

Supporting Information (SI)

**Efficient Solution-Processed Narrowband Green-Emitting
Organic Light Emitting Diodes Sensitized by Thermally
Activated Delayed Fluorescence Polymer**

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1. General information

PEDOT:PSS (Clevios Al4083) was purchased from Heraeus Precious Metals GmbH Co. KG. PFI (Nafion, ≥ 1.00 meq g^{-1} exchange capacity) was purchased from Alfa-Aesar. Poly(TMTPA-DCB) were synthesized according to previously reported methods¹. m-Cz-BNCz (3-(3,6-di-*tert*-butyl-9*H*-carbazol-9-yl)phenyl)-2,6-bis(3,6-di-*tert*-butyl-9*H*carbazol-9-yl) boron), mCP (1,3-bis(carbazol-9-yl)benzene), TSPO1 (diphenyl-4-triphenylsilylphenyl-phosphine oxide), TmPyPB (Tri(m-pyridin-3-ylphenyl)benzene, 1,3,5-Tris(3-pyridyl-3-phenyl)benzene), and LiF were bought from Luminescence Technology Corp.

2. Measurements and characterization

The UV-Vis absorption spectra were recorded by using a PerkinElmer Lambda 35 UV-vis spectrometer. The photoluminescence (PL) spectra, PL quantum yields (PLQYs), and low-temperature phosphorescence spectra were measured by using a Horiba Instruments Inc. FL3C-111 fluorescence spectrometer. And the film samples used for PLQYs were prepared by spin-coating the mCP, poly(TMTPA-DCB), and m-Cz-BNCz mixed solutions on the quartz substrate. The low-temperature phosphorescence spectra were obtained at 77 K under vacuum with a delay time of 0.01 ms to remove the fluorescent component. Transient PL spectra decay characteristics were recorded using a DeltaFlex™ Modular Fluorescence Lifetime System (Horiba Scientific) with an excitation wavelength of 300 nm in N₂. The impedance spectroscopy measurement was carried out in a PARSTAT 3000A potentiostat in a 1.0 MHz - 1.0 Hz frequency range with an applied voltage of 4.0 V and AC signal of 0.1 V. Cyclic voltammetry (CV) measurements were recorded on an EG&G 283 potentiostat/galvanostat system from Princeton Applied Research.

