The Effect of Planar Sinks on the Interstitial Loop Growth under High Temperature Neutron Irradiation

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Synopsis

The role of planar sinks such as surfaces and grain boundaries for the defect structure developments was studied in fission neutron irradiated Ni and Ni alloys of 2 at% Si, Cu, Ge and Sn to the dose of $4\times10^{23}n/m^2$ (>1MeV) at 573 K by comparison between thin foil irradiation and bulk irradiation. The number density of interstitial loops increases and then decreases with the increase of distance from planar sinks. Observed defect structure developments were interpreted in terms of the variation of point defect concentration with the change of sink efficiency. The necessity of the introduction of cascade localization induced bias effect is emphasized.

I. Introduction

The authors pointed out the importance of the role of freely migrating point defects in the defect structure development in high energy neutron irradiated metals, based on the analysis of fusion neutron irradiation experiment of fcc metals such as Au, Ag, Cu, Ni and their dilute alloys[1]. The role of free point defects varies widely depending on irradiation temperatures. At lower temperatures where vacancies and interstitials can form defect clusters directly in cascades, free point defects act to modify these clusters. At higher temperatures where the formation of defect clusters of both interstitials and vacancies is not easy, free point defects become to

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play major role. Point defects to develop dislocation structures and voids are freely migrating ones which escaped from cascade damage zones.

As the concentration of free point defects is influenced by sink geometry, the study of defect structures as a function of sink geometry is important. Specimen surfaces are well defined sinks for both vacancies and interstitials, and grain boundaries also attract our interest from the same reason. In this paper, the influence of the planar sinks for the interstitial loop growth during fast neutron irradiation of Ni and Ni alloys at a high temperature is reported by comparison between thin foil irradiation and bulk irradiation. In thin foil irradiation, specimens are irradiated as electron microscopically observable thin foils and in bulk irradiation they are irradiated as foils of 0.1mm in thickness and observed after thinning. The observed defect structures are interpreted from the variation of point defect concentration with the change of distance from sinks. In practice, the distance from sink is replaced by sink efficiency. Variation of effective sink efficiency of planar sinks by the development of defect structures in the matrix is also discussed.

II. Neutron Irradiation

Ni and four kinds of 2 % Ni alloys, Ni-Si (-5.81), Ni-Cu (7.18), Ni-Ge (14.76) and Ni-Sn (83.40), are irradiated at 573 K using JMTR with an improved temperature control to eliminate the irradiation at transient temperatures[2]. The values in the parentheses are volume size factors of solutes to Ni[3]. Fig. 1 and 2 show examples of thin foil irradiation and bulk irradiation, respectively. In thin foil irradiation, interstitial type dislocation loops are observed in all specimens. Stacking fault tetrahedra are observed only at thin part. In bulk irradiation, the development of dislocation structures together with voids is observed in pure Ni, Ni-Cu and Ni-Ge alloys. In Ni-Si and Ni-Sn alloys only dislocation loops are observed.

The grain boundary also acts as a sink as well as the surface for both types of point defects. The observation of the defect structures near grain boundaries gives us information similar to the cross sectional view of specimen, the variation of defect structures from the surface to the inner part. Fig. 3 shows the defect structures near grain boundary in five bulk specimens. Preferential formation of interstitial type dislocation loops is observed near

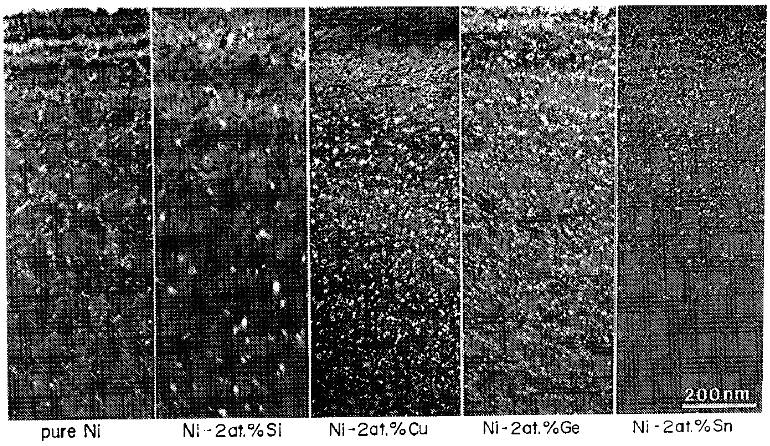

Fig. 1 Comparison of defect structures in thin foil irradiated Ni and Ni alloys at 573 K with JMTR. The fluence was $3.7 \times 10^{23} n/m^2$ (>1MeV).

