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Thermal and Optical Analysis of Quantum-Dot-Converted White LEDs in Harsh Environments

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Abstract: Quantum-dot-converted white LEDs (QD-WLEDs) are promising in both lighting and display applications owing to their high luminous efficiency (LE) and high color-rendering index (CRI). However, their working lifetime is severely limited by the poor reliability of QDs and the LED package. In this work, the variation in photothermal parameters in QD-WLEDs during aging was investigated, and the effect of QDs and the LED package on the optical performance of QD-WLEDs was analyzed. First, the optical properties of phosphor–silicone, QD–silicone, and silicone were measured during the 524 h aging process at 80 °C and 100 °C, respectively. QD-WLEDs with high optical performance were packaged and aged under the same conditions to investigate the variation in their optical parameters and analyze the trends of their CRIs and correlated color temperatures (CCTs). According to the experimental results and the calculation models of the spectra, it was found that the changes in optical parameters are mainly caused by the degradation of QDs, and the aging of QDs has different effects on the CRI and CCT. The analysis of the energy-transfer process shows that the decrease in luminous flux of QD-WLEDs during the aging process is mainly caused by the aging of silicone. Based on optical and thermal analysis, this study proposed different optimization strategies for optical quality and lifetime in the LED design process.

Keywords: quantum dots; white-light-emitting diodes; color-rendering index; packaging reliability; aging analysis

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1. Introduction

White-light-emitting diodes (WLEDs) are the most widely used lighting sources in this century owing to their advantages of being energy saving, safe, and long-lasting [1,2]. Traditional WLEDs consist of a blue LED chip and yellow phosphor composites, which are known as phosphor-converted WLEDs (pc-WLEDs) [3]. The chip converts electrical energy into blue light, and the phosphor converts part of the blue light into yellow light, which is then mixed with unconverted blue light to produce white light [4]. However, the lack of a red component in the spectrum of pc-WLEDs leads to a low color-rendering index (CRI) when it is used for indoor lighting. To solve this problem, some researchers have proposed mixing red fluorescent materials and yellow phosphors to improve the CRI of WLEDs [5–7]. Among them, quantum dots (QDs) have been widely adopted due to their narrow spectrum, feasible tuning range, and high quantum efficiency. Adding red QDs to pc-WLEDs can make up for the lacking red component in the spectrum and achieve a high CRI, which could significantly improve the lighting quality of the light sources [8–10].

However, the development of QD-WLEDs still faces challenges. On the one hand, QDs are sensitive to ambient moisture, oxygen, and heat, which makes them susceptible to degradation, and the underlying mechanisms are quite complicated [11–13]. On the other hand, QD-WLEDs face the same problems as traditional LED devices, such as silicone aging, phosphor carbonization, and chip efficiency degradation [14,15]. Aging of QD-WLEDs can lead to a series of negative effects, such as changes in CCT, degradation of the color-rendering performance, device heating, etc., which eventually results in a shortened working lifetime and reduced lighting performance. The performance degradation of QD-WLEDs is divided into spectral variations and heating, with the former leading to a decrease in lighting quality and the latter leading to a reduction in working lifetime [7,16–19]. Obviously, the performance degradation of QD-WLEDs is related to different parameters. To efficiently optimize the packaging of QD-WLEDs, it is necessary to clarify the specific effects of each factor on their aging performances.

The aging state of QD-WLEDs, as an electro-optical conversion lighting device, is indicated by the changes in optical parameters. According to the previous section, the most important influencing factor affecting the aging of electronic devices is temperature. Therefore, this study analyzed the influence of ambient temperature on the reliability of QD-WLEDs from both optical and thermal perspectives. First, the optical properties of phosphor–silicone, QD–silicone, and silicone were each measured under ambient temperatures of 80 °C and 100 °C for 524 h to explore the degradation characteristics of phosphor, QDs, and silicone. Then, QD-WLEDs were subjected to the same conditions to investigate the variation in the optical parameters and analyze the trends of CRI and CCT. Finally, optical and thermal analysis was combined to reveal the effects of the property changes in QDs, phosphors, and silicone, respectively, on the degradation of QD-WLED optical performances and working lifetimes.

