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Synthesis and Photoluminescence Studies of Eu Doped $Gd_3Al_5O_{12}$ Phosphor

P.B. Lakshmi Prasanna[1], S. KondalaRao[2], K. Suresh[3], B. SubbaRao[1] and **K.V.R. Murthy[4*]**

[1]*Department of Physics, VSR & NVR College, Tenali, A.P., India*
[2]*Department of Physics, QIS College of Engineering, Ongole, A.P., India*
[3]*Department of Physics, The Bapatla College of Arts & Sciences, Bapatla, A.P., India*
[4]*Department of Applied Physics, Faculty of Engineering and Technology, M.S. University of Baroda, Vadodara, India,*

Abstract— *The meta stable garnet lattice of $Gd_3Al_5O_{12}$ have been synthesized by solid state reaction method which then allows an effective incorporation of rare earth ions Eu^{3+} activators for opto-functionality explorations. The characterizations of the products were achieved by combined means of XRD, SEM, and PL/PLE. The LnAG exhibit strong 592 nm and 616nm emissions (the $^5D_0 \rightarrow {}^7F_1$ magnetic dipole transition and $^5D_0 \rightarrow {}^7F_2$ electric dipole transition of Eu^{3+}) upon UV excitation at 275 nm attributed to $^8S_{7/2} \rightarrow {}^6I_J$ with CIE chromaticity coordinates of x=0.62 and y=0.38 (orange red), and the quenching concentration of Eu^{3+} was found. Owing to the improved crystallization of LnAG at a higher temperature, luminescence emission intensity increases significantly, especially above 1000°C. The Eu^{3+} doped GdAG phosphors are expected to be a new type of photoluminescent and scintillation material.*

Keywords— *gadolinium aluminate garnet, Eu^{3+} doping, photoluminescence, lattice structure*

1. INTRODUCTION

Rare-earth aluminate garnets ($Ln_3Al_5O_{12}$, LnAG), especially YAG, are well-known inorganic compounds which have been widely studied for optical and high temperature mechanical applications [1]. When properly activated with luminescent centers, the LnAG compounds are important inorganic phosphors (such as YAG:Ce) finding wide applications in cathode-ray tubes (CRTs), field emission displays (FEDs), scintillation, vacuum fluorescent displays (VFDs), electroluminescence (EL), and so forth [2], because of their high chemical and radiation stabilities, wide band gap and excellent radiation conversion efficiency. The $Gd_3Al_5O_{12}$ (GdAG) based phosphor, though it has higher density than YAG and the Gd^{3+} in this system can Sensitize the $^5D_0 \rightarrow {}^7F_{1,2}$ red emissions of Eu^{3+} through an efficient energy transfer [3], has been rarely reported.

Phase evolution of the as-synthesized precursors upon calcination and luminescence behaviors of the oxide phosphors were studied in detail via the combined techniques of XRD, SEM, and PL/PLE luminescence spectroscopy. In the following sections, we report the synthesis, characterization, and luminescent performance of the GdAG: Eu garnet phosphors.

2. EXPERIMENTAL

To prepare $Gd_3Al_5O_{12}$ with various concentrations of europium (1, 1.5, 2, and 2.5 mol%), stoichiometric amounts of reactant mixture is taken in alumina crucible and is fired in air at 1200°C for 3 hour in a muffle furnace. The Eu^{3+} activated $Gd_3Al_5O_{12}$ phosphors was prepared via high temperature modified solid state diffusion. The starting materials Gd_2O_3, Al_2O_3, were used to prepare the host phosphor and Eu_2O_3 as dopant. The mixture of reagents was grounded together for 45 minute to obtain a homogeneous powder. Powder was transferred to alumina crucible, and then heated in a muffle furnace at 1200 °C for 3 hr. The phosphor materials were cooled to room temperature naturally.

