Bi-layers Red-emitting Sr₂Si₅N₈:Eu²⁺ Phosphor and Yellow-emitting YAG:Ce Phosphor: A New Approach for Improving the Color Rendering Index of the Remote Phosphor Packaging WLEDs

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Due to optimal advances such as chromatic performance, durability, low power consumption, high efficiency, long-lifetime, and excellent environmental friendliness, white LEDs (WLEDs) are widely used in vehicle front lighting, backlighting, decorative lighting, street lighting, and even general lighting. In this paper, the remote packaging WLEDs (RP-WLEDs) with bi-layer red-emitting Sr₂Si₅N₈: Eu²⁺ and yellowemitting YAG:Ce phosphor was proposed and investigated. The simulation results based on the MATLAB software and the commercial software Light Tools indicated that the color rendering index (CRI) of bi-layer phosphor RP-WLEDs had a significant increase. The CRI had a considerable increase from 72 to 94. In conclusion, the results showed that bi-layer red-emitting Sr₂Si₅N₈:Eu²⁺ and yellow-emitting YAG:Ce phosphor could be a prospective approach for manufacturing RP-WLEDs with enhanced optical properties.

Keywords: Remote phosphor, Bi-layer phosphor, Color rendering index, White light emitting diodes (WLEDs)

OCIS codes: (160.4670) Optical materials; (250.0250) Optoelectronics; (160.2100) Electro-optical materials

I. INTRODUCTION

Due to optimal advances such as chromatic performance, durability, low power consumption, high efficiency, longlifetime high, and environmentally friendliness, white LEDs (WLEDs) are widely used in vehicle front lighting, backlighting, decorative lighting, street lighting, and even general lighting. Among the important approaches to generate white light based on the blue LED chips, phosphor-converting WLEDs is the most popular approach because of low cost and simple technology. In phosphor-converting WLEDs, the phosphor particles absorb the blue light from the blue LED chip and then convert the absorbed blue light into yellow emission. Then, the blue and yellow light mix and generate white light based on phosphor-converting processes, light absorption, light scattering and light conversion. To date, there are many research works focused on phosphorconverting processes to improve optical properties of WLEDs. Some researchers tried to control the optical properties of WLEDs by optimization of phosphor thickness, concentration, particle size, geometry, amount, and arrangement. Results showed that these factors play significant roles in improving the optical properties based on reducing the light trapping in WLEDs [1]. By another way, some research just concentrated on doping green or red phosphor on the phosphor compounding to improve lighting performance of WLEDs. Moreover, in [2] TiO2 was used as a dopant in phosphor compounding to enhancing lighting performance of WLEDs, in [3, 4] SiO2 was used for a similar purpose. With these directions, some kinds of phosphor structures were used such as conformal, in-cup, dome, and remote phosphor. The results indicated that the remote phosphor structure has critical advances in comparison with others structures. The remote phosphor structure of

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W-LEDs (RP-WLEDs) 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 thus improve the phosphor efficiency. With these advances, the RP-WLEDs seems to be a prospective solution for manufactured WLEDs [5-12]. However, to date, very rapid work has improved and demonstrated the optical properties of W-LEDs with remote phosphor structure by bi-layer red and yellow phosphor. It is the remaining gap, which could be entirely filled by this proposed research work.

In last twenties years, 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 LED technology. Among these phosphors, $Sr_2Si_5N_8$: Eu^{2+} presented excellent emission characteristics under a blue excitation wavelength of 450 nm, had a uniform particle size distribution and showed high performance in LED packages [13-16]. Besides, with the advantages of excellent thermal and chemical stability, $Sr_2Si_5N_8$: Eu^{2+} could be employed for compensating red light, resulting in increasing the color quality of LED lamps. However, up to now, there are too few previous studies which employ $Sr_2Si_5N_8$: Eu^{2+} for RP-WLEDs.

In this work, the effect of bi-layer red-emitting Sr₂Si₅-N₈:Eu²⁺ phosphor and yellow-emitting YAG:Ce phosphor on the CRI of WLEDs was investigated. In this structure, the upper layer is red-emitting Sr₂Si₅N₈:Eu²⁺ phosphor and the lower is yellow-emitting YAG:Ce phosphor. To make this method convincing, we proposed the physical simulation model and mathematical analysis of WLEDs as the first step. After that, the optical properties of WLEDs (CRI and luminous efficiency) were calculated, analyzed and demonstrated in detail by using the MATLAB software and the commercial software Light Tools. In the simulation process. the concentration of yellow-emitting YAG:Ce phosphor was fixed at 15 % while the concentration of the red one varied from 0% to 9%. The simulation results indicated that the optical properties of WLEDs were critically influenced by the bi-layer phosphor structure and the concentration of the red-emitting Sr₂Si₅N₈:Eu²⁺ phosphor. Finally, bi-layer red-emitting Sr₂Si₅N₈:Eu²⁺ phosphor and yellow-emitting YAG:Ce phosphor could be considered as a prospective approach for manufacturing WLEDs with high optical properties.

