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The effect of electrical hydrogen charging on the strength of 316 stainless steel

S. Sugiyama ^a, H. Ohkubo ^a, M. Takenaka ^b, K. Ohsawa ^b, M.I. Ansari ^c,
N. Tsukuda ^b, E. Kuramoto ^{b,*}

^a Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, 6-1 Kasuga-koen, Kasuga-shi, Fukuoka 816-8580, Japan

^b Research Institute for Applied Mechanics, Kyushu University, 6-1 Kasuga-koen, Kasuga-shi, Fukuoka 816-8580, Japan

^c Department of Materials Science, Bahauddin Zakariya University, Multan 60800, Pakistan

Abstract

The effect of electrical hydrogen charging on the strength of 316 stainless steel specimens has been investigated in tensile tests at 223 K, and the increase of yield stress and the decrease of total elongation were observed. These tendencies increase with increasing hydrogen content of the specimens. This is considered to be due to hydrogen-induced phase transformation from γ (fcc) to ϵ (bcc), α (bcc), which was confirmed by X-ray diffraction method. Hydrogen concentration was determined by elastic recoil detection (ERD) method, the maximum of which reached 40% near the surface region. Positron annihilation lifetime was also measured after electrical hydrogen charging and a longer lifetime of about 300 ps was observed, which suggests the formation of microvoids in the specimens. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

In plasma-facing materials, the increase of hydrogen concentration in the matrix causes a severe change in the material properties, i.e. phase transformation, hydride formation, defect formation and so on. Especially in the austenitic stainless steels, it is well known that phase transformation from γ (fcc) to ϵ (hcp), α (bcc) occurs by hydrogen charging and many investigations have so far been performed [1–4]. Furthermore, it has been recently recognized that the increase in hydrogen concentration might induce defects like vacancies into the specimens [5,6]. These are very important phenomena from a fundamental point of view and must be investigated in detail, and the effect of hydrogen charging on the strength of materials must also be studied in order to give a firm basis for the development of plasma-facing materials. In the present study, the effect of electrical hydrogen

charging on the strength of 316 stainless steel specimens was investigated accompanied by X-ray diffraction experiment, elastic recoil detection (ERD) measurement and positron annihilation lifetime measurement.

2. Experimental

Small size tensile specimens of 316 stainless steel were prepared by punching a thin sheet of 0.1-mm thickness, the actual shape of which and the element analysis are shown in Fig. 1. Tensile specimens were solution-annealed at 1050°C for 1 h. The electrical hydrogen charging was done in a solution of 1 N H₂SO₄ at various current densities. Tensile tests were done by using Instron machine in a bath of liquid alcohol cooled to 223 K by dry ice. The strain rate was $1.67 \times 10^{-4} \text{ s}^{-1}$. The reason why this bath was chosen was to avoid the escape of hydrogen atoms from the specimens. The X-ray diffraction technique was used to detect the phase transformation in specimens electrically hydrogen charged [7]. To obtain the depth distribution of hydrogen atoms in the specimen, the ERD was made for the electrically hydrogen-charged specimens by 3-MeV He ions

* Corresponding author. Tel.: +81-92 583 7766; fax: +81-92 583 7767.

E-mail address: kuramoto@himiko.riam.kyushu-u.ac.jp (E. Kuramoto).

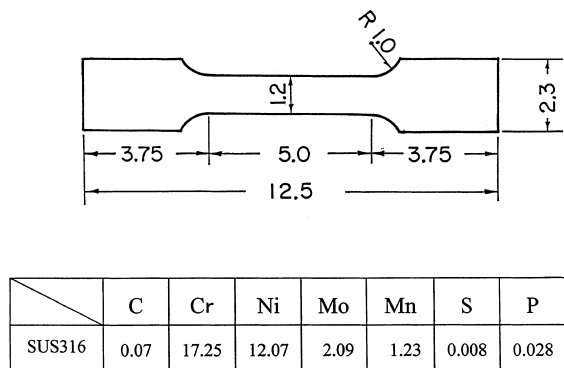


Fig. 1. Shape and size of a small tensile specimen of 316 stainless steel (unit is mm, thickness is 0.1 mm) and chemical analysis of alloying elements in 316 stainless steel (wt%).

accelerated by tandem accelerator, where the specimen was held in a vacuum chamber and recoiled hydrogen atoms were detected as a function of energy [8]. One spectrum was obtained for about half an hour. Positron annihilation lifetime measurement was made for the electrically hydrogen-charged specimens by using the fast-slow coincidence circuit with phototubes (Hamamatsu H3378) and BaF_2 scintillators [9].

