MODELING STUDY OF PRIMARY WATER STRESS CORROSION CRACKING AT DISSIMILAR WELD OF ALLOY 182 OF PRESSURIZED WATER NUCLEAR REACTOR ACCORDING TO HYDROGEN CONCENTRATION

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ABSTRACT

One of the main failure mechanisms of the pressurized water reactors is the primary water stress corrosion cracking (PWSCC) occurring in the Alloy 600 (75Ni-15Cr-9Fe) and weld metals like the Alloy 182 (67 Ni-15Cr-8Fe). It can occurs, among others, at reactor nozzles dissimilarly welded with alloys 182/82 between steel ASTM A-508 G3 and stainless steel AISI316L. These cracks can cause problems that reduce nuclear safety and reliability. The hydrogen which is dissolved to primary water to prevent radiolysis, can also to have influence on the PWSCC behavior.

One departs from a study of Lima (2011) based in experimental data obtained from CDTN-Brazilian Nuclear Technology Development Center, in slow strain rate test (SSRT). It was prepared and used for tests a weld in laboratory, similar to dissimilar weld in pressurizer relief nozzles, operating at Brazilian Nuclear Power Plant Angra # 1. It was simulated for tests, primary water at 325°C and 12.5 MPa containing five levels of dissolved hydrogen.

The objective of this article is to study and discuss an adequate modeling based on these experimental results, for PWSCC growth rate according to the levels of dissolved hydrogen.

The main result was the graphic modeling Crack Growth Rate *versus* Potential Difference from Ni/NiO Transition which comparing to the similar graphic modeling from EPRI-MRP 263 NP (Marks et al.(2012)), showed a much higher peak of CGR, probably due to the test methodology used in Brazilian case: slow strain rate test, instead of others, as in the case of the MRP 263 NP.

INTRODUCTION

The PWSCC is a complex mode of degradation in Pressurized Water Reactor (PWR) ´s thick-walled components of nickel alloy – such as Alloy 600 and its weld metals (Alloy 182 and Alloy 82). It has been identified as an important mode affecting the safety and reliability of this kind of plant operation. A constant effort has been done to develop and identify technologies to mitigate this damage mechanism. Until now, the main ones are hydrogen optimization and zinc injection, during PWR plant operations. Zinc injection is used to delay the PWSCC initiation due to its incorporation in spinel oxide films, thus enhancing their stability (Nordmann (2012)).

Hydrogen injection on primary water is normally applied to prevent radiolysis. Hydrogen optimization which consists in different hydrogen injection levels in primary water of the operating unity has been demonstrated to strongly mitigate the PWSCC growth rate, mainly in Alloy 182 and Alloy 82 weld metals. With respect to the PWSCC initiation time, most of available data did not show a conclusive effect on this, mainly due to the high degree of scatter. Also, the hydrogen increase above current operational levels did not have an enhancement effect on PWSCC initiation time (Marks et al. (2012)).

Also, according to these authors, the best for this case is zinc addition, that has been demonstrated strong mitigation of the initiation of PWSCC in Alloy 600 (further, the zinc addition is an additional way of decreasing occupational radiation exposure of plant staff, according to Nordmann (2011)).

One departs from a study of Lima (2011) which investigates the influence of dissolved hydrogen contents on the susceptibility to PWSCC of Alloy 182, used as weld metal in a dissimilar weld between the steel ASTM A-508 G3 and stainless steel AISI 316L, similar to the weld which exists in a pressurizer nozzle of Angra # 1 nuclear power plant: it was evaluated. In this study was used a simulated PWR primary coolant water chemistry at 325ºC and pressure of 12.5 MPa with five different levels of dissolved hydrogen: 2, 10, 15, 25, 50 cm^3H2/kg H_2O at standard temperature and pressure (STP). Slow strain rate test (SSRT) was used to evaluate the Alloy 182 PWSCC susceptibility. Open circuit potential was measured in different hydrogen concentrations to evaluate their effect in the material electrochemical corrosion: the main study results indicated that Alloy 182 is less susceptible to PWSCC at 50 cm^3H_2 (STP) /kg H_2O at 325ºC, and showed the positive effect in to keep hydrogen concentration in a high level in the PWR primary coolant water.

