

Synthesis and Characterizations of Strontium Cerium Oxide Phosphor

Pradip Z. Zambare ^{1*}, K. D. Girase¹ O. H. Mahajan² ¹Department of Physics, S. V. S's Dadasaheb Rawal College, Dondaicha, India. E-mail pzzambare@rediffmail.com. ²Department of Physics, M. J. College Jalgaon (M.S.), India. E-mail Ohmahajan56@gmail.com.

ABSTRACT

A new Strontium Cerium Oxide ($Sr_4Ce_2O_7$) blue phosphor with, orthorhombic structure was synthesized via standard solid state reaction method using strontium oxide and cerium oxide as a raw materials. The samples were characterized by Thermo analytical techniques (TG, DTA, and DTG), Fourier transformation infrared (FTIR) spectroscopy, Raman spectroscopy, and Photoluminescence studies at room temperature. In excitation spectra two excitation bands were located at 361 and 391 nm respectively. The emission spectrum was a broad band peaking at 474 nm, which was suitable for the doping of rare earth ions.

Keywords

TG, DTA, FTIR, Raman spectroscopy, Sr₄Ce₂O₇, X-ray diffraction .

Academic Discipline And Sub-Disciplines

Physics , Luminescence

SUBJECT CLASSIFICATION

Spectroscopy, Optical properties

TYPE (METHOD/APPROACH)

Solid state reaction method

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1 INTRODUCTION

The development of new compound for ultraviolet- excited phosphor is of great importance for flat panel display and illumination technology. Compared with organic materials and sulfide phosphors. Oxide- based phosphors have the advantages: stable crystalline structure and physical and chemical stability. Therefore oxide based phosphors, especially rare earth (RE) based oxides, are attracting more and more attention [1-5]. For full colour RE- based phosphors, the red and green emissions are generally generated by the f-f transitions of Eu³⁺ and Tb³⁺ while the blue one is caused by the d-f transitions of Eu²⁺ or Ce³⁺. To date, satisfactory red and green phosphors have been commercially available. However, comparable materials for blue emission are still lacking and they are the target of many research works [6-7]. White lightemitting diode(LED), the so- called next generation solid state lighting (SSL), with the advantages of long lifetime, saving energy consumption, and environmental friendly characteristics are attracting more attentions [8]. In view of application, white LEDs are expected to open up a great number of exciting new application field: white light sources to replace traditional incandescent and fluorescent lamps, backlights for portable electronics, medical, and architecture lighting etc [10]. The broad emission band is suitable for the doping of rare earth ions in pursuing new luminescent materials [11]. In this paper, we report here a new composition of blue strontium cerium oxide (Sr₄Ce₂O₇) was synthesized using solid state reaction method in air atmosphere. To the best of my knowledge, there is no literature is available on thermal analysis, Fourier transform infrared (FTIR), Raman spectra and luminescent properties of the synthesized new strontium Cerium Oxide (Sr₄Ce₂O₇).

2 MATERIALS AND METHODS

The starting materials were; Strontium Carbonate $SrCO_3$, Cerium Oxide Ce_2O_3 , of 99.5 % purity. The starting materials were taken in Stoichiometric proportions of (4:1) were thoroughly homogenized in agate mortar for 45 min and then transferred to alumina crucibles for heat treatment in air in muffle furnace. The compounds were subjected to heat treatment at 1050 - 1200 ⁰C for 40 -50 hours with three intermediate grindings and heating and finally cooled to room temperature by furnace shut off. All samples were prepared by same technique.

Thermo gravimetric analysis (TG, DTG, and DTA) of $Sr_2Ce_2O_7$ blue phosphor was carried out in air atmosphere in 30– 1200⁰ C temperature range using a Perkin Elmer Diamond TG/DTA instrument. The initial mass of sample taken for recording the TG/DTA curves was 64.778 mg and hold for 1.0 min at 30^o C, and then heating rate was maintained at 10^o C/min. The powder X- ray diffractograms (XRDs) of the compounds were recorded using an automated Rigaku Miniflex Xray diffractometer (D Max III VC, Japan). The observed (hkl) reflections and their intensities were compared with the calculated ones generated using the computer program POWD (an Interactive Powder Diffraction Data Interpretation and Indexing Program, Version 2.2). The FTIR spectra of $Sr_4Ce_2O_7$ were recorded on SHIMADZU IRAffinity-1 spectrophotometer with KBr pellet method over the wave number range 400–4,000 cm⁻¹. The photoluminescence excitation and emission spectra were recorded at room temperature using Spectrofluorophotometer (SHIMADZU, RF – 5301 PC) equipped with a 150 W Xenon lamp as excitation source

