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U.S. Nuclear Regulatory Commission
ATTN: Mrs. Deborah A. DeMarco
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Subject: Programmatic review of paper titled "Life Prediction of Container Materials for High-Level Nuclear Waste Repository" for presentation at the Asian and Oceanic Congress for Radiation Protection Conference

Dear Mrs. DeMarco:

The paper "Life Prediction of Container Materials for High-Level Nuclear Waste Repository" is being submitted for NRC programmatic review. NRC Form 390A is also attached. This paper was invited for presentation at the First Asian and Oceanic Congress for Radiation Protection, October 20–24, 2002 in Seoul, Korea.

The paper provides a brief summary of the NRC and CNWRA work related to the evaluation of container life prediction. The paper presents a broad cross-section of activities in this area that have been reported elsewhere and summarizes the approach taken by NRC in evaluating container life issues.

Please advise me of the results of your programmatic review. Your cooperation in this matter is appreciated.

Sincerely,

Budhi Sagar
Technical Director

Enclosure

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2. AUTHOR(s)

C S. Brossia, G A. Cragnolino, D.S. Dunn, O. Pensado, N Sridhar, and L T. Yang

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Life Prediction of Container Materials for High-Level Nuclear Waste Repository

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ABSTRACT

This paper focuses on the methodology for evaluating the corrosion performance of high-level waste container materials in the U.S. repository program. The performance of the container material depends critically on the environment contacting it. The composition of the environment contacting the container is determined by evaporation, rewetting, and radiolysis. The time or relative humidity at which condensation of water occurs depends on the dissolved salt concentration. The paper reports recent work related to the measurement of deliquescence point as a function of salt concentration and the use of repassivation and corrosion potentials in determining the conditions for the onset of localized corrosion. The repassivation and corrosion potentials are shown to be functions of solution composition, temperature, and microstructure. The passive dissolution rate is measured as a function of environmental conditions. Mechanistic modeling of passive dissolution is discussed to increase the confidence in performance estimate. Calculations of the failure time of the containers using the EBSFAIL module of the NRC/CNWRA Total-system Performance Assessment (TPA) code are presented.

Key Words: High-level waste repository, corrosion, repassivation potential, container life

INTRODUCTION

In the United States, the Department of Energy (DOE) is responsible for the design and construction of a potential geological repository for the safe disposal of the spent fuel (SF) from nuclear reactors and high-level waste (HLW) from reprocessing operations. If any DOE license application is submitted, the NRC would evaluate it for compliance with the regulatory requirements for the proposed repository at Yucca Mountain, Nevada, contained in the U.S. Code of Federal Regulations (10 CFR Part 63. The current DOE design of the waste package (WP)[1] consists of a 20-mm [0.787 in] thick Alloy 22 (22Cr-13.5Mo-3W-3Fe-balance Ni) outer container surrounding a 50-mm [1.969 in] thick type 316 nuclear grade (NG) stainless steel inner container to provide structural strength only.

For repository conditions that are expected to be reducing, container materials that can provide thermodynamic immunity, such as copper, have been considered. However, the overall redox condition of the environment at the proposed U.S. repository at Yucca Mountain, Nevada is anticipated to be oxidizing.[2] Under such conditions, copper or steel will not provide the long container life desired. Carbon steel was thought to corrode in a predictable, uniform mode, but under certain pH and potential conditions, may suffer localized corrosion.[3] The U.S. Department of Energy has chosen alloys containing chromium among other alloying elements that may provide long life through their low kinetics of corrosion. Low corrosion rates of these alloys result from the presence of a protective oxide film, called passive film. However, under some conditions, the passive film can be breached locally and the ensuing localized corrosion rates or stress corrosion cracking can lead to premature release of radionuclides.

