EURAD

State of Knowledge (SoK) Report

HLW/SF Containers

Domain 3.2.1

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Executive Summary

The aim of this report is to summarise the current state-of-knowledge of all aspects of the selection, design, fabrication, long-term performance and lifetime prediction of containers envisaged for the disposal of radioactive materials in deep underground repositories (or geological disposal facilities, GDFs). In doing so, it has been necessary to be selective in covering the wide range of studies that have been conducted since the mid-1970s. The scope covered in this document is thus limited to a discussion of containers for the disposal of heat-generating high-level waste (HLW) and spent fuels (SF), and excludes the variety of container designs proposed for less radioactive wastes, such as intermediate level waste (ILW). Furthermore, the discussion is limited to conventional engineering alloys, and excludes the use of ceramics (either as coatings or as a bulk container material) or of advanced alloys and coatings. Thus, the main container materials considered here are copper, carbon steel, and some passive alloys (specifically titanium and nickel alloys). Copper coatings applied to a carbon steel or cast iron substrate are also described. In terms of the nature of the repository, the main focus is on 'deep' (>200-300 m depth) repositories located below the water table in either saturated, anoxic (crystalline or sedimentary) host rock formations or in dry salt formations. Relevant information from programmes considering disposal in unsaturated host rock is also included.

Safety functions, performance targets and container requirements are discussed for post-closure and operational periods over the container's lifetime. The primary post-closure safety function of disposal containers for SF and HLW is to provide physical containment of radionuclides for a certain period, while avoiding significant degradation of the performance of other engineered or geological barriers, due to e.g., interactions with heat, gas or corrosion products. Whereas, operational safety functions of the container typically include the ability to enable safe handling, transport, emplacement, and, in specific programmes (e.g. French programme), reversibility of emplacement operations (to allow future generations to take a different approach, should it prove desirable).

The choice of container material and material design is heavily influenced by the geological environment of the GDF. Based on the host geology broader geological setting present or expected in different locations, various waste management organisations have developed their own specific concepts for the design of the disposal system, including the combination of disposal container, its associated buffer material and any surrounding tunnel backfill material. The container material and design need to be selected such that the required lifetime of the containers can be achieved, and so that the overall safety case for disposal meets the requirements of national regulatory authorities. Various container designs, including corrosion-allowance systems and corrosion-resistant systems, as well as the use or otherwise of pre-fabricated emplacement modules, are discussed.

The container is designed to withstand the service environment, which imposes a combination of mechanical loads and corrosive conditions. Depending on the nature of the host rock, the choice of engineered barriers and the container design, mechanical loading arises from residual stress, buffer swelling, hydrostatic pressure, lithostatic loads and, during operations, impact loads and, to a lesser degree, expansion of corrosion products. In the conditions of interest, corrosion processes are influenced by the chemical environment, temperature, presence of oxidants, radiation/radiolysis, microbiological activity, degree of water saturation, and mass transport characteristics of the environment. Evolution of the near-field environment in terms of mechanical loading, temperature and chemical environment (including redox potential) are thus considered in this work.

There are no national or international codes and standards specific to the working conditions and durability requirements of disposal containers. Nevertheless, disposal containers are sealed vessels designed to prevent leakage of their contents into the environment, and as such there are parallels with pressure vessels and other containers designed to prevent leakage of hazardous materials under load. Although an internationally established design methodology does not exist, noting the variability of geological and regulatory boundary conditions, container designs developed for different national programmes fall into similar categories.

Fabrication of the container can be undertaken in several different ways, depending on the design, and can include a range of manufacturing processes. In general designs that reduce the total amount of welded material are preferred, owing the risk of defects and increased susceptibility to corrosion. For all designs, however, sealing of the container must be performed and a range of welding techniques have been investigated for the final closure weld. The presence of flaws in fabricated components and closure welds is inevitable and inspection of the containers is likely to be required to ensure that flaws present are within the acceptance criteria. To achieve this, a range of inspection techniques are available and have been considered for the final closure weld. A key difference to container manufacture in other industries is that disposal containers for highly radioactive waste must be sealed and inspected in a shielded facility, which generate additional complexities.

The long-term durability of the container is discussed in terms of its resistance to mechanical loading and degradation through corrosion. For each material the range of expected degradation mechanisms is described e.g., microbiologically influenced corrosion, galvanic corrosion, environmentally assisted cracking, hydrogen embrittlement, localised corrosion, uniform corrosion, and radiation-assisted corrosion. The main materials considered are copper and carbon steel, but passive materials including nickel and titanium alloys are also compared in terms of their advantages and disadvantages with regards to degradation processes and resulting durability. The mechanical performance of the container is considered based on plastic collapse, creep, fracture and loading under impact.

In safety cases being developed in advanced disposal programmes, container lifetime is considered not deterministically for a single container but for a population of containers emplaced within a disposal facility, whose lifetimes form a probability distribution. In host rock in which substantial groundwater flow through the rock might be expected (e.g., ‘hard’ rocks), longer container lifetimes are desirable (typically greater than100,000 years), as the container may provide the key barrier preventing the release of radionuclides to the biosphere for sufficiently long times to ensure that any radiological dose (and/or radiological risks) to future human populations inhabiting regions surrounding the GDF remains below regulatory limits. In low-permeability sedimentary host rocks, the geological barrier itself is likely to provide sufficient retardation of the transport of radionuclides back to the biosphere on its own. In this case the container lifetime is likely to have more limited influence on the radiological dose to which future human generations (and surface biota) might be exposed to as a result of the release of radioactivity from the GDF. However, a good container durability provides additional confidence in the safety of the GDF and facilitates the reversibility of emplacement operations should it be required. Additionally, corrosion processes in these conditions (particularly when carbon steel containers are envisaged) affect gas generation rates in the repository, which could pressurise the engineered barriers and host rock. The prediction of container lifetimes in this document is discussed based on the definition of container failure, quantification of likely degradation mechanisms and reasoned exclusion of unlikely degradation mechanisms.

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List of abbreviations

ASME	American Society of Mechanical Engineers
BWR	Boiling Water Reactor
CANDU	Canada Deuterium Uranium
CEP	Corrosion evolutionary path
EAC	Environmentally assisted cracking
EB	Electron beam
EBS	Engineered barrier system
ECA	Engineering critical assessment
EDZ	Excavation damaged (or disturbed) zone
FAD	Failure assessment diagram
FSW	Friction stir welding
GDF	Geological disposal facility
GMAW	Gas metal arc welding
GTAW	Gas tungsten arc welding
HAZ	Heat affected zone
HIC	Hydrogen-induced cracking
HLAW	Hybrid laser arc welding
HLW	High-level waste
HSR	High strength rock
ILW	Intermediate level waste
LILW	Low and intermediate-level waste
LSSR	Low strength sedimentary rocks
MAG	Metal active gas
MIC	Microbiologically influenced corrosion
PEM	Prefabricated emplacement module
POCR	Probability of correct rejection
POD	Probability of detection
POI	Probability of identification
PWR	Pressurised Water Reactor
RAC	Radiation assisted corrosion
SCC	Stress corrosion cracking
SCE	Saturated calomel electrode
SF	Spent fuel
SRB	Sulfate reducing bacteria

TIG	Tungsten inert gas
WMO	Waste management organisation

Definitions

Aerobic	Term used to describe the initially oxidising conditions in the repository due to the presence of trapped atmospheric O_2 and/or oxidising radiolysis products and oxidised corrosion products (Cu(II) and Fe(III). In this report, for simplicity, this term is used interchangeably with the term 'oxic' to describe environmental conditions and processes.
Anaerobic	Term used to describe the subsequent period in the evolution of the repository environment characterised by the absence of O_2 and the presence of sulfide. In this report, for simplicity, this term is used interchangeably with the term 'anoxic' to describe environmental conditions and processes.
Anodic	Electrochemical half-reaction in the overall corrosion reaction involving the oxidation of the metal.
Buffer	Material used immediately adjacent to the container to condition the near-field environment, control microbial activity, provide structural support for the container in the repository, and to ultimately delay the transport of radionuclides. Typically in the form of either highly compacted bentonite or a cementitious material.
Cathodic	Electrochemical half-reaction in the overall corrosion reaction involving the reduction of the oxidant.
Corrosion allowance	A classification of container material based on a tendency to corrode uniformly at a predictable rate.
Corrosion resistant	A classification of container material based on the tendency to form a passive film resulting in very low rates of uniform corrosion.
Crevice corrosion	Form of localised corrosion resulting from a locally occluded region.
Environmentally assisted	Various forms of cracking resulting from the degradation of the container cracking material properties due to interaction with the environment, including HIC and SCC.
Fracture	Mechanical degradation mechanism resulting in crack propagation, and which may be ductile or brittle in nature.
Hydrogen-induced cracking	Corrosion mechanism resulting from degradation of the ductility and/or toughness of the container material due to the absorption of hydrogen.
KBS-3 container	Container design based on a thick (typically 50 mm) outer copper corrosion barrier supported by an inner steel or cast iron insert for structural strength.
Localised corrosion	Form of corrosion resulting from the permanent spatial separation of anodic (oxidation) and cathodic (oxidant reduction) processes.
Microbiologically influenced corrosion	Form of corrosion due to microbial activity, either directly on the container corrosion surface associated with a biofilm or due to the transport of corrosive metabolic by-products formed remotely in regions of the repository in which such activity is possible.
Pitting	Form of localised corrosion resulting in local penetrations in metal surface.

Plastic collapse	Mechanical degradation mode resulting from the stress exceeding the yield strength of the material (either globally or locally).
Redox potential	The redox potential is used to describe a system's overall reducing or oxidising capacity and relates to the tendency for chemical species to gain/lose electrons.
Stress corrosion cracking	A form of EAC involving the formation of cracks in a susceptible material exposed to a supportive environment and a tensile stress.
Supercontainer	Concept in which the container and the associated buffer material are assembled and emplaced as a single unit.
Surface roughening	Form of localised corrosion resulting from the non-permanent spatial separation of anodic and cathodic processes and leading to uneven uniform corrosion.

1. Aim/Scope

The concept of a sealed container for the disposal of high-level waste (HLW) and spent fuel (SF) is one that is understandable to both the layperson and the informed expert. As the only absolute barrier in the overall multi-barrier disposal system, much attention is frequently placed on the performance of the container in safety cases for disposal.

The aim of this report is to summarise the current state-of-knowledge of all aspects of the selection, design, fabrication, long-term performance and lifetime prediction of containers envisaged for the disposal of radioactive materials in deep underground repositories (or geological disposal facilities, GDFs). In doing so, it has been necessary to be selective in covering the wide range of studies that have been conducted since the mid-1970s. The scope covered in this document is thus limited to a discussion of containers for the disposal of heat-generating high-level waste (HLW) and spent fuels (SF), and excludes the variety of container designs proposed for less radioactive wastes, such as intermediate level waste (ILW). Furthermore, the discussion is limited to conventional engineering alloys, and excludes the use of ceramics (either as coatings or as a bulk container material) or of advanced alloys and coatings. Information about these latter materials can be found in Gaggiano and Diomidis (2023). Thus, the main container materials considered here are copper, carbon steel, and some passive alloys (specifically titanium and nickel alloys). Copper coatings applied to a carbon steel or cast iron substrate are also described. In terms of the nature of the repository, the main focus is on ‘deep’ (>200-300 m depth) repositories located below the water table in either saturated, anoxic (crystalline or sedimentary) host rock formations or in dry salt formations. Relevant information from programmes considering disposal in unsaturated host rock is also included.

2. Safety functions, performance targets, and container requirements

The primary post-closure safety function of disposal containers for SF and HLW is to provide physical containment of radionuclides for a certain period, while avoiding significant degradation of the performance of other engineered or geological barriers, due to e.g., interactions with heat, gas or corrosion products. Even after failure, the container may continue to provide a containment function, either by acting as a mass-transport barrier, if the breaching of the container wall is localised, or by acting as a redox or sorption barrier. The lifetime of the container is defined differently in various disposal programmes and is in some cases defined by the national regulatory authority. Consequently, it can vary between the period of retrievability or reversibility (a few centuries, e.g. France (Andra 2016), the ‘thermal period’ (a millennium, e.g. Switzerland (Johnson and Zuidema 2013), or significantly longer, in some cases linked to the period until radioactivity decays to levels consistent with ore bodies (up to hundreds of thousands of years, e.g. Sweden, Finland, Canada (SKB 2009, Scully et al. 2016)). Operational functions of the container typically include the safe handling, emplacement and retrieval of the waste. An additional function that is relevant during both post-closure and operational periods is the avoidance of criticality. However, this is primarily linked to the properties and configuration of the waste inside the container and is not discussed further.

To ensure adequate lifetimes, individual container requirements or performance targets are defined. These can vary significantly between national programmes and are typically linked to the type of waste, the selected container material(s) and disposal concept, and the near-field and geological environments. Lifetime-related requirements typically used in performance assessments are related to structural integrity under expected loads, the wall thickness loss due to corrosion, and the degradation of materials properties during disposal, such as embrittlement. For coated containers or other container concepts involving multiple components or materials, the requirements are assigned to each component or material according to its function. An important aspect is the properties of the final seal or weld, which tend to be different from the rest of the container and are typically more difficult to control.

The required structural integrity performance of the disposal container is related to the stresses expected to be applied during disposal, which could be lithostatic, hydrostatic, due to swelling of buffer materials employed in the EBS (particularly important in the case of bentonite, which swells upon water saturation) or due to rock movements such as earthquakes. Apart from the hydrostatic loads which are isotropic, the other types of container loads can be unevenly distributed and thus generate anisotropic stress fields and shear forces. The resulting stresses in the container wall, lid and base, including the seal joint region, must be less than the failure limit for the expected lifetime of the container by an adequate margin. This is typically demonstrated by structural assessments taking into account the strength and fracture toughness of the container material(s), including consideration of the maximum conceivable size of manufacturing defects, and the evolving disposal environment. This type of analysis is required for all conceivable container materials and designs.

The loss of wall thickness due to corrosion over time is typically an important input to the structural analysis described above. When sufficient corrosion has occurred, the wall thickness becomes sufficiently small that stresses in the container wall increase and lead to plastic collapse. This is primarily a concern for container concepts that are based on carbon steel.

Degradation of intrinsic materials properties can occur due to phenomena such as hydrogen uptake, neutron embrittlement or preferential leaching/dissolution (e.g., dealloying). Its importance is strongly dependent on the selected container material. Nevertheless, degradation of material properties often leads to a reduction of fracture toughness and an increased risk of mechanical failure.

Operational safety is strongly linked to container integrity during handling in the surface facility and transportation to the repository, which is ensured by adequate mechanical strength under loads expected during normal operations as well as incidents. Handling loads are related to the size, shape and mass of the loaded containers. The allowed size and mass of containers is usually defined by transport limitations and the size of underground excavations. In some cases, a radiation dose limit is also employed, leading to surface contamination, shielding, and container contents requirements. The emplacement and retrieval concepts vary significantly between national programmes and impose different requirements on each container concept. Finally, operations usually require handling features on the containers or waste packages.

3. Choice of material and container design

Various waste management organisations have developed their own specific concepts for the design of the disposal system, including the combination of disposal container, the surrounding backfill material and the host geology. The container material and design need to be selected such that the required lifetime of the containers can be achieved, and so that the overall safety case for disposal meets the requirements of national regulatory authorities.

For each individual country, the choice of the design will be governed to a certain extent by the geological environment that is available for constructing a GDF. An additional factor affecting the specific design of the container is the type of waste that needs to be contained; for example, in countries where reprocessing of the spent fuel is not undertaken, the container design only needs to accommodate spent fuel, whereas where reprocessing is carried out, the container may also need to accommodate HLW (typically in vitrified form in metallic storage containers fabricated from stainless steel). In some countries, there may be a regulatory requirement that the waste should be retrievable and this would also affect the design of the container and overall EBS. For example, in France a cement-based liner is used to provide the horizontal boreholes in which the disposal container is emplaced sufficient geometric tolerances and durability to enable reversibility of the emplacement operations for one century after disposal.

In broad terms, the host geologies available to waste management organisations can be categorised as hard/crystalline (e.g. granite-like), soft/plastic (e.g. sedimentary mineral deposits) and salt deposits (e.g., halite). Disposal facilities will be constructed at depths of several hundred meters and in the long-term are expected to result in anoxic, aqueous conditions in the vicinity of the waste container, as the indigenous groundwater permeates the facility during the post-closure period (with the exception of salt deposits where the amount of water is extremely limited). It is therefore necessary to identify materials that have the necessary corrosion resistance and mechanical properties to be suitable as waste container materials in such conditions.

