

STUDIES ON PHOTOLUMINESCENCE AND THERMOLUMINESCENCE OF Y₂O₃ DOPED WITH ERBIUM (III) CODOPED WITH YTTERBIUM (III) PHOSPHOR

D. Fekar¹, P. P. Nimje^{1,✉}, and R. Shrivastava²

¹Department of Chemistry, Shri Shankaracharya Professional University,
Bhilai, 490020, Chhattisgarh, India

²Department of Physics, Shri Shankaracharya Professional University,
Bhilai, 490020, Chhattisgarh, India

✉Corresponding author: sspu.drprachi.chem@gmail.com

ABSTRACT

Synthesis of Y₂O₃ doped with Er³⁺ (1.5 mol%) and codoped with various concentrations of Yb³⁺ was reported. The Photoluminescence up-conversion Emission spectra were recorded by exciting the samples through a source of 980 nm. The PL emission spectra exhibited two prominent peaks centred at 560 nm and 660 nm, respectively and were attributed to the transitions ⁴S_{3/2} → ⁴I_{15/2} and ⁴F_{9/2} → ⁴I_{15/2} of Er³⁺, respectively. Thermoluminescence studies of the samples were indicative of the formation of deeper traps with 2nd order kinetics; therefore, materials considered may have a significant potential in the field of LED and display devices.

Keywords: - Phosphor, Up-conversion, Photoluminescence, Thermoluminescence, XRD

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INTRODUCTION

It has been reported that Yb³⁺ and Er³⁺ enhance the upconversion (UC) efficiency, with optimal doping concentrations and synthesis conditions playing crucial roles.

Doping Effects on Up-Conversion Luminescence

Yb³⁺ doping significantly influences the UC photoluminescence (PL) properties of Y₂O₃:Er³⁺ ceramics, with the highest UC emission intensity observed at 5.0% Yb and 0.5% Er, showcasing sharp emission peaks at 550 nm and 660 nm.¹

The UC emission intensity is also dependent on the excitation power, indicating a two-photon process in Er³⁺ transitions.²

Structural and Synthesis Considerations

The incorporation of Li⁺ ions into Y₂O₃ enhances luminescence by reducing defect concentrations, which are known to quench emission.³

Synthesis temperature affects the luminescence lifetime, with higher temperatures leading to improved structural homogeneity and longer lifetimes.⁴

Optimal Composition and Applications

The ideal molar ratio of Er³⁺/Yb³⁺ for maximum luminescence intensity is 1:9, with specific synthesis conditions yielding high-quality UC phosphors suitable for display devices and biosensors.⁵

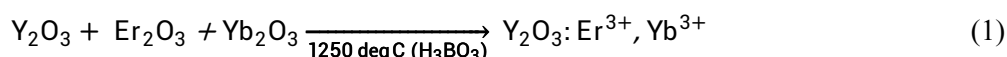
While the up-conversion properties of Y₂O₃: Er³⁺, Yb³⁺ are promising, challenges remain in optimising doping levels and synthesis methods to maximise efficiency and stability for practical applications.

EXPERIMENTAL

Materials and Methods

Yttrium Oxide (Y₂O₃) doped with different concentrations (i.e. 1.0, 1.5, 2.5, and 3.0 mol%) of Ytterbium (III), keeping 1.5 mol% of Erbium (III) (Er³⁺) were prepared using the solid-state reaction technique. Y₂O₃ (99.99%, Himedia), Er₂O₃ (99.99%, Himedia) and Yb₂O₃ (99.99%, Himedia) were mixed together using pastel and mortar in a stoichiometric ratio and kept in the muffle furnace at 1250 °C for about 2 h. 1.6 mol% of H₃BO₃ was also introduced as flux.

