Note:
This is a translation of the ESK discussion paper entitled
“Diskussionspapier zur Kontroverse um die Verwendung kupferbeschichteter Behälter für die Endlagerung hochradioaktiver Abfälle”
In case of discrepancies between the English translation and the German original, the original shall prevail.

E S K

DISCUSSION PAPER of the Nuclear Waste Management Commission (ESK)

Discussion paper on the controversy regarding the use of copper-coated canisters for the disposal of high-level radioactive waste

CONTENTS

1 Introduction

Within the framework of the ongoing site selection procedure in Germany, different disposal concepts in the host rocks rock salt, claystone and crystalline rock are being discussed. In some of the concepts relevant for Germany, copper-coated or copper-encapsulated canisters could play a role in addition to other internationally discussed container concepts (see also [1]). Some time ago, Swedish scientists had questioned the corrosion resistance of copper coatings as envisaged in the Scandinavian KBS-3 concept for the disposal of spent nuclear fuel. Corresponding publications led to an international scientific controversy regarding the corrosion behaviour of copper under anaerobic conditions. This was the reason for the ESK to deal with this topic. The present discussion paper outlines the relevant processes of copper behaviour under disposal conditions, the findings resulting from the controversial discussions and possible consequences for the German site selection procedure.

2 Consultations

At its 76th meeting on 16/17 May 2019, the ESK established the ad hoc working group on copper canisters (Arbeitsgruppe KUPFERBEHÄLTER – AG KUBE). The members were asked to list the problem areas and relevant issues with regard to the controversy about the use of copper-coated canisters for the disposal of high-level radioactive waste, to reconcile with the ESK and to prepare a discussion paper on this basis. At the 77th ESK meeting on 5 September 2019, the ad hoc working group presented a respective concept. The ESK agreed to address the following issues in the paper:

1. Can, according to the current state of the art in science and technology, copper be regarded as a material that is thermodynamically stable in contact with pure, oxygen-free water and for which corrosion under these conditions is negligible?

2. How does the ESK assess the relevance of kinetically driven copper corrosion in sulphide-bearing groundwaters for the safety case of a disposal facility? What implications does this have for the German site selection procedure?

3. Can the conclusions and assumptions made for the Swedish safety concept be applied to the boundary conditions of the German selection procedure? Which further investigations/developments might be necessary to render the Swedish conclusions applicable for the German selection procedure?

Mr Bo Strömberg from the Swedish Radiation Safety Authority (SSM) was invited to give a presentation on this topic at the 74th meeting of the ESK Committee on FINAL DISPOSAL (EL) on 19 June 2020. The aspects resulting from the presentation and the subsequent discussions have been taken into account within the discussion paper.

3 Significance of the canister material for the long-term safety of a disposal facility for high-level radioactive waste

The requirements for a container for high-level radioactive waste are described in a recommendation paper published by the Nuclear Waste Management Commission [2]. They are based on the requirements resulting from the disposal concept in a specific host rock. In rock salt as a host rock, the container has to meet those requirements related to the safe operation of the disposal facility, to the possibility of waste retrieval required by law, and to the facilitation of recovery [3]. In the host rock claystone, the container assumes the function of containing the radioactive waste for a period of several thousand years after emplacement and closure of the disposal facility. For the period thereafter, containment is to be provided by the host rock and the geotechnical barrier [4].

In the concepts envisaged in Sweden, Finland and Canada for the disposal of spent nuclear fuel in crystalline host rock, the container in combination with the backfill material (“buffer”) has to fulfil a significantly longer-term protective function [5]. Swellable bentonite is used as buffer to limit advective groundwater transport in the fractured rock and to protect the corrosion-resistant canister. The design life time of the canister is at least 100,000 years [5].

In the Scandinavian KBS-3 concept, it is planned to dispose spent nuclear fuel in a cast iron canister encapsulated in copper. The thick-walled cast iron canister assumes the function of mechanical stabilisation. According to the current plans a 5 cm thick copper coating serves as a corrosion protection layer. However, “cold spray” coatings are also being discussed internationally where the thickness of the copper layer is only a few millimetres [6]. In combination with a buffer consisting of swellable bentonite, the copper-coated canister forms the essential barrier component that ensures the containment of the radioactive waste. According to [5], a number of factors contribute to the long-term stability of this barrier system in a disposal facility in crystalline host rock:

- a limited diffusive mass transport from and to the canister surface through the compacted bentonite,
- suppression of microbial activities in the nanopore structure of the compacted bentonite surrounding the canister,
- reducing geochemical (anaerobic) conditions in the near field,
- low rates for radiolysis product formation at the surface of the thick-walled canister,
- thermodynamic stability of copper in oxygen-free aqueous solution (in the absence of sulphide),
- limited availability of sulphide in the bentonite and surrounding groundwater,
- the absence of rapid pitting corrosion and stress corrosion cracking processes, and
- structural stability of the thick-walled canister.

The use of a copper-coated canister for high-level radioactive waste can also be suitable for disposal concepts in host rocks other than crystalline rock. Pressure buildup due to hydrogen production during anaerobic iron corrosion is occasionally discussed as a possible threat to the integrity of geotechnical and geological barriers (see e.g. [7]). If the gas pressure reaches the range of the overlying lithostatic pressure, fluid pathways may form. That is why e.g. NAGRA, investigates as well copper-coated canisters as an alternative to the Swiss reference canister concept (forged carbon steel)[8]. In a disposal facility in claystone, the use of copper-coated canisters could significantly reduce gas formation and thus the buildup of relevant pressures (see e.g. [9]).

