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Effect of neutron irradiation on thermal diffusivity of tungsten–rhenium alloys

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Abstract

Thermal diffusivities of W–Re alloys were examined before and after neutron irradiation by a laser-flash method. W and W–Re alloys (up to 25 mass% Re) were irradiated in the Japan materials test reactor (JMTR) reactor at 330 K to thermal and fast neutron fluences of 1.03×10^{20} and 3.37×10^{19} ($E > 1$ MeV), respectively. After irradiation thermal diffusivities of W and W–5% Re decreased, while those of W–10% Re and 25% Re increased. In the unirradiated condition, though the diffusivities of W and W–5% Re decreased with an increase of temperature, those of W–10% Re and W–25% Re increased. These temperature dependencies were the same after the irradiation. After annealing at 973 K for 10 min, the thermal diffusivity was restored for W–5% Re at room temperature, while a difference still existed between those of the irradiated and unirradiated W and W–25% Re. The observed changes are discussed in terms of lattice constant measurements and precipitation behavior.

1. Introduction

Tungsten (W) is considered as one of the candidate materials for structural applications in fusion reactors. However, it has low room temperature ductility and low recrystalization temperature. These disadvantages are improved to some extent with the addition of rhenium (Re). Further, Re is one of the products in the transmutation of W after heavy neutron irradiation [1–3]. According to Noda's estimation of compositional change of W at the armor position [4], W is transmuted to W–5% Re–0.02% Os (osmium), W–10% Re–0.1% Os and W–25% Re–1% Os for the irradiation of 3, 6 and 15.5 MW y/m^2, respectively. On the other hand, one of the important material properties for the plasma facing components is thermal conductivity.

The aims of this study are to investigate the effect of neutron irradiation on the thermal diffusivities of W and W–Re alloys, and their temperature dependence before and after the irraditaion. Measurements of thermal diffusivity and lattice constant of W and W–Re alloys (up to 25 mass% Re) were carried out before and after neutron irradiation in the Japan materials test reactor (JMTR) reactor. The results are discussed from the viewpoint of change in lattice constant and precipitation behavior. As the produced Os content is little compared with that of Re, the effect of Os content on the property was not considered.

2. Experimental procedure

W, W–5.3% Re, W–9.8% Re and W–25.0% Re (in mass%) were used in this study. All the materials were wrought products with 20–30 mm diameter and 10–20 mm thickness. They were stress relieved for 15 min at 1373 K before machining. Specimens for measurement of thermal diffusivity and lattice constant were disks with 8 mm diameter and 1 mm thickness; these were cut by a wire spark cutting tool. All the specimens were polished to #04 before the tests.

Some of the 8 mm disks were irradiated in a fission reactor, JMTR at Oarai Research Establishment of Japan Atomic Energy Research Institute, for up to 144 reactor full power hours at about 330 K. The thermal

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neutron and fast neutron fluences ($E>1$ MeV) were 1.03×10^{20} and 3.37×10^{19} n/cm^2, respectively. After cooling, the effect of the neutron irradiation on the thermal diffusivity was investigated.

A thermal coefficient measuring apparatus (Sinku Rikou) was used for measurement of the thermal diffusivity of the specimens before and after the irradiation. Details of this system can be found elsewhere [5]. The thermal diffusivity of the specimen was evaluated by a conventional 'half time method' [6]. The measurement was carried out from room temperature to 973 K in a vacuum of 6.7×10^{-3} Pa. For unirradiated specimens, the measurement at room temperature was again done after cooling to confirm that oxidization did not occur during heating. The irradiated specimens were first heated to 973 K and held for 10 min, and the treatment is referred to as 'Annealing' in this paper. The thermal diffusivity was measured during heating and continued on cooling from 973 K to room temperature.

Lattice constants of the specimens before and after neutron irradiation, and after the annealing were evaluated by X-ray diffraction with the use of Cu $K\alpha_1$ ($\lambda=1.54051$ Å). Determination of the lattice constants of the specimens was done according to Cohen's method [7].

3. Results and discussion

Fig. 1 shows the effect of Re content on the thermal diffusivity of W before and after the neutron irradiation. The diffusivities of unirradiated W–Re alloys decreased with an increase of Re content parabolically. After the irradiation, diffusivities of W and W–5% Re decreased while those of W–10% Re and W–25% Re increased. Specifically, a significant increase was observed in W–25% Re.

