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Effects of high heat flux hydrogen and helium mixture beam irradiation on surface modification and hydrogen retention in tungsten materials

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ABSTRACT

High heat flux experiments using a hydrogen–helium mixture beam have been carried out on powder metallurgy tungsten (PM-W) and ultra fine grain W–TiC alloy (W–0.5 wt%TiC-H_2). The energy of is 18 keV. Beam flux and heat flux at the beam center is 2.0×10^{21} atoms/m^2s and 7.0 MW/m^2, respectively. Typical ratio of He/D ion is 0.25. Beam duration is 1.5–3 s and interval of beam shot start is 30 s. The samples are irradiated up to a fluence of 10^{22}–10^{24} He/m^2 by the repeated irradiation pulses. After the irradiation, surface modification by the irradiation and hydrogen retention, surface composition have been investigated. Surface modification by hydrogen–helium mixture beams is completely different from results of single beam irradiation. In particular, mixture beam irradiation causes remarkably high hydrogen retention.

1. Introduction

Tungsten materials are potential candidates of the divertor armor and first wall materials of the next fusion devices due to their very low erosion yield and high temperature properties. However, heat load, hydrogen and helium atoms from the plasma, which affect on damage accumulation and mechanical properties, may reduce these superior properties of tungsten.

Hydrogen or helium single beam irradiation experiments have been performed to investigate the synergistic effects of the heat loading and the particle loads (hydrogen or helium) on modification of tungsten. Surface modification by helium and hydrogen beam heating is completely different from results of electron beam heating [1]. In particular, helium beam heating causes remarkable surface modification such as the fine-scale, rough surface at high temperatures due to strong interaction with helium and lattice defects [2]. This modification may influence thermal properties such as thermal conductivity, mechanical properties and tritium retention of the material. Simultaneous irradiation of hydrogen and helium beam has been investigated [3]. However, the armor materials are simultaneously exposed to high heat flux hydrogen isotopes and helium as well as electrons. Very little information is available on the combined effects of high heat flux hydrogen and helium particle implantation under the high heat loading.

In the present study, high heat flux hydrogen and helium mixture beam irradiation experiments have been performed to investigate the synergistic effects simultaneous implantation of high heat flux hydrogen and helium on modification and hydrogen retention of tungsten materials.

2. Experimental

Samples used in the present experiments are powder metallurgy tungsten (PM-W) and ultra fine grain W–0.5 wt%TiC alloy with MA in H_2 which is noted by W-0.5 wt%TiC-H_2. The W–TiC alloy have been developed for good resistance of radiation damage due to neutron irradiation and high temperature embrittlement [4].

Sizes of the PM-W, with size $20 \times 20 \times 5$ mm and $10 \times 10 \times 1$ mm were used. Purity of PM-W was 99.99%. Size of W–TiC alloy was $10 \times 10 \times 1$ mm. The sample surfaces were electropolished.

The samples are mechanically mounted on a Cu holder, actively water cooled and are repeatedly irradiated by hydrogen and helium mixture beam (18 keV, $\sim 2 \times 10^{21}$/m^2s, $\sim$7 MW/m^2, 1.5 $\sim$ 3s) up to a fluence of the orders between 10^{22} and 10^{24}/m^2 using the Divertor Acceptance Testing System (DATS) in JAEA [1].

