ASSESSING THE CORROSION PERFORMANCE OF HIGH-LEVEL NUCLEAR WASTE CONTAINERS

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ABSTRACT

The development of models to assess the corrosion performance of containers for the disposal of high-level nuclear waste has been underway for over 20 years. A review of the rationale behind container design and materials selection, and the bases for the development of corrosion models is discussed.

INTRODUCTION

Methods of disposing of nuclear wastes have been under study for over twenty years. Various geologic formations, supplemented by a series of engineered barriers, have been studied. These include, tuff rock (USA)[1], salt domes (Germany)[2], sedimentary clay deposits (Belgium, Switzerland) [3], and granitic rock sites (Sweden/Canada/Finland)[4]. This multi barrier approach reduces the reliance on any single barrier and compensates for what will inevitably be significant uncertainties in predicted performance.

Regulatory release limits make it necessary to achieve containment for thousands to tens of thousands of years. Within the sequence of barriers, the relative importance of the waste container depends on the uncertainties inherent in the ability of the geologic formation to retard radionuclide transport once groundwater gains access to the wasteform within the failed waste container. It is the only absolute barrier within the system, and its performance can be, at least partially, controlled by the selection of the container material.

In this paper, a number of key features of waste containers are discussed, with emphasis on the container corrosion performance.

WASTE CONTAINER DESIGN

Besides corrosion performance, many other requirements of container design must be taken into consideration. These include; (i) avoidance of nuclear criticality (i.e., a sustained nuclear fuel reaction); (ii) provision of sufficient radiation shielding to ensure safe handling; (iii) ease of fabrication, inspection and welding, since these processes must be performed remotely due to the radiation fields present on the container surface; (iv) sufficient mechanical robustness. The required level of mechanical robustness depends on the location of the waste repository, the rigors imposed by the container emplacement process, and the conditions that prevail once the repository is sealed. Thus, for waste packages emplaced in open excavated drifts within Yucca Mountain (NV, USA), there is the potential for rock fall damage. Containers emplaced in back-filled granitic rock repositories excavated below the water table must be able to withstand hydrostatic pressures as well as the swelling pressure imposed by the self-sealing clays surrounding them. Also, for Northern locations (Canada, Sweden, Finland), in view of the long lifetimes required, it is judicious to fabricate the container with sufficient structural strength to withstand the additional pressure due to the passage over the waste vault of an ice sheet.

For these mechanical reasons, waste containers are dual-walled with an inner wall for structural strength and an outer wall for corrosion resistance. Thus, for sealed granitic environments (Canada, Sweden, Finland) the proposed container will be fabricated with an outer corrosion barrier of 2.54 to 5.0 cm-thick copper internally supported by a cast iron insert [5-8]. The proposed waste package design for the open tuff Yucca Mountain site (USA) has an outer Ni-Cr-Mo alloy corrosion barrier and an inner stainless steel shell [9].

WASTE REPOSITORY CONDITIONS

Any chosen geological formation must possess some clearly identifiable advantageous features, which must not be destroyed by the thermal/chemical transient caused by the excavation/waste emplacement process. Also, the evolution of the repository from the initially disturbed to the final steady-state condition must be predictable with reasonable certainty. Examples of advantageous features include; (i) the arid conditions within Yucca Mountain (NV, USA) which limit the water available to cause container corrosion and to transport radionuclides from the failed container [9]; (ii) the anoxic conditions and extremely low water transport rate in crystalline rock [4]; (iii) the extremely effective retention of radionuclides and the self-sealing plasticity of sedimentary clays [10].

Figure 1 attempts to compare corrosivities of a number of repository environments. The ranges of temperature are from the maximum anticipated (after tens of years of waste emplacement) to that calculated (based on radionuclide decay rates) after $\sim 10^3$ years. Temperatures are adjustable depending on repository design features, such as waste container loadings and container spacing, and the values used in Figure 1 are illustrative rather than definitive. Chloride concentrations are from ground-water analyses. This figure provides only a very general comparison of repository features, and beyond the influence of temperature, does not incorporate the environmental consequences of excavation/emplacement. These disturbances will lead to short term changes specific to individual repositories. Thus, in Yucca Mountain, the periodic contact of dilute, low salinity groundwaters with hot container surfaces could lead to evaporative processes and the development of high ionic strength aqueous environments (up to 10^5 mg/L) possibly at temperatures well above 100^oC. Depending on anionic composition, extreme pH values (≥ 12) could be achieved [9]. To avoid contact of these environments with the waste package, an additional drip shield barrier is included in the repository design. In crystalline rock repositories, oxygen present on sealing, must be consumed before the original anoxic conditions are re-established [11]. In sedimentary clays, oxidation of pyrite (by introduced O_2) will produce corrosive species such as thiosulphate ($S_2O_3^{2-}$) and sulphate, which could either inhibit chloride-induced localized corrosion or react with sulphate-reducing bacteria to initiate MIC [12].

