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Structure, Raman and optical studies of $SrAl_2O_4$: Eu^{3+}, Dy^{3+} phosphor

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Abstract: Synthesis of $SrAl_2O_4:Eu^{3+};Dy^{3+}$ phosphor via low temperature combustion method has been reported.The crystal structure was verified through x-ray diffraction (XRD) characterization and it was revealed that the sample has crystallized into monoclinic structure with space group P 21.The Raman spectrum confirmed lattice structure via the characteristic active Raman modes. The phosphor was further characterized for confirmation of oxide formation via the Fourier transform infer-red (FTIR) spectroscopy, To study the optical properties, we employed UV-Vis spectroscopy and Photoluminescence studies. The lattice parameters calculated were a= 8.4470 Å, b = 8.8160 Å and c = 5.1630Å which confirms sample formation. The average crystallite size calculated using classical Scherer formula was found ≈ 40 nm. The Fourier Transform Infra-red (FTIR) spectra analysis witnessed sample formation via the appearance of signature absorption bands in the data. The UV-Vis spectral study reveals the synthesized phosphor is a wide band gap material and exhibit the bandgap of the order of ≈ 5.1eV. The photoluminescence study revealed the three glow peaks using excitation wave length λ_{ex}= 393nm, two corresponding to Eu^{3+} and one corresponding toDy^{3+}. The peaks are intense and lie in the visible range.

Keywords: Structure; FTIR; Phosphors; UV-Vis spectroscopy; Photoluminescence,.

INTRODUCTION-

Aluminates and their related structure has been a subject of immense interest from the perspective of technological application. The well known phosphor material is Zns but with short durability. To overcome this problem, the alkaline earth aluminates based on $REAl_2O_4$ [RE = Sr, Ba, Mg] were found as alternative which are chemically stable and exhibit durable luminescence and hence in the modern day luminescence devices, they are used as excellent matrix materials

ISSN: 2278-4632
Vol-10 Issue-6 No. 13 June 2020

for better luminescence [1].The luminescence can occur from the ultraviolet to the red region of the electromagnetic spectrum and strongly relies on the nature of host lattice. The photoluminescence of $RE Al_2O_4$ (*RE* = Ca, Sr, Ba) based phosphor aluminates show luminescence with a very long lifetime [2, 3]. Various techniques have been found to synthesize the alkali-earth aluminates family phosphor materials. The excellent luminescent property exhibiting phosphors include blue emitting $CaAl_2O_4$ (Eu^{2+}, Nd^{3+}) phosphor, green $SrAl_2O_4$ (Eu^{2+}, Dy^{3+}) phosphor and $BaAl_2O_4$ (Eu^{2+}, Dy^{3+}). Through the creation of exceptionally dense trapping levels, the doping of Dy^{3+} ions in the matrix of $SrAl_2O_4$: Eu enhances the luminescence property [6]. For long-afterglow characteristics, $SrAl_2O_4$ phosphor doped with Eu and Dy is technologically an indispensable material [7]. To improve the properties of these materials, a frequently used approach is to dope the parent lattice by suitable cations.These properties also depend to great extent on the synthesis method. The chemical and physical properties of inorganic micro/nano structured materials are dependent on their chemical composition,size,morphology, phase and also dimensionality.

In the present study, we report the synthesis of europium doped and dysprosium co-doped strontium aluminate phosphor by low temperature chemical combustion method. This report describes the structural characterization on the basis of X-ray diffraction,Ramanspectra, and optical nature of rare earth doped aluminates with general formula $Sr_{0.994}Eu_{0.005}Dy_{0.001}Al_2O_4$ ionic radii of Eu^{3+} and Dy^{3+} are 1.09 A and 0.908A respectively hence they are expected to replace Sr which has ionic radius of the order of 1.12A without any distortion in the structure.

2 EXPERIMENTATION

2.1 Synthesis

The polycrystalline $Sr_{0.994}Eu_{0.005}Dy_{0.001,}Al_2O_4$ aluminates phosphor was synthesized by solution combustion method. For the synthesis of aluminates phosphor, the raw materials were Strontium nitrate [Sr $(NO_3)_2$], aluminum nitrate [$Al(NO_3).H_2O$], and oxides like europium oxide [Eu_2O_3], and dysprosium oxide [Dy_2O_3] and Urea [$CO(NH_2)_2$]. The Eu_2O_3 and Dy_2O_3 oxides were dissolved in 2ml of concentrated HNO_3 to convert them into nitrates. The starting materials with stoichiometric amounts were mixed with urea and ground using mortar-pestle till a paste of the mixture was obtained. The paste was calcined at 610 ^oC after transferring it to the crucible. The solution catches fire due to fuel (urea) within seconds and a white foam (ash) is formed. The

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Vol-10 Issue-6 No. 13 June 2020

ashes of the solution were turned into fine powder via severe grinding. To remove the impurities for better emission, the final product was annealed at 1050^{o}C.

