



# Advancements in Multicolor Luminescent Phosphors: Synthesis, Characterization, and Potential Applications

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## ARTICLE INFO

## ABSTRACT

Long-lasting luminescence (LPL) phenomenon, exhibited by materials known as long persistent phosphors (LPPs), has garnered significant attention due to its potential applications in various fields. This study focuses on the synthesis and characterization of BaZrSi<sub>3</sub>O<sub>9</sub> phosphors doped with rare earth ions (Eu<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>, Tb<sup>3+</sup>, and Pr<sup>3+</sup>). The materials were prepared using a high-temperature solid-state reaction method, followed by thorough characterization using X-ray diffraction (XRD) and photoluminescence (PL) spectroscopy. Results show successful incorporation of rare earth ions into the BaZrSi<sub>3</sub>O<sub>9</sub> host lattice, leading to distinct emission spectra alongside the host's luminescence. Moreover, afterglow decay curves were analyzed, revealing the presence of multiple trapping centers with varying depths. Thermoluminescence (TL) glow curve experiments provided insights into the trap distribution and their contribution to LPL characteristics. Overall, this research contributes to the understanding of multicolor luminescent phosphors and suggests avenues for the development of new LPPs with diverse applications.

**Keywords:** Long-lasting luminescence (LPL), Rare earth ions, BaZrSi<sub>3</sub>O<sub>9</sub> phosphors, Multicolor luminescent phosphors, Trap distribution.

## Introduction

Long lasting luminescence (LPL) is an optical phenomenon that continues light emission after the excitation source is turned off. Materials capable of exhibiting LPL, known as long persistent phosphors (LPPs), have been extensively researched due to their environmentally beneficial and energy-efficient characteristics [1]. Advanced LPP materials, such as SrAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>, Dy<sup>3+</sup> and Sr<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>:Eu<sup>2+</sup>, Dy<sup>3+</sup>, have been successfully utilized for commercial purposes, showing great potential in various fields such as emergency signage, radiation detection, structural damage sensors, medical diagnostics, optical memory storage, defense identification markers, and optical probes for live bio-imaging. However, the progress of LPPs faces challenges, including the fabrication of white LPPs and the incomplete understanding of LPL processes and the identification of traps. To overcome these obstacles, it is crucial to develop new varieties of LPPs that incorporate multi-color LPL. Multicolor luminescent phosphors (LPPs) have been described by selectively incorporating relevant rare earth ions into acceptable host materials, such as silicate, borate and carbonate with Eu<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>. These LPPs consist of silicate minerals containing Sn or Zr, and are self-activated. The origin of LPL in these uncommon ions is attributed to the transfer of energy from the host material to the luminous activators [2]. This study describes the successful preparation of BaZrSi<sub>3</sub>O<sub>9</sub> phosphors doped with rare earth ions, specifically Eu<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>, Tb<sup>3+</sup>, and Pr<sup>3+</sup>, using luminescent spectra, afterglow decay curves, and thermoluminescence (TL) glow curves. The transfer of energy from the host luminescence to these activators can be verified, and rare earth ions in BaZrSi<sub>3</sub>O<sub>9</sub> can function as both emission centers and aliovalent auxiliary dopants, creating additional trapping centers [3].

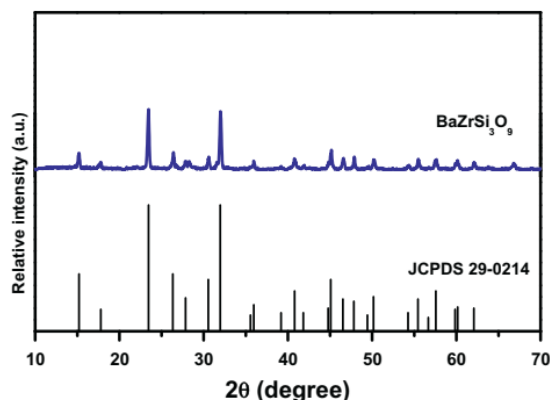


Figure 1. X-ray diffraction (XRD) patterns of undoped  $\text{BaZrSi}_3\text{O}_9$  in both experimental and standard forms.

### Experimental Procedures

The undoped  $\text{BaZrSi}_3\text{O}_9$  and  $\text{BaZrSi}_3\text{O}_9:\text{R}^{3+}$  powder samples were synthesized using a high temperature solid state reaction method [4]. The ingredients used in the preparation included  $\text{BaCO}_3$ ,  $\text{ZrO}_2$ ,  $\text{SiO}_2$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Tb}_4\text{O}_7$ , and  $\text{Pr}_6\text{O}_{11}$ . The stoichiometric ratio was measured, and the powders were combined and pulverized using an agate mortar. The mixtures were then subjected to calcination at  $1400^\circ\text{C}$  for 6 hours in a tubular furnace. After reaching room temperature, the samples were pulverized into fine powders for subsequent measurements. X-ray diffraction (XRD) measurements were conducted on all powders using an XRD powder diffractometer (Model 3010 Analytical Technologies India). The measurements were performed with Cu K $\alpha$  irradiation in the  $2\theta$  range from  $10^\circ$  to  $70^\circ$  [5]. The Hitachi F-7000 Fluorescence Spectrophotometer was used to measure photoluminescence (PL) spectra, and the samples were subjected to UV radiation for 1 minute before taking measurements. The mass of each sample remained constant during the decay and TL curve measurements to ensure accuracy [6]. In conclusion, the undoped  $\text{BaZrSi}_3\text{O}_9$  and  $\text{BaZrSi}_3\text{O}_9:\text{R}^{3+}$  powder samples were synthesized using a high temperature solid state reaction method. The samples were subjected to UV radiation for 1 minute before taking measurements [7-10].

