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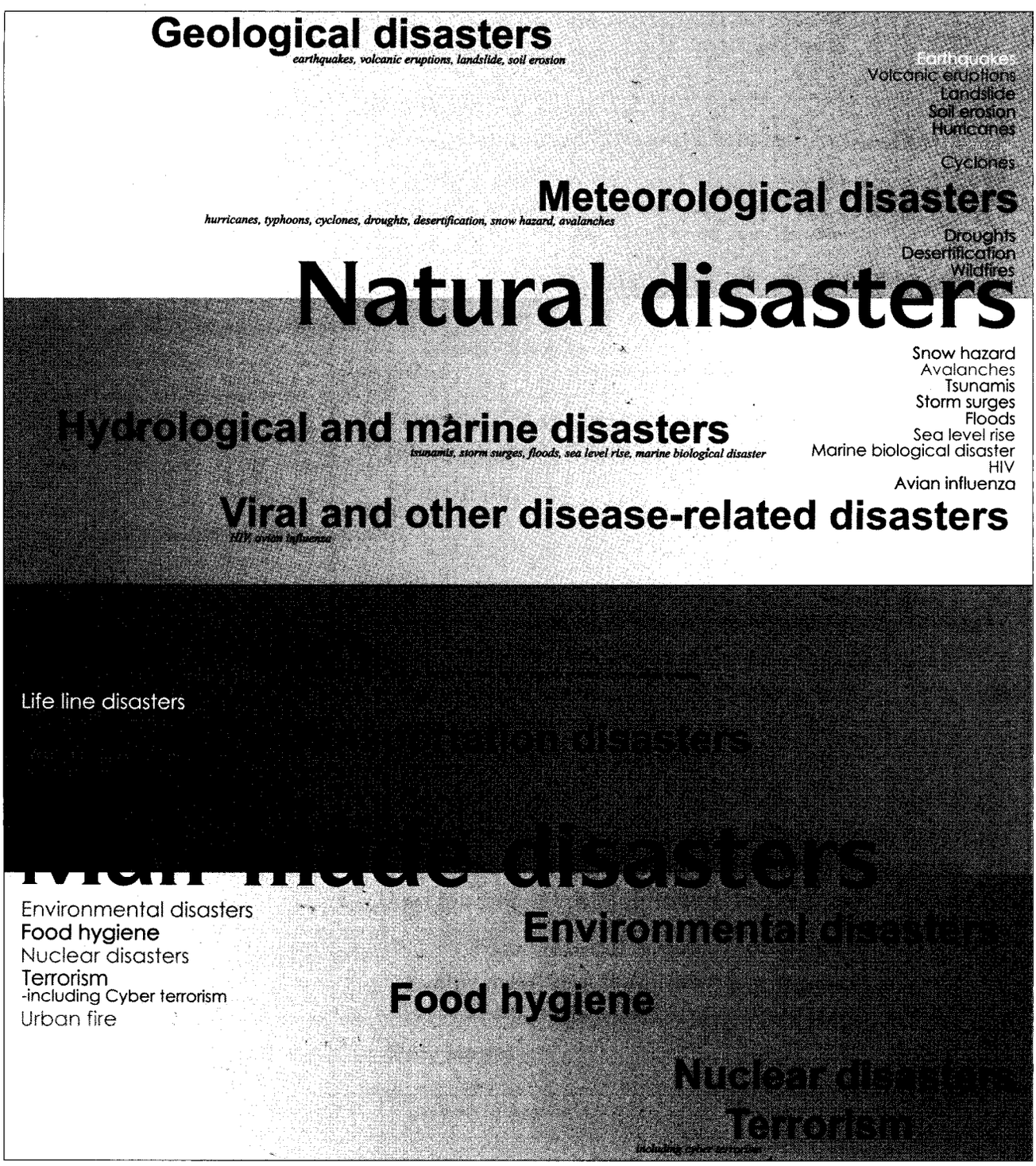

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Paper:

Compared Modeling Study of Primary Water Stress Corrosion Cracking at Dissimilar Weld of Alloy 182 of Pressurized Water Nuclear Reactor According to Hydrogen Concentration

