Positron annihilation lifetime measurements of He-ion-irradiated Fe using pulsed positron beam

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Abstract. The positron annihilation lifetimes of He-ion-irradiated Fe samples were measured by a pulsed positron beam technique. The relative thickness of the surface oxidation layer was measured by Auger microprobe measurements. Unirradiated samples with different surface oxidation layer thicknesses had very similar second lifetime component intensities, indicating that the surface oxidation layer thickness does not affect positron lifetime spectra. In lifetime measurements using 10-keV positrons, the second lifetime component decreased monotonically with increasing irradiation dose. The mean lifetime in the 10-keV measurements was longer than that in 15-keV measurements (which correspond to the peak region of vacancy production); this is thought to be due to vacancy clusters absorbing He atoms.

1. Introduction
In fission and fusion reactors, large amounts of He are introduced into materials by either direct implantation or indirect means such as nuclear transmutation reactions. Because of its extremely low solubility and high mobility in materials, He tends to strongly agglomerate into bubbles, which deteriorates the mechanical properties of materials [1–3]. Therefore, it is critical to investigate the interaction between He and irradiation-induced defects, especially vacancies. Sugano et al. did not observe He bubbles in transmission electron microscopy images of α-Fe samples that been irradiated with 8 keV He ions at room temperature and subsequently annealed at temperatures up to 673 K, but they did observe bubbles in samples that had been annealed at 773 K [4]. To study the growth of He bubbles, it is essential to obtain information about the growth of small vacancy clusters and the interaction between small vacancy clusters and He atoms, which cannot be observed by transmission electron microscopy.

Positron annihilation spectroscopy can detect vacancy clusters and even single vacancies. Positron annihilation spectroscopy using slow-positron beam techniques is very effective for investigating the near-surface microstructure of ion-irradiated materials. Fujinami et al. performed a positron beam study of He–vacancy complexes in He-ion-implanted Si and detected the formation of He–V₂ complexes [5].

In the present study, we investigate the effect of the He irradiation dose on the formation of vacancy clusters in Fe by performing positron annihilation lifetime (PAL) measurements using a
pulsed positron beam. Since oxidation layers readily form on the surfaces of pure Fe samples, the effect of the surface oxidation layer (SOL) on PAL is also discussed.

2. Experimental procedure

Samples were prepared from 99.99% pure Fe that was cold rolled into approximately 0.1-mm-thick sheets. Each sheet was cut into 15 × 15 mm² pieces. All samples were mechanically polished using an abrasive coating with 1-μm-diameter particles. They were then annealed at 1073 K for 2 h in H₂ flow. He ion irradiation was performed at the National Institute of Advanced Industrial Science and Technology (AIST), Japan. The irradiation doses of the incident 150-keV He ions were 1 × 10¹⁸, 1 × 10¹⁹, and 1 × 10²⁰ He⁺/m². The irradiation was performed at room temperature. Calculations using SRIM code [6] gave a displacement damage peak of approximately 400 nm and a He penetration peak depth of about 450 nm. PAL measurements were performed using a variable-energy slow-positron beamline at the AIST linac [7]. The time resolution of the PAL measurements was about 270 ps. 10- and 15-keV positrons were implanted. Positron stopping profiles were calculated using the method described in Refs. 8–10. The average stopping depths of 10- and 15-keV positrons were 212 nm and 408 nm, respectively. Lifetime spectra were decomposed using the PALSfit program [11]. The relative thickness of the SOL was analyzed by a field-emission Auger microprobe (Jeol, JAMP-9500FV) at the Institute of Advanced Energy, Kyoto University, which utilized a 10-keV electron beam (current: 10 nA) to produce Auger electrons and a 1-keV Ar beam (current: 610 nA) for ion etching. The etching time per cycle was 2 s and the etching rate was 10.1 nm/min for Si. No data was obtained for Fe, so only the relative distribution of O atoms from the surface was obtained.

