



Structural and photophysical investigation of gadolinium based Eu^{3+} doped Gd_3GaO_6 phosphor: A red emitter with high color purity for WLEDs application

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ABSTRACT

To understand different properties of Eu^{3+} ions in inorganic phosphors, a series of red emitting $\text{Gd}_3\text{GaO}_6:\text{Eu}^{3+}$ was synthesized via solution combustion method. XRD patterns supported by Rietveld refinement revealed successful formation of crystalline sample with orthorhombic structure and $\text{Cmc}2_1$ space group. Microstructural features and elemental mapping of samples were examined by means of FE-SEM and EDX techniques. Photoluminescence spectra of Eu^{3+} -activated Gd_3GaO_6 revealed red emission at 612 nm due to the $^5\text{D}_0 \rightarrow ^7\text{F}_2$ transition under excitation at 261 nm. A decrease in emission intensity was observed after doping levels exceed 4 mol% indicating energy transfer between activator ions results in concentration quenching governed via dipole-dipole interactions. The intensity parameters Ω_2 and Ω_4 defined by Judd Ofelt theory were calculated utilizing emission spectra to investigate coordination symmetry around Eu^{3+} ions. The photoluminescence decay profile was well described by bi-exponential decay function. The activation energy 0.19873 eV suggests good thermal stability of $\text{GGO}:4 \text{ mol\% } \text{Eu}^{3+}$ phosphor. The high color purity (98.15 %) inferred from CIE coordinates revealed their applicability in white light emitting diodes.

1. Introduction

In the modern period of material science and technology, to meet the growing demand of world in agriculture, power consumption, medicinal chemistry, security, environment *etc.* research is being done to encourage the synthesis of multifunctional materials which is helpful in our day-to-day life [1,2]. Phosphors are the materials used in multiple systems like optical thermometry, WLEDs, fingerprint detection, security ink, therapeutics, semiconductor lighting, energy converters, drug delivery *etc.* [3–6]. Recent advancements in flat electroluminescent, plasma and field emission displays have substantial industrial applications which have forced the increasing demand of materials with enhanced features specifically in terms of stability, brightness and processing capabilities for industrial production [7,8].

Rare earth (RE) elements are generally known as vitamins in industry owing to their remarkable optical, electrical and magnetic properties [9,

10]. These ions are used as dopants in luminescent materials because 4 f electrons are insensitive to surrounding crystal field and site symmetry of host lattice on account of shielding of inner 4 f electrons by fully filled 5 s and 5 p orbitals [11,12]. As a result, weak electron phonon coupling takes place exhibiting narrow emission band owing to intra configurational 4 f luminescence in various hosts. Therefore, electronic levels of trivalent RE ions in different host lattices and those in free state are comparable [13]. Phosphors consist of a variety of host matrices like oxides, silicates, aluminates, phosphates, nitrides, oxynitrides, sulfides, borates *etc.* [14]. Among various host matrices, gallate host materials have been chosen on account of its low cost, high stability, high quantum efficiency, persistent luminescence and color tunable characteristics [15,16]. For example, a recent study demonstrated that co-doping with Bi^{3+} and Er^{3+} in Gd_3GaO_6 yields a multicolor phosphor with distinct emission modes (UV-excited down-shifting and visible up-conversion) confirming that the gallate matrix can support multiple activator ions

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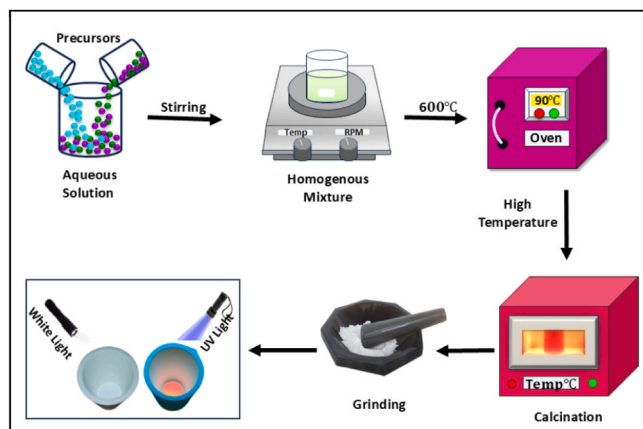


Fig. 1. Representation of solution combustion synthesis employed in nano-phosphor fabrication.

and varied emission channels [17]. More broadly, rare-earth-doped oxide hosts (e.g. RE_2O_3 , gallates, tungstates) such as Gd_2O_3 , Y_2O_3 or mixed gallates have been studied with Eu^{3+} , Tb^{3+} , Dy^{3+} , Sm^{3+} etc. revealing how ionic radius, site symmetry and local environment influence emission color, intensity and concentration quenching [18,19]. Out of various rare earth ions, trivalent europium-doped materials are potential candidates for producing red light emitting phosphors which is widely applied in lighting technology and display devices and gives sharp orange-red emission through intra configurational $^5\text{D}_0 \rightarrow ^7\text{F}_{1,2}$ transitions [20]. Trivalent europium ion displays distinct electric dipole (ED) and magnetic dipole (MD) transitions which are useful in determining local symmetry within prepared phosphors based on the alteration in these two transitions [21]. Intensity of MD $^5\text{D}_0 \rightarrow ^7\text{F}_1$ transition is unaffected by surrounding environment whereas the intensity of hypersensitive $^5\text{D}_0 \rightarrow ^7\text{F}_2$ electric dipole transition depends on the chemical environment around Eu^{3+} ion [22]. Frequently used synthesis methods for preparing phosphors chiefly include solid state synthesis, solution combustion synthesis, sol gel method, hydrothermal method and many more [23–25]. Solution combustion method has the characteristics of simple setup, self-sustained reaction which favors low energy requirement, yield homogenous, pure and single-phase material [26,27]. Thus, in this study Eu^{3+} doped Gd_3GaO_6 phosphors were synthesised by using solution combustion method and their luminescent and structural properties were examined. XRD patterns were gathered and subjected to Rietveld refinement to identify phase purity and unit cell parameters. FE-SEM and EDX techniques were employed to visualize morphology and elemental distribution of the sample, respectively. Luminescence behaviour of the materials was further assessed utilizing PL spectroscopy.

2. Experimental

A series of Eu^{3+} doped Gd_3GaO_6 phosphors was prepared via solution combustion method and is schematically represented in Fig. 1. The starting reagents used for the synthesis were Sigma Aldrich analytical grade reagents involving nitrates of europium, gallium, gadolinium and urea.

The quantity of precursors was calculated and weighed accurately and a clear homogenous solution was formed by adding minimal volume of deionized water. The solution was then stirred on heating plate for 20–25 min to develop a gel-like mixture. The solution mixture was then transferred to pre-heated furnace maintained at 600 °C and kept in this environment for 15 min. An exothermic process took place during this course with the release of gases like CO_2 , N_2 resulting in a voluminous product [28]. The product was cooled to room temperature and grinded utilizing agate mortar to a fine powdered form. Consequently, the

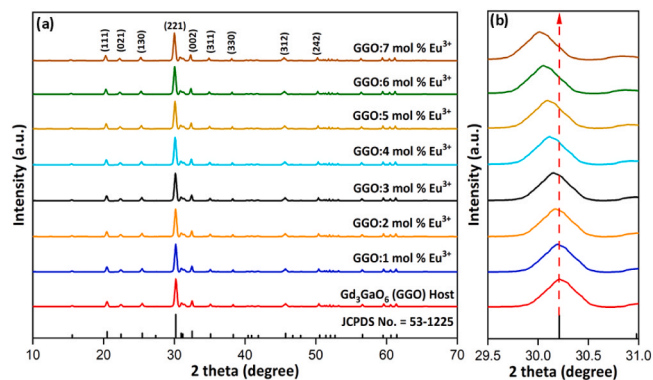


Fig. 2. (a) XRD profiles of Gd_3GaO_6 and $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1-7$ mol%) (b) magnified view highlighting key features of all the samples.

sample was transferred in ceramic crucible and further calcined in high temperature furnace at 1450 °C for 4 h. It was then grinded and used for different characterizations.

3. Characterizations

Various spectroscopic measurements of prepared phosphors were done at room temperature in order to understand the characteristics of the samples. To validate the phase and purity of synthesized substance, XRD patterns were produced on Rigaku Ultima X-ray diffractometer. Nickel crystal was used as filter to obtain only $\text{Cu-K}\alpha$ radiation having wavelength 1.5406 Å and to discard $\text{Cu-K}\beta$ radiations of Cu target. X-ray patterns were investigated by scan speed of 2°/min and angles (2θ) ranging between 10° to 70°. FE-SEM technique was used to ascertain the morphological analysis of desired material performed on Carl Zeiss Sigma 360 instrument. For the confirmation of chemical composition and elemental analysis Ametek EDX spectrometer was employed. In powdered form the assessment of luminescence properties was done on Horiba Jobin YVON Fluorolog Model-FL-3-11 spectrophotometer possessing Xe lamp as an excitation source.

