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Recovery of electrical resistivity of high-purity iron irradiated with 30 MeV electrons at 77 K

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Abstract

The characteristics of radiation-induced defects in high-purity iron subject to 30 MeV electron irradiation at 77 K have been studied using electrical resistivity recovery measurements. For lowest electron dose, the stage II consists of two substages at 165 K and at 180 K. With increasing dose, the 165 K stage remains unshifted whereas the 180 K stage shifts to lower temperatures. These substages are attributed to the free migration of di-interstitials within and outside the displacement cascade, respectively. For higher electron doses, a slight substage (180–190 K) may be associated with the correlated annihilation of vacancies observed after fast neutron irradiation. Furthermore, another marked stage (210–240 K) with the dose dependence of peak temperature shift is ascribed to the monovacancy migration. From these results, it has been verified that the radiation damage produced by 30 MeV electrons comprises two types of defect structures; one associated with displacement cascades and the other one associated with simple displacement processes.

1. Introduction

The use of high energy electrons is effective for producing radiation damage in solids. Their usefulness results from several factors: they have a large penetrating ability and it is believed that they produce a simpler type of damage than heavy particles such as fast neutrons or ions. For electrons with energy of the order of 30 MeV, however, the maximum kinetic energy transferred to primary knock-on atoms (PKAs) exceeds 35 keV in the case of iron, for instance. This energy is comparable to the average PKA energy induced by fast neutrons. This leads us to examine the differences in defect concentration and spatial distribution from low energy electron irradiation. In order to investigate the characteristics of radiation-induced defects in metals subject to 30 MeV electron irradiation, iron has been adopted as an appropriate metal because of its high stage I temperature ($\sim$ 100 K) and isochronal resistivity recovery measurements have been carried out along with those after 2.5 MeV electron and fast neutron irradiation by use of zone-refined high-purity specimens.

2. Experimental

The starting material for the present experiments was pure iron (Atomiron) obtained from Showa Denko K.K. The high-purity iron was prepared by floating zone-refining in a flowing atmosphere of wet and dry hydrogen. The detailed processes for obtaining final specimens for electrical resistivity measurements are indicated in our previous paper [1]. The residual resistance ratio RRR_H ($= \rho_{300\ K}/\rho_{4.2\ K}$ in the presence of magnetic field) of the specimens was $\sim$ 4000.

Frenkel defects and defect clusters were introduced by high energy electron irradiation (2.1×10^{21}–4.1×10^{22} e m^{-2}; $E = 30$ MeV, $T_{irr} = 77$ K) using the electron linear accelerator, by fast neutron irradiation (1.1×10^{21} n m^{-2}; $E > 0.1$ MeV, $T_{irr} = 20$ K) in low temperature loop of KUR at the Research Reactor Institute, Kyoto University, and by low-energy electron irradiation (1.0×10^{21} e m^{-2}; $E = 2.5$ MeV, $T_{irr} = 77$ K) using the Dynamitron at the Takasaki Research Establishment of Japan Atomic Energy Research Institute. For 30 MeV electron irradiation various doses were

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Table 1
Characteristics of high-purity iron specimens irradiated with 30, 2.5 MeV electrons, and fast neutrons[a]

Sample	Specimen irradiation characteristics			
	Fluence ($\times 10^{21}$ m^{-2})	$\Delta\rho_0$(nΩ m)	C_{FP} (ppm)	dpa ($\times 10^{-6}$)
30 MeV electron	2.1	0.39	13	75
30 MeV electron	3.5	0.66	22	130
30 MeV electron	8.0	1.49	50	290
30 MeV electron	20	3.80	130	730
30 MeV electron	41	7.67	260	1500
2.5 MeV electron	10	0.63	21	120
Fast neutron	1.1	0.47	16	90

[a] $\Delta\rho_0$ – radiation-induced resistivity, c_{PF} – concentration of Frenkel pairs. The quantity of c_{PF} was evaluated adopting a specific resistivity of Frenkel pairs as 0.03 nΩ m ppm^{-1} [2].

obtained by varying irradiation duration. Table 1 summarizes the irradiation conditions of the specimens.

All electrical resistivity measurements (w-shaped wire specimen; ~ 0.2 mm in diameter and ~ 40 mm in total length) were made at 4.2 K and in the presence of longitudinal magnetic field of 53 kA m^{-1}, which leads to the reduction of the ferromagnetic contribution of residual resistivity and the reproducibility of resistivity measurements to be within 3×10^{-14} Ω m using Keithley Model 181 Nanovoltmeter. The size-effect correction for the resistivity measurement was neglected.

