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Eu^{3+}activated $Na_2Ba_4(PO_4)Cl$ halophosphate phosphors for UV excitable white light-emitting phosphors.

Naktode PK[1], Akojwar Ashish[1], Shinde KN[1,*] and Kokode NS [2]

[1]N.S. Science and Arts College, Bhadrawati- 442902, India
[2]N.H.College, Bramhapuri - 441206,India
Email: kartik_shinde@rediffmail.com

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ABSTRACT
Red-orange Eu^{3+} activated $Na_2Ba_4(PO_4)$ Cl halophosphate phosphors excitation 396 nm was prepared via combustion method. The structure and properties were characterized by Scanning electron microscope (SEM) and Photoluminescence (PL) technique. The photoluminescence properties were studied by taking excitation and emission spectra and gives red-orange which corresponds $^5D_0 \rightarrow ^7F_1$ and $^5D_0 \rightarrow ^7F_2$ transition of Eu^{3+}. $Na_2Ba_4(PO_4)$ Cl:Eu phosphors may be good candidate for white LED lighting application.

Keywords: Phosphor; photoluminescence; LED.

INTRODUCTION

Rare earth (RE) ion doped materials have drawn great attention due to their significant properties for research on phosphors suitable for fabricating white-light-emitting diodes (LEDs) has attracted more attention [1-4]. White light-emitting diodes (W-LEDs) offer benefits such as high luminous efficiency, low energy consumption, long lifetime, and environment friendly and so on. They are useful to the next generation for solid state lighting, by replacing of conventional incandescent and fluorescent lamps [5]. They have been used mainly for LCD backlighting, traffic lights, and information boards. One of the strategies to produce white light is to utilize a combination of blue LED with yellow YAG:Ce phosphor [6].

However, this strategy has several disadvantages, such as thermal quenching, poor color rendition and narrow visible range. As an alternative, a novel approach to utilize near ultraviolet (NUV) excitation has been suggested. NUV InGaN-based LEDs, having from 350 to 420 nm received more attention because NUV-LED can offer a highly efficient solid-state lighting [7]. It has little effect on the chromaticity coordinate of white LED, i.e., the quality of white color is generally determined by the visible radiation distribution of phosphor between 380 and 730 nm [8]. Therefore, a novel and stable red phosphor is expected, which shows intense emission efficiently upon NUV excitation. It is well known that hosts of red phosphors widely studied are based on phosphates [9]. The emission of Eu^{3+} is situated in the red orange spectral region and consists of transitions from $^5D_0 \rightarrow {}^7F_1$ or $^5D_0 \rightarrow {}^7F_2$ [10]. Hence trivalent europium (Eu^{3+}) has been known to shows a strong emission in red–orange region for solid state lighting applications [11]. In this work, we synthesized $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ phosphors by using the combustion method in order to find the possibility of the applications as rare-earth ions-doped phosphors for NUV excitation. We characterized the structures and properties of $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ phosphors. Also we studied photoluminescent properties of $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ phosphors by taking excitation and emission spectra. We found that the $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ phosphors were red-orange emitting phosphors and had higher efficiency for the operation with the NUV excitation.

METHODOLOGY

The Eu^{3+} activated $Na_2Ba_4(PO_4)Cl$ phosphors were prepared by combustion method. The starting AR grade materials (99.99% purity) were taken where Sodium nitrite ($NaNO_2$), Barium nitrite $Ba(NO_2)_2$, ammonium di-hydrogen phosphate ($NH_4H_2PO_4$), ammonium chloride (NH_4Cl), and europium oxide (Eu_2O_3) also urea used as fuel. In the present investigation, materials were prepared according to the chemical formula $Na_{2-x}Ba_4(PO_4)Cl: Eu_x$ ($0.1 \leq x \leq 1.0$). After mixing all reagents about 15 min, and we will get a homogeneous mixture and transferred into a furnace preheated at 900 ^{0}C and the porous products were obtained. All the samples were checked Photoluminescence (PL) and photoluminescence excitation (PLE) spectra were measured using Shimadzu RF5301PC fluorescence spectrometer

RESULTS AND DISCUSSION

Surface Morphology of $Na_2Ba_4(PO_4)Cl:Eu^{3+}$:

The particle size distribution of the phosphor is an important factor for its application in WLEDs. Fig. 1 show the SEM images of $Na_{2-x}Ba_4(PO_4)Cl:Eu_X^{3+}$ where x=0.1, 0.3, 0.5 phosphors with various concentrations of Eu^{3+} ions prepared by combustion method at 900 ^{0}C. A SEM study was carried out to investigate the surface morphology and crystallite sizes of the synthesized $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ phosphors powders. An average crystallite size is in the sub-micrometer range from 5µm-10µm. In the combustion method, we found the optimal shape and size of the phosphor. The spherical and agglomerated particles are observed. Most particles showed sizes of a few micrometers. These results indicate that the final product is in crystalline forms. Typical morphological images are represented in Fig. 1 It can be seen that the particle of all samples possess same morphology and size.

Photoluminescent Properties of Eu^{3+} activated $Na_{2-x}Ba_4(PO_4)Cl:Eu_x^{3+}$where x=0.1, 0.3, 0.5 phosphers:

The excitation spectrum by monitoring $^5D_0$–$^7F_2$ emission of Eu^{3+} in Na_{2-x} Ba_4 $(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor is given in Fig. 2. The narrow peaks located at wavelengths longer than 396 nm, which is caused by the characteristic $^7F_0$-$^5L_6$ transition of Eu^{3+} [12]. Fig.3. shows the emission spectra of Na_{2-x} Ba_4 $(PO_4)Cl: Eu_x^{3+}$ where x=0.1, 0.3,0.5 and 1m% phosphor under direct excitation the $^7F_0$–$^5L_6$ transition of Eu^{3+} at 396 nm. Na_{2-x} Ba_4 $(PO_4)Cl: Eu_x^{3+}$ where x=0.1, 0.3,0.5 and 1m% phosphor is composed of a series of linear spectra. Typical linear emission peaks of Eu^{3+} can be observed in the range of 592–620 nm and ascribed to the transition $^5D_0$ level to $^7F_1$, $^7F_2$ levels of Eu^{3+}, respectively. The Eu^{3+} doped $Na_{2-x}Ba_4(PO_4)Cl$ where x=0.1,0.3,0.5 and 1m% phosphor have useful significance because excitation and emission of Eu^{3+} is very efficient in several hosts. The first excited 5d configuration lies near to the excited 4f levels and substituted Eu^{3+} ion is supposed to include in the transitional layer. The obtained products emitted the red luminescence of varying intensities, which showed that the activator Eu^{3+} had successfully entered the host lattice of $Na_{2-x}Ba_4$ $(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor. Additionally, due to the large spatial extension of the 5d wave function, the optical spectra due to the 5d-4f transitions usually depend on the surroundings of the Eu^{3+} ions. The emission spectra of Na_{2-x} Ba_4 $(PO_4)Cl$: Eu_x^{3+} where x=0.1, 0.3,0.5 and 1m% phosphor in general, Eu^{3+} has dominant peaks in the emission spectra in many host matrices. The peaks located at 616 (red) corresponds to the hypersensitive transition $^5D_0 \rightarrow {}^7F_2$ (*616 nm*) and another peaks located at 596 nm (red-orange) is due to the transition $^5D_0 \rightarrow {}^7F_1$ (*596 nm*) [13]. It is well-known that the $^5D_0 \rightarrow {}^7F_1$ transition belongs to the magnetic dipole transition which scarcely changes the crystal field strength around the Eu^{3+} ions and this transition is independent of the symmetry and the site occupied by Eu^{3+} ions in the host [14]. While the transition of $^5D_0 \rightarrow {}^7F_2$ belongs to a forced electric dipole transition and its intensity is very sensitive to the site symmetry of the Eu^{3+} ions [15-17]. In this sample preparation phosphor gives the maximum intensity of emission are observed at (Eu^{3+}) 0.5 m% shown in Fig. 4. The excitation spectrum is at 616 nm and it is properties of LED lighting and gives the emission in orange-red region and this phosphor may be good candidates for white LED lighting.

