ARTICLE IN PRESS
JID: NME [m5G;June 22, 2016;22:21]

Nuclear Materials and Energy 000 (2016) 1–6

Contents lists available at ScienceDirect

Nuclear Materials and Energy

journal homepage: www.elsevier.com/locate/nme

Effect of self-ion irradiation on hardening and microstructure of tungsten

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ARTICLE INFO

Article history:
Available online xxx

Keywords:
Tungsten
Tungsten–rhenium alloy
Self-ion irradiation
Microstructure
Nanoindentation hardness

ABSTRACT

The irradiation hardening and microstructures of pure W and W – 3%*Re* for up to 5.0 dpa by self-ion irradiation were investigated in this work. The ion irradiation was conducted using 18 MeV W^{6+} at 500 and 800 °C. A focused ion beam followed by electro-polishing was used to make thin foil specimens for transmission electron microscope observations. Dislocation loops were observed in all the irradiated samples. Voids were observed in all of the specimens except the W–3%*Re* irradiated to 0.2 dpa. The hardness was measured by using nanoindentation. The irradiation hardening was saturated at 1.0 dpa for pure W. In the case of W – 3%*Re*, the irradiation hardening showed a peak at 1.0 dpa. The correlation between the microstructure and hardening was investigated.

1. Introduction

Tungsten (W) is considered to be a candidate materials for plasma-facing components (PFCs) because of its high melting point (3410 °C) [1], high thermal conductivity (166 W/m·K) [1], and high sputtering resistance. The materials for PFCs would be irradiated by neutrons (first wall: 30 dpa for 5 y, divertor: 15 dpa for 5 y) [2] during the operation of a fusion reactor. Thus, it is necessary to study the neutron irradiation effects in order to anticipate the changes in the microstructure and mechanical properties of W. Moreover, it is also important to improve the irradiation resistance of W to prevent the deterioration of its properties by the neutron irradiation. Williams et al. reported the suppression of void nucleation [3] by adding Rhenium (*Re*). Furthermore, Fukuda et al. reported a decrease in irradiation hardening [4], and Klopp reported a decrease in the ductile–brittle transition temperature (DBTT) by the addition of *Re* [5] without irradiation. The largest decrease in the DBTT and irradiation hardening were found when 2–3%*Re* was added to the W. Thus, the authors tried to improve the irradiation resistance of W by adding 3%*Re*.

To demonstrate the improvement in the irradiation resistance by adding a lower level of *Re*, it is necessary to compare the changes in the microstructure and irradiation hardening between W and W–3%*Re*. However, fission neutrons irradiation takes a long time to achieve a particular damage level such as 15 to 30 dpa. Therefore, the decision was made to use self-ion irradiation to obtain a high level of displacement damage without impurities.

In this study, the effects of self-ion irradiation on the microstructure and irradiation hardening of pure W and W–3%*Re* alloy were investigated. For this investigation, the microstructure and hardness were evaluated after self-ion irradiation, and we briefly discuss the microstructure differences after ion and neutron irradiation.

2. Experimental

Pure W and W–3%*Re* plates supplied by A.L.M.T. Corp, Japan, were fabricated using powder metallurgy and hot rolling. These plates were annealed at 900 °C for 20 min to relieve the internal stress introduced by the rolling. The chemical compositions of the specimens are listed in Table 1. To investigate the effects of the displacement damage and irradiation temperature on the microstructure and hardness, the irradiation was conducted at 500 and 800 °C with 18 MeV W^{6+} in vacuum ($< 5 \times 10^{-5}$ Pa).

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http://dx.doi.org/10.1016/j.nme.2016.06.005

Please cite this article as: T. Hwang et al., Effect of self-ion irradiation on hardening and microstructure of tungsten, Nuclear Materials and Energy (2016), http://dx.doi.org/10.1016/j.nme.2016.06.005

Table 1
Chemical composition of pure W and W–3%*Re* (unit: ppm, ∗%).