2. Experimental Methods

The key parameters of WLEDs are the CRI, CCT, and electro-optical conversion efficiency. CRI is the average of the luminaire's ability to display the true color of standard samples. CCT is another important indicator of the lighting source, which will affect human physiology and psychology and should be considered during the use of WLEDs. The electro-optical conversion rate refers to the energy utilization efficiency of the device, which indicates the indirect heat generation of the device. The decrease in electro-optical conversion efficiency is due to the aging of the device, and the increase in heat generation will intensify the aging of the device in turn. The relationships between the aging of fluorescent/packaging materials and the optical and thermal properties of QD-WLEDs are shown in Figure 1.

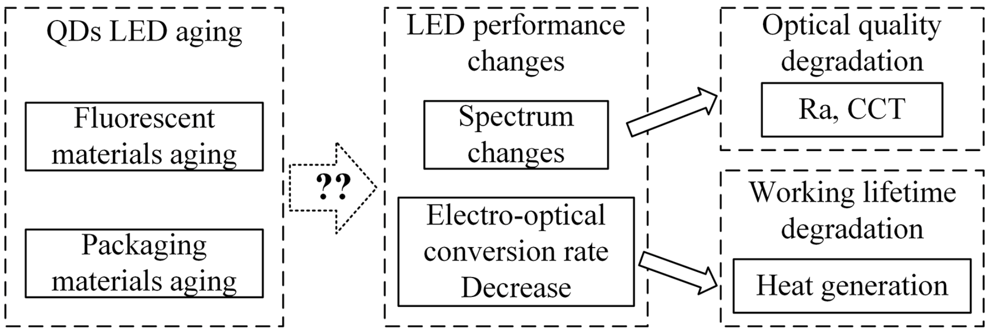

Figure 1. Relationships between the aging of fluorescent/packaging materials and the optical and thermal properties of QD-WLEDs.

As shown in Figure 1, the main parts of LED aging, including luminous-material aging and packaging-material aging, have a complex effect relationship with spectrum changes and electro-optical conversion rate decreases, which is denoted by a question mark. However, there are clear relationships between spectral changes and Ra and CCT parameter variation, and between electro-optical conversion rate decreases and heat generation. In this study, the changes in Ra, CCT parameters, and heat generation were directly analyzed so that the relationship between QD-WLED aging and its performance changes could be deduced. The logic in Figure 1 was the basis for the following experimental design.

As shown in Figure 2a, a 1 Watt LED module with a blue LED chip of 450 nm was used for the experiments. As shown in Figure 2b, four kinds of LED samples were prepared, i.e., LEDs with only silicone encapsulation, LEDs with QD–silicone, LEDs with phosphor–silicone, and WLEDs with QD/phosphor–silicone. Red CdSe/ZnS core/shell QDs (Poly Optoelectronics (Shenzhen, China), 635 nm) and yellow phosphor (Intematix (Fremont, CA, USA) YAG-04, 550 nm) were used. Silicone (DC-184, Dow corning, Auburn Hills, MI, USA) was used to disperse the phosphor and QDs, and then the mixture was covered on the chip and cured at 85 °C for 1 h. Among them, the mass ratio of each component in WLEDs with QDs/phosphor–silicone was set as 0.15 g phosphor: 0.45 mg QDs: 1 g silicone. The CCT was tuned to be 5000~6000 K, and the CRI was 90~94. The optical properties of the as-fabricated WLEDs were tested, and those with relevant CCT, optical power, and other parameters were selected as testing samples to ensure consistency.

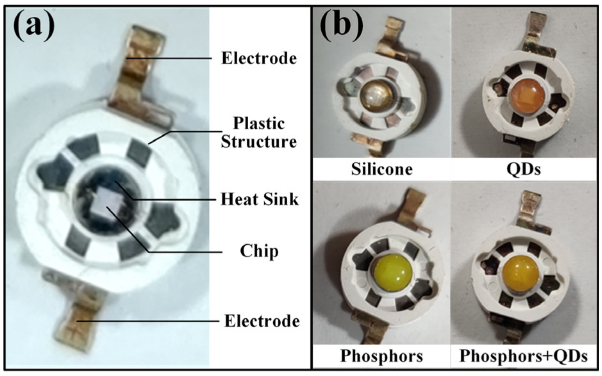

Figure 2. (**a**) Structure of LED sample. (**b**) Photographs of the four kinds of LED samples.

