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Production and annihilation of deuterium traps in He-irradiated vanadium

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Abstract

Deuterium trapping in vanadium pre-irradiated by helium ions was experimentally studied. One side of a vanadium sheet was continuously exposed to deuterium plasma and irradiated by 0.8 MeV ^{3}He ions with a dose of 1.2×10^{21} m^{-2} to introduce radiation damages. Deuterium depth profiles in the irradiated region were observed by a nuclear reaction analysis. Many traps were produced by the irradiation and the deuterium concentration at 348 K increased to 10 times compare to the concentration before the irradiation. The production rate of the trap to atomic displacement was estimated to be 0.021. The trap was fully annihilated at 523 K. These behaviors suggested that the trap was atom vacancies associated with the radiation damages.

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1. Introduction

Vanadium and its alloys are candidates for the structural material of fusion reactors due to their low activation property and high refractory [1]. Evaluation of the tritium inventory in these materials is one of important issues since vanadium is an exothermic metal for hydrogen absorption and it tends to contain much hydrogen isotopes than endothermic metals. Besides, irradiation by energetic particles would produce deep potential sites for hydrogen, so-called traps, to increase the inventory. As for absorption, the evaluation is possible using thermal properties [2] and recombination coefficients [3]. As for trapping, some experimental results [4–6] are available but further informations such as production and annealing behavior of the trap are needed. Effect of helium, which is implanted from fusion plasma and produced by tritium decay in plasma-facing walls, on the trapping is also concerned. In the present work, deuterium trapping in helium-irradiated vanadium is studied using a nuclear reaction analysis (NRA).

2. Experimental

The sample was a vanadium sheet of 0.127 mm thickness (Johnson Matthey Plc.). Its purity was 99.8% in metal basis and major impurities were Si

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(470 ppm), Fe (220 ppm), O (110 ppm), Al (93 ppm), N (78 ppm), Nb (63 ppm) and C (29 ppm). The sample sheet was polished with emery paper up to #1500 mesh and finished with a 0.05 μm alumina powder.

The experimental setup and procedure have been described elsewhere [6] and will be explained briefly. The sample sheet was placed between two vacuum chambers and heated up to 348 K by a lamp. One side of the sheet was exposed to a deuterium rf-plasma and charged with deuterium atoms. A quadrupole mass analyzer at the opposite side of the sheet monitored deuterium permeation through the sample. A probe beam of 1.7 MeV ^{3}He from a 4 MV van de Graaff accelerator of Kyoto University impinged to the plasma-exposing side at 45° to the surface normal. An energy spectrum of protons emitted by a reaction of D (^{3}He, p) ^{4}He was converted into a deuterium depth profile near the surface [7]. The probe depth was 1.6 μm. Next 0.8 MeV ^{3}He ions from the accelerator irradiated the same side with a dose of 1.2×10^{21} m^{-2}. Then the NRA was repeated under different temperature conditions.

In the whole experiment, discharge conditions were kept constant and the sample was continuously exposed to the plasma even at the NRA. The NRA was conducted at the steady state in which the deuterium concentration did not change with time. This was confirmed by the constant permeation flux except for the lowest temperature of 348 K. As the permeation flux was too low at the temperature, the NRA was repeated two times with an appropriate interval of 4 ks to confirm that there is no change in the depth profile. The probe dose for each NRA run was lower than 4×10^{19} m^{-2} and the probe flux was 1×10^{16} m^{-2} to minimize probe effects.

3. Results and discussion

3.1. Depth profiles

Before irradiation of 0.8 MeV ^{3}He ions, the deuterium depth profile had a peak at surface and a plateau in the bulk as shown in Fig. 1. The former and the latter corresponded to absorbed and dissolved deuterium atoms, respectively [3]. It seems that deuterium atoms on surface expanded to both positive and negative depths due to a finite resolution of the NRA detecting system. In order to avoid this effect, the bulk region is defined between 0.2 and 1.5 μm depth hereafter. The average concentration of dissolved deuterium in the bulk was 1.5×10^{26} m^{-3} at 348 K. After the irradiation, the deuterium concentration increased and another peak appeared at 1.0 μm-depth. The peak concentration was 1.8×10^{27} m^{-3}, more than 10 times larger than that before the irradiation. As the terminal solid solubility of deuterium was higher than that concentration [8], deuterium would not be precipitated. These results indicated that the He irradiation produced many trapping sites (traps) for hydrogen isotopes and a large amount of deuterium was trapped.

