

THE APPLICATION OF CALCIUM CARBONATE CaCO_3 AND TITANIA TiO_2 FOR COLOR HOMOGENEITY AND LUMINOUS FLUX ENHANCEMENT IN PC-LEDs

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Abstract. This article studies the development method of pc-LED, a phosphor converted lighting emitting diode, with scattering enhancement particles (SEPs) at 7000 K correlated color temperature. The pc-LED is an advanced lighting solution that has been applied in many different categories; nonetheless, to keep up with the demands of modern lighting, the pc-LEDs need to enhance the color homogeneity and luminous flux. The detailed experiments on the two SEPs used in the articles are also presented. The experiments include combining each of these SEPs with a yellow phosphor $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ to test their properties and influences on the lighting of pc-LEDs. The scattering coefficients, the anisotropic scattering, the reduced scattering, and the scattering amplitudes at 450 nm and 550 nm are the subjects of SEPs study. The LightTools program is used to create the simulation of pc-LED, the results of the optical simulation will then be verified with the Mie-scattering theory. The findings of the research conclude that TiO_2 particles are the best for the growth of color homogeneity while CaCO_3 particles are effective in limiting the color deviation in correlated color temperature. Even though the SEPs benefit the lighting performance, their concentration must be managed to be under an acceptable

amount to ensure desired results and avoid unwanted damages.

Keywords

White LED, Lambert-Beer law, color rendering index, luminous efficacy.

1. Introduction

The pc-LED with formidable characteristics such as high reliability, cost and energy efficiency, and environmental friendliness has earned much attention in the lighting industry in recent years. This lighting solution possesses good luminous efficiency but poor color quality. With the constant growth in demands of lighting applications, it is crucial for pc-LED to achieve a new development in its optical performance. Therefore, the three most important elements in pc-LED production that directly affect the quality of light output are the main concerns [1, 2]. The most common method to fabricate white light is to blend the yellow light from the mixture of yellow phosphor $\text{YAG}:\text{Ce}^{3+}$ and silicone glue with the blue light from the

LED chips. The blue light from the chips will be absorbed by yellow phosphor, thus, creating white light with the desired color temperature [3]. However, while the yellow light tends to grow stronger after absorbing the light from the scattering process, the blue light becomes weaker every time it is absorbed. The mismatch between the blue light and emitted yellow light causing the color distribution to be inconsistent [4, 5]. The issue that can normally arise from the incoherent color distribution is the yellow ring phenomenon, where there is a yellow ring surrounding the perimeter of the pc-LED, causing discomfort to the viewers' eyes. There are many ways to address the deviated color issue, first, we can utilize the differences in range and characteristics of wavelengths in the phosphor to adjust the spatial color homogeneity of pc-LEDs. Besides, the phosphor-in-glass (PiG) with SiO_2 , B_2O_3 , PbO , YAG:Ce^{3+} particles mixed with silicone glue can also reduce the color deviation at an average correlated color temperature of 6000 K from 761 K to 171 K [6]. $\text{HfO}_2/\text{SiO}_2$ DBR film, on the other hand, can lower the color deviation from 1758 K to 280 K at 5000 K CCT [7]. Finally, the micro-patterned is also effective by deducting about 441 K color deviation value at 5337 K CCT [8]. The effects of these mentioned methods in boosting spatial color homogeneity are profound and have been tested, however, the complexity in fabrication as well as the high manufacturing cost prevent them from being widely used. As a result, TiO_2 , ZrO_2 , microspheres, and SiO_2 are the more practical SEPs configuration for the development of a new phosphor compound [9, 10]. There are many research results pointing out the positive impacts of these SEPs on lighting performance of pc-LED, specifically, the Titania (TiO_2) particles, when presents in the phosphor layer at 0.1%, can increase the color quality [11-13]. Also, in regards to the magnification of color homogeneity, the addition of CaCO_3 up to 10% can produce a considerable increase in this property [14]. Moreover, SiO_2 particle is a special SEP that can provide a control over color uniformity when applied to the phosphor layer of pc-LEDs, boost the color quality when placed in a suitable in the phosphor-silicone compound, and adjust the color temperature by changing the particles size [15]. The results all pointed out that SEPs are

