Supplementary Information

Multifunctional Interfacial Molecule for High-Performance Blue Quantum Dot Light-Emitting Diodes

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Experimental section

Materials. Cadmium oxide (CdO, Macklin 99%), zinc oxide (ZnO, Macklin 99%), selenium powder (Se, Alfa 99.999%), sulfur powder (S, Aladdin 99.95%), cadmium acetate dihydrate (CdAc, Aladdin 99.99%), oleic acid (OA, Acros 90%), 1-octadecene (ODE, Macklin 90%), tri-n-octylphosphine (TOP, Aladdin 90%), molybdenum trioxide (MoO_x, Luminescence Technology Corp 99.0%), poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT: PSS, Luminescence Technology Corp), poly(9,9-dioctylfluorene-co-N-(4-(sec-butyl)phenyl)diphenylamine) (TFB, American Dye Source, Inc), 4-bis 4-4-formylphenylphosphanylbenzaldehyde (FPPB, Henan Psai Chemical Products Co. Ltd), magnesium-doped zinc oxide (ZnMgO, Poly OptoElectronics Co. Ltd), methanol (Adamas 99.9%), acetone (Enox AR), octane (J&K 99%), and ethanol (Macklin 99.7%) were purchased and used without any further treatment.

Preparation of 0.2 M cadmium oleate ($Cd(OA)_2$) precursor. First, 20 mmol CdO, 50 mL OA, and 50 mL ODE were mixed and added to a 250 mL three-neck flask. The mixture was degassed at 150 °C for 150 min. Then it was heated to 200 °C under nitrogen for another hour to dissolve the material completely, resulting in the 0.2 M $Cd(OA)_2$ precursor.

Preparation of 2 M Se-TOP and 1 M S-TOP precursors. 40 mmol Se was added to 20 mL TOP solution, and the mixed solution was vibrated for 30 min to obtain 2 M Se-TOP precursor. The 1 M S-TOP precursor was prepared using a similar approach.

Synthesis of CdZnSe/ZnSeS/ZnS core/shell quantum dots. The quantum dots were synthesized according to the reported work with some modifications.[1] In detail, 20 mmol ZnO, 20 mL OA and 30 mL OA ODE were loaded into a 100 mL three-necked flask and degassed at room temperature. The mixture was then heated to 120 °C under vacuum and subsequently to 150 °C under a nitrogen atmosphere until a clear solution was obtained. Next, 3.0 mL of 2 M Se-TOP precursor was swiftly injected into the flask at 270 °C, and the temperature was raised to 310 °C under N_2. After stabilization, 4 mL of 0.2 M $Cd(OA)_2$ precursor was quickly injected, and the reaction temperature was maintained at 310 °C for 120 min to grow CdZnSe cores. For the ZnSeS intermediate shell, a mixture of 1.0 mL 1 M S-TOP and 0.5 mL 2 M Se-TOP was injected at 310 °C, and the reaction was maintained for 60 min. Finally, the outer ZnS shell was formed by dropwise injection of a mixed solution containing 2 mL of 1 M S-TOP, and 8 mL of ODE at a rate of 1 mL min^{-1} for 10 min at 300 °C. The resulting CdZnSe/ZnSeS/ZnS QDs were cooled to room temperature and purified three times using ethanol and hexane.

Preparation of FPPB solution. The preparation processes of FPPB solutions were conducted in a glovebox (with both water and oxygen contents < 0.1 ppm). Using n-butanol as the solvent, FPPB solutions with gradient concentrations (0.05 mg mL^{-1}, 0.1 mg mL^{-1}, 0.2 mg mL^{-1}) were prepared by weighing the corresponding amount of FPPB, dissolving it in n-butanol, and stirring the mixture at room temperature for 1 hour. It should be noted that the FPPB solutions were freshly prepared and used immediately after preparation.

