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Deuterium retention, blistering and local melting at tungsten exposed to high-fluence deuterium plasma

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ABSTRACT

Blistering, deuterium retention and local melting at tungsten exposed to high-fluence (up to 10^{27} D/m^2) of high-flux (10^{22} D$^+$/m^2/s) and low-energy (38 eV) deuterium plasma at varying temperature were examined with scanning electron microscopy (SEM), focused ion beam (FIB), thermal desorption spectroscopy (TDS) and electron probe microanalysis (EPMA). Blisters with various shapes were observed after the plasma exposure, and both deuterium retention and blistering showed a significant dependence upon the exposure temperature and fluence. Local melting and flakes on some grains were observed even if the heat flux is less than 0.1 MW/m^2, suggesting that the local melting is caused by flakes due to the loss of heat conduction between the flakes and the bulk during the plasma exposure.

1. Introduction

Tungsten has been selected as the plasma facing material (PFM) at ITER divertor baffles and dome. Additionally, before the D–T phase of the ITER operation, a full tungsten divertor may be deployed as a potential solution not only for avoiding high in-vessel tritium inventory due to the co-deposition of hydrogen isotopes with carbon but also for avoiding handling and processing large quantities of tritiated water produced by in-vessel oxygen baking. The major safety concern about the tungsten is dust production and dust will be produced not only by melting due to ELMs or disruption but by blistering even at steady operation as well. Hydrogen blistering will occur at tungsten surface, even if the ion energy of deuterium is too low to create displacement damage such as vacancies [1–3]. Blistering at tungsten may also result in high-Z impurity release into the core plasma, leading to instability of the plasma, even disruption. In addition, an increasing tritium inventory in the near surface region of PFCs may become a significant safety issue during the exchange of the divertor cassettes.

In this study, blistering and deuterium retention in tungsten exposed to high-fluence (up to 10^{27} D/m^2) of high-flux (10^{22} D$^+$/m^2/s) and low-energy (38 eV) deuterium plasma at varying temperature were examined with scanning electron microscopy (SEM), focused ion beam (FIB) and thermal desorption spectroscopy (TDS). In addition, local melting on some tungsten grains after the plasma exposure was investigated with SEM and electron probe microanalysis (EPMA).

2. Experimental

The tungsten samples used are the recrystallized tungsten with a purity of 99.99 wt% and principal impurities (in weight ppm) of Mo and Fe around 10, C and O less than 30. The samples were finally recrystallized at 2073 K after being cut and double-sided polished into samples of 10 × 10 × 2 mm. Each sample was cleaned in an acetone ultrasonic bath prior to placing into the deuterium plasma exposure chamber.

A linear plasma generator [1] was used in this study, and the plasma was ignited by an electron-emitting filament made of LaB_6 with a shape of a hollow dual-spiral and maintained by an arc discharge power supply applied between the filament (cathode) and the grounded chamber wall (anode), being assisted by the confinement coils. The water-cooled sample holder is isolated from the grounded chamber wall so that the sample can be negatively biased to adjust the energy of ions impinging onto the sample. The sample is passively heated by the plasma itself and the temperature rise was adjusted by inserting some mica plates of different thickness in total between the sample and the holder. The temperature is monitored using a type K thermocouple tightly pressing the rear of the sample. The ion species can be controlled by adjusting the operational parameters of the plasma generator. In this study, plasma beams highly enriched with a single species

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of D_2^+ to a ratio over 80% were obtained, and the main impurity in the plasma was oxygen with a concentration less than 1 ppm. The area of the sample exposed to the plasma was 9 mm in diameter, and the incident flux and energy were fixed at 10^{22} $D^+/m^2/s$ and 38 eV/D, respectively.

