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Formation process of dislocation loops in iron under irradiations with low-energy helium, hydrogen ions or high-energy electrons

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Abstract

Formation processes of interstitial-type dislocation loops (I loops) in high-purity Fe under irradiations with 5 keV H^+ ions or 1000 keV electrons are examined by in situ transmission electron microscopy at temperatures below room temperature, and the results are compared with that obtained under He^+ ion irradiation. For the electron irradiation, conventional model of I-loop nucleation based on the assumption that di-interstitial atoms are stable nuclei of I loops is questioned. The volume density of I loops by H^+ ion irradiation is one-order of magnitude higher than that by electron irradiation, and several times lower than that by He^+ ion irradiation. The temperature dependence of the volume density of I loops by H^+ ion irradiation supports the idea that such enhancement of I-loop formation is due to trapping of self-interstitial atoms by gas atom-vacancy complexes.

1. Introduction

Plasma facing components in nuclear fusion devices will be heavily exposed to low energy (a few 10 s of eV to a few keV) helium and hydrogen isotopes, and will accumulate high density of defects and injected elements. This may cause degradation of material properties and influence recycling of these elements. Hence, understanding of the damage evolution process in such environment is very relevant to the basic research for the development of plasma facing materials [1].

The formation processes of point defect clusters, such as interstitial-type dislocation loops (I loops) and cavities, and agglomerates of gas atoms under the irradiation with low-energy helium ions (He^+) or hydrogen ions (H^+ or D^+) have been vigorously examined for fcc and bcc metals. Some of these studies have demonstrated using transmission electron microscopy (TEM) that in addition to the formation of cavities, the formation of I loops is also greatly enhanced by these injected ions into fcc Al [2], Cu [3,4] and Ni [5–9] and bcc Fe [10,11], Mo [1,12,13] and W [14,15]. Furthermore, such effect of helium is known to be stronger than that of hydrogen in Ni [5] and Mo [1].

In the past, several mechanisms for the enhanced loop formation have been proposed. For He^+ ion irradiation, a mechanism that He-V-di-I complexes become the nuclei of I loops (model I) [4,5,9,11,12,14] is now considered to be the most reliable one, where V denotes vacancies and I denotes self-interstitial atoms. In contrast, for H^+ (D^+) ion irradiation, two additional mechanisms were proposed [2,5,7,8]. One of them considers that H-di-I complexes become the nuclei of I loops (model II) [3,6,15], and the other is based on the hypothesis that an enrichment of self-interstitial atoms is caused by the formation of H–V complexes (model III) [1,13].

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Recently, the formation process of I loops in bcc Fe has been examined under He^+ ion irradiation at temperatures in the range of 85–770 K [11]. This study supported the model I for the enhanced formation of I loops. In contrast, only a few results on the formation of I loops under H^+ irradiation have been obtained [10]. In addition, the absolute values of I-loop volume densities under electron irradiation, which may be useful for extracting quantitatively the effects of He and H atoms on the I-loop formation processes, have been obtained for only limited conditions [16–18].

The aim of the present study was to acquire systematic data on the formation processes of I loops in bcc Fe under low-energy H^+ ion or high-energy electron irradiations, to compare them with one another, and to extract the effects of He and H atoms on I-loop formation. In order to interpret the I-loop formation process, the irradiation was performed even for temperatures below 220 K below which vacancies are thermally immobile [19]. High-purity iron was used to avoid effects of residual impurities [17].

2. Experimental procedures

The material used in the present work was Fe of 99.999% nominal purity and was supplied by Showa Denko Inc. Sheets of this Fe rolled down to 0.08 mm in thickness were pre-annealed at 1120 K for 1 h in a dry-hydrogen atmosphere purified by ZrH_2. Then, they were electrochemically polished, cut and sandwiched in pairs of molybdenum single meshes of 3 mm diameter for TEM. The surface of the specimen used was near to (0 0 1), and the incident beam orientation was always set approximately parallel to [0 0 1] direction.

