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Co-DOPING RED-EMITTING $Sr_2Si_5N_8:Eu^{2+}$ INTO YELLOW-EMITTING PHOSPHOR-PACKAGING FOR ENHANCING THE OPTICAL PROPERTIES OF THE 8500 K REMOTE-PHOSPHOR PACKAGING WLEDs

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In the last decades, WLEDs attract more and more consideration in both academic and industrial purposes because of its advantages such as fast response time, environment friendliness, small size, long lifetime, and high efficiency. In this research, by doping the red-emitting Sr2Si5N8:Eu^{2+} phosphor particles into yellow-emitting YAG:Ce phosphor-packaging, a new recommendation for enhancing the optical properties (color uniformity, color rendering index, and lumen output) of the 8500 K remote-phosphor packaging WLEDs is presented, investigated, and demonstrated. By using Mat Lab and Light Tools software based on Mie Theory, the obtained results show that the optical properties of the 8500 K remote-phosphor packaging WLEDs significantly depended on Sr2Si5N8:Eu^{2+} concentration. The results have provided a potential practical recommendation for manufacturing remote-phosphor W-LEDs.

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1. Introduction

Nowadays, light-emitting diodes (LEDs), which is considered as a novel type of solid-state lighting (SSL) source, have been applied in many fields like general illumination, automotive lighting, automobile devices, display backlighting, communication devices/networks, and medical applications [1-3]. From that point of view, more interest in the application of high-power white LEDs (WLEDs) (which consume at least 1 W of power) has been increasing in both academic and industrial purposes. In comparison with traditional light sources, WLEDs have many advantages, such as high efficiency, low power consumption, high reliability, long lifetime, and environmental friendliness. Moreover, researchers have carried out some studies on new materials, advanced manufacturing technology, improved packaging technology, thermal management, and reliability of LEDs and associated products [1-4]. With the development of lighting technology, the technique based on a blue LED chip and yellow phosphor (YAG: Ce3 +) is the most popular way, and phosphor converted WLEDs are widely used for mass production. For WLEDs, the white light consists of transmitted chip-emitted blue rays and phosphor-converted yellow rays in the phosphor coating layer. The phosphor coating structure plays an important role in affecting the illumination quality of pc-WLEDs. Many researchers have devoted themselves to the study of phosphor coating structure, which has been thought of as an important factor to influence the CCT uniformity [1-7]. Optical properties of WLEDs can be enhanced by phosphors

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Sr_1-xBaxSi$_2$O$_2$N$_2$:Eu^{2+}(0x1) [8], or by β-SiAlON:Yb^{2+} phosphor [9], or by controlling the phosphor materials and packaging structures [10], or by Red-Emitting Phosphor Li_2SrSiO_4:Eu^{3+}, Sm^{3+} [11], or by co-doping SiO_2 [12,13] to YAG:Ce phosphor-packaging of WLEDs. The studies show the significant effect of method packaging and materials to the optical properties of W-LEDs. From this point of view, research about the LEDs packaging and its materials is the important direction in LEDs industry.

On another side, the remote phosphor structure of W-LEDs is a structure in which the phosphor is moved far away from the LED chip. This structure could significantly reduce the probability of absorption of the re-emitted light by the WLEDs chip, and they could improve the phosphor efficiency. With these advances, the remote phosphor structure of WLEDs seems to be a proposed solution for manufacture WLEDs [14-16]. However, very rare works have improved and demonstrated the lighting performance of W-LEDs with remote phosphor structure by mixing two or more diffusive particles into the phosphor compound. It is the remaining gap, which could be filled by this research work.

In the last decade, SiN_4-base covalent nitride materials such as $M_2Si_5N_8$:Eu^{2+} and $MAlSiN_3$:Eu^{2+} (M= Ca, Sr, Ba) have been extensively considered as excellent materials for LEDs technology. Among these phosphors, $Sr_2Si_5N_8$:Eu^{2+} presented excellent emission characteristics under a blue excitation wavelength of 455 nm, had a uniform particle size distribution and showed high performance in LEDs packages [17-20]. In this research, the red-emitting $Sr_2Si_5N_8$:Eu^{2+} phosphor particles is remotely added into the yellow-emitting YAG:Ce phosphor of the remote packaging WLEDs (RP-WLEDs). By using the commercial software Light Tools 8.4 and Mat Lab software, the influence of the red-emitting $Sr_2Si_5N_8$:Eu^{2+} phosphor particles on the lighting performance of RP-WLEDs is proposed, investigated and demonstrated. This model shows that CCT deviation (D-CCT), Color rending index (CRI), and the lumen output could be improved significantly. The main contribution of this research can be drawn as the following:

1) The physical model of the remote-packaging WLEDs is conducted by using Light Tools and Mat Lab software based on the Monte Carlo simulation and Mie theory.