Blister formation at the surface of tungsten exposed to deuterium plasmas was observed by a SEM (Real Surface View Microscope, KEYENCE VE-9800) at a tilt angle of 45°. Cross-sectional samples of big blisters and lid-removed samples of small blisters were prepared by a FIB microsampling system (HITACHI FB-2000A) with a fine Ga^+ (20 keV) beam, and the cross-sectional images of large blisters and the internal images of lid-removed small blisters were subsequently obtained by the SEM at a tilt angle of 45°. TDS was used to examine the behavior of deuterium release from the tungsten samples after deuterium plasma exposure.

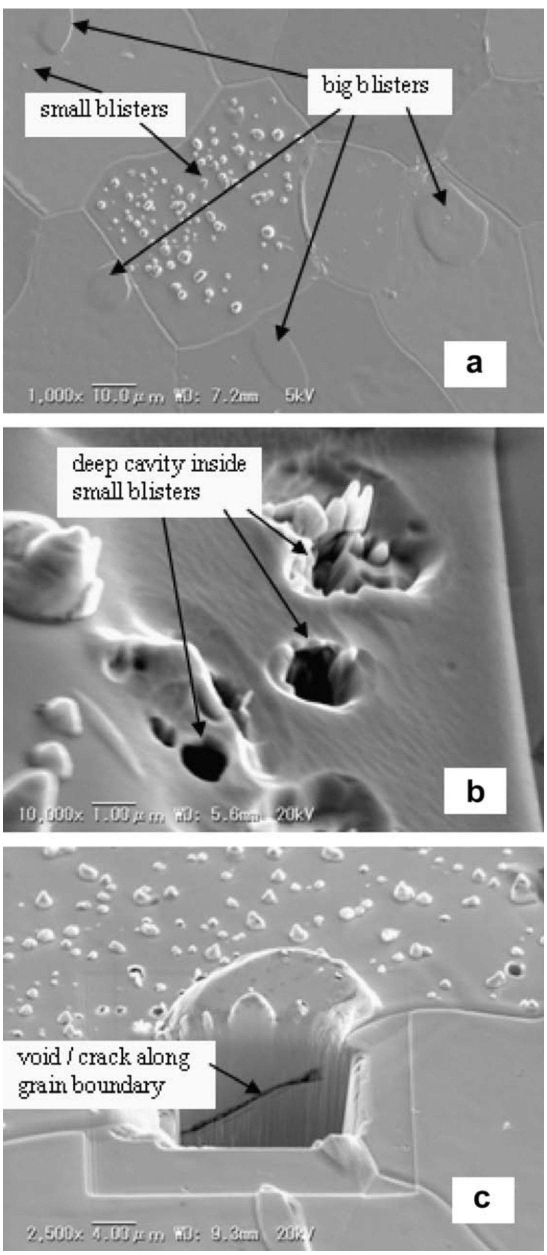

Fig. 1. Two kinds of blisters appearing on tungsten exposed to the fluence (10^{26} D/m^2) of high-flux (10^{22} $D^+/m^2/s$) and low-energy (38 eV) deuterium plasma at 480 K. (a) Surface morphology; (b) inside features of small blisters; and (c) cross-sectional image of a big blister.

