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Synthesis and Luminescence Study of novel $NaBaY(BO_3)_2:Dy^{3+}$ Phosphor for White LED

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ABSTRACT

Dy^{3+}activated $NaBaY(BO_3)_2$ phosphor was prepared by a simple route of solution combustion technique. The synthesis is based upon the exothermic reaction between the fuel (urea) and oxidizer (ammonium nitrate). The heat generated in the reaction is utilized for auto combustion of ingredients. The phase formation and crystal structure of the prepared nanophosphors were confirmed by powder XRD technique. An X-ray structure analysis revealed that the compound crystallizes in space group of R-3m(148), with unit cell parameter a= 5.360 Å, c=36.16 Å. The luminescence properties such as photoluminescence excitation (PLE) and emission (PL) spectra were reported. The emission of Dy^{3+} ions upon 254 nm excitation is observed at 490 nm (blue) due to the $^4F_{9/2}\rightarrow{}^6H_{15/2}$ transitions and 546 nm (yellow) due to $^4F_{9/2}\rightarrow{}^6H_{13/2}$ transitions. All the results imply that the synthesized phosphor could be potentially used as white LEDs.

***Keyword:** Combustion synthesis, Photoluminescence, phosphors, W-LEDs, Rare-earth*

1. INTRODUCTION

Dy^{3+} ion, as an important activator for phosphors, often exhibits two main emissions in the visible region: one in the blue region (~ 480 nm) and one in the yellow region (~ 580 nm), originating from $^4F_{9/2}\rightarrow{}^6H_{13/2}$ and $^4F_{9/2}\rightarrow{}^6H_{15/2}$ transitions of $Dy3^+$ ions, respectively. So, Dy^{3+} ion is often chosen to realize the white-light emission in a single-phase host by suitably adjusting the yellow-to-blue intensity ratio **[1].** Consequently Dy^{3+} activated luminescent material attracted much attention **[2, 3]** because of their significant application as potential single phase white phosphors.

White light-emitting diodes, or w-LEDs, have attracted a lot of interest because of their extended lifespan, superior energy efficiency, and environmental friendliness **[4].** These days, phosphor-converted white LEDs, or PC-wLEDs, have gradually demonstrated their supremacy in terms of industrialization due to their inexpensive cost as well as simple manufacturing method. Optimizing the performance of luminous materials is crucial to the development of PC-wLEDs. White light can be generated by combining an InGaN LED chip that emits blue light between 450 and 470 nm with $Y_3Al_5O_{12}:Ce^{3+}$ yellow phosphors. However, because of the lack of red emission in long wavelength regions and insufficient thermal stability, this approach has significant disadvantages, including a low color rendering index and thermal quenching **[5].** A different approach for creating white LEDs with high CRI and low CCT values involves using near-ultraviolet (near-UV) LED chips to excite red, green, and blue tri-color phosphors **[6-10].**

Therefore, In this work, Dy^{3+} doped $NaBaY(BO_3)_2$ phosphors were synthesized by using solution combustion technique. The corresponding structure and luminescent properties were characterized by XRD and PL spectra.

2. EXPERIMENTAL

The sample were prepared by a novel solution combustion technique; the various steps involve in the preparation were systematically describe in flowchart in Fig. 1. The starting ingredients $K(NO_3)$, $Sr(NO_3)_2$, $Y(NO_3)_3$, $Eu(NO_3)_3$, and H_3BO_3, NH_4NO_3 were used.

The stoichiometric amounts of starting ingredients were calculated as per the balance inorganic reaction listed in table no.1 Firstly, all starting ingredients like Y_2O_3, Eu_2O_3 were converted into $Y(NO_3)_3$ and $Eu(NO_3)_3$ by mixing Y_2O_3 and Eu_2O_3 into few ml of dil. HNO_3.

