



A novel red-emitting NaSrTiTaO₆:Eu³⁺ phosphor for application in high CRI w-LEDs and latent fingerprints

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ABSTRACT

Developing efficient, stable red phosphors is critical for advancing high-quality white light-emitting diodes (w-LEDs) and forensic technologies. The novel Eu³⁺-activated NaSrTiTaO₆ phosphors with perovskite structure were successfully prepared via the standard high-temperature solid-state reaction approach. Structural analysis confirms a cubic phase with negligible lattice distortion upon doping. The photoluminescence (PL) spectra of NaSrTiTaO₆:xEu³⁺ upon 397 nm excitation showed a dominant red emission band at 615 nm, resulting from the hypersensitive ⁵D₀→⁷F₂ transition of Eu³⁺ ions. The NaSrTiTaO₆:40 mol%Eu³⁺, among all the synthesized samples with Eu³⁺ doping levels varying between 0.5 and 50 mol%, demonstrates the best luminescent properties, accompanied by the internal quantum efficiency (IQE) of 76.8% and exceptional color purity (99.9%). Significantly, w-LEDs fabricated using this phosphor deliver superior lighting performance with a notably high color rendering index (CRI) of 90 and near-ideal white chromaticity, addressing the “red-deficiency” challenge in solid-state lighting. Furthermore, under 395 nm light, the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor modified with oleic acid (OA) could enable high-resolution visualization of latent fingerprints (LFPs), clearly displaying the three-level structure of the fingerprint and confirming its dual-functionality. With its excellent thermal stability (*E*_a = 0.31 eV) and versatile applications, these observations suggest that this phosphor can be effectively utilized in w-LEDs and the visualization of LFPs.

1. Introduction

Over recent decades, w-LEDs with phosphor conversion have been regarded as an ideal lighting resource due to their superior luminous efficiency, less energy loss, and environmental friendliness [1–3]. Currently, coating a blue-emitting semiconductor chip with Y₃Al₅O₁₂:Ce³⁺ (YAG) phosphor is the primary way for commercial w-LED manufacturing. Whereas, the performance of this type of w-LED is compromised due to the lack of red components. As a result, it suffers from a low color rendering index and high color temperature [4]. Enhancing the fluorescence efficiency of red phosphors emerges as a crucial objective in the development of w-LEDs.

Fingerprints can be used to distinguish personal information accurately in criminal investigations because of their uniqueness [5,6]. Generally, Fingerprint characteristics are systematically classified into three hierarchical levels. The first level encompasses the fundamental

patterns of fingerprints, including delta, loops, and whorls. The macroscopic details of fingerprints can be shown by level II, including dots, terminations, bridges, and spurs. Lastly, the microscopic details such as sweat pores, creases, and incipient ridges are exhibited by level III. LFP detection methods contain chemical staining methods (iodine 1, 8-diazafluoren-9-one (DFO), silver nitrate, ninhydrin, and liquid cyanoacrylate esters), equipment analysis (photoacoustic imaging, raman spectroscopy, and fourier transform infrared spectroscopy) [7]. The instruments for the equipment analysis technique are expensive and cumbersome, resulting them inaccessible at crime scenes. The chemical staining method is unavoidably hazardous and has the potential to harm the skin, eyes, and DNA of individuals using it [8]. Owing to its robust stability and high safety, the powder dusting method has gained extensive application. Compared with traditional magnetic and metal powders, phosphors offer the advantages of stability, selectivity, and resolution [9]. Thus, it becomes increasingly popular to search for novel

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phosphors to visualize fingerprints.

Due to their ability to emit red fluorescence under ultraviolet excitation, Eu^{3+} -doped inorganic materials can be utilized for the fabrication of w-LEDs and the advancement of LFP. Moreover, the intense and narrow red light can be emitted by Eu^{3+} through the ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ energy level transition, reducing energy loss. Therefore, Inorganic materials doped with Eu^{3+} have consequently garnered significant research interest in recent years. Park et al. reported that the $\text{Y}_2\text{Sn}_2\text{O}_7:\text{Eu}^{3+}$ phosphor exhibits an intense charge transfer band in the ultraviolet spectral region, owing to the charge transfer between O^{2-} ions and Eu^{3+} ions [10]. $\text{SrAlBO}_4:\text{Eu}^{3+}$ exhibits the characteristics of high color purity and efficient orange-red to bright red emission [11]. The novel $\text{Ca}_2\text{MMg}_2\text{V}_3\text{O}_{12}$ ($\text{M}=\text{Li}, \text{K}$) hosts, doped with Eu^{3+} , crystallize in cubic structures and exhibit efficient red emission under near-UV excitation. The VO_4^{3-} groups act as sensitizers, transferring energy to the Eu^{3+} ions to achieve high color purity [12,13]. The $\text{Ca}_7\text{Mg}_2\text{P}_6\text{O}_{24}:\text{Eu}^{3+}/\text{Sm}^{3+}$ possesses excellent thermal stability and chemical stability owing to its 4.95 eV wide band gap [14].

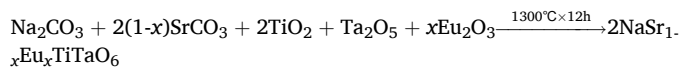
Besides, the luminescence characteristics of phosphors are also affected by the host material, so it is significant to design a suitable substrate for high-quality phosphors. Among all inorganic materials, tantalates are highly esteemed as an outstanding host matrix for phosphors due to their rich physical properties and good chemical stability. For instance, $\text{Sr}_2\text{AlTaO}_6:\text{Pr}^{3+}$ exhibits excellent thermal stability [15], while Pr^{3+} -activated $\text{Li}_2\text{SrTa}_2\text{O}_7$ demonstrates tunable luminescent properties in warm light applications [16]. Moreover, $\text{YTaO}_4:\text{Er}^{3+}$ has been utilized for high-temperature optical thermometry [17]. Recently, Eu^{3+} -doped tantalate phosphors such as $\text{Ca}_2(\text{Y}, \text{Lu}, \text{La})\text{TaO}_6:\text{Eu}^{3+}$, $\text{LiBa}_4\text{Ta}_3\text{O}_{12}:\text{Eu}^{3+}$, and $\text{Gd}_3\text{TaO}_7:\text{Eu}^{3+}$ have been applied in w-LEDs and temperature sensors [18–20]. In 2008, Rajendran et al. demonstrated that NaSrTiTaO_6 has a cubic perovskite structure and the compound or its derivatives might have potential applications as sensors, solid electrolytes, etc. [21]. However, no reports have been found for the luminescent properties of Eu^{3+} -doped NaSrTiTaO_6 up to now.

In this study, the fabrication and comprehensive analysis of novel $\text{NaSrTiTaO}_6:\text{xEu}^{3+}$ luminescent materials with different Eu^{3+} concentrations ($\text{x} = 0.5, 1, 2, 10, 15, 20, 30, 40$, and 50 mol%) are presented. The crystal structure, surface morphology, luminescent properties, thermal stability, lifetime, and quantum efficiency of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor, and its potential applicability in w-LED and fingerprint detection were investigated and evaluated. Additionally, due to the ideal optical properties, $\text{NaSrTiTaO}_6:\text{Eu}^{3+}@\text{OA}$ was proven to possess significant potential for application in the development of LFP.

2. Experiment procedures

2.1. Synthetic process

The synthesis of $\text{NaSrTiTaO}_6:\text{xEu}^{3+}$ (0.5–50 mol%) phosphors was prepared by high-temperature solid-state interaction. Na_2CO_3 (A. R., 99.99%), SrCO_3 (A. R., 99.99%), TiO_2 (A. R., 99.99%), Ta_2O_5 (A. R., 99.99%), and Eu_2O_3 (A. R., 99.99%) were utilized as raw materials for the synthesis of the samples. The starting reagents were accurately weighed according to stoichiometry and subsequently transferred to an Al_2O_3 crucible for thorough grinding. Ultimately, after sintering at 1300 °C for 12 h in a muffle furnace, the synthesized product underwent natural cooling to ambient temperature, followed by secondary grinding using an agate mortar to ensure uniform particle size distribution for subsequent characterization. The synthesized samples exhibit a white coloration with a fine-grained texture, demonstrating intense orange-red luminescence when excited by both 254 nm and 365 nm ultraviolet radiation. The following are the reaction conditions and equations of the experimental reaction:



2.2. Characterization and testing instruments

To acquire the X-ray diffraction (XRD) patterns, the $\text{NaSrTiTaO}_6:\text{xEu}^{3+}$ ($\text{x} = 0.5\text{--}50 \text{ mol}\%$) samples were scanned within the range extended from $10^\circ \leq 2\theta \leq 70^\circ$, using an XRD instrument (D2 PHASER, Bruker, Germany) with $\text{Cu K}\alpha$ ($\lambda = 0.15405 \text{ nm}$) radiation. Simultaneously, the surface morphology, energy dispersive spectrometer (EDS), and elemental composition were examined by the scanning electron microscope (SEM, Nova Nano SEM-450). To explore the potential of the phosphors in LFP visualization, the particle size distribution was analyzed by the laser particle size analyzer (Malvern MS2000, Britain). PL spectra and photoluminescence excitation (PLE) spectra were determined via the fluorescence spectrometer (Edinburgh Instruments, FLS 980).

