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Blister formation on tungsten surface under low energy and high flux hydrogen plasma irradiation in NAGDIS-I

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Abstract

This study presents experimental results on hydrogen blister formation on powder metallurgy tungsten (PM-W) surface under low energy (<100 eV) and high ion flux (>10^{21} $m^{-2}\,s^{-1}$) hydrogen plasma irradiation in a divertor plasma simulator-NAGDIS-I. The tungsten samples were exposed to steady-state hydrogen plasma at various sample temperatures and fluences. Hydrogen blister formations are clearly observed on tungsten surface at the surface temperature below 950 K. The blister size is from a few 10 μm to a few 100 μm. It was found that the blister formations obviously depend on the surface temperature and the incident hydrogen ion fluence. No blisters are observed on tungsten surface at the surface temperature above 950 K even if the fluence is high enough.

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Keywords: Plasma–surface interaction; Divertor plasma simulator; Hydrogen plasma; Tungsten; Bubble and blister formation

1. Introduction

Tungsten has been considered to be a plasma facing material in the divertor and baffle regions of ITER because of its high threshold energy for sputtering, low effective sputtering yield, good thermomechanical properties and high melting point [1,2]. Recently, the experimental results in ASDEX Upgrade divertor tokamak with large area tungsten walls indicate there is no negative influence on the plasma performance except for ITB limiter discharges [3]. Therefore, systematic investigations on tungsten material become more important for the design and future operation of ITER. Most of previous investigations of tungsten material were focused on the use of ion beams at a very high energy range of 1–300 keV and relatively low particle flux. However, plasma facing surfaces are subjected to a particle flux up to 10^{24} $m^{-2}\,s^{-1}$ and the bombarding ions will have a broad energy distribution down to a very low energy of several tens eV in divertor region of ITER. It is thus significant to investigate plasma–tungsten interactions in a divertor plasma simulator [4–7].

We have already reported about helium bubble and hole formation on the tungsten surface irradiated by low ion energy (<50 eV) and high ion flux (>10^{22} $m^{-2}\,s^{-1}$) steady-state helium plasmas at various surface temperatures in NAGDIS-I divertor plasma simulator [4,5]. One of the key issues in future fusion reactor operation is hydrogen isotopes retention in tungsten. The present study represents experimental results on hydrogen blister formation on tungsten surface under low energy (<100 eV) and high ion flux (>10^{21} $m^{-2}\,s^{-1}$) hydrogen plasma irradiation in the linear plasma device Nagoya University Divertor Simulator (NAGDIS-I). It is reported that the blister formations obviously depend on the surface temperature and the incident hydrogen ion fluence.

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2. Experimental setup

The experiments have been performed in NAGDIS-I, in which a high heat flux steady-state plasma is generated by Philips ionization gauge discharge with helium, hydrogen and argon as working gases [8]. The plasma is confined by means of an axial magnetic field up to 0.15 T with a large diameter of 12 cm. The hydrogen plasma density reaches 1×10^{18} m^{-3} in steady-state. The electron temperature measured with a Langmuir probe was 5–10 eV, but the ion temperature is assumed to be much lower than the electron temperature. Therefore, the incident energy of ions is determined by the potential difference between the plasma and the tungsten sample.

In every irradiation experiment, six pure tungsten samples ($25 \times 25 \times 0.2$ mm made by powder metallurgy) were installed on a tantalum plate which was mounted on a movable stage and positioned normally to the magnetic field. The samples were treated by means of ultrasonic cleaning in water before mounting. The tungsten sample is about 2 m away from the plasma source. The diameter of the plasma was limited to 2.5 cm by a tungsten limiter at about 10 cm upstream from the tungsten sample. The sample surface temperature was controlled by means of varying the plasma density. The ion flux was evaluated from saturated ion current of sample. The incident fluences were changed by variation of the irradiation time with a constant ion flux. The tungsten sample surface temperature was monitored through a vacuum window with an infrared thermometer in the wavelength range of 0.8–1.1 μm. Plasma parameters at one cross-section were measured by a reciprocating fast scanning Langmuir probe set about 0.2 m upstream from the tungsten target. Surface analysis was made later by means of scanning electron microscopy (SEM), and the desorption rate of hydrogen gas was measured by means of thermal desorption spectrometry (TDS).

