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Microstructural evolution in beryllium by fusion-relevant low energy helium ion irradiation

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Abstract

An in situ transmission electron microscopy (TEM) study was performed to investigate the microstructural changes in beryllium due to helium-ion implantation. Beryllium samples were irradiated by 8 keV helium ions up to 10^{22} He^+/m^2 at various temperatures between room temperature and 873 K. At all the temperatures examined here, dense tiny bubbles were primarily formed in beryllium above a threshold fluence. The size of the tiny bubbles was almost independent on temperature and was at most 2 nm in diameter. The threshold fluence was slightly dependent on irradiation temperature. More significant effect of irradiation temperature was the growth of the bubbles. Above 673 K, the bubbles were observed to grow up to the size of several tens to hundreds of nanometers in diameter. Cavities with hexagonal shape were also observed above 773 K. After the irradiation and simultaneous TEM observation, surfaces of the samples were also investigated by scanning electron microscopy (SEM). Blisters, exfoliation, flake and pinholes were observed to be formed on the surfaces, and they strongly depended on temperature. Thermal desorption spectroscopy (TDS) was also measured using a quadrupole mass analyzer (QMA) to investigate the amount of helium gas released from the irradiated samples as a function of annealing temperature. The TDS spectra showed that most of the helium gas was released at lower annealing temperature for the samples irradiated at higher fluence. Interrelation among the microstructural evolution in beryllium, the changes in surface shape and the behavior of helium gas implanted in the sample was discussed. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Beryllium has recently been identified [1] as the candidate material for the international thermonuclear experimental reactor (ITER) as a first wall material and as well as a divertor material, because of its good thermal conductivity, reduction of plasma radiation, enhancement of deuteron pumping and reduction in incidence of plasma disruption [2–4]. A plasma-facing material (PFM) is exposed to plasma particle bombardment with a result of displacement damage and subsequent microstructural changes due to strong interaction between plasma particles and lattice defects in a PFM. In comparison with radiation damage by fusion neutrons [5], the characteristic response of materials due to plasma-PFM interaction can be induced by higher flux injection of energetic hydrogen and helium particles near the surface.

Our previous work [6] indicated that bubbles and/or cavities were formed in beryllium irradiated by 8 keV-D_2^+ ions at all irradiation temperatures ranging from room temperature to 873 K. Local swelling by the deuterium bubbles, for example, exceeded 30% at 573 K irradiation. Thermal desorption data indicated that deuterium was detrapped between 800 K and 1000 K, showing that hydrogen particles can still be in the metal in the temperature range where beryllium is supposed to be used as a PFM.

Helium irradiation can cause more profound effects on material properties than hydrogen, due to stronger

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interaction of helium with lattice defects [7]. Defect accumulation by helium ions of keV energy range is considered to induce significant hardening and embrittlement even at high temperature. Bombardment of energetic helium ions can result in unrecoverable damage in PFMs and degrade even material bulk properties. It is reported that a 200 nm thick surface damaged layer by helium ion irradiation can result in embrittlement at low temperatures and ductility loss at high temperatures for a bulk sample of molybdenum [8].

In the present paper, we show an in situ observation study with transmission electron microscopy (TEM) to investigate microstructural changes in beryllium due to helium implantation as a function of irradiation fluence for various temperatures. A scanning electron microscopy (SEM) study was also performed to investigate surface changes of beryllium irradiated by helium ions. Thermal desorption spectroscopy measurements were also employed for beryllium samples irradiated at room temperature to know the details of the retention and desorption of the implanted helium and to identify the responsible traps. Finally, we discuss changes in the microstructures and surface shape as a function of the behaviors of helium gas implanted in beryllium.

2. Experimental

Commercial cast grade samples of 98.7 and 98.5 wt% pure beryllium were provided by NGK Insulators, for TEM/SEM and TDS studies, respectively. The beryllium samples contained 1.00–1.20 BeO, 0.10 Fe, 0.04–0.05 Al, 0.02 Mg, 0.14 C and 0.03–0.04 Si in wt%. The samples were prepared for TEM following standard electropolishing procedures using twin jet polishing unit operating at 5–10 V with an electrolyte solution of 400 ml $HClO_3$, 200 ml C_2H_5OH and 200 ml H_2O.

