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Synthesis & Photoluminescence study of LaPO4: Ce, Eu, Tb Phosphor

***K. G. Chaudhari[1], Y.S.Patil[1], B. Subba Rao[2] and K.V.R.Murthy[3]**

[1]Applied Physics Department, MCT'S Rajiv Gandhi Institute of technology, Versova, Andheri (w), Mumbai
[2]Department of Physics, VSR & NVR College, Tenali, Guntur District, A.P
[3]Display Materials Laboratory, Applied Physics Department, Faculty of Technology and Engineering, The M. S. University of Baroda, Vadodara, India

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ABSTRACT

The present paper reports the Photoluminescence (PL) of the $LaPO_4$ phosphor doped with Ce, Eu and Tb rare-earth ions, keeping Ce, Eu concentration constant and varying Tb concentration as 0.1, 0.5 and 1.5%.The phosphors were synthesized using the standard solid state reaction technique and ground using mortar and pestle, fired at $1200^{O}C$ for 4 hour in a muffle furnace. The crystallinity and phase purity of the product were firstly examined by XRD analysis. We have studied the effect of dopants on the Photoluminescent properties of the samples using Spectrofluorophotometer at room temperature. PL emission of undoped $LaPO_4$ phosphor was observed at 470 nm. Under the excitation of 254nm wavelength, PL emission of doped $LaPO_4$ phosphor shows peaks at 381, 415, 437, 457, 473, 488, 545, 589, 596, 614 and 622nm with good intensity. As the Tb concentration increases the PL intensity also increases.

Keywords: Photoluminescence; phosphor rare-earth ions; XRD; SEM; solid state reaction technique.

INTRODUCTION

Phosphors are widely used in displays and lighting devices. The useful applications of rare earth element compounds, especially lanthanide phosphate doped inorganic materials, have been touched upon broadly. Over the past a few years, they have been applied in many fields, such as optical display panels, cathode ray tubes, optoelectronic, sensitive device, nanoscale electronic and plasma display panels[1−4] due to their special chemical and physical properties. Various solution-phase routes, includingsolid state reaction,sol-gel, precipitation,water oil microemulsion,polyol-mediated process, ultrasonification, hydrothermal, and mechanochemical method[5-8], have been tried to lower the reaction temperature and obtain high-quality $LaPO_4$ based nanoparticles. However, the simple and mass fabrication of $LaPO_4$ nanocrystals with narrow grain size distribution and uniform morphology still remains a challenge. It appears that the best solution both to control powder morphology and to produce low cost thin films is the use of soft chemistry routes. We adopted the standard solid state reaction technique to prepare $LaPO_4$ with good morphologies and fine crystal structures; and its emission and intensity of luminescence were also studied. The present paper reports the Photoluminescence (PL) of the $LaPO_4$ phosphor doped with Ce, Eu and Tb rare-earth ions, keeping Ce, Eu concentration constant and varying Tb concentration.

MATERIALS AND METHODS

All the chemical reagents were analytically pure and used without further purification. $LaPO_4$ phosphor doped with Ce, Eu and Tb rare-earth ions, keeping Ce & Eu concentration constant and varying Tb concentration as 0.1, 0.5, and 1.5% were prepared using solid state synthesis method. Stoichiometric proportions of raw materials namely, Lanthanum Oxide (La_2O_3), Diammonium Hydrogen Phosphate [$(NH_4)_2$ H PO_4), Cerium Oxide (Ce_2O_3), Europium Oxide (Eu_2O_3) and Terbium Oxide (Tb_4O_7) were grinded in an agate motor and mixed and compressed into a crucible and heated at 1200^0C for 4 hours. The prepared samples were again powdered for taking the measurements.

The XRD patterns' of the samples were obtained using Diffractometer system=XPERT-PRO at NCL Pune and the excitation & emission spectra were recorded at room temperature using (SHIMADZU,make Spectroflurophotometer RF – 5301 PC) using Xenon lamp as excitation source at display research Lab., Department of Applied Physics, Faculty of Technology and Engg., M.S.University. Baroda.. The emission and excitation slit were kept at 1.5 nm.

RESULTS AND DISCUSSION

X-ray diffraction study (Phase purity and structure):-

The crystallinity and phase purity of the product were firstly examined by XRD analysis. Fig.1 & Fig.2. Shows the typical X-ray diffraction (XRD) patterns of synthesized samples of pure $LaPO_4$ and $LaPO_4$ dopped with Ce,Eu,Tb. As shown XRD patterns of nanocrystals are in good agreement with the values from JCPDS no.35-731of $LaPO_4$, which shows that all the products are monazite $LaPO_4$ with monoclinic structure. All diffraction patterns were obtained using CuKα radiation ($\lambda = 1.540598$ A^0) at 40 kv and 30 mA, and divergence slit fixed at 1.52 mm. Measurements were made from $2\theta = 10^0$ to 80^0 with steps of 0.008356^0.

