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Effects of Transmutation Elements on Neutron Irradiation Hardening of Tungsten

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Tungsten (W) is a candidate material for Plasma facing materials of fusion reactors. During fusion reactor operation, not only irradiation damages but also transmutation elements such as rhenium (Re) and osmium (Os) are produced by neutron irradiation. As a result, the original pure tungsten changes to W-Re or W-Re-Os alloys. Thus, the mechanical and physical properties are expected to change. The aim of this study is to investigate the effects of transmutation elements on neutron irradiation hardening and microstructure changes of tungsten.

To simulate the effects of transmutation elements, tungsten base model alloys were used in this study. The examined compositions of the alloys were selected from the calculated changes in solid solution area of W-Re-Os alloy. Neutron irradiation was performed in fast test reactor JOYO in JAEA. The irradiation damages and temperature ranges were 0.17–1.54 dpa and 400–750°C respectively. After the irradiation, Vickers hardness test and TEM observation were performed.

There were clear differences between Re and Os in effects on irradiation hardening. In the case of W-Re alloys, when damages were less than 0.40 dpa, the irradiation hardenings were nearly equal to those of pure tungsten independent of Re addition. But when the damage was 1.54 dpa, the irradiation hardenings increased lineally with Re content. Microstructural observations showed that precipitations mainly formed in W-Re alloys. In the case of W-Os alloys, the irradiation hardenings (ΔHv) of W-3Os alloys were larger than those of pure tungsten. And the differences were about 400 independent of dpa and irradiation temperature. Effects of Re and Os on irradiation hardening based on the microstructural observations were discussed. [doi:10.2320/matertrans.MAW200722]

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1. Introduction

Tungsten (W) is a high Z element, and has high melting point, high thermal conductivity, and high sputtering resistance. Thus, tungsten is a candidate material for divertors that are plasma facing components under especially high heat and high particle loads condition in fusion reactors. During fusion reactor operation, due to 14 MeV neutron irradiation, not only irradiation damages but also solid transmutation elements, such as rhenium (Re) and osmium (Os), accumulate in W. Consequently, as neutron fluence increases, original pure W changes to W-Re or W-Re-Os alloys. For example, it is expected that pure W will changes to W-18Re-3Os alloy after 50 dpa irradiation that corresponds to neutron fluence of 10 MWy/m^2 by calculations.[1)]

It is well known that W shows brittle behavior at room temperature though has good high temperature properties. The low temperature embrittlement was improved by Re addition,[2,3)] but the thermal conductivity and diffusivity decrease to about half of pure W by addition of 5 mass%Re.[4,5)] Furthermore irradiation induced precipitations including σ and χ phase were produced and large irradiation hardening was induced in W-26Re alloy by heavy neutron irradiation.[6)] Though Os is a main transmutation element from W and Re and affects properties of W alloys, there are few reports concerning irradiation to W-Os alloys. In order to understand irradiation property changes of W under the fusion reactor condition, systematic investigations on alloy compositions and irradiation doses are necessary.

The aim of this study is fundamental investigations on effects of composition changes on irradiation effects. Binary and ternary W model alloys including Re and Os were fabricated, and neutron irradiations were carried out to investigate synergistic effects of composition changes and irradiation effects. Based on the irradiation experiments, effects of solid transmutation elements, Re and Os, on neutron irradiation hardening and microstructure evolution are discussed.

2. Experimental Procedure

In order to investigate effects of transmutation elements, W base model alloys simulated compositions resulting from transmutation were fabricated in this study. Nominal compositions of examined alloys are shown in Table 1. They were selected along the calculated composition changes in solid solution area of W-Re-Os alloy.[1)] The model alloy fabrication was carried out using an argon arc furnace in Institute of Material Research of Tohoku University. The weight of each alloy ingot was about 20 g. W (99.96%) and

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

W	Re	Os
100	—	—
95	5	—
90	10	—
74	26	—
97	—	3
95	—	5
92	5	3
87	10	3
79	18	3
72	25	3
90	5	5

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Table 2 Neutron irradiation conditions (JOYO, MNTR-01).

Capsule	Irrad. Temp. (°C)	Fluence (10^{25} n/m^2) Fast(En > 0.1 MeV)	dpa
JNC-54	750	12	1.54
JNC-61	740	3.1	0.40
JNC-50	500	2.9	0.37
JNC-60	400	1.3	0.17

MNTR-01: The irradiation rig used in this work.

