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Methodology to make a robust estimation of the carbon steel overpack lifetime with respect to the Belgian Supercontainer design

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ABSTRACT

The Supercontainer (SC) design is the preferred Belgian option for the final disposal of vitrified high-level waste (VHLW) and spent fuel (SF) in deep underground clay layers. The SC consists of a carbon steel overpack, containing VHLW canisters or SF assemblies, surrounded by a thick concrete buffer, which in turn, is entirely encased in a stainless steel envelope. An integrated R&D strategy is developed to demonstrate and defend that the integrity of the carbon steel overpack can be ensured at least during the thermal phase. This integrated approach, proposed to estimate the lifetime of the carbon steel overpack, consists of three steps: lifetime prediction, validation, and confidence building. Under the predicted conditions within the SC (highly alkaline concrete buffer), the carbon steel overpack is expected to undergo uniform corrosion (passive dissolution). The methodology exists in demonstrating that corrosion forms other than uniform corrosion (e.g. localised corrosion such as pitting corrosion, crevice corrosion and stress corrosion cracking) cannot occur ('exclusion principle'). This paper elaborates on how this methodology is implemented.

1. Introduction

Among the options considered for dealing with long-lived radioactive waste (vitrified high-level waste – VHLW – and spent fuel – SF), geological disposal (after a period of storage on the surface to allow gradual decay of both the temperature and radioactivity of the wastes) is the one recommended at the international level and which is scientifically and technically feasible for ensuring a maximal protection of the people and its environment for both the current and future generations. The Belgian radioactive waste management organisation, NIRAS/ONDRAF, is committed to the challenge of developing a concept and design of a disposal facility, and developing the evidence and arguments to prove that such a facility can be constructed in a safe, technically feasible and economically achievable manner, without neglecting the societal aspects.

In geological disposal, ensuring complete containment of the radioactivity at least during the thermal phase, i.e. the period during which the temperature of the host rock is expected to lie above the range of temperatures within which nominal radionuclide migration properties can be relied upon, is essential in providing long-term safety. This primary prerequisite is imposed to avoid the necessity to model contaminant transport at least during the period when the heat output from the wastes is high. According to NIRAS/ONDRAF specifications, the thermal phase is assumed to last for at least hundreds of years for VHLW and, possibly up to a few thousand years for SF, after emplacement of the wastes in the repository.

The planning and implementation of any geological repository is a lengthy process, involving an incremental, step-wise approach. This approach consists of different successive stages including the phase of methodological R&D, the pre-project phase, the construction phase, the operational phase, the phase of repository closure, and the potentially phase of institutional control, surveillance and maintenance. During each of these stages, knowledge is progressively developed and the design of the disposal system is continuously evaluated and improved on the basis of state-of-the-art scientific knowledge and technical developments. The Belgian programme on the development and the implementation of a geological repository for the disposal of VHLW and SF is, at present, in a phase of progress that is described as 'methodological R&D' [1].

The methodological R&D studies were initiated by the Belgian Nuclear Research Centre, SCK•CEN, and have been going on since the 1970s. These studies have been focussed on the Boom Clay, located in the northeast of Belgium beneath the Mol-Dessel nuclear site at a depth between 180 m and 280 m, without however prejudging future decisions on the exact disposal site, or on the final choice of a repository concept and design. The underground research facility, HADES, which was constructed at the beginning of the 1980s at the Mol-Dessel nuclear site, has contributed significantly to these studies [1].

The state of the scientific and technical advancement was presented on a regular basis with a periodicity of approximately 10

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years under the form of preliminary safety and environmental assessments such as the SAFIR-1 [2] and the SAFIR-2 [3] reports that were published in 1989 and 2001, respectively.

To date, the current reference disposal option in Belgium lacks formal political and societal legitimacy. Geological disposal has not yet been confirmed as Belgian policy for managing ILW/VHLW and therefore both Boom Clay and the Mol-Dessel nuclear site are working hypotheses. The progressive confirmation of these hypotheses constitutes the following key objective, since a non confirmation would have a serious impact on the programme. To this end, developing and submitting a first ‘Safety and Feasibility Case’, SFC-1, by the end of 2013, is perceived as the next major scientific and technological milestone [4,5].

