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Vacancy-type defect production in iron under ion beam irradiation investigated with positron beam Doppler broadening technique

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Abstract

Accumulation of vacancy-type defects in iron produced by various ion irradiations at room temperature was investigated with positron beam Doppler broadening technique. Various ion species (He, C, O, Fe) were used for investigation of the effect of PKA energy on vacancy-type defect production. Accumulation of vacancy-type defects leads to sharpening of Doppler broadening spectra, i.e., increase of *S* parameter. *S* parameter was measured as a function of ion fluence for various ion species at a fixed positron energy of 15 keV. At the early stage of irradiation, *S* parameter increases with increasing fluence, and then it reaches some saturated *S* parameter value that varies with ion species. The saturated *S* parameter values tend to be higher for heavier ion irradiation. When compared with the incremental change of *S* parameter at the same low dpa level, heavier ions also lead to a greater change.

1. Introduction

The importance of defect production processes is widely recognized because they are the origin of a variety of radiation effects. Massive efforts have been made to investigate defect production processes in various metals, with recent computational techniques providing many theoretical insights into cascade damage structure. However, it is difficult to obtain the experimental data directly supporting the computational results due to the small dimension (∼nm) and the short time scale (∼ns) of the processes.

The positron annihilation technique is one of the promising tools for this work because of its high sensitivity to small vacancy-type defects, even monovacancies. The slow positron beam technique has demonstrated its capability of analyzing small vacancy-type defects induced by ion irradiation in various materials [1], so we constructed a variable energy positron beam line connected to the ion irradiation chamber for investigation of ion-induced defect production. This enables us to measure Doppler broadening of the 511 keV annihilation photon energy during ion irradiation, and after ion irradiation without exposure of the irradiated samples to the air.

In this paper, the vacancy accumulation process in iron induced by ion irradiation with various ion species is investigated with our variable energy positron beam. Iron is selected because iron is widely investigated both computationally and experimentally, and because vacancy-type defects in iron can be clearly detected due to significant sharpening of the Doppler broadening spectrum. Various ion species are selected as projectiles in order to vary the PKA energy spectrum.

2. Experimental

Samples are 99.99% pure iron sheets annealed at 1023 K for an hour in vacuum. Dimensions of each sample

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are 20 mm×20 mm×0.1 mm. Ion irradiations were carried out at High Fluence Irradiation Facility, Research Center for Nuclear Science and Technology, The University of Tokyo (HIT). Ion beam conditions are (1) 1 MeV He^+, (2) 560 keV He^+, (3) 1.7 MeV C^+, (4) 2.1 MeV O^+, (5) 2.8 MeV Fe^{2+}, where (1) to (4) are generated by 3.75 MV Van de Graaff, and (5) is generated by 1 MV tandem accelerator. Ion energies of (2) to (5) are selected so that the peak depth of defect production is nearly identical. All irradiations were carried out at room temperature without temperature control, which lead to sample heating by the ion beam up to 400 K.

The variable energy positron beam has been equipped at HIT and is connected to the irradiation chamber for the Van de Graaff accelerator for the in-situ or in-chamber positron beam Doppler experiment. A sealed ^{22}Na of 740 MBq is used as a positron source. Positrons emitted from the isotope penetrate into the moderator of annealed tungsten foil located close to the radioisotope, and then they are thermalized in the foil. The thermalized positrons are emitted from the surface with an energy of several eV by their negative work function. Positrons are extracted by the electric field applied between the moderator and an extraction grid mesh (50 V). Magnetic field is applied to the whole source chamber by Helmholtz coils to carry the positrons into the center of the beam transport. The beam transport consists of two straight pipes and two arc pipes of 90°, all of which work as solenoids with wound copper leads. The source chamber and the beam transport along with the connected electrical devices are electrically isolated from ground in order to apply high voltage for acceleration of positrons. Three Helmholtz coils apply magnetic field from the acceleration tube and the sample chamber. The applied magnetic field is around 100 G throughout the beam line. Spatial distribution of the positron beam can be visually seen with a microchannel plate assembly located at the sample position. Beam diameter is estimated to be about 6 mm, and the beam intensity is estimated to be approximately 10^4 e^+/s. The count rate at 511 keV photopeak is about 100–200 cps with a high purity germanium detector adjacent to the sample position. Doppler broadening spectra can be measured at controlled positron energies.

