Supporting Information for

Positional Isomer Engineering of Carbazole-Based Hole Transport Materials for Perovskite Solar Cells

Xiangyu Ni,[a] Cheng Chen,*[a] Ziyang Xia,[a] Ziqi Zhao,[a] Haoxin Wang,*[a] Bin Cai,[a] Ming Cheng[a]

[a] Institute for Energy Research, School of Future Technology, Jiangsu University, Zhenjiang 212013, China.

* E-mail: chencheng@ujs.edu.cn (C. Chen); wanghaoxin@ujs.edu.cn (H. Wang)

Supporting Information

1. Materials

The raw materials and palladium catalysts used for TCBZ - 27 and TCBZ - 36 molecules were purchased from Energy Chemicals, China. Fluorine doped tin oxide (FTO) coated glass (TEC; 7 Ωsq-1, Advanced Election Technology Co., Ltd.), Bis (2,4 - pentane dioic acid) bis (2 - propanoic acid) titanium (IV) (75% in Isopropyl Alcohol, TCI), ethanol (≥99.5%, Aladdin), tin (IV) oxide (SnO2) colloid precursor (15% in H2O colloidal dispersion, Alfa Aesar), polyacrylic acid (PAA, avg. MW = 450000, Sigma-Aldrich), formamidine iodide (FAI; 99.90%, Advanced Election Technology Co., Ltd.), lead (II) iodide (PbI2; 99.999%, Advanced Election Technology Co., Ltd.), Methylammonium tribromoplumbate(II) (MAPbBr3 99%, Xi’an Polymer Light Technology Corp.), Methylammonium chloride (MACl; 99.5%, Xi’an Polymer Light Technology Corp.), Spiro - OMeTAD (99.86%, S1 Advanced Election Technology Co., Ltd.), Lithium bis(trifluoromethanesulfonyl)imide (Li - TFSI; 99%, Xi’an Polymer Light Technology Corp.), 4-tert-butylpyridine (tBP; 99%, Xi ’an Polymer Light Technology Corp.), *N,N*-dimethylformamide (DMF; 99.8%, Sigma-Aldrich), dimethyl sulfoxide (DMSO; 99.9%, Sigma-Aldrich), chlorobenzene (CB; 99.8%; Sigma-Aldrich). All the solvents and chemicals were used as received unless specially stated.

2. Perovskite Solar Cell Fabrication

The n-i-p structured photovoltaic device (FTO/c-TiO_2/SnO_2/$FAPbI_3$/BAI/Spiro-OMeTAD/Au) was constructed through sequential deposition of functional layers. Initially, patterned FTO substrates underwent ultrasonic cleaning in a detergent solution, deionized water, acetone, and ethanol (30 min each).

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Electron Transport Layer Preparation:

c-TiO_2 Layer: A precursor solution was formulated by diluting 1 mL titanium (IV) bis(2,4-glutamate) bis(2-propanolate) (75% isopropanol) with 19 mL anhydrous ethanol. The preheated FTO substrate (500°C) was coated via spray pyrolysis, followed by annealing at 500°C for 60 min and cooling to ambient temperature. Oxygen plasma treatment (25 min) enhanced surface activity.

SnO_2 Layer: A PAA-modified SnO_2 colloid precursor (20× diluted with H_2O, mixed with 1 mg/mL PAA at 80°C for 3 h) was spin-coated (4,000 rpm, 30 s) onto c-TiO_2. Post-annealing at 160°C for 30 min, substrates received oxygen plasma treatment (8 min) prior to transferring to a glovebox.

Perovskite Active Layer Deposition: The $FAPbI_3$ precursor solution—containing PbI_2 (705.2 mg), FAI (240.8 mg), MACl (33.8 mg), and $MAPbBr_3$ (5.8 mg) in DMF: DMSO (8:1)—was spin-coated using a two-step protocol: 1,000 rpm (10 s) and 6,000 rpm (30 s). During the second step, 100 μL chlorobenzene was dripped for film passivation. Thermal annealing at 120°C for 60 min yielded crystallized perovskite films.

Fabrication of the Passivation Layer: A BAI solution was prepared by dissolving 3 mg of butylammonium iodide (BAI) in 10 mL of isopropanol under vigorous stirring at room temperature overnight. The FTO substrate with the pre-deposited perovskite layer was immediately transferred into a nitrogen glovebox. The substrate was placed on a spin coater, and 40 μL of the BAI solution was uniformly spread over the perovskite surface using a pipette. Subsequently, spin-coating was performed at 4000 rpm for 30 s to form the passivation layer.

Hole Transport Layer & Electrode Integration: TCBZ-27 (73 mg/mL in chlorobenzene) doped with 29 μL tBP and 17.5 μL Li-TFSI (520 mg/mL) spin-coated (3,000 rpm, 30 s) onto the perovskite layer. Finally, a 100-nm Au electrode was thermally evaporated to complete the device architecture.

3. Characterization and measurement

Hole mobility was measured by using the space-charge-limited current (SCLC) method with the device structure of ITO/PEDOT: PSS/HTM/Au[1].

$$J = \frac{9\mu e_0 z_r V^2}{8d^3}$$

where μ is the hole mobility, ε_0 is the dielectric constant of the vacuum, ε_r is the dielectric constant of the hole-transporting material, V is the bias voltage applied to the device, d is the film thickness of the hole-transporting material.

The electrical conductivities of the HTM films were determined by using two-probe electrical conductivity measurements. The electrical conductivity (σ) was calculated by using the following equation:

$$\sigma = W/(RLD)$$

where L is the channel length 10 mm, W is the channel width 2 mm, D is the film thickness of the TiO_2 and HTM, and R is the film resistance calculated from the gradients of the curves.

The electrochemical impedance spectroscopy (EIS) was performed using a two-electrode system under dark conditions with electrochemical workstation (Zahner PP211). The EIS spectra were recorded in frequency from 100 mHz to 1 MHz with a bias voltage of 1.20 V.

