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The corrosion behaviour of candidate container materials for the disposal of high-level waste and spent fuel – a summary of the state of the art and opportunities for synergies in future R&D

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ABSTRACT

This paper presents a state-of-the-art analysis of the expected degradation processes of a variety of candidate container materials for the disposal of high-level waste and/or spent nuclear fuel. The work, focusing on the most recent developments, has been performed under the auspices of the Implementing Geological Disposal Technology Platform in the context of an international conference hosted by the Nuclear Waste Management Organisation of Canada (NWMO). The scope of the analysis includes the expected corrosion and environmentally assisted cracking behaviour of copper, carbon steel and titanium in contact with relevant buffer materials (e.g. bentonite, cement) and in conditions expected in an underground disposal facility (long-term anoxic conditions). Considerations relative to the expected evolution of the environmental conditions (especially in the period following backfilling) are also presented. Beyond summarising the current state of knowledge, areas in which opportunities for international collaboration may be present are also highlighted.

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Introduction

Background and aims


In its 2011 Strategic Research Agenda (SRA) [1], the European platform 'Implementing Geological Disposal Technology Platform' (IGD-TP) identified the opportunity to carry out a joint activity (JA11a) to share information on the science and technology underpinning the development of waste containers for the disposal of high-level waste (HLW) and spent nuclear fuel (SNF), in particular on the expected corrosion behaviour of candidate container materials (in some disposal programmes referred to as 'canisters' or 'over-packs'). This information is relevant to most countries expecting to pursue geological disposal, since it may be applicable to most, if not all, HLW and SNF waste and since it can be generalised (with important caveats and observations) to a range of disposal environments and associated disposal concepts (e.g. choice of buffer materials).

Activity JA11a, which was conceived to rationalise past and recent developments in a relatively mature technical area, was completed through a dedicated meeting between European and non-European experts, held on the occasion of the '6th International Workshop on Long-term Prediction of Corrosion Damage in Nuclear Waste Systems' (Toronto, Canada, May 2016) [2]. The workshop, first organised in 2001 [3] and since then held on a regular basis (about every three years) [4–7], is considered by the technical community an effective means of discussing and exchanging information on relevant R&D at an international level (typically with input from European countries as well as from Canada, the U.S.A., Japan and China). As a result, the approach pursued in JA11a was not to create an alternative forum to facilitate the exchange of information but to capitalise on the outputs of a well-established and successful event. The analysis presented in this note draws on the proceedings of the latest workshops (particularly the last one [2]), on review studies

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produced as part of the UK programme (appendices of reference [8]), and on the input of key experts.

The aim of this activity (and the meeting used to help delivering its outcomes) was to discuss key outstanding and/or debated issues for specific disposal systems (and issues broadly applicable to all of them), in order to develop a consensus view about the state-of-the-art and the need for, and direction of, future work. Building on detailed considerations for different disposal systems, this note presents a summary of the current state-of-the-art and highlights areas of research common to different systems in which opportunities to tackle common challenges in a collaborative manner may be present or arise in the future (i.e. it can be used as a basis for identifying future synergies). The intention is to highlight shared views and interests in disposal programmes across (and beyond) European countries, thus helping member states (and the radioactive waste disposal community as a whole) embed existing knowledge in an effective manner and identify and consolidate opportunities for collaborative effort.

The note is intended to present a summary of key points, based on the most recent developments and the general consensus of the technical community. Detailed considerations relative to specific systems are provided in the Appendixes.

Scope of the document

The discussion below presents a high level analysis of the state-of-the-art of the expected corrosion mechanisms, our level of understanding relative to them, and their relevance to disposal of HLW and SNF in a deep geological repository (i.e. anoxic conditions in the long term). Disposal concepts considering disposal in a permanently oxic environment (of the type proposed for Yucca Mountain in the U.S.A.), any permanent or interim storage preceding disposal, and wastes other than HLW and SNF are outside the scope of this document.

