© 2018 IJSRST | Volume 4 | Issue 5 | Print ISSN: 2395-6011 | Online ISSN: 2395-602X
Themed Section: Science and Technology

Combustion Synthesis and Luminescence Properties of Ce^{3+} Doped Strontium Silicate Phosphors

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ABSTRACT

Cerium doped materials attracted the mind of researchers due to characteristic emission in the blue region and near blue region. We have prepared the cerium doped Sr_2SiO_4 phosphors by using the combustion synthesis. The prepared samples are characterized by the X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectra (EDS), ultraviolet visible spectroscopy (UV), Fourier transform infrared spectroscopy (FT- IR) and photoluminescence spectroscopy (PL). From XRD, it is confirmed that the prepared host Sr_2SiO_4 has orthorhombic crystal structure. The formation of the nano-crystalline nature of the samples has been confirmed by the SEM technique. EDS denotes the presence of the Sr, Si and O ions present in the prepared host and Sr, Si, O and Ce atoms present in the cerium doped nano-phosphors. The band gap energies of the host Sr_2SiO_4 and Ce^{3+} doped Sr_2SiO_4 are observed to be 4.5926 and 3.7126 eV. The formation of the Si-O, Si-Si and Sr-O bonds is confirmed by FT- IR. PL depicts the presence of the Ce^{3+} in Sr_2SiO_4 host has shown the emission in the blue region and therefore it is applicable in the blue display devices.

Keywords: XRD, UV, FT- IR, SEM, PL, EDS, Combustion Synthesis and Phosphor.

I. INTRODUCTION

E The rare earth ions activated inorganic phosphors have received much attention because of their wide applications in white LEDs, fluorescent lamps, display devices, solid-state lasers, biological labelling and so on [1,2]. Different rare earth doped materials had been studied by the researchers in last three decade. The emission in the visible region is applicable in different field [1, 2]. Among RE^{3+} ions, the Ce^{3+} doped materials have attracted more interest because their spin and parity allowed optical 4f → 5d transitions which have a fast radiative lifetime of about 10 to 50 ns [3], which is desirable for applications in Scintillators, light-emitting diodes and field emission displays.

Recently, significant efforts have been devoted by several research groups on the synthesis and characterization of various Ce^{3+} doped aluminates [4-5], silicates [6], fluoride and oxide [7-10] materials. The

Ce^{3+} doped Sr_2SiO_4 phosphors were studied using different method of the preparation. The characteristic emission of Ce^{3+} doped silicates is observed in blue or near blue region [5, 11]. To study the effect of method of preparation on the emission properties of Ce^{3+} doped Sr_2SiO_4, we have selected the combustion method to prepare the phosphor material. We have prepared the cerium doped Sr_2SiO_4 phosphors by combustion synthesis by varying the concentration of cerium ions from 1 mole % to 10 mole %.

II. METHODS AND MATERIAL

The Ce^{3+} doped Sr_2SiO_4 phosphors are prepared by combustion synthesis using high purity (AR grade) $(SrNO_3)_2$ $6H_2O$, SiO_2 x H_2O, (NH_4) (NO_3) and Ce $(NO_3)_3$ $9H_2O$ as initial raw materials and urea is used as

fuel agent. All precursors with stoichoimetric ratio are dissolved in the 20 ml distilled water and stirred it for 30 min using magnetic stirrer. The homogeneous solution is obtained, further it is placed in muffle furnace which is maintained at 500 °C temperature. The solution was ignited and foamy powder samples are formed with evolution of gases during ignition of urea. During this ignition, the temperature of the solution becomes about 1400 °C and due to this nano crystalline samples are obtained. The prepared powder samples are characterized for their phase purity and crystallinity by XRD. The PL measurement of excitation and emission spectrum are recorded with slit width of 1.5 nm and equal amount of sample (0.5 gm) for each measurement on the Shimadzu RF5301PC spectrofluorophotometer.

III. RESULTS AND DISCUSSION

A. XRD Studies:

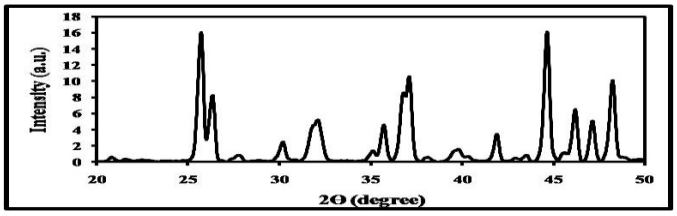


Figure 1. XRD pattern of Sr_2SiO_4(Host)

Figure 1 denotes XRD pattern of the Sr_2SiO_4 host. XRD pattern of Sr_2SiO_4 match with JCPDS file number 39-1256. It has shown orthorhombic structure with cell parameters a= 5.618, b= 9.678, c= 7.181. In the XRD pattern, peaks are well seen with high intensity which confirmed the highly crystalline nature of the prepared host Sr_2SiO_4 sample.

