Influence of He implantation on deuterium trapping at defects induced in W by irradiation with MeV-range W ions

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The effect of helium (He) ion irradiation on the concentration of the ion-induced defects preliminary generated in tungsten (W) was investigated. Polycrystalline mechanically-deformed W samples were damaged by irradiation with 20 MeV W ions at room temperature to 0.5 displacements per atom (dpa) at the damage peak situated at a depth of 1.35 μm, followed by 1 keV He ion irradiation at 473, 623, and 823 K to fluences in the ranges of $(3.7-5)\times10^{22}$ He/m^2. The samples were then loaded with deuterium (D) by D_2 gas exposure at a pressure of 100 kPa at 673 K for 10 h. Concentration of deuterium trapped in the W-ion induced defects was determined by nuclear reaction analysis using the $D(^3He,p)^4He$ reaction. It has been found that the effect of following He irradiation on the concentration of the W-ion-induced traps responsible for D trapping is negligible.

Keywords: Deuterium gas exposure, Deuterium retention, Displacement damage, He ion irradiation, Tungsten

1. Introduction

Due to its favorable physical properties, such as low erosion yield and high melting temperature, tungsten (W) is a candidate material for plasma-facing high heat-flux structures in future fusion reactors. As plasma-facing material, W will be subjected to intensive fluxes of energetic deuterium (D), tritium (T), helium (He) particles as well as 14 MeV neutrons (n) from the D–T fusion reaction. Neutron irradiation causes modification of the W microstructure by creating displacements in the bulk [1, 2]. These processes lead to concerns about tritium inventory in the n-irradiated W after long-term exposure to D-T plasmas.

One of the ways to investigate the influence of n-produced defects on the hydrogen isotope inventory is to simulate displacement damage in tungsten by irradiation with ions at energies in the range from several tenths to several tens of MeV and then to load the damaged W with deuterium. It has been experimentally shown that irradiations of W materials with 0.2−20 MeV H, He, Si, and W ions lead to an increase of deuterium retention in comparison with undamaged materials [3-11].

It has been shown recently that seeding of He ions into deuterium plasmas significantly reduces the deuterium retention at elevated temperatures both in undamaged W materials [12-16] and W preliminary damaged with 20 MeV W ions [8]. Figure 1 taken from Ref. [8] shows the concentration of deuterium (given in D/W) at the depth of the damage peak, ${}^{damage}C_D$, in the damaged mechanically-deformed W exposed to pure D and He-seeded D plasmas at various temperatures. After exposure to the He-seeded D plasma at temperatures above 450 K, ${}^{damage}C_D$ is always lower than that for the pure D plasma.

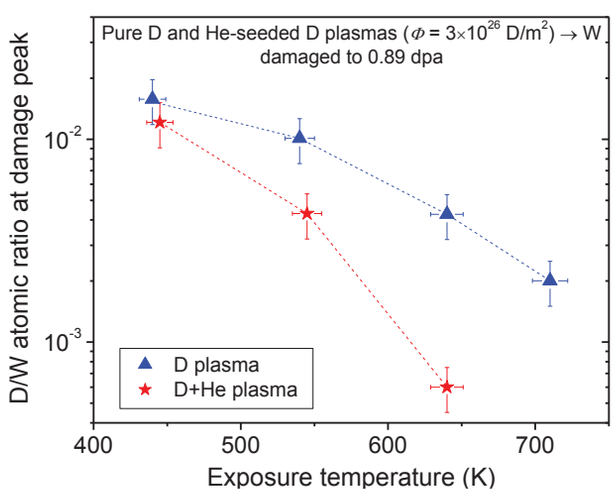

Figure 1. Concentration of deuterium (D/W) retained at a depth of the damage peak in damaged (20 MeV W ions at 300 K to 0.89 dpa) mechanically-deformed W exposed both to pure D plasma (76 eV D_2^+) and He-seeded D plasma [76 eV (D_2^+ + 10% He^+)], as a function of exposure temperature [8].

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It has been proposed that seeding of He ions into the D plasma leads to nano-scale helium bubble formation and development of an open porosity in the near-surface layer. As a consequence, short-circuit paths to the surface are created to enhance the D re-emission and limiting the D diffusion into the bulk [13, 17]. Accordingly, the D concentration in a solid solution reduces significantly and consequently the D concentration in the damage zone decreases [8, 10].

