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Hydrogen Embrittlement in a Coupled Mass Diffusion with Stress near a Blunting Crack Tip for AISI 4135 Pressure Vessel

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***Abstract*—**In pressure vessels contain hydrogen, the role of hydrogen will be important because of hydrogen cracking problem. It is difficult to predict what is happened in metallurgical field spite of a lot of studies have been searched. The main role in controlling the mass diffusion as driving force is related to stress. In this study, finite element analysis is implemented to estimate material's behavior associated with hydrogen embrittlement. For this purpose, one model of a pressure vessel is introduced that it has definite boundary and initial conditions. In fact, finite element is employed to solve the sequentially coupled mass diffusion with stress near a crack front in a pressure vessel. Modeling simulation intergrarnular fracture of AISI 4135 steel due to hydrogen is investigated. So, distribution of hydrogen and stress are obtained and they indicate that their maximum amounts occur near the crack front. This phenomenon is happened exactly the region between elastic and plastic field. Therefore, hydrogen is highly mobile and can diffuse through crystal lattice so that this zone is potential to trap high volume of hydrogen. Consequently, crack growth and fast fracture will be happened.

***Keywords*—**Stress Intensity Factor, Mass Diffusion, FEM, Pressure Vessel

I. INTRODUCTION

COUPLED mass diffusion with stress phenomena occurred by hydrogen diffusion is an important event which causes some problems for materials like, embrittlement, degradation, crack assisting and finally fast fracture and as a result, material will be failed. Krupp et al. [1] proposed a model for describing of hydrogen embrittlement. It said that while hydrogen contacts with surface of a metal, it needs energy to diffuse and cleavages of micro structure of the metal. Indeed, molecule of hydrogen is big enough not to be able enter the metal and puts itself between the molecules and atoms of the metal. In this process, by chemical reactions the molecule of hydrogen is split into two atoms because of that an amount of energy released to help it to diffuse in the metal's surface Fig.1. After, it is occurred, lines of defects are created and caused to accumulate large amount of atoms of hydrogen in these lines.

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So, by piling up them, molecules of hydrogen are constructed and consequently stress is produced. Takeichi et al. [2] suggested hydrogen diffusion occurring in dislocations because of vacancies in a lattice of metal that hydrogen atom can enter them. So, several types of sites were found to trap hydrogen such as grain boundaries, vacancies, voids, dislocations and so on. Symons [3] implemented properties of metal such as ductility changed into brittle happened called degradation. Degradation of mechanical properties is the other effect of hydrogen on metals. Muller et al. [4] specified critical hydrogen concentrations for degradation of the mechanical properties both in the irradiated and unirradiated conditions. Simultaneously, diffusing of hydrogen and degradation of properties, crack stars growing until complete failure of whole material. So, hydrogen can facilitate crack growing that occurs in either of two ways, one is by a strain-controlled mechanism and the other one is by stress-controlled decohesion. In fact, if hydrogen attacks at low temperature and decreasing of fracture toughness are happened because of degradation, absolutely fast fracture will be gone to happen [5, 6].

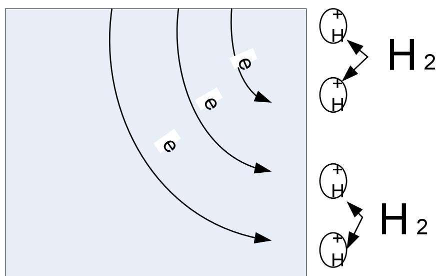


Fig.1 Hydrogen diffusion in surface of a metal

As a result, science of materials and profound mechanical studying of a micro structure in experimental method illustrate the procedure and mechanism of diffusion, degradation, and embrittlement and fracture to some extent but not completely [7]. For this purpose, different models have been proposed by different researchers to describe this mechanism more complete and accurate. There were three basic models that introduced by Zapff-Tetelman, Petch and Troiono. In addition, Zapff-Tetelman's model considered only hydrogen diffusion.