The SFC strongly differs from the previous SAFIR-reports in the sense that whereas the SAFIR-reports were synthesis reports telling what has been done over an arbitrarily fixed period of 10 years, the main objective of the SFC is to build a convincing case to obtain decisions from the national authorities related to construction, operation and closure of a repository [6].

2. The Belgian reference disposal design

The SAFIR-2 design, developed in the late 1980s by ONDRAF/NIRAS, was considered the reference design for the disposal of VHLW and SF in an underground repository up to the beginning of the years 2000. In the SAFIR-2 design, the disposal galleries were lined with concrete, and filled with a clay-based buffer, which surrounded a centralised stainless steel tube (the *disposal tube*). The primary waste package that was encapsulated in an individual stainless steel vessel (the *overpack*) was pushed into the disposal tube. The disposal tube consisted of several sections hermetically welded to one another. The clay-based buffer consisted of precompacted segments made from a mixture of bentonitic FoCa swelling clay (60%), sand (35%) and graphite (5%). FoCa refers to a sedimentary clay from the Paris Basin, extracted in a site near to the Vexin region (Fourges-Cahaignes).

An international peer review of the SAFIR-2 report [7], conducted under the auspices of the Nuclear Energy Agency (NEA) on behalf of the Belgian Government, recommended to rely more on the performance of the engineered barrier system (EBS) in addition to Boom Clay. This review resulted in a re-evaluation of the SAFIR-2 design, which led to the development of a ‘new’ reference disposal design, *viz.* the Supercontainer (SC). The SC is based on the Contained Environment Concept (CEC), the aim of which is to establish and preserve a favourable chemical environment in the immediate vicinity of the metallic overpack, so that it will be exposed to essentially unchanged, benign conditions for a long time, at least for the duration of the thermal phase. The contained environment is the buffer material surrounding the overpack. A key aspect to fulfill this primary aim is to ‘fix’ the immediate environment of the overpack by using a tailor-made material with well-known characteristics (e.g. high chemical buffering capacity) and enclosing it inside a metallic vessel [8].

The SC is a cylindrical container ($L \sim 4$ m; Ø ~ 2 m) made of a 6 mm thick stainless steel casing (the *envelope*). This casing comprises a 30 mm thick carbon steel *overpack*, containing either two VHLW canisters or four SF assemblies, surrounded by a thick concrete *buffer* ($\sim$700 mm). Carbon steel has been chosen for the overpack because it is a material for which a broad experience and knowledge already exists and its corrosion behaviour in a concrete environment, in particular, is well understood and favourable to meeting the requirement for overpack longevity. A Portland cement-based (OPC) concrete has been chosen for the buffer because it will provide a highly alkaline chemical environment, in which the external surface of the overpack will be passivated. Under these conditions, the carbon steel overpack is expected to undergo uniform corrosion (passive dissolution). Additional buffer functions are to provide a low hydraulic conductivity environment to slow the infiltration of external aggressive species to the overpack surface and to provide radiological shielding for the workers (thereby simplifying underground waste transportation and emplacement operations). Once fabricated, the SC will be emplaced horizontally in tunnels excavated in the Boom Clay. During excavation, the tunnels will be immediately lined with concrete wedge blocks to support the plastic clay formation and to limit the convergence of clay (i.e. the gallery walls would collapse without the use of a lining). The void space between the SC and the concrete liner will be backfilled with a cementitious material (the backfill) before the tunnels are sealed with concrete or clay plugs. Fig. 1 presents a schematic diagram of a Supercontainer for VHLW emplaced within galleries excavated in the Boom Clay [4,9–11].

