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Effects of cascade damages on the dynamical behavior of helium bubbles in Cu

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Abstract

Effects of cascade damage on the behavior of helium bubbles in copper have been studied. To examine the dynamical response of the helium bubbles to irradiation with high energy self ions, in situ TEM observations during irradiation with 400 keV Cu^+ at room temperature and 830 K were carried out. Intermittent motion of bubbles of a few nm was observed at room temperature where the bubble was thermally immobile. At high temperature, in addition to Brownian type motions, rapid motion of rather long distances of about 10 nm was observed. The detailed analysis of the frequency and distance of these motions suggested the direct interaction of helium bubbles with sub-cascades and secondary defects.

1. Introduction

The understanding of effects of cascade damage, formed by high energy neutrons, on fusion devices is one of the most critical issues for future nuclear fusion reactors. The in situ observations by transmission electron microscopes (TEM) during high energy ion irradiation have been employed as an efficient technique to investigate these effects [1–3]. In addition, the neutrons generate insoluble helium due to nuclear transmutations. This helium can make the reactor materials brittle. However, since there are few reports on the behavior of helium bubbles during high energy ion irradiation [4,5], the mechanism of the cascade effects on individual bubble behavior for wide temperature ranges has not been clarified yet. The present study is aimed at obtaining fundamental knowledge about the interaction of helium bubbles with cascade damage during the high energy self ion irradiations by means of in situ TEM.

2. Experimental procedure

The material used for this study is polycrystalline Cu of 99.9999 at.% purity supplied by Daido-Kogyo Co. Cu disks of 3 mm diameter were annealed at 873 K for 2 h in a vacuum of 1×10^{-6} Pa and then polished electrochemically for the electron microscopy.

He bubbles were introduced into these specimens at around 573 K by irradiation with 10 keV He^+

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ions to a fluence of 5×10^{19} He^+/m^2 in a JEM-2010 TEM equipped with a low energy ion gun. To examine the dynamical behavior of He bubbles during the formation of cascade damage, in situ observation was carried out with high energy self ions. The 400 keV Cu^+ irradiations were performed in the range room temperature to 830 K, using the intermediate voltage electron microscope (IVEM) facility at Argonne National Laboratory. The beam flux was about 3.6×10^{15} Cu^+/m^2 s. The dynamical behavior of the bubbles was recorded continuously with the video recording system with a time resolution of 1/30 s.

In cases where bubbles are mobile, the relationship between the mean square of the migration distance, $\langle R^2 \rangle$, and time, t, was analyzed. If a proportionality relation was observed, the diffusion coefficient, D, was estimated from the following equation for a series of 2-D video frames according to the random walk theory.

$$\langle R^2 \rangle = 4Dt. \tag{1}$$

Details of the method and theories were shown elsewhere [5,6]. In identifying the positions of bubbles, the center position of each bubble was measured from the video images with an observational error of less than 0.6 nm.

3. Results and discussion

3.1. Depth distribution of he bubbles and cascade damage

To obtain information on the depth distribution of cascade damage and bubbles, Monte Carlo calculations were carried out with TRIM-code [7]. Fig. 1 shows a calculated depth distribution of He bubbles produced by 10 keV He^+ ions in Cu, which was described as a product of the projected ion range and damage deposition. Most bubbles should be located in the 60 nm range from 10 to 70 nm in depth.

In addition, the size distribution of sub-cascades produced by 400 keV Cu^+ ions was also plotted in Fig. 1 as a function of the depth. The distribution of sub-cascades was approximately comparable with the He bubble distribution. In this figure, only sub-cascades consisting of more than 100 vacancies produced by a knock-on atom were plotted because the small sub-cascades are not expected to have tangible impact on the bubble motion. The size was estimated as the statistical standard deviation of the calculated distribution of vacancies contained within a sub-cascade, assuming a sub-cascade was a sphere. The average diameter and number of sub-cascades produced in the region overlapping the He bubbles distribution range 10–70 nm in the depth, were evaluated to be 3.2 nm and 8.2, respectively.