good for the development of pc-LED, yet which SEP is the best is still in question. Therefore, this research employs CaCO_3 and TiO_2 , which are frequently used in pc-LED, for the effectiveness in enhancement tests and then compares the results to pick out the most effective SEP. As the results of previous studies suggested, when giving SEPs the right particle size and concentration, they can enhance the lighting performance, therefore, the parameters that show how the SEPs influence the two lighting properties of pc-LED are also computed using Mie scattering theory and presented within the article. In this paper, the WLED package used for experiments has the correlated color temperature of 7000 K, not 8500 K as the previous one [16]. Moreover, the analysis of scattering property was presented and discussed in more detail to demonstrate the enhancement in color uniformity and lumen output. This is the first time using SEPs has been deeply investigated via the simulated results of 4 scattering properties based on the Mie-scattering theory. This is the new point of this paper. This article developed the idea presented in the previous one by providing the investigated information based on Mie-theory, which has not been demonstrated in previous papers. Specifically, it focused more on analyzing the application of two SEP particles (CaCO_3 and TiO_2). Therefore, the achievements of this paper are a valuable reference for manufacturers. From the attained results of this paper, the manufacturers can choose the appropriate SEP with corresponding concentration and particle size to improve the color quality and lumen performance of LED products. This article presents these experimental results in 2 sections with section 2 demonstrating the scattering amplitude of pc-LED, and section 3 presenting the equations used to calculate optical parameters and comparing the achieved results. Section 4 concludes all the findings in the research and provides the information for SEPs application to enhance the lighting performance of pc-LED.

2. Scattering analysis

The MATLAB, a multi-paradigm numerical computing environment, is used to compute

the parameter of the light scattering process. The scattering processes are the results from SEPs coated on pc-LEDs by conformal coating method and calculated based on Mie-scattering theory [17, 18]. The expressions for computing the scattering coefficient $\mu_{sca}(\lambda)$, anisotropy factor $g(\lambda)$, and reduced scattering coefficient $\delta_{sca}(\lambda)$ can be:

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

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

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

In Eq. (1), $N(r)$ is the distribution density of diffusional particles (mm^3) and C_{sca} indicates the scattering cross sections (mm^2). In Eq. (2), $p(\theta, \lambda, r)$ means the phase function, θ exhibits the scattering angle ($^\circ$), and $f(r)$ indicates the size distribution function of the diffusor in the phosphor film and this parameter can be computed by expressions (4) and (5). Besides that, λ presents the light wavelength (nm), and r is known as radius of diffusional particles (μm).

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

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

$N_{dif}(r)$ and $N_{phos}(r)$ included in $N(r)$ are the density of diffusive and phosphor particles, respectively. Meanwhile, $f_{dif}(r)$ indicates the size distribution function data of the diffusor and $f_{phos}(r)$ represents that of phosphor particle. Here, K_N indicates the diffusor unit quantity for one diffusor concentration. The computation of K_N can be carried out as:

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

$M(r)$ here means the mass distribution of the diffusive unit, and can be reckoned by the following equation:

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

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

The scattering cross sections C_{sca} in Mie-scattering theory can be expressed as:

$$C_{sca} = \frac{2\pi}{k^2} \sum_0^\infty (2n-1)(|a_n|^2 + |b_n|^2) \quad (8)$$

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

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

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

with $x = k.r$, m presents the refractive index, $\psi_n(x)$ and $\xi_n(x)$ are the Riccati - Bessel function. Thus, the computations for the relative refractive indices of diffusor (m_{dif}) and phosphor (m_{phos}) in the silicone can be presented as $m_{dif} = n_{dif}/n_{sil}$ and $m_{phos} = n_{phos}/n_{sil}$; and after that, the phase function $p(\theta, \lambda, r)$ are calculated by:

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

in which $\beta(\theta, \lambda, r)$, $S_1(\theta)$ and $S_2(\theta)$ indicate the angular scattering amplitudes whose computations can be expressed as:

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

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

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

In these equations, θ represents the scattering angle indicated by degrees ($^\circ$). a_n and b_n , in turns, are the expansion coefficient with even symmetry and expansion coefficient with odd symmetry. $\pi_n(\cos \theta)$ and $\tau_n(\cos \theta)$ are the angular dependent functions.

