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**Warm-white luminescence of Dy³⁺ and Sm³⁺ co-doped NaSrPO₄ phosphors
through energy transfer between Dy³⁺ and Sm³⁺ ions**

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Abstract: Series of white-light-emitting NaSrPO₄:Dy³⁺ phosphors were synthesized via a sol-gel method. Small amounts of Sm³⁺ ions were co-doped into the phosphors to enhance their luminescence. The effects of Dy³⁺/Sm³⁺ co-doping concentrations and synthesis temperature on the phosphors' structures and luminescence properties were investigated. The results of X-ray diffraction confirmed that the characteristic diffraction peaks of the phosphors were in accord with those of the standard NaSrPO₄ structure. Energy dispersive spectroscopy indicated that the NaSrPO₄:Dy³⁺ samples were prepared with stoichiometric ratios of elements. Under ultraviolet light excitation of 348 nm, the NaSrPO₄:Dy³⁺ phosphors emitted a white light which was composed of two emission peaks at 484 nm (blue) and 577 nm (yellow) corresponding to the magnetic dipole transition ⁴F_{9/2} → ⁶H_{15/2} and electronic dipole transition ⁴F_{9/2} → ⁶H_{13/2} of Dy³⁺ ions, respectively. Based on Sm³⁺ co-doping, a characteristic red emission at 602 nm arose and a warm-white light containing a lower

CCT value was obtained. Besides, the luminescence lifetime of Dy^{3+} decreased while that of Sm^{3+} increased as dosages of Sm^{3+} were added, indicating efficient energy transfer from Dy^{3+} to Sm^{3+} happened. Therefore, $\text{NaSrPO}_4:\text{Dy}^{3+}$, Sm^{3+} phosphors are promising candidates for application in warm-white light-emitting diodes.

Keywords: NaSrPO_4 ; Rare earth; Sol-gel method; Warm white emission; Energy transfer.

1. Introduction

As environmental pollution and the energy crisis have become increasingly serious, white-light-emitting diodes (w-LEDs) are being increasingly utilized in illuminating systems and display applications [1-4], by virtue of their environmental friendliness, energy savings, and high energy efficiency [5-7]. Typically, widespread w-LEDs are composed of a blue InGaN chip and a yellow $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ (YAG:Ce) phosphor [8-11]. There are shortcomings to this combination, just as highly correlated color temperature ($CCT > 5000$ K) and low color rendering index ($CRI < 80$), since lack of a red emission component [12-15]. Additionally, some researchers have made novel w-LEDs with an ultraviolet (UV) LED chip and tri-color (RGB) phosphors to realize white light. However, this method has several disadvantages related to color ratio control, color reabsorption, and luminous efficiency [16,17]. Therefore, a novel single-component phosphor that produces excellent white light emission under UV-light excitation is urgently needed [18,19].

In phosphors, rare earth Dy^{3+} ions possess two characteristic emission peaks: a blue peak at about 480 nm ($^4\text{F}_{9/2} \rightarrow ^6\text{H}_{15/2}$) and a yellow one at about 580 nm ($^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$) [20,21]. White light is supposed to be achieved by uniting with blue and yellow emissions for w-LEDs applications [22-24]. Recently, numerous phosphors have been reported with admirable luminescence properties, such as

$\text{Na}_3\text{MgZr}(\text{PO}_4)_3:\text{Dy}^{3+}$ [25], $\text{Ba}_3\text{Y}(\text{PO}_4)_3:\text{Dy}^{3+}$ [26], and $\text{Ba}_3\text{La}(\text{PO}_4)_3:\text{Dy}^{3+}$ [27].

However, for the shortage of a red ingredient, the white emission of these Dy^{3+} -doped phosphors have the drawbacks of low *CRI* and high *CCT* values. This is similar to using the combination of an InGaN chip with a YAG:Ce phosphor. In order to ameliorate these defects, red-emitting center Sm^{3+} ions can be introduced into Dy^{3+} -doped phosphors as they have a forceful emission peak relatively at 602nm ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$) that can improve the luminescence properties (*CRI* and *CCT*) of the white emission of Dy^{3+} [28]. By adjusting the ratio of the Dy^{3+} to Sm^{3+} concentration, excellent white light emissions, especially warm white, can be achieved. Phosphates are extensively utilized as phosphor hosts for their low synthesis temperatures and good chemical and thermal stability, including $\text{Ca}_2\text{Sr}(\text{PO}_4)_2:\text{Dy}^{3+}$ [29], $\text{LiCaPO}_4:\text{Ce}$ [30], and $\text{YPO}_4:\text{Dy}^{3+}\text{Eu}^{3+}\text{Tb}^{3+}$ [31]. Wet chemical methods can provide low synthesis temperatures, good crystal structures, and excellent luminescence properties. However, there has been little research on its structure or the preparation of rare-earth-doped NaSrPO_4 by wet chemical methods [32-34], especially in Dy^{3+} -doped NaSrPO_4 white-emitting phosphors [35].

