The Influence of the Thermal Effect on CdSe/ZnS Quantum Dots in Light-Emitting Diodes

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***Abstract*—This study investigates the effect of temperature on CdSe/ZnS quantum dots (QDs) in GaN-based light-emitting diodes (LEDs) using the phosphor conversion efficiency (PCE) and LED junction temperature. In our simulation, the blue chip and CdSe/ZnS QDs temperature are similar because of their minimal thickness. Furthermore, to verify the effect of temperature on CdSe/ZnS QDs, we use continuous wave and pulsed current sources to measure the relationship between the temperature and relative PCE. Higher junction temperatures are observed with greater CdSe/ZnS QD volume in LEDs. This is attributed to the thermal conduction and nonradiative energy between CdSe/ZnS QDs and blue chip. Therefore, if thermal management is improved, CdSe/ZnS QDs are expected to be used as color converting material in LEDs.**

***Index Terms*—Light-emitting diodes (LEDs), GaN, quantum dots (QDs), phosphor.**

I. INTRODUCTION

RECENTLY, the development of light-emitting diodes (LEDs) has been widely studied for practical applications in the field of solid-state lighting. The advantages of LEDs are a longer lifetime, higher energy efficiency, and greater reliability [1], [2]. Several methods are used to produce white light. One of the methods is to combine red, green, and blue LEDs to create white light. Therefore, great efforts have been made to improve the internal quantum efficiency (IQE) of LEDs, especially the large electron–hole wave function overlap quantum wells. The enhancement of spontaneous emission leads to the higher efficiency in green, yellow, and red spectral regime [3]–[7]. Indeed, the surface plasmon approach is also indicated to enhance the emission efficiency for multiple quantum wells (MQWs) [8], [9]. For the phosphor-free white LEDs, the laterally stacked structure of blue and green InGaN/GaN MQWs and monolithic polychromatic (LEDs) are demonstrated to grow white LEDs by multiple emission spectral and become the multicolor light-emitting sources [10], [11]. In addition, the ZnO current spreading is employed to compare the ITO due to lower sheet resistance and lower optical absorption [12].

Other methods use UV-LEDs to excite red, green, and blue phosphors. However, these two methods are not appropriate for high-power white LEDs because of their high cost and low conversion efficiency [13]. Thus, the primary strategy is to combine blue LEDs with yellow luminescence from $Y_3Al_5O_{12}$:Ce^{3+}(YAG:Ce^{3+}) phosphor materials because of their high luminous flux and low cost. Although this strategy is widely used, the primary disadvantage of this type of LEDs is the poor color rendering index (CRI), which is caused by a lack of red components in the spectra. Numerous studies to solve the low CRI of commercially available phosphor, such as through adding red phosphor, are currently in progress. Therefore, red phosphor is recognized as an important issue in current research such as nitride-based, vanadates, and borate red-emitting phosphor [14]–[16]. Consequently, to achieve a high CRI, the phosphor-converted LEDs must have a broad-band-covered visible spectrum [17].

Colloidal CdSe/ZnS quantum dots (QDs) that exhibit quantum confinement effects were created during investigations into possible applications of QDs in solid-state lighting [18], [19]. The high photoluminescence (PL) efficiency wide absorption spectrum and size-tunable bandgap of QDs have become a significant concern [20], [21]. CdSe/ZnS QDs appear feasible for use as color conversion nanophosphors to improve the CRI for solid-state lighting. Although the conversion efficiency of phosphor is higher than QDs, the main problems for the rare-earth-based phosphors suffer the chemically unstable and higher cost when using in the white LEDs [22]. Several studies focus on the improvement of the QDs quantum yield with multishell structure and demonstrated the better photoluminescent efficiency [23]. More importantly, the various diameters of CdSe/ZnS QDs offer the suitable solution for the use in the display devices. Consequently, colloidal QDs have recently been used as color converting material in LEDs. The excellent CRI of white LEDs can be obtained using CdSe QDs and Sr_3SiO_5:Ce^{3+},Li^+ [24]. Nizamoglu *et al.* developed a warm white light that incorporates green and red CdSe/ZnS core-shell nanocrystals on blue chips [25]. The conversion efficiency of green nanorod LEDs can be enhanced by red CdSe QDs [26].

