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SYNTHESIS AND PL STUDY OF $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ PHOSPHOR FOR W-LED APPLICATION

P. A. Nagpure[✉], *N. D. Kherde*, *W. S. Barde*

Shri Shivaji Science College, Amravati, Maharashtra, India

[✉] nagpurepa@yahoo.co.in

Abstract. In the present paper, a single host phosphor for a white light emitting diode (W-LED) $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$, prepared by the co-precipitation method and combustion one, has been reported. Formation of the compound was confirmed by studying the X-ray diffraction pattern. The photoluminescence (PL) properties were studied by fluorescence spectrophotometer F-7000. The $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ exhibited a broad emission band covering the entire visible region centered at 520 nm wavelength and a sharp peak at 613 nm, when excited by 350 nm. The excitation spectrum at 520 nm emission wavelength was found to be suitable for pc-W-LED application.

Keywords: strontium vanadate, co-precipitation, combustion, X-ray diffraction pattern, photoluminescence spectroscopy, W-LED

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СИНТЕЗ И ФОТОЛЮМИНЕСЦЕНТНОЕ ИССЛЕДОВАНИЕ ЛЮМИНОФОРА $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ С ЦЕЛЬЮ ПРИМЕНЕНИЯ В БЕЛЫХ СВЕТОИЗЛУЧАЮЩИХ ДИОДАХ

П. А. Нагпуре[✉], *Н. Д. Херде*, *В. С. Барде*

Научный колледж Шри Шиваджи, г. Амравати, штат Махараштра, Индия

[✉] nagpurepa@yahoo.co.in

Аннотация. В статье исследован люминофор $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$, перспективный для создания белого светоизлучающего диода (W-LED) на основе этого одного соединения. Образцы данного материала были получены двумя способами: соосаждения и сжигания. Образование требуемого вещества было подтверждено рентгенографически. Практически важные свойства объекта изучались методом фотолюминесцентной спектроскопии. В спектрах соединения обнаружены широкая эмиссионная полоса, охватывающая всю видимую область спектра с центром на длине волны 520 нм, а также острый пик на 613 нм. В спектрах возбуждения люминесценции наблюдалась широкая полоса с центром на длине волны 350 нм. Представлена соответствующая диаграмма энергетических уровней. Полученные экспериментальные результаты привели к заключению, что метод соосаждения предпочтителен, а фотолюминесцентные свойства полученного люминофора удобны для создания штучных белых светоизлучающих диодов.

Ключевые слова: ванадат стронция, соосаждение, метод сжигания, фотолюминесценция, белый светоизлучающий диод

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Introduction

White light emitting diodes (W-LEDs) have been found wide use especially for solid state lighting and backlight of liquid crystal display (LCD), traffic signals. W-LEDs have a number of advantages such as long lifetime and lack of pollutant, higher energy efficiency, compactness and reduced power consumption [1]. In a W-LED, white light can be produced either by combining a blue LED with a yellow phosphor or combining the ultraviolet light emitting diodes (UV-LED) with a blend of blue, green and red phosphors. White light emission resulted from a single-phase phosphor compared to combination of two or three phosphors expected to give high luminous efficiency, because it reduces the probability of multiphosphor reabsorption of emission colors [2].

In recent years, vanadates have been used in many high-technology fields, such as biological materials, electrochemistry, optical lasers, catalysis, etc. [3] Luminescence materials have been widely studied and used in white light emitting diodes and flat-panel displays (FPDs), such as $\text{YVO}_4:\text{Eu}^{3+}$, etc. [4]. At ambient pressure orthovanadates exhibit oxygen ionic conductivity and high electronic conductivity, i. e., the migration of electrons between V^{4-} and V^{5-} centers, but electronic conduction is absent in isostructural orthophosphates and orthoarsenates [5]. Vanadates with general formula $\text{M}_3(\text{VO}_4)_2$ ($\text{M} = \text{Ca}, \text{Sr}, \text{Ba}$) have been proved to be good candidates for the luminescent hosts, because they have strong absorption band in the UV region and then transfer the energy to rare-earth activator ions efficiently due to good match of energy levels in the wide region and strong interaction between states of $[\text{VO}_4]^{3-}$ and rare-earth ions. In addition to this, vanadates have the self-activated emitting properties and better chromaticity [6]. $[\text{VO}_4]^{3-}$ group consists of the central metal vanadium ion coordinated by four oxygen ligands in the tetrahedral symmetry and is known to be an efficient luminescent center with broad band emissions from 400 to more than 700 nm with UV or near-UV light excitation [7]. Vanadium oxides have different complex groups of ortho-, meta- and pyrovanadates. Vanadates generally show a short wavelength of the optical absorption edge. This makes them suitable as hosts to accommodate active rare-earth ions [8]. The ytterbium ions Yb^{3+} doped with $\text{Sr}_3(\text{VO}_4)_2$ provide characteristic near-infrared (NIR) emission in the range of 970 – 1050 nm which are suitable to get rid of the spectral mismatch problem of c-Si solar cells [9].

