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Recent findings on blistering and deuterium retention in tungsten exposed to high-fluence deuterium plasma

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ABSTRACT

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 were examined in the temperature range of 315–1000 K with scanning electron microscopy, focused ion beam, thermal desorption spectroscopy and positron annihilation. There were cavities inside small blisters with the maximum ratio of height against diameter of about 0.7, whereas there were voids/holes along the grain boundary beneath most large blisters but no hollow lid formed. Blistering and deuterium retention showed a significant dependence upon fluence and exposure temperature.

1. Introduction

An all tungsten armoured divertor will be adopted for ITER deuterium-tritium phase, because of the great concern of tritium inventory for the use of CFC [1]. However, there remain concerns over what will happen to the melt layer of tungsten target and what effect an uneven re-solidified surface might have on subsequent operations and target life-time. In addition, there is evidence of blistering occurred at tungsten surface even if the ion energy is too low to create displacement damage such as vacancies [2–7]. Tungsten blistering could lead to instability of the plasma due to high-Z impurity release into the core plasma and sudden gas recycling [5].

Up to now, we have observed blister bursting and deuterium bursting release from tungsten exposed to high fluences of high flux and low energy deuterium plasma [5], and investigated the microstructure dependence of deuterium retention and blistering [6] and confirmed with TEM observations that microcracks formed along the grain boundaries before the blister formation [7]. In this paper, the features of high-dome blisters as well as the dependences of blistering and deuterium retention upon exposure temperature and fluences are examined with scanning electron microscopy (SEM), focused ion beam (FIB), thermal desorption spectroscopy (TDS) and positron annihilation (PA).

2. Experimental

The samples used are tungsten recrystallized fully at 2073 K after cutting and polishing, 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 details of the samples and the linear plasma generator used in this study have been described in the previous papers [5–7]. 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 at a tilt angle of 45°. Cross-sectional samples of large blisters and lid-removed samples of small blisters were prepared by a FIB microsampling system. In addition, thermal desorption spectroscopy (TDS) with the time resolution of about 0.3 s was used to examine the behaviour of deuterium release at a ramp rate of 0.5 K/s. Furthermore, the defects in the sample were detected by positron annihilation spectroscopy and quantified by so-called *S* parameter, which is defined as the normalized peak area of the Doppler spectrum [6].

3. Results and discussion

3.1. Features of high-dome blisters

For the tungsten exposed to fluence of 10^{26} D/m^2 at around 500 K, two kinds of blisters namely large blisters and small blisters appeared. The former were greater than a few microns in size and shaped variously with varying dome, and the latter were less than a few microns in diameter and shaped with varying dome. Fig. 1 shows SEM images of the tungsten exposed to fluence of 10^{26} D/m^2 at the exposure temperature of 480 K. Fig. 1(a) shows the overall view of both kinds of blisters, and Fig. 1(b) and (c) show the SEM images after FIB fabrication for a large blister and small blisters, respectively. For most large blisters, there was no hole inside blisters but a hole/void at the grain boundary underneath, as shown in Fig. 1(b). This feature was confirmed by several SEM observations