It is well known that the QDs will fail rapidly when the working temperature is above 100 °C, making it difficult to accurately measure the aging process of LEDs. Moreover, the LED aging is not obvious when the temperature is too low, so the experimental conditions of 80 °C and 100 °C were chosen. Figure 3a,b shows the accelerated aging test and optical testing systems, respectively. The four groups of samples and the bare LED samples without silicone were each placed in a heating oven at 80 °C and 100 °C. The samples were taken out periodically, and then cooled to room temperature. Subsequently, their optical performances were measured under a driving current of 350 mA by using an integrating sphere (ATA-1000, Everfine, Hong Kong, China). The total aging time was set as 524 h.

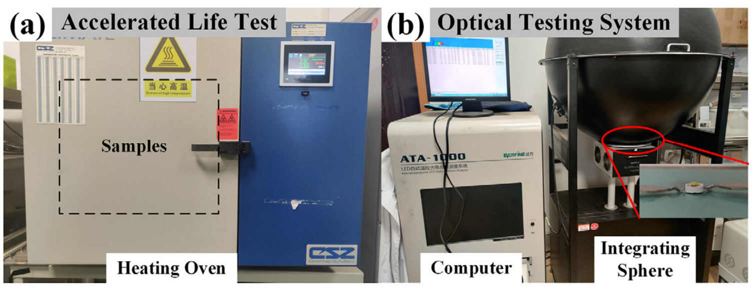

Figure 3. (**a**) Accelerated life test device. (**b**) Optical testing system.

3. Results and Discussion

3.1. Optical Analysis

The experimental results show that after 524 h of aging, the optical power of the basic LED module decreases by less than 1.9%, indicating that the aging of the chip and circuit in the LED module is not obvious and the decrease in optical efficiency is mainly caused by silicone and fluorescent materials. To study the specific effects of each component on the device, the optical parameters of the four samples were analyzed.

Spectral peak changes visually reflected the aging state of luminous materials and chips, in which blue light (peak 450 nm) was the light emitted by the chips, yellow light (peak 550 nm) was converted by the phosphors, and red light (peak 630 nm) was converted by the QDs. Figure 4 shows the spectra and normalized peak intensity of WLEDs at 80 °C (Figure 4a) and 100 °C (Figure 4b). The peak red light of the WLEDs decreased by 33.2% and 42.9%, respectively. It shows that the higher the temperature, the more evident the aging of the quantum dots. Meanwhile, the yellow light peak decreased by 8.7% and 7%, respectively. The yellow light decreased less at higher temperatures because there is a competitive effect between the photoconversion of QDs and phosphors, and the effect is greater than the aging of the phosphors themselves.

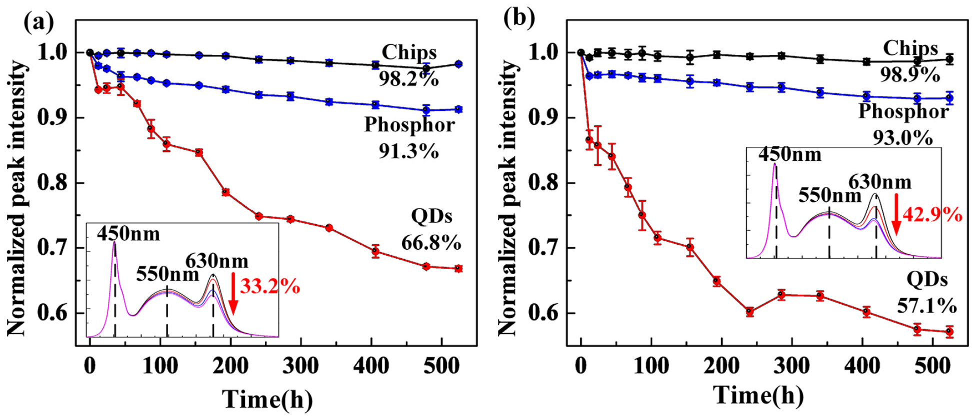

Figure 4. Spectral peaks of WLEDs during aging at (**a**) 80 °C and (**b**) 100 °C. Insets are the spectra of the WLEDs.

