RESULTS ON MODELING OF PRIMARY WATER STRESS CORROSION CRACKING AT CONTROL ROD DRIVE MECHANISM NOZZLES OF PRESSURIZED WATER REACTORS

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ABSTRACT

One of the main failure mechanisms that cause risks to pressurized water reactors (PWR) is the primary water stress corrosion cracking (PWSCC) occurring in alloys like the alloy 600 (75Ni-15Cr-9Fe). It can occur, besides another places, at the control rod drive mechanism (CRDM) nozzles. It is caused by the joint effect of tensile stress, temperature, susceptible metallurgical microstructure and environmental conditions of the primary water. These cracks can cause problems that reduce nuclear safety by blocking the displacement of the control rods and may cause leakage of primary water that requires repair or replacement of the reactor pressure vessel head. In this work it is performed a study of the existing models and proposed a new approach to assess the primary water stress corrosion cracking in nickel-based Alloy 600 CRDM nozzles . The proposed model is obtained from the superposition of electrochemical and fracture mechanics models, and validated using experimental and literature data. The experimental data were obtained from CDTN-Brazilian Nuclear Technology Development Center, in a SSRT equipment, according with Schvartzman et al.(2005).

Staehle (1992) has built a diagram that indicates a thermodynamic condition for the occurrence of some PWSCC submodes in Alloy 600: it was used potential x pH diagrams (Pourbaix diagrams), for Nickel in high temperature primary water (300^0C till 350^0C). The PWSCC submodes were located over it, using experimental data. Also, a third parameter called "stress corrosion strength fraction" was added. However, it is possible to superimpose to this diagram, other parameters expressing PWSCC initiation or growth kinetics from other models.

It is important to mention that the main contribution of this work is from a specific experimental condition of potencial versus pH, it was superposed, an empiric-comparative, according with Staehle (1992), a semi-empirical-probabilistic according with Gorman et al. (1994), an initiation time according with Garud (1997), and a strain rate damage according with Boursier et al.(1995)-models, to quantify respectively the PWSCC susceptibility, the failure time, and in the two lasts, the initiation time of stress corrosion cracking. The results were compared with the literature and it showed to be coherent. From this work was obtained a modeling methodology from experimental data.

The SSRT tests had been realized at a condition of potential =–621 mV_{SHE} and pH= 7.3. The PWSCC strength fraction evaluated was 0.95: this initiates an empirical-comparative model.

The initiation time model obtained was according Eq. (1) with t_i in days, T in K, and σ in MPa. The model was planned for constant load, but some assumptions were done to obtain (1) from slow strain rate tests.

$$\mathbf{t_i = 4{,}88.\ 10^{-23}.\ exp\ (32822,\ 35/T).\ ln\ [1{,}79\ (278{,}5/\sigma)]} \quad \mathbf{(1)}$$

The strain rate damage model obtained was in according to Eq. (2) with t_i in days, $\dot{e}_{SSRT}$ in s^{-1} for primary water temperature 303^0C.

$$\mathbf{t_i = 8{,}28.\ 10^{-3}.\ \dot{e}_{SSRT}^{\ -0{,}67}} \quad \mathbf{(2)}$$

The semi-empirical-probabilistic model was obtained only for its deterministic part since there wasn't enough number of tests for modeling the probabilistic part. Notwithstanding some assumptions can be done over it and compared with literature.

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1 INTRODUCTION

Materials degradation during operation-mainly corrosion, fatigue and irradiation-represents one of the main technological factors that may restrain the reliability and availability of nuclear power plants [1]. One of the main degradation modes that cause risks to PWR is the PWSCC. In the CRDM nozzles of PWR, these cracks, that may be axial or circumferential, may cause problems that reduce nuclear safety and reliability like coolant leaks [2], nozzle components ejection and blocking of the rod drive mechanism [3]. Leakage of coolant/primary water can yet causes general corrosion in the low-alloy vessel head by boron deposits. Most of the western PWR have CRDM penetration in the pressure vessel head made of stainless steel and Alloy 600. This superalloy composition is mainly 72% Ni (min), 14-17 % Cr, 6-10 % Fe [3], [4], [5]. The yield strength of this material varies between 213 and 517 MPa. Normally this material is mill annealed at 885^0C, final anneal for 4 to 6 hours followed by air cooling. Nevertheless this treatment could vary, depending on the vendors. This material operates, with some variation, at 315^0C and 15.5 MPa in pure water [3]. The PWSCC appears in the lower part of each nozzle that is fabricated in Alloy 600 and welded to the internal vessel head surface with dissimilar material Alloy 182. There are typically 40 to 90 penetrations per vessel that may include some spare penetrations which are not fitted with CRDM or through core instrumentation of a PWR [6].