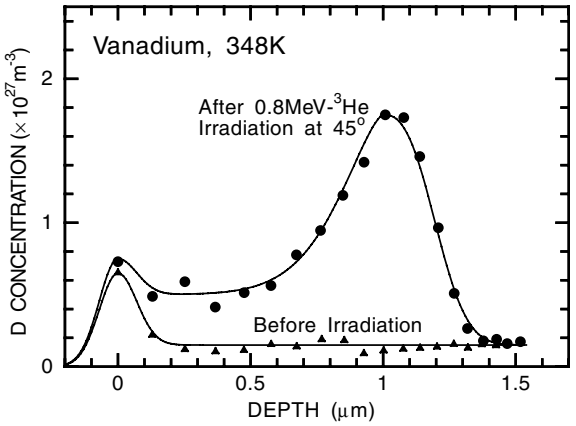

Fig. 1. Deuterium depth profiles in vanadium continuously exposed to deuterium plasma before and after irradiation of 0.8 MeV ^{3}He ions.

The depth profile of trapped deuterium can be obtained by subtracting the profile before the irradiation from that after the irradiation. It is shown in Fig. 2 with distributions of irradiating He ions and displaced host vanadium atoms. These distributions are estimated by the TRIM code [9] in which displacement energy is taken as 20 eV. It is clearly shown that the distribution of displacement is very similar to the depth profile of trapped deuterium. So the trap would be associated with displacement, that is, radiation damages. The He distribution is also similar around the peak but not for a shallow region between 0 and 0.5 μm, in which He ions do not stop nor migrate [10] from deeper region.

The FWHM (full width at half maximum) of the peak in the deuterium profile is 0.42 μm while that in the displacement distribution is 0.33 μm. This difference is larger than the depth resolution. One possible explanation is that the trap migrates from the damaged point to some distance until it is immobilized.

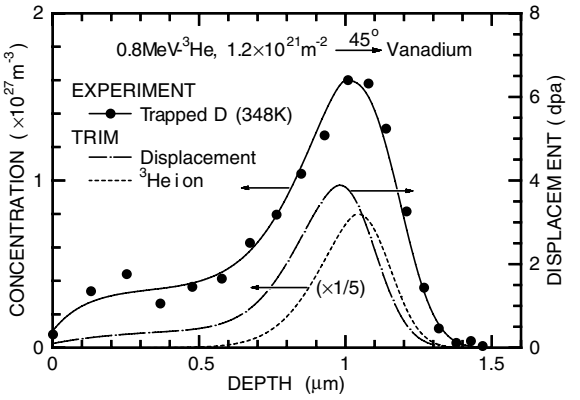

Fig. 2. A depth profile of trapped deuterium in vanadium irradiated by ^{3}He ions at 348 K. Distributions of irradiating ^{3}He and atomic displacement, estimated by the TRIM code, are also shown.

3.2. Trap production rate

An integral of the deuterium concentration from 0.2 to 1.5 μm depth is taken as an areal density of deuterium. Fig. 3 shows evolution of the areal density of trapped deuterium with atomic displacement. The dose of He is also indicated by the upper axis. Filled circles are the data for the He-irradiated sample described above and filled squares for another sample under the same experimental conditions. Although the sample temperatures are slightly different, the data in the two samples are well reproduced.

