ELSEVIER

Journal of Nuclear Materials 283–287 (2000) 302–305

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Microstructure in pure copper irradiated by simultaneous multi-ion beam of hydrogen, helium and self ions

I. Mukouda [a,*], Y. Shimomura [a], T. Iiyama [a], Y. Harada [a], Y. Katano [b], T. Nakazawa [b], D. Yamaki [b], K. Noda [b]

[a] *Applied Physics and Chemistry, Faculty of Engineering, Hiroshima University, 1-4-1 Kagamiyama, Higashi-Hiroshima 739-8527, Japan*

[b] *Department of Materials Science, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 391-1195, Japan*

Abstract

Pure copper was irradiated at 300–500°C by 5 MeV Cu ions (single beam) and Cu ions plus gas atoms (H and He) (dual beam irradiation) simultaneously. The high energy ion irradiation was carried out with the accelerator TIARA at the Takasaki-establishment of JAERI. The ions stop within a few microns from surface level and damage was formed up to this depth. The damage structure was observed as a function of the depth utilizing a focused ion beam (FIB) device. Below 300°C irradiation with a single beam produced a high density of stacking fault tetrahedra (SFT) but void formation was not observed. Large voids were observed with single beam irradiation at 500°C. In specimen irradiated with a dual beam of helium and Ni ions, the number density of voids was increased significantly. In copper irradiated with hydrogen and Ni ions, the number density of voids was not so large. Experimental results show that helium atoms promote void formation. Hydrogen atoms have less effect on void formation than helium atoms in pure copper.

1. Introduction

For fusion reactor applications, there is interest in copper and its alloys. Hydrogen and helium atoms are generated by nuclear transmutation in the fusion environment. These gas atoms play an important role on the evolution of the damage microstructure. It is well known that helium is active in cavity nucleation [1]. Some studies have been carried out by Bullen et al. [2] on hydrogen effects. However, it is not extensively characterized. In the present work, quantitative experiments were carried out to study the role of gas atoms (helium and hydrogen) on the evolution of the damage microstructure in irradiated materials. It is possible to control the concentration of gas atoms in irradiated metals by ion irradiation at high energy. We examined void formation in high energy ion-irradiated pure copper by a single beam (5 MeV Cu) and a dual beam (5 MeV Cu ion and 260 keV H ion, and 5 MeV Ni ion and 600 keV He ion) irradiation. The ion energy was selected so that the projected range of the gas ions in copper coincide with depth of peak damage (1.2 μm) calculated by the TRIM 95 code as shown in Fig. 1. Specimens for transmission electron microscope (TEM) cross-sectional observation was prepared by a focused ion beam (FIB) device. The relation between gas atoms and damage structure was derived from experimental results.

2. Experimental procedure

The specimens used in this study were pure copper (Dowa Ministry) with a nominal purity of 99.9999%. Annealed disks of 3 mm in diameter and 0.05 mm thick were prepared [3]. The high energy ion irradiation was carried out with TIARA accelerator at the Takasaki-establishment of JAERI. The ions stop within 1–2 μm from the surface and damage was formed within this depth. The damage microstructure was observed as a function of the depth by utilizing an SMI9200 (Seiko

* Corresponding author. Tel.: +81-824 24 7857; fax: +81-824 22 7192.

E-mail address: mukouda@ipc.hiroshima-u.ac.jp (I. Mukouda).

0022-3115/00/$ - see front matter
PII: S 0 0 2 2 - 3 1 1 5 (0 0) 0 0 1 3 2 - X

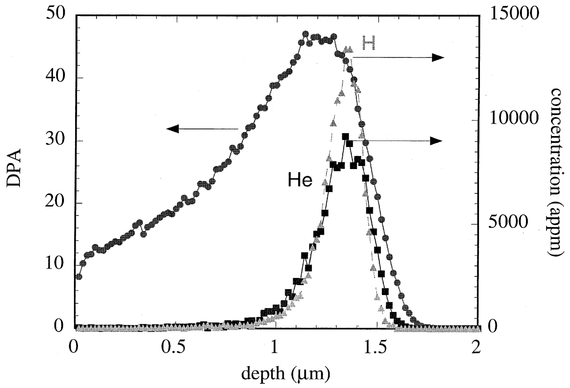

Fig. 1. Depth profile of DPA by 5 MeV Cu ion and gas concentration of 600 keV He and 260 keV H ions in pure copper calculated by the TRIM code.

