Swift heavy ion induced thermoluminescence studies in polycrystalline aluminum oxide

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When energetic swift heavy ions interact with matter, inelastic collision (leading to electronic energy loss \( S_e \)) and elastic collision (leading to nuclear energy loss \( S_n \)) take place. In the present study, the effect of energetic ion species on thermoluminescence (TL) of polycrystalline aluminum oxide (PAO) is reported. PAO pellets of 6 mm diameter are irradiated with energetic \( \text{Au}^{9+} \), \( \text{Ni}^{7+} \) and \( \text{Si}^{7+} \) ions for the fluence of \( 1 \times 10^{13} \text{ ions cm}^{-2} \). A single well resolved prominent TL glow with peak at 538 K is observed in \( \text{Si}^{7+} \) irradiated samples. However, in \( \text{Ni}^{7+} \) and \( \text{Au}^{9+} \) irradiated samples a prominent TL glow with peak at 610 K along with a shoulder at 513 K is observed. On the other hand, when PAO samples are irradiated with \( \gamma \)-rays two well separated TL glows with peaks at 483 K and 638 K are observed. A prominent PL emission with peak at 430 nm besides a weak emission with peak at 480 nm and a shoulder at 525 nm are observed in 120 MeV \( \text{Au}^{9+} \) ion irradiated samples when excited with 320 nm. These PL peaks are attributed to \( F \), \( F_2^{+} \) and \( F_2^{2+} \)-centers respectively. However, in \( \text{Si}^{7+} \) irradiated samples a single PL emission peak at 430 nm is observed and it is attributed to \( F \)-centers.

Thermoluminescence is the luminescence emitted from a previously irradiated insulator or semiconductor as a result of thermal stimulation. The TL intensity is a function of dose of radiation absorbed by the sample and thus is used in radiation dosimetry. The process can be described phenomenologically using the energy band scheme involving delocalization of electrons from traps followed by recombination with holes resulting luminescence emission\(^1\). \( \text{Al}_2\text{O}_3 \) is one of the earlier materials studied for its possible application as a radiation dosimeter owing to its superior thermal and chemical stability and low effective atomic number\(^2\). Further, aluminum oxide has been used historically as an imaging plate for tuning high current electron beam accelerators with good success. The material survives for couple of months and it appears to exhibit monotonic increase of light output with electron beam current\(^3\). Swift heavy ions (SHI) are very useful for modification of the properties of films, foils and surface of bulk solids. It penetrates deep into the target material and produces a long and narrow disordered zone along its trajectory. The passage of SHI induces very rapidly developing processes which are difficult to observe during or immediately after their occurrence. The information about these processes is stored resulting damage, such as size, shape, structure of defects, etc. The degree of disorder can range from point defects to a continuous amorphized zone along the ion path, commonly called latent track\(^4\). In the present study, the thermoluminescence and photoluminescence of SHI irradiated polycrystalline aluminum oxide are reported.

**Theory**

When a fast energetic ion penetrates in a solid it loses its energy mainly by two nearly independent processes: (i) elastic collisions with the nuclei known as the \( S_n \)-nuclear energy loss \( \langle dE/dx \rangle_n \), which dominates at an energy of about 1 keV/amu; and (ii) inelastic collisions of the highly charged projectile ion with the atomic electrons of the matter known as \( S_e \)- electronic energy loss \( \langle dE/dx \rangle_e \) which dominates at an energy of about 1 MeV/amu or more. In the inelastic collision (cross-section \( \sim 10^{-16} \text{ cm}^2 \) ) the energy is transferred from the projectile to the atoms through excitation and ionization of the surrounding...
electrons. The amount of electronic loss in each collision varies from tens of eV to a few keV per Angstrom (Å). The passage of SHI in materials mainly produces electronic excitation of the atoms in the materials. SHI causes exotic effects in different classes of materials which otherwise cannot be generated by any other means. Quantitatively, it is capable of depositing electronic excitation energy of about 1-10 keV/Å in the materials. Such a large electronic excitation brings out various changes in materials. During the passage of SHI through materials, neighboring positive target ions are produced by electronic excitation induced ionization. These positive ions are mutually repulsive. The time to cover atomic sites is short in comparison to the response time of the conduction electrons. So during the passage of the ion a long cylinder containing charged ions is produced.

Figure 1 shows the variation of electronic energy loss ($S_e$) and nuclear energy loss ($S_n$) for Au ions in aluminum oxide target. The variation of electronic energy loss ($S_e$) and nuclear energy loss ($S_n$) for Si ions in aluminum oxide is shown in Fig. 2. The calculation of $S_e$ and $S_n$ is done using SRIM 2003 program. It is observed that, in both the cases the maximum energy deposition takes place by means of inelastic collisions (electronic energy loss-$S_e$) but not by elastic collisions (nuclear energy loss-$S_n$) by the ions having the energies in the range of few MeV to few hundreds of MeV. However, in the present study the maximum $S_e$ deposited in aluminum oxide is observed from gold ions when compared to that with silicon ions.

