Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor: a novel recommendation for improving the lighting performance of the 7000 K remote-packaging white LEDs

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Abstract. In the last few years, white LED lamps (WLEDs) have been popularly used in general lighting because of some excellent properties, such as fast response time, environment friendliness, small size, long lifetime, and high efficiency. In this research, we propose a novel recommendation for improving the lighting performance (in terms of the colour rending index, colour quality scale, and luminous efficiency) of the 7000 K remote-packaging WLEDs by adding the red-emitting Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor to the yellow-emitting YAG:Ce phosphor compound. In the first stage, we use MATLAB to investigate the light scattering process based on Mie Theory. After that, we use the Light Tool software to simulate and demonstrate this process. Finally, the simulation results are verified with analytical analysis, which clearly shows that the lighting performance of the 7000 K remote-packaging WLEDs significantly depended on the Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ concentration. The results provided a potential practical solution for manufacturing remote phosphor WLEDs in the near future.

Key words: remote-packaging WLEDs, red-emitting Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor, colour rendering index, colour uniformity, lumen output.

1. INTRODUCTION

Nowadays, white light-emitting diodes (WLEDs) are considered as the latest type of solid-state lighting, which have unique advantages in comparison with other eight sources [1–3]. Being considered as the fourth generation of light sources, WLEDs have the following advantages: environment friendliness, wide operation temperature, long lifetime, wide range of colour temperatures, energy saving compared to other light sources, and quick startup [1–4]. Commonly, there are two primary ways of producing high-intensity white light using LEDs. One is to use individual LEDs that emit three primary colours – red, green, and blue – and then mix them all to form white light. The other is to use a phosphor material to convert a monochromatic light from a blue or UV LED to a broad-spectrum white light, much in the same way as a fluorescent light bulb works [1–4]. The second method has had a huge advance and is widely used in the public lighting due to the low cost and simple technology of production and use. Furthermore, the blue light from the blue LEDs chip
should be converted into white light by packaging for the second method. From this point of view, the LEDs packaging is considered as an important research direction for improving the lighting performance of WLEDs. A packaging method in LEDs not only can ensure better performance of LED devices by enhancing their reliability and optical characteristics but can also realize control and adjustment of the final lighting performance [4–6]. In the last decade, many papers have focused on improving the lighting performance of WLEDs by improving the packaging method. In more details, lighting properties of WLEDs were enhanced by Sr1-xBaxSi2O2N2:Eu2+(0 x 1) [7], β-SiAlON:Yb2+ phosphor [8], novel packaging structures [9], Li2SrSiO4:Eu3+, Sm3+ [10], or by adding red or green phosphor [11, 12] into the phosphor layer of WLEDs. We can see that this research direction is too hot today.

On another side, the remote-packaging WLEDs are a structure in which the phosphor is moved far away from the LED chip. This structure could significantly reduce the probability of the absorption of the re-emitted light by the WLEDs chip, and thus improve the phosphor efficiency. With these advances, the remote phosphor structure of WLEDs seems to be a solution for the manufacture of WLEDs [13–15]. However, very rare works have improved and demonstrated the lighting performance of WLEDs with a remote phosphor structure by mixing two or more diffusive particles into the phosphor compound. It is the remaining gap that could be filled by this research work.

In the last decade, SiN4-based covalent nitride materials such as M2Si5N8:Eu2+ and MA(Si)N3:Eu2+ (M = Ca, Sr, Ba) have been extensively considered as excellent materials for the LEDs technology. Among these phosphors, Sr2Si5N8:Eu2+ showed excellent emission characteristics under a blue excitation wavelength of 450 nm, had a uniform particle size distribution, and high performance in LED packages [16–18].

In this research, Sr2Si5N8:Eu2+ particles were remotely added into the yellow-emitting YAG:Ce phosphor of the remote-packaging WLEDs in order to improve the lighting performance of WLEDs. Based on Light Tools and MATLAB software, a favourable influence of Sr2Si5N8:Eu2+ on the lighting performance of the remote-packaging WLEDs is suggested, investigated, and demonstrated. The following can be considered as the main contributions of this paper:

1. The 7000 K remote-packaging WLEDs are proposed and simulated by Light Tools.
2. The scattering properties of Sr2Si5N8:Eu2+ particles are investigated by MATLAB.
3. The influence of Sr2Si5N8:Eu2+ concentration on the lighting performance (in terms of correlated colour temperature deviation (D-CCT), Colour Rending Index (CRI), Colour Quality Scale (CQS), and the luminous efficacy) is investigated.

