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Supplementary Information

Non-Halogenated Solvent-Processed TADF OLEDs with Polymer-Based Exciplex Hosts

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Experimental section

Instrumentation

The ¹H and ¹³C nuclear magnetic resonance (NMR) spectra were obtained in CDCl₃ solvents using a Varian Mercury spectrometer. The molecular weight characteristics (M_n and PDI) of PC4-2Cz, PC8-2Cz, and PC12-2Cz were analyzed via gel permeation chromatography (GPC, Agilent 1200 series) using 1,2-dichlorobenzene as the eluent and polystyrene standards. Elemental composition and purity of the compounds were determined using a FlashSmart elemental analyzer (Thermo Fisher Scientific). Film roughness and thickness were measured by atomic force microscopy (AFM) in non-contact mode utilizing a Park Systems XE-100 instrument. Glass transition temperatures (T_g) were measured under an N₂ atmosphere via differential scanning calorimetry (DSC, Mettler STARe). Thermal decomposition temperatures $(T_{\rm d})$ of the materials were assessed by thermogravimetric analysis (TGA, Mettler STARe) at a heating rate of 10 °C min-1 under N2 conditions. Electrochemical properties of the host materials in film form were evaluated by cyclic voltammetry (CV) using a potentiostat (EA161, eDAQ) at a scan rate of 50 mV s⁻¹ to determine oxidation potentials. Host films were prepared by directly coating the materials onto platinum plates, which served as working electrodes. A platinum wire (diameter: 0.5 nm) and an Ag/AgCl electrode were used as the counter and reference electrodes, respectively.

The UV-vis absorption spectra of the host materials in chloroform solutions and thin films were measured using a UV-vis spectrophotometer (Agilent 8453, photodiode array, wavelength range: 190–1100 nm). Fluorescence spectra at 77 K and 298 K, along with phosphorescence spectra at 77 K (delay time: 1.0 ms), were recorded using an F-7100

fluorescence spectrophotometer (HITACHI). The absolute photoluminescence quantum yields (PLQYs) of the thin films were determined using an integrating sphere (ILF-835) in combination with an FP-8500 spectrofluorometer (JASCO). For time-resolved photoluminescence (TRPL) measurements, film samples were placed in a cryostat (Janis VPF-100) under vacuum conditions ($\leq \sim 10^{-2}$ mTorr) and excited by 340 nm pulses generated through third-harmonic conversion of a 1064 nm Nd:YAG laser (Q-smart 850, Lumibird FR). The TRPL signals were recorded using a photomultiplier tube (Hamamatsu Photonics, R955) connected to a 100 MHz digital oscilloscope (DSO-X 3014A, Keysight).

Theoretical calculations

Density functional theory (DFT) calculations were performed using the Gaussian 16 software package with the B3LYP functional and the 6-31G(d) basis set. Optimized molecular geometries, frontier molecular orbitals (HOMO and LUMO), and electronic excited-state energies (S_n and T_n) were determined. Solvation effects in toluene were accounted for using the polarizable continuum model (PCM) with the integral equation formalism variant (IEFPCM).

Space-charge-limited current (SCLC) method for measuring hole mobility

The hole mobilities of the polymer hosts (PC4-2Cz, PC8-2Cz, and PC12-2Cz) were measured using the SCLC method after fabrication in a typical device structure of ITO/PEDOT:PSS (30 nm)/polymer hosts (50 nm)/Al (100 nm). Mobility was calculated using the equation $J = 9\varepsilon_0\varepsilon_r\mu V^2/8L^3$, where J is the current density, ε_0 is the permittivity of the host layer ε_r is the relative dielectric constant, μ is the hole mobility, and L is the thickness of the host layer. The internal voltage was determined using the equation $V = V_{appl} - V_{bi} - V_{a}$, where V_{appl} is the voltage applied to the device, V_{bi} corresponds to the work-function difference between the two electrodes, and V_{a} is the voltage drop.

