

Combustion Synthesis and PL Studies of Red Emitting $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$: Eu^{3+} Lamp Phosphors

G. R. Rahate¹, U. A. Thakare², A. B. Lad³, V. R. Panse⁴, K. V. Sharma⁵

^{1,5}Department of Basic Science & Humanities G. H. Rasoni University, Amravati, Maharashtra, India

²Department of Physics, S. P. M. Science and Gilani Arts, Commerce College Ghatanji Maharashtra, India

³Department of Physics, Amolakchand Mahavidyalaya, Yavatmal, Maharashtra, India

⁴Department of Physics, Late B. S. Arts Prof. N. G. Sci & A. G. Comm College Sakharherda, Maharashtra, India

*Corresponding author : gauravrahategd@gmail.com

ABSTRACT

$\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ luminescent powdered phosphors doped with rare earth elements Eu^{3+} was successfully synthesized using a wet chemical method to identify the most suitable phosphor based on the measurement of their emission spectra at excitation wavelengths. The XRD pattern of the phosphor was well matched with its standard JCPDS file. SEM images revealed irregular morphology. PL excitation and emission spectra clearly indicated that the phosphor containing the Eu^{3+} activated $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphors could be efficiently excited at 396 nm and exhibited an emission mainly including two wavelength peaks at 593 nm and 620 nm.

Keywords : XRD pattern, JCPDS, Vacuum Fluorescent Display, Field Emission Display, Plasma Panel Display

I. INTRODUCTION

These materials recombine with high light capacitance, constructive emission wavelength, faster fluorescence decay with stability of temperature which make them attractive for useful in detectors for elevated energy physics [1] and also medical imaging [2]. Therefore, with the support of outstanding luminescence properties, inorganic materials activated with Eu^{3+} ions are applied in lightings industries, detectors with display systems for ionizing radiation [3-4]. To calculate the 5d energies of another rare earth ion in same host lattices material Eu^{3+} ion also is used as a reference ion [5].

So, the spectroscopic properties investigation of Eu^{3+} in various host material is important for both the actual applications and the fundamental research.

Generation of white light either by combination of basic fundamental colors or corresponding colors due to white light obtained from UV-blue LED by covering on LED suitable inorganic phosphors excitable by LED lights, For such inorganic phosphor transformed LED generating white light concluding development of suitable phosphor materials. Available W-LEDs has lower colors re-generation as they are frequently depends on mixing of a blue LED with yellow emitting inorganic phosphor. Such white LEDs require red and green part of the spectrum of white color. Prepared phosphors are composition of an inert host material and an optically excited activated rare earth ion, normally 3d otherwise 4f metallic electron such as Ce^{3+} , Tb^{3+} or Eu^{3+} , Cr^{3+} Oxide inorganic phosphors material have been found a appropriate for vacuum fluorescent display (VFD), electroluminescent (EL) devices, field emission display (FED) and (PDP)

plasma panel display devices [5-6]. D. Ravichandran *et al.*, reports the luminescence study of Eu^{2+} doped alkaline earth aluminates based phosphors, but there is only some work on red emitting Eu^{3+} doped phosphor. Due to their outstanding properties with application such as higher efficiency of luminous, energy saving capabilities, longer life duration, and short of toxic mercury, white light-emitting diodes becomes considered as next generation lighting sources. With the help of blue/NUVLEDs to generate white light by [1-7-8] different approaches are available. To produce the white light Nichia Co., introduces initial W-LED which has been commercialized in which a blue LED is covered with a yellow color phosphor of $\text{YAG}:\text{Ce}^{3+}$.

Though, in the emission spectra, lack of a red component is observed so this kind of white LED suffers from the disadvantage of yellow/blue color partition and deprived color-rendering index (CRI) observed. One more approach is to combine different lights of colors emitted by diverse chips to get white light. Fabrication of white LEDs reported in 1997, by Nakamura with the help of mixing three primary usual color LEDs (i.e. green as well as blue in GaNSQ W-LEDs with a red AlGaAs LED).[9-10] Though, for such type of mechanism, to balance different color degrades among the red, green and blue chips the complicated IC control system is needed. To excite two or tricolor phosphors and to generate white light the third lumen is with the help of NUV or blue LED sources[11]

II. EXPERIMENTAL

The red-emitting $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ phosphors were prepared by wet chemical method at 100°C . The starting materials were used as $\text{Ca}(\text{NO}_3)_2$ (99.99% purity Merck), NH_4F (99.99% purity Merck), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (99.99% purity Merck), $\text{Eu}(\text{NO}_3)_3$ (99.99% purity Merck), and urea's (purity Merck).

