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Formation mechanism of clustered small loops (rafts) in fission-neutron irradiated Mo at high temperatures

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Abstract

Clustered small loops were observed, by transmission electron microscope, in fission-neutron irradiated molybdenum at high temperatures and referred to as a raft. The groups of small dislocation loops (rafts) were observed at one side of the dislocations or inside of the large interstitial loops. The small loops in a raft aligned in rows. Burgers vector of the small loops was the same as that of the dislocation or large loop. The nature of small loops in the rafts was a vacancy type. It was also observed that the dislocation dipoles were just chopping into small loops. Therefore, formation mechanism of the rafts is as follows: Interstitial atoms aggregate and form loops, which grow to large convoluted loops. The dislocation dipoles on the convoluted loops are chopped off to form the rows of small loops. By this mechanism, the small loops in the rafts have to be opposite type in nature against the large interstitial loops.

1. Introduction

The damage microstructure in neutron-irradiated molybdenum has been extensively investigated because the refractory metals are one of the candidates for first wall materials in fusion reactors [1,2].

A large number of dislocations, dislocation loops and voids were formed by the high temperature irradiation. Especially around 400–600°C irradiation, clusters of small dislocation loops were formed in fission-neutron irradiated molybdenum and referred to as rafts from the form [1,2]. Brimhall and Mastel [1] reported that the rafts were formed by the combination of prismatic glide and conservative climb of small loops with an elastic interaction between them.

In the present work, formation mechanism of the rafts in Mo were studied by transmission electron microscopy and some proofs against the above formation mechanism of the rafts were found.

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2. Experimental procedure

Pure molybdenum specimens (VP grade from MRC Inc.) were fully annealed in ultra-high vacuum ($<5 \times 10^{-7}$ Pa) before the irradiation. The neutron irradiation was carried out at bulky state in the Joyo experimental fast reactor of Japan. After the irradiation, the specimens were electro-polished and observed by electron microscope.

3. Experimental results and discussion

The damage structure are shown in a left part of Fig. 1 for the irradiated molybdenum up to the fluence of 7.9×10^{23} n/m^2 at 400°C. Large loops with complex structure, 50–100 nm in diameter, were distributed heterogeneously in the specimen. The structure of the loops was not clear due to the small size. In general, it is difficult to determine whether the loops are interstitial type or vacancy type for complicated loops due to their shape, however, by the inside–outside method, the nature of some loops could be determined if the size of the loops is large enough. At further irradiation up to 8.1×10^{24} n/m^2 at 400°C, the groups of small dislocation loops, which are called as the rafts, voids and large

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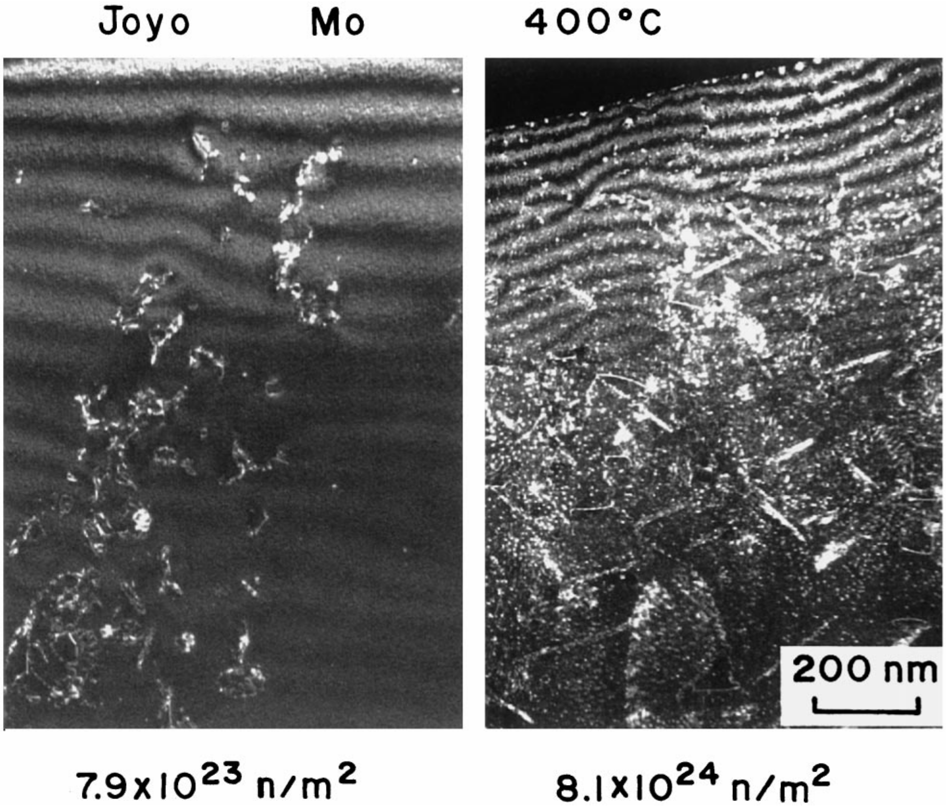

