



Received: 05 September 2017  
Accepted: 31 May 2018  
First Published: 11 June 2018

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Reviewing editor:  
Tim Collins, University of Birmingham, UK

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## MATERIALS ENGINEERING | RESEARCH ARTICLE

# Red-emitting $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$ conversion phosphor: A new selection for enhancing the optical performance of the in-cup packaging MCW-LEDs

Phu Tran Tin<sup>1</sup>, Nguyen Huu Khanh Nhan<sup>2</sup>, Tran Hoang Quang Minh<sup>2\*</sup> and Nguyen Thi Phuong Thao<sup>3,4</sup>

**Abstract:** In this research, the influence of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  convention phosphor on the optical performance of the 7,000 K and 7,700 K in-cup packaging multi-chip white LEDs (MCW-LEDs) is investigated. The effect of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  convention phosphor is demonstrated based on Mie Theory by Mat Lab and Light Tools software. The research results indicated that the optical performance of MCW-LEDs was crucially affected by the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor's concentration. This paper provides an essential recommendation for selecting and developing the phosphor materials for MW-LEDs manufacturing.

**Subjects:** Electronics; Optics & Optoelectronics; Optics, Optoelectronic Effects, Devices & Systems; Semiconductors

**Keywords:** red-emitting phosphor; MCW-LEDs; optical performance; CCT deviation; luminous efficiency; color rendering index; color quality scale

### 1. Introduction

Light-emitting diodes (LEDs) are the modest type of solid-state lighting, which have the most significant advancement in the lighting industry in the last few decades. Phosphor-converted LEDs (pcLED), which combine a blue LED chip, and the yellow emitting phosphor is the most common

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### PUBLIC INTEREST STATEMENT

Light-emitting diodes (LEDs) have found their way into many applications in daily life, from warm-white LEDs for energy-efficient lighting to display backlights for smartphones, tablets, and televisions. The current attention to energy-saving and reduction of CO<sub>2</sub> emission in the atmosphere should, therefore, give an additional boost to the development of LEDs for lighting. In this research, we investigated the influence of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  conversion phosphor on the color quality of the 7,000 K, and 7,700 K in-cup packaging multi-chip white LEDs (MCW-LEDs). The research results indicated that the lighting performance of MCW-LEDs was crucially affected by the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  conversion phosphor's size. This research demonstrated a significant recommendation for selecting and developing the phosphor materials for MCW-LEDs manufacturing.

approach to accomplish white light emission through LEDs packaging (“Design of LED,” 2011; Gibney, 2014; Hu, Luo, & Liu, 2011; Luo, Hu, Liu, & Wang, 2016; Winkler, Trinh, Bodrogi, & Khanh, 2015). In the last few decades, there so many research focuses on improving the optical performance of the pLED in packaging direction. Thickness and concentration of phosphor are considered as the main factors in the white LEDs packaging because the luminous flux and color of LEDs are adjusted mainly through changing the phosphor thickness and concentration after the phosphor converters are chosen. Shuai, He, Tran, and Shi (2011) and Tran and Shi (2008) experimentally studied the effects of phosphor thickness and concentration on LEDs luminous flux and correlated color temperature. From the results, higher luminous efficacy are taken with the lower phosphor concentration and higher phosphor thickness (lower trapping efficiency and fewer backs scattering of light). Sommer et al. firstly studied the effects of phosphor thickness, concentration, and size on the spatial color distribution of white LEDs (Sommer et al., 2008). The spatial color uniformity of white LEDs could be improved by changing the phosphor concentration or thickness. An increase of the phosphor thickness or concentration can alter the color from bluish to yellowish, and the growth in the central zone is faster than that in the border area. In Li et al. (2012) and Liu, Liu, Wang, and Luo (2008a, 2008b, 2008c), the effect of phosphor location on the spatial color distribution was investigated. The results showed that to obtain high color uniformity packaging elements should make the blue light and yellow light have a similar radiation pattern. In the last year, some researchers were concentrated on enhancing the optical performance of multi-chip white LEDs (MCW-LEDs) by adding green or red phosphor into the phosphor compounding (Anh, Quoc, & Lee, 2016; Minh, Quang, Nhan, Anh, & Lee, 2017). It can be found that the optical properties of MCW-LEDs can be enhanced clearly. From this point of view, improving the optical performance of MCW-LEDs by adding diffusers into the phosphor compounding is still needed to be investigated. In this research, we try to fill the remaining gap.

