Journal of Nuclear Materials 385 (2009) 228–230

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Radiation-induced reduction in the void swelling

V.I. Dubinko *, A.G. Guglya, E. Melnichenko, R. Vasilenko

NSC Kharkov Institute of Physics and Technology NAS of Ukraine, Akademicheskaya Str.1, Kharkov 61108, Ukraine

ARTICLE INFO

PACS:
61.72.Ji
61.72.Qq
61.80.Az

ABSTRACT

Vacancy voids have been produced in Ni by 1.2 MeV Cr ion irradiation at 873 K up to the ion fluence of 10^{21} m^{-2}. Subsequent irradiation of specimens containing voids at 798 and 723 K has resulted in the reduction of the void size and number density. Accordingly, the void swelling has decreased by a factor of ~5. The experimental results are explained in the framework of an original model taking into account the interaction of voids with radiation-induced excitations of atomic structure such as focusing collisions and long-propagating self-focusing breathers.

1. Introduction

In the conventional theory of radiation damage, it is assumed that the main effect of irradiation on materials is related to the formation of Frenkel pairs of vacancies and self-interstitial atoms (SIAs) and their clusters. The difference in the ability to absorb vacancies and SIAs by primary or radiation-induced extended defects (EDs) is thought to be the main driving force of microstructural evolution under irradiation. On the other hand, recovery from radiation damage is thought to be driven exclusively by thermal fluctuations resulting in the evaporation of vacancies from voids (void annealing) or dislocations (thermal creep) and in the fluctuation-driven overcoming of obstacles by gliding dislocations (plastic strain). These recovery mechanisms can be efficient only at sufficiently high temperatures. However, there is an increasing evidence that the so-called thermally activated reactions may be modified under irradiation. Thus, results of molecular dynamics (MD) simulation [1] have shown that vacancies can be emitted from voids not only by thermal fluctuations but also by the collision events in the vicinity of voids, which result in a biased formation of vacancies due to the lower energy barrier involved. A subsequent diffusion of the ejected vacancies away from the void under certain conditions may result in the radiation-induced void shrinkage and reduction in the void swelling [2,3]. There is also some evidence from MD simulations that a biased formation of vacancies occurs also in the vicinity of the dislocation cores [4], which results in irradiation creep based on the radiation and stress induced difference in emission of vacancies from dislocations [5].

These are examples of the mechanisms based on the *radiation-induced production of Schottky defects* [6,7], which often act in the opposite direction as compared to the mechanisms based on *Frenkel pair production in the bulk*.

In this work, the phenomenon of radiation-induced void dissolution is investigated by means of irradiation of nickel samples with 1.2 MeV Cr ions at different temperatures. The experimental results are discussed in the framework of the original model taking into account radiation-induced excitations of atomic structure such as focusing collisions and long-propagating breathers.

2. Experimental procedure and results

Four Ni foils of 100 micron thickness have been irradiated with 1.2 MeV Cr ions at 873 K up to the total ion fluence of 10^{21} m^{-2}, which corresponds to the irradiation dose of 25 displacements per atom (dpa) at the dose rate of $K = 7 \times 10^{-3}$s^{-1}. Examination of two control samples in transmission electron microscope TEM-100 has revealed formation of a high number density (~10^{21} m^{-3}) of voids of 40–50 nm in diameter. The third and the fourth foils have been irradiated subsequently up to the ion fluence of 10^{21} m^{-2} at two different temperatures, 798 and 723 K, respectively. The resulting microstructure is shown in Fig. 1, from which it is evident that the irradiation at lower temperatures has made the voids to decrease in size. The quantitative analysis of the void number diameters and swelling confirms this conclusion. According to Fig. 2, the void swelling has decreased by a factor of ~5.

3. Discussion

A few previous experiments have indicated that voids can shrink with decreasing temperature (or increasing dose rate) after they have been formed under more favorable conditions [3,8,9]. According to Steel and Potter [3], voids formed during 180 keV Ni$^+$ ion bombardment of Ni at 923 K shrink rapidly when subjected to further bombardment at temperatures between 298 and 823 K.

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* Corresponding author. Tel./fax: +380 573 351781.
E-mail address: vdubinko@mail.ru (V.I. Dubinko).

