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Simultaneous irradiation effects of hydrogen and helium ions on tungsten

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ABSTRACT

Simultaneous irradiation effects of He on tungsten blistering with hydrogen and carbon mixed ion irradiation were investigated. It was found that only 0.1% addition of He ions to 1 keV H and C mixed ion beam (C: 0.8–1.0%) reduced (473 K) or completely suppressed (653 K and 723 K) blister formation. According to TEM observation, He bubbles with the size of 2 nm or less were formed near the surface, which could be a diffusion barrier of hydrogen into the bulk due to the reduction of diffusion channel or excitation of stress field, leading to the reduction of diffusivity of hydrogen. The reduction rate of hydrogen inward flux by simultaneous He irradiation in our experimental conditions would be more than the factor of three.

1. Introduction

The effects of He ion irradiation on wall materials are important research subject especially for metallic materials. It is known that He ions impinging on the high Z wall materials tend to be trapped at vacancies, grain boundaries, dislocations, and so on [1,2]. As He irradiation fluence increases, He atoms are trapped at He-vacancies complex to form He bubbles. The effects of these bubbles on hydrogen isotope retention in W were studied for He pre-irradiated samples [3,4] and He and D simultaneously irradiated samples [5]. In these studies, it was clearly shown that He irradiation greatly affects hydrogen isotope trapping in W either by bubble formation or by excitation of stress field near the bubbles.

The abovementioned studies provided very important information on hydrogen isotope retention in W by He ion irradiation. Under fusion reactor environments, however, hydrogen isotopes diffuse relatively fast at elevated temperatures in W and would be trapped at traps in the bulk (intrinsic or neutron induced). Therefore, we also need to know the effects of He bubble structure near the surface on hydrogen isotope diffusion into the bulk to correctly evaluate hydrogen isotope behavior in wall materials. However, there have been no studies on this issue.

We have studied hydrogen blistering on W by hydrogen and carbon mixed ion beam irradiation [6,7]. Hydrogen blistering takes place through implantation of H near the surface, diffusion into the bulk, and trapping at grain boundaries which leads to crack formation. The reason for the use of the mixed ion beam in our studies is to deliberately form W and C mixed layer on the tungsten surface [8], which acts as a desorption barrier of hydrogen (low surface recombination rate of WC) and enhances hydrogen inward flux into the bulk. This enhancement of hydrogen inward flux made it possible to cause blistering. By the detailed study on the formation conditions of this hydrogen blistering on W by using this technique, it is possible to study hydrogen diffusion behavior qualitatively.

In this paper, we first present how blistering on hot-rolled tungsten changes with the addition of He ions to H and C mixed ion beam. Then, microstructures of tungsten surface irradiated by mixed ion beam are shown. Finally, He effects on tritium diffusion in W are discussed.

2. Experimental

Ion irradiation was done by an ion beam device HiFIT [9], which has a capability to control small amount of impurities such as He and C with the resolution of about 0.1%. Ion species ratio in the mixed ion beam was measured by a magnetic deflection mass analyzer. Part of the beam ions are subject to neutralization and dissociation in the beam transport region before they entered the analyzer. To correctly evaluate ion species ratio from the mass analyzer signal, we take collision process of beam ions and residual gas molecules into consideration. Details of ion species measurements were described elsewhere [9].

In irradiation experiments, an ion energy was 1 keV with the dominant hydrogen molecular ion component of H_3^+ (50–60%). Hydrogen atom flux was $\sim 2 \times 10^{20}$ H/m^{2}s. In most of the experiments, C concentration was fixed at 0.8–1.0%. Under this condition, C ion irradiation resulted in W and C mixed layer, which made surface conditions for all samples similar [8] (C concentration of 50–60% at the surface). Without C addition to the ion beam, blisters did not form under our ion beam condition [6].