3. Results and discussion

3.1. Tensile tests

In Figs. 2 and 3, the stress–strain curves of 316 stainless steels electrically hydrogen charged under

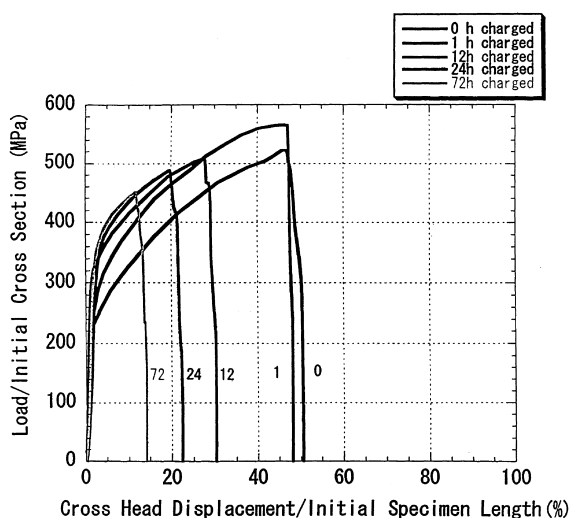


Fig. 2. Stress–strain curves of 316 stainless steels electrically hydrogen charged under 5 mA/cm^2 for various charging times (1, 12, 24, 72 h).

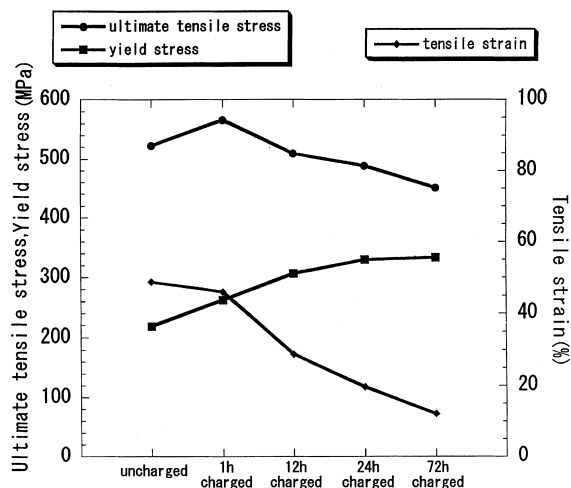


Fig. 3. Change of yield stress (0.2%), ultimate tensile stress, and tensile strain of 316 stainless steels electrically hydrogen charged under 5 mA/cm^2 for various charging times (1, 12, 24, 72 h).

5 mA/cm^2 for various charging times (1, 12, 24, 72 h) are shown. Values of the penetration depth of hydrogen atoms for these charging times can be estimated from $2(Dt)^{1/2}$, where the diffusion constant D of hydrogen in 316 stainless steel is $1 \times 10^{-11} \text{ cm}^2/\text{s}$, giving rise to 3.8, 13.2, 22.8, 32.2 μm , respectively. Hydrogen atoms diffuse into the specimen from both sides, and the thickness of the specimen is 0.1 mm, suggesting that a longer charging time provides hydrogen atoms almost all through the bulk but in the shorter charging time, the central position of the specimen is free from hydrogen atoms, at least just after the charging. All the tensile tests were done at 223 K in order to prevent hydrogen atoms from escaping by diffusion. With increasing charging time, the increase of yield stress (0.2%), and the decrease of tensile strain (total elongation) were observed. Namely, the tendency of embrittlement caused by hydrogen is seen.

In Figs. 4 and 5, the stress–strain curves of 316 stainless steels electrically hydrogen charged under 5 mA/cm^2 for various charging times (1, 12, 24, 72 h) and annealed at room temperature for 3 days are shown. All the tests were again made at 223 K. The recovery of the yield stress and the tensile strain due to 3-day annealing were observed but in the 72 h charged specimen, the recovery is not so complete, that is, the tensile strain is small compared to the uncharged specimen. In Fig. 6, the result of X-ray diffraction analysis for 316 stainless steel electrically hydrogen charged under 10 mA/cm^2 for 22 h and annealed at room temperature is shown. Just after the hydrogen charging, the γ (fcc) peak decreases and the ϵ (hcp) peak and the α (bcc) peak appear, and with increasing annealing time at room temperature, the