When crystallites are less than approximately 100 nm in size, appreciable broadening in X-ray diffraction lines occurs. The crystallite size of powder sample were calculated by using Scherer equation

$$D= 0.9 \lambda / \beta\cos\theta$$

Where β represents full width at half maximum (FWHM) of XRD lines
λ = Wavelength of the X-rays.(0.154 nm in the present case)

θ = Braggs angle of the XRD peak.

The average crystallite size of $LaPO_4$ phosphors is 62 nm and when doped with Ce, Eu, Tb dopants, the crystallite size becomes 75 nm.

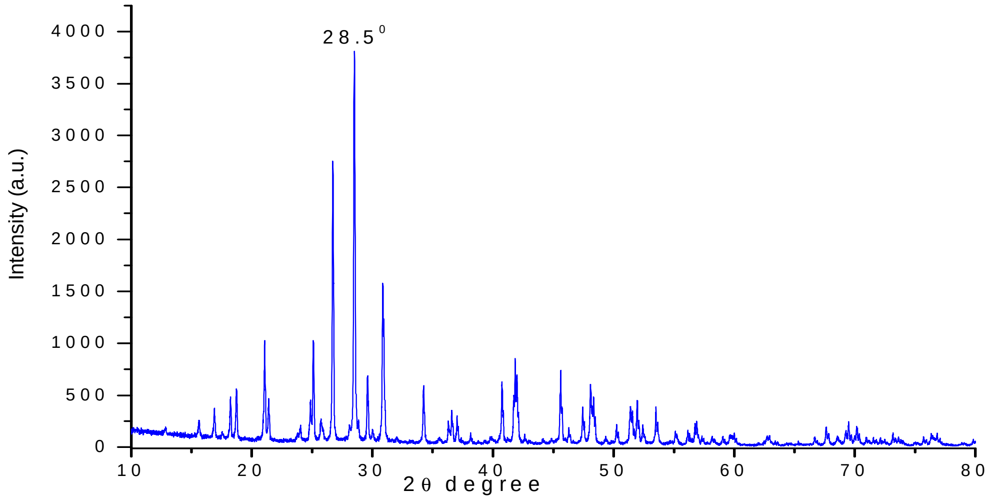

fig.1. XRD Pattern of Base Compound (LaPO4)

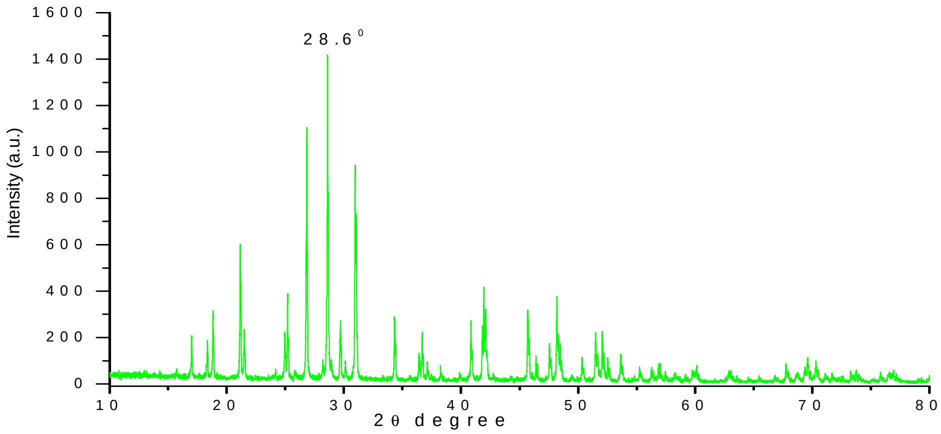

fig.2. XRD Pattern of LaPO4:Ce(1.5%),Tb(1.0% 0,Eu(1.0%)

Scanning Electron Microscope (SEM):-
Fig.3 Shows SEM image of pure $LaPO_4$ at 1200^0C for 4 hours and Fig.4. Shows SEM image of LaPO4: Ce, Eu, Tb. This appears to irregular shape having an average basal diameter of 500 nm and length of 1.5 µm.