II. SIMULATION MODEL AND MATHEMATICAL ANALYSIS

In this paper, the real 7000 K RP-WLED with 9 LED chips was used to simulate the physical model by the commercial LightTools 8.1.0 software (Fig. 1). The LightTools 8.1.0 based on the Monte Carlo ray-tracing method. The real RP-WLEDs have some features:

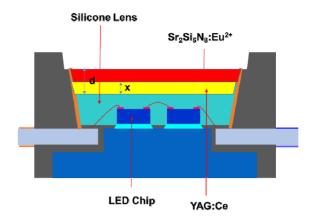


FIG. 1. The physical structure of the RP-WLEDs.

- Each blue LED chip with a 1.14 mm square base and a 0.15 mm height is bonded in the reflector. The power of each blue chip is 1.16 W.
- The reflector has a bottom length of 8 mm, a height of 2.07 mm and a length of 9.85 mm.
- The remote phosphor compounding has the fixed thickness 0.08 mm, which covers the nine chips.

The refractive indexes of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ and YAG:Ce phosphors are 1.93 and 1.83, respectively. The silicone glue has the refractive index of 1.5. The average radius of two types of phosphors is chosen as 7.25 μ m.

The mathematical description could be demonstrated by MATLAB software using Mie-scattering theory [17]. The scattering coefficient $\mu_{sca}(\lambda)$, anisotropy factor $g(\lambda)$, and reduced scattering coefficient $\delta_{sca}(\lambda)$ are calculated by expressions (1), (2), and (3):

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

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

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

where N(r) is the number density distribution of diffusional particles (per cubic millimeter), C_{sca} is the scattering cross sections (per square millimeter), $p(\theta, \lambda, r)$ is the phase function, λ is the wavelength of the incident light (nanometers), r is the radius of particles (micrometers), θ is the scattering angle (degrees), and f(r) is the size distribution function of the diffusers in the phosphor layer.

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

$$N(r) = N_{dif}(r) + N_{phos}(r)$$

$$= K_N \cdot [f_{dif}(r) + f_{phos}(r)]$$
(5)

N(r) is composed of the diffusive particle number density $N_{dij}(r)$ and the phosphor particle number density $N_{phos}(r)$. $f_{dij}(r)$ and $f_{phos}(r)$ are the size distribution function data of the diffusor and phosphor particle. If the phosphor concentration c (milligrams per cubic millimeter) of the mixture is known, K_N denotes the number of the unit diffusor for one diffuser concentration and K_N can be obtained by:

$$c = K_N \int M(r) dr. (6)$$

To obtain K_N , we should first know the mass distribution M(r) (milligrams) of the unit diffusor. The equation below can calculate M \mathbb{R} :

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

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

In Mie theory, C_{sca} is normally presented:

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

where k is the wavenumber $(2\pi/\lambda)$, and a_n and b_n are the expansion coefficients with even symmetry and odd symmetry, respectively. These coefficients can be calculated by equations below:

$$a_{n}(x,m) = \frac{\psi_{n}(mx)\psi_{n}(x) - m\psi_{n}(mx)\psi_{n}(x)}{\psi_{n}(mx)\xi_{n}(x) - m\psi_{n}(mx)\xi_{n}(x)},$$
(9)

$$a_{n}(x,m) = \frac{m\psi_{n}(mx)\psi_{n}(x) - \psi_{n}(mx)\psi_{n}(x)}{m\psi_{n}(mx)\xi_{n}(x) - \psi_{n}(mx)\xi_{n}(x)},$$
(10)

where x is the size parameter (=k,r), m is the refractive index of the diffusive scattering particles, $\psi_n(x)$, $\xi_n(x)$ are the Riccati - Bessel functions.

For small spheres, the phase function $p(\theta, \lambda, r)$ can be calculated according to the following equation) [17]:

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

where $\beta(\theta, \lambda, r)$ is the dimensionless scattering function, which is obtained by the scattering amplitude functions S_1 and S_7 :

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

$$S_{1} = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \begin{bmatrix} a_{n}(x,m)\pi_{n}(\cos\theta) \\ +b_{n}(x,m)\tau_{n}(\cos\theta) \end{bmatrix}, \tag{13}$$

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

In Eqs. (13) and (14), the angular dependent functions $\pi_n(\cos\theta)$ and $\tau_n(\cos\theta)$ are expressed in the angular scattering patterns of the spherical harmonics [18-22].

