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Photoluminescence properties of mixed fuel combustion synthesized Ce^{3+} ions doped $Y_3Al_5O_{12}$ phosphor

N.S. Sawala*, C.B. Palan, A.O. Chauhan, S.K. Omanwar

Department of Physics, Sant Gadge Baba Amravati University, Amravati 444602, India

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ABSTRACT

The series of Ce^{3+} ion doped YAG phosphor have been synthesized by mixed fuel combustion technique. The phase purity and crystalline size of phosphors confirmed by X-ray diffraction (XRD) analysis while surface morphology studied by scanning electron microscopy (SEM). The photoluminescence (PL) studied within 200–700 nm wavelengths. Commission Internationale de l'Eclairage (CIE) co-ordinate of phosphor studied for and found suitable for solid state lighting and white LEDs. The nanometric particle size 15.838 nm and 18.683 nm at 900 °C and 1000 °C, respectively, make the mentioned synthesis method idyllic.

1. Introduction

Due to several advantages such as long operating lifetime, compact in size and high energy efficiency the solid-state white-light emitting diode (w-LED) are found to be champion as next-generation light sources. Additionally, they have environmental merits because they do not contain mercury vapour as a light source [1]. A white light can be obtained by simply mixing red, green, and blue light each with equal proportion. But this move produces a very low quality white light and LED colour properties varies due to manufacturing tolerances makes the multiple colour (red, blue and green) LED impractical. White light can be produced by a combination of a blue LED chip with a yellow emitting phosphor [2]. However, this kind of white light has some drawbacks, such as low colour-rendering index (CRI) and high correlated colour temperature (CCT), owing to the deficiency of red colour emission [3,4]. An alternative way to produce white light with a high CRI may be based on a combination of a near ultraviolet (near-UV) LED chip (380–420 nm) with red, blue and green, emitting phosphors [4–6]. By considering the optical requirements for white light, many new phosphors have been developed to overcome the above noticed. The very common and effortless method to manufacture wLED is based on a combination of a indium gallium nitride (InGaN) chip with a broad-band greenish yellow emitting Ce^{3+} ion doped yttrium aluminium garnet (YAG:Ce^{3+}) phosphor [7,8]. The, YAG:Ce^{3+} has been the mostly accepted wavelength conversion phosphor for blue-emitting InGaN based LEDs as it can effectively absorb blue light and emits bright yellow light despite its shortage of the red portion in the emission spectrum [9,10]. Most of the time YAG phosphors are usually prepared via a solid-state reaction which requires high temperature around 1600–1800 °C and prolonged heating are required to obtain the pure crystalline phase, and this method also requires several hours of sintering and milling. In recent years, a lot of synthesis techniques such as co-precipitation method, sol–gel, combustion, hydrothermal synthesis and spray-pyrolysis synthesis were utilized to obtain the pure desired phase of phosphor [11]. However, they are time consuming and complicated procedures.

The employment of combustion method has been extensively widen and developed looking at experiences of last few years in the preparation of Phosphors such as borates, silicates, aluminates, and oxides [12–14]. The advantages of using the combustion method are low cost, highly efficient and time saving approach to produce highly stable materials. Moreover, under suitable circumstances uniform and narrow distribution of particles with regular crystalline size could be obtained by the combustion synthesis method. Thus, the striking features of combustion synthesis make it attractive as well as focus on research for more development [15].

2. Experimental

2.1. Synthesis of phosphors

The powder samples of Ce^{3+} ion doped in host $Y_3Al_5O_{12}$ (YAG) were prepared by using mix fuel combustion synthesis method. The

* Corresponding author. Tel.: +91 8806091765.
E-mail address: nssawala@gmail.com (N.S. Sawala).

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Table 1
Molar ratio of precursors used for material preparation and corresponding chemical reaction.

Compounds	Molar ratio of the precursors				
$Y_{(3-x)}Ce_xAl_5O_{12}$	$Y(NO_3)_3$	$Al(NO_3)_3{\cdot}9H_2O$	$Ce(NO_3)_3{\cdot}6H_2O$	$C_2H_5NO_2$	CH_4N_2O
	$3-x$	5	x	5	12.5

$(3-x)Y(NO_3)_3 + 5Al(NO_3)_3{\cdot}9H_2O + xCe(NO_3)_3{\cdot}6H_2O + 5C_2H_5NO_2 + 12.5CH_4N_2O \rightarrow \mathbf{Y_{(3-x)}Ce_xAl_5O_{12}}$ + (Gaseous product like NO_3, N_2, CO_2, and H_2O)

