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Neutron irradiation effects on tungsten materials

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ABSTRACT

To understand the irradiation response of tungsten in the ITER and a DEMO-like reactor, irradiation effects on microstructure development, hardening and electrical resistivity of pure W and W-Re-Os alloys are studied using fission reactor irradiation up to about 1–1.5 dpa in the temperature range from 400 to 800 °C. Microstructural development and hardening behavior are summarized here based on the irradiation data. Voids are the major damage structure in pure W, but Re addition clearly suppressed void formation. Irradiation hardening was also suppressed in lower Re content alloys. The hardening was caused by irradiation-induced precipitation of WRe (σ-phase) and WRe_3 (χ-phase), and the hardening behavior strongly depended on the neutron spectrum of the irradiation field. On the basis of these results, a damage structure development prediction map in fusion devices is suggested.

1. Introduction

Tungsten (W) has a high melting point (3380 °C) and high sputtering resistance to energetic particles, hence it is considered to be a candidate for the plasma-facing component (PFC) materials in magnetic confinement fusion reactors such as the first wall of a blanket and a diverter plate [1–3]. During fusion reactor operation, as a result of high-energy neutron exposure, in addition to displacement damage, transmutation elements are also produced in the irradiated materials. Rhenium (Re) is one of the major solid transmutation elements formed from W under fusion reactor conditions. The predicted amounts of displacement damage and Re content that would be produced in W in the ITER and in a DEMO-like fusion power reactor were reported by Bolt et al. [1]. After 5 years of operation in the DEMO-like reactor, the average neutron fluence would be 10 MWa/m^2 at the first wall and 5 MWa/m^2 at the diverter, the displacement damage and Re content would be 30 dpa and 6% in W of the first wall, respectively, and 15 dpa and 3% in the diverter, respectively. Operating temperatures are expected to be 500–800 °C at the first wall, and 600–1300 °C at the diverter, depending on the cooling system used. Other transmuted elements such as helium (He) and hydrogen (H) are also formed; the calculated concentrations of He and H in W after 10 MWa/m^2 neutron irradiation are approximately 30 appm and 60 appm, respectively [4]. In the ITER, a displacement damage of 0.7 dpa and transmutation of 0.15% Re would be produced in the W diverter after a neutron fluence of 0.15 MWa/m^2 at 200–1000 °C [1]; therefore, transmutation may not be the main concern for W in the ITER.

From the 1960s to the 1980s, irradiation behavior of W was investigated using the Experimental Breeder Reactor-II (EBR-II) in the USA to develop space reactor materials. Heavy irradiations up to several dpa at higher temperatures were carried out, but the results were published only in a limited number of papers [5–11]. These data have not been summarized systematically to predict the irradiation behavior of W, because the examined materials were fabricated separately by each researcher individually. We have been studying the irradiation behavior of W using systematically designed W alloys and the same source pure W using several fission reactors to clarify the effects of the irradiation field [12–22].

There is currently no intense fusion neutron source to simulate fusion reactor conditions; therefore, in order to predict irradiation behavior of W in fusion reactor conditions, modeling and accelerator irradiation experiments are required. Systematically summarized real neutron data including irradiation temperature, fluence dependence and impurity effects are indispensable to verify the results of modeling and simulation. The objectives of this work were to summarize and predict neutron irradiation behavior of W from the viewpoint of damage structure development and the effects of transmutation elements based on our experimental studies, and to provide insights into future experimental work to develop radiation-resistant W materials.

2. Experimental

We have been using two different sources of W and W-alloys in our experiments. The first is 0.2 mm-thick hot-rolled sheets of W

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Table 1
Neutron irradiation conditions.

Reactor	Capsule ID	dpa	Irradiation temperature (°C)
Joyo	JNC-54	1.54	750
	JNC-63	0.96	538
	JNC-61	0.40	740
	JNC-50	0.37	500
	JNC-60	0.17	400
	JAEA-04	0.44	531
	JAEA-05	0.47	583
	JAEA-06	0.42	756
JMTR	98M-7U	0.13	800
	98M-7U	0.12	600
	98M-7U	0.10	600–800
	99M-13U	0.09	800
	00M-95U	0.17	800
	05M-19U	0.15	600
	05M-19U	0.22	800
HFIR	14J	0.90	500
	14J	0.98	800