3. Results and discussions

3.1. Blister formation

We fixed the plasma parameters and the sample temperature (920 K), while the incident fluences were changed by varying the irradiation time. The biasing voltage of the sample was −100 V with respect to the vacuum chamber. Fig. 1 shows SEM pictures of the tungsten surface after the hydrogen plasma irradiation with three different incident fluences of 1.4×10^{25}, 2.8×10^{25} and 4.7×10^{25} m^{-2}. The corresponding constant ion flux was 2.0×10^{21} $m^{-2}\,s^{-1}$, and the ion energy ~90 eV. Hydrogen blister formation started to grow at fluences of 1.4×10^{25} m^{-2}, as shown in Fig. 1(a). Size and density of blisters increase with increase in fluence by comparing Fig. 1(a) with (b). The size is developed up to ~300 μm. However, as further increase in fluence as shown in Fig. 1(c), it is found that the density of smaller blisters decreases, and that the average size of blisters increases comparing with Fig. 1(b). For a higher temperature (1150 K), the ion flux was kept at 5×10^{21} $m^{-2}\,s^{-1}$, the ion energy at ~80 eV, no large blister formations were found on the tungsten surface even at the fluence of 2.4×10^{25} m^{-2}, as shown in Fig. 2. A series of

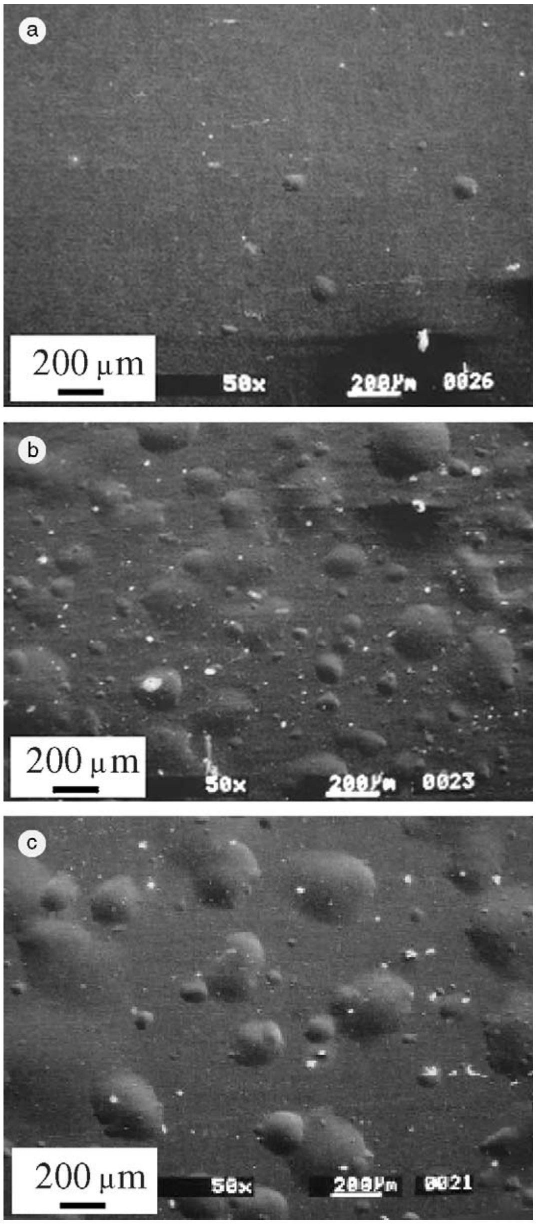

Fig. 1. SEM images of tungsten surface after the hydrogen plasma irradiation at 920 K for an incident fluence of (a) 1.4×10^{25} m^{-2}, (b) 2.8×10^{25} m^{-2} and (c) 4.7×10^{25} m^{-2}.