Instrument) FIB microscope. The FIB generates 30 keV Ga ions and bombards the specimen surface with a glancing angle. Atoms on the surface are removed by sputtering. To preserve the surface position of ion-irradiated metals, we deposited Cu on the irradiated surface by electroplating to prevent erosion of the specimen surface [4,5]. In previous work, it was found that interstitial atoms form cluster throughout FIB-thinned specimens. It is very difficult to distinguish damage defects, which are formed by high energy ion irradiation from FIB damage. To solve this problem, we developed a cryotransfer-FIB thinning technique, in which the specimen is cooled down to 100 K on the FIB thinning holder (the cryo-FIB technique) and transferred to a TEM without warming up. This technique makes it possible to observe voids, even very small ones, in high energy ion-irradiated metals [5]. In ion-irradiated metals

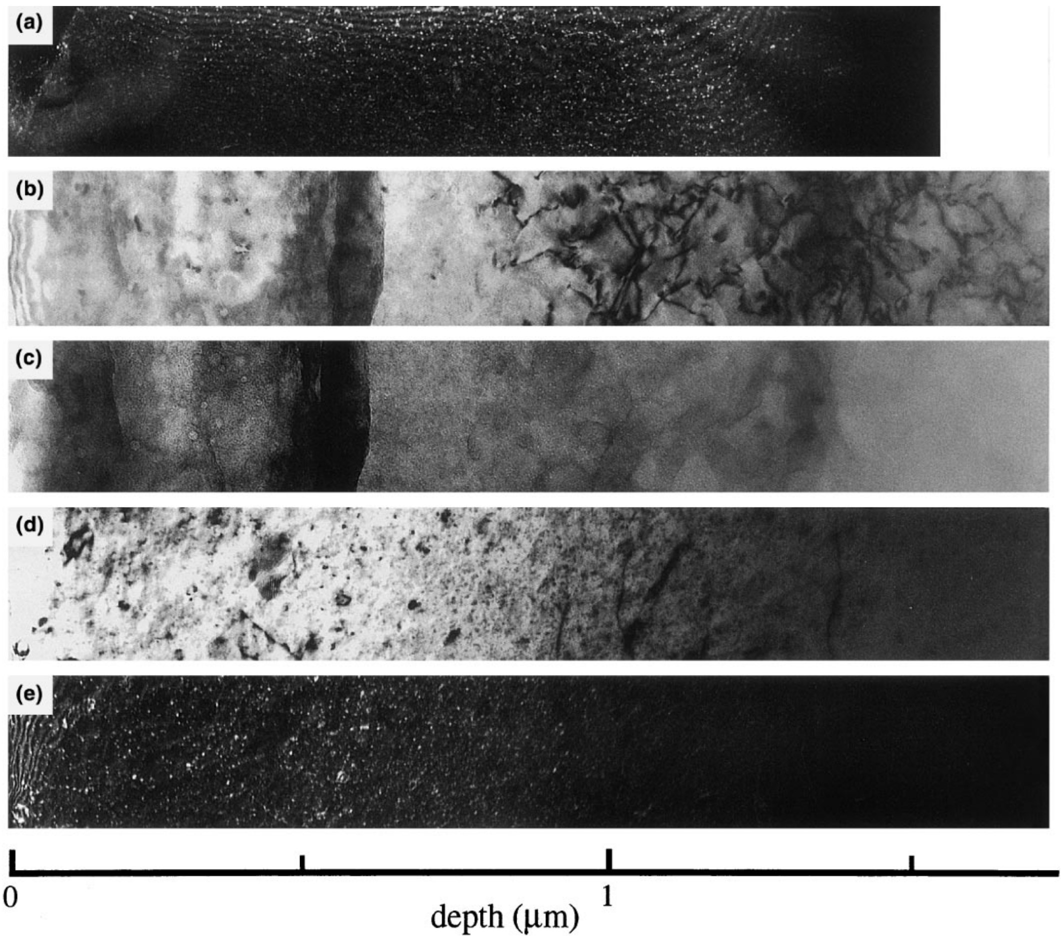

Fig. 2. The microstructure of ion irradiated pure copper at 300°C: (a) 3 MeV Cu ion; (b) 5 MeV Cu + 600 keV He (dislocation contrast); (c) 5 MeV Cu + 600 keV He (void contrast); (d) 5 MeV Cu + 260 keV He (dislocation contrast); (e) 5 MeV Cu + 260 keV He (weak beam image).

