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Characteristic changes of deuterium retention on tungsten surfaces due to low-energy helium plasma pre-exposure

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Abstract

Deuterium retention in tungsten increases substantially when it is pre-exposed to low-energy He plasma at a high temperature (1600 K), while it decreases when pre-exposed at a low temperature (700 K). The increase of deuterium retention could be attributed to the submicron-sized He bubbles and secondary interstitial defects formed on the tungsten surface. Occupation of trap sites by He atoms during the He pre-exposure at low temperature could decreases the effective number of available trap sites for deuterium atoms, which would decreases the deuterium retention on the tungsten surface.

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1. Introduction

Plasma-facing materials in a fusion reactor, like ITER, will be exposed to plasmas with hydrogen isotope and helium (He) ions. Since the divertor plate will be exposed to high heat flux plasmas, tungsten (W) is considered to be one of the important candidates for the divertor material due to its good physical and thermal properties at high temperatures. It has been assumed that ion-induced displacement and physical sputtering do not occur on the W surface with low incident ion energies (<100 eV). However, our previous experiments have shown that damage (such as hole structure) could occur to the W surface by He plasma exposure at high temperature (>1500 K) even though the incident ion energy is below 100 eV [1,2]. The temperature of the divertor plate in ITER could increase to 1500 K or more in the case of repetitive ELMs or transient attached plasma [3]. Hole structure due to He plasma (He ash) exposure could form on the W surface in those cases. The material property, such as hydrogen isotope retention, may

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change drastically with hole structure. Hydrogen isotope retention in tungsten irradiated with ion beams has been investigated in many studies [4–9]. Some of them have shown that D retention in W is affected by He ion pre-irradiation [4–6,8]. The ion energies in these experiments were high (>1 keV), enough to induce displacement in the material, which is considered to be one of the reasons for the characteristic change of D retention. However, there has been no study that concentrates on the influence of the hole structure formation on D retention on W surfaces. In addition, it is very interesting to see how the D retention property of W changes in the case of low-energy and high-flux He plasma pre-exposure, which, while not causing displacement in the material, results in comparatively 3 orders of magnitude higher fluxes and fluences ($\sim 10^{21-22}$ ions/m^{2}s > 10^{25} ions/m^2) than is achievable with ion beam irradiations. The main objective in this study is to investigate the effect of hole structure in the W surface on the D retention property. An interesting result has also been obtained in the case of W even without hole structure.

2. Experimental setup

Three samples are used in this experiment. The samples are powder metallurgy tungsten (PM-W) provided by Nilaco Co. with 99.95% purity and 0.2 mm in thickness. Although as-received samples were cleaned with ultrasonic conditioning before setting up, no pre-treatments, such as annealing and polishing, were done on them. The sample W1 is a virgin sample free from He plasma pre-exposure. He plasma pre-exposure with incident ion energy of 20 eV at low temperature (700 K) was carried out on sample W2 in the linear divertor plasma simulator NAGDIS-I [10] and another sample, W3, was also pre-exposed to a He plasma with incident ion energy of 25 eV at high temperature (1600 K) in NAGDIS-II [11]. He fluences of these two samples were 1.8×10^{26} m^{-2} for W2 and 9.0×10^{25} m^{-2} for W3, respectively. Deuterium plasma exposures with incident ion energy of 80 eV were performed on the samples W1, W2 and W3 under similar temperature (550 K) and fluence (3.0×10^{25} m^{-2}) conditions in NAGDIS-I. The samples were fixed on a water-cooled target stage. The temperature of each sample was measured by a K type thermocouple in contact with the back surface of each sample. Other experimental details are given in Table 1.

After plasma exposures, the samples were analyzed by thermal desorption spectroscopy (TDS) at the Research Institute for Applied Mechanics, Kyushu University. The temperatures of the samples were increased up to 1473 K with a ramping rate of 1 K/s. The released D_2 and He were detected separately. Detail of the TDS device are described elsewhere [8].

