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**STRESS CORROSION CRACKING MODEL FOR HIGH LEVEL
RADIOACTIVE-WASTE PACKAGES**

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ABSTRACT

A stress corrosion cracking (SCC) model has been adapted for performance prediction of high level radioactive-waste packages to be emplaced in the proposed Yucca Mountain repository. For waste packages of the proposed Yucca Mountain repository, the outer barrier material is the highly corrosion-resistant Alloy UNS-N06022 (Alloy 22), the environment is represented by aqueous brine films present on the surface of the waste package from dripping or deliquescence of soluble salts present in any surface deposits, and the tensile stress is principally from weld induced residual stress. SCC has historically been separated into "initiation" and "propagation" phases. Initiation of SCC will not occur on a smooth surface if the surface stress is below a threshold value defined as the threshold stress. Cracks can also initiate at and propagate from flaws (or defects) resulting from manufacturing processes (such as welding); or that develop from corrosion processes such as pitting or dissolution of inclusions. To account for crack propagation, the slip dissolution/film rupture (SDFR) model is adopted to provide mathematical formulae for prediction of the crack growth rate. Once the crack growth rate at an initiated SCC is determined, it can be used by the performance assessment to determine the time

to through-wall penetration for the waste package. This paper presents the development of the SDFR crack growth rate model based on technical information in the literature as well as experimentally determined crack growth rates developed specifically for Alloy UNS-N06022 in environments relevant to high level radioactive-waste packages of the proposed Yucca Mountain radioactive-waste repository. In addition, a seismic damage related SCC crack opening area density model is briefly described.

INTRODUCTION

The engineered barrier system for the proposed Yucca Mountain high level radioactive-waste repository is designed to complement the natural barriers in isolating waste from the environment. A key component of the engineered barrier system is the waste package (WP). According to DOE 2004, a typical waste package (see Fig. 1) would have a dual-metal design containing two concentric cylinders. The inner cylinder would be made of Modified Stainless Steel Type 316 (UNS 31600) equivalent to Type 316NG. The outer cylinder would be made of a corrosion-resistant, nickel-based alloy (UNS-N06022). Alloy UNS-N06022 would protect the stainless steel inner cylinder from corrosion, and Modified Stainless Steel Type 316 (UNS 31600) would provide structural support for the thinner Alloy UNS-N06022 cylinder.

Fig. 1. Typical Waste Package Used for Yucca Mountain Repository.

Each waste package would have outer and inner lids at each end of the cylinder. The outer (closure) lids would be made of Alloy UNS-N06022. The inner lids would be made of Modified Stainless Steel Type 316 (UNS 31600). The loading end of the waste package has a third flat closure lid made of Alloy UNS-N06022, which would be placed between the inner lid of stainless steel and the outer lid of Alloy UNS-N06022. This flat middle closure lid provides an extra barrier against a potential release caused by a breach of the outer lid, such as by stress corrosion cracking (SCC) in the closure weld region. The outer lid closure weld region will be stress mitigated. The current baseline process is laser shock peening but an alternative process (controlled plasticity burnishing) is also under evaluation.

The discussion on SCC in this paper is limited to the Alloy UNS-N06022 waste package outer barrier (WPOB). The stainless steel structural material is not modeled since the waste package performance assessment does not take corrosion-resistance credit from the stainless steel inner barrier of the WP. The stress relevant to SCC in waste packages is primarily the residual tensile stress induced by the welding processes. The entire waste package will be heat treated (annealed) to eliminate the weld residual tensile stresses before the loading of waste contents and welding of the closure lids at the loading end. Therefore, the principal areas of SCC concern are the closure welds of the waste package at the loading end (once the stress mitigated layer depth is removed by general corrosion) since they are not post-weld heat treated. In addition, for low probability seismic events, SCC can potentially occur at seismically damaged regions of the waste package and a seismic crack density model is described that accounts for the effective through-wall crack opening area per unit area of seismic damage. The treatment of SCC described in this paper for the closure lid weld region is illustrated by the flow diagram shown in Fig. 2.

Note: When an indicated value of σ , K_I , or a_{Final} is less than the indicated parameter value, the corresponding crack initiation, crack growth or failure (breach) does not occur

Fig. 2. Flow Diagram for Treatment of Stress Corrosion Cracking.

CRACK INITIATION

SCC has historically been separated into “initiation” and “propagation” phases (Jones and Ricker 1987). For the purpose of lifetime modeling, it is appropriate to associate initiation with microscopic crack formation at localized corrosion or mechanical defect sites. In the area of environmentally assisted cracking (such as SCC), coalescence of microscopically small cracks will take place and develop into deeper cracks. Andresen and Ford (1988) used a crack size of 0.05 mm (50 μm) as an equivalent defect from which to start propagating SCC cracks.

