© 2017 IJSRST | Volume 3 | Issue 8 | Print ISSN: 2395-6011 | Online ISSN: 2395-602X
Themed Section: Science and Technology

Photoluminescence Analytical Study of Eu^{3+} Activated Red Emitting $Ca_{14}Al_{10}Zn_6O_{35}$ Lamp Phosphors

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ABSTRACT

In this work we synthesized the Eu^{3+} doped $Ca_{14}Al_{10}Zn_6O_{35}$ phosphor using solution combustion method. Using powder XRD pattern and SEM was confirmed. Intrinsic absorption due to 4f-4f transition of Eu^{3+} results intense red emission from α-$Ba_3Y_{1-x}(BO_3)$:xEu^{3+}makes it suitable for pc-WLED and confirms centro-inversion symmetry site of Eu^{3+} in host. Red emission at NUV excitation from $Ca_{14}Al_{10}Zn_6O_{35}$: Eu^{3+}results from cross relaxation of Eu^{3+}in host Entire work confirms the importance of synthesized phosphor along with previously reported same host materials. The reported phosphor may be suitable for NUV converted WLED, wavelength conversion devices and high power RGB –WLED.

Keywords : Solution Combustion, Europium, Photo-Luminescence

I. INTRODUCTION

White LEDs based on inorganic phosphors are widely used due to their properties like high energy efficiency, long lifetime,[1-2] dependable stability, Cost-effective, design-friendly, eco-friendly nature. Inorganic luminescent materials have considerable importance in fluorescent lamps, solid-state lighting sources, displays. [3-4-5]Wavelength conversion and color mixing are the most commonly used technology to produce white light [1-6-7]. So the development of efficient phosphors for the wide possible applications is the favorite research area of material science. Rare-earth ions are the most familiar dopants used in the synthesis of inorganic phosphor because of abundant energy levels and the number of possible transitions in the visible/UV light region[7-8]. Rare earth ion, Eu^{3+} is one of the most promising red-emitting activators due to transitions and Eu^{3}+ is the promising red-emitting activator dopant [8-9].

The aluminates compound with formula $Ca_{14}Al_{10}Zn_6O_{35}$ were reported as promising phosphors for W-LED applications. [10-11]The photoluminescence properties under doping are different for both phases. The structure of low temperature phase $Ca_{14}Al_{10}Zn_6O_{35}$ with space group P63 cm is composed of three-fold coordinated atoms, six-fold coordinated yttrium atoms, nine- and six-fold barium atoms [12-13]]. Irish Valerie et.al synthesized the $Ca_{14}Al_{10}Zn_6O_{35}$: Eu^{3+}phosphor via a wet chemical reaction and studied it as a phosphor for w-LEDs using N-UV LED chips[10]. Xiulan Wu.et.al. described $Ca_{14}Al_{10}Zn_6O_{35}$: Eu^{3+}as potential orange –red phosphors for white LEDs[11].Jingjie Yu et.al.

investigated luminescent and energy transfer in α-$Ba_3Y(BO_3)_3:Ce^{3+},Tb^{3+}$[12-14-15].

Luminescent Research groups from all over the world are working for efficient phosphors for WLED had studied and proved the importance of borate host $Ca_{14}Al_{10}Zn_6O_{35}$ But synthesis of $Ca_{14}Al_{10}Zn_6O_{35}$ phosphor by solution combustion method is new approach. The luminescent properties of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ prepared by solution combustion method is not described yet. $Ca_{14}Al_{10}Zn_6O_{35}$ has been investigated for nonlinear optical applications and studied for w-LEDs applications by doping it with Eu^{3+} Photoluminescence (PL) properties, color chromaticity, Energy transfer mechanism for Eu^{3+} is also investigated.

II. Experimental

The blue-emitting $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ phosphors were prepared by solution combustion method. The starting materials were used as $CaNO_3$ (99.99% purity Merck), $Al(NO_3)_3.9H_2O$ (99.99% purity Merck), $ZnNO_3$ (99.99% purity Merck), $(NH_4)_2Ce(NO_3)_6$ (99.99% purity Merck), and ureas (purity Merck).The starting composition of the calcium nitrate, with aluminum nitrate and zink nitrate and urea was depend on the total reducing and oxidising valencies of the oxidizer with the fuel with the concept of propellent chemistry. Constituent of the prepared compound were mixed according to stochiometric ratio in mortar and finally pasty solution was formed, after that solution is shifted in muffle furnace . The flame formed with the foamy powder was created, after that fine powder was collected and further analyzed by XRD and photoluminescence measurement. The phase for purities of the prepared samples was checked by powder X-ray diffraction (XRD); with the help of PAN-analytical diffractometer with Cu Kα radiation (1.5405 Å) at an operating voltage at 30mA and 40 kV, and scaning step time at 10.3377's. Photoluminescence (PL) emission for partucular excitation were measured at our workplace using a Shimadzu RF5301PC spectroflurophotometer recorded at room temperature.