The positron beam line and the ion beam line are connected to the sample chamber so that both beams are merged at the center of the sample chamber at an angle of 30°. The experimental configuration which allows in-situ positron beam Doppler measurement is shown in Fig. 1. The positron beam is perpendicular to specimen surface while the ion beam is tilted at an angle of 30° from the normal to the specimen surface. Depth profiles of defect production calculated with the TRIM-92 code [2] and the stopping probability of 15 keV positrons are shown in Fig. 2. The positron stopping profile was calculated by Eq. (1) [3,4]:

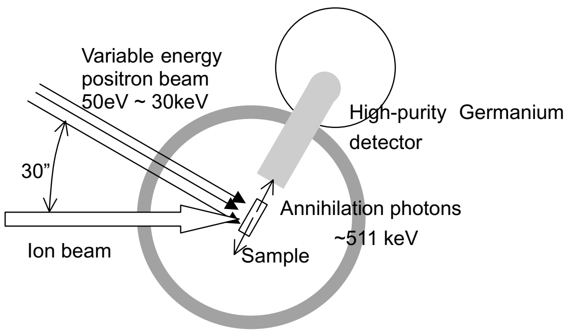

Fig. 1. Experimental configuration of in-situ positron beam Doppler measurement.

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

where $x_0 = 1.13\bar{x}$ and $\bar{x} = \alpha_p/\rho E^n$ (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 material are independent parameters. An evaluation by Vehanen et al. [5] can be employed to yield $m = 2$, $n = 1.62$ and $\alpha_p = 4.0$ μg/cm^{-2} keV$^{-1.62}$ in this calculation. The 15 keV positron beam was used for dose dependence measurement and in-situ measurement so that most of implanted positrons stop in the damage region and are effectively trapped by defects.

It is known that vacancy-type defects in iron 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) to counts in the whole photopeak is used to parameterize the peak shape change. Increase in S parameter means production of vacancy-type defects in iron. In this study, three types of measurement were done: (A) dose dependence of S

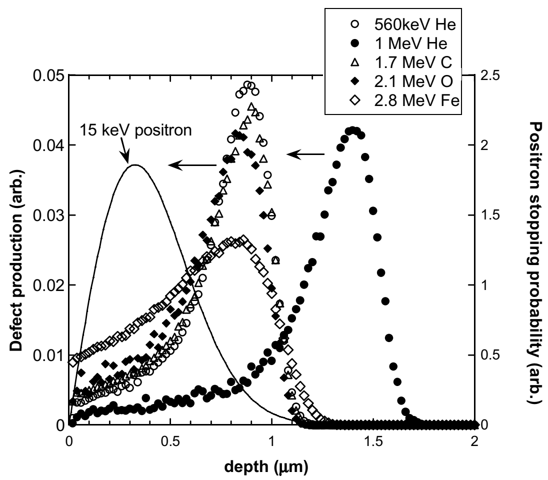

Fig. 2. Depth profile of defect production and stopping probability of 15 keV positron.

parameter by 15 keV positrons, (B) in-situ S parameter measurement by 15 keV positrons, and (C) S parameter measurement after irradiation by positron beam with variable energies.

3. Results and discussion

Fig. 3 shows dose dependence of S parameter for various ion beams along with in-situ measurement results as solid symbols. Dose in this figure means averaged dpa value from surface to 1 μm in depth. It is clear that S parameter increases with increasing dose at initial stage of irradiation, and then tends to show an upper limit. The limit seems to be different value for each ion beam condition.

Since most of 15 keV positrons do not diffuse back to surface, it can be simply considered that implanted and thermalized positrons annihilate either in bulk or at trapping sites. Then a simple trapping model can be employed, and the fractions of positrons annihilating in bulk (f_b) and positrons trapped and annihilating at defects (f_d) can be expressed as follows:

$$f_b = \frac{\lambda_b}{\lambda_b + \kappa C_d}, \tag{2}$$

$$f_d = \frac{\kappa C_d}{\lambda_b + \kappa C_d}, \tag{3}$$

where λ_b is bulk annihilation rate, κ is specific trapping rate for defects, and C_d is defect concentration. The measured S parameter can be considered as a linear sum of specific S parameter values for annihilation fractions by bulk and defects as follows:

$$S_{measured} = S_b f_b + S_d f_d, \tag{4}$$

where S_b and S_d are specific S parameter for bulk annihilation and annihilation at defects, respectively. Using the relationship of $f_b + f_d = 1$, the increment in S parameter (ΔS) can be described as follows:

$$\Delta S = S_{measured} - S_b = (S_d - S_b)\frac{\kappa C_d}{\lambda_b + \kappa C_d}. \tag{5}$$

This form of equation indicates that the measured S parameter will be close to S_d when $\lambda_b \ll \kappa C_d$, i.e., most positrons do not annihilate in bulk but are trapped by defects. Thus, the upper limits of dose dependence of S parameter can be thought to be S_d. S_d is the specific S parameter for annihilation at defects, and it depends on open volume of vacancy-type defects. This means that larger S_d means formation of larger vacancy-type defects. In Fig. 3, heavier ions increase the upper limit values, therefore, heavier ions are likely to produce defects with larger open volume.

