Blister/Hole Formation on Tungsten Surface due to Low-Energy and High-Flux Deuterium/Helium Plasma Exposures

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Abstract. Deuterium/helium plasma exposures on tungsten surface bring serious damages such as blister and hole. Blistering occurs by cleaving along layered structure intrinsic to the press-roll manufacturing process. Mechanical polishing and helium pre-exposure on mirror-finished powder metallurgy tungsten drastically suppress blister formation. Small cracks made by a polishing would become paths to the surface for diffusing deuterium atoms in the substrate, resulting in no gas accumulation and no blister formation on the surface. Helium pre-exposure would make a helium-enriched layer near the surface, which becomes a kind of diffusion barrier for incident deuterium atoms. Blister formation and deuterium retention are suppressed on the surface with helium-enriched layer.

1. Introduction

Plasma-facing materials in a magnetically confined fusion experimental reactor, like ITER, will be exposed to plasmas with high-flux hydrogen isotope and helium (He) ions. It is predicted that the divertor plate will be exposed to low-energy and high-flux plasma with an incident ion energy of below 100 eV and at a flux of ~ $10^{23\text{-}24}$ $m^{-2}s^{-1}$ unlike other first wall materials. Tungsten (W) has been considered to be one of the most important candidates as a divertor material for ITER because of its good material properties like high threshold energy of sputtering, high melting point and low tritium inventory. Impurity release and sudden gas desorption from wall materials due to transient heat load on the plate could lead the core plasma to disrupt. Increasing tritium inventory in the wall is also a significant problem in terms of the safety issue of radioactivity. Although the incident energy of ions to the diverter plate is expected to be below 100 eV, which is lower than the threshold energy of displacement and physical sputtering, recent experiments have shown that low-energy hydrogen-isotopes/helium plasma exposure causes blister and hole structure on tungsten surface 1-4). These damages could enhance the above-mentioned problems. Identification of the physical mechanism of blister and hole formation on material surface is very important from these points of view. It can contribute to a proper selection of damage-free plasma-facing material.

2. Experimental set-up

Ten samples of powder metallurgy tungsten (PM-W) with 0.2 mm in thickness and 99.95 % in purity and one single crystal W (SC-W) with 0.5 mm in thickness and 99.95 % in purity provided by Nilaco Co. were used in the experiments. PM-W samples were mirror-finished.

The five samples W1 ~ W5 were exposed to He and/or D plasma in the linear divertor plasma simulator NAGDIS-I. Figure 1 shows the

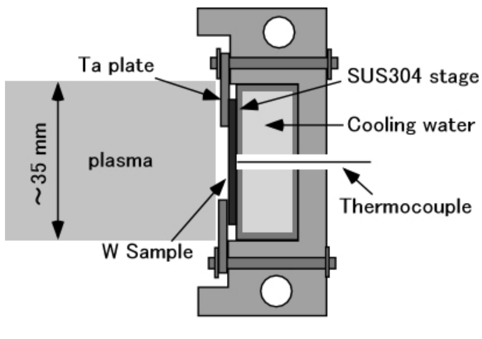

FIG.1 . Schematic view of sample stage.

schematic view of target stage and the experimental configuration of the device. Samples were exposed to plasmas of about 35 mm in diameter. The samples were fixed on the water-cooled target stage with tantalum clip fastening. The temperatures of the samples were measured by K type thermocouple that was contacted with the backside of the sample. The electrical potential of the sample was controlled by a biasing voltage with respect to the vacuum chamber. The value of the incident ion energy was determined by the potential difference between the plasma and the target. The ion flux was found from the ion saturation current to the target. The value of fluence was given by the product of ion flux and exposure time.

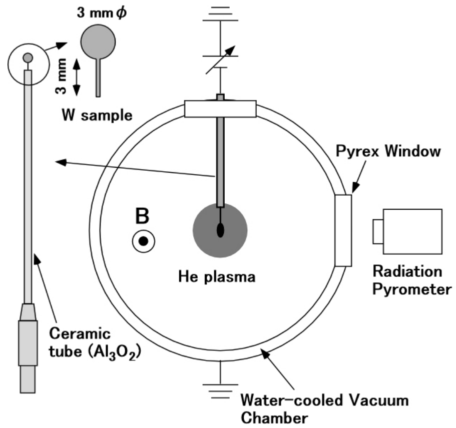

FIG.2 . Schematic view of target probe and experimental configuration.