In this research, the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor is proposed to enhancing the optical performance of the 7,000 K and 7,700 K in-cup packaging MCW-LEDs. The optical performance of the MCW-LEDs is based on the primary factors like the color uniformity or the correlated color temperature deviation (D-CCT), the color rendering index (CRI), the color quality scale (CQS), and the luminous efficacy (lumen output). This paper is divided into three sections. The physical model of the 7,000 K, 7,700 K in-cup packaging MCW-LEDs and the optical properties of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  conversion phosphor are presented Section 1. In Section 2, the mathematical optical description based on Mie theory of red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor with Mat lab software and the simulation results with the commercial Light Tools software (Synopsys) is collected. Finally, the research results are intensely discussed, analyzed, and investigated in Section 3. Then, some conclusions are proposed. From the research results, we can conclude that the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor crucial influenced the optical performance of the 7,000 K and 7,700 K in-cup packaging MCW-LEDs. The red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor could be considered as a prospective approach for MCW-LEDs improvement shortly.

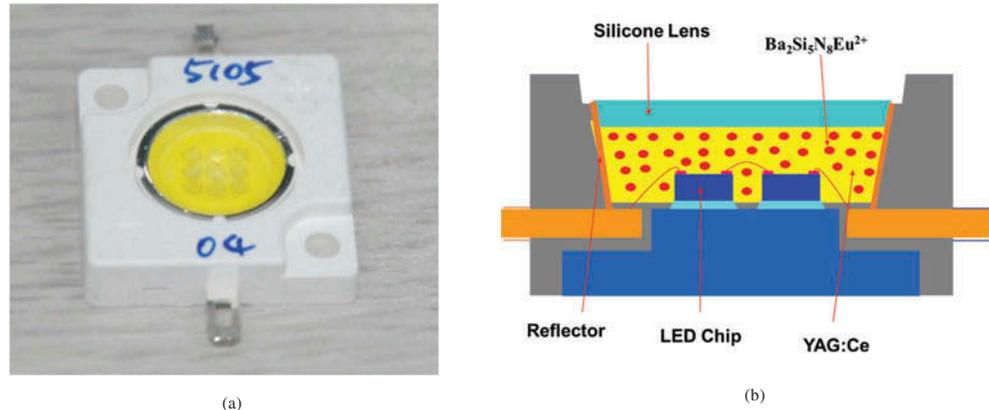
## 2. Research method

### 2.1. The MCW-LEDs physical model

The simulations were carried out using the commercial software package Light Tools. The simulation comprises the set-up of the in-cup packaging MCW-LEDs with average CCT of 7,000 K and 7,700 K. Firstly, to guarantee that our simulation results reflect precisely the impact of our considered parameter (concentration of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor) and are not biased by other factors such as LED’s wavelength, waveform, light intensity, and operating temperature, we use the real-world model of MCW-LEDs (Figure 1(a)). In this simulation stage, we set the main parameters of the in-cup MCW-LEDs as follows:

- (1) The depth, and the inner and outer radius of the reflector to 2.07, 8, and 9.85 mm, respectively.

**Figure 1. (a) The MCW-LED product of the Siliconware Precision Industries Co., Ltd., Taiwan. (b) Illustration of MCW-LEDs with the in-cup phosphor packaging.**



- (2) Nine LED chips are covered with a fixed thickness of 0.08 and 2.07 mm. Each blue chip has a dimension of 1.14 mm × 0.15 mm, the radiant flux of 1.16 W, and the peak wavelength of 453 nm (Figure 1(b)).

In this simulation, the phosphor layer consists of the yellow-emitting YAG:Ce and the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  conversion phosphors particles and the silicone glue, which respectively have the refractive indices of 1.85, 1.93, and 1.50 at room temperature (25°C). Also, the average radius of the yellow-emitting YAG:Ce phosphor particles is set to 7.25  $\mu\text{m}$  for all packages, a value of real particle size. To make the color of MCW-LEDs produce the same when the concentration of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor varies, the yellow-emitting YAG:Ce phosphor concentration should be inversely changed to provide same CCT value. The weight percentage of the MCW-LEDs phosphor layers can be expressed as

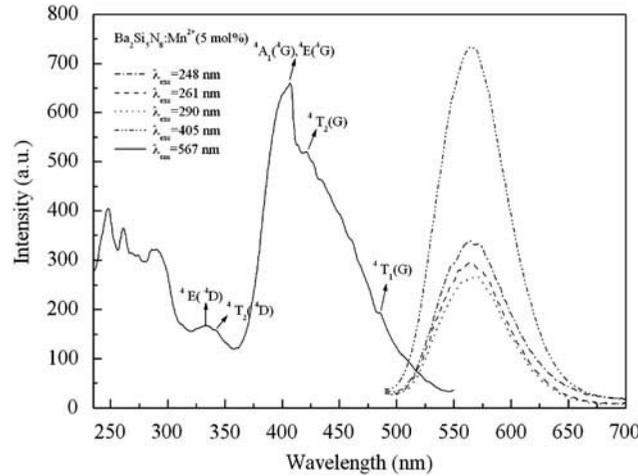
$$\sum W_{\text{pl}} = W_{\text{Yellow phosphor}} + W_{\text{silicone}} + W_{\text{Red phosphor}} = 100\%$$

Here,  $W_{\text{silicone}}$ ,  $W_{\text{Yellow phosphor}}$ , and  $W_{\text{Red phosphor}}$  are in turn the weight percentage of the silicone glue, the yellow-emitting YAG:Ce phosphor, and the red-emitting CeTb phosphors (Anh et al., 2016, 2017; Minh et al., 2017).