The change in thermal diffusivity of unirradiated W–Re alloys with temperature is shown in Fig. 2. The large drop in diffusivity was observed in W; however, only a small change was found in W–Re alloys. Further, though the diffusivities of W and W–5% Re decreased with an increase of test temperature, those of W–10% Re and W–25% Re increased. Hence, the difference among W and W–Re alloys decreased with increasing temperature. Figs. 3–6 denote the thermal diffusivities of irradiated and unirradiated W and W–Re alloys as a function of test temperature. In W, diffusivities of both the specimens before and after the irradiation decreased with an increase of test temperature, and the unirradiated specimen always has larger value than the irradiated specimen (Fig. 3). Annealing the irradiated specimen at 973 K brought about recovery of the diffusivity to some extent.

The diffusivity of irradiated W–5% Re first decreased on heating to around 500 K; but it increased slightly upon heating to 973 K, and on cooling from 973 K it did not change much. At room temperature it coincided with the value of unirradiated W–5% Re (Fig. 4).

W–10% Re did not change largely in its diffusivity with test temperature; and though a slight increase was observed in the irradiated specimen as compared with that of unirradiated one, no large diffusivity difference existed between irradiated and unirradiated specimen upon heating or cooling (Fig. 5).

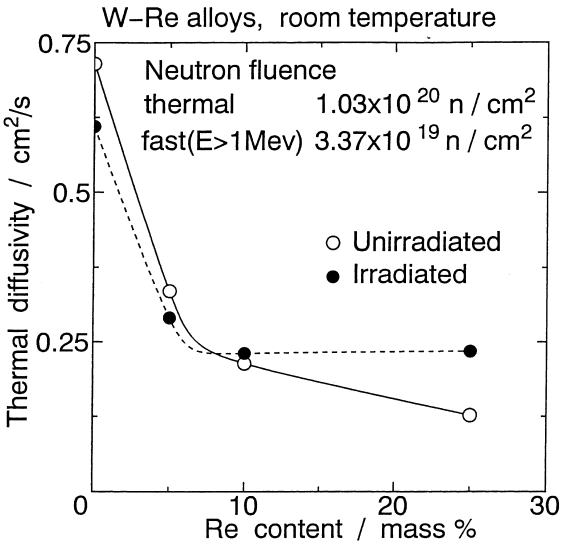

Fig. 1. Thermal diffusivity of W–Re alloys at room temperature as a function of Re content.

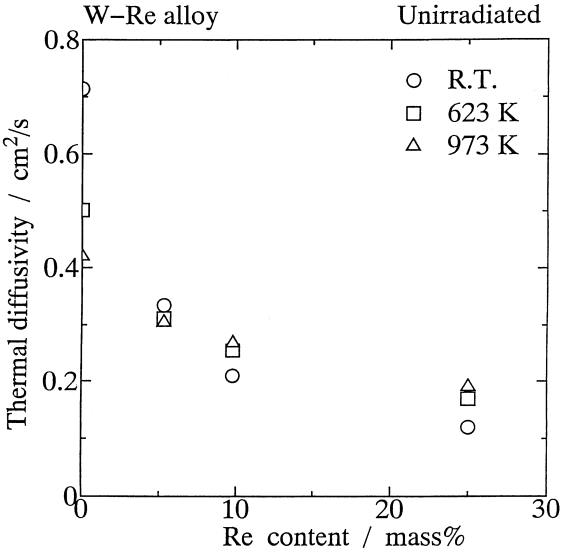

Fig. 2. Thermal diffusivity of unirradiated W–Re alloys at room temperature, 673 and 973 K.

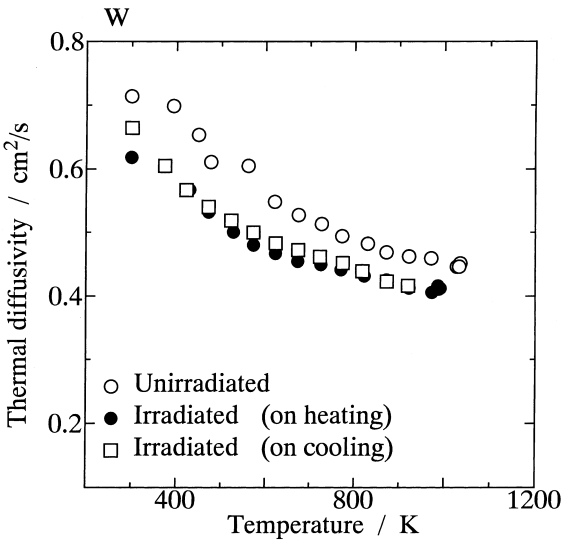

Fig. 3. Thermal diffusivity of W as a function of test temperature.

