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Damage structures in fission-neutron irradiated Ni-based alloys at high temperatures

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Abstract

The defects formed in Ni based (Ni–Si, Ni–Cu and Ni–Fe) alloys which were irradiated with fission-neutrons were examined by electron microscopy. Irradiations were carried out at 473 K and 573 K. In the 473 K irradiated specimens, a high density of large interstitial loops and small vacancy clusters with stacking fault tetrahedra (SFT) were observed. The number densities of these two types of defects did not strongly depend on the amount of solute atoms in each alloy. The density of the loops in Ni–Si alloys was much higher than those in Ni–Cu and Ni–Fe alloys, while the density of SFT only slightly depended on the kind of solute. Also, the size of the loops depended on the kinds and amounts of solute. In 573 K irradiated Ni-Cu specimens, a high density of dislocation lines developed during the growth of interstitial loops. In Ni–Si alloys, the number density and size of the interstitial loops changed as a function of the amount of solute. Voids were formed in Ni–Cu alloys but scarcely formed in Ni–Si alloys. The number density of voids was one hundredth of that of SFT observed in 473 K irradiated Ni–Cu alloys. Possible formation processes of interstitial loops, SFT dislocation lines and voids are discussed.

1. Introduction

Materials in fusion reactors will be irradiated with high energy neutrons at high temperatures. Investigations of damage evolution in fission-neutron irradiated metals at high temperatures have received much interest as a means to simulate the damage produced by fusion-neutron irradiation.

Damage structures have been investigated in metals which were neutron-irradiated at reactor ambient temperatures or higher temperatures, and the formation of various types of defects has been reported previously [1–3]. General trends for the formation of damage microstructures in the metals have been observed. The number density of defect clusters formed by irradiation generally increased with neutron fluence and decreased with irradiation temperature. The size of the defects generally increased with irradiation temperature. However, the observed damage structures varied greatly depending on the specific investigation. For example, the defect morphology differed from one investigation to another even when pure metals were studied. Number densities and sizes of the defects varied among the several investigations. Few systematic studies of the damage were carried out, varying the amount and type of solute in alloys. Systematic studies are needed to clarify the parameters which may affect the damage structures. To determine the mechanisms of damage evolution in metals, an understanding of the nature of the defect structure is essential.

In the present work, to clarify the factors affecting the damage structures and to infer the mechanism of the damage evolution, nickel based alloys with systematic variation of the concentration of solute and thus size of solute atoms was irradiated with fission reactor-neutrons and the damage structures were studied by electron microscopy.

2. Experimental procedure

The Ni based alloys, Ni–Si, Ni–Cu and Ni–Fe, used in this study were fabricated by melting in an argon electric arc furnace. The raw materials were Ni, Si, Cu

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and Fe in the nominal purity of 99.99%, 99.9999%, 99.999% and 99.99%, respectively. The Ni, Cu and Fe were supplied by Johnson Matthey Chemicals and the Si by Osaka Titanium. The concentrations of each solute element in the alloys were 0.1, 1 and 5 at.%, respectively.

The specimens were foils of $50–100 \times 10^{-6}$ m in thickness and were annealed in high vacuum (10^{-4} Pa) between 1073 K and 1173 K. Two irradiations were done in the Japan Material Test Reactor (JMTR). In one of irradiation, the bulk specimens were exposed to fluence up to 2.5×10^{23} n/m^2 ($E > 1$ MeV) at 473 K. In another irradiation, the bulk specimens were exposed to fluences 3.5 and 5×10^{23} n/m^2($E > 1$ MeV) at 573 K.

After irradiations the bulk specimens were electropolished and observed at room temperature using a JEOL-200CX transmission electron microscope.

3. Experimental results and discussion

3.1. 473 K irradiation

In Fig. 1, the damage structure of pure Ni is shown in dark and bright field for the same area. Comparing Fig. 1(a) and (b), two types of images are clearly observed: large loops and small dot defects. In the bright field the complex images with strong strain contrast A, B and C are large loops and the faint small dot images are small dot defects. Many small dot images, which cannot be observed due to faint contrast of the images in the bright field are observed in the dark field. Characteristics of the images are similar to the alloys though the size of the defects changes from alloy to alloy. Hereafter the damage structures for the alloys are only shown with weak beam images in dark fields because the size of defects are inherently shown with weak beam images. In Fig. 2, the damage structures of Ni–Cu, Ni–Si and Ni–Fe alloys are shown for various alloying concentrations with weak beam images in dark field. As mentioned before, there are two types of images especially clear in the alloys of low solute concentration; large dislocation loops and small dot defects scattered among the large dislocation loops. As mentioned later, the large loops are interstitial type in nature and many dot defects are vacancy type defects (stacking fault tetrahedra). Small loops were also observed and distinguished from SFT by their strain contrast, using both the dark field and bright field images at the same area as shown later (in Fig. 5).