For a given alloy, microstructure and environmental condition – and in the absence of cyclic stresses – initiation of SCC will not occur on a 'smooth surface' if the surface stress is below a threshold value defined as the threshold stress (ASM International 1987). SCC crack initiation measurements under constant, active load conditions obtained at GE Global Research Center are summarized in Fig. 3, where the measurements of crack initiation stress are presented as applied stress ratio (the ratio of applied stress to yield strength) vs. time-to-failure (or total exposure time without failure) for specimens subjected to over 21,000 hours of exposure in a hot concentrated salt solution (pH = 10.3 at 105°C). This solution is a more dilute version (15% of

full concentration) of the brine solution known as Basic Saturated Water (BSC 2001), designed to simulate the chemistry of concentrated Yucca Mountain ground water. The test results indicate that Alloy UNS-N06022 exhibits excellent SCC resistance since failure was not observed for any of the 120 Alloy UNS-N06022 specimens covering a variety of metallurgical conditions (including as-welded condition). The applied stress ratios were up to about 2.1 times the yield strength (YS) of the as-received material and up to 2.0 times the yield strength of the welded material. This corresponds to about 89 to 96% of the measured ultimate tensile strength (UTS). Based on the observations of no Alloy 22 SCC at applied stresses of 2.0 time the yield strength, a threshold stress criteria for SCC initiation of 90% of yield strength was conservatively selected for describing initiation on 'smooth' surfaces.

Fig. 3. Failure Stress vs. Time-to-Failure Plot for Crack Initiation Tests.

This high threshold initiation stress is further corroborated by more recent, single and double U-bend tests performed at GE Global Research Center in a Simulated Concentrated Water (SCW) chemistry (SCW is an approximately 1000-fold concentration of J-13 Yucca Mountain site groundwater with a pH of about 10, BSC 2001) at 165 °C. Creviced, mill annealed double U-bends and a range of heat treated, welded Alloy 22 single U-bends (machined from about 1.25 inch thick gas tungsten arc welded plate) have been exposed with the weld located at the U-bend

apex. Specimens were 0.125-inch thickness and were bent using a 1-inch diameter mandrel. The most recent inspection after 4844 hours exposure showed no evidence of stress corrosion crack initiation.

In the case of seismic related impact damage generated residual stresses, if the resultant calculated outer Alloy 22 surface residual stress in the damaged areas exceeds the 90% of yield strength initiation criterion, a high density network of through-wall SCC is assumed. This crack network is based on a close packed hexagonal array of cracks with crack spacing equal to the barrier thickness and the crack opening dimension proportional to the tensile stress normal to the crack plane. This conservative total crack opening area per unit area of seismic damage is used as a scaling factor applied to the seismically damaged areas to obtain the total area through which radionuclides can be potentially released. Depending on Alloy 22 barrier temperature (and resultant yield strength) when the impacts occur, the calculated crack area density can vary from about 1E-02 to 3E-03 or less than about one one-hundredth of the total damaged area.

SLIP DISSOLUTION/FILM RUPTURE MODEL FOR SCC CRACK GROWTH

As stated earlier in this paper, initiation is associated with microscopic crack formation at localized corrosion or mechanical defect sites. Following initiation of single or multiple cracks, SCC develops by transitioning to small crack growth, coalescence of multiple (if initiated) microscopically small cracks, and transition to deep crack response. At any stage, the crack may either arrest or continue to propagate. A lifetime crack propagation prediction model can be developed via a fundamental understanding of the cracking mechanism. For the systems of interest, the slip dissolution/film rupture mechanism has been chosen. This cracking mechanism has been successfully applied to model SCC for stainless steel, low-alloy steel, and nickel-based alloys in light water reactor environments (Ford and Andresen 1988; Andresen and Ford 1988).

For constant load conditions, the crack growth rate V_t (in mm/s) is presented by the following equation:

$$V_t = 7.8 \times 10^{-2} n^{3.6} (4.1 \times 10^{-14})^n (K_I)^{4n} \quad (\text{Eq. 1})$$

where n is the repassivation parameter to be determined experimentally and K_I is the stress intensity factor in $\text{MPa (m)}^{1/2}$. The derivation of Eq. 1 can be found in Lu et al. (2003).