ENVIRONMENT CONTACTING THE CONTAINERS

The repository in the U.S. is proposed to be sited in the unsaturated zone of Yucca Mountain, thus creating nominally dry conditions. For a scenario that assumes 50-year ventilation, followed by permanent closure, the calculated temperature on the surface of the WP and drift wall are shown in Figure 1. This figure represents one of the many design options considered by the DOE. In the first 50 years prior to closure, the repository is considered to be ventilated. After the repository is closed, the temperature initially rises, reaching a peak at about 100 years, followed by a gradual cooling as the radioactivity continues to decay. The predicted relative humidity is shown in Figure 2, along with a critical relative humidity above which water will condense as an aqueous layer on a given surface. The critical relative humidity is a function of container surface condition. Presence of hygroscopic salts or corrosion products can reduce the critical relative humidity and thus promote aqueous corrosion at an earlier time in the repository. It must be noted that certain iron corrosion products (e.g , iron chloride) are especially hygroscopic and corrosion may occur at relative humidity values below about 40 percent.

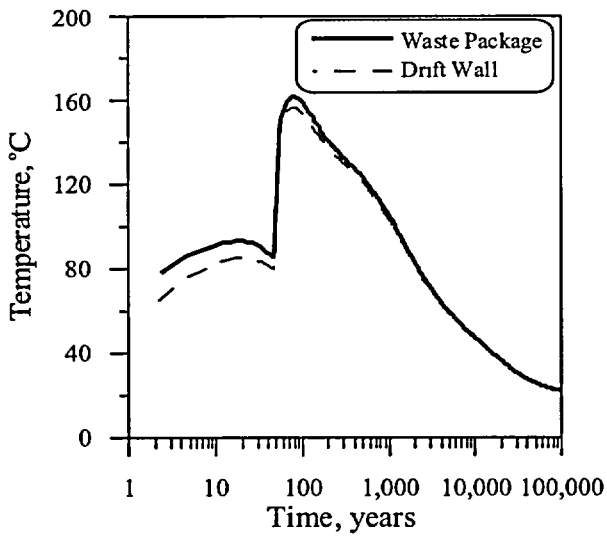

Figure 1. Predicted temperature at two different locations.[13]

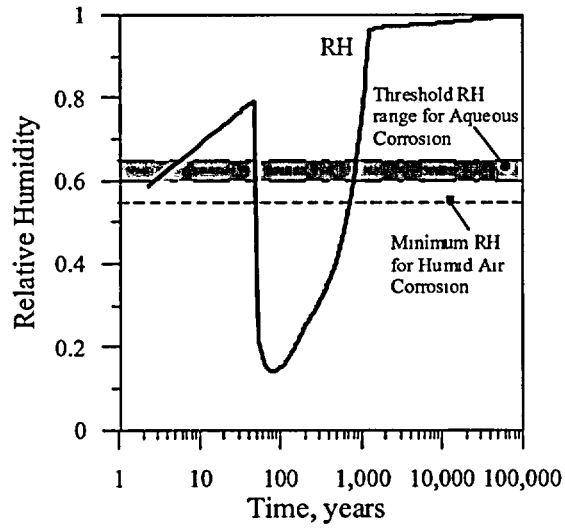

Figure 2. Calculated relative humidity at the drift wall and critical relative humidity.[13]

Deposition of aerosol and dust from ventilation air and the evaporation of water seeping into the drift may cause accumulation of hygroscopic salt on the drip shield and waste package surfaces.[4-6] Thus, the accumulated salt may be mixtures of different salts. Figure 3 shows that the experimental values of deliquescence relative humidity (DRH) of salt mixtures containing Na^+, K^+, NO_3^- and Cl^- are significantly lower than those of the single salt components. At 86 °C, the lowest DRH among the single salts is 65% (for $NaNO_3$), whereas the DRH of the $NaCl+NaNO_3+KNO_3$ mixture is only 43%. Therefore, when two or more salts are present, the DRH of the salt mixture should be used, as the critical relative humidity for aqueous corrosion in model analysis.

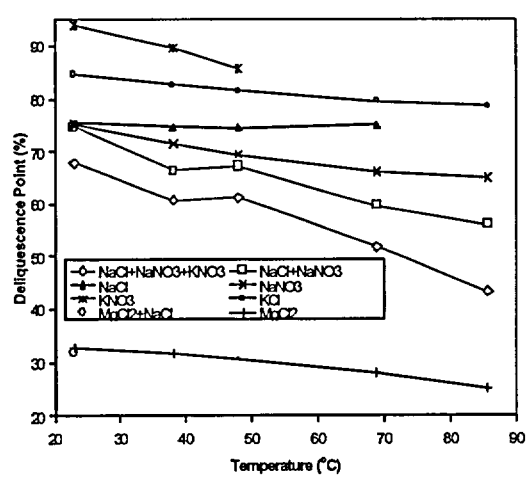

Figure 3. Comparison between the deliquescence relative humidities of pure and mixed salts.[7]