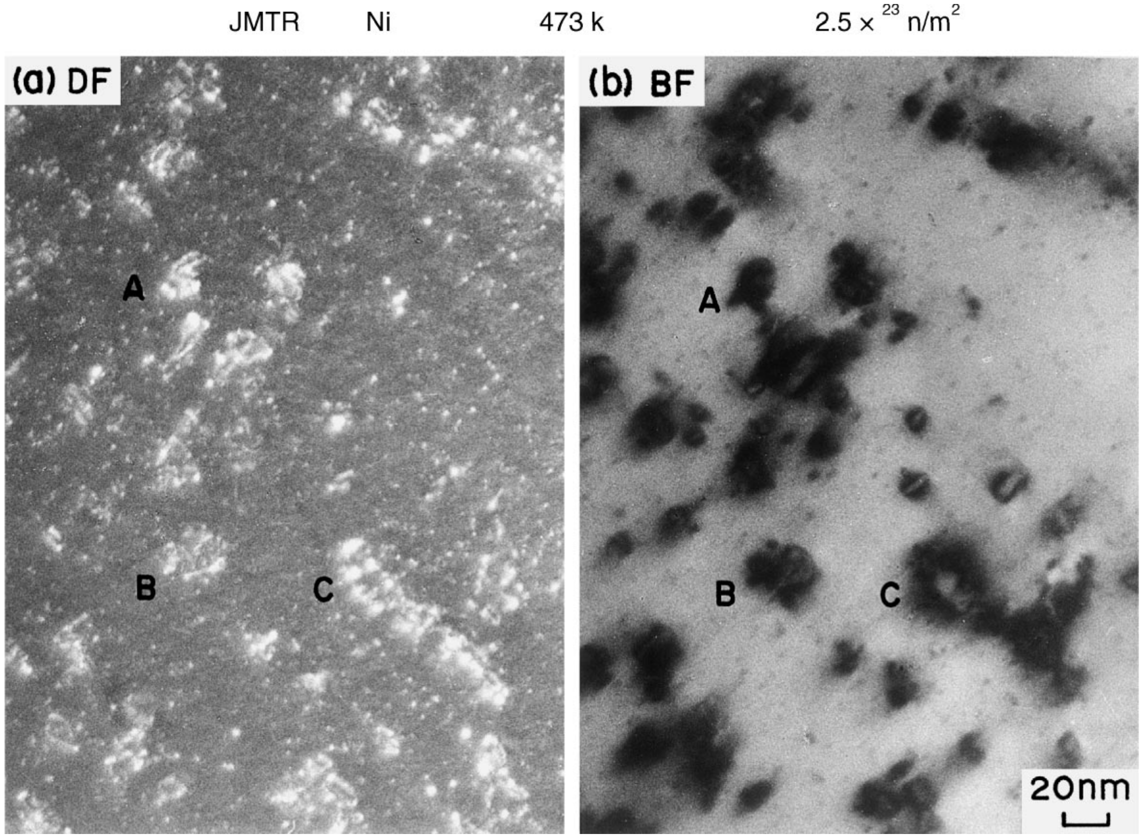

Fig. 1. Images in dark and bright field for the same area in pure Ni irradiated at 473 K up to 2.5×10^{23} n/m^2.