For Alloy UNS-N06022 under constant load condition, the parameter “ n ” can be determined from Eq. 1 based on crack growth rates measured at various levels of applied stress intensity factor, K_I . According to Ford and Andresen (1988), $n = 0.54$ for Type 304 stainless steel in 288°C water. A much higher “ n ” value is expected for Alloy UNS-N06022 because it is highly resistant to SCC. Recent SCC crack growth rate measurements from Andresen et al. (2003) were used for the quantification of the parameter n for the SDFR SCC model to be used for Alloy UNS-N06022. The test data were developed from four Alloy UNS-N06022 compact tension fracture mechanics type specimens tested at 110°C in a fully concentrated mixed salt environment (BSW). The specimens were subjected to cyclic loading in order to initiate crack growth and then followed by constant loading conditions with various hold times. The set of test data to be used as input for establishing the value of n for Alloy UNS-N06022 are summarized in Table 1. These data were selected based on a minimum hold time of 85,400 seconds (or ~24 hours) because Eq. 1 is applicable only to constant loading condition. It is unrealistic to determine the parameter n in this equation based on test data at relative short hold times. The only exception is the data point associated with specimen c144 for which the hold time is relatively short (3,000 seconds or approximately one hour) but cracking appeared to cease, i.e., reaching the constant load state.

Table 1. Summary of source data for Alloy UNS-N06022 SDFR model quantification.

Specimen	Hold time, hours	Tested intensity stress factor, $\text{MPa(m)}^{1/2}$	Measured Crack Growth Rate, mm/s	Calculated "n" value (see Note 3)
c153	CL ^{Note 1}	30	2.50E-10	1.1680
c153	CL ^{Note 1}	30	5.00E-10	1.1190
c144	1	30	1.00E-11 ^{Note 2}	1.3910
c152	24	45	1.00E-11 ^{Note 2}	1.5630
c152	24	45	4.00E-10	1.2810

Notes:

1. CL = Constant Load
2. Growth rate of 1.0E-11 is used for test results where either cracking appeared to cease or the growth rate seemed to arrest.
3. Each of the values in the column "Calculated "n" value" is calculated from Eq. 1.

It can be determined from the n values in the last column of Table 1 that the mean value of n , n_{MEAN} , is 1.304 and the standard deviation (SD), n_{SD} , is 0.160. The base case slip dissolution/film rupture SCC model developed for Alloy UNS-N06022, represented by Eq. 1, is

graphically illustrated in Fig. 4 for n values at 0.984 (-2 standard deviation), 1.304 (mean), and 1.624 (+2 standard deviation), along with test data presented in Table 1 and the graphical representation of Eq. 1 for stainless steel (with $n=0.54$).

Also presented in Figure 4 are independently generated crack growth rate results (listed in Table 2 and obtained at Lawrence Livermore National Laboratory (LLNL) in a broad range of brine environments with pH values between about 2.8 and 10. Excellent resistance to SCC for Alloy UNS-N06022 is clearly illustrated in Fig. 4 where even the higher crack growth rates exhibited by the top curve of Alloy UNS-N06022 with $n=0.984$ (-2 standard deviation) are about 2 orders of magnitude lower than the crack growth rates associated with the stainless steel curve. As can be seen from Figure 4, there is good agreement between the LLNL data and the predictive curves and statistical limits based on the crack growth rate results summarized in Table 1. From Table 2 it can be seen that the prediction error ratio is in the range of -0.93 to 54.87. While the prediction error ratio is generally within the desired 2 orders of magnitude, the prediction model appears to have the tendency of over-prediction (with positive error ratio), i.e., on the conservative side, rather than under-prediction (with negative error ratio). Also, for specimen DCT-22, the measured growth rate is below the crack growth detection limit and thus the comparison between measured and predicted rates is not directly relevant. Furthermore, it can be seen from Figure 4 that measured data fall nicely between the two bounds representing two standard deviations of the mean value. Thus, the good crack growth rate predictive capability of Eq. 1 (benchmarked with the experimental results, presented in Table 1) has been demonstrated with a separate set of measured crack growth rates (see Table 2, column 4). The agreement between the prediction and measured data shown in Figure 4 and quantitatively in Table 2 provides important input to the validation of the SDFR model for Alloy UNS-N06022.

It should be noted that recent crack growth rate data obtained at GE Global Research Center at 40 MPa $\sqrt{m}$ in 150°C SCW solutions on Alloy UNS-N06022 in various conditions (as-welded, as-welded + heat treatments to produce topologically close-packed phases or long range ordering) is similar to these earlier results obtained in BSW at 110°C – the higher temperature, SCW chemistry, and welded/heat treated conditions produced no enhanced SCC growth rates.