All the Ingredients were thoroughly mixed in an Agate Mortar, by adding little amount of double distilled water and place on heating mantle for about 30 minute to obtain homogeneous, clear and thick solution. The aqueous solution was then transferred into preheated muffle furnace maintained at 550ºC. The

solution boils, foams, ignites and undergoes dehydration and then decomposition with liberation of NH_3 and NO_2 gasses and obtained a voluminous foamy powder. The prepared material is then taken out of the furnace and the foamy product is crushed into a fine powder and heated at 800^0C for about 1 hour. Some of the phosphors reported by this technique **[11-15].**

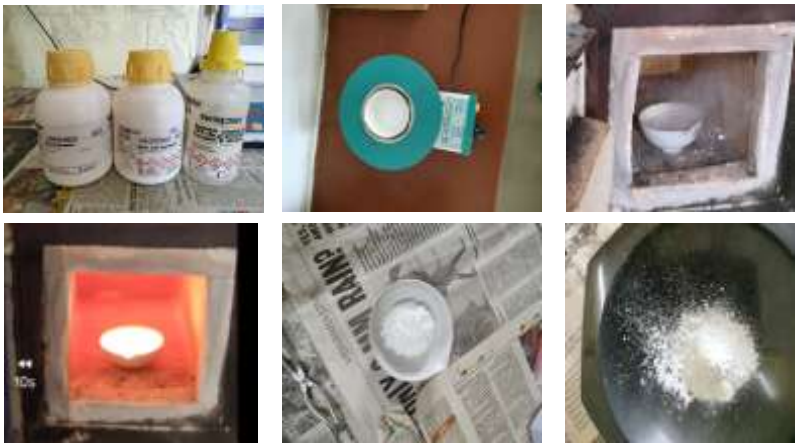

Fig. 1 Stepwise synthesis of phosphor by solution combustion technique.

Table 1. The molar ratios of ingredients used in the synthesis and corresponding balanced chemical reactions.

S.N.	Phosphors.	Balanced chemical reaction with molar ratios of ingredients.
1.	$NaBaY_{(1-x)}(BO_3)_2$: xDy^{3+} (x= 5 mol%)	$Na(NO_3) + Ba(NO_3)_2 + (1-x)Y(NO_3)_3 + 2H_3BO_3 + 9NH_4NO_3 + 10\ CO(NH_2)_2 + xDy(NO_3)_3 \xrightarrow[550\ 0C]{heat} NaBaY(BO_3)_2:Dy^{3+}$ + Gaseous Products ($H_2O\uparrow$, $NH_3\uparrow$ and $NOx\uparrow$)

3. CHARACTERIZATIONS

The XRD pattern of the polycrystalline powder sample $NaBaY(BO_3)_2:Dy^{3+}$ were recorded on Rigaku Miniflex ✕ 600 X-ray Diffractometer using Cu Kα radiation with 1.5418 Å wavelengths. At room temperature, the XRD data was recorded at scanning angles ranging from 10° and 80° in steps of 0.2° per second. The photoluminescence spectra (excitation and emission) were recorded on Hitachi F-7000 fluorescence spectrophotometer.

3.1 Phase Formation

The structure of $NaBaY(BO_3)_2$ consists of layered ortho-borate units and it crystallizes in trigonal crystal system with the space group of R-3m(148) and the lattice parameters are a= 5.360 Å, c=36.16 Å. In this structure, the $(BO_3)^{3-}$ groups form planar layers that exhibit threefold symmetry. The Ba^{2+} and Na+ ions in $NaBaY(BO_3)_2$ occupy distinct cation sites with threefold symmetry, located near the borate layers **[16-17]**. The XRD pattern of $NaBaY(BO_3)_2$ with 5 mol% Dy^{3+} is shown in fig.2. Most peaks are indexed to the $NaBaY(BO_3)_2$ phase, which agrees well with JCPDS No, 48-0307. It indicates that the doping of Dy^{3+} ions does not significantly alter the crystal structure.

