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Dynamical interaction of helium bubbles with grain boundaries in Fe and Fe–9Cr ferritic alloy

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Abstract

Dynamical interaction of helium bubbles with grain boundaries in Fe and Fe–9Cr has been studied by in situ electron microscopy and thermal desorption spectroscopy (TDS). It is clearly demonstrated that most small helium bubbles are swept to the specimen surface by dynamical interaction with the moving grain boundaries during re-crystallization, which induces easy motion along the boundary. At small angle grain boundaries, helium bubbles are frequently trapped at grain boundary dislocations and move along the dislocation line. The bubble diffusivity along the grain boundary dislocation is statistically estimated from the moving path and the value is similar to or a little higher than that in the matrix. Well correlated and enhanced thermal desorption spectra with the sweeping of helium bubbles by moving grain boundaries in Fe and Fe–9Cr are revealed.

1. Introduction

Helium atoms introduced in fusion device materials by nuclear reactions or bombardment of energetic plasma ions frequently precipitate at grain boundaries and result in premature intergranular fracture [1,2]. The problem of helium-induced cracking at grain boundaries during repair welding of irradiated materials is also serious [3]. Therefore, knowledge of the development and dynamical interaction between helium atoms or helium bubbles and grain boundaries is essential for the development of reactor materials. However, only limited information about these subjects is available at present.

Recently, we have successfully revealed easy motion of small helium bubbles along general grain boundaries in Al, where the grain boundary was stationary, and estimated the bubble diffusivity [4,5]. In the present work, similar in situ TEM observation is performed in pure Fe and Fe–9Cr ferritic alloy and dynamical interaction of helium bubbles with moving grain boundaries during re-crystallization is examined. In addition, correlation of the desorption of helium with microstructure change in these specimens is examined by quadru-pole mass-spectroscopy. Also, retention and release property of helium from plasma faced

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components is a key issue for control of the fusion plasma and thus more information about these properties is required.

2. Experimental procedures

Specimens were 99.999 at.% purity Fe supplied by Showa Denko Co., and Fe–9Cr ferritic alloys, which were cast from high purity starting materials supplied by Johnson–Mattey Co. Disk shaped specimens of these materials were annealed at 1220 K for 1 h in a high vacuum furnace and then electrochemically polished for electron microscopy (TEM). The diameter of grain, d_g, in these well annealed specimens of both materials was about 80 μm. Some specimens were annealed at 770 K to produce smaller grains and the average diameter d_g was about 60 μm in Fe and less than 20 μm in Fe–9Cr. The Fe–9Cr specimens included complicated dislocation structure prior to the re-crystallization.

The specimen was irradiated with 10 keV He^+ ions at a temperature ranging from room temperature to 673 K, using a low energy ion accelerator connected to a JEM-2010 electron microscope. After the irradiation, the specimen was warmed up stepwise to 1270 K and the dynamical interaction of helium bubbles with grain boundaries was continuously monitored by in situ electron microscopy. The TEM image of the microstructure of the irradiated specimen was recorded on video tapes by a CCD camera system. The path of the bubble was followed by using an image processor, which allows systematic measurement of the center position of the bubbles as a function of time.

Thermal desorption spectroscopy (TDS) of helium during heating up of similarly irradiated specimens at a constant heating rate was measured by a quadru-pole mass-spectrometer and their correlation to the microstructure change was examined. The size of the thin plate specimen for TDS was $10 \times 10 \times 0.1$ mm.

3. Results and discussion

3.1. Sweeping of helium bubbles by moving grain boundaries

In order to study the dynamical interaction of helium bubbles with grain boundaries, helium bubbles were introduced by irradiation of 10 keV He^+ ions on the specimen which had been annealed near 770 K. The size and density of helium bubbles can be controlled by the irradiation temperature and the ion dose. The results have been presented elsewhere [6].

