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Journal of Nuclear Materials 283–287 (2000) 249–254

journal of nuclear materials

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Development of vacancy clusters in neutron-irradiated copper at high temperature

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Abstract

Pure copper was fission-neutron-irradiated at 473 and 573 K from 0.0003 to 0.14 dpa. In copper, which was irradiated at 573 K to 0.0003 dpa, the number of vacancies which were accumulated in the largest stacking fault tetrahedron (SFT) was 276, while it was 470 in the smallest voids. This is explained by a model in which at 573 K a SFT converts to a void when the number of vacancies exceeds about 400. In 573 K irradiated copper, the number of vacancies in a SFT and a void of average size increases with the neutron fluence. The number of vacancies in a void increases more rapidly than that in a SFT. The reason appears to be that small vacancy clusters relax at 573 K to a string-like cluster, move as a cluster and coalesce. Experimental results are presented which show the movement of voids. In copper which was neutron-irradiated at 573 K, the number density of SFTs and voids peaked at 0.0003 dpa and decreased with fluence. The reason appears to be a low sink efficiency of dislocations for point defect absorption at 0.0003 dpa. Due to a low sink efficiency straight extended dislocations were decorated with many interstitial clusters. After jogs are formed on dislocations by joining with grown interstitial loops, the absorption efficiency of point defects increases significantly which lowers the density of SFTs and voids with increasing of dpa.

1. Introduction

In copper irradiated at high temperature, voids and stacking fault tetrahedra (SFT) form by clustering of vacancies, which are produced in displacement cascades. With these vacancy clusters and interstitial clusters, a damage structure evolves in neutron-irradiated metals. A large number of experimental results have been published previously [1–3]. Singh and Zinkle [4] published a review article in which they summarized experimental results and the atomistic mechanisms of damage structure evolution. Shimomura and Mukouda [5] reported a variation of the number density of voids with neutron fluence that was different from the standard relation reported in the review paper of Singh and Zinkle [4]. In Shimomura and Mukouda's experiment [5], the number density of voids observed in pure copper irradiated at 573 K in a temperature controlled rig at Kyoto University Reactor (KUR) and Japan Materials Testing Reactor (JMTR) was largest at the smallest dose in these experiments and gradually decreased with increasing neutron fluence. These results were not well received in the community due to the significant differences from previous experimental results, and the accuracy of the specimen temperature during irradiation was suspected [6].

Shimomura and Mukouda [7] carried out a computer simulation to investigate the atomistic mechanism of the formation of SFT and voids in irradiated copper. At an elevated temperature, 604 vacancies, which were introduced as small vacancy clusters, relax to a spherically distributed cluster during a molecular dynamics (MD) run [7]. The initial small vacancy clusters are representative of vacancy clusters, which are formed at the core of damage cascades, and the result suggests that a void can be nucleated from a clustering of only vacancies [7].

The objective of the present work is to explain these peculiar results with a unified model which includes the

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0022-3115/00/$ - see front matter
PII: S0022-3115(00)00071-4

dynamic behavior of SFT and voids which were observed experimentally.

2. Experiments and computer simulations

In the experiment, the dynamic behavior of SFT and voids at high temperature was examined. Thin specimens of neutron-irradiated copper were mounted on the heating stage of a double tilting, side-entry holder for a JEM-2000EX electron microscope. Temperature was increased in 50 K steps from 473 K. Observations at each step were made for 30 min. During heating of the specimens, TEM images were recorded on a SONY KCA-60BRS video recorder. This experiment was especially carried out to investigate the movement and disappearance of SFT and voids at high temperature.

Computer simulations were carried out with an isotropic potential of embedded atom method (EAM) potential due to Daw and Baskes [8] which was parameterized with the Foiles' procedure [9] by de la Rubia [10]. The present EAM potential of Cu calculates intrinsic stacking fault energy of 35.4 mJ/m^2. To carry out a simulation of annihilation of interstitial clusters on a dislocation line, we utilize a crystal composed of 26208 atoms. The dipole of an extended edge dislocation was introduced along the y-axis. A periodic boundary condition was applied for planes, which are perpendicular to x-, y- and z-axis. The extended width between two Schockly partial dislocations was $12b$ where b is the nearest neighbor distance.