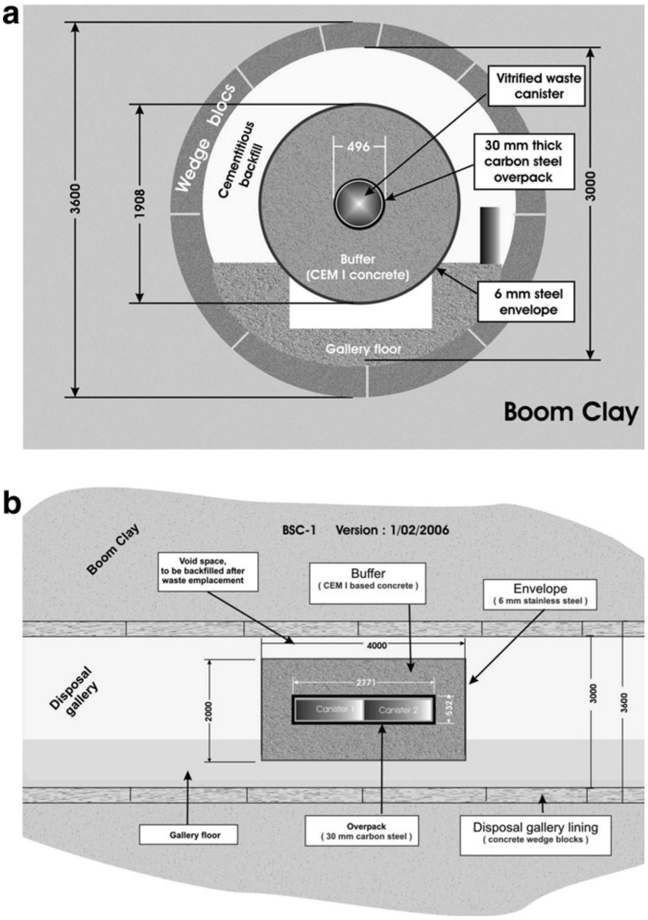

Fig. 1. Schematic diagram of the cross-section (a) and the longitudinal section (b) of a Supercontainer for VHLW emplaced in a disposal gallery [4,11].

3. Integrated approach

In view of the Safety and Feasibility Case I study (SFC I), an integrated R&D methodology is developed to demonstrate and defend that the integrity of the carbon steel overpack can be ensured at least during the thermal phase. Table 1 summarizes the methodology proposed to arrive at a robust estimation of the overpack lifetime. This integrated approach consists of three steps: lifetime prediction, validation, and confidence building.

Table 1
Schematic overview of the methodology proposed to arrive at a robust estimation of the overpack lifetime

What?	How?
Phase 1 – Lifetime prediction	
• Prediction of the CEP[a]	• Geochemical modelling based on different sets of chemical boundary conditions
• Identification of corrosion mechanisms	• Assessing remaining uncertainties (welds, radiolysis, reduced sulphur, ...) • Applying the exclusion principle
• Estimation of corrosion rates of each characteristic phase in the CEP	• Literature data • Information from specialised labs • Expert judgement • Own R&D (if no or insufficient data is available) • Interface models
• Lifetime calculation	• Integration of corrosion rates over time
Phase 2 – Validation of	
• Corrosion evolutionary path • Exclusion principle	By means of e.g. in situ testing
Phase 3 – Confidence building	
• Confidence building in results	• Peer review • Open publications • Organisation of the 4th International Workshop on the ‘Prediction of Long-Term Corrosion Behaviour in Nuclear Waste Systems’ (2010) • Organisation of the International Workshop on ‘Sulphur Assisted Corrosion in Nuclear Waste Disposal systems’ (October 2008)

[a] CEP: Corrosion Evolutionary Path.

3.1. Lifetime prediction

The lifetime of the overpack is predicted on the basis of the corrosion evolutionary path (CEP). The CEP describes the changing environmental conditions the overpack is subjected to in a repository during the different phases of, and also prior to, the disposal period. Environmental conditions surrounding the SC will change with time:

- oxidising conditions will gradually change to reducing conditions following repository closure,
- temperature will decrease as heat production of the radioactive waste decreases, and also
- the geochemistry of the environment surrounding the carbon steel overpack, which is initially governed by the OPC-based buffer chemistry, will gradually be modified as Boom Clay pore water penetrates the concrete lining of the disposal galleries and the concrete buffer material of the SC.