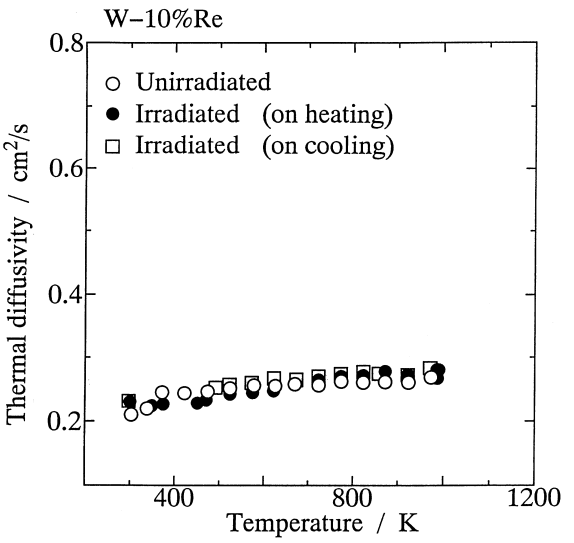

Fig. 5. Thermal diffusivity of W–10% Re as a function of test temperature.

A large difference in diffusivity between irradiated and unirradiated W–25% Re was observed as shown in Fig. 6. The temperature dependence was the same in both specimens and annealing did not seem to bring about the restoration of diffusivity in the irradiated specimen.

Fig. 7 represents the lattice constants of W–Re alloys as a function of Re content. Lattice constants of all the specimens increased after irradiation. Specifically, W–25% Re exhibited a significant increase. Further, after annealing the lattice constants of W, W–10% Re and W–25% Re decreased; however, W–25% Re still has a large lattice constant compared with that of unirradiated specimen. It is very curious that lattice constant of W–5% Re after annealing increased from that of the irradiated specimen.

If one assumes that an approach of the lattice constant of an irradiated specimen after annealing to the value of the unirradiated specimen means recovery, all the specimens besides W–5% Re show recovery to some

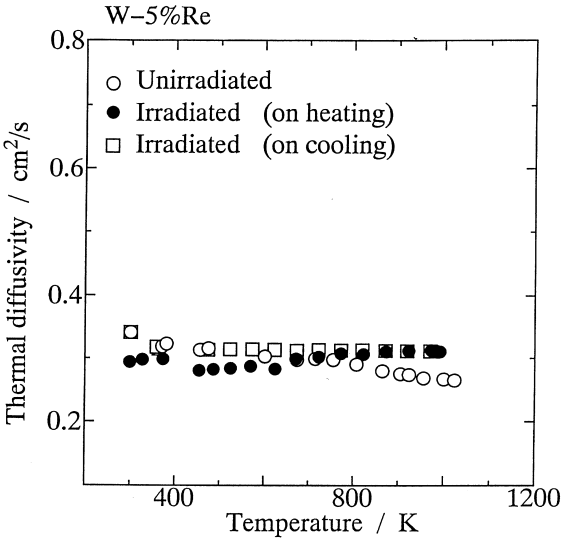

Fig. 4. Thermal diffusivity of W–5% Re as a function of test temperature.

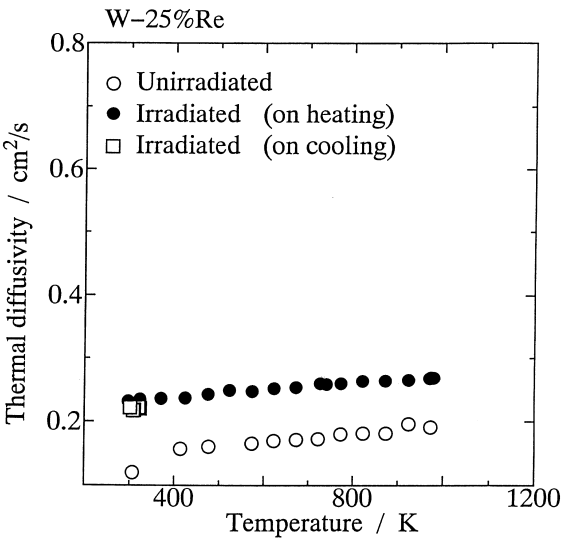

Fig. 6. Thermal diffusivity of W–25% Re as a function of test temperature.

