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Enhanced Reddish-Orange luminescence property of Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$ Phosphors for White-LED Applications

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ABSTRACT

This work, a series of reddish-orange emitting Sm^{3+} doped phosphors based strontium zinc tungstate $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7 and 9 mol%) host were successfully synthesised via solid state reaction route. XRD analysis confirmed synthesis of highly crystalline lattice, indicating that dopant ions were successfully integrated into tungstate host lattice matching with standard JCPDS card no.96-156-3084. Scanning electron microscopy along with Fourier transform infrared spectroscopy characterizations provide insights into the micro structural irregular polyhedron morphology and used for identifying chemical bonds respectively. The optical band gap of $Sr_4Zn_2W_2O_{12}$ phosphors at optimize concentration x=7mol% was identified to be E_g=3.71 eV through diffuse reflectance spectroscopy (DRS). Optical investigation via photoluminescence (PL) studies a prominent excitation peak at 406 nm, under this excitation the phosphor exhibits a highly pure reddish-orange radiation recorded at 645 nm showing $^4G_{5/2}\rightarrow{}^6H_{9/2}$ energy transfer. At an optimal doping level, x=7 mol% concentration quenching phenomenon identified. The optimized phosphor showed CIE chromaticity coordinates (0.596, 0.401) along with outstanding colour purity of 99.99 % and correlated color temperature (CCT) of 1625 K thus making it suitable for reddish-orange emission This optimized material exhibited exceptional color purity of 99.99%, correlated color temperature (CCT) of 1625 K and CIE chromaticity coordinates (0.596, 0.401), making it appropriate for reddish-orange emission. Temperature-Dependent Photoluminescence (TDPL) results highlighted the materials robust thermal stability with retaining 76.43 % emission intensity at 175°C, activation energy to be ΔE= 0.399 eV and quantum efficiency to be 76.9 %. The results suggesting that $Sr_4Zn_2W_2O_{12}$:xSm^{3+} is a superior host for developing high performance w-LEDs.

Keywords: Photoluminescence, phosphors, w-LEDs, activation energy

1. INTRODUCTION

White light-emitting diodes play a crucial role in advancement of solid-state lighting technology primarily due to their superior luminous efficiency and longer lifespan. Compared to traditional incandescent bulbs w-LEDs consume significantly less power which leads to reduction of energy cost. Their compact size allows for the innovation of new lighting designs and application [**Error! Reference source not found.**]. Ongoing advancements in this field will continue to replace the older lighting technologies such as including high-resolution mobile display and advanced automotive illumination [**Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.**]. Ultimately, global transition towards w-LED supports sustainable and renewable sources of energy which makes them a preferrable choice for both residential and commercial areas

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A common fabrication method for w-LEDs involves utilizing a blue-emitting LED as a primary light source in combination with a down-conversion phosphor material. A widely utilized phosphor yttrium aluminum garnet (YAG), which efficiently converts a portion of blue emission to yellow light. By applying layer of phosphor material on the blue diode to create a combined light output that appears white. However, achieving precise color rendering and desired chromaticity through this method remains a technical challenge. Experimenting with 1 different combinations can lead to improved efficiency and color quality [**Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.**].

Full-spectrum light may also be generated by recombination of three light sources: blue, green and red LEDs. When these three sources are combined with equal intensities they produce high-quality white light. However the method represents significant challenges in thermal management. Red LEDs are highly sensitive to temperature fluctuation as they heat up their luminous efficiency drops and their emission wavelength shifts. Consequently, complex sensor 18 system and feedback loops are required to maintain a consistent white light [**Error! Reference source not found.,Error! Reference source not found.**].

To achieving superior efficiency the phosphor materials must possess high quantum yield, robust thermal stability and narrow emission peaks. Various host material includes nitrides $CaAlSiN_3:Eu^{2+}$, $Sr_2Si_5N_8:Eu^{2+}$, silicate includes $(Ba,Sr)_2SiO_4:Eu^{2+}$,$Ca_2SiO_4:Eu^{2+}$,sulfides $ZnS:Cu^{+}$,Al^{3+},$SrS:Eu^{2+}$,oxynitrides $(La,Y)_3Si_6N_{11}:Ce^{3+}$,$BaSi_2O_2N_2:Eu^{2+}$, aluminate include $SrAl_2O_4:Eu^{2+}$, $BaAl_2O_4:Eu^{2+}$, which provide a stable matrix for the phosphor but silicate and aluminate based phosphors are known to suffer for their thermal quenching at elevated operating temperature which leads to a reduction in emission intensity during prolonged LED operation. Nitrides and oxynitrides phosphors provide improved red emission and higher color rendering indices however their synthesis typically requires high temperature, controlled atmosphere and complex processing routes these factors increase the production cost and reduce scalability for large scale applications. In this context tungstate-based host matrices have emerged as a promising alternative offers a high-density, thermally stable framework with a broad excitation range. Specifically the octahedral crystal lattice which act as efficient intrinsic sensitizers and also reduces quenching concentration. Among these strontium zinc tungstate($Sr_2Zn_4W_2O_{12}$) has become a favorable choice as host matrices due to their unique 2 structure and optical advantages which enhance the overall performance of RGB LEDs [**Error! Reference source not found.,Error! Reference source not found.**].