4. Results and discussion

4.1. XRD measurements

Crystal structures of synthesized undoped Gd_3GaO_6 and $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1-7$ mol%) were analysed by powder XRD patterns as shown in Fig. 2(a). XRD analysis revealed prominent diffraction peaks characteristic of orthorhombic structure at 2θ values of 20.435, 22.524, 25.546, 30.206, 32.611, 35.248, 38.407, 45.696 and 50.447 corresponding to (111), (021), (130), (221), (002), (311), (330), (312) and (242) (Fig. 1). The XRD peaks of GGO host sample suggest orthorhombic phase which was further confirmed by the standard reference file JCPDS card no. 53-1225 [29]. Even on incorporation of Eu^{3+} ion in GGO host lattice there is no change in phase of the samples indicating well replacement of Gd^{3+} ions by Eu^{3+} ions.

The inclusion of dopant ion in lattice was further authenticated by estimating radius percentage difference (Δr) among host cation (Gd^{3+}) and dopant ion (Eu^{3+}) by utilizing the relation given in Eq. (1) [30].

$$\Delta r = \frac{|R_B(\text{CN}) - R_D(\text{CN})|}{R_B(\text{CN})} \times 100\% \quad (1)$$

Here, CN, R_B and R_D symbolize coordination number, ionic radii of host ion and dopant ion, respectively. Based on literature studies, a maximum 15 % difference in ionic radii among the dopant and host ions is acceptable for effective doping of activators into an inorganic host matrix [31]. Taking into account the radii of Eu^{3+} ($r = 1.01$ Å, CN = 7) and Gd^{3+} ($r = 1$ Å, CN = 7), the radius percentage difference comes out

Table 1

XRD peak positions (2θ values) and corresponding interplanar spacings (D-values) of Gd_{3-x}GaO₆:xEu³⁺ (x = 1–7 mol%).

Sample	2θ	D-spacing (Å)
GGO:0 mol% Eu ³⁺	30.2062	2.9564
GGO:1 mol% Eu ³⁺	30.1982	2.9571
GGO:2 mol% Eu ³⁺	30.1753	2.9593
GGO:3 mol% Eu ³⁺	30.1525	2.9615
GGO:4 mol% Eu ³⁺	30.1183	2.9648
GGO:5 mol% Eu ³⁺	30.0954	2.9670
GGO:6 mol% Eu ³⁺	30.0498	2.9714
GGO:7 mol% Eu ³⁺	30.0155	2.9747

to be less than 15 % suggesting effective substitution of Gd³⁺ ions by Eu³⁺ ions in GGO host. On introduction of Eu³⁺ ions in Gd₃GaO₆ lattice, X-ray diffraction peaks shift somewhat to lower 2θ values compared to undoped GGO lattice [Fig. 2(b)], according to Bragg equation ($n\lambda = 2d\sin\theta$). The results are consistent with incorporation of larger Eu³⁺ in replacement to smaller Gd³⁺ ions.

Table 1 entails D-spacing values computed using Bragg's equation for most intense diffraction peak for different concentration of dopant ion. Scherrer's Eq. (2), as presented below was applied to determine crystallite size of as-prepared samples and values are tabulated in Table 2.

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (2)$$

In this equation, D denotes average particle size, k is a constant, $\lambda = 0.15416$ nm denotes wavelength of X-rays used, θ refers to diffraction angle and β signifies full width at half maximum of diffraction peak [32, 33]. The crystallite size of Gd₃GaO₆ and Gd_{3-x}GaO₆:xEu³⁺ (x = 1–7 mol %) was found to be in the range of 20–32 nm. Dislocation density (δ) refers to total length of dislocations per unit volume of crystalline material and is determined by applying formula given in Eq. (3) [34].

$$\delta = \frac{1}{D^2} \quad (3)$$

Furthermore, to calculate crystallite size another expression known as Williamson-Hall (W-H) was used which includes lattice strain. The total peak broadening (β_T) arises from the combined contributions of crystallite size broadening (β_D) and microstrain broadening (β_ϵ) and is formulated in Eqs. (4) and (5) [35,36].

$$\beta_T = \beta_D + \beta_\epsilon \quad (4)$$

$$\beta = \frac{K\lambda}{D\cos\theta} + 4\epsilon \tan\theta \quad (5)$$

On reorganizing Eq. (5) we get,

$$\beta\cos\theta = \frac{K\lambda}{D} + 4\epsilon\sin\theta \quad (6)$$

Where, symbol ϵ indicated strain linked to nanoparticles and other symbols are used with their standard meanings as defined earlier. W-H plots were produced with $4\sin\theta$ values on x-axis and $\beta\cos\theta$ values on y-

axis as displayed in Fig. 3.

phosphors.

Crystallite sizes of undoped and doped samples can be derived from intercept ($K\lambda/D$) of linear fit and the values are represented in Table 2. Although the crystallite size values show only minor variation across the different Eu³⁺ concentrations. Such slight changes indicate that the introduction of Eu³⁺ has a limited influence on the overall crystallite growth of the GGO samples. The results suggest that the lattice structure remains largely stable with only minimal effects on crystallite size as the dopant level increases. The crystallite size values calculated from W-H plot are larger than those calculated from Scherrer's formula. This discrepancy is expected because Scherrer formula considers only size broadening and neglects microstrain. In contrast, strain component was included in the W-H formulae which contribute to total broadening.

4.2. Crystal structure and Rietveld refinement

The schematics of crystal structure of Gd₃GaO₆ lattice modelled by Vesta software is shown in Fig. 4. The structure gives information about the presence of luminescent centers and feasibility of dopant ion within the host lattice. The three dissimilar cationic sites and four anionic sites exist in the host matrix involving one gallium [Ga(6)], two gadolinium [Gd(3) and Gd(4)] and four oxygen [O(1), O(2), O(5), O(7)]. As depicted in structure, Gd(3) and Gd(4) resides at 4a and 8b crystallographic sites surrounded by seven oxygen atoms exhibiting [Gd(3)O₇] and [Gd(4)O₇] polyhedral units, correspondingly. Ga(6) occupies 4a site giving rise to [GaO₄] polyhedral unit with coordination number four.

The oxygen atoms O(1), O(2), O(5) and O(7) display different bonding patterns with Gd and Ga. O(1) forms total five bonds with four Gd and one Ga ion and occupies 4a site. Total four bonds are formed by

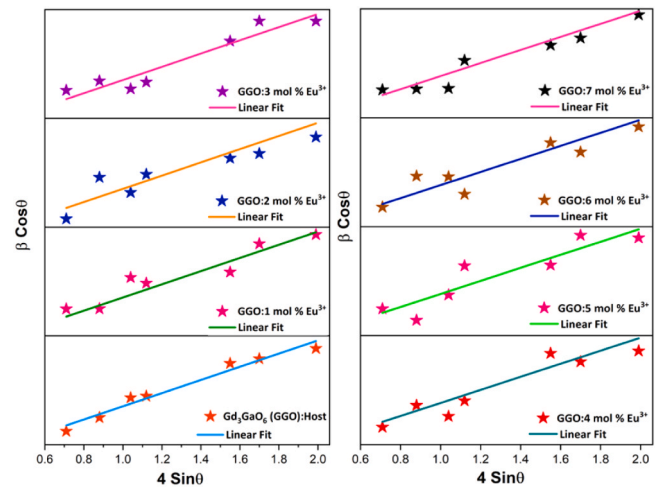


Fig. 3. Williamson-Hall (W-H) plots for Gd₃GaO₆ host and Gd_{3-x}GaO₆:xEu³⁺ (x = 1–7 mol%).

Table 2

Computed XRD parameters (crystallite size, microstrain and dislocation density) of synthesized Gd_{3-x}GaO₆:xEu³⁺ (x = 1–7 mol%) phosphors.

Sample (GGO)	2θ	FWHM	Crystallite Size (nm)		Microstrain ϵ	Dislocation Density
			Scherrer's	W-H		
GGO:0 mol% Eu ³⁺	30.2062	0.34724	24.75	43.35	5.61	1.63×10^{-3}
GGO:1 mol% Eu ³⁺	30.1982	0.36421	23.60	42.24	5.89	1.80×10^{-3}
GGO:2 mol% Eu ³⁺	30.1753	0.39246	21.90	39.93	6.35	2.09×10^{-3}
GGO:3 mol% Eu ³⁺	30.1525	0.32369	26.55	45.72	5.24	1.42×10^{-3}
GGO:4 mol% Eu ³⁺	30.1183	0.31932	26.91	45.97	5.18	1.38×10^{-3}
GGO:5 mol% Eu ³⁺	30.0954	0.27573	31.16	49.38	4.47	1.03×10^{-3}
GGO:6 mol% Eu ³⁺	30.0498	0.26986	31.84	49.91	4.39	9.86×10^{-4}
GGO:7 mol% Eu ³⁺	30.0155	0.37149	23.12	42.56	6.05	1.87×10^{-3}

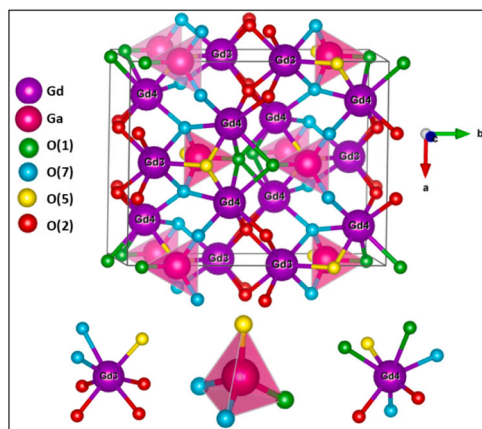


Fig. 4. Visualization of crystal structure of Gd_3GaO_6 illustrating coordinative environment of individual cations.