The anticipated evolution in conditions are shown in Figure 2 for a Canadian repository proposed by AECL [11, 13] and, in Figure 3 for the Yucca Mountain repository [9]. The distributions in parameters show the influence of waste loading and container location with respect to other heat-producing containers. Again, the values shown should be considered illustrative rather than definitive. In Figure 2, the oxygen profile is arbitrary in shape but fixed by two limits: 50 years, the calculated time for all O_2 to be consumed by organic material present in the backfill materials; and 700 years, the time for O_2 to be consumed by Fe^{II}, present in the backfill as the mineral biotite [14]. This definition of the evolution of waste vault/repository parameters, Figures 2 and 3, is an essential first step in the assessment of the long term performance of waste containers, since it defines the initial period, when the exposure environment is disturbed and potentially aggressive, and its relaxation to eventually more benign conditions.

MATERIALS SELECTION

Figure 4 shows the expected corrosion behaviour of different categories of materials [15-18], and can be used to illustrate why specific materials are preferred in different environments. The dark shaded areas show the corrosion potential region within which the specified corrosion process could occur. In interpreting Figure 4, the corrosion potential should not be confused with the redox potential of the exposure environment. The corrosion potential is a measure of that specific materials corrosion response to the environment. The two zones marked at the bottom of the figure are the anticipated environmental redox conditions. Corrosion is a possibility if the corrosion potential of the material overlaps the redox potential range of the environment.

The prevalence of oxidizing conditions at Yucca Mountain dictates the selection of a passive material from one of two categories; Ni alloys (corrosion resistant except under extreme oxidizing conditions) or Ti alloys (corrosion resistant except under extreme reducing conditions). The present uncertainties over the possibility of concentrated environments due to evaporative processes make it judicious to chose extremely resistive alloys such as Alloy-22 (22 wt% Cr, 13 Mo, 3W) and Grade-7 titanium (0.2 Pd). Further studies leading to the elimination of the possibilities of extreme environments could allow the eventual selection of cheaper materials. In salt repositories, especially when acidic Mg^{2+}-containing brines could occur, even the Ni-Cr-Mo alloys may prove unable to maintain passivity, although Ti alloys appear to do so [2].

For anoxic repositories, materials such as carbon steels and copper are primary candidates. The preference for copper (the primary candidate for crystalline rock repositories (Sweden/Finland/Canada)) is based on its ability to achieve thermodynamic stability once O_2, introduced during excavation/emplacement, is consumed. The attraction of iron and carbon steels is their low cost and extensive corrosion history. However, information suggests that the use of steels in dual-walled containers with passive materials (i.e. for the Alloy 22 package proposed for Yucca Mountain) may have more disadvantages than advantages [19]. Under oxidizing conditions, the production of acidic ferric ion solutions could lead to corrosion of the corrosion-resistant second wall, and the formation of voluminous corrosion products could cause deformations and/or assist SCC.

PREDICTING CONTAINER CORROSION PERFORMANCE

While assessment of corrosion performance must be conducted for each material proposed, it is possible to draw a few general conclusions based on environmental evolutions such as those illustrated in Figures 2 and 3. An attempt to categorize the general evolution with time of a series of potential degradation mechanisms is shown schematically in Figure 5.

(a) Localized Corrosion

The probability of localized corrosion (pitting, crevice corrosion) will be greatest during the early disturbed period, when temperatures are high and environments concentrated, Figure 5. As conditions cool and environments become less aggressive, propagation rates should fall and the probability of stifling/repassivation should increase. Considerable effort has been expended to determine susceptibilities to localized corrosion [12, 20], and the value of parameters such as repassivation potentials, critical solution concentrations to stabilize pits and crevices, and critical pit dimensions, have been emphasized [21]. However, because of the time scales and the uncertainties in exposure environments it is difficult to claim immunity, especially for crevice corrosion. Alternatively, performance assessment model must show that the damage sustained will not significantly affect long-term performance.

This requires the development of propagation models. The propagation law, $d = kt^n$, is well established, where d is the maximum penetration depth, k a growth constant, and n a parameter which empirically incorporates environmental and materials factors leading to stifling. For the Yucca Mountain project, its use, coupled to a Monte Carlo approach to uncertainties, has shown that values of $n < 0.3$ are necessary to claim crevice/underdeposit corrosion will not significantly influence the predicted lifetimes of Alloy-22 waste containers within Yucca Mountain [22]. This value is less than the theoretical value of 0.5 for propagation controlled by diffusive or voltage drop, and illustrates the need to show that metallurgical factors will inhibit maintenance of critical solution chemistry, thereby inducing repassivation.

Such an analysis also clearly identifies whether or not the inclusion of an additional barrier, such as the drip shield in Yucca Mountain, is essential to overall waste package/container performance. If laboratory studies show that the propagation of crevice/underdeposit corrosion is potentially rapid and deeply penetrating (i.e., $n > 0.3$ in the propagation law), then the Yucca Mountain Alloy-22 waste package would need protection by the inclusion of a drip shield to allow the environment to evolve to less aggressive conditions before contacting the waste package. If $n < 0.3$, then the use of a drip shield for corrosion protection of the waste package may not be essential. Consequently, while its inclusion to avoid rock fall damage may always be necessary, the choice of a stronger, less corrosion resistant and cheaper material would be an option.