Fig. 2 gives a schematic presentation of the CEP, completed with the potential corrosion modes the carbon steel overpack can be susceptible to during the different phases.

The approach consists in dividing the evolutionary path in different phases (aerobic, anaerobic) and determining the ‘best estimate’ uniform corrosion rate for each of these phases. Lifetime predictions (i.e. the reduction in wall thickness in μm/year) are then estimated by integrating these corrosion rates over the duration of the different phases on the basis of the following equation:

$$d_{\text{overpack}} = \sum v_i \bullet t_i$$

with d_{overpack}: required thickness of the overpack to avoid perforation during the thermal phase; v_i: ‘best estimate’ uniform corrosion

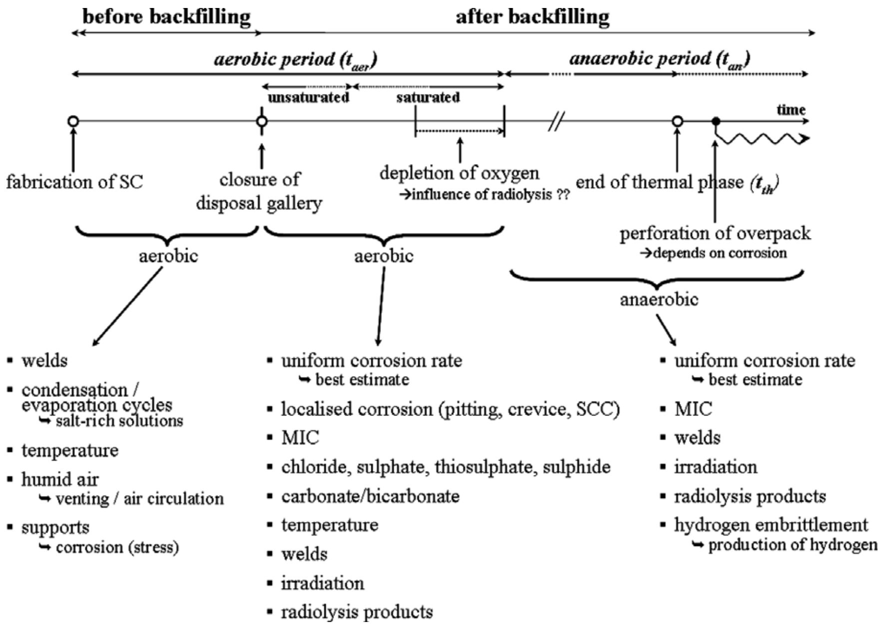

Fig. 2. Schematic of the corrosion evolutionary path (CEP), completed with the potential corrosion modes the carbon steel overpack can be susceptible to during the different phases.

rate during the different phases (aerobic, anaerobic) of the disposal period; t_i: duration of the different phases of the disposal period.

This calculation only provides an estimation of the corrosion margin on the overpack thickness. The thickness required to withstand mechanical stresses is not taken into account.

To arrive at the 'best estimate' of the uniform corrosion rate, studying the carbon steel overpack/concrete buffer interface should not be ignored. Indeed, experimental evidence [12] supports the assumption that uniform corrosion of the carbon steel overpack will decrease in time according to a parabolic law rather than proceeding linearly. Therefore, attention should be paid at developing interface models that couple geochemistry, corrosion and mass transport through the buffer.

In the high pH environment of the SC, the carbon steel overpack is covered with a protective passive film. If this passive film is destroyed locally, the occurrence of localised corrosion (pitting, crevice corrosion) and stress corrosion cracking (SCC) is plausible, which in turn increases the difficulty of predicting the lifetime of the overpack because of the stochastic nature of these local phenomena. Our proposed integrated approach is based on confirming that each corrosion mechanism, other than uniform corrosion, cannot take place under the circumstances described in the corrosion evolutionary path. This is called the 'exclusion principle'. A threshold concentration of aggressive species (e.g. chloride, thiosulfate, sulfide, etc.) exists below which carbon steel does not suffer from the different potential forms of localised corrosion and SCC. The exclusion principle is based on proving that the predicted reference concentrations that can be expected within the SC are situated well below the threshold concentrations. These threshold concentrations are found in literature or determined through a limited set of specific laboratory tests. The occurrence of localised corrosion can be predicted by comparing the critical potentials at which these corrosion modes occur (determined, if available, from literature or experimentally) with the free corrosion potential.

