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Effect of RE Dopant (Ce & Tb) on PL and Crystallites size of Lanthanum Phosphor ($LaPO_4$)

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, 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*

ABSTRACT

The present paper reports the Photoluminescence (PL) of the $LaPO_4$ phosphor doped with Ce,and Tb rare-earth ions, keeping Ce, concentration constant and varying Tb concentration as 0.1, 0.5,1.5%.The phosphors were synthesized using the standard solid state reaction technique and ground using mortar and pestle, fired at 1200^oC for 4 hour in a muffle furnace. The crystallinity and particle morphology of the product were characterized by using XRD, SEM and particle size analysis. Also the effects of dopants on the Photoluminescent properties of the samples were studied using Spectrofluorophotometer at room temperature.

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

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INTRODUCTION

Phosphors are widely used in displays and lighting devices. Morphology of phosphors i.e. shapes and size of the powder particles is one of the key parameters to use phosphors for various applications. Spherical phosphor particle can optimize the optical and geometrical structure of phosphor layer. The particle size of a phosphor affects the amount of phosphor particles needed to produce an optimal coating for a particular application. $LaPO_4$: Ce, Tb is mainly used as a green emitting phosphor for Fluorescent lamps because of its high quantum efficiency and stability at high temperature. Phosphor having smaller particle size can provide equivalent coverage densities at lower powder weights than larger sized particles[1-4]. However, a reduction in size of a phosphor usually results in lower emission brightness because of the higher intrinsic reflection coefficient associated with the smaller particles. The milling process after synthesis of phosphor will also reduce the brightness of the phosphor particles. The $LaPO_4$: Ce, Tb phosphor has high luminescence efficiency under excitation of long wavelength UV radiation

and can used in plasma display panel [6]. However phosphor particles must have good characteristics such as their high brightness, spherical shape, fine size, and narrow size distribution for application in PDP.

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 and Tb rare-earth ions, keeping Ce 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, and Tb rare-earth ions, keeping Ce, 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) 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 & Engg. M.S.U. 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,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[5-7]. Results are shown in Fig.1 and 2 for pure $LaPO_4$ and $LaPO_4$ dopped Ce, Tb. All diffraction patterns were obtained using CuKα radiation (λ = 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 particles 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 LaPO4 phosphors is 62 nm and when doped with RE dopants, the crystallite size becomes 74.5 nm.

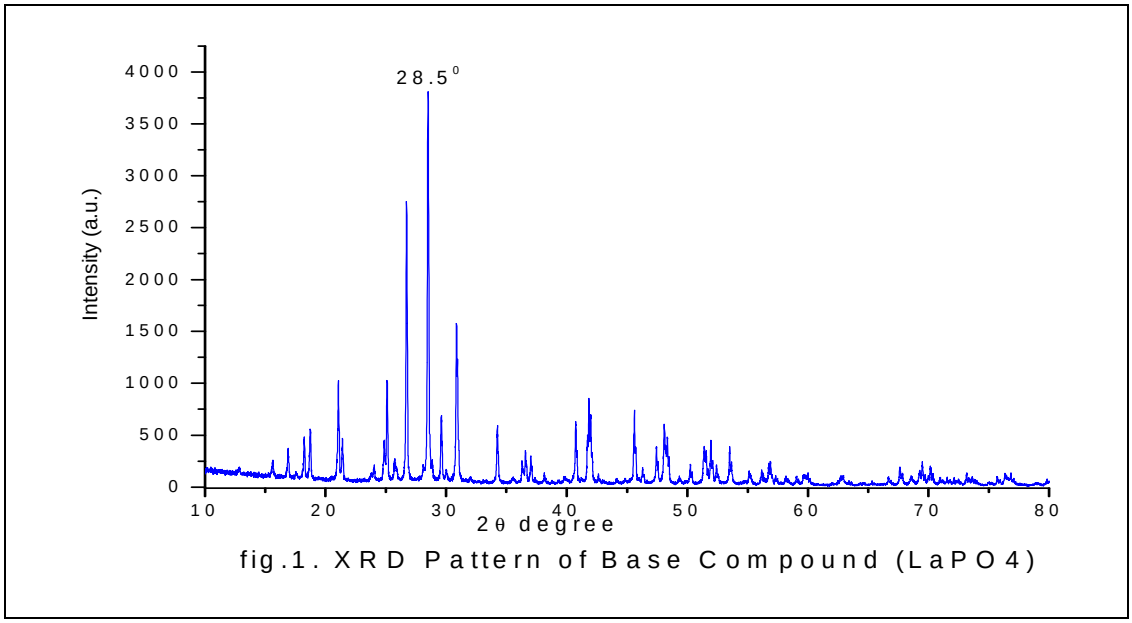

fig.1. XRD Pattern of Base Compound (LaPO4)