The TEM/SEM samples were irradiated by 8 keV helium ions with a typical beam flux of 2×10^{18} $He^+/m^2/s$ using a duoplasmatron ion gun combined to a JEM-2000EX transmission electron microscopy. This enabled us to perform an in situ TEM observation of a sample being irradiated by gas ions with incident energies between 0.1 and 10 keV. The details of the facility is described elsewhere [9]. TRIM code [10] calculation for the implantation by 8 keV helium ions into beryllium indicates a displacement damage distribution with a peak at 60 nm from the irradiated surface and a helium profile distribution with a peak at 75 nm. The microstructural evolution in beryllium during irradiation by helium ions up to 10^{22} He^+/m^2 was observed in situ by TEM at several irradiation temperatures between room temperature and 873 K. This temperature is just below the dissociation temperature of the helium–vacancy complex in beryllium, 900 K [11]. After the irradiation and simultaneous TEM observations, the surfaces of the samples were also examined by JSM-T330 scanning electron microscopy.

To complementarily know the helium gas behavior as a function of the microstructural and surface changes, thermal desorption spectroscopy (TDS) was also applied for irradiated beryllium samples. The samples for the TDS were irradiated by 8 keV helium ions at various irradiation fluences at room temperature. The amount of helium atoms released from the sample was measured with a quadrupole mass analyzer (QMA) during the annealing up to 1400 K which was just below the transformation temperature between α-Be and β-Be. The ramping rate of the annealing temperature during the annealing was constant at 1 K/s.

3. Results

3.1. In situ TEM observation

Fig. 1 shows the transmission electron micrographs of the microstructural evolution in beryllium irradiated by 8 keV helium ions at various irradiation temperatures as a function of the irradiation fluence. At all examined temperatures, tiny bubbles can be seen in the micrographs. The bubbles were suddenly visualized in whole sight of the TEM micrographs at certain threshold fluence. They appeared as if a kind of phase transition occurred in the metal. The threshold fluence at which the primary bubbles appeared was slightly dependent on irradiation temperature. It was in the order of 10^{21} He^+/m^2 at room temperature irradiation, while it was in the order of 10^{20} He^+/m^2 at higher temperatures. The typical size of the bubbles was very small, ranging from 1 to 2 nm in diameter, which was almost independent on irradiation temperature. The primary bubbles were distributed mainly in the primary damaged subsurface region and their density was extremely high. The saturated value of the apparent areal density of the primary bubbles was about 2×10^{17} m^{-2}. It was also independent on irradiation temperature, showing that the primary bubble formation was a kind of athermal process.

There was a clear temperature dependence of the subsequent behavior of the primary bubbles at higher fluence. At lower temperature regime below 573 K, interconnection of the primary bubbles was observed and so-called labyrinth structure [11] was developed at above 10^{21} He^+/m^2. It is interesting to note that in the 573 K irradiation a half part of the micrographic view suddenly became white, which could be related to the surface change of the sample during irradiation, as described below.

At higher temperature regime, on the other hand, the growth of the bubbles was observed. In the 673 K irradiation case, some bubbles grew up to the size of 50 and 100 nm in diameter at the fluences in the order of

