Enhancement of Luminous Efficiency and Uniformity of CCT for Quantum Dot-Converted LEDs by Incorporating With ZnO Nanoparticles

Yong Tang, Zhi Li, Zong-Tao Li, *Member, IEEE,* Jia-Sheng Li, Shu-Dong Yu, and Long-Shi Rao

***Abstract*— ZnO nanoparticles were incorporated into quantum dot (QD)-silicone encapsulation materials of the light-emitting diodes (LEDs) to exploit their strong scattering effect, which were proved by the ZnO-only film experiment. The novel packaging scheme led to a decrease in the radiation flux because of the conversion-energy loss of QDs and backscattering of ZnO nanoparticles. Under the similar color coordinate, the luminous flux of the ZnO-incorporated QD-LED showed a 3.37% increase compared to the conventional structure. This was attributed to the scattering effect, which enhanced the utilization of blue light and the conversion of yellow light. And the angular-dependent correlated color temperature (CCT) deviation was reduced from 862 to 712 K in the range of −70° to 70°. Moreover, the CCT monotonically decreased, and the chromaticity coordinate steadily shifted to the yellow region as the amount of ZnO nanoparticles increased. Therefore, ZnO nanoparticles can be a favorable optical performance enhancer for the future generation of QD-converted LEDs.**

***Index Terms*— Light-emitting diodes (LEDs), quantum dots (QDs), scattering, ZnO nanoparticles.**

I. Introduction

THE quantum dot (QD)—that has recently attracted considerable attention in the scientific and industrial fields—is a new nanomaterial comprising only several atoms. QDs exhibit a remarkable quantum-confinement effect, macroscopic quantum-tunneling effect, surface effect and offer distinctive, and favorable properties [1], [2]. Using high-efficiency and high-quality organic-phase synthesis or the environmental water-soluble hydro-phase synthesis method, common QDs can be synthesized from IIB–VIA-group elements, such as CdS, CdSe, CdTe, and ZnSe, or IIIA–VA-group elements, such as InP and InAs [3], [4]. The successful synthesis of QDs has led to their broad applications for solar cells, biomarkers, chemical catalysts, sensors, and photoelectric devices [5]–[7].

Among the aforementioned applications, QDs have increasingly been used in photoelectric devices in the past few years owing to their extraordinary advantages over the traditional luminescent materials [8], [9], which are as follows.

1) QDs have a higher fluorescence quantum yield than that of traditional luminescent materials: Greytak *et al.* [10] demonstrated the addition of an alternating layer in CdSe/CdS core–shell QDs, yielding fluorescence quantum yields up to 98%.
2) The emission spectra of QDs can be controlled according to their size and chemical composition to cover the visible region [11]. Using the CdTe QD as an example, its emission peak can redshift from 510 to 660 nm as its grain size increases from 2.5 to 4 nm [12].
3) QDs have a narrow and symmetric full-width at half-maximum (FWHM), which means that the color of the emission light is far purer than that of phosphor, and the emission spectra do not overlap when different QDs are used simultaneously, which helps to display the color precisely [13].

The most widely used method for obtaining white light-emitting diodes (LEDs) is packaging blue LEDs with yellow phosphors ($Y_3Al_5O_{12}$:Ce^{3+}) [14], [15]. In light of the advantages of photoluminescent QDs and of replacing phosphors with QDs, the QD-converted white LED has been introduced and gradually attracted considerable research interest [16]. However, many problems remain for the industrial production of QD-LED. The synthesis of QDs is still immature; the luminous efficiency and stability of QD in the LED package are still undesirable [17]. Moreover, the mass production of QDs has not been achieved, leading to a high production costs. While the mass production of QD-converted LEDs requires a large quantity of converted QDs, which is limited to the immature properties of QDs and its high cost [18]. Therefore, increasing QDs usage efficiency in the packaging, in other words enhancing the luminous efficiency of the QD-LED are worthwhile tasks.

Manuscript received August 2, 2017; revised October 18, 2017; accepted November 6, 2017. Date of publication November 28, 2017; date of current version December 27, 2017. This work was supported in part by the National Natural Science Foundation of China under Grant 51405161 and Grant U1401249, in part by the Natural Science Foundation of Guangdong Province under Grant 2014A030312017, and in part by the Science and Technology Program of Guangdong Province under Grant 2015B010132005 and Grant 2015B010114003. The review of this paper was arranged by Editor E. G. Johnson. *(Corresponding author: Zong-Tao Li.)*

Y. Tang, Z. Li, J.-S. Li, S.-D. Yu, and L.-S. Rao are with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology, South China University of Technology, Guangzhou 510640, China.

Z.-T. Li is with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology, South China University of Technology, Guangzhou 510640, China, and also with Foshan Nationstar Optoelectronics Company Ltd., Foshan 528000, China (e-mail: ancient@yeah.net).

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TED.2017.2771785

In the field of traditional phosphor-converted LEDs, the strong scattering effect of nanoparticles has been used to enhance the light-extraction efficiency and correlated color temperature (CCT) performance of devices [19]. Chen *et al.* [20] demonstrated a white LED with a layer of a remote ZrO_2-incorporated phosphor structure that enhanced the luminous efficiency by 2.25% and reduced the deviation of the CCT from 1000 to 420 K in the range of −70° to 70° owing to the strong scattering effect and the refractive-index gradient created by the ZrO_2 nanoparticles. Nevertheless, most of the nanoparticles-incorporated studies aim at phosphor-converted device, the relative research on QD-converted device is still scarce. Zheng *et al.* [21] added a TiO_2/silicone encapsulation layer to the bottom of chip-on-board packaging LEDs to redirect the backscattering blue light to extract the encapsulation materials, the research found that the light-extraction decreased with the phosphor concentration increasing. It indicates that the original scattering ability of phosphor restrains the light-extraction ability of nanoparticles. Hence, it could be inferred that nanoparticles might have a better promotion on the light-extraction performance of the QD-converted LED since QDs present almost nonscattering properties [22]. ZnO nanoparticles are promising because of their excellent thermal and optical properties, and they have been widely applied in piezoelectric devices, sensor, and solar cells [23]. However, there are no reports of enhancements in the utilization rate of QDs, e.g., increasing the luminous efficiency and improving the CCT performance by incorporating ZnO nanoparticles into the encapsulation of QD-converted LEDs.