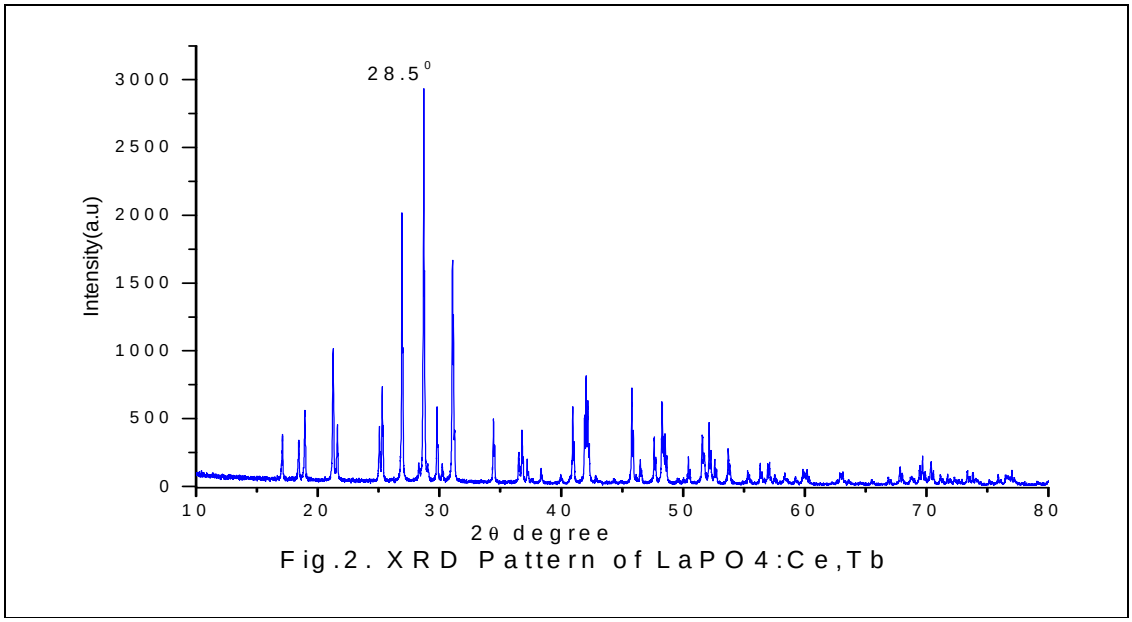

Fig.2. XRD Pattern of LaPO4:Ce,Tb

Photo luminescence study:-

Fig.3 shows the excitation spectra of undopped $LaPO_4$ and fig.4 shows the emission spectra of undopped $LaPO_4$ under the excitation of 254 nm.The bulk materials was doped by Cerium or Terbium or both. Cerium in $LaPO_4$ does not shown any specific emission. However Terbium, is known to give a narrow band emission about 545 nm [10].

