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Review of modeling and simulation of hydrogen induced cold cracking in a low alloy steel weldments

Q. CHEN^{a,b,c}, F. ROGER^a, J. ANGLES^{b,c}, Z. MOUMNI^a, C. ROUBY^a

a. Unité de Mécanique (UME), ENSTA ParisTech, Chemin de la Hunière, 91761 Palaiseau Cedex, France;
b. EDF R&D MRI, 6 quai Wattier, 78400, Chatou, France; c. LaMSID – UMR EDF/CNRS/CEA 8193 EDF R&D, 1 Avenue du Général de Gaulle, 92141 Clamart Cedex, France.

Résumé:

Durant l'opération de soudage des aciers faiblement alliés, l'hydrogène est introduit par l'arc de soudage et se diffuse dans la zone affectée thermiquement où les microstructures telles que la martensite et la bainite sont présentes. Cela entraîne la fragilisation par l'hydrogène. Sous une contrainte résiduelle assez élevée, la fissuration à froid se produit. Ainsi, la fissuration à froid se produit dû aux trois facteurs: I) une microstructure dure et fragile; ii) une teneur en hydrogène assez élevée et iii) une contrainte critique. Cet article résume les travaux dans la littérature et propose un mécanisme de fissuration à froid qui permet de modéliser ce phénomène. L'objectif de la thèse est de déterminer un critère de fissuration à froid à partir des essais expérimentaux et de la simulation numérique par la méthode des éléments finis.

Abstract:

In high strength steel weldments, hydrogen is introduced by the arc of welding and diffuses to the heat affected zone where susceptible microstructures such as martensite and bainite are present. This causes hydrogen embrittlement. At critical residual stress, the Hydrogen Induced Cold Cracking (HICC) occurs. Thus, HICC occurs due to three factors: i) a susceptible microstructure; ii) sufficiently high concentration of diffusible hydrogen and; iii) a critical stress intensity. This article reviews previous works of this subject and proposes a mechanism of HICC which allows further modeling of this phenomenon. The main aim is to determine a cold cracking criterion by experimental tests associated with numerical modeling of these tests using finite element method.

Mots clefs: Cold cracking, hydrogen embrittlement, modeling

1 Introduction

Hydrogen Induced Cold Cracking (HICC) or Cold Cracking (CC) occurs in the low alloy steel weldments as a result of the combination of three factors [1, 3, 9]: i) a susceptible microstructure; ii) sufficiently high concentration of diffusible hydrogen and; iii) a critical stress intensity.

During the welding process, hydrogen is introduced to the weld metal from the moisture or other hydrogenous compounds in the electrode, the covering flux, atmosphere and the weld material [1, 2]. A part of hydrogen diffuses out of weldments during the solidification, another part is trapped in hydrogen traps such as dislocations, grain boundaries, inclusions etc, and the third part diffuses to the Heat Affected Zone (HAZ). This causes Hydrogen Embrittlement (HE) of HAZ microstructure. In low alloy steels, fast cooling rate in the HAZ usually generates hard microstructures such as martensite, bainite which are extremely susceptible to HE. Thus, HICC occurs in the HAZ under local residual stress or restraint stress. Historically, welding parameters were chosen to avoid HICC in the HAZ [4]. But later research has revealed that HICC occurs in the weld metal (WM) as well if high strength filler metal is used. We can identify HICC by several features [1]:

1. it occurs in a delayed manner from several seconds to several days after the welding;
2. temperature lower than 200°C;
3. hydrogen and a susceptible microstructure are both present at the crack tip.

This document is to review the mechanism of HICC and propose a quantitative assessment of the three factors in order to determine a criterion of cold cracking.

