JETIR.ORG ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

Thermoluminescence studies of undoped and Ce-doped $GdPO_4$ phosphor

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Abstract

In the present work, Ce^{3+}-activated $GdPO_4$ phosphors were prepared by the solid-state reaction method. The XRD pattern confirmed that these phosphors encompass a monoclinic phase with space group $P_{21/n(}$14) with cell parameters a = 6.65Å, b = 6.85Å, and c = 6.33Å. The TL studies revealed that the synthesized undoped sample showed a single broad peak centered at 82 °C and Ce doped sample showed a single broad peak centered at 93^{0}C. The optimum TL intensity was observed for samples with 3 mol% of Ce. Since no remarkable variation in the TL intensity with different UV doses was recorded, hence it is not suitable for radiation dosimetry applications.

Keywords: XRD, Thermoluminescence, Dosimetry.

5.1 Introduction

Thermoluminescence is a phenomenon exhibited by certain materials, where absorbed energy from radiation is released as light upon heating. This occurs when an electron or hole is trapped within a material's crystal lattice due to exposure to ionizing radiation, and subsequent heating provides the energy for the trapped charge carrier to escape and recombine, emitting light in the process [1].

The phenomenon of thermally stimulated emission, first named "Thermoluminescence" by Wiedemann in 1903, involves the emission of light from a substance when heated after being exposed to radiation [2,3]. Early investigations into thermoluminescence in the 1930s, particularly the experimental and theoretical work of Urbach in 1930, laid the groundwork for understanding this phenomenon. A significant leap came in 1945 when Randall and Wilkins developed a model that enabled quantitative calculations of thermoluminescence kinetics, pushing the field towards more practical applications. [4–7].

The mid-20th century saw a surge in using thermoluminescence for dosimetry, with pioneers like Daniels in 1953 and Cameron in 1961 exploring the properties of materials for measuring radiation doses. While dosimetry remains a key application, recent years have witnessed the expansion of thermoluminescence studies into diverse fields.

Thermoluminescence now plays a crucial role in archaeology, earth science, solid-state physics, medical science, and more, highlighting its versatility as a research tool. Also because of the ease of preparation, high thermoluminescence (TL) yield, and modification for various applications, phosphate glasses-based dosimeters are present of specific interest in their physical and chemical properties [8-10]. As in the case of powder dosimeters, the particle size of the used sample may show a significant role in thermoluminescence processes, light emission intensity, linearity of dosimetric response, etc. [11-13].

The study of thermoluminescence properties of cerium-doped gadolinium phosphate crystals has garnered significant attention in recent years due to their potential applications in various fields. The gadolinium phosphate phosphor, $GdPO_4$, has garnered significant attention also due to its unique optical properties, particularly its ability to emit bright and efficient luminescence under ultraviolet excitation[14]. The incorporation of rare earth ions, such as terbium (Tb^{3+}), into the $GdPO_4$ host matrix can further enhance its luminescent properties [15]. Previous studies have explored the energy transfer mechanisms between the host lattice and the activator ions, as well as the effects of dopant concentration on the overall luminescence performance [14,16]. Some so many minerals and compounds show thermoluminescence (TL) properties but then only some materials can meet the requirements of an ideal dosimeter. Recently, numerous phosphate materials have been considered for low-dose dosimetry, and among the various phosphates, $ABPO_4$-type material displays interesting TL properties.

5.2 Experimental technique

The following himedia chemicals have been used as starting materials for the synthesis of Gadolinium phosphate and doping materials.

- Gadolinium oxide (Gd_2O_3)
- Di-ammonium hydrogen phosphate $(NH_4)_2HPO_4$
- cerium (III) oxide Ce_2O_3
- Wetting media: Distilled water and Acetone

$GdPO_4$ activated with Ce^{3+} phosphors synthesized through the solid-state reaction method. For the synthesis of host material $GdPO_4$, himedia chemicals of analytical reagent gadolinium oxide (Gd_2O_3) and di-ammonium hydrogen phosphate $(NH_4)_2HPO_4$ were used as a starting material. At the same time, cerium (III) oxide Ce_2O_3 was used as a dopant source with different concentrations from 1 to 5 mol%. According to the SSR method, the stoichiometric proportion of all the raw materials was homogeneously mixed and continuously grounded with the help of a mortar pestle. Wetting media such as acetone and distilled water were used for homogeneous mixing. The homogeneous mixture was transferred into the alumina crucible and calcined at 850°c for 8h in a muffle furnace, cooled at room temperature, and again grounded for 5 mins. The resulting product was then used for further characterization.