The six samples W6 ~ W11 were exposed to He or D/He mixture plasma in NAGDIS-II. Four samples W6 ~ W9 were annealed at 2000 K for 1800 s before He exposure, while no pre-treatment were done on W10 and W11. Figure 2 shows the schematic of the target probe and the experimental configuration. Surface temperatures of samples in NAGDIS-II were measured by a radiation pyrometer through the pyrex window.

3. Experimental results and discussions on deuterium blister

3.1 Deuterium blister formation and impurity generation

Five PM-W samples were exposed to D plasma. Detailed experimental conditions are given in the TABLE I. A mirror-finished PM-W W1 was exposed to D plasma at 500 K for 10800 s with a fluence of 2.7×10^{25} m^{-2} after pre-annealing at 700 K for 7200 s. Figure 3 (a) shows the scanning electron microscopy (SEM) image of large blisters about 100 - 200 μm in diameter formed on the surface. Many small blisters about 10 μm in diameter are also observed on the surface and some small blisters exfoliate their dome-caps as indicated in Fig. 3 (b). This result

TABLE I: Pre-treatments and experimental conditions of samples exposed to D plasma

	W1	W2	W3	W4 [4]	W5 [4]
Pre-treatment	Annealed at 700K for 7200 s	Annealed at 700K for 7200 s	He exposure at 700 K for 7200 s	As-received	He exposure at 700 K for 7200 s
Surface condition	Mirror-finished	Scrached	Mirror-finished	Mirror-finished	Mirror-finished
Surface temp. [K]	500			550	
Fluence [/m^2]	2.7E+25			3.0E+25	
Ion flux [/m^{2}s]	2.5E+21			4.0E+21	
Exposure time [s]	10800			7200	
Ion energy [eV]	80			80	

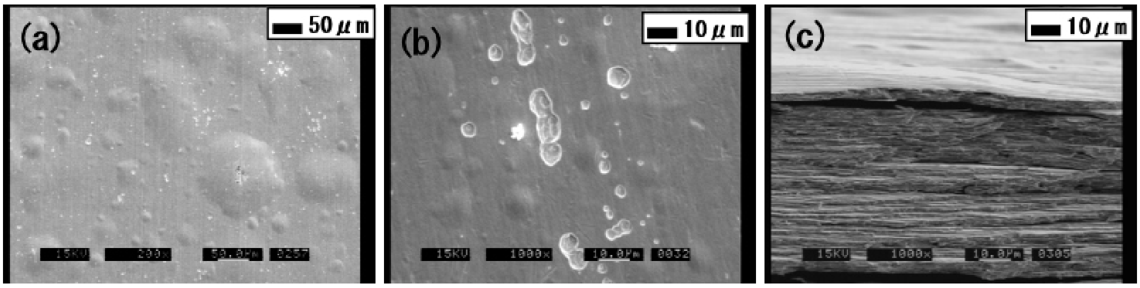

FIG.3 . Deuterium blister formation on mirror-finished W sample W1. (a) Surface view, (b) blister exfoliation and (c) cross-section view of a blister

indicates a possibility that high Z micron-sized impurities are generated due to the blister formation. Figure 3 (c) shows a cross-section of a blister on the surface. A layered structure, which was produced by the press-roll manufacturing process, is clearly seen on the cross-section. Blistering occurs by cleaving along the stratified layers as reported in the previous paper 5).

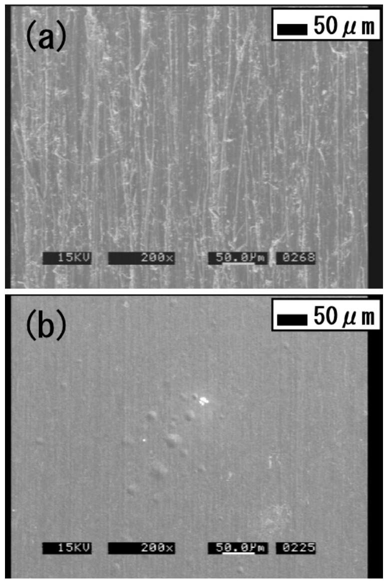

FIG.4 . Surface view of sample W2 and W3 after deuterium plasma exposures. (a) W2: mechanical polished surface and (b) W3:pre-exposed with He plasma

