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U.S. Nuclear Regulatory Commission  
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Subject: Resubmittal of paper titled "Lifetime Prediction of High-Level Radioactive Waste Containers Affected by Corrosion" for presentation at the 15<sup>th</sup> International Corrosion Congress

Dear Mrs. DeMarco:

The enclosed paper with A390 form is being resubmitted for programmatic review in accordance with the new guidelines for presenting papers at the International meetings. This paper has already been submitted for presentation at the 15<sup>th</sup> International Corrosion Congress to be held September 22-27, 2002, in Granada, Spain. If NRC staff review indicates that the paper is programmatically unacceptable for publication, it will be withdrawn from the conference.

Please contact Gustavo Cragolino at (210) 522-5539 if you have any questions regarding this paper. 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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# **Lifetime Prediction of High-Level Radioactive Waste Containers Affected by Corrosion**

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## **Abstract**

The approach currently adopted by the U.S. Nuclear Regulatory Commission (NRC) for estimating the life of containers to be used for the potential geological disposal of spent nuclear fuel and high-level radioactive waste in the proposed repository at Yucca Mountain, Nevada, USA is discussed. Calculations of the failure time of the containers (defined as through-wall penetration) are conducted using the EBSFAIL module of the NRC/CNWRA Total-system Performance Assessment (TPA) code. Uniform passive corrosion and localized (crevice) corrosion are considered the two dominant failure modes limiting container life in the TPA code. It is shown that the approach used to estimate container life could be used for lifetime prediction of structures and components in other industrial applications.

*Keywords: localized and passive corrosion, corrosion potential, Alloys 825, 625 and 22*

## **Introduction**

In the United States, the U.S. 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 a 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 the U.S. Code of Federal Regulations (10 CFR Part 63) before granting or denying authorization for construction and subsequent possession of SF and waste.

The NRC has developed a TPA code [1] to conduct a risk-informed, performance-based safety review of the proposed repository. A module in this code, EBSFAIL, is used to compute the failure time of the containers as a result of corrosion processes that are considered the primary degradation modes of the engineered barriers. 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 [2]. The current DOE design of the waste package (WP) consists of a 20-mm [0.787 in] thick Alloy 22 (Ni-22Cr-13.5Mo-3W-3Fe) outer container surrounding a 50-mm [1.969] thick type 316 nuclear grade (NG) stainless steel inner container to provide structural strength only. An earlier DOE WP design, with Alloy 825 (Ni-21Cr-3Mo-29Fe-2Cu) and A516 steel as inner and outer container materials, was previously evaluated using EBSPAC Version 1.1 [2]. Alloy 625 (Ni-22Cr-9Mo-4Nb-5Fe) was also considered by the DOE as a potential candidate material for such design, instead of Alloy 825, before the selection of Alloy 22.

## **Overview of Processes and Models**

In the proposed repository, alteration of the physical and chemical environment within the

emplacement drifts located in the unsaturated zone could have an important effect on the performance of the WP. After waste emplacement, the environment inside the drift is expected to be hot, dry air because of the combination of the heat generated through radioactive decay and either forced or natural ventilation of the drifts which will be located well above the water table. Water may contact the WP surface by dripping of groundwater, which percolates through rock fractures, or by the formation of a water film on the WP surface when the relative humidity (RH) in the emplacement drifts exceeds a critical RH [1]. This critical RH for water film formation will be affected by the presence of dust or any hygroscopic salt deposit on the WP surface. After water contact with the WP, aqueous corrosion will occur and the rate of aqueous corrosion will depend on the stability of the passive oxide film. As long as the passive film is stable, the rate of corrosion is expected to be low and dictated by the slow passive dissolution of the alloy. However, breakdown of the passive film within occluded areas could lead to the initiation of crevice corrosion causing rapid penetration of the WP wall if localized corrosion propagation is sustained.