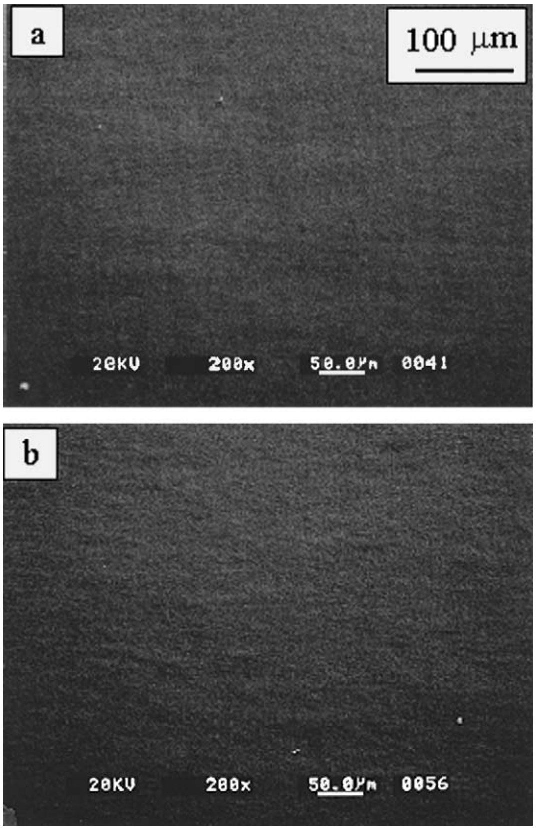

Fig. 2. SEM images of tungsten surface after the hydrogen plasma irradiation at 1150 K for an incident fluence of (a) 6×10^{24} m^{-2} and (b) 2.4×10^{25} m^{-2}.

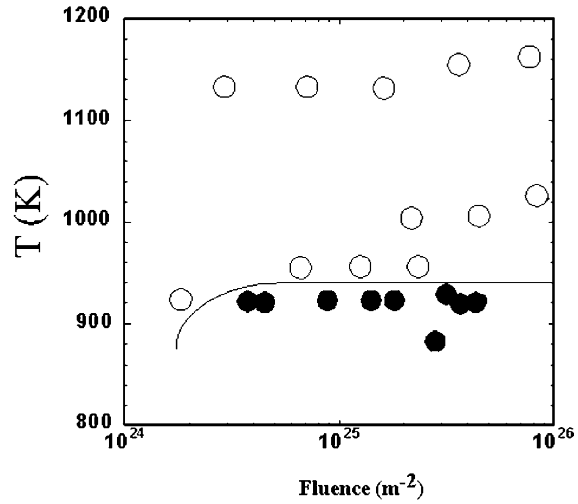

Fig. 3. Dependence of hydrogen blister formation on sample temperature and incident fluence. The symbols ● and ○ represent blister formation and no bilster formation observed in experiments, respectively. The solid line shows the boundary between blister formation region and no blister region.

experiments was carried out for different sample temperatures and fluences. Fig. 3 shows a plot of the dependence of blister formation on sample temperature and incident fluence. The area below the line in Fig. 3 is hydrogen blister formation region, and in the area above the line in Fig. 3 no blister formations occur. It can be clearly seen that as long as fluence is high enough ($> 3 \times 10^{24}$ m^{-2} in Fig. 3) blister formation on the tungsten surface were always observed in the low sample temperature region (<950 K in Fig. 3). No blister was found in W surface in a high sample temperature region.

Although the mechanism in blister formation on metal surfaces was discussed in early studies [9], and at low energy (∼100 eV) deuterium ion irradiation was discussed in recent studies [6,10], the formation mechanism is not still yet fully understood. Especially at low energy (<100 eV) ion irradiation, the trapping sites are not likely produced by the ion bombardment. The implanted H will diffuse in all directions in W, and are trapped dominantly at intrinsic vacancies as well as at grain boundaries. H forms gas bubbles in W, where the bubbles grow by ejecting W atoms from their lattice site (loop punching [9]) as more hydrogen atoms reach the sites. When gas bubbles are formed in W, they finally lead to such large blister formation on W surface due to high gas pressure as shown in Fig. 1.

