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Comparison of calcium carbonate and titania particles on improving color homogeneity and luminous flux of WLEDs

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ABSTRACT

In this paper, the experiments on phosphor-converted LEDs (pc-LEDs) with a correlated color temperature (CCT) of 8500K has been conducted with the scattering enhancement particles (SEPs) to achieve the higher color uniformity and emitted luminous flux of pc-LEDs. Moreover, this paper also introduced about choosing scattering enhancement particles (SEPs), including calcium carbonate ($CaCO_3$) and titania (TiO_2), and compared these particles' properties by adding them into the yellow $Y_3Al_5O_{12}:Ce^{3+}$ phosphor compounding. Afterward, the LightTools program was applied to illustrate the optical simulation, and obtained results was analyzed and verified by applying the Mie-scattering theory. The scattering coefficients, the anisotropic scattering, the reduced scattering, and the scattering amplitudes at 455 nm and 595 nm are included in the scattering computation of SEPs. According to researched results, among the SEPs, TiO_2 particles result in the highest value of color uniformity. However, a rise in their concentration is the cause of a sharp decline in luminous flux. Meanwhile, $CaCO_3$ particles show the ability of reducing the deviated level in correlated color temperature by 620K if there is employed 30% of $CaCO_3$ concentration. Hence, $CaCO_3$ particles are the recommendation for achieving higher chromatic homogeneity and lumen output.

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1. INTRODUCTION

If pc-LEDs are aimed at higher performance, having the scattering event in pc-LEDs enhanced is required. This means there are three crucial optical characteristics must be focused on, consisting of chromatic uniformity, lumen output, and color rendering ability [1, 2]. In fact, a typical method to create a pc-LED is the combination between yellow $Y_3Al_5O_{12}:Ce^{3+}$ ($YAG:Ce^{3+}$) phosphor and silicone glue. Then, the white light having the desired color temperature is created as a result of a process in which the yellow light is stimulated while the exciting blue light is absorbed by the $YAG:Ce^{3+}$ phosphor from the chips [3]. Generally, it is the difference in radiant intensity distributions between phosphor-scattered blue light and the phosphor-emitted yellow light that results in the heterogeneity in the spatial color distribution [4]. In similarity, the existence of a yellow ring in phosphor-converted LEDs of this study can cause human eyes a discomfort. Specifically, the blue light becomes weaker after every time being absorbed by phosphor during the scattering process, while the converted yellow light tends to be stronger after each scattering event. Moreover, there is a complete difference in range and characteristics of the wavelengths in the phosphor layer, and this is one of the factors leading to the more advantageous adjustments for spatial color homogeneity of pc-LEDs. In addition, the usage of the phosphor in glass, containing SiO_2, B_2O_3, PbO, YAG:Ce^{3+} particles and the silicone glue, will contribute to reducing chromatic deviation from 761 K to 171 K at 6000 K average correlated color temperature (ACCT) [5]. Besides that, the fabrication of HfO_2/SiO_2 DBR layer will make a decrease in the deviated level of color from 1758 K to 280 K at a CCT of about 5000 K [6]. Additionally, the application of remote micro-patterned phosphor film can get the color deviation decreased to 441 K at 5537 K ACCT [7]. In short, these phosphor configurations have remarkable benefits in magnifying the spatial color homogeneity. However, the complication and the financial issue have become obstacles in the process of producing these configurations. Therefore, the more realistic configurations SEPs, such as TiO_2 [8], ZrO_2 [9], microspheres [10], and SiO_2 [11, 12], were mixed with the yellow phosphor materials in the purpose of getting novel phosphor compounds generated. Furthermore, the titania (TiO_2) grains were employed as diffusers for pc-LEDs. Specifically, with 0.1% concentration of TiO_2 used in the encapsulation layer case, the color uniformity can perform a higher quality [13]. As a part of efforts to reach the enhancement in color homogeneity, $CaCO_3$ was considered as one of the best suggestions in improving the scattering characteristics of pc-LEDs. As pointed out in the attained results, when there is added 10% of $CaCO_3$, the spatial color homogeneity can be in a sharp increase [14]. What is more, SiO_2 particles are known as a necessary material in putting the spatial color uniformity under control when mixed into pc-LEDs. Besides, as confirmed in some sheds of evidence, the position of SiO_2 particles in the silicone layer rationally relates to the color performance. In addition, the size of SiO_2 particles has also made a significant impact on the color temperature of pc-LEDs [15]. In the studies mentioned above, it is proven that the color performance of pc-LED has been enormously governed by the SEPs. However, the raised question is among different SEPs, which one will be the best selection for achieving better color uniformity and brightness. Additionally, those given pieces of evidence in previous studies have demonstrated the influence of SEPs on simple single-chip packages with low color temperature correlation (CCT) which is the ability in getting the difference in CCT and the luminous efficacy lowered. Actually, if being chosen with appropriate concentrations and sizes, some SEPs can enhance those characteristics of light. Given in this study is a case in which different particles that commonly used to promote the quality of pc-LEDs to a higher state: $CaCO_3$, CaF_2, SiO_2, and TiO_2, are tested for scattering improvement. Moreover, through this research, not only are the best SEP figured out to serve the main purpose, but also the reasons why SEPs can enhance these two optical properties of pc-LEDs are demonstrated based on Mie theory. The specific information for what has been mentioned in this introduction will be given in next sections, which are section 2 the basic scattering analysis of pc-LEDs. Section 3 the optical experiments for achieving the objectives and discussions on the simulation results, and section 4 conclusions for this paper.

