



# Luminescence properties of a novel orange–red $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$ phosphor for near-UV pumped W-LEDs



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## ABSTRACT

A new orange–red  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  phosphor was synthesized by conventional high temperature solid-state reaction method. Its crystal structure, luminescence properties were investigated. This phosphor can be effectively excited by 394 nm near ultraviolet light and exhibit bright orange–red emission with the emission peaks located at 581, 591 and 614 nm corresponding to the  $^5\text{D}_0 \rightarrow ^7\text{F}_j$  ( $j=0-2$ ) transition of  $\text{Eu}^{3+}$  ions, respectively. The optimum  $\text{Eu}^{3+}$  ions concentration in  $\text{CaBi}_2\text{B}_2\text{O}_7$  is 17.0 mol% and the critical transfer distance is determined to be 15.13 Å. Moreover, the CIE chromaticity coordinate and the color purity performances of  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  were also discussed. The present work suggests that the  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  phosphor is a potential orange–red material for the application in the near-UV pumped white light emitting diodes.

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## 1. Introduction

In recent years, white light emitting diodes (W-LEDs) have been considered as a promising technology for next generation solid-state lighting systems due to their unmatched advantages, such as high brightness, low power consumption, longer lifetime, and high color rendering index (CRI) [1–3]. The commercial way to approach white light is by using the blue InGaN chip combined with a broad-band yellow–emitting  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  phosphor [4–6]. However, this method results in low CRI and high correlated color temperature (CCT) due to the deficiency of red emission in the visible spectrum. To solve this problem, an alternative way has been proposed is based on a combination of near ultraviolet (near-UV) LEDs with tri-color (red, green and blue) phosphors [5,7,8]. For example,  $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$ , is for blue component,  $(\text{Sr}, \text{Ba})_2\text{SiO}_4:\text{Eu}^{2+}$  is for green component and  $\text{CaAlSiN}_3:\text{Eu}^{2+}$  is for red component [9–11]. Therefore, it is meaningful to develop a new kind of phosphor with high performance for near-UV based W-LEDs applications.

Borates, which generally feature the advantages of a low synthesis temperature, excellent chemical and physical stability, have been attracted much attention and widely used as luminescent material hosts [1,12–14]. Such as  $\text{Ba}_2\text{Ca}(\text{BO}_3)_2:\text{Ce}^{3+}$ ,  $\text{NaCaBO}_3:\text{Eu}^{3+}$  and  $\text{Sr}_2\text{B}_2\text{O}_5:\text{Eu}^{2+}$  [12,15,16]. Hence, it is a good approach to

develop a novel borate-based phosphor which can be effectively excited by near-UV light. In 2008, a new borate compound  $\text{CaBi}_2\text{B}_2\text{O}_7$  has been reported [17]. However, to the best of our knowledge, there is no investigation has been performed on studying the luminescent properties of  $\text{Eu}^{3+}$  doped  $\text{CaBi}_2\text{B}_2\text{O}_7$  phosphors.

In this paper, we report a new orange–red phosphor  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  obtained by solid state reaction method. Its crystal structure, luminescence properties of  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  were investigated. Moreover, the CIE chromaticity coordinate and the color purity performances of  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  were also studied. The results suggest that the  $\text{CaBi}_2\text{B}_2\text{O}_7:\text{Eu}^{3+}$  phosphor is a potential orange–red material for the application in the near-UV pumped W-LEDs.

## 2. Experimental

All samples were prepared by conventional solid state reaction method at a high temperature. We used  $\text{CaCO}_3$  (AR),  $\text{H}_3\text{BO}_3$  (AR),  $\text{Bi}_2\text{O}_3$  (AR),  $\text{Eu}_2\text{O}_3$  (AR) as starting materials. The stoichiometric materials were weighted and ground together in an agate mortar. Thereafter, the mixture was transferred to a corundum crucible and precalcined at 500 °C for 1 h, and then subsequently further sintered at 800 °C for 3 h in the air atmosphere. After these processes, the samples were prepared.

The purities of samples were tested by X-ray diffraction (XRD) patterns using Rigaku Ultima IV Advanced X-ray diffractometer with a  $\text{Cu K}\alpha$  (40.0 KV, 30.0 mA) radiation ( $\lambda = 1.5418 \text{ \AA}$ ).

