



Short note

Efficient energy transfer and fluorescence in $\text{SrYAl}_3\text{O}_7:\text{Ce}^{3+}, \text{Tb}^{3+}$ phosphorM.S. Mendhe^{a,*}, S.P. Puppallwar^{a,*}, S.J. Dhoble^b^a Department of Physics, Kamla Nehru Mahavidyalaya, Nagpur 440009, India^b Department of Physics, R.T.M. Nagpur University, Nagpur 440033, India

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ABSTRACT

A green light emitting phosphor $\text{SrYAl}_3\text{O}_7:\text{Ce}^{3+}, \text{Tb}^{3+}$ have been synthesized by fast solution combustion route. Structural and photoluminescence properties with energy transfer (ET) mechanism were investigated. Due to the ET from Ce^{3+} to Tb^{3+} , enhancement in the emission intensity of Tb^{3+} is observed. The prepared phosphors exhibit both the weak emission of Ce^{3+} ($5d-4f$) and the strong emission of Tb^{3+} ($^5\text{D}_4-^7\text{F}_j$) with considerable emission intensity. The ET mechanism from Ce^{3+} to Tb^{3+} ion has been determined to be dipole–dipole interaction, and the ET efficiency is obtained over 81%. The effect of Ce^{3+} and Tb^{3+} concentrations on luminescence intensity in single and co-doped phosphors are also studied. The phosphor has remarkable CIE chromaticity coordinates of (0.198, 0.532), which indicate that Ce^{3+} and Tb^{3+} co-doped SrYAl_3O_7 phosphor may be potential UV-convertible candidate with green light emitting for w-LEDs.

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1. Introduction

White light-emitting diodes have been expected as the next generation solid-state lighting sources to replace the incandescent and fluorescent lamps because of their numerous superior advantages such as high efficiency, long lifetime, reliability, small size, good light stability and environmental friendly [1–5]. One of promising approaches to prepare w-LEDs is by pumping blue/green/red tricolor phosphors with a near-ultra violet (360–410 nm) InGaN-based LED as the excitation source in order to obtain excellent color rendering properties. For this reason, it is necessary to develop novel phosphor having the multi-color emission in the field of optical materials [6–9].

It has been widely recognized that ET is a prominent way to develop desired phosphors for w-LEDs. In order to generate white light, co-doping with different RE ions into a proper host is one common strategy to control the emission color via ET processes. Usually, Tb^{3+} ion is used as a significant activator for luminescent materials. Generally, it shows strong green emission at high doping concentration and blue emission at low doping contents due to transitions of $^5\text{D}_4 \rightarrow ^7\text{F}_j$ and $^5\text{D}_3 \rightarrow ^7\text{F}_j$ ($j=6, 5, 4, 3, 2$ and 1), respectively [10–12]. This shows Tb^{3+} emission depends strongly on its concentration. Unfortunately, the absorption peaks of Tb^{3+} are weak and too narrow because those $4f \rightarrow 4f$ transitions are strictly forbidden by the parity selection rule, due to this incompatibility to the tiny emission wavelength shift of LED chips and the poor emission intensity. In order to overcome this problem, Ce^{3+} can be co-doped as a sensitizer. Ce^{3+} ions usually can be strongly excited by ultraviolet irradiation in most oxides due to the strong $4f \rightarrow 5d$ transitions and transfer the harvesting photons to other activators ion. In a proper host, the chance is high indeed that the occurrence of efficient ET from Ce^{3+} to Tb^{3+} when the

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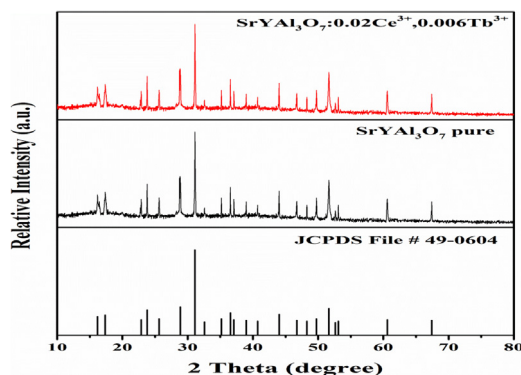


Fig. 1. Powder XRD pattern of prepared samples (a) SrYAl_3O_7 pure and (b) $\text{SrYAl}_3\text{O}_7:0.02\text{Ce}^{3+},0.006\text{Tb}^{3+}$ phosphor along with standard JCPDS. 49-0604.

emission of Ce^{3+} overlaps with the excitation of Tb^{3+} , such as $\text{KCl}:\text{Ce}^{3+},\text{Tb}^{3+}$ [13], $\text{K}_3\text{Gd}(\text{PO}_4):\text{Ce}^{3+},\text{Tb}^{3+}$ [14], $\text{Al}_2\text{O}_6\text{N}:\text{Ce}^{3+},\text{Tb}^{3+}$ [15], $\text{SrMgSi}_2\text{O}_6:\text{Ce},\text{Tb}$ [16]. As a promising sensitizer for Tb^{3+} ions, Ce^{3+} has been widely used in many hosts [17,18].