In the present work, photoluminescence of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor synthesized by co-precipitation and combustion methods has been studied in the context of its application as W-LED phosphor.

Experimental part

Strontium nitrate $\text{Sr}(\text{NO}_3)_2$ (99.9% A.R.), europium oxide Eu_2O_3 (99.9% A.R.) and ammonium vanadate NH_4VO_4 (99.9% A.R.), all from Sigma Aldrich, were used.

Synthesis. The synthesis of phosphor $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$, was attempted using two methods: co-precipitation one and combustion one.

Co-precipitation method. The phosphors $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$ ($x = 0.002, 0.005, 0.010$) were synthesized by co-precipitation method (Fig. 1). The precursors $\text{Sr}(\text{NO}_3)_2$ (99.9% A.R.), Eu_2O_3 (99.9% A.R.) and NH_4VO_4 (99.9% A.R.) were weighed in proper stoichiometric ratio and used for synthesis of the phosphor. The starting chemicals NH_4VO_4 (99.9% A.R.) were dissolved in the double distilled water and heated on a hot plate at about 100°C . The nitric acid HNO_3 was added drop by drop in Eu_2O_3 and a DD water mixture in separate beaker was so as to convert it into

$\text{Eu}(\text{NO}_3)_3$. The hot solution of NH_4VO_4 then added drop wise to the aqueous transparent mixture of nitrates with constant stirring. The entire homogenous solution was then placed on the hot plate maintained at a temperature of 90°C for slow evaporation of water. The dried precipitate was finally crushed and heated at 800°C for 1 hour and 950°C for 2 hours in order to get white crystalline powder of $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$ ($x = 0.002, 0.005, 0.010$).

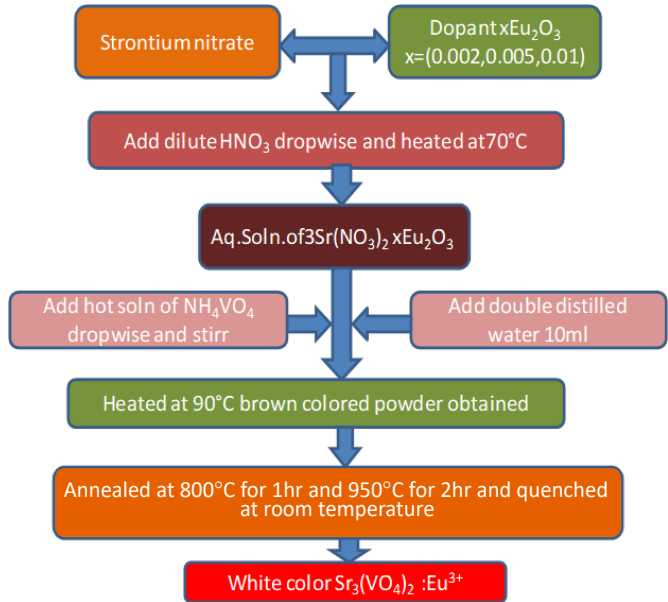


Fig. 1. Flow chart of synthesis of $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$ via co-precipitation method

Combustion method. The stoichiometric amount of precursors, namely, strontium nitrate $\text{Sr}(\text{NO}_3)_2$, ammonium vanadate NH_4VO_3 , urea and Eu_2O_3 converted to $\text{Eu}(\text{NO}_3)_3$ ($x = 0.002, 0.005, 0.010$) were taken in a china clay basin and few drops of DD water added to the mixture. This mixture was heated at 90°C till the precursors dissolved completely, then it was kept into the preheated furnace at 900°C , and then warmed for 5 min. The self-heat generating redox reaction was completed in 2 min and the fine powder of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ was obtained (Fig. 2). This raw powder was sintered for 2 hours at 950°C and quenched to room temperature.

