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Influence of temperature change on microstructure evolution in Ni alloys irradiated with neutrons

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Abstract

To investigate the microstructure evolution during temperature change irradiation, Ni and its binary alloys were irradiated in the Japan Materials Testing Reactor to doses up to 0.46 dpa under two cycles of temperature change of 533/693 K, i.e., 533 K, then 693 K, then 533 K, then 693 K. The irradiation dose at each temperature stage was about the same. The specimen sets were pulled out five times during irradiation. In the beginning of the first irradiation at the high temperature of 693 K after irradiation at 533 K, the densities of interstitial type dislocation loops and voids decreased in pure Ni, Ni–2at.%Cu and Ni–2at.%Ge alloys. In the second irradiation at 533 K, however, the formation of dislocation loops and voids was promoted. The void density continued to increase even after the second irradiation at 693 K. On the other hand, in Ni–2at.%Si and Ni–2at.%Sn alloys, where the formation of voids was suppressed by addition of the minor elements Si or Sn, the microstructure evolution was almost independent of irradiation temperature.

1. Introduction

It is expected that irradiation temperature will change during operation in near future fusion devices such as the International Thermonuclear Experimental Reactor (ITER). The effects of temperature change during irradiation on microstructure evolution have been investigated in some fcc metals, bcc metals and alloys by ion and neutron irradiations in recent years [1–10]. However, except for the final microstructures, it is difficult to investigate the microstructure evolution during neutron irradiation, especially just before and immediately after temperature change. In order to resolve this problem, a special type of in-core irradiation rig was developed in the Japanese Materials Test Reactor (JMTR), where it is possible to pull out several specimen sets during irradiation [11]. In the present work, pure Ni and its binary alloys of Ni–Si, Ni–Cu, Ni–Ge and Ni–Sn, where the role of alloying elements on void swelling have been well studied, were irradiated in the JMTR using the special rig. The purposes of this paper are to investigate the microstructure evolution at each temperature stage during temperature change irradiation, and the difference of alloying element effect on microstructure evolution between constant temperature irradiation and temperature change irradiation.

2. Experimental procedure

Pure nickel (99.99%) and its binary alloys containing 2 at.% Si (−5.81%), Cu (7.18%), Ge (14.76%) and Sn (74.08%), respectively, were tested in this study. The values in parentheses are volume size factors, which are the ratio of difference in volume between the solute and solvent atoms to the volume size of solvent atom [12]. Well annealed specimens were irradiated by the JMTR to doses up to 0.46 dpa under temperature change irradiation of 533/693 K with two cycles. The irradiation doses were 0.1 dpa at 533 K, then 0.12 dpa at 693 K, then 0.11 dpa at 533 K, then 0.13 dpa at 693 K.

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The specimen sets were pulled out five times during irradiation. After irradiation, microstructure features such as dislocations and voids were observed by transmission electron microscopy (TEM). All dislocation and void analyses were done by imaging the $g = [200]$ reflection near the $\langle 011 \rangle$ zone axis. Positron lifetime measurement was carried out to investigate the formation of microvoids which cannot be observed by TEM.

3. Results

3.1. Microstructure evolution in pure Ni

Figs. 1 and 2 show evolutions of the dislocation and void microstructure in pure Ni during temperature change irradiation, respectively. Interstitial type dislocation loops, dislocations and voids were formed even in the first irradiation at 533 K. In the beginning of the irradiation at high temperature (693 K), the density of voids decreased and the voids grew. The changes in void density and void size had the same trend with increasing dose at 693 K. In addition, the small dislocation loops disappeared in the initial irradiation at 693 K, but the dislocation density, which was 8.7×10^{12} m^{-2} and one order of magnitude higher than that in well annealed Ni, scarcely changed during the irradiation at 693 K. Compared with the first irradiation at 533 K, the densities of dislocations and voids increased in the second irradiation at 533 K. Against general expectation that void density could decrease in the second irradiation at high temperature of 693 K, it increased. The dislocation density decreased by 50% during the second irradiation at 693 K. The changes in void density, void size and swelling during temperature change irradiation are shown in Fig. 3. Although the irradiation dose after the second cycle irradiation was two times higher than that after the first cycle irradiation, the swelling in the former was 4.7 times higher than that in the latter.

