



## Photoluminescence properties of BaSr(MoO<sub>4</sub>)<sub>2</sub>:Eu<sup>3+</sup> red Phosphor by a Combustion Synthesis Method

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### Abstract

*A novel Eu<sup>3+</sup> activated BaSr(MoO<sub>4</sub>)<sub>2</sub> Phosphor has been synthesized by combustion method. The prepared phosphor crystallized in scheelite-type tetragonal structure. The incorporation of Mo<sup>6+</sup> into the BaSr(MoO<sub>4</sub>)<sub>2</sub> host matrix can broaden its excitation range and promote tunability to its emission. Photoluminescence (PL) characteristics of the samples has been investigated. The excitation spectra show that the phosphor can be efficiently excited by UV-visible light from 220 nm to 410 nm and exhibited bright red emission when excited with 265 nm UV light. The prepared phosphor gives strong red emission peaking at 610 nm due to characteristic <sup>5</sup>D<sub>0</sub>→<sup>7</sup>F<sub>2</sub> electric dipole transition of Eu<sup>3+</sup> ions. The influence of Eu<sup>3+</sup> concentration on the luminescent properties of Eu<sup>3+</sup> doped BaSr(MoO<sub>4</sub>)<sub>2</sub> was investigated and 5% (mole fraction) was the appropriate dopant molar concentration. The reaction time and temperature had obvious effect on the luminescent properties. The phosphor may be serve as near UV InGaN chip-based red-emitting LED phosphors.*

**Keywords:** Phosphors; Combustion Method; BaSr(MoO<sub>4</sub>)<sub>2</sub>:Eu<sup>3+</sup> Luminescence

### 1. INTRODUCTION:

Solid State lighting is superior in lifetime efficiency, reliability and energy conservation than the conventional incandescent lamps and fluorescent lamps. So the LED based white light sources have been developed rapidly in the last ten years, since the first blue LED is invented in 1996 [1,2]. Especially, white light emission, wherein a blue LED is combined with yellow YAG:Ce<sup>3+</sup> phosphor, has been investigated extensively. Commercialization has been realized due to their applications, such as backlighting for liquid crystal displays and illumination [3-5]. The molybdates with scheelite-type tetragonal structure are characterized by the general formula ABO<sub>4</sub> (A = Ca, Sr, Ba, Pb; B = Mo), space group I4<sub>1</sub>/a, and symmetry C<sub>4h</sub><sup>6</sup> [6, 7]. These materials have been prepared in both powder and film forms by means of several technologies, such as electrochemical method [8], hydrothermal [9], solid-state reaction [10], and sol-gel method [11]. However, these approaches still have some limitations; for example, the as-prepared samples are not only irregular in morphology and large in particle size, but also small and the production cost is high.

Recently, researchers mostly have devoted attention to unit material systems. Shi et al. [12] synthesized Ca<sub>0.5</sub>Sr<sub>0.5</sub>MoO<sub>4</sub>:Eu<sup>3+</sup> powder using a sol-gel method. Few reports on improving the luminescent property of solid-solution powders were reported. In order to fully examine their properties,

the study of alkaline earth molybdate materials is necessary.

Molybdate crystals are presently the subject of considerable interest and detailed evaluation because of their valuable structural, thermal, electrical, and spectroscopic properties which are promising for practical applications in advanced electronic and photonic technologies [13, 14]. The rare-earth molybdate crystals or those doped with rare-earth ions are of special interest due to their specific spectroscopic properties and potential application as laser and luminescent hosts [15]. As a rule, the rare-earth ions in molybdates are in low-symmetry positions; The crystals containing trivalent europium ions are of particular interest, because these ions produce efficient red photoluminescence necessary for the creation of white-light emitting diodes (WLED) devices with the spectral characteristics similar to sun daylight [16-23]. Trivalent europium is a preferable activator because it has a good emission spectrum in red region. Thus, it is necessary to choose a good host material to fabricate red phosphor used for GaN based LED. As is well known, molybdate belongs to scheelite compound: the central Mo ion coordinates with four O<sup>2-</sup> ions in tetrahedral symmetry. Hence molybdate is relatively stable and has been opted as a host material [24-26].