Fig. 1. Microstructures of molybdenum irradiated at 400°C. Complicated loops were observed: 7.9×10^{23} n/m^2. Complicated loops, rafts and voids were observed: 8.1×10^{24} n/m^2.

complicated loops or dislocations are observed with high density as shown in a right part of Fig. 1. The rafts would grow from the complicated loops as mentioned later.

The large complex loops and rafts at 400°C irradiation grow to larger ones at 500°C irradiation as shown in Fig. 2. For larger ones, the nature of the loops could be determined by the inside–outside method. Many small loops exist near the dislocations and at one side of them as A and B in Fig. 2. The small loops in many cases are in the rows from dislocations. At some parts in the specimens, long dislocation dipoles from the dislocations are observed as C and D in Fig. 3. The chopping into small loops of dislocation dipoles is also observed (Fig. 6(a)). The dislocations decorated by small loops were usually observed in neutron irradiated Mo [1,2].

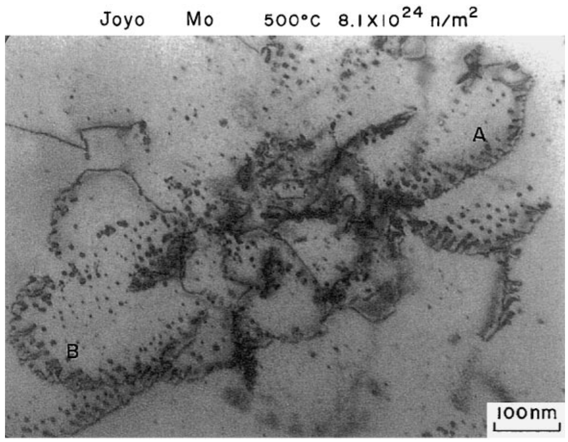

Fig. 2. Microstructures of Mo irradiated at 500°C to a fluence of 8.1×10^{24} n/m^2.

It was difficult to find the isolated large complicated loops as shown in Fig. 2 except in special part of the specimen at 500°C irradiation due to intersection of the loops during irradiation and cutting the loops by the surface during thinning process of the specimen for electron microscopic observation. At a comparatively thicker part and near grain boundary, at where the loop growth was suppressed by migration of point defects to the boundary, we can find large isolated loops, above 250 nm in diameter, with small loops inside of them as shown in Fig. 4. The images were obtained at some abnormal transmission conditions of electrons because

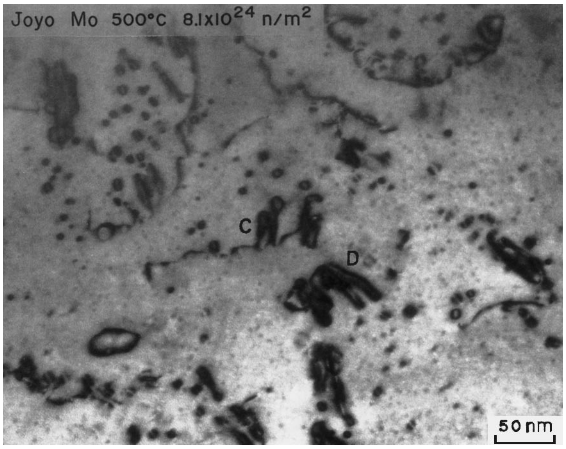

Fig. 3. Examples of parallel dislocation dipoles in irradiated Mo at 500°C.

