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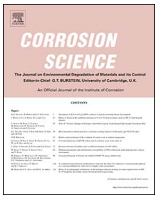

Study on hydrogen enrichment in X80 steel spiral welded pipe

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ABSTRACT

Local hydrogen enrichment would increase the risk of hydrogen embrittlement in welded structures, so the dynamic hydrogen diffusion process and final concentration distribution were investigated numerically by finite element simulation for a hydrogen gas transport X80 steel spiral welded pipe. The results show that the change of microstructure induced by weld thermal cycle would decrease the hydrogen concentration in welded joint, but welding residual stress could cause local hydrogen enrichment. Equivalent hydrogen pressures for base metal, weld metal and three subregions in heat-affected zone were put forward in order to scientifically evaluate the influence of hydrogen enrichment on hydrogen embrittlement.

1. Introduction

Nowadays, about 3000 km of hydrogen transport pipelines operate in the world under a maximum pressure of 10 MPa, and these pipelines are generally built in low-strength grade steels, not higher than X52 [1]. High-strength steel is favorable for reducing pipe wall thickness and increasing operating pressure, however, high-strength steel is prone to hydrogen embrittlement (HE) [2–7]. HE is the process by which metals such as steel become brittle and fracture due to the introduction and subsequent diffusion of hydrogen into the metal. HE caused by service environment is a typical kind of environmental degradation, and it can seriously endanger the safety of hydrogen transport pipeline. Numerous studies have been conducted to investigate the adaptability of applying high-strength pipeline steel in transporting hydrogen gas. Briottet [1] and Moro [8] studied the influence of average hydrogen concentration on mechanical properties of X80 steel, while Nanninga [9], Jin [10] and Park [11] studied the influence of chemical composition, microstructure or inclusions on hydrogen-induced failure. Zhang [12] and da Silva [13] studied the susceptibility of steel welded joints to HE or hydrogen induced stress cracking. However, the influence of local hydrogen enrichment has not been considered. There is severe microstructure inhomogeneity [12,14] and residual stress [15] in welded zones, which would induce hydrogen diffusion [16]. This nonuniform diffusion makes local hydrogen concentration in welded zone perhaps much higher than the average hydrogen concentration in base mental. Sims and Skluev found that the reduction in area at fracture would decrease with the increase of hydrogen concentration (C_H) in steel, which will eventually reach stable value when C_H rises to several parts per million (ppm) [17]. Sanchez also put forward an empirical correlation and showed that the strain at fracture decrease with C_H in high strength steel [18]. Therefore, the welded joints with high C_H will be liable to hydrogen-induced failure. It is essential to determine the distribution of C_H in welded pipe so as to scientifically evaluate the HE risk for hydrogen transport pipeline.

Microstructure inhomogeneity is an important factor that induces hydrogen diffusion and leads to uneven hydrogen distribution. Microstructures of metals are determined by its chemical compositions, thermal process and stress-strain status. Fusion welding process can change microstructures of pipeline steel, so there are great differences between heat-affected zones, weld metal and base metal. Different microstructures would have different crystal lattice parameters and hydrogen traps (such as dislocations, grain boundary, impurity atoms, inclusions, microvoids). Bouhattate found that the effective initial hydrogen concentration increases with the density of hydrogen traps, while the variation of effective diffusion coefficient has a reverse rule [19]. Inhomogeneous microstructure can induce nonuniform hydrogen diffusion in welded area. Studies have been performed on hydrogen permeation behavior influenced by welding heat input [20] and weld thermal cycles [21,22]. However, these studies were conducted zone by zone by using specimens with uniform microstructure, while different zones coexist in an actual welded joint. No reports have been found on the hydrogen distribution in welded steel pipeline in hydrogen environment, not to mention combining the influence of welding residual stress.

Stress gradient is another key factor that induces hydrogen diffusion. Welding residual stress exists objectively in welded joints of

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pipeline. The stress field is not uniform. The stress gradient can induce hydrogen diffusion. By conducting electrochemical permeation experiments under preset constant tensile stresses [23–26] or under slow-strain-rate tension [27], numerous studies on effects of stress/strain on hydrogen solubility, diffusivity and trapping have been performed. However, only the average hydrogen diffusion parameters in the test specimen can be obtained by the above hydrogen permeation tests under tension. The obtained parameters can reflect the change of crystal lattice and hydrogen traps (such as dislocations and microvoids) induced by strain in the material, but cannot reflect the effect of stress gradient. By applying tritium radioluminography [28], scanning Kelvin probe force microscopy [29] or atom probe tomography [30], distribution of hydrogen in materials could be examined. Therefore the hydrogen diffusion induced by stress gradient can be examined as well. Unfortunately, these measurements are very strict with the specimen preparation. Meanwhile, preparation process can release internal stress, and the measured results cannot reflect the influence of stress on hydrogen enrichment in welded joints. No reports have been found on the effective test techniques that study the influence of stress gradient.

