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Effect of Calcination Temperature on Photoluminescence Intensity of Sol-Gel Prepared $Sr_2CeO_4:Eu^{3+}$ Nano Phosphor

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Abstract

Pure Sr_2CeO_4 phosphor and Sr_2CeO_4 doped with europium (0.5, 1.0, 1.5, 2.0, 4.0 mol%) phosphor were synthesized by the sol-gel method. X-ray diffraction (XRD), scanning electron microscopy (SEM) and photoluminescence (PL) spectra were used to characterize the pure Sr_2CeO_4 and $Sr_2CeO_4:Eu^{3+}$ nano phosphor. It is a well-known fact that the crystallinity of the material increases with increase in the calcination temperature. Hence to study the effect of calcination temperature on the sol-gel synthesized phosphor ($Sr_2CeO_4:Eu^{3+}$(2.0 mol%)), the phosphor was fired at various temperatures 1000, 1100 and 1200 ℃ for 2hrs respectively and it was found that the photoluminescence intensity of $Sr_2CeO_4:Eu^{3+}$ nano phosphor increases with the increase of calcination temperature. $Sr_2CeO_4:Eu^{3+}$ nano phosphor has an excellent colour tunability of red light with different concentration of europium and hence it can be a promising candidate for LED lighting Applications.

Keywords: Sr_2CeO_4 Nano Phosphor; Sol-gel technique; Photoluminescence Intensity; Calcination Temperature.
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1. Introduction

The optical properties of the solids have been an area of interest for the past few centuries, but for the past few decades, they have been an area of high interest for those working with nanocrystals. The optical properties can give an insight to the mechanism and the nature of the material formed. These properties have led to the discovery of various inorganic hosts with different technological applications. The concentration of these have mainly been on the metallic nanocrystals[1] and semiconductors (mainly the doped nanocrystals (DNC)), which shows the quantum confined effect. The metallic nanocrystals have been used for various applications like optical data storage, solar energy conversion, and sensors, as catalysts because of their high surface to volume ratio and due to their different shapes. The phosphors with short decay times find applications as essential components in fluorescent lamps, television picture tubes, flat-panel displays, plasma display panels, *etc*.[2,3-9] Table 1 lists phosphors applications in devices. However, phosphors with delayed emission are also extremely useful as sources of light energy (lamps) at the time of sudden power failure or intentional blackouts. These have great potential for luminous paints, watches,[10,11] display industries,[12,13] exit signage and emergency escape route guidance systems. The interest in these phosphors has grown due to the increasing number of applications in radiation detectors, sensors for structural damage,[14,15] and temperature detection.[16] Considering its innumerable applications, the present work was concentrated on the synthesis, characterization, and studies on luminescence properties of multi-color emitting phosphors.

White light emission from a single host lattice is a hot topic today. The quality of the white light depends on many factors such as the correct blue-to-yellow intensity ratio, density, particle size of phosphor, and the coating quality.[17,18] The phosphor research impetus has increased for making better red, green and blue phosphor. The phosphor devices should have high resolution, high packing density (also require low binder content), and higher brightness for better performance, but these can be achieved only when the size of the particle is very small.[19-22] The sol-gel method offers many advantages over conventional solid-state and other techniques are reported.[23] Phosphors. The main aim of the present research work is to develop indigenous blue and white phosphors for Lamps. An attempt was made in this direction.

2. Experimental section

2.1 Sol-gel technique

The starting materials $Sr(NO3)_3$, $Ce(NO3)_3 \cdot 6H_2O$, citric acid, ethylene glycol, and liquid ammonia (NH_3) were

