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Dynamical Behavior of Voids in Neutron-irradiated Copper at Elevated Temperature

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ABSTRACT

We reported previously an experimental result which shows that voids can move in neutron-irradiated copper at elevated temperature. To study the detailed behavior of voids, we carried out annealing experiments and *in-situ* observations in neutron-irradiated copper. Neutron irradiation was carried out in the temperature controlled capsule at KUR (Kyoto University Reactor). Neutron irradiation was performed at 300°C to damage levels between 10^{-4} to 10^{-3} dpa. The annealing temperature was 250°C for 10, 20, 30 min sequentially for neutron- irradiated copper. After annealing TEM observation was carried out at room temperature. Experimental results show that voids moved along the [110] direction. Voids moved during 250°C annealing but small stacking fault tetrahedra (SFT) were not changed. We observed 37 voids and 8 voids moved, the others vanished during annealing. The images of *in-situ* observation, void contrast images were recorded on VTR tape and analyzed frame by frame. At room temperature observation voids were static and were seen as white circles; however at 300°C the contrast changed to an oval shape and sometimes disappeared. After 7 sec contrast was a white circle and moved slightly and similar phenomena were observed quite frequently at 300°C. It is concluded that void moved with dynamical structural relaxation at elevated temperature.

INTRODUCTION

Copper and nickel are used as typical FCC metals in radiation damage studies. Many studies have been carried out and reviewed by Singh and Zinkle [1]. They concluded that there is a lack of information on the microstructure of copper and nickel irradiated to below 10^{-2} dpa at 100 to 300°C [1]. Zinkle and Snead carried out fission neutron irradiation at 230°C to damage levels between 10^{-2} and 10^{-1} dpa [2]. They concluded that a high density of small SFT and dislocation loops was observed in copper and nickel, and small voids were observed in irradiated copper. Recently Shimomura and Mukouda carried out fission neutron irradiation of copper at 200 and 300°C at a similar range of dose, we reported dose dependence of voids and SFTs previously [3-5]. The present work is carried out to examine the evolution of vacancy clusters and voids in neutron-irradiated copper of transient regime at elevated temperatures. To obtain precise results, the irradiation was carried out at the temperature controlled irradiation facility in the KUR.

EXPERIMENTAL PROCEDURES

The specimens used in this study were pure copper (Dowa Ministry Ltd.) with a nominal purity of 99.9999%. Some specimens was degassed by melting in vacuum evacuated to 10^{-5} Pa. Both kinds of specimens were cold-rolled to 0.05 mm and punched out to disks of 3mm in diameter, and annealed in vacuum. Both types of specimens, as-received and degassed ones,

