Decrease in Fatigue Crack Initiation Life by Irreversible Hydrogen in Cold Drawn Eutectoid Steel

M. Nakatani[1], M. Sakihara[1], K. Minoshima[1]
[1]*Osaka University, Osaka, Japan*

1. Introduction

Despite of numerous researches on hydrogen embrittlement in high strength steels, the hydrogen embrittlement mechanism [1-3] is still unclear. This reason is that it is uncertain how hydrogen trapping state in steel is related to the strength degradation by hydrogen embrittlement. Hydrogen trapping state in steel has been quantitatively analyzed by thermal desorption spectrometry [4, 5]. This method is being applied to the research on hydrogen embrittlement [6]. In steel, hydrogen is trapped at the lattice defects such as vacancies, dislocations and second phase particles (precipitates and inclusions). Furthermore, hydrogen trapped at above trap sites is classified into two kinds of states which are respectively called diffusible and irreversible hydrogen trap sites [4]. Diffusible hydrogen can diffuse at room temperature and contributes to hydrogen embrittlement under static loading. On the other hand, irreversible hydrogen does not influence the static strength. This is because binding energy between hydrogen and its trap site is high and irreversible trap sites prevent hydrogen from diffusing to crack tip [5]. From this view point, the generation of irreversible trap site by microstructural control [7, 8] is one of the effective methods to improve the resistance to hydrogen embrittlement.

Machine components are subjected to cyclic loading, and the fatigue properties are very important for the design. Therefore, the influence of hydrogen on the fatigue strength has been investigated by many researchers [9]. However, these researches focused on the influence of diffusible hydrogen on the fatigue strength because of the fact that diffusible hydrogen contributes to hydrogen embrittlement under static loading. Although the generation of irreversible trap site is considered to be a candidate method to improve the resistance to hydrogen embrittlement, the influence of irreversible hydrogen on the fatigue strength has not been investigated. In this study, the fatigue tests were conducted to investigate the influence of irreversible hydrogen on the fatigue strength in cold drawn eutectoid steel. It is found that irreversible hydrogen decreases the fatigue life [10]. To clarify the contribution of irreversible hydrogen to fatigue crack initiation and propagation, we used sharp-notched samples for the fatigue tests, and thereby the fatigue crack propagation rate and crack initiation life was estimated. We discussed the contribution of irreversible hydrogen to crack initiation and/or propagation.

2. Experimental procedures

The material used was cold drawn eutectoid steel wire with diameter of 1.8 mm. The material is consisted of the heavy deformed pearlitic microstructure. Chemical composition and mechanical properties are listed in Tables 1 and 2,

Table 1 Chemical composition (mass %).

C	Si	Mn	P	S	Cu	Ni	Cr	Al	Mo	N	O	Fe
0.80	0.17	0.52	0.008	0.004	0.01	0.01	0.02	0.001	0.01	0.0036	0.0040	bal.

Table 2 Mechanical properties.

Tensile strength [MPa]	Reduction of area [%]
2040	39

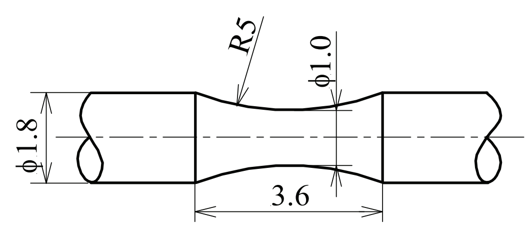


Fig. 1 Shape and dimensions of the center part of a specimen.

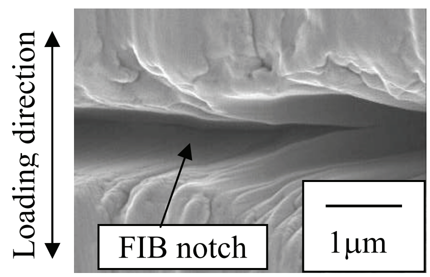


Fig. 2 FESEM images of the cross section of FIB notch.

respectively. Figure 1 shows the shape and dimensions of the center part of a specimen for fatigue tests. The specimen surface was electrolytically polished to remove the work-hardening layer. The stress concentration factor is about 1.05. In experiments, V-shaped small notch (width: 10 μm, height: 1 μm, depth: 5.5 μm) was fabricated to the surface of smooth sample using focused ion beam (FIB) system. Figure 2 shows a field emission type scanning electron microscope (FESEM) image of a FIB notch section. The radius of the curvature of the notch root is about 30 nm, and the stress concentration factor of the notch calculated by finite element method (FEM) was about 9.

