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Formation and migration of helium bubbles in Fe–16Cr–17Ni austenitic alloy at high temperature

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Abstract

The formation and migration of helium bubbles in Fe–16Cr–17Ni austenitic model alloy at temperatures 400–1250°C have been studied by in situ electron microscopy, energy dispersive spectrometry (EDS) and EELS analysis. The nucleation of bubbles under 10 keV He^+ ion irradiation was dominant at 400°C, while the growth was dominant above 600°C. It was revealed that the mean square of the migration distance was proportional to time, which is quantitative evidence of Brownian type motion and yielded diffusivities from 10^{-18} to 10^{-20} m^2/s at 1185°C, depending on the diameter from 1 to 3 nm. These facts suggest that the mobility of helium–vacancy complexes or bubbles is an important factor governing the formation process of helium bubbles. The analysis by scanning transmission electron microscope-electron energy loss spectrometry (STEM-EELS) elucidated Ni precipitation even around small helium bubbles.

1. Introduction

Void swelling behavior in austenitic alloys has been widely studied for the development of fusion reactor materials [1]. The formation and migration of gas bubbles are fundamental processes of the void swelling and understanding of these behavior is basically important and also interesting from an aspect of bubble physics. However, a little information on bubble behavior has been obtained in austenitic alloys. Schroeder et al. [2] studied coalescing mechanisms of helium bubbles in stainless steel, however, direct information on bubble behavior is poor. Therefore, in the present work, we aimed to obtain quantitative data on the formation and migration of helium bubbles in Fe–16Cr–17Ni austenitic model alloy irradiated with such low energy helium ions as in a fusion plasma [3], utilizing in situ observation technique to obtain direct and dynamical information [4,5].

2. Experimental procedures

Disk shaped specimens of Fe–16Cr–17Ni austenitic alloy, which was made from high purity starting materials, were pre-annealed at 1070°C for 1 h in an ultra high vacuum furnace and then polished electrochemically for transmission electron microscopy (TEM). The specimen was mounted on a TEM specimen holder for high temperatures and irradiated with 10 keV He^+ ions, using a low energy ion accelerator connected to a JEOL-2010 electron microscope. In situ TEM observation of the bubble formation was made during ion irradiation with a constant flux of 5.0×10^{17} ions/m^2 s at temperatures from 400°C to 800°C, and the bubble density and the depth distribution were measured by stereo microscopy. Some specimens irradiated with He^+ ions were annealed stepwise from 700°C to 1230°C and the motion of the bubbles was monitored continuously and recorded on a video tape recorder (VTR). The path of a bubble was followed by using an image processor, which allows systematic measurements of center position of bubbles as a function of time.

Precipitation or segregation of alloy elements in irradiated or un-irradiated specimens was examined by

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X-ray energy dispersive spectrometry (EDS) or scanning transmission electron microscope-electron energy loss spectrometry (STEM-EELS), using a Gatan-666 (p-EELS) with 200 kV electrons from a JEOL–Schottky type field emission gun at room temperature. The scanning was made by 0.57 nm space.

3. Results and discussion

3.1. Formation of helium bubbles

Before irradiation, we checked precipitates formed during annealing at temperatures from 400°C to 800°C and confirmed that Cr and Cr oxide were formed at the surface of thin foil specimen.

Areal number densities of helium bubbles formed by irradiation with 10 keV He^+ ions at 400°C, 600°C and 800°C are shown in Fig. 1, as a function of the fluence. The bubble density proportionally increased with fluence without growth at 400°C, while at higher temperatures the bubble growth is remarkable and the density is depressed. That is, the bubble nucleation process is dominant at 400°C, while the growth process is dominant above 600°C. Depth distributions of these bubbles are shown in Fig. 2. The peak depths at 400°C

