

Status of the Understanding of Used Fuel Container Corrosion Processes – Summary of Current Knowledge and Gap Analysis

NWMO TR-2007-09

October 2007

Fraser King

Integrity Corrosion Consulting Limited

nwmo

NUCLEAR WASTE MANAGEMENT ORGANIZATION SOCIÉTÉ DE GESTION DES DÉCHETS NUCLÉAIRES

Nuclear Waste Management Organization
22 St. Clair Avenue East, 6th Floor
Toronto, Ontario
M4T 2S3
Canada

Tel: 416-934-9814
Web: www.nwmo.ca

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ABSTRACT

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Report No.: NWMO TR-2007-09
Author(s): Fraser King
Company: Integrity Corrosion Consulting Limited
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Abstract

This report is a summary of the status of the knowledge of the corrosion behaviour of copper and carbon-steel used-fuel containers, and the gaps in this knowledge.

A brief review of the current level of understanding of used-fuel container corrosion is given. This review draws on information developed as part of the Canadian research and development program for long-term management of Canada's used nuclear fuel and from similar international programs that are also considering the use of copper or carbon-steel containers in their deep geological repository concepts.

Within the Canadian research program, the understanding of the corrosion behaviour of copper is relatively mature, having been the subject of detailed studies over the past 20 years. However, there are still some areas of uncertainty or areas that require additional effort, including corrosion under unsaturated conditions, the potential for microbiologically influenced corrosion, the evolution of corrosion damage from the initial warm, aerobic phase to the long-term cool, anaerobic period, and stress corrosion cracking.

The carbon-steel corrosion research program is much less mature within the Canadian context, although there is considerable experience within the Japanese and various European programs. Much of the existing information is transferable to conditions expected within a Canadian deep geological repository, but there are areas requiring further study, including: corrosion under unsaturated conditions, the evolution of corrosion damage, microbiologically influenced corrosion, stress corrosion cracking, and hydrogen gas generation.

Gaps in the existing knowledge have been identified in these and other research areas. These gaps have been grouped into proposed research areas or programs for future consideration.

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1. INTRODUCTION

With the recent selection by the Government of Canada of the Nuclear Waste Management Organization's (NWMO) recommended approach Adaptive Phased Management (NWMO 2005), the current understanding of the corrosion of used fuel containers (UFC) has been reviewed. This report summarizes both the existing knowledge of the corrosion of copper and carbon steel (C-steel) UFC and the gaps in that understanding. Much of the information contained in this report was drawn from an NWMO-sponsored UFC corrosion workshop held in Toronto from June 12-13, 2007. The NWMO's UFC corrosion workshop brought together Canadian corrosion researchers and experts to discuss the state-of-knowledge on copper and steel corrosion in a deep geological repository and to propose work program activities to address corrosion issues.

The review of the state-of-the-art understanding of UFC corrosion is presented in the context of the Canadian research program for Adaptive Phased Management which includes research into the development of a deep geological repository (DGR) for Canada's used nuclear fuel. Adaptive Phased Management is first briefly described, along with more-detailed descriptions of the UFC design requirements and the expected environmental conditions to which the containers will be exposed in the repository. The review of the existing corrosion knowledge is based on information developed within the Canadian program and from various international programs with similar deep geological repository concepts.

The section of the report describing the gaps in the existing knowledge is largely based on the outcomes of the UFC corrosion workshop. Various areas requiring further study are identified and suggestions have been made with regard to their priority. Proposed individual research areas have been grouped together by topics and further classified into programs common to both candidate UFC materials or specific to either copper or C-steel.

2. BACKGROUND

2.1 Adaptive Phased Management

The Nuclear Waste Management Organization (NWMO) is developing the technology for the long-term management of Canada's used nuclear fuel. The NWMO's recommended approach, Adaptive Phased Management, was accepted by the Government of Canada on June 14, 2007. Adaptive Phased Management includes development of a DGR in a suitable rock formation such as the crystalline rock of the Canadian Shield or Ordovician sedimentary rock (NWMO 2005).

Figure 1 shows a schematic of several different conceptual designs for a DGR in Canada (Russell and Simmons 2003).

Figure 1: Conceptual Designs for a Deep Geological Repository in Canada (Russell and Simmons 2003).

Copper has been selected as the reference corrosion-barrier material for the UFC in a potential deep geological repository (DGR) in a crystalline rock host formation (Maak 1999). Various container designs have been considered (Maak and Simmons 2001), but a possible configuration would involve the isolation of 324 used fuel bundles in a container comprising a carbon steel (C-steel) support vessel and an outer corrosion barrier of oxygen-free, phosphorus-doped (OFP) copper (Figure 2). OFP copper was developed in the Swedish program and is, essentially, oxygen-free copper (equivalent to UNS C10100) with the additional requirements of <5 ppm O, 50-70 ppm P, <0.6 ppm H, and <8 ppm S (Andersson et al. 2004). Phosphorus is added to improve the creep ductility of the material, and the H and S contents are minimized for the same reason. The oxygen specification was based on the need to avoid weld porosity using electron-beam welding, although friction stir welding is now the reference method for the final closure weld in Sweden.

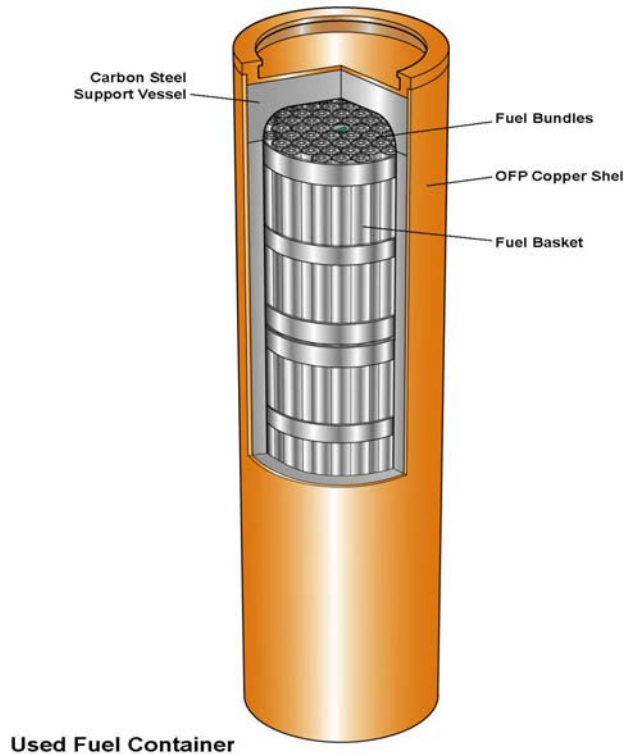


Figure 2: Conceptual Used Fuel Container Design for 324 Used Fuel Bundles Comprising a Carbon Steel Support Vessel and Outer OFP Copper Corrosion Barrier (after Maak and Simmons 2001)

Carbon steel has also been proposed as a candidate container material for a DGR in Ordovician sedimentary deposits (King 2007). Although the lifetime of C-steel containers may be shorter than the lifetime of copper containers, C-steel containers are being studied for possible use in a DGR in sedimentary rocks.

While a detailed design for a C-steel container has not been developed in Canada, there are certain generic requirements that would need to be considered. For example, in order to withstand the external pressure loads in a DGR in sedimentary rock, it is expected that a steel container would need a wall thickness of the order of 100 mm, which is similar to the wall thickness of the inner steel vessel of the reference copper used-fuel container (Maak 2006, Saiedfar and Maak 2000, Maak and Simmons 2001). The detailed container thickness would be determined based on structural analyses that would likely follow the intent of the ASME Boiler and Pressure Vessel Code with a safety factor of 3 for the pre-glaciation period (10,000 years is considered to be within the pre-glaciation period). This means that a steel container with a remaining thickness of about 66 mm (100 mm minus the 34 mm maximum corrosion penetration) should still have adequate mechanical strength with a substantial safety margin to withstand the external pressure for a period of at least 10,000 years. No container failure or full-wall penetration is expected and a steel container should still be capable of providing containment of the used fuel for a period of more than 10,000 years after it is placed in a repository.

2.2 UFC Design Requirements

Various preliminary design requirements for the UFC have been established (Maak 2006). Among the most important functional and performance requirements are (Maak 2006):

Containment and Isolation - The container shall contain the used fuel and isolate it from the surrounding environment from the time of loading through handling, transportation, emplacement and potential retrieval operations, and for its specified design life in the deep geologic repository facility.

Shielding – The container shall provide adequate shielding to ensure that the radiation fields at its surface do not adversely affect other engineered barriers or generate corrosion agents via radiolysis that may adversely affect the integrity of the container.

Design life - For general DGR design purposes, the used fuel container shall be designed for a functional life of not less than 100,000 years. In meeting this requirement the impact of glaciation shall be considered in the design of the container and the repository.

Container designs with a shorter functional life (e.g., between 1,000 to 100,000 years) may also be considered, if the choice of a shorter functional life is supported by a comprehensive site-specific and design-specific environmental and safety assessment and all operational requirements for handling, emplacement and retrieval can be satisfied. This would improve the flexibility of container design and material choices.

Integrity – The container design and its manufacturing, sealing and inspection processes shall be optimised to minimise the probability that a container would have an undetected manufacturing defect that could lead to loss of containment during the container design life.

Mechanical stability and strength - The container shall be designed with sufficient mechanical stability and strength to withstand the mechanical loads that could arise in a DGR and maintain mechanical integrity to provide containment of the used fuel during the design life of the container.

Corrosion resistance - The container shall be designed with sufficient resistance to both general and localized corrosion processes to prevent the possibility of a through-wall penetration throughout its design life.

Breach of a container - In the event of a breach of the containment boundary (i.e., corrosion-barrier vessel) at any time after emplacement, the container shall not exhibit rapid deterioration due to either general or localized corrosion at the breached location.

Adverse effect on other engineered barriers - The container material and its corrosion products shall have no significant adverse effects on the performance of the other engineered barriers.

Internal chemical environment - The container internal structure shall be designed such that it would establish a reducing chemical environment in the case of water ingress into the container.

Total acceptable strain in the container - The total elastic and plastic strain developed in any part of the container shall not exceed the creep-rupture strain of the material over the design life of the container with a specified safety margin (to be determined).

Container surface temperature - During the design life of a used-fuel container, the surface temperature of the container would be designed to ensure that:

1. the integrity of used fuel within the container is not detrimentally affected by temperature;
2. the design of the repository emplacement configuration is feasible and practicable; and
3. the temperature limits on other components of the engineered barriers can be satisfied, including the maximum allowable temperature of 100°C for the bentonite-based sealing materials, which is to avoid undesirable phase transformation and an adverse effect on the swelling and self-sealing properties of a bentonite-based buffer.

Container geometry and capacity - The container design shall be optimised with the intent of maximising the used fuel capacity.

Compatibility of container materials - The container materials shall be chemically compatible with each other and the used fuel to minimise the risk of corrosion.

Retrieval of used fuel - The container shall be designed to facilitate possible retrieval as long as such design features do not compromise the safety of the deep geologic repository facility.

2.3 Environmental Conditions

Evolution of Environmental Conditions

Certain aspects of the chemical and physical environment to which the UFC are exposed change over time as the conditions within the deep geological repository evolve. In general, this evolution of conditions can be described in terms of the changes in (i) temperature, (ii) redox conditions, and (iii) the degree of saturation (King 2005a). In terms of the evolution of temperature and redox conditions, the repository environment is classically divided into an initial warm, oxidizing period and a subsequent, long-term cool, anoxic phase (Figure 3). In terms of the degree of saturation of the repository, the evolution can be alternatively described by an initial dry-out period, followed by a transition phase during which the bentonite-based sealing materials take up moisture, and a long-term saturated phase (Figure 4). In either case, from a corrosion standpoint, the environmental conditions generally evolve from an early period in which localized corrosion processes are possible to a stable long-term period during which corrosion processes are typically slower and more predictable (i.e., uniform corrosion).

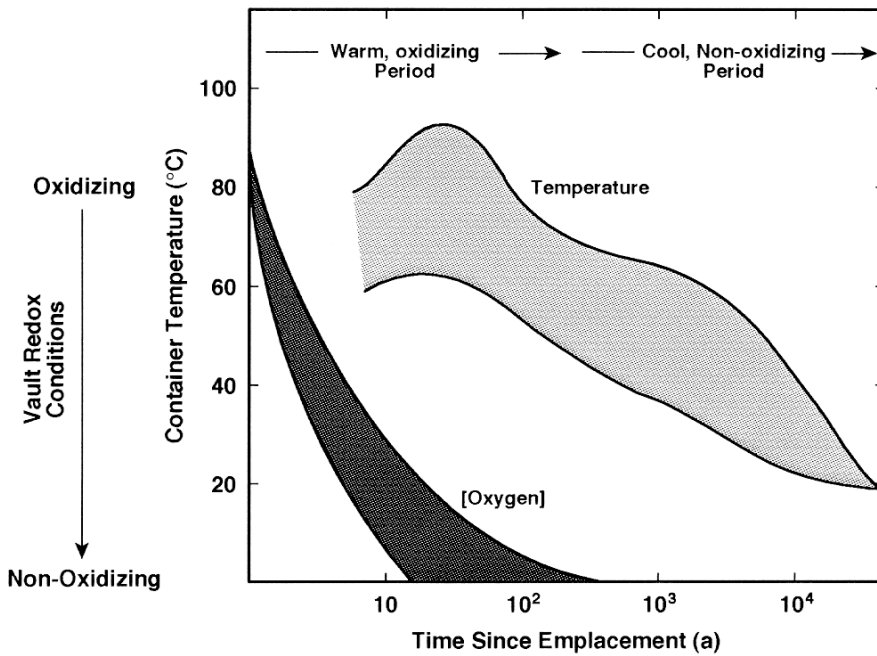


Figure 3: Schematic Representation of the Evolution of Redox Conditions in a Canadian Deep Geologic Repository (King and Kolar 2000).

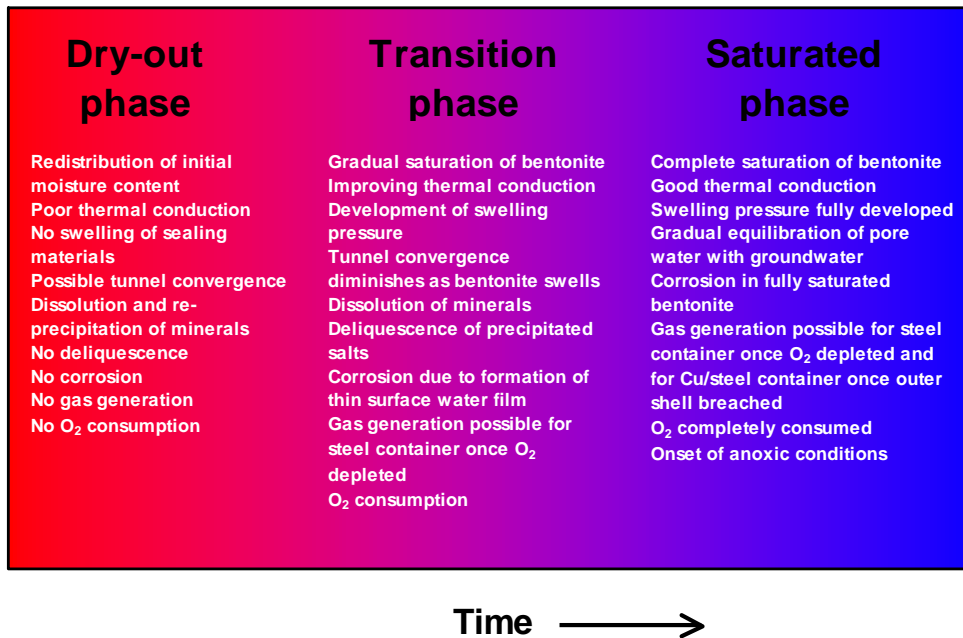


Figure 4: Evolution of Conditions in a Deep Geologic Repository Characterized in Terms of the Stages of Saturation (King 2005a).

The following environmental parameters will affect the corrosion behaviour of the UFC:

- availability of oxidants,
- composition of the groundwater and bentonite pore water,
- temperature,
- the degree of saturation,
- mass transport of species to and from the container surface,
- external loads and residual stresses, and
- the activity of microbes.

Oxidants

The presence of compacted sealing materials in the repository limits the amount of atmospheric O₂ trapped initially in the repository. Depending upon the repository and container design, the initial O₂ inventory is of the order of 1-10 mol_{O₂}/m², which is equivalent to ~10's-100's μm of uniform corrosion (depending upon the container material) if evenly distributed over the container surface. Apart from corrosion of the container, the initially trapped O₂ can be consumed by the oxidation of Fe(II) and other oxidisable minerals in the clay or host rock and by aerobic microbial activity.

As noted above, a design requirement for the UFC is that it be sufficiently self-shielding to preclude the production of radiolysis products that would adversely affect the integrity of the container.

In the absence of sulphide ions, copper is thermodynamically stable in water. Water is, of course, an oxidant for C-steel.

Groundwater

Tables 1 and 2 summarize the expected ranges of the composition of the groundwater at possible repository depths in the Canadian Shield and Ordovician sedimentary rock formations (Maak 2006). In both cases, the salinity of the groundwater increases with depth and, although there are relatively few data for the Ordovician sedimentary deposits, the water is essentially O₂ free at repository depth. For neither host rock is there any evidence for reduced sulphur species in the groundwater or present as mineral phases in the rock.

Table 1: Representative Range of Initial Groundwater Chemistry at Repository Depths between 500 and 1000 m in the Canadian Shield.

Components	pH	Eh (mV)	TDS* (g/L)	Na (g/L)	K (g/L)	Ca (g/L)	Mg (g/L)	Cl (g/L)	SO ₄ (g/L)
Minimum	6 ⁽¹⁾	-225 ⁽²⁾	0.1 ⁽¹⁾	0.001 ⁽¹⁾	0.003 ⁽¹⁾	0.1 ⁽¹⁾	0.002 ⁽¹⁾	0.01 ⁽¹⁾	0.001 ⁽¹⁾
Maximum	9 ^(1,2)	0**	100 ⁽¹⁾	9 ⁽¹⁾	0.2 ⁽¹⁾	28 ⁽¹⁾	0.3 ⁽¹⁾	60 ⁽¹⁾	3.0 ⁽¹⁾

* TDS (Total Dissolved Solids)

** Maximum Eh assumed suitable for repository (to be confirmed)

(1) Frape and Fritz (1987)

(2) Gascoyne (2000)

Table 2: Representative Range of Initial Groundwater Chemistry at Repository Depths between 500 and 600 m in Ordovician Sedimentary Rock.

Components	pH*	Eh* (mV)	TDS* (g/L)	Na** (g/L)	Ca** (g/L)	Cl* (g/L)	SO ₄ ** (g/L)
Shale	Slightly acidic	<0	150-300	25	20	25-150	0.5
Limestone	6.2-6.3	< 0	40-300	25	20	25-200	0.5

* Golder (2003)

** King (2005a)

Bentonite Pore Water

The UFC is not exposed to groundwater, but to conditioned bentonite pore water. Although the pore water will eventually equilibrate with the groundwater, this process could take hundreds to thousands of years. Immediately following placement of the containers and sealing of the repository, the near-field pore-water chemistry is affected by a number of processes (King 2005a, 2006):

1. dissolution of soluble mineral impurities (NaCl, CaCl₂·xH₂O, CaCO₃, CaSO₄·2H₂O) in the water added for compaction of the sealing materials,
2. ion exchange with the sodium bentonite, resulting in the uptake of Ca²⁺ and the release of Na⁺ ions,
3. precipitation of the least soluble minerals as water is driven away from the container surface by the thermal gradient,
4. re-dissolution of precipitated minerals as the sealing materials re-wet, starting with the most soluble (most deliquescent) phases,
5. re-distribution of salts under the thermal gradient,
6. exchange of Ca²⁺ for Na⁺ will continue until the buffer is altered from Na-bentonite to Ca-bentonite,
7. control of the pore-water pH in the range pH ~7.5-8 by calcite precipitation/dissolution,
8. diffusion of dissolved Cu and/or Fe corrosion products in the pore solution, adsorption on the bentonite (displacing Ca²⁺/Na⁺), and possible precipitation,
9. possible cementation of the bentonite by Fe corrosion products,
10. consumption of the initial O₂ inventory by corrosion, oxidation of Fe(II), and aerobic microbial activity,
11. formation of dissolved and gaseous H₂ (in the case of a steel container), and
12. eventual saturation of the sealing materials.

Thus, the early warm, oxidizing, unsaturated period in the evolution of the environment is characterized by a complex series of near-field processes.

Temperature

The maximum UFC surface temperature is determined by a number of design factors, including: the age of the used fuel, container fuel capacity and spacing in an underground repository, the thermal properties of the sealing materials, and the rate of saturation of the repository. Based on a constraint imposed by the need to avoid thermal alteration of the bentonite, a maximum container surface temperature of 100°C has been specified (Maak 2006). The maximum temperature is typically attained approximately 30 years after placing the containers, with the temperature falling to near-ambient after 10-20,000 years (Figure 3).

Bentonite Barriers and Repository Saturation

As illustrated in Figure 1, the UFCs are surrounded by compacted bentonite-based sealing materials. In addition to limiting the amount of initially trapped O₂ and conditioning the pore-water chemistry, these materials will also restrict the mass transport of species to and from the container surface. In fully saturated compacted bentonite, the effective diffusion coefficient of dissolved species is of the order of 10⁻⁷ cm²·s⁻¹, approximately two orders of magnitude lower than in bulk solution (King et al. 1996). In unsaturated material, the effective diffusivity of dissolved species decreases with decreasing moisture content, but the effective diffusivity of gaseous species increases (King 2006). For example, in 80% saturated bentonite (the approximate initial degree of saturation of some of the sealing materials), the effective diffusivity of O₂ is of the order of 10⁻³ cm²·s⁻¹ or four orders of magnitude higher than in saturated bentonite (King et al. 1996).

Many of the near-field processes noted above are driven by the time-dependent degree of saturation. The bentonite-based sealing materials are compacted and placed in an unsaturated condition, typically 33-80% saturated (depending upon the material), although as low as 6% saturation in the case of dry granular bentonite pellets (Russell and Simmons 2003). As shown in Figure 4, there is a period of further drying of the materials closest to the container due to the effect of the thermal gradient. The initial dry-out phase will be followed by period of transient conditions as the bentonite re-saturates, until full saturation is eventually attained. The time required to fully saturate the repository is currently uncertain and will largely depend upon the repository design and local site conditions. As a guide, however, full saturation may occur some 10's to 100's of years following repository closure.

