ROLE OF DISLOCATION LOOPS IN THE DEVELOPMENT OF VACANCY VOIDS

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This paper presents the results of a transmission electron microscopy (TEM)-investigation of the effect of post-irradiation annealing on the structure of molybdenum and nickel doped with helium atoms that was irradiated with high-energy α-particles and fission neutrons. It is shown that:

- The number of interstitial atoms in dislocation loops and the number of vacancies in helium bubbles increase as annealing temperature and time increase.
- The growth of helium bubbles on the grain boundary is mainly observed on the side of the grain body where there are dislocation loops.
- Post-irradiation annealing of dislocation loops formed by 50-MeV α-particles and fission neutrons at 60°C results in the growth of vacancy voids.

Thus, high temperature post-irradiation annealing or irradiation at high temperature (more than $0.3T_{melt}$) constitutes the conditions under which dislocation loops are powerful sources of vacancies. Consequently, if in the crystal structure of the irradiated material there are He_mV_n groups, vacancy oversaturation caused by the thermal emission of vacancies by radiation-induced dislocation loops may be the main driving force for the nucleation and growth of vacancy voids.

Introduction

Radiation swelling, one of the last discovered radiation effects, has been known since 1967 [1] when it was found that irradiation of a material makes its volume (geometrical dimensions) grow. The effect is accounted for by there appearing and developing vacancy voids (VV) in the irradiated material. This unexpected phenomenon, which results in the strenth of reactor building materials becoming catastrophically low, has attracted considerable interest.

Unfortunately, up to date, theories of swelling are unable to quantitatively predict the value of radiation swelling for a wide range of materials and irradiation conditions.

According to up-to-date concepts, the nucleation and growth of VV in irradiated material has oversaturation with vacancies as a driving force. The oversaturation is assumed to result from the fact that the ways vacancies and interstitial atoms interact with dislocations are different, dislocations showing preference to interstitial atoms [2].

However the asumption that dislocations show preferance to interstitial atoms has not been confirmed experimentally. The parameter of preference is difficult to calculate. It depends on the elastic fields of discharge channels , the degree of assimilation of point defects, the barriers to the absorption of point defects on the surface of discharge channels. Therefore, there a need to search for other sources of oversaturation of the crystal lattice with vacancies. In our view, it is reasonable to suppose that the dislocations and dislocation loops of interstitial and vacancy types produced during irradiation may be considered as such sources.

This paper presents the results of a TEM-investigation of the effect of postirradiation annealing on the structure of molybdenum and nickel were doped with helium atoms and also was irradiated with high-energy α-particles and fission neutrons.

Results and discussion

Molybdenum and nickel were chosen as the material to be investigated. The irradiation with neutrons was performed using the WWR reactor; the irradiation with 50 MeV α-particles was carried out on a cyclotron at a temperature of about 60°C.

The postirradiation isothermal and isochronous annealing was performed in a $5x10^{-3}$ Pa vacuum in the temperature range from 100 to 1100°C.

If the critical conditions for dislocation loops to coalesce during their sliding and crawling, which result from the elastic interaction between them, are not met, then dislocation loops may grow by emitting vacancies. For those conditions to be realized, dislocations must be prevented from transferring due to sliding and crawling.

Therefore, dislocation loops can be seen through a TEM to emit vacancies if the following requirements are met:

1. The sliding and crawling of dislocation loops must be eliminated or decreased.
2. The annealing temperatures must be high; sufficiently large loops must be dealt with. This will allow one to decrease the error in measuring the variations in the diameter of dislocation loops and, consequently, the error in measuring the number of the interstitial atoms in them. The dimentions of loops must not be larger than the thickness of the TEM-object (100 nm), their density in the specimen must be sufficient from the statistical standpoint.
3. In the specimen there must be powerful discharge channel to capture the vacancies emitted by dislocation loops in order to prevent them from being captured by the neighbouring loops. Otherwise, the expected increase of interstitial atoms in loops will not be observed.

