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Photoluminescence and structural properties of Eu^{3+} doped SrZnV_2O_7 nanocrystals



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ABSTRACT

Novel $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$ nanoparticles were synthesized employing urea assisted solution combustion process. The Rietveld refinement technique was used to study the effect of Eu^{3+} doping on the crystal structure of the host lattice. It was found to crystallize in monoclinic lattice with the $P12_1/c1$ (14) space group. The photoluminescent spectral analysis showed that upon excitation in the near UV region, these nanophosphors can emit very intense red luminescence (616 nm) corresponding to the hypersensitive $^5\text{D}_0 \rightarrow ^7\text{F}_2$ transition of Eu^{3+} ions. X-ray diffraction studies revealed the low symmetrical co-ordinative environment of the activator ion (trigonal bipyramidal) with no inversion center in the SrZnV_2O_7 host lattice. The optimum concentration of Eu^{3+} ion in SrZnV_2O_7 for better luminescence was found to be 4 mol%. The critical distance for energy transfer (24.1602 Å) imparted the initial idea of intensity variation as a function of dopant concentration. Dipole–dipole (d–d) interactions were successfully accounted for the concentration quenching arising from the over-doping of the activator ions. These near UV light absorbers (395 nm) can be thrivaly used in white LEDs using InGaN chip system, plasma display panels (PDPs) and solid state laser.

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1. Introduction

In the present era, nano-scaled phosphor materials are attracting a lot of research efforts because of their potential applications in a number of devices like light-emitting diodes (LEDs), luminescent solar collectors, biological imaging and scintillation detectors [1–6]. In addition, sharp spectral bands resulting from the 4f electrons intra-configurational transitions induce a special interest to Eu^{3+} doping in inorganic hosts lattices [7]. Various inorganic host materials have been explored to prepare high quality rare earth doped nanophosphors but mixed metal double pyrovanadates with general formula MZnV_2O_7 ($\text{M} = \text{Ba}, \text{Sr}, \text{Ca}$) are quite novel to the research field as far as photoluminescent properties are concerned. The importance of vanadium oxides in material chemistry is due to the diverse structural behavior of the vanadium–oxygen unit. In pyrovanadates, anionic $\text{V}_2\text{O}_4^{4-}$ complex ions are isolated units where both vanadium atoms are corner shared [8–10]. The host, SrZnV_2O_7 exists in a single monoclinic crystallographic form. The standard SrZnV_2O_7 has $P2_1/n$ (14) space group with lattice constant a (Å) = 7.4115, b (Å) = 6.6895, c (Å) = 11.9610 and unit cell volume equal to 589.73 Å³ [11,12].

Several experimental techniques, such as solid state reaction [13], hydrothermal synthesis [14], sol–gel processing [15] and combustion synthesis [16], have been used to fabricate vanadates of high purity and desired crystallinity. In the present work, we have employed the urea assisted solution combustion route to synthesis $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$ nanophosphors. This efficient phosphor processing method has a number of advantages; such as inexpensive raw materials, low synthesis temperature, a relatively simple preparation process and homogenous product with fine particle size [17]. This work is executed in order to investigate the photoluminescence and crystal structure of the single-phased, red emissive $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$ nanophosphor, synthesized via the solution combustion method. The optical and structural characterization not only reveals the variation of luminescence intensity as a function of dopant concentration or temperature but also explains the mechanism behind the non-radiative energy transfer responsible for the concentration quenching arising after the optimal concentration (4 mol%). In addition to the results from Scherrer's equation, the claim of particle size in the nano range was also supported by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The photoluminescent excitation spectra of these nanophosphor shows very strong absorption at 395 nm due to $\text{F}_0 \rightarrow ^5\text{L}_6$ transition and thus claim their promising potential in high efficiency white light emitting diodes (W-LEDs) using near-ultraviolet (NUV) InGaN based LED

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chip coated with red/green/blue tricolor phosphors [18–20]. Furthermore, these nanophosphor finds plausible use in alternating current plasma display panels (AC-PDPs) operating under near ultraviolet and Eu^{3+} doped vanadate based solid state laser [21,22].

2. Experimental

2.1. Material and synthesis

A series of rare earth doped $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$ ($x=0.01-0.06$) nanophosphors were synthesized using the solution combustion method in which constituent raw materials were high purity $\text{Sr}(\text{NO}_3)_2$, $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Eu}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, NH_4VO_3 and urea. The starting materials were weighed in stoichiometric proportion and dissolved in a minimum quantity of deionized water in a 400 ml capacity pyrex beaker. The amount of the urea was calculated using the total oxidizing and reducing valencies of the oxidizer and the fuel [23]. Finally the solution was placed into a preheated furnace maintained at 500 °C. The starting material undergoes rapid dehydration and foaming followed by decomposition, producing combustible gases. These volatile combustible gases ignite and burn with a flame yielding a voluminous solid. Urea was oxidized by nitrate ions and served as a fuel for propellant reaction. The products were then cooled to room temperature and pulverized. A fraction of all the products were calcined at 600 °C and 700 °C for 3 h and kept in desiccator for further characterization.

