Effects of titanium dioxide quantum dots on the color deviation and luminous flux of white light-emitting diodes

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ABSTRACT

The application of quantum dots has been considered as a promising approach to the advancement of phosphor-converted light-emitting diodes (pc-LEDs) since they perform an excellent extinction coefficient. Yet, it is challenging to manage their influences on the optical properties of LEDs due to their different nanometers in size. Hence, the object of this research is to analyze the influences of quantum dot (QDs) to figure out the solution to control the enhancement of LED lighting performances. Particularly, the study worked on investigating the scattering and absorption features of titanium dioxide (TiO_2) QDs. It demonstrated that the radiant efficiency and luminous stability of the TiO_2 QDs-converted LEDs (QC-LEDs) was inferior due to the strong light absorption and reabsorption occurring inside the LED packages. Additionally, it also presented low uniformity of color distribution because the scattering ability of QDs is weak. Therefore, reducing the concentration of QDs when adding to the LED structure seems to be possible to enhance the luminous output of QC-LEDs. We propose 0.05% wt. TiO_2 for white LED to reduce the illumination losing caused by re-absorbent and total internal backscattering, resulting in approximate 31% lumen improvement and high color rendering index (CRI) measured at about 85, at a high color temperature of 7500 K.

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1. INTRODUCTION

The lighting industry has remained its interests in light-emitting diodes (LEDs) since they were invented. The stability, durability, energy efficiency, and high performance are among outstanding features helping them gain much favor with the illuminating market [1]. One of the most frequent ways to make a white light-emitting diode is getting down-conversion material (DCM) excited by the blue LED chips, and the most utilized DCM in packaging white LEDs is the phosphor yttrium aluminum garnet (YAG) [2]. There were many reports on the enhancement of white-LED quality using DCM, which causes the absorption and scattering abilities of the materials. Sommer and co-workers pointed out that the angular color uniformity and radiant efficiency can be promoted via YAG particle size adjustment [3]-[5] since the different diameters of phosphor particles can result in different scattering effects to the packages. Besides, other studies of Sommer *et al*. focused on investigating the performances of phosphor-converted elements (PCEs) and presented that PCEs' absorption feature had considerable effects on their heating and cooling loads [6], [7]. Meanwhile, altering the concentration and size of PCEs in accordance with their absorption and scattering coefficients resulted in remarkable influences on the light transmission and reflection [8], [9]. The LED structure with multiple phosphors was previously examined in our study and the enhancement in LED optical performance was observed. In particular, we applied nitride in rod form as well as globular YAG phosphor particles and modified their concentration to appropriate values, thereby attaining high angle-dependent hue homogeneity (ACU) as well as efficiency of radiant flux. All of these outcomes can be demonstrated by the unique ability in scattering and absorption properties of each phosphor material [10]-[12]. Thus, according to these reported results, it is important to carry out both experimental and theoretical investigations on the light absorption and scattering of DCMs to precisely develop the high-quality white LEDs (WLED) packages. Several pieces of research took this one as their main purpose [13]-[18], and new designs of PCE-embedded WLED structures were developed based on them, including freeform optical surface structure with gradient thickness [19], [20], horizontal-separation layered packaging structures [21], [22], vertical-separation packaging structures [11], [23], and multi-microstructure-combined structures [24], [25]. The similarity among these studies is that they chose the rare-earth-based phosphor materials for WLED structuring and analysis. The rare earth phosphors are well-known for high luminescence efficiency yet the cost to enrich and purify them is too expensive leading to the continuously increasing prices of WLED devices in the lighting market. Therefore, many lamp manufacturers have been looking for a more cost-efficiency phosphor material with abilities to promote high optical performances for WLEDs. Another material called quantum dot (QD) was introduced and quickly gained much attention from researchers and manufacturers. QDs have cost-effectiveness, and easy control over the color properties and therefore been a desirable material for high color-quality WLED and solar cells.