Material	*Re*∗	Al	K	Si	C	O	N	Ca	Cr	Cu	Fe	Mg	Mn	Mo	Ni	Sn	Na
Pure W	–	< 2	< 5	< 5	10	< 10	< 10	< 1	< 1	< 1	3	< 1	< 1	35	< 1	< 2	< 1
W–3%*Re*	3.0	5	< 5	< 5	< 10	< 10	< 10	< 1	6	< 1	78	< 1	< 1	44	< 1	2	3

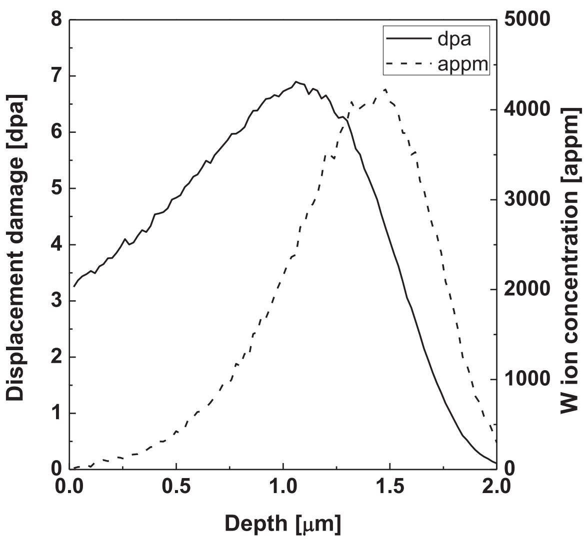

Fig. 1. W ion distribution and displacement damage in pure W calculated by SRIM ($E_d = 90$ eV [6]).

The irradiation experiment was conducted using a 3 MV tandem accelerator at the Takasaki Advanced Radiation Research Institute of the Japan Atomic Energy Agency (JAEA). Calculations of the damage distribution were conducted with the SRIM using 90 eV by the displacement of the shielding [6].

Fig. 1 shows the distribution of the displacement damage and implanted W concentration in pure W after irradiation. In this study, to reduce the effect of the implanted W atoms on the microstructure and mechanical properties, 500 nm was defined as the normal depth for the microstructure observation and hardness measurements.

After the irradiation, thin foil specimens were prepared for transmission electron microscope (TEM) observations using a focused ion beam (FIB) machine. Displacement damage was also induced on the specimen's surface during the FIB process; therefore, electropolishing was carried out to remove the induced defects. Electropolishing was conducted using a 0.5% NaOH solution at 5 ± 3°C and 18 V for 8 ms. After the electropolishing, the microstructure was observed using TEM. Dislocation loops were confirmed by the weak beam method, and voids were distinguished with over/under focus images. The hardness values of the pure W and W – 3%*Re* alloy before and after irradiation were measured using a nanoindenter (ENT-1100a, Elionix inc. Japan) at the International Research Center for Nuclear Materials Science of the Institute for Materials Research (IMR) at Oarai, Japan. Before the nanoindentation hardness measurements, an indentation tip calibration was conducted with the Oliver–Pharr method [7] using fused silica. Hardness measurements were conducted using a Berkovich indenter from the surface to a depth of 0.3 μm at 30 points using the depth-sensing indentation (DSI) method. The number of steps was 500, and the intervals were 20 ms.

3. Results

3.1. Microstructure

Dark-field images of dislocation loops and under-focus images of voids for pure W irradiated to 0.2, 1.0, and 5.0 dpa at 800 °C are shown in Fig. 2. Dislocation loops were observed in all the samples. In the pure W irradiated to 0.2 dpa, the mean size of the dislocation loops was approximately 3.1 nm. The mean size of the voids was 1.0 nm. In the pure W irradiated to 1.0 dpa, the mean size of the dislocation loops was approximately 3.3 nm, and the mean size of the voids was approximately 1.5 nm. In the pure W specimen irradiated to 5.0 dpa, the mean size of the dislocation loops was approximately 3.6 nm. With increasing displacement damage, the number density of the dislocation loops decreased. The size of the dislocation loops tended to increase with increasing damage. The number density of the voids increased with increasing displacement damage. The mean size of the voids increased between the displacement damage levels of 0.2 dpa and 1.0 dpa.