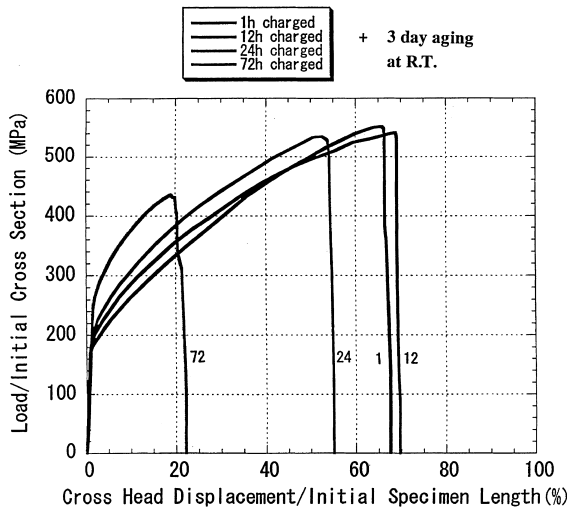


Fig. 4. Stress–strain curves of 316 stainless steels electrically hydrogen charged under 5 mA/cm² for various charging times (1, 12, 24, 72 h) and annealed at room temperature for 3 days.

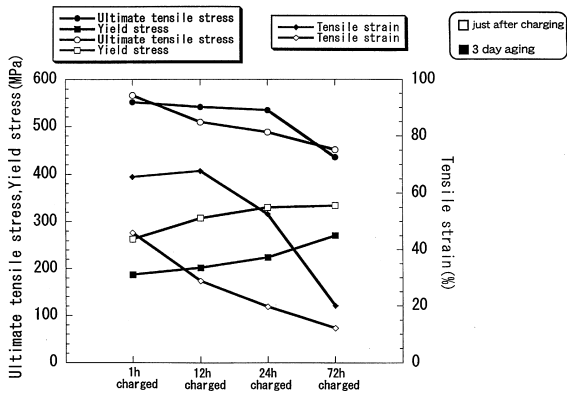


Fig. 5. Change of yield stress (0.2%), ultimate tensile stress, and tensile strain of 316 stainless steels electrically hydrogen charged under 5 mA/cm² for various charging times (1, 12, 24, 72 h) and annealed at room temperature for 3 days.

ϵ peak gradually decreases and the α peak increases with the recovery of the γ peak. The noticeable point is that the angle corresponding to the ϵ peak gradually increases, that is, the lattice constant of the ϵ -phase decreases, suggesting that the hydrogen concentration in this phase decreases. The angle corresponding to the α peak does not change so much during annealing, suggesting that the hydrogen concentration in this phase is basically not so high, probably due to low solubility of hydrogen atoms in the bcc phase. Solubility of hydrogen atoms in the fcc phase is also small, which corresponds to a slight increase of the angle in the γ peak during

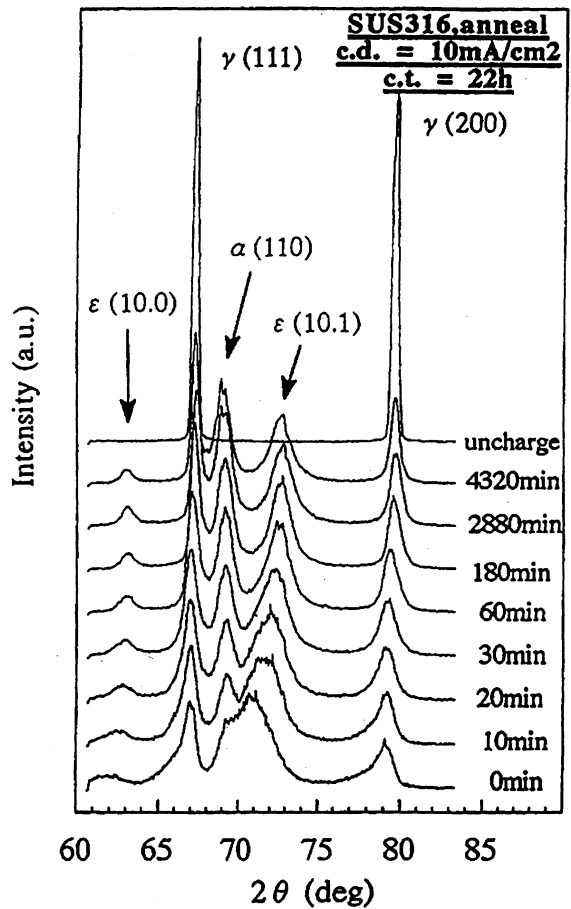


Fig. 6. Result of the X-ray diffraction analysis for a 316 stainless steel electrically hydrogen charged under 10 mA/cm² for 22 h and annealed at room temperature.

