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Application of positron beam Doppler broadening technique to ion beam irradiation in nickel

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Abstract

The formation processes of vacancy-type defects induced by ion irradiation in nickel were investigated with a variable-energy positron beam Doppler broadening technique in order to find the appropriate experimental condition for in situ positron beam experiments. Nickel ion irradiation at 300–343 K induces an *S* parameter increase, but the dependence of the *S* parameter on positron energy is very similar to that of cold-rolled nickel, which suggests positron trapping by small vacancy-type defects such as SFTs. The best experimental condition for investigation of cavity nucleation process by in situ positron beam Doppler experiments is suggested to be around 0.1–0.4 dpa at 773 K.

1. Introduction

Void swelling is one of the characteristic and serious problems of structural materials for fusion reactors. Therefore, the void formation has been widely studied experimentally and theoretically for several decades. Void growth is generally treated on the basis of the difference of vacancy and interstitial fluxes to voids and vacancy emission from voids that are controlled by temperature, dose rate, impurity, and bias factor of sinks [1,2]. On the other hand, void nucleation is less understood than void growth, mainly because of the experimental difficulty in detecting small vacancy clusters at the nucleation stage during irradiation.

Because positron annihilation is well-known for its high sensitivity in detecting small vacancy-type defects, we fabricated a positron beam Doppler broadening instrument connected to an ion beam irradiation chamber in order to detect vacancy-type defects during ion beam irradiation [3,4]. As reported in [3], no significant difference in the *S* parameter between beam-on and beam-off conditions was found for pure iron at room temperature, but this can be attributed to thermal stability of nanovoids in iron at room temperature, i.e. nanovoids produced by ion irradiation remain almost unchanged at room temperature.

In the present work, we are searching for an appropriate experimental condition for in situ

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measurement of void nucleation process using a positron beam. Nickel is a metal that swells like other FCC metals, and the swelling behavior of nickel has been well investigated with transmission electron microscopy (TEM) for both neutron and ion irradiation [5]. Microstructure evolution of nickel during ion irradiation was also investigated by in situ TEM [6–8]. In addition, we have observed differences in the S parameter between beam-on and beam-off conditions for nickel foil at room temperature [9]. For these reasons, nickel was chosen for this positron beam examination of the void nucleation process.

The purpose of the present work was to find the most appropriate irradiation and measurement conditions of nickel for in situ positron beam Doppler broadening measurement. For this purpose, nickel ion irradiation to pure nickel was carried out at various temperatures and variable-energy positron beam Doppler broadening measurements were carried out after irradiation.

2. Experimental

Pure nickel (99.997%) was used for the specimens. Nickel sheets of 1 mm in thickness were cold-rolled to 80% reduction in thickness, and then cut into 20 mm squares. They were annealed at 1073 K for an hour in vacuum. Ion irradiations were carried out with a 1 MV tandem accelerator at the High Fluence Irradiation Facility, Nuclear Professional School, The University of Tokyo (HIT). Accelerated ions were 4 MeV Ni^{3+} for all the irradiations and irradiation temperatures were 773 K, 673 K, 573 K, and room temperature. However, irradiation at room temperature led to beam heating up to 343 K. Maximum fluence level was 1 dpa (averaged value from surface to 1 μm in depth), and the dose rate was $(1.5 \pm 0.5) \times 10^{-4}$ dpa/s.

The variable-energy positron beam has been equipped at HIT and connects to the irradiation chamber of the Van de Graaff accelerator for the in situ or in-chamber positron beam Doppler broadening experiment [3,4]. A sealed ^{22}Na isotope of 740 MBq was used as the positron source and a tungsten foil served as a positron moderator. Generated positrons from β^{+} decay are thermalized and emitted from the moderator, and then extracted by an electric field of 50 V. They are magnetically transported along the S-figure beamline to the sample chamber, where a high purity germanium detector is equipped for Doppler broadening measurement. Doppler broadening spectra can be measured at controlled positron energies from 50 eV to 30 keV.

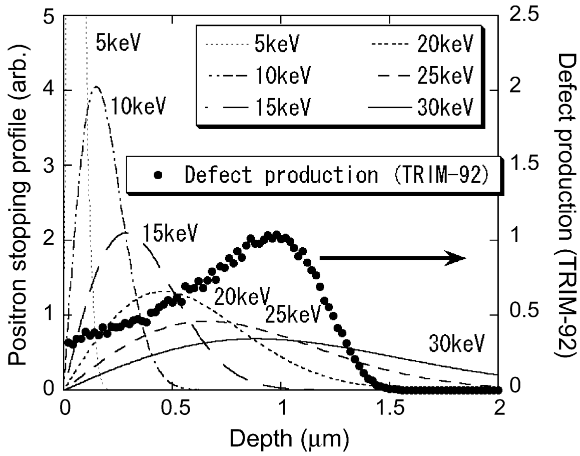

Fig. 1. Depth profiles of defect production (●) by 4 MeV Ni^{3+} calculated with the TRIM-92 code [10] and the stopping probability of positrons of various energies (lines).

