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Thermoluminescence studies of Eu^{3+} doped Calcium Lanthanum borate phosphor

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Abstract

$Ca_3La_2(BO_3)_4$ phosphor doped with Eu^{3+} have been synthesized by high temperature solid-state reaction method. The crystalline structure and elemental analysis of these phosphors are carried out by using X-ray powder diffraction (XRD) and Energy dispersive X-ray spectroscopy (EDS) techniques. The thermoluminescence (TL) properties have been investigated. For TL studies, samples were γ-irradiated with 1450 Gy dose rates using ^{60}Co source. TL of these phosphors showed a sharp glow peak with maxima at 394 K. Incorporation of europium activator in $Ca_3La_2(BO_3)_4$ phosphor resulted in the increase of peak intensity. The trap parameters viz., order of kinetics (b), activation energy (E) and frequency factor (S) associated with the most intensive glow peak of $Ca_3La_2(BO_3)_4:Eu^{3+}$ phosphor were determined using glow curve shape (Chen's) method.

Keywords: Phosphor, XRD, EDS, $Ca_3La_2(BO_3)_4:Eu^{3+}$ Phosphor, Thermoluminescence

1. INTRODUCTION

The study of new thermoluminescent (TL) materials to be used as TL dosimeters has made necessary to have a deeper knowledge of the trapping energy as well as of the average time the electron stays in its trap. This information can be obtained from the glow curve analysis which is obtained after exposing the material to ionizing radiation in order to excite the electrons from the valence band to the conduction band and back to metastable states in the forbidden band over the Fermi level. Then, by heating, those electrons can be released from their traps and recombine with trapped holes

emitting light. Hence, TL is considered as a thermally stimulated process [1]. The glow curve can exhibit various peaks (maximum intensity) originated from different trapping states. There are many different methods for determining the trapping parameters [2] (i.e., the activation energy or trap depth, the kinetic order and the frequency factor). Most of the existing methods of obtaining the frequency factors make use of the determination of the trap depth (activation energy) relaying on some assumptions about the order of the kinetics. This causes inconsistencies in the reported values of the trapping parameters due to appreciable differences in the activation energy obtained by different methods. Then, it is necessary to use a method independent of the determination of the trap depth.

The TL characteristics of borate compounds have been reported such as un-doped and Ce-doped BaB_4O_7 [3], Tb^{3+}-doped $Ba_2Ca(BO_3)_2$ [4], un-doped and Cu- and Mn-doped $K_2B_4O_7$ [5], MgB_4O_7:Dy,Na [6], SrB_4O_7:Dy [7], rare-earth-doped $Sr_2Mg(BO_3)_2$ [8], $Li_2B_4O_7$:Cu,In [9,10] and BaB_4O_7: Dy [11] these results provide useful information concerning the defects and trap structure that are helpful in the search for new borate TLD materials. In this paper, we report the method making use of isothermal decay of the TL was studied to determine the trapping parameters of $Ca_3La_2(BO_3)_4$:Eu^{3+} phosphor by using the glow curve shape method.

2. EXPERIMENTAL

The Phosphor materials of composition $Ca_3La_2(BO_3)_4$ [CLB] doped with europium were synthesized by the solid state reaction method. Stoichiometric amounts of AR grade $CaCO_3$, La_2O_3, Eu_2O_3 and H_3BO_3 (99% Sigma Aldrich Chemicals) were thoroughly mixed and ground together with ethanol in an agate mortar for 5 hours to give homogenous mixture. The resultant powders were initially dried at 100^oC for 1 h, kept in an alumina boat and heated at 1200^oC in air for 3 hrs. Later, the temperature is brought down to 950^oC and the samples were held at that temperature for 1 hour in air. These samples were rapidly cooled down to room temperature and were grinded to get fine powder for further studies.

