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Synthesis, characterization and photoluminescent studies of Eu^{3+} doped LAG phosphor

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Abstract

The present paper reports on the synthesis, characterization and photoluminescent studies of Eu^{3+} doped LAG ($La_2Al_2Gd_2O_9$) phosphor. The dopant concentration in the phosphor is 0.1, 0.2, 0.5, 1.0, 1.5 and 2.0 mol%. The phosphors were synthesized using the standard solid state reaction technique for the first time. To enlarge the fundamental understanding of the phosphors was done by PL, XRD, SEM, FTIR and CIE are determined. From the PL excitation and emission studies, the prepared phosphors are having two excitations 254 and 275nm wavelengths. The emission under these two wavelengths shows interesting results.

Keywords: *Photoluminescence, wavelength, phosphor, solid state reaction*

1. INTRODUCTION

The first reported white light LEDs were based on blue InGaN technology, which uses a combination of blue emission from a blue LED and yellow from YAG:Ce^{3+} phosphor coated. Then uses a combination of blue emission from a blue LED and yellow from YAG:Ce^{3+} phosphor and red from Y_2O_3:Eu^{3+}emissions. However, these single and two-band white LEDs suffer from limited color rendering and color temperature, and are unable to produce all nature-equivalent colors, especially in the red region. To improve the color temperatures and the rendering index of phosphor-converted white LEDs, various phosphors with different components have been developed.

In view of YAG and TAG phosphors we thought to prepare this new LAG phosphor for the first time and studied synthesis, characterization and luminescent properties.

2. EXPERIMENTAL

The inorganic compounds like La_2O_3, Al_2O_3 and Gd_2O_3 of purity (99.9%) were used as starting materials for the host phosphor. Europium oxide Eu_2O_3 is used as activator ion at different concentration (0.1, 0.2, 0.5, 1.0, 1.5 and 2.0 mol%). All the compounds was weighed, added in appropriate proportions and grounded into a fine powder using agate mortar and pestle about an hour. The grounded phosphors were placed in an alumina crucible and heated in air atmosphere at 1200°C for 2 hours in a muffle furnace with a heating rate of 5°C/min. After cooling the phosphors was again ground in to powder and done the following characterizations.

The phosphors were characterized by the Photoluminescence emission and excitation spectra using (SHIMADZU, model RF-5301 PC) Spectrofluorophotometer using xenon lamp as excitation source. X-ray diffraction (XRD) studied using Rigaku-D/max 2500 using Cu Kα radiation. The microstructures of the samples were studied using a scanning electron microscopy (SEM) (XL 30 CP Philips). The FTIR studied using an FTIR spectrometer (Perkin Elmer-Spectrum 100) in the range from 500 to 4000cm^{-1}.

3. RESULTS AND DISCUSSIONS

3.1 Photoluminescence excitation study

Fig.1 is the excitation spectrum of Eu^{3+} doped LAG phosphors monitored under 612nm wavelength. The excitation spectrum was characterized by three main bands. The band was ranging from 220-300nm with peak at 254, 275nm. Among two peaks the 275nm sharp peak is higher than 254nm peak. The third small band peaked at 549nm.

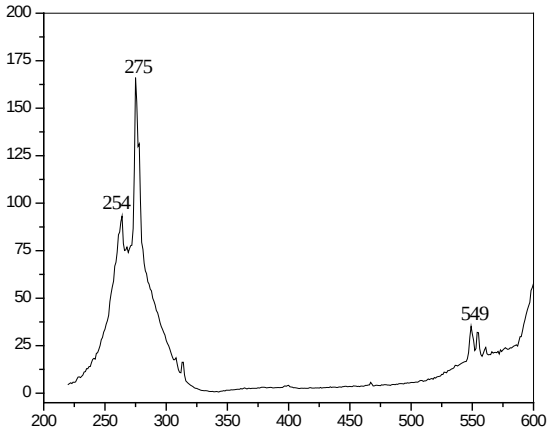

Fig.1 Excitation spectrum of LAG:Eu^{3+} phosphor monitered under 612nm wavelength

3.2 Photoluminescence emission study

Basing on the excitation study it is decided to measure the emission spectrum under 254 and 275nm excitation wavelengths. The emission under 549nm is ignored and not presented due to less intensity.