Fabrication of devices. Indium tin oxide (ITO) coated glass substrates (17 Ω) were cleaned using Decon 90 and deionized water in an ultrasonic cleaner for 30 min. The ITO glass substrates were then dried in an oven at 100 °C for more than 2 hours and treated with UV ozone for 15 min. After that, the PEDOT: PSS was spin-coated on the treated ITO glass substrates at 3000 rpm/min and annealed at 150 °C for 20 min in the atmosphere. Next, the glass substrates were transferred to a glove box. TFB (8 mg mL^{-1}), FPPB, QDs (20 mg mL^{-1}), and ZnMgO (25 mg mL^{-1}) were spin-coated sequentially on the glass substrates at 3000 rpm/min, and the annealing temperatures were 120 °C for 10 min, 100 °C for 10 min, 80 °C for 10 min and 60 °C for 10 min, respectively. Finally, 150 nm thick Al cathode was deposited at 2×10^{-4} Pa.

Characterization. The X-ray diffraction (XRD) curves were measured by Bruker (D8 Discover) and Rigaku Ultima IV. Ultraviolet photoelectron spectroscopy (UPS) was measured by AXIS ULTRA DLD-X-ray photoelectron spectrometer (Shimazu, Japan). The X-ray photoelectron spectroscopy (XPS) spectra were measured with a Thermo Scientific™ ESCALAB™ Xi. The Ultraviolet–visible (UV–vis) absorption was measured using a PerkinElmer Lambda 750 and 950. Temperature-dependent photoluminescence (PL) spectra were obtained from the Fluorescence Spectrometer (PL-TCSPC). PL emission was measured by F-4600. Time-resolved PL (TRPL) was measured by QUANTAURUS-Tau (Hamamatsu, Japan). The Electroluminescence (EL) spectra, Current density and luminance versus voltage ($J-V-L$) characteristics and external quantum efficiency (EQE) values were measured by an EQE measurement system (FS-000ATR-DZ9, FSTAR, Suzhou) containing a luminance meter (CS-2000, KONICA MINOLTA) and a source meter (Keithley 2400). Scanning electron microscope (SEM) characterization was conducted with a Zeiss G500. Surface morphology images of the samples were obtained using an atomic force microscopy (AFM, Cypher-S). The half-lifetime of QLEDs was measured using a commercial LED lifetime test system (64-channel ZJLS-4 type). The infrared thermography images of the device were measured by HIKMICRO K20.

Surface energy calculation procedure.

(1) Selection of probe liquids: Two probe liquids with known surface energy components were used: deionized water (polar liquid) and diiodomethane (non-polar liquid). Their surface energy parameters (at 25 °C) are summarized as follows:

Deionized water: Total surface energy (γ_l) = 72.8 mN·m^{-1}, dispersive component (γ_l^d) = 21.8 mN·m^{-1}, polar component (γ_l^p) = 51.0 mN·m^{-1};

Diiodomethane: Total surface energy (γ_l) = 50.8 mN·m^{-1}, dispersive component (γ_l^d) = 50.8 mN·m^{-1}, polar component (γ_l^p) = 0 mN·m^{-1}.

(2) Contact angle measurement: For each film sample (TFB and TFB/FPPB), three independent contact angle measurements were conducted for each probe liquid using a contact angle goniometer (OCA25). The average values of the measured contact angles (θ) were used for subsequent calculations (as listed in **Table 1** of the original manuscript: TFB: $\theta_{(water)}$=101.1°, $\theta_{(diiodomethane)}$=43.7°; TFB/FPPB: $\theta_{(water)}$=102.2°, $\theta_{(diiodomethane)}$=57.7°).

(3) Application of the Owens–Wendt equation: The Owens–Wendt method assumes that the total surface energy of a solid (γ_s) consists of dispersive (γ_s^d) and polar (

${\gamma_s}^p$) components (${\gamma_s} = {\gamma_s}^d + {\gamma_s}^p$), and the adhesion work between the solid and liquid follows the equation:

$$\gamma_1(1+\cos\theta) = 2\sqrt{{\gamma_s}^d{\gamma_l}^d} + 2\sqrt{{\gamma_s}^p{\gamma_l}^p}$$

By substituting the known surface energy components of the two probe liquids and their measured contact angles on the solid films, two simultaneous equations were established. Solving these equations yielded the dispersive (${\gamma_s}^d$) and polar (${\gamma_s}^p$) components of the solid surface energy, and the total surface energy was obtained by summing these two components.