In order to characterize the phase purity and structure of the as-prepared $Ca_3La_2(BO_3)_4$: Eu^{3+} samples have been studied by X-ray powder diffraction. Powder X-ray diffractograms were recorded on a PANalytical X'pert Analytical X-ray diffractometer (Spectris Instrumentation and Systems, Shanghai, China) using Nickel filtered Cu-Kα radiation of wave length 1.5406 A^o in the 2θ range of 10 ~ 80^0 with a step size of 0.04 ^{o}/ sec. The operation voltage and current of the instrument were maintained at 40 kV and 30 mA respectively. The compositions of the phosphor samples were obtained from energy dispersive spectrum (EDS). The EDS was

attached to the HITACHI S-3700N model instrument. All measurements were recorded at room temperature. Thermoluminescence Analyzer system (type 1007) supplied by Nucleonix Systems Private Ltd., Hyderabad, India. offering an irradiation volume approximately 1000 cc had 2370 curies ^{60}Co source, used for γ-irradiation. The glow curves were recorded by heating the sample at a uniform rate of 2 K Sec-1 with the help of a temperature controller.

3. RESULT AND DISCUSSIONS

3.1. Powder XRD

The powder X-ray diffractograms of CLB are shown in fig.1 and the observed d-lines are indexed for higher concentration of Eu^{3+} . The CLB belongs to the orthorhombic system with space group *Pnam*, and unit cell parameters are show as following: a = 7.279 Å, b = 16.417 Å, c = 8.654 Å. All the compounds are very close to that of reported values for CLB [12]. It is indicated that the doping Eu^{3+} ions do not form new phases in the synthesis process.

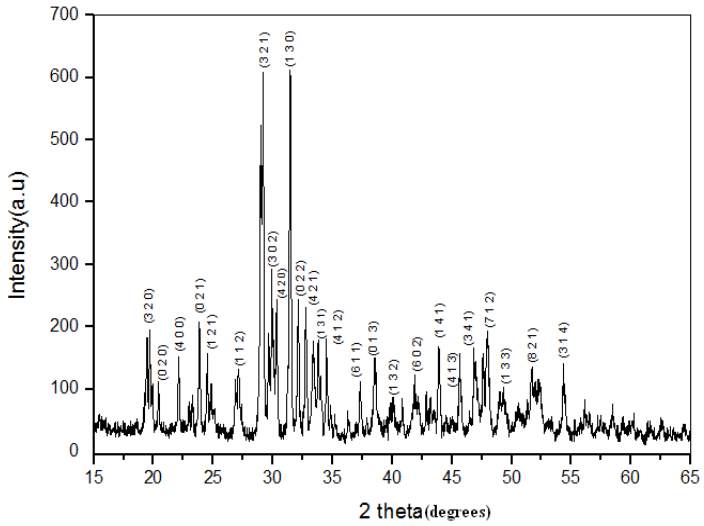

Fig 1 XRD of $Ca_3La_{1.9}Eu_{0.1}(BO_3)_4$ phosphor

3.2. Energy Dispersive Spectrum

The energy dispersive spectrum analysis of 0.1 mol % Eu^{3+} doped CLB sample peaks corresponding to Ca, La, Eu, B and O. The EDS pattern confirms the presence of europium in the CLB powders and its wt% is nearly equal to the doped value of Eu in CLB.

