Determining the suitable SEPs of $CaCO_3$ and TiO_2 for specific optical enhancements of phosphor-converted LEDs

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The development method of pc-LED, a phosphor-converted light emitting diode, with scattering enhancement particles (SEPs) at 8500 K correlated color temperature is analyzed in this article. The pc-LED is an innovative lighting solution that has been used in several different categories, thus this article offers the approach to improve the color homogeneity and luminous flux of the pc-LEDs in order to fulfill the requirements of current lighting. Also, the extensive studies on the two SEPs, calcium carbonate ($CaCO_3$) and titania (TiO_2), are demonstrated in the article. In order to examine their properties and effects on the illumination of pc-LEDs, the tests involve combining both of these SEPs with a yellow phosphor $Y_3Al_5O_{12}:Ce^{3+}$. The analysis of SEPs includes the dispersion coefficients, the anisotropic dispersion, the diminished dispersion, and the dispersion amplitudes at 450 nm and 550 nm. The LightTools program is used to construct the pc-LED simulation, and the Mie-scattering theory can then validate the effects of the optical simulation. The research findings conclude that TiO_2 particles are appropriate for color homogeneity and CCT-deviation reduction, while $CaCO_3$ particles are successful in limiting significant decrease in lumen efficiency and the color deviation at the corresponding color temperature.

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1. Introduction

In recent years, the pc-LED has gained a lot of attention in the lighting industry with formidable features such as high reliability, cost and energy efficiency, and environmental friendliness. It has been demonstrated that this lighting source has good luminous efficiency but low quality of color. It is important for pc-LED to achieve the advancements in its optical performances to keep up with the continuously rising demand for lighting applications. Therefore, ameliorating critical aspects of pc-LED, color characteristics and luminous efficacy, which have tremendous effects on the efficiency of light output is the key concern [1, 2]. Mixing the yellow light from the combination of yellow phosphor $YAG:Ce^{3+}$ and silicone glue with the blue light from the LED chips is the most common process to produce white light. Yellow phosphors would absorb the blue light from the chips, thus producing white light with the desired color temperature [3]. After every scattering event, the yellow light becomes stronger owing to this absorbing feature. Probably, this leads to the insufficient blue-light proportion during the white-light formation. The mismatch between the emitted blue and yellow lights leads the distribution of color into being irregular [4, 5]. The issue that would typically emerge from the inhomogeneous chromatic spread is the phenomenon of the yellow ring. Specifically, a yellow circle around the perimeter of the pc-LED will be formed due to the difference in blue and yellow light distribution, and thus creating confusion in the eyes of the viewers. There are several ways to address the deviated color issue. To change the spatial color homogeneity of pc-LEDs, we can use the variations in ranges and characteristics of wavelengths of the phosphor. In particular, SiO_2, B_2O_3, PbO, $YAG:Ce^{3+}$ phosphor-in-glass (PiG) particles combined with silicone glue can reduce the color variance from 761 K to 171 K at an average correlated color temperature (CCT) of 6000 K [6]. Besides, HfO_2/SiO_2 DBR film reduced the color deviation at 5000 K CCT from 1758 K to 280 K [7]. Also, the micro-patterned is effective by deducting approximately 441 K color deviation value at 5337 K CCT [8]. The effects of these approaches are profound in improving spatial color homogeneity and have been studied, but the difficulties in manufacturing as well as the high cost of production prevent them from being commonly used. As a result, a more practical approach to the enhancement of LED phosphor structure is the scattering enhancement particle (SEP). Some SEPs that have been investigated for a new phosphor compound are TiO_2, ZrO_2, microspheres, and SiO_2 [9, 10]. There are several research findings highlighting the positive effects of these SEPs on the lighting efficiency of pc-LEDs, especially Titania (TiO_2)

particles, which can improve the color quality when being presented in the phosphorous layer at 0.1% [11-15]. In addition, with respect to the magnification of color homogeneity, the addition of up to 10% of $CaCO_3$ can lead to an acute increase in this property [14]. In addition, the SiO_2 particle is a special SEP that can regulate color uniformity, improve color quality when put in a suitable phosphorus-silicone compound, and adjust the color temperature by changing the particle size when being added to the phosphorus layer of pc-LEDs [16]. The results all pointed out that SEPs are good for pc-LED development, yet more details on the SEP selection for the enhancement of a specific LED lighting performance are barely provided.