Fig. 1(c) shows that the blister density does not increase as further increase in the fluence by comparing it with Fig. 1(b). During plasma exposure an equilibrium state is reached when the incident hydrogen equals the sum of hydrogen leaving the surface and diffusing into the material, the defect population in W is a key factor for bubble formation. For our case (Fig. 1), the constant ion flux to the surface acts as a constant source, the fixed surface temperature determined the diffusion rate and the defect population is not likely to be increased by the low energy ion bombardment. Therefore, when the incident fluence exceeds a critical value, the bubble formation arrives at a saturation state. The number of the blister shown in Fig. 1(c) seems to slightly decrease because the smaller bubbles combine into the bigger bubbles by a mechanism of such as inter-bubble fracture [11]. It is clear that such blister formation has obviously a critical temperature (∼950 K) as shown in Fig. 3. Although the diffusion rate increases at the high sample temperature, escaping of the implanted hydrogen from the surface becomes dominating owing to large thermal mobility, and finally no blister formations occur in W surface.

Comparing our experimental results with those reported in the article [10], in which it is reported that the blister size are mainly concentrated on 0.4 μm for 100 eV D^+ ion beam exposure. The blister size increases to 0.6–9 μm and 3–75 μm for the bombardment at 200 eV and 1 keV, respectively. However, our experimental results show the size of formed blister in W at energy about 100 eV is very much large up to ~300 μm. A possible explanation is that our experiments were performed at relatively high flux density ($>10^{21}$ $m^{-2}s^{-1}$) compared with their experiments ($<10^{20}$ $m^{-2}s^{-1}$).

Helium gas bubble formations on tungsten under a low energy helium plasma irradiation has been discussed in our previous publication [4,5]. Fig. 4 shows a plot of the dependence of bubble formation in the parameter space of sample temperature and incident fluence. Helium bubble formation occurs at high sample temperature (>1000 K), as shown in Fig. 4. Comparing hydrogen case with helium one, the temperature dependence is opposite each other. A possible reason for this phenomena is discussed as follows, at the higher temperature, escaping of hydrogen from the surface increases and the traps are depopulated due to large thermal mobility and a low binding energy to trapping site (1.4 eV [12]) so that no bubbles formation occurs when the temperature is high enough (>950 K in Fig. 3). However, for helium case, the high temperature (>1000 K in Fig. 4) increases the diffusion rate of helium in W so that it results in stable helium bubbles formation because of a higher binding energy for trapping site (4.75 eV [13]). At the low surface temperature, the diffusion rate of He in W is too low to form bubbles. Large blister formation in W surface at the low temperature for hydrogen case indicates that hydrogen has a relatively good diffusion in W comparing with helium. But a simple comparison of the calculated diffusion rate of helium from the Arrhenius expression [14] and that of hydrogen from the Frauenfelder expression [12] indicates that the diffusion rate of hydrogen is lower than that of helium in W. Therefore, some enhanced diffusivity of hydrogen in W might be required to explain hydrogen bubble formation at the lower temperature.

It is necessary to point out that hydrogen behavior in W is much influenced by surface impurities. For example, Poon et al. reported that carbon and oxygen impurities appear to affect the deuterium trapping mechanism and the amount of deuterium retained [15]. We did not measure impurities during plasma exposure, but we think that oxygen and carbon as main impurities in NAGDIS-I plasma should be much small. After plasma exposure, although surface analyses of the samples done by EDAX have shown only W in surface of the exposed samples, carbon and oxygen which are not detectable by EDAX. A possible influence from surface impurities in W (e.g. C and O) may exit in our experiments, making a role as trapping sites. Sputtering by impurities might be negligible at the low energy (<100 eV).