2. SCATTERING ANALYSIS

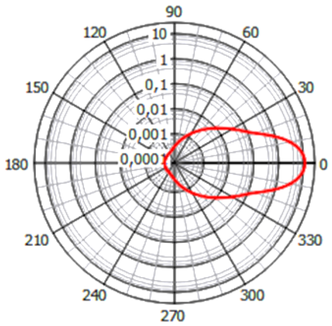
According to Mie-scattering theory, at the time the light scattering occurred as a result of the impact from SEPs in conformal phosphor pc-LEDs, MATLAB will be used as a computing tool in supporting to have the effect determined. As mentioned in previous papers [16-25], the scattering amplitude functions $S_1(\theta)$ and $S_2(\theta)$ can be calculated following these expressions:

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

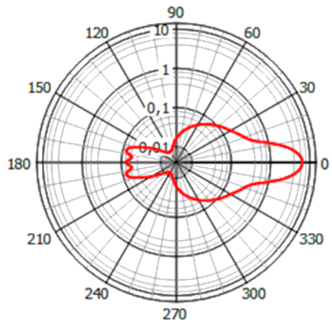
$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n(x,m)\tau_n(\cos\theta) + b_n(x,m)\pi_n(\cos\theta) \right], \tag{2}$$

in which θ presents the scattering angle (degrees), x indicates the size parameter and m is the refractive index. Meanwhile, a_n and b_n are the expansion coefficients with even and odd symmetry, respectively. $\pi_n(\cos\theta)$ and $\tau_n(\cos\theta)$ are known as the angular dependent functions.

From the graph of Figure 1, it is easy to realize that the angular scattering amplitudes of SEPs is reckoned by the MATLAB program. Obviously, with a sufficient blue-light compensation, the reduction of the yellow ring phenomenon, and the rise in the emitted luminous flux, the SEPs have proven their advantages to the blue-light scattering. However, to be able to exploit all the advantages of the SEPs, it is vital to select the appropriate SEPs having angular scattering amplitudes that are approximately equal between blue-light (455 nm) and yellow-light (595 nm). If comparing the $CaCO_3$ and TiO_2 particles, the angular scattering amplitudes of them are significantly discrepant.



(a)



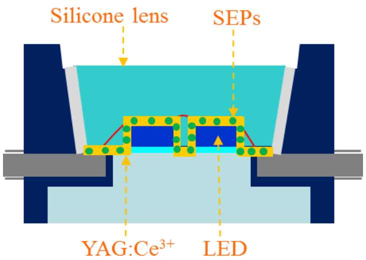
(b)

Figure 1. Angular scattering amplitudes of pc-LEDs adding $CaCO_3$ (a) and TiO_2 (b)

3. COMPUTATION AND DISCUSSION

In this section, the LightTools 8.1.0 software is utilized to calculate the performance of the lighting characteristics of pc-LEDs using different SEPs. Figure 2 is the illustration of a pc-LED's schematic diagram. In this simulation, the parameters of the reflector including depth, inner and outer diameter are approximately 2.1 mm, 8 mm, and 10 mm, respectively.