annealing. The comparison between the results of tensile tests and that of X-ray diffraction suggests that the hydrogen-induced phase transformation largely affects the mechanical property of the 316 stainless steel specimens, i.e. the increase of the yield stress and the decrease of the tensile strain. 3-day annealing at room temperature gives the clear recovery tendency both in the tensile test and the X-ray diffraction profile. During room temperature annealing, three events are taking place together, (i) decrease of the hydrogen concentration, (ii) the reverse transformation from the ϵ -phase to the γ -phase, and (iii) the transformation from the ϵ -phase to the α -phase. Events (i) and (ii) are considered to contribute to the decrease of the yield stress, i.e. recovery of the mechanical property, but the event (iii) contributes to the hardening. These three effects cooperate and finally result in the recovery of hydrogen-induced embrittlement.

3.2. ERD analysis of hydrogen

In order to obtain the concentration and the depth distribution of hydrogen atoms electrically charged in the 316 stainless steel specimens, ERD analysis was performed by using a 3-MeV He ion beam produced by the tandem accelerator. The hydrogen-charged specimen was placed on the mount in the vacuum chamber, being pumped half an hour before starting measurement and during measurement was kept in vacuum for a necessary time for measurement, again half an hour. The obtained counts were accumulated as a function of the energy of recoiled hydrogen atoms and were converted to the relation between the hydrogen concentration and the depth where hydrogen atoms exist. In Fig. 7 is shown one example of the obtained depth distribution of hydrogen atoms in the surface region of the 316 stainless steel specimen electrically hydrogen charged under the current density of 5 mA/cm² for 24 h. Before charging, hydrogen atoms are found only on the surface of the specimen which shows a rather broad distribution because of the low-depth resolution of the system, i.e. about 30 nm. After charging, very high hydrogen concentration, about 40% (H/atom ~0.4) at the peak near the surface is observed and hydrogen atoms distribute towards the central part of the specimen. After 6 h aging at room temperature, the concentration of hydrogen atoms slightly decreases but the behavior of distribution is not so much changed. The high concentration of hydrogen atoms in the 316 stainless steel specimens obtained by the ERD method is considered to be the basis of the hydrogen-induced phase transformation from γ -phase to ϵ -phase or α -phase.

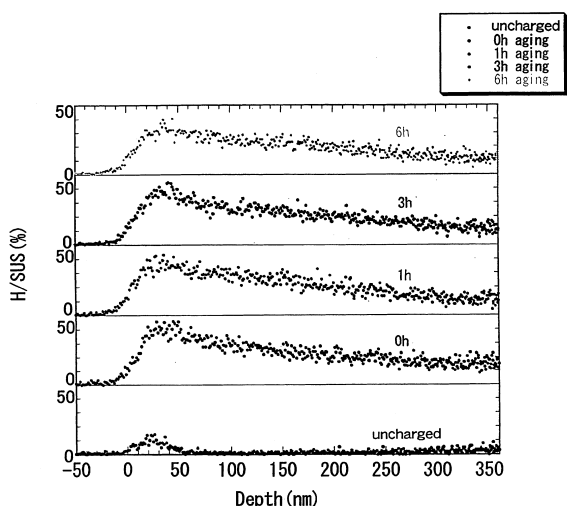


Fig. 7. Result of the ERD experiment for the depth distribution of hydrogen atoms in the surface region of a 316 stainless steel specimen electrically hydrogen charged under the current density of 5 mA/cm² for 24 h.

