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Temperature dependence of one-dimensional motion of interstitial clusters in Fe and Ni

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Abstract

The temperature dependence of one-dimensional (1-D) motion of interstitial clusters and void growth in Fe and Ni was studied to a dose of 0.3 dpa. Two criteria were applied to detect the 1-D motion of interstitial clusters. One is the formation of interstitial type dislocation loops near the dilatational side of edge dislocations. The other is no formation of interstitial type dislocation loops near grain boundaries. In neutron irradiated Ni with improved temperature control at 573 K, the 1-D motion of interstitial clusters existed. However no evidence of 1-D motion was found for a conventional temperature controlled irradiation, where the specimen temperature changed with reactor power. This suggested that there was little 1-D motion of interstitial clusters at lower temperatures during the start-up of the reactor. In neutron irradiated Fe, the 1-D motion of interstitial clusters occurred at 473 K and 623 K, judging from the criteria. A good correlation existed between the 1-D motion of interstitial clusters and void growth. These results are discussed based on the temperature dependence of the interaction between impurities and interstitial clusters.

1. Introduction

The importance of one-dimensional (1-D) motion of interstitial clusters in defect structure evolution has been recognized for many years [1–3]. Computer simulation showed that the activation energy for 1-D migration was very low, lower than that of a single interstitial when the cluster contained less than 100 interstitials in Fe or Ni [4]. No temperature dependence is therefore expected above room temperature. Moreover, if clusters move with the frequency of lattice vibration and activation energy lower than 0.1 eV, it is not possible to observe their movement using transmission electron microscopy (TEM). The movement has been, however, observed by many researchers [5,6]. These results indicated that their actual migration energy is high, even in pure metals. In this paper, in order to clarify the real migration of these clusters in metals, the temperature dependence of 1-D motion of interstitial clusters and void growth in Fe and Ni was studied.

We applied two criteria to detect the 1-D motion of interstitial clusters [7]. The first is decoration of edge dislocations by interstitial type dislocation loops, where many loops are observed at the

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dilatational side of edge dislocations. They are accumulated by 1-D motion of interstitial clusters [8] to the dislocation. The second criterion is no formation of interstitial type dislocation loops near grain boundaries. Interstitial clusters escape easily to grain boundaries by 1-D motion and no loops remain near there. Using these criteria, the temperature dependence of 1-D motion of interstitial clusters is discussed.

2. Experimental

Specimens irradiated were 99.99% pure Ni and Fe (from Johnson Matthey). The specimens were cold-rolled to 0.1 mm thickness and 3 mm diameter discs were punched. Nickel discs were annealed at 1170 K for 1 h in vacuum. Iron discs were annealed 970 K for 0.5 h under dry hydrogen flow. Neutron irradiations were performed in the Japan Materials Testing Reactor (JMTR) and the Kyoto University Reactor (KUR). Two specimen temperature control methods were used for the irradiation [9]. One is an improved temperature control irradiation, where the specimens were kept at the designed temperature using an electric heater. The other is a conventional temperature control irradiation, where the specimens were heated using nuclear heating and gas pressure in a heat gap between specimens and an irradiation capsule. In this control, specimen temperature increased from coolant temperature to designed temperature as the reactor power increased. The irradiation damage in dpa was calculated using a displacement threshold energy of 24 eV for both Ni and Fe.

3. Nickel

In neutron irradiated Ni with the improved temperature control at 573 K, the 1-D motion of interstitial clusters was identified, since decoration of dislocations by interstitial type dislocation loops and no loops near grain boundaries were observed [7,10,11]. An example of the absence of loops near grain boundary is shown in Fig. 1(a). In the grain interior, voids and dislocations developed from interstitial type dislocation loops are observed.

However, the accumulation of interstitial clusters near grain boundaries, which is evidence of no 1-D motion, was observed in the conventional temperature control irradiated Ni at 573 K as shown in Fig. 1(b), where the specimen temperature changed with the reactor power from 363 K to 573 K. The existence of loops depended on the nature of grain boundaries and we selected large angle boundaries in this experiment. The temperature history with reactor power is shown in Fig. 2. Analysis of defect accumulation near grain boundaries was performed based on a rate theory [7]. If mobile defects were