In this study we will demonstrate the analysis on the uses of $CaCO_3$ and TiO_2 particles as they are one of the most commonly used SEPs for the optical enhancement of pc-LED. From the achieved results by calculation and comparison of the scattering characteristics of these two SEPs, it is possible to decide the most effective SEP for the improvement each optical property. The results of previous studies indicated, SEP particles can increase lighting performance when given the proper particle size and concentration, therefore, these parameters are also determined using the Mie scattering theory and provided in this article [17, 18]. In 2 sections, this article addresses these experimental findings, with section 2 displaying the pc-LED scattering amplitude and section 3 presenting the equations used to calculate optical parameters and compare the obtained results. Section 4 concludes all the research results and provides the recommendation for the application of SEPs to improve pc-LED lighting performance.

2. Scattering analysis

A multi-paradigm numerical computing environment, the MATLAB, is used to measure the light scattering process parameters. The scattering properties of SEPs that were coated on the LED chip by the conformal coating method were measured and utilized on the basis of the Mie-scattering theory [19, 20]. The expressions used to measure the scattering coefficient $\mu_{sca}(\lambda)$, the anisotropy factor $g(\lambda)$, and the decreased scattering coefficient $\delta_{sca}(\lambda)$ can be:

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

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

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

In (1), the distribution density of diffusional particles (mm^3) is $N(r)$, and the scattering cross sections are shown by C_{sca} (mm^2). In (2), $p(\theta,\lambda,r)$ means the phase function, θ shows the scattering angle (°*C*), λ indicates the wavelength of light (nm), and r is defined as the diffusion particle radius (μm). $f(r)$ shows the diffuser's size distribution function in the phosphor film, as expressed in (4), and *(r)* can be determined for this parameter.

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

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

In (5), the density of diffusive and phosphor ions, respectively, are $N_{dif}(r)$ and $N_{phos}(r)$ contained in $N(r)$. Meanwhile, $f_{dif(r)}$ shows the diffuser's size distribution function data and $f_{phos(r)}$ represents that of the phosphor particle. Here, for one diffuser concentration, K_N indicates the quantity of the diffuser unit. The K_N computation can be carried out as follows:

$$c = K_N \int M(r) dr \tag{6}$$

$M(r)$ here implies the diffusive unit's mass distribution, which can be determined by the following equation:

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

with $\rho_{diff}(r)$ and $\rho_{phos}(r)$ are the diffusor and phosphor crystal densities, respectively.

In the Mie-scattering theory, the scattering cross section (C_{sca}) can be represented as:

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

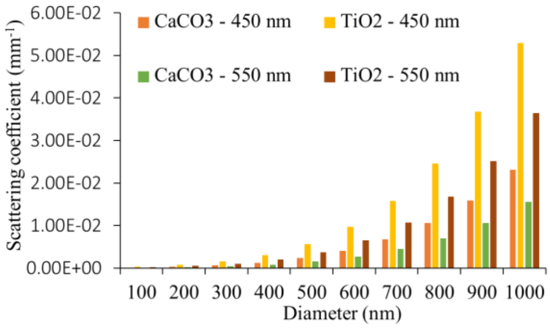
in which $k = 2\pi/\lambda$, a_n and b_n are expressed as follows:

$$a_n(x,m) = \frac{\psi_n'(mx)\psi_n(x) - \mathrm{m}\psi_n(mx)\psi_n'(x)}{\psi_n'(mx)\xi_n(x) - \mathrm{m}\psi_n(mx)\xi_n'(x)} \tag{9}$$

$$b_n(x,m) = \frac{m\psi_n'(mx)\psi_n(x) - \psi_n(mx)\psi_n'(x)}{m\psi_n'(mx)\xi_n(x) - \psi_n(mx)\xi_n'(x)} \tag{10}$$

with $x = k.r$, m presents the refractive index, $\psi_n(x)$ and $\xi_n(x)$ are the Riccati - Bessel functions. The computations for the relative refractive indices of the silicone diffusor (m_{dif}) and phosphor (m_{phos}) can then be described as $m_{dif} = n_{dif}/n_{sil}$ and $m_{phos} = n_{phos}/n_{sil}$.

