

SYNTHESIS AND CHARACTERIZATION OF $\text{Sr}_{(1-x)}\text{TeO}_3:\text{xEu}^{3+}$ PHOSPHORS FOR POTENTIAL SOLID-STATE LIGHTING APPLICATIONS

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Abstract

The $\text{Sr}_{(1-x)}\text{TeO}_3:\text{xEu}_2\text{O}_3$ phosphors were synthesized using the conventional high-temperature solid-state reaction method at 800°C for 3 h in air atmosphere. The synthesized phosphors were characterized using XRD, FTIR, absorption spectra, photoluminescence, and X-ray-induced luminescence. Rigaku SmartLab-SE XRD diffractometers confirmed the crystalline nature of the phosphors. From the study, it was found that the structure of the sample was a mixture of SrTeO_3 and TeO_2 . The increased concentration of Eu_2O_3 seems to contribute to the enhancement or stabilization of SrTeO_3 phase. The UV-VIS-NIR spectrophotometer in reflection mode measured the absorption spectrum, revealing six peaks corresponding to energy transitions of Eu^{3+} ions at 394, 465, 536, 592, 2,207, and 2,070 nm. Photoluminescence analysis demonstrated the strongest luminescence intensity of Eu^{3+} ions at 614 nm (${}^7\text{F}_2$) under 465 nm excitation, resulting in an overall red emission in the CIE 1931 chromaticity. Combining these findings, $\text{Sr}_{(1-x)}\text{TeO}_3:\text{xEu}$ phosphors exhibit promising characteristics for solid-state lighting applications (SSL). The X-ray-induced luminescence shows an increase with increasing concentrations of Eu_2O_3 until 0.10%. The samples with the maximum intensity were compared with bismuth germanate oxide (BGO) crystals, which are used as radiation detectors, and were found to be as effective as 33.65% of the BGO crystals. The results demonstrate that the current samples could be a potential candidate for use in scintillator applications.

Keywords: Eu^{3+} ions; Phosphor; Photoluminescence

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Introduction

In the modern s era, has seen a critical search for novel technologies that can satisfy the demands of both display and illumination due to increasing awareness of energy conservation and environmental protection (Zhong *et al.*, 2018). Over the past few decades, white light-emitting diodes (WLEDs) have emerged as a promising solid-state lighting solution to replace conventional incandescent and fluorescent lamps. Their appeal lies in their energy-saving features, environmental friendliness, and extended lifespan (Wang *et al.*, 2016). Commercially produced WLEDs typically combine a blue InGaN-based LED with a yellow-emitting $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ (YAG: Ce^{3+}) phosphor. However, these WLEDs face challenges, such as a low color-rendering index (CRI) and a high correlated color temperature (CCT), primarily due to the absence of red emission in the visible spectrum. This divergence from natural light hampers their application in certain contexts. To address these challenges, the adoption of tri-color (RGB) phosphors has been proposed. These advanced WLEDs necessitate high-performance blue, green, and red phosphors capable of being excited by a near-UV LED chip (350 nm-420 nm) (Li *et al.*, 2018). The current study, focuses on identifying a red-emitting phosphor suitable for lighting applications by doping Eu^{3+} ions into SrTeO_3 phosphor. The choice of Eu^{3+} ions is grounded in their ability to emit efficient red light when incorporated into an appropriate host material. Specifically, the red emission of Eu^{3+} ions, arising from the $^5\text{D}_0 \rightarrow ^7\text{F}_2$ electric dipole transition, exhibits hypersensitivity to the local chemical environment of Eu^{3+} ions within the lattice (Zhong *et al.*, 2015). Notably, the incorporation of rare earth (RE) elements into SrTeO_3 phosphors has garnered attention due to their high light-emitting efficiency,

holding promise for optical devices operating across the UV and visible ranges (Abubakr *et al.*, 2022). This avenue of research presents exciting possibilities for enhancing the performance and applications of lighting technologies with improved color quality and spectral characteristics.