Note that in various countries (Czech Republic, Russia, People's Republic of China) respective reference disposal concepts in crystalline host rock do not consider copper-coated canisters. The Czech organisation for radioactive waste management SÚRAO, for example, currently also discusses double-walled canisters consisting of an inner structure of stainless steel surrounded by an outer shell of carbon steel [10]. The outer shell is supposed to take over the function of a corrosion allowance to protect the inner part of the canister for a period of 10,000 years. This system is regarded as a “corrosion acceptable concept”, whereby the expected corrosion rates (in the µm/a range) are significantly higher than those known for a corrosion resistance concept, e.g. using a copper-coated canister (in the nm/a range). In the following, the focus is on the consideration of a corrosion resistance concept with a copper-coated canister.

4 Copper controversy

A group of researchers led by Gunnar Hultquist at the Royal Institute of Technology (KTH) in Stockholm published a study in 1986 in which they question the assumption that copper is thermodynamically stable under anaerobic conditions [11]. This then developed into a scientific controversy persisting until today, addressing not only anaerobic copper corrosion but also other corrosion processes. The Swedish nuclear fuel and waste management company SKB took up this topic within the framework of an extensive experimental and theoretical research programme with the participation of various universities [12]. In 2017, the Swedish National Council for Nuclear Waste (SNC) recommended in its statement that the critical aspects of copper corrosion should continue to be addressed in research programmes. In its statement to the Swedish Government in January 2018, the Swedish Land and Environment Court also calls for further investigations into the long-term stability of the disposal containers [13]. In particular, this concerns the following aspects:

- copper corrosion in oxygen-free water,
- pitting corrosion, which can be caused by the impact of sulphide in groundwater and by the so-called “sauna effect”[1] [14, 15, 16],
- stress corrosion cracking processes due to reaction with sulphide also in connection with the “sauna effect”,
- hydrogen embrittlement, and
- the effect of radiation on corrosion processes and hydrogen embrittlement.

The Swedish nuclear licensing authority (SSM) did not express any fundamental concerns about copper corrosion under anaerobic conditions in its recommendation on SKB's licence application [17].

SKB commented on the questions of the Swedish Land and Environment Court in the form of a report with supplementary information on aspects of canister integrity [12] and explains that anaerobic copper corrosion is not considered to be of high relevance since even at the high reaction rates postulated by the KTH scientists, copper corrosion of only about 1 mm can be expected over the period of 1 million years. The SSM assesses the SKB report positively in its statement [18]. In its assessment of the SKB's research and development

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[1] The “sauna effect” represents a scenario where during the phase in which elevated temperatures prevail (up to 90 °C at the canister surface) and the bentonite barrier is not yet saturated. Under such conditions a concentrated brine solution could occur at the canister surface, possibly enhancing copper corrosion.

programme from 2019, the SNC again makes recommendations regarding research on the topic of canister corrosion [19]. SNC comments relate to specific questions on aspects of copper corrosion by sulphide, radiation-induced corrosion and stress corrosion cracking. Interestingly, the SNC no longer addresses the problem of anaerobic copper corrosion in its latest recommendations and thus follows the assessment by the SSM.

The Swedish government's final decision on the repository project is expected to be made in 2022.

5 Mechanisms of copper corrosion

Within the framework of the Swedish safety assessments, a number of processes have been considered and evaluated, as described above, which could potentially affect the stability of the copper layer of a disposal container through corrosion. They can be assigned to different phases of repository evolution (see Fig. 1) [20, 21, 22].

1 Aerobic corrosion before emplacement in the repository.

2 Aerobic corrosion including stress corrosion cracking in the unsaturated geotechnical barrier (bentonite) in the presence of residual oxygen, which will be consumed during a period of several decades to centuries after emplacement.

3 Anaerobic corrosion including stress corrosion cracking in unsaturated bentonite at elevated temperatures (up to 90 °C at the canister surface) especially due to the influence of increasing salt concentrations by the so-called “sauna effect” over a period of several hundred to thousand years after emplacement [16].

4 Anaerobic corrosion by exposure to sulphide (surface corrosion, pitting corrosion, microbial influence) over the entire period of anaerobic conditions [23 , 24].

5 Radiation effects (until decay of the most important fission products) [25].

Other processes investigated were the influence of electromagnetic stray fields from high-voltage power lines [26] and mechanical changes in the copper material (hydrogen embrittlement and cold deformation).