3.2. Microstructure evolution in Ni binary alloys

The evolution of dislocation and void microstructures in Ni binary alloys of Ni–Cu and Ni–Ge were similar to those in pure Ni. In the Ni–Sn alloy as shown in Fig. 4, however, no damage microstructures were observed in the first cycle irradiation of 533/693 K. Only interstitial type dislocation loops were observed in the second irradiation at 533 K. Most of the loops disappeared and the loop density decreased by one order of magnitude in the subsequent irradiation at 693 K. The formation of dislocation and void microstructures was also suppressed in Ni–Si alloy. The change in loop density in the Ni–Si alloy during temperature change irradiation was the same as that in the Ni–Sn alloy. No voids were observed by TEM in both Ni–Si and Ni–Sn alloys. Fig. 5 shows the changes in positron lifetime

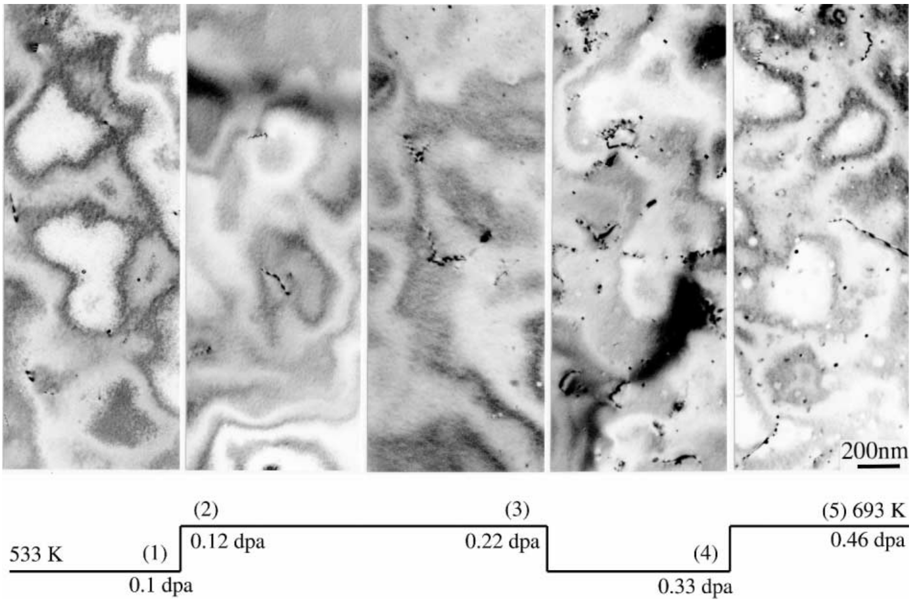

Fig. 1. Dislocation image of Ni during temperature change irradiation of 533/693 K. (1) After 533 K irradiation of 0.1 dpa; (2) after 693 K irradiation of 0.12 dpa; (3) after 693 K irradiation of 0.22 dpa; (4) after 533 K irradiation of 0.33 dpa; (5) after 693 K irradiation of 0.46 dpa.

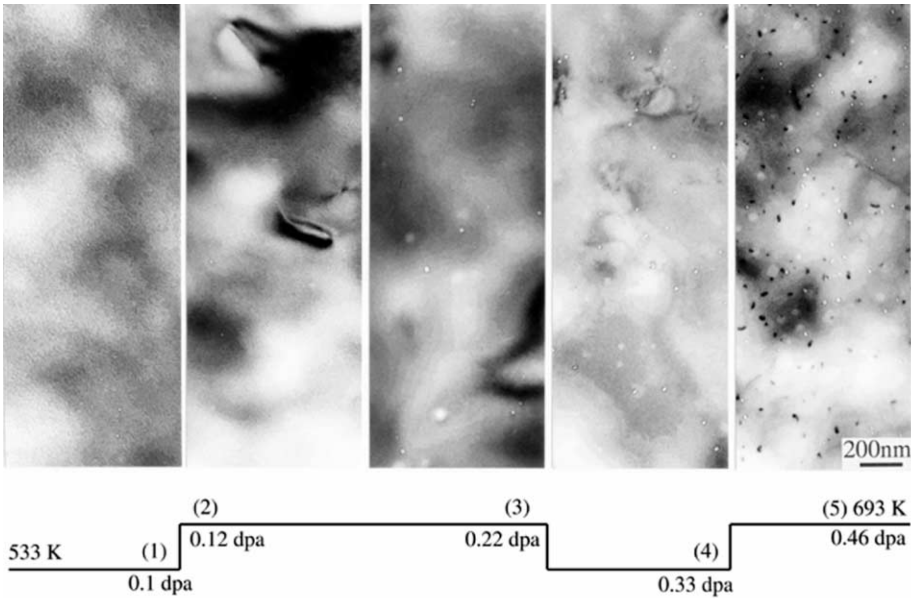

Fig. 2. Void image of Ni during temperature change irradiation of 533/693 K. (1) After 533 K irradiation of 0.1 dpa; (2) after 693 K irradiation of 0.12 dpa; (3) after 693 K irradiation of 0.22 dpa; (4) after 533 K irradiation of 0.33 dpa; (5) after 693 K irradiation of 0.46 dpa.