Experiment

The $\text{Sr}_{(1-x)}\text{TeO}_3: x\text{Eu}_2\text{O}_3$ phosphors ($x = 0.00, 0.01, 0.02, 0.05, 0.10, \text{ and } 0.20\%$) were synthesized through the solid-state reaction method, a meticulous procedure designed to ensure the precision and quality of the resulting materials. Initially, a carefully measured 5 g of the chemical component were combined with acetone. This mixture underwent a thorough ball milling process at 300 rpm for a duration of 3 h, creating a homogenous blend. Once the chemicals achieved complete dryness, a pre-sintering step was implemented at 500°C for a period of 12 h. This pre-sintering process serves to enhance the homogeneity and stability of the compound. Following the pre-sintering, the material was finely ground into a powder, further refining its characteristics and preparing it for the subsequent stages of the synthesis. The next step was to makethe material into pellets after it had been powdered. Employing a pressure of 7,000 kPa, the material was meticulously pressed into uniform pellets, ensuring consistency in the resulting samples. The final crucial stage in the synthesis process was the to sintere the pressed pellets at 800°C for 3 h. The sample was polished and cut to a size of $1 \times 1.5 \times 0.3 \text{ cm}^3$ as shown in Figure 1, to be used to measure various results. This high-temperature sintering process facilitates the consolidation and crystallization of the material, ultimately yielding

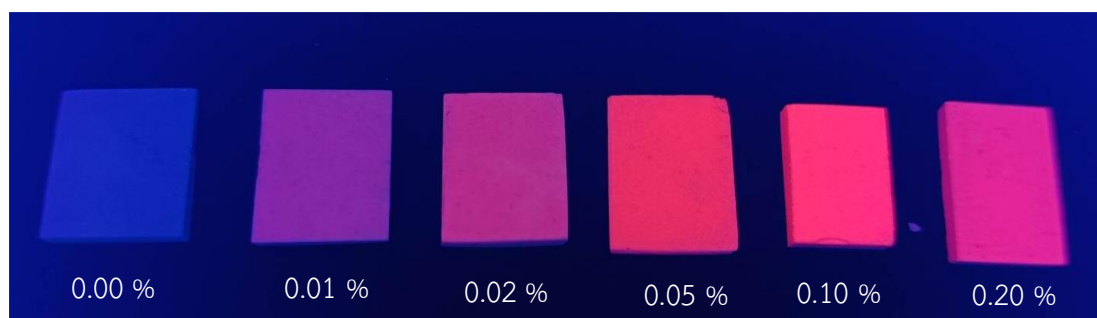


Figure 1. The $\text{Sr}_{(1-x)}\text{TeO}_3$ phosphors at different concentrations of Eu_2O_3

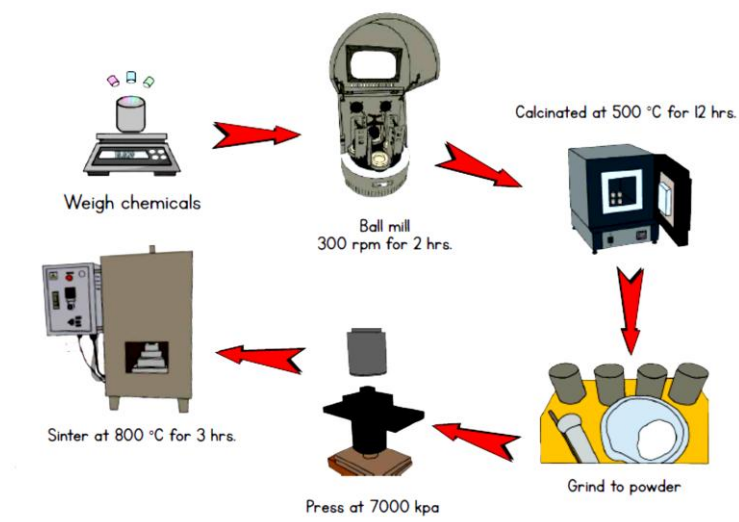


Figure 2. Schematic diagram for the synthesis process of Eu_2O_3 doped SrTeO_3 phosphors

