



Research Paper

PREPARATION OF Eu-ACTIVATED Sr₂SiO₄ PHOSPHOR BY A COMBUSTION METHOD AND ITS OPTICAL PROPERTIES

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ABSTRACT

Eu³⁺ activated Sr₂SiO₄ nanophosphors were prepared by low temperature solution combustion method using urea [CO(NH₂)₂] as a fuel. The Prepared phosphor was well characterized by powder X-ray diffraction and UV-visible spectroscopy. The photoluminescence properties of the nano-size Sr₂SiO₄ : Eu³⁺ phosphors excited under 256nm, showed intense emission in red region. Sr₂SiO₄:Eu³⁺ phosphors exhibited white emissions ranging from 500 to 750 nm when it was excited by near-ultraviolet (near-UV) light, indicating that Sr₂SiO₄:Eu³⁺ was a good light-conversion phosphor for near-UV chip.

KEYWORDS: Photoluminescence, X-ray, Combustion Method, nanocrystalline materials.

1. INTRODUCTION

Light-emitting diode (LED) with the phosphors material to generate white light is the present focus of research in the lighting industry [1]. White LED shows many excellent properties such as low energy consumption, small size, light weight and long lifetime [2]. Moreover, they have an environment benefit in that they do not use mercury in comparison to fluorescent lamps [3]. Therefore, the white LEDs are expected to enable new concepts in lighting field. There are basically two approaches to generate white light using LEDs. One is the combination of light of three primary colors (red, green and blue) emitted from different LED chips. Another is to down-convert the emission from a blue or ultraviolet LED to a longer wavelength light by using down-converting phosphors [4,5]. Strontium orthosilicate exists in two crystallographic phases viz. α -Sr₂SiO₄ (orthorhombic) and β -Sr₂SiO₄(monoclinic) [6].The transition occurs from low temperature β -phase to high temperature α -phase at 385 K, and involve the rearrangement of tetrahedral SiO₄ without change in bonds[7-9].The photoluminescence (PL) is observed in these materials are attributed to the f-f and f-d transitions of rare-earth ions. Whereas the intensity depends on the site symmetry and the nature of the host matrix. In this paper, the Sr₂SiO₄:Eu³⁺phosphors prepared by combustion synthesis were investigated and the effect of Eu³⁺ concentration on micro structural as well as luminescent characteristics of Sr₂SiO₄:Eu³⁺ phosphors were reported.

2. EXPERIMENTAL

Sr₂SiO₄: Eu phosphors were synthesized by combustion method. In combustion method, it is easy to synthesize compound powders of desirable characteristics, including very fine size, narrow size distribution, high purity and high chemical homogeneity. Strontium nitrate Sr(NO₃)₂, silica gel, europium oxide(Eu₂O₃) and urea CO(NH₂)₂ were used as the starting materials in stoichiometry weight. The small amount of ammonium chloride (NH₄Cl) is used as the flux while the urea [CO(NH₂)₂] as a combustion fuel[10]. After mixed sufficiently, the prepared paste is then placed in vertical cylindrical muffle furnace maintained at 700°C. Then the prepared samples were annealed at 800 °C for 2 h under an air atmosphere. The phase composition of the sample are examined by PANanalytical using Cu-

K α radiation ($\lambda=1.5406 \text{ \AA}$). In which a scan range of $2\theta = 20^\circ - 60^\circ$ with a step of 0.0130 and 148.920s as a count time per-step were used. Absorption spectrum was recorded using (Shimadzu UV-1700 UV-Visible) spectrophotometer. The excitation and PL emission were recorded by Spectrofluorometer (PerkinElmer LS45).

3. RESULT AND DISCUSSION

3.1 Optical Absorption Spectra

Optical absorption is important to study the behavior of nano-crystals and a fundamental properties. In figure-1, the optical absorption spectra of pure and europium doped Sr₂SiO₄ phosphors in the range of 200nm-500nm is shown. The absorption profiles exhibit broad band in the UV region (below 350 nm). In the absorption spectra of pure Sr₂SiO₄ absorption edges was found at $\lambda= 225 \text{ nm}$, as in turn the obtained band gap was found equivalent to 5.52 eV. Whereas in the absorption spectra of the Sr₂SiO₄:Eu³⁺ the absorption edges was found at $\lambda= 245 \text{ nm}$ and the calculated was 5.06 eV. This absorption edges may be originated from the charge transfer band (CTB) of Europium oxygen interaction. Several other weak peaks can also be seen in the range of 350-500 nm.

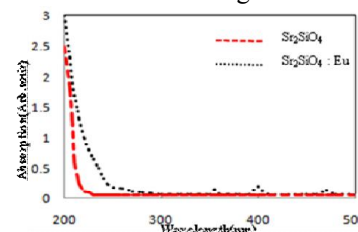


Fig.1. Absorption spectra of Pure and Sr₂SiO₄:Eu³⁺ phosphors.