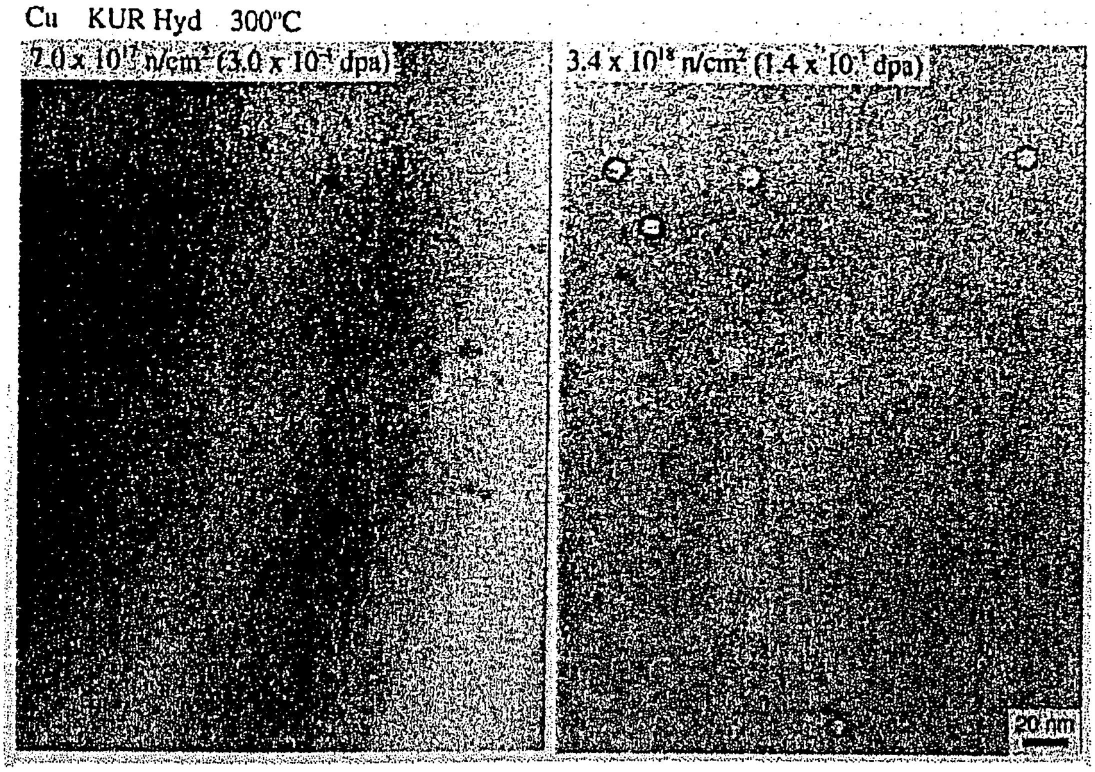

***Figure 1.** TEM observation of neutron-irradiated copper at 300°C (a) 3.0 x 10^{-4} dpa and (b) 1.3 x 10^{-3} dpa by bright field image. The mean size of voids was increased with increasing the dose from 3.0 nm to 7.0 nm.*

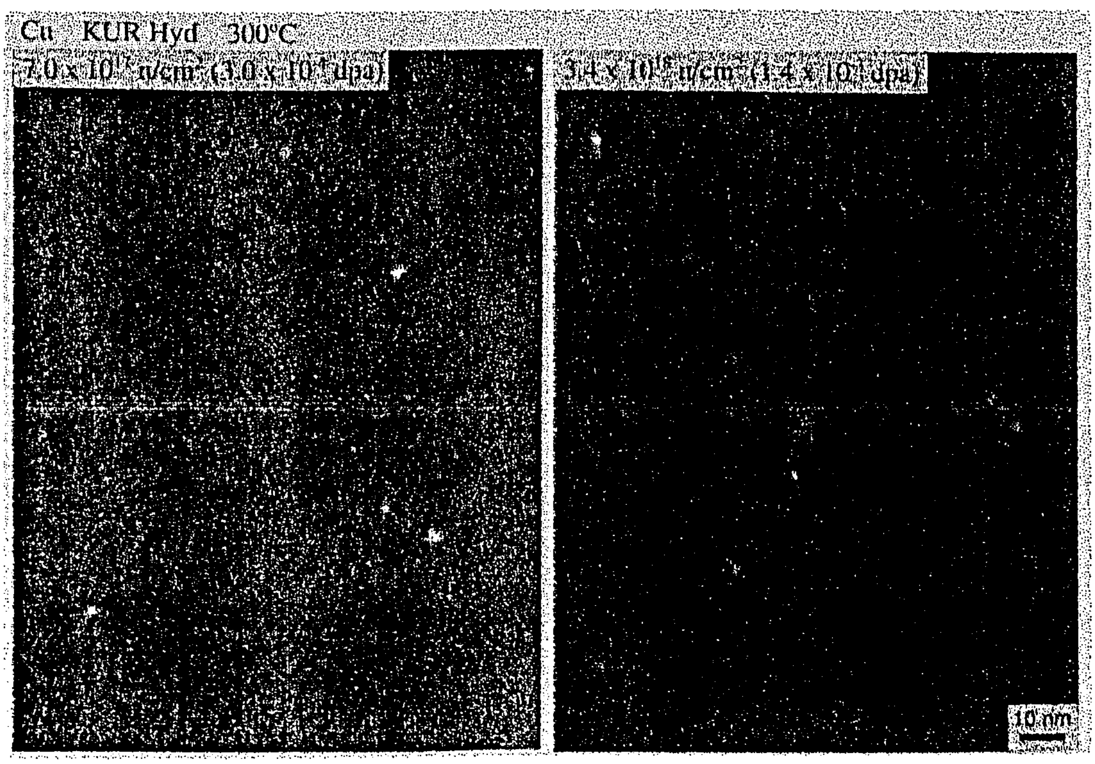

***Figure 2.** TEM observation of neutron-irradiated copper at 300°C (a) 3.0 x 10^{-4} dpa and (b) 1.3 x 10^{-3} dpa by dark field image. The mean size of SFTs was slightly increased with increasing the dose.*

