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Defect structural evolution in high purity tungsten irradiated with electrons using high voltage electron microscope

S. Fukuzumi [a], T. Yoshiie [a,*], Y. Satoh [b], Q. Xu [a], H. Mori [c], M. Kawai [d]

[a] *Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka-fu 590-0494, Japan*
[b] *Institute for Materials Research, Tohoku University, Katahira, Aoba-ku, Sendai 980-8577, Japan*
[c] *Research Center for Ultra-High Voltage Electron Microscopy, Osaka University, Suita 565-0871, Japan*
[d] *High Energy Accelerator Research Organization, Tukuba 305-0801, Japan*

Abstract

Four types of high purity tungsten were irradiated with 2 MeV electrons to 5 dpa using a high voltage electron microscope, and defect structural evolutions were examined as a function of the irradiation temperature and the concentration of impurity atoms. Three of materials were made by sintering of tungsten powder with purity of 99.999% (5N-W), 99.99% (PF-W) and 99.95% (N-W), and one was a chemical vapor deposited tungsten of 99.9999% (CVD-W) purity. The formation of interstitial type dislocation loops is observed above room temperature by electron irradiation. In sintered tungsten, the number density of loops increases with increasing density of impurity atoms, i.e., N-W > PF-W > 5N-W. The density of loops in CVD-W is relatively high, contrary to its purity. In CVD-W, a heterogeneous formation of loops is observed at above 573 K. Loops are aligned on layers, and no loops are formed between the layers. All four types of specimens have a change in slop of the temperature dependence of loop number density at around 500 K which is caused by impurity atoms. Results of radioactivation analysis and hardness testing are also presented.

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1. Introduction

Tungsten is one of the most suitable materials for a solid target spallation neutron source because of the highest neutron yield among non-actinide heavy metal nuclides [1]. Tungsten has two deficiencies, however, for a MW class spallation neutron source. One is the poor corrosion resistance against water coolant. In order to avoid the corrosion of tungsten, Kawai et al. have developed a tantalum-clad tungsten target [1]. The other is high susceptibility to irradiation embrittlement [2,3]. The damage structural evolution in tungsten is affected remarkably by impurity atoms. Sometimes important irradiation results were reported without the purity of tungsten for example [4,5] and we cannot distinguish impurity effects from irradiation effects. Even though a large number of impurity elements are generated as spallation reaction during high dose of high energy proton irradiation, the initial purity is important, because the nucleation of defect clusters finishes at the initial stage of irradiation. In order to clarify the effect

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[*] Corresponding author. Tel.: +81 724 51 2473; fax: +81 724 51 2620.
E-mail address: yoshiie@rri.kyoto-u.ac.jp (T. Yoshiie).

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of impurities, irradiation damage experiments with high purity tungsten are required.

High voltage electron microscopy is a useful technique for the study of irradiation damage in materials [6,7]. Direct observation of defect structural evolution is possible and the displacement damage rate is high (the order of 10^{-2} dpa/s). In this paper, the defect structural evolution in four types of high purity tungsten will be studied. They were irradiated with 2 MeV electrons using a high voltage electron microscope, and defect structures were examined as a function of the irradiation temperature and the concentration of impurity atoms.

2. Experimental procedure

Three different purities of tungsten made by sintering of tungsten powder, the purity of 99.999% (5N-W), 99.99% (PF-W) and 99.95% (N-W), and a chemical vapor deposited tungsten of 99.9999% purity (CVD-W) were used. 5N-W, PF-W and CVD-W were from A.L.M.T. Corp. A list of impurity elements is given in Table 1. N-W was from Plansee AG, and a list of impurity elements was not obtained. Instrumental neutron activation analysis was performed to measure the concentration of trace elements in tungsten. Neutron irradiation was made for 30 s at a core site connected by a pneumatic tube #2 of the Kyoto University Reactor, a 5MW light water reactor. The gamma-rays were measured with a low back ground germanium detector. Hardness test was performed with a dynamic hardness tester (Shimadzu DUH-201) at room temperature with a load of 50 gf.

The specimens were sliced to a thickness of 0.1 mm, shaped into disks with diameter of 3 mm and finally electro-polished for electron microscopy observation. They were irradiated with 2 MeV electrons using a high voltage electron microscope of Osaka University, H-3000, at 300 K and at temperatures of 373–773 K in steps of 100 K for 180 s each. The total displacement damage was 5 dpa estimated by a threshold energy of 35 eV [8].

The thickness of irradiated area of specimens to obtain the number density of defect clusters was measured using thickness fringes taken by two beam condition using a conventional 200 kV electron microscope. Loops were observed near (1 0 0) foil orientation with reflections of several planes to image all of loops.