In last several decade, significant efforts have been devoted to prepare and investigate an important inorganic materials family having general chemical formula ABC_3O_7 , ($\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$; $\text{B} = \text{La}, \text{Gd}, \text{Y}$; $\text{C} = \text{Al}, \text{Ga}$) as these materials have been widely applied in plasma display panels (PDP) for high definition TV (HDTV), all-solid-state lasers, diode laser pumping and tunable laser generation, and white LEDs [19,20]. As one kind of aluminates, SrYAl_3O_7 was selected as a host material in this work. There is not much study on SrYAl_3O_7 host though compounds with similar formula and structure have shows interesting properties. A fast and simple combustion method has been employed to synthesize this material. $\text{CaYAl}_3\text{O}_7:\text{Ce}^{3+},\text{Tb}^{3+}$ [21], $\text{CaYAl}_3\text{O}_7:\text{Eu}^{3+}$ [22], $\text{Er}^{3+}/\text{Yb}^{3+}$ co-doped CaYAl_3O_7 [23], $\text{GdCaAl}_3\text{O}_7:\text{Eu}^{3+}$ [24] and $\text{LaCaAl}_3\text{O}_7$ doped with variety of ns^2 and rare earth activators [25] are prepared by this method and studied earlier. To the best of our knowledge, there is no study about the luminescent properties and ET of Ce^{3+} and Tb^{3+} co-doped SrYAl_3O_7 . In this study, we report luminescent property and ET of $\text{SrYAl}_3\text{O}_7:\text{Ce}^{3+},\text{Tb}^{3+}$ phosphor. The results show that the $\text{Ce}^{3+}-\text{Tb}^{3+}$ is successfully induced in the host by the ET processes from Ce^{3+} to Tb^{3+} .

2. Experimental

$\text{SrY}_{1-x-y}\text{Al}_3\text{O}_7:x\text{Ce}^{3+},y\text{Tb}^{3+}$ phosphors were prepared by a solution combustion method. In a typical preparation, Stoichiometric amounts of $\text{Y}(\text{NO}_3)_3$, $\text{Sr}(\text{NO}_3)_2$, $\text{Al}(\text{NO}_3)_3 \cdot 9(\text{H}_2\text{O})$, $(\text{NH}_4)_2\text{Ce}(\text{NO}_3)_6$, $\text{Tb}(\text{NO}_3)_3 \cdot 6(\text{H}_2\text{O})$ and urea (CON_2H_4) were dissolved in distilled water. A homogeneous solution was obtained after the mixture stirred vigorously for 15–20 min in a glass beaker. The solution was transferred in to a muffle furnace maintained at a temperature of $500 \pm 20^\circ\text{C}$. The reagents decomposed and released large amounts of gases during the evaporation of liquid. Due to the exothermic nature of combustion process a large amount of heat was released with an enormous flame that decomposed the reagents further and released more gases. After 1 min flame damped and the combustion reaction is completed within 5 min. The resulting materials were cooled down to room temperature and ground gently using a pestle and mortar. The obtained powders were calcite at 900°C for 2 h. It was cooled to room temperature and crushed to fine powder and then used for further characterization.

Phase and crystallinity of the prepared materials are checked by a powder XRD pattern using PAN-analytical diffractometer with $\text{Cu K}\alpha$ radiation ($\lambda = 1.5405 \text{ \AA}$) operating at 40 kV, 30 mA in the 2θ range $10-80^\circ$. Particle morphology was studied using SEM at VNIT, Nagpur India, (JSM-6360LV, JEOL, USA). The photoluminescence excitation and emission spectra of the samples were recorded using a SHIMADZU Spectrofluorophotometer (RF-5301 PC). All the measurements are performed at room temperature.

3. Results and discussion

3.1. XRD phase analysis

The XRD patterns of undoped SrYAl_3O_7 and $\text{SrYAl}_3\text{O}_7:0.02\text{Ce}^{3+},0.006\text{Tb}^{3+}$ phosphors calcined at 900°C for 2 h are shown in Fig. 1. It can be clearly observed that all the diffraction peaks are almost in good agreement with the standard data of the SrYAl_3O_7 (JCPDS No. 49-0604). It indicates that the obtained samples are single phase and of same structure as SrYAl_3O_7 . No impurity peaks were observed in Fig. 1, which means that the dopant ions (Ce^{3+} , Tb^{3+}) were completely dissolved in the host lattice. Both of them can be indexed to the tetragonal structure of SrYAl_3O_7 with space group of $\text{P4}_2\text{m}$ and with lattice constants $a = b = 0.7768 \text{ nm}$, and $c = 0.5099 \text{ nm}$. The particles size was calculated using Scherrer's formula and it is approximately found to be in the range of 50–60 nm. Based on the effective ionic radius(r) of cations with different values of coordination number (CN) reported by Shannon [26], the radii of Ce^{3+} (1.01 Å) and Tb^{3+} (0.923 Å) are more nearer to those

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