Application of CaCO$_3$, CaF$_2$, SiO$_2$, and TiO$_2$ particles to silicone lens for enhancing angular color uniformity of white LED lamp

Anh Q. D. Nguyen, Vinh H. Nguyen
Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam
anhkugas@gmail.com

Abstract. In this study, we present an insightful investigation on optimal selection of scattering enhancement particles (SEP) to satisfy each specific optical property of white LEDs (WLEDs). The interested contenders include CaCO$_3$, CaF$_2$, SiO$_2$, and TiO$_2$, each of them is added with YAG:Ce phosphor compounding. The quality improvement on each considered property is demonstrated convincingly by applying Mie-scattering theory together with Monte Carlo simulation on a particular WLEDs which has the color temperature of 8500K. It is observed by simulation results that TiO$_2$ particles provide the highest color uniformity among the SEP, as increasing TiO$_2$ concentration. These results of this work can serve as a practical guideline for manufacturing high-quality WLEDs.

1. Introduction
To improve the overall performance of WLEDs, previous studies have focused on three important optical properties, including color uniformity, luminous flux, and color rendering ability [1]. All these authors’ approaches met at one point, which is to enhance scattering events in phosphor-converted white-LEDs (pcLEDs). Typically, a pcLEDs is formed by combining YAG:Ce phosphor and silicone glue. The YAG:Ce phosphor absorbs the exciting blue light from the chips to stimulate the yellow light, a process that generates white light with desired color temperature [2]. In general, the phosphor-scattered blue light and the phosphor-emitted yellow light have different radiant intensity distributions. As a result, the spatial color becomes non-uniform [3]. Correspondingly, like the conformal phosphor white LEDs which applied in this study, the phenomenon of yellow ring can appear, resulting in disadvantage of vision, see Fig. 1.
Specifically, the blue light during the scattering process is weakened due to the absorption of phosphors, while the energy of converted yellow light is increased after each scattering. Due to the discrepancy of light behavior varying with wavelengths in the phosphor layer, it is necessary and natural to adjust the color performance of pcLEDs. Up to now, there have been studies that propose solutions to improve the color uniformity of LEDs by adding SEP into pcLEDs [4, 5].
In 2010, the group of Lee mingled titania (TiO$_2$) and the encapsulation layer as well as the phosphor layer. This study proposed used 0.1% TiO$_2$ in the encapsulation layer case for obtaining higher color homogeneity (Lee et al. 2010) [6]. In 2013, with the purpose of color homogeneity enhancement, Yang et al. (2013) applied CaCO$_3$ for enhancing the scattering property of WLEDs. The obtained results indicated that the spatial color homogeneity can be increased with 10% CaCO$_3$, significantly [7].

From the work of Anh et al. [1], it has been known that by mixing SiO$_2$ particles to pcLEDs, the spatial color uniformity can be controlled. Moreover, some evidence has been found to confirm a link between the location of SiO$_2$ particles in the silicone layer and the color performance. The color temperature of pcLEDs has also been influenced strongly by the size of SiO$_2$ particles [2].

The above studies have addressed the SEP that can affect the color performance of WLEDs specifically. However, how to select a suitable material among SEP for higher color homogeneity and higher luminous efficiency is still a query. Moreover, the previous studies indicated that the SEP can decrease both the deviation of correlated color temperature (CCT) and the luminous efficiency of the simple single-chip packages having low CCT. In fact, some SEP can enhance the luminous efficiency with a suitable concentration and size.

In this study, we provide an insightful inspection of various scattering enhancement particles including CaCO$_3$, CaF$_2$, SiO$_2$, and TiO$_2$, which are popularly employed for manufacturing higher-quality WLEDs. The target of this work is not only limited on how to select the best SEP but also demonstrated why the SEP can be done by using the Mie theory and the Mote Carlo simulation. The
rest of this paper is organized as follow. The basic analysis of scattering inside WLEDs is provided in Section 2. The detailed optical experiments to achieve the goals of this paper as well as some discussion on the simulation results are described in Section 3. Finally, Section 4 concludes the paper.

2. Scattering analysis

The light scattering effect caused by the SEP in the used conformal phosphor WLEDs structure is determined by using Mie-scattering theory [8] with the support of MATLAB as a computation tool. Recall that the scattering amplitude functions $S_1(\theta)$ and $S_2(\theta)$ is determined as following can be calculated by the following equations:

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

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

where:

- $x$ is the size parameter
- $m$ is the refractive index
- $\theta$ is the scattering angle (degree)
- $a_n$ and $b_n$ are in turn the expansion coefficients with even symmetry and odd symmetry
- $\pi_n(\cos \theta)$ and $\tau_n(\cos \theta)$ describe the angular scattering patterns of the spherical harmonics

The angular scattering amplitudes of SEP are computed by MATLAB program, see Fig. 2. The results show that SEP are of great advantage to blue-light scattering. As compensating blue-light enough, it is not only reduce the yellow ring phenomenon, but also increase lumen output. This depend on the selection of SEP, which has approximate angular scattering amplitudes between blue-light (455 nm) and yellow-light (595 nm). Unlike SiO$_2$ particles, the angular scattering amplitudes of CaCO$_3$ particles present a smallest difference between 455 nm and 595 nm.