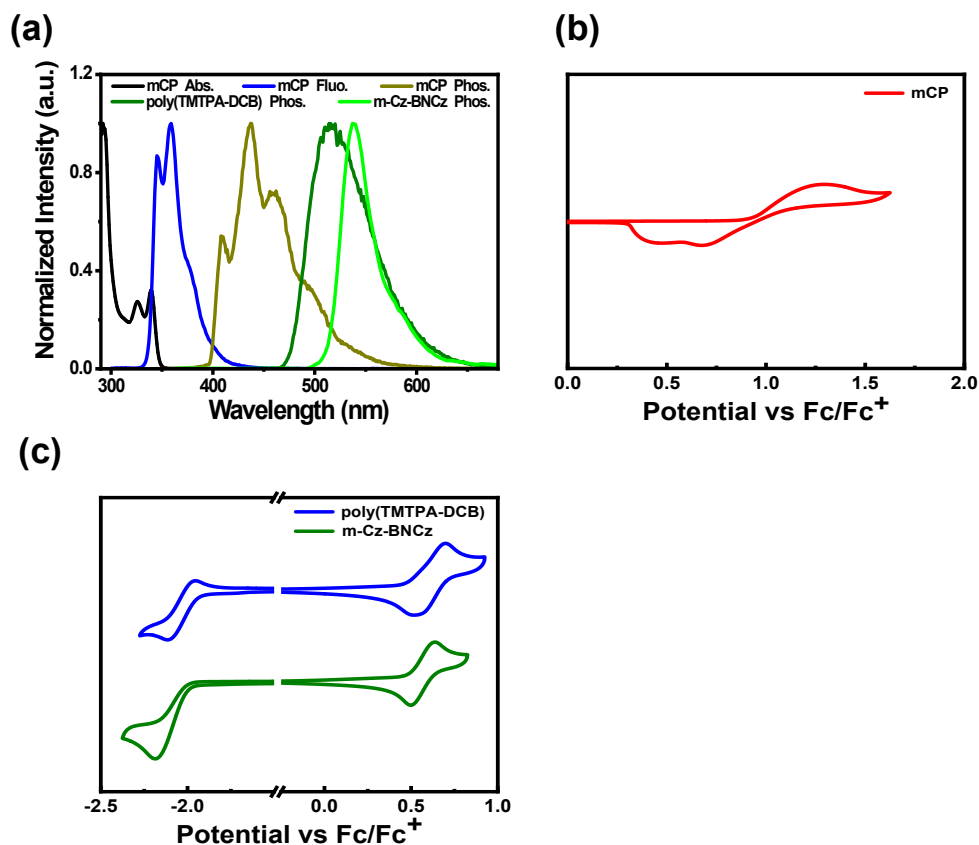


Fig. S1 (a) Normalized UV-vis absorption, PL spectra of mCP; phosphorescence spectra of mCP, poly(TMTPA-DCB) and m-Cz-BNCz; (b) CV curves of mCP; (c) CV curves of poly(TMTPA-DCB) and m-Cz-BNCz.

Table S1. Summary of the photophysical properties of mCP, poly(TMTPA-DCB) and m-Cz-BNCz.

Compound	λ_{abs}^a (nm)	λ_{PL}^b (nm)	FWHM ^c (nm)	E_{S1}^d [eV]	E_{T1}^e [eV]	ΔE_{ST}^f [eV]	E_g^g [eV]	HOMO ^h [eV]	LUMO ⁱ [eV]
mCP	340	344	-	3.67	3.13	0.54	3.54	-5.66	-2.12
poly(TMTPA- DCB)	390	509	84	2.73	2.63	0.10	2.67	-5.23	-2.85
m-Cz-BNCz	484	516	39	2.54	2.42	0.12	2.42	-5.27	-2.77

^a The peak wavelength of the lowest-energy absorption band; ^b The peak wavelength of the PL spectrum (1×10^{-5} M toluene solution, 298 K); ^c Full width at half maximum of the PL spectrum; ^d Singlet energy estimated from the onset of the fluorescence spectrum (1×10^{-5} M toluene solution, 298 K); ^e Triplet energy estimated from the onset of the phosphorescence spectrum (1×10^{-5} M toluene solution, 77 K); ^f $\Delta E_{\text{ST}} = E_{\text{S1}} - E_{\text{T1}}$; ^g Optical band gap estimated from the absorption edges of UV-vis spectrum; ^{h, i} Determined from cyclic voltammetry.

3. Calculation related to FET and DET processes

Firstly, according to the previous report,² we calculated the molar extinction coefficient of m-Cz-BNCz by UV-Vis absorption spectroscopy using the Lambert-Bier law.

$$A = \varepsilon CL\#(S - 1)$$

In equation (S-1), A is the absorbance at the peak position of the first exciton absorption peak, C is the molar concentration of m-Cz-BNCz (mol L⁻¹), L is the optical path length of about 1 cm, ε is the molar extinction coefficient (L mol⁻¹·cm⁻¹).

Then secondly, the Förster energy transfer radius (R_{FET}) is calculated by the expression as:

$$R_{FET}^6 = \frac{9000(\ln 10)k^2\phi_{PL}}{128\pi^5 N_A n^4} \int_0^{\infty} Fh(\lambda)\varepsilon g(\lambda)\lambda^4 d\lambda\#(S - 2)$$

In equation (S-2), Φ_{PL} is PLQY of poly(TMTPA-DCB), κ^2 is the transition dipole orientation factor with the value of 2/3, n is the refractive index with the value of 1.7,

N_A is the Avogadro's number, $\int_0^{\infty} Fh(\lambda)\varepsilon g(\lambda)\lambda^4 d\lambda$ is the overlap integral between the PL of poly(TMTPA-DCB) and the absorption of m-Cz-BNCz (Fig. 2a). In the overlap integral, $Fh(\lambda)$ stands for the normalized donor emission spectrum, and $\varepsilon g(\lambda)$ is the molar absorption coefficient of the acceptor^{2,3}. According to equation (S-2), the R_{FET} calculation results in 4.15 nm for the hybrid of mCP: 10 wt.% poly(TMTPA-DCB): 2.0 wt.% m-Cz-BNCz.