In this paper, $\text{NaSrPO}_4:\text{Dy}^{3+}$, Sm^{3+} phosphors were prepared using a sol-gel method and white light was expected to be obtained from their blue and yellow emissions. Sm^{3+} co-doping would adequately ameliorate the *CRI* and *CCT* values of white light emission. The impacts of $\text{Dy}^{3+}/\text{Sm}^{3+}$ co-doping concentration and synthesis temperature on the structure and white light emission of $\text{NaSrPO}_4:\text{Dy}^{3+}$ samples were investigated. The results provide a reference for research into other white-light-emitting phosphors.

2. Experimental

All $\text{NaSrPO}_4:\text{Dy}^{3+}$, Sm^{3+} specimens were synthesized through a sol-gel method. The

starting materials CH_3COONa , $\text{Sr}(\text{NO}_3)_2$, $\text{NH}_4\text{H}_2\text{PO}_4$, $\text{C}_6\text{H}_8\text{O}_7$, Dy_2O_3 , and Sm_2O_3 were highly purified. The doping concentrations of Dy^{3+} were controlled within 0.5–5 mol%, while that of Sm^{3+} were regulated as 1–3 mol% (relative to the molar amount of Sr^{2+}). All raw materials were weighed out with stoichiometric ratios. First, a certain amount of Dy_2O_3 and Sm_2O_3 were dissolved in nitric acid to produce a nitrate solution, while suitable CH_3COONa , $\text{Sr}(\text{NO}_3)_2$, and $\text{NH}_4\text{H}_2\text{PO}_4$ were added to a conical flask containing distilled water. Afterward, an appropriate amount of citric acid was drizzled into the above solution to form a stable system. The system was stirred fully by vigorous magnetic stirring at 70 °C for 2.5 h to form a transparent sol and heated by an air circulation oven at 80 °C for 36 h until a swelled yellow porous gel was formed. Ultimately, the sample was moved to a furnace and heated at 800–1200 °C for 3 h. The as-prepared phosphors were obtained when cooled naturally to room temperature (*RT*).

The phase structure and purity of these as-prepared specimens were evaluated utilizing X-ray diffraction (XRD) on an X-ray diffractometer (DX-2700, Aolong Company, China) with $\text{Cu K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) and a 2θ scanning speed of 5°/min. The photoluminescence (PL) and lifetime measurements were carried out by a fluorescence spectrophotometer with excitation by a 150 W Xe lamp (F-4600, Hitachi, Japan). The quantum efficiency of as-prepared phosphors was acquired by a Quantum Efficiency Measurement System (QEMS 2000, Labsphere, USA). The morphologies and elemental compositions of the synthesized powders were verified via JSM-7800F scanning electron microscopy (SEM; JEOL, Japan) with energy dispersive spectroscopy (EDS). The absorption spectrogram of relevant powders was monitored through a UV-Vis-NIR diffuse reflectance (DR) spectrophotometer (Cary 5000, Agilent Inc., USA). All the above characterization tests were proceeded at *RT*.

3. Results and discussion

The XRD patterns of NaSrPO₄:Dy³⁺ powders heated at 800–1200 °C are represented in Fig. 1. All diffraction peaks of these specimens present a good match with the standard phase of NaSrPO₄ (PDF#33-1282) [32] and can be indexed as a monoclinic crystal with lattice parameters of $a = 20.414 \text{ \AA}$, $b = 5.429 \text{ \AA}$, and $c = 17.426 \text{ \AA}$. This proves that NaSrPO₄:Dy³⁺ samples with NaSrPO₄ structure are successfully prepared and no obvious impurity phases corresponding to Dy³⁺ appear. In addition, a low Dy³⁺-ion doping concentration does not lead to significant phase changes in the NaSrPO₄ structure in view of the close ionic radii of Sr²⁺ ($r = 0.118 \text{ nm}$) and Dy³⁺ ($r = 0.091 \text{ nm}$). Table 1 displays the corresponding lattice parameters and volumes of these samples. Apparently, increasing the thermal energy enhances diffusion and migration among diverse ions, which causes a slight decrease in cell volume. This can be interpreted as resulting from an increasing number of Dy³⁺ ions entering the host structure to form a luminescence center [36].