III. RESULTS AND DISCUSSION

The scattering, reduced scattering, and backscattering of light inside two phosphor layers are calculated and demonstrated using MATLAB software as shown in Figs. 2 and 3. In this calculation, the concentration of the red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor was varied from 2% to 9 % while the yellow YAG:Ce phosphor concentration was fixed at 15%. From Figs. 2 and 3, it can be shown that scattering and reduced scattering coefficients of blue light (λ = 455 nm), and yellow light (λ = 595 nm) had a rapid rise. Also, the scattering coefficient and the reduced scattering coefficient of blue light increase higher than yellow in both red and yellow phosphor layers. However, the backscattering coefficient of wavelength 455 nm in the red phosphor layer is much greater than for the wavelength 595 nm in the yellow phosphor layer (Fig. 4).

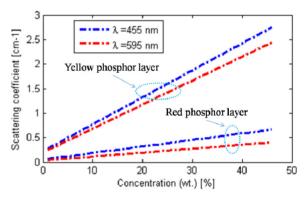


FIG. 2. Scattering coefficient of two phosphor layers of 455 nm, 595 nm wavelengths.

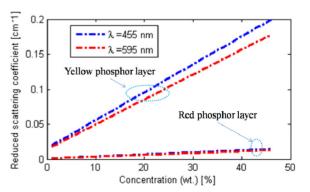


FIG. 3. Reduced scattering coefficient of two phosphor layers of 455 nm, 595 nm wavelengths.

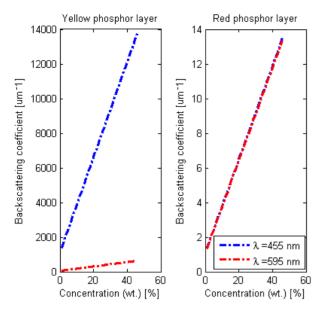


FIG. 4. Backscattering coefficient of two phosphor layers of 455 nm, 595 nm wavelengths.

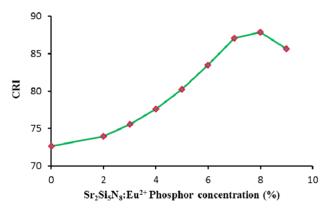


FIG. 5. The CRI of two phosphor layers RP-WLEDs.

The results from the theory-based calculation and optical ray-tracing spectra with LightTools agreed with each other well. With the increase of red phosphor concentrations, the CRI moved up (Fig. 5). In this situation, the CRI had a considerable increase from 72 to 94. This result indicated that for two layers LED packages the chromaticity of LED can be efficiently adjusted by the ratio of blue to yellow and red components since the yellow phosphor does not absorb the backscattering of red emission from the upper red phosphor layer. This is considered to be because the emission energy loss is associated with re-absorption process by different phosphor emission spectrum within the LED packages.

IV. CONCLUSION

In this paper, RP-WLEDs with bi-layer red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor and yellow-emitting YAG:Ce phosphor

are proposed and investigated under different concentrations of red-emitting Sr₂Si₅N₈:Eu²⁺ phosphor. It is found that the color rendering index (CRI) of bi-layer phosphor RP-WLEDs had a significant increase from 72 to 94. These results provide a prospective practical solution for manufacturing bi-layered RP-WLEDs. In future works, the influence of bi-layer red-emitting Sr₂Si₅N₈:Eu²⁺ phosphor and yellow-emitting YAG:Ce phosphor on the lighting performance of RP-WLEDs will be more developed.