While titanium is not the preferred waste container material under anticipated Canadian conditions, it has been extensively studied. A similar model to that described above for Alloy-22 shows that, given the limited oxygen trapped within a sealed granitic repository, a Grade-2 titanium container, emplaced in a borehole and surrounded by sand, clay buffer and clay/rock backfill, Figure 6, would not fail by crevice propagation [23]. Extrapolation of conservative d_{max} values as a function of available O_2 predicts a maximum penetration of about 3mm (of an available wall thickness of 6.5 mm) [24], depending on the availability of O_2 within the borehole (i.e., all available or only that in the sand layer, Figure 6. Since most of the O_2 will react with oxidizable minerals in the clay, this is a conservative calculation. However, the form of the damage function ($d = kt^n$) means a less conservative assumption would have little impact on the predicted penetration, since most of the damage is incurred rapidly at short times. Since the database is sparse (Figure 7) one may subsequently chose to reduce uncertainties and increase safety margins by selecting a more crevice corrosion resistant titanium alloy [13].

When a database of penetration depths is available, it is possible to use an extreme value statistical treatment to calculate the cumulative probability of the deepest penetration site leading to failure of the container. This approach has been used to predict a maximum pit depth of 6 mm after 10^6 years on a Canadian copper container, and is based on a set of penetration depths from an NBS study of buried Cu-alloy pipes [25,26] and a Swedish study of archaelogical bronzes [27]. Since the artifacts are 3000 years old, and the database is extensive, reasonable confidence in the predictions is merited [16].

(b) Stress Corrosion Cracking

SCC is of particular important in container closure welds, where stresses are inevitable. Since stress mitigation procedures must be applied remotely, due to radiation fields, considerable development is necessary to demonstrate they will be successful. Like localized corrosion, SCC is most likely during the early disturbed period and its probability of occurrence will decrease with time as temperatures fall and exposure environments become less aggressive, Figures 2 and 3.

As for localized corrosion, efforts are underway to demonstrate that SCC will not occur. For Yucca Mountain, this involves measurement and calculation (using finite element models) of critical stress intensity factors based on the number density and alignment of flaws within the welds [9]. A key strategy is to demonstrate that stress relief procedures (possibly induction coil heating/laser peening) will produce a thick compressive surface layer, whose corrosion will be sufficiently slow that, by the time development of tensile stresses are feasible, the environment will be too benign to sustain SCC [9,22].

Similarly, for SCC on copper containers (Canada/Sweden/Finland), it is argued that the evolution of the environment and the development of stresses cannot simultaneously produce the required combination of conditions [28]. The potential cracking agents, NO_2^-, from moist air radiolysis, and NH_3, NO_2^-, and organic acids from MIC, are formed at short and long times, respectively, and a short early period of creep should lead to the relief of the hydrostatic and buffer swelling stresses which develop when the local environment becomes saturated, Figure 8. Modeling of crack growth rates is also underway using the film rupture/anodic dissolution model originally developed for stainless steels under operating nuclear reactor conditions [29]. As for localized corrosion, the database to apply such a model to waste containers is sparse.

In comparison to localized corrosion, there is one additional difficulty in predicting the extent of SCC using performance assessment models. For localized corrosion, evidence exists to demonstrate that the slowing of propagation with time will occur, and theories based on voltage drop and diffusion effects support this evidence. However, in the case of SCC, the increasing crack tip stress intensity as crack growth continues complicates arguments that crack arrest, like crevice repassivation, will be probable. Consequently, crack growth rate models rely on the rate decreasing with time to an insignificant level, a feature that will be very difficult to validate experimentally.

(c) Microbially-Induced Corrosion (MIC)

The possible effects of MIC will be awkward to assess since it will be difficult to predict how microbial activity will adapt over repository time periods. Initially, the combined effects of elevated temperatures, gamma radiation, and in particular, a lack of water, will severely limit microbial activity close to the container [30]. For heat-producing containers in compacted clays (Canada/Sweden/Finland) it has been clearly demonstrated that these factors will lead to a microbially-depleted zone. As temperatures fall, radiation fields decay, and the availability of water increases, microbial activity becomes more probable.

To date, few attempts have been made to model MIC under repository conditions [31]. One model [31], for Cu containers within compacted clays, assumes the mean pore size within the clay (0.1 to 0.5 µm) will seriously impede the transport of microbes and nutrients. Thus, once the early dessicated period is over and water re-saturates the clay, microbes will not repopulate the depleted zone to form a biofilm on the container.

However, microbial activity could revive at remote locations, and the inorganic species produced could then be transported to the container surface. For copper, the most dangerous scenario would be the action of SRBs to produce S^{2-}. Reaction between Cu and S^{2-} to produce surface sulphide layers could then catalyze water reduction and Cu corrosion, Figure 9. The key modeling parameters then become the SO_4^{2-} content of the buffer, the level of microbial activity and the transport rate of S^{2-} to the Cu surface. Fortunately, transport rates of species in compacted buffers are ~ 10^2 times less than in open solution. Recently, a more detailed model for MIC under gramitic conditions has been developed [32].