2.3. Fabrication of the LEDs

A red-emitting LED device was fabricated by coating a mixture of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor and an appropriate UV curable adhesive onto a 395 nm near-ultraviolet (n-UV) InGaN substrate. The w-LED was fabricated based on the tri-color RGB method. One w-LED was successfully assembled by combining the commercial phosphor $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$ (BAM, blue), commercial $(\text{Ba}, \text{Sr})_2\text{SiO}_4:\text{Eu}^{2+}$ (green), and prepared $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ red phosphor on an n-UV InGaN chip. The samples were evenly mixed into a photosensitive silicone adhesive, applied onto the n-UV chip, and then cured under a UV light for 5 min. After that, the red LED and w-LED were effectively assembled, respectively. Finally, the electroluminescence (EL) spectra and PL of the LEDs, including CRI, correlated color temperature (CCT), and CIE chromaticity coordinates, and so on, were measured using an Ocean Optics fiber optic spectrometer (model USB 4000).

2.4. Synthesis of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+} @ \text{OA}$ phosphor

The hydrophobic OA surface-modified $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ luminescent powder was synthesized via the following procedure. A homogeneous solution was first prepared by dissolving 5 mL of OA in 25 mL of absolute anhydrous ethanol. Subsequently, 1 g of the $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor was introduced into this mixture. The phosphor was suspended in the solution by ultrasonication for a duration of 1 h. Subsequently, the mixture was transferred into a hydrothermal reaction vessel and subjected to heating at 150 °C for 6 h. Afterward, the sample was then permitted to cool to ambient temperature naturally. In order to eliminate the surplus OA, the precipitate was gathered, centrifuged at a speed of 7000 revolutions per minute for 10 min, and subsequently subjected to ultrasonic cleaning with anhydrous absolute ethanol. After the mixture was centrifuged again and dried at 60 °C for 5 h, the hydrophobic $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+} @ \text{OA}$ phosphor was obtained.

2.5. Acquisition of fingerprint image

To obtain fingerprints, the donor's hands were washed first. Next, the fingerprint was gently pressed onto the aluminum foil. Subsequently, the fluorescent powder was applied to the fingerprint with a fingerprint brush. After gently removing the excess phosphors from the surface of the fingerprints using an ear-washing ball, a digital camera (Canon EOS 700D) was utilized to capture images of the phosphor-visualized fingerprints under both daylight and UV illumination.

3. Results and discussion

3.1. Microstructure and morphology analysis

The atomic arrangement and local coordination spheres in NaSrTiTaO₆ are visualized in Fig. 1, with green spheres representing sodium or strontium atoms, dark blue spheres representing titanium or tantalum atoms, and red spheres representing oxygen atoms, respectively. The NaSrTiTaO₆ exhibits a cubic perovskite structure and belongs to a space group of *Pm3m* [21], with cell parameters $a = b = c = 3.912 \text{ \AA}$, $V = 478.8 \text{ \AA}^3$, and $Z = 2$. The [Na/Sr]O₁₂ group is formed by Na⁺ and Sr²⁺ ions jointly occupying the 1b sites, each coordinating with twelve oxygen atoms. As depicted in Fig. 1(b), all bond lengths connecting the 1b sites and the oxygen atoms are $2.766 \text{ \AA}$.

The tolerance factor (T_f) can be used to assess the stability of the perovskite crystal structure. Its calculation was performed in accordance with the approach described by C.J. Bartel et al. in 2019 by the following equation [22]:

$$T_f = \frac{R_O}{R_B} - n_A \left(n_A - \frac{R_A/R_B}{\ln(R_A/R_B)} \right) \quad (1)$$

where n_A represents the arithmetic mean of the oxidation state of Na⁺ and Sr²⁺, $R_B = (R_{Ti} + R_{Ta})/2$, $R_A = (R_{Sr} + R_{Na})/2$, R_{Na} , R_{Sr} , R_{Ti} , R_{Ta} , and R_O are the ionic radii of Sr²⁺, Na⁺, Ti⁴⁺, Ta⁵⁺, and O²⁻, respectively. A T_f value below 4.18 is indicative of a perovskite structure. Thus, the calculated T_f of 4.08 represents that the NaSrTiTaO₆ is a stable perovskite.

Meanwhile, the relative radius deviation expressed as a percentage of the radius (D_r) in the substance can be calculated according to the presented equation [23]:

$$D_r = \frac{R_s(\text{CN}) - R_d(\text{CN})}{R_s(\text{CN})} \quad (2)$$

This model employs the parameters D_r , R_m , R_d , and CN, which correspond to the relative radius deviation (expressed as a percentage), the radius of the substituted ion, the radius of the Eu³⁺ dopant ion, and the coordination number, respectively. The corresponding determined parameters are listed in Table 2. When CN = 12, the ionic radius of Sr²⁺ and Na⁺ is $1.44 \text{ \AA}$ and $1.39 \text{ \AA}$, respectively, while the ionic radius of the Eu³⁺ ion is $1.12 \text{ \AA}$. The D_r value of Sr²⁺ (22.2%) and Na⁺ (19.42%) are both less than 30% [24]. However, Eu³⁺ substitution at the Na⁺ site introduces a +2 effective charge, necessitating high-energy defects for compensation and reducing stability [25,26]. In contrast, Eu³⁺ substitution for Sr²⁺ yields only a +1 effective charge, while charge compensation is realized by Na⁺ substitution at neighboring Sr²⁺ sites [27]. Therefore, although the ionic radius of Eu³⁺ is closer to that of Na⁺, Eu³⁺ will preferentially substitute for Sr²⁺.

The XRD patterns of NaSrTiTaO₆: $x\text{Eu}^{3+}$ ($x = 0.5\text{--}50 \text{ mol\%}$) are revealed in Fig. 2(a). Evaluation of the samples reveals a perfect correspondence between the measured peaks and the NaSrTiTaO₆ reference data (PDF#04-016-6687), signifying that the synthesized phosphors are sole-phase materials. As visualized in Fig. 2, the refinement output contrasts measured Bragg reflections (black markers) against theoretical predictions (red trace), with misfits emphasized by the blue residual plot. In addition, the short green vertical stripes signify the Bragg positions. The refinement data ($R_{wp} = 11.90\%$, $R_p = 6.97\%$, and $\chi^2 = 3.35$) suggest that the Sr²⁺ ions are effectively replaced by Eu³⁺ ions. Furthermore, the refined lattice parameters of NaSrTiTaO₆:Eu³⁺ phosphor are computed and displayed in Table 1.

For optimal implementation in w-LEDs and LFP detection, the surface morphology and particle size distribution of phosphor represent critical quality parameters. The distribution of particle size is depicted in Fig. 3(a), revealing that the particle-size distribution of NaSrTiTaO₆:40 mol%Eu³⁺ phosphor is mainly distributed at $1.56 \text{ \mu m}$. Thereinto, $d(0.1)$ is $0.13 \text{ \mu m}$, indicating a proportion below 10% for particles measuring under 0.13 micrometers . $d(0.5)$ is $0.18 \text{ \mu m}$, which is the average sample size, illustrating that a median diameter ($d(0.5)$) of $0.18 \text{ \mu m}$, while the $d(0.9)$ value of $2.31 \text{ \mu m}$ confirms that 90% of particles fall below $2.31 \text{ \mu m}$ in size. The dimensions of the sweat pores within the fingerprint are between 88 and $220 \text{ \mu m}$ [28], indicating that these samples can be used for the development of LFP. SEM images of NaSrTiTaO₆:40 mol%Eu³⁺ phosphor at various magnifications are presented in Fig. 3(b) and (c), which distinctly reveal the surface morphology of the phosphor particles. It can be seen that there are still a few agglomerates of phosphor particles, which are caused by the Ostwald ripening and directional adhesion [29].

The elemental composition of the synthesized samples is analyzed using EDS, with the findings presented in Fig. 4(a). The detected peaks in the spectrum correspond to the elements composing the phosphor, and the EDS analysis successfully verifies the presence of all the essential elements, including Na, Sr, Ti, Ta, O, and Eu. Meanwhile, the EDS elemental mapping images for the various components within the phosphor are depicted in Fig. 4(b–g), where orange, light purple, blue, yellow, green, and deep purple correspond to Na, Sr, Ti, Ta, O, and Eu, respectively. These figures show that the different elements are fairly uniformly dispersed throughout the phosphor. Additionally, these results verify the successful incorporation of Eu³⁺ ions into the NaSrTiTaO₆ lattice and the effective preparation of the NaSrTiTaO₆:40 mol% Eu³⁺ phosphor.

