

Wide-Range Correlated Color Temperature Light Generation From Resonant Cavity Hybrid Quantum Dot Light-Emitting Diodes

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Abstract—This study presents extremely uniform colloidal quantum dot white light-emitting diodes (QD-WLEDs) that demonstrate a high color rendering index (CRI) and correlated color temperatures (CCTs) ranging from 2500 to 4500 K. Experimental results indicate that the structure of the distributed Bragg reflector (DBR) containing a stopband in the UV region enhances the intensity output of both monochromatic QD-LEDs and QD-WLEDs by reflecting the unconverted UV light back onto the package to excite the QDs further. Furthermore, the angular CCT uniformity of the QD-WLEDs also improved considerably because of the dependence of the DBR structure on the incident angle. The angular CCT deviation in the range of -70° to 70° decreased to 39 K and the CRI of the WLED is higher than 90. The high-CRI and uniform angular CCT QD-WLED containing the DBR demonstrates potential applicability as the lighting source of next-generation display devices and solid-state lighting.

Index Terms—GaN, light-emitting diodes (LEDs), quantum dots (QDs).

I. INTRODUCTION

LIGHT-emitting diodes (LEDs) have been developed extensively in the past decade because of their high efficiency, long lifetime, and stability in comparison to traditional incandescent and fluorescent lamps [1]–[4]. Currently, the most common technique for producing white LEDs (WLEDs) involves combining blue LEDs with yttrium aluminum garnet (YAG:Ce) phosphor because of high luminous efficiency and

low cost [5], [6]. These types of WLEDs have been commercialized and YAG:Ce has already been developed as a down-converting material; however, the problem of the instability of the color quality remains a critical challenge for phosphor based WLEDs [7]. In general, the characteristics of WLEDs such as the homogenous correlated color temperature (CCT) and high color rendering index (CRI) are considered the key factors that must be used in solid-state lighting (SSL). Therefore, several studies have proposed several structures, such as the patterned remote phosphor structure and ZrO_2 -type phosphor structure, for reducing the angular CCT deviation in WLEDs [8], [9]. Furthermore, to improve the color rendering ability of WLEDs, enhancing the CRI to more than 80 is an essential target that can be achieved by compensating the missing red component in phosphor-converted WLEDs. Consequently, some previous studies have used methods that involve adding red-light emitting phosphors or combining red LEDs to form a hybrid warm WLED package to satisfy strict lighting standards for indoor lighting [10]–[12]. However, the red-light emitting phosphors exhibit some severe limitations such as high cost, large Stokes shift losses, and reabsorption phenomenon, resulting in reduced efficiency in WLEDs [13], [14].

Recently, colloidal quantum dots (QDs) have attracted considerable scientific attention because of their unique properties such as high quantum yield, minimal backscattering, size dependent tunable bandgap, and narrow emission linewidth [15]–[18]. Core-shell structures have been employed to enhance the quantum yield of QDs and derive high photoluminescent efficiency [19], [20]. Major QD applications are currently focused on thin film display, monochromatic displays, and WLEDs [21]–[25]. Regarding the display technology, methods such as transfer printing [21], mist coating [22], inkjet printing [23], and pulse-spray coating [26] have been used for deriving uniform and independent RGB pixels. Furthermore, regarding WLEDs, previous studies have indicated that phosphor converted WLEDs mixed with red QDs could yield high color-rendering properties [27], [28]. Moreover, Nizamoglu *et al.* [29] developed warm WLEDs that incorporate green and red CdSe/ZnS core-shell nanocrystals hybridized with InGaN-GaN blue chips. The balance between color quality and intensity of QD-LEDs are imperative concerns that must be addressed.

