



Photoluminescence studies of Rare earth doped $\text{Y}_3\text{Al}_5\text{O}_{12}$ Phosphor

S.Kondala Rao¹, P.B.L Prasanna¹, P.Indira², and K. V. R. Murthy³

¹ Department of Physics, QIS College of Engineering and Technology, Ongole, AP, India

² Department of Physics, K.R.K Govt. Degree College, Addanki, AP, India

³ Display Materials Laboratory, Applied Physics Department, Faculty of Technology & Engineering, M.S University of Baroda, Baroda, India

Corresponding author: sayana.1980@gmail.com, drmurthykvr@yahoo.com

Abstract - The present paper reports the variation of photoluminescence behavior of $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG) with respect to Eu concentration from 0.5 to 3.0 (mol %). The phosphor was synthesized using the standard solid state reaction technique (SSR). The prepared phosphor materials were characterized using powder X-ray diffractometry [XRD], scanning electron microscopy [SEM], and Photoluminescence [PL] (Emission and Excitation). We have studied the effect of dopants on the photoluminescence YAG phosphor monitoring at 400nm. From the PL emission spectrum as increasing the Eu concentration the luminescence peaks at 592, 610 and 631nm are gradually increased which is the fundamental emission of Eu. It is found the YAG: Eu⁺³ phosphors can act as phosphor in compact fluorescent lamps and these nano phosphors may be good candidates for display devices.

Keywords:-Solid State Reaction, Photoluminescence, Rare Earth ions and X-ray diffraction.

1. INTRODUCTION

Recently various phosphor materials have been actively investigated to improve their luminescent properties and to meet the development of different display and luminescence devices (Shinoya and Yen, 1999). Inorganic compounds doped with rare earth ions form an important class of phosphors as they possess a few interesting characteristics such as excellent chemical stability, high luminescence efficiency and flexible emission colors. YAG garnet phases have been widely used as a host material for lasers and phosphors for their excellent luminescent properties and stable physical and chemical properties. Recently, YAG doped with a small amount of element such as Eu, Ce and Tb has been evolved due to a widely utilized in many fields, such as optical display panels, cathode ray tubes, optoelectronic, sensitive devices, nanoscale electronic and plasma display panels due to their special chemical and physical properties. Phosphors are widely used in displays and lighting devices. Photonic application of such phosphors in the area of CRTs and LED(Y. hinatsu, M. Wakshima, 1999).

2. MATERIALS AND METHODS

2.1 Chemicals and apparatus

All the chemical reagents were analytically pure and used without further purification. For synthesis of YAG phosphor pure and doped with varying concentrations of Eu (0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 mol %) rare-earth ions prepared using solid state

reaction method. Stoichiometric proportions of raw materials namely Yttrium Oxide (Y_2O_3), Aluminum Oxide(Al_2O_3), by adding Europium oxide(Eu_2O_3) were used as starting materials, and grinded in motor and mixed and compressed into a crucible and heated at 1200°C for 4 hours in a muffle furnace at the rate of 300°C per hour. Synthesized materials were characterized by X-ray diffractometer (XRD) (Synchrotron Beam Indus -II) with Cu K α radiation ($\lambda = 0.15406$ nm), Scanning Electron Microscope (SEM) (JEOL, JSM-6380), Spectrofluorophotometer (SHIMADZU, RF-5301 PC) using 150 Watt Xenon lamp as excitation source.

2.2 Synthesis and Physical Characterization

The stoichiometric proportions are weighed and ground into a fine power using agate mortar and pestle for 1 hour, the ground samples were placed in an alumina crucible and heated at 1200°C for 4 hours in muffle furnace with a heating rate of 5°C /min. The samples are allowed to cool to room temperature in the same furnace for about 20 hours. The prepared samples were again ground in to powder for taking the characteristic measurements. All the synthesized phosphor samples were characterized by X-ray diffraction (Synchrotron Beam Indus -II) to identify the crystallinity and phase purity of the phosphor. The Photoluminescence (PL) emission and excitation spectra were measured by Spectrofluorophotometer (SHIMADZU, RF-5301 PC) using 150 Watt Xenon lamp as the excitation source at room temperature.

The emission and excitation slit were kept at 1.5 nm, and recorded at room temperature. The particle morphology of the prepared solid nano powders was characterized by SEM.

3. RESULTS AND DISCUSSION

3.1 Phase characterization

The XRD patterns of YAG: xEu (x=2.5) phosphors are shown in Fig. 1. It can be observed that all the diffraction peaks matched well with the Joint Committee on Powder Diffraction Standards (JCPDS) No.33-0040 (pure YAG). All the reflection peaks can be voluntarily indexed to those of the pure cubic phase with primitive structure of YAG (PDF 33-0040) with space group Ia 3d, No other impurity phases were detected at these doping levels (Kondala Rao. Sayan, Murthy KVR. et.al, 2015). The results showed the expected chemical components in the phosphors. It is obvious that Eu³⁺ ions successfully substituted Y³⁺ ions and the small amount of Eu³⁺ ions in YAG host lattice did not change its crystalline structure.

