Supporting Information

Hybridized Local and Charge-Transfer Excited State Fluorophores Enabling Organic Light-Emitting Diodes with Record High Efficiencies Close to 20%

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1. Experimental Section

1.1 General Methods

Hydrogen (¹H) and carbon (¹³C) nuclear magnetic resonance (NMR) spectra were measured using a Bruker AvanceIII 500HD spectrometer, with deuterated dichloromethane ($CD_2Cl_2-d_2$) and chloroform ($CDCl_3$) as solvent and tetramethylsilane (TMS) as an internal reference. Photoluminescence (PL) spectra and UV-vis absorption spectra were measured using a Shimadzu RF-5301PC spectrometer and a Hitachi U-3900 spectrophotometer, respectively. Transient PL decay

characteristics were recorded on a Quantaurus-Tau system equipped with Oxford Optistat DN. PL quantum yields were recorded on a Horiba JY FL-3 spectrometer equipped with a calibrated integrating sphere. Cyclic voltammetry measurements were carried out on a electrochemical workstation VMP300 (Bio-Logic), by using tetrabutylammoniumhexafluorophosphate (TBAPF₆, 0.1 M) in dichloromethane as electrolyte, a platinum as counter electrode, and a Ag/AgCl as reference electrode (versus ferrocene F_c/F_c^+). Differential scanning calorimetry (DSC) was performed using a NETZSCH DSC 214 Polyma at a heating rate of 10 °C min⁻¹ under N₂ atmosphere. Thermogravimetric analyses (TGA) were conducted with a TA thermal analyzer (A50) under N₂ atmosphere with a heating rate of 20 °C min⁻¹. Time-dependent density functional theory (TD-DFT) calculations were performed with Gaussian 09W program package and produced by m062x/6-311g(d). The first ten singlet and triplet states were simulated using TD-m062x/6-311g(d). Spin-orbit coupling (SOC) matrix were obtained through a PySOC program based on the TD-DFT results.^[1] The models for the SOC calculations were extracted from the corresponding single crystal structures without any further optimization. Natural transition orbital (NTO) calculations were extracted by Multiwfn program.^[2a] The overlaps of the paired orbitals were calculated through Multiwfn program. Reduced density gradient (RDG) and independent gradient model (IGM) are performed by 1.9.3).^[2b] Multiwfn program and plotted by VMD (version Root-mean-square-deviation (RMSD) calculations were carried out by VMD (version 1.9.3).^[2b]

Single-crystal X-ray data were determined using an Oxford Diffraction Germini S Ultra X-ray Single Crystal Diffractometer with a (Cu) X-ray source. Single crystals were obtained through slow evaporation of saturated solution consisting of ethanol and dichloromethane mixed solvents (CCDC number of B2Cz, B2TCz, and B2PTCz is 2021355, 2021331, and 2021326 respectively). These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via <u>www.ccdc.cam.ac.uk/data_request/cif</u>.)

1.2 Devices Fabrication and Characterization

Indium tin oxide (ITO) coated glass substrates with a sheet resistance of 8 Ω sq⁻¹ were firstly ultrasonically cleaned. Then, PEDOT:PSS was spin-coated on the cleaned ITO at a speed of 2000 rpm for 60 seconds, following by thermal annealing treatment for 15 minutes at 200 °C in air. Next, on one hand, for the solution-processed OLEDs, the emissive layer was spin-coated at a speed of 5000 rpm for 40 seconds under room temperature from a chlorobenzene solution with a concentration of 30 mg/mL, wherein B2Cz, B2TCz, and B2PTCz were separately distributed into a common host of CBP, while the chlorobenzene solution was taken into a transition chamber of a nitrogen glovebox for 10 min to remove residual solvent before the spin-coating process. Finally, TPBI and cathode layers were sequentially deposited in a thermal evaporation chamber with vacuum pressure under 4×10^{-4} Pa. On the other hand, for the thermally-evaporated OLEDs, the PEDOT:PSS coated ITO substrates were transferred to the thermal evaporation chamber for deposition of organic and metals layers with vacuum pressure under 4 \times 10⁻⁴ Pa. The active area of the solution-processed and thermally-evaporated **OLEDs** 9 mm^2 . was Current-voltage-luminance characteristics and EL spectra of the unencapsulated OLEDs were measured using a Keithley 2400 source combined with a Photo Research PR735 spectrometer at room temperature, while the external quantum efficiencies (EQEs) were calculated by using a computer program based on previously reported theory.^[3]