The SEM images and corresponding EDS results of NaSrPO₄:Dy³⁺ samples prepared at 800 °C and 1000 °C are shown in Fig. 2. The samples prepared at 800 °C and 1000 °C (Figs. 2(a) and 2(b), respectively) exhibit homogeneous size distributions with particle diameters of about 2 μm and 4 μm at 800 °C and 1000 °C, respectively. Meanwhile, the particle sizes increase rapidly with increases in synthesis temperature. As the synthesis temperature increases, the particle size and degree of bonding are affected to some extent. For these grains acquire an increasing thermal energy, there is a tighter bonding among particles, and the particle size of the phosphor prepared at 1000 °C is larger than that prepared at 800 °C. Moreover, the EDS spectrum shown in Fig. 2(c) provides a distribution of the elemental content at a marker point. The elemental weight percentage of the NaSrPO₄:2%Dy³⁺ sample prepared at 1000 °C and

stoichiometric atom ratio are listed and compared. It can be clearly observed that the sample contained the elements O, Na, P, Sr, and Dy. Compared with the elemental weight percentage of the stoichiometric ratio obtained by theoretical calculation, the as-prepared phosphor has good correspondence to the standard composition, and only a small amount of Na is evaporated at high temperature.

Fig. 3 illustrates the emission spectra of $\text{NaSrPO}_4:\text{Dy}^{3+}$ processed at specified temperatures with an excitation at 348 nm. Two emission peaks located at 484 nm (blue) and 577 nm (yellow), which originated from the magnetic dipole transition $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{15/2}$ and electronic dipole transition $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$ of Dy^{3+} ions, respectively. There are no significant shifts of the emission peaks, although the luminescence intensities of $\text{NaSrPO}_4:\text{Dy}^{3+}$ increases with synthesis temperature up to 1000 °C and later declines. The synthesis temperature produces two concrete roles on its interior structure, which affects the luminescence property of the specimens [14]: On one hand, the crystallization of NaSrPO_4 and diffusion of the Dy^{3+} ions in the structure are favored by high temperatures; on the other hand, heating at high temperatures leads to the formation of large crystal grains. At synthesis temperatures of 800–1000 °C, the first aspect acts a critical section in improving the luminescence performance, while the second affects the luminescence obtained above 1000 °C.

As Dy^{3+} ions are the luminescence center, the Dy^{3+} doping concentration significantly influences the relevant luminescence intensities of these as-prepared powders. The XRD mappings of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ samples with particular Dy^{3+} amounts are represented in Fig. 4. With increased Dy^{3+} doping concentrations, the diffraction mappings of all as-prepared powders exhibit much consistency in the standard NaSrPO_4 structure without impurity peaks, indicating that Dy^{3+} ions do not affect the host structure.

By zooming the patterns in the 2θ angle range of 30° – 34° , an enlarged image of the main diffraction peaks is obtained (Fig. 4(b)). As smaller Dy^{3+} (0.091 nm) ions successfully enter the larger Sr^{2+} ($r = 0.118$ nm) sites in the monoclinic crystal lattice of NaSrPO_4 , the diffraction peaks offset mildly to the larger 2θ angle positions. Moreover, the unit-cell volume of Dy^{3+} -doped NaSrPO_4 is slightly lower than that of the original host, as shown in Table 2.

The excitation spectrum ($\lambda_{\text{em}} = 484$ nm), UV-Visible absorption spectrogram, and emission spectra ($\lambda_{\text{ex}} = 348$ nm) of as-prepared $\text{NaSrPO}_4:\text{x}\text{Dy}^{3+}$ are shown in Fig. 5. In Fig. 5(a), several apparent excitation peaks of $\text{NaSrPO}_4:\text{Dy}^{3+}$ are visible at 348 nm, 384 nm, 362 nm, 322 nm, and 424 nm, which correspond to the ${}^6\text{H}_{15/2} \rightarrow {}^4\text{M}_{15/2}$, ${}^6\text{H}_{15/2} \rightarrow {}^4\text{M}_{21/2}$, ${}^6\text{H}_{15/2} \rightarrow {}^4\text{P}_{3/2}$, ${}^6\text{H}_{15/2} \rightarrow {}^4\text{K}_{15/2}$ and ${}^6\text{H}_{15/2} \rightarrow {}^4\text{I}_{15/2}$ transitions of Dy^{3+} ions, respectively [35]. The excitation spectrum indicates that as-prepared $\text{NaSrPO}_4:\text{Dy}^{3+}$ phosphor is capable to be productively excited by ultraviolet radiation.

