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Journal of Nuclear Materials 275 (1999) 101–107
journal of nuclear materials
www.elsevier.nl/locate/jnucmat

Damage observed in Mo irradiated with 14 MeV neutrons at RTNS-II

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Received 3 August 1998; accepted 4 March 1999

Abstract

The damage in pure molybdenum irradiated at RTNS-II with 14 MeV neutrons was examined by electron microscopy. Annealed specimens were irradiated between 60°C and 450°C at fluences from 4×10^{19} to 7×10^{22} n/m^2. Small defects clusters were observed in the irradiated specimens. The clusters, which become larger with increasing irradiation temperature and fluence, were determined to be interstitial loops from correlation of results from bulk and thin specimens. Groups of small loops were formed for high fluences at high temperatures. Voids were not observed in the specimens. The damage structures formed by fusion-neutron irradiation differed considerably from those formed by fission-neutron irradiation in a previous study. Whereas small interstitial loops are formed for fusion-neutron irradiation, large interstitial loops with groups of small vacancy loops inside them are formed for fission-neutron irradiation. This difference in damage structure is attributed to the change in deposited energy density.

1. Introduction

The component materials of a fusion reactor are irradiated with 14 MeV neutrons. Investigations of damage evolution in metals irradiated by 14 MeV neutrons has therefore been of significant interest. The damage structures of metals and alloys irradiated by fission-neutrons to simulate 14 MeV neutron irradiation have been extensively studied [1–9]. There are two important differences between fusion-neutron irradiation and fission-neutron irradiation. In fusion-neutron irradiation, the energy density in the displacement cascades and concentration of transmutation-produced helium and hydrogen atoms are higher than those in fission-neutron irradiation. Therefore, materials irradiated by 14 MeV neutrons may more reliably be used to investigate damage evolution in fusion reactor materials. Many investigations of damage structures have been carried out using fcc metals such as Au, Cu and Ni [10–12] irradiated with fusion-neutrons. For bcc metals than for fcc metals, especially Mo, we have only limited results concerning damage structures [13,14] due to the lower yield of defect clusters which are observable by electron microscopy. Brimhall et al. [13] observed a low density of defect clusters, 4 nm in diameter in 14 MeV neutron-irradiated Mo and concluded that 14 MeV neutrons produce more damage than fission-neutrons for equivalent dpa levels of irradiation. Yoshida et al. [14] observed small interstitial loops near dislocation lines in Mo irradiated by 14 MeV neutrons.

In the present study, the damage structures in Mo irradiated over a wide range of temperature and fluence by fusion-neutrons were examined by electron microscopy, and the structures were compared with those from fission-neutron irradiation.

2. Experimental procedure

The material used in the present experiment was pure molybdenum foil of VP grade from MRC. The same

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0022-3115/99/$ - see front matter
PII: S0022-3115(99)00093-8

material was previously used in fission-neutron irradiation experiments by the present authors [15]. The details of the specimen were previously reported [15]. The annealed specimens were irradiated by 14 MeV neutrons from a rotating target neutron source (RTNS-II) at the Lawrence Livermore National Laboratory up to 7×10^{22} n/m^2 at several temperatures ranging from 60°C to 450°C. Most of the irradiated specimens were bulk in the present experiment. Pre-thinned specimens, called thin specimens hereafter, were also irradiated at 60°C and 150°C. The bulk specimens were electro-polished and observed together with the thin specimens using a JEOL-200-CX electron microscope at room temperature.

3. Experimental results and discussion

Small point defect clusters were observed in pure Mo specimens which were irradiated at various conditions. The number density, size and distribution of the clusters depended on the experimental conditions.