Aqueous solutions containing critical concentrations of aggressive anionic species such as chloride may promote localized corrosion at sufficiently elevated temperatures if the corrosion potential is higher than the critical potential for the initiation of localized corrosion. For the expected repository environment, the oxygen partial pressure will be equivalent to that in air at the repository horizon. Under such oxidizing conditions, the corrosion mode will be determined by the temperature and the chemical composition and pH of the water in contact with the WP. Additional oxidizing species such as Fe(III) or  $H_2O_2$  may facilitate the initiation of localized corrosion, whereas inhibiting species present in the groundwater such as nitrate may prevent initiation and promote repassivation of localized corrosion

### Thermal and Environment Models

A detailed description of the thermal and environment models has been presented [1–3] and only a brief summary is given here. The temperature and RH in the emplacement drifts are dependent on the average thermal loading that can be modified by altering the drift spacing and the spacing between WPs, in addition to the load of each WP. An analytical conduction-only model is used to calculate the repository horizon average rock temperature. The WP temperature is then calculated as a function of that temperature and the heat output per metric ton of heavy metal in the WP using a conduction, convection and radiation heat transfer analytical model. The model output provides a distribution of drift wall and WP temperatures as a function of time. From these parameters the RH at the WP surface is calculated using the following expression

$$RH = \frac{P_v[\min(T_b, T_w)]}{P_v(T_{WP})} \quad (1)$$

where  $P_v$  is the vapor pressure which is a function of temperature,  $T_b$  is the boiling point temperature at the repository horizon,  $T_w$  is the temperature of the drift wall,  $T_{WP}$  is the temperature of the WP, and  $\min(T_b, T_w)$  is the minimum of the two values.

The environment model, based in the computer code MULTIFLO [4], which accounts for transport of reacting chemical species coupled to evaporation and condensation process involving two-phase fluid flow, provides the chemical environment as a function of time in the vicinity of the WP. The calculated chemical parameters are pH, partial pressure of oxygen, chloride and bicarbonate concentrations, and dissolved silica, but for the purpose of evaluating container life through EBSFAIL only the chloride concentration is used as input.

## Aqueous Corrosion Models

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.

Calculation of corrosion potential. Using appropriate electrochemical and environmental parameters, the corrosion potential,  $E_{\text{corr}}$ , 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 [1–3]. Prior to the occurrence of localized corrosion, uniform distribution of cathodic and anodic areas is assumed for simplicity. Once localized corrosion occurs, it is assumed that the cathodic reaction takes place on the whole available surface because the pitted area is comparatively small.

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,  $i_{\text{O}_2}$ , is then given by

$$i_{\text{O}_2} = \frac{-k_{\text{O}_2} C_{\text{O}_2}^{\text{bulk}} \exp\left(-\frac{z_{\text{O}_2} \beta_{\text{O}_2} F E_{\text{corr}}}{RT}\right)}{1 + \frac{k_{\text{O}_2} \delta \exp\left(-\frac{z_{\text{O}_2} \beta_{\text{O}_2} F E_{\text{corr}}}{RT}\right)}{4FD_{\text{O}_2}}} \quad (2)$$

where  $k_{\text{O}_2}$  is the reaction rate constant for the oxygen reduction reaction,  $\delta$  is the thickness of the water film,  $D_{\text{O}_2}$  is the diffusivity, and  $C_{\text{O}_2}^{\text{bulk}}$  is the bulk concentration of oxygen in solution, which is related to the partial pressure of oxygen through Henry's law.

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

$$i_{\text{H}_2\text{O}} = k_{\text{H}_2\text{O}} \exp\left(-\frac{z_{\text{H}_2\text{O}} \beta_{\text{H}_2\text{O}} F}{RT} E_{\text{corr}}\right) \quad (3)$$

where  $k_{\text{H}_2\text{O}}$  is the reaction rate constant for the water reduction reaction. 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  $E_{\text{corr}}$  is then calculated by solving simultaneously for  $E_{\text{corr}}$  using the passive current density, which is assumed to be independent of potential and temperature, and Eqs. (3) and (4). Details of the derivation and values for the various electrochemical parameters are provided elsewhere [1,2].