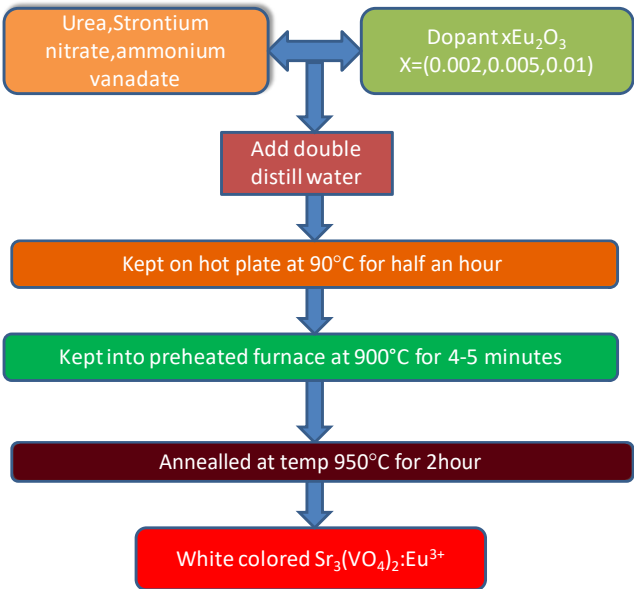


Fig. 2. Flow chart of synthesis of $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$ via combustion method

Characterizations. Formation of the required phase of the compound was confirmed by the X-ray diffraction pattern. The photoluminescence (PL) properties were studied using Hitachi (F-7000) fluorescence spectrophotometer.

Results and discussion

X-ray diffraction (XRD) analysis. The formation of $\text{Sr}_3(\text{VO}_4)_2$ host in the crystalline phase synthesized by both co-precipitation and combustion methods was confirmed by XRD pattern taken from Rigaku miniflex II X-ray diffractometer with scan rate of $2.000^\circ/\text{min}$ and CuK_α ($k = 0.15406 \text{ nm}$) radiation in the range from 10° to 90° (see Fig. 3). The XRD pattern for $\text{Sr}_3(\text{VO}_4)_2$ agreed with the ICDD file (01-081-1844) very well.

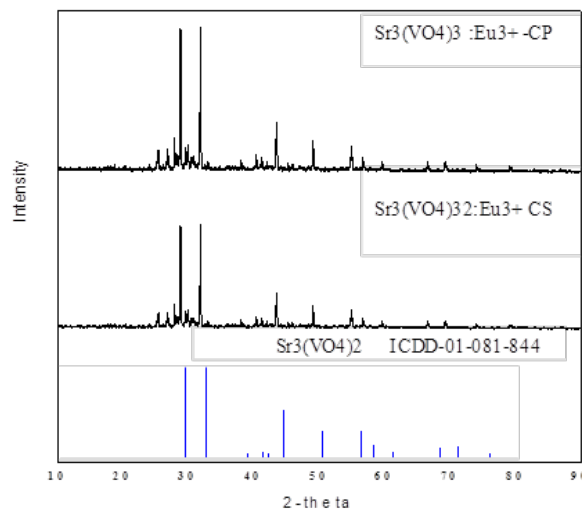


Fig. 3. X-ray diffraction pattern of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$

Photoluminescence characterization. The photoluminescence (PL) emission and excitation spectrum of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor synthesized by co-precipitation and combustion methods are shown in Fig. 4. For the both cases the nature of emission spectra are almost the same but there is a slight difference in the excitation spectra. The emission spectrum exhibits a green broad band centered at 520 nm which is attributed to ${}^3T_{1,2} \rightarrow {}^1A_1$ transition of $[\text{VO}_4]^{3-}$ group, and with a sharp peak at 613 nm in the red region corresponding to a characteristic ${}^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} [10, 11].

The PL emission spectra of the phosphor shows maximum intensity for 0.002 mol doping of Eu when synthesized by the co-precipitation method and for 0.001 mol doping of Eu when synthesized by the combustion one. The overall intensity of PL emission is greater for the phosphor synthesized by the co-precipitation method than that for the phosphor synthesized by the combustion one.

Fig. 5. presents the simplified energy level diagrams of $[\text{VO}_4]^{3-}$ group and Eu^{3+} ion and emission transition processes in the $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor as well.

