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Invisible and visible point defect clusters in neutron irradiated iron

M. Horiki [a,*], T. Yoshiie [b], M. Iseki [a], M. Kiritani [c]

[a] Department of Nuclear Engineering, School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-01, Japan
[b] Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka-fu 590-04, Japan
[c] Department of Electronics, Institute of Hiroshima University, Miyake, Saiki-ku, Hiroshima 731-51, Japan

Abstract

Microstructure evolution in a low-dose neutron-irradiated iron was examined with a transmission electron microscope (TEM). The characteristics of the temperature dependence of defect structures in irradiated iron were as follows, (a) rather lower number density of defect clusters at 473 K than at 573 K, (b) development of dislocation loops and voids at an intermediate temperature of 623 K and (c) formation of irregularly shaped dislocation loops at 673 K. Comparing the defect structures produced by the irradiation with a conventional temperature control and that with an improved temperature control, nucleation of defect clusters was suppressed under neutron irradiation at 473 K. From the defect structures introduced by irradiations in which the irradiation temperature was cyclically changed between two temperatures (T-cycle irradiation), the accumulation of a large number of invisible vacancy type defect clusters by irradiation at 473 K was strongly suggested.

1. Introduction

Iron based alloys, for example ferritic steels, which have a bcc structure are considered as possible structural materials for fission reactors or future fusion reactors. However, in spite of many investigation, their fundamental microstructure evolution under neutron irradiation has not been well understood. For an understanding of the damage structure of these alloys, it is essentially important to clarify the damage structure of iron. Kuramoto et al. [1] suggested the existence of microvoids in neutron irradiated iron from positron annihilation lifetime measurements. One of the authors detected electron microscopically invisible defect clusters in iron irradiated by RTNS-II (Rotating Target Neutron Source) [2].

In this paper, the microstructural evolution in iron neutron-irradiated in the Japan Material Testing Reactor (JMTR) was examined with a transmission electron microscope (TEM). Details of visible and invisible defect clusters are discussed from the results of constant temperature irradiation and temperature-variation irradiation.

2. Experimental procedure

Iron specimens of 99.99% purity were obtained from Johnson-Matthey Chemicals Ltd. The residual resistivity ratio (RRR_H) was about 1000. Punched specimens of 3 mm in diameter were heat-treated under dry hydrogen flow at 970 K for 30 min. Neutron irradiation was performed in JMTR with an improved temperature control in which the sample temperature is maintained at constant temperature during the entire irradiation, and a conventional temperature control [3]. Irradiation temperatures ranged from 473 to 673 K while irradiation dose was about 1.0×10^{24} n/m^2 (>1 MeV). In addition to constant temperature irradiations, temperature variation neutron irradiations in which the irradiation temperature was cyclically changed between two temperatures (T-cyclic irradiation) [4] were also performed.

* Corresponding author. Tel.: +81-52 789 3609; fax: +81-52 789 4685; e-mail: m-horiki@nucl.nagoya-u.ac.jp.

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After irradiation, specimens were electro-polished and defect microstructures were observed with a JEM-200CX operated at 200 kV. In order to identify the nature of defect clusters, neutron-irradiated specimens were irradiated in an ultra-high voltage electron microscope operated at 1000 kV, H-1250ST at Nagoya University.

3. Results and discussion

3.1. Defect structures under constant temperature irradiation

3.1.1. Temperature dependence of defect structures

Fig. 1 shows the defect structures introduced in iron by irradiation with an improved temperature control at various temperatures. The microstructures in the specimens irradiated at 473 and 573 K were very small defect clusters. At an intermediate temperature of 623 K, dislocation loops were developed and voids were observed only at this temperature. At 673 K, the shape of dislocation loops changed to an irregular one.

Fig. 2 shows the size distribution and the number density of defect clusters. The number densities of defect clusters were normalized to the irradiation dose of 1.0×10^{24} n/m^2 by the assumption that the number density of interstitial (I)-loops was proportional to irradiation dose [5] and are indicated in each figure. In the specimen irradiated at 473 K, the number density of defect clusters is rather lower than that irradiated at 573 K. The result that the number density of clusters decreases with lowering temperature has not been found in fcc metals and alloys [3]. On the other hand, the size of defect clusters becomes larger with increasing irradia-