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Photoluminescence excitation (PLE) and emission spectra were measured by Hitachi F-4500 spectrofluorometer equipped with a 150 W Xenon lamp as an excitation source. A cutoff filter was used to avoid the influence of the second-order emission of the source radiation. All the measurements were performed at room temperature.

### 3. Results and discussion

Fig. 1 shows the typical XRD patterns of  $\text{CaBi}_{1.95}\text{B}_2\text{O}_7:0.05\text{Eu}^{3+}$  phosphor. One can see that all of the diffraction peaks can be well fitted with the standard card ICSD #245016 of  $\text{CaBi}_2\text{B}_2\text{O}_7$  with a space group  $Pn\bar{a}21$ , consistent with a  $\text{CaBi}_2\text{B}_2\text{O}_7$  phase of a orthorhombic structure, featuring the lattice parameters of  $a=8.937\text{ \AA}$ ,  $b=5.477\text{ \AA}$ ,  $c=12.591\text{ \AA}$  and  $V=616.33\text{ \AA}^3$  (Fig. 2 shows the detailed crystal structure of  $\text{CaBi}_2\text{B}_2\text{O}_7$ ). There is no impurity peak detected in the experimental range, suggesting that a pure crystalline compound was obtained. The experimental results indicate that the doped  $\text{Eu}^{3+}$  and charge compensator ions have not led to obvious change in the host structure.

The excitation spectrum ( $\lambda_{\text{em}}=591\text{ nm}$ ) of  $\text{CaBi}_{1.95}\text{B}_2\text{O}_7:0.05\text{Eu}^{3+}$  sample is depicted in Fig. 3(a). It can be found that the excitation spectrum is composed of a broad band at short wavelengths ranging from 200 to 350 nm and a group of sharp bands in the region from 350 to 420 nm. The broad band ranging from 200 to 310 nm is attributed to the overlap between the charge transfer band (CTB) of  $\text{O}^{2-} \rightarrow \text{Eu}^{3+}$  [18]. And the group of sharp bands in the region from 350 to 420 nm is consists of a series of sharp peaks at 361 nm, 382 nm, 394 nm and 414 nm, which is belong to the intrinsic 4f–4f transitions of  $\text{Eu}^{3+}$  from the ground state  $^7F_0$  to the excited states  $^7D_4$ ,  $^5G_4$ ,  $^5L_6$ , and  $^5D_3$ , respectively [19], Fig. 3(b)

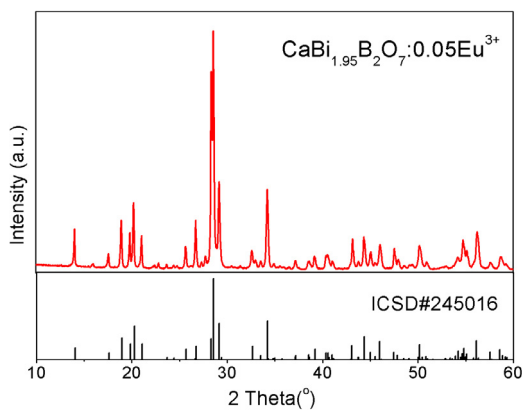


Fig. 1. XRD patterns of  $\text{CaBi}_{1.95}\text{B}_2\text{O}_7:0.05\text{Eu}^{3+}$  phosphor (The standard ICSD#245016 data card of  $\text{CaBi}_2\text{B}_2\text{O}_7$  is provided as a reference.).

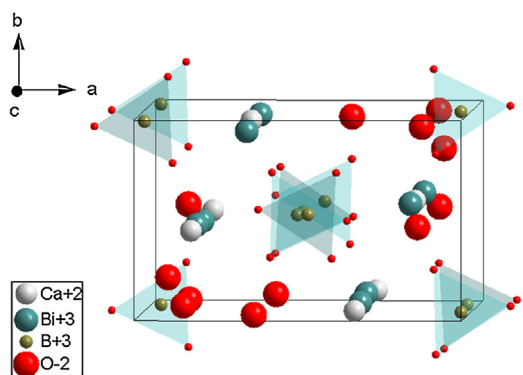


Fig. 2. Crystal structure of  $\text{CaBi}_2\text{B}_2\text{O}_7$ .