Fig. 1(a) compares scattering coefficients with different particle sizes of $CaCO_3$ and TiO_2. Simulation results show that the larger the particle size, the greater the scattering coefficient, at both 450 nm and 550 nm. The scattering coefficient in general evaluates particle scattering ability of the particles. If the scattering coefficient is larger, the scattering intensity will be stronger and the scattering angle will be smaller. The scattering properties in Figs. 1(b), 1(c), 1(d) have similar direction pulses, the larger the particles, the greater the scattering intensity. On the other hand, it is easy to see that the scattering at the wavelength of blue light is greater than the scattering intensity at the yellow light wavelength. This proves that the scattering intensity of blue light is stronger than the yellow-light one. Since the amount of blue light is required to reduce the yellow ring phenomenon, this increase in the blue light proportion has important significance, proving that $CaCO_3$ and TiO_2 are suitable to be applied in this study.



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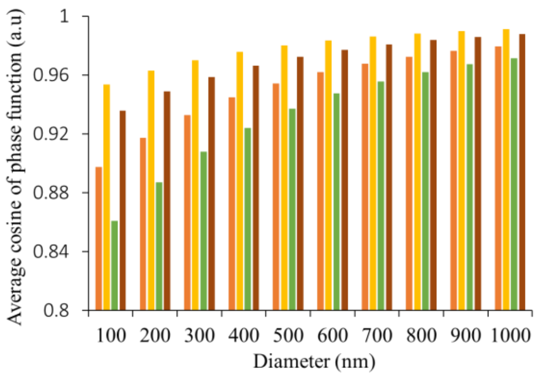

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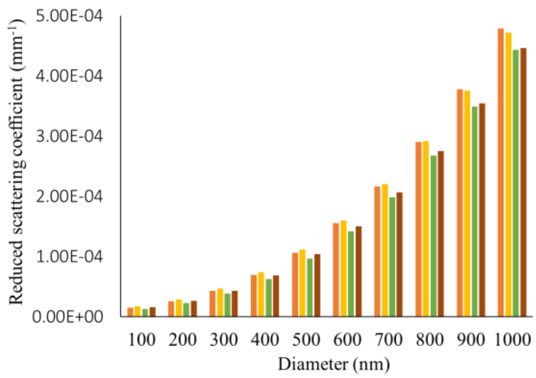

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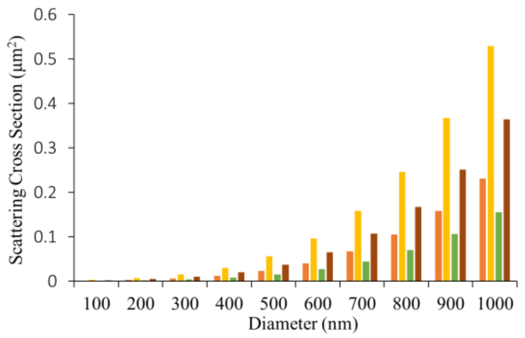


(d)

Fig. 1. Comparison of scattering properties in pc-LEDs with $CaCO_3$ and TiO_2 (color online)

SEPs are mixed in the phosphor layer to increase the scattering, and as the scattering frequency increases, stimulating the blending times between the blue and yellow rays, and eventually generating the more uniform white light. Furthermore, increasing the blue light to the left and right of the phosphor layer helps these blue rays escape and combine with the yellow ring to form white light or limit the yellow ring effect.

