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Capability evaluation of continuous hydrogen-charging method for long-term fatigue test

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ABSTRACT

In hydrogen-precharging method, hydrogen will outgas from the specimen during the test, and thereby, the content and effect of hydrogen decreases with time. This decrease in hydrogen content may be permissible for a short-term test such as a tensile test and a low-cycle fatigue test at high-frequency, while it is a fatal problem for a long-term test such as a high-cycle fatigue test at low frequency. To solve this problem, in the previous study, we have developed a novel fatigue testing method, in which a four-point bending fatigue test was performed with circulation of a hydrogen-charging solution in a pipe specimen over an entire period of the test.

In this study, the four-point bending fatigue tests were carried out for a carbon steel (JIS-S35C) (i) using a pipe specimen with continuous hydrogen-charging and (ii) using a hydrogen-precharged specimen. After crack initiation, the fatigue crack growth tests were conducted under a constant stress intensity factor range of 15 $\mathrm{MPa\cdot m^{1/2}}$ by properly decreasing the stress amplitude. The time variations of total hydrogen content were measured both for a continuous hydrogen-charged specimen and for a hydrogen-precharged specimen. By highlighting the difference of charging method, the relationship between the crack growth rate and the hydrogen content was quantitatively discussed.

1. INTRODUCTION

Recently, the hydrogen energy attracts a great attention as a clean energy. The practical use of hydrogen equipment and infrastructure such as hydrogen fuel cell vehicle and hydrogen station are being proceeded. However, there are some problems to be solved for the realization of hydrogen society.

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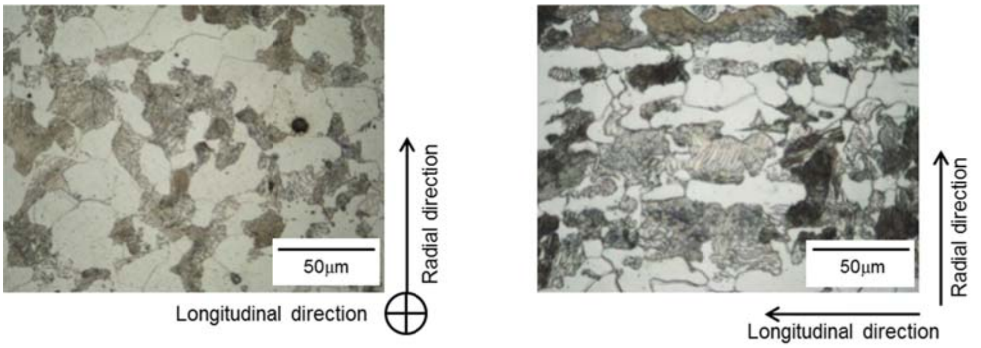
It is well known that hydrogen penetrates into a metallic material and deteriorates its strength properties. This phenomenon is called Hydrogen Embrittlement (HE). To ensure the long-term safety of hydrogen equipment, HE in fatigue needs to be correctly understood. Some components used in hydrogen equipment are subjected to a cyclic load of extremely low frequency, thus the fatigue property at low frequency in the presence of hydrogen is especially a very important issue. However, it is difficult to tackle this issue by the conventional evaluation method of HE, because the conventional method, i.e. testing in air using a hydrogen-precharged specimen (so-called hydrogen precharging method), has a fatal disadvantage, in which hydrogen outgassing from a specimen occurs during the test. In other words, in hydrogen precharging method, the hydrogen effect is evaluated under the temporal decrease in hydrogen content inside a specimen. In short-term testing such as tensile test and low-cycle fatigue test, the effect of temporal decrease in hydrogen content can be regarded as negligible, and thus some researchers have investigated the hydrogen effect on the tensile properties (Matsuoka 2006, Matsunaga 2014) and low-cycle fatigue properties (Murakami 2010) using the hydrogen precharging method. However, the temporal decrease in hydrogen is unclear (probably considerable) in the case of long-term testing such as fatigue test at low frequency, and thereby, the hydrogen effect might not be evaluated properly. To solve this problem, in our previous study, we have developed a new easy yet effective testing method for investigating the HE in long-term fatigue (Yoshimoto 2017). The newly developed testing method is four-point bending fatigue test of a pipe specimen with internal circulation of hydrogen-charging solution. The fatigue crack growth test of common carbon steel using this method demonstrated that the crack growth was accelerated by the continuous circulation of hydrogen-charging solution (Yoshimoto 2017) and the acceleration became pronounced with decrease in test frequency (Yoshimoto 2017). In addition, a certain amount of hydrogen is contained in the specimen even after the 10-day-long fatigue test. Based on these results, hydrogen is expected to be supplied continuously to a specimen over an entire period of testing by continuous circulation of charging solution, but the change in hydrogen content over time has not been measured. In order to verify the effectiveness of the new method, it is necessary to clarify the time variation of hydrogen content both in hydrogen precharging method and in continuous hydrogen charging and their effect on the fatigue properties.