The document focuses on disposal concepts envisaging the use of either a copper, a carbon steel, or a titanium corrosion barrier, reflecting the type of disposal systems typically considered in many countries. While part of the initial ambition of this activity, the use of nickel alloys (and stainless steel) for the disposal of HLW and SNF was not discussed in the meeting (and not reported herein), due to limited time availability and the need to focus the discussion on the most relevant topics. A limited amount of recent information is expected to be available on these materials in the context of HLW/SNF disposal under anoxic conditions, so this is not expected to be a major omission.

The discussion considers the case of disposal in contact with either a clay or a cement-based engineered barrier system (EBS), reflecting the nature of the buffer materials typically envisaged in rocks in which water transport is expected (e.g. granites or clays). Disposal in dry rocks (e.g. evaporites) is not covered in this note, since corrosion processes are expected to be minimal. Disposal with a relatively thin cement/bentonite liner (relevant to the French programme) is also discussed in the section on carbon steel in clay systems. The discussion does not consider in detail whether emplacement in contact with the buffer is carried out *in situ* or upstream in a pre-fabricated emplacement module (PEM, sometimes referred to as 'supercontainer'), an issue which is largely associated with the quality assurance

(QA) of the disposal system rather than its behaviour in the expected conditions.

Processes other than environmental degradation (e.g. mechanical effects) are not considered in detail in this document. However, it is important to emphasise that the mechanical evolution of the system, particularly the likely evolution of tensile stresses (residual and applied) on the surfaces of waste containers, is an important parameter determining the likelihood and impact of some of the environmentally assisted cracking (EAC) processes, which are discussed in some detail in this document. Studies containing a detailed mechanical analysis of the stresses likely to be present in waste containers have been completed in several disposal programmes indicating that, while generally resulting in compressive fields, relatively high magnitude tensile stresses are likely to develop in some locations, and that such locations may vary during the overall mechanical evolution of the disposal system [8,9].

For the environmental degradation effects the discussion focuses on relevant forms of corrosion, for example microbologically influenced corrosion (MIC), and other forms of environmental damage – stress corrosion cracking (SCC) and hydrogen induced cracking (HIC) – that may occur outside waste containers. The discussion reports observations that have emerged from lab-based studies, *in situ* experiments, models and, where appropriate and to a relatively small extent, natural and man-made analogues. Internal corrosion, which is possible in the case of wet wastes (particularly spent fuels that may contain residual water after retrieval and drying from pond storage), have been studied in specific programmes (e.g. [10]) and is expected to lead to very modest amounts of damage. The effect of external corrosion processes on gas generation (a key consideration in disposal concepts in low permeability host rocks) and on the mineralogy of buffer materials are also not covered in this document. In general, the results of *in situ* and demonstration experiments, often focusing on the behaviour of the EBS as a whole, are particularly valuable from this point of view and are discussed, with focus on the corrosion of the waste containers, in relevant parts of the text.

Structure of the document

The main body of the note presents a high level summary of key considerations. Detailed considerations on specific disposal systems are presented in the Appendixes, generally focusing on their expected long-term behaviour.

The text covers in turn systems envisaging the use of copper in a clay buffer, carbon steel in a clay buffer/host rock, carbon steel in a hyperalkaline cement/concrete buffer, and titanium in contact with an undefined buffer/filler material. The type of system currently envisaged in France (making use of a carbon steel waste container emplaced in a steel liner surrounded by a mildly alkaline cement/clay filler disposed in a clay host rock), which presents some peculiarities relative to other concepts, is discussed in the context of clay-based systems but merits its own specific considerations.

Focusing on the transient conditions expected to develop in the initial period after disposal, on which limited emphasis has been placed so far, general considerations on the environmental conditions in contact with waste containers (relevant to all disposal concepts) are also presented. Beyond its

potential impact on the radiological risk of a disposal facility in the long-term (due to the relatively high corrosivity of the environment associated with the presence of oxygen in the system), the corrosion behaviour of waste containers during the transient period is important in disposal programmes envisaging long operational periods of a disposal facility, and particularly the need for ensuring the reversibility of emplacement operations and/or the retrievability of the waste containers, as well as those considering thin corrosion barriers (e.g. copper coatings).