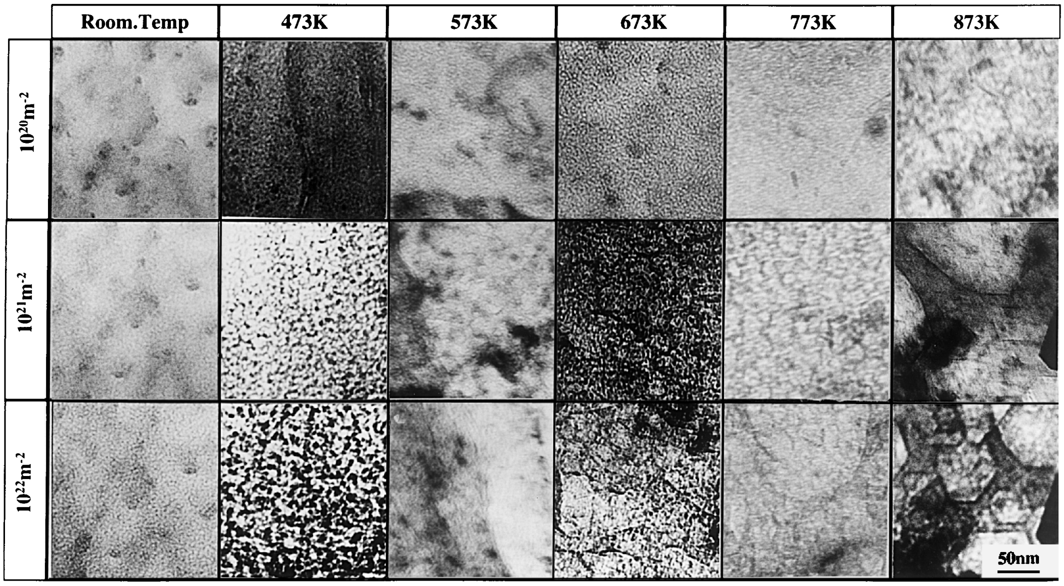

Fig. 1. Transmission electron micrographs of microstructural evolution in beryllium irradiated by 8 keV helium ions at various temperatures as a function of ion beam fluence up to 10^{22} He^+/m^2.

10^{21} and 10^{22} He^+/m^2, respectively. The primary bubbles still existed at the temperature, indicating that the bubble growth largely depends on the distance from the surface. In the 673 K irradiation case, the size of the larger bubbles ranged from 25 to 150 nm in diameter. In the 773 K irradiation, some bubbles were observed to grow up larger at the fluence in the order of 10^{19} He^+/m^2, showing that the threshold fluence for the bubble growth was smaller than that of lower temperature irradiations. Local swelling caused by the bubbles exceeded about 50% for the irradiation of 10^{22} He^+/m^2 at 773 K.

It was observed by TEM at higher temperature regimes that larger bubbles had an amoeba-like structure. The amoeba-like bubbles easily moved in the matrix of the sample and coalesced into even larger ones. Some larger bubbles shrunk and disappeared with a rate of about 1 nm/s. Above 773 K, some large bubbles were observed to be facetted in shape at the fluence in the order of 10^{21} He^+/m^2. The facetted bubble or a cavity was stable and immobile, and its size was about 150 nm in diameter.

3.2. SEM observation

After helium irradiation and in situ TEM observation, the irradiated samples were moreover investigated by SEM. Fig. 2 shows the SEM micrographs of the surfaces of beryllium irradiated by 10^{22} He^+/m^2 as a function of irradiation temperature. As reported for other metals [12], the helium implantation in this fluence regime changed the surface of the samples changed. The surface structure of the irradiated samples strongly depended on irradiation temperature. In the 473 K irradiation, small blisters between 0.1 and 0.2 μm in size were observed as well as large holes between 1 and 2 μm in size. The hole may be a result of the exfoliation of a blister cap or a flake. After irradiation at 573 K holes with two sizes were observed: one was between 2 and 3 μm in diameter and another was between 0.3 and 0.6 μm. Both types of the holes were also observed in the case of 673 K, but the fraction of the smaller size holes was a little greater than after the 573 K irradiation. The total number density of the holes at 673 K was greater than that of 573 K.

At greater temperatures above 873 K, surface structure was clearly different. The blisters or holes of greater size than 1 μm were observed to be roundish, while the submicron-size blisters or holes were scarcely observed at the temperatures. Moreover, in this temperature regime, small pinholes between 3 and 10 nm in size were also observed.

The exfoliation was found to be very temperature dependent, showing the ductile-like and brittle-like shapes at higher and lower temperatures, respectively. For 573 K the exfoliation was observed to be very brittle-like, as shown in Fig. 2, which could correspond to the sudden change visible by TEM (Fig. 1) at the temperature. The formation of the roundish blisters observed at higher temperature can be due to a balance between gas-induced internal pressure in a blister and elasticity of the surface.

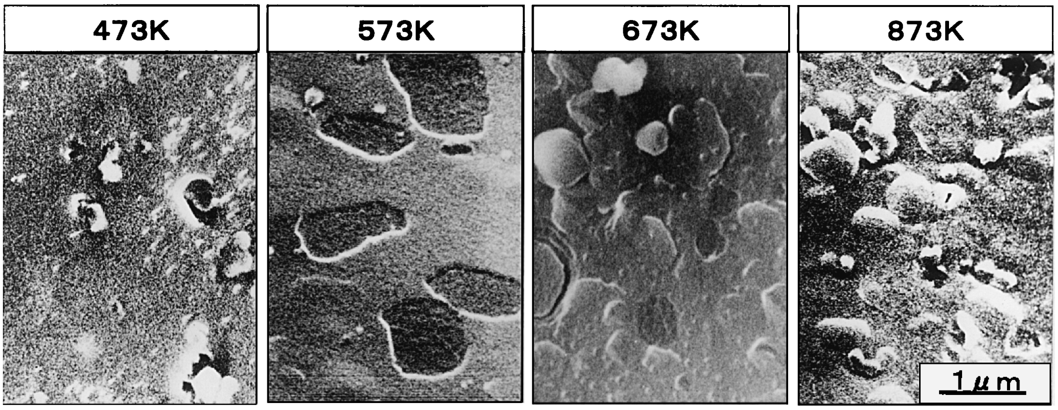

Fig. 2. SEM micrographs of beryllium samples irradiated at the fluence in the order of 10^{22} He^+/m^2 at various temperatures.

