

Colour tuning in Sm^{3+} activated and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-activated $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ phosphors for w-LED applications

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ABSTRACT

$\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (SBT) phosphors activated by Sm^{3+} and co-activated by Eu^{3+} have been synthesized in the current research using a straightforward solid-state reaction technique. X-ray Diffraction (XRD) confirms the sample's crystallinity. Scanning electron microscopy (SEM), energy dispersive x-ray spectroscopy (EDX), and elemental mapping have been used to analyse the morphology and elemental composition of the synthesized phosphor. Fourier-transform infrared spectroscopy (FT-IR) analysis has confirmed the vibrational characteristics of the as prepared phosphor. Photoluminescence studies show increase in luminescence intensity in case of $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-activated phosphors as compared with singly doped Eu^{3+} and Sm^{3+} activated SBT phosphors. Colour tunability was also observed from reddish-orange to pure red region when Sm^{3+} activated phosphors were co-activated with Eu^{3+} ions. These outcomes demonstrate that $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ phosphors activated or co-activated with Sm^{3+} and Eu^{3+} can be deployed in w-LEDs and other display devices applications.

1. Introduction

There is an increasing need for high-quality white light with certain colour temperatures and colour rendering qualities as lighting technology develops [1,2]. Researchers are striving to create phosphors that can accurately and faithfully produce a variety of hues. In numerous optoelectronic devices, such as lasers, LEDs, and photovoltaics, phosphors are crucial [3]. Energy sustainability as a whole is improved by the energy savings brought about by better phosphor materials. Researchers today strive to improve how well can phosphor materials transform light. This entails enhancing phosphors' quantum efficiency, reducing energy losses during the conversion process, and enhancing the spectral dispersion of produced light, among other things. When ultraviolet (UV) and visible light are utilised alternately, photochromism occurs, which is a reversible colour change. Due to their many uses, including photo switches, sensors, 3D homographic memories, and smart windows, photochromic materials have attracted a lot of attention. For this reason,

scientists are looking at new phosphor materials that could enhance the functionality of these devices and open up new possibilities. In contemporary lighting technology, phosphor-based LEDs, sometimes referred to as phosphor-converted LEDs (pc-LEDs), are essential. By employing a blue or UV LED chip to activate a phosphor substance, these LEDs are made to create white light. The phosphor emits light in the visible spectrum, usually at blue and yellow wavelengths that combine to produce white light. Nowadays, w-LEDs are created by coating the GaN blue LED chip with YAG: Ce^{3+} yellow phosphors [4]. In any case, the lack of red emission causes these w-LEDs to exhibit certain drawbacks including high correlated colour temperature (CCT) and low color-rendering index (CRI), which restrict their adaptability in a variety of applications [5]. We can create an acceptable colour temperature and CRI for residential, commercial, and architectural lighting by combining blue or UV LEDs with red-emitting phosphors. For lighting and display technology to provide a wide spectrum of colours, red-emitting phosphors are essential [6].

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To suit the changing needs of many industries, researchers are attempting to create new and superior red-emitting phosphor materials with improved efficiency, stability, and colour purity. Red-emitting phosphors are used by the entertainment and consumer electronics industries to create precise and vibrant colours in colour TVs, monitors, and projectors. Research into more efficient phosphor-based LEDs is motivated by the demand for energy-efficient lighting and decreased energy consumption. Numerous uses in lighting, displays, optoelectronics, and other fields are fuelling the demand for red-emitting phosphors [7,8].

The synthesis and photoluminescence (PL) behaviour of $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (SBT) phosphors co-doped with $\text{Sm}^{3+}/\text{Eu}^{3+}$ ions have been described in this current work. SBT lattices are bismuth layered structure ferroelectrics (BLSFs) with predictable $(\text{Bi}_2\text{O}_2)^{2+}$ layer intergrowth. Due to its unique structure, low phonon energy, high chemical stability, low cost, non-toxic and environmental friendly behaviour, the Aurivillius phase (SBT) materials have attracted a lot of attention of researchers [9,10]. These bismuths layered structures also possesses high Curie temperature, good dielectric, ferroelectric and piezoelectric properties, which made them highly beneficial in other applications like piezoelectric, pyroelectric and non-volatile random access memory etc. [11,12]. On the other hand, due to their potential utility in solid-state lasers, displays, and other technologies, rare earth (RE) ions have been extensively

exploited as luminescence centres throughout the past few decades. Yujian Wu et al., published their work on $\text{SrBi}_4\text{Ti}_4\text{O}_{15}:\text{Er}^{3+}$ emitting phosphor for anti-counterfeiting application [13]. Yuying Zhang et al. have studied the photoluminescence, electrical properties and electron band structure of $(\text{Ho}, \text{Yb})^{2+}$ co-doped $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ multifunctional ceramics [11]. Tong Wei et al. have published articles on high performance temperature sensing and optical heating of Tm^{3+} and Yb^{3+} co-doped $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ up-conversion luminescence nanoparticles and optical multi-functionalities of Er^{3+} and Yb^{3+} sensitized strontium bismuth titanate nanoparticles [10,14]. As per our knowledge, no work has been reported on $\text{Sm}^{3+}/\text{Eu}^{3+}$ SBT phosphors for w-LED applications. In this work, we have used samarium (Sm^{3+}) and europium (Eu^{3+}) RE ions as sensitizers/ activators to study the prospect of tuning orange-red emission to deep red in the as prepared SBT lattice for usage as red emitting component in w-LEDs and other lighting applications.

2. Experimental and characterization section

In this study, a solid-state reaction procedure under high temperature was used to generate a series of SBT: $x\text{Sm}^{3+}$ ($x = 0.5, 1.0, 1.5,$ and 2.0 mol%) and SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0$ mol%, $y = 1.0, 2.0, 3.0, 4.0,$ and 5.0 mol%) doped phosphors under air atmosphere. First, the raw materials SrCO_3 (98 %) (Thermo Fisher), TiO_2 (98 %) (Thermo Fisher),

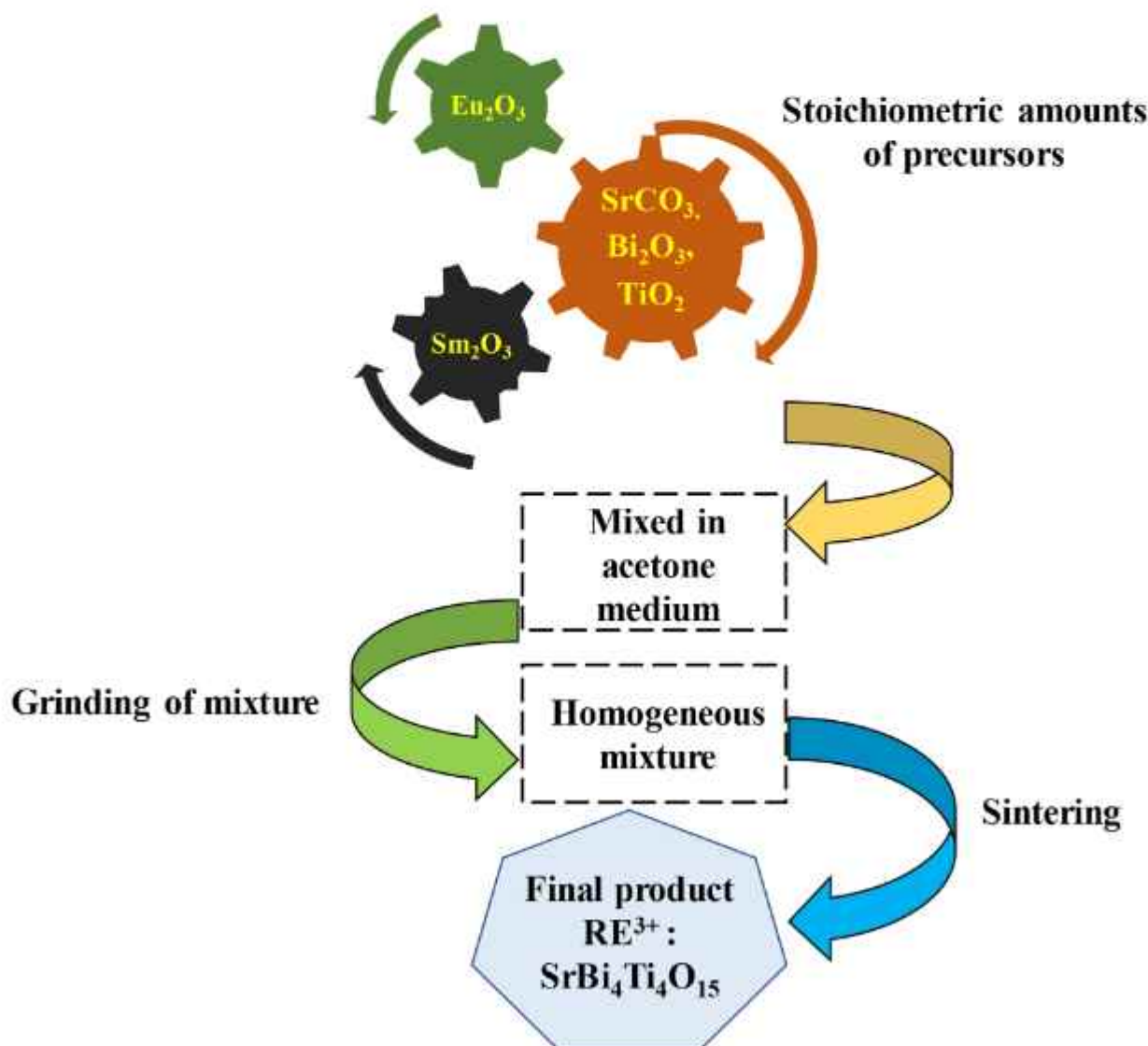


Fig. 1. Flow chart of various steps involved in the synthesis process of SBT phosphor.

Bi_2O_3 (99 %) (Loba Chemie), Sm_2O_3 (99 %) (Loba Chemie), and Eu_2O_3 (99 %) were weighed according to the stoichiometric proportion, placed in an agate mortar, and thoroughly ground for 60 min until a homogeneous mixture was obtained. The mix was then put into an alumina crucible and heated to a high temperature (900 °C) in a programmable furnace. Once the furnace naturally cooled to room temperature, the samples were ground again. Additional characterizations were performed using the obtained phosphor powder. Fig. 1 depicts all synthesis steps in detail.

