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Annealing Behavior of Defect Structures in Ni Irradiated by High Energy H and He Ions

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Abstract

Isochronal annealing experiments for H and He ion irradiated Ni were carried out to study the interactions between gas atoms and vacancy clusters. The accelerating energy of H and He ions was 1.0 MeV and 3.3 MeV, respectively. The total dose was 1.0×10^{17} H ions/cm^2 (0.2 dpa) and 9.6×10^{15} He ions/cm^2 (0.3 dpa), and the irradiation temperature was 300°C. In order to study the recovery behavior of the defect structures, positron annihilation lifetime and coincidence doppler broadening (CDB) measurements were carried out after irradiation and annealing for 1 hour up to 950°C. The microvoids were detected after the H and He ion irradiation. The characteristic difference mainly appeared in the annealing behavior of the defect structures. The results of the positron lifetime and CDB measurements after annealing indicated that the recovery of the defect structures in Ni irradiated by H ions finished completely at 600°C, while that in Ni irradiated by He ions finished at 950°C. In addition, the microvoids formed by the He ion irradiation grew after annealing at 750°C. The stability of the vacancy clusters, H-vacancy complexes, and He-vacancy complexes is discussed from these results.

Keywords:

nickel, hydrogen, helium, ion irradiation, positron spectroscopy, microvoid, gas-vacancy complex

1. Introduction

The effect of H and He atoms on the development of defect structures is an important problem for materials used in fusion reactors, because the production rate of H and He atoms generated by (n,p) and (n,α) nuclear reactions is much higher than in fission reactors, and, in addition the materials used in the fusion reactors are directly damaged by the irradiation of gas atoms with acceleration energy at the MeV level. It is particularly well known that these atoms promote the growth of cavities [1-2] and dislocation loops [3-5]. The gas atoms induce void swelling and radiation embrittlement even at low solubility. In order to study the diffusion and trapping of H and He atoms to defect clusters in pure metal, several studies on the thermal release [6-8] and cavity growth behaviors [9-10] of implanted H and He in Ni have been performed. However, the thermal stability of vacancy clusters by the presence of H and He atoms has not been thoroughly studied. Above all, experimental studies on the microvoid formation in Ni irradiated by H and He ions with high energy for imitating the fusion environment has scarcely been performed.

In the present study, the annealing behaviors of defect structures in Ni irradiated by H and He ions were investigated in order to study the thermal stability of defect structures which were induced by the irradiation of the H and He ions. After the irradiation experiments, isochronal annealing experiments were performed. To

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investigate the formation of vacancy clusters, positron annihilation lifetime and coincidence doppler broadening (CDB) measurements were performed. The positron annihilation technique is effective for investigating microvoid formation [11-13]. The lifetime measurement was used for detecting the size of the microvoids in metals. The CDB measurement, which determines the longitudinal momentum distribution of electrons annihilated with positrons, was used to detect the total amount of residual defects. The CDB measurement is a remarkable technique, which has the great advantage of reducing disturbances from the background spectrum on the higher energy side of the annihilation peak (511 keV) [14-18]. The stability of the vacancy clusters, H-vacancy complexes, and He-vacancy complexes was discussed from the recovery behavior of the defect structures after annealing.

2. Experimental Procedures

High-purity Ni of 99.99% supplied by Johnson & Matthey Chemicals Ltd. was employed for the experiments. The Ni specimens were annealed in a high vacuum below 1.0×10^{-4} Pa at 900°C. The irradiation experiments were carried out by using an accelerator in the Institute for Materials Research, Tohoku University. The total dose was 1.0×10^{17} H ions/cm^2 and 9.6×10^{15} He ions/cm^2, respectively. In order to align the damage and gas-atom range for both irradiations, the acceleration energy for the H and He ions was chosen to be 1.0 MeV and 3.3 MeV, respectively. The damage and gas-atom distributions were calculated by the TRIM code with a displacement energy of 24 eV for Ni. The depth of damage and gas-atom peak was about 6 μm from the surface of the specimen, and the effective gas-containing zone was about 1 μm in the vicinity of the gas peak for both irradiations. The amounts of effective damage and gas-concentration were estimated as the average of the damage and gas-atom distribution in the region of a full-width at half maximum. In the case of the H ion irradiation, the total damage was 0.2 dpa, the damage rate was 2.4×10^{-5} dpa/sec, and the amount of H concentration was about 2 at%. In the case of the He ion irradiation, the total damage was 0.3 dpa, the damage rate was 1.8×10^{-6} dpa/sec, and the amount of He concentration was about 0.3 at%. The irradiation temperature was 300°C.

The system for the positron annihilation lifetime measurement in this study was the conventional fast-fast circuit with two BaF_2 scintillators, and a ^{22}Na source of positrons was used. The time resolution of this system was about 220 psec. The lifetime was determined from the analyses of the annihilation spectra by the positron resolution program. In order to measure the CDB spectra, two Ge-detectors with almost the same energy resolution were used. The overall energy resolution of the two Ge-detectors was 1.6 keV, which was defined by a full-width at half maximum of the 661.6 keV γ-ray generated by the ^{137}Cs standard radiation source.