In this study, the fatigue crack growth tests were carried out both for a pipe specimen with continuous hydrogen charging and for a hydrogen precharged pipe specimen. The relationship between time variation of hydrogen content and crack growth rate was investigated.

2. EXPERIMENTAL METHOD

2.1 Material and Specimen

An annealed carbon steel (JIS-S35C) was used in this study. The Vickers hardness of this material is approximately 156. Figure 1 shows the microstructure, which is of ferrite-pearlite with an elongated texture in the rolling direction. Figure 2 shows the shape and dimensions 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 mm. After polishing, three drilled holes shown in Fig.3 were introduced on the specimen surface as a crack starter. Then, stress relief annealing in vacuum at 600 °C for 1 h was conducted.



(a) Transverse sectional view (b) Longitudinal sectional view

Fig. 1 Microstructure of S35C.

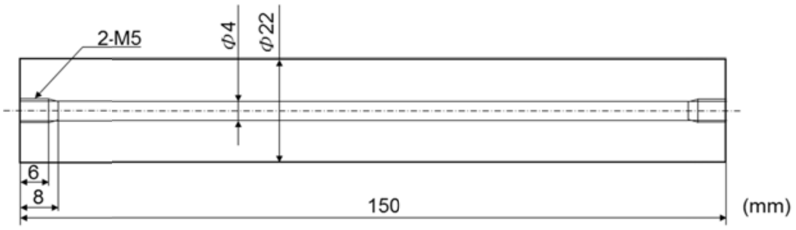


Fig. 2 Dimensions of a pipe specimen.

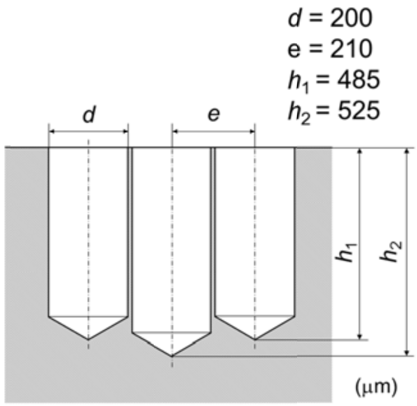


Fig. 3 Dimensions of three drilled holes.

2.2 Hydrogen Charging

Two types of method were conducted for hydrogen-charging of pipe specimen: the continuous hydrogen-charging (CHC) method, developed in the previous study, and the conventional hydrogen-precharging (HPC) method. The continuous hydrogen-charging was conducted by internal circulation of the 20 mass% of aqueous solution of NH_4SCN inside a pipe specimen using a pump and flexible tubes. The circulation of charging solution was started 96 h prior to the test to attain the steady state of hydrogen distribution inside the specimen. A hydrogen-precharged specimen was prepared by soaking in the hydrogen-charging solution for 168 h. This specimen is hereafter called *HPC specimen*, while the specimen with continuous hydrogen-charging is called *CHC specimen*.

2.3 Fatigue Crack Growth Test

Fatigue crack growth tests by four-point bending fatigue were carried out in air at room temperature for both a HPC specimen and a CHC specimen. While precracking, the stress intensity factor (SIF) range, ΔK was increased up to 15 MPa・$m^{1/2}$ under the constant stress amplitude. Then, the ΔK-constant (ΔK = 15 MPa・$m^{1/2}$) tests were conducted at a frequency of 0.002 Hz and a stress ratio of 0.1. The test frequency was determined so that the test takes long to a certain extent, in order to investigate the effect of time variation of hydrogen content on the crack growth.

2.4 Hydrogen Content Measurement

The time variation of hydrogen content during the fatigue test was measured by the following way using a thermal desorption analyzer (TDA). Regarding HPC specimen, a pipe specimen was hydrogen-charged by soaking in charging solution for 168 h. Disk-shaped samples with a thickness of 2 mm were cut from the specimen at a given intervals, and then their hydrogen contents were measured by TDA. Regarding CHC specimen, on the other hand, continuous hydrogen-charging was conducted for the multiple pipe specimens for a given duration and the hydrogen contents of disk samples cut from these CHC specimens were measured. In TDA measurement, the temperature was increased up to 300 °C at a heating rate of 100 °C/h.

