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Journal of Nuclear Materials 283–287 (2000) 179–182

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Correlation between defect structures and hardness in tantalum irradiated by heavy ions

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Abstract

Pure tantalum specimens were irradiated with 2.4 MeV Cu^{2+} ions up to 3 dpa at temperatures between room temperature and 1073 K. Transmission electron microscope (TEM) observation and micro-indentation tests were carried out to correlate the microstructure and the hardness. Significant radiation hardening occurred at temperature ranging from 673 to 873 K. Isochronal annealing of a specimen irradiated at room temperature up to a dose of 0.3 dpa resulted in a rapid increase in hardening between 573 and 673 K and continued to increase up to 873 K. The microstructure showed that the formation of small defect clusters is the major reason for both the radiation hardening and the radiation-anneal hardening. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Tantalum is one of the candidate materials for structural components of diverters because of its high toughness, high-sputtering threshold energy, high fabricability [1] and low-activation properties [2]. Tantalum is also a candidate protective first wall material for fusion reactors [3] and target material for spallation neutron sources [4] which are also subject to a high flux of high-energy neutrons. Our previous papers showed that siginificant change of mechanical properties was expected below ~1100 K because of the formation of a high density of defect clusters [5]. In an authors' previous paper, dose dependence of microstructure and microhardness were compared. In the study, some of the hardness increase was attributed to transmission electron microscope (TEM)-invisible defect clusters [6]. In the present study, further detailed TEM observation, addition and revision of the hardness estimation and post-irradiation annealing were carried out.

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2. Experimental

The material used in this study was 99.95% pure tantalum with impurities of ~30 wt. ppm O, ~20 wt. ppm N and ~30 wt. ppm C. After cold rolling and punching into TEM disks, the specimens were annealed at 2073 K for 300 s in a vacuum of $<3\times10^{-5}$ Pa for recrystallization.

Irradiation was performed with 2.4 MeV Cu^{2+} ion at several temperatures between room temperature and 1073 K using a Tandem type accelerator in Kyushu University. The damage level of the irradiation varied from 0.03 to 3 dpa at its peak position of damage.

After the irradiation, specimens were back-thinned by electropolishing with a solution of 2.5% HF, 5% H_2SO_4 and 92.5% methanol at about 230 K. Microstructures at the depth of 250 nm were observed with a JEM-2000EXII TEM at Kyushu University. The micro-indentation tests were also conducted on both unirradiated and irradiated specimens at room temperature using an Elionix ENT-1100 with a load of 1 gf. A triangular pyramidal diamond indentor (Berkovich type) with a semi-apex angle of 65° was used in this study.

The previous paper indicated that the indentor load (L) and displacement (d) were approximated as follows [7]:

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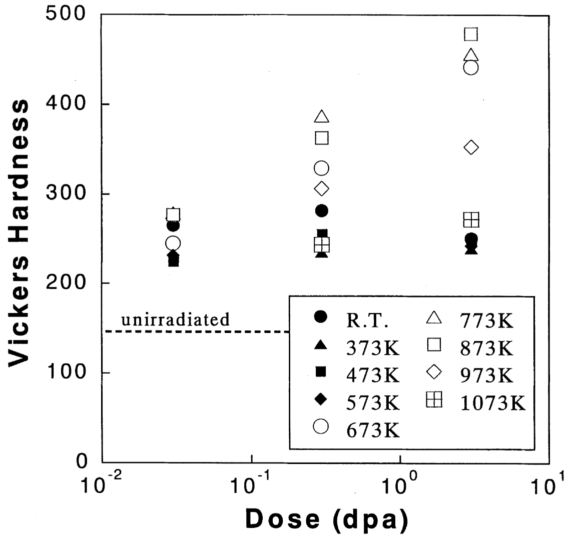

Fig. 1. The dose dependence of hardness of tantalum irradiated with heavy ions at temperatures between room temperature and 1073 K.

$$L/d = Ad + B. \tag{1}$$

They showed good correlation between the coefficient A and Vickers hardness (Hv) for several materials as the following empirical relation:

$$A(\mathrm{GPa}) \sim 0.287\mathrm{Hv}. \tag{2}$$

3. Results

Fig. 1 shows the dose dependence of hardness at temperatures between room temperature and 1073 K. Hardening below 573 K saturates at a low level. On the other hand, at temperatures between 673 and 873 K, the hardness increases with dose. Above 873 K, hardening decreases with increasing temperature. It must be emphasized that the dose dependence of hardening is quite different below 573 K and above 673 K.

To examine the difference in defect structure, the microstructure of specimen irradiated at 573 and 673 K up to 3 dpa was observed and is shown in Fig. 2. Vacancy loops of about 4 nm were observed in dark field images at both temperatures. A high density of voids of <2 nm diameter were formed at 673 K but not at 573 K as shown in the void images in Fig. 2. Small white spots, which are presumably due to strain field around voids, were also observed in the dark field image at 673 K. The density of small voids is about two orders of magnitude higher than that of vacancy loops. Therefore, the difference of hardness at 573 and 673 K is due to the existence of a high density of voids.