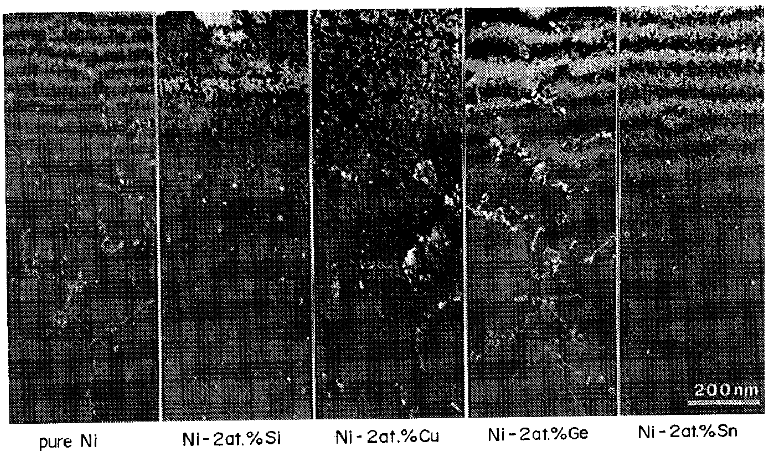

Fig. 2 Comparison of defect structures in bulk irradiated Ni and Ni alloys at 573 K with JMTR. The fluence was $3.7 \times 10^{23} n/m^2$ (>1MeV).

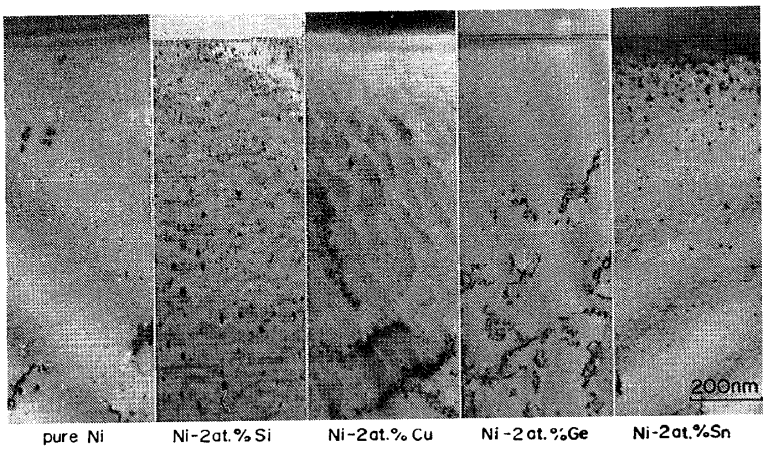

Fig. 3 Comparison of defect structures near grain boundary in bulk irradiated Ni and Ni alloys at 573 K with JMTR. The fluence was $3.7 \times 10^{23} n/m^2$ (>1MeV).

grain boundary only in Ni-Si and Ni-Sn alloys. Well developed dislocation structures with voids are observed in Ni, Ni-Cu and Ni-Ge at the area far from the grain boundary (background area). The defect structure near grain boundary in 14 MeV fusion neutron irradiated pure Ni with RTNS-II at 563 K is shown in Fig. 4 for comparison. Preferential formation of interstitial type dislocation loops near grain boundary is observed in each figure.

Increasing the irradiation dose, the number density of loops in the background increases and the predominance of the loops near grain boundary becomes ambiguous. In the JMTR irradiation, the energy deposition from neutrons in the present examples is 4 times higher than that from the fusion neutrons of highest dose. It can be concluded here that even in fission neutron irradiated pure Ni, dislocation loops are formed initially just the same as in the fusion neutron irradiated Ni and then they are annihilated.

