

Preparation and Photoluminescence Properties of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$ Phosphor

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Abstract— In the present work Photoluminescence behavior of Sm^{3+} doped $\text{CaAl}_2\text{B}_2\text{O}_7$ (CABO) phosphor has been reported for the first time. The series of red-emitting $\text{Ca}_{(1-x)}\text{Al}_2\text{B}_2\text{O}_7:x\text{Sm}^{3+}$ phosphors has been synthesized by using solution combustion synthesis method. Its phase, photoluminescence properties, surface morphology and concentration quenching mechanism were investigated. This phosphor can be effectively excited within the broad near ultraviolet wavelength region from 300 to 450 nm and shows efficient Green-Red emission. The emission spectra exhibit three emission bands corresponding to $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ (564 nm), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ (601 nm), and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (649 nm) transition under the 402 nm excitation wavelength. It has been shown that the 5 mol% doping concentration of Sm^{3+} ions in (CABO) phosphor is optimum, and beyond 5 mol % concentration quenching occurs. The physical mechanism of concentration quenching can be explained by the exchange interaction of Sm^{3+} ions, and the critical transfer distance was determined to be 13.71 Å. The results are promising for Sm^{3+} doped (CABO) phosphor in view of the requirement for solid state lighting.

Keywords— Alumino-borate; combustion synthesis; photoluminescence.

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1. INTRODUCTION

In recent years, there have been reports on the use of alkaline earth aluminates as they have very excellent optical properties which make them part of almost every-display devices. Optical properties of rare earth doped alkaline earth aluminates have been studied in detail because of their wide applications in solid-state optical devices. Among trivalent rare earth ions, the Sm^{3+} ion is one of the most interesting ions with respect to its photoluminescence properties due to different quenching channels exhibited by the $^4\text{G}_{5/2}$ emitting level [1]. However, Sm^{3+} doped materials are less studied compared to the other [2-4] trivalent rare earths ions and in particular, the spectroscopic studies of Sm^{3+} in aluminates host are rarely reported. In the present work we discussed the Sm^{3+} activated $\text{CaAl}_2\text{B}_2\text{O}_7$ (CABO) phosphor with different mole ratios of Sm^{3+} ions synthesized by a solution combustion method. The synthesized materials were characterized through XRD and the morphological study was done through FE-SEM images. After synthesis and characterization of the phosphors, the photoluminescence properties were studied using Spectrofluorometer at room temperature.

2. MATERIALS AND METHOD

The powder samples of Sm^{3+} activated $\text{CaAl}_2\text{B}_2\text{O}_7$ (CABO) phosphor were prepared by a solution

combustion technique. In our previous work [5-6], many borate host materials were successfully synthesized using this technique. The stoichiometric amounts of high purity starting materials, $\text{Ca}(\text{NO}_3)_2$ (A.R.), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (A.R.), Eu_2O_3 (high purity 99.9%), H_3BO_3 (A.R.), $\text{CO}(\text{NH}_2)_2$ (A.R.) have been used for phosphors preparation. The stoichiometric amounts of the ingredients were thoroughly mixed in an Agate Mortar with adding little amount of double distilled water. The materials then transferred into china basin and heated on heating menthol at about 70°C so as to obtained clear solution. The solution was then introduced into a pre-heated muffle furnace maintained at temperature 550 °C for combustion. The solution boils; foams and ignites to burn with flame which gave a voluminous, foamy powder. Following the combustion, the resulting foamy samples were crushed to obtain fine particles and then annealed in a slightly reducing atmosphere provided by burning charcoal at temperature 750°C for 2 hr and suddenly cooled to room temperature. The prepared materials were characterized by powder XRD. Powder X-ray diffraction measurements were taken on Rigaku Miniflex II X-ray Diffractometer and compared with the ICDD files. Surface morphology of the calcined powder sample was observed by Field effect scanning electron microscopy (FE-SEM). PL & PLE measurements at room temperature were performed on Hitachi F-7000 spectrofluorometer with spectral resolution of 2.5 nm.