The objective of this article is to study and to discuss an adequate modeling based on the experimental results from Lima (2011), for PWSCC growth rate according to the levels of dissolved hydrogen. It has been used from Section 7 of EPRI-MRP 263 NP, a numerical model describing the effect of hydrogen on PWSCC growth rate which takes the form of a Gaussian distribution centered at the Ni/NiO transition. This model is function of the difference in electrochemical potential (ECP) between the Ni/NiO transition and the test condition. Typical fitted parameters are the peak width and the peak to baseline ratio (Marks et al. (2012)).

THE ORIGINAL STUDY DESCRIPTION

The original study comprised: a) Dissimilar material weldment from Angra reactor #1 reproduction in CDTN mechanical workshop, according to the ASME Boiler and Pressure Vessel Code – Section IX, Welding and Brazing Qualification, and the AWS specifications to the welding electrodes; b) Chemical-mechanical-structural characterization of weld and related materials, according to ASTM E4, and ASTM E8; c) Evaluation of the corrosion potential of the Alloy 182 at high-temperature; d) Obtaining and characterization of oxide passive film formed in Alloy 182 on primary water environment at high- temperature; e) SSRT with Alloy 182 specimens at different levels of dissolved hydrogen in the test environment, according to ASTM G 49, and ASTM G 129 (Lima (2011)).

The quantification of the PWSCC brittle fracture surface and its depth was obtained departing from the scanning electron microscope (SEM) micro fractographies of the tested specimens: the images were processed by a Quantikov equipment, and the evaluation of the CGR according to a method developed by Totsuka et al., quoted in the references of Lima (2011): see also pp.63-64 from this study for more information.

IMPORTANT RESULTS OF THE ORIGINAL STUDY TO THE MODELING

The main results of Lima study, concerning the value to modeling data, according to EPRI-MRP 263 NP (Marks et al. (2012)), were the Table 1 and Table 2 below reproduced from this: these Tables correspond graphically to the originals Figure 4.38 (page 100), and Figure 4.37 (page 99) from the study of Lima (2011). In Figure 1, one is reproduced the original Figure 4.38.

In the Table 1, E_{cor} represents the corrosion potential, and $\Delta ECP_{Ni/NiO}$ is the electrochemical potential (ECP) difference from that of the Ni/NiO transition: this is a very important parameter, because changes in the hydrogen concentration in primary water enforce that the corrosion potential reaches the Ni/NiO transition line in the Potential versus pH equilibrium diagram between Ni and water, and have a strong influence in the stress corrosion cracking behavior. In the Table 2, A_{IGSCC} is the brittle fracture surface area ratio to the total fracture surface area, remembering that IGSCC is the intergranular stress corrosion cracking, the predominant type of PWSCC in this case.

Table 1: Resulting values of E_{cor} and $\Delta ECP_{Ni/NiO}$ to Alloy 182 in PWR primary water at 325°C.

Test Environment	E_{cor} (mV_{SHE})	$\Delta ECP_{Ni/NiO}$ (mV_{SHE})
2 cm^3 H_2 (STP)/kg H_2O	-717	-18
10 cm^3 H_2 (STP)/kg H_2O	-735	0
25 cm^3 H_2 (STP)/kg H_2O	-756	21
50 cm^3 H_2 (STP)/kg H_2O	-776	41

Table 2: Crack Growth Rate (CGR) to Alloy 182 in PWR primary water at 325°C.

