Enhancement of color quality and luminous flux for remote-phosphor LEDs with red-emitting $\text{CaMgSi}_2\text{O}_6\text{:Eu}^{2+}, \text{Mn}^{2+}$

GUO-FENG LUO¹, NGUYEN THI PHUONG LOAN², LE VAN THO³, NGUYEN DOAN QUOC ANH⁴,
HSIAO-YI LEE^{1,*}

¹Department of Electrical Engineering, National Kaohsiung University of Sciences and Technology, Kaohsiung, Taiwan

²Faculty of Fundamental 2, Posts and Telecommunications Institute of Technology, Ho Chi Minh City, Vietnam

³Institute of Tropical Biology, Vietnam Academy of Science and Technology, Ho Chi Minh City, Vietnam

⁴Faculty of Electrical and Electronics Engineering, Ton Duc Thang University, Ho Chi Minh City, Vietnam

SiO_2 particles and red-emitting $\text{CaMgSi}_2\text{O}_6\text{:Eu}^{2+}, \text{Mn}^{2+}$ phosphor have been added into a yellow phosphor compound YAG:Ce^{3+} to enhance the optical efficiency of white light LEDs whose average correlated color temperature (CCT) is in the range of $5600 \text{ K} \div 8500 \text{ K}$. It was observed that altering $\text{CaMgSi}_2\text{O}_6\text{:Eu}^{2+}, \text{Mn}^{2+}$ concentration from 2 % to 30 % while maintaining 5 % of the SiO_2 strongly influenced the color rendering index (CRI), color quality scale (CQS), and lumen efficiency of the compound. Besides, through the application of Monte Carlo simulation and Mie-scattering theory, it was possible to improve the optical properties by $\text{CaMgSi}_2\text{O}_6\text{:Eu}^{2+}, \text{Mn}^{2+}$ and SiO_2 addition. The results provided a practical approach to achieve higher luminous efficiency and better color uniformity in remote-phosphor white LEDs (RP-WLEDs).

Keywords: remote-phosphor white LEDs; SiO_2 ; $\text{CaMgSi}_2\text{O}_6\text{:Eu}^{2+}, \text{Mn}^{2+}$; color quality scale; Mie-scattering theory.

1. Introduction

In recent years, people have considered using phosphor-converted white LEDs (WLEDs) in lighting applications because of their vital advantages, including flux, energy consumption efficiency, stability, compact size, longevity, and safety, which has created marvelous opportunities for white LEDs in the illumination market [1–3]. Additionally, due to simple fabrication process and low industrial production cost, they have been widely applied in many aspects of life. Nevertheless, typical WLEDs show poor phosphor performance due to the position and arrangement of phosphors in the package [4–7]. Numerous recent studies have focused on enhancing lighting performance of WLED packages with in-cup or conformal phosphor design by non-integrating

or integrating one diffusional particle into a yellow YAG:Ce^{3+} phosphor compound, for example, adding SiO_2 [8, 9] or using $\text{Y}_2\text{O}_3\text{:Eu}^{3+}$ [10]. In 2012, Kwak et al. [11, 12] added red quantum dots to YAG phosphor by layer-by-layer mode for improving the luminous flux and the color rendering index (CRI) of WLEDs. On the other hand, the WLED remote-phosphor structure, in which the phosphor layer and the LED chip are separated from each other, may reduce the absorption ability of the re-emitted light by the LED chip, leading to a better performance of utilized phosphor materials [13, 14]. Nonetheless, very few reports on improving optical efficiency of WLEDs with remote-phosphor packaging design by blending two or more diffusional particles with the original yellow-emitting phosphor materials have been demonstrated. Thus, there remain problems that have not been completely handled: the choice of appropriate diffusive particles and

*E-mail: leehy@nkust.edu.tw

the investigation on how they affect the optical performance of WLEDs. These points will be discussed and presented in this study.

Many phosphor compounds have been recognized as great materials for the development of WLEDs technology [15–19]. Among these kinds of phosphors, $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ possesses superior emission characteristics in 450 nm blue excitation wavelength with a uniform particle size distribution and high performance of LED packages [20–24]. However, there have not been any further studies employing red-emitting $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor to improve the color rendering properties of RP-WLEDs so far.