The present study designed multicolor LEDs by combining colloidal QDs and polymethylmethacrylate (PMMA). The PMMA is a host material that is used to embed the QDs and provide a certain degree of isolation and protection. Furthermore, HfO_2/SiO_2 distributed Bragg reflector (DBR) with a stopband

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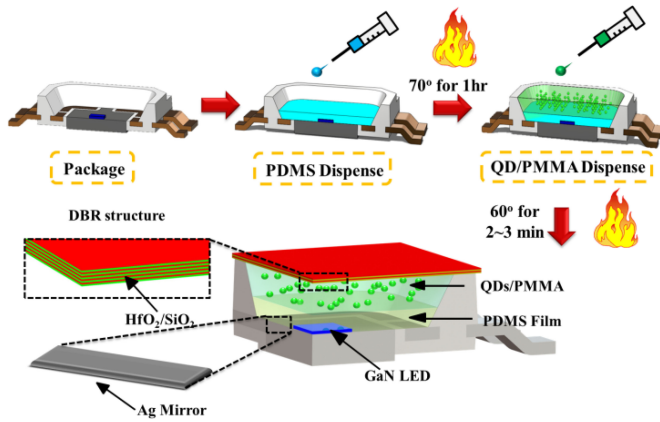


Fig. 1. Schematic flow chart of cross-sectional view DBR embedded remote QD dispense structure.

centered at 400 nm and a full width of approximately 60 nm was employed to enhance the utilization of UV light for producing a higher output intensity. Therefore, the QD-WLEDs demonstrated high CRI and excellent angular CCT ranging from 2500 to 4500 K.

II. EXPERIMENT

Fig. 1 presents a cross-sectional view of a remote QD-dispensing device with a DBR structure. For the conventional dispensing structure, the PMMA was mixed with the QDs to form a QD/PMMA blend [30]. The advantage of PMMA is that its solidifying temperature is lower than that of silicone and epoxy, which can prevent QD degradation during the curing step of the encapsulant. Furthermore, the PMMA can be readily dissolved in chloroform or chlorobenzene, which is commonly used to disperse QDs because of the concern regarding the surface of the capped organic ligands [31], [32]. Because the PMMA is compatible with QD solutions, a highly homogeneous blending of the two chemicals can be realized, and the subsequent self-clustering should be reduced. The QD-LEDs were fabricated using the following steps: 1) The LED package was first half-filled with polydimethylsiloxane (PDMS). 2) The QD/PMMA blend was mixed at a volume ratio of 2:1 and dispensed in the package to fill the remaining space in the package. 3) The QD-LED was baked at 60 °C for 2–3 min, and the DBR structure was finally placed on the surface of the package.

In the finished package, several vital components must be selected carefully to enhance the overall performance. First, the appropriate LED chip was selected. A 1000 × 1000 μm UV LED with a 380 nm emission was used because of the high luminescent efficiency of the QDs under UV excitation [33], [34]. Second, the DBR structure was used to increase the utilization of high energy photons by reflecting them back onto the QD layer. Therefore, HfO₂ and SiO₂ were employed as the dielectric materials in the DBR structure. Third, the ratios between different colors of QDs, which can affect the final CCT considerably, were determined. Table I shows the different CCT output values. Fourth, the PDMS was used to provide heat isolation to the QD layer and prevent the QD/PMMA from sifting through and reaching the LED chip. Silicone is the most frequently used LED filling materials; however, because of its

TABLE I
THE MIXING VOLUME RATIO OF RGB QDS FOR WLEDs
WITH DIFFERENT COLOR TEMPERATURES

CCT	RED	Green	Blue
2500 K	1	240	200
3500 K	1	400	300
4500 K	1	500	700

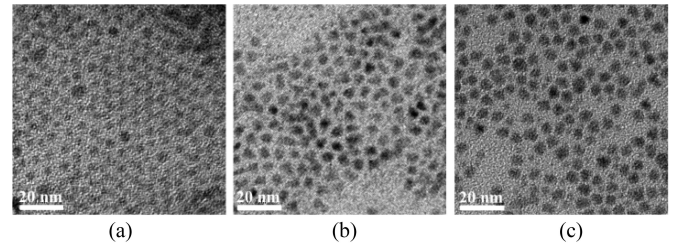


Fig. 2. TEM images of QDs with the emission colors of (a) blue, (b) green, and (c) red.