and W-Re alloys supplied by Plansee Corporation. TEM disk specimens of 3 mm diameter were punched out from the as-received sheets and heat treated at 1300, 1400 or 1600 °C for 1 h in vacuum. The second set of W and W alloys were arc-melted (Arc) ingots that we fabricated in collaboration with the Institute of Materials Research (IMR), Tohoku University. The alloy series of W-xRe-yOs (x = 0, 3, 5, 10, 26; y = 0, 3, 5) was fabricated. TEM disks of 0.2 mm thickness were prepared from these ingots using electro-discharge machining, and these were annealed at 1400 °C for 1 h in vacuum before irradiation. These disk type specimens were loaded in He or He–Ar gas-filled capsules and irradiated in fission reactors. For the neutron irradiation experiments, the Japan Material Testing Reactor (JMTR) and the High Flux Isotope Reactor (HFIR in USA) which are mixed spectrum reactors containing high flux thermal neutrons, and JOYO which is a sodium cooled fast neutron experimental reactor in Japan, were used. The irradiation conditions of temperature and dpa are listed in Table 1. Displacement threshold energy of W used to calculate dpa was 90 eV. On the other hand, the transmutation level depended on the reactor. In the case of JMTR irradiation, the dpa level was less than 0.15–0.22 dpa, the amount of transmuted Re would thus be less than 1.8%. After 1 dpa irradiation in the HFIR, pure W is estimated to transform into W-9Re-5Os alloys. In the case of JOYO irradiation, the amount of Re in pure W was estimated to be about 1.5% after 1.54 dpa irradiation.

Microstructural observations were carried out using a JEM-2010 transmission electron microscope (TEM) operating at 200 kV. Thin foil specimens for the TEM observations were prepared using a twin-jet polishing machine with an electrolyte of 1 mass% NaOH in water at room temperature. Hardness tests were carried out using a micro-Vickers test machine using a 1.96 N load and a loading time of 30 s at room temperature. Electrical resistivity measurements were carried out by the four-probe method at 20 °C using the irradiated 3 mm diameter disks [14] of all the samples.

3. Results and discussions

3.1. Microstructural observation

The typical microstructures of irradiated pure W in a temperature-dpa irradiation matrix are summarized in Fig. 1, along with typical micrographs of pure W [15,17,19,22]. The major defect clusters in pure W under these irradiation conditions were voids. Dislocation loops were also observed at lower dpa levels but their number density was in the range of 1/10–1/100 compared to that of the voids. A void array structure, called the void lattice, was observed above 1 dpa at 538 and 750 °C for irradiation in the JOYO [13,21]. In the case of HFIR irradiated specimens, after 1 dpa at 500 and 800 °C, labeled [HFIR] in the micrographs, a void lattice was not observed. Small amounts of voids and highly dense needle-like precipitates were observed in the pure W. These precipitates were the σ (ReW) or χ (Re_3W) phase [17] as determined by electron diffraction analysis.

The void formation behavior around 1–1.5 dpa was strongly affected by Re addition to W. The JOYO irradiation data showed that the void size and number density tended to decrease significantly with increasing Re content. The defect types in the 5% and 10% Re alloys in this irradiation range were similar, that is a small amount of voids and fine precipitates. The void number density of the W-Re alloys compared to pure W was less than about 1/10–1/100 [19]. The size is also smaller than in pure W. These results show that even a small amount of Re (3%) can suppress void formation, but in the case of simultaneous introduction of Re with displacement damage, the suppression of void formation was not confirmed up to 1.54 dpa at 740 °C. The data show that the main damage structure can be categorized into (1) voids and (2) voids and loops area, as shown in Fig. 1.

3.2. Hardness and electrical resistivity

The irradiation hardening behavior obtained by the fission reactor irradiation as a function of Re addition before irradiation is shown in Fig. 2. The results indicate that irradiation-induced hardening of W depended on the irradiation temperature, dpa and Re composition. It was seen that irradiation hardening strongly depends on the damage microstructure as shown in Fig. 1. Details of microstructure development of W-Re are included in our previous papers [12–21]. The irradiation hardening was caused by the void and loop formation.

Fig. 2 shows irradiation condition dependence on irradiation hardening of W and W-Re alloys. The characteristic point of the hardening behavior is significant hardening of the HFIR-irradiated specimens. This was due to dense formation of precipitates in the HFIR-irradiated pure-W, as shown in Fig. 1(h). In the case of higher irradiation levels for pure-W in the JOYO, precipitates were not observed and a void lattice was observed [13]. Irradiation hardening of the higher irradiation JOYO samples showed similar hardening as indicated in Fig. 2.