Fig.2 shows the combined emission spectrum of Eu^{3+} (0.1, 0.2, 0.5, 1.0, 1.5 and 2 mol%) LAG phosphors under 254nm excitation. The emission spectrum consists of several sharp peaks in the range from 350 – 650nm. The peaks are at 365, 400, 420, 429, 448, 468, 495, 514, 540, 557, 592, 617 and 627nm. These emissions were assigned may be to the Gd^{3+} - O^{2-}. The Gd rare earth ion is having seven unpaired electrons in the outer most orbit.

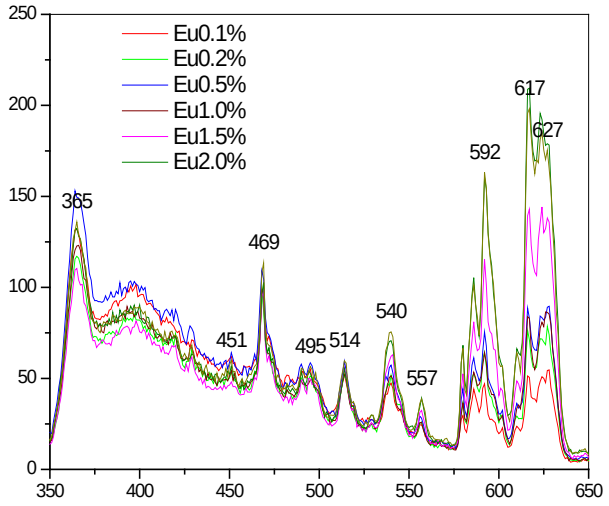

Fig.2 Emission spectrum of Eu doped LAG phosphors under 254nm excitations

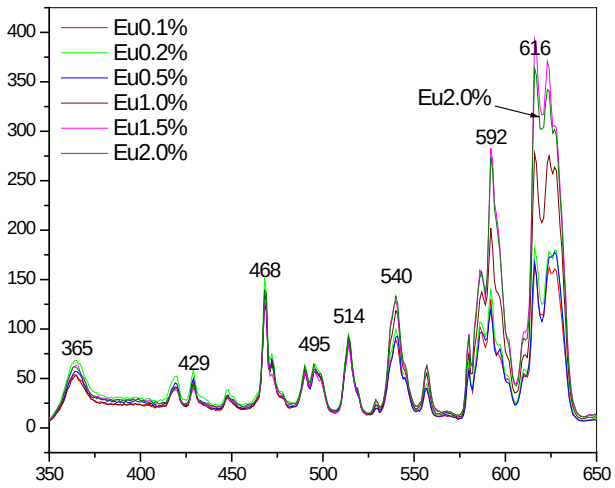

Fig.3 Emission spectrum of Eu doped LAG phosphors under 275nm excitation

Fig.3 shows the combined emission spectrum of Eu^{3+} (0.1, 0.2, 0.5, 1.0, 1.5 and 2 mol%) LAG phosphors under 275nm excitation wavelength. The emission spectrum consists of several sharp peaks in the range from 350 – 650nm same as under 254nm excitation. Fig. 4 shows the combined spectrum of Eu doped LAG phosphors under 275nm excitation in the emission intensity range from 0 – 125 units.

It is observed that under 254nm excitation due to the high crystal field at 365nm the emission peak intensities are decreased as the wavelength increases from 350 – 650nm. The values of emission intensities of the peaks are noted in the table 1.

Whereas under 275nm excitation, due to the less crystal field at 365nm the emission peak intensities are increased from blue to red region only. It clearly indicates that the crystal field playing an important role, one can get different emission colours from the phosphor by changing the excitation wavelength. It is also observed that the red emission increases as the Eu ion concentration increases from 0.1 to 2 mol%.

Table 1

Emission peak wavelength	Emission intensity under	
	254nm	275nm
365	150	68
395	104	32
451	65	34
469	114	154
495	58	68
514	59	91
540	76	134
557	40	63
592	164	283
617	211	392
627	195	369

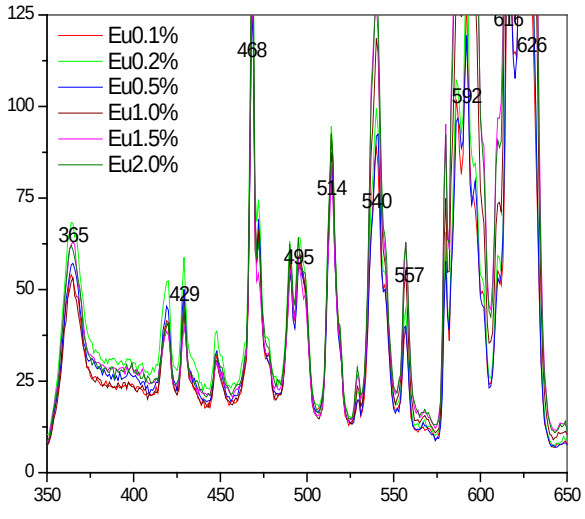

Fig.4 Emission spectrum of Eu doped LAG phosphors under 275nm excitation

3.3 X-Ray Diffraction Analysis

The XRD pattern of LAG phosphor without flux is shown in fig.5. From the XRD pattern the narrow