Table 3

Crystallographic parameters of Gd_3GaO_6 (GGO) host exhibiting orthorhombic crystal system.

Atoms	x	y	z	U	Occ.	Site	Sym.
Gd (3)	0.00000	0.39640	0.42369	0.205	1.000	4a	m.
Gd (4)	0.19300	0.10567	0.46705	0.205	1.000	8b	1
Ga (6)	0.00000	0.26010	0.17000	0.205	1.000	4a	m.
O (1)	0.00000	0.04660	0.12500	0.205	1.000	4a	m.
O (2)	0.00000	0.21400	0.67100	0.205	1.000	4a	m.
O (5)	0.34000	0.01890	0.20600	0.205	1.000	8b	1
O (7)	0.15590	0.26010	0.17000	0.205	1.000	8b	1

O(2) and O(7) which are located at 4a and 8b sites, respectively coordinating three bonds with Gd and one with Ga. Lastly, O(5) like O(1) occupying 8b crystal site forms total four bonds, one with Ga and three with Gd ions. Table 3 presents the information on atomic coordinates and displacement characteristics of host material. A characteristic crystal pattern of the prepared material was formulated by means of rietveld refinement approach to analyze the introduction of Eu^{3+} ion into GGO host.

Fig. 5(a) and (b) portrays rietveld refinement profiles of XRD patterns for pure Gd_3GaO_6 and optimized $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 4$ mol%) samples. In the given plots, black crosses indicate observed pattern and red line symbolises calculated fit. Green vertical lines show Bragg reflections and blue lines at the bottom indicate discrepancy among observed and calculated data. Refinement parameters of undoped and doped $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 4$ mol%) phosphors are demonstrated in Table 4. Both samples were found to be crystallized in orthorhombic

system with $\text{Cmc}2_1$ space group. The refinement results showed R-factors and goodness factor corresponding to undoped GGO ($R_p = 4.192\%$, $R_{wp} = 5.223\%$ and $\chi^2 = 2.104$) and $\text{GGO}:4 \text{ mol}\% \text{Eu}^{3+}$ ($R_p = 4.562\%$, $R_{wp} = 5.756\%$ and $\chi^2 = 2.725$) confirming a close fit of experimental diffraction patterns with that of calculated patterns. Considering the data in Table 4, unit cell parameter (a, b, c) and volume for doped sample was greater as compared to undoped sample which was attributed to replacement of small size host cation by larger Eu^{3+} ion.

4.3. FESEM and EDAX analysis

The surface morphology and chemical composition of the nanoparticles for $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1, 4$ and 7 mol%) sample were evaluated through FE-SEM and EDX spectroscopy. The FE-SEM images of optimum sample are represented in Fig. 6 which reveals random and agglomerated distribution of particles. Observation of micrographs depicts irregular morphology with particle size confined to the range of 60–80 nm. The relationship between particle morphology and Eu^{3+} photoluminescence has been widely reported in the literature.

Previous studies have demonstrated that particle size and the surface-area-to-volume ratio can influence emission intensity and asymmetry due to variations in surface-related non-radiative processes [37–39]. Although the morphological changes in the considered samples

Table 4

Rietveld refinement constraints of Gd_3GaO_6 (GGO) and $\text{Gd}_{2.96}\text{GaO}_6:4 \text{ mol}\% \text{Eu}^{3+}$ phosphor.

Refinement Parameters	Refinement values of Gd_3GaO_6	Refinement values of $\text{Gd}_{2.96}\text{GaO}_6:4 \text{ mol}\% \text{Eu}^{3+}$
Formula weight	637.47	637.26
Crystal system	Orthorhombic	Orthorhombic
2 θ interval	10–70 °	10–70 °
Bravais lattice	C	C
No. of space group	36	36
Hermann-Mauguin symbol	$\text{Cmc}2_1$	$\text{Cmc}2_1$
Hall symbol	C2c-2	C2c-2
Laue class	mmm	mmm
Point group	mm2	mm2
Lattice symbol	oC	oC
Centro-symmetry	Acentric	Acentric
Formula unit (Z)	4	4
Unit Cell Parameters	a = 8.9278 Å, b = 11.3859 Å, c = 5.5060 Å $\alpha = \beta = \gamma = 90^\circ$	a = 8.9885 Å, b = 11.4534 Å, c = 5.5654 Å $\alpha = \beta = \gamma = 90^\circ$
Volume (Å ³)	559.6906	572.9517
R-factors	$R_p = 4.192\%$, $R_{wp} = 5.223\%$	$R_p = 4.562\%$, $R_{wp} = 5.756\%$
Density (g/cm ³)	7.565	7.387
χ^2	2.104	2.725

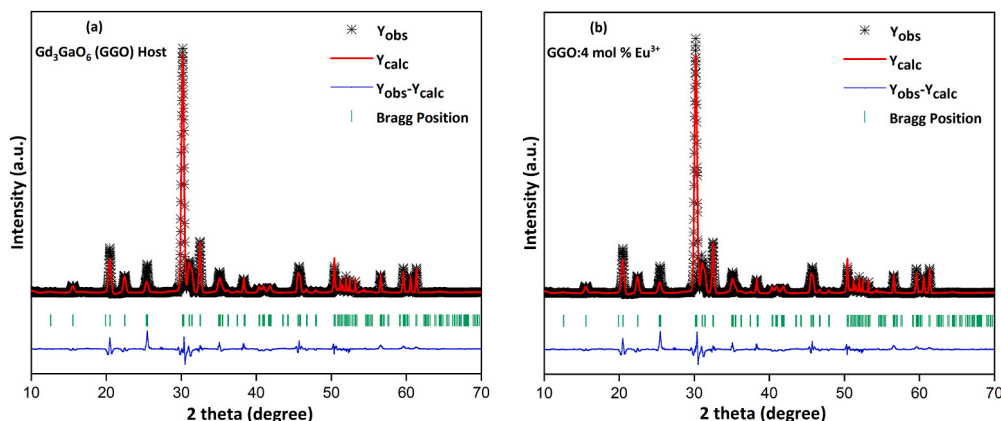


Fig. 5. (a) Rietveld refined XRD profile of Gd_3GaO_6 host. (b) Rietveld refined XRD profile of $\text{Gd}_{2.96}\text{GaO}_6:4 \text{ mol}\% \text{Eu}^{3+}$ sample.

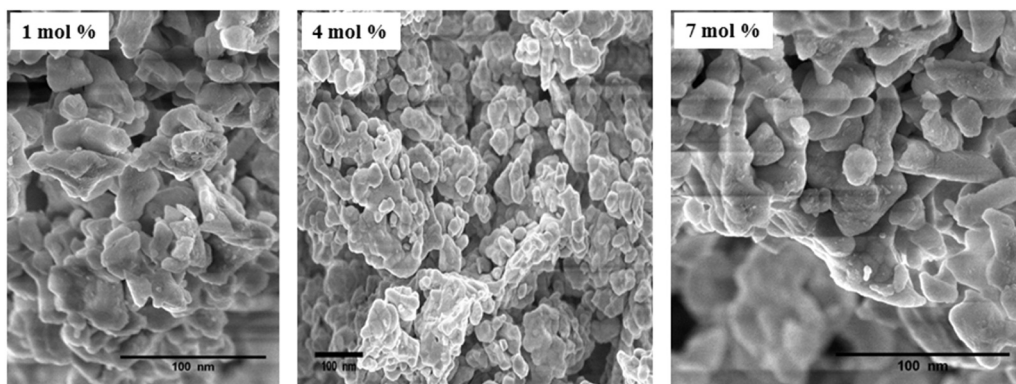


Fig. 6. FESEM images of $\text{Gd}_{3-x}\text{GaO}_6:x\text{Eu}^{3+}$ ($x = 1, 4$ and 7 mol%) showing surface morphology.