2 MODELS AND MODELING

Stress corrosion cracking initiation and propagation are very complex phenomena, one modality of environmentally assisted cracking (EAC), besides corrosion fatigue and hydrogen embrittlement, depending on several parameters that can be classified in microstructural, mechanical and environmental [7],[8]. The microstructural ones are: (1) grain boundary microchemistry and segregation M; (2) thermal treatment TT which can causes intragranular and intergranular metallic carbide distribution; (3) grain size gs and cold work CW or plastic deformation: these two last ones fix the yield stress σ_{ys}. Mechanical ones are: (4) residual stress σ_r; (5) applied stress σ_a – the tensions referred and geometry can be summarized as applied stress intensity factor K_I ; (6) strain ε and strain rate $\dot{\varepsilon}$. Environmental factors include: (7) temperature T; (8) activity of $[H]^+$ or pH; (9) solution or water chemistry SC; (10) inhibitors or pollutants in solution; (11) electrochemical potential V; (12) partial pressure of hydrogen p_{H2} [9]. This environmental cracking susceptibility can be expressed as Eq. (1) [10].

$$\mathbf{SCC=f(M,TT,gs,CW,K_I,\varepsilon,\dot{\varepsilon},T,pH,SC,V,p_{H2})} \quad (1)$$

There are several processes by which above conditions at grain boundaries can be lead to SCC: depassivation caused by film rupture, pitting starting SCC process, grain boundaries weakened by bubbles and creep, hydrogen embrittlement, etc. For more details, see [8], [11].

Thus, there are several approaches to mathematically express these phenomena: the slip dissolution / film rupture of Ford and Andresen [12], the enhanced surface mobility theory of Galvele [13], coupled environment fracture model of Macdonald and Urquidi-Macdonald [14], the internal oxidation mechanism of Scott and Le Calvar [15], numerical model of Rebak and Smialowska [10], hydrogen induced cracking models of Shen and Shewmon, Magnin and others [10]. For a comprehensive review of several of these models see [10]. Mainly for hydrogen action models see [16] and [17]. Two kinetic models, a semi-empirical–probabilistic one and a deterministic strain rate damage model [18], [19] are chosen to compose the present proposal. The semi-empirical-probabilistic model follows Eq. (2) and (3) [19].

$$t_f = A\, t_{ref} \left(\frac{\sigma}{\sigma_{ref}}\right)^n \exp\left[\left(\frac{Q}{R}\right)\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] \quad (2)$$

where: t_f= time to failure; A= non-dimensional material constant that reflects the effect of material properties on time to 1% PWSCC; σ = stress; n= exponent of stress; t_{ref} = time to selected fraction of PWSCC for a reference case; σ_{ref}= reference value of stress; Q= thermal activation energy; T=absolute temperature; R=universal gas constant; T_{ref} = reference value of temperature. The 2-parameter Weibull statistical distribution describes the variation of PWSCC as time function as Eq. 3.

$$F = 1 - \exp\left[-0{,}0101\left(\frac{t}{t_{1\%}}\right)^b\right] \quad (3)$$

where: F= fraction of population of components under consideration all susceptible to the same failure mode that experience PWSCC; t= time normally given in effective full power years (EFPY); b=Weibull slope, a fitted parameter determined by analysis of failure data. The value of $t_{1\%}$ together with an appropriate value for the Weibull slope b determine the complete prediction for PWSSC as time function using Eq. (2). More details of this model and solved examples can be find in [11] and [19].

The strain rate damage model is essentially a semi-empirical model theory of stress corrosion cracking, where strain rate rather than stress is considered to be the main mechanical variable strain rate as a driving force to initiate and propagate the crack. The main parameter of this model is the damage parameter D defined by Eq. (4). The strain rate is formalized as driving factor in a damage model that allows quantitative predictions on serviceable life that depends on SCC. A damage function is defined as linked mode to a component submitted to a strain rate history. When this damage function reaches a critical value, it is predicted the SCC. The critical value of this damage function depends on the concerned material and environment [18],[19].

$$\mathbf{D=\int_0^t A[\dot{e}(t)]^p.\ dt} \qquad (4)$$

where: t= time; $\dot{e}(t)$= total strain rate; A and p = parameters which depend on material-environment combination.