The following chemical equation was used for calculation in this method: -



In order to confirm the proper preparation and structural changes in the pure Y_2O_3 , X-ray diffraction (XRD) pattern was recorded using HORRIBA, JOBIN VYON - FLUOROLOG 3. In the said instrument, k_α anode (Copper) producing the wavelength 1.5405 Angstrom was used. A 365 nm UV excitation source was used in order to record the TL glow curves of the sample with optimum PL emission. The sample was put under the UV exposure for various durations (5, 10, 15 and 20 min) before recording the TL glow curve to understand the dose response of the sample considered. A TLD reader I1009 supplied by Nucleonix, Hyderabad, was used to record the TL glow curves.⁶⁻⁷

RESULTS AND DISCUSSION

X-Ray Diffraction (XRD) Pattern Analysis

In order to confirm the formation of expected phases in the material prepared, the XRD pattern of one of the samples (Y_2O_3 : 1.5 mol% of Er^{3+} and 2.0 mol% of Yb^{3+}) was observed. The observed pattern was matched using Crystal Impact Match 3! Software that expressed a strong resemblance with COD card No. 96-153-7883 with FoM 0.96. The discussed COD card belongs to Y_2O_3 with a cubic lattice type, having I a -3 (206) space group. To visualise the similarities between the observed and calculated XRD patterns, a comparative graph (Fig.-1) is plotted. The calculated pattern was obtained from the crystallographic information file (cif) corresponding to the COD card No. 96-153-7883. <https://www.crystallography.net/cod/>. Figure-1 shows that there is a significant resemblance between the observed and calculated XRD pattern of the material considered.⁷ Lattice parameter refinement was done using the prominent peaks obtained in observed XRD. Celref v.3.0 was used for the lattice parameter refinement process. Figure-2 exhibits the peaks selected for the refinement process. Table - 2 comprises the indexing and refinement results exported from the software Celref. It can be seen from Table-2 that the lattice volume was 1191.3 Å for the COD data card parameters, which is reported for un-doped Y_2O_3 and 1197 Å for Y_2O_3 doped with Er and Yb.