LOCALIZED CORROSION PREDICTION

The most important forms of localized corrosion are pitting and crevice corrosion. It is increasingly believed, at least for the chromium-containing alloys, that the fundamental mechanisms for pitting and crevice corrosion

are the same.[8,9] However, crevice corrosion can be initiated more easily than pits. The parameters that determine whether localized corrosion will occur for a given material-environment combination are the repassivation and corrosion potentials. If the corrosion potential, E_{corr}, exceed the repassivation potential, E_{rp} or E_{rcrev}, localized corrosion is initiated and stabilized. The repassivation potential depends on the alloy composition and the presence of aggressive and inhibiting ionic species. The E_{rp} represents the repassivation potential for pitting, whereas E_{rcrev} represents the repassivation potential for crevice corrosion. Previous work[9] has shown that these potentials are identical for deep pits. The corrosion potential depends on the presence of oxidizing species, as well as the pH and temperature. It also depends on the class of material (e.g. carbon steel), but within a class of material it is not highly sensitive to alloy composition. The effect of environmental conditions on localized corrosion differs for various alloy systems. Two types of metallic materials will be discussed in this paper.

Stainless steels and Nickel-based Alloys

The stainless steels and nickel-base alloys considered by DOE vary in terms of their nickel, chromium, molybdenum, and tungsten contents as well as minor impurities such as carbon. Generally, the higher the Cr, Mo, and W contents, the higher the resistance of the alloy to localized corrosion. The localized corrosion resistance of a number of alloys, including Alloy 22 the current candidate material, to chloride containing solutions is shown in Figure 4. It can be seen that (i) for a given chloride concentration, the repassivation potentials of the alloys are arranged in the order of increasing resistance to localized corrosion as Type 316L SS> Alloy 825 > Alloy 625 > Alloy 22 and; (ii) at the highest chloride concentration studied, all the alloys have similar repassivation potentials. When no localized corrosion occurs, the repassivation potential has no relevance to localized corrosion, but represents a combination of oxygen evolution and transpassive dissolution. The presence of nitrate has a significant inhibitive effect on localized corrosion (Figure 5). The nitrate to chloride ratio needed for inhibition of Alloy 22 at 95°C is approximately 0.2. It is possible that evaporative concentration of the groundwater near the waste containers may result in an increase in both chloride and nitrate. It must be noted that nitrate inhibits crevice corrosion even when it is added after active crevice corrosion has taken place. An increase in temperature results in a reduction in crevice repassivation potential (Figure 6). It is known[10] that in 6 weight percent $FeCl_3$ solution, Alloy 22 has a critical crevice temperature of about 55°C, which means that the alloy will suffer crevice corrosion at temperatures above this value. The data in Figure 6 is consistent with this independent information because the corrosion potential in $FeCl_3$ is approximately 600 mV_{SCE}, which is higher than the repassivation potential at 80°C for a 1.1 molar chloride solution.

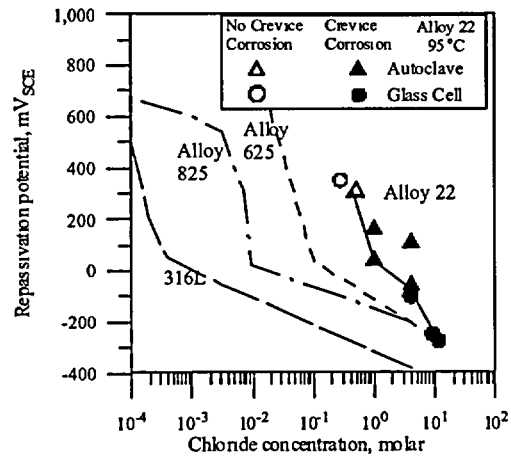

Figure 4. Repassivation potentials of a number of Ni-Fe-Cr-Mo alloys in chloride environments.[2]