OLED device fabrication

OLED devices were fabricated on glass substrates with a 150 nm transparent ITO layer as the anode, featuring a sheet resistance of 15 Ω cm⁻² and an active pattern size of 2 × 2 mm². The substrates underwent sequential ultrasonic cleaning in distilled water (10 min), isopropanol (20 min), and HPLC-grade isopropanol (20 min), followed by drying at 100 °C. A 30 nm hole injection layer was formed by spin-coating PEDOT:PSS onto the ITO surface, followed by annealing at 155 °C for 15 min on a hot plate. The emitting layer was prepared by spin-coating a blend of host materials and t4CzIPN (96:4 wt ratio) using different solvent concentrations: 0.5 wt% in CB and Tol (EML thickness: 40 nm), and 0.1 wt% in 2-MeTHF (EML thickness: 32 nm). A 50 nm BmPyPB layer was introduced as the electron transport layer, while LiF (1 nm) and Al (100 nm) were vacuum-deposited under an inert atmosphere at 5 × 10⁻⁶ Torr. The final OLED structure comprised ITO (150 nm)/ PEDOT:PSS (30 nm)/host:t4CzIPN (40 nm)/BmPyPB (50 nm)/LiF (1 nm)/ Al (100 nm). Device fabrication was conducted under ambient conditions before transferring the substrates to a thermal vacuum evaporator for electron transport and electrode layer deposition.



Fig. S1. ¹H NMR spectrum of 9-(4-bromobutyl)-9'-phenyl-9*H*,9'*H*-3,3'-bicarbazole (2a).



Fig. S2. ¹³C NMR spectrum of 9-(4-bromobutyl)-9'-phenyl-9*H*,9'*H*-3,3'-bicarbazole (**2a**).







Fig. S5. ¹H NMR spectrum of 9-phenyl-9'-(4-(4-vinylphenoxy)butyl)-9*H*,9'*H*-3,3'-bicarbazole (4a).



Fig. S6. ¹³C NMR spectrum of 9-phenyl-9'-(4-(4-vinylphenoxy)butyl)-9*H*,9'*H*-3,3'-bicarbazole (4a).



Fig. S7. ¹H NMR spectrum of PC4-2Cz (5a).



Fig. S8. ¹H NMR spectrum of 9-(8-bromooctyl)-9'-phenyl-9*H*,9'*H*-3,3'-bicarbazole (2b).





Fig. S10. ¹H NMR s yl)octyl)oxy)benzaldehyde (**3b**). 4-((8-(9'-phenyl-9H,9'H-[3,3'-bicarbazol]-9of spectrum



4-((8-(9'-phenyl-9*H*,9'*H*-[3,3'-bicarbazol]-9-¹³C Fig. **S11.** NMR spectrum yl)octyl)oxy)benzaldehyde (3b).



Fig. S12. ¹H NMR spectrum of 9-phenyl-9'-(8-(4-vinylphenoxy)octyl)-9H,9'H-3,3'bicarbazole (4b).



Fig. S13. ¹³C NMR spectrum of 9-phenyl-9'-(8-(4-vinylphenoxy)octyl)-9H,9'H-3,3'-bicarbazole (4b).





Fig. S15. ¹H NMR spectrum of 9-(12-bromododecyl)-9'-phenyl-9*H*,9'*H*-3,3'-bicarbazole (2c).



Fig. S16. ¹³C NMR spectrum of 9-(12-bromododecyl)-9'-phenyl-9*H*,9'*H*-3,3'-bicarbazole (2c).



Fig. S17. ¹H NMR spectrum of 4-((12-(9'-phenyl-9H,9'H-[3,3'-bicarbazol]-9-yl)dodecyl)oxy)benzaldehyde (3c).



Fig. S18. ¹³C NMR spectrum of 4-((12-(9'-phenyl-9*H*,9'*H*-[3,3'-bicarbazol]-9-yl)dodecyl)oxy)benzaldehyde (**3c**).



Fig. S19. ¹H NMR spectrum of 9-phenyl-9'-(12-(4-vinylphenoxy)dodecyl)-9*H*,9'*H*-3,3'-bicarbazole (4c).