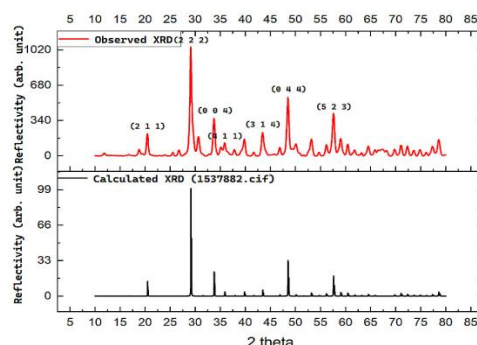


Fig.-1: Comparison between Observed and Calculated XRDs

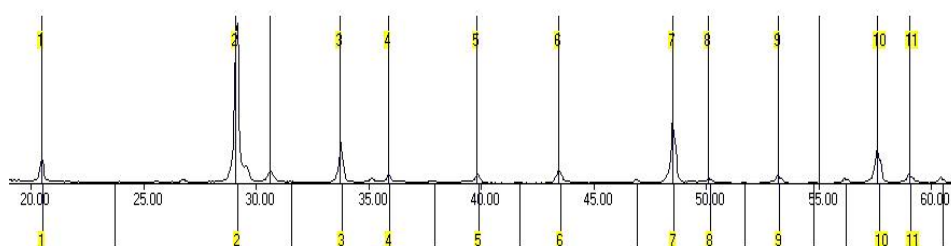


Fig.-2: XRD Peak Selections

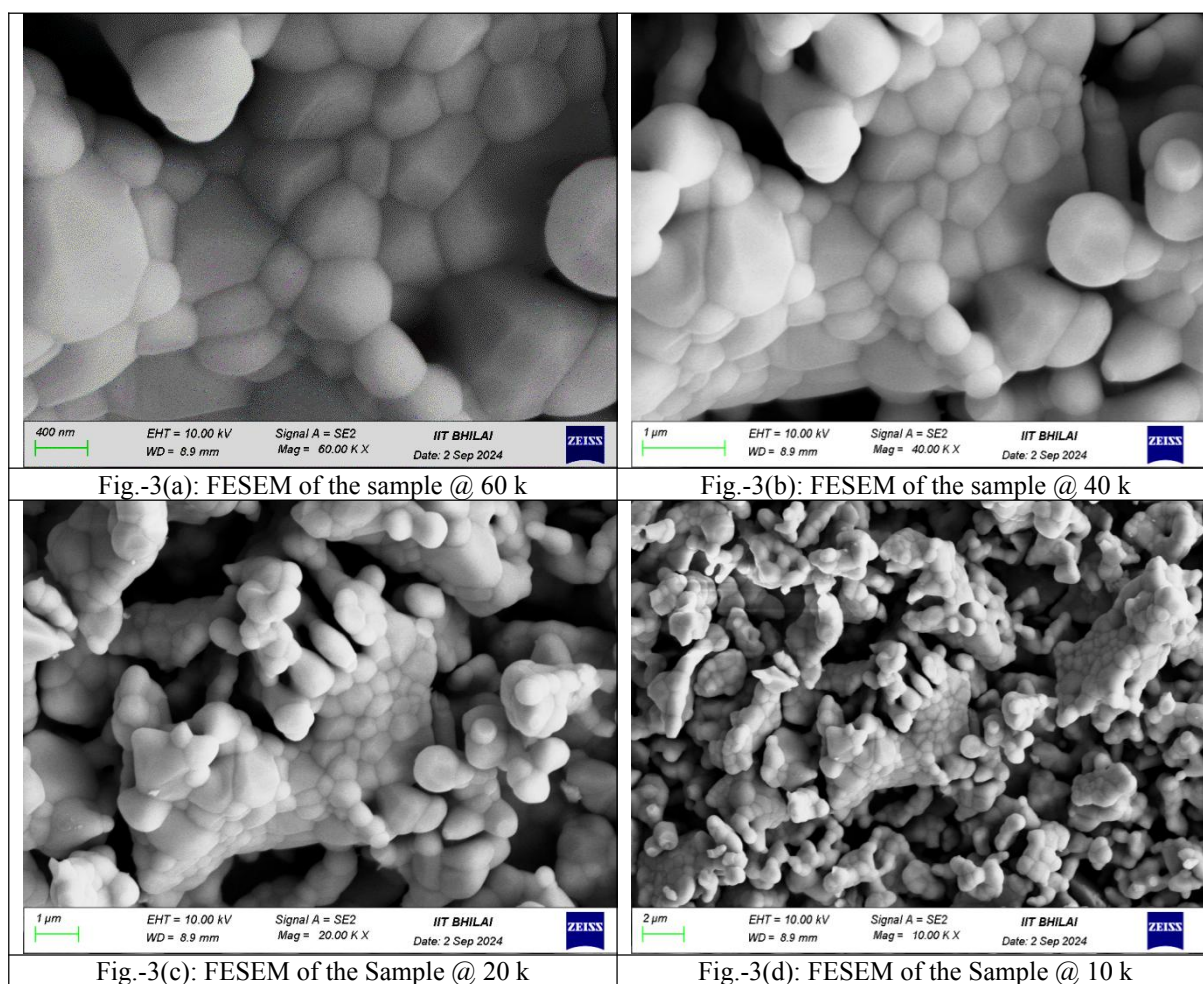
Table-1: Refined Lattice Parameters

Initial values								
Zero	Lambda	a	b	c	alpha	beta	gamma	Vol.
0	1.5418	10.601	10.601	10.601	90	90	90	1191.3
Final Values								
Zero	Lambda	a	b	c	alpha	beta	gamma	Vol.
0	1.5418	10.6177	10.6177	10.6177	90	90	90	1197
h	k	l	2T(Obs)	2T-Zero	2Th(Cal)	DIF		

