

Supporting Information

Blue ZnSeTe Quantum Dot Light-Emitting Diodes with Low Efficiency Roll-Off Enabled by *In-situ* Hybridization of ZnMgO Nanoparticles and Amino Alcohol Molecules

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Experimental section

Materials

Triethylamine (TOA, $\geq 99.0\%$), oleic acid (OA, 90%), oleylamine (OAm, 70%), diethylzinc (Et_2Zn , 1.0 M in hexane), zinc acetate ($\text{Zn}(\text{OAc})_2$, 99.99%), zinc chloride (ZnCl_2 , anhydrous, 99.95%), sulfur (flakes, $\geq 99.99\%$), selenium (granules, 99.99%), tellurium (99.999%), diphenylphosphine (DPP, 98%), HF (48%), trioctylphosphine (TOP, 97%), Zinc acetate dihydrate ($>99\%$), magnesium acetate tetrahydrate ($>98\%$), TMAH ($>97\%$), dimethyl sulfoxide (DMSO, $>99.9\%$), EA (99%), PA (99%) and BA (99%) were purchased from Sigma Aldrich. Anhydrous ethanol (99.5%), anhydrous methanol (99.5%), and ethyl acetate ($>99.5\%$) were purchased from Acros. PEDOT:PSS and TFB were acquired from Aladdin. Chlorobenzene (99.9%) were purchased from Alfa-Aesar. All chemicals were used without further purification.

Synthesis of ZnTeSe cores

OA (13.5 mmol) and OAm (9 mmol) were mixed with TOA (50 ml) in a four-neck flask by stirring and heated to 120 °C under vacuum. Then, the temperature was further increased to 240 °C under N_2 flow (800 ml min^{-1}). Et_2Zn (9 mmol) was added into the reaction medium, and then a mixture of 2 M Se in TOP (4.5 mmol), 0.1 M Te in TOP (0.3 mmol) and DPP (4.5 mmol) was injected quickly to initiate the reaction. During the injection of Et_2Zn , careful manipulation of the pressure under the N_2 flow was required. After 40 min, the ZnTeSe cores were isolated by centrifugation using ethanol and were dispersed in hexane.

Synthesis of ZnTeSe/ZnSe/ZnS QDs

$\text{Zn}(\text{OAc})_2$ (4.8 mmol) and OA (9.6 mmol) were mixed with ODE (20 ml) by stirring and heated to 120 °C under vacuum for 2 h. After the atmosphere was transferred to N_2 , the mixed solution was heated to 300 °C and Se/Te precursors (4.5 mmol Se in 5 mL DPP and 0.3 mmol Te in 3 mL TOP) was injected quickly to the reaction system and kept for 300 °C for 1 hours. For ZnSe and ZnS shell growth, 12 mmol of $\text{Zn}(\text{OA})_2$ in ODE and 1 M TOP-Se (12 mmol) were added and reacted for 1 h. Subsequently, 12 mmol of $\text{Zn}(\text{OA})_2$ in ODE and 1 M TOP-S (12 mmol) were added and further reacted for 1 h. The resulting ZnTeSe/ZnSe/ZnS QDs were centrifuged using ethanol and dispersed in octane.

Synthesis of post-treated ZMO NPs

593 mg of Zn acetate dihydrate and 64.3 mg of Mg acetate tetrahydrate (Zn:Mg = 9:1, molar ratio) were dissolved in 30 mL of DMSO. 10 mL of TMAH solution (0.5 M in ethanol) was added to the Zn:Mg acetate solution, and the mixture solution was stirred for 2 h. After synthesis, the ZMO NPs were precipitated by adding ethyl acetate in a 2:1 ratio to the synthesis mixture and centrifuging. The supernatant was discarded, the dry ZMO NPs were rediluted in ethanol, and 60 μL of ethanolamine was added to help with redispersion and stabilize the colloidal solution. The ZMO NPs were finally washed a second time, and the dry powder was collected and redispersed in ethanol for use.

