Corrigendum

P-12.10: Aging Behavior of Quantum Dot Light-Emitting Diodes Employing Sputtered TiO2 as the Electron Transport Layer

In [1], changes have been made to the following:

• The full version of the article has been added to this version after publication.

Reference

[1] Pan X., Wei J., Li D., Xu Z., Ma J. and Sun X.W. P-12.10: Aging Behavior of Quantum Dot Light-Emitting Diodes Employing Sputtered TiO₂ as the Electron Transport Layer. SID Symposium Digest of Technical Papers. 2024; 55(S1): 1367-1367. https://doi.org/10.1002/sdtp.17369

Aging Behavior of Quantum Dot Light-Emitting Diodes Employing Sputtered TiO₂ as the Electron Transport Layer

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Abstract

This study presents an analysis of quantum dot light-emitting diodes (QLEDs) utilizing sputtered titanium dioxide (TiO₂) films as the electron transport layer (ETL), aiming to enhance device performance controllability and predictability. Our research encompasses the design, fabrication, and comprehensive assessment of QLEDs, focusing on their performance, stability, and aging behavior. By integrating TiO₂ via sputtering, we observed a predictable aging curve.

Author Keywords

Quantum dot light-emitting diodes; Electron transport layer; TiO_2 ; Aging

1. Introduction

In recent years, there has been significant advancement in enhancing the efficiency and device lifetime of quantum dot lightemitting diodes (QLEDs).^[1] The intrinsic ability of QLEDs to precisely manipulate the size and composition of luminescent materials has led to accurate spectral tuning and enhanced color rendition, setting them apart from conventional display technologies. However, despite these technological advances, the widespread commercial adoption and reliability of QLEDs still encounter formidable challenges, especially concerning device lifespan and operational stability.^[2]

A pressing issue in conventional QLED design is the use of ZnO nanocrystals as the electron transport layer (ETL), a choice that, while effective, places limitations on the controllability of the device's performance and the predictability of its lifespan.^[3,4] Due to their chemically active nature, ZnO nanocrystals are susceptible to environmental and electrical field, which can lead to interface deteriorate with quantum dots, electrode. This can adversely affect the performance controllability and predictability and the aging behavior in the lifespan of the QLEDs. Trying to avoid the adverse effects of ZnO, lots of literatures corroborates the potential of alternative ETL materials in remedying the challenges faced by conventional QLEDs.^[6] Among these materials, TiO₂ stands out due to its successful incorporation into solar cell technology and its commendable stability characteristics. A wealth of studies have highlighted the utility of TiO₂ in various optoelectronic devices, accentuating its enduring stability and effectiveness.

This study endeavors to meticulously investigate the aging behavior of QLEDs that incorporate sputtered TiO_2 films as the ETL. Concentrating on pivotal aspects such as device

performance stability, and aging behavior, the goal is to devise a predictable aging curve in the lifespan of QLEDs.

2. Experimental Section

In this study, an inverted bottom-emitting QLED structure is employed, specifically designed to prevent plasma-induced damage to the quantum dot emission layer during the sputtering process. The structure of the device, layered from top to bottom, includes Al/HAT-CN/NPB/TcTa/QDs/TiO₂/ITO/Glass. In the structure, the sputtered TiO₂ film, functions as the ETL. The emission layer is CdSe-based quantum dots. Other layers of materials and fabrication details have been reported in our previous report.

3. **Results and Discussion**



Figure 1. Structure and typical performance of QLED with sputtered TiO_2 film as ETL. (a) Device structure. (b) Voltage-dependent EL spectra. (c) J-V-L, and (d) EQE-L-CE characteristics.

Figure 1(a) details the structure of the device, whilst Figure 1(b) presents the typical voltage-dependent electroluminescent (EL) spectra. The device maintains a stable emission spectrum during the voltage scan, with minimal shifting, akin to a device based on ZnO nanocrystal ETLs. Subsequent device engineering results in the typical characteristics (J-V-L, EQE-L) shown in Figures 1(c) and (d). The optimized maximum external quantum efficiency (EQE) is registered at 7.2%, with peak luminance around 51,200

 cd/m^2 . The device achieves its turn-on voltage (V_T) at 2.3 V (the voltage required for 1 cd/m^2), however, photon detection is feasible under 1.9 V. Such a turn-on voltage is aligned with a standard ZnO-based inverted QLED device, which typically struggles to attain sub-threshold turn-on behavior. This may suggest that the higher turn-on voltage is primarily a result of the limited hole transport and injection process in inverted QLEDs, as further confirmed by our recent investigation demonstrating a noticeable reduction in turn-on voltage when small molecule hole transport materials are replaced with conventional polymers in normal structures, such as TFB.



Figure 2. (a) Aging results (L-t) of devices under different current stress. (b) Lifetime acceleration factor n fitted by Equation 1.

In addition to device performance, operational stability significantly influences QLED application. Figure 2(a) presents the device's aging results under constant current mode. The device luminance shows a pure exponential-like decay with aging time. Results indicate minimal abnormal luminance increase at the beginning, particularly when compared to devices based on ZnO nanocrystal ETLs, which typically exhibit substantial luminance increases during the initial aging stage.