3 RESULT AND DISCUSSIONS

3.1Thermal analysis

Fig.1 shows simultaneous TG, and DTA curves of the precursor prepared by solid state reaction method. Initial mass loss of about 0.417% in TG curve in the range of temperature 116.68 $^{\circ}$ C to 265.93 $^{\circ}$ C arises due to starting of reaction. The mass loss of about 18.66 % occurs through the broad range 565.43 $^{\circ}$ C to 1200 $^{\circ}$ C due to the reaction between SrCO₃ and Ce₂O₃ accompanied by release of CO₂ it is in good agreement with the calculated value 18.83 %. The step inflection point in this stage occurs at 1061.55 $^{\circ}$ C and onset temperature is 965.80 $^{\circ}$ C. After this stage TG curve did not show an appreciable mass change in the temperature range of 1080- 1200 $^{\circ}$ C, indicate that the precursor is thermally stable in this range.

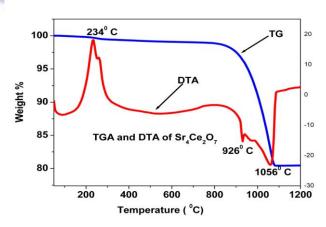


Fig. 1: TG, DTA curve of Sr₄Ce₂O₇



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The endothermic maximum below 100 0 C in DTA curve is due to loss of water absorbed by the samples when handled in air. The exothermic at temperature 233.37 0 C is occurs due to the phase change in starting of reaction between SrCO₃ and Ce₂O₃. The endothermic at 926 0 C and 1056 0 C in DTA curve corresponds to the mass loss due to the release of CO₂.

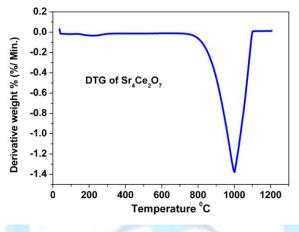


Fig. 2: DTG curve of Sr₄Ce₂O₇

The curve DTG shown in Fig. 2 the sharp endothermic peak at 980 $^{\circ}$ C is attributed due to the loss of CO₂. This endothermic peak observed in the DTG curve indicates that the starts at 965 $^{\circ}$ C and inflection occurs at 1061 $^{\circ}$ C. The peak observed in the DTG curve corresponding to the weight loss in the TG curve. Beyond the temperature 1100 $^{\circ}$ C, the reaction proceed and finally a stable residue Sr₄Ce₂O₇. No further loss is found, suggesting that the Sr₄Ce₂O₇ can be prepared at higher than 1100 $^{\circ}$ C. Following chemical reaction is expected to occur during the decomposition stage,

$$4SrCO_3 + Ce_2O_3 \rightarrow Sr_4Ce_2O_7 + 4CO_2$$

3.2 X-ray diffraction

Several samples of $Sr_4Ce_2O_7$ have been synthesized by solid state reaction method. Duplicate compositions have also been made to check on the consistency of behavior. We kept the muffle Furness temperature at 1200° C for 40 -50 hours then the structure and phase purity of the synthesized $Sr_4Ce_2O_7$ phosphor was investigated by X-Ray Diffraction Method. Results are shown in Fig. 3. All diffraction patterns were obtained using Cu K α radiation (λ = 1.54051 A $^{\circ}$), at 30 kV and 15 mA. Measurements were made from 2θ = 10° to 80° with steps of 0.02°. The crystallite size of powders samples were calculated from X-ray peak broadening of the diffraction using Scherer's equation

D = 0.9λ / β. Cosθ

Where, D is the crystallite size in nm, β represents full width at half maximum (FWHM) of XRD lines, λ is the radiation wavelength of X-ray (λ = 1.54051 Ű), and θ is diffraction peak angle.

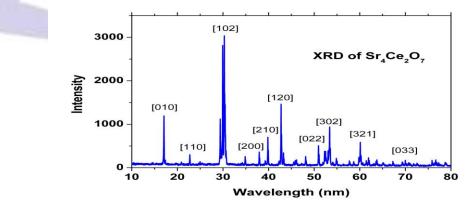


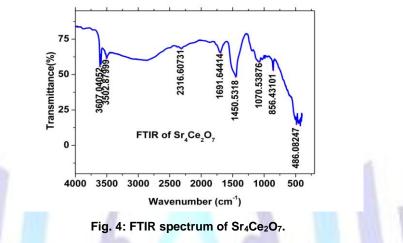
Fig. 3: XRD pattern of Sr₄Ce₂O₇.