2.1 Simplified Strain Rate Damage Model

Concerning the Strain Rate Damage Model, there is a simplified one relating the initiation time of the crack with material activation energy and the material yield strength/ tensile strength ratio, developed to best describe the predictions based on the results of simulation of tube under uniform axial stress, σ, for time to initiation. The failure of a thin-wall tube is reached on time t_i according with Eq. (5). These parameters were found for four conditions of Alloy 600: low temperature mill annealed, high temperature mill annealed, thermally treated at about 710^0C and cold worked. This model is based on assumption of a localized break of passivation film following with repassivation, and interacting with local strain that through the strain rate increases the SCC damage [20].

$$\mathbf{t_i = \alpha_i \exp (Q_i /RT).\ln[A(\sigma_y/\sigma)]} \qquad (5)$$

where t_i =initiation time, α_i = SCC resistance parameter (dimension of time); Q_i = apparent activation energy; R=universal gas constant (1,987 cal/mol); T= absolute temperature in K; A= parameter depending on material-environment interaction; σ_y=yield strenght of material at room temperature and σ = uniform axial stress.

Boursier et al. have proposed a damage model drive by strain rate [21]: especially for Alloy 600 PWSCC strain rate (instead of tension) drives the damage and therefore creep is the growth factor. These authors used experimental data through method developed by Santarini [22] and empirical relationship were obtained for crack tip strain rate in constant load test (CL) and SSRT/CERT. For this last test, Eq. (6) was obtained.

$$\mathbf{\dot{e}_{ct} = C\ \dot{e}_{app} + D\ (da/dt)/\ x^*} \qquad (6)$$

where: $\dot{e}_{ct}$=crack tip strain rate due creep, $\dot{e}_{app}$=apparent macroscopic strain rate, da/dt=crack propagation velocity, x*= calibration lenght, C, D = constants.

According Gras, minimum apparent strain rate to occur Alloy 600 PWSCC is small than $\dot{e}_{app} \sim 2.\ 10^{-7}\ s^{-1}$, and could reach $10^{-7}\ s^{-1}$ if condition were very heavy and the material specially susceptible to PWSCC [23].

According with a diagram developed by these authors [22], it can be unified the results for initiation time and slow crack growth time, to reach a crack depth about 80-90 μm, through Eq. (7).

$$\mathbf{t_f =K.\ \dot{e}_{ct}^{-0,67}} \qquad (7)$$

3 PROPOSED MODEL

It is first proposed an empirical-comparative model similar with the model of Staehle, a three-dimensional diagram according reference [11] that shows the thermodynamic conditions to occur the modes of PWSCC in Alloy 600. The base is a two-dimensional one (Figure 5(a) [11]), the potential x pH or Pourbaix diagram for this material in primary water at high temperature (300 to 350^0 C). It superimposes the corrosion submodes, using literature experimental data. Submodes are determined by regions of potential where the different modes of surface material-environment interactions can occur, like stress corrosion, pitting, generalized corrosion or passivation. The third dimension is the "useful strength" of the material as affected by the environment at that point, the strength fraction. This third variable could be replaced by another one such as crack velocity for the vertical coordinate, instead of the strength fraction because the data are sparse and the component testing with reference to this diagram used different methods of loading states and handling the data [11]. Thus for future development it had been proposed a model framed over the same Pourbaix potential V-pH diagram superposed with following models: a) the empirical-comparative; b) the semi-empirical-probabilistic; c) the strain rate damage simplified of Garud; d) the strain rate dame of Boursier. After, it will be tested the model using literature data and data from the new slow strain rating test (SSRT) equipment installed at CDTN in Brazil [24]. These models can be applied to Brazilian Nuclear Power Plants.

4 MATERIALS AND PROCEDURES

The experimental data utilized for to propose the modelings in this research were obtained at CDTN-Centro de Desenvolvimento de Tecnologia Nuclear, a Brazilian Development of Nuclear Technology Centre, located at Belo Horizonte, through SSRT equipment installed by Nuclear Research Institute Řež (NRI) of Czech Republic [24], [25]. There were utilized not pre-cracked Alloy 600 MA specimens (Figure 1), according composition and mechanic properties described on Table 1.