### Results and Discussion

The X-ray diffraction (XRD) analysis of the BZS host shows that all diffraction peaks can be matched to the standard data of  $\text{BaZrSi}_3\text{O}_9$ . Other samples doped with rare earth ions exhibit XRD patterns similar to those of BZS, and the crystalline structure of each obtained sample is similar to that of  $\text{BaTiSi}_3\text{O}_9$ . The Dexter theory states that for energy transfer to occur, there must be a spectrum overlap between the emission of the donor and the excitation of the acceptor. There is a high likelihood of energy transfer occurring from the host to the doped activators [11]. After stimulation with a wavelength of 254 nm, all of the samples obtained exhibit LPL [12-14]. The afterglow color of the BZS host remains nearly constant regardless of the decay duration. The afterglow emission color of five BZS samples doped with rare earth elements differs to some extent due to the differing decay ratio between the BZS host and rare earth ions. The variable color of LPL is a result of the persistent luminescence emitted by the doped rare earth ions. The decay of afterglow has an initial quick decline followed by a subsequent slow decay, indicating the presence of different traps with varying depths [15].

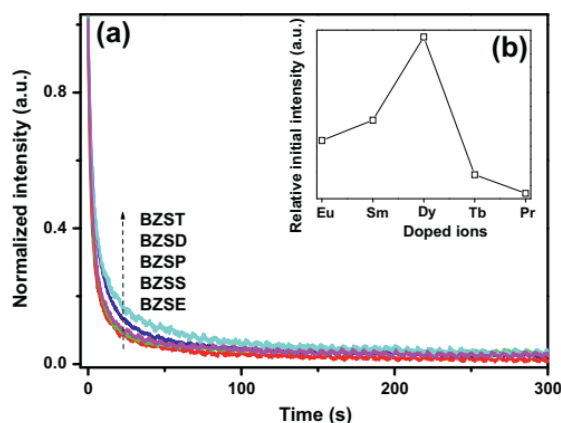


Figure 2. Decay curves (a) and relative initial intensities of afterglow (b) for samples BZSE, BZSS, BZSD, BZST and BZSP.

The afterglow decay curves of rare earth ions doped samples in Fig. 4 can be well fitted with a double-exponential equation (1) as follows:

$$I = A_1 e^{-\frac{t}{\tau_1}} + A_2 e^{-\frac{t}{\tau_2}} \quad \dots\dots (1)$$

where  $I$  represents the intensity;  $A_1$  and  $A_2$  are constants;  $\tau_1$  and  $\tau_2$  are decay times for the exponential components indicating the rapid and slow decay processes, respectively;  $t$  is time. The luminous photoluminescence (LPL) behavior in materials doped with rare earth ions is examined in this study. A significant factor in LPL is the presence or absence of traps in the host lattice, which allow the discharge of charge carriers from designated locations. Both the depth of the trap and its density affect LPL's performance [16]. Deep traps have low or nonexistent LPL because they are extremely immobilized and can't easily revert to their excited state at room temperature. Luminescent phosphor materials that have a high trap density and a suitable trap depth usually create an excellent LPL that has a strong afterglow and lasts for a long time [17]. The presence of foreign ions with charges differing from the cations of the host compounds, high temperatures, or reducing environments can all cause lattice defects in the host material, which in turn can cause traps. In contrast to the inherent traps in undoped BZS, the incorporation of rare earth ions into the BaZrSi<sub>3</sub>O<sub>9</sub> host lattice may produce new traps. To gain important insights into Eu<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>, Tb<sup>3+</sup>, and Pr<sup>3+</sup> doped BaZrSi<sub>3</sub>O<sub>9</sub> systems exhibiting LPL, TL glow curve experiments were carried out. It qualitatively reveals the traps' important information, like how they contribute to the LPL's features, depth, and density. At 39, 107, 150, and 216 °C, two noticeable peaks are seen in the BZS sample; these correspond to the shallowest, deeper, and deepest traps, respectively [18, 19].

### Conclusion

The present study details the discrete doping of five suitable rare earth ions R<sup>3+</sup> into the BaZrSi<sub>3</sub>O<sub>9</sub> host: Eu<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>, Tb<sup>3+</sup>, and Pr<sup>3+</sup>. The distinctive light emitted by these R<sup>3+</sup> ions is clearly visible alongside the blue light emitted by the BaZrSi<sub>3</sub>O<sub>9</sub> host. This is because these R<sup>3+</sup> ions receive energy from the host. The distribution of traps in the host lattice was altered and new traps were formed by the insertion of R<sup>3+</sup> ions. A brief discussion was made of the mechanism of LPL. It is reasonable to assume that by doping the right activator ions into the right hosts, we can create new LPPs with a wider range of LPL colors or even complete colorization.

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### Conflict of Interest Statement

The authors conducted ethical and transparent research, demonstrating the importance of ethical practices in scientific research.

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