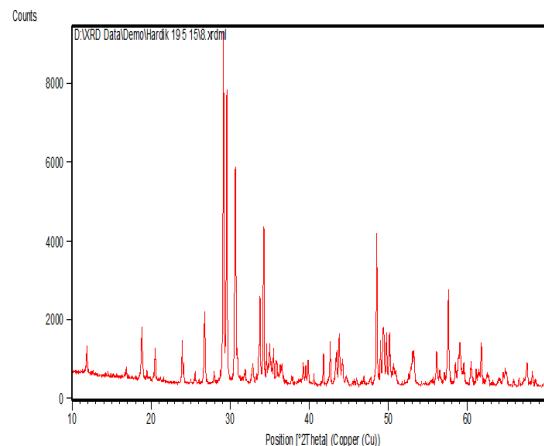


Figure 1. XRD pattern of Eu³⁺ doped YAG phosphor

3.2 The characterization of luminescent properties

Fig 2 is the PL emission and excitation when the pure phosphor is monitored at 400nm, the excitation found at 254nm which is curve-1. In fig 2 curve-2 when the phosphor is excited with 254nm the emissions are found at 365, 400, 469 with intensity 107, 84 and 67 units. The emission at 365 is at strongest which is attributed to crystal field of the material. The emissions 400nm and 469nm are due to release of electron from O²⁻ ions on excitation. This is due to low crystalline density of the phosphor material (R.Sankar, G.V.subba Rao, 2000).

Fig 3 is the PL emission and excitation spectra of Y₃Al₅O₁₂ phosphor with various Eu [0.5, 1, 1.5, 2, 2.5 and 3%] concentrations. All the phosphors are

excited with 254nm and the emission is recorded from 350 to 650nm.

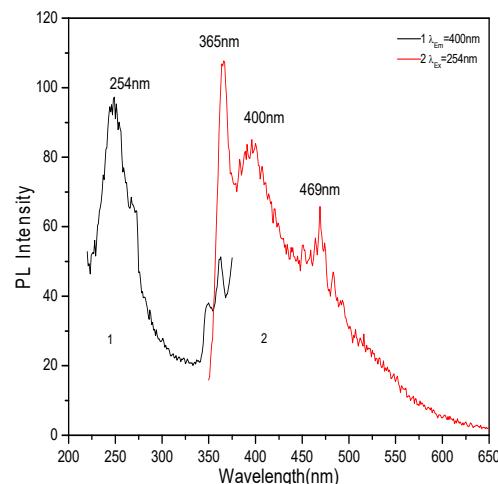


Figure 2. Excitation and Emission spectrum pure YAG phosphor

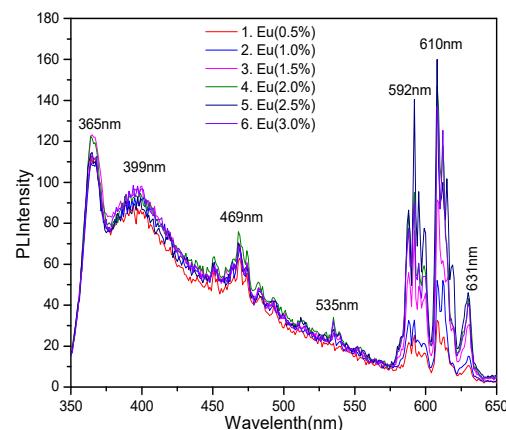


Figure 3. Excitation and Emission spectrum Eu³⁺ doped YAG phosphor

S.N o	Sample	Dopant Concentra tion Mol(%)	Emission peak intensities under 254nm Ex.			
			535	592	610	631
1	Y ₃ Al ₅ O ₁₂	Eu(0.5%)	57	25	32	10
2		Eu(1.0%)	61	32	52	15
3		Eu(1.5%)	59	76	91	30
4		Eu(2.0%)	63	94	145	43
5		Eu(2.5%)	60	140	160	46
6		Eu(3.0%)	59	89	137	41

Table 1. show the effect of Eu on the intensities of 535, 592, 610 and 631nm peaks from the fig it is found as the Eu concentration increases all the Eu³⁺ emission intensities increases linearly. After 2.5%

Eu in the host $\text{Y}_3\text{Al}_5\text{O}_{12}$ phosphor the intensity decreases.

The samples demonstrate strong excitation peaks at 254 nm, which conforms to the value of the charge transfer band in the YAG:Eu³⁺ system. Therefore, stronger charge transfer transitions providing the energy to the excited 5D levels can lead to luminescence enhancement in well dispersed nanocrystals (Feldmann C, Jüstel T, Ronda C R, 2003). This additional transition (CTS) besides the charge transfer band (CTB) will provide energy to the 5D_J states of Eu³⁺, and the absorbed energy in 5D_J states will transfer to 7F_J states. Hence, the charge transfer to Eu³⁺ will become more probable, leading to the enhancement of luminescence emission in the YAG:Eu³⁺ nanoparticles. The 365,399,469nm peaks observed in undoped YAG and also Eu doped phosphors the position and intensities are nearly same. The doping of the Eu generates a hump in green region that is at 535nm whose intensity is nearly same for all the Eu concentration in YAG phosphor. The other emissions from Eu³⁺ in YAG phosphors are 592,610,631 along with other satellite peaks of Eu. As the Eu concentrations increases in YAG phosphors the PL intensity of the 592,611,631 nm peaks increases its intensity up to Eu (2.5%) in YAG phosphor. After 2.5% of Eu in YAG phosphor the intensity marginally decreases.