2.2 Characterizations

The polycrystalline $Sr_{0.994}Eu_{0.005}Dy_{0.001}Al_2O_4$aluminates phosphor was examined for crystal structure and phase formation by X-ray diffraction characterization over the angular range 2θ (10°-80°) using Bruker D8 Advance X-ray diffractometer with CuKα1 (1.5406Å) radiation. The instrument Frontier-Perkin-Elmer FTIR SP 10 STD was exploited to record FTIR spectrum to investigate the finger print region (400–1400cm^{-1}) as well as the functional group region (1400–4000cm^{-1}) of $Sr_{0.994}Eu_{0.005}Dy_{0.001}Al_2O_4$ phosphor by mixing the sample with potassium bromide (KBr).The Raman measurements were carried out using LABRAM - HR800 spectrometer. The sample was subjected to 488 nm radiations (2.53 eV) from an air-cooled Argon Laser and the spectra was recorded in the wavenumber range of 100-800cm^{-1}. For optical band gap study of the sample under investigation, we employed UV–Vis spectrometer (Perkin Elmer, Lambda 950 - USA). Edinburgh Instrument FLS920-s fluorescence spectrometer was used to record the photoluminescence spectra. All the characterizations were carried out at room temperature.

3 Results and Discussions

The $Sr_{0.994}Eu_{0.005}Dy_{0.001}Al_2O_4$ phosphor was synthesized by chemical combustion method successfully. For structural studies, the phosphor was characterized by XRD diffraction technique. For oxide formation, FTIR characterization technique was used, and PL characterization for luminescence studies. The XRD data collected in the angular range of 10^{o} – 80^{o} is displayed in Figure 1. Conveyed the single phase nature of the sample have acquired monoclinic structure with space group P 21/n.crystallite size was calculated using classical Debye-Scherer formula .t=kλ/cosθ and average particle size was estimated 40 nm,The lattice parameter were calculated to be a=**8.3958Å**,b= **8.8018Å**,and c=**5.353Å** and the density was found to be ρ be ρ ≈ 2.853 g/cm^{3}and volume = 383.7974 Å^3.

For further confirmation of the lattice formation, we carried out Raman spectral characterization. The Raman plot is displayed as Figure 2. In the Figure 2, the signature Raman mode appearing at 475 cm^{-1} is assigned to the O–Al–O group bending vibration closely associated to $[AlO_4]^{5-}$ tetrahedral. As this band is the signature vibrational mode, it is major

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Vol-10 Issue-6 No. 13 June 2020

factor the electron-phonon interactions.The Raman mode at 325cm^{-1} corresponds to the Eu-O tension vibration. Regarding the Ramanspectrum, the lack of polarization measurement and possible.