Fig. 3 shows dark-field TEM micrographs and void images of the W–3%*Re* irradiated to 0.2, 1.0, and 5.0 dpa at 800 °C. Dislocation loops were observed in the W and W–3%*Re*. The mean size of these dislocation loops was approximately 2.5 nm in the W–3%*Re* irradiated to 0.2 dpa. With increasing displacement damage to 1.0 dpa, the mean size of the dislocation loops increased to 3.6 nm. No voids were observed in the W–3%*Re* irradiated to 0.2 dpa. However, voids were observed in the W–3%*Re* irradiated to 1.0 dpa. Their mean size was approximately 1.3 nm. After irradiation to 5.0 dpa, the mean size of the voids was approximately 1.4 nm. The number density of the voids decreased between the W–3%*Re* specimens irradiated to 1.0 and 5.0 dpa.

Fig. 4(a) and (b) shows the size histograms of the dislocation loops and voids, respectively, in pure W irradiated at 800 °C. With increasing damage levels, relatively large dislocation loops were frequently observed. In the case of voids, a tendency similar to that of the dislocation loops was also observed. The pure W irradiated to 0.2 dpa showed voids of 1.0 – 1.5 nm in size. After irradiation to 1.0 dpa, voids 1.5 – 2.0 nm in size were observed.

Fig. 5(a) shows the size distributions of the dislocation loops in W – 3%*Re* after irradiation at 800 °C. Dislocation loops 2.0 – 7.0 nm in size were observed in the W–3%*Re* irradiated to 1.0 dpa. After 5.0 dpa of irradiation, dislocation loops 2.0 – 3.0 nm in size were observed in the W–3%*Re*. Fig. 5(b) shows the relationship between the void size and damage level. In the W–3%*Re*, no voids were observed after irradiation to 0.2 dpa. However, after irradiation to 1.0 and 5.0 dpa, voids were observed. In the W–3%*Re* irradiated to 1.0 dpa, voids 1.0 – 1.5 nm in size were observed. As for the size, voids 1.0 – 1.5 nm in size were observed after irradiation to 1.0 dpa. With increasing displacement damage to 5.0 dpa, voids 2.0 nm in size were observed. No precipitates were observed in any of the samples. The microstructure data are summarized in Table 2.

3.2. Irradiation hardening

The results of the nanoindentation hardness measurements and magnitude of the irradiation hardening are shown in Figs. 6 and 7, respectively. The hardness of the unirradiated pure W was 6.6 GPa, which increased to 8.4 GPa after irradiation to 0.2 dpa at 800°C.