Omar F. Aly*, Miguel M. Neto*, Mônica M. A. M. Schvartzman, and Luciana I. L. Lima*****

*IPEN-Energy and Nuclear Research Institute
2242 Av. Lineu Prestes, São Paulo 05508-000, Brazil
E-mail: ofaly@ipen.br; ofaly1@gmail.com
**CDTN-Nuclear Technology Development Center, Belo Horizonte, Brazil
***Vallourec Research and Development-Corrosion, Belo Horizonte, Brazil
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One of the main failure mechanisms of pressurized water reactors (PWR) is primary water stress corrosion cracking (PWSCC), which occurs in alloy 600 (75Ni-15Cr-9Fe) and weld metals such as alloy 182 (70Ni-14Cr-9Fe), and alloy 82 (73Ni-19Cr-2Fe). Corrosion cracking is due, for example, in reactor nozzles welded dissimilarly with alloys 182/82 between ASTM A-508 G3 steel and AISI316L stainless steel. Corrosion cracks can cause problems reducing nuclear installations safety and reliability. Hydrogen dissolved into primary water to prevent radiolysis, also may enhance PWSCC growth. This article begins from a study by Lima et al. (2011) based on experimental data from the CDTN-Brazilian Nuclear Technology Development Center, and related to a slow strain rate test (SSRT). This was prepared and used for testing welds in the laboratory, similar to the dissimilar weld in pressurizer relief nozzles operating at the Brazilian Angra Unit 1 nuclear power plant. It was simulated for tests, primary water at 325°C and 12.5 MPa containing four levels of dissolved hydrogen. Our objective in this article is to clarify, and discuss adequate modeling based on the SSRT experimental results, and to compare them with those from another database and modeling, of the PWSCC growth rate based on levels of dissolved hydrogen.

Keywords: dissolved hydrogen effects, modeling, nickel alloys, pressurized water reactors, stress corrosion cracking

1. Introduction

Primary Water Stress Corrosion Cracking (PWSCC) is a complex mode of degradation that occurs in the pressurized water reactor (PWR)'s thick-walled components composed of nickel alloy – such as alloy 600 and its weld metals (alloy 182 and alloy 82). It has been identified as an important mode of degradation affecting plant operation safety and reliability. Constant efforts have been done to develop and identify technologies to mitigate this damage mechanism. Those developed thus far include hydrogen optimization and zinc injection, during PWR plant operations. Zinc injection delays PWSCC initiation due to its incorporation in spinel oxide film, to enhance film stability [1].

Hydrogen injection in primary water is applied to prevent radiolysis, that is the dissociation of molecules by ionizing radiation. Hydrogen optimization, which consists of different hydrogen injection levels in primary water has been demonstrated to strongly mitigate the PWSCC growth rate, mainly in alloys 182 and 82 weld metals. Most available data do not show a conclusive effect of PWSCC initiation time on these, mainly due to high scattering. Also, the hydrogen increased above normal operating levels, did not enhance PWSCC initiation time [2].

This article departs from the study by Lima et al. [3], which investigates the influence of dissolved hydrogen on the susceptibility to PWSCC of alloy 182, used as a weld metal in a dissimilar weld between ASTM A-508 G3 steel and AISI 316L stainless steel, similar to welds in the pressurizer nozzle of the Angra Unit 1 nuclear power plant. In this article we used simulated PWR primary coolant water chemistry at 325°C and 12.5 MPa of pressure with four different levels of dissolved hydrogen: 2, 10, 25, and 50 cm^3 H_2/kg H_2O at standard temperature and pressure (STP). A slow strain rate test (SSRT) was used to evaluate alloy 182 PWSCC susceptibility. Open circuit potential was measured at different hydrogen concentrations to evaluate their effect on electrochemical corrosion of the material. Main study results indicated that alloy 182 is little susceptible to PWSCC at 50 cm^3 H_2 (STP)/kg H_2O at 325°C, and showed a positive effect in maintaining hydrogen concentration at a high level in PWR primary coolant water.

