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Dynamical process of defect clustering in Ni under the irradiation with low energy helium ions

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Abstract

Dynamical process of defects clustering in pure Ni under the irradiation with low energy (typically 5 keV) He^+ ions has been studied by in situ electron microscopy. Effects of the ion energy (0.5–20 keV), fluence, flux, depth and irradiation temperature on the formation of interstitial type dislocation loops (I-Loops) and bubbles were examined. The density of I-Loops sharply increased with the fluence, but was slightly dependent on the ion flux. It is demonstrated that the formation of I-Loops at room temperature is promoted 3–4 orders magnitude higher than that expected in the case of no assistance of helium atoms on the nucleation. These results suggest a possible nucleation mechanism where helium–vacancy complexes trap the self-interstitial atoms and act as nucleation sites of I-Loops. By irradiation with 20 keV He^+ ions, SFT were formed even at room temperature, coexisting with I-Loops. Bubbles are formed preferentially inside of I-Loops, and by more heavy irradiation, they coalesced or interconnected, leading to a characteristic channel structure.

1. Introduction

Plasma facing materials in a fusion device will be exposed to high flux- low energy-plasma ions such as helium and hydrogen isotopes and then store high density defects and exposed ions. These cause a detrimental surface erosion of the materials and have been the subjects of numerous investigations [1]. However, little information is available about the dynamical process of defect clustering under the irradiation with low energy helium ions, where the defect clustering should be strongly affected by the injected ions through their interactions with point defects, in comparison with a high-energetic-ion irradiation where the displacement cascade is essential.

For Ni irradiated with helium ions, thermal desorption spectroscopy [2], nuclear reaction analysis [3] and theoretical calculation [4] gave worthy information about helium diffusion and the trapping or detrapping at radiation induced defects. More information about the nature of the radiation induced defects is, however, necessary. TEM observations were valuable to reveal the microstructure change after the irradiation [5,6]. However, knowledge about the defect clustering process under the irradiation with He^+ ions, which allow quantitative discussion, and its energy dependence is still poor.

Recently, we have shown the usefulness of the in situ microscopy experiment of Ni under the irradiation with low energy hydrogen ions, and pointed out the effectiveness of hydrogen in the nucleation of I-Loops [7]. In the present work, effects of irradiation with low energy helium ions on the defect clustering in Ni are studied by an examination of influences of the ion energy, fluence, flux, depth and the irradiation temperature.

2. Experimental procedure

Materials used in the present work were pure Ni of 99.997% nominal purity supplied by Johnson-Matthey. Disk shaped specimens formed from the material were pre-annealed at 1200 K in a high vacuum furnace and

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electrochemically polished for electron microscopy. The specimen was irradiated with He^+ ions in an electron microscope, type JEOL-2010, by an ion accelerator connected to the microscope. The incident angle of the ion beam to the specimen surface was about 70°.

Defect clustering processes under the He^+ ion irradiation at several temperatures between room temperature (RT) and 873 K were continuously monitored and recorded on VTR tapes or photo plates. The typical energy of He^+ ions was 5 keV. It was varied from 0.5 to 20 keV and the flux was from 6.4×10^{16} to 6.4×10^{17} ions/m^2 s. The number density and the depth distribution of the defects were measured from stereomicroscopy of weak beam dark field images.

3. Results

3.1. I-Loops formation

Typical electron micrographs which show the microstructure evolution in Ni irradiated with 5 keV He^+ ions at RT are shown in Fig. 1. As seen, the clusters appeared above the fluence around 3×10^{17} ions/m^2 and are identified to be I-Loops. The number density of I-Loops sharply increased with the fluence until their overlapping becomes significant and they converted to high density dislocations above the fluence of 10^{20} ions/m^2. I-Loop formation profiles for two different fluxes, 6.4×10^{16} and 6.4×10^{17} ions/m^2 s, are also compared in the figure. It should be noticed that the increase of I-Loop density was slight at RT, even if the ion flux was increased from 6.4×10^{16} to 6.4×10^{17} ions/m^2 s.