2 Mechanisms of HICC

Microscopic observation is an important step to understand the mechanism of HICC. Alam et al. [3] found by experimental observations that the fracture morphology of crack initiation in HSLA80 (High Strength Low Alloy) and X80 steels was Microvoid Coalescence (MVC) for high hydrogen weld metal and Quasi-Cleavage (QC) or Intergranular (IG) for relatively low hydrogen weld metal. This observation confirms the basic aspect of HE proposed by Thompson and Bernstein [1]: IG, QC and MVC modes. And the QC mode is further stated as the particular characteristic of HE. Other than hydrogen concentration, the stress intensity factor is also a key element to determine the fracture mode. Beacham [5] suggested the brittle IG mode predominates under low level of stress intensity. The fracture mode changes to QC and finally MVC at higher stress intensity. This is because IG mode involves the least amount of plastic deformation and hence needs the least energy to fracture.

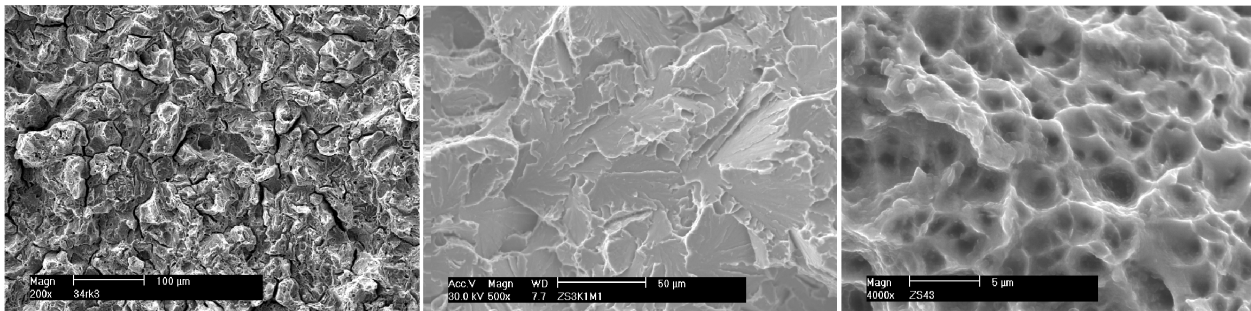


FIG. 1 – Three fracture modes: intergranular and transgranular cleavage fracture (left), quasi-cleavage fracture (middle) and microvoid coalescence fracture (right) [6].

HICC occurs because hydrogen in microstructure causes HE, which leads to be prone to cracking under loading. Embrittlement literally means the loss of ductility of a material, making it brittle. The brittleness is related to fracture toughness which is defined by the energy absorbed per volume before rupture. For microstructures, where the dislocation density is relatively low (low stress or high toughness), the brittle IG mode dominates. For microstructures where the dislocation density is relatively high (high stress or low toughness), the ductile MVC mode dominates.

In the literature, three most investigated models are [7, 10, 22]: i) Hydrogen Enhanced Decohesion (HEDE); ii) Hydrogen Enhanced Localised Plasticity (HELP) and iii) Adsorption Induced Dislocation Emission (AIDE). HEDE was first discovered by Troiano and then developed by Oriani and Gerberich et al.. This mechanism states that atomic hydrogen weakens the interatomic bonds of metal at the crack tip and the surrounding process zone by perturbing the electronic charge density. HELP was first introduced by Beacham and then studied by Birnbaum et al.. It confirms that hydrogen increases dislocation mobility and causes hydrogen to accumulate at voids ahead of the crack tip. This mechanism is considered to be the principal reason of MVC fracture. The third model AIDE has been studied by Lynch [7] and Petch [1, 8]. It proposes that hydrogen absorbed at the crack tip and weakens the interatomic bonds, hence decreasing the surface energy. Thus it further enhances emission of dislocations at the crack tip and causes the propagation of the fissure. This mechanism could be used at the step d of our model. It confirms the fact that hydrogen enhances the propagation of fissures.

Another mechanism [1, 6, 8, 10] used by lots of workers for HSLA steels : Internal Pressure Model by Zapffe and Sims which advocates that atomic hydrogen accumulates at internal defects and combines to molecular hydrogen, which develops high internal pressures;

However, none of these proposals are general enough to account for all experimental observations of HE. This is why we proposed the general schema of HE in FIG. 2 which accounts for most mechanisms. Any particular case of HE can be explained based on this model.