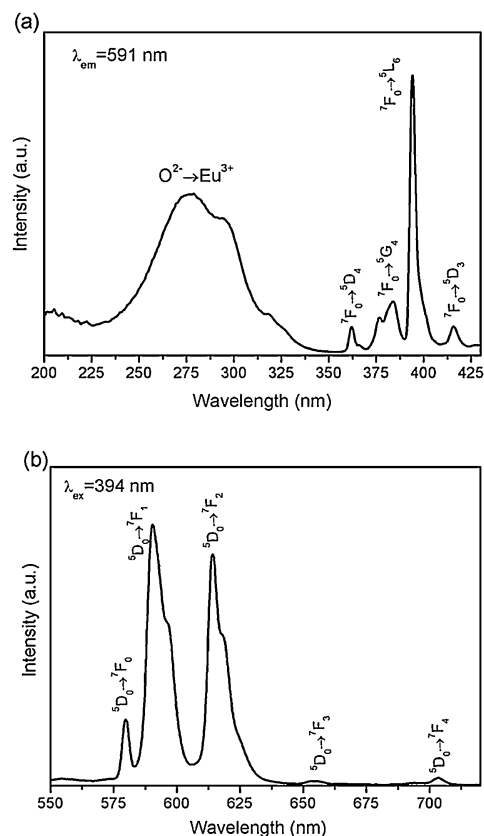


Fig. 3. The PL excitation spectrum ( $\lambda_{\text{em}}=591\text{ nm}$ ) and emission spectrum ( $\lambda_{\text{ex}}=394\text{ nm}$ ) of a typical  $\text{CaBi}_{1.95}\text{B}_2\text{O}_7:0.05\text{Eu}^{3+}$  sample.

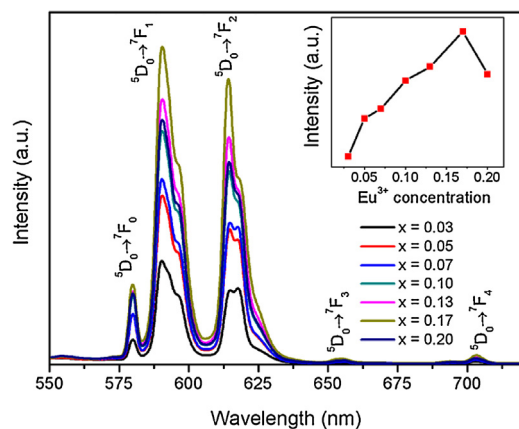


Fig. 4. The emission spectra of  $\text{CaBi}_{2-x}\text{B}_2\text{O}_7:x\text{Eu}^{3+}$  ( $x=0.03, 0.05, 0.07, 0.10, 0.13, 0.17$  and  $0.20$ ) samples excited by 394 nm (Inset: the normalized emission intensity based on the emission intensity of  $^5D_0 \rightarrow ^7F_1$  transition as a function of  $\text{Eu}^{3+}$  ions concentration).

shows the emission spectra of  $\text{CaBi}_{1.95}\text{B}_2\text{O}_7:0.05\text{Eu}^{3+}$  sample under the 394 nm excitation. There are four peaks peaking at 581, 591, 614, 655 and 706 nm due to the  $^5D_0 \rightarrow ^7F_j$  ( $j=0-4$ ) transitions of  $\text{Eu}^{3+}$  ions, respectively [20]. In particular, it can be found that the orange–red emission at 591 nm is dominated.

In order to figure out the optimal doping concentration of  $\text{Eu}^{3+}$  ions in the  $\text{CaBi}_2\text{B}_2\text{O}_7$  host, a series of  $\text{CaBi}_{2-x}\text{B}_2\text{O}_7:x\text{Eu}^{3+}$  ( $x=0.03, 0.05, 0.07, 0.10, 0.13, 0.17$  and  $0.20$ ) samples were prepared. Fig. 4 shows the emission spectra of  $\text{CaBi}_{2-x}\text{B}_2\text{O}_7:x\text{Eu}^{3+}$  as a function of  $\text{Eu}^{3+}$  concentration excited at 394 nm. It can be found that all the spectra of different contents of  $\text{Eu}^{3+}$  doped  $\text{CaBi}_2\text{B}_2\text{O}_7$  are similar and intensity of emission initially increases with the increase of

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