3. Results and discussion

3.1. Effect of surface oxidation layer on PAL

Table 1 lists the PALs of unirradiated samples measured by positrons with penetration energies of 10 and 15 keV. τ₃ probably originates from the background signal due to reemitted positrons or from the experimental apparatus. τ₁ is the lifetime of the matrix. The lifetimes of mechanically polished samples do not differ much from the lifetime of 106–110 ps obtained by conventional methods [12,13]. τ₁ of samples chemically polished with hydrogen fluoride is longer than the matrix lifetime (about 110 ps). This sample did not have a mirror surface, which may be one reason why it has a longer lifetime than the matrix. τ₂ is probably the lifetime of the SOL or the annihilation lifetime of positrons that penetrate inside the sample and then diffuse to the surface. The oxidation layer contains large voids whose lifetimes are about 500 ps [14]. Figure 1 shows the depth distribution of O atoms from the surface in uniradiated Fe. The horizontal axis denotes the ion etching time and the vertical axis corresponds to the amount of O atoms. The number of O atoms decreases with increasing depth. The chemically polished sample has a thinner SOL than the mechanically polished sample. If positrons directly implanted in the oxidation layer influence the positron lifetime, its intensity would vary with its thickness; however, all three treatments gave very similar intensities for τ₂. Therefore, τ₂ is thought to mainly originate from positrons diffusing from the bulk to the oxide interface. In the irradiated samples, positrons are trapped at defects and thus may not diffuse so easily. Consequently, we ignored the effect of SOL when we considered the results for the irradiated samples.

3.2. PAL measurements

Tables 2 and 3 give the PALs of He-ion-irradiated samples with positron penetration energies of 10 and 15 keV, respectively. In all cases, long lifetimes of about 1 ns are obtained for τ₃; they are due to the background signal caused by reemitted positrons or the experimental apparatus. In the 10-keV measurements, τ₁ remains constant and τ₂ decreases monotonically with increasing irradiation dose. In the 15-keV measurements, τ₁ increases for 10²⁰ He⁺/m² irradiation and τ₂ decreases initially and then increases with increasing irradiation dose.
In this study, the peak region of the vacancies and He atoms was measured at a positron penetration energy of 15 keV and the peak height of the generated vacancies was about three times higher than that at a depth of 210 nm (average penetration depth of 10-keV measurements). Troev et al. calculated PALS for vacancy clusters containing He atoms in Fe [15]. The positron lifetime of He bubbles decreases with increasing number of He atoms in vacancy clusters. The lifetime of vacancy clusters composed of 12 vacancies (V_{12}) with no He atoms is about 310 ps, whereas the lifetime of V_{12} containing 9 He atoms is about 200 ps. \( \tau_1 \) was about 140 ps. It is the mixed lifetime of single vacancy–He atoms complexes (V–He_n) and dislocation loops. \( \tau_2 \) (262–355 ps) is the lifetime for vacancy cluster–He atoms complexes (V_{m}–He_n).

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**Table 1.** PALs of unirradiated samples measured by positrons with penetration energies of 10 and 15 keV. MP: mechanical polishing; CP: chemical polishing; SS: soft sheet; HS: hard sheet. “\( \sim \)” indicates that it was not possible to decompose the spectra into three components.

| Positron energy | Surface treatment | \( \tau_1 \) (ps) | \( I_1 \) (%) | \( \tau_2 \) (ps) | \( I_2 \) (%) | \( \tau_3 \) (ps) | \( I_3 \) (%) |
|-----------------|-------------------|-------------------|---------------|-----------------|---------------|-----------------|---------------|
| 10 keV          | MP with SS        | 107 ± 6           | 68 ± 1        | 394 ± 3        | 31 ± 1        | 1141 ± 44      | 1 ± 0.1       |
| 10 keV          | MP with HS        | 110 ± 1           | 70 ± 1        | 407 ± 1        | 30 ± 1        | -              | -             |
| 15 keV          | CP                | 124 ± 1           | 71 ± 1        | 351 ± 2        | 29 ± 1        | -              | -             |
| 15 keV          | MP with SS        | 115 ± 1           | 84 ± 1        | 403 ± 3        | 15 ± 1        | 1023 ± 35      | 1 ± 0.1       |