Rare earth elements are used for doping in phosphor materials because of their unique 4f electron shell, as they are shielded through outermost 5s and 5p energy levels which results 20 precise and narrow emission peaks and have high color purity [**Error! Reference source not found.,Error! Reference source not found.**]. Doping with Sm^{3+} ions offer several advantages

like efficient energy transfer, enhancing the luminescent property of phosphor. Its unique electronic structure allows for tunable emission wavelength, enabling better color rendering index. Samarium-doped phosphors demonstrate good compatibility with various host matrices, making them versatile for different application. Incorporating samarium ions leads to improved performance and efficiency in w-LEDs technology. Samarium ion absorbs high energy photons and re-emit them at lower energy levels, resulting in longer wavelength of light which is known as down-shifting [**Error! Reference source not found.**]. Sm^{3+} ion shows emission of reddish orange light because after excitation it shows radiative decay from $^4G_{5/2}$ state to lower energy levels such that its emission wavelength is around 645 nm which represents the emission of light in visible reddish orange region. The process enhances the luminescent properties of phosphor making it more effective in applications such as in led lighting and in display technology.

Tungstate-based compounds have emerged as highly promising host materials when activated with rare-earth (RE) ion [**Error! Reference source not found.**]. For instance, Ruijin reported that $Ba_2CaWO_6:Sm^{3+}$ phosphors exhibit intense orange-red emission [**Error! Reference source not found.**]. Similarly Zeng et al demonstrated that $Sr_9Eu_2W_4O_{24}$ phosphors show significant red emission, where the intensity increases proportional to the Sm^{3+}ion concentration [**Error! Reference source not found.**]. Furthermore Peng Chen developed $Ba_2ZnWO_6:Sm^{3+}$ which displays strong orange emission making them suitable candidate for orange light emitting devices [**Error! Reference source not found.**]. Despite these advancements, limited research has been directed toward the synthesis and application of Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$ phosphors for white LED applications.

The present work, Sm^{3+}-activated phosphors were synthesized via conventional high-temperature solid-state reaction route. XRD was used to analyze the phase formation and structural purity of synthesized samples and EDS was used to verify the elemental composition. The functional group of host lattice was identified and bonding properties were examined using FT-IR analysis. photoluminescence (PL) measurements including concentration and time-dependent studies, were performed to evaluate optical properties of phosphor material. Additionally, decay curve analysis was utilized to determine the luminescence lifetime of Sm^{3+} ions. No previous reports are available regarding photoluminescence behavior and morphological investigation of Sm^{3+}-doped Strontium Zinc Tungstate ($Sr_4Zn_2W_2O_{12}$) phosphors.

2. Experimental Section

2.1. *Synthesize of Sm^{3+}-doped strontium zinc tungstate phosphors*

Fig.1 shows $Sr_4Zn_2W_2O_{12}$ phosphor host lattice synthesized via solid-state reaction route. High purity precursors, specifically $SrCO_3$(99.0%), ZnO (99%), and WO_3 (99.99%) were employed as phosphors. Calculated quantities of these reagents were combined together and uniformly blended using agate mortar and pestle. Acetone added as grinding medium for homogeneous mixing and fine particle production. The obtained homogenous mixture was moved into ceramic crucible and thermally treated in heating chamber at temperature 1200°C for 7 hours. After thermal treatment, samples were allowed naturally cool to ambient temperature.

Synthesized powders were regrind for the consistency of particle size. From this approach, Sm^{3+}-activated $Sr_4Zn_2W_2O_{12}$ phosphors were effectively produced with different dopant concentrations (x= 3,5,7 and 9 mol%).

2.2. Characterization Techniques

The crystal lattice and phase homogeneity of synthesized phosphor samples were examined using XRD. A Bruker D8 advanced diffractometer based on Cu-K_α radiation (1.54 Å) filtered via Nickel applied to record the diffraction pattern. For precise phase identification, a broad range of 2(θ) data was collected. A JOEL 7610 F SEM utilized for examine sample morphology along with specific microstructural characteristics. EDS was used simultaneously to verify the final product stoichiometric ratios and elemental composition. Using a PerkinElmer Frontier FT-IR spectrometer, Mid-infrared FT-IR spectra were recorded to identify specific functional groups and examine type of bonding within host lattice. Diffuse reflectance spectra (DRS) from the JASCO V-923 model UV-Vis-NIR spectrophotometer were employed to examine photon absorption properties regarding Sm^{3+}-doped phosphor. The JASCO FP-8300 spectrofluorometer with a xenon lamp as primary excitation source was used to measure photoluminescence (PL) parameters, including excitation and emission spectra. The luminescence durations were further verified by recording time-resolved photoluminescence decay (TRPL) profiles using an Edinburgh FL920 fluorescence spectrometer. Lastly, the Ocean Optics Flame-S-XRL-ES spectrometer was used to detect temperature-dependent PL in order to evaluate the phosphors thermal stability.