Hydrogen was charged by a cathodic charging technique in buffer solution (0.2 mol/L CH_3CHOOH + 0.17 mol/L CH_3COONa, pH= 4.8). The current density was kept constant at 500 A/m^2 with charging time of 6 h. Thermal desorption spectrometry (TDS) analysis [11] was conducted in the temperature range from room temperature to 773 K to analyze the hydrogen desorption behavior with a heating rate of 100 K/h. Figure 3 shows the hydrogen evolution profiles. After hydrogen charge, two peaks of hydrogen desorption, shown by dashed line, appeared at 348 K and 553 K, respectively. When the sample was conditioned in dry air for 60 h after hydrogen charge, the hydrogen desorption peak at 348 K disappeared. This means that the hydrogen desorbed out of the sample during 60 h conditioning in dry air. On the other hand, hydrogen desorption peak at 553 K hardly changed after 7 day conditioning. These results indicate that hydrogen desorbed at peak temperature of 348 K and 553 K corresponds to diffusible and irreversible hydrogen, respectively. The hydrogen trap site in a heavy deformed pearlitic microstructure was reported as follows [12]: diffusible hydrogen is

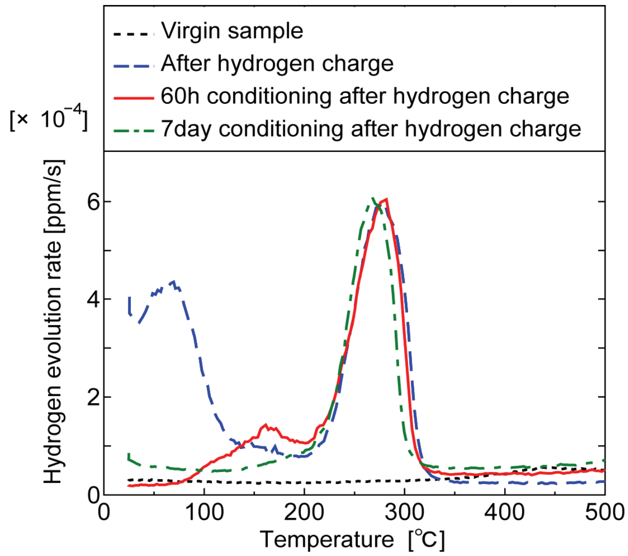


Fig. 3 Hydrogen evolution profile measured by TDS.

mainly trapped at dislocation caused during cold drawing, and irreversible hydrogen is trapped at strained interfaces between ferrite and cementite.
Fatigue tests were performed at a stress ratio R = 0 and a stress cyclic frequency of 30 Hz using an electro-hydraulic servo-controlled fatigue testing machine. To evaluate the influence of irreversible hydrogen on the fatigue strength, internal hydrogen states were changed as follows: (a) virgin sample, (b) irreversible hydrogen charged sample that contained only irreversible hydrogen, which was obtained by 72 h-conditioning in dry air after hydrogen pre-charge. The fatigue tests were conducted in dry air (dew point: 203 K) to prevent hydrogen entry from the testing environment.

3. Experimental results

3. 1 Influence of irreversible hydrogen on the fatigue strength
Figure 4 shows the influence of irreversible hydrogen on the *S*-*N* curves. When a sample did not failure until 10^7 fatigue cycles, higher stress range was applied to the sample until final failure to confirm the crack initation by cyclic loading. The subscripts (a-k) indicate that the same sample used for this low-high two-step fatigue test. The crack initiation site was internal Al_2O_3 inclusion in the case of smooth sample. The influence of irreversible hydrogen on the fatigue strength is not clear because of a large scatter. This scatter of fatigue life was resulted from the scatter of the inclusion size. In the FIB notched sample, on the other hand, the fatigue life and fatigue limit of FIB notched sample decreased compared with that of the smooth sample. Moreover, the slope of the *S*-*N* curve was steeper than that of smooth sample. Note that the influence of irreversible hydrogen was not observed in notched sample.

