The decrease of tensile strength for the notched specimens in the hydrogen gas

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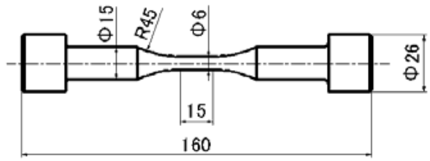
Abstract. We investigated the effect of hydrogen gas environment to 1300MPa tensile strength of chromium-molybdenum steel (JIS SCM435) with a sharp notched specimen. Tensile tests were carried out in hydrogen gas and in helium gas environments. In hydrogen gas environment, the specimen which was not exposed to hydrogen gas and the specimen which was exposed to hydrogen gas for 48h were used. A thermal desorption spectrometry (TDS) analysis was conducted, in order to decide the exposure time to hydrogen gas. As the result of tensile tests, the notch tensile strengths in hydrogen gas exposed specimens was stronger than that of in hydrogen gas non-exposed specimens. We also investigated the crack initiation of notched specimens in hydrogen gas and found that the crack initiation load at the hydrogen gas non-exposed specimen was lower than that of the exposed specimen.

Introduction

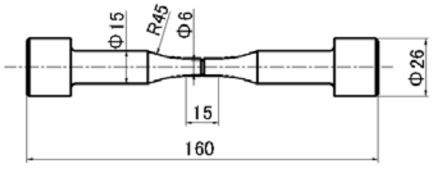
Recently, hydrogen energy is expected a next energy of fossil fuels and several products which use hydrogen energy are developed. For example, development of fuel cell vehicle and hydrogen automobile are advanced, and many parts of these products and infrastructure installation of hydrogen supply; pipe lying and storage tank are manufactured from a variety of steels. Steels cause hydrogen embrittlement, especially, high strength steels exhibit high hydrogen embrittlement susceptibility [1-2]. Although many researches have been studied to understand hydrogen embrittlement [3-5], it is not yet satisfactorily understood. The purpose of the present study is to investigate effect of hydrogen gas environment to 1300MPa tensile strength of SCM435 with a sharp notched specimen, especially considering the hydrogen exposure time.

Specimens and experimental procedure

Specimens. Quenched and tempered SCM435 was used in the present study with the chemical composition listed in Table 1. The circumferentially notched round bar specimens with a notch root radius of ρ=0.03mm, and minimum cross-section diameter of d=6mm and various depth of notch of t=0(smooth specimen), 0.02, 0.1, 0.2, 0.5, and 1.0mm are shown in Fig. 1. The surface of smooth specimen and the notch root of notched specimen were polished by alumina powder (powder size: 0.05μm) to avoid the effect of machined flaw or surface roughness.



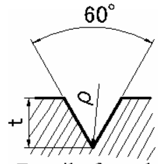
(a) Smooth specimen.



(b) Notched specimen.

Table1 Chemical composition (wt%)

C	Si	Mn	P	S	Ni	Cr	Mo	Cu
0.38	0.22	0.84	0.024	0.021	0.08	1.11	0.15	0.12



Detail of notch

ρ =0.03mm

t =0.02, 0.1, 0.2, 0.5, 1.0mm

Fig. 1 Specimen configuration.

Experimental procedure. Tensile tests were carried out at a cross head speed was 5.0×10^{-4}mm/s and in hydrogen gas and helium gas environments with gas pressure 0.6MPa at room temperature. In hydrogen gas environment, a specimen which was not exposed to hydrogen gas and a specimen which was exposed to hydrogen gas for 48h were used. Exposure condition was gas pressure 0.6MPa at room temperature. Tensile tests in helium gas and hydrogen gas non-exposed specimen were started right after filling a gas. Tensile tests in hydrogen gas exposed specimen were exposed to hydrogen gas for 48h before starting the tests. On later, describe a specimen tested in helium gas environment as ‘helium gas specimen’, a specimen which is not exposed to hydrogen gas environment as ‘hydrogen gas non-exposed specimen’ and a specimen which is exposed to hydrogen gas environment for 48h as ‘hydrogen gas exposed specimen’.