Kiritani has schematically shown the progressive variation of the time dependence of the accumulation of vacancy-clustered defects during irradiation with collision cascades [6–8]. He proposed four stages of accumulation. Stage I is typical at the lowest dose range, where number density of vacancy clusters is proportional to the square of the irradiation dose. At Stage II number density of vacancy clusters is simply proportional to the irradiation dose, during which all vacancy clusters formed directly from cascades are preserved. At Stage III the number density of vacancy clusters is proportional to the square root of the irradiation dose. This behaviour is typical of an irradiation effect in which interstitials are free to annihilate vacancy clusters. At Stage IV, the number of vacancy clusters exponentially saturates to a certain level due to spatial overlap of cascades. Thus C_d in Eq. (5) can be assumed to follow one of these four types of dose dependence. The result of fitting analysis of the dose dependence of the measured S parameter indicates that assumption of Stage III gives the best among the four. This implies that vacancy type defects detected with positrons accumulate as a balance between direct formation from cascades and annihilation by interstitials escaping from cascades.

Although Fig. 3 includes data of positron beam Doppler measurement under irradiation as solid symbols, these data do not differ from those of off-beam conditions. Since all of the in-situ data lie in the saturation region, these values can be interpreted as a reflection of the sizes of open volume in a cluster rather than number density. Thus size of vacancy-type defects is not likely to change under and after irradiation in this temperature range, and this can be reasonably attributed

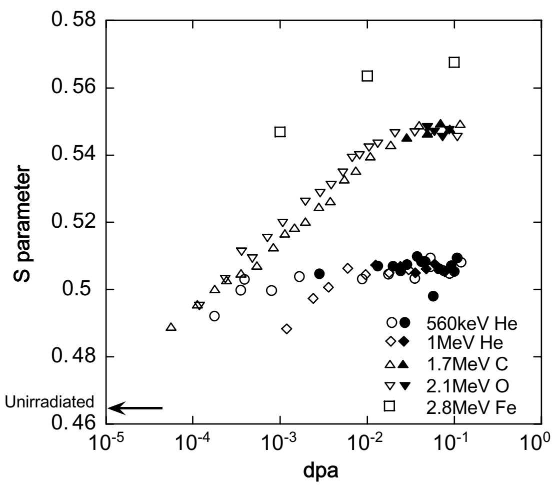

Fig. 3. Dose dependence of S parameter change in iron induced by various ion irradiations at room temperature. Measured with 15 keV positrons. Solid symbols express in-situ measurement.

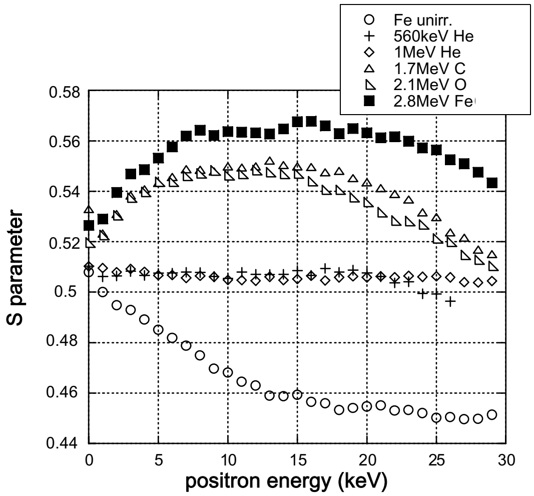

Fig. 4. Dependence of S parameter on positron energy measured after irradiation of 0.1 dpa.

to thermal stability of vacancy-type defects at room temperature.

Dependence of the S parameter on positron energy as measured after irradiation is shown in Fig. 4. For the defect-free specimen (circle), the S parameter decreases with increasing positron energy to settle down at the value S_b because of decreasing back diffusion fraction to surface. For the irradiated specimens, the S parameter initially goes up with increasing positron energy, and then reaches constant values for each condition, and finally goes down due to the increasing fraction of positrons passing through the defect region to reach defect-free region. However, data from 1 MeV He stay at S_d and do not go down because the damage region produced by 1 MeV He is deeper than others. Since those constant values can be regarded as S_d, this plot also indicates that heavier ions make larger vacancy clusters.

There are two possible reasons why heavier ions tend to produce larger open volume. One is difference in PKA energies among projectile ions. Heavier ions produce more PKAs with high energy (keV∼MeV in this study) than lighter ions, which cause displacement cascades. Computer simulations suggest PKAs with higher energy produce larger vacancy clustering after cascade cooling [9]. Those vacancy clusters might act as nuclei for subsequent clustering of vacancies nearby. The other reason is a difference in the number of PKAs per ion. A heavier ion produces more PKAs along its path simultaneously than a lighter ion, and those PKAs can effectively interact with each other due to their close spacing.

4. Conclusions

Accumulation of vacancy-type defects in iron by ion irradiation at room temperature was investigated with positron beam Doppler broadening measurement. S parameter rises with increasing dose and then it reaches a saturation value that depends on open volume from vacancy-type defects. Heavier ions are likely to produce larger open volume in vacancy-type defects than lighter ions. A difference in open volume between in-situ and post-irradiation was not obtained in this experimental condition.

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