In order to obtain the annealing induced change in the effects of irradiation on the hardness change as a function of isochronal annealing temperature of the specimen irradiated at room temperature and 0.3 dpa is shown in Fig. 3. Solid and open circles represent irradiated and unirradiated specimens, respectively. For irradiated specimens, there is a prominent hardness increase at 573–673 K. The high hardening is maintained up to 873 K, and then decreases with increasing the annealing temperature. On the other hand, hardness change is not observed in unirradiated specimens at any temperature.

Fig. 4 shows the microstructure after annealing at 723 and 1073 K for 1 h irradiated at room temperature and at 0.3 dpa. Dark field image shows that small loops and a high density of small dot defects of ~1 nm, which are probably vacancy clusters, appeared at 723 K. At

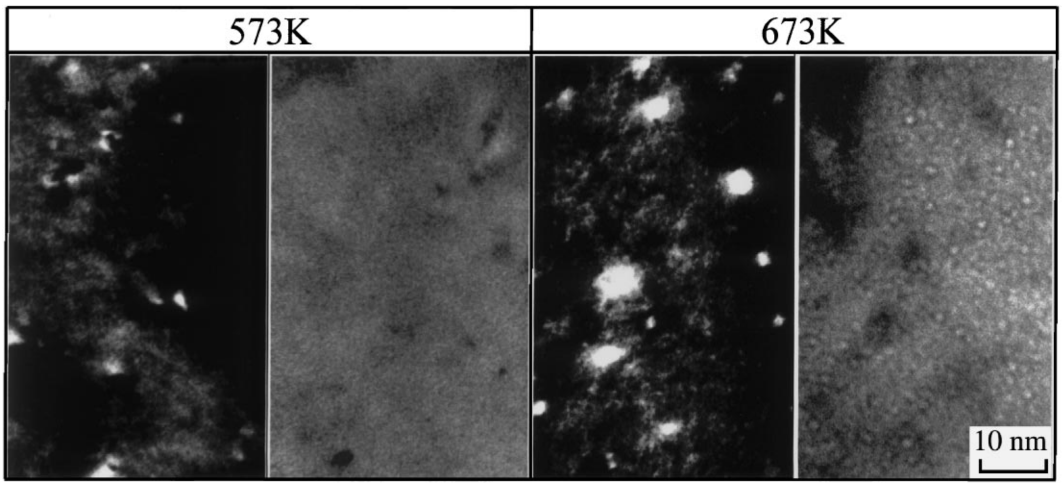

Fig. 2. The microstructure of tantalum irradiated to 3 dpa at 573 and 673 K.

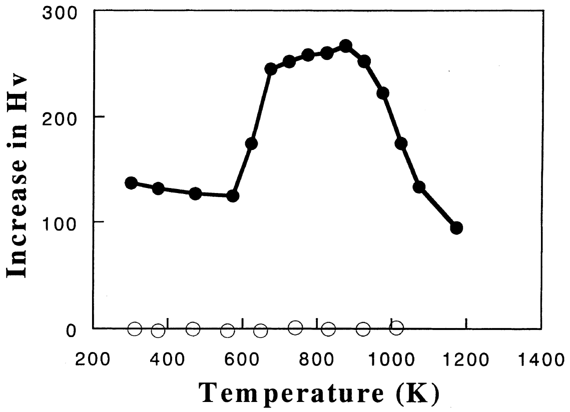

Fig. 3. Hardness changes of tantalum irradiated at room temperature to 0.3 dpa as a function of annealing temperature. The specimens were annealed for 1 h at each temperature.

1073 K, dot defects are not observed in dark field images. Instead, a low density of voids of about 2 nm was formed. These results suggest that the hardness increase above 573 K is attributed to small vacancy clusters as observed 723 K.

4. Discussion

The effect of the observed defect clusters on yield strength increase was estimated for the case of irradiation at 3 dpa. Yield strength increment is given by

$$\Delta\sigma_y = M\alpha\mu b(Nd)^{1/2}, \tag{3}$$

where M is the Taylor factor which has a value of 3 [8], α the barrier strength which depends on the nature of defect cluster, μ the shear modulus, b the Burgers vector magnitude, and N and d are the density and size of the defect clusters, respectively. In this study, the total increase in yield strength was calculated using the following equation [8]:

$$\Delta\sigma_{\text{total}} = \left[\Delta\sigma^2_{\text{loops}} + \Delta\sigma^2_{\text{voids}}\right]^{1/2} + \Delta\sigma_{\text{disl}}. \tag{4}$$