To compensate the scatter of inclusion size, we evaluated the fatigue strength in terms of stress intensity factor range calculated by the following equation [13]:

$$\Delta K_{\mathrm{ini}} = \begin{cases} 0.5\Delta\sigma\sqrt{\pi\sqrt{area_{\mathrm{ini}}}} & \text{(Internal)} \\ 0.65\Delta\sigma\sqrt{\pi\sqrt{area_{\mathrm{ini}}}} & \text{(Surface)} \end{cases} \tag{1}$$

where ΔK_{ini} is stress intensity factor range for the inclusion or FIB notch, $\Delta\sigma$ is applied stress range, and $area_{\mathrm{ini}}$ is the projected area of an inclusion or FIB notch. Figure 5 shows the relationship between ΔK_{ini} and N_f. The data with the subscript "e" in Fig. 4 was excluded in Fig. 5 because the fracture surface suffered from post-corrosion, and the crack initiation site could not be verified. The difference of the fatigue life between the smooth and FIB notched sample increased as ΔK_{ini} became smaller. It is considered that this was resulted from the difference of the fatigue crack initiation life between smooth and FIB notched sample. This would be related to the difference of stress concentration at crack initiation site. According to FEM calculation, the stress concentration factor of an inclusion was about 1.3 and that of FIB notch was about 9. In a smooth sample, some number of cyclic loading is required for the crack initiation due to low stress concentration. In the FIB notched sample, on the contrary, the crack can initiate at the FIB notch root in the early stage of fatigue because of high stress concentration.

In the case of smooth sample, the fatigue life of irreversible hydrogen charged sample decreased compared with that of virgin sample. The threshold stress intensity factor range of irreversible hydrogen charged sample was smaller than that of virgin sample, where the threshold was defined by maximum ΔK_{ini} at which a sample did not fracture up to 10^7 fatigue cycles. This means that a decrease in the fatigue strength was brought about even by only irreversible hydrogen. However, in the FIB notched sample, irreversible hydrogen did not influence the fatigue strength.

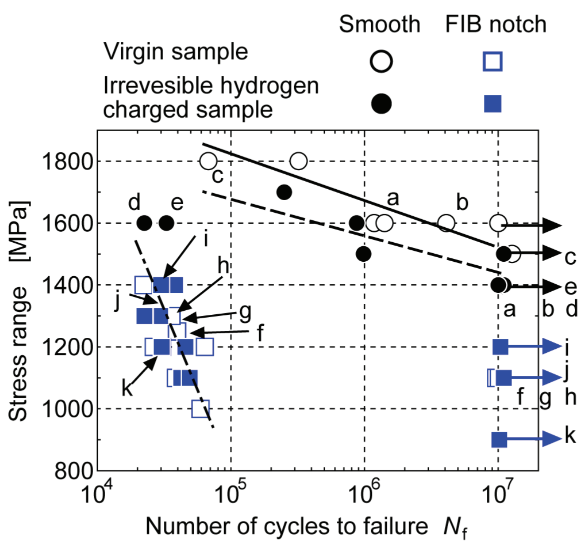


Fig. 4 *S-N* curves of smooth and FIB nothced samples.