However, thermal issues also have a strong effect on the performance of LEDs. As heat accumulates, the junction temper-

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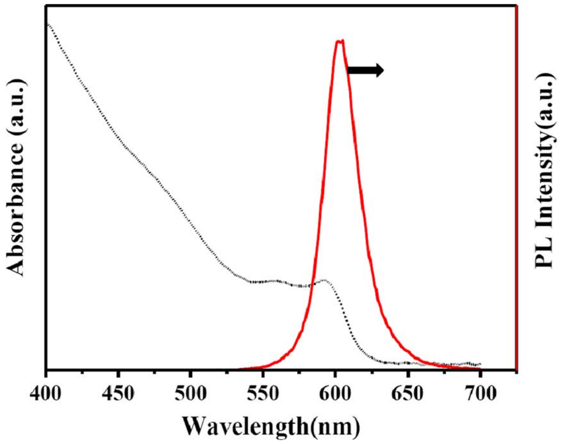

Fig. 1. PL and absorption spectra of CdSe/ZnS QDs.

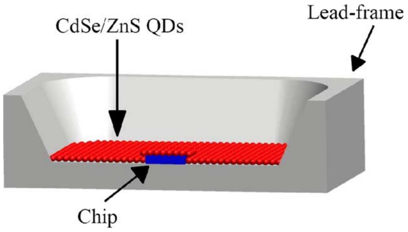

Fig. 2. Schematic diagram of a CdSe/ZnS QDs LED device.

ature increases. Although numerous studies have incorporated QDs with LEDs to improve the CRI value, few studies [27] have investigated the effect of temperature on CdSe/ZnS QDs.

Therefore, this study examines the effect on CdSe/ZnS QDs in LEDs and characterizes the activated energy. We simulate the temperature of blue chips and CdSe/ZnS QDs of varying thicknesses using finite element method (FEM) simulations. The temperature increases with a higher volume of CdSe/ZnS QDs. Furthermore, we discuss the influence of thermal effects on CdSe/ZnS QDs using the phosphor conversion efficiency (PCE) and junction temperature.

II. Experiment

The CdSe/ZnS QDs were synthesized using the hot-injection method [28], [29]. Cadmium acetate was dissolved into trioctylphoshine oxide and hexadecylamine solvent at 300 °C. The Se precursor was dissolved in trioctylphosphine. Under an Ar atmosphere, the temperature was increased to the injection temperature, and the TOP solution was injected rapidly. After cooling, CdSe QDs were obtained. To achieve greater efficiency, a zinc precursor solution and sodium sulfide were mixed into the previous solution. The concentration of CdSe/ZnS QDs with toluene was 5 mg/mL.

Fig. 1 shows the PL and the absorption spectra of CdSe/ZnS QDs. The emission peak wavelength of CdSe/ZnS QDs occurs at 605 nm, with a full-width at half-maximum (FWHM) of 29 nm. The absorption spectrum shows that nearly all visible wavelengths, including blue LEDs, can be absorbed by CdSe/ZnS QDs. A schematic diagram of CdSe/ZnS LEDs is shown in Fig. 2. The chip size of the LED was 600 × 600 μm^2, and the peak emission wavelength was 450 nm, with an FWHM of 20 nm. The blue chips were bonded in a lead frame with silver glue; the radiant fluxes were 100 mW at 120 mA. The CdSe/ZnS QDs were spin-coated onto the surface of blue LEDs [26]. The deviation of QDs thickness is approximately 10–15% with current coating method. The uniformity could be much improved with new techniques [30], [31].

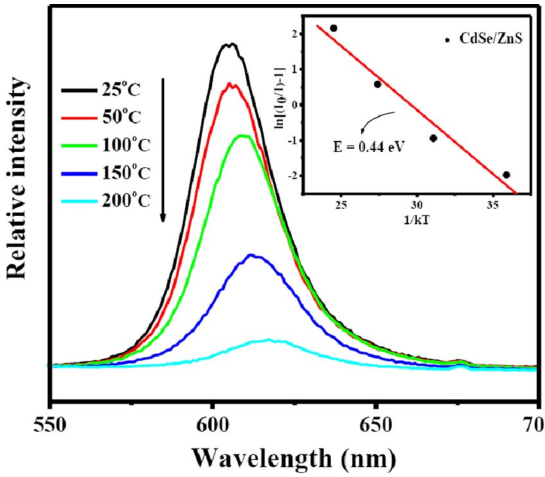

Fig. 3. Temperature dependence of the CdSe/ZnS QD emission spectra. The inset shows the fitted activation energy for thermal quenching.