The HE takes the following steps:

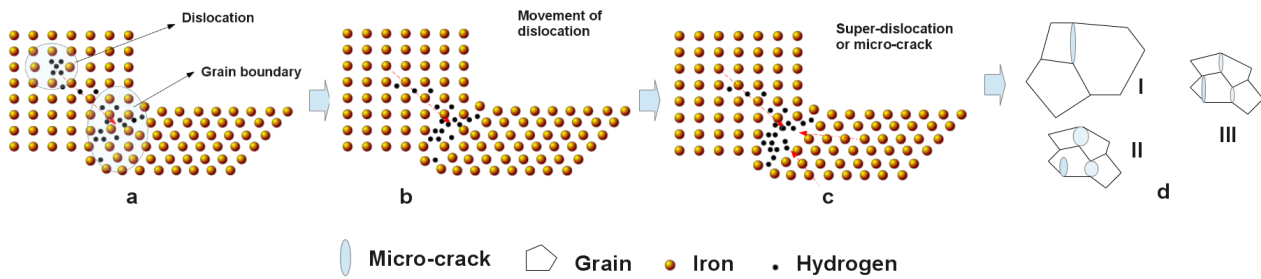


FIG. 2 – Hydrogen embrittlement mechanism

- Hydrogen is trapped in dislocation and increases its mobility. On the mean while, dislocation transports hydrogen.
- Hydrogen trapped in grain boundaries weakens the boundary bonds, and dislocations are stopped at grain boundaries. Other defects such as inclusion or micro-cracks can also be the obstacles for the movement of dislocations.
- More dislocations pile up at grain boundaries and create a cluster of super-dislocation or a micro-crack. Depending on the grain size, the hardness of grain, and weakness effect of lattice bond by hydrogen, micro-cracks show different forms.
- For those materials, whose grains are coarse and hard, the shape of micro-cracks are sharp and acute (case **d I** in the figure). Stress at the tip of cracks is high, while the bond at boundaries is weakened by hydrogen, so cracks propagates in long distance. The fracture is IG mode. For those whose grain size is fine and not hard (case **d II**), the micro-cracks become micro-voids. Voids grow under stress and internal hydrogen pressure due to high hydrogen concentration until coalescence. For those with fine grain size, but hard grain (case **d III**), micro-cracks can both grow through grain boundaries and in the grain, it's the QC mode of fracture, which blends cleavage facets with areas of dimple rupture.

3 Modeling and simulation of HICC

Once we have understood the HICC mechanism, our aim is to find a way to predict this risk. The modeling of HICC consists of evaluating the three factors quantitatively and finds out their relationship in order to determine a criterion of cracking.

3.1 Microstructure: the HAZ hardness

High strength steels are prone to HICC. Debiez [12] used implant test (a type of cold cracking tests) to study the weldability of different high strength steels and concluded that the yield strength could be used to evaluate the susceptibility to HICC. Yurioka and Suzuki [1] propose the HAZ hardness as a rough index. They proposed a formula to model the HAZ hardness by investigating two factors: post-welding cooling rate and chemical composition (or carbon equivalent) of steels. Davidson et al. [9] summarize the results of several researchers and conclude that neither hardness nor strength is a reliable indicator of HICC susceptibility. They have also investigated some particular microstructures like second phase particles, grain boundary ferrite, and conclude that these particular microstructures are more reliable indicators. Since some microstructures such as martensite and bainite are particularly susceptible to HICC, Yurioka and Suzuki [1] also propose the hardenability which represents how easily martensite can be obtained instead of hardness to be the HICC susceptibility indicator.

Our model of HE in FIG. 2 supposes that hardness is a general index of HICC susceptibility, but some particular microstructures such as second phase particle or grain size can cause different mode of fracture. Since IG and QC fractures tend to occur at lower stress intensity, a microstructure that promotes IG fracture seems to be more susceptible to HICC. For different steels, hardness may not be a suitable indicator since some particular elements can promote HICC. But if we study the HICC susceptibility of one steel, hardness can be regarded as HICC susceptibility indicator.