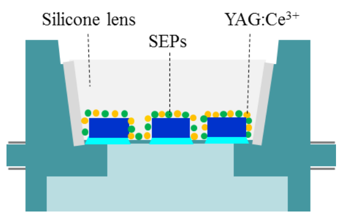
This means less yellow ring, more uniform chromaticity. Yellow rings are yellow streaks of light appearing on the receiving surface of the light. This reduction in wall has been being improved by scientists so far. Therefore, the results of this study are meaningful to the concentration and size selection of SEPs for the quality improvement of LED lamps. The smaller the particle size, the better the color quality, because the small particle size offers larger scattering angle, causing the process of mixing light rays to occur more often than the particle of large size does. However, this multi-mixing process reduces the released energy, which in turn increases the brightness of the LED. Depending on the requirements, the manufacturer chooses the right size.

The scattering of the particles remains the core to adjust the color quality as well as the clearance of the LED. In this study, this is the first time the application of SEPs has been analyzed deeply through the results of Mie theory simulation. The results have important implications for the manufacturer to select the suitable SEP radius when being applied in production. Another concern in using SEPs for pc-LEDs is their concentration. In Section 3, the effects of $CaCO_3$ and TiO_2 will be discussed and examined with the support of the results from the scattering simulation in Fig. 1.

3. Computation and discussion

Fig. 2 shows the physical configuration of the pc-LED used in the experiments with various types of SEPs. The detailed lighting performance measurements are computed using LightTools 8.1.0. A 0.08 mm phosphor layer and 9 LED chips make up the pc-LED model. The reflector parameters include a depth of 2.1 mm, an inner diameter of 8 mm and a surface diameter of 10 mm. The particles of $CaCO_3$ and TiO_2 are spherical and have a diameter of approximately 0.5 µm, with a refractive index of 1.66 and 2.87, respectively.

(a)



(b)

Fig. 2. (a) pc-LED model used in the research, (b) 2D schematic image of pc-LEDs model (color online)

Managing the diffusional particle density to preserve the predetermined associated color temperature by balancing the weight percentages between the components in the phosphor layer is the key objective of adding a SEP to the phosphorus packaging of a pc-LED. To properly adjust the particle density and optimize the CCT and light output, the following equation can be used:

$$W_{phosphor} + W_{silicone} + W_{SEP} = 100\% \qquad (11)$$

In this equation, $W_{silicone}$ is the weight percentage of silicone, $W_{phosphor}$ is the weight percentage of phosphor particle, and W_{SEP} is the weight percentage of SEP. Equation (11) indicates the constituent weight percentages leading to the pc-LED phosphor layer. According to the equation, to maintain the average associated color temperature, which is 8500 K in this analysis, as the concentration of SEPs increases and leads to a rise in the weight percentage of SEP, the weight percentage of the yellow phosphor YAG:Ce^{3+} must decrease the same amount.

CCT deviation, the principal reason that causes the phenomenon of yellow-ring and chromatic inhomogeneity, is strongly linked to light emission angles. In order to determine the light quality of pc-LEDs, the minimization in CCT differences at various emission angles is therefore important and needs to be included in the analysis. We can use the following equation to calculate the angular CCT deviation:

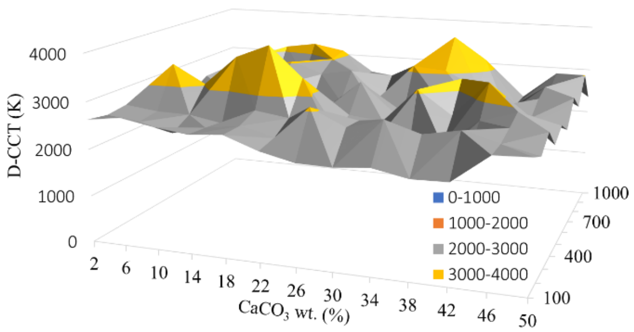
$$\Delta CCT = CCT_{(Max)} - CCT_{(Min)} \qquad (12)$$

Based on the viewing angle, where $CCT_{(Max)}$ reflects the highest CCT at 0°, and $CCT_{(Min)}$ is the lowest CCT at 90°.