3. Simulation and discussion

In this section, the optical properties of WLEDs that employ different SEP is evaluated by using the LightTools 8.1.0 software. The refractive index of the SEP including CaCO$_3$, CaF$_2$, SiO$_2$, and TiO$_2$ are 1.66, 1.44, 1.47, and 2.87, respectively. All SEP are assumed to be spherical with radius of 0.5 µm. The average radius of a phosphor particle is 7.25 µm and its refractive index is equal to 1.83 at all wavelengths in visible band. The refractive index of the silicone glue is 1.5. The diffusional particle density is varied for optimizing the CCT uniformity and the output efficiency.

$$W_{\text{phosphor}} + W_{\text{silicone}} + W_{\text{SEP}} = 100\%$$  \hspace{1cm} (7)
$W_{\text{phosphor}}$, $W_{\text{silicone}}$ and $W_{\text{SEP}}$ are the weight percentages of the silicone, phosphor and SEP of the phosphor layer in WLEDs, respectively. If the weight percentage of the SEP is increased, the weight of YAG:Ce phosphor needs to be reduced to maintain the mean CCT value at 8500K. To evaluate the light quality of WLEDs, we determine how much different between CCT values in proportion to angles. This is an important standard for evaluating the performance of solid-state lighting applications. A large deviation in CCT values in proportion to angles will cause the yellow ring phenomenon and generate the non-uniform white color at different angles. The angular CCT deviation is calculated as

$$D_{\text{CCT}} = \text{CCT}_{(\text{Max})} - \text{CCT}_{(\text{Min})} \quad (8)$$

where $\text{CCT}_{(\text{Max})}$ and $\text{CCT}_{(\text{Min})}$ represent the maximal CCT at the zero degree and the minimal CCT occurs at the 70 degree of viewing angle, respectively.

In fact, the reason that causes variation in optical properties of WLEDs is due to the difference among the scattered light of each particle in pcLEDs. The key idea is that this CCT deviation can be reduced significantly as long as the scattered blue light is enhanced sufficiently. As displayed in Fig. 2, the deviation of angular scattering amplitude of CaCO$_3$ particles for 455 nm and 595 nm is smallest in comparison with the other SEP. It means that there is a smallest variation in radiant intensity distributions for the phosphor-scattered blue light and the phosphor-emitted yellow light as using CaCO$_3$.

Moreover, like all the applied SEP, the angular scattering amplitude of CaCO$_3$ particles for 455 nm is higher than 595 nm. The scattered blue light not only combines with the converted yellow light, but also with the yellow ring for emitting white light, which would alleviate the yellow ring phenomenon in WLEDs. Conversely, the CCT deviation will become larger under condition of lack or redundancy of scattered blue light in WLEDs. Also indicated in Fig. 2, the deviations of angular scattering amplitude for 455 nm and 595 nm can be occurred to 4 times with SiO$_2$, CaF$_2$ and TiO$_2$ particles. Therefore, this is not of much benefit to control the color uniformity and the lumen output.

All discussion above can be illustrated in Fig. 3, in which the CCT deviations for CaCO$_3$ and TiO$_2$ cases have a tendency to go downward. Specially, the angular CCT deviation can be decreased significantly by using TiO$_2$. For CaF$_2$ and SiO$_2$ cases, it’s interesting to observe that the CCT deviations in both cases grow with the concentration of corresponding particles.

4. Conclusion

In this paper, we analyze the effects of various scattering enhancement particles on two optical properties of WLEDs for achieving higher color uniformity. Based on the Mie-scattering theory and the Monte Carlo simulation method, we can verify that the scattered light enhancement achieved by using each type of particles in pcLEDs is quite different from each other. This opens a new door to access some methodology for effectively controlling of optical properties of WLEDs by selecting appropriate scattering enhancement particles with appropriate concentration. We found some
interesting results that can serve as a guideline for practical manufacturing of W-LEDs to meet some pre-defined specification on light quality. The CCT deviation would reduce when the concentration of CaCO$_3$ and TiO$_2$ increases. In particular, the best color uniformity of W-LEDs can be obtained in TiO$_2$ case.

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