For passive materials, the long term prospects for supporting MIC are much less likely. While the possibility of microbial activity may increase with time, the probability that pitting or crevice corrosion would initiate and propagate would decrease. This is especially so for the extremely resistant passive alloys, Alloy 22 and Grade-7 titanium [33] chosen for the Yucca Mountain Repository.

(d) Hydrogen-Induced Cracking (HIC)

A second process potentially more dangerous at long times is HIC. This process requires the accumulation of sufficient hydrogen within the container to significantly reduce its fracture toughness. As indicated in Figure 4 such a process is only anticipated for materials such as titanium.This process has been modeled for titanium alloys when hydrogen absorption occurs initially due to crevice corrosion (disturbed period) but eventually due to passive corrosion once crevice propagation ceases when anoxic conditions prevail. During crevice corrosion, hydrogen absorption could be rapid since active corrosion occurs under acidic conditions. However, passive corrosion will be slow, and H absorption inefficient, through the low permeability passive film. Failure is assumed to occur once a criticial hydrogen concentration (established by mechanical testing) is achieved. A detailed description of this model has been given elsewhere [34].

(e) General Corrosion

If failure by localized corrosion can be ruled out, then eventual failure will be by general corrosion. Intuitively, this process should be relative simple to model. For actively corroding materials this may be so, but, for passive materials, very little is known about the long term evolution of the passive state, making validation based on short term experimental observations difficult.

For active materials such as copper and carbon steels, predictions have been based on deterministic mixed potential models [28,35,36]. The model for copper [28] couples the interfacial electrochemical processes to the precipitation/dissolution, redox, adsorption/desorption and mass transport processes that will occur in the surrounding buffer and backfill materials. A one dimensional, four layer, spatial grid is used to describe the repository, consisting of buffer (clay), backfill (clay/rock), an excavation damaged zone (EDZ) on the surface of the rock, and a region of intact (sparsely fractured) rock. One boundary is the container, the other a major, water-carrying fracture assumed present in the rock, Figure 10. These layers are defined in terms of their diffusivities, adsorptive properties, and redox capacity. A numerical solution is obtained using finite-difference methods. Application of this model yields the evolution in corrosion rate with time, which on integration yields the extent of wall penetration; ~ 11 µm by the time all the oxygen is consumed and general corrosion stops.

For passively corroding systems, such as Alloy-22 and titanium Grade-7 under Yucca Mountain conditions, present models assume a constant corrosion rate and account for uncertainties using Monte Carlo sampling of a large database of rates fitted to a Weibull distribution [9,22,37]. Since these rates are only tens of nm/year, extremely long lifetimes of 10^4 to 10^6 are predicted. Presently, a model is under development based on mixed potential principles in which the growth/dissolution of the oxide is modeled using the point defect model.

However, considerable uncertainty remains in these predictions since the long term evolution of passive film behaviour is unknown. A number of possible long term scenarios by which passive corrosion rates could exceed those measured in short term tests have recently been discussed by various review panels [38,39]. Possible processes include; (i) the accumulation of damage from metastable passivity breakdown events, which could be exacerbated by long term aging effects leading to the precipitation of intermetallic compounds; (ii) the surface segregation of minor impurities such as S, which could undermine passivity; (iii) long term ennoblement which could push the corrosion potential into the transpassive region.

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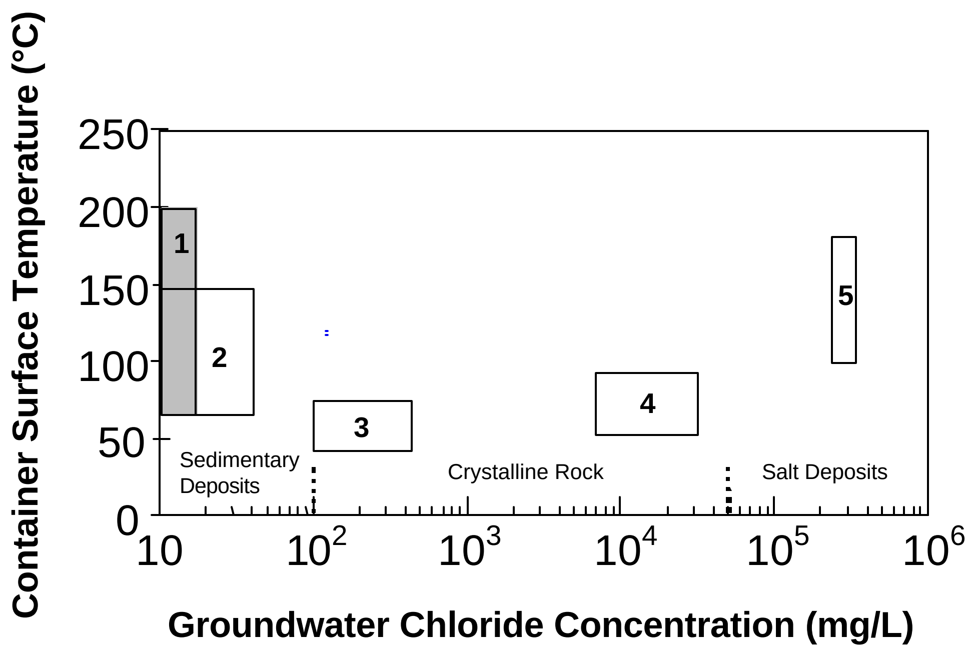

Figure 1; Corrosivities of various proposed waste repositories: (1) tuff (USA); (2) clay (Belgium); Granite (Sweden); (4) granite (Canada); (5) salt (Germany).