## 2.2. The optical properties of the red-emitting phosphor

Recently, the compounds  $\text{M}_2\text{Si}_5\text{N}_8$  (M: Ca, Sr, Ba) have been intensively studied as host lattice in the exploration of LED conversion phosphors. Due to the high covalency and large crystal field effect of nitrogen anion,  $\text{Ce}^{3+}$  and  $\text{Eu}^{2+}$  show long wavelength emission in  $\text{M}_2\text{Si}_5\text{N}_8$  host lattice (M = Ca, Sr, Ba). These rare-earth doped materials have already been demonstrated to be promising conversion phosphors for the white-light LED application (Chen & Xie, 2014; Duan, Otten, Delsing, & Hintzen, 2008; Fukuda, Okada, & Albessard, 2012; Li, Xie, Hirosaki, Takeda, & Zhou, 2009; Li et al., 2006; Zeuner, Schmidt, & Schnick, 2009). Luminescence due to  $\text{Mn}^{2+}$  is known to occur in a lot of inorganic compounds. As for the application of  $\text{Mn}^{2+}$ -doped phosphors in the field of white-light LED, many investigations have been done (Chen & Xie, 2014; Duan et al., 2008; Fukuda et al., 2012; Li et al., 2006, 2009; Zeuner et al., 2009). These phosphors efficiently emit visible light from yellow-green to red range. Also, compared with oxide and sulfide phosphors, they show superior thermal and chemical stability. Among these (oxy) nitride phosphors,  $\text{M}_2\text{Si}_5\text{N}_8:\text{Eu}^{2+}$  (M: Ca, Sr, Ba) shows unusually long wavelength broadband emission between 570 and 680 nm depending on the type of the M ion and  $\text{Eu}^{2+}$  concentration under near-UV to blue light irradiation (370–465 nm) and has high quantum efficiency and very low thermal quenching, which makes it an encouraging yellow-red phosphor for white LEDs to improve the CRI (Figure 2) (Chen & Xie, 2014; Duan et al., 2008; Fukuda et al., 2012; Li et al., 2006, 2009; Zeuner et al., 2009).

**Figure 2. Excitation and emission spectra of the Mn<sup>2+</sup>-activated Ba<sub>2</sub>Si<sub>5</sub>N<sub>8</sub> phosphor.**



**2.3. The mathematical description of the red-emitting phosphor**

The influence of the concentration of the red-emitting Ba<sub>2</sub>Si<sub>5</sub>N<sub>8</sub>Eu<sup>2+</sup> phosphor on the optical performance of the MCW-LEDs can be demonstrated using Mie theory (“Design of LED,” 2011; Winkler et al., 2015). Here, we can apply Mie-scattering theory (“Beyond Mie’s theory II,” 2011; Frisvad, Christensen, & Jensen, 2012; Mackowski, 2012; Wriedt, 2012). The scattering coefficient  $\mu_{sca}(\lambda)$  (mm<sup>-1</sup>), the absorption coefficient  $\mu_{abs}(\lambda)$  (mm<sup>-1</sup>), anisotropy factor  $g(\lambda)$  (mm<sup>-1</sup>), and reduced scattering coefficient  $\delta_{sca}(\lambda)$  (mm<sup>-1</sup>) can be computed by the following expressions (1)–(4):

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

$$\mu_{abs}(\lambda) = \int N(r)C_{abs}(\lambda, r)dr \tag{2}$$

$$g(\lambda) = 2\pi \int_{-1}^1 p(\theta, \lambda, r)f(r) \cos \theta d \cos \theta dr \tag{3}$$

$$\delta_{sca} = \mu_{sca}(1 - g) \tag{4}$$

In these equations,  $N(r)$  indicates the distribution density of diffusional particles (mm<sup>3</sup>).  $C_{abs}$  and  $C_{sca}$  are the absorption and scattering cross sections (mm<sup>2</sup>),  $p(\theta, \lambda, r)$  is the phase function,  $\lambda$  is the light wavelength (nm),  $r$  is the radius of diffusional particles (μm), and  $\theta$  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

$$f(r) = f_{dif}(r) + f_{phos}(r) \tag{5}$$

$$N(r) = N_{dif}(r) + N_{phos}(r) = K_N \cdot [f_{dif}(r) + f_{phos}(r)] \tag{6}$$