But, this model couldn't see stress parameter. Petch' model just proposed effects of fracture energy on hydrogen diffusion phenomena. But there was some defects like incomplete model involved diffusion. Troiono's model predicted interatomic band and focused on micro scales reactions between hydrogen atoms which diffused. However, it was unable to foresee and make judgment for macro scales and a coupled mass diffusion with stress in a metal [8]. Another model defined the role of strain rate at a crack tip consisting a cylinder subjected to tension is parallel to cylinder axes [9]. Toribio [10] proposed a model for elastic field on crack tip. Kim et al. (2005) considered this problem as independent system between H and C without metal components. Really, he presented a thermodynamic analysis of metal-hydrogen model to fine a relation between concentration and stress. But the biggest problem was related not to be able to describe fracture mechanics. A model has been developed describing both the microscopic and macroscopic features of embrittlement due to dissolving or internal hydrogen [11]. These are models which described above converged to find a more reliable solution to predict material's behavior during coupled mass diffusion with stress and answer certainly and surely to many questions about mass diffusion. According to the problems have been occurred due to mass diffusion phenomena in industry especially for pressure vessels; encourage us to research in this field. One of the main concerning during an operation of a pressure vessel is susceptibility of it to an embrittlement. To have a required safety margin against a fracture, should be tried with ASME Cod Section 3 and 4 [12]. There are two important points to evaluate the condition of a pressure vessel for operating. First is fracture toughness and second is the ratio of stress intensity factor to fracture toughness [13]. Diffusion occurs when there are gradient of concentration and stress as driving forces [14, 15]. The effects of hydrogen on various metals and use of metals which are well to tolerate them are very important point [16]. So, it is should be constructed a physical model in order to solve a diffusion equation to predict the treatment of mass diffusion and stress.

In this study, for having more accurate analysis and prefect evaluation of the coupled mass diffusion with stress equation, it is essential to have a model can describe and illustrate what happen during mass diffusion and how efficiently it confirms two main points such as accumulating of hydrogen concentration always is happened near the crack tip and maximum hydrostatic stress is occurred there. If one model is associated with these two main characteristics, it is possible to accept it as a reliable model for analysis of a coupled mass diffusion with stress equation. Therefore, a model for a pressure vessel exposed to mass diffusion is made and coupled mass diffusion with stress is investigated how hydrogen embrittlement can influence on the fracture toughness of its material by FEM. In addition, mass diffusion is described by Fick's law. There are two driving forces, the first one is gradient of concentration and the second is gradient of stress. So, these two gradients give us the flux of coupled mass diffusion with stress. This formulation is the extended fick's law introduced by Sofronis [17]. Furthermore, the evaluation is done in LEFM and also critical crack length and changing the hydrogen distribution for a path involved crack in along the thickness in comparison with a path involved no crack in are evaluated by FEM. Also, distribution of hydrogen is obtained along the thickness for two reasons. First, hydrogen embrittlement is controlled by the diffusion of hydrogen into the crack tip region and is influenced by hydrostatic stress. So, we need to see the treatment of metal during time of diffusion and guess what happens in future. Then it's essential to consider diffusion and fracture equations and solve them simultaneously. For this purpose, it is tried to obtain distribution of hydrogen concentration. Second, it obtained to compare with experimental distribution of concentration. It should be noted that the computational modeling analysis presented here is qualitative in nature.

II. The Concept of Finite Element for Mass Diffusion Analysis

The hydrogen diffusion model is based on the concept of an elastic interaction between the hydrostatic pressure and interstitial hydrogen atom. So, localization of hydrogen atoms strongly can be explained by hydrostatic stress as driving force. For this purpose, the relation between hydrostatic stress and concentration has been introduced by Boltzman [18] as follow:

$$C = C_0 \exp\left(-\frac{\sigma V_H}{RT}\right) \quad (1)$$

where C_0 is the initial concentration and V_H is the partial molar volume of hydrogen and σ is hydrostatic stress. The governing equations for hydrogen diffusion used in ABAQUS are an extension of Fick's equations.

$$J = -D\left(\nabla C + SK_P \nabla \sigma\right) \quad (2)$$

where J is the flux of concentration of the hydrogen gas, D is the diffusivity, S is the solubility of the hydrogen gas, KP is the present stress factor.

Stress assisted-diffusion is specified by defining the pressure stress factor, K_P which presented by Lui et. al [19] as

$$K_P = \left(\frac{\phi V_H}{RT}\right) \quad (3)$$

where R is the universal gas constant equal to 8.31432 J/mol(K), T is temperature in Kelvin and $\emptyset$ is the normalized hydrogen concentration defined as the ratio of mass concentration of hydrogen in material, C, to its solubility, S, i.e. $\emptyset = c/s$.

