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One dimensional motion of interstitial clusters and void growth in Ni and Ni alloys

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Abstract

One dimensional (1-D) motion of interstitial clusters is important for the microstructural evolution in metals. In this paper, the effect of 2 at.% alloying with elements Si (volume size factor to Ni: −5.81%), Cu (7.18%), Ge (14.76%) and Sn (74.08%) in Ni on 1-D motion of interstitial clusters and void growth was studied. In neutron irradiated pure Ni, Ni–Cu and Ni–Ge, well developed dislocation networks and voids in the matrix, and no defects near grain boundaries were observed at 573 K to a dose of 0.4 dpa by transmission electron microscopy. No voids were formed and only interstitial type dislocation loops were observed near grain boundaries in Ni–Si and Ni–Sn. The reaction kinetics analysis which included the point defect flow into planar sink revealed the existence of 1-D motion of interstitial clusters in Ni, Ni–Cu and Ni–Ge, and lack of such motion in Ni–Si and Ni–Sn. In Ni–Sn and Ni–Si, the alloying elements will trap interstitial clusters and thereby reduce the cluster mobility, which lead to the reduction in void growth.

1. Introduction

The importance of one dimensional (1-D) motion of interstitial clusters on microstructural evolution in metals has been pointed out by many researchers [1–6]. 1-D motion of interstitial clusters was first observed during the electron irradiation with high voltage electron microscopy [7,8]. Movement of interstitial clusters was also often observed in neutron irradiated metals by transmission electron microscopy (TEM) [9]. Formation of interstitial type dislocation loops near the dilatational side of edge dislocations [10,11] and alignment of interstitial type dislocation loops on ⟨1 1 0⟩ directions [12] are considered to be the result of movement of clusters. By molecular dynamics simulation of cascade damage, the escape of interstitial clusters was shown [13]. Analytical calculations have demonstrated that 1-D transport of interstitial clusters is likely to cause decoration of dislocations by interstitial clusters as formation of rafts of interstitial type dislocation loops [5].

Alloying elements are expected to affect the motion of interstitial clusters, and thus it is possible to investigate the role of 1-D motion of interstitial clusters on microstructural evolution. The authors have studied earlier the effect of alloying elements in Ni [14–17]. In neutron irradiated pure Ni, well developed dislocation network and voids were observed at 573 K at a dose of 0.026 dpa by TEM. Existence of microvoids was detected by positron lifetime measurement even at a low dose level of 0.001 dpa [17]. The addition of 2 at.% Cu (7.18%: volume size factor to Ni [18]) and Ge (14.76%) does not change microstructures so much. On the other hand, after the addition of Si (−5.81%) and Sn (74.08%), no voids were detected by TEM observation [16] or positron lifetime measurement [17].

In this paper, the effect of alloying elements on 1-D motion of interstitial clusters and void growth is reported. Special attention was paid on the difference of defect structures between in the matrix and near grain boundaries in these alloys, since interstitial clusters were expected to escape to grain boundaries by 1-D motion.

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2. Experimental

Specimens irradiated were 99.99 pure Ni (Johnson Matthey) and Ni based binary alloys, which contain Si, Cu, Ge, and Sn as solute atoms. The concentration of solute atoms was 2 at.%. The alloys were cold-rolled to 0.1 mm thickness and 3 mm diameter discs were punched out. These discs were annealed at 1170 K for 1 h in a vacuum of 10^{-4} Pa and the average grain size after annealing was 0.03 mm.

Neutron irradiation was performed to a dose level of 0.4 dpa (damage rate: 1.5×10^{-7} dpa/s) with the Japan materials testing reactor at Japan Atomic Energy Research Institute. After irradiation each specimen was electro-polished for electron microscopy. Another type of specimens which were thinned before the irradiation for TEM was also observed (thin foil irradiation).

3. Results

The defect microstructures in neutron irradiated Ni and Ni alloys to a dose level of 0.4 dpa at 573 K were studied. In the matrix, well developed dislocation microstructures and voids were observed in Ni, Ni–Cu and Ni–Ge. Near grain boundaries, dislocation network, interstitial type dislocation loops (within 600 nm) and voids (within 200 nm) were not observed. Fig. 1 shows an example of Ni with a diffraction condition for dislocation observation. With increasing dose, the growth of interstitial type dislocation loops and dislocation network was observed in the matrix, not near grain boundaries. The growth of voids was also observed as shown in Fig. 2, where the micrographs were taken with so called void contrast condition. These results were obtained under the improved temperature control irradiation, where the specimen temperature was kept at the irradiation temperature before the start of reactor to avoid the temperature history [19,20]. Under conventional irradiation, some loops near grain boundary were observed due to an easy nucleation of loops during irradiation of the temperature transient [15].