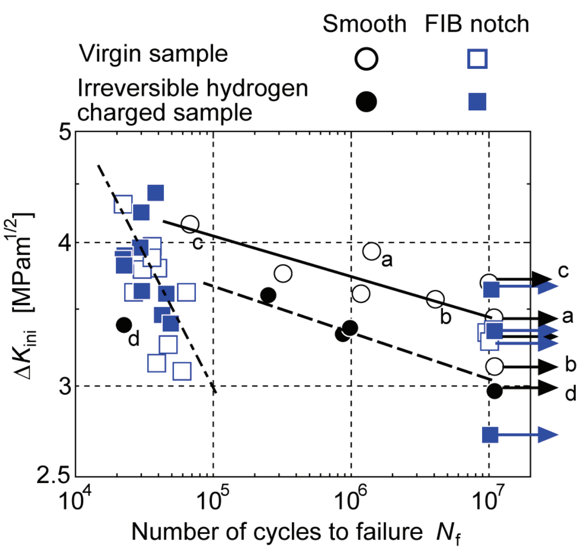
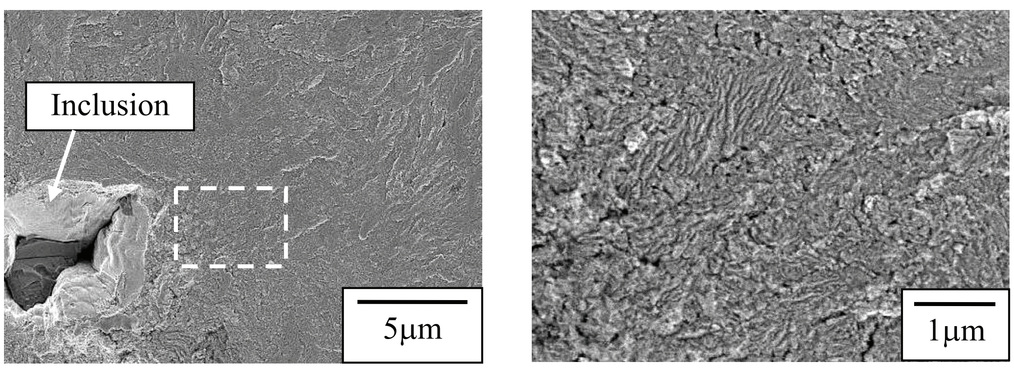


Fig. 5 Relationship between ΔK_{ini} and N_f in the smooth and FIB notched samples.

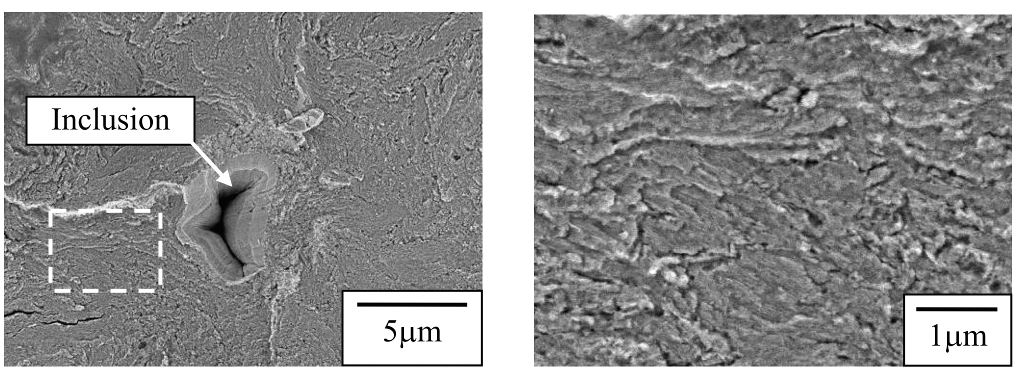
3. 2 Fatigue fracture behavior

To investigate the influence of irreversible hydrogen on the fracture behavior, the fatigue fracture surface was observed by FESEM. Figure 6 shows fracture surfaces near fatigue crack initiation site in a smooth sample. For virgin sample, the ductile granular fracture surface, which looked different from "Optical Dark Area" [14], was observed around the crack initiation sites as shown in Fig. 6(a). For the irreversible hydrogen charged sample, the fracture surface exhibited different fracture surface morphology which was related to the heavy deformed pearlitic microstructure. This change of fracture surface morphology by irreversible hydrogen indicates that irreversible hydrogen influences the fatigue fracture mechanism.