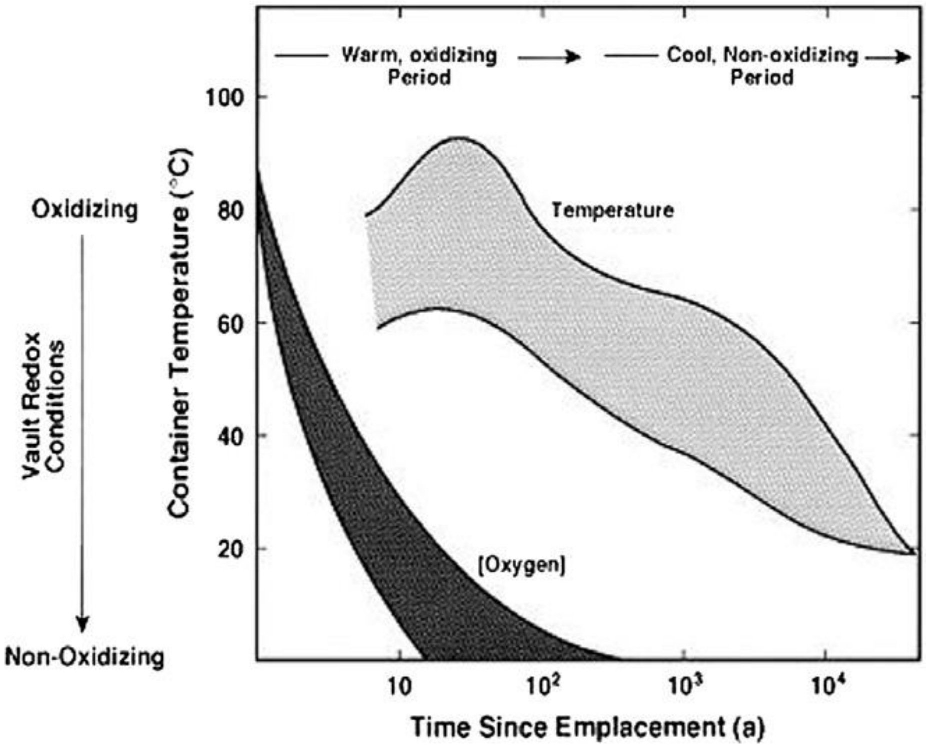


Fig. 1: Schematic illustration of the evolution of physical-chemical conditions in the emplacement area. The figure shows the transition from initially warm and oxidising conditions to cool non-oxidising (anaerobic) conditions (from [20]).

The discussion paper focuses on the following two aspects:

- anaerobic copper corrosion, as it has been a central issue of the "copper controversy", and
- sulphide-induced corrosion, which is generally recognised as a relevant mechanism of long-term copper corrosion under disposal conditions.

5.1 Corrosion in contact with oxygen-free water

Based on the standard model of thermodynamics, the system Cu-H_2O can be described via the equilibrium

$$2\,Cu \ + \ H_2O \quad \rightleftharpoons \ Cu_2O \ + \ H_2$$

The standard Gibbs energy of reaction is calculated as follows:

$$\Delta G° = \ \Delta_f G° \, (Cu_2O_{(cr)}) \ + \ \Delta_f G° \, (H_2) \ - \ \Delta_f G° \, (H_2O) \ - 2 \ \Delta_f G° \, (Cu_{(cr)})$$

Using the standard Gibbs energies of formation $\Delta_f G°$ for the individual species (see Table 1), the standard Gibbs energy of reaction is $\Delta G° = 91,1$ kJ/mol.

Table 1: Standard Gibbs energies of formation $\Delta_f G°$ for the Cu-H_2O and Cu-H_2S system [27].

Species	**$\Delta_f G°$ [kJ/mol]**
Cu^{2+} (aq)	65.5
Cu^{+} (aq)	50.0
Cu_2O (cr)	-146.0
CuO (cr)	-129.7
Cu_2S (cr)	-86.2
CuS (cr)	-53.6
$CuSO_4$ (cr)	-662.2
Cu (cr)	0
HS^- (aq)	-16.3
H^+ (aq)	0
H_2 (g)	0
H_2O (l)	-237.1
OH^- (aq)	-157.2

At equilibrium: $\Delta G° = -RT \ln K°$ (R=8.314471 J/mol K; T= absolute temperature)

with $$K^0 = \frac{a_{H_2} \cdot a_{Cu_2O}}{a_{Cu}^2 \cdot a_{H_2O}},$$

where $a_{Cu_2O} = 1$, $a_{Cu} = 1$ and a_{H_2O} in highly diluted systems almost assumes the value 1. As a result $\Delta G° = -RT \ln p_{H2}$ and for the equilibrium hydrogen partial pressure a value of about 10^{-16} bar is calculated for T = 298,15 K. As soon as a hydrogen partial pressure of 10^{-16} bar is reached in the system under consideration, the “corrosion reaction” stops and no further copper will react. The total amount of copper reacting by corrosion is negligibly small. By increasing the ionic strength in the system and thus reducing the water activity to values < 1, lower equilibrium partial pressures for hydrogen establish. Hence, based on the thermodynamic data, copper is stable in oxygen-free water. This is visualized in the redox potential pH diagram of the Cu-H_2O system (Fig. 2). Copper is stable under the redox conditions in a disposal facility.

