Synthesis and Photoluminescence Properties of Eu³⁺ Doped Strontium Cerium Sulfide Phosphor

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Abstract: Eu (0.5 mol%)-doped strontium cerium sulfide (SrCeS) phosphor was synthesized by solid state reaction method. The photoluminescent properties of the synthesized phosphor were analyzed by Spectrofluorophotometer. Efficient energy transition has been observed by photoluminescence investigation upon UV – visible (263, 320, 466, 535 and 650 nm) excitations and results in red emissions at about 595nm and 611nm. The excitation spectrum monitored under 611nm wavelength consists of broad and sharp peaks covering from 220-650nm. The photoluminescence intensity varies with excitation wavelength. The Commission International de l'Eclairage (CIE) co-ordinates were calculated by the spectrophotometric method using the spectral energy distribution of 595nm peak is (0.602, 0.397) and of 611nm peak is (0.668, 0.332) confirm the red emission. Chromaticity coordinates does not change distinctly with the change of excitation wavelengths. The phosphor exhibit stable red light indicates that it has potential application in display and in w-LED.

Keywords: Phosphor, Photoluminescence, Excitation, Emission, Spectral energy, CIE

1. Introduction

In recent years LEDs are potential candidates for replacing conventional incandescent and fluorescent lamps due to the advantages of long lifetime, saving energy, high efficiency, reliability and environmental friend characteristics [1, 2]. During the past few years, more and more cities have begun replacing traditional streetlights (sodium, mercury lamps) with newer LEDs. The past few decades have seen a lot of work reported on the use of divalent/trivalent Europium as a dopant in phosphors as they have very good optical properties. The luminescence associated with 'Eu' contained in different host lattices has found applications related to its red light emission which is associated with the MD (${}^{5}D_{0} \rightarrow$ ${}^{7}F_{1}$) and ED (${}^{5}D_{0} \rightarrow {}^{7}F_{2}$) transitions respectively, which is important in the fields of displays, sensors, lasers and LED's. Among all the rare-earth ions, Eu^{2+} and Eu^{3+} is the most extensively studied, owing to the simplicity of its spectra and also its use in commercial red phosphors.

Strontium cerium oxide is a blue phosphor which emits red light by doping Eu ion was studied extensively and reported by several researchers [3-19]. Our group made an attempt to study PL properties of strontium cerium sulfide phosphor for w-LED applications. Sulfide based phosphors were suitable for w-LED applications due to their excitation and emission wavelengths are in visible region. In this letter, we report the Eu-doped strontium cerium sulfide phosphor by high temperature solid state reaction method. Photo luminescent properties of the powder were investigated. The emission, excitation spectra and CIE were done on the prepared sample at room temperature.

2. Experimental Work

Assay 99.9% purity compounds like Strontium sulfide (SrS), Cerium Oxide (CeO₂) and Europium Oxide (Eu₂O₃) were used as starting materials for Eu-doped SrCeS phosphor synthesis. These starting materials were weighed according to stoichiometric equation and mixed/ground into a fine powder using agate mortar and pestle about an hour. The grounded sample was placed in an alumina crucible and fired at 1000°C for 3 hours in a muffle furnace with a heating rate of 5°C/min. After heating, the sample was cooled down to room temperature in the same furnace. The sample was again ground to powder for PL characterization. The emission and excitation spectra were measured by Spectrofluorophotometer (SHIMADZU, RF-5301 PC) using Xenon lamp as excitation source. All the spectra were recorded at room temperature. The emission colour can be seen by CIE colour coordinates were calculated by the spectrophotometric method using the spectral energy distribution on 1931 chart using Radiant Imaging software.

3. Results and Discussion

3.1 Excitation Study

Fig.1 presents the excitation and emission spectra of SrCeS: Eu³⁺ phosphor fired at 1000°C in air for 3 h. The excitation spectrum (below part of fig.1) was obtained by monitoring the 611 nm emission of $Eu^{\overline{3}+}$ (${}^{5}D_{0} \rightarrow {}^{7}F_{2}$) transition. It can be seen clearly that the excitation spectrum consists of an intense broad band with the maximum at 263 and 320 nm and some sharp weak lines in the range from 395 to 585 nm. From the spectrum it is observed that there is an obvious charge imbalance in the lattice when Eu³⁺ ions substituting for Sr²⁺ ions in the phosphor. The excitation spectrum consists of two main features, a broad band with high intensity from 220 - 350nm centered at 263 and 320nm which is assigned to the charge transfer band (CTB), such result shows that the phosphor can be excited efficiently by low-pressure mercury lamp. The sharp weak lines located at 395, 466, 533, 580 and 595 nm are related to the intra configurational 4f-4f transitions of Eu³⁺ ions in the host