The remaining part of this paper is organized as follows. The system model and mathematical description are simulated and proposed in Section 2. Section 3 presents the results and some discussion. Section 4 concludes this paper.

2. SYSTEM MODEL AND MATHEMATICAL DESCRIPTION

In this research, 7000 K remote-packaging WLEDs were simulated by using Light Tools (Fig. 1). This model of remote-packaging WLEDs has the following key parameters:

1. The parameters of the reflector: an 8 mm bottom length, a 2.07 mm height, and a 9.85 mm length.
2. The remote phosphor layer has a fixed thickness of 0.08 mm.
3. The parameters of the LED chip: a 1.14 mm square base and a 0.15 mm height. Each blue chip has a 1.16 W luminous flux [10–11].

In the simulation, we set the Sr2Si5N8:Eu2+ concentration from 2% to 22%. The refractive indexes of Sr2Si5N8:Eu2+, yellow-emitting phosphors, and the silicone glue were chosen as 1.80, 1.83, and 1.50, respectively. The selected average radius of the phosphor particles was at 7.25 μm. The diffusional particle density was varied to fix the average correlated colour temperature (CCT) value. When the weight percentage of the diffusers was increased, it was necessary to reduce the weight percentage of YAG:Ce phosphor to maintain the average CCT value [10–11].

![Fig. 1. Physical structure of 7000 K remote-packaging WLEDs.](image)
Applying Mie theory [17,18], we can formulate:

- the scattering coefficient $\mu_{\text{sca}}(\lambda)$:
  \[
  \mu_{\text{sca}}(\lambda) = \int N(r) C_{\text{sca}}(\lambda, r) dr; \tag{1}
  \]

- the anisotropy factor $g(\lambda)$:
  \[
  g(\lambda) = 2\pi \int_{-1}^{1} p(\theta, \lambda, r) f(r) \cos \theta d \cos \theta dr; \tag{2}
  \]

- the reduced scattering coefficient $\delta_{\text{sca}}(\lambda)$:
  \[
  \delta_{\text{sca}} = \mu_{\text{sca}} (1 - g). \tag{3}
  \]

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

3. RESULTS AND DISCUSSION

The scattering and reduced scattering coefficients of the Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ particles, obtained with using MATLAB software, are presented in Fig. 2 and Fig. 3, respectively. From this analysis, we can see that the scattering coefficients grew with increasing Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ concentration (Fig. 2). This means that the white-light quality can be enhanced by controlling the Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ concentration. The scattering effects of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ particles were significantly influenced on remote-packaging WLEDs (Fig. 2). The Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor has a higher absorption ability for the blue light from LEDs. Therefore, the domination of the emitted red light could be applied for compensating red light in remote packaging WLEDs. Moreover, the reduced scattering coefficients of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ at wavelengths 453 nm, 555 nm, and 680 nm are approximately equal to each other (Fig. 3). This indicates that the scattering stability of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ is useful for controlling the colour quality of the remote packaging WLEDs.

The $\Delta$CCT of the 7000 K remote-packaging WLEDs at different concentrations of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor particles (2–22%) was determined by using Light Tools (Fig. 4). The influence of particle concentration on the colour rendering index (CRI) and the colour quality scale (CQS) is illustrated in Fig. 5 and Fig. 6, respectively. As we can see, the CRI increased from 68 to 75 and the CQS from 64 to 68 while the concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ rose continuously from 2% to 22%.

![Fig. 2. Scattering coefficients of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor.](image)

![Fig. 3. Reduced scattering coefficient of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor.](image)

![Fig. 4. Colour–temperature (CCT) deviation of the remote-packaging WLEDs at adding red-emitting Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor particles.](image)
The highest values of the CRI and CQS were respectively about 75 and 68 at the 20% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor concentration. The D-CCT of WLEDs decreased significantly, from 6600 K to 5700 K, at the increasing concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor particles and reached the lowest value at the 12–22% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor concentration (Fig. 4). The decrease of the D-CCT at the increasing concentration of the Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ particles could be caused by the overscattering properties of light from the blue chips in the phosphor layer. There are maximums and valleys on the trendline because the scattering process in the phosphor layer is a random process and may be caused by some significant changes in some simulation point in the trendline. The lighting performance of the 7000 K remote-packaging WLEDs is influenced by each light (from the blue chips) scattering process in the phosphor compounding.

