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Supporting Information

Title: Highly-efficient (EQE >27%) Yellow OLEDs using Spiro[fluorene-9,9'-phenanthren-10'-one]- Carbazole -Based Donor-Acceptor-Donor Host Materials

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1. Experimental

All reactions and manipulations were performed under a nitrogen atmosphere unless specified, and solvents were freshly distilled according to standard procedures. The ¹H and ¹³C NMR were recorded on a Bruker AVIII HD 400. The ¹H and ¹³C NMR chemical shifts were reported on δ scale downfield from (CH₃)₄Si. The coupling constants (J) were given in hertz (Hz). Absorption spectra were recorded on a Shimadzu UV-1800 spectrophotometer, and emission spectra were obtained using a Hitachi F-4500 spectrofluorimeter. Emission spectra in solutions were measured using a spectral grade solvent and right-angle detection. Redox potentials were measured using cyclic voltammetry on a Zahner Ennium analyzer. All measurements were performed in CH₂Cl₂ solution containing 0.1 M tetrabutylammonium hexafluorophosphate a as supporting electrolyte under ambient conditions after purging the solution with N₂ for 10 min. The conventional three-electrode configuration was employed which consists of a glassy carbon working electrode, a platinum counter electrode, and an Ag/Ag⁺ reference electrode calibrated with ferrocene/ferrocenium (Fc/Fc⁺) as an internal reference. Mass spectra were recorded on a Jeol JMS-700 double-focusing mass spectrometer. Analytical thin layer chromatography (TLC) was performed on Silica gel 60 F 254 Merck. Column chromatography was performed using the silica gel from Merck (Kieselgel Si 60; 40-63 µm). Solvent THF was distilled with sodium benzophenone ketyl. Toluene, 1,2dimethoxyethane (DME), and dichloromethane were distilled with CaH₂. Other solvents and reagents were reagent grade, purchased from Acros, Alfa, Merck, Lancaster, TCI, Sigma-Aldrich, and Showa, which were directly used without purification.

Fabrication and Characterization of OLEDs:

Organic materials purchased from Shine Materials Technology were subjected to temperature-gradient sublimation in a high vacuum before use. After a cleaning procedure

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of ultrasonication of the ITO-coated glass in deionized water and organic solvents, the ITO substrate was pretreated with plasma for 5 minutes. The organic and metal layers were deposited by thermal evaporation in a vacuum chamber with a base pressure of $< 10^{-6}$ Torr. Device fabrication was completed in a single cycle without breaking the vacuum. The deposition rate of organic materials was kept at around 0.1 nm s⁻¹. The active area was defined by the shadow mask (2 × 2 mm²). Current density-voltage-luminance characterization was done using two Keysight B2901A current source-measure units equipped with a calibrated Si-photodiode. The electroluminescent spectra of the devices were recorded using an Ocean Optics spectrometer (Ocean Optics 2000).

Quantum Chemistry Computations: Geometry optimization of the dyes were accomplished by Gaussian G16W program (B3LYP/6-31G* hybrid functional). For excited states, the time-dependent density functional theory (DFT) and the B3LYP functional were used. The frontier orbital plots of the highest occupied molecular orbital (HOMO), the lowest unoccupied molecular orbital (LUMO), and the electrostatic potential (ESP) map were drawn by using GaussView 06.



Scheme S1 Synthetic conditions of MS-host: (i) CuCN, 202 °C, 24 h; (ii) $Pd(PPh_3)_2Cl_2$, 2M K₂CO_{3 (aq)}, Toluene/EtOH/H₂O (4/2/1) 90 °C, 18 h.

Compounds 1, 3, 4, MS-OC, and MS-PC were reported in our previous reports.[1, 2]

N,N-diphenyl-4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)aniline (2).

