Effect of Reversible Hydrogen Trapping on Crack Propagation in the API 5CT P110 Steel – A Numerical Simulation

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ABSTRACT

This paper presents a numerical simulation of the reversible hydrogen trapping effect on crack propagation in the API 5CT P110 steel using a model based on a synthesis of fracture mechanics and continuum damage mechanics. The trapping term at the diffusion equation of this model was replaced by the equivalent term of McNab & Foster's model. Was simulated an C(T) specimen loaded in the mode I, in linear elastic regime, in plane strain state, and under the action of a static mechanical loading and hydrogen effect. The simulations showed that the material degradation ahead of crack tip increases with increasing in hydrogen concentration due the trapping with low interaction energies. Furthermore, the process of onset and crack growth in material with reversible traps is faster than the material free of traps. These results show a good correlation and consistency with macroscopic observations of the trapping effect, providing a better understanding of the hydrogen embrittlement in structural steels.

Keywords: Hydrogen Trapping, Hydrogen Embrittlement, Continuum Damage Mechanics

1. INTRODUCTION

The possibility of development of hydrogen embrittlement processes increases in the cathodic protection systems when *cathodic overprotection* occurs, and large amounts hydrogen are produced. The hydrogen can be absorbed in metals and metallic alloys, but is not loosely transported by diffusion through the Normal Interstitial Lattice Sites - NILS, because part of it is trapped in some microstructural heterogeneities denominated *traps*, giving rise to the so-called *hydrogen trapping* [1]. The traps are classified into reversible and irreversible, depending on their interaction energy. Are reversible when they have low interaction energy and hydrogen can easily abandon them. Are irreversible when a much larger amount of energy must be applied for the hydrogen is released. The reversible traps are associated with a limited time of hydrogen residence in the lattice while in the irreversible traps the hydrogen has a permanent residence time for a given temperature [2].

When the hydrogen accumulates in traps, the diffusion process is hindered because the residence time in them is greater than the residence time in NILS. Because the main effect of traps is to reduce the hydrogen transport rate through the material, was established that the trapping is a very important part of the hydrogen embrittlement [3] and that hydrogen susceptibility of the steels is closely correlated with trapping. Some studies show the beneficial effect of hydrogen trapping, others indicate the potential of traps to start cracks when are saturated with hydrogen [4]. Studies conducted in different steels coincide on qualify the mobile hydrogen (atoms in NILS and in reversible traps) as the most harmful in relation to its contribution in the process of embrittlement, because it can more easily migrate to sites of crack initiation [5]. However, it is widely accepted that the manipulation and control of microstructure is an effective method to obtain resistance to hydrogen embrittlement in steels [6].

Several mathematical models have been proposed to evaluate the trapping effect on the hydrogen transport [7], the most referenced are the McNabb & Foster's model [8] and the Oriani's model [9]. The first model is based on probabilistic considerations and places no restrictions on the mechanism of trapping, so it is of general applicability. The second model assumes that for any stage of diffusion, hydrogen can reside on local equilibrium in NILS and traps, and that the concentration in these sites is a function of concentration in NILS. Kanayama *et al*. [10] showed that the McNab & Foster's model [8] is more appropriate than the Oriani's model [9] when one should to consider the concentration in traps.

This work presents a numerical simulation of the effect of hydrogen trapping on crack propagation in the API 5CT P110 steel using a model based on a synthesis of fracture mechanics and continuum damage mechanics proposed by Bolotin & Shipkov [11]. In order to simulate the effects of hydrogen trapping in a way closer to reality, the trapping term of hydrogen transport equation of this model was replaced by the equivalent term of McNabb & Foster's model [8].

Was simulated a C(T) specimen loaded in the opening mode, in linear-elastic regimen, in plane strain state and under the effects of a static mechanical load and an environmental action, characterized by the hydrogen concentration at the tip of crack, the diffusion in the region ahead the tip and the reversible trapping in microstructure of material.