On the other hand, no voids and few interstitial type dislocation loops were visible in the matrix of Ni–Si and Ni–Sn. Interstitial type dislocation loops were observed preferentially near grain boundaries in Ni–Sn as shown in Fig. 3. The loops were on each of four {1 1 1} plains with almost the same ratio. Large loops were also observed near grain boundaries in Ni–Si. The easy escape of interstitial clusters to the surface in Ni and no escape of them in Ni–Si and Ni–Sn were also demonstrated by the thin foil irradiated specimens at 673 K (Fig. 4). No defects were observed in Ni, while large interstitial type dislocation loops were observed in Ni–Si and Ni–Sn. These results clearly indicate the difference of defect processes between two groups.

4. Reaction kinetics analysis

A reaction kinetics analysis which included the point defect flow into planar sink (surface or grain boundary) was performed without including 1-D motion of point defect clusters as used in a previous paper [15]. The

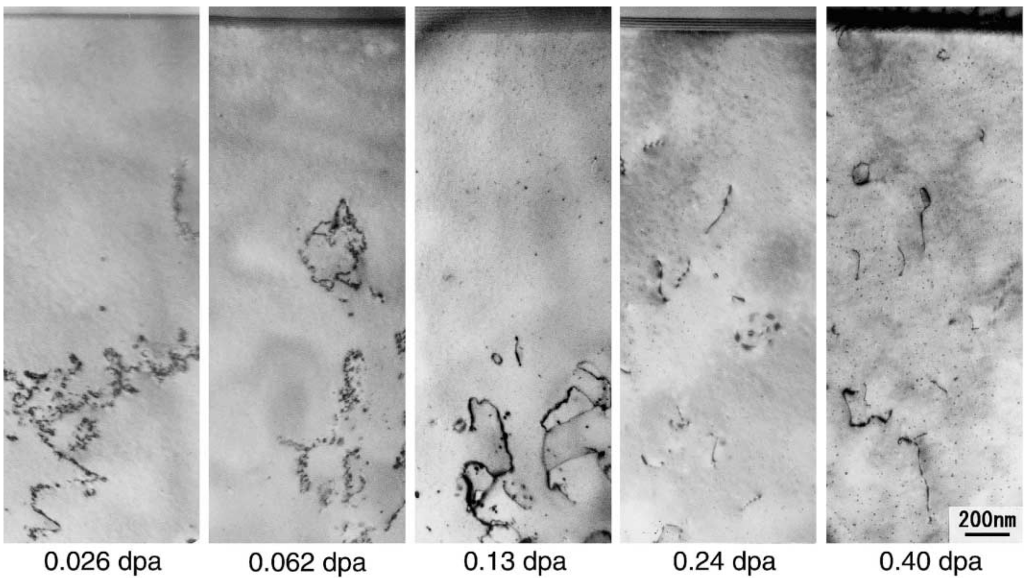

Fig. 1. Dislocation images in neutron irradiated Ni near grain boundaries at 573 K. Grain boundaries are at the top of each photograph.

