INFLUENCE OF SCATTERING ENHANCEMENT PARTICLES $CaCO_3$, CaF_2, SiO_2 AND TiO_2 ON COLOR UNIFORMITY OF WHITE LEDS

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Abstract. *In this paper, the influence of scattering enhancement particles $CaCO_3$, CaF_2, SiO_2 and TiO_2, adding to YAG:Ce phosphor compounding, on color uniformity of white LEDs (W-LEDs) was presented. Firstly, the physical model of multi-chip W-LEDs is simulated and demonstrated by using commercial LightTools 8.1.0 program. After that, the influence of scattering enhancement particles on color uniformity is calculated and analyzed. With using the Monte Carlo simulation and the Mie-scattering theory, the color uniformity improvement of an 8500 K W-LEDs is demonstrated convincingly. From the researched results, the best color uniformity can be accomplished with TiO_2 particles. The results and discussions provided a practical approach for higher-quality manufacturing W-LEDs.*

Keywords

$CaCO_3$, CaF_2, color uniformity, SiO_2, TiO_2, white LEDs.

1. Introduction

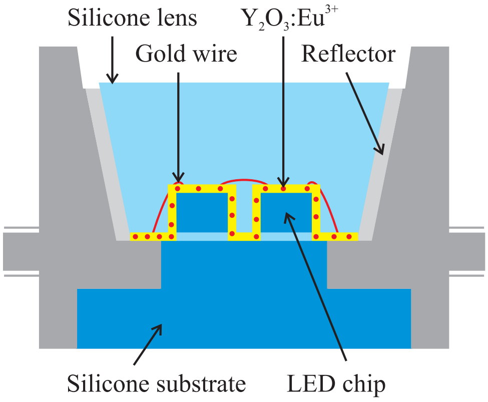
Nowadays, W-LEDs are becoming increasingly important light sources for illumination applications, because they are long-life, compact, mercury-free and energy-efficient. Color uniformity is the main optical properties of W-LEDs and it could be improved in many previous papers [1], [2] and [3]. All these studies started from the scattering enhancement in phosphor-converted white-LEDs (PC-LEDs). In fact, the structure of PC-LEDs is the combination of YAG:Ce phosphor and silicone glue. The YAG:Ce phosphor absorbs the exciting blue light from the chips to stimulate the yellow light and thus result in white light with the desired color temperature [4]. In other words, in these studies, the color uniformity of LEDs was improved by optimizing the state of the phosphor or the optical structure of PC-LEDs. In conclusions, the spatial color uniformity of PC-LEDs can be controlled by the thickness and the concentration of the phosphor [9]. Moreover, the location of phosphor material in the silicone layer significantly effects on the color performance. The color temperature of PC-LEDs has demonstrated the strong influence of the refractive indexes of the silicone matrix and the phosphor materials and the size of phosphor particles [10].

In this study, we concentrated on finding one particle from scattering enhancement particles $CaCO_3$, CaF_2, SiO_2 and TiO_2, which is employed for manufacturing higher-quality W-LEDs. The target of study is an improvement the color uniformity of W-LEDs. This research paper can be divided into three main sections: In Section 2. , the physical model of 8500 K W-LEDs is simulated and demonstrated by using commercial LightTools 8.1.0 program. In Section 3. , by adding one of scattering enhancement particles $CaCO_3$, CaF_2, SiO_2 and TiO_2 to YAG:Ce phosphor compounding, the color uniformity is simulated, calculated and analyzed: In Section 4. , the simulation can be convinced by using the Monte Carlo simulation and the Mie-scattering theory. In this study, the results demonstrated that the best color uniformity of 8500 K W-LEDs could be accomplished with TiO_2 particles. This results can consider the prospective solution for higher-quality manufacturing W-LEDs in the near future.

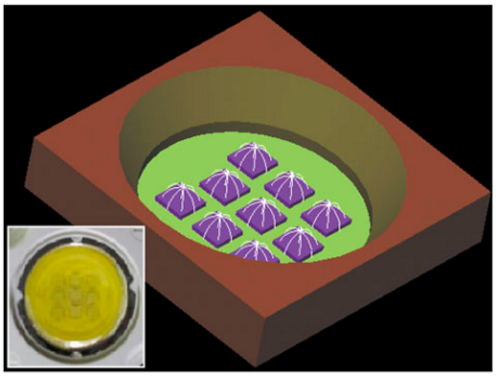
2. Physical Model

In this work, an 8500 K W-LEDs with the conformal phosphor structure is simulated by using the commercial LightTools software based on the Monte Carlo ray-tracing method. To perform optical simulations, we built 3-D models (Fig. 1). In this research, W-LEDs has commonly configured:

- The reflector has a bottom length of 8 mm, a height of 2.07 mm and a length of 9.85 mm at its top surface.
- The conformal phosphor layer with a fixed thickness of 0.08 mm covers the 9 LED chips.
- Each LED chip with a square base of 1.14 mm and a height of 0.15 mm is bound in the cavity of the reflector (Fig. 1(b)).The radiant flux of each blue chip is 1.16 W at wavelength 455 nm.



