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Effects of helium bombardment on the deuterium behavior in tungsten

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Abstract

The effects of pre-irradiation with helium ions of fusion relevant energy on trapping of injected deuterium in W was studied by thermal desorption spectrometry technique using high-resolution quadrupole mass spectrometer. Pre-irradiation with He ions caused remarkable effects on the trapping of injected deuterium. Most of the injected deuterium is desorbed between 400 and 600 K for the case without helium pre-irradiation, while additional desorption occurs between 600 and 800 K for the helium pre-irradiation case. Total amount of the trapped deuterium for irradiations of 2.0×10^{21} He/m^2 and 1.0×10^{22} D_2/m^2 is 6.2×10^{20} D_2/m^2, which is more than three times higher than that in the case of no helium pre-irradiation. The present result indicates that irradiation effects of He bombardment must be taken into account to understand and evaluate the behavior of hydrogen isotopes in fusion environment.

1. Introduction

In thermonuclear fusion reactors the behavior of hydrogen isotopes in the plasma facing materials will influence plasma parameters via re-emission of hydrogen atoms. Furthermore, the injected hydrogen isotopes and helium atoms cause problems for the wall material with respect to structural integrity and tritium retention. It is well established that hydrogen isotopes may be trapped at particular defect sites, such as vacancies, dislocations, grain boundaries and precipitates [1,2]. In the future D–T reactors, implantation of helium as fusion and transmutation product will be added. Early experiments showed that helium was strongly trapped at radiation induced defects such as vacancies in metals [3]. Therefore, helium irradiation of metals would cause much stronger influences in comparison with hydrogen irradiation [4,5]. Furthermore, helium atoms are trapped strongly by themselves in metals even in the absence of radiation induced defects [6]. It is expected, therefore, that co-bombardment of helium and hydrogen isotopes in fusion environments may affect not only the properties of the plasma facing materials but also the behavior of hydrogen isotopes in the materials. Previous experiments using nuclear reaction analysis have demonstrated that pre-irradiation by helium ions produces a significant amount of trapping sites for hydrogen isotopes in many metals [7–9]. However, the mechanism of interaction between the hydrogen isotopes and the helium induced defects is not clarified well.

High-Z materials such as W are potential candidates for the armor materials of the plasma facing components. In a previous paper, it was reported that the retention of hydrogen isotopes in tungsten is very low especially for doses less than 10^{22} ions/m^2 [10]. In the present work, effects of pre-irradiation with helium ions at fusion relevant energies on the trapping of deuterium in tungsten were studied by thermal desorption spectrometry (TDS).

2. Experimental procedures

Specimen used in the present work was high purity (99.95%) powder metallurgy W containing 40 wppm

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Mo, 20 wppm Fe, 15 wppm C and 5 wppm O and N. After cutting into the size of $5 \times 10 \times 0.1$ mm^3, mechanical polishing and electro-polishing were carried out successively. The specimens were pre-irradiated with 8 keV-He^+ at room temperature, and then irradiated with 8 keV-D_2^+ at several temperatures between room temperature and 673 K. The projected range of 4 keV-D^+ (8 keV-D_2^+) and 8 keV-He^+ ions are quite similar. After irradiation, the specimens were transferred into the TDS apparatus, where the thermally desorbed deuterium gas was measured with a high-resolution quadrupole mass spectrometer [11]. This device makes it possible to distinguish the small difference in mass of He (^{4}He: $m = 4.0026$ amu) and deuterium atoms (D_2: $m = 4.0282$ amu). TDS measurement was started after 4–8 h to get sufficient vacuum condition. The desorption rate of D_2 was quantitatively calibrated by comparing with a He standard leak with a specific relative ionization efficiency. The temperature of the specimens was increased up to 1400 K with a ramping rate of 1 K/s. Since the main desorption stage of He is very high temperature, we could not take up the TDS spectra of He in this study.

3. Results

Fig. 1 shows TDS of deuterium injected in the specimens with the pre-irradiations of 8 keV-He^+ at room temperature to doses of 1.0×10^{20} and 2.0×10^{21} He^+/m^2. Deuterium ion irradiation was performed subsequently at room temperature up to 1×10^{21} D_2/m^2. Desorption of deuterium from the sample without pre-irradiation of helium is also shown in the figure for comparison. Majority of deuterium is desorbed between

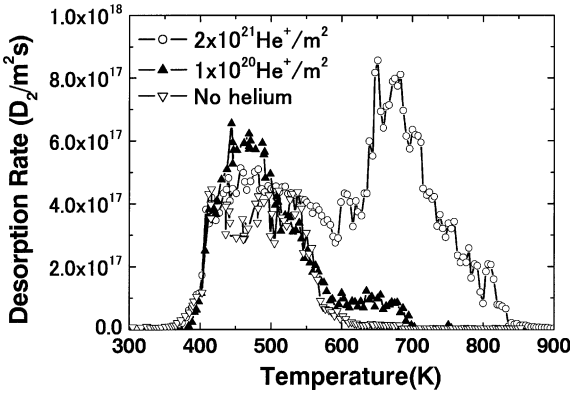

Fig. 1. Thermal desorption spectra of deuterium for samples without pre-irradiation and with pre-irradiation of 8 keV-He^+ to doses of 1.0×10^{20} and 2.0×10^{21} He^+/m^2 at room temperature.