3.2 Suppressions of blister formation by mechanical polishing and He pre-exposure

A mirror-finished PM-W W2 was annealed at 700 K for 7200 s like W1. The purpose of pre-annealing at 700 K for 7200 s on W1 (Fig. 3 (a)) and W2 was to have the same conditions in the temperature history with that of the sample W3 which was pre-exposed to He plasma at 700 K for 7200 s. After the annealing, the sample W2 was mechanicaly polished with commercially available abrasive paper (Kovax Co. #P80), followed by ultrasonic cleaning. The sample W3 with mirror-finished surface was pre-exposed to He plasma at 700 K for 7200 s with the incident ion energy of 90 eV for He ions with a fluence of 2.7×10^{25} m^{-2}. He pre-exposure on W3 at 700 K did not cause any visible surface modification, such as blistering and hole structure. The samples W2 and W3 were exposed to D plasma with the same condition as W1. Figure 4 shows the surface modifications of the samples after D exposures. No blister formation was seen on the sample W2. The surface after D exposure does not change compared with the surface before D exposure. It is not clear why blister formation was suppressed on the scratched surface of W2. Probably, the mechanical polishing can create

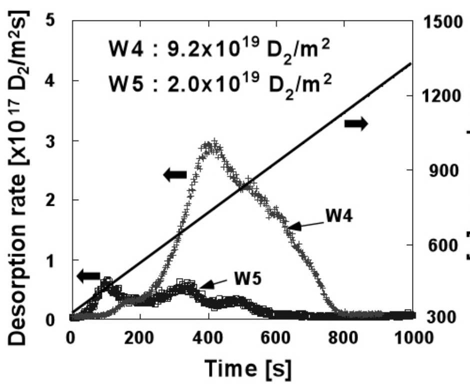

FIG.5 . Thermal desorption spectra of deuterium. (a) W4: virgin W and (b) W5: pre-exposed with He plasma

many small cracks on the surface, which could be paths to the surface for the diffusing D atoms, resulting in no D accumulation in the substrate, which is necessary for cleaving the stratified layers (see Sec. 3.1). Although some blisters are seen on W3 (Fig. 4 (b)), the size and the number of the blisters drastically reduced compared with those of W1, which means that the He plasma pre-exposure produces a suppressive effect for the blister formation on the mirror-finished PM-W. An interesting result that He plasma pre-exposure on PM-W sample at 700 K for 7200 s decreases the D retention compared with that of virgin PM-W was observed 4). Figure 5 shows the thermal desorption spectrum of D_2 for the two samples W4 and W5 as reported in the Ref. 4. The sample W4 without pre-treatment was exposed to D plasma at 550 K for 7200 s with a fluence of $3x10^{25}$ m^{-2} with the incident ion energy of 80 eV for D ions at a flux of $4x10^{21}$ $m^{-2}s^{-1}$. The sample W5 was pre-exposed to He plasma at 700 K for 7200 s ($1.8x10^{25}$ m^{-2}, 20 eV, $2.5x10^{22}$ $m^{-2}s^{-1}$) followed by subsequent D plasma exposure with the same conditions as W4. The D retention of W5 was $R_{D2W5} = 2.0x10^{19} m^{-2}$, which was about one-fifth of that of W4 ($R_{D2W4} = 9.2x10^{19}$ m^{-2}). No blisters were formed both on W4 and on W5 due to short exposure time. Then almost desorbed D atoms/molecules were trapped in interstitial trap sites. Two possibilities were suggested for the reason why D retention of He pre-exposed sample was reduced compared with that of virgin sample. The first was that He pre-exposure at 700 K gives an annealing effect for the sample, which removed some trap site for D atoms/molecules such as strain. The second was that pre-occupation of trap sites due to He atoms decreases the effective number of trap sites available for D atoms. However, large blisters 100-200 μm in diameter as formed on the pre-annealed sample W1 were observed on an non-annealed mirror-finished PM-W (not shown here) after D exposure with the same conditions as W1, which means that pre-annealing at 700 K for 7200 s does not have special effect for blistering suppression and probably for D retention on PM-W. Therefore, it is valid to consider that incident He ions were trapped in the surface layer to fill intrinsic interstitial defects in PM-W, which decreases the effective number of trap site available for D atoms. In addition, this He-enrich surface layer could be a diffusion barrier for incident D atoms. D desorption of W4 occurs over a broader temperature range (from RT to 1150 K) than that of W5 (from RT to 900 K). This indicates a possibility that D atoms in the sample without He pre-exposure were released from the surface after diffusing from the trap sites existing deep inside the substrate. That is, D atoms could diffuse deep into the virgin substate, while they could not do so due to the diffusion barrier of He-enriched layer in the case of the sample with He pre-exposure.