Dissolved Hydrogen	Crack Depth (mm)	A_{IGSCC} (%)
2 cm^3 H_2 (STP)/kg H_2O	0.836	14
10 cm^3 H_2 (STP)/kg H_2O	1.3	33
15 cm^3 H_2 (STP)/kg H_2O	1.18	25
25 cm^3 H_2 (STP)/kg H_2O	1.04	20
50 cm^3 H_2 (STP)/kg H_2O	0.573	6
Dissolved Hydrogen	Time to failure t_f (h)	CGR (mm/s)
2 cm^3 H_2 (STP)/kg H_2O	324	1.3×10^{-7}
10 cm^3 H_2 (STP)/kg H_2O	216	5.0×10^{-7}
15 cm^3 H_2 (STP)/kg H_2O	273	2.9×10^{-7}
25 cm^3 H_2 (STP)/kg H_2O	278	2.1×10^{-7}
50 cm^3 H_2 (STP)/kg H_2O	384	2.9×10^{-8}

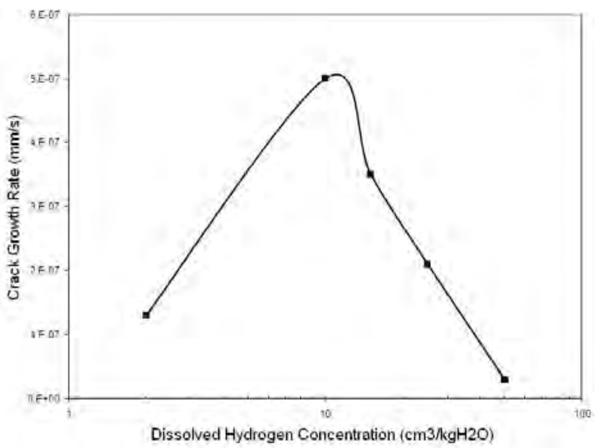

Figure 1. Typical experimental result for Alloy 182 SSR tested at 325°C in primary water with strain rate 3.10^{-7} s^{-1}, from Lima (2011).

MODELING BASIS

The Sections 6 and 7 of EPRI-MRP 263 NP (Marks et al. (2012)) treat about respectively mitigation of PWSCC initiation through elevated hydrogen content, and mitigation of PWSCC propagation through elevated hydrogen content. The conclusion of section 6 is based only in the Alloy 600 case, and is that according to the available data did not exist a hydrogen effect on PWSCC initiation in this nickel alloy: the case of Alloy 182 was not considered. But in Section 7, it was studied the hydrogen effect on Alloy 600, and Alloy 182 (here the interest case). The considered model described the effect of hydrogen on PWSCC propagation in the form of a Gaussian distribution centered at the Ni/NiO transition. This model is function of the difference in ECP between the Ni/NiO transition and the test condition (Equation (1)). Typical fitted parameters are the peak width (c) and the peak to baseline ratio (P). The concentration of hydrogen corresponding to the potential at the Ni/NiO transition is temperature dependent (Equations (2) and (3)).

$$CGR = CGR_{\max}\left[\left[\frac{1}{P}+\frac{(P-1)}{P}\exp\left(\left(-0.5\left(\frac{\Delta ECP_{Ni/NiO}}{c}\right)^{2}\right)\right]\right] \quad (1)$$

$$\Delta ECP_{Ni/NiO} = 29.58\left(\frac{T_{ref}+273.15}{298.15}\right)\log\left(\frac{[H_2]}{[H_2]_{Ni/NiO}}\right) \quad (2)$$

$$[H_2]_{Ni/NiO} = 10^{(0.0111Tref-2.59)} \quad (3)$$

where: CGR is the crack growth rate, CGR_{max} is the maximum CGR at the Ni/NiO transition, P is the ratio of the maximum to minimum expected CGR, c is the peak width, $\Delta ECP_{Ni/NiO}$ is the ECP difference from the Ni/NiO transition, T_{ref} is the reference temperature (in Celsius degrees) in test condition, $[H_2]$ is the hydrogen concentration on environment, $[H_2]_{\mathrm{Ni/NiO}}$ is the $[H_2]$ on the Ni/NiO transition.