Fig. 4. Comparison of the SDFR prediction models for Type 304 stainless steel and Alloy UNS-N06022 with measured data for Alloy UNS-N06022

SDFR CRACK GROWTH MODEL FOR UNS N06022 AT 288°C

Since the SDFR was initially developed for stainless steels and nickel-based alloys Inconel 600/182 under higher temperature (~288°C) light water reactor coolant conditions, additional confidence in the applicability of this model to Alloy UNS-N06022 can be gained from observation of the response of Alloy UNS-N06022 under similar light water reactor coolant conditions. Recent test results performed at the GE Global Research Center (GEGRC) for the Yucca Mountain Program indicate the crack growth rate response of Alloy 22 exposed to 288°C relatively pure water (2 ppm O₂) is broadly consistent with other materials, such as Alloys 600 and 182 (Andresen et al. 2002b) and austenitic stainless steel (Andresen et al. 2002a) under these same conditions. The test results at GEGRC were obtained for Alloy UNS-N06022 forged at room temperature to 21% reduction in thickness, fabricated to a 0.5T CT specimen, and assembled and

tested using techniques identical to those describe in Andresen et al. (2003). The measured crack growth rates show a similar dependency to parameters like corrosion potential and water purity (sulfate). For example, in all test cases, the change in corrosion potential from $\sim +0.2 V_{SHE}$ to $\sim -0.5 V_{SHE}$ (due to a change from 2 ppm O_2 to H_2 -deaerated water) causes a drop of at least one order of magnitude in the crack growth rate, as indicated by Table 3. Also, as expected, Alloy UNS-N06022 shows crack growth rates under repository type oxidizing conditions, i.e., at $0.2V_{SHE}$ which are about one order of magnitude lower than those of the other materials under identical test conditions, demonstrating its superiority as a structural material under conditions where stress corrosion cracking is a concern.

Table 2. Comparison of Predicted and Measured Crack Growth Rates for Alloy UNS-N06022

Specimen ID	Nominal Test Temperature (°C)	AVERAGE Stress Intensity $MPa(m)^{0.5}$	MEASURED CRACK Growth Rate (mm/s)	PREDICTED CRACK Growth Rate (mm/s)	PREDICTION ERROR RATIO
DCT-13	100	45.13	2.12E-09	3.02E-10	-0.86
DCT-14	100	44.88	4.23E-09	2.93E-10	-0.93
DCT-16	100	46.38	1.41E-09	3.48E-10	-0.75
DCT-18	94	45.07	2.12E-10	3.00E-10	0.42
DCT-19	94	45.08	1.41E-11	3.00E-10	20.28
DCT-20	95	45.11	4.23E-10	3.01E-10	-0.29
DCT-21	95	44.68	2.82E-11	2.87E-10	9.18
DCT-22	95	44.37	4.94E-12	2.76E-10	54.87

Notes: (1) Values in Column 6 are obtained from Eq. 1 with $n = 1.304$.

(2) Prediction error ratio (Col. 7) = (Col.. 5 – Col. 6) / Col. 5

Table 3. Measured crack growth rates (mm/s) due to drop in corrosion potential

Alloy	Stress Intensity Factor $MPa(m)^{0.5}$	Corrosion Potential $+0.2 V_{SHE}$	Corrosion Potential $-0.5 V_{SHE}$	Note
Alloy 22	27.5 [25 ksi (in) ^{0.5}]	2.3×10^{-8}	4.0×10^{-9}	Measured data at GEGRC
Alloy 182	28.4	3.0×10^{-7}	6.0×10^{-9}	Andresen et al. (2002b)
Alloy 600	30.0	3.3×10^{-7}	3.7×10^{-8}	Andresen et al. (2002b)
Unsensitized Type 316L Stainless Steel	27.5 [25 ksi (in) ^{0.5}]	4.1×10^{-7}	$>2.0 \times 10^{-8}$	Andresen et al. (2002a)

CONCLUSIONS

This paper deals with the description and validation of the process-level model (see Fig. 2) developed for the performance assessment of the Alloy UNS-N06022 waste package outer barrier subjected to stress corrosion cracking due to weld induced stress in the final closure welds once the planned stress mitigated layer is removed by general corrosion. The slip dissolution/film rupture model relates crack initiation and the subsequent advance to the metal oxidation that occurs when the protective film at the crack tip is ruptured.

As demonstrated previously, the SDFR model with $n = 0.54$ is a good prediction model for sensitized Type 304 stainless steel in 288°C water. Similarly, the good crack growth rate predictive capability of Eq. 1 (benchmarked with the experimental results, presented in Table 1) has been validated with a separate set of crack growth rates measured at LLNL (see Table 2). As can be seen from Figure 4, there is good agreement between the LLNL data and the predictive curves and statistical limits. Fig 4 further indicates that measured data fall nicely between the two bounds representing two standard deviations of the mean value. The agreement between the prediction and measured data shown in Figure 4 provides important input to the validation of the SDFR model for Alloy UNS-N06022.

In the case of SCC in mechanically damaged areas resulting from low probability seismic events, a crack area density model is described that can be applied to conservatively calculate the total crack opening area through which radionuclides can be potentially released.

In conclusion, this paper has clearly demonstrated that the slip dissolution/film rupture model can be applied to assess the breach (or the lack of it) of the high level radioactive-waste packages due to SCC crack propagation of initiated SCC under the combined effects of stress and environment.

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