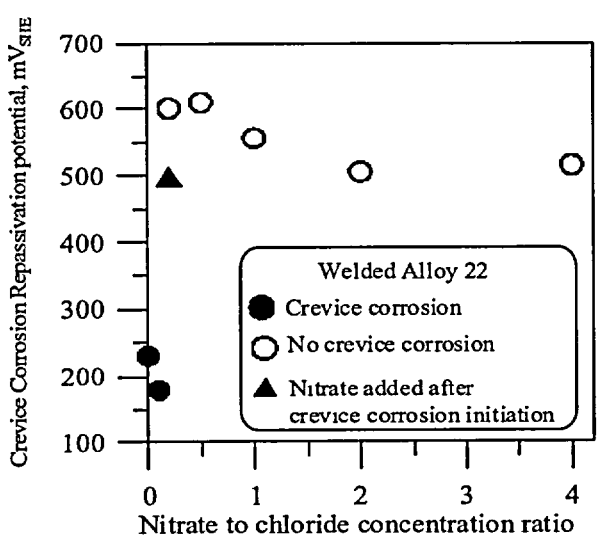

Figure 5. Effect of nitrate in inhibiting localized corrosion of Alloy 22.

LOCALIZED CORROSION PROPAGATION

If localized corrosion occurs under conditions discussed in the previous section, the growth rate of such corrosion is important to determine the time of complete penetration of the container. The growth rate of pits and crevices have been examined by many investigators[9] and has been found in general to be determined by the transport of metal-chloride complexes from within the pit to the external environment. An example of the growth rate of crevice

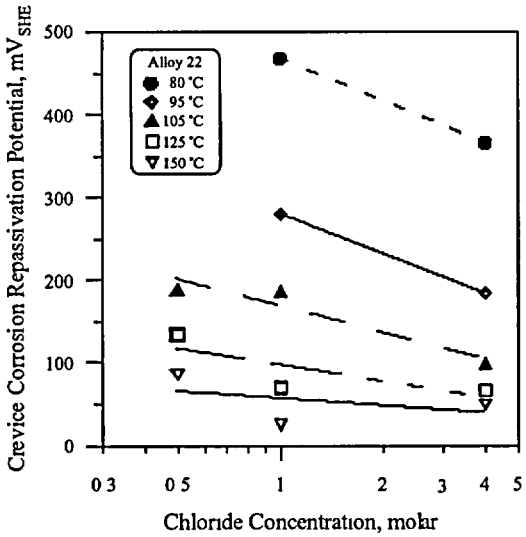

Figure 6. Effect of temperature on repassivation potential of Alloy 22.[2]

corrosion for Type 316L stainless steel is shown in Figure 7. The growth rate in this figure was measured during exposure to a naturally aerated solution. The measurement is performed by galvanically coupling a creviced sample to a large open sample of Type 316L stainless steel through a zero-resistance ammeter and measuring the resulting current. The current is then converted to a crevice corrosion growth rate. The value of the exponent indicates that the morphology of the pitting within the crevice is not perfectly hemispherical. It is also important to note that the crevice corrosion growth rates are extremely high. Therefore, long container life can not be obtained for these materials when they suffer localized corrosion.

PASSIVE DISSOLUTION

Passive dissolution occurs when an alloy is covered by an non-porous, oxide film. Passive dissolution may occur through a variety of mechanisms, some of which have implications on the long-term life of the container. Because of the extremely low dissolution rates and the continual formation and dissolution of the oxide film, measurements using conventional weight-change techniques are not accurate. Electrochemical techniques can provide greater resolution, but need to be interpreted carefully since multiple electrochemical reactions contribute to the measured currents. The dissolution rates shown in Figure 8 for Alloy 22 are independent of potential up to about 600 mV_{SCE}, beyond which they increase with potential. The region of steep increase in current with potential is governed by a combination of oxygen evolution and transpassive dissolution in which Cr in the metal dissolves as Cr (VI) species. However, from a practical point of view the transpassive regime is not of relevance to repository environment. This is because even in the presence of highly oxidizing radiolytic species the corrosion potential of the alloy is not expected to attain such high values. The passive dissolution rates in Figure 8 are also relatively independent of the chloride concentration and pH in the environment. The range of current densities in the passive dissolution regime correspond to uniform corrosion rates of 10^{-4} to 10^{-3} mm/year.