3. Result and discussion

3.1. XRD analysis:

The XRD method was performed to evaluate the atomic arrangement structure and phase purity of both undoped and Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$ phosphors. Fig. 2(a) along with 2(b) illustrate XRD patterns of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} samples at different temperature and concentration (x=0 and 7mol%) alongside the standard reference from JCPDS database card no.96-156-3084 including lattice constants a = 5.6320 Å, b = 5.6080 Å, c = 9.7210 Å, α = γ= 90°, β =125.36°.

The XRD pattern of the undoped $Sr_4Zn_2W_2O_{12}$ phosphor at various temperatures are illustrates in Fig. 2(a). The Bragg diffraction peaks are very sharp and well-defined indicates high crystallinity, which is essential factor for luminescent efficiency. It was observed that Sm^{3+} doping does not significantly modified the host lattice framework, suggesting dopant ions are successfully incorporated into $Sr_4Zn_2W_2O_{12}$. The substitution is highly favourable due to comparable ionic radii of constituent ions; specifically, Sm^{3+} (0.958Å) can substitute for Zn^{2+} (0.74Å). The alignment of the observed diffraction peaks with the pure phase confirms that a 7 mol% doping concentration does not lead to any noticeable impurity or secondary phase represented in Fig. 2(b). Using obtained XRD pattern, fundamental structural parameters-including phase identification, interplanar spacing(d) and lattice constants were determined.

24 Furthermore, average crystallite size (D) was calculated using Debey-Scherrer equation [**Error! Reference source not found.**]:

$$D = \frac{K\lambda}{\beta cos\theta} \quad (1)$$

Where λ denotes X-Ray wavelength that is 1.54Å, β is FWHM, K is Scherrer constant typically taken (0.9), D represents average grain size and θ is diffraction angle.

The calculated grain size of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=0,7 mol%) are 35.39 nm and 32.47 nm respectively. Beyond Debey-Scherrer estimation, lattice strain (ε) and crystallite size (D) of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} phosphors were further evaluated using Williamson-Hall (W-H) approach. W-H relationship expressed as follows [**Error! Reference source not found.**]:

$$\beta cos\theta = \frac{k\lambda}{D} + 4\varepsilon sin\theta \quad (2)$$

In this approach, a linear plot is constructed with $\beta cos\theta$ on y-axis and $4sin\theta$ on x-axis. The intercept of resulting linear fit corresponds to $\frac{k\lambda}{D}$, enabling a more refined calculation of the average crystallite size (D), while slope of line provides value of lattice strain (ε) as shown in Fig. 2(c) [**Error! Reference source not found.**]. Fig. 2(d) represents W-H plot of $Sr_4Zn_2W_2O_{12}$:xSm^{3+}(x= 7 mol%) phosphor. The calculated crystallite size of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=0,7 mol%) by W-H approach are 80.87 nm and 90.81 nm respectively with estimated lattice strain values are 0.00125 and 0.00159. The comparatively smaller crystallite size obtained via the Debye-Scherrer equation can be attributed to its inherent limitation as it only considers size-induced peak broadening. Conversely, the Williamson-Hall method provides a more comprehensive structural assessment by decoupling the contributions of both crystallite size and microstrain to the diffraction peak width.

The detection of microstrain in phosphor materials is generally associated with internal lattice distortions, structural imperfections, or stress arising from variations in crystallographic planes. The low strain value suggests that the $Sr_4Zn_2W_2O_{12}$:Sm^{3+} system has relatively defect-free crystal frame and minimal lattice distortion. This implies that the stability of the host lattice structure is not affected by the addition of Sm^{3+} ions.

3.2. SEM: surface analysis

SEM was utilized to investigate microstructures, morphology, topography, particle dimensions moreover size allocation of Sm^{3+}-doped phosphors. The synthesized phosphors are composed of particles with morphologies ranging from irregular polyhedron presented in Fig. 3. The samples exhibits a non-uniform particle size distribution along with a moderate degree of agglomeration. This agglomeration is likely a consequence of high temperature solid state synthesis and manual grinding processes employed during preparation. The particle size for both the undoped and Sm^{3+}-activated samples were found to reside predominantly with in the micrometer range such that 4µm - 10µm range respectively. These dimensions are highly comparable to commercially available phosphor materials. Such a particle size distribution is

considered advantageous for the industrial coating process, as it facilitates the creation of a uniform phosphor layer, thereby enhancing the overall performance and reliability of the resulting LED devices [**Error! Reference source not found.**].