Fig. S20. ¹³C NMR spectrum of 9-phenyl-9'-(12-(4-vinylphenoxy)dodecyl)-9*H*,9'*H*-3,3'-bicarbazole (**4c**).



Fig. S21. ¹H NMR spectrum of PC12-2Cz (5c).



Fig. S22. TGA (a) and DSC (b) thermograms of PC4-2Cz, PC8-2Cz and PC12-2Cz.

Tab	le.	S 1.	Properties	of the	three po	lymer	hosts
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Host	M _n [kDa]	PDI	$T_d^{onset} / T_g^{onset} (^{\circ}C)$
PC4-2Cz	14.7	2.10	476 / 150
PC8-2Cz	11.4	2.38	450 / 109
PC12-2Cz	27.2	2.27	450 / 93



Fig. S23. Cyclic voltammograms of PC4-2Cz, PC8-2Cz and PC12-2Cz in the film state.



Fig. S24. Optimized geometries, HOMO, LUMO, S_1 , and T_1 energy levels of 2Cz, PC4-2Cz, PC8-2Cz, PC12-2Cz, and tPDBA obtained using the DFT method with B3LYP/6-31G(d). For polymers, calculations were performed using a simplified model based on the repeating unit.



Fig. S25. Photoluminescence (PL) spectra of PCm-2Cz:tPDBA (m = 4, 8, 12) blends at various mixing ratios (x:y wt%).

The newly synthesized p-type polymer was used as an exciplex host with tPDBA as the n-type material. To determine the optimal blending ratio, PL spectra of blend films were measured and optimized. For PC4-2Cz:tPDBA (1.5:1 wt%), PC8-2Cz:tPDBA (1.6:1 wt%), and PC12-2Cz:tPDBA (1.7:1 wt%), exciplex emission dominated while single-host emissions were minimized. These weight ratios were converted to a 1:1 mol% ratio based on molecular weights, identifying 1:1 mol% as the optimal ratio for device fabrication.



Fig. S26. UV-vis absorption and PL spectra of (a) PC4-2Cz:tPDBA (1:1 mol%), (c) PC8-2Cz:tPDBA (1:1 mol%) and (d) PC12-2Cz:tPDBA (1:1 mol%) in the chloroform solution and film states. The phosphorescence (Phos) spectra were measured in the film state at 77 K.

From these results, we determined the ΔE_{ST} values in the film state as follows: (a) PC4-2Cz:tPDBA - 0.06 eV, (b) PC8-2Cz:tPDBA - 0.06 eV, and (c) PC12-2Cz:tPDBA - 0.05 eV.

Table S2. Photophysical parameters and rate constants for three polymer-based exciplex host systems

Materials (1:1 mol%)	$\Phi_{\rm PL}^{\ a}(\%)$	$\Phi_{p}^{\ b}(\%)$	$\Phi_{\rm d}^{\rm \ c}(\%)$	$\tau_{p}^{\ d}(ns)$	$\tau_{d}^{\ e}(\mu s)$	$k_{\rm ISC}^{\rm f} (\times 10^7 {\rm s}^{-1})$	$k_{\rm RISC}^{g} (\times 10^7 {\rm s}^{-1})$	$k_{\rm nr,S}^{\rm h} (\times 10^7 {\rm s}^{-1})$
PC4-2Cz:tPDBA	24.3	8.1	16.2	20.1	0.122	3.32	2.46	1.25
PC8-2Cz:tPDBA	23.0	7.6	15.4	19.7	0.103	3.40	2.94	1.29
PC12-2Cz:tPDBA	21.3	8.0	13.3	15.0	0.089	4.18	3.00	1.97

^a Overall absolute PLQY obtained using an integrating sphere; ^b PLQY of the prompt fluorescence(PF); ^c PLQY of the delayed fluorescence (DF); ^d PF lifetime; ^e DF lifetime; ^f Intersystem crossing rate constant; ^g Reverse intersystem crossing rate constant; ^h Nonradiative decay rate constant of the singlet excited state.



Fig. S27. Atomic force microscopy (AFM) images of (a) the neat film of the small-molecule host film (Me-2Cz) and (b) the exciplex host film blended with tPDBA.