(a)



(b)

Figure 2. (a) Photograph of WLEDs sample, (b) Illustration of 2D WLEDs model

The nine LED chips are coated with a phosphor layer having 0.08 mm fixed thickness. In addition, the refractive index of $CaCO_3$ is 1.66 while that of TiO_2 is equal to 2.87. Besides, it is assumed that all the SEPs are spherical with a 0.5 μm radius. Meanwhile, a phosphor particle has an average radius of 7.25 μm and a refractive index of 1.83 at all wavelengths in the visible band. The silicone glue has a 1.5 refractive index. The density of diffusion particle is adjusted to optimize the uniformity of CCT and the output efficacy.

$$W_{phosphor} + W_{silicone} + W_{SEP} = 100\%. \tag{3}$$

$W_{silicone}$, $W_{phosphor}$, and W_{SEP} indicate the weight percentages of the silicone, phosphor, and SEP in the phosphor layer of pc-LEDs, respectively. When there is a rise in the weight percentage of SEPs, the $YAG:Ce^{3+}$ phosphor's weight have to be reduced for keeping the average CCT value at 8500 K. In an effort of evaluating the light quality of pc-LEDs, the determination of the number of variations among CCT values at different angles is in need. In this way, it is easier to assess the efficiency of solid-state lighting applications. Specifically, a sharp CCT deviation in connection with angles causes the yellow ring to occur and then generate inhomogeneous color for white lights at many angles. The calculation of angular CCT deviation is expressed:

$$\Delta CCT = CCT_{(Max)} - CCT_{(Min)} \quad (4)$$

in which $CCT_{(Max)}$ is the maximal CCT at 0^0 and $CCT_{(Min)}$ presents the minimal CCT at 90^0 viewing angles.

In fact, that the scattered light of each phosphor particle in optical characteristics of pc-LED is different from each other is the cause of such existing variations. Here is the noticeable point, it is able to greatly reduce this CCT deviation, as long as the blue light scattering is sufficiently promoted. In Figure 1 is the deviated value of angular scattering amplitude of $CaCO_3$ particles which is smaller than that of TiO_2 grains. This means that compared to the other SEPs, when $CaCO_3$ is applied to the phosphor layer, it can optimally reduce the differences in radiant intensity distributions for the scattered blue light and the emitted yellow light of the phosphor particles. After that, the white light is generated by the combination between the scattered blue light and either the converted yellow light or the yellow ring, resulting in reducing the effect of the yellow ring phenomenon in pc-LEDs. In contrast, the deviation in CCT will increase because of the lack or redundancy of scattered blue light in pc-LEDs. Therefore, it is possible to control the color uniformity and the lumen output more advantageously. In Figure 3 all the key points from the aforementioned discussions are summarized. This figure shows that the CCT deviations tend to decline in $CaCO_3$ and TiO_2 cases. It can be seen in Figure 3 that the CCT deviation decreased from 2670 K to 2050 K with the usage of 30% $CaCO_3$, dropped by 620 K compared to the case not using any SEP.

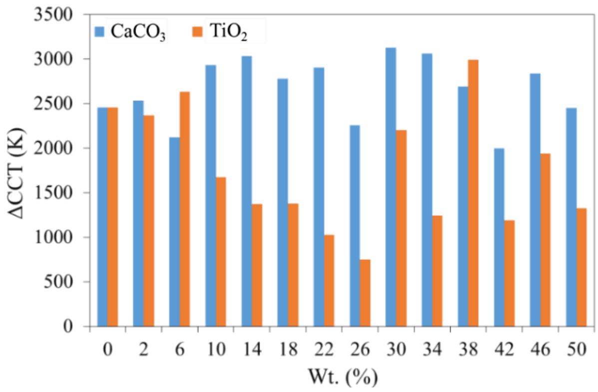


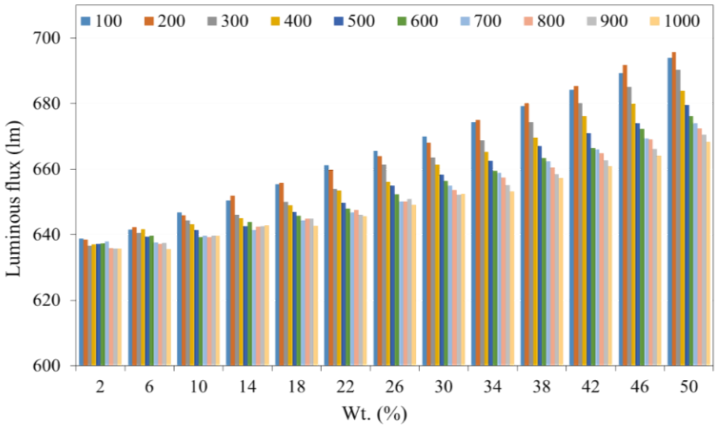
Figure 3. Comparison of CCT deviation of pc-LEDs adding $CaCO_3$ and TiO_2

