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Microscopic deformation of tungsten surfaces by high energy and high flux helium/hydrogen particle bombardment with short pulses

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High energy and high flux helium and hydrogen particles were irradiated with polycrystalline tungsten specimen by using neutral beam injection (NBI) facility in National Institute of Advanced Industrial Science and Technology (AIST). Incidence energy and flux of NBI shot are 25 keV and 8.9×10^{22} ions/m^{2}s, respectively. Duration time of each shot was 30 ms with 6 min interval. Surface temperature may be reached over 1800 K. In the cases of helium irradiation, total fluence was selected 2 cases of 1.5×10^{22} ions/m^{2}s and 4.1×10^{22} He/m^{2}s. The former case, large sized blisters with the diameter of 500 nm were densely observed. While, the latter case, blisters was disappeared and fine nano-branch structures appeared instead of blisters. Cross-sectional transmission electron microscope (TEM) observation was performed by using focused ion beam (FIB) technique. Fig. 1 shows that TEM image after irradiated with 4.1×10^{22} He/m^{2}s. It was clear that very dense fine helium bubbles with the size of 1-50 nm were observed, the density of tungsten matrix was drastically decreased by void swelling with growth of helium bubbles. In the hydrogen irradiation case, these damages were not observed.

Keywords: Helium, Hydrogen, High energy and high fluence, Tungsten, Nano-scale structure, Blister, Bubble

1. Introduction

Tungsten is a candidate of divertor armor materials in international thermonuclear experimental reactor (ITER). In ITER conditions, strong erosion and radiation damages on the divertor surface caused by high heat and particle loading due to the disruptions and edge localized modes (ELMs) are critical issues for achievement of its good thermal and mechanical properties. Thus, heat loading tests or particle loading tests have been performed using several electron beam [1,2] or plasma machines [3,4], respectively. However, evaluation of the synergistic loading effects of high heat and particles with short pulses is necessary for confirming the reliability of tungsten armor tiles. Although some proceeded experiment about it has already been performed [5-7], they have mainly focused on the micro scale or macro scale phenomena. For understanding the fundamental mechanism of the damage evolution, we have to focus on nano-scale structures.

Especially, helium irradiation effects with high heat loads should be noted because helium once injected into metals causes serious damaging effects and it does not release until high temperature due to the strong interaction with lattice defects such as vacancies and dislocation loops [8]. Actually, tungsten irradiation tests to divertor plasmas in Large Helical Device (LHD) indicated that synergistic effects due to high heat and helium/hydrogen particle load lead to damage and erosion of tungsten surface [9,10].

Therefore, the helium or hydrogen irradiation experiment with high heat loading is important for not only elucidation of the damage evolution but also safety assessment of tungsten walls for fusion devices. In this study, high energy and high flux helium and hydrogen particles were irradiated with polycrystalline tungsten specimen by using neutral beam injection (NBI) facility in National Institute of Advanced Industrial Science and Technology (AIST). After the exposure, nano-scale deformation and damage evolution were investigated by using focused ion beam (FIB) fabrication technique and transmission electron microscope (TEM) observation. Furthermore, to investigate the effect of bombardment of helium and hydrogen particles, optical reflectivity was measured by means of spectrophotometer for the wavelength between 190 and 2500 nm. It is expected that from the investigation of correlativity of reflectance and damage evolutions, radiation damages caused by heat and particle loads may be able to be predicted from measurement of optical reflectivity.

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This study is fundamental research focused on the nano-scale damage evolution in tungsten due to the energetic particle bombardment under high heat loading.

2. Experimental procedures

2.1. Material irradiation in NBI facility

High current and high current-density NBI beam system with strong focusing characteristics for measure to the behavior of alpha particle characteristics in nuclear fusion device were successfully developed in AIST [11]. Another application of this focused beam is the irradiation test of materials.