3.3. EDS Analysis

The elemental structure and chemical composition of host lattice were verified through EDS spectroscopy. EDS spectra for a Sm^{3+}-activated sample of $Sr_4Zn_2W_2O_{12}$ phosphor at x = 7 mol% are shown in Fig. 4. The analysis reveals the presence of different constituent elements such as strontium (Sr), zinc (Zn), Tungsten (W) and oxygen (O), including the samarium (Sm) dopant as the activated sample. The different peaks verify that all precursor reagents were successfully incorporated into the crystalline phosphor lattice. Additionally, the spectra validate absence of elemental impurities, confirming the high phase purity previously suggested by the XRD results. The results provide that high-temperature solid-state reaction method successfully produced $Sr_4Zn_2W_2O_{12}$:Sm^{3+} in perfect elemental proportion [**Error! Reference source not found.**].

3.4. Fourier Transform Infrared Spectroscopy Analysis

FT-IR spectroscopy was utilized to investigate the internal molecular structure and identify the chemical bonding characteristics within the synthesised material by analysing its infrared absorption. Fig. 5 illustrates the FT-IR spectrum of undoped $Sr_4Zn_2W_2O_{12}$ phosphor across spectral interval 400-4000 cm^{-1}. Several absorption peaks were identified by 496, 697, 806, 855, 1071,1445, 1770, 2484, 2878 and 2981 cm^{-1} corresponding specific vibrational modes of chemical bonds within lattice. The absorption peaks observed between 400 and 800 cm^{-1} are primarily attributed for metal-oxygen (M-O) bond stretching vibrations, confirming successful formation of the crystal lattice. Specifically The lowest peak is at 496 cm^{-1}, where the sharp bands located between 806 and 855 cm^{-1} are characteristic of W-O-W stretching vibrations [**Error! Reference source not found.**]. The peak near 1071cm^{-1} represents the asymmetric stretching mode of WO_6 octahedral units [**Error! Reference source not found.**,**Error! Reference source not found.**]. High-intensity peak at 1445cm^{-1} confirms the presence of Sr-O bond within the host structure [**Error! Reference source not found.**]. Asymmetric stretching mode of C-O bonds is identified as the vibrational bond at 1770 cm^{-1} [**Error! Reference source not found.**]. Minor peaks detected in 2878-2981cm^{-1} region indicate traces of organic impurities, associated to the asymmetric stretching of CO_2 molecules absorbed on the surface [**Error! Reference source not found.**].

3.5. Diffuse Reflectance Spectra and Optical Band Gap Determination

DRS was used to examine the optical properties of phosphor materials because they are opaque and made in powder form. The DRS spectra of the synthetic $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7 and 9 mol%) phosphors are displayed in Fig. 6. The spectra were recorded between 200-1600 nm. Electronic excitation from lowest energy level $^6H_{15/2}$ to several high energy states $^3H_{7/2}$, $^4F_{7/2}$, $^4I_{11/2}$, $^6F_{11/2}$, $^6F_{9/2}$, $^6F_{7/2}$, $^6F_{5/2}$, $^6H_{13/2}$ and $^6H_{11/2}$ are represented by the observed peaks. These transitions occur at respective wavelengths 324, 408, 476, 954, 1084, 1236, 1383, 1488 and 1551 nm due to absorption of incident light energy.

To determine the optical band gap from the reflectance data, reflectance (R) was converted into absorption coefficient (α) using the Kubelka-Muck (K-M) function: [**Error! Reference source not found.**]

$$F(R) = \frac{(1-R^2)}{2R} = \frac{\alpha}{S} \quad (3)$$

Where S represents the scattering coefficient. This equation allows for isolation of material's absorption characteristics. Subsequently, optical band gap (E_g) was calculated using the Tauc relations [**Error! Reference source not found.**]:

$$\alpha h\nu = A(h\nu - E_g)^n \quad (4)$$

The Tauc equation (4), E_g represents the optical band gap energy, A is constant related to material's transition probability and hν denotes the incident photon energy. The exponent n characterizes transition type, where n = 1/2 or n = 2 indicate allowed direct as well as allowed indirect transitions correspondingly. Given that Kubelka-Muck function F(R) is linearly related to absorption coefficient (α), substituted into Tauc equation as follows [**Error! Reference source not found.**].

$$F(R) = A(h\nu - E_g)^n \quad (5)$$

By plotting $[F(R)h\nu]^{1/2}$ vs (hν) along with extrapolating linear section of resulting Tauc plot to horizontal axis, values of E_g were obtained. As shown in Fig.8, calculated band gap energies of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7, 9 mol%) are 3.74, 3.72, 3.71 and 3.68 eV in order. As Sm^{3+} concentration (x) rises, band gap energy is found to decrease illustrates in Fig. 7.