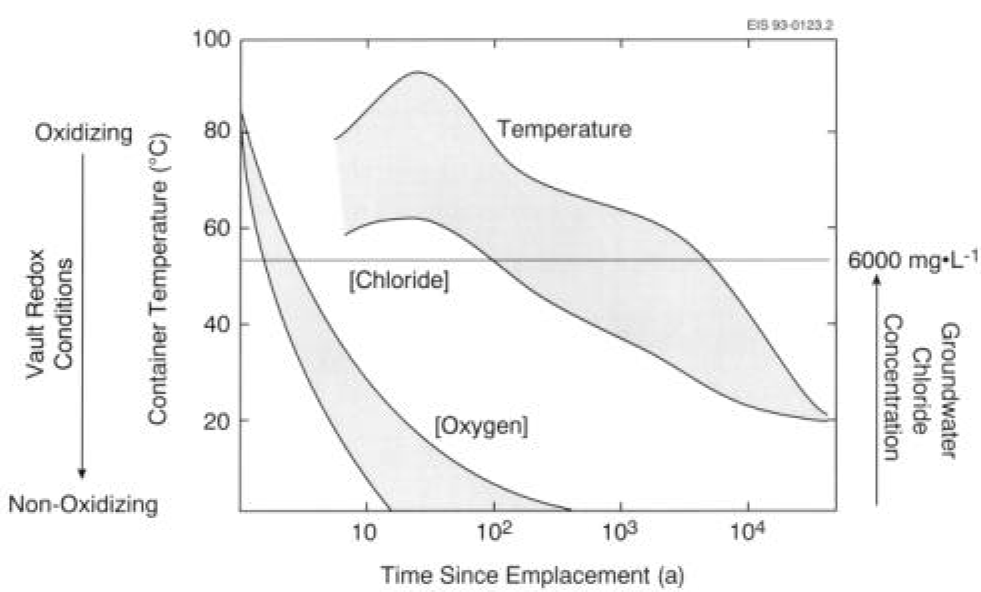

Figure 2: Anticipated evolution of the key environmental parameters within a Canadian waste repository.

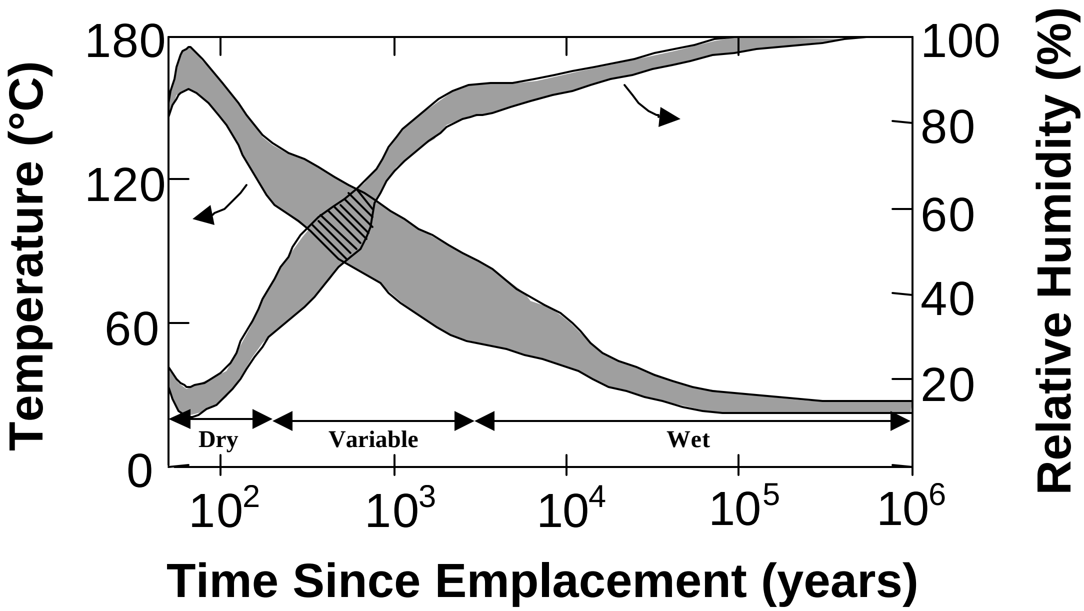

Figure 3; Anticipated evolution of key parameters within the Yucca mountain (USA) repository for the high temperature mode [6]