The environment immediately adjacent to the waste container can be controlled by using a carefully selected buffer material that will modify the local physico-chemical conditions such that they are conducive to extended lifetimes for the container materials. The most common buffer material for the disposal of HLW and SF is bentonite, a clay predominantly composed of the layered mineral montmorillonite, which swells when it is wetted. In specific cases (Belgian disposal programme) cementitious buffers are envisaged. The pH value of the porewater is approximately 8.4 for bentonite and >12.5 for a cementitious backfill, although the latter pH value may decrease in the long-term due to carbonation, reactions with the aggregates or leaching of its alkaline components and eventual equilibration with groundwater. The pH of the environment is very important when considering the corrosion behaviour of the container material.

The candidate materials that have been considered for disposal containers can be categorised as ‘corrosion allowance materials’ and ‘corrosion resistant materials’. The former category includes carbon steel, copper (including copper-coated substrates), whereas the latter includes stainless steels (including austenitic and duplex grades), nickel alloys (e.g., Hastelloys, Inconels) and titanium alloys (e.g. Grade-2). Extensive reviews of possible container materials have been conducted by waste management organisations (e.g., King and Padovani 2011, King et al. 2016).

With respect to achieving the required container lifetime, the advantages and disadvantages of the various possible combinations of host geology-backfill-container material are summarised at a high level in Table 1 below.

3.1 Corrosion-allowance systems

The concept of using a copper container in a bentonite backfill in a HSR environment originated in Sweden in the 1970s and is known as the KBS-3 concept; it has also been adopted in Finland and it is one of the main concepts under consideration in several other countries. A summary diagram of the concept is shown in Figure 1. The container is sealed by welding, with friction stir welding currently being the preferred technique in both Finland and Sweden. Under anoxic conditions, copper is thermodynamically stable and immune to corrosion unless certain species, such as sulfide, are present (see Section 7.1.1). This makes this design very durable with long lifetimes possible.

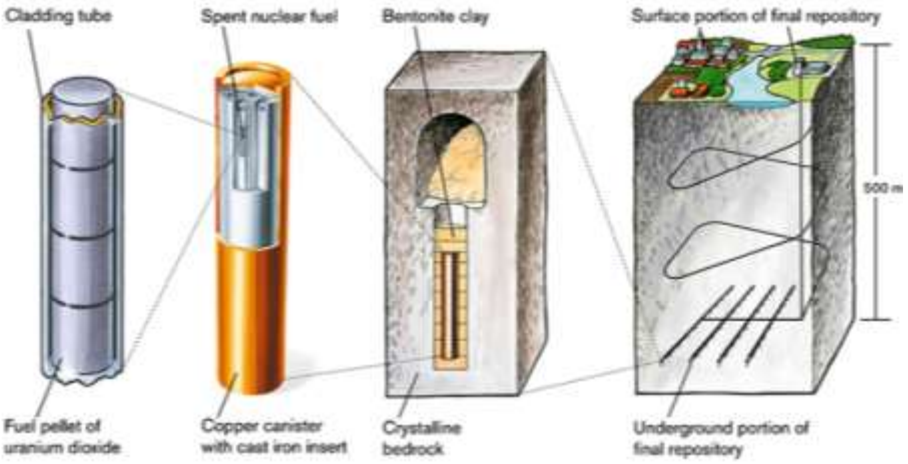

Figure 1: Schematic diagram illustrating the KBS-3 waste disposal concept.

Table 1: Advantages and disadvantages of various material combinations in current waste disposal concepts

Host geology	Buffer material in contact with container	Container material	Advantages	Disadvantages
Crystalline (hard) rock	Bentonite	Copper (including copper-coated carbon steel)	Very little hydrogen produced by anaerobic corrosion Predictable uniform corrosion rates Low tendency for localised corrosion or EAC Possibility of very long lifetimes Supported by natural and archaeological analogues Dual-wall design less sensitive to joint mechanical-corrosion degradation modes than single-walled containers (e.g. coatings may be subjected to lower tensile stress fields than critical regions of the body of a single wall container such as the final closure weld)	Possibility of creep for dual-wall design (with gap between copper and steel/iron substrate) Corrosion performance strongly tied to properties of buffer Potentially more expensive depending on container design
Sedimentary (plastic) rock		Copper (including copper-coated carbon steel)	As above	As above
		Carbon steel	Inexpensive material Predictable uniform corrosion rates Low tendency for localised corrosion Excellent mechanical performance Extensive manufacturing experience at-scale Supported by archaeological analogues	Corrosion products may affect the local performance of the bentonite Production of hydrogen during the anoxic phase Single-shell design subject to joint mechanical-corrosion degradation modes
Sedimentary (plastic) rock	Cement	Carbon steel	Inexpensive material. Very low uniform corrosion rates.	Need to assess risk of localised corrosion process and EAC

Host geology	Buffer material in contact with container	Container material	Advantages	Disadvantages
			Excellent mechanical performance	
Evaporite[1]	Crushed host rock	Carbon steel	Inexpensive material. Excellent mechanical performance Extensive manufacturing experience at-scale	Degradation of material properties and decrease of fracture toughness in the presence of hydrogen
Any	Cement	Stainless steel	Very low uniform corrosion rates	Relatively expensive Susceptible to localised corrosion in the presence of sufficiently corrosive conditions
Any	Any	Nickel alloys	Very low uniform corrosion rates Wide range of alloys with varying degrees of corrosion resistance available Less reliant on properties of buffer than other materials	Expensive Susceptible to localised corrosion in the presence of sufficiently corrosive conditions (more corrosive than for stainless steels) Sealing techniques unproven.
Any	Bentonite, no buffer	Titanium and titanium alloys	Very low uniform corrosion rates Highly resistant to MIC Immune to pitting Less reliant on properties of buffer than other materials	Very expensive, especially Pd-containing alloys Some grades susceptible to crevice corrosion Some grades susceptible to hydride-induced cracking

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[1] The corrosion performance of the disposal canisters is of secondary importance in an evaporite host rock because the rock is expected to be dry. In this case the selection of the material is driven by costs and mechanical performance.

In Sweden and Finland, a phosphorus-doped, oxygen-free copper has been specified for the outer corrosion barrier. Approximately 30-100 ppm phosphorus is added to oxygen-free copper to confer a higher creep resistance (SKB, 2001). In recent years the option of using a copper-based coating on a carbon steel shell has been investigated, particularly in Canada (Keech et al. 2021), Switzerland (Diomidis and King 2022) and Japan (Suzuki et al. 2021). The advantage of such an approach is that the design, in which the copper corrosion barrier is metallurgically bonded to the underlying carbon steel substrate, is not sensitive to creep as in the KBS-3 design, in which a gap exists between the copper shell and the structural insert. For the Canadian programme, which focuses on the disposal of relatively small spent fuel assemblies (CANDU fuel), the material used for the steel substrate is also commercially available in the required size, thus removing the need for bespoke manufacture. The latter, coupled with the ability to tailor the amount of copper required around its corrosion performance, rather than both its corrosion and mechanical performance, enables an overall reduction in cost. Several application methods for copper coatings are under development (e.g., cold-spray, electrodeposition).

The other commonly proposed container material in the ‘corrosion allowance’ category is carbon steel. The main advantages of this material are that it is relatively inexpensive, manufacturing experience at scale is commonly available, and that, like copper, in the conditions typically expected in a disposal facility it corrodes uniformly and at a predictable rate. A wide range of different grades of carbon steel are available with similar corrosion behaviour but with a range of mechanical properties. Since thick-walled containers are required to provide sufficient corrosion allowance, a low-strength alloy that is more-resistant to hydrogen-related degradation mechanisms can be selected while still providing sufficient structural integrity. A major consideration in repository safety cases regarding the use of carbon steel is that when it corrodes under anoxic conditions it generates gaseous hydrogen at a rate that depends on the environmental conditions, particularly on the pH value of the aqueous phase in contact with it. If a significant pressure of gaseous hydrogen were to be produced in the EBS, it would have the potential to disrupt the structural integrity of the EBS and/or host rock as well as accelerate the transport of radionuclides. The behaviour of hydrogen also depends on the local geology, since gas transport will be more rapid through a fractured crystalline rock than through a relatively impermeable sedimentary rock such as clay (Diomidis et al. 2016). Carbon steel can be used with both bentonite and cementitious buffer materials (in the latter case, due to passivation processes in highly alkaline environments, as a passive, i.e. corrosion-resistant material). For both copper and iron, there is a significant body of archaeological and natural analogue evidence to support predictions of their long-term corrosion behaviour, including iron and bronze artefacts from prehistoric times and, in the case of copper, natural metallic ore bodies (e.g., native copper). Although the limitations of using analogues to make lifetime predictions need to be recognised (e.g., the environmental conditions are not known exactly, and they may not reflect repository conditions sufficiently closely), analogues provide a very important basis to demonstrate mechanistic understanding of corrosion processes over much longer times than available with testing, and thus build confidence in long-term predictions.

3.2 Corrosion-resistant systems

In some disposal programmes, disposal containers making use of passive materials (e.g., titanium, nickel alloys or stainless steel) or passivated materials (e.g., carbon steel in cement systems) have been considered. For example, a titanium-based container was historically considered in the Canadian programme, whilst a carbon steel / cement system is currently considered in the Belgian programme.

Passive materials are an alternative to the use of copper and carbon steel and offer certain advantages. Passive materials exhibit low rates of uniform corrosion due to the formation of a highly protective (passive) surface film. For this reason, passive alloys can be used in thinner sections than corrosion allowance materials, which may result in cost saving or containers that are less heavy and easier to handle. However, a thin outer corrosion-resistant layer would likely need internal structural support by a steel or cast iron insert, which might then offset any weight advantage and also complicate container fabrication. A wide range of Ti- and Ni-based alloys are available with an equally wide range of corrosion resistance, enabling the container material to be selected for the expected service environment.

Because the corrosion resistance of passive materials is conferred by the properties of the surface film, these materials are less reliant on the buffer material to maintain a suitable near-surface environment, thus ensuring that the performance of the container is independent of that of the buffer. Unlike copper, and to a large degree carbon steel, certain passive materials could be used without any buffer material, which may be an advantage in some repository designs.

From a corrosion perspective, the main disadvantage of using passive materials is that, although highly corrosion resistant in most circumstances, Ti- and Ni-based alloys are susceptible to localised corrosion in some environments (see Section 7.1.4). Since the rates of localised corrosion propagation (and, to some extent, the likelihood of initiation of localised corrosion) can be more difficult to predict than the rates of uniform corrosion, a historic (and still relevant) perception is that the development of well-supported safety cases may be more difficult with these materials. The lack of natural and man-made analogues for their long-term corrosion performance makes it more difficult to build confidence in long-term predictions of their behaviour. For these reasons, the corrosion allowance approach has become, and remains, the favoured option in many disposal programmes.

3.3 Use of pre-fabricated emplacement modules

Some disposal concepts make use of an approach in which the primary container containing the HLW/SF is mounted inside a secondary envelope and surrounded by a buffer material. The resulting assembly is variously referred to as a “supercontainer” in the Belgian concept or as a Prefabricated Emplacement Module (PEM) in other programmes. This approach involves placing the buffer material around the container under more controlled conditions at the surface and should, therefore, ensure greater quality of the emplaced buffer. However, it increases the overall weight of the waste package, which may affect the ease of handling during emplacement. The inner container provides the corrosion barrier, the purpose of the outer envelope is largely to enable handling. The buffer material provides a controlled environment around the primary containment. Good examples of this approach are the design proposed for use in Belgium, Canada and, as an alternative option, in Sweden and Finland. In the Belgian design (shown schematically in Figure 2), the waste is encased inside a carbon steel overpack (which acts as a disposal container), which is mounted in a cylindrical stainless steel envelope filled with a cementitious buffer material (Kursten, et al., 2021). In the Swedish/Finnish programmes, the KBS-3H concept envisages using horizontally-placed modules made of a copper-based container within a bentonite-based buffer encased within a titanium envelope (Johannesson, 2014) (Figure 3). In the Canadian programme a copper-coated steel container is emplaced within pre-compacted bentonite blocks (Figure 4).

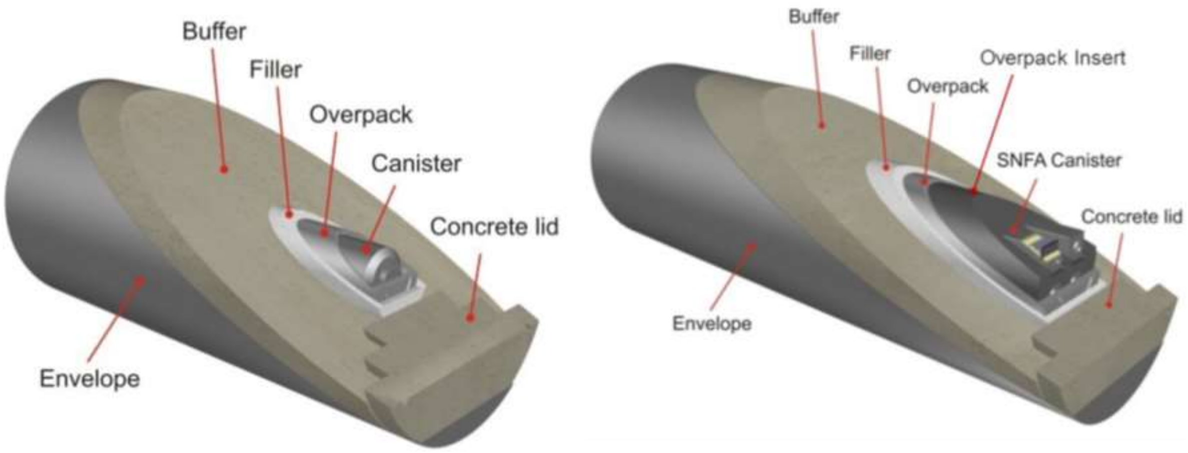

Figure 2: Schematic diagram illustrating the Belgian supercontainer waste disposal concept.

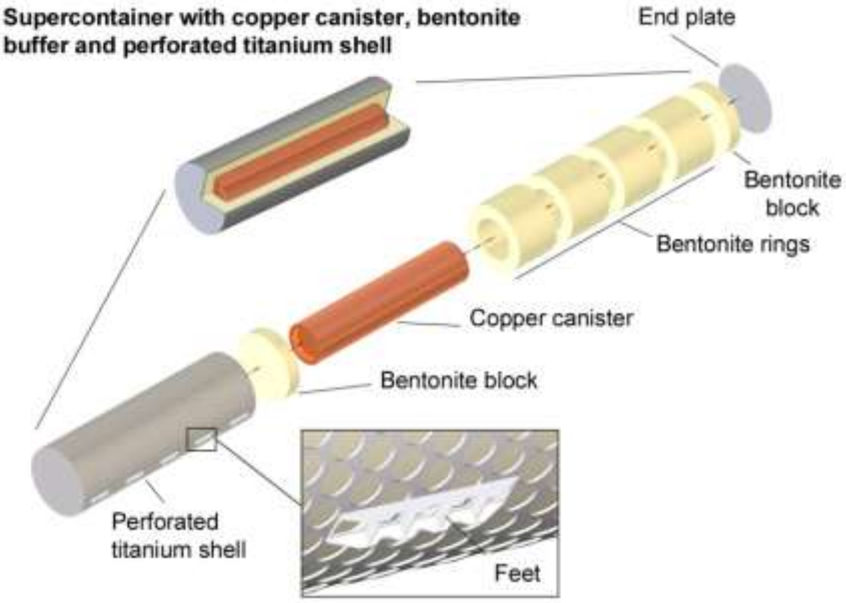

Figure 3: Schematic diagram illustrating the use of supercontainers in the Scandinavian KBS-3H waste disposal concept.

