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Defect accumulation behavior in iron irradiated with energetic ions and electrons at ~80 K

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Abstract

Defect accumulation behavior in α-Fe irradiated with 0.5–2.0 MeV ^{1}H, ^{4}He, ^{12}C, ^{20}Ne and ^{40}Ar ions and 2.0 MeV electrons was studied. The change in electrical resistivity of the specimen at ~80 K was measured as a function of particle fluence in order to obtain the defect production curve. After irradiation, the defect recovery spectra were measured up to ~250 K. From the experimental results, the defect production cross-section, the recombination volume, the damage efficiency (the ratio of the experimental defect production cross-section to the calculated one) and the fraction of the stage-I recovery were derived for each irradiation. Primary Knock-on Atom (PKA) energy dependence of the damage efficiency, the recombination volume and the fraction of the stage-I recovery is discussed.

1. Introduction

Properties of materials are remarkably changed when they are exposed to radiation field. As a consequence, it is of prime importance to investigate the radiation effects of materials in order to determine their lasting qualities regarding radiation and to select the optimum materials under extreme radiation conditions (e.g. fusion reactor).

For evaluating the radiation effects of fusion reactor materials, the estimation of the number of lattice defects or displacements per atom (dpa) produced by irradiation is required. In NRT model [1], it has been considered that the number of irradiation-produced defects is simply proportional to the Primary Knock-on Atom (PKA) energy. However, the defect recombination due to lattice vibration induced by the energy transfer to target atoms takes place. Thus, the number of defects surviving a collision cascade does not show a linear dependence on the PKA energy. In order to predict the number of defects, computer simulation studies by molecular dynamics are now in progress for some metals [2]. The empirical relationship between the number of defects and the PKA energy has been proposed in Ref. [2].

In actual irradiation, however, the PKA energy spectrum distributes widely. On the average, the PKA energy for neutron-irradiation is much higher (by several orders of magnitude) than that for electron-irradiation. In the case of ion-irradiation, the PKA energy spectrum can be systematically varied by changing the ion mass and the ion energy. Therefore, ion-irradiation is quite appropriate for the study on the PKA energy dependence of radiation damage process. So far, a large number of electron- and neutron-irradiation effects in metals have been studied in detail [3]. These irradiation experiments have been performed at low temperatures in order to freeze the thermal diffusion of defects. Concerning the ion-irradiations, although several studies on FCC metals were performed [4–7], there are few systematic studies on BCC metals irradiated with energetic ions except GeV heavy ions [8].

In the present work, we performed irradiations of α-Fe, which is one of the typical BCC metals, with energetic ions and electrons at low temperature and studied the defect accumulation behavior for these irradiations.

2. Experimental procedure

Iron thin films about 200–250 nm thick were deposited on α-Al_2O_3 single crystal substrates by rf magnetron sputtering with an Fe target (99.99%) using Ar gas. The

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electrical resistivity of the specimen at ~80 K is typically 4–5 μΩ cm. These films were irradiated with 0.5 MeV ^{1}H, 1.0 MeV ^{4}He, 1.0 MeV ^{12}C, 1.0 MeV ^{20}Ne and 2.0 MeV ^{40}Ar ions at ~80 K using a 2 MV Van de Graaff accelerator at JAERI (Japan Atomic Energy Research Institute)-Tokai and with 2.0 MeV electrons using a 3 MV single-ended accelerator at JAERI-Takasaki.

The projected ranges of ions and electrons in Fe for the present experiments are much larger than the thickness of the specimen. Therefore, most of the irradiating particles can pass through the specimen without remaining as impurities and the defects produced by these particles are distributed uniformly in the specimen.