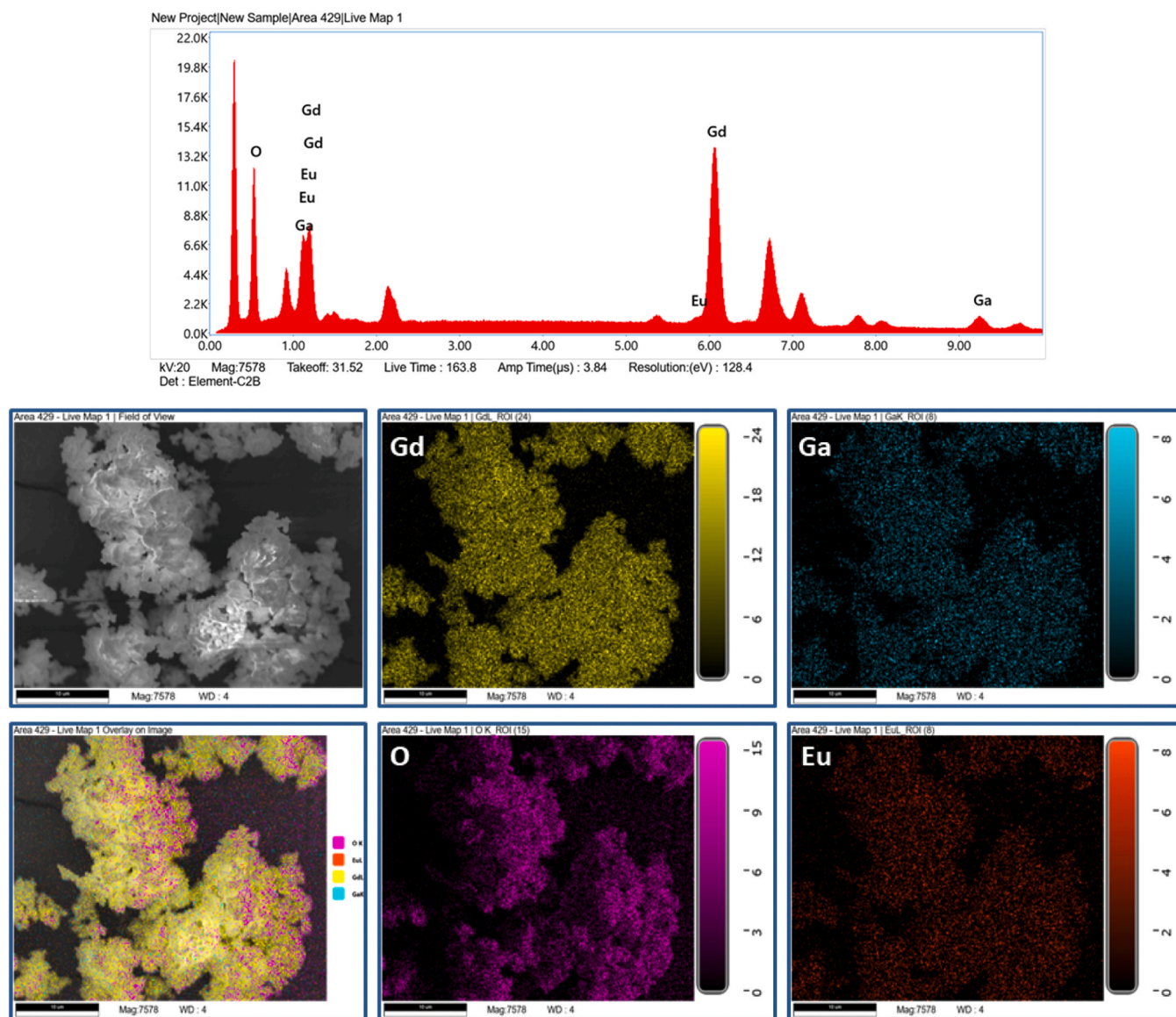


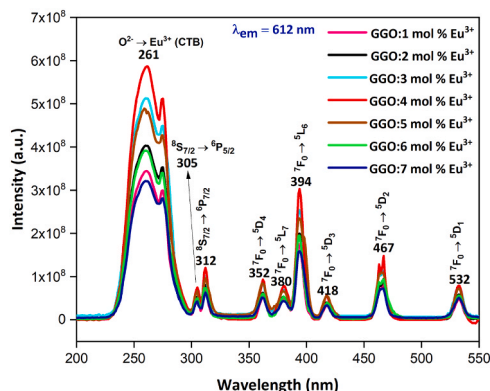
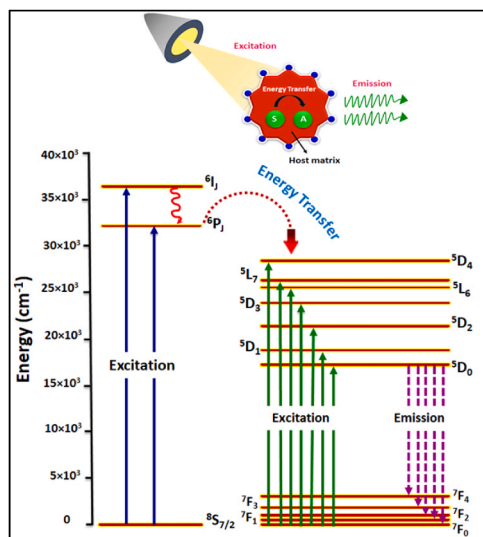
Fig. 7. (a) EDX spectrum of the optimized $\text{Gd}_{2.96}\text{GaO}_6:4 \text{ mol\% Eu}^{3+}$ nanophosphor. (b) EDX mapping images for $\text{Gd}_{2.96}\text{GaO}_6:4 \text{ mol\% Eu}^{3+}$ nanophosphor displaying spatial distribution of Gd, Ga, Eu and O elements.

are modest, these works support the idea that even small differences in particle size or surface characteristics can affect Eu^{3+} luminescence. As seen from Fig. 7(a), the EDX spectra of optimized sample represents

various peaks of elemental constituents-gadolinium (Gd), gallium (Ga), oxygen (O) and europium (Eu) consistent with designed composition of sample. There are no additional peaks in EDX spectra confirming the

Table 5Atomic and weight percentages for $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 4$ mol%) phosphor.

Element	Series	Atomic %	Weight %
Gadolinium (Gd)	L-series	27.4	73.6
Gallium (Ga)	K-series	5.4	6.4
Oxygen (O)	K-series	66.6	18.2
Europium (Eu)	L-series	0.7	1.8

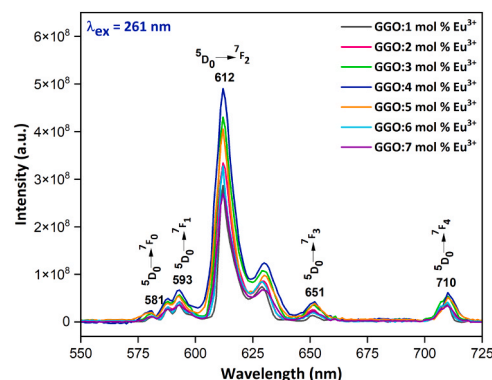
**Fig. 8.** Excitation spectra of $\text{Gd}_{3-x}\text{GaO}_6:\text{x mol \% Eu}^{3+}$ ($x = 1\text{--}7$ mol %) phosphors.**Fig. 9.** Depiction of electronic transitions in Eu^{3+} and energy transfer from Gd^{3+} ions under UV excitation.

formation of pure phase of prepared phosphor. The EDX mapping technique was utilized for examination of chemical homogeneity of prepared sample. Fig. 7(b) illustrates elemental maps of Ga, Gd, O and Eu demonstrating homogenous distribution of europium ion in the host. The elemental weight percentage and atomic percentage of the phosphors are displayed in Table 5.

4.4. Photoluminescence study

4.4.1. Analysis of excitation and emission spectra

Fig. 8 demonstrates PL excitation spectra of $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1\text{--}7$ mol%) monitored at emission wavelength of 612 nm. At 261 nm, a broad charge transfer peak was exhibited corresponding to charge transfer from O^{2-} to Eu^{3+} ($2p$ to $4f$) [40]. The excitation spectrum entails various peaks due to dopant Eu^{3+} and Gd^{3+} ion. A strong

**Fig. 10.** Emission spectra of $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1\text{--}7$ mol%) nanophosphors.

peak centered at 275 nm arising from the transition $8S_{7/2} \rightarrow 6I_{11/2}$ and weak intensity peak at 305 nm and 312 nm arising from $8S_{7/2} \rightarrow 6P_{5/2}$ and $8S_{7/2} \rightarrow 6P_{7/2}$, respectively were due to absorption of Gd^{3+} ions present in the lattice [41,42]. The high intensity of peak at 275 nm indicates efficient energy transfer from Gd^{3+} to Eu^{3+} ions. The other characteristic peaks observed in the excitation spectra in 320–550 nm wavelength region resulting from intra-configurational $4f\text{--}4f$ transitions in Eu^{3+} ion. These peaks are displayed at 352 nm, 380 nm, 394 nm, 418 nm, 467 nm and 532 nm corresponding to $7F_0 \rightarrow 5D_4$, $7F_0 \rightarrow 5L_7$, $7F_0 \rightarrow 5D_3$, $7F_0 \rightarrow 5D_2$ and $7F_0 \rightarrow 5D_1$ transitions [43].

Fig. 9 displays a characteristic energy-transfer schematics for PL emissions of Eu^{3+} and Gd^{3+} ions excited at 261 nm. Due to its half-filled $4f$ configuration, Gd^{3+} has a highly stable $8S_{7/2}$ ground state. Gd^{3+} ions can be excited by ultraviolet light at a wavelength of 261 nm promoting electrons from ground state $8S_{7/2}$ to higher $6I_J$ energy states. These excited levels lose energy non-radiatively allowing the ions to relax into lower-energy $6P_J$ ($J = 7/2$ and $5/2$) states. The relaxation of electrons in Gd^{3+} ions from $6P_J$ levels to ground state can occur through two possible routes: energy transfer to Eu^{3+} ($5D_4$) or photon emission corresponding to $6P_J \rightarrow 8S_{7/2}$ transition. Energy transfer from Gd^{3+} to Eu^{3+} is facilitated due to small energy gap among $6P_J$ states of Gd^{3+} and $5D_4$ states of Eu^{3+} . Consequently, trivalent europium ions possess multiple excited levels; $5D_4$, $5L_7$, $5L_6$, $5D_3$, $5D_2$ and $5D_1$ above ground state $7F_0$. Following excitation, Eu^{3+} ions undergo non-radiative relaxation to $5D_0$ level from which characteristic emissions are observed. The emission spectra of Gd_3GaO_6 doped with trivalent europium ions was obtained under 261 nm excitation wavelength.