Figure 4 illustrated the comparison of luminous flux using the different particles $CaCO_3$ and TiO_2 with the corresponding concentration and size. In this experiment, the applied range for the concentration of these SEPs is from 0 to 50%, in regard to the particle size from 100 nm to 1000 nm. In the case of $CaCO_3$, the increase of the observed luminous flux has a close connection with the concentration and particle size, which means its changes depend on the value of concentration and particle sizes. It is undeniable that in consequence of applying the larger particle size, the scattering in the phosphor layer will be lower, and the luminous flux is promoted to the higher performance, in this way. With the existence of TiO_2, the luminous flux performs an increase by about 0-10%, and when the TiO_2 concentration goes up at all particle sizes, it tends to decrease. In accordance with the demonstration of the decrease in the concentration of SEPs, the Lambert-Beer law and the Mie theory are applied.

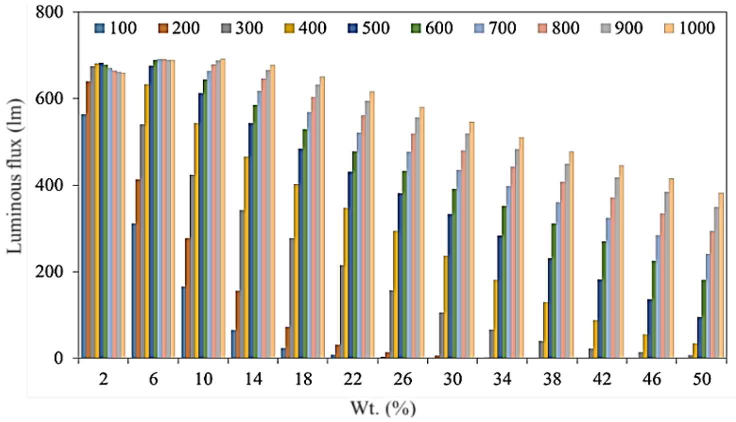
The analysis of SEPs' scattering was conducted with the application of the Mie-theory. In addition, Mie-theory also helps the scattering cross section C_{sca} for spherical particles to be calculated following the formula below. Besides, by applying the Lambert-Beer law, the transmitted light power can be reckoned:

$$I = I_0 \exp(-\mu_{ext} L) \quad (5)$$

where I_0, L, and μ_{ext} are the incident light power, the phosphor layer thickness (mm) and the extinction coefficient, respectively. Additionally, the extinction coefficient μ_{ext} can be calculated as: $\mu_{ext} = N_r.C_{ext}$, in which N_r indicates the number density distribution of particles (mm^{-3}), and C_{ext} (mm^2) presents the extinction cross-section of phosphor particles. In (5) has come up with a conclusion, the higher the concentration of SEPs is, the lower the luminous flux of WLEDs becomes. There are two reasons explaining this result. The first one is that the transmitted energy declines because of excessive scattering in the phosphor layer; and the second one is the dependence of the rise in the scattering on the concentration of SEPs.



(a)



(b)

Figure 4. Comparison of luminous flux of pc-LEDs adding $CaCO_3$ (a) and TiO_2 (b)

4. CONCLUSIONS

In brief, this paper has described and analyzed the effects of $CaCO_3$ and TiO_2 particles on two optical performances of pc-LEDs: chromatic homogeneity and luminous flux. By using the Mie-scattering theory, a noticeable result that each type of SEPs applied to pc-LEDs can significantly enhance the achieved scattered light has been verified. This has formed a crucial basis of keeping the optical properties of pc-LEDs under control with the appropriate concentration of added SEPs. From these attained results, manufacturers could have fundamental and valuable information for applying to practical manufacturing of W-LEDs so that the predefined specifications on lighting quality can be fulfilled. Initially, as the consequence of the rise of $CaCO_3$ and TiO_2 concentration, the CCT deviation will be reduced. TiO_2 is particularly considered as playing a vital role in minimizing the color deviation to the lowest value. However, while rising with the addition of $CaCO_3$ concentration, the luminous flux will present a sharp decline if there is an enormous increase in TiO_2 concentration. On the other hand, the CCT deviation can be dropped by approximately 620 K after 30% of $CaCO_3$ is added. Therefore, $CaCO_3$ particle is considered as the best selection in achieving the goal of improving the optical characteristics of pc-LEDs in general applications to manufacture an expected novel WLEDs generation.

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