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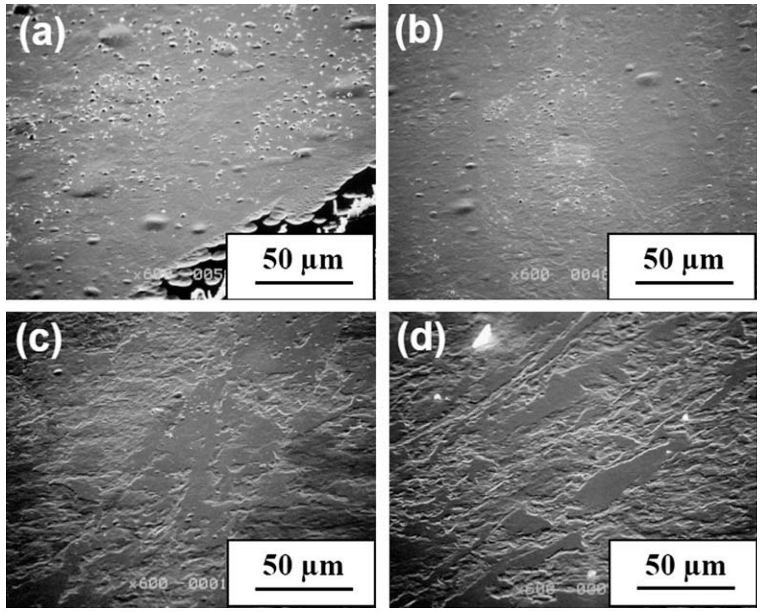

Fig. 1. Surface morphology of ion irradiated W by 1 keV H, C, and He mixed ion beam. Carbon concentration was about 1.0%. Helium concentrations in the beam were 0% (a), 0.1% (b), 1.0% (c), 4.3% (d).

Samples used for this study is sintered and hot-rolled tungsten plates (purity of 99.99%, A.L.M.T. Co.) with the thickness of 1 mm. Heat treatment at 900 °C for half an hour was done for stress relief. Sample surface was mirror polished to the roughness of ~0.01 μm by diamond powder prior to ion irradiation.

3. Effects of He bubbles on blistering

It was found that small amount of He in the ion beam affected blister formation on W. The typical result is shown in Fig. 1. Here, H and C mixed ion beam was irradiated to the fluence of $7.5 \times 10^{24}\ m^{-2}$ at 453 K. Carbon concentration in the ion beam was about 1.0%. For no He case (Fig. 1(a)), a lot of small blisters with diameters less than 10 μm and a few large blisters (>10 μm) appeared on the surface (number density of blisters was ~8500/mm^2). Only 0.1% addition of He to the mixed ion beam greatly reduced the number of blisters, see Fig. 1(b). Both small and large blisters were reduced in number density (~4000/mm^2). For 1.0% (Fig. 1(c)) and 4.3% (Fig. 1(d)) He cases, almost no blisters appeared

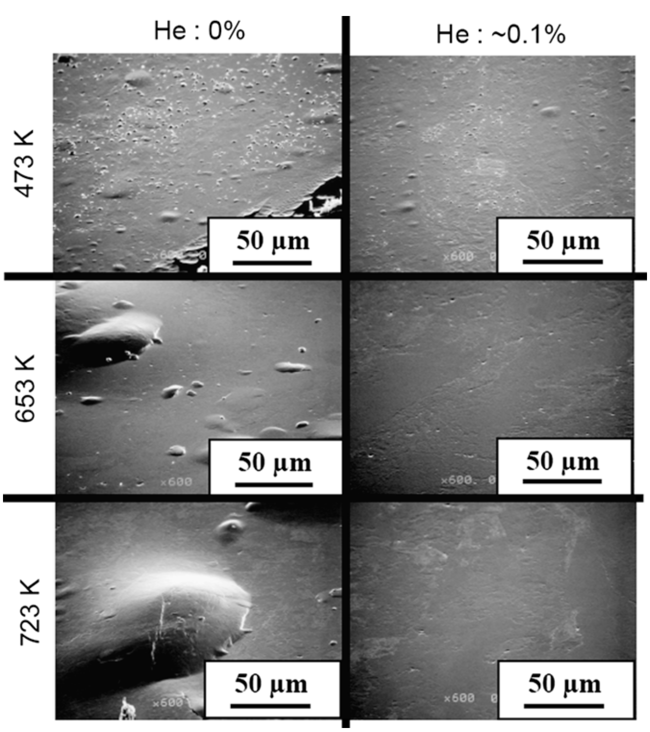

Fig. 2. Temperature dependence of surface morphology of ion irradiated W by 1 keV H, C, and He mixed ion beam. Carbon concentration was about 1.0%.