W-26Re (W: 74.0 ± 0.2%, Re: 26.0 ± 0.2%) rods supplied by Plansee Ltd. and Os powder (99.9%) were used as raw materials to obtain the compositions. After setting the raw materials, 3 times gas substitutions were practiced before melting for purity of atmosphere. Interstitial impurity levels of the fabricated alloys were in the range of C: 40∼200 ppm, O: 20∼40 ppm and N: <12 ppm. Irradiation specimens with a diameter of 3 mm and a thickness of 0.3 mm were cut out from the ingots by spark wire method and mechanically grinded and polished to 0.2 mm thickness, and annealed at 1400°C for 1 hour in vacuum ($<10^{-5}$ Pa).[7]

The irradiations were carried out in fast test reactor JOYO in JAEA. Irradiation conditions are shown in Table 2 (fluences of fast neutron (En > 0.1 Mev) are shown). Displacement damages (dpa) were calculated by NPRIM-1.3 code[8] using 90 eV as the displacement threshold energy.[9] With these fluences, composition changes during irradiation can be almost disregarded because the cross section of transmutation of W with the neutron spectrum in JOYO is small. After cooling, Vickers hardness (Hv) test and microstructure observation were carried out.

Vickers hardness tests were carried out on a MM-2 micro-hardness tester with load of 200 gf and loading time of 30 s. In this study, irradiation hardening (ΔHv) is defined as the difference of Hv number between before and after irradiation.

Thin foil specimens for microstructure observations using the transmission electron microscopy (TEM) were prepared using a twin-jet polishing machine with an electrolyte of 1 mass% NaOH in water. The observations were carried out on a JEM-2010 operating at 200 kV.

3. Result

3.1 Irradiation hardening

Figure 1 shows results of irradiation hardenings of W-Re alloys as a function of Re content and irradiation conditions. When the irradiation level is less than 0.40 dpa, the values were nearly equal to pure W independently Re addition. Only hardening of W-26Re which was irradiated to 0.40 dpa at 740°C was larger than others. In the case of 1.54 dpa, hardenings increased linearly with Re content.

Figure 2 shows results of irradiation hardenings of W-Os alloys as a function of Os content and irradiation conditions. Irradiation hardenings of W-3Os were about 400 larger than those of pure W independently on dpa. Remarkable difference was not observed between Os content of 3% and 5%.

Figure 3 shows results of irradiation hardenings of W-xRe-3Os alloys as a function of Re content and irradiation conditions. In the case of 1.54 dpa at 750°C, there was not clear correlation between Re contents and irradiation hardenings. But when irradiation temperature and dpa was lower, hardening tended to decrease with Re content.

Figure 4 shows results of irradiation hardenings of W-5Re-xOs alloys as a function of Re content and irradiation

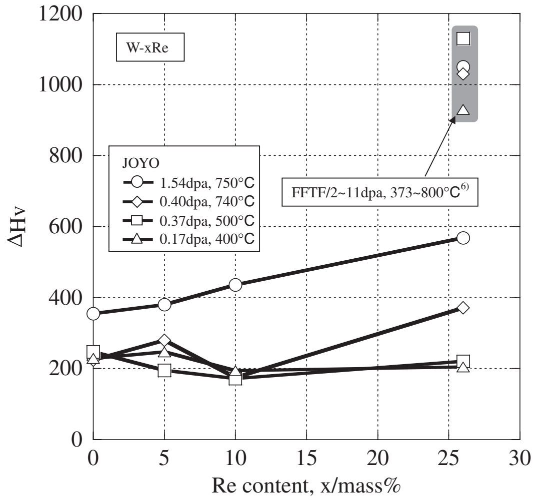

Fig. 1 Hardness test results of W-*x*Re alloys as function of Re content and dpa.

