Scripta Materialia 154 (2018) 101–105

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Regular article

Peculiar temperature dependence of hydrogen-enhanced fatigue crack growth of low-carbon steel in gaseous hydrogen

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ARTICLE INFO

Article history:
Received 6 March 2018
Received in revised form 16 May 2018
Accepted 21 May 2018
Available online xxxx

Keywords:
Fatigue crack growth
Low-carbon steel
Hydrogen embrittlement
Trap-site occupancy
Dislocation

ABSTRACT

The peculiar temperature dependence of hydrogen-enhanced fatigue crack growth (HEFCG) of low-carbon steel in hydrogen gas was successfully interpreted in terms of 'trap-site occupancy' of hydrogen. HEFCG decreased with increasing temperature in hydrogen gas at 0.7 MPa and 298 to 423 K due to lower occupancy of trap sites at higher temperatures. In hydrogen gas at 90 MPa, HEFCG was insensitive to the temperature because most of the trap sites were occupied by hydrogen, regardless of the temperature. Trap sites with a binding energy of 47 kJ/mol, corresponding approximately to the dislocation core, dominated the temperature dependence of HEFCG.

The temperature dependence of crack growth remains of considerable interest in relation to the understanding of fracture phenomena associated with hydrogen embrittlement (HE) and stress corrosion cracking (SCC), as the fracture process is intricately affected by both chemical reactions (for example, trapping of hydrogen or dissolution of material) and physical effects (for example, crack growth). For example, time-dependent crack growth (da/dt) in fully hardened AISI4130 in hydrogen gas at pressures below 0.1 MPa decreases with increasing temperature in the range from 193 to 273 K [1], but at temperatures from 298 to 353 K, the opposite tendency is observed. Williams and Nelson [1] pointed out that the process of bulk transport of hydrogen is not an important factor in crack growth, because the measured crack-growth rates were two to five orders of magnitude faster than the maximum expected diffusion rate. They concluded that the fractional coverage of initial adsorption sites at a crack tip is important for crack advancement. The rate of fatigue crack growth (FCG), da/dN, in 2Ni–Cr–Mo–V rotor steel in hydrogen gas at 0.24 MPa also decreases with increasing temperature in the range 296 to 358 K [2]. The same tendency has also been reported by Marrow et al. [3], who mention the importance of the occupancy of hydrogen traps and transport to the vicinity of cracks, although they did not perform a specific analysis of the relationship between trap-site occupancy and hydrogen-enhanced FCG (HEFCG). Note that these tendencies were observed in low-pressure hydrogen-gas environments only.

The general opinion regarding the temperature dependence of SCC is that higher temperatures lead to increased rates of fracture, i.e., da/dt and/or da/dN [4–7]. The progress of SCC, especially intergranular SCC, has been associated with an apparent activation energy for grain boundaries, as the fracture process is markedly dependent on chemical reactions at grain boundaries [7]. In the temperature dependence of SCC reported by Landes and Wei [4], Saito [5], Gerberich et al. [6], and Arioka et al. [7] the value of da/dt or da/dN increased with increasing temperature T, a tendency opposite to that reported by Williams and Nelson [1], Smith and Stewart [2], and Marrow et al. [3]. These opposing tendencies associated with hydrogen-gas embrittlement and SCC might be important in relation to achieving a physical understanding of the intrinsic effect of hydrogen gas on the crack-growth process.

Although the temperature dependence of FCG in low- to high-pressure hydrogen gas is a critical issue in achieving a comprehensive understanding of the mechanism of HEFCG, no experimental results obtained in high-pressure hydrogen gas have been reported, as the availability of appropriate facilities has hitherto been extremely limited. We recently developed systems for performing FCG tests in hydrogen gas in high pressures of up to 140 MPa and at temperatures from 233 to 423 K, which permit the verification of the temperature dependence of HEFCG in high-pressure hydrogen gas. Various experimental findings revealed that the rate of FCG in high-pressure hydrogen gas has an upper bound at test frequency ranging from 0.001 Hz to 5 Hz in low-

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https://doi.org/10.1016/j.scriptamat.2018.05.035
1359-6462/

carbon steels [8], low-alloy steels [9], and austenitic stainless steels [10,11].