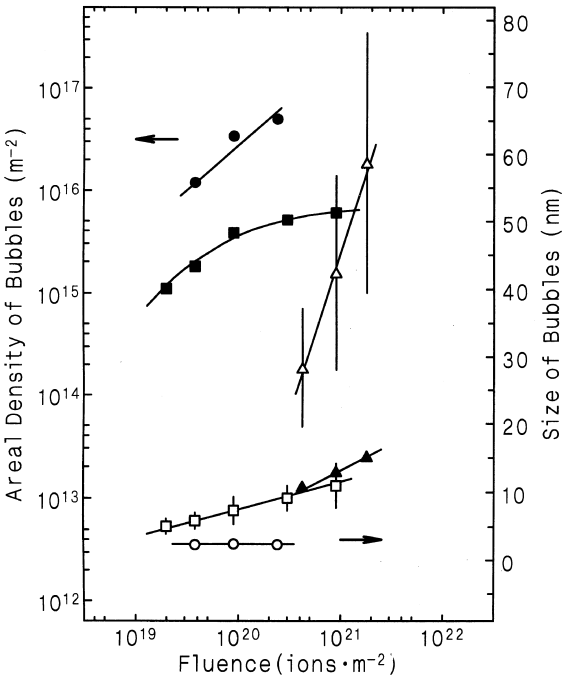


Fig. 1. Areal number density of helium bubbles (filled markers) and their average diameter (open markers) in the specimen irradiated at 400°C (●,○), 600°C (■,□) and 800°C (▲,△) versus the fluence of 10 keV He^+ ions.

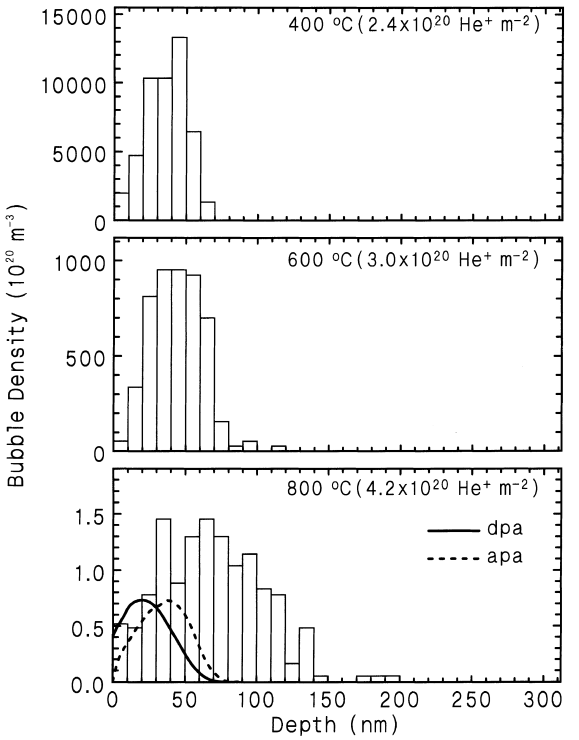


Fig. 2. Bubble density versus the depth from the specimen surface. In the lowest part of the figure, dpa- and apa-rate depositions calculated by the TRIM code are shown for comparison.

are in good agreement with the maximum apa-rate deposition calculated by TRIM code [6], while bubbles formed at 600°C and 800°C distributed even in deeper region.

3.2. Annealing and migration of helium bubbles

After irradiation with helium ions, the specimen was isochronally annealed for 20 min at higher temperatures and changes in the bubble density and the size were examined. It was found that the number density of bubbles remarkably decreased above 900°C and at the same temperature smaller bubbles (diameter $r \sim 1$ nm) disappeared and larger bubbles ($r \sim 6$–12 nm) appeared. This suggests that the migration of smaller bubbles ($r \sim 1$ nm) and their coalescence to form larger ones become significant above 900°C. The disappearance of the smaller bubbles at the specimen surface is also likely during the motion. No shrinkage of bubbles was observed, which was suggested from Ostwald ripening mechanism [2].