Fig. 10 represents several distinguishable bands in 580–720 nm wavelength range of visible region. The emission spectra entail peaks positioned in green region between 500 and 595 nm located at 581 nm, 593 nm and in red region located at 612 nm and 651 nm. The pronounced peak centred at 612 nm was observed corresponding to $5D_0 \rightarrow 7F_2$ transition. The $5D_0 \rightarrow 7F_2$ transition is hypersensitive with its intensity being highly dependent on the local site symmetry and ligand field experienced by Eu^{3+} ions [44]. All the synthesized samples exhibit narrow emission bands attributed to $5D_0 \rightarrow 7F_J$ ($J = 0, 1, 2, 3, 4$) transitions. Emission peaks at 581 nm, 593 nm, 612 nm, 651 nm and 710 nm correspond to $5D_0 \rightarrow 7F_0$, $5D_0 \rightarrow 7F_1$, $5D_0 \rightarrow 7F_2$, $5D_0 \rightarrow 7F_3$ and $5D_0 \rightarrow 7F_4$, respectively [45].

Transitions $5D_0 \rightarrow 7F_{1,3}$ are categorised as allowed MD transitions and are largely insensitive to ligand field surrounding the Eu^{3+} ions whereas $5D_0 \rightarrow 7F_{2,4}$ are electric dipole forbidden transitions [46]. The relative strength of electric and magnetic dipole transitions is a measure of local site symmetry around Eu^{3+} ions. When $5D_0 \rightarrow 7F_1$ magnetic dipole transition is dominant, it reflects the presence of inversion symmetry at Eu^{3+} site. In contrast, stronger electric dipole emission relative to magnetic dipole emission implies absence of inversion symmetry [47]. The $5D_0 \rightarrow 7F_1$ transition is responsible for orange luminescence whereas $5D_0 \rightarrow 7F_2$ transition gives typical red luminescence. In current study,

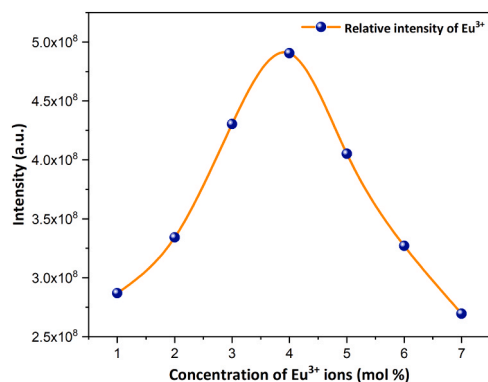


Fig. 11. Emission intensity dependence on Eu^{3+} concentration in $\text{Gd}_{3-x}\text{GaO}_6:x\text{Eu}^{3+}$ ($x = 1-7$ mol%) phosphors.

$^5\text{D}_0 \rightarrow ^7\text{F}_2$ electric dipole transition (red luminescence) situated at 612 nm predominate over $^5\text{D}_0 \rightarrow ^7\text{F}_1$ magnetic dipole transition (orange luminescence) indicating that Eu^{3+} ions placed at a lattice site lacking center of symmetry. To evaluate the performance of $\text{GdO}_3:\text{Eu}^{3+}$, its luminous intensity was compared with representative commercial red phosphor $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$. Under 261 nm excitation, $\text{Gd}_{3-x}\text{GaO}_6:x\text{Eu}^{3+}$ ($x = 4$ mol%) phosphor exhibits higher luminous intensity than that of $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ [48].

4.4.2. Concentration quenching

The PL intensity of Eu^{3+} doped Gd_3GaO_6 host lattice quenches on rising Eu^{3+} ion concentration above 4 mol% which is maximum doping concentration after which the intensity decreases with dopant concentration as given in Fig. 11. Concentration quenching is the phenomenon responsible for this decrease of intensity after an optimum Eu^{3+} ion concentration.

It is assumed that at lower dopant concentration there is negligible interaction among the ions and at elevated dopant levels the spacing between dopant ions is small enough for the interaction and non radiative energy transfer among them results in quenching of PL emission intensity. In this process, the excitation energy is transferred from one ion to other randomly and finally reaches to acceptor ions leading to concentration quenching [49,50]. There are various interactions accountable for ion-ion relaxation including radiation re-absorption, exchange interactions and electric multipolar interactions. Radiation re-absorption occurs when transition energies of donor and acceptor ions are distinct. This energy difference leads to emission or absorption of phonons particularly when emission spectra of sensitizer and activator ion overlap [51]. Non radiative energy transfer through radiation reabsorption is unlikely to occur in the present study as there is no broad overlap among emission and excitation spectra. Exchange interactions come into existence when two dopant ions are close to one another i.e. average critical energy transfer distance (R_c) is considerably smaller than 5 Å. Critical distance (R_c) is a significant parameter to understand the phenomenon of energy transfer. R_c for energy transfer between $\text{Eu}^{3+}-\text{Eu}^{3+}$ ions was assessed by Blasse's Eq. (7).

$$R_c = 2 \left(\frac{3V}{4\pi x_c Z} \right)^{\frac{1}{3}} \quad (7)$$

Where, x_c denotes optimal dopant ion concentration at which quenching occurs, V signifies unit cell volume and Z denotes number of crystallographic cationic positions available in unit cell [52]. R_c was calculated by substituting the values of $V = 572.9517 \text{ Å}^3$, $Z = 4$, $x_c = 0.04$ in above equation and comes out to be about 18.98 Å. R_c value exceeded 5 Å suggesting that quenching is not governed by exchange interaction mechanism referring to theory of radiationless energy transfer between RE elements given by Van Uitert. Thus, multipole-multipole interactions

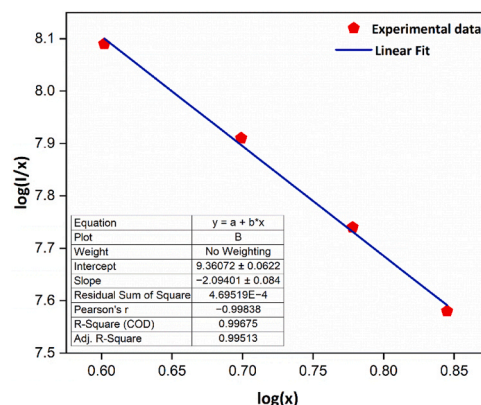


Fig. 12. Graph of synthesized materials with linear fitting applied after concentration quenching.

play active role for non radiative energy transfer in Eu^{3+} doped Gd_3GaO_6 host matrix. Eq. (8), based on Dexter's theory was utilized to compute specific type of multi-polar interactions [53].

$$\frac{I}{x} = k \left[1 + \beta(x)^{\theta/3} \right]^{-1} \quad (8)$$

In this context, I/x is defined as emission intensity per unit concentration of Eu^{3+} ions, variable θ denotes multi-polar interactions, k and β serves as constants.

The value of $\theta = 6, 8$ and 10 corresponds to dipole-dipole, dipole-quadrupole and quadrupole-quadrupole interactions, respectively. Eq. (8) can be modified to Eq. (9) when $\beta(x)^{\theta/3} \gg 1$ [54,55].

$$\log\left(\frac{I}{x}\right) = -\frac{\theta}{3} \log x + k' \quad (9)$$

Where, k' is a constant. The $\log(I/x)$ is plotted against $\log(x)$ and is represented in Fig. 12. A linear correlation exists between $\log(I/x)$ and $\log(x)$ where slope is determined to be -2.094 . Consequently, θ was calculated to be 6.282 which is approximately 6. On the basis of result, the main cause of energy transfer (concentration quenching) amongst $\text{Eu}^{3+}-\text{Eu}^{3+}$ ions in Gd_3GaO_6 phosphor appears to be dipole-dipole interaction.

4.5. Judd Ofelt analysis

Judd-Ofelt (JO) theory is extensively applied approach for interpreting parity forbidden

f-f transitions. Through determination of J-O parameters, valuable information can be obtained about local site symmetry and bonding in Eu^{3+} doped host lattice. This theory is applied for computation of various radiative characteristics such as radiative transition probability (A_R), branching ratio (β) and asymmetry ratio (R) for various levels of dopant ions using photoluminescence emission spectra. The three transitions $^5\text{D}_0 \rightarrow ^7\text{F}_J$ ($J = 2, 4$ and 6) enable independent determination of intensity parameters Ω_2, Ω_4 and Ω_6 , respectively. Since intensity of magnetic dipole (MD) $^5\text{D}_0 \rightarrow ^7\text{F}_1$ transition is insensitive to crystal field environment, the related parameters are derived from PL spectra and this transition is used as a standard reference for the analysis of ED $^5\text{D}_0 \rightarrow ^7\text{F}_J$ ($J = 2, 4$ and 6) transitions. Based on Judd-Ofelt approach, spontaneous emission probability of MD ($^5\text{D}_0 \rightarrow ^7\text{F}_1$) transition is described by the expression given in Eq. (10).

$$A_{01}^{MD} = \frac{64\pi^4 \nu^3}{3h(2J+1)} \eta^3 S_{MD} \quad (10)$$

In above formula, h , η , $(2J+1)$, ν and S_{MD} variables represent planck's constant, refractive index, degeneracy, wavenumber and strength of $^5\text{D}_0 \rightarrow ^7\text{F}_1$ MD transition, respectively [56]. Furthermore,

Table 6

Judd-Ofelt parameters and asymmetry ratio for GGO:Eu³⁺ phosphors with varying Eu³⁺ content (x = 1–7 mol%).

