ELSEVIER

Journal of Nuclear Materials 283–287 (2000) 229–233

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

The effect of alloying elements on the defect structural evolution in neutron irradiated Ni alloys

T. Yoshiie [a,*], Q. Xu [a], Y. Satoh [a], H. Ohkubo [a], M. Kiritani [b]

[a] *Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka-fu 590-0494, Japan*
[b] *Department of Electronics, Hiroshima Institute of Technology, Miyake, Saeki-ku, Hiroshima 731-5193, Japan*

Abstract

The effect of alloying elements, Si (−5.8%: the volume size factor in Ni), Ge (+14.76%) and Sn (+74.08%), on void swelling in neutron irradiated Ni at 573 K was studied by transmission electron microscope (TEM) observation and positron annihilation lifetime measurement. Neutron irradiation dose was changed widely from 0.001 to 0.4 dpa using two reactors, the Kyoto University reactor (KUR) and the Japan materials testing reactor (JMTR). Voids were observed in pure Ni by TEM even after very small irradiation dose of 0.001 dpa. With increasing dose, the density of voids did not change much while their size increased. The same tendency was observed in Ni–2at.%Ge. In Ni–2at.%Sn and Ni–2at.%Si, however, no voids were observed by TEM at a damage dose of 0.4 dpa. But positron lifetime measurement revealed the existence of microvoids at a medium dose of irradiation. When irradiation dose increased to 0.4 dpa in Ni–2at.%Si and 0.13 dpa in Ni–2at.%Sn, their existence was not detected. Suppression of microvoids in these alloys is discussed from the standpoint of solute point defect interactions.

1. Introduction

Understanding the role of alloying elements on the defect structural evolution is important for the development of fusion reactor materials. We have studied the damage structures of pure Ni and its binary alloys, which contain Si (−5.8%), Cu (+7.18%), Ge (+14.76%) and Sn (+74.08%) [1–4]. They were chosen to change the volume size factor in Ni systematically as shown above in parentheses [5]. Well developed dislocation networks and voids were observed in Ni, Ni–Cu and Ni–Ge by fission neutron irradiation 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 using the Japan Materials Testing Reactor (JMTR), which is a 50 MW light-water-moderated reactor of the Japan Atomic Energy Research Institute [2,4]. On the other hand, no voids were observed in Ni–Si and Ni–Sn at the same irradiation condition; instead, a small number of interstitial type dislocation loops were observed.

Recently, a special type of in-core irradiation rig was developed in the JMTR [6], making it possible to pull out several specimen sets during irradiation. In the Kyoto University Reactor (KUR), a 5 MW light-water-moderated reactor, it is also possible to irradiate materials to a desired dose up to 0.05 dpa. In this paper, the growth behavior of voids in Ni and its alloys was studied over a wide range of irradiation dose using these two irradiation facilities.

2. Experimental

Specimens irradiated were 99.99% pure nickel (Johnson Matthey) and nickel base binary alloys, which contain Si, Ge and Sn as solute atoms. The concentration of solute atoms was 2 at.%. The alloys were rolled to 0.1 mm in thickness and 3 mm diameter discs were punched out. These discs were annealed at 1170 K for 1 h in vacuum.

Irradiation with fission neutrons was performed with the KUR to a fluence of 3.5×10^{22} n/m^2 (0.0049 dpa) and with the JMTR to a fluence of 1.3×10^{24} n/m^2 (0.4 dpa). The dpa was obtained using the SPECTER code [7] with

---

[*] Corresponding author. Tel.: +81-724 51 2620; fax: +81-724 51 2473.
E-mail address: yoshiie@rri.kyoto-u.ac.jp (T. Yoshiie).

0022-3115/00/$ - see front matter © 2000 Elsevier Science B.V. All rights reserved.
PII: S 0 0 2 2 - 3 1 1 5 (0 0) 0 0 0 7 0 - 2