The influence of the red-emitting phosphor concentration on the luminous efficacy of the 7000 K WLEDs is presented in Fig. 7. The figure shows that the luminous efficacy increased significantly from 600 lm to 680 lm while the concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor was increased from 2% to 16%. After that, the lumen output had a considerable decrease to 660 lm as the concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor increased to 18–20%. The lumen output can reach the highest value at 16% to 18% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor. This effect was caused by the overscattering and backscattering process in WLEDs when the concentration of the red-emitting phosphor was higher than 18%. The overscattering process was linked to the lost energy of the light and the decrease of the luminous efficacy of the WLEDs.

4. CONCLUSIONS

The use of the red-emitting Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor particles to improve the lighting performance of 7000 K remote-packaging WLEDs was suggested and its influence was analysed. From the results and theoretical analysis, some conclusions can be drawn:

(1) Colour uniformity (CCT deviation) significantly decreased (from 6600 K to 5700 K) with the rising concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor particles.

(2) The values of CRI and CQS increased from 68 to 75 and from 64 to 68, respectively, while the concentration of Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor rose continuously from 2% to 22%. The highest values of CRI and CQS (75 and 68) were at 22% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor concentration.

(3) The lumen output had an increase from 600 lm to 680 lm, then a decrease to 660 lm after the optimal value. The lumen output had a massive decrease of 18–20% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor. The highest value of the lumen output was recorded at 16% to 18% Sr$_2$Si$_5$N$_8$:Eu$^{2+}$ phosphor.
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REFERENCES

1. Liu, S. and Luo, X. \textit{LED Packaging for Lighting Applications: Design, Manufacturing and Testing}. John Wiley & Sons, 2011.
2. Gibney, E. Nobel for blue LED that revolutionized lighting. \textit{Nature}, 2014, \textbf{514}, 152–153.
3. Paisnik, K., Rang, G., and Rang, T. Life-time characterization of LEDs. \textit{Estonian J. Eng.}, 2011, \textbf{17}, 241–251.
4. Luo, X. and Hu, R. Chip packaging: encapsulation of nitride LEDs. In \textit{Nitr id Semiconductor Light-Emitting Diodes ( LEDs )}, 2014, 441–481.
5. Luo, X., Hu, R., Liu, S., and Wang, K. Heat and fluid flow in high-power LED packaging and applications. \textit{Prog. Energy Combust. Sci.}, 2016, \textbf{56}, 1–32.
6. Paisnik, K., Poppe, A., Rang, T., and Rang, G. Physics related modeling of power LEDs. In \textit{13th Biennial Baltic Electronics Conference}, 2012.
7. Schubert, E. F. Solid-state light sources getting smart. \textit{Science}, 2005, \textbf{308}, 1274–1278.
8. Seibald, M., Rosenthal, T., Oeckler, O., and Schnick, W. Highly efficient pc-LED phosphors Sr\textsubscript{1-x}Ba\textsubscript{x}Si\textsubscript{2}O\textsubscript{2}N\textsubscript{2}:Eu\textsuperscript{2+} (0 \leq x \leq 1) – crystal structures and luminescence properties revisited. \textit{C r i t. Rev. Solid State Mater. Sci.}, 2014, \textbf{39}, 215–229.
9. Shuai, Y., He, Y., Tran, N. T., and Shi, F. G. Angular CCT uniformity of phosphor converted white LEDs: effects of phosphor materials and packaging structures. \textit{IEEE Photon. Techn. L.}, 2011, \textbf{23}, 137–139.
10. Minh, T. H. Q., Nhan, N. H. K., Anh, N. D. Q., and Lee, H-Y. Red-emitting α-SrO·3B\textsubscript{2}O\textsubscript{3}:Sm\textsuperscript{2+} phosphor: an innovative application for increasing color quality and luminous flux of remote phosphor white LEDs. \textit{J. Chin. Inst. Eng.}, 2014, \textbf{40}(4), 313–317.
11. Nhan, N. H. K., Minh, T. H. Q., Nguyen, T. N., and Voznak, M. Co-doping green-emitting Ca\textsubscript{2}F\textsubscript{2}:Ce\textsuperscript{3+}:Tb\textsuperscript{3+} and yellow-emitting phosphor particles for improving the CCT deviation and luminous efficacy of the in-cup phosphor packaging WLEDs. \textit{Dig. J. Nanomater. Bios.}, 2011, \textbf{12}(3), 891–898.
12. Ying, S-P. and Shiu, A-Y. Investigation of remote-phosphor white light-emitting diodes with improved scattered photon extraction structure. \textit{Appl. Opt.}, 2015, \textbf{54}(28), E30–34.
13. Li, C., Rao, H., Zhang, W., Zhou, C., Zhang, Q., and Zhang, K. Self-adaptive conformal–remote phosphor coating of phosphor-converted white light emitting diodes. \textit{J. Displ. Technol.}, 2016, \textbf{12}, 946–950.
14. Kim, J. K., Luo, H., Schubert, E. F., Cho, J., Sone, C., and Park, Y. Strongly enhanced phosphor efficiency in GaInN white light-emitting diodes using remote phosphor configuration and diffuse reflector cup. \textit{Jpn. J. Appl. Phys.}, 2005, \textbf{44}.
15. Luong, V. D., Zhang, W., and Lee, H-R. Chem inform abstract: preparation of Sr\textsubscript{2}Si\textsubscript{5}N\textsubscript{8}: Eu\textsuperscript{2+} for white light-emitting diodes by multi-step heat treatment. \textit{ChemInform}, 2011, \textbf{42}(33).
16. Piao, X., Horikawa, T., Hanzawa, H., and Machida, K-I. Characterization and luminescence properties of Sr\textsubscript{2}Si\textsubscript{5}N\textsubscript{8}: Eu\textsuperscript{2+} phosphor for white light-emitting-diode illumination. \textit{Appl. Phys. Lett.}, 2006, \textbf{88}, 161908.
17. Nersisyan, H., Won, H. I., and Won, C. W. Highly effective synthesis and photoluminescence of Sr\textsubscript{2}Si\textsubscript{5}N\textsubscript{8}: Eu\textsuperscript{2+} red-emitting phosphor for LEDs. \textit{Chem. Commun.}, 2011, \textbf{47}, 11897.
18. Tapaninen, O., Myohanen, P., Majanen, M., Sitomaniemi, A., Ollkonen, J., Hildenbrand, V., et al. Optical and thermal simulation chain for LED package. In \textit{17th International Conference on Thermal, Mechanical and Multi-Physics Simulation and Experiments in Microelectronics and Microsystems (EuroSimE)}, 2016.

