A positron beam study on vacancy formation in iron by ion beam irradiation at low temperature

T Iwai, K Murakami, Y Katano, T Iwata, T Onitsuka and H Abe

1 Nuclear Professional School, School of Engineering, The University of Tokyo, 2-22 Shirakata-Shirane, Tokai-mura, Ibaraki 319-1188, Japan
2 Department of Nuclear Engineering and Management, School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8654, Japan
3 Nuclear Safety Research Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Ibaraki 319-1195, Japan
4 Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan

iwai@nuclear.jp

Abstract. This study intends to investigate cascade damage structure produced by energetic ion irradiation. Cascade damage structure is preserved at low temperature below stage I where interstitial atoms begin to migrate. Then positron beam is implanted to the irradiated surface as a vacancy probe to evaluate vacancy concentration remained in the irradiated specimens. By this method, defect production efficiency was evaluated for iron irradiated with proton and carbon ions. The defect production efficiency values indicate enhanced recombination for carbon irradiation due to primary knock-on atoms (PKA) with higher energies.

1. Introduction

Point defect production by energetic particle irradiation is a common process in materials used for nuclear systems. This phenomenon leads to many kinds of degradation in the materials exposed to radiations, especially fast neutrons. Initial defect production processes (cascade damage) are important because they control the subsequent microstructural evolution and the macroscopic property change of the materials. Many efforts, mainly computational works, have been put into this fundamental process [1,2] to obtain damage parameters such as defect production efficiency, freely migrating defect production, and in-cascade clustering. But direct experimental validation is not sufficient because of the difficulty that comes from the time scale and the size (<nm) of the processes. There is a great need for experimental validation to raise the reliability and accountability of the computational works.

In this study, aiming at providing some experimental outcome of defect production processes, positron annihilation technique is applied to iron irradiated at low temperature below stage I where point defect configuration is preserved. Pure iron is chosen not only because steels are main structural materials in nuclear systems but also because many simulation results of iron exist for further comparison. Positron annihilation spectroscopy is a well established technique to probe vacancies and their clusters in iron, and slow positron beam is suitable for the detection of vacancies in ion-irradiated iron [3]. In this paper, an experimental evaluation of cascade damage parameters was challenged by combination of ion irradiation and positron beam Doppler broadening spectroscopy at low temperature below Stage I.
2. Experimental

Samples are 99.998% pure iron sheets annealed at 1023 K for an hour in vacuum. Dimensions of each sample are 15mm x 15mm x 0.1mm. Ion irradiations were carried out with a 3.75MV single ended Van de Graaff accelerator at High Fluence Irradiation Facility, The University of Tokyo (HIT). Ion beam conditions are 1 MeV H\(^+\) and 2.8 MeV C\(^+\) with an angle of incidence of 30°. All irradiations were carried out at 12 K with a cryostat system (IWATANI Cryo Mini 4K Cryocooler HE05).

The variable energy positron beam has been equipped at HIT and is connected to the irradiation chamber of the Van de Graaff accelerator for the in-situ or in-chamber positron beam Doppler broadening experiment [3,4]. This uses a sealed \(^{22}\)Na isotope of 740 MBq as a positron source and a tungsten foil as a positron moderator. Generated positrons from \(\beta^+\) decay are thermalized and emitted from the moderator, and then extracted by electric field of 50 V. They are magnetically transported along the S-figure beamline with solenoid to sample chamber, where a high purity germanium detector (ORTEC GEM 20180-P) is equipped for Doppler broadening measurement. Doppler broadening spectra can be measured at controlled positron energies from 50 eV to 30 keV. In this study positron beam Doppler broadening spectra were measured by fixed energy positrons: 20 keV for 1MeV H\(^+\) and 15 keV for 2.8MeV C\(^+\). Depth profiles of defect production calculated by SRIM-2008 [5] and positron stopping of Makhov profile [6] are shown in Figure 1. Irradiation and Doppler measurement are sequentially repeated at 12 K to obtain detailed dose dependence of S parameter defined as the ratio of counts in the central region (511 ± 0.8 keV) to counts in the whole photo peak.

![Figure 1](https://example.com/figure1.png)

**Figure 1** Depth profile of defect production and positron stopping (A) 20keV e\(^+\) on 1MeV H\(^+\) (B) 15keV e\(^+\) on 2.8MeV C\(^+\).