a displacement threshold energy of 24 eV. The irradiation temperature was 573 K in all irradiations. Post-irradiation defect structure observation was performed with a JEOL JEM-2010 transmission electron microscope (TEM). All the positron lifetime measurements were performed at room temperature using a conventional apparatus with a time resolution of about 250 ps (FWHM). After source-background subtraction, lifetime spectra were resolved into two lifetime components, using the fitting program POSITRONFIT [8]. The lifetime of the annealed and unirradiated Ni and Ni alloys was about 109 ps and was not resolved into two components. As the lifetime of vacancies in Ni is 175 ps [9], a longer lifetime than 175 ps corresponds to three-dimensional vacancy clusters (microvoids). In Ni, TEM observations showed that voids have a monomial distribution [10]. So the lifetime of microvoids is also assumed to be one component. The short lifetime (100–170 ps) in Ni represents an average of lifetime characteristics of free positrons in bulk state and trapped positrons at simple defects such as edge dislocations (113 ps) and stacking fault tetrahedral (28 vacancies: 130 ps, 10 vacancies: 170 ps) in Ni [9]. Although positron lifetime calculations have not been performed yet on vacancy clusters and dislocations in Ni alloys, almost the same value (Ni–Si) or a little less value (Ni–Ge and Ni–Sn) is expected [11].

3. Results

Voids were observed in Ni by TEM even after a low irradiation dose of 0.026 dpa at 573 K as shown in Fig. 1. The microvoids were taken with an under-focus condition in order to observe voids. The void density and the swelling between 0.026 and 0.4 dpa are shown in Fig. 2. With increased dose, void swelling increased. While above 0.13 dpa, the density of voids decreased a little, up to a dose of 0.0049 dpa, voids were not yet observed with an under-focus condition. Interstitial type dislocation loops, 25 nm in size on an average, and small clusters, less than 2 nm in size, were observed under weak beam conditions. Although some of the small clusters were identified to be stacking fault tetrahedra by their TEM image contrast, the existence of voids could not be confirmed. Positron lifetime measurements showed the existence of a long lifetime component from the very low dose of 0.001 dpa as shown in Fig. 3. The mean lifetime stands for the single lifetime component or an averaged lifetime between the short and the long ones. The intensity of the long lifetime component increased with increasing irradiation dose. The existence of microvoids in an early stage of irradiation was also detected in Ni–Ge as shown in Fig. 4. In this case, the increase of the intensity of the long lifetime component was not significant. Voids were observed by TEM above a dose of 0.026 dpa.

In Ni–Sn and Ni–Si, no voids were observed, and only a low density of interstitial type dislocation loops was observed by TEM up to a dose of 0.4 dpa. By positron lifetime measurement, however, the existence of vacancies and their clusters was detected in some stage of irradiation in both alloys. In the case of Ni–Si, the existence of microvoids was detected by a dose of 0.001 dpa as shown in Fig. 5. On the other hand, only in the irradiation dose between 0.0049 and 0.062 dpa, the

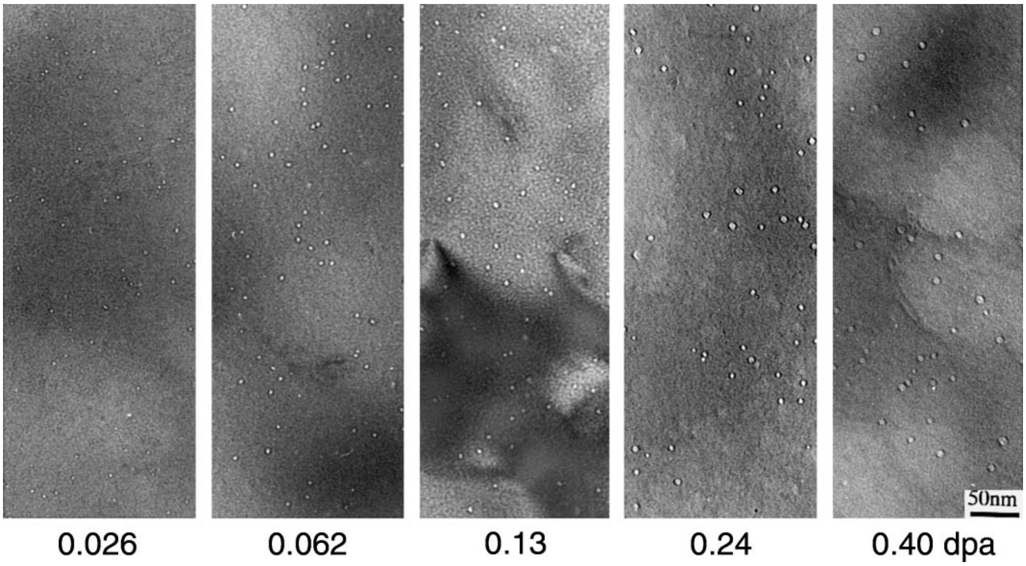

Fig. 1. Comparison of defect structures in Ni irradiated at five irradiation doses by JMTR at 573 K.