In this work, the method of mixing SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphors with the $\text{YAG}:\text{Ce}^{3+}$ phosphor compound in RP-WLEDs to improve the lighting efficacy of RP-WLEDs (CRI, CQS, and lumen output) is demonstrated and recommended. As the concentration of SiO_2 has been fixed at 5 wt.%, the effects of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration on CRI, CQS, and lumen output of the WLEDs have been investigated and clarified. The content of this research paper includes two main sections: (1) presenting the setting-up simulation physical model of WLEDs using LightTools software, adding and mixing SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphors into the yellow phosphor composite; (2) analyzing and discussing how $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration affects the optical properties of WLEDs. The results indicate a considerable increase in CRI, CQS and luminous flux by changing $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor content from 2 % to 30 %. Subsequently, the enhancement in RP-WLEDs optical properties by adding SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ particles has been demonstrated using the Mie-scattering theory and Monte Carlo simulations.

2. Preparation and simulation

2.1. Preparation of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor

Red-emitting $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor is frequently employed for very high loading

and long lifetime fluorescent lamps due to the high quantum efficiency, chemical and thermal stability of this material. $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ composition is a result of chemical processes between the materials such as CaO , MgO , SiO_2 , Eu_2O_3 , MnCO_3 and NH_4Cl . This phosphor compound can be considered to be an efficient candidate for improving luminous efficacy of WLEDs. The detailed composition of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor is given in Table 1. The process of creating $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ consists of six steps: mixing, drying, double firing, washing and drying again. Each step plays an important role and is connected with the step(s) that happened previously. All these steps must be taken in following order to achieve optimal effect:

Firstly, mix the materials making their slurry in methanol. The main point of this step is how to soak the materials with a few cubic centimeters liquid. Secondly, dry the mixture in air. After that, fire the dried material in a capped quartz tube. Simultaneously, it has to be combined with N_2 at a temperature of 1000 °C for 1 hour. Then, powdering by frying in a capped quartz tube filled with CO for 1 hour at a temperature of 1150 °C. Next, pick up the product and wash it in water several times. Finally, let it dry to obtain perfect $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$.

2.2. Simulation of RP-WLEDs

A commercial software LightTools 8.1.0 based on the Monte Carlo ray-tracing method was employed to simulate RP-WLEDs which had the average CCT of 8500 K, 7700 K, 7000 K, 6600 K, and 5600 K with the remote phosphor structure. The 3-D simulation physical model of WLEDs for performing optical simulations of remote package WLEDs is shown in Fig. 1.

The input parameters of this model are as follows: the reflector is 2.07 mm high, 8 mm bottom long, and 9.85 mm top long. Nine blue LED chips whose parameters including square base, height and radiant flux at 455 nm wavelength are 1.14 mm, 0.15 mm and 1.16 W, respectively, are attached to the cavity. The parameters of the model are listed in Table 2. The

Table 1. Composition of red-emitting $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor.

Ingredient	Mole [%]	By weight [g]	Molar mass [g/mol]	Mole [mol]	Ions	Mole [mol]	Mole [%]
CaO	45.69	150	56.0774	2.67	Ca^{2+}	2.67	0.19
MgO	16.95	40	40.304	0.99	Mg^{2+}	0.99	0.07
SiO_2	35.82	126	60.08	2.10	Si^{4+}	2.10	0.15
Eu_2O_3	0.17	3.5	351.926	0.01	O^{2-}	8.13	0.58
MnCO_3	1.37	9.2	114.9469	0.08	Eu^{2+}	0.02	0.0014
NH_4Cl	–	5.4	53.49	–	Mn^{2+}	0.08	0.0057

blue chips are covered with a 0.08 mm thick remote phosphor layer, as in Fig. 1b. The concentration of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor particles changed continuously from 2 % to 30 % while SiO_2 content in the compound was kept at 5 %. Nevertheless, each average CCT of the WLED was maintained by controlling the amount of $\text{YAG}:\text{Ce}^{3+}$.

of 7.25 μm and a refractive index of 1.83 for all light wavelengths, while the silicone glue has a refractive index of 1.5. The density of diffusional particles is tailored in order to fix the average CCT figures. If the concentration of the diffuses increases, $\text{YAG}:\text{Ce}^{3+}$ phosphor concentration needs to be reduced to stabilize the average CCTs.

