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Microstructure dependence of deuterium retention and blistering in the near-surface region of tungsten exposed to high flux deuterium plasmas of 38 eV at 315 K

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Abstract
Four kinds of tungsten samples were exposed to 76 eV D_2^+ ions (38 eV D^{-1}) at a flux of 10^{22} D $m^{-2}s^{-1}$. For plasma exposure at 315 K, blistering occurred more significantly on the unrecrystallized and single crystal W samples than the partially and fully recrystallized W samples. The un-recrystallized W sample showed the largest retention ratio at the same fluence. For samples exposed to higher fluences (up to 10^{27} D m^{-2}), the phenomena of bursting release of deuterium and blister bursting were clearly observed. Preliminary positron annihilation measurements indicated that the vacancy concentration in the near-surface region of tungsten increased after the deuterium plasma exposure. The results of electron back-scattering diffraction showed a strong dependence of blistering upon grain orientation, suggesting the possibility of alleviating blistering by selective texturing of tungsten for future fusion reactors.

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1. Introduction

Tungsten has been selected as the plasma facing material at the divertor baffles and dome in the ITER design [1], and all plasma facing components may be made of high-Z materials in future fusion reactors so as to avoid the high tritium inventory of carbon. However, blistering occurs for high-Z materials even if the ion energy is too low to create displacement damage [2–5], with the potential for instability and even disruption of the plasma due to high-Z impurity release into the core plasma and sudden gas recycling. Actually, oscillations of heat load, recycling flux and molybdenum impurity have been observed in the TRIAM-1M, where molybdenum was used at divertor plates and limiters, and this kind of oscillation would lead to termination of the discharges [6].

In this study, blistering and deuterium retention were investigated by exposing various kinds of tungsten samples to high fluences of a high-flux and low-energy deuterium plasma, and the dependence of deuterium retention and blistering upon the microstructures of tungsten was studied with a variety of techniques, such as scanning electron microscopy (SEM), thermal desorption spectroscopy (TDS), positron annihilation (PA) and electron back-scattering diffraction (EBSD).

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2. Experimental

Four kinds of tungsten samples were used, i.e., the un-recrystallized (U), the partially recrystallized (P), the fully recrystallized (F) and the single crystal (111) (S). The S sample (size: $\Phi 9 \times 2$ mm) has a purity of 99.95 wt% and principal impurities are Mo and Fe with 100 and 40 weight ppm, respectively. The sample was annealed in vacuum at 1300 K for 30 min for reducing the density of dislocations that were produced during the sample preparation such as polishing. The others (U, P and F) with sizes of $10 \times 10 \times 2$ mm have a purity of 99.99 wt% with principal impurities (in weight ppm) of Mo and Fe around 10, C and O less than 30. After being cut and double-sided polished, the surface layers of the U sample were removed *in situ* by argon-sputtering prior to deuterium plasma exposure, while P and F samples were partially and fully recrystallized at 1473 and 2073 K, respectively. Each sample was cleaned in an acetone ultrasonic bath prior to placing into the plasma exposure chamber.

The linear plasma generator used in this study is capable of delivering plasma beams comparable to the edge plasma at ITER divertor. The ion species can be controlled by adjusting the operational parameters of the plasma generator and the main impurity in the plasma is oxygen with a concentration less than 1 ppm. The incident flux and energy were fixed at $10^{22}\,\mathrm{D\,m^{-2}\,s^{-1}}$ and $38\,\mathrm{eV\,D^{-1}}$, respectively. The ion energy was determined from the bias voltage of -80 V and the plasma potential of -4 V measured by a Langmuir probe, taking into account the predominant ion species of D_2^+.

Blister formation at the surface of tungsten after the plasma exposure was observed by a SEM at a tilt angle of 45°. TDS at a ramp rate of $0.5\,\mathrm{K\,s^{-1}}$ was used to examine the behaviour of deuterium release from the tungsten samples, and a standard deuterium leak with an accuracy higher than 90% was employed to calibrate the quadrupole mass spectrometer prior to each TDS analysis. In addition, for PA spectroscopy, positrons generated by a ^{22}Na source were thermalized in a moderator, extracted and accelerated to an energy of 0.2–30 keV before being finally delivered to the sample. The defects in the sample were detected by measuring Doppler spectra of the annihilation γ-rays with a high resolution Ge detector and quantified by the so-called *S* parameter, which is defined as the normalized peak area of the Doppler spectrum. Furthermore, the dependence of blistering behaviour on the orientation of grains was examined by EBSD using the electron beam of a field emission SEM.