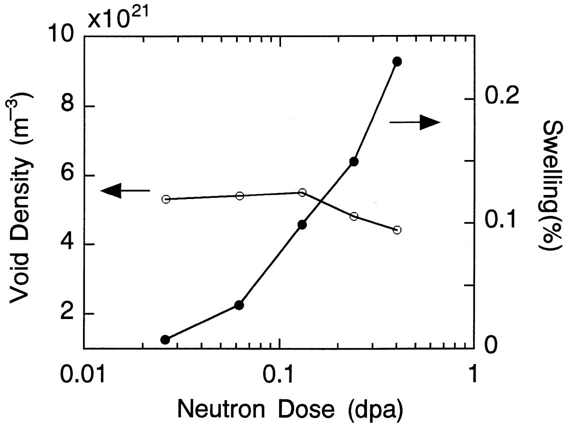

Fig. 2. Variation of void density and swelling in Ni as a function of irradiation dose.

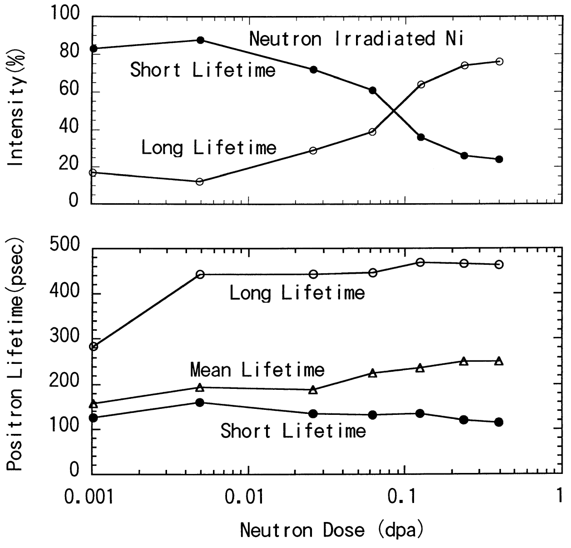

Fig. 3. Variation of positron mean lifetime, long lifetime, short lifetime, and their intensities in Ni as a function of irradiation dose.

lifetime corresponding to vacancies was detected in Ni–Sn (Fig. 6). When irradiation dose increased to 0.4 dpa in Ni–2at.%Si and 0.13 dpa in Ni–2at.%Sn, two-component decomposition of the lifetime was not possible. The summary of the TEM observation and the positron lifetime measurement is in Table 1.

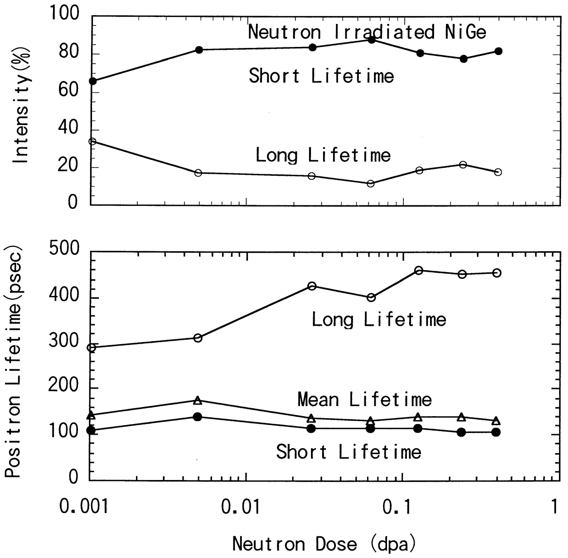

Fig. 4. Variation of positron mean lifetime, long lifetime, short lifetime, and their intensities in Ni–2at.%Ge as a function of irradiation dose.

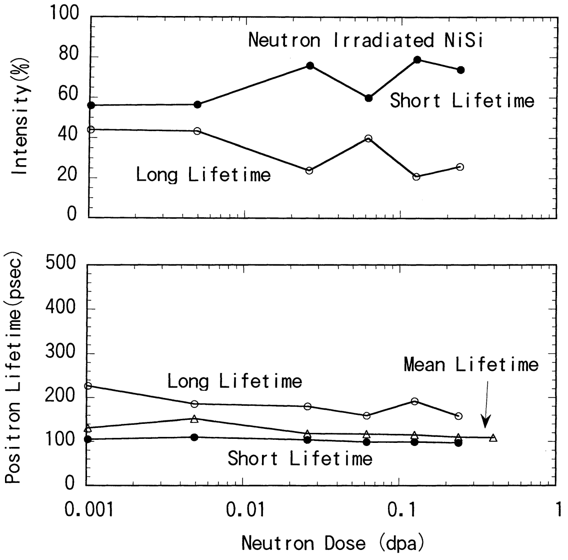

Fig. 5. Variation of positron mean lifetime, long lifetime, short lifetime, and their intensities in Ni–2at.%Si as a function of irradiation dose.