To further explore the effect of aging of the fluorescent material on the optical performance of the system, the CCT and CRI trends of the WLED samples during the experiment were analyzed. CCT and CRI parameters are determined by the spectrum, which shows the optical performance of the WLED device in use and is the main optimization target in WLED design. As shown in Figure 5a, the CCT of the two sets of WLED devices with different temperature conditions increased by 1105 K and 1721 K after 524 h of the experiment, respectively. Figure 5b gives the CRI trend of the WLED devices during the experiment. Both groups of samples underwent a process of fluctuation and then stabilization, with a greater range of CRI variation at higher temperatures and a more pronounced decrease in the CRI at final stabilization. The above results indicate that the aging of the QD materials could cause an increase in the CCT and a decrease in the CRI.

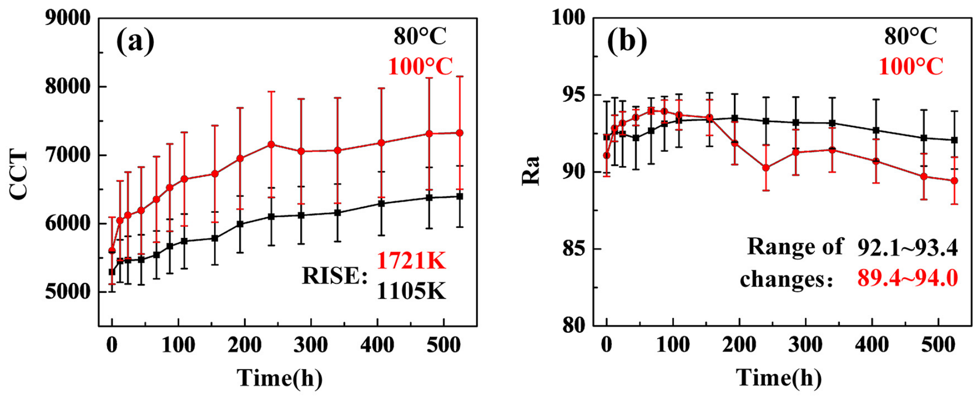

Figure 5. (**a**) CCT and (**b**) CRI changes in white LEDs during aging.

The experimental results show the optical performance trend of LEDs during aging, the influence of which comes from the combined effect of several parameters. To explore the effect of QD degradation on LED devices and the specific mechanism of this effect, numerical calculations are required.

The first step is establishing a spectral model of the WLEDs. During the experiment, the attenuation of the chip and phosphors can be neglected. The main attenuation appeared at 650 nm, which corresponds to the failure of the QDs. The luminescence of the chip and phosphors was considered, and the light emitted from the QDs was superimposed to establish a spectral model with time. A typical sample consisting of chip and phosphors was selected as the standard for the first set of spectral parameters. The spectra of the QDs are also measured with experiments.

The second step is setting an attenuation coefficient k, which represents the ratio of the light conversion efficiency of the QDs to the initial value. The overall spectrum is expressed as

$$S(\lambda) = S_{\text{chip+phosphor}}(\lambda) + (1-k) \times S_{\text{QDs}}(\lambda) \quad (1)$$

where the range of the attenuation coefficient k is set from 0.5 to 1 based on the variation range of the QD light conversion efficiency in practical applications at 100 °C.

The third step is calculating the CCT and CRI corresponding to the spectrum in accordance with the method developed by CIE. First, the CCT of the light source to be measured was calculated. Then, the color-rendering ability of the light source for the eight standard color samples was calculated further based on the CCT calculation results, the average value of which is the color-rendering index Ra. The calculation results are shown in Figure 6.