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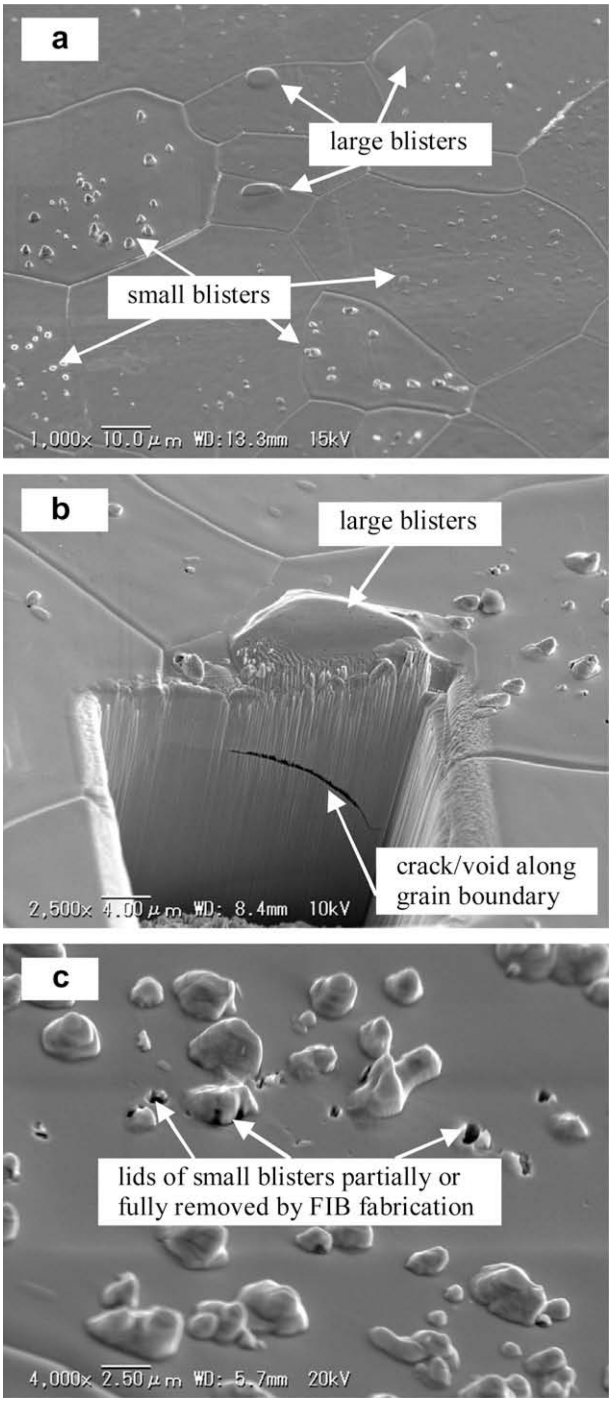

Fig. 1. SEM images of tungsten exposed to fluence of 10^{26} D/m^2 at 480 K (45° tilt). (a) An overall image; (b) cross-sectional image of a large blister; and (c) internal image of small blisters.