3. Results

3.1. The variation of dislocation structure with neutron fluence in pure copper irradiated at 573 K

Figs. 1(a)–(c) show the dislocation structure observed in pure copper irradiated at 573 K to 0.0003, 0.0014 and 0.046 dpa, respectively. In copper irradiated to 0.0003 dpa (Fig. 1(a)), interstitial clusters decorate straight dislocations. In Fig. 1(b), these interstitial clusters coalesce to grow large interstitial loops near line dislocations during neutron irradiation. These interstitial loops join the dislocation line and form a jogged shape dislocation. Fig. 1(c) shows a dislocation bowed by absorbing interstitial clusters, which was observed in 0.046 dpa irradiated copper. Small interstitial clusters can be seen at the expansion side of the dislocation. The segregation of interstitial clusters along the dislocation as seen in Fig. 1(a) could be due to the difficulty of absorption of interstitial atoms on dislocations, which creates extended jogs.

3.2. The variation of the number density and vacancy content of SFT and voids with the neutron fluence at 473 and 573 K

Fig. 2 shows that the variation of the number density of SFT and voids with dose (dpa). Although part of the results were reported previously by Shimomura et al. [5], they are included here to see how that density decreases with increasing dose (dpa). Fig. 3 shows the variation of the number of vacancies contained in an average sized SFT and void with dose (dpa). At 573 K, the number of vacancies in both SFT and voids increase with increasing dose. The number of vacancies in voids rapidly increases with dpa faster than that in SFT. The increase is smaller for SFT and void for irradiations at 473 K. As seen in Fig. 3, the number of vacancies, which are accumulated in a void, is very large compared with that in a SFT. In copper neutron-irradiated at 573 K to 0.0003 dpa, the maximum SFT size was 6.1 nm of edge length (276 vacancies) while the minimum void size was 2.2 nm in diameter (470 vacancies).

3.3. Coalescence of SFT and movement of voids during high temperature annealing in copper

Mukouda and Shimomura [11] previously reported that voids move as a cluster during high temperature annealing. Figs. 4(a) and (b) show voids before and after their movement at 523 K. These voids move along $\langle 1\,1\,0\rangle$ as shown in Fig. 5. Detailed examination showed that voids showed pre-movement relaxation. Before a void moves, it moves back and forth along a $\langle 1\,1\,0\rangle$ direction and relaxes to an elongated shape along the $\langle 1\,1\,0\rangle$ axis. Voids were not imaged during movement, but were observed after motion at a position displaced from the original position. It was also observed in the present work that a void moved to SFT and the two coalesced.

During heating of TEM specimens, SFTs coalesced [12]. It is frequently observed that two or three SFTs coalesced during observations. It is noted that SFTs disappear abruptly at high temperature without shrinking in size.

4. Discussion and summary

As shown in experimental results, interstitial clusters were observed to segregate along dislocation lines. These straight dislocation lines were thought to exist in annealed pure copper before neutron-irradiation. Fig. 6 shows the result of a computer simulation of segregation of interstitial clusters along an extended edge dislocation dipole. After a model crystal, which included an extended edge dislocation dipole, is thermally equilibrated, 68 interstitial atoms (20 single interstitial plus 12 tetra-interstitial cluster) were introduced at the random

