

Supporting Information

Hole-transport-layer-free CdSe/ZnS core/shell red quantum-dot light-emitting diodes sensitized by TADF polymer

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Materials

Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) (Heraeus Clevios P VP AI 4083) was used as hole injecting layer, and the solution was purchased from Xi'an Polymer Light Technology Corporation. The modified PEDOT:PSS was prepared according to the previous work by our group[18, 20]. Poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA) was used as hole transporting layer. The host materials 1,3-bis(carbazol-9-yl)benzene (mCP) and 10,10'-(4,4'-Sulfonylbis(4,1-phenylene))bis(9,9-dimethyl-9,10-dihydroacridine (DMAC-DPS) were purchased from Luminescence Technology Corporation. Bis-[3-(9,9-dimethyl-9,10-dihydroacridine)-phenyl]-sulfone (m-ACSO₂) was synthesized by our group. The

TADF polymer PCzAQC0.5 was synthesized according to literature[1]. 8-hydroxyquinolinolate (Liq), and 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB), and bis[2-(diphenylphosphino)phenyl]ether oxide (DPEPO) were used as purchased without further purification.

Device fabrication and characterization

The indium tin oxide (ITO) substrates were cleaned with acetone and ethanol consecutively for 10 minutes in the ultrasonic bath. After being dried by N₂, the substrates were treated with UV-ozone for 20 minutes to form a hydrophilic surface. The modified PEDOT:PSS layer were obtained by spin-coating from the aqueous solution on the ITO substrate at 4000 rpm for 30s. Then the substrates were transferred to a glovebox to conduct a thermal annealing at 120 °C for 10 minutes. After these steps, the quantum-dot-based emitting layers were spin-coated from the toluene solution at 1000 rpm for 30s, followed with an annealing process at 50 °C for 10 minutes. DPEPO, TmPyPB, Liq, and Al were thermally deposited in a vacuum chamber, sequentially. After encapsulation with UV epoxy, all the devices were tested at ambient air. The voltage-current-luminance characteristics and the EL spectra were simultaneously measured with PR735 SpectraScan Photometer and Keithley 2400 sourcemeter unit under ambient atmosphere at room temperature.

Table S1. Comparison of the electroluminescent performances of the devices.

EML	HTL	EQE ^a (%)	CE _{max} ^b (cd/A)	PE _{max} ^c (lm/W)	FWHM ^d (nm)	λ _{max} ^e (nm)	CIE ^f (x,y)	Color purity ^g (%)
Non-doped	HTL-Free	N.A. ⁱ	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
doped	HTL-Free	12.4	15.9	5.6	36	620	(0.65, 0.35)	95.9
doped	PTAA	6.1	7.6	3.0	45	620	(0.64, 0.35)	92.6

^aExternal quantum efficiency (EQE). ^bMaximum current efficiency (CE). ^cMaximum power efficiency (PE). ^dFull width at half maxima. ^ePeak wavelength. ^fThe Commission Internationale de l'Eclairage color coordinates recorded at 10V. ^gColor purity calculated by Equation S1. ⁱNot available since the HTL-free QLED with the neat QD film as the emitting layer wasn't working.

$$color\ purity = \frac{\sqrt{(x_s - x_i)^2 + (y_s - y_i)^2}}{\sqrt{(x_d - x_i)^2 + (y_d - y_i)^2}} \times 100\% \quad (1)$$

In Equation 1, the parameters (x_s, y_s) represent the color coordinates of the devices. (x_i, y_i) are the color coordinates of the 1931 CIE standard equal-energy point, i.e., (0.333, 0.333) used in this investigation. (x_d, y_d) refer to the color coordinates of the dominant wavelength[2,3].

Table S2. Comparison of the electroluminescent performances of the devices with different hosts.

Host	QD ^a (%)	EML ^b (nm)	EQE ^c (%)	CE ^d (cd/A)	PE ^e (lm/W)	FWHM ^f (nm)	λ_{\max} ^g (nm)	CIE ^h (x,y)	Color purity ⁱ (%)
DMAC-DPS	20	40	10.8	16.3	8.5	32	620	(0.64,0.35)	87.6
m-ACSO ₂	20	40	5.3	7.6	3.0	32	620	(0.55,0.35)	76.2
mCP	20	40	17.7	25.6	10.7	32	620	(0.65,0.33)	97.0
mCP	20	30	17.2	26.6	18.6	30	620	(0.63,0.32)	88.9
mCP	30	30	18.1	29.2	20.4	30	620	(0.64,0.33)	91.1

^aDoping concentration of QD. ^bThickness of the emitting layer. ^cExternal quantum efficiency (EQE). ^dMaximum current efficiency (CE). ^eMaximum power efficiency (PE). ^fFull width at half maxima. ^gPeak EL wavelength. ^hThe Commission Internationale de l'Eclairage coordinates recorded at 10V. ⁱColor purity calculated following Equation S1.

