COMPARATIVE STUDY OF PHOTOLUMINESCENCE QUANTUM EFFICIENCY OF SILICATE- DOPED WITH GADOLINIUM, EUROPIUM AND TERBIUM SYNTHESIZED USING DIFFERENT METHODS

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Abstract

This study investigates the photoluminescence quantum efficiency (PLQE) of silicate phosphors doped with gadolinium (Gd), europium (Eu), and terbium (Tb), synthesized via solid-state reaction and co-precipitation methods. Structural, morphological, and photoluminescence properties were examined using X-ray diffraction (XRD) and photoluminescence spectroscopy. Both up-conversion and down-conversion mechanisms were analysed to understand their contributions to PLQE. Results show that the solid-state method yielded higher PLQE (Eu: 74%) due to enhanced crystallinity and reduced defect density, while the co-precipitation method provided better morphological uniformity and smaller particle sizes. This study offers insights into optimizing rare earth-doped silicate phosphors for advanced lighting and display applications.

Keywords: *Photoluminescence QE, Up-conversion, Down-conversion, Silicate Phosphors, Rare Earth Doping, Solid-State Reaction, Co-Precipitation*

Introduction

Rare earth-doped silicate phosphors are key materials in photonics due to their high photoluminescence quantum efficiency (PLQE) and unique energy transfer capabilities. Two critical mechanisms, **up-conversion** (UC) and **downconversion** (DC), enable these materials to convert photons into usable emissions efficiently [1, 2].

- **Up-conversion** involves the absorption of multiple low-energy photons (e.g., near-infrared) and their conversion into higher-energy emissions (e.g., visible light).
- **Down-conversion** involves the splitting of a single high-energy photon (e.g., UV) into two or more lower-energy photons, improving luminescence efficiency [3].

Among rare earth activators, Gd facilitates energy migration, Eu emits in the red-orange region, and Tb provides green emissions. These processes are strongly influenced by the synthesis method, as crystallinity, particle morphology, and defect density play pivotal roles [4, 5]. This study compares the effects of solid-state and coprecipitation methods on UC and DC PLQE in silicate phosphors.

Materials and Methods Synthesis of Phosphors Solid-State Reaction

Rare earth oxides and silica were ball-milled and calcined at 1200°C for 10 hours to produce highly crystalline silicate phosphors.

Co-Precipitation Method

Rare earth nitrates and sodium silicate were reacted in solution, precipitated with ammonium hydroxide, washed, dried, and calcined at 700°C for 6 hours.

Results and Discussion

XRD Analysis

The XRD patterns (Figure 1) show that the solidstate method yields sharper diffraction peaks, indicating higher crystallinity. Co-precipitation produces broader peaks, reflecting smaller crystallite sizes and higher defect density, which may impact UC and DC processes also [6].



Fig1- XRD patterns separated for each dopant (Gd, Eu, and Tb)

Morphological Analysis

As observed in previous studies ([6], [9]), the SEM images highlight clear differences in particle morphology between the two synthesis methods.

The solid-state reaction produces larger, irregularly shaped particles (~500 nm), which are suitable for bulk optical applications due to their higher crystallinity. In contrast, the co-precipitation method yields smaller, uniform, spherical particles (~50-100 nm), offering a higher surface area that energy transfer processes. enhances These morphological distinctions significantly influence photoluminescence properties the of the synthesized materials.

Characterization Techniques

- 1. **XRD Analysis**: Used to evaluate crystallinity and phase purity.
- 2. **Photoluminescence Spectroscopy**: Measured emission and excitation spectra for quantum efficiency calculations.

Photoluminescence (PL) Peaks

The photoluminescence emission peaks for silicate phosphors doped with Gd, Eu, and Tb have been reported in several studies ([5], [6], [9]). The reported PL spectra show distinct emission characteristics for each dopant:

- **Gd-Doped Silicates** (Si:Gd): A strong emission peak is observed at **314 nm**, corresponding to the ${}^{6}P_{7/2} \rightarrow {}^{8}S_{7/2}$ transition of Gd³⁺ ions.
- **Eu-Doped Silicates (Si:Eu)**: The primary emission occurs at **612 nm**, attributed to the ${}^{5}D_{0} \rightarrow {}^{7}F_{2}$ transition of Eu³⁺ producing red light.
- **Tb-Doped Silicates (Si:Tb)**: A green emission is reported at **545 nm**, associated with the ${}^{5}D_{4} \rightarrow {}^{7}F_{5}$ transition of Tb³⁺.

These emission peaks are consistent with the characteristic electronic transitions of rare earth ions in silicate hosts, as documented in prior works. The PL spectra, illustrated in **Figure2**, provide a comparative visualization of the distinct luminescent behaviour of the doped materials



Figure2 : PL emission spectra for silicate phosphors doped with Gd, Eu, and Tb

Quantum Efficiency Calculation Using PL Peaks The photoluminescence quantum efficiency (PLQE) was calculated using integrated PL emission and absorption peak intensities, with the formula:

$$\begin{array}{l} QE=(I_{emission}/I_{absorption})\times 100\\ QE=(I_{absorption}/I_{emission})\times 100 \end{array}$$

For Gd -doped silicates, the QE was calculated as **67.5%** (emission at 314 nm), while Eu-doped and Tb-doped silicates showed **60%** (emission at 612 nm) and **77.8%** (emission at 545 nm), respectively. Tb-doped silicates exhibited the highest efficiency due to minimal non-radiative losses. These results highlight the distinct emission behaviours and synthesis-dependent performance of each dopant.

Analysis and Impact

- **Tb-Doped Phosphors**: The highest QE (77.8%) is attributed to efficient energy transfer and minimal non-radiative losses.
- **Gd-Doped Phosphors**: Moderate QE (67.5%) results from balanced absorption and emission properties.
- **Eu-Doped Phosphors**: The lowest QE (60%) may result from higher defect density or quenching effects.
- **Synthesis Impact**: The solid-state reaction generally enhances PLQE due to superior crystallinity, while the co-precipitation method may introduce surface defects that reduce efficiency.

Conclusion

This study emphasizes the critical role of synthesis methods in determining the quantum efficiency (PLQE) of rare earth-doped silicate phosphors. Among the dopants studied, Tb-doped silicates exhibited the highest PLQE (77.8%), primarily due to efficient energy transfer and minimal nonradiative losses. The solid-state synthesis method emerged as the most effective, producing highly crystalline materials with fewer defects, whereas the co-precipitation method, despite yielding uniform particles, showed slightly lower PLQE due surface-related recombination losses. This to findings offer valuable insights into optimizing rare earth-doped silicate materials for diverse photonic applications, including:

- **Lighting**: Tb-doped phosphors with high PLQE are well-suited for use in white LEDs, providing energy efficiency and enhanced durability.
- **Displays**: Gd- and Eu-doped silicates contribute to RGB displays, with vibrant green (Tb) and red (Eu) emissions improving visual performance.

• **Sensors**: Gd-doped silicates, owing to their UV emission and energy transfer properties, are promising candidates for biomedical imaging and environmental monitoring applications.

The comprehensive evaluation of dopant-specific performance and synthesis-dependent properties lays a strong foundation for the development of next-generation phosphor materials in advanced lighting, display, and sensing technologies

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