The isochronal annealing for Ni irradiated by the H and He ions was carried out for 1 hour by a 50°C increment from 300°C. The positron annihilation lifetime and CDB measurements were performed after each annealing process.

3. Results

3.1 Positron lifetime measurements

Figures 1 and 2 show the results of the positron annihilation long lifetime and the mean lifetime for the

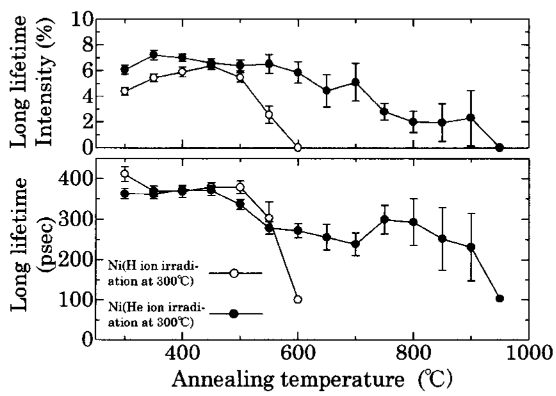

Fig. 1 The isochronal annealing behavior of a long lifetime component and the intensity in Ni irradiated by H and He ions at 300°C.

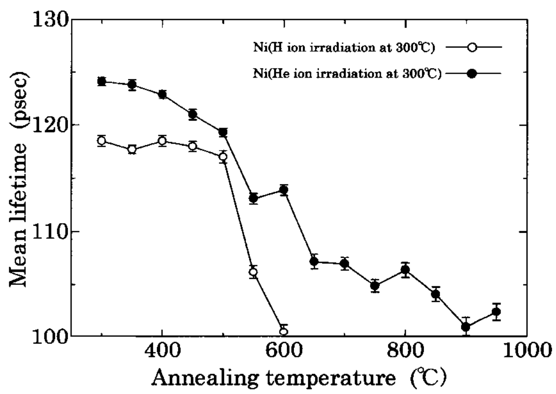

Fig. 2 The isochronal annealing behavior of a mean lifetime component in Ni irradiated by H and He ions at 300°C.

Ni irradiated by H and H ions at 300°C as a function of the annealing temperature. The long lifetime corresponds to the size of the vacancy clusters, its intensity corresponds to the density of the vacancy clusters, mainly microvoids, and the mean lifetime corresponds to the total amount of defects. As can be seen from these results, the formation of microvoids was observed after the H and He ion irradiation. However, the difference between the H and He ion irradiation was mainly marked in the annealing behavior of the long and mean lifetimes. In the case of the H ion irradiation, the long lifetime component disappeared at 600°C, while in the case of the He ion irradiation the long lifetime component disappeared at 950°C. This suggests that the recovery of microvoids formed by the H ion irradiation occurred completely at 600°C, whereas that by the He ion irradiation occurred at 950°C. Another feature was a slight increase of the long lifetime after annealing above 750°C in the case of the He ion irradiation. This means that the vacancy clusters grew above this temperature.

3.2 Coincidence doppler broadening (CDB) measurements

Figure 3 shows the CDB spectra of un-irradiated Ni and H ion irradiated Ni at 300°C (0.2 dpa). P_l is the longitudinal momentum of an electron annihilated with a positron. The mc of the x-axis unit means the momentum, where m is the electron rest mass and c is the light speed. The unit of the y-axis is the count ratio for each momentum, which is normalized by total counts. A remarkable feature in this result is that the count ratio at the low momentum region for the H ion irradiated Ni was larger than that before irradiation. Positrons trapped at open-volume defects such as vacancies are mainly annihilate with low-momentum electrons, i.e., conduction and valence electrons [19]. The increase of the count ratio at the low momentum region in the CDB spectra signifies that open-volume defects exist in the specimen. This low momentum region is called the S-region, and the S-region of Ni at our CDB measurement system was less than 5.0×10^{-3} mc. The ratio of the S-region to total counts is called the S-parameter, which is used for measuring a total amount of vacancy-type defects. For example, if the S-parameter ratio of high energy particle irradiated specimens to un-irradiated specimens is larger than 1.0, it is considered that radiation induced defects will be formed in the irradiated specimen.

In Fig. 4, the S-parameter ratio of the Ni irradiated by H and He ions at 300°C to the un-irradiated

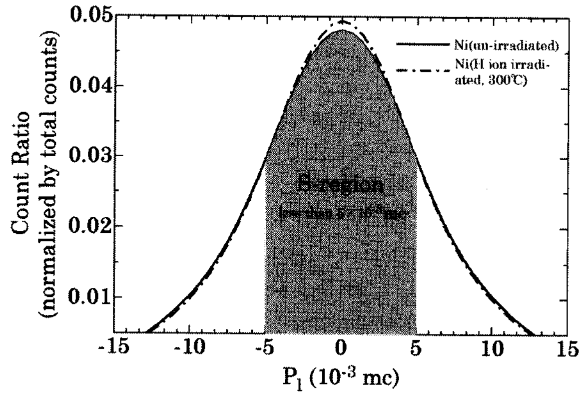

Fig. 3 Comparison of spectra from the CDB measurement between un-irradiated Ni and H ion irradiated Ni at 300°C.