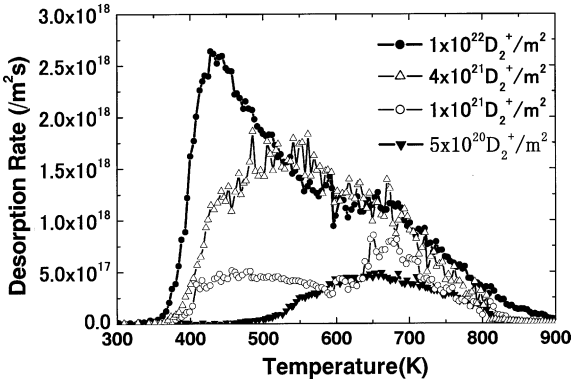

Fig. 2. Thermal desorption spectra of deuterium for tungsten pre-irradiated with 8 keV-He^+ to 2×10^{21} He^+/m^2 at room temperature for deuterium doses ranging from 5×10^{20} to 1×10^{22} D_2^+/m^2 introduced at room temperature.

400 and 600 K for the non-pre-irradiation case, while additional desorption occurs between 550 and 850 K depending on the dose of He pre-irradiation. The former and the later desorption stages are named here the low temperature and high, respectively. At the low dose helium pre-irradiation (1.0×10^{20} He^+/m^2), an additional desorption appeared above 600 K. However, this is very small compared with the major desorption stage on the low temperature side.

Dependence of TDS on deuterium dose is shown in Fig. 2 for the helium pre-irradiated specimens (8 keV-He^+, 2×10^{21} He^+/m^2, room temperature). At low dose (5×10^{20} D_2^+/m^2), the majority of the trapped deuterium desorbed at the high temperature stage (500–800 K) with almost no desorption at the low temperature stage. With increasing dose of deuterium ions, the desorption at the high temperature stage saturates at about $4.0 \times 10^{21} D_2^+/m^2$. In contrast, the desorption below about 500 K increased with increasing deuterium dose, with no sign of saturation even at 1×10^{22} D_2/m^2.

Dependence of TDS on the temperature of deuterium ion irradiation was also examined. Fig. 3 shows results for pre-irradiation of helium performed at room temperature up to 2×10^{21} He^+/m^2 and subsequent deuterium irradiation of 1×10^{21} D_2/m^2 at several temperatures up to 673 K. In the case of deuterium irradiation at 473 K, the desorption at the low temperature stage is diminished, but the large desorption at the high temperature stage still exists. One should note that trapping of deuterium for irradiation above 473 K is very small for non-helium pre-irradiated specimens [10]. With increasing irradiation temperature of deuterium, retention of deuterium becomes smaller and smaller but some desorption of deuterium at the high temperature stage is still observed even at 673 K.

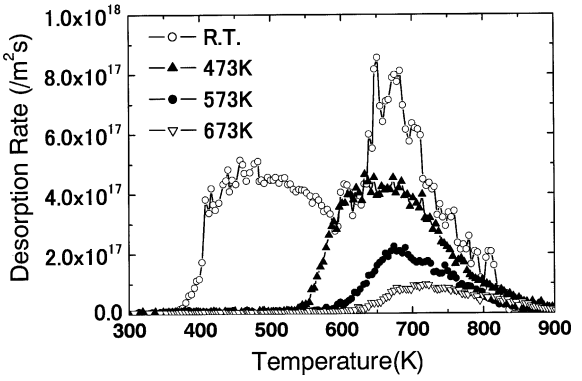

Fig. 3. Dependence of the thermal desorption spectra on deuterium irradiation temperature after pre-damaged by 8 keV-He^+ to 2×10^{21} He^+/m^2 at room temperature.

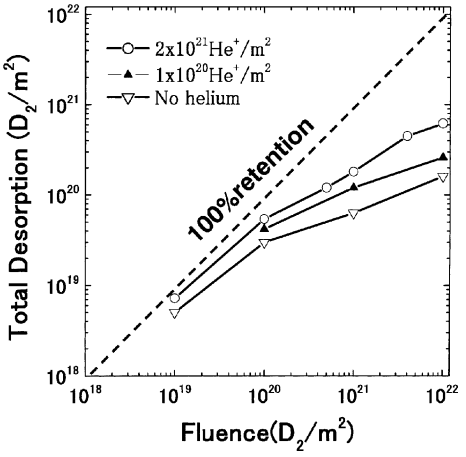

Fig. 4. Dose dependence of total amount of desorbed D_2 from the W for no pre-irradiation and for 8 keV-He^+ pre-irradiation to fluences of 1×10^{20} and 2×10^{21} He^+/m^2.

Total amounts of desorbed deuterium are plotted in Fig. 4 against deuterium dose, with and without helium pre-irradiations (1×10^{20} and 2×10^{21} He^+/m^2). The straight dashed line shows 100% retention. It is clear that the pre-irradiation of helium increases the retention of deuterium more than three times for the high dose of helium and deuterium.