$N(r)$  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 diffuser and phosphor particle. Here,  $K_N$  is the number of the unit diffuser for one diffuser concentration and can be calculated by the following equation:

$$c = K_N \int M(r)dr \tag{7}$$

where  $M(r)$  is the mass distribution of the unit diffuser and can be proposed by the following equation:

$$M(r) = \frac{4}{3} \pi r^3 [\rho_{\text{dif}} f_{\text{dif}}(r) + \rho_{\text{phos}} f_{\text{phos}}(r)] \tag{8}$$

Here,  $\rho_{\text{dif}}(r)$  and  $\rho_{\text{phos}}(r)$  are the density of diffuser and phosphor crystal (Jonasz & Fournier, 2007; Luo & Hu, 2014; Mishchenko, Travis, & Lacis, 2002; Sommer et al., 2011; Zhong et al., 2011).

In Mie theory,  $C_{\text{sca}}$  and  $C_{\text{abs}}$  can be obtained by the following expressions:

$$C_{\text{sca}} = \frac{2\pi}{k^2} \sum_0^\infty (2n+1)(|a_n|^2 + |b_n|^2) \tag{9}$$

$$C_{\text{ext}} = \frac{2\pi}{k^2} \sum_1^\infty (2n+1) \text{Re}(a_n + b_n) \tag{10}$$

$$C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}} \tag{11}$$

where  $k = 2\pi/\lambda$ , and  $a_n$  and  $b_n$  are the expansion coefficients with even symmetry and odd symmetry, respectively. They can be calculated by

$$a_n(x, m) = \frac{\psi'_n(mx)\psi_n(x) - m\psi_n(mx)\psi'_n(x)}{\psi'_n(mx)\xi_n(x) - m\psi_n(mx)\xi'_n(x)} \tag{12}$$

$$b_n(x, m) = \frac{m\psi'_n(mx)\psi_n(x) - \psi_n(mx)\psi'_n(x)}{m\psi'_n(mx)\xi_n(x) - \psi_n(mx)\xi'_n(x)} \tag{13}$$

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

Moreover, the phase function  $p(\theta, \lambda, r)$  can be calculated according to

$$p(\theta, \lambda, r) = \frac{4\pi\beta(\theta, \lambda, r)}{k^2 C_{\text{sca}}(\lambda, r)} \tag{14}$$

Here,  $\beta(\theta, \lambda, r)$  is the dimensionless scattering function, which is obtained by the scattering amplitude functions  $S_1(\theta)$  and  $S_2(\theta)$ .

$$\beta(\theta) = (1/2)[|S_1(\theta)|^2 + |S_2(\theta)|^2] \tag{15}$$

$$S_1(\theta) = \sum_{n=1}^\infty \frac{2n+1}{n(n+1)} \left[ a_n \frac{P_n^1(\cos\theta)}{\sin\theta} + b_n \frac{dP_n^1(\cos\theta)}{d\theta} \right] \tag{16}$$

$$S_2(\theta) = \sum_{n=1}^\infty \frac{2n+1}{n(n+1)} \left[ b_n \frac{P_n^1(\cos\theta)}{\sin\theta} + a_n \frac{dP_n^1(\cos\theta)}{d\theta} \right] \tag{17}$$

where  $P_n^1(\cos\theta)$  is the associated Legendre polynomial (“Beyond Mie’s theory II,” 2011; Frisvad et al., 2012; Mackowski, 2012; Wriedt, 2012).

### 3. Results and discussion

As displayed in Figure 3, the scattering coefficients grew with increasing red phosphor concentration. It means that the white-light quality can be enhanced by controlling red phosphor concentration. The reduced scattering coefficient of red phosphor with wavelengths 453, 555, and 680 nm is the same as each other’s (Figure 4). It is indicated that the scattering stability of red phosphor is used for controlling the color quality of MCW-LEDs. The results indicated that red phosphor particles had a significant advantage in producing blue light. The more the blue light emitted, the more the yellow ring phenomenon reduced. Figure 5 shows that the anisotropy coefficient is the same as the red and the blue light. However, the anisotropy coefficient of the yellow light is higher than others. Moreover, the scattering amplitude of the blue light is the highest, and the scattering amplitude of the red light is the lowest compared with each other (Figure 6). This is in connection with the scattering and reduced scattering processes in the phosphor layer according to Mie theory.

Figure 3. Scattering coefficients of the red-emitting phosphor  $Ba_2Si_5N_8Eu^{2+}$ .