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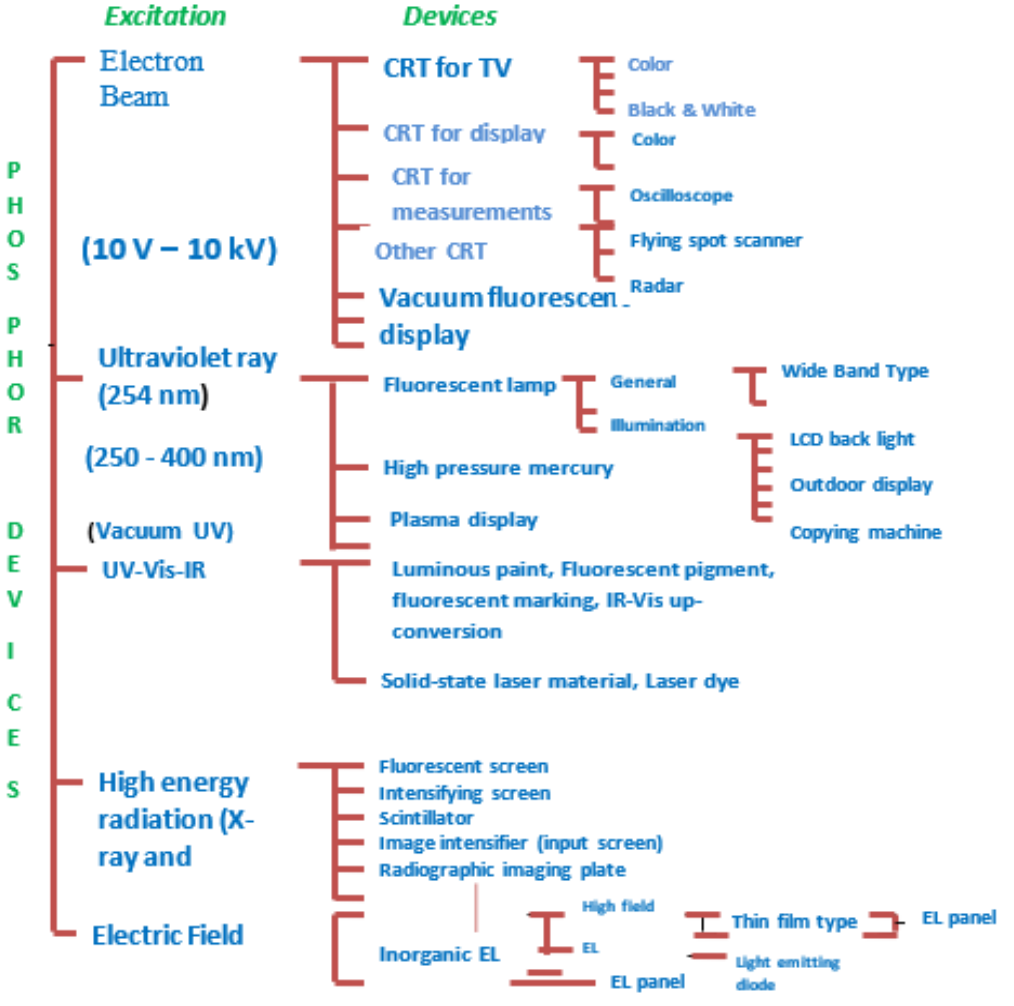
Table 1. Phosphor Devices and their Applications.



purchased from S.D. fine chemicals (Boisar, Mumbai, India). The stoichiometric ratio of Ce: Se was taken as 1:2. The nitrates were first dissolved in around 20 ml of double distilled water and kept for stirring on a magnetic plate at room temperature until it becomes transparent in color and all the salt has mixed. Gradually the temperature of the plate was increased to 40-50 °C. After a certain time of continuous stirring, citric acid was added to the transparent solution. The pH of the resulting solution was maintained 6-7 by adding droplets of ammonia. The temperature of the solution was raised to 60- 80 °C. At this stage, the solution becomes milky in color due to precipitation and citric acid acts as a chelating agent. After a few hours of stirring ethylene glycol was added to the solution and stirring was maintained. At this time, the solution gradually turned into a yellow viscous gel.The gel is then kept in an oven at 110 °C for 4 hours to remove water content if any and for drying. After drying gel was made into various parts. Each part was given different heat treatment. The doping of the europium into the phosphor was done at the initial stage of the reaction using the Eu_2O_3 (99.9%) purchased from S.D Fine chemicals. For some of the synthesis in the sol-gel case the Eu_2O_3 was first dissolved with concentrated HNO_3 (36% w/v) from Merck, Mumbai, heated and then washed several times with double-distilled water.

2.2 X-ray Diffraction Studies

Phase identification of the powders was carried out by the X-ray powder diffraction using RIGAKU D'MAX III Diffractometer having Cu Kα radiation (λ=1.54 nm). The scan range was kept from 50 to 800 at the scan speed of 0.050 per second.

2.3 Scanning Electron Microscope (SEM)

The Scanning Electron Micrograph images of the samples were taken using JEOL make JSM-5610 LV for studying the morphology of the compound.

2.4 Photoluminescence studies

The photoluminescence (Emission and Excitation spectra) were studied at room temperature using Spectrofluorophotometer RF-5301 PC of SHIMADZU. Xenon lamp is used as Source. The slit width for the emission and excitation was kept at 1.5 nm for all the measurements. A filter was used in order to remove the second-order peak of the excitation light in the PL measurements.

