



Blue-yellow emitting Dy³⁺ doped K₂Y₂B₂O₇ novel phosphor for WLED application

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Abstract In this study, we synthesized a set of novel phosphors by introducing Dy³⁺ ions into K₂Y₂B₂O₇ through a combustion synthesis method, employing various Dy³⁺ ion concentrations. X-ray diffraction (XRD) analysis of the prepared phosphor is carried out to confirm the formation of phase pure compound. Fourier Transform Infrared (FTIR) analysis confirms the formations of Y–O and B–O bonds and affirming the absence of contaminants. The photoluminescence investigation of the phosphor unveiled a noteworthy finding: the optimal Dy³⁺ concentration for emission was determined to be 0.5 mol%, with a subsequent concentration quenching effect observed beyond this level. The excitation spectra of K₂Y₂B₂O₇:Dy³⁺ indicates several bands in near UV region, with notable peaks at approximately 351 and 388 nm. Photoluminescence emission spectra of the phosphor gives blue emission at 481 nm and yellow emission at 575 nm under the 350 nm near ultra violet excitation. Furthermore, we determined the Commission Internationale de l’Eclairage (CIE) chromaticity coordinates (Cx, Cy) for these emissions, with values of (0.087, 0.144) for blue and (0.478, 0.520) for yellow, respectively. The findings of this investigation imply a promising application for the synthesized phosphor as an effective blue and yellow emitting component in the realm of white LED technology.

Keywords Combustion synthesis · Phosphor · K₂Y₂B₂O₇:Dy³⁺ · Photoluminescence · FTIR

Introduction

Luminescent materials have found applications in various fields which includes optoelectronic devices, radiation dosimetry, solar cells, paints, etc. These materials have main applications in solid state lighting and display technology. Recently, the alkali metal, alkaline earth metal, and transition metal yttrium have given rise to an extensive array of compounds exhibiting appealing photoluminescent characteristics [1, 2]. Yttrium borate as a host for light emitting materials under UV excitation have attracted much attention due to wide variety of structure [3]. Borate possesses a good chemical and mechanical stability and has a wide spectral range [4–6].

Borates are good host for rare earth ion activated phosphors due to relatively easy and low temperature synthesis, high luminous efficiency, good colour purity, strong absorption near UV region [7, 8]. Its spectroscopic, magneto electric properties make it promising candidate for technical applications [9, 10]. Rare earth doped yttrium-based phosphors are used in the bioimaging, lasers, plasma display panels, fluorescent lamps, etc. [11]

In 2013, Nagpure et al. [12] reported combustion synthesis of borate phosphors viz. YBO₃:Eu³⁺, BaZr(BO₃)₂:Eu³⁺, YCaBO₄:Eu³⁺, YAl₃(BO₃)₄:Eu³⁺, YAl₃(BO₃)₄:Tb³⁺, LaBaB₉O₁₆:Tb³⁺, LaBaB₉O₁₆:(Ce³⁺, Tb³⁺), and Na₃La₂(BO₃)₃:Tb³⁺. These phosphors emit radiations in red and green region under 173 nm excitation. These phosphors are promising candidates for Plasma Display Pannels (PDP) and lamp applications.

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The alkali trivalent R.E. borate with general formula $M_2RE_2B_2O_7$ ($M=Na, K, RE=Y, La$) have been studied for their excellent photoluminescence properties, high crystallinity, high radiation efficiency [9]. Ce^{3+} , Tb^{3+} , Eu^{3+} doped $Na_2Y_2B_2O_7$ phosphor material which gives intense emission from violet region to red region are studied [13]. Koparkar et al. [14] in 2014 reported the synthesis and structural properties of $K_2Y_2B_2O_7:Eu$ by replacing Na by K which shows red emission. The phosphors doped with Dy^{3+} can emit light in various colours, including blue, green, yellow and red, depending on the specific energy levels of the dopant ions [15–17]. This versatility in emission colours makes them crucial for creating full-colour displays and energy-efficient lighting sources. In this paper we introduce a host $K_2Y_2B_2O_7$ doped with Dy^{3+} phosphor synthesized using combustion method. Luminescence properties, crystal structure was studied using XRD and FTIR. The colour coordinates of sample were analyzed with CIE 1931 color chromaticity system.

Experimental

The compounds of $K_2Y_{2(1-x)}B_2O_7:xDy^{3+}$ ($x=0.001, 0.003, 0.005, 0.01, 0.02$) presented in this work were synthesized by combustion method using urea as a fuel. All the starting materials of high purity (AR grade) (99.99%) were used viz. yttrium nitrate hexahydrate ($Y(NO_3)_3 \cdot 6H_2O$), potassium nitrate (KNO_3), boric acid (H_3BO_3), Urea ($NH_2-CO-NH_2$), and dysprosium oxide (Dy_2O_3).

