Improved Efficiency of All-Inorganic Quantum-Dot Light-Emitting Diodes via Interface Engineering

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As the charge transport layer of quantum dot (QD) light-emitting diodes (QLEDs), metal oxides are expected to be more stable compared with organic materials. However, the efficiency of metal oxide-based all-inorganic QLEDs is still far behind that of organic–inorganic hybrid ones. The main reason is the strong interaction between metal oxide and QDs leading to the emission quenching of QDs. Here, we demonstrated nickel oxide (NiO$_x$)-based all-inorganic QLEDs with a maximum current efficiency of 20.4 cd A$^{-1}$ and external quantum efficiency (EQE) of 5.5%, which is among the most efficient all-inorganic QLEDs. The high efficiency is mainly attributed to the aluminum oxide (Al$_2$O$_3$) deposited at the NiO$_x$/QDs interface to suppress the strong quenching effect of NiO$_x$ on the QD emission, together with the molybdenum oxide (MoO$_x$) that reduced the leakage current and facilitated hole injection, more than 300% enhancement was achieved compared with the pristine NiO$_x$-based QLEDs. Our study confirmed the effect of decorating the NiO$_x$/QDs interface on the performance enhancement of the all-inorganic QLEDs.

Keywords: NiO$_x$, all-inorganic, quantum dots, light-emitting devices, high efficiency

INTRODUCTION

Quantum dots (QDs) have many advantages including high color purity, high photoluminescence (PL) quantum yield (QY), and high stability, which make them promising luminescent materials for light-emitting diodes (LEDs) (Anikeeva et al., 2009; Bae et al., 2013; Shirasaki et al., 2013; Shen et al., 2015; Chen et al., 2018; Cao et al., 2019; Zhang et al., 2019). Recently, the performance of QD LEDs (QLEDs) has been improved greatly, the external quantum efficiencies (EQEs) for tricolor QLEDs have all surpassed 20%, with peak EQEs of 30.4% for red, 22.9% for green, and 19.8% for blue QLEDs, respectively (Wang et al., 2017; Shen et al., 2019; Song et al., 2019). At present, highly efficient QLEDs are mainly based on hybrid organic–inorganic structure, in which poly(3,4-ethylendioxythiophene) polystyrene sulfonate (PEDOT:PSS) is widely used as the hole injection layer (HIL); poly[N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)benzidine] (Poly-TPD), poly[9,9-dioctylfluorene-co-N-[4-(3-methylpropyl)]diphenylamine] (TFB), or poly(N-vinyl carbazole) (PVK) are adopted as the hole transport layer (HTL); and zinc oxide (ZnO) nanoparticles (NPs) are used as the electron transport layer (ETL) (Qian et al., 2011; Dai et al., 2014; Zhang et al., 2019). As we know, organic materials are sensitive to moisture and may degrade under high operating currents, which affect the stability of devices, and consequently, the strict encapsulation technology is indispensable. To solve this problem, it is necessary to seek more stable hole transport materials that can endure a high carrier density at high luminance.
Many inorganic metal oxides [nickel oxide (NiOx), tungsten oxide (WOx), molybdenum oxide (MoOx), vanadium oxide (VOx), etc.] have been applied as HIL in optical electronic devices to improve the device stability (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Moreover, NiOx possesses relatively proper band energy for efficient hole injection and electron blocking to confine the excitons in the QD emitting layer (Murase and Yang, 2012; Huu Tuan et al., 2014; Yang et al., 2014; Zhang et al., 2017), and NiOx is a promising hole transport material among them due to its nature of intrinsic p-type semiconductor with a wide bandgap and high transparency. Additionally, the QDs in octane solution exhibited a green emission, which is mainly attributed to two reasons. First, the excitons formed near the NiOx layer are subject to the surface of NiOx, and a large number of free carriers and defects/traps on the surface of adjacent NiOx HTL leads to the quenching of QD emission (Caruge et al., 2006, 2008; Wu and Yeow, 2010). It is reported that many dipolar surface species of NiOOH are present on the solution-processed NiOx films and induce a strong localized electric field, which facilitates radiationless decay channels with a charge-transfer/charging and/or energy transfer processes and leads to a severe decrease of device efficiency (Ratcliffe et al., 2011; Liu et al., 2015).

