

High Efficiency Cadmium-free Red Quantum Dot-Light Emitting Diodes

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ABSTRACT

We report a high efficiency inverted red indium phosphide (InP) based quantum dot-light emitting diode (QD-LED) by optimizing InP-QD properties as well as interfacial contact between electron transport layer and emissive QDs, and applying self-aging approach. Our QD-LED exhibits substantial improvement in the external quantum efficiency from 4.42 to 10.2% after several days of self-aging.

1. INTRODUCTION

Quantum dot-light emitting diode (QD-LED) has been considered as one of the favorable technology for the next generation display applications owing to its numerous advantages such as high color purity, wide color gamut, and simple and cost-effective fabrication process.¹ In recent years, significant research works have been performed on the development of efficient cadmium based QD structure and improvement of its properties for instance high photoluminescence quantum yield (PLQY), and air stability, etc. Therefore, cadmium-based QD-LED revealed significantly high and comparable efficiency (especially for red and green) with organic light emitting diode. Similarly, device stability is also steadily improving towards the required practical limit. However, cadmium-based QDs have limited practical applications considering its impact on the environment due to cadmium toxicity.² As an alternative InP based QDs have been reported as a promising contender for next-generation display applications because of its no effect on the health and environment.³ However, the efficiency and stability of InP based QD-LEDs are still lagging compared with the cadmium counterparts due to low PLQY in film state and poor air-stability. Likewise, the device efficiency is also limited by the charge balance issue in the emissive QDs, which is mostly associated with the noteworthy charge injection barrier between the transporting layers and emissive QDs as well as the mobility of transporting layers. Previously, several approaches such as new transport layers for improvement of charge injection into the emissive QDs, QD structure modifications have been largely reported to improve the performances of QD-LED.^{4,5} Very recently, Su et. al and Acharya et. al reported an improvement in the conventional cadmium based QD-LED efficiency after several days of aging.^{6,7} They demonstrated two different mechanisms, for

instance the interfacial reaction between electron transport layer (ETL) and Al cathode as well as the reaction between ETL and encapsulation resin behind the improvement of device efficiency. Therefore, developing cadmium-free QD-LED with high efficiency and long device lifetime is vital to make this technology a feasible alternative for next-generation display applications.

The main objective of this research work is to present an efficient approach to attain high efficiency in cadmium-free QD-LED. Here, we developed a high-efficiency red cadmium-free inverted QD-LED by optimizing an interfacial contact between emissive InP-QDs and electron transporting layer and applying aging test. Our inverted red InP based QD-LED with optimized interfacial contact exhibits a maximum external quantum efficiency (EQE) of about 4.42% and it is further improved to 10.2% after several days of aging. This result shows an almost 2.3-fold improvement in the efficiency of the device with several days of aging. We anticipate that our approach will raise the flexibility of InP comprised QD-LED for the development of next-generation QD based display applications.

2. EXPERIMENT

The indium tin oxide (ITO) patterned glass substrates were sequentially cleaned using acetone, isopropyl alcohol and deionized water followed by UV-ozone treatment for 10 min. Initially, ZnMgO nanoparticles (NPs) dispersed in ethanol were spin-coated onto the ITO/glass substrates and annealed at 230 °C for 30 min. Subsequently, red InP-QDs (10 mg/ml in octane) were spin-coated at 3000 rpm in the nitrogen filled glove box. Finally, organic hole transport and injection layers and cathode were successively deposited onto InP-QD layer using thermal evaporator under high-vacuum evaporation chamber.

3. RESULTS AND DISCUSSION

Here, red InP-QDs with core/shell structure is used for the evaluation of QD-LED performances. Fig. 1 shows the absorption and photoluminescence spectra of our colloidal InP-QDs. This InP based QDs displays maximum photoluminescence peak at 626 nm with a

narrow FWHM of about 38 nm. Additionally, our InP-QDs exhibit an absolute PLQY of about 80%, which is

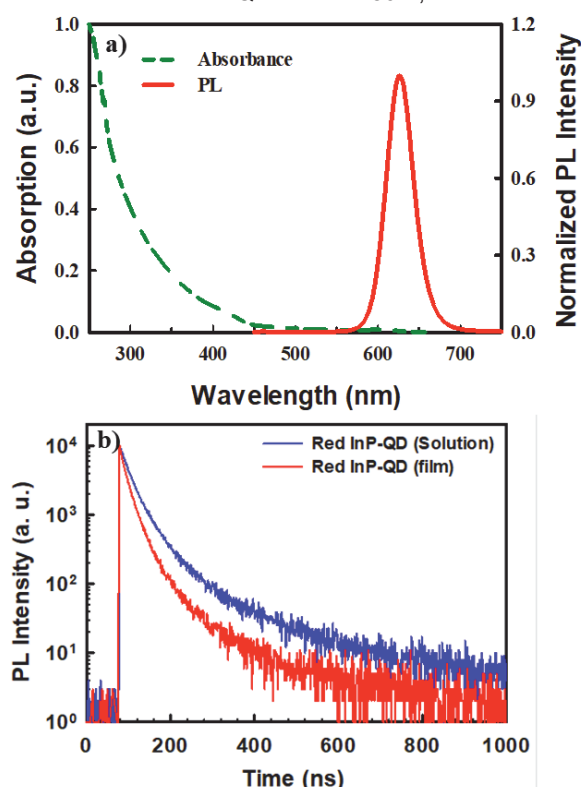


Fig. 1 Absorption and photoluminescence spectra of our InP-QDs.

comparable with the widely used cadmium based QDs.⁸ Likewise, our QDs also exhibit an exciton decay lifetime of about 48 ns and 28 ns, in the solution and film states, respectively.