Localized corrosion model. Based on experimental measurements [5], it is postulated in the EBSFAIL module that, above a threshold chloride concentration, the lowest value of the critical potential for the initiation of crevice corrosion is defined by the repassivation potential,  $E_{\text{rcrev}}$ . For simplicity,  $E_{\text{rcrev}}$  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 [3,6]. Above a critical chloride concentration and a threshold temperature which are both dependent on the alloy, the dependence of  $E_{\text{rcrev}}$  on chloride concentration and temperature for the three Ni-Cr-Mo alloys, as determined experimentally [5,6], is given by

$$E_{\text{rcrev}} = E_{\text{rcrev}}^0(T) + B(T) \log[\text{Cl}^-] \quad (4)$$

where the constants  $E_{\text{rcrev}}^0(T)$  and  $B(T)$  are linear functions of temperature given by

$$E_{\text{rcrev}}^0(T) = A_1 + A_2 T \quad (5)$$

$$B(T) = B_1 + B_2 T \quad (6)$$

and the coefficients of these equations are listed in Table 1.

**Table 1. Coefficients of the Crevice Corrosion Repassivation Potential Expressions**

Alloy	T (°C)	$[\text{Cl}^-]_{\text{crit}}$ (M)	$A_1$ (mV <sub>SHE</sub> )	$A_2$ (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  $E_{\text{corr}}$  is higher than  $E_{\text{rcrev}}$ , localized (crevice) corrosion penetration is given by

$$P = At^n \quad (7)$$

where  $P$  is the depth of the localized attack,  $t$  is time, and  $A$  and  $n$  are experimentally determined constants.

Passive corrosion model. If  $E_{\text{corr}}$  is less than  $E_{\text{rcrev}}$ , penetration of the WP wall will occur by passive dissolution and the corrosion rate is calculated using the following expression

$$CR = \frac{i_{\text{pass}} EW}{F \rho} \quad (8)$$

where  $i_{\text{pass}}$  is the passive dissolution current density,  $EW$  is the equivalent weight, and  $\rho$  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 [7,8], using a modification of the Point Defect Model [9], suggest that in the long term, dissolution is stoichiometric because vacancy diffusivity in the bulk of the metal is very slow. Although preferential dissolution of Ni has been observed in short-term experiments, passive dissolution will be stoichiometric in the long term, presumably accompanied by periodic spalling of the passive film, unless that either fast penetrating paths of the corrosion front or substantial changes in the reactive surface area occur [7,8].

## Results of Computations

After waste emplacement, natural ventilation removes some fraction of the heat generated by the WPs limiting the maximum WP temperature to approximately 100 °C [212 °F] according to calculations for an average thermal loading of 60 MTU per acre. Following repository closure, a temperature increase is observed 50 years after emplacement reaching a peak WP

temperature of 160 °C [320 °F] after 100 years. The WP temperature then remains above boiling for approximately 1,000 years.

Initially the calculated RH at the WP surface is approximately 55 percent but within a few years after waste emplacement, the RH exceeds the critical RH for aqueous corrosion, which is normally distributed in the range of 60 to 65 percent RH. The large decrease in RH after 50 years of waste emplacement occurs as a result of repository closure and coincides with a sharp temperature increase. After the maximum WP temperature is reached and the WPs start to cool down, the RH increases and exceeds the critical RH for aqueous corrosion at approximately 1,000 years after waste emplacement. The RH remains higher than 95 percent at all times greater than 2,000 years.