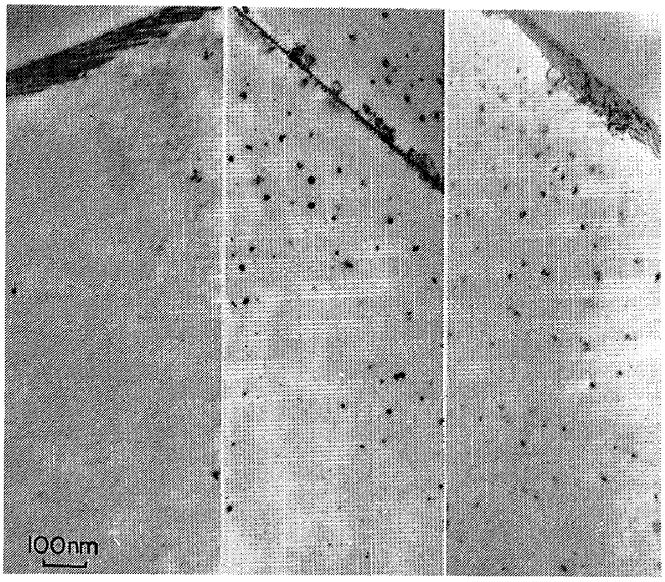

Fig. 4 Comparison of defect structures near grain boundary in bulk irradiated Ni with RTNS-II at 563K.

III. The Analysis of Planar Sink Efficiency as a Function of Distance from the Sink

The kinetics analysis, in which the distance to planar sink is replaced by sink efficiency, is performed based on rate equations in order to understand the observed defect structure variation. The nucleation of interstitial loops has been understood to take place directly in cascades[4], though their nucleation strongly depends on temperatures and alloying elements. Therefore making the problems simple, the nucleation process is neglected and it is assumed that the nuclei of interstitial loops exist already homogeneously in the matrix, and only the growth of them are treated. We also assume, as a high temperature case, the homogeneous formation of interstitials and vacancies in the matrix ignoring the effect of localized formation of point defects in cascades. The effect will be discussed at the last section of this paper.

The rate equations which express the variation of interstitial concentration C_I and vacancy concentration C_V during the neutron irradiation are given by

$$dC_I/dt = P - \pi(L/a)C_L Z_{IL} M_I C_I - S_I M_I C_I - Z_{IV} M_I C_I C_V,$$

and

$$dC_V/dt = P - \pi(L/a)C_L Z_{VL} M_V C_V - S_V M_V C_V - Z_{IV} M_I C_I C_V.$$

The symbols used are listed in table 1. Each term in equations

Table 1 Symbols and their values used in calculation

Symbol	Quantity	Value
P	Point defect production rate	10^{-8}
M_I	Interstitial mobility	0.13×10^{13}/s
M_V	Vacancy mobility	100/s
Z_{IV}	Number of site for recombination of interstitials and vacancies	100
Z_{IL}	Absorption cross section of loops for interstitials	44
Z_{VL}	Absorption cross section of loops for vacancies	40
S_V	Planar sink efficiency for vacancies	
S_I	Planar sink efficiency for interstitials	
C_L	Number density of interstitial loops	1×10^{-7}
a	Change of loop radius by absorption of one point defect at a site	0.25nm

expresses the production rate of Frenkel pairs, the absorption rate of point defects at interstitial loops, the absorption rate of point defects at the surface or a grain boundary and the mutual annihilation rate between interstitials and vacancies. From the random walk theory, the surface sink efficiency S is estimated to be about $(a/h)^2$ at the center of a foil of thickness $2h$ or the distance h from a grain boundary.

