IV OPTICALLY STIMULATED LUMINESCENCE

IV-1 INTRODUCTION

On irradiation of a phosphor by high-energy radiation like UV, X-rays or γ-rays, electrons in the samples get excited and trapped in the metastable levels of the phosphor. One of the methods to detrap these electrons is to stimulate the sample by electromagnetic radiation of energy relatively very much lower than that used for excitation. These optically detrapped electrons on recombination at luminescence centres produce luminescence, which is called optically stimulated luminescence (OSL).

As stimulation of the excited sample continues, the detrapping process depletes the population of the metastable levels, thereby decreasing the rate of detrapping, resulting in the gradual decrease of OSL intensity. Thus, the OSL decay curve is an indirect display of the population levels of such traps and detrapping rates. Preliminary surveys have shown that most of the commonly used thermoluminescent materials like LiF:Mg:Ti, LiF:Mg:Cu:P, CaSO₄:Dy, Al₂O₃:C etc. produce OSL signals in a lesser or greater extent of which Al₂O₃:C is having high OSL sensitivity.

The key property in determining whether a particular phosphor is a good OSL material is the photo ionization cross-section of the traps under study. All the traps cannot be emptied optically with high efficiency using visible or UV wavelengths. In all applications of OSL, we are relying on the fact that the thermoluminescence from these materials is optically unstable, i.e. TL signal is observed to fade if the sample is exposed to visible light.
OSL studies have been carried out by various workers\textsuperscript{(100,108)}, using both coherent and incoherent radiation. Most of the works in OSL are related to dosimetry for dating purposes. It has been found that quartz and feldspar show OSL using radiations from Infra Red Light Emitting Diode (IR LED) as stimulant\textsuperscript{(102,103)}. Huntley was the first one to use green light from argon laser (514.5 nm) to stimulate luminescence from quartz for dating sediment\textsuperscript{(104)}. Later, broadband emitters such as incandescent or arc lamps, in conjunction with carefully selected filters were used to produce both infrared and visible light stimulated luminescence from feldspar and quartz samples\textsuperscript{(105,106)}.

Gasiot et al., has studied the OSL emission from CaS:Eu,Sm on stimulation with Nd:YAG laser\textsuperscript{(83)}. The study showed that high resolution OSL imaging of spatial dose distribution is possible via stimulation of the dose proportional luminescence with a focused scanning laser beam. Chakrabarti et al. studied the OSL spectra of CaS:Ce,Sm and obtained Ce\textsuperscript{3+} doublet emission\textsuperscript{(107)}. It is explained as due to the recombination of optically stimulated electron from Sm\textsuperscript{3+} ions at holes trapped in Ce\textsuperscript{3+} ions.

OSL spectra of CaS:Eu,Sm was studied by Tamura and Shibukawa using monochromatic light from spectrophotometer\textsuperscript{(108)}. They studied the concentration dependence of infrared stimulated luminescence intensity. According to Hutt and Jack the traps related to OSL from CaS doped with Bi and In can be attributed to intrinsic defects in CaS and to single In\textsuperscript{3+} or Bi\textsuperscript{3+} states\textsuperscript{(109)}.

Gong et al., studied the OSL emission spectra of CaS: Sm\textsuperscript{3+} using YAG laser of 1064nm\textsuperscript{(110)}. The OSL spectrum was found to be similar to fluorescence emission spectrum. Using pico second infrared pulse from YAG laser, Fan et al. studied the OSL response of CaS:Eu,Sm\textsuperscript{(111)}. According to them the stimulated emission from the phosphor decays according to the law $I = I_0(1 + \tau t^{1/2})^{-\tau}$ where, $\tau = 1.56$. 

\textsuperscript{67}
The description of OSL process is based on a phenomenological model for luminescent dosimetric materials, as given in Fig. 4.1.

![Phenomenological model for OSL diagram](image)

**Fig. 4.1**

**Phenomenological model for OSL**

(1) shallow trapping level, (2) main dosimetric trap, (3) thermally disconnected trap, (4) recombination site, \(a\) – optical stimulation, \(b\) – recombination resulting in OSL, \(c\) – retrapping in deeper trap (3) and \(d\) – optical release of charge from deeper trap (3).

This model consists of a main dosimetric trap (2) in Fig. 4.1, from which electrons may be released optically (in OSL) or thermally (in thermoluminescence). Recombination of these released electrons with holes at recombination sites (4) in Fig. 4.1 produces OSL or thermoluminescence (TL) emission. In addition to main dosimetric traps most of the luminescent materials can be expected to contain shallow trapping levels (1) in Fig. 4.1, which are incapable of storing trapped charges over long periods at normal temperatures. Although these levels are normally not directly employed in most dosimetric applications, they do contribute to the overall characteristics of the
luminescence behavior of the material. Also we can expect the existence of deep thermally disconnected traps (3) in Fig.4.1, which are normally too deep to be thermally drained during normal TL applications. Such traps contribute to sensitization and non-linear growth of TL signal with dose, and can be problematic in TL dosimeters. Although such deep traps may be inaccessible thermally, the optical release of the charge carriers from these states is a real possibility in many materials depending on the chosen stimulation wavelength. This advantage of OSL over TL makes the possibility of using deep traps as a source of dosimetric information.

Fig.4.1 also shows the transition occurring during the OSL process. Transition $a$ represents the optical stimulation and transition $b$ the recombination yielding the OSL signal. Although retrapping in to deeper traps may occur $c$, by a suitable choice of wave length one can optically release $d$ the charge from these deeper levels either simultaneously with transition $a$ or subsequent to it. So loss processes due to retrapping and recombination can be considerably reduced. Many TL materials suffer from the problem of thermal quenching in which luminescence efficiency is decreased as the temperature is increased. Since OSL process is taking place at room temperature, the problem of thermal quenching can be avoided and we can expect a higher sensitivity for OSL process as compared with TL process.