The X-ray diffraction (XRD) patterns of the synthesised phosphors were captured using a Bruker D8 Advanced Powder X-ray diffractometer and a Cu-K radiation source ($\lambda=1.54 \text{ \AA}$, 40 kV, 40 mA) in the 2θ range of 20–80°. Using a JEOL 7610F Plus microscope, the morphological, elemental, and EDX analyses of the materials were performed. The Perkin Elmer Spectrum 2 was used to examine the phosphor's vibrational modes and functional groups. At room temperature, diffuse reflectance spectra of synthesised samples were captured using a UV-Vis spectrophotometer (Jasco V-770 Spectrophotometer). Using a xenon lamp as the excitation source, the samples' excitation and emission spectra were recorded using a Jasco FP-8300 Spectrofluorometer. PL decay curves were recorded on a Hameg Instruments HM 1507 digital oscilloscope running at 150 MHz. Using an ocean optics system, the temperature-dependent photoluminescence (TDPL) emission spectra were measured. All PL measurements, except for TDPL, were recorded at room temperature.

3. Results and discussion

3.1. XRD analysis

The initial evaluation of the purity of the as-prepared sample involves phase confirmation using XRD. Fig. 2 displays the XRD patterns of the pure, undoped $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ base material. All the diffraction peaks observed in the sample match the standard data for $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ (ICDD no. 43-0973). There are no indications of the presence of any other phases, suggesting that the obtained samples are composed of a single phase. $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ possesses an orthorhombic crystal structure with a space group denoted as $A21am$ (36). It has specific lattice parameters: $a = 5.4280 \text{ \AA}$, $b = 5.4380 \text{ \AA}$, $c = 40.9400 \text{ \AA}$, and angles α , β , and γ are all 90°.

Fig. 3 depicts the structural configuration of $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$, showcasing the positioning of Sr^{2+} , Bi^{3+} , and Ti^{4+} atoms within their respective coordination environments. $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ is classified as an Aurivillius-type material, characterized by a pseudo-perovskite structure. This classification was first established by Aurivillius in 1949. Aurivillius-type oxides follow a general formula $[\text{Bi}_2\text{O}_2][\text{A}_{n-1}\text{B}_n\text{O}_{3n+1}]$ ($n = 1, 2, 3, 4$), where $[\text{Bi}_2\text{O}_2]^{2+}$ layers intertwine with perovskite-like $[\text{A}_{n-1}\text{B}_n\text{O}_{3n+1}]^{2-}$ layers [15]. $\text{ABi}_4\text{Ti}_4\text{O}_{15}$ ($A = \text{Ba}, \text{Sr}, \text{and Pb}$) belong to the $n = 4$ series within the Aurivillius family. In the crystal structure of $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$, titanium occupies the MO_6 position in the perovskite layer [16]. The coordination of atoms is as follows: Sr is coordinated with 8 oxygen atoms, Bi(1) and Bi(2) with 6 oxygen atoms, while Bi(3) is coordinated with 5 oxygen atoms. Hence, based on the ionic radii and valency of the doped rare earth ion, it is anticipated that rare earth ions will substitute Sr and Bi sites in the crystal structure. As we know, these

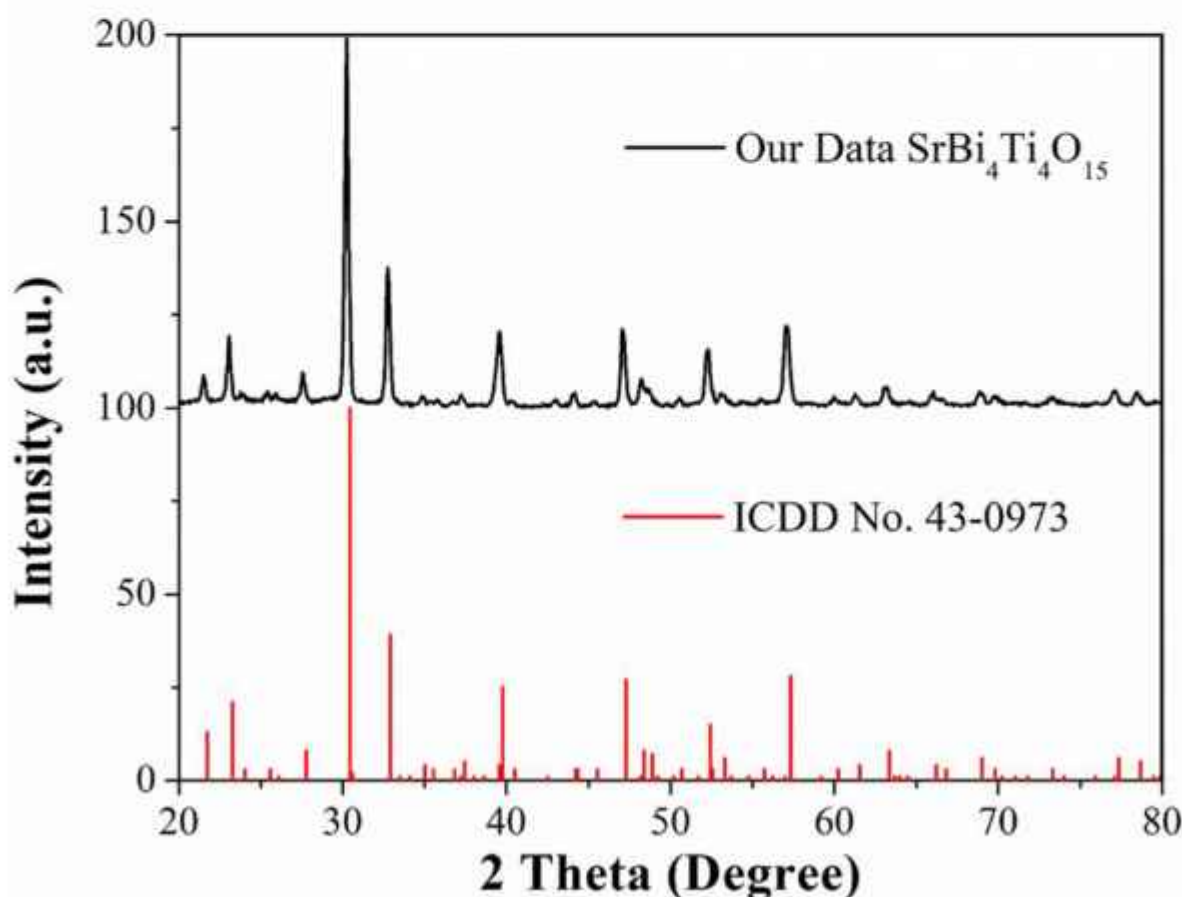


Fig. 2. XRD pattern of the $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$ and the reference data of ICDD no. 43-0973.

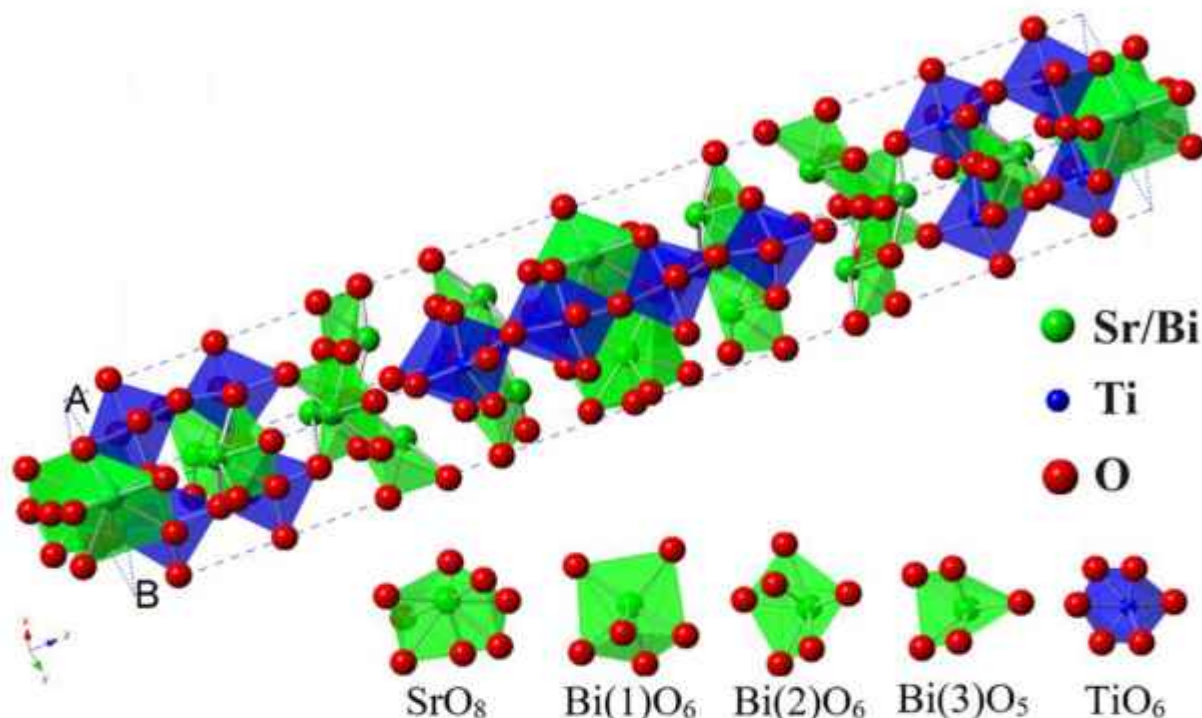


Fig. 3. Crystal structure of $\text{SrBi}_4\text{Ti}_4\text{O}_{15}$, emphasizing the coordination environment of Sr, Bi, Ti and O atoms.

lanthanides, exhibit unique electronic and magnetic properties that make them attractive for various technological applications. Substituting the RE ions in the crystal lattice of a phosphor can lead to changes in the emission wavelengths and color of the emitted light. This property is particularly important in applications like light-emitting diodes (LEDs) and display technologies.

3.2. SEM and EDX analysis

Sm^{3+} doped SBT phosphor and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped SBT phosphor's respective morphologies are shown in SEM micrographs presented in Fig 4(a & b). Small nanoparticles with variable orientation that resemble plates were observed [10]. It is thought that the intrinsic anisotropy of the bismuth layered perovskite structure is what that caused the plate-like grain development [14]. The range of prepared phosphors' particle sizes is 150–200 nm.

The EDX spectrum and elemental mapping of SBT:1mol% Sm^{3+} , 1mol% Eu^{3+} phosphor have been displayed in Fig. 5 in order to examine its composition and element distribution [17]. According to the sample's EDX spectrum, Eu^{3+} , Sm^{3+} , Sr^{3+} , Ti^{4+} , and Bi^{3+} elements make up the sample. These are the precursor substances that were used to create this phosphor. These elements are distributed equally according to the results of the elemental mapping, which further demonstrates the sample's accurate synthesis.

