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Journal of Nuclear Materials 283–287 (2000) 282–285

journal of nuclear materials

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Defect structures introduced in iron under varying temperature neutron irradiation

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Abstract

Defect structures in iron under varying temperature neutron irradiation and those under constant-temperature irradiation were examined by a transmission electron microscope (TEM). For irradiation at a low temperature (473 K), microvoids were detected by positron annihilation lifetime (PAL) measurement. For irradiation at a high temperature (673 K), with increasing the irradiation dose a few interstitial (I)-type dislocation loops grew and the total number density of the loops decreased. In a varying temperature irradiation between 473 and 673 K, after a shift to the high temperature the absorption of interstitials to I-type dislocation loops was suppressed by microvoids introduced at the low temperature and the loops shrank or disappeared by vacancies released from microvoids. As the cycle of varying temperature irradiation became short, the effect of the suppression of the defect development became small.

1. Introduction

Although the damage structure in neutron irradiated iron is not well understood yet, the characteristics that have been clarified so far [1–4] are as follows: (1) compared to metals and alloys with a face centered cubic structure, the formation efficiency of defect clusters is quite low; (2) voids are observed only at 623 K by electron microscopy; (3) invisible defect clusters exist at low temperatures (<473 K) and (4) irregular shaped dislocation loops are formed at high temperatures (~673 K).

In this paper, we examined the microstructural evolution in iron under varying temperature irradiation in which the temperature is changed between the higher and lower of characteristic temperature (573 K) at which small vacancy clusters become unstable [4], and under constant-temperature irradiation at the higher and the lower temperatures in the Japan Material Testing Reactor (JMTR).

2. Experimental procedure

Iron specimens examined were 99.99% purity from Johnson-Matthey Chemicals. Specimens of 3 mm in diameter and of 0.1 mm thickness were heat treated under dry hydrogen flow at 970 K for 30 min. The grain size of specimens was 0.05 mm. The specimens were irradiated under constant temperature at 473 and 673 K, and under varying temperature between about 473 and 673 K with three various cycles (irradiation numbers: 94M-13U, 95M-5U and 91M-26US) in the JMTR. The irradiation dose ranged from 2.0×10^{21} to 1.7×10^{24} n/m^2 (>1 MeV). Fig. 1 shows diagrams of varying temperature irradiation for the 94M-13U and the 95M-5U. Arrows show points at which specimens were pulled with a specially designed irradiation rig [5]. Numbers surrounded by circle correspond to those shown in Figs. 4 and 5. In the 91M-26US, specimens were irradiated between 473 and 673 K with a cycle of 44 h [6]. After

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0022-3115/00/$ - see front matter
PII: S0022-3115(00)00105-7

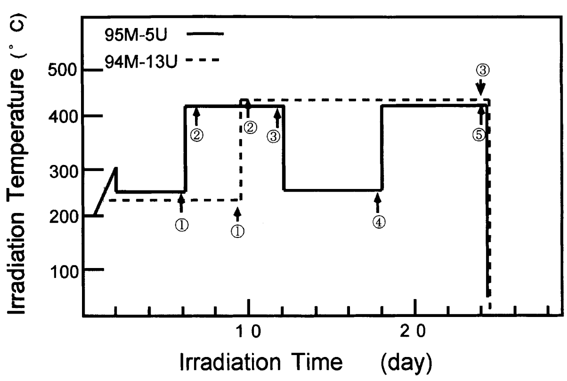

Fig. 1. Diagrams of varying temperature neutron irradiations for the 95M-5U and the 94M-13U in the JMTR.

irradiation, defect microstructures were observed with a 200 kV transmission electron microscope (TEM), JEM-200CX. For some specimens, positron annihilation lifetime (PAL) was measured. All the PAL measurements were performed at room temperature using a conventional apparatus with a time resolution of about 250 ps. After source-background subtraction, lifetime spectra were resolved into two lifetime components using the fitting program POSITRONFIT [7]. The standard deviation of the fits was less than 3%. Before irradiation the lifetime of the specimen was 107–109 ps and was not resolved into two components.