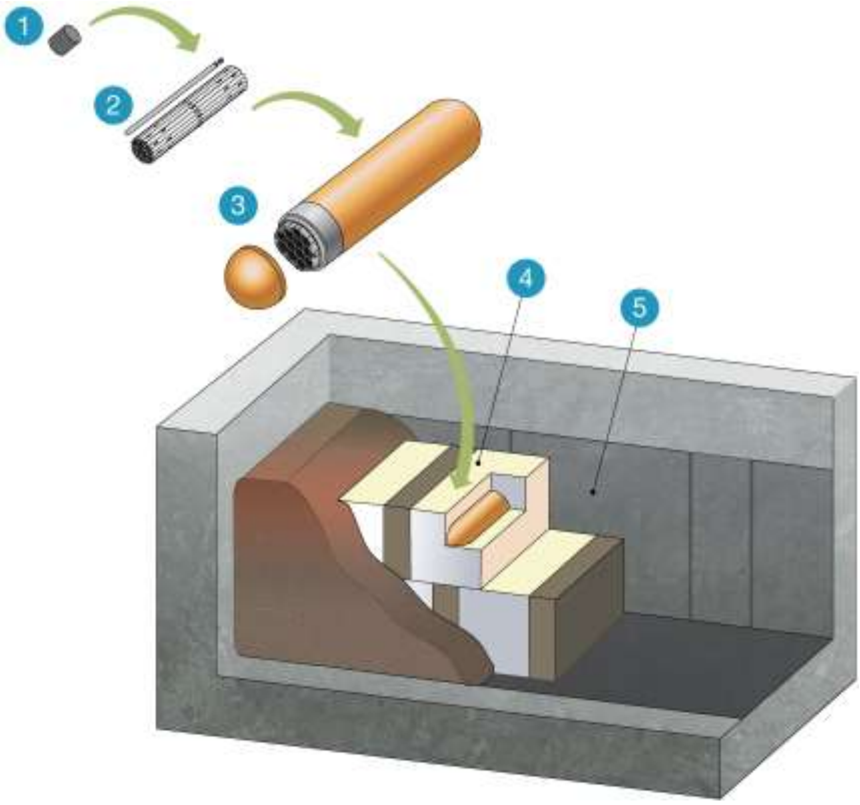

Figure 4: Canadian buffer box arrangement of a copper-coated container embedded in pre-compacted buffer blocks (https://www.nwmo.ca/en/A-Safe-Approach/Facilities/Deep-Geological-Repository/Multiple-Barrier-System). (1) fuel pellet, (2) CANDU fuel bundle, (3) copper-coated steel container and end cap, (4) pre-compacted bentonite buffer, (5) disposal tunnel excavated in host rock.

4. Service (environnemental) conditions

4.1 Factors contributing to structural degradation (mechanical loads)

The container is designed to withstand mechanical loads in the form of applied and residual stresses. In addition to their effect on the structural integrity of the container, these loads can, in combination with a suitable corrosive environment, also lead to joint mechanical-corrosion degradation modes, such as environmentally assisted cracking (EAC). The various sources of applied and residual stress include:

- Residual stress: if not relieved by some form of post-weld heat treatment, tensile or compressive residual stresses could be present during the unsaturated phase. Surface tensile residual stress could lead to EAC under unsaturated conditions and can aggregate with external tensile loads (both hydrostatic and lithostatic) as the repository saturates. The level of residual stress will depend on the fabrication method (commonly the final sealing weld), with an overall balance of tensile and compressive stresses. Conservatively, the maximum tensile residual stress can be assumed to be equal to the yield strength of the material.
- Buffer swelling pressure: bentonite-based buffer materials develop a swelling pressure when confined and upon saturation with incoming groundwater, with the swelling pressure dependent (among other factors) on the smectite (montmorillonite) content. The swelling pressure may be isostatic or, in the case of uneven density distribution or non-uniform wetting, asymmetric. Asymmetric swelling can result in a bending load on the container and a tensile stress component, as opposed to the compressive surface stresses that develop due to uniform swelling. The buffer swelling pressure is typically in the range 2-10 MPa, depending on buffer density and composition (smectite content), porewater salinity, and temperature.
- Hydrostatic pressure: for repositories located in the saturated zone in which groundwater transport is likely to occur through advection in faults or slow resistance paths (e.g. repositories built in hard rocks), a hydrostatic pressure will develop due to the water column extending from the surface to repository depth. The height of this water column may be extended during periods of glaciation if a continuous water path exists from the surface to the base of the icesheet. The hydrostatic pressure will be isotropic. The hydrostatic pressure amounts to 1 MPa for every 100 m depth of the repository (below ground level or below the surface of the icesheet). In some countries, ice sheets of up to 4-5 km in thickness are considered, resulting in a maximum total hydrostatic pressure of 40-50 MPa.
- Lithostatic loads: in lower strength rocks, such as claystones and salt deposits, an isostatic or asymmetric lithostatic load can develop on the container as a result of rock movement (creep) re-filling previously excavated areas (deposition tunnels). The extent of such loads depends on the depth of the repository, the nature of the host rock, and the level of in situ stress.
- Shear loads: in the event of seismic-induced motion across a fracture that intersects the container located in the repository, a shear load could be transferred to the container (depending on the properties of the surrounding buffer material, if any is present). If the shear load is high enough, the container could be plastically deformed, with a resulting residual stress.
- Impact loads: impact loads are possible during handling and could result in localised deformation, cold work or, in the extreme, container breaching.
- Expansive corrosion: Stresses can be generated in waste container components by the formation of corrosion products that have a larger molar volume than the original metal if they develop in a confined space, a phenomenon that is sometimes called ‘oxide jacking’. This effect is possible if the Pilling-Bedworth ratio is greater than 1 and the mechanical properties of the particular corrosion product are appropriate. Possible locations for the effect include between mating components in the design of containers. For example, the possibility of generating stresses through expansive corrosion at the interface between the cast iron insert in the KBS container design and the surrounding copper canister has been investigated (Smart et al. 2001, 2003, 2004). Factors to consider include the expected corrosion rates and the extent to which corrosion products are precipitated as opposed to diffusing into the surrounding buffer material.

4.2 Factors contributing to corrosion degradation (chemical loads)

The most important environmental factors affecting the corrosion behaviour of the container are listed below and discussed at a high level in the context of disposal of HLW and SF in the following sub-sections:

- Chemical environment (pH, pore-water composition)
- Temperature
- Concentration of oxidants (including oxygen)
- Radiation and radiolysis of the near-field environment
- Microbiological activity
- Degree of water saturation
- Mass transport characteristics of the environment

The **chemical environment** covers factors such as the pH of the aqueous phase in contact with the container surface and the concentration in solution of anions, cations and other species, such as organics. In the context of waste disposal, the aqueous phase may be in the form of bulk water, such as groundwater (pH near neutral), or a porewater in a buffer material matrix, such as bentonite porewater (pH ~8.5), or in a cementitious matrix porewater (pH normally >12.5, depending on the particular mix of components). A wide range of ions are present in natural groundwaters, but the most significant in the context of corrosion are chloride, which may cause breakdown of protective oxide films on a metal surface, and sulfide, which can lead to the corrosion of a range of metals. The pH value has a strong effect on the stability, and hence the protectiveness, of oxy-hydroxy films formed on the surface of the container. For example, on carbon steel, the corrosion product film is more protective at the high pH values found in cement porewater than at the near-neutral pH that occurs in bentonite porewater.

The **temperature** affects the rate of the chemical and electrochemical reactions occurring on the surface of the metal. The temperature experienced by a waste container will range from ambient temperature to a maximum temperature determined by the power output of the waste container and the thermal conductivity of the environment. In an underground disposal facility, the ambient temperature will typically be 10-15 °C in the vicinity of the surface and increase according to the geothermal gradient as the depth increases (typically about 1°C per 100 m). Given their thermal output, however, the maximum temperature experienced by containers for the disposal of HLW/SF may be 100-200°C depending on the disposal concept. The repository would be typically engineered for the spacing between containers to comply with thermal limits driven by the properties and functional requirements of the buffer materials, as well as the host rock.

For corrosion to occur, an **oxidant** must be present to oxidise the metal. At the time of closure, a geological repository would be initially expected to experience naturally aerated conditions. In these conditions, the dominant oxidant is oxygen, present mainly as gaseous O_2 in the unsaturated pores of the buffer and backfill materials but also as dissolved O_2 in the buffer porewater. However, in a sealed disposal facility, oxygen is expected to be rapidly consumed by a range of processes (for example by corrosion of the disposal containers or microbial activity in the near-field). Once the oxygen has been consumed, the most readily available oxidant is water which, for some metals (e.g., iron), can be reduced to release hydrogen. The oxidising power of the environment is often described by the redox potential (E_h) value.

Radioactive waste can emit alpha, beta and gamma **radiation**[2], but the only form of radiation that is sufficiently penetrating and energetic to affect the external corrosion processes of a disposal container is gamma radiation. Gamma radiation has minimal interactions with the microstructure of materials but can interact with their electronic structures, generating alteration in energy states and generation of heat (Compton effect). Of relevance to aqueous corrosion processes, gamma radiation can interact with

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[2] In the case of radioactive waste disposal, the corrosion behaviour of disposal containers is primarily affected by gamma radiation, since alpha and beta radiation will not penetrate their wall thickness. Neutron irradiation is minimal (outside of a nuclear reactor neutrons are only generated by rare spontaneous fission events).

water, generating oxidising (and reducing) radicals, metastable and stable species (gamma radiolysis). Gamma radiolysis of aqueous solutions can result in the formation of a wide range of reactive radiolysis products, including free radicals and oxidising species, such as hydrogen peroxide. These species may affect the corrosion potential of the container metal, either in a beneficial way, for example by thickening the passive film, or in a negative way, for example by increasing the driving force for localised corrosion. It is also possible that altered energetic states of the metal surfaces affect reaction kinetics. The extent of any effect of radiation on the corrosivity of the environment depends on the intensity of the radiation reaching the surface (i.e., the dose rate) and the particular corrosion reactions involved. Even in the case of HLW and SF disposal, the level of radiation on the external surface is typically low because of the shielding provided by the container, with resulting effects on corrosion expected to be minimal (Farnan et al., 2018). In specific cases (e.g., Belgian and Canadian disposal concepts), the wall thickness of the disposal container is sufficiently low to bring dose rates to a level requiring due consideration (Padovani et al. 2017). However, it is still unclear whether it is the dose rate or the total absorbed dose that is the most important characteristic determining the corrosion behaviour, and this is an area of ongoing investigation.

Some kinds of **microbial activity** can increase the risk of corrosion. In a disposal environment, the species of most significance to corrosion are sulfate reducing bacteria, SRB. Several species have been identified, each with their own specific nutritional requirements, but they are all capable of reducing sulfate to sulfide, which is aggressive to most metals (King et al. 2011b).

In a post-closure situation, the supply of water to the surface of the metal container may be a controlling factor for the corrosion rate in a complex manner. For example, for a disposal system made of carbon steel and bentonite, the rate of groundwater resaturation may not only determine the long-term supply of oxidant (water) to the surface, but also determines the rate of **saturation** of the bentonite, which in turn determines the viability of microbes that are only active above a threshold water activity.

In some situations, the corrosion rate of the container material will be controlled by the rate of supply of the oxidant to the surface of the container (i.e., by **mass transport** processes). For copper, the overall long-term corrosion rate in a bentonite-filled repository is typically assumed to be determined by the rate of sulfide diffusion from the regions outside the bentonite buffer to the surface of the container.

4.3 Evolution of the disposal environment

An important aspect of the near-field environment is that it evolves with time (King et al. 2017). The evolution of the environment can be summarised in a diagram such as the one shown schematically in Figure 5 and is characterised by the following key factors:

- Initially, the redox potential (E_h) will be high due to the presence of residual oxygen that has been trapped in the near-field, but as the oxygen is consumed, the E_h value will become more negative, corresponding to more reducing conditions. Recent in situ experiments indicate that oxygen depletion processes might occur very rapidly in a repository, leading to the generation of reducing conditions within a few months or years (Giroud et al. 2018).
- The temperature will also rise initially and then decline as the heat output decreases as the waste decays. The temperature will decrease from the peak temperature attained approximately a decade after emplacement to the values close to that of the host rock over much longer timescales than the redox conditions (after several millennia).

In the long-term the conditions will reach a steady state, which will be cool (i.e., close to ambient temperature), low E_h and with low radiation levels. These are the conditions that must be demonstrated to lead to acceptable levels of long-term degradation of the disposal containers, predominantly by corrosion processes.

In terms of the evolution of the redox potential, anaerobic conditions will prevail for the vast majority of the container service life. For example, for copper containers with an expected lifetime exceeding 1 million years, the environmental conditions will be anaerobic for >99.999% of the time. This is an important consideration because aerobic corrosion processes, which exhibit a greater tendency to localised penetration and, possibly, are more difficult to predict, will only be possible for a small fraction of the container lifetime. If the container remains intact following this relatively brief period of aggressive conditions, then long-term containment is feasible.

The stresses experienced by containers within the near-field also evolve with time, an example of which is shown in Figure 6. In a clay repository, the initial stresses are caused by tunnel convergence and swelling of the buffer as it hydrates. Stress then increases over time due to recovery of the hydrostatic pressure within the borehole and possibly also from rock deformation and compaction of bentonite, depending on the mechanical properties of the host rock (Patel et al., 2012).

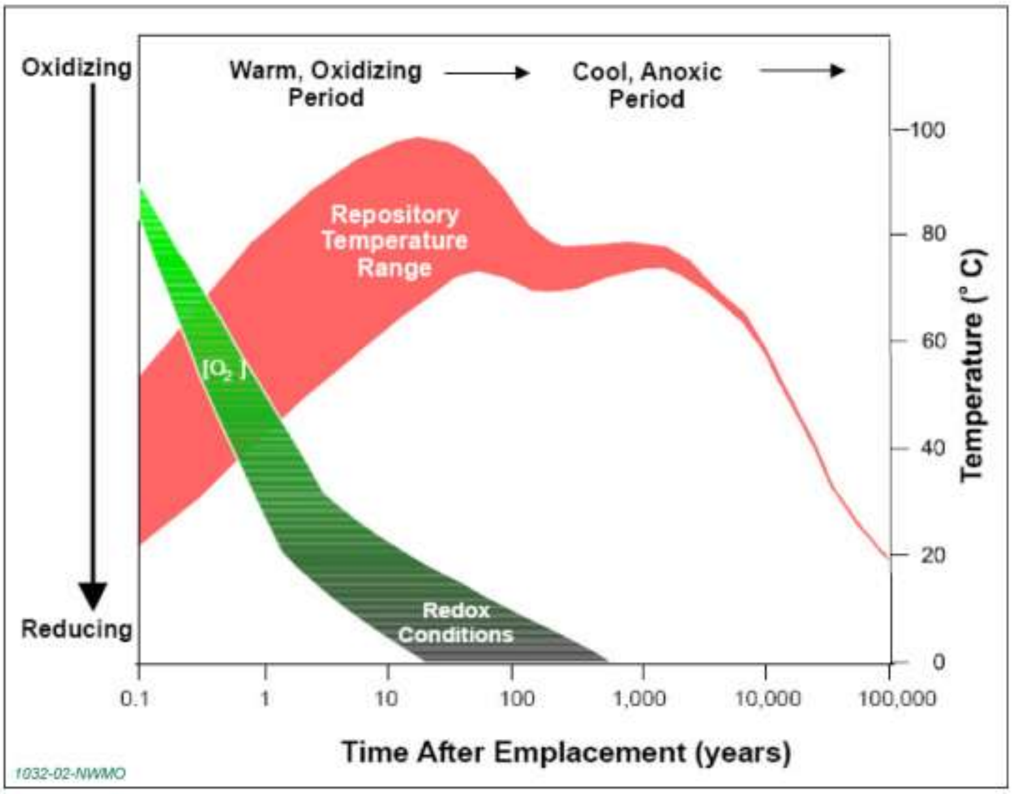

Figure 5: An illustration of the evolution of temperature and redox conditions in the EBS in the Canadian disposal concept for spent fuel (McMurry, et al., 2003)*. The duration of different periods is likely to vary significantly between different host rock types and disposal concepts.*

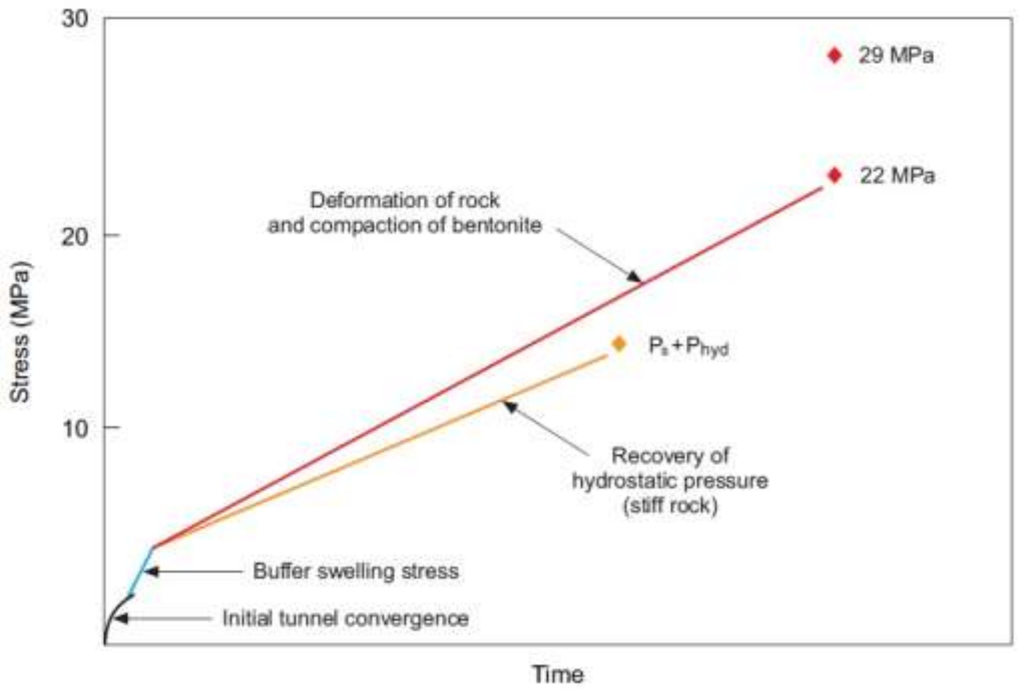

Figure 6: Possible evolution of stresses experienced by a container inside a bentonite barrier in a repository in Opalinus Clay at 900 m. The two stress evolution paths represent two alternative possibilities depending on the mechanical properties of the rock. The maximum conceivable stress at 900 m is 29 MPa horizontal and 22 MPa vertical (Landolt et al. 2009).