2. MODEL OF ONSET AND CRACK GROWTH IN AGGRESSIVE ENVIRONMENTS

In this model [11], the crack propagation is considered as the result of the interactions between the stability conditions of the cracked body as a mechanical system, and the process of damage accumulation at the crack tip. The model includes the kinetic equations for the accumulation of each damage type, the equation of evolution of the crack tip radius and the equation that describes the influence of the damage accumulation process on the generalized resistance forces.

Will be employed the general theory for the propagation of cracks developed by Bolotin [12] and additionally, will be considered a mass transfer model. Their coupling with the kinetic equations of damage accumulation, associated to the equilibrium conditions, stability and crack propagation, will make possible the modeling of the growth of a crack subject to a static load under the hydrogen effect.

2.1 CRACKED BODY MECHANICS

From the viewpoint of mechanics of deformable solids, crack initiation and growth are a result of the interaction of two mechanisms: damage accumulation near the crack tip and the general balance of forces and energy in the system cracked body-loading-environment. Therefore, the crack behavior depends on the relationship between the generalized driving force, G, and the generalized resistance force, Γ; in addition, the generalized resistance force depends on the damage accumulated at the crack tip zone. G can be associated with the energy release rate in linear fracture mechanics, and Γ with the critical magnitudes of this rate. In terms of these forces, the crack does not grow while the condition $G < \Gamma$ is respected. The crack growth will begin and propagates stably until the next arrest or until failure when $G = \Gamma$. In the case of $G > \Gamma$, the system is unstable, and the crack propagates until fracture of the component [12].

These conditions are valid to model the crack growth and the final fracture for a mechanical loading including the effect of an environmental phenomenon, such as the hydrogen embrittlement.

2.2 HYDROGEN ASSISTED DAMAGE

To model the process of damage accumulation, it is introduced a special measure for each damage type, mechanical or environmental, and the correspondent kinetic equation that governs the evolution in the time. For example, when mechanical actions are considered, the damage caused by statics loadings is differentiated from the damage due to other mechanical loadings, as the cyclic loadings. Also, when environmental actions are considered, the damage caused by hydrogen diffusion is differentiated of the damage produced by a corrosion process. Therefore, the entire damage field is represented by a set of scalar damage fields, where the hydrogen damage is an additive component of the total damage magnitude.

The evolution of the static load damage at the crack tip can be expressed by:

$$\frac{d\psi_s}{dt} = \frac{1}{t_c}\left(\frac{\sigma - \sigma_{th}}{\sigma_s}\right)^{m_s} \tag{1}$$

where σ is the average value of the tensile stress in a considered material point. σ_s characterize the resistance to the damage produced by the static loading and σ_{th} is a resistance threshold parameter.

The evolution of the hydrogen damage ahead the crack tip can be expressed by [13]:

$$\frac{d\psi_{rh}}{dt} = \frac{1}{t_c}\left(\frac{(c_r - c_{th})}{c_d}\right)^{m_{rh}} \tag{2}$$

where ψ_h is the magnitude of hydrogen damage at the crack tip; c_r is the evolution of the concentration in the time at the distance r ahead of the tip, which varies in the processing zone of hydrogen damage evolution, λ_h. c_{th} is a threshold concentration and c_d characterizes the resistance to hydrogen damage. In both equations the m_{rh} and m_s exponents are similar to the exponents of the equations of fatigue and crack growth rate curves, and t_c is a time constant.

The evolution of the crack tip radius can be expressed by:

$$\frac{d\rho}{dt} = \left(\frac{\rho_s - p}{\lambda_a}\right)\frac{da}{dt} + (\rho_b - \rho)\frac{d(\psi_s + \psi_{rh})}{dt} \tag{3}$$

The first term on the right side describes the crack tip sharpening evolution until the magnitude ρ_s, that is the sharp tip radius. The second describes its blunting due to damage accumulation until the magnitude ρ_b, that is the blunt tip radius. When the critical value of this radius is reached, is initiated the growth and crack propagation. λ_a is a parameter with the length dimension.

The generalized resistance forces to propagation are related to measures of damage ahead the crack tip using the following equation:

$$\Gamma = \Gamma_0 [1 - \chi(\psi_s + \psi_{rh})^\alpha] \tag{4}$$

where Γ_0 is the fracture specific work for an undamaged body, χ characterizes the residual fracture toughness and α is a material parameter.