The samples were characterized by using Photoluminescence (PL), XRD and SEM. The photoluminescence (PL) emission and excitation spectra were recorded at room temperature by use of a Shimadzu RF-5301 PC spectrofluorophotometer

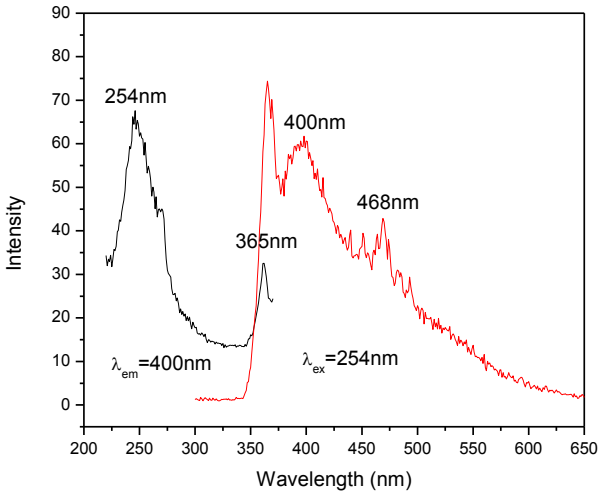

Fig. 1: PLE and PL spectra of $Gd_3Al_5O_{12}$ phosphor

* *Corresponding Author Email: drmurthykvr@yahoo.com*

3. RESULTS AND DISCUSSION

3.1 Photoluminescence Study of Base Phosphor

Fig.1 depicts the PLE and PL spectra of sample $Gd_3Al_5O_{12}$ calcined at 1200°C for 3 h. A general observation is that the PLE spectrum is composed of a strong CTB band located at 254 nm Upon UV excitation at 254nm, the phosphors exhibit the typical emission in the 350-500 region, with peaks at 400, 468nm.

3.2 $Eu3^+$ Doping for Red Luminescence

The PLE spectra of $Gd_3Al_5O_{12}:Eu^{3+}$ by monitoring $^5D_0 \rightarrow {}^7F_J$ emission of Eu^{3+} at 616nm was measured and shown in Fig.2. The spectra are scaled on the $^8S_{7/2} \rightarrow {}^6I_J$ excitation (275nm) intensity. A series of much weaker intra-4*f*6 electronic transitions of Eu^{3+} in the longer wavelength region as marked in the figure. It should be noted that the typical $^8S_{7/2} \rightarrow {}^6I_J$ intra *f-f* transition of Gd^{3+} is clearly observed at 275 nm with high intensity, and this result is in good agreement with the result reported by Ji-Guang Li et al., from providing direct evidence of an energy transfer from Gd^{3+} to the Eu^{3+} activators.

Fig.3 shows the PL spectra of red-emitting $Gd_3Al_5O_{12}:Eu^{3+}$ upon excitation in the ^6I_J level of Gd^{3+} at 275nm. The emission measured from 400 – 650nm range, observed several peaks 419, 430, 467, 490, 543, 557, 592, 595, 599, 616 and 628nm. All the peaks are attributed to Eu3+ ion transitions from $^5D_J \rightarrow {}^7F_J$. Among these emissions high intense MD and ED transitions are observed. As the Eu concentration increases the MD and ED transitions of Eu^{3+} ion emission increases. Fig.3a shows the emission of MD and ED under 275nm excitation wavelength.

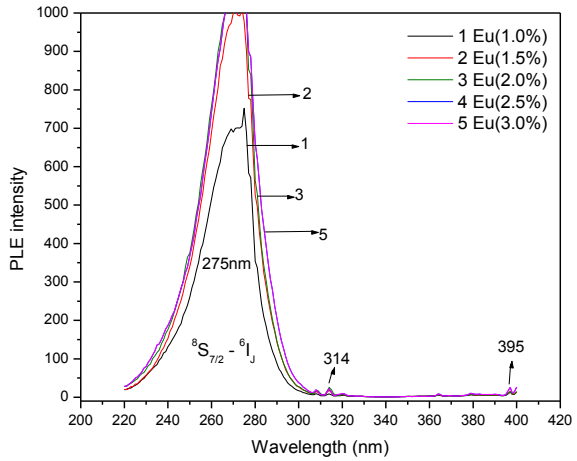

Fig. 2: PLE spectra of Eu^{3+} doped $Gd_3Al_5O_{12}$ phosphor monitored under 616nm wavelength