3.2. Validation

The data of the corrosion evolutionary path, the suitability of the 'exclusion principle' and the interface models should be validated through a limited set of well-defined tests (e.g. an *in situ* experiment).

3.3. Confidence building

An active publication policy (*a.o.* publications in peer reviewed journals), together with a periodical review of the programme by a panel of experts in the field, should help to enhance the confidence in the predicted lifetime of the SC. SCK•CEN is also planning to organise an International Workshop on 'Sulphur Assisted Corrosion in Nuclear Waste Disposal Systems' (SACNUC) in 2008 and the 4th International Workshop on 'Prediction of Long-Term Corrosion Behaviour in Nuclear Waste Systems' in 2010. This will be an excellent opportunity to focus on long-term corrosion prediction in the framework of the SC concept and to present, to an audience of peers, the results of the Belgian programme on overpack corrosion.

4. Current status

4.1. Time evolution of the near-field environment surrounding a disposal gallery

Based on our current knowledge and experience from the excavation and construction of the extension of the underground research facility, HADES, a better understanding of the time evolution of the near-field around a radioactive waste disposal gallery for the current Belgian reference design of the Supercontainer has been developed. Fig. 3 schematically illustrates, from right to left, the time evolution of the near-field environment surrounding a disposal gallery. In the early stage, excavation of the gallery creates fractures within the surrounding host rock up to about 1 m. Moreover, oxygen will come into contact with the initially anoxic Boom Clay. Mainly pyrite and organic matter, two important compounds of Boom Clay, will interact with this oxygen. Along the fractures, the oxygen can easily intrude up to the same depth of about 1 m. Because the plastic nature of Boom Clay, the fractures will seal fast. Water is continuously drained towards the gallery, but oxygen will dissolve in the pore water and continue to in-diffuse into the host formation. Neglecting the reactivity of the oxygen with remaining pyrite and organic matter, the in-diffusion will not exceed about 2 m, even after 20 years of ventilation [13]. Consequently, the extent of the oxidised zone remains limited to the first meters, while the degree of oxidation of the host formation will increase. During the early closure phase, the heat-emitting waste will cause a temperature increase, lasting for at least several hundreds to thousands of years. Meanwhile, reactions leading to an equilibrium between the high pH concrete and the surrounding host rock are commencing (alkaline plume). As this is a very slow process, these reactions will mainly continue throughout the late closure phase. A high pH within the concrete is maintained during at least 80 000 years, while in the meantime mineralogical and geochemical changes occur within the surrounding host rock. The extent of this alkaline plume within the Boom Clay is also limited to about 2.5 m after 100 000 years [14].

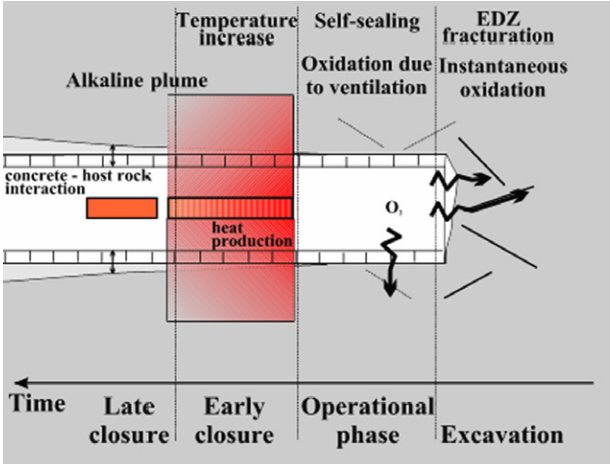

Fig. 3. Schematic of the time evolution of the near-field around a disposal gallery.