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3. RESULT AND DISCUSSION

3.1 X-Ray Diffraction Pattern

The X-ray diffraction (XRD) technique have been used to identify the phase composition, structure and their crystallinity with CuK α radiation ($\lambda = 1.5405 \text{ \AA}$). Fig. 1 shows the XRD pattern of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$ phosphor. The XRD-pattern of the as prepared phosphor powder shows good agreement with standard ICDD File no.00-019-0206.

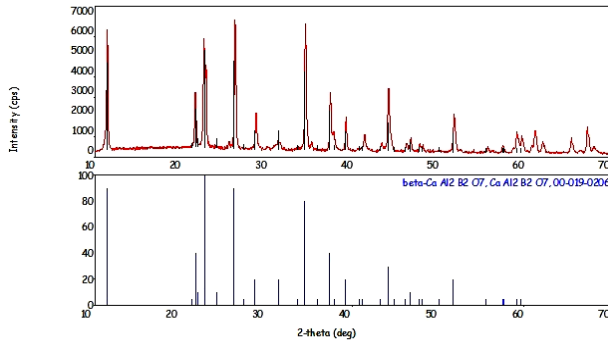


Fig. 1: X-ray powder diffraction powder of $\text{CaAl}_2\text{B}_2\text{O}_7$ host phosphor.

3.2 FE-SEM Images of Phosphor Powders

The SEM photographs of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$ powder produced by solution combustion method is as shown in Fig. 2. The shape of the particles was observed to be irregular with agglomerate phenomenon.

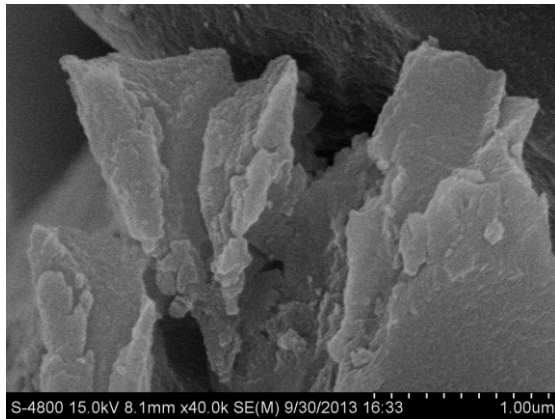


Fig. 2: FE-SEM images of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$

3.3 Photoluminescence of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$

To study the photoluminescence characteristics, the excitation spectrum was recorded for Sm^{3+} doped $\text{CaAl}_2\text{B}_2\text{O}_7$ phosphor by monitoring the emission at 601 nm as depicted in Fig. 3. Among the observed excitation bands, the band centered at 402 nm due to $4f \rightarrow 4f$ inner shell transition of Sm^{3+} ion is very sharp and intense. Upon this excitation, the photoluminescence spectra for different concentrations of Sm^{3+} doped $\text{CaAl}_2\text{B}_2\text{O}_7$ phosphors were recorded in the spectral range 450–750

nm. These spectra exhibit three emission bands centered at 564, 601 and 649 nm corresponding to the $^4\text{G}_{5/2} \rightarrow ^6\text{H}_J$ ($J=5/2, 7/2, 9/2$) transitions of Sm^{3+} ions, respectively. Among the observed emission transitions, the $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{11/2}$ transitions are purely electric dipole in nature ($\Delta J \leq 6$) whereas the other two transitions ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2, 7/2}$) contain both electric and magnetic contributions ($\Delta J=0, \pm 1$) [7]. The emission band corresponding to the $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ transition possesses highest intensity. The luminescence intensity of Sm^{3+} in $\text{CaAl}_2\text{B}_2\text{O}_7$ phosphors increases with the increase of its concentration, reaching a maximum at 5.0 mol% and then decrease for further rise of Sm^{3+} concentration.