5. Container Design

By their nature, disposal containers are specific in their design and manufacture to their intended inventory and disposal environment. WMOs have adopted different approaches to developing container solutions for their respective national programmes, governed by regulatory and geological boundary conditions. There are no national or international codes and standards specific to the working conditions and timescales for such containers. Nevertheless, disposal containers are sealed vessels designed to prevent leakage of their contents into the environment, and as such there are parallels with pressure vessels and other containers designed to prevent leakage of hazardous materials under load. Therefore, in some cases, pressure vessel or similar codes, especially related to power plant components, are partly employed or used as general guidance (since nuclear safety regulators tend to be familiar with them).

The American (ASME) Boiler and Pressure Vessel Code Section III provides Rules for Construction of Nuclear Facility Components (ASME, 2019). Division 1 covers general Pressure Vessels, while Division 3 covers Containment Systems & Transport Packaging for Spent Nuclear Fuel & High Level Radioactive Waste. As nuclear pressure vessels and transport containers need to be of high integrity, the methods and quality criteria defined in ASME III could also be relevant for disposal containers. However, containers are not under high internal pressure (although external pressure is generally expected) and their prime function requires long periods without access for inspection. The ASME III code, therefore, is not directly applicable.

The European standard for the design of unfired pressure vessels is EN 13445 (EN 13445, 2021). While this standard does not cover the loading and other disposal conditions of containers, it could provide a basis for their construction within a European context. EN 13445 cites existing European specifications for materials, welding and inspection. For the application of EN 13445 to disposal containers, special requirements would need to be considered for aspects that would not fall under normal design or manufacturing practice. These could include the automated and remote welding and inspection of the final closure weld, the lack of access for a pressure test, pre-service and in-service inspection, and the particular loading and environmental conditions that containers might experience during disposal. The specification of containers whose lifetime might be thousands of years is well outside the scope of any existing code or standard.

An important aspect of container design development is the closure weld. This final sealing operation will need to be performed under non-standard conditions (in the presence of radiation, usually in a shielded area or hot cell). Ensuring that the weld is of adequate quality, and thus defining the weld requirements, is of primary importance for container performance. From the aspect of weld flaw acceptability, British procedures BS 7910 (BS 7910, 2013) and R6 (EDF Energy, 2015) do not predefine flaw acceptance criteria. In general, to the authors knowledge, there are currently no available procedures that provide guidance for the sealing of nuclear waste containers.

The Swedish Radiation Safety Authority (SSM) has published a handbook (Dillström et al., 2008), where a procedure for the assessment of detected cracks or crack-like defects, and for defect tolerance analysis, is described. This procedure is based on the R6 Option 1 procedure (similar to BS 7910 Option 1). According to Raiko et al. (2010) the acceptance criteria in this SSM handbook are based on stress intensity factors and not critical flaw size.

In the absence of relevant codes, a methodology based on “design-by-analysis” can be employed for the development of container design concepts (see Annex B of EN 13445 Part 3). This provides acceptance criteria for different limit states, based on the results of an elastic-plastic finite element analysis of the structure subject to the relevant loadings and yield conditions containing partial safety factors and safety margins. The advantage of the design-by-analysis approach is that it covers geometric and loading configurations for which there are no design formulae. It contains provisions for the limit states that are relevant to disposal containers: ductile collapse (unstable gross plastic yielding), excessive local plastic strain (ductility exhaustion), fracture (brittle/ductile), and instability (buckling). A distinction is made between normal design and exceptional loads with regard to the safety factor. The conceptual designs can consider materials, corrosion, structural performance, weld design, inspection and manufacturing issues. The container requirements and the boundary conditions set by the WMOs and/or regulators form the basis of the conceptual design, and bespoke assessments define how these requirements are met or exceeded. The basic design focuses principally on meeting the requirements that are associated with maintaining structural integrity throughout the timescale being considered.

There is inevitably uncertainty in predicting the loading and environmental conditions acting on the container and changes in its materials’ properties over its lifetime. Conservative estimates may be made based on current knowledge but, if the extremes of all the factors are taken, this may result in a design that is unreasonable when the balance of risks is considered. To treat these uncertainties, one possible engineering approach is to define upper bound, lower bound and best estimate values, and to do a sensitivity analysis. A more sophisticated approach would be to define probability distributions to describe the uncertainties with a best estimate and standard deviation. It would then be possible to undertake a probabilistic analysis to determine the probability of a limit state being reached and to specify the acceptability of the design based on a limiting probability value. Another approach is to apply partial safety factors on the best estimate values. An example of a workflow diagram for the design of disposal containers is shown in Figure 7.

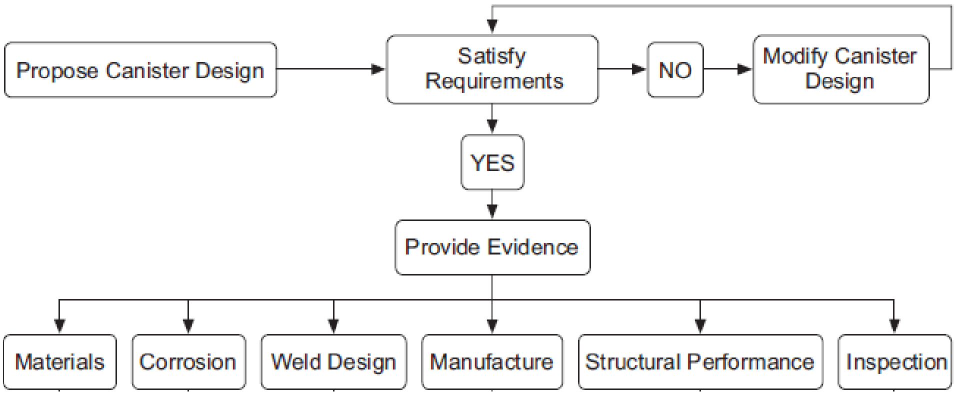

Figure 7: Example of workflow diagram for the development of SF/HLW disposal container concepts following the design-by-analysis approach (adapted from Patel et al. 2012).

Despite the lack of an internationally established design methodology and the variability of geological and regulatory boundary conditions, container designs developed for different national programmes are quite similar. Containers tend to be cylindrical in shape, with sizes defined by the dimensions of the waste, and are emplaced in the repository surrounded by a buffer material. See Section 3 for some typical examples.

6. Container fabrication

Container fabrication involves the application of conventional industrial casting, forging, welding, and inspection techniques. Although a high level of quality is required, these requirements do not exceed those of other safety-critical industrial components routinely manufactured for the nuclear, chemical, shipbuilding, and aerospace industries. Indeed, it is generally required that the containers can be fabricated and sealed using existing, proven methods. The only special requirement for HLW/SF containers is that the final sealing and inspection must be carried out in a shielded facility.

6.1 Fabrication of Container Components

The body and lid of the container can be pre-fabricated, inspected, and then delivered to the encapsulation facility. Depending upon the design, the container body can be fabricated using a number of conventional technologies, including (Patel et al. 2012):

- Casting
- Forging (either a single piece or of multiple pieces, with or without an integral base)
- Extrusion
- Pierce and draw (with or without an integral base)
- Welded rolled plate.

Because of the possibility of the preferential corrosion of welds and of the presence of defects, fabrication methods that limit the total number and length of welds are preferred. However, prior to loading of the waste, the body of the container can be heat-treated and inspected in a conventional facility to ensure the absence of defects and to impart the desired metallurgical and microstructural properties.

For dual-wall container designs, there is an additional cast iron or carbon steel inner vessel that supports the outer corrosion barrier. This inner vessel may be fabricated by any of the methods listed above; for example by casting, as in the case of the cast iron insert for the KBS-3 design, or from thick-walled carbon steel. If the two barriers are separate, then the outer corrosion barrier and inner structural vessel are fabricated separately and then typically assembled in the container fabrication facility after first loading the HLW/SF into the inner vessel. For coated container designs, it would be expected that the coating would be applied to the body and lid of the container prior to subsequent loading and assembly of the container in the shielded encapsulation facility.

In addition to the container body, insert, and lid, some type of internal structure is also generally required to support the SF assemblies or HLW containers inside the disposal container. In the KBS-3 design with the cast iron insert, this internal structure is provided by carbon steel channels that also serve as a mould for the casting process. Alternatively, a simple steel structure can be fabricated that fits inside the container, if required.

A number of full-scale containers and weld mock-ups have been fabricated in various national programmes. Over 50 cast iron inserts and a similar number of cast copper ingots and extruded copper tubes have been fabricated in Sweden and Finland as part of the KBS-3 container development, and dedicated fabrication laboratories have been built at Oskarshamn in Sweden and at the proposed Olkiluoto repository site in Finland. Prototype containers have also been fabricated in France (Andra 2005), Japan (JNC 2000) and Canada (Giallonardo et al. 2017).

6.2 Sealing

Following loading of the HLW/SF in a hot cell, the container must then be sealed remotely by a suitable welding operation. Some programmes specify two lids, an inner lid to prevent contamination within the shielded area, which may or may not be welded, and an outer welded lid that provides containment. A range of welding techniques have been investigated for the final closure weld, including:

- Electron beam (EB) and friction stir welding (FSW) of the 50-mm-thick copper shell in the Swedish and Finnish programmes (Cederqvist 2004, Raiko 2013)
- Narrow-gap gas-tungsten arc welding (GTAW, also referred to as tungsten inert gas TIG) and electron beam (EB) welding for 140-mm-thick carbon steel in the Nagra programme (Allen et al. 2016, Pike et al. 2010, Patel et al. 2012)
- GTAW and gas metal arc welding (GMAW, also referred to as metal active gas MAG) for the welding of 190-mm-thick carbon steel in the Japanese programme (Asano et al. 2005; 2006a,b)
- Hybrid laser arc welding (HLAW) for partial-penetration welds in carbon steel vessel of NWMO’s copper-coated container (Boyle and Martel 2015).

Some of these procedures require control over the environment during the welding process, such as the need for a vacuum for EB welding or the provision of a gas shield to avoid the incorporation of oxygen into FSW for copper (Björck et al. 2017, Raiko 2013).

Prior to welding, there may be a need to ensure an inert atmosphere inside the container or the absence of moisture. The possible presence of internal water is of particular concern when SF is removed directly from a storage pond and encapsulated without adequate drying or an extended period of interim dry storage. Water may then be entrained in damaged fuel pins resulting in fuel oxidation and/or internal corrosion of the container.

For coated container designs, the final fabrication step involves applying a coating over the seal weld between the body and lid of the inner structural vessel. For the copper-coated container design being developed by NWMO and Nagra, this final application would involve a cold spray technique, again applied remotely in a shielded facility.

Unless suitably treated, any welding will introduce residual stresses. This is a particular concern for the final closure weld, for which post-weld heat treatments might be difficult to perform. The presence of residual stresses in this region could adversely impact the long-term corrosion and mechanical performance of the container. Because FSW does not involve melting and recrystallisation of the metal, and because the parent material is in the annealed condition, the level of residual stress for the thick-walled copper corrosion barrier of the KBS-3 design is generally low (less than a few tens of MPa) (Raiko et al. 2010, Raiko 2013). Higher residual stresses can exist in thick-walled carbon steel container designs because of the nature of the welding techniques typically envisaged. Thermal stress relief may or may not be feasible, as the “soak temperature” required (typically in the range of 600°C) and the large thermal mass of the container may lead to temperatures that exceed the thermal limit for the waste (expected to be around 400-500°C, depending on the waste) (Patel et al. 2012). Non-thermal, surface stress relief techniques can be used in this case. These techniques induce compressive residual stresses to a depth of a few mm (King 2016). Ideally, the depth of compressive stress should be equal to or greater than the corrosion allowance, as otherwise there is the possibility of surface tensile stresses following a period of corrosion. For copper-coated designs, the as-deposited cold spray material requires annealing to restore sufficient ductility (Boyle and Meguin 2016).

6.3 Inspection

All fabricated components and closure welds will contain flaws. Whether these flaws are of a sufficient size to adversely affect the mechanical and corrosion performance of the container (i.e., whether the flaw should be classified as a defect) is assessed by performing an engineering critical assessment based on the loads to which the container will be exposed. Based on this assessment, the tolerable flaw sizes can be defined, which in turn define the required inspection capability.

Flaws in the body of the container and, especially, the final closure weld, can take a number of forms (BS 7910 2013):

- Volumetric flaws, such as large cavities or smaller regions of porosity (e.g., “worm holes”), solid inclusions, or areas of local wall thinning.
- Planar flaws, such as cracks, or lack of fusion.
- Shape imperfections, such as misalignment or imperfect profile.

In addition to their type, other important characteristics of flaws are their size, location (surface or embedded), orientation (radial, axial, circumferential), and the degree of interaction between them. Surface-breaking and embedded flaws will interact differently with applied loads, as will any flaw depending in its orientation. For corrosion-allowance materials, an embedded flaw could become surface-breaking after a period of corrosion. The prevalence of these different types of flaws will typically depend on the welding process, while the number of such flaws will typically depend on the quality of the inspection.

A minimum of two complementary inspection techniques would be expected to be required for both the pre-fabricated container components and for the final closure weld. An example of complementary techniques would be eddy current testing to detect surface-breaking flaws and phased-array ultrasonic testing to detect sub-surface volumetric and planar-type flaws. Many inspection techniques are available for both the pre-fabricated components and final closure weld. These include:

- Liquid penetrant for surface-breaking crack-like flaws
- Magnetic particle inspection for surface-breaking cracks in ferromagnetic materials
- Radiography for sub-surface volumetric flaws
- Phased-array ultrasonic testing to detect and size sub-surface volumetric and planar flaws
- Time-of-flight diffraction – an ultrasonic technique for improved sizing of sub-surface flaws
- Eddy current testing for surface-breaking flaws.

Inspection techniques can be assessed based on the probability of detecting and sizing of flaws, as characterised by the probability of identification (POI), probability of detection (POD), probability of correct rejection (POCR), and other similar measures. Because of the easier access and the greater range of applicable methods, flaws in the pre-fabricated components should be able to be detected and sized with a high degree of confidence. The need to inspect remotely in a high radiation environment will inevitably reduce the choice of inspection capabilities for flaws in the closure weld.

Whatever inspection methodologies are selected, it is necessary to be able to demonstrate that flaws larger than the critical flaw size can be detected with a sufficient level of confidence.