3.1. Thickness dependence

In Fig. 1, the correlation between bulk and thin specimens is shown. The damage structures were imaged by weak-beam dark-field conditions. Although both thin and bulk specimens were irradiated at the same time (1.2×10^{21} n/m^2 at 60°C), the low density of small defect clusters observed in the bulk specimens was not observed in the thin specimens. The diameter of the clusters was smaller than 3 nm. Similar results were obtained for the specimens irradiated at 150°C (1.3×10^{21} n/m^2). In this temperature range, interstitial atoms in Mo easily migrate due to the small migration energy [16]. On the other hand, vacancies can scarcely migrate during neutron irradiation. The migration energy of vacancies is 1.30 eV and a vacancy-related recovery stage appears at around 300°C when the specimens are annealed for 10 min [17]. In the bulk specimens of 50 μm thickness, most interstitial atoms would not escape to the surface during the irradiation. On the other hand, the interstitials in the thin specimens would mostly be lost at the surface. Therefore, interstitial clusters in the bulk specimens would nucleate and grow into an observable cluster size but in the thin specimens the clusters would not be formed. Thus, the small clusters observed in the bulk specimens should be interstitial in nature. English observed small interstitial loops at low neutron fluence in fission-neutron irradiated Mo [1]. If the small defect clusters were vacancy type, the clusters should also be observed in the thin specimen. Vacancies cannot escape from even the thin specimen because vacancy migration is very slow in this temperature range.

In bulk specimens, the small clusters were homogeneously formed during irradiation at 60°C to 1.2×10^{21} n/m^2. Clusters could not be observed at fluences lower than 7.5×10^{19} n/m^2 and 9.3×10^{19} n/m^2 for irradiation at 60°C and 150°C, respectively. These results are

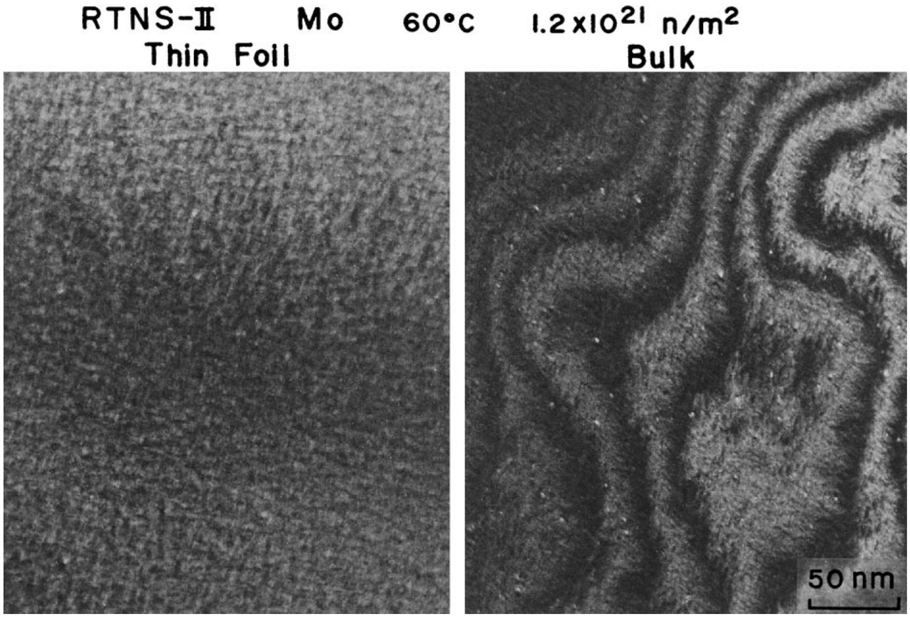

Fig. 1. The damage structures in bulk specimens and thin specimens irradiated at 60°C. The specimens were irradiated with 14 MeV neutrons at RTNS-II to a fluence of 1.2×10^{21} n/m^2.

consistent with interstitial atoms, ejected from damage cascades, migrating and aggregating to form small clusters during irradiation. After low fluence irradiation, interstitial clusters are not observed because of their small size. The clusters grow into an observable size as the fluence increases.