3.3. FT-IR analysis

The FT-IR spectrum at room temperature has been shown in Fig. 6 in the range of 400–4000 cm^{-1} . At 557 and 853 cm^{-1} , two strong absorption peaks were seen wherein, the former peak could be attributed to Ti-O bond stretching and the latter was attributed to Bi-O bond stretching vibration [18]. The creation of a layered-perovskite structure

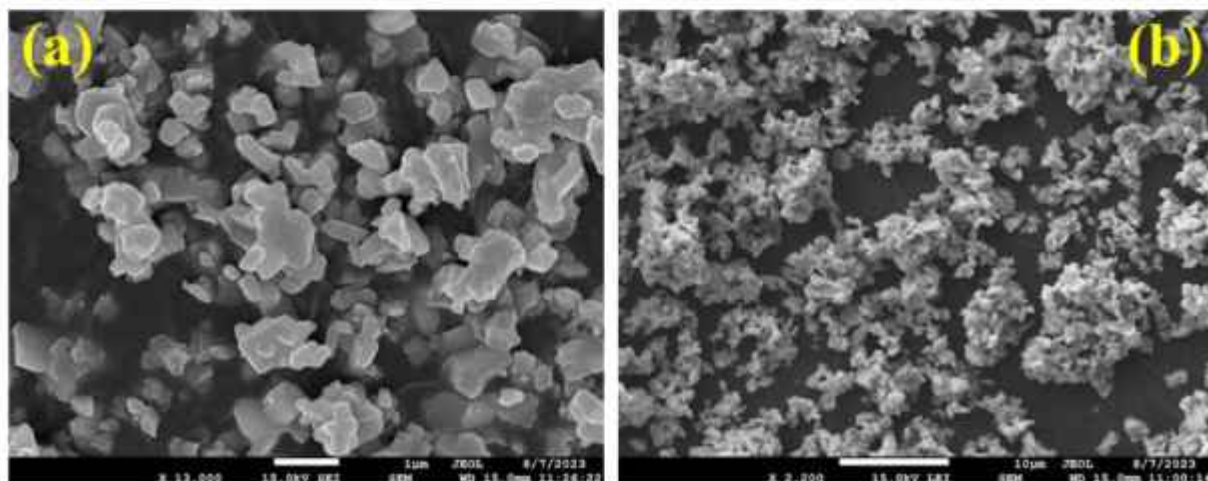


Fig. 4. SEM images of Sm^{3+} doped SBT phosphor and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped SBT phosphors.

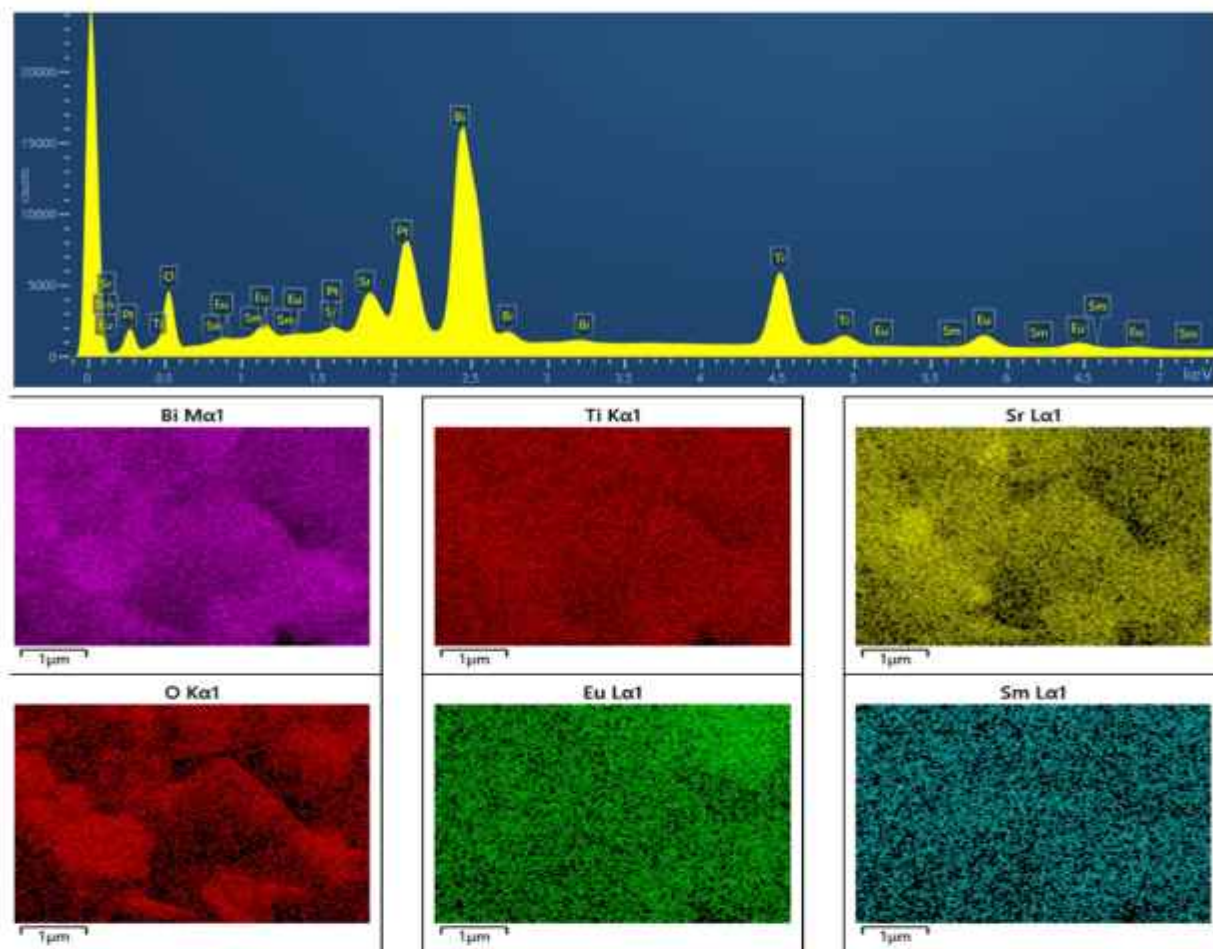


Fig. 5. EDX images and elemental mapping results of SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1 \text{ mol\%}$, $y = 1 \text{ mol\%}$) phosphor.

was also indicated by these two distinctive peaks. The symmetric and asymmetric stretching modes of the C—O bond are represented by the absorption peaks of CO_3^{2-} at 1050 and 1490 cm^{-1} , respectively [3]. There are two little humps at 2858 and 2931 cm^{-1} , which indicates hydrogen bonding [2]. The fundamental O—H stretching vibrations are represented by the absorption band at 3786 cm^{-1} .

3.4. Diffuse reflectance spectroscopy

The DRS spectra of SBT: $x\text{Sm}^{3+}$ ($x = 1.0 \text{ mol\%}$) and SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0 \text{ mol\%}$, $y = 1.0, 2.0, 3.0, 4.0,$ and 5.0 mol\%) phosphors are shown in Fig. 7. The charge transfer band transition between $\text{Sm}^{3+}-\text{O}^{2-}$ and $\text{Eu}^{3+}-\text{O}^{2-}$ is represented by the absorption band at 230 nm [19].

The Kubelka-Munk theory was applied to calculate the band gap E_g using the following equations [20,21]:

$$[F(R)hv]^{1/n} = A(h\nu - E_g) \quad (1)$$

$$[F(R)] = \frac{(1-R)^2}{2R} \quad (2)$$

Here, R is the coefficient of reflection and $F(R)$ represents the Kubelka-Munk function. Planck's constant is given by h , ν stands for frequency and c for speed of light. A and n are constants. The sample's characteristics will determine the value of n . For the direct allowed transitions, direct forbidden transitions, indirect allowed transitions, and indirect forbidden transitions, n is given by $1/2$, $1/3$, 2 , or 3 respectively. An evaluation of the E_g band gap can be made by plotting $[F(R)hv]^2$ vs photon energy $h\nu$. The E_g band gap for the SBT: 1 mol\% Sm^{3+} is 4.59 eV

and SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ co-activated phosphors is in the range of 4.70 – 4.78 eV , as illustrated in Fig. 7 (b&d). This band gap value is comparable to the previously reported band gap values of $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped host lattices. Mukesh K. Sahu et al. reported their work on synthesis and enhancement of photoluminescent properties in spherical shaped $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped NaCaPO_4 phosphor particles for w-LEDs and the band gaps for NaCaPO_4 : $x\text{Sm}^{3+}$ ($x = 1.0 \text{ mol\%}$) and NaCaPO_4 : $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = y = 1.0 \text{ mol\%}$) phosphors were estimated to be about 4.489 and 4.472 eV , respectively [22]. Dongcheng Jiang et al. reported the photoluminescence properties and energy transfer in the Sm^{3+} and Eu^{3+} co-doped $\text{Ca}_2\text{Bi}(\text{PO}_4)_3$ red phosphor and bandgap calculated for CBP: 0.08Sm^{3+} , 0.05Eu^{3+} is 4.04 eV [23]. These band gap values are favourable for the w-LED fabrication.