Figure 1(a) presents the comparison of scattering coefficient (SC) among different particles sizes of CaCO_3 and TiO_2 . The results show that the bigger the particle diameter is, the higher the scattering coefficient becomes, at the two wavelengths of 450 nm and 550 nm. The scattering coefficient is the parameter used to evaluate the

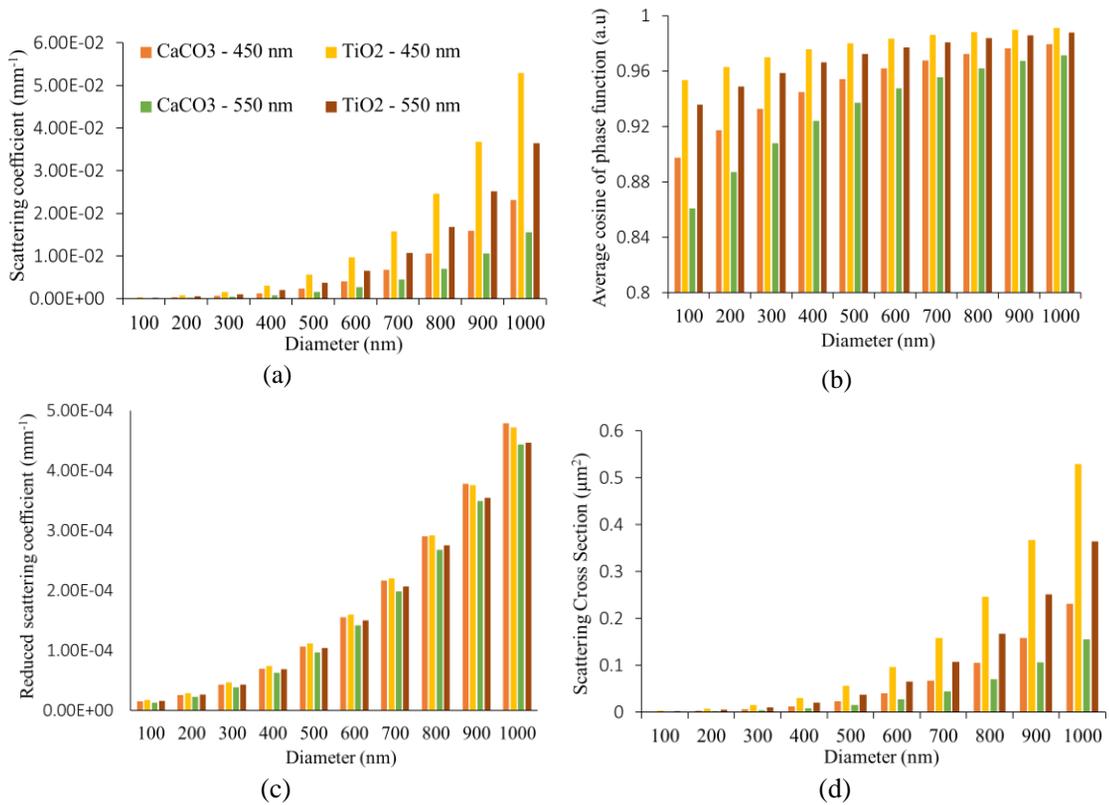


Fig. 1: Comparison of scattering properties in pcLEDs with CaCO₃ and TiO₂.

