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Hydrogen traps in ion-irradiated F82H steel observed by NRA

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ABSTRACT

Characteristics of irradiation-induced trapping of hydrogen were investigated for quantitative evaluation of tritium retention in F82H steel. Before and after irradiation of 0.8-MeV ^{4}He or 0.3-MeV H ions, deuterium depth profiles near the surface of a disk sample were observed by nuclear reaction analysis under continuous exposure of deuterium plasma. One type of trap, with a trapping energy of 0.66 eV, was observed after each irradiation. The ratio of trap production rate to atomic displacement was 0.0046 and 0.0014 for He and H irradiation, respectively. Annihilation occurred around 600 K for H irradiation but was not observed even at 691 K for He irradiation. Traps are likely to be interstitial-like sites associated with dislocation loops. This study also indicates that helium plays a role in inhibiting trap annihilation. In addition, the deuterium diffusion coefficient in non-irradiated F82H was determined by a time-lag permeation experiment.

1. Introduction

F82H ferritic/martensitic steel is a candidate material for structural components of a fusion reactor due to its low-activation property [1]. When the components are heavily irradiated by fast neutrons, irradiation damage and irradiation-induced defects are produced that can act as hydrogen traps and increase tritium retention. Quantitative evaluation of tritium retention requires an understanding of the characteristics of the traps (e.g., trapping energy and trap density). However, limited data are available regarding hydrogen trapping in irradiated low-activation steel.

Forcey et al. [2] observed transient deuterium permeation through MANET II martensitic steel and derived the trapping energy (0.62 eV) and the diffusion coefficient. Serra et al. [3] used a similar permeation technique to obtain the trapping energies of 0.58 eV for F82H and 0.45 eV for BATMAN, respectively. These studies suggest intrinsic traps for hydrogen isotopes in ferritic/martensitic steels. When steel is irradiated, irradiation-induced traps are added to the intrinsic traps and analysis of permeation behavior becomes more complex.

The present work investigates the characteristics of hydrogen traps in F82H steel using an in situ observation technique [4]. This technique allows for the observation of deuterium concentration in a sample under steady-state permeation conditions and, therefore, for estimating the amount of deuterium in irradiation-induced traps (as will be shown later).

2. Experiments

Two experiments on deuterium trapping were conducted. One was a transient permeation experiment to estimate diffusion coefficient and observe the intrinsic traps. The other was an in situ observation of deuterium depth profiles before/after ion bombardment to characterize irradiation-induced traps.

2.1. Transient permeation

A disk sample with thickness of 2.0 mm and diameter of 21 mm was cut from a F82H block and mechanically polished with 0.3-μm alumina. A general composition of F82H is 0.1 C, 8 Cr, 2 W, 0.2 V, and 0.04 Ta in % and Fe [5].

The experimental setup has been detailed elsewhere [6] and only briefly described here. The sample was set between two vacuum chambers and one side of the sample was exposed to deuterium radio-frequency (RF) plasma. The RF power was 20 W and discharge pressure was 1 Pa. Because the energy of deuterium particles incident on the sample was very low, typically 1 eV [7], there were no effects of defect formation or temperature change. A mechanical shutter with one large and several small holes was located between the plasma and the sample. As the position of the shutter was switched between the large hole and the small holes, the incident deuterium flux was changed instantaneously. A quadrupole mass analyzer was used to observe the transient behavior of deuterium permeation through the sample between 408 and 667 K.

The transient permeation behavior was analyzed by the time-lag method [8]. In this experiment, the incident flux never becomes

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zero and the apparent diffusion coefficient (D_a) is related to lag-time (τ) by [9].

$$\tau = \frac{L^2}{6D_a}\left(1 - \frac{J_i}{J_f}\right) \quad (1)$$

where J_i and J_f are the permeation flux at the steady state before and after the shutter is switched, respectively, and L the sample thickness. The lag time (τ) is an intercept of the asymptotic line, which is the time-integrated function of the permeation flux.

2.2. In-situ observation of D trapping

Sample material, size, and polishing were the same as described above, as was the experimental setup except that the shutter was removed and the chamber connected to a beam duct of the 4 MV van de Graaff accelerator of Kyoto University. Details of the experimental setup are explained elsewhere [10].

Deuterium depth profiles near the plasma-exposed surface of the sample were measured by nuclear reaction analysis (NRA) using the reaction D(^{3}He,p)^{4}He. A $^3He^+$ beam with 1.7 MeV energy was injected in the sample at 45° and produced protons emitted at 174.3° were detected by a solid-state detector. The proton energy spectrum was converted to a deuterium depth profile from surface to 1.5 μm-depth. The ion flux was kept below 1×10^{16} m^{-2} s^{-1} to avoid temperature increases in the sample.

To introduce damage, the sample was irradiated with 0.8 MeV ^{4}He ions or 0.9 MeV H_3 ions at 45°. Typical ion flux was 3.5×10^{16} and 2×10^{16} m^{-2} s^{-1} for ^{4}He and H_3, respectively. A total particle dose was 1.5×10^{21} and 1.0×10^{22} m^{-2} for ^{4}He and H, respectively. Before and after the irradiation, NRA was conducted to observe changes in the deuterium concentration. It should be noted that one side of the sample was continuously exposed to plasma and the deuterium concentration was observed under steady state permeation conditions.