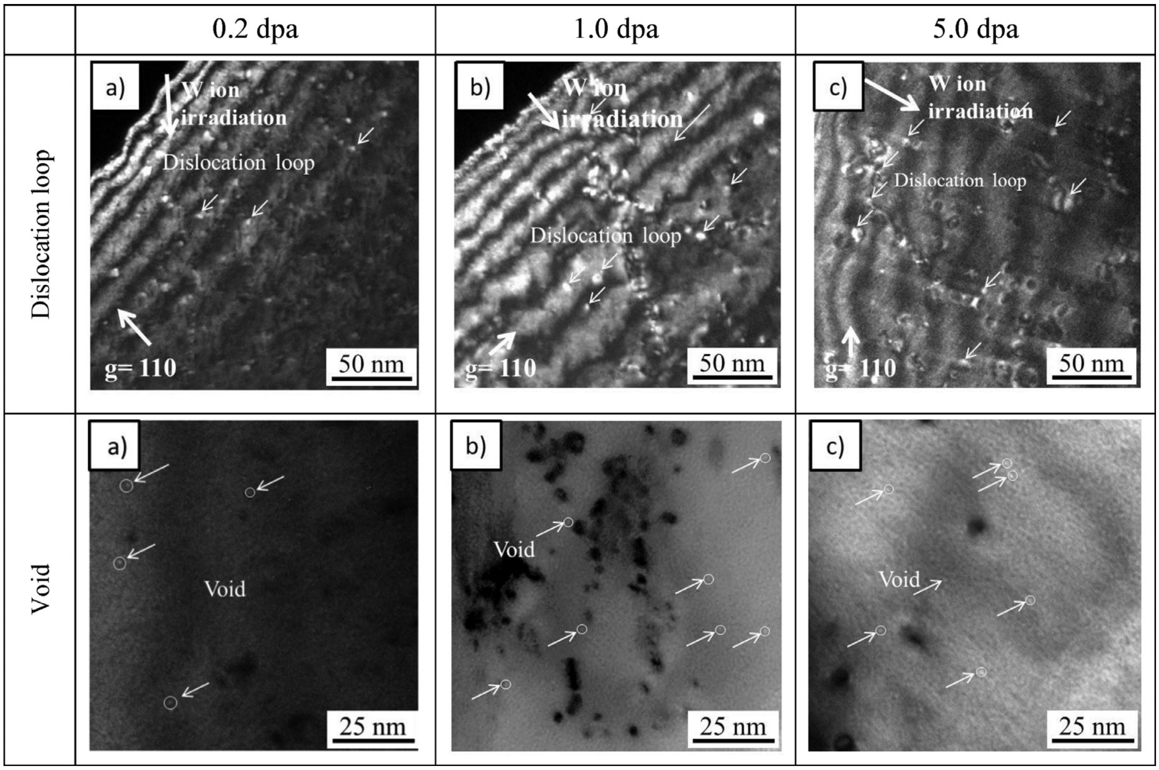

Fig. 2. Dark-field TEM micrographs and void images of pure W irradiated to (a) 0.2, (b) 1.0, and (c) 5.0 dpa at 800 °C.

Table 2
Relationship between ion irradiation results and neutron irradiation results [10].

Irradiation condition	Material	Damage level [dpa]	Irradiation damage rate [dpa/s]	Voids		Dislocation loops	
				Number density $[10^{22}/m^3]$	Size [nm]	Number density $[10^{22}/m^3]$	Size [nm]
Self-Ion	Pure W	1.0	4.3×10^{-5}	7.1	1.5	6.1	3.3
		5.0	3.0×10^{-4}	8.9	1.6	6.0	3.6
	W-3%*Re*	1.0	4.3×10^{-5}	13	1.3	3.8	3.6
		5.0	3.0×10^{-4}	7.0	1.4	3.9	3.0
Neutron	Pure W	1.5	1.5×10^{-7}	12	4.7	–	–

The hardness further increased to 9.5 GPa with an increase in the damage level to 1.0 dpa. However, no obvious differences in the magnitudes of the irradiation hardening between 1.0 and 5.0 dpa were observed, and the irradiation hardening was saturated at damage levels above 1.0 dpa. In the case of W – 3%*Re*, the hardness of the unirradiated specimen was 6.0 GPa, which increased to 8.4 GPa with an increase in the damage level to 0.2 dpa at 800 °C. The hardness of the W – 3%*Re* irradiated to 1.0 dpa was almost the same as that of the specimen irradiated to 0.2 dpa. However, the hardness decreased to 7.4 GPa after irradiation to 5.0 dpa.

The hardness of the pure W irradiated to 0.2 dpa at 500 °C was 7.9 GPa. With an increase in the damage level to 1.0 dpa, the hardness of the specimen increased to 9.3 GPa. In the case of irradiation at 500°C, there was no obvious hardness difference between the specimens irradiated to 1.0 and 5.0 dpa. The W – 3%*Re* irradiated at 500°C showed the same hardness tendency as the specimen irradiated at 800°C. The hardness of the W – 3%*Re* irradiated to 0.2 dpa was 8.6 GPa. The hardness then increased to 9.3 GPa with an increase in the damage level to 1.0 dpa. However, the hardness of the specimen irradiated to 5.0 dpa showed a decrease in the hardness to 8.4 GPa.