Titanium dioxide (TiO_2) quantum dots having high efficiency in high photocatalytic activity would be viewed as a potential replacement to the conventional down-converted phosphor materials. Moreover, TiO_2 QDs present advantages that are beneficial to lighting production, such as high stability, non-toxicity, and environmental friendliness, while can be synthesized via simple methods such as the sol-gel method which significantly reduces the production cost for manufacturers. Additionally, their stability and quantum performance are promoted by physical and chemical modifications, which were researched in some previous results. However, TiO_2 QDs have been facing obstacles that prevent them from expanding their application in lighting-solution development. The most considerable one is the poor lumen efficiency leading to low reliability. Thereby, the structures using quantum dots as the converted elements (QDCEs) were extensively studied. Moreover, to enhance the chromatic stability of WLED with different injection currents, a concave lens constructed for chips was introduced to heighten the homogeneity in thermal absorption of QDCEs. Some studies demonstrated the enhancement in QD-converted LEDs (QC-LEDs) based on the LED using the personal consumption expenditures (PCEs). Particularly, these investigations pointed out noticeable differences in lighting properties of QC-LEDs and conventional phosphor-converted LEDs (PC-LEDs). At the same concentration, for example, the down-conversion efficacy achieved from QC-LEDs was higher than that from PC-LEDs. Specifically, the QC-LEDs attained high down-conversion efficacy with just 1% wt. QD used. Meanwhile, the PC-LEDs required a higher concentration of applied phosphor, which is more than 5% or even 50% wt., to yield the efficiency as high as the QC-LEDs did. Nevertheless, the down-conversion efficiency in QC-LEDs was impacted by the injection currents and the distribution of incident light, while these two factors were not present effects on this property of PC-LEDs. In addition to that, combining PCE structures with QDCE structure to boost the LED performance faced critical problems, the incompatibility between the QDCEs and the PCE structure, for instance, the vertically separated structure of yellow and red phosphor layers [11]. These articles obviously showed that the QD and phosphor materials own different scattering and absorption characteristics. Besides, the packaging structure of QC-LEDs has been the main concern of many recent research papers. This means that the scattering and absorption influences of specific QD on the packaging structure and its optical performances have not been deeply analyzed. Therefore, applying QD into high-performance LED fabrication is limited.

The intention of this research is to provide a demonstration in details about QD scattering and absorption effects on the QC-LED packaging structure. Particularly, the theorical and experimental analysis carried out on these two characteristics of TiO_2 QDs has been performed to examine the optic features of QC-LEDs. Moreover, the effectiveness in using TiO_2 QDs will be proven by comparing with the impacts of traditional yellow phosphor YAG. The Mie-scattering theory was used for this study to build the mathematic systems for measuring and evaluating the illuminating diffusion and also illuminating absorbent in TiO_2-QD LED packages. To make the research paper easy to follow, the specifications and structure diagrams of the QC-LED using TiO_2 QDs was presented in the next section. Consequently, the calculating expressions and explanations of achieved results was detailed to provide a more thorough understanding on QDs' usage for LED enhancement. Finally, the conclusion was drawn to summarize the achievements for references. We believe that the manuscript about TiO_2 QDs can contribute greatly to the development of QDCEs and help the WLED manufacturers enhance their product quality.

2. EXPERIMENTS

In Figure 1, the essential simulation data of the studied WLED is provided. Specifically, the specifications and images of the actual WLED we used in this study are displayed in Figures 1(a)-(c) is the illustration of the simulated QD-LED package for experiments. We examined the LED packages with nine blue LED chips with the size of 0.36×0.71 mm and a peak emission wavelength of 455 nm. The chips were attached to the 2.8×3.5 mm lead frame and covered with a silicone-phosphor layer. The silicone used to combine with phosphors and QDs has a 1.54 index of refraction. TiO_2 QDs and YAG yellow phosphor selected for the experiments have more than 80% quantum yield and green-low light emission. The mixture of silicone and QDs and yellow phosphor was dispensed over the chips to form the QDCE and PCE structures for later comparison. As mentioned in the introduction, the concentrations of QDs and phosphors hold a necessary part in specifying the optic productivity of LED packages. Here, we decided to adjust the amount of QDs and yellow YAG phosphor to 1.5% wt. and 12% wt., respectively. At these concentrations, the QDs and phosphors presented the same down-conversion efficiency. We simulated the WLED at the temperature of 25 ^{o}C and injected it with a 60-mA current. Instrument Systems' integrating sphere was used to measure the spectra of QDCEs and PCEs structures while an adjustable Keithley direct current (DC) power supply was utilized for their injection currents.