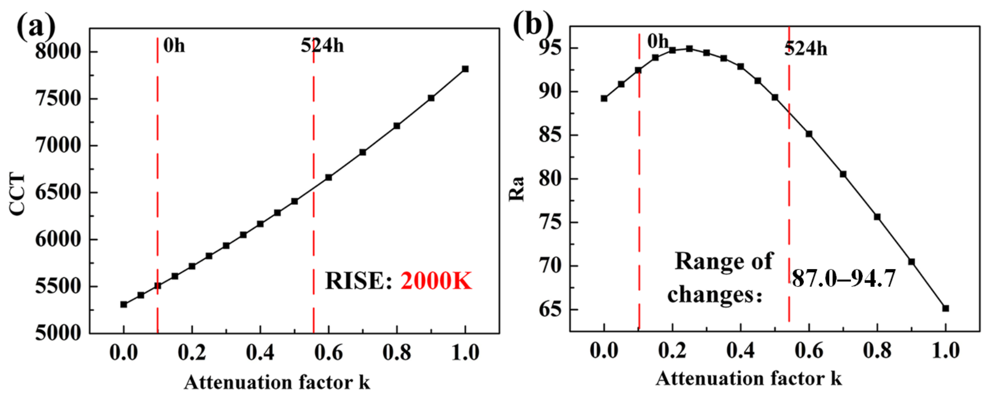

Figure 6. (**a**) CCT and (**b**) CRI of simulated spectrum changes with attenuation factor k.

Figure 6a,b shows that the CCT increases uniformly, whereas Ra first increases and then decreases as aging progresses. The trends are basically consistent with the experimental results. Similar trends indicate the results of the study, which simulated the aging of QDs through changes in spectral components, are accurate. And further comparison of the variation intervals of experimental and simulated values enables to study the effect of QDs aging in the variation in LED optical parameters. The red dashed line corresponds to the interval of attenuation factor k in the experiment, and the rise in CCT is 2000 K and the variation in Ra is 87.8 to 94.7, which is consistent with the experimental results (CCT is 1721 K and the variation in Ra is 89.4 to 94.0).

The determinants of the spectral structure include light generation by the chip, light conversion by phosphors and QDs, and light absorption by the packaging structure. In the simulation, the changes in the chip-generated light, phosphor-converted light, and packaging structure-absorbed light during the aging process were ignored, while only the QD-converted light was considered. Therefore, the comparison of the simulation results with the experimental results shows the proportion of the QD-converted light and the ignored parts in the changes in spectral parameters. The smaller the difference between simulation and experiment, the greater the effect of QD aging on the changes in optical parameters of the device. In this study, the simulation results demonstrated that the changes in the optical parameters of the LED devices are mainly caused by the partial attenuation of the red light of the spectrum due to the aging of QDs.

3.2. Thermal Analysis

The main factor affecting the lifespans of electric equipment is heat generation. The heat generation of QD-WLEDs is compensated for through optical absorption and efficiency loss, while the efficiency loss included an electro-optical conversion efficiency part and an optical conversion efficiency part. In Section 3.1, the optical effects of aging of QDs, which leads to a decrease in light conversion efficiency, have been demonstrated. However, the effects on QD-WLED heat generation need to be further investigated. To determine the percentage of different parts in the QD-WLED heating generation, the energy transfer process was analyzed.

The energy transfer process of LEDs is shown in Figure 7. E_{in} is the input electric power of the whole system, part of which is converted into blue light by the chip, and the rest is converted into thermal energy E_1, which includes the conversion heat loss of the chip and the blue light absorbed by the chip itself. Part of the blue light is absorbed by the phosphor and QDs and converted into yellow and red lights, in which a portion of the light energy is converted into thermal energy E_2. The three wavelengths of light are mixed to form white light, and the device structure (mainly silica gel in these samples) absorbs a portion of the energy E_3, with the remaining energy being the final output white light E_{optical}.

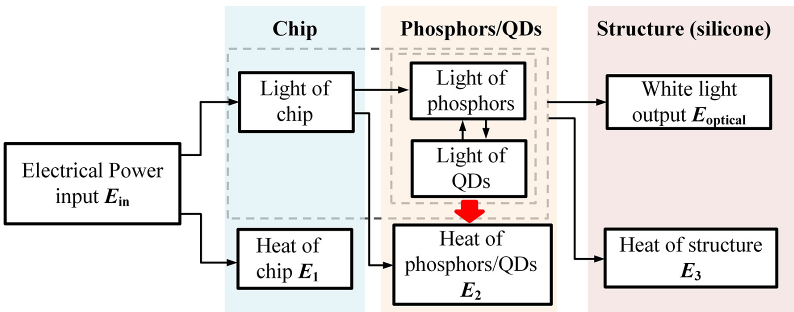

Figure 7. The energy transfer process of QD white LEDs during operation.