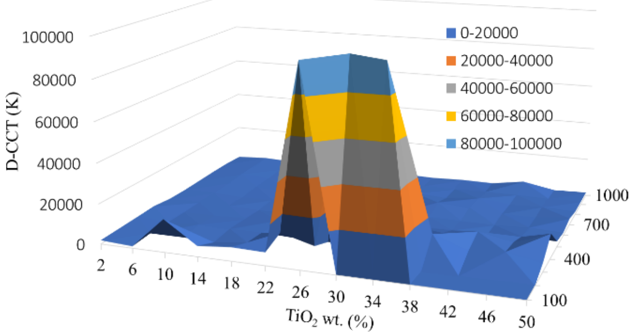
$CaCO_3$ will decrease deviated color lower than any other SEPs, as stated in an earlier SEP study, since its distribution of light intensity between the scattered blue light and the emitted yellow light shows the smallest variations. It is noted that the CCT deviation is caused by the dissimilarity of optical properties between the phosphor particles inside the pc-LED; in other words, the hue differences of a LED are probably caused by the inequal generated blue and yellow lights. Thus, it is understandable why the CCT deviation value can be reduced by increasing the emitted blue light to an amount that can suit the emitted yellow light. Therefore, owing to the scattered-blue-light absorption of yellow phosphors, the lack of required blue-light proportion occurs, resulting in the excessive yellow light, and finally degrading the color quality of pc-LEDs. As white light is a combination of blue light and yellow light, the more the blue lights are emitted, the more the unnecessary yellow lights that cause the phenomenon of yellow-ring are decreased, resulting in the enhanced white light and the eradicated yellow-ring. For the management and enhancement of WLED optical properties with SEPs of $CaCO_3$ and TiO_2, the results in color homogeneity and luminous flux can be useful.

As can be seen from Fig. 3 illustrating the CCT deviation of $CaCO_3$- and TiO_2-doped pc-LED configurations, both SEPs are effective in reducing the distribution difference of radiant intensity. In the case of using $CaCO_3$ at 30% concentration, the CCT deviation falls by 620 K, from 2670 K to 2050 K. With 30% TiO_2 added in the phosphor package, the CCT deviation decreases from around 2600 K to 1300 K, which is two times lower than the CCT deviation with 0% TiO_2. This means the TiO_2 is more suitable for reducing the color deviation and enhancing the color uniformity.

Luminous flux, which can be measured in Fig. 4, is the last critical optical property for pc-LEDs. The luminous efficiency of pc-LEDs using $CaCO_3$ and TiO_2 at various concentrations, from 0% to 50%, is shown in Fig. 4. Furthermore, their particle size is adjusted from 100 nm to 1000 nm. As can be seen in Fig. 4 (a), the luminous flux becomes higher as the concentration of $CaCO_3$ increases, at all particle sizes. Besides, Fig. 4 (b) shows that the lumen output shows its enhancement when using TiO_2 at below 10% and starts to decrease gradually as the TiO_2 concentration continuously grows from 10%, regardless of the particle sizes. The results of the pc-LED with $CaCO_3$ indicate that the lighting performance is greatly influenced by the concentration and particle size of this SEP. In particular, as the particle size grows, the scattering of light is degraded, while the luminous flux receives benefits from this increase in diameter. It is also indicated from Fig. 4 that when using small particle sizes of $CaCO_3$ (smaller than 500 nm), the concentration of this SEP should not be over 30% to avoid the unwanted decrease in lumen output.