Sample	$\Omega_2 (\times 10^{20} \text{ cm}^2)$	$\Omega_4 (\times 10^{20} \text{ cm}^2)$	Asymmetric ratio (R)
1 mol% Eu ³⁺	11.679 ± 0.0265 (11.421)	2.236 ± 0.0125 (2.090)	8.250 (7.973)
2 mol% Eu ³⁺	11.491 ± 0.0312 (11.797)	1.957 ± 0.0328 (1.871)	8.246 (8.252)
3 mol% Eu ³⁺	11.368 ± 0.0354 (11.021)	1.938 ± 0.0225 (2.097)	7.869 (7.639)
4 mol% Eu ³⁺	11.845 ± 0.0359 (11.753)	1.736 ± 0.0452 (1.810)	8.379 (8.235)
5 mol% Eu ³⁺	11.654 ± 0.0567 (11.462)	2.394 ± 0.0321 (2.125)	9.086 (8.018)
6 mol% Eu ³⁺	11.791 ± 0.0398 (11.904)	2.083 ± 0.0147 (2.216)	8.847 (8.336)
7 mol% Eu ³⁺	11.286 ± 0.0591 (11.021)	1.865 ± 0.0276 (2.097)	7.825 (7.639)

Theoretical values are given in ().

radiative transition rates for ED ⁵D₀→⁷F₂, ⁵D₀→⁷F₄ and ⁵D₀→⁷F₆ transitions were estimated by Eq. (11).

$$A_{0J}^{ED} = \frac{64\pi^4 \nu^3 e^2}{3h(2J+1)} \frac{n(n^2+2)^2}{9} \Omega_2 \langle \Psi J | U \lambda | \Psi J \rangle^2 \quad (11)$$

Where, e signifies charge on an electron and $\langle \Psi J | U \lambda | \Psi J \rangle^2$ represents reduced matrix elements and has non zero values for transitions from ⁵D₀ state to ⁷F_J ($J = 2, 4$ and 6). The values of $\langle \Psi J | U \lambda | \Psi J \rangle^2$ for $J = 2, 4$ and 6 are 0.0032, 0.0023 and 0.00023, correspondingly [57]. The values of JO parameters (Ω_2 and Ω_4) for different doping concentration of Eu³⁺ ion in GGO host lattice are depicted in Table 6.

The intensity parameter Ω_2 being a short-range sensitive parameter reflects distortions in immediate environment of Eu³⁺ ion and is closely related to strength covalency between Eu³⁺ center and coordinating ligands. Ω_4 on the other hand gives insight of rigidity or stiffness of host lattice representing long range interactions [58]. The Ω_6 value is not taken into account in this particular study as ⁵D₀→⁷F₆ transition lies in IR region and is not detected in emission spectra. The approach established by A. Cirić *et al.* gives a theoretical pathway for determination of J-O parameters from JOES software [59]. Both experimental and theoretical optical characteristics are summarized in Table 6. The experimental outcomes for all prepared samples are strongly related to theoretical data. The consistent pattern of $\Omega_2 > \Omega_4$ for GGO:xEu³⁺ ($x = 1-7$ mol%) suggests covalent interaction among Eu³⁺ ions and ligands which further indicates low symmetry environment of trivalent europium ions in Gd₃GaO₆. The total radiative transition probability (A_R) is the sum of all individual radiative rates (A_{0J}) for ⁵D₀→⁷F_J transition and is expressed as Eq. (12).

$$A_R = \sum_J A_{0J} = A_{01} \frac{\nu_{01}}{I_{01}} \sum_{J=0}^4 \frac{I_{0J}}{\nu_{0J}} \quad (12)$$

The terms ν_{01} and ν_{0J} in above formula symbolizes energy

Table 7

Radiative transition probabilities (A_r) and branching ratio (β) measured for Gd_{3-x}GaO₆:xEu³⁺ ($x = 1-7$ mol%) phosphors.

Sample	Branching ratio (β)			Radiative transition probabilities (A_r in sec ⁻¹)		
	$\beta (^5D_0 \rightarrow ^7F_1)$	$\beta (^5D_0 \rightarrow ^7F_2)$	$\beta (^5D_0 \rightarrow ^7F_4)$	$A_r (^5D_0 \rightarrow ^7F_1)$	$A_r (^5D_0 \rightarrow ^7F_2)$	$A_r (^5D_0 \rightarrow ^7F_4)$
1 mol% Eu ³⁺	0.095 (0.105)	0.786 (0.823)	0.073 (0.072)	115.217	899.703	78.516
2 mol% Eu ³⁺	0.095 (0.103)	0.788 (0.834)	0.057 (0.063)	115.239	932.057	70.057
3 mol% Eu ³⁺	0.098 (0.109)	0.775 (0.817)	0.067 (0.074)	115.542	868.418	78.431
4 mol% Eu ³⁺	0.093 (0.104)	0.782 (0.835)	0.055 (0.061)	115.564	927.575	67.216
5 mol% Eu ³⁺	0.086 (0.106)	0.783 (0.818)	0.062 (0.072)	115.707	906.041	78.803
6 mol% Eu ³⁺	0.089 (0.101)	0.784 (0.826)	0.071 (0.073)	115.037	937.504	82.901
7 mol% Eu ³⁺	0.098 (0.102)	0.766 (0.821)	0.071 (0.074)	115.564	903.288	81.774

Theoretical values are given in ().

barycenters for transition ⁵D₀ to ⁷F₁ and ⁵D₀ to ⁷F_J whereas I_{0J} and A_{0J} corresponds to area under PL emission curve and Einstein coefficient of ⁵D₀→⁷F_J transition, individually [60]. The results of radiative transition probability (A_R) for GGO:xEu³⁺ ($x = 1-7$ mol%) are displayed in Table 7.

One of the key parameters of photoluminescence related emission tunability is asymmetry ratio which is described as intensity proportion of ED (⁵D₀→⁷F₂) and MD (⁵D₀→⁷F₁) transition and is a measure of covalency between Eu³⁺ and O²⁻. Mathematically, the values of asymmetry ratio for various Eu³⁺ doped samples were evaluated using Eq. (13) and the results are tabulated in Table 6 [61].

$$R = \frac{I(^5D_0 \rightarrow ^7F_2)}{I(^5D_0 \rightarrow ^7F_1)} \quad (13)$$

Larger difference in the intensities of two transitions indicates higher degree of asymmetry and covalency. The ⁵D₀→⁷F₂ transition of Eu³⁺ ion is forbidden at sites with an inversion centre according to Judd Ofelt theory. Consequently, deviations from centrosymmetry in local environment leads to enhanced luminescence intensity of this transition owing to the relaxation in selection rule. In other words, emission intensity is directly related to the degree of distortion from inversion symmetry [62]. In present study, in all the samples under consideration, ⁵D₀→⁷F₂ peak is more intense than ⁵D₀→⁷F₁ resulting into high red to orange (R/O) ratio which is further correlated with the findings of J-O parameters.

4.6. Branching ratio

Table 7 contains the branching ratio (β) values of various Eu³⁺ doped GGO samples for ⁵D₀→⁷F_{1,2,4} transitions. Branching ratio itself is defined as the fraction of each transition to overall integrated area of emission spectrum. The values are derived from Eq. (14) which expresses the ratio of individual peak's integrated area to the combined integrated area of aforementioned peaks [63].

$$\beta = \frac{A_{0J}}{\sum A_{0J}} \quad (14)$$

Here, A_{0J} signifies integrated area of particular transition in emission spectra of GGO:xEu³⁺ ($x = 1-7$ mol%).

4.7. Luminescence lifetime

The decay profile was analysed to understand the luminescence mechanism and determine the luminescent centres. Fig. 13(a) displays the decay curve obtained for optimized Gd_{3-x}GaO₆:xEu³⁺ ($x = 4$ mol%) phosphor monitored at an excitation of 261 nm and emission wavelength of 612 nm. The experimental results fit accurately to double exponential decay function as defined in Eq. (15) [64,65].

$$I_t = I_0 + A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) \quad (15)$$

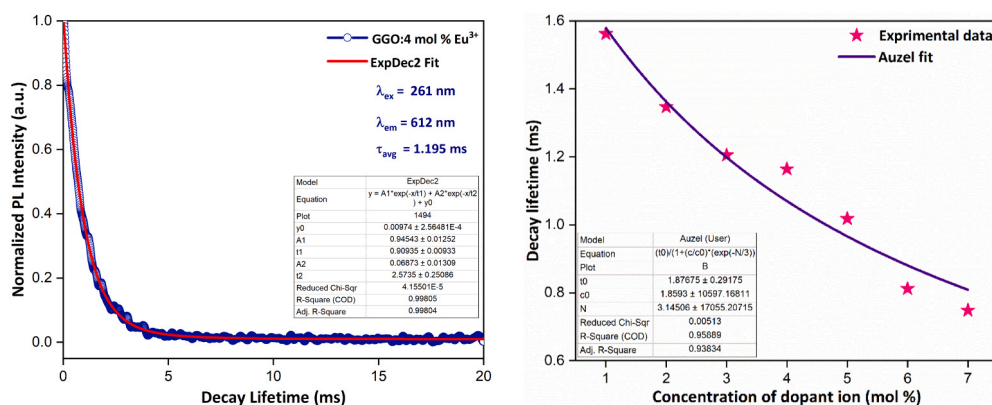


Fig. 13. (a) Decay lifetime curve of Gd_{2.96}GaO₆:4 mol% Eu³⁺ phosphor. (b) Auzel fitting curve of all prepared samples.

