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Multi-scale observation of hydrogen-induced, localized plastic deformation in fatigue-crack propagation in a pure iron

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ABSTRACT

In order to study the influence of hydrogen on plastic deformation behavior in the vicinity of the fatigue crack-tip in a pure iron, a multi-scale observation technique was employed, comprising electron channeling contrast imaging (ECCI), electron back-scattered diffraction (EBSD) and transmission electron microscopy (TEM). The analyses successfully demonstrated that hydrogen greatly reduces the dislocation structure evolution around the fracture path and localizes the plastic flow in the crack-tip region. Such clear evidence can reinforce the existing model in which this type of localized plasticity contributes to crack-growth acceleration in metals in hydrogen atmosphere, which has not yet been experimentally elucidated.

Hydrogen-assisted fatigue crack-growth (HAFCG) in structural steels [1–4] is of significant concern, particularly with respect to the storage of compressed gaseous hydrogen in a future hydrogen energy-based economy. In the presence of hydrogen, two types of cracking behavior are generally accepted in steels, i.e., stress-controlled fracture in high-strength steels and strain-controlled, quasi-static fracture in low-to medium-strength steels [5]. In the former case, hydrogen triggers brittle cracking along the grain boundaries and the intensity of crack acceleration becomes more pronounced with a lower loading rate (delayed fracture) [6,7]. In contrast, crack extension is mediated by a quantity of plastic flow and the fracture is frequently characterized by quasi-cleavage, showing no delayed cracking, as in the latter case [8]. Whereas several successful models have been proposed to demonstrate the behavior of stress-controlled fracture [6,7], due to its complex nature, the detailed mechanisms of strain-controlled cracking are still not fully understood.

Extensive investigations for the strain-controlled HAFCG have also been conducted by the authors' research group, with the resulting proposal of one possible model to describe the series of phenomena; hydrogen-induced successive crack-growth (HISCG) [9–13]. The main role of hydrogen in this model is to concentrate plastic flow in crack-tip region, in accordance with the hydrogen-enhanced localized plasticity (HELP) [14,15], leading to suppressed crack-tip blunting and faster crack propagation. However, critical evidence is still lacking to justify the use of the model, although similar mechanisms have also been implied by several researchers, based on the fractographic features of fatigue-tested or statically-cracked samples in hydrogen atmosphere [16,17].

One of the reasons for this lack of strong evidence is that most of the experiments have been performed on materials with complex microstructures, several alloying elements and sometimes even two or more phases. All of these factors make interpretation difficult and thus, the analysis of the hydrogen effect on the fracture processes is further complicated. Hence, in this study, pure Fe was targeted in order to avoid those complications. Additionally, most of the past studies have been conducted either on macro- or nano-scale, lacking any link between the phenomena of the two different scales. Dislocation structures

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have recently attracted attention as the key to understand the mechanism of hydrogen-induced degradation [18–23]. Transmission electron microscopy (TEM) is used for analyzing dislocation structures, but characteristics of the TEM samples (e.g., local plastic deformation and crack morphology) are often not clarified in as much detail as can be obtained via other techniques. In this study, we present an example of the successful utilization of electron channeling contrast imaging (ECCI) and orientation imaging microscopy (OIM), in combination with TEM imaging, with the aim of elucidating the crack-tip deformation in HAFCG on different scales. Specifically, ECCI and OIM are used to characterize crack morphology on a microscale, while TEM reveals the dislocation structures at specific locations on a smaller scale.

In order to demonstrate the hydrogen-enhanced, localized, plastic deformation in the fatigue crack-tip region, fatigue crack-growth (FCG) tests were performed in a hydrogen atmosphere. Various observation techniques were employed to prove that hydrogen dramatically reduces the plasticity expansion around the fatigue crack-tip, while significantly affecting dislocation structure evolution just beneath the fracture surface.

The material used was a commercial, pure Fe plate, with the chemical composition of 0.001%C-0.07%Mn-0.010%P-0.003%S. The yield stress, σ_Y, and tensile strength, σ_B were 133 MPa and 252 MPa, respectively. Compact-tension (CT) specimens with a width, W, of 50.8 mm, and a thickness, B, of 10.0 mm, were cut from L–T orientation of the plate.

FCG tests were performed in air and 0.7 MPa hydrogen at room temperature (RT), according to the ASTM E647 [24] standard, under a constant load range condition (ΔP-constant test), in order to identify the relationship between the FCG rate, da/dN, and the stress intensity factor range, ΔK. The load range, R, and the test frequency, f, were 0.1 and 1 Hz, respectively. By using a low carbon steel, it was confirmed that FCG rate in 0.7 MPa hydrogen peaks out at f of 0.1–1 Hz [11]. Therefore, we chose $f = 1$ Hz as a preferable test frequency to characterize the HAFCG phenomena also in pure iron. Moreover, FCG tests were also conducted under the ΔK-constant condition so as to observe the hydrogen effect on the crack propagation morphology at a fixed ΔK value ($\Delta K = 17\ \text{MPa}\cdot\text{m}^{1/2}$).