This study is the first to examine FCG in hydrogen gas at 0.7 and 90 MPa over a wide range of temperatures (298–423 K), with the aim of verifying the temperature dependence of HEFCG in a low-carbon steel (JIS-SM490B). The steel used in this study was of commercial grade with a chemical composition (mass%) of C 0.16, Si 0.44, Mn 1.43, P 0.017, S 0.004 and Fe balance. Its 0.2% proof stress and tensile strength were 360 and 537 MPa, respectively. Compact-tension specimens with a width of 50.8 mm and a thickness of 10.0 mm were used for all the FCG tests. Before the FCG tests, a pre-crack was introduced from the starter notch in each specimen by applying cyclic loading in air at room temperature (RT ≈ 298 K). According to ASTM standard E647-05 [12], FCG tests under a constant load range (ΔP) were performed to measure the FCG rate (da/dN) versus stress intensity factor range (ΔK) at a test frequency of 1 Hz and a stress ratio of 0.1 in hydrogen gas at 0.7 and 90 MPa at temperatures ranging from 298 K to 423 K, as well as in air.

The da/dN–ΔK curve (Fig. 1) clearly showed that HEFCG occurred under all the test conditions. HEFCG in hydrogen gas at 0.7 MPa was most significant at RT (~298 K), and its intensity decreased with increasing temperature, as reported previously [1–3]. The relative rate of FCG rate compared with that in air at 298 K, $(da/dN)_H/(da/dN)_{air}$, at ΔK = 25 MPa·m$^{1/2}$ was ~27 at 298 K and ~2.5 at 423 K. Interestingly, in hydrogen gas at 90 MPa, the intensity of HEFCG was not particularly dependent on the test temperature; there was an intense acceleration of FCG in 90 MPa hydrogen gas, even at 393 K, unlike the FCG tests in 0.7-MPa hydrogen gas. In 0.7-MPa hydrogen gas at 298 K (Fig. 2(a)) and 90-MPa hydrogen gas at 298 K (Fig. 2(b)), a remarkable FCG acceleration occurred, the fatigue crack became straighter, and slip deformation was localized, compared with FCGs in air (Fig. 2(c)) or in 0.7-MPa hydrogen gas at 423 K (Fig. 2(d)). Furthermore, as shown in Fig. 2 (e) and (f), striation resulting from fatigue crack propagation under the remarkable FCG acceleration was less distinct compared with that in air (Fig. 2(g)) or under a slight FCG acceleration (Fig. 2(h)). The FCG rate, da/dN at ΔK = 25 MPa m$^{1/2}$ for four environmental conditions: p_H = 0.7 MPa at T = 298 K in Fig. 2(e), p_H = 90 MPa at T = 298 K in (f), in air in (g), and p_H = 0.7 MPa at T = 423 K in (h) are 1.76 × 10^{-6}, 2.42 × 10^{-6}, 1.02 × 10^{-7}, and 2.67 × 10^{-7} m/cycle, respectively. The averaged striation spacing, s, corresponds approximately to the FCG rate for each condition as represented in Fig. 2(e)–(h), respectively.

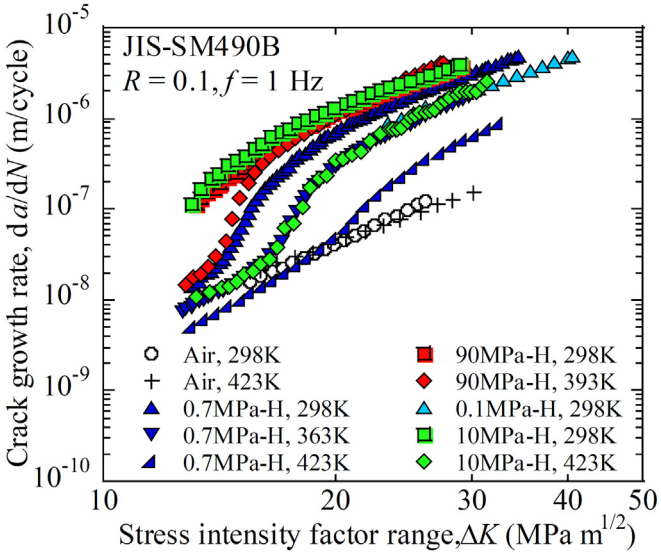

Fig. 1. FCG curve for the low-carbon steel JIS-SM490B in air and in hydrogen gas at pressures of 0.7 and 90 MPa at 298–423 K.