This study employed AFM measurements to examine the impact of 2Cz incorporation into the side chains of three synthesized polymers on thin-film morphology. Films coated with 2Cz exhibited significant irregularities and high roughness compared to polymer films with 2Cz in

their side chains. AFM data revealed poor film-forming properties for Me-2Cz and its blend with tPDBA, with Rq values of 29.7 nm and 32.5 nm for $5.0 \,\mu\text{m} \times 5.0 \,\mu\text{m}$ scan areas, indicating uneven surfaces with prominent protrusions. Height profiles confirmed substantial surface variations due to aggregated domains and defects.

Therefore, incorporating Me-2Cz in solution-processed films increases surface roughness and heterogeneity, making it unsuitable for OLED EMLs, where smooth, uniform films are essential for optimal performance.



Fig. S28. Interaction radius (R_a) of PC4-2Cz, PC8-2Cz and PC12-2Cz in each solvent.

Table S3. Hansen solubility parameters and interaction radius of PC4-2Cz, PC8-2Cz, PC12-2Cz.

Host	δ_{d}	δ_{p}	$\delta_{\rm h}$	R _a (Toluene)	<i>R</i> _a (Chlorobenzene)	R _a (Chloroform)	R _a (2-MeTHF)	R _a (Ethanol)
PC4-2Cz	23.3	3.94	6.54	11.81	9.73	11.06	13.92	20.35
PC8-2Cz	22.1	3.25	5.94	9.28	7.42	8.60	11.55	19.25
PC12-2Cz	21.3	2.77	5.48	7.59	5.97	7.01	10.02	18.74

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Functional Group	Occurences	V (cm ³ mol ⁻¹)	$F_{ m di} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$	$F_{ m pi} \over ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$	$E_{ m hi} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$
-CH ₂ -	5	16.1 (80.5)	270 (1350)	0	0
>CH-	1	-1.0 (-1.0)	80 (80)	0	0
=C<	13	-5.5 (-71.5)	70 (910)	0	0
=CH-	23	13.5 (310.5)	200 (4,600)	0	0
>N-	2	-9.0 (-18.0)	20 (40)	800	5000 (10,000)
-0-	1	3.8	100	400	3000
Σ Total	-	304.3	7,080	1,440,000	13,000
δ	-	-	$\delta_d = 23.3$	$\delta_p = 3.94$	$\delta_h = 6.54$

Table S4. Functional group contribution terms used for calculating Hansen solubility parameters (HSPs) of PC4-2Cz.

Functional group contribution values were taken from reference. ^{S1}

Table S5. HSPs for the various organic solvents and calculated R_a values of PC4-2Cz

Salvant		$R_{\rm a}$ (MPa ^{1/2})		
Solvent	δ_d	δ_p	δ_{h}	to PC4-2Cz
Toluene	18	1.4	2	11.81
Chlorobenzene	19	4.3	2	9.73
Chloroform	17.8	3.1	5.7	11.06
2-MeTHF	16.4	4.8	4.9	13.92
Ethanol	15.8	8.8	19.4	20.35

All the HSP values were taken from reference. S2

Table S6. Functional group contribution terms used for calculating Hansen solubility parameters (HSPs) of PC8-2Cz.

Functional Group	Occurences	V (cm ³ mol ⁻¹)	$F_{ m di} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$	$F_{ m pi} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$	$E_{ m hi} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$
-CH ₂ -	9	16.1 (144.9)	270 (2430)	0	0
>CH-	1	-1.0 (-1.0)	80 (80)	0	0
=C<	13	-5.5 (-71.5)	70 (910)	0	0
=CH-	23	13.5 (310.5)	200 (4,600)	0	0
>N-	2	-9.0 (-18.0)	20 (40)	800	5000 (10,000)
-0-	1	3.8	100	400	3000
Σ Total	-	368.7	8,160	1,440,000	13,000
δ	-	-	$\delta_d = 22.1$	$\delta_p=3.25$	$\delta_{\rm h} = 5.94$

