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Effect of helium irradiation on trapping and thermal release of deuterium implanted in tungsten

S. Nagata *, K. Takahiro

Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan

Abstract

Retention and thermal release of D atoms and lattice disorder in W single crystals pre-irradiated with He were studied by ion beam analysis techniques. The retention of 1 keV D ions implanted in W crystals was strongly enhanced by the 10 keV ^{4}He pre-irradiation to a small dose where a large distortion was created at room temperature. The total amount of D retained in near surface layer of the W crystal did not vary with a range of He pre-implantation dose between 10^{19} He/m^2 and 10^{21} He/m^2. However, the lattice disorder was significantly accumulated in near surface layer above 10^{21} He/m^2 and the thermal release of retained D atoms occurred at higher temperature. For a specimen pre-irradiated at 850 K, the crystal lattice was not heavily damaged, and trapping sites for the D atoms were uniformly distributed in the He implanted region probably owing to ordered He bubbles. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Deuterium; Defects; Helium; Retention; Tungsten

1. Introduction

In near-term fusion devices, W is a candidate material for plasma facing components because of its superior physical and mechanical properties at high temperature [1]. The plasma facing materials will suffer exposure not only of hydrogen isotopes, but also of He particles produced by fusion reactions and of He ions during the discharge cleaning. It is well known that He irradiation of metals causes much heavier damages in comparison to hydrogen irradiation [2] and efficiently enhances the retention of hydrogen atoms in the penetrated region [3,4]. The interaction between hydrogen isotopes and He irradiation-induced defects is of importance because it can modify the transport behavior of hydrogen in the surface materials, and affect the fuel recycling and tritium inventory. Although the effects of irradiation-induced defects in some metals on the hydrogen isotopes has been extensively investigated, a relatively small number of quantitative studies on the hydrogen–helium interaction in W has been reported.

In the present paper, influence of the He pre-irradiation on retention and release of the D atoms post-implanted in W single crystals was studied by using ion beam analysis techniques. Concentration depth profiles of retained D atoms implanted into W single crystals were investigated in connection with and irradiation-induced lattice disorder created by the He pre-irradiation. Thermal release of the retained D atoms and the annealing of the irradiation-induced damage were also examined to clarify the responsible trapping mechanism.

2. Experimental

The specimens used were W single crystals with $\langle 1\,1\,1 \rangle$ orientation. A single crystal rod of about 8 mm diameter was prepared from powder fabricated material (99.95 wt%) by floating zone melting methods. The rod was cut into disks of about 0.5 mm thickness, and electropolished in 0.1 N NaOH solution.

Ion implantation and the ion beam analyses were performed in a scattering chamber connected to a 1.7 MV tandem accelerator. The He and D ion implantation

* Corresponding author. Tel.: +81-22 215 2058; fax: +81-22 215 2061.
E-mail address: nagata@wani.imr.tohoku.ac.jp (S. Nagata).

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was carried out using an ion gun with a mass filter situated at the scattering chamber. Prior to the D ion implantation, He irradiation was performed with the incident energy of 10 keV at a temperature range between 290 and 860 K. The current density of the incident He was about 1×10^{18} He/m^{2}s, and the maximum irradiation dose was about 6×10^{21} He/m^2. To study the lattice disorder produced by the He ion irradiation, the ion channeling experiments along the $\langle 1\,1\,1 \rangle$ axis of the specimen were performed using the backscattering of ^{4}He ions with an incident energy varied from 0.5 to 4.0 MeV. After the He ion implantation, 1 keV D ions were implanted at room temperature and the concentration depth profiles of D atoms retained in near surface of the W crystals were measured by the elastic recoil detection (ERD). Thermal release of the retained D atoms and changes of the lattice disorder in the irradiated region were measured after each stage of the isochronal annealing for 600 s in a temperature range between 300 and 850 K. Details of the experimental setup and conditions are described elsewhere [5].