1.3 Materials

Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)(PEDOT:PSS),m-bis(N-carbazolyl)benzene(MCP)and1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBI) were used as purchased from

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Xi'an Polymer Light Technology Co. Ltd. All the chemicals were used as received without any purification.

2. Synthesis and Characterization

B2Cz, B2TCz and B2PTCz were synthesized following Scheme S1 as below.



Scheme S1. Synthetic routes of B2Cz, B2TCz and B2PTCz.

Synthesis of 4,7-di(9H-carbazol-9-yl)benzo[c][1,2,5]thiadiazole (B2Cz)

4,7-dibromobenzo[c][1,2,5]thiadiazole (0.5 g, 1.70 mmol) and 9H-carbazole (0.85 g, 5.10 mmol), K_2CO_3 (1.41 g, 10.20 mmol), copper(I) iodide (18 mg, 0.10 mmol), o-Phenanthroline (18 mg, 0.10 mmol) and 18-Crown-6 (26 mg, 0.10 mmol) were added to dry DMF (20 mL). The mixture was stirred at 180 °C for 12 h, then cooled to room temperature. The mixture was poured to 250 mL of salt water, the crude product was collected by filtration. the product was purified by silica gel column chromatography with dichloromethane/n-hexane (v/v=1:2.5) as eluent. B2Cz was obtained as an orange powder in 63% yield (0.50 g).

Synthesis of 4,7-bis(3,6-di-tert-butyl-9H-carbazol-9-yl)benzo[c][1,2,5]thiadiazole (B2TCz)

4,7-dibromobenzo[c][1,2,5]thiadiazole (0.5 g, 1.70 mmol) and 3,6-di-tert-butyl-9H-carbazole (1.43 g, 5.10 mmol), K_2CO_3 (1.41 g, 10.20 mmol), copper(I) iodide (18 mg, 0.10 mmol), o-Phenanthroline (18 mg, 0.10 mmol) and 18-Crown-6 (26 mg, 0.10 mmol) were added to dry DMF (30 mL). The mixture was stirred at 180 °C for 12 h, then cooled to room temperature. The mixture was poured to 250 mL of salt water, the crude product was collected by filtration. the product was purified by silica gel column chromatography with dichloromethane/n-hexane (v/v=1:2.2) as eluent. B2TCz was obtained as a yellow powder in 50% yield (0.60 g).

Synthesis of 4,7-bis(4-(3,6-di-tert-butyl-9H-carbazol-9-yl)phenyl)benzo[c][1,2,5] thiadiazole (B2PTCz)

4,7-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzo[c][1,2,5]thiadiazole (0.4 g, 1.03 mmol) and 9-(4-bromophenyl)-3,6-di-tert-butyl-9H-carbazole (1.12 g, 2.58 mmol) were dissolved in THF (30 mL), then 2 M K₂CO₃ solution (2.5 mL) was added. Then catalytic amount of Pd(PPh₃)₄ was added into the mixture under an argon atmosphere. The resulting mixture was stirred at 90 °C for 12 h. After cooling down to ambient temperature, the product was purified by silica gel column chromatography with dichloromethane/n-hexane (v/v=1:2.5) as eluent. B2PTCz was obtained as a yellow powder in 51% yield (0.56 g).

3. Supplementary Figures and Tables

¹H NMR (400 MHz, CD2Cl2) δ = 8.29, 8.27, 8.16, 8.13, 8.11, 7.52, 7.50, 7.48, 7.46, 7.43, 7.41, 7.39, 7.37, 7.29, 7.27, 7.25



Figure S1. ¹H NMR spectra of BCz.

