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Preparation and Luminescent Properties of $Sr_3B_2O_6$: Eu^{3+} Phosphor

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Abstract*— Eu^{3+} ion doped $Sr_3B_2O_6$ phosphors were prepared by combustion method at 590°C in 6 minutes with Eu^{3+} ion concentration was changed from 0,5 to 7%mol, later the phosphors were annealed at temperature 800, 900, 1000 and 1100°C in the air. The crystalline structure of product was confirmed by X-ray diagram. The effects of annealing temperature and concentration of Eu^{3+} ion on luminescent properties were investigated by the photoluminescence and the excitation spectra.

***Index Terms*— Europium, $Sr_3B_2O_6$, red phosphors, combustion.**

I. INTRODUCTION

The energy levels of Eu^{3+} ion have been investigated fully in many host lattices. The emission of Eu^{3+} ion which originates from transitions between 4f levels is predominantly due to electric - dipole or magnetic – dipole interaction [1], [2], [3]. In case an Eu^{3+} ion positions at a site in crystal lattice which has inversion symmetry, magnetic-dipole transitions $^5D_0$-$^7F_1$ dominate within emission transitions between levels of the $4f^n$ configuration. In the opposite case, if there is no inversion symmetry in the site, electric-dipole transitions $^5D_0$-$^7F_2$ is dominant [2]. Eu^{3+} ion is used as a structural sensor besides it is also used as activator for red-emitting phosphor, such as Y_2O_3:Eu^{3+}, $Ba_2Mg(BO_3)_2$:Eu^{3+} [2], [4].

Among many materials using as host lattices to dope Eu^{3+}, borates are good candidates as a host structure due to their low synthetic temperature and easy preparation [2], [5], [6], [7]. Besides, combustion method was used to synthesis this material in short time and low temperature [5], [8]. This paper presents the effect of annealing temperature and Eu^{3+} concentration on luminescent properties of $Sr_3B_2O_6$:Eu^{3+} phosphors.

II. EXPERIMENTAL

In first target, to survey the effect of annealing temperature on luminescent properties, the $Sr_3B_2O_6$ phosphors doped with x = 0.5%mol Eu^{3+} ions were fabricated by urea - nitrate solution combustion method. The initial materials include $Sr(NO_3)_2$ (Merk), H_3BO_3 (AR), $Eu(NO_3)_3$ (Sigma). Urea (NH_4NO_2) was used to supply fuel for combustion process. The theoretical equation for the formation of $Sr_3B_2O_6$:Eu^{3+} of these phosphors is shown in below Equation:

$$(3-x)Sr(NO_3)_2 + xEu(NO_3)_3 + 2.H_3BO_3 + 6.67CH_4N_2O \rightarrow Sr_{(3-x)}B_2O_6 : xEu^{3+}$$

After that process the precursors were mixed and heated at 70°C by magnetic stirrer in 3 hours to become a gel. Gel was annealed at 590°C in 5 munites then it was decreased down to room temperature, obtained powder was white. Next step, this powder was heated at 800, 900, 1000 and 1100°C in air in 1 hour to obtain final products.

In second target, investigation the effect of Eu^{3+} concentration on luminescent properties, the $Sr_3B_2O_6$ phosphors were synthesized with various Eu^{3+} concentration from 0,5 to 7%mol and annealing temperature at 900°C.

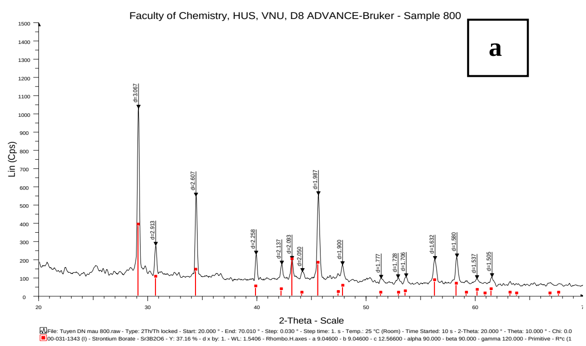
The structure of the synthesized products were characterized by X-ray diffraction (XRD) using a Bruker D8-Advance X-ray diffractometer. The emission and excitation spectra were recorded by a Fluorog 3-22 (Horiba Jobin-Yvon) with 450W Xe lamp at room temperature.

III. RESULTS AND DISCUSSIONS

A. Influence of the annealing temperature on luminescent properties of $Sr_3B_2O_6$:Eu^{3+}.

To demonstrate the pure phase and structure of $Sr_3B_2O_6$:Eu^{3+} phosphors, powder X-ray diffraction (XRD) measurements of these samples were carried out and shown in the Fig 1. It is clearly observed that all samples have Rhombo single phase.