For device characterization, all current-voltage (I-V) characteristics of the devices were measured under simulated AM1.5G irradiation (100 mW/cm^2) using a Xe lamp-based SS-F5-3A Solar Simulator. A Xe lamp equipped with an AM1.5G filter was used as the white light source. The light intensity was controlled with an NREL-calibrated Si solar cell with a KG-5 filter. The devices were measured both in reverse scan (1.2 V→-0.2 V, step 0.02 V) and forward scan (-0.2 V→1.2 V, step 0.02 V) with 2 ms delay time. The external quantum efficiency (EQE) was measured by a QE-R3011 measurement system.

4. Synthesis and Characterization of materials

The synthetic routes and molecular structures of the newly developed hole-transport materials (HTMs) TCBZ-27 and TCBZ-36 are shown in Scheme 1. Compounds 1, 2, 4, and 5 were synthesized according to reported literature procedures[2-5]. All reactions involving air- or moisture-sensitive reagents were carried out under a dry nitrogen atmosphere using standard Schlenk techniques unless otherwise stated. Glassware was oven-dried at 120°C for at least 12 h and cooled under nitrogen prior to use. Organic solvents were dried and degassed before use when required. Both HTMs were obtained through a five-step synthetic route starting from low-cost commercial precursors, with an overall yield of approximately 33%.

Synthesis of Compound 3

A dry 100 mL two-neck round-bottom flask equipped with a magnetic stir bar was evacuated and backfilled with nitrogen three times. Under a nitrogen atmosphere, 6-

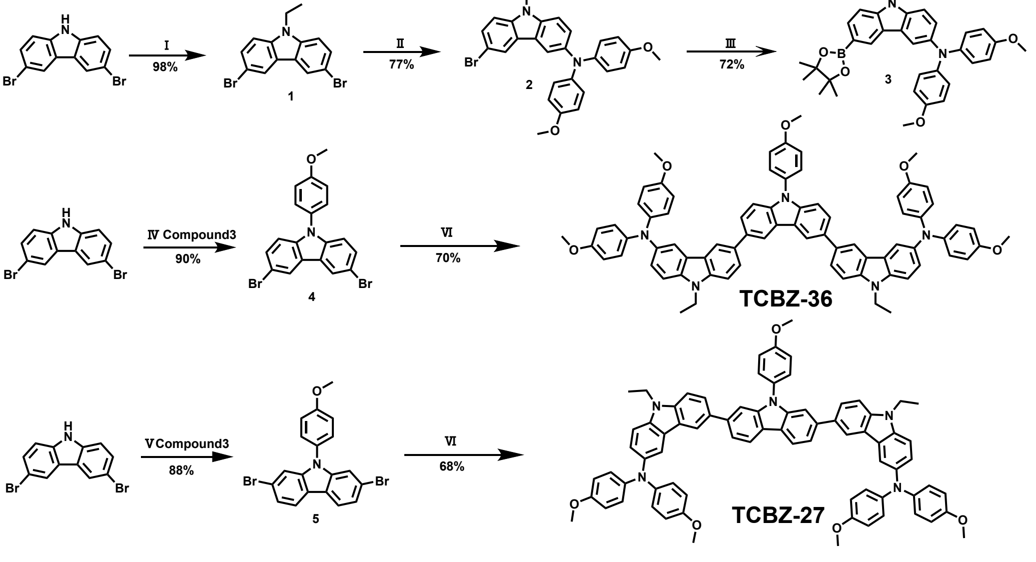
bromo-9-ethyl-N,N-bis(4-methoxyphenyl)-9H-carbazol-3-amine (compound 2, 1.50 g, 2.99 mmol), bis(pinacolato)diboron (1.14 g, 4.48 mmol, 1.5equiv), potassium acetate (KOAc, 880 mg, 8.97 mmol, 3.0equiv), and Pd(dppf)Cl_2(70 mg, 0.09 mmol, 3 mol%) were added to the flask. Anhydrous 1,4-dioxane (20 mL, reaction concentration≈ 0.15 M with respect to compound 2) was degassed by bubbling nitrogen for 20 min and then added to the reaction mixture via syringe. The resulting suspension was heated to 100°C and stirred under nitrogen for 24 h. After completion of the reaction (monitored by TLC), the mixture was cooled to room temperature and diluted with ethyl acetate (30 mL). The organic phase was washed with deionized water (3× 30 mL) to remove inorganic salts. The combined organic layers were dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. The crude product was passed through a short silica gel plug using ethyl acetate as the eluent. Final drying under vacuum afforded compound 3 as a white solid (1.18 g, 72%). 1H NMR (400MHz, DMSO) δ 8.27 (s, 1H), 7.78 (d, J = 2.2 Hz, 1H), 7.71 (dd, J = 8.2, 1.1 Hz, 1H), 7.53 (dd, J = 8.5, 5.4 Hz, 2H), 7.13 (dd, J = 8.7, 2.1 Hz, 1H), 6.91 (d, J = 8.7 Hz, 4H), 6.88 – 6.79 (m, 3H), 4.38 (q, J = 7.0 Hz, 2H), 3.70 (s, 5H), 1.30 (d, J = 7.0 Hz, 2H), 1.28 (s, 11H), 1.15 (s, 2H).

