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Radiation damage and radiation-induced segregation in single crystal stainless steel by RBS and PIXE channeling

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Abstract

The radiation damage and the radiation-induced segregation (RIS) in type 304 austenitic stainless steel single crystal were investigated with 1.6 MeV He ion irradiation up to 2.5×10^{17} cm^{-2} at RT by using RBS and PIXE with channeling conditions (RBS-C and PIXE-C). By the RBS-C experiments, it is found that radiation-induced damage for (1 1 0) orientation increased more slowly than those for (1 0 0) and (1 1 1) orientations with He ion dose. By the PIXE-C experiments, it is found that the Si atoms are displaced from the lattice sites and segregate significantly to the surfaces of (1 0 0), (1 1 0) and (1 1 1) orientations and the S atoms also seem to be enriched slightly in the (1 0 0) and (1 1 0) surfaces during He ion irradiation.

1. Introduction

The radiation effect of stainless steels has been studied extensively as one of the most common complex alloys and one of the most useful materials for nuclear reactor devices. The effect of neutron irradiation is a very important factor to make use of the materials for fusion reactors, for it may cause the degradation on the strength of materials after prolonged service time. Especially, radiation-induced segregation (RIS) is one of the most stimulating topics in the development of nuclear reactor materials [1–11]. To avoid the activation of samples during long period experiments, ion beam irradiation is often adopted instead of neutron irradiation.

Most of the investigations were performed using polycrystalline stainless steels for the purpose of practical use. Recently it has been shown that single crystals are advantageous to examine the orientation dependence on mechanical and electrical properties [12–14]. Moreover, it has been shown that the microscopic investigation and the microstructural evolution for single crystal stainless steels by Rutherford backscattering spectroscopy and by particle induced X-ray emission under channeling conditions (RBS-C and PIXE-C) are powerful tools for the study on depth profiles of implants or impurities and transformed layers, and radiation-induced segregation [15–19].

In this paper, we have investigated the radiation damage and the radiation-induced segregation in single crystals of type 304 austenitic stainless steel (SUS 304) grown by the modified Bridgman method. The effect of 1.6 MeV He ion irradiation on the SUS 304 specimens of the (1 0 0), (1 1 0) and (1 1 1) orientations was examined by means of RBS-C and PIXE-C measurements.

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2. Experimental

2.1. Sample preparation

Single crystal specimens used in this experiment were grown by the modified Bridgman method using a commercial purity rod of type 304 stainless steel (SUS 304) at Himeji Institute of Technology. The original rod was moved at a falling rate of 5.6×10^{-4} cm s^{-1} in purified argon ambient at 1500°C and then homogenized at 1300°C for 4.0 h [13,18,19]. Table 1 shows the chemical compositions of a polycrystal and a single crystal, which are satisfactorily similar to each other. From a cylindrical single crystal of the fcc structure (9 mm in diameter and 600 mm in length), the specimens with a thickness of 1 mm were prepared for the (1 0 0), (1 1 0) and (1 1 1) orientations by spark cutting, mechanical polishing and electropolishing. The orientation of each specimen was determined by the back-reflection Laue method. Then, all specimens encapsuled in quartz tubes in a vacuum were annealed at 1100°C for 1.0 h, and subsequently quenched in water (solution annealing). The quality of the single crystals has been examined and it has been shown that the solution annealing process is absolutely necessary not only for the production of polycrystals but also for single crystals [18].

2.2. RBS-C and PIXE-C measurements

Single crystalline specimens of (1 0 0), (1 1 0) and (1 1 1) oriented SUS 304 were bombarded with 1.6 MeV $^4He^+$ ions incident at 4° to the surface normal at room temperature to avoid the channeling effect in the radiation damage and the radiation-induced segregation. The He ion irradiation was performed at a 2 MV van de Graaff accelerator facility of the Osaka National Research Institute (ONRI) in Ikeda. The He ion doses ranged from 1.3×10^{16} to 2.5×10^{17} cm^{-2}, corresponding to 0.11–0.21 dpa. At the same time, the radiation damage at the surface region and the behavior of component elements in SUS 304 single crystals were determined by RBS-C and PIXE-C measurements using the same 1.6 MeV $^4He^+$ beam at ONRI.

