Nuclear Instruments and Methods in Physics Research B 379 (2016) 69–72

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Luminescence properties of $La_2O_3:Eu^{3+}$ nanophosphor prepared by sol–gel method

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A R T I C L E I N F O

Article history:
Received 12 November 2015
Received in revised form 8 April 2016
Accepted 12 April 2016
Available online 19 April 2016

Keywords:
X-ray diffraction
Gamma rays
Thermoluminescence

A B S T R A C T

Undoped and Eu^{3+} doped La_2O_3 nanophosphor are synthesized by low temperature sol–gel technique. The synthesized samples are characterized by X-ray diffraction (XRD) and average crystallite size is found to be ~18 nm and ~23 nm for undoped and Eu^{3+} doped La_2O_3 respectively. Gamma ray irradiated undoped La_2O_3 shows high intense thermoluminescence (TL) glow peak at 640 K and weak TL glow peak at 443 K and the high intense peak intensity is sub linear increase with γ-dose. Whereas Eu^{3+} doped La_2O_3 nanophosphor show a prominent TL glow peak at 640 K and its TL intensity linearly increases up to 1 kGy. The kinetic parameters are estimated using glow curve deconvoluted (GCD) technique. TL emission of γ-ray irradiated Eu^{3+} doped La_2O_3 show peaks at 508, 586, 619 and 706 nm are attributed to Eu^{3+} transition peaks.

1. Introduction

Rare earth sesquioxides have been extensively studied in recent years due to their unique electronic, optical, and chemical properties [1–3]. Among these oxides lanthanum oxide (La_2O_3) has a number of industrial and technological applications [4,5]. Sesquioxides can be obtained in the form of thin films, nanoparticles and single crystals, have been investigated for their potential applications in luminescent displays, dosimetry, and light emitting diodes (LEDs). Further Ln^{3+} ions can easily substitute in La^{3+} lattice due to similar chemical, ionic radii and electro negativity of La^{3+} ion and resulting in high efficient optical, electrical and magnetic properties. Due to quantum confinement effect, optical and luminescent properties more efficient in nano size than bulk materials.

Thermoluminescence (TL) is observed when a material irradiated for gamma, X-ray, beta and heavy ion beams etc, part of the irradiation energy is used to transfer electrons to traps. This energy, stored in the form of the trapped electrons, is released by raising the temperature of the material, and the released energy is converted to luminescence. Material such as LiF:Mg, $CaSO_4$:Dy, CaF_2:Dy and Al_2O_3:C are good thermoluminescence materials because of their applications in radiation dose measurement, age determination and defect structure analysis in solids [6,7]. However, these materials not suitable for high dose measurement because TL intensity saturates at lower doses due to the overlapping of ionized zones [8]. Hence, research has been going on to improve the dosimetry applications of nano materials by changing synthesis technique or adding inorganic and organic agents [9,10].

A wide variety of techniques are available for preparation of nano materials such a solid state reaction, sol gel, microwave synthesis, ball milling, hydrothermal, solution combustion and co-precipitation methods [9]. Among these techniques sol–gel technique is adopted for the synthesis of La_2O_3 nano particles because, this technique yields products of a high surface area to volume ratio, high homogeneity and allows fine control of the chemical composition [10,11]. In the present work, gamma ray irradiated TL glow curve and TL emission properties of undoped and Eu^{3+} doped La_2O_3 nanoparticles are studied.

2. Materials and methods

Undoped and Europium doped lanthanum oxide nano materials are synthesized by sol–gel technique using lanthanum oxide [99.8% pure (La_2O_3), Aldrich chemicals], anhydrous citric acid [99.5% pure GR ($C_6H_8O_7$), Merck chemicals], Europium oxide [99.9% pure (Eu_2O_3), Aldrich chemicals] and nitric acid [(HNO_3), Merck chemicals] as raw materials. The ratio of citric acid to La^{3+} is considered as 2.0 [10]. Stoichiometric amount of lanthanum oxide and europium oxide are separately dissolved with dilute

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http://dx.doi.org/10.1016/j.nimb.2016.04.033
0168-583X/

nitric acid to get lanthanum nitrate and europium nitrate followed by dissolving in 50 ml of double distilled water. The resultant solutions are mixed and refluxed at room temperature for 3 h. Finally, the stoichiometric amount of citric acid is slowly added, to act as a chelating agent and refluxed at 60–65 °C for 3 h. During refluxing, the solution slowly evaporated and turned into a reddish brown gel. Then gel is dried at 110 °C in an oven to obtain powder and annealed at 800 °C for 4 h.

The Phase purity of the samples is characterized by X-ray diffraction (XRD) technique using Rigaku Miniflex II diffractometer with Cu Kα (λ = 1.541 Å) radiation. For TL measurements, 30 mg of nanophosphor is exposed to γ-rays (^{60}Co) in a dose range 0.05–12.00 kGy. TL measurements are carried out using TL Reader (model: TL1009I; Nucleonix Systems Pvt Ltd, India) in the temperature range 325–750 K. TL emission recorded using Ocean Optics spectrometer (USB4000).

3. Results and discussions

3.1. X-ray diffraction

Fig. 1 shows the XRD patterns of annealed undoped and Eu^{3+} doped La_2O_3. All the diffraction peaks are well indexed as a hexagonal phase with space group P3m-1 (JCPDS card No. 05-0602) and the lattice parameter are found to be a = 4.036 Å, c = 6.201 Å and a = 3.397 Å, c = 6.129 Å for undoped and doped La_2O_3 respectively [12]. It is observed that weak diffraction peaks at 36.07°, 46.95° and 56.25° are indexed to La $(OH)_3$. Presence of hydroxide peaks in the sample due to the hygroscopic nature of La_2O_3 at room temperature. The average crystallite size is calculated by Scherrer's formula and found to be ~18 nm and ~23 nm for undoped and Eu^{3+} doped La_2O_3 respectively.