The particle size of the prepared material from high intensity peck of XRD pattern is calculated by scherrer's formula and it is found to be 79 nm.

B. SEM Analysis:

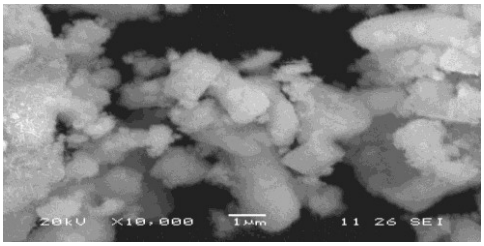


Figure 2. SEM image of Sr_2SiO_4

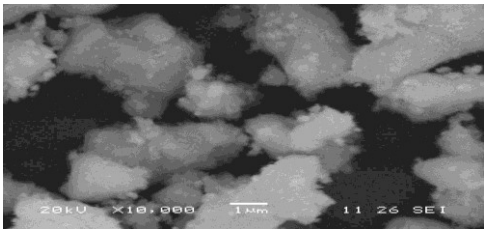
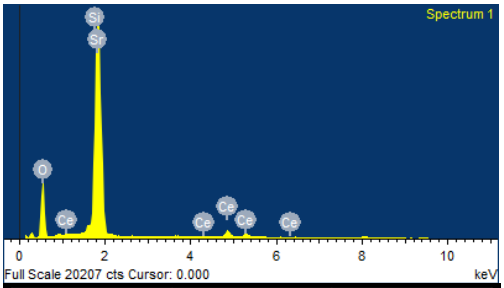


Figure 3. SEM image of $Sr_{2-x}SiO_4$: Ce_x

The morphological analysis is carried out by using scanning electron microscopy (SEM). Figure 2 and 3 represent the SEM images of prepared Sr_2SiO_4 and Cerium doped Sr_2SiO_4 phosphors respectively. From these SEM images, it is observed that Sr_2SiO_4 phosphor having nanostructure but these are highly agglomerated and the particles are having irregular size. The average particle sizes of phosphors calculated by using Image J software and it found near 60 nm.

C. EDS Analysis:



Element	Weight%	Atomic%
O K	48.60	80.55
Si K	6.79	6.41
Sr K	40.51	12.26
Ce K	4.10	0.78

Figure 4. EDS Spectrum and Elemental Analysis of $Sr_{2-x}SiO_4$: Ce_x

Figure 4 represents the EDS spectrum and elemental composition of Ce^{3+} doped Sr_2SiO_4. The EDS spectrum of Ce^{3+} doped Sr_2SiO_4 shows the signals for Sr, Si, O and Ce in prepared host material. These observations combined with XRD analysis, confirm successfully preparation Ce^{3+} doped Sr_2SiO_4 phosphors.

D. UV Measurements :

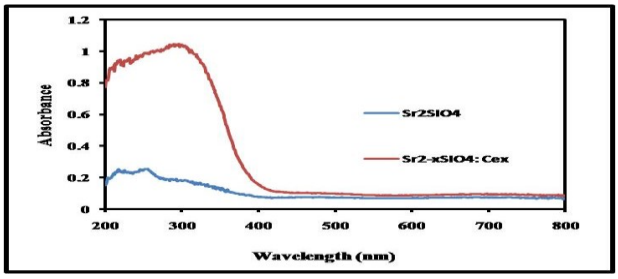


Figure 5. UV- Visible Spectra of $Sr_{2-x}SiO_4$: Ce_x

Figure 5 denotes the UV of the prepared Ce^{3+} doped Sr_2SiO_4 phosphor. The calculated energy band gaps for host Sr_2SiO_4 and Cerium doped Sr_2SiO_4 are 4.5926 eV and 3.7126 eV respectively. The band gap of host material decreases to 3.7126 eV with 10 m% doping of Ce^{3+}. The red shift is occur for absorption edge wavelength and this might be happen due the Ce^{3+} ions accquires the energy levels in the band gap of the host material. Also there is the change in intensity is observed for Ce^{3+} doping and this change in the intesity is occured due to the high absorption of energy due to the Ce^{3+} ions.