In this paper, different amounts of ZnO nanoparticles were incorporated uniformly into the silicone encapsulation materials of CdSe QD-converted white LEDs. The scattering ability of the ZnO nanoparticles was determined by ZnO-only films. The influence of the content of ZnO nanoparticles on the radiation flux, luminous efficiency, and CCT was experimentally investigated. The influence of the incorporating with ZnO nanoparticles on the optical performance of the QD-converted LED was studied in detail, providing a possible solution for increasing the utilization rate of QDs in the QD-converted LEDs mass production.

II. Experiment

Fig. 1 shows a transmission electron microscopy (TEM) image of CdSe QDs from Beijing Beida Jubang Co., Ltd., with an average diameter of 10 nm and a photoluminescence quantum yield as high as 80%. Fig. 2 shows the luminescence spectrum under 450-nm blue-light excitation and the ultraviolet absorption spectrum of the CdSe QDs tested by fluorospectro photometer. Here, we observe that the QDs had a small absorption peak at 525–550 nm. The emission peak wavelength was ∼555 nm, and the FWHM of emission was ∼40 nm. The ZnO nanoparticles are supplied from Nanjing XFNANO Co., Ltd., with the diameter ranging from 20 to 30 nm. The ZnO-incorporated QD-converted LEDs were prepared as follows.

1) A 6.6 mg of CdSe QD powder was completely dissolved into 3 mL of chloroform mixed with 0.75 g of A-silicone and 2.25 g of B-silicone; the QD concentration is 0.22 wt%.
2) The QD-silicone-chloroform solution was evaporated at 50 °C until the chloroform volatilized completely.
3) The QD-silicone mixture was incorporated with ZnO nanoparticles.
4) The mixture was placed into a vacuum de-aeration machine to disperse the nanoparticles thoroughly and remove the air bubbles.
5) The encapsulation mixture was dispensed into the LED package and cured at 150 °C for 3 h.

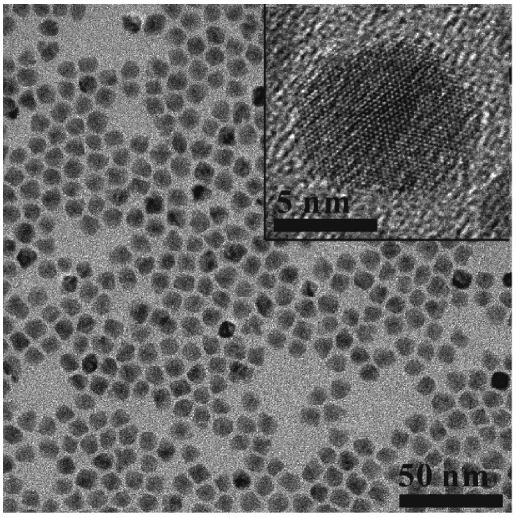

Fig. 1. High-resolution TEM image of CdSe QDs dispersed uniformly in the chloroform solution.

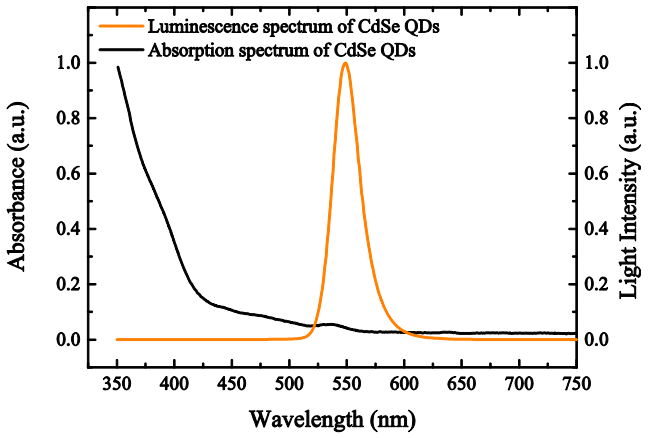

Fig. 2. Luminescence spectrum under 450-nm blue-light excitation and ultraviolet absorption spectrum of CdSe QDs.

In the reference sample, only the QDs were uniformly mixed with the silicone encapsulant. The GaN-based blue chip with an emission wavelength of 450 nm was bonded in the lead-frame package. Fig. 3 shows a cross-sectional schematic of a ZnO-incorporated QD-converted LED and a conventional QD-converted LED. LEDs with the same original optical performance are selected for comparison at a driving current of 350 mA.