3. Results and discussion

3.1. Defect accumulation processes in constant-temperature irradiation

In specimens irradiated at 473 K, no defect clusters were observed up to 8.0×10^{22} n/m^2 by electron microscopy. As the irradiation dose increases more than 4×10^{23} n/m^2, very small defect clusters (<5 nm) began to form and their number density increased with the irradiation dose. These small clusters had been identified previously to be of interstitial (I)-type [1]. Fig. 2 shows the change of PAL in the specimens irradiated at 473 K. The existence of microvoids was detected from an appearance of long lifetime component of PAL (τ_2) even in a specimen irradiated to the lowest dose (2×10^{21} n/m^2) and the intensity of long lifetime component decreased at the highest dose. This decrease may be due to coarsening of microvoids by increasing the dose.

Fig. 3 shows progressive changes of microstructures in specimens irradiated at 673 K. In the specimens irradiated at 673 K, considerably large dislocation loops were observed even in a specimen irradiated to the lowest dose (1.1×10^{23} n/m^2). The nature of the loops is I-type. As the irradiation proceeds, a few of the loops

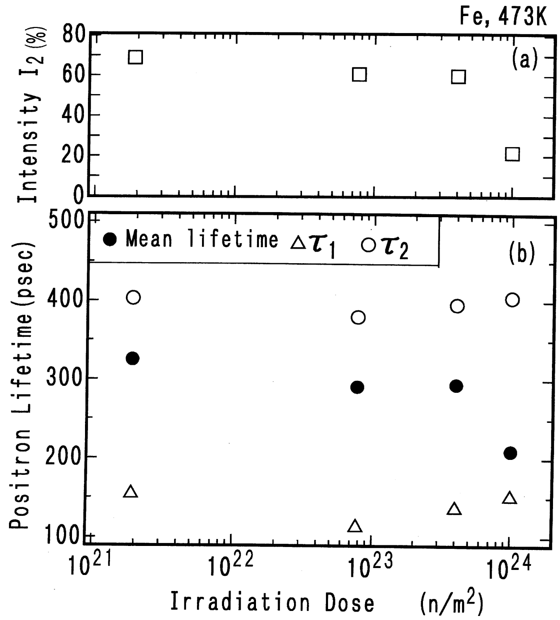

Fig. 2. Change of the positron annihilation lifetime in neutron irradiated iron at 473 K. τ_1 is the lifetime of matrix component. τ_2 and I_2 are the lifetime and intensity of long lifetime component, respectively.

grew very large selectively, while the size of the other loops did not change very much and the total number density of loops decreased. Voids and microvoids were not found by electron microscopy and PAL measurement, respectively. Although the mechanism of selective growth is not well understood, larger loops grow by absorbing more interstitials than smaller loops and the smaller I-type loops shrink or disappear by the reaction with vacancies remained in matrix.

3.2. Defect accumulation processes in varying temperature irradiation

Figs. 4 and 5(a) and (b) show defect structures and the change of the size distribution of I-type loops, respectively at each step of the 95M-5U. In a specimen irradiated at 533 K for about 5.6 days, I-type dislocation loops having an irregular shape formed. By an irradiation for 1 day after a shift to 693 K, the number density of the loops having a relatively small size (less than 40 nm) decreased but some loops grew as shown in Fig. 5(a) (1 → 2). The shrinkage of small I-type loops was also observed in a shift from a low temperature to a high temperature in another varying temperature irradiation, 94M-13U (Fig. 5(c); 1 → 2). Although the initial size distributions of the loops in the 95M-5U and the 94M-13U are slightly different because of the difference of the low irradiation temperatures, similar behavior is