The backscattered He ions were detected with a solid state particle detector (EGG&G Ortec BA-13-25-100 or BA-14-100-100) set at 165° to the incident projectiles. The X-ray spectra were measured with a Si(Li) detector with 15 μm Be window (Horiba SLP-914, 170 eV FWHM at 5.9 keV) at 135°. A polyester foil of 7 μm thickness was used for protecting from the radiation damage of the Be window by the backscattered He ions.

3. Results and discussion

Fig. 1 shows random and aligned spectra of backscattered helium ions ($\Theta = 165°$) from the (1 1 0) oriented SUS 304 crystal for an incident energy of 1.6 MeV where the RBS edges of Cr, Fe and Ni are 1.18, 1.21 and 1.22 MeV, respectively. In these experiments, we could not distinguish these elements from the RBS-C spectra. These RBS-C measurements show that the specimen crystal is nearly perfect, for the value of χ_{min} at the (1 1 0) surface is 3.5% and very close to the value of 2.0% obtained by the calculation. Here the minimum yield, χ_{min}, is defined as the ratio of the yield at aligned orientation to that at random one. PIXE-C measurements were also performed simultaneously, in order to distinguish each element in SUS 304 crystal. As shown in Fig. 2, PIXE-C enables the detection of characteristic

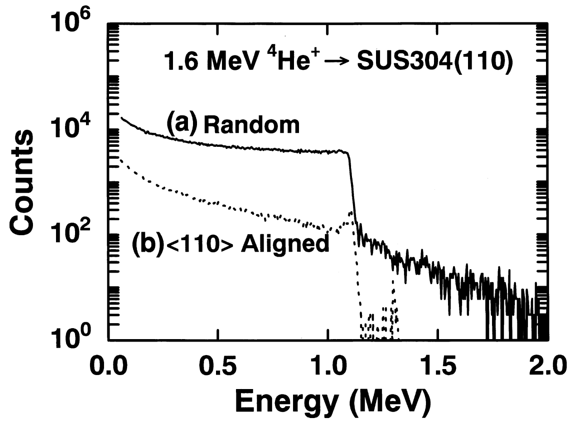

Fig. 1. Rutherford backscattering spectra of 1.6 MeV $^4He^+$ ions at 165° from unimplanted SUS 304 (1 1 0) single crystal; (a) aligned, (b) random direction.

Table 1
Nominal alloy composition (wt%) of SUS 304

SUS 304	C	Si	Mn	P	S	Ni	Cr	Fe
Poly	0.05	0.04	1.42	0.034	0.001	9.18	18.29	Balance
Single [a]	0.05	0.80	0.68	0.049	0.002	9.50	18.05	Balance
Single [b]	0.03	0.64	0.85	0.023	0.001	9.25	17.80	Balance

[a] The upper part.
[b] The lower part.

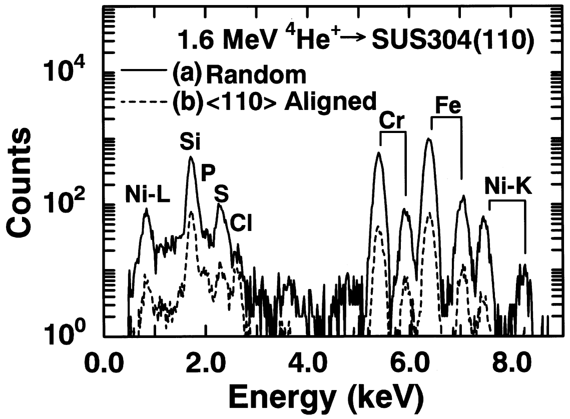

Fig. 2. PIXE spectra of 1.6 MeV $^4He^+$ ions from unimplanted SUS 304 (1 1 0) single crystal; (a) aligned, (b) random direction. They were measured simultaneously with RBS spectra shown in Fig. 1.