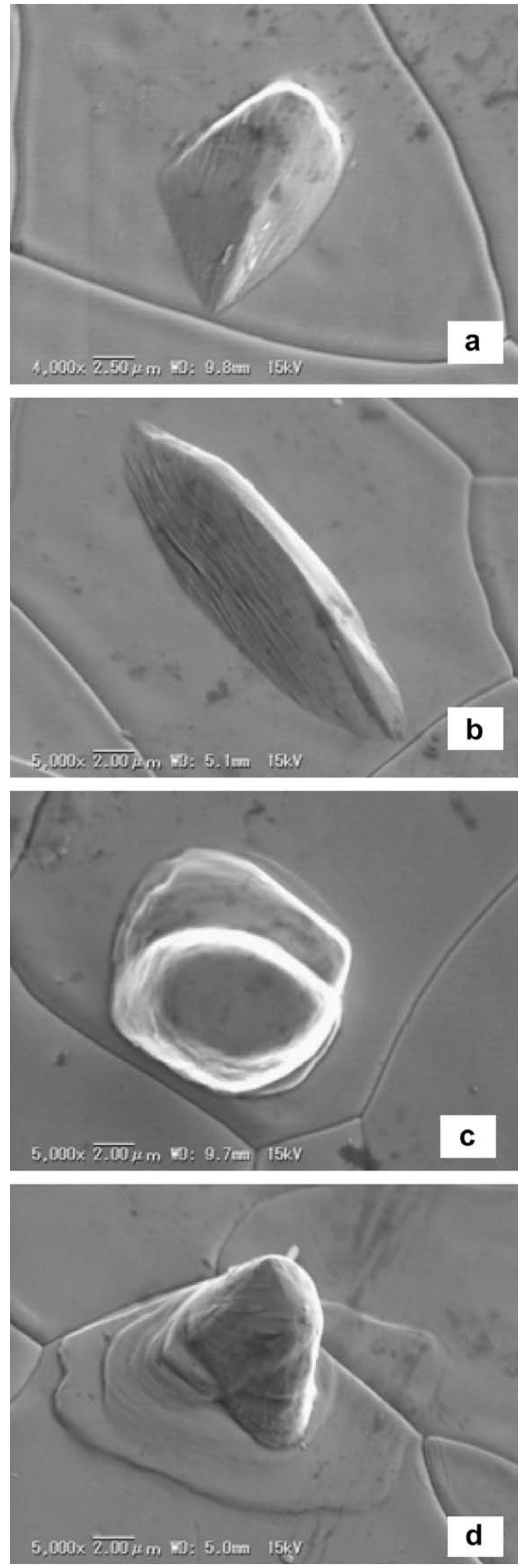

Fig. 2. Various shapes of big blisters appearing on tungsten exposed to the fluence of 10^{26} D/m^2 at 520 K. (a) a pyramid; (b) an upside-down boat; (c) a two-storied dome; and (d) a hill of cow dung.

A standard deuterium leak with an inaccuracy lower than 10% was employed to calibrate the quadrupole mass spectrometer prior to each TDS analysis so that the calibrated release rate during TDS could be obtained. During TDS, an infrared heater was used to heat the exposed samples at a ramp rate of 0.5 K/s and the sample temperature was raised to over 1000 K. Local melting on some tungsten grains after the plasma exposure was observed with the SEM, and then confirmed with an EPMA (JEOL JXA-8600MX) at the acceleration voltage of 15 kV. Four electron probe microanalysis attachments (NewSTE, PET, TAP, LIF) were used for compositional analysis on tungsten surfaces.

3. Results and discussion

3.1. Blistering and deuterium retention

Fig. 1 shows the SEM images of both the external looks and internal features of two kinds of blisters appeared on tungsten after deuterium plasma exposure at the fluence of 10^{26} D/m^2 and temperature of 480 K. One is a big blister with magnitude of greater than a few microns and various shapes, and the other is a small blister with size of less than a few microns and varying dome, as show in Fig. 1(a). Deep cavities (holes or pits) were formed in the internal blisters for all of the small blisters as indicated in Fig. 1(b), whereas there was no hollow lid formed but void/crack along the grain boundary beneath for most big blisters as seen from Fig. 1(c). This feature is contrary to the conventional definition, in which blisters are plastic dome-shaped buildings of the surface layer and a lenticular cavity is included between the blister lid and the bulk material [4].

The SEM images of some big blisters with various shapes are shown in Fig. 2 for tungsten exposed to deuterium plasma at the fluence of 10^{26} D/m^2 and temperature of 520 K. These blisters looked like a pyramid (a), an upside-down boat (b), a two-storied dome (c) and even a hill of cow dung (d), respectively. It seems that the shape of the big blister depends on the orientation and the shape of the grain itself. In addition, the blisters showed a multi-layered structure like steps, as shown clearly in Fig. 2(c) and (d). Almost all the blisters were limited by the grain boundaries and most of them appeared near the grain boundaries.