2.3 HYDROGEN TRANSPORT

The hydrogen atoms that enter metals and metallic alloys come from several sources, such as electrochemical reactions or the environmental moisture. Hydrogen is transported from the crack mouth to the crack tip, adsorbed on the metal surface, absorbed by the metal interface and transported through the lattice. This latter process includes interstitial diffusion and trapping mechanism.

A phenomenological approach to the hydrogen transport within the hole crack, is given by:

$$\frac{dc_t}{dt} = \frac{c_h - c_t}{\tau_D} + \frac{c_b - c_t}{\lambda_D} \frac{da}{dt} \tag{5}$$

where the first term on the right side represents the diffusion mechanism, and the second term, the crack propagation effect. λ_D is the characteristic length of the diffusion zone and τ_D the characteristic time of diffusion. c_t is the hydrogen concentration at the crack tip, c_h characterizes the stationary concentration reach for a stationary crack in the time $t >> \tau_D$, and depends on the entrance concentration and of the crack length. The parameter c_b is a characteristic concentration for the fast growth of the crack.

Hydrogen diffuses through the material due to the existence of a concentration gradient. However, when a stress field is applied, the chemical potential of hydrogen is changed and atoms diffuse toward regions where their chemical potential is lower [3]. Thus, when a mechanical load is applied, the hydrogen can diffuse from regions of low stress towards regions of high stress, and the flow comes to depend not only of the concentration gradient, but of the stress gradient and hydrogen trapping process. The hydrogen transport without considering the hydrogen trapping, was simulated in [13], where was verified the importance of mechanical effect on damage evolution in hydrogen assisted processes.

The hydrogen transport in the solid phase, ahead of the crack tip, considering the hydrogen trapping in the microstructural defects, is given by:

$$\frac{\partial c_t}{\partial t} = \frac{\partial}{\partial x}\left(D_s \frac{\partial c_t}{\partial x} \right) - \frac{\partial}{\partial x}\left(\frac{D_s \overline{V}_H}{RT} c_t \frac{\partial \sigma_h}{\partial x} \right) - \frac{\partial c_T}{\partial t} \tag{6}$$

where D_s is the diffusivity in the solid and σ_h the hydrostatic stress. $\overline{V}_H$ is the partial molar volume of hydrogen in the metal, R is the universal gas constant, T the room temperature and c_T the hydrogen concentration in the traps.

The last term of the right side of Eq. (6) describes the trapping effect in terms of threshold concentrations at the crack tip and in the traps, but not characterizes the trap type. In this work this term has been replaced by the corresponding term of McNabb & Foster's model [8], with the purpose to obtain a better characterization of the real phenomenon.

After substitution, Eq. (6) becomes:

$$\frac{\partial c_t}{\partial t} = \frac{\partial}{\partial x}\left(D_s \frac{\partial c_t}{\partial x} \right) - \frac{\partial}{\partial x}\left(\frac{D_s \overline{V}_H}{RT} c_t \frac{\partial \sigma_h}{\partial x} \right) - N_T \frac{\partial \theta_T}{\partial t} \tag{7}$$

where N_T is the trap density and θ_T the coverage of these sites. The coverage rate is given by:

$$\frac{\partial \theta_T}{\partial t} = kc_{\mathrm{L}}(1-\theta_T) - p\theta_T \tag{8}$$

where k and p are the hydrogen capture and hydrogen release rates, respectively.

3. NUMERICAL FORMULATION

The purpose of the formulation of this problem is to evaluate the hydrogen trapping effect on crack propagation in a hydrogenated media using the mathematical model described previously. The crack is loaded in the tensile opening mode, in plane strain state. The geometric configuration of the specimen chosen for the simulation and the brittle state of material ensure that their behavior is in the elastic regime. The evolutions of damage, crack tip radius and crack length variables, and the generalized resistance forces, form a 1st order differential equations system that is solved through the 4th order Runge-Kutta method. The problem of solid-solid diffusion ahead the crack tip and trapping is solved by the finite difference method. It is assumed a high coverage of traps with the same interaction energy, and is considered only the trapping reversible mechanism.