In this paper trivalent europium ion (Eu<sup>3+</sup>) activated Barium strontium molybdate [BaSr(MoO<sub>4</sub>)<sub>2</sub>] phosphors was prepared by

combustion synthesis and its luminescent properties were investigated.

## 2. EXPERIMENTAL

Stoichiometric amount of Strontium nitrate  $\text{Sr}(\text{NO}_3)_2$ , Ammonium Molybdate  $(\text{NH}_4)_2\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ , Barium Nitrate  $\text{Ba}(\text{NO}_3)_2$  were taken in a mortar pestle. Then various amount equimolar mixture of Urea  $(\text{NH}_2\text{CONH}_2)$  was added to it. Then europium oxide  $\text{Eu}_2\text{O}_3$  (5 M%) dissolved in Con.  $\text{HNO}_3$  was added and heating on magnetic stirrer to prepare Europium Nitrate then whole constituent was crushed about 2 hour until pasty mass was formed as most nitrate and urea are hygroscopic and absorb moisture during mixing. The absorption of moisture is not undesirable, on contrary it helps to form a homogeneous mixture. These mixtures were transferred to china crucible and placed in a furnace maintained at temperature  $550^\circ\text{C}$ . The product is formed along with foam with evolution of gases. Flame was observed. At the completion of reaction, the china crucible was removed from the furnace and this sample was crushed in mortar pestle to obtain Eu activated  $\text{BaSr}(\text{MoO}_4)_2$  phosphor.

## 3. RESULTS AND DISCUSSION

### PL Characteristics of $\text{BaSr}(\text{MoO}_4)_2$ Phosphor:

PL spectra are useful in identifying the valence state of the Eu ion.  $\text{Eu}^{3+}$  gives characteristic emission in the orange-red region of the spectrum corresponding to the transitions  $^5\text{D}_0 \rightarrow ^7\text{F}_2$ . These transitions are parity forbidden.  $\text{Eu}^{2+}$  emission, on the other hand, is lattice dependent and it originates from  $4f_6^5d \rightarrow 4f_7$  transition.

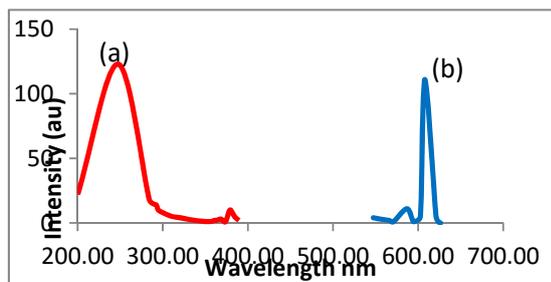


Fig. (1): PL emission spectra of  $\text{BaSr}(\text{MoO}_4)_2$ :  $0.05 \text{Eu}^{3+}$  monitored at 268 nm excitation.

Fig.(1) shows the excitation spectrum of  $\text{BaSr}(\text{MoO}_4)_2$ :  $\text{Eu}^{3+}$ , monitored at 610 nm. It shows broad bands with the maximum at about 268 nm, which can be attributed to the charge transfer state (CTS) this is related to the excitation of an electron from the oxygen 2p state to a  $\text{Eu}^{3+} 4f$  state [27].  $\text{Eu}^{3+}$  PL is in general weak, unless there is excitation by charge transfer or energy transfer from a sensitizer.  $\text{Eu}^{3+}$  excitation usually occurs through CT band or from host lattice absorption and energy transfer,