Experimental results

Hydrogen contents. To decide the exposure time to hydrogen gas, a thermal desorption spectrometry (TDS) analysis was conducted. The condition of hydrogen gas exposure was to saturate hydrogen in a specimen.

Theoretical curve of Hydrogen contents. Consider an infinite cylinder of radius r_0, the rate of C/C_0; C is the concentration at the point r at time t and C_0 is the concentration at the point of cylinder surface ($r= r_0$) of the cylindrical ordinate, is derived from Fick’s equation at constant temperature [6].

$$C/C_0 = 1 - 2\sum_{n=1}^{\infty}\frac{\exp(\frac{-D\beta_n^2 t}{r_0^2})J_0(\frac{\beta_n r}{r_0})}{\beta_n J_1(\beta_n)} \qquad (1)$$

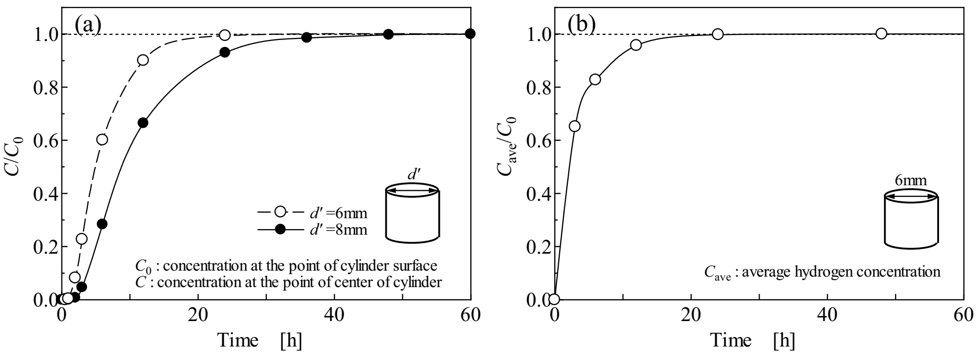
Where D is the diffusion coefficient under experimental conditions (D=1.0×10^{-10} for SCM435 [7]), J_0, J_1 are Bessel coefficients of zero and first order, β_n is a root of $J_0(\beta)$=0. From the Eq. 1, the rate of C_{ave}/C_0; C_{ave} is the average hydrogen concentration in an infinite cylinder at time t, is follows,

$$C_{ave}/C_0 = 1 - 4\sum_{n=1}^{\infty}\frac{\exp(\frac{-D\beta_n^2 t}{r_0^2})}{\beta_n^{\ 2}} \qquad (2)$$

The theoretical curves of hydrogen concentration at center of infinite cylinder with diameter d'=6mm and d'=8mm were derived from Eq. 1 are shown in Fig. 2. These cylinder's size were decided from the size of minimum cross-section diameter (6mm) and maximum cross-section diameter (8mm) for tensile tests specimens. Hydrogen diffusion time is adequate for 30-40h at the center of cylinder according to Fig. 2(a). The theoretical curve of average hydrogen concentration of infinite cylinder with diameter d'=6mm derived from Eq. 2 is shown in Fig. 2(b). Data obtained from TDS analysis are average hydrogen content in measurement sample, so the results of TDS analysis are effective by compared with Fig. 2(b).

Results of TDS analysis. Hydrogen exposure and TDS analysis were conducted with referring the theoretical exposure time obtained in previous chapter. The exposure condition was gas pressure 0.6MPa at room temperature and the exposures were used specimens with diameter of d=6mm, length of L=30mm.Exposure time were 0, 6, 24, 48, and 96h. After exposed, the specimens were cut off for the measurement samples with thickness 2.0mm and measured by TDS, where the measurement sample's thickness was shorter than the exposure tests specimen's length sufficiently, hydrogen diffusion condition of the measurement samples was same as that of an infinite cylinder.