4. Experimental results and discussions on micron-sized helium bubble and hole structure

4.1 Hole growth and surface degradation

Hole structure and micron-sized He bubbles are formed on W surface especially in a higher surface temperature range than the recrystallization temperature (1400-1500 K for W). Figure 6 shows the dependence of the fluence for the hole/bubble growth on PM-W samples exposed to He plasma at 2200 K with 25 eV He ions at a flux of $8x10^{22}$ $m^{-2}s^{-1}$. Submicron-sized holes and bubbles were already appeared on the sample W6 after only 1000 s exposure with a fluence of $8x10^{25}$ m^{-2}. They grew to micron-sized hole/bubble after 10000 s exposure with a flunece of $8x10^{26}$ m^{-2}. The surface became rough and ragged after 75000 s exposure. Although holes/bubbles did not grow so much in the vertical direction, bubble coalescence spread over the whole surface and created a labyrinth-like structure as indicated in Fig. 6 (c2).

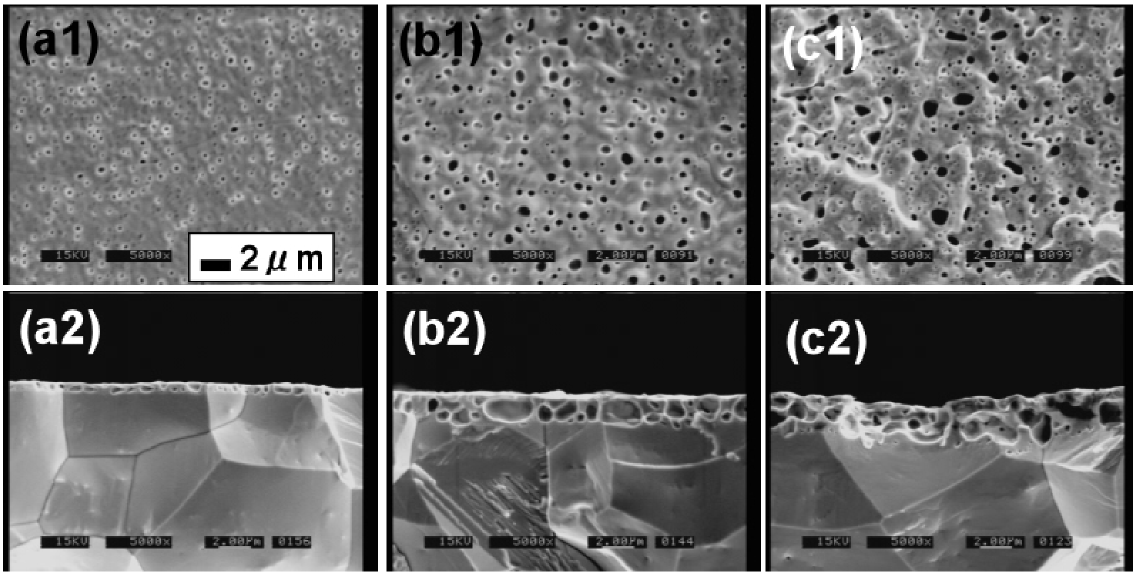

FIG.6 . Fluence dependence for Hole/He-bubble growth. (a1) W6:1000 s exposure, (b1) W7:10000 s exposure and (c1)W8:75000 s exposure. (a2), (b2) and (c2) are cross-section view of each sample.

4.2 Hole formation on single crystal tungsten

In order to investigate the crystal structure dependence of hole formation, one single crystal W sample W9 was exposed to He plasma with the same conditions of W7 (2200 K, 8×10^{26} m^{-2}, 8×10^{22} $m^{-2}s^{-1}$, 10000 s, 25 eV). Holes are observed on the surface similar to the PM-W. Micron-sized He bubbles are also seen within the depth of 5 µm from the top surface. Although diameters of holes on the SC-W are several times larger than those on the PM-W sample W7, it may be said that there is no essential difference between the hole formations in PM-W and SC-W. One characteristic difference is that no grain boundaries are observed on the surface or even on the fractured surface of the SC-W (Fig. 7 (c)).

Intrinsic vacancies in SC-W are much less than those of in PM-W and no radiation-induced vacancies were produced during the exposure due to a very low incident ion energy (25 eV). Thermal vacancy would be considered to be a vacancy supply source, which is necessary for the nucleation and growth of He bubbles.