Usually, to freeze the thermal diffusion of defects in metals, irradiation is performed at liquid-He temperature. However, the present irradiation experiments were performed at liquid-N_2 temperature, because most of irradiation-produced defects in Fe cannot move up to ~80 K. We measured electrical resistance of the specimens before, during and after irradiation by means of a conventional four-probe method. During irradiation, the measurement was performed at appropriate fluence intervals until the change in resistivity of the specimen, $\Delta\rho$, reached ~0.3 or 3.0 μΩ cm. It can be assumed that the change in resistivity is proportional to the amount of defects produced by irradiation in metals; $\Delta\rho = \rho_F \cdot C$, where ρ_F is the resistivity of a Frenkel pair and C is the concentration of irradiation-produced defects. In the present work, $\rho_F = 1250$ μΩ cm was employed for Fe [9]. After irradiation, the recovery behavior of irradiation-produced defects was observed during annealing up to ~250 K at a constant heating rate (~2 K/min). The difference in the resistivity between before and after irradiations at the same temperature corresponds to the amount of defects surviving the recombination due to the thermal diffusion at this temperature.

The influence of the size effect on electrical resistivity of the specimen is negligible in the present experiment. The correction of the change in resistivity by irradiation, $\Delta\rho$, for the size effect is $\leqslant 0.3\%$ at ~80 K [10].

3. Results and discussion

Fig. 1(a)–(c) shows the defect production curves, i.e., the resistivity change, $\Delta\rho$, as a function of particle fluence, Φ, for ^{1}H and ^{40}Ar ion- and electron-irradiations. For ion-irradiations, these curves exhibit a saturation behavior as Φ increases. This indicates that the irradiation-produced defects are annihilated due to the interactions both between defects themselves and between defects and incident particles in the region of rather high fluence.

The defect production curve used to be analyzed by the following conventional equation [11]:

$$\frac{\mathrm{d}C}{\mathrm{d}\Phi} = \sigma_\mathrm{d}(1 - 2v_0 C), \tag{1}$$

where σ_d is the defect production cross-section and v_0 is the spontaneous recombination volume. From the solution of Eq. (1), $\Delta\rho$ is given by

$$\Delta\rho = \rho_\mathrm{F} C = \frac{\rho_F}{2v_0}\{1 - \exp(-2v_0\sigma_\mathrm{d}\Phi)\}. \tag{2}$$

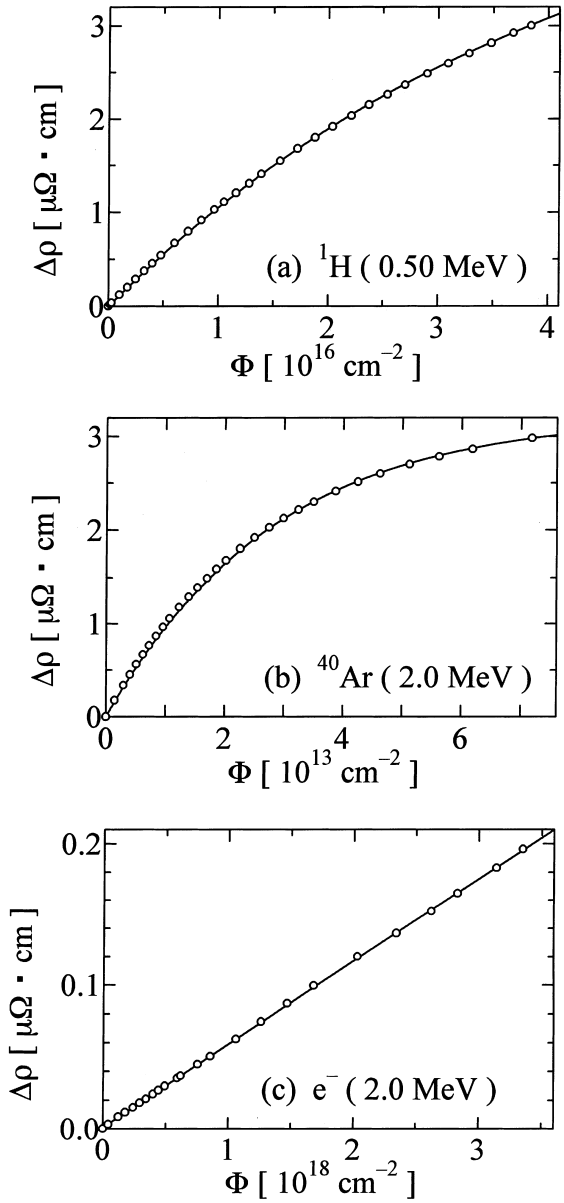

Fig. 1. Change in electrical resistivity of the specimen, $\Delta\rho$, as a function of particle fluence, Φ, at ~80 K for (a) 0.5 MeV ^{1}H ion-, (b) 2.0 MeV ^{40}Ar ion- and (c) 2.0 MeV electron-irradiations. The solid curves are drawn by the least-squares fitting of the data to Eq. (7) for (a) and (b) and to Eq. (3) for (c).