The fracture morphology in 0.7-MPa hydrogen gas approached that in air at higher temperatures. A large crack blunting led to clearly visible striations in Fig. 2(g) and (h). A general mechanism for striation formation under FCG has been well described by Laird and Smith [13], Bichler and Pippan [14]. The indistinct striation under HEFCG implies that the crack maintains a sharp shape without blunting during the loading process. A macroscopic mechanism for HEFCG has been proposed in the Hydrogen-Induced Successive Crack Growth (HISCG) model of Matsuoka et al. [15], which was derived from the earlier Hydrogen-Enhanced Successive Fatigue Crack Growth (HESFCG) model [16].

On the basis of our experimental evidence, we related the concept of trap-site occupancy based on thermal-equilibrium theory [17] to HEFCG; this permits discussion of the temperature dependence of the HEFCG over a wide range of hydrogen-gas pressures. The relationship between the trap-site occupancy θ_X [N_{HX}/N_X] and the lattice-site occupancy θ_L [N_{HL}/N_L] (where N_X is the number of trap sites in a unit cell and N_{HX} is the number of trap sites occupied by hydrogen in a unit cell) is given by following equation if $\theta_X \gg \theta_L$, as is the accepted case for a bcc system:

$$\frac{\theta_X}{1-\theta_X} = \theta_L \exp\left(\frac{E_b}{RT}\right) \tag{1}$$

Here, E_b is the binding energy of a trap site (kJ/mol), R is the universal gas constant (8.314 J·K^{-1}·mol^{-1}), and T is the absolute temperate (K). The value of θ_L can be obtained by unit conversion from [H atom/Fe atom] to lattice site occupancy [N_{HL}/N_L] in the empirical equation [18], if we assume that hydrogen is preferentially trapped at tetrahedral sites:

$$\theta_L = 3.08 \times 10^{-4}\sqrt{F}\exp\left(-\frac{3440}{T}\right) \tag{2}$$

where F is the fugacity (MPa), as defined in Eq. (3):

$$F = p_H \exp\left(\frac{bp_H}{RT}\right) \tag{3}$$

Here, p_H is the hydrogen-gas pressure (MPa) and b is a constant (1.584 × 10^{-5} m^3/mol) [19]. A lower value of T or a higher value of p_H results in a higher value of θ_X. The temperature dependence of θ_X therefore becomes smaller as the pressure rises.

To make it easier to understand the temperature dependence of HEFCG in hydrogen gas at various pressures, Fig. 3(a) shows the FCG acceleration ratio $(da/dN)_H/(da/dN)_{air}$ at ΔK = 25 MPa·m$^{1/2}$ as a function of p_H. The trap site occupancy, θ_X, was calculated by the following steps. The value of θ_L was calculated by using Eqs. (2) and (3), considering that hydrogen is preferentially trapped at tetrahedral site in BCC system. Secondary, $(da/dN)_H/(da/dN)_{air}$ was assumed to be proportional to θ_X: $(da/dN)_H/(da/dN)_{air} = \alpha\theta_X + 1$, where α is a proportionality coefficient. Then, two unknown parameters: the binding energy of a trap site, E_b, and the proportionality coefficient, α, were determined by fitting $(da/dN)_H/(da/dN)_{air} = \alpha\theta_X + 1$ to the experimental data with the least-squares method; consequently, it was obtained that α = 25 and E_b = 47 kJ/mol. Finally, the values of θ_X under various pressures and temperatures were calculated from Eq. (1) by using the obtained values of θ_L and E_b as shown in Fig. 3(c), then plotted in Fig. 3(b). For all the experimental data, the assumption that $(da/dN)_H/(da/dN)_{air}$ was proportional to θ_X seems to be acceptable from Fig. 3(b) and the tendency of HEFCG was roughly described by the fitted curves as known from Fig. 3(a). This means that the temperature dependence of HEFCG is a result of trapping of hydrogen by sites with a binding energy of 47 kJ/mol, which are mainly attributed to the dislocation core [20]. The temperature dependence of the trap-site occupancy θ_X decreases with increasing hydrogen-gas pressure. At a pressure of 90 MPa, most trap sites with E_b = 47 kJ/mol are occupied by hydrogen, regardless of the