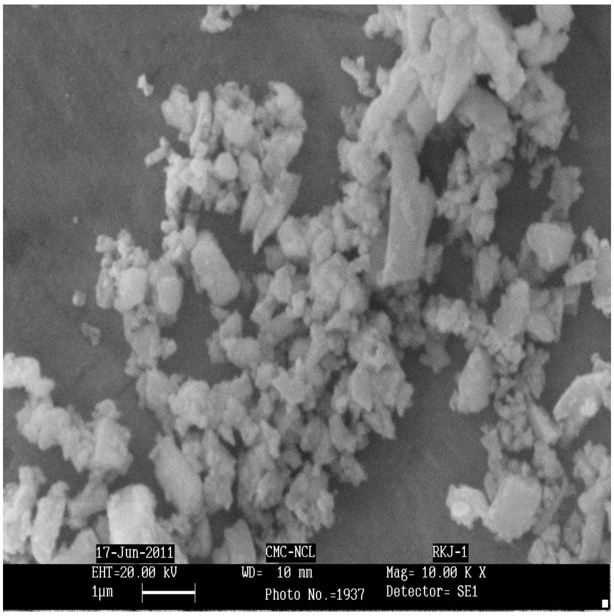

Fig.3 SEM Image of $LaPO_4$

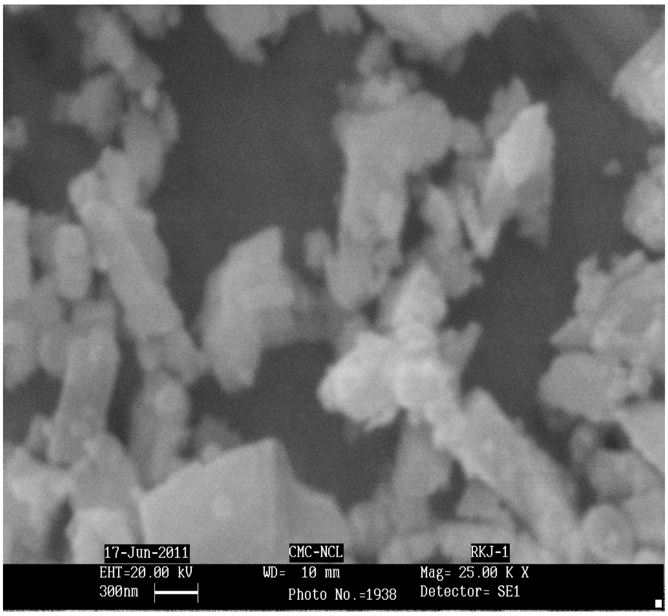

Fig.4. SEM Image of $LaPO_4$: Ce, Eu, Tb

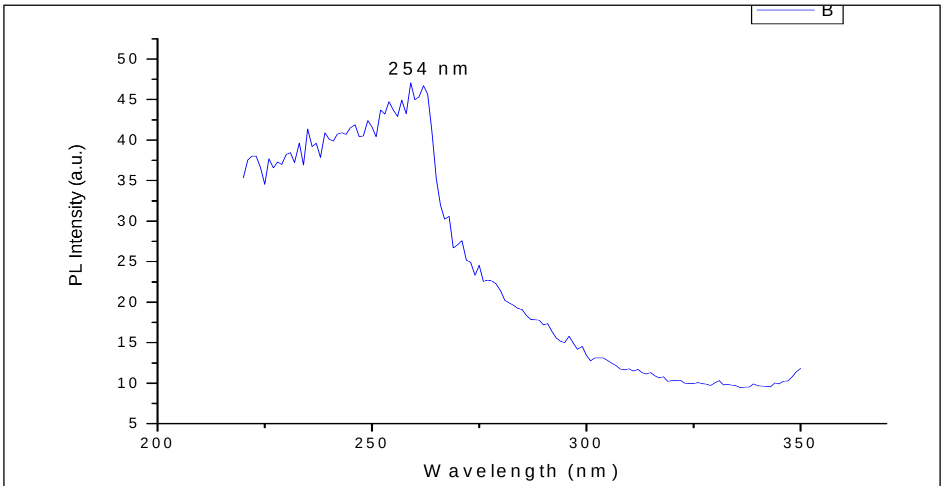

Fig 5. Excitation Spectra of undoped $LaPO_4$

Photo luminescence study:-

Fig.5. shows the excitation spectra of undoped $LaPO_4$ phosphor and fig.6 shows emission spectra of undoped LaPO4. The PL emission of pure LaPO4 was observed at 470 nm wavelength. Fig.7. shows the PL emission of doped $LaPO_4$: Ce, Eu, Tb phosphor under the excitation of 254 nm wavelength. The phosphor shows the peaks at 381, 415, 437, 457, 473, 488, 545, 589, 596, 614 and 622 nm with good intensity. In the trivalent rare earth ions, the luminescence arises mainly due to transactions within the 4 f shell. The efficiency of emission depends on the number of electrons in the 4f shell. The Tb^{3+} ion has 8 electrons in the 4f shell, which can be excited in the 4f-5d excitation band [7]. The electron in the excited $4f^7$ - 5d state remains at the surface of the ion and comes under the strong influence of the crystal field resulting in the splitting of the excitation band. The excitation Spectra thus has multiple peaks. The excited ion in the $4f^7$ - 5D

