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Utilizing $CaCO_3$, CaF_2, SiO_2, and TiO_2 phosphors as approaches to the improved color uniformity and lumen efficacy of WLEDs

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ABSTRACT

The two elements that are most favorable in the quality evaluation for phosphor-converted LEDs (pcLEDs) these days are the chromatic homogeneity and the lumen output. In this study, a thorough research on enhancing color uniformity and luminous flux of pcLEDs that have a high correlated color temperature (CCT) of 8500K is carried out. The scattering enhancement particles (SEPs): $CaCO_3$, CaF_2, SiO_2, and TiO_2 are used to accomplish the goal by adding them to a yellow phosphor compounding $Y_3Al_5O_{12}:Ce^{3+}$, and comparing their characteristics afterwards. LightTools program is used to build an optical simulation and Mie-scattering theory helps to examine the achieved results. Specifically, the parameters included in SEPs' scattering calculation are the scattering coefficients, the anisotropic scattering, the reduced scattering, and the scattering amplitudes at 455 nm and 595 nm. The outcomes presented that compared to other SEPs, TiO_2 particles can yield the highest chromatic homogeneity. However, the lumen output reduces considerably as TiO_2 concentration greatly increases while it can be bettered when using SiO_2 particles with any particle size. For $CaCO_3$ particles, the color deviation of 620 K CCT can be reduced with 30% concentration, leading to the recommendation of using $CaCO_3$ to promote the CCT homogeneity and luminescence efficiency.

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1. INTRODUCTION

The increase in internal scattering of phosphor-converted LEDs (pcLEDs) is considered as a crucial requirement for getting their quality improved. Thus, three optical properties consisting of chromatic uniformity, lumen efficiency, and color rendering ability (CRI) must be brought into a sharp focus [1, 2]. Generally, a typical pcLED is comprised of blue chips, the yellow $Y_3Al_5O_{12}:Ce^{3+}$ phosphor, and the silicone substance. The process of simulating the yellow light and $Y_3Al_5O_{12}:Ce^{3+}$ absorbing the blue lights produced by the blue chips will generate the white light having desired color temperature [3]. Besides, that the scattered-blue-light radiant intensity distribution is different from the phosphor-emitting yellow-light one results in the non-uniformity of spatial color distribution [4]. This also causes the yellow ring phenomenon to happen to the phosphor-converted LEDs, which discomforts the human eyes. Particularly, in the time the scattering events occur, the energy of the blue photons is degraded when being absorbed by the phosphor but the converted yellow light power tends to go up after every scattering. However, the phosphor layer has completely different wavelength bands and wavelength characteristics, and this helps to modify the spatial color uniformity of pcLEDs in a more advantageous way. A phosphor-in-glass comprised of SiO_2, B_2O_3, PbO, $Y_3Al_5O_{12}:Ce^{3+}$ phosphors, and the silicone is applied to lessen the variance of 6000 K average correlated color temperature (CCT) from 761 K to 171 K [5]. Another method introduced to minimize the color deviation is the HfO_2/SiO_2 DBR film whose result was the reduction from 1758 K to 280 K at around 5000 K CCT [6]. Besides that, the remote micro-patterned phosphor film could reduce the color deviation by 441 K at an ACCT of 5537 K [7]. In general, these phosphor geometries actually resulted in better spatial color homogeneity yet they are complicated to fabricate and also have high production cost. Hence, using scattering enhancement particles (SEPs), including TiO_2 [8], ZrO_2 [9], microspheres [10], and SiO_2 [11, 12], has been considered as a more practical approach. Each SEP was combined with the yellow phosphor grains to make an advanced phosphor compounding. Diffusers of pcLEDs created from the application of the titania (TiO_2) particles showed positive result in color uniformity enhancement, which is that when adding 0.1% TiO_2 to the encapsulation layer case, the chromatic homogeneity can achieve a higher level [13]. Continuing working on the methods to promote the color uniformity, $CaCO_3$ particles were proposed to serve the purpose of improving the scattering properties inside pcLEDs; and the outcome demonstrated a sharp increase in the spatial chromatic homogeneity when using 10% concentration of $CaCO_3$ [14]. Besides the mentioned SEPs, SiO_2 particles are also known as a good material in managing the spatial chromatic homogeneity of pcLEDs. Moreover, there are some pieces of evidences proving that SiO_2 position in the silicone film and the chromatic quality have a rational relation. In addition, their particles sizes considerably impact the pcLEDs' color temperature [15].