Synthesis of *in-situ* hybrid ZMO NPs

593 mg of Zn acetate dihydrate and 64.3 mg of Mg acetate tetrahydrate (Zn:Mg = 9:1, molar ratio) were dissolved in 30 mL of DMSO. Add an appropriate amount of amino alcohol (metal cation: amino alcohol=2:1, molar ratio) to the precursor solution and stir for 5 min. 10 mL of TMAH solution (0.5 M in ethanol) was added to the Zn:Mg acetate solution, and the mixture solution was stirred for 2 h. After synthesis, the ZMO NPs were precipitated by adding ethyl acetate in a 2:1 ratio to the synthesis mixture and centrifuging, and the dry powder was collected and redispersed in ethanol for use.

Fabrication of QLEDs

ITO substrate was cleaned by ultrasonic stirring with detergent, deionized water, acetone, and isopropanol, and then treated with ultraviolet ozone for 30 min. PEDOT:PSS solution was spin-coated at 4000 rpm for 40 s and thermally backed at 150 °C for 15 min. Then, the TFB dissolved in chlorobenzene with a concentration of 10 mg mL⁻¹ was spin-coated at 3000 rpm and annealed at 120 °C for 20 min. QD solution (15.0 mg mL⁻¹, dispersed in n-octane) was spin-coated at 3000 rpm for 40 s and annealed at 80 °C for 10 min. After that, different types of ETL (30 mg mL⁻¹) were dynamic-spin-coated on the QDs layer and then annealed at 90 °C for 20 min. Finally, the Al electrode (120 nm) was deposited by thermal evaporation under vacuum of $\approx 3 \times 10^{-4}$ Pa. For TFB/R/B/ETL QD double layer QLEDs, 3 mg mL⁻¹ of red QDs and 15 mg mL⁻¹ of blue QDs were coated with 3000 rpm 40 s annealed at 80 °C, sequentially. Other functional layers were coated as in the regular devices.

Characterization

UV-vis absorption spectroscopy was conducted using a U-3100 spectrophotometer (Hitachi). The PL spectra were measured using an Edinburgh FLS920 PL spectrometer. FTIR analysis was performed with a Thermo Scientific Nicolet iS20 instrument. TEM imaging was carried out using JEM1011 (JEOL) electron microscopes at an operating voltage of 100 kV. XRD analysis was conducted using a Bruker D8 Advance diffractometer. Time-resolved PL measurements were collected using a fluorescence lifetime measurement system (QM/TM/NIR, PTI, America). Elemental analysis was studied using XPS spectrum analysis with a Perkin-Elmer model PHI 5600 instrument. Photoluminescence quantum yield was measured using an Edinburgh fluorescence spectrometer FS5. UPS measurements were performed using a Thermo Fischer ESCALAB 250Xi instrument. The performance of QLEDs was investigated using a Keithley 2400 source meter and a PR-670 Spectra Colorimeter. The half-lifetime of QLEDs was tested using a ZJZCL-1 OLED ageing lifespan test instrument.

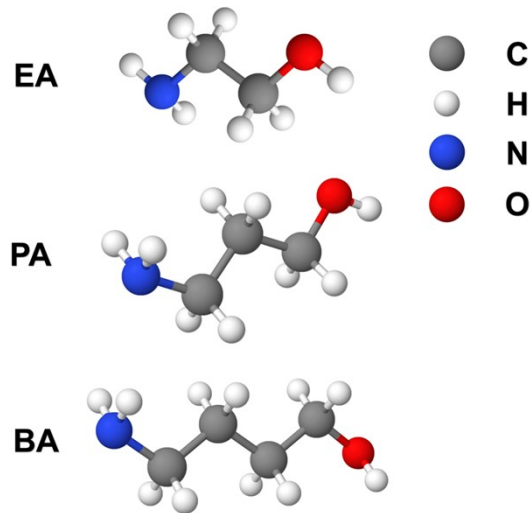


Figure S1. Chemical structure formula of different amino alcohols (EA, PA, BA).