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lattices, which can be assigned to ${}^{7}F_{0}\rightarrow{}^{5}L_{6}$, ${}^{7}F_{0}\rightarrow{}^{5}D_{2}$, ${}^{7}F_{0}\rightarrow{}^{5}D_{1}$, ${}^{7}F_{0}\rightarrow{}^{5}D_{0}$ transitions, respectively. Among these excitation transitions 533nm is the most intense one than others. It is well known that 4f-4f transition and CTB are two types for excitation of the trivalent lanthanide ions in crystals. The 4f orbital is shielded from the surroundings by the filled ${}^{5}s_{2}$ and ${}^{5}p_{6}$ orbital. Therefore, the influence of the host lattice on the optical transitions are sharp lines. The CTB absorption shows a broad band character which is intense than 4f-4f transitions. The intra-configurational f-f transitions of Eu³⁺ implies that as prepared phosphor can be effectively excited by radiations of wavelength not only UV but also blue light (466nm). This can be made use of suitable luminescent material in white LED applications.

3.2 Emission Study

The emission spectrum (above part of fig.1) was measured under 263, 320 and 466 nm excitations. 263 and 320nm are corresponds to the broad CTB absorption. The emission spectrum shows the typical emissions of Eu³⁺ ions. All the spectra are composed of several narrow lines in the range 550-700 highest emission peaks are located at 595nm corresponding to the Eu³⁺ magnetic dipole (MD) transition of ${}^{5}D_{0} \rightarrow {}^{7}F_{1}$, and electric dipole (ED) transition of ${}^{5}D_{0} \rightarrow {}^{7}F_{2}$. The MD transition is much stronger than the ED one is due to the Eu³⁺ ions mainly occupy centrosymmetric sites and the MD is dominant in the host lattice. The most intensive narrow emission with the FWHM is less than 4nm, which indicates that the prepared phosphor has excellent crystallinity.

Fig.2 shows the emission spectra of the prepared phosphor under 533nm and 670nm excitations. From fig.1 & 2 it is observed that the phosphor exhibit good intense emission under visible excitation wavelengths, which is useful for LED applications. It is observed that when the phosphor is excited with 670nm, the phosphor exhibited the same emission with good intensity i.e. energy transfer from low to high which is a novel emission.

3.3 CIE Colour Coordinates Study

The CIE chromaticity coordinates for Eu-doped SrCeS phosphor were calculated based on the measured emission spectra. Fig.3 is the Commission Internationale de l'Eclairage (CIE) coordinates which are calculated using the spectral energy distribution (1931 chart). Fig.3 shows corresponding CIE chromaticity diagram of the Eu-doped SrCeS phosphor under 263, 320, 466, 533, 670nm excitations. The dark point A and B indicates the coordinates of 595nm (x=0.602, y=0.397) peak and 611nm (x=0.668, y=0.332) peak respectively. As the excitation wavelength changed the colour coordinates are not changed.



Figure 1: PLE spectrum (below) and PL spectra (above) of Eu-doped SrCeS phosphor



Figure 2: PL emission spectra of Eu-doped SrCeS phosphor under 533 and 670nmexcitations



Figure 3: CIE co-ordinates of Eu-doped SrCeS phosphor depict on 1931 chart A) 595nm peak B) 611nm peak

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4. Conclusions

In summary, Eu^{3+} (0.5 mol%) doped SrCeS phosphor was synthesized by solid state reaction method. The characteristic emission peaks of Eu^{3+} ions located at 595nm and 611nm are observed. The Eu^{3+} doped SrCeS phosphor exhibit stable red emission when the excitation wavelengths changes from UV- visible. Under 320nm excitation the phosphor shows high emission intensity than other excitations. This phosphor transfers energy from low to high under 670nm excitation. The results presented reveal the potential applications are in display, lamps and w-LEDs.

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