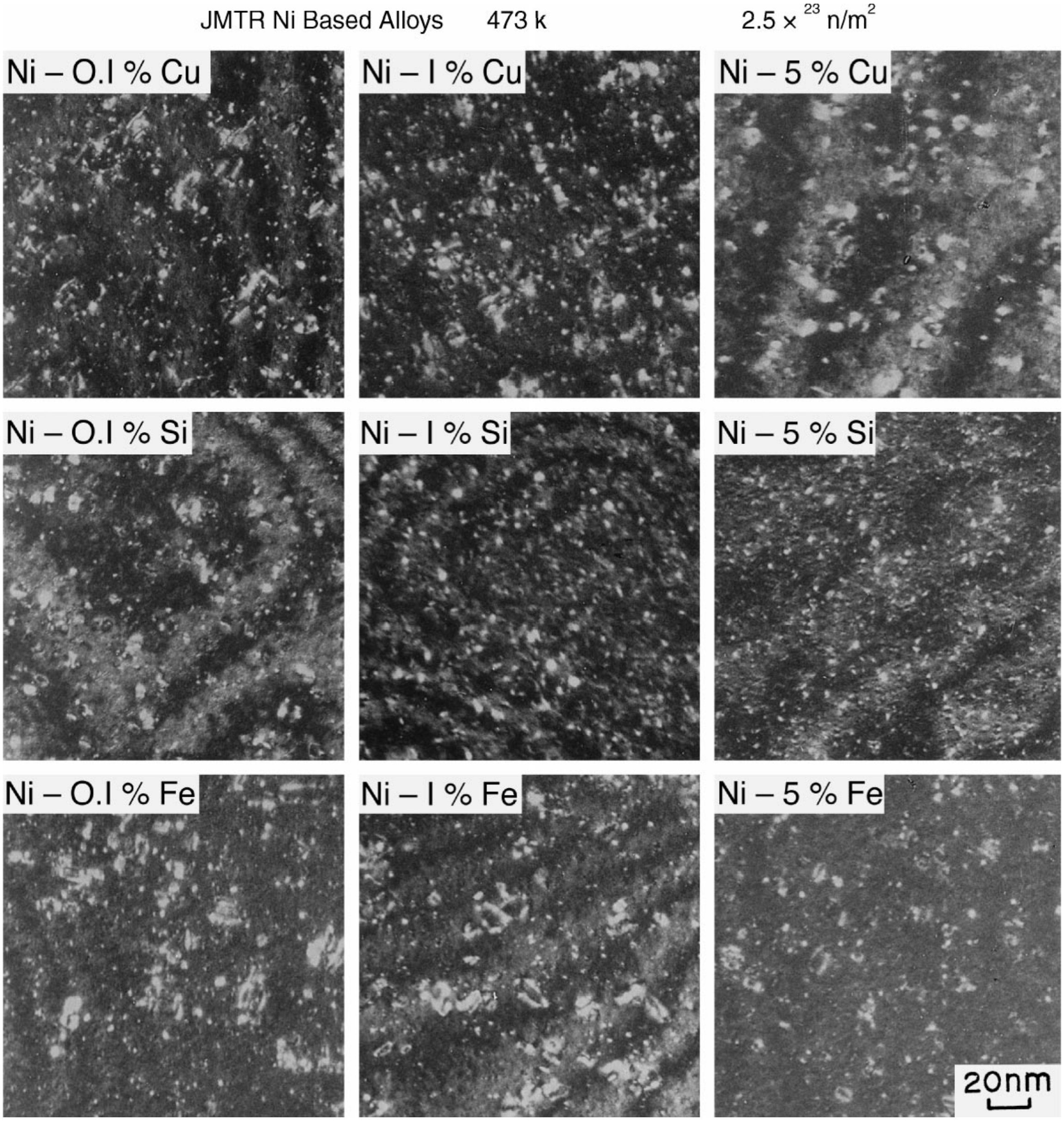

Fig. 2. Damage structures in Ni–Cu, Ni–Si and Ni–Fe alloys irradiated at 473 K up to 2.5×10^{23} n/m^2. The loops in Ni–Si alloys were significantly smaller than the loops in Ni–Cu and Ni–Fe alloys.

Some of the large dislocation loops showed the fringe of stacking faults. The large loops were often decorated by some small loops and had complex shapes. The interstitial loops in Ni–Si alloys were considerably smaller in size and higher in number densities than those in Ni–Cu and Ni–Fe alloys. The mean sizes of the loops in Ni–Cu alloys were slightly larger than those in Ni–Fe alloys. The volume size factors of Fe, Cu and Si atoms to Ni atoms are 10.57%, 7.18% and −5.81%, respectively [4]. The volume size factor, V_{sf} is defined as

$$V_{sf} = (V_A - V_{Ni})/V_{Ni}, \tag{1}$$

where V_A and V_{Ni} are atomic volume of solute, A and solvent, Ni. The relative size of the loops did not follow in order of the volume size factors. In Cu–Mn and Cu–Ni alloys, the size of interstitial loops also did not follow the order of the size factors [5]. In all alloys, the dislocation loops became smaller with increasing solute content. On the other hand, the size of small dot defects did not clearly depend on the contents. Detection of