3.5. Photoluminescence (PL) investigation of single Sm^{3+} and co-doped $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped phosphors

Fig. 8(a) shows the PL excitation spectrum of SBT: $x\text{Sm}^{3+}$ ($x = 0.5 \text{ mol\%}$) phosphor. The PL excitation spectra of this sample are obtained by monitoring $\lambda_{em} = 600 \text{ nm}$. Several sharp peaks between 350 and 500 nm can be seen in the spectrum. The peaks at 365 , 380 , 409 , 422 , 440 , 465 and 481 correspond to transitions from ${}^6\text{H}_{5/2}$ to ${}^4\text{D}_{3/2}$, ${}^4\text{D}_{1/2}$, ${}^4\text{F}_{7/2}$, ${}^4\text{M}_{19/2}$, ${}^4\text{G}_{9/2}$, ${}^4\text{I}_{13/2}$ and ${}^4\text{I}_{11/2}$ respectively. The emission spectra of SBT: $x\text{Sm}^{3+}$ ($x = 0.5, 1.0, 1.5$ and 2.0 mol\%) phosphors under 409 nm excitation wavelength have been shown in Fig. 8(b). Four bands at 564 , 600 , 646 and 708 correspond to transitions ${}^6\text{G}_{5/2} \rightarrow {}^6\text{H}_{5/2}$, ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$, ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{9/2}$, and ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{11/2}$ respectively can be observed [24]. PL emission intensity seems to increase till 1.0 mol\% of Sm^{3+} ion concentration but decreases beyond that. This can be

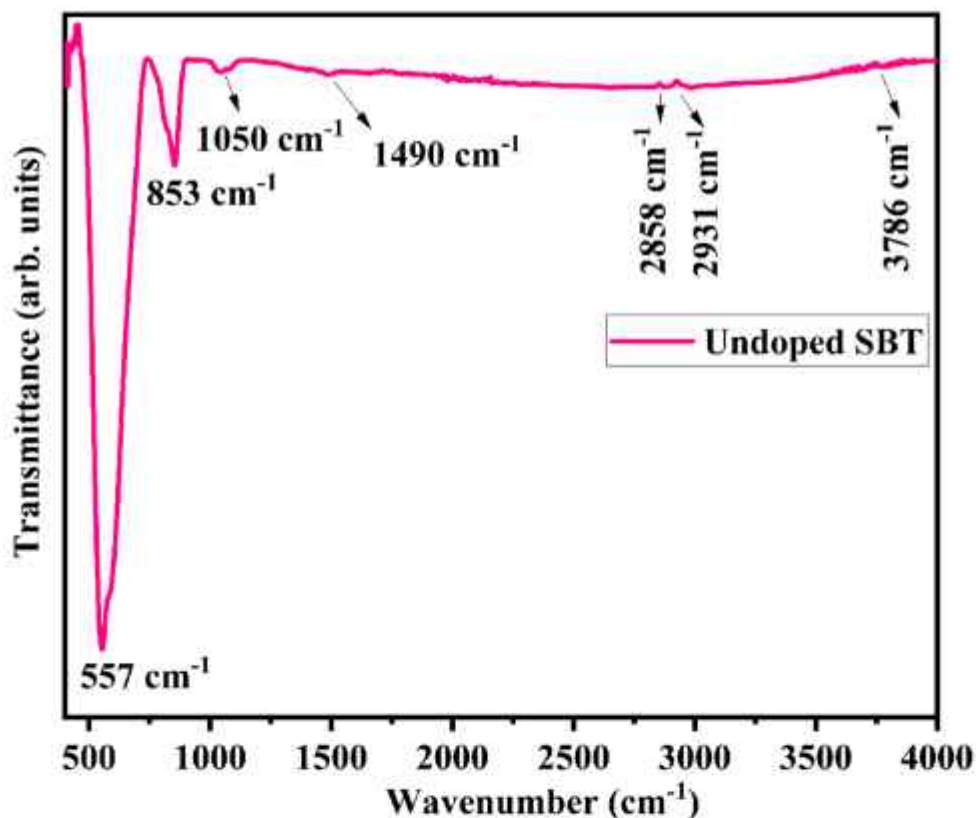


Fig. 6. FTIR spectra of un-doped SBT phosphor.

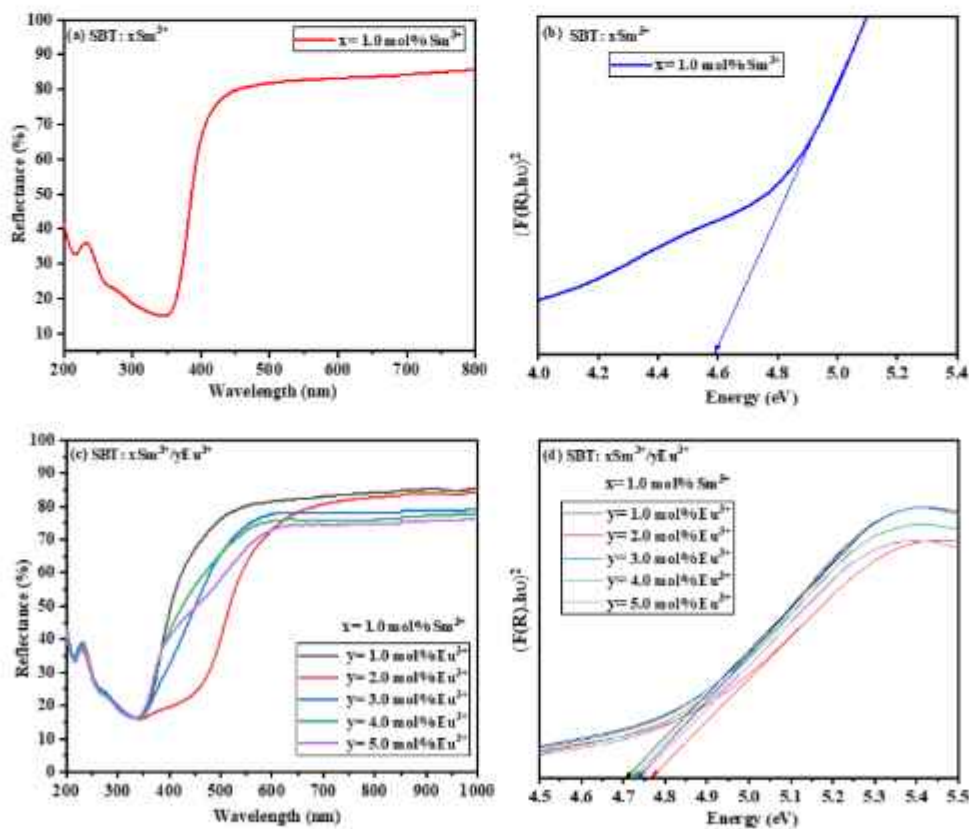


Fig. 7. DRS spectra of SBT: $x\text{Sm}^{3+}$ ($x = 1 \text{ mol\%}$) and SBT: $x\text{Sm}^{3+}/y\text{Eu}^{2+}$ ($x = 1 \text{ mol\%}$, $y = 1, 2, 3, 4$ and 5 mol\%) phosphor.

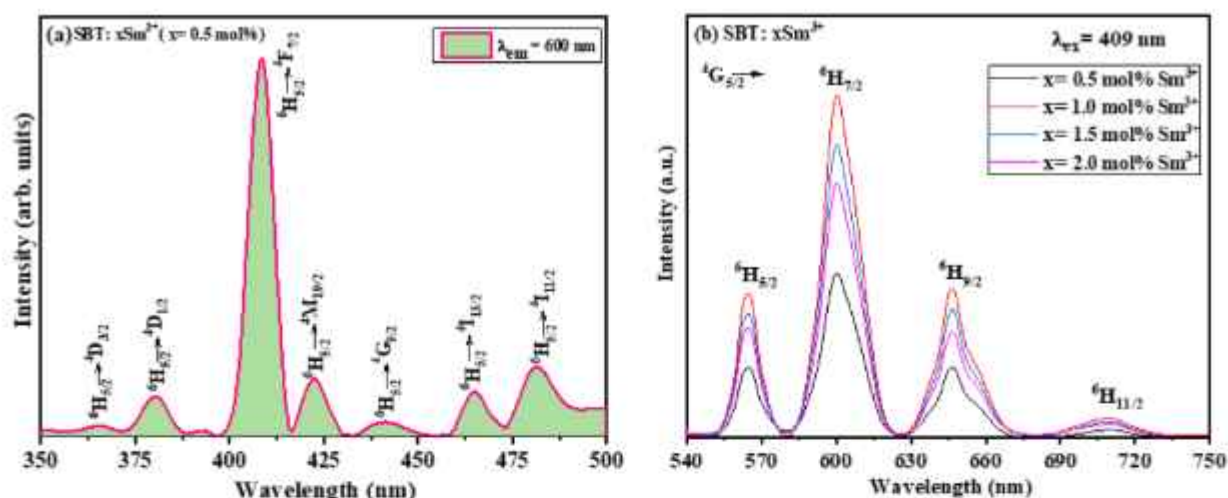


Fig. 8. PL excitation spectrum (a) of SBT: $x\text{Sm}^{3+}$ ($x = 0.5\text{mol}\%$) phosphor and emission spectra (b) of SBT: $x\text{Sm}^{3+}$ ($x = 0.5, 1.0, 1.5$ and $2.0\text{mol}\%$) phosphors.

attributed to the phenomenon of concentration quenching wherein the activator ions undergo non-radiative losses due to excessive ions in each other's vicinity.

Fig. 9 illustrates the excitation and emission spectra of SBT: $y\text{Eu}^{3+}$ ($y = 1\text{mol}\%$) phosphor. The excitation spectrum, recorded at $\lambda_{\text{em}} = 615\text{nm}$, comprises peaks at 353, 396 and 465 nm corresponding to the transitions from excited state ${}^7\text{F}_0$ to the ${}^5\text{D}_4$, ${}^5\text{L}_6$ and ${}^5\text{D}_2$ emission states of Eu^{3+} respectively. The emission spectra were recorded in the wavelength range 560–760 nm on pumping the phosphor at three different excitation wavelengths $\lambda_{\text{ex}} = 353, 396$ and 465nm . Peaks at 593, 615, 652 and 703 nm owing to transitions ${}^5\text{D}_0 \rightarrow {}^7\text{F}_1$, ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$, ${}^5\text{D}_0 \rightarrow {}^7\text{F}_3$, and ${}^5\text{D}_0 \rightarrow {}^7\text{F}_4$ respectively were observed [25]. Out of these peaks, the one at 615 nm was the most intense and the highest emission intensity was observed at $\lambda_{\text{ex}} = 353\text{nm}$ wavelength.