For all the ion-irradiations, σ_d and v_0 were derived by fitting the defect production curve to Eq. (2). In the case of electron-irradiation, no indication of saturation was found since the electron fluence was too small. Therefore, σ_d for electron-irradiation was derived by fitting the defect production curve to

$$\Delta\rho = \rho_F \sigma_d \Phi. \tag{3}$$

Thus, v_0 for electron-irradiation could not be obtained from the present experimental data.

The defect production cross-section, σ_d, means the concentration of defects produced by the unit fluence of incident particles. The damage efficiency, ξ, is defined as $\xi = \sigma_d^{exp}/\sigma_d^{cal}$, where σ_d^{exp} is the experimental value of σ_d, and σ_d^{cal} is the value of σ_d calculated by assuming that all the energy transferred from an incident particle to target atoms through elastic interactions is used for the defect production. Hence, ξ indicates how effectively the energy of the incident particle is used for the defect production.

The values of σ_d^{cal} for all irradiations except electron-irradiation were calculated by using TRIM-92 computer code [12] with a threshold displacement energy, E_d, of 24 eV [9]. For electron-irradiation, σ_d^{cal} was calculated by

$$\sigma_d^{cal} = \int_{E_d}^{T_{max}} \nu(T) \frac{d\sigma(E,T)}{dT} dT, \tag{4}$$

where T_{max} is the maximum energy of PKAs (in case of head-on collision), T is the PKA energy, E is the energy of an incident electron, $\nu(T)$ is the damage function which means the number of Frenkel pairs produced by a PKA with energy T, and $d\sigma(E,T)/dT$ is the differential scattering cross-section between an incident electron and a target atom. The NRT model was employed for $\nu(T)$ [1]. As 2.0 MeV electrons are in the relativistic range of velocity, T_{max} and $d\sigma(E,T)/dT$ given in Ref. [13] were used.

As a matter of fact, the damage efficiency for electron-irradiation, ξ_e, must be close to 1, because irradiating electrons produce mainly simple Frenkel pairs and not cascade damage. Therefore, in Fig. 2, the normalized damage efficiency, ξ/ξ_e, for each irradiation is plotted as a function of the PKA median energy, $T_{1/2}$, which is the characteristic of the PKA energy spectrum and is defined as the PKA energy above which half the defects are produced [4,5]. The normalized damage efficiency, ξ/ξ_e, decreases monotonically as $T_{1/2}$ increases. The deviation of ξ/ξ_e from 1 indicates that the energy transferred from the incident particle to the target atoms is used not only for the defect production but also for the defect annihilation within the cascade, which has not been considered in the calculation of σ_d.

The spontaneous recombination volume, v_0, for each ion-irradiation was obtained by fitting the defect production curve to Eq. (2). According to its definition, v_0 should depend only on the target material. Therefore, v_0 for all of the present irradiations should be constant. However, experimental v_0 varies with the kind of irradiating particle. This suggests that defects are annihilated not only through the spontaneous recombination but also through the subthreshold recombination, which is caused by interactions between the incident particle and pre-existing defects, while Eq. (1) does not contain this effect. Instead of Eq. (1), therefore, we have to use the following equation,