3. Result and discussion

Figure 2 shows a dose dependence of S parameter in iron at 12 K for H\(^+\) and C\(^+\) irradiation. Displacement per atom calculated from Norgett-Robinson-Torrens model (NRT model) [7] by SRIM-2008 is employed as the dose unit. This value is averaged between the surface and 1\(\mu\)m in depth. S parameter for both irradiations was increased with dose, then tended to saturate. When defect concentration is sufficiently high, most of positrons should be trapped at defects and measured S parameter can be treated as defect-specific S parameter \(S_d\). In this experiment, this S parameter saturation should occur due to high concentration of defects preserved without thermal process hereafter. Then this saturated value in figure 1 can be treated as \(S_d\). Increase in S parameter of H\(^+\) irradiation is faster than that of C\(^+\) irradiation, and the saturated S value is similar for each ions. This S value is much lower than \(S_d\) for larger vacancy clusters in the reference [3] of Fe\(^{2+}\)-irradiated iron at room temperature.

In simple case, measured S parameter can be generally expressed as a linear combination of specific S parameters and each positron fraction:
where subscript \( b, d \) and \( s \) denote bulk, defect and surface, respectively [8]. At 15 and 20 keV \( e^+ \) in irradiated specimens, contribution of back diffusion to surface should be minimal. Along with the two state trapping model, defect concentration \( (C_d) \) can be expressed as:

\[
C_d = \frac{(S_{\text{measured}} - S_b) \lambda_b}{(S_d - S_{\text{measured}}) \mu}
\]  

(2)

where \( \mu \) and \( \lambda_b \) denote specific trapping rate for defects and bulk annihilation rate \((=9.1 \times 10^9 \text{ s}^{-1})\), respectively.

From computational researches, majority of vacancies exist as monovacancies, even in a cascade (only \( \sim 3\% \) for vacancies in clusters if nearest neighbor criterion is applied [9]). Assuming that the almost all the trapping sites in this experiment are monovacancies is reasonable and consistent with these terms. Then \( \mu \) value for monovacancies estimated by Vehanen et al. \(((1.1 \pm 0.2) \times 10^{15} \text{ s}^{-1})\) [10] can be applied. By equation (2), \( C_v \) (vacancy concentration) can be estimated.

In this temperature range below stage I assigned by Takaki et al. [11], vacancies and interstitials are immobile to suppress mutual annihilation and clustering after cascade processes. Under this condition where cascade structure is preserved, the ratio of vacancy concentration to NRT dpa can be treated as defect production efficiency \( \eta \). Figure 3 shows the dose dependence of the vacancy concentration along with a line which indicates that \( \eta=1 \). Linear increase of \( C_v \) with NRT dpa was observed up to 0.3 mdpa as expected, and the slopes are ‘apparent’ defect production efficiencies. They are estimated to be \( 0.3 \pm 0.1 \) for \( H^+ \) and \( 0.2 \pm 0.1 \) for \( C^+ \).

For comparison, defect production efficiency was roughly estimated by PKA energy spectrum from SRIM-2008 calculation and published computational efficiency values compiled by Malerba [2]. The estimated values were 0.52 for 1MeV \( H^+ \) and 0.42 for 2.8MeV \( C^+ \). Although our experimental values qualitatively represent the difference of ion species well, our values are approximately half of the computational values. These values are not necessary to coincide, but we cannot still exclude possibilities of several phenomena to reduce our experimental values. The one possibility is the positron trapping at vacancies locally concentrated near PKAs (cascade). The \( \mu \) value for monovacancies estimated by Vehanen et al. was estimated by combined experiment of electric resistivity and positron annihilation spectroscopy for electron-irradiated iron. In that case, Frenkel pairs are uniformly distributed to trap positrons effectively. But locally concentrated vacancies by cascade damage might reduce positron trapping probability compared to homogeneous defect

![Figure 2](image_url)
distribution due to larger defect free volume. This effect may reduce apparent vacancy concentration. Another possibility is modest migration of interstitials to occur recombination of closed Frenkel pairs, possibly caused by electronic excitation accompanied with ion irradiation.

In-cascade clustering is another important subject of cascade damage structure. However, our experimental method was not suitable for detecting clustering in cascade because considerable concentration of monovacancies around clusters effectively trap positrons to hide clusters from positrons.

![Figure 3](image-url)

**Figure 3** Dose dependence of vacancy concentration calculated from measured $S$ parameter.

4. Conclusions
Vacancy accumulation by ion irradiations at low temperature below stage I was investigated with positron beam Doppler broadening. Formed vacancy-type defects were likely to monovacancies, independent of PKA energy spectrum. ‘Apparent’ defect production efficiency values qualitatively represent the difference in PKA energy spectrum of $H^+$ and $C^+$. The values were lower than simulation-based ones, possibly due to inhomogeneous distribution of vacancies caused by cascades and enhanced mutual annihilation of Frenkel pairs. Positrons are too sensitive to monovacancies nearby to probe in-cascade vacancy clustering.

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