Those requirements are likely to be met in specimens doped with helium atoms. In such specimens the appropriate annealing results in dislocation loops of the interstitial type and helium bubbles being formed. The dislocation loops serve as sources of vacancies and the bubbles as discharge channels for them.

Indeed, in Mo specimens doped with helium up to 4.5x10^{-2} at% and annealed at 1000^0 C, dislocation loops and helium bubbles are formed whose density and dimentions comply with the requirements of paragraphs 2, 3.

The presence of helium bubbles on dislocation loops prevent them from conservative motion, and the high density of helium bubbles in the space between dislocation loops provides a lot of efficient and thermally stable discharge channels for vacancies.

The specimens were isothermally annealed at temperatures of 1000 and 1100°C for 1, 5, 10, 50 and 100 hours.

As shown in Fig. 1, the longer the annealing time is, the larger the size of dislocation loops are and the lower their density is. The TEM analysis carried out showed that dislocation loops, which are mainly of the interstitial type, are situated in planes of the {111} type and have Burgers's vector of the 1/2a <111> type.

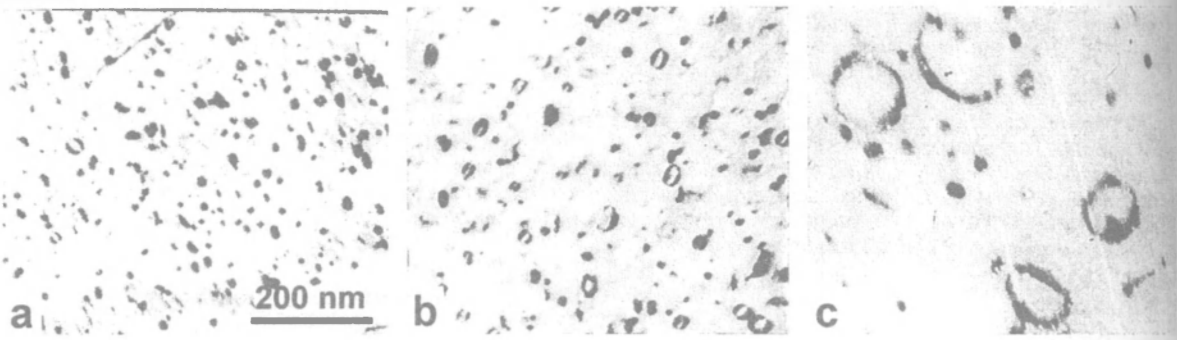

Fig.1. Development of dislocation loops in Mo doped with helium (4.5x10^{-2} at.%) during postradiation annealing at 1000°C, the annealing time being 1 hr (a), 10 hrs (b), 50 hrs (c)

On this basis, the number of interstitial atoms in dislocation loops was calculated as a function of annealing time at 1000 and 1100^{0}C (Fig. 2). Fig. 2 shows that the number of interstitial atoms in dislocation loops is a linear function of annealing time. As seen, the number of interstitial atoms increases at much larger rate at 1100^{0}C than at 1000^{0}C. Moreover, it is significant that the increases in the number of interstitial atoms in loops closely correlate with the increases in the number of vacancies in helium bubbles.

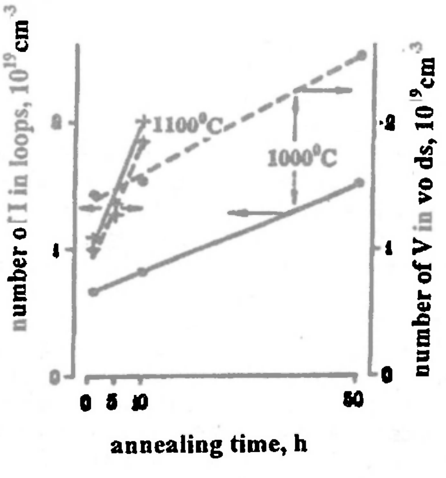

Fig.2. The number of interstitial atoms in dislocation loops and the number of vacancies in helium bubbles in Mo as a function of postradiation annealing time at 1000 and 1100°C.

