Benefits of using TiO₂ quantum dots in producing low-cost and high-quality white light-emitting diodes

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ABSTRACT

Quantum dots (QDs) is considered as a potential material for the improvement of light-emitting diodes (LEDs). However, different from the traditional phosphor materials, they have unique scattering and absorption properties affected by their several nanometers sizes, which makes their application in the production of LED confront more challenges. In addition to this, their influences on QDs-converted LEDs (QCLEDs) are rarely investigated. So as to propose solutions for those problems, in this article, we experimentally and theoretically investigated the impacts of titanium dioxide (TiO2) QDs' scattering and absorption on the light quality of QCLEDs by drawing a thorough comparison between their properties and the traditional yttrium aluminum garnet phosphors characteristics. The outcomes showed that QCLEDs have poor radiant efficacy and stability due to QDs' strong characteristic of absorption (reabsorption) while their weak scattering property causes a low uniformity in correlated color temperature (CCT). For achieving high efficiency and stability white LEDs, we highly suggest using QDs with a low concentration to get reductions in the reabsorption and total internal reflection losses. With 0.05 concentration of TiO₂ nanoparticles (TiO₂ NPs), the white LEDs can simultaneously achieve a high CCT (approximately 7500 K) and a high color rendering index (around 85).

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1. INTRODUCTION

Light-emitting diodes (LEDs) have gradually spread their application in lighting industry because they own many outstanding characteristics, such as long lifespan, stability and high efficiency [1]. To generate white-light LEDs, there were various practical methods proposed and one of them is exciting the down-conversion material (DCM), for instance, yttrium aluminum garnet (YAG) phosphor by utilizing blue LED chips [2]. Many previous researches showed that DCM had a great effect on the quality of white light LEDs (W-LEDs) due to its light scattering and absorption. According to previous studies [3]-[5], it is possible to enhance the angular correlated color temperature (CCT) uniformity and the radiant flux of W-LEDs by adjusting the particle size of YAG phosphor with different scattering functions. In addition to this, their works also demonstrated the significant impact of the absorption characteristic of phosphor-converted

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elements (PCEs) on their ethermal load [6], [7]. According to the scattering and absorption coefficients of PCEs, other studies carried out the investigation on the transmissivity and the reflection of PCEs while their particle sizes and concentrations were adjusted [8], [9]. Besides, in our previous studies, we determined that when using a multi-phosphor structure with appropriate proportion of rod-shaped nitride and spherical-shaped YAG phosphor, W-LEDs can accomplish high angular uniformity and luminous flux [10]. This achievement can be explained by the different scattering and absorption properties of the used phosphor [11], [12]. Moreover, analyzing the DCM's scattering and absorption plays a crucial role in constructing a precise model W-LEDs from experiments and theory, and thus there were many studies focus on these two properties of DCM [13]-[15]. Furthermore, these studies inspired researchers to design new structures using PCEs for white LEDs, such as freeform surface (gradient thickness) [16], horizontal separated [17], vertical separated [18], and microstructures combined structures [19]. Nonetheless, they only mentioned structures with the rare-earth-based phosphor DCM, an expensive and ineluctable material. Another material called solution-processed colloidal quantum dot (QD) has drawn a great attention to researchers because of their color-tunable and chromatic saturation characteristics. Therefore, they are popular in improving the performances of optoelectronic equipment including LED lights and photovoltaic cells.

Titanium dioxide (TiO₂) (core/shell) QD, which has unique photocatalytic properties, is considered as a great alternative to the traditional phosphor DCM in fabricating a high-quality white LED generation. In addition, the studies with relative topics have been conducted to get their quantum yield and stability enhanced. Nevertheless, for their applications in W-LED packages, they still confront some considerable existing challenges consisting of poor lumen efficiency and reliability. For overcoming these problems, in recent years, research papers have directed their concentration to the packaging designs of QDs-converted LEDs (QCLEDs). Additionally, they also constructed a concave lens for chips in order to enhance the absorption (thermal) homogeneity of QCLEDs, and improving the chromatic constancy at various injection currents as a result. Besides, some previous articles proposed PCE structures in terms of accomplishing the optimization of QDCE structures. Most of the findings in these studies presented that QCLEDs showed different characteristics in comparison with that of the conventional phosphor-converted LEDs (pc-LEDs). For instance, QCLEDs can yield high down-conversion performance with low QDs' concentration, less than 1 wt%, while pc-LEDs need to have its concentration increased over 5 wt% and can be up to 50 wt% to achieve the same result. However, the injection current and incident light distributions largely affect the down-conversion efficiency of QCLEDs. Meanwhile, that of pc-LEDs seems not to be influenced by these two factors. Furthermore, QCLEDs are not compatible with some efficient PCE designs, for example, the one constructed by vertically separating the yellow and red phosphors mentioned in the previous research [20]. From these results, it can be implied that the effects of scattering and absorption properties between QD and phosphor are not the same, which is probably the main obstacle that prevents the QCLEDs from enhancing their performances. However, researchers have just focused on studying OCLED packages recently, and thus, the optical efficiency of OCLEDs in accordance with the effects of the scattering and absorption characteristics of QD has not been demonstrated in any article. Therefore, there is not enough reference for establishing the most effective packing design for OCLEDs.