III. Coupled Mass Diffusion with Stress Equation

Hydrogen embrittlement is controlled by the diffusion of hydrogen into the crack front region and is influenced by hydrostatic stress state. So, it is needed to consider the treatment of metal during time period of diffusion and guess what happens in future. Then it's essential to consider diffusion and fracture equations and solve them simultaneously to get one equation through that a designer can guess exactly when the metal damaged and failed. For this purpose, it is tried to obtain distribution of hydrogen concentration. Besides, mass diffusion is described by Fick's law. It is used for cylindrical coordination with assumption that the diffusion occurred. There are two driving forces the first one is gradient of concentration and the second is gradient of stress. So, these two gradients give us the flux of the coupled mass diffusion with stress. This formulation is the extended fick's law introduced by Sofronis et al. [17]. Fig. 2 shows the model which is a pressure vessel with an elliptical crack.

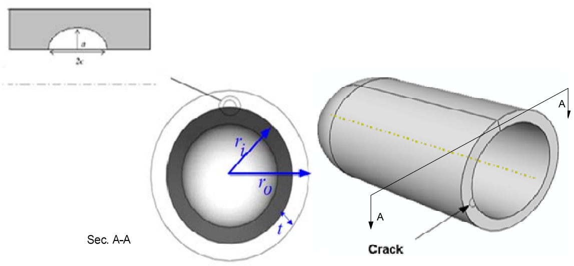


Fig.2 Schematic of a pressure vessel with an elliptical crack

$$\frac{\partial C}{\partial t} = D\nabla^2 C - M\left(\nabla C.\nabla\sigma\right) \tag{4}$$

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{1}{r^2}\frac{\partial^2 C}{\partial \theta^2} + \frac{\partial^2 C}{\partial z^2}\right) - M\left(\left(\frac{\partial C}{\partial r} + \frac{1}{r}\frac{\partial C}{\partial \theta} + \frac{\partial C}{\partial z}\right)\cdot\left(\frac{\partial \sigma}{\partial r} + \frac{1}{r}\frac{\partial \sigma}{\partial \theta} + \frac{\partial \sigma}{\partial z}\right)\right), \tag{5}$$

$M = DV_H/RT$ is mobility coefficient which is constant.

The boundary and initial conditions of the cylinder are:

$$C(r,0) = C_0 \quad \text{(5a)}$$

$$C(r_i,t) = C_B \quad \text{(5b)}$$

$$C(r_o,t) = 0 \quad \text{(5c)}$$

where C_0 and C_B is constant.

The mechanical boundary condition is that both the inner surface is subjected to inner hydrogen pressure, i.e., the pressure P. Therefore, for solution the 'Eq. (5)' needed to have stress distrubution near the crack front.

For elastic region [15]:

$$\sigma_e = \frac{2}{3}(1+\upsilon)\frac{K_I}{\sqrt{2\pi r}}\cos\frac{\theta}{2} + \frac{1}{3}\sigma_0 \tag{6}$$

According to 'Eq. (6)' for gradient of hydrostatic stress in elastic region which this relation is defined by

$$\nabla\sigma = \frac{\partial\sigma}{\partial r}\vec{e}_r + \frac{1}{r}\frac{\partial\sigma}{\partial\theta}\vec{e}_\theta = \left(\frac{-K_I}{\sqrt{2\pi}}\frac{1}{3}(1+\upsilon)Cos\frac{\theta}{2}\right).r^{-1.5}\vec{e}_r - \frac{1}{r}\frac{K_I}{\sqrt{2\pi r}}\frac{1}{3}(1+\upsilon)Sin\frac{\theta}{2}\vec{e}_\theta \tag{7}$$

By putting 'Eq. (7)' into 'Eq. (5)' is written as:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{1}{r^2}\frac{\partial^2 C}{\partial \theta^2} + \frac{\partial^2 C}{\partial z^2}\right) - M\left(\left(\frac{\partial C}{\partial r} + \frac{1}{r}\frac{\partial C}{\partial \theta} + \frac{\partial C}{\partial z}\right)\cdot\left(\frac{-K_I}{\sqrt{2\pi}}\frac{1}{3}(1+\upsilon)Cos\frac{\theta}{2}\right).r^{-1.5}\vec{e}_r - \frac{1}{r}\frac{K_I}{\sqrt{2\pi r}}\frac{1}{3}(1+\upsilon)Sin\frac{\theta}{2}\vec{e}_\theta\right) \tag{8}$$

For solving this equation, finite element analysis is employed.