Fig. 10 (a, b & c) illustrates the comparison of emission spectra of singly Eu^{3+} doped SBT: $y\text{Eu}^{3+}$ ($y = 1.0\text{mol}\%$) and co-doped SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0\text{mol}\%$, $1.0\text{mol}\%$) phosphors under three different excitations, $\lambda_{\text{ex}} = 353, 396$ and 465nm respectively. Higher luminescence intensity for $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-activated phosphors can be seen as compared to single Eu^{3+} ion activated phosphors. This shows that energy transfer takes place from Sm^{3+} to Eu^{3+} ions due to which emission intensity increases multiple times. Co-doping with an activator ion increases the absorption cross-section and subsequently enhances luminescence efficiency. Since there is a very small energy difference between the energy level ${}^4\text{G}_{5/2}$ of the Sm^{3+} ion and the ${}^5\text{D}_0$ level of the Eu^{3+} ion, Sm^{3+} act as an efficient sensitizer and transfers its energy to Eu^{3+} ions which in results in enhanced luminescence. Energy transfer between Sm^{3+} and Eu^{3+} ions can tune the luminescent colour of the

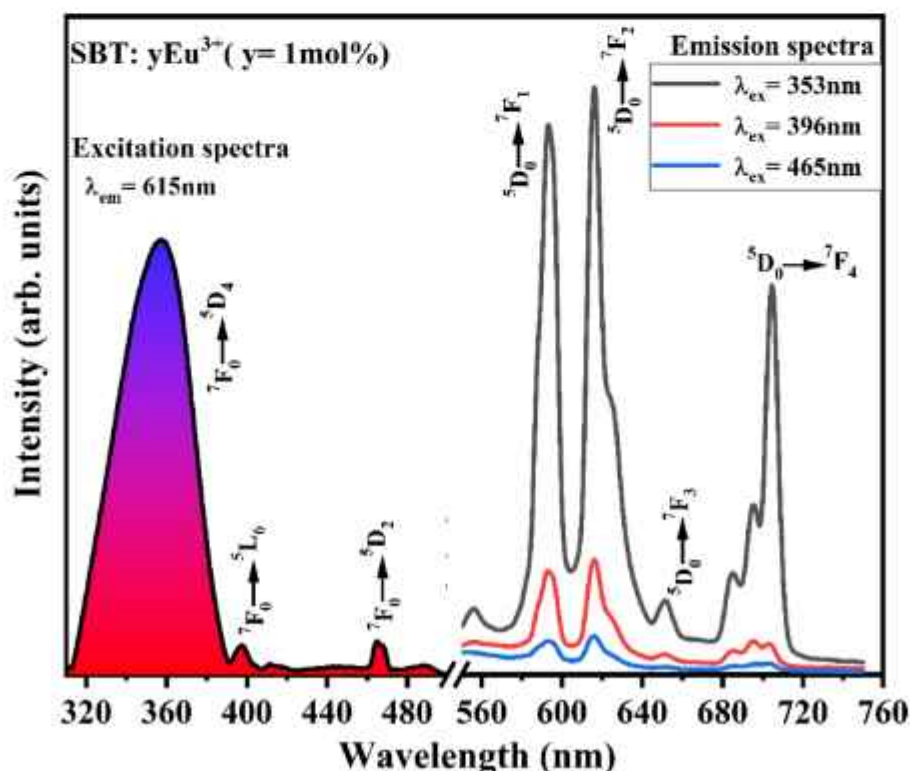


Fig. 9. Excitation and emission spectra of SBT: $y\text{Eu}^{3+}$ ($y = 1\text{mol}\%$) phosphor.

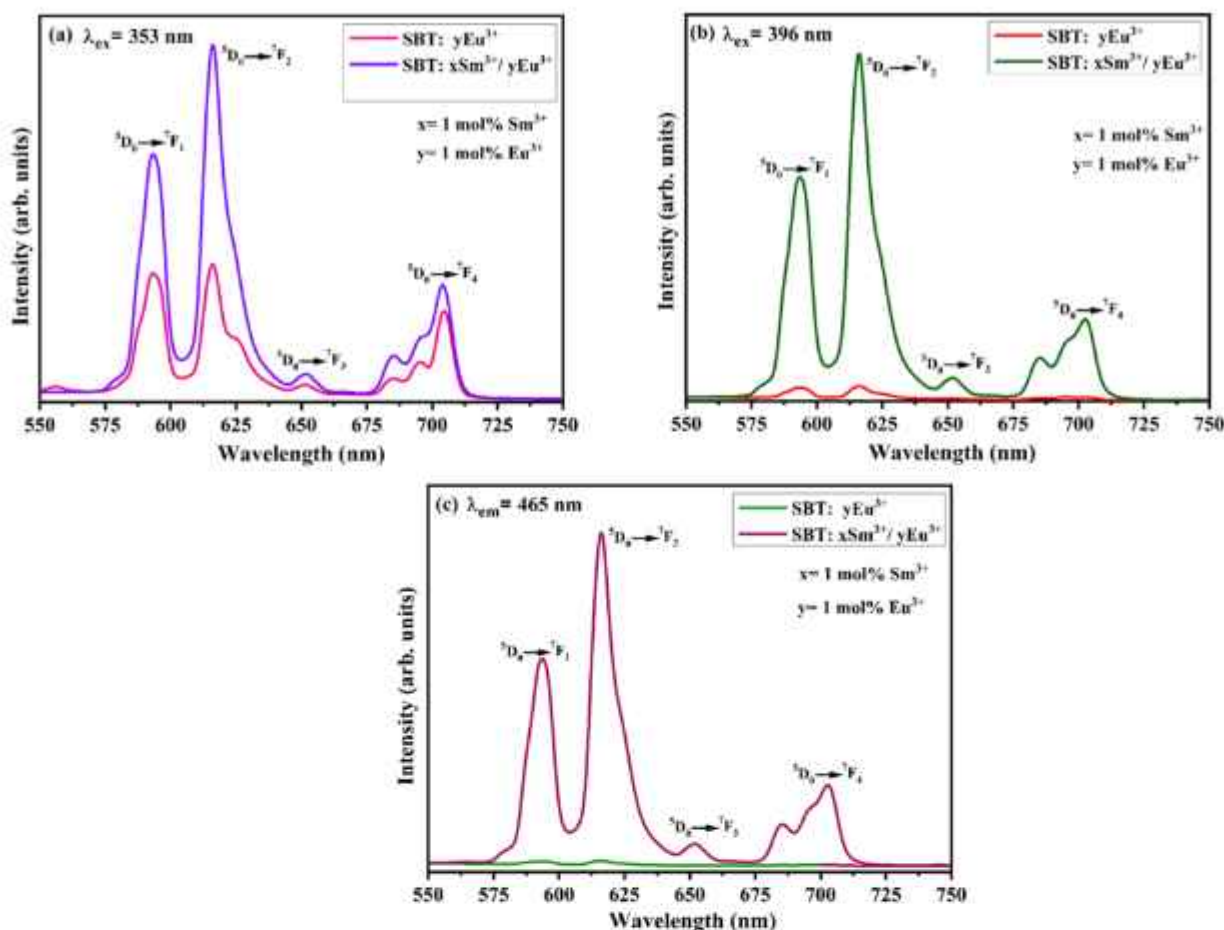


Fig. 10. (a, b & c). Comparison of emission spectra of SBT: $y\text{Eu}^{3+}$ ($y = 1.0$ mol%) and co-doped SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0$ mol%, 1.0 mol%) phosphors.

phosphor. This is especially important in LED applications where different light sources or applications require specific colours. The combination of Sm^{3+} and Eu^{3+} ions helps achieve better color rendering index (CRI) in LED lighting. The high color rendering index makes the illuminated object look natural and vibrant. The electron exchange process between Sm^{3+} and Eu^{3+} ions can achieve energy conversion, enabling the LED to emit increasingly brighter light.

Emission spectra of SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1$ mol%, $y = 1, 2, 3, 4$ and 5 mol%) have been shown in Fig. 11(a, b, c & d). The spectra were recorded at four different excitation wavelengths 353, 396, 409 and 465 nm respectively. The spectra comprise of several peaks at positions 593, 615, 652 and 703 nm owing to transitions ${}^3\text{D}_0 \rightarrow {}^7\text{F}_1$, ${}^7\text{F}_2$, ${}^7\text{F}_3$, and ${}^7\text{F}_4$ respectively. It can be clearly seen that increasing Eu^{3+} concentration results in increase in emission intensity up to 2 mol% of Eu^{3+} ions. After that, emission intensity decreases gradually. That means 1.0 mol% of Sm^{3+} ion concentration in singly Sm^{3+} activated phosphors and 2 mol% of Eu^{3+} ion concentration in co-activated phosphors is the optimum concentration of RE ions for this particular host lattice. Fig. 12 shows the energy level diagram showing various transitions exhibited by Sm^{3+} and Eu^{3+} ions [26–28].

Dexter's theory was applied along with Reisfeld's approximation, to check the type of multipolar interaction by using the given equation -

$$\frac{I_{50}}{I_{5U}} \propto C^{n/3}$$

where I_{50} and I_{5U} are the luminescence intensities of the SBT: $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped phosphor with the absence and presence of Eu^{3+} ions respectively. C is the sum of the concentration of Sm^{3+} and Eu^{3+} ; $n = 6, 8, 10$ represent dipole-dipole ($d-d$), dipole-quadrupole ($d-q$), and

quadrupole-quadrupole ($q-q$) interactions, respectively. Fig. 13 shows the linear regression between I_{50}/I_{5U} versus $C^{n/3}$. The best linear fit was observed for $n = 8$, means the type of interaction involved here is dipole-quadrupole interaction.

3.6. Quantum yield

To assess the quantitative emission performance of the phosphor the quantum yield (QY) measurement was done using Horiba QuantaPhi-2 integrating sphere. The scattered 396 nm excitation spectra (Rayleigh Spectra) were recorded for both the blank cup (R_b), and the optimized sample (SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$, $x = 1.0$ mol%, $y = 2$ mol%) (R_s). Integration of instrument corrected Rayleigh intensities was performed from 386 nm to 406 nm as shown in Fig. 14(a).

Thereafter, the fluorescence emission for blank and sample were measured and integration of the emission signals E_b (blank) and E_s (sample) was performed from 550 nm to 750 nm as shown in Fig. 14(b).

The quantum Yield (Φ) is estimated using the following relation:

$$(\Phi) = 100\% \times \frac{E_s - E_b}{R_b - R_s}$$

Since the integration time constant used to collect the E_b and E_s was 8 time longer than for R_b and R_s during the measurement so the difference $E_s - E_b$ is divided by 8 in the equations.