Figure 1 shows the results of  $E_{\text{corr}}$  and  $E_{\text{rcrev}}$  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 [3]. Two periods where aqueous corrosion occurs, as determined by  $\text{RH} > \text{RH}_{\text{aq corr}}$ , 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  $E_{\text{corr}}$  is calculated using both a low current density of  $5.0 \times 10^{-9} \text{ A cm}^{-2}$  [ $32.26 \times 10^{-9} \text{ A in}^{-2}$ ] and a high current density of  $5.4 \times 10^{-8} \text{ A cm}^{-2}$  [ $34.84 \times 10^{-8} \text{ A in}^{-2}$ ], corresponding to corrosion rates of  $4.9 \times 10^{-5}$  and  $5.3 \times 10^{-4} \text{ mm yr}^{-1}$  [ $1.93 \times 10^{-6}$  and  $2.09 \times 10^{-5}$  in  $\text{yr}^{-1}$ ] [3,8]. The  $E_{\text{corr}}$  varies approximately 50 mV with such variation in corrosion rates, and the highest  $E_{\text{corr}}$  is approximately 200 mV<sub>SHE</sub>. The  $E_{\text{rcrev}}$  is defined for  $[\text{Cl}^-] > [\text{Cl}^-]_{\text{crit}}$  when the  $\text{RH} > \text{RH}_{\text{aq corr}}$ . During the initial wet period after waste emplacement, the  $E_{\text{rcrev}}$  is calculated for both a low and a high chloride concentration. The low chloride concentration (1.8 M  $\text{Cl}^-$ ) is based on the total  $\text{Na}^+$  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  $\text{Cl}^-$ ) is that corresponding to saturated NaCl in water with no other dissolved sodium salts. Localized corrosion is not initiated because  $E_{\text{corr}}$  never exceeds  $E_{\text{rcrev}}$  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  $E_{\text{rcrev}}$  becomes very high because the chloride concentration in the concentrated solutions is

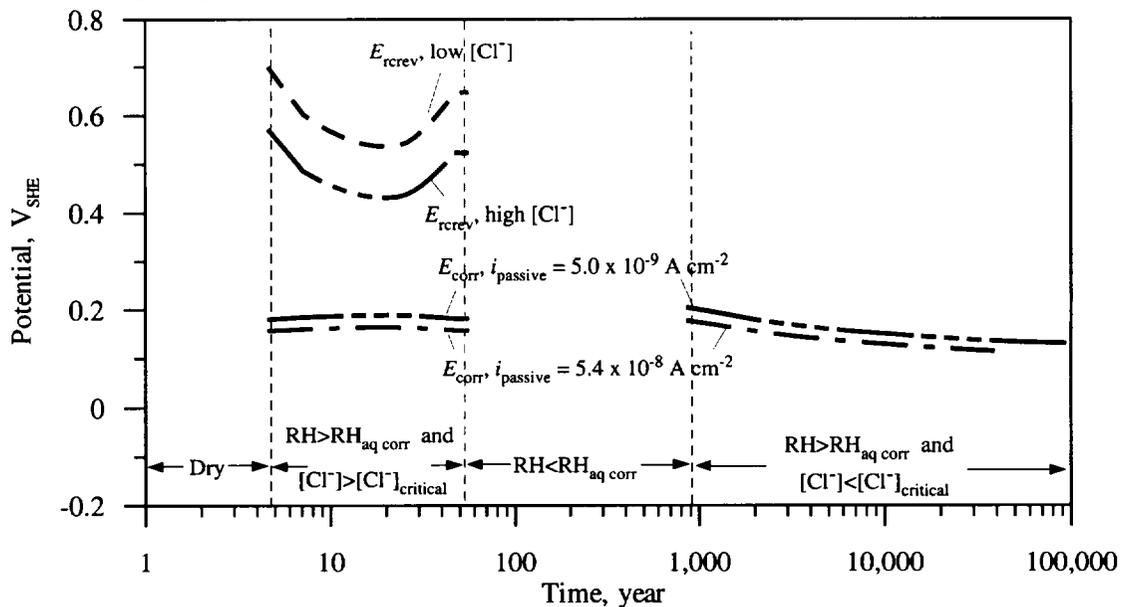
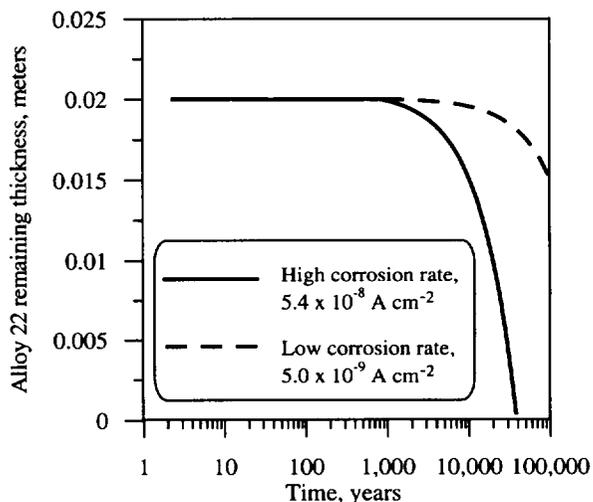