precursors Y_2O_3 (99.9% AR), HNO_3 (69% AR), $Al(NO_3)_3{\cdot}9H_2O$ (99.9% AR), $Ce(NO_3)_3{\cdot}6H_2O$ (99.9% AR)and CH_4NO_2 (urea 99% AR) and $C_2H_5NO_2$ (glycine 99% AR) were used for synthesis of YAG doped with Ce^{3+} ions. The mixed (glycine and urea) fuel was used for synthesis since glycine has exothermic reaction with yttrium nitrate and urea with aluminium nitrate [16]. The composition of each chemical weighed in proper stoichiometric ratio. Most of the precursor components were available in the form of nitrate salts. Yttria (Y_2O_3) was converted into corresponding nitrate salts ($Y(NO_3)_3$) by suitable reaction with nitric acid (HNO_3). First sample of YAG doped with Ce^{3+} ion having molar concentration of 0.005 mol was prepared at different sintering temperature. The effect of sintering temperature on PL properties was studied and optimum sintering temperature was decided. The stoichiometric amounts of the ingredients were thoroughly mixed in an Agate Mortar with adding little amount of double distilled water. The materials then transferred into china basin and heated on heating menthol at about 80 °C so as to obtained clear solution. The solution was then introduced into a pre-heated muffle furnace maintained at temperature 500 °C for few minutes. The mixture molted first and gases like CO_2, N_2, and H_2O etc were evolved. Frothing and swelling of mixture took place and then combusted with the formation of foam. The homogeneous solution ignites to burn with yellow flame sustain for few seconds which gave a voluminous, foamy powder. The resulting foamy sample was gently pressed and crushed to obtain fine powder and then annealed in air at temperature 500, 800, 900 and 1000 °C for 2 h, respectively. From the study of PL, XRD and SEM the optimum annealing temperature was decided to be 900 °C for further series of $Y_{(3-x)}Ce_xAl_5O_{12}$ (x = 0.01, 0.03, 0.05 and 0.07 mol) phosphor. The detail of stoichiometric molar amount of each precursor used for phosphors synthesis is given in Table 1.

2.2. Characterizations of materials

The confirmation of as prepared materials was done by XRD method by using Rigaku miniflex II X-ray diffractometer with scan speed of 2.000°/min and Cu K_α (λ = 0.15406 nm) radiation in the range 10° to 80°. The structural and morphological characteristics i.e., particle size and shape of particle sample was studied using a SEM analysis. The measurement was performed using a ZEISS EVO/18 Research. In this study, sample in powder form (100–150 μm) was placed directly on sample holder of SEM for imaging. The PL emission and excitation spectra were recorded at room temperature on (Hitachi F-7000) fluorescence spectrometer associated with 450 W Xenon discharge lamp. The measuring parameter such as width of monochromatic slit (1.0 nm), photomultiplier tube (PMT) detector voltage, scan speed (240 nm/min), spectral resolution were kept constant throughout the analysis of materials.

3. Results and discussion

3.1. Structural confirmation

Fig. 1 represents the XRD of YAG doped with 0.005 mol of Ce^{3+} ions at different temperatures 500 °C for few minutes, 500, 800, 900 and 1000 °C for 2 h, respectively, represented by curves (a)–(e) in Fig. 1. The formation of the YAG in the crystalline phase prepared by

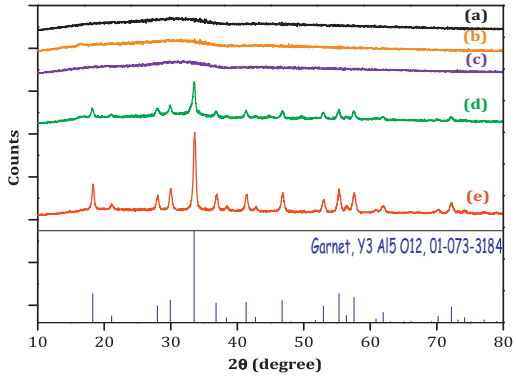

Fig. 1. XRD patterns of Ce^{3+} ion doped YAG phosphors annealed d at different temperatures (a) 500 °C for few minutes, (b) 500 (c) 800 (d) 900 and (e) 1000 °C for 2 h, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

mixed fuel combustion method was confirmed by XRD pattern. The XRD pattern for Ce^{3+} doped YAG annealed at 900 °C (d) and 1000 °C (e) for 2 h agreed well with the standard data from ICDD file (01-073-3184). The Ce^{3+} doped YAG has cubic crystal structure with space group *Ia*-3*d* (2 3 0) and lattice parameter a = 11.990 Å. The high intensity peaks were observed at 33.39, 18.11, 55.20, 57.47, 29.78 and 46.66 which are corresponding to (4 2 0), (2 1 1), (6 4 0), (6 4 2), (4 0 0) and (5 3 2), respectively. The XRD also showed that the prepared material is completely crystalline and was in single phase. Moreover, from analysis of the XRD pattern it is understood that the introduction of activator Ce^{3+} ion does not influence the crystal structure of the YAG sample, because both Ce^{3+} and Y^{3+} ions have similar ionic radius (Ce^{3+}: 1.14 Å, Y^{3+}: 1.02 Å) and the cerium ion enters the lattice substitution ally in yttrium sites. This expected as concentration of Ce^{3+} ion was very less. The average crystalline size of this phosphor was found to be 15.838 nm and 18.683 nm at 900 °C and 1000 °C, respectively, as estimated by following Debye-Scherrer's formula.

$$D = \frac{K\lambda}{\beta \cos\theta}$$

where θ is Bragg angle of diffraction lines, K is a shape factor taken as 0.90, λ is wavelength of incident X-rays (λ = 0.154 nm) and β is full-width at half maximum (FWHM in radians).

The crystalline increases with increase of temperature which is obvious since the particles start to bond together and the size enlarges with elevation in temperature.

Fig. 2 showed the XRD patterns for $Y_{(3-x)}Ce_xAl_5O_{12}$ (x = 0.005, 0.01, 0.03, 0.05 and 0.07 mol) which again well agreed with the standard data from ICDD file (01-073-3184). The rest of the crystal parameters remain unaffected even at high concentration of Ce^{3+} ion [17]. This is possibly due to similar ionic radii of both trivalent ions. Moreover their periodic properties are almost similar as both are d block elements.

3.2. Surface morphology

Fig. 3 shows the SEM images of Ce^{3+} doped (0.005 mol) YAG phosphors annealed at 900 °C (d) and 1000 °C (e) prepared by