4-bromo-*N*,*N*-diphenylaniline (1.00 g, 3.08 mmol), 4,4,4',4',5,5,5',5'-octamethyl-2,2'-bi-(1,3,2-dioxaborolane) (0.94 g, 3.7 mmol), 1,1'-bis(diphenylphosphino)ferrocene palladium(II)dichloride (Pd(dppf)Cl₂) (0.11 g, 0.15 mmol), and potassium acetate (KOAc) (0.76 g, 7.7 mmol) were dissolved in Dioxane under nitrogen atmosphere. The reaction mixture was stirred for 15 hours at 100 °C. After the reaction was completed, it was cooled to room temperature, poured into excess brine and extracted with dichloromethane (CH₂Cl₂). The organic portions were dried with anhydrous Na₂SO₄, filtered, and concentrated. The crude reaction mixture was purified via column chromatography (dichloromethane/hexane = 1/2) and concentrated to afford **2** (0.96 g, 84%) as a brown solid. ¹H NMR (400 MHz, CDCl₃): 7.66 (d, 2H, J = 8.4 Hz), 7.26 (t, 5H, J = 8.4), 7.11–7.02 (m, 8H), 1.33 (s, 12H).

10'-oxo-10'H-spiro[fluorene-9,9'-phenanthrene]-2,7-dicarbonitrile (MS-CN).

2,7-dibromo-10'H-spiro[fluorene-9,9'-phenanthren]-10'-one (1.00 g, 1.99 mmol) and copper(I) cyanide (CuCN) (0.54 g, 3.0 mmol) were added to an oven-dried round-bottomed flask, followed by three vacuum/nitrogen cycles. Dry 1-methyl-2-pyrrolidone (NMP) (3.37 mL) were purged under nitrogen for 20 min and added to the reaction mixture. The reaction was refluxed for 24 hours. After the reaction was completed, it was cooled to room temperature, copper was removed via a suction filter, NMP was removed via a reduced pressure distillation, poured into excess water and extracted with ethyl acetate. The organic portions were dried with anhydrous Na₂SO₄, filtered, and concentrated. The crude reaction mixture was purified via column chromatography (n-hexane/ethyl acetate = 4/1) and concentrated to afford **MS-CN** (0.50 g, 63%) as a white solid. ¹H NMR (400 MHz, CDCl₃): 8.23 (d, 1H, J = 8.0 Hz), 8.16 (d, 1H, J = 7.8 Hz), 7.98–7.86 (m, 4H), 7.75 (dd, 2H, J = 7.9 Hz, 1.3 Hz), 7.54–7.45 (m, 2H), 7.36 (s, 2H), 7.17–7.13 (m, 1H), 6.49 (dd, 1H, J = 7.8 Hz, 1.0 Hz);¹³C NMR (100 MHz, CDCl₃): 195.01, 148.69, 144.21, 137.81, 136.18, 136.09, 133.05, 130.70, 129.85, 129.43, 129.21, 129.14, 128.97, 128.83, 127.71, 125.09, 123.86, 122.32, 118.57, 112.95, 68.71. MS (HR-FAB): m/z calcd. for C₂₈H₁₅N₂O 394.1106, found 395.1187 [M+H]⁺.

2,7-bis(4-(diphenylamino)phenyl)-10'H-spiro[fluorene-9,9'-phenanthren]-10'-one (MS-TPA).

2,7-dibromo-10'H-spiro[fluorene-9,9'-phenanthren]-10'-one (1.00 g, 1.99 mmol), *N*,*N*-diphenyl-4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)aniline (1.62 g, 4.38 mmol), potassium carbonate (K₂CO₃) (0.83 g, 6.0 mmol), and bis(triphenylphosphine)palladium(II) dichloride (Pd(PPh₃)₂Cl₂) (40 mg, 0.06 mmol) were dissolved in a mixed solution which contained toluene (20 mL), THF (10 mL), and distilled water (5 mL) under nitrogen atmosphere. The reaction mixture was stirred for 15 hours at 130 °C. After the reaction was completed, it was cooled to room temperature, poured into excess water and extracted with dichloromethane (CH₂Cl₂). The organic portions were dried with anhydrous Na₂SO₄, filtered, and concentrated. The crude reaction mixture was purified via column chromatography (dichloromethane/hexane = 1/1) and concentrated to afford **MS-TPA** (1.14 g, 69%) as a yellow solid. ¹H NMR (400 MHz, CDCl₃): 8.20 (d, 1H, *J* = 8.1 Hz), 8.10 (d, 1H, *J* = 7.9 Hz), 8.03 (d, 1H, *J* = 7.2 Hz), 7.85–7.77 (m, 3H), 7.62 (dd, 2H, *J* = 8.0 Hz, 1.2 Hz), 7.46 (t, 2H, *J* = 7.5 Hz),