The work of Yurioka et al. [14] takes account of chemical composition and martensite fraction. Their formula predicts the HAZ hardness for almost all the ferritic steels. Later, Nicholas and Abson [15] have summarized the work of Yurioka et al. and the work of Lorenz and Düren, and give the empirical equations

which consider both the cooling time and the carbon equivalent (CE). For fully transformed martensite, the HAZ hardness depends only on the CE. For partially transformed martensite, the HAZ hardness depends both on the CE and the cooling rate. So the key point in determining the HAZ hardness is to evaluate the martensitic transformation and the cooling rate since the CE is already known for a particular steel. In our work, we use a Continuous Cooling Transformation (CCT) phase diagram to model the martensitic transformation.

3.2 Diffusible hydrogen

Hydrogen induced degradation has been discussed in section 2. Here, we are more interested in evaluating and controlling hydrogen concentration in the HAZ. As we have addressed, hydrogen is dissociated at high temperature and enters into the weld metal. A huge part will diffuse out during the cooling stage. Another part is trapped by defects. The third part diffuses to the HAZ. It's this third part called *diffusible hydrogen* which is attributable to HICC [7, 16, 17]. This can be justified by the fact that a post-heating of 150°C which allows diffusible hydrogen to diffuse out of HAZ can eliminate the HICC. This is why Olson et al. [18] proposed the approach of decreasing diffusible hydrogen by introducing some special additions to the WM to serve as hydrogen traps.

Another method of controlling diffusible hydrogen is by the selection of weld metal martensite start temperature (M_s) [18, 19]. The solubility of hydrogen in austenite is very high but its diffusion rate is relatively low, in contrast to ferrite and martensite. So a higher M_s in the WM causes a rapid hydrogen transport to the HAZ and creates a peak in the fusion line, since the low diffusion rate in the HAZ doesn't allow efficient diffusion of hydrogen to the parent metal. This is usually the case of underbead HAZ cracking in high strength parent metal. By contrast, a higher M_s in the HAZ can efficiently decrease hydrogen concentration in the HAZ since it allows hydrogen to transport a significant long distance to the parent metal. This will transfer HICC from the HAZ to the WM.

In order to modeling hydrogen diffusion, we use the Fick law which postulates that the flux J of hydrogen is proportional to the gradient of hydrogen concentration C :

$$J = -D \frac{dC}{dx} \quad (1)$$

where D is the diffusion coefficient.

But this law is no longer valid in the case of non-uniform solubility S . For a metal where external forces like hydrostatic stress or temperature gradient result in non-uniform solubility. At saturation state, the concentration of hydrogen equals the solubility. Since the solubility is not equally the same, there exists a gradient of concentration which, by the Fick law, should lead to a flux. This is obviously contradictory since saturation state is a stationary state where non additional flux is allowed. This is our particular case because a huge temperature gradient exists during the welding process. Thus, we use the more general *Einstein relation* [16] which postulates that the flux is proportional to the gradient of activity $A = \frac{C}{S}$.

$$J = -DS \frac{dA}{dx} \quad (2)$$

External forces can be regarded as resulting in variation of the solubility. The Sievert's law takes account of the temperature T effect in the solubility:

$$S = S_0 \sqrt{P_{H_2}} e^{\frac{-\Delta H}{RT}} \quad (3)$$

where S_0 is a constant; P_{H_2} is the pressure of external hydrogen; ΔH is the activation energy of hydrogen mobility; R is the gas constant.