(4) Calculation results: For the TFB film, the calculated ${\gamma_s}^d$ = 32.1 mN·m^{-1}and ${\gamma_s}^p$ = 7.2 mN·m^{-1}, leading to a total surface energy of 39.3 mN·m^{-1}. For the TFB/FPPB film, ${\gamma_s}^d$ = 28.5 mN·m^{-1} and ${\gamma_s}^p$ = 1.9 mN·m^{-1}, resulting in a total surface energy of 30.4 mN·m^{-1}.

Lifetime conversion equation. The device lifetime (T) follows a power-law dependence on the initial luminance (L), expressed as[2]:

$$L^n \cdot t = constant$$

Here, n is defined as the accelerating factor, which can be obtained by fitting the values at multiple initial luminance. In our case, the accelerating factor is about 1.81. This yields an estimated T_{100} of approximately 5.5 × 10^4 min, corresponding to 3,817 h.

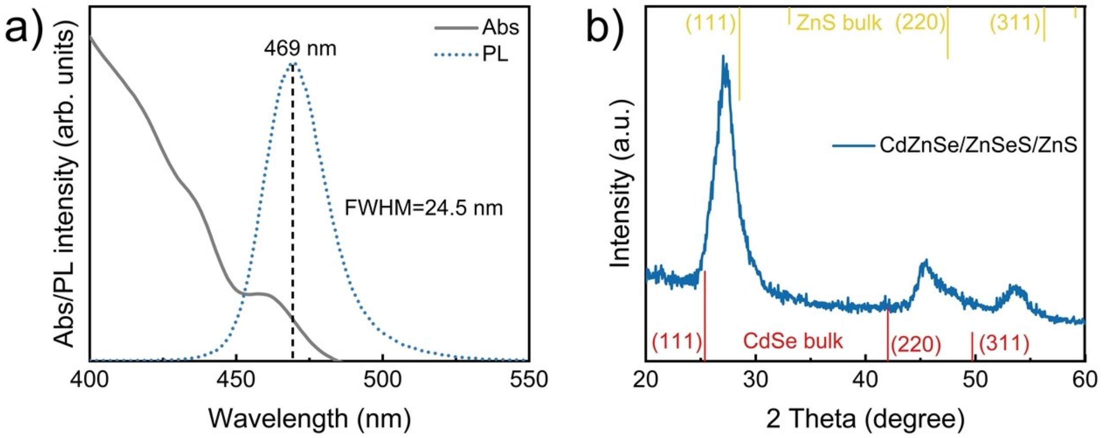

Fig. S1 a) Ultraviolet–visible (UV–vis) absorption and photoluminescence (PL) spectra of QDs; b) X-ray diffraction (XRD) patterns of blue QDs with CdZnSe/ZnSeS/ZnS core/shell.

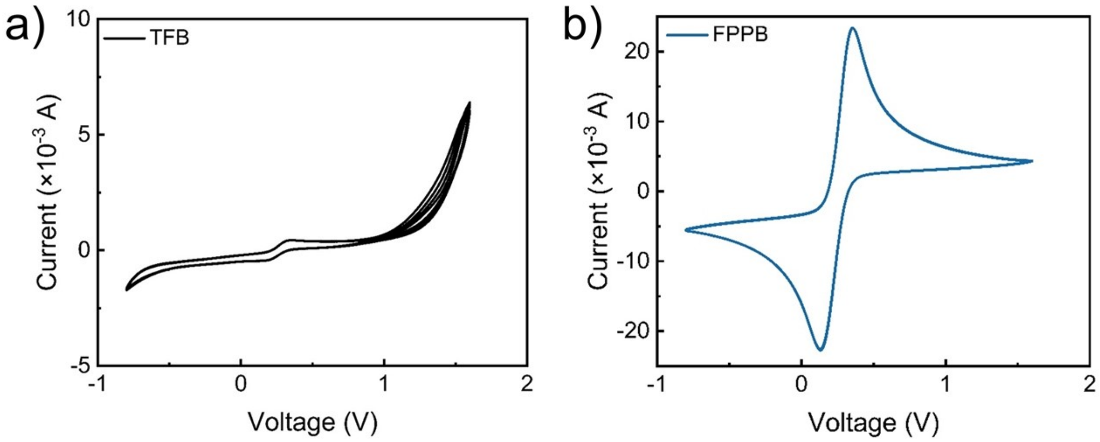

Fig. S2 Cyclic voltammetric curves of a) TFB and b) FPPB.