Synthesis of TCBZ-36

A dry 100 mL two-neck round-bottom flask was evacuated and refilled with nitrogen three times. Compound 3 (2.85 g, 5.19 mmol), 3,6-dibromo-9-(4-methoxyphenyl)-9H-carbazole (compound 4, 1.00 g, 2.33 mmol), and Pd(PPh_3)$_4$ (100 mg, 0.09 mmol, ~4 mol%) were added under nitrogen atmosphere. Degassed anhydrous toluene (20 mL, reaction concentration≈ 0.12 M with respect to compound 4) and saturated aqueous potassium carbonate solution (4 mL) were sequentially injected into the flask via syringe. The biphasic mixture was bubbled with nitrogen for an additional 20 min and then heated to 110°C with vigorous stirring for 24 h. After cooling to room temperature, the reaction mixture was diluted with ethyl acetate (30 mL). The organic phase was separated, and the aqueous layer was extracted with ethyl acetate (2× 20 mL). The combined organic layers were washed with deionized water (2× 30 mL), dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. The crude product was purified by silica gel column chromatography using petroleum ether/ethyl acetate (10:1, v/v) as the eluent. The desired fractions were collected and evaporated to afford TCBZ-36 as a white solid (1.82 g, 70%).1H NMR (400 MHz, DMSO) δ 8.78 – 8.73 (m, 1H), 8.48 (s, 1H), 7.97 – 7.92 (m, 1H), 7.92 – 7.85 (m, 1H), 7.85 – 7.79 (m, 1H), 7.66 (d, J = 8.6 Hz, 1H), 7.55 (dd, J = 8.7, 4.5 Hz, 2H), 7.33 (d, J = 8.5 Hz, 1H), 7.22 (d, J = 8.8 Hz, 1H), 7.18 – 7.10 (m, 1H), 6.90 (d, J = 8.6 Hz, 4H), 6.81 (d, J = 9.0 Hz, 4H), 4.43 (d, J = 8.0 Hz, 2H), 3.87 (s, 2H), 3.66 (s, 6H), 1.34 (t, J = 7.1 Hz, 3H), 1.30 – 1.20 (m, 0H), 1.20 (s, 1H), 0.88 – 0.70 (m, 4H). ^{13}C NMR (101 MHz, CDCl3) δ 158.84, 140.86, 130.55, 128.45, 125.80, 123.98, 123.29, 119.18, 118.86, 115.14, 109.98, 77.26, 55.63, 55.54, 33.62, 29.73, 1.06.

Synthesis of TCBZ-27

TCBZ-27 was synthesized following the same procedure as described for TCBZ-36, except that compound 5 was used instead of compound 4 in the final Suzuki coupling step. All reaction conditions, solvent volumes, catalyst loading, inert atmosphere control, work-up, and purification procedures were identical. After silica gel column chromatography (petroleum ether/ethyl acetate = 10:1, v/v), TCBZ-27 was obtained as a white solid (1.76 g, 68%).1H NMR (400 MHz, DMSO) δ 8.39 (s, 1H), 8.23 (d, J = 8.1 Hz, 1H), 7.96 (d, J = 2.1 Hz, 1H), 7.73 – 7.49 (m, 5H), 7.19 (d, J = 8.5 Hz, 1H), 7.12 (dd, J = 8.7, 2.2 Hz, 1H), 6.89 (d, J = 8.7 Hz, 4H), 6.81 (d, J = 9.0 Hz, 4H), 4.40 (d, J = 7.4 Hz, 2H), 3.85 (s, 1H), 3.69 (s, 5H), 1.31 (t, J = 7.1 Hz, 3H), 1.20 (s, 2H), 0.83 (s, 1H), 0.85 – 0.71 (m, 1H). 13C NMR (101 MHz, CDCl3) δ 158.94, 142.64, 130.39, 128.87, 123.25, 121.69, 120.38, 119.92, 119.46, 115.34, 108.16, 77.28, 55.59, 55.55, 29.75, 29.49, 22.87, 1.08.



Scheme S1. Synthetic routes and chemical structure of TCBZ-27 and TCBZ-36

I). NBS, THF, DMF, 0°C. II): Bis-(4-methoxyphenyl)-amine, Pd$_2$(dba)$_3$, Tri-tert-butylphosphine, Sodium tert-butoxide, Toluene, 100°C. III): bis(pinacolato)diboron, KOAc, Pd(dppf)Cl$_2$, 1,4-dioxane, 100 °C. IV): 4-Iodoanisole, CuI, K$_3$PO$_4$, 1,4-dioxane, 110 °C. V): 4-Iodoanisole, CuI, K$_3$PO$_4$, 1,4-dioxane, 110 °C. VI): K$_2$CO$_3$, Pd(pph$_3$)$_4$, Toluene, 110°C.

Table S1. Detailed calculation of cost for the synthesis of 1 g TCBZ-27.

	Commercially available chemicals	**Price**	**Dosage**	**Cost**
Step 1	9-Ethylcarbazole	25 g/177.75 RMB	0.97 g	6.88 RMB
	NBS	500 g/70 RMB	0.88 g	0.07 RMB
	DMF	500 mL/58 RMB	5 mL	0.12 RMB
	THF	500 mL/68 RMB	30 mL	4.08 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 2	Benzenamine	125 g/25 RMB	0.23 g	0.05 RMB
	Tris(dibenzylideneacetone)dipalladium	1 g/336 RMB	0.05 g	0.01 RMB
	Bis-(4-methoxyphenyl)-amine	25g/321 RMB	1.13g	14.5 RMB
	Sodium tert-butoxide	500g/111.7 RMB	0.95g	0.2 RMB
	Toluene	500 mL/24 RMB	40 mL	1.92 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 3	bis(pinacolato)diboron	500 g/950 RMB	1.02 g	1.94 RMB
	Pd(dppf)Cl$_2$	10 g/800 RMB	0.148 g	11.8 RMB
	KOAc	500 g/40 RMB	1.077 g	0.09 RMB
	1,4-dioxane	500 mL/58 RMB	30 mL	3.48 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 5	3,7-Dibromodibenzosulfolane	5 g/48.75 RMB	0.63g	6.14 RMB
	Hexamethyldisilazane	50ml/148.8 RMB	0.5ml	1.48 RMB
	Sodium tert-butoxide	500 g/111.7RMB	0.8	0.18 RMB
	THF	500 mL/68 RMB	20 mL	2.72 RMB
	4-methoxyaniline	100 g/46.5 RMB	0.2 g	0.09 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 6	Pd(pph$_3$)$_4$	10 g/679 RMB	0.054 g	3.73 RMB
	K$_2$CO$_3$	500 g/22 RMB	1 g	0.04 RMB
	Toluene	500 mL/24 RMB	20 mL	0.96 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Total cost：135.52 RMB (18.6 US$)				