The structures and the energy transfer of the three samples are shown in the Figure 8. The different effects of chips, silicone, QDs and phosphors energy transfer process were analyzed through comparison. According to the conservation of energy, we can obtain the following relationship.

For the LED chip, as shown in Figure 8a, its optical power can be calculated by

$$E_{\text{opitcal1}} = N_1 \times E_{\text{in}} = E_{\text{in}} - E_1 \tag{2}$$

For the LED chips with pure silicone, as shown in Figure 8b, its optical power can be calculated by

$$E_{\text{opitcal2}} = N_2 \times E_{\text{in}} = E_{\text{in}} - (E_1 + E_3) + K \tag{3}$$

where K is the light output energy enhanced by silicone.

For the LED chips with silicone and luminous materials, as shown in Figure 8c, its optical power can be calculated by

$$E_{\text{opitcal3}} = N_3 \times E_{\text{in}} = E_{\text{in}} - (E_1 + E_2 + E_3) + K \tag{4}$$

where N_1, N_2 and N_3 denote the electro-optical conversion efficiency of the three samples, respectively. Considering E_1 + E_3 − K as device structure heat generation, the following relationships are obtained:

$$\frac{E_1 + E_3 - K}{E_{in}} = 1 - N_2 \tag{5}$$

$$\frac{E_2}{E_{in}} = N_2 - N_3 \tag{6}$$

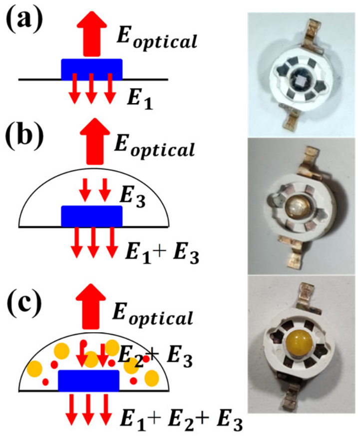

Figure 8. The energy transfer of (**a**) an LED chip, (**b**) an LED chip with silicone, and (**c**) an LED chip with silicone and luminous materials.

The heat generation E_{thermal} of the whole device can be calculated by

$$E_{\text{thermal}} = 1 - \frac{E_{\text{optical}}}{E_{in}} \tag{7}$$

During the aging process of QD-WLEDs, each component causes heat generation due to their own properties. Besides, heir aging state also leads to a change in thermal properties, which affects the QD-WLED heat generation. Typically, the aging of each component leads to an increase in heat generation, which accelerates the aging of QD-WLEDs. Therefore, it is also necessary to compare the parameter changes before and after the aging of the device. The process of thermal parameter change indicates the change in heat generation of LED device in the aging process. At last, by analyzing the thermal parameters, the effect of different parts on the heat generation of QD-WLEDs during the aging process could be investigated.

After aging at 80 °C for 524 h, the electro-optical conversion efficiency of each sample is shown in Table 1. After aging, the electro-optical conversion efficiency of chip (N_1) only decreased by 1.41% and that of silicone (N_2) could reach as high as 4.9%, respectively. When it comes to phosphor–silicone, QD–silicone and phosphor/QD–silicone (N_3), the value is 6.07%, 6.1% and 4.43%, respectively. When silicone/luminous materials are present in the structure, the increase in heat generation of the sample during the experiment is significantly higher than in the structure without silicone/luminous materials. After excluding the effect of heat generation from the chip, we analyzed the comparisons of the silicone and luminous materials on their effect to the increase in heat generation. The results of the analysis are presented in Table 2.

Table 1. The electro-optical conversion efficiency of each sample after aging at 80 °C for 524 h.

Electro-Optical Conversion Efficiency	Before Aging	After Aging
N_1 (chip)	50.34%	48.93%
N_2 (Silicone)	64.06%	59.16%

Table 1. *Cont.*

Electro-Optical Conversion Efficiency	Before Aging	After Aging
N_3 (Phosphor–silicone)	41.04%	34.97%
N_3 (QD–silicone)	30.59%	24.49%
N_3 (Phosphor/QD–silicone)	33.69%	29.26%

Table 2. The change ratio of device structure heat generation ($(E_1 + E_3 - K)/E_{\text{in}}$), heat generation of the whole device ($E_{\text{thermal}}/E_{\text{in}}$) and luminous material heat generation (E_2/E_{in}) before and after aging were calculated for different samples, respectively.