III. Results and Discussion

The blue lights emitted from the chip cannot be effectively scattered because the particle size of the QDs is only

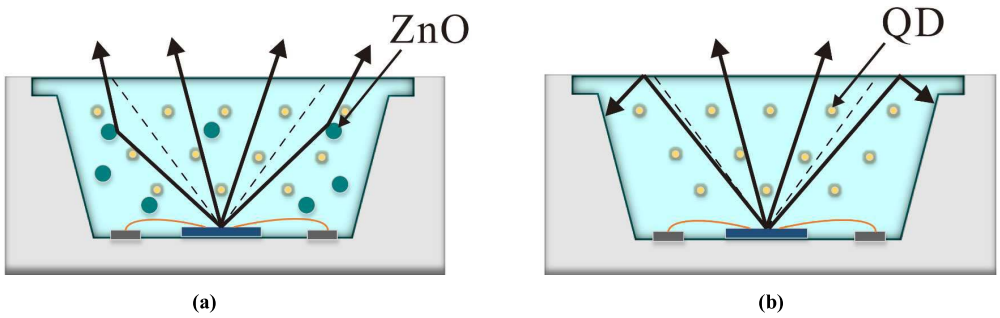

Fig. 3. (a) Schematic cross-sectional view of ZnO-incorporated QD-converted LED. (b) Schematic cross-sectional view of conventional QD-converted LED.

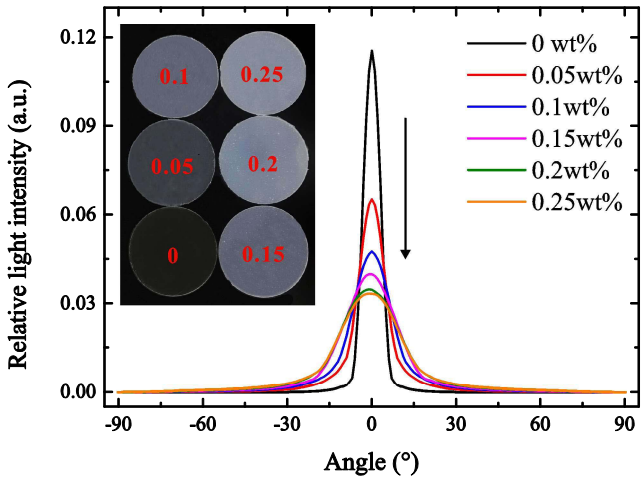

Fig. 4. Light intensity distribution of different concentrations of ZnO-only films. The inset shows the photograph of ZnO-only films.

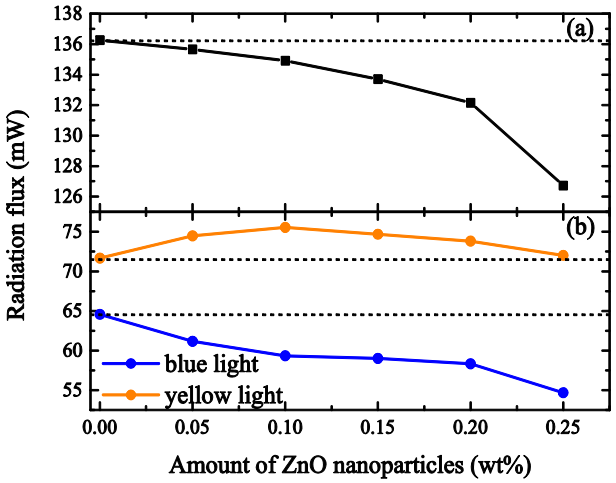

Fig. 5. (a) Radiation flux of the QD-converted LEDs with different contents of ZnO nanoparticles under the measured current of 350 mA. (b) Corresponding blue-light radiation-flux integrated from 380 to 510 nm and yellow-light radiation-flux integrated from 510 to 700 nm.

several nanometers. The ineffective scattering of light might give rise to issues such as severe total internal reflection (TIR) and a low light conversion efficiency. First of all, in order to study the scattering impact of ZnO nanoparticles, a series of ZnO nanoparticles-only films with the concentration varied from 0 to 0.25 wt% has been introduced. The thickness of the films is 0.5 mm, and the light intensity distribution was tested by spatial light distribution instrument under a 450-nm laser light [24]. As illustrated in Fig. 4, when the ZnO nanoparticles concentration became larger, the light intensity distribution became more and more scattered. Hence, it is confirmed that the ZnO nanoparticles can bring effective scattering ability, and the scattering ability strengthens as the concentration becomes larger. With this law, we studied the optical impact of ZnO nanoparticles on the QD-converted LEDs later and Fig. 5(a) shows the radiation flux of the QD-converted LEDs incorporating with different contents of ZnO nanoparticles, the test was conducted by calibrated integrating sphere under a measured current of 350 mA. The QD concentration was the same for all the tested samples. The experimental results indicated that the radiation flux monotonically decreased as the amount of ZnO nanoparticles increased, which means that the ZnO nanoparticles intensified the internal light energy loss. The energy bandgap of bulk ZnO is 3.3 eV, which corresponds to approximately 375 nm, and it should not be possible that these ZnO nanoparticles absorbed the light of the blue LED and QDs which emitted at around 450–460 nm and 535–575 nm, respectively [25]. The experimental results differ from those of conventional nanoparticle-incorporated phosphor-converted LEDs that exhibited a stable radiation flux as the content of nanoparticles changed [20].

The ZnO mechanisms of light extraction might not be exactly identical for conventional phosphor-converted LEDs and QD-converted LEDs. To further investigate this issue, the blue-light radiation flux from 380 to 510 nm and yellow-light radiation flux from 510 to 700 nm were individually extracted by integrating the corresponding energy of the spectra. The corresponding statistics are shown in Fig. 5(b).