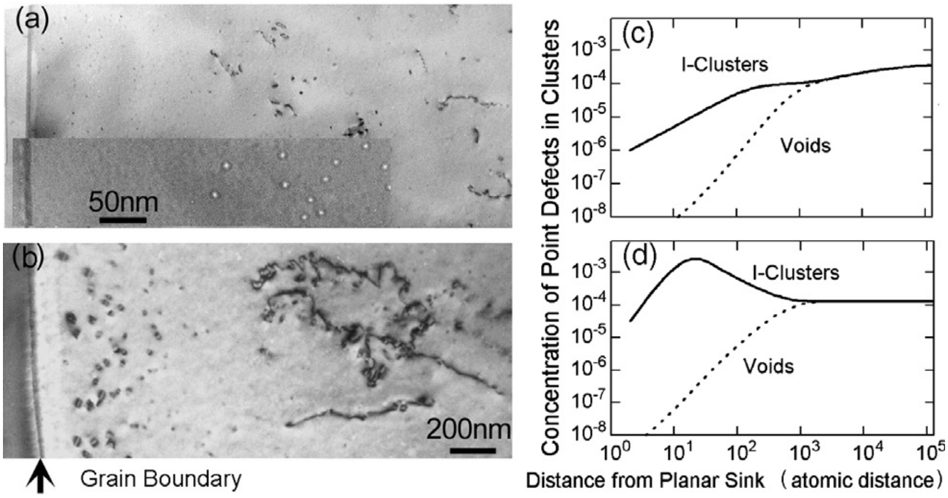


Fig. 1. Damage structures in Ni neutron irradiated at 573 K. (a) Improved temperature control irradiation to 0.091 dpa, (b) conventional temperature control irradiation to 0.14 dpa. The figure inserted in (a) is a void contrast image. (c) and (d) are the result of rate equation analysis under the migration of interstitials, interstitial clusters and vacancies, and under the migration of interstitials and vacancies, respectively.

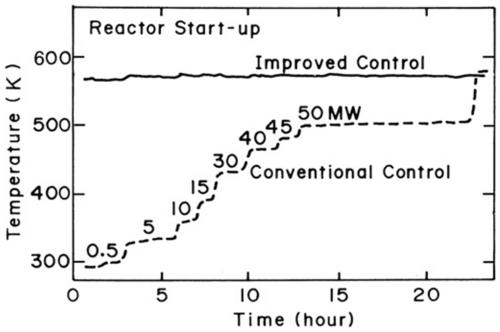


Fig. 2. Temperature history of specimens irradiated in the JMTR with improved temperature control and conventional temperature control.

vacancies, interstitials and interstitial clusters in void forming condition, the defect structure near grain boundary (Fig. 1(c)) was just the same as in Fig. 1(a). If mobile defects were interstitials and vacancies in void forming condition, interstitial type dislocation loops existed near grain boundaries as shown in Fig. 1(d), just the same as in Fig. 1(b). As the 1-D motion existed at 573 K and interstitial clusters formed at 573 K should escape to grain boundaries, loops near grain boundaries were believed to be nucleated by low temperature irradiation before the specimen temperature got to design temperature. The specimens were held at 500 K for 9 h before the adjustment of temperature to 573 K using gas pressure as shown in Fig. 2. This indicate that the 1-D motion did not occur at temperature lower than 573 K.

Another example of the conventional temperature control irradiation is shown in Fig. 3(a). In this case, Ni was irradiated at 673 K as a thin foil which was electro-polished for TEM before neutron irradiation. In the thinnest area, interstitial type dislocation loops exist. With increasing foil thickness, the defect structure changes from no defects existing area to voids and interstitial type dislocation loops coexisting area. In Ni irradiated with the improved temperature control at 673 K, at the thin area no loops existed as shown in Fig. 3(b). In Ni irradiated at 473 K with improved temperature control, many loops were observed even at the thin area as shown in Fig. 3(c). No loops near thin area in thin foil irradiation is again evidence of 1-D motion [7], since the 1-D motion of interstitial clusters does not exist at 473 K irradiation. Therefore loops in the thin area by 673 K conventional temperature control irradiation were also concluded to be nucleated by irradiation at low temperatures at around 473 K where no 1-D motion existed.