Figure 1. Corrosion potential and crevice corrosion repassivation potential as a function of time for the Alloy 22 waste package outer container

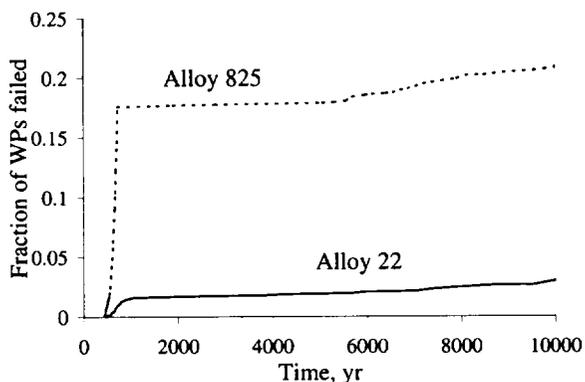
assumed to rapidly decrease to very low values when more water contacts the WPs and hence only passive dissolution occurs.

Plots of the remaining WP wall thickness for two values of passive current density are shown in Figure 2 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  $5.4 \times 10^{-8} \text{ A cm}^{-2}$  [ $34.84 \times 10^{-8} \text{ A in}^2$ ] is used but life is extended beyond 100,000 years if a value of  $5.0 \times 10^{-9} \text{ A cm}^{-2}$  [ $32.26 \times 10^{-9} \text{ A in}^2$ ] 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  $[\text{Cl}^-]_{\text{crit}}$  and  $E_{\text{rrev}}$ .

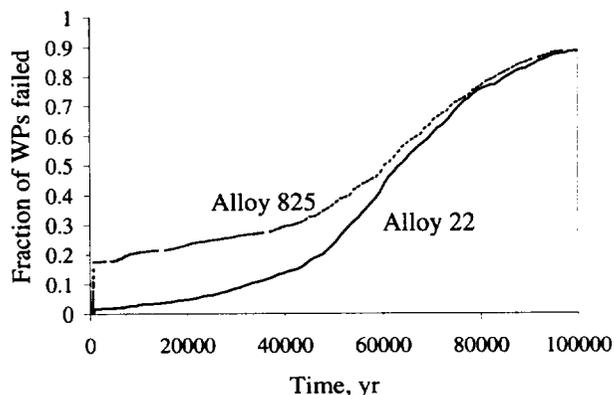


**Figure 2. Remaining wall thickness for the Alloy 22 WP as a function of time after waste emplacement**

The fraction of WPs failed as a function of time is plotted as cumulative distribution functions for 10,000 and 100,000 years in Figures 3 and 4. As an example of the computations conducted with the probabilistic TPA code, a comparison between Alloys 825 and 22 as outer container materials for approximately 8,900 emplaced WPs is shown in both figures using the mean values of 200 realizations. In this example, the chloride concentration is assumed to decrease from 0.06 M at 1,000 years to 0.003 M after 100,000 years and a multiplication factor uniformly distributed from 1 to 30 is used to account for variations in different areas of the repository. It is clearly seen in Figure 3 that approximately 18 percent of the Alloy 825 WPs fail in less than 1,000 years as a result of wall penetration due to localized corrosion. On the other hand, only a small fraction (< 2 percent) of Alloy 22 WPs fails in the same period by localized corrosion. It is shown in Figure 4 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 2. It should be noted that a uniform distribution of



**Figure 3. Cumulative distribution function for failure of Alloys 825 and 22 waste packages in 10,000 years**



**Figure 4. Cumulative distribution function for failure of Alloys 825 and 22 waste packages in 100,000 years**

corrosion rates corresponding to the range of passive current densities listed in Figure 2, and identical for both alloys, is used in the calculations plotted in Figures 3 and 4. Therefore, the fraction of failed WPs becomes equal for both Alloy 825 and 22 WPs after 100,000 years.