at high temperature, voids, interstitial clusters and dislocations are formed. Even with the FIB technique mentioned above, it is difficult to observe the dislocation structure, which is developed by clustering of interstitial atoms in irradiated specimen. To overcome this difficulty, we developed a TEM specimen preparation method, which is a combination of cryo-FIB thinning and electro-polishing. In annealed pure copper, many dot defects are observed with FIB thinning at room temperature. However in the case of thinning at 100 K, defects were smaller than with thinning at room temperature. Furthermore to remove regions damaged by the Ga ions, the specimens were electro-polished in a solution of 33% HNO_3 and 67% ethanol cooled to −40°C at an applied potential of 6 V for 6 s. After electro-polishing, no dot defects were observed. It was found that the damage microstructure of ion-irradiated metals such as copper could be observed without FIB damage.

3. Results and discussion

Fig. 2(a) shows the microstructure of pure copper irradiated at 300°C by 3 MeV Cu ion irradiation. Many stacking fault tetrahedron (SFT) were observed in the irradiated region. Figs. 2(b) and (c) show the microstructure observed by dislocation and void contrast in Cu and He dual beam irradiated copper at 300°C, respectively. Many voids were observed and the density of voids was 3.5×10^{22} m^{-3} at the peak of implanted He atoms as shown in Fig. 3(a). Number density and size of voids depended on the depth. The number density of voids gradually increased and the size of voids decreased with increasing dose and He concentration. In Cu and He irradiated copper at 400°C, the number density of voids increases significantly at the depth corresponding to the range of helium atoms as shown in Fig. 3(b). Figs. 2(d) and (e) show the microstructure observed by dislocation and weak beam images in Cu and H dual beam irradiated copper at 300°C, respectively. Figs. 3(c) and (d) show the microstructure observed by weak beam dark field image in Cu and H ions dual beam irradiated copper at 300°C and 400°C, respectively. Many SFT were observed in the irradiated region, similar to the single ion irradiation results. In Cu and H irradiated copper at 300°C and 400°C, no voids were observed. The number density and mean size of vacancy clusters at the damage peak (depth = 1.2 μm) are summarized in Table 1. It is concluded that the total number of vacancies for Cu and H ion irradiation was smaller than that for Cu and He ion irradiation. It is considered that a large number of invisible vacancy clusters combined with hydrogen atoms or molecules are formed in the specimen irradiated with Cu and H ion. These results suggest that the mobility of vacancy clusters, which trap hydrogen atoms, is suppressed. In the pure copper irradiated at 500°C by a single beam (Ni ion), voids were observed, but the number density of voids was very low [6]. In the dual beam (Ni and He ion) irradiated copper, the number density of voids increased significantly at the depth corresponding to the range of helium ions. In dual beam (Ni and H ion) irradiated copper, only very large size voids were observed and the number density of voids was lower than that for the He dual beam irradiation [5]. The depth profile of voids was uniform in the