4. Discussion

For determination of the mechanism of the large retention of deuterium in the helium pre-irradiated specimens, information about the microstructural evolution is essential. Fig. 5 shows transmission-electron micrograph of tungsten irradiated at room temperature by helium ions at 8 keV with doses of (a) 1×10^{20} He^+/m^2 and (b) 2×10^{21} He^+/m^2 [6]. At the low dose (1×10^{20} He^+/m^2), a large number of interstitial type dislocation loops was formed. Though it is not observable by TEM due to the limitation of the resolution, atomic size complexes of vacancies and helium atoms must be densely accumulated in the matrix, because their mobility and dissociation rates are expected to be very low at room temperature. With increasing dose of helium, the vacancy–helium complexes grow and become visible as helium bubbles as observed in Fig. 5(b). On the other hand, only dislocation loops were observed by TEM in the deuterium ion irradiated tungsten at room temperature even after doses up to 1×10^{22} ions/m^2 [12]. Accumulation of high density of vacancies is also expected. The experimental result that retention of deuterium is significant for helium pre-irradiation at 473 K but not for the non-pre-irradiation case, indicates that dislocation loops and individual vacancies do not work as trapping sites for deuterium above 473 K. Judging from the microstructure shown in Fig. 5(b), the strong trapping must be associated with the high density of small helium bubbles. It has been already reported that deuterium trapping sites in iron, pre-irradiated with helium was stronger than other defect traps [13,14]. Abramov and co-workers determined the trap strengths in nickel pre-irradiated with helium using a D_2 gas charging method [15]. The binding enthalpy was estimated to be in the range of 0.4–0.6 eV. Disadvantage of this method is, however, the difficulty in estimating the amount of injected deuterium. The present results, shown in Fig. 2, indicate that there is a clear saturation for the deuterium trapping by the strong trapping sites corresponding to the high temperature stage. For the deuterium irradiation at 473 K, most of the injected deuterium was trapped by helium bubbles. The estimated amount of the trapped deuterium is 6×10^{19} D_2/m^2. Assuming that the helium bubble is uniformly distributed up to 60 nm in depth and its volume density is $1.3 \times 10^{25}/m^3$, 154 deuterium atoms can be bound to a single helium bubble.

In the case where the helium bubbles are noticeably formed, rather weak but dense deuterium trapping sites are formed corresponding to the low temperature stage. This type of trapping increases the total amount of the retained deuterium drastically. The mechanism for the strong retention in the low temperature stage of the helium pre-irradiated case may be interpreted as follows; at low temperature, helium bubbles can grow by ejecting interstitials and interstitial loops around helium bubbles. As a result, a large number of loops are formed in the surrounding of each bubble. These interstitial loops around helium bubbles may act as trapping sites for the low temperature stage.

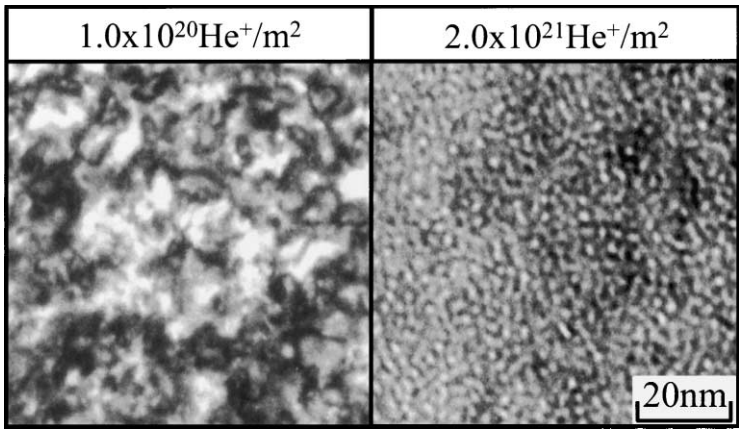

Fig. 5. Transmission-electron micrograph of W irradiated with 8 keV-He^+ to 1×10^{20} and 2×10^{21} He^+/m^2 at room temperature.

Though it is known that retention of hydrogen isotopes in tungsten is low in general, and this is one of the advantage of this material, the present results indicate that synergistic effects of helium bombardment must be taken into account to understand and to evaluate the behavior of hydrogen isotopes in plasma facing materials used in fusion environment.

5. Conclusions

TDS technique was used to study the trapping of deuterium atoms in tungsten pre-irradiated with helium ions. Pre-irradiation with helium ions caused remarkable effects on the trapping of injected deuterium. The strong trapping sites were shown to be related to the helium bubbles. Possible trapping sites for the desorption above 600 K is the strong stress field around the high-pressure helium bubbles. In the case where the helium bubbles were noticeably formed, rather weak but dense deuterium trapping sites were also formed. As a result, total amount of the retained deuterium increase drastically. Possibly dislocation loops punched by helium bubbles are the responsible trapping sites.

The present result indicates that irradiation effects of He must be taken into account to understand and to evaluate the behavior of hydrogen isotopes in fusion environment.

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