3. Results and discussions

The result of gamma-ray spectroscopic measurements after neutron irradiation is shown in Fig. 1. No significant differences are found in the four types of specimens. In addition to the impurities listed in Table 1, Ti (2614.5 keV) is detected in all specimens. Only in CVD-W, Ac (911.1 keV) and Bi (1120.3 keV) are present. The quantitative analysis has not been performed yet. In Table 2, the results of hardness tests are presented. No change of hardness is detected with the difference of the concentration of impurity atoms.

During electron irradiation using the high voltage electron microscope, the formation of interstitial type dislocation loops is observed as shown in Figs. 2–5. The nucleation of loops occurs at an initial stage of irradiation and the number density does not change during irradiation. The density has strong irradiation temperature dependence. By increasing irradiation temperature, the number density of loops decreases and their growth speed increases. At temperatures higher than 773 K no loops are formed. A difference in defect structural evolution among the four types of specimens is detected as shown in Fig. 6. In sintered tungsten, the number density of loops increases with increasing impurity atoms, i.e., N-W > PF-W > 5N-W.

Yoshida et al. [9] and Kiritani [8] explained the variation of impurity effects with temperatures as follows. In the case where the di-interstitals are stable nuclei of interstitial type dislocation loops, the nucleation rate of loops is expressed as the sum of the interstitial-to-interstitial combination rate and interstitial-to-interstitial-impurity complex combination rate. At high temperatures (the temperature region I in Fig. 7), an interstitial-impurity complex dissociates before a next interstitial comes to form a stable nucleus, and the impurity atoms have no influence on the nucleation of loops.

Table 1
Impurity elements in tungsten (unit: ppm)

	O	N	C	Na	K	Al	Ca	Cr	Cu	Fe
PF-W	<10	<10	<10	4	<5	3	<1	<1	<1	5
5N-W	<10	<10	<10	0.1	<0.1	<0.1	0.2	0.4	<0.1	1.5
CVD-W	0.065	0.025	0.13	0.011	0.005	0.001	0.04	0.001	0.001	0.047

	Mg	Mn	Mo	Ni	Si	Sn
PF-W	<1	<1	11	2	<5	<2

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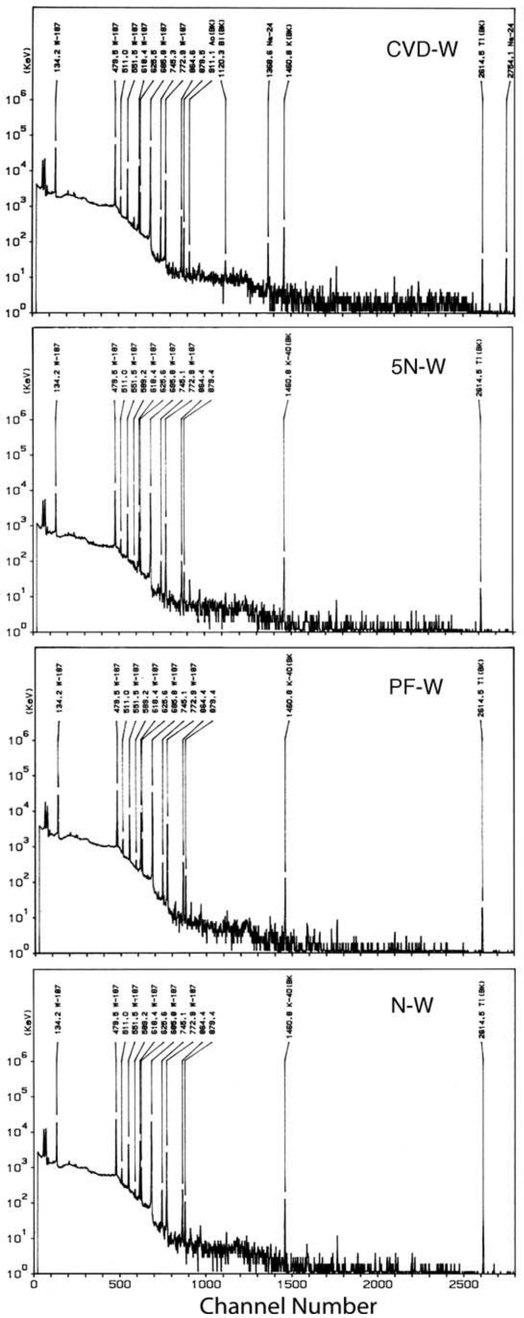

Fig. 1. The result of radioactivation analysis of the four types of tungsten specimens.