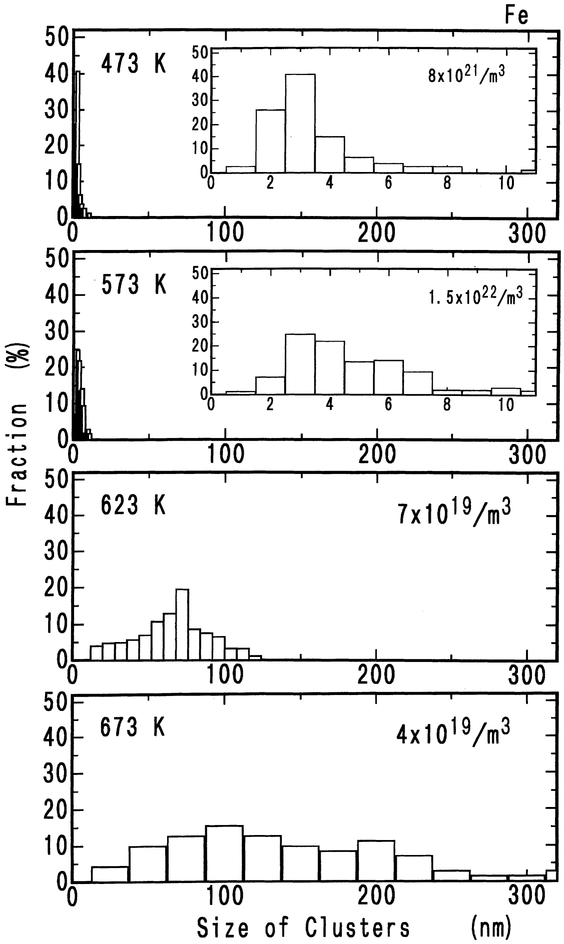

Fig. 2. Variation of the size distribution and the number density of defect clusters in the iron irradiated.

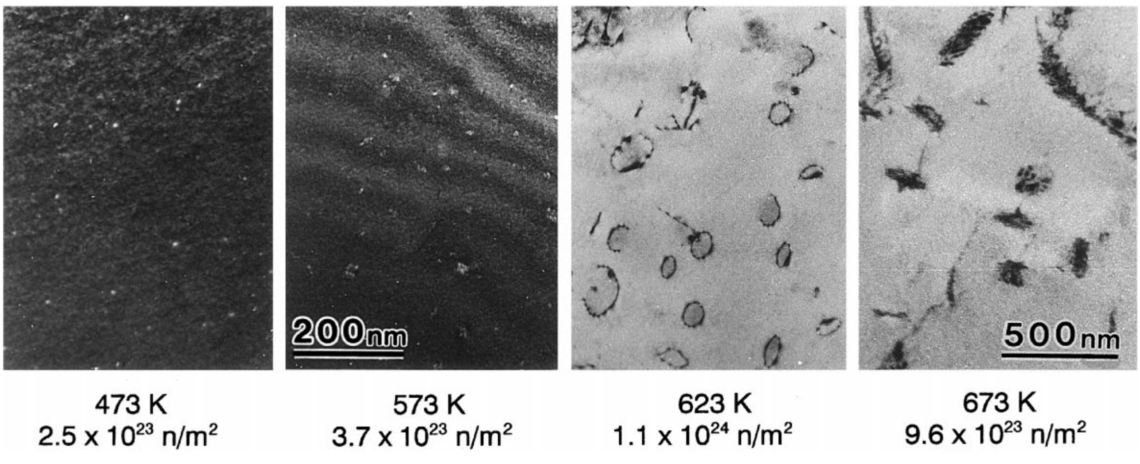

Fig. 1. Defect structures of iron neutron-irradiated at four temperatures. Images of specimens irradiated at 473 and 573 K have the same magnification and images of specimens irradiated at 623 and 673 K have the same magnification.

tion temperature, especially at higher temperature than 623 K.

3.1.2. Defect structures near dislocations at 473 K

Though the number of defect clusters in the matrix of the specimen irradiated at 473 K was considerably low, as shown in Fig. 1, a high number density of defect clusters were observed at one side of a dislocation.

Recently, we have established a method to identify the nature of defect clusters in neutron-irradiated specimens by means of electron irradiation [6]. Fig. 3 shows progressive change of microstructure near a dislocation in a specimen neutron-irradiated at 473 K during electron irradiation at 473 K. By electron irradiation, almost all of clusters including those far from the dislocation grow. A couple of grown clusters are indicated by arrows in each photograph. Since interstitial atoms are dominant at an early stage of electron irradiation [7], I-type clusters grow and V-type clusters shrink or disappear. From the behavior of defect clusters during electron irradiation, the nature of clusters formed near the dislocation as well as in the matrix is concluded to be of I-type.