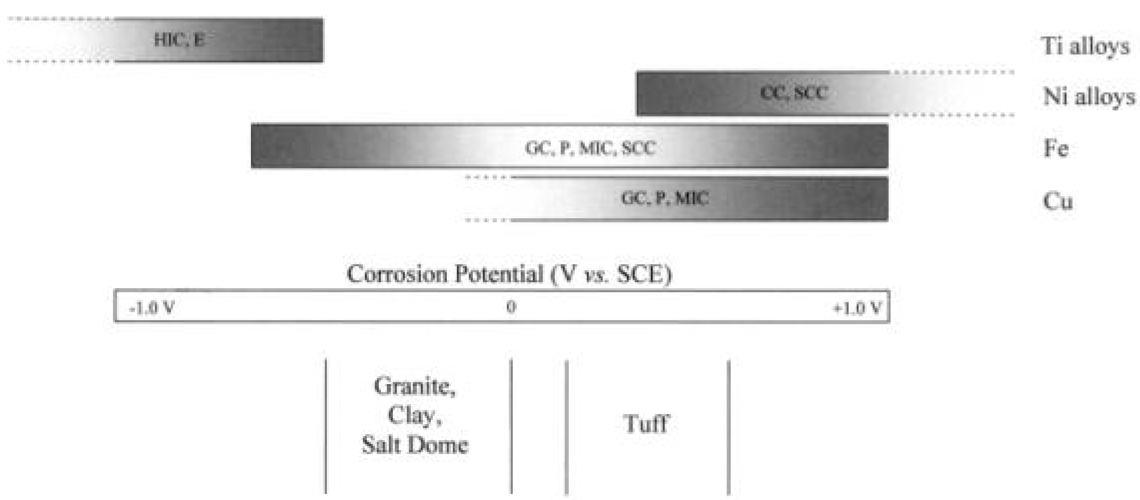

Figure 4: Anticipated corrosion behaviour of various materials as a function of redox conditions (expressed as a corrosion potential); P – pitting; MIC – Microbially-induced corrosion; SCC –

Stress corrosion cracking; CC – Crevice corrosion; HIC – Hydrogen-Induced Cracking; GC – General Corrosion; E - Embrittlement

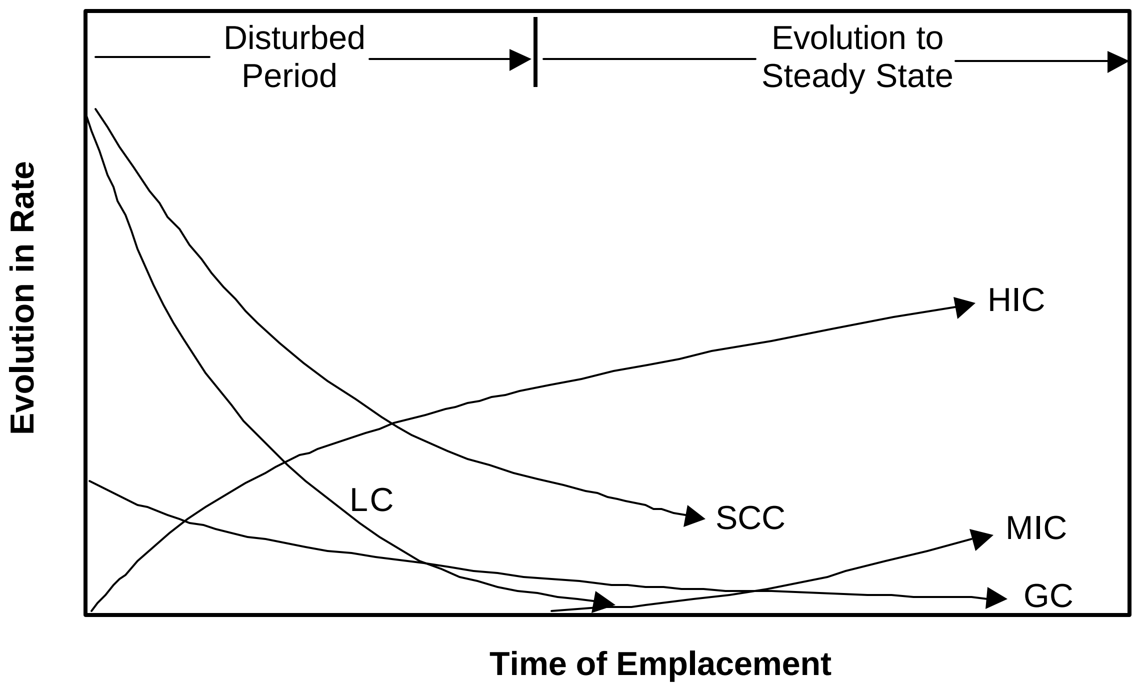

Figure 5; Anticipated evolution in importance of a number of container corrosion modes as repository conditions evolve.

