P71

Surface morphology and deuterium retention in tungsten exposed to low-energy, high flux pure and helium-seeded deuterium plasmas

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Abstract

Blistering and deuterium retention in re-crystallized tungsten exposed to low-energy, high flux pure and helium-seeded D plasmas to a fluence of 10^{27} D/m^2 have been examined with scanning electron microscopy, thermal desorption spectroscopy, and the D(^{3}He,p)^{4}He nuclear reaction at a ^{3}He energy varied from 0.69 to 4.0 MeV. In the case of exposure to pure D plasma (38 eV/D), blisters with various shapes and sizes depending on the exposure temperature are found on the W surface. No blisters appear at temperatures above 700 K. The deuterium retention increases with the exposure temperature, reaching a maximum value of about 10^{22} D/m^2 at 480 K, and then decreases as the temperature rises further. Seeding of 76 eV He ions into the D plasma significantly reduces the D retention at elevated temperatures and prevents formation of the blisters.

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1. Introduction

As plasma-facing material in the tokamak divertor area, tungsten (W) will be subjected to bombardment with low-energy, high flux deuterium (D) and tritium particles including helium (He) ash. It is known that He irradiation leads to such structure modification as bubble formation, vacancy swelling, blistering, flaking, porous surface structure, depending on the irradiation conditions [1-7]. Retention of hydrogen isotopes in W is enhanced by pre-irradiation with He ions at energies of several keV [8-12], whereas sequential 500 eV He^+-D^+ irradiation at 300 K results in a ~70% reduction in deuterium retention compared to D^+-only irradiation [13]. For simultaneous 500 eV He^+-D^+ irradiation at 300 K, deuterium retention is similar to D^+-only levels. However, the presence of He enhances deuterium trapping in the near surface, while limiting D diffusion into the bulk [14]. Simultaneous irradiation with 1 keV H_3^+ and He^+ ions at 473K significantly reduces the number of blisters [15].

In this study, surface morphology and D retention in tungsten exposed to low-energy, high flux pure and helium-seeded D plasmas at various temperatures were investigated.

2. Experimental

Polycrystalline tungsten (A.L.M.T. Corp., Japan) re-crystallized fully at 2070 K after cutting and polishing, with a purity of 99.99 wt%, was used. The linear plasma generator used for delivering plasma beam comparable to the edge plasma at ITER divertor is described elsewhere [16]. To generate pure D plasma, D_2 gas was filled in the plasma generation section to a pressure of 1 Pa. In doing so, plasma beam highly enriched with species of D_2^+ to over 80% of ions was obtained [16]. The bias voltage of -80 V was applied to the W sample resulting to the incident energy of 38 eV/D, taking into account the plasma potential of about -4 V measured by the Langmuir probe. The incident deuterium ion flux and fluence were fixed at 10^{22} D/m^{2}s and 10^{27} D/m^2, respectively. The exposure temperature was set by the thermal contact between the sample and holder.

To generate helium-seeded D plasmas, ^{4}He was added to D_2. The He partial pressure in the plasma generation section was 4×10^{-3} and 1×10^{-1} Pa, whereas the D_2 partial pressure was fixed at

1 Pa. Percentage of He in the He+D_2 gas mixture was determined by means of a high resolution quadrupole mass-spectrometer (HRQMS) QMA 410 (Balzers Instruments). The energies of D and He ions were fixed at 38 eV/D and 76 eV/He, respectively.

Concentration of He ions in the helium-seeded D plasmas was determined by measuring sputtering yields of W target exposed to the helium-seeded D plasmas with various He percentages. For these experiments the bias potential was chosen to be -200 V, therefore the energy of D_2^+ and He^+ ions was 196 eV. The sputtering yield, Y, of W exposed to the pure D and He plasmas was determined to be $Y_{D2} = (1.2\pm0.6)\times10^{-5}$ W/D_2^+ and $Y_{He} = 2.7\pm0.5\times10^{-4}$ W/He^+, respectively. Linear dependence of the sputtering yield on the percentage of He atoms in the He+D_2 gas mixture used for generation of the helium-seeded D plasma allows conclusion that the He ion concentration in the plasmas equals to the He atom concentration in the gas mixtures.

Deuterium and helium retention in the W samples was monitored ex-situ using thermal desorption spectrometry (TDS). An infrared heater was used to heat the samples at a ramp rate of 0.5 K/s and the sample temperature was raised to 1300 K. HD, ^{4}He and D_2 molecules released during TDS run were monitored by the HRQMS. To calculate the relative contribution of the recorded HD and D_2 masses to the total release of deuterium, the partial currents of the HRQMS were normalized as described in Ref. [17]. Standard D_2 and He leaks with an inaccuracy lower than 10% were employed to calibrate the HRQMS after each TDS analysis.