scattering ability of a phosphor particle. The higher the SC is, the stronger the scattering intensity and the smaller the scattering angle become. Besides, the other scattering properties in Fig. 1(b, c, d) perform the similar trend, the larger particles lead to the higher scattering intensity. It can be also observed that the scattering at the blue-light wavelength (450 nm) is stronger than the light intensity at the yellow wavelength (550 nm). This means the scattering intensity of blue lights is stronger than that of the yellow ones. Moreover, the enhancement of the blue light scattering plays a vital role in minimizing the yellow ring phenomenon. Hence, CaCO₃ and TiO₂ are suitable to address this problem. In particular, SEPs are integrated into the phosphor layer to boost the internal scattering. When the scattering events are multiplied, there are more occurrences in which the blue and yellow lights are mixed. As a result, the generated white light becomes more and more homogenous. Furthermore, the increase in blue

lights at the right and left sides of the phosphor film promotes more blue light extracted. Then, these blue lights combine with the yellowing light to create the white light, which implies that the yellow ring phenomenon is reduced and the color uniformity is improved. As researchers have been working on diminishing the yellow ring, a phenomenon where a circle of yellow light appears on the illuminated surface, the results of this study is important for selecting the right SEPs' concentrations and sizes to enhance the color quality of LED products. The smaller the size of SEP particles, the greater the color quality. This is because using smaller SEP particles results in wide scattering angle and scattering rays, causing the mixing process of lights to be repeated multiple times, compared to utilizing the larger particles. However, this leads to the decrease in lumen output; in other words, the brightness of LED lights is degraded. Thus, the suitable SEP diameter should be chosen depending on the quality requirements. On the other

hand, the concentration of SEPs in the phosphor layer greatly impacts the performance of WLEDs. Specifically, when the concentration is excessive, there will be too much scattering events, and the lumen efficiency is probably reduced.

The simulated results in Fig. 1 support to demonstrated the calculated results in Section 3 to prove the effectiveness of using CaCO_3 and TiO_2 in enhancing the WLED quality. In short, the scattering particles is the core of adjusting WLED color quality and luminous flux. Besides, this study has, for the first time, presented an in-depth analysis of the effects of applying SEPs in WLED structure based on the Mie-theory simulated figures. This has a crucial meaning for WLED companies to choose the right SEPs' parameters for their products.

3. Computation and discussion

The physical model of the pc-LED used in the experiments with different types of SEPs is presented in Fig. 2. The detailed measurements of the lighting performance are computed using the LightTools 8.1.0. The pc-LED model consists of a 0.08 mm phosphor layer and 9 LED chips. The parameters of the reflector are 2.1 mm in depth, 8 mm inner diameter, and 10 mm surface diameter. CaCO_3 and TiO_2 particles are spherical and have the diameter of 0.5 μm approximately, while their refractive indexes are 1.66 and 2.87, respectively. Besides, the yellow phosphor has a refractive index of 1.83 at all wavelengths within the visible spectrum. Meanwhile, the silicone glue used to mix with the SEP has 1.5 refractive index.

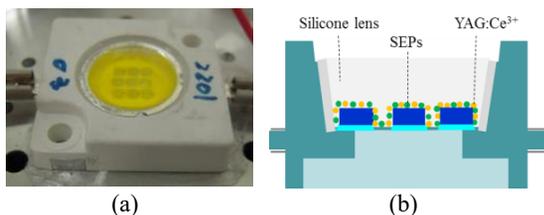


Fig. 2: (a) pc-LED model used in the research, (b) 2D schematic image of pc-LEDs model.

