Investigation of defect production in iron on gamma irradiation using the positron Doppler broadening technique

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The parameters S and W corresponding to the positron Doppler broadening technique have been used to study displacement type defects produced in iron samples irradiated by various imparted doses of gamma ranging from 20 Gy to 2kGy. For the logistic support of the experimental observations, Monte Carlo simulation of displacements has also been performed. Of the increase of S parameter with dose up to a limit, it is revealed that vacancy type defects dominate. From both the experimental observations and the Monte Carlo study, it is inferred that on irradiation monovacancies are produced and under the given irradiation conditions there is negligible chance of the atom+atom collision cascade which may give rise to multiple type vacancies.

Keywords: Gamma irradiation, Positron Doppler broadening, S and W parameters, Positron annihilation

1 Introduction

On passage of radiation through matter, displacements are produced which manifest in the form of microstructures, mono-vacancies and/or the clusters. Doppler broadening technique based on the physical process of positron-electron annihilation is a state of the art technique and it is generally, employed to investigate the concentration and nature of vacancies or the clusters1. Mono-vacancies will correspond to a different electronic configuration as compared to a cluster. The positron annihilation parameters are very sensitive to the fraction of which the positrons annihilate with a valence or a core electron. Analytically, S-parameter corresponds to rise of the photopeak and W-parameter to the width of the photopeak of annihilation of gamma energy spectrum. For more details and practical utility of DB analysis reader is advised to refer to Iwai et al2.

Defects in iron and iron-alloys have been studied using the two photon Coincidence Doppler Broadening (CDB) technique in case of their irradiation by neutrons3. Similarly, vacancy formation in iron by the proton and carbon beams have also been studied4. Very little experimental data of vacancy formation are available in case of iron irradiated by gammas. Such a study is required in case of a reactor vessel where vessel steel is irradiated both by neutrons and gamma and embrittlement of the vessel is considered to be significant5 because of gammas. Nature of vacancies created by gamma radiation may be investigated using the DB technique. Thus, the iron samples irradiated to 6 different doses of gamma spectrum of cobalt-60 source have been investigated and results on the status of vacancies are presented in this paper after incorporating the results of Monte Carlo simulation of the irradiation.

2 Experimental Details

Pure iron samples (99.5+% pure) each of 0.039 cm thickness were irradiated with gamma doses ranging from 2 Gy to 2 kGy from a cobalt-60 source strength, 1.7667×1014 Bq (= 4775 Ci) available at the gamma irradiation facility GC-1200, IUAC, New Delhi. Exposure dose rate at the time of irradiation was 5.538 kGy / h. Description of sample size, exposure time, total absorbed dose and total number of incident photons falling on the samples are given in Table 1 alongwith the irradiation temperature.

A schematic diagram of the experimental set-up is shown in Fig. 1 in which 22Na positron source of initial radioactivity, 1.295×109 Bq (35 mCi) is installed in a lead cavity at 15.24 cm below the sample holder. Positrons of endpoint energy 0.541 MeV lose ~ 0.041 MeV in the air and finally
strike the sample with a leftover average energy \( \sim 0.219 \text{ MeV} \). Monte Carlo simulation of the depth profile of positrons in iron has been studied and according to that a positron will penetrate up to 0.01 cm in the sample. Also, the positron source emits gamma of 1.28 MeV, which will also damage the sample during DB measurements. In our case, measurements are made for 8 h on each sample and in this period only \( 4.8 \times 10^9 \) gammas emitted from the positron source that strike the sample. According to MC simulation, there are only \( \sim 159 \) displacements produced by these gammas in a sample. This number is very small as compared to the number of displacements, \( N_d \sim 10^9 \) due to the original irradiation. Resolution of the HPGe detector used in the experiment is 0.004 at 511 keV and 0.003 at 1 MeV.