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Ratio of hydrogen and helium beam is determined by an optical spectroscopic measurement. The ratio of hydrogen and helium beam was calculated by comparison of peak counts of emission from hydrogen and helium of the single hydrogen beam, the single helium beam and the hydrogen–helium mixture beam with same heat flux. Typical ratio of He/D ion is 0.25. In addition to the mixture beam irradiations, single beam of hydrogen and helium irradiation experiments are also carried out to compare with the results by the mixture beam irradiation. The surface temperature of the sample is measured with a two-color optical pyrometer and with a two-dimensional surface temperature distribution of the samples is also monitored with an IR camera. Maximum temperatures of the samples during the irradiations change from 1124 to 2244 °C, which depends on heat load, duration time of the beam, sample size and thermal contact with the sample and Cu holder. After the irradiation experiments, surface modification is examined with a scanning electron microscope (SEM). In addition, quantitative analyses of the implanted hydrogen and helium in the samples are carried out by means of elastic recoil detection (ERD) by using an oxygen ($^{16}O^{4+}$) analyzing beam with the energy of 5.0 MeV. The compositional depth profiles near the surface of the specimens were measured using TOF-SIMS. A Ga and a Cs ion beam with an energy of 3 and 25 keV, incident on the specimen at 45°, was used as primary and sputter ion beam, respectively. The secondary negative ions emitted from the samples surface by sputtering were collected by a time of flight mass spectrometer. In addition, depth profiles of dpa and hydrogen and helium deposition rate in W irradiated with 18 keV H and He ions were calculated by the TRIM-code.

3. Results

A temperature evolution during hydrogen–helium mixture beam irradiation is shown in Fig. 1. The surface temperatures gradually increase and reach a peak temperatures and starts to decrease after beam turn-off. Temperature changes also depend on the size of the samples, heat flux and thermal contact with the sample and the Cu holder.

In the case of PM-W, surface modification such as micro-relief occurs and corresponds to the grains from 10 to 100 μm. As the sputtering depends on the grain orientation, the surface modification may be formed by micro-structure change near the surface and erosion due to the sputtering. In addition, grain dependence on modification by hydrogen–helium irradiation is relatively small. Shown in Fig. 2 is typical SEM images taken from the surface of PM-W irradiated to 1.0×10^{24} atoms/m^2 of (a) hydrogen beam, (b) helium beam and (c) Hydrogen and helium mixture beam. It is clearly seem that surface modification due to the mixture beam irradiation is different from that of single hydrogen and helium beam irradiation. Roughness of the surface of mixture beam irradiation is smaller than that of single beam irradiation. In the case of the mixture beam irradiation at PM-W, surface is not rough comparing with that of single hydrogen and helium beam.

On the other hand, in the case of W–0.5 wt%TiC-H_2 alloy, surface modification is uniformly formed. The reason may be that W–0.5 wt%TiC-H_2 alloy has very fine grains with the size of a few 100 nm. Shown in Fig. 3 is SEM images taken from the surface of W–0.5 wt%TiC-H_2 alloy irradiated to 7.1×10^{23} atoms/m^2 of (a) hydrogen beam, (b) helium beam and (c) hydrogen and helium mixture beam. The surface modification depends on kind of

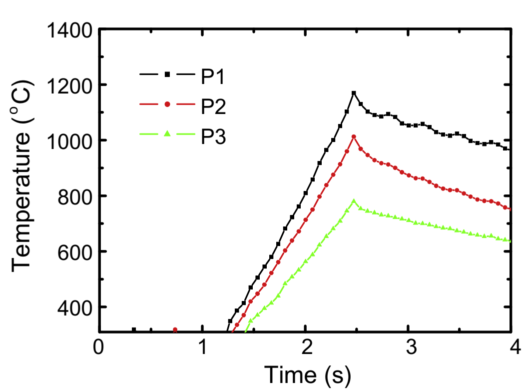

Fig. 1. Time evolution of surface temperatures measured by IR camera by H–He beam irradiation. Beam duration is 2.5 s. P-1: 7 MW/m^2, 20 × 20 × 5 mm, P-2: 5 M/W/m^2, 10 × 10 × 1 mm, P-3: 1.8 MW/m^2, 20 × 20 × 0.1 mm.