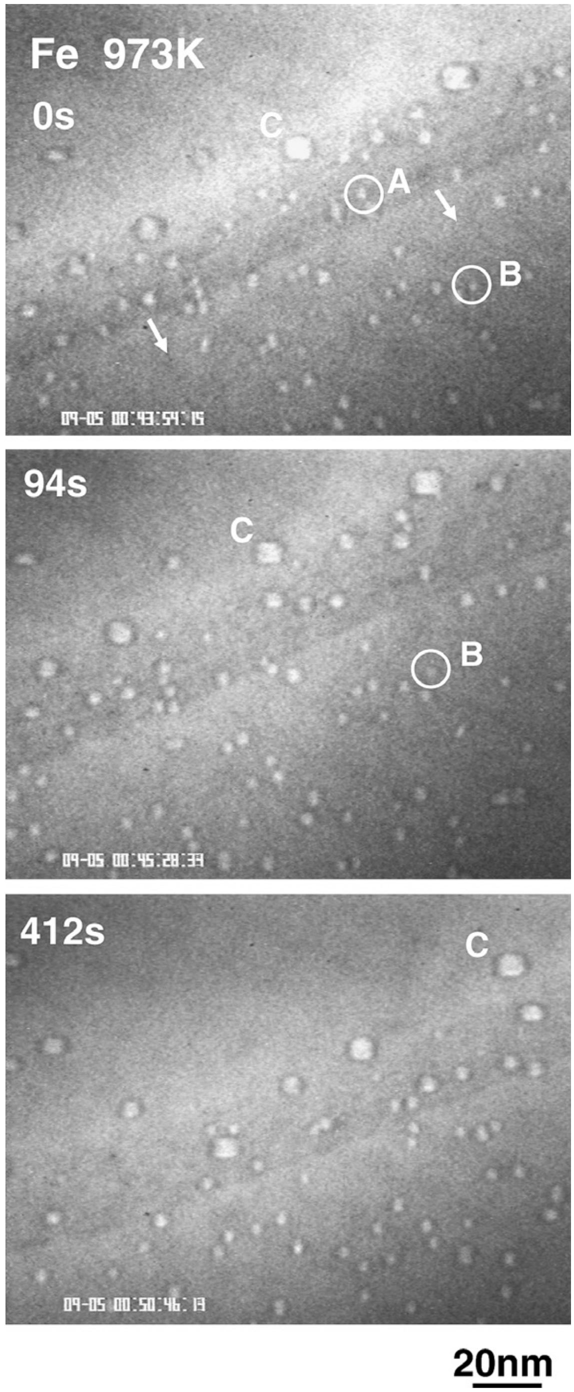

Fig. 1. A series of video frames which shows sweeping of helium bubbles during the motion of general grain boundary from upper left to lower right at 973 K in Fe.

When the irradiated specimen was warmed to higher temperatures, the grain boundary movement due to re-crystallization took place and characteristic interaction between moving grain boundaries and helium bubbles was observed. Fig. 1 shows a case of general grain boundary in pure Fe irradiated to a dose of 6×10^{19} ions/m^2 at 573 K and warmed stepwise to 973 K. The grain boundary was moving from upper left to lower right in the direction shown by the arrows. The interacting bubbles with the grain boundary moved easily along the moving boundary like the bubble A. Some bubbles coalesced and some disappeared at the specimen surface. As a result, most helium bubbles were swept out with the progress of the boundary movement, but some bubbles coarsened by coalescence and were left behind the boundary, like bubble C. Fig. 2 shows the path of the representative bubble A (the diameter: $d = 2.2$ nm), seen in Fig. 1. The bubble A moved following the moving grain boundary and then disappeared at the specimen surface at 110 s.

In contrast, the bubbles in the matrix, like B, moved randomly as shown in the previous work [6]. Fig. 3 shows the mean square of the migration distance versus time for bubble B with diameter 2.5 nm. According to the random walk theory, we can estimate the diffusion coefficient of the bubble, D, from the proportional relation between the mean square of the migration distance and time t by $D = \langle R^2 \rangle / 4t$, which yields $D = 4.0 \times 10^{-19}$ m^2/s at 973 K.

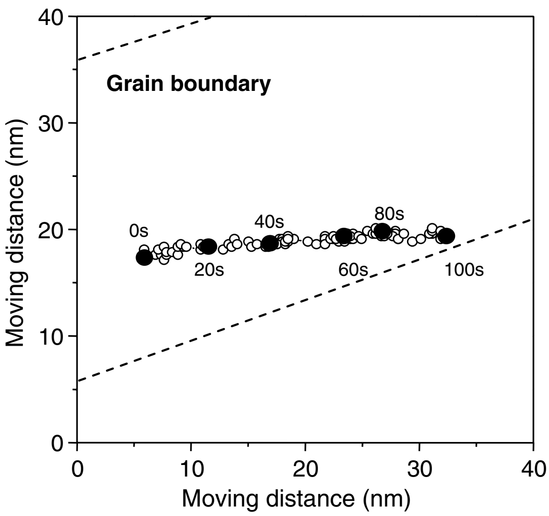

Fig. 2. The path of bubble A shown in Fig. 1, from pictures taken every 2 s for 100 s. The moving bubble followed the moving grain boundary and then disappeared at the specimen surface at 110 s.