4. Discussion

The size distribution of the dislocation loops in the pure W irradiated at 800 °C showed that the size of the major dislocation loops increased with an increase in the damage level. In the case of voids, the same tendency was observed. The size of the dislocation loops in the W – 3%*Re* irradiated at 800 °C also increased with an increase in the damage level. However, the W - 3%*Re* specimen irradiated to 0.2 dpa was different from the pure W irradiated to 0.2 dpa, and no voids were observed in the W – 3%*Re*. According to a report by Suzudo et al. [8], a W – *Re* mixed dumbbell was formed by combining self-interstitial atoms (SIAs) and *Re* substitutional atoms. This might cause an increase in the probability of recombination between the vacancies and interstitial atoms and suppress the void nucleation and growth. As a result, the formation of voids was suppressed in the W – 3%*Re* irradiated to 0.2 dpa compared to pure W that was irradiated to 0.2 dpa.

The irradiation hardening of the W – 3%*Re* specimen irradiated to 0.2 dpa at 800°C was higher than that of the pure W. This was possibly caused by irradiation–induced *Re* cluster formation [9]. On the other hand, irradiation, the smaller irradiation hardening in the W – 3%*Re* irradiated to 5.0 dpa at 800 °C compared to that irradiated to 1.0 dpa at 800°C could have been caused by decreases in