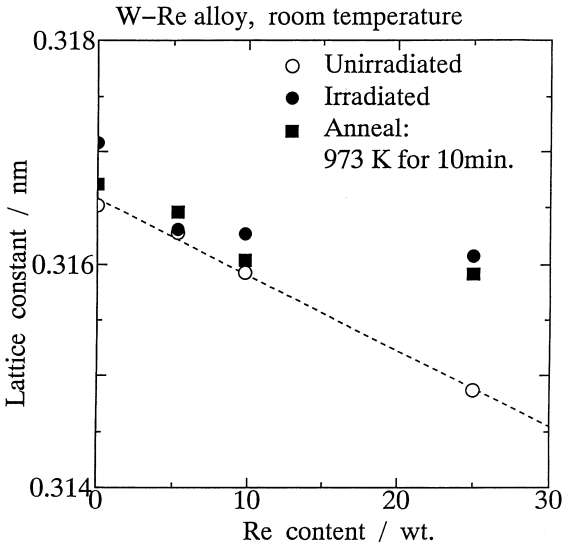

Fig. 7. Lattice constant of W–Re alloys as a function of Re content.

extent. From the thermal diffusivity data, the data of W and W–25% Re suggest this phenomena. As for W–10% Re, as all the data, not only at room temperature but also at high temperatures, were almost the same, it was difficult to discern recovery.

The room temperature thermal diffusivity of W–5% Re after annealing coincided with that of unirradiated one, while its lattice constant after annealing increased further from that of the irradiated specimen. The temperature dependence of the data on heating and also on cooling behaved in a different manner from those of W and W–10% Re, suggesting an occurrence of anomaly during heating and cooling processes of the test. This anomaly would bring about the observed increase in lattice constant. At present, the reason is unknown.

There are many problems to solve besides the anomaly mentioned above. First is the difference of the temperature dependence between W and W–5% Re, and W–10% Re and W–25% Re. Fabritsiev et al. reported the thermal conductivity data of Mo–Re alloys [8]. Their data on the conductivity at room temperature coincide with our data; thermal conductivity decreases with an increase of Re content. Further the temperature dependence of the thermal conductivity in Mo was the same as that in W. However, the conductivity of Mo–Re alloys decreased with an increase of temperature; no anomaly like the one observed in W–Re alloys was evident.

Second is the significant increase of thermal diffusivity and the lattice constant after irradiation that was observed in W–25% Re. The cause of this phenomenon is at present also not understood. Intermetallics of WRe (σ phase and/or χ phase [9]) may precipitate, due to defects introduced through neutron irradiation, during heating. If so, the Re content in α solid solution would decrease and this may lead to the increase in the lattice constant. However, at least X-ray diffraction did not reveal the existence of these phases.

Further study is needed to solve these problems through examining other physical properties such as electric resistivity and metallography.

4. Conclusions

The effect of neutron irradiation (neutron fluences: thermal 1.03×10^{20}, fast ($E>1$ MeV) 3.37×10^{19} n/cm^2, respectively) on the thermal diffusivities of W and W–Re alloys (up to 25 mass% Re) and their temperature dependence before and after the irradiation were investigated.

The results obtained were as follows:

1. At room temperature, thermal diffusivities of W and W–5% Re decreased after irradiation, while those of W–10% Re and W–25% Re increased.
2. The thermal diffusivities of W and W–5% Re decreased with an increase in test temperature, while those of W–10% Re and W–25% Re increased. The difference among them decreased with increasing test temperature. These same temperature dependencies were exhibited after irradiation.
3. After annealing at 973 K for 10 min, the thermal diffusivity of irradiated W–5% Re was restored at room temperature, while there existed large differences between the thermal diffusivities of the irradiated and unirradiated W and W–25% Re.
4. The lattice constants of W and W–Re alloys increased after irradiation. Specifically, W–25% Re exhibited a large increase.

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