were irradiated in temperature controlled rigs at KUR. Neutron irradiation was carried out in the temperature controlled capsule at KUR reactor in Hyd for 5 and 24 hours. The specimen temperature was kept at 300°C during irradiation. The neutron fluence (E>0.1MeV) was 7.0 x 10^{17} and 3.4 x 10^{18} n/cm^2 corresponding to 3 x 10^{-4} and 1.4 x 10^{-3} dpa, respectively. After radiation cooling, specimens were electro-polished and using JEOL-2000EX TEM at an accelerating voltage of 200kV. Void images were observed by bright field technique of off-Bragg diffraction condition (void contrast) and weak beam dark field (WBDF) image.

RESULTS AND DISCUSSIONS

In neutron-irradiated copper at 200°C to 10^{-1} dpa, pre-existing dislocation was observed to be decorated by interstitial clusters as reported previously [3]. In neutron-irradiated copper at 300°C to 3.0 x 10^{-4} dpa, pre-existing dislocations were observed to be decorated by interstitial clusters [6]. At 300°C irradiation, segregation of interstitial clusters were clearly seen at lower dose because mobility of clusters was larger than for irradiation at 200°C. Interstitial clusters grew by absorption of small clusters by strain field effects when the dose increased to 1.4 x 10^{-3} dpa. These clusters were perfect loops without stacking fault contrast. If interstitial clusters grew and touch dislocation lines, the dislocation line changed to a cusp shape. The number of interstitial clusters observed along the dislocation decreased with increasing dose. Uniformly distributed straight dislocations developed at the stage of 10^{-2} to 10^{-1} dpa irradiated at 300°C and the most of the voids were connected dislocation lines. The number of interstitial clusters along dislocations decreased with increasing dose. This result shows that vacancy and interstitial clusters aggregate along dislocations, while vacancies and their clusters form SFTs and voids in the medium. The number density of SFT slightly decreased with increasing dose and the mean size was 2.6 nm, no difference was observed between as-received and residual-gas-free specimen. Voids were observed and the number density of voids decreased with increasing dose [5]. The mean size of voids increased with increasing dose from 3.0 to 7.0 nm as shown in Fig. 1. However the mean size of SFTs increased slightly as shown in Fig. 2. The number of vacancies to be accumulated in a void is larger than in a SFT. This result suggests that voids move and coalesce on increasing the dose. To investigate movement of voids, annealing experiments were carried out. The annealing temperature was 250°C for 10, 20, 30 min sequentially, after annealing TEM observation was carried out at room temperature. Fig. 3 shows that the weak beam dark field image of the void position corresponding to the annealing step. The void moved along [110] direction as shown in Fig. 3(e). Voids moved during 250°C annealing but SFTs were not changed. We observed 37 voids and 8 voids moved, the rest of them vanished or did not move during annealing. We think that movement of voids is due to strain field sensitive motion because small vacancy clusters move with string shape in MD computer simulation by Shimomura et al. [7]. Fig 4 shows images from *in-situ* observations of a 3.0 nm void. Void contrast images were recorded on VTR tape and analyzed frameby frame (1/30 sec). At room temperature voids were static and exhibited white circle contrast as shown in Fig. 4(a), however at 300°C contrast changed to an oval shape (Fig. 4(b)) and sometimes contrast disappeared as shown in Fig. 4(c). After 7 sec contrast was a white circle and moved slightly as shown in Fig. 4(d). For 30 min observations similar phenomena were observed quite frequently at 300°C. It is concluded that the void moved with dynamical structural relaxation at high temperature like Fig. 4(b). A large void, size 10.0 nm, also moved frequently. Fig. 5 shows the void images observed at 350°C for 70 sec. The void stands still and sometimes moves. The structure relaxation in the process of movement is observed again in the oval shape as in Fig. 4(b).