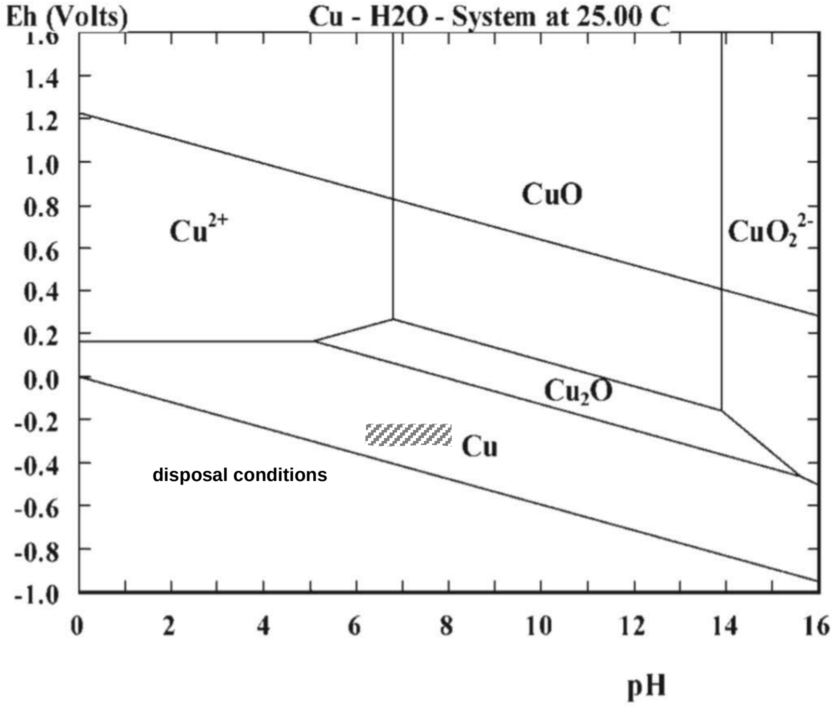


Fig. 2: Redox potential pH diagram of the Cu-H_2O system (from [5]); redox potential pH range of disposal conditions after closure and bentonite saturation from [21].

Significantly higher equilibrium hydrogen partial pressures and thus noticeable copper corrosion would only be conceivable if either another solid phase with correspondingly different standard Gibbs energies of formation or strong Cu(I) complexes were formed in the aqueous phase. The latter can be ruled out according to current knowledge and the generally accepted data [27]. A controversial discussion has developed in recent years on the formation of an alternative solid Cu phase (corrosion product).

Experimental investigations by Hultquist et al. [11] on copper corrosion showed hydrogen partial pressures in the Cu-H_2O system that were many orders of magnitude higher, up to approx. 1 mbar. The study concludes that copper corrosion takes place via a different mechanism and a copper hydroxide solid phase is formed, e.g. CuOH or copper hydroxyoxides. Such or similarly composed solid phases were postulated by the authors but could not be identified yet and thus, no standard Gibbs energy of formation could be experimentally determined for the postulated phases. There are only theoretical estimates for the Gibbs energy of reaction for such a postulated reaction.

Based on experiments commissioned by SKB [28], it could be shown that the significantly increased hydrogen partial pressures observed in experiments are very likely due to sources other than the corrosion reaction, e.g. hydrogen dissolved in the metallic copper.

The stability of copper can also be shown in sulphide-free mineralised groundwaters on the basis of the thermodynamic data. Although stable Cu complexes can form in the aqueous phase depending on the concentration of complexing anions in the groundwater, such as chloro or carbonato complexes, the corrosion resistance of copper is only marginally influenced by this. Experiments have shown that even in highly concentrated chloride solutions (4.5 mol/L), the long-term corrosion rates of copper-coated canisters are < 1 nm/a [24, 29].

5.2 Corrosion in the presence of sulphide

Sulphide reacts with elemental copper according to the equation:

$$2\,Cu(cr) + HS^- + H^+ \rightleftharpoons Cu_2S(cr) + H_2$$

Using the data from Table 1 yields a standard Gibbs energy of reaction of ΔG° = -69.9 kJ/mol. In contrast to the situation in oxygen-free water in the absence of sulphide, the reaction equilibrium is on the right side, i.e. copper is thermodynamically unstable in the presence of sulphide. Sulphide minerals such as chalcocite ($Cu_2S(cr)$), covellite ($CuS(cr)$) and djurleite ($Cu_{1,9375}S(cr)$) are formed [30]. Fig. 3 illustrates this fact for the expected redox and pH conditions in emplacement areas after closure of the repository. Not elemental copper but copper sulphide is stable.

If sparingly soluble sulphide corrosion products cover the entire copper surface, they can act as a passivation layer protecting copper against further corrosion. However, the formation of a uniform passivation layer cannot be taken as granted, and layer damage cannot be ruled out. Thus, there is a risk of pitting corrosion by which the copper coating can be severely attacked locally. Long-term predictions for corrosion rates thus become less reliable so that pitting corrosion must be excluded as far as possible. Local corrosion processes could also occur as a result of the conversion of the copper oxide layer formed under the initial oxic conditions to copper sulphide and by mechanical loads in the form of stress corrosion cracking [19]. Such reactions are discussed as possibly relevant for the initial phase before saturation of the bentonite barrier. All these processes are considered by both SKB and SSM to be of little relevance for the long-term stability of the copper shell from today's perspective. Nevertheless, questions of detailed understanding are still the subject of investigations.

Since elemental copper is not thermodynamically stable in the presence of sulphide, the lifetime of the copper shell is determined by the corrosion rate, which in turn depends on the sulphide concentration and the transport rate of the sulphide through the bentonite buffer to the canister surface. The sulphide concentrations determined in groundwaters in Sweden and Finland range between 10^{-6} and 10^{-4} mol/L and are limited by the formation of sparingly soluble iron sulphides [31]. The sulphate present in much higher concentrations can theoretically be reduced to sulphide and thus increase the sulphide concentration. This reaction, however, proceeds extremely slowly unless sulphate-reducing microbes are present [25]. Microbial activities can lead to a significant increase in the sulphide concentration in case of sufficiently high nutrient availability [23]. The bentonite buffer that tightly surrounds the canister plays a crucial role in this regard for two reasons:

1 It forms a diffusion barrier that allows only slow sulphide transport to the canister surface.

2 The swellable material fills void spaces around the canister so that only nanometre-scale pores remain that do not provide habitats for microbes.