It has been considered that the high-dome blisters are formed by deuterium-induced local superplasticity [5]. First, deuterium-induced vacancies are generated due to at least the lowering of vacancy formation energy by trapping of deuterium [6,7]. Subsequently deuterium-vacancy clusters are formed and these deuterium-vacancy clusters diffuse deeply into the bulk such as somewhere near the surface in the grains and even grain boundaries (i.e. diffusion of tungsten atoms to the surface) and agglomerate, resulting in small blisters and big blisters, respectively. In addition, every agglomeration of the deuterium-vacancy clusters results in a step print on the blister.

Fig. 3 shows the TDS spectra of tungsten exposed to the fluence of 2×10^{26} D/m^2 at 400 K, in which bursting releases of deuterium were observed with sudden peaks due to bursts of blisters. The bursting releases as well as the high-temperature release peak (around 840 K) indicate that deuterium retains in tungsten mostly in the form of molecules at voids, corresponding to the formation of blisters. During TDS heating, deuterium releases directly when blisters burst or firstly dissociates from molecules into atoms (thus

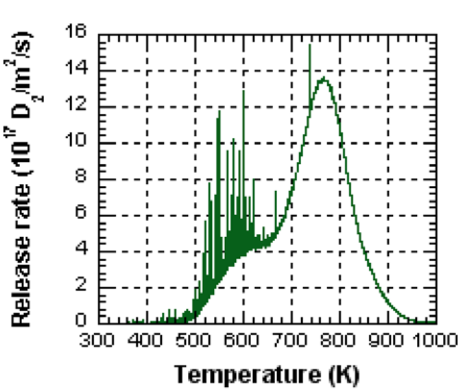

Fig. 3. TDS spectra of tungsten exposed to the fluence of 2×10^{26} D/m^2 at 400 K (heating rate: 0.5 K/s).

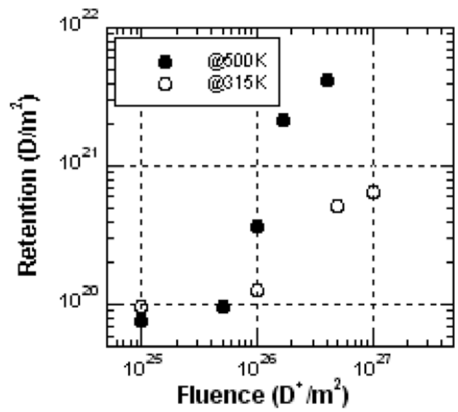

Fig. 4. Fluence dependence of deuterium retention in tungsten exposed at temperature of 500 K and 315 K.