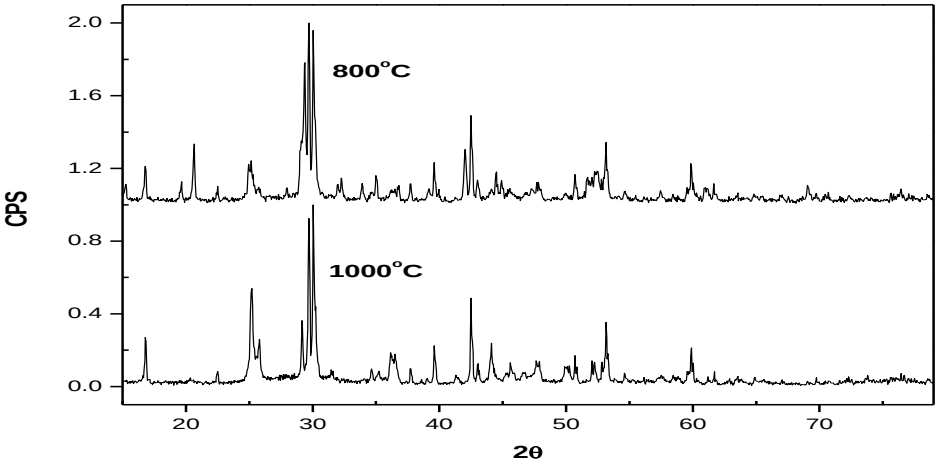


Fig. 1 The X-ray diffraction pattern of the europium doped Sr_2CeO_4 synthesized with a sol-gel technique (heated at at 800 ℃ & 1000 ℃ temperatures).

3. Results and discussion

3.1 X-ray Diffraction result

The crystallinity of the compound as revealed by the XRD pattern increased on raising the calcining temperature. This was also observed by Shikao Shi *et al*.[24] From the analysis of the XRD pattern, it was understood that the introduction of activator Eu^{3+} did not influence the crystal structure of the phosphor matrix. The calculated average crystal size of the sample calculated by measuring the full-width half maxima was found to be about 40 nm. Fig. 1 is the sol-gel synthesized Sr_2CeO_4 sample heated at different temperatures of 800 ℃ and 1000 ℃. The result shows that the phase of the material is almost pure with traces of $SrCeO_3$ present in it. It is observed from both the curves that the pure Sr_2CeO_4 has not formed at these temperatures. This concludes that if the pure phase has to be achieved then the sample has to be heated at1200 ℃. The X-ray diffraction pattern of the europium doped Sr_2CeO_4 synthesized with sol-gel and solid-state reaction technique heated at 1200 ℃ is shown in Fig. 2. XRD study confirms the single phase of the prepared sample with orthogonal structure.

3.2 Scanning Electron Microscope (SEM) result

From the SEM micrographs, the sol-gel synthesized samples are spheroidal in shape and even at high temperature of annealing they appear less agglomerated. The sol-gel synthesized samples are spheroidal in shape[25-27] and they are very small in size compared to solid-state ones.[28] This elaborates on another positive of the sol-gel synthesis technique. As the firing temperature in the case of sol-gel synthesized sample was increased from 400 ℃ to 1200 ℃, the shape and the size of the particles became spherical in shape. The spherical morphology has a positive effect on the optical property, as the scattering of the light is less and hence it directly influences the luminescence intensity. The micrographs of the sample are prepared from the europium doped Sr_2CeO_4 and one plain shown in Fig. 3 for comparison.

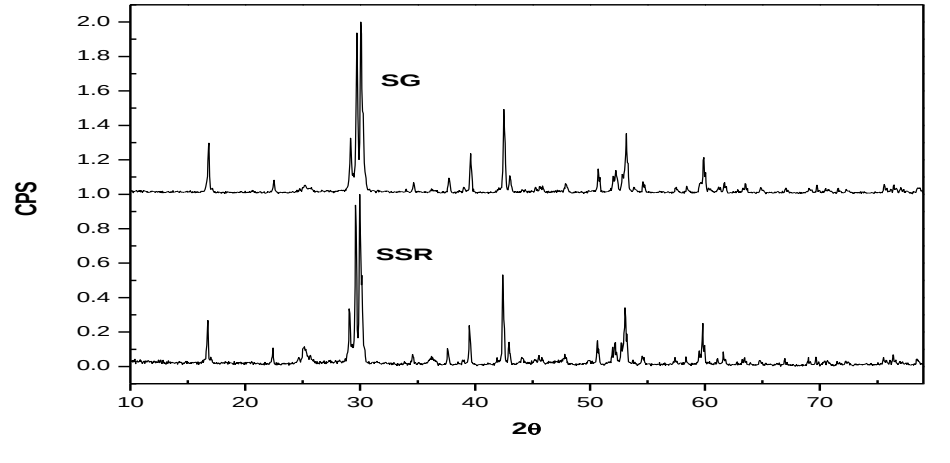


Fig. 2 The X-ray diffraction pattern of the europium doped Sr_2CeO_4 synthesized with sol-gel and solid-state reaction technique heated at 1200 ℃.

