Development of four-point bending fatigue test method using continuously hydrogen-charging pipe specimen

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Abstract. To evaluate hydrogen embrittlement, the following two types of testing method are available: (i) testing in high-pressure hydrogen gas environment and (ii) testing in ambient air using hydrogen precharged specimen. Testing in high-pressure hydrogen gas environment is technically difficult and expensive because high-pressure gas equipments, such as high-pressure vessel and pipe, have to be installed in the laboratory. On the other hand, in the case of precharging method, outgassing of hydrogen from the specimen occurs during the test. Therefore, hydrogen embrittlement can hardly be evaluated properly, especially, in long-term testing such as high cycle fatigue test at low frequency. In this study, to effectively evaluate the hydrogen embrittlement in fatigue, an experimental method, which was the four-point bending fatigue test system with a mechanism of internal circulation of hydrogen-charging solution in a pipe specimen, was developed. By using this method, the fatigue crack growth properties in the presence of hydrogen were investigated at frequencies of 0.05 Hz and 1 Hz.

1. Introduction

Hydrogen attracts attention as clean energy which does not emit carbon dioxide and air pollutant and the practical use of hydrogen energy is being proceeded in USA, Europe, Japan and so on. A lot of effort has been put on the development of, in particular, hydrogen fuel cell vehicle (HFCV) and hydrogen station. However, in order to ensure the safety of these hydrogen equipments, hydrogen embrittlement: degradation of strength properties of metallic material due to hydrogen penetration into the material, needs to be clarified.

To evaluate hydrogen embrittlement, the following two types of testing method are commonly used: (i) testing in high-pressure hydrogen gas environment and (ii) testing in ambient air using hydrogen precharged specimen. The behavior of materials used in hydrogen equipments can be simulated by the testing in high-pressure hydrogen gas. In recent research [1, 2], the testing in high-pressure hydrogen gas demonstrated that fatigue crack growth properties were affected by hydrogen pressure, test frequency, test temperature, etc. However, this testing method is technically difficult and
expensive because high-pressure gas equipments such as a high-pressure vessel and a compressor must be installed in a laboratory satisfying sufficient safety and in addition, the installation cost is of the order of hundred million yen in Japan. On the other hand, the precharging method is easy to handle and inexpensive, while it has a disadvantage that outgassing of hydrogen from the specimen occurs during the test. Figure 1 shows the time variation of hydrogen outgassing from hydrogen precharged carbon steels. Hydrogen content of precharged carbon steels decreased to almost the same level as that of uncharged material during the first 200 h after hydrogen precharging. Thus, hydrogen embrittlement can hardly be evaluated properly, especially in long-term testing such as high cycle fatigue test at low frequency. Investigation of the hydrogen embrittlement in fatigue is essential for assurance of long-term integrity of hydrogen equipment, and therefore, it is important to establish an easy yet effective evaluation method for hydrogen embrittlement in fatigue.

In this study, to effectively evaluate hydrogen embrittlement, an experimental method was developed, which was the four-point bending fatigue test system with a mechanism of circulation of hydrogen-charging solution in a pipe specimen.

![Figure 1. Relationship between hydrogen content and time after hydrogen-charging.](image)

2. Experimental method

2.1 A developed testing setup
Figure 2 shows the concept of testing method developed in this study. The fatigue crack growth test was conducted by the four-point bending fatigue test system with a mechanism of internal circulation of hydrogen-charging solution in a pipe specimen. The hydrogen-charging solution (20 mass% NH$_4$SCN) was pumped into the pipe specimen through a flexible tube. Two screw holes were introduced on both ends of the specimen, which was connected to a flexible tube by a screw type fitting.

There are the following advantages by employing the four-point bending fatigue test system. The screw holes of both ends of specimen are not subjected to bending moment because the bending moment is zero at the outer side of load points, as indicated by the bending moment diagram shown in Fig. 3. In addition, the through-hole was present on the neutral plane where the bending stress is zero, see Fig. 4. Therefore, the unintentional fracture from the screw holes or the through-hole is avoidable in spite of the direct exposure to hydrogen-charging solution. This specimen is hereafter called the H-charging specimen, while the specimen tested without hydrogen-charging is called the Non-charging specimen.
2.2 Material and specimen

An annealed carbon steel (JIS-S35C) was used in this study. The Vickers hardness of this material is approximately 156. The microstructure is of ferrite-pearlite with an elongated texture in the rolling direction, as shown in Fig. 5.