3.3. TDS measurement

Fig. 3 indicates thermal desorption spectra (TDS) of helium atoms released from beryllium samples irradiated at room temperature at various fluences. Note that the ordinates in the figure are different depending on the fluence.

The TDS spectra obtained in the present study greatly depend on the irradiation fluence. At the fluence in the order of 10^{19} He^+/m^2, almost all the helium atoms were thermally released between 1000 and 1200 K, where annihilation of the bubble and dissociation of vacancy–helium complexes could take place in beryllium. The spectrum in this fluence regime is very consistent with the data obtained by Eleveld et al. [13]. In the order of more than 10^{20} He^+/m^2, a new TDS peak develops between 600 and 800 K. This temperature regime is well consistent with the temperature where the vacancy-assisted migration of a vacancy–helium complex can take place [11]. It also corresponds to the TEM observation where the growth of bubbles occurred. The bubble growth might also be due to the helium–vacancy complex migration. Above about 10^{21} He^+/m^2 new peaks were observed to appear at around 400 K, where most of helium atoms was released.

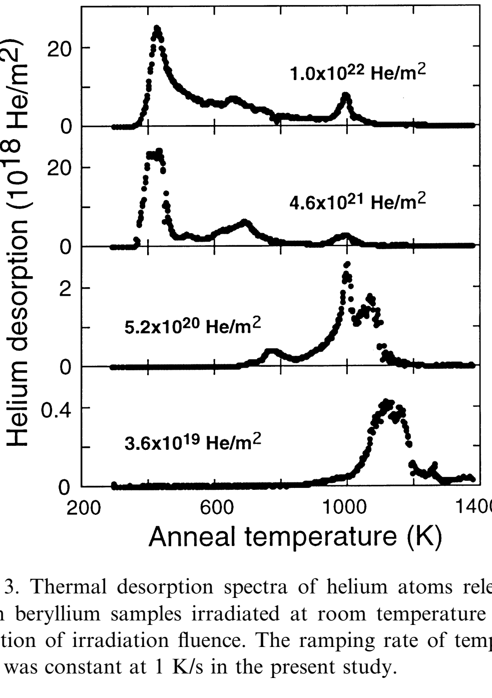

Fig. 3. Thermal desorption spectra of helium atoms released from beryllium samples irradiated at room temperature as a function of irradiation fluence. The ramping rate of temperature was constant at 1 K/s in the present study.

Fig. 4 is a plot of the amount of helium atoms retained in beryllium which were irradiated at room temperature, as a function of irradiation fluence. This plot is obtained from the TDS spectra of Fig. 3. As Fig. 4 shows the amount of helium atoms retained in beryllium deviated from the amount of implanted helium atoms at

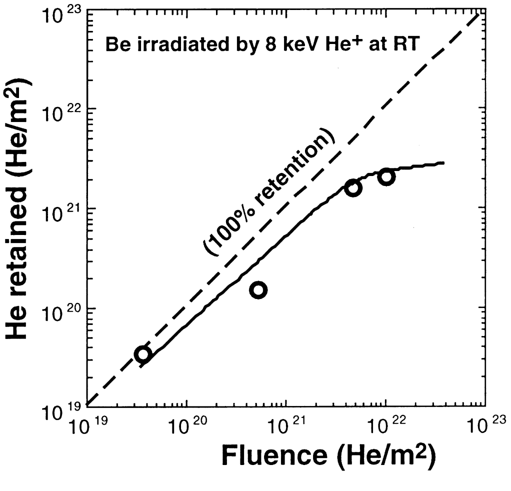

Fig. 4. Retention behavior of helium gas in beryllium irradiated by 8 keV helium ions at room temperature.

above 10^{21} He^+/m^2, indicating that capability of retaining helium atoms was exhausted. The behavior of helium atom retention in beryllium was well consistent with the findings of Pontau et al. [14].