A correlation between the yield strength and hardness was used as follows [9]:

$$\text{Hv} = \sigma/3. \tag{5}$$

A comparison between the micro-indentation measurements and the yield strength increment calculated from the microstructure is given in Fig. 5. The barrier strength α of 0.2 was applied for dislocations. Assuming that the difference of hardness between 573 and 673 K at 3 dpa is caused by small void formation as shown in Fig. 2, we can obtain the value α of 0.2 and 0.05 for void and vacancy loop, respectively. Vacancy loops and dislocations caused an increase in yield strength below 573 K. The

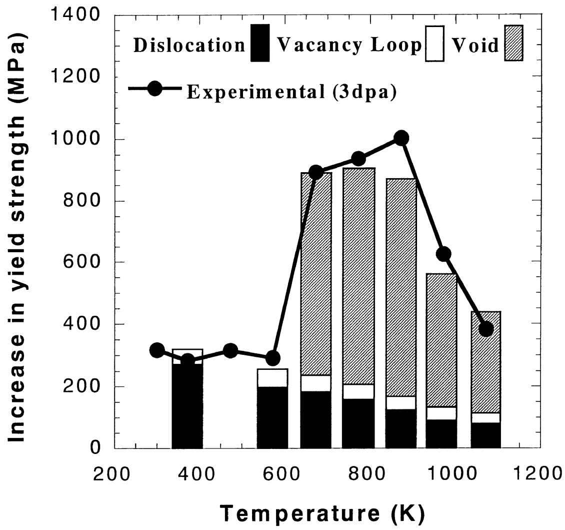

Fig. 5. The comparison between experimental and calculated yield strength increase of tantalum irradiated at temperatures between room temperature and 1073 K to 3 dpa.

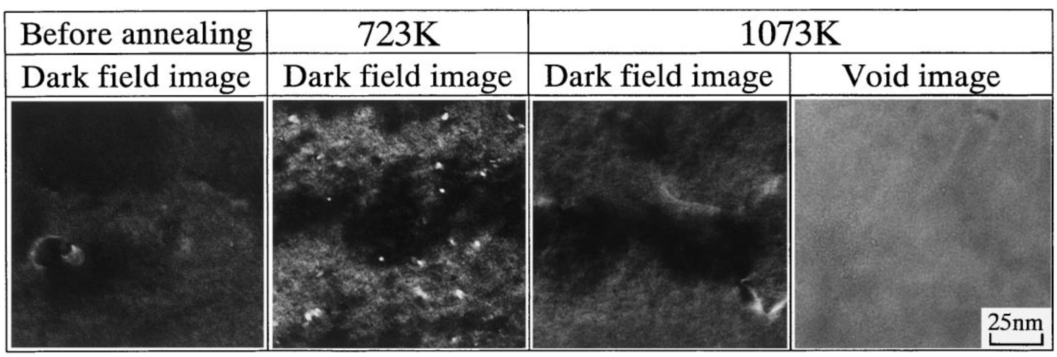

Fig. 4. The microstructure of tantalum irradiated at room temperature to 0.3 dpa after annealing for 1 h at 723 and 1073 K.

additional formation of small voids resulted in significant hardening between 673 and 873 K.

Below 573 K, lower hardening occurred comparing with higher temperatures. From the hardness test, evidence of radiation damage saturation could be seen. The hardness only becomes two times higher than that of unirradiated value at a dose as high as 3 dpa. These results suggest that the process of mutual recombination is dominating below 573 K, which resulted in suppressing void formation. Similar results were reported on tantalum irradiated 800 MeV proton at room temperature [10]. In the study, tensile strength was increased by a factor of two at the dose of 1.5 dpa. The total elongation remained about 14%. However, strain hardening was greatly reduced compared with the unirradiated specimen. According to the loss of work hardening ability resulting from dislocation channeling observed in Nb [11], it is suggested that the reduction in uniform elongation would lead to brittle failure called plastic instability in pure tantalum irradiated below 573 K.

The annealing experiment of specimen irradiated at room temperature also produced an increase in hardness at temperatures between 573–673 K. This phenomenon is known as radiation annealing hardening (RAH), and is observed in V [12], Nb [13] and Mo [14]. These studies indicate that the RAH was due to the formation of impurity-defect complexes [12], small vacancy clusters [14] and the condensation of vacancies onto dislocations [13,14]. In this study, post-irradiation annealing at 723 K caused the formation of small dot defects that were presumably vacancy aggregates. Therefore, small vacancy clusters seem to be responsible for the RAH in pure tantalum. This result suggests that vacancies were accumulated as mono-vacancies or very small aggregates due to their low mobility during irradiation at room temperature, and were then rearranged during the annealing.

5. Conclusions

Correlation of microstructure and hardness in heavy ion-irradiated pure tantalum has been studied. Significant hardening occurred under irradiation or post-irradiation annealing between 573–673 K. TEM observation revealed that the formation of small voids is mainly responsible for radiation hardening in this temperture regime.

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