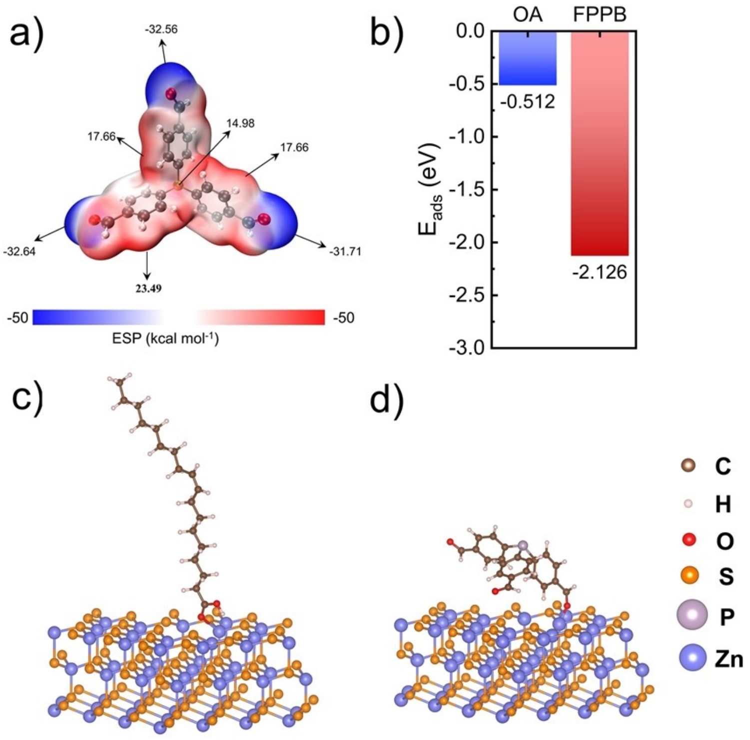

Fig. S3 a) Surface electrostatic potential (ESP) distribution map of FPPB. b) Calculated adsorption energies of FPPB and OA. c) Adsorption model of intrinsic ligand OA and d) FPPB on the surface of QDs.

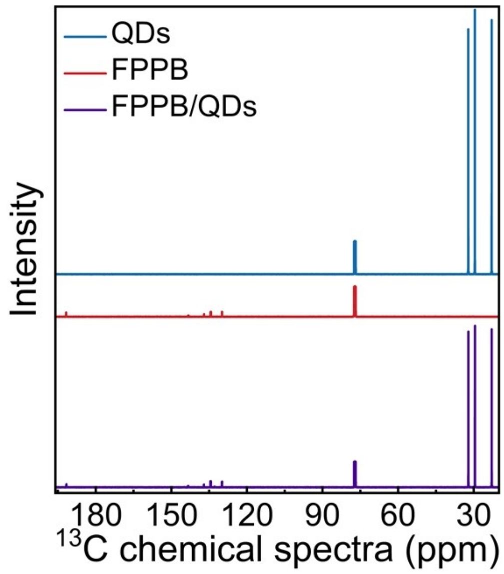

Fig. S4 ^{13}C nuclear magnetic resonance (NMR) spectra of QDs, FPPB, and FPPB/QDs mixture in the deuterated chloroform ($CDCl_3$) solution.

