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Combustion Synthesis and Photo- luminescent properties of $NaBaY(BO_3)_2:Sm^{3+}$ Phosphor for Solid State Lightning

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ABSTRACT

Sm^{3+} activated $NaBaY(BO_3)_2$ phosphor was prepared by a simple route of solution combustion technique. The synthesis is based upon an exothermic reaction between urea, acting as the fuel, and ammonium nitrate, serving as the oxidizer. The heat generated in the reaction is utilized for auto combustion of ingredients. The phase formation, surface morphology and luminescent properties of phosphor were investigated using XRD, SEM and Photoluminescence analysis respectively. An X-ray structure analysis revealed that the compound crystallizes in space group of R-3m(148), with unit cell parameter a= 5.355 Å, c=36.26 Å. Under NUV excitation Sm^3 ions exhibit emission at 606 nm which corresponding to the $^4G_{5/2} \rightarrow {}^6H_{7/2}$ transition of Sm^{3+}.

Keywords: - Combustion synthesis, Photoluminescence, Phosphors, Solid-state lightning, Rare-earth

1. INTRODUCTION

Over the past two decades, inorganic phosphor materials doped with rare-earth ion have garnered significant interest owing to their extensive luminescent emission across various colors, originating from f-f or f-d transitions, and their potential applications in solid-state lighting (SSL) and white light-emitting devices (W-LEDs) [1-4]. Additionally, trivalent rare-earth elements have emerged as the most promising activators of phosphors owing to their luminescence properties in the ultraviolet (UV) and visible regions [5]. W-LEDs are getting particular attention due to their energy efficiency, prolonged lifespan, and environmentally favourable characteristics [6-7]. Furthermore, these materials emit white luminescence, offering significant advantages through the replacement of traditional light sources such as incandescent and fluorescent lamps [8-10]. Phosphors are generally attracting a lot of attention because they primarily occur in a variety of inorganic compounds that contain oxygen, including silicates, aluminates, borates, oxides, and alumina-borates. Inorganic rare-earth doped phosphors incorporated into a suitable host matrix have a wide range of applications, including W-LED, FED, fiber optic communications, opto-electronic devices, solid-state lasers, and optical fiber amplifiers etc.

Borate phosphors are ideal host materials for luminescence due to their many applications like large electronic band gap, low synthesis temperature, chemical and environmental stability, and high UV-VUV region transparency [11]. The selection of host material with preferred concentration of doping ion is much important to be considered in developing more effective results [12]. Therefore, trivalent RE activator ion Sm^{3+} plays an important role to produce intense emission in the reddish-orange region for the selected host matrix. In the present work, we have successfully synthesized and investigated for Sodium-barium-yttrium borate (NBYB) phosphors of different doping concentrations of Sm^{3+} ion.

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 $Na(NO_3)$, $Ba(NO_3)_2$, $Y(NO_3)_3$, $Sm(NO_3)_3$ 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. The starting ingredients like Yttrium-oxide and Samarium-oxide were transformed into Yttrium-nitrate and Samarium-nitrate by mixing them into few ml dil. nitric acid. All the materials were mixed thoroughly in an Agate Mortar with a small amount of double-distilled water and placed 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 and suddenly cooled to room temnperature.

