ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна

E15-98-364

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VOID FORMATION DURING ANNEALING OF NEUTRON AND ALPHA-PARTICLE IRRADIATED MOLYBDENUM

Submitted to «Journal of Nuclear Materials»

1998

Introduction

Numerous theoretical and experimental works on vacancy voids forming in metals and alloys during high-temperature irradiation is connected with establishment of the regularities and mechanisms of its development. The question on the mechanism of vacancy void nucleation is far from its solution that is caused by details of its formation under simultaneous creation and mobility of Frenkel pairs. The basis of the mechanism of the vacancy voids nucleation and growth is in the supposition that the required vacancy oversaturation of the grain body at irradiation is provided at the expense of a preferred absorption of interstitial atoms by dislocation [1]. From the other hand, in the process of irradiation the dislocation loops (of interstitial type, mainly) are generated.

Among the mechanisms of their thermal growth, the mechanism of their growth at the expense of sliding and by way of emitting the vacancies by them, are taken into account. Although the latter mechanism is rather difficult to fix experimentally, it also can serve as a reason for the oversaturation of the crystal lattice by vacancies, which is required for nucleation and growth the vacancy voids.

In this connection it is interesting to study the nucleation and development of vacancy void under conditions when the interstitial atoms are not generated though they can be present as dislocation loops. Similar model conditions can apparently result in postirradiation annealing of defect structure produced at low-temperature irradiation.

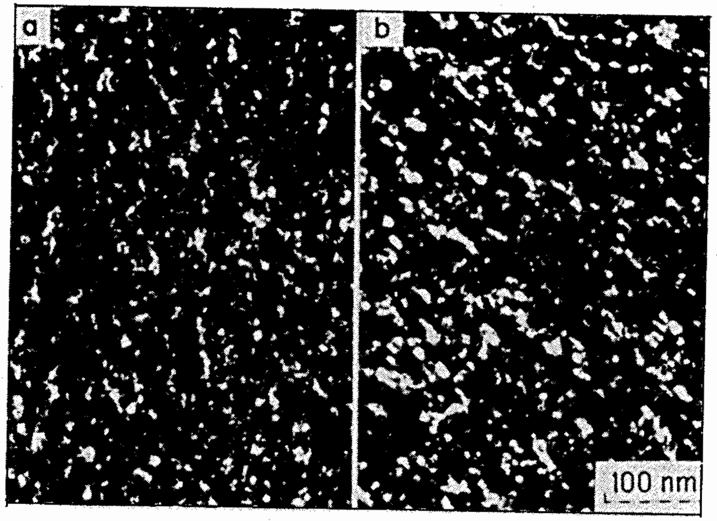
The object of the present paper was establishment of the fact of formation and peculiarities of thermal evolution of vacancy voids from the defect structure created during low-temperature irradiation of molybdenum by high-energy alpha-particles and fission neutrons.

Experiment

The molybdenum of 99.97% purity with 100 mm thickness irradiated by fission neutrons and alpha-particles "throughout" irradiation (i.e. without helium doping) with 50 MeV initial energy at temperature not greater than 100°C up to $7 \cdot 10^{-2}$dpa was taken. Postirradiation annealing was carried in 10^{-3}Pa vacuum in 100-1100°C temperature range with 100°C-step. Electron-microscopic investigations were performed on JEM-100CX microscope.

Results and discussion

After neutron and alpha-particle irradiation in molybdenum were formed morphologically identical defect structures that consist of vacancy aggregations and interstitial dislocation loops (Fig.1). Total numerical densities of the observed



aggregations of radiation defects and histogram of size distributions appeared to be similar.

The present irradiation dose produces the defect density so high and their overlapping so essential that the defect identification appeared to be extremely difficult. Nevertheless, the investigations performed on the samples irradiated up to the less (10^{-2}dpa) damage level [2] made possible to set that the most of the defects with size less than 5nm are vacancy aggregations; larger ones are interstitial aggregations. Apparently, it should be expected that this tendency is kept after the given irradiation dose, too.

Up to 200°C annealing the defect structures in neutron and alpha-particle irradiated molybdenum are the same as in state following the irradiation. Thermal stability of the radiation defect aggregations observed in TEM in the given temperature range (to 200°C) was confirmed by observation in the electron microscope column equipped with hot-stage. Some enlargement of dislocation loops with simultaneous disappearance of the defect aggregations up to 2 nm in size begins at 300°C and higher. The most typical evolution of the defect structure takes place at temperature more than 500°C (Fig.2 and 3). At 800°C practically complete disappearance of the defect aggregations less than 5 nm in size and formation of vacancy voids with average size of 1.4 nm (Fig.4 and 5) occurs. The treatment that the observed voids are vacancy ones is based on the following experimental facts:

-formation of voids from the gas impurities diluted in molybdenum is excluded as far as they are not observed in the unirradiated material,

-as it is seen from Fig.6, change of relative volume of voids in alpha-particle irradiated molybdenum with annealing temperature has the bell shape. In case of bubbles formation from the atom of unsoluble gases, helium for example, swelling is constantly increased with temperature rise [3],

-helium bubbles [4] are mainly formed on dislocation loops. In the present work voids are not observed on the dislocation defects.