4.2. Geochemical modelling of the concrete buffer surrounding the carbon steel overpack

Scoping calculations [15] have been carried out to simulate the evolution of pH and the concentration of anionic species, some of which can be detrimental to the metallic barriers, from the Boom Clay at the carbon steel overpack and the stainless steel envelope as a function of time. The outcome of these scoping calculations is summarised in Table 2. These calculations are based on a chemical coupled reactive transport 1D radial model. The concentration evolution of the different species is determined only by diffusion, which is driven by concentration gradient. A flow-through model [15] was applied assuming a local equilibrium between Boom Clay pore water and concrete components to derive compositions of concrete pore fluids for three stages in time:

- young concrete fluid: this water is still in equilibrium with dissolved alkalis;

Table 2
Modelled pore water chemistry of the concrete buffer surrounding the overpack within the supercontainer

Time scale (yr)	Unsaturated $t = 0$	Saturated $t = 5 \sim 10$	Young concrete water $t = 1000 \sim 10000$	Evolved concrete water $t > 80000$	CSH water
[Element], mM					
Ca	0.7	0.7	0.7	15.3	0.8/1.3(a)
Na	141	141	141	15.1	15.1
K	367	367	367	0.2	0.2
Al	0.06	0.06	0.06	0.005	9.4/2.9(a)
Si(a)	0.05/0.3	0.05/0.3	0.05/0.3	$6 \times 10^{-3}/3 \times 10^{-3}$	0.8/6.3
Mg	$\sim 10^{-7}$	$\sim 10^{-7}$	$\sim 10^{-7}$	4×10^{-6}	10^{-6}
Fe	10^{-5}	10^{-5}	10^{-5}	10^{-6}	10^{-7}
C (TIC)	0.3	0.3	0.3	8×10^{-3}	0.02
SO_4^{2-}	2	2	2	7×10^{-3}	0.05
$S_2O_3^{2-}$	–	–/1.9[b]	1.9–6.4	–	–
HS^-/S^{2-}	–	–/0.15[b]	0.15–0.5	–	–
Cl^-	–	0.2/3.6[b]	0.2–12	0.2	0.2
pH	∼12[c]	∼12[c]	13.5	12.5	∼12
E_h, mV	100–200	100–200	∼−800	∼−800	∼-800

[a] Controlled by afwillite and CSH_1.8.

[b] Saturated with the reference or ‘worst-case’ water.

[c] Temperature at 80 °C of thermal phase.

- evolved concrete fluid: after leaching out of the dissolved alkalis, the pH of this water is controlled by dissolution of portlandite;
- CSH (calcium-silicate-hydrates) concrete fluid: the portlandite has completely dissolved away.

The initial pH of the concrete pore fluid is about 13.5, controlled by the dissolved alkalis (K^+ and Na^+), and decreases to 12.5, regulated by portlandite solubility, in about 1000 years. The pH 12.5 is predicted to maintain for at least 80000 years, after which it will slowly start to drop. The temperature increase (∼80 °C) during the thermal phase of the repository operation will decrease the pH to about 12, owing to the effect of temperature on hydrolysis properties of the system.

In the most conservative case, i.e. a fast ingress of the ‘worst-case’ Boom Clay pore water, a chloride concentration of 12 mM might reach the overpack in 400 years. The ‘worst-case’ Boom Clay pore water assumes that the in-diffusing Boom Clay pore water is oxidised as a result of repository excavation and operation. Taking the retardation of chloride (i.e. the binding of chloride by cement constituents), which is generally observed in a cementitious system, into account, the same chloride concentration will only reach the overpack after 8000 years.

The concentration of sulfate in the ‘worst-case’ water (∼10 mM) may reach the overpack in about 500 years considering no retardation. This is conservative as sulfate will likely interact with cement constituents. Taking ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}{:}26H_2O$) as the sulfate controlling mineral in the model system, the sulfate concentration is about 2 mM when the pH is regulated by KOH/NaOH and 7 μM if the pH is buffered by the portlandite dissolution.