III. Results and discussion

X-ray diffraction pattern

Fig 1 shows the XRD patterns of as prepared $Ca_{14}Al_{10}Zn_6O_{35}$ 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 for study of the XRD pattern of prepared powder compound was recorded using Cu-Kα of radiation (1.54060 nm) with a scanning scan step time 10.3377s with continuous scan type. We prepared this $Ca_{14}Al_{10}Zn_6O_{35}$ materials Eu^{3+} ion on the reference of standard JCPDS data source file phosphor and found it is in good agreement with JCPDS data source file no. 050-0426. The XRD pattern of the $Ca_{14}Al_{10}Zn_6O_{35}$ compound is as fallows.

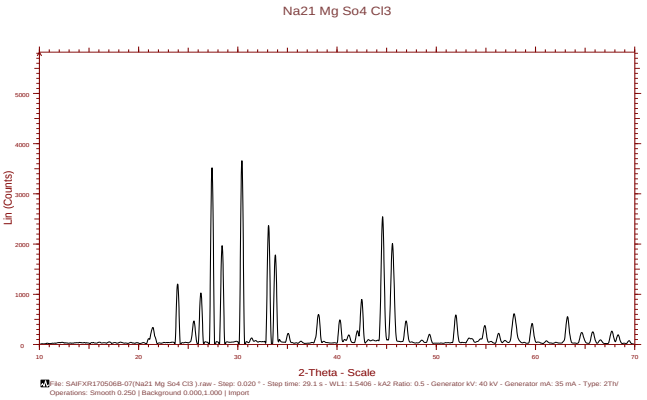

Fig. 1 XRD-pattern of $Ca_{14}Al_{10}Zn_6O_{35}$ lamp phosphors.

SEM Study

The SEM images are represented in fig 2 for combustion synthesized $Ca_{14}Al_{10}Zn_6O_{35}$ phosphor under few micron to sub few micron. It indicate that phosphor prepared with the help of modified combustion synthesized method have sharp shape surface morphology and have grains of crystalline nature. 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.

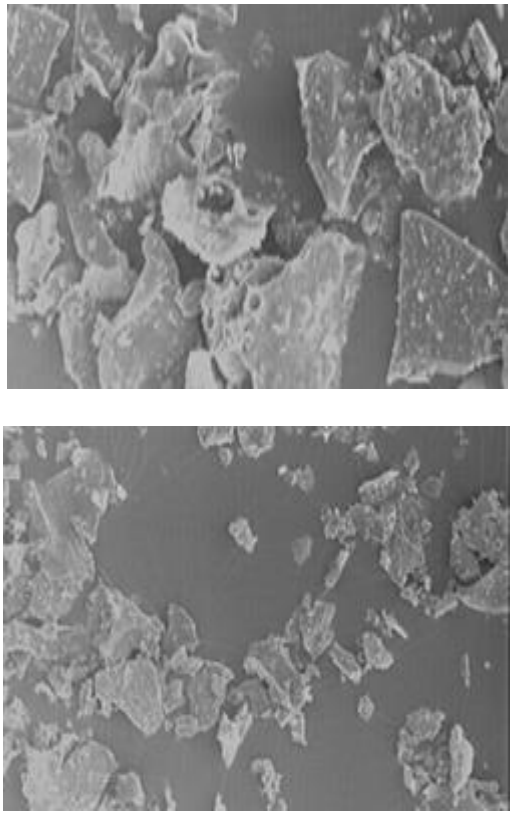

Fig. 2 Morphology of the combustion synthesized $Ca_{14}Al_{10}Zn_6O_{35}$ phosphors

Luminescent property of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ Phosphor

Excitation spectrum of $Ca_{14}Al_{10}Zn_6O_{35}$: 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.