High purity (~99.95 %) powder metallurgy (PM) tungsten specimens of 10 × 5 × 1 mm^3 were mounted in the focused point of the NBI beam by using special specimen stage as shown in Fig. 1 and then, irradiated with helium, hydrogen and He+H NBI beam. Incidence energy and beam current were fixed as 25 keV and 40 A, respectively. The diameter of the focused beam was estimated as ~60 mm. Then, the power density and flux were 300 MW/m^2 and 8.8×10^{22} $particles/m^2s$. Duration time of each shot was 30 ms with 6 min interval. Total irradiation time and fluence of each irradiation pattern were summarized in table. 1. In the case of He+H irradiation, the ratio of hydrogen and helium was estimated at about He : H = 7.9 : 2.1 [12]. Although surface temperature was measured by using an optical pyrometer and thermocouples embedded to just behind the specimen, unfortunately, its exact value was not able to be measured. The expected temperature from optical pyrometer is near 1800 K.

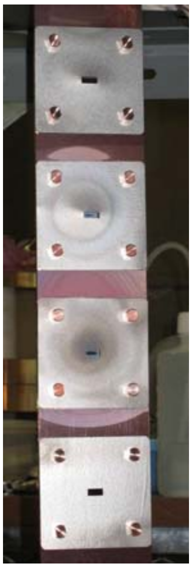

Fig. 1 Specimen stage

2.2. Material analysis

After the exposure, surface morphology was analyzed by means of scanning electron microscopy (SEM). For clarify the depth distribution of nano-scale damages, cross-sectional microscopic observation was conducted by using focused ion beam (FIB) fabrication technique and transmission electron microscopy (TEM) observation.

Furthermore, to investigate the effect of bombardment of helium and hydrogen particles on optical reflectivity, change of optical reflectivity was measured by means of spectrophotometer.

	Pulse x shot	Total Irr. time	Power density	Fluence
He	~30ms x 7shot	171ms	$300MW/m^2$	$1.5x10^{22}He/m^2$
	~30ms x 16shot	461ms	$300MW/m^2$	$4.0x10^{22}He/m^2$
H	~30ms x 5shot	146ms	$300MW/m^2$	$1.3x10^{22}H/m^2$
He+H	~30ms x 15shot	447ms	$300MW/m^2$	$3.9x10^{22}He+H/m^2$

Table 1 Total irradiation time, power density and total fluence for each irradiation pattern.

3. Results and discussions

3.1. Surface morphology

Fig. 2 shows surface photo and SEM image of tungsten specimens after irradiated with helium, hydrogen and He+H beams. Irradiated area on specimens was about 8 × 3 mm^2. In the cases of helium irradiation, total fluence was selected 2 patterns of 1.5×10^{22} He/m^2 and 4.1×10^{22} He/m^2. The former pattern, surface color was changed to gray and large blisters with the diameter of 500 nm were densely observed. While, the latter pattern, blisters were disappeared due to the sputtering erosion, and surface become smoother at low magnification SEM image. However, at high magnification image, fine nano-branch structures appeared instead of blisters. This structure must be affected the degradation of optical reflectivity in visible wavelength, actually, the surface color has been turned to dark brown. The detailed discussion of optical reflectivity will be mentioned in section 3.5.

In the case of hydrogen irradiation, although intensive grain growth was observed, remarkable deformation did not show any sign. These discrepancies between hydrogen and helium seem to be the result of the difference of the behavior of them in metals. In the case of helium, surface modifications are considered to be due to the formation, coalescence and migration of helium bubbles near the surface during the pulse high heat loading. While, in the case of hydrogen, these effects does not occur. It is known that retention of hydrogen isotopes in tungsten is low in general, and this is one of

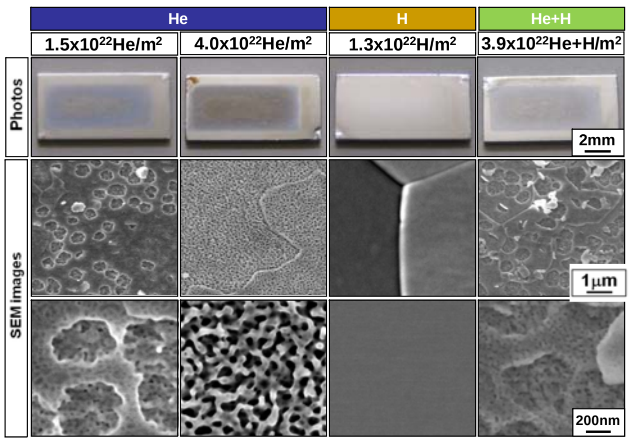

Fig. 2 Surface photo and SEM image of tungsten specimens

the advantages of this material [13].