3. RESULTS AND DISCUSSION

3.1 Time variation of hydrogen content

Figure 4 shows the time variations of hydrogen content both for CHC specimen and for HPC specimen. The hydrogen content in the HPC specimen showed the maximum value just after hydrogen charging. Then it gradually decreased with time, and reached the almost same level as uncharged material 500 h after hydrogen charging. In contrast, in CHC specimen, the hydrogen content inside the specimen started to increase just after beginning of charging, and it was saturated after about 100

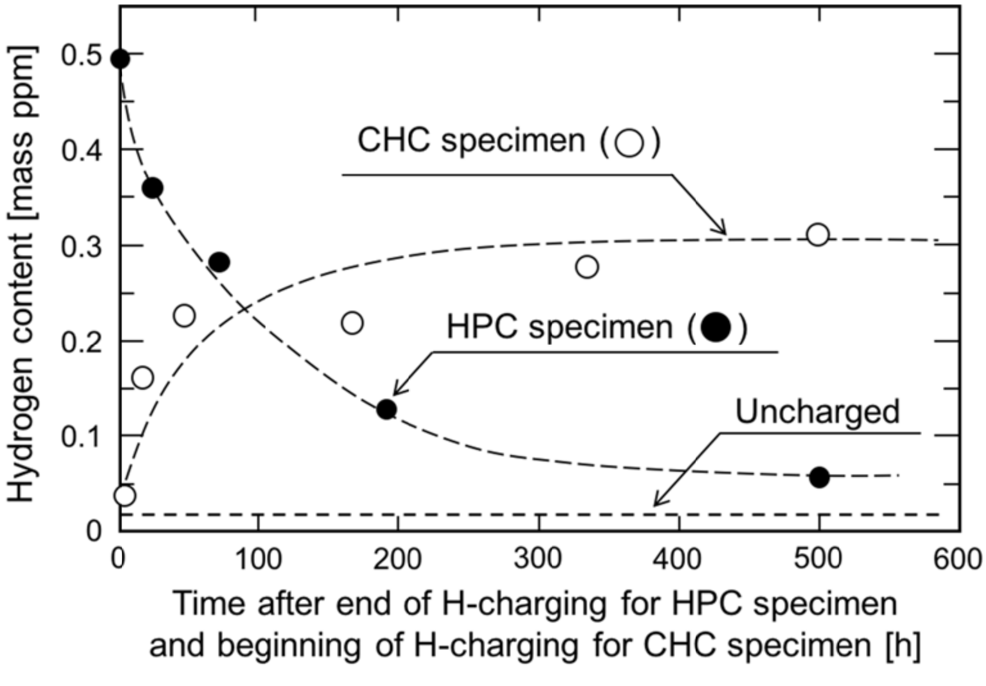


Fig. 4 Time variations of hydrogen content for HPC specimen and CHC specimen.

h. This means that the amount of hydrogen supply to a specimen became balanced with that of hydrogen outgassing from a specimen. In addition, it should be noted that the saturated hydrogen content of CHC specimen was lower than that of HPC specimen. This difference can be explained by taking the hydrogen distribution inside the specimen into account. Namely,

the hydrogen distribution of HPC specimen is uniform just after hydrogen charging, whereas the hydrogen distribution has a gradient in CHC specimen. In other words, in CHC specimen, the local hydrogen concentration is maximum at the inner surface, which is considered to be equivalent to the saturated hydrogen content of HPC specimen, and the local concentration decreases toward the outer surface. As a result, the total hydrogen content of CHC specimen is somewhat lower than that of HPC specimen.

3.2 The effect of time variation of hydrogen content on the crack growth rate

Fatigue crack growth tests were conducted at ΔK of 15 MPa・m$^{1/2}$ both for HPC specimen and for CHC specimen. Figure 5 shows the relationship between crack growth rate and time. The crack growth rate of HPC specimen was also decreased with time. In HPC specimen, the crack growth rate correlates significantly with the hydrogen content of specimen. The acceleration ratio became almost 1 (no acceleration), when the hydrogen content reached to the almost same level as uncharged material. Based on these results, it can be concluded that the degree of hydrogen embrittlement is affected considerably by the decrease in hydrogen content with time (i.e. it declines over time) and thereby, the hydrogen embrittlement is not evaluated properly using a hydrogen-precharged specimen in the case of long-term fatigue test.