peaks indicates bigger the crystallite size and thereby bigger the particle size. It is clearly observed that the maximum peak obtained at 28.5°. The calculated crystallite size using Scherer's formula $D = K.\lambda / \beta \cos\theta$ is 41nm, where k the constant (0.9), λ the wavelength of the X-ray (1.5418 Å), β the full-width at half maxima (FWHM), θ the Bragg angle. From XRD pattern it is found the phosphor may not be in single phase. Many crystallites agglomerate together and form a particle. In the present case it is interesting to note the calculated crystallite size is around 41nm, which conclude us the solid state reaction route is good to prepare nano crystallites also.

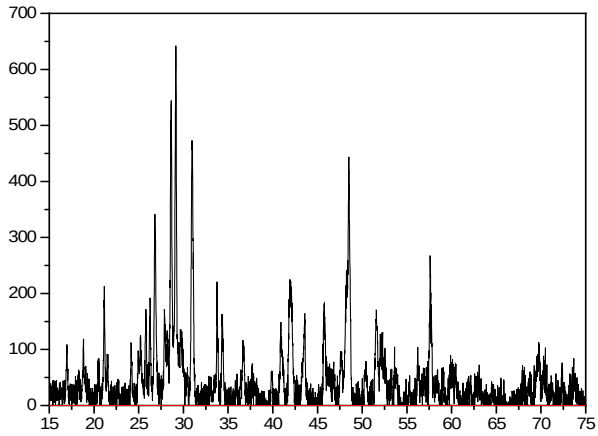

Fig.5 XRD pattern of Eu (2%) doped LAG phosphor

3.4 SEM Analysis

Fig. 6a & 6b shows the SEM micrographs of the Eu doped LAG phosphor under different resolutions. From the Scanning Electron Micrographs with the measuring scale is 10 & 5 microns. The microstructure appears to be mostly irregular shape with smooth surface having an average basal diameter less than one micron. From SEM micrograph it is observed good particles having mostly spherical shape of varying sizes mostly agglomarated together.

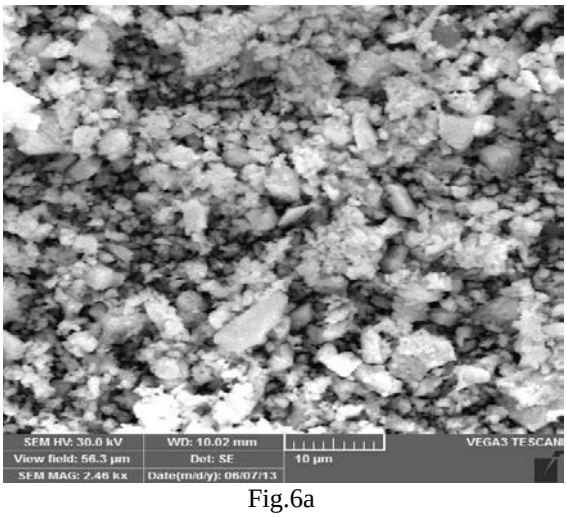

Fig.6a

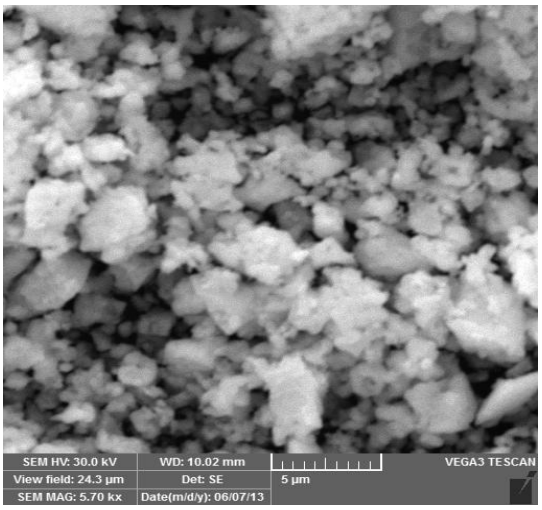

Fig.6a & 6b SEM micrographs of Eu (2%) doped LAG phosphor under different resolutions

3.5 FTIR Analysis

Fig.7 shows the FTIR spectrum of Eu doped LAG phosphor. From the figure the observed peaks are at 3625, 2375, 1425, 850, 650, 500 cm^{-1}. From FTIR it is observed that most of the bonds are due to C-O, Sr-O, La-O and Al-O stretching and the O-H stretching band is observed at 3625cm^{-1}. The band at 3625cm^{-1} is due to the H-OH stretching of absorbed water molecule from the atmosphere.