3.6. Photoluminescence (PL) Analysis

Photoluminescence spectroscopy was used to analyse the optical characteristics of Sm^{3+}-activated $Sr_4Zn_2W_2O_{12}$ phosphors. For different Sm^{3+} concentrations (x=3, 5, 7 and 9 mol%) excitation spectra were recorded in 225–500 nm region. Fig. 9 depicts PL excitation spectra of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7, 9 mol%) phosphors under 645 nm emission wavelength. Ten distinct electronic transitions originating from $^6H_{5/2}$ ground state were identified. Specifically, these peaks correspond the transitions to higher energy levels $^4D_{5/2}$, $^4D_{3/2}$, $^6P_{7/2}$, $^4G_{11/2}$, $^4F_{7/2}$, $^4M_{19/2}$, $^4I_{13/2}$, $^4I_{11/2}$, $^4M_{15/2}$ and $^4I_{9/2}$. The observed excitation peaks were located at wavelengths of 347, 364, 378, 391, 406, 421, 440, 463, 480 and 491 nm respectively[**Error! Reference source not found.**,**Error! Reference source not found.**,**Error! Reference source not found.**,**Error! Reference source not found.**]. Among these, most prominent excitation peak is centred at 406 nm, which corresponds to $^6H_{5/2} \rightarrow {}^4F_{7/2}$ spectral line. The highest peak confirms that synthesised $Sr_4Zn_2W_2O_{12}$:Sm^{3+} are highly compatible with ultraviolet (UV) and near-UV radiation sources, making them suitable candidate for solid-state lighting technologies.

Emission spectra, which illustrate radiative relaxation of electrons from excited energy levels to ground state shown, are presented in Fig. 10. Four primary emission peaks were identified at wavelengths of 536, 598, 645 and 706 nm. The corresponding 4f→4f conversion of Sm^{3+} ions is represented by these peaks: $^4G_{5/2} \rightarrow {}^6H_n$ (n= 5/2, 7/2, 9/2 and 11/2) in order [**Error! Reference source not found.**]. The dominant emission peak occurs around 645nm, originating from $^4G_{5/2} \rightarrow {}^6H_{9/2}$ transition. The transition resides within visible spectrum and produces a vivid reddish-orange luminescence. These optical characteristics demonstrate that Sm^{3+} ions

effectively sensitised within the $Sr_4Zn_2W_2O_{12}$ host lattice, enabling the phosphor to serve as an efficient component for high-performance white-LED applications [**Error! Reference source not found.**].

The emission band centred at 645nm, originating from $^4G_{5/2} \rightarrow {}^6H_{9/2}$ peak, primarily governed by an electric doublet Mechanism ($\Delta J \leq 6$; J or J′= 0, 2, 4, 6). In contrast, the emission at 536 nm, corresponding to $^4G_{5/2} \rightarrow {}^6H_{5/2}$ emission, arises solely from magnetic doublet transitions (ΔJ = +1, 0, -1). The intermediate emission peak at 598nm involves $^4G_{5/2} \rightarrow {}^6H_{7/2}$ which contributes mixed multipole transition [**Error! Reference source not found.**].

In general, electric dipole transitions are parity-forbidden for 4f→4f electronic transitions, however their relative intensity compared to magnetic dipole transitions was highly sensitive to local symmetry surrounding the optically active ion [**Error! Reference source not found.**]. A dominant electric dipole transitions indicates a highly asymmetric local environment, whereas magnetic dipole transitions are largely Independent of site symmetry. In the current $Sr_4Zn_2W_2O_{12}$ lattice, the pronounced electric dipole emission associated with the $^4G_{5/2} \rightarrow {}^6H_{9/2}$ transitions at 645nm significantly surpasses the magnetic dipole transitions, confirming that Sm^{3+} ion occupy sites with low symmetry. This highly asymmetric crystal field facilitates intense reddish-orange emission with excellent colour purity.

The variation of photoluminescence emission intensity as function of Sm^{3+} ion concentration was depicted in Fig. 11. The PL intensity initially increases with rising Sm^{3+} concentration, reaching a maximum at x = 7 mol%. Beyond this threshold, a subsequent decrease in intensity is observed, a phenomenon known as concentration quenching.

The quenching effect was attributed by resonance energy transfer pathways within Sm^{3+} ions. Heavily doped levels, the inter-ionic distance decreases, which enhance the probability of non-radiative energy migration and effectively triggers quenching effects. Therefore, an optimal doping level of x=7 mol% was identified for maximizing the luminescent performance of the $Sr_4Zn_2W_2O_{12}:Sm^{3+}$ phosphor system.