CONTAINER PERFORMANCE ASSESSMENT

The NRC has developed a Total-System Performance Assessment (TPA) code[11] to support a risk-informed, performance-based safety review of the proposed repository. A module in this code, EBSFAIL, is used to compute

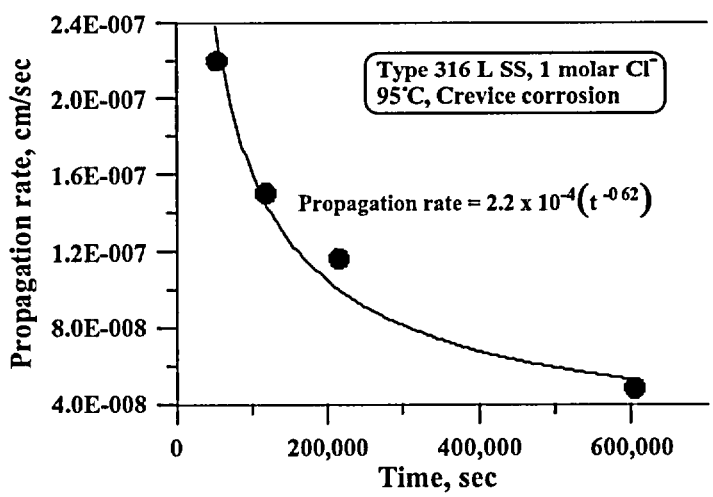

Figure 7. Crevice corrosion propagation rate of Type 316L stainless steel in NaCl

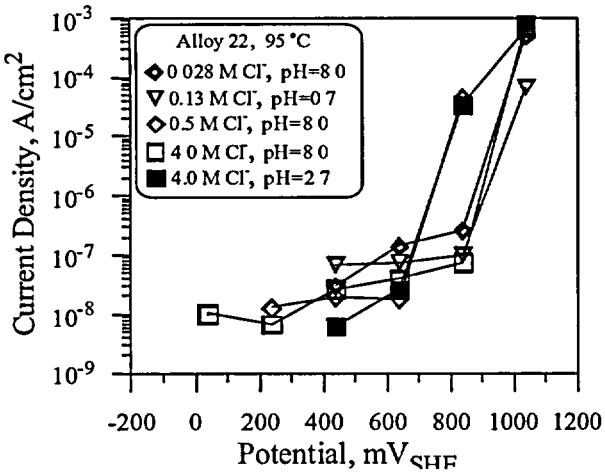

Figure 8. Uniform passive dissolution in the absence of localized corrosion as measured by electrochemical techniques.[2]

the failure time of the containers as a result of corrosion processes that are considered the primary degradation modes. The Engineered Barrier System Performance Assessment Code (EBSPAC) Version 1.1, which served as a basis for the development of the EBSFAIL module, is described in detail in a previous paper[12]. As mentioned previously, aqueous corrosion models are used to estimate container life by calculating the uniform corrosion rate using measured values of the passive current density if the corrosion potential is lower than the critical potential for the initiation of localized (crevice) corrosion. Otherwise, if the corrosion potential is higher than the critical potential, the corrosion rate is calculated using estimated values of the penetration rate due to crevice corrosion, assuming that the initiation time for this process is negligible.

Using appropriate electrochemical and environmental parameters, the corrosion potential, E_{corr}, defined as the potential at which the current due to all the cathodic processes is equal to the current due to the electrochemical dissolution of the metal, is calculated in the EBSFAIL module as a function of time.[11,12] The two cathodic reactions implemented in the EBSFAIL module are the oxygen reduction and the hydrogen evolution reactions. The oxygen reduction reaction is assumed to be limited by a combination of activation-controlled charge transfer and molecular diffusion-controlled transport processes. The current density for oxygen reduction, i_{O_2}, is then given by Eq. (1)

$$i_{O_2} = \frac{-k_{O_2} C_{O_2}^{bulk} \exp\left(-\frac{z_{O_2}\beta_{O_2} F E_{corr}}{RT}\right)}{1 + \frac{\left[k_{O_2}\delta \exp\left(-\frac{z_{O_2}\beta_{O_2} F E_{corr}}{RT}\right)\right]}{4FD_{O_2}}} \tag{1}$$

where k_{O_2} is the reaction rate constant for the oxygen reduction reaction, d is the thickness of the water film, D_{O_2} is the oxygen diffusivity in the water film, $C_{O_2}^{bulk}$ is the bulk concentration of oxygen in solution, which is related to the partial pressure of oxygen through Henry's law, z_{O_2} is the charge associated with the oxygen reduction, β_{O_2} is the transfer coefficient, F is the Faraday's constant, and E_{corr} is the corrosion potential.