Depth profiles of defect production were calculated with the TRIM-92 code [10] and the stopping probability of positrons are shown in Fig. 1. The positron stopping profile was calculated by the following equation [11,12]:

$$P(x) = (m/x_0)(x/x_0)^{m-1} \exp\{-(x/x_0)^m\}, \quad (1)$$

where $x_0 = 1.13\bar{x}$ and $\bar{x} = \alpha_p E^n/\rho$ (the mean depth), E is the positron energy in keV, ρ is the density of the material in g/cm^3, and α_p, m, and n are material independent parameters. Values of $m = 2$, $n = 1.7$, and $\alpha_p = 2.75$ μg/cm^{-2} keV^{-n}, experimentally determined by Gebauer et al. [13], were used for this calculation. This plot shows the variation of energies from 50 eV to 30 keV can sufficiently cover the whole damaged region by 4 MeV Ni^{3+} beam.

Vacancy-type defects in nickel are known to sharpen the energy spectrum of annihilation photons, and the S parameter, which is defined as the ratio of counts in the central region (511 ± 0.4 keV in this paper) to counts in the whole photo peak, is used to parameterize the peak shape change. Increase in the S parameter means production of vacancy-type defects in nickel and, in general, the S parameter can be affected by both number density and open volume size. However, when most of positrons annihilates at trapping sites, an S parameter value reflects its open volume size. The dependence of S parameter on positron energy was analyzed with a fitting program called 'VEP-

FIT' so as to extract information on depth profile under an assumption of either Gaussian or box-type depth profile of defects [14]. In the fitting analysis, the data of 50 eV positrons are excluded because contribution of epithermal positrons is significant at this energy.

3. Results and discussion

3.1. Annealed and cold-rolled nickel

Fig. 2 shows the dependence of the *S* parameter on implanted positron energy (*S*–*E* curve) for annealed and cold-rolled nickel. The curve for annealed nickel is a typical curve shape for annealed metals, which gradually decreases with increasing positron energy and approaches an *S* parameter value for bulk annihilation (S_b). The curve for cold-rolled nickel is clearly different from that for the annealed one in two aspects, one is the lower positron energy where the *S* parameter reaches a stable value, and the other is the large *S* parameter value that does not depend on positron energy above 10 keV. The lines in Fig. 2 shows the fitting curves calculated with VEPFIT [14] using a simple assumption of two annihilation sites (surface and bulk). The fitting estimates the diffusion length of positrons to be 184 nm for the annealed and 59 nm for the cold-rolled, and this reduction means the formation of trapping sites by cold rolling. The large *S* parameter indicates the trapping sites seem to have open volume like vacancy-type defects. Ohkubo et al. reported the effect of cold rolling on positron lifetime with a value of 170 ps being found for cold-rolled nickel when the reduction in thickness is greater than 20% [15]. Kuramoto et al. also used positron lifetime spectroscopy for cold-rolled nickel of 5% reduction in thickness and obtained about 185 ps or τ_2 [16]. As these lifetime values are attributed to monovacancies [15] and divacancies on dislocations [16], the observed trapping sites are thought to be such small vacancy-type defects, and the *S* parameter value (0.48) should correspond to such defects with small open volume.

3.2. Ni ion irradiation at 573 K and below 573 K

Fig. 3 shows *S*–*E* curves for ion-irradiated nickel at 573 K to 0.1 and 1 dpa, and at 300–343 K to 0.1 dpa. It is notable that the measured *S* parameter for that irradiated at 300–343 K is quite similar to that for the cold-rolled nickel, which indicates that the trapping sites are small vacancy-type defects. In this irradiation temperature range close to the Stage III of nickel, individual vacancies introduced by irradiation can act as the positron trapping sites and lead to a similar *S* value to the cold-rolled nickel. Vacancy clusters are also known to exist as stacking fault tetrahedra (SFTs) in nickel irradiated with neutrons [17]. Positron lifetime of SFTs was calculated by Kuramoto et al. to vary from 183 ps (3 vacancies) to 130 ps (28 vacancies) [16]. This calculation suggests the open volume of SFTs is