To Identify the specific nature of energy transfer among Sm^{3+} ions, critical distance was calculated using Blasse's equation [**Error! Reference source not found.**]:

$$R_c \approx 2\left[\frac{3V}{4\pi X_c N}\right]^{1/3} \quad (6)$$

Here, N denotes number of available cation sites identified as 8. V is the unit cell volume determined to be 276.70 $Å^3$ and X_C is the critical Sm^{3+} concentration (x = 0.07). The computed R_C value is 9.810 Å exceeds the 5Å threshold, confirming that electric multipolar- interactions rather than exchange interactions dominate energy transfer process in these phosphors.

To further elucidates the specific multipolar interaction type, Dexter's theory was applied to evaluate the relationship between activator concentration (x) and emission Intensity (I) [**Error! Reference source not found.**]:

$$log\left(\frac{I}{x}\right) = A - \frac{slog(x)}{d} \quad (7)$$

Inside equation, d=3 for three-dimensional compounds, A is constant and the parameter s determines the interaction type. Specifically, permanent dipole-dipole interactions are represented by s=6, dipole-quadrupole by s=8 and quadrupole-quadrupole interactions by s=10 [**Error! Reference source not found.**]. Fig. 12 revels analysis of linear fit in between log(I/x) and log(x) yielded s = 5.1, which is closest to 6. This finding identifies dipole-dipole

interactions as primary mechanism responsible for concentration quenching in the $Sr_4Zn_2W_2O_{12}:Sm^{3+}$ phosphor system.

Fig. 13(a) illustrate various energy transfer pathways within Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$ systems and Fig. 13(b) represents cross-relaxation channels (CRC) and general energy transfer (ET) processes. Upon excitation, Sm^{3+} ions are promoted to higher-energy levels, followed by rapid non-radiative relaxation to metastable $^4G_{5/2}$ state. Radiative transitions then occur out of $^4G_{5/2}$ level to lower energy levels, specifically $^6H_{5/2}$, $^6H_{7/2}$ along with $^6H_{9/2}$.

For higher dopant concentrations, interactions between proximate Sm^{3+} ions promote self-quenching via non-radiative channels, which includes resonant energy transfer and cross relaxation. These mechanisms lead to a decrease in luminescence efficiency once the critical concentration is exceeded [**Error! Reference source not found.,Error! Reference source not found.**].

CRC 1: ($^4G_{5/2} + {}^6H_{5/2} \rightarrow {}^6F_{5/2} + {}^6F_{11/2}$),

In first cross-relaxation channel, excited Sm^{3+} ion undergoes non- radiative transition from the $^4G_{5/2}$ state to $^6F_{5/2}$. The energy released in this decay is simultaneously transferred to a neighbouring ground-state, promoting it from $^6H_{5/2}$ to the $^6F_{11/2}$ level.

CRC 2: ($^4G_{5/2} + {}^6H_{5/2} \rightarrow {}^6F_{7/2} + {}^6F_{9/2}$),

In second cross-relaxation channel, excited Sm^{3+} ion undergoes non- radiative transition from the $^4G_{5/2}$ state to $^6F_{7/2}$. The energy released in this decay is simultaneously transferred to a neighbouring ground-state, promoting it from $^6H_{5/2}$ to the $^6F_{9/2}$ level.

CRC 3: ($^4G_{5/2} + {}^6H_{5/2} \rightarrow {}^6F_{9/2} + {}^6F_{7/2}$),

In third cross-relaxation channel, excited Sm^{3+} ion undergoes non- radiative transition from the $^4G_{5/2}$ state to $^6F_{9/2}$. The energy released in this decay is simultaneously transferred to a neighbouring ground-state, promoting it from $^6H_{5/2}$ to the $^6F_{7/2}$ level.

CRC 4: ($^4G_{5/2} + {}^6H_{5/2} \rightarrow {}^6F_{11/2} + {}^6F_{5/2}$),

In fourth cross-relaxation channel, excited Sm^{3+} ion undergoes non- radiative transition from the $^4G_{5/2}$ state to $^6F_{11/2}$. The energy released in this decay is simultaneously transferred to a neighbouring ground-state, promoting it from $^6H_{5/2}$ to $^6F_{5/2}$ level.

3.7. Colorimetric Characteristics CIE and CCT Analysis

The chromatic performance of the synthesized $Sr_4Zn_2W_2O_{12}:xSm^{3+}$ (x=3, 5, 7, 9 mol%) phosphors was evaluated by determining Commission Internationale de l'Eclairage (CIE) coordinates from emission data recorded under 406 nm excitation. As detailed in Table 1 and illustrated in Fig. 14 emission coordinates regarding all dopant Concentrations are consistently localized within the reddish-orange spectral domain.