3.2. Irradiation temperature dependence

In Fig. 2 the damage structures of the bulk Mo specimen are shown for various irradiation temperatures between 90°C and 450°C. For 200°C and 400°C irradiations, the highest density areas of the clusters are shown. The number density of the small clusters decreased with irradiation temperature. At low fluence, the size did not change with irradiation temperature but at high fluence, the size increased with temperature. The distribution of clusters was more inhomogeneous than that formed at low irradiation temperature. The lowest fluence at which cluster could still be observed, increased with irradiation temperature. For example, no clusters were visible after irradiation at 60°C to 7.5×10^{19} n/m^2 but clusters were formed for a fluence of 1.2×10^{21} n/m^2; clusters were rarely observed for a fluence of 5.3×10^{21} n/m^2 at 450°C.

3.3. Fluence dependence

Fig. 3 shows how the damage structures in specimens irradiated at 90°C develop with increasing fluence. Both the density and size of the small clusters increased with fluence. The small clusters were homogeneously distributed in specimens irradiated at 90°C up to 1.5×10^{22} n/m^2. The fluence dependence of damage structure for irradiations at 200°C and 450°C is shown in Figs. 4 and 5, respectively. The small clusters formed by low temperature irradiation were distributed more homogeneously than those formed by high temperature irradiation. Fig. 6 shows the size distributions of clusters for increasing fluence after irradiation at 200°C and 450°C. With increasing fluence, the clusters exhibit significant growth and the distribution range broadens. For low fluence at high irradiation temperature, isolated groups of small clusters were formed. On the other hand, in low fluence at low irradiation temperature, small clusters were formed homogeneously. One example of the groups in the specimen irradiated at 450°C to 5.4×10^{21} n/m^2 is shown in Fig. 7. Each defect cluster in the groups appears to have been formed separately as shown in the figure and the size of groups was variable. Maher et al. [4–8] investigated systematically the influence of crystal perfection, irradiation dose and irradiation temperature on the distribution and nature of the visible defect clusters and their behavior on post-irradiation annealing in Mo irradiated with fission-neutrons by electron microscopy. They showed heterogeneous formation of small loops at impurities and formation of small interstitial loops at early stage of irradiation and discussed the growth of loops by glide and climb of the small loops. English [1] and Yoshida [14] observed

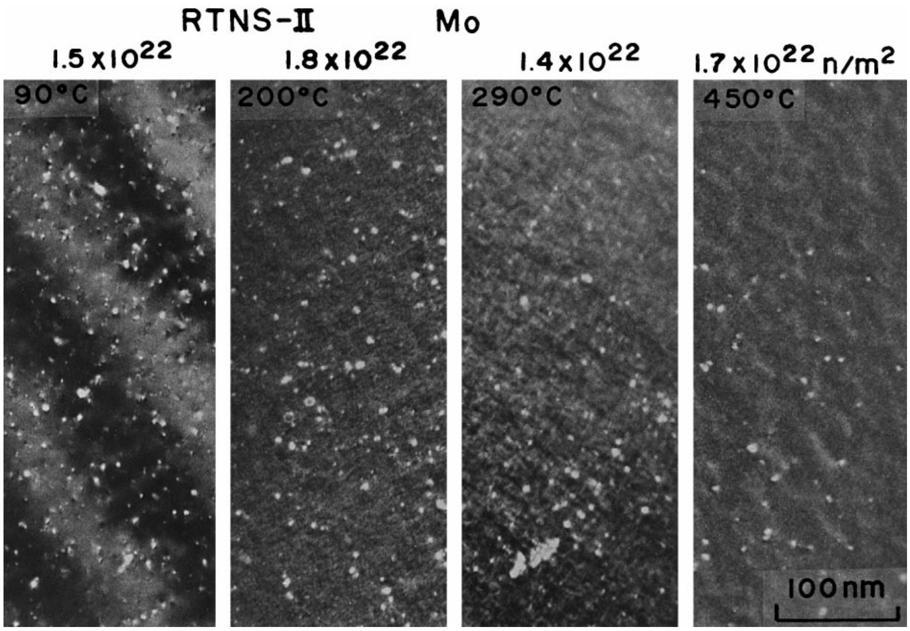

Fig. 2. The damage structures in bulk Mo specimens between 90°C and 450°C. The specimens were irradiated with 14 MeV neutrons at RTNS-II to 1.6×10^{22} n/m^2. The characteristic of the damage is that the cluster density decreases with irradiation temperature.