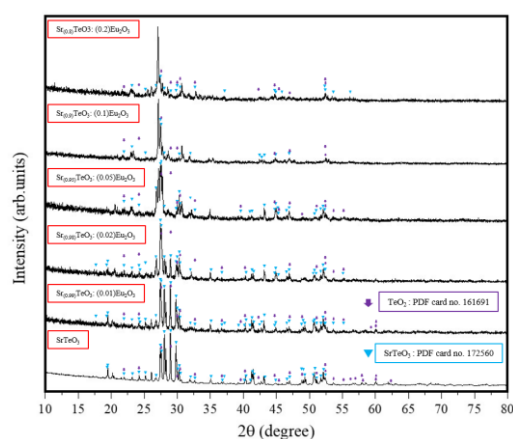


Figure 3. The XRD pattern of Eu_2O_3 doped SrTeO_3 phosphors

the desired samples. The results of this comprehensive synthesis process are illustrated in Figure 2, providing a visual representation of the achieved materials characteristics.

Results and Discussions

Crystal Phases of SrTeO_3 Phosphors

In Figure 3, the XRD patterns of SrTeO_3 phosphors are presented, highlighting variations in the patterns corresponding to different concentrations of Eu_2O_3 . Observing Figure 3, it becomes evident that all samples exhibit a mixed-phase composition, characterized by the presence of TeO_2 (PDF card no. 161691) and SrTeO_3 (PDF card no. 172560). However, with increasing

concentration of Eu_2O_3 discernible shift in phase composition occurs. Specifically, the SrTeO_3 phase becomes more prominent and pronounced in the XRD patterns. This shift in phase dominance indicates the influence of Eu_2O_3 doping on the crystal structure of the SrTeO_3 phosphors. The increased concentration of Eu_2O_3 appears to contribute to the enhancement or stabilization of the SrTeO_3 phase, potentially influencing the overall crystalline structure and properties of the synthesized materials. Further interpretation of these results could provide deeper insights into the interplay between dopant concentration and the crystalline phases, offering valuable information for optimizing the synthesis process and tailoring the material properties for specific applications.

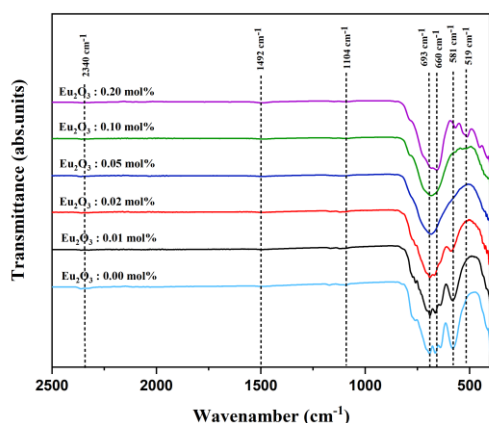


Figure 4. The FT infrared of Eu_2O_3 doped SrTeO_3 phosphors

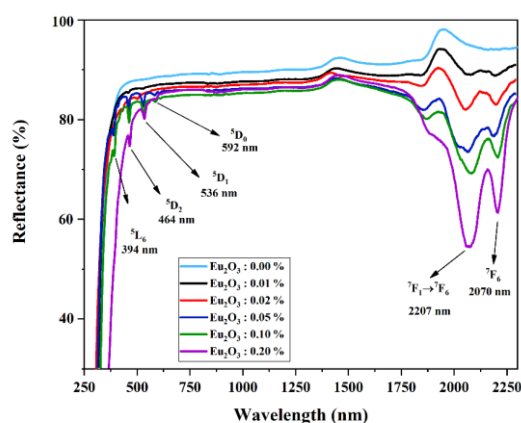


Figure 5. The reflectance spectra of Eu_2O_3 doped SrTeO_3 phosphors

Table 1. FTIR band positions and their peak assignments of the Eu_2O_3 doped SrTeO_3 phosphors