2	1	1	20.456	20.456	20.4886	-0.03s26		
2	2	2	29.108	29.108	29.134	-0.026		
0	0	4	33.752	33.752	33.7664	-0.0144		
4	1	1	35.878	35.878	35.8821	-0.0041		
3	2	3	39.807	39.807	39.8207	-0.0137		
3	1	4	43.447	43.447	43.4579	-0.0109		
0	4	4	48.52	48.52	48.4999	0.0201		
4	3	3	50.088	50.088	50.0936	-0.0056		
5	2	3	53.181	53.181	53.1757	0.0053		
2	2	6	57.597	57.597	57.5811	0.0159		
3	1	6	59.024	59.024	59.0013	0.0227		

Morphological Studies

To analyse the morphology of the samples prepared, Field Effect Scanning Electron Microscopy (FESEM) Images of the sample with optimum PL emission was considered. The Imaging was done at various zoom levels, i.e. 10 k, 20 k, 40 k, and 60 k. The FESEM images for various resolutions were provided in Fig.-3(a-d). It looks as if the particles are agglomerated, and when high-resolution images were observed, they indicated the shape of the particles to be circular. In a 10-k resolution image, the flower-like structure is also noticed.



UP-Conversion Photoluminescence Studies

It could be noticed from the literature review that Erbium and Ytterbium have the ability to exhibit up-conversion photoluminescence. Therefore, prepared samples underwent the excitation of 980 nm, and then the emission spectra for various samples were recorded. Figure-4 exhibits the emission spectra for various samples. It is noticed from the figure that PL emission intensity increases with increasing concentration of Ytterbium (III). The PL emission was recorded to be optimum when Ytterbium's concentration was 2.0 mol%, and increasing the concentration further results in concentration quenching, hence the PL intensity decreased. The PL emission spectra exhibited two

prominent peaks centred at 560 nm and 660 nm, respectively (Fig.-4). These peaks were attributed to the transitions $^4S_{3/2} \rightarrow ^4I_{15/2}$ and $^4F_{9/2} \rightarrow ^4I_{15/2}$ of Erbium (III), respectively^{6,8} (Fig.-5). Figure-6 provides the proposed mechanism for this Photoluminescence process. It is already reported that Erbium (III) has the ability to act dually. It may act like activator and also like sensitizer. In Figure 06, Er (01) transfers energy to other Erbium, i.e. Er (02), Ytterbium also functions like sensitizer and transfers some energy to Er (02), which is functioning like an activator. Er (02) reaches the excited state energy after receiving energies from Er (01) and Yb. Then various transitions take place results in emission. Er (01) and Yb were excited by an IR wave of wavelength 980 nm, but the energy transfers enabled the activator Erbium to function like an activator and produce the emission spectra.⁸⁻⁹

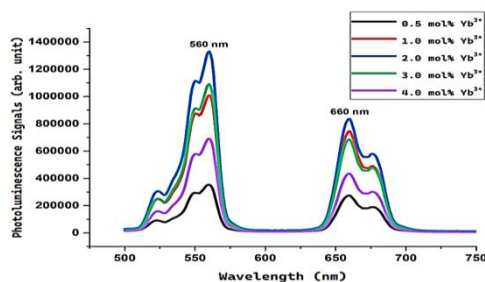


Fig.-3: Photoluminescence Emission Spectra of Y_2O_3 Doped with Various Concentrations of Yb, keeping 1.5 mol % of Er^{3+} Constant