Furthermore, in the case of simultaneous irradiation of He and H, surface morphology was quite similar to the lower fluence case of helium irradiation (1.5×10^{22} He/m^2). This means that damage evolution of the surface does not depend on the hydrogen irradiation.

3.2. Depth distribution of nano-scale damages

To clarify depth distribution of radiation damages, we need cross-sectional image of the sub-surface region in nanometer level. FIB fabrication technology was used for obtain the cross-sectional TEM samples for nano-observations. Fig. 3-(a~d) shows TEM images of four exposure patterns. In the case of lower helium irradiation (a), helium bubbles with size of about 1~50 nm were densely observed up to the depth of 200 nm. One should note that many of the bubbles are larger than 20 nm and some of them have an oddly shaped image like an ellipse or even a gourd. This indicates that the bubbles have grown by coalescence with the abrupt temperature increase and the non-equilibrium shape is "frozen" by rapid cool-down after the termination of the NBI beams. Although average penetration depth of 25 keV-He in tungsten was estimated to be about 60 nm by TRIM-code, heavy damaged region was much deeper than that of this value. We can consider that this discrepancy is caused by not only diffusion of injected helium atoms but also void swelling of the surface. In the case of (b), highest helium irradiation, helium bubbles with size of about 1~100 nm were densely observed with nano-branch structures, many of the bubbles are larger than 50 nm. It was much larger than that of case (a). Thus, density of tungsten matrix was drastically decreased by void swelling, and its depth distribution was over 300 nm. On the surface of this specimen shown in Fig. 2, blisters lid which can be observed in case (a) was already disappeared due to the sputtering erosion. In this irradiation stage, the accumulation of helium atoms by implantation in the matrix and loss by sputtering erosion and exfoliation of blisters has reached a balance. Therefore, if the helium were continuously irradiated, nano-branch structures will not be disappeared.

In the case of hydrogen irradiation, (c), nano-scale damages scarcely formed, which corresponds to the surface observation result as shown in Fig. 2. It is considered that since hydrogen atoms scarcely interact with lattice defects, radiation damages could not form even at the high energy and high flux condition.

Furthermore, result of a simultaneous irradiation of hydrogen and helium, (d), shows that size and density of helium bubbles and its depth distribution were very similar to the case (a). As mentioned above, damage evolution does not depend on the hydrogen irradiation. In contrast, helium once injected into metals, they cause serious damaging effects due to the strong interaction with lattice defects such as vacancies and dislocation loops. Thus, helium irradiation effects in tungsten indicate that serious consideration should be given to the surface erosion of tungsten walls.

3.5. Optical reflectivity

Fig. 4-(a~d) shows the optical reflectivity of tungsten specimens after exposed to NBI beams. Reflectivity of virgin specimen was also plotted together. It is clear that strong reduction of reflectivity has occurred in all helium irradiation cases (a, b, d). In particular, about 500 nm or less is remarkable. Referring to our previous study [14], it is considered that reduction of optical reflectivity is due