Table S2. Detailed calculation of cost for the synthesis of 1 g TCBZ-36

	Commercially available chemicals	**Price**	**Dosage**	**Cost**
Step 1	9-Ethylcarbazole	25 g/177.75 RMB	0.97 g	6.88 RMB
	NBS	500 g/70 RMB	0.88 g	0.07 RMB
	DMF	500 mL/58 RMB	5 mL	0.12 RMB
	THF	500 mL/68 RMB	30 mL	4.08 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 2	Benzenamine	125 g/25 RMB	0.23 g	0.05 RMB
	Tris(dibenzylideneacetone)dipalladium	1 g/336 RMB	0.05 g	0.01 RMB
	Bis-(4-methoxyphenyl)-amine	25g/321 RMB	1.13g	14.5 RMB
	Sodium tert-butoxide	500g/111.7 RMB	0.95g	0.2 RMB
	Toluene	500 mL/24 RMB	40 mL	1.92 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 3	bis(pinacolato)diboron	500 g/950 RMB	1.02 g	1.94 RMB
	$Pd(dppf)Cl_2$	10 g/800 RMB	0.148 g	11.8 RMB
	KOAc	500 g/40 RMB	1.077 g	0.09 RMB
	1,4-dioxane	500 mL/58 RMB	30 mL	3.48 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 4	3,6-Dibromocarbazole	5 g/24 RMB	0.37g	1.78 RMB
	Hexamethyldisilazane	50ml/148.8 RMB	0.5ml	1.48 RMB
	Sodium tert-butoxide	500 g/111.7RMB	0.8	0.18 RMB
	THF	500 mL/68 RMB	20 mL	2.72 RMB
	4-methoxyaniline	100 g/46.5 RMB	0.2 g	0.09 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Step 6	$Pd(pph_3)_4$	10 g/679 RMB	0.054 g	3.73 RMB
	K_2CO_3	500 g/22 RMB	1 g	0.04 RMB
	Toluene	500 mL/24 RMB	20 mL	0.96 RMB
	Petroleum ether	500 mL/12 RMB	150 mL	3.6 RMB
	ethyl acetate	500 mL/27 RMB	250 mL	13.5 RMB
Total cost：131.17 RMB (18.3 US$)				

Table S3. Comparison of synthesis cost and commercial price of TCBZ-27 with some widely used HTMs.

Compound	Materials cost	Commercial price

	(US \$/1g)	(US \$/1g)
TCBZ-27	18.6	
TCBZ-36	18.3	
Spiro-OMeTAD	91.67[6]	500[7]

Table S4. Summarizes the optical and electrochemical properties of TCBZ-36 and TCBZ-27.

HTMs	λ_{int} [a] /nm	E_g [b] /eV	E_{HOMO} /eV	E_{LUMO} [c] /eV	Hole mobility /$cm^2 \cdot V^{-1} \cdot s^{-1}$	Conductivity /$S \cdot cm^{-1}$
TCBZ-36	384	3.23	-5.15	-1.92	2.48×10^{-4}	1.22×10^{-4}
TCBZ-27	408	3.04	-5.15	-2.11	3.25×10^{-4}	4.04×10^{-4}

[a] Starting absorption wavelength in CH_2Cl_2 solution: point of crossing of UV absorption spectrum and fluorescence emission spectrum.[b] Estimated from the UV onset absorption wavelength using the formula $E_g=1240/\lambda_{int.}$ [c] $E_{LUMO} = E_{HOMO} + E_g$.

Table S5. Best photovoltaic performance of PSCs based on different HTMs.

HTMs	*Voc*/V	*Jsc*/mA·cm^{-2}	*FF*/%	PCE/%
Spiro-OMeTAD	1.14	25.7	84.5	24.7
TCBZ-36	1.12	25.2	79.8	22.5
TCBZ-27	1.16	26.2	83.5	25.2

Table S6. TRPL fitting parameters for perovskite films and hole-transporting materials.

Sample	τ_1 (ns)	R_1	τ_2 (ns)	R_2	τ_{avg} (ns)
perovskite	25	0.13	425.66	0.87	373.57
Perovskite/TCBZ-36	8.49	0.84	232.8	0.16	44.38

Perovskite/TCBZ-27	5.97	0.63	40.04	0.37	18.64

Table S7. Key PV parameters (Voc, Jsc, FF, and PCE) based on TCBZ-27

TCBZ-27	*Voc*/V	*Jsc*/mA·cm^{-2}	*FF*/%	PCE/%
1	1.14	26.0	82.1	24.3
2	1.18	25.5	81.8	24.3
3	1.12	26.5	82.3	24.3
4	1.15	25.7	83.1	24.4
5	1.16	25.9	82.6	24.5
6	1.13	26.4	82.8	24.5
7	1.14	26.3	83.0	24.6
8	1.17	25.8	82.5	24.7
9	1.15	26.1	83.2	24.8
10	1.16	26.2	83.5	25.2

Table S8. Key PV parameters (Voc, Jsc, FF, and PCE) based on TCBZ-36

TCBZ-36	*Voc*/V	*Jsc*/mA·cm^{-2}	*FF*/%	PCE/%
1	1.12	25.2	79.8	22.5
2	1.13	25.0	79.5	22.1
3	1.11	25.3	79.2	21.8
4	1.14	24.8	78.9	21.7
5	1.12	24.7	79.3	21.5
6	1.09	25.5	78.5	21.3

7	1.10	25.4	78.7	21.5
8	1.15	24.5	78.3	21.2
9	1.08	25.1	78.1	21.1
10	1.11	24.6	77.9	21.0