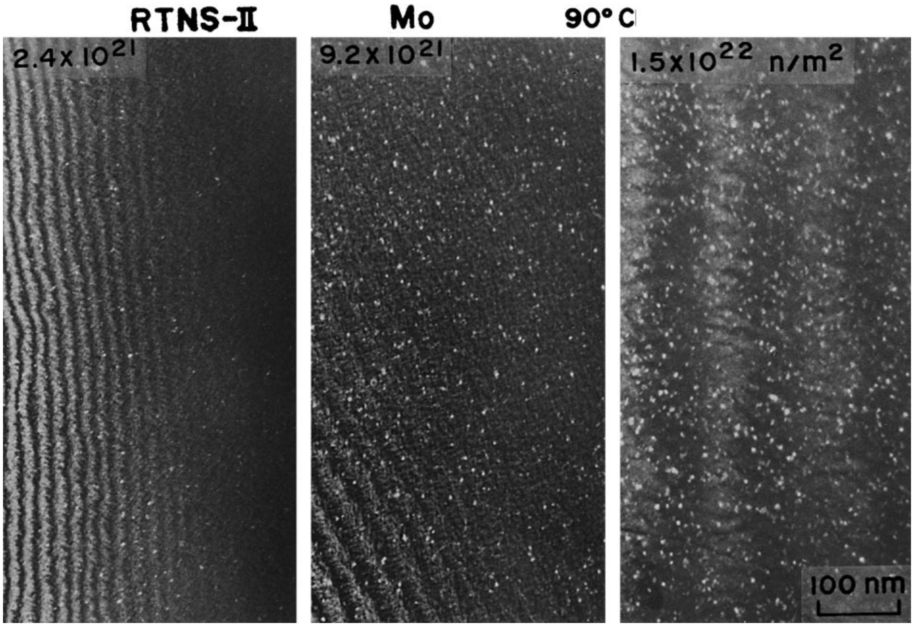

Fig. 3. The damage structures in bulk specimens irradiated at 90°C to fluences up to 1.5×10^{22} n/m^2. The small clusters are formed homogeneously in the specimens.

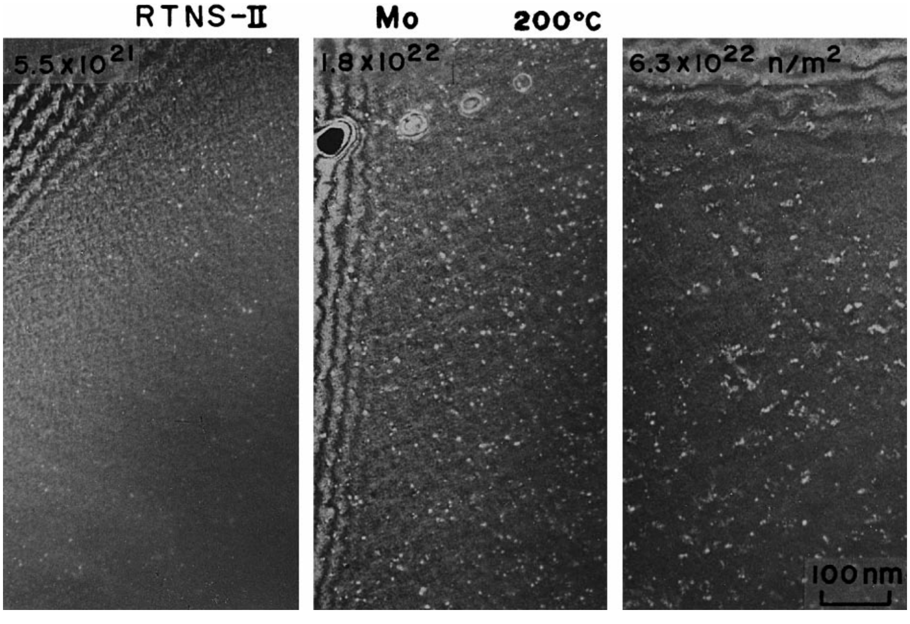

Fig. 4. The damage in bulk specimens irradiated at 200°C at various fluences.