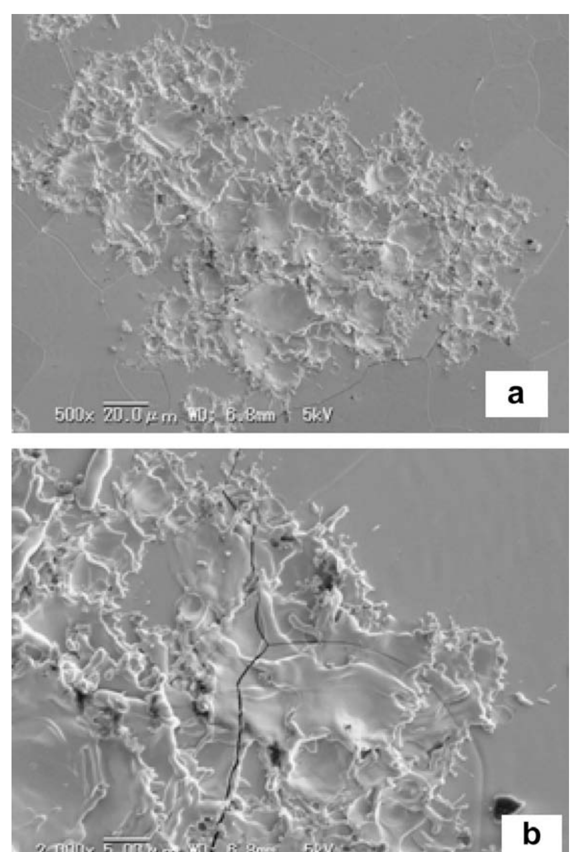

Fig. 5. SEM images of local melting on some grains of tungsten exposed to 10^{26} D/m^2 at 618 K. (a) Severe melting; and (b) micro-cracks along grain boundaries.

high-temperature is required) and then atoms diffuse to the surface where they recombine and release as molecules.

Both deuterium retention and blistering showed a significant dependence upon the exposure temperature and fluence. The fluence dependence of retention is shown in Fig. 4 for exposure temperature of 500 K and 315 K, respectively. The amount of deuterium retained in tungsten increased with the increasing fluece, but the relationship between the retention and the fluence is not a simple function. Especially, a rapid increase in the retention occurred at the fluence of 10^{26} D/m^2 for the samples exposed at 500, corresponding to the appearance of two kinds of high-dome blisters.

3.2. Local melting

During plasma disruption, a high heat flux will be deposited onto the plasma facing materials for a very shot time causing melting of the surface, and evaporation may take place depending on the intensity. In the tests of bulk tungsten 9 MW/m^2 had been applied for 15 s leading to surface temperature above 2773 K, resulting micro-cracks [8]. The impact of micro-cracks on the performance had seemed to be negligible for this degree of cyclic thermal loading, 7–8 MW/m^2 for 10 s up to 150 cycles, since no significant temperature increase had been observed [9]. In this study, however, local melting on some grains was observed, even if the heat flux is less than 0.1 MW/m^2. Fig. 5 shows the SEM images of local melting of tungsten exposed to 10^{26} D/m^2 at 618 K. Severe melting occurred on some grains, as seen from Fig. 5(a), and micro-cracks along grain boundaries were observed accompanying with the local melting, as indicated in Fig. 5(b).

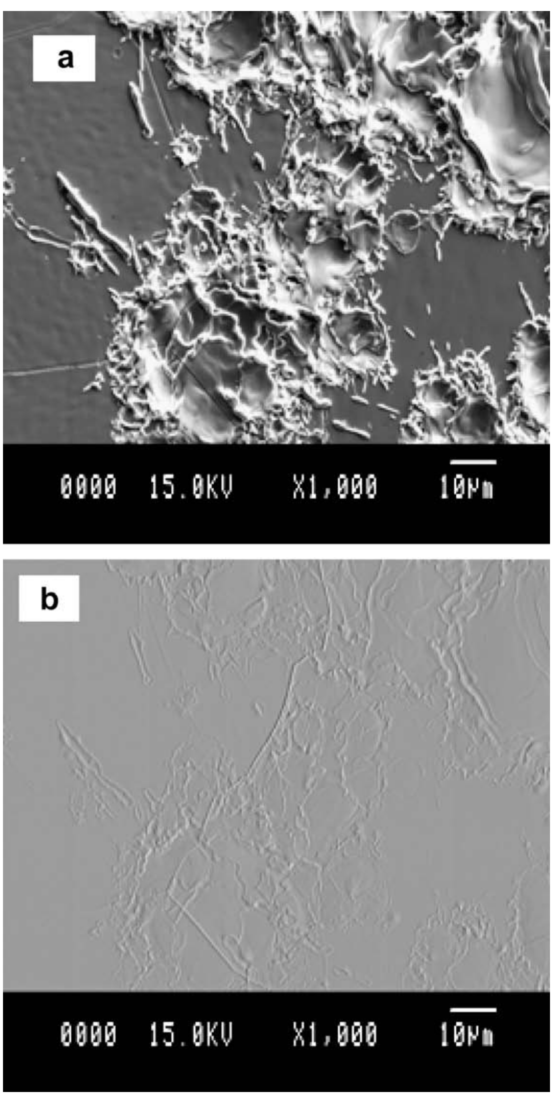

Fig. 6. EPMA images of local melting on some grains of tungsten exposed to 10^{26} D/m^2 at 618 K. (a) Secondary electron image; and (b) backscattered electron compositional image.