### Extension of the Approach to other Industrial Applications

The approach used to estimate container life based on the use of the  $E_{\text{corr}}$  and  $E_{\text{rcrev}}$  could be extended to other applications to predict if localized corrosion may occur and estimate at least approximately a component life. The validity of these electrochemical parameters as predictors has been discussed previously on the basis of limited field experience [5,10]. Table 2 is a compilation of several examples in which  $E_{\text{rcrev}}$  values calculated with Eqs. (4), (5) and (6), and the coefficients of Table 1, as well as values measured for 316L SS [10], are compared to measured  $E_{\text{corr}}$  values to predict the occurrence of crevice corrosion. In many cases in which crevice corrosion was observed, as indicated by CC, a good correspondence with values of  $E_{\text{corr}}$  greater than those of  $E_{\text{rcrev}}$  was obtained, as shown in Table 2.

**Table 2. Use of Repassivation Potential for Prediction of Localized Corrosion in Some Industrial Applications (Original references are provided in Reference 10)**

Application	Marine	Marine	Marine	Pulp and Paper	Geothermal Energy
Environment	Seawater	Seawater + Cl <sub>2</sub>	3%NaCl + Air	0.5%Cl <sup>-</sup> , pH3	12%Cl <sup>-</sup> , pH3
Temp. (°C/°F)	25/77	42/107.6	80/176	25/77	100/212
$E_{\text{corr}}$ (mV <sub>SHE</sub> )	440 to 540	440 to 670	40	340 to 740	40 to 50
316L SS	CC	CC	CC	CC	—
$E_{\text{rcrev}}$ (mV <sub>SHE</sub> )	240	—	-120	210 (317L SS)	—
Alloy 825	CC	CC	No CC	—	—
$E_{\text{rcrev}}$ (mV <sub>SHE</sub> )	346	221	133	—	—
Alloy 625	CC	CC	No CC	—	No CC
$E_{\text{rcrev}}$ (mV <sub>SHE</sub> )	—	—	147	—	50
Alloy 22	No CC	No CC	No CC	—	No CC
$E_{\text{rcrev}}$ (mV <sub>SHE</sub> )	—	—	546	—	48

### Summary and Conclusions

The long-term performance of the WPs in the proposed repository at Yucca Mountain was evaluated using simplified models based on the expected thermal and environmental conditions within the emplacement drifts and data from laboratory investigations. The  $E_{\text{corr}}$ , which is amenable to experimental verification, is calculated and compared at each time step with  $E_{\text{rcrev}}$  to determine if localized corrosion can be initiated. Model calculations suggest that only a limited fraction of Alloy 22 WP outer containers may be susceptible to localized corrosion under the environmental conditions prevailing in the drifts and most of the failures will be the result of slow passive dissolution rate. There are uncertainties in the parameters used in the EBSFAIL module, which are related to the definition of the environment, the long-term stability of the passive film, and the influence of fabrication factors on the corrosion processes.

A similar approach to that used in the performance evaluation of the engineered barriers for the proposed repository can be used in other industrial applications. Limited examples from marine applications, pulp and paper industry, and geothermal energy generation illustrate the

advantages of this methodology to prevent the occurrence of localized (crevice) corrosion. The use of  $E_{\text{crev}}$  as a critical potential for the initiation of localized corrosion and  $E_{\text{corr}}$  as a driving force of its occurrence can eliminate many uncertainties in design and selection of materials particularly when the environmental conditions are relatively well defined. Indeed, one of the most important limitations confronted in the prediction of the material behavior for the proposed repository is related to the uncertainties associated with the evolution in the composition of the aqueous environment in contact with the WPs.

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