Table 2. The actual parameters of LED chip.

LED vendor	Epistar
LED chip	V45H
Voltage [V]	3.5~3.6
Peak wavelength [nm]	455
Power [mW]	320~340
Lead frame	4.7 mm Jentech Size-S
Die attach	Sumitomo 1295SA
Bonding diagram

Optical properties of SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphors have been determined by using LightTools 8.1.0 software. The refractive index of diffusional particles, such as SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ phosphor, is 1.54 and 1.80. According to the Mie simulation, the average diameter of SiO_2 particles should be of 3 μm . In the conventional phosphor compounds, the yellow phosphor grains have an average radius

3. Computation and discussion

Based on Mie-scattering theory, the relations for the scattering coefficient $\mu_{\text{sca}}(\lambda)$, anisotropy factor $g(\lambda)$, and reduced scattering coefficient $\delta_{\text{sca}}(\lambda)$ are:

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

$$g(\lambda) = 2\pi \int_{-1}^1 \int_0^1 p(\theta, \lambda, r)f(r) \cos \theta d \cos \theta dr \quad (2)$$

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

$N(r)$ and C_{sca} represent the distribution density of diffusional particles (mm^3) and the scattering cross sections (mm^2), respectively, while $p(\theta, \lambda, r)$ shows the phase function. λ means the light wavelength (nm), r indicates the radius of diffusional particles (μm), θ represents the scattering angle ($^\circ$), and $f(r)$ is the symbol of the size distribution function of diffuser in the phosphorous layer. In addition, $f(r)$ could be computed by using the following equation:

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

Fig. 1. (a) photograph of WLEDs sample, (b) illustration of RP-WLEDs with SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ particles in $\text{YAG}:\text{Ce}^{3+}$ compound, (c) simulation of the RP-WLEDs, (d) the simulated WLEDs model.

$$\begin{aligned} N(r) &= N_{\text{dif}}(r) + N_{\text{phos}}(r) \\ &= K_N \cdot [f_{\text{dif}}(r) + f_{\text{phos}}(r)] \end{aligned} \quad (5)$$

$N(r)$ is determined from the diffusional particle density $N_{\text{dif}}(r)$ and the phosphor particle density $N_{\text{phos}}(r)$. $f_{\text{dif}}(r)$ and $f_{\text{phos}}(r)$ are the size distribution function data of the diffuser and phosphor grains. K_N here is the number of the diffuser unit for one diffuser concentration and can be calculated by applying equation 6:

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

In this formula, $M(r)$ indicates the mass distribution of the diffusive unit, defined by:

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

in which, $\rho_{\text{dif}}(r)$ and $\rho_{\text{phos}}(r)$ are the diffuser and phosphor densities.

According to Mie-scattering theory, the expression for C_{sca} is:

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

$k = 2\pi/\lambda$, while a_n and b_n are reckoned by:

$$a_n(x, m) = \frac{\psi'_n(mx) \psi_n(x) - m \psi_n(mx) \psi'_n(x)}{\psi'_n(mx) \xi_n(x) - m \psi_n(mx) \xi'_n(x)} \quad (9)$$

$$b_n(x, m) = \frac{m \psi'_n(mx) \psi_n(x) - \psi_n(mx) \psi'_n(x)}{m \psi'_n(mx) \xi_n(x) - \psi_n(mx) \xi'_n(x)} \quad (10)$$

Here, x is the size parameter, m represents the index of refraction, $\psi_n(x)$ and $\xi_n(x)$ are the Riccati-Bessel functions.

As shown in Fig. 2, the scattering coefficients increase with the concentration of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor. The scattering effect of both $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ and SiO_2 particles influences RP-WLEDs considerably. Due to the strong absorption properties of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ particles, more blue light emitted from LED chips is absorbed. Hence, the

Fig. 2. Scattering coefficients of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ at the wavelengths of 453 nm, 555 nm and 680 nm.

emitted red light is dominant, which can be used as the compensation for red-light components in RP-WLEDs. Furthermore, 5 wt.% of SiO_2 applied to enhance the scattering events contributes to increasing the absorption capability of PC-LEDs. For this reason, $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ and SiO_2 particles are used to produce white LEDs with higher color quality.