Assuming that defects (i.e., flaws that exceed the critical flaw size), can be detected, a decision has to be made about whether the defect can be repaired or whether the container needs to be scrapped or reworked.

6.4 Prefabricated emplacement modules (supercontainers)

In the case of PEMs, there is a further fabrication step required upon assembly, in which the HLW/SF container is embedded in the clay-based or cementitious buffer material. Figures 2 and 3 show the various components of the Belgian 'supercontainer' design (with integral cementitious buffer) and of the KBS-3H design (with horizontally-emplaced copper container and compacted bentonite rings), respectively. The additional steps of pouring the concrete backfill or of assembling the pre-compacted buffer rings around the container must obviously be performed in a shielded facility. Not only does this represent a fabrication challenge, but the resulting mass of the assembly (several tens of tonnes) must then be handled and moved from the surface facility to the underground disposal tunnels. Because of the relatively small size of CANDU SF bundles, and of the resulting container, the proposed NWMO buffer box arrangement results in a PEM design (Figure 4) that can be handled and emplaced with conventional fork-lift equipment (Birch and Mielcarek 2017). However, this advantage is the result of the small size of the container (approximately 2 m in length) and would not be present in the case of larger containers.

7. Container performance

7.1 Corrosion performance

7.1.1 Corrosion mechanisms and considerations common to different EBS designs

Containers envisaged for the disposal of HLW and SF are expected to be subjected to a range of potential corrosion processes. In most cases, the choice of the container and buffer material is designed to exclude or mitigate the possibility of either rapid and/or difficult to predict corrosion mechanisms and/or of factors complicating the treatment of such processes in performance assessments. In this section we first provide a high-level description of the main corrosion mechanisms of relevance to disposal containers. We then consider common strategies used to exclude or mitigate mechanisms potentially applying to a range of different EBS systems (particularly for RAC and MIC). Detailed considerations of the effect of corrosion mechanisms on different materials are then discussed in the next sections.

Uniform corrosion, resulting in the relatively even and gradual loss of material from a reactive surface, is generally the key corrosion mechanism of concern for most EBS designs. Detailed considerations about its occurrence in different systems are provided in the sections below.

Localised corrosion in the form of pitting or crevice corrosion[3] is the result of the spatial separation of the anodic (oxidation) and cathodic (reduction) reactions that constitute the overall corrosion process. Spatial separation can be caused by several different processes, including the localised breakdown of a passive film or due to geometrical factors. However, for such processes to occur, there must also be a sufficient driving force to sustain the electrochemical potential difference between anodic and cathodic processes, and for this reason localised corrosion is primarily an issue under aerobic (or more broadly oxidising) conditions. If spatial separation of anodic and cathodic processes is not maintained, corrosion tends to take the form of non-uniform corrosion rather than deep localised penetrations. In radioactive waste disposal, localised corrosion is typically mitigated through the choice of container material (an 'active' material such as copper or carbon steel), buffer (ensuring a 'non-passivating' environment, e.g., bentonite) and design choices (e.g., no mating joints). The occurrence and likely extent in different systems are discussed in detail below.

Environmentally-assisted cracking (EAC), is a general term to cover a range of potential cracking mechanisms including stress corrosion cracking (SCC) and hydrogen-induced cracking (HIC) requiring

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[3] Crevice corrosion occurs when there is close contact between the metal surface and another solid surface, resulting in the formation of an 'occluded' cell next to part of the canister surface, in which the chemical environment develops into a more aggressive environment than the bulk environment. Due to the design of the canister and the near-field environment such crevices are avoided.

due consideration when assessing the durability of disposal containers. The likelihood of cracking is typically mitigated by a suitable choice of material (more ductile materials such as copper and low-strength carbon steel are not very susceptible), environment (an anaerobic environment tends to reduce the risk of SCC, although might favour HIC), and manufacturing technologies (e.g., welding techniques and surface finishing able to reduce tensile stresses and the presence of crack-initiating defects). Detailed considerations about its occurrence in different systems are provided in the sections below.

Microbiologically-influenced corrosion (MIC) may occur either under biofilms if microbial communities colonise the container surface, or as a result of corrosive species produced by microbial activity further away from the container. MIC is primarily controlled by selection of the buffer materials, such that microbial activity is hindered in the vicinity of the container. Most disposal programmes achieve this by:

- using compacted bentonite of a sufficiently high dry density (generally accepted limit of 1.45 g/cm^3), which upon saturation will limit water activity and pore space and lead to a sufficiently high swelling pressure (Taborowski et al. 2019).
- controlling the chemical environment to make the nearfield inhospitable to microorganisms (e.g., by using high pH cement, Aerts 2009).

In the case of bentonite, assuming the emplacement process ensures a sufficiently high initial density (which is necessary to achieve the required swelling pressure upon saturation), neither microbial activity nor biofilm formation are considered to be possible in the buffer material or on the container surface. Therefore, microbial activity is only considered to be of concern if corrosive metabolic by-products, formed in regions of the repository where microbial activity is possible, subsequently diffuse to the container surface. A number of in situ experiments have included study of the potential for MIC of copper, carbon steel and other candidate container materials in conditions relevant to disposal with a bentonite buffer (Diomidis and King 2020). In general, MIC is not observed in sufficiently dense compacted buffer, whereas microbial activity can result in extensive corrosion in the absence of compacted buffer or buffer of a low density.

Radiation-assisted corrosion (RAC), which in the case of disposal containers could occur as a result of the generation of oxidising radicals due to γ radiation, is often designed out through the use of sufficiently thick disposal containers to reduce dose rates below levels of concern. Some studies suggest that dose rates expected in many container designs (< 1 Gy/hr) are insufficient to cause any observable effect (Shoesmith and King 1999). However, this is an area of ongoing investigation.

Galvanic corrosion, which is the accelerated corrosion of a metal when electrically coupled (typically joined or touching) with a more noble metal, is typically designed out in disposal containers by using a single metal for the corrosion barrier (e.g., copper shell). Galvanic corrosion, however, is particularly relevant to programmes (e.g., the Canadian programme) envisaging the use of a thin copper layer on top of a steel substrate. In this case, the greater nobility of copper could accelerate the corrosion of the steel substrate in the presence of a surface defect. However, the low redox conditions expected in the EBS in the long-term substantially decreases the impact of any galvanic coupling (Standish et al. 2016).

Corrosion at welds. Due to their differences in microstructure, crystallography and chemical composition compared to the parent material, welds and the heat-affected zone around them often tend to exhibit a different corrosion performance than the base metal. In combination with the propensity for flaws, and the difficulty of inspection (at least for the final closure weld), this makes the welds an area of increased concern. For carbon steel, the welds may exhibit slightly higher corrosion rates than the base metal, and slightly increased roughness (Reddy et al., 2021).

Atmospheric corrosion, which will occur on the disposal container during manufacturing, handling, emplacement in the GDF and during the early post-closure period, until all the available oxygen is consumed. The overall damage due to atmospheric corrosion in the early stages of waste disposal is limited, compared to the wall thickness loss due to uniform corrosion under anaerobic conditions and can easily be accommodated in the design of the container.

7.1.2 Copper

Copper has been considered as the preferred container material in a number of European and international waste management programmes, including in Sweden (SKB 2010a, 2019), Finland (Posiva 2021), Switzerland (Diomidis and King 2022), Canada (Hall et al. 2021), South Korea (Lee et al. 2011), and Taiwan (Hung et al. 2017). Pure copper is a "near-noble" material, meaning that it is essentially thermodynamically stable in repository environments in the absence of O_2, oxidising radiolysis products and sulfide ions (in the presence of which copper corrodes with the evolution of H_2). Invariably, copper containers would be used in conjunction with a bentonite-based buffer to restrict microbial activity and to limit the supply of remotely produced sulfide. Thus, in this environment, copper offers the advantage that the extent of corrosion is either limited, due to the limited amount of available oxidants (trapped O_2 and oxidants generated by the low gamma radiation fields typically present (Padovani et al. 2017), or occurs very slowly and predictably, in the case of the transport-limited corrosion by sulfide. The other primary advantage of the use of copper is that, because chloride ions are often dominant in deep groundwaters, copper corrosion in a disposal facility is generally uniform in nature with little tendency towards passivation and, hence, either localised corrosion or SCC.

As discussed in Section 4, the corrosion behaviour of the container is dependent on the nature of the near-field environment and how it evolves with time. Two important features of the near-field in the case of a copper container are the general lack of oxidants and the very slow rate of mass-transport imposed by compacted buffer material. For example, in various repository designs, the rate of supply of sulfide to the container surface is estimated to be of the order of 10^{-17}-10^{-16} $mol \cdot cm^{-2} \cdot s^{-1}$ (Cloet et al. 2017, King et al. 2017, Posiva 2021). Laboratory experiments to evaluate the corrosion behaviour of copper in relevant conditions have been typically conducted with higher sulfide concentrations and in bulk solution (rather than in bentonite), with a corresponding sulfide flux of 10^{-11}-10^{-10} $mol \cdot cm^{-2} \cdot s^{-1}$. As a consequence, corrosion processes that might be observed under laboratory conditions may or may not be relevant to a container in the repository. Therefore, for copper, as well as for other candidate container materials, great care should be taken when inferring the performance of the container based on the results of laboratory experiments.

As discussed in Section 3, there are two main designs for the copper corrosion barrier. In the KBS-3 container design, the corrosion barrier is 50 mm thick and vastly exceeds the amount of corrosion that could be conceivably induced by the initially trapped O_2 in the repository or the amount of oxidising radiolysis products that could be produced over the lifetime of the container. For the copper-coated container designs being developed in Canada and Switzerland, however, the corrosion barrier is only a few mm thick. Therefore, not only is it necessary to have more certainty about the rate and extent of corrosion but it is also more important to ensure that corrosion is uniform in nature rather than being localised, so as to avoid risks of localised perforations.

Uniform corrosion. Uniform corrosion of copper is possible under both aerobic and anaerobic conditions. The amount of trapped atmospheric O_2 is generally small, equivalent to a few tens of µm of uniform corrosion even if all of it causes corrosion. The extent of radiation-induced corrosion is also limited (again, of the order of a few tens of µm, (King and Behazin 2021) because the dose rates at the external surface of the container are generally <1 Gy/hr).

Sulfide ions, present either naturally in the groundwater or produced by the microbial reduction of sulfate in locations away from the container surface where such activity is possible, will also result in uniform corrosion of the container. Because of the restrictive mass-transport conditions in the repository, the rate of uniform corrosion of copper under anaerobic conditions is determined by the rate of supply of sulfide to the container surface (King et al. 2017). Transport control of the corrosion reaction has important consequences not only for the localised corrosion and SCC behaviour of the container (see below), but also for the properties of Cu_2S films that form as a result of uniform corrosion. At the extremely low sulfide fluxes expected under repository conditions, there is extensive evidence indicating that the Cu_2S films are porous rather than passive (Guo et al. 2021, Chen et al. 2014, Posiva 2021).

This distinction is important as passive Cu_2S films could otherwise render the underlying surface susceptible to localised corrosion and pitting.

Copper does not undergo sustained corrosion in the absence of O_2 (or of radiolytic oxidants) or sulfide. There have been claims that sustained copper corrosion with the evolution of H_2 is possible in O_2-free pure H_2O (Hultquist 1986, Szakálos et al. 2007), but extensive investigation has not been able to substantiate the original claims (Hedin et al. 2018). Neither is the presence of Cl^- ions in the groundwater (which stabilise dissolved Cu(I) in the form of cuprous-chloro complex ions) sufficient to shift the equilibrium in favour of the dissolution reaction to result in significant corrosion, even over repository timescales (Lilja et al. 2021). Both of these systems, Cu in O_2-free H_2O and high $[Cl^-]$, are examples of corrosion under “near-equilibrium” conditions (Posiva 2021). In other words, as corrosion proceeds and as corrosion products (dissolved Cu(I) and H_2) accumulate, the system shifts to a position of equilibrium and corrosion stops. In the case of Cu corrosion in O_2-free H_2O, the extent of corrosion necessary to bring the system to equilibrium is infinitesimally small (the equilibrium dissolved [Cu(I)] and H_2 pressure are 3×10^{-12} mol/L and 10^{-9} atm, respectively, at 25°C and pH 7) and corrosion would cease after a few atoms of corrosion.

Localised corrosion. Under aerobic conditions, mechanisms for the spatial separation of anodic and cathodic reactions are possible, resulting in the risk of both pitting and crevice corrosion. However, particularly in the presence of Cl^- ions, copper tends to corrode actively with little tendency for passivation (Qin et al. 2017). Only in moderately alkaline (pH >8), low $[Cl^-]$ (<0.01 mol/L) porewaters is passivation likely, but such conditions are considered to be unlikely in repositories considered in national programmes envisaging the use of copper containers (Briggs et al. 2021). Non-uniform wetting of the container surface, which is possible during the initial saturation transient period of the near-field evolution, may also lead to geometrical separation of anodic and cathodic processes due to the classic Evans’ droplet mechanism for atmospheric corrosion (Posiva 2021). Although the initiation of localised corrosion in these conditions is possible, there are many potential stifling mechanisms that will limit pit growth. Chief among these mechanisms is the consumption of the limited amount of trapped O_2 by processes other than container corrosion, such as aerobic microbial activity or the oxidation of Fe(II) minerals in the buffer and host rock. It is also possible that, depending on the relative rates of saturation and O_2 consumption, there may be no oxic corrosion phase since all of the O_2 may be consumed before the container surface is wetted sufficiently to sustain electrochemical corrosion processes (Posiva 2021).

Under anaerobic conditions, neither process for the spatial separation of anodic and cathodic reactions is likely. As noted above, Cu_2S films formed under repository conditions will be porous and non-protective, with no possibility of localised film breakdown. Equally, since the rate of uniform corrosion is transport limited, the interfacial HS^- concentration is zero and it is therefore impossible to establish a differential sulfide concentration cell on the surface on the basis of geometrical factors. Furthermore, even if localised corrosion does initiate, it will not be able to penetrate far beyond the uniform corrosion front because the sulfide concentration gradient between the mouth and the base of the pit will be practically zero and there will be no driving force for the transport of HS^- to the base of the pit.

For the mechanistic reasons discussed above, corrosion of copper containers is, at worst, expected to take the form of uneven uniform corrosion, rather than the more localised penetrations associated with permanent separation of anodic and cathodic reactions.

Microbiologically influenced corrosion. Like the majority of engineering alloys, copper and copper alloys are susceptible to MIC (Little et al. 1991). The main metabolic by-product of concern for copper containers is sulfide produced by the action of SRB in the excavation disturbed zone (EDZ) surrounding the deposition holes or tunnels in which the containers are emplaced. The extent of sulfate reduction may be limited by a number of factors, such as the availability of nutrients and electron donors or of sulfate ions, and the resulting supply of sulfide to the container surface will be further limited by the slow rate of mass transport through the compacted buffer.

Environmentally assisted cracking. The susceptibility of copper to H-related degradation mechanisms is limited due to the low solubility and diffusivity of H atoms in the fcc (face-centred cubic) matrix. Furthermore, high stress gradients for the concentration of H cannot be sustained for the low-strength copper alloys selected for use as container materials (with yield strengths of the order of 60 MPa). Therefore, although effects of absorbed H on the mechanical behaviour of copper can be observed under the aggressive H-charging conditions sometimes used in laboratory tests, copper containers are not considered to be susceptible to H-related degradation mechanisms under repository conditions (SKB 2019, Posiva 2021).

Copper is susceptible, however, to stress corrosion cracking. SCC of copper is known to occur under aerobic conditions due to the presence of ammonia (or ammonium ions), acetate, and nitrite (King 2021a). Cracking occurs via different mechanisms but, in all cases, a relatively positive corrosion potential is required, inferring the need for oxidising conditions in the repository. The electrochemical conditions associated with cracking indicate that the surface must be passive for SCC to occur, as well as the need for there to be sufficient SCC agent present at the container surface. These various requirements for SCC to occur in copper make it unlikely that the environmental conditions found in the laboratory to be necessary for cracking will exist at the container surface in the repository (King 2021a). Furthermore, apart from the effects of possible uneven buffer swelling, it is unlikely that there will be sufficient tensile stress for SCC (Salonen et al. 2021, Posiva 2021). Overall, therefore, SCC of copper containers under repository conditions is considered unlikely.