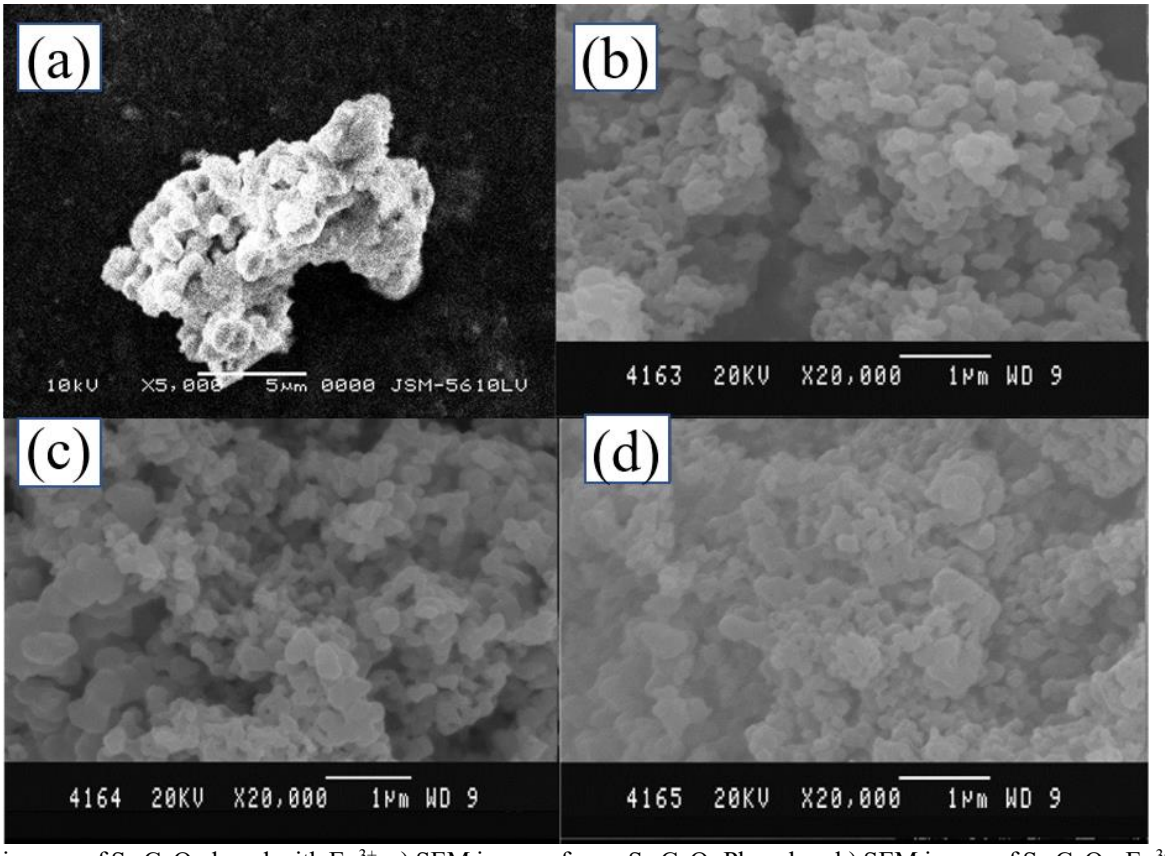


Fig. 3 SEM images of Sr_2CeO_4 doped with Eu^{3+}, a) SEM image of pure Sr_2CeO_4 Phosphor, b) SEM image of Sr_2CeO_4: Eu^{3+} Phosphor at 1000 °C, c) SEM image of pure Sr_2CeO_4: Eu^{3+} Phosphor at 1100 °C, d) SEM image of pure Sr_2CeO_4 Phosphor at 1200 °C.