Table 1
Experimental parameters of W1, W2 and W3

	Sample name	W1	W2	W3
Pre-He	Surface temperature [K]	–	700	1600
	Fluence [m^{-2}]	–	1.8E+25	9.0E+25
	Ion flux [m^{-2}s^{-1}]	–	2.5E+21	4.8E+22
	Exposure time [s]	–	7200	1800
	Ion energy [eV]	–	20	25
	Hole formation	–	No	Formed
D	Surface temperature [K]		550 ± 30	
	Fluence [m^{-2}]		3.0E+25	
	Ion flux [m^{-2}s^{-1}]		4.0E+21	
	Exposure time [s]		7200	
	Ion energy [eV]		80	
	Blister formation		No blister formation	

3. Experimental result

Scanning electron microscopy (SEM) analysis was performed on each sample. Fig. 1 shows the surface condition of virgin sample (a) and the surface morphologies of the samples W2 (b) and W3 (c) after He plasma pre-exposures. No hole structure is observed on W2 after He pre-exposure at 700 K, while it is clearly evident on W3. The surface condition of W2 seems to be smoother than that of the virgin sample. SEM analysis showed no additional surface modification on these three samples after D plasma exposures.

Fig. 2(a) shows the TDS spectra of D from the three samples after D plasma exposures. The background spectrum is also shown in the figure. The background desorption levels are not included in the total desorption fluences of the samples. A peak emission exists at 700 K and a small shoulder is observed at 500 K in W1, which was not pre-exposed to the He plasma. The total desorption fluence (or retention) of W1 is $R_{\mathrm{D2W1}} = 9.2 \times 10^{19}$ m^{-2}. Desorption occurs over a broad temperature range, from room temperature (RT) to 1150 K with appreciable emission. Sample W2 which was pre-exposed to a He plasma at 700 K has three peaks at 450 K, 680 K and 850 K. The total desorption fluence is $R_{\mathrm{D2W2}} = 2.0 \times 10^{19}$ m^{-2}, which is only about one-fifth of that of W1. For sample W3, which was pre-exposed to a He plasma at 1600 K and exhibited a hole structure on its surface, two peaks are observed at temperatures of 550 K and 800 K. The total D retention is $R_{\mathrm{D2W3}} = 5.1 \times 10^{20}$ m^{-2}, which is about six times as large as that of W1. In this case, the dominant desorption peak is at 550 K.

4. Discussion

Iwakiri et al. indicated that He bubbles and dislocation loops punched by He bubbles provide trap sites