This article demonstrates experimental and theoretical investigations about the influence of the light scattering and absorption of TiO_2 QD on the W-LED optical properties, which is conducted by drawing a comparison between this nanoparticle and the traditional YAG phosphor. Specifically, the lighting performances of QCLEDs and pc-LEDs are examined and presented in experiments, initially. After that, there is an introduction of the measurement method that combines finite-difference time domain (FDTD) and ray tracing (RT). This introduction shows and explains the mechanisms of these results, and also considers the unique light scattering and absorption effects of QD which is distinct from that of YAG phosphor. Finally, QCLEDs and pc-LEDs are compared from theoretical aspects, and then, some potential approaches to enhance the performance of QCLEDs are given based on the difference concluded from the aforementioned comparison. The findings from this manuscript can become a good reference offering a route to a better understanding of designing QCLEDs' packing structures from the aspect of scattering and absorption effects.

2. EXPERIMENT

Figure 1(a) illustrates the structure of white LEDs. The blue LED chips and the commercial lead frame used in WLED production have the size as follows, in turn: $0.36 \text{ mm} \times 0.71 \text{ mm}$ and $2.8 \text{ mm} \times 3.5 \text{ mm}$. In addition, the LED chips also have the emission wavelength centered at 455 nm, and the silicone participating in constructing the LED packages has refractive index of 1.54, see Figure 1(b) and Figure 1(c). The TiO₂ QDs and YAG phosphor, whose green-yellow emission light and quantum yield are larger than 80%, are blended with silicone, and then being dispersed on the chips in the lead frame in order to achieve QCLEDs and PCEs, respectively. Besides, for the purpose of comparing these two structures,

the concentration of QDs is adjusted to 1.5 wt% while that of YAG phosphor is 12 wt%, as these are the concentrations that could help the QCLEDs and pc-LEDs have an equal down-conversion efficiency which is known as the radiant power proportion of QD and the phosphor light to the proportion of the total emission light. Both of the WLED models are developed in the environment temperature of 25 °C and with 60 mA injection current. Their spectra measurement is carried out by using the integrating sphere from instrument systems, and their injection currents is provided by a Keithley adjustable dc source.



Figure 1. Schematic diagrams of phosphor-converted MCW-LEDs as doping TiO₂: (a) the actual MCW-LEDs, (b) its parameters, and (c) structure diagram of MCW-LEDs

3. COMPUTATION AND DISCUSSION

Figure 2 shows the deviation of CCT of WLED in accordance with the concentrations of TiO_2 QDs. There is one feature that was not mentioned in Figure 2 when TiO_2 QDs concentration is 0 wt%, the CCT of WLEDs is infinite. However, that infinity of CCT of WLEDs having single type of QD can be reduced to about 2300 K by the growth of TiO_2 QDs concentration, due to the scattering effect of TiO_2 QDs. It is possible to achieve a high value of color rendering index (CRI) when the concentration of TiO_2 ranges from 0.17 to 0.41 wt%. When CRI is at around 85, the WLEDs package can generate lights having a color of cold white with a CCT of approximately 7500 K. This may be caused by the proper yellow-red color proportion filled in the spectra. In addition, when TiO_2 concentration is lowered to a sufficient amount, either the CCT or the CRI are not sensitive to the TiO_2 nanoparticles (TiO_2 NPs)-doped CDs layer (TCL) mass, and the CCT shows a sharp change [21], [22]. Hence, if TiO_2 concentration is too low, adjusting the CCT and CRI will be more difficult. Thus, to be able to manage the CCT and CRI, we can use a higher concentration of TiO_2 , and then varying its concentration and the TCL mass.