The UV-Vis absorption spectrogram of $\text{NaSrPO}_4:\text{Dy}^{3+}$ is shown in Fig. 5(b) for verification. The sample has an intense absorption region between 200 nm and 300 nm, which match with the transient state of charge transfer between phosphorus and oxygen. Meanwhile, two visible absorption peaks at 348 nm and 384 nm that are highly consistent with the excitation spectrum and are also attributed to the ${}^6\text{H}_{15/2} \rightarrow {}^4\text{M}_{15/2}$ and ${}^6\text{H}_{15/2} \rightarrow {}^4\text{M}_{21/2}$ electronic transitions of Dy^{3+} ions. Moreover, the stronger absorption at 348 nm indicates that there is suitable excitation of Dy^{3+} ions, which can lead to the excellent photoluminescence property of $\text{NaSrPO}_4:\text{Dy}^{3+}$.

The emission spectra of $\text{NaSrPO}_4:\text{x}\text{Dy}^{3+}$ phosphors with Dy^{3+} concentrations ($x = 0.5$ – 4%) are presented in Fig. 5(c). Under 348 nm excitation, two high-intensity emission peaks from Dy^{3+} are detected at 484 nm and 577 nm. Therefore, the concentration of Dy^{3+} doping only affects the luminescence intensities but not the

peak positions. The luminescence intensities of phosphors increase with increases in Dy^{3+} concentration from 0.5% to 2%, resulting in increases in the luminescence centers, which reach a maximum at a doping concentration of 2%. As the concentration increases further, a sudden fall in luminescence intensity appears because of the concentration quenching phenomenon.

White light can be generated by assembling the characteristic emission peaks of blue and yellow of Dy^{3+} , so their luminescence intensities are important. Fig. 5(d) exhibits the relevance of the luminescence intensities of the two corresponding peaks on the specified Dy^{3+} concentration, which can explain the correlation between dosage concentration and the luminescence intensity ratios of the two corresponding peaks located in 484 nm and 577 nm ($I_{484 \text{ nm}}/I_{577 \text{ nm}}$). The $I_{484 \text{ nm}}/I_{577 \text{ nm}}$ ratios of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ ($x = 0.5\%, 1\%, 2\%, 3\%$, and 4%) are calculated as 1.139, 1.050, 1.078, 1.027, and 1.044, respectively. The calculation results reveal that the $I_{484 \text{ nm}}/I_{577 \text{ nm}}$ ratio can be altered by the Dy^{3+} doping concentration, although they are close to 1. Hence, we suppose that better quality white light can be obtained through adjusting the ratio of $I_{484 \text{ nm}}/I_{577 \text{ nm}}$.

The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ ($x = 0.5\%, 1\%, 2\%, 3\%$, and 4%) phosphors are shown in Fig. 6. It indicates the relevant coordinates of the samples sit in white-light area and are near to the standard white coordinates (0.33, 0.33). The coordinates of all phosphors are distributed on a line between the blue and yellow light points. Due to the influence of the blue-yellow ratio, the positions of the color coordinates also vary slightly.

Therefore, it is suggested that ideal white-light will hopefully be manufactured via regulating the blue-yellow ratio and color coordinates.

To further explore the concentration quenching phenomenon of luminescence among adjacent Dy^{3+} ions, the critical radius distance (R_c) can be counted as [37]:

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

where R_c denotes the critical distance, V is the unit cell volume, N represents the cation sites from a unit cell in NaSrPO_4 host, and x_c refers the critical concentration of Dy^{3+} . In this case, R_c is calculated to be 22.50 Å via applying the following reference values: $V = 1907.85 \text{ Å}^3$, $x_c = 0.02$, $N = 16$. A larger distance among Dy^{3+} ions makes the exchange interactions less effective, while a multipole interaction is superior. Accordingly, There are two types of energy transfer: one is exchange interaction, another is multipole-multipole interaction. Based on the calculation of R_c ($> 5 \text{ Å}$), the conclusion of multipole-multipole interaction mechanism plays main role in energy transfer of Dy^{3+} ions can be offered. To further analyze the mechanism, the following equation can be used according to Dexter's theory [38]:

$$I/x = k/[1 + \beta(x)^{Q/3}] \quad (2)$$

where k and β indicate two constants in a NaSrPO_4 host; and I and x represent the luminescence intensity of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ ($x = 2\%, 3\%, 4\%, 5\%, 6\%$) and molar fraction of the specific Dy^{3+} concentration, respectively. Q shows the form of interaction among dipole-dipole, dipole-quadrupole, and quadrupole-quadrupole interactions, correspond to $Q = 6, 8$, and 10 , respectively. Provided $\beta(x)^{Q/3}$ is much larger than 1, the above equation can be equivalently converted to the following [39]:

$$\log(I/x) = C - \frac{Q}{3}\log(x); (C = \log k - \log \beta) \quad (3)$$

Fig. 7 demonstrates the relevance of $\log(I/x)$ and $\log(x)$ of the Dy^{3+} -ion peaks at 484 nm and 577 nm. By fitting, the slopes of the lines are -2.3259 and -2.1890 , respectively. The Q values are calculated as 6.9778 and 6.5670, which are near to 6. Hence, the dominating cause of concentration quenching of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ can be

considered to involve dipole-dipole interactions.

The above consequences are verified that white light is promising to be achieved through uniting the two characteristic blue and yellow emissions of NaSrPO₄:Dy³⁺ phosphors. Moreover, Sm³⁺ ions should be co-doped in the phosphors to improve luminescence, especially the *CCT* value.

The XRD patterns of NaSrPO₄:2%Dy³⁺, *y*Sm³⁺ (*y* = 1%, 2%, 3%) phosphors are shown in Fig. 8. Like Fig. 4, the patterns for NaSrPO₄:Dy³⁺, Sm³⁺ specimens are in keeping with the standard structure of NaSrPO₄; hence, Dy³⁺, Sm³⁺ co-doping does not cause apparent variations in the host structure. Meanwhile, the lattice parameters and volumes of NaSrPO₄:Dy³⁺, Sm³⁺ are revealed in Table 3 which interpret that small amounts of Dy³⁺ and Sm³⁺ ion doping have no distinct effect in the overall structure of the samples by cause of the close ionic radius among Sr²⁺ (*r* = 0.118 nm), Dy³⁺ (*r* = 0.091 nm), and Sm³⁺ (*r* = 0.096 nm). Compared with the standard NaSrPO₄ structure, by virtue of the tiny difference in ion radius, the relevant cell volumes and lattice parameters of the as-prepared specimens decrease and distort slightly. In addition, this phenomenon shows that Dy³⁺ and Sm³⁺ ions enter the host structure without altering its structure.

The emission spectra ($\lambda_{\text{ex}} = 348 \text{ nm}$) and corresponding energy transfer efficiency of NaSrPO₄:2%Dy³⁺, *y*Sm³⁺ phosphors with specified Sm³⁺ co-doping concentrations are put forward in Fig. 9. Apart from the two original peaks of Dy³⁺ ions located at 484 nm and 577 nm, a new emission peak appears at 602 nm (Fig. 9(a)), which stems from the characteristic ⁴G_{5/2}→⁶H_{7/2} transition from Sm³⁺. The results show that as the co-doping concentration of Sm³⁺ increases gradually, the luminescence intensities of the Sm³⁺ characteristic peak at 602 nm increase, while the luminescence intensities of the Dy³⁺ characteristic emission peaks decrease. This phenomenon is interpreted as

the result of transmission in energy from Dy^{3+} to Sm^{3+} .

The energy transfer efficiency (η_{ET}) between Dy^{3+} and Sm^{3+} can be calculated as follows:

$$\eta_{\text{ET}} = 1 - \frac{I_s}{I_{s0}} \quad (4)$$

where I_{s0} expresses the intrinsic luminescence intensity of single-doped Dy^{3+} and I_s refers to the luminescence intensity of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ co-doping Sm^{3+} . Hence, the η_{ET} values, accompanying Sm^{3+} concentrations of 1%, 2%, and 3% are calculated as 34.7%, 47.6%, and 70.6%, respectively (Fig. 9(b)).