Wavenumber (cm ⁻¹)	Assignments	Reference
≥1,104	The vibration of OH group.	Selvi <i>et al.</i> (2017)
~693	The tributed to the Te-O bending vibrations of TeO_3 and TeO_6 structural units.	Selvi <i>et al.</i> (2017)
~660	The stretching vibration of Te-O bonds in the TeO_4 units.	Sidek <i>et al.</i> (2009)
~519, 581	The Sr-O stretching vibration of SrO units.	Ouis and Marzouk (2020)

Fourier Transform Infrared Spectroscopy

The Fourier Transform Infrared Spectroscopy (FTIR) analysis of Eu_2O_3 doped SrTeO_3 phosphors was conducted over the range of 400-2,500 cm^{-1} , revealing valuable insights into the molecular composition and bonding characteristics of the synthesized materials, as depicted in Figure 4. The obtained IR spectra exhibit three distinct broad bands, each indicating characteristic component peaks. A comprehensive summary of the band assignments is presented in Table 1.

1) The band appearing after 1,104 cm^{-1} is attributed to the OH vibrations, and the band appearing around 693 cm^{-1} is attributed to the Te-O bending vibrations of both TeO_3 and TeO_6 structural units (Selvi *et al.*, 2017). This specific vibrational mode provides valuable information about the arrangement and interactions of tellurium and oxygen within the molecular structure, contributing to the overall understanding of the phosphors composition.

2) Bands observed at 651 cm^{-1} have been attributed to the stretching vibration of Te-O bonds within the TeO_4 units (Sidek *et al.*, 2009). The presence and characteristics of these vibrations offer insights into the specific bonding configurations of tellurium and oxygen, shedding light on the structural integrity of the TeO_4 units within the phosphor.

3) A distinct wavenumber at 575 cm^{-1} is assigned to the Sr-O stretching vibration of SrO units (Ouis and Marzouk, 2020). This vibrational mode provides information about the strontium-oxygen interactions, aiding in the analysis of the SrO units structural role and their contribution to the overall material properties.

It is noteworthy that a decrease in intensity observed at the wavenumber 561 cm^{-1} is attributed to the escalating concentration of Eu_2O_3 doping, indicating a potential influence on the strontium-related vibrations. This observation suggests that the substitution of strontium with Eu_2O_3 may lead to alterations in the vibrational characteristics of SrO units, further influencing the material's infrared response.

Absorption Spectra Study

A notable observation from Figure 5 is the discernible increase in the sharpness of these absorption bands with an elevated concentration of Eu_2O_3 . This indicates that the introduction of higher concentrations of Eu_2O_3 correlates with a more defined and pronounced absorption behavior. The enhanced sharpness of the absorption bands could be attributed to the increased influence of Eu_2O_3 on the electronic structure of the SrTeO_3 phosphor, potentially altering the energy levels involved in the absorption transitions.

The reflectance spectra of all Eu_2O_3 doped SrTeO_3 phosphors at 250 to 2,300 nm are shown in Figure 5. A part of the deflated curve in the spectra performs the absorption ability of phosphor. These spectra show six absorption bands at 394 ($^7\text{F}_0 \rightarrow ^5\text{L}_6$), 464 ($^7\text{F}_0 \rightarrow ^5\text{D}_2$), 536 ($^7\text{F}_0 \rightarrow ^5\text{D}_1$), 592 ($^7\text{F}_0 \rightarrow ^5\text{D}_0$), 2,207 ($^7\text{F}_1 \rightarrow ^5\text{F}_6$) and 2,070 ($^7\text{F}_0 \rightarrow ^5\text{F}_6$) nm respectively (Deng *et al.*, 2017). Furthermore, it was found that the sharpness of absorption bands increased with increasing of Eu_2O_3 concentration as shown on Figure 5.