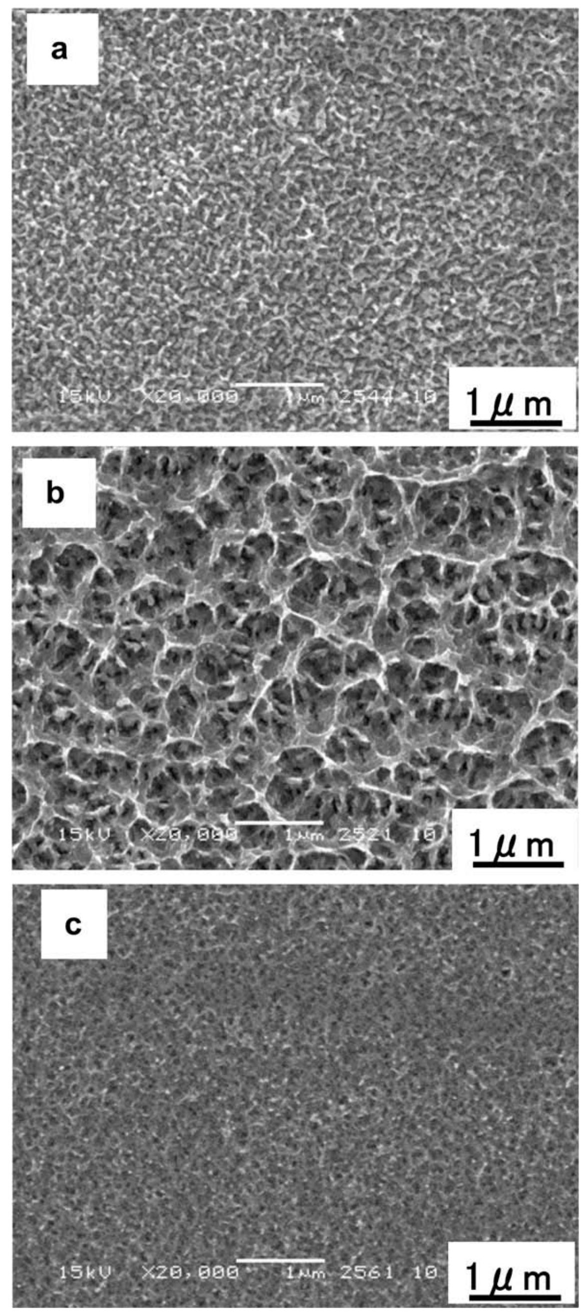

Fig. 2. SEM image taken from the surface of PM-W irradiated to 1.0×10^{24} atoms/m^2. (a) Hydrogen beam (7 MW/m^2). The peak temperature of surface is 1127 °C. (b) Helium beam (7 MW/m^2). The peak temperature of surface is 1258 °C. (c) Hydrogen and helium mixture beam (7 MW/m^2). The peak temperature of surface is 1165 °C.

irradiation beam. It is clearly also seem that surface modification due to the mixture beam irradiation is different from that of the single hydrogen and helium beam irradiation in the case of W–0.5 wt%TiC-H_2 alloy. In the case of (b), small holes with a diameter of a few 100 nm are observed. These holes may be formed by exfoliation of grains due to embrittlement by helium irradiation. On the other hand, in the case of mixture beam irradiation, surface is relatively smooth comparing with the single beam irradiation.

Fig. 4 shows backscattering spectra for 5 MeV $^{16}O^{4+}$ ion beam (a) and energy spectra (b) of He and H recoil particles from PM-W after irradiated with 18 keV H–He mixture beam at fluence of 1.8×10^{23} atoms/m^2. The peak temperature is 893 °C. The result of RBS measurement indicates that impurities are not detected because the scattering energy and cross section of W is large. In addition, retained He and H in the surface region is detected by ERD.