3.2 Structural analysis: X-ray Diffraction (XRD)

The typical XRD pattern of Sr₂SiO₄ : Eu³⁺ Phosphor with Eu³⁺ concentration 0.01 mol% is presented in figure 2. From which it can be seen that all the diffraction peaks agree well with Ref. code: 98-003-5667, indicating that the orthorhombic phase of α -Sr₂SiO₄ with space group Pnma. It is well known that there are two crystallographic modifications of strontium orthosilicate (Sr₂SiO₄), β -Sr₂SiO₄ (monoclinic) and α' - Sr₂SiO₄ (orthorhombic). The β and α' - forms of Sr₂SiO₄ have closely related crystal structures made of SiO₄ tetrahedra. The only difference is the small tilting in the SiO₄ tetrahedra (T_d) leading to the absence of a mirror plane parallel to the (100) plane in the case of β - Sr₂SiO₄ [7-9]

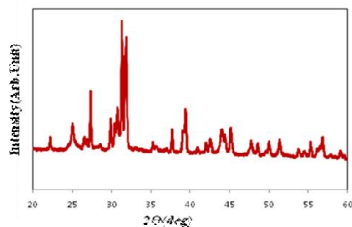


Fig. 2: XRD pattern of Eu^{3+} doped Sr_2SiO_4 phosphors.

3.3. Excitation and emission spectrum

The excitation spectra of nano-sized $\text{Sr}_2\text{SiO}_4:\text{Eu}^{3+}$ phosphors shown in fig. 3(a). The spectrum of $\text{Sr}_2\text{SiO}_4:\text{Eu}^{3+}$ phosphors exhibit a broad band in the UV region centered at 256 nm and sharp lines between 300-500 nm. The broad absorption band is called charge transfer band (CTB) due to the europium-oxygen interactions, which is caused by the electron transfer from filled 2p orbital of O^{2-} ions to vacant 4f orbital of Eu^{3+} ions [11]. The sharp excitation peaks located at 317, 364, 383, 395 and 465 nm are related to the inter-configuration 4f-4f transitions of Eu^{3+} ions in the host lattices, were assigned to electronic transitions of ${}^7\text{F}_0 \rightarrow {}^5\text{H}_6$, ${}^5\text{D}_4$, ${}^5\text{G}_2$, ${}^7\text{L}_6$ and ${}^5\text{D}_2$ respectively. The peak positions are good agreement with the emission spectra reported earlier [12-13]. The excitation spectra indicate that the intensities of both CTB and 4f-4f transition.

After excitation, the emission spectrum in fig. 3(b) are described by well-known ${}^5\text{D}_0 \rightarrow {}^7\text{F}_J$ ($J=0, 1, 2, 3, \dots$) emission lines of the Eu^{3+} ion, with strong emission at 613 nm (${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$). In our case, the intermediate state is thought to populate the lowest ${}^5\text{D}_0$ state leading to the emission at 613 nm. Other emission bands were observed at 575 nm, 593 nm, 651 nm and 704 nm [12-13]. In the current study, the concentration quenching effect is observed in excitation at 256 nm. The possibility of transition in any symmetry is based on the prevailing selection rules. For non centre of inversion symmetry, electron dipole transitions can occur and corresponds to $J = \pm 2$ as selection rules. But if, Eu^{3+} ion is at the centre of inversion symmetry for surrounding atoms in the lattice, only magnetic dipole transition are allowed and selection rules of $J = \pm 1$ are operative. It is well known that the ${}^5\text{D}_0 \rightarrow {}^7\text{F}_1$ (593 nm) transition is a weak electric dipole transition, it is allowed in low-symmetry sites. The ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ (613 nm) transition is magnetic dipole, which indicates that the Eu^{3+} ions lie in central symmetric sites.

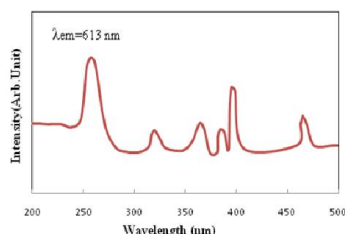


Fig.3(a): photoluminescence excitation spectra of $\text{Sr}_2\text{SiO}_4:\text{Eu}^{3+}$ phosphors

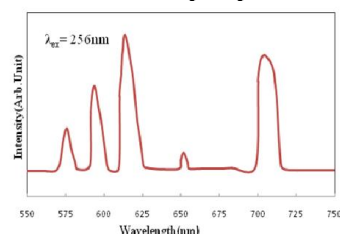


Fig.3(b) photoluminescence emission spectrum of $\text{Sr}_2\text{SiO}_4:\text{Eu}^{3+}$ phosphors ($\lambda_{\text{ex}}=256$ nm)

4. CONCLUSION

In this work $\text{Sr}_2\text{SiO}_4:\text{Eu}^{3+}$ phosphors prepared by combustion method. XRD studies confirmed the formation of a single phase compound. The absorption spectra of pure are widened as compare to Eu^{3+} Doped Sr_2SiO_4 . The photoluminescence intensity of the reddish emission at 575 nm, 593 nm, 613 nm, 651 nm, 704 nm under excitation 256 nm was appeared, so these PL intensity may have good potential for producing the high-quality white LEDs.

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