	Before Aging	After Aging	Change Ratio
E_1/E_{in}	49.66%	51.07%	2.84%
$(E_1 + E_3 - K)/E_{\text{in}}$	35.94%	40.84%	13.65%
Phosphor–silicone			
$E_{\text{thermal}}/E_{\text{in}}$	58.96%	65.03%	10.30%
E_2/E_{in}	23.03%	24.19%	5.03%
QD–silicone			
$E_{\text{thermal}}/E_{\text{in}}$	69.41%	75.51%	8.79%
E_2/E_{in}	33.48%	34.67%	3.56%
Phosphor/QD–silicone			
$E_{\text{thermal}}/E_{\text{in}}$	66.31%	70.74%	6.68%
E_2/E_{in}	30.37%	29.90%	−1.55%

Table 2 gives the change ratio of device structure heat generation ($(E_1 + E_3 - K)/E_{\text{in}}$), heat generation of the whole device ($E_{\text{thermal}}/E_{\text{in}}$), and luminous material heat generation (E_2/E_{in}) before and after aging were calculated for different samples. The change ratio of the chip heat generation was 2.84% while that of the device structure heat generation was 13.65%. This result is consistent with the conclusion in the previous paragraph that the increase in heat generation of chip is not significant. For the phosphor–silicone, after the aging, the change ratio of the whole device heat generation was 10.30%, while the change ratio of the luminous material heat generation was only 5.03%. For QD–silicone, the change ratio of the whole device heat generation was 8.79%, while the change ratio of the luminous material heat generation was only 3.56%. For phosphor/QD–silicone, the change ratio of the whole device heat generation was 6.68%, while the change ratio of the luminous material heat generation was −1.55%. Three samples containing both silicone and luminous material had similar experimental results, showing that the increase in heat generation due to the aging of silicone is greater than that due to the aging of luminous material for all three samples. Comparing the different luminous materials in the three kind of samples, although the heat generation increase in both is less than that of silicone, the experimental results show that the heat increase in phosphor is higher than that of QDs. This is due to the quantum efficiency of phosphor being lower than that of QDs, so more phosphor needs to be added when preparing the WLED samples, which causes a higher total heat generation than that of QDs. A higher total amount of heat generation means a higher amount of heat generation increase when thermal performance changes.

According to the above results, it can be concluded that the heat generation increase during aging of the WLEDs is mainly due to the failure of the silicone, while the secondary influencing factor is the failure of the phosphor, and the smallest influencing factor is the failure of the QDs. Effective thermal management strategies toward phosphor–silicone layer are essential for the optical performances maintenance of WLEDs [20,21].

4. Conclusions

During the aging process of QD-WLEDs, the optical properties changed significantly. Optical parameters such as CRI and CCT could severely damage the lighting performance of QD-WLEDs. In this work, it was found that the failure of the QDs in the device is the main reason for the degradation of the optical performances of the QD-WLEDs. According to the calculation models, as the QDs ages, the WLEDs' CRI fluctuates, and high temperatures can exacerbate this phenomenon. The effect of QDs aging on CCT is greater than the effect on color-rendering index.

Moreover, from the experiments, it could be concluded that the decrease in luminous flux of the device is mainly due to the aging of the silicone. The heat generation of the QD-WLED increases during the aging process, mainly due to the aging of silicone, while the phosphor aging is a secondary influence factor. Moreover, with the aging of silicone, the device heat generation increases, making the device aging faster in turn.

In WLED manufacturing and applications, the following improvements can be made based on the experimental results.

1. In the WLED packaging process, in addition to the impact of luminous-flux decline during the work process, the changes in lighting quality should also be considered.
2. In the case of noncritical working environments, the WLEDs could be lit first as the initial parameters until the CRI is stable.
3. In long-term use, QD-WLED devices with a color temperature slightly lower than the target could be chosen so that the color temperature of the LED could meet the requirements for a longer time.
4. In addition to the calculation results of the CRI, the specific spectrum and CRI trends during long-term use should also be considered.
5. The device's heating problem can be improved by choosing a silicone with better temperature resistance or by using replaceable separated silicone film to extend the service life.

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