4. Discussion

Vacancies in Ni and its alloys are thermally mobile above 473 K [12]. The nucleation of voids in pure Ni finished in an early stage of irradiation at 573 K. With increasing irradiation dose, the lifetime of the long lifetime component increased and saturated at a dose of 0.0049 dpa, while its intensity continued to increase above 0.0049 dpa. The lifetime is known to be sensitive to the size of voids when their size is small, while it becomes constant as they grow larger [13]. On the other hand, the intensity of the long lifetime component is

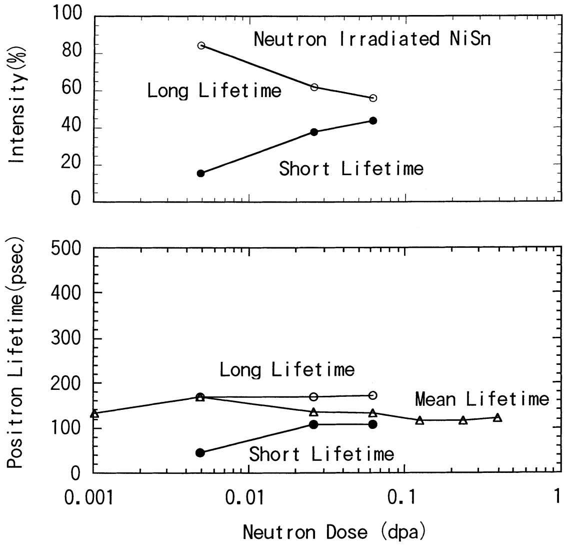

Fig. 6. Variation of positron mean lifetime, long lifetime, short lifetime, and their intensities in Ni–2at.%Sn as a function of irradiation dose.

Table 1
The summary of TEM observation and positron lifetime measurement

TEM observations

Dose (dpa)	Ni−Si	Ni	Ni−Ge	Ni−Sn
0.0010	Low density of I−loops	Low density Of I − loops and SFT	Low density of I−loops and SFT	Low density of I−loops
0.0049				
0.026		Voids Void size increases ↓	Voids Void size increases	
0.062				
0.13				
0.24				
0.40				

Increasing volume size factor →

Positron lifetime Measurements

Dose (dpa)	Ni−Si	Ni	Ni−Ge	Ni−Sn
0.0010	Microvoids	Microvoids The size increases ↓	Microvoids The size increases ↓	
0.0049	Vacancies (Microvoids)			Vacancies (Microvoids)
0.026				
0.062				
0.13				
0.24				
0.40				

generally related to the density of the trapping site: the number density of voids in this case. So the constant intensity and increasing lifetime in pure Ni to the dose of 0.0049 dpa suggest that voids grew larger without changing their number density during the irradiation. The increase of the intensity above 0.0049 dpa is not consistent with the results of TEM observation: the void concentration did not increase as can be clearly seen in Fig. 2. So we can conclude that the increase also corresponds to an increase in the void size. With increasing void size, the probability of positrons to be trapped by voids will increase, even if the number density of voids is constant.

In Ni–Ge, the lifetime of the long lifetime component increases with increasing irradiation dose up to 0.13 dpa, while the intensity did not change. This suggests that the concentration of microvoids did not change so much and only voids grew larger up to the final dose. The essential difference between pure Ni and Ni–Ge is the lower growth rate of voids in Ni–Ge. This can be explained by a decrease in vacancy mobility by the solute vacancy interaction.

Nuclei of microvoids will be formed directly in a cascade area, where vacancy concentration is high. So the existence of microvoids in Ni–Si is expected in an early stage of irradiation. As Si is an undersize element in Ni, interstitials will be trapped by Si easily. With increasing dose, a large amount of trapped interstitials and their clusters will be formed. They work as an effective site for the annihilation of freely migrating vacancies, and suppress the growth of microvoids.