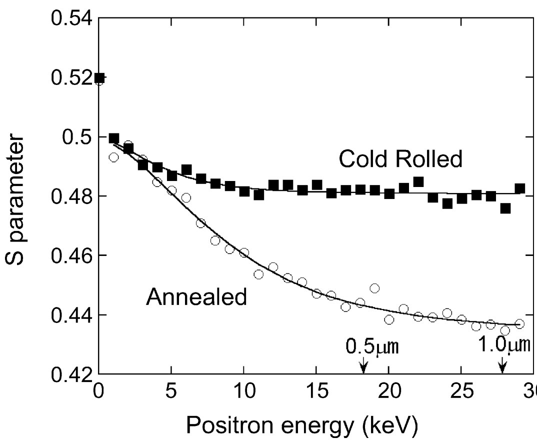

Fig. 2. Dependence of *S* parameter on implanted positron energy for annealed (○) and cold-rolled (■) nickel. Lines are the fitted curves by VEPFIT [14]. Arrows with 0.5 and 1.0 μm indicate the energy corresponding to the mean implantation depth of positrons.

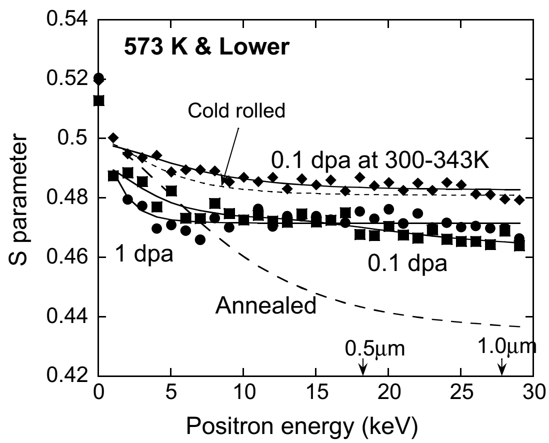

Fig. 3. Dependence of *S* parameter on implanted positron energy for ion-irradiated nickel at 573 K to 0.1 dpa (■) and 1 dpa (●), and ion-irradiated nickel at 300–343 K to 0.1 dpa (◆). The curves for the annealed and cold-rolled nickel are also plotted for reference. Lines are the fitted curves by VEPFIT [14].

similar to or less than monovacancies. The observed large S value indicates that SFTs are not dominant for trapping positrons at this temperature.

As for irradiation at 573 K, the curve shape that reaches a constant S value at low positron energies indicates so large a population of trapping sites that most of positrons should be trapped. Larger population of trapping sites for 1 dpa leads to reduction in the fraction of positrons which escape to surface than 0.1 dpa, and it apparently makes S parameter slightly lower for low energy positrons. The constant S value, which is lower than that of the cold-rolled and the irradiated at room temperature, indicates that the open volume of trapping sites is smaller than vacancies and SFTs formed at room temperature. Although we cannot identify the nature of trapping sites for that irradiated at 573 K, larger SFTs and/or dislocations are possibly acting as trapping sites because larger SFTs have smaller open volume which would result in lower positron lifetime [16,18].

3.3. Ni ion irradiation at 673 K

Fig. 4 shows the S–E curves for ion-irradiated nickel at 673 K to 0.1, 0.4 and 1 dpa. The S–E curve for 0.1 dpa at 673 K shows an increase at overall positron energies compared to the unirradiated one, but the increment is smaller than that for irradiation at 573 K. The shape of S–E curve does not indicate significant decrease in diffusion length of positrons compared to the annealed nickel, which means the population of trapping sites are not so high as the situation where most of positrons are trapped.

On the other hand, the curves for 0.4 and 1 dpa have an interesting shape. The S parameters for low energy positrons between 3 and 12 keV are significantly increased by irradiation up to nearly a constant value of 0.50, and the S parameters decrease with increasing positron energy above 12 keV. This shape and the large constant S value (∼0.50) imply that formation of vacancy clusters occurs mainly in the shallow region below the surface with few in the deeper region. The lines in Fig. 4 are the fitted curves by VEPFIT under the assumption of Gaussian depth profile of trapping sites, but the best fit is achieved when the distance of Gaussian center from the surface is negative, which means the actual depth profile is estimated to be monotonically decreasing from the surface. As interstitials can easily escape from the matrix to the surface in the shallow region (like the so-called 'thin-foil' condition), more vacancy clusters can survive recombination compared to the deep region (bulk condition).