To investigate the thermal effects between LEDs and CdSe/ZnS QDs, we adopted a temperature-dependent PL measurement for QDs and simulated the temperature of different layers. Additionally, we used both continuous wave (CW) and pulse current sources to verify the effect of temperature on the PCE of QDs in LEDs. Finally, the junction temperature of LEDs with QDs was also examined.

III. Result and Discussion

Fig. 3 shows the temperature-dependent PL measurement of CdSe/ZnS QDs between 25 °C and 200 °C. The excitation wavelength was 450 nm, which equals the emission of blue LEDs. The emission intensity of QDs decreases as the temperature increases. The relative peak intensity of CdSe/ZnS QDs reaches 76% of the initial value at 100 °C and 43% at 150 °C. With an increase in temperature, the ligand is broken, and the QDs are aggregated and enlarged [32]. According to the quantum confinement effect, the band structure is altered, leading to the red shift emission wavelength.

To understand the thermal behavior and determine the activation energy for thermal quenching at various temperatures, the temperature-dependent results were fitted using the Arrhenius equation [33]:

$$I(T) = \frac{I_0}{1 + c\,e^{-E/KT}}$$

where I_0 is the initial intensity, $I(T)$ is the intensity at a given temperature T, c is a constant, E is the activation energy for thermal quenching, and K is the Boltzmann's constant. The fitting curve of the PL results is shown in the inset of Fig. 3. The activation energy (E) of CdSe/ZnS QDs is 0.44 eV. The low activation energy leads to QD instability against heat.

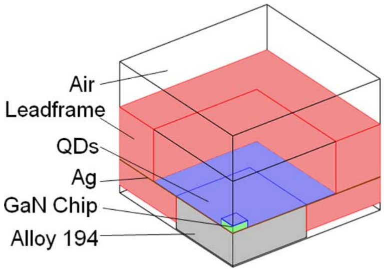

Fig. 4. Schematic cross-sectional view of CdSe/ZnS LEDs in simulation.

TABLE I
THICKNESS PARAMETER AND THERMAL CONDUCTIVITY OF DIFFERENT LAYERS

No.	Layer	Thickness (mm)	Thermal conductivity (W/m · K)
1	**Alloy 194**	**0.5**	**260**
2	**Ag**	**0.0025**	**428**
3	**GaN Chip**	**0.111**	**245**
4	**Leadframe**	**1.2**	**0.34**
5	**CdSe/ZnS**	**0.00008**	**4**
	QDs	**0.00016**	**4**
		0.00024	**4**

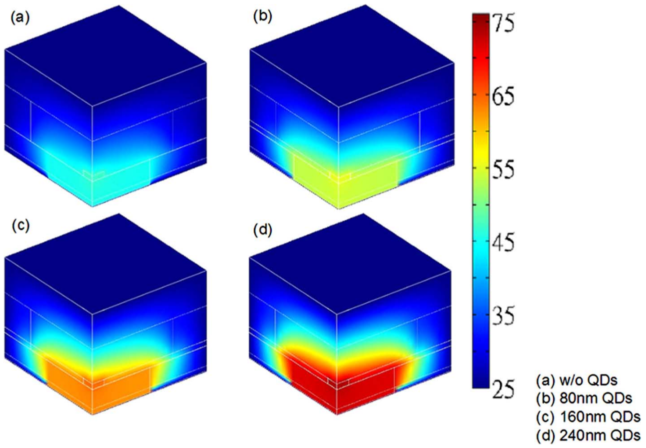

Fig. 5. Simulated thermal distribution in (a) blue chip with a thickness of (b) 80 nm (c) 160 nm (d) and 240 nm in a CdSe/ZnS LED.

To explore the thermal phenomenon, we use the FEM to simulate the temperature of CdSe/ZnS LEDs. The simulated model, which is similar to the real sample, is shown in Fig. 4. Furthermore, the thickness and thermal conductivity of each layer of CdSe/ZnS QDs are listed in Table I.