The growth speed of an interstitial loop of the diameter L is given by

$$dL/dt = 2aZ_{IL}M_I C_I - 2aZ_{VL}M_V C_V.$$

The steady state condition of production and annihilation of point defects is given by $dC_I/dt = dC_V/dt = 0$, and each concentration of point defects is expressed as

$C_V = (A + B) / C,$
and
$C_I = (A + B) / D,$
where
$A = - M_V(Z_{VL} + S_V)(Z_{IL} + S_I),$
$B = (M_V^2(Z_{VL} + S_V)^2(Z_{IL} + S_I)^2 + 4M_V(Z_{VL} + S_V)(S_I + Z_{IL})Z_{IV}P)^{1/2},$
$C = 2Z_{IV}(Z_{VL} + S_I),$
and
$D = 2Z_{IV}(Z_{IL} + S_V).$

Figure 5 shows the variation of C_I and C_V and dL/dt as a function of S (= S_I = S_V) using the values in table 1, which corresponds to a typical case of neutron irradiated Ni with JMTR at 573 K. Here 10 % of dislocation bias to interstitials ($Z_{IL} = 1.1Z_{VL}$) is assumed. The point defect production rate P is determined under the assumption of homogeneous formation of point defect in the matrix. The concentrations of C_I and C_V increase with foil thickness and saturate, while the growth speed of loops dL/dt has a maximum and starts to decrease. The result of this variation of loop growth as a function of distance from sinks is explained as follows.

(1) Very thin foil (large S): The planar sink is the dominant sink and the concentrations of interstitials and vacancies are low by

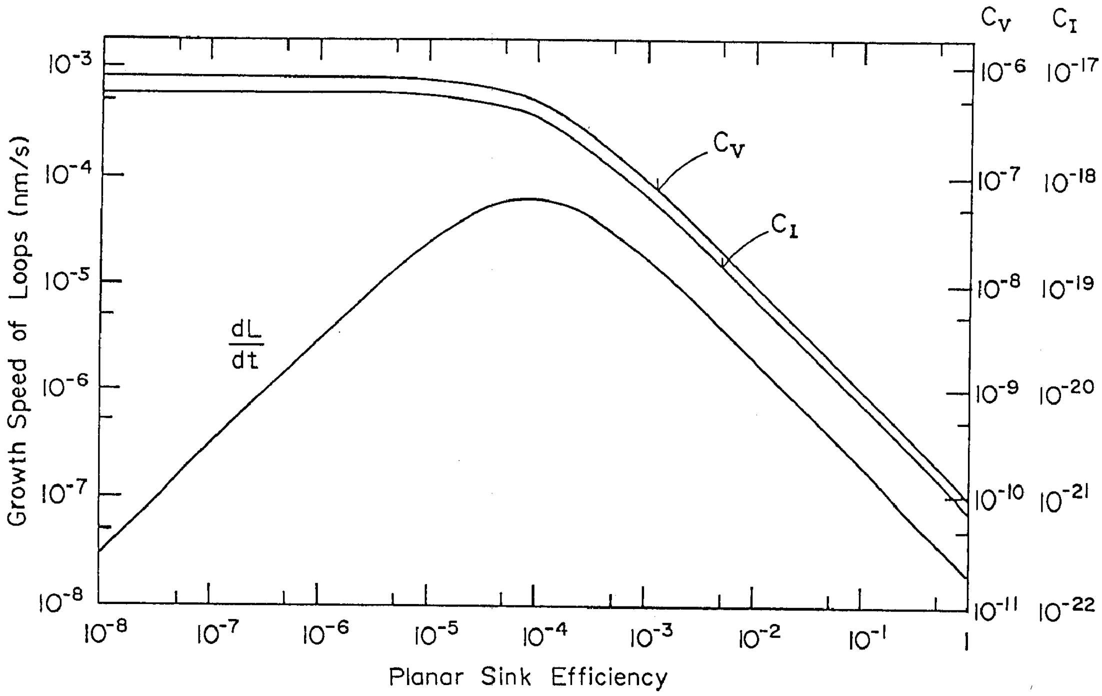

Fig. 5 Results of simulation of the concentrations of vacancies C_V and interstitials C_I, and the growth speed of interstitial loops as a function of the planar sink efficiency.