The D concentration was measured by means of the D(^{3}He,α)H reaction, where both, the α particles and protons were analyzed. To determine the D concentration at larger depths, an analyzing beam of ^{3}He ions with energies varied from 0.69 to 4.0 MeV was used, and the proton yields measured at different ^{3}He ion energies allowed to measure the D depth profile at depths of up to 7 μm [18].

The surface morphology of the plasma-exposed W samples was examined by a scanning electron microscope (SEM) (Real Surface View Microscope, KEYENCE VE-9800) at a tilt angle of 45º.

3. Results and discussion

After exposure to the pure D plasma at temperatures, T_{exp}, in the range from 320 to 370 K, low-dome blisters with sizes up to 10 microns are observed (Fig. 1 (a)). At T_{exp} = 460-515 K, two kinds of blisters appear: large blisters with sizes of a few tens of microns and small blisters with chords of less than a few microns. Some of the small blisters have openings in the blister lids (Fig. 1 (c)). No blisters appear at exposure temperatures above 700 K.

Seeding of helium ions into the D plasma to a concentration of 0.2% does not change the surface morphology at exposure temperature of 335 K, while at $T_{\rm exp}$ = 495 K the blisters become much sparser and disappear at $T_{\rm exp} \geq 600$ K. An increase of the He ion concentration up to 5% at $T_{\rm exp}$ = 340-430 K leads to formation of less blisters (Fig. 1 (b)). At exposure temperatures of 460-530 K no blisters appear after exposure to the helium-seeded D plasma (Fig. 1 (d)). However, small blisters 0.2-0.5 μm in size appear at $T_{\rm exp}$ = 630-720 K and disappear at $T_{\rm exp}$ = 810 K.

In W exposed to pure D plasma at $T_{\rm exp}$ = 320 K, the deuterium depth profile is characterized by a sharp near-surface concentration maximum of 2-3 at.%, a concentration of about 0.2 at.% at depths of 1-2 μm, and a decreasing concentration tail into the bulk (Fig. 2 (a)). As the exposure temperature increases up to 460 K, the D concentration in the near-surface slightly decreases, whereas the concentration at depths above 1 μm increases reaching the maximum value of about 1 at.%. This high D concentration could be due to accumulation of D_2 molecules in cavities created during the D plasma exposure [19, 20]. Further increase of the exposure temperature leads to a decrease of the D concentration (Fig. 2 a).

In W exposed to helium-seeded D plasma with 5% of He ions, the D concentration in the near-surface layer decreases from ~4 down to ~0.4 at.% as the exposure temperature increases from 345 to 725 K, while the D concentration at depths from 1 to 7 μm decreases even stronger and, at $T_{\rm exp} \geq 630$ K, it is below the NRA detection limit (10^{-4} at.%) (Fig. 2 (b)).

In W exposed to pure D plasma, the D retention is about 2×10^{21} D/m^2 at $T_{\rm exp}$ = 330 K and, as exposure temperature increases, rises to its maximum of about 10^{22} D/m^2 at $T_{\rm exp}$ = 480 K and then decreases down to about 10^{19} D/m^2 at $T_{\rm exp}$ = 800 K (Fig. 3). One can recognize from the comparison of the TDS and NRA data that less than 50% of retained deuterium is accumulated in the sub-surface layer up to 7 μm.

Seeding of He ions into the plasma at exposure temperature below 350 K does not change strongly the D retention, while at the temperatures above 400 K the D retention decreases significantly compared to that for the pure plasma exposure. At $T_{\rm exp} \geq 600$ K, the D retention is about 10^{19} D/m^2 independent of the He ion concentration (Fig. 3). Good agreement of the TDS and NRA data after exposure to the helium-seeded D plasmas (Fig. 3) shows that deuterium is accumulated only in the near-surface layer.

For exposure to helium-seeded D plasmas, the He retention does not depend practically on the He ion concentration in the D plasma and increase with the exposure temperature from about 2×10^{19} He/m^2 at $T_{\mathrm{exp}} = 340$ K to about 3×10^{20} He/m^2 at $T_{\mathrm{exp}} = 810$ K (Fig. 3).