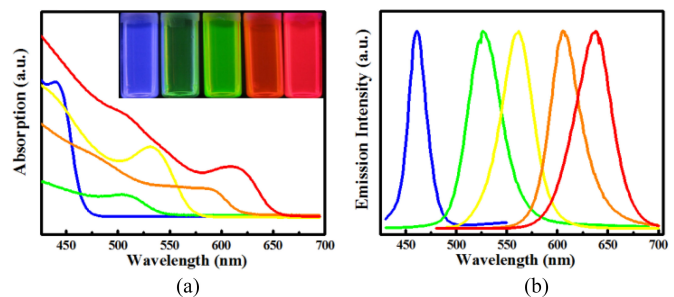


Fig. 3. (a) Absorption spectra and (b) emission spectra of blue, green, yellow, orange, and red QDs.

chemical reactivity with chlorobenzene in QD solutions, it is not suitable for this device.

III. RESULT AND DISCUSSION

Fig. 2 shows the transmission electron microscope (TEM) images of blue, green, and red QDs. The average diameters of the blue, green, and red QD samples are estimated as 3.58 ± 0.22 , 4.16 ± 0.21 , and 5.41 ± 0.34 nm, respectively; these QD samples are spherical and their sizes are uniform. Fig. 3 shows the UV-visible absorption and photoluminescence (PL) spectra of five QD products (Sigma-Aldrich Company) used in this experiment. The wavelength of peak emission of the CdS QDs is located at 460 nm and that of the CdSe/ZnS QDs is located at 530, 560, 610, and 650 nm; the full-widths at half-maximum of these products are approximately 20, 40, 40, 35, and 40 nm, respectively.

Fig. 4(a) and (b) illustrate the measured reflectivity and absorption spectra of the DBR structure comprising 11 pairs of HfO₂/SiO₂. The DBR structure was produced using an ion-assisted e-gun system to evaporate the 11 pairs of HfO₂/SiO₂. The refractive indices of the HfO₂ and SiO₂ layers are 1.9 and 1.46, respectively. The maximum reflectivity was designed for a central wavelength of 400 nm, and more than 90% of the

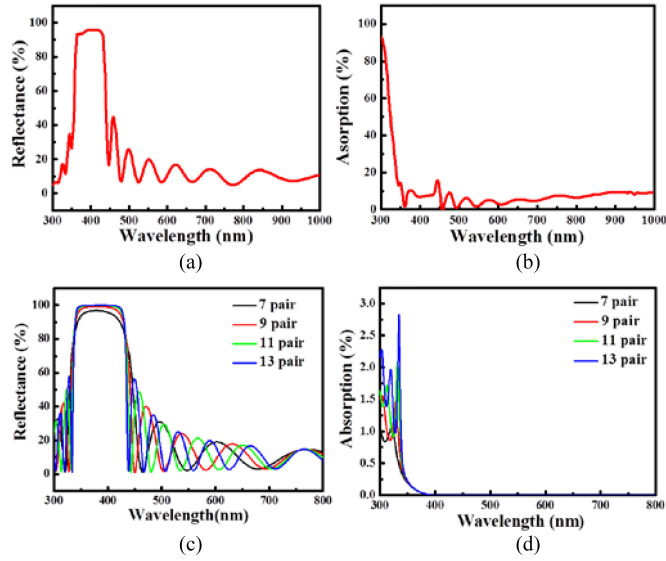


Fig. 4. The measured (a) reflectivity and (b) absorption spectra of 11 pairs of $\text{HfO}_2/\text{SiO}_2$ DBR structures with wavelengths ranging from 300 to 800 nm. The simulated (c) reflectance and (d) absorption spectra of different pairs of $\text{HfO}_2/\text{SiO}_2$ DBR structures with wavelength ranging from 300 to 800 nm.

reflectivity was maintained between 365 and 430 nm, which is sufficient for the wavelength of the InGaN UV-LED ($\lambda = 380$ nm) in the experiment conducted in this study. The reflectivity is relatively low in the visible spectral range, which enabled the transmission of visible photons produced by the QDs. The absorption loss is approximately 10% at 380 nm, and less than 10% between 450 and 700 nm. This result indicates that the DBR structure is ideal for use in the WLED. Fig. 4(c) and (d) depicts the simulated reflection and absorption spectra (for the wavelength range of 300–800 nm) of the DBR structure containing different pairs of $\text{HfO}_2/\text{SiO}_2$. Although the reflectivity increases as we increase the pair number, a sharp rise of absorption for 13 pairs of DBR indicates the upper limit of pair number shall be between 11 and 13. Thus the choice of 11 pairs can be conservative but safe to avoid extra absorption loss at UV wavelength.