Photoluminescence of Eu_2O_3

In Figure 6(a), the excitation spectra of Eu_2O_3 at room temperature are illustrated, with a focus on monitoring the emission wavelength at 614 nm. Seven distinct excitation peaks are observed at wavelengths 362, 382, 394, 415, 465, 525, and 535 nm, corresponding to the $^5\text{D}_4$, $^5\text{G}_3$, $^5\text{D}_3$, $^5\text{L}_6$, $^5\text{D}_2$, $^5\text{D}_1$, and $^5\text{D}_0$ energy levels, respectively, as shown on

Figure 7. Notably, the excitation peak at 465 nm, corresponding to the $^7\text{F}_0 \rightarrow ^5\text{D}_2$ transition, is particularly intense, making it the chosen wavelength for the subsequent measurement of the emission spectra of the phosphors. Figure 6(b) shows the room-temperature emission spectra of Eu_2O_3 -doped SrTeO_3 phosphors under the excitation wavelength of 465 nm. The emission peaks demonstrate characteristic lines of the Eu^{3+} ion, corresponding to the $^5\text{D}_0 \rightarrow ^7\text{F}_Y$ ($Y = 0, 1, 2, 3, 4$) transitions (Zhang *et al.*, 2010). The intensity of these transition peaks increases with an increase of Eu_2O_3 concentration until it reaches 0.10%, beyond this value, the peak intensity decreased. This decrease indicates the concentration quenching in SrTeO_3 phosphors at 0.10% of Eu_2O_3 . The phenomenon of concentration quenching is attributed to non-radiative energy transfer mechanisms, such as cross-relaxation and resonant energy transfer. As the concentration of Eu^{3+} ions increases, the proximity between the ions decreases,

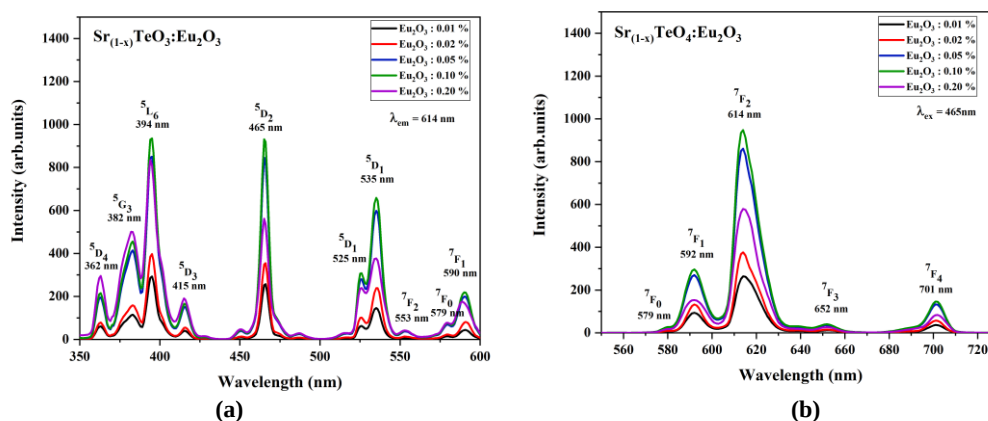


Figure 6. (a) The excitation spectra of $\text{SrTeO}_3:\text{Eu}^{3+}$ ion phosphors, (b) The emission spectra of $\text{SrTeO}_3:\text{Eu}^{3+}$ ion phosphors