IV. FEM Method for Simulation

Three equations are solved by FEM such as fracture mechanics, material's properties and mass diffusion equations in a sequentially method. It means that first stress is analysis and then mass diffusion is evaluated. It is possible to be used this method When the effect of hydrogen on the stress-strain response of the steel is negligible. So, Fig. 3 shows this [19].

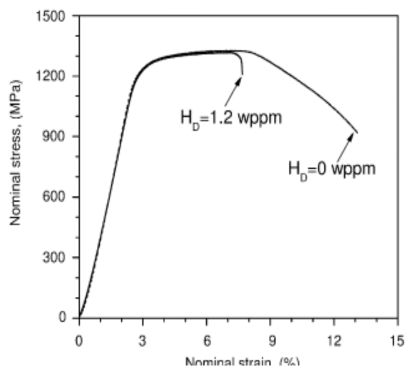


Fig.3 Effect of hydrogen embrittlement on stress-strain curve during diffusion [19]

ABAQUS v.7.1 soft ware is employed to predict it because of its capacity in analysis of coupled mass diffusion with stress.

V. Modeling of a Pressure Vessel

A. Geometrical simulation

A pressure vessel is simulated by finite element in Fig. 4. In this figure, the geometrical measurements are defined. This pressure vessel involves an elliptical crack. Only 1/8 of the pressure vessel is simulated. Sizes of geometric: L1=2300 mm, L2=250 mm. Ri=250 mm, t=20 mm, ri=250 mm.

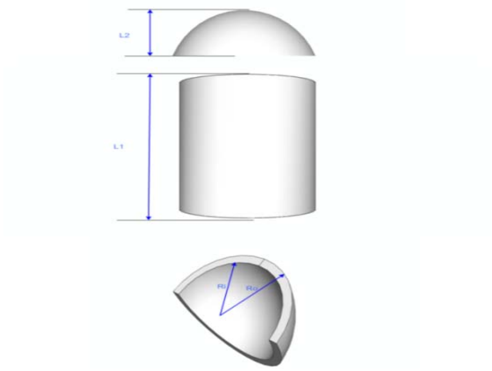


Fig.4 Schematic view of different parts of a pressure vessel

The length of the pressure vessel is 3000 mm and its inner radius and thickness are respectively 250 and 20 mm. Also, Fig. 5 illustrates singular elements which are used around the crack front. The basic concept of these elements is that they introduce the solution dominated by the singularity at the crack front. Moreover, Fig.5 shows different parts of the crack such as crack face, crack front and crack block.

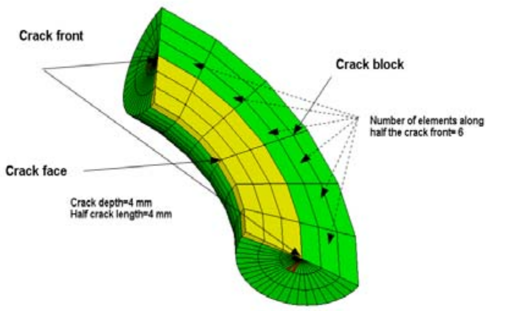


Fig.5 Schematic of a crack with three parts and singularity of the crack front

For more accuracy in crack simulation, many different partitions are employed, Fig. 6.

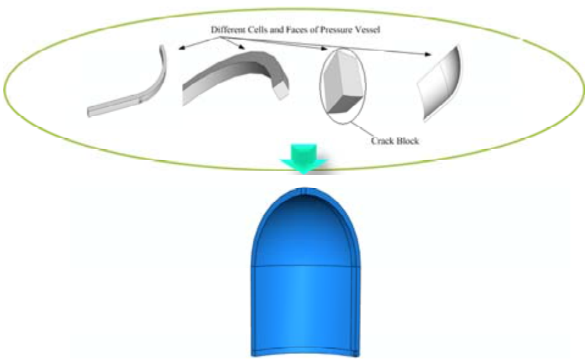


Fig.6 Different partitions for a pressure vessel

B. Definition of material property

The Finite element analyses are model a pressure vessel to solve the boundary value problems of blunting crack front under modeI (tensile) opening and small scale yielding conditions. Small scale yielding refers to the situation where the plasticity is so localized at the crack front.