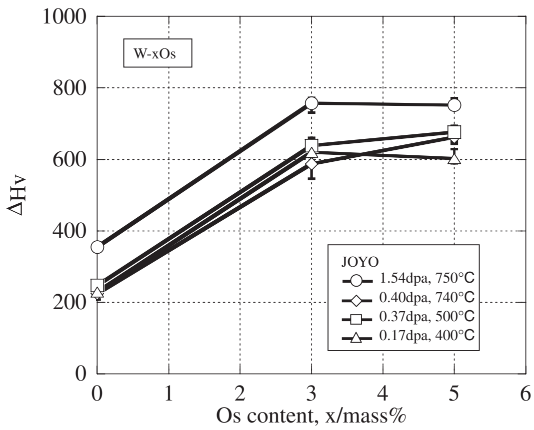

Fig. 2 Hardness test results of W-*x*Os alloys as function of Os content and dpa.

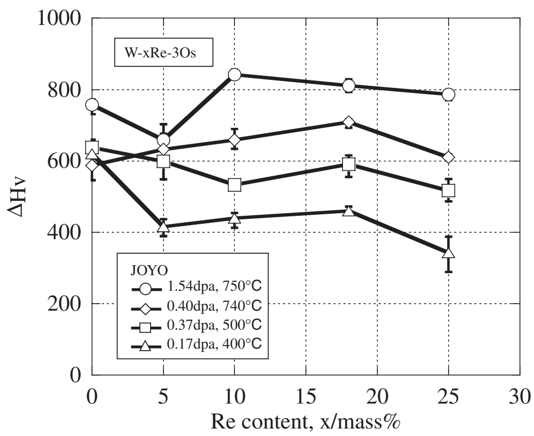

Fig. 3 Hardness test results of W-*x*Re-3Os alloys as function of Re content and dpa.

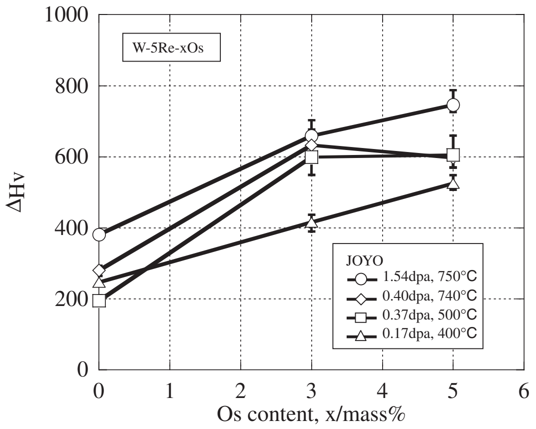

Fig. 4 Hardness test results of W-5Re-*x*Os alloys as function of Os content and dpa.

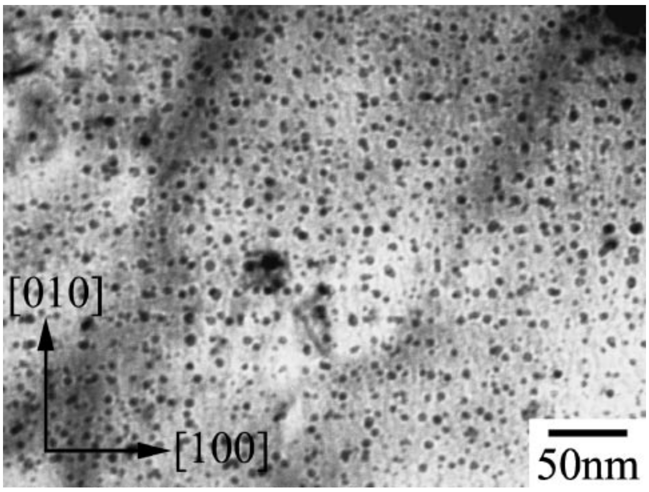

Fig. 5 TEM image of pure W irradiated 1.54 dpa at 750°C. Void lattice in matrix.

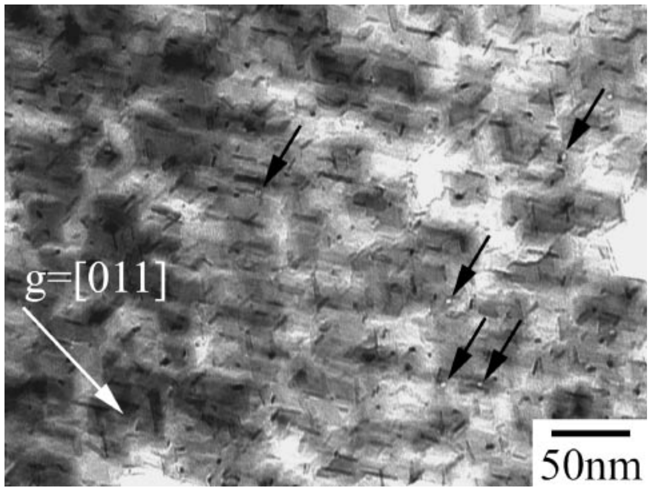

Fig. 6 TEM image of W-10Re irradiated 1.54 dpa at 750°C. A number of precipitations in matrix. Black allows mark voids in matrix.