The blue-light radiation flux monotonically decreased as the amount of ZnO nanoparticles increased, for which we consider two possible reasons. First, a larger content of ZnO nanoparticles yielded a greater scattering effect. The scattered blue light experienced more complicated optical paths in the encapsulation materials; thus, more blue light was absorbed by the QDs. Second, an excessive content of ZnO nanoparticles increased the backscattering of the blue light; thus, more blue light was absorbed by the substrate and the sidewall of the LEDs. In contrast, the yellow-light radiation flux initially

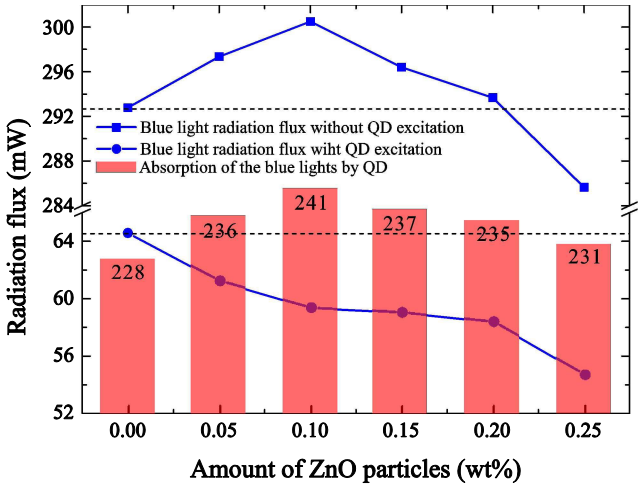

Fig. 6. Blue-light radiation flux with and without QD excitation and the corresponding blue-light absorption of QDs.

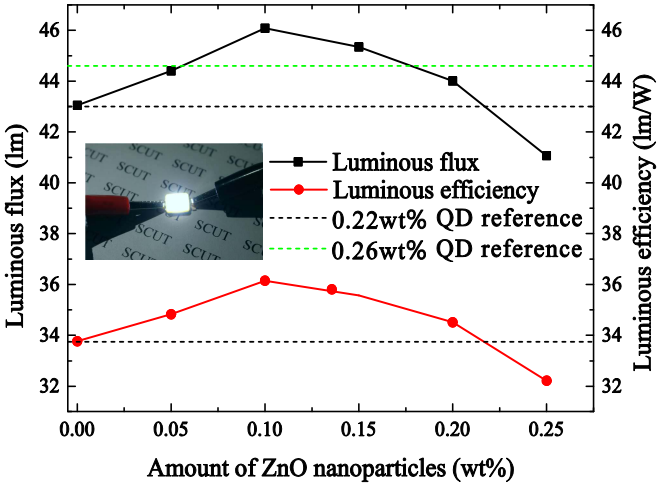

Fig. 7. Luminous flux and luminous efficiency of QD-converted (0.22 wt%) LEDs with different contents of ZnO nanoparticles (green dashed line represents the luminous flux of 0.26 wt% QD reference sample). The inset shows the QD-LED (0.1 wt% ZnO incorporated) under operation.

increased and reached its maximum value when the concentration of ZnO nanoparticles was 0.1 wt%. This value was 5.44% higher than that of the QD-LEDs without ZnO incorporating, probably because as the content of ZnO nanoparticles increased, the scattering ability of the nanoparticles increased; thus, the blue lights emitted from the LED chip were scattered more uniformly in the encapsulation materials and then absorbed and transferred by the QDs more thoroughly, and the yellow-light radiation flux increased. However, the yellow-light radiation flux began to decrease as the amount of ZnO nanoparticles continued to increase. Because the absorption ability of QDs is intense, we hypothesize that the absorption of the QDs had already been saturated when the amount of ZnO nanoparticles exceeded a certain value [26], and when the transferred yellow lights reached the peak, the continued increase of ZnO nanoparticles only caused the unnecessary absorption of yellow lights by the package structure and reabsorption by the QDs. The latter phenomenon is indicated by the absorption spectrum shown in Fig. 2, which contains an absorptance peak in the wavelength range of 525–555 nm.

The aforementioned analyses are partially based on speculation and are insufficient for explaining the decreasing curve of the radiation flux. To clarify the physical mechanism causing the phenomena, blue LEDs-incorporated with ZnO nanoparticles were prepared. The blue-light radiation flux without QD excitation was tested, as illustrated in Fig. 6. The blue-light radiation flux without QD excitation initially increased and reached its maximum value when the amount of ZnO nanoparticles was 0.1 wt%. This value was 2.63% higher than that of a reference blue LED without ZnO, indicating that the scattering effect of the ZnO nanoparticles effectively reduced the TIR, and that the radiation-flux increase of blue lights might compensate for the QD conversion-energy loss. As the amount of ZnO nanoparticles increased, the radiation flux began to decrease. When the concentration of ZnO nanoparticles was 0.25 wt%, a negative radiation-flux performance was observed compared with the reference blue LED without ZnO. The cause of this appeared to be the backscattering of ZnO nanoparticles. To investigate further, we considered that the backscattering and other energy losses in the ZnO-incorporated only blue LEDs and the ZnO-incorporated QD-converted LEDs were the same and used the corresponding blue-light radiation flux without QD excitation minus the blue-light radiation flux with QD excitation to represent the absorption of the blue light by the QDs, as plotted in Fig. 6. The absorption reached its peak when the concentration of ZnO nanoparticles was 0.1 wt% and then decreased as the concentration of ZnO nanoparticles increased. Considering the monotonically decreasing curve of the radiation flux, these phenomena can be understood as follows.

1) When the content of ZnO nanoparticles was relatively low, increasing the content promoted the blue-light absorption by the QDs because of the enhanced scattering effect. However, as the yellow-light transfer increased, the conversion-energy loss was larger than the energy enhancement caused by the promotion of the scattering ability, which caused the radiation flux to decrease at first.
2) As the ZnO nanoparticles concentration continued increased, the absorption of QD was saturated, the positive effect of increasing blue-light absorption reached its peak and the negative effect of aggravating energy loss still increased.
3) An excessive content of ZnO nanoparticles increased the backscattering, yielding the monotonically decreasing curve of the radiation flux.