**Table 2.** PALs of He-ion-irradiated samples with a positron penetration energy of 10 keV. \( \tau_{av} \) was calculated using \( (\tau_1I_1+\tau_2I_2)/(I_1+I_2) \).

| Irradiation dose | \( \tau_{av} \) (ps) | \( \tau_1 \) (ps) | \( I_1 \) (%) | \( \tau_2 \) (ps) | \( I_2 \) (%) | \( \tau_3 \) (ps) | \( I_3 \) (%) |
|------------------|----------------------|-------------------|---------------|-----------------|---------------|-----------------|---------------|
| \( 10^{18} \) He/m² | 228                  | 138 ± 10          | 58 ± 1        | 355 ± 4        | 41 ± 1        | 1456 ± 64      | 1 ± 0.1       |
| \( 10^{19} \) He/m² | 223                  | 148 ± 9           | 58 ± 2        | 330 ± 6        | 41 ± 2        | 1132 ± 39      | 1 ± 0.1       |
| \( 10^{20} \) He/m² | 224                  | 148 ± 11          | 53 ± 3        | 311 ± 6        | 46 ± 2        | 935 ± 25       | 1 ± 0.1       |

**Table 3.** PALs of He-ion-irradiated samples with a positron penetration energy of 15 keV. \( \tau_{av} \) was calculated using \( (\tau_1I_1+\tau_2I_2)/(I_1+I_2) \).

| Irradiation dose | \( \tau_{av} \) (ps) | \( \tau_1 \) (ps) | \( I_1 \) (%) | \( \tau_2 \) (ps) | \( I_2 \) (%) | \( \tau_3 \) (ps) | \( I_3 \) (%) |
|------------------|----------------------|-------------------|---------------|-----------------|---------------|-----------------|---------------|
| \( 10^{18} \) He/m² | 195                  | 142 ± 3           | 66 ± 2        | 301 ± 5        | 33 ± 2        | 1053 ± 32       | 1 ± 0.1       |
| \( 10^{19} \) He/m² | 192                  | 145 ± 6           | 59 ± 4        | 262 ± 7        | 40 ± 4        | 978 ± 24        | 1 ± 0.1       |
| \( 10^{20} \) He/m² | 221                  | 186 ± 4           | 74 ± 4        | 326 ± 12       | 25 ± 4        | 944 ± 37        | 1 ± 0.1       |

**Figure 1.** The depth distribution of O atoms from the surface in unirradiated Fe. MP: mechanically polished; CP: chemically polished; SS: soft sheet; HS: hard sheet.
When $\tau_{av}$ was measured for the same irradiation dose and at a different positron penetration energy; $\tau_{av}$ for 15-keV measurements is shorter than that of 10-keV measurements despite the high amount of vacancies produced. For $10^{18}$ and $10^{19}$ He$^{+}$/m$^2$ irradiation, $\tau_{2}$ of 15-keV measurements is also shorter than that of 10-keV measurements. This is because the amount of He atoms increased in the defect peak region so that more He atoms were absorbed by vacancy clusters.

In 10-keV measurements, vacancy clusters are formed by low dose irradiation and the vacancy clusters absorb He atoms by high dose irradiation. Consequently, the mean lifetime decreases with increasing irradiation dose. In 15-keV measurements, vacancy clusters also absorb He atoms. However, the production rate of vacancies is high in the defect peak region, and the growth speed of vacancy clusters exceeds the reduced lifetime caused due to the absorption of He atoms. These phenomena need to be demonstrated by performing simulations based on rate theory.

4. Concluding remarks
The PAL of Fe irradiated with He ions was measured by a pulsed positron beam technique. The relative thickness of the SOL was obtained using an Auger microprobe. The SOL thickness did not influence the lifetime spectra. PAL measurements detected the growth of vacancy clusters and $V_{n}$–He$^{m}$ complexes. The PAL decreases as the number of He atoms vacancy clusters that are absorbed increases. In the next step, the isochronal annealing behaviour of PAL will be investigated using a pulsed positron beam.

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