For the water reduction reaction, the cathodic current is assumed to be controlled only by the charge transfer process and the cathodic current density is given by Eq. (2)

$$i_{H_2O} = k_{H_2O} \exp\left(-\frac{z_{H_2O}\beta_{H_2O} F}{RT} E_{corr}\right) \tag{2}$$

where k_{H_2O} is the reaction rate constant for the water reduction reaction and z and β represent similar parameters as for Eq. (1) for water reduction. The temperature dependence of the reaction rate constants for the oxygen evolution and the water reduction reactions is given by an Arrhenius-type relationship. The E_{corr} is then calculated by solving simultaneously for E_{corr} using the passive current density, which is assumed to be independent of potential and temperature, and Eqs. (1) and (2). Details of the derivation and values for the various electrochemical parameters are provided elsewhere.[12] For simplicity, E_{rcrev} is assumed to depend only on chloride concentration and temperature, even

though both nitrate and sulfate may act as inhibitors above a certain concentration ratio of nitrate plus sulfate to chloride [13,14] Above a critical chloride concentration and a threshold temperature which are both dependent on the alloy, the dependence of E_{rcrev} on chloride concentration and temperature for the three Ni-Cr-Mo alloys, as determined experimentally[12], is given by Eq. (3)

$$E_{rcrev} = E^0_{rcrev}(T) + B(T)\log[Cl^-] \tag{3}$$

where the constants $E^0_{rcrev}(T)$ and $B(T)$ are linear functions of temperature given by Eq.(4) and (5).

$$E^0_{rcrev}(T) = A_1 + A_2 T \tag{4}$$

$$B(T) = B_1 + B_2 T \tag{5}$$

The coefficients of these equations are listed in Table 1.

TABLE 1. COEFFICIENTS OF THE CREVICE CORROSION REPASSIVATION POTENTIAL EXPRESSIONS

Alloy	T (°C)	$[Cl^-]_{crit}$ (M)	A_1 (mV_{SHE})	A_2 (mV/°C)	B_1 (mV)	B_2 (mV/°C)
825	50–100	0.002	422.8	-4.1	-64.0	-0.80
625	95	0.03	98.5	*	-160.8	*
22	80–125	0.5	1,540	-13.1	-362.7	2.3
*Not measured; data only for 95 °C.						

If E_{corr} is higher than E_{rcrev}, localized (crevice) corrosion penetration is given by Eq. (6)

$$P = At^n \tag{6}$$

where P is the depth of the localized attack, t is time, and A and n are experimentally determined constants. If E_{corr} is less than E_{rcrev}, penetration of the WP wall will occur by passive dissolution and the corrosion rate is calculated using Eq. (7)

$$CR = \frac{i_{pass} EW}{F\rho} \tag{7}$$

where i_{pass} is the passive dissolution current density, EW is the equivalent weight, and ρ is the density of the alloy. It should be noted, however, that the life of the Alloy 22 WP outer container under passive conditions depends on the long-term stability of the protective Cr (III)-rich oxide film. Recent modeling of the long-term passive dissolution of Ni-Cr-Mo alloys, using a modification of the Point Defect Model,[13] suggest that in the long term, dissolution is stoichiometric. Although preferential dissolution of Ni has been observed in short-term experiments, passive dissolution will be stoichimetric in the long term, presumably accompanied by periodic spalling of the passive film, unless either fast penetrating paths of the corrosion front (e.g., grain boundaries) or substantial changes in the reactive surface area occur.[13]