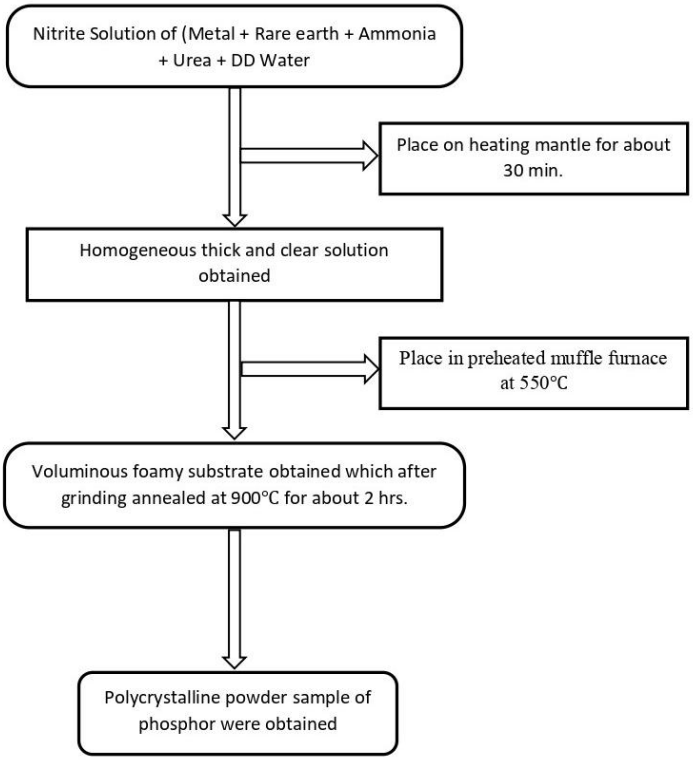

Fig. 1 Flowchart of solution combustion technique

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.	$NaBaY_{(1-x)}(BO_3)_2:xSm^{3+}$ (x= 0.01, 0.03, 0.05, 0.07, 0.09)	$Na(NO_3) + Ba(NO_3)_2 + (1\text{-}x)Y(NO_3)_3 + 2H_3BO_3 + 9NH_4NO_3 + 10\ CO(NH_2)_2 + xSm(NO_3)_3 \xrightarrow[550^oC]{heat} NaBaY(BO_3)_2:Sm^{3+}$ + Gaseous Products ($H_2O\uparrow$, $NH_3\uparrow$ and $NOx\uparrow$)

3. CHARACTERIZATIONS

The XRD pattern of the polycrystalline powder sample NaBaY(BO3)2:Sm^{3+} were recorded on Rigaku Miniflex × 600 X-ray Diffractometer using Cu Kα radiation with 0.15405 Å wavelengths. The XRD data was recorded at scanning angles ranging from 10° and 80° in steps of 0.2° per second at room temperature,. The photoluminescence spectra (excitation and emission) were recorded on Hitachi F-7000 fluorescence spectrophotometer serial number: 2475-005 with 450 W Xenon lamp as the excitation source. The surface properties and particle size of synthesized phosphor were studied by using SEM instrument Jeol JSM. (Model IT 200) having standard tungsten filament.

3.1 XRD Analysis

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= b=5.355 Å, c=36.26 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 [13-14]. The XRD pattern of $NaBaY(BO_3)_2$ 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 Sm^{3+} ions does not significantly alter the crystal structure. The unit cell structure of synthesized borate material is very much identical with the experimental results reported by T N Svetlyakova et al. [15].