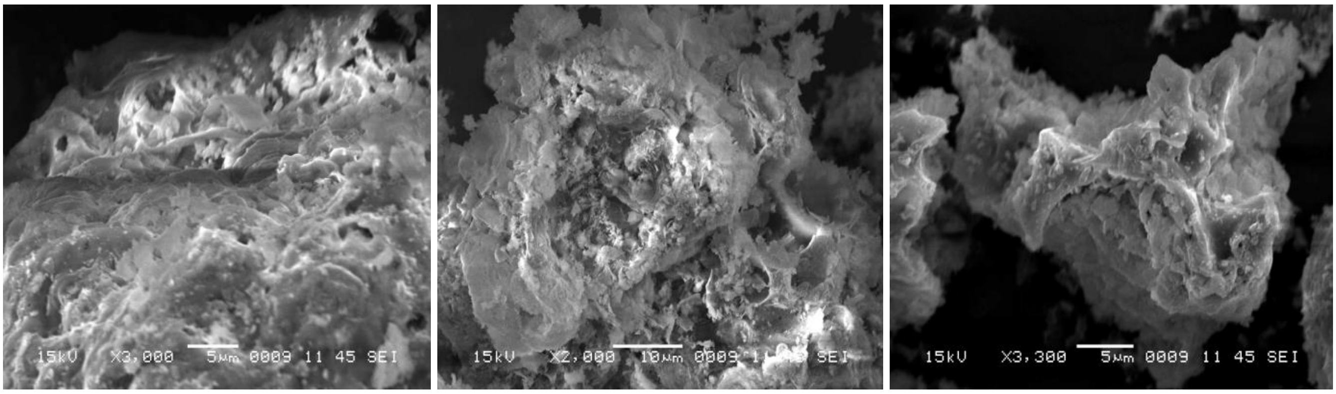

Fig.1 represent the SEM images of the Na_{2-x} Ba_4 $(PO_4)Cl$: Eu_x $(0.1 \leq x \leq 1.0)$ powders