The main purpose of adding a SEP to phosphor packaging of a pc-LED is managing the diffusional particle density to maintain the predetermined correlated color temperature by balancing the weight percentages between the components in the phosphor layer. The following equation can be used to properly adjust the particle density and optimize the CCT and light output:

$$W_{\text{phosphor}} + W_{\text{silicone}} + W_{\text{SEP}} = 100\% \quad (15)$$

Equation (15) demonstrates the constituent weight percentages contributing to the pc-LED phosphor layer, in this equation W_{silicone} is weight percentage of the silicone, W_{phosphor} , is weight percentage of phosphor, and W_{SEP} is weight percentage of SEP. According to the equation, when the concentration of SEPs rises and leads to an increase in the weight percentage of SEP, the weight percentage of the yellow phosphor YAG:Ce^{3+} must decrease the same amount to maintain the average correlated color temperature, which is 7000 K in this research.

CCT deviation, the main reason that causes yellow-ring phenomenon and induces chromatic inhomogeneity, is strongly related to the angles of light emission. Therefore, CCT variation at different emission angles are crucial to assess the light quality of pc-LED and need to be included in the research. To calculate the angular CCT deviation, we can use the following equation:

$$\Delta\text{CCT} = \text{CCT}_{(\text{Max})} - \text{CCT}_{(\text{Min})} \quad (16)$$

where $\text{CCT}_{(\text{Max})}$ represents the maximal CCT at 0° , and $\text{CCT}_{(\text{Min})}$ is the minimal CCT at 90° based on the viewing angle.

As mentioned in previous studies of SEPs, CaCO_3 can reduce deviated color lower than any other SEPs because its light intensity distribution between the scattered blue light and emitted yellow light showed the smallest differences. Since it is noted that CCT deviation is caused by the dissimilarity in optical properties between the phosphor particles within the pc-LED, it is obvious why boosting the amount of emitted blue light to compatible with that of the emitted yellow light can limit the CCT deviation value. The white light is a combination of blue light and yellow light, therefore, the lack of scattered

blue light needed for absorption process results in excessive yellow light and degrades the color quality of pc-LEDs. The more emitted blue light the more redundant yellow light that caused the yellow-ring phenomenon is reduced, thus, resulting in better white light and yellow-ring eradication. These findings in color homogeneity and luminous flux can be useful for their management and improvement.

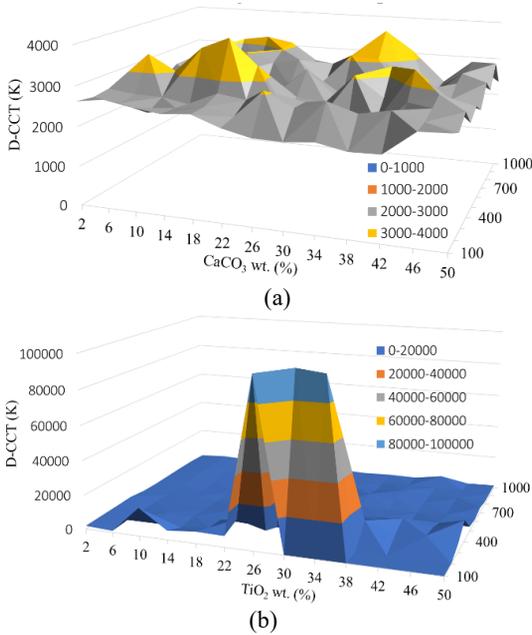


Fig. 3: The fluctuation of CCT deviation in pcLEDs employing (a) CaCO₃ and (b) TiO₂.

As can be seen from Fig. 3 demonstrating CCT deviation of pc-LED configurations with CaCO₃ and TiO₂, both SEPs are effective in reducing the variation of radiant intensity distribution. In the case that CaCO₃ concentration is at 30%, the CCT deviation decreases by 390 K, from 2070 K to 1680 K. The last crucial optical property for pc-LED is luminous flux, which can be assessed from Fig. 4. Figure 4 demonstrates the luminous efficiency of pc-LEDs utilizing CaCO₃ and TiO₂ with different concentrations, from 0% to 50%. In addition, the particle size of them are changed from 100 nm to 1000 nm. The results of the pc-LED with CaCO₃ suggest that the concentration and particle size of this SEP greatly affect the lighting performance. Specifically, the scattering of light is degraded as the particle size increases while

the luminous flux gets benefits from this diameter increase. In the case of TiO₂, the increase of luminous flux is from 0 – 10%, however, any increase in concentration of TiO₂ at any particle size may cause the luminous flux to decline.