In Fig. 2, the areal number densities of I-Loops formed by irradiation with 5 keV He^+ ions under the constant flux of 6.4×10^{16} ions/m^2 s at RT, 473, 573, 673, 773 and 873 K are plotted as a function of the fluence. As seen, the density decreased with increasing irradiation temperature. Even at 473 K, a significant decrease in the I-Loop density was seen. At 873 K, the formation of I-Loops under the flux of 6.4×10^{16} ions/m^2 s was scarce.

Fig. 3 shows depth distributions of I-Loops formed by irradiation at RT and 673 K, respectively. In the

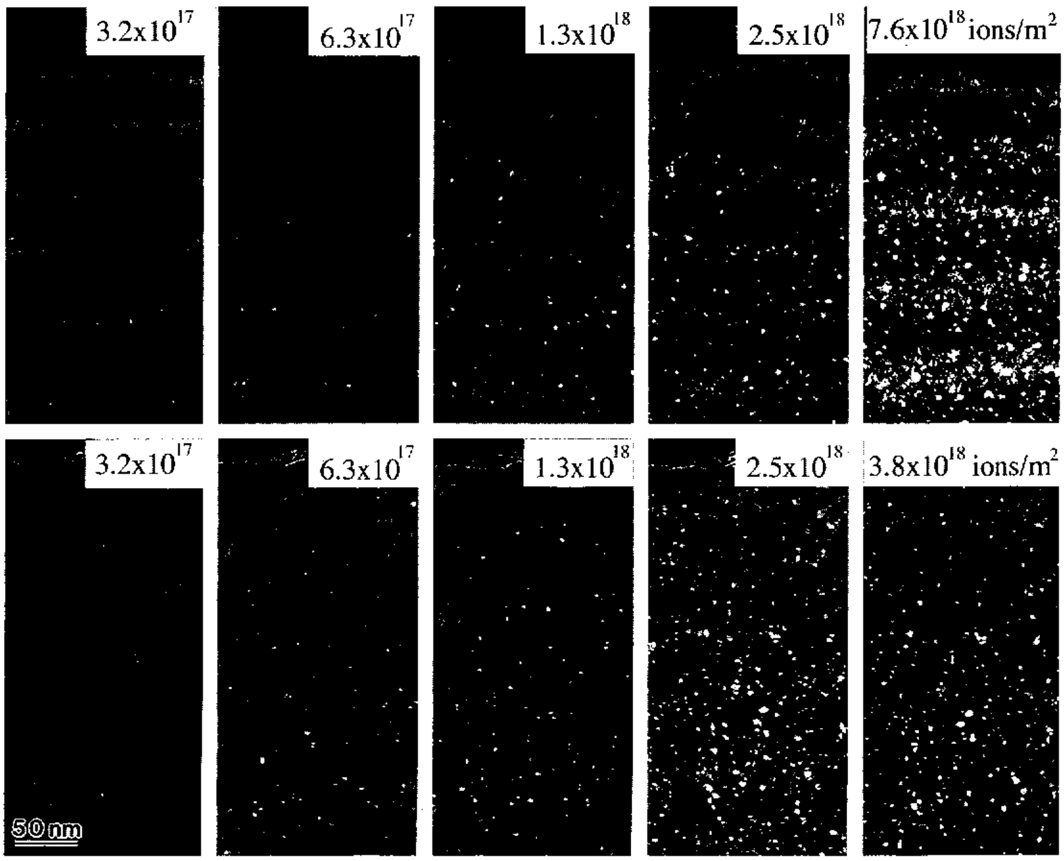

Fig. 1. Evolution of I-Loops with the fluence in pure Ni which was irradiated with 5 keV He^+ ions under the fluxes of 6.4×10^{16} ions/m^2 s (upper photographs) and 6.4×10^{17} ions/m^2 s (lower ones) at RT.