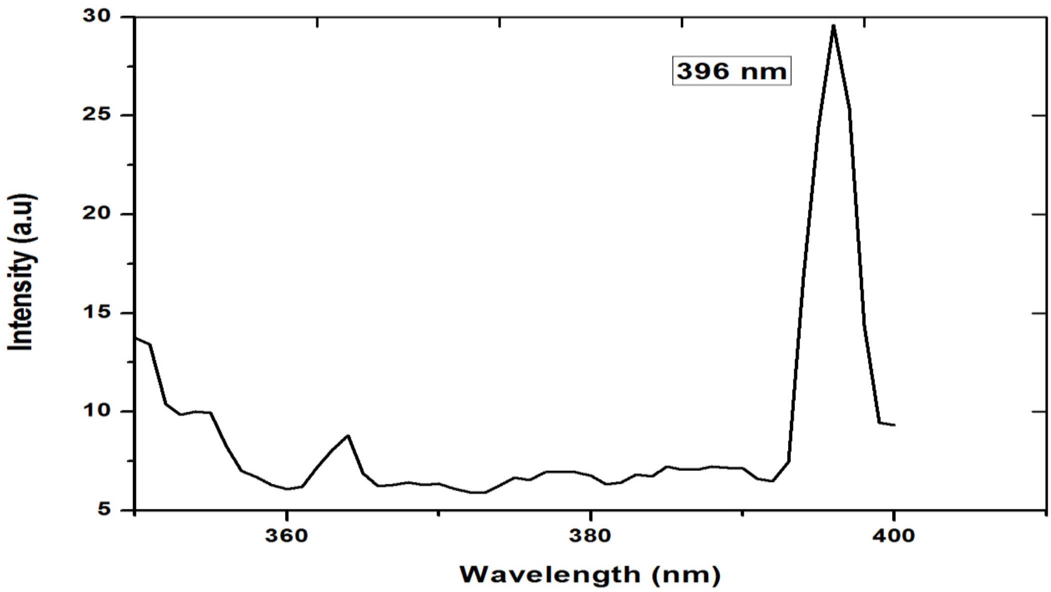

Fig.1 Exitation spectrum $Na_2Ba_4(PO_4)Cl:Eu^{3+}$ Phosphor monitore at 616nm

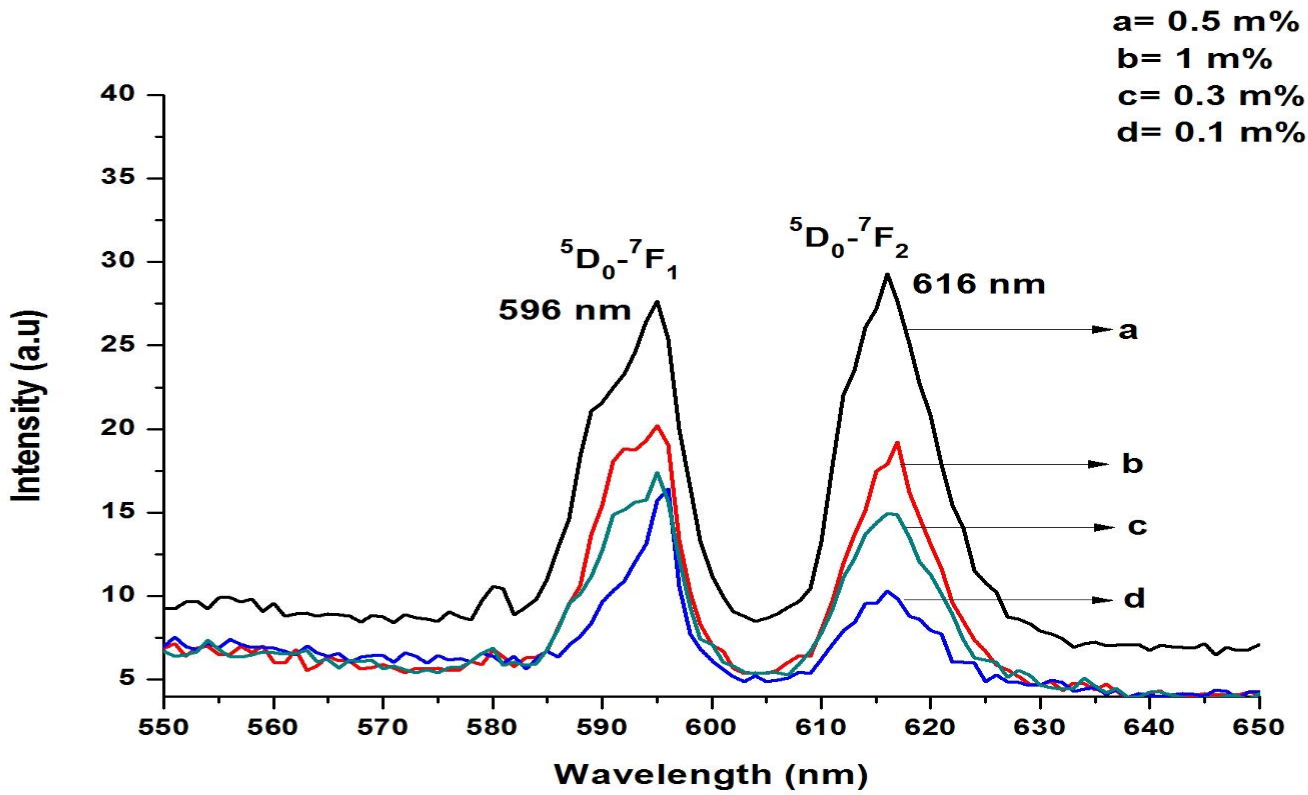

Fig. 3: Emission spectra of $Na_{2-x}Ba_4(PO_4)Cl$: Eu_x^{3+} phosphor ($0.1 \leq x \leq 1.0$) (excited at 396 nm).