All the ingredients were weighted in stoichiometric proportion and added in mortar except Dy_2O_3 . Firstly, Dy_2O_3 dissolved in dilute nitric acid and excess of HNO_3 was removed at high temperature and then added in mortar. The mixture was ground together for 25 min to get a thick paste. Thus obtained paste was transferred in to the crucible and transferred in preheated muffle furnace maintained at $(540 \pm 10)^\circ C$. After 5 min, mixture undergoes dehydration and liberated N_2 , CO_2 gases and the paste burn with flame; lastly a foamy powder obtained. The entire procedure took between 5 and 10 min. After removing the crucible from the furnace, it was allowed to cool, and the foamy powder was crushed and transformed into a fine powder.

Result and discussion

XRD Analysis of $K_2Y_2B_2O_7:Dy^{3+}$

Figure 1 represents the XRD spectrum of (0.005) Dy^{3+} doped $K_2Y_2B_2O_7$ phosphor. The 2θ ranges from 10° to 90° with the steps of 0.02° . The highest peak intensity is

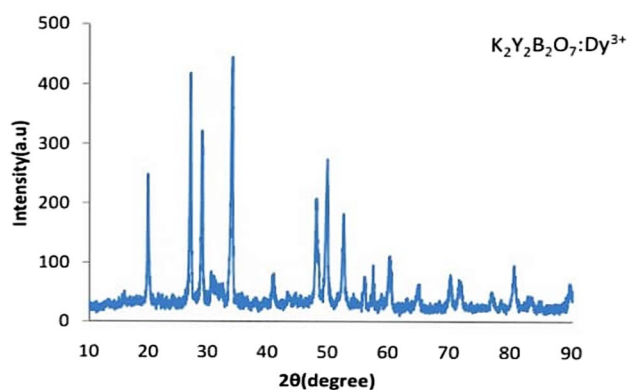


Fig. 1 XRD pattern of $K_2Y_2B_2O_7:Dy^{3+}$ (0.005)

located at the value of $2\theta = 33.924$. The XRD pattern did not show the peaks of precursors like $Y(NO_3)_3 \cdot 6H_2O$, KNO_3 , H_3BO_3 and other likely these, it indicates that the synthesized sample is not the simple chemical mixture of precursors $Y(NO_3)_3 \cdot 6H_2O$, KNO_3 , H_3BO_3 . Furthermore, the XRD data also matches well with the one presented by Koparkar et al. [14] which suggest the prepared substance may be a single host. The detailed structural study of this material is still under investigation, hence, only XRD data is given in this report.

