

Technical Note: Hydrogen Diffusion in Welded Joints and Its Relation to Lamellar Tearing

BY E. BETZ AND H. K. LEUNG

ABSTRACT. A mathematical model is developed to analyze the transient hydrogen diffusion in typical welded joints and its effect on stress fields that lead to lamellar tearing.

The study indicates that if lamellar tearing occurs at an early stage of the cooling cycle, viz. at 200 to 300 C (392 to 572 F), hydrogen does not contribute to the failure. However, if tearing takes place at a later stage, viz. after the weld has cooled to ambient, then three factors—through-thickness ductility, boundary restraints, and hydrogen—interact to cause the failure. Thus it is suggested that hydrogen only plays a supporting role in lamellar tearing.

Introduction

During a welding process, hydrogen is introduced into the weld through the consumed electrode coating, flux, and inherent surface moisture. When the weld cools, it diffuses into the base metal, where it stays and creates various unfavorable effects.¹⁻³ For example, lamellar tearing, which is a cracking phenomenon featured by step-like tears inside the base metal, has been related to hydrogen embrittlement in the weld.

At present, the significance of hydrogen in the mechanism of lamellar tearing is still not clear. Earlier researchers believe that the role hydrogen plays is minor.⁴⁻⁶ Later studies indicate that hydrogen embrittlement can be a prime factor in lamellar tearing.^{7,8} However, no conclusive proof has been offered, nor is there a well defined theory behind these two schools of thought.

The authors have attempted to postulate a theory in which lamellar tearing is considered to be a two-stage possibility in the crack formation process.

At the early stage of the cooling cycle, the ductility of steel is lowest at

200 to 300 C (392 to 572 F). This lack of ductility can cause tears to initiate at inclusion sites just outside the heat-affected zone, but before hydrogen can diffuse into this region. Otherwise, a second stage possibility comes into play. As the metal cools to room temperature and hydrogen is diffused into the base metal, hydrogen interaction at inclusion sites can further reduce the ductility and cracking may be initiated. This theory implies that hydrogen acts in a supporting role in the failure of lamellar tearing.

In order to verify the above theory, and to study quantitatively the interlocking relationship of thermal cycle, hydrogen embrittlement, inclusions, and stress-strain levels, a computer-aided numerical analysis of typical joints has been made. This note describes the mathematical model used in the hydrogen diffusion analysis, and from the results, assesses the role that hydrogen has at both stages in which lamellar tearing can initiate.

Formulation of Model

A modified form of Fick's 2nd Law on diffusion of hydrogen in steel is used as a basis for developing a three dimensional model which takes into account the coupled plastic strains, various void and inclusion populations, and the evolution of free hydrogen at the surfaces of the joint.

Thus if C is defined as the volume concentration, then:⁹

$$\begin{aligned} C &= N/V, \\ C^* &= N/L, \text{ and} \\ C &= \phi \cdot C^* \end{aligned}$$

where N = number of hydrogen atoms, V = volume, C^* = dislocation

concentration, and L = dislocation length.

ϕ is called the local dislocation density and is related to the plastic strain ϵ_p in the stress field by the following equation:

$$\phi = \alpha \cdot \epsilon_p + \beta$$

in which α and β are temperature dependent parameters. If the hydrogen flux J is assumed to be caused by the gradient of C^* , viz.,

$$J = -\gamma \cdot D \cdot \frac{\delta C^*}{\delta x}$$

with γ being a proportional constant and D the coefficient of diffusion, then the equivalence to Fick's 2nd Law can be obtained from conservation of matter as:

$$\frac{\delta C}{\delta t} = \gamma \cdot D \cdot \frac{\delta^2 C^*}{\delta x^2},$$

where $\frac{\delta C}{\delta t}$ is the change of concentration of hydrogen with time.

