Preparation, characterization and luminescent properties of LiBO₂:Tb³⁺ green emitting phosphor for solid state lighting

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Abstract

Introduction: In our work we report the preparation, characterization and luminescence mechanism of Tb³⁺ activated LiBO₂; green emitting phosphor for solids state lighting. Tb³⁺ has been extensively used as green-emitting activator for luminescent phosphor materials; the excitation and emission spectra indicate that the prepared lithium borate based phosphor can be effectively excited by 353 nm, to showing a bright green emission at 545 nm corresponding to the f→f transition of Tb³⁺ ions. All the characteristics indicated that LiBO₂:Tb³⁺ is an excellent phosphor for solid state lighting.

Keywords: Phosphor, Photoluminescence, Solid state lighting, CIE, XRD, SEM

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Received Date: 21/08/2014  Accepted Date: 01/09/2014

INTRODUCTION

Generally in most of the phosphor rare earth activated borates and oxyborates have been extensively investigated as host lattice materials for several applications. They possess exceptional properties such as high ultraviolet transparency, outstanding optical damage threshold, broad band gap and high constancy which make them attractive for several convenient practical applications.¹,² The combustion synthesis method is found to be extremely useful for the generation of light from the luminescent materials particularly for silicates, oxides, borates and aluminates.³,⁴ The particle size of the product prepared by using this method is uniform and narrow. Furthermore the method is very simple, easy and time consuming. Thus for the synthesis of lithium borates we have attempted combustion synthesis method. From the literature it is found that borate compounds are known for their wide band gap. They are the preeminent hosts for different activators and find numerous exciting applications, e.g. barium borate is a non-linear optical (NLO) material used for laser harmonic generation⁵ and for forming of green component of tricolor TV phosphor⁶ and solid state lighting. In borate compounds boron atom is synchronized by oxygen atoms to form a mixture of atomic groups that affect the physical and optical properties⁷. LiBO₂:Tb³⁺ phosphor is previously reported by O. Dincer and co worker. They studied the luminescence investigation of Tb³⁺ ion by solid state method.⁸ Within shell because of their distinct 4f transitions rare earth activated phosphor has attracted more and more attention⁹. With due of their emission properties rare earth activators considered as the most important and promising activators for phosphors for solid state lighting and extra other applications.³,⁸ Tb³⁺ ion has a sharp emission in the region of 541 to 545 nm with high emission intensity which is nearer to the three basic primary color centers value of theoretical principle wavelength; therefore for different phosphors it is used as the most important green-emitting activators¹⁰,¹¹. In some extent borates are steady chemical compound which gives without serious problems to attempts doping them with TSL sensitizers⁸. A necessity for understanding about the results of Tb³⁺ activator has motivated our work and the exact potentialities of these phosphor materials for the applications in solid state lighting.⁸ In our work, LiBO₂:Tb³⁺ prepared phosphor was successfully prepared by using combustion synthesis methods and then it was further characterized by X-ray diffraction (XRD),
scanning electron microscopy (SEM), photoluminescence (PL) techniques and chromaticity behavior.

**MATERIALS AND METHODS**

**Experimental**

During this synthesis, the stoichiometric amounts of high purity starting materials, Li$_2$NO$_3$, Ca(NO$_3$)$_2$4H$_2$O, H$_3$BO$_3$, C$_2$H$_5$NO$_2$, were mixed thoroughly in agate mortar for about 15 to 20 min with drop by drop addition of double distilled water, so that the paste was formed. A stock solution of stoichiometric amount of dopant prepared by dissolving oxides of terbium into concentrated HNO$_3$ with double distilled water was then mixed in paste. The prepared paste was kept in heating some time till we got a clear and completely dissolved solution. Then again crush it for 10 min then as prepared solution was then transferred into the pre-heated furnace (550 °C) after warming it for 5 min. The self heat generating redox reaction was completed and the fine powder of LiBO$_2$: Tb was finally obtained. During the combustion synthesis proper molar ratios of fuel and oxidizers depending on the moles of precursors are very necessary for evacuation of unwanted various molecules so that fine powders of required product could be finally obtained. Therefore the reaction is to be exactly balanced.

**OBSERVATION AND RESULTS**

**X-ray Diffraction**

The X-ray diffraction pattern of the prepared host material powder was recorded with the help of X-ray diffractometer by Cu-Kα radiation (1.54060 nm) with scanning step time of about 10.3377s duration. The scan type observed was continuous. The XRD pattern of the LiBO$_2$ compound is shown in Fig. 1.