On the Table 7-2 of EPRI-MRP 263 NP are presented model parameters for Alloy 182 data sets from various authors, as well as its average and standard deviation values. On the Figures 7-7 to 7-10 of EPRI-MRP 263 NP are presented graphics CGR *versus* $\Delta ECP_{Ni/NiO}$ departing from raw data for Alloy 182 from researchers Andresen and Toloczko (Marks et al. (2012)).

MODELING RESULT

Using the data on the Table 1 and Table 2, one replaced the values on Equations (1) to (3):

● Baseline Ratio P= 5.0 x 10^{-7}/ 2.9 x 10^{-8}= 17. 241

a) Entering with the values from the Table 1 and the Table 2 on Equation (1) one has:

1-CGR=CGR_{max} [1/17.241 + (17.241-1)/17,241x exp (-0.5 $(0)^2$], to DH= 10 cm^3 H_2 (STP)/kg H_2O.

CGR= 0.945 CGR_{max}; to CGR_{max}= 5.0 x 10^{-7}mm/s, one obtained the value CGR= 4.727x10^{-7}mm/s.

To dissolved hydrogen (DH) = 25 cm^3 H_2 (STP)/kg H_2O, Equation (1) resulted:

2- CGR=CGR_{max} [1/17.241 + (17.241-1)/17,241x exp (-0.5 $(21/c)^2$]

Resulting c=15.2

CGR_{max} = 5.0x10^{-7}mm/s = 1.7 mil/day

Let´s to compare with to the Andresen´s raw data, retired from Table 7-1 and Table 7-2 ((Marks et al. 2012)). This comparison is on the Table 3.

Table 3. Comparison between CDTN data (Lima (2011)) with Andresen data (Marks et al. (2012)) to Alloy 182.

	CDTN	Andresen T=325°C	Deviation %
Peak Width c (mV)	15.2	12.1	+ 25.6
Height of CGR_{max} (mils/day)	1.7	0.371	+358.2
Baseline Ratio P	17.2	10.5	+ 63.8

The equation (1) model fit to Alloy 182 raw data from Andresen (red) (Marks et al. (2012)) compared to modeling of the CDTN data (blue) (Lima (2011)), is showed in the Figure 2.

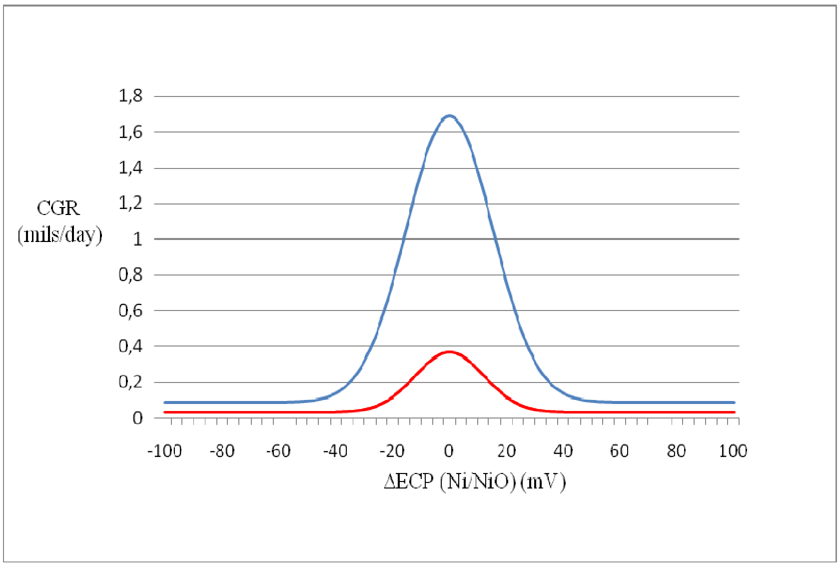

Figure 2. Equation (1) model fit to Alloy 182 raw data from Andresen (red) (Marks et al. (2012)) compared to CDTN data (blue) (Lima (2011)). Plotted through Microsoft Excel 2007.