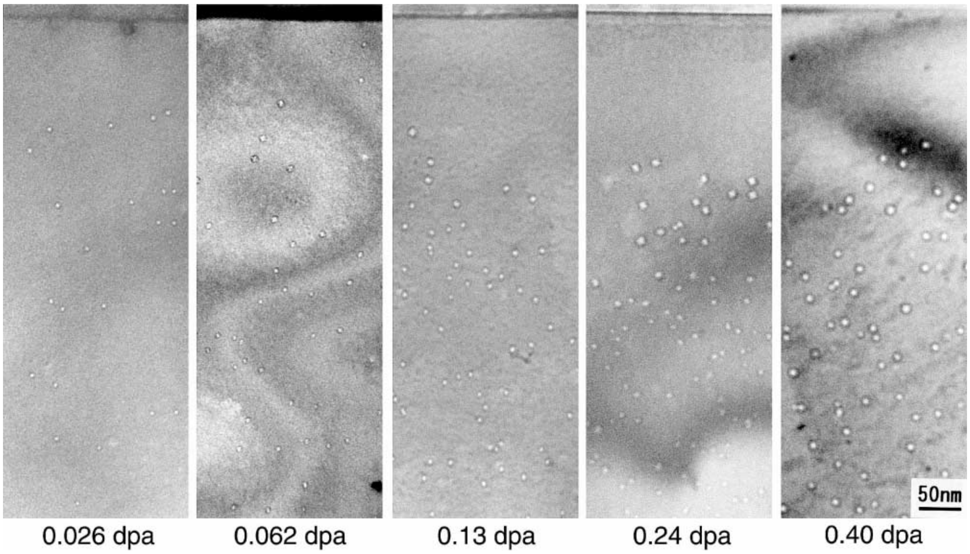

Fig. 2. Void images in neutron irradiated Ni near grain boundaries at 573 K. Grain boundaries are at the top of each photograph.

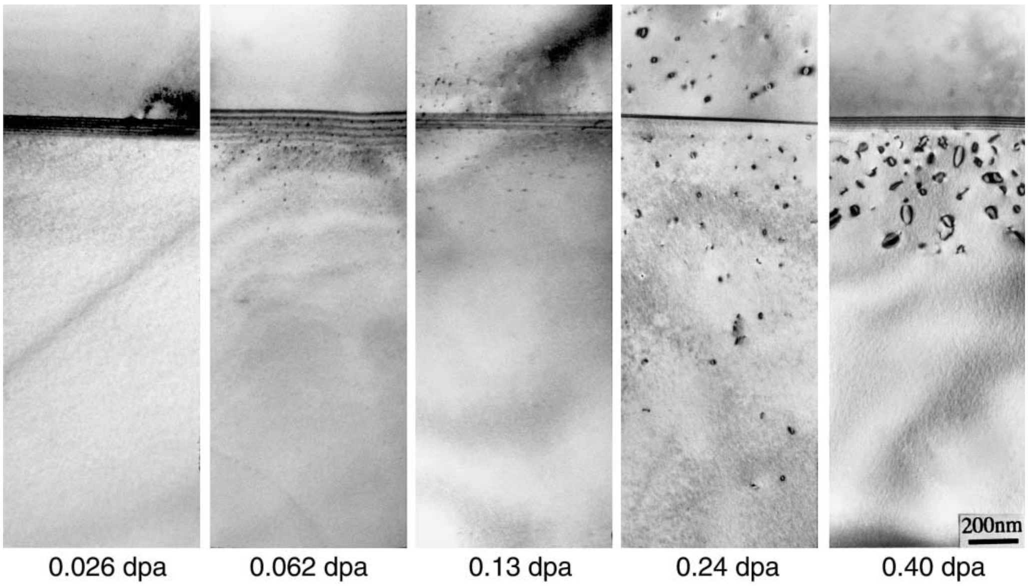

Fig. 3. Dislocation images in neutron irradiated Ni–Sn near grain boundaries at 573 K. Grain boundaries are at the upper part of each photograph.

variation of interstitial concentration C_{I} and vacancy concentration C_{V} at time t during irradiation is expressed as

$$\frac{\mathrm{d}C_{\mathrm{I}}}{\mathrm{d}t} = P_{\mathrm{I}} + \nabla^2 D_{\mathrm{I}} C_{\mathrm{I}} - Z(M_{\mathrm{I}} + M_{\mathrm{V}}) C_{\mathrm{I}} C_{\mathrm{V}} - Z_{\mathrm{I,IC}} S_{\mathrm{I}} M_{\mathrm{I}} C_{\mathrm{I}} - Z_{\mathrm{I,VC}} S_{\mathrm{V}} M_{\mathrm{I}} C_{\mathrm{I}} - T_{\mathrm{I}},$$

$$\frac{\mathrm{d}C_{\mathrm{V}}}{\mathrm{d}t} = P_{\mathrm{V}} + \nabla^2 D_{\mathrm{V}} C_{\mathrm{V}} - Z(M_{\mathrm{I}} + M_{\mathrm{V}}) C_{\mathrm{I}} C_{\mathrm{V}} - Z_{\mathrm{V,VC}} S_{\mathrm{V}} M_{\mathrm{V}} C_{\mathrm{V}} - Z_{\mathrm{V,IC}} S_{\mathrm{I}} M_{\mathrm{V}} C_{\mathrm{V}} - T_{\mathrm{V}},$$

where subscripts, I, V, IC and VC denote interstitials, vacancies, interstitial clusters and vacancy clusters, respectively. P is the point defect production rate by