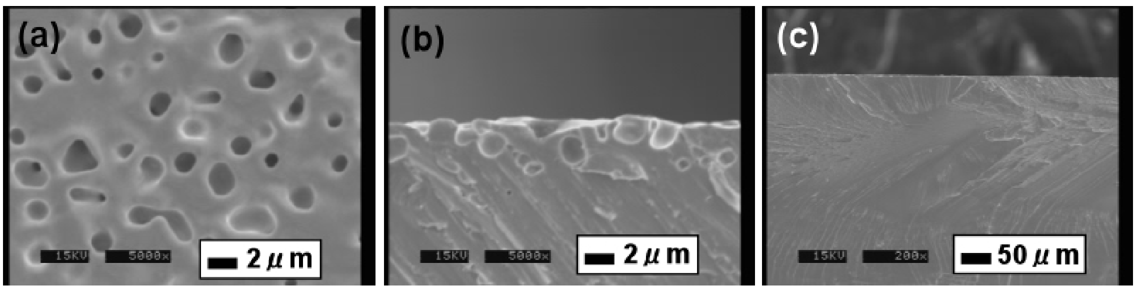

FIG.7 . Hole structure on single crystal W sample W9 after helium plasma exposure at 2200 K. (a) surface, (b) cross-section near the surface and (c) wide view of cross-section.

4.3 Are there any suppression techniques for micron-sized He bubble?

As we reported in the previous paper 4), W surface with a hole structure increases D retention compared with that of the virgin W surface. It is considered that sublimation of W atoms on the surface is very much enhanced due to the decrease of the heat capacity and conduction of the surface layer with hole/labyrinth-like structure. It can increase the impurity contamination of the core plasma in fusion devices. A technique for the suppression of hole structure on W surface is therefore required to solve these problems. Figure 8 shows surface modifications of two PM-W samples W10 and W11 after D and He mixture plasma exposure at temperatures of 1150 K for W10 (Fig. 8 (a)) and 1700 K for W11 (Fig. 8 (b)) with a fluence of 1 x 10^{26} m^{-2} with 10 eV ions for 7200 s 6). Neither D blister nor hole structure was formed on the surface of W10 while clear hole structure was seen on the surface of W11. No D desorptions were observed on the two samples. Almost all D atoms might have been released from the substrate during the exposures due to high temperatures. The hole structure will not be formed even when the temperature is higher than 1500 K if the incident ion energy of He ions is less than about 5 eV because the surface potential barrier of W prevents implantation of He 3,6). However, it seems to be difficult to decrease the incident ion energy below 5 eV even in detached plasmas. Therefore, keeping the surface temperature of W below the recrystallization temperature is the best way to suppress the nucleation and the growth of micron-sized hole structure.

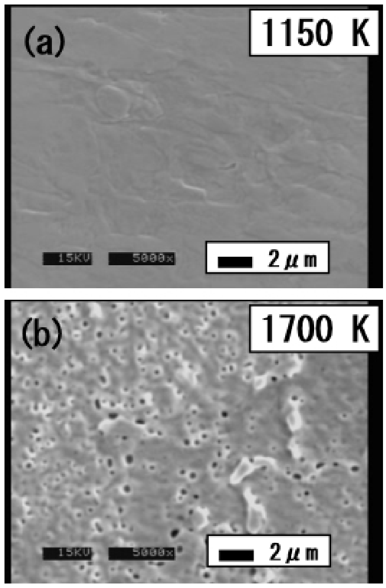

FIG.8 . Surface temperature dependence for hole formation with D and He mixture plasma exposure 6). (a)W10: at 1150 K and (b)W11:at 1700 K

5. Summary

Powder metallurgy tungsten samples are exposed to low-energy and high-flux deuterium/helium plasma to investigate the formation mechanisms and suppression techniques of blister and hole structure. The blistering occurs by cleaving along the stratified layers that were formed during roll-press manufacturing process. Exfoliated dome caps of relatively small blisters less than 10 μm in diameter released from the surface could become micron-sized high Z impurities. Mechanical polishing on mirror-finished powder metallurgy tungsten surface by abrasive paper exhibited a very favorable result that no blisters were formed on the scratched surface. Although small blisters with a few tens of μm in diameter were still formed, the blister formation was drastically suppressed on a mirror-finished PM-W surface which was pre-exposed to a He plasma at 700 K for 7200 s. It is believed that the He-enriched surface layer prevents D atoms from diffusing further deep inside of the material, which was deduced from the D desorption spectra of another He pre-exposed sample.

Micron-sized hole structures were observed both on powder metallurgy tungsten and on single crystal tungsten at higher surface temperature than the recrystallization temperature of W even with the incidental ion energy of He is below the threshold energy of ion displacement. There is no essential diference in hole formation between them. Bubble coalescence spreads over whole surface resulting in labyrinth-like structure as He ion fluence increases. Micron-sized hole structure is suppressed at the surface temperature below the recrystallization temperature of W.

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