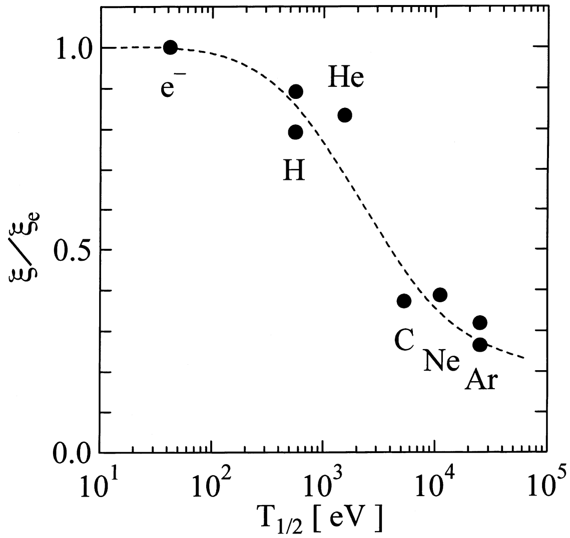

Fig. 2. Normalized damage efficiency, ξ/ξ_e, plotted against the PKA median energy, $T_{1/2}$, for each irradiation.

$$\frac{dC}{d\Phi} = \sigma_d(1 - 2v_0 C) - \sigma_r C, \tag{5a}$$

where σ_r is the subthreshold recombination cross-section. Then, Eq. (5a) is rewritten as

$$\frac{dC}{d\Phi} = \sigma_d\left\{1 - 2\left(v_0 + \frac{\sigma_r}{2\sigma_d}\right)C\right\} \equiv \sigma_d(1 - 2v_0^* C), \tag{5b}$$

where v_0^* is the effective recombination volume defined as

$$v_0^* \equiv v_0 + \frac{\sigma_r}{2\sigma_d}. \tag{6}$$

From the solution of Eq. (5b), $\Delta\rho$ is given by

$$\Delta\rho = \frac{\rho_F}{2v_0^*}\left\{1 - \exp\left(-2v_0^*\sigma_d\Phi\right)\right\}, \tag{7}$$

which is the same form as Eq. (2).

Fig. 3 shows the normalized effective recombination volume, v_0^*/v_{0e}^*, as a function of the PKA median energy, $T_{1/2}$, for the present ion-irradiations, where v_{0e}^* is the effective recombination volume for the electron-irradiation and obtained from the experimental data shown in Ref. [14] in the same manner as used in the present work. In contrast with the damage efficiency, v_0^*/v_{0e}^* increases

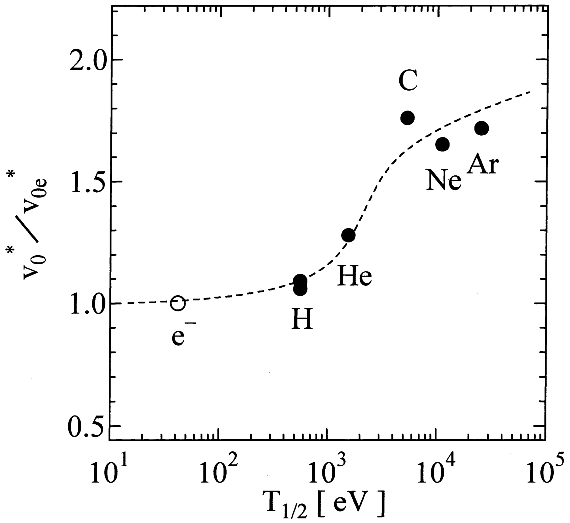

Fig. 3. Dependence of normalized effective recombination volume, v_0^*/v_{0e}^*, on the PKA median energy, $T_{1/2}$, for the present ion-irradiations (solid circles) and for electron-irradiation [14] (open circle).

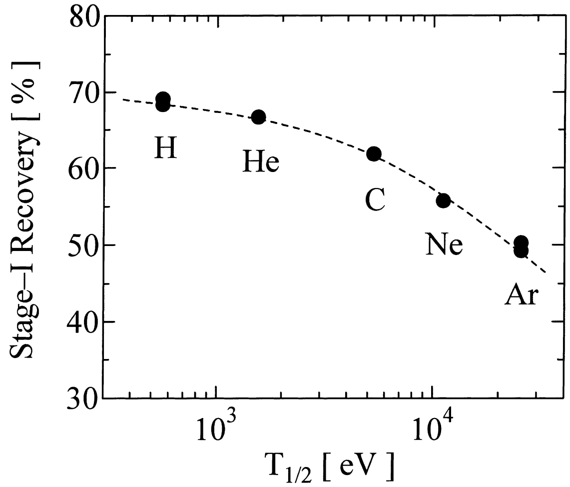

Fig. 5. Fraction of stage-I recovery as a function of the PKA median energy, $T_{1/2}$, for each ion-irradiation, which was derived from results in Fig. 4.