Though previous studies emphasized and confirmed the immense influences of SEPs in advancing the chromatic quality of pcLEDs, there is a remaining issue: which one will be the most suitable material for pcLEDs to accomplish better brightness and color homogeneity. Moreover, these aforementioned papers presented that besides increasing color quality by reducing the difference in CCTs, SEPs also decrease the lumen efficacy of the pcLEDs having low CCT and simple single-chip design. Literally, when SEPs are applied with appropriate concentrations and particle sizes, they can improve the effectiveness of the lighting output.

In this article, the common SEPs applied in high-quality pcLEDs production, including $CaCO_3$, CaF_2, SiO_2, and TiO_2, are examined to figure out their benefits and drawbacks, from which a suitable selection of SEPs can be decided to meet the requirements of manufacturers. Besides that, the explanations of the way SEPs can better the two important optical characteristics of pcLEDs are provided with the support from Mie-scattering theory, which are demonstrated in the following sections. Section 2 presents the internal scattering analysis for pcLEDs. Section 3 details the optical experiments carried out for reaching the study's objectives and also discusses the simulation results. The last section, section 4, shows the conclusion of the article.

2. SCATTERING ANALYSIS

Considering the pcLEDs with conformal phosphor structure and based on Mie-scattering theory [16-18], the influence of light scattering occurring when SEPs are added to the structure can be computed with calculation tool MATLAB. The scattering computation of SEPs includes the scattering coefficient $\mu_{sca}(\lambda)$, the anisotropy factor $g(\lambda)$, the reduced scattering coefficient $\delta_{sca}(\lambda)$, and the scattering amplitude functions $S_1(\theta)$ and $S_2(\theta)$, all of which are expressed as follows:

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

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

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

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n(x, m)\pi_n(\cos\theta) + b_n(x, m)\tau_n(\cos\theta)] \quad (4)$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n(x, m)\tau_n(\cos\theta) + b_n(x, m)\pi_n(\cos\theta)] \quad (5)$$

in which $N(r)$ indicates the density distribution of diffusional particles (per cubic millimeter). The scattering cross-section presented by C_{sca} is measured in square millimeters. $p(\theta,\lambda,r)$ represents the phase function where λ (nm), r (μm), and θ (in degrees) are the incident light wavelength, the particle radius, and the scattering angle, respectively. The size distribution of SEPs in the phosphor film is $f(r)$. x means the size parameter, and m shows the index of refraction. a_n demonstrates the expansion coefficient with enven symmetry while b_n is odd-symmetry expansion coefficient. The other parameters: $\pi_n(\cos\theta)$ and $\tau_n(\cos\theta)$ are the angular dependent functions.

Figure 1 and Figure 2 exhibit the increase of scattering coefficients which is dependent on the concentration of used SEPs. Moreover, they also imply that when SEPs are added with high concentration, the blue light absorption tends to be higher. As can be seen in two figures, $CaCO_3$ is the one getting the scattering coefficient reached the highest point, compared to the other SEPs. Besides that, the dissimilarity of scattering coefficients between 455 nm and 595 nm is reduced to the minimum number as $CaCO_3$ appears in the phosphor structure, which means $CaCO_3$ can balance the blue and yellow photons' intensity distributions. Also, in the bar charts of Figure 1 and Figure 2, the anisotropy scattering $g(\lambda)$ values are presented based on the calculated results from (2). Unlike the scattering coefficients, the changes of $g(\lambda)$ are not dependent much on the variance of SEPs concentration. Furthermore, the growth in $g(\lambda)$ following the density of SEPs density is very slight which is not significant enough to be considered.