of a sufficiently thick part of the specimen. In this image condition the voids were not observed clearly at thicker part of the specimen. But at void imaging condition, the high density of voids was observed clearly at thinner part of the same specimen. In this thicker part, the voids were also formed in high density because the specimen was irradiated at bulky state. Nature of these large loops was determined as interstitial type by the inside–outside method. The analysis was performed on the loops shown in Fig. 4 which is a set of micrographs with the reversal of ***g*** vectors with the same sign of deviation parameter *s*. The planes of large loops were determined by tilting the specimen.

A set of micrographs with reverse ***g*** vectors is shown in Fig. 5 for the small loops and large loop. The small loops inside of the large loop exhibit opposite size change to the outside large loop. The small loops forming a raft in the large loop are on the same plane because the small loops have almost the same double arc image direction one another. Therefore these small loops are vacancy type in nature if the small loops are on parallel plane to that of the large loop. It was difficult to obtain the loop plane of the small loops, in this case, because the loops are small and located at the thick part of the specimen.

However, it is inferred from the fact observed below that the plane of the small loops is the same as that of each large loop. The dislocation or large loop and small loops accompanied with it had the same Burgers vector ***b***. One of the example for $\boldsymbol{g}\cdot\boldsymbol{b} = 0$ is shown in Fig. 6. In Fig. 6(a) the dislocation and small loops are observed at bright field and in Fig. 6(b) and (c) the same area is observed with $\boldsymbol{g} = [1\,\bar{1}\,0]$ and $\boldsymbol{g} = [\bar{1}\,0\,1]$, respectively, at dark field. In Fig. 6(c) the dislocation and small loops disappeared at the same time by the condition of $\boldsymbol{g}\cdot\boldsymbol{b} = 0$. Small and weak dot images in a back ground are the

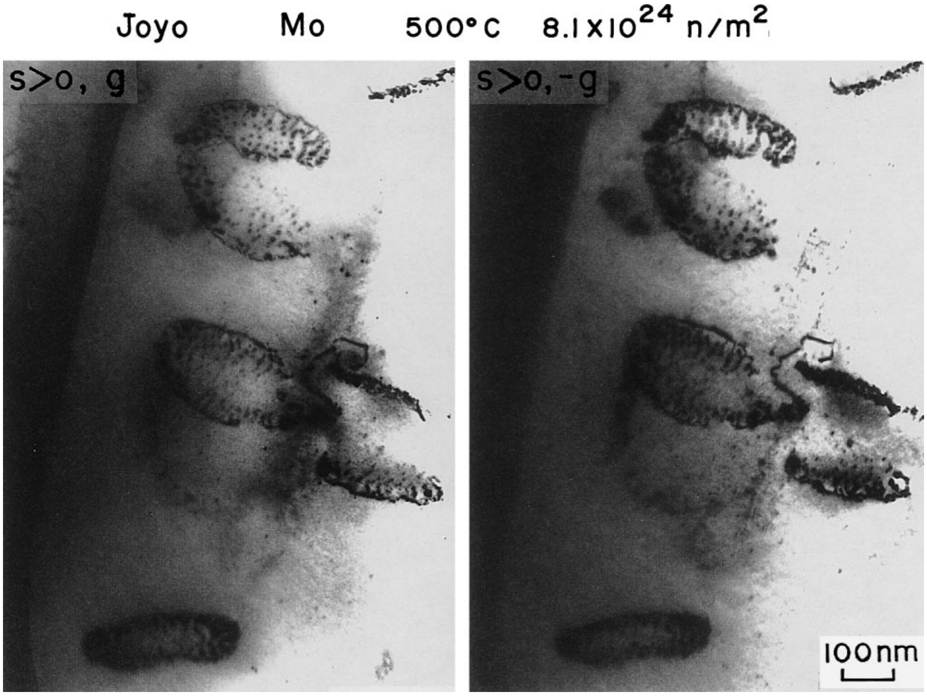

Fig. 4. A set of micrographs with reverse ***g*** vectors for the large loops with positive deviation parameter *s*. The large loops change the size by the reversion.