inhomogeneous distribution of small loops around dislocations. Inhomogeneous formation of the groups may be caused by aggregation of interstitials around dislocations and/or regions containing impurity atoms. At higher fluence, some clusters appear to have coalesced to make larger loops with complicated shapes and with three dimensional distribution. They have no planar and row-like distribution as in the rafts mentioned below. This tendency was most pronounced at high temperature and high fluence (4.1×10^{22} n/m^2 at 450°C). The formation of groups of the loops cannot be explained by simple growth of the loops by absorbing interstitial atoms produced by further neutron-irradiation. The fluence increase in Fig. 5 (from middle to right) is only

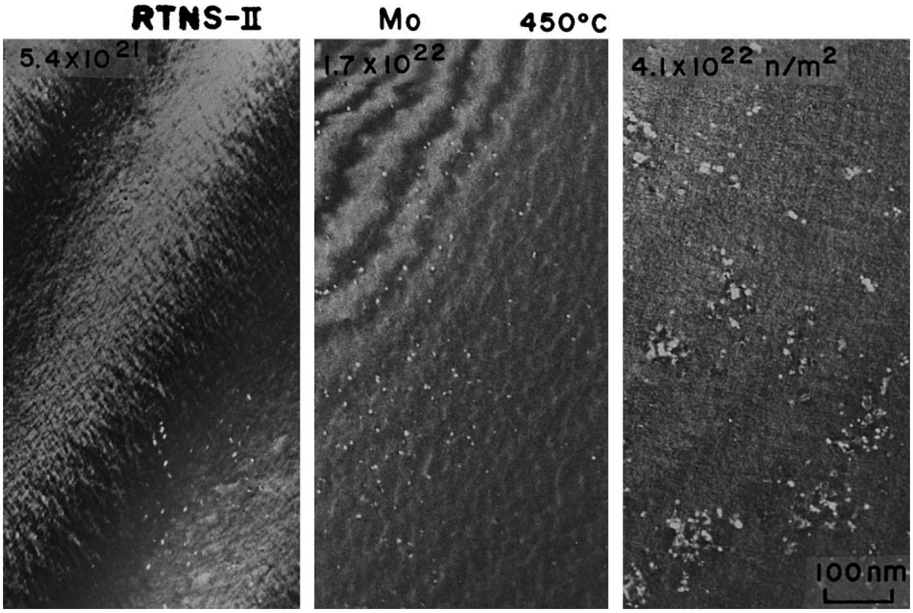

Fig. 5. The damage in bulk specimens irradiated at 450°C at various fluences.

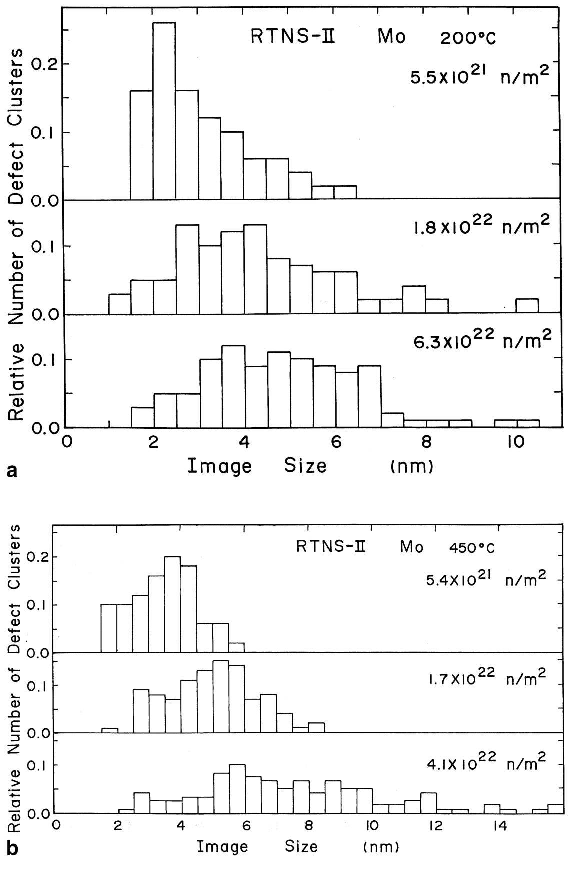

Fig. 6. Changes in size distributions of clusters with fluence. (a) 200°C (b) 450°C.