Figure 2. Color deviation of phosphor-converted MCW-LEDs as doping TiO₂

Figure 3 is the illustration of the luminous flux of WLEDs with different concentrations of TiO₂ QDs in the applied phosphor compounding. The results show that when there is 0.5 wt% TiO₂, the LEDs can get the maximum luminous intensity which is higher than the one without the presence of TiO_2 QDs. One of the reasons behind this is the increased the amount of color with long wavelength which emerges from the increase in reabsorption through scattering when growing the concentration of TiO_2 NPs. Especially, when concentration of TiO_2 NPs is smaller than 1.38 wt%, there is an increase in the green-vellow color proportion that is sensitive to the human eyes following the rise of TiO_2 NPs concentration. What is more, with an optimized proportion of TiO₂, it can reduce the total internal reflection (TIR) at the TCL-air interface by scattering. As pointed out in Figure 3, when the concentration of TiO_2 QDs grows from 0 to 0.05 wt%, the WLED having smaller TCL exhibits a better enhancement in lumen efficiency. Nevertheless, with that concentration, the proportion of green-yellow color also exhibits a smaller improvement. Thus, the reduction in TIR is considered as one of the most important factors to improve the lumen efficiency. Therefore, the most appropriate TiO₂ concentration for WLEDs is 0.05 wt% by which the luminous intensity can reach its maximum value of 31%, higher than the flux from the LEDs packages using phosphor silicone encapsulant. However, when the concentration of TiO₂ continues to increase, the luminous intensity has a significant decrease, which mainly caused by the development of the back-scattered and down-conversion losses occurring following the continuous growth in TiO₂ concentration. It is possible for the LEDs to accomplish various spectral ranges by adjusting the TiO₂ QDs concentration, but the luminous intensity is largely affected by the amount of TiO_2 NPs, and it even bottoms out for less than 10 mcd when TiO_2 concentration is 3.25 wt%. Moreover, the lumen output of the device is totally lower than 0.3 cd/W (1.4 l m/W), due to the low radiant efficiency of UV source and the low quantum yield of QDs. Therefore, to reach an enhancement in lumen efficacy, it needs to be improved the QDs quantum yield and the radiant efficiency of the UV source.

It is essential to calculate the scattering coefficient μ_{sca} before measuring other optical properties of the phosphor compound in MCW-LED. Based on the Mie-theory [23]-[25], the scattering coefficient (SC) μ_{sca} , are expressed as (1)-(4):

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

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

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

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

In which, f(D) is the size distribution function and c is the phosphor concentration (g/cm^3) . $\bar{C}_{sca}(\lambda)$, $C_{sca,D}$ indicates the scattering cross-section and the scattering cross-section of the phosphor with D indicates the particle diameter, respectively. \bar{m} represents the particle mass of the phosphor integrated on f(D). The scattering power and emission intensity are $P_{sca}(\lambda)$ and $I_{inc}(\lambda)$, in turn.

Figure 4(a) illustrates the scattering coefficient (SC) of WLED with different sizes of TiO₂ particles. As can be seen, the fluctuation in SC is mainly caused by the size of TiO₂; the larger the particle size of TiO₂ the wider the fluctuation of the scattering coefficients. Particularly, the rise of the scattering coefficient follows the increase of phosphor concentration, and this phenomenon becomes stronger in the wavelength band of 680 nm and above. During the experiment, the obtained SC is always higher when there is a higher phosphor concentration, regardless of the size of TiO₂ size which remains at 200 nm or 400 nm, leading to better color uniformity. However, with the TiO₂ phosphor particles of 200 nm or 400 nm, the SC tends to be more stable regardless of the rise in the concentration of TiO₂, which is beneficial to the color quality scale (CQS). Thus, based on the illustration in Figure 4(b), it can be concluded that 400 nm is a suitable size of TiO₂ particles for applying in WLED when the aim is to enhance the color quality. From the results of this section, it can be affirmed that not only the concentration of TiO₂ phosphor but also its size can determine the performance of WLED. Moreover, manufacturers can freely change the suggested parameters depending on their targets, which implies that TiO₂ QD is an effective material using for the enhancement of both luminous efficiency and color uniformity of WLEDs.



Figure 3. Luminous flux of phosphor-converted MCW-LEDs as doping TiO2



Figure 4. The computed values of (a) scattering coefficients and (b) average cosine of phase function of phosphor compounding doping TiO₂ particles

4. CONCLUSION

This paper demonstrated the optical performance of WLEDs with silicone encapsulant having the phosphor compounding added TiO_2 QDs. Using TiO_2 NPs is to enhance the scattering property of the silicone encapsulant, and then resulting in the significant probability of light reabsorption and extraction. As can be seen from the findings of the article, changing the concentration of TiO_2 NPs can adjust the emission spectra of the encapsulant layer, and thus acquiring various color qualities of WLEDs with a certain and small amount of QDs. In particular, with the optimized concentration of TiO_2 of 0.05 wt%, the LEDs can attain high values of CCT and CRI which are approximately 7500 K and 85, respectively. Furthermore, that concentration also results in the increase in the luminous intensity by 31%. Therefore, TiO_2 QDs can become an effective alternative to traditional phosphor material in fabricating low-cost and high-quality WLEDs.

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