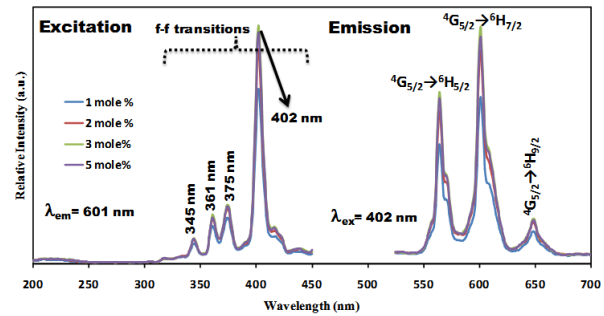


Fig. 3: Excitation and Emission spectra of Sm^{3+} in $\text{CaAl}_{2(1-x)}\text{B}_2\text{O}_7 : x\text{Tb}^{3+}$ ($x = 0.01, 0.02, 0.03, 0.05$).

According to Blasse [8], the critical transfer distance at optimum concentration can be calculated using Eq. (1):

$$R_c = 2\left(\frac{3V}{4\pi\chi_c N}\right)^{1/3} \quad (1)$$

Where χ_c is the optimal concentration, V is the volume of the unit cell, N is the number of cations in the unit cell. The values of V and N of $\text{CaAl}_2\text{B}_2\text{O}_7$ phosphor are 404.8 \AA^3 and 6. Using the optimal concentration χ_c , the critical transfer distance (R_c) for Sm^{3+} doped CABO was approximately found to be 13.71 \AA . The $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$ phosphor exhibit intense green-red luminescence with

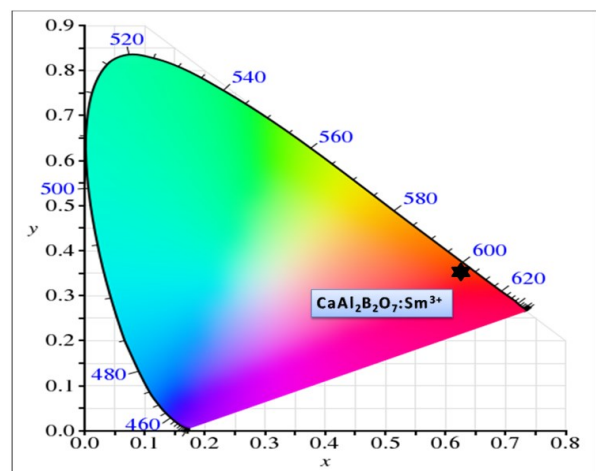


Fig. 4: CIE chromatic point and spectral locus of XY-coordinates of emission wavelength of $\text{CaAl}_2\text{B}_2\text{O}_7:\text{Sm}^{3+}$.

Commission International de l'Eclairage in (CIE) chromaticity coordinates ($x=0.63$, $y=0.36$) which are duly located in the red-green region of CIE chromaticity diagram shown in Fig. 4[9].

4. CONCLUSION

Sm^{3+} activated $\text{CaAl}_2\text{B}_2\text{O}_7$ phosphor was successfully synthesized by using solution combustion technique. The XRD patterns confirmed its phase structure and the FE-SEM images showed the closely packed particles with agglomerate phenomenon. The PL emission spectrum of Sm^{3+} ions at 402 nm excitation gives three bands centered at 564, 601 and 649 nm which corresponds to the $^4\text{G}_{5/2} \rightarrow ^6\text{H}_J$ ($J=5/2, 7/2, 9/2$) transition of Sm^{3+} ions as the surrounding offers high frequency vibrations. Based on the theoretical calculations, it is found that the exchange interaction is the major mechanism for concentration quenching of CABO phosphor and the critical transfer distance was found 13.71 Å. Thus this phosphor may be useful for solid state lighting and LED applications.

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