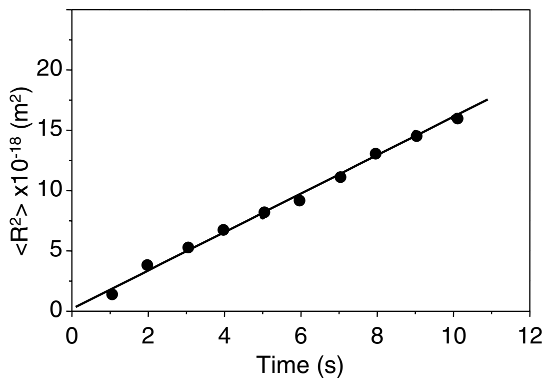

Fig. 3. Mean square of the migration distance for bubble B in the matrix, shown in Fig. 1, versus time. The proportional relation yields the diffusivity $D = 4.0 \times 10^{-19}$ m^2/s for the bubble B ($d = 2.5$ nm).

3.2. Migration of bubbles along grain boundary dislocations

A small angle grain boundary frequently includes boundary dislocations. At the boundary dislocations, trapping of helium bubbles during random motion along the grain boundary was frequently observed. They moved along the boundary dislocation and some coalesced and were stabilized. Fig. 4(a) shows a photograph of such a bubble ($d = 3.6$ nm) moving along the boundary dislocation at 973 K in Fe, where the boundary was stationary because of the long time annealing at the temperature. Fig. 4(b) shows the one dimensional path of the moving bubble indicated by the arrow in Fig. 4(a). To quantify the bubble motion along the grain boundary dislocation, the mean square of the migration distance $\langle R_{gd}^2 \rangle$ is plotted in Fig. 5 as a function of time t. From the proportional relation, the diffusivity of the bubble, D_{gd}, is estimated from the relation, $D_{gd} = \langle R_{gd}^2 \rangle / t$, to be 4.9×10^{-19} m^2/s. This is the first measurement of diffusivity along a grain boundary dislocation. Here, the value of D_{gd} was estimated from the projected TEM image of the boundary dislocation, but its correction is less than 15% because of a small angle tilting. It is, therefore, known that the diffusivity along the grain boundary dislocation is similar to or a little higher than that in the matrix, taking in to account the size dependence of the bubble diffusivity, reported as $D \propto d^{-4} \sim d^{-5}$ [7].

In Fe–9Cr ferritic alloy, the precipitation of helium bubbles at grain boundaries is similarly observed at higher temperatures, around 1000 K, but the profile is complicated by irregular precipi-

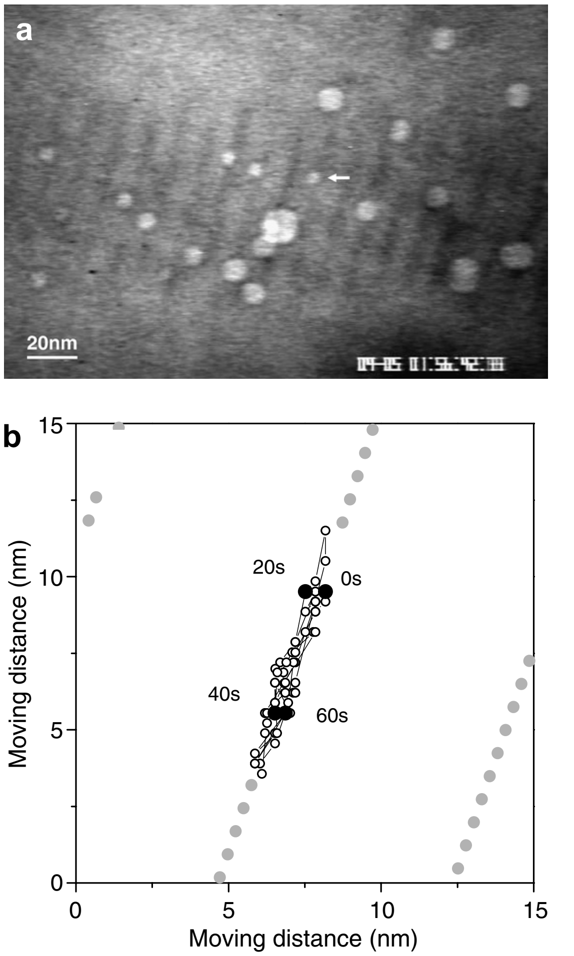

Fig. 4. (a) A photograph of the video frame which shows bubble motion along the grain boundary dislocations at 973 K in Fe. (b) The path of the bubble indicated by the arrow in the figure (a), from photos taken every 2 s. One dimensional motion along the dislocation is seen.

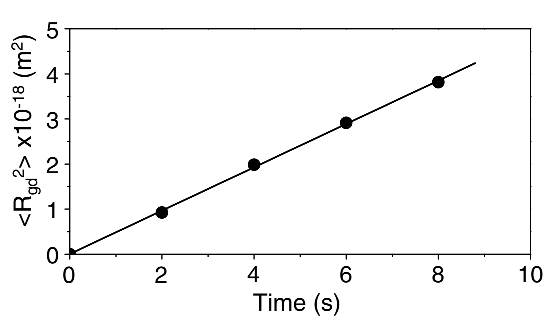

Fig. 5. Mean square of the bubble migration distance along the grain boundary dislocation, shown in Fig. 4(b), versus time. The diffusivity for the bubble ($d = 3.6$ nm) estimated from the inclination is $D_{gd} = 4.9 \times 10^{-19}$ m^2/s at 973 K.

tates of Cr on grain boundaries. For quantitative discussions, more study is necessary.