At medium temperatures (the temperature region III in Fig. 7), the dissociation rate of interstitial-impurity complexes is small and all impurity atoms become the nuclei of loops. If the nucleation of loops through interstitial-to-interstitial formation is small, the number density of loops saturates and has no temperature dependence. At low temperatures (the temperature region V in

Table 2
Result of dynamic hardness test (arbitrary unit)

	CVD-W	5N-W	PF-W	N-W
Hardness	520.4 ± 14.9	521.18 ± 12.4	510.5 ± 10.3	517.4 ± 15.6

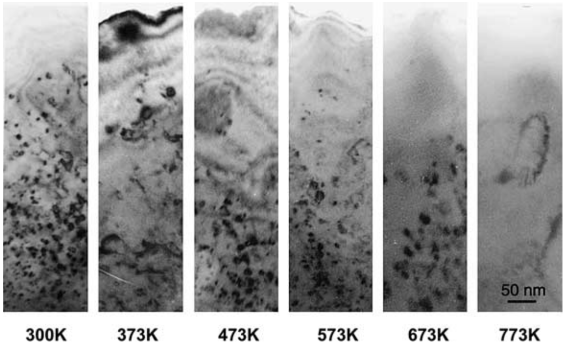

Fig. 2. Electron irradiated CVD-W at each temperature for 3 min (5 dpa).

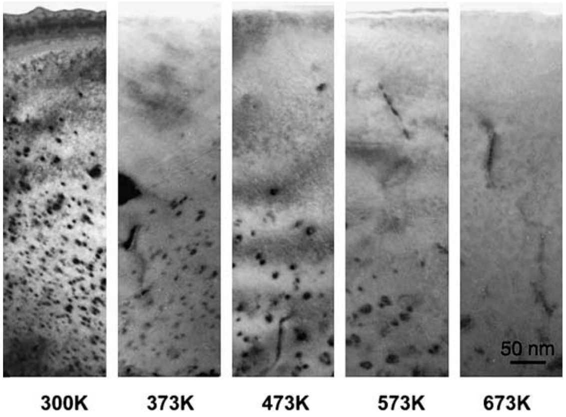

Fig. 3. Electron irradiated 5N-W at each temperature for 3 min (5 dpa).

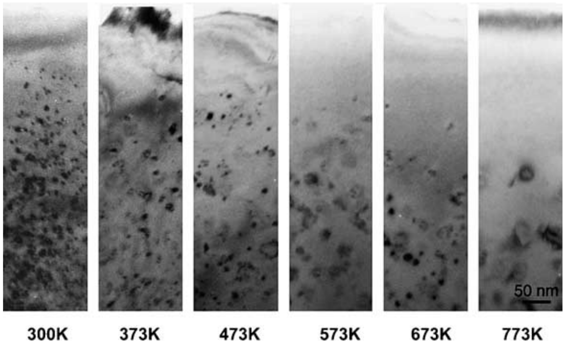

Fig. 4. Electron irradiated PF-W at each temperature for 3 min (5 dpa).

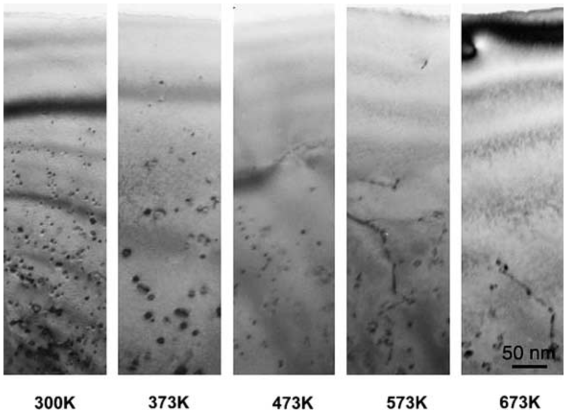

Fig. 5. Electron irradiated N-W at each temperature for 3 min (5 dpa).

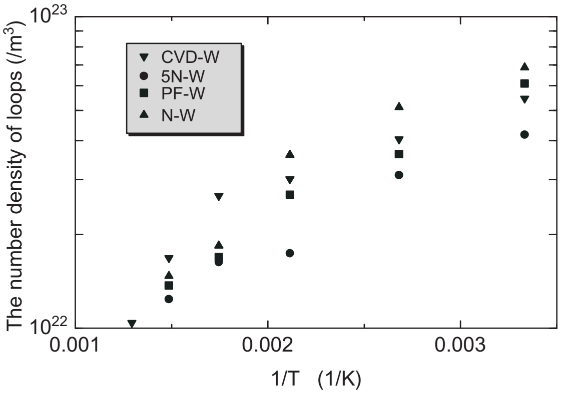

Fig. 6. The temperature dependence of the number density of interstitial type dislocation loops.

Fig. 7), the mobility of the interstitials is extremely low and the interstitial concentration increases. The formation rate of interstitial-to-interstitial combination rate exceeds the interstitial-to-interstitial impurity combination rate. The majority of loops nucleate without the help of impurity atoms. The temperature regions II and IV are transient regions.