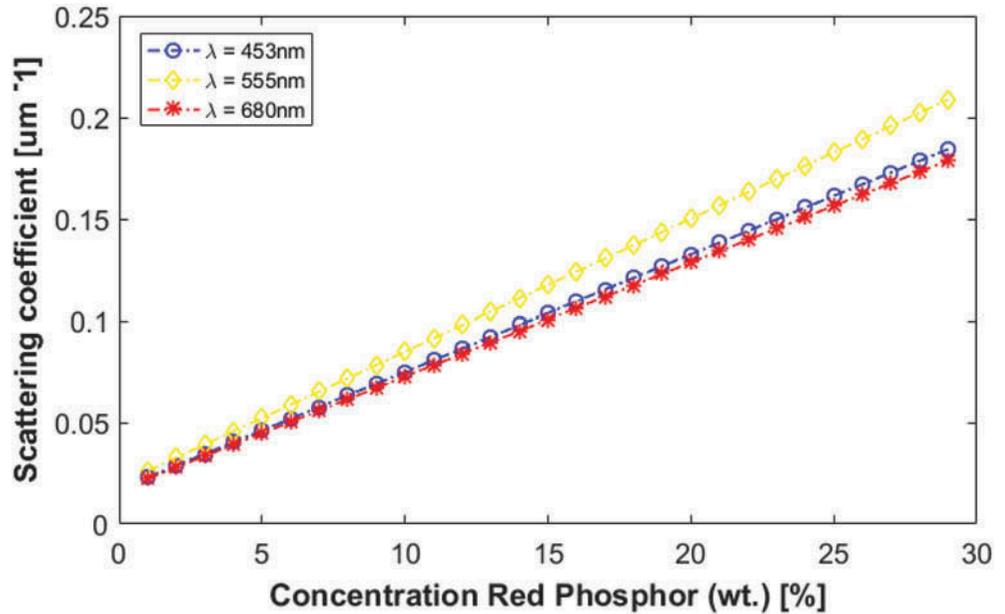
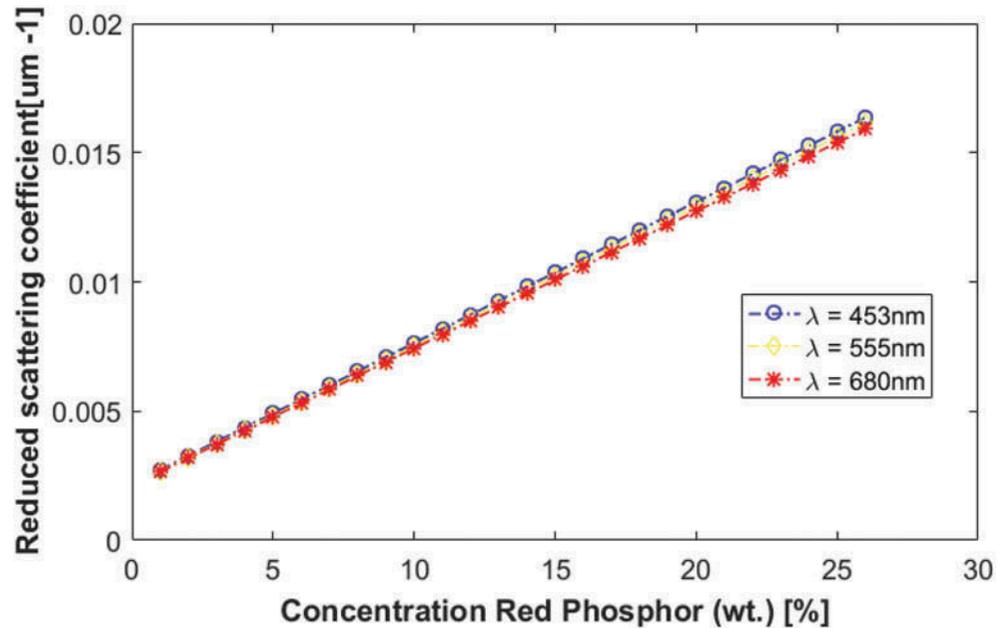


Figure 4. Reduced scattering coefficient of the red-emitting phosphor  $Ba_2Si_5N_8Eu^{2+}$  with wavelengths of 453, 555, and 680 nm.



In this segment, the D-CCT, CRI, CQS, and the lumen output of the 7,700 K and 7,000 K MCW-LEDs are calculated and obtained by using the commercial software Light Tools. In this simulation, the concentration of the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor varied continuously from 0% to 0.4%. From the simulation results, the corresponding values of D-CCT are calculated and are indicated in Figure 7. As shown in Figure 7, the D-CCT had a slight decrease when the concentration of the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor varied from 0% to 0.26%. Then, the D-CCT of the MCW-LEDs increased while the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor changed to 0.4%. The most interesting aspect of this graph is the lowest value of the D-CCT is obtained at

Figure 5. Anisotropy coefficient of the red-emitting phosphor  $Ba_2Si_5N_8Eu^{2+}$  with wavelengths of 453, 555, and 680 nm.