External Loads and Residual Stress

The containers will be subject to both external loads and the impact of residual stress. Table 3 summarizes the external loads for DGR's in both crystalline rock and sedimentary deposits, with and without the effect of glacial loading (Maak 2006). Additional loading may result from the expansion of corrosion products (in the case of steel containers) and residual stress from the final closure weld. In the case of external loading, compressive stresses will be present on most of the external surface of the container. However, relatively low tensile stresses (i.e., about 5% of the yield strength of C-steel) will be present in a small area on the wall of the container near the lid region (Saiedfar and Maak 2000). Residual stresses can be minimized by suitable weld procedures or by post-weld stress relief by shot peening (Eadie et al. 2002) or by laser-peening or low-plasticity burnishing (BSC 2004).

Table 3: External Pressure Loads in Deep Geological Repository Designs in Crystalline Rock and Sedimentary Rock (glacial load estimated based on the conservative assumption of a full, continuous column of unfrozen water exerting hydraulic pressure on the repository).

	Ordovician Sedimentary Limestone	Ordovician Sedimentary Shale	Crystalline Rock
Assumed average saturated rock density above shale (kg/m ³)	Not applicable (n/a)	2600	n/a
Repository depth (m)	1000	1000	1000
Water density (kg/m ³)		1000	
Hydrostatic water pressure (MPa)	10	10	10
Submerged rock density (kg/m ³)	n/a	1600 ^a	n/a
Effective lithostatic pressure of overburden rock above shale (MPa)	n/a	16 ^b	n/a
Buffer swelling pressure (MPa)	4	n/a	5
Total estimated external pressure load w/o glacial load	14	26^c	15
Hydrostatic pressure of groundwater in a 3000-m ice sheet (MPa)	30	30	30
Total estimated external pressure load with glacial load	44	56	45

Notes: a. Submerged rock density = saturated rock density – water density

b. Effective lithostatic pressure is derived from the submerged rock density

c. Sum of effective lithostatic pressure and hydrostatic water pressure

Microbial Activity

It is accepted that microbes exist at repository depth and are either contained in the sealing materials or will otherwise be introduced into the repository during operations. In order to reduce microbiologically influenced corrosion of UFCs, there is a need to inhibit microbial activity at or near the container surface in the repository. The microbial studies to date indicate that microbial activity and the possible formation of biofilms at the container surface can be potentially inhibited by enclosing the container in highly compacted bentonite (HCB) buffer material (Stroes-Gascoyne et al. 2006). In order to prevent significant microbial activity, the HCB material would need to satisfy one or more the following conditions in the near-field environment of the container:

1. a water activity <0.96, or
2. a swelling pressure ≥2 MPa together with an *in situ* dry density of 1.6 Mg·m⁻³ (i.e., an effective montmorillonite dry density of 1.4 g·cm⁻³) for a pore-water salinity <100 g·dm⁻³, or
3. a pore-water salinity >100 g·dm⁻³.

Although corrosive microbial metabolic by-products could be produced some distance away from the container, they would first need to diffuse through the HCB to cause corrosion.

3. CURRENT UFC CORROSION KNOWLEDGE

The following discussion is not a comprehensive discussion of the detailed knowledge of the corrosion behaviour of Cu and C-steel UFC in the expected repository environments. Instead, this Section presents a summary of what is known about the various forms of corrosion for each container material. More-detailed discussions can be found in Andra (2001); JNC (2000); Johnson and King (2003); Johnson et al. (1994); King and Stroes-Gascoyne (2000); King et al. (2001, 2002b); King (2005b, 2007); and Shoesmith (2006).

3.1 Copper UFC

3.1.1 Canadian Studies

An extensive experimental and modelling program has been carried out on the corrosion of copper UFC in Canada over the past 20 years (King and Kolar 2000, King 2005b). Experimental studies have focussed on uniform corrosion and stress corrosion cracking (SCC). Modelling studies have addressed uniform and localized (pitting) corrosion, SCC, microbiologically influenced corrosion (MIC), and predictions of the evolution of environmental conditions within a deep geological repository.

In terms of the uniform corrosion of copper, studies have been performed in the following areas:

1. anodic dissolution of Cu in Cl⁻ solutions (King and Litke 1989a,b),
2. anodic dissolution of Cu in Cl⁻/SO₄²⁻ solutions (King and Tang 1998),
3. anodic dissolution of Cu in Cl⁻/NH₃ solutions (King et al. 1997a),
4. effect of moderately alkaline pH on the anodic dissolution of Cu in Cl⁻ solutions (King et al. 2001),
5. cathodic reduction of O₂ and the effect of surface films (King et al. 1995a,b),
6. dissolution behaviour of CuCl₂·3Cu(OH)₂ and Cu₂O (King and Kolar 2000),
7. effect of mass transport, [Cl⁻], and [O₂] on the corrosion potential (E_{CORR}) of both rotating-disc and clay-covered electrodes (King et al. 1995c),
8. effect of HS⁻ on the E_{CORR} of rotating-disc and clay-covered electrodes and the simulation of the evolution of E_{CORR} in a repository (King and Stroes-Gascoyne 1997),
9. corrosion behaviour of Cu in compacted bentonite, including measurements of the corrosion rate, [Cu] profile, and the nature of the corrosion products (King et al. 1992, 1997b; Litke et al. 1992),
10. the extent of "localized" corrosion of Cu exposed to compacted bentonite (Litke et al. 1992),
11. effect of γ-radiation on the corrosion of Cu (King et al. 1992), and
12. measurements of the sorption of Cu(II) on both loose and compacted bentonite (Ryan and King 1994).

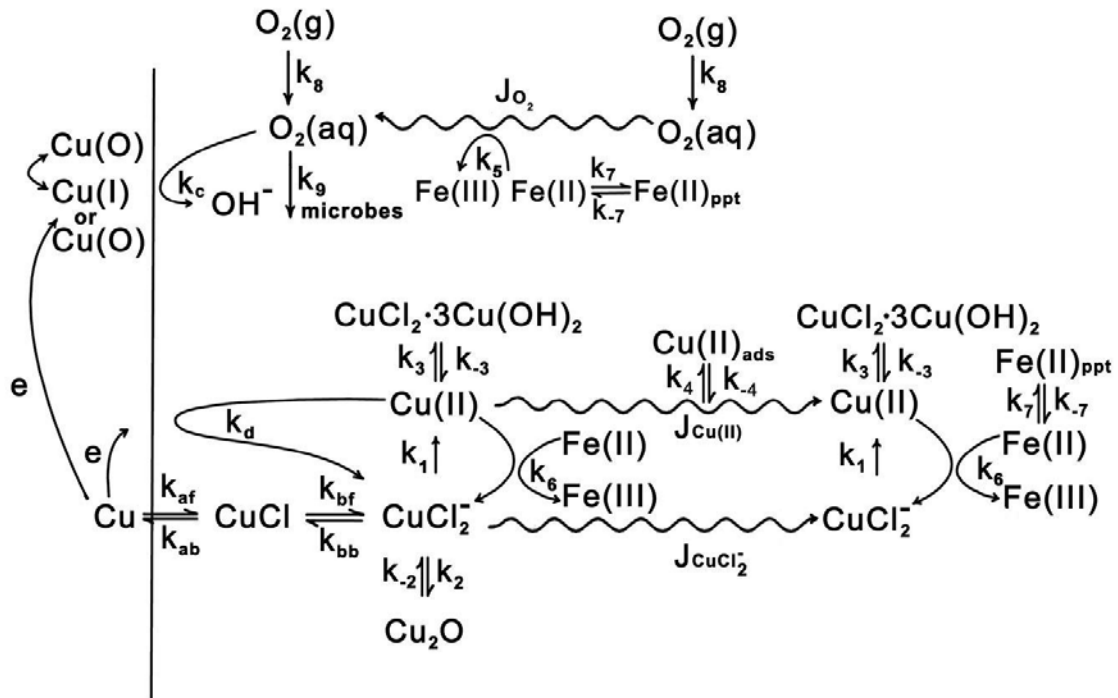


Figure 5: Reaction Scheme for the Corrosion of Copper in O₂-containing Chloride Environments in the Presence of Bentonite Clay (King and Kolar 2000).

Based on the results of these studies and on data from the literature, a reaction mechanism has been developed to account for the corrosion of Cu in compacted bentonite in the presence of Cl⁻-dominated pore water (Figure 5). This reaction scheme accounts for the various electrochemical, chemical, redox, adsorption/desorption, precipitation/dissolution, and mass transport processes involved in the overall mechanism.

The extent of localized (pitting) corrosion of Cu containers has been assessed using extreme-value analysis of literature pit-depth data (King and LeNeveu 1992) and by comparison of predicted E_{CORR} values to breakdown and re-passivation potential data from the literature (King et al. 2001). However, it is apparent from the nature of the corrosion profiles observed following exposure of copper to simulated repository conditions that the surface does not undergo pitting in the classical sense but rather a form of under-deposit corrosion characterized by the non-permanent spatial separation of anodic and cathodic processes (King and Kolar 2000). Thus, it is inappropriate to refer to this form of corrosion as "pitting" since the extent of damage is much less than would occur if the anodic and cathodic processes were permanently separated as occurs, for example, in the pitting of copper water pipes.

Stress corrosion cracking of copper and copper alloys is well known and is considered to be a potential container corrosion failure mechanism. The occurrence of SCC requires that a susceptible metal be exposed to sufficient tensile stress and an active SCC agent. Nitrite, ammonia and acetate are known SCC agents. A common feature for SCC of copper appears to be the formation of a thin oxide or tarnish film (Ikeda and King 2001). When this film does not form, SCC is not observed.

Tests have been performed to assess the SCC behaviour of copper in nitrite-, ammonia- and acetate-containing environments (Ikeda and Litke 2000, Ikeda et al. 2004, Litke and Ikeda 2006)). Although these species are not normally found in the natural groundwaters of the Canadian Shield, they could be introduced by mining activities, microbial activity or gamma radiolysis. The test results indicate that copper SCC susceptibility would decrease with decreasing concentrations of the SCC agents. There seems to be a threshold concentration level for each agent below which SCC would not occur. The experimental results also indicate that chloride in the saline pore-water of the bentonite material has an inhibiting effect on copper SCC in nitrite, ammonia and acetate environments. The SCC susceptibility appears to decrease with increasing chloride concentration. Most of these experiments have been carried out at room temperature. A number of SCC tests were carried out at elevated temperatures of about 100°C and 130°C in nitrite-only and nitrite/chloride mixtures (Ikeda and Litke 2007). The results indicate that elevated test temperatures would significantly reduce the SCC susceptibility in these test solutions. The inhibiting effect of chloride on SCC susceptibility also appears to be enhanced by the elevated test temperatures.

An extensive program has been conducted to determine whether microbes will be active in the near field close to the container surface (Stroes-Gascoyne et al. 2006). This program has demonstrated that:

- a water activity (a_w) <0.96 severely limits microbial culturability
- swelling pressures >2 MPa kill bacteria
- the evolution of the repository environment can be divided into three stages: Stage I – characterized by hot, unsaturated bentonite, during which the potential for significant MIC effects is small; Stage II – characterized by decreasing temperature and slow re-wetting of the bentonite, during which it is uncertain whether microbial activity can occur in cracks in the bentonite or at the UFC/bentonite and bentonite/host rock interfaces, and Stage III – characterized by saturated bentonite, high swelling pressure and uniform dry density, during which microbial activity will not occur in the bentonite, although there will remain a large "viable" microbial population.

In the absence of microbial activity close to the container, it is possible to claim that:

- biofilms will not form on the UFC surface, and
- the only form of MIC possible is that due to the diffusion of remotely produced metabolic by-products through the compacted bentonite to the container surface.

A series of reactive-transport models, based partly on the reaction scheme shown in Figure 5, has been developed in the Canadian program. These models have been used to make long-term predictions of various aspects of the performance of the container and repository, including:

1. uniform corrosion of Cu (King and Kolar 2000),
2. the possibility of SCC of Cu containers (King and Kolar 2004, Maak and King 2005),
3. the extent of MIC of Cu containers (King et al. 2002a, 2004),

4. consumption of O₂ and evolution of the repository environment (King and Kolar 2006),
5. effect of repository design and geological conditions on the consumption of O₂ and the corrosion of Cu UFC (King and Kolar 1997a), and
6. corrosion under unsaturated conditions (King and Kolar 1997b).

A number of analogue studies have also been performed, including the use of pit data from Bronze-Age artifacts and from a long-term underground corrosion study (King and LeNeveu 1992) and analysis of a bronze cannon submerged in sediments in the Baltic Sea for ~300 years (King 1995).

3.1.2 International Studies

Sweden

The Swedish nuclear waste management program was the first to develop a concept for a deep geological repository for spent nuclear fuel, leading to the development of the KBS-3 repository concept which has formed the basis of a number of other national programs (SKB 1983). The KBS-3 design is similar to the conceptual design for crystalline rock in Canada and comprises spent fuel assemblies inside an OFP-Cu canister supported by a cast iron insert, placed in a DGR in crystalline rock and sealed with HCB and backfill materials (Figure 6). The current Cu canister wall thickness is 50 mm, having been reduced from the original 200 mm (Andersson et al. 2004). The cast iron insert has a minimum membrane thickness of 60 mm. As a consequence, the canister is effectively self-shielding.

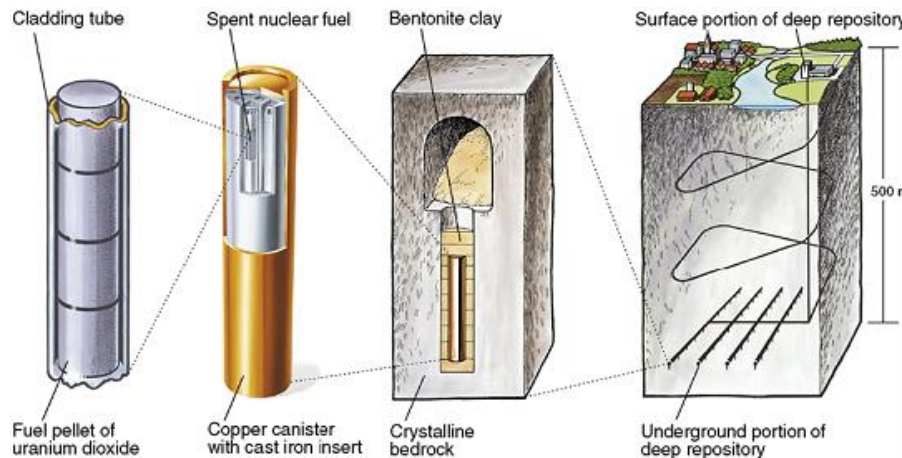


Figure 6: Schematic of the KBS-3 Repository Concept.

Environmental conditions for Cu canisters in a Swedish DGR may be similar to those expected in a Canadian repository in crystalline rock. Swedish groundwaters are anoxic and saline at repository depth with a composition similar to those given in Table 1 for the Canadian Shield, with the exception of the presence of 0.03-0.3 mg/L HS⁻ in Swedish waters (King et al. 2001).

The maximum canister surface temperature and temperature profile are similar to those in the Canadian program (Figure 3). The sealing system and target bentonite compaction density would also be similar, with a criterion of a swelling pressure >2 MPa for the suppression of microbial activity (Hedin 2006). External loads would also be similar to those expected in a Canadian repository (Table 3).

For the purposes of predicting the long-term corrosion behaviour, the canisters are assumed to be subject to uniform corrosion and pitting. The extent of uniform corrosion is assessed through a combination of mass-balance and mass-transport calculations (King et al. 2001). Corrosion caused by the initially trapped O₂ is accounted for using a mass-balance argument and is equivalent to ~80 µm of wall penetration. Corrosion due to sulphide, either from the groundwater, from pyrite impurities in the clay, or from microbial activity, is calculated based on a mass-transport basis and accounts for an additional 0.27 mm of corrosion over a period of 10⁶ years. The extent of pitting is assessed using a pitting factor, with a value of between 2 (considered realistic) and 5 (considered to be conservative). In total, therefore, the maximum wall penetration is predicted to be 1.5-2.0 mm after 10⁶ years.

Other corrosion processes, such as SCC and MIC, are considered to be unlikely in the repository environment (King et al. 2001). An allowance is made for microbially produced HS⁻ in the lifetime assessment, but the major argument against MIC is the use of HCB with a swelling pressure of >2 MPa which it is claimed precludes microbial activity in the near field (Hedin 2006). For SCC, it is claimed that cracking is only likely under the initial oxidizing conditions and then only in the unlikely event that certain species (acetate, nitrite, and ammonia) are present (SKB 2006). Furthermore, creep will tend to relieve the stress at the crack tip.

Experimentally, the Swedish program has focussed its efforts on specific areas, including:

1. measurement of creep properties of oxygen-free Cu and the effects of alloying additions and impurities (Henderson et al. 1992, Yao and Sandström 2000),
2. corrosion behaviour of the cast iron insert and the effect on the outer Cu shell of expansive corrosion products (Smart et al. 2001, 2002, 2006),
3. stress corrosion cracking (Benjamin et al. 1988, Khanzhin and Nikulin 2005, Pettersson and Oskarsson 2000, Rosborg 1998, Rosborg and Svensson 1994, Rosborg and Werme 2001),
4. *in situ* corrosion tests at Äspö (Karnland et al. 2000; Rosborg et al. 2002, 2004), and
5. corrosion of Cu in sulphide environments (Smith et al. 2007).

Finland

The Finnish program is based on the KBS-3 concept, but is somewhat more advanced in that a site has been selected (Olkiluoto) and construction of the ONKALO underground characterization facility is under way at the repository site.

An alternative “supercanister” concept is being considered for the KBS-3H concept for horizontal emplacement. The super-container would comprise a loaded Cu/cast iron canister and pre-compacted bentonite blocks supported by a cage-like structure.

The environmental conditions expected in the Finnish repository are similar to those in a Swedish repository, except that the groundwater at Olkiluoto is more saline (up to 16,000 mg/L Cl⁻), has a higher [HS⁻] (up to 1 mg/L) and contains CH₄ (up to 400 ml/L). In addition, the possibility of ammonia formation in the groundwater is acknowledged, although the concentration is likely to be below the detection limit (<0.05 mg/L, King et al. 2002b).

Like the Swedes, the Finns have performed a number of focussed experimental studies on the corrosion behaviour of Cu. These studies have included:

1. the corrosion of Cu in deaerated Cl⁻ solutions (Bojinov and Mäkelä 2003),
2. SCC of Cu under deaerated synthetic groundwater with and without ammonium ions (Aaltonen et al. 1984, Arilahti et al. 2000), and
3. SCC of Cu in acetate solutions (Betova et al. 2005).

There is no evidence that H₂O acts as an oxidant for Cu in highly saline groundwaters ([Cl⁻] > 1 mol·dm⁻³). Furthermore, the SCC studies have not shown any evidence for cracking under anoxic conditions.

Switzerland

Although the Swiss at one time considered the construction of a DGR in crystalline rock, the focus of the research program is now entirely on a possible deep geological repository in sedimentary Opalinus Clay deposits (Figure 7) (Nagra 2002). The reference canister material is C-steel, although the possibility of using a Cu canister is retained should there be a need for longer-term containment (Johnson and King 2003).

There is currently no reference design for a Cu canister in the Swiss program.

The Opalinus Clay host rock possesses a very low permeability ($K \sim 2 \times 10^{-14}$ m/s), so that less reliance would need be placed on the canister as an absolute barrier (Nagra 2002). Pore water salinity is of the order of ~ 0.2 mol·dm⁻³ and the Opalinus Clay contains ~ 1 wt.% pyrite, some of which could be oxidized to sulphate and/or thiosulphate during the operational phase and the early aerobic period.

Canisters would be placed on blocks of pre-compacted bentonite and the remaining space in the disposal tunnels filled with granular bentonite to a dry density of ~ 1.5 Mg·m⁻³ using an auger system (Figure 7). After complete saturation, bentonite placed in this fashion would have an hydraulic conductivity of $< 10^{-13}$ m/s and a swelling pressure of 2-4 MPa.

Geological repository (SF/HLW/ILW)

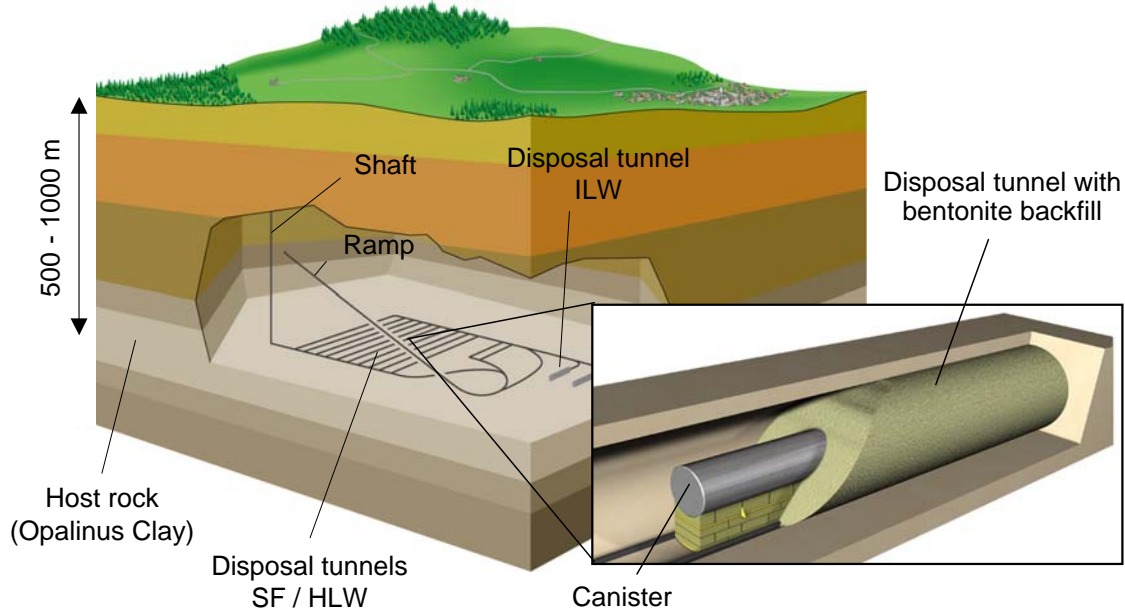


Figure 7: Conceptual Design for a Swiss Deep Geological Repository in Opalinus Clay.