On the other hand, there have been reports of crack-like features formed on copper in the presence of a sufficient concentration of sulfide (Becker and Öijerholm 2017, Forsström et al. 2021, Taniguchi and Kawasaki 2008). However, other researchers have failed to observe cracking under similar conditions, while others suggest that the “cracks” are actually a case of intergranular attack at grain boundaries that are subsequently opened up by the excessive strains typically used in such experiments (SKB 2019; Taxén et al. 2018, 2019; Posiva 2021). Regardless of whether or not this is a true case of SCC, the various observations of the presence or absence of crack-like features can be rationalised in terms of a threshold sulfide flux (Salonen et al. 2021, Posiva 2021). Because the uniform corrosion of copper in sulfide environments is generally transport limited, propagation of a crack (or of any form of localised feature) ahead of the uniform corrosion front is only possible if the sulfide flux exceeds the transport-limited corrosion rate. Only then will there be a sufficient driving force for sulfide to be transported to the tip of the crack. Thus, the apparent threshold sulfide concentration reported by some researchers is in fact a threshold flux. Confirmation of this mechanism comes from the work of Taxén et al. (2018, 2019), who, having initially failed to observe crack-like features, subsequently added phosphate to their experiments in an attempt to exactly replicate the earlier work of Becker and Öijerholm (2017). Cracking was then observed at exactly the same sulfide flux reported by Becker and Öijerholm (2017), whereas no cracks had been previously found by Taxén et al in the absence of the phosphate. In addition to acting as a pH buffer (the original purpose for the phosphate addition), phosphate is also a corrosion inhibitor. The addition of phosphate, therefore, had reduced the rate of the interfacial sulfide reactions and the system was no longer under sulfide-transport control. The inferred threshold flux for the formation of cracks in sulfide environments is at least 4-5 orders of magnitude higher than the maximum flux of sulfide to the container surface expected in the repository, ruling out the possibility of SCC of copper containers under anaerobic conditions (Salonen et al. 2021, Posiva 2021).

Radiation-assisted corrosion. Gamma radiolysis of the near-field environment will produce oxidising radiolysis products which will contribute to the inventory of oxidants under aerobic conditions. However, the γ-dose rates for copper containers are generally low (<1 Gy/hr) and the extent of RAC will be limited (King and Behazin 2021). Similarly, the degree of ennoblement of the free corrosion potential (E_{CORR}) will be insufficient to induce localised corrosion even if the copper surface was passivated (not expected).

Galvanic corrosion. Galvanic corrosion between the copper corrosion barrier and steel or cast iron inner components is possible if there is a through-wall defect in the copper, but the effect is only significant under aerobic conditions (Smart et al. 2014, Standish et al. 2016). Such a situation is unlikely to exist in the repository, except possibly for an undetected defect in the thin copper coating of a coated container design in the early post-closure period (whilst oxygen is still present and radiation fields are high).

7.1.3 Carbon steel

Even though different WMOs specify different steel alloys for their container, in most cases low-strength, low-carbon alloys are considered, primarily due to their predictable corrosion performance and the reduced risk of HIC. Several corrosion mechanisms prevail during the disposal period, evolving with the near-field environment:

Uniform corrosion. This is the main corrosion mechanism expected on low carbon steel under long-term disposal conditions (i.e., saturated, anaerobic environments) proceeding through the reduction of water and the release of hydrogen (King 2008, Féron et al. 2009).

In the near-neutral pH environments associated with clays, the formation of corrosion products (e.g., oxides, carbonates, silicates) is unlikely to fully passivate the surface but may lead, over time, to a degree of protection, resulting in a decrease in corrosion rate (Martin et al. 2014, Necib et al. 2017, Taniguchi et al. 2010, Diomidis and Reddy 2022). The main solid corrosion product is magnetite, which in practice may be non-stoichiometric or may incorporate species from the buffer or host rock porewater. Uniform corrosion of carbon steel under anaerobic conditions is a relatively slow and predictable process leading to container wall thickness loss and a consequent increase of the risk of plastic collapse. As a result, it is primarily addressed by designing the disposal containers with sufficient wall thickness for the required lifetimes. However, the presence of hydrogen can lead to degradation of material properties and decrease of fracture toughness.

The primary concern related to uniform corrosion is the availability of corrosion rate data that are representative of disposal conditions and timescales. Since the corrosion rate decreases with time, as protective corrosion products develop on the steel surface, experiments with sufficiently long durations are needed, in order to increase the accuracy of extrapolation over repository time scales. Average corrosion rates of 1-2 µm/yr are generally reported (Martin et al. 2014), values that are typically obtained after a few years. However, the longest running representative experiment known to the authors exhibits instantaneous corrosion rates of 0.1-0.2 µm/yr which are still decreasing after 13 years (Diomidis and Reddy 2022). Some degree of non-uniformity of the corrosion processes (surface roughening) typically occurs and it is usually accounted for by an additional corrosion allowance when designing the container. With roughness values of a few 10s of micrometres, the overall impact of surface roughening is relatively small.

Corrosion of a steel container leads to the generation of hydrogen, which can be detrimental to the properties of the metal and give rise to mechanical-corrosion interactions. Hydrogen can also accumulate and lead to pressurisation of the EBS and, in the case of a low permeability host rock, the host rock itself (Diomidis et al. 2016). Soluble intermediate corrosion products (e.g., Fe^{2+} ions) are also typically formed (at least in near-neutral pH), which can diffuse into the bentonite buffer and interact with the clay minerals. Experimental evidence indicates that ferrous ions can exchange with interlayer cations in bentonite, but no mineral alteration takes place (Diomidis & Reddy 2022). Due to the limited solubility of Fe^{2+} at high pH values, such interactions are limited when a cementitious buffer is employed.

Localised corrosion. As in many other systems, localised corrosion may occur in the form of pitting or crevice corrosion when local breakdown of the passive surface film occurs. In bentonite, with its associated porewater pH value of ~8.4 and limited carbonate content, carbon steel is not passive (JNC 2000). Carbon steel is passive, however, in the alkaline pH of a cementitious buffer porewater (Sharifi-Asl et al. 2013). Several studies conducted in the framework of the Belgian radioactive waste disposal programme have investigated the risk of pitting of the carbon steel overpack and have concluded that, at the very low chloride concentrations expected in a Belgian GDF (Boom Clay) the risk is very low (Smart et al. 2017, Kursten et al 2017). In principle, pitting could occur in the presence of both bentonite and cement for the short period of aerobic conditions in the GDF. However, any localisation of corrosion can be taken into account through the use of a suitable ‘pitting factor’ or extreme value analysis. Such approaches indicate that, as the overall depth of corrosion increases, the degree of corrosion localisation is expected to decrease (Féron et al. 2009, Landolt et al. 2009, Taniguchi et al. 2011). In some cases the pitting factor is assumed to include a contribution for MIC, since components buried in the sub-surface environment are arguably exposed to the same, or higher, degree of microbiological activity than metallic containers in an underground repository (Féron and Crusset 2014).

Microbiologically influenced corrosion. As in the case of copper, with the possibility of the growth of microbial communities to form biofilms on the container surface being very small, the only MIC-related concern for carbon steel containers is the diffusion of metabolic by-products from locations further from the container. Sulfide is the primary corrosive agent that could result from microbial activity (associated with SRB). Due to solubility limitations and the slow diffusion processes of sulfide in a bentonite buffer the flux of sulfide to the container surface is expected to be extremely small (see Section 7.1.2). As a result, the overall wall thickness loss due to sulfide-induced corrosion is several orders of magnitude smaller than the loss due to oxidation by water.

Hydrogen-induced cracking. Steels are susceptible to HIC, and hydrogen produced by anaerobic corrosion may lead to increased hydrogen partial pressure at the container surface. Some of this hydrogen will enter the steel in the form of H atoms and progressively diffuse through to the interior of the container. However, most of the hydrogen atoms generated will recombine to form molecular hydrogen in solution (Turnbull 2009). Hydrogen present in the steel will be distributed between lattice and microstructural trap sites, with the greatest concentration in the latter for the temperatures expected under repository conditions. Trap sites include grain boundaries, phase boundaries, dislocations, vacancies and solute atoms. The result of hydrogen entrapment is a loss of ductility and fracture toughness of the material. Cracking will occur if the stress intensity factor is higher than a threshold value, which can occur due to residual or applied stresses. Measures to decrease the risk of cracking include selection of a steel alloy with no or low susceptibility to hydrogen embrittlement (low strength steels) or designing the container so that it is structurally sound when considering the properties of the embrittled material.

Stress corrosion cracking. Carbon steel is known to be susceptible to SCC in a number of environments similar to those expected in the repository. Near-neutral pH SCC has been found to occur in dilute bicarbonate solution (in the range 0.001-0.01 mol/L) with a pH in the range 5.5 to 7.5 and electrochemical potentials corresponding to those of anaerobic environments. Cracking in these conditions is promoted by cyclic loading, which is not expected on disposal containers. High-pH SCC occurs in concentrated carbonate/bicarbonate solutions (0.1 to >1 mol/L) at pH values > 9.3. Under these conditions, the steel surface is passive. Cracking occurs in a specific range of potentials (e.g.,-0.525 to -0.675 V vs SCE at room temperature), with crack growth rates increasing with temperature. Even at high pH, ongoing work suggests that cracking is unlikely at the low chloride concentrations assumed in a Belgian disposal facility (Kursten and Gaggiano 2017).

Results from French (Didot et al. 2017, Necib et al. 2017) and Japanese (Ogawa et al. 2017) programmes provide evidence for crack initiation on carbon steel under severe loading conditions when exposed to repository-relevant environments. However, no evidence for sustained crack growth exists.

7.1.4 Passive alloys

Stainless steels have seldom been considered for the disposal of HLW/SF because of perceived risks associated with localised corrosion. These risks, however, depend on the corrosivity of the groundwater (e.g., its chloride content), whether any corrosive groundwater would come into contact with the container before or after oxygen is consumed (and more broadly, before or after low Eh conditions have been established), and on the efficacy of buffer materials (e.g., cements) in inhibiting localised corrosion and MIC. The rapid consumption of oxygen observed in many in situ experiments would suggest the risk of localised corrosion might be modest (especially with a cement buffer), risks associated with MIC (typically managed through the use of a bentonite buffer) would also need to be ruled out. Currently the

Czech programme envisages the use of a stainless steel inner vessel, a carbon steel outer vessel and bentonite buffer (Pospiskova et al. 2017).

Nickel alloys can be divided into six groups based on their elemental composition, but only those containing Ni-Cr-Mo have been typically considered for use in HLW/SF programmes (King, 2014a). As for stainless steels, in sufficiently corrosive conditions these alloys may exhibit localised corrosion. However, the use of very resistant grades can substantially mitigate this risk. The general compositions and international naming conventions of these alloys are given in Table 2.

There is a wide range of titanium alloys commercially available. Those considered as possible container materials can be split broadly into single-phase alloys (containing α-phase only) and dual phase alloys (containing α-phase and β-phase). Commercially pure (CP) titanium and various titanium alloys (Grades 1, 2, 5, 7, 9, 11, 12, 16, 17 and 24) have been considered for use in several national nuclear waste management programmes (Kursten et al., 2004, Johnson et al., 2004, DOE, 2008, Hua et al., 2005, King, 2014b and Holdsworth et al, 2018). With the possible exception of crevice corrosion and HIC, titanium alloys are considered resistant to most forms of corrosion expected in a GDF, including MIC.

Table 2: Composition of nickel alloys considered for use in various HLW/SF programmes.

Alloy	**UNS**	**Alloy Class**	**Ni**	**Cr**	**Fe**	**Mo**	**C**	**Si**	**Mn**	**Ti**	**Other**
Alloy 22	N06022	Ni-Cr-Mo	56.0	22.0	3.0	13.0	0.015	0.08	-	-	3.0 W
Hastelloy C-4	N06455	Ni-Cr-Mo	Bal.	14.0 - 18.0	3.0	14.0 - 17.0	0.015	0.08	1.0	0.70	2.0 Co. 0.03 S, 0.04 P
Hastelloy C-276	N10276	Ni-Cr-Mo	Bal.	14.5 - 16.5	4.0 - 7.0	15.0 - 17.0	0.02	0.08	1.0	-	3.0-4.5 W, 2.5 Co, 0.35 V, 0.03 S, 0.03 P
Alloy 625	N06625	Ni-Cr-Mo	58.0 min	20.0 - 23.0	5.0	8.0 - 10.0	0.10	0.5	0.5	0.4	0.4 Al, 0.015 S, 3.15-4.15 Nb
Alloy 825	N08825	Ni-Fe-Cr-Mo	36.0 - 46.0	19.5 - 23.5	24 - 40	2.5 - 3.5	0.05	0.5	1.0	1.6 - 1.2	1.5-3.0 Cu, 0.2 Al, 0.03 S

Advantages over carbon steels and copper alloys

The main advantage of using passive alloys as opposed to carbon steel or copper alloys is the improved resistance to uniform corrosion conferred by the presence of a passive film. Nickel alloys and stainless steels gain their passivity from the formation of a chromium rich oxide/hydroxide passive film, which is thermodynamically stable over a wide range of pH and potential values that span those anticipated in a GDF (see Figure 8). Whilst this passive film is present, the uniform corrosion rate in both oxic and anoxic environments is low and decreases with increasing chromium content of the alloy. The passive corrosion rate increases with increasing temperature and is typically in the range of sub-nm/y to 10s of nm/y in simulated GDF environments (Enos & Bryan, 2015).

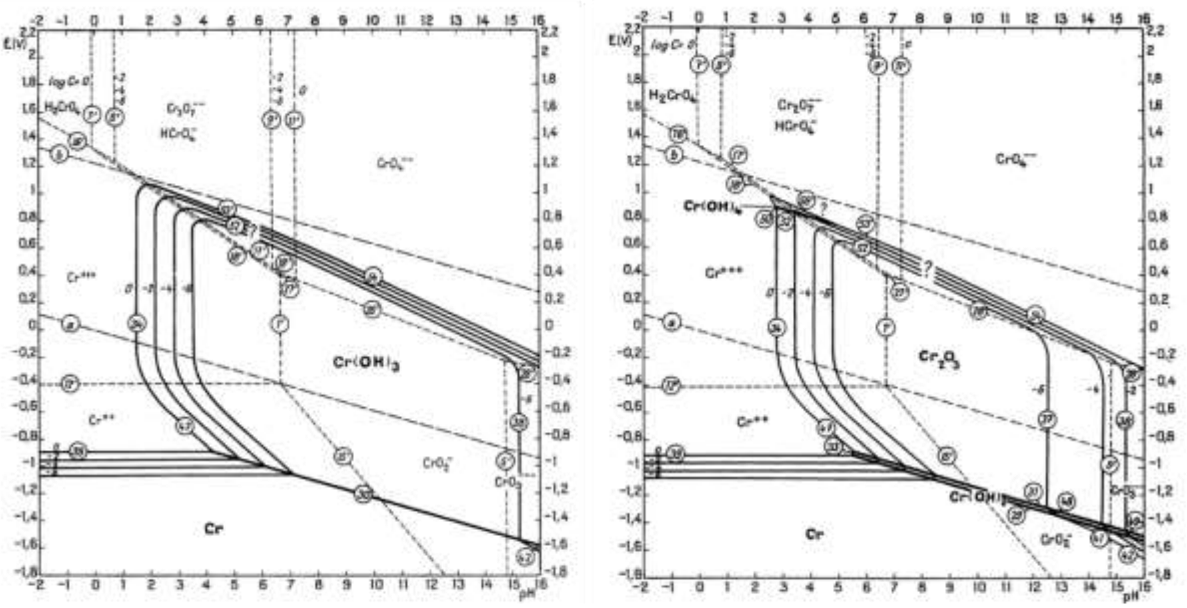

Figure 8: Potential-pH equilibrium diagram for the system chromium-water, at 25 °C. In solution containing chloride, established considering Cr(OH)$_3$ (left) and anhydrous Cr$_2$O$_3$ (right) (Pourbaix 1974)

Titanium alloys form a protective TiO_2 film over a broad potential and pH range (see Figure 9) extending beyond those anticipated in a GDF, including those of early post-closure in which oxygen may be present. Typical corrosion rates for titanium alloys exposed to environmental conditions anticipated in a GDF are of the order of 10s of nm/y (Hua et al., 2005). In environments consisting of compacted bentonite saturated with dilute synthetic ground water at 95°C, the corrosion rates of a commercial grade (Grade 2) and of a more expensive Ti-0.2Pd alloy were estimated as being of the order of 1 nm/y following 6 years of exposure (Mattsson and Olefjord, 1984, 1990).