Normally, ZnO is widely used as an ETL in QD-LED because of its facile solution processing and good optical as well as electrical properties. However, it shows strong exciton quenching at the interface between emissive QDs and ZnO NPs by the surface defects, and interfacial charge transfer process, and thereby noteworthy reduction in the device performances. Hence, to suppress an exciton quenching at the interface of ZnO/InP-QD, we optimize the interfacial property of ZnO by doping different concentration of Mg. The exciton decay lifetime of the InP-QD deposited onto ZnMgO (24 ns) is significantly improved compared with the pristine ZnO layer (11.3 ns). Such improvement in the exciton decay lifetime of ZnMgO is originated from the reduced interfacial charge transfer process and suppressed non-radiative process. To confirm the influence of different Mg doping concentrations (ZnMgO NPs) on the performances of InP comprised QD-LED, inverted red devices are fabricated with the following configuration. Device: Glass/ITO/ETL/InP-QD/TCTA/TAPC/HATCN/Al. For a valid comparison of device performances, the control device is also fabricated with widely used ZnO as

the ETL.

Our QD-LED with the ZnMgO NPs (optimized Mg doping concentration) layer displays a relatively high

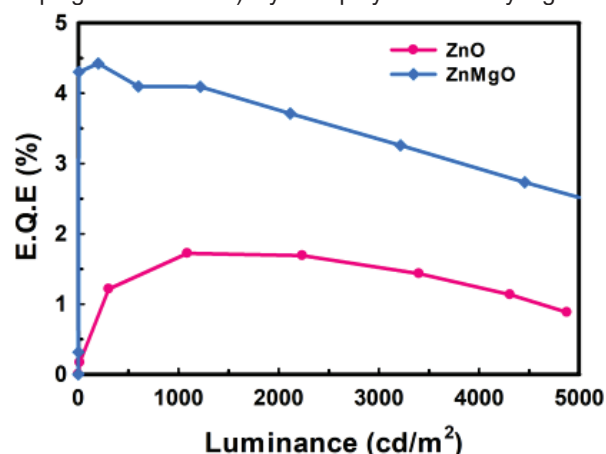


Fig. 2 EQE versus luminance characteristics of inverted red InP based QD-LED with ZnO and ZnMgO based ETL layer.

driving voltage of about 4.36 V (at the luminance of 1000 cd/m²) compared with the ZnO layer (3.47 V). Such a high driving voltage in ZnMgO based device is attributed to its lower conductivity and relatively higher conduction band minimum. The QD-LED with ZnMgO also reveals significant improvement in maximum EQE of about 4.42%, which is almost 1.8 times higher than the reference device (1.72%) (Fig. 2). Similarly, the luminance of the ZnMgO based device is improved from 4875 to 7813 cd/m². These results indicate a significant improvement in the charge balance of the ZnMgO based QD-LED through the reduced conductivity of the ETL and suppressed exciton quenching.

By considering the impact of aging on the conventional device efficiency, we also investigate the effect of aging on the performances of our optimized inverted red InP based QD-LED. Fig. 3 shows the EQE versus luminance characteristics of an inverted red QD-

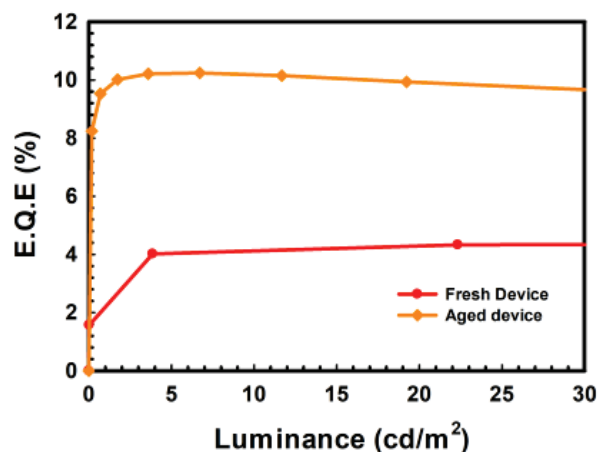


Fig. 3 EQE versus luminance characteristics of optimized inverted red InP based QD-LED before and after several days of aging.

LED device with aging. As shown in Fig. 3, EQE increases with an increasing aging period. The device with several days of aging shows a maximum EQE of about 10.2%. This result shows a more than two-fold increase in efficiency after several days of aging. To the best of our knowledge, this is one of the highest EQE values reported for InP based QD-LED so far. To find the exact reason behind improvement of efficiency in QD-LED after several days of aging, several measurements such as single carrier devices (HOD and EOD), ultraviolet photoelectron spectroscopy, time-resolved PL and X-ray photoelectron spectroscopy are performed. These measurements show decreased in the conductivity and variations in the conduction band minimum of ZnMgO after several days of aging. More results about this investigation will be covered in the presentation.

4. CONCLUSIONS

Here, we demonstrate a high performance inverted red InP based QD-LED by optimizing an interfacial contact between emissive QDs and ETL. Additionally, we also investigate the effect of aging on the performances of optimized inverted red InP based QD-LED. Our optimized QD-LED reveals an improvement of maximum EQE from 4.42 to 10.2%, which is more than 2 times higher than the fresh device. To the best of our knowledge, this is one of the highest EQE value reported for inverted red InP based QD-LED. We anticipate that our findings will be very helpful in attaining high performances in the next generation QD based display devices.

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