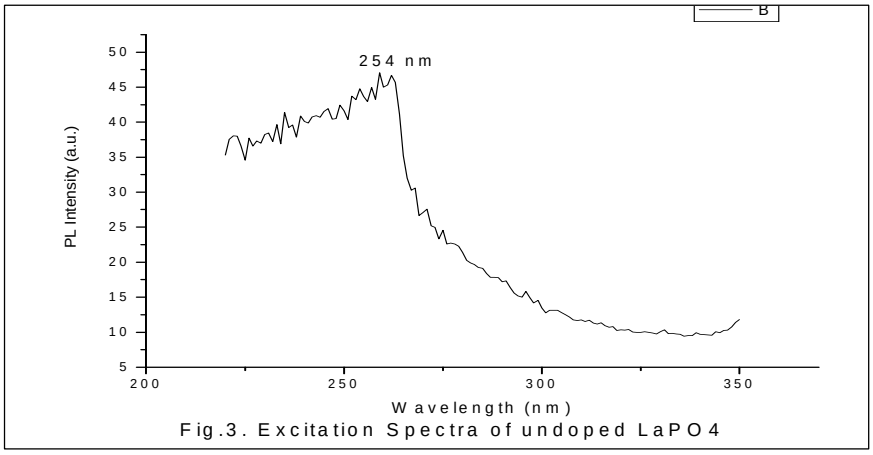

Fig.3. Excitation Spectra of undoped LaPO4

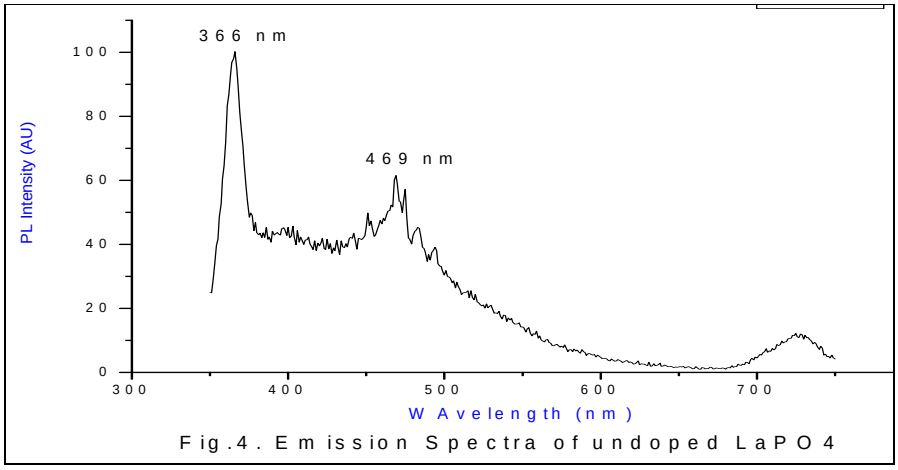

Fig.4. Emission Spectra of undoped LaPO4

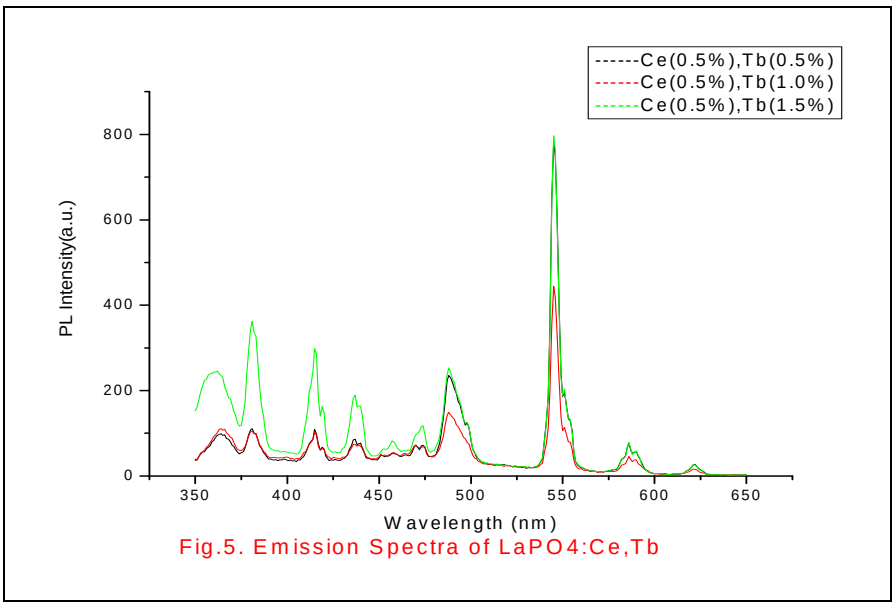

Fig.5. Emission Spectra of LaPO4:Ce,Tb

Fig.5 shows the emission spectrum of $LaPO_4$: Ce,Tb .The 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 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 th $4f^7$ - 5D state decays stepwise from this state to the luminescent levels $5D4f_3$ or $5d4f_4$ by giving up phonons to the lattice [8-9]. 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.

CONCLUSION

$LaPO_4$ phosphor doped with Ce and Tb rare-earth ions, keeping Ce 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 28.5^0 corresponding to a d- value of about $3.10A^0$, 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. The PL intensity is very high therefore; the $LaPO_4$:Ce, Tb phosphors can be easily applied in various types of lamp and display.

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