Both functions of the bentonite barrier contribute significantly to the protection of the copper shell. Recently published experimental data assume a low corrosion rate of < 1 nm/a due to sulphide, which decreases even more with increasing reaction time over several years [24]. A recent SKB study concludes that the corrosion depth due to the influence of sulphide is less than 10 µm within 1 million years [32].

Swedish safety studies consider scenarios in which meltwater from retreating glaciers can penetrate into the emplacement area and (partially) erode away the bentonite barrier. For this case, where the bentonite barrier no longer protects the copper shell, microbially induced sulphate reduction cannot be excluded [23]. However, from the SKB's point of view [33], it can be expected that i) sulphate concentrations in low-mineralised meltwater will tend to be lower than in today's local groundwaters and ii) concentrations of dissolved organic matter that could serve as nutrients for microbes will tend to be low in intruding meltwater. Resulting sulphide concentrations should therefore be lower than today. Safety studies considering such scenarios [33], taking into account unfavourable conditions regarding fracture distributions and resulting flows as well as sulphide concentrations in groundwater, result in a failure of one out of 6,000 emplaced canisters on average due to corrosion over a period of 1 million years.

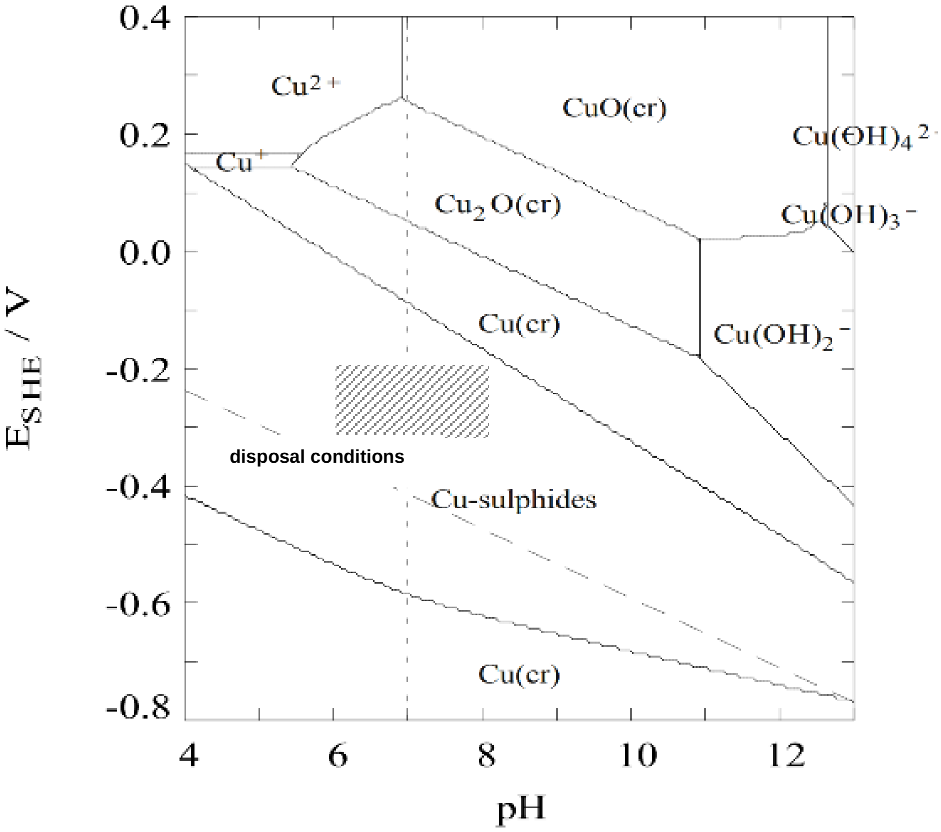


Fig. 3: Redox potential pH diagram of the Cu-H_2O-H_2S system (from [30], calculated at room temperature); redox potential pH range of disposal conditions after closure and bentonite saturation from [21].

6 Summary and conclusions for the German site selection procedure

Can, according to the current state of the art in science and technology, copper be regarded as a material that is thermodynamically stable in contact with pure, oxygen-free water and for which corrosion under these conditions is negligible?

An essential element of the KBS-3 concept is based on the corrosion resistance of copper in oxygen-free (and sulphide-free) aqueous solutions, which is derived from existing thermodynamic data and from current knowledge. Experimental findings of a group of researchers that deviated from this could be explained within the framework of a comprehensive investigation programme, and the thermodynamic stability of copper could be substantiated with experimental data. The dissent among the experts on the issue of anaerobic copper corrosion has not been completely resolved but the SNC no longer mentions this topic in its questions to SKB. In the opinion of the ESK, thermodynamic stability of copper in aqueous solutions under anaerobic conditions can be assumed according to the current state of knowledge.