This pressure vessel contains hydrogen gas with $0.9\sigma_B$. The material used for this main is AISI 4135 steel. The high strength steel contacts with hydrogen at ambient temperature. The properties of AISI 4135 are in Tables 1and 2 [19].

TABLE I
CHEMICAL COMPOSITION OF THE STEEL INVESTIGATED (MASS%) [19]

C	Si	Mn	P	S	Cr	Mo	FeCu
0.35	**0.19**	**0.79**	**0.019**	**0.014**	**16**	**0.16**	**0.17**

TABLE II
MECHANICAL PROPERTIES OF THE AISI 4135 STEEL AFTER TEMPERING AT 633 K [19]

Speciment	Tempering temperatre (K)	Tensile strength σ_B(Mpa)	Yield strength σ_y (Mpa)	Young's Modulus E (Mpa)	Poisson's ratio ϑ
B15	**633**	**1450**	**1320**	**195**	**0.3**

In addition, other important parameters for hydrogen diffusion are in Table III. Initial concentration is 0.17 ppm.

TABLE III
PARAMETERS OF HYDROGEN DIFFUSION ANALYSIS [19]

Diffuse coefficient ($m.s^{-1}$)	Temperature (K)	Material density ($kg.m^{-3}$)	Partial molar volume of hydrogen $\overline{V_H}(m^3.mol^{-1})$	Solubility (*wppm*)
2.5×10^{-11}	**298**	**7700**	**2.1×10^{-6}**	**0.4**

VI. USING TIE CONSTRAIN TECHNIQUE

In order to decrease run time, allowing variations in element shape and element interpolation would require tie constraints between varying regions. This technique can help decreasing the run time by refine meshing. The concept of this technique consists of making size of mesh changed everywhere is necessary.

So, for having accurate results in studied model, the crack block cell should have the smallest mesh of all cells of the pressure vessel because this makes results more precise. For this purpose, the model is separated into two parts. One part is crack block and the other part is the rest of the pressure vessel. So, it tried to make two parts constrain together to act as one part. Precisely, it is useful to use tie constraint technique.

VII. STRESS AND MASS DIFFUSION ANALYSIS

Finite element can be able to evaluate stress analysis in CAE (Complete Abaqus Environment). In this analysis, one modeled pressure vessel with an elliptical crack subjected to inner pressure is analyzed. As a result, contours of stress are created near the crack front. Also, mass diffusion has two responses like steady state and transition. At the beginning, diffused concentration increases as a function of time in the crack until that it reaches to saturated condition near the crack front. After that, there is no change happened for amount of concentration near the crack front. So, finite element is employed to evaluate this phenomenon to predict the behavior of the material which exposed to mass diffusion.

According to the figure, the master surface, which is the surface of the larger part and slave surface, is smaller part. Fig. 7 shows the schematic of tie constrain technique to make meshes of the crack block small to obtain accurate results.

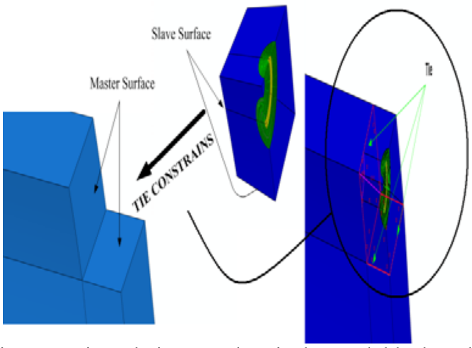


Fig. 7 Tie constrain technique used to tie the crack block to the rest of the pressure vessel

VIII. Boundary conditions for stress analysis

Axisymmetric model allows 1/8 of the pressure vessel to be modeled. This pressure vessel is subjected to inside pressure and its faces are restricted according to Fig. 8.

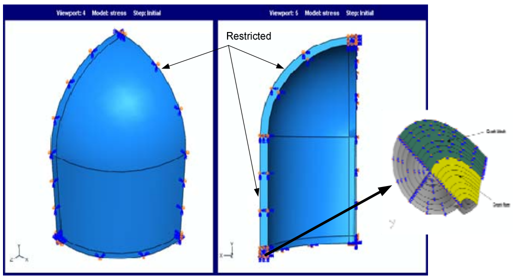


Fig.8 Schematic of 1/8 pressure vessel with boundary conditions

A. Boundary and initial conditions for mass diffusion analysis

For mass diffusion analysis, it is needed to have one initial and two boundary conditions. These conditions are derived from experimental paper (Maoqiu Wang, 2005). According to this reference, in initial time the value of the hydrogen concentration is 0.17 ppm that is changed linearly along the thickness. Also, the value of pressure is $0.9\sigma_B$ Mpa. For outside surface, there is not any flux of concentration, so the value of concentration is zero. Fig. 11 shows these boundaries more clear. Moreover, the type of element is illustrated in this Fig.9.