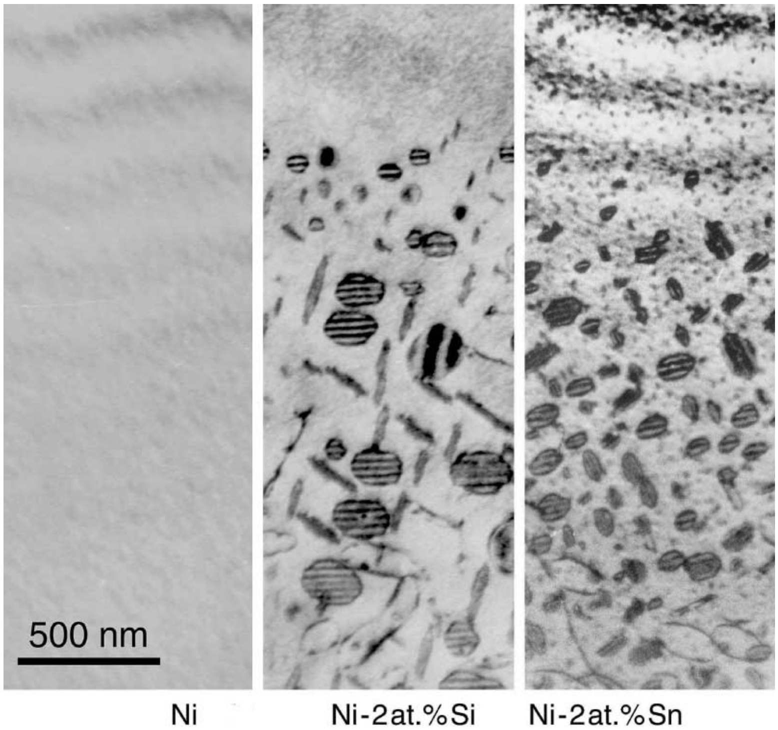

Fig. 4. Differences of defect microstructures in thin foils of Ni, Ni–Si and Ni–Sn neutron irradiated at 673 K to 0.31 dpa.

neutron irradiation. D is the diffusion coefficient, Z is the number of sites for the reaction with subscripts of reacting components, M is the mobility, S is the sink efficiency of point defect clusters formed during the irradiation and T is the thermal dissociation rate of point defects from clusters. The nucleation rate of interstitial clusters C_{IC} and vacancy clusters C_{VC} are

$$\frac{\mathrm{d}C_{\mathrm{IC}}}{\mathrm{d}t} = P_{\mathrm{IC}},$$
$$\frac{\mathrm{d}C_{\mathrm{VC}}}{\mathrm{d}t} = P_{\mathrm{VC}},$$

where P_{IC} and P_{VC} are the production rates of interstitial and vacancy clusters directly in cascades, respectively.

The accumulation rate of interstitials and vacancies in their clusters is

$$\frac{\mathrm{d}R_{\mathrm{I}}}{\mathrm{d}t} = Z_{\mathrm{I,IC}}S_{\mathrm{I}}M_{\mathrm{I}}C_{\mathrm{I}} - Z_{\mathrm{V,IC}}S_{\mathrm{I}}M_{\mathrm{V}}C_{\mathrm{V}} + P_{\mathrm{IC}}N_{\mathrm{I}},$$
$$\frac{\mathrm{d}R_{\mathrm{V}}}{\mathrm{d}t} = Z_{\mathrm{V,VC}}S_{\mathrm{V}}M_{\mathrm{V}}C_{\mathrm{V}} - Z_{\mathrm{I,VC}}S_{\mathrm{V}}M_{\mathrm{I}}C_{\mathrm{I}} + P_{\mathrm{VC}}N_{\mathrm{V}},$$

where R_{I} and R_{V} are total interstitials and vacancies in interstitial and vacancy clusters, respectively. N is the number of point defects in a cluster formed directly from the cascade damage.

As for the point defect clusters, we select two types; interstitial type dislocation loops and voids. The sink efficiencies of these defect clusters are

$$S_{\mathrm{I}} = 2\pi L_{\mathrm{I}}C_{\mathrm{IC}},$$
$$S_{\mathrm{V}} = 4\pi L_{\mathrm{V}}C_{\mathrm{VC}},$$
$$\frac{4\pi}{3}L_{\mathrm{V}}^3C_{\mathrm{VC}} = R_{\mathrm{V}},$$
$$\pi L_{\mathrm{I}}^2C_{\mathrm{IC}} = R_{\mathrm{I}},$$

where L is the radius of defect clusters in atomic distance unit.