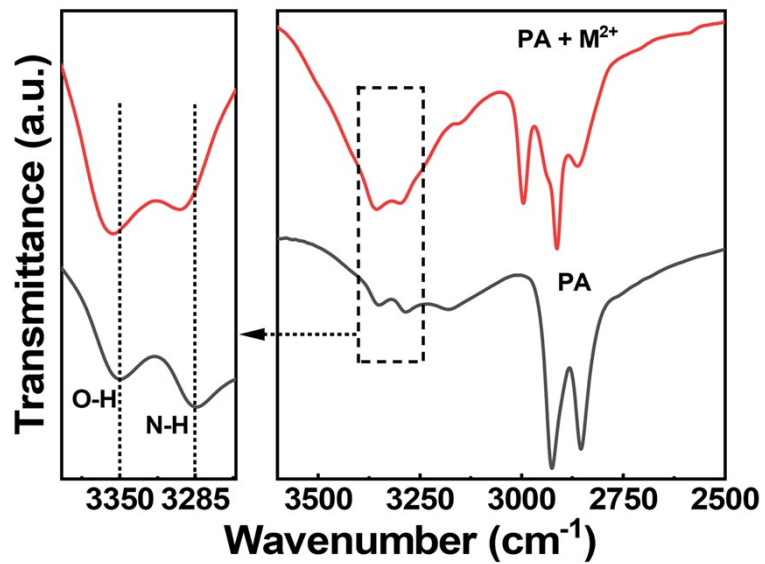


Figure S2. FTIR spectra of PA and M^{2+} with PA molecules.

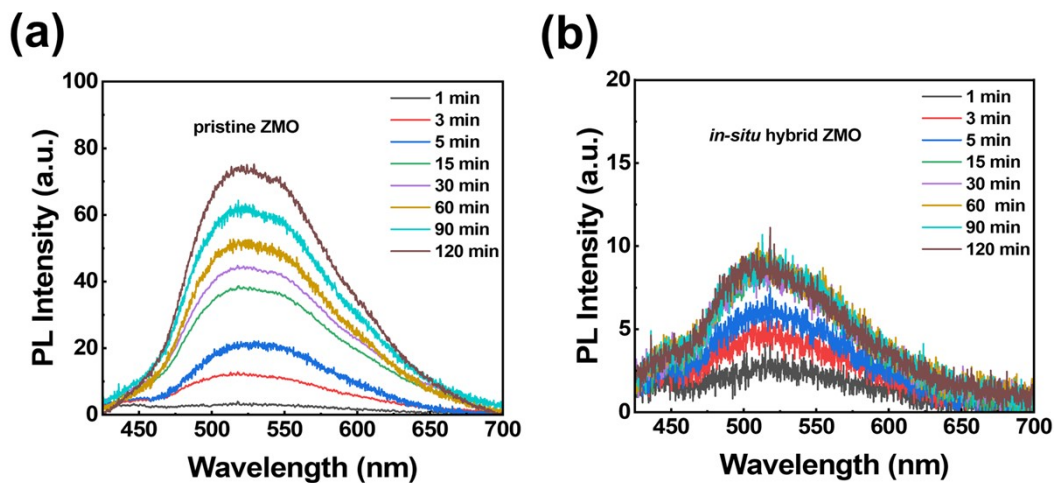


Figure S3. Variation of PL spectra of (a) the pristine ZMO NPs and (b) *in-situ* hybrid

ZMO NPs as a function of reaction time, respectively.

