Ni ion damage structures and hardness changes in austenitic stainless steels and their model alloys

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Abstract. Defect structures of austenitic stainless steels and their model alloys after 6 MeV Ni ion irradiation were studied using positron annihilation lifetime measurements and Vickers hardness obtained from micro-hardness measurements. After irradiation at room temperature, a long lifetime of 150–180 ps was obtained and the Vickers hardness increased with increasing the irradiation dose. After irradiation at 573 K, all the alloys exhibited lifetime spectra that were not decomposed into two components. Their hardness also increased with the irradiation dose, but the enhancement was small compared with that at room temperature. Defects introduced by the ion irradiation could be detected near the surface using a Na-22 source. The hardness is correlated with the density and size of the defect clusters.

1. Introduction

To study the irradiation effects on nuclear materials, ions are commonly used as incident particles instead of neutrons (simulation irradiation). Ion irradiation provides many advantages such as high damage rates and easy setting of irradiation conditions, e.g., temperatures and stresses. One of the disadvantages is the localized formation of irradiation damage in the specimens because of a determined ion range. In the case of self ion irradiation of structural materials of nuclear systems such as steels with several MeV, the ion range is the order of microns and it is necessary to extract the bulk properties of the irradiation effects from the property changes at a thin area near the surface. Micro-hardness testing is one of the most widely used methods to measure mechanical properties of thin layers. This technique measures the penetration under different indentation loads to obtain an indentation curve. From these measured data, the mechanical properties such as hardness and elastic modulus can be evaluated [1].

Positron annihilation spectroscopy is a very powerful technique to detect open volume defects in materials because of their ability to trap positrons. Positron annihilation lifetime measurements and coincidence Doppler broadening measurements using Na-22 as a positron source have been adopted to study irradiation damage in bulk materials. Slow positron beam is the best way to detect irradiation defects in the near-surface region of ion-irradiated materials [2]. We have demonstrated, however, that it is possible to detect irradiation damages of 6 MeV Fe ion irradiated ferritic stainless steels by conventional sandwich configuration on positron source [3]. In the case of high ion irradiation, even if
the damages are localized near the surface area, some of the positrons (the probability for the positrons to stop within ~3μm in depth in Fe is approximately 25%) are trapped there, thus the lifetime of the defects can be measured.

Austenitic stainless steels are important nuclear materials. Owing to the irradiation-induced swelling, they can experience, however, dimensional changes at very high dose neutron irradiation. It is known that the defect evolution strongly depends on the alloying elements [4-6]. In our previous studies, irradiation damages by electron irradiation and neutron irradiation were induced below 10⁻² dpa and below 0.2 dpa, respectively, and the defect structural evolution before void swelling was studied [5-6]. In this study, defect structural evolutions of austenitic stainless steels and their model alloys were investigated at higher irradiation dose by Ni ion irradiation. Damaged structures and changes in the mechanical properties were investigated by micro-hardness tester and positron annihilation lifetime spectroscopy with Na-22.

2. Experimental
The test materials used in this study were Ni of 99.99% in purity, four model alloys, and four commercial alloys, as listed in Table 1. Ti-added modified SUS316SS (J) is an austenitic stainless steel with high Ni concentration to reduce void swelling, and designed for the first wall component of near term fusion devices [7-8]. The concentrations of the elements in the four model alloys, A, B, C, and D, were determined by the concentrations of the alloy constituents in J. For this experiment, thin disk specimens with the dimensions of 0.1 mm in thickness and 3 mm in diameter were machined and annealed at 1323 K and 10⁻⁴ Pa for 1 h, followed by cooling to room temperature. The cooling rate was sufficiently rapid, so that no precipitates were observed by transmission electron microscopy.

Specimens were irradiated by Ni ions (6 MeV) with an accelerator of Quantum Science and Engineering Center, Kyoto University at room temperature and at 573 K. The irradiation dose was 0.037, 0.37, 1.85, and 6.8 dpa (defect peak position). Figure 1 shows the implantation profiles of the Ni ions in Ni and the corresponding damage distributions calculated using the SRIM code [9]. The depth of the peak position of the defects is about 1.8 μm. We chose Ni as its crystal structure is the same and the lattice constant is almost the same as those of the austenitic stainless steels.