Fracture surfaces near FIB notch root are shown in Fig. 7. Except for this FIB notch, there were not other crack initiation sites such as surface defect or internal inclusion. The fracture surfaces of irreversible hydrogen charged sample were similar to smooth samples which contained irreversible hydrogen. This suggested that irreversible hydrogen, similar to the case of smooth sample, influenced the fatigue fracture of notched sample. In the case of fracture surface away from the crack initiation site, the fracture surface of virgin sample showed ductile transgranular fracture and that of irreversible hydrogen charged sample looked

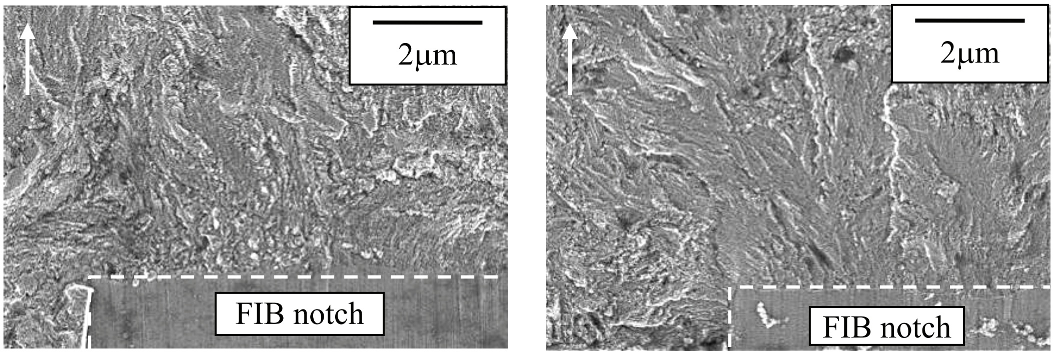


(a) Virgin sample



(b) Irreversible hydroben chaged sample

Fig. 6 Fracture surface near crack initiation site in the smooth sample. Right images are enlarged images of the region enclosed with the dashed rectangle of left figure.



(a) Virgin sample (b) Irreversible hydroben chaged sample

Fig. 7 Fracture surface near crack initiation site in the FIB notched sample. The arrows indicate the crack propagation direction.

brittle in both smooth and FIB notched samples. However, in high ΔK region, $\Delta K > 20$ MPam$^{1/2}$, the fracture surface of irreversible hydrogen charged sample became similar to that of virgin sample, indicating that irreversible hydrogen did not influence the fatigue fracture mechanism. These results suggested that irreversible hydrogen influences not only crack initiation process but also crack propagation process of low ΔK region.

3. 3 Estimation of fatigue crack propagation rate

As mentioned before, the crack propagation life dominates the fatigue life of a FIB notched sample because of high stress concentration at notch root. Based on the relationship between ΔK_{ini} and N_{f} shown in Fig. 5, the fatigue crack propagation rate in cold drawn wire was estimated. Assumed that crack propagation rate da/dN follows Paris's law [15], fatigue life can be calculated by integrating Paris's law from initial crack length or notch depth a_{ini} to final crack length a_{f}. When the crack length at final failure is still longer than initial crack length, the relationship between stress intensity factor range for initial crack length ΔK_{ini} and fatigue life N_{f} is expressed as follow:

$$\Delta K_{\mathrm{ini}}{}^{m} N_{\mathrm{f}} = \frac{2a_{\mathrm{ini}}}{(m-2)C} \tag{2}$$

where m and C are coefficient in Paris's law. Thus, we can calculate m-value by fitting the data shown in Fig. 5 by using the method of least squears. The obtained m-value was about 4.6 irrespective of internal hydrogen state. C-value was then determined to minimize the difference between the experimental fatigue data and the life given by the eq.(2). Then we obtained the crack propagation rate in virgin and irreversible hydrogen charged sample, as follows:

$$(\mathrm{d}a/\mathrm{d}N)_{\mathrm{v}} = 3.7\times 10^{-13} \times \left(\Delta K\right)^{4.6} \tag{3}$$

$$(\mathrm{d}a/\mathrm{d}N)_{\mathrm{h}} = 3.0\times 10^{-13} \times \left(\Delta K\right)^{4.6} \tag{4}$$

where subscripts "v" and "h" indicate virgin and irreversible hydrogen charged samples, respectively. These relations show that irreversible hydrogen hardly influences the crack propagation rate though the fatigue fracture surface of low ΔK region looked brittle.