The phase for purities of the prepared samples was checked by powder X-ray diffraction (XRD); with the help of PAN-analytical diffractometer with $\text{Cu K}\alpha$ radiation (1.5405 \AA) at an operating voltage at 30mA and 40 kV, and scanning step time at 10.3377's. Photoluminescence (PL) emission for particular excitation were measured at our workplace using a Shimadzu RF5301PC spectrofluorophotometer recorded at room temperature.

III. RESULTS AND DISCUSSION

X-ray diffraction pattern of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ phosphors

For this prepared sample study of the crystalline nature of the phosphor products, XRD measurements were carried out at room temperature. Fig 01 shows the XRD patterns of as prepared $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ host phosphors. The prepared powder compound was characterized for study of its phase purity and crystallinity by X-ray powder diffraction (XRD) using a PAN Analytical X'pert Pro diffractometer XRD diffractometer and found it is in good agreement with JCPDS data source file no. 53-1232. The XRD pattern of the $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ compound is as follows. The XRD-pattern shows phosphor have good crystalline nature.

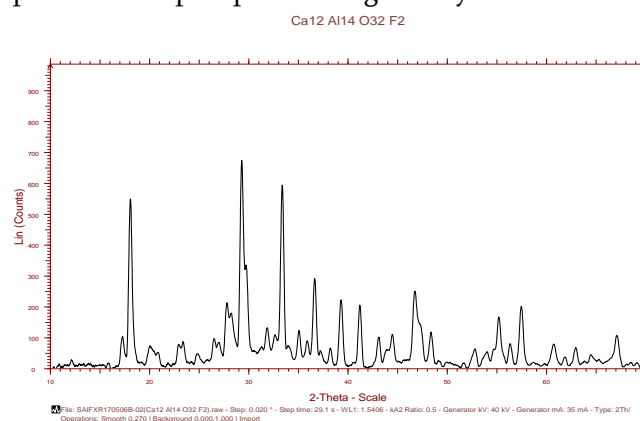


Fig. 01 XRD-pattern of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ lamp phosphors. Morphology of the combustion synthesized $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphors

The SEM images are represented in fig 02 for combustion synthesized $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphor under few micron to sub few micron. The particles

prepared possess like phomy morphology shaped as of highly agglomerated crystallites particles. The usual size of crystallite is in the range of sub micrometer as notified in SEM images, for all the compositions prepared as crystallite sizes are nearly same.

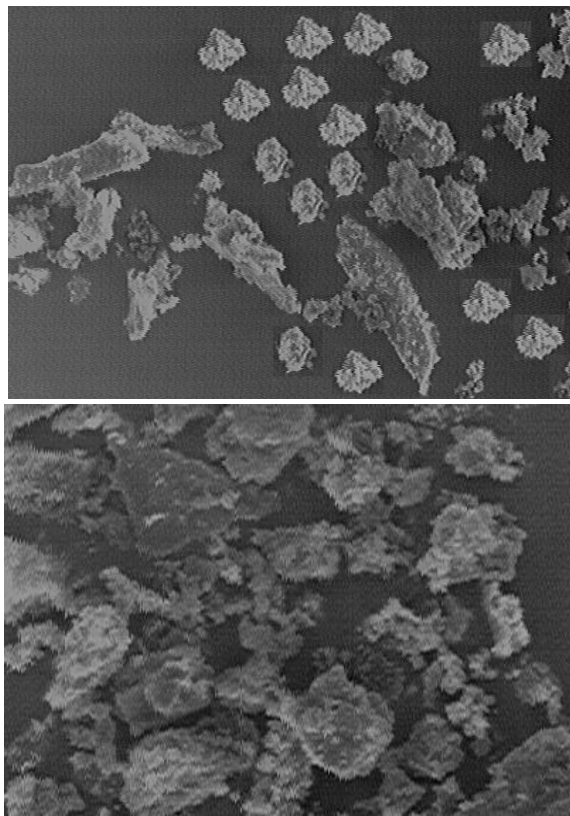


Fig. 02 - Morphology of the combustion synthesized $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphors

Luminescent property of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ Phosphor

Excitation spectrum of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ sample (Eu^{3+} was 1 to 0.1mol.%) prepared using wet chemical synthesis method using urea as a flux . While emission & excitation spectrum showing in fig 03 & 04 showing sharp red emission used for the purpose of solid state lighting. While preparing the material the entire corresponding reagent taken as per the stoichiometric ratio of the prepared composition calculated due to the total oxidizing with reducing fuel valancy which behaves as a statistical and numerical coefficient for the stoichiometric equilibrium position.[12] Excitations spectra appears at 396 nm for the 593nm & 620 nm emission which is Hg free excitation suitable

for the purpose of solid state lighting showing sharp emission peak ,observed in the red visible region.