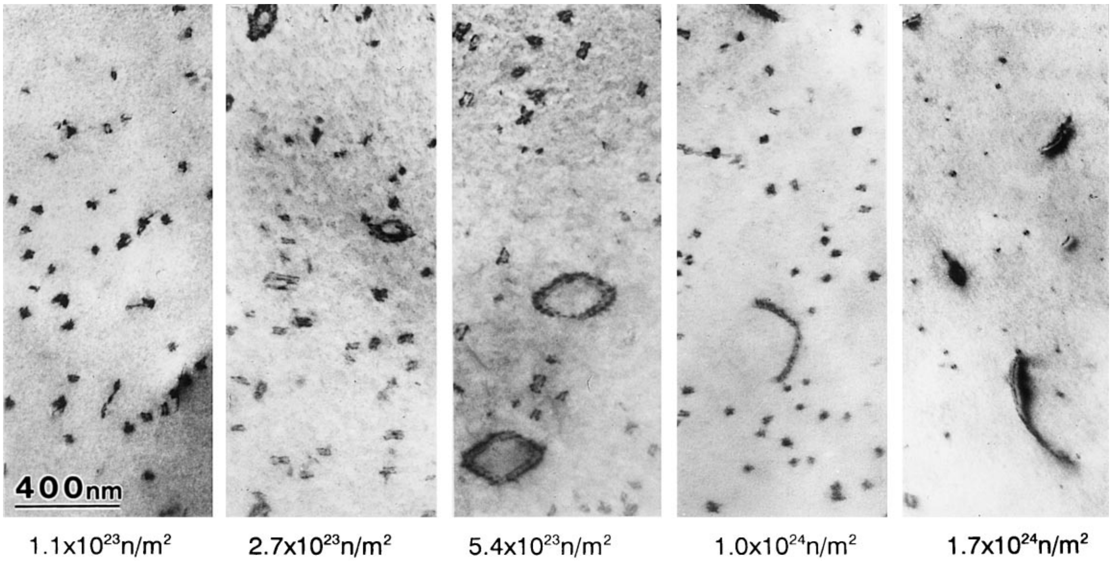

Fig. 3. Progressive change of microstructures introduced in neutron irradiated iron at 673 K.

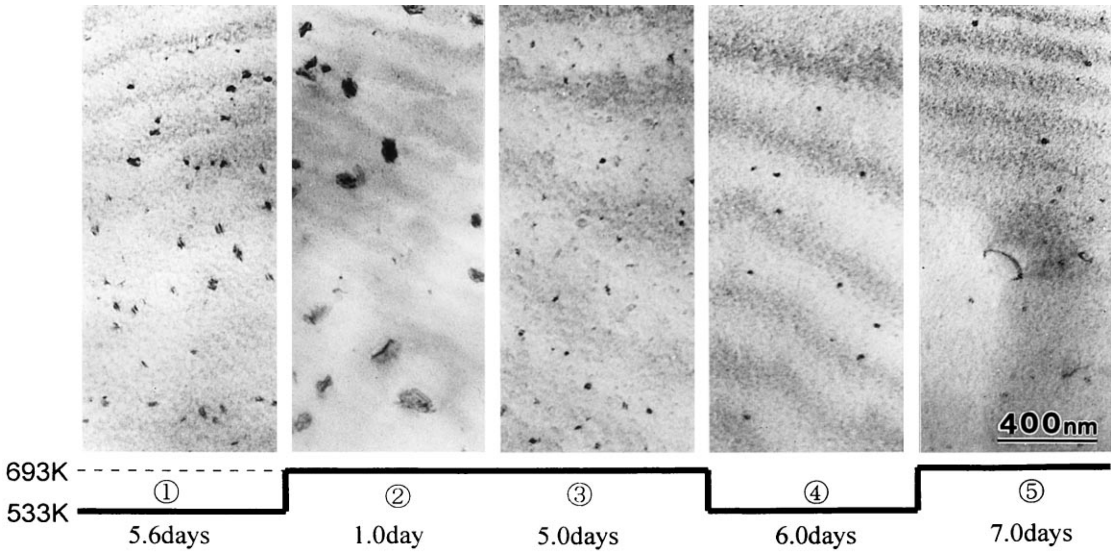

Fig. 4. Defect structures introduced by a varying temperature irradiation with a temperature combination of 533/693 K (95M-5U).

observed. In the case of 94M-13U, the shrinkage of I-type loops occurs for only 3 h irradiation after the shift from the low temperature to the high temperature. Hence this process may be induced by the migration of very unstable vacancy (V)-type defects due to an annealing during temperature shift as pointed out by Xu [8].