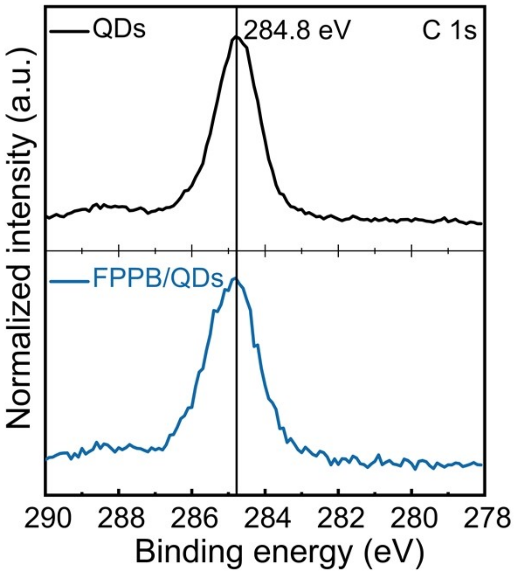

Fig. S5 X-ray photoelectron spectroscopy (XPS) spectra of C 1s core-level peaks for QD and FPPB-modified QD films.

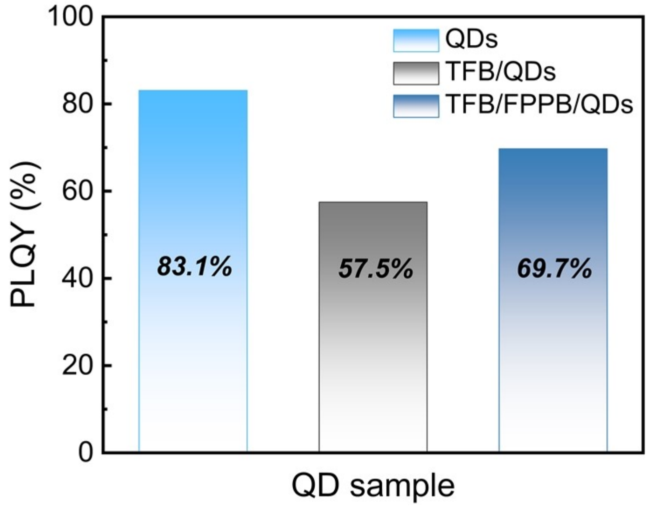

Fig. S6 Photoluminescence quantum yield (PLQY) results of QD, TFB/QD, TFB/FPPB/QD films on glass substrates.