Besides that, it is easy to see that the reduced scattering coefficients $\delta_{sca}(\lambda)$ of SEPs at 455nm have a tiny difference from that at 595nm, for example, $CaCO_3$ grains have g (595 nm) = 0.9 while their g (455 nm) = 0.88, the gap is just 0.02. In other words, the $\delta_{sca}(\lambda)$ between 455nm and 595nm are quite equal. Thus, applying the stability of $CaCO_3$ and TiO_2 scattering ability will make the enhancing process of the spatial chromatic homogeneity become easier and simpler.

Figure 3 shows SEPs' angular scattering amplitudes calculated by MATLAB program. Obviously, SEPs have tremendous benefits to the scattering of blue rays as they provide a sufficient compensation to the blue light, minimize the effect of yellow ring phenomenon, and improve the emitted luminous flux. Nonetheless, to achieve the best result, it is crucial to select the SEPs having relatively equal angular scattering amplitudes between the blue (455 nm) and yellow (595 nm) lights. Considering the angular scattering amplitudes at 455 nm and 595 nm between $CaCO_3$ and SiO_2 particles, $CaCO_3$ results in a smaller difference than SiO_2 does, and this outcome could be observed in Figure 3.

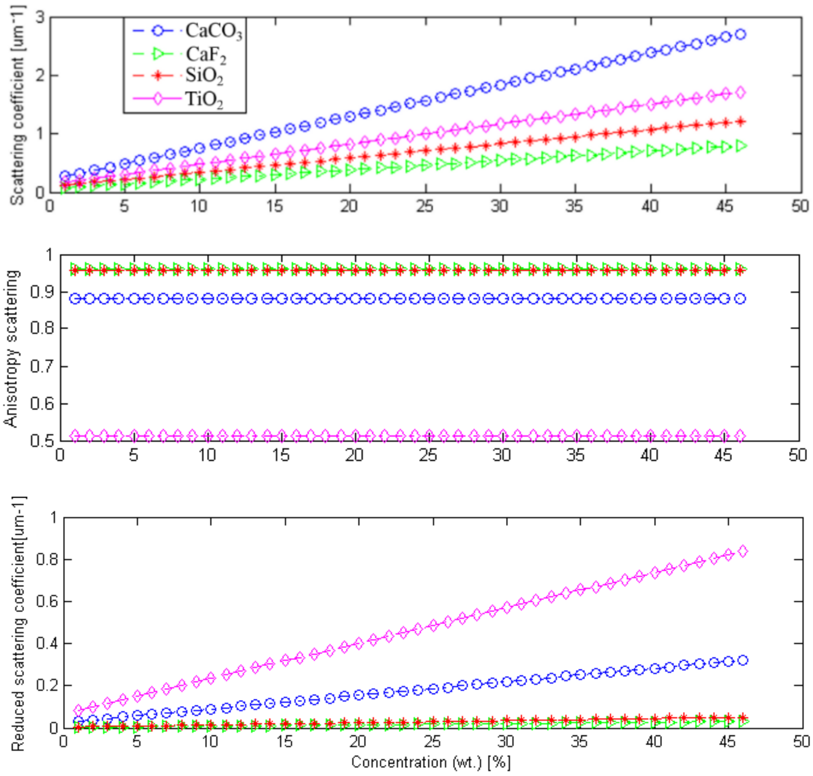


Figure 1. Computation of scattering coefficient, anisotropic scattering and reduced scattering coefficient of the SEPs at 455 nm