In a subsequent irradiation at the high temperature of the 95M-5U, selective growth of a few dislocation loops observed in the constant-temperature irradiation at 673 K was not found at all and the size of loops decreased homogeneously (Fig. 5(a); 2 → 3). The absorption of interstitials to the loops was obviously suppressed by microvoids introduced at low temperatures and I-type dislocation loops shrank by the reaction with vacancies released from microvoids.

3.3. The effect of the cycle in varying temperature irradiation

We reported the damage structures of iron during the 91M-26US in a previous paper [1]. By the comparison of the defect structures among three varying temperature

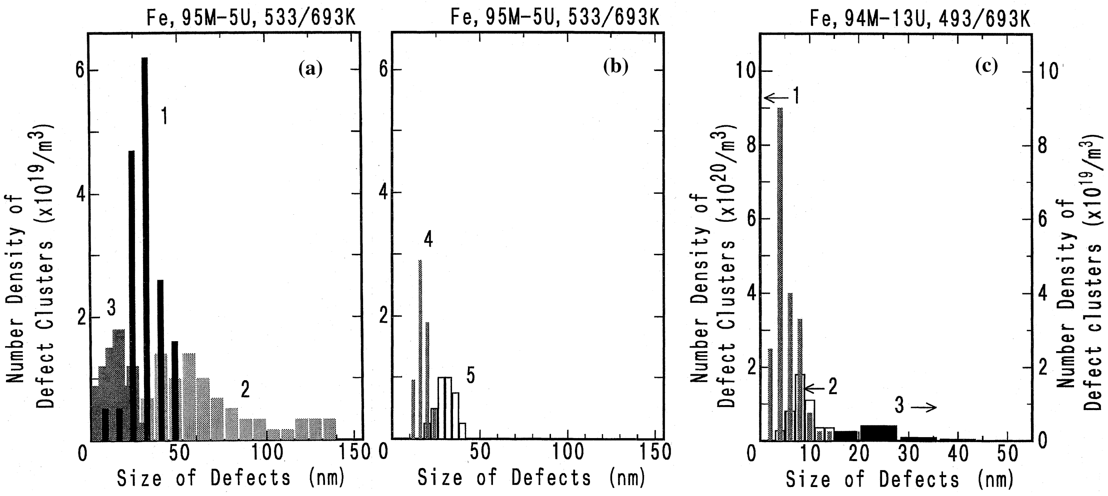

Fig. 5. Change of the size distribution in two varying temperature irradiations (95M-5U and 94M-13U). The numbers in the figure correspond to those in Figs. 1 and 4.

irradiations, the size and the number density of I-type loops became larger with decreasing the cycle of the varying temperature. As mentioned in Section 3.2, a characteristic process during the varying temperature irradiation (473/673 K) is the suppression of the growth of I-type loops at high temperatures by microvoids introduced at low temperatures. If varying temperature is repeated, the effect of repeating is also included. As shown in Fig. 5(a) and (b), in the first cycle of the 95M-5U, only shrinkage of the I-type loops was observed. But in the second cycle the growth of I-type loops was observed. While in the case of the 94M-13U, it is considered that most of I-type loops introduced at low temperature shrink once and only a few loops grow at latter stage of the irradiation at the high temperature, as shown in Fig. 5(c). But I-type loops hardly grow. Therefore the high concentration of microvoids at low temperatures is thought to induce heavy suppression of the development of I-type loops.

4. Conclusions

By comparing the defect structures under varying temperature irradiation at about 473/673 K and under constant-temperature irradiation at 473 and 673 K, the following facts are clarified.

For irradiation at 473 K, microvoids formed even in a specimen irradiated to a quite low dose and the growth of microvoids occurred concurrent with the formation of I-type defect clusters. For irradiation at 673 K, selective growth of a few I-type dislocation loops was observed as irradiation proceeds. In the varying temperature irradiation (473/673 K), the growth of I-type loops was suppressed at the high temperature by microvoids introduced at the low temperature. As the cycle of varying temperature became long, the development of defect clusters was suppressed very strongly.

Acknowledgements

The authors would like to thank to the staff of Oarai branch of Tohoku University for the reactor irradiation of the specimens and post-irradiation experiments.

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