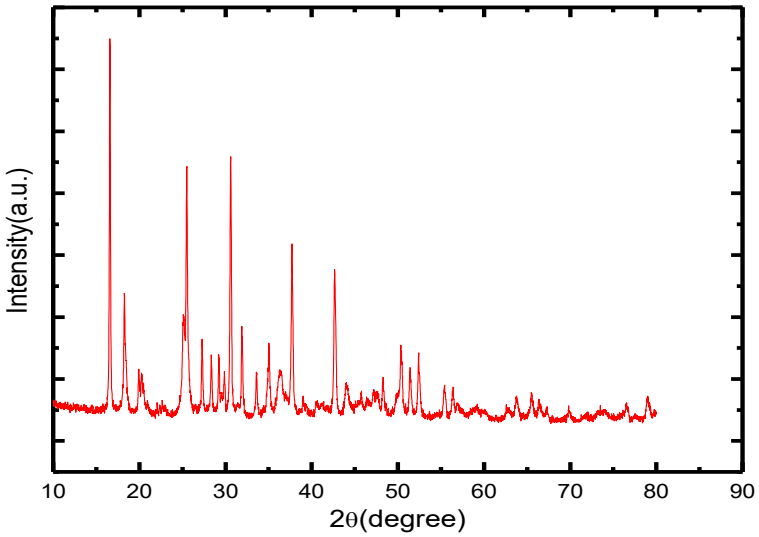
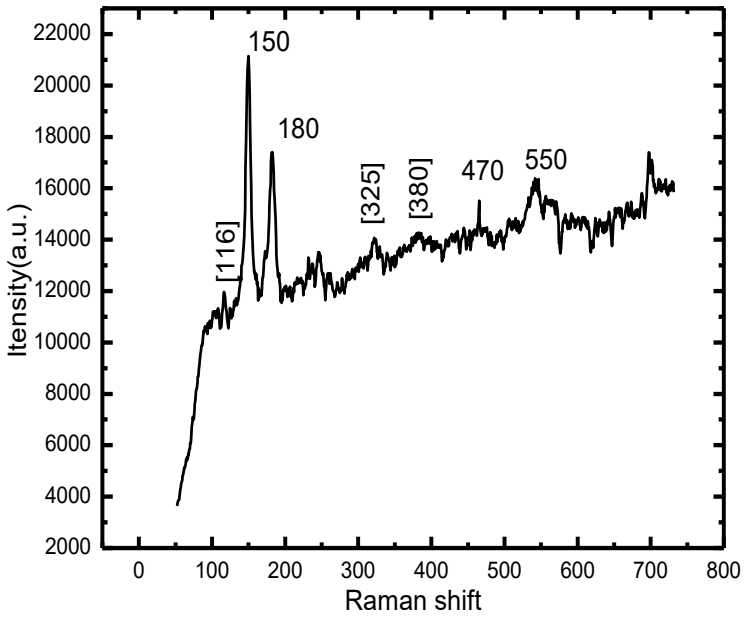


Figure 1: XDR diffractogram of $SrAl_2O_4$:Eu;Dy phosphor

Interference of atomic shifts makes it difficult to assign the Raman modes. For tetrahedral MO4 unit, we as approximation, assign modes appearing at frequency higher than 600cm^{-1}to Al-O stretching vibrations. The low frequency bands below 250cm^{-1} are assigned to tetrahedral vibrations or tilts. The assignment of intermediate is typical but by analogy with other



ISSN: 2278-4632
Vol-10 Issue-6 No. 13 June 2020

compounds, they can be assigned.

Figure 2: Raman Spectrum of $SrAl_2O_4$:Eu, Dy phosphor

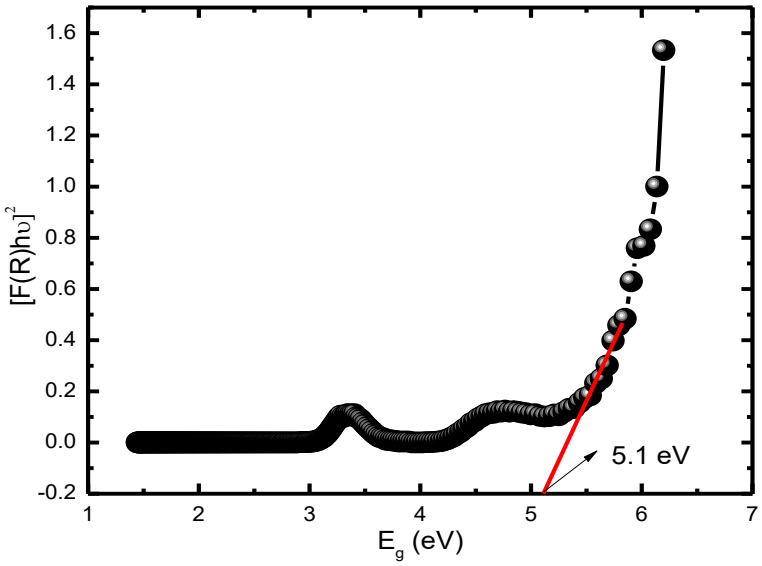


Figure 3: UV-Vis DRS spectrum of $SrAl_2O_4$:Eu;Dy phosphor

The band gap estimation of a material is important characterization to check its validity for the optical device applications. For this purpose, the UV-visible absorption spectra of $Eu^{3+}Dy^{3+}$doped $SrAl_2O_4$ phosphor was recorded using a diffuse reflection spectroscopy. The band gap was estimated by plotting the Kubelka-Munk-function $[F(R)h\nu]^n$ against energy [hν] where R is reflectance [13]. For the plot, the best fit was obtained for n = 1/2, displayed in Figure 3. An extrapolated straight line along the sharp edge of the curve intercepts the energy axis at a point which gives the estimation of the band gap and the value of optical band gap in the current case is 5.1eV [14].