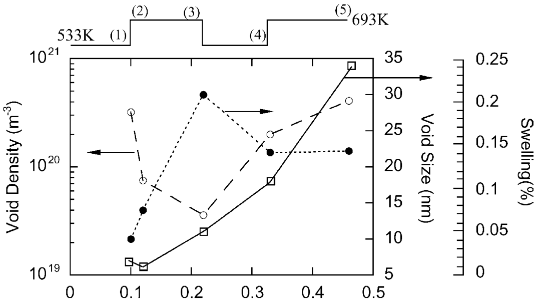

Fig. 3. Changes of void density, void size and swelling during temperature change irradiation of 533/693 K.

during the irradiation in the Ni–Si alloy, where τ_m is the positron mean lifetime. The shorter lifetime τ_1 mainly comes from free positrons in the matrix, and the longer lifetime τ_2 results from positrons trapped at defects, such as microvoids. The microvoids were formed in Ni–Si in the first irradiation at 533 K. The density of microvoids decreased and microvoids grew during the first irradiation at 693 K. In the second cycle irradiation of 533/693 K, however, two-lifetime-component analysis of measured lifetime spectra could not be carried out. It means that the microvoids disappeared in the second cycle irradiation of 533/693 K. While in Ni–Sn alloy, as the long lifetime component did not exist, no microvoids were formed during the temperature change irradiation.

4. Discussion

4.1. Mechanism of microstructure evolution in pure Ni

The densities of dislocation loops and voids decreased in Ni during the first irradiation at high temperature of 693 K. This behavior agrees with the results obtained in many experiments and simulations under temperature change irradiation [5–8,10,13,14]. The mechanism has been explained by the appearance of a vacancy-predominant condition after changing the temperature from low to high. Two vacancy sources are believed to exist. One is the reaction of vacancy clusters formed by the irradiation at low temperature and interstitials produced by the irradiation at high temperature [13]. Small vacancy clusters are dissolved into freely migrating vacancies by absorbing interstitials. The other is a dislocation bias effect, i.e., preferential absorption of interstitials at dislocations.

After the first irradiation of 533/693 K, the density of vacancy clusters decreased with increasing decomposition of small vacancy clusters. While the small dislocation loops disappeared by absorbing the excess vacancies at high temperature. The irradiation dose was almost the same in the first and the second irradiation at 533 K, but the nucleation and growth of dislocation loops formed by cascade damage and growth of voids were higher in the latter. The edge dislocations were formed after the first cycle irradiation of 533/693 K. Ogasawara [15] irradiated a neutron-irradiated Ni–Cu

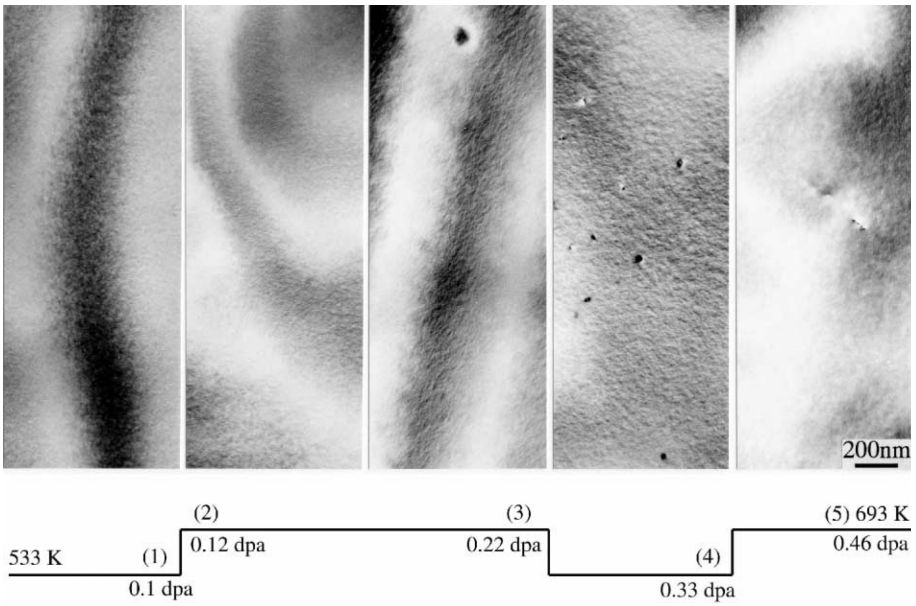

Fig. 4. Dislocation image of Ni–Sn alloy during temperature change irradiation of 533/693 K. (1) After 533 K irradiation of 0.1 dpa; (2) after 693 K irradiation of 0.12 dpa; (3) after 693 K irradiation of 0.22 dpa; (4) after 533 K irradiation of 0.33 dpa; (5) after 693 K irradiation of 0.46 dpa.