3.2. Photoluminescence properties

The excitation characteristics of NaSrTiTaO₆:40 mol%Eu³⁺ phosphor, measured at 615 nm emission, are illustrated in Fig. 5. The charge

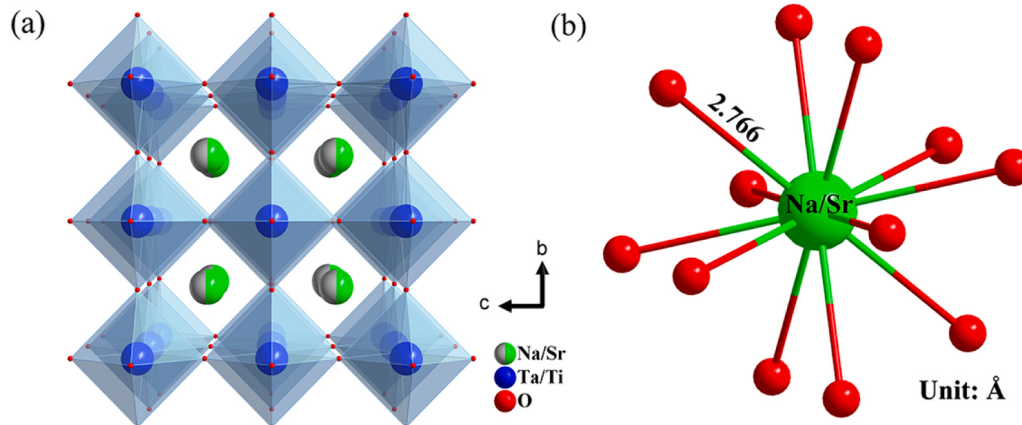


Fig. 1. (a) The crystal structure of perovskite NaSrTiTaO₆, (b) The coordination environment of sodium or strontium ions (Bond length unit: Å).

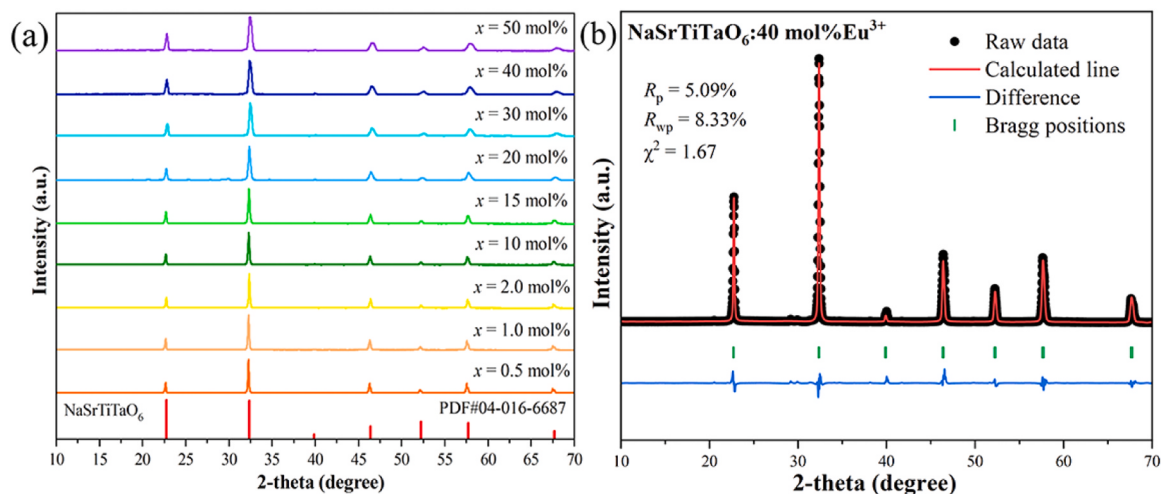


Fig. 2. (a) The XRD patterns of the $\text{NaSrTiTaO}_6\text{:}x\text{Eu}^{3+}$ ($x = 0.5\text{--}50\text{ mol\%}$) phosphors, (b) Rietveld refinement pattern of the $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$.

Table 1

The crystallographic parameters of the $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$ obtained by rietveld refinement.

Atom	Wyck.	Occup.	x	y	z	Uiso
Na	1b	0.5000	0.5000	0.5000	0.5000	0.012
Sr	1b	0.5000	0.5000	0.5000	0.5000	0.012
Ti	1a	0.5000	0.0000	0.5000	0.0000	0.012
Ta	1a	0.5000	0.0000	0.5000	0.0000	0.012
O	3d	1.0000	0.5000	0.5000	0.0000	0.012

Table 2

D_r of doped ions versus potential substitution sites within the NaSrTiTaO_6 lattice.

Ion	CN	Ion radius ($\text{\AA}$)	D_r (with Eu^{3+})
Ta^{5+}	6	0.64	48.0%
Ti^{4+}	6	0.605	56.5%
Sr^{2+}	12	1.44	22.2%
Na^+	12	1.39	19.42%

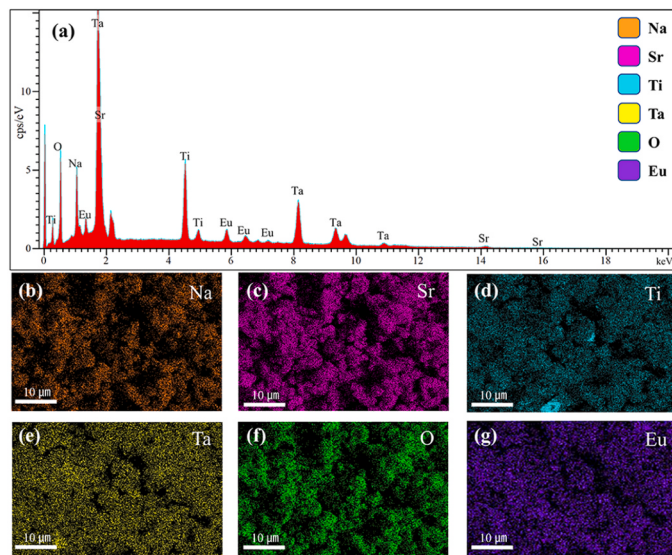


Fig. 4. (a) EDS spectrum, (b–g) Elemental mapping of the $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$ phosphor.

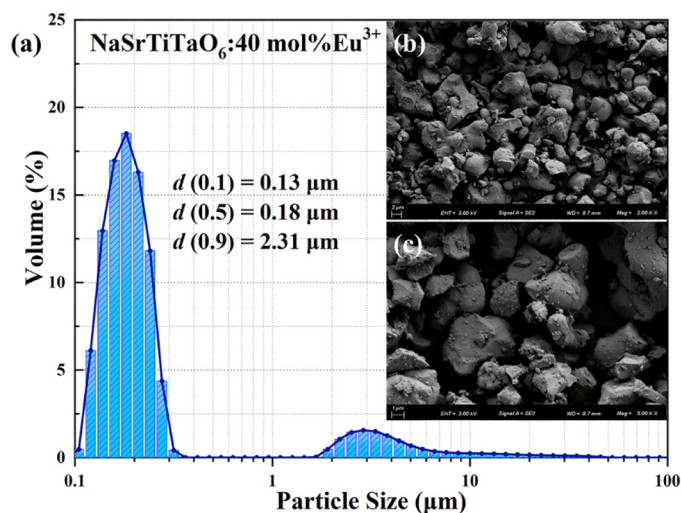


Fig. 3. (a) Particle size distribution of the $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$, (b, c) the SEM image of $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$.

transfer band (CTB), ranging from 250 to 350 nm, originates from the

electronic transition involving charge transfer from the negatively charged oxygen ion ($2p^6$) to the unoccupied state of the $4f^7$ orbital of the Eu^{3+} ion, as well as the charge transfer from O^{2-} to Ta^{6+} . All of the intense excitation bands among 350 and 500 nm are on account of the $4f\text{--}4f$ transition generated by the ${}^7\text{F}_0\text{--}{}^5\text{D}_4$ (363 nm), ${}^7\text{F}_0\text{--}{}^5\text{G}_3$ (376 nm), ${}^7\text{F}_0\text{--}{}^5\text{L}_6$ (397 nm), ${}^7\text{F}_0\text{--}{}^5\text{D}_3$ (413 nm), and ${}^7\text{F}_0\text{--}{}^5\text{D}_2$ (465 nm), respectively [30]. Among the peaks, the ${}^7\text{F}_0\text{--}{}^5\text{L}_6$ transition of Eu^{3+} ions leads to the sharpest absorption peak, which is located at around 397 nm, as shown in Fig. 5(a). The two stronger emission peaks centered at 397 and 465 nm mean that the $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$ phosphor exhibits strong excitation under both UV and blue light, demonstrating its suitability for integration into w-LED devices and anti-counterfeiting ink [29,31].