On the other hand, the crack growth rate of CHC specimen was kept constant 120 h to 220 h after the beginning of hydrogen charging. In this period, the hydrogen content of

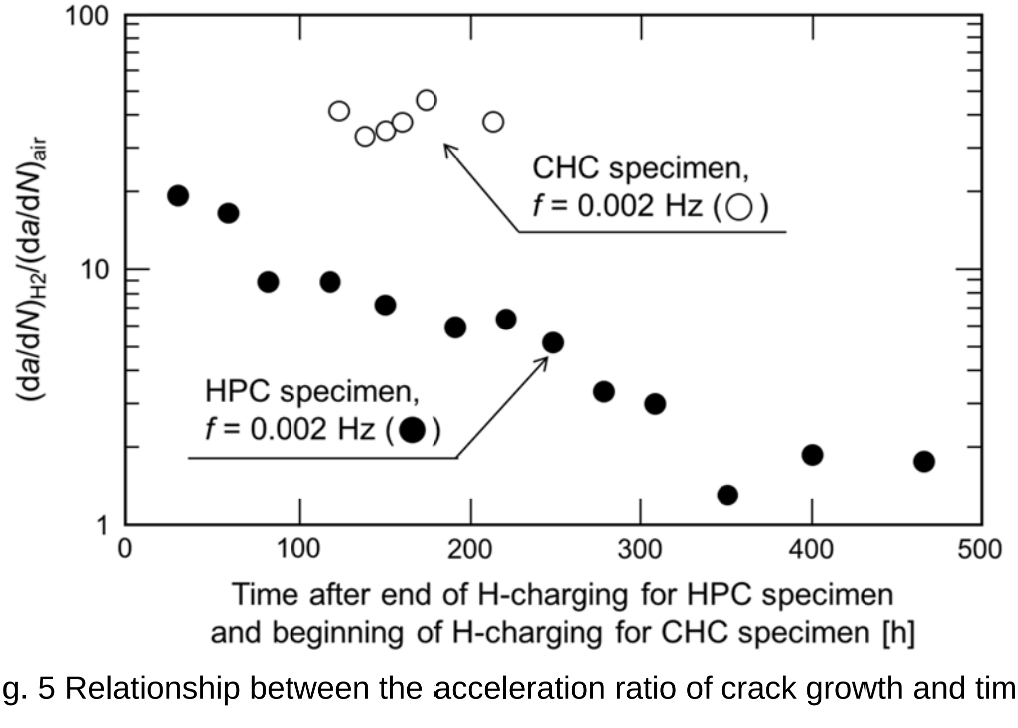


Fig. 5 Relationship between the acceleration ratio of crack growth and time.

CHC specimen was almost constant, as shown Fig. 4. In this continuous hydrogen charging method, therefore, once the hydrogen content is saturated (i.e. the hydrogen inflow and outflow are balanced), it is possible to evaluate the hydrogen embrittlement under the constant condition even in long-term test. In this point, the continuous hydrogen charging method we developed has an advantage over the conventional hydrogen precharging method.

The saturated hydrogen content of CHC specimen was lower than that of HPC specimen, as mentioned above. Nevertheless, the acceleration ratio is larger for CHC specimen than HPC specimen. Accordingly, it is considered that the degree of hydrogen effect of CHC specimen depends on the local hydrogen concentration rather than on the average hydrogen content, because the hydrogen distribution of CHC specimen is not uniform. Therefore, the effect of hydrogen distribution on the hydrogen embrittlement is another important issue for verification of the continuous hydrogen charging method, and this issue will be investigated in near future.

4. CONCLUSIONS

The four-point bending fatigue tests were carried out at constant ΔK of 15 MPa・$m^{1/2}$ for a carbon steel (JIS-S35C) (i) using a pipe specimen with continuous hydrogen-charging and (ii) using a hydrogen-precharged specimen. The following results were obtained:

(1) The hydrogen content of hydrogen-precharged (HPC) specimen decreased with time and it reached to almost same level as uncharged specimen about 500 h after hydrogen-charging. On the other hand, in continuous hydrogen-charging (CHC) specimen, the hydrogen content kept constant after the inflow and outflow of hydrogen were balanced.
(2) The fatigue crack growth acceleration due to hydrogen correlates with hydrogen content. Thus, the acceleration ratio of HPC specimen decreased with decrease in hydrogen content, whereas that of CHC specimen kept constant over an entire period of testing.
(3) From these experimental results, it can be concluded that the newly developed method of continuous hydrogen-charging is more effective to long-term fatigue test in the presence of hydrogen than conventional hydrogen-precharging method.

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