3. Results and discussion

3.1. Microstructure dependence of blistering and the nucleation of blisters

Both P (the partially recrystallized) and F (the fully recrystallized) samples were exposed in a fluence range of 10^{25} to $10^{27}\,\mathrm{D\,m^{-2}}$, while U (the un-recrystallized) and S (the single crystal (111)) samples were only exposed to 10^{25} and $10^{26}\,\mathrm{D\,m^{-2}}$, respectively. Some SEM images of the four kinds of tungsten samples after the plasma exposure at 315 K are shown in figure 1(a)–(d), respectively. For the U sample (a), blisters with diameters of 1–5 μm appeared after exposure at $10^{25}\,\mathrm{D\,m^{-2}}$. For both P and F samples, however, only a few visible blisters could be observed up to the fluence of $10^{26}\,\mathrm{D\,m^{-2}}$. After the exposure with the fluence of $10^{27}\,\mathrm{D\,m^{-2}}$, blisters with diameters smaller than 1 and 5 μm appeared on the P sample (b) and the F sample (c), respectively. In addition, bursting tails (traces of blister bursting) were also observed on the samples after high fluence

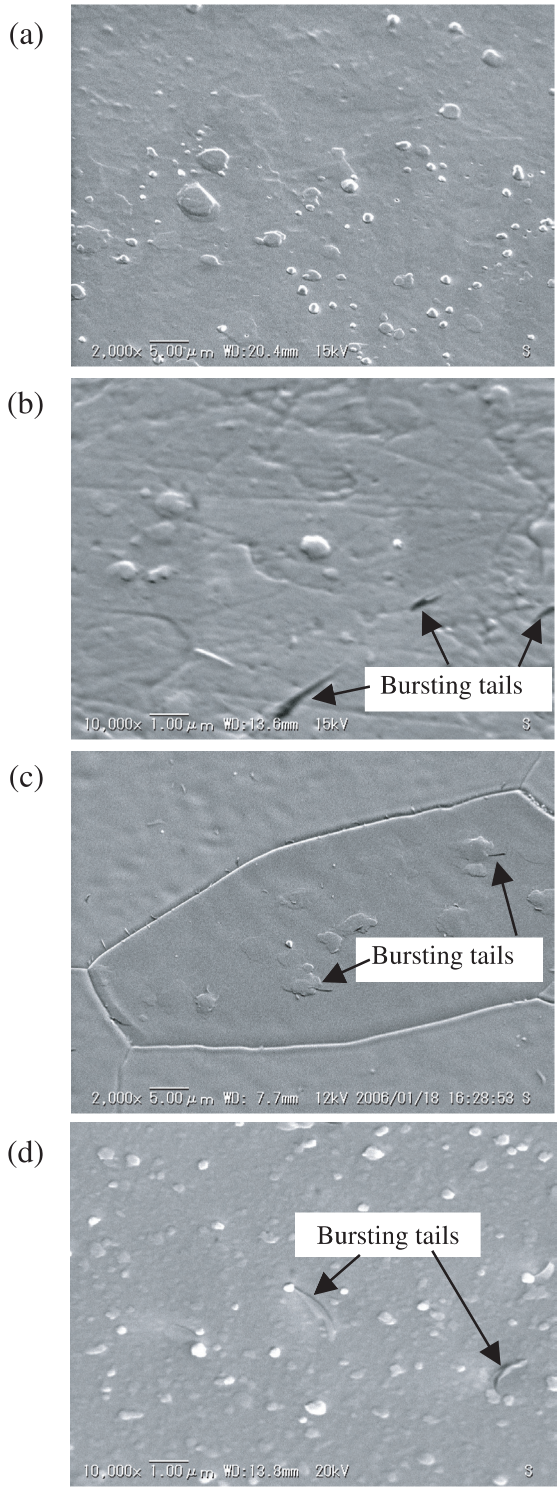

Figure 1. Some SEM images of the four kinds of tungsten after the plasma exposure at 315 K (45° tilt). (a) U sample exposed to $10^{25}\,\mathrm{D\,m^{-2}}$; (b) P sample exposed to $10^{27}\,\mathrm{D\,m^{-2}}$; (c) F sample exposed to $10^{27}\,\mathrm{D\,m^{-2}}$; and (d) S sample exposed to $10^{26}\,\mathrm{D\,m^{-2}}$.