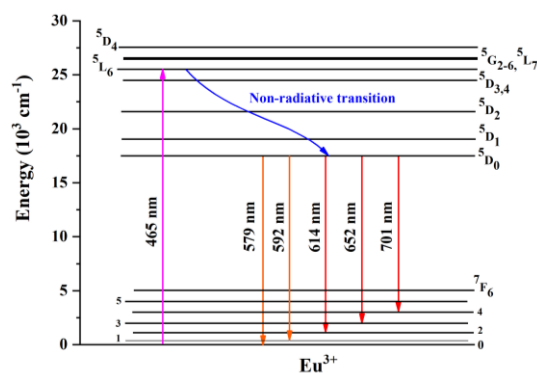


Figure 7. The energy level diagram of Eu^{3+} ion in SrTeO_3 doped Eu_2O_3

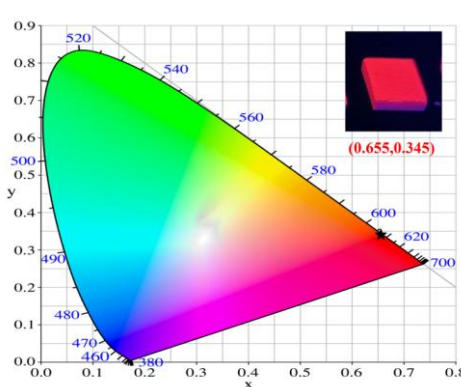


Figure 8. The CIE chromaticity diagram of SrTeO_3 doped Eu_2O_3 at 0.10%

leading to an increase in energy transfer through cross-relaxation (Zaman *et al.*, 2016). To characterize the color of the emitted light, the CIE chromaticity diagram is employed, providing a color coordinates for the SrTeO_3 doped with 0.10% Eu_2O_3 phosphors. The chromaticity coordinates (x, y) of these phosphors were determined to be $x = 0.655$ and $y = 0.345$, respectively. The estimated values are located on the reddish-orange region as depicted in Figure 8. This positioning makes these phosphors suitable for applications requiring a reddish-orange hue. Furthermore, the correlated color temperature (CCT) is utilized to assess the light source quality. The obtained CCT value for the prepared samples is 2,700 K, indicating a warm white light. This characteristic makes the phosphors promising for the development of solid-state lighting with a reddish-orange tint (Bashar *et al.*, 2020).

X-ray-induced Luminescence

Eu^{3+} ion doped SrTeO_3 phosphors were subjected to X-ray irradiation with 50 kV and 30 mA, and the resulting emission spectra are depicted in Figure 9. Five distinctive peaks were identified at wavelengths 535, 591, 612, 652, and 702 nm, corresponding to specific electronic transitions of Eu^{3+} ions. Notably, the intensity of these peaks exhibited a notable increase with the increasing concentration of Eu_2O_3 until reaching a concentration of 0.10%, after which a decline was observed. The occurrence of a double peak at the wavelength position of 612 nm is attributed to crystal field splitting, revealing insights into the electronic structure and symmetry of the doped SrTeO_3 phosphors (Dorenbos, 2002). To assess the effectiveness of these phosphors in comparison to established scintillator materials, the sample exhibiting the highest intensity was compared with BGO (Bismuth Germanate) crystals, commonly used in scintillator detectors. The X-ray luminescence spectra, presented in Figure 10, showcasing the scintillation efficiency of SrTeO_3 phosphors with 0.10% Eu_2O_3 ions when compared to BGO crystals. Remarkably, the integral scintillation efficiency of the phosphors was determined to be 33.65% of that of BGO crystals. This signifies that SrTeO_3 phosphors, particularly when doped with 0.10% Eu_2O_3 ions, exhibit a significant scintillation efficiency suitable for the development of scintillator devices in X-ray detectors.

Lifetimes

The lifetimes of the $^5\text{D}_2$ energy level were measured for various concentrations of Eu_2O_3 , and the experimental decay curves are presented in

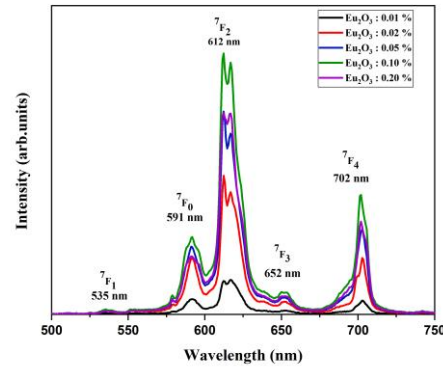


Figure 9. The X-rays induced optical luminescence spectra of SrTeO_3 doped Eu^{3+} ions phosphors