The hydrogen concentration at the crack mouth, c_e, is considered constant, and the concentration at the crack tip provides the concentration gradient required to initiate the hydrogen diffusion ahead of the crack tip; moreover, the hydrostatic stress gradient leads some of the hydrogen to the crack tip. The boundary conditions for the diffusion problems in the crack hole, a, and in the process zone of hydrogen damage in front of the crack tip, λ_h, shown in Fig. 1, are:

$$\begin{array}{llcll} c = c_e, & \text{for } x = 0 & ; & D(\partial c/\partial x) = 0, & \text{for } x = a; \\ c = c_t, & \text{for } x = a + dx & ; & D(\partial c/\partial x) = D_s(\partial c_t/\partial x), & \text{for } x = \lambda_h. \end{array}$$

The approximate thickness of hydrogenation film, equivalent to the length of the hydrogen damage process zone ahead the crack tip, can be calculated from:

$$\lambda_h = a_0 \frac{D_s}{D_0} \tag{9}$$

where a_0 is the initial crack size and D_0 is the constant of solute/solvent system.

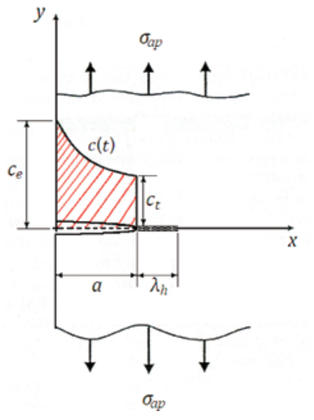

Figure 1: Hydrogen distribution in the hole crack [1]

In the context of linear elastic fracture mechanics, it is possible to relate the generalized propagation forces with the elastic strain energy, and quantify them through the Irwin's equation:

$$G = \frac{K_I^2}{E}(1 - v^2) \tag{10}$$

where E is Young's modulus, v the Poisson's modulus and K_I the stress intensity factor. The formula for the calculation of this parameter can be found in the ASTM E1820-09 standard [14].

To evaluate the mechanical damage, the stress fields in the body during the considered process should be known. Some simple approaches based on an analogy among the stress concentration factor, K_t, and the stress intensity factor, K_I, can be used. An approximate formula for the distribution of the normal stress in crack tip front is:

$$\sigma = K_t \sigma_a \left[1 - \frac{4(x-a)}{\rho}\right]^{-\frac{1}{2}} \tag{11}$$

where:

$$K_t = 1 + 2Y\left(\frac{a}{\rho}\right)^{\frac{1}{2}} \tag{12}$$

4. RESULTS AND DISCUSSION

To evaluate the applicability of the model at the conditions of study previously discussed, was simulated a C(T) specimen with dimensions: l = 48 mm, w = 50 mm, t = 10 mm, and a_0 = 20.3 mm (Fig. 2). The material chosen was the API 5CT P110 steel, whose physical and mechanical properties are shown in Table 1.

The specific fracture work for the undamaged body was assumed as Γ_0 = 31.2 kJ/m^2, which, within the framework of linear fracture mechanics, corresponds to a fracture toughness of 86 MPa√m. The subsurface hydrogen concentration for the API 5CT P110 steel, that characterizes the overprotection condition in seawater (–1100 mV$_{Ag/AgCl}$) is 6.27 ppm [13,15].