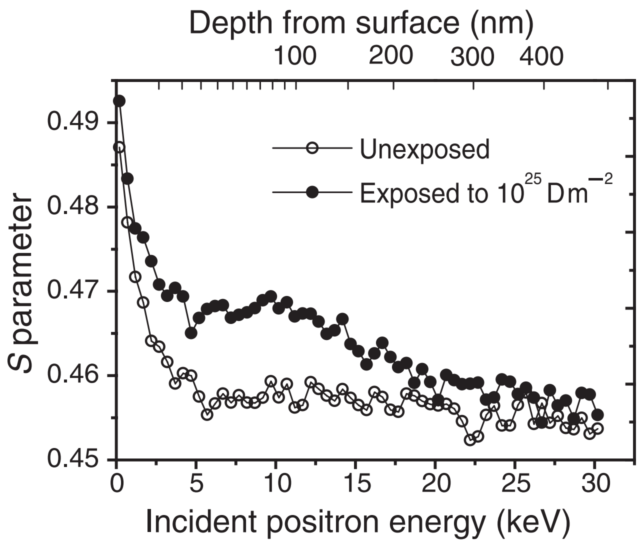

Figure 2. PA results of the P sample exposed to 10^{25} D m^{-2} at 315 K in comparison with that prior to plasma exposure.

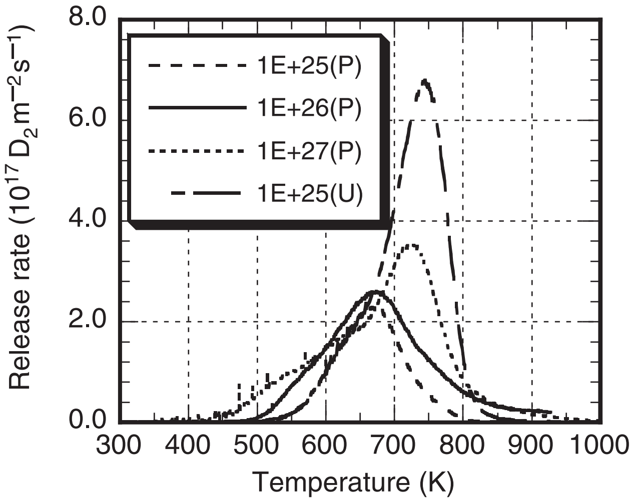

Figure 3. TDS spectra of P samples with varying fluences in comparison with that of the U sample.

Table 1. Retention characteristics of the four kinds of tungsten exposed to varying fluences at 315 K.

Samples	Release peaks (K)	Release shoulders	Retention (D_2 m^{-2})	Retention ratio
1E+25(U)	740	660	1.6×10^{20}	3.2×10^{-5}
1E+25(P)	660		5.8×10^{19}	1.2×10^{-5}
1E+26(P)	660	850	9.4×10^{19}	1.9×10^{-6}
1E+27(P)	740	540, 660, 850	1.3×10^{20}	2.6×10^{-7}
1E+25(F)	640	540, 780	4.8×10^{19}	9.6×10^{-6}
1E+26(F)	640	540, 780, 850	6.4×10^{19}	1.3×10^{-6}
5E+26(F)	640, 780	850	2.6×10^{20}	1.0×10^{-6}
1E+27(F)	640, 780	850	3.3×10^{20}	6.6×10^{-7}
1E+26(S)	540, 780	640, 850	8.7×10^{19}	1.7×10^{-6}

exposure. Furthermore, small blisters with diameters less than 1 μm and blister bursting were observed even for the S sample (d).

It is clear that the blistering behaviour is definitely influenced by the microstructures. For plasma exposure at 315 K, blistering occurred more significantly on U and S samples than P and F samples, indicating that recrystallization can alleviate blistering. PA measurements confirmed that the densities of defects in the near-surface regions of both P and F samples prior to plasma exposure were much smaller than those of both U and S samples.

However, it is still an open question how blisters nucleate during plasma exposure with energies well below the displacement threshold. Poon *et al* [3] argued that the recoil implantation of oxygen and carbon impurities arriving at the surface created vacancies in tungsten exposed to a 500 eV D^+ plasma. Alimov *et al* [4] observed that during 200 eV D^+ irradiation the deuterium concentration in the implantation zone greatly exceeds the solubility limit, and assumed that the matrix lattice is stressed and plastic deformation occurs resulting in formation of voids and vacancy clusters so as to alleviate these tensions. On the other hand, Shu *et al* [5] considered that Fukai's theory of formation of superabundant vacancies (vacancy–hydrogen clusters) [7], where the formation energy of a vacancy is decreased by cluster formation and the configurational entropy of the system at high hydrogen concentrations, might be valid in the present case of tungsten exposed to a high-flux and low-energy deuterium plasma. This consideration was thus examined preliminarily by PA measurements. As shown in figure 2, the S parameter of the P sample exposed to 10^{25} D m^{-2} at 315 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. Further detailed examinations are required.