To address this issue, a modification of the NiOx/QD interface is needed. Several kinds of buffer layer have been inserted to suppress the exciton quenching induced by NiOx. By introducing ultrathin aluminum oxide (Al2O3) layer at the NiOx/QD layer interface (Zhang et al., 2016; Ji et al., 2017, 2018), Ji et al. fabricated highly efficient all-inorganic QLEDs, in which over 800% enhancement for the current efficiency/EQE of up to 34.1 cd·A−1/8.1% was achieved when the Al2O3 layer was obtained by atomic layer deposition (ALD), and this represented the highest EQE for all-inorganic QLEDs reported ever. With ultrathin lithium fluoride (LiF) being inserted at the NiOx/QD interface and ultrathin Al2O3 being inserted between the QDs and ZnO layer (Yang et al., 2018), Yang et al. reported highly efficient all-inorganic QLEDs with a maximum EQE of 6.52% and a long device lifetime of 16,120 h at 100 cd·m−2. Li et al. reported all-inorganic QLEDs of the highest maximum brightness of 40,000 cd·m−2 by sputtering ultrathin MgO at the NiMgO/QD interface; however, the maximum EQE is only 1.5% (Jiang et al., 2019). These results indicate the importance of decoration of the NiOx/QD interface on suppressing the QD emission quenching and improving the performance of the all-inorganic QLED efficiency. Among them, the Al2O3 buffer layer obtained by ALD technology has more advantages since the film thickness can be precisely controlled at atomic level by alternating the exposure cycles of trimethylaluminium [Al(CH3)3] and H2O, and the as-prepared films possess good uniformity over large substrates and excellent conformality on three-dimensional surface topologies. Furthermore, the hydroxyl (-OH) in the NiOOH species can be consumed during the exposure to Al(CH3)3 deposition cycles. Nevertheless, the maximum EQE for all-inorganic QLEDs with Al2O3 buffer layer is still very low, which is likely due to imbalanced carrier transport in devices resulting from the inefficient hole injection from indium tin oxide (ITO) to the NiOx layer and the relatively higher energy barrier between the NiOx layer and QD layer. To solve this problem, more researches are still needed to optimize the structure of NiOx-based all-inorganic QLEDs and improve the device efficiency.

Here, we demonstrated highly efficient all-inorganic QLEDs with an optimized structure of ITO/solution-processed MoOx (sMoOx)/NiOx/Al2O3/QDs/ZnO/Al through all solution-process method except for Al2O3 layer and the electrodes. The ultrathin Al2O3 inserted at the NiOx/QDs interface was to suppress the strong quenching effect of NiOx on the emission of QD. And the sMoOx introduced before the NiOx layer was aimed to reduce leakage current and facilitate the hole injection from anode to the emitting layer and minimize the hole-blocking effect of Al2O3 layer. Our resultant all-inorganic QLEDs reached a high current efficiency of 20.4 cd·A−1 and a maximum EQE of 5.5%, more than 300% enhancement was achieved compared with the pristine NiOx-based QLEDs.

**MATERIALS AND METHODS**

**Preparation of Green Quantum Dots and Metal Oxide Solution**

Cadmium selenide (CdSe)/zinc sulfide (ZnS) QDs were synthesized according to the method reported in the literature (Li et al., 2019). The QDs in octane solution exhibited a green emission with the PL peak at 525 nm (Supplementary Figure 1). The NiOx precursor was prepared by a modified method (Mashford et al., 2010); the mixture of nickel acetate tetrahydrate [Ni(OAc)2·4H2O; purchased from Aldrich] and equimolar quantity of monoethanolamine (MEA; purchased from Aldrich) in ethanol was heated at 60°C for 2 h and stirred overnight. 0.1 M MoOx solutions were synthesized by a thermal decomposition method using ammonium heptamolybdate [(NH4)2Mo7O24·4H2O] as a precursor (Murase and Yang, 2012; Vu et al., 2016). The ZnO NPs were prepared by slowly mixing 0.1 M zinc acetate in dimethyl sulfoxide (DMSO) and 0.3 M tetramethylammonium hydroxide (TMAH) in ethanol together for 1 h, and the ZnO particles were precipitated by adding hexane/ethanol to the solution.