The results of TDS analysis are shown in Fig. 3.As a result, the exposure time for 48h was adopted as the tensile test condition by comparing the results of TDS analysis and theoretical curve.



(a) Hydrogen concentration at center of infinite cylinder of diameter d'=6 and 8mm.

(b) Hydrogen concentration of infinite cylinder of diameter d'=6mm.

Fig. 2 Theoretical curves of hydrogen concentration.

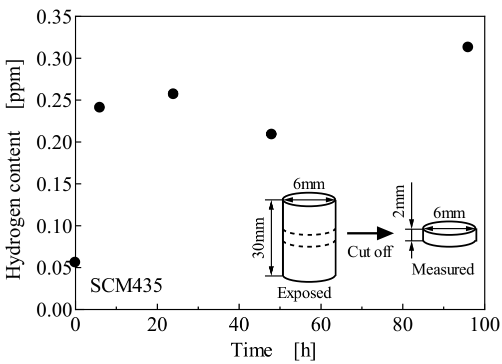
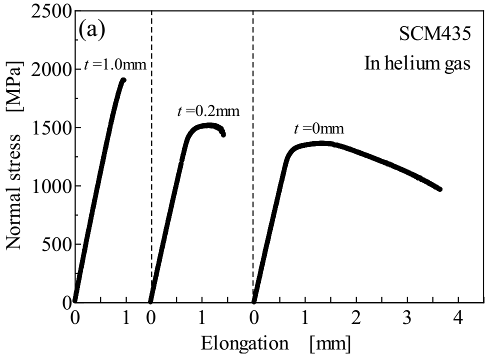
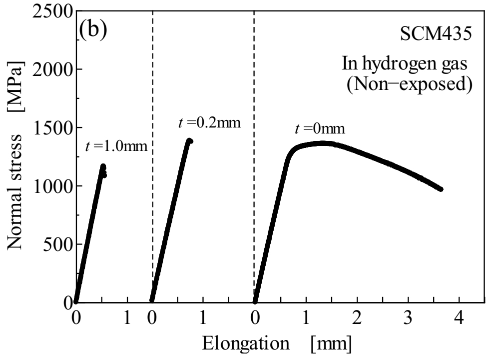


Fig. 3 Relationship between hydrogen content and exposed time.

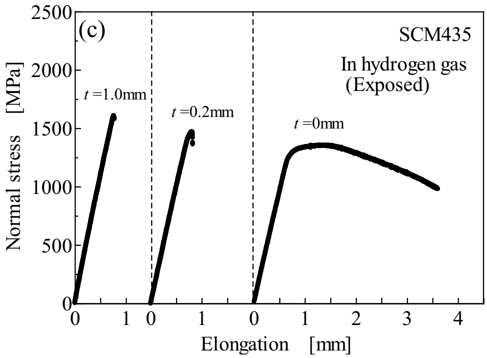
Results of tensile tests. Stress-elongation curves of tensile tests of helium gas specimens and hydrogen gas exposed specimens and non-exposed specimens are shown in Fig. 4(a), (b) and (c), and relationship between tensile strength and depth of notch is shown in Fig. 5. The difference of tensile strength with smooth specimens between helium gas specimen and hydrogen gas specimen is not appeared. In contrast, the difference of notch tensile strength is appeared, the strongest is a helium gas specimen, the second is a hydrogen gas exposed specimen and the third is a hydrogen gas



(a) In helium gas.



(b) In hydrogen gas (non-exposed).



(c) In hydrogen gas (exposed).

Fig. 4 Relationship between nominal stress and elongation.