The low initial moisture content of the granular bentonite (~10% saturation) has a number of effects on the evolution of environmental conditions, including:

- higher initial inventory of O_2 than in bentonite compacted with a higher initial degree of saturation (although the quantity of O_2 only amounts to $\sim 10 \text{ mol}_{O_2} \cdot \text{m}^{-2}$),
- longer re-saturation times, although a saturation time of only ~ 100 years is predicted, and
- lower thermal conductivity resulting in higher canister temperatures.

Partly as a result of the lower thermal conductivity and partly a result of the higher ambient temperature at repository depth, the maximum canister surface temperature is expected to be in the range $150\text{-}160^\circ\text{C}$ (Johnson and King 2003).

The maximum external load due to lithostatic stress from the Opalinus Clay and buffer swelling, the hydrostatic pressure, and the pressure of H_2 gas (for a steel canister) is expected to be in the range $30\text{-}35 \text{ MPa}$ (Johnson and King).

Microbial activity close to the canister surface is not considered because of the use of highly compacted bentonite.

Nagra has not performed any specific corrosion studies for their back-up Cu canister design. Their lifetime assessment follows that carried out by SKB, with a slightly higher flux of HS^- due to dissolution of pyrite in the host Opalinus Clay (Johnson and King 2003).

Japan

The Japanese program is based on a generic site with a bentonite-based sealing material. The reference canister material is C-steel, although both titanium and copper have also been considered (JNC 2000). Approximately 40,000 overpacks would be required to dispose of the projected inventory of high-level waste in Japan. Site selection is based on the concept of volunteerism.

Since no specific site has been selected in Japan, the detailed environmental conditions in the DGR have not been defined. To support ongoing R&D studies, reference “fresh” and “saline” groundwaters have been defined. Repository-specific environmental conditions have not been defined, but are likely to be similar to those expected in other bentonite-backfilled designs.

There have been relatively few studies in Japan in support of the use of Cu containers. Imai et al. (1996) studied the anodic polarization of Cu in the presence of various groundwater species, but no further studies have been published. More recently, Taniguchi and Kawasaki (2007) reported the SCC of Cu in sulphide environments. An apparent threshold $[HS^-]$ of $\sim 10^{-3} \text{ mol}\cdot\text{dm}^{-3}$ was reported, although further work is required to confirm that the cracking observed is due to environmental effects rather than creep.

A conservative approach was taken for the estimation of lifetimes of Cu containers (JNC (2000)). A mass-balance argument was used to predict the extent of corrosion by O_2 (0.6-3.6 mm) and due to HS^- produced by SRB (0.1-1.1 mm). In the latter case, it was assumed that all SO_4^{2-} in the bentonite was reduced to HS^- by SRB, despite the fact that the specified bentonite dry density of $1.6 \text{ Mg}\cdot\text{m}^{-3}$ would be sufficient to suppress microbial activity. In addition, sulphide present in the groundwater was estimated to contribute an additional 8.1 mm of corrosion over a period of 1000 years, as the result of assuming a groundwater $[HS^-]$ of $0.03 \text{ mol}\cdot\text{dm}^{-3}$, a concentration more than two orders of magnitude higher than in the groundwater. Pitting was assessed using a pitting factor of 3 (based on Swedish data), resulting in a total wall loss of 27-39 mm in 1000 years. This very conservative estimate of lifetimes for Cu containers is clearly at odds with those from other national programs considering the use of Cu as a corrosion barrier.

3.1.3 Summary of Current Knowledge of Copper UFC Corrosion

Based on the brief description of the state-of-the-art above, this Section provides a summary of the existing knowledge of various aspects of Cu UFC corrosion.

Uniform Corrosion

In general, there is a good understanding of the uniform corrosion behaviour of Cu in conditions expected in a DGR in either crystalline rock or Ordovician sedimentary deposits. The mechanism of corrosion in O_2 -containing chloride environments is understood (Figure 5) and many of the necessary rate constants and other parameters required for modelling are available.

Areas in which there is abundant knowledge from either the nuclear waste management or open corrosion literature include:

1. the mechanism of the uniform corrosion of Cu in O₂-containing chloride environments in porous media,
2. anodic dissolution of Cu in Cl⁻ and Cl⁻/SO₄²⁻ mixtures,
3. cathodic reduction of O₂ on Cu,
4. the diffusivity of species in compacted buffer material,
5. adsorption/desorption behaviour of Cu(I) and Cu(II) on bentonite,
6. the kinetics of redox reactions between O₂ and Fe(II),
7. the effect of γ -radiation on Cu corrosion (although the extent of knowledge is limited, so too is the impact at the expected radiation levels),
8. a mechanistically-based mixed-potential model for predicting E_{CORR}, and
9. the effect of [Cl⁻] on the corrosion of Cu.

Localized Corrosion

It is now generally accepted that a Cu UFC will not undergo pitting corrosion in the classical sense of the term, but instead a form of surface roughening characterized by temporary spatial separation of anodic and cathodic sites.

There is also information available for assessing the extent of localized corrosion of Cu UFC using alternative approaches, including

1. a model for predicting E_{CORR} for comparison with the film breakdown or re-passivation potentials, and
2. a database of pitting factors for statistical analysis (King and LeNeveu 1992)

Stress Corrosion Cracking

The effects of stress corrosion cracking (SCC) agents, chloride concentration, and oxidant on the SCC behaviour of OFP copper have been studied using short-term SCC experiments. There is a reasonable understanding of SCC behaviour of OFP copper in nitrite, ammonia and acetate environments (with and without chloride) during the initial aerobic phase.

An approach to predicting the SCC behaviour of Cu UFC during the initial aerobic phase has been developed based on a decision-tree approach (King and Kolar 2005). This approach is supported by existing information on the bulk potential and pH for the SCC of Cu in nitrite solutions, partly determined in the Canadian R&D program.

Microbiologically Influenced Corrosion

There is widespread international consensus that the culturability and/or viability of microbes is compromised in compacted bentonite with a dry density of $\geq 1.6 \text{ Mg}\cdot\text{m}^{-3}$. Microbial activity can also be limited by the presence of highly saline pore water with a salinity $>100 \text{ g/L}$.

The current approach to assessing the extent of MIC is based on an argument that microbial activity will not occur close to the container surface, thus precluding biofilm formation and limiting corrosion to that resulting from the diffusion of aggressive metabolic by-products produced remote from the container. This argument is, in turn, based partly on the following observations and conclusions:

1. a bentonite dry density of $1.6 \text{ Mg}\cdot\text{m}^{-3}$ is sufficient to prevent significant microbial activity in compacted bentonite,

2. a pore-water salinity of >100 g/L is sufficient to prevent microbial activity in compacted bentonite,
3. a water activity (a_w) of less than 0.96 is sufficient to prevent significant microbial activity in compacted bentonite, and
4. biofilm formation on the container surface can be prevented through the use of highly compacted bentonite.

A predictive model for microbial activity in a repository has been developed based on this approach (King et al. 2002a, 2004).

Preferential Weld Corrosion

A limited amount of information is available regarding the susceptibility of welded OFP Cu to various forms of corrosion. There is some information on the SCC susceptibility of EB-welded Cu from the Canadian program (Ikeda and Litke 2000) and information on grain boundary attack from the Swedish program (Gubner et al. 2006). Although the weld metal and parent material exhibit similar crack growth rates in $0.1 \text{ mol-dm}^{-3} \text{ NaNO}_2$ solution at room temperature, the weld metal exhibits a lower threshold conditional stress intensity for crack initiation (Ikeda and Litke 2000). No grain boundary attack was observed for either EB- or friction stir welded Cu, but higher uniform corrosion rates are possible because the weld metal is anodic to the base metal (Gubner et al. 2006).

Corrosion During the Unsaturated Period

Conceptually, the processes occurring during the unsaturated phase are understood (King 2006). Initially, the UFC surface will dry sufficiently that aqueous corrosion will not occur. Subsequent deliquescence of salts on the surface will lead to wetting of the container, at first at isolated salt crystals (leading to the possibility of localized attack) and then more uniformly as the relative humidity increases and the temperature decreases. Depending upon the time dependence of the UFC surface temperature and relative humidity, the period of unsaturated conditions could persist for a few 100's of years, during which much if not all of the initially trapped O_2 would be consumed. Therefore, the initial aerobic phase could correspond to the period of unsaturated conditions; conditions that have not been studied in detail.

No specific experimental studies of the corrosion of copper UFC under unsaturated conditions have been performed, although a conceptual understanding of the important processes during the unsaturated phase is available and deliquescence data to predict the onset of wetting are available (King 2006).

Hydrogen Gas Issues

Hydrogen generation is clearly less of an issue for a Cu UFC than for a C-steel container, but still needs to be considered for those initially defective Cu containers or in the event of early UFC failure. Exposure of the C-steel support vessel will result in H_2 generation under anoxic conditions. Corrosion rates of cast iron galvanically coupled to Cu under anaerobic conditions are similar to those for uncoupled iron (Smart et al. 2005).

Differences Between Crystalline and Sedimentary Host Rocks

Until such time that detailed repository-depth, site-specific information is available from potential sites for a deep geological repository in Canada, it is difficult to be certain of the exact nature of the environmental differences between crystalline and sedimentary host rocks. However, available generic information on these two potential host rock formations suggests that the major differences from a corrosion viewpoint between these two types of sites are (i) the potential higher salinity, especially [Cl⁻], for Ordovician sedimentary deposits, and (ii) the potential lower hydraulic conductivity of sedimentary deposits, possibly resulting in longer re-saturation times and a longer period of unsaturated conditions.

Based on our current understanding of environmental conditions in sedimentary deposits, however, there is an adequate understanding of the following processes:

1. the effect of [Cl⁻] on the electrochemical behaviour of Cu and on the E_{CORR} of the container (King and Kolar 2000), and
2. conceptual understanding of the processes during the unsaturated phase (King 2006).

Lifetime Predictions

In general, prediction of the UFC lifetime requires:

1. a numerical prediction of the extent of damage and/or mechanical degradation of the container leading to failure, and
2. a demonstration of sufficient mechanistic understanding to justify such a prediction.

The actual numerical prediction may be based on either empirical data (e.g., measured corrosion rates, pitting factor from samples exposed to corrosive environments) or mechanistic modelling (e.g., reactive-transport models, mixed-potential models).

The methodology for lifetime predictions of Cu UFC is relatively mature. A combination of mechanistic and empirical damage prediction supported by mechanistic understanding is used to estimate the extent of uniform and localized corrosion and to support the exclusion of SCC and MIC. A series of 1-D reactive-transport models (the Copper Corrosion Model, CCM) has been developed to predict the consequences and/or probability of uniform corrosion (CCM-UC), SCC (CCM-SCC), and MIC (CCM-MIC) (King and Kolar 2000, 2004, 2006; King et al. 2002a, 2004; Maak and King 2005). An important component of these models is a mixed-potential model for predicting the time dependence of E_{CORR} of the container, which can be used to predict the probability of various localized corrosion processes. Furthermore, these reactive-transport models predict the evolution of environmental conditions (redox, pore-water salinity, extent of adsorption and precipitation of corrosion products) in the buffer and backfill sealing materials.

In addition to the reactive-transport models for uniform corrosion, SCC, and MIC, statistical approaches for the analysis of literature pit-depth data are also available (King and Kolar 2000).

Natural and Archaeological Analogues

Natural analogues and archaeological artifacts, used judiciously, can provide useful supporting evidence for UFC lifetime predictions. This is particularly so for the Cu UFC because of the existence of natural analogues (in the form of native Cu deposits) and ancient Bronze Age artifacts (Bresle et al. 1983).

For Cu UFC, the following studies of natural analogs and archaeological artefacts have been reported:

1. native Cu deposits,
2. Bronze Age archaeological artifacts (Bresle et al. 1983),
3. copper lightning conductor plates (Hallberg et al. 1984),
4. a bronze cannon from the Kronan shipwreck (Hallberg et al. 1988, King 1995),
5. copper-uranium deposits (Mildowski et al. 2002),
6. Ferryland historical site, NL (McMurry et al. 2001), and
7. U.S. NBS long-term underground corrosion studies (Romanoff 1989).

3.2 Carbon Steel Containers

3.2.1 Canadian Studies

Recently, the Canadian Technical R&D Program has investigated the possibility of using C-steel as a container material for the isolation of used fuel in Canada (King 2005b). The focus has been on the use of this material for a potential DGR in Ordovician sedimentary deposits, which exhibit a hydraulic conductivity approaching that of the Opalinus Clay in Switzerland. As in the Swiss program, the use of C-steel UFC may be acceptable from a safety perspective because of the low permeability of the geosphere.

A detailed design for a Canadian C-steel UFC is not yet available. The grade of material and the container fabrication and sealing procedures have also not yet been defined.

With some exceptions, the environmental conditions for C-steel UFC would be the same as those for Cu containers summarized above. However, important differences will exist, including:

1. the presence of higher salinity groundwaters in Ordovician sedimentary deposits compared to those in the crystalline rock of the Canadian Shield (Table 2),
2. higher external loads due to lithostatic pressures in shale formations (Table 3),
3. the formation of a H₂ gas phase and resulting gas pressure,
4. compaction of the bentonite in contact with the container due to the formation of expansive corrosion products, and
5. the potential alteration of smectite by reaction with Fe(II) corrosion products with the formation of non-swelling illite, and the consequences for microbial activity at the container surface.

Preliminary corrosion estimates in Canada have been based largely on the approaches developed in the Swiss and Japanese programs (King 2005b, 2007). Corrosion during the aerobic phase is estimated based on mass-balance arguments. A constant corrosion rate is assumed during the long-term anoxic period. The extent of localized corrosion (or surface

roughening) is estimated based on a depth- (or a time-) dependent pitting factor derived from an analysis of long-term underground corrosion studies supplemented by short-term laboratory data. The estimated ranges of wall penetration are 9-34 mm after 10,000 years (1-10 mm as uniform corrosion and 8-24 mm as pitting) and 34-175 mm after 100,000 years (10-100 mm as uniform corrosion and 24 to 75 mm as pitting). The variability results primarily from the uncertainty in the long-term anaerobic corrosion rate ($0.1-1 \mu\text{m}\cdot\text{a}^{-1}$) (King 2005b, 2007).

Other corrosion processes, such as SCC, H-related effects, MIC, and preferential weld corrosion are not considered in the lifetime assessment. It is assumed that these processes can be excluded from consideration based on proper specification of the composition of the steel, the welding and post-weld stress-relief procedures, or (in the case of MIC) the bentonite properties (King 2007). These assumptions remain to be validated.

3.2.2 International Studies

Switzerland

A feasibility study of the disposal of spent fuel, high-level waste, and long-lived ILW (Project Entsorgungsnachweis (EN)) was submitted to the regulatory authorities in 2002. In June 2006, the Swiss Federal Council stated that the project had successfully demonstrated that disposal of SF/HLW and ILW is technically feasible in Switzerland. The Swiss program undertook a major safety case in 2002 in support of their Opalinus Clay repository concept (Nagra 2002). The concept is illustrated in Figure 7.

Experimental studies of the corrosion behaviour of C-steel were performed in Switzerland in the 1980's (Kreis 1991, Simpson 1989, Simpson and Valloton 1986). The studies largely involved the measurement of corrosion rates in Swiss groundwaters in support of the then reference concept in crystalline rock. A number of measurements of the corrosion rate and rate of H_2 generation under anoxic conditions were performed at that time. No specific corrosion studies have been performed in support of the Opalinus Clay repository concept.

The review of the expected corrosion behaviour and estimated canister lifetimes by Johnson and King (2003) borrowed significantly from the earlier Japanese assessment (JNC 2000). Thus, the extent of uniform corrosion and pitting were quantitatively assessed, with reasoned arguments made for excluding SCC, H-related effects, and MIC. Aerobic corrosion was calculated based on the amount of trapped O_2 in the sealing materials. Corrosion during the anaerobic phase was estimated using a constant anaerobic corrosion rates of $1 \mu\text{m}\cdot\text{a}^{-1}$ with an additional $0.02 \mu\text{m}\cdot\text{a}^{-1}$ to account for HS^- diffusion from the host Opalinus Clay. A constant pitting factor of 10 was used, based on empirical data from the U.S. National Bureau of Standards underground corrosion study (Romanoff 1989) supplemented by Japanese laboratory data (JNC 2000).

Of the processes not included in the lifetime prediction, SCC was discounted based either on the absence of the appropriate environment (in the case of caustic, nitrate, phosphate, CO-CO_2 , high-temperature water, and concentrated $\text{HCO}_3^-/\text{CO}_3^{2-}$ environments) or the absence of cyclic load (in the case of $\text{HCO}_3^-/\text{CO}_3^{2-}$ environments, i.e., the so-called high- and near-neutral pH $\text{HCO}_3^-/\text{CO}_3^{2-}$ environments known to cause SCC of pipeline steels).

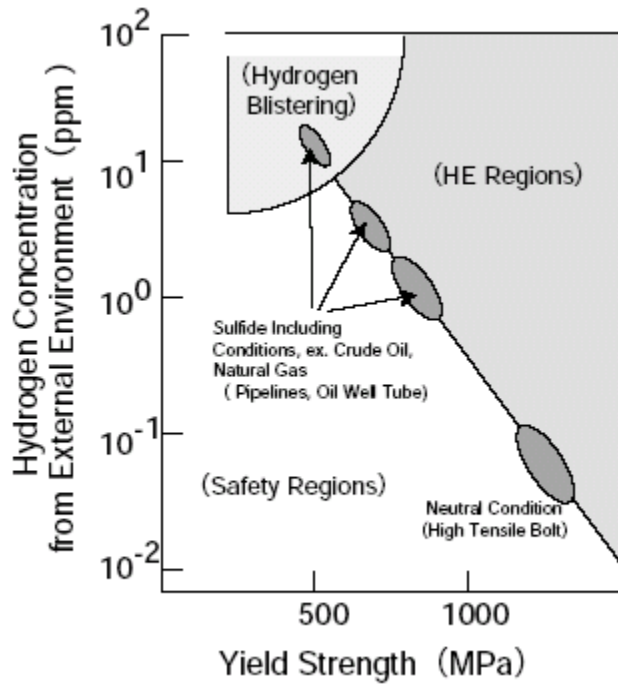


Figure 8: Schematic Illustrating the Zones of Susceptibility of Carbon Steel to Hydrogen-related Degradation Mechanisms Based on the Absorbed Hydrogen Concentration and the Material Yield Strength (JNC 2000).

The possibility of H-related phenomena was determined by comparing the sub-surface absorbed H concentration (c_H^0) to the (temperature-dependent) threshold concentration (c_H^0)_{TH} (Figure 8). The criterion for the absence of H-related effects is

$$c_H^0 < (c_H^0)_{TH} \quad (1)$$

Experimental c_H^0 values determined in dilute groundwaters are of the order of 1 ppm (Cheng et al. 2000, King et al. 2005), suggesting that blistering and hydrogen-induced cracking will not occur for the reference C-steel yield strength for the canister of 256 MPa (Johnson and King 2003). Furthermore, the data on which Figure 8 is based were obtained from "dirty" ferrite-pearlite steels that are more susceptible to hydrogen effects than modern steels. Modern steels are more resistant to hydrogen effects because of better control of the number and shape of MnS inclusions and because of small alloying additions that limit the generation and absorption of hydrogen (King 2007). Therefore, it is likely that a UFC manufactured from a modern grade of steel would be less likely to fail because of hydrogen-related degradation mechanisms.

The reasoned argument for MIC is the same as that used in Sweden and Canada, i.e., that biofilm formation on the canister surface will not occur as microbial activity in the near-field will be prevented through the use of highly compacted bentonite and that only corrosive metabolic by products formed further away need to be taken into account (Johnson and King 2003).

Japan

The Japanese have performed a significant number of experimental studies on the corrosion of C-steel. Much of this work has been focussed on the passivation and localized corrosion of C-steel under repository conditions (see, for example, Fukaya and Akashi 1999; Fukuda and Akashi 1995; Fukuda et al. 1995; Nakayama and Akashi 1991; Taniguchi et al. 1998, 1999, 2002; Tsuchiya et al. 1996). It appears unlikely that a C-steel overpack would be passive in compacted bentonite at the expected pore-water pH of ~7.5-8, and voltammetric measurements in compacted bentonite appear to confirm this conclusion (JNC 2000). Honda et al. (1991) reported an extensive study of the corrosion of C-steel in compacted bentonite under aerobic conditions, studying the effects of pore-water salinity, exposure time, bentonite density, and temperature. The corrosion behaviour is dominated by mass-transport of O₂ to the corroding surface. A good summary of the pre-2000 work is given in JNC (2000).

Kojima et al. (1995) raised the issue of whether the long-term anoxic corrosion rate could increase with time due to galvanic coupling between Fe₃O₄ corrosion product (acting as the cathode) and the underlying C-steel. Taniguchi (2003) confirmed the earlier observations but concluded that the effect would be transitory and have little impact on the UFC lifetime.

More recently, attention has been paid to the properties and corrosion performance of welded C-steel (Asano and Aritomi 2005; Asano et al. 2005, 2006a,b; Mitsui et al. 2005, 2006a-c). Some work has also been performed on the effects of bentonite clay on SRB, although no English-language version of this work is available (N. Taniguchi, private communication 2005).

A number of lifetime assessments have been performed using a combination of empirical and mechanistic data (JNC 2000). Both statistical and empirical (pitting factor) approaches have been used to assess the extent of localized corrosion (Akashi et al. 1990, Asano et al. 1992, JNC 2000). A simple electrochemical model, as well as an empirical corrosion rate, have been used to assess the extent of uniform corrosion (JNC 2000). Hydrogen-related degradation mechanisms have not been considered in lifetime assessments based on the criterion in Equation 1. Stress corrosion cracking has not been specifically addressed. Microbial effects are either excluded based on the non-survivability of microbes in compacted bentonite or conservatively assessed by assuming that all SO₄²⁻ ions are converted to HS⁻ (JNC 2000).

Belgium

Belgium has had an ongoing high-level nuclear waste program for the past 20-30 years, principally focussed on sedimentary deposits such as Boom Clay. A major safety assessment (SAFIR-2) was published in 2002 (ONDRAF-NIRAS 2002). An expert Corrosion Study Panel reported on the Belgian concept in 2004 (ONDRAF-NIRAS 2004). Partly as a result of that review, the Belgian program has recently re-designed its repository concept in favour of a supercontainer comprising a C-steel (or 2¼Cr 1Mo steel) overpack encased in a cementitious buffer material surrounded by a stainless steel envelope (Bel et al. 2006). The supercontainers would be placed in horizontal boreholes in Boom Clay sedimentary deposits (Figure 9). One of the major driving forces behind the re-design was the desire to create an alkaline near-field environment for improved corrosion resistance of the steel canister.