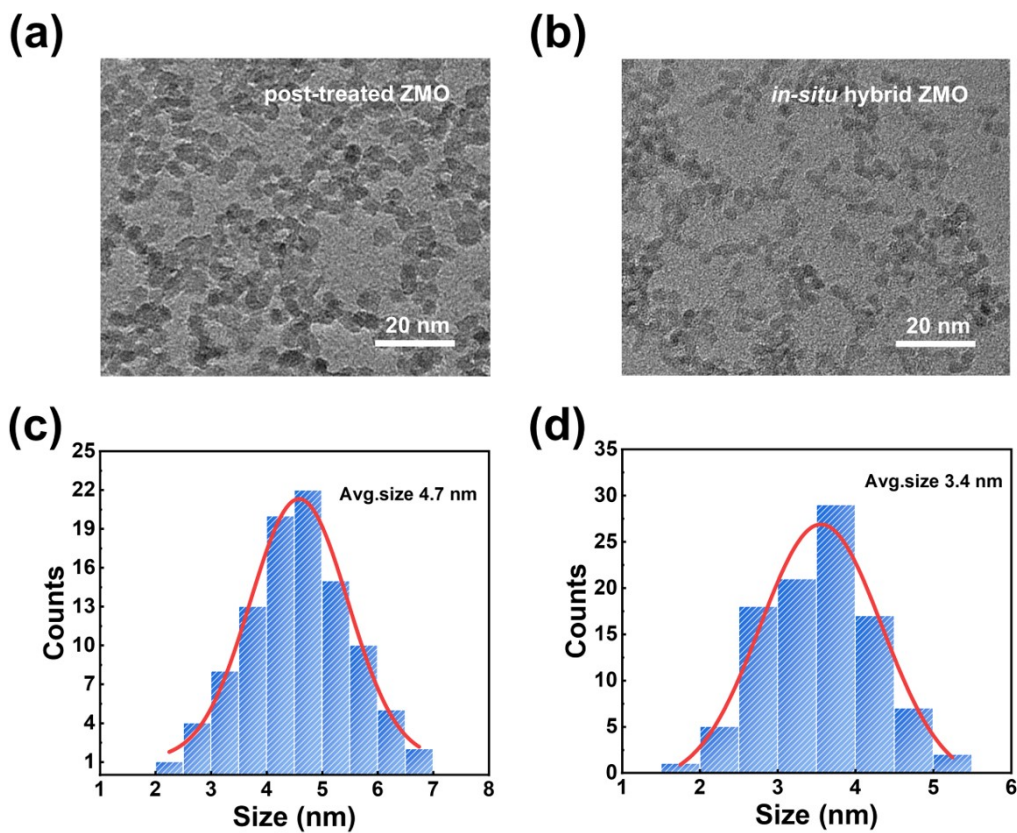


Figure S4. TEM images of (a) the post-treated and (b) *in-situ* hybrid ZMO NPs, respectively. (c) and (d) are the responding size histograms.

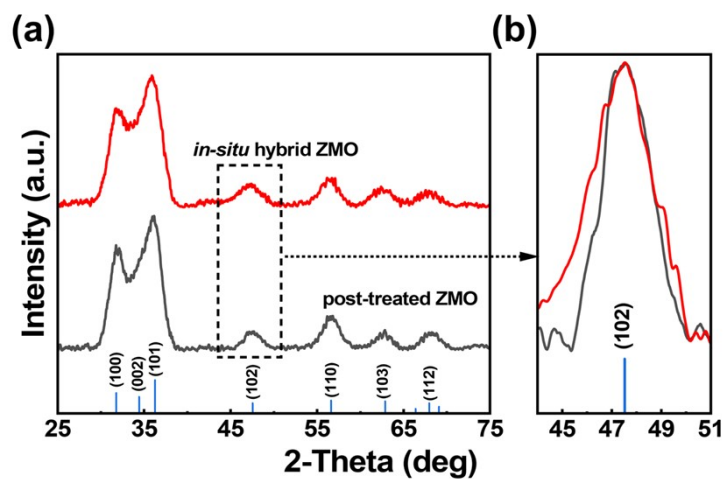


Figure S5. (a) XRD pattern of the post-treated and *in-situ* hybrid ZMO NPs. (b) Zoomed-in views of the diffraction peak at $\sim 47.5^\circ$ in (a).

Table S1. The exciton lifetime fitting from the time-resolved PL data.