Our objective in this article is to study, compare and discuss adequate modeling based on the experimental results of Lima et al. [3], as these related to the PWSCC growth rate based on dissolved hydrogen levels. From Section 7 of EPRI-MRP 263 NP, a numerical model

has been used to describe the effect of hydrogen on the PWSCC growth rate, which takes the form of a Gaussian distribution centered on the Ni/NiO transition. This model is represented by a function of the difference in electrochemical potential (ΔECP) between the Ni/NiO transition and the test condition. Typical fitted parameters are peak width (c) and peak to baseline ratio (P) [2].

2. Original Study Description

The original study by Lima et al. [3], comprised the following:

a) Dissimilar material welded from Angra reactor unit 1 reproduction in the CDTN mechanical workshop, based on ASME Boiler and Pressure Vessel Code – Section IX, Welding and Brazing Qualification, and AWS specifications for welding electrodes.

b) Chemical, mechanical, and structural characterization of welds and related materials, based on ASTM E4, and ASTM E8;

c) Evaluation of alloy 182 corrosion potential at high temperatures;

d) Acquisition and characterization of oxide passive film formed in alloy 182 in a primary water environment at high temperature;

e) SSRT with alloy 182 specimens at different levels of dissolved hydrogen in the test environment, based on ASTM G 49, and ASTM G 129 [3].

The PWSCC brittle fracture surface and its depth were quantified departing from scanning electron microscope (SEM) micro-fractographies of tested specimens (approximately cylindrical as showed in Figures 13 to 16, reference [3]: images were processed using Quantikov equipment, and the crack growth rate (CGR) was evaluated based on a method developed by Totsuka et al. [4, 5] quoted by Lima et al. [3]: it consists basically of evaluating the crack surface and its depth around the specimen's cross bar, and of calculating the CGR as the crack depth multiplied by the IGSCC surface area divided by the product between of the total surface area and time to failure.

3. Important Results of the Original Study

The main results of the study by Lima et al. [3], on the modeling data based on EPRI-MRP 263 NP [2], are shown in **Tables 1** and **2**, reproduced from [3];

E_{cor} in **Table 1** represents corrosion potential. $\Delta ECP_{Ni/NiO}$ 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 force that the corrosion potential reaches the Ni/NiO transition, and consequently have a strong influence in the stress corrosion cracking behavior.

Table 1. Values of E_{cor} and $\Delta ECP_{Ni/NiO}$ of alloy 182 in PWR primary water at 325°C [3].

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. *CGR* of alloy 182 in PWR primary water at 325°C [3].

Dissolved Hydrogen	Deepest Crack (mm)	A_{IGSCC} (%)
2 cm^3 H_2(STP)/kg H_2O	0.84	14
10 cm^3 H_2(STP)/kg H_2O	1.30	33
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}
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}

A_{IGSCC} in **Table 2** is the brittle fracture surface area ratio with the total fracture surface area. Note that the intergranular stress corrosion cracking (IGSCC) is the predominant type of PWSCC in this case.

When dissolved hydrogen grows from 2 to 50 cm^3 H_2(STP)/kg H_2O, the CGR increases and the time to failure decreases to a maximum value at 10 cm^3 H_2(STP)/kg H_2O – this maximum peak is caused by reaching the Ni/NiO potential – then decreases to a very low value at 50 cm^3 H_2(STP)/kg H_2O.

4. Basis of Modeling

Section 6 of EPRI-MRP 263 NP [2] treats the mitigation of PWSCC initiation through elevated hydrogen content and Section 7 treats the mitigation of PWSCC propagation through elevated hydrogen content. The conclusion in Section 6 is based only on the case of alloy 600, that is based on available data, no hydrogen effect existed on PWSCC initiation in this nickel alloy: the case of alloy 182 was not considered. Section 7 deals with the hydrogen effect on alloys 600 and 182 – alloy 182 being of interest here. The hydrogen effect model describes the effect of hydrogen on PWSCC propagation in the form of a Gaussian distribution centered on the Ni/NiO transition. This model is a function of the difference in ECP between the Ni/NiO transition and the test condition (Eq. (1)). Typical fitted parameters are the peak width (c) and the peak to baseline ratio of the maximum to minimum expected CGR (P). The maximum CGR in this case is near the Ni/NiO transition, 5.0×10^{-7} mm/s corresponding to the

10 cm^3 H_2(STP)/kg H_2O hydrogen dissolved point in **Table 2**. The minimum CGR is 2.9×10^{-8} mm/s that corresponds to the 50 cm^3 H_2(STP)/kg H_2O hydrogen dissolved point in **Table 2**. The concentration of hydrogen corresponding to the potential at the Ni/NiO transition is temperature dependent (Eqs. (2) and (3)).