Fig. 5 shows the blue, green, yellow, orange, and red monochromatic QD-LEDs at a driving current of 150 mA with peak wavelengths of 460, 530, 560, 610, and 640 nm, respectively. The results demonstrate the outstanding characteristics of the QDs, such as narrow bandwidth and size-tunable bandgap, as well as the substantial enhancement after capping the DBR structure. Additional evaluations indicated that the power intensity enhancement ratio between the samples with and without DBR for the blue, green, yellow, orange and red QD-LEDs are 14.6%, 37.8%, 38.9%, 29.4%, and 39.4%, respectively. Furthermore, the 30 nm emission bandwidth of the QDs can yield a high degree of color purity compared with the estimated 100 nm bandwidth of the monochromatic phosphor-converted LEDs [35], [36]. Fig. 5 also shows the photographs of the red, green, blue, yellow, and orange monochromatic QD-LEDs operated at 100 mA. The peak wavelength of the monochromatic QD-LEDs demonstrates a slight red shift and broadened bandwidth, which can be attributed to the CdSe QD aggregation inside the PMMA resin [37].

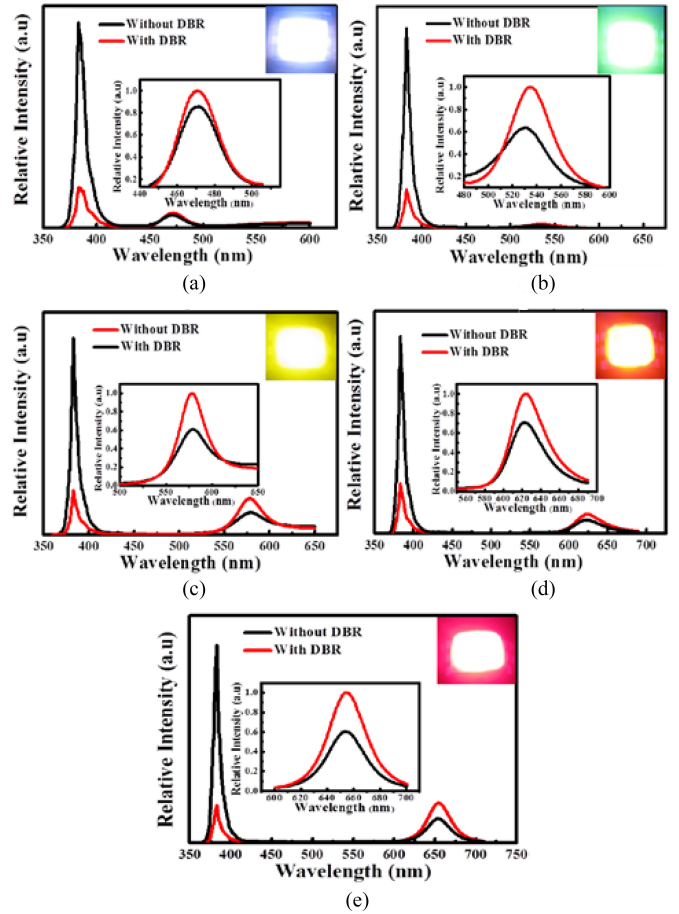


Fig. 5. (a)–(e) Relative emission spectra of monochromatic QDs with and without the DBR structure at 150 mA, and the insets indicate the images of the device with the DBR structure.

Apart from single color devices, appropriately mixed QDs can generate white light at different CCTs. WLEDs demonstrating three CCTs with and without DBR can be created using the ratios listed in Table I. Moreover, the intensity of red light clearly demonstrates a relatively high enhancement after capping the DBR. This can be attributed to reabsorption of green and blue light as well as the substantially high stability of the red QDs. The CIE color coordinates of the QD-WLEDs with DBR for three CCTs are (0.35, 0.33), (0.40, 0.41), and (0.44, 0.37), as shown in Fig. 6(d)–(f). In particular, the CIE color coordinates of the 4500 K WLED are similar to the Planckian locus, and the coordinates of the other CCT WLEDs are also consistent with the trend of the Planckian locus. When this trichromatic QD-LED method is used, the WLEDs can render light similar to natural sunlight at various CCTs and have the potential to replace traditional lighting devices.