RESULTS OF COMPUTATIONS

Figure 9 shows the results of E_{corr} and E_{rcrev} calculations as a function of time for the Alloy 22 WP using both chloride concentration and temperature as inputs at a constant pH equal to 9.0. Two periods where aqueous corrosion occurs, as determined by RH > $RH_{aq\ corr}$, are identified in Figure 1. The first period is relatively short, lasting from 5 to 50 years after waste emplacement. The second period occurs after 1,000 years when the WPs cool to temperatures below boiling. The E_{corr} is calculated using both a low current density of 5.0×10^{-9} A cm^{-2} [32.26×10^{-9} A in^2] and a high current density of 5.4×10^{-8} A cm^{-2} [34.84×10^{-8} A in^2], corresponding to corrosion rates of 4.9×10^{-5} and 5.3×10^{-4} mm yr^{-1} [1.93×10^{-6} and 2.09×10^{-5} in yr^{-1}] The E_{corr} varies approximately 50 mV with such variation in corrosion rates, and the highest E_{corr} is approximately 200 mV_{SHE}. The E_{rcrev} is defined for [Cl^-] >

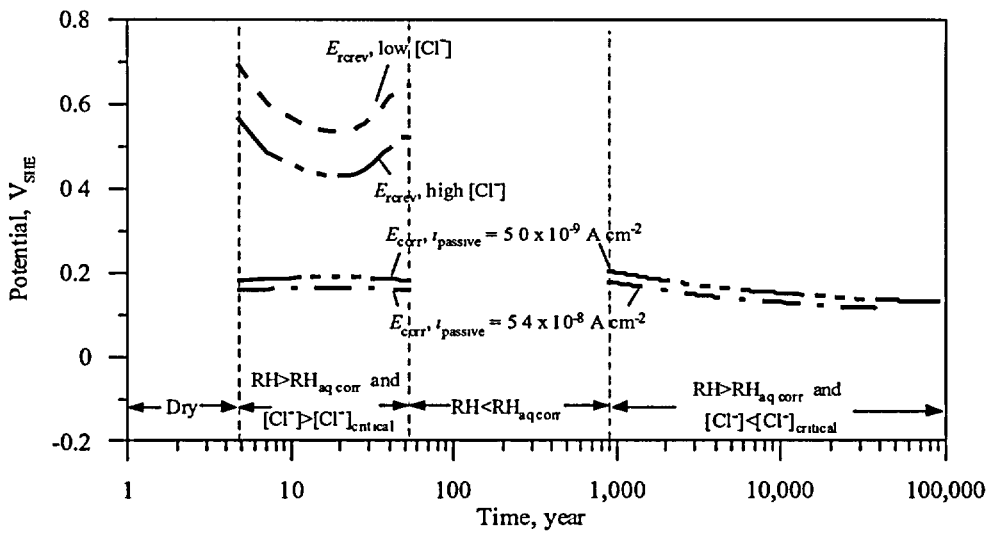

Figure 9. Corrosion potential and crevice corrosion repassivation potential as a function of time for the Alloy 22 waste package outer container.[15]

$[Cl^-]_{crit}$ when the RH > $RH_{aq\ corr}$. During the initial wet period after waste emplacement, the E_{rcrev} is calculated for both a low and a high chloride concentration. The low chloride concentration (1.8 M Cl^-) is based on the total Na^+ concentration in J–13 water (a reference groundwater from the vicinity of Yucca Mountain) saturated with respect to halite (NaCl), whereas the high chloride concentration (6.6 M Cl^-) is that corresponding to saturated NaCl in water with no other dissolved sodium salts. Localized corrosion is not initiated because E_{corr} never exceeds E_{rcrev} for either the low or the high chloride concentration, and only passive dissolution of Alloy 22 WPs occurs in the initial wet period. After 1,000 years when the WPs cool down to temperatures below boiling, the value of E_{rcrev} becomes very high because the chloride concentration in the concentrated solutions is assumed to rapidly decrease to very low values when more water contacts the WPs and hence only passive dissolution occurs.

Plots of the remaining WP wall thickness for two values of passive current density are shown in Figure 10 as a function of time. Even though the regulations require assessment for 10,000 years, the calculations are extended to 100,000 years to illustrate the effect of uncertainties in the values of the corrosion rate. Penetration of the WP wall by uniform corrosion is estimated to occur after 37,000 years if a passive current density of 5.4×10^{-8} A cm^{-2} [34.84 $\times 10^{-8}$ A in^2] is used but life is extended beyond 100,000 years if a value of 5.0×10^{-9} A cm^{-2} [32.26 $\times 10^{-9}$ A in^2] is adopted. It is apparent that the extremely long WP lifetime is the result of the excellent resistance of Alloy 22 to localized corrosion as indicated by the high values of $[Cl^-]_{crit}$ and E_{rcrev}.