To measure the mobility of helium bubbles, video frames of moving bubble were analyzed. Some representative TV frames taken from a series obtained on a

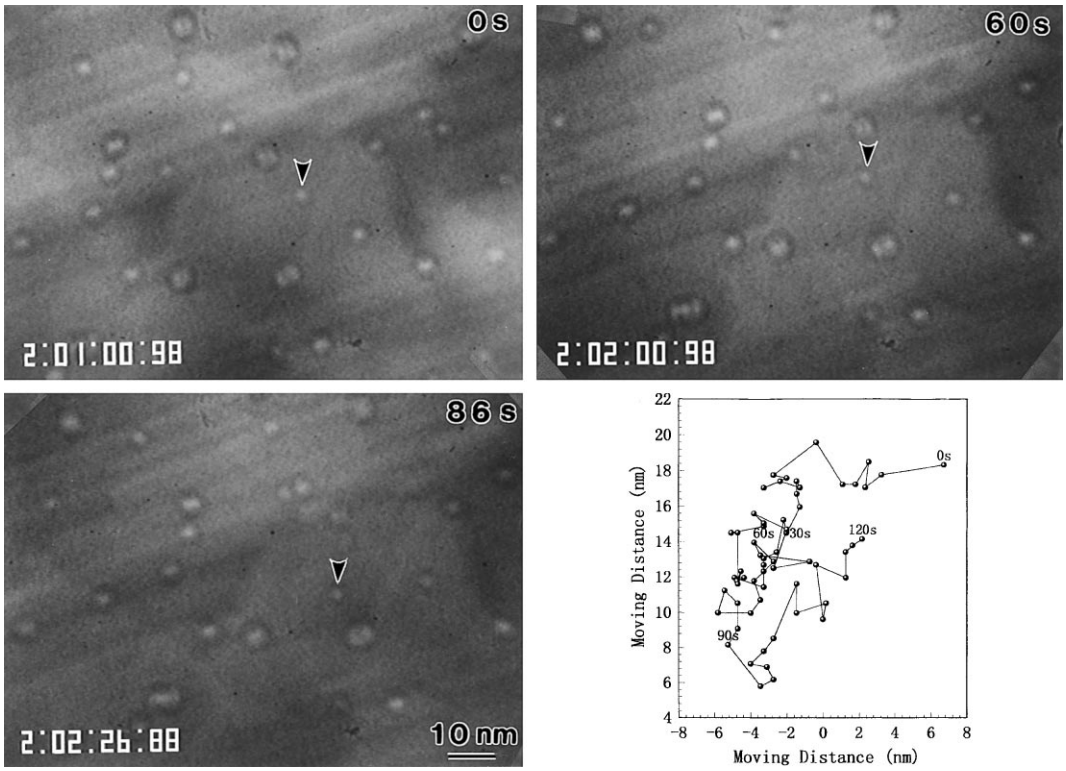


Fig. 3. A sequence of TV frames, which show the bubble motion at 1185°C and a path of the bubble marked with the arrow, being measured at 2 s intervals.

specimen irradiated to a dose of 7.5×10^{18} ions/m^2 at 1185°C are shown in Fig. 3. The path of the bubble marked with the arrow is also shown in the figure, where the center positions were measured for 2 min at 2 s intervals. The motion in random directions is clear. To quantify the bubble mobility, the mean square of the bubble migration distance, $\langle R^2 \rangle$, was plotted in Fig. 4, as a function of the time, t. It is evident that $\langle R^2 \rangle$ s for the same bubble are proportional to time for both temperatures of 1185°C and 1215°C. The similar relation was found for various sizes of bubbles. This is quantitative evidence of Brownian type motion of the bubbles and is the first demonstration in the austenitic alloy. During Brownian type motion, some bubbles coalesced and some disappeared at the specimen surface.

The random walk theory allows us to evaluate the diffusivity of the bubble, D, from the relation $D = \langle R^2 \rangle / 4t$. Diffusivities thus obtained are strongly dependent on the bubble diameter, as shown in Fig. 5, where D values at 1185°C vary in the order from 10^{-18} to 10^{-20} m^2/s, depending on the bubble diameter from 1 to 3 nm. The gradient of the solid line seems to favor a r^{-4} relation proposed by a surface diffusion model [7] rather than a r^{-3} relation proposed by a volume diffusion model [8]. Larger bubbles ($r > 5$ nm) frequently had