**Fabrication of Quantum Dot Light-Emitting Diode Devices**

The all-inorganic QLED structure consists of ITO/MoOx/NiOx/Al2O3 (x cycles)/QDs/ZnO/Al. The NiOx, QDs, and ZnO are used as HTL, emission layer, and ETL, respectively. Before fabricating the devices, the ITO substrates were ultrasonically cleaned in detergent, DI water, acetone, and isopropyl alcohol for 15 min successively followed by an ex situ UV ozone treatment in air for 15 min. This as-prepared MoOx precursor solution was spin-coated onto the UV ozone-treated ITO substrates at 4,000 rpm and then baked at 120°C for 10 min to get the MoOx film. Then, the NiOx precursor was spin-coated at 2,000 rpm and annealed at 275°C for 30 min in air to obtain.
a highly conductive layer. The Al₂O₃ layer was deposited by alternating exposures of Al(CH₃)₃ and H₂O with the same substrate and maintaining the temperature at 200°C, and the thickness is approximately 0.1 nm for each ALD cycle. Al₂O₃ layers with different thicknesses were deposited on the NiOₓ films for device A (zero cycle), B (one cycle), C (two cycles), and D (three cycles), respectively. Note that no MoO₃ layers were inserted for devices A to C, and device A is a control device without the Al cathode lines with a width of 2.0 mm were deposited orthogonally to the 2 mm ITO anode lines to form a 4 mm² active area.

Measurements and Characterization

Current density–voltage–luminance (J–V–L) characteristics of QLEDs were tested using a Keithley 2400 source meter and a picoammeter (Keithley 6485) with a calibrated Newport silicon diode under ambient conditions. The luminance was calibrated using a Minolta luminance meter (CS-100). The electroluminescence spectra were obtained with an Ocean Optics spectrometer (USB2000, relative irradiance mode) and a Keithley 2400 source meter. The room temperature PL spectrum of the QLEDs was assessed by atomic force microscopy (AFM; Bruker Multimode-8). The UV photoelectron spectroscopy (UPS; Thermo Scientific ESCALAB 250 XI) measurement was performed using a He I discharge lamp (hv = 21.22 eV) under high vacuum (2.5 × 10⁻⁸ mbar) and the UPS spectra of MoO₃ and NiOₓ was measured (Supplementary Figure 4).

RESULTS AND DISCUSSION

The composition of the solution-processed NiOₓ films was studied by XPS analysis. Figure 1A shows the XPS spectrum for Ni 2p₃/2 state possessing three peaks. The first peak centered at a binding energy of 854.2 eV corresponds to Ni²⁺ in the standard Ni-O octahedral bonding configuration in cubic rock salt NiOₓ. The adjacent peak shoulder located at 855.9 eV was ascribed to Ni²⁺ vacancy-induced Ni³⁺ ion and NiOOH (Sasi and Gopchandran, 2007; Manders et al., 2013). The broad peak centered at 861.0 eV has been ascribed to a shake-up process in the NiO structure. Figure 1B shows the XPS spectrum for the O 1s state possessing three peaks. The first peak centered at 531.2 eV is indicative of nickel hydroxides and oxyhydroxides, including defective NiOₓ with hydroxyl groups adsorbed on the surface (Han et al., 2006; Ratcliff et al., 2011).

The morphology evolution of each layer within the QLEDs was assessed by atomic force microscopy (AFM; Figure 2). The root-mean-square (RMS) roughness of pure ITO (Supplementary Figure 2) is 2.38 nm, and the value decreased slightly as the layer number increased, which showed 1.51 nm for QD layer and 2.12 nm for ZnO layer, respectively. Since Al₂O₃ is an insulating material, it is very important to control its thickness precisely via the ALD process. To get the optical thickness of Al₂O₃, we first fabricated all-inorganic QLEDs consisting of a structure of ITO/NiOₓ/Al₂O₃/QDs/ZnO/Al. Different deposition cycles of Al₂O₃ (0C, 1C, 2C, 3C) were applied at the NiOₓ/QD interface, and the corresponding photoelectrical properties of devices were characterized and shown in Figure 3. Al₂O₃ showed

![FIGURE 1](image-url) | The X-ray photoelectron spectroscopy (XPS) spectra for Ni 2p (A) and O1s (B).
a remarkable influence on the performance of all-inorganic QLEDs. The current density decreased evidently with the increasing thickness of AlO

| Sample       | PL Peak (nm) | Lifetime (ns) |
|--------------|--------------|---------------|
| QD in octane | 525          |               |
| F1: Glass/QD | 528          | 7.3           |
| F2: ITO/NiO/QD | 532        | 5.2           |
| F3: ITO/NiO/AlO

To study the effect of ultrathin AlO

It is reported that the sMoO

$\text{Al}_2\text{O}_3$, aluminum oxide; ITO, indium tin oxide; NiO

Summary of the PL peak and the decay lifetime for different samples.

samples were summarized in Table 1. It can be seen that the emission of QD film on glass substrate was peaked at 528 nm with an exciton lifetime of 7.3 ns, while that on NiO

FIGURE 2 | Atomic force microscopy (AFM) images of (A) indium tin oxide (ITO)/nickel oxide (NiO

TABLE 1 | Summary of the PL peak and the decay lifetime for different samples.