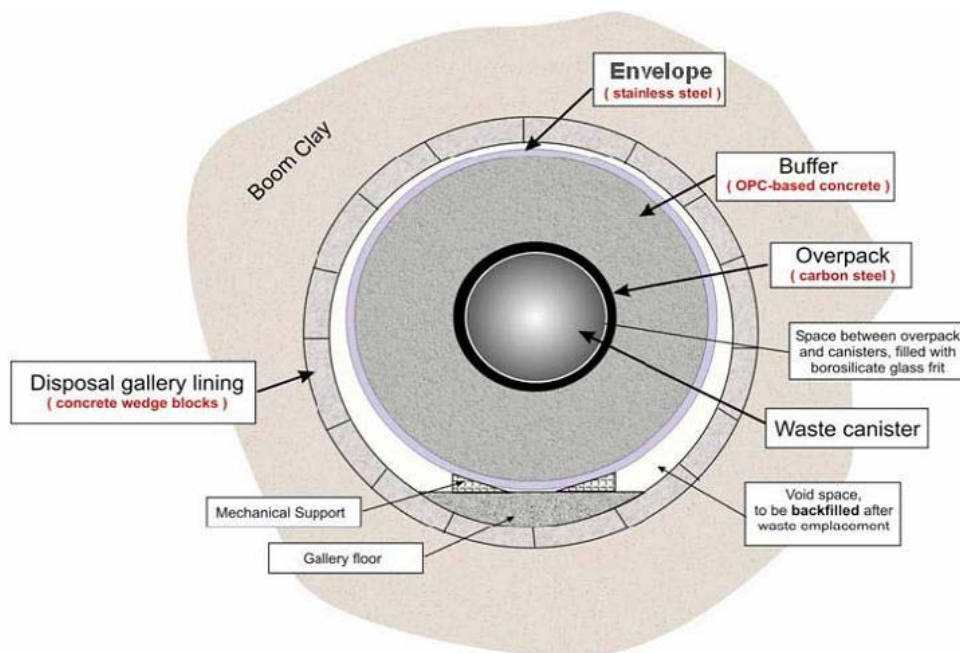


Figure 9: Schematic of the Conceptual Supercontainer Design for the Belgian Program (Bel et al. 2006).

The design life of the engineered barrier system is about 2,000 years (ONDRAF-NIRAS 2004).

Like many sedimentary deposits being considered worldwide, Boom Clay exhibits a low hydraulic conductivity and is, therefore, considered to be an excellent barrier to radionuclide migration. The steel overpacks would be placed in concrete-lined galleries with an integral cementitious buffer.

Interstitial waters in the Boom Clay contain ~10 ppm Cl^- and ~1 ppm SO_4^{2-} , but oxidation of pyrite inclusions can lead to 1000's ppm SO_4^{2-} and 100's ppm $\text{S}_2\text{O}_3^{2-}$ (thiosulphate).

A wide range of materials have been considered for the overpack in the Belgian program, including C-steel, Ni alloys, Ti alloys, and stainless steels (Casteels et al. 1985, Debruyne 1990, Debruyne et al. 1991, Kursten and Druyts 2000, Kursten et al. 1996). A major concern throughout the program has been the aggressiveness of environments formed by the oxidation of impurities in the clay and during unsaturated conditions. Concerns over the localized corrosion of C-steel by thiosulphate and excessive rates of general corrosion resulted in a recommendation against the use of this material prior to the adoption of the supercontainer design illustrated in Figure 9.

From a corrosion viewpoint, the rationale behind the supercontainer is (ONDRAF-NIRAS 2004):

- the use of a pitting-resistant grade of stainless steel for the outer envelope, although some localized penetration is considered acceptable,
- the avoidance of SCC failure of the outer stainless steel envelope,
- pH-conditioning by the cementitious buffer material to ensure passivation and corrosion resistance of the overpack material (C-steel or 2¼Cr 1Mo steel), and
- 20-mm-thick overpack to provide adequate corrosion resistance for the thermal period.

France

France is considering the horizontal placement of C-steel canisters containing HLW and, possibly, spent fuel in boreholes in argillaceous clay (Figure 10). The boreholes would not be backfilled to allow for retrieval for a period of up to 300 a.

Interstitial pore waters in the Callo-Oxfordian argillaceous clay deposits tend to be Ca-Na-Cl-dominated solutions with a [Cl] of ~0.03-0.04 mol/kg (Andra 2001). The waters themselves are anoxic ($E_h -138 \text{ mV}_{\text{NHE}}$) and the environment within the boreholes is expected to become anoxic once the initially present atmospheric O_2 has been consumed. The possibility of an extended humid period is acknowledged (up to a few 100 years), with temperatures as high as 185°C.

A range of materials has been considered for the canister material, including Cu and Ti alloys, but only non-alloy steels and Fe-Cr-Ni-Mo steels are considered potential materials at this stage (Andra 2001).

In the near-term, conditions are expected to be dominated by unsaturated atmospheric conditions, resulting in uniform corrosion at a predictable rate. A 55-mm-thick C-steel container is considered to provide sufficient containment for the necessary period of reversibility and to be suitable for long-term disposal if necessary (Andra 2005).

United Kingdom

The high-level nuclear waste program in the UK has recently been re-started under the auspices of the Nuclear Decommissioning Authority (NDA). Although the current reference concept is based on the KBS-3V design with Cu/cast iron canisters, work in the 1980's focussed on the possible use of C-steel.

The current repository environmental conditions are uncertain until such time that a site has been selected. In the 1980's, however, the focus was on a granitic host rock, although some work was also done in other geomedias. Much of this earlier work, therefore, involved the use of both dilute granitic and saline waters. The reference concept at that time included the use of a bentonite-based backfill material. The environmental conditions considered were similar to those expected in Canada, albeit with lower salinity granitic groundwater.

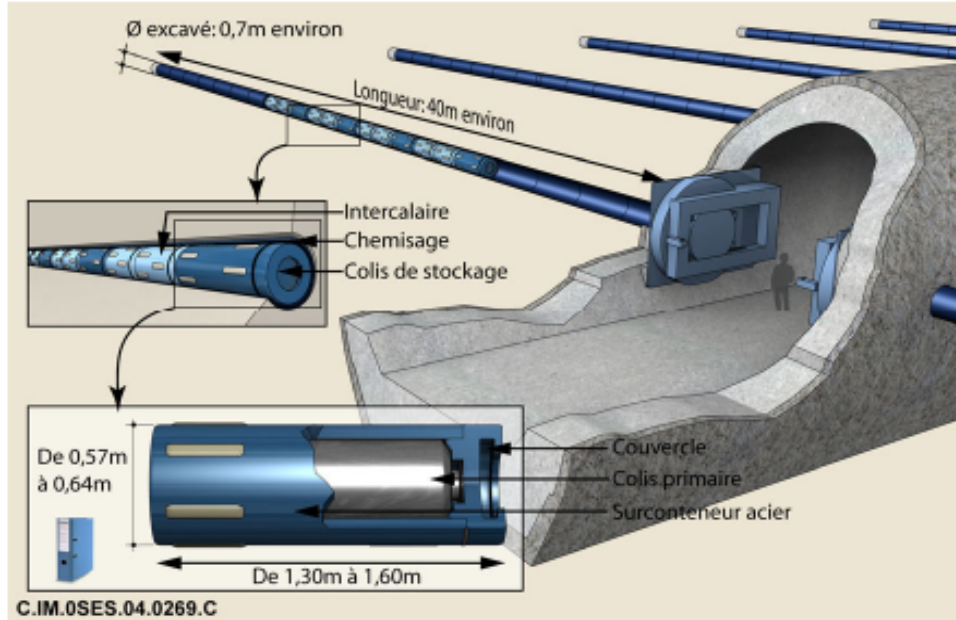


Figure 10: Conceptual Designs for the Long-term Management of High-level Waste in France (Andra 2005).

Marsh and co-workers (Marsh and Taylor 1988; Marsh et al. 1987, 1988, 1989) conducted various experimental corrosion studies on C-steel in support of the UK program. Corrosion processes considered included uniform corrosion, localized (pitting) corrosion, SCC, and MIC. Uniform corrosion was treated using either empirical corrosion rate data (and a mass-balance argument for the aerobic period) or a film-free electrochemical model. The effects of radiolysis were also included. Localized corrosion was predicted using extreme-value statistical analysis of laboratory pit-depth data for the period of passivity of the container (if any). The period of passivity was equated to the period for which the transport-limited supply of O_2 exceeded the passive current density under the particular environmental conditions. SCC was excluded from consideration by arguing that post-weld heat treatment could be used to anneal the container. When addressed, the extent of MIC was determined based on the assumption that the amount of organic carbon limited the extent of HS^- production by SRB (Marsh and Taylor 1988).

Sweden

Although the Swedish program is based on the use of a Cu outer corrosion barrier, SKB have performed a number of experimental projects on the corrosion of the cast iron insert.

The major areas of interest are:

1. the anaerobic corrosion rate of cast iron and C-steel, and the effect of H_2 gas generation on the integrity of the bentonite buffer, and
2. the expansion of steel corrosion products, and the effect on the integrity of the outer Cu corrosion barrier.

Smart et al. (2001, 2002) have measured the anaerobic corrosion rate in dilute granitic water, the main conclusions being (Smart et al. 2001):

- “Carbon steel undergoes active anaerobic corrosion in artificial Swedish granitic groundwater, although a magnetite film retards the rate of corrosion. The outer layer is a loose porous film that is easily disrupted, while the inner layer is adherent and crystalline and forms the main rate-controlling layer.
- The anaerobic corrosion rate of bare carbon steel at 30°C, 50°C and 85°C is of the order of 10 to 30 $\mu\text{m}\cdot\text{yr}^{-1}$ initially, but as an oxide film forms the rate decreases to $<0.1 \mu\text{m}\cdot\text{yr}^{-1}$. The initial corrosion rate increases with increasing temperature.
- Hydrogen overpressures up to 100 atmospheres have no significant effect on the corrosion rate or the corrosion potential. This suggests that the corrosion reaction is therefore anodically limited, rather than cathodically limited.
- Higher anaerobic corrosion rates occur above an ionic strength threshold.
- The anaerobic corrosion rate is not limited by the concentration of dissolved ferrous ion.
- The presence of the radiolysis products ammonia and nitric acid do not significantly affect the anaerobic corrosion rate.
- No localized corrosion initiates under anoxic conditions.
- Welding has no effect on the anaerobic corrosion rate in granitic groundwaters.
- The long-term anaerobic corrosion rate of cast iron is approximately five times lower than for carbon steel in the same solution, at 50°C.
- The long-term anaerobic corrosion rates of both cast iron and carbon steel are lower in groundwaters at higher pH, due to a more stable oxide film.
- In both aerated and deaerated conditions, physical damage to the surface has only a short transient effect on the corrosion rate. The rate of film reformation does not depend on the presence of dissolved oxygen, because the oxygen required to form the film is provided by direct reaction between bare steel and the water. If a passive Fe_2O_3 film exists before anoxic conditions develop, the film can undergo a reduction process to provide the current required to regenerate a magnetite film.”

Studies of the effects of γ -radiation (Smart and Rance 2005) and galvanic coupling between Cu and cast iron (Smart et al. 2005) have also been reported.

Recent studies of the effect of expansive iron corrosion products suggest that little if any effect on the structural integrity of an outer Cu corrosion barrier can be expected (Smart et al. 2006). Magnetite corrosion products were found to be easily deformed and more compliant than similar oxides formed at high temperature. So-called “mini-can” experiments are being conducted at the Äspö Hard Rock Laboratory to further investigate the consequences of the anaerobic corrosion of cast iron on the outer Cu corrosion barrier (N. Smart, unpublished work, 2007).

3.2.3 Summary of Current Knowledge of Carbon Steel UFC Corrosion

This Section provides a summary of the state-of-the-art knowledge of C-steel UFC corrosion. For more-detailed discussions see the reviews by Andra (2001); JNC (2000); Johnson and King (2003); and King (2005b, 2007).

Uniform Corrosion

There is a large amount of information on the uniform corrosion of C-steel, both from nuclear waste management studies and from the broader corrosion literature. The nuclear waste management literature includes a relatively consistent set of data, albeit with wide variations in the reported corrosion rate largely resulting from the wide range of experimental conditions employed (King and Stroes-Gascoyne 2000). Typically, the extent of uniform corrosion is assessed empirically and is based on a mass-balance calculation for the aerobic phase and a constant corrosion rate for the anaerobic period. Such a relatively simple methodology is acceptable provided there is a sufficient underlying mechanistic understanding of the corrosion processes to justify the prediction.

Specific areas of adequate knowledge include:

1. uniform corrosion of C-steel in compacted buffer material under aerobic conditions,
2. range of uniform corrosion rates of C-steel under anaerobic conditions,
3. cathodic reduction of O₂ on C-steel and its corrosion products, and
4. anodic dissolution of Fe in Cl⁻-dominated environments.

Localized Corrosion

Localized corrosion of steel UFC is typically treated using a simple empirical pitting factor derived from suitable laboratory or field data. There is evidence that the degree of localization (i.e., the value of the pitting factor) decreases with increasing depth of corrosion (JNC 2000; King and Johnson 2003; King 2005b, 2007), which may also reflect a tendency for the localized corrosion to stifle.

Available databases and statistical models for the localized corrosion of C-steel include:

1. empirical database of pit-depth data (Romanoff 1989),
2. depth-dependent pitting factor (JNC 2000, Johnson and King 2003), and
3. extreme-value analyses of pit-depth data (Akashi et al. 1990, Asano et al. 1992).

Hydrogen-related Effects

The methodology for determining whether the material selected for C-steel UFC is susceptible to H-related degradation mechanisms is well established (JNC 2000, Johnson and King 2003, King 2007). As defined by Equation (1), H-related degradation only occurs if the sub-surface absorbed [H] (C_H^0) is greater than the threshold value ($(C_H^0)_{TH}$) for damage. The value of $(C_H^0)_{TH}$ is temperature, material, and damage-mode dependent.

There is existing knowledge of both c_H^0 and $(c_H^0)_{TH}$ values for older ferrite-pearlite steels for various hydrogen degradation mechanisms, although the equivalent database for modern, more-resistant alloys is limited.

Stress Corrosion Cracking

Various arguments have been used for excluding the possibility of SCC of steel containers, including:

1. the absence of suitable environment (in the case of cracking in caustic, phosphate, nitrate, CO-CO₂, high-temperature water, and concentrated HCO₃⁻/CO₃²⁻ environments),
2. the absence of cyclic loading (for dilute and concentrated HCO₃⁻/CO₃²⁻ environments), and
3. the assumption of adequate post-weld stress relief (all environments).

The evidence for these arguments comes from the substantial database on the SCC of ferrite-pearlite pipeline steels in dilute and concentrated HCO₃⁻/CO₃²⁻ environments under cyclic load conditions.

Microbiologically Influenced Corrosion

The existing state-of-the-art for the MIC of C-steel UFC is based on the same arguments regarding the likely absence of microbial activity made for Cu UFC, described above in detail.

Preferential Weld Corrosion

Conventional arc welding processes are likely to be employed for fabricating the steel container. These welding processes will result in the development of hardened microstructures in the heat-affected zone, which will be more susceptible to SCC than the base metal. The welding processes will also cause the development of residual stresses in the welded area. Elevated-temperature post-weld heat treatment can be used to temper and reduce the hardness of the heat-affect zone, which will also reduce residual stresses and the hydrogen content in the weld metal and heat-affected zone for all the container welds (except perhaps for the final closure weld). However, if PWHT is not permitted due to the concern over thermal damage to the used fuel, the temper bead welding technique can be potentially used to produce a tempered heat-affected zone with acceptable hardness without the need for PWHT at high temperatures. Other types of stress relief processes can be used to reduce the residual stresses of the final closure weld.

Corrosion During the Unsaturated Period

As for Cu UFC, C-steel containers will be subject to corrosion during the unsaturated period in the evolution of the repository environment. The same principals are involved for C-steel as for copper.

Hydrogen Gas Issues

The transport of H₂ away from the container surface and the impact on the buffer and backfill sealing materials and host rock is perhaps the most important issue for C-steel containers. Much work on this topic has been performed by Nagra, particularly in the area of H₂ transport mechanisms. The properties of the geosphere and, possibly, the near-field may be sufficiently similar to those expected in a possible DGR in Ordovician sedimentary deposits that much of the information developed in the Swiss program can be adapted for a Canadian DGR.

There is a good understanding of gas transport through sedimentary deposits (at least for Opalinus Clay) and through bentonite (Nagra 2002).

Differences Between Crystalline and Sedimentary Host Rocks

The major differences between crystalline and sedimentary host rock formations from a corrosion viewpoint are (i) the potential higher salinity, especially [Cl⁻], for Ordovician sedimentary deposits, and (ii) the potential lower hydraulic conductivity of sedimentary deposits, possibly resulting in longer re-saturation times and a longer period of unsaturated conditions.

Conceptually, the effects of a longer re-saturation time are understood in terms of the processes determining when the UFC surface will become wetted and corrosion occur (King 2006).

Lifetime Predictions

To date, lifetime predictions for C-steel UFC have been based on an empirical damage prediction supported by mechanistic understanding (JNC 2000, Johnson and King 2003, King 2005b, Marsh and Taylor 1988). This approach has been applied to the estimation of the extent of aerobic and anaerobic corrosion and of the maximum depth of localized corrosion. Reasoned arguments have been used to exclude other possible corrosion processes, such as SCC, MIC, and hydrogen-related effects.

Archaeological Artifacts

Archaeological artifacts do exist for C-steel and use has been made of some of these studies (e.g., Crossland 2005, McMurry et al. 2001). Both the French and Japanese nuclear waste management programs are investing a lot of effort in the study of archaeological artifacts. The French program uses evidence from artifacts to affirm the long-term corrosion mechanism(s) (Neff et al. 2006), whereas the Japanese program is also using these studies to provide estimates of long-term corrosion rates (Yoshikawa et al. 2007).

4. KNOWLEDGE GAPS AND RESEARCH AREAS

At the NWMO corrosion workshop, a large number of topics were considered, both those for which there was deemed to be sufficient information and those where further work is required.

4.1 Knowledge Gaps and Issues

Bases on the summary of the state of the existing knowledge of UFC corrosion, there is a good understanding of the long-term corrosion behaviour of copper and C-steel containers. However, a number of areas requiring further study are apparent from the above discussion and are summarized here. Also included here are additional issues and gaps identified during the NWMO corrosion workshop.

This Section is a summary of only those topics that were identified as gaps in the various areas considered at the corrosion workshop. Prioritization of these issues for future study and consideration and the grouping of the gaps into research areas are discussed in the next Section.

4.1.1 Copper UFC

Uniform Corrosion

Although there is a good understanding of the uniform corrosion behaviour of copper UFC, there are some areas of remaining uncertainty. Most importantly, there is a need to validate the mixed-potential model predictions, especially since these predictions form the basis of the Copper Corrosion Model and the prediction of the possibility of SCC, MIC, and localized corrosion, in addition to uniform corrosion. Validation would not only require a database of measured E_{CORR} values under various conditions (some of which already exist), but also determination or confirmation of various parameters and processes included in the model.

In addition to validation of the mixed-potential model, a number of other areas of uncertainty were identified, including the corrosion behaviour of copper in unsaturated bentonite and the thermodynamic stability of copper in O_2 -free concentrated Cl^- solutions.

Areas in which the current state of knowledge is limited include:

1. rates of precipitation and dissolution of Cu_2O , $CuCl_2 \cdot 3Cu(OH)_2$, and other Cu corrosion products,
2. limited E_{CORR} data in compacted bentonite systems for validation of mixed-potential models,
3. limited large-scale test data for validation of CCM predictions,
4. corrosion of Cu in unsaturated bentonite,
5. rate of Cu(II) reduction on corroded Cu surfaces,
6. availability of Fe(II) in buffer and backfill pore water for redox reactions with O_2 ,
7. properties of oxide films,
8. thermodynamic stability of Cu in high $[Cl^-]$,

9. effect of film formation on E_{CORR} , and
10. measurement of the interfacial pH.

Localized Corrosion

The major challenges in the area of the localized corrosion of copper UFC are (i) gaining acceptance that the form of corrosion observed is a type of surface roughening rather than discrete localized corrosion or pitting, and (ii) developing a suitable model for making long-term predictions of the observed phenomena. Corrosion in simulated disposal environments takes the form of roughened uniform corrosion in which all areas of the surface are corroded, but to differing degrees. This form of corrosion is characterized by the non-permanent spatial separation of anodic and cathodic processes. Over time, this form of corrosion is expected to result in surface roughening, rather than discrete localized attack.

It is now recognized that spatial separation of anodic and cathodic processes (possibly leading to localized attack) is possible during the initial saturation phase in the evolution of the repository environment. Non-uniform wetting of the surface due to deliquescence of surface impurities could result in temporary localized attack. Although a conceptual model for this form of corrosion has been developed (King 2006), there is no experimental database to support this model.

Specific areas of uncertainty include:

1. the mechanism of the corrosion of Cu in unsaturated bentonite and the possibility of localized corrosion
2. evolution of the surface roughness from the initial aerobic phase to the long-term anaerobic period
3. model for predicting surface roughening and how it evolves with time, and
4. insufficient validation of localized corrosion predictions

Stress Corrosion Cracking

Although a substantial amount of work has been done on the SCC behaviour of copper UFC, there are a number of gaps in our existing knowledge. These areas are listed below, in no particular order:

1. incomplete mechanistic understanding of SCC of Cu under repository conditions,
2. inadequate understanding of the apparent inhibitive effects of Cl^- , particularly under highly saline environments for a DGR in sedimentary rock,
3. insufficient bulk potential and pH data for SCC in acetate and ammonia environments,
4. uncertainty whether SCC could occur under anaerobic conditions,
5. dependence of SCC on loading conditions,
6. lack of validation of predicted interfacial pH values from CCM-SCC code,
7. assessment of effectiveness of non-thermal, post-weld stress relief treatments,
8. effect of phosphorus on SCC behaviour,
9. effect of material properties (e.g., degree of cold work, yield strength, grain size, etc.), and
10. the properties of oxide films and their role in SCC.

Microbiologically Influenced Corrosion

The current approach to predicting the extent of MIC is to determine under what conditions microbial activity is, and is not, possible during the evolution of the repository environment. Significant progress has been made in determining the ranges of bentonite density, swelling pressure, and water activity that support microbial activity. It appears unlikely that extensive microbial activity can occur in the near-field, but there is remaining uncertainty about (i) the possibility of some activity during the transitional stage prior to the full development of the bentonite swelling pressure and (ii) whether it will ever be possible to categorically state that microbial activity cannot occur in the near field at any time.