Summary of the state-of-the-art

The corrosion behaviour of the types of containment systems (waste container and, if any, buffer materials) typically envisaged in geological disposal is generally well understood, although, after many years of active research, areas of some uncertainty (typically covered by existing R&D programmes) still exist. The understanding of degradation mechanisms and resulting durability estimates is particularly mature for disposal concepts envisaging the use of copper and carbon steels in conjunction with clay buffers (particularly bentonite) but also for concepts making use of concrete buffers/liners. A brief summary of the state-of-the-art for specific systems is presented below. A recent analysis is also available in Ref. [11]. Building on this, areas in which further R&D may be beneficial, particularly across a number of systems, are discussed in the next section.

In general, the impact of corrosion processes is either managed by designing structures of sufficient thickness to tolerate expected damage (typically the case of general corrosion in the so called 'corrosion allowance' designs) or excluded by the selection of container and buffer materials (typically the case for localised corrosion, MIC and EAC). In the case of thick-walled containers, wall thicknesses are sufficiently high to provide additional confidence in the robustness of the design to processes potentially leading to faster penetration rates (e.g. pitting corrosion, MIC), were they to occur. Additional confidence in the design robustness to EAC, conversely, typically relies on the choice of materials with limited susceptibility and/or suitable design, manufacture and stress-relief of key components (e.g. welds).

Copper-clay systems

The durability of copper in many disposal conditions (particularly near-neutral pH, chloride-rich porewaters containing up to moderate amounts of sulphide) is expected to be determined by well-understood degradation processes and typically result in a very high durability (dictated by the thickness of the corrosion barrier, the expected concentration of sulphide outside disposal containers, and the rate of transport of sulphide to the container). While it is possible that a copper-clay system may provide a very high durability also in conditions of higher sulphide and lower chloride than those currently expected in the Swedish, Finnish and Canadian programmes, it is important to understand that challenges to the current understanding (i.e. likely to require additional R&D) or to the likely applicability based on current understanding (i.e. unlikely to require additional R&D) may arise in the case of porewater/groundwater compositions poor in chloride, and particularly rich in sulphide and/or carbonate (see for example [12]). Hydrogen generation processes reported in pure water remain an uncertainty, although recent work

is indicating that gas generation is either likely to be associated with processes other than corrosion (degassing of tested material or equipment) or, if associated with corrosion processes, likely to occur at a rate less than that expected for sulphide-induced corrosion, thus bearing limited consequence on the durability of waste containers. Effects that are particularly important in the case of thin-walled (e.g. coated) containers, specifically the effects of radiation on general corrosion, localised forms of corrosion and possibly hydrogen absorption, may also require further R&D and are being investigated. Further analysis of natural and archaeological analogues focusing on the nature of degradation processes and corrosion products in environmental conditions similar to a repository (e.g. native copper in a clay host structure) may provide additional confidence in the understanding gained so far.

Carbon steel-clay systems

The corrosion behaviour of carbon steel in a clay environment is well understood and, within a relatively wide envelope of conditions, expected to result in modest long-term general corrosion rates, leading to long lifetimes. Care, however, needs to be taken in designing the EBS to ensure that carbon steel containers are not exposed to transient periods in which acidic conditions may develop, which may not only lead to higher rates of general corrosion during the transient period itself but also in the longer term. The presence of modest concentrations of carbonate in the environment may lead to a risk of SCC (or probably, more appropriately, HIC). This requires bespoke consideration, including selection of suitable alloys (low strength, suitable microstructure) and manufacturing (e.g. welding) techniques. The presence of radiation, depending on the level of shielding provided by the waste containers, is expected to have a modest or minimal effect on corrosion rates and, if required, can be conservatively taken into account in the container design and/or durability estimate.

Carbon steel-cement/concrete systems

The corrosion behaviour of carbon steel in a cement/concrete environment is well understood and, within systems containing limited amounts of chloride, expected to result in a very long durability, associated with passivation of the metal surface. These considerations may extend to systems containing relatively high amounts of chlorides, depending on differences between the rate of groundwater resaturation and oxygen consumption in the near field. Sulphur-containing species (e.g. thiosulphate, sulphide), known to be corrosive to a number of metals, are not expected to be able to offset the steel passivity at the high pH considered in this type of system. Even at relatively high dose rates, radiation does not seem to affect the behaviour of the system. Studies of the long-term behaviour of the passive film in relevant conditions indicate that the film is stable in reducing conditions but that the likelihood of pit initiation, while low, may not be negligible. While evidence from studies in a variety of systems indicates that, in anoxic conditions, deep propagation of localised corrosion is unlikely, this would benefit from experimental and theoretical confirmation to reinforce the current state of understanding. Studies aimed at evaluating the characteristics of the film, underpinning estimates of the likelihood of corrosion initiation (pitting and SCC), are continuing.