It is reported that the sMoO

$\text{Al}_2\text{O}_3$, aluminum oxide; ITO, indium tin oxide; NiO

$\text{Al}_2\text{O}_3$ showed a higher work function of 5.6 eV, better transparency, and smoother surface morphology, providing the QLEDs with good Ohmic contact and small charge transfer resistance (He et al., 2013; Vu et al., 2016). The device structure was further optimized by using sMoO

$\text{Al}_2\text{O}_3$ modified layer, which reduced the leakage current and led to a more balanced carrier injection in emitting layer. For device III possessing sMoO

The exciton lifetimes for different film
FIGURE 3 | Photoelectric properties of quantum dot light-emitting diodes (QLEDs). (A) Voltage vs. current density (V–J), (B) voltage vs. luminance (V–L), (C) current density–luminous efficiency, and (D) current density–external quantum efficiency. 0 C means 0 cycle deposition of aluminum oxide (Al₂O₃), each cycle is about 0.1 nm.

FIGURE 4 | (A) Steady-state and (B) time-resolved photoluminescence (PL) spectra of samples with and without the aluminum oxide (Al₂O₃) layer.

TABLE 2 | Summary of the electrical properties of the QLEDs.

| Device | λ_max (nm) | V_T (V) | L_{max} (cd/m²) | EQE_{max} (%) | η_{Amax} (cd/A) | η_{Pmax} (lm/W) |
|--------|------------|---------|-----------------|---------------|----------------|----------------|
| I      | 532        | 3.9     | 3,786 (7.6 V)   | 1.7 (5.3 V)   | 7.5 (5.3 V)    | 4.5 (5.3 V)    |
| II     | 534        | 4.3     | 4,930 (8.5 V)   | 4.3 (5.2 V)   | 15.9 (5.2 V)   | 9.6 (5.2 V)    |
| III    | 534        | 4.7     | 9,140 (9.0 V)   | 5.5 (6.2 V)   | 20.4 (6.2 V)   | 10.7 (5.8 V)   |

EQE, external quantum efficiency; QLED, quantum dot light-emitting diode.
FIGURE 5 | Photoelectric properties of devices I, II, and III. (A) Voltage vs. current density (V–J), (B) voltage vs. luminance (V–L), (C) luminance–current efficiency, and (D) luminance–external quantum efficiency. The inset in (B) is the energy level diagrams of quantum dot light-emitting diode (QLED) III.

FIGURE 6 | Electroluminescence (EL) spectra of devices I (A) and III (B) under different voltages.

and better tolerance to higher operating voltage for device III than the other two. It is also confirmed from the EL spectra under increasing driving voltage of devices I and III (Figure 6). The EL peak for device I without Al₂O₃ layer exhibited a red shift of 4 nm as the voltage increased to 8 V, while that for device III kept its profile from 5 to 10 V. Despite the slightly higher turn-on voltage, the insertion of sMoOₓ layer combining Al₂O₃ layer in NiOₓ-based all-organic QLEDs improved not only the device efficiency but also the performance stability.

A comparison of the performance of all-inorganic QLEDs between our work and others in literature was summarized (see Supplementary Table 2).

CONCLUSION

All-inorganic QLEDs with high efficiency were fabricated using solution-processed NiOₓ as the HTL and ZnO as the ETL, and ultrathin Al₂O₃ was deposited at the NiOₓ/QDs interface by the
ALD process to reduce the strong quenching effect of NiOx on the QD emission. The corresponding all-inorganic QLEDs exhibited a maximum current efficiency of 19.8 cd A\(^{-1}\) and EQE of 4.5\%, which is 260\% enhancement compared with the QLEDs without Alox insertion, making them among the highest efficient inorganic QLEDs. This result suggests that the Alox passivating layer is critical to device efficiency improvement by suppressing QDs emission quenching induced by NiOx. Despite great device improvement, the maximum EQE for NiO\(_x\) all-inorganic QLEDs is still below 10\%, which is probably due to the relatively lower hole mobility of NiOx and higher energy barrier for hole transfer from NiOx to the QD layer, resulting in an imbalanced charge injection in devices. The energy level regulating as well as improving electrical performance of NiOx are vital strategies to fabricate high-performance all-inorganic QLEDs.

DATA AVAILABILITY STATEMENT

All datasets generated for this study are included in the article/Supplementary Material.

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AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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SUPPLEMENTARY MATERIAL

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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