Additionally, the dominant wavelength approach was used to determine the color purity (CP) [**Error! Reference source not found.**]:

$$Color\ Purity = \frac{\sqrt{(x-x_i)^2+(y-y_i)^2}}{\sqrt{(x_d-x_i)^2+(y_d-y_i)^2}} \quad (8)$$

Here, (x, y) stands for chromaticity coordinates of synthesized sample, (x_i, y_i) denotes equal energy point (0.333,0.333) and (x_d, y_d) is coordinates of dominant emission wavelength. According to the analysis an exceptional colour purity of 99.99% was achieved at x=7 mol% Sm^{3+} concentration [**Error! Reference source not found.**]. In contrast to phosphor lattice that was previously reported this result signifies a huge improvement. The high color purity with excitation in the near-UV spectrum, indicates $Sr_4Zn_2W_2O_{12}$:xSm^{3+} is a perfect lattice for the applications of high-quality reddish-orange emission.
To find color appearance of light source, Correlated Color Temperature (CCT) was determined. This parameter reflects the black-body temperature that mostly matches the source's chromaticity. McCamy proposed a cubic polynomial approach described as follows [**Error! Reference source not found.**]

$$CCT = -449w^3 + 3525w^2 - 6823.20w + 5520.30 \quad (9)$$

Where w is calculated using the relationship $w = \frac{x-0.3320}{y-0.1858}$. The lower CCT values indicate warmer tones, while higher values indicates cool light [**Error! Reference source not found.**]. The Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$ phosphors exhibits CCT values under 3000 K, highlighting their potential for warm light applications.

3.8. Time–Resolved Photoluminescence Analysis

The decay kinetics of reddish-orange emission from synthesized $Sr_4Zn_2W_2O_{12}$:xSm^{3+}(x=3, 5, 7, 9 mol%) phosphors were investigated at room temperature. Analysis was conducted under excitation wavelength of 406 nm, while monitoring emission peak at 645 nm. To accurately described the experimental data, several decay models were evaluated; a bi-exponential decay function was found to provide the most precise fit shown in Fig. 15 [**Error! Reference source not found.**].

Emission decay behaviour is mathematically represented by the following relation [**Error! Reference source not found.,Error! Reference source not found.**]:

$$I = I_0 + D_1 e^{-t/\tau_1} + D_2 e^{-t/\tau_2} \quad (10)$$

Wherein I denotes luminescence intensity by specific time (t), I_0 represents initial intensity at t=0. The constants D_1 and D_2 correspond to fractional contribution of respective decay components, while τ_1 and τ_2 represents the individual decay lifetimes.
Furthermore, average lifetime (τ_{avg}) of phosphor was determined using weighted expression [**Error! Reference source not found.,Error! Reference source not found.,Error! Reference source not found.**]:

$$< \tau_{avg.} > = \frac{D_1 \tau_1^2 + D_2 \tau_2^2}{D_1\tau_1 + D_2 \tau_2} \quad (11)$$

Calculated mean lifetime (τ_{avg}) values for $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7, 9 mol%) phosphors are 0.687, 0.622, 0.621 and 0.608 ms exhibits a progressive decline Sm^{3+} concentration. This trend linked to enhanced resonance energy transfer among adjacent activator charged species. The reduction in τ_{avg} at higher doping levels confirms a higher probability of ion- ion interaction and cross-relaxation processes. Similar quenching behavior

has been documented in various rare-earth-doped oxide systems, reinforcing their suitability for solid-state lighting and optical sensing applications [**Error! Reference source not found.**]. Fig. 16 represents reduction in decay lifetime as a function of Sm^{3+} concentration, Auzel's model was employed using the following relationship [**Error! Reference source not found.**]:

$$\tau_{avg} = \frac{\tau_0}{1 + \frac{c}{c_0 e^{-\frac{N}{3}}}} \quad (12)$$

where c represents the Sm^{3+} dopant concentration, c_0 is a fitting constant, and N denotes number of photons involved in relaxation process. Here, τ_0 signifies intrinsic radiative lifetime. The application of Auzel's model to the $Sr_4Zn_2W_2O_{12}$:xSm^{3+} phosphor series yielded the radiative lifetime value(τ_0) to be 0.806 ms.

Furthermore, critical photophysical parameters, including quantum efficiency (η_{QE}) and energy-transfer efficiency (η_{ET}) were determined using stablished lifetime-based relationships [**Error! Reference source not found.**,**Error! Reference source not found.**]. The internal quantum efficiency was derived from ratio of calculated average lifespan to intrinsic radiative lifetime (τ_0)[**Error! Reference source not found.**]:

$$\text{Quantum efficiency (\%)} = \frac{\tau_{avg}}{\tau_0} \times 100 \quad (13)$$

Additionally, the energy transfer efficiency (η_{ET}), which quantifies the fraction of excitation energy exchanged via ion-ion interaction, was calculated as follows [**Error! Reference source not found.**]:

$$\eta_{ET} = 1 - \frac{\tau_{avg}}{\tau_0} \times 100 \quad (14)$$

The values of Decay lifetimes, radiative lifetime, Quantum efficiency and energy transfer efficiency η_{ET} for $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7, 9 mol%) are represented in Table 2.