Figure 6 shows the geometry of pipe specimen. The specimen surface was polished with a #2000 emery paper and subsequently buff-polished using alumina paste with a particle diameter of 1 μm. After polishing, three drilled holes shown in Fig. 7 were introduced on the specimen surface as a crack starter. Then, stress relief annealing in vacuum at 600 °C for 1h was conducted.
2.3 Fatigue crack growth test

Fatigue crack growth tests were carried out in the four-point bending fatigue using H-charging specimens in air at room temperature. The stress ratio was 0.1. The test frequency was 0.05 Hz or 1 Hz. The maximum bending stress at specimen surface was controlled to be constant, so that the stress intensity factor (SIF) range increased with crack growth. The crack growth behavior was observed by an optical microscope at the appointed numbers of cycles to measure the crack length. The fracture surface was observed by a scanning electron microscope.

3. Results and discussion

3.1 Effects of continuous hydrogen-charging on the fatigue crack growth rate

The fatigue crack growth test were conducted under the both conditions of continuous hydrogen-charging and of no hydrogen-charging at a stress ratio of $R = 0.1$ and at a test frequency of $f = 1$ Hz. The initial SIF range was about $5.5 \text{ MPa} \cdot \text{m}^{1/2}$ and the SIF range increased up to about $11 \text{ MPa} \cdot \text{m}^{1/2}$ with crack growth.

Figure 8 shows the relationship between the crack growth rate, $da/dN$, and the SIF range, $\Delta K$. At the relatively low $\Delta K$ regime ($\Delta K < 7 \text{ MPa} \cdot \text{m}^{1/2}$), the crack growth was unstable and the crack growth rate has a large scatter. It appears that there was no noticeable acceleration of crack growth by hydrogen in this low $\Delta K$ regime. At the relatively high $\Delta K$ regime ($\Delta K > 7 \text{ MPa} \cdot \text{m}^{1/2}$), however, the crack growth rate for H-charging specimen was higher than that for Non-charging specimen and the difference in crack growth rate increased with increase in $\Delta K$. As mentioned in the introduction, there are already two conventional methods, in which the test is performed in hydrogen gas or using a hydrogen-precharged specimen. For comparison, in Fig. 8, the present test results are shown together with the conventional test results obtained using CT specimens [10]. Although these results cannot be simply compared owing to the diversity of testing conditions including material, specimen geometry, test frequency, hydrogen content and $\Delta K$-increasing/decreasing loading, no significant difference was found among these three methods. In addition, when the test frequency was dropped from 1 Hz to 0.05 Hz, the crack growth acceleration by hydrogen became more pronounced. At the SIF range of $\Delta K = 10 \text{ MPa} \cdot \text{m}^{1/2}$, the acceleration ratio of crack growth rate with hydrogen to that without hydrogen was 4.5 at 0.05 Hz, while it was 2.2 at 1 Hz. This frequency dependence upon hydrogen-induced crack growth acceleration is well known as a feature of hydrogen-related fracture [3-7] and it has been explained by taking the increase in hydrogen accumulation at the crack tip with decrease in test frequency into account.
Figure 8. Relationship between crack growth rate and $\Delta K$.

3.2 Fracture surface

Figure 9 (a) shows the fracture surface of Non-charging specimen tested at 1 Hz and Figs. 9 (b) and (c) show the fracture surfaces of H-charging specimens tested at 1 Hz and at 0.05 Hz, respectively. All images were taken at the SIF range of 10 MPa·m$^{1/2}$. At the SIF range of $\Delta K > 7$ MPa·m$^{1/2}$ where the crack growth rate was accelerated by continuous hydrogen-charging, flat fracture facets could be observed partially, as shown in Fig. 9 (b) and (c). These facets are seen to be attributed to intergranular cracking. This facet-type fracture was also observed in conventional testing method in some previous studies [5,7-9]. It is considered that the facet-type fracture induced by continuous hydrogen-charging would account for the acceleration of fatigue crack growth. Focusing on the frequency effect on hydrogen-induced facet formation, there is no marked difference between 1 Hz and 0.05 Hz (Fig. 9 (b) and (c)), though the crack growth acceleration due to hydrogen was more pronounced for $f = 0.05$ Hz.
3.3 Residual hydrogen content

The residual hydrogen content was measured just after fatigue test using a thermal desorption spectrometer (TDS). The testing sample was a chip cut from the region about 7 mm away from the fracture surface.