The luminescence characteristics of $\text{NaSrTiTaO}_6\text{:}40\text{ mol\%Eu}^{3+}$ under 397 nm excitation are presented in Fig. 5(b), revealing distinct emission bands. The phosphor material demonstrates pronounced red emission, predominantly originating from the electric dipole ${}^5\text{D}_0\text{--}{}^7\text{F}_2$ transition of Eu^{3+} ions. Meanwhile, the attained PL spectrum features other weak peaks caused by ${}^5\text{D}_0\text{--}{}^7\text{F}_0$, ${}^5\text{D}_0\text{--}{}^7\text{F}_1$, ${}^5\text{D}_0\text{--}{}^7\text{F}_3$, and ${}^5\text{D}_0\text{--}{}^7\text{F}_4$ transitions, which correspond to 578, 593, 651, and 703 nm, respectively [32,33]. In the red region, the emission peak has the strongest

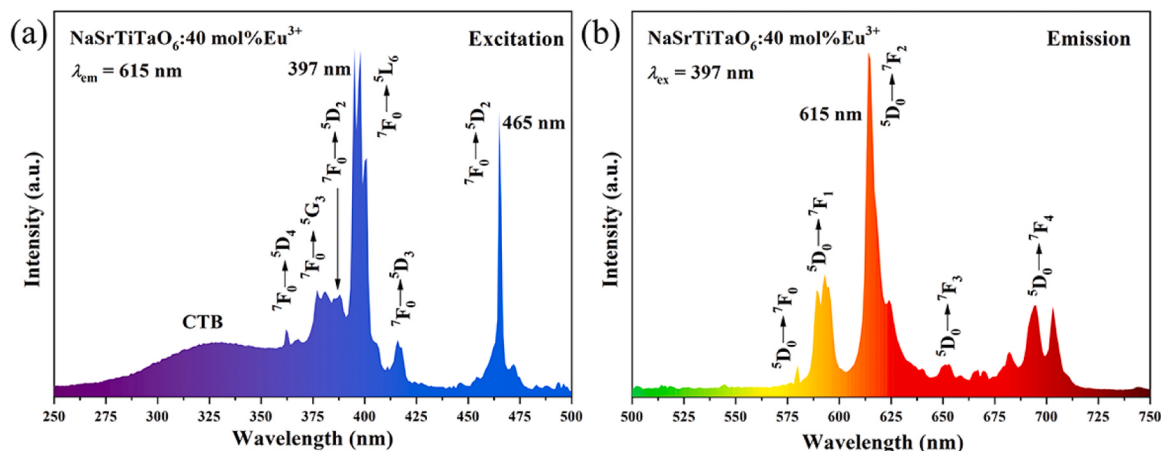


Fig. 5. (a) The excitation spectrum of the NaSrTiTaO₆:40 mol%Eu³⁺ ($\lambda_{em} = 615$ nm), (b) The emission spectrum of the NaSrTiTaO₆:40 mol%Eu³⁺ ($\lambda_{ex} = 397$ nm).

intensity at 615 nm ($^5D_0 \rightarrow ^7F_2$), suggesting that the product has the potential to be applied in red light-emitting devices.

Fig. 6 displays an energy level diagram derived from excitation and emission spectra. Electrons absorb photon energy at specific points (363, 376, 397, 413, and 465 nm) and are excited to high energy levels. Following excitation, electrons undergo non-radiative decay to the 5D_0 state, subsequently emitting radiation when transitioning to various 7F_J levels ($J = 0-4$).

The luminescence intensity of NaSrTiTaO₆:xEu³⁺ ($x = 0.5-50$ mol %), varying with the doping concentration of Eu³⁺ ions, is illustrated in Fig. 7(a). Evidently, comparative analysis reveals nearly identical luminescence profiles for all composition variants in the NaSrTiTaO₆:xEu³⁺ system. The observed spectral data clearly demonstrate that Eu³⁺ doping levels exert negligible influence on the characteristic emission peak locations in the photoluminescence spectra. Fig. 7(b) exhibits the normalized integral of luminescence intensity of NaSrTiTaO₆:xEu³⁺ ($x = 0.5-50$ mol%) phosphors. As depicted in Fig. 7(b), the luminescence intensity of the phosphor rises accordingly as the Eu³⁺ ion concentration increases at first. After Eu³⁺ ion concentration exceeds 40 mol%, the luminescence intensity of the NaSrTiTaO₆:Eu³⁺ phosphors decreases with increasing concentration of Eu³⁺ ions. This concentration quenching phenomenon originates from the presence of NR transitions. The NR transition is subsequently enhanced when the ion concentration of Eu³⁺ increases, resulting in a decline of the luminescence intensity of the phosphor [34].

To elucidate the concentration-dependent effects of Eu³⁺ ions on phosphor properties, the following equation is able to be employed to determine the critical transfer distance (R_c) [35]

$$R_c \approx 2 \left(\frac{3V}{4\pi x_c N} \right)^{\frac{1}{3}} \quad (3)$$

In this equation, x_c represents the optimal content in NaSrTiTaO₆:xEu³⁺ ($x = 0.5-50$ mol%), which is 40 mol%. N denotes the number of equation units per unit cell, and for NaSrTiTaO₆ phosphor, $N = 2$. V serves as the volume per crystal cell, and in this case, for NaSrTiTaO₆ phosphor, $V = 478.8 \text{ \AA}^3$. The calculation shows that the R_c of the Eu³⁺ ion in NaSrTiTaO₆ is approximately $10.46 \text{ \AA}$, which surpasses $5 \text{ \AA}$, indicating that the quenching of the luminous material concentration is due to the electric multipolar interaction between the ions [36].

A comprehensive understanding of high-concentration Eu³⁺ interactions requires the theoretical calculation of the cascaded energy transfer mechanism as formulated in the following relation [37]:

$$\frac{I}{x} = K [1 + \beta(x)^{Q/3}]^{-1} \quad (4)$$

In this equation, K and β are the constants of each interaction under identical reference conditions, while I denote the luminescence intensity of NaSrTiTaO₆:Eu³⁺ phosphor, x denotes the doping concentration of Eu³⁺ in NaSrTiTaO₆, and the multistage interaction mechanism type is Q , $Q = 3, 6, 8$, and 10 indicating the nearest ion, dipole-dipole, dipole-quadrupole, and quadrupole-quadrupole, respectively. As can be seen in Fig. 8(a), a linear relationship exists between the $\lg I/x$ and $\lg x$ of the NaSrTiTaO₆:xEu³⁺ phosphors. The slope of the line is -0.73 , so Q is calculated to be 2.19 , which is nearly 3 . Therefore, the nearest-neighbor ion interaction accounts for the concentration quenching in NaSrTiTaO₆:Eu³⁺ phosphors.

Generally, the ED transition is highly responsive to the surrounding chemical conditions of the dopants and the symmetry of the crystal field, where the dopants are exposed, and the symmetry of the crystal field, whereas the MD transition is not [38]. Substituting Sr²⁺ with Eu³⁺ causes lattice contraction and local distortion due to ionic radius mismatch, breaking the inversion symmetry [39]. According to the Judd-Ofelt theory, this distortion enhances the probability of the electric dipole transition $^7F_0 \rightarrow ^5D_2$ [40]. The increase of the Eu³⁺ doping level progressively amplifies the degree of crystal field distortion, continuously reduces the site symmetry, and consequently improves the intensity of the hypersensitive transition [41]. Therefore, Eu³⁺ ions can be used as effective probes to assess the symmetry of lattice positions. According to Judd-Ofelt theory, the symmetry of the Eu³⁺ ion in its chemical conditions can be analyzed by the asymmetry ratio (R/O) [42], and the asymmetry can be obtained by integral I_{ED}/I_{MD} .

Fig. 8(b) exhibits the corresponding asymmetric ratios of the NaSrTiTaO₆:Eu³⁺ phosphors at different concentrations. The asymmetric ratio values are $2.49, 2.51, 2.53, 2.62, 2.63, 2.66$, and 2.70 ,

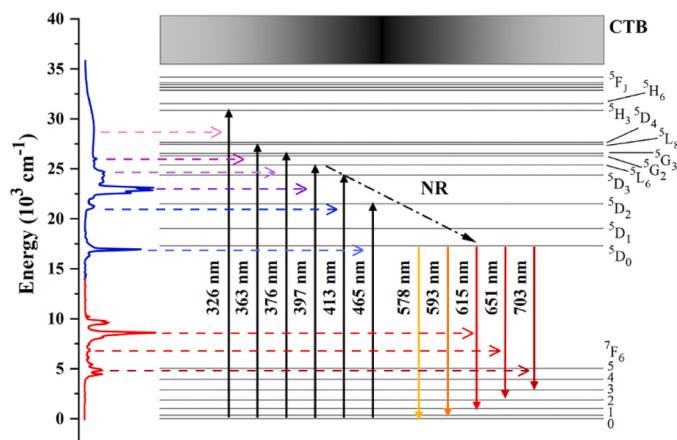


Fig. 6. The schematic energy level diagram for Eu³⁺ ions.

