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Effects of transmutation elements on the microstructural evolution and electrical resistivity of neutron-irradiated tungsten

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A B S T R A C T

During fusion reactor operation, transmutation elements such as rhenium (Re) and osmium (Os) are produced from tungsten (W) upon neutron irradiation. Thus, the pure W becomes W–Re or W–Re–Os alloys and its physical properties gradually change. The irradiation hardening, microstructural changes, and physical properties of these transmutation elements of W are here investigated. Tungsten-based model alloys are fabricated and neutron irradiation is performed in the JOYO fast test reactor. The irradiation dose and temperature are 0.17–1.54 dpa and 400–750 °C, respectively. Vickers hardness measurements, microstructural observations, and electrical resistivity measurements are subsequently performed. The effects of the microstructural evolution on the irradiation hardening and electrical resistivity are discussed.

1. Introduction

Tungsten (W) has a high melting point, high thermal conductivity, and high sputtering resistance. Thus, W is a good candidate material for plasma components. During fusion reactor operation, as a result of the high-energy neutron exposure, solid transmutation elements, such as rhenium (Re) and osmium (Os), will be produced and accumulate within W. As the neutron fluence increases, W will become W–Re or W–Re–Os alloys; for example, calculations predict that pure W will become the W–18Re–3Os alloy after 50 dpa of irradiation [1].

The thermal conductivity and diffusivity decrease to half those of pure W by addition of only 5% Re [2,3]. Furthermore, a drift in the temperature reading of a W–Re thermocouple will occur due to precipitation and compositional changes during irradiation [4]. Therefore, it is important to study the effects of compositional changes due to transmutation on the physical properties of W upon neutron irradiation.

In this work, the effects of displacement damage and compositional changes due to transmutation were investigated by fabricating and irradiating W model alloys. A difference in the irradiation hardening of W–Re and W–Os alloys was found [5]. Although the hardening of W–Re alloys is small and depends on the irradiation dose, that of the W–Os alloys is substantial and independent of the dose. This paper reports the results of microstructural observations and electrical resistivity measurements on these materials, and the relation between the irradiation hardening and the microstructure.

2. Experimental procedure

Tungsten-based model alloys incorporating Re and Os were fabricated to simulate compositional changes due to transmutation by neutron irradiation of up to 50 dpa. The raw materials were pure W (99.96%) and W–26Re (W: 74.0 ± 0.2%, Re: 26.0 ± 0.2%) rods supplied by Plansee and Os (99.9%) powder supplied by Kojundo Chemical Laboratory. Fabrication of the model alloys was performed using an argon arc furnace. Nominal compositions of the alloys are given in Table 1. Interstitial impurity levels of the fabricated alloys were in the range of 40–200 wppm for carbon (C), 20–40 wppm for oxygen (O), and <12 wppm for nitrogen (N). The samples were disk-shaped with a diameter of 3 mm and a thickness of 0.2 mm and they were annealed at 1400 °C for 1 h in vacuum before irradiation.

The irradiation test was performed in the JOYO fast test reactor at JAEA (Japanese Atomic Energy Agency) under the conditions listed in Table 2. Displacement damage (dpa) was calculated using the NPRIM-1.3 code [6] with 90 eV displacement threshold energy [7]. Greenwood and Garner [8] calculated the compositional changes due to transmutation during irradiation in a fast reactor. After irradiation, Vickers hardness measurements, microstructural observations, and electrical resistivity measurements were performed.

Microstructural observations were carried out using a JEM-2010 transmission electron microscope (TEM) at 200 kV. Thin foil

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Table 1
Nominal compositions of alloys fabricated in this work (mass%).

W	Re	Os
100	–	–
95	5	–
90	10	–
74	26	–
97	–	3
95	–	5

Table 2
Irradiation conditions in JOYO.

Irradiation temperature(°C)	Fluence (En > 0.1 MeV) (10^{25} n/m^2)	dpa
400	1.3	0.17
500	2.9	0.37
740	3.1	0.40
750	12	1.54

specimens were prepared using a twin-jet polishing machine with an electrolyte of 1% NaOH in water at room temperature.

Electrical resistivity measurements were carried out by the 4-probe method at room temperature. The measurement was performed with 100 mA load current I, 0.635 mm space between probes S, and 0.98 N load P. The electrical resistivity was calculated as

$$\rho = Vt/I \times \text{C.F.} \quad (1)$$

where V was the measured voltage drop, t was the specimen thickness, and C.F. is a conversion factor that depends on the sample shape and measurement conditions [9]. The C.F. was 3.5242 in this work.

3. Results and discussion

3.1. Microstructure

Fig. 1(a) and (b) show microstructures of irradiated W. A void lattice was formed on the {1 1 0} plane upon 1.54 dpa irradiation at 750 °C. This dose level to form a void lattice is smaller than that previously reported (10 dpa) [10]. The mean size and spacing of the voids were 4.7 nm and 15 nm, respectively. The resulting expansion calculated from this number density and size was 0.48%. The size of the observed dislocation loops was less than 20 nm and the number density was less than a twentieth of that of the voids. In W irradiated at less than 0.40 dpa, smaller (2–3 nm) voids were formed at random locations in the matrix and the number density was same as for W irradiated at 1.54 dpa, except that localized black dots were also observed.