E. FT-IR Spectroscopy Analysis:

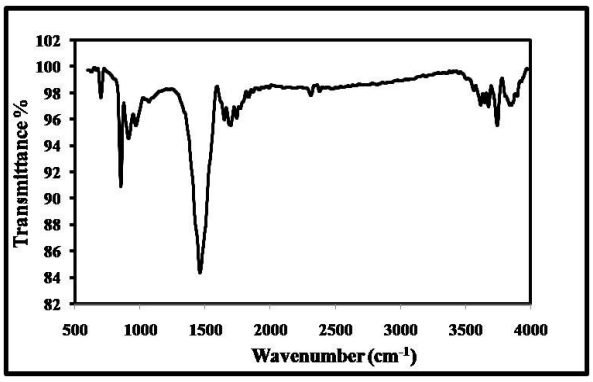


Figure 6. FT- IR Spectrum of Sr_2SiO_4 host

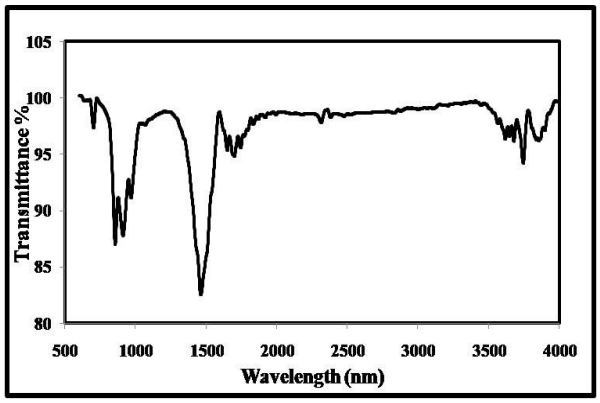


Figure 7. FT- IR Spectrum of $Sr_{2-x}SiO_4$: Ce_x

The molecular structure of Sr_2SiO_4 host and Ce^{3+} doped Sr_2SiO_4 is analysed by FT-IR spectroscopy technique. Figure 6 and 7 show the FT- IR spectra of Sr_2SiO_4 host and Ce^{3+} (10 m %) doped Sr_2SiO_4 phosphor powder respectively. The absorption band at 1480 cm^{-1} is ascribed to Sr-O stretching vibration [12]. The absorption in the range of 400-557 cm^{-1} is due to stretching vibrations of and Si-O bending vibration [13]. The absorption peak at 3428 cm^{-1} is due to H-OH bond vibration of absorbed water [12-13]. The absorption band in the 800-1000 cm^{-1} region indicates the stretching vibrations of Si-O-Si linkages in the SiO_4 tetrahedron unit [12-13]. From FT- IR analysis it is cleared that the bonding nature of the sample is consistent with the XRD analysis.

F. PL Measurement:

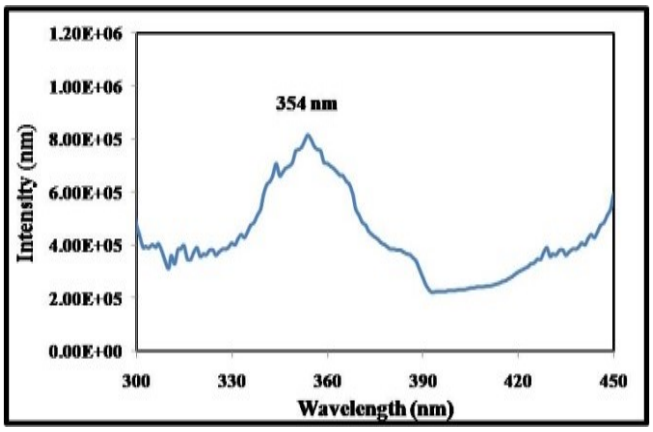


Figure 8. PL Excitation spectra of $Sr_{2-x}SiO_4$: Ce_x (x= 10 m%) monitored under 458 nm