Fig.4 shows the emission under different excitation wavelengths. Fig.5 reveals the energy level diagram for the Gd^{3+}- Eu^{3+} system, showing the possible visible quantum cutting by a two-step energy transfer from Gd^{3+} to Eu^{3+} where 1 and 2 denote cross relaxation and direct energy transfer. Table 1 shows the emission peak wavelengths and their intensities under different excitation wavelengths.

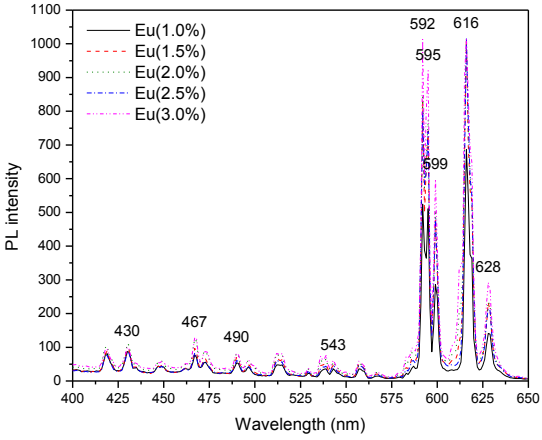

Fig. 3: PL spectra of Eu^{3+} doped $Gd_3Al_5O_{12}$ phosphor under 275nm excitation

Table 1: Emission peak wavelengths and their intensities under different excitation

S. No	*Sample*	*Emission peak Wavelengths (nm)*	*Emission peak Intensities under*		
			λ_{Ex}=*254nm*	λ_{Ex}=*269nm*	λ_{Ex}=*275nm*
1	$Gd_3Al_5O_{12}$:Eu (3.0%)	419	109	101	94
2		430	95	103	103
3		467	151	143	129
4		557	47	64	60
5		592	592	>1000	>1000
6		595	592	950	960
7		599	350	600	615
8		616	843	>1000	>1000
9		628	189	296	292

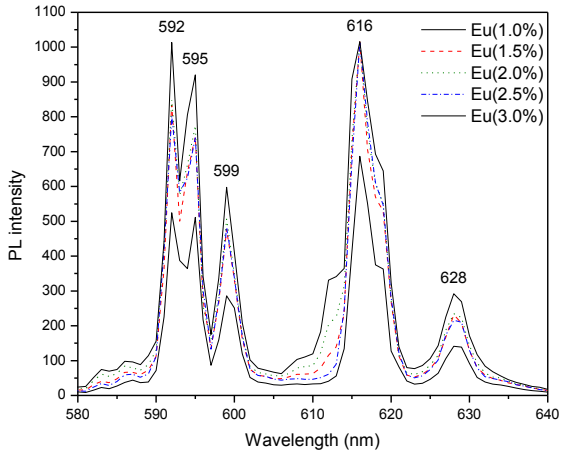

Fig. 3(a): PL spectra of Eu^{3+} doped $Gd_3Al_5O_{12}$ phosphor under 275nm excitation range from 580-640nm

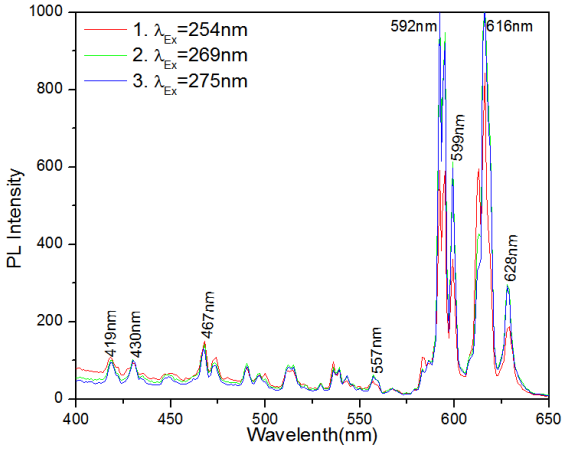

Fig. 4: PL spectra of Eu^{3+} (3.0%) doped $Gd_3Al_5O_{12}$ phosphor under different excitations