In case of H irradiation [6], the areal density of trapped deuterium at 366 K was 3.1×10^{20} m^{-2} under 0.3 MeV H irradiation with 1.2×10^{21} m^{-2} dose. Irradiating H ions readily diffused away and the trap was considered to be atom vacancies. For comparison with the present work, the data are plotted as open triangles with the same displacement scale in Fig. 3. It is found that dependence of the areal density on displacement for H and He irradiation is expressed as a single relation as shown in a dashed line, indicating that the same trap was produced.

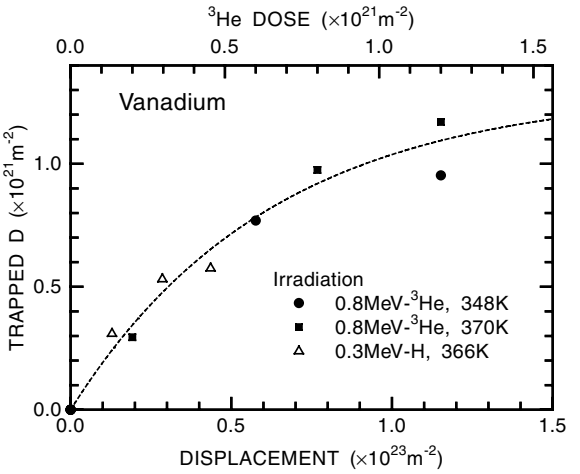

Fig. 3. Evolution of the areal density of trapped deuterium in vanadium with atomic displacement. The data for H irradiation [6] are plotted on the lower axis.

The areal density tends to saturation at higher doses. Irradiation by energetic particles produces interstitial host atoms and vacancies. The interstitials would migrate to escape from the damaged region while the vacancies would not. Consequently the number of vacancies, or trap, increase linearly with the irradiation dose. The vacancies accumulate as the irradiation progresses until they begin to be annihilated by the interstitials newly produced. At very high doses, production and annihilation of the vacancies would become balanced.

The trap was fully occupied by deuterium atoms at low temperature [6] and the amount of trapped deuterium was equal to that of the vacancies. Here the production rate of the trap is defined as a ratio of the number of the trapped atoms to the displacements. From the initial slope of the dashed line in Fig. 3, the maximum production rate is estimated to be 0.021. The value is comparable to 0.015 for nickel under the same experimental conditions [11]. As most irradiation defects would recover during short time annealing just after a collision cascade, it is plausible that these values are much less than unity.

3.3. Thermal annihilation

After the irradiation of 348 K, the sample temperature was increased to 523 K and then decreased to 333 K. The observed data of the deuterium areal density are shown in Fig. 4. An open circle represents the start point, corresponding to the profile before the irradiation in Fig. 1. Arrows A and B indicate the 0.8 MeV ^{3}He irradiation and the subsequent temperature increase, respectively. With increasing the sample temperature, the concentration of dissolved deuterium decreased due to increasing the recombination coefficient [3] and the amount of trapped deuterium decreased due to enhancement of detrapping. Consequently the areal density decreased as shown in the arrow B. At this stage, the trap would not be thermally annihilated

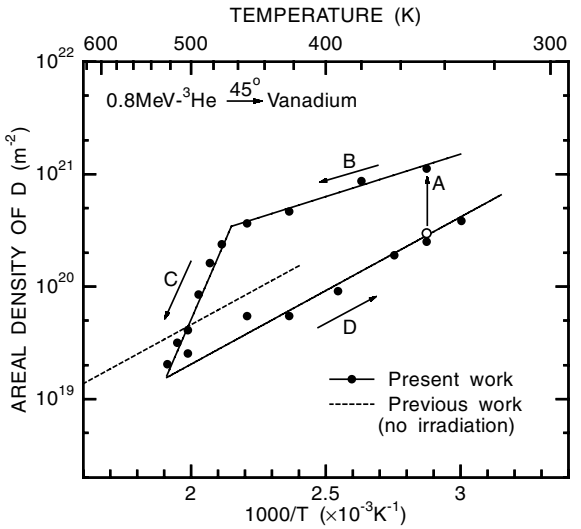

Fig. 4. Four sequential stages of the amount of deuterium; increase with ^{3}He irradiation (A), decrease with increasing temperature (B), subsequent sudden decrease (C) and gradual increase with decreasing temperature (D). An open circle represents the start point. A dashed line indicates the data without irradiation [3].