In Ni–Sn, the nucleation of microvoids directly in a cascade area is also expected just the same as other alloys. But in this case, the growth of microvoids will be delayed by Sn, which will trap the vacancies. Accordingly, microvoids were not detected in Ni–Sn in an early stage of irradiation. At an irradiation dose between 0.0049 dpa and 0.062 dpa, the existence of a long lifetime component 180 ps, which almost corresponds to single vacancies in pure Ni, was detected as shown in Fig. 6. However, the lifetime is probably not that of vacancies, but of microvoids, which grow with increasing dose, because the positron lifetime of trapped vacancies with oversize element is shorter than that of free vacancies [11]. At higher irradiation dose, the concentration of interstitial clusters becomes high, and the growth of microvoids is prevented owing to the absorption of vacancies by the interstitial clusters.

The increase of interstitial clusters in Ni–Si and Ni–Sn also prevents the detection of a long lifetime component of microvoids. In positron lifetime experiments, interstitial clusters produce much shorter lifetimes (<113 ps [9]) than vacancies. If the trapping site of interstitial clusters is superior to that of microvoids, two-component decomposition of positron lifetime becomes difficult even if long lifetime component exists.

In this paper, we compared the results obtained in two different reactors using only the dpa. It is well known that the dpa is not sufficient to account for the irradiation damage in metals. It ignores the distribution of displaced atoms in a cascade. Two reactors are, however, light-water-moderated ones and the difference of the spectra is not significant. The detailed discussion will be presented in a paper [14].

5. Conclusion

The effect of alloying elements on void growth in Ni was studied. In Ni, the existence of microvoids was detected at a dose of 0.001 dpa by positron lifetime measurement, and at 0.026 dpa, voids were observed by TEM. The same tendency was observed in Ni–Ge. Although no voids were observed by TEM in binaries containing the undersize element Si and the oversize element Sn, microvoids were detected between 0.001 and 0.24 dpa in Ni–Si, and between 0.0049 pa and 0.062 dpa in Ni–Sn by positron lifetime measurement. With increasing irradiation dose, interstitial clusters formed, and acted as an effective sink for vacancies, preventing the growth of microvoids in these alloys.

References

[1] S. Kojima, T. Yoshiie, M. Kiritani, J. Nucl. Mater. 155–157 (1988) 1249.

[2] M. Kiritani, T. Yoshiie, S. Kojima, Y. Satoh, K. Hamada, J. Nucl. Mater. 174 (1990) 327.

[3] T. Yoshiie, S. Kojima, Y. Satoh, K. Hamada, M. Kiritani, J. Nucl. Mater. 191–194 (1992) 1160.

[4] K. Hamada, S. Kojima, Y. Ogasawara, T. Yoshiie, M. Kiritani, J. Nucl. Mater. 212–215 (1994) 270.

[5] H.W. King, J. Mater. Sci. 1 (1966) 79.

[6] M. Narui, H. Kurishita, H. Kayano, T. Sagawa, N. Yoshida, M. Kiritani, J. Nucl. Mater. 212–215 (1994) 1665.

[7] L.R. Greenwood, ANL/FPP/TM-197, Argonne National Laboratory, Argonne, IL, January 1985.

[8] P. Kirkegaard, N.J. Pedersen, M. Eldrup, Riso-M-2740, Riso National Laboratory, Denmark, 1989.

[9] E. Kuramoto, T. Tsutsumi, K. Ueno, M. Ohmura, Y. Kamimura, Computational Mater. Sci. 14 (1999) 28.

[10] M. Kiritani, T. Yoshiie, S. Kojima, Y. Satoh, K. Hamada, J. Nucl. Mater. 174 (1990) 327.

[11] Y. Shirai, T. Hashimoto, T. Takeshita, K. Furukawa, J. Takamura, in: J. Takamura, M. Doyama, M. Kiritani (Eds.), Proceedings of the Fifth Yamada Conference on Point Defects and Defect Interactions in Metals, Kyoto, 1981, p. 441.

[12] M. Kiritani, M. Konno, T. Yoshiie, S. Kojima, Mater. Sci. Forum 15–18 (1987) 181.

[13] M.J. Puska, R.M. Nieminen, J. Phys. F 13 (1983) 333.

[14] T. Yoshiie, X. Xu. Q. Xu, S. Yanakita, Y. Satoh, in: J.G. Williams, D.W. Vehar, F.H. Ruddy, D.M. Gilliam (Eds.), Proceedings of the 10th International Symposium on Reactor Dosimetry, ASTM STP 1398, 2000, in press.