An alternative approach to assessing the threat from MIC is to estimate the maximum damage that could occur if microbial activity near the container is possible. The extent of MIC could be limited by a number of factors, including the availability of nutrients or electron acceptors. Estimates made on mass-balance arguments could be used to bound the maximum possible extent of MIC, should it prove impossible to categorically exclude the possibility of microbial activity at some stage during the container service life.

The following studies would support the argument that microbial activity will not occur in the near field or at the container surface:

1. identification of the physiological mechanism(s) for the suppression of microbial activity in compacted bentonite,
2. the development of engineering confidence that criteria for the suppression of microbial activity can be guaranteed in the repository (e.g., at interfaces between blocks of compacted bentonite), and
3. development of experimental and modelling evidence that microbial activity can be suppressed during the transition phase as the bentonite saturates.

The following studies would support the approach of determining the maximum amount of damage by MIC if near-field microbial activity occurs:

1. prediction of the maximum amount of metabolic by-products that could be formed,
2. mass-balance calculations to predict the extent of MIC for various assumed limiting processes,
3. sensitivity analyses using the CCM-MIC to determine the maximum amount of damage under various scenarios,
4. development of a biofilm MIC model, and
5. targeted experimental studies under realistic exposure conditions.

Preferential Weld Corrosion

The current knowledge of the effects of different welding procedures on the subsequent corrosion behaviour of the UFC is limited. Further work on welded OFP Cu material is warranted, including the possibility of enhanced uniform corrosion, the susceptibility to localized attack, and further study of the susceptibility to SCC.

Corrosion During the Unsaturated Period

A conceptual understanding of the processes leading to the initial and subsequent wetting of the UFC surface during the saturation phase has recently been developed (King 2006). Wetting is likely to be non-uniform initially as deliquescence of surface contaminants occurs as the container cools and the relative humidity increases. Since conditions will be aerobic during this period, the possibility of spatial separation of anodic and cathodic processes cannot be excluded. Corrosion should become more uniform as the surface progressively wets.

Although the conceptual understanding is in place, no experimental data have been determined to confirm the proposed model. Therefore, the following gaps have been identified:

1. experimental evidence for the degree of localization of corrosion during the unsaturated phase,
2. experimental evidence for the nature of the surface contaminants on Cu UFC and their deliquescence behaviour,
3. model for assessing the extent of damage during the unsaturated phase, and
4. an estimate of the time-surface temperature-relative humidity relationship during the unsaturated phase.

Hydrogen Gas Issues

The issue of gas generation from a failed copper UFC (with C-steel insert) is generally of secondary importance, especially compared with the magnitude of the issue for a C-steel UFC. Scoping analyses performed for the Third Case Study showed that gas generation from one failed container has a negligible effect on the safety of the repository (Gierszewski et al. 2004).

Although perceived to be of relatively low priority (see below), the following gaps exist in our current understanding:

1. an assessment of the rate of H₂ generation from C-steel corrosion for a defected Cu outer corrosion barrier
2. an assessment of the range of defect sizes in the Cu corrosion barrier

Differences Between Crystalline and Sedimentary Host Rocks

The major differences in the environmental conditions for the two potential host rocks are (i) the potential higher salinity, especially [Cl⁻], for Ordovician sedimentary deposits, and (ii) the potential lower hydraulic conductivity of sedimentary deposits, which may result in longer re-saturation times and a longer period of unsaturated conditions. In principal, the effects of these two factors on the subsequent corrosion behaviour are known, or can be inferred from our existing knowledge. However, where areas in which our current understanding could be improved include:

1. estimation of the duration of, and the surface temperature-relative humidity relationship during, the unsaturated phase, and
2. inadequate understanding of the apparent inhibitive effects of Cl⁻ on the SCC of Cu.

Lifetime Predictions

The methodology for predicting the long-term corrosion performance of copper UFC (based on the CCM suite of models) is relatively mature. There are, however, a number of areas where these models could be improved and/or extended, including:

1. the effect of pore-water species other than Cl^- (e.g., SO_4^{2-} , $\text{CO}_3^{2-}/\text{HCO}_3^-$) on speciation of Cu corrosion products,
2. multi-dimensional reactive-transport modelling,
3. validation of reactive-transport models against laboratory or large-scale experimental data,
4. development of a corrosion model for unsaturated conditions,
5. behaviour of UFC following penetration of the Cu corrosion barrier (i.e., time dependence of the fractional failed area), and
6. assessment of the maximum allowable temperature for Cu UFC from a corrosion viewpoint.

Natural and Archaeological Analogues

The study of natural analogues and archaeological artifacts provides a useful validation of long-term predictions of the corrosion behaviour of UFC. This is particularly so for copper containers since there are abundant examples available and they provide credible evidence for the longevity of copper-based materials in natural environments. A number of such studies have been performed, as noted above.

At this time, there does not appear to be a specific need to pursue further such studies. However, the option of additional studies should be retained in case an interesting analogue is discovered in the future.

4.1.2 Carbon Steel UFC

The possible use of C-steel UFC has recently been considered in the Canadian R&D program, although there has been little work on the corrosion behaviour under conditions expected in a Canadian repository. However, much of the information developed in other national programs is expected to be transferable to the Canadian context. The gaps in our current understanding identified below take into account this existing information from other countries.

Uniform Corrosion

There have been a number of studies of the uniform corrosion of steel under simulated repository conditions, with a wide range of reported corrosion rates (King and Stroes-Gascoyne 2000; King 2005b, 2007). Corrosion rates under aerobic conditions are typically of the order of several 10's of $\mu\text{m}\cdot\text{a}^{-1}$, with anaerobic rates in the range $0.1\text{-}10\ \mu\text{m}\cdot\text{a}^{-1}$. In addition to determining the rate of corrosion, it is also necessary to develop an understanding of the corrosion mechanism, since justification of long-term predictions requires that the processes controlling the rate of corrosion be understood.

Based on these needs, a number of gaps in the application of the existing knowledge to a repository in Canada can be identified, namely:

1. aerobic corrosion rate(s) of C-steel under anticipated “Canadian” conditions in a deep geological repository,
2. anaerobic corrosion rate(s) of C-steel in a “Canadian” DGR under unsaturated conditions,
3. anaerobic corrosion rate(s) of C-steel in a “Canadian” DGR under saturated conditions,
4. kinetics of H₂O reduction on C-steel and its corrosion products, especially Fe₃O₄,
5. effect of film formation on the anodic dissolution kinetics,
6. mechanism of the long-term anaerobic corrosion of C-steel,
7. understanding the transition in corrosion behaviour between aerobic and anaerobic conditions,
8. mixed-potential model for the prediction of E_{CORR} of C-steel containers, including the effects of film formation and transformation and the evolution of the repository environment,
9. effect of high [Cl⁻] on aerobic and anaerobic corrosion rates of C-steel,
10. effect of H₂ partial pressure on the rate of anaerobic corrosion, E_{CORR}, and the H₂ evolution reaction,
11. effect of CO₂ partial pressure on the anaerobic corrosion rate of C-steel,
12. effect of HCO₃⁻/CO₃²⁻ on passivation,
13. alteration of bentonite by reaction with Fe(II),
14. mechanism of passivation in compacted bentonite, and
15. effect of material composition on passivation.

Localized Corrosion

The “uniform” corrosion of C-steel generally exhibits a certain degree of unevenness, ranging from discrete pits to surface roughening (King 2005b, 2007). The available evidence suggests that the degree of localization of the corrosion diminishes with either increasing time or increasing depth of corrosion. Localized corrosion is most likely during the initial aerobic phase, both because of the possibility of passivation of the surface and because of the initial non-uniform wetting of the surface during saturation of the bentonite. Regardless, the consequence is that the allowance that must be made for localization of the corrosion in lifetime assessments decreases with time. This is an important conclusion and one that requires a full understanding since it has a significant impact on how UFC lifetimes are predicted.

Given this background knowledge, the gaps in the existing knowledge include:

1. the degree of surface roughness associated with the uniform corrosion of C-steel in compacted bentonite,
2. whether a C-steel UFC will passivate under repository conditions and, if so, the duration of passivity,
3. mechanistic understanding of the depth-dependent pitting factor and/or stifling mechanism of localized corrosion of C-steel,
4. effect of unsaturated conditions on the initiation of localized corrosion,
5. localized corrosion due to reductive dissolution of Fe(III) films during the aerobic-anaerobic transition, and
6. assessment of the extent of localized corrosion of C-steel UFC under Canadian conditions.

Hydrogen-related Effects

The susceptibility of steels to hydrogen-related degradation mechanisms increases with increasing yield strength of the material (Figure 8). Recent improvements of the resistance of steels to hydrogen effects, especially in the oil and gas industry, should minimize the impact of this integrity threat to the UFC. There is a large database of existing knowledge of hydrogen effects for older, “dirtier” ferrite-pearlite C-steels, but less for modern micro-alloyed steels designed for use in sour service.

Information is required on hydrogen-related effects on modern steels to help guide material specification for a C-steel UFC. Gaps in the existing knowledge include:

1. measurement of sub-surface hydrogen concentrations (c_H^0) for candidate UFC steels under repository conditions,
2. measurement of threshold hydrogen concentrations ($(c_H^0)_{TH}$) for candidate UFC steels for various hydrogen degradation mechanisms,
3. assessment of the probability of H-related degradation mechanisms for C-steel UFC, and
4. the effect of welding on the susceptibility to hydrogen embrittlement.

Stress Corrosion Cracking

There is a large database of SCC data for C-steels, particularly for pipeline steels in HCO_3^-/CO_3^{2-} environments (King 2005b, 2007). Although some of this information is transferable to the conditions that are expected in a DGR, there is a lack of information for more modern steels representative of those from which a UFC might be fabricated. In addition, environments to which pipelines may be exposed differ from those expected in the repository in a number of significant ways, not least the absence of cyclic stress for the latter.

There are a number of methods for minimizing the residual stress on the container, and hence the possibility of SCC. Firstly, welding procedures can be optimized, such as the use of the temper bead welding technique to produce a tempered heat-affected zone with acceptable hardness. Secondly, residual stresses can be minimized by post-weld stress relief by shot peening (Eadie et al. 2002) or by laser-peening or low-plasticity burnishing (BSC 2004). It would seem to be sound design practice to follow these routes in order to reduce or eliminate the threat from SCC.

A number of gaps in the existing knowledge can be identified, including:

1. assessment of welding procedures to minimize residual stress,
2. assessment of non-thermal, post-weld stress relief treatments,
3. assessment of whether near-neutral pH SCC conditions will develop at the UFC surface in the repository,
4. SCC behaviour of candidate UFC material(s) in dilute HCO_3^-/CO_3^{2-} environments under cyclic and static load, and
5. SCC of candidate UFC material(s) under a range of repository conditions.

Microbiologically Influenced Corrosion

The treatment of MIC of C-steel UFC follows a similar argument to that for copper UFC, namely: (i) demonstrating that microbial activity is unlikely at the container surface and in the near-field and (ii) even in the unlikely event that MIC does occur, demonstrating that the extent of corrosion is limited. Thus, many of the studies proposed to address MIC apply equally to both UFC materials.

The only additional studies specific to C-steel are (i) the effect of low levels of HS^- on the corrosion behaviour (uniform and localized corrosion, H absorption, and SCC) and (ii) alteration of bentonite by reaction with Fe(II) limiting the swelling pressure and the effects on microbial culturability (this effect will be offset to some degree in sedimentary host rock by the effect of saline pore waters on microbial culturability).

Preferential Weld Corrosion

At some stage in the development of C-steel UFC there will be a need to assess the corrosion behaviour of the container closure welds. There is currently limited knowledge in the Canadian program in the following areas:

1. demonstration of candidate welding procedures for fabricating a C-steel UFC and the fabrication of prototype weld material for laboratory testing,
2. uniform and localized corrosion behaviour of base, HAZ, and weld metal for all container welds including the final closure weld, and
3. susceptibility of base, HAZ, and weld metal to H-related degradation mechanisms and SCC for all container welds including the final closure weld.

Corrosion During the Unsaturated Period

Carbon steel is a possible container material for a DGR in sedimentary host rock. The period of saturation could be longer than that for a DGR in crystalline rock because of the lower hydraulic conductivity of sedimentary deposits. Therefore, it is important to consider the effects of unsaturated conditions on the corrosion behaviour of C-steel UFC.

The issues associated with corrosion of C-steel UFC during the unsaturated phase are similar to those defined above for copper containers. The unsaturated phase is a period when localized corrosion is possible because of the aerobic conditions (initially, at least) and because of the possibility of non-uniform wetting of the surface. Because of the potentially extended unsaturated phase in sedimentary deposits, it is necessary to also consider the rate and mechanism of the uniform corrosion of C-steel in unsaturated anaerobic conditions.

Hydrogen Gas Issues

The generation and transport of hydrogen are clearly major issues for a C-steel UFC. Issues related to the rate of H_2 generation are dealt with here. The equally important issues of how fast H_2 can be transported away from the container surface through the near field and geosphere should be addressed in other programs.

In the corrosion area, gaps in the existing knowledge include:

1. the benefits (and drawbacks) of reducing the rate of H₂ generation through small alloying additions, and
2. the long-term H₂ generation (or anaerobic corrosion) rate under Canadian conditions.

Differences Between Crystalline and Sedimentary Host Rocks

As noted above, the major differences in the environmental conditions for the two potential host rocks are (i) the potential higher salinity, especially [Cl⁻], for Ordovician sedimentary deposits, and (ii) the potential lower hydraulic conductivity of sedimentary deposits, possibly resulting in longer re-saturation times and a longer period of unsaturated conditions.

Gaps in our existing knowledge include:

1. an estimate of the time-surface temperature-relative humidity relationship during the unsaturated phase,
2. the effect of Cl⁻ on the potential for passivation and, if a passive film does exist, on passivity breakdown for C-steel UFC under repository conditions,
3. the effect of Cl⁻ on the anaerobic corrosion rate of C-steel, and
4. the effect of host rock properties on the mechanism(s) and rate of H₂ transport.

Lifetime Predictions

A number of lifetime estimates have been made for C-steel UFC, both in the Canadian program (King 2005b, 2007) and internationally (JNC 2000, Johnson and King 2003).

Two approaches can be taken to lifetime predictions: (i) simple extrapolations based on empirical data supported by a detailed understanding of the corrosion processes involved and/or (ii) detailed mathematical modelling based on the identified corrosion mechanism(s). In either case it is necessary to have a detailed mechanistic understanding of the processes involved.

Lifetime predictions to date have been based on simple extrapolations of empirical corrosion data. Gaps in our existing approaches include:

1. development of a mixed-potential model for C-steel UFC,
2. assessment of the extent of localized corrosion of C-steel UFC under Canadian conditions, including during the unsaturated period,
3. definition of (the range of) uniform corrosion rate(s) of C-steel under anaerobic conditions,
4. an assessment of the likelihood of H-related degradation mechanisms under Canadian conditions,
5. an assessment of the likelihood of SCC under Canadian conditions, and
6. assessment of the maximum allowable temperature for C-steel UFC from a corrosion viewpoint.

Archaeological Artifacts

As demonstrated by their use in the French (Neff et al. 2006) and Japanese (Yoshikawa et al. 2007) programs, archaeological artifacts can provide useful supporting evidence for predictions of the long-term corrosion behaviour of C-steel UFC.

No specific needs or gaps have been identified in the Canadian program, although such a study should be considered in future if a useful or interesting archaeological artifact study should be found.

4.2 Research Areas

Having identified the various gaps in the existing knowledge of the corrosion behaviour of copper and C-steel UFC, it is necessary to (i) group the gaps into proposed research areas or programs and (ii) prioritize both the individual project ideas or gaps and the larger research areas.

Grouping of the gaps can be done in a number of ways. The process used here to identify the gaps was to consider different corrosion processes for each material (e.g., uniform corrosion, SCC, preferential weld attack, etc.), along with consideration of different phases and aspects of the DGR environment (e.g., the unsaturated phase, differences between crystalline and sedimentary host rocks). The gaps identified above are at the level of individual projects.

Research areas or programs could be defined based on different phases in the evolution of the repository environment (e.g., aerobic corrosion of copper and C-steel, corrosion of UFC materials during the unsaturated period, etc.), or by different corrosion processes (e.g., SCC of copper and C-steel, MIC of UFC materials, etc.), or material-specific (e.g., corrosion of copper welds, H₂ generation from C-steel, etc.).

Here, a hybrid approach has been taken, with three research areas defined that are common to both materials (MIC, corrosion during the unsaturated phase, and evolution of corrosion damage), with the remainder of the areas defined by specific corrosion processes for each material separately. This process results in the definition of fourteen research areas or programs. Further rationalization of these 14 areas could be performed by combining studies of a particular corrosion process for both materials into a single program (e.g., SCC of UFC materials, lifetime prediction models, corrosion of welds, etc.).

The second task is to prioritize individual projects within a research area and the different research areas themselves. At the NWMO UFC corrosion workshop in June 2007, projects and programs were prioritized based on their "impact" and "achievability"

The impact of a research project depends on a number of factors, including:

- What are the consequences of not having this information?
- Is this a "must-have" piece of information?
- Is this information needed for other, linked studies?
- Is there currently sufficient understanding?
- How urgently is the information needed?

Similarly, the achievability of a project depends on:

- How much effort is involve (funds, resources, duration)?
- Is there a long lead time or can it be done at any time?
- Are the necessary skills available (in Canada, at all)?
- What is the probability of success (speculative idea, new or established techniques, reliant on other developments, extension of existing knowledge)?
- Is the project complex (many partners, inter-dependent tasks)?
- Are the project objectives/deliverables well defined?

Research projects were classified as either “high” or “low” impact and either “high” or “low” achievability. Clearly, projects that are both “high” impact and “high” achievability are a higher priority than projects that are rated “low” in both categories.

Prioritization of future studies was based on one of two criteria:

1. the need to demonstrate sufficient scientific and engineering understanding of a given corrosion phenomenon, and
2. the need to develop DGR designs for the program over the next 2-3 years.

4.2.1 Common Research Areas

Microbiologically Influenced Corrosion

The two aims of the MIC program are:

1. To demonstrate the range of repository environmental conditions under which microbes can be reasonably expected to be non-culturable (inactive).
2. To determine the nature and maximum extent of corrosion damage to the UFC if microbes were to be active in the DGR.

The first aim is a continuation of an effort over the past 5-10 years to try to demonstrate that microbial activity will not occur close to the UFC surface because of the inhospitable nature of the environment. Factors such as the elevated temperature, low water activity, and high bentonite swelling pressure are expected to severely restrict the extent of microbial activity and, possibly, prevent any activity (and, hence, the possibility of biofilm formation) at the container surface.

It may be impossible to categorically exclude the possibility of some microbial activity close to the UFC. Consequently, the second aim of the MIC research area is to estimate the maximum amount of damage that could result if microbes are indeed active. This could involve both modelling and experimental studies. Modelling studies could involve simple mass-balance approaches or more-detailed predictions based on sensitivity analyses using the CCM-MIC code. In addition, recent advances in the development of modelling procedures for biofilm models may make it feasible to predict the consequences of microbial activity at the UFC surface. As an alternative to modelling approaches, the nature and extent of damage could be estimated experimentally. However, any such experimental program must be carefully defined and be performed under realistic environmental conditions. Great care must be exercised in attempting to accelerate such tests in order to simulate long exposure times.

A number of gaps were identified for each of these aims (Table 4). The nature of the gap, the rationale for why this information is needed, and the priority assigned to the gap are summarized below. Because many of these issues apply equally to copper and C-steel UFC, a single MIC program would address both materials.

For the program aimed at demonstrating that microbial activity will not occur in the repository, the gaps in the current knowledge are (Table 4(a)):

Gap: Can biofilm formation on the container surface be prevented through the use of highly compacted bentonite?

Rationale: The current work focussing on the effects of water activity and bentonite swelling pressure on microbial activity needs to be completed.

Impact/achievability: High/low

Priority: Medium - a significant amount of effort has already gone into this topic, although some additional effort is required to complete the work.

Gap: What is the physiological mechanism(s) for the suppression of microbial activity in compacted bentonite?

Rationale: A physiological explanation for the apparent effect of high bentonite swelling pressure and/or low water activity is required to support the argument for the absence of microbial activity in compacted bentonite.

Impact/achievability: High/low

Priority: Medium – this understanding should come from on-going studies of the factors that limit microbial activity in the repository.

Gap: Need to establish engineering confidence that criteria for the suppression of microbial activity can be guaranteed in the repository.

Rationale: Need to demonstrate that microbial activity is suppressed at all times and at all locations within the repository, especially at interfaces and especially during the saturation phase.

Impact/achievability: High/low

Priority: High – this represents a significant uncertainty in the current understanding of the time evolution of microbial activity in the DGR.

Gap: Can an upper temperature limit for microbial activity in the repository be established?

Rationale: It is likely that high temperature (>100°C?) will kill microbes and could produce a microbial-depleted zone around the UFC. This would be a strong argument in support of the current model that no microbial activity will occur in the near field, provided the necessary temperature can be attained in the repository without deleterious effects on the container or bentonite.

Impact/achievability: High/low

Priority: Medium – useful approach if bentonite can tolerate higher temperatures.

For the program aimed at bounding the maximum extent of damage if microbial activity were to occur in the repository, the gaps in the current knowledge are (Table 4(b)):

- Gap:* Prediction of maximum amount of metabolic by-products that can be formed if microbial activity at container surface is possible.
- Rationale:* An estimation of the maximum amount of aggressive metabolic by-products is required in the event that microbial activity is possible in the near field. Different species would be considered for the two different container materials.
- Impact/achievability:* High/high
- Priority:* High – needed to support the approach of bounding the extent of MIC.
-
- Gap:* Mass-balance calculations to predict the amount of damage due to an assumed limiting factor or process (e.g., organic C, electron acceptor)
- Rationale:* Need to determine the corrosion consequences of the production of metabolic by products from microbial activity for both copper and C-steel UFC.
- Impact/achievability:* High/high
- Priority:* High - needed to support the approach of bounding the extent of MIC.
-
- Gap:* Sensitivity analyses with CCM-MIC to determine worst-case effect of assuming limitation by organic C, electron acceptor, etc.
- Rationale:* The CCM-MIC is an ideal tool for assessing the maximum amount of metabolic by-products for different assumptions regarding the limiting species/process.
- Impact/achievability:* High/high
- Priority:* High - needed to support approach of bounding extent of MIC.
-
- Gap:* Development of UFC biofilm model
- Rationale:* Recent developments in the modelling of biofilms now makes it possible to develop biofilm MIC models to predict the extent of attack and the degree of localization.
- Impact/achievability:* H/L
- Priority:* Medium – biofilm modelling is in its infancy and it may be difficult to justify and/or validate predictions.
-
- Gap:* Specific Cu/Fe corrosion experiments to assess extent of damage under realistic exposure conditions
- Rationale:* To support predictions of the maximum damage that could occur, well-planned, targeted experiments under realistic exposure conditions should be considered.
- Impact/achievability:* High/high
- Priority:* High – will be needed to support arguments based on bounding microbial activity, but care should be exercised in planning of experiments.