Fig. 1(c) shows the microstructure of W–26Re irradiated at 0.40 dpa at 740 °C. Needle- or plate-like precipitates were observed in irradiated W–10Re and W–26Re alloys. The diffraction pattern confirmed that precipitates aligned symmetrically along the {1 1 0} plane. In addition, spherical precipitates were observed in W–26Re. In the case of irradiation at 0.40 dpa at 740 °C, the needle-like precipitates grew and their number density increased, although the density of spherical precipitates decreased in W–26Re. The needle-like precipitates appeared to be χ phase (Re_3W) [4]. They must have been formed by the irradiation because the χ phase cannot be formed in W–26Re at thermal equilibrium. The volume fraction of the needle-like precipitates in W–26Re irradiated at 0.40 dpa at 740 °C was 4–5%. The volume fraction of the spherical precipitates was less than 0.1%.

Fig. 1(d) depicts the microstructure of W–3Os irradiated at 1.54 dpa at 750 °C. Needle-like precipitates were again observed in W–3Os irradiated at 1.54 dpa at 750 °C and aligned symmetrically along the {1 1 0} plane. The precipitates might be σ phase (WOs) and their volume fraction was up to 0.8%.

3.2. Electrical resistivity

Fig. 2(a) and (b) graph the change in electrical resistivity of W–Re and W–Os alloys due to irradiation. The resistivity increase of pure W upon irradiation was about 20%, much smaller than for the alloys, and it lacked a clear dependence on the fluence.

The electrical resistivity of unirradiated W–Re and W–Os alloys increased linearly with Re or Os content up to 10% before saturating. To model the effects of impurities, the electrical resistivity can be described by

$$\rho = \rho_0 + \Delta\rho \quad (2)$$

with

$$\Delta\rho = A(C/100)(1 - C/100) \quad (3)$$

where σ_0 is the resistivity of the pure matrix, C is the impurity atom content (at.%), and A is a factor that depends on the matrix and on the impurity. Because the atomic masses of Re and Os are nearly equal to that of W,Eq. (2) is equivalent to

$$\rho = \rho_0 + A(x/100)(1 - x/100) \quad (4)$$

where x is the impurity mass content (mass%).

For pure W at room temperature, σ_0 = 6.01 μΩ cm, while the Re factor A_{Re} and Os factor A_{Os} were 145 and 640, respectively. Although the resistivity is also affected by phonons (which depends on the temperature), the measured changes were only due to the added impurities because the sample temperature was held approximately constant.

The resistivity changes of the W–Re alloys due to irradiation were small and did not depend on the irradiation conditions. However, the resistivity of W–26Re increased by irradiation of 1.54 dpa at 750 °C, and the observed precipitate fraction in W–26Re was up to 5% after irradiation. The resistivity of a multi-phase alloy is

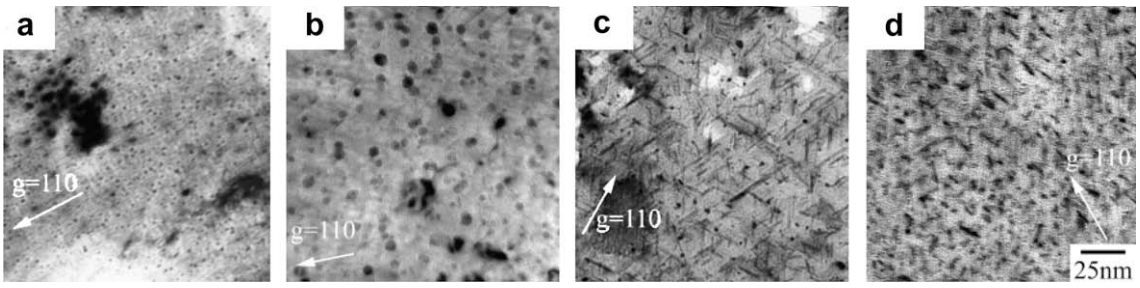

Fig. 1. TEM micrographs of (a) pure W irradiated at 0.17 dpa at 400 °C, (b) pure W irradiated at 1.54 dpa at 750 °C, (c) W–26Re irradiated at 0.40 dpa at 740 °C, and (d) W–3Os irradiated at 1.54 dpa at 750 °C.