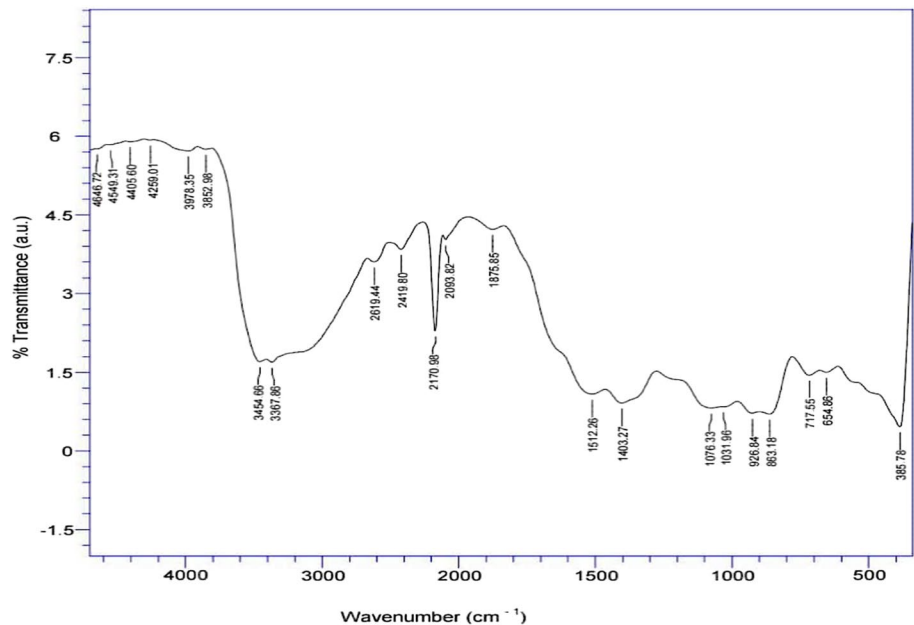
FTIR studies of $K_2Y_2B_2O_7:Dy^{3+}$

Figure 2 illustrates the Fourier Transform Infrared Spectrum of a $K_2Y_2B_2O_7$ phosphor doped with Dy^{3+} (0.005) across wave numbers spanning from 400 cm^{-1} to 4500 cm^{-1} . The region ranging from 400 cm^{-1} to 1600 cm^{-1} is the fingerprint of the sample. The spectrum shows broad peaks, the peaks 400 cm^{-1} – 1078 cm^{-1} which represent the Y–O stretching vibration [18] and the absorption peaks 1403 – 1512 cm^{-1} assigned the B–O stretching mode in BO_3 unit which are component of B_3O_6 group [19]. This confirms the existence of B_2O_4 in $K_2Y_2B_2O_7$. In high frequency region, the peaks due to the stretching and in plane bending vibration of OH group are observed at 3367 cm^{-1} and 3454 cm^{-1} due to the atmospheric moisture.

Photoluminescence studies of $K_2Y_2B_2O_7:Dy^{3+}$ phosphor

Figure 3a shows the PL excitation property of $K_2Y_2B_2O_7:Dy^{3+}$ phosphor. The emission spectra indicate that the phosphor shows good response in near UV region. The excitation spectrum is monitored at 481 nm consist of a several narrow lines peaks at wavelength 325, 351, 366, 388, 427 and 456 nm. These peaks can be attributed to matrix

Fig. 2 FTIR spectra of $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ phosphor



absorption ${}^6\text{H}_{15/2} \rightarrow {}^6\text{P}_{3/2}$, ${}^6\text{H}_{15/2} \rightarrow {}^4\text{I}_{11/2} + {}^4\text{M}_{15/2} + {}^6\text{P}_{7/2}$, ${}^6\text{H}_{15/2} \rightarrow {}^4\text{P}_{3/2} + {}^6\text{P}_{3/2,5/2}$ and ${}^6\text{H}_{15/2} \rightarrow {}^4\text{I}_{13/2} + {}^4\text{F}_{7/2} + {}^4\text{K}_{17/2} + {}^4\text{M}_{19/2,21/2}$ of Dy^{3+} [20]. The emission band observed around 351 nm is strongest one.

Figure 3b shows the emission spectra of $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ monitored at 351 nm shows two visible emission bands, at 483 nm (blue) which is attributed to ${}^4\text{F}_{9/2}$ to ${}^6\text{H}_{15/2}$ and at 575 nm (yellow) corresponds to ${}^4\text{F}_{9/2}$ to ${}^6\text{H}_{13/2}$ [21]. Generally, the optical properties of the phosphors are depends on the structure of the host matrix. When Dy^{3+} resides in a high symmetry site (possessing an inversion center), it exhibits stronger blue emission, whereas positioning Dy^{3+} in a low symmetry site (lacking an inversion center) results in stronger yellow emission. The photoluminescence emission spectrum of the $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ in Fig. 3b reveals a more pronounced blue emission at 483 nm as compared to the yellow emission which is observed at 575 nm., This observation implies that Dy^{3+} occupies a high symmetry site in $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7$ [22, 23]. With an increase in the Dy^{3+} concentration (ranging from 0.1, 0.3, 0.5, 1, 2 mol %), the emission intensity initially rises and subsequently declines. The optimum concentration of Dy^{3+} is 0.5 mol%. The emission intensity decreased beyond this doping concentration due to concentration quenching, which is a

result of cross relaxation between the neighbouring rare earth ions [24]. Figure 3c shows the variation in emission intensity with the concentration of Dy^{3+} ions in $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ phosphor.

Chromatic properties of $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$

The CIE chromaticity of $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ is represented in Fig. 4. The CIE coordinates for wavelength 481 and 575 nm are (0.087 and 0.144) and (0.478 and 0.520) respectively which shows the high degree of colour purity [25–27].