The hardening of W-Re alloys was suppressed at lower dpa levels but it significantly increased after 0.4 dpa or higher irradiation [16]. The results of microstructural observation showed that fine and dense precipitates were formed in the hardened specimens and the precipitates might cause the hardening [14,17]. The results revealed that hardening of W-Re alloys was suppressed by suppression of void formation, but with increasing dpa, precipitates nucleated and grew due to the irradiation-induced precipitation, then hardening became larger.

Fig. 3 shows the electrical resistivity of the W-Re alloys before and after neutron irradiation under different conditions [14,15]. In the case of the JOYO and JMTR irradiations, no significant change in the electrical resistivity was observed in pure W and in the W-Re alloys. It is well known that an increase in the resistivity is due to an increase in the concentration of solutes before irradiation. In the case of HFIR irradiation, the increase in electrical resistivity of pure W is larger than that in the case of JOYO irradiation, suggesting a higher transmutation rate of Re in the HFIR irradiation than that in the JOYO.

Previous microstructure observation showed that irradiation induced precipitates were formed in W-Re after a certain irradiation fluence, and the precipitation behavior depended on the Re content. Even in pure W, under fusion reactor conditions, transmutation products increased in the matrix as irradiation fluence increased.

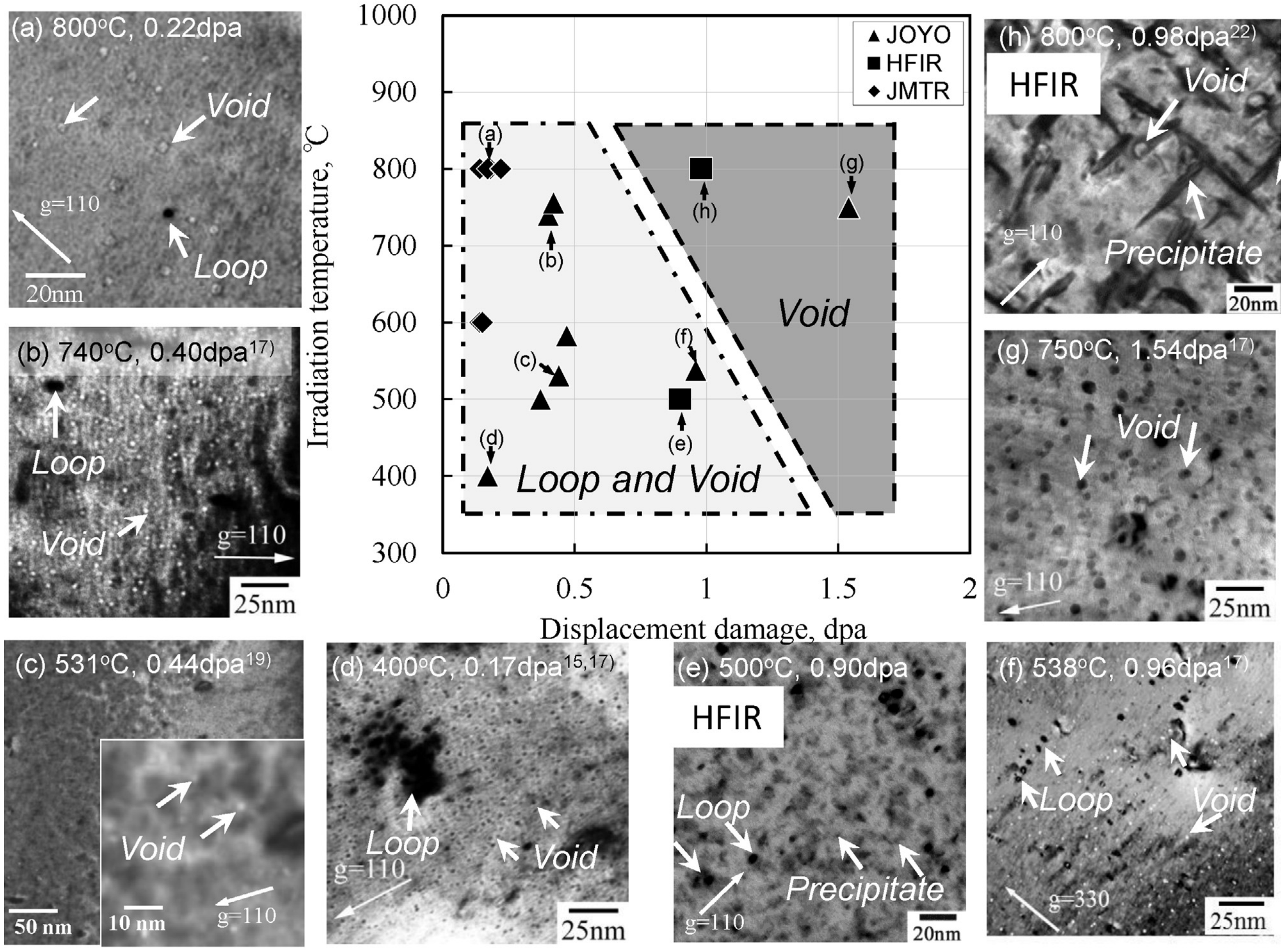

Fig. 1. Summary of irradiation conditions and typical microstructure of pure W obtained by TEM. Void images (a)–(c) and (f) were taken in an under-focus condition, (d) and (g) were taken in an over-focus condition, and (e) and (h) were taken in the focus condition.