How does the ESK assess the relevance of kinetically controlled copper corrosion in sulphide-bearing groundwaters for the safety case of a disposal facility? What implications does this have for the German site selection procedure?

Sulphide-induced copper corrosion is a multifaceted phenomenon, which, according to the current state of knowledge, does not enhance corrosion rates above the permissible values under the boundary conditions relevant in Scandinavia. But there are still open questions. With the available thermodynamic data, the corrosion of copper can be calculated as a function of the sulphide concentration in the groundwater in relation to the geochemical conditions. The rate of sulphide transport through the bentonite buffer thus determines the corrosion rate of the canister, which, according to Scandinavian and Canadian safety assessments, is in the range of nm/a and does not endanger the integrity of the canister over a period of 1 million years. However, other coupled processes – such as microbiological activities at the copper/bentonite interface (sulphate reduction), the influence of sulphide on pitting corrosion, stress corrosion cracking – determine the long-term stability of the copper. There is also still a need for research on some of these processes (see e.g. also [34]).

In principle, all aspects considered relevant in Scandinavian safety assessments are to be considered in the site selection procedure in Germany as well:

- What are the sulphide/sulphate concentrations in the groundwaters?
- What microbiological boundary conditions are to be expected (microbial activity, presence of organic nutrients)?
- What is the degree of fracturing in the crystalline rock under investigation and how is the hydraulic regime of the site?

- How should the effects of a scenario be assessed under which low-mineralised meltwater from retreating glaciers can penetrate the emplacement area and erode the bentonite barrier protecting the copper layer against sulphide corrosion?

Can the conclusions and assumptions made for the Swedish safety concept be applied to the boundary conditions of the German selection procedure? Which further investigations/developments might be necessary to render the Swedish conclusions applicable to the German selection procedure?

The findings on scenarios obtained in Scandinavian safety assessments are to be checked for their applicability to German sites in crystalline rock and for their compatibility with German safety requirements. This also applies to possible damage to the disposal container that is not due to biogeochemical corrosion but is caused, for example, by mechanical shearing due to postglacial earthquakes or isostatic loads.

The Swedish regulations (SSMFS 2008:21 [35]) require graded safety analyses for specific time periods. The primary basis for assessing safety in connection with disposal is provided by quantitative dose and risk calculations for the period of 1,000 years following the closure of the repository [36]. For the period thereafter, various possible evolution scenarios are to be considered. An analysis is required for the time period for which the maintenance of the barrier function is to be ensured, but at least for 10,000 years. However, safety analyses should also consider scenarios covering the time period of the expected next glacial cycle, i.e. 100,000 years. The outcome of radionuclide transport calculations over significantly longer periods (up to 1 million years) resulting in values for radionuclide fluxes, radionuclide concentrations in the biosphere or dose values for the population can be used as additional safety indicators for the safety assessment. This time-graded approach is justified by the fact that statements are associated with greater uncertainties the further they extend into the future. In a similar way, the Finnish waste management organisation POSIVA also structures its safety analyses according to time periods [37]. The initial focus is on the containment function of the canister-buffer system over a period of 10,000 years. The entire analysis, however, covers a period of 1 million years.

The German Site Selection Act (StandAG, [38]) does not introduce such a gradation, but states 1 million years as the assessment period. This results in uncertainty as to the extent to which a safety case for a disposal container according to Swedish regulatory requirements can satisfy the requirements applicable in Germany (see also [39]). However, this problem only exists when considering the use of a copper-coated canister in a disposal facility in crystalline rock. In contrast to other host rocks, the containment function of the disposal container (in combination with the bentonite buffer) must be demonstrated as a main barrier over the entire assessment period, unless a containment-providing rock zone can be identified. This means that the main burden of ensuring safe containment of the waste over a period of 1 million years is on the engineered/geotechnical barrier combination as stipulated in the safety requirements applicable in Germany (see Disposal Facility Safety Requirements Ordinance EndlSiAnfV § 4 (3, 5 [40]). For a repository in other host rocks (claystone, rock salt), the container is not considered as a main barrier. In those cases, the containment-providing rock zone consisting of a geological barrier in interaction with engineered and geotechnical seals is considered as main barrier. Waste containment by the disposal container is required for significantly shorter periods of time.

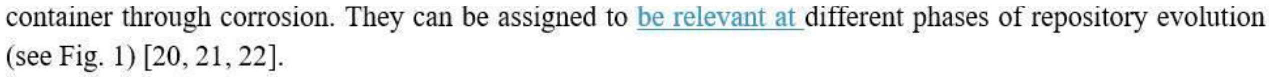
Scandinavian safety analyses come to the conclusion that when considering scenarios in which partial erosion of the bentonite barrier is postulated, taking into account unfavourable conditions, one canister failure due to corrosion can be expected on average in a period of 1 million years. Corresponding studies on the relevant boundary conditions of an investigation area in crystalline rock would also have to be carried out in Germany. The extent to which the resulting radionuclide release rates fulfil the requirement of the German safety requirements [40] in terms of the safe containment of radioactive waste is to be examined.