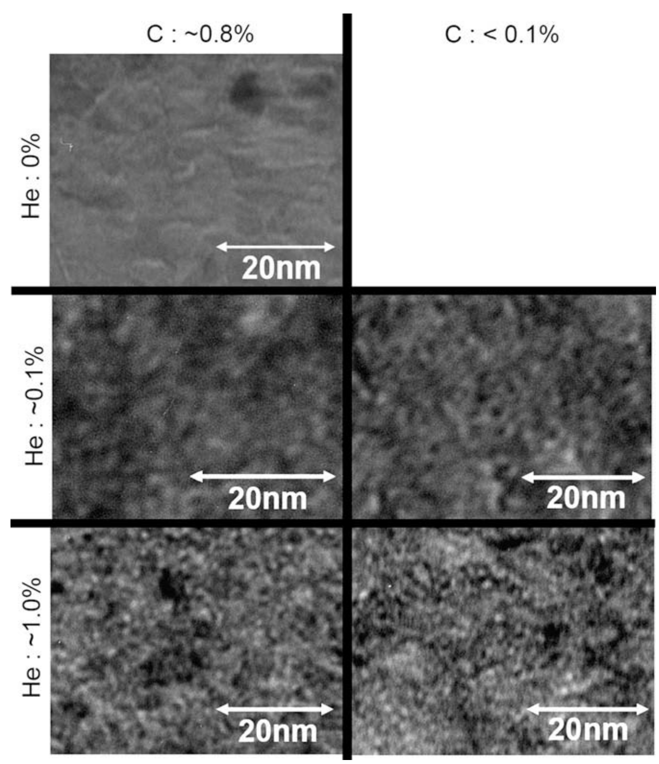

Fig. 3. Microstructure of ion irradiated W by H and C mixed ion beam as a function of He concentration in the beam.

on the surface and surface roughness was increased due to sputtering erosion by He ions.

Temperature dependence of He effects from 473 K to 723 K was shown in Fig. 2. At elevated temperatures ($\geqslant$653 K), small blisters with diameters less than 10 μm disappeared and mostly large blisters ($\geqslant$10 μm) were formed for no He case. Addition of 0.1% He ions completely suppressed their formation. At 473 K, however, this complete suppression of blisters by 0.1% He addition did not occur, though significant reduction of blister numbers was observed. The blister characteristics could be different for those at elevated temperatures ($\geqslant$653 K) and at low-temperatures (473 K). The high-temperature blisters have dome-like shapes with the size of more than 10 μm (up to an order of mm), while the low-temperature blisters have plateau-like irregular shapes with the size of an order of μm. Therefore, it is suggested that formation mechanism of these blisters are different and this difference could relate to sensitivity to He effects.

4. Microstructure of He bubbles formed by mixed ion irradiation

Microstructure near the top surfaces of ion irradiated W samples was observed by a 200 keV transmission electron microscope [1]. Pre-thinned samples were used for the final preparation by twin-jet electro-polishing. The thickness of observation region was roughly 20 nm. Results are shown in Fig. 3. He bubbles with the sizes of 2 nm or less were observed for He: 1.0% cases. For the case of He: 0.1%, He bubbles were also observed. The density of He bubbles was higher for He: 1.0% than that for He: 0.1%.

To compare the cases of high C concentration (C: 0.8%) and low C concentration (C: <0.1%) in the ion beam, He bubble sizes and densities are almost similar, see Fig. 3. For the high C case (C: 0.8%), the top surface was covered by W and C mixed layer (50–60% C) with the thickness of 10–15 nm [8]. This mixed layer consisted of mainly WC confirmed by XRD [8]. The TEM diffraction pattern also indicated the presence of WC. On the other hand, for low C case (C: <0.1%) the TEM diffraction pattern showed mostly pure W in the surface layer.

In Fig. 4, ion range distribution of H and He in W (a) and in WC (b) is shown. The density of WC is assumed to be 15.8 g/cm^3. Our ion beam contained several hydrogen molecular ions such as H_3^+, H_2^+, and H^+ with atomic species ratio of ∼70% (0.33 keV), ∼20% (0.5 keV), and ∼10% (1 keV), respectively. The ion range of the dominant species, H_3^+ (three 0.33 keV H atoms), in W was shorter than the ion range of 1 keV He, see Fig. 4(a). The ion ranges in WC is similar to those in W for H and He. Therefore, most of hydrogen species impinged in the shallower region than He, no matter how much carbon ions were included in the beam.