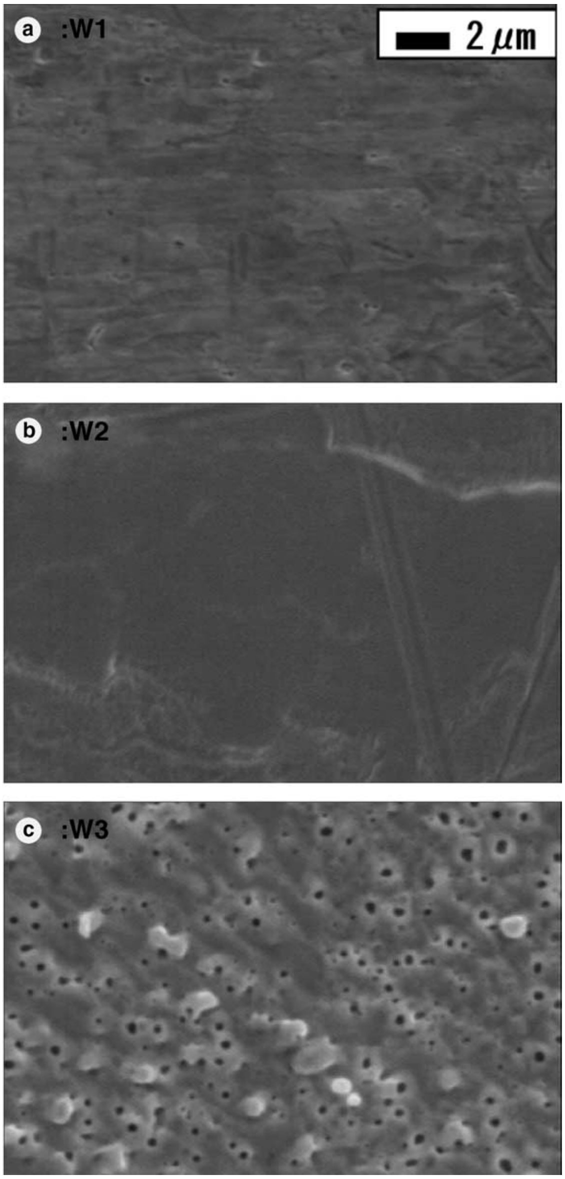

Fig. 1. SEM photographs of tungsten samples. (a) W1: virgin sample, (b) W2 after He pre-exposure at 700 K and (c) W3 after He pre-exposure at 1600 K.

for D atoms [8]. Fig. 3 shows the cross-section of a W sample exposed to a He plasma with the same conditions as W3, and which has the same scale hole structure on its surface as was observed on W3. Submicron-sized He bubbles are seen within a depth of 1 μm from the top surface. D atoms/molecules could be trapped inside these large He bubbles. An increase of D retention on W3 could be attributed to submicron-sized He bubbles and to secondary interstitial defects surrounding He bubbles.

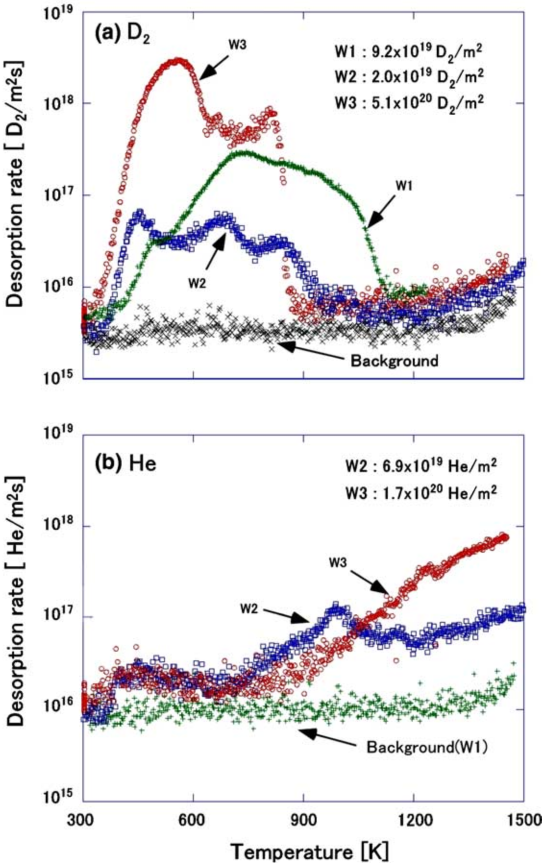

Fig. 2. Thermal desorption spectra of (a) deuterium and (b) helium for the tungsten samples pre-exposed by He plasma (W2 and W3) with temperature of 700 K and 1600 K and the sample without He pre-exposure (W1).

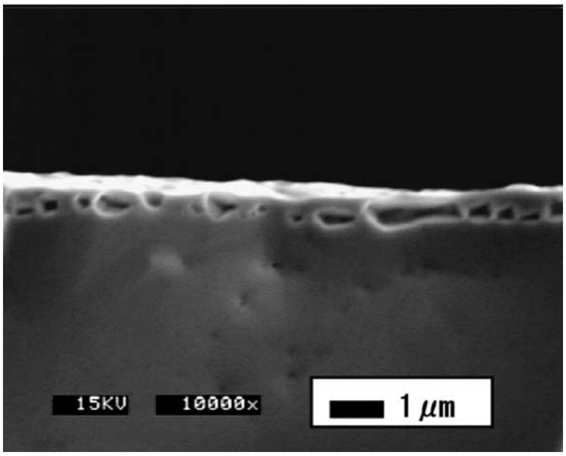

Fig. 3. SEM photographs of the cross-section of a tungsten sample exposed to He plasma with the same conditions as W3.