The results of the relevant investigations on (copper)corrosion of disposal containers in Sweden, Finland and Canada are to be reviewed for their respective applicability to the geochemical boundary conditions of German sites. In this context it should be noted that container durability depends on a variety of geological and geochemical conditions. A comparison of the geological situation (e.g. fracturing) and groundwater compositions at sites in crystalline rock in Germany with those in Scandinavia would therefore be necessary. According to [41], with a few exceptions, most deep waters in Germany are highly mineralised and thus may have significantly higher chloride contents than at the sites investigated in Scandinavia and could have a corrosive effect. pH ranges of groundwaters in German crystalline rock formations are between 6.6 and 11.5. In recent studies, however, only low copper corrosion rates were found even in saturated NaCl solutions (see discussion above).

The use of a copper-coated canister for high-level radioactive waste can also be suitable for disposal concepts in host rocks other than crystalline rock. Pressure build-up due to hydrogen produced during iron corrosion is discussed in various disposal concepts as a possible threat to the integrity of geotechnical and geological barriers (see e.g. [7]). In a disposal facility in claystone, the use of copper-coated canisters could significantly reduce gas formation and thus the build-up of relevant pressures (see e.g. [9]). As an alternative to the Swiss reference container concept (use of forged carbon steel), NAGRA therefore also carries out investigations on copper-coated canisters [8].

The safety concept for the disposal in rock salt consists of preventing solutions from entering the emplacement area of the disposal containers. The advantage of a copper-coated canister over a thick-walled structural steel container is therefore not obvious. Even if postulating the intrusion of limited brine volumes, such scenarios would still have to be considered in which corrosion-promoting sulphide is formed in concentrated brines by thermal and microbial sulphate reduction in the presence of natural hydrocarbons (see [42]). Thus, in rock salt as host rock, a copper-coated canister does not necessarily lead to an additional increase of safety.

7 References

[1] 

container through corrosion. They can be assigned to be relevant at different phases of repository evolution (see Fig. 1) [20, 21, 22].

, C. Herold, S. Prignitz, T. Orellana Pérez, A. Wunderlich, H. Völzke, D. Wolff, ABSCHLUSSBERICHT zum FuE-Verbundvorhaben KoBrA, Anforderungen und Konzepte für Behälter zur Endlagerung von Wärme entwickelnden radioaktiven Abfällen und ausgedienten Brennelementen in Steinsalz, Tonstein und Kristallingestein, 2020, BGE TEC 2020-19, BAM 3.4/3205-1

[2] EMPFEHLUNG der Entsorgungskommission: Anforderungen an Endlagergebinde zur Endlagerung Wärme entwickelnder radioaktiver Abfälle, ESK, 20.01.2017

[3] J. Mönig, D. Buhmann, A. Rübel, J. Wolf, B. Baltes, K. Fischer-Appelt, Sicherheits- und Nachweiskonzept; Bericht zum Arbeitspaket 4, Vorläufige Sicherheitsanalyse für den Standort Gorleben, Juni 2012

[4] Safety Options Report - Post-Closure Part (DOS-AF), 2016, FRENCH NATIONAL RADIOACTIVE WASTE MANAGEMENT AGENCY, ANDRA, CG-TE-D-NTE-AMOA-SR2-0000-15-0062/A

[5] F. King, C. Lilja, Scientific basis for corrosion of copper in water and implications for canister lifetimes Corrosion Engineering, Science and Technology 2011, 46, 153-158

[6] J.R. Scully, D. Féron, H. Hänninen, Review of the NWMO Copper Corrosion Program, Nuclear Waste Management Organization, Canada, NWMO-TR-2016-11, August 2016

[7] A. Rübel, J. Mönig, Gase in Endlagern, GRS 242, November 2008

[8] N. Diomidis, L.H. Johnson, P. Bastid, C. Allen, Design development of a copper-coated canister for the disposal of spent fuel in a deep geological repository in Opalinus Clay, Corrosion Engineering Science and Technology, 2017, 52, 31–39

[9] Effects of Post-disposal Gas Generation in a Repository for Spent Fuel, High-Level Waste and Long-lived Intermediate Level Waste Sited in Opaliounus Clay, NAGRA Technical Report, NTB 04-06, July 2004

[10] I. Pospiskova, D. Dobrev, M. Kouril, J. Stoulil, D. Novikova, P. Kotnour, O. Matal, Czech national programme and disposal canister concept, CORROSION ENGINEERING, SCIENCE AND TECHNOLOGY, 2017, 52 (S1), 6–10

[11] G. Hultquist, Hydrogen evolution in corrosion of copper in pure water, 1986, Corros. Sci., 26, 173–177.

[12] Supplementary information on canister integrity issues; SKB Technical Report TR-19-15, March 2019

[13] http://www.mkg.se/en/translation-into-english-of-the-swedish-environmental-court-s-opinion-on-the-final-repository-for-sp

[14] O. Karnland, T. Sandén, L.E. Johannesson, T.E. Eriksen, M. Jansson, S. Wold, K. Pedersen,M. Motamedi, B. Rosborg, Long term test of buffer material, Final report on the pilot parcels. SKB Technical Report, TR-00-22, 2000

[15] P. Szakálos, C. Leygraf, Review of SKB‘s Supplementary Information on copper canister integrity issues, TECHNICAL NOTE Date: 2019-09-19, SSM2019-2484-8

[16] M. Birgersson, R. Goudarzi, Summary report on “sauna” effects, SKB TR-17-07, August 2017

[17] SSM 2018, Yttrande över ansökningar om tillstånd till anläggningar för slutligt omhändertagande av använt kärnbränsle, SSM2011-1135-23, Strålsäkerhetsmyndigheten