Fig. 5 shows the temperature distribution in blue chips and differing CdSe/ZnS QD thicknesses. Regarding the thermal distribution, the temperature increases with the thickness of CdSe/ZnS QDs and the maximum temperature exists in the center of LEDs. In addition, the junction temperature and the temperature of CdSe/ZnS QDs are similar for each thickness, as shown in Fig. 6. This is attributed to the minimal thickness of CdSe/ZnS QDs, which differ from phosphor.

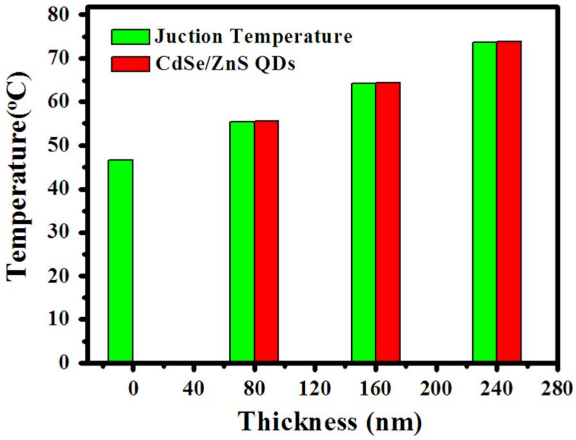

Fig. 6. Simulated junction temperature and CdSe/ZnS QDs temperature with different QDs layer thicknesses.

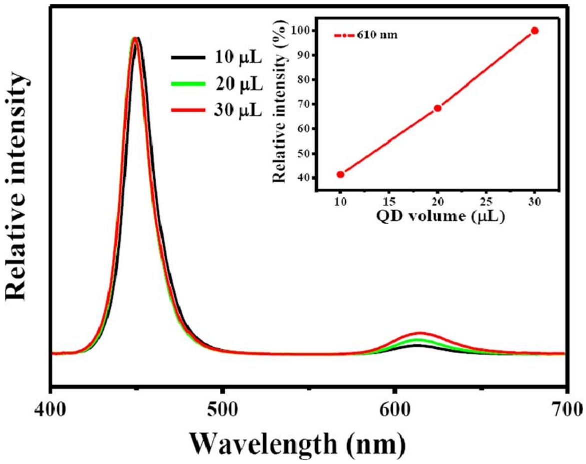

Fig. 7. EL spectra of blue chips with different ratios of CdSe/ZnS QDs at 120 mA; the inset plots the enlargement EL from 550 to 700 nm.

Furthermore, to understand the interacting effect of temperature between QDs and LEDs, we combined CdSe/ZnS QDs with InGaN blue chips using the spin-coating method. The normalized electroluminescence (EL) spectra of LEDs with various volumes of CdSe/ZnS QDs at 120 mA are shown in Fig. 7. Two emission bands occurred at 450 nm (blue band) and 613 nm (red band), which were contributed by InGaN blue chips and CdSe/ZnS QDs, respectively. The inset shows that the red band increases with the volume of CdSe/ZnS QDs at 120 mA. With greater CdSe/ZnS QD volume, more red emission can be obtained, thereby achieving high CRI in LEDs. Differing red band volumes do not cause a red shift under the use of a pulse current source, which isolates the interval of each current source. The thermal effect on CdSe/ZnS QDs is significantly related to the PCE, which can be defined as [17]

$$\eta_{\text{pce}} = \frac{P_R}{P_{B_0} - P_B}$$

where P_R is the emission intensity of the red band; P_{B0} and P_B are the emission intensity of blue LEDs with and without the addition of CdSe/ZnS QDs, respectively. Fig. 8 demonstrates the relative efficiency of a constant current source and a pulse

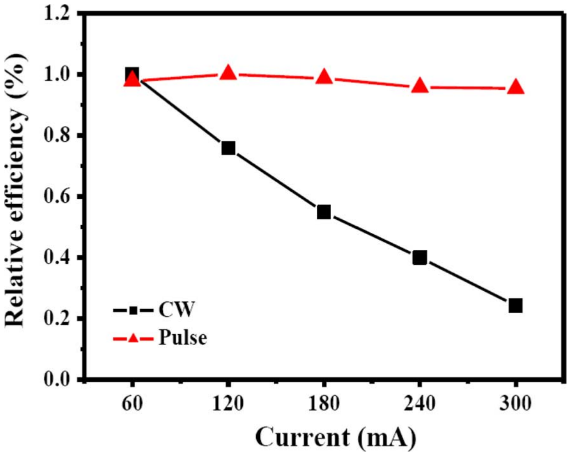

Fig. 8. Relative efficiency of LEDs pumped by a CW current source and a pulse source at different currents.