¹³C NMR (126 MHz, CDCl₃) δ = 152.66, 141.05, 129.74, 127.77, 126.11, 125.83, 124.11, 120.83, 120.60, 110.45, 110.00.



Figure S2. ¹³C NMR spectrum of BCz.



Figure S3. EI-mass spectrum of BCz.



Figure S4. High resolution mass spectrum of BCz.

¹H NMR (400 MHz, CD2Cl2) δ = 8.29, 8.27, 8.16, 8.13, 8.11, 7.52, 7.50, 7.48, 7.46, 7.43, 7.41, 7.39, 7.37, 7.29, 7.27, 7.25



Figure S5. ¹H NMR spectra of BTCz.

¹³C NMR (126 MHz, CDCl₃) δ = 152.64, 143.66, 139.50, 129.82, 127.19, 124.10, 123.72, 116.54, 109.90, 34.81, 32.02.







Figure S7. EI-mass spectrum of BTCz.



Figure S8. High resolution mass spectrum of BTCz.

¹H NMR (400 MHz, CD2Cl2) δ = 8.35, 8.33, 8.24, 8.05, 7.84, 7.82, 7.57, 1.53.



Figure S9. ¹H NMR spectra of BPTCz.

13C NMR (101 MHz, CDCl3) δ= 54.12, 143.10, 139.10, 138.48, 135.70, 132.66, 130.59, 128.30, 126.67, 123.68, 123.58, 116.31, 109.40, 34.78, 32.04.



Figure S10. ¹³C NMR spectrum of BPTCz.



Figure S11. EI-mass spectrum of BPTCz.



Figure S12. High resolution mass spectrum of BPTCz.



Figure S13. (a) TGA and (b) DSC curves of BTCz and BPTCz.



Figure S14. UV-Vis absorption and PL spectra of BCz, BTCz and BPTCz in solution state.

Commenced	Abs	$\lambda_{\rm PL}$	T _d	$T_{ m g}$	$T_{\rm m}$	τ	НОМО	LUMO	$E_{ m g}$	PLQY
Compound	(nm)	(nm)	(°C)	(°C)	(°C)	(ns)	(eV)	(eV)	(eV)	(%)
BCz	463	590	340 ^a	127 ^a	326 ^a	15.0	5.7	3.4	2.3	73.0 ^b , 81.0 ^c
BTCz	477	614	413	134	347	14.0	5.3	3.1	2.2	45.5 ^b , 48.0 ^c
BPTCz	430	564	479	-	437	6.1	5.4	3.0	2.4	90.6 ^b , 99.0 ^c

Table S1. Photophysical and thermal properties of the compounds in solid state

Abs: Absorption peak in THF solution; λ_{PL} : PL emission peak; T_d : decomposition temperature with 5% weight loss measured by thermogravimetric analysis; T_g , T_m : glass transition temperature and melting temperature measured by differential scanning calorimetry, respectively; τ : decay lifetime; HOMO: the highest occupied molecular orbital; LUMO: the lowest unoccupied molecular orbital; E_g : energy bandgap; PLQY: PL quantum yield.

^a The data are obtained from previous report.^[5]

^{b,c} PLQY of the neat compound and CBP doped compound films, respectively.



Figure S15. Cyclic voltammetry measurements of BCz, BTCz and BPTCz. The HOMO energy levels can be determined by HOMO = $-4.8-(E_{onset}-E1/2(Fc))$,^[4] where E_{onset} is the onset oxidation potential, E1/2(Fc) is the oxidation potential of ferrocene (Fc/Fc+) versus Ag/AgCl electrode. Meanwhile the, energy bandgaps (E_g) were determined from the onset of absorption spectra in dilute solutions (Figure 1b), and thereafter the lowest unoccupied molecular orbital (LUMO) levels could be estimated.