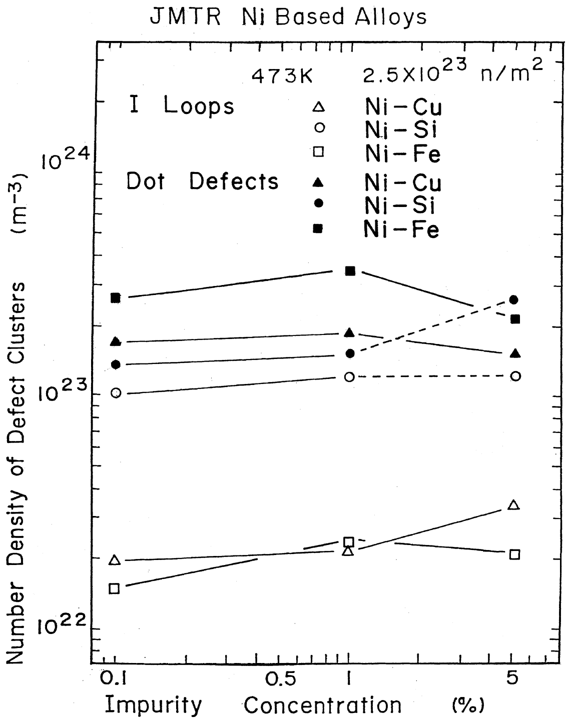

Fig. 3. Number densities of defect clusters for Ni–Si, Ni–Cu and Ni–Fe alloys against the amount of solute atoms. I loops: Interstitial loops.

small changes in size is difficult for small defects, especially at diameters in the 2 nm range.

Number densities of the defect clusters are shown in Fig. 3 for various Ni based alloys as a function of solute concentration. Both the loops and dot defects showed nearly constant densities in the range of 0.1–5 at.% solute atoms. We consider from the results of these alloys that the loops in the Ni–1 at.% Si and 5 at.% Si alloy are also interstitial type though the nature cannot be determined by the inside–outside contrast method due to the small size. In the Ni-5 at.% Si alloy, the images with strong contrast were regarded as loops and the images with weak contrast as dot defects. But for the images of medium contrast, the distinction between the two types was difficult. This ambiguity may lead to a higher number density of small dot defects in the 5 at.% Si alloy. The number densities of the dot defects for Ni–Cu and Ni–Fe alloys were one order of magnitude larger than those of interstitial loops.

The nature of large dislocation loops in Ni based alloys was determined by the inside–outside contrast method. In Fig. 4, the large dislocation loops are shown with opposite reflection vector $\boldsymbol{g}$ with the same sign of deviation parameter s ($s < 0$) for a dark field image. In Fig. 4(a), the images of the loops A and B are smaller than the images in Fig. 4(b), in which the reflection vector $\boldsymbol{g}$ is reversed as indicated. On the other hand, the images of C and E loop are larger than corresponding loop. The planes of the loops were determined by the change of the shape of the loops when the specimens were tilted. The nature of these large loops was interstitial type.

Small dot defects were observed at under and over focused conditions but these defects did not show void contrast. This is not caused by the smallness of the voids and therefore it can be concluded that there are no voids in the specimens at this low irradiation temperature. The shapes of some dot defects are shown in Fig. 5 in a bright field image. At marks, A, B, C and D, the small square images are observed. Geometrical directions are also shown in the figure. The edges of the squares are ⟨1 0 0⟩ when the defects were observed from [1 0 0]. From ⟨1 1 0⟩ or ⟨1 1 2⟩ directions, small triangular images were observed. Therefore, many of the small dot defects are SFT (vacancy clusters). The images at the marks, E and F are from small loops. The results observed at 473 K irradiation are summarized in Table 1 together with those at 573 K irradiation. I-type and SFT are interstitial type loops and stacking faulted tetrahedra, respectively. The marks L, M and S mean comparative size of defects for each alloy system.

In Ni–Si alloys, self interstitial atoms (SIA) have a larger binding energy to Si atoms compared to Cu and Fe atoms [6,7]. Interstitial loops would nucleate from aggregation of SIA–Si complexes. Therefore, the density of the loops formed in Ni–Si alloys was higher than those in Ni–Cu and Ni–Fe alloys. This high density of the loops explains the small size of the loops.