Nevertheless, different amounts of radiation flux are needed for different wavelengths of monochromatic light to produce the same visual effect to the human eyes. In the visible spectrum, eyes are most sensitive to yellow light at the wavelength of 555 nm and become less sensitive to the two sides of the spectrum. Hence, we use the luminous flux to define the radiant power that can be sensed by the eyes, which is equal to the product of the radiant power in some wavelength band and the corresponding relative vision rate per unit time [27]. The luminous flux and luminous efficiency of the QD-converted LEDs with different contents of ZnO nanoparticles are plotted in Fig. 7; the measurement was conducted by a calibrated

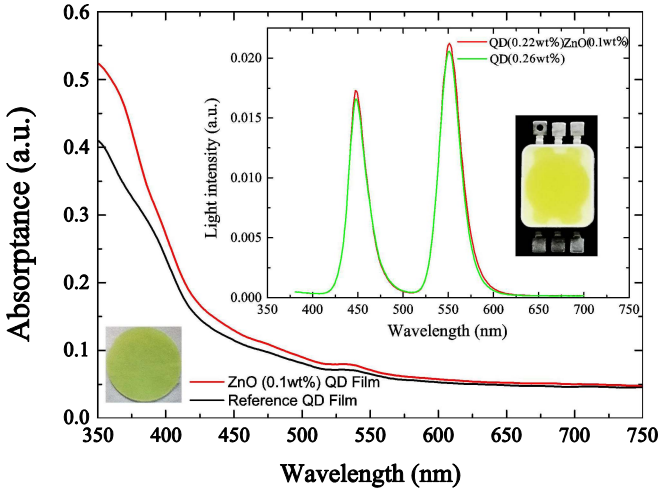

Fig. 8. Absorption spectra of the reference QD film (without ZnO) and the optimal QD film (0.1 wt% ZnO incorporated). The inset shows the emission spectra of 0.1 wt% ZnO-incorporated QD-LED (0.22 wt% QD), and none-ZnO QD-LED (0.26 wt% QD) under the similar color coordinate.

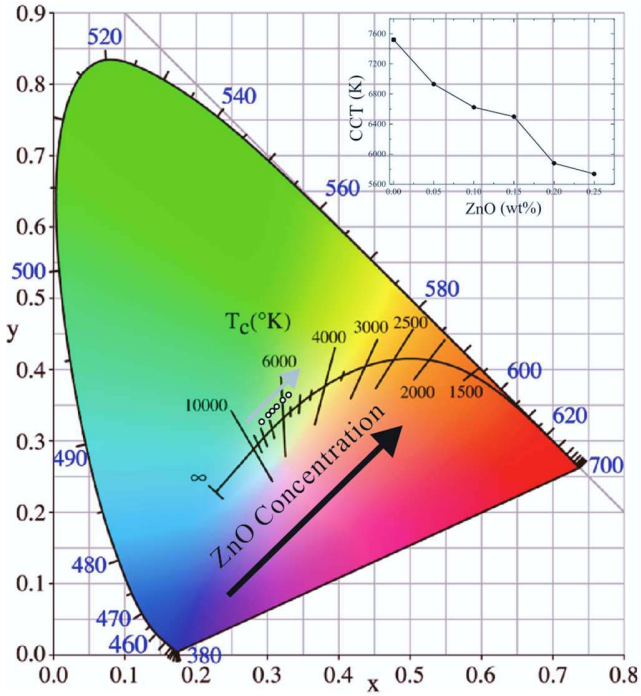

Fig. 9. Chromaticity coordinates and CCT of the QD-converted LEDs with different amounts of ZnO nanoparticles.

integrating sphere. In contrast to the radiation flux, the statistics show that the luminous flux initially increased with the amount of ZnO particles and reached its peak value when the content of ZnO nanoparticles was 0.1 wt%. This value was 7.05% higher than that of the reference QD-converted LED without ZnO. The results indicate that incorporating with ZnO nanoparticles enhanced the scattering ability of the blue light inside the encapsulation materials, resulting in the increase of the yellow-light conversion by the QDs, and the higher yellow-light ratio eventually led to a higher luminous-flux performance. When the content of ZnO nanoparticles increased to 0.25 wt%, the luminous flux decreased to 4.61% less than that of the reference QD-converted LED. This is because the whole radiation flux seriously decreased due to the strong backscattering of ZnO nanoparticles, and the higher yellow light-to-blue light ratio did not help to increase the luminous flux in this situation.

To support the aforementioned analysis, the relevant absorption spectra of the reference QD film and the optimal 0.1 wt% ZnO-incorporated QD film were tested and showed in Fig. 8. Compared with that of the conventional reference structure, the absorptance in the 0.1 wt% ZnO nanoparticles incorporating structure was found to have an 11.61% increase at the wavelength of 450 nm, which proved that ZnO nanoparticles could effectively increase the blue light absorption rate of QDs. For the exact comparison of luminous flux, the color coordinate of the LED samples must be kept as a constant value, thus we make another none-ZnO QD-converted LED ($x = 0.30048$, $y = 0.34643$) which has the similar color coordinate with the 0.1 wt% ZnO-incorporated QD-converted LED ($x = 0.30378$, $y = 0.34822$) by increasing the content of QDs (increased from 0.22 to 0.26 wt%). The inset of Fig. 8 shows the emission spectra of these two kinds of QD-converted LED correspondingly. Comparing to the 0.26 wt% QD content LED, 0.1 wt% ZnO-incorporated QD-converted LED has higher blue- and yellow-lights energy simultaneously. As marked with the green dashed line in Fig. 7, the 0.1 wt% ZnO-incorporated QD-converted LED could reach a 3.37% higher luminous-flux performance even with less QD content under the similar color coordinate, which means ZnO could reduce TIR and effectively increase the transfer efficiency of QDs. To sum up, the absorption and emission spectra strongly indicate that incorporating low content of ZnO nanoparticles could effectively enhance the luminous efficiency of the QD-converted LED under the sufficient radiation flux. Besides, since the QDs price is still high in the market, it is promising to reduce the dosage of QDs while maintain the luminous flux of QD-converted LED by incorporating ZnO nanoparticles.