3.4. Nickel ion irradiation at 773 K

Fig. 5 shows S–E curves for nickel irradiated at 773 K. For 0.1 dpa, the S–E curve is almost identical to that of the irradiated at 673 K. The S–E curves go up with increasing irradiation dose, and large increase in S parameter is at 10–20 keV positrons. An assumption of Gaussian defect population reproduces the S–E curve quite well, and the depth

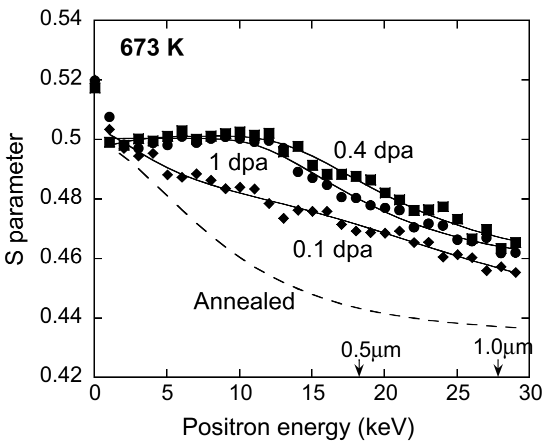

Fig. 4. Dependence of S parameter on implanted positron energy for ion-irradiated nickel at 673 K to 0.1 dpa (◆), 0.4 dpa (■) and 1 dpa (●). Lines are the fitted curves by VEPFIT [14].

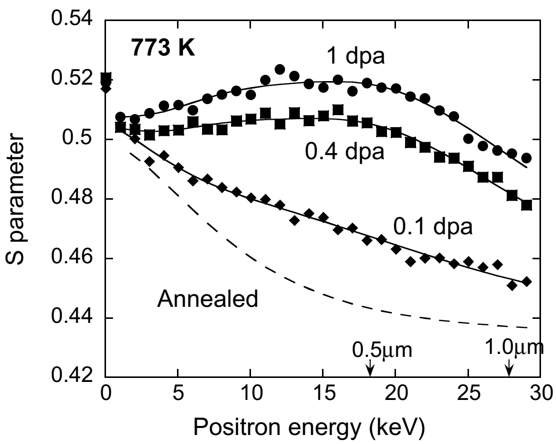

Fig. 5. Dependence of S parameter on implanted positron energy for ion-irradiated nickel at 773 K to 0.1 dpa (◆), 0.4 dpa (■) and 1 dpa (●). Lines are the fitted curves by VEPFIT [14].

of Gaussian center is estimated to lie around 550 nm. As the maximum value in the curve for 0.4 and 1 dpa significantly exceeds the constant value for the cold-rolled ($S = 0.48$), the trapping sites have considerable open volume. At 773 K, where SFTs and v-loops are thermally unstable, it is natural that the large S parameter is attributed to cavity formation. This explanation is consistent with the previous ion irradiation studies in nickel at this temperature by Packan et al. [5] and Ishino et al. [6,7]. The dose dependence of the S–E curves indicates that cavity formation begins at around 0.1 dpa.

3.5. Perspective on in situ positron beam experiment

Compared to the established method of in situ TEM observation of radiation damage, in situ positron beam measurement of radiation damage is a challenging and difficult method to obtain meaningful data. Although the positron beam has an advantage of high sensitivity, even to monovacancies, time resolution is 4–5 order of magnitude worse than that of in situ TEM observation (typically 1/30 s; depending on the video frame rate) because it takes at least several minutes to collect a statistically sufficient number of counts from positron irradiation. This requires a relatively slow dose rate to follow the dose-dependent transient. From the previous section, cavity nucleation is likely to occur around 0.1 dpa at 773 K. In order to follow the condition in detail, a lower dose rate is desirable for an in situ positron beam experiment. It may be effective to use sequential irradiations in which irradiation at higher dose rate is employed for microstructural evolution and then irradiation at lower dose rate is carried out for an in situ positron beam experiment.

4. Conclusions

Formation processes of vacancy-type defects induced by ion irradiation up to 773 K in nickel were investigated with a variable-energy positron beam Doppler broadening technique in order to find the appropriate experimental condition for in situ positron beam experiments. Irradiations at 573 K and lower temperature were not sufficient for cavity formation. At 673 K and 773 K, vacancy cluster formation was indicated by a large increase in the S parameter, but it was limited to a shallow region at 673 K. The best experimental condition for investigation of cavity nucleation process by in situ positron beam Doppler experiments is suggested to be around 0.1–0.4 dpa at 773 K, and such experiments will be done in the future.

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