3.2. 573 K irradiation

At 573 K irradiation, a large number of dislocation lines, small loops marked with A and small dot defects are observed for Ni–Cu alloy specimens, as shown in Fig. 6. The damage structures in Ni–Fe alloys irradiated at 573 K have not been investigated by us. The stacking fault tetrahedra as in the specimens at 473 K irradiation are not observed in Ni–Si and Ni–Cu alloys. The features of micro-structures are similar to that of pure Ni, as shown in the figure, but in detail both the number density and size of voids increase slightly with the amount of solute atoms. The thinner part of the specimen is shown in Fig. 7 at under-focused condition (left) and focused condition (right) for the same area of the specimen. White dot images in the left figure are voids which show black dot images at an over-focused condition. A large part of small dot defects showed void images. The small dot images with strong strain contrast in the right figure are small dislocation loops found by comparing with the images in the left figure. The nature of these loops cannot be determined for the small size of the loops. The number densities of the voids in Ni–Cu

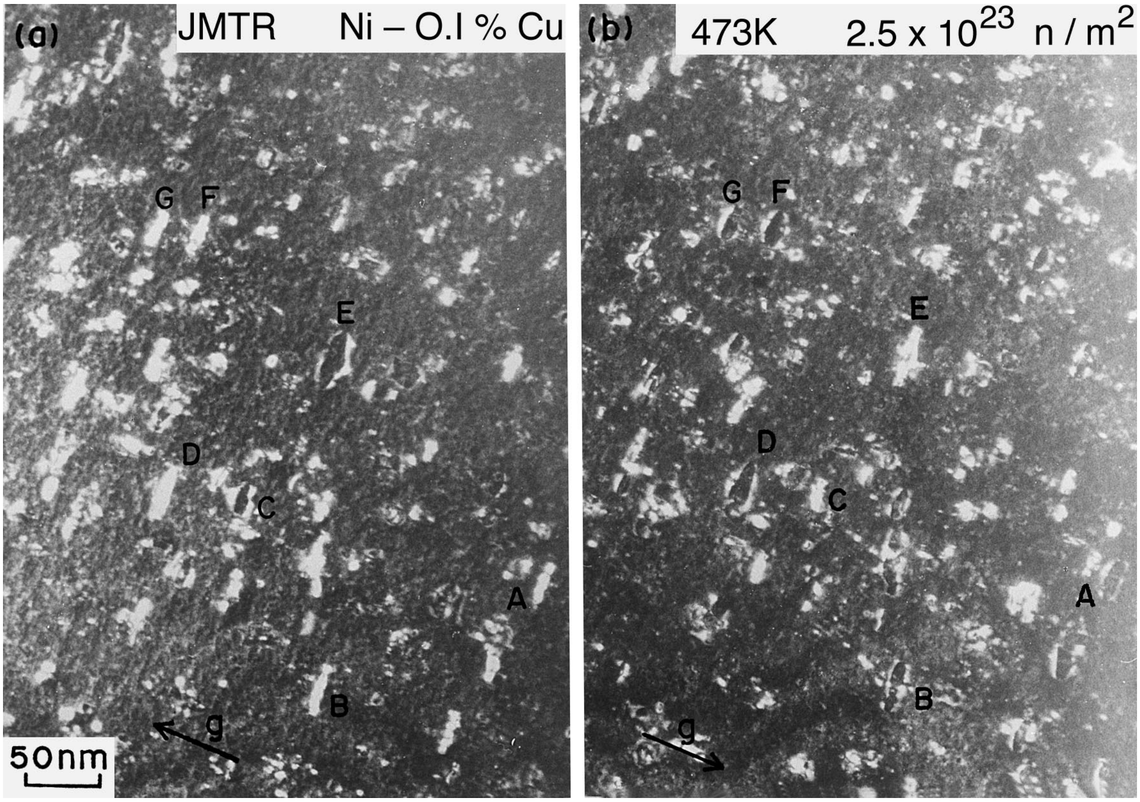

Fig. 4. Nature determination of the large loops in Ni–0.1at. % Cu alloy. The loops, A–G clearly change the size with reversing the reflection vector $\boldsymbol{g}$ with $s < 0$ as shown in (a) and (b).

alloys were two orders of magnitude lower than those of vacancy-type defects in the 473 K irradiation. In Ni-2 at.% Cu alloy, Kojima et al. have also observed a high density of voids in the specimen irradiated with 14 MeV neutrons at 563 K [3].