3. 4 Estimation of fatigue crack initiation life

As discussed in previously, the estimate fatigue crack propagation rate was not affected by irreversible hydrogen. Therefore, this means that irreversible hydrogen influenced the fatigue crack initiation in the smooth sample. We, then, estimated the fatigue crack initiation life in smooth sample in order to clarify the influence of irreversible hydrogen on the fatigue crack initiation. Intenal fatigue fracture process can be divided into fatigue crack initiation, intenal fatigue crack propagation in fish-eye region and surface fatigue crack propagation. Therefore, fatigue life of smooth sample is expressed as follow:

$$N_{\mathrm{f}} = N_{\mathrm{ini}} + N_{\mathrm{fish}} + N_{\mathrm{sur}} \tag{5}$$

where N_{f} the fatigue life, N_{ini} the crack initiation life, N_{fish} the number of cycles which required for internal crack to reach the specimen surface and N_{sur} the number of cycles required from fish-eye crack to final failure. If the fatigue crack propagation properties are known, the fatigue crack initiation life can be obtained by calculating the fatigue crack propagation lives of intenal and surface crack region. The stress intensity factor for internal and surface crack was calcurated from eq. (1). For the surface crack, assuming that the surface crack propagates with constant aspect ratio a/b=1.25 which was an average aspect ratio at final failure of all smooth specimens, the surface fatigue crack propagation life was calculated using the obtained fatigue crack propagation rate, as expressed by eqs. (3) and (4). In the fish-eye region, however, the crack propagation rate may decelerate compared with that in air because of high vaccum [16]. We refered the crack propagation rate in fish-eye region which was obtained by fatigue tests of high strength steel JIS-SCM435 [17]. Note that the influence of irreversible hydrogen on the intenal crack propagation rate was neglected because irreversible hydrogen hardly influence the estimated surface fatigue crack propagation rate.

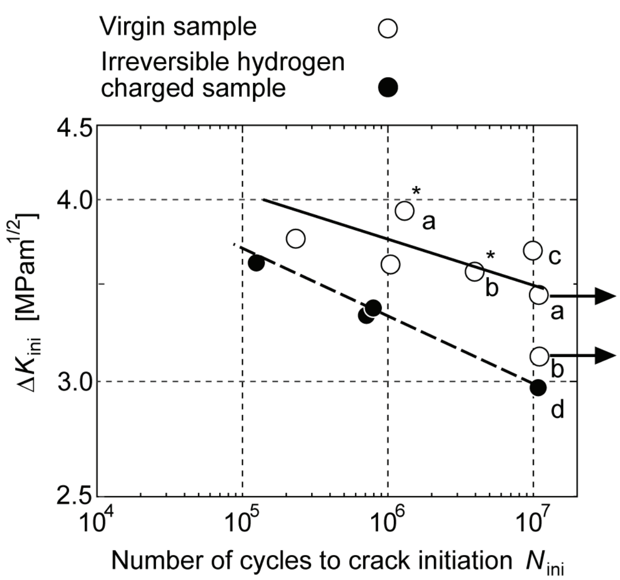


Fig. 8 Influence of irreversible hydrogen on number of cycles to crack initiation from internal inclusion. When the crack was not initiated at low level stress, the arrows are added to the data point. The mark "*" indicates that the crack was initated at high level stress for the first time.

Based on these assumptions, the fatigue crack initiation life in the smooth sample was obtained, and the result is showed in Fig. 8. The alphabets in this figure correspond to Fig. 5. In the case of low-high two-step fatigue test, N_{ini} was evaluated assuming that two step or low-high loading does not influence the crack propagation rate. When the crack was not initiated at low level stress, the arrows are added to the data point. The mark "*" indicates that the crack was initated at high level stress for the first time. When the crack was estimated to be initiated at low level loading, the fatigue crack initiation life at low level loading was plotted. The crack initiation life shared a large part of the fatigue life, and the ratio of crack initiation life to fatigue life increased as ΔK_{ini} became small. It is clear that a decrease in the fatigue crack initiation life was caused by irreversible hydrogen. Moreover, there was a tendency that a decrease in the fatigue crack initiation life by irreversible hydrogen increases as fatigue life become longer: irreversible hydrogen remarkably influence the fatigue crack initiation compared with fatigue crack propagation, and this is the reason of a decrease in fatigue life of smooth samples by irreversible hydrogen.