Functional group contribution values were taken from the reference ^{S1}

Colvert	-	$R_{\rm a}({\rm MPa^{1/2}})$		
Solvent	δ_d	δ_p	$\delta_{\rm h}$	to PC8-2Cz
Toluene	18	1.4	2	9.28
Chlorobenzene	19	4.3	2	7.42
Chloroform	17.8	3.1	5.7	8.60
2-MeTHF	16.4	4.8	4.9	11.55
Ethanol	15.8	8.8	19.4	19.25

Table S7. HSPs for the various organic solvents and calculated R_a values of PC8-2Cz

All the HSP values were taken from the reference ^{S2}

 Table S8. Functional group contribution terms used for calculating Hansen solubility parameters (HSPs) of PC12-2Cz.

Functional Group	Occurences	V (cm ³ mol ⁻¹)	$F_{ m di} \ ({ m J}^{1/2}{ m cm}^{3/2}{ m mol}^{-1})$	$F_{ m pi} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$	$E_{ m hi} \ ({ m J}^{1/2}~{ m cm}^{3/2}~{ m mol}^{-1})$
-CH ₂ -	9	16.1 (209.3)	270 (3510)	0	0
>CH-	1	-1.0 (-1.0)	80 (80)	0	0
=C<	13	-5.5 (-71.5)	70 (910)	0	0
=CH-	23	13.5 (310.5)	200 (4,600)	0	0
>N-	2	-9.0 (-18.0)	20 (40)	800	5000 (10,000)
-0-	1	3.8	100	400	3000
Σ Total	-	433.1	9,240	1,440,000	13,000
δ	-	-	$\delta_d = 21.3$	$\delta_p = 2.77$	$\delta_{\rm h}=5.48$

Functional group contribution values were taken from the reference S1

Table S9. HSPs for the various organic solvents and calculated R_a values of PC12-2Cz

Calment]		$R_{\rm a}({\rm MPa}^{1/2})$	
Solvent	δ_d	δ_p	$\delta_{\rm h}$	to PC12-2Cz
Toluene	18	1.4	2	7.59
Chlorobenzene	19	4.3	2	5.97
Chloroform	17.8	3.1	5.7	7.01
2-MeTHF	16.4	4.8	4.9	10.02
Ethanol	15.8	8.8	19.4	18.74

All HSP values were sourced from the reference. ^{S2}

	Classifica		Solubility		
Solvent	Halogenated or Non-halogenated	Aromatic or Aliphatic	PC4-2Cz	PC8-2Cz	PC12-2Cz
Dichloromethane	Halogenated	Aliphatic	0	0	0
Chloroform	Halogenated	Aliphatic	0	0	0
Chlorobenzene	Halogenated	Aromatic	0	0	0
Toluene	Non-halogenated	Aromatic	Х	Ο	0
2-MeTHF	Non-halogenated	Aliphatic	Х	Х	0
Ethanol	Non-halogenated	Aliphatic	Х	Х	Х

Table S10. Solubility of PC4-2Cz, PC8-2Cz and PC12-2Cz in each solvent.

O: Soluble, \triangle = slightly soluble, X: Insoluble at RT



Fig. S29. Photographs of solutions of (a) PC4-2Cz, (b) PC8-2Cz, and (c) PC12-2Cz in 0.2 wt% EML solvents, including dichloromethane (DCM), chloroform (CF), chlorobenzene (CB), toluene (Tol), 2-methyltetrahydrofuran (2-MeTHF), and ethanol (EtOH). The polymer hosts were dissolved at room temperature.



Fig. S30. Space charge-limited current (SCLC) measurements of the hole-only devices fabricated using p-type polymer host films (PC4-2Cz, PC8-2Cz, and PC12-2Cz).



Fig S31. Chemical structures of the materials employed in the devices.



Fig S32. (a) Device configurations and energy diagram of the solution-processed TADF-OLEDs. (b) Current density–voltage–luminance (J-V-L) curves and (c) external quantum efficiency (EQE) of devices based on polymer single hosts and exciplex hosts.