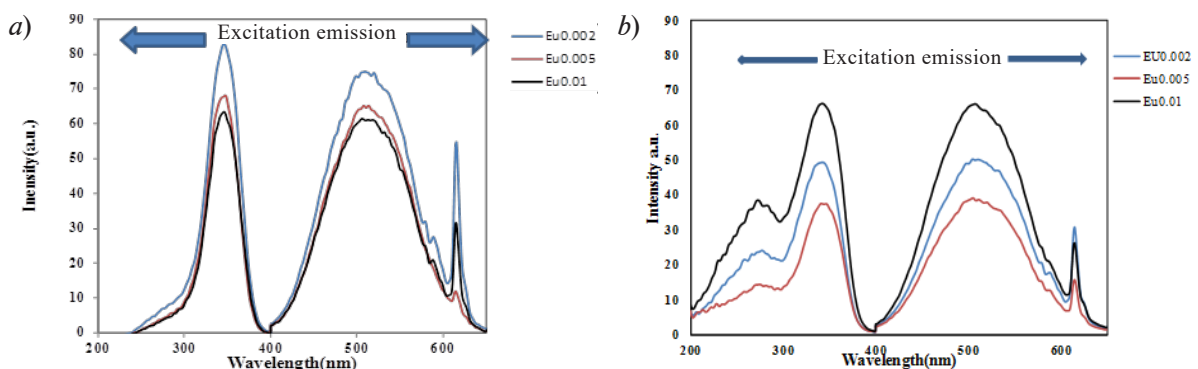


Fig. 4. Photoluminescence excitation and emission spectra of $\text{Sr}_{3-x}(\text{VO}_4)_2:(\text{Eu}^{3+})_x$ synthesized by co-precipitation (a) and combustion (b) methods

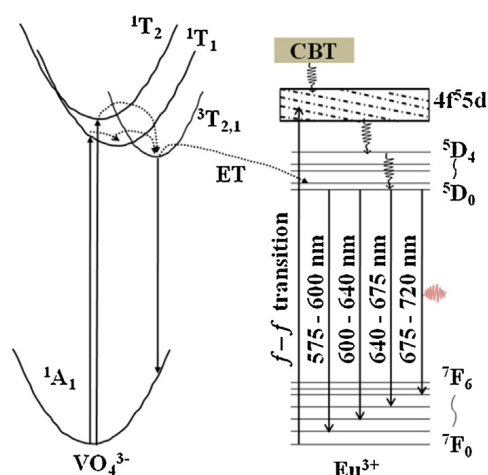


Fig. 5. The simplified energy level diagrams of $[\text{VO}_4]^{3-}$ group and Eu^{3+} ion and emission transition (ET) processes in the $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor

Summary

A series of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphors were successfully synthesized by co-precipitation and combustion methods. The XRD patterns confirmed the pure crystalline phase of $\text{Sr}_3(\text{VO}_4)_2$. The photoluminescence excitation spectra showed the broad excitation band with maximum at 350 nm. The photoluminescence emission spectra showed the broad band (400 – 650 nm) with a maximum at 520 nm attributed to the ${}^3T_{1,2} \rightarrow {}^1A_1$ transitions of $[\text{VO}_4]^{3-}$ group and a sharp peak at 613 nm attributed to the ${}^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} . The co-precipitation method was found to be suitable and efficient for synthesis of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$.

The photoluminescence spectra of the $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ exhibited a potential application of the prepared phosphor in energy-efficient solid-state lighting, optoelectronic devices and organic composite solar cells.

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THE AUTHORS

NAGPURE Pankaj A.

Shri Shivaji Science College, Amravati
Shivaji Nagar, Morshi Road, Amravati, 444603, India
nagpurepa@yahoo.co.in
ORCID: 0000-0001-6767-1907

KHERDE Nivedita D.

Shri Shivaji Science College, Amravati
Shivaji Nagar, Morshi Road, Amravati, 444603, India
niveditaholey@gmail.com

BARDE Waman S.

Shri Shivaji Science College, Amravati
Shivaji Nagar, Morshi Road, Amravati, 444603, India
wamanbarde81@gmail.com

**СВЕДЕНИЯ ОБ АВТОРАХ**

НАГПУРЕ Панкадж А. – *Ph.D.*, доцент кафедры физики Научного колледжа Шри Шиваджи, г. Амравати, штат Махараштра, Индия.

Shivaji Nagar, Morshi Road, Amravati, 444603, India
nagpurepa@yahoo.co.in
ORCID: 0000-0001-6767-1907

ХЕРДЕ Ниведита Д. – *сотрудник кафедры физики Научного колледжа Шри Шиваджи, г. Амравати, штат Махараштра, Индия.*

Shivaji Nagar, Morshi Road, Amravati, 444603, India
niveditaholey@gmail.com

БАРДЕ Ваман С. – *Ph.D.*, адъюнкт-профессор кафедры физики Научного колледжа Шри Шиваджи, г. Амравати, штат Махараштра, Индия.

Shivaji Nagar, Morshi Road, Amravati, 444603, India
wamanbarde81@gmail.com

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