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

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

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

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

In 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. In Figures 7-7 to 7-10 of EPRI-MRP 263 NP (pages 7-10 to 7-11) are presented graphics CGR versus $\Delta ECP_{Ni/NiO}$ departing from raw data for alloy 182 from researchers Andresen (quoted references [58] and [61] from reference [2]) and Toloczko (quoted in reference [64] from reference [2]).

5. Modeling Result

Values in Eqs. (1) to (3) are replaced using the data from **Tables 1** and **2** as follows:

· Baseline Ratio $P = 5.0 \times 10^{-7}/2.9 \times 10^{-8} = 17.241$.

a) Values from the **Tables 1** and **2** in Eq. (1) yields:

1) To dissolved hydrogen DH = 10 cm^3 H_2(STP)/kg H_2O, Eq. (1) yields:

$$CGR = CGR_{\max}[1/17.241 + (17.241-1)/17.241 \times \exp(-0.5(0)^2)]$$

$$CGR = CGR_{\max} = 5.0 \times 10^{-7} \text{ mm/s}.$$

2) To dissolved hydrogen DH = 25 cm^3 H_2 (STP)/kg H_2O, Eq. (1) yields:

$$CGR = CGR_{\max}[1/17.241 + (17.241-1)/17.241 \times \exp(-0.5(21/c)^2)]$$

3) Resulting the peak width.

· $c = 15.2$ to $CGR_{\max} = 5.0 \times 10{-}7$ mm/s = 1.7 mil/day, and to $CGR_{\min} = 2.9 \times 10^{-8}$ mm/s.

Table 3. Comparison of CDTN data [3] to Andresen's data [2] 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

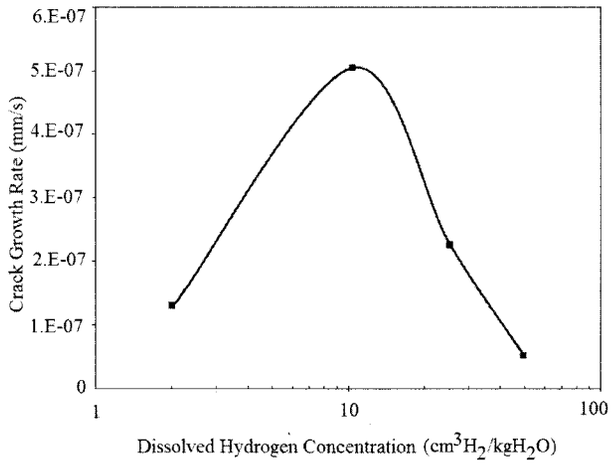

Fig. 1. Typical experimental results for alloy 182 SSR tested at 325°C in primary water with strain rate 3.10^{-7} s^{-1} [3].

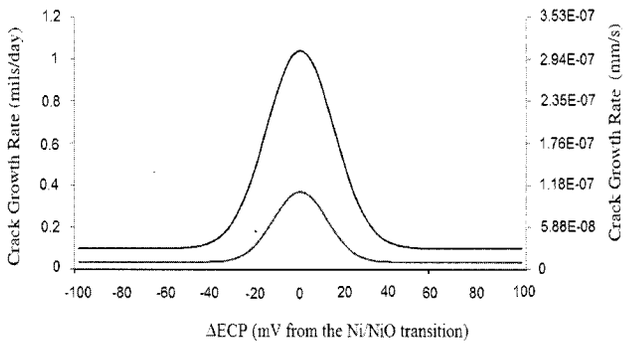

Fig. 2. Eq. (1) model fitted to raw alloy 182 data from Andresen in red [2] compared to the CDTN data in blue [3].