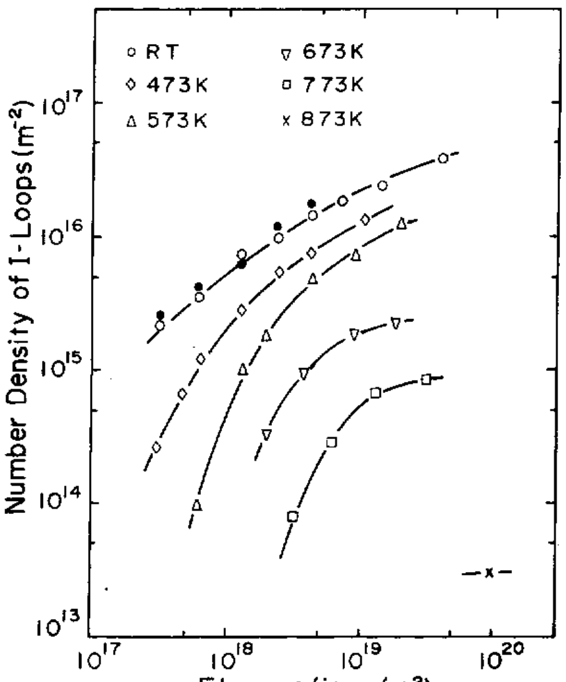

Fig. 2. Areal number density of I-Loops versus the fluence. All are irradiations with the constant flux of 6.4×10^{16} ions/m^2 s at the temperatures denoted, except (●) with the flux of 6.4×10^{17} ions/m^2 s at RT, as seen in Fig. 1.

lower part of the figure, the corresponding dpa- and apa-rate (He^+ ions/atoms s) depositions calculated by TRIM code [8] are compared. It should be noticed that the peaks of the I-Loop distribution at RT are located around 10–30 nm, which coincide with that of helium ion deposition. The I-Loop density in this region increased sharply with the fluence in the early stage of the irradiation. I-Loops were located gradually in larger depth at higher temperatures above 470 K. At 673 K, as seen in the figure, they were formed in the depth 10–70 nm with the peak around 35 n.

3.2. Bubble formation

Bubbles are remarkably formed at temperatures between 573 and 873 K by irradiation with 5 keV He^+ ions to the fluence above about 1×10^{19} ions/m^2. They appeared preferentially inside of the grown-up I-Loops, as seen in Fig. 4(a). With the increase of the fluence, the number density of the bubbles increased and they frequently appeared along the dislocation lines converted from I-Loops. Above the fluence about 2×10^{20} ions/m^2 at 673 K, the helium irradiated area was filled with high density bubbles and dislocations. At the fluence of 5.9×10^{20} ions/m^2, these bubbles coalesced or became interconnected and grew to a characteristic channel structure as seen in Fig. 4(b), which leads to surface blistering.