For tungsten implanted with D ions at energies below the displacement threshold, the mechanism of plastic deformation due to deuterium super-saturation has been considered for formation of trapping sites for deuterium [21, 22]. During D plasma exposure, the D concentration in the implantation zone greatly exceeds the solubility limit and stresses the matrix lattice until plastic deformation occurs to alleviate these tensions. This deformation is assumed to be responsible for the generation of vacancies, vacancy complexes and microscopic cavities at depths of several micrometers and the concurrent accumulation of diffusing deuterium. It may be suggested that the stress-induced plasticity of tungsten appears at high concentrations of soluble hydrogen and increases as the temperature grows. At long-term irradiation the diffusing D atoms recombine on the cavity surfaces, increasing thus the gas pressure inside these cavities. At near-room temperatures, as more deuterium is deposited, cooperative fracture between the cavities suddenly becomes an easy way of relieving their overpressure, thus initiating cracks, allowing internal gas release. At elevated temperatures high deuterium pressure inside the cavities leads to extrusion of metal and formation of blisters [20].

According to Iwakiri et al. [3], in W irradiated with low-energy He ions, He platelets and bubbles are formed in the implantation zone. Based on results obtained for simultaneous irradiation of W with helium and hydrogen ions, Ueda et al. [15] have concluded that hydrogen atoms can be trapped at the periphery of He bubbles reducing thus hydrogen inward flux. He bubbles formed in the near-surface layer under exposure to the He-seeded D plasmas could increase stress field around the high-pressure bubbles generating defects in the crystal lattice [8, 13-15], in doing so the defects could serve as trapping sites for D atoms. On the other hand, a dynamic mechanism of nano-scale helium bubble formation can lead to development of an open porosity in the near-surface layer and can create short-circuit paths to the surface enhancing thus the D re-emission and limiting the D diffusion into the bulk. Additionally, the porous near-surface structure may serve as a damper layer to dissipate the compressive stresses induced by the local deuterium super-saturation. As this takes place, the D trapping sites are not generated in the sub-surface layer.

4. Summary

Significant temperature dependence of blistering and deuterium retention is found for re-crystallized W exposed to low-energy (38 eV/D), high flux (10^{22} D/m^{2}s) D plasma at an ion fluence of 10^{27} D/m^2. At 320 K, only sparse blisters with chords of 0.5-2 μm are formed on the W surface. In this case the D depth profiles are characterized by a near-surface concentration maximum of about 2 at.% and, in the sub-surface layer (at depths from 1 to 7 μm), by a concentration of about 0.2 at.% slowly decreasing into the bulk. As the exposure temperature increases, the blisters become much denser. At 460-515 K, two kinds of blisters appear: large blisters with sizes of 10-30 μm and small cone-shaped blisters with chords of less than a few μm. As this takes place, the D concentration in the sub-surface layer reaches 1 at.%. No blisters appear at temperatures above 700 K, and the D concentration at depths of several micrometers is about 10^{-3} at.%. The D retention is about 2×10^{21} D/m^2 at 320 K and, as exposure temperature increases, rises to its maximum of 10^{22} D/m^2 at 480 K and then decreases to 10^{19} D/m^2 at 800 K.

Seeding of helium ions (76 eV/He) into the D plasma significantly reduces the D retention at elevated temperatures (400-700 K) and prevents the blister formation. In doing so, deuterium is accumulated only in the near-surface layer.

D plasma exposure with ion energies well below the displacement threshold modifies the W structure to depths of several micrometers. Stress-induced plastic deformation caused by deuterium super-saturation within the near-surface layer is suggested as a mechanism for D_2-filled cavity formation. Seeding of He ions into the D plasma leads to formation of the He porous structure in the near-surface layer which serves as a damper layer to dissipate the compressive stresses induced by the local deuterium super-saturation. In doing so, the D trapping sites in the sub-surface layer are not generated.