The temperature intervals and pulse annealing durations for the isochronal anneals were as follows: 3 K/3 min, for $T = 77$–200 K; 4 K/4 min, for $T = 200$–300 K; 5 K/5 min, for $T > 300$ K.

3. Results and discussion

Fig. 1 demonstrates the isochronal recovery spectra of electrical resistivity of high-purity iron subject to 30 MeV electron irradiation together with those obtained for 2.5 MeV electron and fast neutron irradiation under the conditions of similar $\Delta\rho_0$. The quantity $\Delta\rho_0$ is defined as the radiation-induced resistivity. At the end of each recovery stage: stage I; 77–150 K, stage II; 150–200 K, stage III; 200–300 K, it turns out to be the significant differences in retained resistivity between 30 and 2.5 MeV electron irradiation. In particular, the marked difference in resistivity recovery in stage I has been observed: ~ 80% for 2.5 MeV, ~ 60% for 30 MeV electron irradiation. This result reveals the preferential retention of interstitial clusters in 30 MeV electron irradiation. In comparison with electrons, the corresponding value accounts for ~ 40% in fast neutron irradiation and ~ 70% in 10 MeV proton irradiation ($T_{irr} = 17$ K) [3].

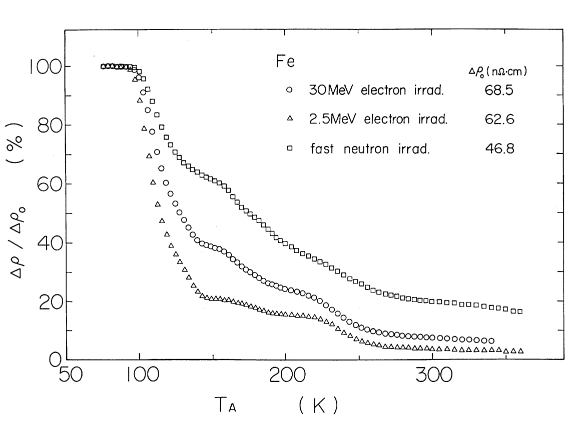

Fig. 1. Comparison of isochronal recovery spectra of high purity iron subject to 30 MeV electron irradiation with those of 2.5 MeV electron and fast neutron irradiation.The amount of $\Delta\rho_0$ indicates induced resistivity by irradiation.

Fig. 2 shows the differential recovery spectra corresponding to Fig. 1, and Figs. 3–5 the variation of recovery spectra with increase of electron dose in stages I–III, where $\Delta\rho_{I}$, $\Delta\rho_{II}$ and $\Delta\rho_{III}$ represent overall recovery in stages I, II and III, respectively.

As revealed in Figs. 2 and 3, the two substages observed after 30 MeV electron irradiation correspond to the correlated annihilation stage I_{D2} (~ 108 K) and uncorrelated stage I_E (~ 120–150 K) appearing in

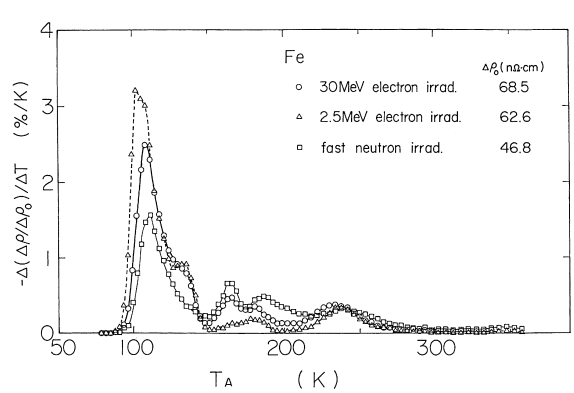

Fig. 2. Comparison of differential recovery spectra corresponding to Fig. 1.

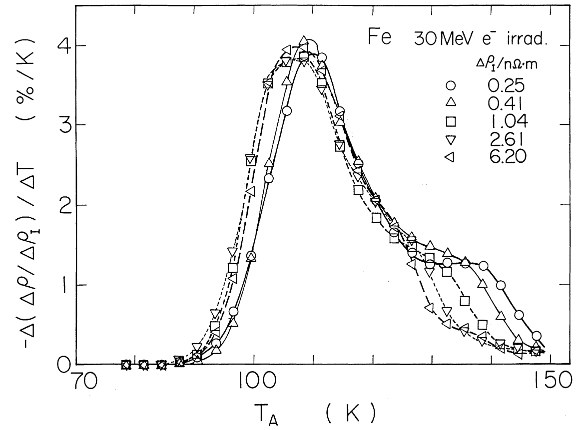

Fig. 3. Dependence of $\Delta\rho_{\mathrm{I}}$ on differential recovery spectra between 77 and 149 K of high purity iron irradiated with 30 MeV electrons.