Fig. 9 shows the chromaticity coordinates of the QD-converted LEDs with different ZnO contents. The chromaticity coordinates gradually shifted to the yellow region as the concentration of ZnO nanoparticles increased, which can be explained by the relatively high yellow light-to-blue light ratios caused by the stronger scattering effect. Correspondingly, the CCTs tested by the integrating spheres are shown in the inset, as the ZnO nanoparticles concentration increased, the CCT monotonically decreased from 7521 to 5757 K. This phenomenon indicates that a soft-optical performance QD-device with a low QD concentration can be achieved by incorporating ZnO nanoparticles to reduce the CCT.

To further investigate the scattering effect of the ZnO nanoparticles and discuss the light distribution in the encapsulation materials thoroughly, the angular-dependent CCT of the QD-converted LEDs with different ZnO contents was tested by the spatial light distribution instrument and plotted in Fig. 10 with black line. In general, the uniformity of the CCT is defined as the maximum CCT minus the minimum

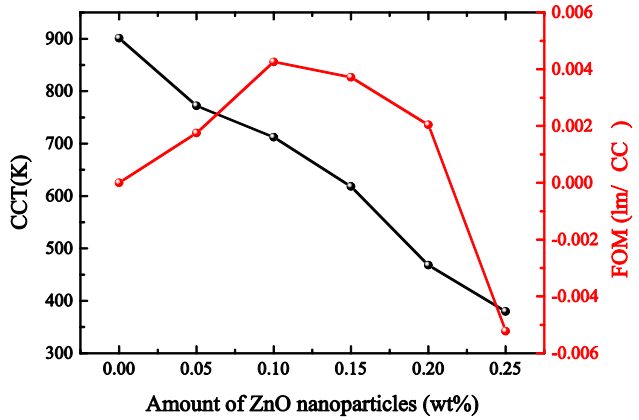

Fig. 10. CCT deviation and FOM of the QD-converted LEDs with different amounts of ZnO nanoparticles.

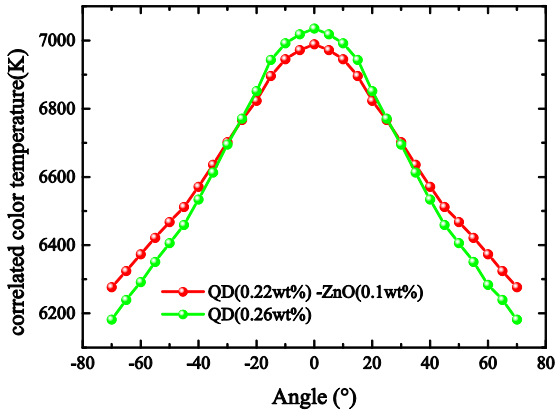

Fig. 11. Angular-dependent CCT of conventional and ZnO-incorporated QD-LED under the similar color coordinate.

CCT, and the results show that the CCT deviation decreased monotonically as the content of ZnO nanoparticles increased. Compared with the reference QD-converted LED without ZnO, the CCT deviation of the 0.25 wt% ZnO-incorporated sample was improved from 901 to 380 K in the range of −70° to 70°. This is because the blue LED chip is a Lambertian emitter, and the blue-light intensity is highest in the middle and lowest in the margin. However, the emission of yellow light from the QDs was uniform; thus, the extracted yellow light and blue light mixed ineffectively and led to an extremely high yellow light-to-blue light ratio and low CCT at large angles. With the ZnO incorporating, the QD-converted LEDs had a lower CCT deviation. This phenomenon indicates that the blue-light intensity increased at large angles because the scattering effect of the ZnO nanoparticles redirected the Lambertian blue-light source more uniformly in the encapsulation materials. Thus, the blue light and yellow light remixed more uniformly and were extracted from the package together to reach a higher CCT at large angle. To make exact comparison, we measured the angular-dependent CCT of 0.26 wt% QD-LED and 0.1 wt% ZnO–0.22 wt% QD-LED with the similar color coordinate, and the result is illustrated in Fig. 11. It is clear that the CCT deviation of conventional and ZnO-incorporated structure are improved from 862 to 712 K in the range of −70° to 70°.

Besides, for the luminous flux, 0.1 wt% ZnO content is best for QD-converted LED. However, the uniformity of CCT is proportional to the amount of ZnO. Thus, we introduce the figure of merit (FOM) to optimize the content of ZnO to satisfy both properties in QD-converted LED [19], defined as

$$\mathrm{FOM} = \frac{\mathrm{Lumen}_{\mathrm{ZnO}} - \mathrm{Lumen}_{\mathrm{No\,ZnO}}}{\Delta \mathrm{CCT}}. \tag{1}$$

The relative result is shown in Fig. 10 with red line, according to the definition of FOM, the optimal ZnO-incorporated concentration was discovered to be at 0.1 wt%.