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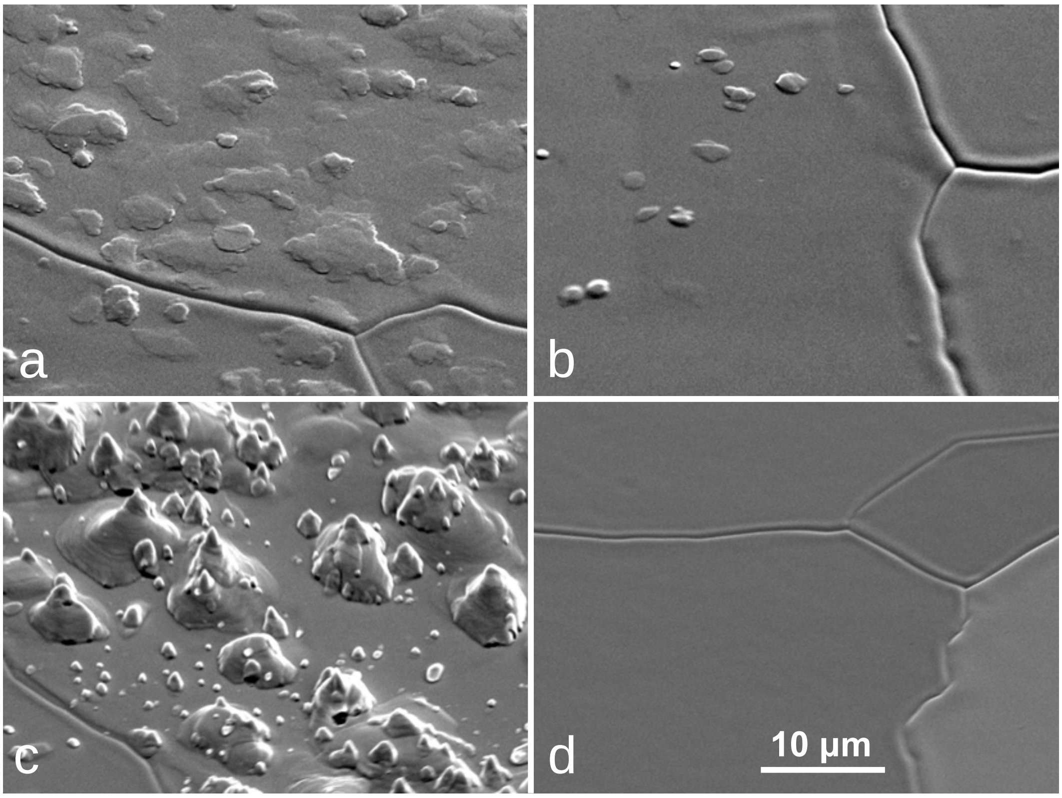
Figure captures

Figure 1. SEM images of re-crystallized W exposed to pure D plasma (38 eV/D) (a, c) and helium-seeded D plasma (38eV/D + 76 eV/He, 5% of He ions) (b, d) at temperatures of 360 K (a), 345 K (b), 460 K (c), and 470 K (d) to D ion fluence of 10^{27} D/m^2. The scale bar (d) is the same for all images.

Figure 2. Depth profiles of deuterium retained in re-crystallized W exposed to pure D plasma (38 eV/D) (a) and helium-seeded D plasma (38eV/D + 76 eV/He, 5% of He ions) (b) to D ion fluence of 10^{27} D/m^2 at various temperatures.

Figure 3. Deuterium and helium retention in re-crystallized W exposed to pure D plasma (38 eV/D) and helium-seeded D plasmas (38eV/D + 76 eV/He, 0.2 and 5% of He ions) to D ion fluence of 10^{27} D/m^2, as a function of the exposure temperature. The deuterium and helium retention was determined by thermal desorption spectrometry (TDS), whereas the D retention up to a depth of 7 μm was calculated from D depth profiles measured by the D(^{3}He,α)H nuclear reaction analysis (NRA).