The photoluminescence emission (PL) spectra of samples annealed at different temperatures under excited radiation $\lambda_{ex} = 394nm$ are shown in Fig 2(a). The peaks in PL spectra are ascribed to the transitions of electron from excited state $^5D_0$ to ground state ^7F_j (j=0, 1, 2, 3, 4) within Eu^{3+} ion. The intensity of allowed electric dipole transition $^5D_0$-$^7F_2$ (610 nm) was presented in Fig 2(b). The samples annealed at 900°C and 1000°C have the highest intensity.



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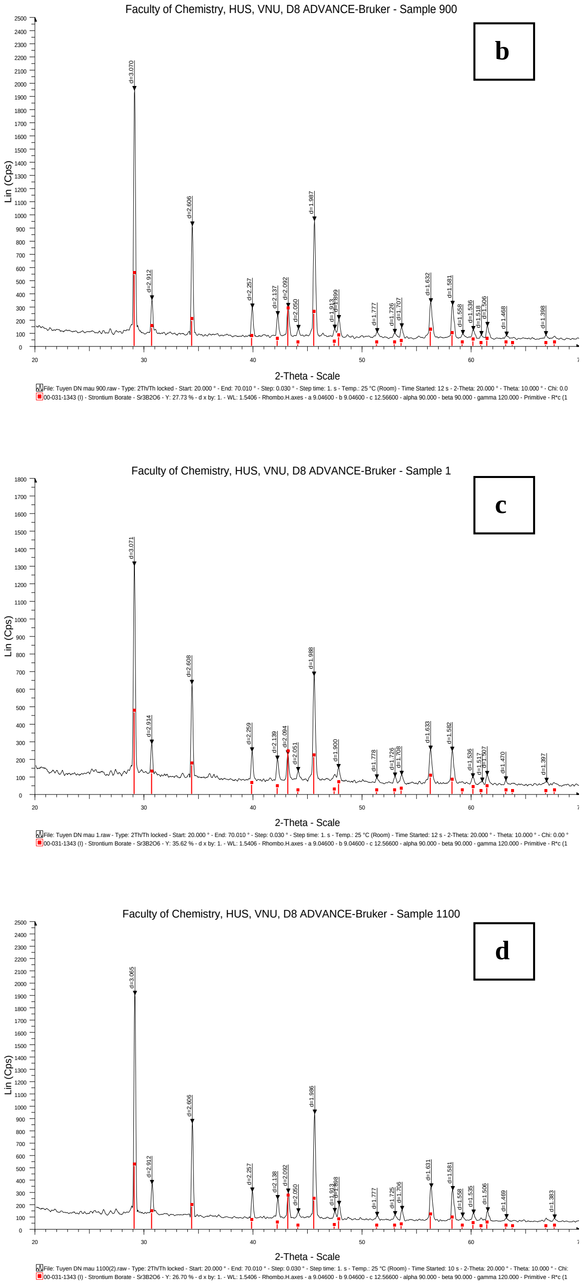

Fig. 1. XRD of samples annealed at the different temperature (a) 800°C, (b) 900°C, (c) 1000°C, (d) 1100°C.

On another hand, luminescent efficiency of Eu^{3+} in $Sr_3B_2O_6:Eu^{3+}$ phosphors is obtained from R ratio which is defined by $R=I(^5D_0\text{-}^7F_2)/I(^5D_0\text{-}^7F_1)$. In optical transition of Eu^{3+} ion, $^5D_0\text{-}^7F_2$ is an allowed electric dipole transition and its intensity is hypersensitive to the variation of the bonding environment of the Eu^{3+} ions. While $^5D_0\text{-}^7F_1$ is a magnetic dipole allowed transition and its emission intensity is hard to change with the bonding environment of the Eu^{3+} ions [3], [9]. Thus, intensity ratio R is also used to know the bonding environment of Eu^{3+} ions. The luminescent efficiency R which changes over annealing temperature of $Sr_3B_2O_6:Eu^{3+}$ phosphors was showed in Table I. The change of luminescent efficiency among these samples is little, sample with annealing temperature at 900°C is better than other samples.