Table S9. Key PV parameters (Voc, Jsc, FF, and PCE) based on Spiro-OMeTAD

Spiro-OMeTAD	*Voc*/V	*Jsc*/mA·cm^{-2}	*FF*/%	PCE/%
1	1.14	25.7	84.5	24.7
2	1.15	25.5	83.2	24.2
3	1.13	25.8	82.8	23.9
4	1.16	24.9	83.0	23.7
5	1.12	25.9	82.5	23.6
6	1.14	24.8	82.7	23.5
7	1.11	26.0	82.1	23.4
8	1.10	25.6	81.8	23.2
9	1.13	24.7	82.3	23.1
10	1.12	25.3	81.5	22.7

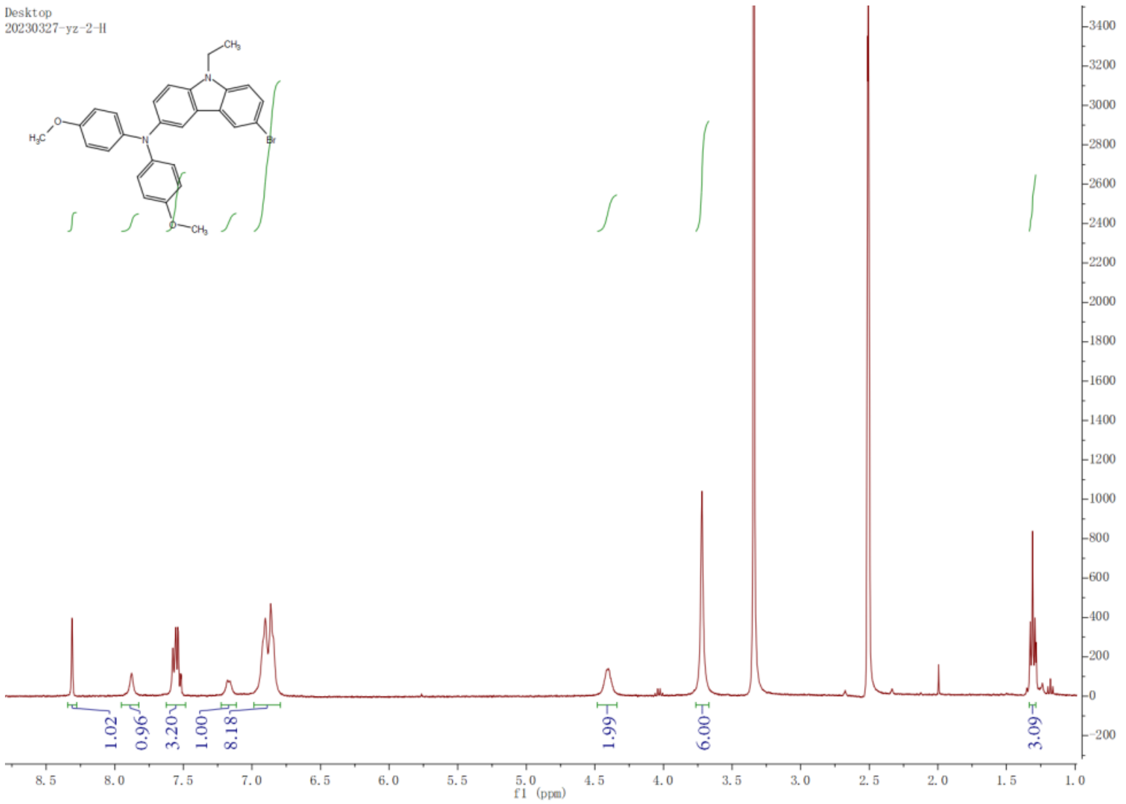


Figure S1. ^{1}H NMR spectrum of compound 2 recorded in DMSO-d_6

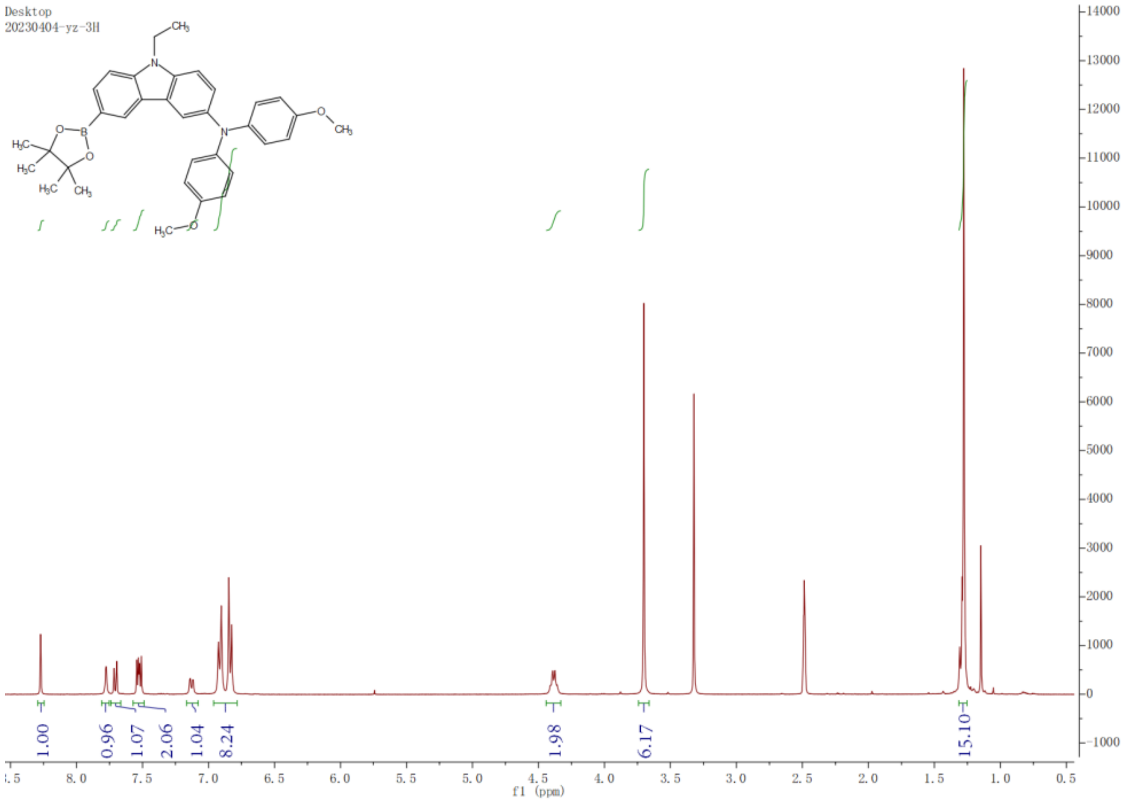


Figure S2. ^{1}H NMR spectrum of compound 3 recorded in DMSO-d_6

Supporting Information

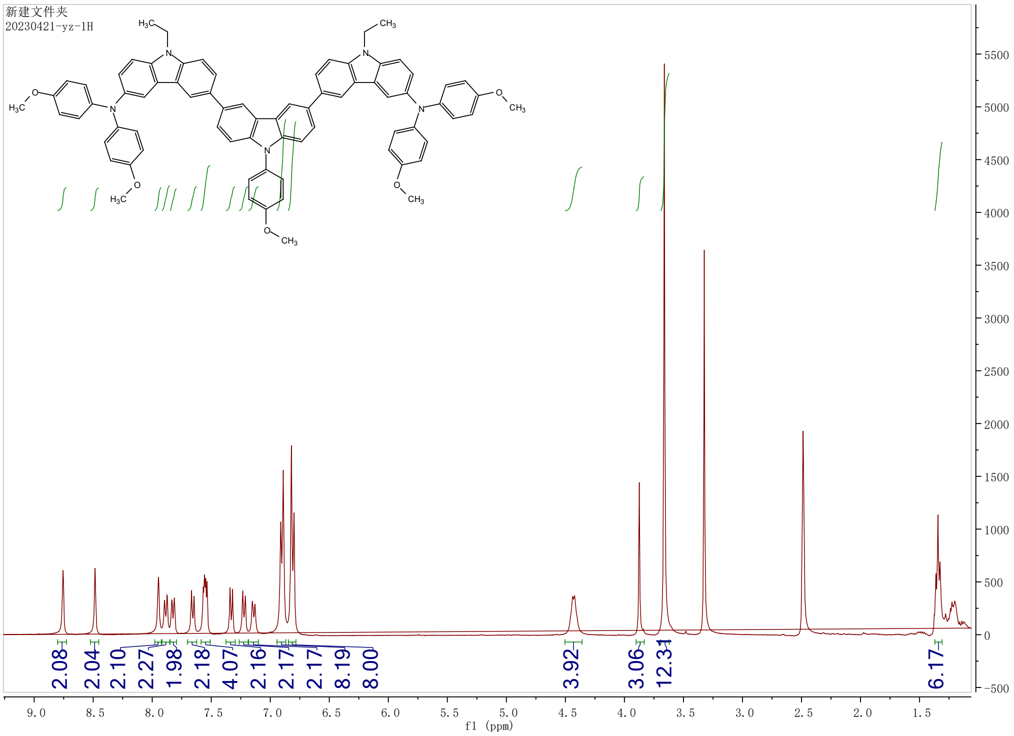