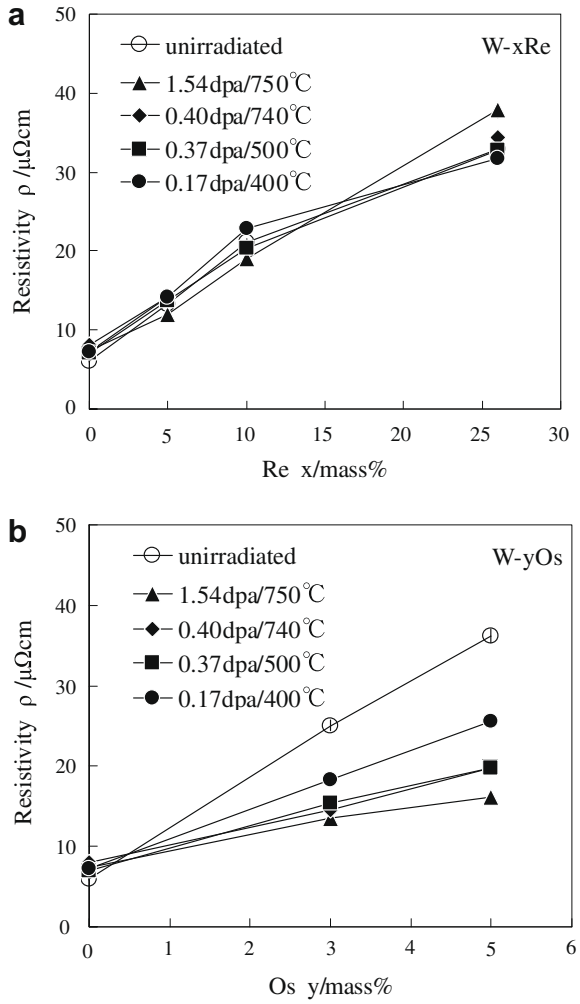

Fig. 2. Electrical resistivity changes of (a) W–Re and (b) W–Os alloys after irradiation. Dashed line means supposition of linear increase related to Re content.

calculated using the complex rule for the volume fraction of each phase,

$$\rho = \Sigma \rho_i v_i \quad (5)$$

where i is the phase and v is the volume fraction. Thus, as a result of the increase in the precipitate fraction, the electrical resistivity of the precipitates as a second phase can alter the bulk resistivity.

The resistivities of the W–Os alloys decreased greatly upon irradiation. Further, the electrical resistivity increased linearly with Os content both before and after irradiation. The resistivity reduction therefore depends on the Os content. This behavior is strikingly different from that of pure W and of the W–Re alloys. The resistivity reduction increased with the dose, but there was no significant change due to the irradiation temperature difference at 0.40 dpa. The precipitate has negligible effect because the observed precipitate fraction of W–3Os after irradiation was only 0.8%. The solute Os content changes due to irradiation were estimated, based on the measured resistivity changes, as listed in Table 3.

Table 3
Estimated Os solute content change due to irradiation (mass%).

Os content before irradiation	Os solute content after irradiation estimated by electrical resistivity			
	1.54 dpa 750 °C	0.40 dpa 740 °C	0.37 dpa 500 °C	0.17 dpa 400 °C
3	1.2	1.4	1.5	2.0
5	1.6	2.2	2.2	3.2

3.3. Irradiation hardening and precipitation

Irradiation hardening of the alloys examined in this study has been previously reported [5]. The difference in the number density and size of the voids in pure W due to irradiation is significant, although the electrical resistivity change is much smaller.

In the case of W–Re alloys, as the fluence increases, the irradiation hardening increases due to precipitation. As a result, the solute Re content in the matrix decreases. However, the resistivity of W–26Re was increased by irradiation. Thus, the effect on the bulk resistivity of precipitation of intermetallic Re_3W is larger than that of solute Re in the matrix.

The solute Os content in the matrix of W–Os alloys decreases due to irradiation-induced precipitation. However, precipitated Os (estimated by microstructural observation) was as much as 20% of the added Os, although it was much less than that estimated from the resistivity. The solute Os content level and amount of precipitates cannot explain the large irradiation hardening by Os. Thus, there must be some other hardening mechanism related to Os.

4. Conclusion

The effects of solid transmutation elements were investigated by hardness tests, electrical resistivity measurements, and microstructural observations. The experimental results can be summarized as follows:

(1) The threshold level for displacement damage of the void lattice due to neutron irradiation was between 0.40 and 1.54 dpa at 750 °C for pure W. Although void formation induced irradiation hardening, it did not alter the electrical resistivity significantly.
(2) The electrical resistivity of unirradiated alloys increased linearly with Re or Os content up to 10% and then became saturated. The effect of Os on electrical resistivity was 5 times larger than that of Re.
(3) Needle-like precipitates were induced in W–Re alloys during irradiation. However, the amount of precipitated Re was at most 10% of what was added, and the electrical resistivity change was small. With increased precipitation, the electrical resistivity of the precipitates would probably alter the bulk resistivity.
(4) Needle-like precipitates were induced in W–Os alloys during irradiation and the electrical resistivity decreased greatly. The resistivity reduction tended to increase as the dose and Os content increased. There must be other mechanism of irradiation hardening for W–Os alloys than solution or precipitation hardening.

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