Note here that the possibility exists that different c values could be drawn from CDTN test data at different H_2 levels: if DH = 2 cm^3 H_2 (STP)/kg H_2O was used, the c value yields 10.3. This indicates as expected, a non-deterministic relationship based on Eq. (1), and better modeling should be obtained with least squares data regression determining the (P,c) normal curve as applied in [2]. The approach to modeling that we used here is immediate but somewhat rough, and we used it only in a quick engineering application.

Table 3 Compares CDTN results to Andresen's raw data, from Tables 7-1 and 7-2 [2].

The Eq. (1) model fits alloy 182 raw data from Andresen in red – Fig. 7-7 page 7-10 from reference [2] compared to modeling of CDTN data in blue [3], as shown in **Fig. 2**.

Note that the blue curve in **Fig. 2**, is the modeled curve of results presented in **Fig. 1**, referring to data obtained at the CDTN.

Note also that the profile of the plotted blue curve in **Fig. 2** differs from the curve in **Fig. 1**, because the blue curve is a statistical regression for data in **Table 2** plotted in **Fig. 1**.

6. Comparison of Data and Modeling

For other important model databases, such as from EPRI-MRP-115 [6], USNRC [7], stress intensity factor (K) evaluation is necessary to proceed to comparison. The SSRT of the CDTN was not performed with controlled stress intensity factor (K) variation, but it is possible to estimate an approximate K-value for this test. To estimate this K-value from Eq. (4) which represents K in mode I to a fully circumferential crack in a bar [8], we consider a simplification related to the actual SSRT results, because the axial crack depth after SSRT is not completely constant: note that the specimens are cylindrical round bars approximately fully circumferential crack in a bar, as shown in Figures 13 to 17 from reference [3].

$$K_I = (Y\sigma)\sqrt{\pi a} \quad \ldots\ldots\ldots\ldots \quad (4)$$

where: σ = stress, a = crack depth, Y = factor representing the total contribution of primary and secondary stresses.

Values considered for being used in Eq. (4) were: σ = 440 MPa, $a = 1.04 \times 10^{-3}$ m – both values corresponding to the case where the hydrogen concentration is equal to 25 cm^3H_2/kg H_2O – see Fig. 12 from Lima et al.[3], showing stress versus strain curves obtained on SSRT at the CDTN: the stress value used is the average stress of the SSRT applied to the specimen at 325°C; the value for $Y \approx 1$, considering contribution of primary stress far greater than the contribution of secondary stress. The estimated K-value is then 25.15 MPa$\sqrt{}$m.

EPRI-MRP-115 and USNRC databases {crack growth rate, stress intensity}are compared in reference [9]: EPRI disposition curves show good accordance with Brazilian CDTN data point estimation at 25 cm^3H_2/kg H_2O and 25°C, equal to $\{2.1 \times 10^{-7}$ mm/s; 25 MPa$\sqrt{}$m$\}$. The EPRI model could be used in principle to evaluate CDTN tests: more of these tests are required to confirm EPRI disposition curves. For the USNRC model results of the CDTN experimental CGR-value compared to the corresponding modeling value based on the USNRC, is about 9.5% higher. For details see reference [9].

Further example of how is done these comparisons (such as in reference [9]): we choose another model to compare with Brazilian CDTN tests. This is the EdF (Eletricité de France) model based on Eq. (5) [6].

$$CGR = 2.2 \times 10^{-10}(K-9)^{0.1} \quad \text{[m/s]} \ldots\ldots \quad (5)$$

where: CGR is the crack growth rate in m/s; and K the stress intensity factor in MPa$\sqrt{}$m.

Replacing K equal to the estimated K-value of 25.15 MPa$\sqrt{}$m corresponding to the value estimated for Brazilian CDTN tests, results in $CGR = 2.9 \times 10^{-7}$ mm/s. The evaluation for the CDTN case was $CGR = 2.1 \times 10^{-7}$ mm/s. However, so for the considered actual point of CDTN data, the EdF model is 38.1% less conservative. But is only an estimate: so, more points should be obtained by using fracture mechanics specimens at the CDTN, to construct a true model curve.