3.3. Thermal desorption spectroscopy

Thermal desorption spectra from well annealed specimen which was similarly irradiated by helium ions has been reported [8] and good correlation to the microstructure change during heating at a constant heating rate was demonstrated. In the present work, thermal desorption spectra from Fe ($d_g =$ 60 μm) and Fe–9Cr ($d_g < 20$ μm) specimens which were annealed at 770 K and irradiated to 2.0×10^{20} ions/m^2 at room temperature were measured. Fig. 6 shows their thermal desorption spectra during heating at a constant heating rate of 6 K/min, comparing with the one from Fe specimen ($d_g = 80$ μm) annealed at 1220 K and irradiated similarly. The peaks near 990 K can be correlated with the migration of small helium bubbles [8]. It is clearly seen that peaks above 1000 K in Fe ($d_g = 60$ μm) and Fe–9Cr ($d_g < 20$ μm) specimens annealed at 770 K are much higher than that of Fe ($d_g = 80$ μm) annealed at 1270 K. Total amount of helium atoms evolved up to 1270 K was 88, 78 and 22% of irradiated helium ions in Fe–9Cr ($d_g < 20$ μm), Fe ($d_g = 60$ μm) and Fe ($d_g = 80$ μm), respectively. This means that in pure Fe with larger size grain, more of the implanted helium still remains in the coarsened bubbles. These results suggest that high density of grain boundaries for a small grain size could trap high density bubbles and easily sweep out them to the specimen surface

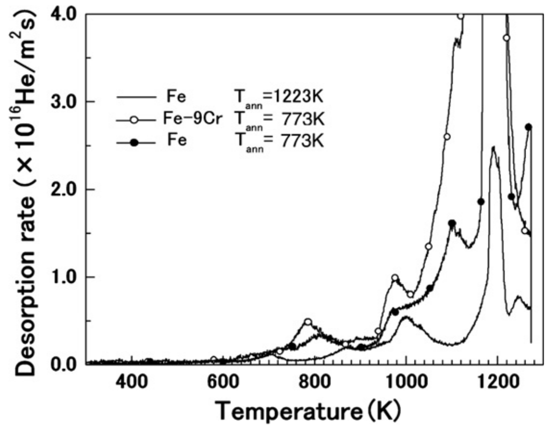

Fig. 6. Thermal desorption spectra of helium from Fe ($d_g = 60$ μm) and Fe–9Cr ($d_g < 20$ μm) annealed at 770 K and Fe ($d_g = 80$ μm) annealed at 1270 K, which were irradiated with 10 keV He^+ ions to a dose of 2.0×10^{20} ions/m^2 at room temperature and warmed up at a constant heating rate of 6 K/min. Enhancement of the helium evolution during re-crystallization above 1000 K is clearly seen.

through the easy motion along the moving grain boundaries during re-crystallization. These facts are well correlating to the microstructure change shown in Fig. 1.

The TDS peaks around 800–900 K can be correlated with release of helium trapped at interstitial type dislocations, because these loops disappear at these temperatures [8]. The sharp peaks at 1190 K and 1140 K were correlated with the α–γ phase transformation in Fe and Fe–9Cr, respectively [8].

The accumulation of helium bubbles by moving grain boundaries and migration and coalescence at grain boundaries, as described above, may be a general feature of the interaction between helium bubbles and grain boundaries, although their mobility depends on materials. This accumulation mechanism should play a significant role during repair welding of irradiated materials, which leads to helium-induced cracking at grain boundaries, instead of a free migration model of helium bubbles to grain boundaries [9].

4. Conclusions

The dynamical interaction of helium bubbles with grain boundaries in Fe and Fe–9Cr was examined by in situ TEM and TDS. The main conclusions of this work are as follows:

1. Sweeping of helium bubbles by moving grain boundaries during re-crystallization was demonstrated using continuous TEM observation.
2. Thermal desorption spectra above 940 K was correlated with the microstructure change of bubble migration and sweeping of helium bubbles by moving grain boundaries during re-crystallization.
3. One dimensional migration of helium bubbles along grain boundary dislocations was clearly demonstrated.
4. The bubble diffusivity along the grain boundary dislocation was estimated for the first time using a proportional relation ship between the mean square of the migration distance and time. The value determined was similar or a little higher than that of bubble diffusion in the matrix.

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