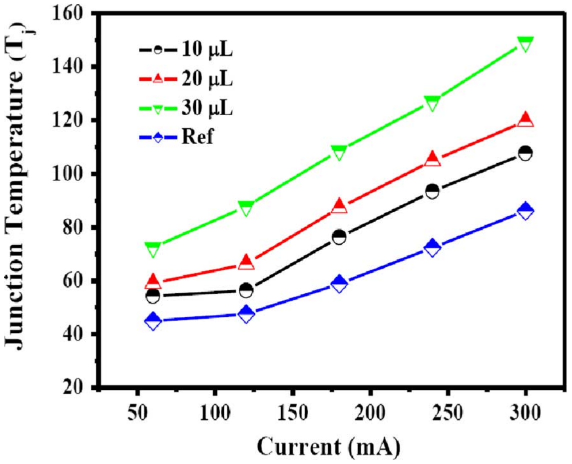

Fig. 9. Junction temperatures of differing volumes of CdSe/ZnS QDs under currents between 60 and 300 mA.

source at different currents. The pulsewidth and duty cycle was set at 1 μs and 1%, respectively. Assuming that both the LEDs and CdSe/ZnS QDs generate no heat under pulse operation, the relative PCE exhibits a similar value with various operation currents. This phenomenon indicates that the reduction of PCE was not because of QD saturation, but primarily because of the thermal effect.

The junction temperature can be measured using the forward voltage method, which is related to the thermal characteristics. Additionally, the influence of CdSe/ZnS QDs on LEDs was presented using another method, i.e., the junction temperature of LEDs with varying volumes of QDs and operation currents, as shown in Fig. 9. LEDs with 10, 20, and 30 μL of CdSe/ZnS QDs were compared with bare LEDs. The junction temperature of LEDs increased as the current increased. However, the junction temperature was further increased in correlation with the volume of QDs. The junction temperature was 47.52 °C, 56.56 °C, 66.31 °C, and 87.71°C at 120 mA for LEDs without QDs and with 10, 20, and 30 μL of CdSe/ZnS QDs, respectively. The higher junction temperature is attributed to thermal conduction and nonradiative energy, which reduces the relative PCE. We found that the junction temperature increases with differing volumes of QDs under the same current.

Therefore, extra heat is produced and back-scattered to the LED chip. This indicates that the thermal quenching issue is a substantial concern for LEDs that use QDs as color converting material. In addition, the greater volume of QDs in LEDs provides a greater backscattering of light.

The thermal effect of QDs emphasizes their importance in LEDs. Because the PCE of CdSe/ZnS or II-VI QDs is extremely sensitive to heat, the IQE of GaN LEDs and the PCE of QDs should be enhanced. To increase the PCE, the effect of temperature on CdSe/ZnSe QDs, such as in remote type LEDs, must be reduced. Therefore, QDs have significant potential for use as color converting material in LEDs if the thermal atmosphere and PCE is improved.

IV. CONCLUSION

This study presents the influence of thermal effects on CdSe/ZnS QDs in LEDs using the PCE and junction temperature. The results correspond with the simulation results using differing thicknesses of CdSe/ZnS QDs. Furthermore, the PCE and junction temperature indicate that CdSe/ZnS QDs can be used reliably in LEDs. CdSe/ZnS QDs are easily influenced by the temperature of the atmosphere. Higher junction temperatures are attributed to the thermal conduction and nonradiative energy between CdSe/ZnS QDs and blue chips. Therefore, temperature is likely to become an essential factor of CdSe/ZnS QDs LEDs. We aim to conduct a future study on avoiding the temperature influence of CdSe/ZnS QDs.

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