To describe the energy transfer ($\text{Dy}^{3+} \rightarrow \text{Sm}^{3+}$) and their level structure more clearly, a schematic plot of energy transfer is presented in Fig. 10. Under UV-light irradiation, the electrons in the 4f electronic shell of Dy^{3+} in the ground-state orbitals $^6\text{H}_{15/2}$ absorb light and leap into higher energy levels ($^6\text{P}_{7/2}$, $^6\text{P}_{5/2}$, $^4\text{I}_{15/2}$, etc.). Radiative transition occurs among high energy levels and results in the electrons falling into the $^4\text{F}_{9/2}$ level. Subsequently, absorbed energy is released as the Dy^{3+} electrons get back to the ground-state orbitals of $^6\text{H}_{15/2}$ and $^6\text{H}_{13/2}$, realizing two characteristic blue light (484 nm) and yellow light emissions (577 nm). Meanwhile, some of the energy is transferred to Sm^{3+} electrons from Dy^{3+} . Electrons of Sm^{3+} in ground state transition to an excited state $^6\text{P}_{3/2}$ after gaining transfer energy. Accompanied by radiative transition, the high-energy electrons of Sm^{3+} migrate to three ground states $^6\text{H}_{5/2}$ (566 nm) $^6\text{H}_{7/2}$ (602 nm) and $^6\text{H}_{9/2}$ (649 nm) from the $^4\text{G}_{5/2}$ excited state and a characteristic red emission is produced.

To further verify the $\text{Dy}^{3+} \rightarrow \text{Sm}^{3+}$ energy transfer, decay curves are depicted to evaluate the luminescence lifetimes of Dy^{3+} ($^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$) and Sm^{3+} ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$) in $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ ($y = 1\%, 2\%, 3\%$) phosphors (Fig. 11). The decay curves which identify with an especial function of double exponential, can be fitted via the

formula [40]:

$$I(t) = I_0 + A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \quad (5)$$

where A_1 and A_2 are fitting constants; while τ_1 and τ_2 signify the lifetimes of exponential sections; I_0 and $I(t)$ represent the luminescence intensities at times 0 and t , respectively. Conforming to the above parameters, the decay times for different concentrations of Sm^{3+} co-doped $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ phosphors can be assessed through the following functional equation [41]:

$$\tau = (A_1\tau_1^2 + A_2\tau_2^2)/(A_1\tau_1 + A_2\tau_2) \quad (6)$$

Compared with Dy^{3+} -doped phosphors, Dy^{3+} - and Sm^{3+} -codoped samples monitored at 484 nm show decay-time decreases from 1.2823 ms to 0.9564 ms, along with the specified Sm^{3+} doping concentrations gradually increase from 1% to 3%. Meanwhile, the decay times monitored at 602 nm increase from 2.1274 ms to 2.8006 ms. Briefly, energy is capable of being transferred effectively from Dy^{3+} to Sm^{3+} .

The CIE chromaticity coordinates of $\text{NaSrPO}_4:\text{Dy}^{3+}$, Sm^{3+} phosphors are measured and summarized in Table 4. Compared with the standard white-light point (0.33, 0.33), all the calculated values move from the white to the near red region. The CCT , a standard measure of color temperature used to reflect lighting quality, can be provided via the equation [42]:

$$CCT = -437n^3 + 3601n^2 - 6861n + 5514.31 \quad (7)$$

where $n = (x - x_e)/(y - y_e)$; and x_e and y_e both equal 0.33; and y_e equal 0.1858.

Although the $\text{NaSrPO}_4:\text{Dy}^{3+}$ phosphor with the 2% doping concentration has a higher luminescence intensity, its CCT value is as high as 5695 K, which can be considered a “cold” white light (blueish). Fig.12 represents the CIE chromaticity coordinates of samples under different doping contents. Along with the specific dosage of Sm^{3+} are added into $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ phosphors, the CCT values decrease from 4506 K to

3989 K, accompanied by a variation in light color from neutral white to “warm” white (reddish). Hence, it illustrates that the CIE chromaticity coordinates are effectively alterable by regulating the doping ratios of Dy^{3+} and Sm^{3+} . The external quantum efficiencies of $\text{NaSrPO}_4\text{:}2\%\text{Dy}^{3+}$, $y\text{Sm}^{3+}$ ($y=1\%$, 2%) phosphors are measured as 25.2% and 21.4%, respectively. All results indicate that the $\text{NaSrPO}_4\text{:Dy}^{3+}$, Sm^{3+} phosphor has great potential for use in commercial applications requiring warm white light.