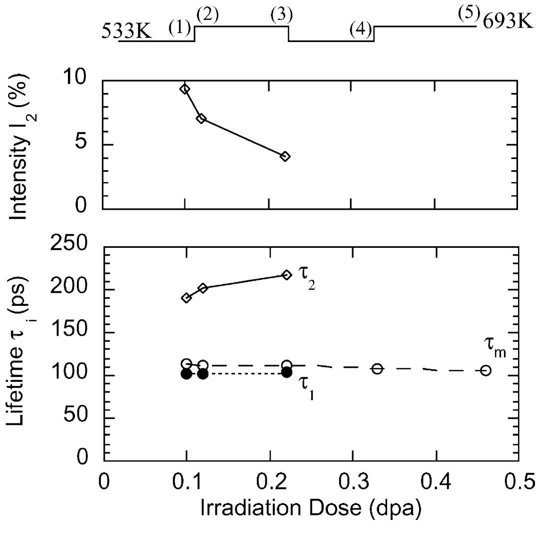

Fig. 5. Change of lifetime in Ni–Si alloy during temperature change irradiation of 533/693 K.

sample by high voltage electron microscopy, and found that the growth of interstitial type dislocation loops was enhanced at the dilatational side of edge dislocations. He suggested that migration efficiency of interstitials defined as M_IC_I was higher than that of vacancies M_VC_V in the dilatational side of edge dislocation, where M is defect mobility and C is defect concentration. The growth of interstitial type dislocation loops was promoted under interstitial-predominant condition. In the present study, the increase in dislocation density in the second irradiation at 533 K is also explained by this mechanism, because dislocation density before the second irradiation at 533 K was one order of magnitude higher than that before the first irradiation at 533 K. Compared with the voids formed in the first irradiation at 533 K, the void size was large in the second irradiation at 533 K. The higher growth of voids was caused by a dislocation bias effect, since more dislocations existed after the first irradiation at 693 K. During the second irradiation at 693 K, interstitials produced at the high temperature are annihilated at dislocations by their bias. The remaining vacancies contributed to nucleate voids. The biggest difference between the first and second irradiations of 533/693 K was the existence of dislocations.

4.2. Effects of alloy elements on microstructure evolution during temperature change irradiation

Damage microstructure evolution in Ni and its binary alloys has been well studied by the authors. Dislocation networks and voids were observed in Ni, Ni–Cu and Ni–Ge alloys irradiated by fission neutrons to a fluence of 3.7×10^{23} n/m^2 (0.11 dpa) at 573 K and 9.6×10^{23} n/m^2 (0.3 dpa) at 673 K [16,17]. The effects of alloy elements Cu and Ge on microstructure evolution in Ni are small. On the other hand, interstitial type dislocation loops with low density and no voids were observed in Ni–Si and Ni–Sn alloys. The nucleation and growth of interstitial type dislocation loops and voids were suppressed strongly in Ni–Si and Ni–Sn alloys due to increased recombination of interstitials and vacancies by the addition of Si or Sn in Ni [18,19]. In the present study, the effects of Cu and Ge on microstructure evolution in Ni under temperature change irradiation were the same as those in constant temperature irradiation at 573 and 673 K. Whereas the suppression of damage microstructure formation in Ni–Si and Ni–Sn alloys was stronger in the temperature change irradiation than in constant temperature irradiation. The small defect clusters formed at low temperature irradiation with low density disappeared at the next irradiation at high temperature. Consequently, almost no defect clusters remained in Ni–Si and Ni–Sn alloys after temperature change irradiation.

5. Conclusion

In order to investigate the effects of alloy elements on microstructure evolution in Ni under temperature change irradiation, Ni and its binary alloys were irradiated by fission neutrons with two cycles of 533/693 K temperature changes. It is clear that the damage microstructures formed in the first cycle irradiation of 533/693 K affect the nucleation and growth of damage microstructures in the next cycle irradiation. It was found that Cu and Ge scarcely change the microstructure evolution in Ni in temperature change irradiation. On the other hand, the microstructure evolution in Ni–Si and Ni–Sn alloys was more suppressed in the temperature change irradiation than that in constant temperature irradiation.

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