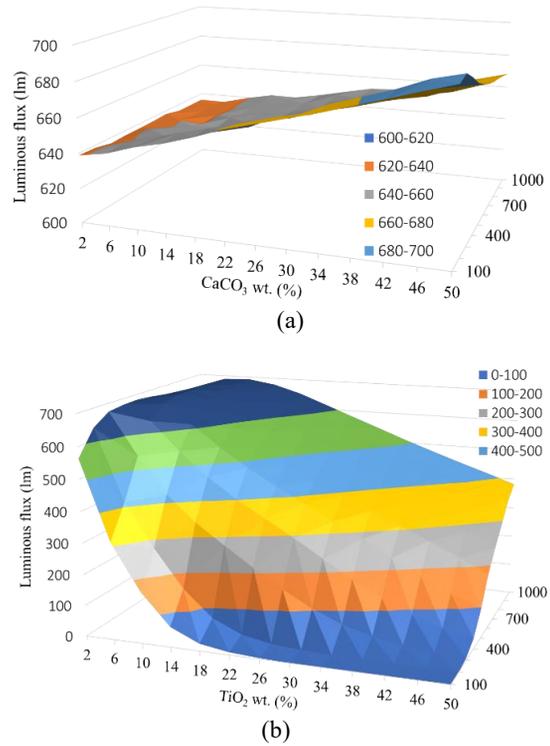


Fig. 4: Luminous flux yielded from pcLEDs with (a) CaCO₃ and (b) TiO₂.

To verify the fluctuation in luminous flux in both cases with CaCO₃ and TiO₂, we based on the SEP scattering properties computed using the Mie-scattering theory. The equation below can be used to calculate the scattering cross section C_{sca} for spherical particles, and in combination with the Lambert-Beer law [18], the transmitted light power can also be determined:

$$I = I_0 \exp(-\mu_{ext}L) \tag{17}$$

In this formula, I_0 is the incident light power, L is the phosphor layer thickness (mm) and μ_{ext} is the extinction coefficient, which can be computed by: $\mu_{ext} = N_r \cdot C_{ext}$, with N_r as the number density distribution of particles (mm⁻³). C_{ext} (mm²) is the extinction cross-section of phosphor particles. Through the application of Eq. (5), it can be concluded that the increase

in concentration of SEPs results in the lower luminous flux of WLEDs. The excessive scattering in the phosphor layer induces light loss from backscattering, thus, damages the transmitted energy. Meanwhile, the scattering events in the phosphor package has a close connection with the concentration of the SEP. In other words, the scattering increases when the concentration of the SEP increases and becomes excessive if the concentration rises over a certain level. According to these two reasons, it can be concluded that the higher the SEP concentration, the lower the luminous efficiency.

4. Conclusions

The study of CaCO_3 and TiO_2 influences on color uniformity and light output in this research verified their benefits to the improvement of these LED optical properties. In particular, the rise in CaCO_3 and TiO_2 concentrations initiates a decrease in CCT deviation, especially, TiO_2 is superior as it can reduce the deviated color temperature to the lower value, compared to CaCO_3 . In the case of CaCO_3 , when the concentration of this SEP reaches 30% the color deviation can be reduced by 390 K. On the other hand, if the concentration of TiO_2 increases to this level, the luminous flux of pc-LED will be negatively affected and result in a plunge. Therefore, it is important to manage the SEPs concentration when employing CaCO_3 and TiO_2 for the optical property management of the manufactured pc-LED. The optical properties of CaCO_3 and TiO_2 particles are computed and verified using the Mie scattering theory to ensure accuracy and reliability. With these results, it seems that CaCO_3 particles are more suitable in manufacturing high-performance pc-LED that is compatible with the specifications of advanced lighting applications.

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