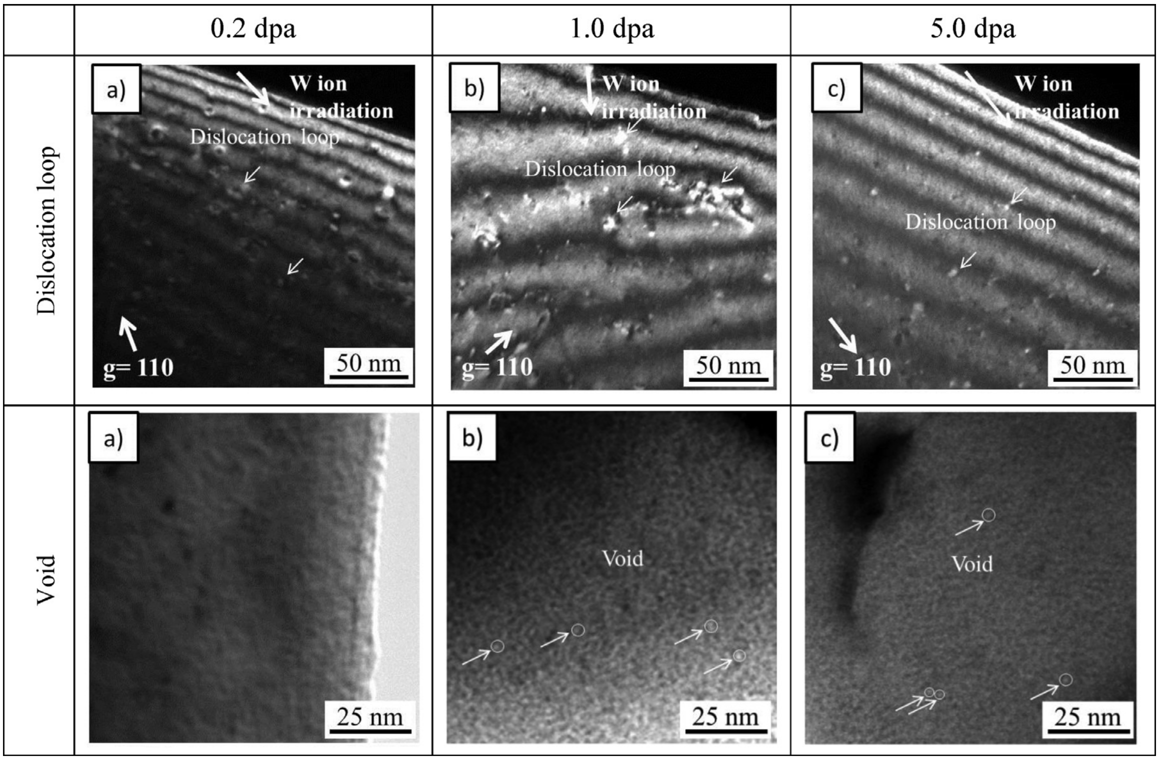

Fig. 3. Dark-field TEM micrographs and void images of W–3%*Re* irradiated to (a) 0.2, (b) 1.0, and (c) 5.0 dpa at 800 °C.

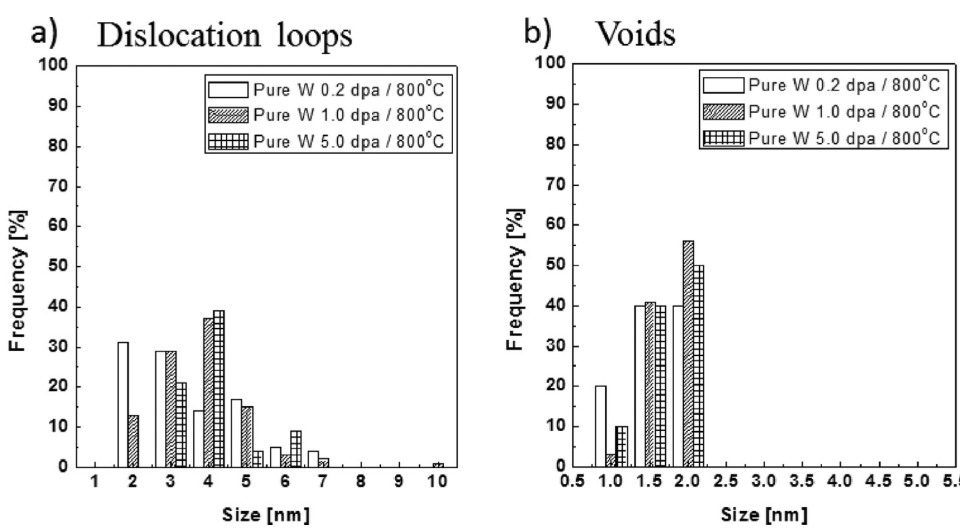

Fig. 4. Size distribution of (a) dislocation loops and (b) voids in pure W after irradiation at 800 °C.

the number density of the voids and size of the dislocation loops. This might have been caused by decreasing of defect cluster by mutual recombination of vacancy and SIA trapped by the W–*Re* dumbbells or by the recovery of the dislocations introduced by the material fabrication process before irradiation.

Table 2 also lists our previous neutron irradiation data [11]. Neutron irradiation was carried out using Joyo at JAEA. As a result of the neutron irradiation of pure W to 1.5 dpa at 750°C, only voids were observed, which formed a lattice structure. In the case of self-ion irradiation, the number density and size of the voids were smaller than those with neutron irradiation. Dislocation loops were observed in the pure W after self-ion irradiation. This difference was probably caused by the damage rates (neutrons: 1.5×10^{-7} dpa/s [11], ions: 4.3×10^{-5} dpa/s). The neutron irradiation period was approximately 100 times longer than the ion irradiation. Thus, point defects could have had enough time to grow in the case of the neutron irradiation. The microstructure without dislocation loops after the neutron irradiation might be attributed to the recovery of dislocation loops and the vacancy absorption at this temperature.