Figure S3. ^{1}H NMR spectrum of TCBZ-36 recorded in DMSO-d_6

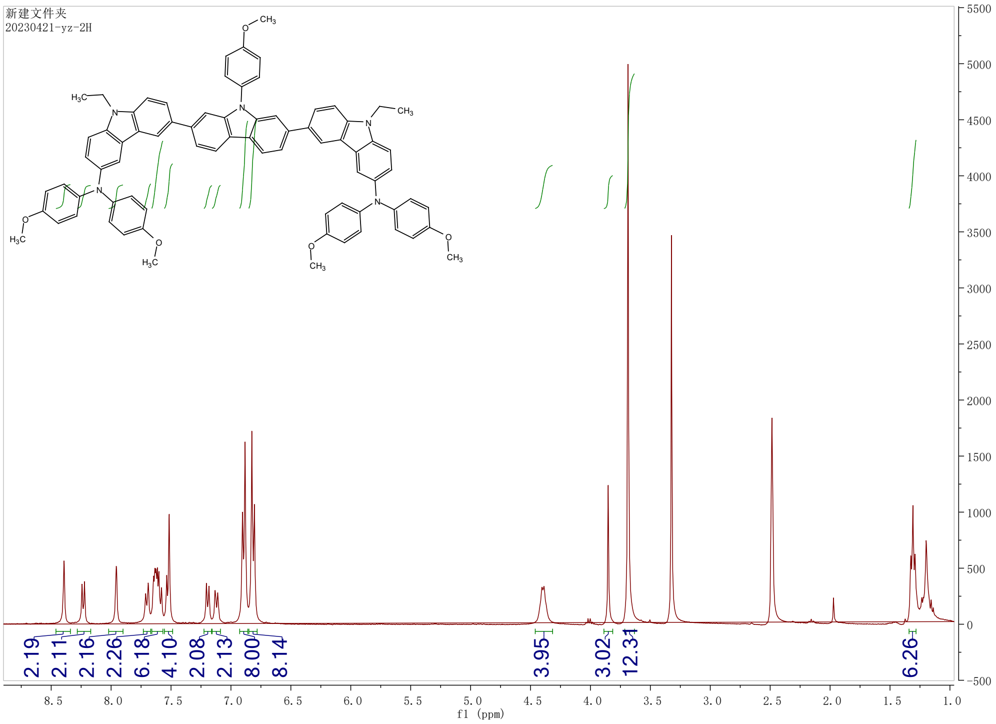


Figure S4. ^{1}H NMR spectrum of TCBZ-27 recorded in DMSO-d_6

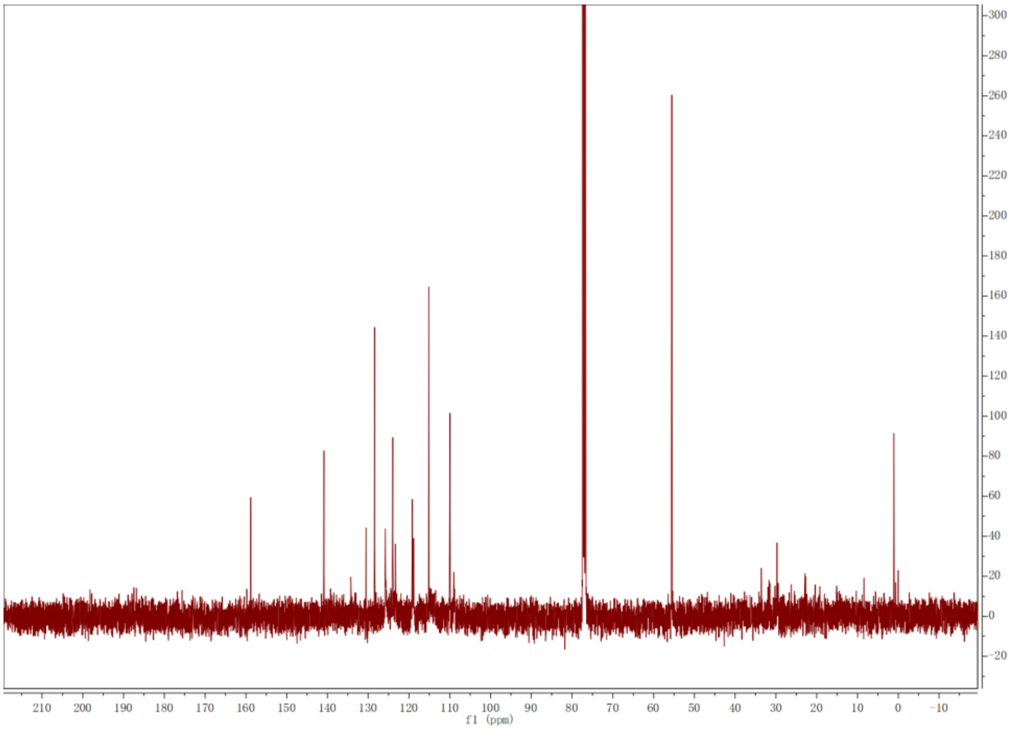


Figure S5. ^{13}C NMR spectrum of TCBZ-36 recorded in CDCl$_3$

