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Detection of interstitial clusters in neutron irradiated Ni–Hf alloy by perturbed angular correlation and positron annihilation lifetime measurements

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Abstract

In order to understand the effect of alloying elements on the damage structure evolution in metals, defect clusters near Hf in Ni were studied using the perturbed angular correlation (PAC) technique and positron annihilation spectroscopy (PAS). The volume size factor of Hf in Ni was determined to be 61% by measuring the lattice parameter. The positron annihilation mean lifetime of Ni–0.5 at.%Hf after neutron irradiation at 473 K up to a dose of 0.0053 dpa was 132 ps. The lifetime decreased with increasing annealing temperature and by annealing at 723 K the recovery was finished. The PAC spectrum of ^{181}Ta ($\leftarrow$^{181}Hf) taken after the irradiation indicated that there were two components. The first component consisted of a Larmor frequency of 534 mega-radian/s (Mrad/s). The second consisted of a very broad range of frequencies which brought destructive interference among them and made the contribution to the spectrum almost zero. The component disappeared with annealing at 873 K. It was concluded that the first component and the second component represent Hf in the regular substitutional site and Hf with defect clusters, respectively. From the difference of annealing out temperature of defect clusters, clusters annihilated by 723 K were assigned as stacking fault tetrahedra (SFTs) and those by 873 K as interstitial clusters.

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1. Introduction

The understanding of the roles of alloying elements in defect structure evolution is important for the design of fusion reactor materials. Usually we assume that oversized elements trap vacancies and prevent void growth, and that undersized elements interact with interstitials and delay the growth of interstitial type dislocation loops. The authors have studied the effect of alloying elements on the defect structure evolution in Ni with changing volume size factors schematically using the transmission electron microscopy and the positron annihilation spectroscopy (PAS) [1–4]. The volume size factor represents the ratio of volume difference of solute element to solvent. Alloying elements used were Si (−5.81), Cu (7.18), Ge (14.76) and Sn (74.08), where the value in the parenthesis is the volume size factor in Ni [5]. It has been clarified that void formation by neutron irradiation is delayed by the addition of Si and Sn to the irradiation dose of 0.5 dpa. Recently the importance of one dimensional motion of interstitial clusters for void growth has been pointed out by many researchers. Oversized elements are expected to trap interstitial clusters and prevent their one dimensional motion. The experimental evidence of the interaction between interstitial clusters and oversized elements is, however, scarce.

Perturbed angular correlation (PAC) of gamma-rays is sensitive to the local atomic arrangements around radioactive probe atoms. The interaction between the nuclear dipole and quadrupole moments of the probe

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and the local hyperfine fields caused by a non-cubic lattice or by nearby defects is detectable. Hf is one of elements suitable for PAC measurement through the formation of ^{181}Hf with irradiation by thermal neutrons and is oversized in Ni. In this paper, we report detection of vacancy clusters and interstitial clusters near Hf in Ni using the PAC and the PAS techniques.

2. Experimental procedure

Ni–Hf alloys (1, 0.5 and 0.1 at.%Hf) were prepared by arc melting. They were cold-rolled to 0.1 mm and quenched in water from 1473 K, since the equilibrium Hf concentration at room temperature is almost zero. The lattice constant of each specimen was measured using a conventional X-ray diffractometer (RINT2000, Rigaku). X-ray analysis indicated the existence of Hf precipitates only in Ni–1 at.%Hf. So we mainly used Ni–0.5 at.%Hf in this experiment. The specimens were irradiated with fission neutrons at 473 K up to a dose of 0.0053 dpa using the Materials Controlled Irradiation Facility (SSS) in the Kyoto University Reactor (KUR) [6]. During this irradiation, damage was formed by high energy neutrons and ^{181}Hf was created by thermal neutrons at the same time. In order to obtain information before irradiation, the specimens were exposed to thermal neutrons using the Pneumatic Tube of the KUR. For the comparison of defect structure evolution, pure Ni, Ni–2 at.%Ge and Ni–2 at.%Sn were also irradiated in the SSS at the same irradiation condition. Damage structures were investigated by PAC and PAS measurements at room temperature. The PAC study was carried out using a standard BaF_2 detector system to observe the cascade gamma-rays through the 482 keV excited level of ^{181}Ta, the daughter nucleus of the ^{181}Hf decay. The PAS was performed using the fast-fast coincidence system with ^{22}Na, whose lifetime resolution was about 190 ps. The isochronal annealing of specimens for 1 h was performed up to 873 K.

3. Experimental results

3.1. Volume size factor of Hf in Ni

The volume size factor of Hf in Ni was determined according to the method described in Ref. [5]. The atomic volume of solid solution alloys, which was obtained taking the volume of the unit cell divided by the number of atoms in the cell, was measured with the change of Hf concentration as shown in Fig. 1. The effective atomic volume of the Hf was obtained by linear extrapolation of the volume plot to 100% and the volume size factor of Hf in Ni was determined to be 61 ± 5% oversized.