This abnormal increase in luminance significantly impacts the predicted lifetime of QLEDs based on ZnO nanocrystals, particularly at low initial luminance levels. Typically, the correlation between the initial luminance and the point of degradation is delineated by the following equation:

$$L_0^{\ n}T_{50} = C$$
 (1)

In Equation 1, L_0 denotes the initial luminance at the onset of the aging stage, while T_{50} represents the time the device takes to degrade to half of L_0 . The acceleration factor, n, is often empirically determined through aging at different and is linked to the materials, device structure, and even the process conditions. Any change in conditions demands a retest of n. For traditional QLEDs with ZnO-based nanocrystals as ETLs, n is commonly approximated to be ~1.8. It should be noted that multiple lifetime reports of QLEDs have overlooked the influence of initial abnormal luminance increases on device lifetime predictions. This oversight can lead to inaccurate estimations of device lifetimes, typically overestimating the actual lifetime of the device.

However, in the case of QLEDs with a sputtered TiO_2 film serving as the ETL, the aging curves display a predominantly exponential decay, with minimal abnormal luminance increase during the initial aging stage. The fitted results presented in Figure 2(b) show that the acceleration factor, n, for a QLED with a sputtered TiO₂ film as ETL is approximately 0.84. This figure is considerably lower than that observed with QLEDs based on ZnO nanocrystals (~1.8). A smaller n signifies expedited degradation of the QLED with a sputtered TiO₂ film as ETL. This could be attributed to enhanced quenching at the QD/TiO₂ interface compared to the QD/ZnO-based nanocrystal interface. The energy of quenched excitons contributes to the degradation of the organic hole transport layer or QDs. Table 1 summarizes the typical device parameters of QLED with sputtered TiO₂ film as ETL.

Table 1. Typical device parameters of QLED with sputtered TiO₂ film as ETL.

Device	V _T (V)	EQE (%)	L _{Max} (cd/m ²)	$T_{50} @ 100 cd/m$ (h)	n*
Sputter ed TiO ₂ Film a s ETL	2.3	7.2	51,200	205.5	0.84

*Life time acceleration factor

4. Conclusions

Sputtered TiO₂ has been deployed as an ETL within QLEDs. Although these QLEDs exhibit lower efficiency and a reduced lifetime compared to conventional QLEDs utilizing ZnO-based nanocrystals, the use of sputtered TiO₂ films as ETL in inverted QLEDs yields a predominantly exponential luminance decay aging curve with little abnormal luminance increase at initial aging stage.

Acknowledgments

This work was supported by the National Key Research and Development Program of China (No. 2022YFB3602903, 2021YFB3602703, and 2022YFB3606504), National Natural Science Foundation of China (No. 62122034), Guangdong University Key Laboratory for Advanced Quantum Dot Displays and Lighting (No. 2017KSYS007), Shenzhen Key Laboratory for Advanced Quantum Dot Displays and Lighting (No. ZDSYS201707281632549), Shenzhen Science and Technology Program (No. JCYJ20220818100411025), and Shenzhen Development and Reform Commission Project (Grant No. XMHT20220114005).

References

- 1. Jang E, Jang H. Quantum dot light-emitting diodes[J]. Chemical Reviews, 2023, 123(8): 4663-4692.
- Qu X, Ma J, Liu P, et al. On the voltage behavior of quantum dot light-emitting diode[J]. Nano Research, 2023, 16(4).
- Zhang W, Chen X, Ma Y, et al. Positive aging effect of ZnO nanoparticles induced by surface stabilization[J]. The Journal of Physical Chemistry Letters, 2020, 11(15): 5863-5870.
- He S, Tang X, Deng Y, et al. Anomalous efficiency elevation of quantum-dot light-emitting diodes induced by operational degradation[J]. Nature Communications, 2023, 14(1): 7785.

- Choi J, Song S, Hörantner M T, et al. Well-defined nanostructured, single-crystalline TiO₂ electron transport layer for efficient planar perovskite solar cells[J]. ACS nano, 2016, 10(6): 6029-6036.
- Zaiats G, Ikeda S, Kamat P V. Optimization of the electron transport layer in quantum dot light-emitting devices[J]. NPG Asia Materials, 2020, 12(1): 57.
- Hamad S, Catlow C R A, Woodley S M, et al. Structure and stability of small TiO₂ nanoparticles[J]. The Journal of Physical Chemistry B, 2005, 109(33): 15741-15748.
- Lettieri S, Pavone M, Fioravanti A, et al. Charge carrier processes and optical properties in TiO2 and TiO2-based heterojunction photocatalysts: A review[J]. Materials, 2021, 14(7): 1645.

- 9. Kim J, Hahm D, Bae W K, et al. Transient dynamics of charges and excitons in quantum dot light-emitting diodes[J]. Small, 2022, 18(29): 2202290.
- Li Y, Fan X, Shen C, et al. Charge balance in red QLEDs for high efficiency and stability via ionic liquid doping[J]. Advanced Functional Materials, 2022, 32(32): 2203641.
- Davidson-Hall T, Aziz H. Perspective: Toward highly stable electroluminescent quantum dot light-emitting devices in the visible range[J]. Applied Physics Letters, 2020, 116(1).