Sample	A ₁ (%)	τ_1 (ns)	A ₂ (%)	τ_2 (ns)	τ_{avg} (ns)
Glass/QDs	81.1	18.5	18.9	49.2	24.30
Glass/post-treated ZMO/QDs	89.7	6.9	10.3	22.3	8.49
Glass/ <i>in-situ</i> hybrid ZMO/QDs	86.5	10.4	13.5	34.4	13.64

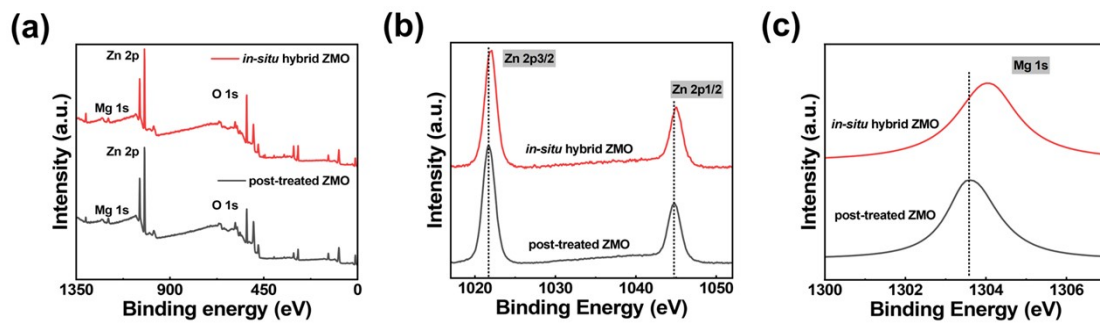


Figure S6. XPS spectra of (a) wide scans, (b) Zn 2p and (c) Mg 1s of the post-treated and *in-situ* hybrid ZMO films.

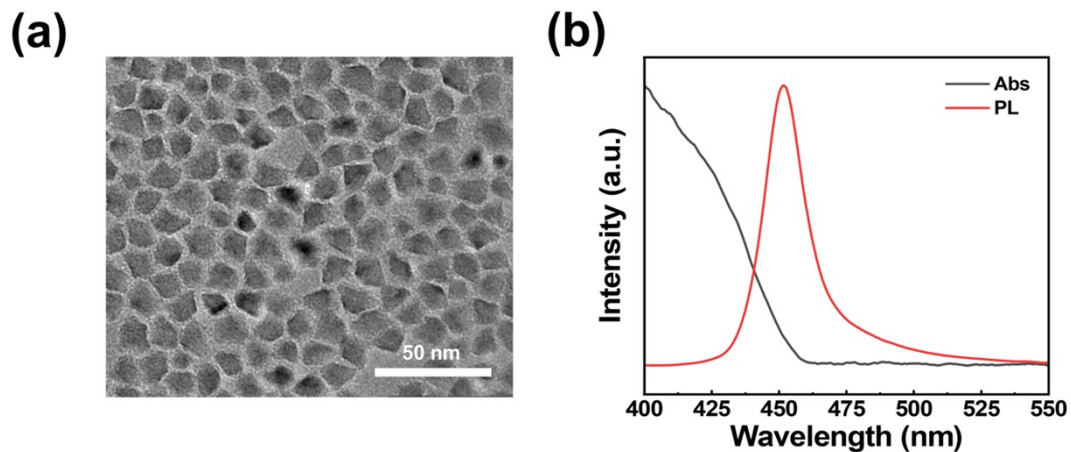


Figure S7. (a) TEM image and (b) PL & absorption spectra of blue ZnSeTe QDs.