The next, Förster energy transfer rate (k_{FET}) values were calculated by following the expression as (S-3~S-6).

$$k_p = \frac{1}{\tau_p}\#(S - 3)$$

$$\tau_p^{polymer} = \frac{1}{k_{ISC} + k_r^s + k_{nr}^s}\#(S - 4)$$

In equation (S-3, S-4), τ_p indicates a nanosecond-scale prompt fluorescence component for the poly(TMTPA-DCB), k_{ISC} is a rate constant of intersystem crossing,

k_r^S and k_{nr}^S stand for the radiative and nonradiative decay rate, respectively⁴. Therefore, the τ_p of the doped Emitting Layer with the FET process can be expressed by the expressions **(S-5)** and **(S-6)** (Dye_x denotes the m-Cz-BNCz content).

$$\tau_p^{Dye_x} = \frac{1}{k_{ISC} + k_r^S + k_{nr}^S + k_{FET}} \#(S-5)$$

$$k_{FET} = \frac{1}{\tau_p^{Dye_x}} - \frac{1}{\tau_p^{polymer}} \#(S-6)$$

Finally, the average distance between poly(TMTPA-DCB) and m-Cz-BNCz (R) was calculated following the expression as **(S-7)**:

$$k_{FRET} = \frac{1}{\tau} \left(\frac{R_{FET}}{R} \right)^6 \#(S-7)$$

Taking mCP: 10 wt.% poly(TMTPA-DCB): 2.0 wt.% m-Cz-BNCz as an example, k_{FET} and R were determined to be $7.32 \times 10^7 \text{ s}^{-1}$ and 3.54 nm, respectively. Compared with the FET radius ($R_{FET} = 4.15 \text{ nm}$), the lower R means that the FET could happen from poly(TMTPA-DCB) to m-Cz-BNCz (Table S3).

In addition, we also calculated k_{FET} and k_{DET} for different m-Cz-BNCz doping concentrations by expressions **(S-3)**, **(S-7 ~ S-11)**. (Table S2 and Fig. S2)

$$k_D = \frac{1}{\tau_D} \#(S-8)$$

$$k_{ISC} = k_P * (1 - \Phi_P) \#(S-9)$$

$$k_{RISC} = \left(\frac{k_P k_D}{k_{ISC}} \right) * \left(\frac{\Phi_D}{\Phi_P} \right) \#(S-10)$$

$$k_{DET} = \frac{1}{\tau_D^x} - \frac{1}{\tau_D^{polymer}} + k_{ISC} k_{RISC} * (\tau_P^x - \tau_P^{polymer}) \# (S-11)$$

Table S2. Summary of the PL characteristics and rate constants of mCP: 10 wt.% poly(TMTPA-DCB): x wt.% m-Cz-BNCz films (x = 0.0, 0.2, 0.4, 0.6, 1.0, 2.0) and mCP: 2.0 wt.% m-Cz-BNCz film.

x wt.%	PLQY (%)	τ_p (ns)	τ_d (μ s)	Φ_P (%)	Φ_D (%)	k_{RISC} (10^5 s^{-1})	k_{ISC} (10^6 s^{-1})	k_{FET} (10^7 s^{-1})	k_{DET} (10^4 s^{-1})
0.0	97.58	35.44	5.19	63.42	34.16	2.84	1.03	-	-