In the present experiment, the change of the number density of loops corresponds to the temperature regions of II, III, IV and V in Fig. 7. All four types of specimens have a change in slop at around 500 K which corresponds to the regions from II to IV. 5N-W and CVD-W have a clear temperature region in which the number density of loops does not change with irradiation temperatures (region III). From the number density of loops at this region in 5N-W, the impurity concentration is estimated to be 2.7×10^{-7}. This value is two orders lower than the actual concentration of impurity atoms (Table 1) if we assume that all impurities nucleate loops.

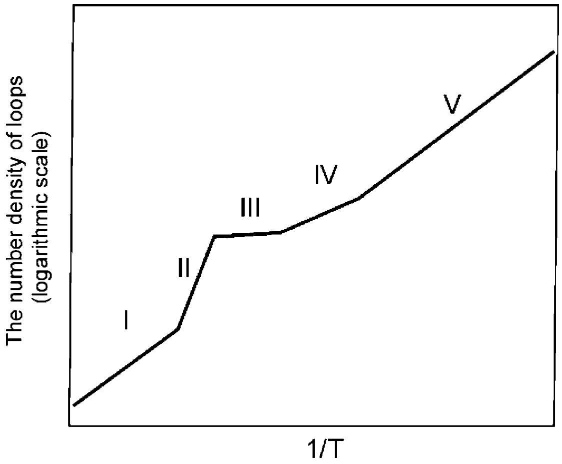

Fig. 7. Expected variation of the number density of interstitial type dislocation loops with irradiation temperatures.

The value 2.7×10^{-7} should be considered to be the concentration of effective impurity atoms which contribute to the formation of interstitial-impurity complexes. At the temperature region V, the effect of impurity atoms is not significant. The activation energy of interstitial migration in 5N-W is estimated to be 0.15 eV using the formula by Kiritani et al. [7] at this temperature region. This value is important to discuss the formation and growth of interstitial clusters.

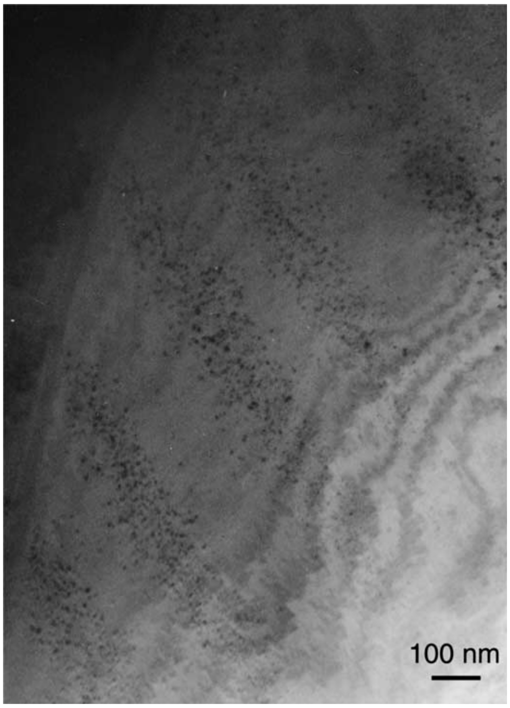

Fig. 8. Electron irradiated CVD-W at 573 K for 3 min (5 dpa).

The number density of interstitial type dislocation loops in CVD-W is relatively high inspite of its purity. Heterogeneous distribution of loops is observed at above 573 K as shown in Fig. 8. Loops are aligned on layers and no loops are formed between the layers. These are probably caused by the heterogeneous distribution of impurity atoms in CVD-W.

CVD-W was formed by the reaction of WF_6 and H_2 at 973 K. These gases are introduced to a reaction chamber and a tungsten layer is formed on a rotating table in the chamber. The estimation of the rotating speed and the amount of stacking thickness during one turn indicate that the separation of impurity layers almost corresponds to the stacking thickness of one turn. Therefore, we conclude that the impurity level is high at one part of the turning table during vapor deposition. The impurity atoms cause the preferential growth of interstitial type dislocation loops by electron irradiation. This result indicates the possibility to fabricate an arbitrary defect structure by controlling the impurity distribution in high purity metals.

4. Conclusions

Four types of tungsten specimens were irradiated with electrons by the high voltage electron microscope in the temperature range between room temperature and 773 K. In sintered tungsten, the number density of interstitial type dislocation loops is in the order of the concentration of impurity atoms. That in CVD-W is relatively high, and a heterogeneous formation of loops is observed at above 573 K. They are formed by the heterogeneous distribution of impurity atoms during the fabrication of the specimen.

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