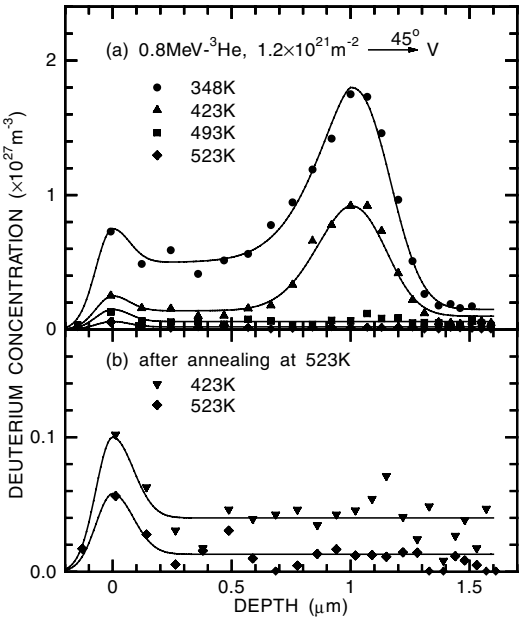

Fig. 5. Deuterium depth profiles at several temperatures of (a) stage B and (b) stage D. Symbols of B and D are denoted in Fig. 4.

which was supported by the fact that the depth profile at 423 K still had the peak in the bulk as shown in Fig. 5(a).

The areal density abruptly decreased above 470 K as indicated by an arrow C in Fig. 4 and the peak in the depth profile disappeared at 493 K in Fig. 5(a). These suggested thermal annihilation of the trap. When the sample temperature was again set to 423 K, the peak was no longer seen in Fig. 5(b). Note that the vertical axis is expanded to ten times as that in Fig. 5(a). The areal density increased with decreasing temperature as indicated by an arrow D in Fig. 4 until it just returned to the initial value before the irradiation. A dashed line in the figure represents the data observed in a non-irradiated sample in our previous work [3]. One can say that the dashed line agrees with the data at stage D, considering some difficulties in reproducing intensity of the plasma. The trap was fully annihilated by thermal processes during the stage C and all deuterium atoms existed in solution sites at stage D.

Free vacancies generally recover at very low temperature [12] but vacancies produced by irradiation recover at higher temperatures of 410–470 K [13–15] probably because they are immobilized by impurity atoms or irradiation defects. Similar annihilation behavior has been observed in H-irradiated vanadium [6]. Agreement of these recovery stages of the annihilation temperature supports the trap being the vacancies.

There was not a steady state in stage C where the amount of the trap continuously decreased. Hence quantitative consideration is difficult and only the time sequence at stage C will be described. It took 2.4 ks to vary the temperature from 453 to 473 K. Annihilation began during this time period. After 37 ks from 453 K, the temperature reached to 523 K and the trap was fully annihilated. The time and temperature required for annihilation of the trap agreed with the case of H irradiation [6] because of the same kind of the trap. As for production and annihilation of the trap, no differences were seen between He and H irradiation.

4. Summary

The depth profiling of deuterium in ^{3}He-irradiated vanadium was conducted with the NRA technique. Due to continuous charge of deuterium into the sample, the amount of trapped deuterium could be observed even at elevated temperatures. The results of the depth profile of trapped deuterium, the evolution of the deuterium amount with the irradiating dose and the annihilation behavior of the trap indicated that the trap was the vacancies, probably immobilized by the impurity atoms or the

irradiation defects. The trap began to disappear above 470 K until it was fully annihilated at 523 K. The production rate of the trap to displacement was estimated to be 0.021 at initial irradiation. These results of the annihilation temperature and the production rate were very similar to those in the case of H irradiation so helium itself would not so much affect trapping of hydrogen isotopes in vanadium. In more actual cases of vanadium alloys, characteristics of the traps are not well known and will be investigated in future works.

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