Change of numerical density and average size of dislocation loops depending on temperature are shown in Fig.7.

One should note the following circumstances indicating the similarity and distinctions of thermal evolution or the defect structures formed at low-temperatureof neutron and alpha-particle molybdenum irradiations:

- in the range of 100-900°C annealing temperature the change of dislocation loop numerical density does not practically depend on the irradiation type. The difference is observed in temperatures up to complete annealing only. So in case of neutron irradiation molybdenum the dislocation loops are annealed at 1000°C while in case of alpha-particle irradiation at 1100°C.

-annealing temperature at which the visible in TEM voids appear is about 800°C for the both cases, their numerical density in alpha-particle irradiated molybdenum is three times higher than that in case of neutron irradiation.

-peculiar differences in thermal evolution of the defect structures in neutron and alpha-particle irradiated molybdenum are observed in the change of size

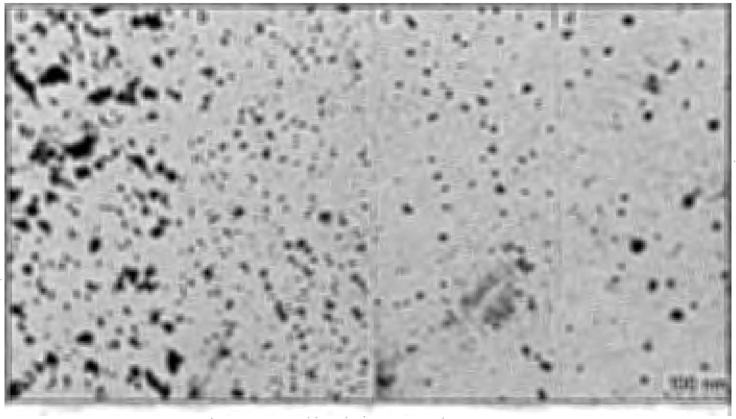


Fig.2. Typical change of dislocation loop structures of neutron irradiated molybdenum as a result of isochronous annealings:
a- 500°C, b-700°C, c- 800°C, d- 900°C.

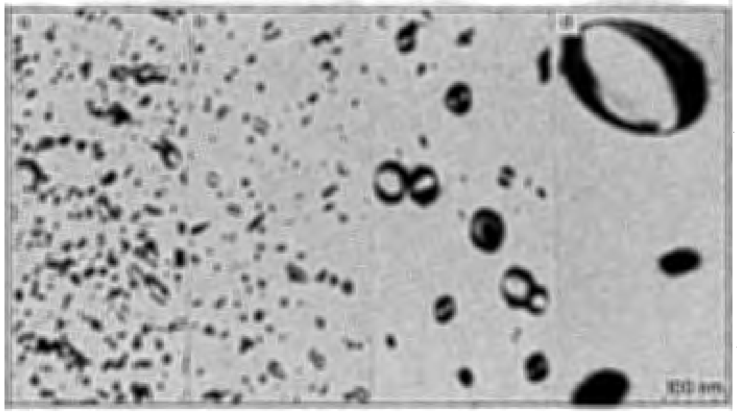


Fig.3. Typical change of dislocation loop structures of alpha-particle irradiated molybdenum as a result of isochronous annealings:
a- 500°C, b-700°C, c-800°C, d- 1000°C

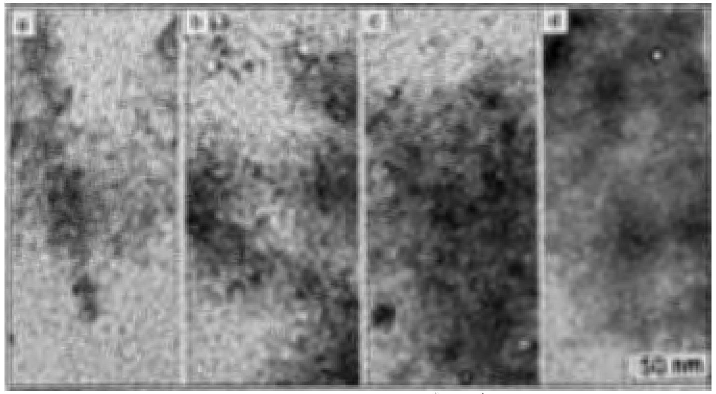


Fig.4. Vacancy voids in alpha-particle irradiated molybdenum after one-hour isochronous annealings:
a- 800°C, b- 900°C, c- 1000°C, d- 1100°C.