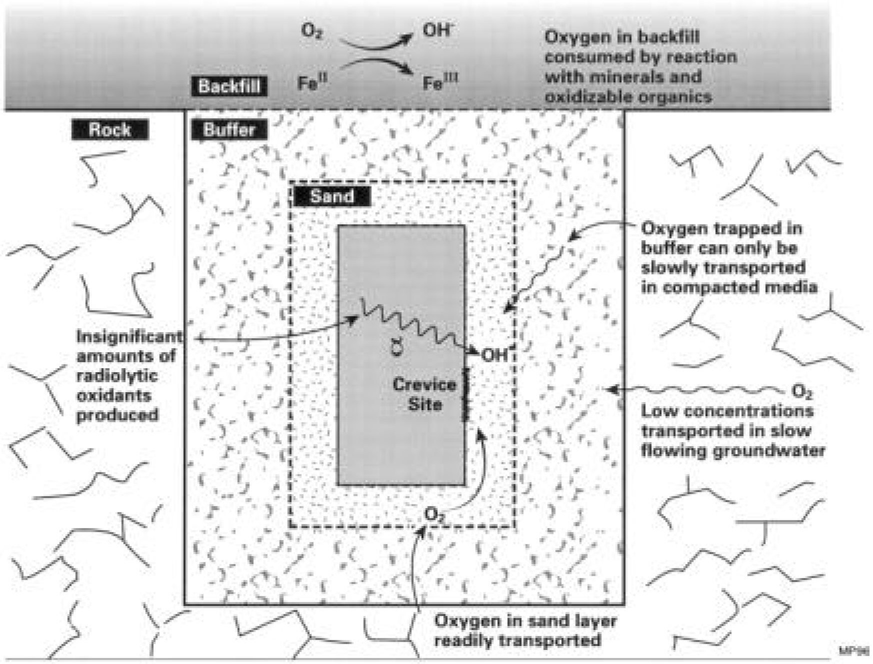

Figure 6; Schematic showing the sources of O_2 potentially available to drive crevice corrosion of Ti-2 in a borehole in the floor of a Canadian repository.

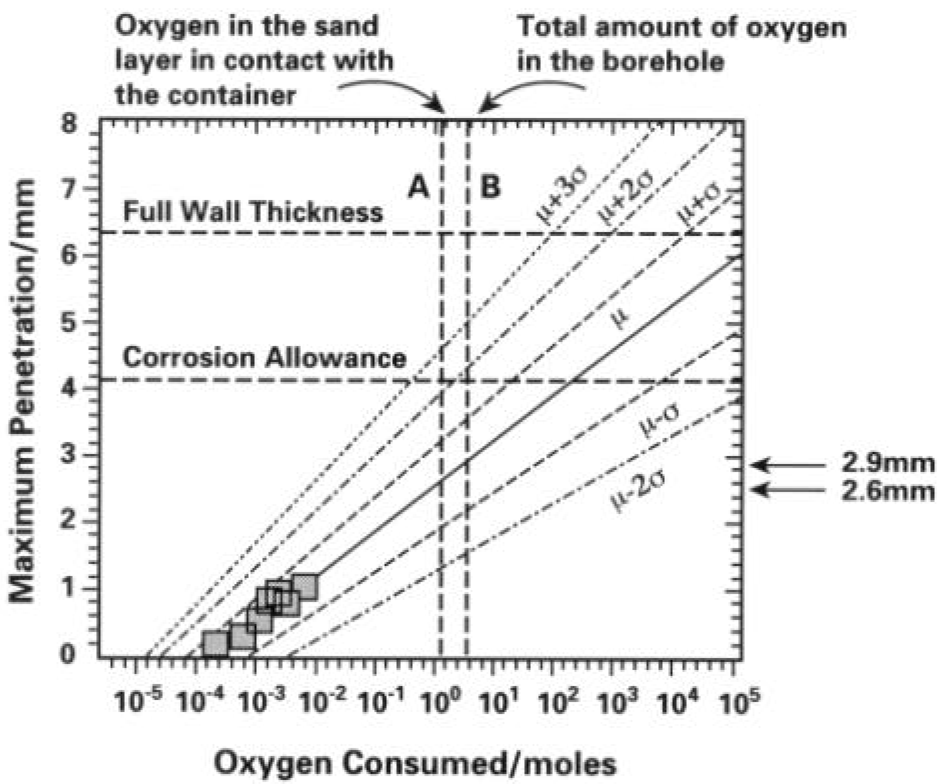

Figure 7; Extrapolated damage function to predict the maximum depth of crevice corrosion on Ti-2 as a function of available O_2 in a borehole in the floor of a Canadian repository.