their escape to planar sinks, and the growth rate is very small.
(2) Thick foil (small S): The concentrations of both interstitials and vacancies increase by their difficulty to escape to planar sinks and by their low mutual annihilation rate, even with their increase of concentration. The interstitial loops can grow by the difference caused by the bias to point defects.
(3) Very thick foil (bulk condition, $S \simeq 0$): The planar sink does not act as a sink any more. The concentrations are determined with a balance between point defect production rate and mutual annihilation rate between interstitials and vacancies and become constant. Even though the concentrations of interstitials and vacancies are high, in this case, dislocation loops absorb the same number of interstitials and vacancies, and can not grow larger. In other words, an additional increase of vacancy concentration caused by the preferential absorption of interstitials to dislocations compensates the predominance of interstitial absorption to the dislocation loops, which is realized in the case (2).

The observed dislocation loop growth as a function of specimen thickness, i.e., predominant loop growth in a foil irradiated Ni and Ni alloys is explained in this model calculation as the case of (2). High density of loops near grain boundary and the decrease of loop density at the inner part of bulk irradiated Ni-Si and Ni-Sn alloys are also understood as the transition from the case (2) to the case (3).

In Ni, Ni-Cu and Ni-Ge alloys, no loops near grain boundary and well developed dislocation structures with voids are observed. The kinetics analysis of planar sink efficiency to point defects mentioned above does not explain these structure changes, even when the parameters used are changed widely. No loop formation near grain boundary is strongly related to the development of dislocation structures in the background. In the next section, the development of dislocation structures with void growth is discussed first by cascade localization induced bias effect and then no loop formation near grain boundary in Ni, Ni-Cu and Ni-Ge alloys is discussed.

IV. Cascade Localization Induced Bias Effect

The development of dislocation structures with void growth has been generally interpreted in terms of the bias effects of dislocations to point defects. While the authors pointed out the importance of the effect of localized formation of point defects by cascade damage[5] and have proposed the cascade localization induced bias effect (CLIB effect) for accounting the appearance of a vacancy dominant atmosphere during neutron irradiation[6]. In a cascade, the initial local distributions are different between interstitials and vacancies, i.e., a vacancy rich region which occupies a smaller volume is surrounded by a larger volume of interstitial rich region. When the diffusion of vacancies starts from this localized volume, the vacancy rich atmosphere is formed until their annihilation. The growth of voids and dislocation structures for various initial vacancy-interstitial distributions was calculated and the possibility of the void swelling of 1 %/dpa in austenitic stainless steels was demonstrated with this mechanism. This is entirely different sequence of defect structure development from those by dislocation bias in which voids are formed after dislocation structure development[7].

According to CLIB effect, the appearance of vacancy rich atmosphere during neutron irradiation make it possible to form the voids and dislocation structures simultaneously. No loop formation near grain boundary in pure Ni, Ni-Cu and Ni-Ge alloys is explained as follows. The growth of interstitial loops near grain boundary takes place in an early stage of irradiation where the dislocation structures have not developed yet even in Ni, Ni-Gu and Ni-Ge alloys as shown in the case (2) of the calculation and also in the examples of low dose irradiated Ni (see Fig. 4). With increasing the irradiation dose, dislocation structures with voids develop in the background. The dislocations act as strong sinks for interstitials, which leads to the flow of excess vacancies to the grain boundary and the increase of vacancy concentration near grain boundary. Consequently pre-existing loops near grain boundary are annihilated after the growth of dislocation structures.

As mentioned above, loops near grain boundary are annihilated with the development of dislocation structures. In other words, high density of loops observed near grain boundary in Ni-Si and Ni-Sn alloys results from no development of dislocation structures in the background. The reason why the dislocation structures with voids do not develop in Ni-Si and Ni-Sn alloys is explained based on the alloying effect. Alloying elements Si and Sn are considered to suppress the mobility of interstitials and increase the interstitial concentration[8], which leads to the easy nucleation of interstitial loops. High density nuclei of interstitial loops will act as sinks for both interstitials and vacancies. CLIB effect does not work effectively in this case, since there are too many sinks in cascades and long range diffusion of point defects, which is required condition for CLIB effect to be effective, is prevented. Accordingly the development of defect structures in bulk does not take place and preferential growth of loops near grain boundary remains.

Acknowledgments

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