Table S3. The calculated photoluminescence properties of the doped QD films.

mCP:PCzAQC0.5:QD ^a	Φ (%) ^b	Φ_p/Φ_d (%) ^c	τ_p (ns) ^d	τ_d (μ s) ^e	k_r^S (10^7 s^{-1}) ^f	k_{nr}^S (10^7 s^{-1}) ^g
70:0:30	58	58/-	75.3	-	0.75	0.55
70:30:0	76	40.0/36	43.7	2.01	0.92	0.29

^aWeight ratio of the emitting layer. ^bPhotoluminescence quantum yield (PLQY). ^cPLQY ratio of the prompt and delayed components, respectively. ^dLifetime constant of the prompt fluorescence. ^eLifetime constant of the delayed fluorescence. ^fRate constant of radiative recombination decay. ^gRate constant of non-radiative recombination decay.

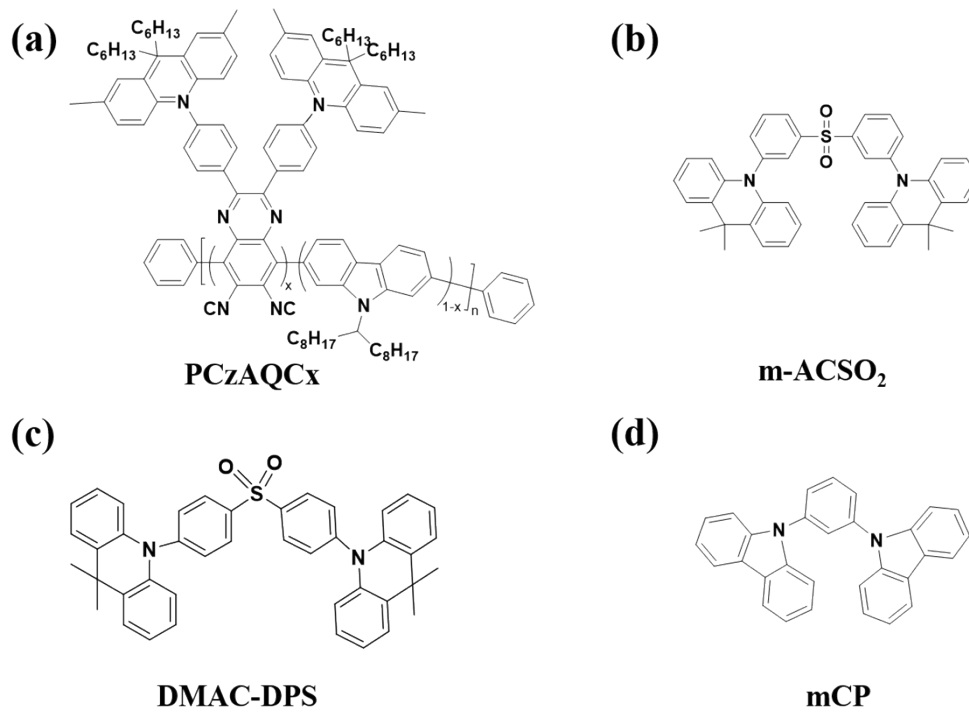


Figure S1. Chemical structures of the organic materials used in the emitting layers: (a) PCzAQC25, (b) m-ACSO₂, (c) DMAC-DPS, and (d) mCP.