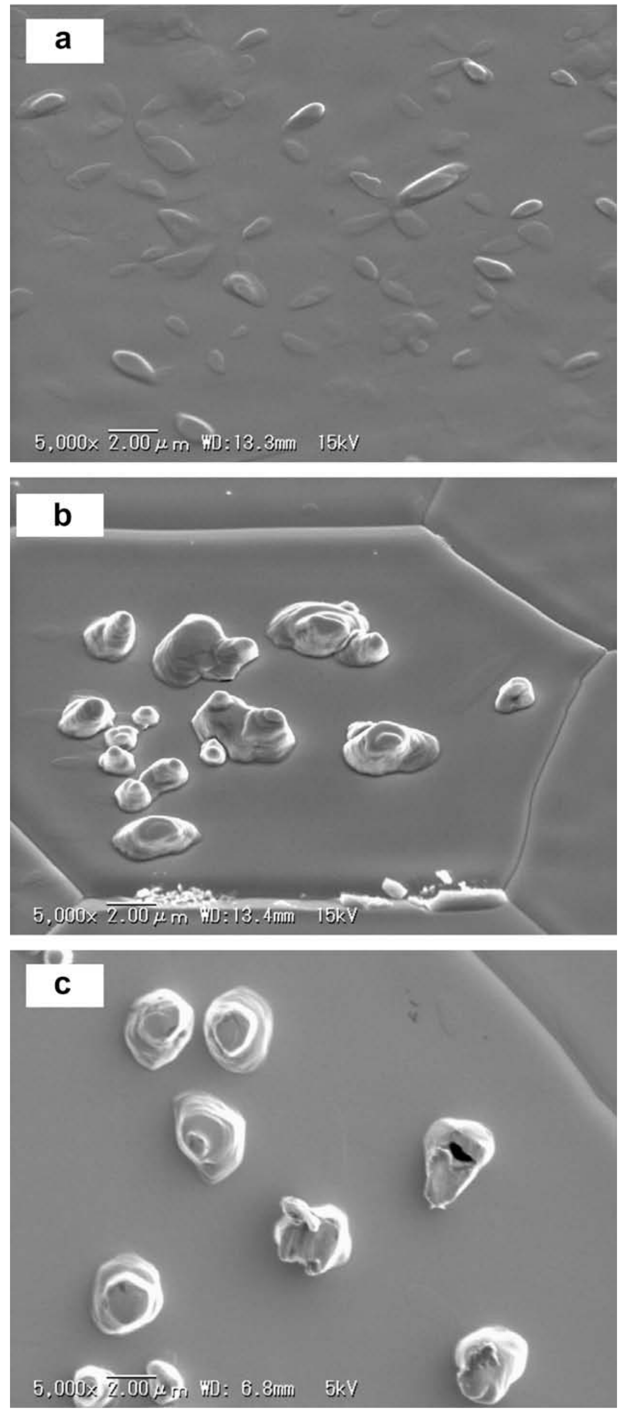

Fig. 2. SEM images of the small blisters appearing at tungsten exposed to the fluence of 10^{26} D/m^2 at 480 K (45° tilt). (a) An initial stage; (b) growing; and (c) bursting.

during the FIB fabrication, which is contrary to the conventional blister with hollow lid [8]. On the other hand, for the small blisters, internal blister was a hole or pit for all the small blisters, as shown in Fig. 1(c). The maximum ratio of height against diameter reached about 0.7. This ratio is about one order of magnitude greater than the typical blister reported before [8], in which the cavity between the surface layer and the bulk had been generally assumed to be initially of a lenticular shape (the ratio of blister height against inner chord was around 0.05).

Fig. 2 shows the growth of small blisters that appeared at tungsten exposed to fluence of 10^{26} D/m^2 at 480 K. In a certain early stage, four blisters with different heights of domes appeared closely like a flower, as seen from Fig. 2(a). Subsequently, the low-dome blisters disappeared, and one or two high-dome blisters grew among these four blisters, as shown in Fig. 2(b). Finally, some blisters burst with an opening, as indicated in Fig. 2(c).

Some large blisters are shown in Fig. 3 for tungsten exposed to fluence of 10^{26} D/m^2 at 520 K. First of all, these large blisters had various shapes and varying domes, as seen from Fig. 3(a). Secondly, the blisters showed a multi-layered structure like steps, as shown clearly in Fig. 3(b) and (c). Additionally there was no hollow lid