Although there are intrinsic defects such as vacancies and voids in W2, additional defects and large He bubbles as observed on W3 are believed to be not formed by He plasma pre-exposure at a surface temperature of 700 K. The thermal vacancy density of W and the mobility of vacancies and voids in W, which are considered to be essential factors for He bubble growth [2], are negligible at that temperature. Radiation-induced vacancies are also not expected to be formed during He pre-exposures because of the low incident ion energy (<25 eV). This means that the density of intrinsic defects in W2 after He pre-exposure is at a level similar to that see in the virgin sample. However, some trap sites such as strain field may be removed at a temperature of 700 K, although intrinsic vacancies and voids may be more difficult to remove at this temperature. The annealing effect during He pre-exposure at 700 K may decrease the number of interstitial trap sites, partially contributing to the observed decrease of D retention on W2. In addition, some trap sites (e.g., vacancies and grain boundaries), may be occupied by He atoms during He pre-exposure and few trapped He atoms are released during the subsequent D plasma exposure at the relatively low temperature of 550 K. This He occupation of trap sites can lead to a decrease in the effective number of trap sites available for D atoms, and as a result, the D retention of W2 becomes smaller than that of W1. The difference of D retention between W1 and W2 is $\Delta R_{\mathrm{D2}} = R_{\mathrm{D2W1}} - R_{\mathrm{D2W2}} = 7.2 \times 10^{19}\ \mathrm{D_2/m^{-2}}$, which corresponds to D atom retention of $\Delta R_{\mathrm{D1}} = 1.4 \times 10^{20}\ \mathrm{D/m^2}$. By comparison, He retention of W2 is $R_{\mathrm{HeW2}} = 6.9 \times 10^{19}\ \mathrm{He/m^2}$ (Fig. 2(b)), which is half of this difference of D atom retention i.e., $R_{\mathrm{HeW2}} \sim \Delta R_{\mathrm{D1}}/2$. However, not all the He atoms are expected to be released from the material because some He atoms could be deeply trapped in the material even at high temperature [12]. Supposing that the "annealing effect" is negligible for the decrease of trap sites, the total retention of He in W2, including He atoms remaining in the material, may be close to the difference of the D atom retention in W1 and W2.

5. Summary

Deuterium (D) and helium (He) gas retention of powder metallurgy tungsten (W) samples after low-energy He plasma pre-exposure and subsequent D plasma exposure were studied by using thermal desorption spectroscopy (TDS) techniques. All experiments were performed with incident ion (both He and D) energies of less than 80 eV, which means that the W samples were free from either radiation-induced defects or physical sputtering.

For the sample, on which many holes and submicron-seized He bubbles were formed during He plasma pre-exposure at 1600 K, the subsequent D plasma exposure at 550 K resulted in a total D retention which was ~6 times higher than that observed for the virgin sample (without He plasma pre-exposure); also, the maximum desorption peak occurred at a lower temperature compared to that of the virgin sample. Submicron-sized He bubble formation in the material and secondary interstitial defects surrounding the He bubbles that could be trap sites for D atoms are considered to be the main causes for the increased amount of D retention.

The total D retention of the sample pre-exposed to a He plasma at 700 K, and then exposed to a D plasma at 550 K, was about one-fifth of that seen for the virgin sample for the same D exposure conditions. No additional trap sites were introduced in the sample because neither radiation-induced defects nor thermal vacancies are formed in the sample exposed to He plasma at 700 K. The annealing effect and/or the occupation of trap sites by He atoms would decrease the number of available trap sites for D atoms and as a result, D retention decreases for the sample pre-exposed to He at 700 K.

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