The TDS measurement demonstrated that the residual hydrogen content of the chip was about 0.24 mass ppm. Figure 10 shows the hydrogen outgassing properties of the hydrogen-precharged carbon steels (JIS-S10C, S45C) for reference. As seen in this figure, about 200 h after the hydrogen-charging, the hydrogen content of those carbon steels becomes nearly equal to that of uncharged material (nearly equal to or less than 0.1 mass ppm), whereas it is 0.30-0.50 mass ppm just after the hydrogen-charging. The fatigue test conducted in this study spent about 10 days (i.e. 240 h). Nevertheless, much more amount of hydrogen remained in the pipe specimen, compared to precharged carbon steels. It should be noted that a sufficient amount of hydrogen was being supplied into the pipe specimen by the continuous circulation of hydrogen-charging solution over an entire period of testing.

The fatigue test based on the continuous hydrogen-charging demonstrated the influence of hydrogen on the crack growth acceleration, its frequency dependence and the fracture surface morphology. Accordingly, it can be concluded that the developed testing method has an advantage over the conventional precharging method in a long-term fatigue test because of its capability for hydrogen supply over an entire period of testing.

Figure 10. Decrease in hydrogen content in ambient air of hydrogen precharged carbon steel (JIS-S10C, S45C).
4. Conclusions
A fatigue testing method was developed for investigating the effect of hydrogen on the material property, which utilized continuous circulation of hydrogen-charging solution in a pipe specimen. The conclusions can be summarized as follows:

1. The acceleration of fatigue crack growth rate occurred by this method at the relatively high $\Delta K$ regime ($\Delta K > 7 \text{ MPa} \cdot \text{m}^{1/2}$). The acceleration was associated with flat facet formed by intergranular fracture.
2. Fatigue crack growth acceleration caused by this method became more pronounced with decrease in the test frequency from $f = 1 \text{ Hz}$ to $f = 0.05 \text{ Hz}$.
3. Sufficient amount of hydrogen was contained in a specimen after the fatigue test of a long period of 10 days.
4. For a long-term fatigue test, the testing method developed in this study has an advantage over the conventional precharging method in terms of its sufficient capability for hydrogen supply over an entire period of testing.

References
[1] Matsuoka S, Tanaka H, Homma N and Murakami Y 2011 Int. J. Fracture 168 101
[2] Marchi C S, Somerday B P, Nibur K A, Stalheim D G, Boggess T and Jansto S 2010 Proc. Asme Pressure Vessels and Piping Con 6 939
[3] Yamabe J, Yoshikawa M, Matsunaga H and Matsuoka S 2016 Procedia Structural Integrity 2 525
[4] Murakami Y, Kanezaki T, Mine Y and Matsuoka S 2008 Metal. Mater. Trans. A 39 1327
[5] Macadre A, Artamonov M, Matsuoka S and Furtado J 2011 Eng. Frac. Mech. 78 3196
[6] Vergani L, Colombo C, Gobbi G, Bolzoni F M and Fumagalli G 2014 Procedia Engineering 74 468
[7] Tanaka H, Homma N, Matsuoka S and Murakami Y 2007 Trans. Jpn. Soc. Mech. Eng. 73 1358
[8] Somerday B P, Sofronis P, Nibur K A, San Marchi C and Kirchheim R 2013 Acta Mater. 61 6153
[9] Sun Z, Benoit G, Moriconi C, Hamon F, Halm D, Hamon F and Henaff G 2011 Int. J. Hydrogen Energy 36 8641
[10] Takeuchi E, Furuya Y, Hirukawa S, Matsuo T and Matsuoka S 2013 Trans.Jpn. Soc. Mech. Eng. 79 1030