To anticipate the irradiation hardening from the microstructure, the calculations were based on the following equation (Orowan equation) [12,13]:

$$\Delta\sigma_y = M\alpha\mu b\sqrt{Nd} \tag{1}$$

where M is a Taylor factor, α is a constant depending on the irradiated alloy and defect cluster type, μ is the shear modulus, and b is the Burgers vector. The values of M, μ, and b are 2.0, 151 GPa,

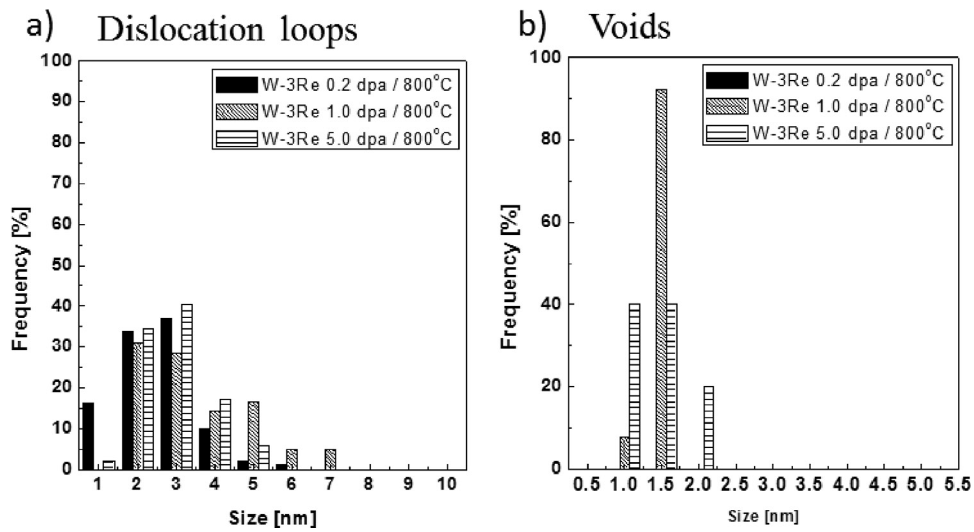

Fig. 5. Size distribution of (a) dislocation loops and (b) voids in W–3%*Re* after irradiation at 800 °C.

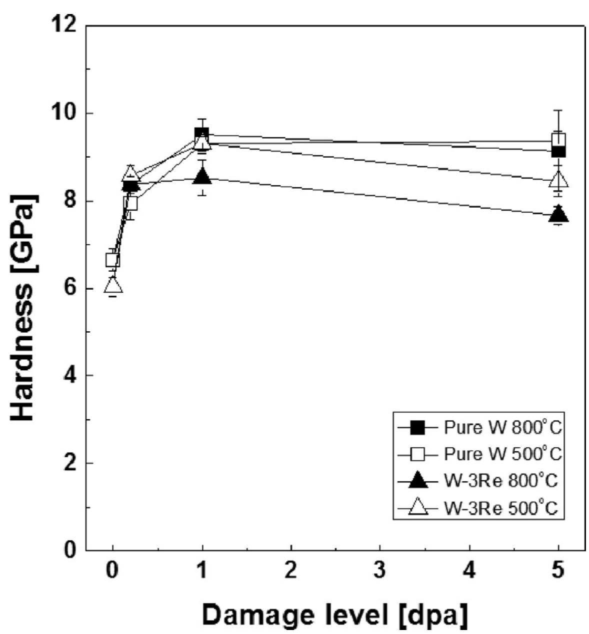

Fig. 6. Relationship between damage level and hardness.