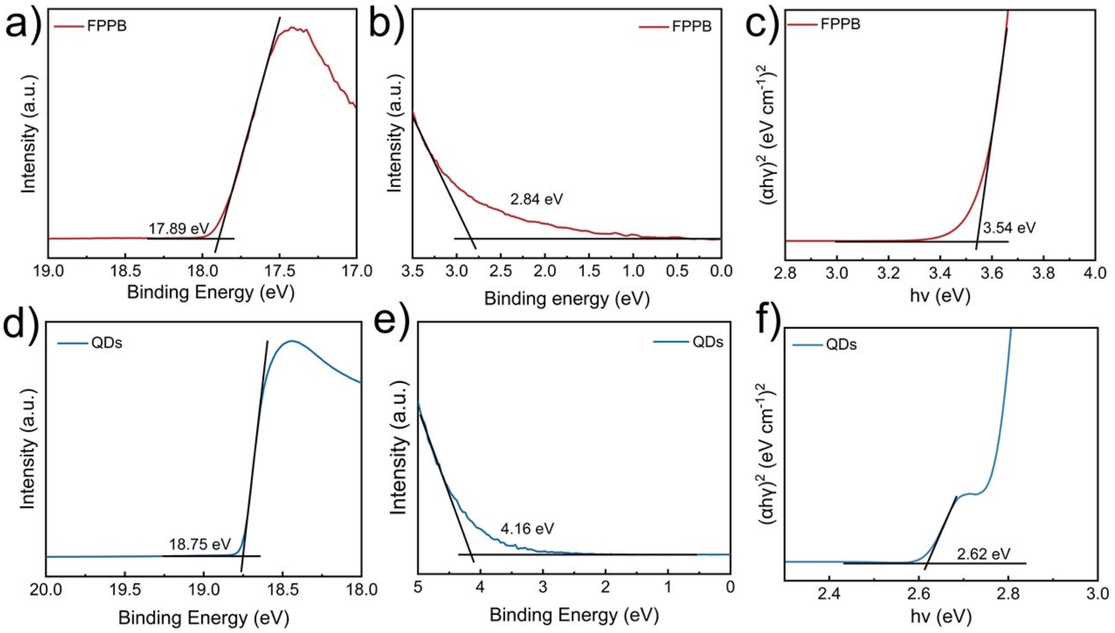

Fig. S7 Ultraviolet photoelectron spectroscopy (UPS) of a,b) FPPB and d,e) QDs. UV-vis absorption spectra of c) FPPB and f) QDs.

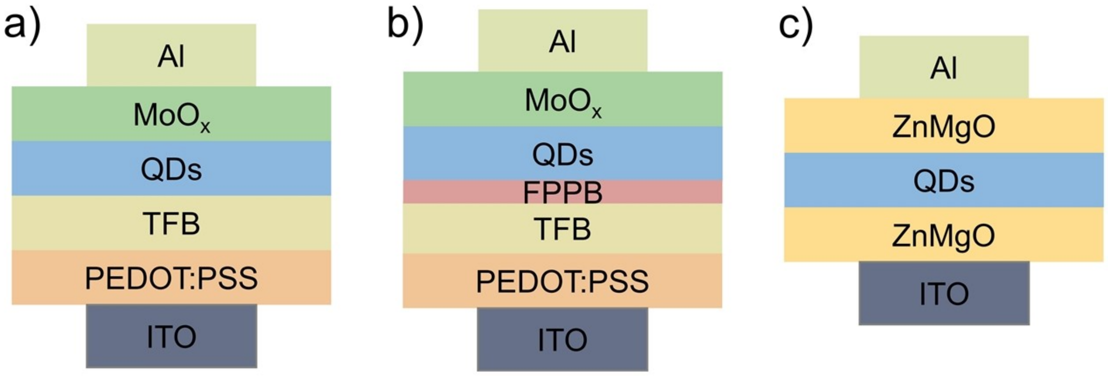

Fig. S8 Device structure of hole-only devices (HODs) a) without and b) with FPPB. c) Device structure of electron-only device (EOD).

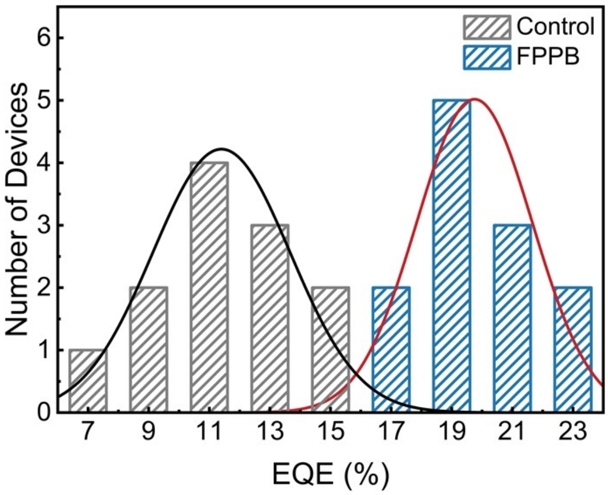

Fig. S9 Statistical histogram of EQE for control and devices with 0.1 mg mL^{-1} FPPB.