Expressing the above equation in three dimensions gives:

$$\frac{\delta C}{\delta t} = \gamma \cdot D \cdot \Delta^2 C^* \quad (1)$$

$$\text{or} \quad \Delta^2 \left(\frac{C}{\phi} \right) = \frac{1}{\gamma D} \cdot \frac{\delta C}{\delta t} \quad (1a)$$

Note that for no plastic strain, $\phi = \beta = \gamma$, giving the simple Fick's 2nd Law:

$$\frac{\delta C}{\delta t} = \frac{\delta}{\delta x} \left(D \cdot \frac{\delta C}{\delta x} \right) \quad (2)$$

Due to the existence of micro-voids, a further modification of the diffusion equation is required to provide accommodation of hydrogen in interstitial solution and for hydrogen molecules occupying a void. Thus, there is a

E. BETZ is Associate Professor in Mechanical Engineering and H. K. LEUNG is a Post Doctoral Research Fellow, The University of Newcastle, New South Wales, Australia.

state of equilibrium existing between the two forms of hydrogen and their relationship can be represented by the following differential equation of diffusion:

$$\Delta^2 C_1 = \frac{1}{D} \cdot \frac{\delta}{\delta t} (C_1 + C_v) \quad (3)$$

where C_1 is the interstitial hydrogen concentration and C_v is the hydrogen concentration in the void (both in gm H_2/mm^3 metal), and equilibrium is governed by the following equation:¹⁰

$$C_1 = K \sqrt{\frac{C_v R \cdot T}{v_o F}} \cdot \exp(b \cdot F \cdot C_v R / v_o) \quad (4)$$

where $K = 4.883 \times 10^{-3} \cdot \exp(-3453/T)$, (cm · dyne^{-1/2}); T = temperature, (°K); $R = 3.709 \times 10^3$, (erg · °K⁻¹ cm⁻³); $b = 7.049 \times 10^{-7}$, (°K · cm² dyne⁻¹); $F = 1.11 \times 10^7$, (cm³ H_2 · mm³ metal/gm H_2 · cm³ metal); and v_o = fractional void volume, (mm³ void/mm³ metal).

Again, introducing ϕ as per equation (1a), equation (3) is modified to give:

$$\Delta^2 \left(\frac{C_1}{\phi} \right) = \frac{1}{\gamma D \xi} \cdot \frac{\delta}{\delta t} (C_1 + C_v) \quad (5)$$

where $D\xi$ is the coefficient of diffusion (mm²/sec) of hydrogen in a virtually clean steel with no plastic strain, and is a function of temperature.

Equation (5) is the diffusion equation in which there is no hydrogen exchange with the boundary. If surface evolution is to be included, the equation becomes:

$$\sum_{j=1}^3 \frac{a_j h_j}{\Delta l_j} \cdot \frac{(C_a - C_1)}{\gamma} + \Delta^2 \left(\frac{C_1}{\phi} \right) = \frac{1}{\gamma D \xi} \cdot \frac{\delta}{\delta t} (C_1 + C_v) \quad (6)$$

where h_j = surface evolution coefficient (1/mm); Δl_j = small finite control length (mm); a_j = multiplier whose value depends on the location where evolution occurs; and C_a = ambient hydrogen concentration (gm H_2/mm^3 metal).

Finally, if external hydrogen input is considered (as a result of hydrogen transfer during the welding process), with mass flux \dot{m} gm H_2/s say, then the complete diffusion equation is:

$$\sum_{j=1}^3 \frac{a_j h_j}{\Delta l_j} \cdot \frac{(C_a - C_1)}{\gamma} + \Delta^2 \left(\frac{C_1}{\phi} \right) + \frac{1}{\gamma D \xi} \cdot \left(\frac{\dot{m}}{\Delta u} \right) = \frac{1}{\gamma D \xi} \cdot \frac{\delta}{\delta t} (C_1 + C_v) \quad (7)$$

in which Δu is the control volume of a

small element (mm³) and the initial condition

$C_{1o} + C_{vo} = C_o$, the initial hydrogen content specified in the steel component (gm H_2/mm^3 metal), is assumed.