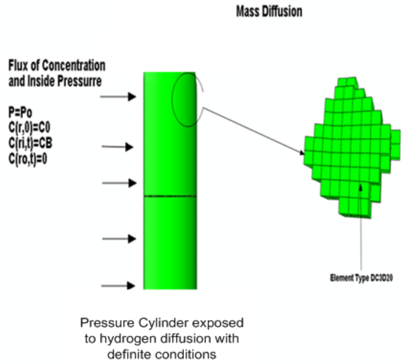


Fig. 9 Schematic of boundary and initial conditions for mass diffusion

B. Using suitable elements and property meshing

Three different kinds of element are used for modeling and analysis such as structure element (C3D20R), wedge element (C3D15R) and mass diffusion element (DC3D20). The structure elements are used in crack block and for showing singularity at crack front, the wedge elements are used. Shapes of these elements are found in Fig. 10.

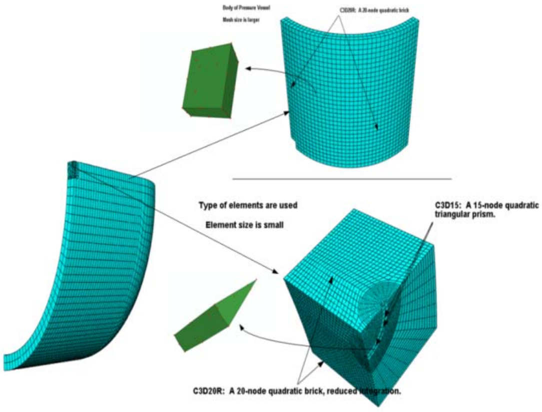


Fig.10 shape of two kinds of elements, structure element and wedge element

IX. FEM Results and Discussion

It is not necessary to solve couple diffusion because the stress-strain response of the steel is negligible. Therefore, the stress distribution is first calculated by the above finite element analysis, and then the hydrogen diffusion is analyzed.

A. Stress analysis

Finite element can be able to evaluate stress analysis in CAE (Complete Abaqus Environment). In this analysis, one modeled pressure vessel with an elliptical crack subjected to inner pressure is analyzed. As a result, contours of stress are created near the crack front. Fig.11 and Fig.12 respectively show contours of stress in mode I and stress distribution. It must be said that the maximum hydrostatic stress is happened near the crack front and Fig. 12 indicates to this fact.