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doi:10.1016/j.jnucmat.2008.11.028

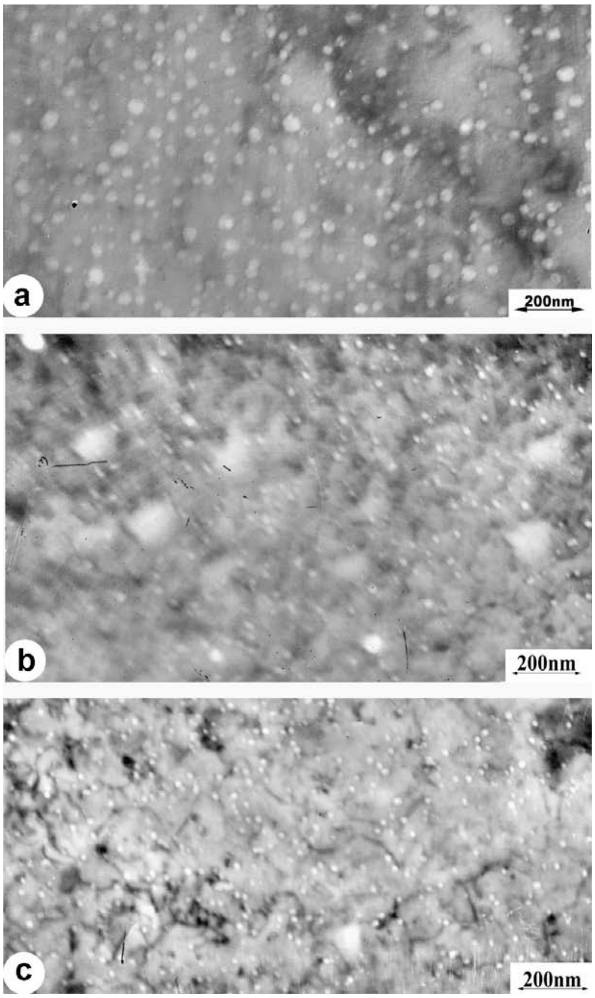

Fig. 1. TEM micrographs showing voids in (a) Ni irradiated with Cr ions up to 25 dpa at 873 K; (b) 25 dpa at 873 K + 25 dpa at 798 K; (c) 25 dpa at 873 K + 25 dpa at 723 K.