Corrosion During the Unsaturated Period

The aim of this program is to assess the importance of the unsaturated period in terms of the extent and nature of corrosion damage, in particular the extent to which corrosion is localized as a result of non-uniform wetting of the container surface.

Based partly on a report by King (2006), there is now a realization that the period of unsaturated conditions could be more significant than previously thought. At a typical atmospheric corrosion

rate of $1\text{-}2\ \mu\text{m}\cdot\text{a}^{-1}$, virtually all of the initially trapped O_2 in the repository would be consumed during an unsaturated phase that lasted of the order of 100 a. Indeed, if the re-saturation period were longer than this, there could be an extended period of anaerobic corrosion under unsaturated conditions.

An uncertainty for the unsaturated phase is the degree to which non-uniform wetting of the UFC surface will induce localized corrosion.

There are a number of issues that need to be resolved to develop an adequate understanding of the nature and extent of damage during this period. Table 5 defines a number of gaps, many of which apply equally to copper and C-steel UFC.

Gap: *Experimental evidence for the nature of the surface contaminants on copper and C-steel UFC and their deliquescence behaviour.*

Rationale: The wetting of the UFC surface, both in terms of the time of wetting and the distribution of surface moisture, will be determined by the distribution and deliquescence behaviour of surface contaminants.

Impact/achievability: High/high

Priority: High – need to know what surface contaminants are present before assessing the consequences.

Gap: *Experimental evidence for the degree of localization of corrosion of copper UFC during the unsaturated phase.*

Rationale: Initial wetting of the UFC surface is likely to be non-uniform and may result in localized corrosion of the container.

Impact/achievability: High/high

Priority: High – need to understand extent of issue for copper UFC.

Gap: *Experimental evidence for the degree of localization of corrosion of C-steel UFC during the unsaturated phase.*

Rationale: Initial wetting of the UFC surface is likely to be non-uniform and may result in localized corrosion of the container.

Impact/achievability: High/high

Priority: High - need to understand extent of issue for C-steel UFC.

Gap: *Range of uniform corrosion rates of C-steel under anaerobic unsaturated conditions.*

Rationale: In sedimentary host rock, it is likely that the DGR will become anaerobic before it fully saturates. Consequently, there may be a period of anaerobic corrosion under unsaturated conditions, for which there are few reported corrosion rates.

Impact/achievability: High/high

Priority: High – there could be an extended period of anaerobic, unsaturated conditions in a DGR in sedimentary host rock.

Gap: Model for assessing the extent of damage of copper and C-steel UFC during the unsaturated phase.

Rationale: Corrosion during the unsaturated phase is not currently explicitly included in the lifetime predictions models for copper and C-steel containers, particularly the possibility of localized attack.

Impact/achievability: High/high

Priority: Medium – model development should await further experimental evidence.

Gap: Estimation of time-temperature-relative humidity relationship during the unsaturated phase.

Rationale: The time at which the surface first wets is determined, in part, by the time dependence of the relative humidity and temperature at the container surface.

Impact/achievability: High/high

Priority: Medium – this information will be needed for predicting extent of corrosion under unsaturated conditions.

Evolution of Corrosion Damage

The aim of this program is to describe how the nature of the corrosion damage evolves as the environment evolves from initially warm and aerobic to eventually cool and anaerobic. Specifically, the aim is to understand how localized damage that may occur initially evolves into more uniform corrosion over time.

For both copper and C-steel, localized attack of the surface is likely to take the form of non-uniform corrosion rather than the discrete pitting or crevice corrosion associated with passive materials. There will be a degree of surface roughening, but neither material is expected to undergo sustained localized attack. Localized attack may be initiated by non-uniform wetting of the surface during the initial unsaturated phase or by temporary spatial separation of anodic and cathodic reactions due to the precipitation of corrosion products or mineral salts on the surface.

Another characteristic of the early aerobic phase is the precipitation of oxidized corrosion products, such as $\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$ or various forms of $\text{FeOOH} \cdot x\text{H}_2\text{O}$. These Cu(II) and Fe(III) species are thermodynamically unstable under anaerobic conditions, implying that reductive dissolution could occur during the aerobic-anaerobic transition. Reductive dissolution processes can also lead to localized corrosion (King 2006), although the period of localized attack is often limited.

There is a need, therefore, for a program that describes how the corrosion front evolves with time as the environmental conditions evolve. Such a program should address issues such as the time-dependent localization of corrosion, the associated mechanisms, and how the surface profile evolves with time.

Many of these mechanisms, and the gaps in the current understanding, are common to both materials (Table 6).

Gap: Evolution of the surface roughness of Cu from the initial aerobic phase to the long-term anoxic period.

- Rationale: Corrosion of copper in compacted bentonite is known to exhibit surface roughening, but how that roughness evolves over time is uncertain.
Impact/achievability: High/high
Priority: High – needed to demonstrate understanding of corrosion mechanism.
- Gap: Model for predicting surface roughening of Cu and how it evolves with time.*
- Rationale: A mechanistically based model for predicting surface roughening of copper is needed.
Impact/achievability: High/high
Priority: High - needed to demonstrate understanding of corrosion mechanism.
- Gap: Degree of surface roughness associated with the uniform corrosion of C-steel in compacted bentonite.*
- Rationale: The degree of surface roughening of C-steel in contact with bentonite is uncertain.
Impact/achievability: High/high
Priority: High – experimental data not currently available for C-steel.
- Gap: Whether a C-steel UFC will passivate under repository conditions and, if so, the duration of passivity.*
- Rationale: Localized corrosion of C-steel is more likely if the surface passivates under repository conditions. Japanese evidence suggests that passivation is unlikely, but the potential for an extended unsaturated phase in a DGR in sedimentary host rock makes passivation more likely.
Impact/achievability: High/high
Priority: Medium – passivity appears unlikely, but issue should be resolved at some stage.
- Gap: Mechanistic understanding of the depth-dependent pitting factor and/or stifling of localized corrosion of C-steel.*
- Rationale: Currently, localized attack on C-steel is predicted based on a time- (or depth-) dependent pitting factor. The empirical data suggest that localized corrosion stifles, but the mechanism is uncertain.
Impact/achievability: High/high
Priority: Medium – need mechanistic understanding for empirical observation in order to justify long-term extrapolation.
- Gap: Database of film breakdown and re-passivation potentials for C-steel under repository conditions.*
- Rationale: An alternative approach to predicting the localized corrosion of C-steel is through comparison of the corrosion potential E_{CORR} to pitting or re-passivation potentials. A database of such potentials is required.
Impact/achievability: Low/high
Priority: Low – prediction based on comparison of E_{CORR} and pitting or re-passivation potentials is not considered the most appropriate approach for predicting localized corrosion of C-steel.
- Gap: Localized corrosion due to reductive dissolution of Fe(III) films during the aerobic-anaerobic transition.*

Rationale: During the evolution of the environmental conditions in the repository, previously formed Fe(III) corrosion products will be reduced to the Fe(II) state, a process that may induce localized corrosion.

Impact/achievability: High/high

Priority: Medium – some information available from analogous systems, but no direct evidence under DGR conditions.

4.2.2 Copper UFC Research Areas

Stress Corrosion Cracking of Copper

The aim of the copper SCC program is to define under what conditions a UFC might be susceptible to cracking and to determine the impact on container lifetimes. A list of gaps is given in Table 7.

Gap: Mechanistic understanding of SCC of Cu under repository conditions.

Rationale: The nature of the processes controlling crack initiation and growth need to be determined to justify long-term predictions.

Impact/achievability: High/high

Priority: Medium – mechanistic understanding should come from continuing experimental efforts.

Gap: Understanding of the apparent inhibitive effect(s) of Cl⁻.

Rationale: Experimental evidence is available indicating that Cl⁻ ions inhibit the SCC of copper in nitrite, ammonia, and acetate environments, but the precise mechanism is uncertain. This information is required to support long-term SCC predictions.

Impact/achievability: High/high

Priority: Medium – some evidence currently available, but mechanistic understanding still required.

Gap: Insufficient bulk potential and pH data for SCC in acetate and ammonia environments.

Rationale: A more-extensive database of potentials and pH values for the SCC of copper in acetate and ammonia is needed to support the current modelling approach based on a threshold potential and pH for cracking.

Impact/achievability: High/low

Priority: Medium – requires a large experimental effort.

Gap: Uncertainty whether SCC could occur under anaerobic conditions.

Rationale: Recent Japanese data suggest the possibility of SCC of copper under anaerobic conditions in the presence of sulphide. Although the concentration of sulphide in a Canadian DGR is likely to be lower than the reported threshold concentration, there remains the question of whether SCC can occur under the long-term anaerobic conditions.

Impact/achievability: High/low

Priority: Medium/low – would be significant if SCC can occur under anaerobic conditions, but relatively little sulphide expected in a Canadian repository.

Gap: Lack of validation of predicted interfacial pH values from CCM-SCC code.

Rationale: The current SCC modelling approach is based on a comparison of predicted potentials and pH values with a criterion based on the equilibrium E/pH for Cu₂O/CuO. The predicted pH values need validation against experimental measurements.

Impact/achievability: High/high

Priority: Medium – evidence required at some stage to validate CCM-SCC predictions.

Gap: *Assessment of effectiveness of non-thermal, post-weld stress relief treatments.*

Rationale: Sound engineering practice dictates that the residual stress of the final closure weld be minimized to limit the probability of SCC. The ability of various low-temperature methods for reducing the residual stress should be assessed.

Impact/achievability: High/low

Priority: Low – can be deferred until UFC design and fabrication procedure are defined.

Gap: *Effect of phosphorus on SCC behaviour.*

Rationale: The reference OFP Cu material contains 50-70 ppm P to improve the creep ductility. However, these levels of P have been shown to increase the susceptibility of Cu to SCC in ammonia environments.

Impact/achievability: Low/high

Priority: Medium – P content at low end of range shown to have effect on SCC susceptibility.

Gap: *Effect of Cu properties (degree of cold work, yield stress, grain size, etc.).*

Rationale: The degree of cold work, mechanical properties, and grain size are known to affect the SCC susceptibility of Cu. An understanding of these effects is required to address the impact of container fabrication processes and potential upset conditions (e.g., the accidental introduction of cold work due to impacts during handling and emplacement of the container).

Impact/achievability: Low/high

Priority: Medium – will need addressing at some later stage.

Gap: *Properties of oxide films and their role in SCC.*

Rationale: SCC of copper is observed under environmental conditions that support the formation of a surface film. The properties of these films need to be understood in order to support long-term predictions.

Impact/achievability: High/high

Priority: High – surface films play central role in SCC.

Corrosion of Copper Welds

The aim of this program is to determine the corrosion behaviour of welded regions on the container surface and to establish whether these areas are more susceptible to corrosion.

Relatively little work has been done on the corrosion behaviour of welded copper in the context of the UFC corrosion program and at some stage a focussed program will be required (Table 8).

Gap: Possibility of enhanced uniform corrosion, localized attack, and SCC of welds.

Rationale: Manufactured components frequently fail at weld locations. An assessment will be required of the possible enhanced susceptibility of welds on a copper UFC to uniform corrosion, localized attack, and SCC.

Impact/achievability: Low/high

Priority: Medium – gap should be addressed once closure weld technique has been defined.

Uniform Corrosion of Copper

In addition to investigating the mechanism of the uniform corrosion of copper, the aim of this program is to develop the necessary database to enable mixed-potential modelling of the corrosion potential of copper UFC. A sound mechanistic understanding is already in place, although some minor gaps have been identified (Table 9). Among the more-important gaps are validation of the CCM predictions and determining the effect of surface films on E_{CORR} .

Gap: Rates of precipitation and dissolution of Cu_2O , $CuCl_2 \cdot 3Cu(OH)_2$, and other Cu corrosion products.

Rationale: Precipitation of corrosion products is treated in the CCM codes, but reliable data for the rates of precipitation and dissolution are not available.

Impact/achievability: High/low

Priority: Medium/low – currently included in model, albeit with uncertain rate parameters.

Gap: Limited E_{CORR} data in compacted bentonite for validation of mixed-potential models.

Rationale: One of the main features of the CCM codes is the prediction of E_{CORR} . However, there are few E_{CORR} measurements in compacted bentonite against which to validate the model.

Impact/achievability: High/high

Priority: High – urgent need to validate CCM predictions.

Gap: Limited large-scale test data for validation of CCM predictions.

Rationale: The CCM code predicts the spatial and temporal distributions of various dissolved Cu species, as well as the corrosion rate and the potential for localized corrosion. There has been limited validation of the model. Data from large-scale tests are ideal for validation purposes.

Impact/achievability: High/low

Priority: High – need to validate CCM predictions.

Gap: *Corrosion of Cu in unsaturated bentonite.*
Rationale: If the surface of a copper UFC becomes uniformly wetted during the unsaturated phase before the complete consumption of O₂, there could be a period of uniform corrosion under unsaturated conditions. No experimental data are available to support predictions of the rate of corrosion under these conditions.

Impact/achievability: High/high
Priority: High – as noted above, need to address corrosion under unsaturated conditions.

Gap: *Extent of Cu(II) reduction on corroded Cu surfaces.*
Rationale: In most O₂-containing natural waters, the actual oxidant is Cu(II), produced by the homogeneous oxidation of Cu(I) by O₂, rather than O₂ itself. The interfacial reduction of Cu(II) is included in the CCM codes, but no direct measurements of the rate constant under simulated repository conditions have been made.

Impact/achievability: Low/high
Priority: Low/medium – currently included in model, albeit with uncertain rate parameters.

Gap: *Availability of Fe(II) in buffer and backfill pore water for redox reactions with O₂.*

Rationale: Part of the O₂ initially trapped in the repository is consumed by reaction with Fe(II) dissolved from mineral impurities. Dissolved Fe(II) may precipitate as a secondary phase if the concentration exceeds the solubility. Dissolution of Fe(II) is modelled based on biotite dissolution, but the validity of this treatment has not been demonstrated

Impact/achievability: Low/high
Priority: Low - currently included in model, albeit with uncertain rate parameters.

Gap: *Properties of oxide films.*
Rationale: Oxide films are known to form on copper under simulated disposal conditions, but film formation is only included in the CCM code as a sink for Cu(I). The possible effects of Cu₂O on the rate of interfacial processes, the probability for SCC, or for localized corrosion are not currently included in lifetime prediction models.

Impact/achievability: High/high
Priority: High – could have impact on E_{CORR}.

Gap: *Thermodynamic stability of Cu at high [Cl].*
Rationale: There are repeated suggestions that copper may be thermodynamically unstable with respect to H₂O in high salinity groundwaters. Despite various attempts by Posiva, no credible evidence for this claim has been obtained, but the question may arise again, especially if Cu is considered as a container material for the more highly saline groundwaters expected for DGR in sedimentary host rock.

Impact/achievability: Low/low
Priority: Low – evidence available from other programs.

Gap: *Effect of film formation on E_{CORR}.*

Rationale: Film formation typically results in the ennoblement of E_{CORR} . Although film formation is known to occur under simulated repository conditions, the effects of film formation on E_{CORR} are not currently included in the CCM codes.

Impact/achievability: High/high

Priority: High/medium – could have significant effect on E_{CORR} .

Gap: *Measurement of interfacial pH.*

Rationale: The CCM-SCC code includes the prediction of the interfacial pH. There are no experimental measurements of the interfacial pH of corroding Cu in contact with compacted bentonite for validation of the model predictions.

Impact/achievability: High/high

Priority: High – needed for validation of CCM-SCC model predictions.

Lifetime Prediction of Copper Containers

The aim of this program is to develop mechanistically based corrosion models to make justifiable predictions of the long-term corrosion behaviour of copper UFC.

Lifetime predictions for copper UFC have been based on a series of reactive-transport models based around the mechanism of the uniform corrosion of copper in contact with compacted buffer material. At this stage of the program, these models are deemed to be sufficiently developed, with further development awaiting additional mechanistic information from experimental programs.

However, two high-priority and a number of lower priority gaps have been identified (Table 10):

Gap: *Insufficient validation of localized corrosion predictions.*

Rationale: Historically, localized corrosion of copper UFC has been predicted on the basis of a maximum pit depth or through use of a pitting factor. Localized corrosion is now believed to take the form of surface roughening rather than discrete pitting, but no formal procedure has been developed for predicting the extent of this surface roughening.

Impact/achievability: High/high

Priority: Medium – need to validate surface roughening mechanism.

Gap: *Effect of pore-water species other than Cl⁻ (e.g., SO₄²⁻, HCO₃⁻/CO₃²⁻) on speciation of Cu corrosion products.*

Rationale: The CCM codes are based on the assumption of a Cl⁻ -dominated pore water. However, other pore-water species, such as SO₄²⁻, HCO₃⁻/CO₃²⁻, could impact the speciation of dissolved copper and the propensity for passivation.

Impact/achievability: High/low

Priority: Medium – evidence that system is Cl⁻ dominated needs to be verified.

Gap: *Multi-dimensional reactive-transport modelling.*
Rationale: The CCM codes are based on one-dimensional reaction-diffusion equations. This dimensionality causes some difficulties when simulating a 3-dimensional repository design.

Impact/achievability: Low/low
Priority: Low – no immediate need for multi-dimensional modelling.

Gap: *Validation of reactive-transport models.*
Rationale: There has been limited validation of the CCM codes against experimental data.

Impact/achievability: High/high
Priority: High – limited CCM validation to date.

Gap: *Corrosion model for unsaturated conditions.*
Rationale: The effects of unsaturated conditions on the uniform and localized corrosion behaviour of copper UFC are not explicitly included in the CCM codes.

Impact/achievability: High/high
Priority: High – need to address corrosion during unsaturated period.

Gap: *Behaviour of UFC following penetration of the Cu corrosion barrier.*
Rationale: Release of radionuclides from a failed copper UFC will depend in part on the time dependence of the size and shape of the aperture in the outer Cu corrosion barrier. No assessment has been made of the effect of corrosion of the inner C-steel vessel or the effect of the possible expansion of corrosion products on the size of the opening in the Cu shell.

Impact/achievability: High/low
Priority: Low – will need addressing at some future stage.

Gap: *Assessment of the maximum allowable temperature for Cu UFC from a corrosion viewpoint.*

Rationale: There may be some advantages in having a higher maximum design temperature for the container (e.g., closer UFC spacing, avoidance of MIC issues in the near field). The maximum container surface from the viewpoint of corrosion has not been determined.

Impact/achievability: Low/high
Priority: Low – must first establish whether elevated temperature is acceptable for other barriers, particularly with regard to bentonite stability.

Gas Generation from Failed Copper Containers

The aim of this program is to determine the impact of gas generation from a failed Cu/C-steel UFC on the safety and performance of the repository. The identified gaps are of low priority (Table 11), partly because of the minor safety consequences found for this scenario in the Third case study (Gierszewski et al. 2004).

Gap: *Assessment of the rate of H₂ generation from C-steel corrosion for a defected Cu outer corrosion barrier.*

Rationale: Hydrogen will be generated from anaerobic corrosion of the C-steel inner vessel once the outer copper corrosion barrier has been penetrated. Estimates from the Third Case study suggest that the rate of H₂ generation is small.

Impact/achievability: Low/low

Priority: Low – safety consequences expected to be minimal.

Gap: Assessment of the range of defect sizes in the Cu corrosion barrier, and effects of expansivity of corrosion products.

Rationale: The rate of generation of H₂ from a defected Cu/C-steel container depends on the size of the aperture in the Cu corrosion barrier. The time dependence of the size and shape of the aperture is currently uncertain.

Impact/achievability: Low/low

Priority: Low – existing evidence suggests expansivity of C-steel corrosion products is minor.

4.2.3 Carbon Steel Research Areas

Stress Corrosion Cracking of Carbon Steel

C-steel is known to be susceptible to SCC under certain conditions and the aim of this program is to determine the impact of this form of corrosion on the lifetime of a C-steel UFC.

In general, SCC of C-steel UFC appears unlikely based on the nature of the environmental conditions and the expected low level of tensile stress on the container. However, there may be some benefit in having a limited program so that conjectural questions can be addressed; for example, does mechanical damage to a container during handling increase the likelihood of SCC?

The gaps in the existing knowledge that have been identified include (Table 12):

Gap: Assessment of welding procedures to minimize residual stress.

Rationale: Welding procedures can be optimized to minimize the residual stress and, in turn, minimize the possibility of SCC. Definition of appropriate welding procedures, and possibly some development work, are needed.

Impact/achievability: High/low

Priority: Medium – needs to be addressed once welding procedure defined.

Gap: Assessment of non-thermal, post-weld stress relief treatments.

Rationale: An alternative approach to minimizing residual stress is to apply some form of post-weld stress relief. There are a number techniques available that do not require heating the UFC above the cladding temperature limit. These techniques need to be assessed to determine their practicality.

Impact/achievability: High/low

Priority: Medium – needs to be addressed once welding procedure and stress-relief treatments have been defined.

Gap: Assessment of whether near-neutral pH SCC conditions will develop at the UFC surface in the repository.

Rationale: Of any of the environments known to cause SCC of C-steels, that most likely to form at the surface of a UFC is the so-called near-neutral pH SCC environment known to cause SCC of pipeline steels. This form of cracking could be excluded from consideration if it could be shown that the necessary environment does not form in the repository near field.

Impact/achievability: High/high

Priority: Medium – could exclude this form of SCC from consideration.

Gap: *SCC of candidate UFC material(s) in dilute $\text{HCO}_3^-/\text{CO}_3^{2-}$ environments under cyclic, SSRT, and static load.*

Rationale: Although most literature reports of the near-neutral pH SCC of pipeline steels indicate that some degree of cyclic loading is necessary to sustain cracking, there are periodic reports of SCC under static load (as would exist on a C-steel UFC in the repository). The possibility of this form of SCC under the loading conditions existing in the repository needs to be assessed.

