Electronic Supplementary Information

Significant Enhancement in Quantum-dot Light Emitting Device Stability via a ZnO:Polyethylenimine Mixture in the Electron Transport Layer

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TRPL of QD exciton lifetime on ZnO:PEI

QD exciton lifetime was evaluated by time-resolved photoluminescence (TRPL). For comparison, QD solution was spin-coated on ZnO as well as ZnO:PEI with PEI concentrations of 0.1 wt%, 0.3 wt%, and 0.6 wt%. QD on ZnO:PEI exhibits longer lifetime than QD on ZnO, suggesting exciton quenching sites in ZnO are passivated by dispersed PEI molecules in blended film. As can be seen in figure S1, higher PEI concentration does not extensively improve QD exciton lifetime. Comparing to QD exciton lifetime on ZnO/PEI structure, blending PEI does not fully remove structural defects in ZnO within the range of concentration in this experiment.

Driving voltage dependance of FIrpic emission in QDLEDs with marking layer

10 nm of CBP:FIrpic (10%) marking layer was inserted in HTL for examining excessive electrons in HTL. Figure S2 (a) and (b) depicts the EL spectra of QDLEDs with a marking layer with an applied bias of 6V to 10V. Figure S3 (a) shows that QDLEDs with ZnO exhibit negligible emission from FIrpic in entire voltage range, suggesting excessive electrons cannot reach CBP:FIrpic marking region. In figure S3 (b), FIrpic emission in a QDLED with ZnO/PEI slightly increases at 10V, suggesting that excessive electrons reach CBP:FIrpic marking layer. It could be expected that higher applying voltage will increase FIrpic emission in QDLED with ZnO/PEI. FIrpic emission greatly increases along with voltage range from 6V to 10V in QDLEDs with ZnO:PEI 0.1 wt% and 0.3 wt% as can be shown in figure S3 (c) and (e). On the other hand, in higher voltage range in figure S3 (d) and (f), FIrpic emission ratio decreases as voltage increases. This may be because either electrons and holes move further toward the opposite direction, i.e electrons and holes drift toward anode and QD, respectively, or excitons in CBP:FIrpic layer are dissociated by strong electric field. This result indicates electric field forces excessive electrons to overcome the QD/HTL interface.

Delayed electroluminescence measurement

The experimental setup for delayed EL measurements is schematically illustrated in figure S4. An optical chopper (shutter) is opened and closed for on-off transition of EL data acquisition. The chopper is closed until shortly after the forward bias ends so that the optical fiber does not capture the prompt EL signal. Once the chopper opens, delayed EL signal is measured by the optical fiber and amplified through a photomultiplier tube. In order to investigate the recombination and residual charge distribution effects, a 0.2 ms reverse bias pulse is applied during the delayed EL data acquisition period.

Figures



Figure S1. TRPL characteristics of the QDs on ZnO:PEI with different PEI concentrations.



Figure S2. EL spectra from devices containing the CBP:Firpic marking layer with the various ETLs at (a) 6V and (b) 10V.



Figure S3. EL spectra from devices containing the CBP:FIrpic marking layer with (a) ZnO ETL, (b) ZnO/PEI ETL, (c-d) ZnO:PEI_{0.1} ETL, and (e-f) ZnO:PEI_{0.3} ETL, at various forward bias voltages



Figure S4 Schematic illustration of possible changes in charge distribution in the HTL and QD layers in the devices with (a) ZnO ETL, (b) ZnO/PEI ETL, and (c) ZnO:PEI ETL.



Figure S5. Schematic images of delayed electroluminescence measurement setup. Timings of shutter open/close, bias pulse, and electroluminescence are illustrated.

ETL	LT50 for L_0 at 20 mA cm ⁻²	LT50 for $\rm L_0$ 100 cd $\rm m^{-2}$	LT50 for $\rm L_0$ 1000 cd $\rm m^{-2}$	Maximum EQE
ZnO	46 hours for 2,500 cd $m^{\text{-}2}$	15,100 hours	239 hours	7.7 %
ZnO/PEI	62 hours for 3,030 cd $m^{\text{-}2}$	28,773 hours	456 hours	11.2 %
ZnO:PEI (0.1 wt%)	140 hours for 3,000 cd $m^{\text{-}2}$	65,630 hours	1,011 hours	11 %
ZnO:PEI (0.2 wt%)	101 hours for 3,050 cd $m^{\text{-}2}$	42,430 hours	752 hours	13.8 %
ZnO:PEI (0.3 wt%)	292 hours for 3,250 cd $m^{\text{-}2}$	153,735 hours	2,437 hours	12.1 %

Table S1 LT50 and EQE of QDLEDs with different ETL structures. Expected LT50 for $L_0=100$ cd m⁻² and $L_0=1000$ cd m⁻² are calculated by lifetime relation equation, $L_0^n LT50 = constant$. Acceleration factor n of 1.8 is used and LT50 is based on a measured value under 20 mA cm⁻² of constant current.