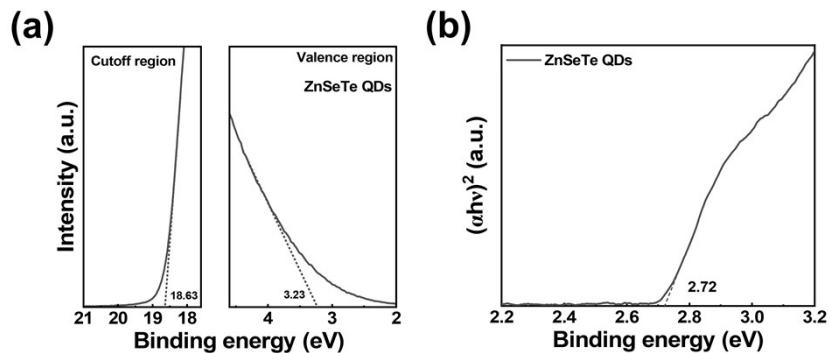


Figure S8. (a) UPS spectra of the secondary-electron cut-off region and the valence band edge region of ZnSeTe QDs film. (b) Dependence of $(\alpha h\nu)^2$ of ZnSeTe QDs film upon the incident photon energy ($h\nu$).

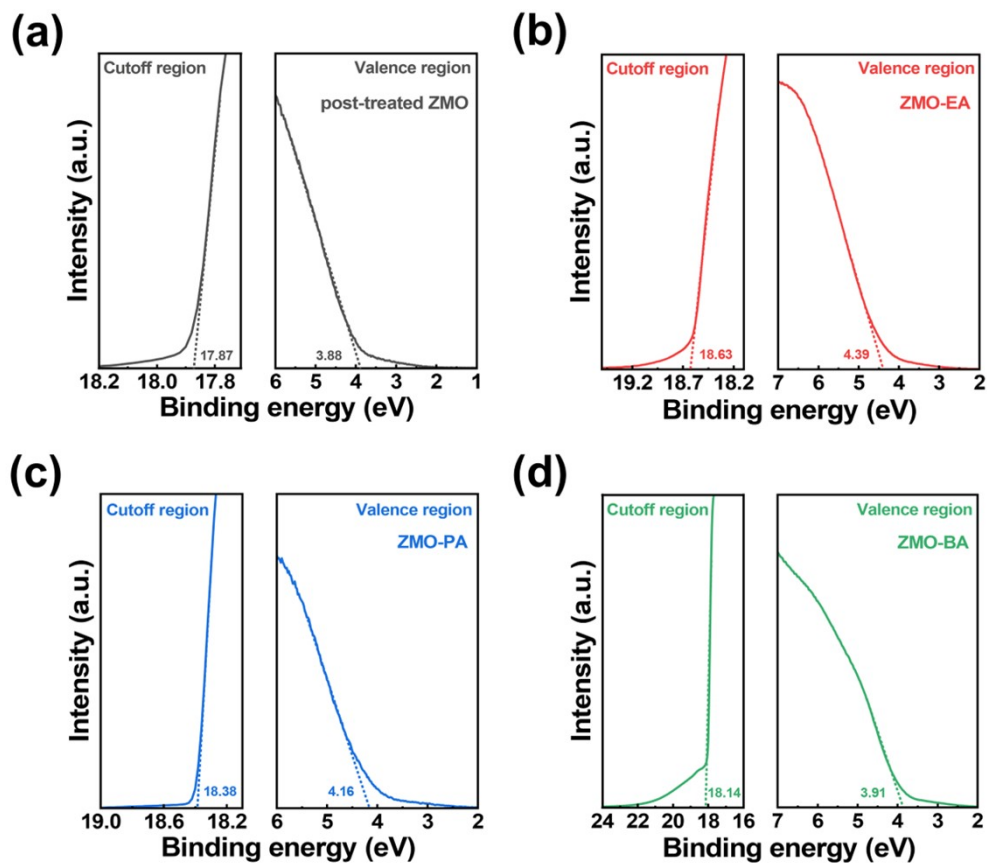


Figure S9. UPS spectra of the secondary-electron cut-off region and the valence band edge region of (a) the post-treated ZMO, (b) ZMO-EA, (c) ZMO-PA and (d) ZMO-BA films.