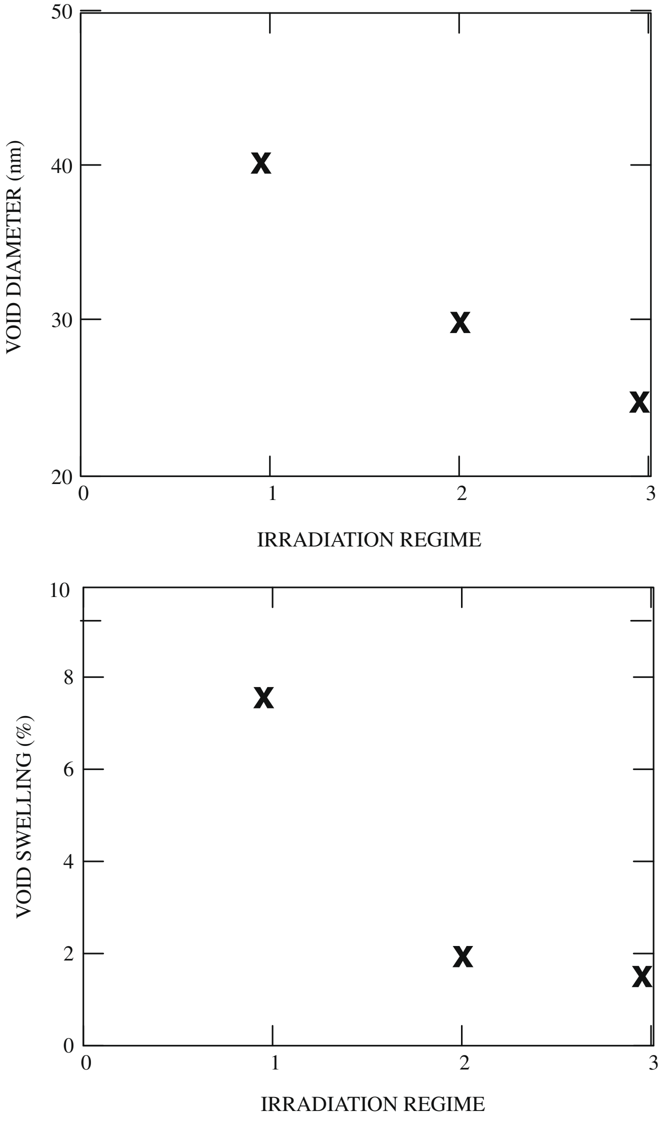

Fig. 2. Mean void diameter and swelling in Ni irradiated with Cr ions up to 25 dpa at 873 K (regime 1); 25 dpa at 873 K + 25 dpa at 798 K (regime 2) and 25 dpa at 873 K + 25 dpa at 723 K (regime 3). The size of symbols '*x*' corresponds to the mean error in void measurements.

The authors have attempted to explain the observations using the rate theory modified to include the interstitials injected by the ion beam. However, this effect was shown to be negligible due to a very low 'production bias' introduced by injected SIAs (about 0.1%) [7]. What is more, in the present experiment the energy of Cr ions was an order of magnitude higher than that in Ref. [3]. Accordingly, the Cr ions came to rest at a distance of about 10^4 nm, which exceeded the depth, at which voids have been produced, by orders of magnitude. The explanation of the void shrinkage presented in our previous papers [1,2,7] was based on the mechanism of radiation-induced vacancy emission from voids due to interaction with unstable Frenkel pairs (UFP) and focused collision sequences (focusons). It was concluded that the focuson effect dominates over the UFP effect if the focuson propagation length exceeds 50*b* where *b* is the inter-atomic distance. The focusons are unstable against thermal motion because they depend on the alignment of atoms. Typically, at ambient temperatures, the focuson range is limited to about ten unit cells. However, there exists another, essentially non-linear, mechanism of the excitation propagation over great distances, which is called self-focusing breathers [10–12]. As the incident focuson energy is dispersed, on-site potentials and long range co-operative interactions between atoms can influence the subsequent dispersal of energy in the lattice by the creation of self-focusing breathers (SFB). Russell and Eilbeck have presented evidence for the existence of energetic, mobile, highly localized lattice excitations that propagate great distances in atomic-chain directions in crystals of muscovite, an insulating solid with a layered crystal structure. Specifically, when a crystal of muscovite was bombarded at a given point, atoms were ejected from remote points on another face of the crystal, lying in atomic-chain directions at more than 10^7 unit cells distance from the site of bombardment. This points to the possibility of atoms being ejected from the void surface by the same mechanism, in a process of breather-induced vacancy emission from voids.

Although these results relate to layered crystals there is evidence that breathers can occur in non-layered crystals, but with shorter path lengths of order 10^4 unit cells [12]. This was reported in connection with radiation damage studies in silicon and with diffusion of interstitial ions in austenitic stainless steel. The demonstrated stability of breathers against thermal motion and ubiquitous occurrence makes this mechanism a promising candidate for the explanation of the radiation-induced void dissolution and related phenomena. Fig. 3 shows the calculated temperature dependence of the void growth/shrinkage rate in Ni irradiated under the present irradiation conditions for different ranges of the breather