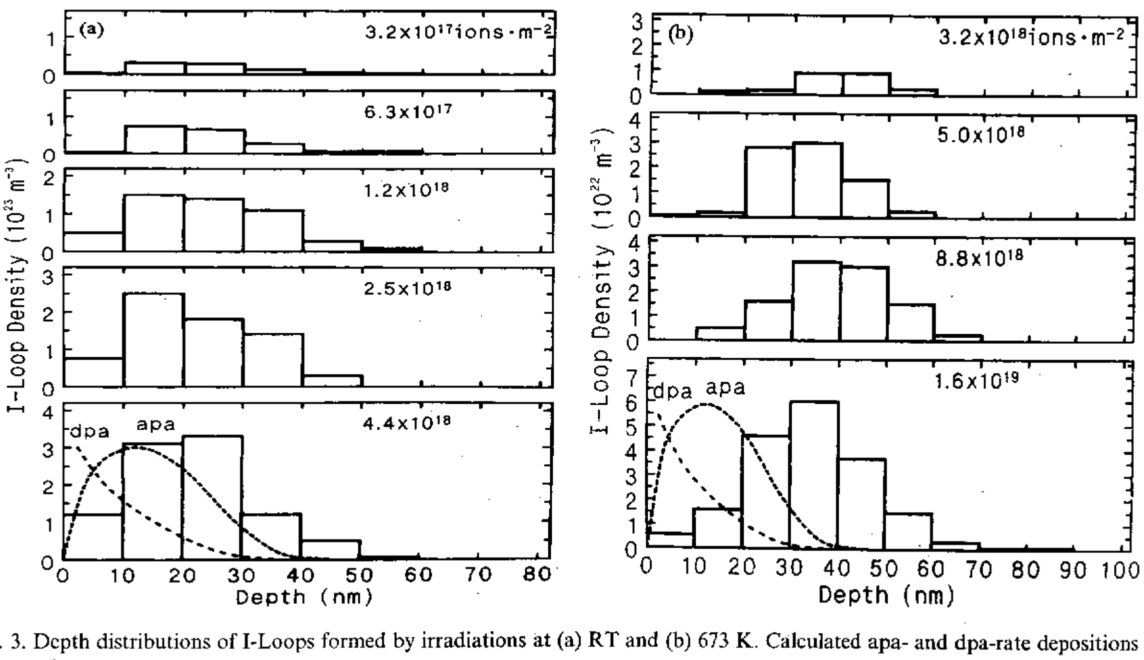

Fig. 3. Depth distributions of I-Loops formed by irradiations at (a) RT and (b) 673 K. Calculated apa- and dpa-rate depositions are compared.

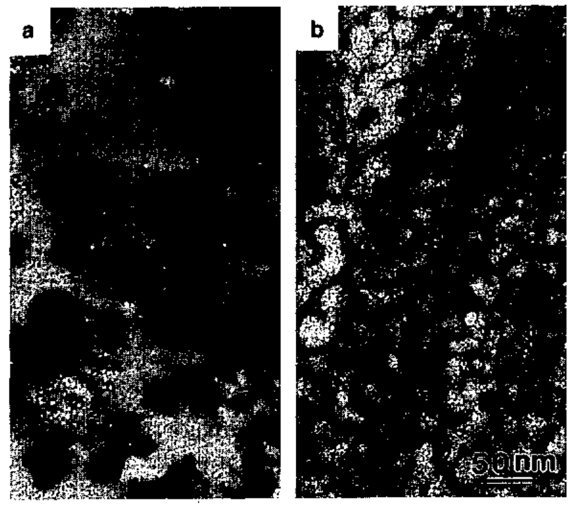

Fig. 4. (a) Bubbles which appeared preferentially inside of I-Loops by irradiation to 3.8×10^{19} ions/m^2 at 773 K. (b) Coalesced or interconnected bubbles with a characteristic channel structure, which were formed by irradiation to 5.9×10^{20} ions/m^2 at 673 K.

3.3. Energy dependence

For comparison with the above results of 5 keV He^+ ions irradiation, the energy of He^+ ions was changed. By irradiation with 0.5 keV He^+ ions, high density I-Loops and dislocation networks converted from the I-Loops were also formed at room temperature, although higher fluence than the case of 5 keV He^+ ions was necessary.

By irradiation with 20 keV He^+ ions at room temperature, stacking fault tetrahedra (SFT) were observed in a thinner part of the specimen, coexisting with I-Loops, as seen in Fig. 5, although SFT were never observed by 5 keV He^+ irradiation.

4. Discussion

To make clear the quantitative difference in I-Loop formation profiles between the present results and a case of no helium effects on the nucleation, we have calculated concentrations of vacancies C_V, the self-interstitial atoms C_I, helium atoms C_{He}, helium–vacancy complexes $C_{He\text{-}V}$ and I-Loops C_L, as a function of the irradiation time and the depth from the specimen surface, and compared the results with the present experimental results. The calculation was numerically made according to the nucleation kinetics in electron irradiated pure metals [9], using the dpa and apa rates which are given by TRIM code [8] (for example, dpa rate = 2.5×10^{-4}/s and apa rate = 1.3×10^{-5}/s in the depth 10–20 nm), the migration energies of the self-interstitial atoms 0.15 eV [10], vacancies 1.2 eV [11] and helium atoms 0.35 eV [12].