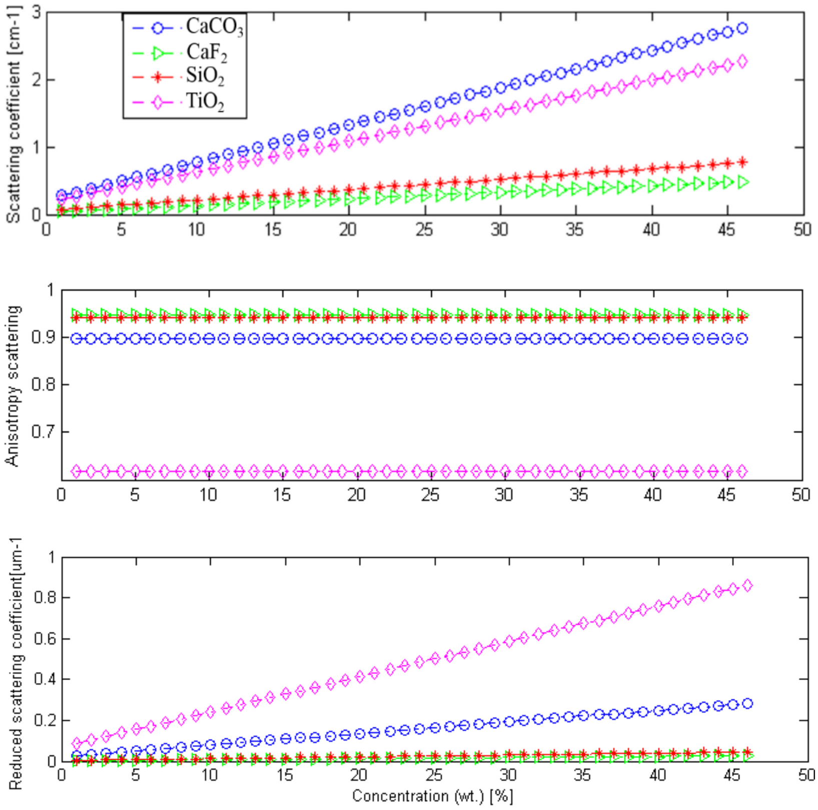
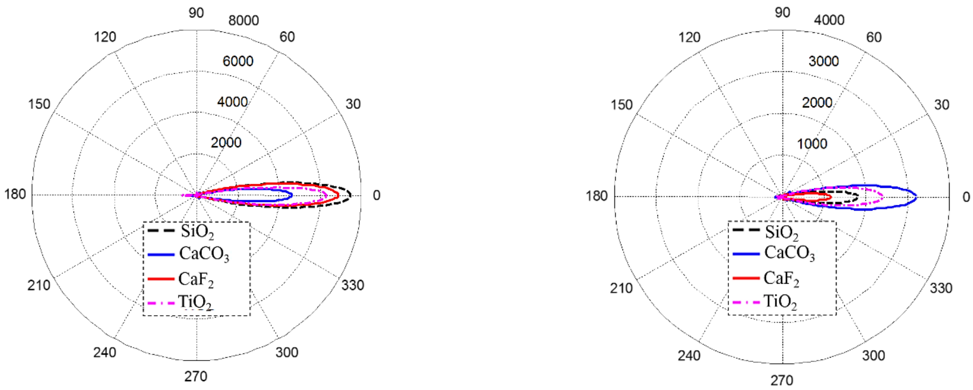


Figure 2. Computation of scattering coefficient, anisotropic scattering and reduced scattering coefficient of the SEPs at 595 nm



(a) (b)

Figure. 3. Angular scattering amplitudes of different SEPs at (a) 455 nm and (b) 595 nm

3. COMPUTATION AND DISCUSSION

Presented in this part is the computed results about the optical performances of pcLEDs when SEPs are applied which are verified by LightTools 8.1.0 software. Moreover, there are some discussions on the impacts of those SEPs on the lighting outcomes of pcLEDs to find out the suitable SEPs for each requirement of producers. In Figure 4 is the schematic diagram of pcLEDs. The input parameters of each element of a pcLED structure must be determined and constant. The reflector of it has the approximate measurement of 2.1 mm, 8 mm, and 10 mm for depth, inner diameter, and outer diameter, respectively. The phosphor layer placed over nine LED chips has 0.08 mm fixed thickness, approximately. For the SEPs examined in this research, including $CaCO_3$, CaF_2, SiO_2, and TiO_2, they are supposed to be spherical particles having 0.5 μm radius. Each of them has different refractive indexes, specifically, 1.66 for $CaCO_3$, 1.44 for CaF_2, 1.47 for SiO_2, and 2.87 for TiO_2. Meanwhile, the yellow phosphor's average radius and index of refraction are set at 7.25 µm and 1.83, respectively, in the visible wavelength range; additionally, 1.5 is the refractive index of the silicone material. Besides that, the density of the diffusional particle is variable parameter for the optimization of CCT homogeneity as well as luminescence efficacy [19, 20].