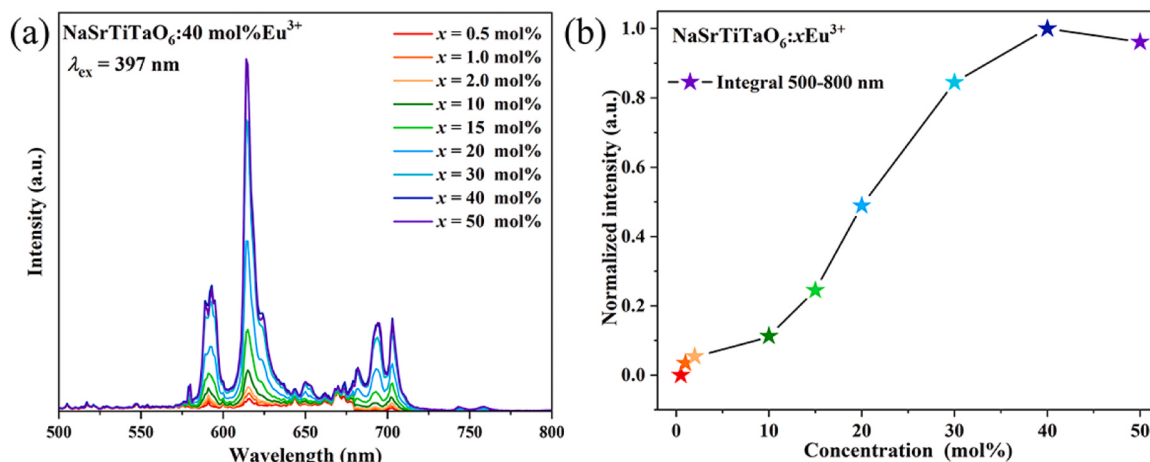


Fig. 7. (a) The PL spectra of the NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50$ mol%) at 397 nm, (b) The effect of Eu³⁺ concentration on normalized emission intensity.

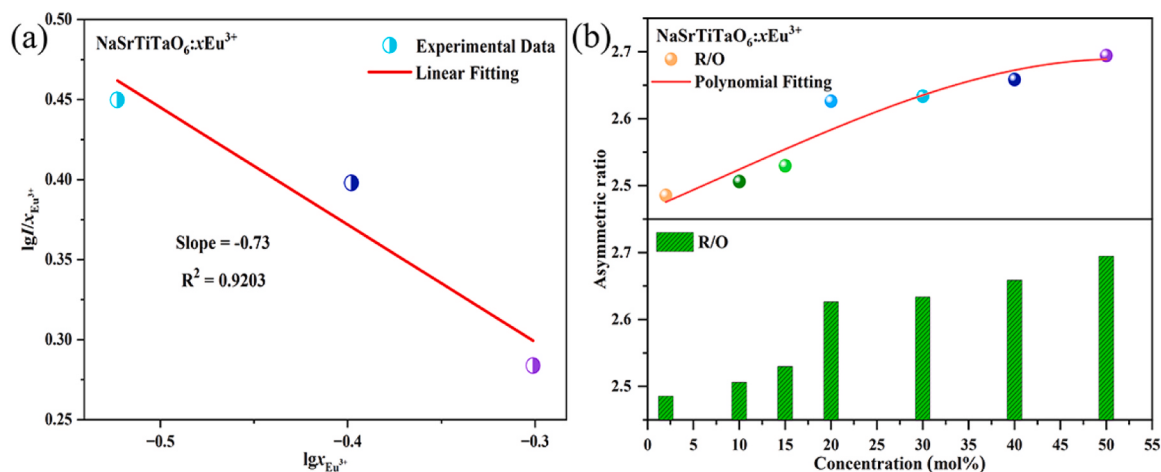


Fig. 8. (a) The relationship between $\lg(I/x)$ and $\lg x$ in the NaSrTiTaO₆:Eu³⁺, (b) R/O changes with the concentration of Eu³⁺.

corresponding to the concentration of Eu³⁺, 2, 10, 15, 20, 30, 40, and 50 mol%, respectively. The corresponding asymmetry ratio increases with the increase of Eu³⁺ concentration, indicating that the local symmetry around the Eu³⁺ ions is gradually being disrupted. $R/O > 1$

indicates that the ED transition is predominant and Eu³⁺ ions are positioned in sites lacking inversion symmetry within the NaSrTiTaO₆ host lattice [43,44], which indicates the advantages of high red color purity.

The luminescent performance of phosphors can be effectively

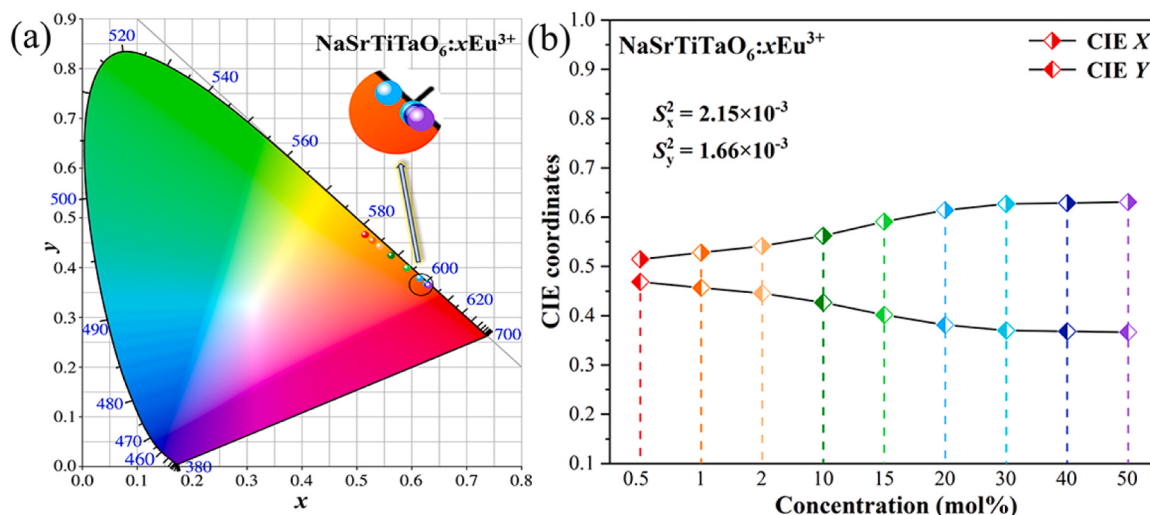


Fig. 9. (a) The CIE chromaticity coordinates of NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50$ mol%), (b) The variation between CIE coordinates and the concentration of Eu³⁺.

assessed through their CIE chromaticity coordinates, which serve as crucial indicators in this evaluation. The emission spectra provide the means to determine the CIE chromaticity coordinates for the NaSrTiTaO₆:Eu³⁺ phosphor samples, which are visually represented by the dots in Fig. 9(a). The chromaticity coordinates of NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50\text{ mol}\%$) phosphors are (0.515, 0.469), (0.528, 0.457), (0.541, 0.446), (0.562, 0.427), (0.591, 0.402), (0.614, 0.381), (0.627, 0.370), (0.629, 0.368), and (0.631, 0.367), respectively. All of these chromaticity coordinates points are situated within the red region and lie at the periphery of the CIE 1931 color chromaticity diagram. This positioning indicates that the NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50\text{ mol}\%$) phosphors exhibit high color purity. The following equation is used to calculate the color purity [45]:

$$\text{Color purity} = \frac{\sqrt{(x_s - x_i)^2 + (y_s - y_i)^2}}{\sqrt{(x_d - x_i)^2 + (y_d - y_i)^2}} \times 100\% \quad (5)$$

Here, the coordinates (x_i, y_i) represent the white light at (0.333, 0.333), while (x, y) denotes the chromaticity coordinates of the products. Additionally, (x_d, y_d) corresponds to the chromaticity coordinates of the dominant wavelength points. Through calculations, the NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50\text{ mol}\%$) samples demonstrate the excellent color purity ranging from 99.7% to 99.9%.

Fig. 9(b) illustrates how the CIE coordinates shift as the concentration of Eu³⁺ ions varies. With concentration ascending, the resulting chromaticity values (x, y) exhibit only a minimal variation. With the following equation, we can get the variance (S^2) of the coordinates:

$$S^2 = \frac{\sum (\bar{\alpha} - \alpha_i)^2}{n - 1} \quad (6)$$

where α_i is the coordinate value, and the $\bar{\alpha}$ is the average of α_i . The calculated variance values ($S_x^2 = 2.15 \times 10^{-3}$, $S_y^2 = 1.66 \times 10^{-3}$) indicate the deviation between the data values and the average. In Fig. 9(b), the CIE x values slightly rise as the ascend of concentration, whereas the CIE y values display an inverse trend. Table 3 provides a comprehensive summary of the computed color purity, CIE chromaticity coordinates, and CCT of the NaSrTiTaO₆:xEu³⁺ phosphors, with x ranging from 0.5 to 50 mol%.