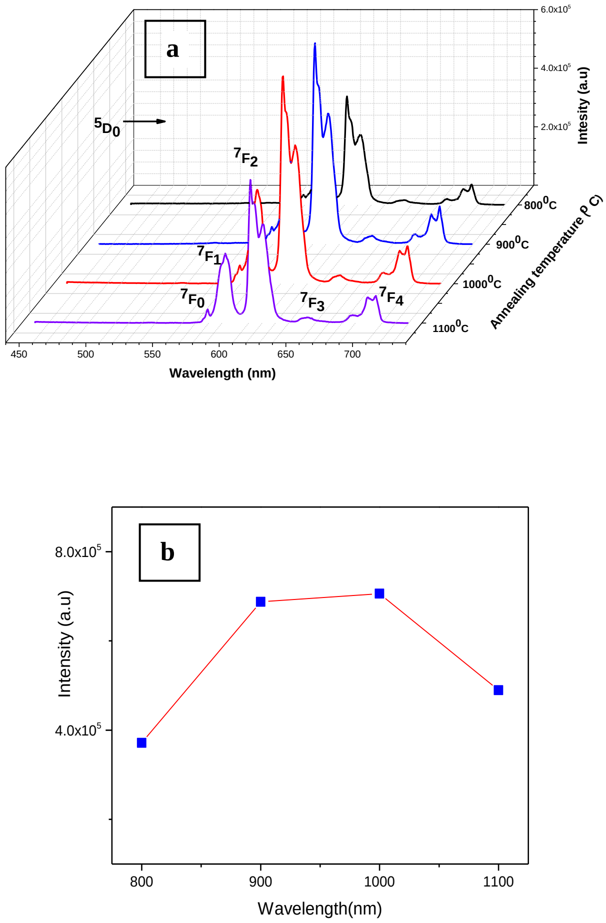

Fig. 2. PL spectra (a) and $^5D_0\text{-}^7F_2$ intensity (b) of samples annealed at different temperature.

TABLE I. R RATIO OF SAMPLES ANNEALED AT THE DIFFERENT TEMPERATURE

Samples	800°C	900°C	1000°C	1100°C
R ratio	2.66	2.67	2.51	2.34

Photo luminescent excitation (PLE) spectra monitored at 610 nm of $Sr_3B_2O_6:Eu^{3+}$ phosphors that annealed at the different temperature are shown in Fig 3. The excitation spectra consist of a broad band and several narrow lines. This broad band in a range from 250 to 310 nm is due to charge-transfer band of the Eu^{3+} ion [9]. And narrow lines located at about 320, 363, 383, 394, 416, 466 and 526 nm are attributed to the f-f transitions within the $4f^6$ configuration of Eu^{3+} ion. In temperature range from 800 to 1100°C, the excitation transition $^7F_0\text{-}^5D_6$ (394nm) of sample annealed at 900°C gets the highest intensity. As shown on XRD, PLE and PL spectra, the optimum temperature to synthesis the $Sr_3B_2O_6:Eu^{3+}$ red phosphor is 900°C.

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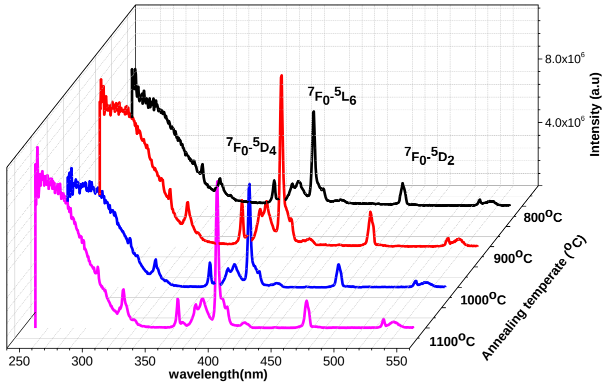
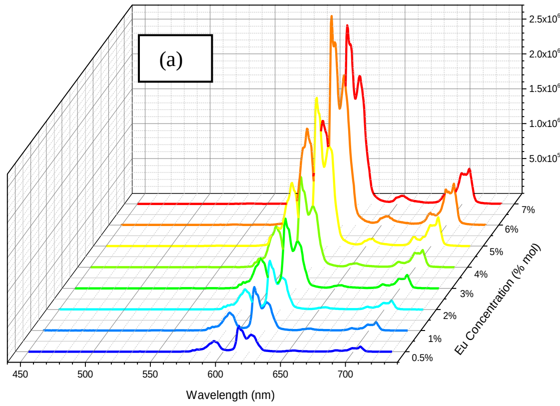


Fig. 3. PLE spectra of samples annealed at different temperature.

B. Influence of Eu^{3+} ion concentration on luminescent properties of $Sr_3B_2O_6$: Eu^{3+}

Phosphors were fabricated with different concentration of Eu^{3+} ion at the annealing temperature 900^{o}C. Fig 4 shows the PL spectra (a) and maximum intensity of transition $^5D_0$-$^7F_2$ (610nm) (b) of samples with various Eu^{3+} concentrations from 0.5 to 7%mol.