3. Results and discussions

Fig. 1 shows backscattering spectra for 2 MeV ^{4}He incident along the $\langle 1\,1\,1 \rangle$ direction in W crystals irradiated by 10 keV ^{4}He to various doses at room temperature. Backscattering yields increased with an increase of the irradiation dose of He ions, although no clear backscattering peak was observed for the He penetrated depth in the specimens irradiated to doses below 1×10^{21} He/m^2. This indicated that irradiation damage in the implanted zone accumulated during He irradiation, but did not contain a large number of point scattering centers, which contribute direct backscatering yields. He irradiation with higher dose caused significant misalignment in the implant layer because a pronounced peak appeared for the specimen irradiated to 2.5×10^{21} He/m^2. A similar defect accumulation during the He irradiation was reported for Nb single crystals [6].

The dechanneling yields of a backscattering spectrum for the D implanted crystal can be related to the number of defects in the path of the incident ions [7]. Moreover, the dechanneling cross-section has different dependence on incident ion energy according to the type of defect. Therefore, energy dependent analysis approach using dechanneling parameter allows us to examine the nature of the disorder [8]. In Fig. 2, dechanneling parameter obtained for $\langle 1\,1\,1 \rangle$ axis channeling of W crystals irradiated by 10 keV ^{4}He at room temperature are plotted as a function of the square root of the analyzing beam energy. The dechanneling parameter at the depth of about 200 nm was estimated in the same way as described elsewhere [5]. A linear increase of the dechanneling parameter was clearly seen for the specimen irradiated to a dose below 1×10^{20} He/m^2, as was

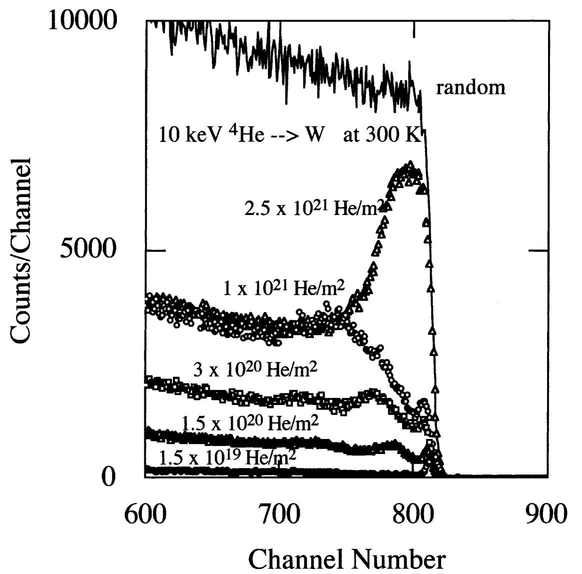

Fig. 1. Backscattering spectra for 2 MeV ^{4}He incident along the $\langle 1\,1\,1 \rangle$ direction in W crystals irradiated by 10 keV ^{4}He to various doses at room temperature.

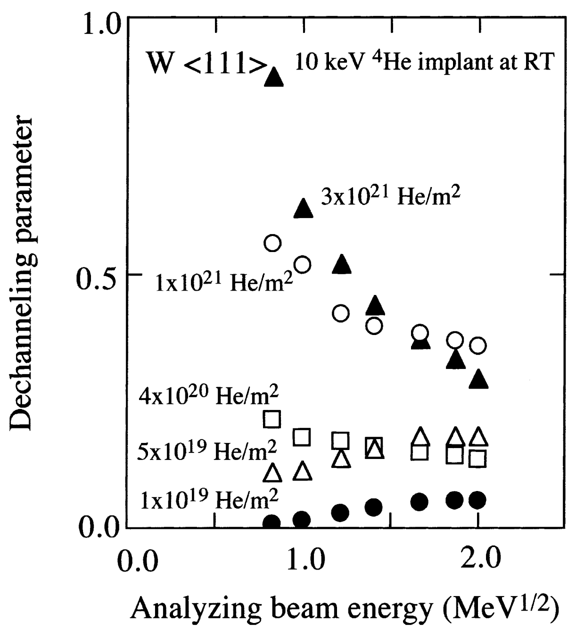

Fig. 2. Dechanneling parameter obtained for $\langle 1\,1\,1 \rangle$ axis channeling of W crystals irradiated by 10 keV ^{4}He at room temperature plotted as a function of the square root of the analyzing beam energy. Irradiation dose of He ions for each specimen is indicated in the figure.