$$W_{phosphor} + W_{silicone} + W_{SEP} = 100\%. \tag{6}$$

As can be seen in (6) the weight percentage of each material: yellow phosphor ($W_{phosphor}$), silicone ($W_{silicone}$), and SEP (W_{SEP}) in the pcLED phosphor film must be modified to maintain the stability of the average CCT (ACCT). In particular, the weight percentage of $Y_3Al_5O_{12}:Ce^{3+}$ yellow phosphor must be decreased when W_{SEP} increases to keep the ACCT at 8500K. Moreover, it is vital to determine the amount of variance among CCT values at different angles so as to make it easier to assess the quality of illumination with solid-state lighting technology. In fact, when there is a sharp angular deviation in CCT [21, 22], the yellow ring phenomenon occurs, leading to the inhomogeneous color of white lights at different angles. The computation of CCT deviation dependent on the viewing angles can be expressed as:

$$\text{D-CCT} = CCT_{(Max)} - CCT_{(Min)} \tag{7}$$

$CCT_{(Max)}$ and $CCT_{(Min)}$ are the maximum and minimum CCTs the viewing angles of 0^0 and 90^0, respectively.

Clearly, the variation in optical properties of pcLED are caused by the scattered light diversity of each particle applied in the structure. The key to the reduction of this CCT deviation is the sufficient amount of scattered blue lights. Back to Figure 3, it is obvious that among these SEPs, $CaCO_3$ gives the angular-dependent scattering amplitude the smallest deviation between 455 nm and 595 nm. In other words, using $CaCO_3$ can minimize the radiant intensity distribution differences of the blue lights scattered and the yellow lights emitted by the phosphors to the smallest value. In addition, similar to the other SEPs, $CaCO_3$ grains have higher angular scattering amplitude at 455 nm than at 595 nm. The white lights are generated when phosphor-scattered blue rays blend with phosphor-converted yellow lights and also the yellow ring, and this process remarkedly lessens the effect of the yellow ring phenomenon for pcLEDs. However, a shortage or excess of scattered blue lights in pcLEDs will be followed by a larger CCT deviation. Figure 3 also shows that $CaCO_3$ accomplishes a thin line of difference between 455 and 595 nm angular scattering amplitudes which is nearly three times smaller than that of the other SEPs. Thus, using $CaCO_3$ is more advantageous to keep the uniformity of white light color and the luminous performance under control.

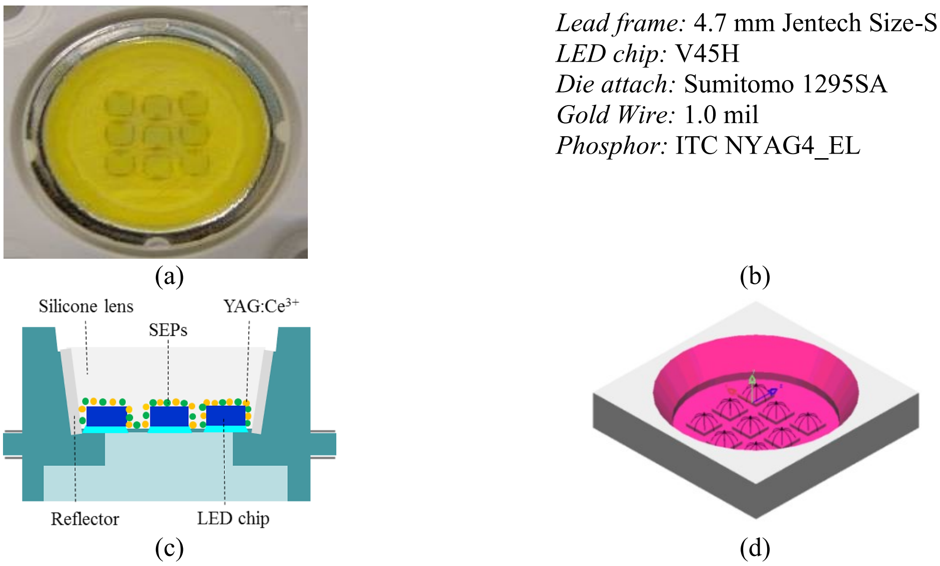


Figure 4. (a) Photograph of WLEDs sample, (b) Manufacturing parameter of WLEDs, (c) Illustration of 2D WLEDs model, (d) the simulated WLEDs model