The concentration of sulfide in the ‘worst-case’ water is 0.5 mM. This concentration might reach the overpack in about 500 years if no retardation of the species is considered. As mechanisms of chemical interactions between sulfide species and cementitious components were unknown at the time of the modelling exercises, these were not taken into account at this stage.

The highest thiosulfate concentration (‘worst-case’ water) is 6.4 mM, which may arrive at the overpack in about 500 years.

The concentration of inorganic carbon is controlled by the dissolution of calcite within the domain and therefore dependent on pH. When the pH is controlled by NaOH/KOH, the concentration of inorganic carbon at the overpack is about 0.4 mM. Afterwards, the concentration decreases to 8 μM when the pH is regulated by the dissolution of portlandite [15].

5. Impact on the integrity of the carbon steel overpack (from a corrosion point of view)

Modelling calculations have indicated that the concrete buffer surrounding the overpack will maintain a high pH (>12.5) during at least 80000 years [15]. Therefore, the carbon steel overpack is expected to remain in a passive state over the entire duration of the thermal phase.

During excavation and operational phase, pyrite (FeS_2) will oxidise due to the ingress of oxygen. This results in the production of sulfates and Fe-(oxi-)hydroxides. It is assumed that the dissolved sulfates will accumulate at the interface between the SC-envelope and the concrete gallery lining. This is consistent with observations made in the underground research facility, HADES, of sulfate salt precipitation on the concrete tunnel liner [16]. Sulfates might be transformed into reduced sulfur species due to the presence of sulfate-reducing bacteria (SRB), which existence has been demonstrated and activity cannot be excluded after closure of the repository [17]. Several experiments and modelling calculations have been performed (or are still ongoing), in order to understand these perturbations and their effects on the geochemistry. The main uncertainty lies in the demonstration of the presence or absence of bacterial activity within the near-field and the maximum concentration of reduced sulfur species to be expected [14].

6. Conclusions

The Supercontainer (SC) design is the preferred Belgian option for the final disposal of VHLW and SF in deep underground clay layers. The SC consists of a carbon steel overpack, containing VHLW canisters or SF assemblies, surrounded by a thick concrete buffer, which in turn, is entirely encased in a stainless steel envelope. Under the predicted conditions within the SC (highly alkaline concrete buffer), the carbon steel overpack is expected to undergo uniform corrosion (passive dissolution).

An integrated R&D strategy is developed to demonstrate and defend that the integrity of the carbon steel overpack can be ensured at least during the thermal phase. This integrated approach, proposed to estimate the lifetime of the carbon steel overpack, consists of three steps:

- *Lifetime prediction.* The lifetime of the overpack is predicted on the basis of the corrosion evolutionary path, which describes the changing geochemical environment the overpack is subjected to during the different phases of (and also prior to) the disposal phase. The evolutionary path is divided in different phases (aerobic, anaerobic) and the ‘best estimate’ uniform corrosion rate for each of these phases is determined. The reduction in wall thickness is obtained by integrating the corrosion rate, expressed as loss in wall thickness (in μm/year), over time. The thus predicted lifetime is only valid if corrosion occurs uniformly over the entire overpack surface. Therefore, the approach exists in confirming that other forms of corrosion (localised corrosion such as pitting and crevice corrosion, and stress corrosion cracking) cannot take place under the circumstances described in the corrosion evolutionary path. This is called the ‘exclusion principle’.

- *Validation.* The data of the corrosion evolutionary path, the suitability of the ‘exclusion principle’ and the interface models should be validated through a limited set of well-defined tests (*e.g.* an *in situ* experiment).
- *Confidence building.* An active publication policy (*a.o.* publications in peer reviewed journals), together with a periodical review of the programme by a panel of experts in the field, should help to enhance the confidence in the predicted lifetime of the SC.

Scoping calculations based on a local equilibrium-diffusion transport model have been performed and indicated that the near-field will likely remain alkaline (pH > 12.5) for a geological time span (>80000 years).

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