observed for the W crystals irradiated by 5 keV D ions [5]. A flat energy dependence was shown for the doses between 10^{20} and 10^{21} He/m^2, indicating bubble formation. For higher irradiation doses above 1×10^{21} He/m^2, dechanneling parameter decreased with an increase of the analyzing beam energy. The results suggest that the lattice distortion probably due to interstitial loops was accumulated initially, and voids and/or bubbles might be created at higher irradiation doses. Above 10^{21} He/m^2, the large number of W atoms were detected as randomly distributed scattering centers, possibly caused by the blister formation which was observed for Nb crystals irradiated by He [6]. The present results about the defect accumulation during the He irradiation are consistent with the previous TEM observation on Mo irradiated by 8 keV He ions [9].

In order to investigate the influence of the He irradiation-induced damage on the D retention, the D ions with the energy of 1 keV were implanted to the same spot irradiated by 10 keV ^{4}He with various doses at room temperature. Because the incident energy of 1 keV is considered to be around threshold energy for producing displacements [10], no large number of displacements were expected during 1 keV D implantation. Fig. 3 shows retention of D atoms in the near surface layer of W crystals at room temperature plotted against the ^{4}He pre-irradiation dose. The D retention shows a sharp rise at a He irradiation dose of about 1×10^{19} He/m^2, then the D retention was nearly constant while the pre-irradiation dose of He changed in two orders of magnitude. The concentration of D atoms in the He penetrated layer seemed to be saturated and no significant increase of the retention of D atoms was observed for higher implantation dose of D ions. The maximum D concentration at room temperature is about 15 at.% which is comparable to that obtained for 5 keV D ion implantation [5].

Fig. 4 shows total amounts of D atoms in the near surface layer of W crystals pre-irradiated by ^{4}He ions plotted as a function of annealing temperature. The irradiation of 10 keV ^{4}He ions were carried out at 300 and 850 K, followed by the ion implantation of D ions with 1 keV to a dose of 2×10^{21} D/m^2 at room temperature. The D retention was evaluated by integrating the retained D atoms from surface to 150 nm after each stage of the isochronal annealing for 600 s. The release curve for the specimen irradiated with 3×10^{20} He/m^2 is nearly identical to that for the crystal without He pre-irradiation but irradiated with 5 keV D ions to 1×10^{22} D/m^2. It has been shown [5] that implantation with energetic D ions creates a large lattice distortion, probably, ion induced interstitial loops. Therefore, for low He doses, the trapping of D atoms may be related to strain fields around interstitial loops and/or around He-vacancy complexes. The large lattice distortion created by the energetic D ion may be attributed to the formation of D_2 molecules, which can induce large strain fields around them. The thermal release occurred at the higher temperature for the crystal irradiated to the higher dose,

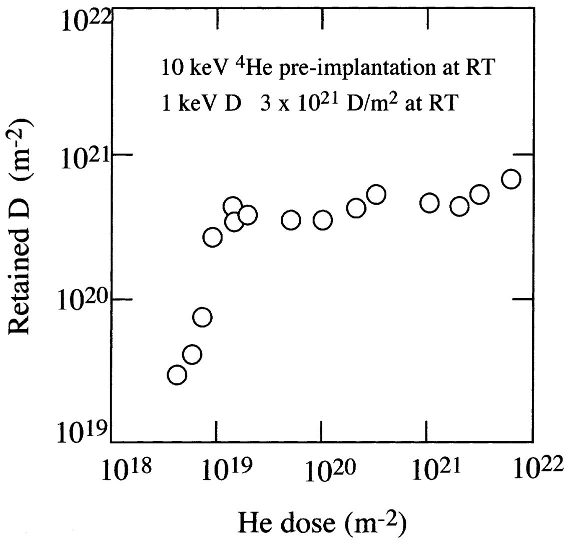

Fig. 3. Retention of D atoms in the near surface layer of W crystals at room temperature plotted against the ^{4}He pre-irradiation dose.