Impact/achievability: Low/high

Priority: Medium – need for cyclic load would exclude possibility of SCC.

Gap: *SCC of candidate UFC material(s) under a range of repository conditions.*

Rationale: Once the candidate C-steel material has been defined, its susceptibility to SCC should be compared with the known susceptibility of other materials in environments known to cause SCC and which could form in a DGR.

Impact/achievability: High/high

Priority: Medium – need to exclude possibility of SCC of C-steel UFC.

Corrosion of Carbon Steel Welds

The aim of this program is to determine whether welding of C-steel UFC leads to an increase in the rate of uniform corrosion or an increased susceptibility to SCC or H-related degradation mechanisms.

As with the program on the effects of copper UFC welding, this issue needs to be addressed at some stage although it is currently of lower priority than other areas (Table 13).

Gaps in existing knowledge that have been identified include:

Gap: *Candidate welding procedures for a C-steel UFC and fabrication of prototype weld material for laboratory testing.*

Rationale: A welding technique will need to be defined for the final closure weld on the container. The welding technique should be selected based on the following criteria: ease and speed of weld production, suitability for remote operation, level of residual stress, inspectability, etc. Once a technique has been selected, prototype weld material should be prepared for laboratory testing.

Impact/achievability: High/high

Priority: Low- can be deferred until later.

Gap: *Uniform and localized corrosion behaviour of base, HAZ, and weld metal.*

Rationale: Candidate weld material should be tested for possible enhanced uniform and localized corrosion of the base and weld metal and HAZ.
Impact/achievability: High/low
Priority: Medium – can be deferred until later.

Gap: Susceptibility of base, HAZ, and weld metal to H-related degradation mechanisms and SCC.

Rationale: Candidate weld material should be tested for possible enhanced hydrogen-related degradation mechanisms and SCC of the base and weld metal and HAZ.

Impact/achievability: High/low
Priority: Medium – can be deferred until later.

Uniform Corrosion of Carbon Steel

Uniform corrosion is one of the major corrosion mechanisms for C-steel UFC and a program is required to develop an understanding of the mechanisms of corrosion under both aerobic and anaerobic conditions and to develop input data for predictive models.

Unlike the well-developed program for the uniform corrosion of copper, the DGRTP has relatively little information regarding the mechanisms of the uniform corrosion of C-steel. Although information is available in the literature and from other international programs, similar studies under Canadian repository conditions should be performed.

The gaps in the current understanding include (Table 14):

Gap: Uniform corrosion of C-steel in compacted buffer material under aerobic conditions representative of a DGR in sedimentary host rock.

Rationale: Although the aerobic period is expected to be short, the factors controlling the rate of uniform corrosion should be established.

Impact/achievability: High/high
Priority: High – require information on this major corrosion mechanism for C-steel.

Gap: Range of uniform corrosion rates of C-steel under anaerobic saturated conditions representative of a DGR in sedimentary host rock.

Rationale: Following consumption of the initially trapped O₂ and saturation of the repository, corrosion of C-steel UFC will occur under anaerobic, saturated conditions. The rate of anaerobic corrosion not only determines the lifetime of the container but also the rate of H₂ generation. There is a wide range of reported rates in the literature. A value for the conditions expected in a Canadian DGR should be established.

Impact/achievability: High/high
Priority: Medium – existing database from international programs, but data needed for Canadian conditions.

Gap: Range of uniform corrosion rates of C-steel under anaerobic unsaturated conditions representative of a DGR in sedimentary host rock.

Rationale: If the rate of saturation of a DGR in sedimentary host rock is slow because of the low hydraulic conductivity, there could be an extended period of unsaturated, anaerobic corrosion of C-steel in unsaturated bentonite or humid atmospheres.

Impact/achievability: High/high

Priority: High – few existing data available.

Gap: Kinetics of H₂O reduction on C-steel and its corrosion products, especially Fe₃O₄.

Rationale: The cathodic reduction of H₂O supports the anaerobic corrosion of C-steel. Although the rate of corrosion is likely to be anodically, rather than cathodically, limited, studies of the reduction of H₂O are still important. For example, if H₂O reduction occurs on Fe₃O₄ then the effective cathodic surface area could increase with time. Alternatively, H₂O reduction on a defected or porous Fe₃O₄ corrosion product could support localized corrosion during the anaerobic phase.

Impact/achievability: High/high

Priority: High – potentially important for long-term corrosion performance.

Gap: Effect of film formation on the anodic dissolution kinetics.

Rationale: It is likely that the rate of anaerobic corrosion is anodically controlled, possibly due to a transport process across a precipitated film. The nature and properties of the anodic film formed under freely corroding conditions need to be established.

Impact/achievability: High/high

Priority: Medium – potentially important effect.

Gap: Mechanism of the long-term anaerobic corrosion of C-steel.

Rationale: In conjunction with studies of the individual anodic and cathodic processes, the mechanism of the anaerobic corrosion of C-steel needs to be established in order to justify long-term predictions of the corrosion behaviour of C-steel UFC. This mechanistic description should identify the overall rate-controlling process.

Impact/achievability: High/high

Priority: High – needed to justify long-term predictions.

Gap: Mixed-potential model for the prediction of E_{CORR} of C-steel containers, including the effects of film formation and transformation and the evolution of the repository environment.

Rationale: The development of a mixed-potential model for copper UFC has proved useful in not only predicting the long-term corrosion behaviour of the containers (uniform and localized corrosion, SCC, and MIC), but also the evolution of the repository environment. A similar model is required for C-steel containers to predict the corrosion behaviour and the rate of H₂ generation.

Impact/achievability: High/high

Priority: High – may take several years to develop, linked to other programs.

Gap: Effect of high [Cl] on anaerobic corrosion rates of C-steel.
Rationale: A characteristic of the sedimentary host rock in which C-steel UFC could be used is the highly saline groundwater. There is conflicting evidence in the literature on the effect of [Cl] on the corrosion rate of C-steel.

Impact/achievability: High/low
Priority: Medium – some evidence currently available.

Gap: Effect of H₂ partial pressure on C-steel corrosion rate, H₂ evolution reaction, and E_{CORR}.

Rationale: Thermodynamically, corrosion of C-steel should cease in the presence of a H₂ over-pressure equal to the equilibrium partial pressure of H₂ for the corrosion reaction. Previous studies have shown no effect of H₂ pressure on either the corrosion rate or the corrosion potential. Confirmation of these observations under Canadian repository conditions is required.

Impact/achievability: High/high
Priority: High – useful argument to be able to make if valid. Could be experimentally challenging because of high H₂ partial pressures required.

Gap: Effect of CO₂ partial pressure on corrosion behaviour under anaerobic conditions.

Rationale: The presence of carbonate minerals in the bentonite and repository host rock could lead to elevated CO₂ partial pressures, which in turn may influence the corrosion behaviour of a C-steel UFC. The effect of CO₂ on the corrosion of C-steel should be reviewed.

Impact/achievability: Low/high
Priority: Medium –existing data from the oil and gas industry.

Lifetime Prediction of Carbon Steel Containers

This program aims to develop modelling capability to make long-term, justifiable predictions of the lifetimes of C-steel UFC. A conceptual, semi-empirical container lifetime model is currently available for C-steel UFC and lifetime predictions have been made (King 2007). At this stage of the program, there is limited need to further develop this, or an alternative, model until such time that further mechanistic information becomes available.

The following gaps have been identified (Table 15):

Gap: Development of a mixed-potential model for C-steel UFC.

Rationale: Mixed-potential models are useful for predicting the corrosion behaviour of the UFC and the evolution of the repository environment. A C-steel model would allow the prediction of the rate of uniform corrosion and, under anaerobic conditions, the corresponding rate of H₂ production. The predicted E_{CORR} values would also be of use in predicting the probability of localized corrosion and SCC. In addition to predicting the rate of H₂ production, the model would also predict the rate of Fe(II) release into the bentonite.

Impact/achievability: High/high
Priority: High – basic tool for predicting C-steel UFC lifetimes.

Gap: Assessment of the extent of localized corrosion of C-steel UFC under Canadian repository conditions, including during the unsaturated period.

Rationale: Localized corrosion is an important potential failure mechanism for C-steel UFC. Current information suggests that the degree of localized corrosion should diminish with time. Once further experimental evidence for the mechanism of localized corrosion becomes available, the extent of attack on a UFC should be assessed.

Impact/achievability: Low/high

Priority: Low – defer until more experimental data are available.

Gap: Definition of (range of) uniform corrosion rate(s) of C-steel under anaerobic conditions.

Rationale: Defining the rate of uniform corrosion under anaerobic conditions is important for lifetime prediction, as well as estimation of the rate of H₂ generation. Currently, there is a wide range of reported values in the literature. Measurements under “Canadian” DGR conditions are required, which can be used for lifetime prediction and/or validation of a C-steel mixed-potential model.

Impact/achievability: Low/high

Priority: Low – should wait until more experimental data are available.

Gap: Assessment of the likelihood of H-related degradation mechanisms under Canadian repository conditions.

Rationale: C-steel is susceptible to H-related degradation mechanisms. Specification of a resistant grade of steel should minimize the impact of H-related mechanisms, but the probability of failure should be assessed once additional information becomes available.

Impact/achievability: Low/high

Priority: Medium – defer until more experimental data become available.

Gap: Assessment of the likelihood of SCC under Canadian repository conditions.

Rationale: Based on existing SCC mechanisms for C-steel, the probability of failure by cracking seems low. At some stage, this assessment should be reviewed in light of newly acquired data.

Impact/achievability: Low/high

Priority: Low - defer until more experimental data become available, but probably more existing knowledge than for H-related effects.

Gap: Assessment of the maximum allowable temperature for C-steel UFC from a corrosion viewpoint.

Rationale: There may be some advantages in having a higher maximum design temperature for the container (e.g., closer UFC spacing, avoidance of MIC issues in the near field). The maximum container surface from the viewpoint of corrosion has not been determined.

Impact/achievability: Low/high

Priority: Low - must first establish whether elevated temperature is acceptable for other barriers, particularly with regard to bentonite stability

Gas Generation from Carbon Steel Containers

Gas generation is one of the major issues with the use of a C-steel UFC and the aim of this program is to assess the impact of H₂ generated by corrosion on the safety of the repository.

Corrosion-related gaps in the existing knowledge include (Table 16):

Gap: *Benefits (and drawbacks) of reducing the rate of H₂ generation through small alloying additions.*

Rationale: If the rate of H₂ generation from C-steel is found to be higher than that which can be safely transported away from the UFC surface through the near-field and geosphere, alloying of the material could be considered to reduce the corrosion (and H₂ generation) rate. Although the addition of alloying elements (such as small amounts of Ni or Cr) will tend to passivate the surface and, therefore, reduce the corrosion rate, passivation may render the surface more susceptible to localized corrosion, SCC, or H effects. The relative benefits of alloying would need to be assessed.

Impact/achievability: High/high

Priority: Medium – need first to establish whether H₂ can be effectively transported away for unalloyed C-steel UFC.

Gap: *Long-term H₂ generation (or anaerobic corrosion) rate under saturated Canadian repository conditions.*

Rationale: The rate of anaerobic corrosion under saturated conditions is needed to assess the impact of H₂ generation on repository safety.

Impact/achievability: High/high

Priority: Medium – information required for lifetime prediction as well.

Gap: *Long-term H₂ generation (or anaerobic corrosion) rate under unsaturated Canadian repository conditions.*

Rationale: There could be a significant period of unsaturated conditions in a DGR in sedimentary host rock because of the expected low hydraulic conductivity. The rate of anaerobic corrosion under unsaturated conditions is needed to assess the impact of H₂ generation on repository safety.

Impact/achievability: High/high

Priority: High – information required for lifetime prediction as well.

Hydrogen-related Degradation Mechanisms

The aim of this program is to assess the impact of absorbed hydrogen on the lifetime of C-steel UFC as a result of various H-related degradation mechanisms. A distinction can be drawn between H-related mechanisms that require an applied or residual stress and those that do not. In both cases, the probability of H-related failure is believed to be small although a number of gaps can be identified (Table 17):

Gap: *Measurement of c_H^0 values for candidate steels under repository conditions.*

Rationale: Hydrogen-related effects are generally assessed by comparing the threshold value for various failure mechanisms ($(c_H^0)_{TH}$) to measured sub-

surface hydrogen concentrations (c_H^0) determined under the expected service conditions. Currently, there is a small database of c_H^0 values for modern steels representative of those from which a C-steel UFC would be manufactured.

Impact/achievability: High/high
Priority: Medium – gaps in knowledge, but not considered likely UFC failure mechanism.

Gap: Measurement of $(c_H^0)_{TH}$ values for candidate UFC steels for various hydrogen degradation mechanisms.

Rationale: Hydrogen-related effects are generally assessed by comparing the threshold value for various failure mechanisms ($(c_H^0)_{TH}$) to measured sub-surface hydrogen concentrations (c_H^0) determined under the expected service conditions. Currently, there is a small database of $(c_H^0)_{TH}$ values for modern steels representative of those from which a C-steel UFC would be manufactured.

Impact/achievability: High/high
Priority: Medium – gaps in knowledge, but not considered likely UFC failure mechanism.

Gap: Effect of corrosion product on H uptake.

Rationale: Precipitated corrosion product will tend to block the uptake of H, thus reducing the potential susceptibility to H-related failure mechanisms. However, if H_2 evolution is catalyzed by corrosion product films (e.g., Fe_3O_4), the susceptibility could be enhanced.

Impact/achievability: High/high
Priority: Medium – gaps in knowledge, but not considered likely UFC failure mechanism.

Gap: Effect of welding on H-degradation mechanisms.

Rationale: Welds in C-steel have traditionally been subject to hydrogen cracking. Proper weld techniques and procedures are required to minimize the effect of hydrogen cracking.

Impact/achievability: High/high
Priority: Medium – gaps in knowledge, but not considered likely UFC failure mechanism.

Gap: Assessment of the probability of H-related degradation mechanisms for C-steel UFC.

Rationale: The overall susceptibility of C-steel UFC to H-related failure mechanisms needs to be addressed.

Impact/achievability: High/high
Priority: Medium – gaps in knowledge, but not considered likely UFC failure mechanism.

4.2.4 Summary of Research Areas

Table 18 gives a summary of each of the proposed corrosion research areas along with their aims and the overall priority rating. These priority ratings are based on an average from each of the individual gaps identified in each area. However, there may be High priority individual gaps within an area that is otherwise rated as Medium or Low priority.

5. SUMMARY

The current state-of-the-art and gaps in our understanding of the corrosion behaviour of copper and C-steel used fuel containers have been reviewed and documented. The state-of-the-art review covers not only experience in the Canadian Technical R&D Program, but also experience from international programs with similar repository concepts and container materials.

Gaps in the current understanding of the corrosion behaviour were developed, in part, during a corrosion workshop sponsored by the NWMO in June 2007. This report serves as a partial record of the outcome of that meeting.

A number of gaps have been identified and further categorized into proposed research areas. The fourteen proposed research areas are:

1. Microbiologically influenced corrosion (copper and C-steel)
2. Corrosion during the unsaturated period (copper and C-steel)
3. Evolution of corrosion damage (copper and C-steel)
4. Stress corrosion cracking of copper
5. Corrosion of copper welds
6. Uniform corrosion of copper
7. Lifetime prediction for copper containers
8. Gas generation from failed copper containers
9. Stress corrosion cracking of C-steel
10. Corrosion of C-steel welds
11. Uniform corrosion of C-steel
12. Lifetime prediction for C-steel containers
13. Gas generation from C-steel containers
14. Hydrogen-related C-steel degradation mechanisms

Each individual gap and the larger research areas have been prioritized. Prioritization of the gaps and proposed future studies was based on one of two criteria:

1. the need to demonstrate sufficient scientific and engineering understanding of a given corrosion phenomenon, and
2. the need to develop deep geological repository designs for the program over the next 2-3 years.

The contents of this report should be of use for planning of the Canadian Technical R&D Program future corrosion studies.

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Table 4: Knowledge Gaps for Microbiologically Influenced Corrosion of UFC.

(a) Prevention of Microbial Activity in Repository

Gap	Comment/rationale	Impact/ achievability	Priority
1. Can biofilm formation on the container surface be prevented through the use of highly compacted bentonite?	The current work focussing on the effects of water activity and bentonite swelling pressure on microbial activity needs to be completed.	H/L	M
2. What is the physiological mechanism(s) for the suppression of microbial activity in compacted bentonite?	A physiological explanation for the apparent effect of high bentonite swelling pressure and/or low water activity is required to support the argument for the absence of microbial activity in compacted bentonite.	H/L	M
3. Need to establish engineering confidence that criteria for the suppression of microbial activity can be guaranteed in the repository	Need to demonstrate that microbial activity is suppressed at all times and at all locations within the repository, especially at interfaces and especially during the saturation phase.	H/L	H
4. Can an upper temperature limit for microbial activity in the repository be established?	It is likely that high temperature (>100°C?) will kill microbes and could produce a microbially-depleted zone around the UFC. This would be a strong argument in support of the current model that no microbial activity will occur in the near field, provided the necessary temperature can be attained in the repository without deleterious effects on the container or bentonite.	H/L	M

Table 4: Knowledge Gaps for Microbiologically Influenced Corrosion of UFC.

(b) Assessment of Maximum Damage Due to Microbial Activity in Repository

Gap	Comment/rationale	Impact/achievability	Priority
5. Prediction of maximum amount of metabolic by-products that can be formed if microbial activity at container surface is possible	An estimation of the maximum amount of aggressive metabolic by-products is required in the event that microbial activity is possible in the near field. Different species would be considered for the two different container materials.	H/H	H
6. Mass-balance calculations to predict amount of damage due to assumed limiting factor (e.g., organic C, electron acceptor)	Based on the above calculations, the maximum amount of damage that could result from microbial activity if it were possible needs to be determined, for both copper and C-steel UFC.	H/H	H
7. Sensitivity analyses with CCM-MIC to determine worst-case effect of assuming limitation by organic C, electron acceptor, etc.	The CCM-MIC is an ideal tool for assessing the maximum amount of metabolic by-products for different assumptions regarding the limiting species/process.	H/H	H
8. Development of UFC biofilm model	Recent developments in the modelling of biofilms now makes it possible to develop biofilm MIC models to predict the extent of attack and the degree of localization.	H/L	M
9. Specific Cu/Fe corrosion experiments to assess extent of damage under <u>realistic</u> exposure conditions	To support predictions of the maximum damage that could occur, well-planned, targeted experiments under realistic exposure conditions should be considered.	H/H	H

Table 5: Knowledge Gaps for UFC Corrosion During the Unsaturated Phase.

Gap	Comment/rationale	Impact/achievability	Priority
Experimental evidence for the nature of the surface contaminants on copper and C-steel UFC and their deliquescence behaviour	The wetting of the UFC surface, both in terms of the time of wetting and the distribution of surface moisture, will be determined by the deliquescence behaviour of surface contaminants.	H/H	H
Experimental evidence for the degree of localization of corrosion of copper UFC during the unsaturated phase	Initial wetting of the UFC surface is likely to be non-uniform and may result in localized corrosion of the container.	H/H	H
Experimental evidence for the degree of localization of corrosion of C-steel UFC during the unsaturated phase	Initial wetting of the UFC surface is likely to be non-uniform and may result in localized corrosion of the container.	H/H	H
Range of uniform corrosion rates of C-steel under anaerobic unsaturated conditions	In sedimentary host rock, it is likely that the DGR will become anaerobic before it fully saturates. Consequently, there may be a period of anaerobic corrosion under unsaturated conditions, for which there are few reported corrosion rates.	H/H	H
Model for assessing the extent of damage of copper and C-steel UFC during the unsaturated phase	Corrosion during the unsaturated phase is not currently explicitly included in the lifetime prediction models for copper and C-steel containers, particularly the possibility of localized attack.	H/H	M
Estimation of time-temperature-relative humidity relationship during the unsaturated phase	The time at which the surface first wets is determined, in part, by the time dependence of the relative humidity and temperature at the container surface.	H/H	M

Table 6: Knowledge Gaps for the Evolution of Corrosion Damage.

Gap	Comment/rationale	Impact/ achievability	Priority
Evolution of the surface roughness of Cu from the initial aerobic phase to the long-term anoxic period	Corrosion of copper in compacted bentonite is known to exhibit surface roughening, but how that roughness evolves over time is uncertain.	H/H	H
Model for predicting surface roughening of Cu and how it evolves with time	A mechanistically based model for predicting surface roughening of copper is needed.	H/H	H
Degree of surface roughness associated with the uniform corrosion of C-steel in compacted bentonite	The degree of surface roughening of C-steel in contact with bentonite is uncertain.	H/H	H
Whether a C-steel UFC will passivate under repository conditions and, if so, the duration of passivity	Localized corrosion of C-steel is more likely if the surface passivates under repository conditions. Japanese evidence suggests that passivation is unlikely, but the potential for an extended unsaturated phase in a DGR in sedimentary host rock makes passivation more likely.	H/H	M
Mechanistic understanding of the depth-dependent pitting factor and/or stifling of localized corrosion of C-steel	Currently, localized attack on C-steel is predicted based on a time- (or depth-) dependent pitting factor. The empirical data suggest that localized corrosion stifles, but the mechanism is uncertain.	H/H	M
Database of film breakdown and re-passivation potentials for C-steel under repository conditions	An alternative approach to predicting the localized corrosion of C-steel is through comparison of the corrosion potential E_{CORR} to pitting or re-passivation potentials. A database of such potentials is required.	L/H	L
Localized corrosion due to reductive dissolution of Fe(III) films during the aerobic-anaerobic transition	During the evolution of the environmental conditions in the repository, previously formed Fe(III) corrosion products will be reduced to the Fe(II) state, a process that may induce localized corrosion.	H/H	M

Table 7: Knowledge Gaps for Stress Corrosion Cracking of Copper UFC.

