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Interface Engineering of Quantum Dot Light-Emitting Diodes Based on MoO_3 Interlayer

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Abstract. Quantum dot light-emitting diodes (QLEDs) are strong contenders in display technology due to their excellent color purity and wide color gamut. Nevertheless, the strong acidity and hygroscopicity of the conventional hole injection layer (HIL) PEDOT:PSS lead to ITO anode corrosion and unbalanced carrier injection, severely limiting device efficiency and stability. In this work, MoO_3 prepared by sol–gel method was employed as an interfacial interlayer between ITO and PEDOT:PSS to construct a hybrid dual-hole injection system, and the effects of annealing temperature (100–170°C) on film morphology and device performance were systematically studied. Results reveal that the MoO_3 interlayer effectively blocks ITO corrosion by PEDOT:PSS and optimizes hole injection via energy level matching. The optimized red QLED with MoO_3 annealed at 130°C exhibits outstanding performance: the maximum current efficiency (CE_{max}) reaches 121.21 cd/A, the maximum external quantum efficiency (EQE_{max}) is 17.06%, and the maximum brightness climbs up to 97340 cd/m². More importantly, the T50 lifetime at 100 cd/m² is extended to 8511 h, which is nearly 7 times that of the standard PEDOT:PSS–based device. This study offers a reliable interface engineering strategy for fabricating high–efficiency, long–lifetime all–solution–processed QLEDs, laying a foundation for their industrial applications in advanced displays and lighting.

Keywords: Hole Injection Layer, MoO_3, Interface Engineering, Quantum Dot Light-Emitting Diodes

1. Introduction

Quantum dot light-emitting diodes have the advantages of excellent monochromaticity, high color purity, wide color gamut and long service life, so they have developed rapidly.Therefore, QLED devices are expected to become the mainstream display and lighting technology, applied in smart terminals, ultra-high-definition displays, high-end lighting and other fields. After years of continuous development, the luminous brightness and luminous efficiency of QLEDs have been greatly improved compared with the early stage, and they are expected to become high-performance, large-area, low-cost electroluminescent devices [1-5].

Constructing QLED devices not only needs to consider the carrier injection efficiency but also achieve high carrier mobility. Therefore, selecting a suitable hole injection layer material to achieve better carrier injection balance is particularly important for obtaining high-efficiency and high-

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performance light-emitting devices. At present, PEDOT:PSS is one of the most popular materials as a buffer layer. However, PEDOT:PSS itself is relatively sensitive to water and oxygen in the air and is prone to degradation, thereby corroding the ITO substrate, resulting in poor device stability, and its low conductivity limits its application in all-solution QLED devices. Therefore, people choose transition metal oxide MoO_x as an interlayer to isolate the corrosion of ITO electrodes by PEDOT to overcome the shortcomings of organic materials [6-10].

Previously, many researchers worldwide have adopted double hole-injection layers composed of transition metal oxides/PEDOT:PSS or MoO_x to construct QLEDs, which has improved the device's efficiency, brightness, and operational lifetime to some degree. In 2018, Yang Xuyong et al. [11] designed an all-inorganic structure of ITO/MoO_3/NiO/QD/ZnO/Al with MoO_3 as the hole injection layer. Compared with the previous use of only NiO, the introduction of MoO_3optimized the energy band structure of this device. In 2019, Zhu et al. [12] doped MoO_x into PEDOT:PSS to construct high-performance red QLEDs. This strategy elevated the Fermi level of the hole injection layer, achieving better energy level matching, enhanced hole injection capability, and thereby improved device performance. In 2022, Yu Hsin-Chieh et al. [13] reported QLEDs with various transition metal oxides as the hole injection layer and compared their performance with that of QLEDs based on PEDOT:PSS. When MoO_x was used as the hole injection layer, the maximum brightness of the device reached 61,699 cd/m². In 2023, Yang Ji-Hun et al. [14] investigated the effect of using MoO_xnanoparticles as the hole injection layer on the performance of QLEDs. The experimental results showed that the maximum brightness (Lmax) was 68,240.7 cd/m², the EQEmax was 13.2%, and the CEmax was 56.0 cd/A. In this work, a high-quality MoO_x film annealed at 130°C was used as an intermediate layer to isolate the anode from the PEDOT:PSS layer, with the aim of achieving highly efficient and durable red quantum dot light-emitting diodes.

2. Luminous principle

The device architectures of QLEDs include multi-layered, three-layered, double-layered and single-layered structures, among which the triple-layer structure is the most widely adopted. Fig. 1 presents a typical working mechanism diagram illustrating the charge injection and recombination process. Within the quantum dot emission layer, holes and electrons meet and interact to form excitons, which subsequently generate photons through radiative recombination to realize electroluminescence.