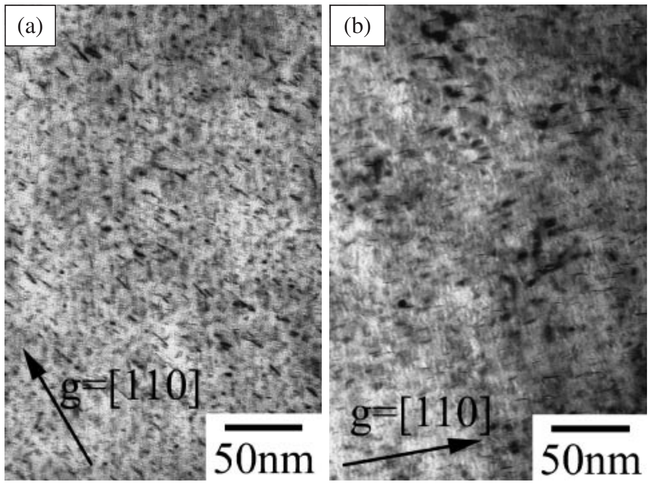

Fig. 7 TEM images of W-3Os irradiated 1.54 dpa at 750°C. (a) Needle like precipitations ($Z = [111]$, 7 g) and (b) black dots ($Z = [111]$, 1 g) in matrix.

conditions. The behaviors were similar to W-Os binary alloys. Furthermore hardenings of W-5Re-3Os and/or W-5Re-5Os were about 400 larger than W-5Re without Os. But in the case of 0.17 dpa at 400°C, the difference was only about 200 between hardening of W-5Re-5Os and W-5Re.

3.2 Microstructural observation

Figure 5, 6 and 7 are TEM images of pure W, W-10Re and W-3Os respectively, irradiated to 1.54 dpa at 750°C. Void lattice was formed in pure W. The lattice tended to align with direction [100]. Void diameter and lattice spacing were about 5 nm and 20 nm respectively. Small dislocation loops and other structure except for voids were found scarcely. Although void lattice was formed by irradiation to over 10 dpa according to previous papers,[10,11] the lattice was formed with smaller dpa in present experiments.

Precipitations were mainly observed in W-10Re. Precipitations are seemed needle like. These could not be identified from diffraction pattern because the size and density of them were small. Small dislocation loops were found scarcely, and void density was much smaller than that of pure W.

Needle like precipitations and black dots were mainly observed in W-3Os. But precipitations could not been identified because there is not spot of precipitations on diffraction pattern. Black dots may be interstitial defect clusters or dislocation loops. Voids were scarcely found in this irradiation conditions.

4. Discussion

4.1 Binary alloys

Irradiation hardening (ΔHv) of pure W irradiated to 1.54 dpa was 100 larger than that of irradiated up to 0.40 dpa without remarkable dependence on dpa. Since void lattice was formed in W irradiated to 1.54 dpa, it is supposed that irradiation hardening due to voids appeared over 0.40 dpa and under 1.54 dpa, hardening over 1.54 dpa saturate due to void lattice formation.

Irradiation hardening can be suppressed by Re addition up to 0.40 dpa. However W-26Re alloys show significant hardening and embrittlement after irradiation to 2∼11 dpa because of irradiation induced precipitation of σ (ReW) or χ (Re_3W) phases.[6] The results of this work show that irradiation hardenings of W-Re alloys increase linearly with increasing of Re content, and hardening rate depends on dpa when irradiation damage and temperature is over 1.54 dpa and 750°C respectively. Thus the effect of precipitations on irradiation hardening increases in proportion with Re content. Comparing results of W-26Re irradiated to 0.40 dpa or less, irradiation hardening of only irradiated to 0.40 dpa at 740°C was larger than others. It is expected that precipitation forming is enhanced and irradiation hardening occurs when irradiation temperature is high.