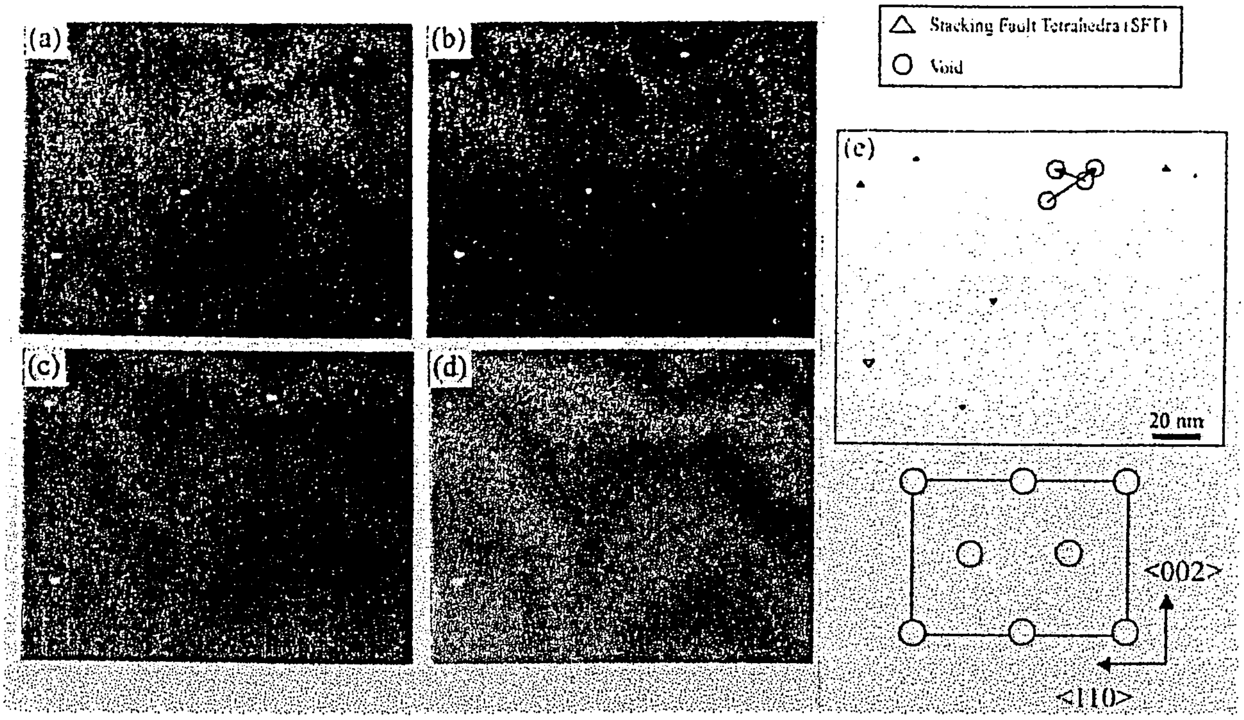

Figure 3. Change of annealing step of neutron-irradiated copper at 300°C irradiated to 3.0 x10^{-4} dpa (a) 250°C 0 min (b) 250°C 10 min (c) 250°C 30 min (d) 250°C 60 min (e) schematic drawing of this region.