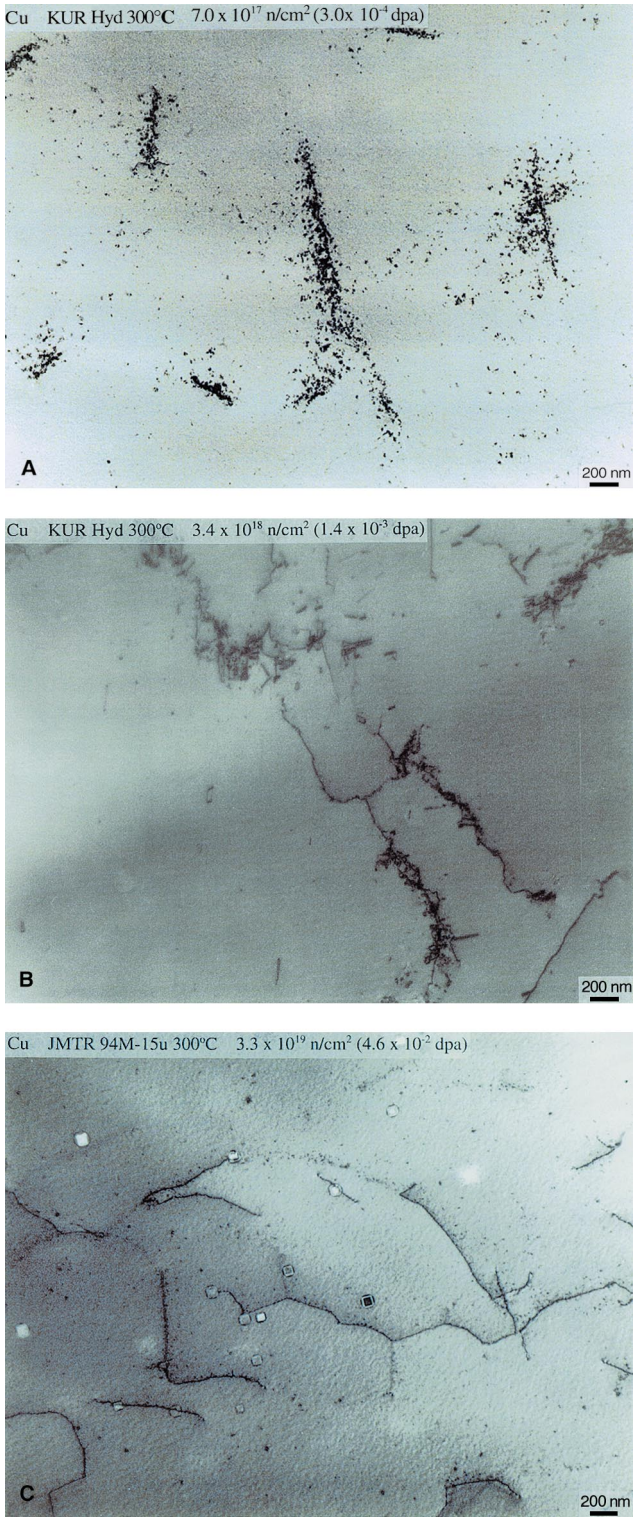

Fig. 1. Development of dislocation structure in neutron-irradiated pure copper at 573 K at different stage of dose. (a) 0.0003; (b) 0.0014; (c) 0.046 dpa.

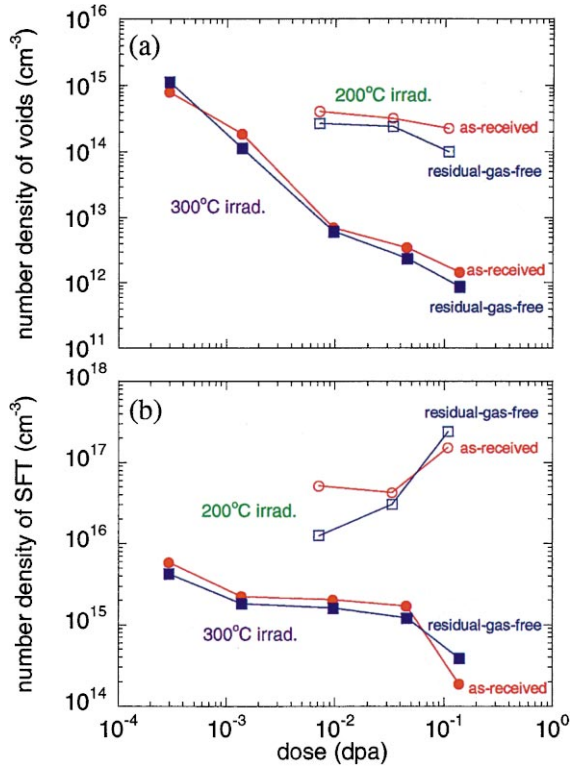

Fig. 2. The number density of SFT and void versus dpa in neutron-irradiated copper at 473 and 573 K.