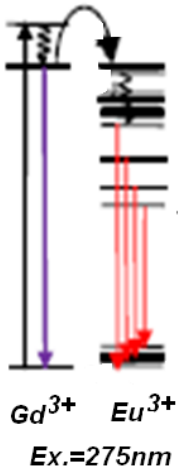

Fig. 5: Charge transfer mechanism

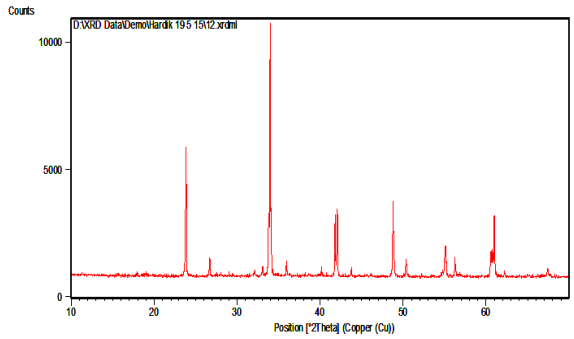

Fig. 6: XRD pattern of Eu^{3+} (3.0%) doped $Gd_3Al_5O_{12}$ phosphor

Fig.7 is the SEM micrograph of the phosphor understudy. It is found from the micrograph agglomerated particles with average size is around 3-5 microns.

Table.2 is the emission wavelengths and allowed transitions of the Gd and Eu in 3+ states. Fig.5 is the charge transfer mechanism and the possible transfer of energy from Gd to Eu in 3+ states.

Table 2: The following are the allowed transitions of RE^{3+}

Emission peak Wavelength (nm)	*Transition*
430	$^5D_3 \rightarrow {}^7F_0$
467	$^5D_2 \rightarrow {}^7F_0$
557	$^5D_1 \rightarrow {}^7F_0$
592	$^5D_0 \rightarrow {}^7F_1$
595	$^5D_0 \rightarrow {}^7F_1$
599	$^5D_0 \rightarrow {}^7F_1$
616	$^5D_0 \rightarrow {}^7F_2$
628	$^5D_0 \rightarrow {}^7F_2$

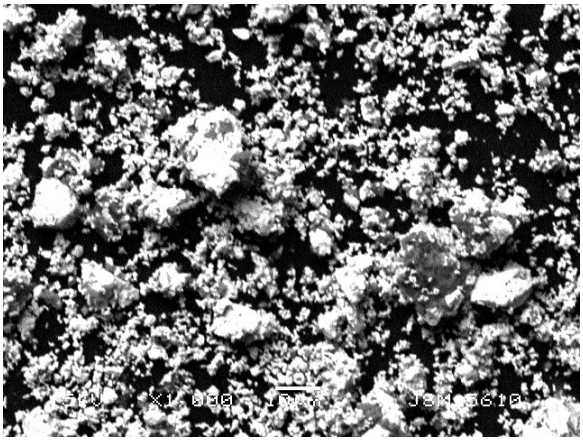

Fig. 7: SEM image of Eu^{3+} (3.0%) doped $Gd_3Al_5O_{12}$ phosphor

4. CONCLUSIONS

The Eu^{3+} (3.0%) doped $Gd_3Al_5O_{12}$ phosphors were synthesized successfully via a solid-state reaction. Photoluminescence exhibit orange red (594nm) and red (616, 627nm) dominant luminescence along with blue and green emissions. From XRD studies the compound is mostly in single phase. The results in this work demonstrate that this phosphor is expected to be promising candidates for application in display devices.

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