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REFERENCES

- S. C. Allen and A. J. Steckl, "A nearly ideal phosphorconverted white light-emitted diode," J. Appl. Phys. 92, 143309-143311 (2008).
- H. Masui, S. Nakamura, and S. P. Denbaars, "Effects of phosphor application geometry on white light-emitting diodes," Jpn. J. Appl. Phys. 45(34), 910-912 (2006).
- 3. N. D. Q. Anh, M. F. Lai, H. Y. Ma, H. Y. Lee, "Enhancing of correlated color temperature uniformity for multichip white light LEDs by adding SiO₂ to phosphor layer," J. Chin. Inst. Eng. **38**(3), 297-303 (2015).
- N. D. Q. Anh and H. Y. Lee. "Improving the angular color uniformity and the lumen output for white LED lamps by green Ce0.67 Tb0.33 MgAl11 O19:Ce,Tb. Phosphor," J. Chin. Inst. Eng. 40, 871-875 (2016).
- M. H. Crawford, "LEDs for solid-state lighting: performance challenges and recent advances," IEEE J. Sel. Topics Quantum Electron. 15, 1028-1040 (2009).
- H. Run, L. Xiaobing, F. Han, and L. Sheng, "Effect of phosphor settling on the optical performance of phosphorconverted white light-emitting diode," J. Lumin. 132(5), 1252-1256 (2012).
- H. Zheng, X. Luo, R. Hu, B. Cao, X. Fu, Y. Wang, and S. Liu, "Conformal phosphor coating using capillary microchannel for controlling color deviation of phosphor-converted white light-emitting diodes," Opt. Express 20, 5092-5098 (2012).
- 8. R. Hu, X. Luo, and S. Liu, "Study on the optical properties of conformal coating light-emitting diode by monte carlo simulation," IEEE Photon. Technol. Lett. **23**, 1673-1675 (2011).
- I. Zhmakin, "Enhancement of light extraction from light emitting diodes," Phys. Rep. 498, 189-241 (2011).
- Z. Y. Liu, S. Liu, K. Wang, and X. B. Luo, "Studies on optical consistency of white LEDs affected by phosphor thickness and concentration using optical simulation," IEEE Trans. Compon. Packag. Tech. 33, 680-687 (2010).
- 11. R. Yu, S. Jin, S. Cen, and P. Liang, "Effect of the phosphor

- geometry on the luminous flux of phosphor-converted light-emitting diodes," IEEE Photon. Tech. Lett. **22**, 1765-1767 (2010).
- 12. Z. Y. Liu, C. Li, B. H. Yu, Y. H. Wang, and H. B. Niu, "Effects of YAG:Ce phosphor particle size on luminous flux and angular color uniformity of phosphor-converted white LEDs," J. Disp. Technol. **8**, 329-335 (2012).
- 13. R. Hu, B. Cao, Y. Zou, Y. Zhu, S. Liu, and X. Luo, "Modeling the light extraction efficiency of bi-layer phosphors in white LEDs," IEEE Photon. Tech. Lett. **25**, 1141-1144 (2013).
- V. D. Luong, W. Zhang, and H. Lee, "Preparation of Sr₂Si₅N₈:Eu²⁺ for white light-emitting diodes by multi-step heat treatment," J. Alloys Compd. 509, 7525-7528 (2011).
- X. Piao, T. Horikawa, H. Hanzawa, and K. Machida, "Characterization and luminescence properties of Sr₂Si₅N₈:Eu²⁺ phosphor for white light-emitting-diode illumination," Appl. Phys. Lett. 88, 161908 (2006).
- 16. H. Nersisyan, H. I. Won, and C. W. Won, "Highly effective synthesis and photoluminescence of Sr₂Si₅N₈:Eu²⁺ red-emitting phosphor for LEDs," Chem. Commun. **47**(43), 11897 (2011).
- 17. B. Zhang, J.-W. Wang, L.-Y. Hao, X. Xu, S. Agathopoulos,

- L.-J. Yin, C.-M. Wang, and H. T. Hintzen, "Highly stable red-emitting $Sr_2Si_5N_8:Eu^{2+}$ phosphor with a hydrophobic surface," J. Am. Ceram. Soc. **100**(1) 257-264 (2016).
- J. Zhong, M. Xie, Z. Ou, R. Zhang, M. Huang, and F. Zhao, "Mie theory simulation of the effect on light extraction by 2-D nanostructure fabrication," in *Proc. Symposium on Photonics and Optoelectronics (SOPO)*, IEEE (2011).
- 19. R. Hu, B. Cao, Y. Zou, Y. Zhu, and X. Luo, "Modeling the light extraction efficiency of bi-layer phosphors in white LEDs," IEEE Photon. Tech. Lett. **25**, 1141-1144 (2013).
- C. H. Chiang, S. J. Gong, T. S. Zhan, and S. Y. Chu, "White light-emitting diodes with high color rendering index and tunable color temperature fabricated using separated phosphor layer structure," IEEE Electron Device Lett. 37(7), 898-901 (2016).
- Y. Zhu and N. Narendran, "Investigation of remote-phosphor white light-emitting diodes with multi-phosphor layers," Jpn. J. Appl. Phys. 49, 100203 (2010).
- W. M. Yen and M. J. Weber, "Inorganic phosphors: compositions, preparation and optical properties," Washington, D. C. CRC Press (2004).