(a) The conformal phosphor structure.



(b) The original lamps and physical model.

Fig. 1: W-LEDs structure.

To maintain the average Correlated Color Temperature (CCT) of 8500 K, the YAG:Ce concentration changes to the concentration of $CaCO_3$, CaF_2, SiO_2 and TiO_2.The refractive index of the diffusors such as $CaCO_3$, CaF_2, SiO_2 and TiO_2 are chosen as 1.66, 1.44, 1.47 and 2.87, respectively. The diffusers are assumed to be spherical and have radius 0.5 µm. The average radius of the phosphor particles are 7.25 µm and have a refractive index of 1.83 at all wavelengths of light. The refractive index of the silicone glue is 1.5. The diffusional particle density is varied for optimizing illumination CCT uniformity and output efficiency by the expression:

$$W_{phosphor} + W_{silicone} + W_{diffusor} = 100\ \%, \qquad (1)$$

where $W_{silicone}$, $W_{phosphor}$ and $W_{diffusor}$ are the weight percentages of the silicone, phosphor and diffuser of the W-LEDs, respectively. To maintain the mean CCT value of 8500 K, the weight of YAG:Ce phosphor should be decreased when the weight percentage of the diffuser is increased.

3. Results and Discussion

For improving the light quality of the W-LEDs, the difference of angular CCT Deviation (D-CCT) between the normal and large angle is an important standard to evaluate in the solid-state lighting application [9]. The larger D-CCT can cause the yellow ring phenomenon and generate the non-uniform white color at the different angle [14]. In this study, the D-CCT is expressed as D-CCT = CCT (Max) − CCT (Min). Here CCT (Max) and CCT (Min) are the maximal CCT at the zero degree of viewing angle and minimal CCT at the 70 degree of viewing angle, respectively. The scattered light of each particle in PC-LEDs is different, resulting in varying the optical properties of W-LEDs. If the scattered blue light is enhanced enough, the D-CCT can be reduced significantly. Conversely, the D-CCT should be increased with lack or redundancy of the scattered blue light in W-LEDs. The scattered blue light not only combines with the converted yellow but also combine the yellow ring for emitting white light, resulting in a reduction of yellow ring phenomenon of W-LEDs. It can be seen in Fig. 2, where the D-CCT of $CaCO_3$ and TiO_2 cases have a downward trend. Meanwhile, the D-CCT of CaF_2 and SiO_2 cases grow with their concentration.

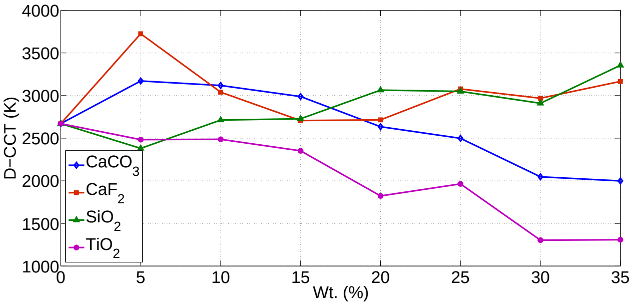


Fig. 2: The impact of the diffusive particles concentration on CCT deviations.

4. Scattering Description

Simulation results can be investigated and demonstrated by Matlab software using Mie-scattering theory [11]. The scattering coefficient $\mu_{sca}(\lambda)$, anisotropy factor $g(\lambda)$ and reduced scattering coefficient $\delta_{sca}(\lambda)$ are calculated by expression Eq. (2), Eq. (3) and Eq. (4):

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

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

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

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 (degree) and $f(r)$ is the size distribution function of the diffusers in the phosphor layer.

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

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

where $N(r)$ is composed of the diffusive particle number density $N_{dif}(r)$ and the phosphor particle number density $N_{phos}(r)$. $f_{dif}(r)$ 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:

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

To obtain K_N, we should first know the mass distribution $M(r)$ (milligrams) of the unit diffusor. Below equation can calculate $M(r)$:

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

where ρ_{dif} and ρ_{phos} are the density of diffusor and phosphor crystal.

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

$$C_{sca} = \frac{2\pi}{k^2} \sum_{0}^{\infty} (2n-1)(|a_n|^2 + |b_n|^2), \tag{9}$$

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:

$$a_n(x,m) = \frac{\Psi'_n(mx)\Psi_n(x) - m\Psi_n(mx)\Psi'_n(x)}{\Psi'_n(mx)\xi_n(x) - m\Psi_n(mx)\xi'_n(x)}, \tag{10}$$

$$a_n(x,m) = \frac{m\Psi'_n(mx)\Psi_n(x) - \Psi_n(mx)\Psi'_n(x)}{m\Psi'_n(mx)\xi_n(x) - \Psi_n(mx)\xi'_n(x)}, \tag{11}$$

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

According to Eq. (3), the theoretical results of $g(\lambda)$ are calculated and shown in Fig. 3, Fig. 4 and Fig. 3. Results show that the variation of the diffuser concentration has a slight impact on the anisotropy factor $g(\lambda)$ and the increase of $g(\lambda)$ by the diffusional particle density is so small that the increase can be neglected. The anisotropy factor of particles for a long wavelength should be larger than that of a short wavelength. It means that the particles should present stronger a scattering effect for a short wavelength. This theoretical result can be modified in the following angular scattering amplitudes simulation shown in Fig. 3, Fig. 4 and Fig. 5.