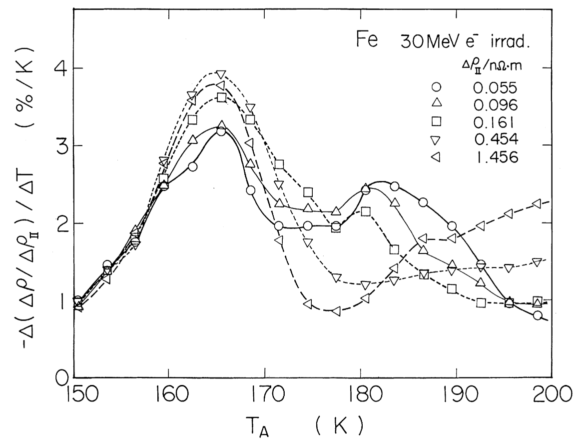

Fig. 4. Dependence of $\Delta\rho_{\mathrm{II}}$ on differential recovery spectra between 149 and 200 K of high purity iron irradiated with 30 MeV electrons.

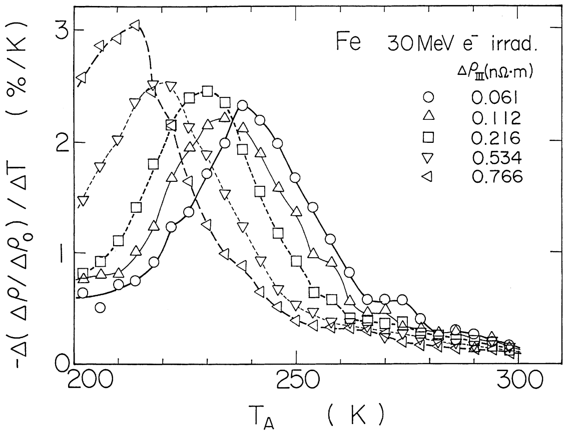

Fig. 5. Dependence of $\Delta\rho_{\mathrm{III}}$ on differential recovery spectra between 149 and 200 K of high purity iron irradiated with 30 MeV electrons.

ordinary electron irradiation [4]. The ‘correlated’ refers to the recombination of an interstitial with its own vacancy, and the ‘uncorrelated’ the recombination of an interstitial with another vacancy different from the place where it was created. The correlated one, which lowers down slightly its peak temperature with dose increase, appears also in fcc metals subject to α-particle or fast neutron irradiation [5]. On the contrary, the uncorrelated one has the marked dose dependence of peak temperature shift characteristic of low energy electron irradiation [4].

Figs. 2 and 4 have provided the evidence that the 180 K stage appearing at the lowest dose shifts its position toward lower temperatures with dose increase and at higher doses merges into another 165 K stage relevant to fast neutron irradiation [6,7]. For the 165 K recovery stage, an activation energy of 0.42 eV has been determined by the slope change method. This value is in good agreement with those obtained with fast neutron irradiation by Matsui et al. [7] and electron irradiation by Takaki et al. [4], which leads to the ascription of this stage to di-interstitial migration. This allows us to regard these two recovery stages as those due to di-interstitial migration, respectively, within and outside the displacement cascades.

Furthermore, at higher doses, the indication of another discernible recovery (180–190 K) has been found to be in the same temperature range as the correlated monovacancy migration stage observed after fast neutron irradiation [6,7]. This slight and broad stage corresponds to the one observed in positron annihilation life-time experiment [6]. This is attributed to the correlated monovacancy migration stage within the displacement cascades because of the increase in the long life-time component.

On the other hand, Fig. 5 represents the appearance of another marked stage (210–240 K) with the dependence of the peak shift on the dose. On the basis of the additional result that the activation energy of this stage (0.56 eV) is in good agreement with those corresponding to low electron [4] and fast neutron irradiation [7], it is verified that this stage is ascribed to the monovacancy migration within and outside the displacement cascades.