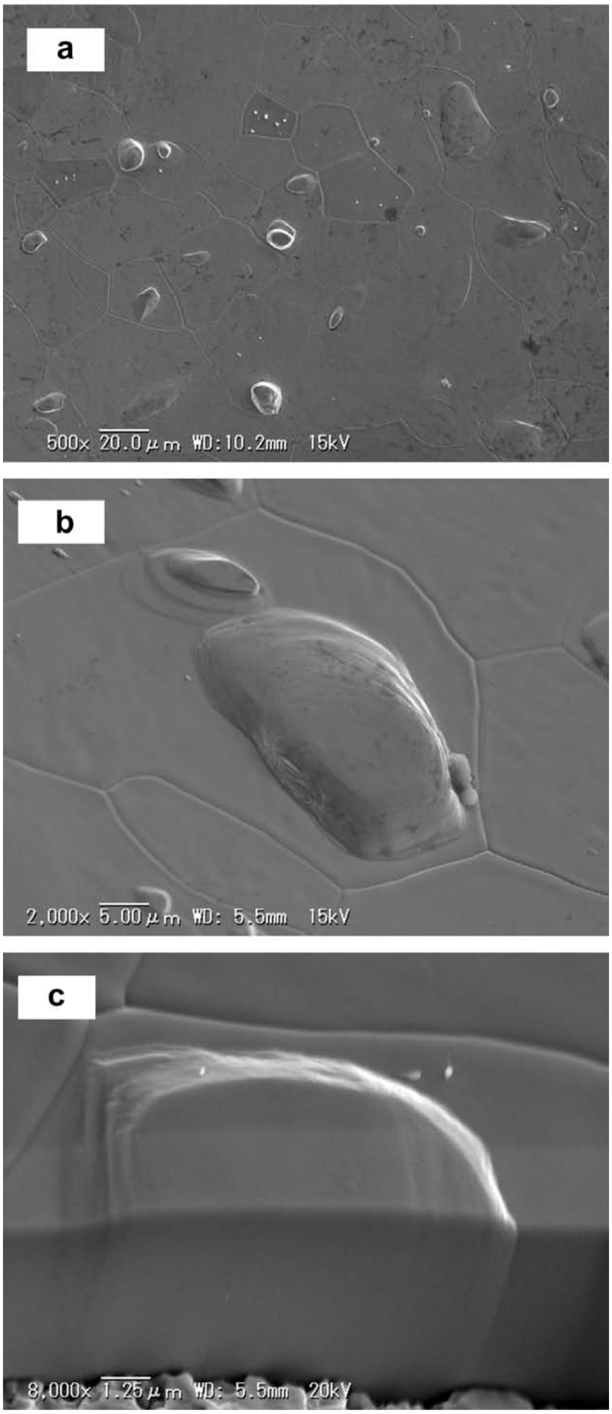

Fig. 3. SEM images of the large blisters appearing at tungsten exposed to the fluence of 10^{26} D/m^2 at 520 K (45° tilt). (a) An overall image; (b) growing step by step; and (c) cross-section of a blister.

formed inside most large blisters as indicated in Fig. 3(c). Finally, almost all the blisters were limited by the grain boundaries and most of them appeared near the grain boundaries. Since the ductile-to-brittle transition temperature (DBTT) for tungsten is around 1073 K [9], it is clear that the blisters cannot be generated by normal plasticity, namely the movement of dislocations.

How do blisters nucleate and grow during plasma exposure with energies well below the displacement threshold? In order to answer this question, the surface state after plasma exposure was examined by the PA measurements and compared with that prior to exposure. As shown in Fig. 4, the *S* parameter of tungsten exposed to 10^{26} D/m^2 at 520 K and TDS-treated to 1093 K was larger than that prior to plasma exposure, indicating that the vacancy concentration in the near-surface region of tungsten increased after the deuterium plasma exposure. From the analyses based on the one-dimensional diffusion equation of positron [10], a bulk layer was obtained for the unexposed sample, while a defect layer 296 nm deep was obtained for the sample after the plasma exposure and TDS treatment, resulting in a specific *S* parameter of 1.03 for the defect layer.

Fukai et al. [11] proposed a theory of formation of superabundant vacancies (hydrogen–vacancy clusters), where the formation energy of a vacancy was decreased by cluster formation and the configurational entropy of the system when metal was exposed to high-pressure hydrogen atmosphere at high temperature. This mechanism is considered here to be valid in the present case of tungsten exposed to high flux and low energy deuterium plasma. Blisters would be caused by the generation of the hydrogen-induced vacancies and subsequent formation and clustering of hydrogen and vacancies which diffuse deeply into the bulk such as grain boundaries or even somewhere in the grains (i.e. diffusion of tungsten atoms to the surface) and agglomerate.

3.2. Dependence of blistering and deuterium retention upon fluence and temperature

The dependence of blistering and deuterium retention upon fluence was examined for the tungsten exposed at around 500 K. At the fluence of 10^{25} D/m^2, only sparse low-dome blisters with chords of a few microns or less appeared. Even when the fluence was increased to 5×10^{25} D/m^2 the blisters were still small and sparse. A peculiar change occurred at the fluence of 10^{26} D/m^2, where both large blisters and small blisters appeared. As the fluence reached 4×10^{26} D/m^2, the small blisters became larger and their domes became higher. On the other hand, a peculiar phenomenon of bursting release of deuterium was found obviously in TDS spectra. As shown in Fig. 5, numerous bursting peaks appeared at a wide temperature range of 380–740 K for tungsten exposed to the fluence of 10^{26} D/m^2. The extreme point of deuterium release shifted from 720 K at the fluence of 10^{25} D/m^2 to 880 K at