The three dimensional finite difference explicit method is used to solve the diffusion problem. The finite difference interpretation of equation (7) for anywhere (both inside and on the boundary) of a variable grid model can be expressed in the equation:

$$(C_1 + C_v)_{l,m,n}^p = (C_1 + C_v)_{l,m,n}^{p-1} + \gamma \cdot \Delta t \cdot (D^p [e] [A] [E^p])_{l,m,n} \quad (8)$$

where the second term on the R.H.S. contains a scalar function obtained by multiplication of the three matrices:

[e], a 1×71 element matrix, the values of which depend on the location of the point in the grid,

[A], a 71×10 constant matrix in which the coefficients of the differential equations are stored, and

[E^p], a 10×1 matrix containing terms of diffusion, evolution, and mass transfer of hydrogen at any time p for any point l,m,n.

Application of the Hydrogen Diffusion Model in Lamellar Tearing

There are two aspects by which the participation of hydrogen in lamellar tearing could be evaluated. They are:

1. *Hydrostatic pressure set up by hydrogen existing at voids and debonded inclusion sites.* For a real gas (such as hydrogen), its state can be expressed by van der Waals' equation:¹¹

$$\left(P + \frac{a \cdot n^2}{V^2} \right) \cdot (V - n \cdot b) = n \cdot R \cdot T \quad (9)$$

where n = number of mole of gas present and a , b are constants.

Applying the appropriate constants to a material with fractional void volume v_o and rearranging gives:

$$P = 2.89 \times 10^{-4} C_v T / (v_o - 6.32 \times 10^{-5} C_v) \quad (\text{atmo.}) \quad (10)$$

Hence hydrostatic pressure P is a function of temperature, hydrogen concentration in voids, and fractional void volume.

2. *The effects of hydrogen on material properties in calculating transient stresses, strains, and ductility losses during a welding cycle.* The temperature history of the weld is first determined using techniques similar to the hydrogen diffusion problem. The hydrogen diffusion program "HYDROGEN" then runs jointly with a three dimensional finite element program "WELDSTRESS," by which the stress-strain field developed due to temperature changes and boundary constraints

is calculated.

It is noted that in the formation of the hydrogen diffusion model, the term ϕ is used to incorporate the effect of plastic strains on hydrogen diffusion. The hydrogen concentration effect is in turn accounted for when considering the properties of the material during the stress-strain analysis. If we define:

$$DL_z = \frac{\epsilon_z(x,y,z,t,T)}{\epsilon_{totz}(z,t,T,C_T)} \quad (11)$$

where $\epsilon_z(x,y,z,t,T)$ is the transient through-thickness strain for any point at time t after the weld has been started, and $\epsilon_{totz}(z,t,T,C_T)$ is the maximum strain possible in the through-thickness direction at temperature T when the material has the additional effect of hydrogen imposed on it, then DL_z is readily obtainable from "WELDSTRESS." From the increase in the DL_z values due to hydrogen at the region just outside the heat affected zone, the effects of hydrogen in causing possible onset of cracks can be assessed.

Interpretation of Results and Discussions

For discussion purposes, hydrogen diffusion results are considered for a typical T-joint using electrodes of hydrogen content equal to 15 cc/100 gm deposited metal. Here, it is found that hydrogen concentration reaches a maximum of 5.22 cc/100 gm metal on the weld face in the short time of 0.5 s after the electrode has passed the point concerned. Then, very rapidly, the hydrogen atoms diffuse towards the parent metal and the concentration on the weld face remains very low throughout the rest of the cooling cycle.

Just inside the heat-affected zone, a maximum concentration of 2.6 cc/100 gm metal is reached in 4 s after the electrode has passed. In this region, hydrogen first exists as interstitial atoms; then the majority of it is transformed into gas molecules, occupying the space of the voids, producing a hydrostatic pressure. The gas molecules, however, do not stay in the voids permanently. As more interstitial atoms diffuse into regions of lower concentrations, the remaining atoms and molecules reorganize themselves until equilibrium is again restored. The diffusion rates are very sluggish, and it takes more than 1 hour for hydrogen to diffuse out of the heat affected zone to a significant extent.

Studies also show that hydrogen concentration and hydrostatic pres-

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