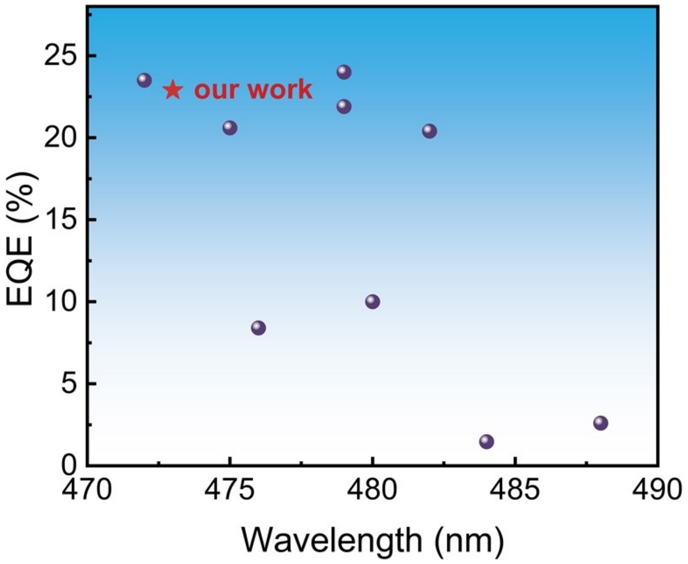

Fig. S10 Performance comparison of state-of-the-art sky blue QLEDs reported in the literature and this work.

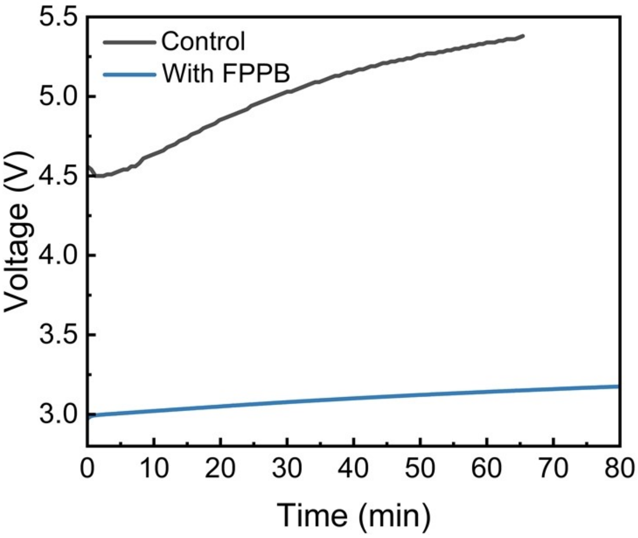

Fig. S11 Voltage variation over time during lifetime test for QLEDs without and with 0.1 mg mL^{-1} FPPB.

Table S1. Comparison of device performance among reported sky blue QLEDs.

EL peak	Device structure	EQE (%)	L_{max} (cd m^{-2})	T_{50} (h)	CIE	Year[ref]
480	ITO/PEDOT:PSS/Poly-TPD/PVK/CdSeS/ZnSeS/ZnS/ZnMgO/Al	10.0	NA	10000	NA	2020[3]
484	ITO/PEDOT:PSS/TFB/InP/ZnS/ZnS/ZnO/Al	1.47	125	NA	NA	2021[4]
475	ITO/PEDOT:PSS/PVK/ZnSe/ZnS/ZnCdSe/ZnS/ZnMgO/Al	20.6	24020	2297	NA	2022[5]
476	ITO/PEDOT:PSS/TFB/CdZnSe/ZnSeS/ZnS/CdZnS/ZnMgO/Al	8.4	12000	10.6	(0.11,0.13)	2022[1]
479	ITO/PEDOT:PSS/PF8Cz/CdZnSe/ZnS/ZnMgO/Al	21.9	NA	24000	(0.100, 0.157)	2022[6]
488	ITO/PEDOT:PSS/TFB/InP/ZnS/ZnS/ZnMgO/Al	2.6	422	NA	NA	2022[7]
482	ITO/PEDOT:PSS/TFB/ZnCdSe/ZnCdSeS/ZnS/ZnO/Al	20.4	NA	80377	(0.098,0.177)	2023[8]
472	ITO/PEDOT:PSS/PF8Cz/CdZnSe/CdZnS/ZnS/ZnMgO/Al	23.5	25510	3402	(0.12, 0.11)	2025[9]
479	ITO/PEDOT:PSS/PVK/g-CdZnSeS/ZnS/ZnMgO/Al	24.0	57000	27000	NA	2025[10]
474	**ITO/PEDOT:PSS/TFB/FPPB/CdZnSe/ZnSeS/ZnS/ZnMgO/Al**	**22.9**	**23370**	**3817**	**(0.112,0.130)**	**This work**

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