Ion irradiations of the specimens were carried out in an electron microscope, type JEOL-2010, by an ion accelerator RIB-20S connected to the microscope [20]. H^+ ion irradiation was performed at an acceleration voltage of 5 kV to the fluence of about 1×10^{20} H^+/m^2, where dpa (displacement per atom) at the peak depth was calculated to be 1×10^{-1} by TRIM code [21], or more at 140, 180, 235 and 300 K. The beam flux was 6.5×10^{17} $H^+/m^2\,s$, where the dpa rate at the peak depth was estimated to be 5.0×10^{-4} dpa/s. By such low-energy irradiation, direct formation of complexes of self-interstitial atoms or vacancies by collision cascades are considered to occur only rarely [11,22], hence the formation of I loops are expected to be due to the interaction among self-interstitial atoms, vacancies and implanted hydrogen atoms. The process of microstructure evolution during ion irradiation was monitored continuously by TEM, operated at 200 kV. After the irradiation, annealing of the specimens was performed in order to examine the characters of radiation-induced defects.

High-energy electron irradiation and in situ observation was performed in a high-voltage electron microscope H-3000 (Hitachi) operated at 1000 kV and at temperatures ranging from 20 to 300 K. The beam fluxes were 9.2×10^{22} and 9.2×10^{23} $e^-/m^2\,s$ which correspond to dpa rates of 5.0×10^{-4} and 5.0×10^{-3} dpa/s, respectively. The beam flux for H^+ irradiation and former flux for electron irradiation were set so that the dpa rate at the peak depth was equal to that for the previous He^+ irradiation [11].

For in situ TEM, kinematical and dynamical bright-field and weak-beam dark-field imaging were used. The reflection adopted was mainly $\mathbf{g} = 110$ with the deviation parameter from the exact Bragg condition, s, ranging from 0.06 to 0.2 nm^{-1}. For H^+ ion irradiation, stereomicroscopy was used in order to obtain the volume density of I loops at each depth from the ion-incident surface. The areal or volume densities of I loops were obtained from the regions about 150 nm thick, which is larger than the depth distributions of I loops.

3. Results and discussion

3.1. H^+ ion irradiation

Under H^+ ion irradiation, dot-like defects were observed. The density and size of these clusters increased with fluence. Fig. 1 shows the effect of irradiation temperature on the microstructural evolution. The defect clusters induced at 235 and 300 K clearly indicate the contrast of dislocation loops. Dot-like defects smaller than several nm in diameter rarely performed one-dimensionally back-and-forth motion. Such a phenomenon has been observed during electron irradiation [23] and has been considered to be an intrinsic behavior of small perfect dislocation loops [24].

No cavities were observed during the irradiation at the irradiation temperatures of 140, 180, 235 and 300 K. Instead, the post-irradiation annealing at 300 K led to the formation of small cavities (smaller than 4 nm in diameter) in the specimens irradiated below 300 K.

It has been reported that hydrogen platelets showing faint contrast in comparison with dislocation loops are formed under 5 keV H^+ ion irradiation at the fluence of 1×10^{20} H^+/m^2 at 300 K [10]. However, in the present study, such contrast was not observed even for the irradiation to the fluence of 2.3×10^{21} H^+/m^2 at the same temperature. For the irradiation at the other temperatures, such contrast was not found either. However, in the process of the annealing of the specimen irradiated at 140 K, the objects having the same contrast as that reported in Ref. [10] were formed below 180 K. The density of these objects decreased with increasing annealing temperature and almost all of them disappeared at around 300 K, and finally only a few dot-like defects