The results are shown in Fig. 5 for two systems, where the values of parameters were the same as in the previous paper [15]. The point defect flow into the planar sink is taken into above equations as boundary conditions, i.e., the concentration of point defects and their clusters is 0 at the planar sink. In the system containing interstitial type dislocation loops and voids (System I, $P_{\mathrm{IC}} \neq 0$ and $P_{\mathrm{VC}} \neq 0$), voids and loops grew in the matrix and the growth of loops was pronounced near the planar sink. In this system, as a result of the growth of loops, excess vacancies accumulated in voids in the matrix. Near the planar sink, vacancies can escape there, and accordingly more loops remained. In the other system in which only interstitial type dislocation loops were assumed (System II, $P_{\mathrm{IC}} \neq 0$ and $P_{\mathrm{VC}} = 0$),

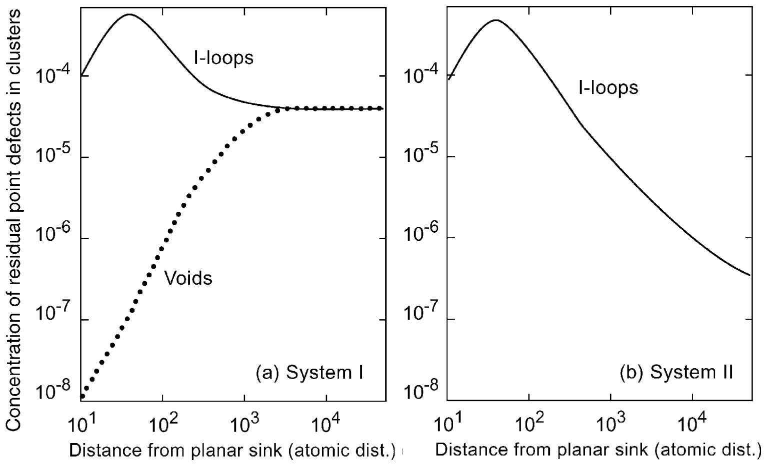

Fig. 5. The residual point defects in clusters after neutron irradiation to 0.1 dpa as a function of the distance from a planar sink in (a) system I (interstitial loops and void co-existing system) and in (b) system II (only interstitial loops containing system).

loops grew only in a narrow volume near the planar sink. In this system, vacancies produced in the matrix were absorbed by loops, reducing the number of interstitials in loops. The cases of Ni, Ni–Cu and Ni–Ge belong to System I: well developed interstitial type dislocation loops, dislocation network and voids co-existing in the matrix. Absence of loops near the planar sink was different from the model analysis. System II corresponds to the cases of Ni–Sn and Ni–Si, and TEM observation almost supports the calculated result.

5. Discussion

The discrepancy between experimental and model analysis in System I is explained by the escape of interstitial clusters by 1-D motion to grain boundaries, which was not included in the model analysis. If interstitial clusters jump more than one atomic distance and vacancies jump only one atomic distance, the reaction efficiency of interstitials with other defects is lower than that of vacancies, which leads a vacancy dominant atmosphere and void growth in the matrix.

The effect of alloying elements on void growth has been usually explained by the interaction with point defects. Oversize elements will interact with vacancies and delay the void growth. Undersize elements will interact with interstitials. As the mobility of interstitials decreases, many interstitial type dislocation loops are formed. They are expected to act as an effective annihilation site for vacancies. If alloying elements affect 1-D motion of interstitial clusters, another explanation for the effect of alloying elements is possible, i.e., alloying elements, such as Sn (largely over size) and Si (under size) in Ni, will trap interstitial clusters and prevent their motion. The low reaction efficiency of interstitials is no longer accomplished there and the lack of 1-D motion of interstitial clusters prevents void growth in these alloys.

6. Conclusion

The effect of alloying elements in Ni on void growth was explained by 1-D motion of interstitial clusters. Dislocation network and voids were observed in Ni, Ni–Cu and Ni–Ge and no interstitial type dislocation loops were observed near grain boundaries. In Ni–Si and Ni–Sn, no voids were observed and interstitial type dislocation loops were preferentially formed near grain boundaries. The reaction kinetics analysis showed the existence of 1-D motion of interstitial clusters in Ni, Ni–Cu and Ni–Ge, and no existence in Ni–Si and Ni–Sn. These results indicate the importance of 1-D motion of interstitial clusters for void growth.

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