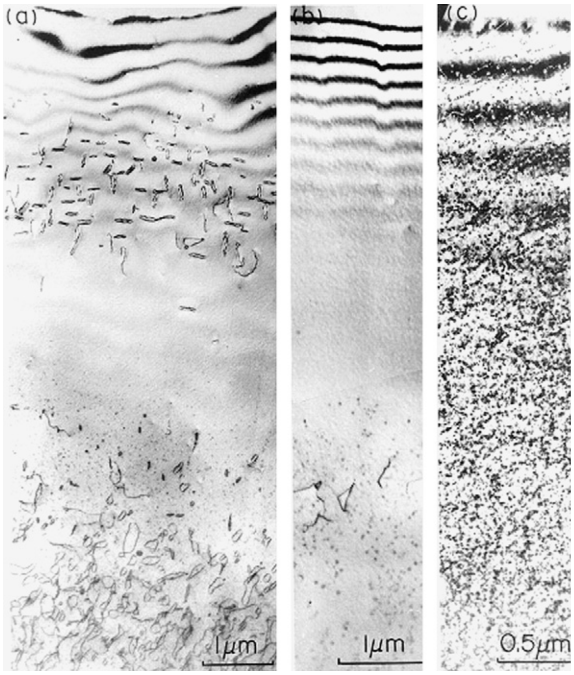
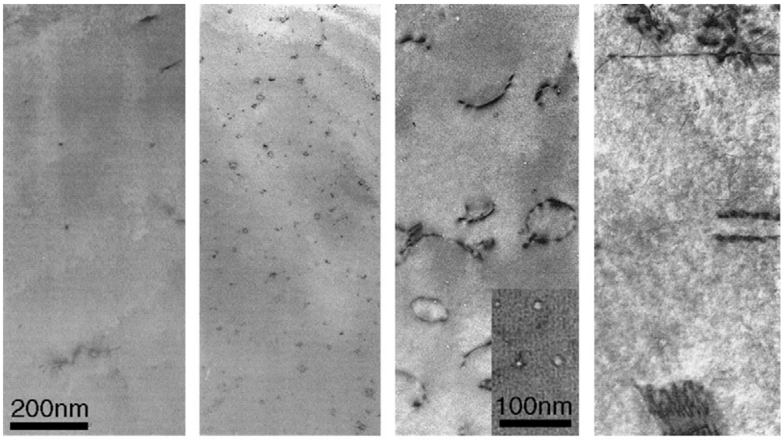


Fig. 3. Damage structures in Ni neutron irradiated as thin foils in the JMTR. (a) Conventional temperature control irradiation to 0.28 dpa at 673 K, (b) improved temperature control irradiation to 0.30 dpa at 673 K and (c) improved temperature control irradiation to 0.077 dpa at 473 K.

4. Iron

The irradiation temperature dependence of defect structures in irradiated Fe is shown in Fig. 4. All irradiations of Fe were performed with improved temperature control and the TEM observation was performed after electro-polishing of the specimens following neutron irradiation. Interstitial type dislocation loops were observed at all these irradiation temperatures. Large loops identified were Burgers vector of ⟨100⟩ type. In the irradiation range of 0.3 dpa, voids were only observed by TEM for 623 K irradiation [12]. Positron annihilation spectroscopy was performed to detect small vacancy type defects which were not observed by TEM [13]. Micro-voids existed at 473 K as shown in Table 1. Though micro-voids were identified for irradiation at 573 K for low dose irradiation, no micro-voids were detected for a dose of 0.19 dpa. Decoration of dislocations by interstitial type dislocation loops was observed at 473 K, and is shown in



473K 0.061dpa 573K 0.091dpa 623K 0.27dpa 673K 0.24dpa

Fig. 4. Damage structures in Fe neutron irradiated in the JMTR. The figure inserted at 623 K is a void contrast image.

Table 1
Two component positron lifetime analysis in Fe neutron irradiated in the KUR and the JMTR

Temperature (K)	473			573			673		
Damage (dpa)	0.0046	0.019	0.098	0.021	0.10	0.19	0.066	0.13	0.42
Mean lifetime (ps)	294	325	291	117	123	109	114	113	109
Short lifetime (ps)	136	154	113	112	114	–	–	–	–
Long lifetime (ps)	396	403	380	342	285	–	–	–	–
Intensity (%)	60	69	61	6.4	14	–	–	–	–

‘–’ Denotes that two component decomposition was not possible, indicating the absence of voids.