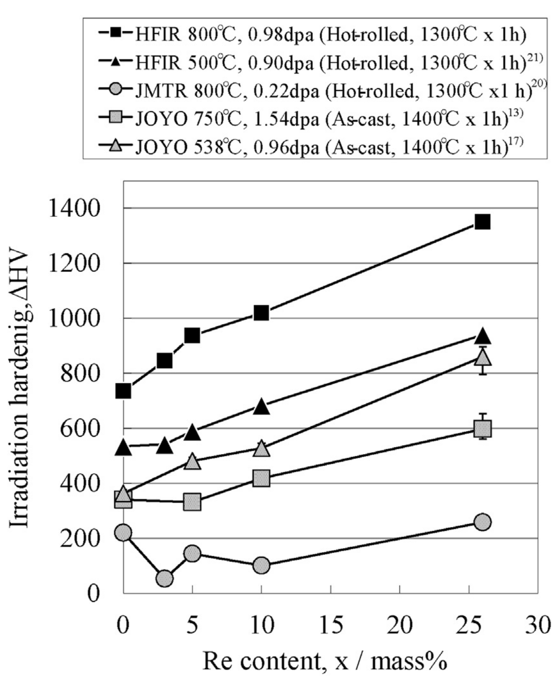

Fig. 2. Irradiation condition dependence on hardness of W and W-Re alloys.

Then the irradiation microstructure, such as voids or void lattice, should be changed because of the increase of the solute element in the matrix. This also affects material properties, such as thermal conductivity and electrical resistivity. Comparing data for the HFIR and JOYO irradiations, the transmutation effect on microstructure development was revealed in this work.

3.3. *Prediction of microstructural development of pure W under fusion reactor conditions*

In order to predict irradiation behavior of W by modeling, multi-scale modeling needs to be established, but the modeling is in an on-going state even in Fe-base alloys [23], which have been studied intensively. Many unknown parameters of W such as dose rate (dpa/s) effects, temperature dependence and impurity effects need to be clarified. In this work, as a first order approximation of irradiation behavior, microstructural development of pure W in fusion devices in the temperature range of 600–800 °C is suggested in Fig. 4 based on our neutron irradiation data for pure W and W-Re alloys. The arrows toward the top right from the lower left means as change in the Re concentration with neutron irradiation (dpa). Gray arrows indicate real composition change by neutron irradiation in our fission reactor irradiations and the black arrows indicate those in the fusion devices, ITER and DEMO-like reactors. The Re composition change of the W-Re alloys in JMTR and JOYO below 1 dpa is almost negligible in the scale of this figure, therefore, results for W-3Re and W-5Re are indicated as horizontal arrows in the figure.