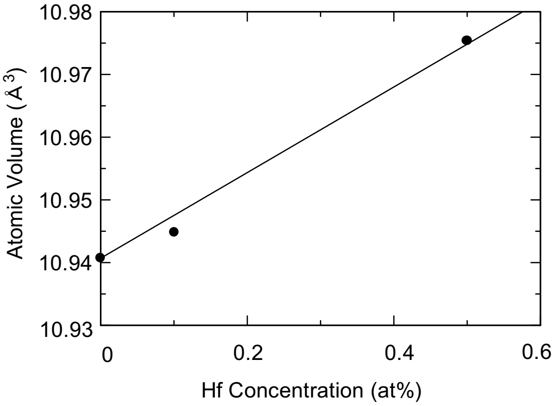

Fig. 1. The change of atomic volume as a function of Hf concentration in Ni.

3.2. Positron annihilation lifetime measurements

The results of positron annihilation lifetime measurement of neutron irradiated Ni, Ni–2 at.%Ge, Ni–2 at.%Sn and Ni–0.5 at.%Hf to 0.0053 dpa at 473 K are shown in Fig. 2. A long lifetime more than 280 ps which indicated the existence of microvoids was obtained in Ni and Ni–2 at.%Ge. The long lifetime of Ni–0.5 at.%Hf was 135 ps and almost the same as its mean lifetime of 132 ps. This lifetime indicates no void formation in Ni–0.5 at.%Hf as well as Ni–2 at.%Sn. The mean lifetime of

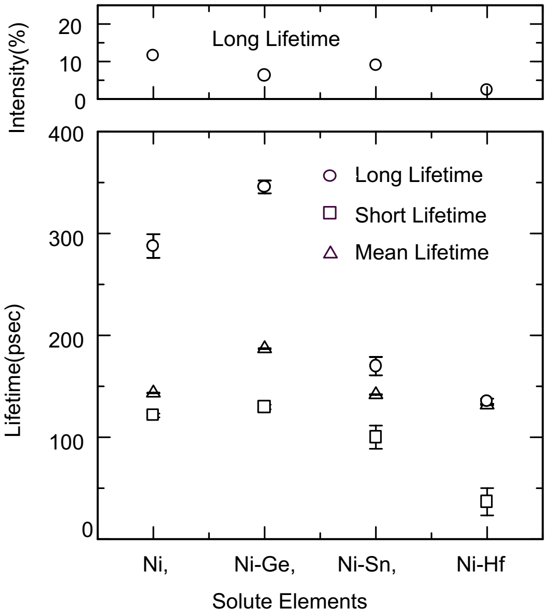

Fig. 2. The result of positron annihilation lifetime measurements in neutron irradiated Ni, Ni–2 at.%Ge, Ni–2 at.%Sn and Ni–0.5 at.%Hf at 473 K to 0.0053 dpa.

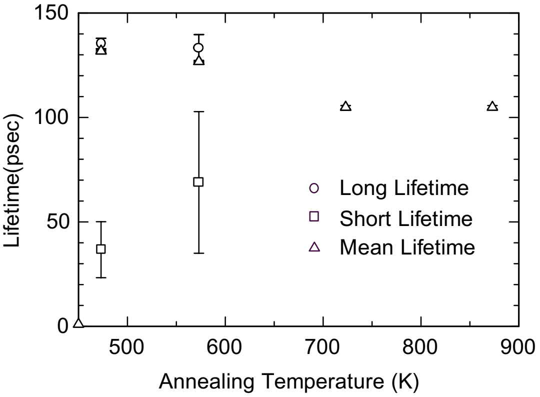

Fig. 3. The annealing behavior of positron annihilation lifetime in neutron irradiated Ni–0.5 at.%Hf at 473 K to 0.0053 dpa.

Ni–0.5 at.%Hf decreased with increasing annealing temperatures as shown in Fig. 3. Following annealing at 723 K, the recovery was completed.

3.3. PAC measurements

Fig. 4 shows the PAC spectra after the Pneumatic Tube irradiation (a), after the SSS irradiation at 473 K (b), after annealing at 573 K (c), 723 K (d) and 873 K (e). In each spectrum a sharp modulation is observed. The PAC spectra were analyzed by three parameters using the following equation

$$R(t) = A(1 + 2\cos\omega t + 2\cos 2\omega t) + C,$$

where A, ω and C are the effective angular correlation coefficient, the Larmor frequency and a constant, respectively. The Larmor frequency of the Pneumatic Tube irradiated specimen (Fig. 4(a)) was 534 Mrad/s. The PAC spectrum taken after the SSS irradiation (Fig. 4(b)) indicated that there were two components. The first component consists of a Larmor frequency of 534 Mrad/s. The second which disappears after the annealing at 873 K consists of very a broad range of frequencies which brings destructive interference among them so that this component dose not contribute to the PAC spectrum.