3.9. Temperature-Dependent Photoluminescence (TDPL)

The presence of structural defects, often referred to as trap states, can lead to the loss of excitation energy through non-radiative pathways. To investigate the energy barriers associated with these defects and evaluate the thermal quenching behaviour of the material, Temperature-Dependent Photoluminescence (TDPL) analysis was performed. The TDPL spectra were recorded by monitoring emission intensity against wavelength, using an excitation wavelength of 406 nm over temperature range 50°C to 175°C as shown in Fig.17. $Sr_4Zn_2W_2O_{12}$:xSm^{3+}(x=7 mol%) phosphor demonstrates significant resistance to thermal degradation. Taking the emission intensity at 50° C as 100 %, a gradual decrease in luminescence is observed while temperature rises. As shown in Fig. 18, the emission intensity at highest temperature of 175°C is 76.43% belonging to its original value. This intensity shows that the synthesized phosphor series achieves maximal thermal stability at a concentration of x=7 mol%.

The Arrhenius equation was applied to temperature-dependent emission data in order to further determine activation energy (ΔE). ΔE represents thermal energy required per carrier for escape from a trap state or luminescent centre to the quenching level via non-radiative recombination.

The relationship between photoluminescence (PL) intensity and temperature is expressed as follows [**Error! Reference source not found.**].

$$I_T = \frac{I_0}{1+C\,\mathrm{ex}\left(\frac{-\triangle E}{K_B T}\right)} \quad (14)$$

In this expression, I_T denotes the PL intensity at a specific absolute temperature T, I_0 represents the initial PL intensity extrapolated to 0 K, A is a frequency factor or a constant related to the ratio of non-radiative to radiative recombination rates, k_B is the Boltzmann constant ($8.617*10^{-5}$eV/K). The value of ΔE was determined by linearising the Arrhenius equation and plotting $\ln[(I_0/I(T)) -1]$ vs $1/k_BT$, as depicted in Fig. 19. Following a linear fit of the experimental data, R^2 value 0.983 was achieved, indicating high degree of corelation. From slope of linear fit, activation energy ΔE for the $Sr_4Zn_2W_2O_{12}$:xSm^{3+}(x=7 mol%) phosphor was calculated to 0.399 eV. Table 3 illustrates CIE coordinates, color purity and Activation energy of previously reported Sm^{3+} ions doped phosphors.

4. conclusion

In this study, Sm^{3+}- activated tungstate-based phosphors were successfully synthesised through solid-state reaction route. A systematically characterization has been done to determine their structural, morphological, optical and luminescence properties were conducted. XRD analysis verified generation of single-phase crystalline lattice. The calculated grain size using Debey-Scherrer equation of $Sr_4Zn_2W_2O_{12}$:xSm^{3+}(x=0, 7 mol%) are 35.39 nm and 32.47 nm respectively. FT-IR spectroscopy verified the characteristics metal-oxygen (M-O) bonding within host framework. Electron microscopy revealed irregular, polyhedron-shaped particles with mild agglomeration and particles sizes ranging from 0.4-10 µm making them suitable for LED fabrication and phosphor coating. Tauc plot analysis revealed that the optical band gap at optimized concentration x=7 mol% was 3.71 eV. From a prominent reddish-orange radiation wavelength towards 645 nm, which corresponds to $^4G_{5/2} \rightarrow {}^6H_{9/2}$ peak, photoluminescence investigates photoexcitation peak at 406 nm, which is attributable to $^6H_{5/2} \rightarrow {}^4F_{7/2}$ transition. Dexter's dipole-dipole interaction model accurately captured the quenching concentration at x=7 mol%. The CIE coordinates of optimized concentration is (0.596, 0.401) with excellent color purity of 99.99 %. The average lifetimes of $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7, 9 mol%) phosphor are 0.687, 0.622, 0.621 and 0.608 ms described by time-resolved photoluminescence that followed a bi-exponential decay which decreased as Sm^{3+} concentration increased due to cross-relaxation processes. Additionally, TDPL investigates an activation energy of ΔE = 0.399 eV while maintaining 76.43% emission intensity at 175 °C. All of these results signify that Sm^{3+}-doped $Sr_4Zn_2W_2O_{12}$:xSm^{3+} (x=3, 5, 7 and 9 mol%) phosphor as the prominent host lattice for the applications of warm w-LEDs and display technologies.