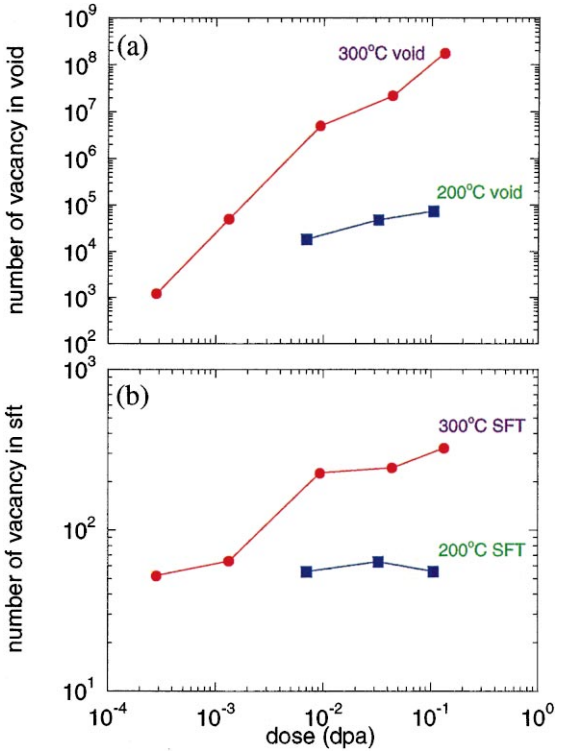

Fig. 3. The number of vacancy accumulated in a SFT and a void versus dpa in neutron-irradiated copper at 473 and 573 K.

positions in the crystal. An interstitial atom was introduced in the crystal at an octahedral position as an extra-atom. After MD of 60 ps at 500 K, all interstitial arrived at the dislocations. We carefully examined graphically the structure of the dislocation to see if the interstitial atoms were absorbed as extended jogs. Interstitial atoms sat near the dislocation but were not absorbed as extended jogs. Interstitial clusters in Fig. 6 grew on the expansion side of the dislocation.

An explanation for the observations goes as follows. As shown in Fig. 1(a), the decoration of interstitial clusters along a straight dislocation was observed only in copper irradiated to the lowest fluence of 0.0003 dpa. This can be explained by the difficulty of absorption of interstitial clusters on extended straight dislocations because this creates extended jogs. With increasing neutron fluence, these interstitial clusters start to aggregate near the dislocation. The large interstitial loops then join the dislocation as shown in Fig. 1(b), and the dislocation changes to a jogged shape, which can absorb point defects. After the formation of the jogged structure, the decoration of interstitial clusters along dislocation disappears as shown in Fig. 1(b). With increasing neutron fluence, the dislocation grows to a curved shape on which many jogs exist. Dislocations at this stage can absorb both interstitial clusters and vacancy clusters if they migrate to the dislocations. The sink efficiency for interstitial atoms and vacancies thus changes significantly and depends on the neutron-irradiation dose.

For the 573 K irradiation at the lowest fluence in the present work, the dislocations could not absorb point defects fast enough, and interstitial clusters are segregated near the dislocation through the dislocation strain field. Vacancy clusters are left in the center of the grain. At 573 K these vacancy clusters migrate as clusters and coalesce with each other to grow in size. As shown in Fig. 3, the average size of SFT in 573 K irradiated copper increases with neutron fluence. The results suggest that vacancy clusters grow by absorbing vacancies during irradiation. As indicated by Osetsky et al. [13], SFT larger than 6 nm are not observed in neutron-irradiated copper. Shimomura and Mukouda [7] proposed a model for conversion of a SFT to a void at high temperature if the number of vacancy exceeds roughly 400. At high temperature vacancies in a cluster tend to distribute spherically rather than forming a platelet [7]. The vacancy cluster, which contains vacancies of more than 400, relaxes to a void. Void nucleation by this mechanism occurs without gas atoms in the clusters. This is consistent with experimental observations [5]. It is