3.3. Positron annihilation lifetime measurement

Since positron technique is a very powerful tool to detect vacancy-type defects, positron lifetime measurement was performed for the hydrogen-charged 316 stainless steel specimen. It is considered that a high concentration of hydrogen atoms in the bulk not only causes phase transformation, but also produces some amount of vacancies or vacancy-type defects. In Fig. 8, the mean positron lifetime for the 316 stainless steel specimen electrically hydrogen charged under the current density of 100 mA/cm² for 24 h is shown as a function of aging time at room temperature. It is seen that just after hydrogen charging, the positron lifetime increases from 106 ps (bulk of stainless steel) to 118 ps and after that the lifetime gradually decreases. In Fig. 9, the result of two-component analysis is shown, where a longer lifetime, about 300 ps, is seen. This long lifetime may be due to vacancy clusters, i.e. microvoids of about 10 vacancies. These microvoids are considered to be formed by clustering of vacancies which are produced in the matrix or in the surrounding region of phase-transformed part of the matrix, under the presence of high concentration of hydrogen atoms. There might be some hydrogen molecules in the microvoids, but the microvoids are not completely filled with hydrogen molecules because if so, longer lifetime cannot be observed. The long lifetime component gradually decreases during room temperature aging, probably due to the conversion from microvoids to collapsed type defects like stacking fault tetrahedra.

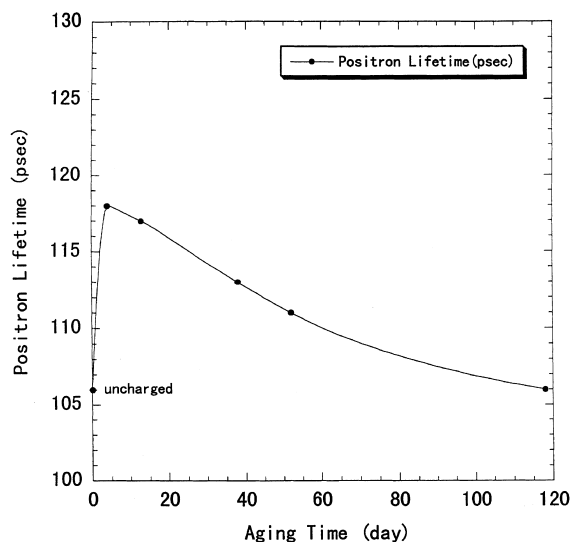


Fig. 8. Mean positron lifetime experimentally obtained for a 316 stainless steel specimen electrically hydrogen charged under the current density of 100 mA/cm² for 24 h as a function of aging time at room temperature.

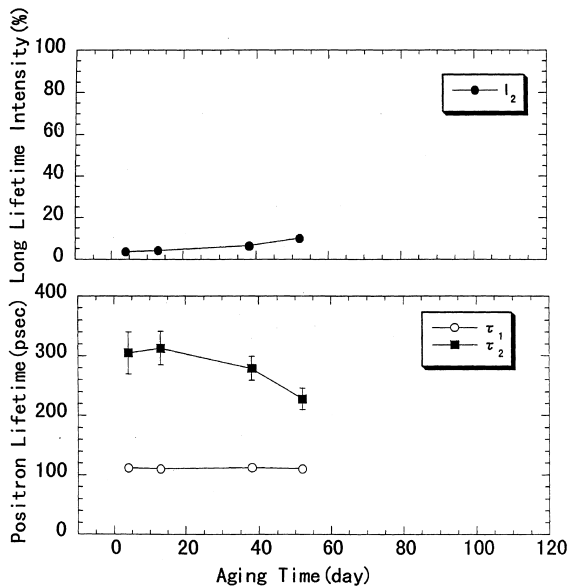


Fig. 9. Result of two-component analysis of positron lifetime spectrum obtained for a 316 stainless steel specimen electrically hydrogen charged under the current density of 100 mA/cm² for 24 h as a function of aging time at room temperature.

4. Conclusion

The electrically hydrogen charged 316 stainless steel specimens showed the hydrogen-induced embrittlement, i.e. the increase of the yield stress and the decrease of the tensile strain. This phenomenon coincides with the appearance of the ϵ -phase and α -phase and decrease of the γ peak in the X-ray diffraction analysis for the charged

specimens. 3-day aging causes the recovery from the hydrogen embrittlement in these specimens. Hydrogen concentration was measured by the ERD method and a very high concentration of hydrogen atoms, H/atom ~ 0.4 was obtained, which causes the phase transformation in the γ -phase. Positron annihilation lifetime measurement was made for the charged specimens and longer lifetime about 300 ps was observed, which corresponds to microvoids of about 10 vacancies (probably larger than this, because hydrogen atoms exist inside). These vacancies might be formed by the presence of high concentration of hydrogen atoms, probably near the phase-transformed region.

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