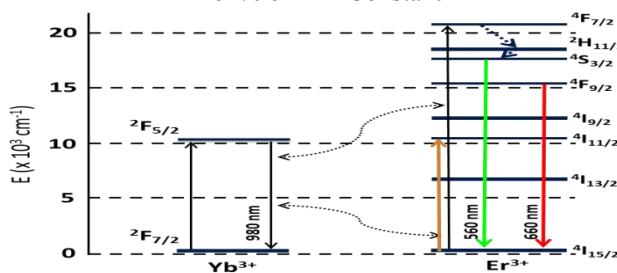


Fig.-4: Various Transitions Involved⁸⁻⁹

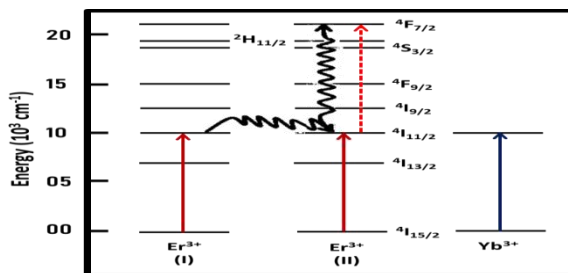


Fig.-5: Up-conversion Mechanism⁹

Thermoluminescence Studies

In order to further study the application of the prepared phosphor, Thermoluminescence (TL) studies were done. The phosphor with optimum PL intensity was considered for TL studies. A sample with 1.5 mol% of Er^{3+} and 2.0 mol% of Yb^{3+} was given various UV exposures (05, 10, 15, 20, 25, 30 and 35 min). It could be noticed that the TL intensity increased with increasing UV exposure time and reached its optimum value when exposure time was 30 minutes, after which quenching occurred, and intensity decreased abruptly for 35 minutes of UV exposure. It is also observed that the peak temperature for various UV exposure ranges between 241 – 258°C, which is quite a high temperature and indicative of the formation of deeper traps in this case. Formation of a deeper trap is one of the criteria for a phosphor to be a good candidate for LED applications. Kinetic parameters using the peak shape method were also calculated. The shape factor for various glow curves was found to be in the range of 0.528 - 0.579, which means these glow curves follow 2nd order Kinetics. The activation energy calculated was in the range of 0.52-0.61 eV, therefore indicative of the formation of deeper traps. 2nd Order kinetics and formation of deeper traps enable us to conclude that the materials considered may have a significant potential in the field of LED and display devices.⁷

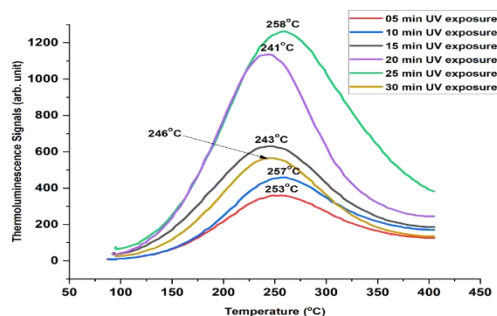


Fig.-7: Thermoluminescence Glow Curves for Various UV Exposure

Table-2: Thermoluminescence Kinetic Parameters

UV Min	T ₁	T _m	T ₂	τ	δ	ω	μ = δ / ω	Activation energy (eV)	Frequency Factor (s ⁻¹)
5	189.00	253.00	338.00	64	85	149	0.570	0.574	22780.681
10	196.00	257.00	341.00	61	84	145	0.579	0.614	52806.385
15	176.00	243.00	324.00	67	81	148	0.547	0.521	8270.887
20	183.00	241.00	306.00	58	65	123	0.528	0.592	50143.285
25	188.00	258.00	350.00	70	92	162	0.567	0.533	17180.374
30	183.00	246.00	317.00	63	71	134	0.530	0.556	17873.855

CONCLUSION

Up-conversion properties of Y₂O₃ doped with various concentrations of Ytterbium (III), keeping Erbium (III) to be 1.5 mol% in each of the case, were carried out. It was noticed that PL emission spectra exhibited two distinct emission peaks centred at 560 nm and 660 nm, respectively, upon excitation at 980 nm. Erbium (III) was considered to be functioning like an activator, whereas Ytterbium was considered as the sensitizer, responsible for up-conversion properties. Thermoluminescence Glow curves of a sample that provided the highest PL intensity were recorded by providing them various UV doses. The Glow curve indicated the existence of 2nd order kinetics. The activation energy suggested the formation of deeper trap levels. This information allows us to consider this phosphor to be a suitable candidate for LED or display device applications.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-7: Affordable and Clean Energy*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

Dhaleshwari Fekar  <https://orcid.org/0000-0002-2617-8697>

Prachi P. Nimje  <https://orcid.org/0009-0000-4655-8972>

Ravi Srivastava  <https://orcid.org/0000-0002-2617-8697>

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