2) The effect of the red-emitting phosphor particles' concentration on D-CCT, CRI and lumen output is derived.

The end of this paper can introduce by the following sections. Firstly, the physical model of RP-WLEDs is conducted by using the commercial software Light Tools, and the mathematical description is calculated using Mat Lab software. Then, the simulation results are analyzed and demonstrated. Finally, some conclusions are drawn from the research results.

2. Physical and mathematical model

In this section, the 8500 K remote-packaging WLEDs is simulated by using the commercial Light Tools 8.4 software based on the Monte Carlo ray-tracing method (Fig. 1). In this physical model of RP-WLEDs, the component parameters of the remote-packaging WLEDs are shown below:

1) The reflector has an 8 mm bottom length, a 2.07 mm height, and a 9.85 mm length.

2) The remote phosphor layer with a fixed thickness of 0.08 mm covers the four LEDs chips.

3) The LEDs chip with a 1.14 mm square base and a 0.15 mm height. The radiant flux of each blue chip is 1.16 W at wavelength 455 nm [24-28].

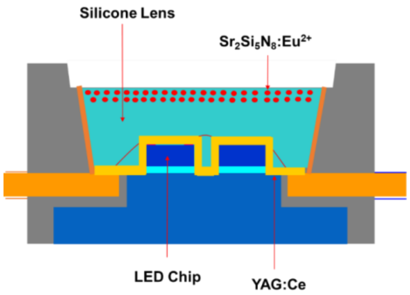

Fig. 1. The RP-WLEDs physical structure in Light Tools 8.4 software.

In this section, the optical properties (In term of D-CCT, CRI, and lumen output) of the remote-packaging WLEDs by co-doping the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particles into the phosphor packaging are details demonstrated by using the Light Tools 8.4 software. For investigating the effect of the red-phosphor particle's concentration on the optical properties of WLEDs, we vary the concentration of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particle continuously from 2% to 20%. Furthermore, the refractive indexes of the red-emitting and yellow-emitting phosphors in the Light Tools software are chosen at 1.80 and 1.83, respectively. The average radius of the phosphor particles are 7.25 μm and the refractive index of the silicone glue was chosen 1.5 [24-28].

By applying Mie theory [21], the scattering coefficient $\mu_{sca}(\lambda)$, anisotropy factor $g(\lambda)$, and reduced scattering coefficient $\delta_{sca}(\lambda)$ can be computed by the below expressions (1), (2), and (3):

$$\mu_{sca}(\lambda) = \int N(r) C_{sca}(\lambda, r) dr \tag{1}$$

$$g(\lambda) = 2\pi \int \int_{-1}^{1} p(\theta, \lambda, r) f(r) \cos\theta \, d\cos\theta \, dr \tag{2}$$

$$\delta_{sca} = \mu_{sca}(1 - g) \tag{3}$$

In these equations, $N(r)$ indicates the distribution density of diffusional particles (mm^3). C_{sca} is the scattering cross sections (mm^2), $p(\theta,\lambda,r)$ is the phase function, λ is the light wavelength (nm), r is the radius of diffusional particles (μm), and θ is the scattering angle (°), and $f(r))$ is the size distribution function of the diffuser in the phosphorous layer. Moreover, $f(r)$ and $N(r)$ can be calculated by:

$$f(r) = f_{dif}(r) + f_{phos}(r) \tag{4}$$

$$N(r) = N_{dif}(r) + N_{phos}(r) = K_N.[f_{dif}(r) + f_{phos}(r)] \tag{5}$$

$N(r)$ is composed of the diffusive particle number density $N_{dif}(r)$ and the phosphor particle number density $N_{phos}(r)$. In these equations, $f_{dif}(r)$ and $f_{phos}(r)$ are the size distribution function data of the diffusor and phosphor particle. Here K_N is the number of the unit diffusor for one diffuser concentration and can be calculated by the following equation:

$$c = K_N \int M(r) dr \tag{6}$$

Where $M(r)$ *is* the mass distribution of the unit diffuser and can be proposed by the below equation:

$$M(r) = \frac{4}{3}\pi r^3 [\rho_{dif} f_{dif}(r) + \rho_{phos} f_{phos}(r)] \tag{7}$$

Here $\rho_{diff}(r)$ and $\rho_{phos}(r)$ are the density of diffuser and phosphor crystal.