7.36 (t, 1H, J = 7.3 Hz), 7.31 (d, 4H, J = 8.6 Hz), 7.23 (d, 11H, J = 7.4 Hz), 7.09–7.00 (m, 16H), 6.72 (d, 1H, J = 7.7Hz); ¹³C NMR (100 MHz, CDCl₃): 197.11, 148.27, 147.60, 147.17, 140.30, 140.14, 139.30, 138.16, 135.03, 134.74, 130.53, 129.99, 129.35, 139.26, 128.68, 128.49, 128.39, 128.07, 127.66, 126.96, 124.42, 124.13, 123.74, 123.33, 122.95, 122.83, 120.82, 68.64. MS (HR-FAB): m/z calcd. for C₆₂H₄₂N₂O 830.3297, found 830.3288 [M]⁺.

2. ¹H and ¹³C NMR spectra



Fig. S1 ¹H NMR spectra of 2.



Fig. S2 ¹H NMR (upper) and ¹³C NMR (lower) spectra of MS-CN.



Fig. S3 ¹H NMR (upper) and ¹³C NMR (lower) spectra of MS-TPA.

3. Thermogravimetric analysis (TGA) curves



Fig. S4 Thermogravimetric analysis (TGA) curves of MSs.

4. Differential scanning calorimetry (DSC) curves



Fig. S5 Differential scanning calorimetry (DSC) curves of MSs.

5. CV voltammograms



Fig. S6 Cyclic voltammetry diagrams of MSs in CH₂Cl₂.

	MS-CN	MS-TPA	MS-OC	MS-PC
LUMO+1				
[eV]	-2.48	-1.56	-1.86	-1.90
LUMO	•			
[eV]	-2.70	-2.08	-2.06	-2.11
НОМО	••• <u>•</u> • • ••••••••••••••••••••••••••••••	BING CONT	zing	NEW CONTRACTOR
[eV]	-6.84	-5.07	-5.34	-5.14
HOMO-1	-4000			
[eV]	-6.93	-5.23	-5.53	-5.49

6. Theoretical calculation

Fig. S7 HOMO/LUMO levels of MSs.

	MS-CN	MS-TPA	MS-OC	MS-PC
S ₀ (eV)	-6.8354	-5.0700	-5.34	-5.14
S ₁ (eV)	-2.6981	-2.0797	-2.06	-2.11
T ₁ (eV)	-3.020	-3.010	-3.016	-3.015
S ₀ -S ₁ Energy Difference (eV)	4.1373	2.9903	3.28	3.03
$\Delta E_{ST} (eV)$	0.3219	0.9303	0.956	0.905
S₀-T₁ Energy Difference (eV)	3.8154	2.0600	2.324	2.125
Dipole moment (debye)	2.7475	2.9914	5.6686	2.4960

Table S1 Theoretically calculated energy level of first excited triplet state (T₁), HOMO-LUMO energy gap (E_g), and singlet-triplet energy difference (ΔE_{ST}) of **MS** fluorescent emitters



Fig. S8 Theoretically optimized structures, dihedral angles, and ESP map of MS-OLED hosts.

7. PL spectra of the thin film samples



Fig. S9 PL spectra of the thin film samples consisting of CN-T2T and the synthesized MSs.