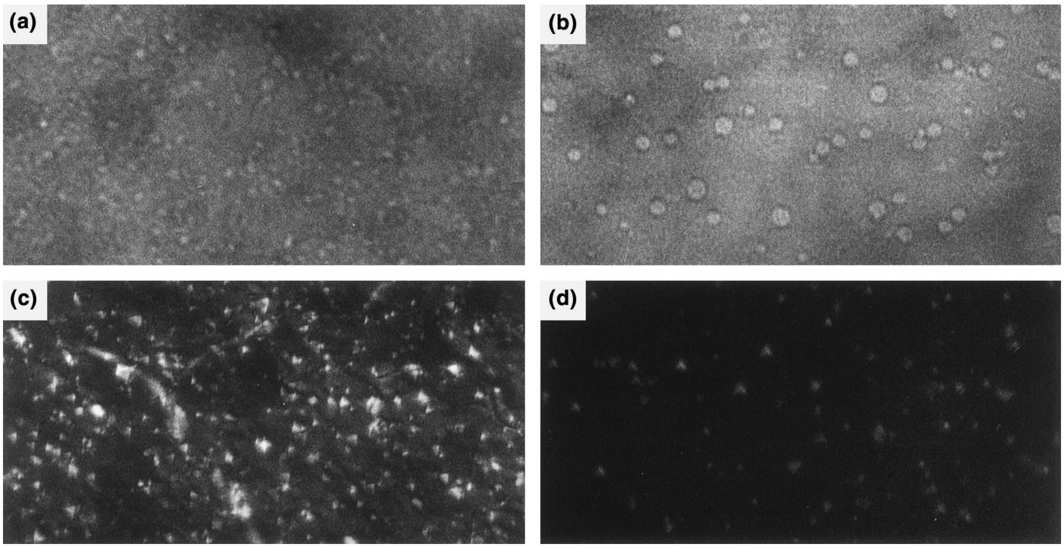

Fig. 3. Void contrast images at the damage peak in pure copper irradiated with Cu and He ion at: (a) 300°C; (b) 400°C, respectively. Weak beam dark field images at the damage peak in pure copper irradiated with Cu and H ion at: (c) 300°C; (d) 400°C, respectively.

Table 1
Number density and mean size of vacancy clusters at the damage peak (depth = 1.2 μm)[a]

	5 MeV Cu + 600 keV He irrad.		5 MeV Cu + 260 keV H irrad.	
	N_{void} (m^{-3})	d_{void} (nm)	N_{SFT} (m^{-3})	d_{SFT} (nm)
300°C	3.5×10^{22}	1.8	4.9×10^{22}	3.8
400°C	1.5×10^{22}	4.7	2.2×10^{22}	4.3

[a] *N*: Number density, *d*: mean size.

Cu and H ion dual beam irradiation. Since diffusion of hydrogen is much higher than helium, effects of hydrogen can occur outside the calculated implanted hydrogen distribution. In the triple beam (Ni, He and H ion) irradiated copper, the number density of voids was similar to Cu and He dual beam irradiation results. He effects on void nucleation appear to be much larger than H effects in Cu.

4. Summary

In pure copper, helium atoms promote void nucleation for irradiation at 300°C, 400°C and 500°C. While hydrogen atoms suppressed void formation at 300°C and 400°C, more voids were observed at 500°C than for the specimen exposed to single (Cu ion) beam irradiation. These results show that hydrogen atoms trap small vacancy clusters at low temperature. Vacancy clusters and H atoms or molecules dissolved with increasing temperature and very large voids were formed at 500°C. In pure copper, helium atoms promote void formation. Hydrogen atoms have less effect on void formation than helium atoms in pure copper.

Acknowledgements

The authors are grateful to Dr H. Watanabe for a part of ion irradiation experiment. This work was supported partly by grand-in aid from to Ministry of Education, Science, Sports and Culture, Japan. This work has been carried out as a part of the Universities-JAERI Joint Research Project for the Advanced Radiation Research.

References

[1] K. Farrell, Radiat. Eff. 53 (1980) 175.
[2] D.B. Bullen, G.L. Kulcinski, R.A. Dodd, J. Nucl. Mater. 122&123 (1984) 584.
[3] I. Mukouda, Y. Shimomura, T. Iiyama, Y. Katano, D. Yamaki, T. Nakazawa, K. Noda, Mater. Res. Soc. Symp. Proc. 540 (1999) 549.
[4] S.J. Zinkle, R.L. Sindelar, Nucl. Instrum. and Meth. B 16 (1986) 154.
[5] S.J. Zinkle, G.L. Kulcinski, R.W. Knoll, J. Nucl. Mater. 138 (1986) 46.
[6] Y. Shimomura, I. Mukouda, Mater. Res. Soc. Symp. Proc. 540 (1999) 527.