At temperatures sufficiently high to activate the detrapping process, trapping-detrapping equilibrium can be attained, and the fraction of traps, θ_t, occupied with deuterium at temperature T is expressed as

$$\theta_t/(1-\theta_t) = \theta_L \exp(E_{bin}/kT), \quad (1)$$

where θ_L is the fraction of occupied interstitial sites, E_{bin} is the binding energy (the enthalpy difference between deuterium atom in a solid solution state and in a trapped state), and k is the Boltzmann constant [18]. This means θ_t varies with the D concentration in the solid solution state, C_{SS}, being proportional to θ_L. Note that

$$C_{SS} = C_L\, \theta_L, \quad (2)$$

where C_L is the concentration of interstitial sites.

Under exposure to D plasmas, C_{SS} is determined by the balance between penetrating flux $\alpha\phi_{in}$ and re-emitted flux ϕ_r of deuterium. Here, α is the sticking coefficient and ϕ_{in} is the incident flux. In the first approximation, in a steady state

$$\alpha\, \phi_{in} = \phi_r = 2\sigma\, k_r\, C_{SS\text{-}S}^2, \quad (3)$$

where $C_{SS\text{-}S}$ is the concentration of solute D atoms beneath the surface, σ is the surface roughness factor [19], and k_r is the recombination rate constant. Hence,

$$C_{SS\text{-}S} = (\alpha\, \phi_{in}/2\sigma\, k_r)^{1/2}, \quad (4)$$

and $C_{SS\text{-}S}$ decreases as the surface roughness increases.

It should be noted that when tungsten is exposed to D_2 gas, C_{SS} is determined by Sieverts' law:

$$C_{SS} = k_0 \exp(-E_S/kT) P^{1/2}, \quad (5)$$

where k_0 is the solubility constant, E_S is the heat of solution and P is the D_2 pressure.

However, there is another assumption that formation of He bubbles and accompanying emission of self-interstitial atoms and dislocation loops [20, 21] can reduce the concentration of W-ion-induced defects responsible for trapping of D atoms in the damage zone.

The objective of this work is, therefore, to study the effect of He ion irradiation on the concentration of the ion-induced defects preliminary generated in W. W samples damaged with 20 MeV W ions were irradiated with 1 keV He ions and then loaded with D by exposure to D_2 gas at 673 K and 100 kPa. In contrast to plasma exposure, D loading by gas exposure is an equilibrium process. The concentration of solute D and consequently trap occupancy by D are determined solely by D_2 gas pressure and temperature as formulated above, but open porosity has no influence on it. Therefore, the change in the defect concentration should be clearly detected if it occurred.

In this work polycrystalline mechanically-deformed W was mostly used, and recrystallized W was also used in some of experiments for comparison. It should be noted that after irradiation with 20 MeV W ions, the D concentration in the damage zone doesn't depend on the initial microstructure of these W materials [9].

2. Experimental

Mechanically-deformed polycrystalline W from A.L.M.T. Corp., Japan, having a purity of 99.99 wt.% was used for irradiation with He ions. The microstructure of this W material consists of anisotropically elongated grains along the deformation axis. The grain size is 1-3 μm in section and up to 5 μm in length. Individual elongated cracks observed between grains are due to the deformation treatment [22]. Square-shaped samples, 10×10 mm^2 in size and 2 mm in thickness, were cut from mechanically-deformed polycrystalline W rods annealed at 1173 K for 30 min in a hydrogen atmosphere to relieve internal stresses occurred during the manufacturing process. The samples were prepared such that the irradiated surfaces were perpendicular to the deformation axis, which corresponds to the ITER specification [23]. The samples were mechanically polished, cleaned in an acetone ultrasonic bath, and then annealed in vacuum at 1173 K for 30 min to relieve stresses induced in the polishing process.

The W samples were irradiated with 20 MeV W ions to a fluence of 8×10^{17} W/m^2 at room temperature using a tandem accelerator at IPP Garching. As a result, the near-surface layer of the samples was damaged to 0.5 displacements per atom (dpa) at the damage peak situated at a depth of 1.35 μm. The damage profile was calculated using the program SRIM 2008.03 [24], "full cascade option", with a displacement threshold energy of E_d = 90 eV [25]. In what follows the W-ion-irradiated W samples will be designated as "damaged" W samples, and a value of the damage level will be indicated as a number of dpa at the damage peak.

The damaged W samples were irradiated with 1 keV He ions at temperatures of 473, 623, and 823 K using the HiFIT device at Osaka University [26]. The ion flux was $(2.7-3.7)\times10^{19}$ He/m^{2}s, and the fluences were 3.7×10^{22} and 5×10^{22} He/m^2. The sample was heated during the irradiation by using an infrared heater placed behind the sample. Temperature was measured by a type K thermocouple attached at the front surface of samples. The depth distribution of implanted He atoms was characterized by the projected range of about 4 nm and longitudinal straggling of about 7 nm [21].