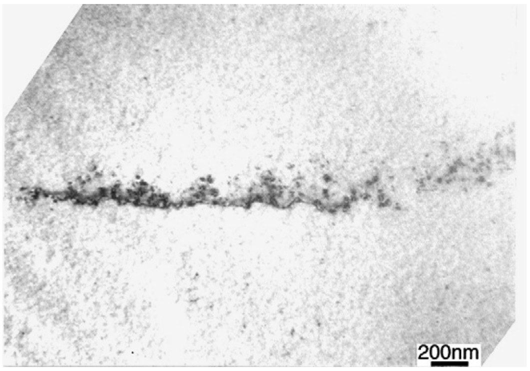
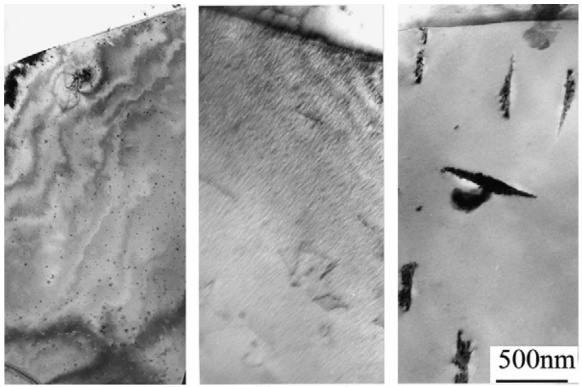


Fig. 5. Decoration of edge dislocation by interstitial type dislocation loops in Fe irradiated at 473 K to 0.061 dpa.

Fig. 5. Interstitial type dislocation loops near grain boundaries were observed for the irradiation at 573 K and 673 K but not at 623 K as shown in Fig. 6. If we apply the criteria of 1-D motion of interstitial clusters, the 1-D motion exists at 473 K and 623 K. In the case of Fe, as voids are observed at 473 K and 623 K, there exists a good correlation between 1-D motion of interstitial clusters and void formation.



573K 0.11dpa 623K 0.34dpa 673K 0.29dpa

Fig. 6. Damage structures in Fe neutron irradiated in the JMTR. Grain boundaries are at the upper part of the figures.

5. Discussion

The temperature dependence of 1-D motion of interstitial clusters was observed in Ni and Fe for our irradiation condition. In the case of Ni, at temperature lower than 573 K, the 1-D motion of clusters did not occur. In the case of Fe, the 1-D motion also did not occur at 573 K and 673 K. In previous papers, we showed that oversized element Sn and undersized element Si in Ni prevent the 1-D motion of interstitial clusters [7,10,11]. Sato et al. have studied the effect of foreign atoms on the migration of interstitial clusters as a bundle of crowdions [14]. They showed that oversized element Au (oversize by 63.6% [15]) in Ni increased the migration energy. Their result indicates that the actual long range migration of interstitial clusters is very effectively prevented by impurities in the migration path of interstitial clusters. Therefore the temperature dependence of 1-D motion of interstitial clusters is expected by that of the interaction between interstitial clusters and impurities.

In the case of Ni, we conclude that the 1-D motion of interstitial clusters is prevented by impurities at temperature lower than 573 K and that the 1-D motion of clusters overcome the barrier of impurities at temperature above 573 K. Residual loops near grain boundaries and surfaces are faulted loops with Burgers vector of $1/3\langle 111\rangle$. As mobile interstitial clusters are perfect loops with Burgers vector of $1/2\langle 110\rangle$, trapping of loops by impurities may induce Burgers vector change. Impurities in the interstitial planes reduce the stacking fault energy, which contributes to the formation of faulted loops.

The 1-D motion of interstitial clusters in Fe is not simple. Though the vacancy migration energy is low [16], except for highly pure Fe, vacancies actually move above 473 K aided by the influence of impurities [17]. If impurity pairs and clusters are formed by the migration of impurities with vacancies at 573 K, they act as strong obstacles and prevent the 1-D motion of interstitial clusters. At around 623 K interstitial clusters may overcome the barrier. At higher temperature, stronger obstacles, such as phonons or newly formed strong clusters, again prevent the motion. As Burgers vector of residual loops is $\langle 100\rangle$, impurities may contribute to Burgers vector change from mobile $1/2\langle 111\rangle$ type to immobile $\langle 100\rangle$ type.

6. Conclusion

The temperature dependence of 1-D motion of interstitial clusters has been identified by experimental results. It was shown that the temperature dependence was caused by the interaction between impurities and the clusters. Our understanding of 1-D motion of interstitial clusters has increased and identified an important mechanism for void growth and mechanical property changes. Most of studies were performed, however, in pure metals. If the 1-D motion of interstitial clusters is affected by impurities, the study of 1-D motion in real alloys is important to the development of fusion reactor materials.

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