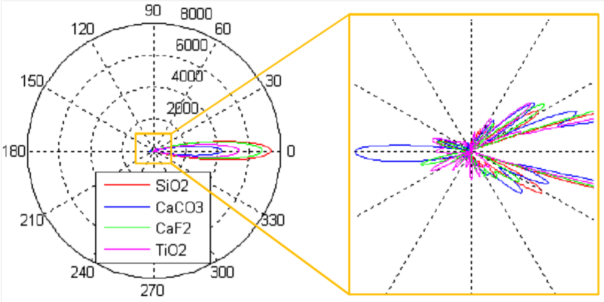


Fig. 3: The angular scattering amplitudes of the various diffusional particles with sphere diameter = 1 µm for blue light = 455 nm.

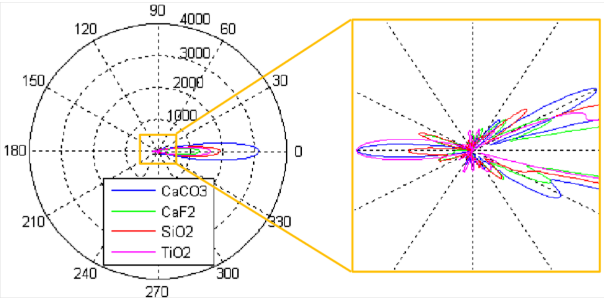


Fig. 4: The angular scattering amplitudes of the various diffusional particles with sphere diameter = 1 µm for yellow light = 595 nm.

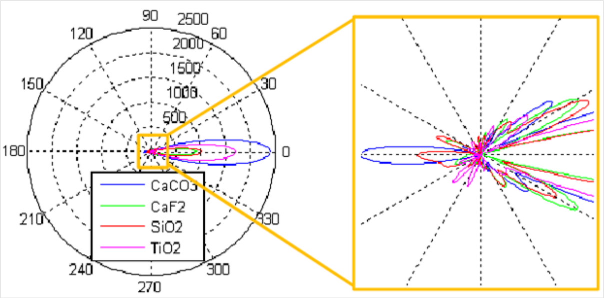


Fig. 5: The angular scattering amplitudes of the various diffusional particles with sphere diameter = 1 μm for red light = 680 nm.

In the mixture of phosphor, diffusor and silicone, the refractive index of embedded silicone (n_{sil}) is 1.53 and the refractive index of diffusor (n_{dif}) are 1.66, 1.44, 1.47 and 2.87 respectively. Silicone and diffusors are considered to be transparent for the blue light and the yellow light. The refractive index of the phosphor particle (n_{phos}) has a complex form. Therefore, the relative refractive indices of diffusor (m_{dif}) and phosphor (m_{phos}) in the silicone are $m_{dif} = n_{dif} \cdot n_{sil}^{-1}$ and $m_{phos} = n_{phos} \cdot n_{sil}^{-1}$. For small spheres, the phase function $p(\theta, \lambda, r)$ can be calculated according to the following equation [12] and [13]:

$$p(\theta, \lambda, r) = \frac{4\pi\beta(\theta, \lambda, r)}{k^2 C_{sca}(\lambda, r)}, \tag{12}$$

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

$$\beta(\theta, \lambda, r) = \frac{1}{2}\left[|S_1(\theta)|^2 + |S_2(\theta)|^2\right]. \tag{13}$$

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \begin{bmatrix} a_n(x,m)\pi_n(\cos\theta) \\ +b_n(x,m)\tau_n(\cos\theta) \end{bmatrix}. \tag{14}$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \begin{bmatrix} a_n(x,m)\tau_n(\cos\theta) \\ +b_n(x,m)\pi_n(\cos\theta) \end{bmatrix}. \tag{15}$$

In equations Eq. (14) and Eq. (15), the angular dependent functions and are expressed in the angular scattering patterns of the spherical harmonics.

5. Conclusion

In this research, the influence of $CaCO_3$, CaF_2, SiO_2 and TiO_2 on color uniformity of 8500 K MCW-LEDs was presented, calculated, analyzed and demonstrated. From the researched results, some conclusions are proposed:

- The CCT deviation has a decreasing tendency when the concentration of $CaCO_3$ and TiO_2 increases.
- Meanwhile the CCT deviation of CaF_2 and SiO_2 cases grow with their concentration.
- The best color uniformity of W-LEDs can be obtained in TiO_2 case. In summary, TiO_2 particles should be chosen for improving the color uniformity of W-LEDs. This research provided an important technical implication for the selection of phosphors in WLED manufacturing and development of phosphor materials for WLED applications. In further research, color rendering index and luminous efficiency of MCW-LEDs by adding $CaCO_3$, CaF_2, SiO_2 and TiO_2 particle into the phosphor compounding is necessary to analyze and demonstrate.

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