The luminous efficiency of the LED device with the DBR structure and containing a UV LED source is approximately 8.4 lm/W, and this is consistent with the results of previous studies [38]–[40]. This improvement can be attributed to the enhancement of the UV excitation between the DBR structure and the highly reflective silver mirror at the bottom of the LED package, which excites the RGB QD and derives a higher intensity. In addition, Fig. 7(b) depicts the current-dependent CRI.

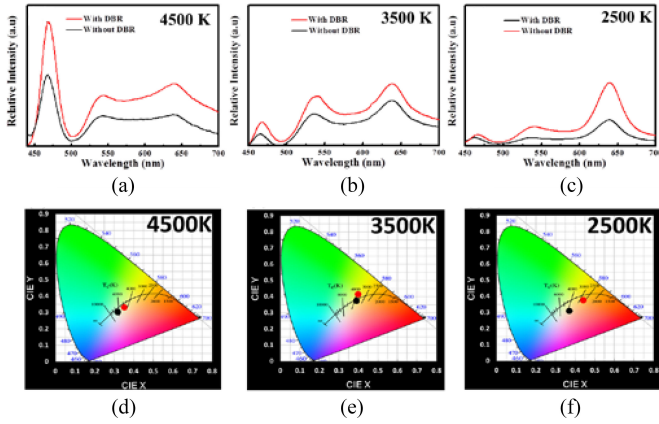


Fig. 6. (a)–(c) Emission spectra and (d)–(f) CIE chromaticity coordinate of RGB QD-WLED at CCTs of 4500, 3500, and 2500 K with and without the DBR at 150 mA.

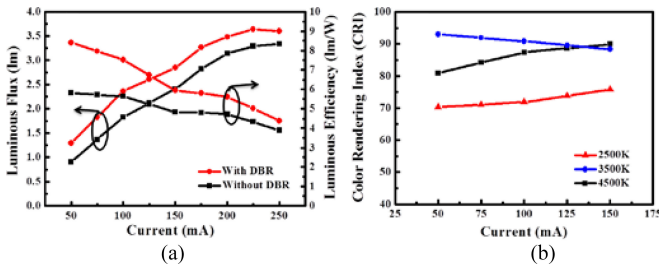


Fig. 7. Current-dependent (a) lumen and luminous efficiency of LED the device with and without the DBR structure at 3500 K (b) CRI of the RGB QD-WLEDs at various CCTs.

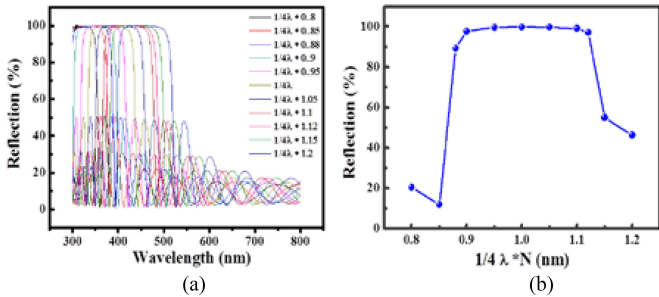


Fig. 8. (a) Simulated reflection spectra at wavelengths ranging from 300 to 800 nm. (b) The calculated reflectivity at 380nm with various DBR thickness conditions.

The 3500 and 4500 K samples register a CRI of over 90, which is prominent for indoor lighting. The CRI of the 2500 K WLEDs is low because the intensity of red light is substantially higher than that of the blue and green components. Therefore, the CRI can be improved further by fine-tuning the mixing ratio of the RGB QDs to ensure the uniformity of the relative intensities of the RGB components.