The lifetime decay of the samples has important implications for the study of the phosphors. Fig. 10 illustrates the luminescence decay curves of the NaSrTiTaO₆:xEu³⁺ ($0.5\text{ mol}\% \leq x \leq 50\text{ mol}\%$) samples monitored at 615 nm with excitation at 397 nm. With increasing Eu³⁺ ions, the average decay time for the sample is expected to be determined from the following equation [46]:

$$\tau_m = \frac{\int_0^\infty t \times I(t) dt}{\int_0^\infty I(t) dt} \quad (7)$$

In this equation, $I(t)$ denote the luminescence intensity of NaSrTiTaO₆:xEu³⁺ ($0.5\text{ mol}\% \leq x \leq 50\text{ mol}\%$), and τ_m represents the average

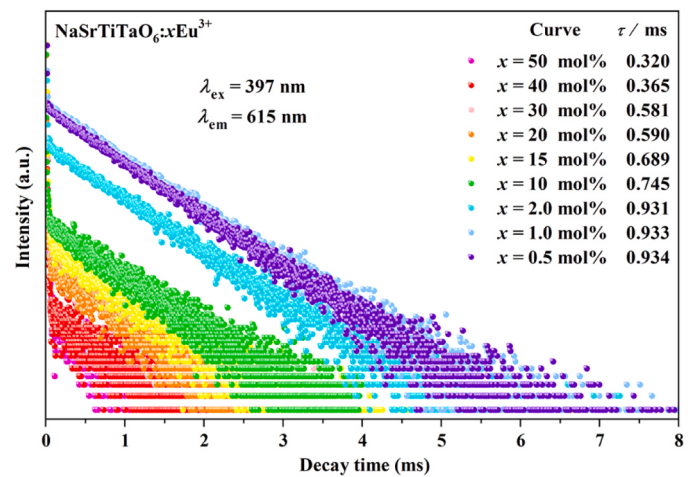


Fig. 10. The luminescence lifetime curves of NaSrTiTaO₆:xEu³⁺ ($x = 0.5\text{--}50\text{ mol}\%$).

decay time. At different doped concentrations of 0.5, 1.0, 2.0, 10, 15, 20, 30, 40, and 50 mol%, the decay lifetime of the samples is 0.934, 0.933, 0.931, 0.745, 0.689, 0.590, 0.581, 0.365, and 0.320 ms, indicating that the decay lifetime exhibits a downward trend as the doping concentration rises.

Generally, the luminescent intensity of phosphors is significantly influenced by temperature. Fig. 11 (a) displays the PL spectra of the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor measured at various temperatures, using an excitation wavelength of 397 nm. At different temperatures, the emission peaks maintain a uniform shape. As the temperature rises from 300 K to 480 K, the PL intensity of the phosphor diminishes, likely attributed to the enhanced NR transition. Fig. 11 (b) illustrates the PL intensity for the NaSrTiTaO₆:40 mol%Eu³⁺ product with temperature. The $T_{0.5}$ represents the temperature at which the normalized emission intensity decreases to half of its initial value at a temperature of 300 K. The normal operating temperature of the LED device is 420 K. In this experiment, $T_{0.5}$ is above 420 K, which indicates its potential application for w-LEDs.

Fig. 12 (a) intuitively illustrates the correlation among the wavelength, intensity, and temperature for the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor. The color shift from red to blue reflects the decreasing emission intensity of the samples. One of the most crucial parameters for LED phosphors is the activation energy (E_a) of thermal quenching with temperature. The correlation among $\ln[(I_0/I)-1]$ and $1/T$ for NaSrTiTaO₆:40 mol%Eu³⁺ phosphor is depicted in Fig. 12 (b). The E_a of the phosphor during the thermal quenching process is calculated by the Arrhenius equation [47,48]:

$$I(T) = \frac{I_0}{1 + c \exp\left(\frac{-E_a}{kT}\right)} \quad (8)$$

Here, $I(T)$ denote the luminescence intensity of the phosphors measured at various temperatures (T), I_0 represents the initial intensity, and c and k are constants. As displayed in Fig. 12 (b), every original data point aligns closely with the fitted line ($R^2 = 0.9972$). The activation energy E_a is derived to 0.31 eV through the linear fitted curve for the NaSrTiTaO₆:40 mol%Eu³⁺ product. Compared to several reported phosphors, such as BaZnO₂:Eu³⁺ (0.26 eV) [49], NaGd_{0.95}MgWO₆:0.05Eu³⁺ (0.2279 eV) [50], and BaLiZn₃(PO₄)₃:0.05Eu³⁺ [51], the obtained E_a of NaSrTiTaO₆:40 mol%Eu³⁺ is higher value, which suggests that the product has outstanding thermal stability.

The IQE of the phosphor is one of the most important complications in evaluating its applicability in w-LEDs. Fig. 13 depicts the comparison between the emission spectrum of the NaSrTiTaO₆:40 mol%Eu³⁺

Table 3

CIE (x, y), CCT, and color purity of NaSrTiTaO₆:Eu³⁺ phosphors at different concentrations.

Concentration (mol%)	CIE (x, y)	CCT (K)	Color purity (%)
0.5	(0.515, 0.469)	2461	99.7
1.0	(0.528, 0.457)	2260	99.7
2.0	(0.541, 0.446)	2080	99.8
10	(0.562, 0.427)	1829	99.8
15	(0.591, 0.402)	1633	99.8
20	(0.614, 0.381)	1667	99.9
30	(0.627, 0.370)	1792	99.9
40	(0.629, 0.368)	1822	99.9
50	(0.631, 0.367)	1844	99.9

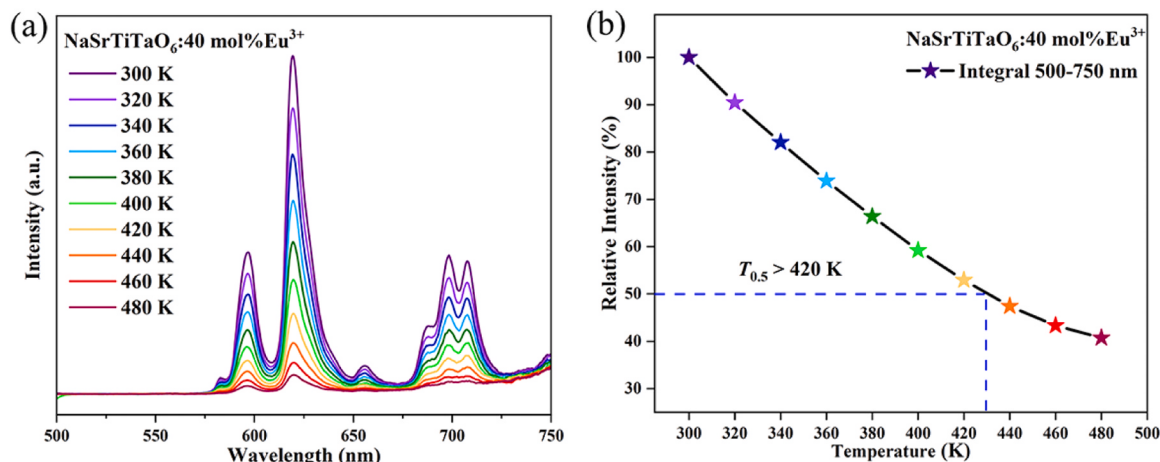


Fig. 11. (a) NaSrTiTaO₆:40 mol%Eu³⁺ emission spectra at different temperatures; (b) PL intensity with the temperature.

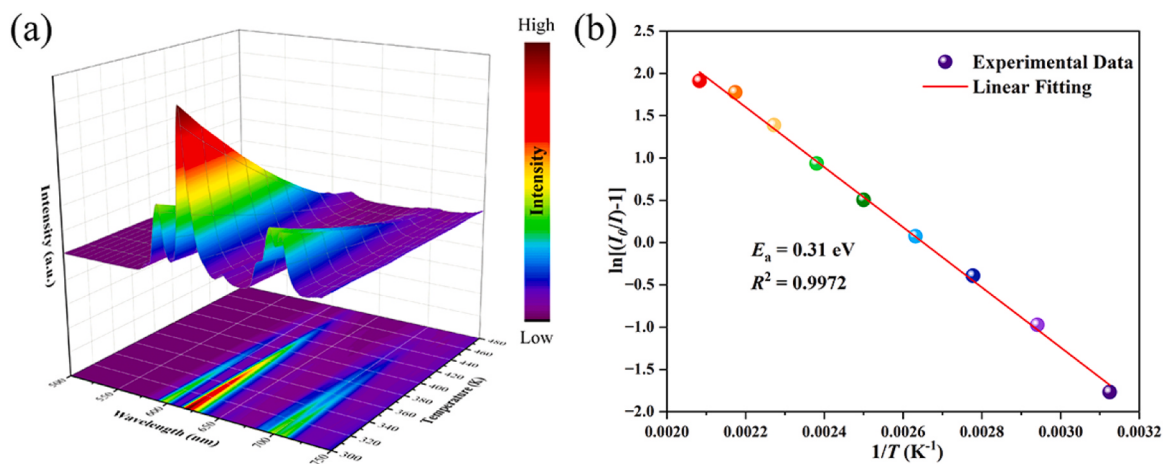


Fig. 12. (a) Three-dimensional thermal quenching diagram for the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor, (b) Fitting graph of $\ln(I_0/I_T - 1)$ and $1/T$.