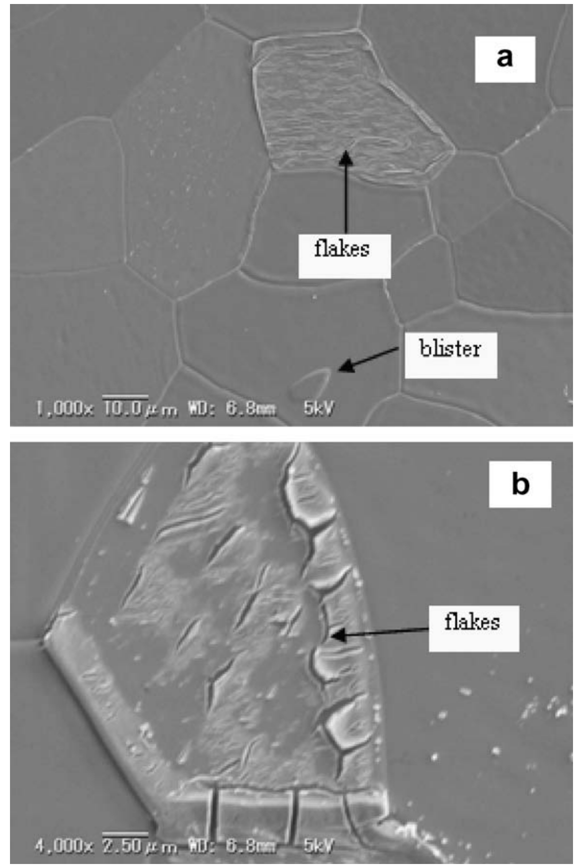

Fig. 7. SEM images of flakes on some grains of tungsten exposed to 10^{26} D/m^2 at 618 K. (a) A blister and flakes; and (b) flakes.

Why local melting occurred at the relatively low heat flux? We first analyzed the chemical composition of melted area with EPMA. Both the secondary electron image and the backscattered electron compositional image of a melted area are shown in Fig. 6(a) and (b), respectively. The EPMA results showed that the melted area is pure tungsten and even the intensity of carbon impurity is negligibly small. Subsequently the surface morphology of the sample with local melting was examined with SEM. Flakes on some grains were found, as shown in Fig. 7(a) and (b). It is considered here that local melting may occur when the flakes are exposed to the plasma due to the loss of heat conduction between the flakes and the bulk. The EPMA results showed that the flakes are also pure tungsten and the intensity of carbon impurity is negligibly small. The mechanisms of flake formation should be further investigated.

4. Conclusions

(1) Small blisters and big blisters with various shapes like a pyramid, an upside-down boat, a two-storied dome and even a hill of cow dung appeared on tungsten after the plasma exposure. It suggests that the shape of the big blister depends on the orientation and the shape of the grain itself.

(2) Though the amount of deuterium retained in tungsten increased with the increasing fluece, the relationship between the retention and the fluence is very complicated. A rapid increase in the retention occurred at the fluence of 10^{26} D/m^2 for the samples exposed at 500 K, corresponding to the appearance of two kinds of high-dome blisters.

(3) Local melting on some grains was observed, even if the heat flux is less than 0.1 MW/m^2. The EPMA results showed that the melted area is pure tungsten and even the intensity of carbon impurity is negligibly small. Flakes are considered to be the cause of local melting due to the loss of heat conduction between the flakes and the bulk during the plasma exposure.

Acknowledgements

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