ANALYTICAL STUDY OF PHOSPHORS FOR RADIATION DOSIMETRY

Pankaj Pathak¹, Manisha Singh², Pankaj Kumar Mishra³
¹,²,³Department of Applied Physics, Amity School of Engineering & Technology, Amity University, Gwalior (MP)
Email: pankaj2002@hotmail.com¹

Abstract
The main aim of the study is to analyse the different luminescence properties of phosphors synthesised by Combustion Synthesis (CS) Technique for Radiation Dosimetric Applications. Different electrical, optical and structural properties are seen in inorganic and organic materials in nano-crystallized form as compared to those in the bulk form. Out of these, the ones suited as phosphor host material show considerable size dependent luminescence properties when an impurity is doped in a quantum-confined structure. Pioneering studies of Thermoluminescence (TL) properties of nanostructure materials have produced encouraging results. With an objective of finding a new dosimetric material with higher sensitivity, good reproducibility, low hygroscopicity, and good response at high doses in radiotherapy and in mixed radiation fields and thereby advancing the dosimeters hence developing highly efficient materials, the present investigation is undertaken. Activated Nano/Micro phosphor in regulated condition can be obtained by employing different Synthesis methods. Luminescent Properties exhibited by so prepared Phosphors can be tailored by impurities present, type of dopants used, molar composition of dopants and methods employed for their Synthesis, and temperature of synthesis. The materials of required properties can be synthesized by employing, hydrothermal reaction, sol-gel synthesis, micro-emulsion synthesis and mechanochemical synthesis.

Key-words: Phosphors, Luminescence, Radiation Dosimetry

Introduction:
Thermally stimulated luminescence also termed as thermoluminescence (TL) is a powerful technique extensively used for dosimetry of ionizing radiations. TL dosimeter (TLD) materials presently in use are inorganic crystalline materials. They are in the form of chips, single crystals or microcrystalline size powder. The most popular of them are LiF: Mg, Ti, LiF: Mg, Cu, P, CaF₂, Li₂B₄O₇, CaSO₄: Dy, CaF₂:Dy. The TL results of the recently reported nanomaterials have revealed very imperative characteristics such as high sensitivity and saturation at very high doses. However, recent TL studies of different luminescent ceramic micro or nanomaterials showed that they have a potential application in radiation dosimetry. These materials spread over various applications such as medical imaging, high energy physics, and nondestructive testing. During the last two decades, numerous ceramic materials have been proposed to be used as radiation detectors. In order to provide efficient dosimeters for X or γ rays, the choice of the oxide matrix is crucial. The possibility to prepare nanocrystalline powder could allow the preparation of ceramics that could replace traditional thermoluminescent material. Another very attractive solution is the direct preparation of radiation detecting thin films. Thin detecting films are particularly valuable in fundamental spectroscopic studies when the absorption coefficient of the material is high or when the excitation energy is close to the absorption edge.

The commercially available Phosphors and their radiative efficiencies are tabulated in Table 1 & Table 2 [1-5]:

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Table 1: Different TLD dosimeter-based personnel monitoring (PM) systems being used for individual monitoring of radiation workers.

<table>
<thead>
<tr>
<th>System</th>
<th>Dosimeter type/Country where the PM system is being used</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harshaw TLD Badge (Thermo Electron Corp., USA + many other commercial systems in Europe)</td>
<td>LiF:Mg,Cu,P or LiF:Mg,Ti</td>
</tr>
<tr>
<td>Teledyne Isotopes, USA</td>
<td>CaSO4:Dy Teflon Tape Card: a) Korea, b) some labs in USA.</td>
</tr>
<tr>
<td>TLD badge system based on CaSO4:Dy teflon discs</td>
<td>a) India, b) Australia, c) Brazil</td>
</tr>
<tr>
<td>Panasonic TLD badge (Japan)</td>
<td>Li2B4O7:Cu &amp; CaSO4:Tm</td>
</tr>
<tr>
<td>Rados, Finland</td>
<td>Li2B4O7:Mn,Si</td>
</tr>
<tr>
<td>Vinca, Serbia and Montenegro</td>
<td>Li2B4O7:Cu,Ag,P MgB4O7:Dy,Na</td>
</tr>
<tr>
<td>Landauer Inc., USA</td>
<td>Al2O3:C OSL badge</td>
</tr>
<tr>
<td>Chiyoda Technol Corporation, Japan</td>
<td>Silver-activated phosphate glass dosimetry system: Japan and France</td>
</tr>
</tbody>
</table>

Table 2: General characteristics of some commercially available thermoluminescent dosimeters relevant for personnel dosimetry.