### Table 1. Chemical compositions of the specimens.

| Specimens | Composition |
|-----------|-------------|
| Ni        | Pure Ni (99.99%) |
| A         | Fe-16.1Cr-17.0Ni |
| B         | Fe-15.4Cr-15.9Ni-2.68Mo-1.89Mn |
| C         | Fe-15.3Cr-15.8Ni-2.66Mo-1.88Mn-0.53Si |
| D         | Fe-15.3Cr-15.8Ni-2.66Mo-1.88Mn-0.53Si-0.24Ti |
| J         | Ti added modified SUS316SS: Fe-15.3Cr-15.8Ni-2.66Mo-1.88Mn-0.53Si-0.24Ti-0.055C-0.024P |
| E         | SUS316L SS: Fe-17.4Cr-12.05Ni-2.08Mo-0.85Mn-0.47Si-0.019C-0.027P-0.001S |
| F         | SUS316SS: Fe-19.2Cr-12.58Ni-2.26Mo-1.82Mn-0.39Si-0.05C-0.019P-0.004S |
| G         | SUS304SS: Fe-18.1Cr-9.08Ni-0.85Mn-0.48Si-0.05C-0.027P-0.0025S |
After irradiation, the damage structures were studied using positron annihilation lifetime measurements. The measurements were performed at room temperature using the fast-fast coincidence system, whose lifetime resolution was 190 ps (full width at half maximum, FWHM). The lifetime spectra were collected with a total count of about $10^6$. The spectra were analyzed using the PALSfit program [10]. The total amount of residual defects can be derived from the mean positron lifetime. The amount of positron annihilation that occurs in the matrix can be determined from the short lifetime component. The size of the vacancy-type defects can be derived from the long lifetime component, and their density can be obtained from the relative intensity of the long lifetime component. The calculated positron annihilation lifetimes for single-vacancy and 13-vacancy clusters in Ni are 169 and 341 ps, respectively [11]. When the vacancy clusters grow, their positron lifetime becomes longer. A series of hardness tests was also performed by a micro-hardness tester (DUH201, Shimadzu Ltd.). The diamond indenter had a pyramidal shape with angles of 22° on each side with respect to the horizontal plane. The loading rate was 13.24 mN/s and the holding time was 5 s. The Vickers hardness was obtained by the following equation [12]:

$$H_v = 189.1 \left( \frac{P}{d^2} \right),$$

(1)

where $P$ is the load in mN ($P = 10$ mN) and $d$ is the average length of the diagonal left by the indenter in μm.

3. Results and discussion

Figures 2–4 show the positron annihilation lifetime of austenitic stainless steels and their model alloys irradiated with Ni ions at room temperature and 0.037, 0.37, and 1.85 dpa, respectively. Before Ni ion irradiation, the lifetimes of Ni and the alloys were between 106 and 108 ps. A longer lifetime of approximately 150–200 ps was obtained after irradiation. The formation of large vacancy clusters was not detected. These tendencies closely resemble the results of neutron and electron irradiation experiments [5,6]. This is due to the fact that vacancies in these alloys are not mobile at room temperature (vacancy migration energies in Ni and austenitic stainless steels are 1.25 and 1.1–1.4 eV, respectively [13,14]). The lifetimes of commercial stainless steels were a little bit shorter than their model alloys. Figures 5–7 show the Vickers hardness. In all the specimens, the Vickers hardness increased with increasing the irradiation dose, owing to the fact that the amount of defects increases with the irradiation dose.
Figure 2. Positron annihilation lifetime of specimens irradiated with Ni ions at room temperature and 0.037 dpa.

Figure 3. Positron annihilation lifetime of specimens irradiated with Ni ions at room temperature and 0.37 dpa.

Figure 4. Positron annihilation lifetime of specimens irradiated with Ni ions at room temperature and 1.85 dpa.

Figure 5. Vickers hardness of specimens irradiated with Ni ions at room temperature and 0.037 dpa.
Figures 8–10 show the positron annihilation lifetime of austenitic stainless steels and their model alloys irradiated with Ni ions at 573 K and 0.037, 0.37, and 6.8 dpa, respectively. Figures 11–12 show the Vickers hardness change of austenitic stainless steels and their model alloys. At 0.037 dpa, the mean lifetime increased after irradiation, but the lifetime spectra were not decomposed into two components. This is due to the formation of interstitial type dislocation loops or large stacking fault tetrahedra by the movement of vacancies. At 0.37 and 6.8 dpa, small vacancy clusters were detected only in pure Ni. In other alloys, the lifetime spectra were not decomposed into two components, and the large vacancy clusters could not be detected. These results are also identical to those obtained by neutron and electron irradiation experiments at 573 K [5, 6]. Even at the high irradiation dose of 6.8 dpa, no voids were detected. For the formation of stable voids, an increase of the dislocation density
by the growth of interstitial type dislocation loops is required (dislocation bias theory for void swelling [15]). The void swelling peak by ion irradiation is higher than that by neutron irradiation because of the high damage rate induced by ion irradiation [16]. In the present irradiation conditions, a temperature of 573 K may be too low for void swelling. Furthermore, after irradiation at 573 K, the hardness increased with the irradiation dose, but the change was small compared with that observed after irradiation at room temperature because of a lower density of the defect clusters. The low defect cluster density prevented the decomposition of the positron annihilation lifetime spectra of Ni-ion-irradiated austenitic stainless steels and their model alloys into two components.

**Figure 10.** Positron annihilation lifetime of specimens irradiated with Ni ions at 573 K and 6.8 dpa.

**Figure 11.** Vickers hardness of specimens irradiated with Ni ions at 573 K and 0.037 dpa.

**Figure 12.** Vickers hardness of specimens irradiated with Ni ions at 573 K to 0.37 dpa.
4. Summary
Positron annihilation lifetime measurements and Vickers hardness tests of Ni-ion-irradiated stainless steels and their model alloys were performed. After irradiation at room temperature and at 573 K, the hardness increased with increasing the irradiation dose. However, the observed enhancement at 573 K was less intense. After irradiation at room temperature, a long lifetime of 150–180 ps was obtained, while after irradiation at 573 K the lifetime spectra were not decomposed into two components except for the Ni spectrum. These results are the same as those obtained by neutron and electron irradiation experiments [5,6].

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