5.3 Structural characterization

The phase identification and crystal structure of the synthesized powder were evaluated by a PANalytical X'pert Powder X-ray Diffractometer with CuK_α radiation of λ= 1.5406 Å. To examine the crystallinity nature of synthesized samples the "Joint Committee of Powder Diffraction Standards (JCPDS)" of Powder Diffraction Files (PDF) was used. The X-ray diffraction (XRD) pattern of the $GdPO_4$:Ce sample was recorded using a scanning mode for the 2Θ range from 10 to 55° with a scanning step size of 0.02. The diffraction pattern of $GdPO_4$:Ce is shown in Figure 1, which indicates that a sharp intense peak was obtained and matched very well with the JCPDS card no. 84-0920. The crystal structure of prepared $GdPO_4$: Ce is identified as a monoclinic phase with space group P21/n(14) with cell parameters a = 6.65Å, b = 6.85Å, and c = 6.33Å. A small quantity of doping of Ce^{3+} does not affect the crystal structure of $GdPO_4$.

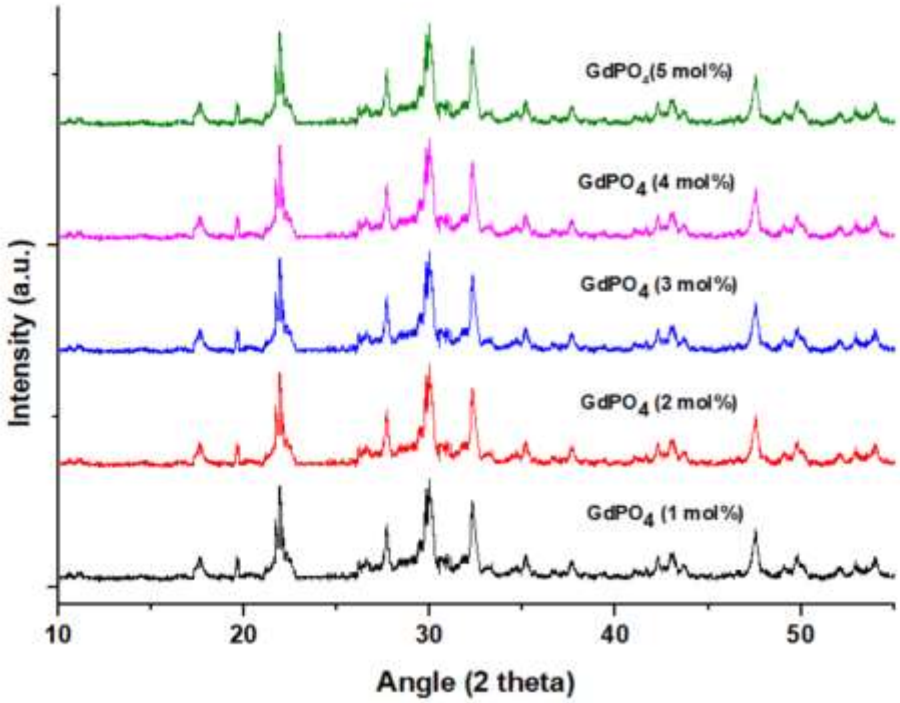

Figure 1 X-ray diffraction (XRD) pattern of $GdPO_4:Ce^{3+}$ (1-5mol%) phosphors.

5.4 Result and Discussion

5.4.1 TL glow curve of undoped and Ce doped $GdPO_4$ phosphor and effect of dopant concentration in phosphor

The thermoluminescence properties of undoped and cerium-doped $GdPO_4$ were investigated using a programmable Nucleonics TL Reader. Samples were irradiated with UV light for 25minute at 354 nm and subsequently heated at a rate of 5 °C/s [17–19]. At a wavelength of 354 nm, the prepared sample was irradiated through UV light for 5 min. Figures 2 and 3 show the TL glow curve of undoped and Ce-doped $GdPO_4$. The TL glow peak of undoped $GdPO_4$ is around 82 °C, and the TL intensity for Ce-doped $GdPO_4$ increases with an increase in temperature and attains a maximum peak for a temperature of 93 °C. After achieving the maximum value, it gradually starts to reduce with an increased dopant concentration, this is because of concentration quenching.