0.2	89.40	20.93	4.53	53.09	36.31	2.84	1.03	1.96	2.82
0.4	86.00	13.86	4.08	50.03	35.97	2.84	1.03	4.39	5.22
0.6	82.80	12.45	3.94	48.73	34.07	2.84	1.03	5.21	6.10
1.0	74.00	11.05	3.25	47.69	26.31	2.84	1.03	6.23	11.51
2.0	68.90	9.86	2.26	42.16	26.74	2.84	1.03	7.32	24.93
2.0 (without polymer)	23.10	8.94	1.45	13.65	9.45	2.84	1.03	-	-

Experimental data is collected in neat films under nitrogen atmosphere; τ_p and τ_d are prompt and delayed lifetimes; Φ_p and Φ_d are PLQYs of prompt and delayed components; k_{ISC} and k_{RISC} are rate constants of intersystem crossing from S_1 to T_1 and reverse intersystem crossing from T_1 to S_1 in the poly(TMTPA-DCB); k_{FET} and k_{DET} are Förster energy transfer rate and Dexter energy transfer rate from poly(TMTPA-DCB) to m-Cz-BNCz, respectively.

Table S3. Summary of the FET calculation from poly(TMTPA-DCB) to m-Cz-BNCz.

Compound	Dopant concentration (wt. %)	PLQY ^a	$J(\lambda)^b$ ($\text{mol}^{-1}\text{dm}^3\text{cm}^{-1}\text{nm}^4$)	R_{FET}^c (nm)	R^d (nm)
m-Cz-BNCz	2.0	69.9	1.30×10^{15}	4.15	3.54

^a PLQY measured using an integrating sphere in thin film; ^b spectral overlap between PL emission of poly(TMTPA-DCB) and absorption spectrum of m-Cz-BNCz; ^c FET radius; ^d intermolecular distance; ^e prompt fluorescence component of doped thin film; ^f FET rate constant.

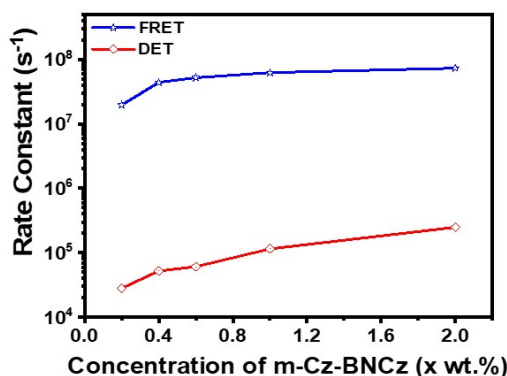


Fig. S2 The FET and DET rate constants in the doped films of mCP: 10 wt.% poly(TMTPA-DCB): x wt.% m-Cz-BNCz (x = 0.0, 0.2, 0.4, 0.6, 1.0, and 2.0, respectively).

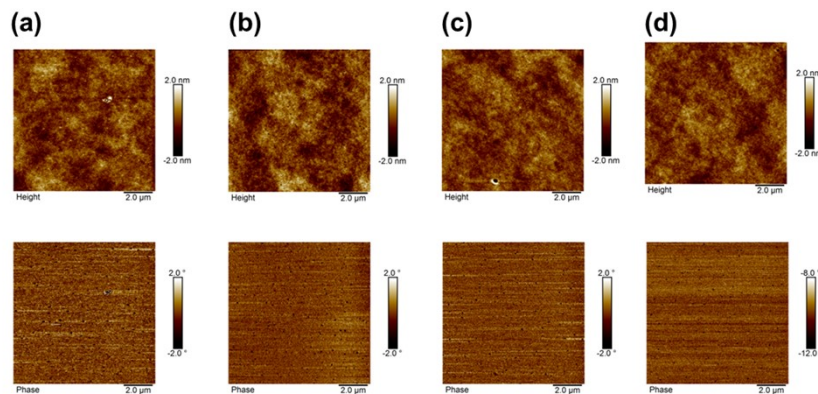


Fig. S3 AFM morphology images for mCP: 10 wt.% poly(TMTPA-DCB): x wt.% m-Cz-BNCz ($x = 0.2$ (a), 0.4 (b), 0.6 (c), 1.0 (d)). The RMS values were measured as 1.054, 1.092, 1.154 and 1.176 nm for m-Cz-BNCz doping concentrations ranging from 0.2 wt.% to 1.0 wt.%.