**Experimental Procedure**

Polycrystalline aluminum oxide used in the present investigations is synthesized by combustion technique. In order to handle the phosphor easily, 100 mg of polycrystalline $\text{Al}_2\text{O}_3$ powder was grained into a fine powder using an agate and mortar and mixed with a drop of polyvinyl alcohol in a cylindrical pilot of the palletizer to make pellets. Pellets of 6 mm diameter and 1 mm thickness from the above mixture were obtained by applying pressure of about 70 MPa using a home-made palletizer at room temperature. As prepared pellets were irradiated with SHI for fluence $1 \times 10^{13}$ ions cm$^{-2}$ using 15UD Tandem Pelletron Accelerator in Materials Science beam line at Inter University
Accelerator Centre, New Delhi, India. The details of ions used for irradiation are given in Table 1. TL measurements were made using TL reader (Nucleonix, India) connected to a PC. The TL signal was integrated from RT up to 673 K at a heating rate of 5 Ks⁻¹. The PL measurements were made using Perkin Elmer LS55 spectrometer. The sample was excited using 320 nm of Xenon lamp. All experiments are confined to room temperature only.

**Results and Discussion**

Thermoluminescence glow curves of polycrystalline Al₂O₃ irradiated with swift heavy ions for the fluence 1×10¹³ ions cm⁻² is shown in Fig. 3. Two TL glows - a well resolved one with peak at ~610 K and a weak one at ~513 K are recorded in 120 MeV Au⁹⁺ ion irradiated sample. When the samples are irradiated with 100 MeV Ni⁷⁺ ions a well resolved TL glow with peak at 610 K and an unresolved shoulder at 513 K is observed. In the case of 100 MeV Si⁷⁺ ions single prominent well resolved TL glow with peak at 537 K is observed. It is observed that the shoulder at 513 K is absent in silicon irradiated samples. This may be attributed to less amount of energy deposition leading to formation of simple point defects. Further, when polycrystalline aluminum oxide samples are irradiated with γ-rays two well separated TL glows with peak at 483 K and 638 K are observed. ⁶

Figure 4 shows the photoluminescence emission spectra of swift heavy ions irradiated polycrystalline Al₂O₃ for 1×10¹³ ions cm⁻².

![Fig. 3—Thermoluminescence glow curves of polycrystalline Al₂O₃ irradiated with swift heavy ions for the fluence 1×10¹³ ions cm⁻²](image1)

![Fig. 4—Photoluminescence emission spectra of swift heavy ions irradiated polycrystalline Al₂O₃ for 1×10¹³ ions cm⁻²](image2)

**Table 1—Ion species, its mass number, energy, charge state, \( S_e, S_n \) and range used for irradiation**

<table>
<thead>
<tr>
<th>Ion species</th>
<th>Mass number</th>
<th>Energy (MeV)</th>
<th>Charge state</th>
<th>Electronic energy loss (keV nm⁻¹)</th>
<th>Nuclear energy loss (keV nm⁻¹)</th>
<th>Range (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>196.26</td>
<td>120</td>
<td>+9</td>
<td>24.80</td>
<td>0.361</td>
<td>09.18</td>
</tr>
<tr>
<td>Ag</td>
<td>108.87</td>
<td>100</td>
<td>+7</td>
<td>19.76</td>
<td>0.012</td>
<td>08.75</td>
</tr>
<tr>
<td>Si</td>
<td>28.08</td>
<td>100</td>
<td>+7</td>
<td>04.41</td>
<td>0.033×10⁻³</td>
<td>19.66</td>
</tr>
</tbody>
</table>

The TL measurements were made using TL reader (Nucleonix, India) connected to a PC. The TL signal was integrated from RT up to 673 K at a heating rate of 5 Ks⁻¹. The PL measurements were made using Perkin Elmer LS55 spectrometer. The sample was excited using 320 nm of Xenon lamp. All experiments are confined to room temperature only.
observed and it is attributed to F-centers. However, the PL peaks at 480 and 525 nm are not resolved in this case probably due to overlapping with the 430 nm emission. The extra TL shoulder observed is attributed to complex defect centers created due to large amount of energy deposited through electronic energy loss. Therefore, it is believed that the ions with high $S_e$ deposition lead to formation of complex defect centers. The prominent TL glow (610 K) in the present studies may be attributed to recombination of electrons (F-centers) with holes. The weak and unresolved TL glow (513 K) might be due to recombination of M-centers with another type of hole centers. However, optical absorption/photoacoustic studies of aluminum oxide reveal the nature of electrons (F and F aggregate) and hole centers.

**Conclusions**

Two TL glows one with peaks at 610 K and another at ~513 K are observed in both 120 MeV Au$^{9+}$ and 100 MeV Ni$^{7+}$ ion irradiated Al$_2$O$_3$ samples. A single and well resolved TL glow with peak at 537 K is observed in 100 MeV Si$^{7+}$ ion irradiated Al$_2$O$_3$ with absence of 513 K. This might be due to less amount of energy deposition in aluminum oxide to form higher complex centers. The well resolved PL emission peaks at 430, 480 and 525 nm in Au ion irradiated alumina are attributed to F, F$_2$ and F$_2^{2-}$-centers respectively. It is believed that the ions with high $S_e$ deposition lead to formation of complex defect centers.

**References**

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