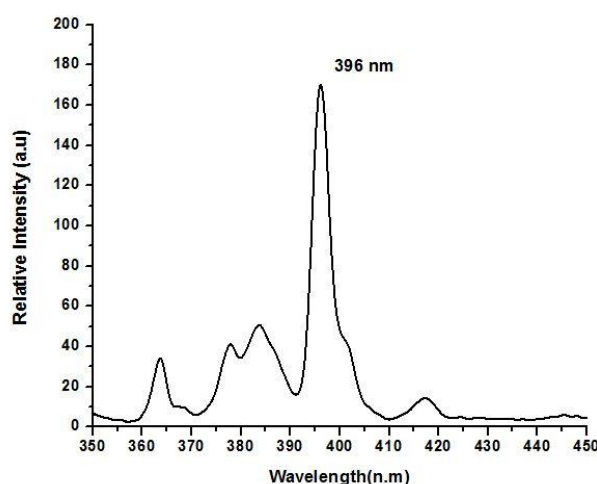


Fig.03 Excitation spectra of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ monitoring the emission at 620 nm

The Eu^{3+} activated $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphor, having doping concentration varying from 1m% to 0.1m % excited by using wavelength 396 nm showing main emission peaks at 593 nm and 620 nm. As the concentration of trivalent europium ion increases, the significant virtual intensity of both transitions such as 593 ($^5\text{D}_0 \rightarrow ^7\text{F}_1$) and 620 ($^5\text{D}_0 \rightarrow ^7\text{F}_2$) increases. From the emission spectrum of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ phosphor , suitable for red emission for the purpose of solid state lighting.

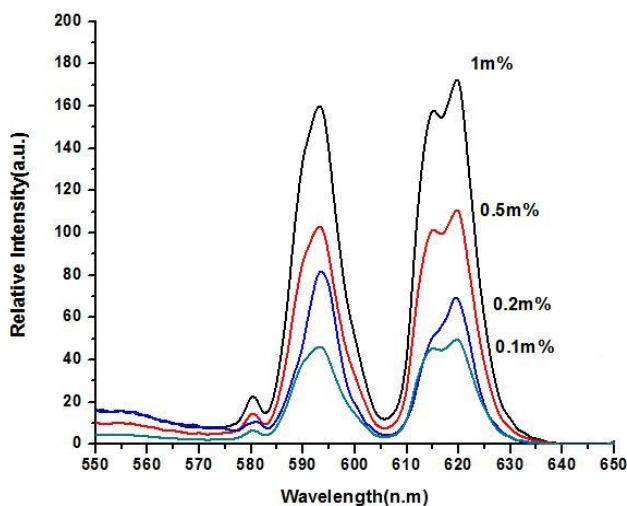


Fig 04 Emission spectra of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ under excitation at 396 nm

Chromatic properties of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ phosphor

We think that as Eu^{3+} is present with host lattice then the quantity of energy can be shifted to the activator ion, resulting from the distinctive unique emission peak of these activator ions. [13] It is renowned fact that the outcome obtained from the luminescence properties of prepared inorganic phosphors in powder form depends on the concentration of activator ion, therefore the recognition with concentration of dopant is necessary [14]. Consider the emission spectrum of Eu^{3+} which was located in red region, was chosen for further analytical study and characterization, the luminescent properties of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ red emitting phosphors and achieve the complete emission of color. Here we determine the coordinate of chromaticity indexed with the help of the emission spectra of Eu^{3+} .

IV. CONCLUSION

The investigation on the photoluminescence characteristics of Eu^{3+} activated red-emitting $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphors in the near UV-vis range shows the excitation bands at 396 nm, respectively. Emission characteristics of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ shows broad emission band at 593 nm & 620 nm. XRD-pattern of prepared $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2$ phosphor indicates the good crystalline nature. Scanning electron microscopic images shows morphology of the phosphor at microns to sub few microns. The prepared phosphors have prospective red application for the lamp industries.