**SEM analysis**

The SEM of the combustion synthesized LiBO$_2$:Tb$^{3+}$ phosphors shown in fig 02. With due progress of huge amount of gases a well expected sharp chip shape morphology is observed. In combustion synthesis process instantaneous oxidation with reduction process done$^{12}$.

**DISCUSSIONS**

**Photoluminescence Study**

For phosphors material because of their emission and excitation characteristics in ultraviolet and visible region rare earth (RE) elements have been considered as the most significant and promising dopant$^{13}$. Tb$^{3+}$ ion shows a sharp emission around 541nm-545 nm with elevated strength which is very nearer to the theoretical ideal wavelength suitable for the green element of three primary basic color centers, therefore it is the most significant activator for obtaining green emitting luminescence for several phosphors$^{14,15}$. Basically on particle shape and size, the photoluminescent properties of phosphors depend which optimized to get the utmost quantum effectiveness through absorption of energy$^{16}$. The emission and excitation spectra of LiBO$_2$:Tb$^{3+}$ microcrystalline phosphors shown in figure 03 and 04. Upon the excitation of 353 nm, the emission spectrum exhibits five major emission bands. The emission peaks are located at 440, 491, 545, 586 and 625 nm, corresponding to the $^5$D$_3$$\rightarrow$F$_{4}$, $^5$D$_3$$\rightarrow$F$_{6}$, $^5$D$_4$$\rightarrow$F$_{5}$, $^5$D$_4$$\rightarrow$F$_{4}$ and $^5$D$_4$$\rightarrow$F$_{3}$ typical transitions of Tb$^{3+}$, respectively. The strongest one appears at 545 nm. The excitation spectrum was obtained by monitoring the emission of Tb$^{3+}$ at 545 nm. The overall excitation spectrum can be divided into two parts. One band in the range from 240 to 280 nm represents the 4f$^8$$\rightarrow$4f$^6$5d$^1$ broad band absorbability. The other band in the range from 350 to 400 nm, and the excitation peaks located at 353,372 and 381 nm correspond to the f-f transitions of Tb$^{3+}$, respectively. The excitation and emission spectra indicate that this phosphor can be effectively excited by UV LED. The green emission observed for the excitation 353 nm explains that as we increased the trivalent terbium...
concentration the emission intensity considerably increases. LiBO$_2$:Tb$^{3+}$ phosphor a strong green emission located at 545 nm, which indicate that it could be a good green phosphor candidate for creating white light in solid state lighting.

**Chromaticity Behavior**

In general specifications of lighting refer to color in terms of the 1931 CIE chromatic color coordinates which helps to explain that the human visual system prefers three basic primary colors: red, green, and blue$^{13,14}$. In color space the color of any light source can be represented on the (u, v) coordinates$^{19}$. The CIE chromaticity diagram of LiBO$_2$:Tb$^{3+}$ shown in Fig. 5. The coordinates of the LiBO$_2$:Tb$^{3+}$ phosphor arc, in green color (Cx = 0.265, Cy =0.723) shown in Fig. 8. Which indicates that the position of present phosphors founds are very close to the CIE diagram edge, which reposts the high color purity of these phosphors. By connecting these points [including a white light point (0.31, 0.32)], the intermediate compositions can generate white light for solid state lighting.

**CONCLUSION**

In our work LiBO$_2$:Tb$^{3+}$ green emitting phosphor was synthesized by using combustion synthesis method. The prepared sample is good agreement with the standard JCPDS file available 01-076-2212. SEM micrographs showed the existence of tiny particles and the crystalline grain was observed in typical, in the sub µm range. The study of spectroscopic properties has provided essential information about the luminescence features of Tb$^{3+}$ ions in lithium borate. The emission band observed at 545nm of CIE coordinates (Cx = 0.265, Cy = 0.723) is attributed to $5d_{4s}$→$^1I_{5/2}$ transition of Tb$^{3+}$ ion, which corresponds to the deep green region of the spectrum excited at 353nm in mercury free excitation. Thus, our prepared phosphor has probable green emitting phosphor for solid state lighting.

**ACKNOWLEDGMENT**

One of the author, VRP thankful to, Dr. K.R. Dixit (Director), Dr. P.A. Potdukhe (Principal). Shri. Santosh Shrigadiwar (Registrar), Namdeo Rao Poreddiwar College of Engineering and Technology, Gadchiroli for their kind support during this research work.

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