Table 8

Computed decay lifetimes and quantum efficiencies of Gd_{3-x}GaO₆:xEu³⁺ (x = 1–7 mol%) phosphors.

Sample	τ_{avg} (ms)	η (%)
GGO:1 mol% Eu ³⁺	1.562	83.08
GGO:2 mol% Eu ³⁺	1.347	71.65
GGO:3 mol% Eu ³⁺	1.205	64.09
GGO:4 mol% Eu ³⁺	1.195	63.56
GGO:5 mol% Eu ³⁺	1.049	55.79
GGO:6 mol% Eu ³⁺	0.812	43.19
GGO:7 mol% Eu ³⁺	0.638	33.94

In this equation, t represents time variable, I_0 and I_t refer to the luminescence intensities at $t = 0$ and at time t , respectively, τ_1 and τ_2 correspond to decay lifetimes of two exponential components whereas A_1 and A_2 denote fitting constants. The values of decay time τ_1 and τ_2 were measured to be 0.909 ms and 2.573 ms, respectively. Furthermore, the average lifetime was determined employing the following Eq. (16) [66].

$$\tau_{avg} = \frac{(A_1 \tau_1^2 + A_2 \tau_2^2)}{(A_1 \tau_1 + A_2 \tau_2)} \quad (16)$$

The average decay lifetime for the considered sample was estimated to be 1.195 ms. The average lifetimes of all the doped samples are summarized in Table 8.

It was observed that decay times decrease with increase in Eu³⁺ ion concentration. This occurs due to closer proximity of ions which facilitates faster energy transfer introducing an additional decay path with shorter decay time [67]. Using Auzel's fitting method as depicted in

Fig. 13(b), the value of τ_0 was determined to be 1.88 ms.

$$\tau_c = \frac{\tau_0}{1 + \frac{C}{C_0} e^{-N/3}} \quad (17)$$

Here, τ_0 corresponds to intrinsic radiative lifetime, τ_c represents measured fluorescence lifetime, N indicates number of photons and C_0 is a constant term.

Moreover, the quantum efficiency (η) of activated phosphor material was evaluated by Eq. (18) which is defined as the ratio of measured average lifetime to the radiative decay lifetime [68].

$$\eta = \tau_{avg} / \tau_0 \quad (18)$$

The quantum efficiency of synthesized samples was determined and listed in Table 8. The results indicate that the prepared phosphors are well suited for WLED applications.

4.8. Temperature dependent photoluminescence

To investigate the potential of prepared phosphor material for WLED applications, the temperature dependent photoluminescence spectra of the optimized GGO:4 mol% sample were recorded between 298 and 448 K range as illustrated in Fig. 14(a). As depicted in the graph, emission peak position does not change while luminescence of phosphor decreases continuously with rise in temperature owing to thermal quenching effect [69]. For a more comprehensive understanding of the behaviour of thermally activated phosphor a graphical representation of temperature dependent luminous intensity is displayed in Fig. 14(b). It was observed that emission intensity retains 81.3 % of its initial value even at 448 K indicating its superior thermal stability. The activation energy of Gd_{3-x}GaO₆:xEu³⁺ (x = 4 mol%) phosphor was estimated

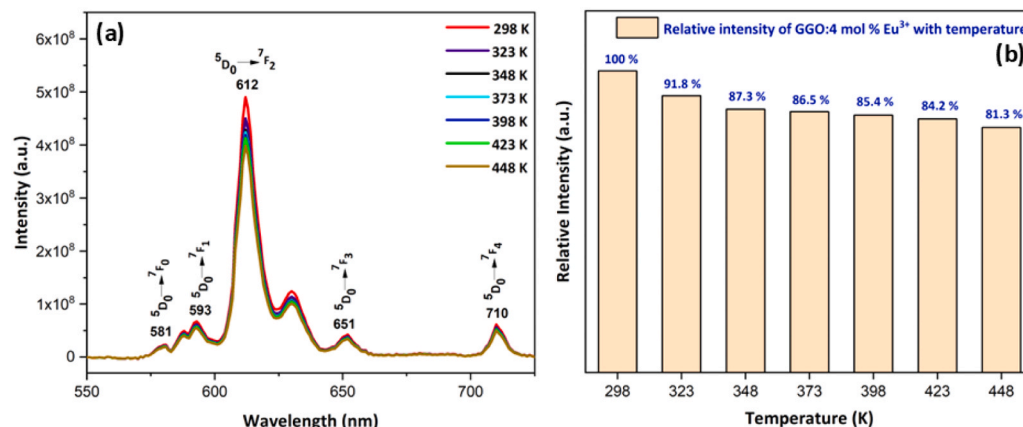


Fig. 14. (a) Temperature dependent photoluminescence and (b) the variation of relative emission intensity with temperature for Gd_{2.96}GaO₆:4 mol% Eu³⁺ phosphor.

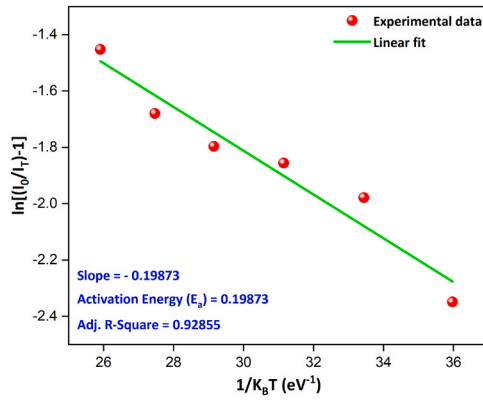


Fig. 15. Linear fit plot for activation energy calculation.

applying Arrhenius relation (19) [70].

$$I_T = \frac{I_0}{1 + A \exp\left(-\frac{E_a}{K_B T}\right)} \quad (19)$$

In the expression, E_a stands for the activation energy of thermal quenching process, I_T denotes PL intensity measured at temperature T , I_0 corresponds to PL intensity at room temperature, K_B represents Boltzmann's constant (8.617105×10^{-5} eV/K) and A is a constant term. Consequently, the dependence of $\ln[(I_0/I_T) - 1]$ on $1/K_B T$ can be formulated as given in Eq. (20) [71].

$$\ln\left(\frac{I_0}{I_T} - 1\right) = \ln A - \frac{E_a}{K_B T} \quad (20)$$

Based on slope of linearly fitted plot (Fig. 15), the activation energy was roughly estimated to be about 0.19873 eV implying high thermal stability of the prepared phosphor at higher temperature range.

4.9. Colorimetric investigations

The standard color space known as CIE 1931 (Commission International de l'Eclairage) is utilized to analyse luminescence behaviour of the phosphor materials and to identify actual emission color of prepared samples. Commonly recognized, CIE diagram expresses composition of any color using three primary colors-red, blue and green. The CIE co-ordinates (x , y) were evaluated utilizing emission spectra of as synthesised phosphors and can be represented by following Eq. (21) [72].

$$x = \frac{X}{X + Y + Z} \quad y = \frac{Y}{X + Y + Z} \quad (21)$$

Here, X , Y and Z are tristimulus values of a given color known as artificial colors and x , y represents CIE 1931 co-ordinates. The calculated values of color coordinates for prepared sample $\text{Gd}_{3-x}\text{GaO}_6:\text{xEu}^{3+}$ ($x = 1-7$ mol%) are listed in Table 9 and represented in color chart of CIE 1931 in Fig. 16. The observed color coordinates (x , y) lie in red region of CIE triangle. Therefore, prepared gallate based phosphor can be used as a promising material for lighting devices.

Another chromaticity coordinates, CIE 1976 (u' , v') were introduced to study photometric characterizations (Fig. 17). These can be established by following Eq. (22) using x , y coordinates [73].

Table 9

CIE (x , y and u' , v') coordinates, CCT (K), color purity and LER for $\text{GGO}:\text{Eu}^{3+}$ phosphors at different Eu^{3+} doping levels.