7. Discussion

In discussing the previous sections, we would first like to comment that the tests used for modeling were not obtained under the same conditions as for EPRI-MRP 263 NP [2]. In this it was been used samples of fracture mechanics, rather than the cylindrical specimens used in CDTN tests, where there is no control of the stress intensity factor K. This is a fact that could harm experimental results – such as scattering – because P and K values are not complete independent, as required by the modeling theoretically expressed in Eq. (1). The modeling done here should also be improved using c and P regression of the normal curve using the least squares method, as suggested by a peer reviewer, and done in EPRI-MRP 263 NP [2].

Notwithstanding these issues, the possible cause of deviations in the EPRI-MRP 263 NP hydrogen model [2] result (Section 5), may be the different testing methodologies involved. Although Andresen references are not available in reference [2], Andresen usually uses constant load tests and "reverse U-bend (RUB)" tests, rather than the SSRT.

The SSRT is basically an accelerated test in which CGR values may exceed constant load tests CGR-values. The deviations of CGR-values obtained in the CDTN are therefore upwards related to Andresen's results and consequently the P-value (baseline ratio) is greater than the P-value obtained for Andresen raw testing data. Modeling is nevertheless qualitatively valid for testing based on SSRT in the CDTN, although apparently there are no SSRT data sets for comparison in EPRI-MRP-263 NP [2].

Another explanation for why the P-value is greater in SSRT than the same in constant load tests is given by authors such as Rios, and Magnin [10], who propose a corrosion-enhanced plasticity model in which a strong interaction exists between corrosion and local crack tip plastic conditions. If plasticity in the constant load test is enhanced in SSRT, then hydrogen could enhance corrosion, accelerating the CGR. Notwithstanding this possibility, these CGR differences should be more investigated, through more comparisons between Andresen's tests, and SSRT.

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

In the case of CDTN tests it would be very interesting to do constant loads tests and tests using the frac-

ture mechanics specimens, both to be compared quantitatively with MRP-263 NP, and to completely model of the stress corrosion cracking propagation rate with the hydrogen influence, which could be done using Eq. (1) in EPRI-MRP-263 NP [2]. This equation is a product of effects of materials, stress intensity, temperature, and hydrogen, as showed below.

$$CGR = \alpha f_{\text{weld}} (K - K_{\text{th}})^{\beta} \exp\left[\frac{-Q}{R}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right] \times \left[\frac{1}{P} + \frac{P-1}{P} \exp\left(\left(-0.5\left(\frac{\Delta ECP_{Ni/NiO}}{c}\right)^2\right)\right)\right] \quad (6)$$

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

Other modeling could be tried, such as that using a propagation model as detailed in MRP-307 [11]. In this MRP, the model proposed is also based on Eq. (1). The Fracture Research Institute (FRI-Japan) model can also be used, which includes the effects of material stress and strain characteristics and CGR on the crack tip strain rate [12]. The MRP-307 contains data sets with a substantial range in dissolved hydrogen, and also enables the hydrogen variation/ΔECP models for weld metals to be compared. In EPRI-MRP-115 [13], there is a data collection of tests from several laboratories from around the world that has been used to develop CGR curves for the weld metals selected for use with the alloy 600 base material (alloys 82, 182, and 132) – but in this case the hydrogen variation effect is not provided. More adequate tests base on the stress intensity variation are necessary to input adequate data in these models.

Concerning other important modeled databases, such as from EPRI-MRP-115 [6], USNRC [7], and EdF [6], that consider crack propagation rate versus stress intensity variation (reference [6]), it is possible to estimate K-values for SSRT results, as done in Section 6: in Brazilian CDTN tests a K-value was estimated to be compared as a single point with modeled curves in references [6] and [7]. This K-value estimative has been shown to be reasonable, with some deviations according mainly to USNRC (less conservative) or EdF (more conservative). To confirm these deviations, however a complete curve should be constructed with Brazilian CDTN tests using fracture mechanics specimens.