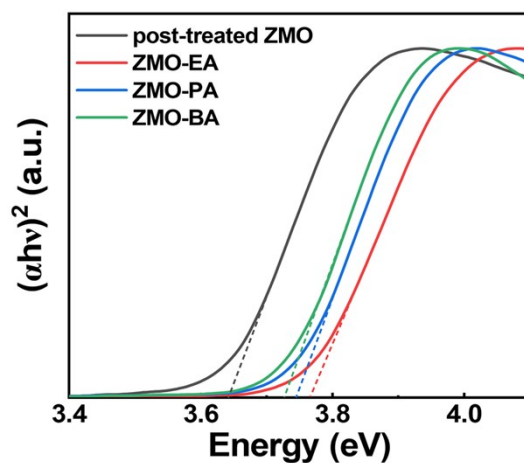


Figure S10. Dependence of $(\alpha hv)^2$ of the post-treated ZMO, ZMO-EA, ZMO-PA and ZMO-BA film upon the incident photon energy ($h\nu$).

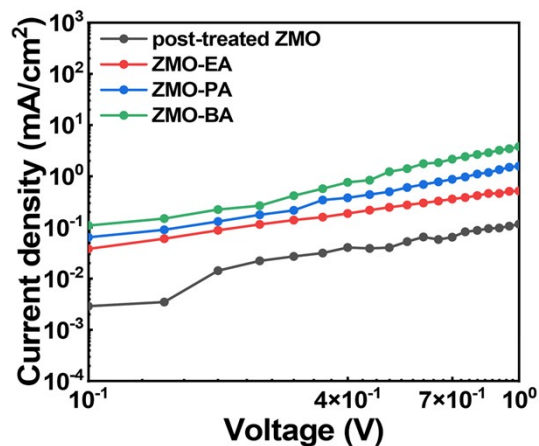


Figure S11. Comparison of current density-voltage of EODs integrated with the post-treated ZMO, ZMO-EA, ZMO-PA and ZMO-BA NPs (device structure: ITO/ZMO NPs/QDs/ETL/Al).

Table S2. The detailed performance parameters for QLEDs in Figure 3.

Devices	V _{on} (V)	Max.Luminance (cd m ⁻²)	Max.CE (cd A ⁻¹)	Max.EQE (%)	Max.PE (lm W ⁻¹)	CIE(x,y)
post-treated ZMO	3.5	6668	3.6	4.5	2.4	(0.15,0.06)
ZMO-EA	2.5	12410	4.5	7.0	3.7	
ZMO-PA	2.5	14125	5.4	8.6	4.2	
ZMO-BA	2.5	13109	4.1	6.7	3.9	

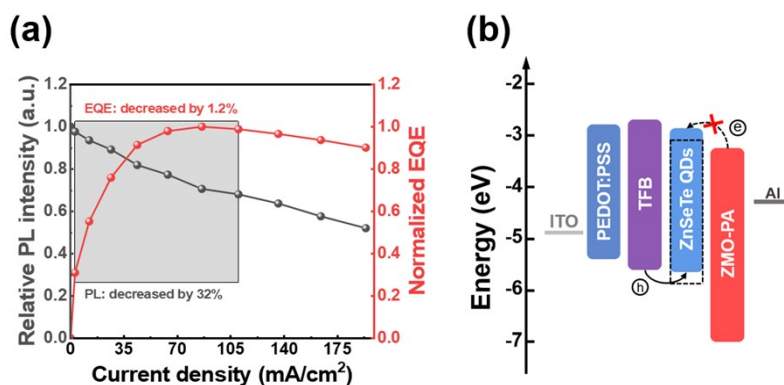


Figure S12. (a) EQE and relative PL intensity of the QLED at different current densities. (b) Schematic diagram for charge injection into negatively-charged QDs.