The fracture surfaces formed in ΔP-constant tests were observed by SEM operated at 15 kV. After the ΔK-constant tests, specimens were cut along near mid-thickness section and the surfaces were polished with colloidal SiO_2. Crystallographic orientations were analyzed along the fracture path by EBSD, with a beam step size of 1 μm. Based on the crystal orientation maps, the strain distribution along the cracks was also examined using grain reference orientation deviation (GROD) method. Here, the average orientation of each grain was used as an orientation reference, with a deviation angle from the reference at every scanned point being mapped for each grain. Additionally, ECCI was conducted with an accelerating voltage of 30 k for obtaining an overview of the microstructural evolution along the fracture path. TEM samples perpendicular to the mid-thickness surfaces of the ΔK-constant test specimens were extracted using the JEOL JIB-4500 multi-beam machine. The samples were evaluated using a JEOL 2100F, field-emission-TEM, operated at 200 kV in scanning mode (STEM).

The ΔP-constant tests revealed the existence of two distinctive regions on the da/dN-ΔK curve in hydrogen, as indicated in Fig. 1: (i) the low ΔK regime, where the da/dN value was approximately equal to that in air (Stage I) and (ii) the high ΔK regime, with a high acceleration rate (Stage II). Yoshikawa et al. [11] investigated the HAFCG of a low-carbon steel in 0.7 MPa hydrogen gas, reporting that the material exhibited an upper limit of FCG acceleration, about 20 times greater in hydrogen than in air, regardless of testing frequency, viz., the material did not exhibit delayed cracking and the strain-controlled fracture was dominant even in hydrogen atmosphere. FCG rates in the low-carbon steel, as obtained in the same environments [11], were also plotted in Fig. 1. The FCG curves are consistent with each other, indicating that the crack propagated accordingly via the identical mechanism in the two materials.

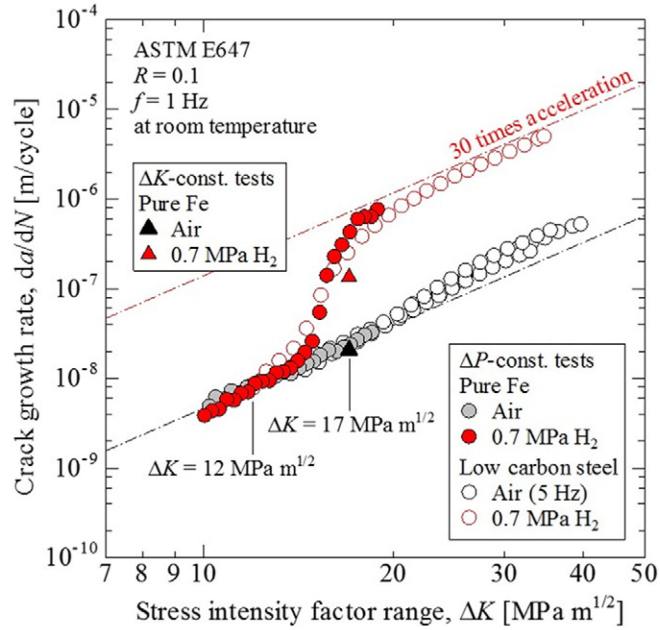

Fig. 1. FCG curves of pure Fe in air and in 0.7 MPa hydrogen gas, including the results obtained for a low-carbon steel, as reported in a previous paper [11].

Fig. 2 presents the fracture surfaces observed via SEM. Whereas ductile striations were formed throughout the entire ΔK range in air, the fracture morphology in hydrogen changed dramatically. Fig. 2 (b) reveals the fracture surface in Stage I, as formed in hydrogen. Even though there was no acceleration of FCG at this stage, surprisingly, almost half of the fracture surface was covered by intergranular (IG) facets with a brittle appearance while none of IG features were visible in case of air. The IG fracture in HAFCG was also reported in previous researches [4, 25,26]. However, even though the IG facets seemed brittle macroscopically, numerous slip traces were found at higher magnifications (region A in Fig. 2 (b)), implying the mediation of plasticity during the fracture process. Martin et al. [21] reported the formation of similar slip traces on the IG fracture surface of hydrogen-charged pure Ni, and concluded that dislocation movements play an important role in the grain boundary (GB) separation. Specifically, the hydrogen distributed inside the grain was transported by moving dislocations, then deposited into the GBs, in conjunction with the piling-up of dislocations, resulting in the local satisfaction of stress concentration and hydrogen agglomeration for GB decohesion [27]. Similar explanations may be applicable in the present case, although the direct correlation between the IG fracture and the macroscopic FCG rate remains unclear and the detailed analysis of stage I is beyond the scope of this article.

In association with the onset of FCG acceleration, an abrupt transition of fracture morphology was observed in hydrogen (Fig. 2 (c)). In contrast to the IG fracture in Stage I, the fracture in Stage II was characterized by faceted, transgranular planes (quasi-cleavage = QC), frequently observed on the hydrogen-affected fracture surfaces of low- to medium-strength steels [1,8,11]. Moreover, a magnified image of this QC (region B in Fig. 2 (c)) revealed the existence of brittle-like striations which ran perpendicular to the macroscopic FCG direction. It was confirmed that the average striation spacing was roughly consistent with the macroscopic FCG rate at the corresponding ΔK value. Chen and Gerberich [28] reported similar striations on {100} cleavage planes of Fe-3%Si single crystals during sustained-load experiments in hydrogen, as the evidence of intermittent, time-dependent, crack propagation. However, consistency of striation-spacing and the FCG rate in the present study implies that the striations were formed on a cycle-by-cycle basis, and their formation mechanism was different from that reported by Chen and Gerberich. The formation mechanism of such brittle-like striations in hydrogen was studied by Matsuoka et al. [10,29].