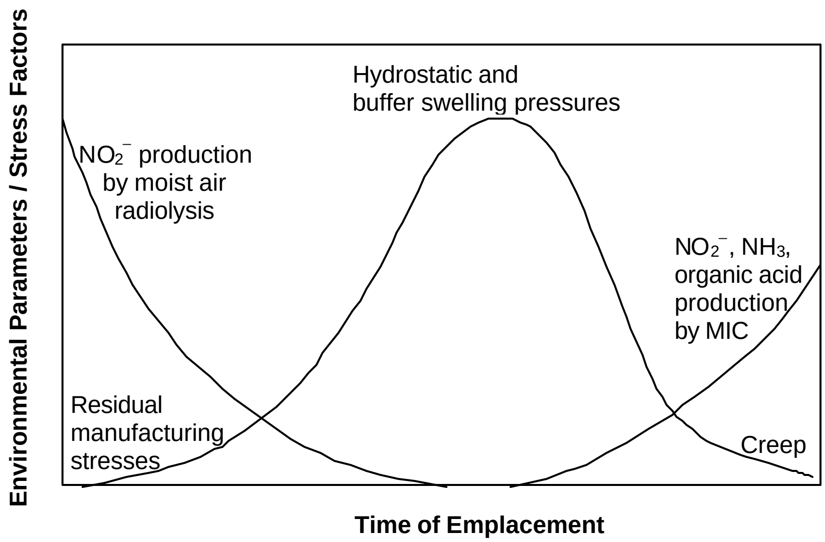

Figure 8; Schematic illustrating the non-coincidence of the prerequisites for SCC of Cu containers under Canadian repository conditions.

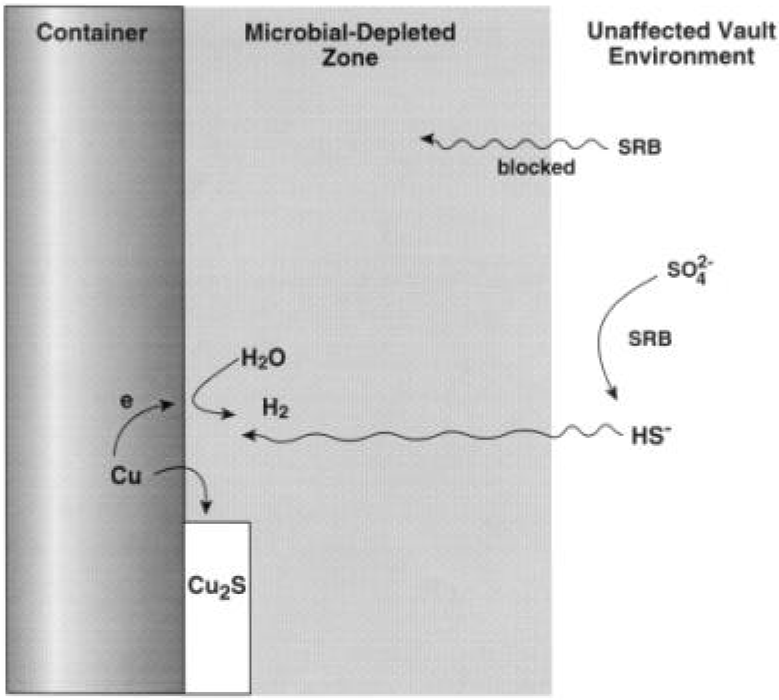

Figure 9; Schematic illustrating the possible long term effects of sulphate reducing bacteria on MIC of Cu containers under repository conditions

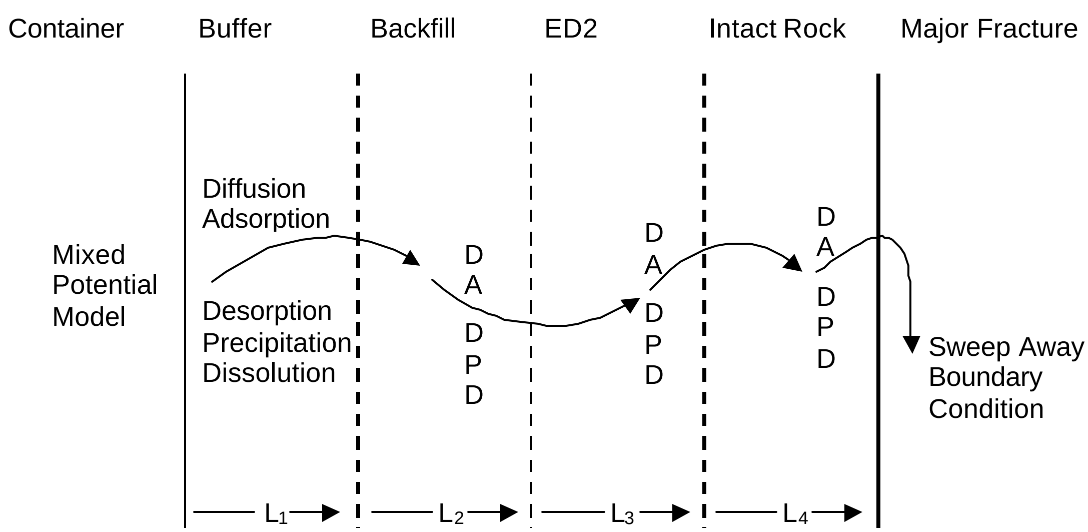

Figure 10; Simplified one-dimensional representation of the various mass transport barriers involved in the general corrosion of Cu in a Canadian repository [25]; L_n are the barrier lengths. The important processes within each barrier are indicated.