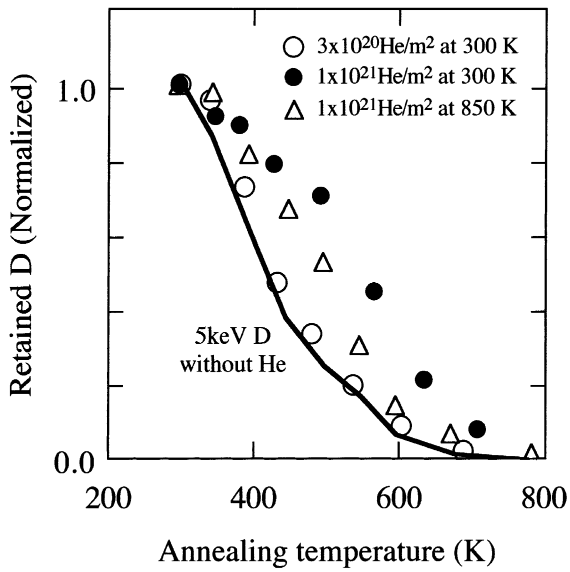

Fig. 4. Total amounts of D atoms retained in the near surface layer of W crystals pre-irradiated to a dose of 3×10^{20} He/m^2 at 300 K (○), 1×10^{21} He/m^2 at 300 K (●) and 1×10^{21} He/m^2 at 850 K (△) plotted as a function of annealing temperature. The solid line refers to the results for the W crystal irradiated 5 keV D ions to a dose of 1.4×10^{22} D/m^2 without He pre-irradiation [5].

at which He bubbles were most likely created. However, previous TDS experiments reported that D release from He bubbles contributed an extra peak at 650 K [11], which is considerably higher than the release temperature obtained in the present results. Furthermore, the D release occurred at lower temperature for the specimen pre-irradiated at 850 K, where bubbles are thought to be created more effectively than at room temperature. This means that a large fraction of the D atoms may not be bound at the bubble walls in the present experiments.

Fig. 5 shows dechanneling parameter on $\langle 1\,1\,1 \rangle$ axis channeling for specimens irradiated to a dose of about 1×10^{21} He/m^2 at 300, 650 and 850 K, plotted as a function of the square root of the analyzing beam energy. Above 650 K, the dechanneling parameter weakly depended on the analyzing beam energy and had smaller values, indicating bubble formation with less lattice disorder in comparison with the irradiation at room temperature. The dechanneling parameter obtained for the crystal irradiated with ^{4}He at room temperature followed by heat treatment at 750 K for 10 min is also shown in Fig. 5. It was the same specimen that was irradiated at 300 K, which is also plotted in the figure. The dechanneling parameter became slightly larger after the annealing, especially for higher incident energy. This enlargement of the dechanneling parameter may correspond to the growth of the He-vacancy clusters and/or He bubbles. A clear peak appeared in the backscattering spectrum at the He implanted depth, after the heat treatment up to 750 K. It also indicates rearrangements of the He-vacancy complexes and/or clusters created by He irradiation during the annealing process. The retardation of the release of retained D can be explained by the formation of defect clusters or He bubbles [12] during the isochronal annealing process.

Fig. 6 shows concentration depth profiles of retained D atoms in the near surface of the W crystals pre-irradiated by ^{4}He to a dose of 1×10^{21} He/m^2 at 300 and 850 K. Depth profile of D atoms in the crystal irradiated at 300 K was similar to the vacancy distribution calculated by TRIM95. On the other hand, D atoms flatly distributed in the near surface layer of the crystal irradiated at 850 K, and the maximum concentration was lower than that in the crystal irradiated at 300 K. It is known that ordered bcc bubble super lattices are formed in W following high-dose He implantation at high temperature [13]. Therefore, the uniform distribution of retained D atoms seems to correspond to the depth profile of the ordered bubbles, indicating the trapping of D atoms by He bubbles. As shown in Fig. 4, however, the present results of thermal release experiment do not clearly suggest the adsorption of D atoms at the He bubble walls, if one assumes relatively high dissociation enthalpy [11] for it. Because He bubble pressures can be very high, the strain filed around the bubbles may play an important role for trapping of D atoms and/or molecules.