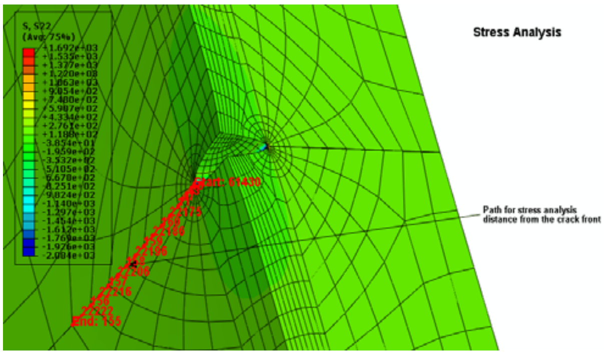


Fig.11 contour of stress near the crack front for mode I Stress

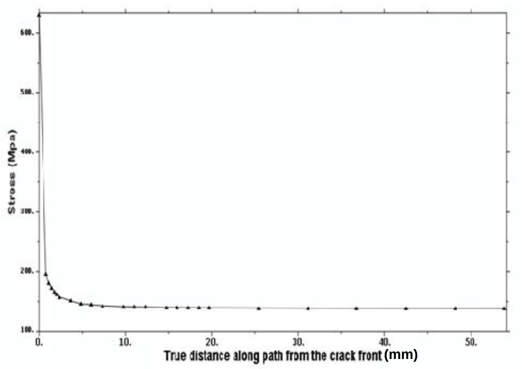
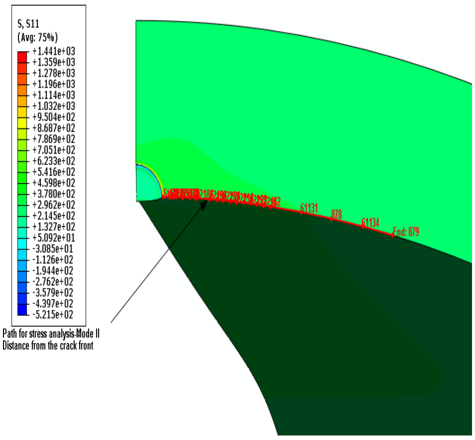
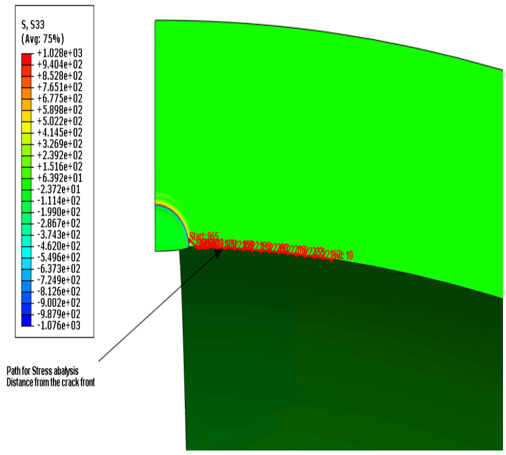


Fig.12 Stress distribution as distance from crack front for one increment-Mode I Stress

If the mode I stress is compared with other modes, definitely the value of stress is biggest. Fig. 13 shows this comparison.



(a)



(b)

Fig.13 Different view port for two mode of stress (a) mode II Stress, (b) mode III Stress

Fig.14 and Fig.15 respectively explain decreasing amount of hydrostatic as increasing distance for mode II and III. Moreover, it should be said that the amount of stress for mode I is the highest value. On the other hand, maximum stress happens near the crack front.

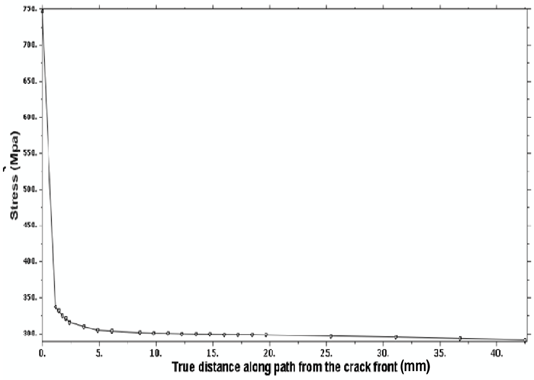


Fig.14 Stress distribution as a function of distance from the crack front- Mode II Stress

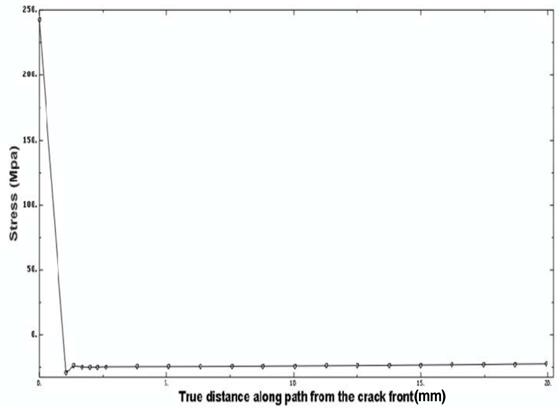


Fig.15 Stress distribution as a function of distance from the crack front- Mode III Stress

B. Mass diffusion analysis

Hydrogen causes to make the material brittle and fast fracture will be happened. It is the significant effect of hydrogen on material properties that always is concerned about it. So, for solving this problem and having safe design, it needed to have distribution of concentration. It is very critical to estimate the life time of the pressure vessel. During the hydrogen diffusion, it leads to accumulate concentration of hydrogen near the crack front. Therefore, this causes to decrease yield stress. The strain region of the high hydrogen concentration in trap site is very small so, using the LEFM has good and acceptable results. At high loading time, the lattice sites which are depleted by trapping are rapidly filled again by hydrogen diffusion and peak to appear at the same position of the hydrostatic peak which is shown in Fig. 16 and Fig. 17.