All key points in discussions are demonstrated in Figure 5. As can be seen, $CaCO_3$ and TiO_2 result in the decrease of the CCT deviations. Additionally, the CCT deviation obviously declines from 2670 K to 2050 K when adding 30% $CaCO_3$, which means a reduction of 620 K CCT deviation can be accomplish with 30% concentration of $CaCO_3$, compared to that without using any SEP. In contrast, the CCT deviations tend to go up when CaF_2 and SiO_2 are applied. The trends of lumen efficiency in accordance with the concentrations and the particle sizes of $CaCO_3$, CaF_2, SiO_2, and TiO_2 are exhibited in Figure 6. The range of concentration for these particles in the experiment is 0-50%, while the diameter of those SEPs varies in the range of 100-1000 nm. In the cases of $CaCO_3$ and SiO_2, the luminous fluxes rise as their concentrations and particle sizes increase. However, with CaF_2, the lumen output increases when the concentration of CaF_2 varies from 0 to 20% at all particle sizes. Then, as CaF_2 concentration surpasses 20% the flux starts to fall down regardless of the particle sizes. Obviously, the bigger SEP diameter leads to the higher degradation of scattering events in the phosphor film, which probably lifts up the luminous flux of pcLEDs. The trend of luminous flux in TiO_2 case is similar to the results of using CaF_2, which is going up in the TiO_2 concentration range from 0 to 10% and after that going down steeply along with higher concentration, at any particle size. The downward trend in luminous flux of SEPs is demonstrated by the Lambert-Beer law and the Mie-scattering theory.