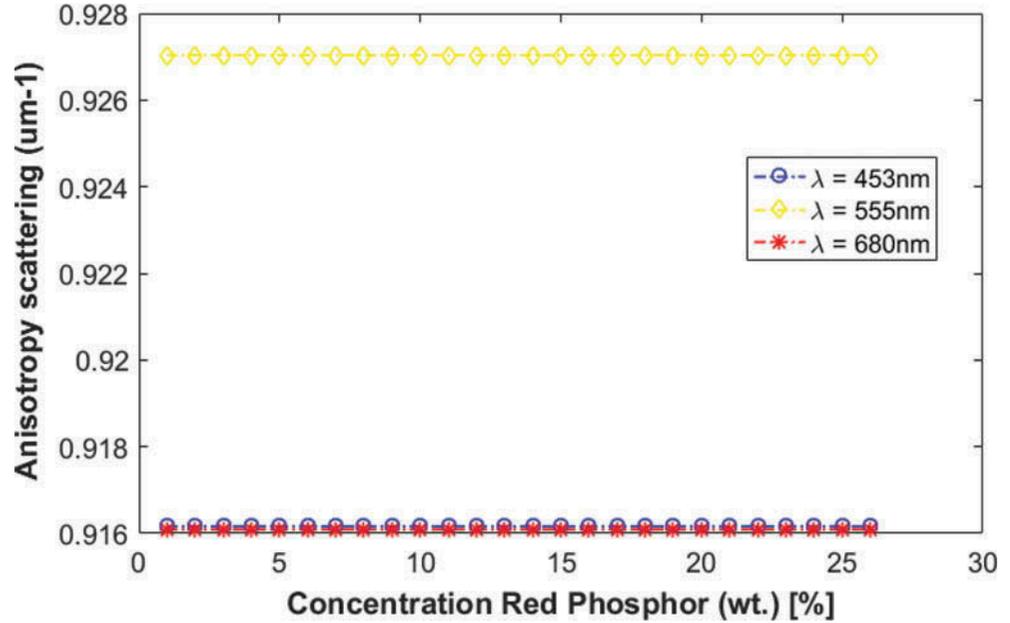
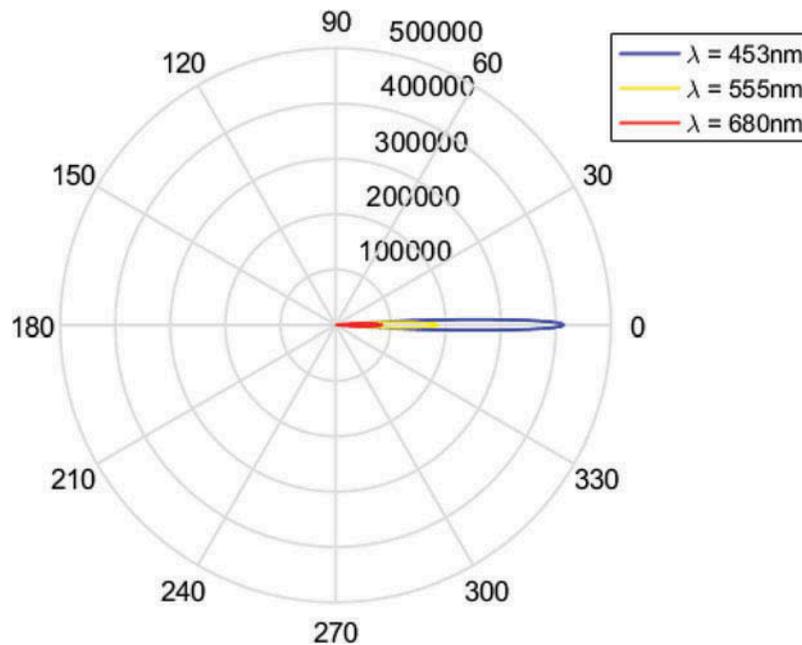


Figure 6. Scattering amplitude of the red-emitting phosphor  $Ba_2Si_5N_8Eu^{2+}$  with wavelengths of 453, 555, and 680 nm.



0.16–0.26% concentration of the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor. Moreover, Figures 8 and 9 show the CQS and CRI of the in-cup packaging MCW-LEDs while the concentration of the red phosphor changed from 0% to 0.4%. It can be indicated that the CQS and CRI grow with the concentration red phosphor in the range from 0% to nearly 0.26%. The highest CQS and CRI is obtained with the red weight varying from 0.24% to 0.26%. As shown in Figures 8 and 9, the

Figure 7. The D-CCT at average CCTs of 7,000 K and 7,700 K.