8. EL characteristics of OLED device A and device C

$I(t) = A + B_1 \exp(-t/\tau_1) + B_2 \exp(-t/\tau_2)^{a}$										
Sample	\mathbf{B}_1	$\tau_1(ns)$	B ₂	$\tau_2(ns)$						
MS-CN : CN-T2T (1:1)	9820	1.68	277	17.4						
MS-TPA : CN-T2T (1:1)	3540	5.07	589	25.6						
MS-PC : CN-T2T (1:1)	3650	3.09	412	14.4						
MS-OC : CN-T2T (1:1)	3906	4.81	1577	25.6						

 Table S2 Photophysical characteristics of the mixed films.

^a Measured under an ambient atmosphere, and the decay components were fitted with two exponential decay models.



Fig. S10 Current density-voltage (J-V) characteristics of the hole-only and electron-only devices.



Fig. S11 (a) Normalized EL spectra at a luminance of 10^3 cd m⁻²; (b) current density-voltageluminance (*J*-*V*-*L*) characteristics; (c) external quantum efficiency vs luminance; (e) luminance/power efficiency vs luminance for devices BR1, BR2, BR3, and BR4.



Fig. S12 (a) Normalized EL spectra at a luminance of 10^3 cd m⁻²; (b) current density-voltageluminance (*J*-*V*-*L*) characteristics; (c) external quantum efficiency vs luminance; (e) luminance/power efficiency vs luminance for devices BG1 and BG4.

Device		А	.G		А	Y		AR				
No.		1	4	1	2	3	4	1	2	3	4	
Host		MS-CN	MS-OC	MS-CN	MS-TPA	MS-PC	MS-OC	MS-CN	MS-TPA	MS-PC	MS-OC	
Emitter		Ir(p	py) ₃		РО	-01			Ir(piq) ₂ acac		
External	[a]	0.7	22.0	9.5	12.0	22.8	22.5	8.5	9.8	14.4	14.9	
efficiency [%]	[b]	0.6	18.7	5.1	11.4	22.1	21.5	3.8	8.8	13.6	13.4	
Luminance	[a]	1.9	77.7	27.9	36.7	66.2	67.1	5.3	6.6	8.1	8.7	
$[cd A^{-1}]$	[b]	1.7	66.4	15.1	35.0	64.0	64.1	2.4	5.9	7.6	7.9	
Power	[a]	1.8	98.5	30.3	52.4	86.0	81.1	5.6	9.5	10.5	10.5	
[lm W ⁻¹]	[b]	1.0	63.5	8.4	47.1	70.9	62.0	1.2	6.7	7.9	6.5	
$V_{\rm on}[V]$	[c]	3.0	2.4	3.3	2.0	2.4	2.6	3.5	2.1	2.4	2.8	
$J_{1/2}$ [mA cm ⁻²]	[d]	754.5	75.6	1.0	46.2	78.3	90.5	2.5	90.1	215.8	108.8	
Max luminan [cd m ⁻²] [V	ice]	36152 [10.8]	8211 [11.4]	112382 [11.8]	29428 [11.8]	41953 [11.0]	144592 [11.2]	159521 [10.6]	5390 [12.2]	9604 [9.4]	27260 [11.0]	
CIE1931 coordinates	[b]	(0.45, 0.53)	(0.32, 0.63)	(0.50, 0.49)	(0.50, 0.49)	(0.51, 0.48)	(0.50, 0.49)	(0.68, 0.32)	(0.68, 0.31)	(0.69, 0.31)	(0.69, 0.31)	

Table S3 EL characteristics of the device A series with different hosts and emitters.

[a] Maximum efficiency; [b] measured at 10² cd m⁻²; [c] turn-on voltage measured at 1 cd m⁻²; [d] the current density at the half of peak external quantum efficiency.