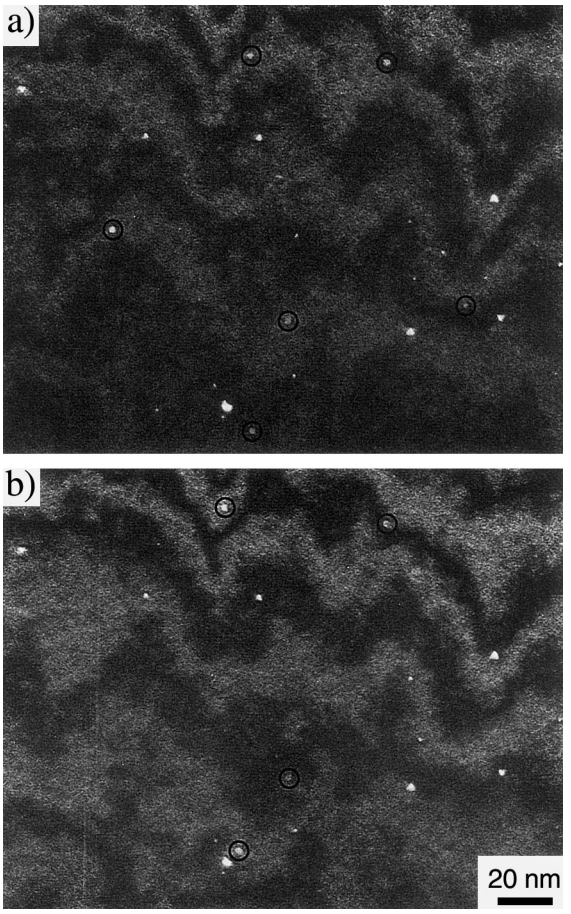

Fig. 4. Movements of voids during annealing. TEM pictures of (a) and (b) were taken from the same place with the same diffration condition. A specimen was annealed at 573 K for 20 min. after photographing of (a) and then taken a picture of (b).

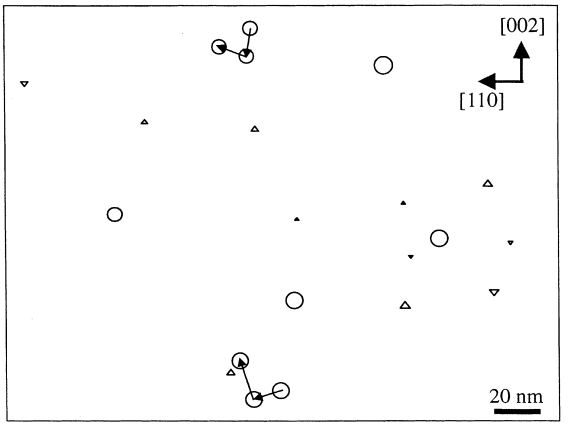

Fig. 5. ⟨1 1 0⟩ movement of voids. The movement of voids was examined from the picture of (a) and (b) of Fig. 4.

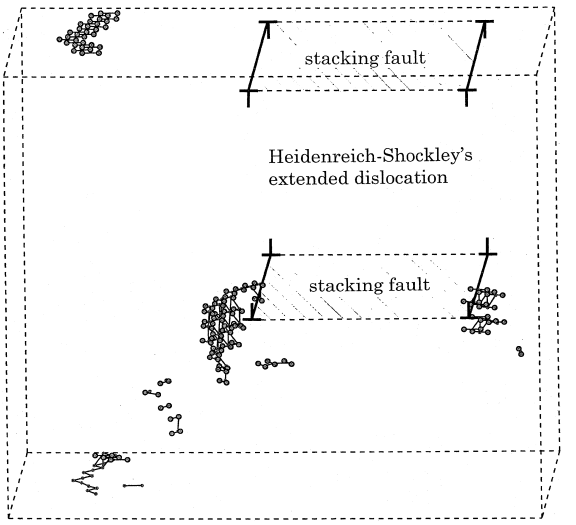

Fig. 6. Computer simulation of segregation of interstitial cluster to dislocations.

noted that the void nucleation is due to a process-sensitive reaction and not through the minimum energy path.

With the increasing sink efficiency of dislocations for point defect absorption, mobile vacancy clusters disappear at dislocations, leading to a decrease in number density with increasing neutron fluence. At a dose of 0.0014 dpa, voids grew to an average size of 7.3 nm in diameter, but even at this size of voids, the number density of voids decreased with increasing neutron fluence. To explain the experimental results on the variation of void number density with neutron fluence, voids of this range of size have to disappear to sinks during an irradiation. The TEM annealing experiments of thin foils which contains SFT and voids shows that voids move at high temperature.

SFT disappear abruptly at high temperature without a decrease in size. Computer simulations suggest that an atom located outside the tetrahedron jumps into a tetrahedron. Subsequently a SFT changes to a string-like structure [12]. In the case of voids a similar process occurs in which a void relaxes to a string-like vacancy cluster and moves. At a dose of 0.0014 dpa, a dislocation line had a zone denuded of voids. At doses of 0.0014 dpa, voids grew to a fairly large size (9.5 nm in diameter). Such voids can be intersected by dislocations climbing rapidly in the crystal. Such a large void does not shrink even if it is connected to a dislocation. This is partly due to the inclusion of gas atoms in voids. Some voids on dislocation lines can be unified into a large stable void through a pipe diffusion of vacancies. This can cause a decrease in the number density of voids. At 0.014 dpa, it was observed that many dislocation lines intersect a void [14]. This structure is typical of voids which were observed in heavily irradiated copper at FFTF-MOTA [15].

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