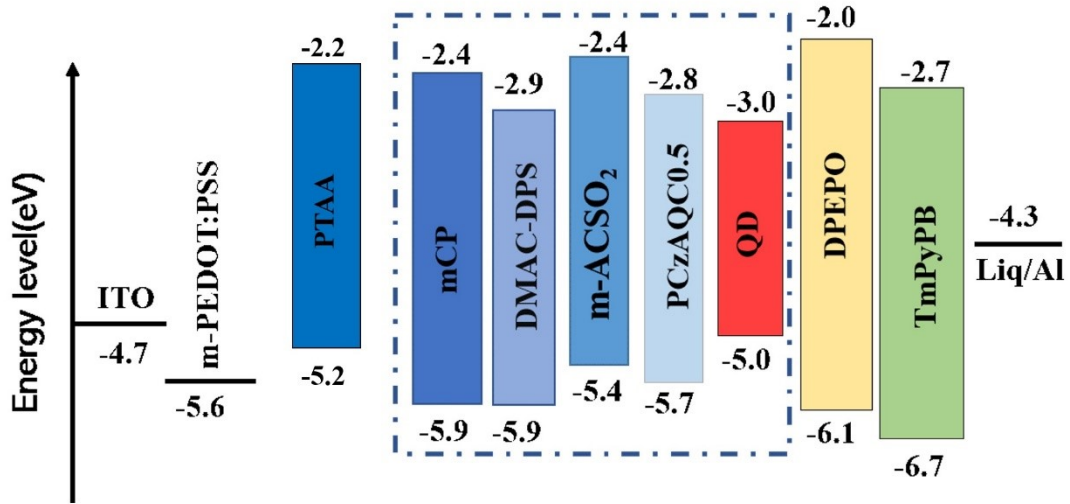


Figure S2. Schematic energy diagram of the prepared QLEDs.

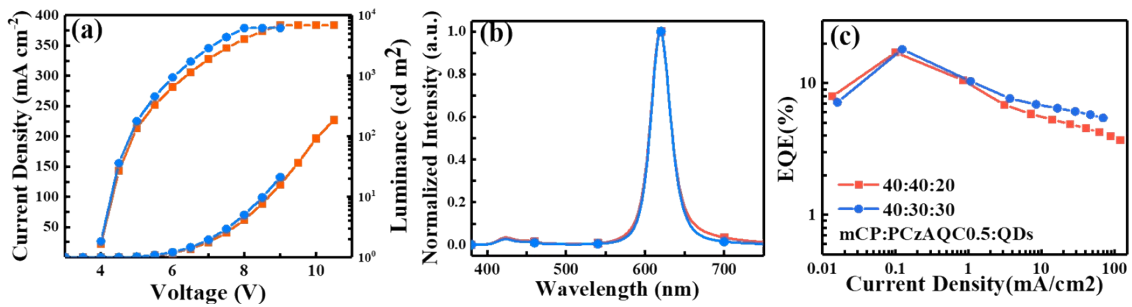


Figure S3. (a) Current density-luminance-voltage (J-L-V) characteristics, (b) current efficiency characteristics, and (c) EQE characteristics of QLEDs with different QD concentration.

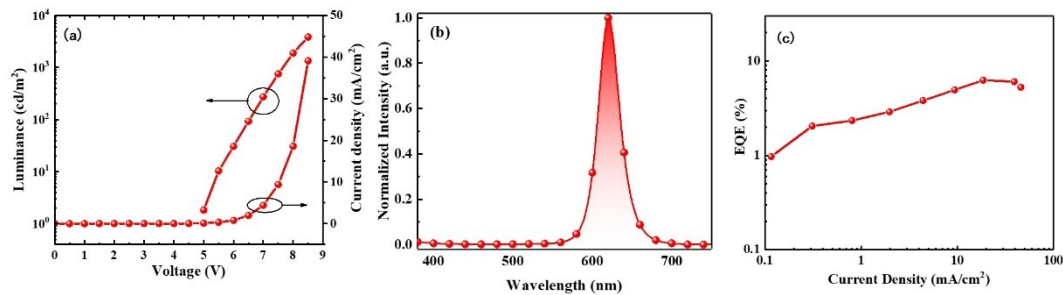


Figure S4. (a) Current density-luminance-voltage (J-L-V) characteristics, (b) current efficiency characteristics, and (c) EQE characteristics of the device with mCP:QD = 80:20 as the emitting layer.

- [1] T. Wang, K. Li, B. Yao, Y. Chen, H. Zhan, Z. Xie, G. Xie, X. Yi and Y. Cheng, *Advanced Functional Materials*, 2020, 30, 2002493.
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- [3] T. Zhao, H. Liu, M. E. Ziffer, A. Rajagopal, L. Zuo, D. S. Ginger, X. Li and A. K. Y. Jen, *ACS Energy Letters* 2018; 3(7): 1662-1669.