Some authors calculated the distribution of hydrogen concentration in front of a crack by numerical simulation [31,32], and similarly the stress gradient induction on hydrogen diffusion in steel welded joint can be studied. For example, finite element simulation was conducted by Jiang [33] to investigate the effect of welding residual stress on hydrogen diffusion in welded joint of a spherical tank. Nevertheless, Jiang's calculation considered the welded joint as a homogeneous material, and various welded zones adopted the same hydrogen diffusion parameters, thus ignored the influence of microstructure change on hydrogen enrichment. In addition, the precondition of simulation calculation is to known the sub-surface concentration of atomic hydrogen (c_0) at the charging surface and the solubility (s) and diffusion coefficient (D) of hydrogen in the studied material. Presently, electrochemical charging in liquid electrolyte is widely used to study the impact of heat treatment conditions [12], welding heat input [20], and stress levels [34] on hydrogen diffusion parameters of pipeline steel. However, the atomic hydrogen concentration c_0 at the charging surface is determined by charging environment. The electrochemical reactions that occur on the charging side of specimen in liquid electrolyte would lead to the generation of deposits [35], therefore, the c_0 obtained from electrochemical charging cannot reflect the c_0 in gaseous charging conditions. Gaseous charging, however, has been usually performed at high temperature and relative low pressure [36–38], and the hydrogen parameters would be totally different from that of hydrogen gas at ambient temperature and high pressure. Furthermore, the matching gas stream measurement technique is less efficient than the electrochemical hydrogen detection [39]. The authors developed a hydrogen permeation device which combines gaseous charging with electrochemical hydrogen detection. Using this device, hydrogen permeation tests could be conducted on various materials in a high-pressure hydrogen-containing gas [21]. The obtained hydrogen permeation parameters can be used as the initial or boundary conditions for calculating hydrogen distribution in welded joints of hydrogen-containing gas transporting pipeline.

In the present work, a combined experimental and finite-element modeling approach for an effective study of hydrogen diffusion behavior in X80 steel spiral welded pipe was carried out. Hydrogen permeation parameters, such as c_0, s, and D, were measured in base metal, heat affected zones and weld metal using the gaseous charging − electrochemical detection technique. A comprehensive three-dimensional modeling considering the interaction of hydrogen diffusion, microstructural inhomogeneity and transient stress-strain was established to perform transient hydrogen analysis in the welded joint of X80 pipeline steel. Equivalent hydrogen pressures for various zones in welded joint were put forward in order to scientifically evaluate the influence of hydrogen enrichment on hydrogen embrittlement.

2. Basic theory of hydrogen diffusion

Using finite element method to investigate hydrogen diffusion based primarily on the control equation and constitutive equations [40]. Considering hydrogen diffusion in a body with volume V and surface A, mass conservation requires that the rate of total hydrogen inside V is equal to the flux through A:

$$\int_V \frac{dc}{dt}dV+\int_A n\cdot JdA = 0 \tag{1}$$

where c is the hydrogen concentration, J is the flux of hydrogen concentration, n is the outward normal to A, and $n{\cdot}J$ is the flux of concentration leaving A.

The derivative from Formula (1) using divergence theorem [41] is then determined:

$$\int_V \left(\frac{dc}{dt} + \frac{\partial}{\partial X}\cdot J\right)dV = 0 \tag{2}$$

Because the volume is arbitrary, this provides the pointwise equation:

$$\frac{dc}{dt} + \frac{\partial}{\partial X}\cdot J = 0 \tag{3}$$

The equivalent weak form is

$$\int_V \delta\phi\left(\frac{dc}{dt} + \frac{\partial}{\partial X}\cdot J\right)dV = 0 \tag{4}$$

Where $\delta\phi$ is an arbitrary, suitably continuous, scalar field.

This statement can be rewritten as

$$\int_V \left[\delta\phi\left(\frac{dc}{dt}\right) + \frac{\partial}{\partial X}\cdot(\delta\phi J) - J\frac{\partial\delta\phi}{\partial X}\right]dV = 0 \tag{5}$$

Using the divergence theorem again yields

$$\int_V \left[\delta\phi\left(\frac{dc}{dt}\right) - \frac{\partial\delta\phi}{\partial X}\cdot J\right]dV + \int_A \delta\phi n\cdot JdA = 0 \tag{6}$$

The diffusion is assumed to be driven by the gradient of a chemical potential, $\frac{\partial\mu}{\partial X}$, and the relationship between $\frac{\partial\mu}{\partial X}$ and J can be expressed as [42]:

$$J = -\frac{Dc}{R(T-T_Z)}\cdot\frac{\partial\mu}{\partial X} \tag{7}$$

where D is the diffusion coefficient of hydrogen, R is the gas constant, T denotes the absolute temperature, T_Z is the absolute zero on the temperature scale used, μ is chemical potential. Here, μ is defined as:

$$\mu = \mu_0 + R(T - T_Z)ln\phi + \sigma_h V_H \tag{8}$$

where μ_0 is a fixed datum without considering the influence of stress, V_H is the partial molar volume of hydrogen in the solid solution, here is $2\ cm^3{\cdot}mol^{-1}$ [42], σ_h is the hydrostatic stress, ϕ is the activity of hydrogen, and

$$\phi = \frac{c}{s} \tag{9}$$

where c is the mass concentration of hydrogen, s is its solubility in the metal.

Introducing the Eqs. (8) and (9) into Eq. (7) yields

$$J = -SD\cdot\left[\frac{\partial\phi}{\partial X} + \phi ln\phi\frac{\partial}{\partial X}(ln(T-T_Z)) + \phi\frac{V_H}{R(T-T_Z)}\frac{\partial p}{\partial X}\right] \tag{10}$$

Let K_s is the "Soret effect" factor, providing diffusion because of the temperature gradient, K_p is the pressure stress factor, providing diffusion driven by the gradient of the equivalent pressure stress, $p = -\sigma_h$. Data for K_s and K_p can be calculated from the equations

$$K_s = \phi ln\phi \tag{11}$$