4. Device fabrication and testing

The solution-processed devices were fabricated with a configuration of ITO/PEDOT: PSS + PFI (40 nm)/ EML (30 nm)/ TSPO1(8 nm)/ TmPyPB (42 nm)/ LiF (1 nm)/ Al (200 nm). Firstly, the cleaned ITO-coated substrates (15Ω per square) were treated with Ultraviolet-ozone (UVO) for 1 h. Secondly, the PEDOT: PSS (Clevios PVP A14083) mixed with PFI (volume ratio = 2:3) was spin-coated on the substrate in the air at a speed of 5000 rpm, annealed at $120 \text{ }^\circ\text{C}$ for 1 h, and then transferred into a nitrogen-filled glove box. Then the chlorobenzene solution of m-Cz-BNCz doped into mCP and poly(TMTPA-DCB) at different concentrations were spin-coated at a speed of 1800 rpm for 60 s to form the 30 nm thickness of emitting layer (EML). After being transferred into a vacuum chamber, other functional layers including TSPO1 (8 nm), TmPyPB (42 nm), LiF (1 nm), and Al (200 nm) were thermally deposited layer-by-layer in sequence at a pressure under 3×10^{-4} Pa. The current density-voltage-luminance (J-V-L) characteristics of all fabricated devices were measured using Keithley 2000 and Keithley 2400 (fitted with calibrated silicon photodiode) systems. The EL spectra and CIE coordinates were measured using a CS2000 spectrometer. EQE was calculated from the EL spectrum, luminance, and current density assuming a

Lambertian emission distribution. All the measurements were performed at room temperature and air atmosphere.