4. Discussion

The microstructural development in beryllium observed by TEM was mainly due to bubbles and cavities in the present study. An interstitial-type defect, e.g., an interstitial loop, was scarcely observed at all the temperatures investigated here. The deuterium ion irradiation also hardly produces an interstitial loop [6]. A high voltage electron microscopy (HVEM) study with 1 MeV electrons indicated that no interstitial loops were produced in beryllium irradiated up to 0.1 dpa [15]. These facts may show that the nucleation of interstitial loops is very difficult in beryllium.

Dense tiny bubbles were produced in beryllium by 8 keV helium irradiation at any irradiation temperatures. The production of the tiny bubbles was an athermal process [12,16], which was based on a gas-driven process and radiation-induced vacancy migration. However, the threshold fluence for the tiny bubble formation is slightly depended on temperature. Thermal vacancy migration would assist the process at higher temperature. Helium atoms implanted in beryllium could be held in the bubbles. From the TEM and TDS experiments, apparent number density of bubbles and the number of helium atoms retained in beryllium indicated that several thousands of helium atoms were captured in a single tiny bubble.

At the fluence in the order of more than 10^{21} He^+/m^2, the tiny bubbles were observed to connect with each other at lower temperature regime. This shows that interfracture of the bubbles occurred probably due to the high pressure of helium gas in the bubble. The interconnection of the bubbles indicates the formation of microchannels [11] which may be a path for helium gas to move around in the metal and to escape to the outside. Another significant effect of helium-gas pressure was indicated by the shape change of the irradiated surface. At fluences where the microchannels were developed, formation of the blisters, exfoliation, flakes and pinholes was observed. They all can be gas-driven phenomena. In this way, when the capacity for retaining helium in beryllium was almost exhausted as shown in Fig. 4, such helium-gas driven phenomena were dominant as the microchannel development and the surface shape change. In this higher fluence regime, most of helium-gas was released at relatively low temperature around 400 K in the TDS measurement. The helium release at the temperature could correspond to escaping of helium atoms through the microchannel to the surface, the exfoliation of blister and the pinholes on the blister. The behavior of helium gas may depend very much on the development of the microstructure in beryllium and its surface shape.

Significant effect of the irradiation temperature on the microstructural evolution was the growth of the bubble with an amoeba-like structure up to about 100–200 nm in diameter. The growth could be a result of the vacancy-assisted diffusion of helium–vacancy complexes [11]. It is interesting to note that in situ observation showed an easy movement, coalescence or shrinkage of the amoeba-like bubbles. This behavior of the amoeba-like bubbles must be gas-driven: The internal pressure in the bubbles and surface migration of beryllium atoms in the bubbles could play important roles on the phenomena. The number density of the amoeba-like bubbles of 100–200 nm in diameter that were produced at the temperature between 573 and 873 K, was about 10^{14} m^{-2}, which was 5–10 times greater than that of the blisters of such sizes observed by SEM. The amoeba-like bubbles produced near the surface could be related to the blisters observed by SEM. In the irradiation temperature of more than 773 K, the bubbles grew to create a hexagonal void-like structure, i.e., cavity. The transformation from the roundish bubble into the hexagonal cavity could be related to reduction in gas pressure in the bubble. At this temperature regime, the radiation-induced or thermal vacancies could be easily supplied for the bubble growth, resulting in an increase in size and reduction in pressure. Moreover, escaping of helium gas from the sample could also be related to reduction in pressure. Microstructural evolution due to the large bubbles was also dependent on behavior of helium gas atoms.

5. Summary

The changes in the microstructure and its surface shape were investigated for beryllium samples irradiated by 8 keV helium ions at several temperatures between room temperature and 873 K. Helium release behavior during post-irradiation annealing was also investigated from beryllium samples irradiated at room temperature. The amount of helium atoms retained in beryllium at room temperature was saturated in the fluence of more than 10^{21} He^+/m^2, where the interbubbule fracture and blister formation were observed. In this higher fluence regime, most of retained helium was released during annealing at very low temperature around 400 K, where a divertor material of the ITER can be experienced. It indicates that helium re-emission from beryllium can be in problem during operation of the ITER.

The microstructural and surface evolution in beryllium can be all helium-gas-driven, and transport of the helium gas can be greatly dependent on the changes in the microstructure and surface shape. All the behaviors greatly depend on temperature.

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