rather than through 4f-5d absorption, though this is also possible in some solids by UV radiation in the region 200 – 250 nm. Sometimes, absorption within 4f shell also takes place as the spin and parity prohibitions are partially lifted by mixing of levels promoted by crystal field. Blasse et. al. [28-31] studied  $\text{Eu}^{3+}$  emission extensively in various hosts. In general, narrow emission bands may be observed at about 570, 590, 610, 650 and 700nm corresponding to transitions  $^5\text{D}_0 \rightarrow ^7\text{F}_0$ ,  $^7\text{F}_1$ ,  $^7\text{F}_2$ ,  $^7\text{F}_3$ ,  $^7\text{F}_4$ , respectively.  $\text{Eu}^{3+}$  emission usually occurs from  $^5\text{D}_0 \rightarrow ^7\text{F}_j$  transitions. There are three transitions which are of prime importance  $^5\text{D}_0 \rightarrow ^7\text{F}_0$  (around 570 nm),  $^5\text{D}_0 \rightarrow ^7\text{F}_1$  (around 595 nm) and  $^5\text{D}_0 \rightarrow ^7\text{F}_2$  (around 610 nm). The first one is strongly forbidden transition and yet observed with appreciable intensity in some hosts.  $^5\text{D}_0 \rightarrow ^7\text{F}_1$  transition is forbidden as electric dipole, but allowed as magnetic dipole. This is the only transition when  $\text{Eu}^{3+}$  occupies a site coinciding with a centre of symmetry. When  $\text{Eu}^{3+}$  ion is situated at a site, which lacks the inversion symmetry, then the transitions corresponding to even values of J (except 0) are electric dipole allowed and red emission can be observed.  $^5\text{D}_0 \rightarrow ^7\text{F}_1$  transition can also be observed as magnetic dipole allowed transition. Further, all the lines corresponding to these transitions split into number of components decided by the local symmetry [32]. The spectra are also sensitive to sizes of cations [33] and chemical bonding [34].

The most intense line at 610 nm corresponds to the hypersensitive transition between the  $^5\text{D}_0$  and  $^7\text{F}_j$  level of  $\text{Eu}^{3+}$  ion, which is electric dipole forbidden and sensitive to the ligand environment. The highest photoluminescence intensity was found at a doped- $\text{Eu}^{3+}$  Concentration of 0.05 mol fraction.

FIG. [2] shows the CIE diagram indicating the color coordinates from emission spectrum (under 268 nm excitation) of  $\text{BaSr}(\text{MoO}_4)_2$ :  $0.05 \text{Eu}^{3+}$  phosphors and the NTSC standard value. It can be clearly seen that the color coordinates of  $\text{BaSr}(\text{MoO}_4)_2$ :  $0.05 \text{Eu}^{3+}$  phosphor is very close to the NTSC standard value. Hence  $\text{BaSr}(\text{MoO}_4)_2$ :  $0.05 \text{Eu}^{3+}$  phosphor is considered as promising red phosphor for near UV InGaN chip-based red-emitting LED light.

## 4. CONCLUSION

The red phosphor  $\text{BaSr}(\text{MoO}_4)_2$ : $\text{Eu}^{3+}$  was successfully fabricated with combustion synthesis method. Its excitation spectrum covered the range between 200-400 nm it was excited effectively at 268 nm. Its emission was composed of some sharp lines, characteristic of  $\text{Eu}^{3+}$ . It emitted red light located at 610nm; the CIE chromaticity coordinates ( $x=0.661$ ,  $y=0.333$ ) are close to the NTSC standard values



( $x=0.66$ ,  $y=0.33$ ), and it may be serve as near UV InGaN chip-based red-emitting LED phosphors.

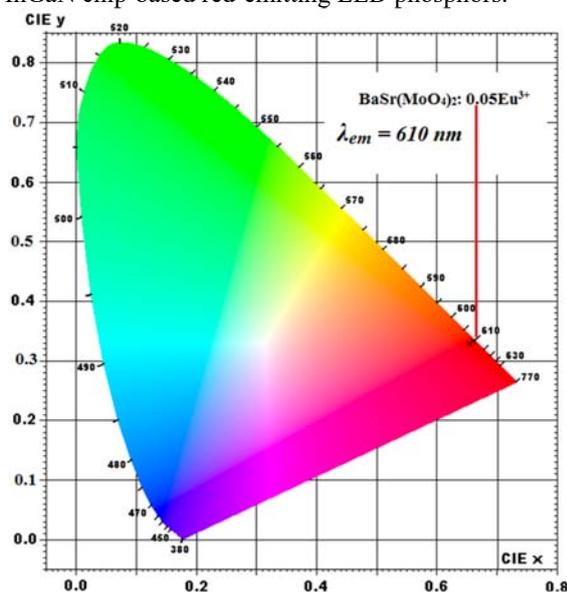


FIG.2 CIE DIAGRAM INDICATING THE COLOR COORDINATES FROM EMISSION SPECTRUM (UNDER 268 nm EXCITATION) of  $\text{BaSr}(\text{MoO}_4)_2:\text{Eu}^{3+}$  PHOSPHORS and THE NTSC STANDARD VALUE.

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