2.1 Monte Carlo Simulation

A Monte Carlo code\(^6,7\), JA-IPU has been used to study the defect profile due to the incident gamma spectrum of the Cobalt-60 source. For the purpose of simulation, we have generated histories of \( 2 \times 10^8 \) photons to fall on the sample perpendicularly. An incident photon transfers energy to an electron of the medium by way of three main process i. e. photoelectric effect, Compton scattering and pair-production. Kinetically, an ejected energetic electron having kinetic energy greater than 0.629 MeV may displace an atom from the iron lattice. The details of the simulation scheme and the data libraries used in simulation have been discussed\(^8\).

### Results of Simulation and the Experiment

#### 3.1 Depth Profile of Defects and Positron Stopping Range

Distribution of positions of displacements/defects produced in the sample on impact of \( 2 \times 10^8 \) photons is shown in Fig. 2 in the form of the number of displacements per incident photon (\( N_d /\text{ph} \)). It can be seen that the \( N_d /\text{ph} \) are uniformly distributed over 0.01 cm depth of the sample.

For the stopping probability of the positron, Makhov profile method described in Ref. (8) has been used. According to this:

\[
P(z, E) = \frac{m}{z_0^m} \exp \left[ -\left( \frac{z}{z_0} \right)^n \right] \quad \text{with} \quad z_0 = 0.113 \frac{\alpha E_n}{\rho}
\]

where \( m, n \) and \( \alpha \) are empirical parameters independent of the material. Vehanen et al.\(^8\) have given the values \( m = 2, n = 1.62 \) and \( \alpha = 4 \mu g/cm^2\cdot keV \) and \( E \) is the energy of positrons in

### Table 1 — Details of sample size, exposure time and exposure dose and the total number of incident photons on a sample at 300 K

<table>
<thead>
<tr>
<th>Sr. No.</th>
<th>Sample size (s)</th>
<th>Exposure time (s)</th>
<th>Exposure dose (Gy)</th>
<th>Number of incident photons</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.05x0.5x0.039</td>
<td>13</td>
<td>20</td>
<td>2.22x10^14</td>
</tr>
<tr>
<td>2</td>
<td>1.10x0.8x0.039</td>
<td>32.3</td>
<td>50</td>
<td>9.32x10^14</td>
</tr>
<tr>
<td>3</td>
<td>1.05x0.6x0.039</td>
<td>65.1</td>
<td>100</td>
<td>1.21x10^15</td>
</tr>
<tr>
<td>4</td>
<td>0.8x0.63x0.039</td>
<td>650.1</td>
<td>1000</td>
<td>1.07x10^16</td>
</tr>
<tr>
<td>5</td>
<td>0.8x0.63x0.039</td>
<td>975.1</td>
<td>1500</td>
<td>1.60x10^16</td>
</tr>
<tr>
<td>6</td>
<td>0.8x0.65x0.039</td>
<td>1300.1</td>
<td>2000</td>
<td>2.20x10^16</td>
</tr>
</tbody>
</table>

Fig. 1 — Schematic diagram of the experimental set-up of Doppler’s Broadening measurements
keV and ρ is the density of material in g/cm³. It is clear from the plot of the positron absorption profile shown in Fig. 2 that almost all of the incident positrons will be trapped in 0.01 cm thickness of a sample.

3.2 PKA Energy Distribution

Figure 3 shows the results of Monte Carlo simulation of kinetic energy of primary knocked-out atom (PKA) in a sample and the distribution is qualitatively in agreement with that envisaged by Brinkman. This indicates that the energy transferred by produced electron to the PKA is in between 40 to 62.5 eV. Because of the fact that PKA energy is less than 2Ed, therefore, chances of development of an atomic cascade are negligibly small and we can safely assume that due to irradiation only monovacancies are created in the sample. Unfortunately, motion of the monovacancies dependence on temperature has not been considered.

3.3 Identification of Vacancies by DB

Figure 4 shows the S parameter dependence on the absorbed dose. Dose displayed along X-axis means the dose absorbed in only 0.01 cm depth of the sample. In Fig. 4, the normalized value of S parameter with respect to the bulk material of the sample i.e. S/Sb is plotted. An increase in the S/Sb up to an absorbed dose say 0.0397 Gy, shows that available vacancies or the defects are such that positrons annihilate with the valence electrons. At higher doses, decline of S/Sb shows that positrons annihilate dominantly with the core electrons.