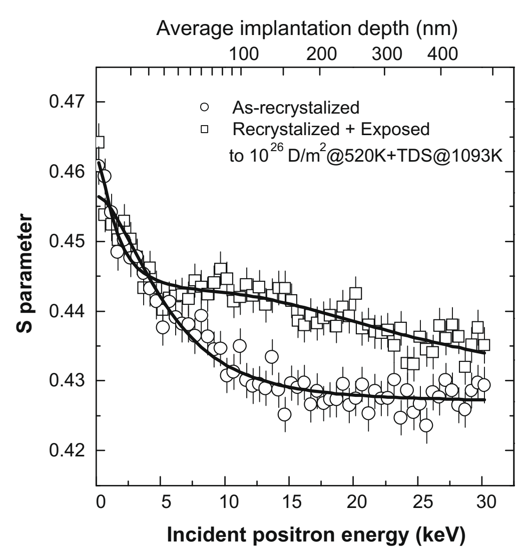

Fig. 4. *S* parameter of tungsten after the exposure to 10^{26} D/m^2 at 520 K and TDS treatment to 1093 K (open squares) as a function of incident positron energy in comparison with that of prior to plasma exposure (open circles). Solid lines indicate *S* parameters calculated based on the one-dimensional diffusion equation of positrons.

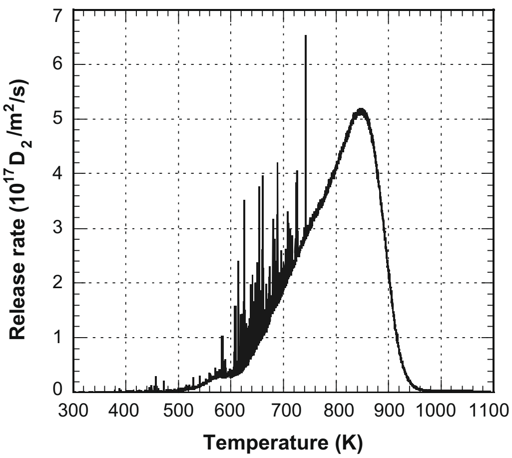

Fig. 5. TDS spectra of tungsten exposed to the fluence of 10^{26} D/m^2 at 500 K (heating rate: 0.5 K/s).

4×10^{26} D/m^2, suggesting a deeper retention of deuterium with increasing the fluence or the formation of more stable hydrogen–vacancy clusters even without noticeable change in retention depth. In addition, the total amount of deuterium retained in tungsten increased from 7.6×10^{19} D/m^2 at 10^{25} D/m^2 to 4.2×10^{21} D/m^2 at 4×10^{26} D/m^2.

The dependence of blistering and deuterium retention upon the exposure temperature was also examined for tungsten exposed at the fluence of 10^{26} D/m^2. At 315 K, only sparse low-dome blisters with a chord of a few microns or less appeared even the fluence was increased to 10^{27} D/m^2. At around 400 K, the blisters became much denser and the dome of blisters became a little higher. A peculiar change occurred at around 500 K, where both large blisters and small blisters appeared. In high temperature region (higher than 600 K), the blisters became much sparser with the increasing temperature and disappeared at 1000 K. On the other hand, the peak temperature of the deuterium release rate increased simply with the increasing exposure temperature, suggesting that deuterium would diffuse to a deeper depth at higher exposure temperature. However, the sample exposed at around 500 K retained the maximum amount of deuterium among the samples exposed to the same fluence.

4. Conclusions

Two kinds of high-dome blisters appeared at tungsten exposed to the fluence greater than 10^{26} D/m^2 at about 500 K. For most large blisters, there were voids/holes along the grain boundary beneath but no hollow lid formed. On the contrary, there were cavities inside small blisters with the maximum ratio of height against diameter of about 0.7. These high-dome blisters are considered to be generated by the formation of hydrogen–vacancy clusters. In addition, there was significant dependence of blistering and deuterium retention upon fluence and exposure temperature.

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