Titanium systems

The conditions leading to the initiation of corrosion on titanium in near-neutral solutions simulating porewater that may be expected in contact with a clay buffer or other filling material are relatively well understood. However, specific aspects of its propagation in conditions in which corrosion is viable are not. Current understanding indicates that if crevice corrosion was not able to initiate and/or propagate deep into the metal during the oxic period, a very high durability may be expected. In particular, current knowledge indicates that, even if initiation during the oxic phase could not be excluded, corrosion propagation may be inherently limited by mechanistic effects, resulting in limited amount of physical damage and, with that, limited amount of embrittlement due to hydrogen absorption.

Disregarding the crevice corrosion behaviour, however, the reduction in mechanical properties associated with the long-term absorption of hydrogen during general corrosion would need to be taken into account when designing disposal containers. Whether, if true, such arguments could be proved convincingly may require further R&D. In particular, it would be desirable to gain a better understanding of the rate of hydrogen pick-up, the critical levels of hydrogen required to alter the mechanical properties of the metal and of the mechanical properties of 'embrittled' (i.e. hydrogen-containing) titanium, as well as additional confidence in the long-term stability of the passive film.

If recent observations on the (rapid) rate of oxygen depletion in a disposal facility (see below) were applicable to a range of disposal concepts and geological environments, confidence in the likely effectiveness of titanium as a long-term corrosion barrier would substantially increase.

Environmental conditions

The nature of environmental conditions expected to develop in the disposal system after the initial transient period have been studied for many years and are generally well understood. However, given its limited duration, the conditions expected to develop in the near field during the transient period immediately following backfilling and sealing of deposition tunnels have not been considered in great detail and may require further investigation. During this period, depending on the system in question, fast degradation mechanisms linked for instance to the localisation of anodic and cathodic areas or to the development of SCC, may need to be avoided.

In particular, the analysis of *in situ* experiments is gradually indicating that, due to microbial activity, corrosion, and/or mineral (e.g. pyrite) oxidation, oxygen consumption in the near-field may occur more quickly than previously estimated. This is particularly important in concepts for which fast degradation mechanisms, particularly SCC, may only be possible in oxic conditions (notwithstanding the fact that, in anoxic conditions other potentially 'fast' degradation mechanisms, particularly HIC, may also be possible).

Conversely, a number of *in situ* experiments indicate that swelling of the bentonite in contact with incoming groundwater is a relatively slow and, in some cases, heterogeneous process. Depending on whether the main factor controlling the risk of MIC is the bentonite swelling pressure or the water activity, this may have important consequences for the likely behaviour of the system. If the risk of MIC is controlled by water activity, then such risk is likely to be low even before

the bentonite is saturated, given that the limited water activity before resaturation will control microbial activity (after resaturation, water activity will remain low due to the swelling processes). Conversely, if the risk of MIC in bentonite is directly controlled by its swelling pressure, a long resaturation process increases the risk of MIC during the transient period, associated with incompletely or unevenly swollen areas of the buffer. In disposal studies, instances of microbial activity, and even MIC, have been found in components deliberately or accidentally exposed to below-specification swelling pressures, but the overall risk of MIC during the transient period in an otherwise well specified buffer and the mechanisms controlling it remain an important uncertainty.

A greater understanding of the likely changes in mineralogical and transport properties associated with heating and resaturation of cement buffers (studies already ongoing in the context of clay-based buffers) may also be desirable to complement existing studies of the likely pH evolution (notwithstanding the observation that current cement systems envisage the use of a 'supercontainer' in which the conditions of the concrete in contact with the waste container are well defined and likely to remain so for long periods of time).