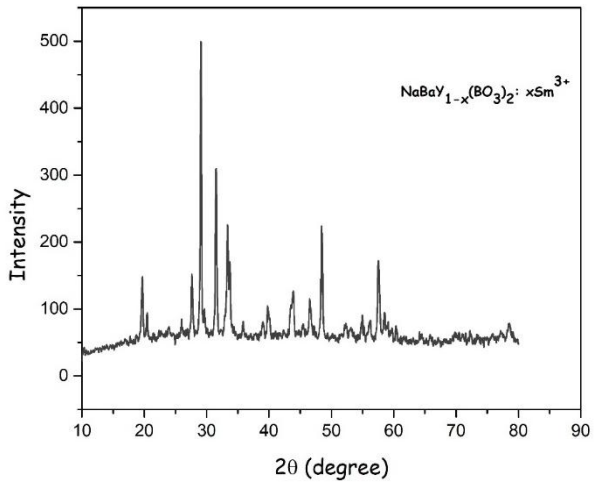

Fig. 2 XRD pattern of $NaBaY(BO_3)_2$ prepared by solution combustion technique

3.2 Photoluminescence Analysis

The photoluminescence spectra of $NaBaY_{1-x}(BO_3)_2:xSm^{3+}$ (x=0.01, 0.03, 0.05, 0.07, 0.09) is depicted in Fig. 3. The excitation spectra Fig. 3 (b) monitored at 600 nm shows two main broad band peaking at 375 nm and 405 nm leads to the transition of Sm^{3+} ions $^6H_{5/2} \rightarrow {}^4D_{1/2}$ and $^6H_{5/2} \rightarrow {}^4F_{7/2}$ [16]. The narrowband peaking at 405 nm was the dominant in the excitation peaks and this wavelength was selected for recording the emission spectra. The emission spectrum under 405 nm excitation within a scanning range of 500-700 nm, consists of multiple peak at 566 nm, 606 nm and 656 nm which corresponds to the $^4G_{5/2} \rightarrow {}^6H_j$ (j=5/2, 7/2, 9/2) transition of Sm^{3+}. The emission from the electric dipole transition $^4G_{5/2} \rightarrow {}^6H_{7/2}$ (606 nm) is stronger than the emission from the magnetic dipole transition $^4G_{5/2} \rightarrow {}^6H_{5/2}$ (566 nm) which indicates that Sm^{3+} ions occupy the sites with low symmetry. All these assigned transitions were in good agreement with the earlier reports.

It was noted that as the concentration of Sm^{3+} in the host matrix increases, the intensity of the emission correspondingly increases. This intensity of emission is optimum for Sm^{3+} concentration at 0.05 mol. Due to the cross relaxation of Sm^{3+} ions, the concentration quenching effect was occurred at the 0.09 mol and hence that emission intensity was decreased as shown in Fig. 4.

3.3 Surface Morphology

Fig.5 shows the FE-SEM micrograph of as-synthesized phosphor. The SEM images shows that the particles are irregularly shaped and consist of agglomerated grains which is typical for phosphor synthesized by solution combustion method. The particle size is found to be in the range of 5-10μm, with some degree of size dispersion. No significant change in the morphology of material is observed upon doping of RE ions, indicating that the incorporation of rare-earth ions does not significantly affect the surface texture and growth behaviour of the $NaBaY(BO_3)_2$ host lattice.

4. CONCLUSION

We successfully synthesized an intense reddish-orange emitting Sm^{3+} activated sodium-barium-yttrium borate phosphors (NBYB) by solution combustion technique and their structural, morphological and luminescence properties were investigated. The XRD result shows the highly crystalline and phase purity of the synthesized phosphor. These samples with various doping concentrations of Sm^{3+} were monitored by NUV excitation wavelength at 405 nm which results four emission bands at 566 nm, 606 nm and 656 nm due to the distinctive electronic transitions $^4G_{5/2} \rightarrow {}^6H_j$ (j=5/2, 7/2, 9/2) of Sm^{3+} respectively. Hence this synthesized $NaBaY_{1-x}(BO_3)_2:xSm^3$ phosphor might be a promising candidate in reddish-orange lighting devices with WLEDs pumped at NUV region.

5. ACNOLEDMENT

The authors sincerely acknowledge the support of the Head of the Department, Government Vidarbha Institute of Science and Technology for providing the necessary laboratory facilities and also grateful to Shri. Shivaji Science College, Amravati for granting access to advanced characterization facilities, including XRD, SEM, and PL. The constant encouragement and access to advanced experimental facilities are greatly appreciated.