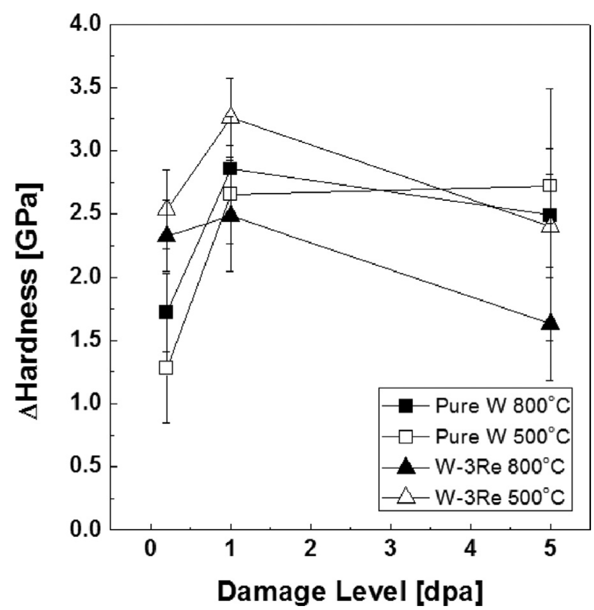

Fig. 7. Relationship between damage level and magnitude of hardness.

and 0.2741 nm, respectively [13, 14]. The values of α_l and α_v are 0.2 for the dislocation loops and 0.6 for the voids, respectively [10, 15].

In addition, the following equations were used to calculate the values of the nanoindentation hardness change $\Delta H_{cal.}$ using the values for the yield stress change calculated using Eq. 1. Eqs. 2 and 3 show the relation between the changes in yield stress $\Delta\sigma_y$ and Vickers hardness H_V and between Vickers hardness H_V and nanoindentation hardness H_I, respectively. The value of 3.06 in Eq. 2 is the correlation value between the yield stress change and Vickers hardness change [16]. The values of 0.8 and 9.8 in Eq. 3 are the correlation value between the nanoindentation hardness and Vickers hardness which obtained from experimental results and the value used to convert the units from kilogram-force to Newtons.

$$\Delta\sigma_y = 3.06\Delta H_v\sqrt{Nd} \tag{2}$$

$$H_v = (0.8/9.8) \times 10^3 \times H_I \tag{3}$$

$$\Delta H_{cal.} = 4.0 \times 10^{-3}\Delta\sigma_y \tag{4}$$

The result of the calculated and measured hardening for the pure W showed almost the same value. However, the measured hardening of the W – 3%*Re* was smaller than the calculated hardness. This might have been caused by the size of the voids. Voids and dislocation loops were the main obstacles to the dislocation motion. However, the size of the voids in this work was too small to prevent dislocation motion [17].

5. Summary

In order to investigate the effect of self-ion irradiation on the hardening and microstructure of pure W and W–3%*Re*, microstructure observations and nanoindentation tests were carried out on self-ion irradiated specimens up to 5.0 dpa. The following results were obtained:

1. In the case of pure W irradiated at 800 °C, the sizes of the observed dislocation loops and voids increased with an increase in the damage level.
2. The hardness of pure W increased with an increase in the damage level to 1.0 dpa at 800 °C. The hardness then remained at almost the same level with a further increase in the damage level.
3. The saturation of the irradiation hardening in pure W was also observed after irradiation to 1.0 dpa at 500 °C.
4. The size distribution of the dislocation loops in the W–3%*Re* at 800 °C showed the same tendency as the pure W, but no voids were observed in the specimen irradiated to 0.2 dpa.
5. W–3%*Re* samples showed a decrease in the irradiation hardening after irradiation to 5.0 dpa at 500 and 800 °C.
6. Decrease of irradiation hardening in W–3%*Re* irradiated to 5.0 dpa at 500 and 800 °C might be caused by decreasing of defect cluster by mutual recombination of vacancy and SIA trapped by the W–*Re* dumbbells or by the recovery of the dislocations introduced by the material fabrication process before irradiation.

Acknowledgements

Ion irradiation was carried out for this study at TIARA in the Takasaki Advanced Radiation Research Institute of the Japan Atomic Energy Agency (JAEA). Post–irradiation experiments were conducted at Kyoto University, IMR in Oarai and Tohoku University. The authors thank the staff of TIARA, JAEA in Rokkasho, Kyoto University, and Tohoku University. This work was supported by JSPS KAKENHI Grant Number JP24246151.

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