In Mie theory, C_{sca} can be obtained by the following expression:

$$C_{sca} = \frac{2\pi}{k^2}\sum_{0}^{\infty}(2n-1)(|a_n|^2 + |b_n|^2) \tag{8}$$

where $k = 2\pi/\lambda$, and a_n and b_n are calculated by:

$$a_n(x,m) = \frac{\psi_n^{'}(mx)\psi_n(x) - \mathrm{m}\psi_n(mx)\psi_n^{'}(x)}{\psi_n^{'}(mx)\xi_n(x) - \mathrm{m}\psi_n(mx)\xi_n^{'}(x)} \tag{9}$$

$$b_n(x,m) = \frac{m\psi_n^{'}(mx)\psi_n(x) - \psi_n(mx)\psi_n^{'}(x)}{m\psi_n^{'}(mx)\xi_n(x) - \psi_n(mx)\xi_n^{'}(x)} \tag{10}$$

Where $x = k.r$, m is the refractive index, and $\psi_n(x)$ and $\xi_n(x)$ are the Riccati - Bessel function.

In RP-WLEDs, the relative refractive indices of diffusor (m_{dif}) and phosphor (m_{phos}) in the silicone can be calculated by $m_{dif} = n_{dif} / n_{sil}$ and $m_{phos} = n_{phos} / n_{sil}$. Then the phase function $p(\theta,\lambda,r)$ can be expressed by the below equation:

$$p(\theta,\lambda,r) = \frac{4\pi\beta(\theta,\lambda,r)}{k^2 C_{sca}(\lambda,r)} \tag{11}$$

Where $\beta(\theta,\lambda,r)$ $S_1(\theta)$ and $S_2(\theta)$ are calculated by below equations:

$$\beta(\theta,\lambda,r) = \frac{1}{2}[|S_1(\theta)|^2 + |S_2(\theta)|^2] \tag{12}$$

$$S_1 = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\begin{bmatrix} a_n(x,m)\pi_n(\cos\theta) \\ +b_n(x,m)\tau_n(\cos\theta) \end{bmatrix} \tag{13}$$

$$S_2 = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\begin{bmatrix} a_n(x,m)\tau_n(\cos\theta) \\ +b_n(x,m)\pi_n(\cos\theta) \end{bmatrix} \tag{14}$$

In equations (13) and (14), τ_n and π_n are the angular dependent functions [24-28]

3. Results and discussion

First of all, the scattering coefficients (SC) has a massive increase with increasing red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particle's concentration. It bases on the fact that the white-light quality can be enhanced by controlling red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particle's concentration. The scattering effects of both red-emitting $Sr_2Si_5N_8:Eu^{2+}$ and yellow-emitting particles are massively influenced on the remote-packaging WLEDs. The red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor has a higher absorption ability for the blue light from WLEDs. Moreover, the domination of emitted red light could be done for compensating red-light in the remote-packaging WLEDs.

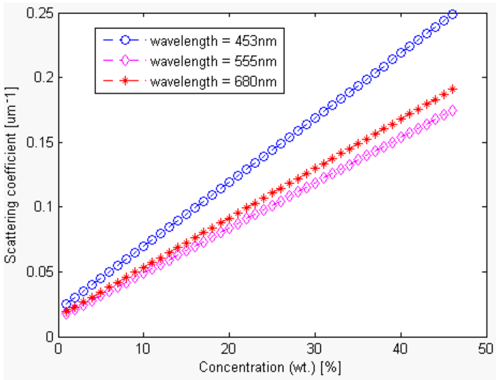

Fig. 2. Scattering coefficients of Sr2Si5N8:Eu2+ in proportion to wavelengths of 453nm, 555nm, and 680nm.

Fig. 3 shows that the reduced scattering coefficient (RSC) of red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor with wavelengths 453nm, 555nm and 680nm are the same with each other. It can be observed that the scattering stability of red-emitting $Sr_2Si_5N_8:Eu^{2+}$ is enough for controlling the optical properties of the remote-packaging WLEDs.

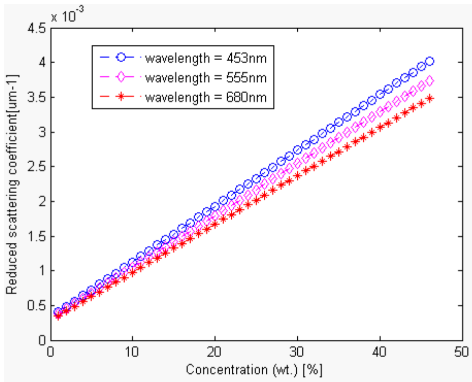

Fig. 3. Reduced scattering coefficient of Sr2Si5N8:Eu2+ with wavelengths of 453nm, 555nm, and 680nm.