4. Conclusion

In summary, $\text{NaSrPO}_4\text{:Dy}^{3+}$, Sm^{3+} phosphors were synthesized through a sol-gel method and the white-light emissions from Dy^{3+} and Sm^{3+} were surveyed in detail. The PL spectra indicate that $\text{NaSrPO}_4\text{:Dy}^{3+}$ samples are effectively excited by UV light of 348 nm, and emissions with characteristic peaks at 484 nm and 577 nm combine into white light. The quenching concentration of $\text{NaSrPO}_4\text{:Dy}^{3+}$ is 2%, which coincides with the dipole-dipole interactions. By co-doping with Sm^{3+} ions, $\text{NaSrPO}_4\text{:Dy}^{3+}$ phosphors are supposed to be regulated to realize warm white light emission with a lower *CCT* value. Meanwhile, energy transfer from Dy^{3+} to Sm^{3+} ions in the NaSrPO_4 structure is proven. The results suggest that $\text{NaSrPO}_4\text{:Dy}^{3+}$, Sm^{3+} phosphors are strong candidates for application to w-LEDs.

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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Tables

Table 1 Lattice parameters and volumes of standard NaSrPO₄ and NaSrPO₄:2% Dy³⁺ samples prepared at different temperatures.

Sample	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	<i>V</i> (Å ³)
Standard	20.414	5.429	17.246	1911.33
900 °C	20.409	5.425	17.240	1909.12
1000 °C	20.408	5.421	17.245	1907.85
1100 °C	20.411	5.419	17.237	1906.54

Table 2 Lattice parameters and volumes of NaSrPO₄ doped with Dy³⁺ doping concentrations.

Sample	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	<i>V</i> (Å ³)
NaSrPO ₄ :1%Dy ³⁺	20.411	5.428	17.236	1909.59
NaSrPO ₄ :2%Dy ³⁺	20.405	5.423	17.241	1907.83
NaSrPO ₄ :3%Dy ³⁺	20.406	5.422	17.216	1904.80
NaSrPO ₄ :4%Dy ³⁺	20.403	5.418	17.187	1899.91

Table 3 Lattice parameters and volumes of NaSrPO₄ co-doped with Dy³⁺ and Sm³⁺ co-doping concentrations.

Sample	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	<i>V</i> (Å ³)
2%Dy ³⁺ , 1%Sm ³⁺	20.359	5.426	17.246	1905.13
2%Dy ³⁺ , 2%Sm ³⁺	20.378	5.424	17.212	1902.45
2%Dy ³⁺ , 3%Sm ³⁺	20.399	5.414	17.218	1901.56

Table 4 CIE parameters of the $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ and $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ phosphors.

Point	Sample	CIE(x,y)	<i>CCT</i> /K	Light color
a	$\text{NaSrPO}_4:2\%\text{Dy}^{3+}$	(0.3408, 0.3722)	5129	Cold white light
b	$\text{NaSrPO}_4:2\%\text{Dy}^{3+}, 1\%\text{Sm}^{3+}$	(0.3592, 0.3697)	4506	Neutral white light
c	$\text{NaSrPO}_4:2\%\text{Dy}^{3+}, 2\%\text{Sm}^{3+}$	(0.3821, 0.4084)	4100	Neutral white light
d	$\text{NaSrPO}_4:2\%\text{Dy}^{3+}, 3\%\text{Sm}^{3+}$	(0.3893, 0.4197)	3989	Warm white light

Figure captions

Figure 1 XRD patterns of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ samples prepared at various synthesis temperatures.

Figure 2 SEM images of $2\%\text{Dy}^{3+}$ -doped NaSrPO_4 phosphor prepared at (a) $800\text{ }^\circ\text{C}$ and (b) $1000\text{ }^\circ\text{C}$ and (c) corresponding EDS results.

Figure 3 Emission spectra of $\text{NaSrPO}_4:\text{Dy}^{3+}$ phosphors under excitation of 348 nm synthesized at specific temperatures.

Figure 4 (a) XRD patterns of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ ($x = 1\%, 2\%, 3\%, 4\%$) with specified Dy^{3+} ion concentrations and (b) enlarged patterns in the 2θ ranged $30^\circ\text{--}34^\circ$.