592	$^5\text{D}_0 \rightarrow ^7\text{F}_1$	2.103eV
610	$^5\text{D}_0 \rightarrow ^7\text{F}_2$	2.031eV
631	$^5\text{D}_0 \rightarrow ^7\text{F}_3$	1.961eV

This is due to the standard quenching effect. Which is attributed to the columbic repulsion of nearby Eu ions leads to non release of electrons from 4f shells. The emissions 592,610,631 are standard Eu³⁺ ions, which is attributed to the following transitions of RE³⁺. The emission peaks are produced by Eu³⁺. 592nm ($^5\text{D}_0 \rightarrow ^7\text{F}_1$), 610nm ($^5\text{D}_0 \rightarrow ^7\text{F}_2$) and 631($^5\text{D}_0 \rightarrow ^7\text{F}_3$) (S. Kondala Rao, P.Indira, et al, 2015), it is interesting to note here the emissions below 600 are considered magnetic dipole component and above 600 components are attributed to hyper sensitive electric component of the phosphor material.

3.3 SEM analysis

The typical SEM micrographs of the phosphor powders are depicted in Figure. 4. The morphology of resulting sample which reveals the formation of polycrystalline material with grain size shape distribution is irregular and average grain size is in sub-micrometer range, due to agglomeration of smaller grains which forms due to thorough crushing and high temperature processing of the samples

(Danielson E, et al, 1998). This proves the solid state synthesis method is approving for synthesis macro structured samples of reported phosphor. The particle sizes are in the size range of 2–5 μm . From SEM images,(Ropp, R.C, 1991) it has been noted that the agglomeration of synthesized powder phosphors has been increased with the change in Eu content, owing to the density variation.

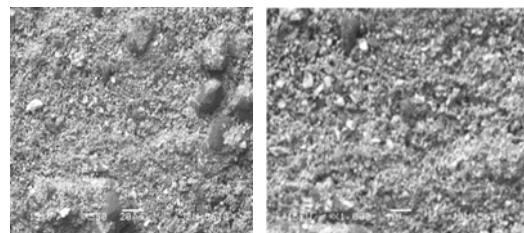


Figure.4a, 4b SEM of Eu³⁺ doped YAG phosphor

3.4 EDS Study

Energy-dispersive X-ray spectroscopy (EDS), sometimes called energy dispersive X-ray is an analytical technique used for the elemental analysis or chemical characterization of a sample. It relies on an interaction of some source of X-ray excitation and a sample. Its characterization capabilities are due in large part to the fundamental principle that each element has a unique atomic structure allowing a unique set of peaks on its electromagnetic emission spectrum

From the spectra of EDS mostly elements of base materials are seen in the EDS spectrums. There fore in general it is normally concluded that the phosphors under EDS study are pure.

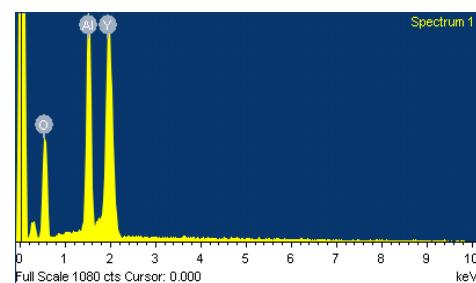


Fig.5. EDS of Eu (2.5%) doped $\text{Y}_3\text{Al}_5\text{O}_{12}$ Phosphor

CONCLUSIONS

The YAG base and Eu³⁺ doped YAG phosphors were successfully synthesized by conventional solid state reaction method and its photoluminescence studies were investigated in detail. The YAG based orange red emitting phosphor, tested by X-ray diffraction pattern and SEM micrographs show the formation of microcrystalline sample. The photoluminescence studies of synthesized Eu³⁺ doped YAG phosphor material shows intense orange



red emissions at 592nm and 610nm & 631nm corresponding to characteristic transitions corresponding to MD ($^5D_0 \rightarrow ^7F_1$) and ED ($^5D_0 \rightarrow ^7F_2$ & $^5D_0 \rightarrow ^7F_3$) of Eu³⁺ ion under the excitation wavelength of UV 254 nm respectively. These phosphors with fine shape emissions may find potential applications in the fields of minute color displays in the near future (K.V.R. Murthy et al, 2007). The luminescence enrichment observed in the nano phosphors is of practical importance for this system to be applied to field emission devices. The results indicated that present phosphor could find application in white light emitting CFLs which can be used in back light producing lamps for liquid crystal display (LCDs) and other optoelectronic devices.

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