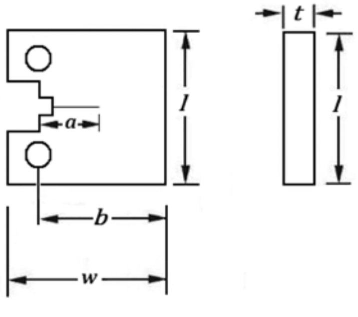

Figure 2: C(T) specimen used in simulation

Table 1: Physical and mechanical properties of API 5CT P110 steel [15]

σ_{UTM} [*Pa*]	σ_Y [*Pa*]	*E* [*Pa*]	*v* [ad.]
$9.77 \cdot 10^9$	$8.73 \cdot 10^8$	$2.16 \cdot 10^{11}$	0.30

Table 2: Material parameters [11, 15]

α	c_h	c_b	c_e	c_d	t_c	τ_D	ρ_s	ρ_b	λ_D	λ_a	ρ
[ad.]	[ppm]	[ppm]	[ppm]	[ppm]	[s]	[s]	[μm]	[μm]	[μm]	[μm]	[μm]
1.0	2.0	3.0	4.5	1.2	$1{\cdot}10^3$	$1{\cdot}10^2$	10.0	$1{\cdot}10^2$	10.0	$1{\cdot}10^2$	50.0

λ_h	λ_s	χ	m_s	m_{rh}	σ_s	σ_{th}	$\bar{V}_H$	D_s	k	θ_T	N_T
[μm]	[μm]	[ad.]	[ad.]	[ad.]	[Pa]	[Pa]	[m^3/mol]	[m^3/s]	[m^3/s]	[ad.]	[sites/cm^3]
63.5	$1{\cdot}10^2$	1	1.0	1.0	$5{\cdot}10^9$	$40{\cdot}10^6$	$1{\cdot}10^{-6}$	$4.78{\cdot}10^{-11}$	$4.5{\cdot}10^{-25}$	0.002	$1.49{\cdot}10^{15}$

The material parameters adopted for the simulation and solution of this problem are shown in Table 2.

The results from the application of the model, for σ_{ap} = 50 MPa, are shown in the graphs below: Figure 3, Fig. 4 and Fig. 5 show the hydrogen distribution in the damage process zone ahead the crack tip for different diffusion times. Fig. 3 was obtained considering only the concentration gradient in Eq. (7). The hydrogen distribution without considering the trapping term in Eq. (7) is show in Fig. 4. The application of the full Eq. (7) is show in Fig. 5.

In Figure 3 can be observed that the diffusion process is conditioned uniquely by the concentration gradient. The hydrogen flux goes towards the smaller concentration and the steady state is reached. When the stress is applied, some of the hydrogen migrates for the close area to the crack tip, as seen in Fig. 4, which accelerates the reduction of fracture resistance of material.

When considering the reversible trapping mechanism, hydrogen is trapped in reversible traps, hindering the diffusion process and making available more hydrogen atoms to be displaced until regions of high stress concentration. As observed in Fig. 5, traps retain the atomic hydrogen in the damage process zone, and the hydrogen trapped migrates to the crack

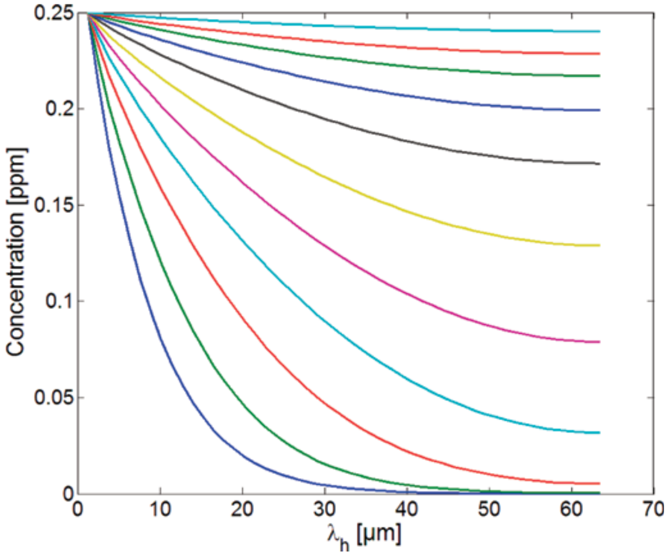

Figure 3: Hydrogen transport in the damage process zone ahead the crack tip considering only the concentration gradient in Eq. (7)