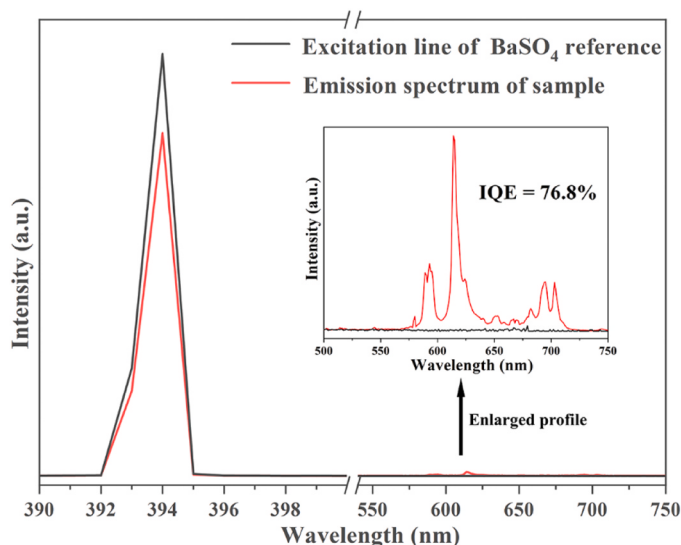


Fig. 13. IQE of the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor.

phosphor and the excitation line of the BaSO₄ reference. The formula below provides a method for computing the IQE [52]:

$$\eta = \frac{\int L_S}{\int E_R - \int E_S} \# \quad (9)$$

The IQE (η) is calculated using the emission spectrum (L_S) along with two absorption spectra: E_R (BaSO₄ reference) and E_S (sample). The IQE value of the NaSrTiTaO₆:40 mol%Eu³⁺ samples exhibits an impressive IQ of 76.8%, significantly surpassing Gd₂MoO₆:Eu³⁺ (47.5%) [53], K₂Mg₃Zn₂Si₁₂O₃₀:Eu³⁺ (10.55%) [54], and BaSrYZrO_{5.5}:Eu³⁺ (15.86%) [7]. This result reveals that NaSrTiTaO₆:40 mol%Eu³⁺ phosphor has high efficiency and superior luminescence performance.

3.3. Assembling and properties of the w-LED device

EL spectral characteristics for the single-color LED are presented in Fig. 14 (a). The monochromatic LED presents a resemblance to the red emission spectrum of the NaSrTiTaO₆:40 mol%Eu³⁺ phosphor. Fig. 14 (b) depicts the EL spectrum of the assembled w-LED prepared by combining red (NaSrTiTaO₆:40 mol%Eu³⁺), blue (BAM), and green ((Ba, Sr)₂SiO₄:Eu²⁺) phosphors. The McCamy equation provides a computational method to determine CCT, which serves as one of the most important indicators for phosphor characterization [55]:

$$\text{CCT} = -449n^3 + 3525n^2 - 6823n + 5520.33 \quad (10)$$

Examination of the electroluminescence spectra reveals that the developed white LED devices demonstrate favorable color characteristics, with a CCT of 5864 K and an excellent color rendering index

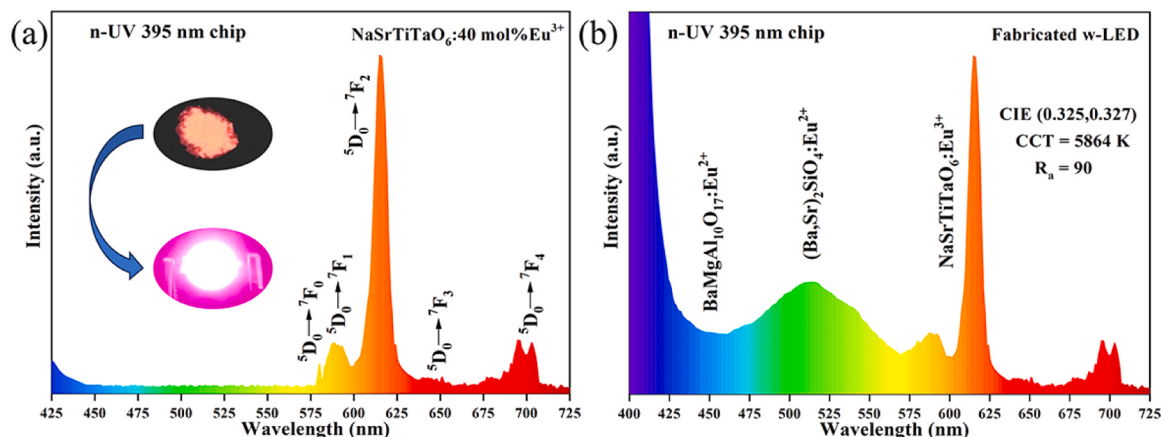


Fig. 14. EL spectrum of the fabricated red LED with $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ (a) and the packaged w-LED (b).

reaching 90. This remarkable R_a value surpasses those of several reported phosphors, such as $\text{Gd}_2\text{MoO}_6:\text{Eu}^{3+}$ ($R_a = 72$) [53], $\text{SrLaLiTeO}_6:\text{Dy}^{3+}$, Eu^{3+} ($R_a = 87$) [56], and $\text{Ca}_3\text{Y}_2\text{Ge}_3\text{O}_{12}:\text{Tb}^{3+}$, Eu^{3+} ($R_a = 87$) [57]. Furthermore, the EL spectrum of the prepared red LED overlaps well with the absorption peaks of plant photosynthetic pigments, indicating potential for plant growth lighting and wavelength-converting agricultural films.

The CIE chromaticity coordinate of the fabricated w-LED is shown in Fig. 15 (a). The fabricated w-LED demonstrates chromaticity coordinates (0.325, 0.327) within the white area of the CIE diagram, approaching the equal energy point at (0.333, 0.333). Thus, it can be concluded that the mixture of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor and suitable green and blue phosphors is an effective method to realize white-light emission. The CRI, including its sub-parameters (R_1 – R_{14}), is also a crucial metric for assessing the suitability of phosphors for use in w-LEDs. Comparisons between the CRI (R_1 – R_{14}) of the experimentally fabricated w-LED and the commercial w-LED combining $\text{YAG}:\text{Ce}^{3+}$ phosphor and UV chip are shown in Fig. 15 (b). Comparative analysis reveals that the fabricated w-LED consistently demonstrates superior color rendering performance relative to its commercial counterpart. In particular, the values of R_9 , serving as a metric to assess the quality of vivid red reproduction, outperform present commercial w-LEDs. The fabricated w-LED is characterized by an R_9 color rendering index of 80, a value 5.33 times higher than that of conventional commercial devices. This result demonstrates the promising application prospects of

$\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ in white light-emitting diode technology.

3.4. Application of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ + @OA phosphor for LFP development

FT-IR spectra of samples can examine the surface properties of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$. Fig. 16 (a) depicts the comparison of FT-IR spectra between the unmodified $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ and the $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ @OA sample. The FT-IR spectrum in Fig. 16 (a) shows characteristic absorptions around 3800 cm^{-1} and 1800 cm^{-1} . The former corresponds to $\text{H}-\text{O}$ stretching, confirming trace water content in the $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ @OA material, while the latter is characteristic of the $\text{C}=\text{O}$ group in oleic acid. Fig. 16 (b) exhibited enlarged FT-IR spectra ranging from 3100 cm^{-1} to 2750 cm^{-1} . Compared with unmodified $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$, FT-IR analysis of $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ @OA revealed two distinct absorption bands at 2987 cm^{-1} and 2902 cm^{-1} . The observed vibrational bands correspond to the stretching vibrations of $\text{C}-\text{H}$ groups of oleic acid, confirming successful surface modification of the $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor [58,59]. Specifically, the carboxyl groups of oleic acid coordinate with surface Eu^{3+} ions on the $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ phosphor. This interaction drives the self-assembly of oleic acid under hydrothermal conditions, forming a hydrophobic coating. Consequently, the modified particles exhibit high hydrophobicity, enabling them to selectively adsorb non-polar sebum constituents, such as oils,

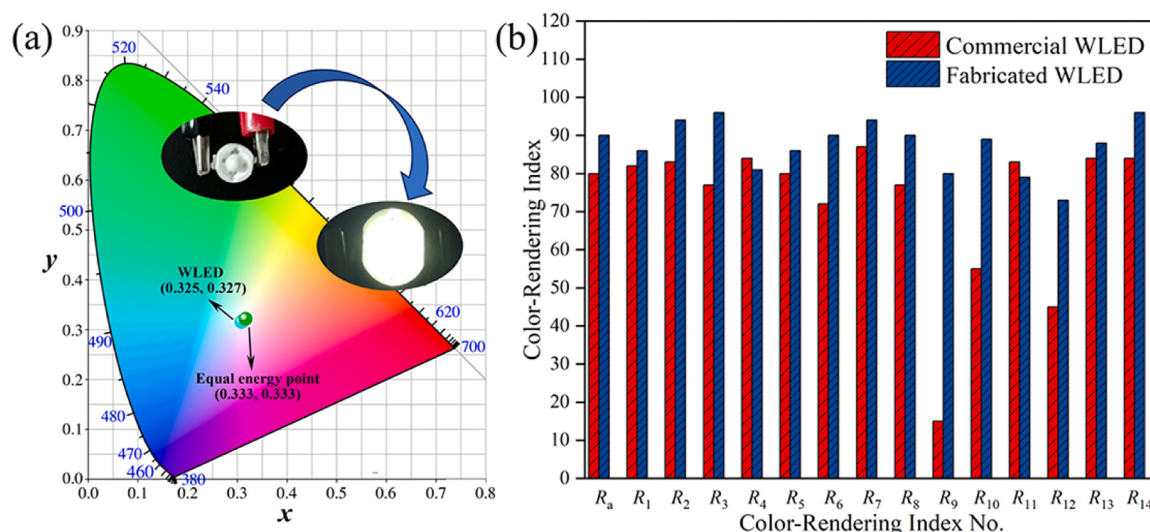


Fig. 15. (a) CIE coordinates and photos of the w-LED. (b) Comparison of the CRI between the fabricated w-LED and the commercial w-LED.