Fig. 6 shows the fraction of displacements produced by PKAs of energy up to T in iron for 30 MeV electrons together with those for low energy 2.5 MeV electrons and fast neutrons [8]. In this figure, E_{d}, T_{m} and $v(T)$ represent the average threshold energy for single displacement, the maximum PKA energy and the number of displacements induced by one PKA of energy T, respectively and 40 eV is adopted as E_{d} [9]. For the damage production by electrons the differential

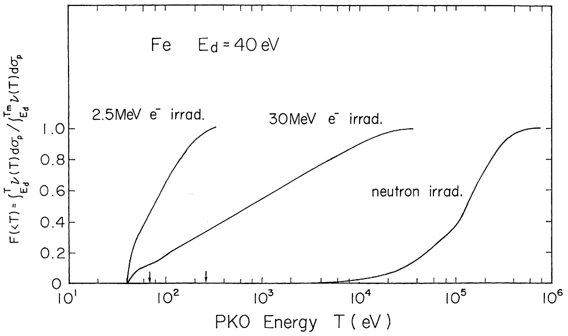

Fig. 6. Comparison of the fractional damage produced by PKA energy less than T for iron subject to 30 MeV electron irradiation with those subject to 2.5 MeV electron and fast neutron irradiation. A value of 40 eV is adopted as the average threshold energy for single displacement. In this figure, the fractional damage relevant to fast neutrons is quoted from a paper by Marwick [8].

cross-section $\sigma_p(T)$ derived by Mckinley and Feshbach [10,11] was used. If a value of 1 keV is assumed for the threshold energy of PKAs producing displacement cascades, the fraction of damage production by PKAs greater than 1 keV is found from this figure to exceed 40% of the overall damage. The average number of surviving interstitials in cascades produced by PKAs with energy (5–6 for 1 keV, 20 for 5 keV, 30 for 10 keV, 60 for 20 keV) [12], however, is substantially smaller compared with ~100 in the average clusters produced by fast neutrons. Hence the radiation-induced damage consists of almost randomly distributed Frenkel pairs and typical small displacement cascades.

For bcc metals, the cascade structures have been extensively [12–16] studied only for iron. The recent molecular dynamics (MD) simulation of the cascades in iron (simulation at 100 K) showed that surviving interstitials are distributed at the periphery of the cascades and the large fraction of them is mono- or di-interstitials, and vacancies are concentrated in the cascade core, in contrast but the possibility of significant vacancy clustering [15,16] is not predicted.

Peisl et al. [17], by means of diffuse X-ray scattering technique, studied the distribution of interstitials in iron after fast neutron irradiation at 4.5 K and obtained the results that the interstitials in a typical displacement cascade are primarily present as singles and ~ 50% di-interstitials. On the other hand, Vetrano et al. [18] observed small loops attributable to collapsed vacancy clusters, in iron subject to 100 keV self-ion irradiation at 30 K. These experimental results are in fairly good agreement with MD simulation.

The enhanced defect retention in Fig. 1 after stage I for 30 MeV electron irradiation in comparison with 2.5 MeV electron irradiation and the marked stage at 165 K due to di-interstitial migration provide the evidence of the high concentration of di-interstitials in the displacement cascade.

As for vacancy clusters, from the long life-time component observed by positron annihilation (~ 200 ps) after fast neutron irradiation at 77 K [6] compared with ~ 180 ps after low energy electron irradiation [19], the presence of small clusters (di- or tri-vacancies as the average) [20] was confirmed.

On the contrary, Kawaguchi et al. [21] observed no appreciable increase in positron annihilation lifetime in the as-irradiated state of high-purity specimens and the subsequent pronounced increase between 150 and 200 K (correlated vacancy migration stage) after 30 MeV electron irradiation at 77 K. This is consistent with the MD simulation result [22] that no significant vacancy clustering occurs in iron for cascades of up to 5 keV in energy since the fractional displacements associated with these cascades occupy ~ 80% of the total displacements as shown in Fig. 6. This suggests that most of the vacancies produced in the cascade core by 30 MeV electrons are not in the form of clusters but distributed in close proximity to each other.

4. Conclusion

The conclusions obtained from the electrical resistivity spectra of high-purity iron irradiated with 30 MeV electrons are as follows:

1. The recovery spectra consist of three stages corresponding to the migration of single interstitials (77–150 K), di-interstitials (150–200 K) and vacancies (180–240 K).
2. Each stage has two substages; the one appearing at higher temperatures characteristic of low energy electron irradiation with the dependence of the peak temperature shift on the electron dose and the other appearing at lower temperatures characteristic of fast neutron irradiation with no dependence of the peak temperature shift on the electron dose.
3. The stages associated with low energy electron and neutron irradiation correspond to the radiation damage recovery outside and within the displacement cascades, respectively.

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