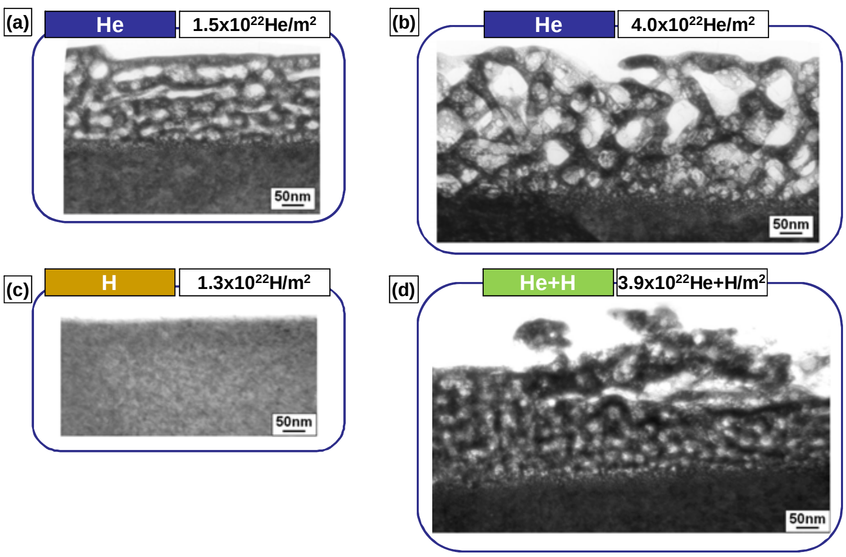

Fig. 3 Cross-sectional TEM images after irradiated with
(a): 1.5×10^{22} He/m^2, (b): 4.0×10^{22} He/m^2, (c): 1.3×10^{22} He/m^2, (d): 3.9×10^{22} He+H/m^2

to the multiple scattering of light by the dense helium bubbles in the sub-surface region. In the case of (a) and (b), the spectrum is very similar in the range of 190 to 2500 nm. We can consider that since surface morphology and depth distribution of nano-scale damage of them were similar in Fig. 2 and Fig. 3, reduction of reflectivity may be dependent on them.

On the other hand, highest helium irradiation, (b), although drastic degradation has occurred in short wave length (190~1000 nm), its recovery has identified in long wave length (2000~2500 nm). We can consider the following mechanisms for this phenomenon. Reduction of reflectivity must be due to the surface roughening and fine bubble formation, drastic degradation in short wave length was caused by formation of nano-branch structures with nano-scale helium bubbles as shown in Fig. 3-(b). While, in the long wave length region, its recovery has been caused by smoothening the surface due to the progress of the sputtering erosion as shown in Fig. 2.

In the case of hydrogen irradiation, (c), although slight degradation was observed in the long wave length (1500-2500 nm), remarkable degradation was not confirmed in whole wave length regions. We should note that effect of helium bombardment for optical reflectivity is also much higher than that of hydrogen bombardment. This result is important for the design and operation of plasma diagnostics using first mirror.

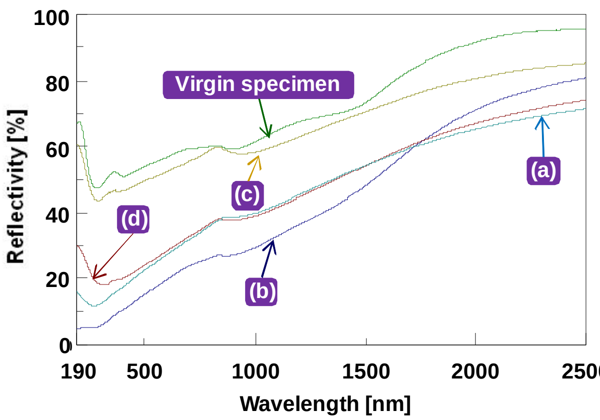

Fig. 4 Optical reflectivity of specimen after exposed to NBI beams. (a): 1.5×10^{22} He/m^2, (b): 4.0×10^{22} He/m^2, (c): 1.3×10^{22} He/m^2, (d): 3.9×10^{22} He+H/m^2

4. Summary

High energy and high flux helium and hydrogen particles were irradiated with tungsten specimen by using NBI facility in AIST, and then, nano-scale deformation and damage evolution in tungsten were investigated.

In the case of helium irradiations, very dense fine helium bubbles with the size of 1~100 nm were observed, the density of tungsten matrix was drastically decreased by void swelling with growth of helium bubbles. In the hydrogen irradiation case, these damages were not observed.

To investigate the effect of bombardment of helium and hydrogen particles, optical reflectivity was measured. The hydrogen irradiation case, it was scarcely changed before and after irradiation. However, reduction of reflectivity has occurred in helium irradiation case, and it near 500 nm or less and also infrared region was remarkable. It is considered that reduction of optical reflectivity is due to the multiple scattering of light by the helium bubbles in the sub-surface region.

High energy and high flux helium irradiation effects in tungsten indicate that serious consideration should be given to the surface erosion of tungsten walls. Furthermore, if we use the tungsten for first mirror, its design and operation will become also serious problems.

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