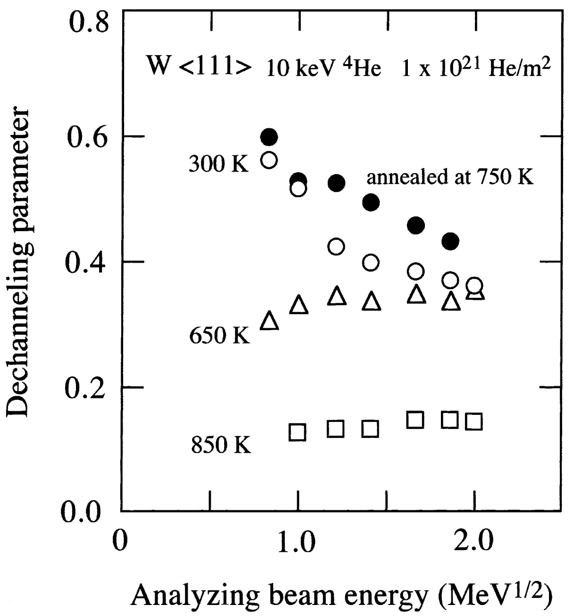

Fig. 5. Dechanneling parameter obtained for $\langle 1\,1\,1 \rangle$ axis channeling of W crystals irradiated by 10 keV ^{4}He ions to a dose of about $1 \times {}^{21}$ He/m^2 at 300 K (○), 650 K (△) and 850 K (□) plotted as a function of the square root of the analyzing beam energy. Close circles (●) indicate the parameter obtained for the specimen annealed at 750 K for 10 min after the He irradiation at 300 K.

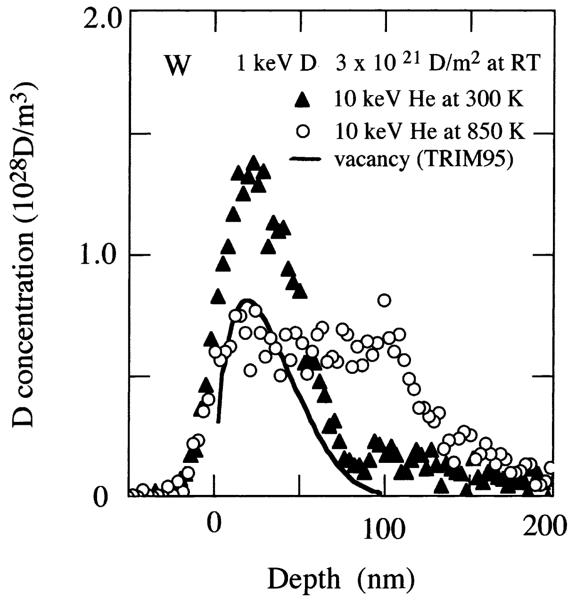

Fig. 6. Concentration depth profiles of retained D atoms in the near surface of the W crystals pre-irradiated by ^{4}He to a dose of 1×10^{21} He/m^2 at 300 K (▲) and 850 K (○). Calculated vacancy distribution (solid line) is inserted for comparison.

4. Conclusions

Retention of D atoms implanted with the energy of 1 keV to W single crystals at room temperature was strongly enhanced by He pre-irradiation to a small dose, where a large lattice distortion probably due to interstitial loops was created in the He implant zone. For the W crystal pre-irradiated by He ions below 10^{21} He/m^2, defect accumulation, trapping and thermal release behavior of D atoms were similar to those in a crystal implanted by 5 keV D ions without He pre-irradiation. This suggests that trapping mechanism of D atoms is identical for both energetic D and He irradiation. The large lattice distortion created by the energetic D ion may be attributed to the formation of D_2 molecules.

With increasing the He dose, the lattice disorder accumulated significantly in the He implanted zone, and thermal release of D atoms shifted to higher temperatures. A flat distribution of D atoms in the W crystals pre-irradiated at high temperature suggests the interaction between ordered helium bubbles and D atoms, although the present results do not directly indicate the adsorption at the He bubble walls. The strain filed around the He bubbles may also play an important role for trapping of D atoms.

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