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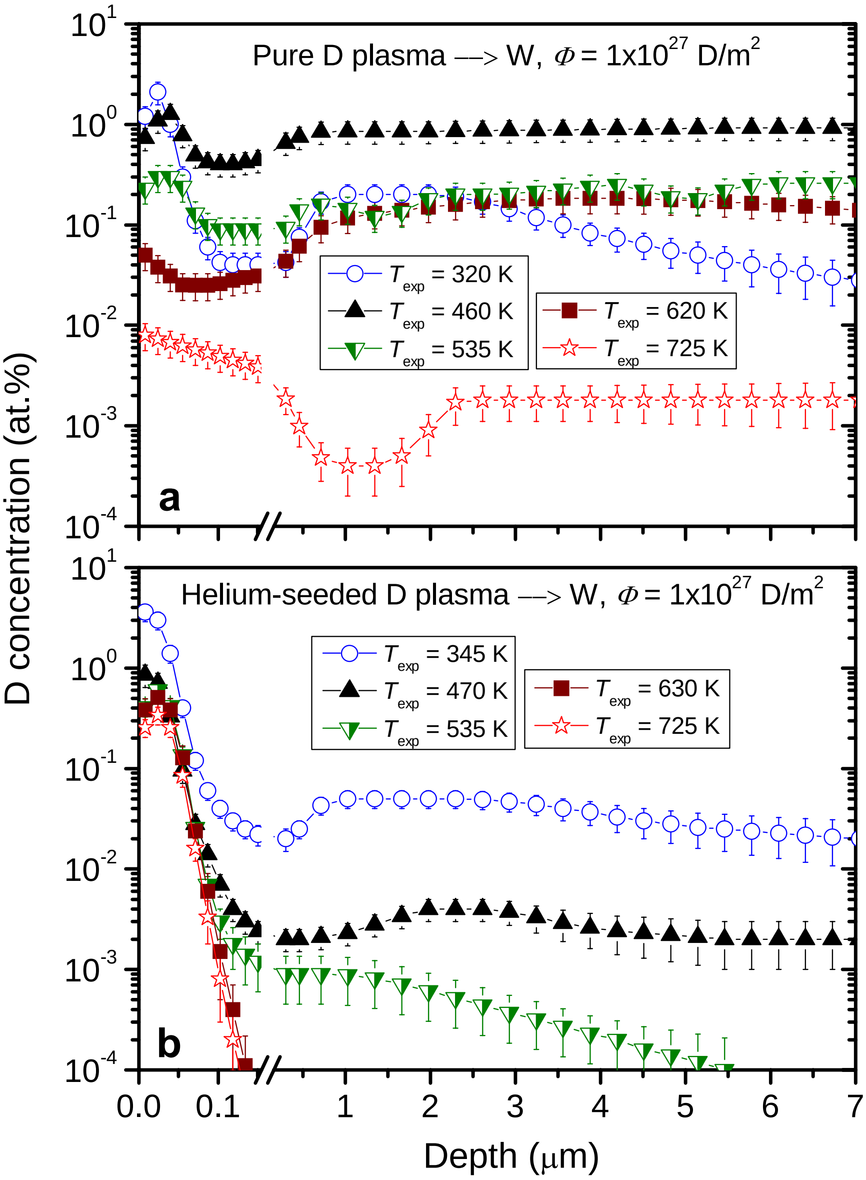

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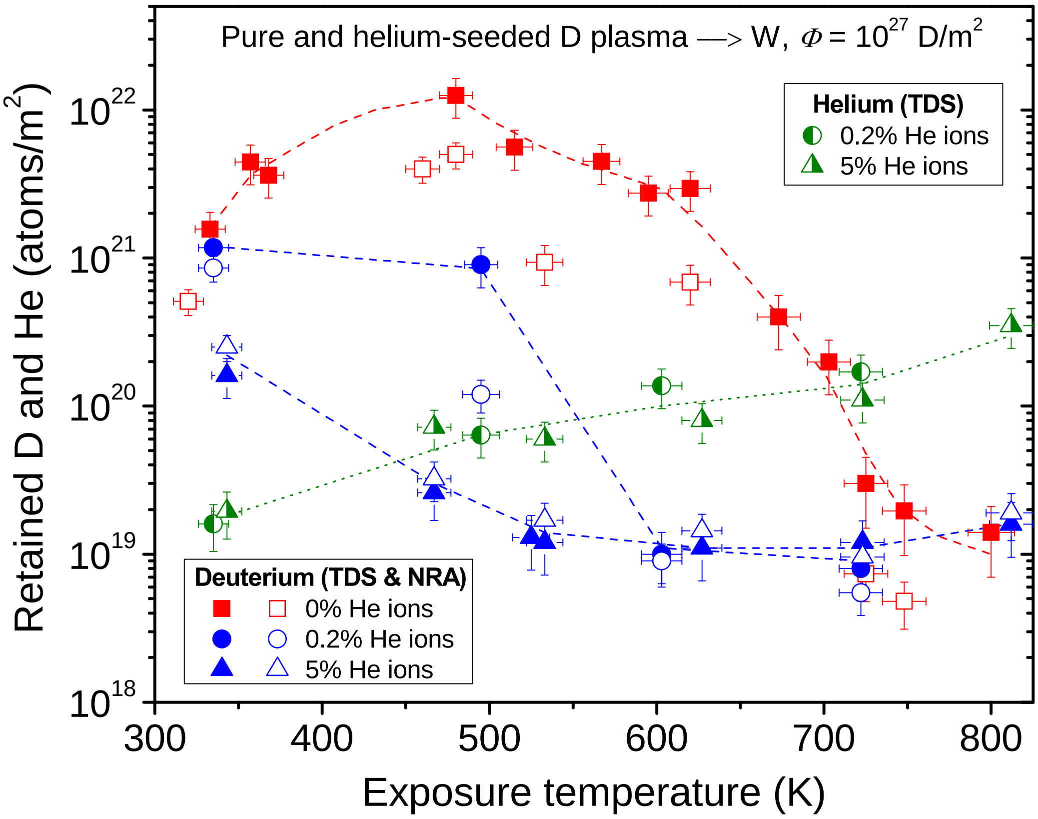

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