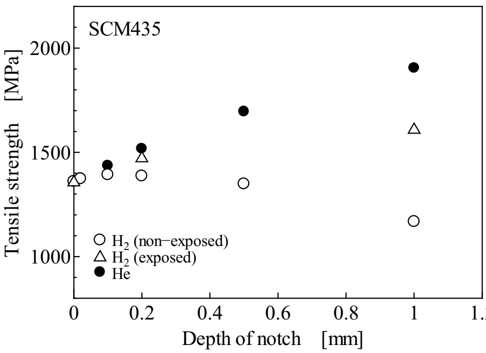
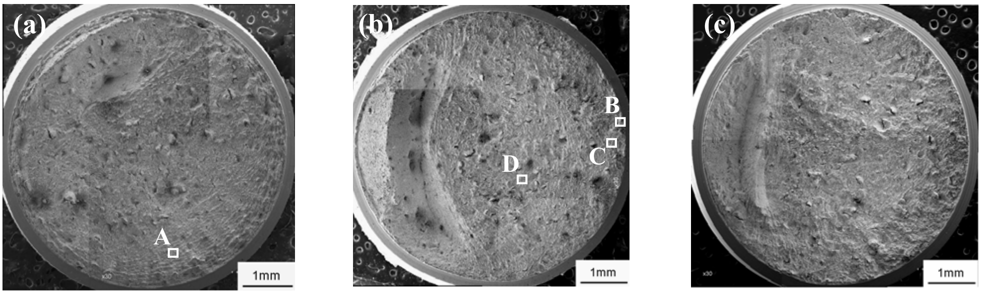


Fig. 5 Relationship between tensile strength and depth of notch.

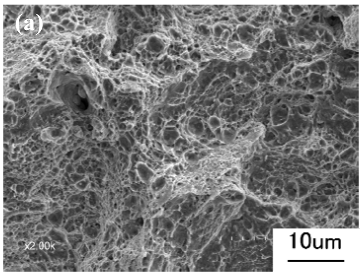
non-exposed specimen at each depth of notch.

Fractography. The fracture surfaces were observed on the scanning electron microscope (SEM). The fracture surfaces of notched specimens whose depth of notch is 0.2mm are shown in Fig. 6. The helium gas specimens exhibited a fracture mode of microvoid coalescence in all part of surfaces (Fig. 7(a)). In contrast the hydrogen gas specimens exhibited an area which seemed to be ductile fracture in the very vicinity of notch root (Fig. 7(b)), an intergranular fracture in the vicinity of the notch root (Fig. 7(c)) and a quasi-cleavage fracture inside of an intergranular fracture appearance area (Fig. 7(d)).

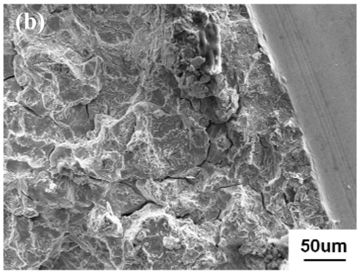


(a) In helium gas. (b) In hydrogen gas (non-exposed). (c) In hydrogen gas

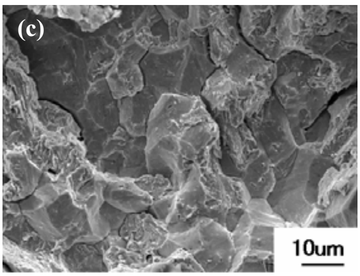
Fig. 6 Fracture surfaces of notched specimens (t=0.2mm).



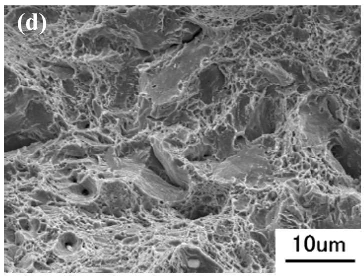
(a) Microvoid (A in Fig. 6(a))



(b) Ductile fracture (B in Fig. 6(b))