According to Iwakiri et al. [1], He bubbles are formed near the implantation zone. They showed that dense He bubbles with the size of 2 nm or more were formed when the fluence reached in the middle of 10^{20} He/m^2 at the temperature of 873 K or less at the energy of 0.25 keV. In our experiments, total helium ion fluence to the samples are 7.5×10^{22} He/m^2 for the case of He: 1.0% and 7.5×10^{21} He/m^2 for the case of He: 0.1%. Even if He atoms was not desorbed from the sample, He atoms remaining in the samples were much less than these values due to the sputtering erosion. For high C concentration case (C: 0.8%) erosion depth was about 300 nm, while it was about 100 nm for low C case (C: <0.1%). Since width of the implantation zone of He was about 10 nm (see Fig. 4), actual fluence to the samples were lower than that of injected He ions by an order of magnitude. However, even for the low He case (He: 0.1%) its actual fluence was still in the order of 10^{20} H/m^2, enough to form He bubbles according to Iwakiri et al. [1]. It is noted that these He bubbles formed not only in the W layer but also the W and C mixed layer.

From the abovementioned results, it is concluded that He bubbles formed near the surface significantly reduced hydrogen inward flux, which is schematically shown in Fig. 5. Since hydrogen atoms can be trapped at the periphery of He bubble, the bubbles become diffusion barrier for hydrogen atoms. He bubbles could also increase stress field around the bubbles, leading to the reduction of a diffusion of hydrogen atoms through this region. We made

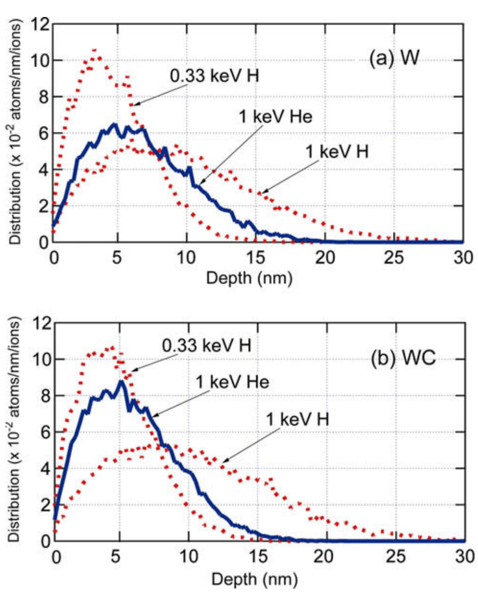

Fig. 4. Ion range distribution of 0.33 keV H, 1 keV H, and 1 keV He in pure W (a) and WC (b).

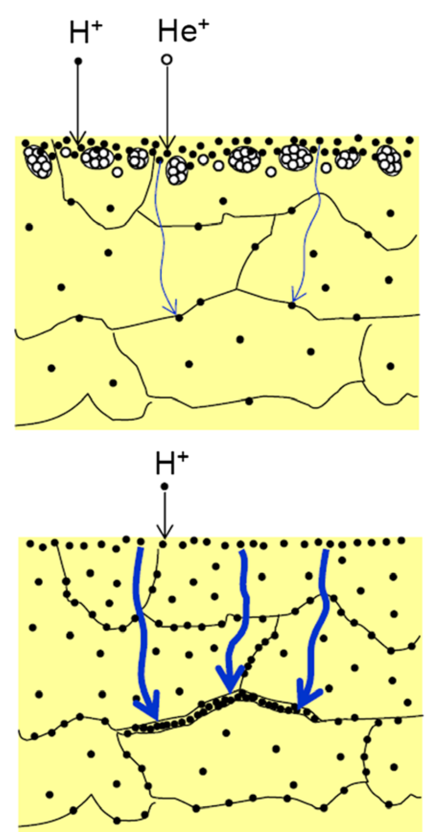

Fig. 5. Schematics of He bubble effects on hydrogen blistering of W. Top figure: with He bubbles, bottom figure: without He.

the reduced flux experiments without He in the flux of $0.7 \times 10^{20}\,\mathrm{m^{-2}s^{-1}}$ (about 1/3 of the irradiation flux of the above-mentioned experiments). Under this condition, blistering also took place. This suggests that the addition of 0.1% He (no blistering, see Fig. 2) reduced hydrogen inward flux into the bulk by more than a factor of three.

5. Conclusion

The effects of He on tungsten blistering were studied by 1 keV H and C mixed ion beam irradiation to W. It was found that only 0.1% addition of He to the mixed ion beam significantly reduced the number of blisters. Especially at elevated temperatures (>653 K), this completely suppressed blister formation. By TEM observation, He bubbles were observed for He: 0.1% case in both W and WC layers. Since these He bubbles were formed a bit deeper than the ion range of dominant hydrogen ion species (H_3^+), it is concluded that He bubble structure significantly reduced H diffusion into the bulk. These results suggest that the He effects on hydrogen isotope diffusion need to be studied to correctly evaluate hydrogen isotope behavior in W under burning plasma conditions.

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