The QY of the optimal sample SBT: 1 mol% Sm^{3+} , 2 mol% Eu^{3+} is calculated as 58.9 which is relatively higher than the previous literature values reported for $\text{Sr}_2\text{Y}(\text{BO}_3)_2:0.07\text{Sm}^{3+}, 0.15\text{Eu}^{3+}$ (38.7 %) [29], $\text{Sr}_2\text{Y}_2\text{W}_4\text{O}_{24}:0.025\text{Sm}^{3+}/0.05\text{mol\% Eu}^{3+}$ (51.2 %) [30], $\text{NaLa}_{0.65}\text{MgWO}_6:0.05\text{Sm}^{3+}/0.3\text{Eu}^{3+}$ (48 %) [31] phosphors. This high

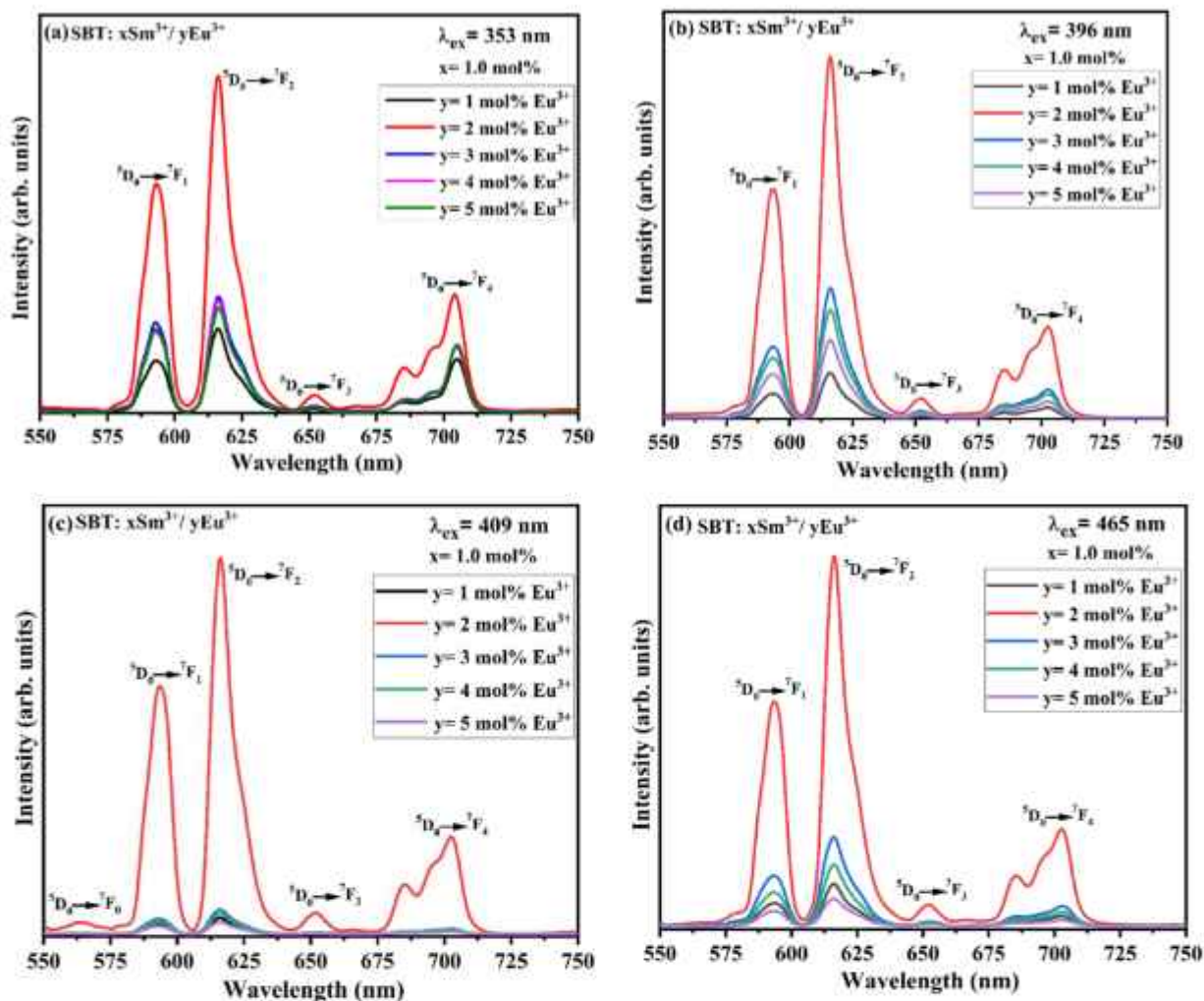


Fig. 11. (a, b, c & d). Eu^{2+} concentration dependent emission spectra of $\text{SBT}: x\text{Sm}^{3+}/y\text{Eu}^{2+}$ ($x = 1\text{mol}\%$, $y = 1, 2, 3, 4$ and $5\text{mol}\%$).

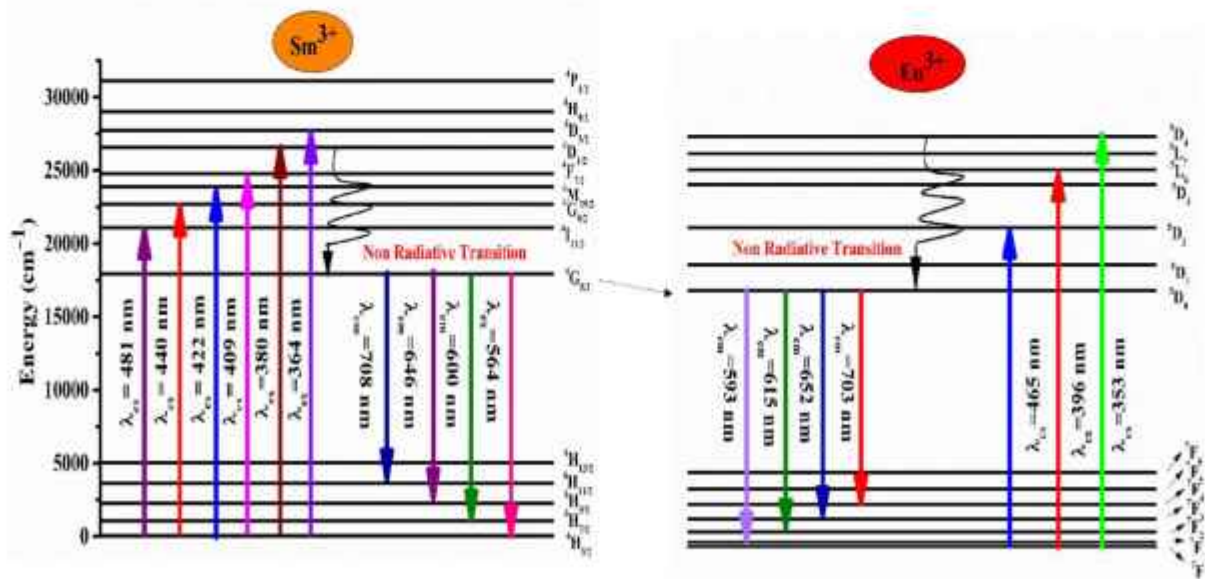


Fig. 12. Energy level diagram showing various transitions exhibited by Sm^{3+} and Eu^{3+} ions.

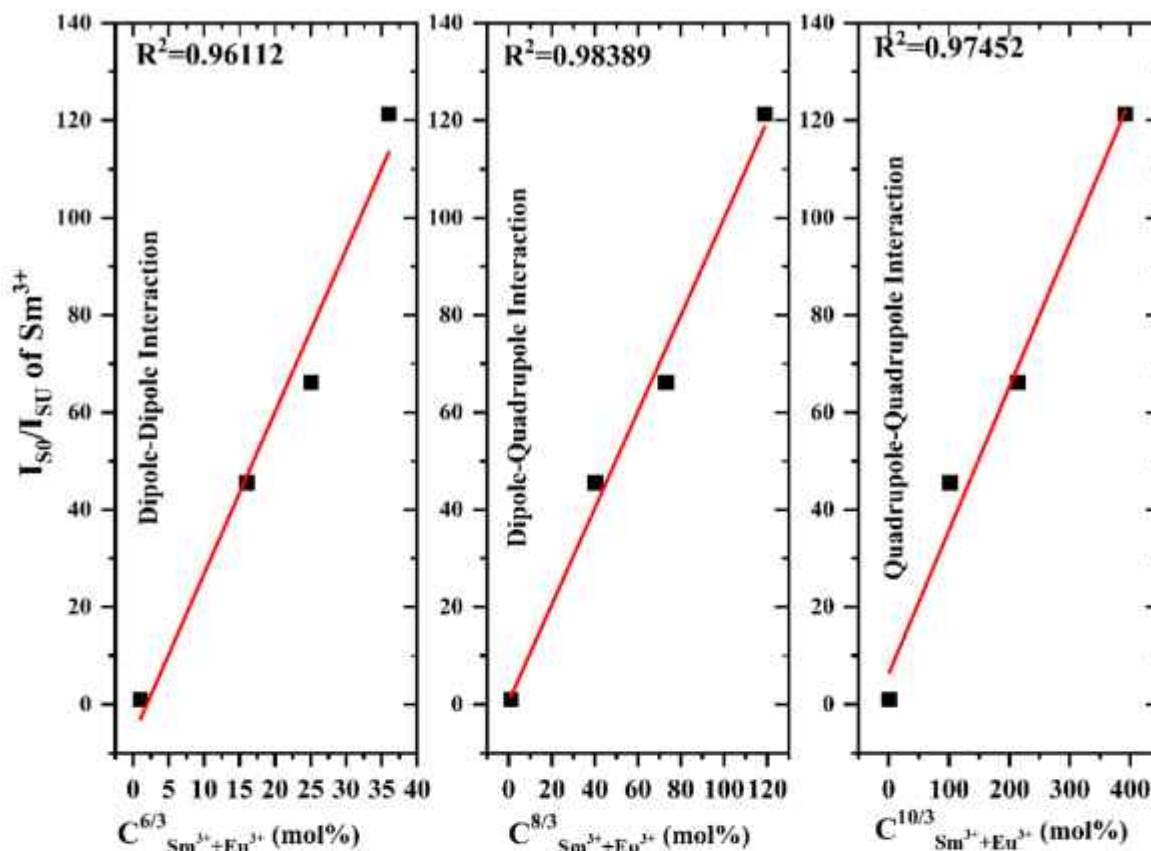


Fig. 13. Variation of I_{S0}/I_{SU} of Sm^{3+} ion w.r.t. $C^{6/3}$, $C^{8/3}$ and $C^{10/3}$ for SBT: xSm^{3+}/yEu^{3+} ($x = 1.0\text{mol}\%$, $y = 1, 2, 3, 4$ and $5\text{mol}\%$) co-doped phosphors under 409 nm excitation wavelength.