Device		C	G	CY CR						R	
No.		1	4	1	2	3	4	1	2	3	4
Host		MS-CN	MS-OC	MS-CN	MS-TPA	MS-PC	MS-OC	MS-CN	MS-TPA	MS-PC	MS-OC
Emitter		Ir(p	py) ₃		РО	-01			Ir(piq) ₂ acac	
External	[a]	4.6	22.1	9.5	12.0	22.8	22.5	12.2	18.4	18.3	20.1
efficiency [%]	[b]	3.8	21.9	5.1	11.4	22.1	21.5	7.3	18.4	17.5	18.5
Luminance	[a]	14.4	77.9	27.9	36.7	66.2	67.1	7.2	10.8	10.4	11.5
$[cd A^{-1}]$	[b]	11.9	77.0	15.1	35.0	64.0	64.1	4.3	10.7	9.9	10.6
Power	[a]	10.2	107.0	30.3	52.4	86.0	81.1	8.7	15.2	14.8	16.4
$[\text{Im W}^{-1}]$	[b]	9.7	95.8	8.4	47.1	70.9	62.0	2.9	11.6	11.8	11.9
$V_{\rm on}[V]$	[c]	2.6	2.1	3.3	2.0	2.4	2.6	3.0	2.0	2.2	2.2
$J_{1/2}$ [mA cm ⁻²]	[d]	580	95.3	1.0	46.2	78.3	90.5	7.7	163.8	131.5	82.8
Max luminan [cd m ⁻²] [V	ice]	36152 [10.8]	54657 [11.0]	135477 [9.6]	29428 [11.8]	41953 [11.0]	144592 [11.2]	159521 [10.6]	10600 [11.4]	28016 [11.4]	27849 [11.2]
CIE1931 coordinates	[b]	(0.41, 0.56)	(0.34, 0.61)	(0.50, 0.49)	(0.50, 0.49)	(0.51, 0.48)	(0.50, 0.49)	(0.69, 0.31)	(0.69, 0.31)	(0.69, 0.31)	(0.69, 0.31)

Table S4 EL characteristics of the device C series with different hosts and emitters.

[a] Maximum efficiency; [b] measured at 10² cd m⁻²; [c] turn-on voltage measured at 1 cd m⁻²; [d] the current density at the half of peak external quantum efficiency.

9. EL characteristics of PO-01-based devices reported in the literature

Year	Host	EQE _{peak} (%)	CE _{peak} (cd/A)	PE _{peak} (lm/W)	Max. Luminance (cd/m ²)	V _{on} ^a (V)	Ref
2005	CBP	—	43.56	30.41	10,999	4.5	[3]
2013	mCP	10.2	_	-	_	_	[4]
2014	ACDCN	25.7	_	_	_	-	[5]
2015	ACRFLCN	26.9	_	70.4	—	-	[6]
2013	BBPI	16.3	49.1	52.0	—	2.6	[7]
2016	BBPI	12.9	36.8	38.6	_	2.6	[8]
2017	mCP	17.1	57.3	_	_	_	[9]
2017	m-DCz-S	26.1	80.9	79.4	61,000	3.0	[10]
2018	TPAPOPPI	18.9	57.3	60.0	—	2.5	[11]
2019	TP-PPI	26.1	80.6	82.4	—	2.6	[12]
	tcaz-tcaz	11.0	36.0	35.6	2,956	3.3	[13]
2020	CBP	13.1	42.1	29.0	16,170	3.2	[14]
2020	CF-2-BzF	25.3	77.2	59.8	—	-	[15]
	TCTA: DCb-BPP (55:45)	24.5	_	74.0	174,000	2.56	[16]
	H2	10.9	35.8	32.2	23,190	3.5	[17]
2021	IPNCz	25.6	82.4	85.9	44,866	2.6	[18]
	Trz-PhCz	27.3	80.0	98.9	_	3.0 ^b	[19]
2022	MS-OC : CN-T2T (1:1)	27.1	80.0	113.0	142,464	2.1	This work

 Table S5 EL characteristics of PO-01-based devices reported in the literature.

^{a.} Turn-on measured at 1 cd m⁻². ^{b.} Operation voltage measured at 100 cd m⁻².

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