Device fabrication includes the cleaning of indium tin oxide (ITO) substrates, spin-coating of various functional layers, as well as the evaporation and encapsulation of aluminum electrodes.In the experiment, four annealing temperatures (Starting from 100 degrees Celsius, set a stable temperature every 20 degrees, and the final temperature will be 170 degrees Celsius.) were set to select high-quality molybdenum oxide (MoO_x) thin films for subsequent device fabrication.The current density, brightness, current efficiency, and other device parameters were measured using a Keithley 2400 digital source meter.The MoO_x solution was prepared via the sol-gel method using ammonium molybdate as the raw material and deionized water as the solvent.A commercially available aqueous ammonium molybdate solution was diluted with deionized water to a specific concentration, then heated and stirred in air for approximately one hour to obtain a MoO_x solution with a concentration of 6% (w/t).The solution was then filtered through a 0.45-micron filter head for subsequent spin-coating. Each functional layer was spin-coated in a glove box, and aluminum electrodes were evaporated in a Mini Spectra compact thermal evaporation system.

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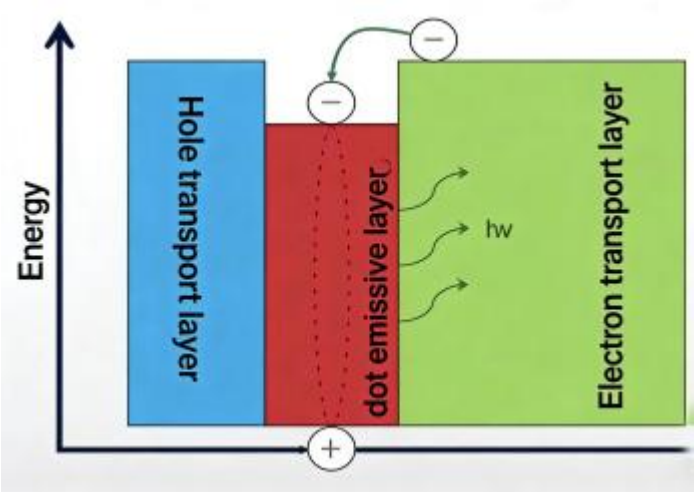

Figure 1. Luminescence mechanism diagram of a typical QLED

3. Results and discussion

Compared with the traditional hole-injection layer material PEDOT:PSS, which exhibits strong acidity (pH = 1.2–2.2) and hygroscopicity, transition metal oxides possess many advantages, such as suitable work function, high transmittance, solution processability, abundant sources, large electron affinity, and low cost.

Among them, MoO_x, as an n-type semiconductor, has the advantages of non-toxicity, high work function, good environmental stability, and deep electronic states. Its chemical composition affects its energy band structure; therefore, MoO_x films with energy levels suitable for QLED devices can be obtained via annealing treatment.

Therefore, this study investigated the changes in surface roughness of MoO_x films prepared by the sol-gel method (under different annealing temperatures). During the experiment, the films were placed in the middle layer to separate the ITO electrode and PEDOT:PSS. The MoO_x interlayer can prevent the corrosion of the ITO electrode by PEDOT:PSS, enhance the hole injection capability of QLED devices, and achieve simultaneous improvement in device efficiency and lifetime.

As illustrated in Figure 2, the luminescence image of the fabricated red QLED device is displayed.



Figure 2. Luminescence diagram of red QLED device

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Due to the existence of oxygen vacancies, MoO_x thin films are n-type semiconductor materials, and the role of oxygen vacancies in the thin films is similar to electron doping. According to existing literature reports, there are performance differences between stoichiometric MoO_x and oxygen-deficient MoO_x in device applications. Therefore, on the basis of the optimal thickness of MoO_x, it is speculated that the annealing temperature will affect the composition and transmittance of the thin film.

Therefore, in this experiment, ammonium molybdate was used as the precursor and deionized water as the solvent, and the film was formed by a simple spin-coating method. Then the prepared MoO_x thin films were treated at different annealing temperatures (100℃, 130℃, 150℃, 170℃). It was found that the surface roughness of the thin films was all within 2.0 nm. The atomic force microscope (AFM) test is shown in Figure 3. The MoO_x thin film was very flat after annealing at 130℃, with the lowest roughness.