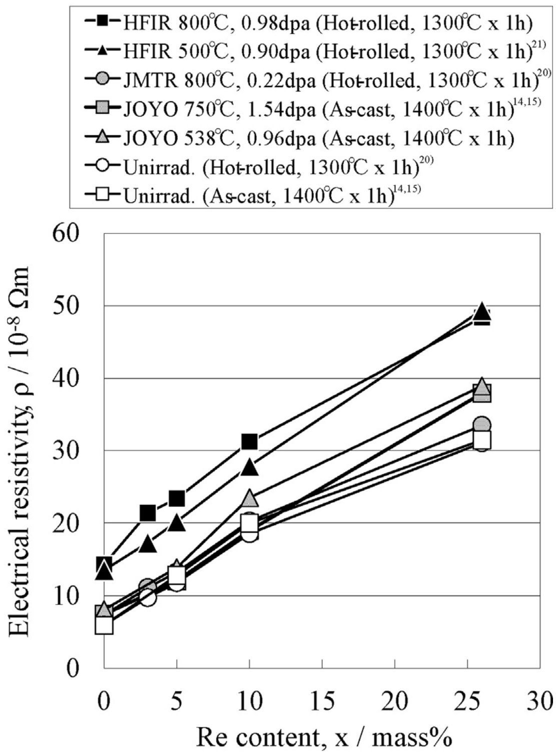

Fig. 3. Irradiation condition dependence on electrical resistivity of W and W-Re alloys.

The Re content in pure W after 0.15 dpa irradiation in the JMTR was about 1.2% and it was about 9.6% for 1 dpa irradiation in the HFIR.

Typical schematic microstructures are also shown in Fig. 4. As mentioned before, in the JOYO irradiated pure W, a void lattice formed after 1.54 dpa at 740 °C, and voids and loops were observed after 0.4 dpa at 583 °C. Coarse and large precipitates were observed after 1 dpa at 800 °C by HFIR irradiation.

Microstructure observation results of W-5Re and 10Re after the JMTR and JOYO irradiations suggested that in the early stage of the irradiation, voids and dislocation loops were formed at lower dpa levels in pure W and the W-Re alloys. When neutron irradiation proceeded with increasing Re concentration, the voids formed in the early stages of irradiation might have disappeared and precipitates appeared when the Re contents in the matrix was above ∼5%.

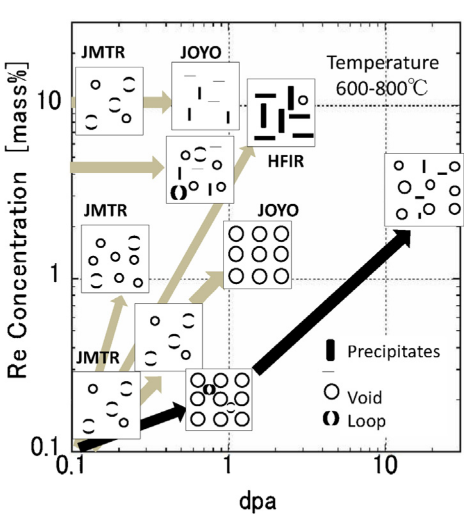

Fig. 4. Prediction of microstructural development of W under fusion conditions based on fission reactor irradiation data.

The mechanism of Re effects on void formation is been considered to be as follows [10]. Re is an under sized atom in the W matrix therefore substitutional Re atom tend to bind self-interstitial atoms (SIA) because their lattice strain field is opposite. Once the Re-SIA complex is formed, the mobility of the SIA decreases, enhancing vacancy-interstitial recombination. Therefore, void growth is suppressed with enrichment of Re, and might induce into an embryo of the χ-phase precipitation.

On the basis of these results, the microstructural changes in W by neutron irradiation can be predicted. In pure W, voids are formed at the early stage of irradiation and the void lattice structure appears at around 1 dpa at 600–800 °C. The void lattice is a well-known damage structure in many metals but the formation mechanism has not been clarified yet. Once the void lattice is formed, it is expected to be stable up to a higher dpa region [24]. The stability of the void lattice structure means the balance of formation and annihilation of voids maintained during the process.

In the case of W, with increasing neutron fluence up to above 10 dpa, the concentration of transmuted Re increases to several mass %, and the balance between point defect production and annihilation in the void lattice structure changes so that the voids might shrink and precipitates are formed in the W matrix simultaneously. The threshold concentration level of Re from in-solution to precipitation dominant level might be about several mass%. To improve accuracy of the prediction, acquiring accelerator irradiation data is effective to expand the higher dpa region and quantitative analysis using modeling is needed.

4. Summary

Using fission neutron irradiation data of W and W-Re alloys, microstructure development under irradiation up to 1.54 dpa in the temperature range of 400–800 °C was determined. The irradiation behavior of the hardness and electrical resistivity change of W are also summarized. Based on these results, qualitative prediction of microstructural development in pure W under neutron irradiation was suggested.

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