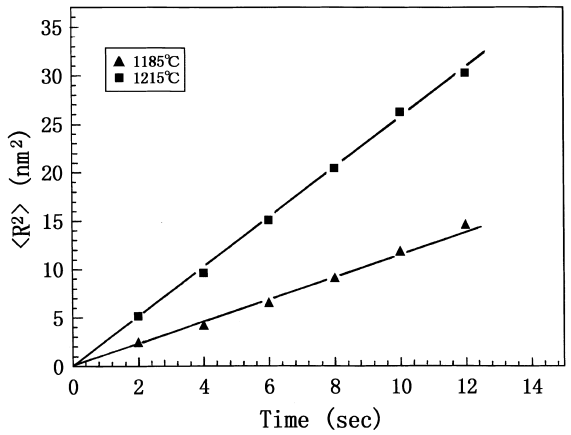


Fig. 4. Mean square of the migration distance of bubble with $r = 1.8$ nm versus time. It is noted that $\langle R^2 \rangle$ is proportional to time for both temperatures.

faceted faces and their diffusivities became lower than $\sim 10^{-21}$ m^2/s at 1185°C.

The diffusivities shown in Fig. 5 suggest that very small bubbles move rapidly. According to the diffusion coefficient of helium–vacancy complex in AISI 316 steel

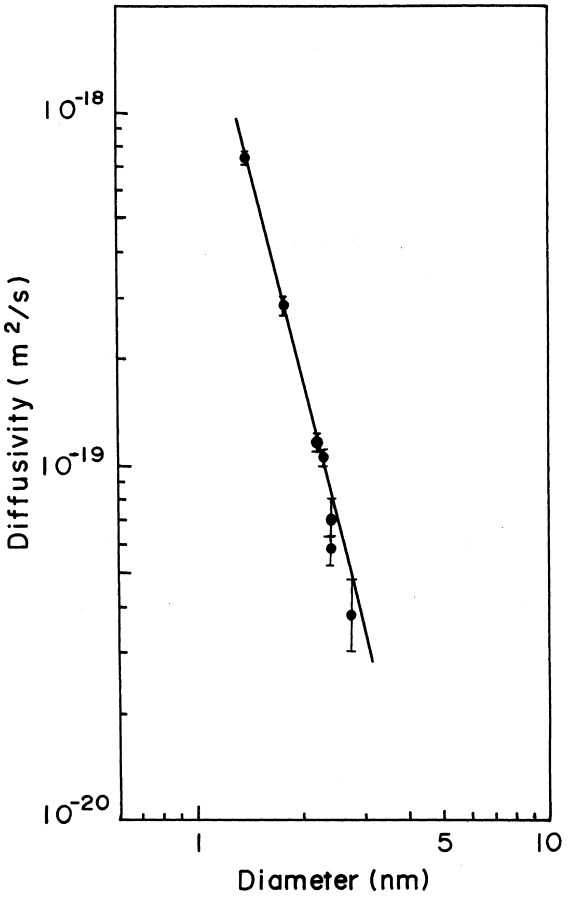
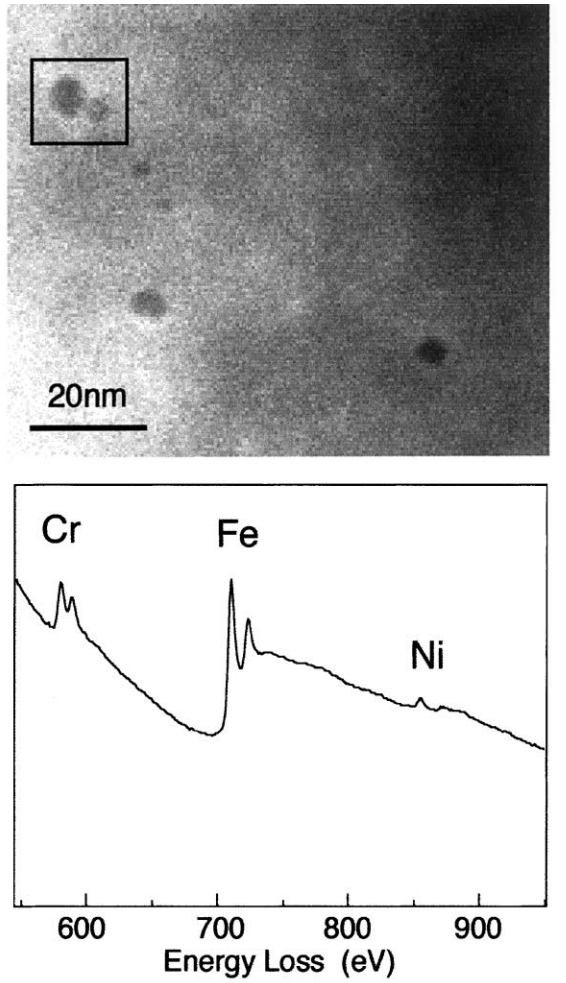


Fig. 5. Diffusivity of bubble at 1185°C as a function of the bubble diameter.