IV. Conclusion

Different mass fractions of ZnO nanoparticles were incorporated into the encapsulation materials to investigate the optical performance of ZnO-incorporated QD-converted LEDs, whereby a higher luminous flux and superior uniformity of the angular CCT were achieved. The ZnO-only film was introduced first and proved that the higher concentration of ZnO nanoparticles yields the stronger scattering ability. The radiation flux decreased monotonically as the amount of ZnO nanoparticles increased because an excessive content of ZnO nanoparticles saturated the absorption of QDs, stopping the yellow light from further exciting, while the backscattering of the ZnO nanoparticles led to continuous energy loss. Under the similar color coordinate, the luminous flux of the 0.1 wt% ZnO-incorporated QD-converted LED increased by 3.37% compared with the conventional device without ZnO, because the strong scattering effect of the ZnO nanoparticles caused higher blue-light utilization and higher yellow-light conversion ratio under a sufficient radiation-flux performance. Besides, this novel structure reduced angular-dependent CCT deviations from 862 to 712 K in the range of −70° to 70° for redistributing the blue-light extraction by ZnO nanoparticles. Moreover, the CCT monotonically decreased, and the chromaticity coordinate steadily shifted to the yellow region as the amount of ZnO nanoparticles increased.

In addition to the promotion of luminous efficiency and CCT, we plan to mix yellow QDs and red QDs in the ZnO-incorporated LEDs to reach high color quality in the future, exploring the effect of nanoparticles on the multiple QDs encapsulation. We believe that our findings will significantly contribute to the development of QD-LED lighting applications.

References

[1] Y. Jin and X. Gao, "Plasmonic fluorescent quantum dots," *Nature Nanotechnol.*, vol. 4, no. 9, pp. 571–576, 2009, doi: 10.1038/nnano.2009.193.

[2] D. V. Talapin and J. Steckel, "Quantum dot light-emitting devices," *MRS Bull.*, vol. 38, no. 9, pp. 685–695, 2013, doi: 10.1557/mrs.2013.204.

[3] R. Chen, B. Han, L. Yang, Y. Yang, Y. Xu, and Y. Mai, "Controllable synthesis and characterization of CdS quantum dots by a microemulsion-mediated hydrothermal method," *J. Luminescence*, vol. 172, pp. 197–200, Apr. 2016, doi: 10.1016/j.jlumin.2015.12.006.

[4] X. Li *et al.*, "$CsPbX_3$ quantum dots for lighting and displays: Room-temperature synthesis, photoluminescence superiorities, underlying origins and white light-emitting diodes," *Adv. Funct. Mater.*, vol. 26, no. 15, pp. 2435–2445, 2016, doi: 10.1002/adfm.201600109.