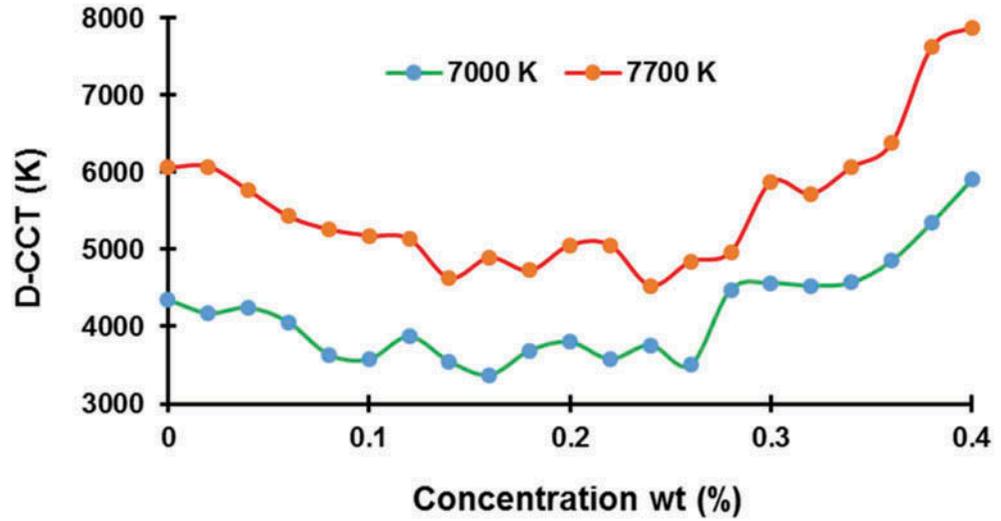
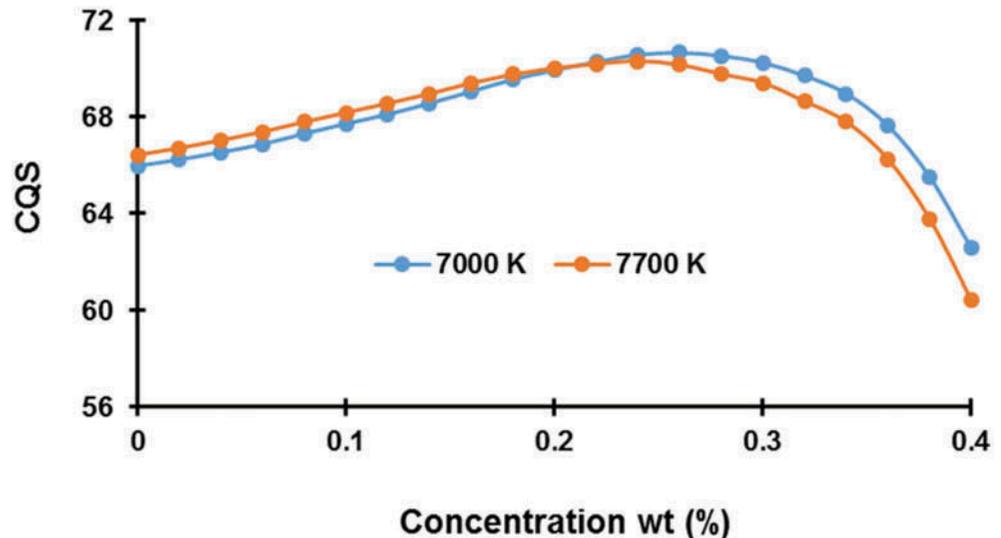


Figure 8. The color quality scale (CQS) at average CCTs of 7,000 K and 7,700 K.



CQS and CRI could obtain the highest value near 70 and 85, respectively. Likewise, the influence of the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor's concentration on the luminous efficacy of the fin-cup packaging MCW-LEDs is shown in Figure 10. The results showed that the lumen output had a considerable decrease while the concentration of the red-emitting phosphor varied from 0.1% to 0.4%. This point of view means that the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor had a harmful impact on the luminous efficacy because of the highly reduced scattering of the blue light in the phosphor compound. It is linked to the loss of the energy of the white light. From that point of view, the CQS, CRI, and D-CCT of high CCT MCW-LEDs can be controlled by adding red-emitting phosphor in phosphor compound and varying its concentration. The best value of the D-CCT, CRI, and CQS can be obtained when the concentration of the red-emitting  $Ba_2Si_5N_8Eu^{2+}$  phosphor is nearly 0.24–0.26%.

Figure 9. The CRI at average CCTs of 7,000 K and 7,700 K.