As shown in Fig. 8, for Ni–Si alloys large loops were observed with stacking faults and these became larger with increasing solute concentration. The size of the loops was larger than that for the 473 K irradiation. The nature of the loops was determined as interstitial type by the inside–outside contrast method [8]. Dislocation lines were not observed in the Ni–Si alloys different from the Ni–Cu alloys. The void density in Ni-0.1 at.% Si alloy was one order of magnitude lower than in the Ni–Cu alloys. Few voids were observed in the Ni-1 at.% Si alloy and not observed in the 5 at.% Si alloy. The results at 573 K irradiation are summarized in Table 1 together with those at 473 K irradiation. In another, voids were investigated by electron microscopy and positron annihilation for Ni and Ni-0.8 at.% Si alloy after electron irradiation [7]. Void formation was suppressed in the Ni-0.8 at.% Si alloy. This observation was explained by considering the slow migration of SIAs when strongly bound to Si solute atoms. The number of surviving interstitial atoms, which is estimated from the product of the density and the areal size of interstitial loops, did not considerably differ in the Ni–Si alloys. The number of

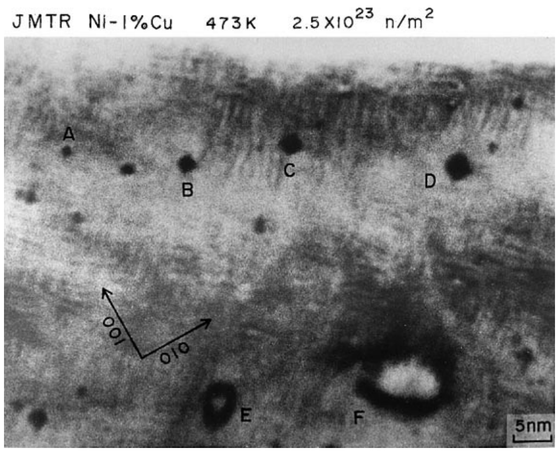

Fig. 5. Dot defects observed from [1 0 0] with high magnification at bright field in Ni-1 at.% Cu specimen irradiated to 2.5×10^{23} n/m^2 at 473 K.

Table 1
Observed defect images for Ni based alloys. $T_{irr.}$ and Cs in the table are irradiation temperature and solute concentration in atomic percent. The marks L,M and S mean comparative size of defects for each alloy system

	T_{irr}:	Observed defects							
		473 K Irradiation			573 K Irradiation				
		Loops	SFT	Voids	Loops		SFT	Voids	Dislocations
Alloys	Cs (%)	I-type			I-type	V or I-type			
Ni–Fe	0.1	○L	○	–					
	1	○M	○	–					
	5	○S	○	–					
Ni–Cu	0.1	○L	○	–		○	–	○	○
	1	○M	○	–		○	–	○	○
	5	○S	○	–		○	–	○	○
Ni–Si	0.1	○L	○	–	○S		–	○	–
	1	○M	○	–	○M	–	–	△	–
	5	○S	○	–	○L	–	–	–	–

(○) Observed; (△) Observed a few; (–) Not observed.