V. REFERENCES

- [1]. J. J. Carvajal, I. Parreu, F. D'iaz, X. Solans, M. Aguil'o, Chem. Mater. 18 (2006) 221.
- [2]. Jna. Gavalda, I. Parreu, R. Sol'e, J. Massons, F. D'iaz, M. Aguil'o, Chem. Mater. 15 (2003) 5059.
- [3]. K. N. Shinde and S. J. Dhoble, Adv. Mat. Lett., 1(3) (2010) 254.
- [4]. E. Tadashi, Y. H. Wang, L. He, C. F. Wu, J. Cryst. Growth. 268 (2004) 568.
- [5]. W. Qin. Y. Jin, J. Solid State Chem. 181 (2008) 724.
- [6]. G. Panzer, R. Ternane, M T H Cohen-Adad. Opt. Mater. 16 (2001) 291.
- [7]. I. M. Nagpure, S. J. Dhoble, Manoj Mohapatra, Vinay Kumar, Shreyas
- [8]. S. Pitale, O. M. Ntwaeaborwa, S. V. Godbole, H. C. Swart, J. Alloys Compd. 509 (2011) 2544.
- [9]. K. N. Shinde, S. J. Dhoble, Animesh Kumar, J. Lumin. 131 (2011) 931.
- [10]. X. Yan, W. Li, K. Sun, Mater. Res. Bull. 46 (2011) 87.

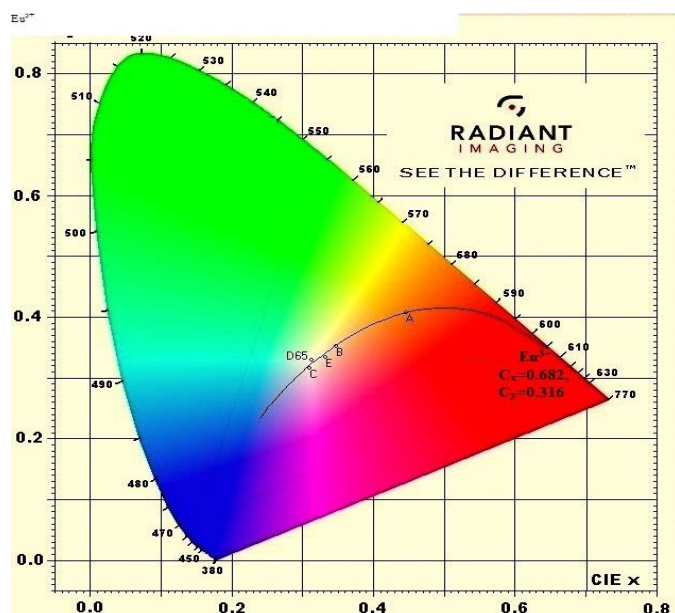


Fig 05. CIE chromatic diagram for $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ phosphor.

The CIE diagram of $\text{Ca}_{12}\text{Al}_{14}\text{O}_{32}\text{F}_2:\text{Eu}^{3+}$ red emitting phosphor shown in $0.682 C_y = 0.316$. With the help of CIE diagram it is easy to explain that the $\text{CaAl}_{10}\text{O}_{15}\text{F}_2:\text{Eu}^{3+}$ phosphors are very near to the CIE graph frame, which easily shows the utmost color clarity of prepared phosphor material.

- [11]. D. A. Steigerwald, J. C. Bhat, D. Collins, R. M. Fletcher, M. Ochiai Holcomb, M. J. Ludowise, P. S. Martin, S. L. Rudaz, IEEE J. Quantum. Elect. 8 (2002) 310.
- [12]. K. S. Sohn, B. I. Kim, N. Shin, J. Electrochem. Soc. 151 (2004) H243.
- [13]. Y. Hu, W. Zhuang, H. Ye, S. Zhang, Y. Fang, X. Huang., J. Lumin. 111 (2005) 139.
- [14]. S. H. M. Poort, W. Janssen, G. Blasse, J. Alloys Compd. 260 (1997) 93.
- [15]. F. Gu, S. F. Shu, M. K. Lu, G. J. Zhou, S. W. Liu, D. Xu, D. R. Yuan, Chem. Phys. Lett. 380 (2003) 185.