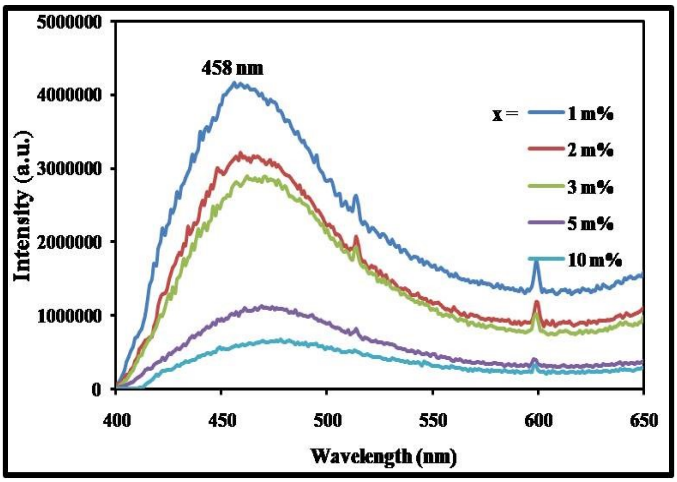


Figure 9. PL Emission spectra of $Sr_{2-x}SiO_4$: Ce_x monitored under 354 nm

We have done characterization of photoluminescence spectra for Ce $^{3+}$ doped Sr_2SiO_4. The excitation spectra for Ce^{3+} doped Sr_2SiO_4 is shown in Figure 8. The excitation spectrum is carried out at 450 nm emission wavelength. This broad peak at the 354 nm wavelengths is due to the characteristic 4f →5d transition of Ce^{3+} ion and this wavelength is used as excitation wavelength for emission specta of the sample. The emission spectra are shown in Figure 9 above. It has shown broad emission peak at 458 nm and which is due to transitions from the relaxed lowest 5d excited state of Ce^{3+} to the 4f ground state levels $^2F_{5/2}$ and $^2F_{7/2}$. The broad emission peck at 458 nm is used for excitation spectrum.

We have varied the Ce^{3+} in the Sr_2SiO_4 host as 1m%, 2m%, 3m%, 5m% and 10 m%. The enhancement in the intensity of Ce $^{3+}$ doped Sr_2SiO_4 is observed due to doping of Cerium ion. The maximum intensity is observed for 10 m% doped Ce^{3+} Sr_2SiO_4. Therefore, the prepared samples of Ce^{3+} doped Sr_2SiO_4 may be used in UV excitation and blue emission LED's.

G. Color Co-ordinate Calculations:

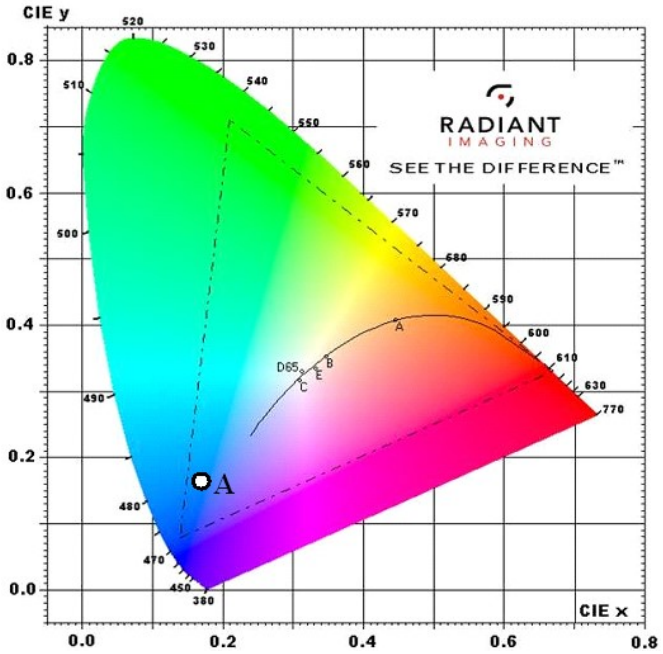


Figure 10. CIE chromatic diagram showing the chromatic coordinates of Ce^{3+} doped Sr_2SiO_4

The lighting calculations refer to color in terms of the 1931 CIE chromatic color coordinates which recognize that the human visual system uses three primary colors: red, green, and blue [14]. In general, the color of any light source can be represented on the (x, y) coordinate in this color space. The color purity was compared to the 1931 CIE Standard Source C (illuminant Cs (0.3101, 0.3162)). The chromatic coordinates (x, y) was calculated using the color calculator program radiant imaging [15]. The coordinates of the Ce^{3+} doped $SrSi_2O_4$ phosphors of blue color (x ≈ 0.1536, y ≈ 0.1705 are shown in Figure 10 by point A (white circle). This indicates that the color properties of the phosphor powder prepared by combustion synthesis does approaching those requirements of Solid state lighting.

IV.CONCLUSION

The Ce^{3+} doped Sr_2SiO_4 are prepared by the combustion synthesis. The nanostructure nature of the phosphor is confirmed by the SEM and XRD analysis. FT- IR analysis confirmed the bonding

nature of the prepared Ce^{3+} doped Sr_2SiO_4 phosphor. PL denotes the emission in the blue region of prepared Ce^{3+} doped Sr_2SiO_4 due to the 5d excited states to 4f ground state. Therefore this phosphor may be applicable in solid state lighting for blue emission.

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