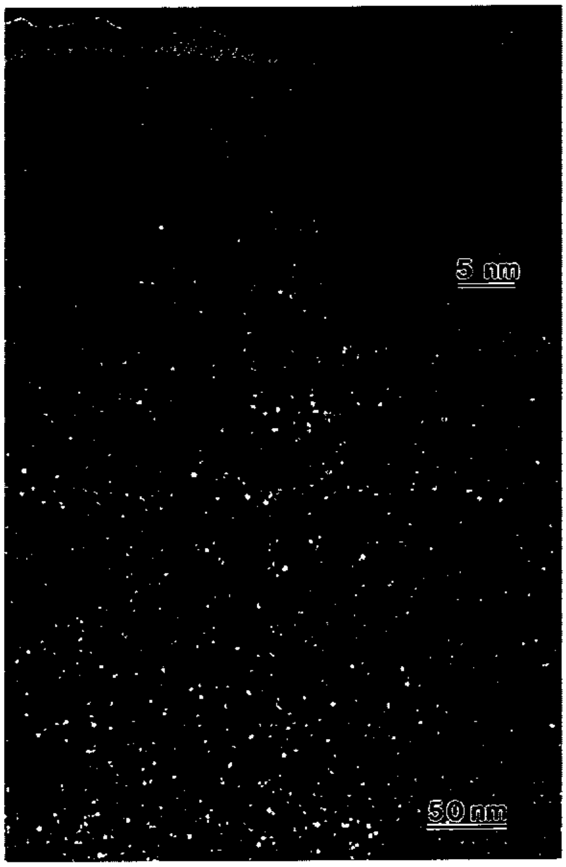

Fig. 5. I-Loops and SFT formed by irradiation with 20 keV He^+ ions to the fluence of 1.0×10^{19} ions/m^2 at RT. Enlarged figure of SFT which was observed in a thinner part of the specimen is inserted.

The dissociation energy of helium–vacancy complex is comparatively large [13], so we assumed that the complex is stable at RT, but pops out a helium atom by absorbing a self-interstitial atom [14]. A representative result of the calculation and the experimentally observed I-Loop concentrations in the depth 10–20 nm at RT are compared in Fig. 6. With the other examples of the calculation, it is clearly known that the observed results are much different from the calculated results in the following points: (i) The observed I-Loop concentration is 3–4 orders magnitude higher than the expected one from the di-interstitial nucleation model calculated. (ii) Although the sharp increase of I-Loop concentration with the fluence was observed, the increase in the calculation is small. (iii) As seen in Fig. 2, the experimental flux dependence of I-Loops at RT is slight against the expectation of the calculation. (iv) Sharp distributions of I-Loops with the peak around 10–30 nm at RT were observed as seen in Fig. 3, while the calculated distribution became more broad because of high mobility of the interstitial atoms at RT.