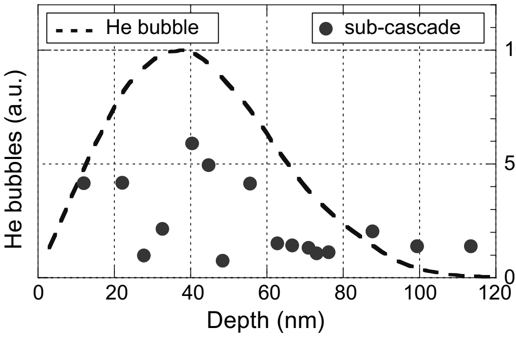

Fig. 1. Depth distributions of He bubbles produced by 10 keV He^+ ions and sub-cascades produced by 400 keV Cu^+ ions in Cu, which were calculated by the TRIM-code [7].

3.2. Room temperature irradiation

Under high energy self ion irradiations at room temperature where the bubbles are thermally immobile, intermittent bubble motions of a few nm were observed. Fig. 2 shows a couple of images obtained from an area of helium-implanted Cu during 10 s of irradiation at 300 K. The bubble indicated by the arrow moved a distance evaluated to be 2 nm. The velocity of the bubble was measured as a function of time during the irradiation from the series of video frames obtained every 1/3 s, and the result is shown in Fig. 3. As shown in this figure, sporadic motion of the bubble occurred at around 6 and 9 s in time. This kind of motion with a moving distance of several nm or less took place in random direc-

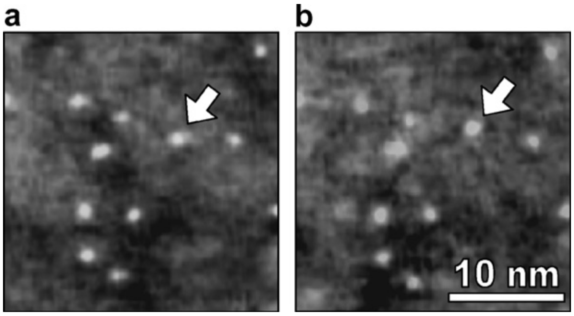

Fig. 2. He bubble motion in Cu during room temperature irradiation with 400 keV Cu^+ (b) was taken 10 s later than (a).

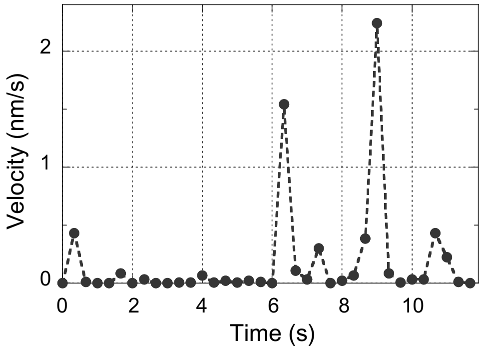

Fig. 3. Velocity of the bubble motion under room temperature irradiation with 400 keV Cu$^+$.

tions under the irradiation. The changes in the position of bubbles are plotted for the 10 s irradiation in Fig. 4. In this figure, some bubbles were neglected, because the bubbles images disappeared occasionally due to a change of diffraction conditions or bending of the specimen. Counting the bubbles which moved more than 1 nm, and considering an observational error, 21% of bubbles moved on average during a 10 s irradiation.

According to Ref. [4], irradiation induced bubble motion can be described as an interaction process between bubbles and cascade zones. The probability of the interaction is the product of the sub-cascade production rate and the interaction zone volume surrounding the bubble. The interaction rate, N, is given by

$$N = \frac{4\pi}{3}\left[(r_b + r_c)^3 - r_b^3\right] n\phi/t, \tag{2}$$

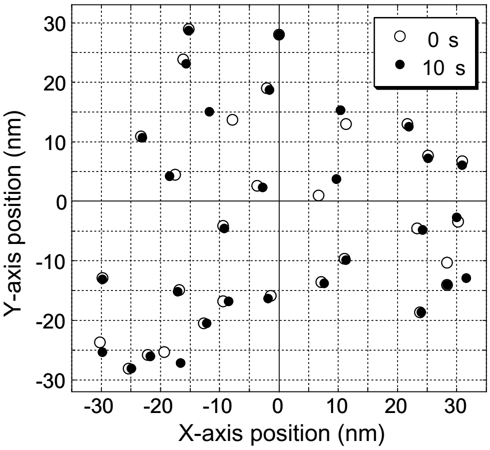

Fig. 4. Changes of the positions of He bubbles for a 10 s irradiation with 400 keV Cu$^+$ at room temperature.