2.2. Materials characterization

The crystal phases of $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$, as-prepared and calcined at different temperatures were characterized using a high resolution Rigaku Ultima-IV X-ray powder (XRD) diffractometer fitted with a dual position graphite monochromator in the diffracted beam. The diffraction pattern was recorded from $2\theta=10^\circ-80^\circ$ at a scanning speed of 2°min^{-1} using $\text{Cu K}\alpha$ radiation at 40 kV tube voltage and 40 mA tube current. The qualitative and quantitative phase analysis of samples as prepared, calcined at 600 °C and calcined at 700 °C was also carried out using Rietveld refinement technique by MAUD program for Rietveld refinement

[24–27]. The particle size was calculated by the Scherrer equation using the X-ray diffraction pattern.

The surface morphological properties were studied using a Jeol JSM-6510 scanning electron microscope (SEM) and a Tecnai G² FEI transmission electron microscope (TEM). In order to obtain the clear SEM micrographs, secondary electron imaging (SEI) was preferred over the backscattered electron imaging (BSEI) due to its higher surface sensitivity and greater resolution. TEM images were obtained using the electron beam accelerated at 80 kV with a magnification of 10^5 . SEM and TEM analysis was also used to confirm the particle size in the nano range as given by the X-ray diffraction studies.

The Fourier transform infrared (FT-IR) spectra were recorded in the transmission mode using a Perkin-Elmer FT-IR/RZX spectrometer in the spectral range of $4000-400\text{ cm}^{-1}$ following the KBr pellet technique with a resolution of 0.5 cm^{-1} . The background correction has been made in the FT-IR experiment. The complete FT-IR spectrum is consisted of 64 scans.

The photoluminescence excitation and emission spectra (scanning rate: 1200 nm min^{-1} , PMT voltage: 400 V, widths of the excitation slit and emission slit: 5.0 nm) and fluorescence decay curve and color co-ordinates of the $\text{SrZn}_{1-x}\text{Eu}_x\text{V}_2\text{O}_7$ nanocrystals in the UV-vis region were recorded and investigated at room temperature using a Hitachi F-7000 fluorescence spectrophotometer with Xe-lamp as the excitation source.

3. Results and discussion

3.1. Crystal structure

Phase identification and crystal structure analysis of the sample as-prepared, calcined at 600 °C and 700 °C were carried out using the Rietveld refinement technique over the experimental data of X-ray diffraction profiles. Fig. 1 depicts that the as-prepared sample is a mixture of two phases $\text{Sr}(\text{NO}_3)_2$ (54.79%) (JCPDS Card no. 76-1375) and $\text{Zn}_2\text{V}_2\text{O}_7$ (45.19%) (JCPDS Card no.=29-1396) [28,29]. The blue dot line is the experimental pattern observed for the sample as prepared at 500 °C and solid red and green lines are the simulated diffraction data from $\text{Sr}(\text{NO}_3)_2$ and $\text{Zn}_2\text{V}_2\text{O}_7$ respectively. The qualitative and quantitative phase analysis of the sample calcined

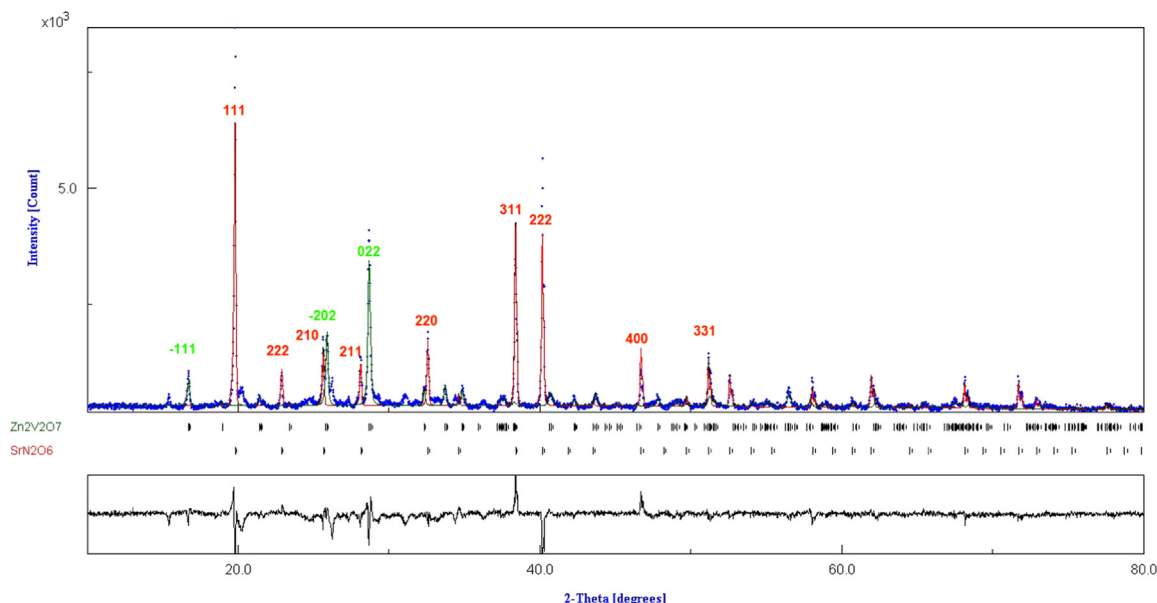


Fig. 1. Rietveld refinement of $\text{Sr}(\text{NO}_3)_2 + \text{Zn}_2\text{V}_2\text{O}_7$ with sample as-prepared. $\chi_r=3.12$, $R_{\text{wp}}(\%)=15.80$, $R_p(\%)=12.26$, $R_{\text{exp}}(\%)=5.06$, SrNO_6 weight % = 54.79, $\text{Zn}_2\text{V}_2\text{O}_7$ weight % = 45.19. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

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