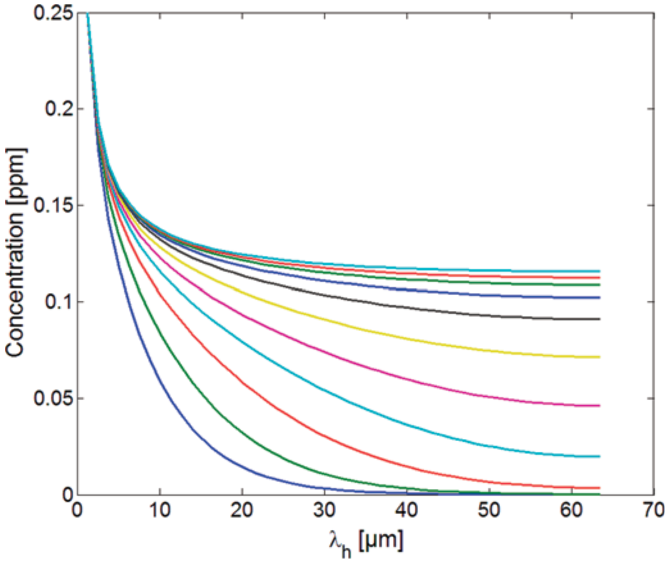

Figure 4: Hydrogen transport in the damage process zone ahead the crack tip without considering the trapping term in Eq. (7)

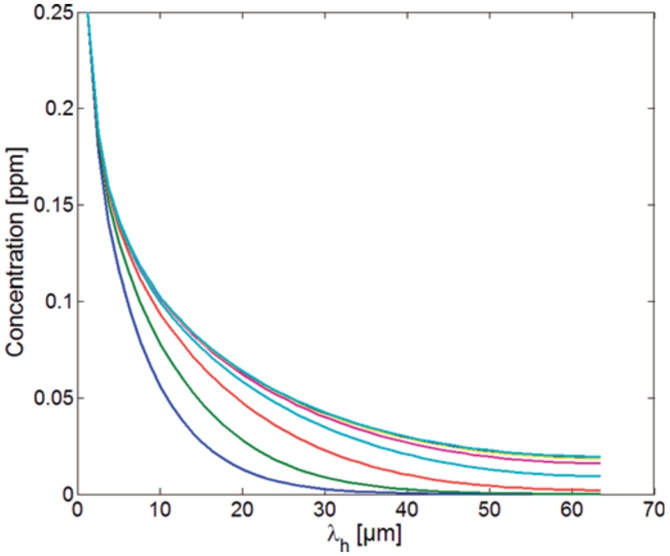

Figure 5: Hydrogen transport in the damage process zone ahead the crack tip considering all terms in Eq. (7)

tip after application of mechanical load. As will be seen below, the degradation process of material in this condition is more intense that in the cases described above.

The intensification process of material degradation due to trapping effect can be best observed in Fig. 6, which shows the evolution at the crack tip for three different trapping energies (), 20 kJ mol^{-1}, 30 kJ mol^{-1} and 59 kJ mol^{-1} [6], correspondent to three reversible traps sites: grain contour, dislocations core (screw) and dislocation core (mixed). In all curves we observe that the closest region to the crack tip is that more undergoes the degradation process, as a direct effect of the hydrogen concentration in this zone, associated to the mechanical loading applied and the hydrogen trapping: the magnitude of damage is higher in so far as the trapping energy is less. As result, the damage is greater at the crack tip, but also is important in the processing region ahead the crack tip.

This degradation profile is associated with the hydrogen released from traps along the processing zone and is most evident for the trapping with lower interaction energy, as the grain boundaries. The effect of different types of traps on the hydrogen damage evolution can be easily perceived: when the trapping interaction energy is greater, as in the case of dislocations, there are more difficulty so that the hydrogen atoms are released when a stress is applied, and cannot actively participate in the degradation processes occurring in the region ahead of the crack tip. The hydrogen damage is more severe in presence of reversible traps with lower interaction energy, indicating that the material with these heterogeneities is more susceptible to hydrogen effect, what is according with results of experimental observations reported in the scientific literature [7].