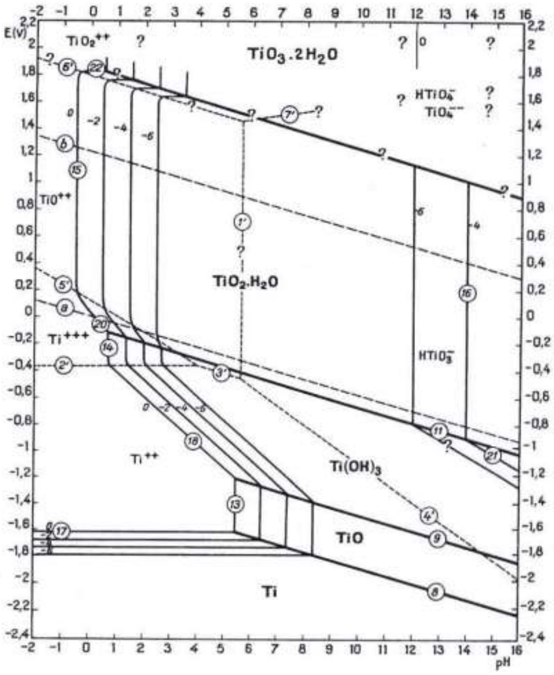

Figure 9: Potential-pH equilibrium diagram for the system titanium-water, at 25 °C assuming TiO_2 H_2O to be the stable solid phase (Pourbaix, 1974).

Titanium has been shown to be resistant to attack by sulfur species such as H_2S, elemental sulfur and thiosulfates (ASM, 2005) and the stability of the TiO_2 film confers a high resistance to depassivation in acidic conditions (Hua et al., 2005). These characteristics make titanium highly resistant to MIC, which typically proceeds due to the formation of sulfides or organic acids. Nickel alloys also exhibit a high resistance to MIC due to the stability of their passive films (Lloyd et al., 2004).

Both titanium and nickel alloys are highly resistant to SCC in the conditions anticipated within a GDF (Kursten, et al., 2004). SCC testing of titanium in the presence of concentrated brine at temperatures between 90 – 200°C showed no initiation of SCC (Kursten, et al., 2004). Immunity to SCC was also observed following U-bend tests performed in brines simulating Yucca Mountain environments (DOE, 2008).

Disadvantages over carbon steels and copper alloys

A key disadvantage of using passive materials over copper and carbon steel is that the corrosion resistance and corrosion rate are less predictable. This is primarily due to the risk of localised corrosion, which can increase the rate of wall penetration by many orders of magnitude compared to conditions where the alloy remains passive. This makes it necessary to pay careful consideration to the electrochemical environment with regards to the stability of the passive film. The main risk of localised corrosion to both titanium and nickel alloys is from crevice attack. Titanium alloys have been shown to be susceptible to crevice corrosion in oxidising environments in the presence of Cl^- at elevated temperatures (90-100 °C), but the degree of susceptibility is heavily influenced by the alloy composition and the chloride concentration (King 2014b). Crevice repassivation potentials have been established for a wide range of nickel alloys as a function of temperature and environmental chemistry; an illustrative example of the effect of temperature and chloride concentration for Alloy 22 is given in Figure *10* (Cragnolino et al., 1999).

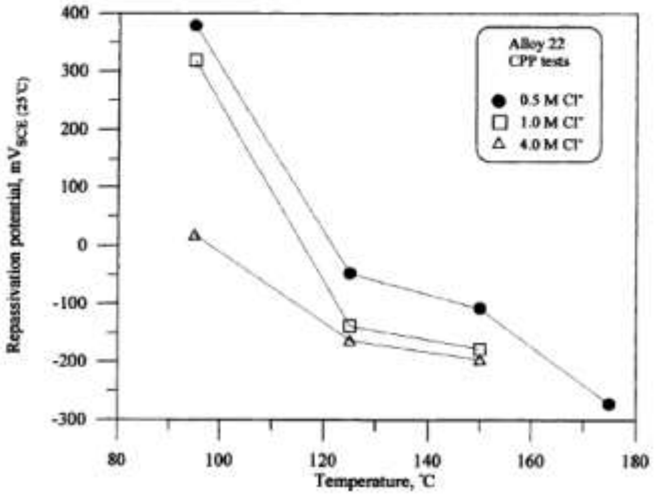

Figure 10: Effect of temperature on the crevice re-passivation potential of Alloy 22 in various chloride solutions (Cragnolino et al., 1999).

Titanium alloys are susceptible to hydrogen induced cracking (HIC) in certain environments. This can arise due to the formation of brittle hydride phases if enough hydrogen is absorbed, which could occur during localised or uniform corrosion, or during cathodic charging, which could occur when coupled to a less noble metal (e.g., Al, Mg, Zn). Once sufficient hydride phases have formed, the susceptibility of the alloy to fast fracture is markedly increased, with the threshold depending on the exact alloy composition (Hua et al., 2005, King, 2014b). HIC could be of particular concern if crevice corrosion is possible as the reduction of protons and the absence of a passive film within the acidified occluded region can lead to extensive hydrogen absorption (Yan et al. 2011).

7.2 Mechanical Performance

The key mechanical failure modes and degradation mechanisms that are of concern for HLW/SF containers are described below. Other modes of mechanical failure such as fatigue and erosion are omitted from the discussion, as they are prevented by the design of the container and service conditions. The behaviour of specific materials is not considered, as the methods of analysis are applicable to most standard engineering materials.

- **Plastic collapse (Mechanical overload)**

Mechanical overload occurs when the container is subjected to stresses exceeding its yield stress resulting in plastic deformation. This can happen at a large scale and can cause plastic collapse of the container or can happen at a small scale resulting in local plastic strain.

In a GDF, mechanical loading is anticipated to arise from buffer swelling (for repositories that utilise a compact clay buffer), hydrostatic pressure, lithostatic pressure (depending on the host rock) and seismic activity (i.e., rock shear), as outlined in Section 4.1. The extent of mechanical loading that arises via these mechanisms has been evaluated against the relevant mechanical properties of container materials for a range of disposal concepts as discussed in further detail below.

Finite element analysis of the susceptibility to the steel container described in the Swiss disposal concept demonstrated that, under conservative loading conditions combined with conservative corrosion losses (20 mm and 40 mm), the maximum stresses were less than 1.4 times lower than the limit load as determined by EN 13445 (Patel, et al., 2012). Loading of the container was bounded by considering anisotropic pressures of 29 MPa and 22 MPa in the horizontal and vertical directions respectively, that were selected based on the possible in situ stresses in Opalinus Clay at a depth of 900 m. The mechanical properties used in the analysis were those of the selected steel, grade taken as having a yield strength of 220 MPa at room temperature and 175 MPa at 150 °C, corresponding to the maximum expected container temperature of 130-140°C (Patel, et al., 2012).

For copper containers that rely on a cast-iron insert to resist collapse under loading, consideration of the load bearing capability of the insert are considered independently of the copper outer layer (Jonsson et al. 2018). For Swedish repository conditions, the maximum external pressure a container is expected to be exposed to is 45 MPa, arising from swelling of the bentonite buffer, and hydrostatic pressure of the groundwater (including an additional hydrostatic contribution from glacial water during a hypothetical future glaciation event). The stress on the container is increased beyond that imposed by hydrostatic loading when also considering asymmetrical expansion of the buffer and shear of the host rock due to seismic activity (Jonsson et al. 2018). The collapse pressures for disposal containers designed to hold BWR and PWR were found to be 99 MPa and 128 MPa respectively, which gives a robust safety margin against the maximum hydrostatic loading of 45 MPa. Under the most severe uneven buffer swelling loading, the estimated bending stress of the container is below the yield strength of the insert. Similarly, under a shear displacement of 5 cm applied at ¾ height of the container, the maximum stress is below the insert yield strength. Given that bending of the container and shear of the host rock are considered low probability events, the simultaneous occurrence of both situations is not considered. For copper-coated containers, it is the mechanical performance of the underlying substructure that is primarily of interest (Diomidis and King 2022).

- **Creep**

Creep is the time-dependent plastic straining of a material subjected to sustained stress levels that are below the yield strength. Creep is highly sensitive to temperature and occurs when diffusion processes of lattice defects contribute significantly to material flow at stress levels below yield (above yield, dislocation movements are activated). As a rule of thumb, this typically occurs when the temperature of a material reaches 30-40% of its melting point (on the Kelvin scale).

The creep process is often subdivided into three stages:

- Primary creep – the rate of strain decreases with increasing time; for metals, this is attributable to strain hardening in the material.
- Secondary creep (or steady state creep) – the rate of strain is constant and driven by the diffusion of lattice defects; in many cases this is the most important stage when determining the service lifetime of components subjected to creep.
- Tertiary creep – the rate of strain increases with time until fracture occurs.

Creep under loads imposed within a GDF is a key consideration for dual-wall copper containers such as the KBS-3 concept. The creep properties of copper under repository conditions have been studied extensively over the past 35+ years and have focussed on alloy composition (including the sulfur and phosphorus content), microstructure (e.g., grain size, and welding and cold work), effect of temperature, effect of loading rate, influence of hydrogen pickup, and effect of processing line inclusions and oxide streaks (Andersson-Östling, 2020).

In the dual wall design, during the development of swelling loads and hydrostatic loads, the copper container is anticipated to deform around the inner cast iron insert via creep processes; compared to the copper, the extent of creep of the cast iron at repository temperatures is negligible. Consequently, creep of the copper is considered a beneficial process, since it accommodates plastic deformation that might otherwise result in cracking. For this reason, the KBS-3 concept utilises a phosphorus-doped oxygen free copper alloy, which has more favourable ductility under creep loading than conventional oxygen free copper. The addition of 50 ppm of phosphorous to oxygen free copper has been found to increase the creep ductility from between 0-10% to between 30-50% at temperatures below 300 °C (Andersson-Östling, 2020).

The creep properties have been evaluated through modelling and experimentation, and consider both the base metal, the weld metal (from the friction stir weld) and material from the adjacent heat affected zone (HAZ), with experimental work typically being accelerated by being performed at temperatures greater than those anticipated in the repository. For phosphorus-doped oxygen-free copper, the base metal, weld metal and HAZ all exhibit good creep ductility, with experimental testing demonstrating a tendency for failure to occur in the HAZ as opposed to the base metal or weld (Andersson-Östling, 2020).

In the KBS-3 concept, loading arising from bentonite swelling and hydrostatic forces is expected to cause a pressure on the container of 10-18 MPa. These pressures are low in comparison to the strength of the material, which yields at around 70 MPa. As a result, secondary creep is considered to be negligible in the loading conditions expected in the GDF. Of more relevance is primary creep and the strain induced by progressive loading during initial saturation of the buffer (Andersson-Östling, 2020).

- **Fracture**

Fracture describes the process by which a material is separated into two or more pieces by the propagation of a crack. Cracks are often initiated at regions of high local tensile stress, such as a defect (e.g., a corrosion pit) or another stress raising feature (e.g., a weld root). The presence of a crack in a tensile stress field increases the magnitude of the stress ahead of the crack tip enabling continued propagation. Crack arrest can occur if the crack propagates into a region of reduced tensile stress (e.g., due to changing geometry, or from opposing compressive stresses such as those encountered in a peened surface). Cracking can occur due to externally-applied stresses that are static, quasi-static or dynamic (including cyclic loading i.e., fatigue), or due to residual stress (e.g., induced by welding).

The risk of fracture due to the presence of residual stress at the final sealing weld was evaluated for the steel container design used in the Swiss concept by calculation of the critical flaw size (Patel, et al., 2012). Finite element analysis was used in conjunction with engineering critical assessment following the failure assessment diagram approach described in BS 7910. For the most severe conditions, which assumed a residual stress equal to the yield strength of the material (220 MPa) and a fracture toughness of 85 MPa$\sqrt{m}$ (which includes the effect of hydrogen, see Section 7.1.3), the critical flaw size for fracture was 22 mm. Given the common practice of adopting a safety factor of 10 between the critical flaw size and the maximum tolerable flaw size, this would give a maximum tolerable flaw of 2.2 mm. For a residual stress of 70% of the yield strength the critical flaw size for fracture is increased to 35 mm (Patel, et al., 2012).

For copper-coated steel containers as described in the Canadian concept (Keech et al., 2021), a key consideration is that the ~3 mm copper coating is expected to have the strength, ductility, and adhesion to resist loading in a GDF environment without fracture. In this disposal concept, as in many other concepts envisaging the use of a bentonite buffer, the main source of pressure is from swelling of the bentonite upon hydration, accompanying the hydrostatic load associated with groundwater at depth (which could include an additional glacial load). Under these conditions, it is expected that the coating will undergo a total strain of the order of 1% (Boyle & Shaker, 2015). Further evaluation by finite element modelling indicated that electrodeposited copper on a steel substrate exhibited superior mechanical properties to wrought SKB OFP-copper in terms of both strength and ductility. Cold sprayed copper (envisaged to protect the weld regions) proved to be unsuitable in its as-deposited form but was substantially improved after annealing. For cold sprayed copper, the conditions of the post-manufacture annealing treatment governed whether fracture occurred at the copper-steel interface or within the copper coating, and optimisation of this process was highlighted as a key activity (Boyle & Shaker, 2015).

- **Impact loads**

The main sources of impact load anticipated for radioactive waste containers arise during handling prior to emplacement (e.g., being dropped). The damage that a container undergoes during impact loading is complex as it is heavily dependent on the exact mechanics of the impact (e.g., speed, orientation, energy of collision, nonlinear material properties), which necessitates the use of dynamic finite element analysis to model effectively. Resistance of a container to impact is often evaluated experimentally using a standard test methodology such as drop tests, which can be designed to simulate different scenarios (Hawkins, 2019). However, models are also used to predict the stresses arising from different drop scenarios. For example, the SF container developed by Nagra was used to demonstrate that the impact from a 0.5m drop onto concrete would not lead to breaching (Patel, et al., 2012).

8. Lifetime Prediction

8.1 Lifetime Prediction Methodology

8.1.1 Introduction

It is both unlikely and, from the standpoint of the radiological consequences, undesirable, for there to be a single container lifetime. Instead, there will be a distribution of container lifetimes due to the spatial variability of environmental conditions within the repository and because of variability in the properties of the container (for example, due to the presence and nature of flaws). Furthermore, despite the wealth of available knowledge on the properties and performance of the container, there inevitably are remaining uncertainties. It is the purpose, therefore, of the lifetime prediction models described below to capture these sources of variability and uncertainty and to account for them in the prediction of the distribution of container lifetimes.

The importance of the container and its predicted lifetime differs for the various types of disposal concepts and host rock types. For permeable crystalline host geologies, the delay in the release of radionuclides due to long-lived container may be an important contributor to ensuring that the dose to the specified receptor remains below the regulatory limit. For such cases, a robust and justifiable lifetime prediction is important and, in many cases, a long container lifetime (greater than, say, 100,000 years) is desirable to order to demonstrate the safety of the disposal system. In low-permeability sedimentary host rocks, on the other hand, the geological barrier itself provides sufficient retardation of the transport of radionuclides that the container lifetime has little influence on the ultimate dose that future human generations (and surface biota) might be exposed to as a result of the release of radioactivity from the GDF. Therefore, there is no radiological benefit from a long-lived container provided the containment remains intact for the required period of retrievability (if any) and for the initially hotter period (thermal-period), in which degradation processes of the EBS and transport processes of the radionuclides (including in the gas phase) might be accelerated.

8.1.2 General considerations

Container lifetime prediction models generally serve one of two purposes. Firstly, to build the overall safety case in support of the proposed disposal system, it is desirable to demonstrate a thorough understanding of the underlying mechanisms of the different corrosion processes, often supported by detailed deterministic models on a microscopic, if not atomistic, scale. Such detailed models are typically too complex, however, to use in a safety assessment of the radiological consequences of container failure and subsequent radionuclide transport, and for such purposes an abstracted or simplified lifetime model is usually required.

In developing a container lifetime prediction model, the various corrosion processes can be divided into two types. For those corrosion processes that will occur and which will result in penetration of the corrosion barrier (or corrosion allowance), a quantitative measure of the extent or rate of corrosion is required. For other corrosion processes that are considered unlikely to occur, it is necessary to develop a ‘reasoned argument’ for excluding the process from the lifetime prediction. The reasoned argument (also referred to as the ’exclusion principle‘) may or may not involve a quantitative assessment of the extent of corrosion.