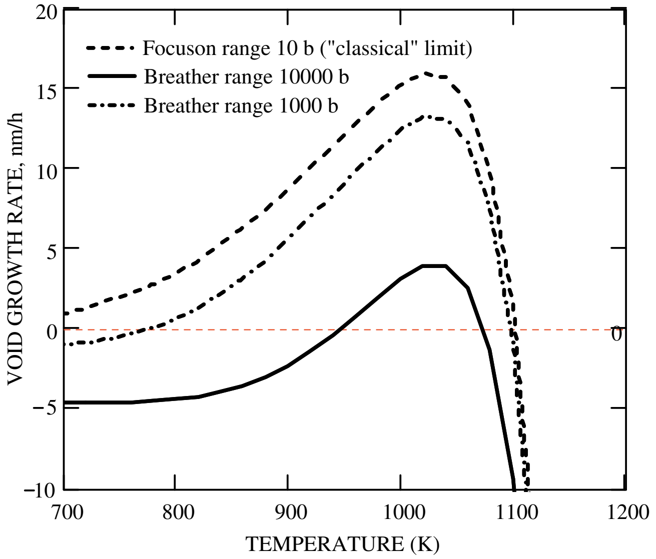

Fig. 3. Temperature dependence of the void growth/shrinkage rate in Ni irradiated at the dose rate of $K = 7 \times 10^{-3}\mathrm{s}^{-1}$ for different ranges of breather propagation. The void radius was assumed to be 10 nm and other material parameters are presented in Ref. [7]. The void shrinkage with decreasing irradiation temperature can be explained by the present model assuming the breather propagation range to be between 10^3 and 10^4 unit cells. The focuson range is limited to about ten unit cells, which would result in negligible deviance from the conventional theory shown as the 'classical' limit.

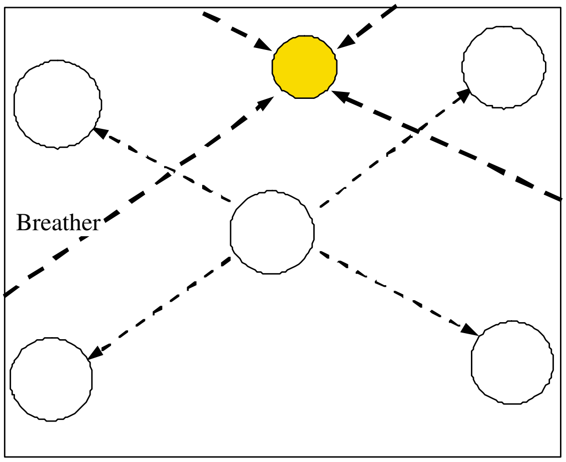

Fig. 4. Illustration of the dissolution of a void in the 'interstitial' position due to the absorption of breathers coming from larger distances as compared to 'regular' voids that shield each other from the breather fluxes along the close packed directions.

propagation. The void shrinkage observed in our experiment with decreasing irradiation temperature can be explained by the present model assuming the breather propagation range to be between 10^3 and 10^4 unit cells. The void shrinkage rate given by the breather-induced mechanism is essentially temperature independent as compared to the void growth rate due to biased vacancy absorption from the bulk [2,7]. The bias-induced void growth rate decreases with decreasing irradiation temperature due to the bulk recombination of Frenkel pairs, which results in the observed void shrinkage with decreasing irradiation temperature.

The radiation-induced void dissolution phenomenon is intrinsically connected with a void ordering observed in very different radiation environments ranging from metals to ionic crystals [13–19]. One might suggest that ordering phenomena is consequence of the *energy transfer* along the close packed directions provided by self-focusing breathers. Since the breather propagating range is larger than the void spacing, the voids can shield each other from breather fluxes along the close packed directions, which would provide a driving force for the void ordering, as illustrated in Fig. 4. An analytical model of the void ordering has been developed in Ref. [17], in which small dislocation loops provided the *interstitial transfer* along close-packed directions. The model can be modified to include breathers as an alternative ordering mechanism, which will be done elsewhere.

4. Summary and outstanding problems

As has been noted in [7], non-equilibrium fluctuations of energy states of the atoms surrounding crystal defects arise as a result of their interaction with radiation-induced excitations in the ionic system. These fluctuations result in radiation-induced recovery processes such as the void shrinkage and plastic strain, which should be taken into account in modeling of the microstructural evolution under irradiation.

Experimental results presented above give additional evidence in favor of this concept, while the recent findings demonstrating ubiquitous occurrence and stability of breathers against thermal motion makes this mechanism a promising candidate for the explanation of the radiation-induced void dissolution and related phenomena. More detailed data are needed for development of this direction of research both from the side of theory and experiment. Specifically, the dependence of the breather range on the crystal structure, type and concentration of impurities is of particular importance for evaluation of the material response to irradiation.

Acknowledgement

This study has been supported by the STCU Grant No. 4368.

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