Conclusion

In summary, a series of $\text{K}_2\text{Y}_2\text{B}_2\text{O}_7:\text{Dy}^{3+}$ phosphors with different concentrations of Dy^{3+} were successfully synthesized by combustion method. The XRD pattern of the compound indicates that the material exhibited a high level of crystallinity, with multiple peaks associated with precursors or dopants. The presence of function group composition in the precursor was confirmed by FTIR study. The phosphor exhibits good photoluminescence properties. Several

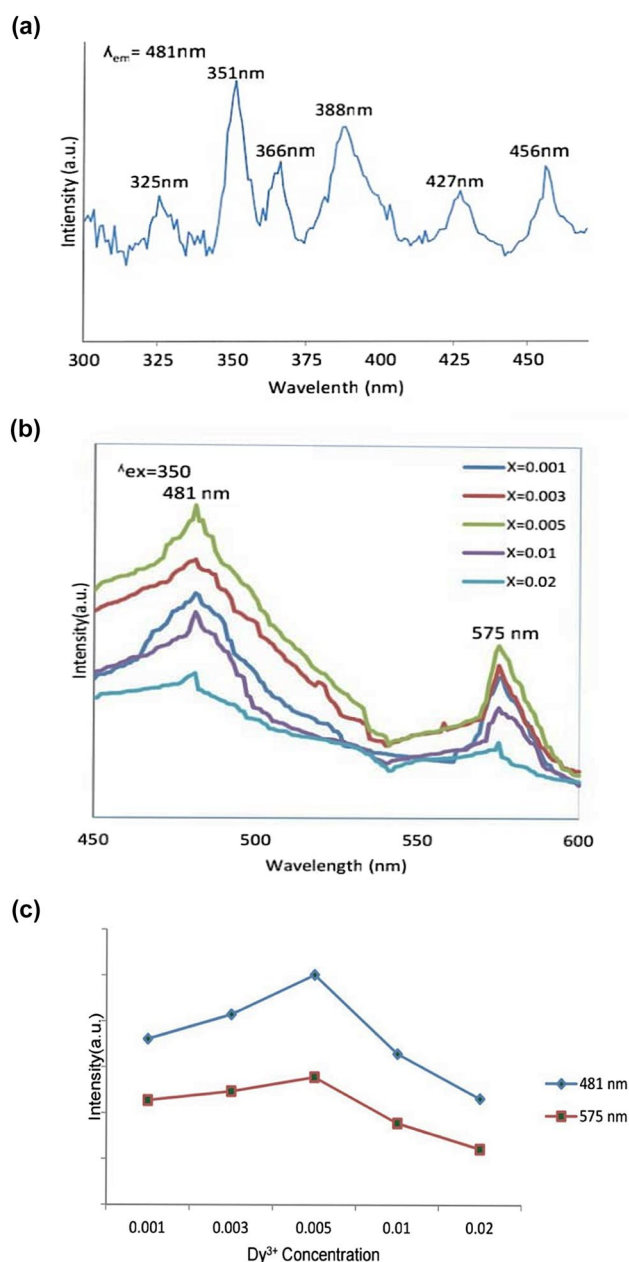


Fig. 3 **a** Excitation spectra of $K_2Y_2B_2O_7:Dy^{3+}$ phosphor monitored at 481 nm emission. **b** Emission spectra of $K_2Y_2B_2O_7:Dy^{3+}$ phosphor monitored at 350 nm excitation. **c** Variation in the emission intensity with different doping concentration of Dy^{3+} in the $K_2Y_2B_2O_7:Dy^{3+}$ phosphor

excitation peaks are observed from 325 to 456 nm. The sample shows the blue and yellow emission at 483 nm and 575 nm respectively. The result of PL spectra indicates that the phosphor $K_2Y_2B_2O_7:Dy^{3+}$ has potential application in fluorescent lamp or in white LEDs.

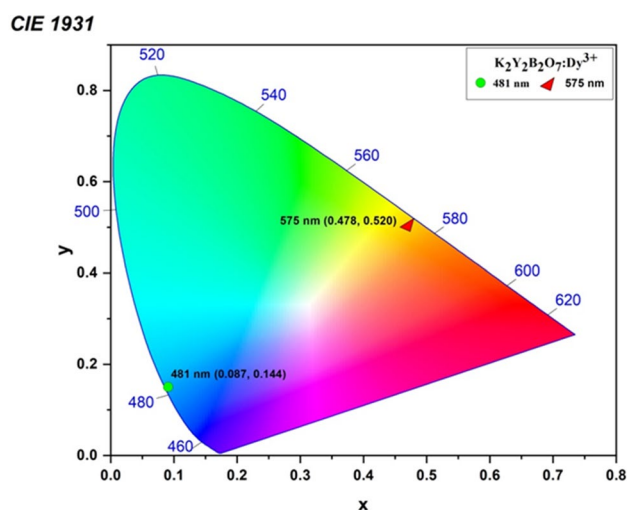


Fig. 4 CIE diagram of $K_2Y_2B_2O_7:Dy^{3+}$ Phosphors

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Declaration

Conflict of interest The authors declare that they do not have any known competing financial interests or personal relationships that may seem to have influenced the work reported in this paper.

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