\begin{align*}
\text{Sr}_2\text{Si}_3\text{N}_8: \text{Eu}^{2+} \text{ fosfoor: uudne soovitus 7000 K kaugpakendatud valge valgusdioodi (WLED) valgustamisomaduste täiustamiseks} \\
\text{Phu Tran Tin, Nguyen Huu Khanh Nhan, Tran Hoang Quang Minh, Tan N. Nguyen, Miroslav Voznak ja Tran Thanh Trang} \\
\text{Viimastel aastatel on valged valgusdioodivalgust (WLED) üldvalgustistes väga populuaarseks saanud. Põhjuseks on nende valgustite väike ümberlihitusaeg, keskkonnasõbralikkus, vääkised mõõtmed, pikk kasutusiga ja suur kasutegur. Käesolevas uurimises on esitatud uudne soovitus nende dioodivalgustite valgustamisomaduste (värvuse ümberpäigutamise indeksi (CRI), värvuse kvaliteediastmiiku (CQS), valgusjõu efektiivsuse) täiustamiseks rakendatuna 7000 K kaugpakendatud valge valgusdioodile, lisades punast fosfoori Sr\textsubscript{2}Si\textsubscript{5}N\textsubscript{8}: Eu\textsuperscript{2+} kollaselle põhiumendatud YAG:Ce. Soovituse aluseks on programmiga MATLAB tehtud simulatsioonid, kasutades Mic teooriat valguse hajuse ja kohta. Hajumisprotsesside täpsuse kõrvaldamiseks on kasutatud ka programmi Light Tool.}
\end{align*}

On näidatud, et uuritud valge valgusdioodi omadused on parendatavad fosfoori Sr\textsubscript{2}Si\textsubscript{5}N\textsubscript{8}: Eu\textsuperscript{2+} lisamisega ja valgusdioodi valgustamisomadused sõltuvad fosfoori koncentраtsioonist. Soovitus pakub praktilist abi 7000 K kaugpakendatud valge valgusdioodi (WLED) valgustamisomaduste täiustamiseks.