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Synthesis and photoluminescent properties of LaPO₄: Ga³⁺ phosphor

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ABSTRACT

Pure LaPO₄ and LaPO₄ : Ga^{3+} (0.5 mol%) phosphor was synthesized by the conventional solid-state reaction method. Photoluminescence (PL) properties of pure LaPO₄ and LaPO₄ : Ga^{3+} phosphor was investigated. The excitation spectrum of pure LaPO₄ and LaPO₄ : Ga^{3+} phosphor monitored under 400nm wavelength was characterized by a broad band ranging from 220-280nm with a maximum intensity peak at 249nm(4.98eV) and 248nm(5.0eV) respectively. Upon excitation at 254nm wavelength, the emission spectrum of pure LaPO₄ and LaPO₄ : Ga^{3+} phosphor emits a broad band range from 400-650nm with maximum intensity peak at 469nm(2.64eV, Blue) and 594nm(2.08eV, Red) with the full width at half maximum (3nm). The color coordinates for the LaPO₄ : Ga^{3+} (0.5 mol%) phosphor are x=0.5691 and y= 0.4303. LaPO₄ : Ga^{3+} (0.5 mol%) phosphor having excellent colour tunability of red light.

Keywords: Photoluminescence; XRD; SEM; FTIR; phosphor; rare-earth ions; solid state reaction technique;CIE; particle size analysis.

INTRODUCTION

The optical properties of the lanthanides in inorganic compounds, their preferred valence, and their electron donating or accepting properties are all determined by the electronic structure, i.e., by the relative and absolute level energies of the lanthanide impurity states and the host conduction and upper valence band states. Rare-earth orthophosphates(REPO₄) are a very interesting class of host lattices of activator ions due to their physic-chemical inercy(high insolubility, high thermal stability), thus providing durable phosphors[1]. Lanthanide orthophosphate (LnPO₄) belongs to two polymorphic types, the monoclinic monazite type (for La to Gd) and quadratic xenotime type (for Tb to Lu). The LaPO4 crystalline matrix is widely used for development of phosphors for compact fluorescent lamps, plasma display panels, field emission display, optical amplifiers, laser active mediums and electrical conduction. Process ability of this material and resistance for atmospheric influence caused the interest to study of the luminescent properties of LaPO₄ nanoparticles. Monoclinic lanthanum phosphate is a compound with extremely low solubility in water, and high thermal and chemical stability; it has been proposed for use in broad applications. The luminescent properties of rare-earth phosphates can be conferred by the presence of lanthanide(III) ions as activators due to their intense and narrow emission bands arising from f-f transitions, which are proper for the generation of individual colours in multiphosphor devices [2-4]. Lanthanide orthophosphate (LaPO₄) and lanthanide (III)-doped lanthanide orthophosphate have attracted much attention due to their unique photoluminescent properties and various potential applications in various areas, including color displays, light sources, field-effect transistors, solar cells, and biomedical labels,nanoscintillators for radiotherapy [5,6]. It is known that the LaPO₄ has a monoclinic phase of monazite structure crystallographically, wherein La³⁺ ion is nine coordinated to oxygen atoms, four oxygens forming a distorted tetrahedron interpenetrating a quasiplanar pentagon formed by another five [7-10]. The La³⁺ ion site in the monazite structure can be easily substituted by any other lanthanide ions. To improve luminescent properties of nanocrystalline phosphors, many preparation methods have been used, such as solid state reactions, sol-gel techniques, hydroxide precipitation, hydrothermal synthesis, spray pyrolysis, laser-heated evaporation, and combustion synthesis[11-13].

In this paper pure LaPO₄ and LaPO₄ :Ga³⁺ (0.5 mol%) phosphor was prepared by the solid state reaction method in air at 1200⁰C, and their luminescent properties were studied. Optimization of the concentration of activator ions incorporated into the host lattice during the synthesis of the phosphor powders is essential for developing highly luminescent RE³⁺ doped nanocrystalline phosphors as well as for the growth of grain particles. Photoluminescence studies and CIE co-ordinates of pure LaPO₄ and LaPO₄ :Ga³⁺ phosphor reveals that the emission colour having excellent colour tunability of red light So this material may be a potential luminescent material.

MATERIALS AND METHODS

Synthesis:

The pure LaPO₄ and LaPO₄ :Ga³⁺ (0.5 mol%) phosphor was synthesized by using the conventional solid-state reaction method. The formation of the phosphor powder occurs according to the following chemical equation.

For pure LaPO₄ and LaPO₄: Ga (0.5%)

 $La_2O_3 + 2 (NH_4) H_2PO_4 \rightarrow 2LaPO_4 + 2NH_4OH \uparrow + H_2O\uparrow$

 $La_2O_3 + 2 (NH_4)H_2PO_4 + GaO \rightarrow 2LaPO_4$: Ga (0.5%)

The starting materials were lanthanum oxide (La_2O_3), Diammonium Hydrogen Phosphate [(NH₄)₂ H₂ PO₄),Galium oxide(GaO) of 99.9% purity. They were weighed with a certain stoichiometric ratio. The composite powders were grinded in an agate mortar and then placed in an alumina crucible with the lid closed. After the powders had been sintered at 1200 °C for 3 hr in a muffle furnace and then cooled to room temperature. All the samples were again ground into fine powder using an agate mortar and pestle about an hour. Finally, the powders were sieved again through 100 μ m sieve. The emission and the excitation spectra of the synthesized powders were characterized with a spectroflurophotometer(Shimadzu RF – 5301 PC) with xenon lamp as excitation source. The Commission International de l'Eclairage (CIE) co-ordinates were calculated by the spectrophotometric method using the spectral energy distribution. The chromatic coordinates (x, y) of prepared materials were calculated with colour calculator version 2, software from Radiant Imaging[14].