[5] R. Freeman, J. Girsh, and I. Willner, "Nucleic acid/quantum dots (QDs) hybrid systems for optical and photoelectrochemical sensing," *ACS Appl. Mater. Inter.*, vol. 5, no. 8, pp. 2815–2834, 2013, doi: 10.1021/am303189h.
[6] M. Stanisavljevic, S. Krizkova, M. Vaculovicova, R. Kizek, and V. Adam, "Quantum dots-fluorescence resonance energy transfer-based nanosensors and their application," *Biosensors Bioelectron.*, vol. 74, pp. 562–574, Dec. 2015, doi: 10.1016/j.bios.2015.06.076.
[7] J. Zhou, Y. Yang, and C.-Y. Zhang, "Toward biocompatible semiconductor quantum dots: From biosynthesis and bioconjugation to biomedical application," *Chem. Rev.*, vol. 115, no. 21, pp. 11669–11717, 2015, doi: 10.1021/acs.chemrev.5b00049.
[8] J. S. Li *et al.*, "ACU optimization of pcLEDs by combining the pulsed spray and feedback method," *J. Display Technol.*, vol. 12, pp. 1229–1234, 2016, doi: 10.1109/JDT.2016.2588419.
[9] R. Liang *et al.*, "Quantum dots-based flexible films and their application as the phosphor in white light-emitting diodes," *Chem. Mater.*, vol. 26, no. 8, pp. 2595–2600, 2014, doi: 10.1021/cm404218y.
[10] A. B. Greytak *et al.*, "Alternating layer addition approach to CdSe/CdS core/shell quantum dots with near-unity quantum yield and high on-time fractions," *Chem. Sci.*, vol. 3, no. 6, pp. 2028–2034, 2012, doi: 10.1039/C2SC00561A.
[11] C. Ippen *et al.*, "ZnSe/ZnS quantum dots as emitting material in blue QD-LEDs with narrow emission peak and wavelength tunability," *Organic Electron.*, vol. 15, no. 1, pp. 126–131, 2014, doi: 10.1016/j.orgel.2013.11.003.
[12] V. Dzhagan, I. Lokteva, C. Himcinschi, X. Jin, J. Kolny-Olesiak, and D. R. T. Zahn, "Phonon Raman spectra of colloidal CdTe nanocrystals: Effect of size, non-stoichiometry and ligand exchange," *Nanosc. Res. Lett.*, vol. 6, no. 1, p. 79, 2011, doi: 10.1186/1556-276X-6-79.
[13] Y.-K. Kim, K.-C. Choi, S.-H. Ahn, and Y.-S. Cho, "A facile synthesis of SiO_2-based nanocomposites containing multiple quantum dots at high concentration for LED applications," *RSC Adv.*, vol. 2, no. 16, pp. 6411–6413, 2012, doi: 10.1039/c2ra20780j.
[14] J. Li, Z. Li, Z. Li, Y. Tang, X. Ding, and B. Yu, "Improvement in optical performance and color uniformity by optimizing the remote phosphor caps geometry for chip-on-board light emitting diodes," *Solid State Electron.*, vol. 126, pp. 36–45, Dec. 2016, doi: 10.1016/j.sse.2016.09.020.
[15] Z.-T. Li, Q.-H. Wang, Y. Tang, C. Li, X.-R. Ding, and Z.-H. He, "Light extraction improvement for LED COB devices by introducing a patterned leadframe substrate configuration," *IEEE Trans. Electron Devices*, vol. 60, no. 4, pp. 1397–1403, Apr. 2013, doi: 10.1109/TED.2013.2248063.
[16] M.-H. Shin, H.-G. Hong, H.-J. Kim, and Y.-J. Kim, "Enhancement of optical extraction efficiency in white LED package with quantum dot phosphors and air-gap structure," *Appl. Phys. Exp.*, vol. 7, no. 5, pp. 052101-1–052101-4, 2014, doi: 10.7567/APEX.7.052101.
[17] B. Xie, R. Hu, X. Yu, B. Shang, Y. Ma, and X. Luo, "Effect of packaging method on performance of light-emitting diodes with quantum dot phosphor," *IEEE Photon. Technol. Lett.*, vol. 28, no. 10, pp. 1115–1118, May 15, 2016, doi: 10.1109/LPT.2016.2531794.
[18] D. W. Ayele *et al.*, "Controlled synthesis of CdSe quantum dots by a microwave-enhanced process: A green approach for mass production," *Chem. Eur. J.*, vol. 17, no. 20, pp. 5737–5744, 2011, doi: 10.1002/chem.201003686.
[19] K.-J. Chen *et al.*, "White light emitting diodes with enhanced CCT uniformity and luminous flux using ZrO_2 nanoparticles," *Nanoscale*, vol. 6, no. 10, pp. 5378–5383, 2014, doi: 10.1039/c3nr06894c.
[20] H.-C. Chen *et al.*, "Improvement in uniformity of emission by ZrO_2 nano-particles for white LEDs," *Nanotechnology*, vol. 23, no. 26, p. 265201, 2012, doi: 10.1088/0957-4484/23/26/265201.
[21] H. Zheng, L. Li, X. Lei, X. Yu, S. Liu, and X. Luo, "Optical performance enhancement for chip-on-board packaging LEDs by adding TiO_2/silicone encapsulation layer," *IEEE Electron Device Lett.*, vol. 35, no. 10, pp. 1046–1048, Oct. 2014, doi: 10.1109/LED.2014.2349951.
[22] T.-H. Kim, S. Jun, K.-S. Cho, B. L. Choi, and E. Jang, "Bright and stable quantum dots and their applications in full-color displays," *MRS Bull.*, vol. 38, no. 9, pp. 712–720, 2013, doi: 10.1557/mrs.2013.184.
[23] J. Lang *et al.*, "Synthesis, characterization and photoluminescence property of La-doped ZnO nanoparticles," *Appl. Phys. A Mater. Sci. Process.*, vol. 122, no. 10, p. 873, 2016, doi: 10.1007/s00339-016-0408-x.
[24] Q. Chen, Z. Li, K. Chen, Y. Tang, X. Ding, and B. Yu, "CCT-tunable LED device with excellent ACU by using micro-structure array film," *Opt. Exp.*, vol. 24, no. 15, pp. 16695–16704, 2016, doi: 10.1364/OE.24.016695.
[25] T. Monteiro *et al.*, "Optical and structural analysis of bulk ZnO samples undoped and rare earth doped by ion implantation," *Superlattices Microstruct.*, vol. 39, nos. 1–4, pp. 202–210, 2006, doi: 10.1016/j.spmi.2005.08.043.
[26] Y.-J. Lee, C.-J. Lee, and C.-M. Cheng, "Enhancing the conversion efficiency of red emission by spin-coating CdSe quantum dots on the green nanorod light-emitting diode," *Opt. Exp.*, vol. 4, no. 23, pp. A554–A561, 2010, doi: 10.1364/OE.18.00A554.
[27] P. P. Zak and N. N. Trofi, "Spectral dependence of visual functions when comparing characteristics of white light emitting diodes," *Light & Eng.*, vol. 22, no. 1, pp. 57–60, 2014.

Yong Tang received the Ph.D. degree in mechanical engineering from the South China University of Technology, Guangzhou, China, in 1994.

He is currently a Professor with the School of Mechanical and Automotive Engineering, South China University of Technology, where he is also the Director of the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology.

Zhi Li received the B.E. degree in mechanical engineering and automation from the South China University of Technology, Guangzhou, China, in 2016, where he is currently pursuing the master's degree with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology.

His current research interests include LED packaging process and reliability.

Zong-Tao Li (M'16) received the Ph.D. degrees in microelectronics manufacture engineering and mechanical engineering from the South China University of Technology, Guangzhou, China, in 2014.

He is currently with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology, South China University of Technology, and also with the Optoelectronics Engineering Technology Research and Development Center, Foshan Nationstar Optoelectronics Co., Ltd., Foshan, China.

Jia-Sheng Li received the M.E. degree in mechanical engineering from the South China University of Technology, Guangzhou, China, in 2017, where he is currently pursuing the Ph.D. degree with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology.

His current research interests include simulation, packaging, and application of LED.

Shu-Dong Yu received the M.E. degree in mechanical engineering from the South China University of Technology, Guangzhou, China, in 2017, where he is currently pursuing the Ph.D. degree with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology.

His current research interests include packaging and application of quantum dots-LED.

Long-Shi Rao received the M.E. degree in mechanical engineering from the South China University of Technology, Guangzhou, China, in 2017, where he is currently pursuing the Ph.D. degree with the Engineering Research Center of Green Manufacturing for Energy-Saving and New-Energy Technology.

His current research interests include synthesis, application, and reliability of quantum dots.