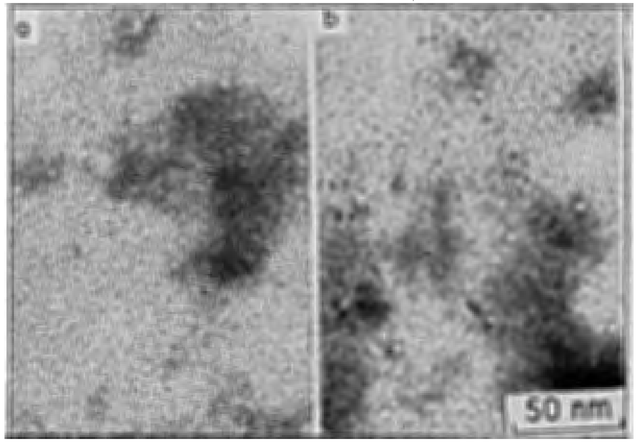


Fig.5. Vacancy voids in neutron irradiated molybdenum after one-hour isochronous annealings:
a- 800°C, b- 900°C.

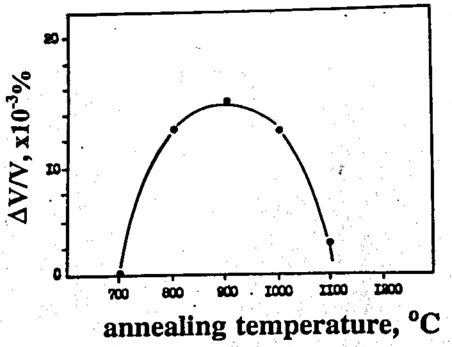


Fig.6. Change of relative volume of vacancy voids in alpha-particle irradiated molybdenum in the process of isochronous annealings.

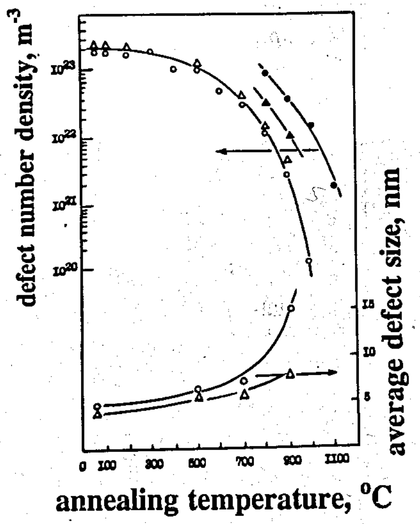


Fig.7. Change of numerical density void average size of radiation defect aggregations observed in TEM after irradiation up to $7\cdot10^{-2}$dpa damage level as a result of one-hour isochronous annealings: O,Δ- dislocation loops; ●,▲- vacancy voids for the case of alpha-particle and neutron irradiations, respectively.

distribution histograms of dislocation loops. Thus, in alpha-particle irradiated samples the rate of dislocation loop growth in temperature range more than 700°C is significantly higher, larger ones up to 300nm being reached, while in case of neutron irradiation the maximum loop size is not more than 20nm.

In comparison with neutron irradiation alpha-particle one causes, higher rates of helium atom accumulation at the values of the larger cross-section of defect formation and (α,nα) type nuclear reactions and alpha-particle back scattering. In this case the effect interaction of helium atoms with the isolated vacancies and vacancies in cascades give their greater preservation in annealing process. The latter causes the observed significant growth and survival of interstitial loops in alpha-particle irradiated molybdenum in temperature range from 700 to 1100°C.

The experimentally observed equality between number of vacancies in voids and number of interstitial atoms in dislocation loops in wide range of annealing temperature makes possible to suppose that in formation of vacancy voids is determined by their nucleation on HemVn-complexes and by the growth in the conditions of the oversaturation of the crystal lattice by vacancies, at the expense of their intensive emission by the dislocation loops growing at high annealing temperature.

Thus, the present results give possibility to make the following inclusions:

-nucleation and development of vacancy voids take place during postirradiation annealing of molybdenum irradiated by fission neutrons and high-energetic alpha-particles at low (60°C) temperature;

-it is shown that evolution of molybdenum defect structure as a result of annealings in 100-1100°C temperature range depends on irradiation type. Thus, in alpha-particle irradiated samples the rate of dislocation loop growth is significantly higher than in neutron irradiated ones.

-there was an assumption that the nucleation and growth of the vacancy voids resulting from the postirradiation annealing of the dislocation -loop structure Mo, formed at low temperature irradiation by neutrons and alpha-particles is determined by thermal emission of vacancies by dislocation loops.

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Received by Publishing Department
on December 21, 1998.