Table S11. EL performance of solution-processed TADF-OLEDs based on polymer hosts.

Host	Emitter	Doping Conc.	$V_{ m on}{}^{ m a)}$ (V)	$\eta_{c, \max}^{b)}$ (cd/A)	$\eta_{\rm p, max}^{\rm c)}$ (lm/W)	$L^{d)}$ (cd/m ²)	$\eta_{\mathrm{ext, max}}^{\mathrm{e})}$ (%)	FWHM (nm)	$\text{CIE}\left(x,y\right){}^{\text{f})}$
PC4-2Cz			2.71	34.48	30.949	12380	10.101	88	(0.38, 0.57)
PC8-2Cz	t4CzIPN	4wt%	2.67	35.43	37.102	7582	10.432	88	(0.38, 0.57)
PC12-2Cz			2.67	33.105	34.667	7411	9.7902	88	(0.38, 0.57)

^{a)} Turn-on voltage at 1 cd/m². ^{b)} Maximum CE. ^{c)} Maximum PE. ^{d)} Maximum luminance. ^{e)} Maximum EQE. ^{f)} CIE color coordinates at 1000 cd/m².

Table S12. EL performance of solution-processed TADF-OLEDs based on polymer exciplex hosts.

Host (1:1 mol%)	Emitter	Doping Conc.	V _{on} ^{a)} (V)	$\eta_{ m c,max}^{ m b)}$ (cd/A)	$\eta_{\rm p, max}^{\rm c)}$ (lm/W)	$L^{d)}$ (cd/m ²)	$\eta_{\mathrm{ext,max}}^{\mathrm{e)}}$	FWHM (nm)	$CIE\left(x,y\right){}^{\rm f)}$
PC4-2Cz:tPDBA			2.73	58.353	52.378	22500	16.850	88	(0.37, 0.58)
PC8-2Cz:tPDBA	t4CzIPN	4wt%	2.73	54.530	57.104	17060	16.044	88	(0.37, 0.58)
PC12-2Cz:tPDBA			2.73	55.339	49.672	16780	16.024	88	(0.37, 0.58)

^{a)} Turn-on voltage at 1 cd/m². ^{b)} Maximum CE. ^{c)} Maximum PE. ^{d)} Maximum luminance. ^{e)} Maximum EQE. ^{f)} CIE color coordinates at 1000 cd/m².



Photoluminescence quantum yield of single and mixed host doped films

Fig. S33. PLQY values of (a) PC4-2Cz, PC8-2Cz and PC12-2Cz, and (b) blend films of PC4-2Cz:tPDBA (1:1 mol%), PC8-2Cz:tPDBA (1:1 mol%) and PC12-2Cz:tPDBA (1:1 mol%). All films were measured at an excitation wavelength of 300 nm.



Fig. S34. PLQY values for: (a) PC4-2Cz, PC8-2Cz and PC12-2Cz (b) exciplex host systems: PC4-2Cz:tPDBA (1:1 mol%), PC8-2Cz:tPDBA (1:1 mol%), and PC12-2Cz:tPDBA (1:1 mol%) with 4 wt% t4CzIPN in the film state. The excitation wavelength is 300 nm.

The photoluminescence quantum yield (PLQY) of p-type polymers PC4-2Cz, PC8-2Cz, and PC12-2Cz was compared with exciplex host films blended with the tPDBA acceptor. The results revealed a significantly lower PLQY in pristine polymer films than in exciplex systems, underscoring the critical role of tPDBA in enhancing PLQY. Suitable acceptors like tPDBA

create an optimal electronic environment for charge separation and recombination, thereby improving quantum yield. This behavior aligns with previous studies showing that optimized charge transport and exciton formation enhance photoluminescence.

References

- S1. C. M. Hansen, *Hansen Solubility Parameters: A User's Handbook*, CRC Press, Boca Raton, 2007.
- S2. D.W. van Krevelen, K. te Nijenhuis, *Properties of Polymers: Their correlation with chemical structures; Their numerical estimation and prediction from additive group contributions*, Elsevier Science, Amsterdam, The Netherlands, 2009.