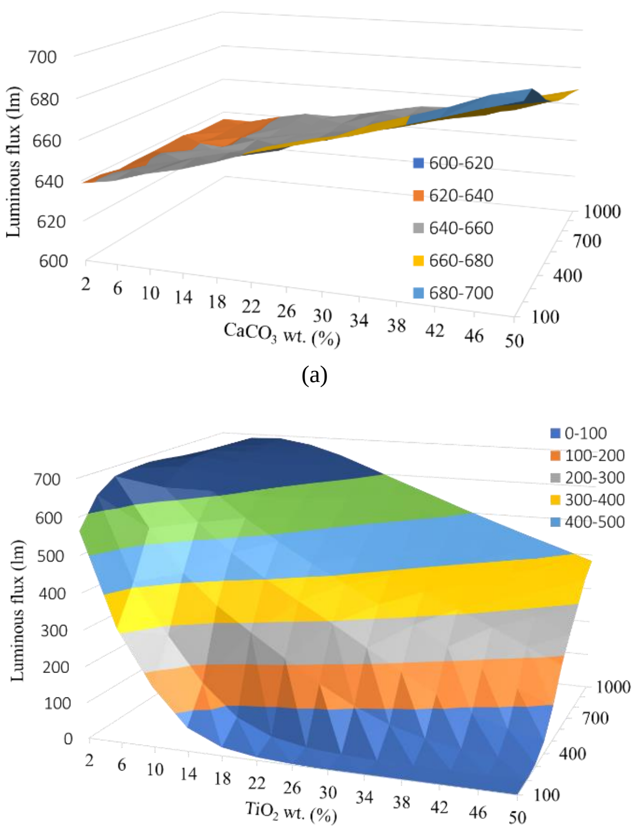


(a)



(b)

Fig. 3. The fluctuation of CCT deviation in pcLEDs employing (a) $CaCO_3$ and (b) TiO_2 (color online)



(a)

(b)

Fig. 4. Luminous flux yielded from pcLEDs with (a) $CaCO_3$ and (b) TiO_2 (color online)

We based on the SEP scattering properties determined using the Mie-scattering theory to validate the fluctuation of luminous flux with $CaCO_3$ and TiO_2 in both cases. The following equation can be used to calculate the scattering cross-section C_{sca} for spherical particles, and the transmitted light power can also be calculated in combination with the Lambert-Beer law [21]:

$$I = I_0 \exp(-\mu_{ext} L) \tag{13}$$

I_0 is the incident light power in this formula, L is the thickness of the phosphor layer (mm), and μ_{ext} is the extinction coefficient that can be calculated by:

$\mu_{ext} = N_r.C_{ext}$, with N_r as the particles' number density distribution (mm^{-3}). The extinction cross-section of phosphor particles is C_{ext} (mm^2). It can be inferred that the rise in the concentration of SEPs results in a lower luminous flux of WLEDs by applying Equation 5. The excessive scattering in the phosphor layer allows backscattering to cause light loss, thus damaging the energy transmitted. The scattering events in the phosphor package, meanwhile, have a strong relation to the SEP concentration. In other words, the scattering increases as the concentration of the SEP increases and becomes excessive if the concentration grows above a certain level. It can be inferred from these two reasons that the greater the SEP concentration, the lower the luminous efficiency.

4. Conclusions

In this study, the study of $CaCO_3$ and TiO_2 effects on color uniformity and light output has verified their advantages in enhancing these two optical properties. In particular, a decrease in CCT deviation is initiated by the increase in $CaCO_3$ and TiO_2 concentrations, yet TiO_2 is superior since it can minimize the deviated color temperature to a lower value than $CaCO_3$. Meanwhile, as the concentration of $CaCO_3$ is around 30%, the color deviation can be decreased by 620 K. In terms of luminous output, $CaCO_3$ is the better SEP as it can increase the efficiency by increasing the utilized concentration. It is advisable to keep the concentration of $CaCO_3$ not exceeding 30% to prevent the reduction in luminescence when applying this SEP with small sizes. On the other hand, if TiO_2 concentration rises above 10%, the pc-LED luminous flux will be adversely affected and will result in a plunge. Therefore, when using $CaCO_3$ and TiO_2 for the optical property management of the manufactured pc-LED, it is important to control the SEPs concentration. With these results, $CaCO_3$ particles tend to be more ideal for pc-LEDs manufacturing with high luminescent properties, while TiO_2 particles are more appropriate for pc-LEDs requiring high color uniformity.

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