Figure 5 (a) Excitation spectrum ($\lambda_{\text{em}} = 484\text{ nm}$) and (b) UV–vis spectrum of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ at $1000\text{ }^\circ\text{C}$; (c) Emission spectra of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ with different Dy^{3+} concentrations; (d) Relevance of the luminescence intensities of two corresponding peaks (blue and yellow) on Dy^{3+} doping concentration.

Figure 6 CIE coordinates of $\text{NaSrPO}_4:x\text{Dy}^{3+}$ with specified Dy^{3+} ion concentrations.

Figure 7 (a) Fitting results of $\log(I/x)$ and $\log(x)$ for the $^4\text{F}_{9/2}\rightarrow^6\text{H}_{15/2}$ (484 nm) and (b) $^4\text{F}_{9/2}\rightarrow^6\text{H}_{13/2}$ (577 nm) transitions of Dy^{3+} ions in $\text{NaSrPO}_4:\text{Dy}^{3+}$ phosphors.

Figure 8 XRD patterns of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ ($y = 1\%, 2\%, 3\%$) with specified Sm^{3+} ion concentrations.

Figure 9 (a) Emission spectra of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ with specified Sm^{3+} ion concentrations and (b) corresponding energy transfer efficiency from Dy^{3+} to Sm^{3+} in NaSrPO_4 structure.

Figure 10 Schematic diagram of energy transfer from Dy^{3+} to Sm^{3+} in $\text{NaSrPO}_4:\text{Dy}^{3+}, \text{Sm}^{3+}$ phosphor.

Figure 11 Decay curves of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ ($y = 1\%, 2\%, \text{ and } 3\%$)

phosphors inspected at (a) 484 nm and (b) 602 nm.

Figure 12 CIE coordinates of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ and $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ phosphors.

Figure 1

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Figures

Figure 1

XRD patterns of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}$ samples prepared at various synthesis temperatures.

Figure 2

SEM images of 2%Dy³⁺-doped NaSrPO₄ phosphor prepared at (a) 800 °C and (b) 1000 °C and (c) corresponding EDS results.

Figure 3

Emission spectra of NaSrPO₄:Dy³⁺ phosphors under excitation of 348 nm synthesized at specific temperatures.

Figure 4

(a) XRD patterns of NaSrPO_4 : $x\text{Dy}^{3+}$ ($x = 1\%, 2\%, 3\%, 4\%$) with specified Dy^{3+} ion concentrations and (b) enlarged patterns in the 2θ ranged 30° – 34° .

Figure 5

(a) Excitation spectrum (λ_{em} = 484 nm) and (b) UV–vis spectrum of NaSrPO₄:2%Dy³⁺ at 1000 Å; (c) Emission spectra of NaSrPO₄:x%Dy³⁺ with different Dy³⁺ concentrations; (d) Relevance of the luminescence intensities of two corresponding peaks (blue and yellow) on Dy³⁺ doping concentration.

Figure 6

CIE coordinates of NaSrPO₄:xDy³⁺ with specified Dy³⁺ ion concentrations.

Figure 7

(a) Fitting results of $\log(I/x)$ and $\log(x)$ for the 4F_{9/2} → 6H_{15/2} (484 nm) and (b) 4F_{9/2} → 6H_{13/2} (577 nm) transitions of Dy³⁺ ions in NaSrPO₄:Dy³⁺ phosphors.

Figure 8

XRD patterns of NaSrPO_4 :2% Dy^{3+} , $y\text{Sm}^{3+}$ ($y = 1\%, 2\%, 3\%$) with specified Sm^{3+} ion concentrations.

Figure 9

(a) Emission spectra of $\text{NaSrPO}_4:2\%\text{Dy}^{3+}, y\text{Sm}^{3+}$ with specified Sm^{3+} ion concentrations and (b) corresponding energy transfer efficiency from Dy^{3+} to Sm^{3+} in NaSrPO_4 structure.

Figure 10

Schematic diagram of energy transfer from Dy^{3+} to Sm^{3+} in $\text{NaSrPO}_4:\text{Dy}^{3+}, \text{Sm}^{3+}$ phosphor.

Figure 11

Decay curves of NaSrPO₄:2%Dy³⁺, ySm³⁺ (y = 1%, 2%, and 3%) phosphors inspected at (a) 484 nm and (b) 602 nm.

Figure 12

CIE coordinates of NaSrPO₄:2%Dy³⁺ and NaSrPO₄:2%Dy³⁺, ySm³⁺ phosphors.