Using this film as an interlayer to separate the ITO and PEDOT:PSS layers, the device structure is shown in Figure 4. This structure can effectively reduce the energy barrier for holes to inject from ITO into the organic layer, and at the same time inhibit the corrosion of ITO by the acidic PEDOT:PSS.

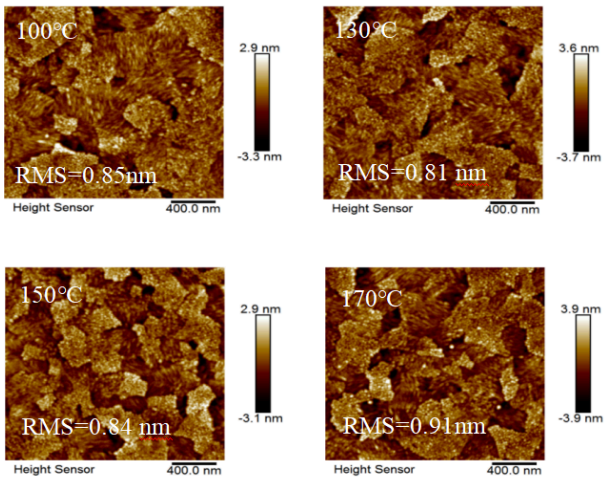

Figure 3. AFM topography of MoO_3 thin films at different annealing temperatures

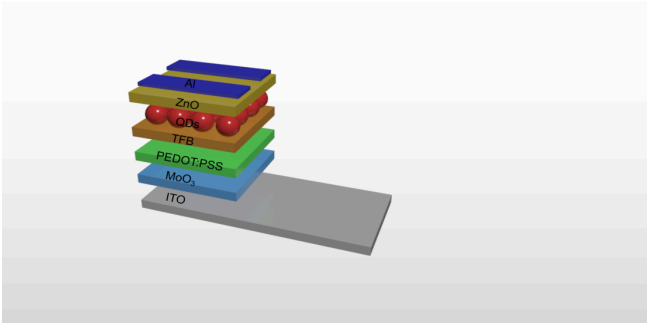

Figure 4. Structure diagram of QLED device with MoO_x/PEDOT:PSS hole injection layer

The red device was tested, and as shown in Figures 5 and 6, the CE_{max} reached 121.21 cd/A, the maximum EQE reached 17.06%, and the maximum brightness reached 97340 cd/m². The significant improvement in performance is due to the effective regulation of charge injection and interfacial

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exciton behavior by MoO_3. The introduction of MoO_3 with high work function forms a reasonable energy level transition between the ITO anode and PEDOT:PSS, which significantly enhances the hole injection effect and helps solve the problem of unbalanced carrier injection in QLED devices.

The balance of carrier injection directly affects the brightness of the device, which explains why this device has high brightness. The previous annealing treatment improved the interface quality, effectively reducing useless energy loss paths, increasing the proportion of effective light emission and improving efficiency. At the same time, the flatter and cleaner interface reduces light loss caused by electrode diffusion or interface defects.

In addition, the introduced inorganic modification layer can restrict excitons, allowing them to be more efficiently confined in the light-emitting area for effective light emission. In short, the cooperative design of this inorganic/organic hybrid hole injection layer provides a new idea for realizing high-efficiency and high-brightness QLED devices.