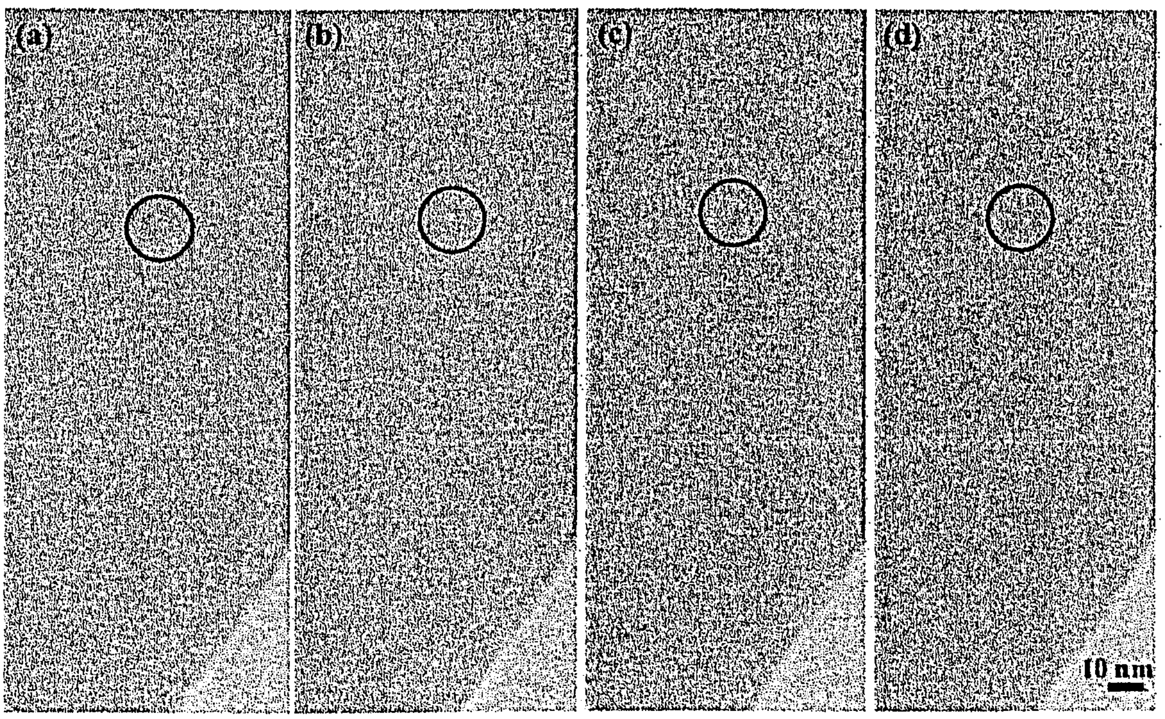

Figure 4. In-situ observation at 300°C of neutron-irradiated copper at 300°C 3.0 x 10^{-4} dpa (a) 1' 32" 24 (b) 1'33"26 (c) 1'34"00 (d) 1'38"24. Diameter of the void is 3.0 nm.