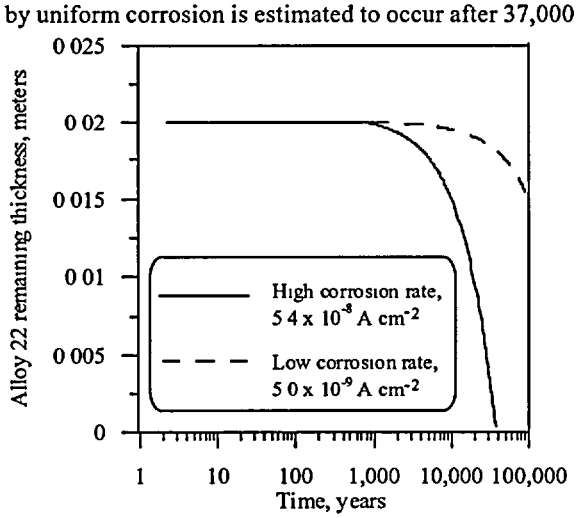

Figure 10. Remaining wall thickness for the Alloy 22 WP as a function of time after waste emplacement.[13]

The fraction of WPs failed as a function of time is plotted in Figure 11, as an example of the computations conducted with the probabilistic TPA code, Version 4.0. The 95th and 5th percentiles and the mean fraction of waste packages failed are shown in this example. Failure is assumed to be penetration of the outer container of the waste package, since no credit is assumed for the performance of the inner container made out of Type 316L stainless steel. It is shown in Figure 11 that the fraction of failed WPs starts to increase significantly beyond the 10,000-yr compliance period as a result of slow uniform corrosion under passive conditions in agreement with the results of the deterministic calculations plotted in Figure 10.

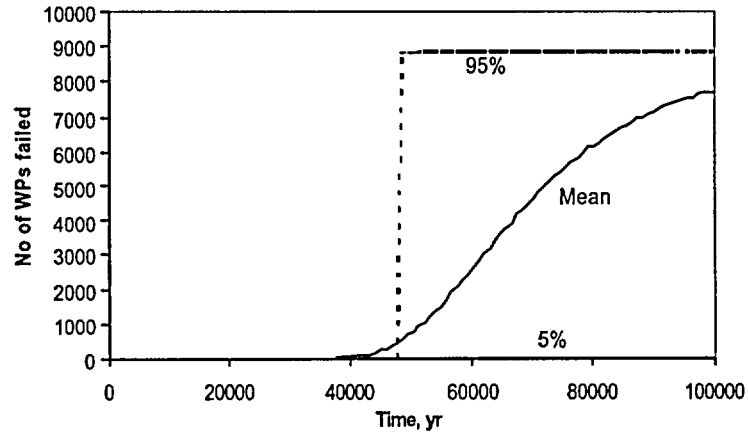

Figure 11. Calculated number of Alloy 22 waste packages that fail at various times.

SUMMARY

This paper outlines the approach used to model container life in the proposed repository at Yucca Mountain. The approach involves (i) an estimation of the temperature and chemistry of water contacting the waste package, (ii) calculation of the corrosion potential, (iii) calculation of the repassivation potential, (iv) calculation fo the time at which localized corrosion would occur, and (v) calculation of the rates of localized and uniform corrosion rates. Based on this approach, it has been shown that container life depends on the chemical composition of the environment, temperature, and passive dissolution rate. The performance assessment calculations presented in this paper, are not intended to demonstrate container life, but are intended to determine the factors that are important to the overall performance of the containers. More detailed, process level and mechanistic models are being evaluated to increase the confidence in the performance assessment calculations Long-term laboratory studies,[2] and studies of natural analogs will provide additional confidence to the model assumptions and parameters.[17] Finally, methods are being evaluated for confirmation of container performance in the proposed repository using remotely monitored sensors.[18]

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