8. Conclusions and Remarks

It had been shown that is it possible to apply a dissolved hydrogen model exposed on EPRI-MRP-263 NP to an experimental data set for alloy 182 with hydrogen variation in a primary water environment, realized at CDTN-Brazil. Qualitatively this data set is based on the EPRI document. More work is needed to compare SSRT data sets adequately, and more experimental data with stress intensity variation should be done in complete stress corrosion modeling as proposed in EPRI documents. The modeling we have shown here should be improved through an adequate regression of the normal curve through a proper adjustment of c and P values using the least squares method. It is also possible to estimate a pair of CGR and K-values for Brazilian tests, and to compare it to models considering K variation. In this case also, more tests should be done to construct Brazilian test disposition curves, which should then be compared adequately to these existing models. Research should now be put in progress based on EPRI recommendations, and other global data comparisons. The final future consideration is to propose a simulation program for SCC in such plant components, based for example on reference [15].

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The paper that this one continues, is originally to be published (in press) in SMiRT 22 Transactions [14], and with similarity to reference [9], and contains improved changes to text, and an additional discussion. It also includes a new Section 6 that added the EdF model comparison with ours. These three papers use the same experimental database.

Lastly we thank the four anonymous JDR peer reviewers for their excellent reviews, which significantly improved this article.

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Name:
Omar Fernandes Aly

Affiliation:
Post-Doctored Researcher, IPEN/CNEN-USP São Paulo University, SP, Brazil

Address:
2242 Avenida Lineu Prestes, São Paulo 05508-000, Brazil

Brief Career:
1978-1999 EMAE/Eletropaulo/Light Electricity Services: Maintenance Mechanic Engineer at Thermoelectric Power Plant
2006- Doctor in Science-Nuclear Materials Technology at IPEN
2013- Post Doctor at IPEN, Nuclear Engineering Center
2013-2015 Volunteer Researcher at IPEN, Nuclear Engineering Center

Selected Publications:
• O. F. Aly, M. M. Neto, and M. M. A. M. Schvartzman, "Stress Corrosion Prediction Modeling Software – A Proposal," Recent Patents on Corrosion Science, Vol.2, No.2, pp. 112-117, 2012.

Name:
Miguel Mattar Neto

Affiliation:
Senior Researcher, Engineer, Nuclear and Energy Research Institute, IPEN-CNEN/SP

Address:
2242 Av. Prof. Lineu Prestes, Cidade Universitária, CEP 05508-000 São Paulo, Brazil

Brief Career:
1991- Ph.D. in Structural Engineering, University of Sao Paulo, Brazil

Selected Publications:
• "On the structural integrity assessment of cracked piping of PWR nuclear reactors primary systems," Progress in Nuclear Energy, Vol.50, Issue 7, pp. 800-817, September 2008.

Academic Societies & Scientific Organizations:
• Brazilian Society of Mechanical Sciences and Engineering (ABCM)
• Brazilian Society of Nuclear Energy (ABEN)

Name:
Mônica Maria de Abreu Mendonça Schvartzman

Affiliation:
Senior Researcher, Engineer, Centre of Nuclear Technology Development, CDTN, CNEN/MG

Address:
6627 Av. Pres. Antônio Carlos, Pampulha, CEP 31270-901, Belo Horizonte – MG, Brazil

Brief Career:
1999- Ph.D. in Chemical Engineering, University of Campinas – UNICAMP, São Paulo, Brazil

Selected Publications:
• "Stress corrosion cracking behavior of Alloy 182 weld in PWR primary water environment at 325°C," Corrosion, Vol.67, Issue 8, pp. 085004-1-085004-9, August 2011.

Academic Societies & Scientific Organizations:
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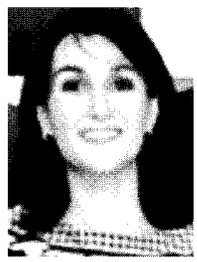

Name:
Luciana Iglésias Lourenço Lima

Affiliation:
Engineer, Vallourec Competence Center

Address:
485 Rua Paulo Emídio Barbosa, Parque Tecnológico, Cidade Universitária – CEP, 21941-615 Rio de Janeiro – RJ, Brazil

Brief Career:
2011- Ph.D. in Mechanical Engineering, Federal University of Minas Gerais, joined Vallourec Brazil

Selected Publications:
• "Stress corrosion cracking behavior of Alloy 182 weld in PWR primary water environment at 325°C," Corrosion, Vol.67, Issue 8, pp. 085004-1-085004-9, August 2011.