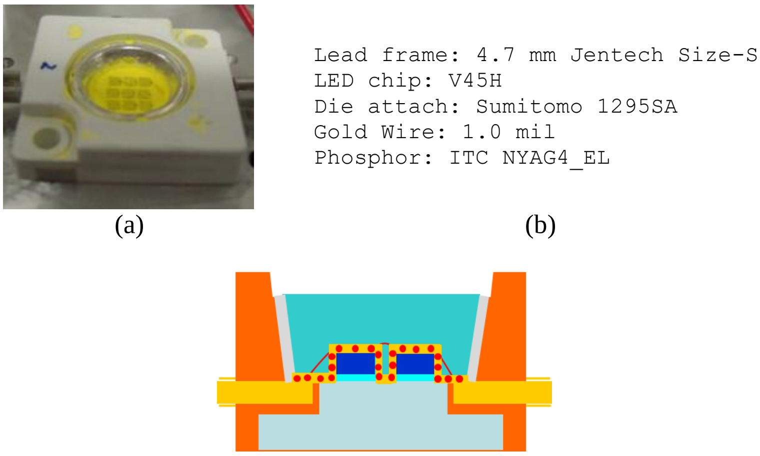

Figure 1. Picture of WLEDs configuration as doping TiO_2 (a) the real MCW-LED device along with, (b) its information, and (c) structure diagram of the device

3. COMPUTATION AND DISCUSSION

The color deviation of WLED packages with TiO_2 QDs was presented in Figure 2. It is noted that when the TiO_2 concentration is 0%, the deviation of correlated color temperature (CCT) is extremely large. However, as TiO_2 is added, the deviation tends to decline. Particularly, it is possible to get 2300 K CCT-deviation just by growing the content of TiO_2 particles to optimize their scattering property. CRI, amongst the most popular parameters to evaluate the hue standard of a WLED package, can be also heightened when increasing TiO_2 concentration to 0.41% wt. At 85 CRI, the LED can generate cool white light with the CCT of approximately 7500 K. Such an outcome is caused by the sufficient yellow-red emission color added into the white-light emission spectra. Moreover, when the TiO_2 QDs concentration was lowered to a proper amount, the TCL mass did not have much influence on CCT and color rendering index (CRI), while the CCT showed significant changes. According to this point, it is difficult to achieve complete control over the color temperature as well as CRI in QC-LEDs with low TiO_2 concentrations. Therefore, using high TiO_2 concentration seems to be advantageous to manage both CCT and CRI values. However, it is essential to adjust the particle concentrations and TCL mass to get the highest results.

The changes in the luminescence of QD-doped LED packages in relation to the TiO_2 QDs concentration were demonstrated in Figure 3. The lumen output of the package became higher at 0.5% wt. TiO_2 concentration, in comparison with that of 0% wt. TiO_2. The demonstration for this change probably is the higher portion of long-wavelength visible light color. This is caused by the heightened light reabsorption following the bettered scattering ability when the TiO_2 concentration doped in the package increased. Besides, when growing the TiO_2 concentration from 0% wt. to below 1.38% wt., the rise in green-yellow color, to which human eyes shows sensitivity, can be observed. Additionally, with an appropriate TiO_2