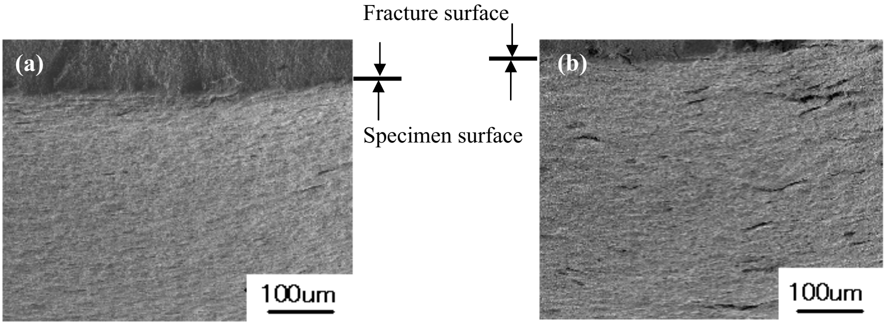


(c) Intergranular (C in Fig. 6(b))



(d) Quasi-cleavage (D in Fig. 6(b))

Fig. 7 SEM images of fracture surfaces.



(a) In hydrogen gas (non-exposed). (b) In hydrogen gas (exposed).

Fig. 8 Specimen surfaces near fracture surface.

The smooth specimens exhibited a cup and corn fracture mode and the difference between helium gas specimen and hydrogen gas specimens was not clear. In the hydrogen gas non-exposed specimen, however, the specimen surface exhibited many cracks at near the fracture surface (Fig. 8).

Discussions

Tensile strength in hydrogen gas. The reason why tensile strengths of hydrogen gas specimens decrease compared with that of helium gas specimens is thought from the fractography. The helium gas specimens had the crack initiation site in the vicinity of the notch root which indicates a ductile fracture, therefore, the fracture is caused by plastic deformation. On the other hand, the hydrogen gas specimens had the area which seemed to be ductile fracture and the intergranular cracking in the vicinity of the notch root where hydrogen concentrates by high stress field.

Difference between hydrogen gas non-exposed specimen and exposed specimen. Consider the difference of the notch tensile strength between hydrogen gas non-exposed specimen and exposed specimen. Non-exposed specimen and exposed specimen are similar at fracture surface; intergranular or quasi cleavage, so the characteristic distinguish was not appeared. It makes no difference about the fracture mechanism to exposure time. The difference of the notch tensile strength is able to explain from the cracks which were shown in surface of smooth specimen for hydrogen gas non-exposed

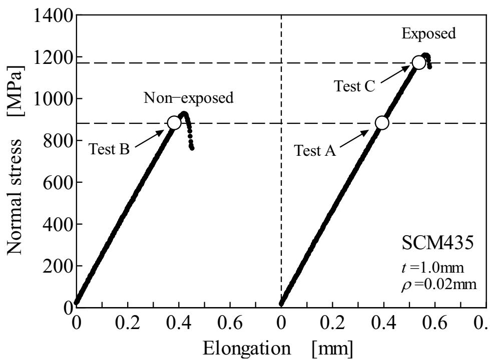


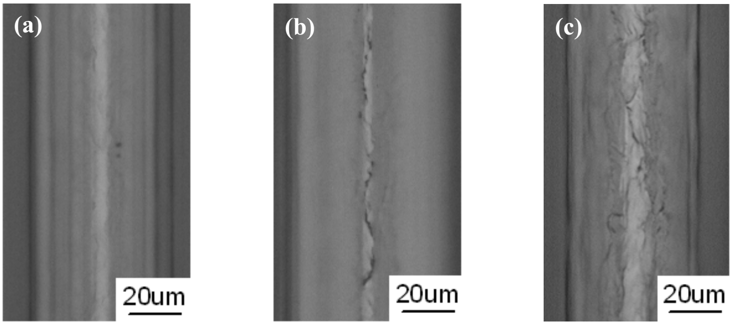
Fig. 9 Test conditions.

specimen (Fig. 8). As these cracks were exhibited in the specimen surface near the fracture surface, the cracks initiated at surface where hydrogen concentrates after necking. If the same phenomenon is occurred to the notched specimen, it is thought that the crack initiation of hydrogen gas non-exposed specimen is faster than that of hydrogen gas exposed specimen, and as a result, the tensile strength is decreased.