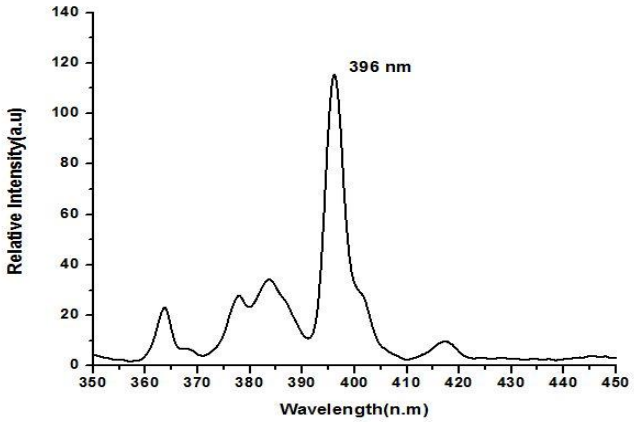

Fig.03 Excitation spectra of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ monitoring the emission at 620 nm

The Eu^{3+} activated $Ca_{14}Al_{10}Zn_6O_{35}$ 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 ($^5D_0 \rightarrow {}^7F_1$) and 620 ($^5D_0 \rightarrow {}^7F_2$) increases. From the emission spectrum of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ phosphor , suitable for red emission for the purpose of solid state lighting.

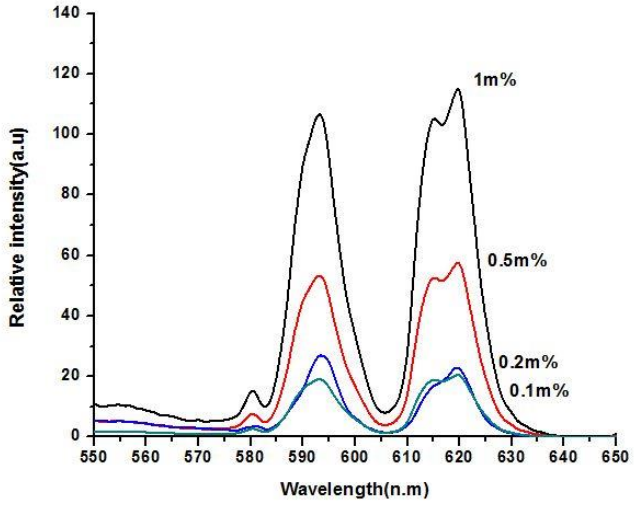

Fig 04 Emission spectra of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ under excitation at 396 nm

Chromatic properties of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ 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. [13-6] 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 [14-15-16-17]. 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 $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ 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+}.

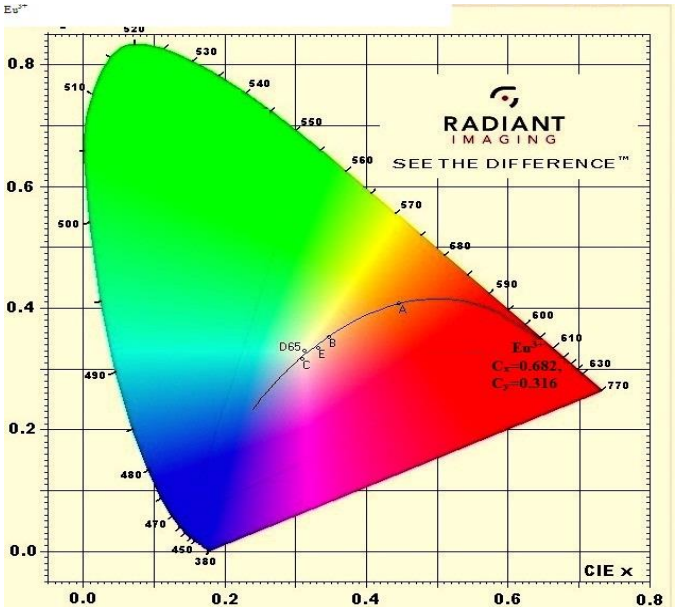

Fig 05. CIE chromatic diagram for $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ phosphor.

The CIE diagram of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ red emitting phosphor shown in 0.682 C_y =0.316) . With the help of CIE diagram it is easy to explains that the $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ phosphors are very near to the CIE graph frame, which easy to shows the utmost color clarity of prepared phosphor material.

IV. CONCLUSION

The investigation on the photoluminescence characteristics of Eu^{3+} activated red-emitting $Ca_{14}Al_{10}Zn_6O_{35}$ phosphors in the near UV–vis range shows the excitation bands at 396 nm , respectively. Emission characteristics of $Ca_{14}Al_{10}Zn_6O_{35}:Eu^{3+}$ shows broad emission band at 593 nm & 620 nm.XRD-pattern of prepared $Ca_{14}Al_{10}Zn_6O_{35}$ 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.

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