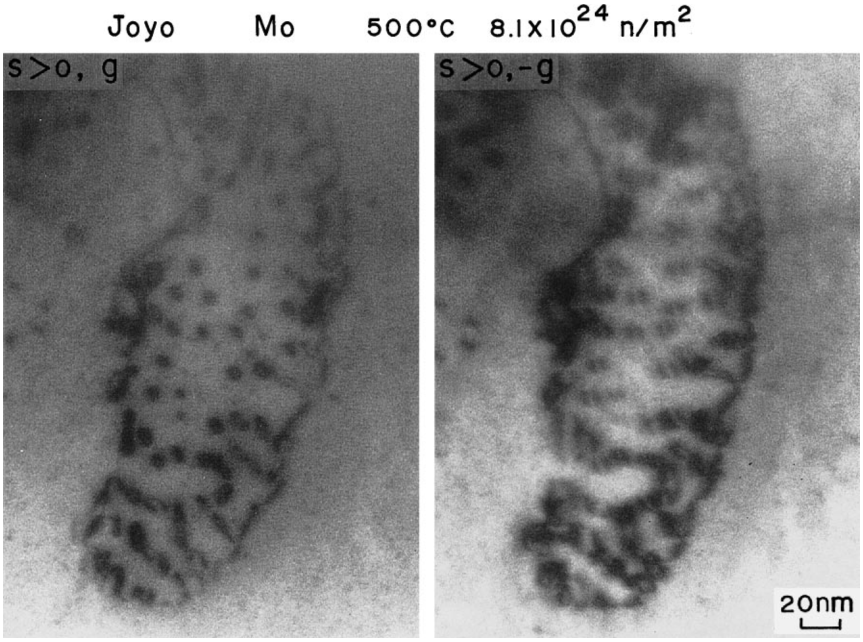

Fig. 5. A set of micrographs with reverse ***g*** vectors for the large loop and small loops.

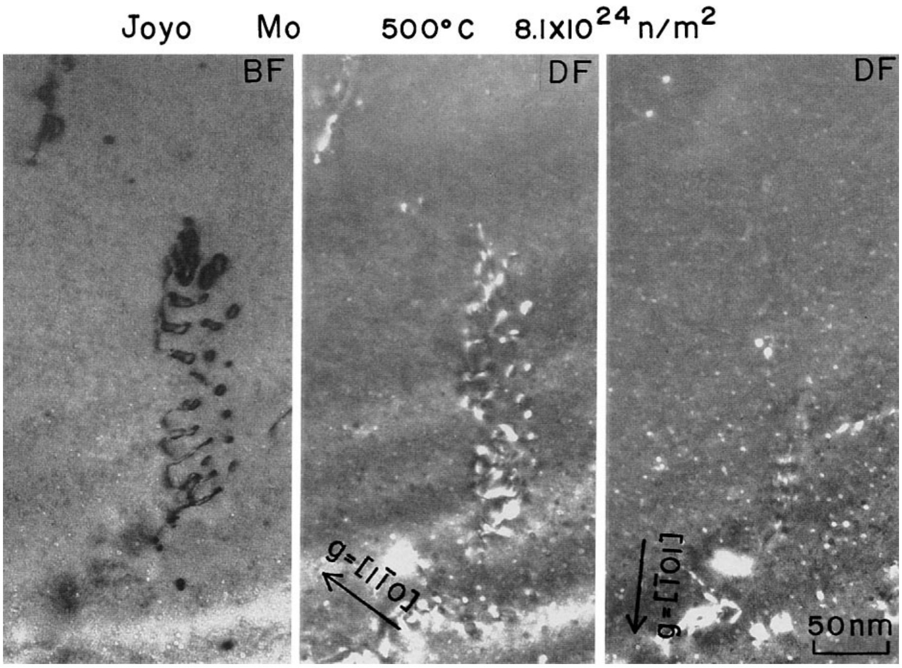

Fig. 6. Small loops (rafts) and parent dislocation. (a): bright field image. (b) and (c): dark field images.

voids. Further indication for the parallel plane would be that the raft and the large loop have a layer structure in the plane of the large loop. When the rafts together with the large loops were observed from their sides, they had rather narrow widths.