The dominant formation of defect clusters near a dislocation was observed in nickel and copper irradiated with RTNS-II. Satoh showed by calculation [8] that migration efficiency (the product of the mobility and concentration of point defects) of interstitials is larger than that of vacancies at the dilatational side of a dislocation and vice versa at the compressional side of a dislocation for copper neutron-irradiated at 473 K. In the case of iron, similar to nickel and copper, a high number density of I-type clusters are formed at the dilatational side of a dislocation due to the dominant reactivity of interstitials.

3.2. Defect structures under temperature-variation irradiation

3.2.1. Comparison of defect structures produced by irradiation with the conventional temperature control and the improved temperature control

Fig. 4 shows microstructures produced by the conventional irradiation and by the improved irradiation both at 573 and 673 K. The number density of defect clusters in a specimen irradiated by the conventional irradiation at 573 K was lower than that by the improved irradiation. On the other hand, the number density of defect clusters by the conventional irradiation at 673 K was higher than that by the improved irradiation. The conventional temperature control irradiation at 573 and 673 K had a transient irradiation below each designed temperature for 1 day. From these results, the nucleation of defect clusters is suppressed during transient irradiation below 573 K but the suppression effect disappears during transient irradiation between 573 and 673 K.

3.2.2. Defect structures by T-cycle irradiation

Fig. 5 shows defect structures produced by two T-cycle irradiations with a combination of six cycles of irradiation at 473 K for 44 h and 673 K for 44 h (473/673 K), and at 573 and 673 K (573/673 K). The formation of well-developed I-type dislocation loops at 473/673 K and at 573/673 K are similar to those produced at 673 K. Fig. 6 shows the size distribution and the number density of defect clusters after two T-cycle irradiations.

The number density of clusters by 473/673 K is nearly the same as that produced at 673 K, but the average size of clusters at 473/673 K is much smaller than that at 673 K. This means that the growth of I-type dislocation loops is much suppressed during irradiation at 473/673

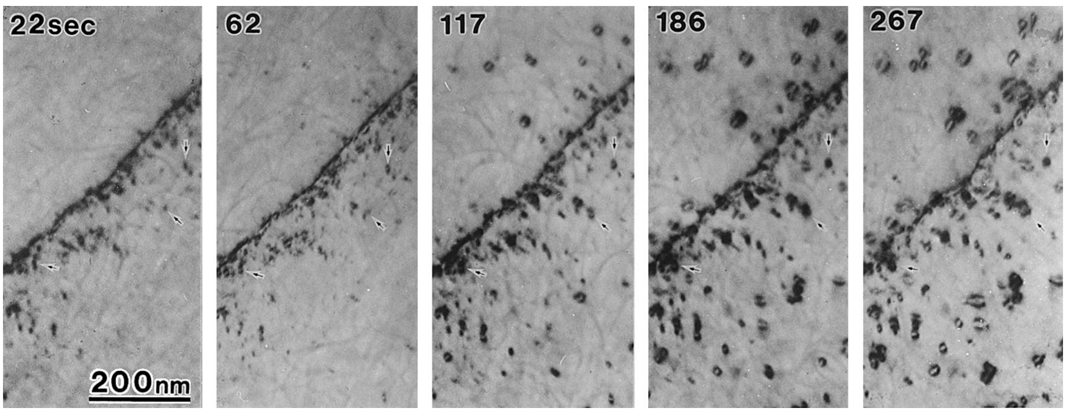

Fig. 3. Progressive change of microstructure near a dislocation in a neutron-irradiated iron (1.0×10^{24} n/m^2) at 473 K during electron irradiation (1000 kV, 2.0×10^{22} e/m^{2}s) at 473 K.