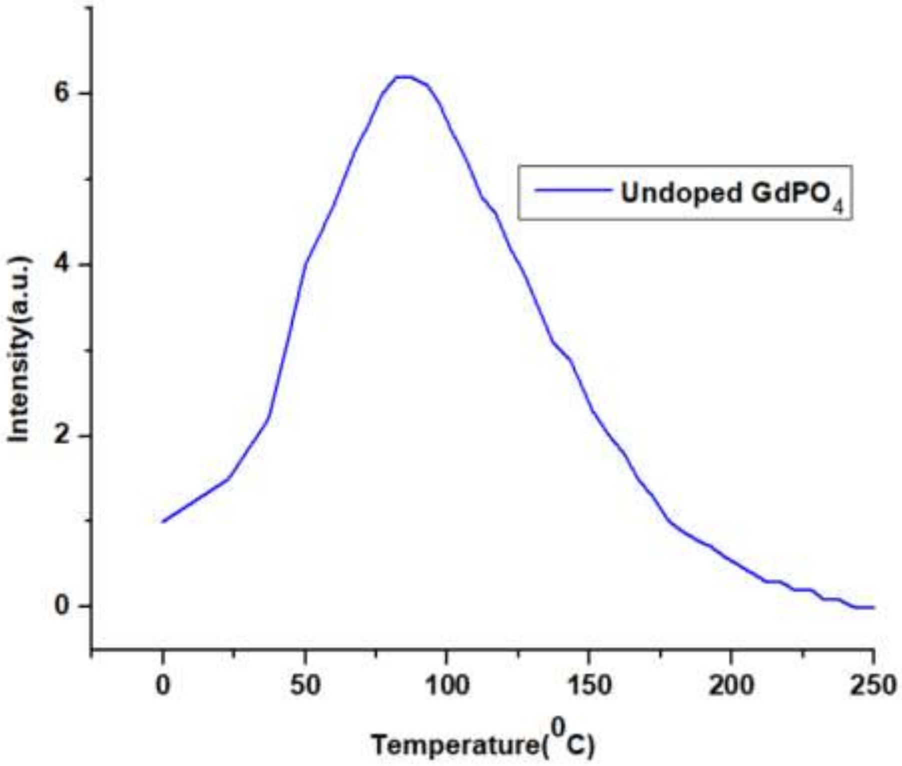

Figure 2 TL Glow curve of undoped $GdPO_4$.

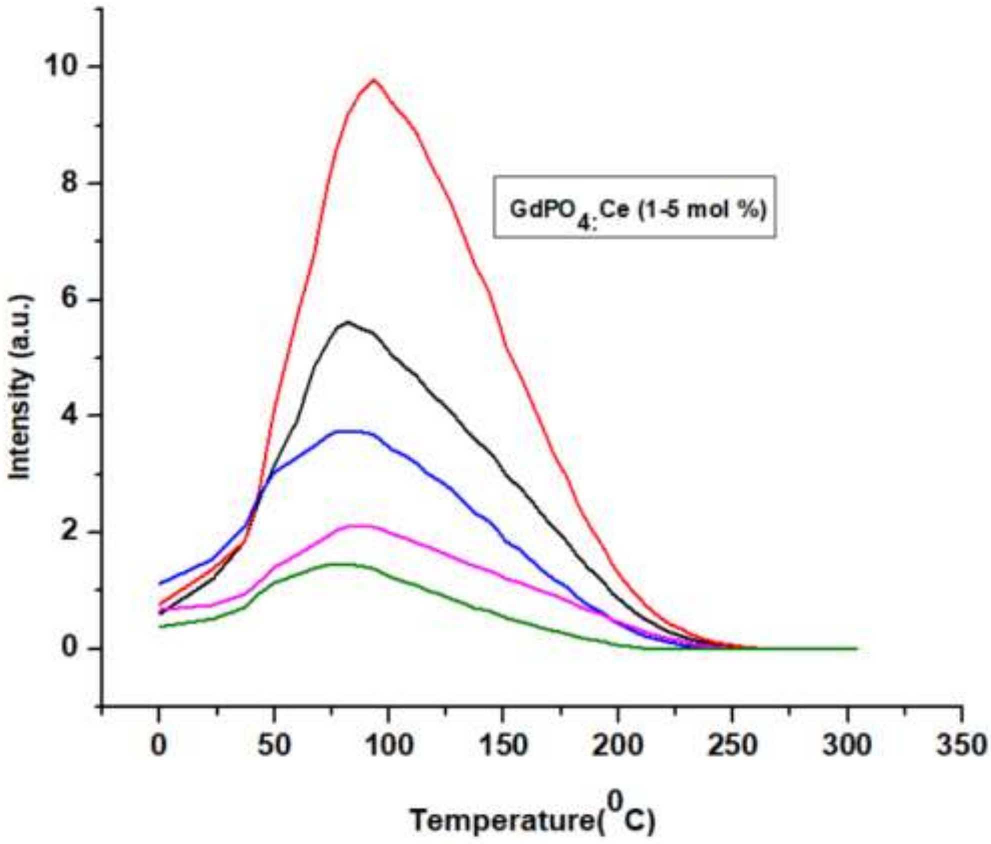

Figure 3 TL Glow curve of Ce doped $GdPO_4$ (1 – 5 mol %).