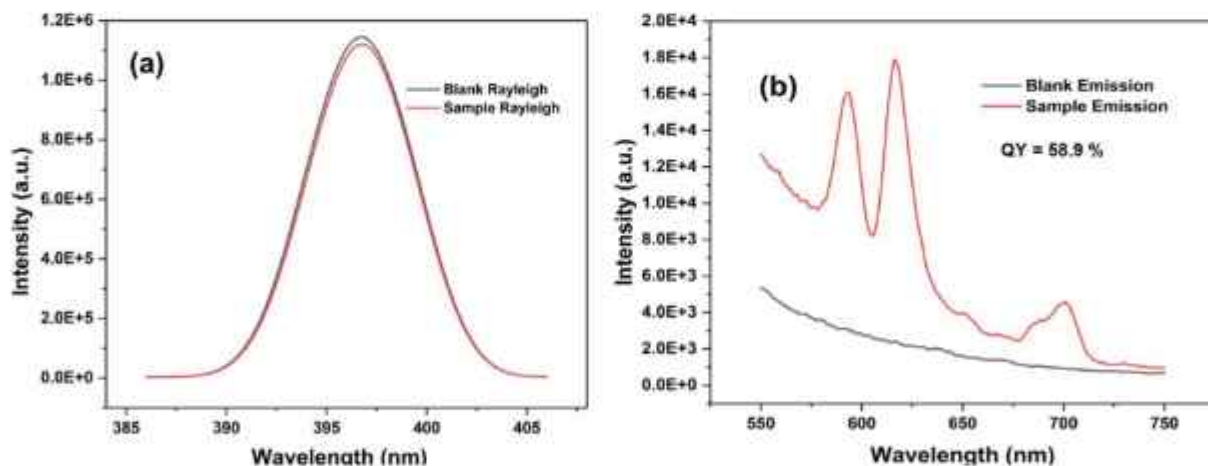


Fig. 14. Determination of quantum yield for SBT: xSm^{3+}/yEu^{3+} ($x = 1.0\text{mol}\%$, $y = 2\text{mol}\%$) phosphor.

QY result allows us to consider the use of the above phosphor in the application of photonic devices.

3.7. PL decay curves

Fig. 15 shows the SBT: xSm^{3+}/yEu^{3+} co-doped phosphors' normalised PL decay curves at 409 nm excitation wavelength. A double-exponential function using the following formula was used to fit the decay curves [32]:

$$I = I_0 + B_1 \exp\left(\frac{-t}{\tau_1}\right) + B_2 \exp\left(\frac{-t}{\tau_2}\right) \quad (3)$$

Where I_0 and I represent the intensities at time 0 and after t seconds, respectively; B_1 and B_2 are the fitting constants and τ_1 and τ_2 , respectively stand for the fast and slow decay time components. The average phosphor lifespan is calculated using the relation shown below [33]:

$$\tau_{avg} = \frac{A_1 \tau_1^2 + A_2 \tau_2^2}{A_1 \tau_1 + A_2 \tau_2} \quad (4)$$

The calculated lifetime values of the co-doped phosphors are listed in Table 1. The lifetimes values decrease with increasing Eu^{3+} concentration thereby implying an effective energy transfer from Sm^{3+} to Eu^{3+} ions [34].

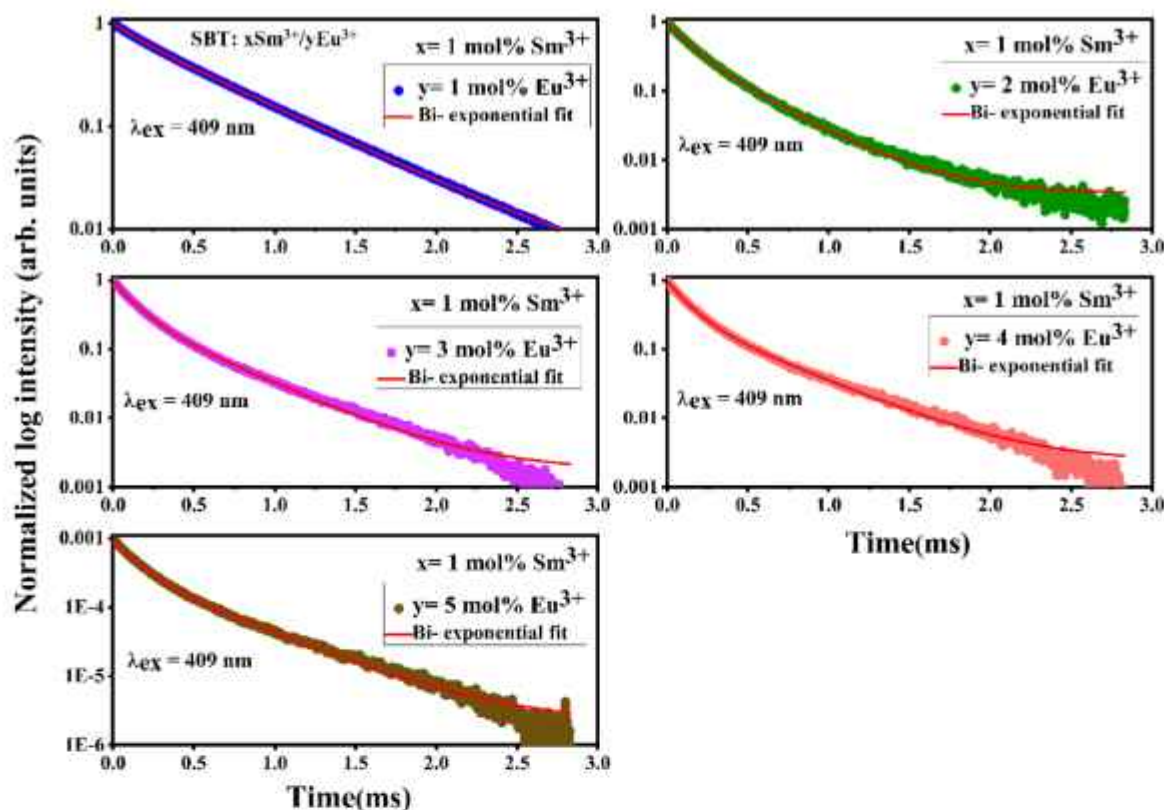


Fig. 15. PL decay curves of SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ co-doped phosphors at 409 nm excitation wavelength.

Table 1

Lifetime decay values of SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ co-doped phosphors.

Sample ID ($x\text{Sm}^{3+}/y\text{Eu}^{3+}$ series)	Lifetime values (ms)
$x = 1.0\text{mol}\%, y = 1\text{mol}\%$	5.68
$x = 1.0\text{mol}\%, y = 2\text{mol}\%$	2.77
$x = 1.0\text{mol}\%, y = 3\text{mol}\%$	3.13
$x = 1.0\text{mol}\%, y = 4\text{mol}\%$	3.27
$x = 1.0\text{mol}\%, y = 5\text{mol}\%$	3.57

3.8. Colorimetric analysis

The optimal Sm^{3+} doped and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped SBT phosphors' chromaticity diagrams from the Commission Internationale de l'éclairage (CIE) are shown in Fig. 16. The emission spectra of Sm^{3+} doped phosphors that was recorded at excitation wavelength 409 nm and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped phosphors that were recorded at excitation wavelengths 353, 396, and 409 nm, respectively, were used to calculate CIE points. The CIE coordinates of optimized Sm^{3+} ion doped phosphor fall in orange-reddish region. From Fig. 16, we can find that CIE coordinates for $\text{Sm}^{3+}/\text{Eu}^{3+}$ ion co-doped SBT phosphors shifted from orange-reddish to deep red region. Thus, the CIE coordinates show a color tunability in $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-activated phosphors as compared to singly doped Sm^{3+} activated SBT phosphors. Color tunability allows users to adjust the lighting environment to their preferences and specific needs. This change is beneficial in creating different climates in residential, commercial and industrial areas. Adjustable LED lighting can be used to simulate natural day changes, support circadian rhythms and promote better sleep. In medical facilities, lighting can be adjusted according to the comfort and health of the patient [35]. This capability opens up new possibilities in various fields, enabling the customization of lighting and display technologies to meet specific requirements and enhance visual experiences.

Correlated colour temperature (CCT) is an additional important

parameter for illustrating the suitability and importance of RE^{3+} doped phosphor. The CCT represents the colour hue of a light source emitted from a phosphor when it is excited by an energy source. With the use of the empirical McCamy equation, the CCT values was computed [36]:

$$\text{CCT} = -449\pi^3 + 352\pi^2 - 6823.3\pi + 5520.33 \quad (5)$$

Where (x, y) is the CIE coordinate, (x_c, y_c) is the chromaticity epicentre with coordinates (0.336, 0.186), and (x_e, y_e) is the chromaticity epicentre, and $\pi = (x-x_e)/(y-y_e)$ is the inverse slope line. Table 2 contains a list of the calculated CCT values and CIE coordinates.

3.9. Temperature dependent PL (TDPL)

Temperature dependent PL is an essential characterisation technique to assess the thermal stability of the as prepared phosphor especially if such materials are being targeted for usage in w-LEDs and other lighting applications where operating temperatures are very high. Here, the temperature-dependent emission spectra of a $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0\text{mol}\%, y = 2\text{mol}\%$) co-doped SBT phosphor were recorded in the temperature range of 298 to 423 K at intervals of 25 K under 409 nm excitation (Fig. 17a). The spectra show that the thermal quenching effect causes the emission intensity to steadily decrease as temperature rises. The intensity measured at 298 K decreased by 72 percent at 398 K, showing that the synthesised co-doped phosphors had some thermal stability and might be used to create effective lighting and display systems. Additionally, the Arrhenius relation can be used to compute the activation energy (ΔE), which is a crucial quantity to determine the thermal stability of the phosphors [37,38].