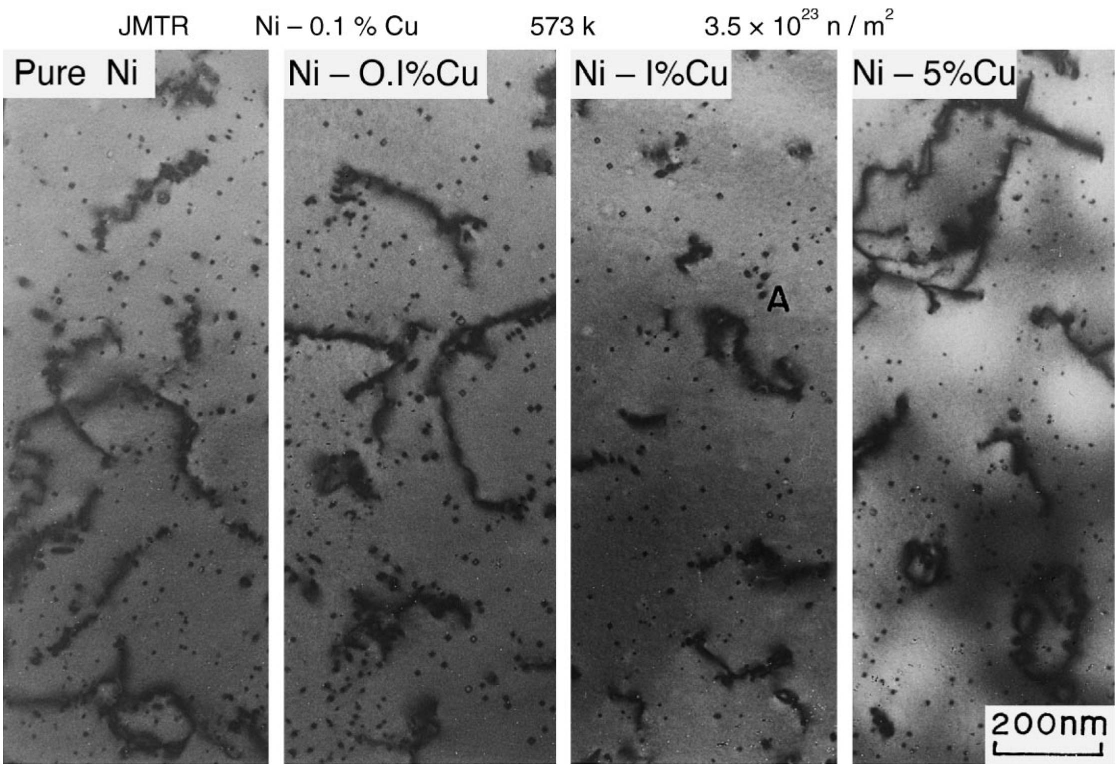

Fig. 6. Damage structures in pure Ni and Ni–Cu alloys irradiated at 573 K up to 5×10^{23} n/m^2.

surviving vacancies may be nearly the same to the number of above surviving interstitials though the electron microscopic images of vacancy clusters are not observed in the Ni-1 at.% and 5 at.% Si alloy. The vacancy clusters in the alloys are then micro-voids.

Nucleation of interstitial loops in the Ni–Si alloys decreases at higher irradiation temperature. Similarly the nucleation of the loops in the Ni–Cu alloys also decreases with temperature. Thus the loops in the Ni–Cu alloys can grow and intersect to form dislocation lines.