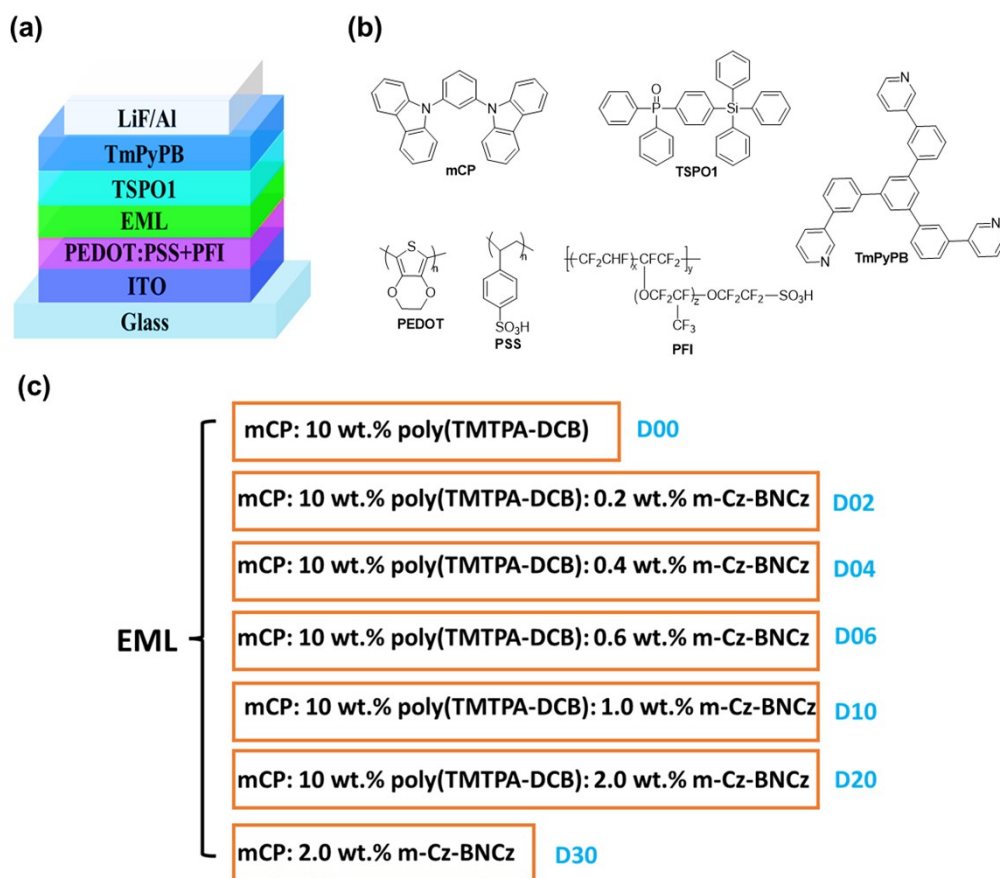


Fig. S4 (a) Solution processed device structure in this work; (b) The molecular structures of used EL materials; (c) The specific constituents of EML.

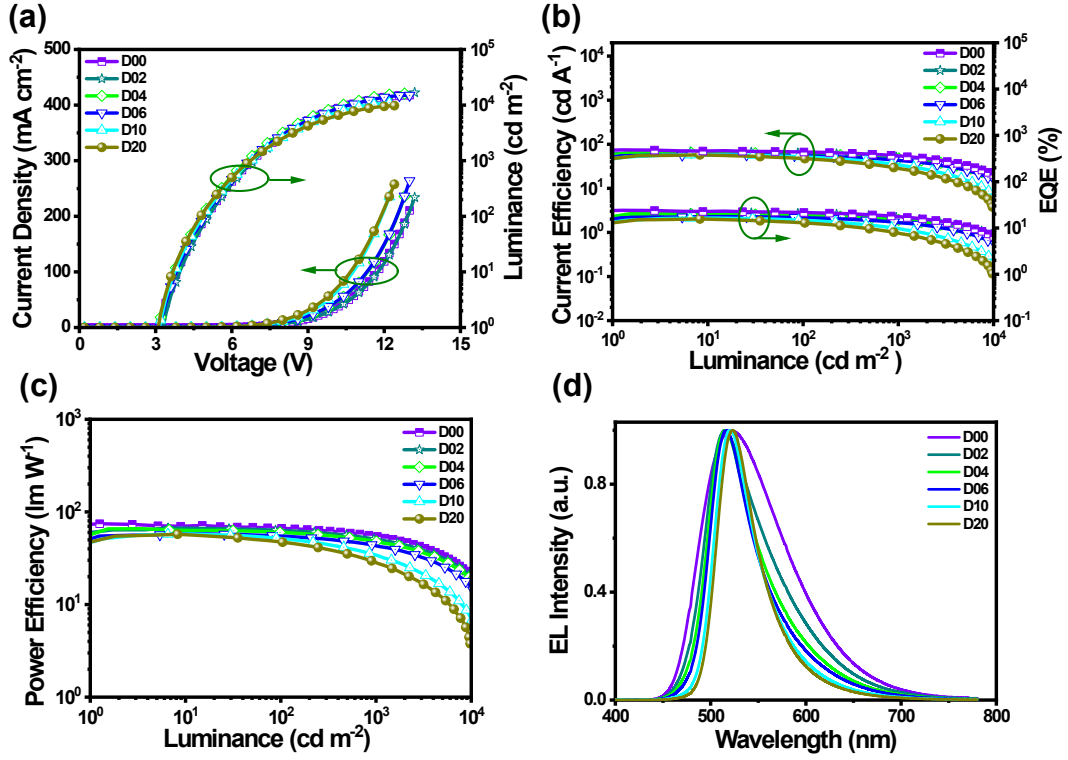


Fig. S5 Device performance of D00, D02, D04, D06, D10 and D20: (a) Current density-voltage-luminance characteristics; (b) Current efficiency and EQE vs luminance; (c) Power efficiency vs luminance; (d) EL spectra at 1000 cd m^{-2} .

Table S4. Summary of the device performance of D00, D02, D04, D06, D10 and D20.