$$I_T = \frac{I_0}{1 + C \exp\left(\frac{\Delta E}{k_B T}\right)} \quad (6)$$

Here, C is a constant and k_B is the Boltzmann's constant, I_0 is the PL

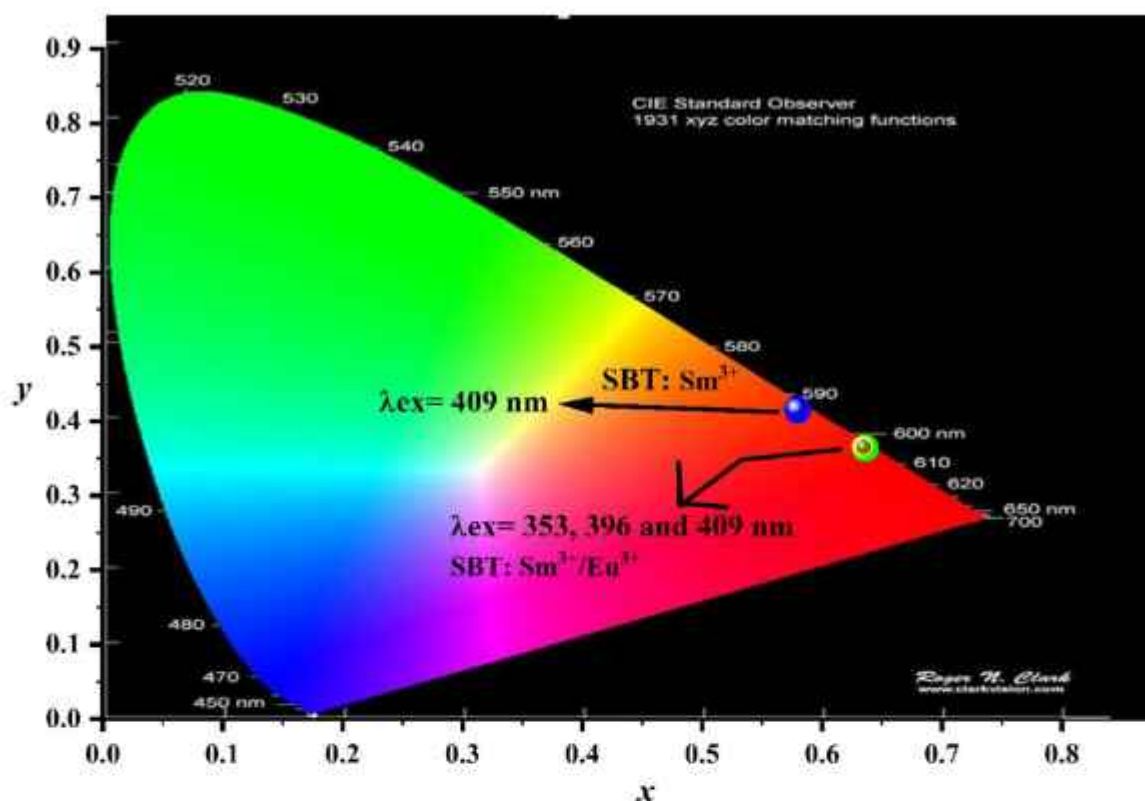


Fig. 16. CIE co-ordinates of optimal Sm^{3+} doped and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped SBT phosphors.

Table 2

Variations of CIE Coordinates and CCT values for optimized singly Sm^{3+} ion doped and co doped $\text{Sm}^{3+}/\text{Eu}^{3+}$ SBT phosphor.

Sample ID ($x\text{Sm}^{3+}/y\text{Eu}^{3+}$ series)	λ_{exc}	CIE Co-ordinates	CCT(K)
$x = 1.0\text{mol}\%$, $y = 0\text{ mol}\%$	409 nm	(0.579, 0.415)	1765
$x = 1.0\text{mol}\%$, $y = 2\text{ mol}\%$	353 nm	(0.635, 0.365)	2105
$x = 1.0\text{mol}\%$, $y = 2\text{ mol}\%$	396 nm	(0.636, 0.363)	2138
$x = 1.0\text{mol}\%$, $y = 2\text{ mol}\%$	409 nm	(0.634, 0.366)	2061

intensity at 298 K and I_T is the intensity at various testing temperatures. The slope of graph $\ln[(I_0/I_T) - 1]$ vs. $1/K_B T$ shown in Fig 17(b) can be used to determine the value of ΔE . As a result, ΔE calculated value is 0.385 eV which is relatively higher than the previously reported values for $\text{Ba}_2\text{MoTiO}_6:x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.5\text{ mol}\%$, $y = 4\text{ mol}\%$) ($\Delta E = 0.306$) [37], $\text{Sr}_2\text{Y}_2\text{W}_4\text{O}_{24}:x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 2\text{ mol}\%$, $y = 5\text{ mol}\%$) ($\Delta E = 0.306$) [30], $\text{Zn}_{0.99-y}\text{Nb}_2\text{O}_6:0.01\text{Sm}^{3+}, 0.25\text{Eu}^{3+}$ ($\Delta E = 0.235$) [39]. Higher activation energy generally corresponds to a slower reaction rate. Substances with higher activation energy can withstand higher temperatures without undergoing significant chemical changes because the thermal energy available at those temperatures may not be sufficient to overcome the activation energy barrier.

4. Conclusion

In conclusion, the current paper presents morphological and photoluminescent analysis of $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped SBT phosphors produced using a typical solid-state reaction approach. Studies have been done on the structural, optical, and thermal stability of as prepared phosphors. All phosphors have orthorhombic crystal structures, which is confirmed by the XRD investigation. FT-IR analysis gave information about the vibrational states and functional groups present in the lattice of the phosphors. From the UV-Vis-NIR absorption investigation, it was seen that the band gap of the phosphors changes after doping with rare earth

ions possibly as a result of the development of extra trap states in the conduction and valance band region. PL emission and excitation studies on singly doped Sm^{3+} SBT phosphors were conducted at 409 (${}^6\text{H}_{5/2} \rightarrow {}^4\text{F}_{7/2}$) nm excitation and 600 nm (${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$) emission wavelengths, respectively. PL excitation and emission investigations on $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-doped phosphors were also recorded at various excitation wavelengths ($\lambda_{\text{exc}} = 353, 396,$ and 409 nm). Energy transfer from Sm^{3+} to Eu^{3+} ions inside the host lattice was studied by comparing the emission spectra of Sm^{3+} and $\text{Sm}^{3+}/\text{Eu}^{3+}$ co-activated phosphors. Using temperature dependent PL analysis, the thermal stability of the phosphors was investigated in the temperature range of 298–423 K. After Eu^{3+} co-doping in Sm^{3+} doped phosphors, a considerable shift in the CIE co-ordinates from the reddish-orange to the pure red region was seen. As a result, our research supports the idea that Sm^{3+} and Eu^{3+} co-doped SBT phosphors, when used in the right proportions, can trigger efficient energy transfer phenomena and can be used as deep red emitting component of w-LEDs and in thermally stable lighting and display technologies.

CRediT authorship contribution statement

Pooja Rohilla: Writing – original draft, Visualization. Sheetal Kumari: Writing – review & editing, Software, Investigation. Ravita: Investigation, Formal analysis. Samarthya Diwakar: Visualization, Methodology. Rupesh Talewar: Investigation. Ankur Shandilya: Investigation. Kartika Maheshwari: Investigation. M. Venkateswarlu: Formal analysis. Aman Prasad: Validation, Supervision, Funding acquisition, Data curation. A.S. Rao: Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

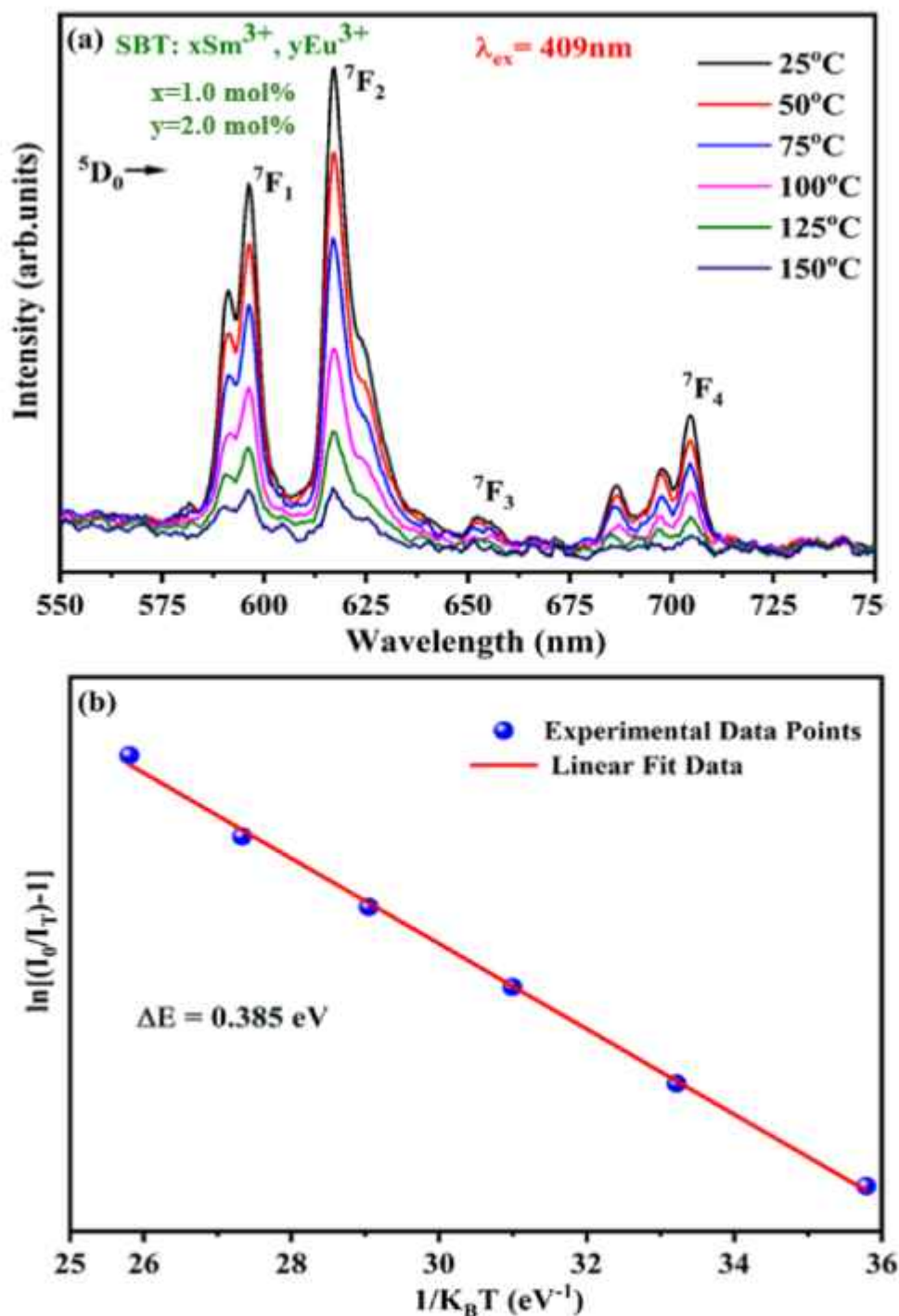


Fig. 17. (a). Temperature dependent PL spectra of SBT: $x\text{Sm}^{3+}/y\text{Eu}^{3+}$ ($x = 1.0\text{mol}\%$, $y = 2\text{mol}\%$) phosphor. (b). $\ln[(I_0/I_T) - 1]$ vs. $1/K_B T$ plot for activation energy.

Data availability

The data that support the findings of this study are available on request from the corresponding author Dr. Aman Prasad. The data are not publicly available due to restrictions, e.g. their containing information that could compromise the privacy of research participants.

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