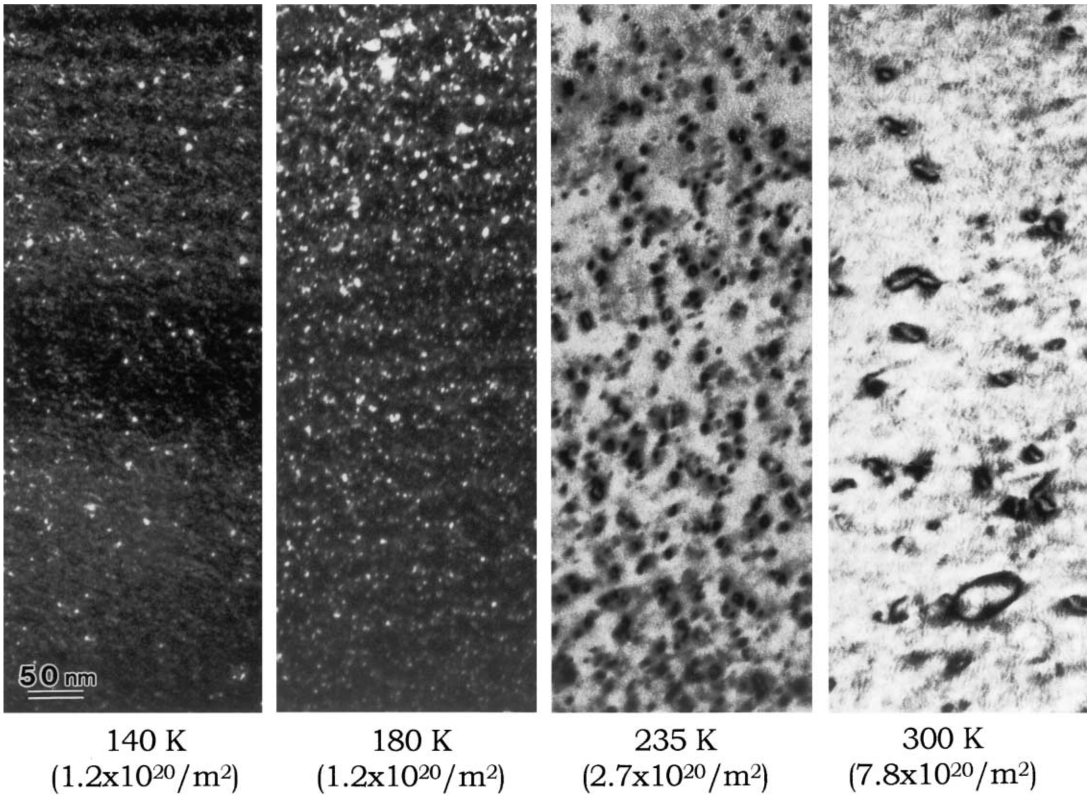

Fig. 1. Temperature dependence of microstructure in Fe induced by irradiation with 5 keV H^+ ions with a beam flux of 6.5×10^{17} H^+/m^2 s.

remained. Fig. 2 shows the change in the contrast of the objects with the change in the sign of diffraction vector **g**. As shown in the figure, their contrasts are converted from black into white or from white into black with the change in the sign of **g**. Such contrast change was pointed out to be unique for the hydrogen platelets [10]. These objects were not found by annealing of the specimens irradiated above 140 K.

From the above results, the dot-like defects formed under the irradiation at temperatures above 140 K are considered to be almost all dislocation loops. In addition, the dislocation loops introduced at 300 K were determined to be of interstitial type by the inside–outside contrast change with the change in the sign of **g** [25]. As shown in Fig. 1, their number density increases and their size decreases monotonously with decreasing irradiation temperature. Hence, almost all of the dislocation loops are considered to be of interstitial type (I loops).

3.2. Electron irradiation

I loops were formed during the electron irradiation at both beam intensities and at all the temperatures examined. They became visible below the fluence of 1×10^{24} e^-/m^2. They grew gradually with the fluence. The size of I loops is larger at higher temperature. Some of the smaller I loops made intermittent one-dimensional back-and-forth motion as in the case of the H^+ ion irradiation even at temperatures below 120 K where self-interstitial atoms are thermally immobile [19]. The direction of the I-loop motion was along $\langle 111 \rangle$ orientations, which were parallel to their Burgers vectors $1/2\langle 111 \rangle$. Some of the moving I loops coalesced with other I loops, or escaped from the specimen. When mobile I-loops interact with other I loops, they often changed their Burgers vectors into $\langle 100 \rangle$, which are those of sessile I loops. Such coalescence or escape occurred frequently during the irradiation with the higher beam intensity and the volume density of I loops decreased with the fluence. In contrast, with the weaker beam intensity, the density of I loops saturated soon after their appearance.