2.4 fold. The groups of the loops could be formed by loop glide and climb with some loop coalescence. Such a mechanism is necessary to account for such abrupt changes in loop size and loop distribution with a small fluence change.

Brimhall and Mastel [9] investigated the damage in Mo irradiated with fission-neutrons by transmission electron microscopy. Small dislocation loops distributed uniformly in their Mo irradiated at low temperatures. Then groups of small loops (rafts) were observed as characteristic damage at 400–600°C. The rafts were distributed in plane and the Burgers vector was the same for all small loops within a raft. They accounted for the observed microstructures by a combination of glide and climb of small loops.

In Mo fission neutron-irradiated in the Joyo experimental fast reactor, we observed large interstitial loops with some complicated structures inside them, groups of small loops (so called raft [9]), voids and a high density of dislocation lines in specimens irradiated between 400°C and 600°C [15]. With a fluence of 7.9×10^{23} n/m^2 ($E > 0.1$ MeV) at 400°C, large dislocation loops, which were somewhat inhomogeneously distributed in the specimens, were formed. With further irradiation, the dislocation loops grew to larger ones with small interior vacancy loops. The groups of small vacancy loops were formed by a geometrical reaction, which was different from the mechanism presented by Brimhall and Mastel [9], during growth of the large convoluted dislocation loops absorbing interstitials [18]. The groups of small dislocation loops (rafts) were observed at one side of the dislocations or inside of the large interstitial loops and

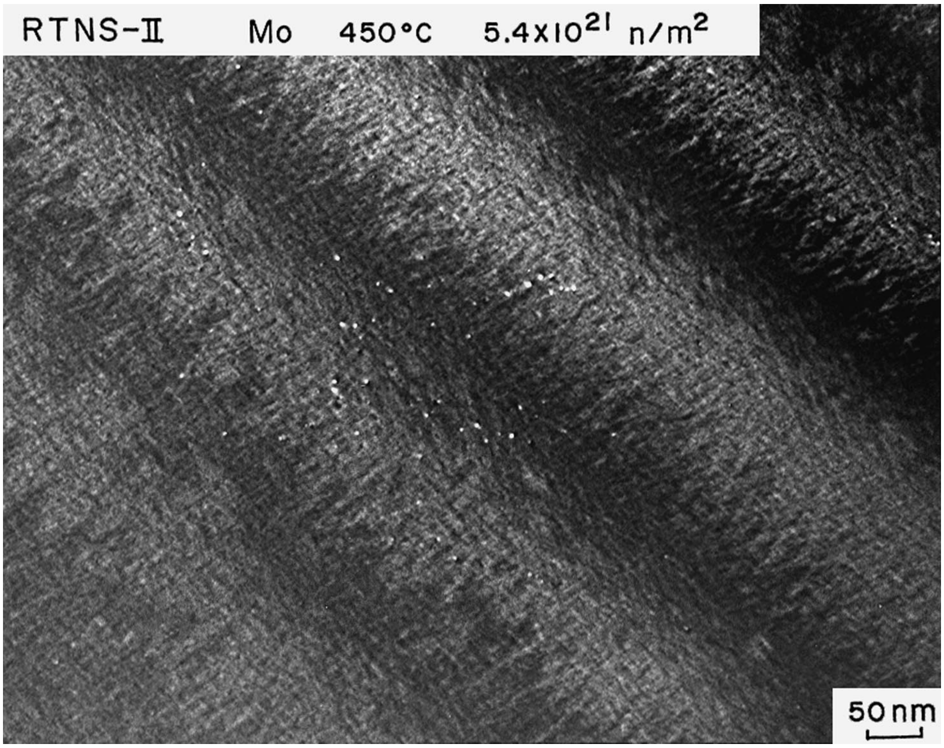

Fig. 7. Inhomogeneous distribution of damage in the matrix irradiated at 450°C to 5.4×10^{21} n/m^2.

aligned in rows. Burgers vector of the small loops was the same as that of the dislocation or large loop.