4. Discussion

The positron annihilation long lifetime of Ni–0.5 at.%Hf just after neutron irradiation by the SSS corresponds to the annihilation lifetime of positrons at SFTs [7]. The disappearance temperature of the long lifetime component is between 573 and 723 K. This temperature region is higher than the annihilation temperature of SFTs in pure Ni, between 423 and 523 K [8]. The increase of annealing temperature is, however, explained by the stabilization of stacking fault tetrahedra (SFTs) by the addition of oversized elements [9] and we conclude that the main annihilation sites of positrons are SFTs.

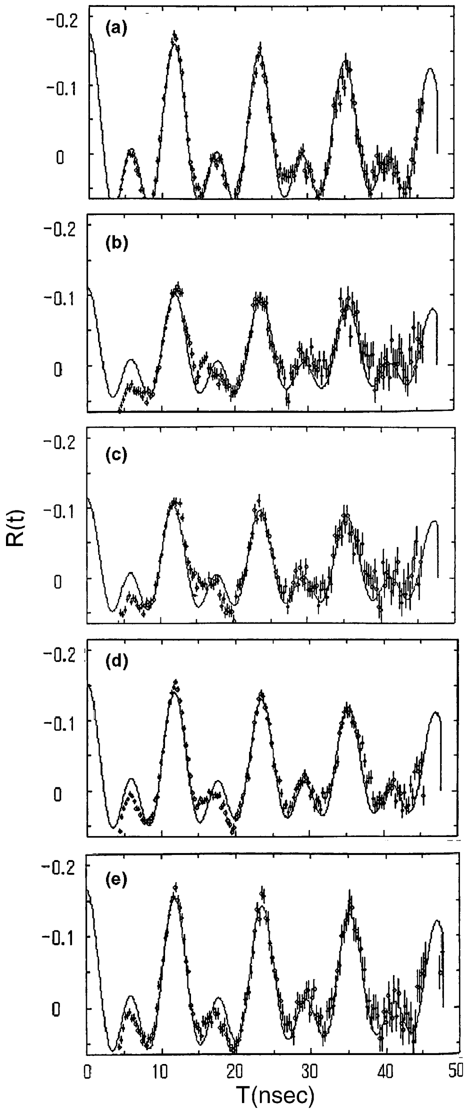

Fig. 4. The result of PAC measurement of Ni–0.5 at.%Hf. After the Pneumatic Tube irradiation at 300 K (a), after the SSS irradiation at 473 K to 0.0053 dpa (b), and after subsequent annealing of the SSS irradiated specimen at 573 K (c), 723 K (d) and 873 K (e) for 1 h.

The Larmor frequency of the Pneumatic Tube irradiated specimen and the first component of the SSS irradiated specimen at 473 K was 534 Mrad/s which is essentially the same as the value 532 Mrad/s obtained by Kaufman et al. [10]. They assigned this frequency to Hf in the substitutional site of Ni. We conclude that the first

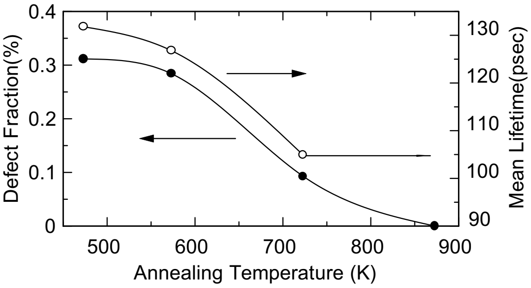

Fig. 5. The annealing behavior of the defect fraction $D(T)$. The annealing behavior of the positron annihilation mean lifetime is also shown for comparison.

component and the second component represent Hf in the regular substitutional site and Hf with defects, respectively.

In order to study the annealing behavior of defects with Hf, we defined a defect fraction $D(T)$ as a function of annealing temperature T as follows,

$$D(T) = \{A\ (873\ K) - A(T)\}/A\ (873\ K).$$

Fig. 5 shows the change of $D(T)$ with annealing temperature. The change of positron mean lifetime is also shown in the same figure. If the defects are single vacancies adjacent to Hf, a well defined frequency component is expected [10]. Therefore, SFTs and interstitial clusters are considered as the defects. There are many kinds of different atomic configurations for Hf in SFTs. Interstitial clusters of a bundle of crowd ions are expected to be trapped by oversized element like Hf. There are also many atomic configurations near Hf according to the number of interstitials. Hf with these defects causes a broad range of frequencies.

The difference of the annealing temperatures of defects between the PAC measurement and the PAS measurement is also understood by this assignment. With positron annihilation lifetime measurements, it is possible to detect SFTs. Interstitial clusters which are stable above 723 K are not easy to detect with the positron annihilation lifetime measurement, since the lifetime of positrons at interstitial clusters is almost the same as that at dislocations. Therefore, above 723 K, interstitial clusters were observed only by the PAC measurement.

5. Summary

The results are summarized as follows. (1) The volume size factor of Hf in Ni was determined to be 61 ± 5%. (2) The existence of interstitial clusters near Hf in neutron irradiated Ni–0.5 at.%Hf was confirmed by comparison between the PAC measurement and the positron annihilation lifetime measurement.

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