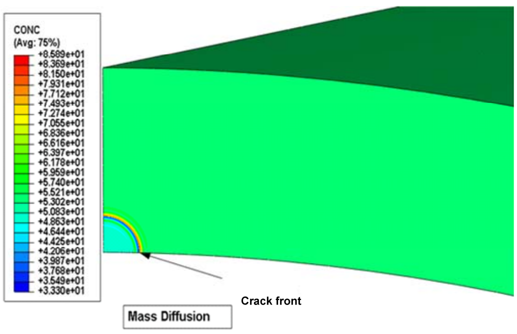


Fig.16 contour of mass diffusion near the crack front

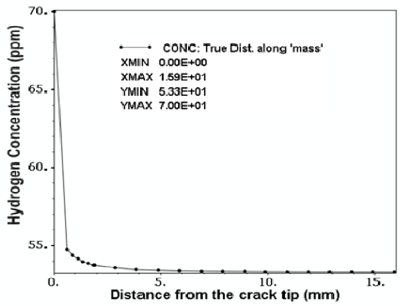


Fig.17 Hydrogen concentration as a function of distance from the crack front or crack tip

Now, it turns to draw a diagram to indicate variety of concentration as a function of time. So, the first work is drawn the different hydrogen concentration for different increments near the crack front. As a result of calculation, the concentration from the crack front is shown in Fig.18. Moreover, this figure shows in each increment, hydrogen increases and reaches to saturated value in the crack front.

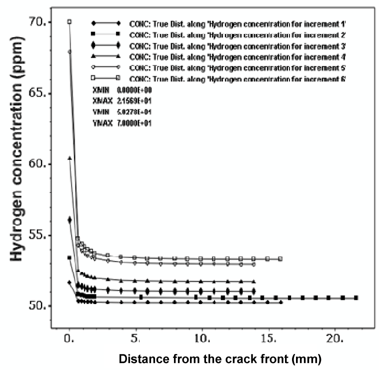


Fig.18 Calculated hydrogen concentration distribution in distance from the crack front for different increment

As a result of FEM, a valid model for stress-assisted hydrogen embrittlement should have some qualities like accumulation of hydrogen happened in a region where hydrostatic stress is maximum and this is the vital parameter exists in the studied model.

C. Validation of results

After modeling of a pressure vessel with a blunting crack, finite element analysis employed to solve sequentially coupled mass diffusion with stress, then the results of FEM model must be valid by experimental work. Fig.19 expresses a comparison experimental hydrogen distribution with finite element analysis. Analysis results are in good agreement with work of Maoqui et al. [19].

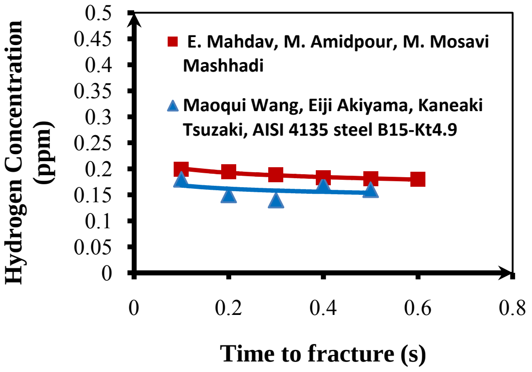


Fig.19 Comparison of hydrogen concentration distribution as function of time for finite element analysis with experimental hydrogen distribution

As it said, calculation of coupled mass diffusion with stress equation to predict the activity of hydrogen embrittlement is the main purpose of this study. Also, it is important for prevention of hydrogen embrittlement failure. Moreover, it is described the mechanism of diffusion. It is essential to know that for providing the conditions to make the material stronger and probably the best choice of it.

Finite element is solved the sequentially coupled mass diffusion with stress problem for AISI 4135 steel b15-Kt. This method is suitable because influence of hydrogen on stress-strain response is not significant on the material which is used and accuracy of comparison confirms that sequentially couple is perfect.

X. Conclusion

Hydrogen embrittlement has great effect on material properties. While hydrogen is diffusing the ductile property is shifted to brittle behavior and consequently causes fast fracture. So, it is essential to have distribution of concentration while hydrogen diffuses in metal. For this purpose, a finite element analysis is implemented to estimate the effect of hydrogen diffusion coupled with stress near the crack front in 3D model. According to the conception and the fact discussed, the maximum hydrostatic and the hydrogen concentration occurred near the crack front and by increasing distance from it, they reduced. As a result, this FEM method is compared with experimental work and there is a good agreement between them.

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