3.2. Thermoluminescence

Fig. 2 shows TL glow curve of La_2O_3 samples irradiated with γ-rays (^{60}Co) with energy 1.2 MeV for different exposures in the range 0.05–12.0 kGy recorded at a heating rate of 5 Ks^{-1}. Undoped sample shows two TL glows with high intense peak at 640 K and weak one with a peak at 443 K. These TL glows are attributed to the presence of oxygen vacancies in the sample [12,13]. Fig. 3 shows TL glow peak (640 K) intensity and glow peak temperature as a function of γ-irradiation dose. It is observed that the glow peak

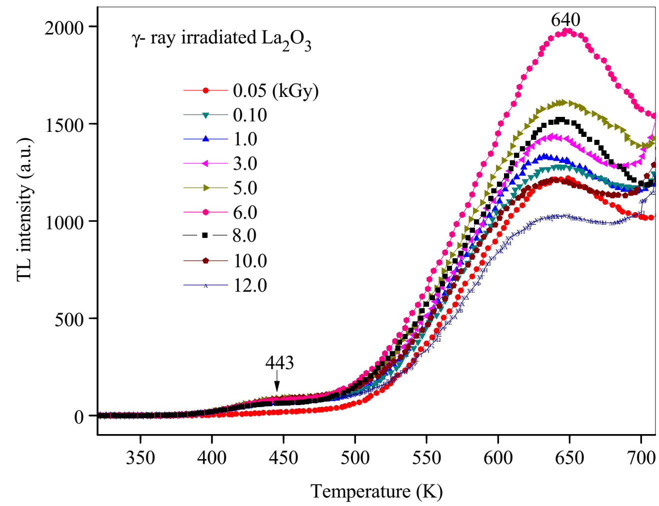

Fig. 2. TL glow curves of γ-ray irradiated La_2O_3 nanoparticles.

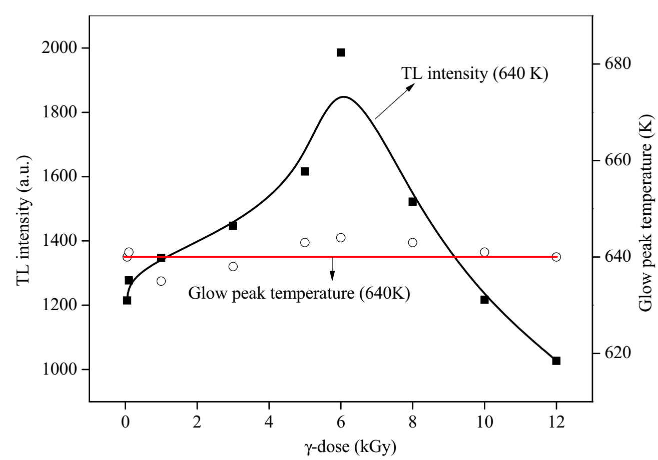

Fig. 3. Variation of TL glow peak intensity and glow peak temperature with γ-dose for undoped La_2O_3 nanoparticles.

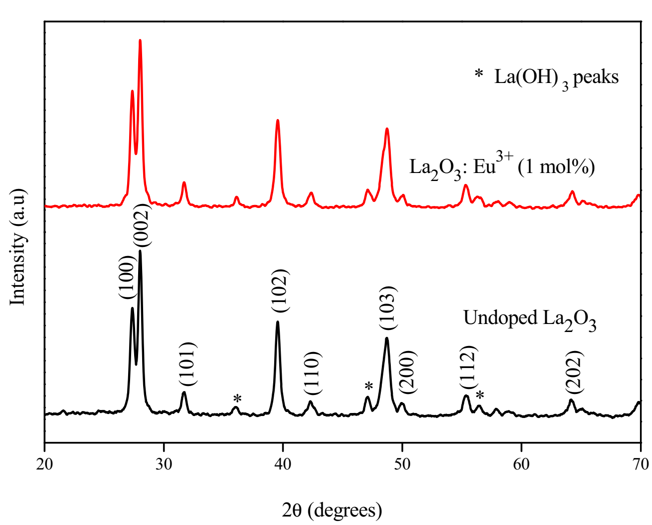

Fig. 1. X-ray diffraction pattern of undoped and Eu^{3+} (1 mol%) doped La_2O_3 nanoparticles.

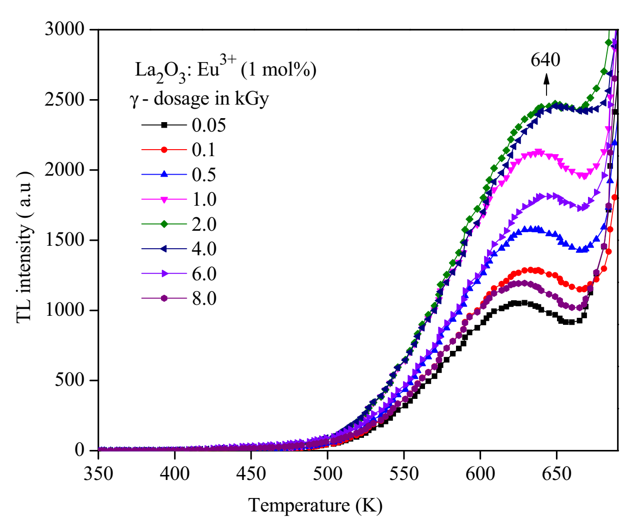

Fig. 4. TL glow curves of γ-ray irradiated $La_2O_3:Eu^{3+}$ (1 mol%) nanoparticles.