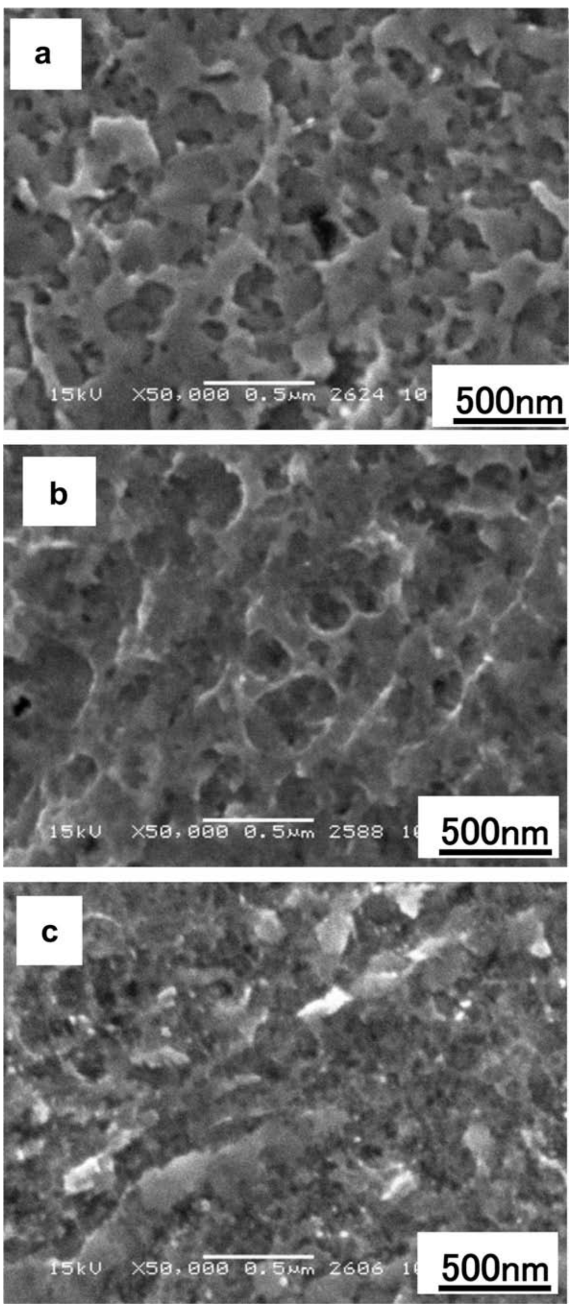

Fig. 3. SEM image taken from the surface of W–0.5 wt%TiC-H_2 irradiated to 7.1×10^{23} atoms/m^2. (a) Hydrogen beam (5 MW/m^2). The peak temperature of surface is 1698 °C. (b) Helium beam (5 MW/m^2). The peak temperature of surface is 1698 °C. (c) Hydrogen and helium mixture beam (5 MW/m^2). The peak temperature of surface is 1677 °C.

Figs. 5 and 6 show concentration depth profiles of H in the PM-W and W–0.5 wt%TiC-H_2 irradiated by H, He and H–He mixture beam estimated using the ERD depth profiles. In the case of He irradiation, large amount of H is retained on the sample surface. On the other hand, depth profile of the mixture beam irradiation has two peaks. One has the surface peak and the other has the peak with a depth of about 200 nm. TOF-SIMS analyses showed that oxygen exists in the near surface.

4. Discussion

In the high heat beam experiments in the present work, the peak sample temperatures were higher than that of the temperatures which hydrogen retain in the W samples. However, hydrogen is retained in the samples. This is believed to be due to the pulse temperature increase. Therefore, it is expected that as the sample cooled after the beam irradiation, the diffusing deuterium atoms already in the sample may become trapped in sites that remain unoccupied during the high temperature in the present irradiation experiment.

It is well-known that He ion implantation can provide strong hydrogen trapping site, which are related to hydrogen adsorption on the He bubbles walls [5] and in the stress/strain field around bubbles [6]. In addition, the beam irradiated surface was activated for creating oxides surface, which play an important role for hydrogen dissociation. In this experiment, the TOF-SIMS analyses shows oxygen was exists near surface and it is expected W-oxides layer was formed. In the case of helium irradiation, hydrogen diffuses from surface to deeper area of the sample regarding from the hydrogen depth profile.