Gap	Comment/rationale	Impact/ achievability	Priority
Mechanistic understanding of SCC of Cu under repository conditions	The nature of the processes controlling crack initiation and growth need to be determined to support long-term predictions.	H/H	M
Understanding of the apparent inhibitive effect(s) of Cl ⁻	Experimental evidence is available indicating that Cl ⁻ ions inhibit the SCC of copper in nitrite, ammonia, and acetate environments, but the precise mechanism is uncertain. This information is required to support long-term SCC predictions.	H/H	M
Insufficient bulk potential and pH data for SCC in acetate and ammonia environments	A more-extensive database of potentials and pH values for the SCC of copper in acetate and ammonia is needed to support the current modelling approach based on a threshold potential and pH for cracking.	H/L	M
Uncertainty whether SCC could occur under anaerobic conditions	Recent Japanese data suggest the possibility of SCC of copper under anaerobic conditions in the presence of sulphide. Although the concentration of sulphide in a Canadian DGR is likely to be lower than the reported threshold concentration, there remains the question of whether SCC can occur under long-term anaerobic conditions.	H/L	M/L
Lack of validation of predicted interfacial pH values from CCM-SCC code	The current SCC modelling approach is based on a comparison of predicted potentials and pH values with a criterion based on the equilibrium E/pH for Cu ₂ O/CuO. The predicted pH values need validation against experimental measurements.	H/H	M
Assessment of effectiveness of non-thermal, post-weld stress relief treatments	Sound engineering practice dictates that the residual stress of the final closure weld be minimized to limit the probability of SCC. The ability of various low-temperature methods for reducing the residual stress should be assessed.	H/L	L

..... continued.

Table 7: Knowledge Gaps for Stress Corrosion Cracking of Copper UFC (concluded).

Gap	Comment/rationale	Impact/ achievability	Priority
Effect of phosphorus on SCC behaviour	The reference OFP Cu material contains 50-70 ppm P to improve the creep ductility. However, these levels of P have been shown to increase the susceptibility of Cu to SCC in ammonia environments.	L/H	M
Effect of Cu properties (degree of cold work, yield stress, grain size, etc.)	The degree of cold work, mechanical properties, and grain size are known to affect the SCC susceptibility of Cu. An understanding of these effects is required to address the impact of container fabrication processes and potential upset conditions (e.g., the accidental introduction of cold work due to impacts during handling and emplacement of the container).	L/H	M
Properties of oxide films and their role in SCC	SCC of copper is observed under environmental conditions that support the formation of a surface film. The properties of these films need to be understood in order to support long-term predictions.	H/H	H

Table 8: Knowledge Gaps for Corrosion of Copper Welds.

Gap	Comment/rationale	Impact/ achievability	Priority
Possibility of enhanced uniform corrosion, localized attack, and SCC of welds	Manufactured components frequently fail at weld locations. An assessment will be required of the possible enhanced susceptibility of welds on a copper UFC to uniform corrosion, localized attack, and SCC.	L/H	M

Table 9: Knowledge Gaps for Uniform Corrosion of Copper.

Gap	Comment/rationale	Impact/ achievability	Priority
Rates of precipitation and dissolution of Cu_2O , $\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$, and other Cu corrosion products	Precipitation of corrosion products is treated in the CCM codes, but reliable data for the rates of precipitation and dissolution are not available.	H/L	M/L
Limited E_{CORR} data in compacted bentonite for validation of mixed-potential models	One of the main features of the CCM codes is the prediction of E_{CORR} . However, there are few E_{CORR} measurements in compacted bentonite against which to validate the model.	H/H	H
Limited large-scale test data for validation of CCM predictions	The CCM code predicts the spatial and temporal distributions of various dissolved Cu species, as well as the corrosion rate and the potential for localized corrosion. There has been limited validation of the model. Data from large-scale tests are ideal for validation purposes.	H/L	H
Corrosion of Cu in unsaturated bentonite	If the surface of a copper UFC becomes uniformly wetted during the unsaturated phase before the complete consumption of O_2 , there could be a period of uniform corrosion under unsaturated conditions. No experimental data are available to support predictions of the rate of corrosion under these conditions.	H/H	H
Extent of Cu(II) reduction on corroded Cu surfaces	In most O_2 -containing natural waters, the actual oxidant is Cu(II), produced by the homogeneous oxidation of Cu(I) by O_2 , rather than O_2 itself. The interfacial reduction of Cu(II) is included in the CCM codes, but no direct measurements of the rate constant under simulated repository conditions have been made.	L/H	L/M
Availability of Fe(II) in buffer and backfill pore water for redox reactions with O_2	Part of the O_2 initially trapped in the repository is consumed by reaction with Fe(II) dissolved from mineral impurities. Dissolved Fe(II) may precipitate as a secondary phase if the concentration exceeds the solubility. Dissolution of Fe(II) is modelled based on biotite dissolution, but the validity of this treatment has not been demonstrated.	L/H	L

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Table 9: Knowledge Gaps for Uniform Corrosion of Copper (concluded).

Gap	Comment/rationale	Impact/ achievability	Priority
Properties of oxide films	Oxide films are known to form on copper under simulated disposal conditions, but film formation is only included in the CCM code as a sink for Cu(I). The possible effects of Cu ₂ O on the rate of interfacial processes, the probability for SCC, or for localized corrosion are not currently included in lifetime prediction models.	H/H	H
Thermodynamic stability of Cu at high [Cl ⁻]	There are repeated suggestions that copper may be thermodynamically unstable with respect to H ₂ O in high salinity groundwaters. Despite various attempts by Posiva, no credible evidence for this suggestion has been obtained, but the question may arise again, especially if Cu is considered as a container material for the more highly saline groundwaters expected for DGR in sedimentary host rock.	L/L	L
Effect of film formation on E _{CORR}	Film formation typically results in the ennoblement of E _{CORR} . Although film formation is known to occur under simulated repository conditions, the effects of film formation on E _{CORR} are not currently included in the CCM codes.	H/H	H/M
Measurement of interfacial pH	The CCM-SCC code includes the prediction of the interfacial pH. There are no experimental measurements of the interfacial pH of corroding Cu in contact with compacted bentonite for validation of the model predictions.	H/H	H

Table 10: Knowledge Gaps for Lifetime Prediction of Copper Containers.

Gap	Comment/rationale	Impact/ achievability	Priority
Insufficient validation of localized corrosion predictions	Historically, localized corrosion of copper UFC has been predicted based on the basis of a maximum pit depth or through use of a pitting factor. The attack is now believed to take the form of surface roughening rather than discrete pitting, but no formal procedure has been developed for predicting the extent of this surface roughening.	H/H	M
Effect of pore-water species other than Cl ⁻ (e.g., SO ₄ ²⁻ , HCO ₃ ⁻ /CO ₃ ²⁻) on speciation of Cu corrosion products	The CCM codes are based on the assumption of a Cl ⁻ -dominated pore water. However, other pore-water species, such as SO ₄ ²⁻ , HCO ₃ ⁻ /CO ₃ ²⁻ , could impact the speciation of dissolved copper and the propensity for passivation.	H/L	M
Multi-dimensional reactive-transport modelling	The CCM codes are based on one-dimensional reaction-diffusion equations. This dimensionality causes some difficulties when simulating a 3-dimensional repository design.	L/L	L
Validation of reactive-transport models	There has been limited validation of the CCM codes against experimental data.	H/H	H
Corrosion model for unsaturated conditions	The effects of unsaturated conditions on the uniform and localized corrosion behaviour of the copper UFC are not explicitly included in the CCM codes.	H/H	H
Behaviour of UFC following penetration of the Cu corrosion barrier	Release of radionuclides from a failed copper UFC will depend in part on the time dependence of the size and shape of the aperture in the outer Cu corrosion barrier. No assessment has been made of the effect of corrosion of the inner C-steel vessel and the possible expansion of corrosion products on the size of the opening in the Cu shell.	H/L	L
Assessment of the maximum allowable temperature for Cu UFC from a corrosion viewpoint	There may be some advantages in having a higher maximum design temperature for the container (e.g., closer UFC spacing, avoidance of MIC issues in the near field). The maximum container surface from the viewpoint of corrosion has not been determined.	L/H	L

Table 11: Knowledge Gaps for Gas Generation from Failed Copper Containers.

Gap	Comment/rationale	Impact/ achievability	Priority
Assessment of the rate of H ₂ generation from C-steel corrosion for a defected Cu outer corrosion barrier	Hydrogen will be generated from anaerobic corrosion of the C-steel inner vessel once the outer copper corrosion barrier has been penetrated. Estimates from the Third Case Study suggest that the rate of H ₂ generation is small.	L/L	L
Assessment of the range of defect sizes in the Cu corrosion barrier, and effects of expansivity of corrosion products	The rate of generation of H ₂ from a defected Cu/C-steel container depends on the size of the aperture in the Cu corrosion barrier. The time dependence of the size and shape of the aperture is currently uncertain.	L/L	L

Table 12: Knowledge Gaps for Stress Corrosion Cracking of C-steel.

Gap	Comment/rationale	Impact/achievability	Priority
Assessment of welding procedures to minimize residual stress	Welding procedures can be optimized to minimize the residual stress and, in turn, minimize the possibility of SCC. Definition of appropriate welding procedures, and possibly some development work, are needed.	H/L	M
Assessment of non-thermal, post-weld stress relief treatments	An alternative approach to minimizing residual stress is to apply some form of post-weld stress relief. There are a number techniques available that do not require heating the UFC above the cladding temperature limit. These techniques need to be assessed to determine their practicality.	H/L	M
Assessment of whether near-neutral pH SCC conditions will develop at the UFC surface in the repository	Of any of the environments known to cause SCC of C-steels, that most likely to form at the surface of a UFC is the so-called near-neutral pH SCC environment known to cause SCC of pipeline steels. This form of cracking could be excluded from consideration if it could be shown that the necessary environment does not form in the repository environment.	H/H	M
SCC of candidate UFC material(s) in dilute $\text{HCO}_3^-/\text{CO}_3^{2-}$ environments under cyclic, SSRT, and static load	Although most literature reports of the near-neutral pH SCC of pipeline steels indicate that some degree of cyclic loading is necessary to sustain cracking, there are periodic reports of SCC under static load (as would exist on a C-steel UFC in the repository). The possibility of this form of SCC under the loading conditions existing in the repository needs to be assessed.	L/H	M
SCC of candidate UFC material(s) under a range of repository conditions	Once the candidate C-steel material has been defined, its susceptibility to SCC should be compared with the known susceptibility of other materials in environments known to cause SCC and which could form in a DGR.	H/H	M

Table 13: Knowledge Gaps for Corrosion of C-steel Welds.

Gap	Comment/rationale	Impact/ achievability	Priority
Candidate welding procedures for a C-steel UFC and fabrication of prototype weld material for laboratory testing	A welding technique will need to be defined for the final closure weld on the container. The welding technique should be selected based on the following criteria: ease and speed of weld production, suitability for remote closure weld, level of residual stress, inspectability, etc. Once a technique has been selected, prototype weld material should be prepared for laboratory testing.	H/H	L
Uniform and localized corrosion behaviour of base, HAZ, and weld metal	Candidate weld material should be tested for possible enhanced uniform and localized corrosion of the base and weld metal and HAZ.	H/L	M
Susceptibility of base, HAZ, and weld metal to H-related degradation mechanisms and SCC	Candidate weld material should be tested for possible enhanced hydrogen-related degradation mechanisms and SCC of the base and weld metal and HAZ.	H/L	M

Table 14: Knowledge Gaps for Uniform Corrosion of C-steel.

Gap	Comment/rationale	Impact/ achievability	Priority
Uniform corrosion of C-steel in compacted buffer material under aerobic conditions representative of a DGR in sedimentary host rock	Although the aerobic period is expected to be short, the factors controlling the rate of uniform corrosion should be established.	H/H	H
Range of uniform corrosion rates of C-steel under anaerobic saturated conditions representative of a DGR in sedimentary host rock	Following consumption of the initially trapped O ₂ and saturation of the repository, corrosion of C-steel UFC will occur under anaerobic, saturated conditions. The rate of anaerobic corrosion not only determines the lifetime of the container but also the rate of H ₂ generation. There is a wide range of reported rates in the literature. A value for the conditions expected in a Canadian DGR should be established.	H/H	M
Range of uniform corrosion rates of C-steel under anaerobic unsaturated conditions representative of a DGR in sedimentary host rock	If the rate of saturation of a DGR in sedimentary host rock is slow because of the low hydraulic conductivity, then there could be an extended period of anaerobic corrosion of C-steel in unsaturated bentonite or a humid atmosphere.	H/H	H
Kinetics of H ₂ O reduction on C-steel and its corrosion products, especially Fe ₃ O ₄	The cathodic reduction of H ₂ O supports the anaerobic corrosion of C-steel. Although the rate of corrosion is likely to be anodically, rather than cathodically, limited, studies of the reduction of H ₂ O are still important. For example, if H ₂ O reduction occurs on Fe ₃ O ₄ then the effective cathodic surface area could increase with time. Alternatively, H ₂ O reduction on a defected or porous Fe ₃ O ₄ corrosion product could support localized corrosion during the anaerobic phase.	H/H	H
Effect of film formation on the anodic dissolution kinetics	It is likely that the rate of anaerobic corrosion is anodically controlled, possibly due to a transport process across a precipitated film. The nature and properties of the anodic film formed under freely corroding conditions need to be established.	H/H	M

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Table 14: Knowledge Gaps for Uniform Corrosion of C-steel (concluded).

Gap	Comment/rationale	Impact/ achievability	Priority
Mechanism of the long-term anaerobic corrosion of C-steel	In conjunction with studies of the individual anodic and cathodic processes, the mechanism of the anaerobic corrosion of C-steel needs to be established in order to justify long-term predictions of the corrosion behaviour of C-steel UFC. This mechanistic description should identify the overall rate-controlling process.	H/H	H
Mixed-potential model for the prediction of E_{CORR} of C-steel containers, including the effects of film formation and transformation and the evolution of the repository environment	The development of a mixed-potential model for copper UFC has proved useful in not only predicting the long-term corrosion behaviour of the containers (uniform and localized corrosion, SCC, and MIC), but also the evolution of the repository environment. A similar model is required for C-steel containers to predict the corrosion behaviour and the rate of H_2 generation.	H/H	H
Effect of high $[Cl^-]$ on anaerobic corrosion rates of C-steel	A characteristic of the sedimentary host rock in which C-steel UFC could be used is the highly saline groundwater. There is conflicting evidence in the literature on the effect of $[Cl^-]$ on the corrosion rate of C-steel.	H/L	M
Effect of H_2 partial pressure on C-steel corrosion rate, H_2 evolution reaction, and E_{CORR}	Thermodynamically, corrosion of C-steel should cease in the presence of a H_2 over-pressure equal to the equilibrium partial pressure of H_2 for the corrosion reaction. Previous studies have shown no effect of H_2 pressure on either the corrosion rate or the corrosion potential. Confirmation of these observations under Canadian repository conditions is required.	H/H	H
Effect of CO_2 partial pressure on corrosion behaviour under anaerobic conditions	The presence of carbonate minerals in the bentonite and repository host rock could lead to elevated CO_2 partial pressures, which in turn may influence the corrosion behaviour of a C-steel UFC. The effect of CO_2 on the corrosion of C-steel should be reviewed.	L/H	M

Table 15: Knowledge Gaps for Lifetime Prediction for C-steel Containers.

Gap	Comment/rationale	Impact/ achievability	Priority
Development of a mixed-potential model for C-steel UFC	Mixed-potential models are useful for predicting the corrosion behaviour of the UFC and the evolution of the repository environment. A C-steel model would allow the prediction of the rate of uniform corrosion and, under anaerobic conditions, the corresponding rate of H ₂ production. The predicted E _{CORR} values would also be of use in predicting the probability of localized corrosion and SCC. In addition to predicting the rate of H ₂ production, the model would also predict the rate of Fe(II) release into the bentonite.	H/H	H
Assessment of the extent of localized corrosion of C-steel UFC under Canadian repository conditions, including during the unsaturated period	Localized corrosion is an important potential failure mechanism for C-steel UFC. Current information suggests that the degree of localized corrosion should diminish with time. Once further experimental evidence for the mechanism of localized corrosion becomes available, the extent of attack on a UFC should be assessed.	L/H	L
Definition of (range of) uniform corrosion rate(s) of C-steel under anaerobic conditions	Defining the rate of uniform corrosion under anaerobic conditions is important for lifetime prediction, as well as estimation of the rate of H ₂ generation. Currently, there is a wide range of reported values in the literature. Measurements under "Canadian" DGR conditions are required, which can be used for lifetime prediction and/or validation of a C-steel mixed-potential model.	L/H	L
Assessment of the likelihood of H-related degradation mechanisms under Canadian repository conditions	C-steel is susceptible to H-related degradation mechanisms. Specification of a resistant grade of steel should minimize the impact of H-related mechanisms, but the probability of failure should be assessed once additional information becomes available.	L/H	M
Assessment of the likelihood of SCC under Canadian repository conditions	Based on existing SCC mechanisms for C-steel, the probability of failure by cracking seems low. At some stage, this assessment should be reviewed in light of newly acquired data.	L/H	L
Assessment of the maximum allowable temperature for C-steel UFC from a corrosion viewpoint	There may be some advantages in having a higher maximum design temperature for the container (e.g., closer UFC spacing, avoidance of MIC issues in the near field). The maximum container surface from the viewpoint of corrosion has not been determined.	L/H	L

Table 16: Knowledge Gaps for Gas Generation from C-steel Containers.

Gap	Comment/rationale	Impact/ achievability	Priority
Benefits (and drawbacks) of reducing rate of H ₂ generation through small alloying additions	If the rate of H ₂ generation from C-steel is found to be higher than that which can be safely transported away from the UFC surface through the near-field and geosphere, alloying of the material could be considered to reduce the corrosion (and H ₂ generation) rate. Although the addition of elements (such as small amounts of Ni or Cr) will tend to passivate the surface and, therefore, reduce the corrosion rate, passivation may render the surface more susceptible to localized corrosion, SCC, or H effects. The relative benefits of alloying would need to be assessed.	H/H	M
Long-term H ₂ generation (or anaerobic corrosion) rate under Canadian repository saturated conditions	The rate of anaerobic corrosion under saturated conditions is needed to assess the impact of H ₂ generation on repository safety.	H/H	M
Long-term H ₂ generation (or anaerobic corrosion) rate under Canadian repository unsaturated conditions	There could be a significant period of unsaturated conditions in a DGR in sedimentary host rock because of the expected low hydraulic conductivity. The rate of anaerobic corrosion under unsaturated conditions is needed to assess the impact of H ₂ generation on repository safety.	H/H	H

Table 17: Knowledge Gaps for Hydrogen-related C-steel Degradation Mechanisms.

Gap	Comment/rationale	Impact/ achievability	Priority
Measurement of c_H^0 values for candidate steels under repository conditions	Hydrogen-related effects are generally assessed by comparing the threshold value for various failure mechanisms ($(c_H^0)_{TH}$) to measured sub-surface hydrogen concentrations (c_H^0) determined under the expected service conditions. Currently, there is a small database of c_H^0 values for modern steels representative of those from which a C-steel UFC would be manufactured.	H/H	M
Measurement of $(c_H^0)_{TH}$ values for candidate UFC steels for various hydrogen degradation mechanisms	Hydrogen-related effects are generally assessed by comparing the threshold value for various failure mechanisms ($(c_H^0)_{TH}$) to measured sub-surface hydrogen concentrations (c_H^0) determined under the expected service conditions. Currently, there is a small database of $(c_H^0)_{TH}$ values for modern steels representative of those from which a C-steel UFC would be manufactured.	H/H	M
Effect of corrosion product on H uptake	Precipitated corrosion product will tend to block the uptake of H, thus reducing the potential susceptibility to H-related failure mechanisms. However, if H_2 evolution is catalyzed by corrosion product films (e.g., Fe_3O_4), the susceptibility could be enhanced.	H/H	M
Effect of welding on H-degradation mechanisms	Welds in C-steel have traditionally been subject to hydrogen cracking. Proper weld techniques and procedures are required to minimize the effect of hydrogen cracking.	H/H	M
Assessment of the probability of H-related degradation mechanisms for C-steel UFC	The overall susceptibility of C-steel UFC to H-related failure mechanisms needs to be addressed.	H/H	M

Table 18: Overall Summary of Proposed Research Areas, Their Aims, and Prioritization.

(a) Common Research Areas

	Program	Aim	Priority
1	Microbiologically Influenced Corrosion	<ol style="list-style-type: none"> 1. Demonstrate the range of environmental conditions in a deep geological repository (DGR) under which microbes can be reasonably expected to be non-culturable (i.e., inactive) 2. Determine the nature and maximum extent of corrosion damage to the UFC if microbes were to be active in the DGR 	H
2	Corrosion During the Unsaturated Period	Assess the importance of the unsaturated period in terms of the extent and nature of corrosion damage, in particular the extent to which corrosion is localized as a result of non-uniform wetting of the container surface	H
3	Evolution of Corrosion Damage	Describe how the nature of the corrosion damage evolves as the environment evolves from initially warm and aerobic to eventually cool and anaerobic	H

(b) Copper UFC Research Areas

	Program	Aim	Priority
1	Stress Corrosion Cracking of Copper	Define under what conditions a copper used fuel container (UFC) might be susceptible to cracking and to determine the impact on container lifetimes	M
2	Corrosion of Copper Welds	Determine the corrosion behaviour of welded regions on the container surface and to establish whether these areas are more susceptible to corrosion	M
3	Uniform Corrosion of Copper	Investigate the mechanism of the uniform corrosion of copper and develop the necessary database to enable mixed-potential modelling of the corrosion potential of copper UFC	L
4	Lifetime Prediction for Copper Containers	Develop mechanistically based corrosion models to make justifiable predictions of the long-term corrosion behaviour of copper UFC	L
5	Gas Generation from Failed Copper Containers	Determine the impact of gas generation from a failed copper/carbon-steel UFC on the safety and performance of the repository	L

Table 18: Overall Summary of Proposed Research Programs, Their Aims, and Prioritization (continued).

(c) Carbon-steel UFC Research Programs

	Program	Aim	Priority
1	Stress Corrosion Cracking of Carbon Steel	Determine the impact of stress corrosion cracking (SCC) on the lifetime of a carbon-steel (C-steel) UFC	M
2	Corrosion of Carbon Steel Welds	Determine whether welding of C-steel UFC leads to an increase in the rate of uniform corrosion or an increased susceptibility to SCC or hydrogen-related degradation mechanisms	L/M
3	Uniform Corrosion of Carbon Steel	Develop an understanding of the mechanisms of corrosion under both aerobic and anaerobic conditions and develop input data for predictive models	H
4	Lifetime Prediction of Carbon Steel Containers	Develop modelling capability to make long-term, justifiable predictions of the lifetimes of C-steel UFC	L
5	Gas Generation from Carbon Steel Containers	Assess the impact of hydrogen gas (H ₂) generated by corrosion on the safety of the repository	H
6	Hydrogen-related Degradation Mechanisms	Assess the impact of absorbed hydrogen on the lifetime of C-steel UFC as a result of various hydrogen-related degradation mechanisms	M/H