3.2. Microstructure dependence of deuterium retention

Figure 3 shows TDS spectra of P samples with varying fluences in comparison with that of a U sample. As the fluence was increased, the amount of deuterium retained in P samples increased, and the peak temperature shifted from 660 K for the samples exposed to 10^{25} D m^{-2} to 740 K for the sample exposed to 10^{27} D m^{-2}. In addition, the amount of deuterium retained in the U sample exposed to 10^{25} D m^{-2} was even greater than that of the P sample exposed to 10^{27} D m^{-2}, indicating a strong dependence of deuterium retention on microstructures of tungsten. Furthermore, the phenomenon of bursts in the release rate of deuterium was observed for the sample exposed to the highest fluence. It is clear that these sudden releases were accompanied with bursting of blisters. The phenomena of blister bursting and bursting release of deuterium agree well with the relaxation oscillations of recycling flux and molybdenum impurity observed in the plasma of TRIAM-1M [6].

Table 1 lists the retention characteristics of the four kinds of tungsten samples exposed to varying fluences at 315 K. Release peaks and shoulders changed from 540 to 850 K, and U and S samples as well as P and F samples with the highest fluence showed higher release peaks. In addition, the U sample showed the largest retention ratio at the same fluence. Furthermore, as fluence increased, the retention increased whereas the retention ratio decreased for both P and F samples.

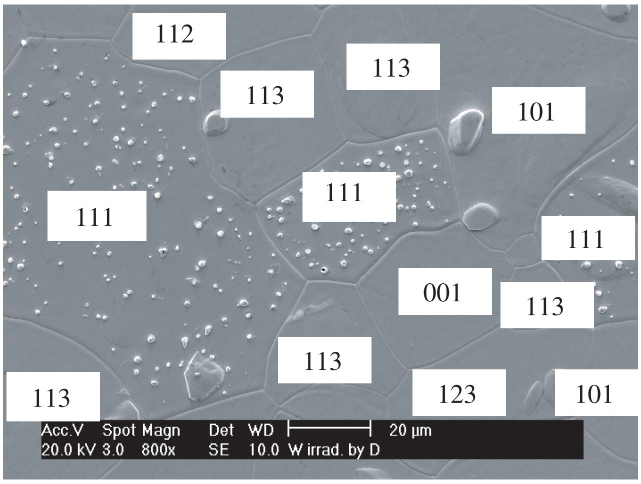

Figure 4. Dependence of blistering upon the grain orientation for an F sample exposed to 10^{26} D m^{-2} at 520 K.

3.3. Orientation dependence of blistering and ideas for alleviating blistering

The blistering dependence upon the grain orientation is shown in figure 4 for an F sample exposed to 10^{26} D m^{-2} at 520 K, at which deuterium retention was highest [8]. In addition, blisters did not appear uniformly over the whole surface. There exist some grains where no visible blisters appeared. The results of EBSD showed that small blisters only appeared on grains with nearly (111) surface orientation, suggesting that it is possible to control or alleviate both blistering and retention by arranging suitable orientations (by selective texturing) of tungsten for future fusion reactors.

Another idea for alleviating blister is to maintain tungsten at an elevated temperature (above 900 K) where the trapping sites are no longer able to hold hydrogen isotopes [8]. However, maintaining tungsten at high temperature may bring about the problem of helium blistering [9]. Thus, it is necessary to investigate the blistering behaviour of tungsten exposed to a He–H mixed plasma.

4. Conclusions

1. Both blistering and deuterium retention were strongly influenced by the microstructures of tungsten. Blistering occurred more significantly on the un-recrystallized and the single crystal W samples than the partially and the fully recrystallized W samples, with the plasma exposure at 315 K. The un-recrystallized W sample showed the largest retention ratio at the same fluence. Preliminary PA measurements suggested vacancy generation due to the exposure.
2. Bursting release of deuterium and blister bursting occurred for samples exposed to the higher fluences (up to 10^{27} D m^{-2}), but these phenomena could be alleviated by selective texturing of tungsten due to the strong dependence of blistering upon the grain orientation.

Acknowledgments

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