By using commercial program Light Tools 8.4 and varying concentration red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particles from 2% to 20%, the D-CCT of the 8500K remote-packaging WLEDs are presented in Fig. 4. After that, Fig. 5 plots the effect of red-emitting phosphor particle concentration on CRI. From these results, CRI remarkably increased in connection with the concentration of red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor rose continuously from 2% to 20%. The maximum value of CRI was about 77 with 20% red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor concentration. As shown in Fig. 4, the D-CCT of W-LEDs decreased significantly by increasing concentration of red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particles and got the minimum value at 12-20% red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor. The effect of the red-emitting phosphor particle's concentration on the luminous efficacy of the 8500K remote-packaging WLEDs is illustrated in Fig. 6. Fig. 6 showed that the luminous efficacy increased crucially while the concentration of red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor grown from 2% to 14 %. After the optimal value, the lumen output had a considerable decrease with 15% to 20 % red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor. The maximum value of the lumen output reaches at 13% to 15 % red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor.

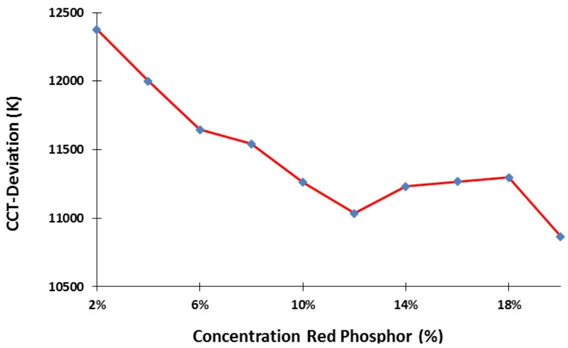

Fig. 4. The D-CCT of the remote-packaging MCW-LEDs by adding red-emitting Sr2Si5N8:Eu2+ phosphor particles.

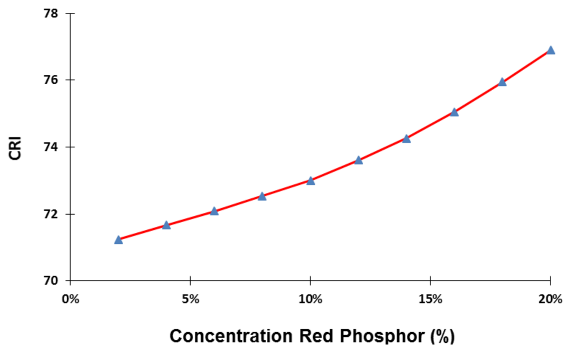

Fig. 5. CRI of the remote-packaging MCW-LEDs grow by adding Red-emitting Sr2Si5N8:Eu2+ phosphor particles.

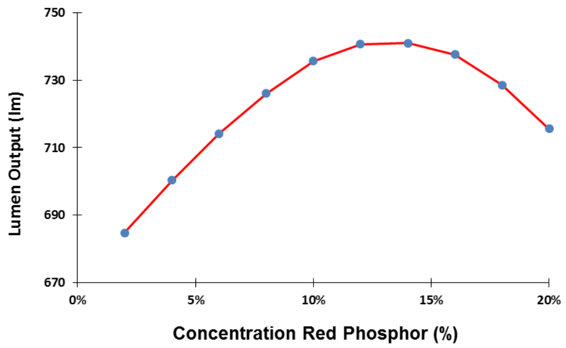

Fig. 6. Lumen output of the remote-packaging MCW-LEDs grow by adding Red-emitting Sr2Si5N8:Eu2+ phosphor particles.

4. Conclusions

In this research, we investigate the effect of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particle's concentration on the optical properties (in term of D-CCT, CRI and lumen output) of the 8500K remote-packaging WLEDs. From results and theory analysis, some conclusions are drawn:

D-CCT remarkably decreased with upping concentration of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor particles. CRI crucially increased while the concentration of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor rose continuously from 2% to 20%. The highest value of CRI was 77 with 20% $Sr_2Si_5N_8:Eu^{2+}$ phosphor concentration. In the beginning, the lumen output had a considerable increase and then decrease in the end. The lumen output had a massive decrease with 15% to 20 % $Sr_2Si_5N_8:Eu^{2+}$ phosphor. The highest value of the lumen output can reach 13% to 15 % $Sr_2Si_5N_8:Eu^{2+}$ phosphor.

This study provided a potential technical recommendation for W-LEDs manufacturing and materials development for W-LEDs applications.

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