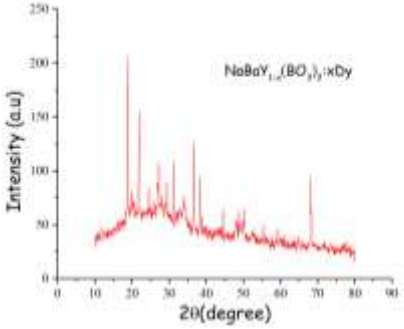

Fig .2 XRD pattern of $NaBaY_{1-x}(BO_3)_2:xDy^{3+}$

3.2 Photoluminescence properties

Fig.3 shows the emission spectrum of $NaBaY(BO_3)_2:Dy^{3+}$ phosphor under 254 nm excitation with 5mol % Dy^{3+}. The main peak is observed at 490 nm (blue) due to the $^4F_{9/2}\rightarrow{}^6H_{15/2}$ transitions and 546 nm (yellow) due to $^4F_{9/2}\rightarrow{}^6H_{13/2}$ transitions of Dy^{3+}, which is because Dy^{3+} occupies the noncentrosymmetric position in the crystal structure of $NaBaY(BO_3)_2$

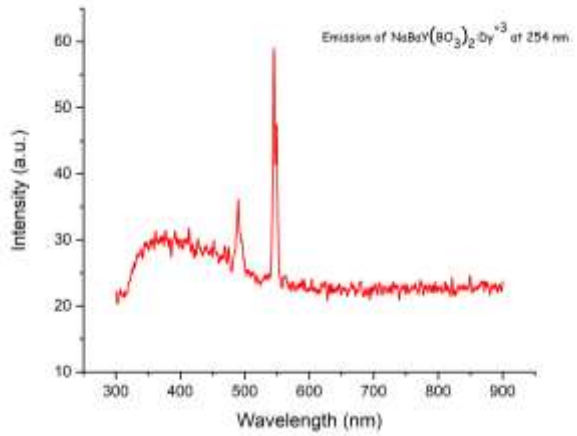

Fig. 3 Emission spectrum of $NaBaY_{0.95}(BO3)_2:Dy_{0.05}$ (λ_{ex}=254nm).

Fig. 4 shows the excitation spectra of $NaBaY(BO_3)_2:Dy^{3+}$ by monitoring 546 nm ($^4F_{9/2}\rightarrow{}^6H_{13/2}$ transition of Dy^{3+}). The spectrum presents the transition lines with the strong peak at 206 nm and 254 nm. The present phosphor has excitation in the near-UV light.

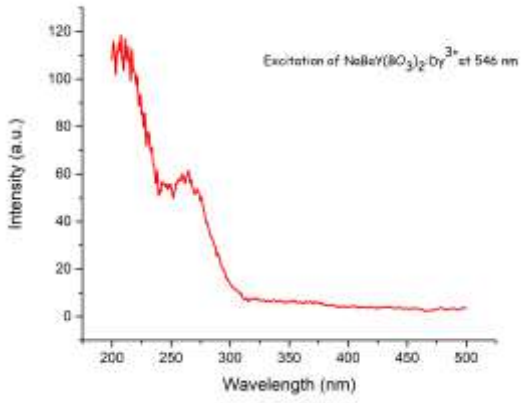

Fig. 4 Excitation spectrum of NaBaY0.95 (BO3)2:Dy0.05 (λem=546nm)

4. CONCLUSION

In summary, the phosphors $NaBaY(BO_3)_2:Dy^{3+}$ was synthesized successfully by a simple, time saving, low cost and rapid prototype solution combustion method. Powder XRD pattern confirmed the formation desired product. The study presented that the $NaBaY_{1-x}(BO_3)_2:xDy^{3+}$ could be excited by near ultraviolet light and the emission peak located at 490 nm (blue) and 546 nm (yellow). These results indicates that $NaBaY(BO_3)_2:Dy^{3+}$ Phosphor shows potential application in NUV white light generating applications.

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