with $T_{1/2}$. It indicates that the subthreshold recombination is promoted with an increase in $T_{1/2}$. At $T_{1/2} < \sim 10^3$ eV, however, v_0^*/v_{0e}^* becomes nearly constant. It seems that v_0^*/v_{0e}^* is close to 1 at low $T_{1/2}$. For electron-irradiation, therefore, the subthreshold recombination

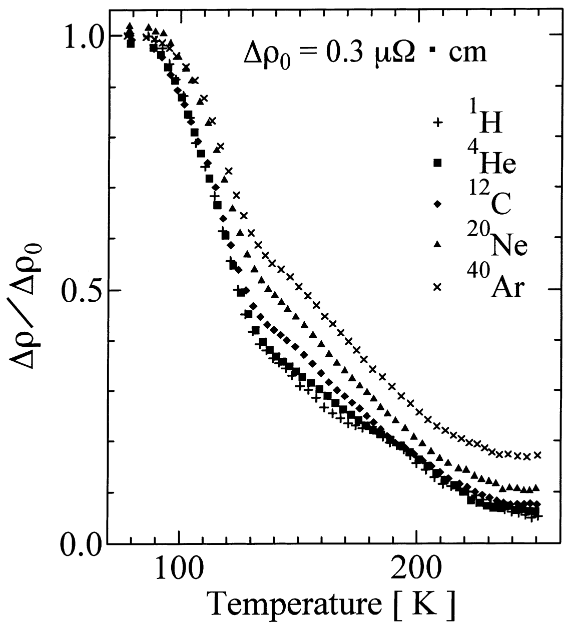

Fig. 4. Defect recovery spectra for ion-irradiations. Resistivity change of the specimen by irradiation before annealing, $\Delta\rho_0$, was $\sim$0.3 μΩ cm.

can be neglected as compared with the spontaneous recombination.

After each ion-irradiation, we obtained the defect recovery spectrum, which is shown in Fig. 4. The change in electrical resistivity of the specimen by each irradiation before annealing, $\Delta\rho_0$, was $\sim$0.3 μΩ cm which corresponded to the defect concentration of $\sim$240 ppm. The defect recovery is promoted at $\sim$120 K, which is called the stage-I recovery in Fe, and it is known that comparatively free interstitials recombine with vacancies around the stage-I temperature [15]. Fig. 5 shows the fraction of the stage-I recovery for each irradiation as a function of the PKA median energy, $T_{1/2}$, assuming that the recovery up to 150 K corresponds to the stage-I recovery. The fraction of the stage-I recovery, i.e., the fraction of the free interstitials surviving recombination tends to decrease with an increase in $T_{1/2}$. It implies that at higher PKA energy some of the free interstitials had already recombined with vacancies during irradiation and/or the free interstitials were not produced so many because of the recombination of defects within the cascade, which resulted in the decrease in the damage efficiency.

4. Summary

The defect accumulation behavior in α-Fe irradiated with energetic ions and electrons at $\sim$80 K was studied. The defect production curves and the defect recovery spectra were obtained by measuring the resistivity of the specimen before, during and after each irradiation. Then, the defect production cross-section, the effective recombination volume, the damage efficiency and the fraction of the stage-I recovery were evaluated. The damage efficiency decreases with increasing the PKA median energy, $T_{1/2}$, as a whole. It indicates that the defect annihilation within the cascade is promoted at higher $T_{1/2}$. The effective recombination volume increases with $T_{1/2}$ in contrast with the damage efficiency. It implies that the subthreshold recombination is enhanced at high $T_{1/2}$. The fraction of the stage-I recovery also decreases gradually as $T_{1/2}$ increases. It is associated with the decrease in the damage efficiency. The dependence of the damage efficiency, the recombination volume and the stage-I recovery on $T_{1/2}$ for α-Fe is similar to that for some fcc metals [4–7].

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