After He ion irradiation the damaged W samples were exposed to D_2 gas with a pressure of 100 kPa at

673 K for 10 h in a gas exposure apparatus at University of Toyama. Damaged W sample without He ion irradiation was also exposed to D_2 gas for comparison. The sample was placed inside the quartz tube connected to the high-vacuum pumping system and heated in a vacuum with the use of an external ohmic heater. The temperature was monitored using a type K thermocouple contacted directly to the sample inside the tube. As the sample temperature reached the required value, a valve between the tube and the pumping system was closed and the tube was filled with D_2 gas. The background pressure inside the tube was measured with an ionization gauge, whereas the D_2 gas pressure was controlled with a Baratron capacitance manometer. After reaching required exposure duration, D_2 gas evacuation and sample cooling started simultaneously. D_2 gas was evacuated in several seconds, while the sample was cooled down in several minutes.

After the D_2 gas exposure, the near-surface deuterium depth profiles in the W samples were determined by nuclear reaction analysis (NRA) at IPP Garching. The $D(^3He,p)^4He$ reaction was utilized, and both the α particles and protons were analyzed. An analyzing beam of ^3He ions with energies varied from 0.69 to 4.0 MeV was used, which allows to determine the D concentration to depths of 6 μm. For the deconvolution of the proton yields measured at different ^3He ion energies, the program SIMNRA [27] was used. A deuterium depth distribution was assumed taking into account the near-surface depth profile obtained from the α particle energetic spectrum, and the proton yield was calculated as a function of incident ^3He energy. The shape of the D depth profile was then varied using an iterative technique until the calculated curve matched the measured proton yields [28].

3. Results and discussion

Deuterium depth profiles in damaged (20 MeV W ions at 300 K to 0.5 dpa) W samples (Fig. 2) show that in the course of D_2 gas exposure (at 673 K and 100 kPa for 10 h) D atoms dissolve into W matrix, diffuse through the metal lattice, and become trapped at defects (or traps) like vacancies and vacancy clusters [29-31] generated from the W-ion-induced displacement damage. Note that the D concentration beyond the damage zone (at depths ≥ 3 μm) is $(3-6)\times10^{-3}$ at.% that is significantly higher than the solute D concentration. Obviously, the intrinsic defects such as intergranular cracks in the bulk of the mechanically-deformed W serve as traps for diffusing D atoms.

Figure 3 shows the D concentration (given as D/W) at the damage peak (at a depth of ~1.35 μm) after D_2 gas exposure at 673 K as a function of annealing / He irradiation temperature. Below 623 K the D concentration at a depth of the damage peak, $^{damage}C_D$, is roughly constant regardless of prior He irradiation or the He irradiation temperature, while $^{damage}C_D$ becomes slightly lower after He ion irradiation at 823 K.

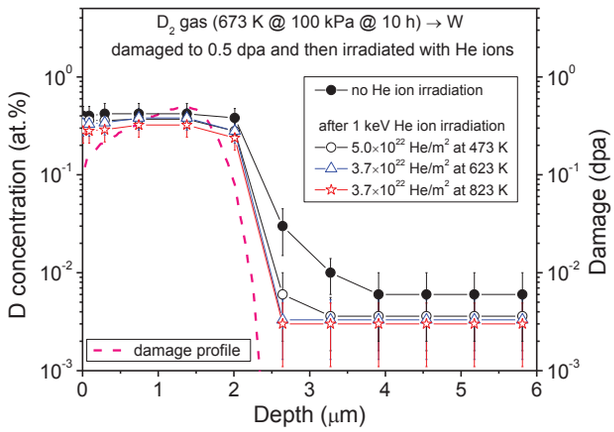

Figure 2. Depth profiles of deuterium retained in damaged (20 MeV W ions at 300 K to 0.5 dpa) mechanically-deformed W samples without and with following irradiation with 1 keV He ions, after exposure to D_2 gas (at 673 K and 100 kPa for 10 h). The He ion irradiation temperatures and fluences are indicated in the legend. Damage profile is plotted with the use of right ordinate axis.