Fig. 3 shows the temperature dependence of the volume density of I loops for two beam intensities. The volume densities shown in the figure are those of I loops before the densities begin to decrease due to I-loop motion. Nucleation of I loops under the presence of single-interstitial atoms at temperatures where vacancies

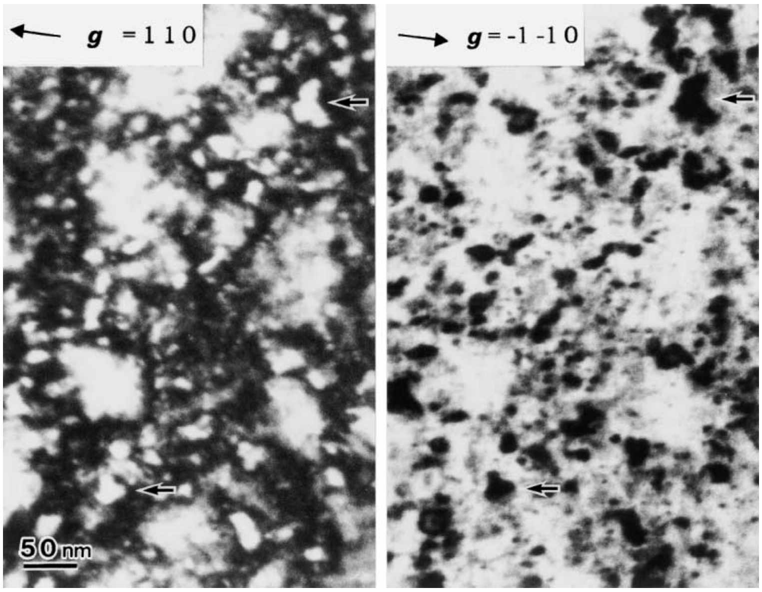

Fig. 2. Change in the microstructure images with the sign of the diffraction vector **g**. The specimen was irradiated with 5 keV H^+ ions with a beam flux of 6.5×10^{17} H^+/m^2 s to a fluence of 2.2×10^{20} H^+/m^2 at 140 K, then subsequently annealed up to 180 K. Observation axis is approximately [0 0 1] direction.

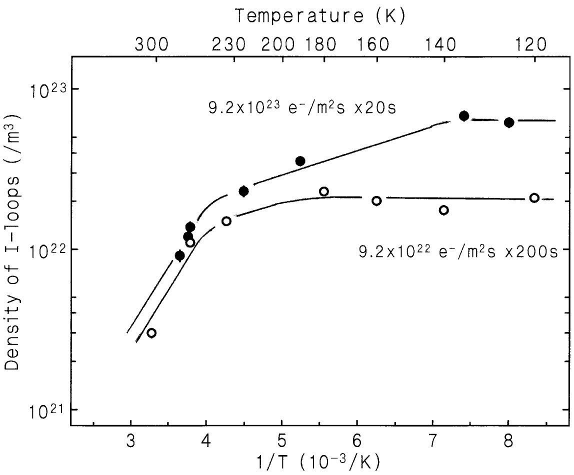

Fig. 3. Temperature dependence of volume density of I loops induced by irradiation with 1000 keV electrons with beam fluxes of (●) 9.2×10^{22} e^-/m^2 s and (○) 9.2×10^{23} e^-/m^2 s.

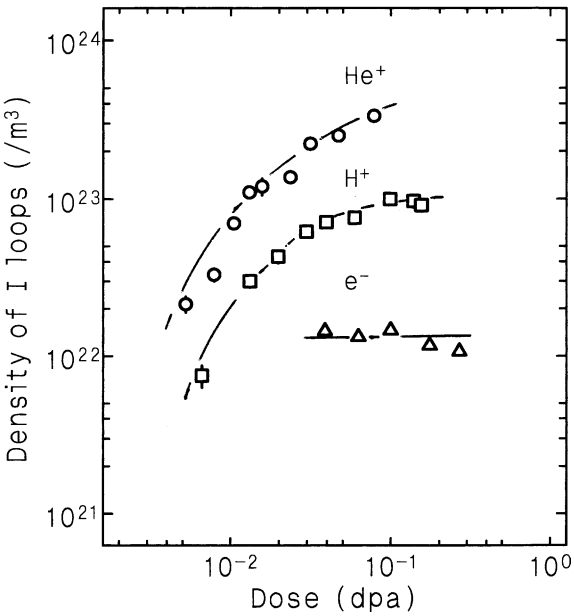

Fig. 4. Comparison among volume densities of I loops formed under irradiation with 5 keV He^+ ions, 5 keV H^+ ions or 1000 keV electrons at 235 K. The vertical axis is the density of I loops at each peak depth of I-loop density for (○) He^+ ion irradiation (20–30 nm, 2.6×10^{-4} dpa/s), (□) H^+ ion irradiation (20–30 nm, 3.3×10^{-4} dpa/s), and (△) electron irradiation (5.1×10^{-4} dpa/s).