Fig. 3. Anisotropic scattering of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ at the wavelengths of 453 nm, 555 nm and 680 nm.

Fig. 3 presents the anisotropy factor of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ particles for 453 nm, 555 nm, and 680 nm wavelengths. The figure indicates that the anisotropic scattering at 680 nm is

higher than at 555 nm. Moreover, the highest value of anisotropy factor has been obtained at 453 nm. To put it another way, $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ particles benefit color uniformity of RP-WLEDs.

The relative refractive indices of diffusor (m_{dif}) and phosphor (m_{phos}) in the silicone compound of RP-WLEDs are expressed by $m_{\text{dif}} = n_{\text{dif}}/n_{\text{sil}}$ and $m_{\text{phos}} = n_{\text{phos}}/n_{\text{sil}}$, where n_{sil} and n_{phos} indicate the refractive indices of silicone (1.53) and phosphor particle, respectively. The phase function can be attained by:

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

where $\beta(\theta, \lambda, r)$, $S_1(\theta)$ and $S_2(\theta)$ are the angular scattering amplitudes that can be obtained by applying the equations:

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

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n(x, m) \pi_n(\cos \theta) + b_n(x, m) \tau_n(\cos \theta) \right] \quad (13)$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n(x, m) \tau_n(\cos \theta) + b_n(x, m) \pi_n(\cos \theta) \right] \quad (14)$$

In Fig. 4, the computed reduced scattering coefficient of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ at three different wavelengths of 453 nm, 555 nm, and 680 nm has been shown. The scattering stability of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ is beneficial to the chromatic quality of RP-WLEDs. Next, computation of the angle-dependent scattering amplitudes of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ has been carried out using MATLAB program. The results reveal that $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ particles have a huge advantage to the scattering characteristics of blue light. In particular, the higher the amount of emitted blue rays, the lower chance that the yellow ring phenomenon occurs. Correspondingly, $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ particles are employed for compensating not only red-light, but also blue-light, as shown in Fig. 5. Results of calculations for CRI, CQS and lumen output depending on $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Mn}^{2+}$ concentration have been shown in Fig. 6 to Fig. 8, respectively.

Fig. 4. Reduced scattering coefficient of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}$ at the wavelengths of 453 nm, 555 nm and 680 nm.

Fig. 5. Angular scattering amplitude of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ at the wavelengths of 453 nm, 555 nm and 680 nm.

Fig. 6. CRI of RP-WLEDs as a function of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration.

Fig. 7. CQS properties of RP-WLEDs as a function of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration.

Fig. 8. Lumen output as a function of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration.

As shown in Fig. 6 and Fig. 7, either CRI or CQS develop extensively if red phosphor concentration varies from 2 % to 30 %. When $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}$ attains 30 %, CRI and CQS values reach their peaks of 85 and 71, respectively. Moreover, the value of angular scattering amplitude of $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ at 453 nm is the highest in comparison with that of 555 nm or 680 nm, as demonstrated in Fig. 5. It means that the blue-light scattering gets a great benefit from the added $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$. This is an advantage not only for improving the color quality, but also for enhancing the luminous flux. However, a significant backscattering event may occur if $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ concentration increases over 15 %, causing the decrease of luminous flux. Therefore, the luminous output increases remarkably at first, reaches the highest value and then decreases slightly, as illustrated in Fig. 8.

4. Conclusions

In this research paper, the influence of diffusional particles SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ on CRI, CQS and luminous flux of WLEDs are presented. Based on the simulation results and the theoretical analysis, the following conclusions can be drawn:

1. A huge increase in CRI and CQS values is observed when $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphor concentration grows from 2 % to 30 %. At 30 wt.% red-emitting phosphor, WLEDs can achieve the best CRI and CQS values which are 85 and 71, respectively.
2. Luminous flux of WLEDs firstly increases remarkably, up to the highest value, and then decreases slightly.

In conclusion, the article offers an important technical implication for WLEDs fabrication and an appropriate phosphor material selection for WLED enhancement. Besides, the effects of particle sizes of SiO_2 and $\text{CaMgSi}_2\text{O}_6:\text{Eu}^{2+},\text{Mn}^{2+}$ phosphors on WLEDs optical performance will be presented, analyzed and demonstrated in our further works.

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