Fig. 8 shows the simulated reflection spectra at wavelengths ranging from 300 to 800 nm as well as the reflection at 380 nm. The reflectivity at 380 nm is focused here because it can be linearly correlated towards the enhancement of visible light emis-

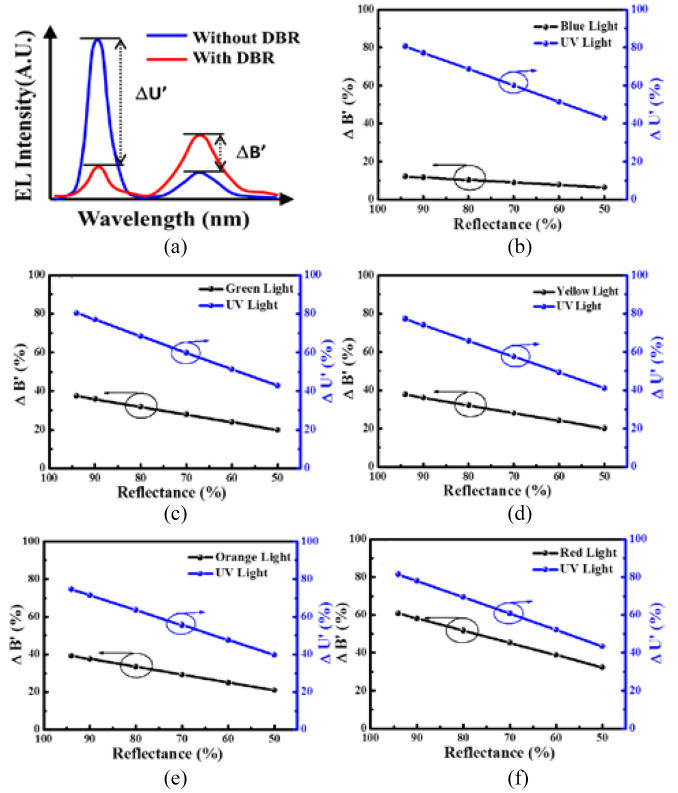


Fig. 9. (a) The illustration of the intensity spectrum for hybrid QD LED with and without DBR structure. (b)–(f) illustrate the plots of $\Delta U'$ for UV light reduction and $\Delta B'$ for the enhancement of blue, green, yellow, orange, and red monochromatic QD-LEDs. All the calculation is based on the linear extrapolation of the measured results.

sion. Under the same quantum yield of CQDs, more reflection on UV photons means more emission of visible ones.

In Fig. 8(b), a variation of 2% reflectivity at 380 nm can be maintained within the regular $\pm 10\%$ thickness variation. Only when the layer changed over 15% of thickness, the reflectivity drops sharply to 40% of the original values. So from this simulation, we can conclude that a change of 10% in DBR structure is tolerable in our device.

As for the influences on the device performance, we could use a simple linear model to obtain the first order estimation. Shown in Fig. 9, compare the spectra with and without DBR, the amount of UV reduction ($\Delta U'$) can be correlated to enhancement of visible ($\Delta B'$). This $\Delta U'$ changes linearly with the reflectivity at 380 nm, and when reflectivity becomes zero, no enhancement can be detected. So a linear fit for various colors can be demonstrated in Fig. 9(b)–(f), according to various reflectivities at 380 nm. To set a standard of 90% of the current enhancement, the reflectivity cannot be lower than 85%, which also comply with our previous $\pm 10\%$ DBR fabrication tolerance.

To evaluate the light quality of three CCT WLEDs further, this study analyzed the deviation of the angular CCT. The deviation of the angular CCT can be determined by subtracting the minimum CCT from the maximum CCT in the range between -70° and 70° . Fig. 10 depicts the distribution of the angular CCT of the QD-WLED sample at 2500, 3500, and 4500 K; the deviation of the angular CCT in the WLEDs can be improved considerably