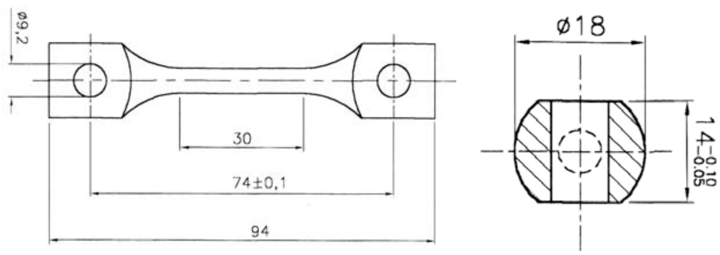


Figure 1. Alloy 600 MA specimen geometry used on CDTN SSRT [25].

The standard utilized for tests was ASTM G-129-00 [25]. It were realized three tests with strain rate $\dot{e}$= 3. 10^{-7} s^{-1}, velocity v_T= 33 μm/h, one of these tests in neutral environment (N_2).

Table 1. Chemical composition (%), main mechanical properties at 22^0C and primary water environment conditions of tests of material specimens [21]

Ni	Cr	Fe	Mn	C	Si	S	P
75,05	15,61	8,81	0,22	0,042	0,18	0,0002	0,008
Co	Cu	Al	Ti	Nb	σ_Y(MPa)	σ_R(MPa)	
0,10	0,03	0,08	0,20	0,20	302	632	
Environment primary water: pressure = 10MPa, temperature T= 303°C, 1200 ppm H_3BO_3, 2,2 ppm LiOH, 35 $cm^3 H_2$/kg H_2O, 5 ppb O_2							

The slow strain rate test (SSRT) is a dynamic test where it is imposed to the specimen a slow strain rate through external force over a monitored section, or over a notched region of this specimen, or over a fatigue pre-cracked to the evaluation of SCC material resistance. The imposed slow strain rate is normally between 10^{-4} and 10^{-7} s^{-1}. The local strain rate should be slow enough to make time to occur corrosion processes, and quick enough to cause cracks or damage in a specimen during a reasonable time [25]. In Figure 2 is showed a lateral surface of a SSRT test specimen, PWSCC cracked (b and c), compared with another one in neutral environment (a).

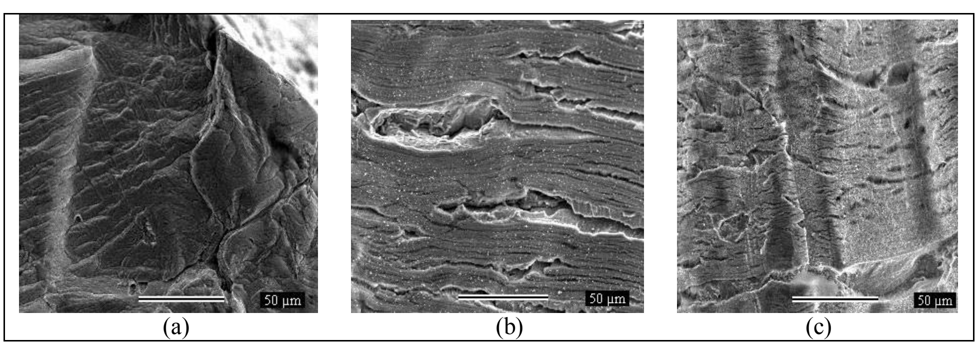


Figure 2. Microfractographies obtained through ESM, increased 500 times. Is showed the Alloy 600 MA specimen's lateral surfaces. The tests were realized with slow rate test at 303 ºC and 10 MPa. Rate test was $3,0x10^{-7}$ s^{-1}. (a) SSRT 01 (N_2 environment), (b) SSRT 02 (PW environment), (c) SSRT 03 (PW environment) [25]

In Figures 3 and 4 are showed respectively the stress-strain curve, and stress-time of three tests, one in neutral and two in primary water environment, in CDTN-SSRT. [25]

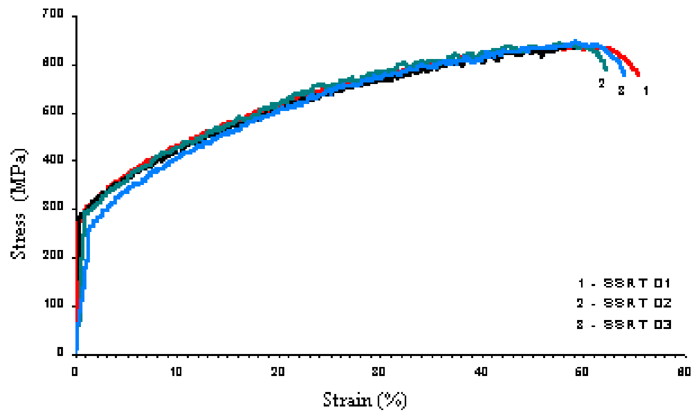


Figure 3. Alloy 600 MA specimens stress-strain curves. SSRT 01 (neutral environment of N_2), SSRT 02 and 03 (PW environment at 303 ºC and 10 MPa). Initial strain rate of $3{,}0x10^{-7}s^{-1}$ [25]