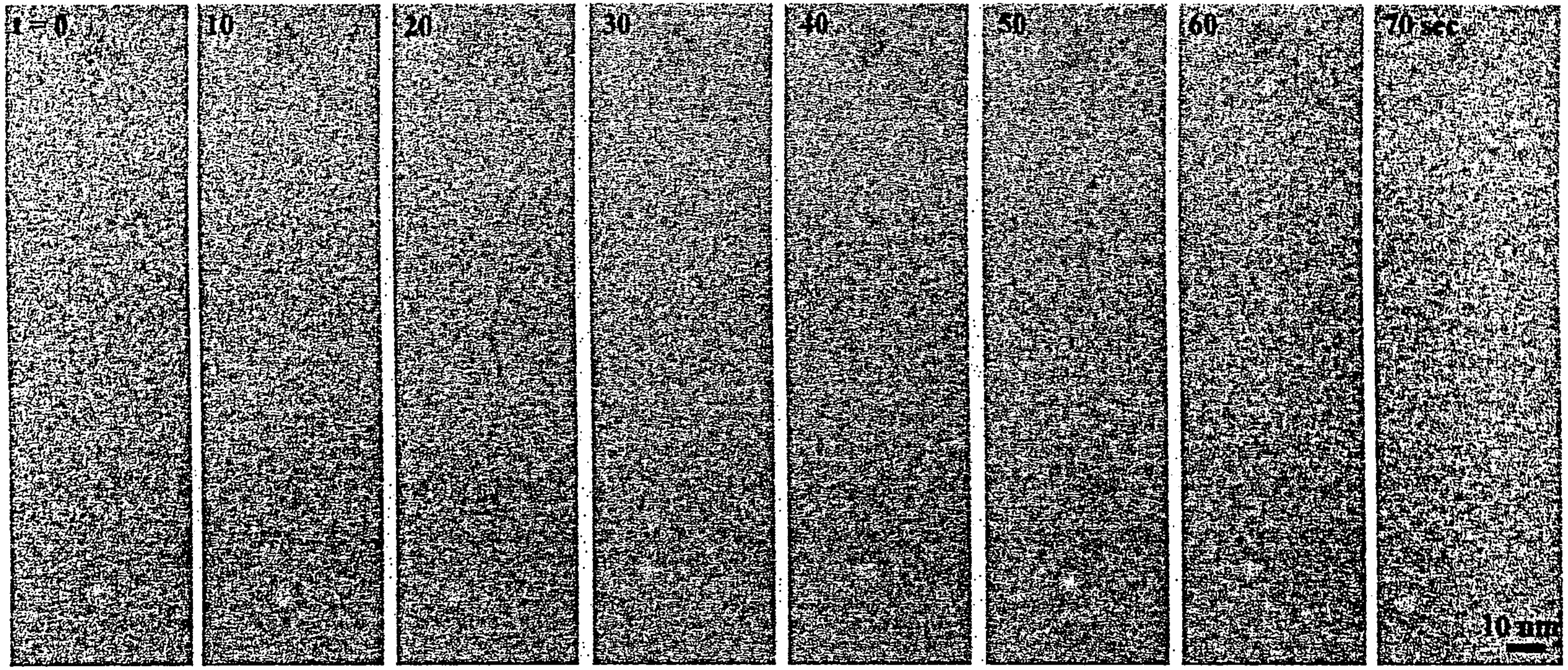

Figure 5. In-situ observation at 300°C of neutron-irradiated copper at 350°C 1.4 x 10^{-3} dpa for 70 seconds. Diameter of the void is 10.0 nm. Each frame is shown every 10 seconds. The black spot of the upper part of each frame is the contamination on the surface of specimen.

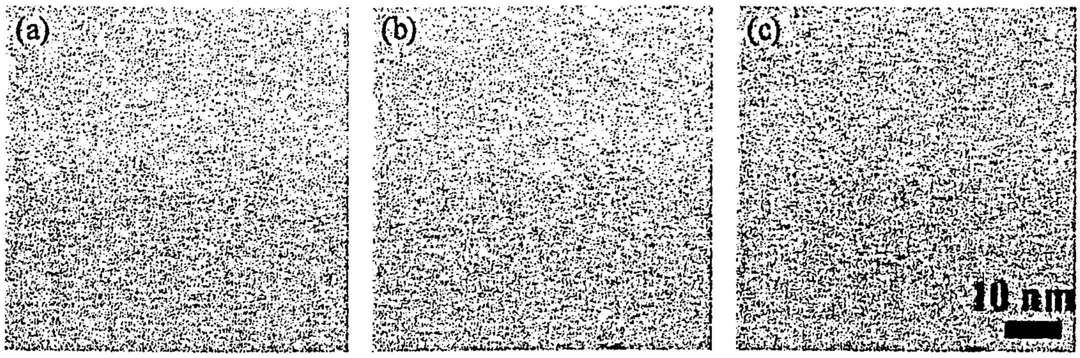

Figure 6. In-situ observation at 350°C of neutron-irradiated copper at 300°C 1.4 x 10^{-3} dpa. Diameter of the void is 16.0 nm. At first void was one, after a few seconds void divided in three as shown in (b).

Furthermore a large void was observed to split as shown in Fig. 6(b) and (c). The size of void was 16.0 nm. At first there was a single void, after a few seconds void divided in three as shown in Fig 6(b). After that the small voids changed structure as shown by the oval white contrast.

SUMMARY

It was observed that interstitial clusters in copper segregate along dislocations irradiated at 300°C to low dose of order in 3 x 10^{-4} dpa. These interstitial clusters start to aggregate to a larger cluster along dislocation and form perfect dislocation loops. Dislocation developed jogged structure with increasing dose. Interstitial clusters along dislocations decreased with increasing dose. Vacancies and their clusters form voids and SFTs, voids can move above 250°C. This result suggests that the growth of voids occurs by the coalescence of vacancy clusters. As a result voids grow with increasing dose in neutron-irradiated copper.

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