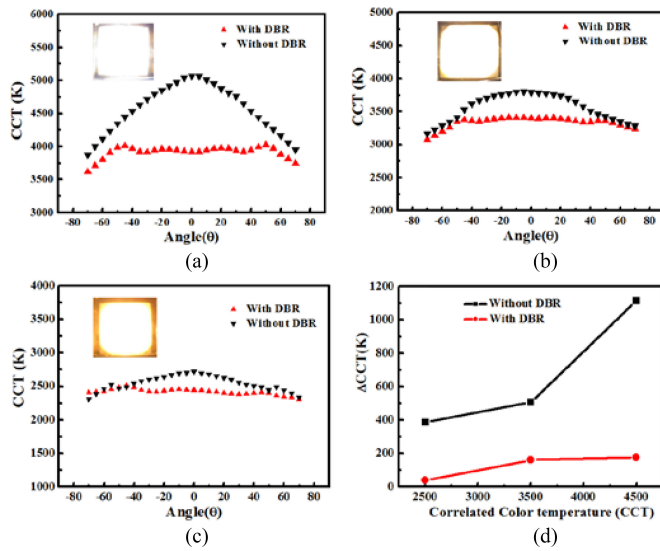


Fig. 10. Angular-dependent CCT with and without DBR of (a) 4500 K, (b) 3500 K, and (c) 2500 K. (d) The CCT deviation for three samples with and without DBR.

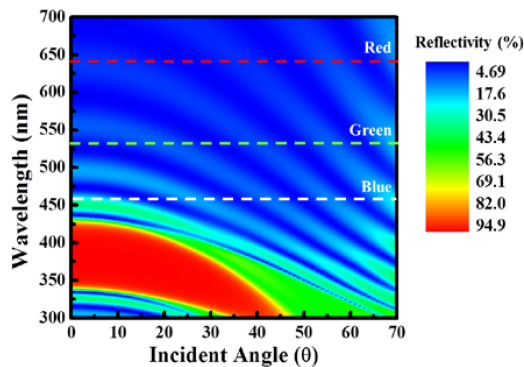


Fig. 11. Color mapping plot of the reflectivity of the DBR at the various incident angles.

by capping the top of the package with a DBR. The CCT deviations of the QD-WLEDs with and without DBR at 4500 and 3500 K improved from 1114 and 621 K to 177 and 160 K.

Specifically, the CCT of the QD-WLEDs with the DBR structure at 2500 K decreased to 39 K, which indicates excellent color uniformity at various angles. With such uniformity, unwanted phenomena, such as yellow rings, can be avoided. In general, the conventional WLED structure has an inferior angular CCT deviation because of the poor extraction of blue light at wide angles. With DBR structure, as highlighted in the following paragraph, the ratio of blue over other colors can be appropriately adjusted such that blue photons are suppressed in the normal direction and the CCT uniformity is improved.

To examine the variation of the DBR reflection and its effect on the CCT uniformity improvement, this study used the TFCalc simulation software to analyze the characteristics of the DBR structure. Fig. 11 shows the reflectivity of the DBR structure at various angles. The reflectivity of the DBR structure in the normal direction demonstrates an almost total reflection of the

UV light, and the reflection of the blue light region is higher than that of the green and red light regions. Therefore, the CCT can be suppressed in the normal direction because of the lower transmittance of blue light. In addition, the reflectivity of the DBR structure at the peak wavelength of the UV chip (380 nm) considerably decreased by 40% when the incident angle exceeded 35°. Moreover, the reflectivity of the blue light region also decreased by the same amount as the green and red light regions when the incident angle exceeded 35°. The simulated results indicate that the short wavelength reflectivity of DBR at an angle greater than 35° can be suppressed, and more blue photons are permitted at wide angles than in the normal direction. This reduction in reflectivity can change the ratio of the blue to yellow photons and thus modify the CCT at wide angles. Furthermore, the extra cost due to the addition of DBR structure can be estimated as only 0.2% of the overall QD-WLED package budget. Therefore, the technology of QD-WLEDs with DBR can provide light sources with high CRIs and uniform CCTs, which can potentially replace traditional lighting in the next generation of SSL.

IV. CONCLUSION

This study presents DBR capped monochromatic QD-WLEDs with various CCTs that can be possible candidates for the future generation of SSL. The DBR capped onto the top of the package can increase the intensity of the QD emission by reflecting the UV light for both monochromatic QD-LEDs and QD-WLEDs. Moreover, the CRI of the WLED is greater than 90. Therefore, the CCT angular deviation of the QD-WLED can decrease to 39 K because of the reflectivity variation of the DBR at various incident angles, which is an attractive approach for high quality lighting applications.

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