The positive hydrostatic stress can be considered as increasing the activation energy because it requires more energy for hydrogen to move in the metal lattice. Plastic deformation or dislocation can be regarded as increasing the solubility proportionally since it can trap more hydrogen. It can otherwise be modeled as traps which are described by the Mac Nabb&Foster equation [20]:

$$\frac{\partial \theta}{\partial t} = kC(1 - \theta) - \mu\theta \quad (4)$$

where θ is the coverage ratio of the trap sites; k is the rate constant of capture in the trap sites; μ is the rate constant of escape from the trap sites;

By applying the principle of mass conservation, the modeling of hydrogen diffusion can be described by:

$$\begin{cases} \frac{\partial C}{\partial t} + \frac{\partial(N\theta)}{\partial t} = -\nabla J \\ \frac{\partial \theta}{\partial t} = kC - \mu\theta \end{cases} \quad (5)$$

where N is the trap site density per unit volume.

The laser induced breakdown spectroscopy can follow the distribution of hydrogen across the weldment and verify the hydrogen diffusion model [18].

3.3 Stress state

Stress in welding process can be from either i) residual welding stress due to the non-uniform temperature distribution or ii) external loading. This leads to two types of different experimental tests [21]: i) self-restraint cold cracking tests such as CTS, Tekken, Lehigh etc; ii) externally loaded cold cracking tests such as Implant test. These tests usually use notched specimens in order to create a local structural high stress to promote initiation of cracking. So the structural effect should be taken account when evaluating the critical stress. We'll use the Tekken test in the investigation of HICC because this test is very close to the real HICC problem in our industrial application.

Another factor that can significantly affect the stress is the phase transformations in both WM and HAZ. Modeling of residual stress requires a full consideration of these factors by using thermal elastoplastic analysis for low temperature and viscoplastic analysis for high temperature. A lot of researchers (Inoue et al., Leblond et al., Fisher et al., Moumni et al., Waeckel) [23] have tried to model the residual stress after welding, especially by taking account of the transformation induced plasticity. The work of Moumni et al. [23] on the 16MnD5 steel has taken our particular attention since we use the same material in our research. The results in their work show a fair agreement with experimental tests. This gives us a confidential evaluation of residual stress in our tests. Further validation of modeling of residual stress in our tests could be done by measurement of local residual stress. The work of Wingrove [24] offers several methods of measurement of restraining loads and hence can be used in our work.

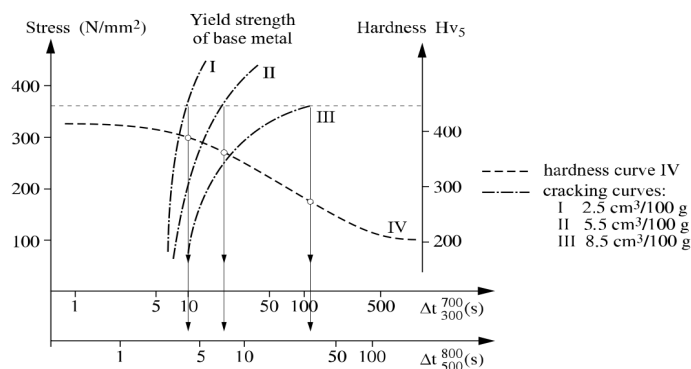


FIG. 3 – Criterion of hydrogen induced cold cracking [25].

So we can propose a theoretical criterion of the following general form:

There exists a functional f , expressed in terms of some internal variables $\alpha_1, \alpha_2, \dots$, defines the security state of HICC. The security threshold is defined by $f(\alpha_1, \alpha_2, \dots) < 0$.

We can easily identify these internal variables by the three factors: the HAZ hardness, hydrogen concentration and critical stress. FIG. 3 illustrates this general idea of HICC criterion. This review doesn't give an explicit formulation of this functional, but rather offers a methodology to arrive at this point.

4 Conclusion and perspective

We have discussed the mechanism and methodology of modeling the three factors of HICC. In order to explicitly reveal the criterion formulae of HICC, the cold cracking tests should be done by varying welding conditions to investigate the effect of each factor. By changing the hydrogen proportion in the protection gas of welding, we can vary hydrogen concentration in the HAZ. By using different welding energy, we can change the HAZ hardness, diffusible hydrogen concentration and residual stress. Finally, different geometries of specimen in the Tekken tests can change significantly the local residual stress magnitude. The final failure or not of specimens allows us to give the explicit expression of f .

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