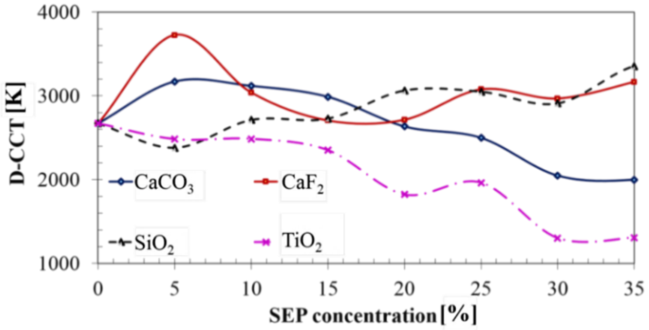


Figure 5. Comparison of CCT deviation of pcLEDs using different SEPs

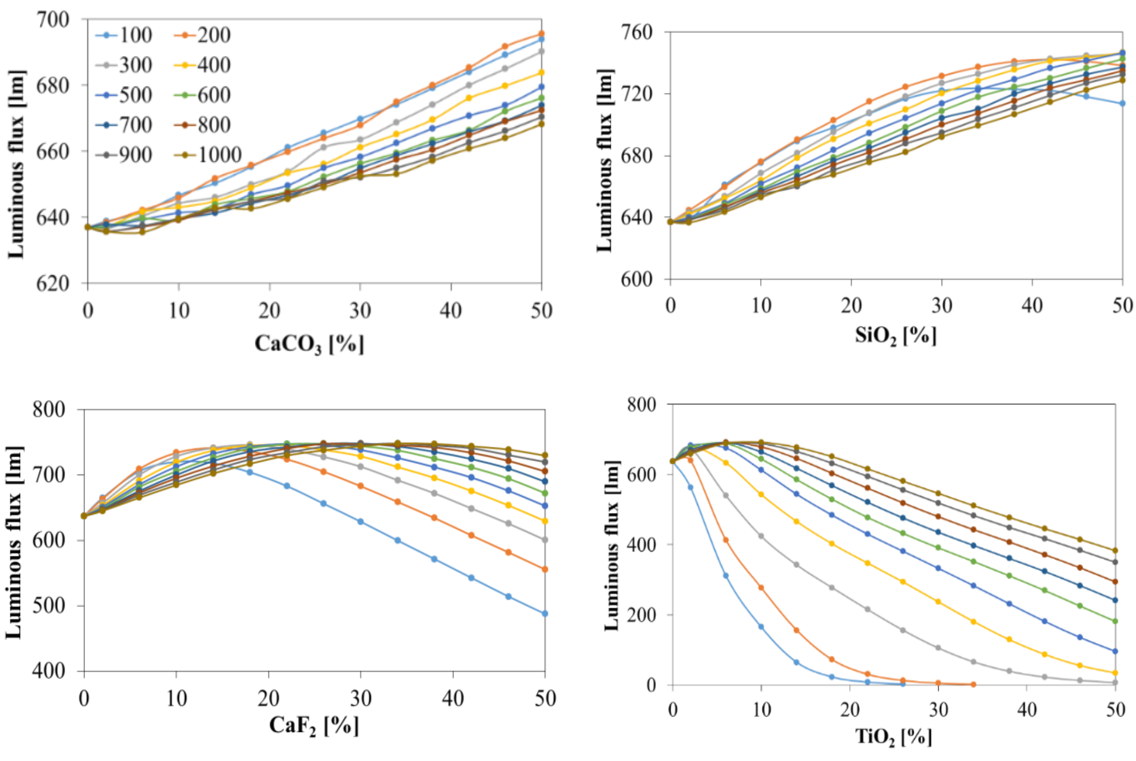


Figure 6. Comparison of luminous flux of pcLEDs using different SEPs

The downward trend in luminous flux of SEPs is demonstrated by the Beer's law and the Mie theory of scattering. Specifically, the Mie-theory is applied to examine and compute the scattering ability of SEPs and the spherical-particle scattering cross section C_{sca}. Meanwhile, the Lambert-Beer law [23-25] is responsible for calculating the power of transmitted lights, which is expressed as:

$$I = I_0 \, exp(-\mu_{ext} L) \quad (8)$$

I_0 indicates the incident light power, L presents the phosphor layer thickness (mm), and μ_{ext} is the extinction coefficient expressed as: $\mu_{ext} = N_r . C_{ext}$, with N_r (mm^{-3}) and C_{ext} (mm^2) are the number density distribution of particles and the extinction cross-section of phosphor particles, respectively. In (8) implies that the enhancement in the luminescence output of pcLEDs follows the growth in the concentration of SEPs. This can be explained by these two reasons: 1) the excess of the scattering events in the phosphorus film is the cause of the decrease in transmitted energy; 2) the scattering improvement relies on the concentration of SEPs.

4. CONCLUSION

This paper has achieved the main purpose of analyzing and demonstrating the influences of SEPs ($CaCO_3$, CaF_2, SiO_2, and TiO_2) on the performance of the color uniformity and luminous output of pcLEDs. Mie-scattering theory is used to examine the ability of each SEPs, and the results show that the scattered lights obtain a considerable improvement as SEPs are in the pcLED's phosphor structure. Thus, with this useful finding, the work of enhancing the performance of pcLEDs is limited to the focus on figuring out the appropriate SEPs concentrations. This study indicated that when the concentrations of TiO_2 and $CaCO_3$ develop, the CCT deviation can be minimized. TiO_2 particularly is advantageous in reducing the CCT deviation to the minimal value. Yet, the luminous performance tends to decrease steeply as TiO_2 concentration continues to be higher. In contrast, the lumen output shows an upward trend with the concentrations of $CaCO_3$, CaF_2, and SiO_2, especially SiO_2, the one has a tremendous benefit in yielding better lumen values. Besides, $CaCO_3$ is also beneficial to reduce the color deviation, by nearly 620 K, with a concentration of 30%. Therefore, it can be said that the most effective SEP for the optical improvement of pcLEDs is $CaCO_3$. The outcomes of this article give a solid foundation about the use of SEPs and a valuable reference to the work of bettering pcLEDs' performance. Manufacturers can use this for their practical production of pcLED products that fulfill the requirements of the lighting market.

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