According to Iwai et al., the S parameter can be considered as a linear sum of specific S values multiplied by the fraction of positron annihilation in bulk at trapping sites or in the produced defects:

\[ S = S_b f_b + S_d f_d \]

where Sb and Sd denote the specific S parameter for the bulk and defect, respectively. Also, fb and fd are the fraction of positron annihilation in bulk and the defect site, respectively and they can be calculated from the following relations:

\[ f_b = \frac{\lambda_b}{\lambda_b + \mu C_d} \quad \text{and} \quad f_d = \frac{\mu C_d}{\lambda_b + \mu C_d} \]
where \( \mu \) is the specific trapping rate at the defects (for iron its value is \( 1.1 \times 10^{15} \text{ s}^{-1} \) for the mono vacancies\(^1\)) and \( \lambda_b \) is the bulk annihilation rate (for iron\(^2\) it is taken to be \( 9.1 \times 10^9 \text{ s}^{-1} \)). The defect concentration, \( C_d \) can be calculated\(^10\) from the following relation:

\[
C_d = \frac{(S - S_b) \lambda_b}{(S_d - DS) \mu}
\]

\[
\mu C_d = \left( \frac{S}{S_b} - 1 \right) \lambda_b = \left( \frac{S_d}{S_b} - S \right) \left( \frac{S}{S_b} - S_b \right)
\]

The values of \( S/S_b \) based on the measured values of \( S \) and \( S_b \) along with calculated values of various other parameters are given as a function of the absorbed dose. \( S/S_b \) has been plotted as a function of dose in Fig. 4. The displayed data may be divided in two segments, i.e. (i) rising part and (ii) decline part. In case \( \lambda_b \leq \mu C_d \) measured value \( S/S_b \leq S_d/S_b \) and \( S/S_b \) rises with the dose and at 0.0397 Gy the rising trend nearly stops. Probably, this is the condition when \( S/S_b \sim S_d/S_b \) where the fraction of the annihilation at monovacancies reaches a saturation. In case of the last dose, value of \( S/S_b = 1.010273 \) is slightly less than the saturation value. The difference can easily be neglected due to large errors. If at all it has meaning then it indicates that the positron annihilation takes place at sites where a paired core electronic configuration exists and these sites are different to the pristine lattice sites. If this is the case, then the value of \( W/W_b \)-parameter should be enhanced in case of all the doses after 0.0397 Gy. This can be verified from the data displayed in Fig. 5 that after the absorbed dose 0.0397Gy, value of \( W/W_b \) starts increasing.

From the calculation of fractions of positrons annihilating at defect site, (FD), we can say that in case of first 5 doses more than 85% positrons are trapped in defect positions. Sudden drop in case of maximum absorbed dose at 0.08 Gy only 36.9% of positron are trapped in reduced vacancy type defects. These conclusions are supported by the data of \( W/W_b \) plotted as a function of dose.

In case of single type vacancies W-S plot\(^1\) is supposed to follow a straight line bound in between \( (S_b, W_b) \) and \( (S_d, W_d) \) values. In Fig. 6, \( W/W_b \) has been plotted as a function of \( S/S_b \) and it is evident that the data satisfies the aforesaid condition.

### 4 Discussion and Conclusions

Simulation of the depth profile of displacements using the JA-IPU code has been useful in understanding that the defects are uniformly distributed. Also, from the distribution of kinetic energy of PKA as shown in Fig. 3, it may be inferred that existing vacancies are monovacancy and there is no chance of multiple type displacements or vacancies due to atomic cascades.

From the experimental study of Doppler broadening parameters following conclusions can be drawn:

1. At the initial low doses of gamma radiation, the value of \( S/S_b \) increases with increasing dose, and then tends to saturate.
2. It is evident that in the saturation region more than 85% positrons are trapped at defect sites where
annihilation with valence electrons takes place. At later, higher doses decline in $S/S_b$ and complimentary rise of $W/W_b$ indicates that annihilation takes place, preferably with the core electrons.

3 The second source of better information can be an estimation of free volumes as function of gamma radiation dose using the positron annihilation lifetime analysis (PAL).

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References