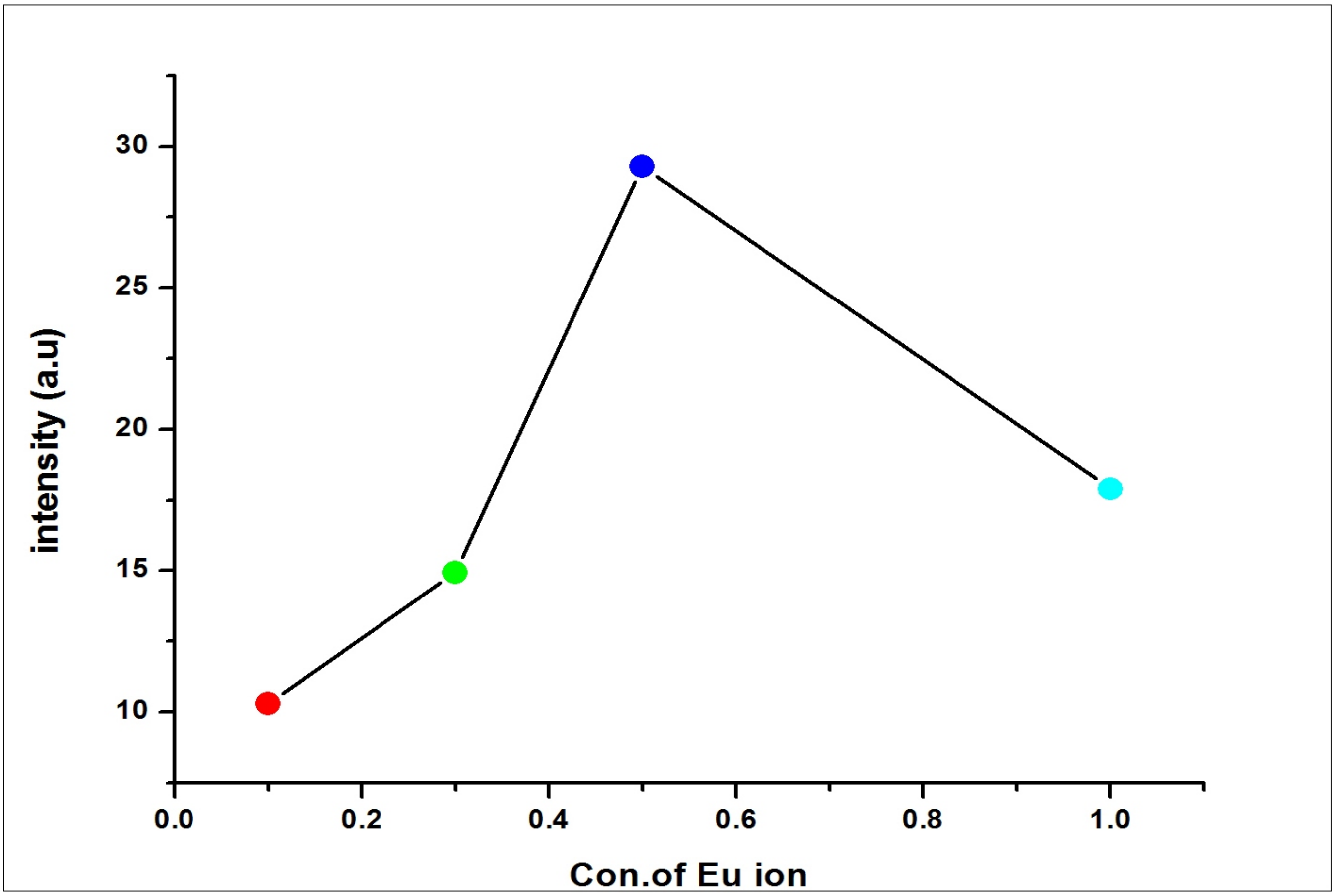

Fig. 4: Variation in the PL intensity due to Eu^{3+} ion concentrations

CONCLUSION

In the present work, the Eu^{3+} ions activated $Na_{2-x}Ba_4(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor have been synthesized by combustion method. SEM images show with average crystallite size in sub-micrometer. The spherical and agglomerated particles are observed. The PL emission is strongly observed in the red region of the spectrum due to transition of Eu^{3+} ions in the $Na_{2-x}Ba_4(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor. The strong red emission is observed in $Na_{2-x}Ba_4(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor by near UV excitation. This is useful as red component in near UV LED applications. Orange/red emission observed in $Na_{2-x}Ba_4(PO_4)Cl$ where x=0.1, 0.3,0.5 and 1m% phosphor due to transitions from $^5D_0$ excited states to ^7F_J (J=0–4) ground states of Eu^{3+} ions under the 396 nm and it is more favorable of solid state lighting.

Conflicts of interest: The authors stated that no conflicts of interest.

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