Sample	CIE Co-ordinates		Color Purity (%)		CCT (K)	LER (lm/W)
	x	y	u'	v'		
GGO:1 mol% Eu^{3+}	0.6628	0.3369	0.4638	0.5303	97.63	275
GGO:2 mol% Eu^{3+}	0.6590	0.3407	0.4568	0.5314	96.60	283
GGO:3 mol% Eu^{3+}	0.6622	0.3375	0.4627	0.5305	97.63	272
GGO:4 mol% Eu^{3+}	0.6623	0.3374	0.4629	0.5305	98.15	277
GGO:5 mol% Eu^{3+}	0.6524	0.3472	0.4451	0.5331	94.81	280
GGO:6 mol% Eu^{3+}	0.6585	0.3412	0.4559	0.5315	96.33	279
GGO:7 mol% Eu^{3+}	0.6620	0.3377	0.4622	0.5306	97.37	274

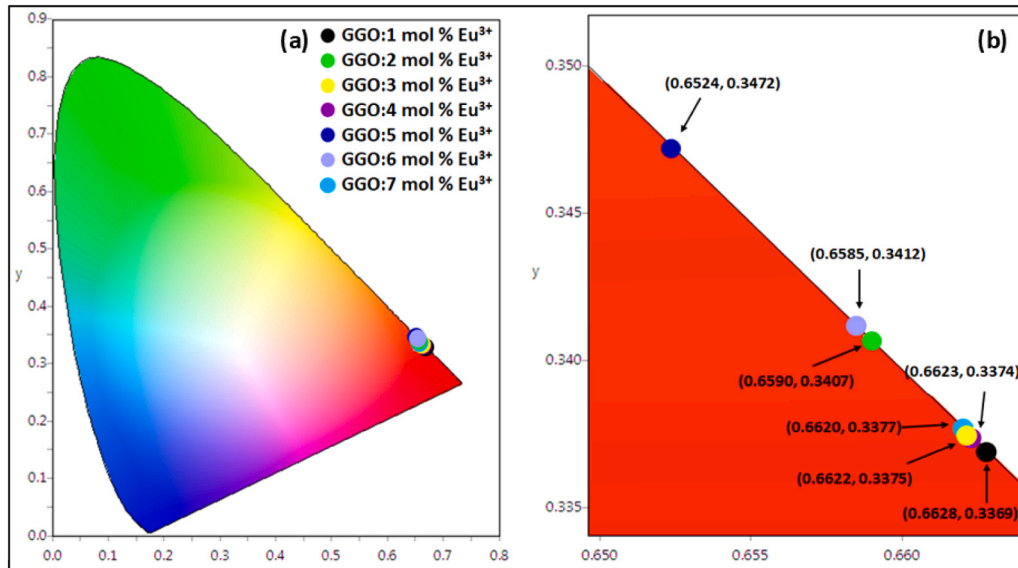


Fig. 16. (a) Representation of chromaticity coordinates (x , y) plotted on CIE 1931 diagram (b) Enlarged view showing detailed spread of coordinates.

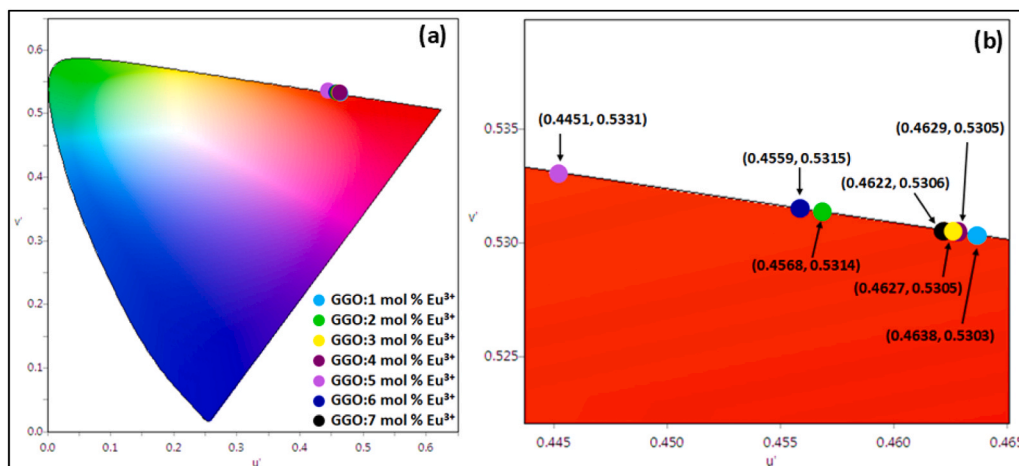


Fig. 17. (a) Representation of chromaticity coordinates (u' , v') plotted on CIE 1976 diagram (b) Enlarged view showing detailed spread of coordinates.

Table 10

Comparative optical performance of the synthesized GGO:Eu³⁺ with Eu³⁺-activated phosphors.

Host lattice	Activator ion	Emission peak (nm)	CIE (x, y)	Color Purity (%)	References
Gd ₃ GaO ₆	Eu ³⁺	612 nm	0.6623, 0.3374	98.15	-
Sr ₂ InTaO ₆	Eu ³⁺	624 nm	0.6265, 0.3693	96.40	[70]
Ba ₃ Lu ₄ O ₉	Eu ³⁺	616 nm	0.6687, 0.3307	98.00	[71]
KBaScSi ₃ O ₉	Eu ³⁺	611 nm	0.6542, 0.3455	98.80	[72]
Y ₂ O ₃	Eu ³⁺	611 nm	0.6590, 0.3320	96.00	[73]
Na ₂ CaSiO ₄	Eu ³⁺	595 nm	0.5840, 0.4210	95.65	[74]

$$u' = \frac{4x}{-2x + 12y + 3} \quad v' = \frac{9y}{-2x + 12y + 3} \quad (22)$$

To determine the color purity and perfect color emission of prepared phosphor, chromaticity coordinates of CIE diagram are extremely useful. It is unitless parameter and its values lies between 0 % and 100 %. Color purity is significant factor used to analyze quality of red color emission and is calculated by following Eq. (23) [74,75].

$$CP = \frac{\sqrt{(x - x_i)^2 + (y - y_i)^2}}{\sqrt{(x_d - x_i)^2 + (y_d - y_i)^2}} \times 100 \quad (23)$$

In this equation, the point x , y denotes chromaticity coordinates, x_d , y_d represents coordinates of dominant wavelength and $x_i = 0.333$, $y_i = 0.333$ is the point to white illuminant. The color purity values for prepared samples were grouped together in Table 9 and value for 4 mol % sample Gd_{3-x}GaO₆:Eu³⁺ was found to be 98.15 %. The observed color purity is comparable to the reported literature (Table 10) which supports its relevancy in light emitting devices applications [76–80]. CCT is a parameter used to evaluate the warmth of white light and measure of emission color of phosphors. CCT value indicates whether the light source appearance is cool or warm. If the value of CCT is low then the color will lie in warm region and if it is high it will lie in cool region. Thus, light source having CCT > 4000 K and CCT < 3200 K are supposed to be lying in cool and warm region, respectively [81]. The CCT can be evaluated from CIE coordinates (x , y) by using McCamy method.

$$CCT = -437n^3 + 3601n^2 - 6861n + 5514.31 \quad (24)$$

$$n = \frac{(x - x_e)}{(y - y_e)} \quad (25)$$

Where, n is the inverse slope line, (x_e , y_e) indicates epicenter of convergence in CIE 1931 diagram and has values (0.332, 0.186) [82]. The CCT values of synthesized samples are tabulated in Table 9 and found to be less than 3200 K for optimized sample demonstrating

warmth of the phosphor.

The luminous efficiency of radiation (LER), which reflects the perceived brightness of light by the human eye in lm/W. Based on the emission spectrum $S(\lambda)$ the LER was determined through Eq. (26) [83].

$$LER = K_m \frac{\int_{\lambda} v(\lambda) S(\lambda) d\lambda}{\int_{\lambda} S(\lambda) d\lambda} \quad (26)$$

In this expression, $v(\lambda)$ denotes eye-sensitivity curve and K_m represents the maximum luminous efficiency achievable by a light source. The values of LER for Gd_{3-x}GaO₆:xEu³⁺ ($x = 1-7$ mol%) phosphors are listed in Table 9.

5. Conclusion

In this study, red emitting Gd_{3-x}GaO₆:xEu³⁺ ($x = 1-7$ mol%) phosphors have been developed by facile solution combustion method. The crystalline nature and orthorhombic phase (Cmc2₁) of synthesized materials were confirmed through XRD technique. The replacement of Gd³⁺ by Eu³⁺ ion in GGO host is supported by Rietveld refinement results. FE-SEM and EDX analysis was carried out for optimized sample. FE-SEM images illustrate well distributed nanoparticles with dimensions lying in nanometer range and EDX spectra validates the elemental composition. Under 261 nm excitation, Gd_{3-x}GaO₆:xEu³⁺ ($x = 1-7$ mol %) phosphors show red color emission at 612 nm owing to prominent ED (⁵D₀ → ⁷F₂) transition of Eu³⁺ ions. Critical quenching concentration of GGO:Eu³⁺ was estimated to be 4 mol% with corresponding critical energy transfer distance of 18.98 Å. The quenching mechanism was attributed to dipole-dipole interactions. The Judd-Ofelt parameters comprising intensity parameters (Ω_2 and Ω_4), radiative probabilities (A_R), branching ratio (β) and asymmetry ratios (R) were calculated for europium doped GGO phosphors. The thermal quenching profile of the optimized sample reflects its high thermal stability. Finally, photometric analysis revealed high color purity (98.15 %), low CCT values (2600–3200 K) and CIE coordinates positioned within red region of CIE diagram demonstrating their potential applications in lighting.

CRediT authorship contribution statement

Ramesh Kumar: Visualization. **Varun Kumar:** Software. **Harish Kumar:** Formal analysis. **Rinki Jangra:** Validation. **Pawan Kumar:** Project administration. **Reshu Kajal:** Writing – original draft, Methodology, Investigation, Data curation. **Devender Singh:** Writing – review & editing, Supervision, Resources.

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.

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Data availability statement

Data will be made available on request.

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