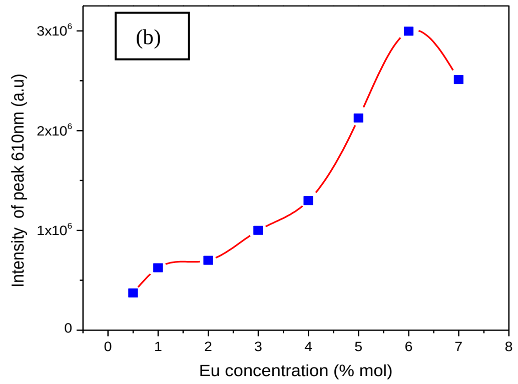


Fig.4. PL spectra (a) and $^5D_0$-$^7F_2$ intensity (b) of samples with various Eu^{3+} ion doped concentration.

The intensity of $^5D_0$-$^7F_2$ (610nm) transition increases and reaches maximum at 6%mol. But when Eu^{3+}concentration is more than 6%mol luminescent intensity decreases, it is due to the concentration quenching phenomenon. Besides, R ratios of these phosphors were listed on the Table II. The sample doped 6%mol Eu^{3+} ion has optimum luminescent efficiency.

TABLE II. R RATIOS OF SAMPLES WITH VARIOUS Eu^{3+} CONCENTRATION

Samples	0.5 %mol	1 %mol	2 %mol	3 %mol	4 %mol	5 %mol	6 %mol	7 %mol
R ratio	2.63	2.84	2.82	2.81	2.55	2.81	3.11	2.85

The PLE spectra of the $Sr_3B_2O_6$:Eu^{3+} phosphors which were fabricated with the different concentration of Eu^{3+} ion is shown in Fig 5. The excitation spectra indicates that the intensity of narrow line at 394nm ($^7F_0$ - $^5L_6$ transition of Eu^{3+} ion) changes over concentration.

In addition, the structure of $Sr_3B_2O_6$:Eu^{3+} (5%mol) was also confirmed by XRD diagram which presents in the Fig 6. From the XRD diagram we can see that this concentration (5%mol) of Eu^{3+} does not have any affect on the structure of the material.

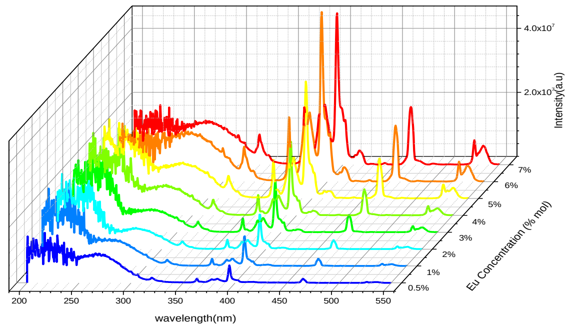


Fig. 5. PLE spectra of samples with changed concentration of Eu^{3+} ion.

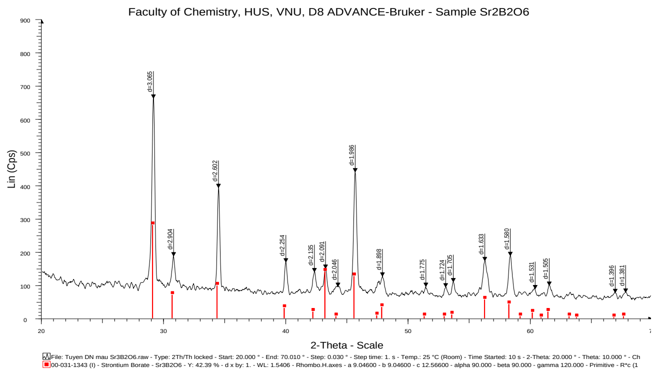


Fig. 6. XRD diagram of $Sr_3B_2O_6$: Eu^{3+} (5%mol).

IV. CONCLUSIONS

$Sr_3B_2O_6$:Eu^{3+} red emitting phosphors were prepared by urea-nitrate solution combustion method, after combustion

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process samples were annealed in air. These phosphors have rhombohedral single phase structure. The results from PL spectra and R ratio display that the intensity $^5D_0$-$^7F_2$ (610nm) of phosphors changes over annealing temperature and achieves the highest intensity at 900^{o}C. With the increase of Eu^{3+} concentration, the emission intensity of the phosphors increased but maximum emission wavelength of spectra did not change. Optimum concentration of Eu^{3+} ion was 6 %mol in order to achieve the highest emission.

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