concentration, the total inner reflection (TIR) of lights occurring at the TCL-air interface can be minimized by taking advantage of the scattering property. Figure 3 also presented that when using 0.05% wt. TiO_2 in the LED package, the one with smaller TCL had higher lumen efficacy but insignificant improvement in green-yellow color emission. It has turned out that getting TIR decreased can be an effective approach to achieve better luminescence for WLEDs. Here, we observed the TiO_2 concentration at about 0.05 %wt. was beneficial to the luminous intensity and promoted it to be higher than 31%, compared to that of the configuration without using TiO_2. However, the decline in lumen output can be observed as the TiO_2 concentration kept becoming higher. This luminescence reduction can be demonstrated by high back-scattered and down-conversion losses. Though a higher concentration of TiO_2 would lead to better color uniformity, it can damage the luminous efficiency. The lumen output of QC-LEDs with TiO_2 QDs approximately 3.25% wt. showed the lowest values, below 10 mcd, in comparison with other lower TiO_2 concentrations. Additionally, the low radiant efficiency of the UV source and the fact that QDs have low quantum yield are the reasons resulting in the inferior total luminous efficiency that stayed below 0.3 cd/W (~1.4 l m/W).

The scattering coefficient μ_{sca} is the important parameter in analyzing the effects of TiO_2 on the LED optical properties. The scattering computation of TiO_2 was performed rely on Mie-scattering theory [23]-[26], and can be demonstrated as:

$$\mu_{sca}(\lambda) = \frac{c}{\bar{m}} \bar{C}_{sca}(\lambda) \tag{1}$$

$$\bar{C}_{sca}(\lambda) = \frac{\int^{\int} C_{sca,D}(\lambda) f(D) dD}{\int^{\int} f(D) dD} \tag{2}$$

$$\bar{m} = \frac{\int^{\int} m_i(D) f(D) dD}{\int^{\int} f(D) dD} \tag{3}$$

$$C_{sca}(\lambda) = \frac{P_{sca}(\lambda)}{I_{inc}(\lambda)} \tag{4}$$

Here, λ and c indicate the wavelength (nm), the concentration of QD particle (g/cm^3), respectively. *f(D)* presents the size distributing function, and $\underline{m}$ is the particle mass of the integrated QDs. $\underline{C}_{sca}(\lambda)$ displays the diffusing cross-section and $C_{sca,D}$ means the scattering cross-section of phosphor with D diameter. $P_{sca}(\lambda)$ shows diffusing energy and $I_{inc}(\lambda)$ is the emission intensity.

Figure 4 demonstrated the calculated scattering parameters of TiO_2 at the two wavelengths measured at 450 nm and 550 nm. The study focused on the scattering results in the wavelengths measured at 450 nm and 550 nm as the lights emitted at these two wavelengths are the main lighting elements in the phosphor layer. Particularly, Figure 4 presented the scattering cross-section following the change of TiO_2 particle size. The scattering of TiO_2 at 450 nm is much better than at 550 nm wavelength. The scattering coefficient is the crucial parameter in evaluating the scattering ability of a particle. This result is good for the blue-light scattering. In other words, this enhances the blue-light proportion in the phosphor layer, leading to the efficiency in minimizing the yellow ring phenomenon. Besides, this blue-light scattering enhancement brings better color quality because the illuminations of blue and yellow can be blended and spread uniformly extra times. However, all these potential improvements can be ruined completely by the inappropriate particle size and concentration of TiO_2. Hence, our study investigating the influence of TiO_2 diameters is probably a valuable reference for manufacturers to select the most optimal particle size for their WLED products. According to Figure 4, the scattering of lights at 450 nm wavelength is always better than at the 550 nm one. This emphasized that TiO_2 is a suitable particle for achieving higher color quality of WLED lamps. Small TiO_2 particle leads to a better mixing process of lights and thus heightening the color homogeneity. This means, the lower the particle size, the larger the light-scattering angle and higher light-blending efficiency [27], [28]. Yet, the higher scattering means the higher energy consumption. In contrast, the larger particle will be advantageous to the luminous efficiency since it allows the lights to be transmitted easily, leading to lower scattering events and narrower scattering angles. Therefore, determining the particle size and proportion that can balance the chromaticity and lumen output becomes much more important to the application of TiO_2 QDs [29], [30]. Combining the results illustrated in Figure 3 and 4, it seems that with the particle size of TiO_2 of 400 nm, the LED packages can have smaller color deviation and the scattering coefficient become more stable. In addition to that, the luminous output was achieved with 400-nm TiO_2 at 0.05% wt. showed high values. Thus, 400 nm or 0.05 % wt. of TiO_2 QDs can be selected for the optimization of WLED properties.