Figure S16. (a) UV-Vis absorption spectra, (b) PL spectra, and (c) linear correlation of orientation polarization (*f*) of solvent media with the Stokes shift (ν_{a} - ν_{t}) for BCz, BTCz and BPTCz in different solutions.



Figure S17. Temperature-dependent transient decay of BCz in neat film.



Figure S18. Temperature-dependent transient decay of BTCz in neat film.



Figure S19. Temperature-dependent transient decay of BPTCz in neat film.

Commenced		τ/	ns	
Compound -	77 K	100 K	200 K	300 K
BCz	21.68	21.75	21.98	18.61
BTCz	14.72	14.69	14.47	13.50
BPTCz	6.23	6.11	5.76	5.19

Table S2. Temperature dependent lifetime of the compounds in neat film.



Figure S20. HOMO and LUMO distributions of BCz, BTCz and BPTCz.



Figure S21. Natural transition orbitals of BCz for the S0, S1, T1, and T2 states.



Figure S22. Natural transition orbitals of BTCz for the S0, S1, T1, and T2 states.



Figure S23. Natural transition orbitals of BPTCz for the S0, S1, T1, and T2 states.



Figure S24. Molecular packing mode of BPTCz in single crystal.



Figure S25. Intermolecular interaction (π - π stacking) of BPTCz in single crystal.



Figure S26. Molecular packing modes of (a) BCz and (b) BTCz in single crystals.

Compound	Lm	CEm	PEm	EQEm	λel	CIE	EMI	Ref
Compound	(cd/m ²)	(cd/A)	(Im/W)	(%)	(nm)	(x , y)	ENIL	
TPP-TXO2	10480	11.1		10.5	443	(0.152,0.065)	Doping	6
	370	2.83		6.01	440	(0.153,0.051)	Doping	-
ТҒРВІ	7490	2.89		5.74	448	(0.152,0.054)	Non-doping	1
PPI-2TPA	14850	4.4	4.6	7.2	440	(0.150,0.063)		
PPI-2NPA	18105	3.98	3.88	6.33	480	(0.151,0.066)	Non-doping	8
mPAC	3995	5.61	3.48	6.76	448	(0.16,0.09)	Non-doping	9
Cz-TPB	3298	3.61	3.54	4.32	449	(0.15,0.09)	Non-doping	10
2TriPE-BPI-MCN	6129	4.72	3.17	4.60	452	(0.153,0.147)	Non-doping	11
PAIDO	~ 700	7.64	7.27	8.82	452	(0.15,0.09)	Doping	12
	3652	7.83	7.68	9.03	436	(0.155,0.102)	Doping	12
2CZPn-CNNPI	5980	6.65	5.66	5.09	454	(0.154,0.143)	Non-doping	15
PAC	16776	12.37	6.94	10.48	458	(0.15,0.13)	Non-doping	14
PyPPA	30344	9.16		8.47	464	(0.14,0.13)		15
PyPPAC	50046	8.74		7.52	468	(0.15,0.21)	Non-doping	
TPAAnPI	29900	18.09	12.35	11.47	470		Non-doping	16
DDPPPA	10 582	10.5	8.3	5.7	480	(0.15,0.10)	Non-doping	17
DDPBA	12 634	12.0	9.2	6.0	470	(0.15,0.08)	Non-doping	17
TPEPO	17007	15.86	10.90	6.62	484		Non-doping	18
ppCTPI	31070	16.32	18.46	7.16	503	(0.211,0.402)	NY 1 '	10
mpCTPI	36030	18.27	19.12	6.71	506	(0.243,0.447)	Non-doping	19
CzP-BZP		23.99	16.38	6.95	538	(0.34,0.60)	Non-doping	•
TPA-BZP		8.84	7.18	3.80	588	(0.55,0.45)	Non-doping	20
2DMeTPA-BT	64 947	25.02	20.37	7.29	544	(0.38,0.58)		
2MeTPA-BT	40 151	30.40	23.67	8.47	560	(0.44,0.55)	Doping	21
2TPA-BT	26 767	11.05	6.01	4.43	602	(0.52,0.47)		
TDA 5007	15740	12.74	10.07	3.73	532		Doping	22
IFA-DPPZ	61951	11.05	8.18	3.42	568		Non-doping	22
BPTCz	33190	65.5	37.4	19.5	546	(0.397,0.562)	Doping	Our