**IV-2 SAMPLE PREPARATION**

OSL from Ce doped and undoped CaS was studied. Method of preparation of the samples is explained in detail in section 3 in chapter II.

**IV-3 EXPERIMENTAL TECHNIQUES**

The experimental set up used for the OSL studies is given in Fig.4.2. and its photograph is given in Fig.4.2.(a). It consists of 9 infra red LEDs arranged conically so as to concentrate radiation to 1.8cm$^2$ area of the heater plate.
Fig. 4.2
Experimental setup for OSL studies
Fig. 4.2(a)

Photograph of Experimental Setup used for OSL Studies
To ensure uniform current flow to all the LEDs they are connected in a series parallel combination\(^{(112)}\). The LED's connected on the printed circuit board made in the laboratory for this is given in Fig.4.3(a) and the circuit diagram is shown in Fig.4.3(b). 365 nm radiation from mercury source was used to excite the samples. By a shutter arrangement a photomultiplier tube (PMT) window can be exposed to the sample by simultaneously screening off the exciting radiation. The sample kept on the heater plate is heated to about 500\(^{0}\)C to release the electrons trapped earlier in it. After cooling, the sample is excited using UV radiation and kept for phosphorescence decay to be completed. The sample is then stimulated by using infrared radiation from the LED array. Emitted light is sensed by the PMT, output of which is amplified and plotted as a function of time using a strip chart recorder. The sample cell is designed in such away that the scattered light from the LED or any other source directly falling on the PMT is avoided.

Exposure of samples to the UV radiation is varied by changing the excitation time. OSL characteristics are also studied by changing the intensity of stimulant radiation keeping the radiation time fixed. In order to study the effect of dopant concentration on OSL studies are done by changing Ce concentration in the case of CaS:Ce

**IV-4 RESULTS AND DISCUSSION**

The observations made during the study are

1. OSL decays very fast in the case of undoped CaS while it lasts for more than one minute in the case of doped CaS, Fig.4.4.
2. There is an initial rise in emission before the decay sets in during OSL process. Fig.4.5.
3. Emission intensity of OSL increases with increasing duration of UV excitation. The increase is not linear. Fig.4.6.
4. Concentration quenching of OSL emission is observed when the concentration of the dopant is increased.
Fig. 4.3
(a) Printed circuit board for fixing the IR LED's
(b) Circuit diagram for connecting IR LED's
Fig. 4.4

OSL decay curves of (a) undoped CaS and (b) CaS doped with Ce
Fig. 4.5

Variation of OSL intensity with dopant (Ce) concentrations (mole%)

(1) 0.0175, (2) 0.035, (3) 0.175 & (4) 0.35
Variation of OSL emission intensity maximum with excitation time.

Fig. 4.6
5. Decrease of OSL emission exhibits three clear regions in the decay process corresponding to different decay constants. Fig.4.7. The decay constant values are given in Table 4.1.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Decay Constant</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$b_1$</td>
<td>$b_2$</td>
<td>$b_3$</td>
</tr>
<tr>
<td>CaS:Ce</td>
<td>0.25</td>
<td>0.53</td>
<td>0.95</td>
</tr>
<tr>
<td>CaS:Pr</td>
<td>0.30</td>
<td>0.60</td>
<td>0.61</td>
</tr>
<tr>
<td>CaS:Nd</td>
<td>0.30</td>
<td>0.50</td>
<td>0.48</td>
</tr>
<tr>
<td>CaS:Sm</td>
<td>0.32</td>
<td>0.29</td>
<td>0.64</td>
</tr>
</tbody>
</table>

6. OSL increases with intensity of stimulant radiation from the LED array. Stimulant radiation intensity is changed by changing the current flowing through the LED bank. Fig.4.8 shows the plot of OSL intensity versus LED current.

OSL decay curves of doped samples exhibit a higher initial rise time as compared to those of undoped ones. The initial rise time increases with dopant concentration. This is illustrated in Fig.4.5 in the case of CaS:Ce. When the phosphor is excited, some deeper traps also get filled in addition to shallow ones. During stimulation process, it is probable that the electrons in the deeper levels get detrapped and retrapped in the shallow levels. The time of retrapping obviously depend on the number of deeper levels, which in turn depend on the concentration of the dopant. This explains the enhancement of initial rise time with dopant concentration. Another interesting observation is that intensity of OSL emission decreases with increase in the atomic number of the dopant Fig.4.9. This is due to the non-radiative processes taking place in the sample.
Fig. 4.7

Logarithm of OSL intensity (\(\ln I\)) versus logarithm of time (\(\ln t\)) curves for
(1) CaS:Ce, (2) CaS:Pr and (3) CaS:Nd phosphors
Fig. 4.8

Variation of OSL emission intensity maximum with diode current
Fig. 4.9

Variation of OSL emission intensity of doped CaS with atomic number of the dopant.
during OSL emission. As the atomic number of the dopant increases such non-radiative relaxation become more predominant due to enhancement in the collision cross section between dopant and other species.

**IV-5 CONCLUSION**

Certain characteristics of optically stimulated luminescence emission from UV excited CaS:Ce phosphor samples were studied. Experimental setup was designed and fabricated for the study of OSL. It was found that phenomena like trapping and detrapping of the electrons characterize the OSL during the initial period. Heavy atom effect resulting in to decrease in OSL emission is one of the interesting observations.