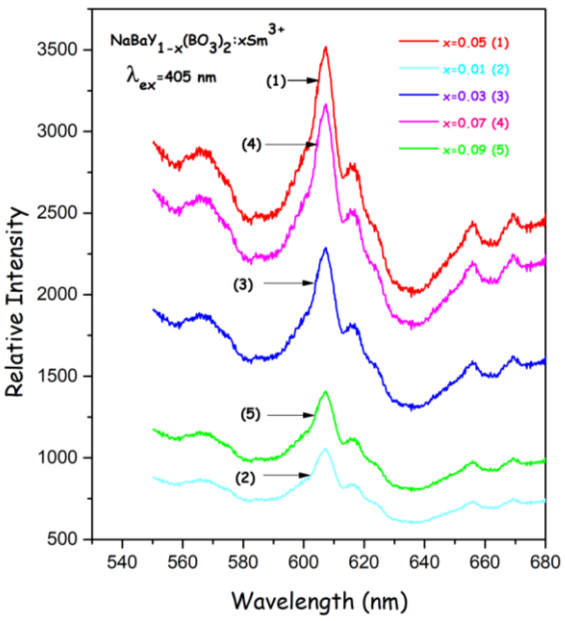

Fig.3 (a) Emission spectra of $NaBaY_{1-x}(BO_3)_2$: xSm^{3+} (x=0.01-0.09) for λ_{ex}=405 nm

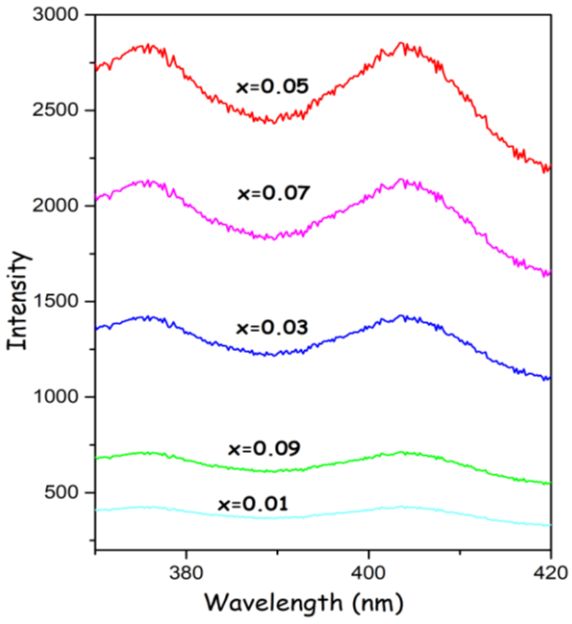

Fig.3 (b) Excitation spectra of $NaBaY_{1-x}(BO_3)_2$:xSm^{3+} (x=0.01-0.09) for λ_{em}=600 nm

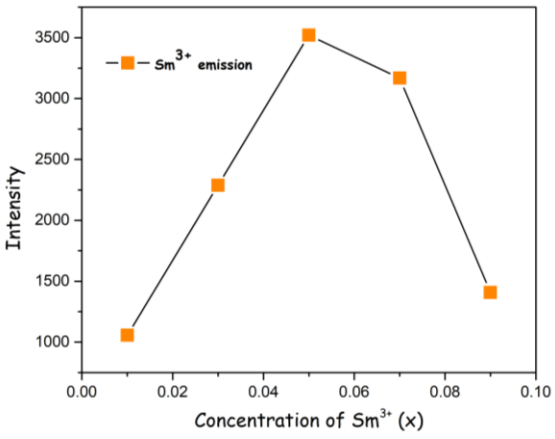

Fig. 4 Dependence of the emission intensity $^{+}$ on the concentration of Sm^{3}

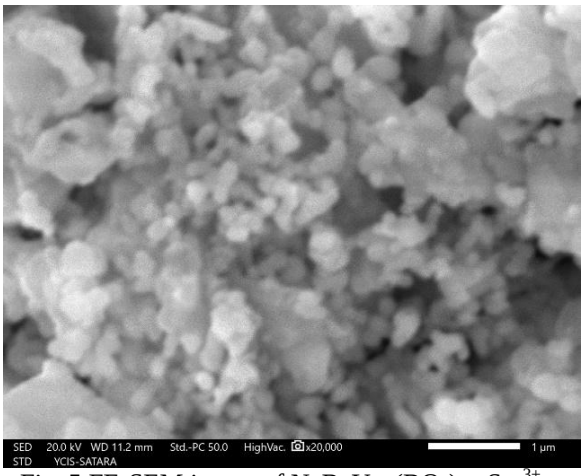

Fig. 5 FE-SEM image of $NaBaY_{1-x}(BO_3)_2$:xSm^{3+}

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