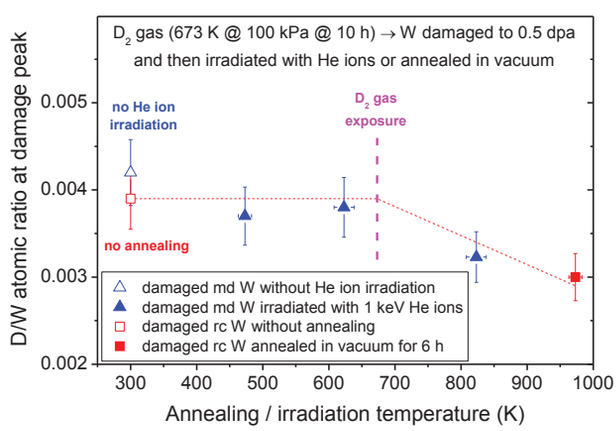

Figure 3. Concentration of deuterium (D/W) retained at a depth of the damage peak in (i) damaged (20 MeV W ions at 300 K to 0.5 dpa) mechanically-deformed (md) W samples, without and with following irradiation with 1 keV He ions, and (ii) in damaged (20 MeV W ions at 300 K to 0.5 dpa) recrystallized (rc) W samples, without and with following annealing at 973 K for 6 h, after exposure to D_2 gas (at 673 K and 100 kPa for 10 h), as a function of the exposure and annealing temperature.

Figure 3 also shows the D/W at damage peak obtained from damaged (20 MeV W ions at 300 K to 0.5 dpa) recrystallized W, without and with after-damaging annealing in vacuum at 973 K for 6 h, followed by D_2 gas exposure under the same exposure conditions (at 673 K and 100 kPa for 10 h) [32]. Comparison of these data

suggests that the $^{damage}C_D$ is more influenced by the annealing temperature rather than the prior He ion irradiation. In other words, the effect of irradiation with He ions on the concentration of the W-ion-induced traps responsible for trapping of hydrogen isotopes is virtually negligible. It appears that the self-interstitial atoms of W and dislocation loops produced beneath the surface by He implantation did not diffuse to deeper regions.

Recently, ion-driven permeation of D through W under simultaneous He–D irradiation at temperatures in the range from 800 to 1050 K was studied by Lee et al. [33]. Reduced deuterium permeation from 1 keV He ion implantation was observed, and the permeation flux reached steady state following an implanted He fluence of ~10^{21} He/m^2. It has been concluded that under simultaneous He–D irradiation a reduction in diffusive D concentration takes place [33]. However, the results reported in [33] don't allow distinguishing effects of (i) enhancement of the D re-emission and (ii) retardation of D diffusion caused by trapping of D atoms at defects generated by 1 keV He ion irradiation.

As may be inferred from the D depth profiles shown in Fig. 2, trapping of deuterium at the He-ion-induced defects is negligible. Otherwise, after D_2 gas exposure, the near-surface D concentration (i.e., within the range of He ions) in the He-ion-irradiated damaged W would be higher than that for the damaged by only W self-irradiation. In fact, the near-surface D concentration in the He-ion-irradiated damaged W is slightly lower.

It should be also noted that in damaged W the ion-induced traps are filled by D atoms diffusing from the surface [11], and, as the D loading time increases, the boundary separating layers unsaturated and saturated with deuterium migrates from the surface into the bulk. Thus, the plateau-like D depth profiles shown in Fig. 2 indicate saturation of the damage zone with deuterium.

A remark is in order that the plateau-like D depth profiles observed in damaged mechanically-deformed W after exposure to He-seeded D plasma [8] also suggest that reduction of the D concentration in damaged zone caused by seeding of He ions into D plasma can not be explained by retardation of D diffusion due to trapping of D atoms at W-ion-induced defects.

Thereby, significant decrease of the D concentration in the damage zone of W materials exposed to He-seeded D plasmas at elevated temperatures [8] can be explained by significant reduction of the D solute concentration [10]. It is safe to assume that under exposure of W materials to the He-seeded D plasmas at temperatures above 400 K, a mechanism of nano-scale helium bubble formation leads to the development of an open porosity in the near-surface layer and creates short-circuit paths to the surface. As a consequence, the D re-emission flux is enhanced and the flux of D atoms diffusing into the bulk decreases [13, 17]. As this takes place, the D concentration in the solid state is reduced as compared to the case of pure D plasma exposure. As a result, the D retention decreases in W materials, both without and with W-ion-induced damages.

4. Summary

It has been revealed that irradiation of W, beforehand damaged with 20 MeV W ions, with energetic He ions doesn't influence the concentration of the ion-induced defects.

Significant decreasing of the D concentration in the damage zone of W materials exposed to He-seeded D plasmas at elevated temperatures can be, therefore, explained by significant reduction of the D concentration in a solute solution due to formation of He-ion-induced short-circuit paths to the surface enhancing the D re-emission flux.

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