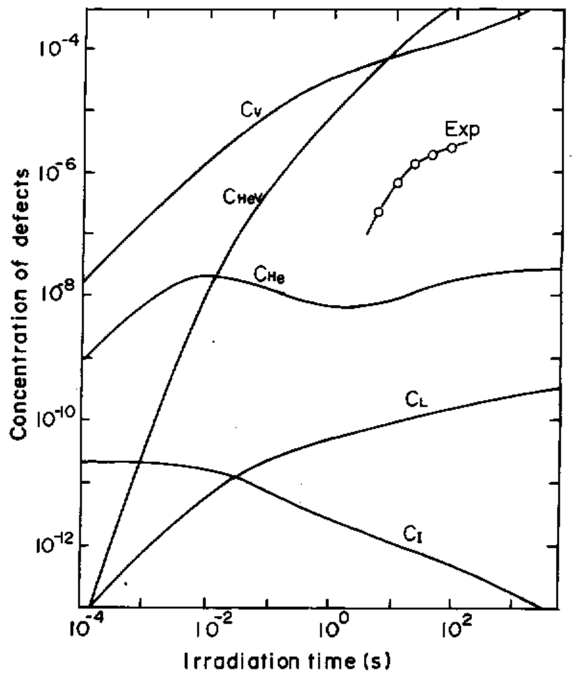

Fig. 6. Defects concentrations in the depth 10–20 nm at RT, which were calculated according to a di-interstitial nucleation model [9]. (o) indicates the observed concentration of I-Loops shown in Fig. 3.

The comparison of these results suggests that trapping centers of the self-interstitial atoms, which assist the nucleation of I-Loops, are formed during the He^+ ions irradiation and the density of the trapping center increases sharply with the fluence. The nature of trapping center is also characterized from the experimental facts of the depth distributions of I-Loops shown in Fig. 3. As seen, at RT, the observed peak coincides with the apa rate deposition, not the dpa rate deposition. At temperatures above 470 K, the distribution becomes broad and the peak tends to shift into a deeper region. These facts must be a result affected by the temperature depended distribution of the trapping centers. Therefore, it is most probable that the trapping center of the interstitial atoms consists of helium-vacancy complex, because vacancies in Ni become mobile at around 470 K and their complex with the helium atoms should become mobile at higher temperatures.

The complexes of helium-vacancy should cause a relaxation of the surrounding lattice and act as trapping sites of the self-interstitial atoms which grow up to I-Loops, unless the vacancy is fully filled with or overpressurized with helium atoms. Similar idea was reported in Mo [15] and Ni [5]. The present work give advanced experimental results which allow more quantitative discussion.

In the irradiation with 5 keV He^+ ions at RT, SFT were never observed, in contrast to the irradiation with 20 keV He^+ ions. These facts suggest that a treatment of the defect clustering process in the irradiation with 5 keV He^+ ions as a kinetic reaction process of Frenkel defects and helium atoms is valuable. However, in the irradiation with 20 keV He^+ ions, SFT should be produced even at RT by cascade effects and the escape of the self-interstitial atoms to the specimen surface, as similarly pointed out in Cu irradiated with He^+ ions [16], but not in Ref. [5]. The radiation induced diffusion of vacancies around the collision may also assist the formation of SFT at RT.

The calculation by TRIM code [8] indicates that 0.5 keV He^+ ions irradiated to Ni produce several Frenkel pairs per ion in the depth region of 0–2 nm and the helium atoms locate in the depth 0–5 nm, although 60% of irradiated He^+ ions are back scattered. This and the present experimental results suggest that strong trapping centers for the self-interstitial atoms are formed by the irradiation, overcoming a disappearance of the self-interstitial atoms to the specimen surface and should support the present nucleation model.

Zell et al. [17] reported a temperature dependent nucleation of helium bubbles in Ni, which was attributed to different mechanisms of helium diffusion at temperatures below 800 K and above. However, the present results shown in Sections 3.1 and 3.2 indicate that the existence of I-Loops is very effective to the formation of bubbles below 873 K. Therefore, one of reasons for the temperature dependent bubble formation should come from the formation or existence of I-Loops below around 800 K.

5. Conclusion

Effects of the ion energy, fluence, flux, depth and irradiation temperature on the formation of I-Loops and bubbles were examined. It was demonstrated that the I-Loop formation was much enhanced by helium irradiation. As a possible mechanism of I-Loop nucleation, it was concluded that helium-vacancy complexes trap the self-interstitial atoms and act as nucleation sites of I-Loops. Bubble formation was also much influenced by existence of dislocation loops. High density bubbles led to a characteristic channel structure.

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