are immobile has been studied by use of reaction kinetics theory [16]. According to such studies, the nuclei of I loops in metals under high-energy electron irradiation are stable di-interstitials and complexes of impurity atom and self-interstitial atom [17]. On the other hand, small interstitial complexes were pointed out to be highly mobile from MD calculations [26,27], which may be consistent with the present result of the one-dimensional motion of I loops. Hence, the validity of the above nucleation model is considered to be doubtful. The detail of the kinetics of I-loop nucleation under these conditions will be reported in the near future [28].

3.3. Comparison among He^+, H^+ ion and electron irradiation

Fig. 4 shows an example of the dose dependence of I-loop volume density for He^+ [11], H^+ ion and electron irradiation. In the figure, the vertical axis is the volume density at each peak depth of the I-loop density. As shown in Fig. 4, I-loop density by He^+ ion irradiation is several times higher than that by H^+ ion irradiation, and the density by H^+ ion irradiation is one order of magnitude higher than that by the electron irradiation. This indicates that H atoms enhances I-loop formation, and that the effect is stronger for He atoms than that for H atoms in Fe, as well as in other metals such as Ni [5] and Mo [1].

Fig. 5 shows the depth distributions of the volume density of I loops by H^+ ion irradiation for three temperatures. As shown in Fig. 5, at 180 K, the peak depth of I-loop volume density is situated at the depth of dpa peak. In contrast, at 235 K, the peak depth is deeper, and the profile of the distribution is broader. Such change in the distribution at 235 K is considered to occur since vacancies become thermally mobile. Hence, the enhancement of I-loop formation by H atoms is expected to be due to the model I, as well as that by He atoms [11].

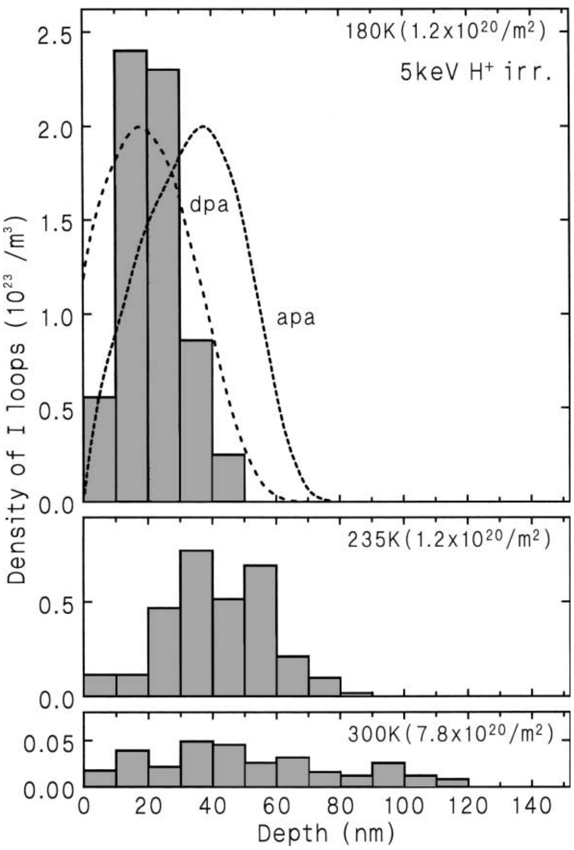

Fig. 5. Depth distributions of volume density of I loops formed during irradiation with 5 keV H^+ ions at several temperatures. Calculated dpa- and apa (deposited H atom per Fe atom)-rate profiles are compared (in arbitrary units).

4. Conclusions

This study provides significant data on the formation of interstitial-type dislocation loops (I loops) in high-purity Fe under irradiation with low-energy H^+ ions and high-energy electrons. It has been shown that I-loop formation is enhanced by H^+ irradiation and that the effect of H atoms is weaker than that of He atoms. It is suggested that hydrogen-vacancy complexes trap self-interstitial atoms and act as nucleation sites for I loops. This may be considered to be the mechanism for the enhancement of I-loop formation.

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