where r_b, r_c are the mean radius of the bubble and the sub-cascade respectively, n is the number of sub-cascades per ion, ϕ is the ion flux, and t is the thickness of the interaction zone. The interaction rate of 3.4%/s is obtained from this equation, using $r_c = 1.6$ nm, $n = 8.2$, $\phi = 3.6 \times 10^{15}$ ions/m^2 s and $t = 60$ nm as mentioned above, and $r_b = 1$ nm obtained from the micrograph. As a result, the fraction of the number of moving bubbles is calculated to be 29% for a 10 s irradiation. On the other hand, 21% were obtained as the experimental value. This was slightly smaller than the value given by Eq. (2). However, considering the distance of all bubble motions were less than the sub-cascade size and the short range motions less than 1 nm were neglected, this direct interaction between bubbles and sub-cascades seems to provide a good explanation of the bubble motion under high energy ion irradiation at room temperature. These results also suggest a useful method to evaluate cascade effects with He bubbles. This method makes it possible to observe single events of a cascade directly under the microscope.

3.3. High temperature irradiation

It has been reported that smaller He bubbles in Cu move with Brownian motion at temperatures above about 773 K under normal conditions, whereas He bubbles showed erratic motion under irradiation at high temperatures. Fig. 5 shows a series of video frames of bubbles in Cu specimen during 400 keV Cu$^+$ irradiation at 823 K. The bubble indicated by the arrow in the figure moved rapidly through a distance of more than 10 nm. This kind of rapid motion was observed regardless of bubble sizes. Fig. 6 shows the trajectory of the same He bubble during beam off (a) and beam on (b) periods. The position of the bubble was plotted every 0.5 s for 30 s. From the comparison of the motions

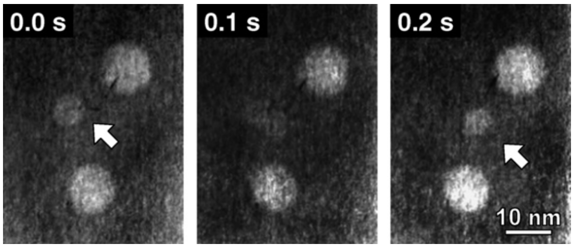

Fig. 5. Rapid motion of a bubble under irradiation with 400 keV Cu$^+$ at 823 K.

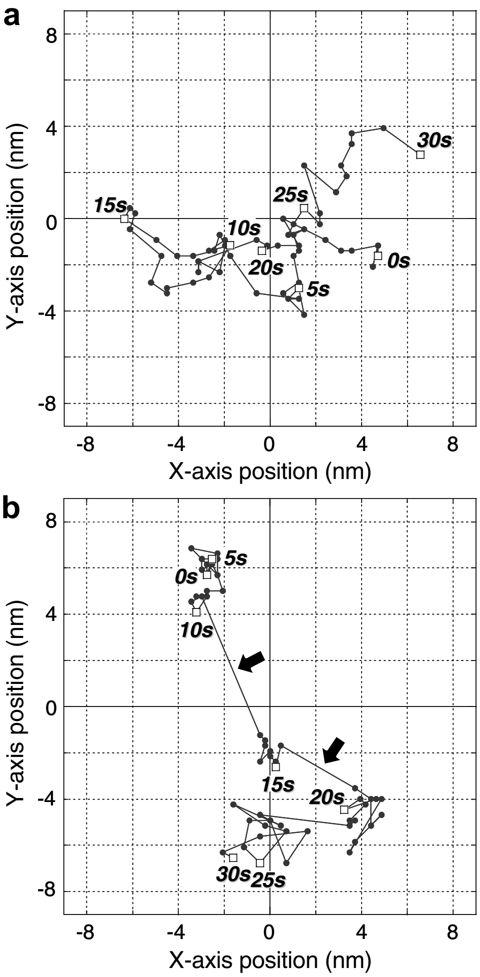

Fig. 6. The trajectories of the same bubble during beam off (a) and beam on periods (b). The positions of the bubble were plotted every 0.5 s for 30 s.