Device	V_{on}^{a} (V)	$L_{\text{max}}^{\text{b}}$ (cd m^{-2})	CE^{c} (cd A^{-1})	PE^{c} (lm W^{-1})	EQE^{c} (%)	CIE^{d} (x, y)	SE^{e} (%)	$\lambda_{\text{EL}}^{\text{f}}$ (nm)	FWHM^{g} (nm)
D00	3.2	15067	74.5/54.3	73.1/25.1	24.1/17.6	(0.32,0.55)	-	522	102
D02	3.2	16735	66.9/50.9	55.8/24.5	21.3/16.9	(0.31,0.58)	60.97	514	77
D04	3.2	16034	65.9/48.0	60.8/22.8	20.5/14.9	(0.29,0.60)	70.80	515	63
D06	3.2	14969	58.0/44.2	53.8/21.0	17.7/13.4	(0.29,0.61)	76.88	517	57
D10	3.2	11028	57.8/33.9	52.1/15.6	16.4/9.6	(0.28,0.64)	86.28	521	52
D20	3.2	9738	57.4/27.7	53.7/12.8	15.7/7.1	(0.28,0.65)	91.69	523	48

^a Turn-on voltage at 1 cd m^{-2} ; ^b Maximum luminance; ^c Data at maximum and 1000 cd m^{-2} ; ^d Data at a driving voltage of 6 V; ^e Sensitization efficiency; ^f EL peak wavelength; ^g Full width at half maximum; CE: Current efficiency; PE: power efficiency; EQE: external quantum efficiency; CIE: Commission Internationale de l'Eclairage.

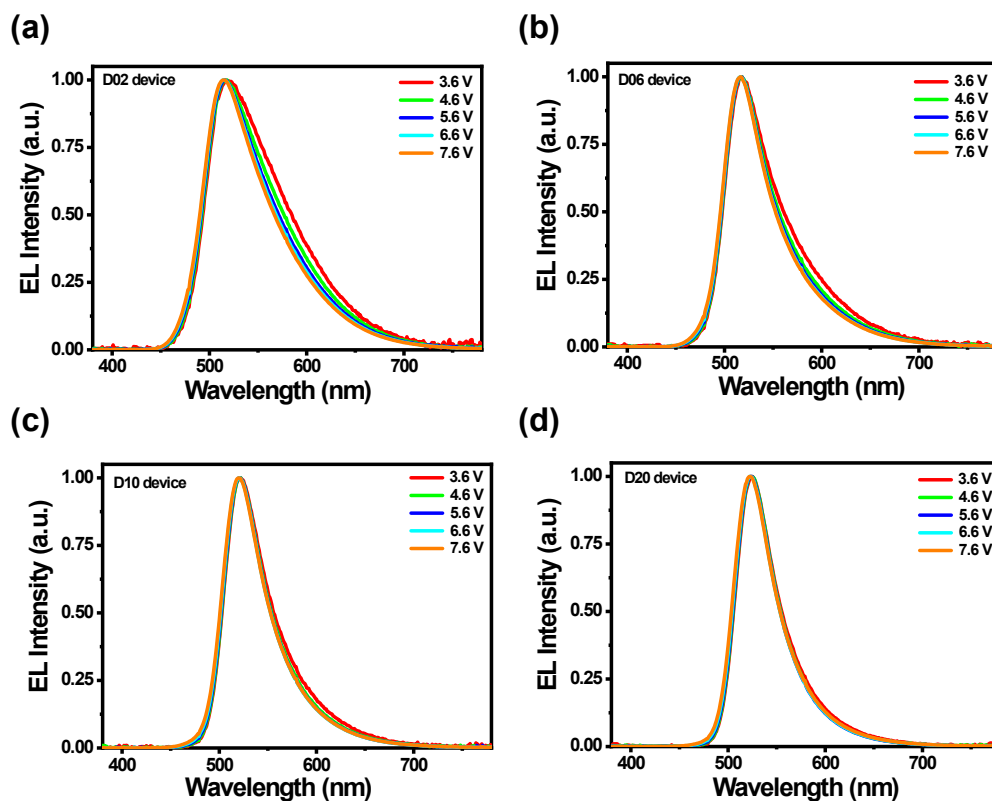


Fig. S6 Voltage dependence of the EL spectra for device D02 (a), D06 (b), D10 (c) and D20 (d).