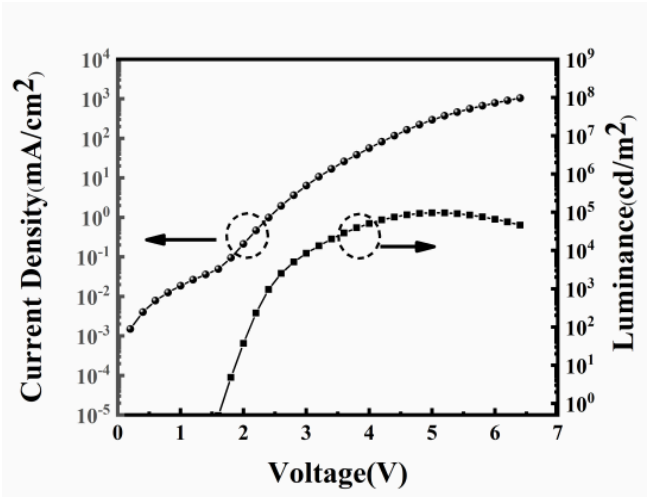

Figure 5. Current density-voltage-brightness relationship diagram

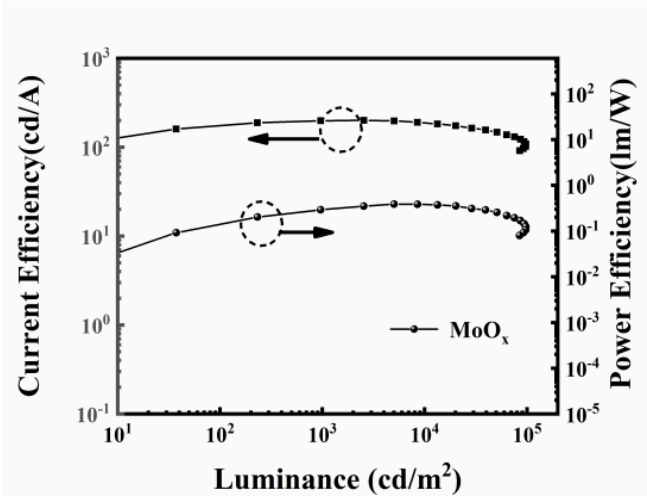

Figure 6. Current efficiency-brightness-power efficiency relationship diagram

Next, to further compare the stability of QLED devices employing MoO_x/PEDOT:PSS, the service life of the standard device was tested in addition to the QLED device with a single-layer MoO_x hole injection layer.

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In Figure 7(a), when driven by a direct current corresponding to an initial luminance of 11610 cd/m², the T_{50} lifetime of the standard QLED device using PEDOT:PSS as the hole injection layer was 0.975 hours.

As shown in Figure 7(b), when driven by a direct current corresponding to an initial brightness of 21400 cd/m², the T50 service life of the QLED device designed with MoO_x/PEDOT:PSS as the hole injection layer was 2.897 hours.

As shown in Figure 7(c), when operated under a direct current corresponding to an initial brightness of 7069 cd/m², the T_{50} operating lifetime of the QLED device designed with MoO_x as the hole injection layer was 0.365 hours.

Using the formula $L_0^n * T_{50} = K$ (where K is a constant and $1 < n < 2$), with an acceleration factor n = 1.5, we converted their T_{50} service lives to an initial brightness of 100 cd/m², which were 1210 hours, 220 hours and 8511 hours respectively. Compared with the standard device, the service life of the MoO_x/PEDOT:PSS-based device was increased by about 7 times, while that of the MoO_x-based QLED device was reduced by 5.5 times. The test results show that introducing MoO_x between ITO and PEDOT:PSS can improve both the performance and lifetime of QLED devices.

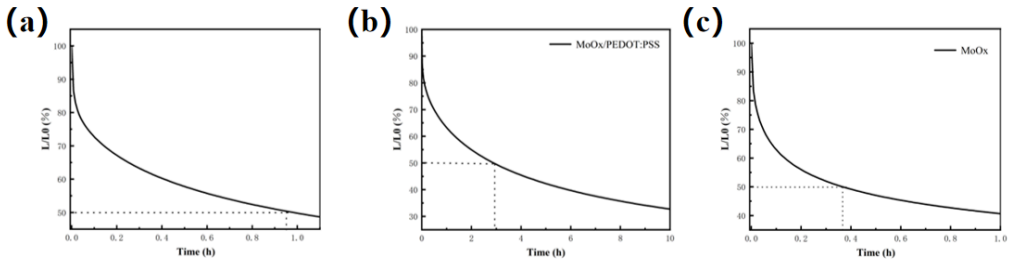

Figure 7. (a) Lifetime testing curve of the standard device at an initial brightness of 11610 cd/m².(b) Lifetime testing curve of the MoO_x/PEDOT:PSS-based QLED device at an initial brightness of 21400 cd/m².(c) Lifetime testing curve of the MoOx-based QLED device at an initial brightness of 7069 cd/m²

4. Conclusion

This work systematically explores the influence of MoO_3 interlayers on the optoelectronic performance of quantum dot light-emitting diodes (QLEDs). The introduction of the MoO_3 modification layer solved the problem of electrode corrosion by the traditional organic injection layer. Benefiting from better energy level matching, its maximum external quantum efficiency reached 17.06%, maximum current efficiency reached 121.21 cd/A, and maximum brightness reached 97340 cd/m². The service life of the improved device has been increased by 7 times.This proves the superiority of the inorganic/organic hybrid injection layer in regulating carrier balance. Although this study has made progress in improving QLED efficiency, future research can try to develop an all-inorganic charge transport system to completely eliminate the problem of easy aging of organic materials, so as to meet the requirements of harsh environments such as automotive displays and high-end lighting. Alternatively, further optimize the rheological parameters of the MoO_3 precursor solution to make it suitable for large-area inkjet printing technology, promoting the advancement of QLED from laboratory research to industrial mass production.

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