Hence, it is impossible to predict the actual corrosion effects in the LOT-experiments if γ -radiation would have been present.

Concluding remark

This LOT-study shows, under all circumstances, that the anoxic copper corrosion rate in Swedish groundwater is catastrophic with respect to the KBS-3 model and this conclusion can be made without further considering the radiation induced corrosion (radiolysis), stress corrosion cracking and hydrogen embrittlement.



Tekn. Dr. Peter Szakálos



Prof. em. Christofer Leygraf

References

- [1] A. J. Johansson et al., “Corrosion of copper after 20 years exposure in the bentonite field tests LOT S2 and A3”, SKB TR-20-14 (2020).
- [2] I. Puigdomenech et al., “O₂ depletion in granitic media: The REX project” SKB TR-01-05 (2001).
- [3] Müller H. et al., “Implementation of the full-scale emplacement (FE) experiment at the Mont Terri rock laboratory. Swiss J. Geosci. 110:287–306 (2017).
- [4] P. Szakalos and C. Leygraf, Part II, Technical note SSM-2019:22 (2019).
- [5] P. Szakalos and S. Seetharaman.”, Corrosion of Copper Canister” Page 13 in Technical note SSM 2012-17 (2012).
- [6] Y. Cao et al., “Paratacamite from Gypsum Veins in Sandstones of the Kuqa Basin, Xinjiang, China: Implications for a New Epigenetic Cu Enrichment Mechanism”, Resour. Geol. Vol. 66, No. 2: 114–126 (2015)
- [7] Wersin P. and Kober F, FEBEX-DP, Metal Corrosion and Iron-Bentonite Interaction Studies. Arbeitsbericht NAB 16-16 (2017).
- [8] Åsa Björkbacka, Radiation induced corrosion of copper, Doctoral Thesis, KTH Royal Institute of Technology, Stockholm, Sweden, ISBN 978-91-7595-710-4 (2015).