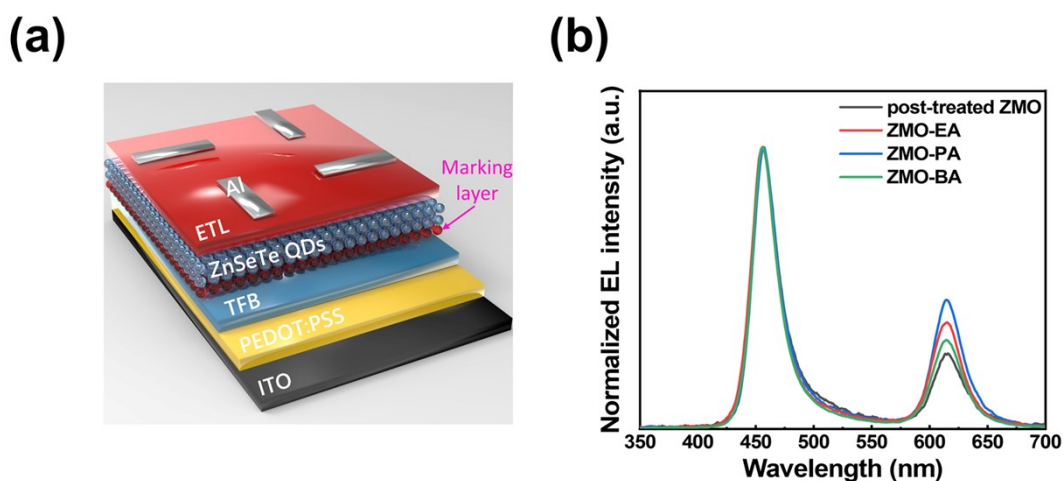


Figure S13. (a) Schematic diagram of the device structure with a red QD marking layer. (b) Normalized EL spectra from the QLEDs under 20 mA cm^{-2} .

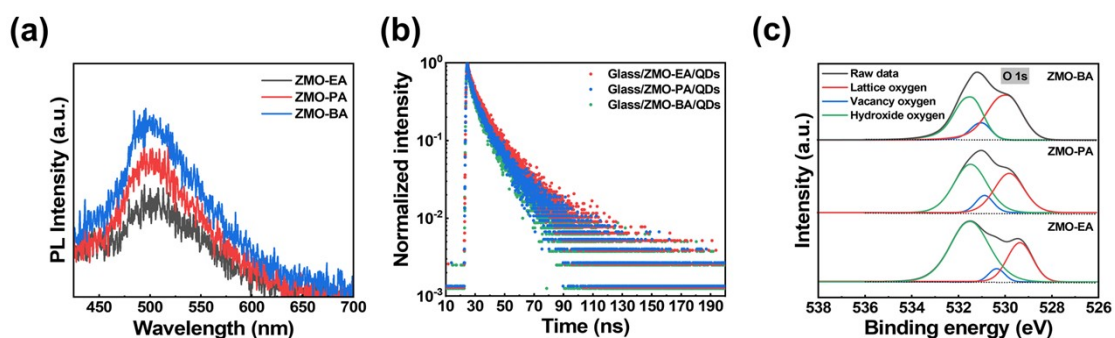


Figure S14. (a) PL spectra of ZMO-EA, ZMO-PA and ZMO-BA NPs. (b) time-resolved PL decay profile of ZnSeTe QDs films on different substrates. (c) O 1s XPS spectra of ZMO-EA, ZMO-PA and ZMO-BA films.

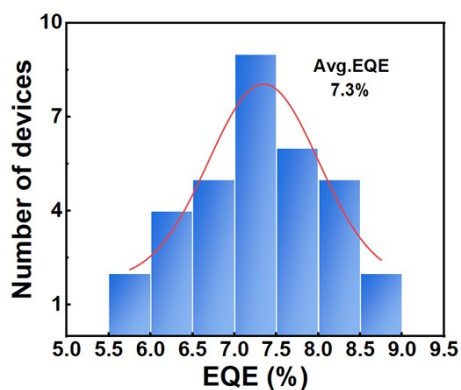


Figure S15. Histograms of peak EQEs from 33 QLEDs with MZO-PA.