The calculated average crystallite size of the Sr₄Ce₂O₇ phosphor is 49 nm. The computer program POWD (an Interactive Powder Diffraction Data Interpretation and Indexing Program, Version 2.2) was used to calculate hkl and d values which are reported in table 1. The XRD patterns of the powders revealed that the structure of Sr₄Ce₂O₇ is orthorhombic having lattice parameters a= 5.9431 A°, b= 5.1929 A°, c=7.0644 A° and cell volume V=218.02 (A°)³. JCPDS data card of Sr₄Ce₂O₇ material is not available.



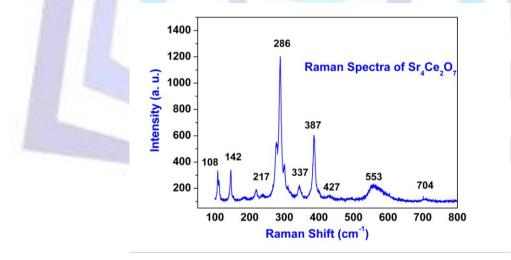
3.3 FT-IR Spectrometry

The synthesized $Sr_4Ce_2O_7$ by solid state reaction method have been subjected to Fourier transform infrared studies, which are used to analyze qualitatively the presence of functional group in the powder. The FTIR spectrums of powders were recorded using IR affinity-1 made by Shimadzu FTIR Spectrometer by KBr pellet technique. The FTIR spectrum of the $Sr_4Ce_2O_7$ is shown in Fig. 4. The peaks at 3700 cm⁻¹ are assigned to water molecules that may be present due to absorption of moisture. 3450 and 1110 cm⁻¹ is assigned to the hydrogen bonding in water and impurities, usually present in KBr respectively. The absorption peaks at 1770, 1450, 1022, 856, 705 and 699 cm⁻¹ were assigned to stretching characteristics of $SrCO_3$ [4]. The absorption peak between 600-300 cm⁻¹ is assigned to the metal oxide frequency band.



3.4 Raman spectrometry

Raman spectra are used as a complement of FTIR spectra for studying phase and structure of $Sr_4Ce_2O_7$. The Raman shift at 704 cm⁻¹ is assigned to symmetric stretching mode of $SrCO_3$ which coincide well with the IR features. The shift at 553 cm⁻¹ is attributed to antisymmetric bending vibration. Two strong Raman shift at 286 and 387 cm⁻¹ are detected, which can be attributed to the stretching modes of the Ce-O₂ and Ce-O₁ of CeO₆ octahedra in $Sr_4Ce_2O_7$ respectively. So the contribution of Ce-O₂ bonds increases corresponding with Ce-O₁ bonds to induce the charge transfer [4], which related to the luminescence of this material.





3.5 Luminescent properties

Fig. 5 shows the excitation spectra of $Sr_4Ce_2O_7$ blue phosphor. The excitation spectra consist of two bands observed at low and high wavelength; respectively. The luminescence of $Sr_4Ce_2O_7$ is thought to originate from a legend-to-metal charge transfer. The emission spectrum excited by 320 nm as shown in fig. 6. The spectrum shows broad band in the region 300 - 700 nm with a peak around 474 nm. The emission band can be assigned to transitions of Ce³⁺ ions.

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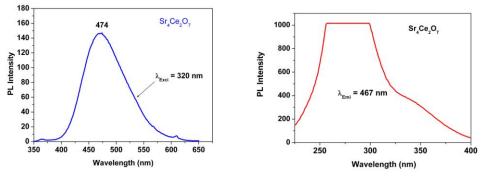


Fig.6: Excitation spectrum of Sr₄Ce₂O₇ F

Fig. 7: Emission spectrum of Sr₄Ce₂O₇

4 CONCLUSIONS

- A new Strontium Cerium Oxide Sr₄Ce₂O₇ was successfully synthesized by solid state reaction method.
- The XRD patterns of the powders revealed that the structure of Sr₄Ce₂O₇ is Orthorhombic,
- The emission spectra of Sr₄Ce₂O₇ phosphor was observed under excitation 320 nm, phosphor shows broad emission from 350 – 650 peaking at 474nm.
- The as synthesized Sr₄Ce₂O₇ phosphor with strong blue- white emitting could be a good candidate for advanced display devices.