Figure 7 displays three curves that correspond to the evolution of crack growth for three situations: first, when the structure is under the mechanical loading and hydrogen effect, without considering trapping; second, when considered traps with medium interaction energy as dislocations core (mixed); third, when considered traps with low interaction energy, as grain contour. The crack growth evolution is conditioned by the static loading and the hydrogen effect, which in turn, is conditioned by trapping. When evaluated the effect of traps with different interaction energy in the two last conditions, can be observed that as trapping energy decreases, the time of onset and crack propagation decreases also, as in real situations. In the three curves, the highest point represents the onset of unstable propagation and final fracture of the component.

This type of behavior was already expected, since the trapping mechanisms have a very important influence on hydrogen transport in the lattice, which affects the concentration levels in the places where degradation processes occur, which in turn, affects the mechanical behavior of the material.

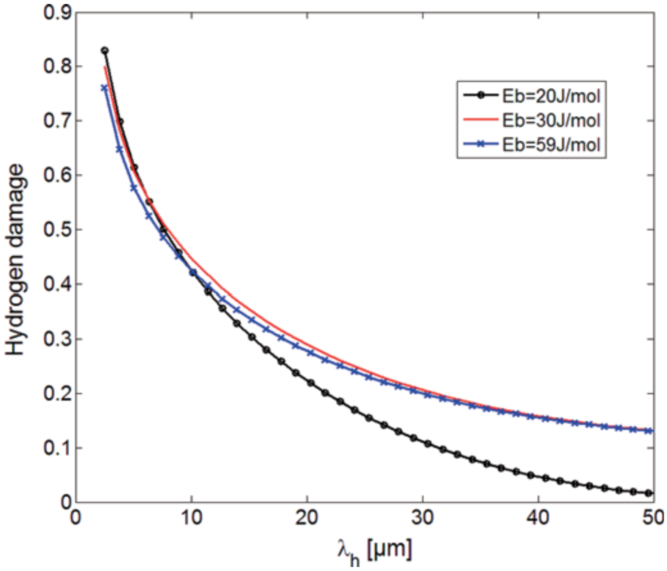

Figure 6: Hydrogen damage evolution at the crack tip

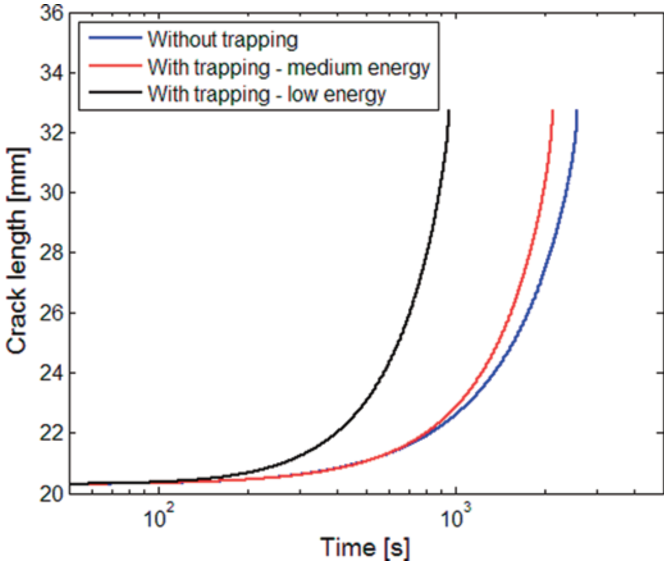

Figure 7: Crack growth evolution

5. CONCLUSIONS

The damage mechanics model adopted in this study showed good flexibility to be coupled with others models, such the McNabb & Foster's model. The coupling of the trapping term in the transport equation of damage model allowed a simulation closer to reality, and a better understanding of the studied phenomenon.

The simulation showed that the time of onset and crack growth decreases with the presence of reversible traps in the microstructure, and increases when the trapping effect is not considered. As was assumed that the rate of hydrogen release of traps is high, characterizing the traps as reversible, these results were expected, since experimental work papers published by different authors coincide in indicating that the reversible traps are dangerous.

The results showed good consistency with observations of the hydrogen embrittlement phenomenon, well described in scientific literature, allowing infer the validation of model for the conditions and hypotheses outlined in the problem.

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