<table>
<thead>
<tr>
<th>TLD type</th>
<th>Effective atomic number Zeff</th>
<th>Main peak (°C)</th>
<th>Emission maximum (nm)</th>
<th>Relative sensitivity</th>
<th>Fading (at 25°C) for storage in dark</th>
<th>Useful dose Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiF:Mg,Ti</td>
<td>8.2</td>
<td>200</td>
<td>400</td>
<td>1</td>
<td>5%/year</td>
<td>20 μGy-10 Gy</td>
</tr>
<tr>
<td>LiF:Mg,Cu,P</td>
<td>8.2</td>
<td>210</td>
<td>400</td>
<td>25</td>
<td>5%/year</td>
<td>0.2 μGy-10 Gy</td>
</tr>
<tr>
<td>Li2B4O7:Cu</td>
<td>7.4</td>
<td>205</td>
<td>368</td>
<td>2</td>
<td>10%/2 months</td>
<td>10 μGy-103 Gy</td>
</tr>
<tr>
<td>MgB4O7:Dy</td>
<td>8.4</td>
<td>190</td>
<td>490</td>
<td>10</td>
<td>4%/month</td>
<td>5 μGy-50 Gy</td>
</tr>
<tr>
<td>Mg2SiO4:Tb</td>
<td>11</td>
<td>200</td>
<td>380-400</td>
<td>negligible</td>
<td>10 μGy-1 Gy</td>
<td></td>
</tr>
<tr>
<td>CaSO4:Dy</td>
<td>15.3</td>
<td>220</td>
<td>480, 570</td>
<td>30</td>
<td>1%/2 months</td>
<td>2 μGy-10 Gy</td>
</tr>
<tr>
<td>CaSO4:Tm</td>
<td>15.3</td>
<td>220</td>
<td>452</td>
<td>30</td>
<td>1-2%/2 months</td>
<td>2 μGy-10 Gy</td>
</tr>
<tr>
<td>Al2O3:C</td>
<td>10.2</td>
<td>190</td>
<td>420</td>
<td>60</td>
<td>5%/year</td>
<td>1 μGy-10 Gy</td>
</tr>
</tbody>
</table>

Analysis:

Transitions of electrons between the valence band and the conduction band are allowed and they produce free electrons in the conduction band and free holes in the valence band[6]. The energy difference between the two bands is denoted by the band-gap energy $E_g$ (as shown in Figure 1). The transition of electrons directly from a metastable state to ground state is forbidden. The metastable state represents a shallow electron trap and electrons returning from it to the excited state require energy. This energy can be supplied in the form of optical radiation (photo stimulation) or as heat (thermal stimulation). The probability ($p$) per unit time that a trapped electron will escape from a...
metastable state to an excited state is governed by the Boltzmann equation.

\[ p = s \cdot \exp\left(-\frac{E}{kT}\right) \]  

(1)

Where \( s \) is the frequency factor (s\(^{-1}\)), depending on the frequency of the number of hits of an electron in the trap which can be considered as a potential well, \( E \) is the thermal activation energy required to liberate a trapped charge carrier called trap depth (eV), \( k \) is Boltzmann's constant and \( T \) is the absolute temperature (K).

First, the intensity of thermo luminescent emission does not remain constant at constant temperature, but decreases with time and eventually ceases altogether. Second, the spectrum of the Thermo luminescence is highly dependent on the composition of the material and is only slightly affected by the temperature of heating. The Thermo luminescence emission mainly is used in solid state dosimetry for measurement of ionizing radiation dose. Initially radiation dose was given by simple mechanism of thermo luminescence.