Similarly, Sharma examined the glow curve for undoped $GdPO_4$ shows a maximum peak at 365 K with an approximate trap depth (activation energy, E_a) of 0.78 eV calculated using a first-order kinetic equation, $25k_BT_m$, where k_B is Boltzmann's constant and T_m is the glow curve maximum. This glow curve maximum at 365 K is comparable to that detected for $DyPO_4$, $SmPO_4$, and $EuPO_4$ samples [18]. Thus, it is indicated that the origin of this glow curve maximum is an intrinsic defect that is created due to the host lattice and is also independent of the choice of the rare earth ion.

Gasiorowski et al also studied the influence of particle size of Gd_2O_3 and Dy_2O_3 doped phosphate glasses on the intensity of thermoluminescence (TL) signals. The obtained data and studied presented that the phosphate glasses show good thermoluminescence properties for high-energy beta radiation dose measurement applications [20].

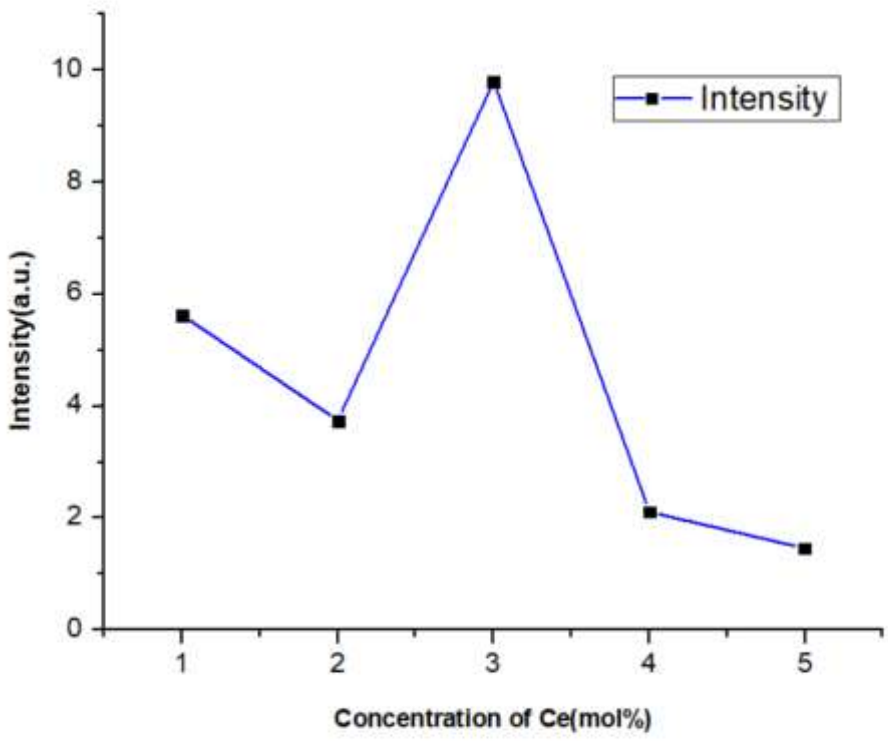

Figure 4 TL intensity variation with a varied dopant concentration of Ce

Figure 4 shows the TL intensity variation with Ce ion concentration. The TL intensity reaches its optimum condition for a 3 mol% dopant concentration of Ce, after that it decreases due to concentration quenching. Hence, 3mol% Ce might be the optimum dopant concentration for maximum TL intensity. Concentration mainly affects the TL intensity and not its shape. Since the sample does not show any remarkable variation with UV dose, hence it is not a suitable candidate for radiation dosimetry applications.

5.5Conclusion

$GdPO_4:Ce^{3+}$ phosphors have been successfully synthesized by solid-state reaction method with a varying concentration ranging from 1 to 5 mol%. The XRD pattern confirmed that these phosphors enclose a monoclinic phase with space group $P_{21/n}$(14) with cell parameters a = 6.65Å, b = 6.85Å, and c = 6.33Å. The TL glow curve of the synthesized undoped sample showed a single broad peak centered at 82 ^{0}C. The TL glow curve of the synthesized Ce doped sample showed a single broad peak centered at 93 ^{0}C. The TL intensity for different concentrations of Ce did not show much variation. The optimum TL intensity was observed for samples with 3 mol% of Ce. With further increase in doping concentration, concentration quenching occurs. No remarkable variation in the TL intensity with different UV doses was recorded. As the sample does not show any remarkable variation with UV dose, hence it is not suitable for radiation dosimetry applications.

5.8 References

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