4. Disccusion

As described first, hydrogen embrittlement under static loading is caused by diffusible hydrogen because hydrogen can diffuse to crack tip or stress concentration field. On the contrary, irreversible hydrogen which can not diffuse due to a high binding energy between hydrogen and its trap site does not influence the static strength. Irreversible hydrogen, however, caused a decrese in fatigue strength of smooth sample as shown in Figs. 5 and 8. This indicates that irreversible hydrogen desorbed from its trap site by cyclic loading and the desorbed hydrogen caused a decrease in fatigue strength. Irreverisible hydrogen trapped at the interface between an inclusion and base metal [18] might be related to the fatigue strength degradation. The estimated crack initiation life showed a large number of loading cycles was required for the crack initiation from internal inclusion. Further, the fatigue life decrease by irreversible hydrogen was larger when longer fatigue life. This reason is considered as follow: as the cyclic loading period to the fatigue crack initiation becomes long, more hydrogen, that is desorbed from the trap site due to cyclic loading, may diffuse to the crack initiation site and promoted the fatigue crack initiation. In the case of FIB notched sample, however, the crack was initiated in the early stage of fatigue fracture, and the fatigue crack propagation process dominated the fatigue fracture. However, the fatigue crack propagation rate was not influenced by irreversible hydrogen. Hence, irreversible hydrogen did not influence the fatigue strength of notched sample. These yielded the conclusions that irreversible hydrogen contributed to the fatigue crack initiation rather than the fatigue crack propagation.

In the irreversible hydrogen charged samples, the sharp edges that would originate in the pearlitic lamella were observed on the fracture surface near the the crack initiation site had sharp edge, as shown in Fig. 7(b). This indicates that the fatigue crack propagation mechanism might be affected by irreversible hydrogen. In the high ΔK region, on the other hand, irreversible hydrogen hardly influenced the fracture surface morphology. These results suggests that the influence of irreversible hydrogen on the fatigue crack propagation depends on ΔK or crack propagation rate even if irreversible hydrogen is desorbed from its trap site by cyclic loading. This is the result that the crack propagates before hydrogen concentrates to the crack tip sufficiently at high ΔK [9]. There is a posibility that the fatigue crack propagation rate in the irreversible hydrogen charged sample might be accelerated compared with that of virgin sample because the fracture surfaces of irreversible hydrogen charged sample showed brittle nature. However, the acceleration of the fatigue crack propagation rate by irreversible hydrogen was small considerating that the fatigue life of the notched sample was not affected by irreversible hydrogen. The fatigue crack propagation tests are required to clarify whether irreversible hydrogen influences the fatigue crack propagation rate or not.

5. Conclusions

Fatigue tests were condudted for smooth and FIB notched samples to investigate the influence of irreversible hydrogen on the fatigue strength of cold drawn eutectoid wire. A special attention was paid to the both crack initiation and propagation life, and the investigation yielded the following conclusion.

1. Irreversible hydrogen induced a decrease in the fatigue life of smooth sample. On the contrary, the fatigue life of notched sample was not affected by irreversible hydrogen.
2. The fatigue crack propagation rate was estimated by using the relationship between initial stress intensity factor range ΔK_{ini} and fatigue life N_{f}: the fatigue crack propagation rate of irreversible hydrogen charged sample was almost the same as that of a virgin sample.
3. Based on the estimated crack propagation rate, the crack initiation life from internal inclusion in the smooth sample was obtained. Firstly the crack initiation life of both hydrogen states shared a large part of the fatigue life. Secondaly, the crack initiation life decreased by irreversible hydrogen. A decrease in fatigue crack initiation life by irreversible hydrogen increases as fatigue life become longer.
4. Irreversible hydrogen contributed to the fatigue crack initiation rather than to the fatigue crack propagation.
5. In the irreversible hydrogen charged sample, fracture surface near the crack initiation site exhibited brittle fracture, whereas the crack propagation region at high ΔK was ductile fatigue fracture surface similar to that of virgin sample. These indicated that irreversible hydrogen might influence the fatigue crack propagation behavior.Further invesitigations are required to clarify the influences of irreversible hydrogen on the fatigue crack propagation.

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