Opportunities for synergies in future R&D programmes

Based on the analysis presented above, this section highlights opportunities for collaborative working and/or mutual learning in addressing technical challenges that may be common or similar across a number of disposal concepts. The analysis of future R&D that may benefit specific disposal concepts is beyond the remit of this note but, to an extent, maybe inferred from the description of the state-of-the-art presented above. Considerations applicable to most disposal concepts are:

- Current understanding indicates that, at the dose rates typically expected from HLW and spent fuel at the time of disposal, designing disposal concepts able to take into account radiation effects (through a suitable wall thickness and/or by accounting for enhanced corrosion rates for a limited duration of time) is not expected to be particularly challenging. Radiation effects, however, may become more important during the optimisation of current designs, typically leading to a reduction in thickness of structural and corrosion-barrier components, and/or for programmes envisaging the disposal of waste which will not have been subjected to long decay times. In many circumstances, expected dose rates may be bounded by tests already carried out or continuing. However, there may be opportunities to develop a greater mechanistic understanding of the effect of radiation on the behaviour of metallic interfaces and on the likelihood of radiation to induce, beyond changes in corrosion behaviour, degradation of mechanical properties, particularly HIC (mechanical degradation processes experienced in nuclear reactors, in particular neutron embrittlement, are not expected in disposal containers). Table 1 reports values of the external dose rates of waste containers from currently available information, which can be used as a basis to plan and contextualise further studies in this area.

Table 1. Dose rates on the external surfaces of HLW/SNF containers estimated in different national programmes for a variety of container designs and cooling periods assumed in the calculations.

Country	Container design and typical/minimal wall thickness	Cooling time before disposal assumed in calculation	Approximate external dose rate
Belgium	Carbon steel container (30 mm)	50 years (60 years currently assumed in the programme)	25 Gy h ⁻¹
Canada	Carbon steel member (30–47 mm) with copper coating (3 mm)	10 years (30 years currently assumed in the programme)	<3 Gy h ⁻¹
France	Carbon steel container (65 mm)	60 years	10 Gy h ⁻¹ *
Japan	Carbon steel container (190 mm including 40 mm corrosion allowance. Dose rate estimation is for 150 mm thickness.)	50 years	0.03 Gy h ⁻¹
Sweden	Copper shell (50 mm) with massive cast iron insert	30 years	0.2 Gy h ⁻¹
Switzerland	Carbon steel container (120 mm)	30 years	0.1–0.2 Gy h ⁻¹
UK (Variant 1)	Copper shell (50 mm) with massive cast iron insert	25 years	0.03 Gy h ⁻¹
UK (Variant 2)	Carbon steel container (min 70 mm)	25 years	0.3 Gy h ⁻¹

The dose rates are indicative, since they will typically display a modest dependence on location, including local wall thickness. Note that other assumptions (particularly waste type) are also important to define dose rates but are expected to be relatively similar across different countries.

*Note that the value reported for the French programmes is a conservative 'working value', not based on actual calculation.

- Understanding the behaviour of the EBS during the transient period requires a greater understanding of the relative rates of oxygen consumption and resaturation (particularly for passive or passivated materials), as well as greater understanding of the result of heterogeneous wetting and evolution of the chemistry at the interface between the container and the buffer. Further studies of the inhibition of MIC and its controlling mechanism during this period seem also important, with a number of *in situ* experiments continuing.
- Slightly peripheral to this note, but becoming of increasing importance in many disposal programmes, there may be opportunities to carry out studies of the design, manufacture and subsequent testing of realistic waste container components (including alloy selection, fabrication and welding processes, subsequent treatment and, in the case of copper-coated designs, coating deposition) in a collaborative manner. Beyond increasing confidence in the feasibility (already achieved in many disposal programmes), these studies would increase confidence in the ability to mitigate, with suitable engineering approaches, potentially detrimental processes (particularly SCC/HIC on carbon steels and creep/HIC on copper). *In situ* tests would be particularly valuable to gain confidence in the behaviour of the system beyond lab-based studies.
- There may be further opportunities to develop and refine container failure time distributions for probabilistic safety assessments as well as providing further confidence in the durability of the disposal system by additional studies on natural and man-made analogues (particularly for copper systems).

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