K X-rays of Si and S and L X-rays of Ni in addition to Cr, Fe and Ni K X-rays. From the angular dependence of the K X-ray yields from these atoms for the 1.6 MeV He^+ beam along the axes $\langle 100\rangle$, $\langle 110\rangle$ and $\langle 111\rangle$, these elements were found to occupy the fcc lattice sites in SUS 304 single crystal.

In order to investigate the atomic displacement processes in more detail, the dose dependence of χ_{min} for RBS-C and PIXE-C was measured. An appropriate approximation to the concentration of displaced atoms at the surface, $N_d(0)$, is taking the difference between aligned and virgin spectra at the surface region. The disorder concentration $N_d(0)$ is given by

$$N_d(0) = N[\chi(0) - \chi_v(0)]/[1 - \chi_v(0)], \quad (1)$$

where N is the number of atoms per cubic centimeter and χ, the ratio of aligned to random yields at the energy region of interest. For a virgin crystal, $\chi_v(0)$ is equal to the minimum yield χ_{min} [20]. Fig. 3 shows the dose dependence of the displacement atom ratio (DAR) $N_d(0)/N$ for the (1 0 0), (1 1 0) and (1 1 1) orientations. The values of DAR for (1 1 0) increases linearly and reaches 0.1 at 2.5×10^{17} cm^{-2}. Those for (1 0 0) and (1 1 1) increases more rapidly at the initial stage and reaches 0.15 and 0.14 at the same dose, respectively. The dose dependence for (1 1 0) is fairly different from the other two orientations. It suggested that the production of the lattice defects depends on the orientation. However, we cannot conclude in this stage if this difference is associated with the specified orientation of atomic defects.

The dose dependence of the PIXE-C spectra shows that X-ray yields for the aligned orientation increase similarly for all elements. This means that all elements including the impurity atoms are displaced more or less from the fcc lattice positions by the implanted He atoms. In order to investigate the atomic displacement process for each element, the dose dependence of χ_{min} of PIXE-C for (1 0 0), (1 1 0) and (1 1 1) oriented crystals was also measured. As an example, the (1 1 0) oriented specimen is given in Fig. 4. For all elements, χ_{min} increases linearly up to a dose of 1.3×10^{17} cm^{-2}, for S, Cr, Fe and Ni atoms it increases more gradually, and for Si atoms it still increases linearly up to a dose of 2.5×10^{17} cm^{-2}. It may be considered that Si atoms are displaced in perpendicular to the $\langle 110\rangle$ direction and not influenced so much by the recovery process of host lattice. In the other orientations (1 0 0) and (1 1 1), the dose dependence of χ_{min} is very similar to each other.

Fig. 5 shows a part of the PIXE-C spectra from the random orientation of SUS 304 single crystal with the (1 1 0) surface before and after 1.6 MeV He ion

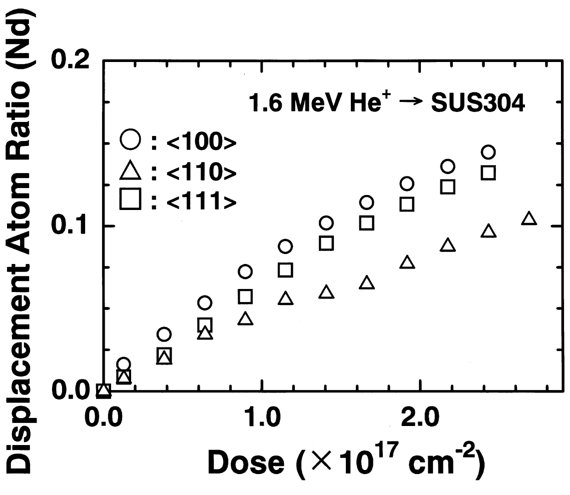

Fig. 3. Dose dependence of the DAR in SUS 304 single crystal for the (1 0 0), (1 1 0) and (1 1 1) oriented surfaces irradiated with 1.6 MeV $^4He^+$ ions from the RBS-C measurements.