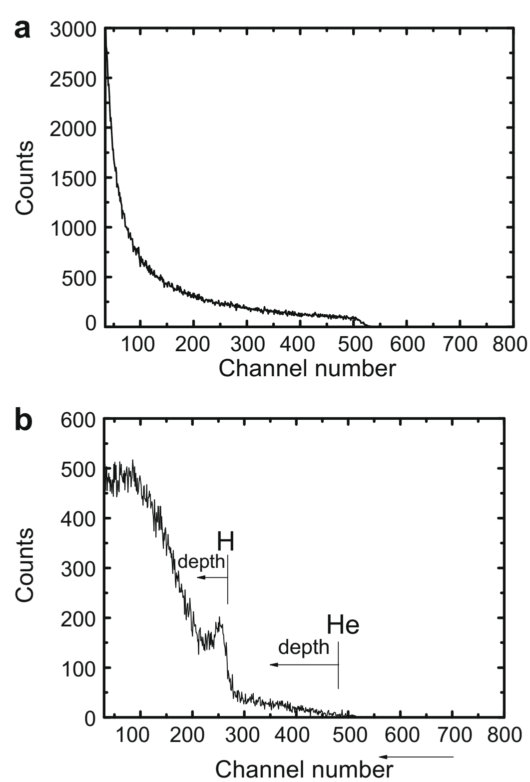

Fig. 4. Backscattering spectra for 5 MeV $^{16}O^{4+}$ ion beam (a) and energy spectra (b) of He and H recoil particles from PM-W after irradiated with 18 keV H–He mixture beam (1.3 MW/m^2) at fluence of 1.8×10^{23} atoms/m^2. The peak temperature is 893 °C.

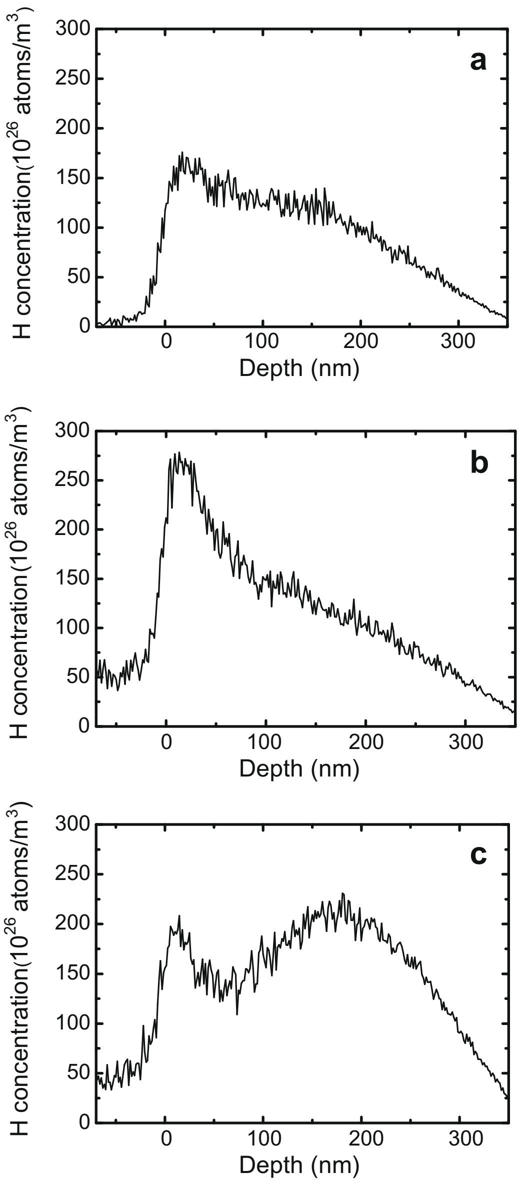

Fig. 5. Concentration depth profiles of H in the PM-W irradiated by H, He and H–He mixture beam. (a) Hydrogen beam (1.3 MW/m^2) to a fluence of 1.8×10^{23} atoms/m^2 at the peak surface temperature 573 °C. (b) Helium beam (3.3 MW/m^2) to a fluence of 5.0×10^{23} atoms/m^2 at the peak surface temperature 1310 °C. (c) Hydrogen and helium mixture beam (1.3 MW/m^2) to a fluence of 1.8×10^{23} atoms/m^2 at the peak surface temperature of 893 °C.