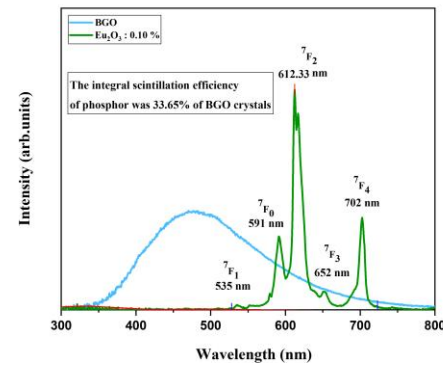


Figure 10. The x-rays induced optical luminescence spectra of SrTeO_3 doped 0.10% of Eu_2O_3 ions phosphors compared with BGO crystals

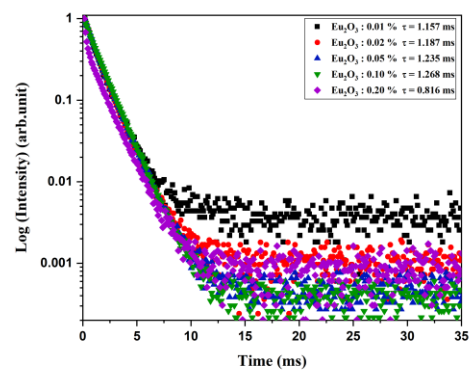


Figure 11. The lifetimes of Eu^{3+} ions doped SrTeO_3 phosphors

Figure 11. A noticeable trend emerges where an increase in the concentration of Eu^{3+} ions until 0.20% After that it decreases. This phenomenon can be attributed to the influence of increased Eu^{3+} ions

on the decay process, primarily through energy transfer mechanisms, particularly cross-relaxation. When Eu^{3+} ions are excited above the $^5\text{D}_2$ level, the electrons undergo rapid non-radiative relaxation, causing a decay to the $^5\text{D}_2$ level. Subsequently, emissions occur as the electrons transition from this energy level to lower energy levels (Suhasini *et al.*, 2009). The observed increase in lifetime with increasing Eu^{3+} ion concentration suggests a less efficient energy transfer process between the excited and ground state ions, predominantly facilitated by cross-relaxation mechanisms. In summary, the experimental decay curves in Figure 11 provide a visual representation of the lifetime variations at the $^5\text{D}_2$ energy level for different concentrations of Eu_2O_3 . The increase in lifetime with increasing Eu^{3+} ion concentration underscores the impact of cross-relaxation on the decay dynamics, shedding light on the intricate interplay between dopant concentration and the luminescent properties of the SrTeO_3 phosphors.

Conclusions

In summary, this research focused on Eu^{3+} ion-doped SrTeO_3 phosphors, examining their optical and luminescent properties. XRD analysis revealed a mixed phase composition, with an increasing dominance of SrTeO_3 phase as the concentration of Eu_2O_3 increased. FTIR spectroscopy provided insights into molecular vibrations, indicating the presence of specific TeO_3 , TeO_4 , and SrO units. Reflectance spectra displayed distinctive absorption bands corresponding to various electronic transitions of Eu^{3+} ions. Excitation spectra highlighted a strong peak at 465 nm, which was selected for emission measurements. The emission spectra exhibited characteristic lines of Eu^{3+} ions, with intensity increasing until 0.10% Eu_2O_3 concentration, followed by concentration quenching. X-ray irradiation produced emission spectra, unveiling five peaks indicative of Eu^{3+} transitions. The scintillation efficiency of SrTeO_3 phosphors at 0.10% Eu_2O_3 was found to be 33.65% when compared to BGO crystals, showcasing their potential as scintillator materials for X-ray detectors. Lifetime measurements at the $^5\text{D}_2$ energy level demonstrated an increase with increasing Eu^{3+} ion concentration, implicating energy transfer via cross-relaxation. These findings collectively contribute to understanding the optical and luminescent behavior of Eu^{3+} ion-doped SrTeO_3 phosphors, positioning them as promising candidates for applications in solid-state lighting and scintillator devices.

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