8.1.3 Mechanical versus corrosion degradation modes

Typically, containers will fail by mechanical overload following a period of corrosion, during which the load-bearing component is reduced in thickness by uniform corrosion. (The one exception to this scenario is if the wall is penetrated locally by a deep pit or crack, in which case the container structural integrity may be maintained but containment is lost). To account for these two degradation modes, it has been usual to define a minimum wall thickness necessary for mechanical stability, with the remaining wall thickness designated as a corrosion allowance. The container lifetime can then be assessed based on the time required to consume or to penetrate the corrosion allowance. This approach is appropriate for dual-wall container designs where the two components serve different functions; the outer layer acting as the corrosion barrier and the inner component providing structural support (Keech et al. 2021, Posiva 2021, SKB 2010b). However, a similar approach has also been used for carbon steel containers where the one component provides both corrosion and mechanical resistance (Ogawa et al. 2017).

More recently, attention has been focussed on the effects of joint mechanical-corrosion degradation modes on container integrity (King et al. 2016). Such interactions are generally limited for dual-wall container designs (Posiva 2021), but can be important for single-shell designs, particularly if there is time-dependent degradation of the material properties. An example of such a situation is the corrosion of carbon steel containers under anoxic conditions and the subsequent absorption of hydrogen leading to a time-dependent reduction in the ductility and fracture toughness of the material (King et al. 2014b). For these circumstances, it is necessary to take into account both corrosion and mechanical aspects in order to predict the container lifetime.

8.1.4 Definition of container failure

In order to estimate the container lifetime, it is first necessary to define what is meant by container ‘failure’. The simplest, and most conservative, definition of failure is penetration of the corrosion barrier, although, for a dual-wall container design this may not result in contact of the waste by incoming buffer porewater. This definition also takes no account of the mode of failure and the residual barrier function provided by the ‘failed’ container. For example, if penetration of the corrosion barrier is the result of isolated pits or tight SCC cracks then the remainder of the container wall still provides a significant mass-transport barrier to the ingress of water and the egress of radionuclides. Credit has been taken for this continuing barrier function of a nominally penetrated container wall in some safety assessments (DOE 2008). Usually, however, it is conservatively assumed that the entire container effectively disappears at the time of the initial penetration.

8.1.5 Lifetime prediction methods for different forms of corrosion

Many different approaches have been taken to the prediction of the evolution of the corrosion behaviour and of the ultimate container lifetime. The discussion here addresses both the more-detailed modelling approaches used to build confidence in the overall safety case, as well as the abstracted modelling approaches used to predict container lifetimes for the safety assessment. Table 3 summarises these various approaches for the four container materials considered here (copper, carbon steel and cast iron, Ti alloys and Ni alloys) for the four main types of corrosion of interest (uniform corrosion; localised corrosion in the form of pitting, crevice corrosion, or surface roughening; EAC in the form of either SCC or H-related degradation mechanisms; and MIC).

Uniform corrosion

Uniform corrosion occurs under both aerobic and anaerobic conditions. The most common approach used in safety assessments in the case of aerobic conditions resulting from the initially trapped atmospheric O_2, is simply to use a mass-balance argument to calculate the maximum depth of uniform corrosion. These conservative estimates can be supported by detailed reactive-transport models which, in part, can be used to assess what fraction of the initial O_2 may be consumed by processes other than container corrosion, such as the oxidation of Fe(II) mineral phases in the buffer or host rock or by aerobic microbial processes. For the permanently aerobic conditions of the unsaturated repository at Yucca Mountain, the corrosion rate of the Alloy 22 waste package and Ti-7 drip shield were based on empirically measured values.

Since the external γ-radiation fields are small, it has been argued that the effect of irradiation is insignificant and that it can be excluded from consideration (Johnson and King 2003, Ogawa et al. 2017, Suzuki et al. 2021). Alternatively, the yield of oxidising species can be predicted based on radiolysis models and used to predict the extent of corrosion (King and Behazin 2021, SKB 2010b).

Under anaerobic conditions, the corrosion rate of carbon steel is typically based on empirically measured values from long-term experiments (Andra 2005, Diomidis and Johnson 2014, Ogawa et al. 2017). Reactive-transport models have also been developed for the uniform corrosion of carbon steel in both bentonite (King et al. 2014a) and cementitious buffer (Lu et al. 2014). Copper is essentially thermodynamically stable in the anoxic repository environment, except in the presence of sulfide. The corrosion rate is determined by the rate of supply of sulfide to the container surface (King et al. 2017) and various reactive-transport models have been developed to assess the extent of corrosion due to microbially produced sulfide (Briggs et al. 2017; Cloet et al. 2017, Idiart et al. 2019, King et al. 2021, Pekala et al. 2019). Depending upon the repository design and the location of the sulfide source with respect to that of the containers, interesting 3-D effects are predicted (Briggs et al. 2017, Pekala et al. 2019, Posiva 2021), with the depth of corrosion varying over the surface of the container.

Localised corrosion

The three forms of localised corrosion that have been assessed for different container materials are: surface roughening, pitting, and crevice corrosion. Surface roughening results from the non-permanent spatial separation of anodic and cathodic reactions and occurs for the corrosion allowance container materials copper and carbon steel. For copper, a surface roughening allowance is generally defined based on empirical data. For carbon steel, a cellular automata model has been developed based on the principle of shifting anodes and cathodes (Pérez-Brokate et al. 2017).

Even though copper and carbon steel are more likely to undergo surface roughening than discrete pit formation, various approaches have been developed to predict the extent of pitting for these materials. Empirical pitting factors, the ratio of the maximum to the average penetration depths, have been used for both copper (Werme et al. 1992) and carbon steel (Féron et al. 2009, JNC 2000), typically based on measurements from archaeological artifacts and more-recently buried objects. In the case of carbon steel, the pitting factor decreases with increasing average penetration depth (Féron et al. 2009), which is consistent with deep pits degenerating into surface roughening as the extent of corrosion increases. The same database of pit depths from long-term buried objects can be analysed statistically using Gumbel-type extreme-value distributions (King and LeNeveu 1992, Ogawa et al. 2017) and then using them to predict the depth of the deepest pit on the container.

The methods described above are focussed on predicting the extent of propagation. Alternatively, the likelihood of pit initiation can be assessed electrochemically by comparing the predicted corrosion potential (E_{CORR}) to the film breakdown potential (E_B). This approach presupposes that the container surface is in the passive state. Since pit initiation is a stochastic process, it is appropriate to use a probabilistic approach to account for the uncertainty and variability in E_B and E_{CORR}, as well as the conditions that lead to passivation. Briggs et al. (2021) describe a probabilistic model for assessing the likelihood of pit initiation on copper under aerobic, saturated conditions, with the extent of pit growth based on empirical propagation rates.

The assessment of pit initiation for passive alloys has also been based on electrochemical principles and, in particular, whether the value of E_{CORR} exceeds the repassivation potential (E_{RP}), which is a more conservative criterion for initiation than E_B. For Ti alloys, E_{RP} has a value of several volts in repository-relevant environments and the possibility of pit initiation can be excluded (Hua et al. 2005). Pitting is far more likely for Ni-based alloys, but the focus of localised corrosion models for these materials has been on crevice corrosion (Hua and Gordon 2004), which initiates at lower potentials than pitting corrosion.

Because corrosion initiates more readily in occluded regions than on exposed surfaces, the localised corrosion behaviour of Ti- and Ni-based has been assessed based on crevice corrosion rather than pitting. Inherent in this approach is the assumption that an occluded region suitable for the initiation of crevice corrosion exists on the container surface, which is not necessarily the case but which is nevertheless a conservative assumption to make. Not all grades of Ti alloy considered as container materials are susceptible to crevice corrosion under repository conditions. The commercially pure Grades 1 and 2 are susceptible in aerobic warm Cl^- environments, as is the Ni-Mo-containing Grade 12. The addition of Pd (as in Grades 7 and 16) greatly improves the resistance to crevice initiation and these alloys can be considered to be immune under repository conditions. For the susceptible Grade 2 material, a model has been developed on the basis of the rate of crevice propagation (Johnson et al., 1994). For Ni alloys, a large database of empirical E_{RP} (Figure 10) and E_{CORR} values has been used to predict the likelihood of crevice initiation under aerobic conditions for the Yucca Mountain repository (DOE 2008). Conservatively, it was assumed that the rate of propagation was fast and that through-wall penetration would rapidly follow, although there is empirical evidence and mechanistic arguments to suggest that stifling of the crevice could occur prior to container failure (Mon et al. 2005).

Environmentally assisted cracking

Typically, the rate of crack growth is so high relative to the required service life of the container, that it is necessary to demonstrate that the conditions required for EAC do not exist in the repository. If such conditions can exist, then failure is generally assumed to be instantaneous once the threshold criterion for cracking is met. Thus, the SCC of both copper (King 2021a) and carbon steel containers (Johnson and King 2008) are excluded as possible failure modes based on the argument that the pre-requisite conditions for a suitably corrosive environment, a sufficient tensile stress, and a susceptible material do not co-exist in the repository at any time. These arguments can be strengthened by a suitable choice of a less-susceptible grade of material (Crusset et al. 2017).

Carbon steel and Ti alloys are also susceptible to hydrogen-related degradation mechanisms. The hydrogen-induced cracking of carbon steel has been excluded based on the argument that the absorbed hydrogen concentration is less than that required for HIC for the relatively low-strength grades considered as container materials (JNC 2000). However, higher absorbed hydrogen concentrations are possible if an external H_2 pressure develops at the container surface due to the low permeability of the bentonite buffer. Under these circumstances, the possibility of HIC can be assessed by comparison of the stress or stress intensity to the threshold value for sub-critical crack growth in the presence of a given hydrogen pressure (King et al. 2014b).

Titanium alloys are susceptible to hydride formation and hydride-induced cracking (Hua et al. 2005). The point at which fast fracture occurs can be predicted based either on the time to reach a threshold absorbed hydrogen concentration (Hua et al. 2005, Qin and Shoesmith 2008) or a critical hydride-layer thickness (Nakayama et al. 2008). Since the major source of absorbed H is the cathodic reduction of protons inside an actively propagating crevice, one of the most effective methods for excluding the HIC of Ti alloys is to select one of the crevice-corrosion-resistant Pd-containing grades.

The one exception to the general assumption of fast crack propagation is the attempt to predict the rate of SCC crack growth for Alloy 22 waste packages based on a slip dissolution mechanism (Andresen et al. 2005). However, arguing on the basis of limited propagation requires that the crack driving force (for example, either the supply of oxidant or the magnitude of the tensile stress) diminishes with time, as otherwise through-wall crack propagation would seem to be inevitable for the timescales of interest.

Microbiologically influenced corrosion

Corrosion science has not yet developed to the point at which it is possible to predict the extent or morphology of corrosion underneath a biofilm with any degree of confidence. Therefore, except for Ti alloys and the more-highly-resistant Ni alloys for which it may be possible to construct a reasoned argument that these materials are immune to MIC, it is necessary to demonstrate that microbial activity at the container surface will not occur. Both highly compacted bentonite (Taborowski et al. 2019) and cementitious backfill (Aerts 2009) suppress microbial activity, so MIC is only of concern if remotely produced metabolic by-products diffuse to the container surface (King 2009). This process is the basis for many of the sulfide-transport models developed to account for the corrosion of copper containers under anaerobic conditions (Idiart et al. 2019, King et al. 2021, Pekala et al. 2019). In such models, the extent of microbial sulfate production is generally limited by the availability of either the electron acceptor (sulfate) or of the electron donor (organic carbon or H_2) (King et al. 2021). It is possible, therefore, to approximate the extent of corrosion based on a mass-balance argument involving the different reactants (Marsh and Taylor 1988).

Table 3: Summary of methods used to predict various corrosion processes for copper, carbon steel, and titanium and nickel alloys. See text for specific examples of each prediction methodology

	Uniform corrosion	Localised corrosion	EAC	MIC
Copper	Mass-balance or reactive-transport modelling for initially trapped O_2 Reasoned argument or radiolysis modelling for radiation-induced corrosion Mass-transport modelling for corrosion due to sulfide	Pitting assessed using pitting factor, extreme-value analysis of maximum pit depth, or probabilistic pitting model for aerobic saturated conditions Corrosion allowance for surface roughening Reasoned argument for exclusion of pitting in presence of sulfide	Reasoned argument for absence of SCC based on joint mechanical-corrosion assessment of environmental and loading conditions	Reasoned argument based on suppression of microbial activity by highly compacted bentonite Sulfide-transport model for remote SRB activity
Carbon steel/iron	Mass-balance or electrochemical modelling for initially trapped O_2 Reasoned argument for excluding radiation-induced corrosion Empirical corrosion rates of reactive-transport modelling for anaerobic corrosion	Pitting assessed using depth-dependent pitting factor or extreme-value analysis of maximum pit depth Cellular automata model for surface roughening	Reasoned argument for absence of SCC or specification of a resistant alloy Reasoned argument for absence of HIC based on low absorbed H concentration and use of low-strength alloys Joint mechanical-corrosion analysis of failure by HIC	Mass-balance argument based on limitation by either organic carbon or sulfate Reasoned argument based on suppression of microbial activity by highly compacted bentonite or cementitious buffer
Ti alloys	Empirical corrosion rates	Limited propagation argument for Grades 2 and 12 Ti, or use of resistant alloys Grades 7, 16, 29	HIC based on either critical absorbed H concentration or a critical hydride layer thickness	Considered to be immune
Ni alloys	Empirical corrosion rates	Model based on exceedance of threshold potential, followed by rapid propagation Reasoned argument for stifling of propagating crevice	Slip dissolution model for limited crack growth	Enhancement factor for uniform corrosion

8.2 Justification and Robustness of Lifetime Predictions

An important challenge in the development and performance assessment of disposal containers is the justification and demonstration of robustness of the container lifetime predictions. The most important step is the ability to demonstrate a sound mechanistic understanding of the corrosion and mechanical processes involved. Regardless of whether lifetime predictions are deterministic (i.e., based on mechanistically based models, Macdonald 2003) or empirical (e.g., based on the extrapolation of observed corrosion rates), it is essential to be able to demonstrate a mechanistic understanding of the underlying corrosion processes in order to justify predictions over such huge timescales. This can be further substantiated by demonstration at appropriate scales of space and time. A thorough mechanistic understanding should underpin any long-term prediction, whether that prediction is based on a simple extrapolation of an empirically determined corrosion rate or on the basis of a detailed reactive-transport model. Only by having a mechanistic understanding is it possible to justify the exclusion of corrosion processes that are not considered possible under repository conditions.

In situ tests and demonstration experiments in underground research laboratories (URL) simulate, as closely as possible, the actual conditions in a deep geological repository (Diomidis and King 2020). Such experiments may be conducted at full-scale to demonstrate the behaviour of the proposed system or, where the scale of the experiment is not important to the aim of the test, at partial scale in a borehole or by making use of some other aspect unique to the URL environment. In both cases, however, in situ testing takes advantage of one or more thermal, hydraulic, chemical, or microbiological aspects of the underground environment that are otherwise difficult to simulate.

Copper- and iron-based materials have been in use by humankind for approximately 5,000 years. On a much greater timescale, deposits of native, metallic copper exist in many countries around the world. These natural and archaeological analogues exist because the material is either thermodynamically or kinetically stable under the specific environmental conditions and can provide evidence of timescales similar to those typically used in safety assessment (Dillmann et al. 2014, King 2021b, Neff et al. 2010, Yoshikawa et al. 2008). Analogues can be used to confirm the long-term kinetics and the validity of the mechanisms identified in typical experimental durations, and for building confidence in the robustness of predictions. An important challenge in the interpretation of corrosion data from analogues, and in the justification of their validity is the generally high uncertainty in the evolution of environmental conditions during their time underground.

Another way to demonstrate the robustness of container lifetime predictions is by applying alternative models for the prediction of a given process. Commonly, there are multiple methods for predicting the rate or extent of a given corrosion process. For example, the extent of corrosion of the container due to the atmospheric O_2 initially present in the buffer material can be variously estimated based on a mass-balance calculation, empirical corrosion rate data, or detailed reactive-transport modelling. The use of alternative approaches to predict the same, or similar, outcome provides confidence in the long-term prediction.

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