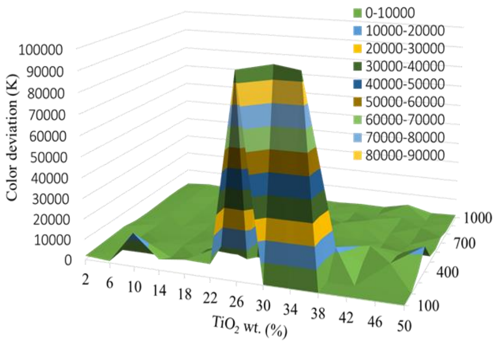

Figure 2. Color deviation of phosphor-transformed MCW-LEDs as doping TiO_2

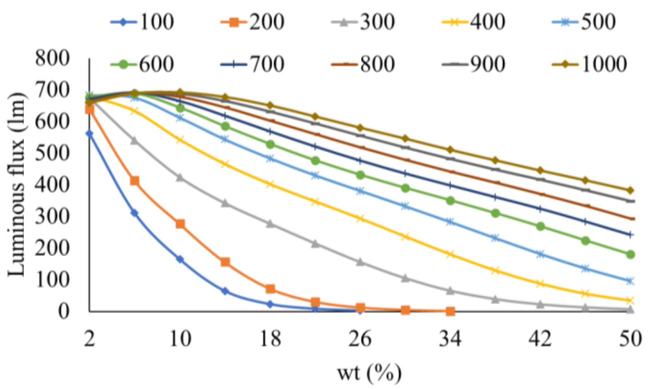

Figure 3. Luminous flux of phosphor-transformed MCW-LEDs as doping TiO_2

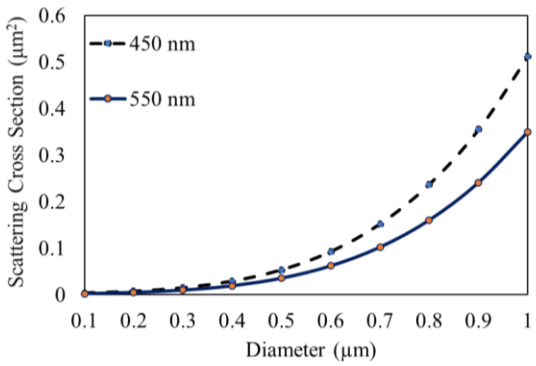

Figure 4. The computed scattering cross-section values of phosphor compounding doping TiO_2 particles

4. CONCLUSION

The effects of TiO_2 quantum dots on the white-light quality when being doped into the phosphor structure of WLED packages were introduced and discussed in this article. It is proved that TiO_2 is suitable material for further WLED development as this quantum dot showed great ability to improve the light scattering, absorption and extraction efficiency. The research findings indicated that the diameter and concentration of TiO_2 QDs play important role in the control over color quality and luminous efficacy. It pointed out 0.05% wt. is the sufficient concentration for TiO_2 to be applied to the LED package to color adequacy. Particularly, at this concentration, TiO_2 can promote high CCT of about 7,500K and CRI of 85 for LED devices. Moreover, 0.05% TiO_2 led to 31% improvement in luminous intensity. Besides, in terms of diameter, it was reported that TiO_2 particle size of 400 nm can considerably reduce the color deviation while maintaining the stability of scattering coefficient values. Additionally, high lumen output can be observed with 0.05% concentration and 400 nm diameter of TiO_2. Hence, the suggestion for producing higher-quality WLEDs can be using 400-nm TiO_2 with a concentration of 0.05% wt.

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