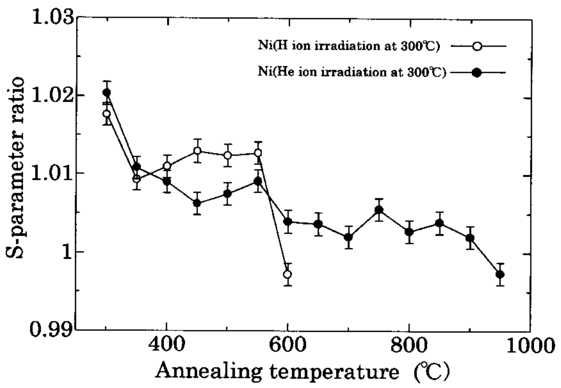

Fig. 4 The isochronal annealing behavior of the S-parameter ratio in Ni irradiated by H and He ions at 300°C.

specimen is plotted as a function of the annealing temperature. Fig. 4 implies the same temperature-dependence as the positron lifetime measurement. The S-parameter ratio for the H ion irradiation was less than 1.0 after annealing at 600°C, while that for the He ion irradiation was less that 1.0 at 950°C. It was also clear from the result of the CDB measurement that the microvoids formed by the H ion irradiation decomposed below 600°C, whereas those formed by the He ion irradiation did not decompose until 950°C.

4. Discussion

The results of the positron annihilation lifetime and CDB measurements indicate the difference in thermal stability between the H and He-vacancy clusters, i.e., He-vacancy clusters were much more stable than H-vacancy clusters. According to Niwase *et al.* [9], the cavities in Ni formed by the 20 keV D ion irradiation at

360°C disappeared after annealing at 625°C. Though there is a difference between the D and H ion, it can be considered that the same recovery of defect clusters occurs below 600°C in H ion irradiated Ni. In the case of the H atom in Ni, the migration activation energy of the H atom is 0.41 eV [20], and the binding energy of the H-vacancy complex is 0.57 eV [21]. The de-trapping energy, a sum of the migration energy and binding energy, of the H-vacancy complex in Ni is about 1 eV. Therefore, H atoms can easily escape from vacancies or small vacancy clusters by annealing at 350°C. It can also be explained from the thermal desorption behavior of H atoms in Ni. Tanabe *et al.* [6] have reported that the thermal desorption of D atoms from Ni finished over 350°C in the case of D ion implantation into Ni by the acceleration energy of under 30 keV with fluences of 4.0×10^{14} up to 9.0×10^{17} ions/cm^2 at room temperature (RT). Therefore, the recovery behavior of vacancy clusters was not influenced by H atoms above 350°C. It was concluded that in the case of H ion irradiation, the thermal decomposition of vacancy clusters occurred between 350°C and 600°C.

However, according to Yamauchi *et al.* [7], the thermal release peak of He atoms from Ni was above 900°C in the case of He ion implantation into Ni by the acceleration energy of 20 keV with a fluence of 1.0×10^{17} ions/cm^2 at RT. This is consistent with the present results, which showed that the long lifetime component disappeared and the S-parameter ratio was under 1.0 after annealing at 950°C. Regarding the He bubbles, the growth of the bubbles by heating above 750°C was observed in Ni implanted by 20 keV He ions with a fluence of 1.8×10^{17} ions/cm^2 at RT [22]. The present positron lifetime measurements supported this result. The size of the He-vacancy clusters increased above 750°C, while the density of the clusters suddenly decreased. This can be explained from the de-trapping energy of the He-vacancy complex. According to Reed [23], the migration activation energy of He is 0.08 eV, and the binding energy of the He-vacancy complex is 3.16 eV. Therefore, the de-trapping energy of the He-vacancy complex in Ni is 3.24 eV. Once He atoms can escape from vacancies or small vacancy clusters at a high temperature, He atoms will easily migrate in crystal lattice due to the low migration energy. Since the migration activation energy of vacancies in Ni is 1.2 eV [24], free vacancies are able to migrate easily above 750°C. It suggests that the He atoms started to escape from vacancies or small vacancy clusters above 750°C, and the growth of He-vacancy clusters occurred by the coalescence between free vacancies and vacancy clusters. After annealing at 950°C, the desorption of He came to end, and at the same time, the decomposition of the vacancy clusters fully occurred.

5. Conclusions

Based on the positron annihilation measurements for Ni irradiated by H and He ions at 300°C after isochronal annealing, the following conclusions were drawn. In the case of the H ion irradiation, the vacancy clusters disappeared after annealing at 600°C, which indicated that the desorption of H atoms might occur after annealing at 350°C, and that the vacancy clusters decomposed between 350°C and 600°C. The recovery of the vacancy clusters was not influenced by H atoms above 350°C. In the case of the He ion irradiation, the vacancy clusters grew by annealing above 750°C, and disappeared after annealing at 950°C. This was explained by the unstableness of the He-vacancy complex above 750°C, and the thermal decomposition of the He-vacancy clusters ended below 950°C.

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