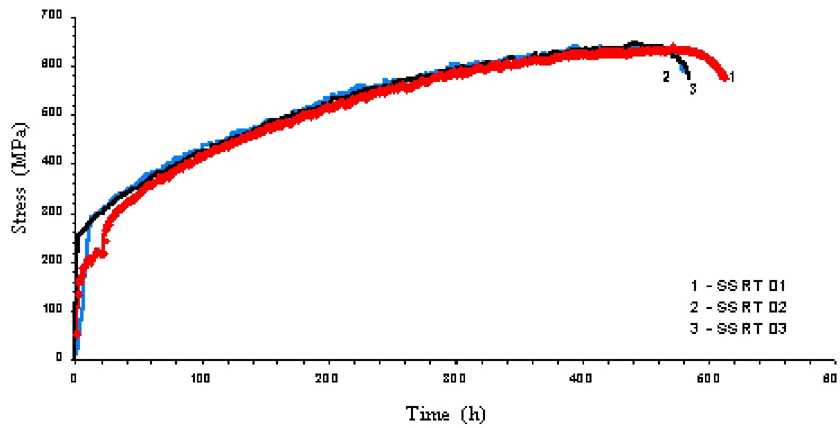


Figure 4. Alloy 600 MA specimens stress-time curves. SSRT 01 (neutral environment of N_2), SSRT 02 and 03 (PW environment at 303 ºC and 10 MPa). Initial strain rate of $3{,}0x10^{-7}s^{-1}$ [25]

5 RESULTS AND DISCUSSION

5.1. Concerning the Empiric-Comparative Model [11]

It were obtained values in Table 2 related to Alloy 600 PWSCC according with susceptibility parameters [25].

Since this information it had been fixed a representative number of the PWSCC strength fraction, the arithmetic mean value between the two tests failure time rate ~ 0,95; this value had been plotted on z-axis that represents the PWSCC strength fraction, according Figure 5 (b) [11]. Also, it had been obtained through estimating since the electrochemical experiments in CDTN [26], the pair of values (potential, pH) for SSRT tests, (-621 mV_{SHE} and pH=7,3): this value is near the bordeline between corrosion submode I_{PAS} and the submode III_{SCC} (Figure 5(a)), corroborating the expectation for Alloy 600 PWSCC initiation on test condition, agreeing literature [11].

Then it is possible to build an empirical-comparative model according with Staehle [11], but this is not a practical one, because it will be necessary several tests to build it and validate it how has done in reference [11]. However in this same reference it had been recommended that experimental data of testing following to be accumulate, to contribute on validation of diagrams like Figure 5 [11].

Table 2. Parameters of semi – quantitative evaluation to PWSCC according with tests in CDTN[25]

	Test SSRT n°. 1	Test SSRT n°. 2
Failure time rate (test environment/neutral environment)	0,94	0,97
Strain rate (test environment/neutral environment)	0,96	0,97
Reduction of area rate (test environment/neutral environment)	0,92	0,92

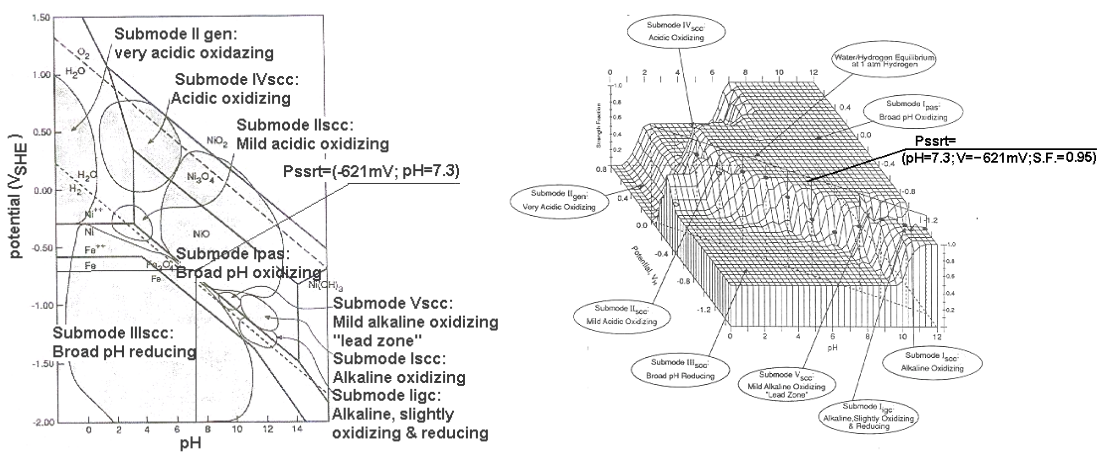
5.2. Concerning the Semi-Empirical-Probabilistic Model [19]