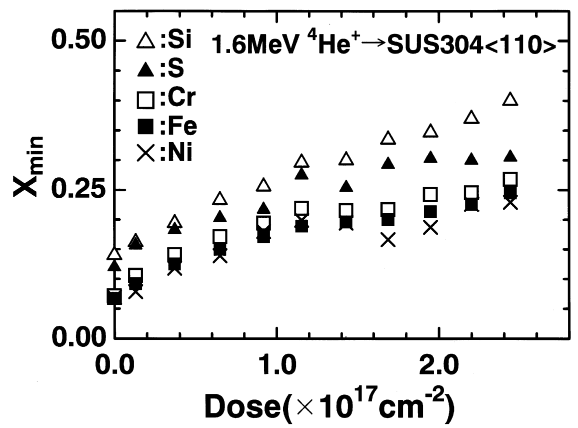

Fig. 4. Dose dependence of the χ_{min} of each element for the (1 1 0) oriented surface in the PIXE measurements for SUS 304 single crystal.

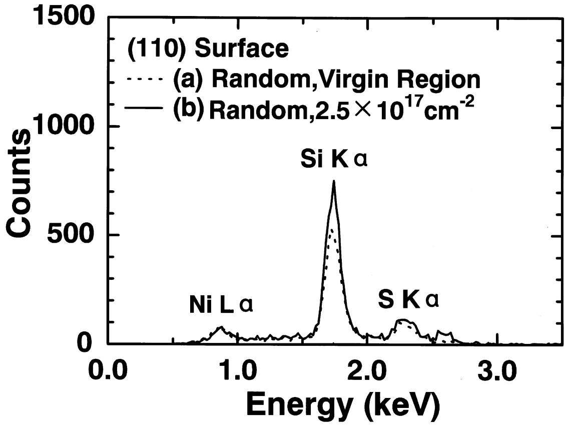

Fig. 5. PIXE spectra of 1.6 MeV $^4He^+$ ions from the random orientation of SUS 304 single crystal oriented to the (1 1 0) surface before and after 2.5×10^{17} cm^{-2} He irradiation; (a) dashed curve indicates before irradiation and (b) solid curve, after irradiation, respectively.

irradiation of 2.5×10^{17} cm^{-2}. The PIXE-C spectra were also measured for the other surfaces of (1 0 0) and (1 1 1) directions. K X-ray spectra of Cr, Fe and Ni are confirmed to be equal in all six spectra, respectively. On the other hand, the K X-ray yields from the Si atoms after 2.5×10^{17} cm^{-2} He ion irradiation increase by 20% for (1 0 0) and (1 1 0), and by 15% for (1 1 1), comparing with that of virgin specimens. Those from the S atoms increased only by a few % for the (1 0 0) and (1 1 0) facing specimens. It is necessary to examine in more detail the changes in the K X-ray yields observed for the S atoms. This enrichment of Si and S atoms to the surface of SUS 304 crystal during He ion irradiation is due to RIS of Si and S atoms to the (1 0 0), (1 1 0) or (1 1 1) surface. Especially, a large amount of RIS of Si atoms was found in the (1 0 0) and (1 1 0) surfaces. This result does not always correspond to the results of orientation dependence observed for radiation-induced defects from RBS-C and PIXE-C measurements. However, it should be noted that the value of χ_{min} for Si atoms in the (1 1 0) surface increases more than other atoms up to a dose of 2.5×10^{17} cm^{-2}. Now, it is most important that the orientation dependence of RIS was clearly observed by PIXE-C measurements. In the study of the RIS process, heavy ion, electron and neutron irradiation are commonly used at elevated temperatures and up to high doses. It is the first time that the phenomena of RIS is observed in stainless steels at room temperature ion irradiation. The light impurity elements, such as Si and S are very sensitive to the ion irradiation, but the heavy elements such as Cr, Fe and Ni are not so sensitive to the room temperature ion irradiation. The study of RIS for the heavy elements such as Cr, Fe and Ni may require high temperature irradiation. The RBS-C and PIXE-C method proved to be a very sensitive technique to investigate the radiation damage and RIS mechanisms in single crystal stainless steels.

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