DISCUSSION

The possible cause of these deviations may be different testing methodologies: although Andresen references are not available on Marks et al. (2012), this researcher normally uses constant load test and "Reverse U-Bend (RUB)" test, instead of SSRT. It also had been used samples of Fracture Mechanics, instead of the cylindrical specimens used in CDTN tests, where there is no control of the stress intensity factor K. The SSRT test is basically an accelerated test where CGR values are greater than the constant load tests CGR values. So, the deviations of CGR values obtained at CDTN, are upwards and consequently the P-parameter (baseline ratio) is greater than the P-parameter obtained for Andresen raw testing data. Nevertheless modeling is valid qualitatively for testing according SSRT in CDTN, although apparently there are no SSRT data sets for comparison in the EPRI –MRP-263 NP (Marks et al. (2012)).

Another explanation why the P-parameter is greater in SSRT than the same at constant load tests is given by some authors as Rios, Magnin and others (Hua and Rebak (2008)): these proposed a corrosion-enhanced plasticity model where there is a strong interaction between corrosion and local crack tip plastic conditions. If the plasticity in constant load test is enhanced in SSRT, then the hydrogen could enhance the corrosion, accelerating the CGR.

Based on this issue, the recommendation to EPRI is to add a comparative data set done in SSRT at MRP-263 NP to check the comparisons of the P-values.

In the case of CDTN tests it would be very interesting to do constant loads tests, and tests using the Fracture Mechanics specimens, not only to compare quantitatively with MRP-263 NP, but also to do the complete modeling of the stress corrosion cracking propagation rate with the hydrogen influence, which could be done using the Equation (4) given in EPRI –MRP-263 NP (Marks et al. (2012)). This Equation is a product of material effects, stress intensity effects, temperature effects, and hydrogen effects, as showed below.

$$CGR = \alpha f_{weld}(K - K_{th})^{\beta} \exp\left[\frac{-Q}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right]\left[\frac{1}{P} + \frac{(P-1)}{P}\exp\left(\left(-0.5\left(\frac{\Delta ECP_{Ni/NiO}}{c}\right)^{2}\right)\right)\right] \tag{4}$$

where: CGR is the crack growth rate, α is a material constant, f_{weld} is the weld factor, K is the stress intensity, K_{th} is the stress intensity threshold, β is an exponent, Q is the activation energy, R is the universal gas constant, T is the absolute temperature, T_{ref} is the absolute reference temperature, P is the ratio of the maximum to minimum expected CGR, c is the peak width, $\Delta ECP_{Ni/NiO}$ is the ECP difference from the Ni/NiO transition.

One can try another modeling, such as the one using a propagation model as detailed in MRP-307- Marks et al. (2011). In this document, the model proposed is also according Equation (4). It can be used also the Fracture Research Institute (FRI-Japan) model, which includes the effects of material stress and strain characteristics and CGR on the crack tip strain rate (Eason (2008)). In this document are included data sets with substantial range in dissolved hydrogen, and also it allows to compare the hydrogen variation/ΔECP submodels for weld metals. In EPRI-MRP-115 (White et al. (2004)) there is a data collection of tests from several world laboratories which has been used to develop the CGR curves for the weld metals selected for use with Alloy 600 base material (Alloy 82, 182, and 132): but in this case is not provided the hydrogen variation effect. However, is necessary to do more tests according to the stress intensity variation to input adequate data in these models.

CONCLUSIONS AND REMARKS

It had been showed in this article that is possible to apply the dissolved hydrogen submodel exposed on EPRI-MRP-263 NP to an experimental data set for Alloy 182 with hydrogen variation on primary water environment, realized at CDTN-Brazil: qualitatively this data set is according to the EPRI document, but not quantitatively; more efforts should be done to compare adequately SSRT data sets, and more experimental data with stress intensity variation should be done to do the complete stress corrosion modeling as proposed in the EPRI documents. This research work is in progress to put a modeling objective through Brazilian experimental data, according to the EPRI recommendations, and other global data comparisons.

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