As mentioned before, Brimhall and Mastel [1] observed the rafts in neutron-irradiated Mo at high temperatures. At 400–600°C irradiation they observed that there were complex structures consisting of large loops surrounded by small loops and that the small loops as the rafts had the same Burgers vector one another, though the nature of the small loops was not determined. However, they claimed that the small loops in the rafts are interstitial type, similar to those observed by Eyre et al. [3] who analyzed the nature of the small loops ('black spots') in neutron irradiated Mo. It must be clearly noted that Eyre et al. analyzed for the small loops formed in the matrix on early stage of irradiation but not for the small loops forming the rafts. Their black spots formed first in the specimen subsequently grow to our large interstitial loops at high irradiation doses. The large loops irradiated above 600°C were interstitial in nature [1]. Furthermore they suggested that the irregular configurations of the large loops or dislocations are due to the coalescence of the mobile small loops to form the large loops and the rafts are formed by a combination of prismatic glide and conservative climb of the small loops.

Their assertion about the nature of the small loops forming the rafts has important discrepancy with the present experimental result and could not explain the rows of small loops.

In irradiated metals it has been pointed out by many authors that the damage resolvable by TEM is inhomogeneously distributed near the dislocations especially at high temperatures [4,5]. They discussed the origin of the damage structures. Some of the results are interpreted with sink effect of dislocations by the dislocation bias for interstitials and vacancies [4,5]. For these cases, as the clusters are nucleated by super saturation of the point defects due to depletion of another point defects, they would be formed with various Burgers vectors against that of the parent dislocation [4]. It is not the case in the present experiment because the small loops in the raft have the same Burgers vector with the parent dislocation.

The complicated loops were also observed in electron irradiated Mo in high voltage electron microscope [6]. The loops have a characteristic structure containing many petalous parts inside the loops and are interstitial type in nature. Formation mechanism of the petalous parts of the loops has been proposed by Kiritani et al. [6]. The flower-like shape of their loops is explained by directional flow of interstitial atoms from the side of compression to that of expansion along the strain field gradient around an edge dislocation. If a part of a straight dislocation have a curved shape, the tendency will be strengthened more and more by faster growth of the concave part than the convex part. The difference of the growth speed during irradiation is attributed to the geometry of the volume from which interstitial atoms flow into each segment of the dislocation [6]. From the present results and the model of Kiritani et al., the formation mechanism of the raft is explained as follows. As observed by many authors, in neutron irradiated Mo at high temperatures the interstitial loops are formed and become larger during irradiation by absorbing the interstitial atoms. During growth the loops become convoluted ones by the mechanism of Kiritani et al. [6]. Therefore, even they are comparatively small interstitial loops they show complicated structure in them (7.9×10^{23} n/m^2, 400°C). By further irradiation (8.1×10^{24} n/m^2, 400°C), they become larger and visible as the rafts. At higher irradiation temperature, 500°C, various raft like structures are easily observed because they become larger. The dislocation dipoles (petalous parts) of the convoluted loops are chopped off to form the rows of small loops. Therefore, the small loops as the rafts are formed inside of the large loops or one side of the dislocation lines which are formed by intersection of the large loops or specimen surfaces etc. The small loops have to be vacancy type in nature as determined by present experiment by geometrical reason if the large loops are interstitial type. Then the small loops forming the raft and the large parent dislocation loop must have the same Burgers vector each other. This is also good agreement with the present experimental results.

It is noted that small loops as the rafts were also observed at the inside of helical dislocations. If the above formation mechanism of the rafts are applied to the dislocations forming the helixes, the rows of small loops must be formed inside of the helixes.

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