[18] SSM 2019, Remiss av Svensk Kärnbränslehantering AB:s kompletterande yttrande i ärende om tillståndsprövning enligt lagen (1984:3) om kärnteknisk verksamhet av anläggningar i ett sammanhängande system för slutförvaring av använt kärnbränsle och kärnavfall, SSM2019-3168-7, Strålsäkerhetsmyndigheten

[19] The Swedish National Council for Nuclear Waste's Review of SKB's RD&D Programme 2019, Swedish National Council for Nuclear Waste, SOU 2020:39

[20] F. King, M. Kolar, P. Maak, Reactive-transport model for the prediction of the uniform corrosion behaviour of copper used fuel containers, Journal of Nuclear Materials 379 (2008) 133–141

[21] F. King, L. Ahonen, C. Taxén, U. Vuorinen, L. Werme, Copper corrosion under expected conditions in a deep geologic repository, SKB Technical Report, TR-01-23, 2001

[22] Bo Strömberg, Lena Sonnerfelt, and Henrik Öberg, Exploratory what-if analysis of some debated canister failure modes in the review of a licence application for the construction and operation of a spent nuclear fuel repository in Sweden, Adv. Geosci., 49, 67–75, 2019

[23] A. Gordon, H. Pahverk, E. Börjesson, L. Sjögren, Corrosion morphology of copper in anoxic sulphide environments, SKB TR-18-14, December 2018

[24] N. A. Senior, T. Martino, J. Binns, P. Keech, The anoxic corrosion behaviour of copper in the presence of chloride and sulphide, Materials and Corrosion. 2021, 72, 282–292

[25] F. King, C. Lilja, K. Pedersen, P. Pitkänen, M. Vähänen, An update of the state-of-the-art report on the corrosion of copper under expected conditions in a deep geologic repository, SKB Technical Report TR-10-67, December 2010

[26] C. Taxén, B. Sandberg, C. Lilja, Possible influence from stray currents from high voltage DC power transmission on copper canisters, SKB Technical Report, TR-14-15, October 2014

[27] Cox, J. D., Wagman, D. D., and Medvedev, V. A., CODATA Key Values for Thermodynamics, Hemisphere Publishing Corp., New York, 1989

[28] RD&D Programme 2019, Programme for research, development and demonstration of methods for the management and disposal of nuclear waste, SKB Technical Report TR-19-24, December 2019

[29] Lilja C, King F, Puigdomenech I, Pastina B. Speciation of copper in high chloride concentrations, in the context of corrosion of copper canisters. Materials and Corrosion. 2020;1–7

[30] I. Puigdomenech, C. Taxén, Thermodynamic data for copper - Implications for the corrosion of copper under repository conditions, SKB Technical Report TR-00-13, August 2000

[31] E-L. Tullborg, J. Smellie, A.-Ch. Nilsson, M. J. Gimeno, L.F. Auqué, V. Brüchert, J. Molinero, SR-Site – sulphide content in the groundwater at Forsmark, SKB Technical Report TR-10-39, December 2010

[31] SKB, Long-term safety for the final repository for spent nuclear fuel at Forsmark; Main report of the SR-Site project, Volume I, SKB Technical Report TR-11-01, March 2011.

[32] F. King, M. Kolář, Copper Sulfide Model (CSM) - Model improvements, sensitivity analyses, and results from the Integrated Sulfide Project inter-model comparison exercise, SKB Technical Report, TR-18-08, December 2019

[33] SKB, Long-term safety for the final repository for spent nuclear fuel at Forsmark; Main report of the SR-Site project, Volume I, SKB Technical Report TR-11-01, March 2011.

[34] D.S. Hall, M. Behazin, W.J. Binns, P.G. Keech, An evaluation of corrosion processes affecting copper-coated nuclear waste containers in a deep geological repository, Progress in Materials Science, 2021, 118, 100766

[35] Swedish Radiation Safety Authority Regulatory Code, SSMFS 2008:21

[36] Swedish Radiation Safety Authority Regulatory Code, SSMFS 2008:37

[37] Safety Case for the Disposal of Spent Nuclear Fuel at Olkiluoto, Posiva 2012-04

[38] Gesetz zur Fortentwicklung des Gesetzes zur Suche und Auswahl eines Standortes für ein Endlager für Wärme entwickelnde radioaktive Abfälle und anderer Gesetze (StandAG), Bundesgesetzblatt Jahrgang 2017 Teil I, Nr. 26, 5. Mai, 2017

[39] Sicherheitskonzeptionelle Anforderungen an das Barrierensystem eines Endlagers für hoch radioaktive Abfälle und deren Umsetzbarkeit, STELLUNGNAHME der Entsorgungskommission, 21.02.2019

[40] Endlagersicherheitsanforderungsverordnung vom 6. Oktober 2020 (BGBl. I S. 2094)

[41] Referenzdatensätze zur Anwendung der geowissenschaftlichen Abwägungskriterien im Rahmen von § 13 StandAG, Grundlagen, BGE, Stand 01.09.2020

[42] G. Bracke, T. Popp, W. Püttmann, B. Kienzler, A. Lommerzheim, H.C. Moog, Berücksichtigung der Kohlenwasserstoffvorkommen in Gorleben, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS 285, August 2012