The emission spectrum of $SrAl_2O_4$: Eu^{3+}, Dy^{3+} phosphor synthesized via solution combustion method with excitation wavelength 396nm is depicted in the Figure 4. The presence of a sharp emission peak at 594 nm, a characteristic feature of Eu^{3+} emission due to the $^5D_0 \rightarrow {}^7F_1$ transition, indicates the presence of Eu^{3+} ions. Attributes to the transition between $^4F_{9/2} \rightarrow {}^6F_{13/2}$energy levels [12, 15].and the excitation spectra of $SrAl_2O_4$: Eu^{3+}, Dy^{3+} shows a sharp excitation peak at 346,356 attributed to the transition $4f^7$-$4f^65d^1$.

ISSN: 2278-4632
Vol-10 Issue-6 No. 13 June 2020

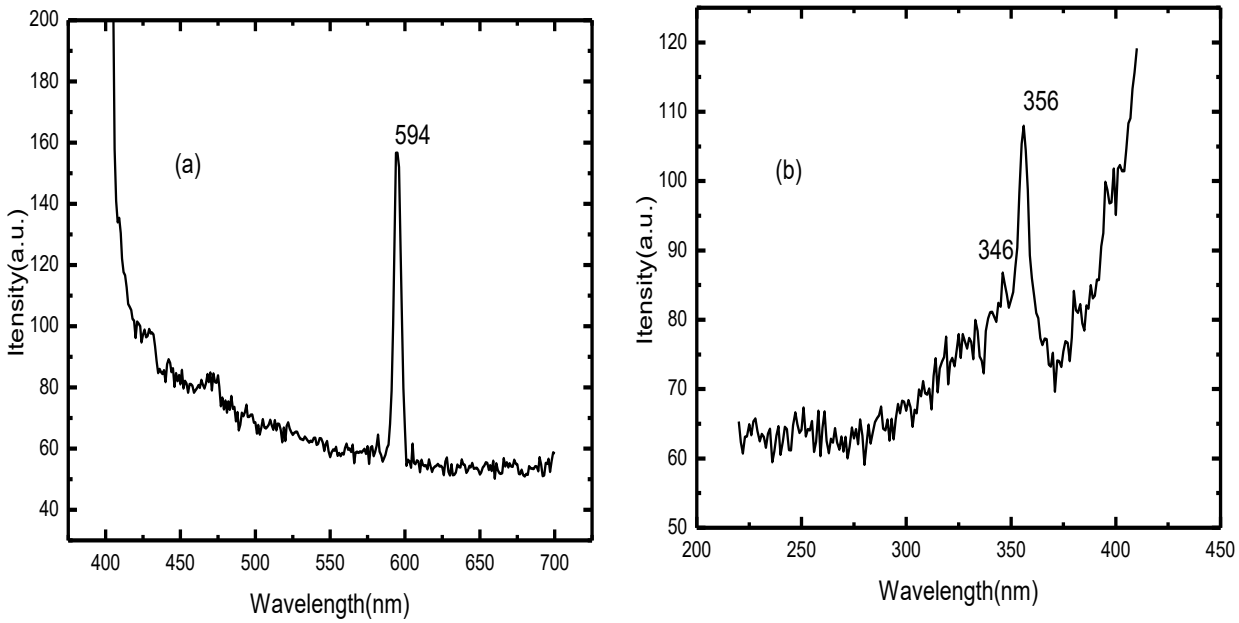


Figure 4: (a) emission spectra and (b) Excitation Spectra of SrAl2O4: Eu;Dy phosphor

In conclusion, the phosphor $SrAl_2O_4$: Eu^{3+}, Dy^{3+} was successfully prepared via solution combustion method. The type of phase and crystal structure was verified from XRD data revealing sample to be single phase of monoclinic structure having space group P 21/n. The Raman spectra analysis conveys the sample formation via the signature modes of vibration particularly the mode of vibration at 465cm-1. Optical bandgap study revealed the sample to be wide bandgap material with E_g= 5.1 eV. The PL emission spectra displayed characteristic glow peaks corresponding to Eu^{3+} and Dy^{3+} in the visible range.

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