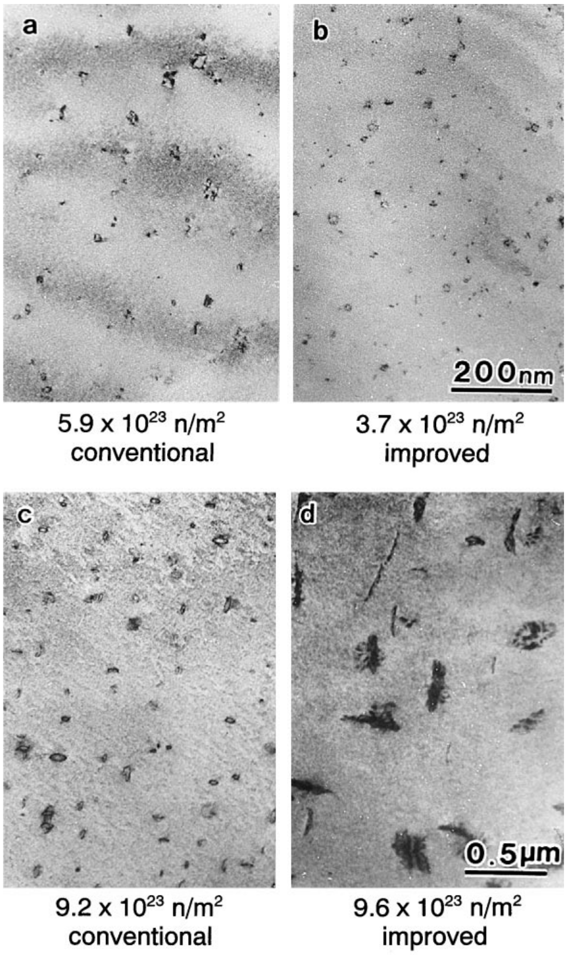

Fig. 4. Comparison of defect structures produced by irradiation between with the conventional temperature control and the improved temperature control at 573 K (a) (b) and 673 K (c) (d).

K comparing with that at 673 K. While the number density of clusters at 573/673 K is larger than that at 673 K, the average size of clusters at 573/673 K is smaller than that at 673 K. These results are understood as follows. The nucleation of I-type dislocation loops is suppressed by the irradiation at 473 K and promoted by the irradiation at 573 K, consistent with the results of the Section 3.2.1.

We reported in a previous paper [4] that in fcc metals and alloys irradiated by T-cycle irradiation the formation of I-type dislocation loops was extremely suppressed by the decomposition of V-type clusters at high temperature which had previously been introduced at low temperature. Since the number density of interstitials in I-type dislocation loops produced by 473/673 K is much lower than that at 673 K, a similar effect of V-type clusters is expected. Thus, it is strongly suggested

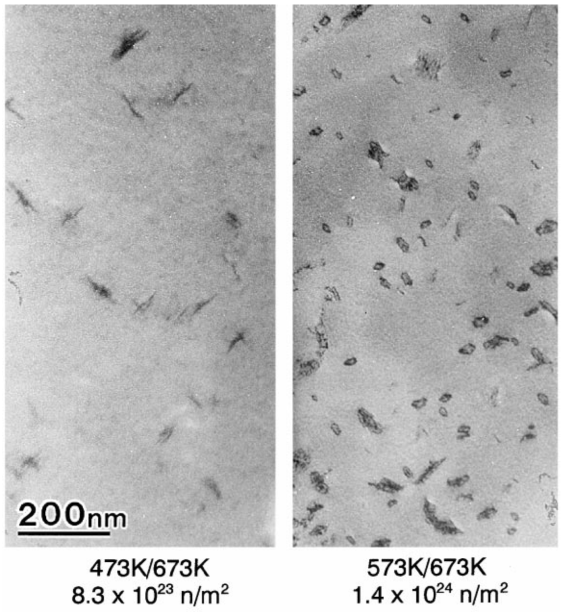

Fig. 5. Defect structures by two T-cycle irradiations with a combination of irradiation at 473 and 673 K and a combination of irradiation at 573 and 673 K.

that a large number of invisible V-type clusters are accumulated at low temperature (473 K), and evaporated at high temperature, resulting in the suppression of the growth of I-type dislocation loops. V-type defect clusters are not easy to nucleate at 573 K and only small number of voids are thought to nucleate. Actually voids are observed by irradiation at 623 K. Voids will not

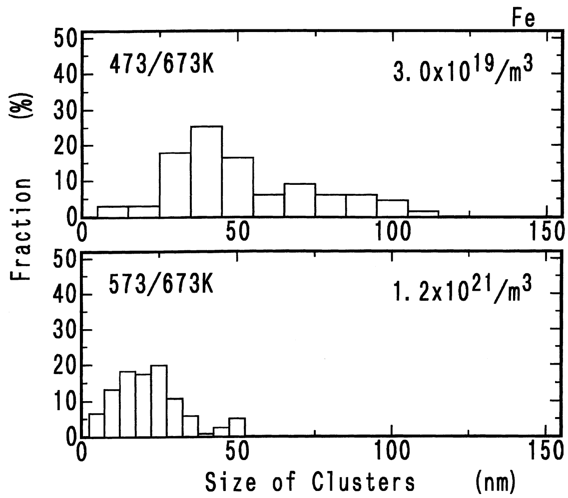

Fig. 6. Size distribution and number density of defect clusters produced by the two T-cycle irradiations.

suppress the growth of I-type dislocation loops at the higher temperature of T-cycle irradiation.

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