



Synthesis and luminescence properties of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor and emission enhancement by co-doping Li^+ ion

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ABSTRACT

$\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ and $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$, Li^+ phosphors are synthesized by solid-state reaction method in air. X-ray diffraction patterns indicate that all samples are pure phase $\text{Sr}_3(\text{VO}_4)_2$. The host $\text{Sr}_3(\text{VO}_4)_2$ with excitation 348 nm emits yellow-green light. Emission spectrum of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor with excitation 335 nm contains a broad emission band peaking at ~ 540 nm due to $\text{V}^{5+} \rightarrow \text{O}^{2-}$ charge transfer and some narrow emission bands owing to the $4f - 4f$ electron transitions of Eu^{3+} ion in the range of 400–780 nm. With excitation 394 nm, only emission bands derived from Eu^{3+} ion are observed. Emission intensity of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor can be improved obviously due to charge compensation and fluxing agent role of Li^+ ion when Li^+ ion is co-doped. The optimal Eu^{3+} doping concentration is ~ 5 mol%. The energy transfer process between $[\text{VO}_4]^{3-}$ group and Eu^{3+} ion is observed and analyzed by emission spectrum in $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor. The luminous mechanism of $\text{Sr}_3(\text{VO}_4)_2:\text{Eu}^{3+}$ phosphor is explained by energy transfer and energy level diagram of $[\text{VO}_4]^{3-}$ group and Eu^{3+} ion.

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1. Introduction

Vanadates have been applied in many high-technology fields, such as electrochemistry, optical lasers, catalysis, biological materials [1–3]. Vanadate luminescence materials have been widely studied and used in white light emitting diodes (LEDs) and flat-panel displays (FPDs), such as $\text{YVO}_4:\text{Eu}^{3+}$ [4,5]. Recently, rare-earth (RE) ion doped vanadate phosphors have attracted more attentions due to energy transfer (ET) from $[\text{VO}_4]^{3-}$ group to RE ions, the self-activated emitting properties of $[\text{VO}_4]^{3-}$ group, and better chromaticity [6]. $[\text{VO}_4]^{3-}$ group is composed of the central metal ion V coordinated by four oxygen ligands in a tetrahedral (T_d) symmetry and known to be an efficient luminescent center with broad band emissions [7]. Numbers of vanadate phosphors may show broadband emission from 400 to more than 700 nm with excitation ultraviolet (UV) or near UV light because of the $[\text{VO}_4]^{3-}$ group and the transition in its energy levels [8]. The excitation wavelength matches well with the emission of AlGaIn-based near UV LED chips.

Eu^{3+} ion is one of important RE ions as activator-ions of luminescent materials. Eu^{3+} -doped phosphors usually exhibit intense sharp line emission owing to the ${}^5\text{D}_0 \rightarrow {}^7\text{F}_j$ ($j=1-6$) electronic transitions [9]. The typical absorption bands peaking at ~ 395 and 465 nm due to the ${}^7\text{F}_0 \rightarrow ({}^5\text{L}_6 \text{ and } {}^5\text{D}_2)$ transitions of Eu^{3+} are well matched with the commercial n-UV and

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blue LED chips. The alkaline earth metal orthovanadates with general formula, $M_3(VO_4)_2$ ($M = Ca, Sr, Ba$) can be used as host materials for the doped RE ion [10]. Eu^{3+} -doped vanadates phosphors usually show not only effective typical $f-f$ transitions of Eu^{3+} ion but also a broad and intense charge transfer band (CTB) absorption in the UV region from oxygen to metal ions. In recent years, many researchers pay more attentions to the fluorescence properties of $M_3(VO_4)_2$ ($M = Ca, Sr, Ba$) doped with RE ions due to the good nonlinear optical properties [11]. In addition, it is well known that charge compensation is one of important methods to improve the luminescence properties of phosphors.

In the paper, a series of $Sr_3(VO_4)_2:xEu^{3+}$ ($0 \leq x \leq 6$ mol%) and $Sr_3(VO_4)_2:3\%Eu^{3+}, 3\%Li^+$ phosphors are synthesized by high-temperature solid-state reaction method in air. The crystal structures and luminescence properties are investigated. ET process between $[VO_4]^{3-}$ group and Eu^{3+} ion is observed and analyzed by emission spectrum and energy level diagram. The influences of Eu^{3+} doping concentration to luminescence properties and lifetime are discussed. The improvement of Li^+ ion to luminescence properties is shown and explained. The luminous mechanism of $Sr_3(VO_4)_2:Eu^{3+}$ phosphor is explained by energy level diagram of $[VO_4]^{3-}$ group and Eu^{3+} ion.