Figure 1

An optical storage phosphor or material is defined as to be a system which undergoes electronic or structural change that allows an optical readout of radiation exposure. luminescent phosphors where the radiation induced centres are metastable or stable and can be repetitively read out by photoexcitation[7]. The impurity incorporation transfers the dominant recombination route from the surface states to impurity states. If the impurity-induced transition can be localized as in the case of the transition metals or the rare earth elements, the radiative efficiency of the impurity- induced emission increases significantly[8]. Dosimetric characteristics of the phosphor like Thermo luminescence glow curve, TL emission spectra, dose–response, fading studies, reproducibility and reusability studies are the factors which measures the efficiency and application potential of synthesized phosphors[9].

**GLOW CURVES:**

Thermo luminescence properties to be considered in choosing a suitable dosimetric system for radiation therapy dosimetry are linearity between radiation dose and response, sensitive to the signal, TL glow curve, acceptable accuracy and precision, and a good characterized relationship between the dosimeter response in the medium in which dose is to be measured and the calibration radiation field. Further its radiation application was measured and analysed by reading the following parameters:

**TL glow curve:**

Shallow traps, those nearest to the conduction band, are easily emptied at room temperature (RT), leading to measurable fading of the TL signal. The dosimetric traps require somewhat more energy to release trapped electrons, normally forming the peak within which maximum TL yield is obtained and hence are used as the principal peak in dosimetric evaluation. The deep seated traps require appreciable energy in order to be emptied, obtained by high temperature annealing. The maximum peak of TL intensity increase with increasing the irradiation dose.
TL response
One of the important characteristic to be a good Thermo luminescence dosimeter is linear relationship between TL emission and the absorbed dose. The particular Thermo luminescence material gives the great effect to the linearity range. Generally, the response of TL phosphors is linear at low absorbed dose value than becomes supralinear and finally saturates at high values.

Energy dependence
The energy response should be flat, i.e., the system calibration should be independent of energy over a certain range of radiation qualities. In reality, the energy correction has to be included in the determination of the quantity Q for most measurement situations.

Sensitivity
TL sensitivity can be define as the amount of light released by phosphor per unit of radiation exposure. There are many factors that affect to TL sensitivity dosimeter such as kind and concentration of activators, system of the readout, heating rate, etc. The Figure 2 shows the characteristic of TL-glow curve with the function of temperature & relative Intensities of Phosphors.

Thermoluminescence glow curves of CaSO$_4$:Dy phosphors

![Thermoluminescence glow curves of CaSO$_4$:Dy phosphors](image)

Figure 2

Results and Discussion:
After the synthesis and characterization of formed phosphor exhibits thermo luminescence when heated after exposure to radiation [eg.gamma rays (Co-60 teletherapy machine), X-rays and particulate radiation (Linear accelerator with 4, 6,15MV photon energies & 4-18Mev electron energies)], the intensity of the thermo luminescence being in proportion to the dosage of radiation of the phosphor, i.e., the dosage of radiation to which the phosphor is exposed. The TLD reader heats the TL element, measures the thermoluminescence emanating from the element by means of a light measuring circuit including a photomultiplier tube, and displays a reading of the dosage of radiation corresponding to the measured intensity of the thermo luminescence in an analog or digital value. TL glow curves will be recorded for all the samples with various treatments. Characteristic of natural, natural plus irradiation induced TL and together with annealed sample The characteristics of formed phosphors will give the following properties when analyzed with TLD-Reader (a) Trap Depth (b) Value of frequency facto: (c) Order of Kinetics (d) The trap density of Dopants & Co-Dopants changes will be analysed: (e) TL peak temperature (Tm):

APPLICATIONS:
One of the important applications of TL Phosphors has been in the field of medical physics for their application in radiodiagnosis,
nuclear medicine and radiotherapy. The rmoluminescent dosimeters (TLDs) have become popular in these fields due to their high sensitivity, miniature size, tissue equivalence, high stability to environmental conditions, low TL fading, reusability, linear dose response and sufficient precision and accuracy.

References:


9. Thermoluminescence dosimetric characteristics of thulium-doped Zn(BO₂)₂ phosphor; Annalakshmi, O. T.; http://dx.doi.org/10.1016/j.jlumin.2013.09.042