Thus, these results testify that interstitial loops grow by emitting vacancies. Moreover, estimates of the activation energy for loop growth give the value about 3eV, which is close to the energy required for producing vacancies in molybdenum (3.3 eV [3]).

In another series of experiments in which the peculiarities of the growth of helium bubbles near grain boundaries in helium-doped Ni were studied, data were obtained that testify that interstitial dislocation loops are quite powerful sources of vacancies.

Near grain boundaries in Ni specimens, zones were created that were depleted in dislocation loops, and that contained helium bubbles (Fig. 3a). On further annealing, as interstitial dislocation loops in the grain body keep on growing, the diameter of helium pores is seen to increase not near grain boundaries (which are commonly accepted powerful sources of vacancies) but in the opposite region, which borders on the area of the grain body where there is high density of interstitial dislocation loops (Fig. 3b,c). We think that in this case the flow of vacancies from the area of the grain body where there are dislocation loops prevails over the flow of vacancies from the grain boundaries.

Thus, all the above-stated experimental results, in our opinion, strongly testify that interstitial and, of course, vacancy dislocation loops are powerful sources of vacancies and, therefore, may be responsible for the oversaturation of the crystal lattice with vacancies at appropriate temperatures. And, if there are centres for them to condense on, particularly He_mV_n, this may result in the nucleation and growth of vacancy voids. So, what is of fundamental importance, the process concerned can be accounted for without using the hypothesis of so-called preference

It is known that owing to interstitial atoms being highly mobile at ordinary temperature at which irradiation is carried out, interstitial atoms in aggregations are in the form dislocation loops. As a

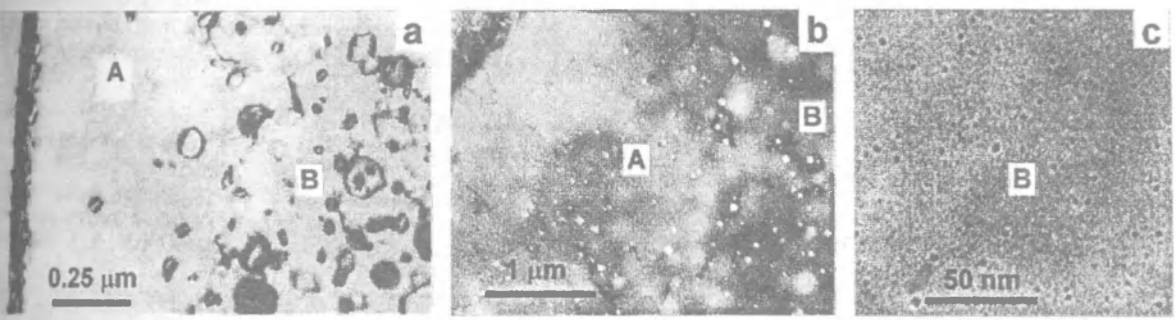

Fig.3. Dislocation loops in Ni doped with helium up to $2x10^{-2}$ at.% and annealed at 600^0C for 1 hr.(a). Development of helium bubbles in Ni ($2x10^{-2}$ at.% He) as a result of annealing at 800^0C: b- in region A lying near the grain boundary, which is poor in dislocation loops (see Fig.3a); c- in the grain body (region B, see Fig. 3a).

result, during postradiation annealing, they are in no way involved in the evolition of dislocation loops and much less in the formation of vacancy voids.

This and above-stated results suggest that vacancy porosity may be formed in specimens irradiated at low (room) temperatures and then annealed.

For this purpose, Mo (99.97%) in the form of 100 mkm foil was irradiated with neutrons in a nuclear reactor at about 60^0C and with 50 MeV α-particles, ensuring shooting through, up to $7x10^{-2}$ dpa.

The postirradiation annealing was performed in a $5x10^{-3}$ Pa vacuum in the temperature range from 100 to 1100^0C, the step being 100^0C.

Immediately after irradiation of specimens with neutrons and α-particles there appears a morphologically identical imperfect structure in the form of vacancy clusters/loops and interstitial dislocation loops (Fig. 4a,b). The average density of defects and their size distribution also proved to be similar. Defects of size less than 5 nm were mainly of the vacancy type whereas dislocation loops of larger size were of the interstitial type.