2. Experimental

All the chemicals are purchased from the Aladdin Chemical Reagent Company in Shanghai China, such as $SrCO_3$ (A.R. 99.5%), Li_2CO_3 (A.R. 99.5%), V_2O_5 (A.R. 99.9%), and Eu_2O_3 (99.99%). $Sr_3(VO_4)_2:xEu^{3+}$ ($x = 0, 1, 2, 3, 4, 5,$ and 6 mol%) and $Sr_3(VO_4)_2:3\%Eu^{3+}, 3\%Li^+$ phosphors are synthesized by high-temperature solid-state reaction method in air. The stoichiometric amount of raw materials are well grounded in an agate mortar without further purification, then sintered at $500^\circ C$ for 6 h, and subsequently $900^\circ C$ for 5 h in air. All products are obtained after natural cooling to room temperature. Repeated grindings are performed between two sintering processes to improve the homogeneity.

The crystal structures of all phosphors are checked by X-Ray Powder Diffraction (XRD) (Philips Model PW1830) with $Cu-K\alpha$ radiation at 40 kV and 40 mA at room temperature. The data are collected in the 2θ range of $10-90^\circ$. Luminescence properties and fluorescence lifetimes of all phosphors are investigated by using a steady-state FLS980 spectrofluorimeter (Edinburgh Instruments, UK, Edinburgh) with a high spectral resolution (signal to noise ratio > 12000:1) at room temperature. A 450 W ozone free xenon lamp is used for steady-state measurements. A microsecond pulsed xenon flash lamp $\mu F900$ with an average power of 60 W is available to record the emission decay curves for lifetimes.

3. Results and discussion

The unit cell of $Sr_3(VO_4)_2$ drawn on the basis of the Inorganic Crystal Structure Database (ICSD) #73258 is shown in Fig. 1. $Sr_3(VO_4)_2$ is described as the rhombohedral crystal system with space-group $R-3m$ (166) and the lattice parameters $a = 5.619(1) \text{ \AA}$, $c = 20.100(4) \text{ \AA}$, $v = 549.60(22) \text{ \AA}^3$, and $z = 3$ [12]. $Sr_3(VO_4)_2$ crystal structure contains $[VO_4]$ tetrahedra, $[Sr(2)O_{10}]$ polyhedra, and $[Sr(1)O_{12}]$ icosahedra. There is a layered structure by linking $[VO_4]$ tetrahedra. $[Sr(2)O_{10}]$ polyhedra is described as a combination of a half cuboctahedron and a hexagonal pyramid, and the polyhedral around central atom V is tetrahedral structure [13]. The Eu^{3+} ions occupy the Sr^{2+} ions site in the host lattice owing to their similar ionic radii (Eu^{3+} : $\sim 0.95 \text{ \AA}$ and Sr^{2+} : $\sim 1.13 \text{ \AA}$).

XRD patterns of Joint Committee on Powder Diffraction Standards (JCPDS) card no. 19-1289 ($Sr_3(VO_4)_2$), blank $Sr_3(VO_4)_2$, $Sr_3(VO_4)_2:xEu^{3+}$ ($x = 1, 3,$ and 6 mol%), and $Sr_3(VO_4)_2:3\%Eu^{3+}, 3\%Li^+$ are shown in Fig. 2(a). The XRD patterns of these samples all match well with the standard data of JCPDS card (no. 19-1289). The XRD patterns of other $Sr_3(VO_4)_2:xEu^{3+}$ ($0 \leq x \leq 6$ mol%) phosphors are not displayed in Fig. 2(a), but those patterns are also in line with those of JCPDS card (no. 19-1289). No other crystalline phase is formed after Eu^{3+} ions are doped. This is said that all samples are pure phase $Sr_3(VO_4)_2$. After Eu^{3+} and Li^+

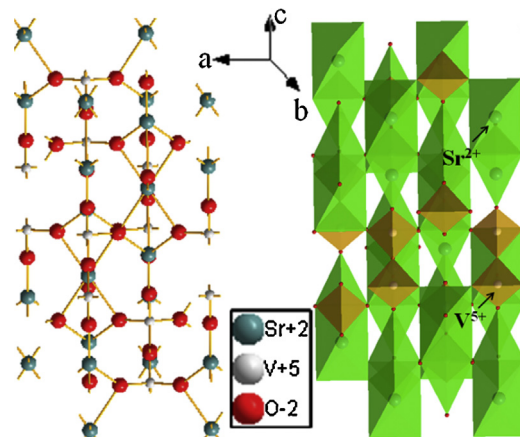


Fig. 1. The unit cell of $Sr_3(VO_4)_2$ drawn on the basis of ICSD #73258.

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