RESULTS AND DISCUSSION

Photoluminescence Study of pure LaPO₄ and LaPO₄: Ga³⁺ (0.5 mol%) phosphor

Fig.1and Fig.2 shows PL excitation and emission spectra of pure LaPO₄ and LaPO₄ :Ga³⁺ (0.5 mol%) phosphor recorded at room temperature. The excitation spectrum of LaPO₄ :Ga³⁺ (0.5 mol%) phosphor monitored under 400nm wavelength was characterized by a broad band ranging from 220-400nm with a maximum intensity peak at 264nm. The La³⁺ ion site in the monazite structure can be easily substituted by any other lanthanide ions. The La³⁺ ion site in the monazite structure can be easily substituted by any other lanthanide ions. The luminescent characteristics of the particles depend on its size and other properties including the degree of crystallization, defects and the valence state of the doped activator ions. The shape of the emission spectra and emission peak wavelength is independent of the excitation wavelengths. Upon excitation at 254nm wavelength, the emission spectrum of LaPO₄ :Ga³⁺ (0.5 mol%) phosphor emits a broad band range from 400-650nm with maximum intensity peak at 589(red)nm with the full width at half maximum (3nm).

CIE (1931-Chart) Coordinates of pure LaPO₄ and LaPO₄:Ga³⁺ (0.5 mol%) phosphor

Most lighting specifications refer to color in terms of the 1931 CIE chromatic colour coordinates which recognizes that the human visual system uses three primary colours: red,

green, and blue. In general, the color of any light source can be represented on the (x, y) coordinate in this color space. The color purity was compared to the 1931 CIE Standard Source C (illuminant Cs (0.3101, 0.3162)). The chromatic coordinates (x, y), was calculated using the color calculator program radiant imaging.

Fig.4 shows the CIE co-ordinates of (chart -1931) of pure $LaPO_4$ and $LaPO_4$: Ga^{3+} (0.5 mol%) phosphor. The color co-ordinates for the pure $LaPO_4$ are x=0.1609 and y=0.0305 and $LaPO_4$: Ga^{3+} (0.5 mol%) phosphor are x=0.5691and y=0.4303. The location of the colour coordinates of the monophosphate powder on the CIE chromaticity diagram presented in Fig.4 indicates that the colour properties of the phosphor powder prepared by solid state

reaction method are approaching those required for field emission displays. This phosphor is having excellent colour tunability of red light.

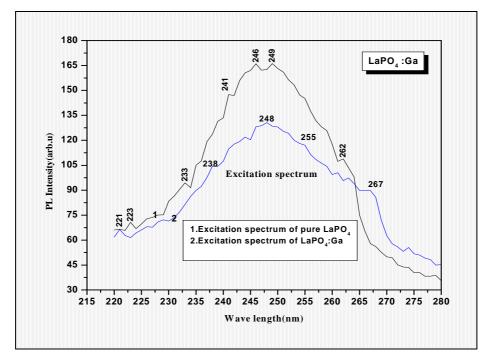


Figure 1 Exicitation spectrum of Pure LaPO₄ and LaPO₄: Ga³⁺ (0.5%) phosphor

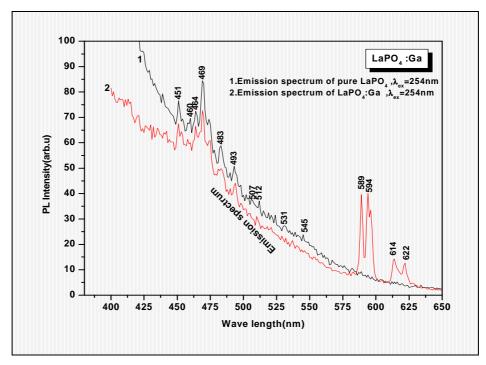


Figure.2 Emission spectra of Pure LaPO₄ and LaPO₄ :Ga $^{3+}$ (0.5 mol%) phosphor

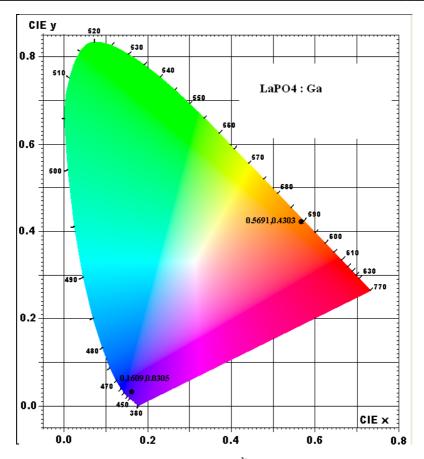


Figure. 3 CIE Co-ordinates of pure LaPO₄ and LaPO₄ :Ga³⁺ (0.5 mol%) phosphor depicted on 1931 chart

CONCLUSION

Pure LaPO₄ and LaPO₄ : Ga^{3+} (0.5 mol%) phosphor powder were successfully synthesized through solid state reaction method at high temperature($1200^{0}C$) and the luminescent properties of sample was studied. The PL characterization demonstrates that the pure LaPO₄ and LaPO₄ : Ga^{3+} (0.5 mol%) phosphor shows the most intense emission. The Commission International de 1'Eclairage [CIE] co-ordinates of LaPO₄ : Ga^{3+} (0.5 mol%) phosphor exhibit the excellent colour tunability of red. Therefore, the LaPO₄ : Ga^{3+} (0.5 mol%) phosphor can be easily applied in various types of lamp and display due to its good PL performance.

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