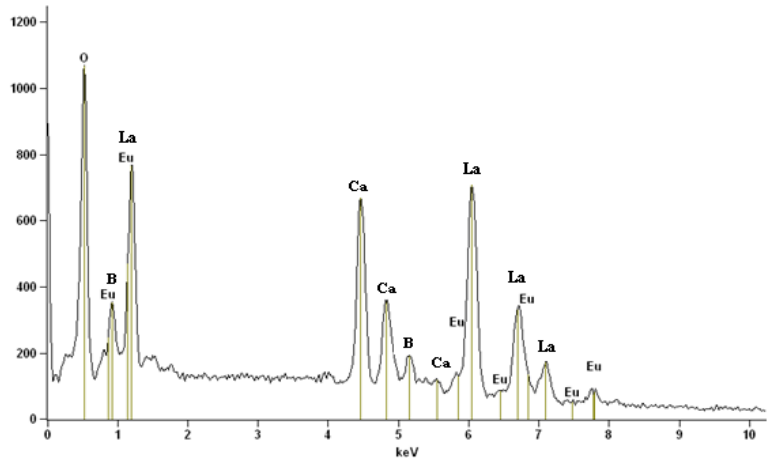

Fig 2. EDS of $Ca_3La_{1.9}Eu_{0.1}(BO_3)_4$ phosphor

3.3.1 Thermoluminescence of $Ca_3La_2(BO_3)_4$: Eu^{3+}

The observation of Eu^{3+} afterglow in the $Ca_3La(BO_3)_4$ host, it is necessary to take the TL spectrum into consideration. Fig. 3 represents the TL glow curve of $Ca_3La_{1.9}Eu_{0.1}(BO_3)_4$ phosphor. During this study, glow curves of Eu^{3+} doped $Ca_3La(BO_3)_4$ samples were recorded at dose rate of γ-irradiation 1400 Gy at room temperature. A strong TSL glow peak is observed at 394 K. It is observed that the intensity of this glow peak is found to increase with the increase of Eu concentration.

3.3.2. Glow curve shape method

The glow curve of $Ca_3La_{1.9}Eu_{0.1}(BO_3)_4$ phosphor shown in Fig.3. The method based on the shape of glow curve proposed by Chen was used to verify the trapping parameters. To determine these parameters the following shape parameters were determined: the total half intensity width ($\omega = T_2 - T_1$), the high temperature half width ($\delta = T_2 - T_m$), the low temperature half width ($\tau = T_m - T_1$), where Tm is the peak temperature and T_1 and T_2 are temperature on either side of T_m corresponding to half peak intensity. In order to calculate the trapping parameters associated with the 394°C glow peak by glow curve shape method using chen's method [13,14] and given in Table 1.

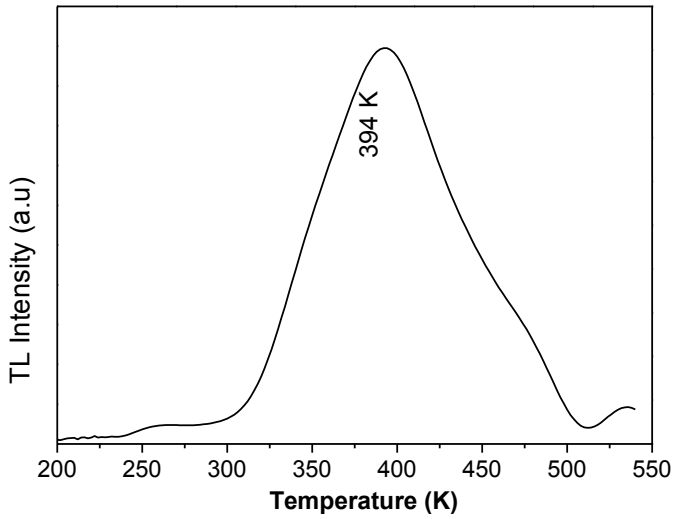

Fig 3 TL glow curve of $Ca_3La_{1.9}Eu_{0.1}(BO_3)_4$ phosphor

Sample	Dose (Gy)	T_m ±2K	τ (K)	δ (K)	Ω (K)	$^{\mu}g$	Activation Energy (eV)				S (Sec^{-1})
							$^{E}\tau$	$^{E}\delta$	$^{E}\omega$	^{E}av	
$Ca_3La_2(BO_3)_4$	1320	394	32	44	76	0.579	0.677	0.648	0.659	0.661	2.704×10^7
$Ca_3La_{1.9}Eu_{0.10}(BO_3)_4$		394	35	46	81	0.568	0.596	0.596	0.596	0.596	3.523×10^6
$Ca_3La_{1.9}Eu_{0.10}BO_3)_4$		394	31	42	73	0.575	0.700	0.671	0.683	0.684	5.522×10^7
$Ca_3La_{1.9}Eu_{0.10}(BO_3)_4$		394	35	49	84	0.583	0.609	0.591	0.598	0.599	3.860×10^6

4. CONCLUSIONS

In this work, we report the chemical synthesis of a new $Ca_3La_2(BO_3)_4$ thermoluminescent phosphor. The experimental evidence presented shows that these novel materials exhibit very good thermoluminescence properties, as to be considered for the development of new dosimeter phosphor materials.

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