Specimens before fractured were observed to investigate material failure near the notch root. Failure of the hydrogen non-exposed specimen and exposed specimen were observed on the optical micro scope. The tests were conducted with the notched specimens whose depths of notch were 1.0mm at three conditions. The tests conditions are shown in Fig. 9. The one condition was the test with the hydrogen gas non-exposed specimen loading at 95% of tensile strength of the hydrogen gas non-exposed specimen (test A). The second condition was the same as test A except for using the hydrogen gas exposed specimen (test B). The third condition was the test with the hydrogen gas exposed specimen loading at 95% of tensile strength of the hydrogen gas exposed specimen (test C).

The failures at each condition are shown in Fig. 9. Compared with the failure of test A and test B, hydrogen gas non-exposed specimen (test B) exhibited several large failures. These cracks were shown in the semiperimeter of specimen. The exposed specimen exhibited a few failures. Hydrogen gas exposed specimen exhibited many comparatively small failures on the all circumferences of notch root at test C.

In order to understand this failure, test B specimen was cut at a longitudinal direction. The result is shown in Fig. 11. Result of these observations, it turns out that crack initiation at hydrogen gas non-exposed specimen is faster than hydrogen gas exposed specimen. As the fracture mechanism is seemed to be same, the difference between the non-exposed specimens and the exposed specimens is only hydrogen concentration distribution by difference of exposure time to hydrogen gas.



(a) Test A. (b) Test B. (c) Test C.

Fig. 10 Failure observed in notch root at the Fig. 9 conditions.

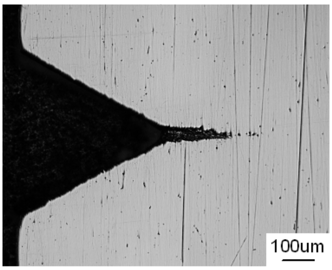


Fig. 11 Fractography of crack in longitudinal direction of test B in Fig. 10.

Conclusions.

The effect of hydrogen gas to 1300MPa tensile strength of SCM435 steel with a sharp notched specimen has been investigated by means tensile tests and fractography. The results of this study can be summarized as follows.

(1) The notch tensile strengths of hydrogen gas exposed specimens are stronger than that of hydrogen gas non-exposed specimens.
(2) The crack initiation at the hydrogen gas non-exposed specimen is faster than that of the hydrogen gas exposed specimen.
(3) It is thought that the difference of the notch tensile strength between hydrogen gas non-exposed specimen and hydrogen gas exposed specimen is caused by the difference of a hydrogen concentration distribution.

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References

[1] S. Matsuyama, in: *Delayed Fracture*, The Nikkan Kogyo Shimbun, LTD., Tokyo (1989).

[2] Y. Fukai, K. Tanaka and H. Uchida, in: *Hydrogen and Metal*, Uchida Rokakuho, Tokyo (1998).

[3] S. Matsuoka, N. Homma, H. Tanaka, Y. Fukushima and M. Yukitaka: The Japan Institute of Metals, Vol.70, No.12 (2006), pp. 1002-1011.

[4] M. Wang, E. Akiyama, K. Tsuzaki: Corrosion Science 49 (2007), pp.4081-4097.

[5] T.L. Chang, L.W. Tsay, C. Chen: Material Science and Engineering, A316 (2001) pp.153-160.

[6] A. Demaret, A. G. Hock, F. A. Meunier: Acta Metallurgia, Vol.2, (1954) 3 pp214.

[7] J. Nagata, N. Guy and Y. Murakami: Journal of the Society of Materials Science, Japan, Vol.54 No.12 (2005) pp1217-1224.