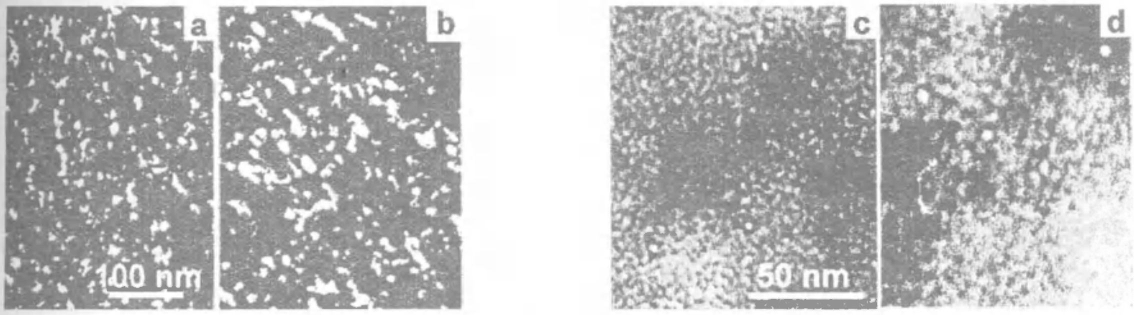

Fig.4. Imperfect structure in Mo on irradiation at 60^0C, a - with fission neutrons ($7x10^{-2}$ dpa), b - with 50 MeV α-particles ($7x10^{-2}$ dpa). Vacancy voids in Mo: c - irradiated with neutrons and annealed at 900^0C, d - irradiated with α-particles and annealed at 900^0C

The imperfect structure begin evolving at 300^0C: defects of size about 2 nm disappear. The marked evolution of the loop structure occurs at temperatures higher than 500^0C. In the temperature range from 800 to 1100^0C voids are initiated and begin to grow (Fig. 4c,d). The way in which the characteristics of voids depend on annealing temperature, that is, that the volume of voids increases at 900^0C, and that they are practically annealed at 1100^0C, points to the fact that they are of a vacancy nature.

Comparison of the increase in the number of interstitial atoms in dislocation loops and the increase in the number of vacancies in voids indicates that interstitial dislocation loops are the sources of vacancies for voids to grow.

He_mV_n complexes seem to be the centres at which voids are nicleated. In the case of neutron irradiation, helium accumulates as a result of (n,α) reactions. Under irradiation with α-particles helium accumulates both owing to the corresponding channels of nuclear reactions of the $(\alpha,x\alpha)$ type and owing to the backward scattering of the bombarding α-particles in the sample. Therefore, there being more helium atoms in Mo specimens irradiated with high-energy α-particles as compared to those irradiated with fission neutrons seems to account for the larger density and sizes of voids observed in the case of irradiation with α-particles.

Thus, the above results not only point to the possibility of the growth of vacancy voids during annealing of the imperfect structure formed under irradiation at low temperature but also give strong evidence of emission of vacancies from dislocation loops, which grow at the appropriate temperatures.

Conclusions

The presented experimental results show that:

- the number of interstitial atoms in dislocation loops and the number of vacancies in helium pores increase as annealing temperature and time increase;

- the growth of helium bubbles on the grain boundary is mainly observed on the side of the grain body where there are dislocation loops;
- the postirradiation annealing of the structure with dislocation loops which was formed by irradiating with 50 MeV α- particles and fission neutrons at 60^0C results in the growth of vacancy voids.

The foregoing gives grounds to state that high temperature annealing or irradiation at high temperature (more than $0.3T_{melt}$) constitutes the conditions under which dislocation loops are powerful sources of vacancies. Therefore, on condition that in the crystal structure of the irradiated material there are groups of the He_mV_n type, vacancy oversaturation accounted for by thermal emission of vacancies by dislocation loops of radiation origin may be the main driving force for the nucleation and growth of vacancy voids.

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