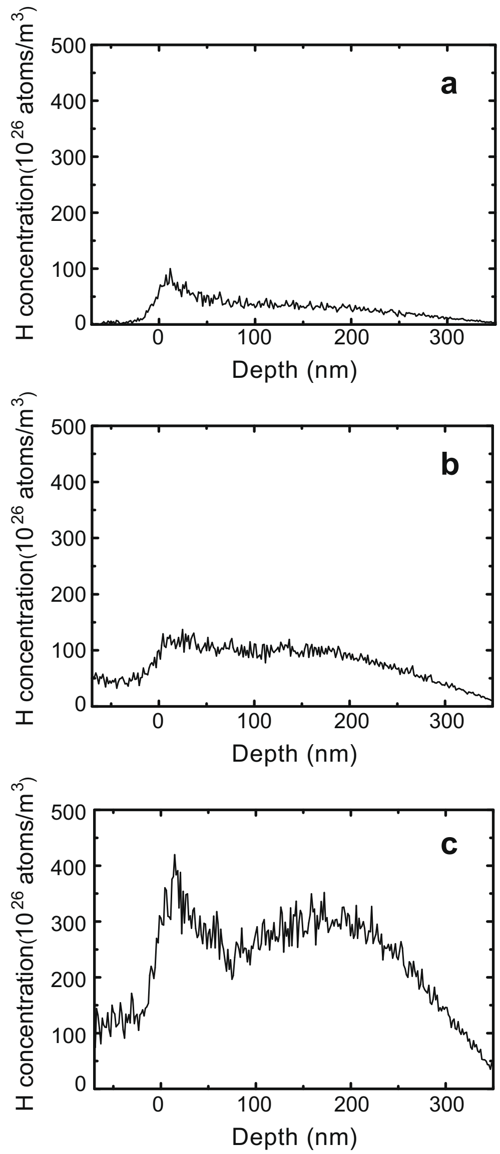

Fig. 6. Concentration depth profiles of H in the W–0.5 wt%TiC-H_2 irradiated by H, He and H–He mixture beam. (a) Hydrogen beam (5.0 MW/m^2) to a fluence of 7.1×10^{23} atoms/m^2 at the peak surface temperature 1213 °C. (b) Helium beam to a fluence (5.0 MW/m^2) of 7.1×10^{23} atoms/m^2 at the peak surface temperature 1698 °C. (c) Hydrogen and helium mixture beam (5.0 MW/m^2) to a fluence of 7.1×10^{23} atoms/m^2 at the peak surface temperature of 1677 °C.

On the other hand, in the case of hydrogen–helium mixture beam irradiation, hydrogen exists on surface and the area with a depth of about 200 nm. It is expected that implanted hydrogen is retained trapping site which is formed by helium beam irradiation. The depth profiles calculated by the TRIM-code show that hydrogen and helium atoms are mostly concentrated near the surface and distributed up to 200 and 140 nm in depth, respectively. In addition, the radiation damage by hydrogen and helium are also mostly concentrated near the surface and distributed up 175 and 120 nm in depth, respectively. In this experiment, vacancy and interstitial atom as well as hydrogen can easily move to deeper area in the sample temperature. As a result, hydrogen exists in the deeper area rather than the implanted area. In the case of W–0.5 wt%TiC-H_2, behavior is similar to that of PM-W; however, quantitative value is different from that of PM-W. In the present irradiation experiments, sample temperature control was very different because the sample was fixed mechanically and thermal diffusion from the sample to Cu holder was very sensitive. Further, systematic investigation is needed to clarify micro-structure dependence for the mixture beam irradiation.

5. Conclusion

High heat flux hydrogen and helium mixture beam irradiation experiments have been performed to investigate the synergistic effects simultaneous implantation of high heat flux hydrogen and helium on modification and hydrogen retention of tungsten materials. Surface modification by hydrogen–helium mixture beams is completely different from results of single beam irradiation. In particular, mixture beam irradiation causes remarkably high hydrogen retention. Therefore, surface modification, erosion and hydrogen isotope retention will be evaluated in consideration of helium simultaneous irradiation.

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