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REFERENCES

- [1] Danielson E, Devenney M, Giaquinta D M, Golden J H, Haushalter, R C, McFarland E W, Poojary D M, Reaves C M Weinberg, A rare earth phosphor containing one-dimensional chains identified through combinatorial methods. 1998; Science, 279: 837-839.
- [2] Fu Shi-Liu, Yin Tao, and Chai Fei, Synthesis and characterization of Ca₂Sn_{1-x}Ce_xO₄ with blue luminescence originating from Ce⁴⁺ charge transfer transition, 2007 Chinese Physics,;16(10): 3129-3133.
- [3] Xiuzhen Xiao, Bing Yan. Sr₂CeO₄:Eu³⁺ and Sr₂CeO₄:5 mol% Eu³⁺, 3 mol% Dy³⁺ microphosphors: Wet chemistry synthesis from hybrid precursor and photoluminescence properties. 2008 J. of Phy. and Chem. of Solid,;69:1665-1668.
- [4] Chunxiang Zhang, Wenjun Jiang, Xujie Yang, Qiaofeng Han, Qingli Hao, Xin Wang. Synthesi and luminescent property of Sr₂CeO₄ phosphor via EDTA-complexing process, 2009; J. of Alloys and Compounds, 474: 287-291.
- [5] Yong Dong Jiang, Fuli Zhang and Christopher J. Summers, and Zhong Lin Wang. Synthesis and properties of Sr₂CeO₄ blue phosphor for field emission displays, 1997; Applied Physics Letters, 74:1677-1679.
- [6] Rahul Ghildiyal, Pallavi Page, and Murthy KVR. Photoluminescence and thermoluminescence properties of Sr₃Al₂O₆:Tb³⁺ 2008 Mat. *Research Bulletin.*; 43:353-360.
- [7] Niyaz Pravin Shaik, N V Poornachandra Rao, B. Subbarao, Murthy K V R, Photoluminescence study on Sr₂CeO₄ Nanophosphor, . 2011; *World J. of Chemistry*6(2):115-117.
- [8] Qiao Yanmin, Zhang Xinbo, Ye Xiao, Chen Yan, Guo Hai. Photoluminescent properties of Sr₂SiO₄: Eu³⁺ and Sr₂SiO₄:Eu²⁺ Phosphors prepared by solid state reaction method, 2009; Journal of Rare Earths, 27(2): 223-226.
- [9] Sue Jin Kim, Hyung II Won, Nersisyan Hayk, Chang Whan Won, Duk Young Jeon, and Artavazd G. Kirakosyan, Preparation and characterization of Sr₄Al₂O₇: Eu³⁺, Eu²⁺ phosphors, 2011; Material Science and Engineering B, 176(18):1521-1525.
- [10] Haiyan JIAO, Yuhuab WANG, Jiachi Zhang, Novel red phosphors for light emitting diodes: Sr_{2-y}Ce_{1-x}Ti_xO₄: yEu³⁺, 2009 Journal of Physics : Conference Series 152, 012089.
- [11] Zhang Chunxiang, Shi Jianshe, Yang Xujie, Lu Lude and Wang Xin, Preparation, characterization and luminescence of Sm³⁺ or Eu³⁺ doped Sr₂CeO₄ by a modified sol- gel method, 2010, *J. of Rare earths*, 28: 513-518.



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Author' biography with Photo

1)



1988. He is presently working as Associate professor in Department of Physics, S.V.S's Dadasaheb Rawal, College, Dondaicha, India. He worked on rare earth ions doped inorganic phosphor materials. His current research interest is in the synthesis and characterization of blue emitting phosphor for solid state lighting. He has published many research papers in various journals. He is a life member of Luminescence Society of India [LSI].

Pradip Z. Zambare obtained M.Sc. degree in Physics from M. S. University, Baroda, India in



K. D. Girase obtained M.Sc. degree in Physics from University of Poona, India in 1988. He 2) obtained Ph.D. degree in Physics from North Maharashtra University, Jalgaon India. He is presently working as vice Principal and associate professor in S.V.S's Dadasaheb Rawal, College, Dondaicha, India. He worked on crystal growth, thin film and rare earth ions doped inorganic phosphor materials for lamp industry. He has published many research papers in various journals. He is a life member of Luminescence Society of India [LSI].



O. H. Mahajan obtained M. Sc. Degree and Ph.D. degree in Applied Physics from 3) Faculty of Technology and Engineering, MS University of Baroda, Vaodara. He is presently working as Head, and Associate Professor in Physics Department, M.J. College Jalgaon, India. During his research carrier, he is involved in the Luminescence (For Lamps and Sources; Specific area of expertise: Lamp Phosphors Thermo luminescence). He has published many research papers in various journals. He is a Fellow of Luminescence

Society of India.