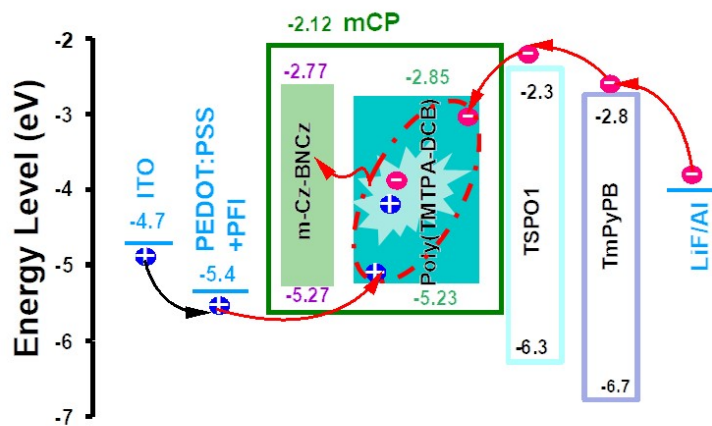


Fig. S7 The energy level and excitation transfer diagram of the device.

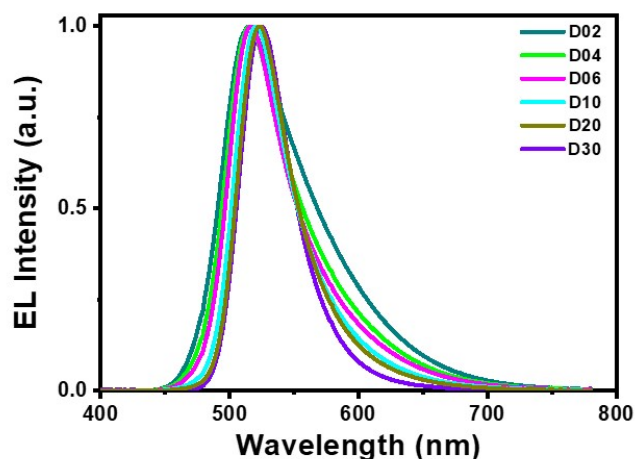


Fig. S8 The calculation of sensitization efficiency (SE) for sensitized TADF devices. The SE values were determined to be 60.97%, 70.80%, 76.88%, 86.28%, and 91.69% with the doping concentrations ranging from 0.2 wt% to 2.0 wt%, respectively, utilizing the equation $SE = 1 - P_{\text{sen}}/P_{\text{total}}$, where P_{sen} represents the emission intensity of the TADF sensitizer in the EL spectra and P_{total} denotes the total emission intensity of the EL spectra.

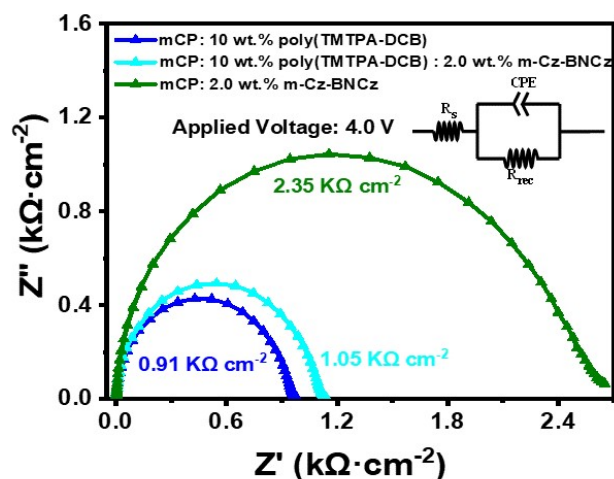


Fig. S9 Nyquist plots with a forward voltage of 4.0 V (Inset: simplified equivalent circuit).

5. References

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