In W-3Os irradiated 1.54 dpa at 750°C, there were a number of precipitations and black dots but voids were observed scarcely. Thus it is considered that void suppression effect of Os is stronger than that of Re, and Os can enhance to form black dots such as interstitial defect cluster. Because there is not large difference between irradiation hardening with Os content of 3 and 5 mass% independently on dpa and irradiation temperature, it is considered that enhanced irradiation hardening compared to pure W started at Os content less than 3 mass%. The mechanism of irradiation hardening of W-Os alloys is considered to be formation of black dots and dislocation loops. It is also considered that interaction between the loops and precipitations enhanced the hardening.

The results of this work were shown that under size elements, Re and Os in a bcc metal W, suppress formation of voids. In the case of vanadium alloys, under size elements such as iron, silicon and chromium enhanced void swelling of vanadium.[12] To explain the difference, microstructural observations of other specimens are necessary.

4.2 Ternary alloys

The present results of irradiation hardening of W-xRe-3Os and W-5Re-xOs alloys demonstrate that Re suppresses irradiation hardening due to Os when irradiated to less than 0.17 dpa at 400°C or below. It is considered that the magnitude of suppression effect does not clearly depend on Re content when Os content is less than 3 mass% and Re content is over 5 mass%. There would be little suppression effect of Re on the hardening when Os content is 5 mass%.

During fusion reactor operation, large irradiation hardening due to Os may be occur when the Os content in W reaches about 3% by transmutation. Consequently irradiation hardening rate may increase with dpa.

5. Conclusion

In order to investigate effects of solid transmutation elements on neutron irradiation hardening of W, fabrications of W model alloys simulated composition changes by transmutation in fusion reactors and neutron irradiation from 0.17 dpa to 1.54 dpa at 400∼750°C were carried out. Followings were obtained from hardness tests and microstructural observations.

(1) Irradiation hardening of pure W was observed with the irradiation conditions. Formation of void lattice by irradiation over 1.54 dpa at 750°C was observed.
(2) For W-Re binary alloys, irradiation hardening was obtained but the hardening did not clearly depend on irradiation temperature on neutron irradiation. Hardening due to irradiation induced precipitation occurs when alloys are irradiated to over 1.54 dpa at 750°C. This hardening linearly increases with Re content.
(3) For W-Os binary alloys, irradiation hardening (ΔHv) of W-3Os is 400 larger than pure W independently on dpa.
(4) For W-Re-Os ternary alloys irradiated to 0.17 dpa at 400°C, Re would suppress irradiation hardening due to Os. However it is considered that Os dominantly affects irradiation hardening independently on Re content when irradiation temperature and damage are over 740°C and 0.40 dpa respectively.

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REFERENCES

1) T. Noda, M. Fujita and M. Okada: J. Nucl. Mater. **258–263** (1998) 934–939.
2) P. Makarov and K. Povarova: J. Refractory Metal & Hard Materials **20** (2002) 277–285.
3) H. P. Gao and R. H. Zee: J. Mater. Sci. Lett. **20** (2001) 885–887.
4) M. Fujitsuka, B. Tsuchiya, I. Mutou, T. Tanabe and T. shikama: J. Nucl. Mater. **283–287** (2000) 1148–1151.
5) T. Tanabe, C. Eamchotchawalit, C. Busabok, S. Taweethavorn, M. Fujitsuka and T. Shikama: Mater. Lett. **57** (2003) 2950–2953.
6) Y. Nemoto, A. Hasegawa, M. Satou and K. Abe: J. Nucl. Mater. **283–287** (2000) 1144–1147.
7) J. C. He, A. Hasegawa, M. Fujiwara, M. Satou, T. Shishido and K. Abe: Mater. Trans. **45** No. 8 (2004) 2657–2660.
8) S. Shimakawa, N. Sekimura and N. Nojiri: Proc. the 2002 Sympo. on Nuclear Data (2003) pp. 283–288.
9) C. H. M. Broeders and A. Yu. Konobeyev: J. Nucl. Mater. **328** (2004) 197–214.
10) K. Krishan: Radiat. Eff. **66** (1982) 121–155.
11) V. K. Sikka and J. Moteff: J. Appl. Phys. **43** (1972) 4942.
12) H. Nakajima, S. Yoshida, Y. Kohno and H. Matsui: J. Nucl. mater. **191–194** (1992) 952–955.