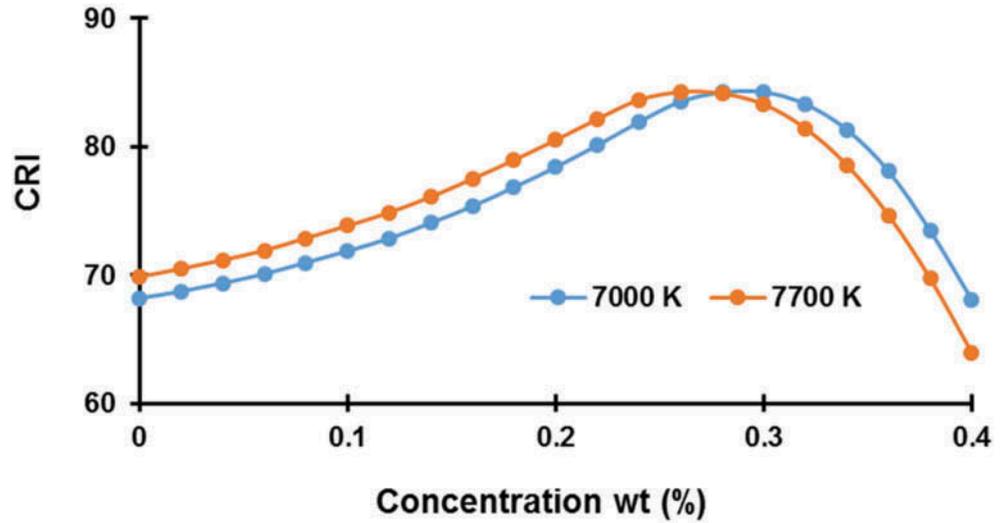
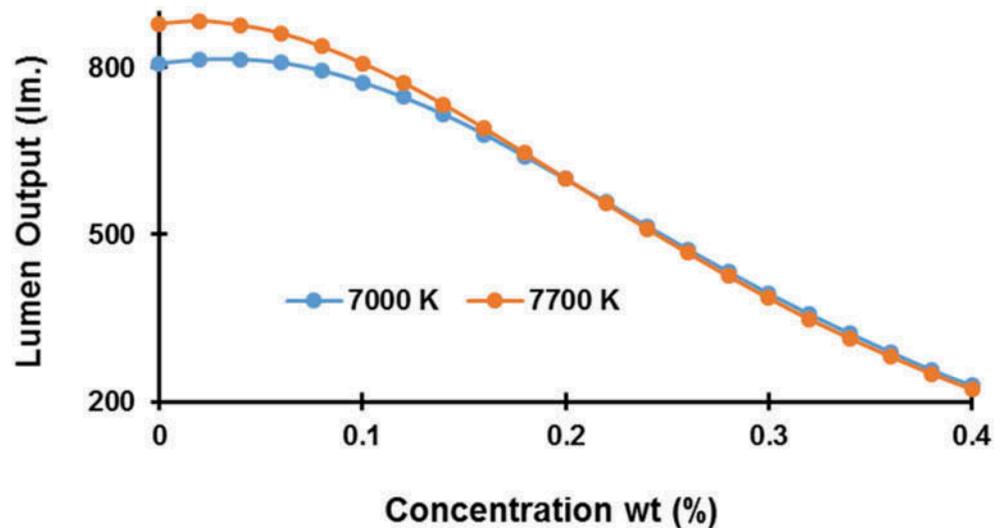


Figure 10. The lumen output at average CCTs of 7,000 K and 7,700 K.



#### 4. Conclusion

The purpose of this paper is to determine the influence of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  conversion phosphor concentration on the optical performance of the 7,000 K and 7,700 K in-cup packaging MCW-LEDs. The results of this investigation show that the concentration of the red phosphor particles crucially influenced the optical performance of the in-cup packaging MCW-LEDs. D-CCT, CRI, and CQS can obtain the best value at the 0.24–0.26% concentration of the red-emitting  $\text{Ca}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor. Further studies need to be carried out to validate the influence of the red-emitting  $\text{Ba}_2\text{Si}_5\text{N}_8\text{Eu}^{2+}$  phosphor size on the optical performance of the in-cup packaging MCW-LEDs.

### Acknowledgement

The authors appreciate the support of Professor Hsiao-Yi Lee, Department of Electrical Engineering, National Kaohsiung University of Applied Sciences, Kaohsiung City, Taiwan.

### Funding

The authors received no direct funding for this research.

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### Citation information

Cite this article as: Red-emitting Ba<sub>2</sub>Si<sub>5</sub>N<sub>8</sub>Eu<sup>2+</sup> conversion phosphor: A new selection for enhancing the optical performance of the in-cup packaging MCW-LEDs, Phu Tran Tin, Nguyen Huu Khanh Nhan, Tran Hoang Quang Minh & Nguyen Thi Phuong Thao, *Cogent Engineering* (2018), 5: 1486153.

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