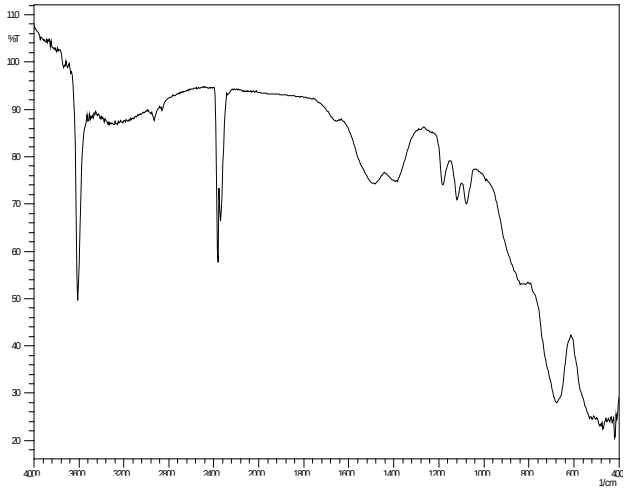

Fig.7 FTIR pattern of Eu (2%) doped LAG phosphor

3.5 CIE Analysis

Fig.8 shows the CIE colour co-ordinates of the Eu (2%) doped LAG phosphor under 254 and 275nm excitation wavelength. The colour co-ordinates of the Eu doped LAG phosphor under 254nm and 275nm are x=0.490, y=0.317 and x=0.571, y=0.348. From the figure the phosphors emitting red colour and are useful in producing white light in the field of lamps and display devices. The calculated correlated colour temperature

(CCT) of 254nm excited phosphor is 1692K whereas the CCT of 275nm excited phosphor undefined.

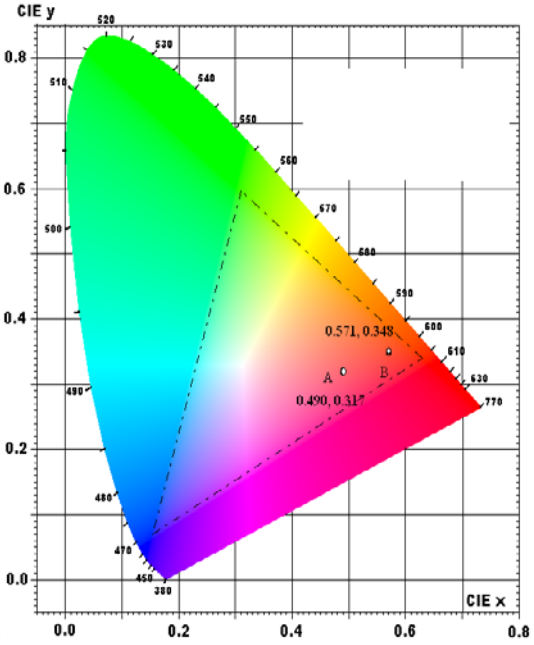

Fig.8 CIE colour co-ordinates of Eu doped LAG A) 254nm excited phosphor B) 254nm excited phosphor

Conclusions:

The Eu^{3+} doped LAG ($La_2Al_2Gd_2O_9$) phosphor is successfully synthesized. The emission spectrum of Eu^{3+} doped LAG phosphors under 254nm excitation consists of several sharp peaks in the range from 350 – 650nm. The peaks are at 365, 400, 420, 429, 448, 468, 495, 514, 540, 557, 592, 617 and 627nm with good intensity. These emissions were assigned to the Gd^{3+} - O^{2-} and Eu^{3+} - O^{2-} bonds stretching's and or emission arises from the un paired electrons in the outer most orbit of six electrons in Eu^{3+} and seven electrons in Gd^{3+} which are behaving in such a way to tune them self gives rise the combined luminescence leading to generation of near white light from single host which is Eu^{3+} doped LAG ($La_2Al_2Gd_2O_9$) phosphor.

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