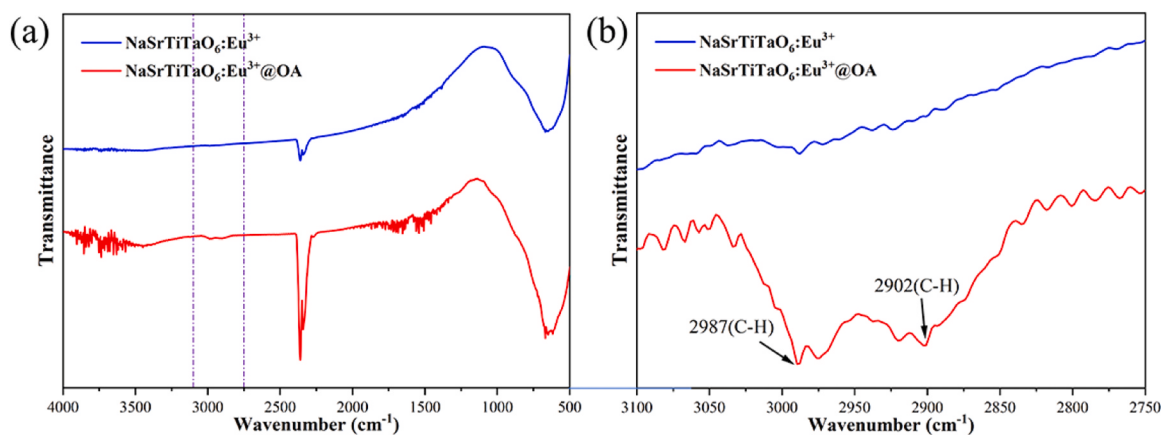


Fig. 16. (a) The FT-IR spectra of the unmodified and OA-modified $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+}$ products, (b) The enlarged FT-IR spectra in the range of 3100 cm^{-1} to 2750 cm^{-1} .

fatty acids, and lipids, within latent fingerprint residues. This strong affinity ensures precise adherence to fingerprint ridges, significantly enhancing the resolution and clarity of fingerprint visualization.

Fig. 17 displays the fluorescence patterns of latent fingerprints developed with $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+} @ \text{OA}$ phosphor, which obviously present characteristics at different levels. The whorl (1) and delta (2) constitute primary Level I features that serve as morphological markers for basic fingerprint categorization. By comparing Level I, Level II, and Level III features, identifying fingerprints achieves greater

precision through the analysis of island (3), bifurcation (4), termination (5), spur (6), sweat pore (7), and crease (8). Through the three-level characteristics of the fingerprint, the fingerprint structure can be accurately identified, which confirms $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+} @ \text{OA}$ phosphor as a highly promising luminescent material for latent fingerprint visualization applications. Meanwhile, the pixel intensity distribution across the designated region is illustrated in Fig. 17. The high red and low values clearly distinguish the ridge and groove, which demonstrates the superior resolving capability of latent fingerprint

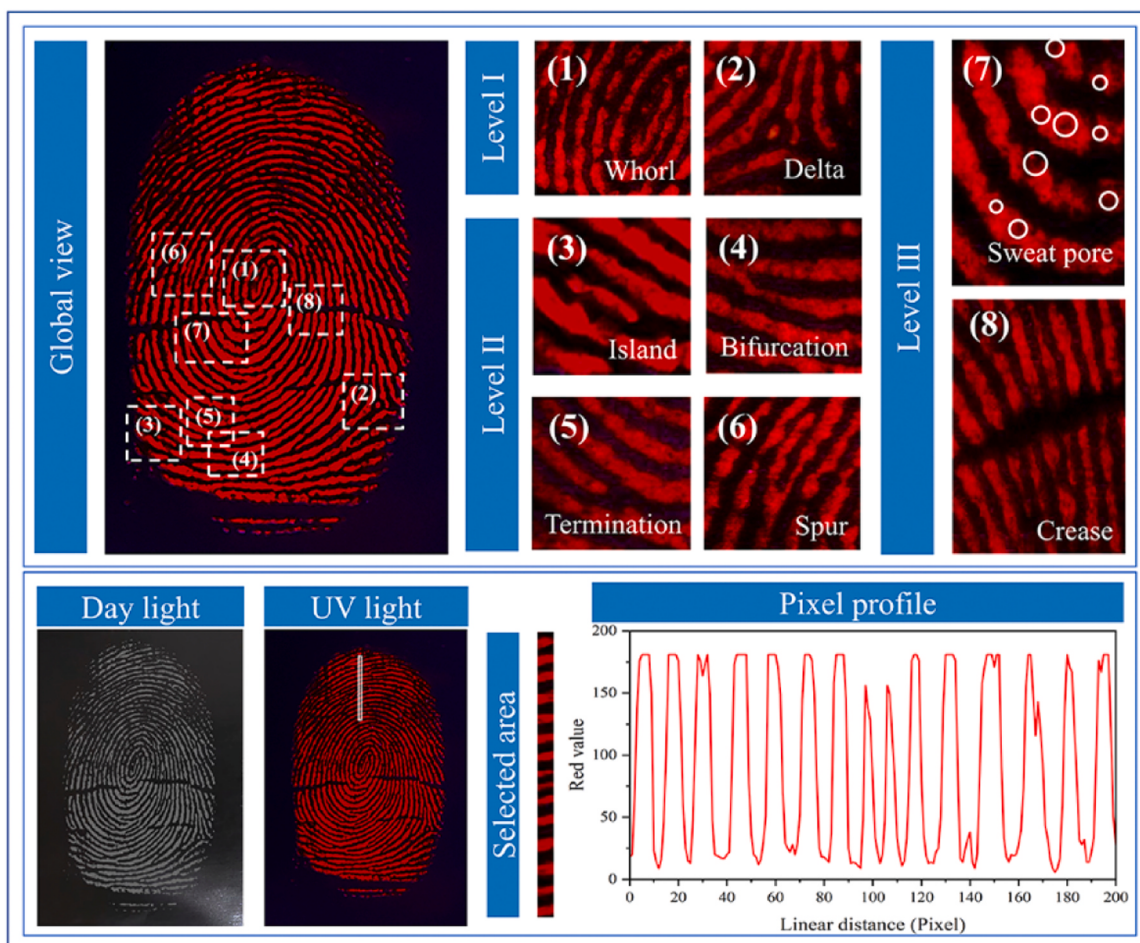


Fig. 17. Global view, microscopic pictures (Level I-III), and the pixel profile of the LFP on aluminum foil stained by $\text{NaSrTiTaO}_6:40 \text{ mol}\% \text{Eu}^{3+} @ \text{OA}$.

development.

4. Conclusions

A series of Eu^{3+} -activated NaSrTiTaO_6 phosphors were synthesized via a high-temperature solid-state reaction. The phase purity and cubic perovskite structure (space group $Pm\bar{3}m$) are confirmed by structural analyses, while EDS mapping indicates a uniform elemental dispersion. Spectral analysis reveals that the $\text{NaSrTiTaO}_6:\text{Eu}^{3+}$ phosphor exhibits optimal excitation at 397 nm and intense red emission dominated by the $^5\text{D}_0 \rightarrow ^7\text{F}_2$ transition at 615 nm. Optimization studies identified 40 mol% as the ideal doping concentration, yielding maximum emission intensity and high color purity (99.9%). The material demonstrates a microsecond-scale fluorescence lifetime and a quantum efficiency of 76.8%. The w-LEDs fabricated with this phosphor achieve near-ideal chromaticity coordinates (0.325, 0.327) and high color rendering ($R_a = 90$), indicating its suitability as a candidate red component for w-LED applications. Furthermore, the EL spectrum suggests promise for plant growth lighting, anti-counterfeiting ink, and agricultural films. Surface-modified phosphors enable high-resolution visualization of latent fingerprints across Levels I–III. These results highlight the multifunctional potential of the $\text{NaSrTiTaO}_6:\text{Eu}^{3+}$ phosphor.

Compliance with Ethical Standards

This article does not contain any studies with human participants or animals performed by any of the authors.

CRediT authorship contribution statement

Daixin Hu: Visualization. **Jingyue Yan:** Formal analysis. **Junjie Feng:** Visualization. **Anyu Cheng:** Funding acquisition. **Jingfei Wang:** Investigation. **Fei Li:** Software. **Leyi Zhang:** Writing – original draft, Data curation. **Xianchao Du:** Resources. **Ruijin Yu:** Writing – review & editing, Resources. **Dan Zhang:** Writing – review & editing.

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

All data included in this study are available upon request by contact with the corresponding author.

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