Based on CDTN-results (initiation time of 482.4 h, and 494.4 h, in two tests with primary water environment [25]) it had been obtained the empirical part of the model; it had been considered the mean value between these two values to propose only the deterministic part of this model, according Eq. (8):

$$\mathbf{t_i = 1,45.\ 10^{-13}.\ \sigma^{-4}.\ exp\ (32882,35/T)} \qquad \textbf{(8)}$$

with t_i = initiation time in days; σ= stress in MPa and T=absolute temperature in K; the not experimental parameters of modeling were taken off reference [19].

It should be noted that experimental data of CDTN are compatible with literature values in references [11], [20], [27], but in a very conservative way, assuming very early crack initiation: in any way, this shall be investigate through additional tests, also because it's necessary to obtain the probabilistic part of the model, that demands more tests to be apply the Weibull statistical distribution..



(a) (b)

Figure 5. (a) Tridimensional diagram base, the Pourbaix pH x potential V_{SHE}; (b) Tridimensional diagram: PWSCC strength fraction x pH x potential V_{SHE} with plotted point obtained on CDTN tests: S.F.~0,95; V=-621mV; pH=7,3. Based diagrams are from reference [11]

5.3. Concerning the Simplified Damage Model [20]

It had been obtained Eq.9 according parameters presented in reference [20] and the CDTN tests results [25].

$$\mathbf{t_i = 4,88.\ 10^{-23}.\ exp\ (32822,\ 35/T).\ ln\ [1,79\ (278,5/\sigma)]} \qquad \textbf{(9)}$$

with t_i = initiation time in days; σ= stress in MPa e T=absolute temperature in K.

The not experimental parameters of modeling were taken off reference [20]. It had been also assumed to Eq. (3), that instead of an applied uniform axial stress σ (original model was conceived for a constant load tube test), it had been considered an average stress σ_A=450 MPa (according Figure 3), since the SSRT test load is variable (with constant strain rate): this average value corresponds to the mean between elastic ultimate stress, and yield ultimate stress reached on testing.

5.4. Concerning the Damage Model of Boursier [21]

It had been obtained Eq.10 according reference [21] and the CDTN tests results [25].

$$\mathbf{t_i = 8{,}28.\ 10^{-3}.\ \dot{e}_{SSRT}^{\ -0{,}67}} \qquad \mathbf{(10)}$$

with t_i = initiation time in days; $\dot{e}_{SSRT}$=strain rate used in SSRT/CERT corresponding to apparent macroscopic strain rate, in s^{-1} and for a fixed temperature of 303^0C.

The Eq. (10) corresponds to a particular case of the Eq. (7) considering crack initiation (crack growth velocity~0), and $\dot{e}_{SSRT}$ like or smaller than $2,5 \cdot 10^{-7}\ s^{-1}$, to say when the relationship between crack type strain rate $\dot{e}_{CT}$ with $\dot{e}_{SSRT}$ is around 3, according with reference [21]. Then, the Eq. (10) is restrained to SSRT, and is a laboratory model in a limited strain rate range. It's necessary more tests to confirm validity of the Eq. (10). It's possible also to mean about a field application using extensometry tests, and then considering a similar model.

6 CONCLUSION

It's possible to use SSRT to modeling Alloy 600 PWSCC, departing from some selected models: in this paper were realized modelings, based in some tests done at CDTN-Brazil. It were used the models empirical-comparative, semi-empirical-probabilistic (only the deterministic part), simplified strain rate damage, and strain rate damage of Boursier. The use of Pourbaix diagram (potential x pH) to determine the thermodynamics conditions to occur various corrosion submodes, combined with kinetic models of crack initiation and growth, has the advantage to do better predicting of the very complex PWSCC, and also to proportionate a research methodology for this kind of cracking. Notwithstanding , in all cases, it's necessary to do more tests to confirm obtained results and to following modeling.

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