3.2. Thermal desorption spectrometry results

TDS measurements were carried out for some irradiated samples. The sample was heated by means of infrared heating method. The sample temperature was increased gradually as 1 K/s. TDS data were evaluated by subtracting a TDS data of an unexposed W sample from those of an exposed W sample. Fig. 5 shows desorption rate of hydrogen gas from one of tungsten samples after hydrogen plasma irradiation with the fluence of 3.7×10^{25} m^{-2}, the ion energy ~96 eV, and the sample temperature 630 K. The result indicates that a peak of hydrogen gas desorption is about 8×10^{18} $m^{-2}s^{-1}$ at the sample

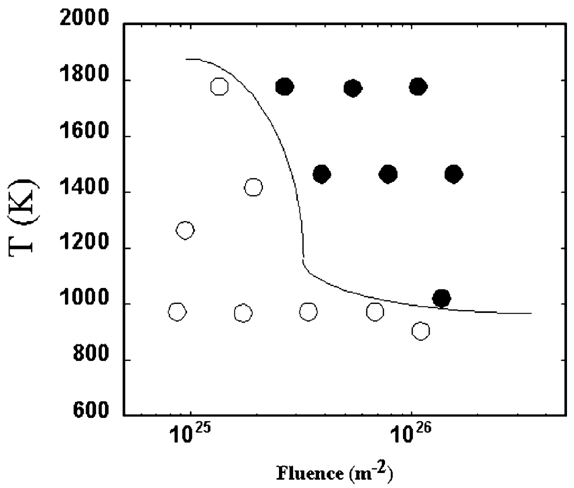

Fig. 4. Dependence of helium bubble formation on sample temperature and incident fluence. The symbols ●, ○ and the solid line have the same meaning as Fig. 3.

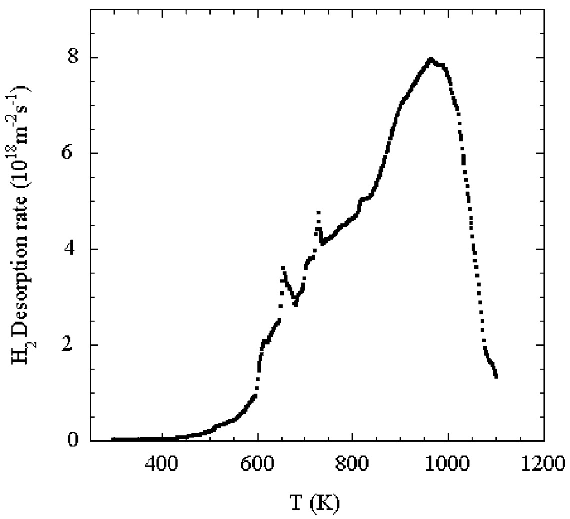

Fig. 5. The desorption rate of hydrogen gas from tungsten sample after hydrogen plasma irradiation with the fluence of 3.7×10^{25} m^{-2}, the ion energy ~96 eV, and sample temperature 630 K.

temperature of 950–1000 K. The quantity of the total absorption of hydrogen is about 2.5×10^{21} m^{-2}, which agree with the results reported in the article [6]. The peak temperature position is in good agreement with the sample temperature threshold of blister formation in Fig. 3.

4. Summary

Blister formation is clearly observed on tungsten surface under a low energy (<100 eV) and high flux (>10^{21} m^{-2} s^{-1}) hydrogen plasma irradiation at the surface temperature less than 950 K and the fluence higher than 3×10^{24} m^{-2}. The size of maximum hydrogen blisters is up to ~300 μm at higher fluence irradiation. The dependence of blister formation on sample temperature is clearly obtained. When the surface temperature is higher than 950 K no blisters are observed on tungsten surface at various incident fluences. These results indicate that such large hydrogen blister formation due to low energy particles irradiation at low temperature can also make a great effect on the hydrogen retention in fusion devices and reactors. For future work, it will be investigated to consider the effect of impurities in the next step.

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