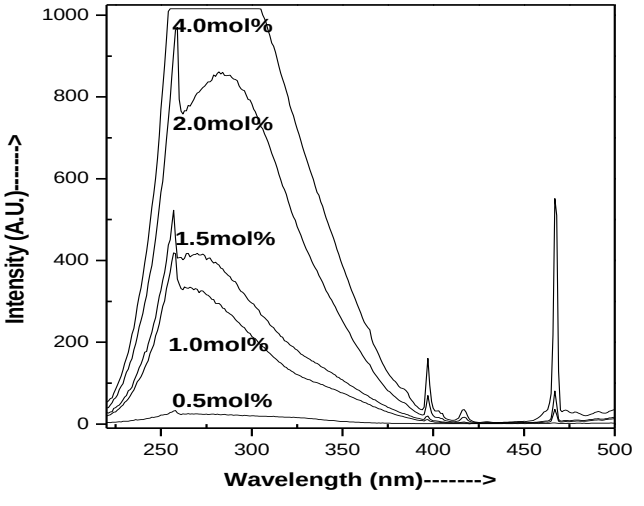


Fig. 4 The Photoluminescence excitation spectra of the Europium doped Sr_2CeO_4 Synthesized by sol-gel technique.

3.3 Photoluminescence Spectra of europium doped Sr_2CeO_4

Fig. 4 shows the excitation spectra of Sr_2CeO_4: Eu^{3+} (when monitored with 617 nm wavelength) measured at room temperature. It was found that at low concentration of Eu^{3+} (less than 2 mol %) only the charge transfer excitation band at 254 nm of Ce^{4+} is observed, but as the concentration of the Eu^{3+} increases (≥2 mol%), the excitation band shows the characteristic lines at 397, 417 and 467 nm, which along with the charge transfer band of Ce^{4+}, it due to the transitions $7F_{0,1}\rightarrow5L_6$ and $7F_0\rightarrow5D_2$ of Eu^{3+}, respectively. The excitation spectrum shows the presence of a strong peak at 467 nm at a high concentration of europium (≥2 mol %). This shows that the absorption of energy takes place in the $^5D_2$ level ($^5D_0\rightarrow{}^7F_2$). However, there is a non-radiative transition from $^5D_2\rightarrow{}^5D_0$ from where emission starts showing up with a high peak intensity for $^5D_0\rightarrow{}^7F_2$ transition. This is possible at a high concentration of Eu^{3+} ions.[29] The excitation spectra follow the same pattern for the entire emission wavelength. In our study, we can see that occupancy of Eu^{3+} ions are present at two different sites with totally ruled out the existence of different local environment. At higher concentration the stark splitting becomes negligible and this can be seen from the emission peak of 4 mol % europium doped sample, where the splitting of the $^5D_0\rightarrow{}^7F_2$ merged into a single peak. It is a well-known fact that the crystallinity of the material increases with an increase in the calcination temperature. As the degree of crystallinity of nanoparticles increase with increasing the calcination temperature and the intensity of photoluminescence increases with increasing crystallinity.[30] Hence to study the effect of calcination temperature on the sol-gel synthesized phosphor (Sr_2CeO_4:Eu^{3+} (2.0 mol %)), the phosphor was fired at various temperatures 1000 °C, 1100 °C, and 1200 °C for 2hrs, respectively. From Fig. 5, it is clear that

the photoluminescence intensity increases with an increase in the calcination temperature. No definite inferences regarding the peak shift with respect to different annealing temperatures can be drawn.

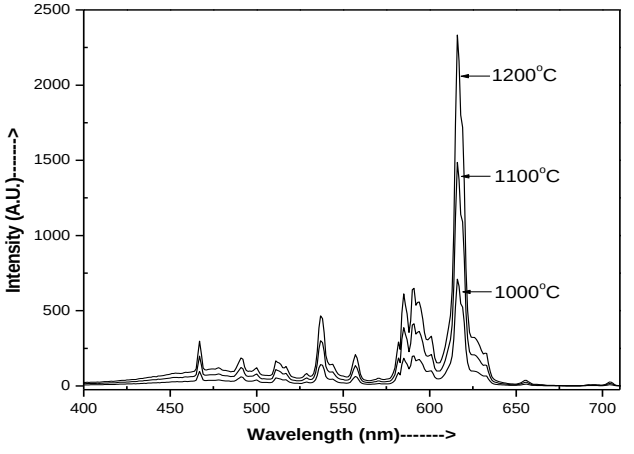


Fig. 5 The effect of calcination temperature on the sol-gel synthesized phosphor Sr_2CeO_4 doped europium.

4. Conclusions

The main conclusions that can be drawn by studying Europium doping on the luminescence properties of the Sr_2CeO_4 are as follows:

Europium doped Sr_2CeO_4 phosphor was prepared successfully by sol-gel synthesis. Doping of europium at various concentrations was done and a very rare europium emission was observed at 467 nm for the transition $5D_2 \rightarrow 7F_0$ photoluminescence measurements at room temperature. Excellent tunability of phosphor observed when doped with various concentrations of europium**.** The photoluminescence intensity is increased by increasing the calcination temperature.

Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

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