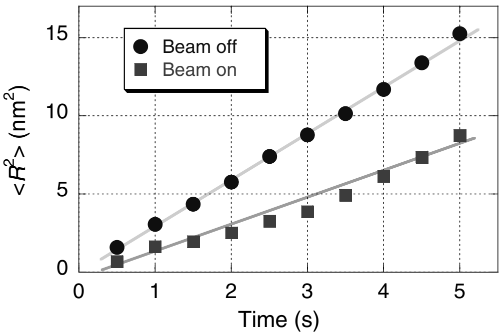

Fig. 7. Mean square of the same bubble migration distances, $\langle R^2\rangle$, versus time. The data for the bubble during the beam off period was obtained through a process of exclusion of the influence of the rapid rectilinear motion.

shown in this figure, whereas the bubble merely has a random motion during the beam off period, the bubble under the irradiation clearly has rapid long distance rectilinear motions in addition to random motion within a relatively short region. To compare the behavior of these bubbles quantitatively, the mean squares of the migration distances, $\langle R_x^2\rangle$, were analyzed as a function of time. In Fig. 7, data obtained from the same 10 nm diameter bubble during the beam off and the beam on period were plotted, where the data for the bubble during the beam off period was obtained through a process of exclusion of the influence of the rapid rectilinear motion, assuming that the motion under irradiation consists of two components, the random motion and the rapid rectilinear motion. It is clear that these mean square distances are proportional to time, which indicates Brownian type motion. From the slopes in the figure, $D_{\text{Beam-off}} = 7.5 \times 10^{-19}\ \text{m}^2/\text{s}$ and $D_{\text{Beam-on}} = 4.3 \times 10^{-19}\ \text{m}^2/\text{s}$ were found as diffusivity coefficients by use of Eq. (1). The component of Brownian motion of the bubbles under irradiation is suppressed in comparison with the motion during beam off period. This retardation during the irradiation has been confirmed in previous work on Al [5], and may be caused by absorption of excess vacancies that survive through kinetic reactions among point defects in the cascade damage region.

In spite of this retardation, the mobility of bubbles under irradiation increased remarkably due to the long distance rapid motions. These rapid motions occurred at an average rate of 0.048 jumps/s per bubble. Assuming a simple direct interaction between bubbles and sub-cascades and adapting the Eq. (2) with mean bubbles radius $r_b = 6.0$ nm to this case, the frequency rate is estimated to be 0.46 jumps/s. This value differs by about one order magnitude from the experimental value, and the distance of the rapid motion is much longer than the sub-cascades radius. These mean that the simple interaction model cannot account for the erratic motion of bubbles under high temperature irradiation. A possible mechanism is the interaction with secondary defects, such as dislocation loops. Taking into account the increase of bubble mobility along edge dislocations and grain boundaries [5,8], the motion could be induced by the dislocation loops which formed and annihilated during the irradiation. Also, since the coinstantaneous formation of several defect clusters within a narrow range of 30 nm and less has been reported under similar irradiation condition [3], a relation with the rapid motions with the distance of about 10 nm was expected.

As discussed previously, the cascade damage has significant effect on the dynamical behavior of bubble motion. Similar effects should be expected to be caused by the high energy neutron irradiation of plasma facing materials in fusion reactors. Hence, the cascade damage is likely to cause serious problems related to not only deterioration of materials but also control of the plasma, connected to the retention and release of fusion plasma particles, including He, D and T species.

4. Summary

The dynamical response of helium bubbles to irradiation with high energy self ions in Cu has been studied by means of the in situ TEM observation. It was shown that cascade damage formed by irradiation has significant effects on the dynamical behavior of bubble motion. At room temperature where the bubble was thermally immobile, the direct interaction between bubbles and sub-cascades caused the intermittent bubble motion of a few nm. In the high temperature irradiation, the retardation of Brownian type motion and rapid rectilinear motions were observed.

These changes of dynamical behaviour of He bubbles seem to have impacts on the retention and release properties of He atoms in the material. Taking into account the multiple radiation effects in plasma facing materials, the retention properties under cascade damage should be examined in the future.

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