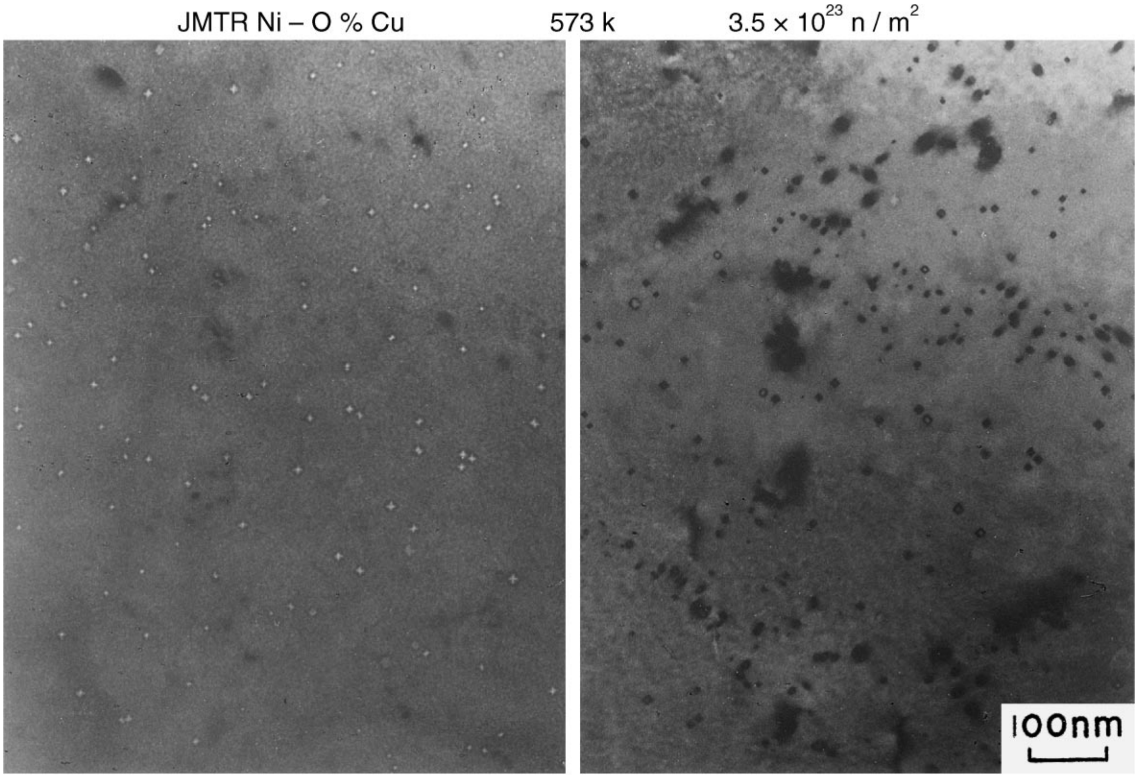

Fig. 7. The images at under-focused condition (left) and focused condition (right) at the same area in the specimen for bright field conditions. White dot images in the left figure are voids. Many small dislocation loops with strong strain contrast are observed in the right figure.

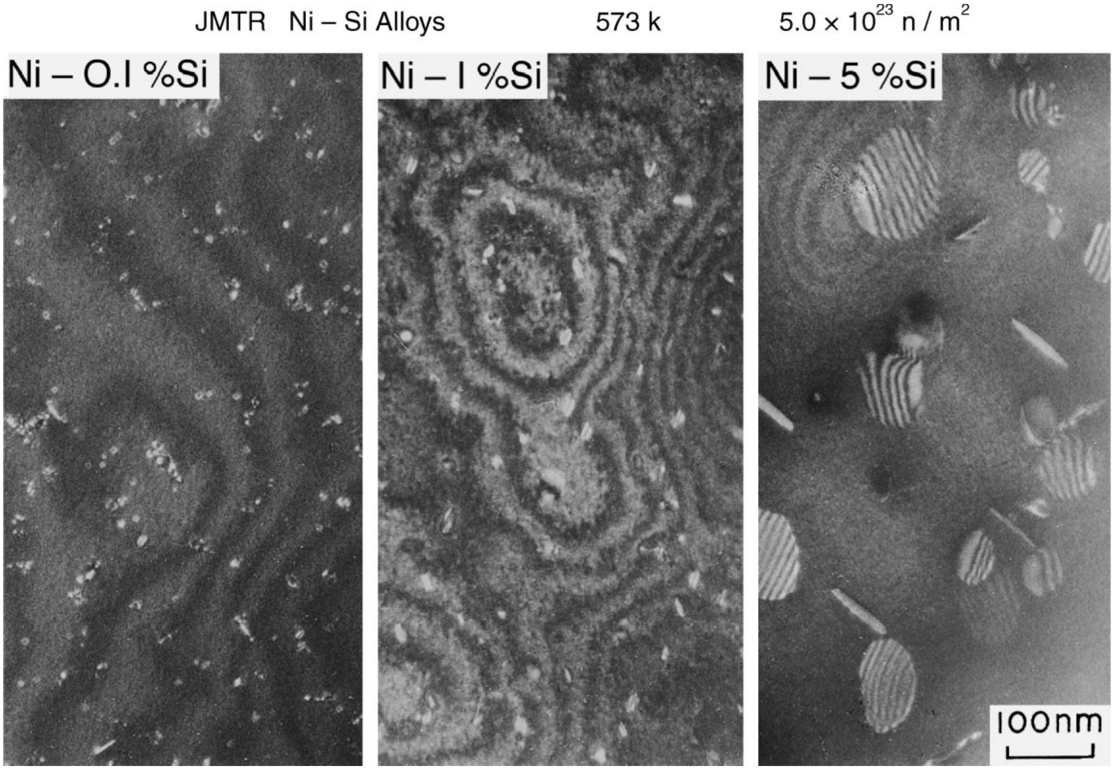

Fig. 8. Damage structures in Ni–Si alloys irradiated at 573 K up to 5×10^{23} n/m^2.

This study suggests that damage structures in Ni based alloys evolve at high temperatures in the following manner. Interstitial atoms produced by neutron irradiation aggregate and grow to large loops by absorbing interstitials subsequently produced. During growth of the loops, the large loops in Ni–Cu alloys intersect either with the foil surfaces or other loops, and the density of dislocation line segments increases. By the consumption of the interstitials at sinks (interstitial loops, dislocations or grain boundaries), vacancy concentration increases and becomes high enough to form vacancy type defects (voids, stacking faulted tetrahedra or vacancy loops). Which defects are formed depends on the stability of the defects at the irradiation temperature.

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