The amount of damage in Mo was calculated by Nishiguchi for comparison of the RTNS-II irradiation and Joyo irradiation [19]. Damage was 1.75×10^{-25} dpa/(n/m^2) for the RTNS-II irradiation and 1.46×10^{-26} dpa/(n/m^2) for the Joyo irradiation. Therefore, fluences of 7.9×10^{23} n/m^2 in the Joyo irradiation and 4.1×10^{22} n/m^2 in the RTNS-II irradiation correspond to 1.2×10^{-2} dpa and 7.2×10^{-3} dpa, respectively. Remarkable differences were observed between the damage structures formed by fusion neutrons and those formed by fission neutrons. Large interstitial loops with groups of small vacancy loops in them were formed during fission-neutron irradiation. On the other hand, groups of small interstitial loops were formed during fusion-neutron irradiation. In fusion-neutron irradiated Mo, displacement damage cascades are generated with high deposited energy density. The local concentration of interstitial atoms would be expected to be higher than those from fission-neutron irradiation. Thus, the number density of nuclei of small interstitial clusters should be higher than for fission-neutron irradiation. After nucleation, the growth speed of the loops is slow during fusion-neutron irradiation. At the same dpa, the mean number of interstitial atoms absorbed per loops for fusion-neutron irradiation is less than for fission-neutron irradiation.

In fusion-neutron irradiation, no visible voids were formed in all specimens irradiated up to 4.1×10^{22} n/m^2, which corresponds to 7.2×10^{-3} dpa. In fission-neutron irradiation, we could not observe voids after 7.9×10^{23} n/m^2 (1.2×10^{-2} dpa) at 400°C, but high number densities of small voids were formed in specimens irradiated to 8.1×10^{24} n/m^2 (1.2×10^{-1} dpa) at 400°C and 500°C.

During fusion-neutron irradiation, vacancy type defects are expected to form at high fluence just as voids formed after high fluence Joyo irradiation. It is probable that void could not be observed due to the smallness and/or low number density after 450°C irradiation in the RTNS-II. Further heavy irradiation in the RTNS-II is necessary for valid comparisons with void formation in Mo irradiated by fission-neutrons. The number density, size and distribution of the voids could well be different for the two irradiations.

In conclusion, the damage evolution in pure Mo irradiated by fusion-neutrons is suggested as follows. The interstitial atoms produced by irradiation aggregate and small interstitial loops are formed. The small loops grow by absorbing the subsequently produced interstitials. Upon further irradiation at high temperature, the groups of the loops are formed. The damage microstructures from fusion-neutron irradiation differ considerably from those of fission-neutron irradiation. During the early stage of irradiation, high number densities of small interstitial loops formed during fusion-neutron irradiation. On the other hand, a low density of large interstitial loops formed during fission-neutron irradiation. The difference is probably caused by the difference in deposited energy density of the two irradiations. To clarify the fusion–fission correlation of the microstructure in Mo, we need fusion-neutron irradiation at high fluences, equivalent to damage levels at which observable voids were formed in fission irradiations. During evolution of the damage structure by the consumption of interstitials at sinks, the vacancy concentration should increase and vacancy defects might cluster to form sub-microscopic size voids at high temperatures for this fluence level.

Acknowledgements

We would like to express our thanks to Professors M. Kiritani, Hiroshima Institute of Technology and S. Ishino, Tokai University for organization of the program on RTNS-II Utilization. We are also grateful to Professor H. Kayano and other members of the Oarai branch of Tohoku University for JMTR Utilization for the post irradiation experiments.

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