$D_{\mathrm{He\text{-}V}} = 5 \times 10^{-6} \exp(-2.1\,\mathrm{eV/kT})$ $\mathrm{m^2/s}$ reported by Sekiguchi et al. [9] and present results shown in Figs. 1 and 2, we can deduce that helium–vacancy complexes are less mobile at 400°C, where nucleation occurs without significant bubble growth. The clusters become mobile above 600°C, where bubble growth occurs even in regions deeper than the helium deposition. Thus, the mobility of helium–vacancy complexes or bubbles is an important factor, which governs the formation process of helium bubbles at high temperature.

3.3. STEM-EELS analysis

In order to examine segregation of the alloy elements on the bubble surface and its effects on the motion, an analysis by STEM-EELS was carried. Fig. 6 shows an annular dark field image (ADF image) of bubbles in a specimen irradiated to 1.8×10^{20} $\mathrm{He^+/m^2}$ at 700°C, energy loss spectra from L_{23} edges and the magnified ADF image and elemental maps for Fe, Cr and Ni around the bubbles in the marked region (20.66×17.79 nm) of the



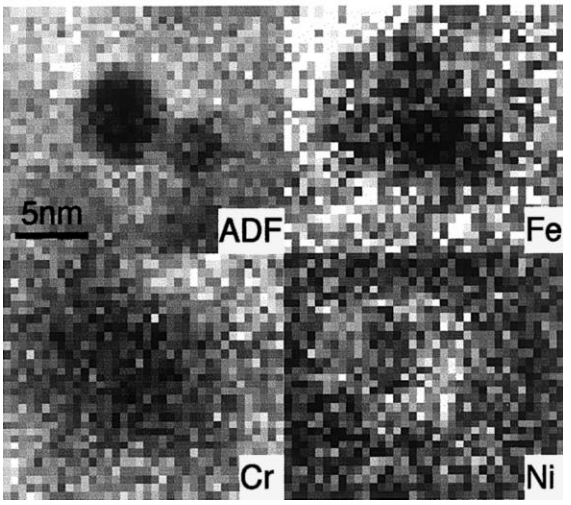


Fig. 6. ADF image of bubbles formed at 700°C (top), electron energy loss spectra from L_{23} edges (middle), magnified ADF image and elemental maps around the bubbles in the enclosed region of the ADF image with mark (bottom).

ADF image. It is clear from the elemental maps that Cr and Fe were depleted, but Ni was enriched even around the small bubble. Similar enrichment of Ni around very large size voids in neutron irradiated Fe–Ni–Cr alloys was previously observed by EDS [10]. The Ni coating of bubble surface is considered to affect the bubble motion, because reported diffusivities of alloy elements in Fe–Ni–Cr alloys were different in the order $D_{Cr} > D_{Fe} > D_{Ni}$ with $D_{Cr}/D_{Ni} \cong 2.5$ [11].

4. Conclusions

We have examined the formation and migration of helium bubbles in Fe–16Cr–17Ni austenitic alloy by in situ TEM, EDS and EELS analyses and conclude the following:

1. The nucleation process of bubbles is dominant at 400°C, while the growth process is dominant above 600°C. The mobility of helium–vacancy complexes or bubbles is an important factor governing these processes.
2. Brownian type motion of helium bubbles has been demonstrated in the alloy by the observation that the mean square of the migration distance is proportional to time.
3. The diffusivity of the bubble has been derived as a function of the bubble diameter and the values range from 10^{-18} to 10^{-20} m^2/s at 1185°C.
4. A Ni coating on the bubble surface has been found, which may affect the bubble mobility.

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