 Table S3. Device performance summary of reported HLCT-based OLEDs

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	23640	57.5	30.1	17.8	552	(0.424,0.547)	Non-doping	
BCz	18150	28.5	17.9	9.3	568	(0.468,0.507)	Doping	
	15590	23.4	14.7	9.1	583	(0.508,0.479)	Non-doping	
	7680	14.4	7.5	7.3	598	(0.543,0.437)	Doping	
BTCz	10960	14.8	9.3	7.4	600	(0.556,0.432)	Non-doping	
		11.1	12.0	6.6	588	(0.572,0.426)	Doping	
2TPA-BT-N-2P		10.2	10.5	5.8	608	(0.577,0.421)	Non-doping	23
TPABTPA	5760	17.7	12.4	11.1	608	(0.59,0.40)		24
ТРАВСНО	6630	4.7	2.5	5.0	640	(0.64,0.36)	Non-doping	
2TPA-BZN	3892	2.25	1.54	5.35	665			
2TPA-NZN	2814	2.1	1.83	5.06	666		Doping	25
	2644	5.15	4.27	5.75	630	(0.61,0.36)	Doping	
BTDF-TTPA	147	0.11	0.09	0.83	690	(0.70,0.30)	Non-doping	
BTDF-TtTPA	2044	2.98	2.6	4.94	642	(0.63,0.33)	Doping	26
	163	0.19	0.16	1.44	690	(0.71,0.29)	Non-doping	
NZ2TPA	6330			3.9	696	(0.70,0.30)	Non-doping	27

L_m.: max luminance, CE_m: max current efficiency, PE_m: max power efficiency, EQE_m: max external quantum efficiency, λ_{EL} : EL peak wavelength, EML: emissive layer.



Figure S27. Chemical structures of reported HLCT materials listed in Table S2.

Compound	Lm (cd/m ²)	CE _m (cd/A)	PE _m (lm/W)	EQE _m (%)	λ _{EL} (nm)	CIE (x,y)	EML	Ref	
	~2000	12.0	5.4	4.5	505	(0.10,0.55)	Non-doping		
2EHO-TPA-CNPE	~1100	10.1	4.0	5.5	475	(0.09,0.45)	Doping	28	
	~240	2.23		2.36	473	(0.20,0.15)	Non-doping		
2EHO-CNPE	~310	5.91		7.06	452	(0.15,0.09)	Doping	29	
CzPAF-TFMP	3738	5.19	3.99	4.6	442	(0.16,0.14)	Doping	30	
4	1438	2.5	1.5	6.5	428	(0.16,0.06)	Doping	31	
1	1910	23.1	14.5	7.0	532	(0.31,0.55)	Doping		
2	2476	24.9	15.6	8.1	528	(0.29,0.51)	Doping	32	

Table S4. Device performance summary of solution-processed HLCT-based OLEDs



Figure S28. Chemical structures of reported HLCT materials listed in Table S4.



Figure S29. (a) Current density–voltage–luminance curves, and (b) EQE–luminance curves of non-doped OLEDs based on BCz, BTCz and BPTCz, inset: EL spectra.



Figure S30. (a) Current density–voltage–luminance curves, and (b) EQE–luminance curves for solution-processed OLEDs with a device configuration given as: ITO/PEDOT:PSS (30 nm)/ EML (28 nm)/ TPBI (40 nm)/LiF(1 nm)/Al (100 nm), wherein EML = CBP:40wt%BCz, CBP:40wt%BTCz or CBP:40wt%BPTCz, inset: EL spectra.

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