3. Results and discussion

3.1. Deuterium diffusion

Fig. 1 shows typical permeation curves of deuterium observed at 626 K after the shutter was switched to the large hole (increase) and to the small holes (decrease). Permeation curves calculated with D_a [9], which is derived by the time-lag method, agree with the experimental data as shown in Fig. 1. At other temperatures between 408 and 667 K, all the experimental data are well reproduced by D_a. This indicates that the permeation is limited by diffusion.

As shown in Fig. 2, D_a data obtained in the above experiment lie on a straight line at higher temperatures but deviate below 550 K due to a delay from the intrinsic traps. Assuming a local equilibrium exists between deuterium atoms in solution sites and in traps and that the trap occupation is small, D_a can be related to the diffusion coefficient D by [2,11],

$$D_a = D[1 + (C_0/hN)\exp(E_t/kT)]^{-1} \quad (2)$$

where C_0 and hN are the density of the trap and the solution site, respectively, E_t is the trapping energy, k is the Boltzmann constant, T is the temperature, h is the number of solution sites per host atom, and N is the host atomic density. In general, $h = 6$ for martensitic steel [2]. As D_a approaches D at higher temperatures, D is estimated from the data between 556 and 667 K, that is,

$$D = 3.6 \times 10^{-8}\exp(-0.076\ (\mathrm{eV})/kT) \quad \mathrm{m^2 s^{-1}} \quad (3)$$

A solid line in Fig. 2 is the best fit result of the data with Eq. (2) and it is found that

$$C_0/hN = 2.5 \times 10^{-7} \quad \text{and} \quad E_t = 0.62\ \mathrm{eV}.$$

In addition, Fig. 2 shows D_a by other researchers (i.e., Serra et al. [3], Forcey et al. [2], and Dolinsky et al. [12]). The data for F82H (E_t = 0.58 eV) and BATMAN (E_t = 0.45 eV) [3] are very close to the present data, indicating the same type of trap. The data of MANET II (E_t = 0.62 eV) [2] is separate from these data; however, the trapping energy is nearly the same. Discussions by Serra et al. [3] and Forcey et al. [2] indicate that the trap is likely to be associated with the lath boundaries and with dislocations. This is supported by the fact [12] that the traps in F82H are annihilated by careful annealing at high temperature.

3.2. Deuterium depth profiles

Fig. 3 shows deuterium depth profiles at 376 K before and after ^{4}He irradiation. Before irradiation, represented as 0 dose in the fig-

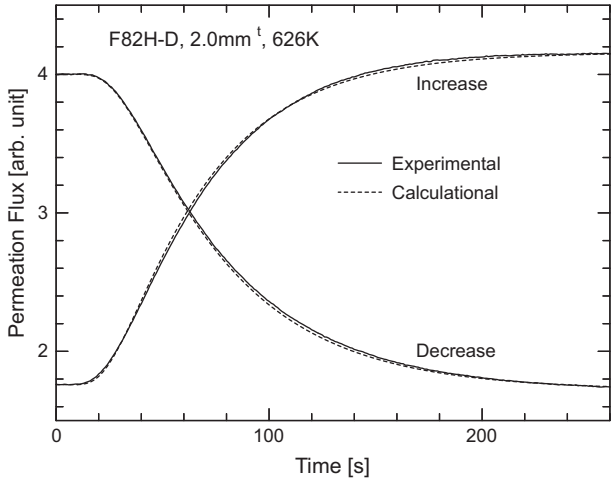

Fig. 1. Transient permeation flux through F82H sample after the shutter is switched to the large hole (increase) and to the small holes (decrease).

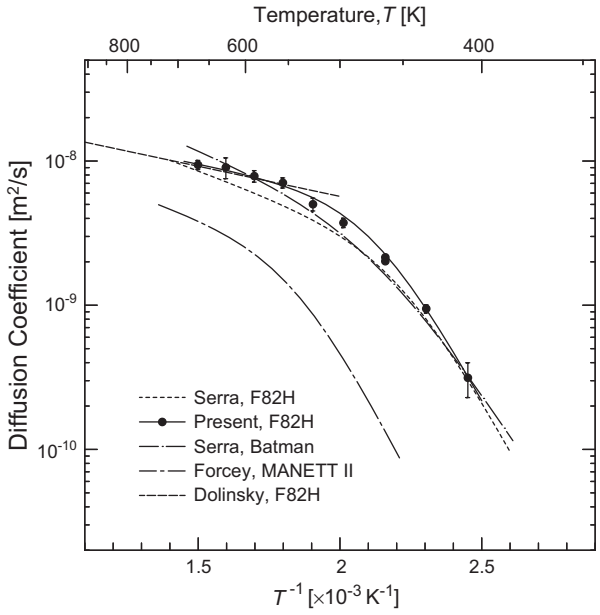

Fig. 2. Apparent diffusion coefficient of deuterium in F82H estimated by the time-lag method. Other experimental works on ferritic/martensitic steel are also shown.