State decays stepwise from this state to the luminescent levels $5D4f_3$ or $5d4f_4$ by giving up phonons to the lattice [8]. Luminescence emission occurs from either of these states, with the ion returning to the ground state. The emission line in the green region lying at 545 nm is due to the transition $5D_4$ -$7F_6$, 585 nm due to $^5D_4$-$^7F_4$ and 620 nm due to $^5D_4$-$^7F_5$. There are in fact multiple emission lines at each of these due to the crystal field splitting of the ground state of the emitting ions [8]. As the Tb concentration increases the PL intensity also increases.

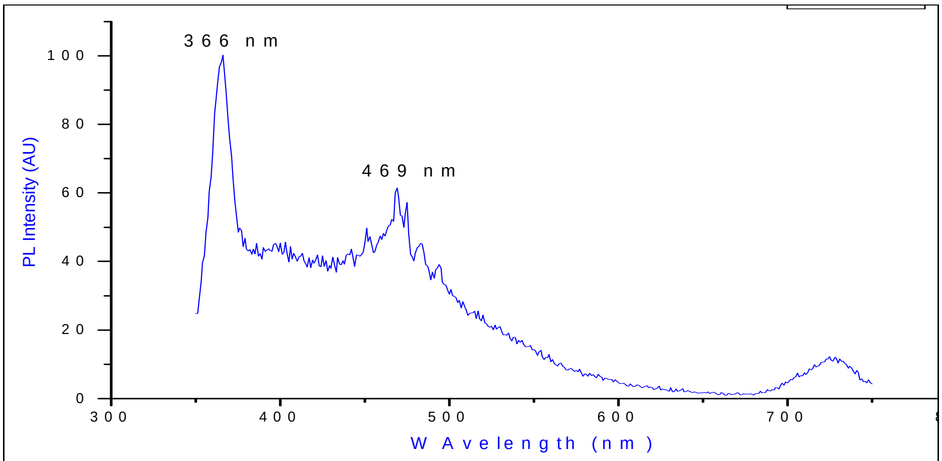

Fig 6. Emission Spectra of undoped $LaPO_4$

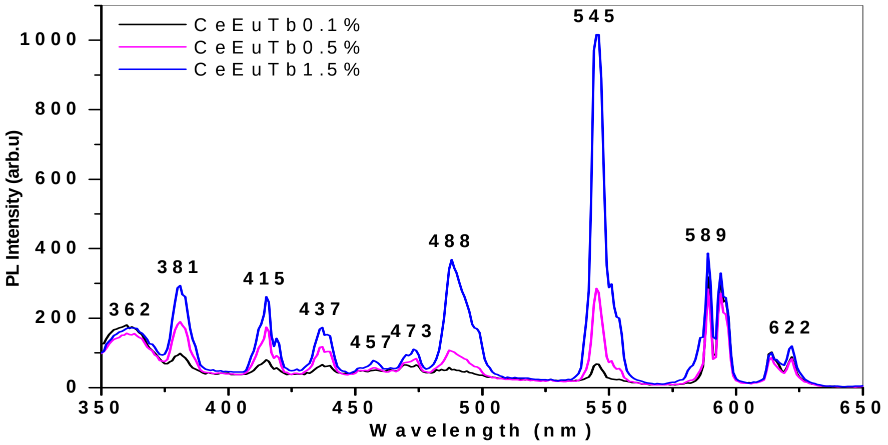

fig 7.**Emission spectrum of $LaPO_4$:Ce, Eu & Tb at 254nm Excitation**

CONCLUSION

LaPO4:Eu phosphor doped with Ce, Eu and Tb rare-earth ions, keeping Ce, Eu concentration constant and varying Tb concentration as 0.1, 0.5 and 1.5% were prepared using solid state synthesis method are successfully synthesized. The main peak in XRD pattern was found around 28.6^0 corresponding to a d- value of about $3.13A^0$, followed by other less intense peaks corresponds to the monoclinic system of crystal structure of Lanthanum Phosphate. As the Tb concentration increases the PL intensity also increases. Therefore, the $LaPO_4$: Ce, Eu, Tb phosphors can be easily applied in various types of lamp and display due to its good PL performance. In this regard, our target product is a very promising phosphor.

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