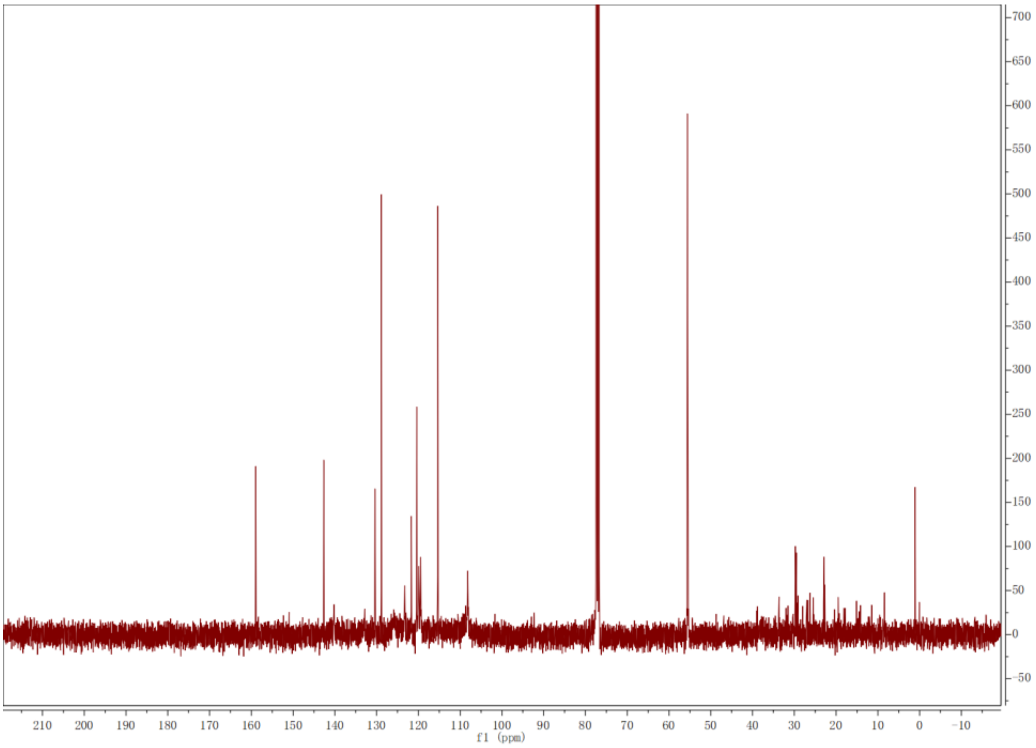


Figure S6. ^{13}C NMR spectrum of TCBZ-27 recorded in CDCl$_3$

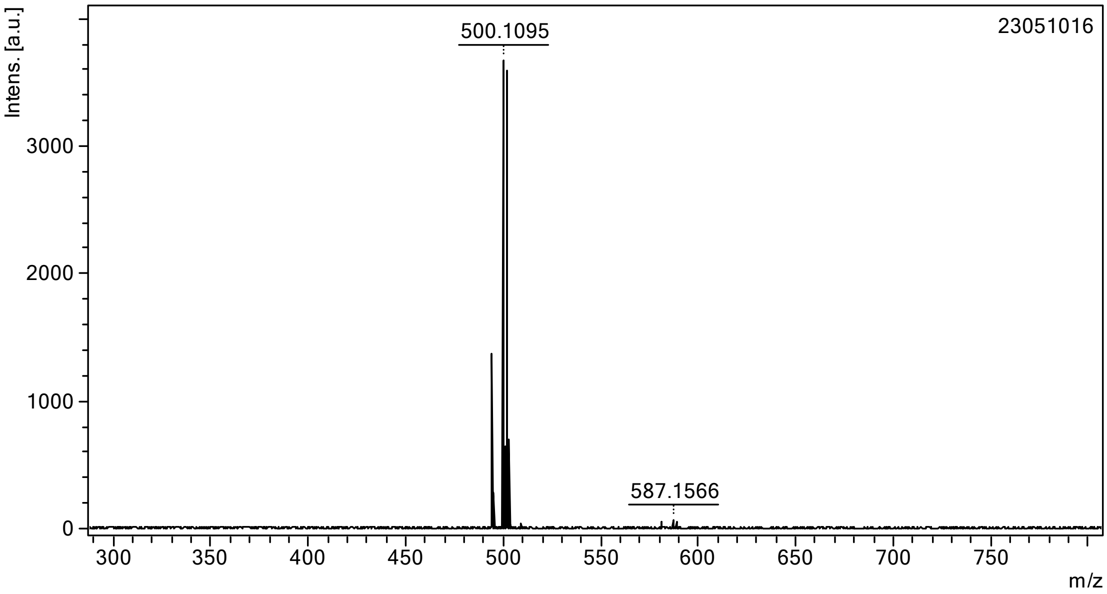


Figure S7. HR-MS of compound 2

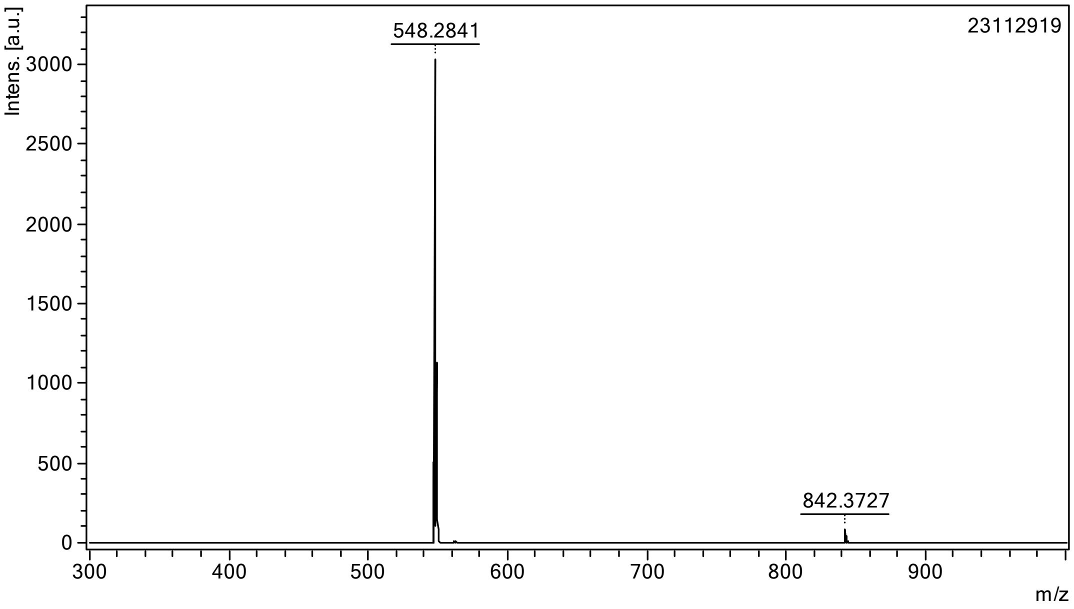


Figure S8. HR-MS of compound 3

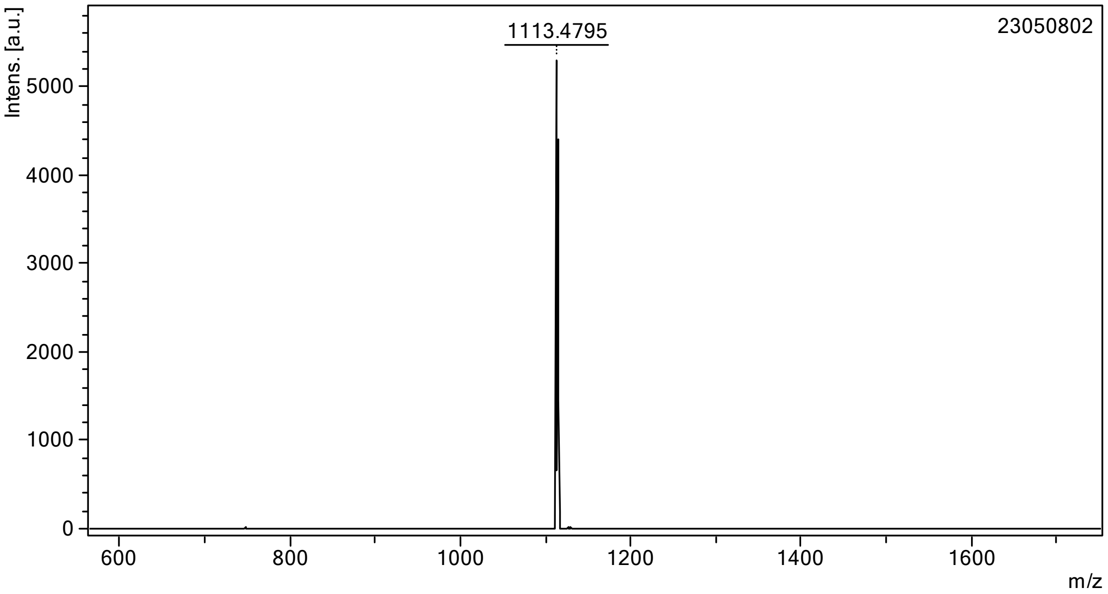


Figure S9. HR-MS of TCBZ-36

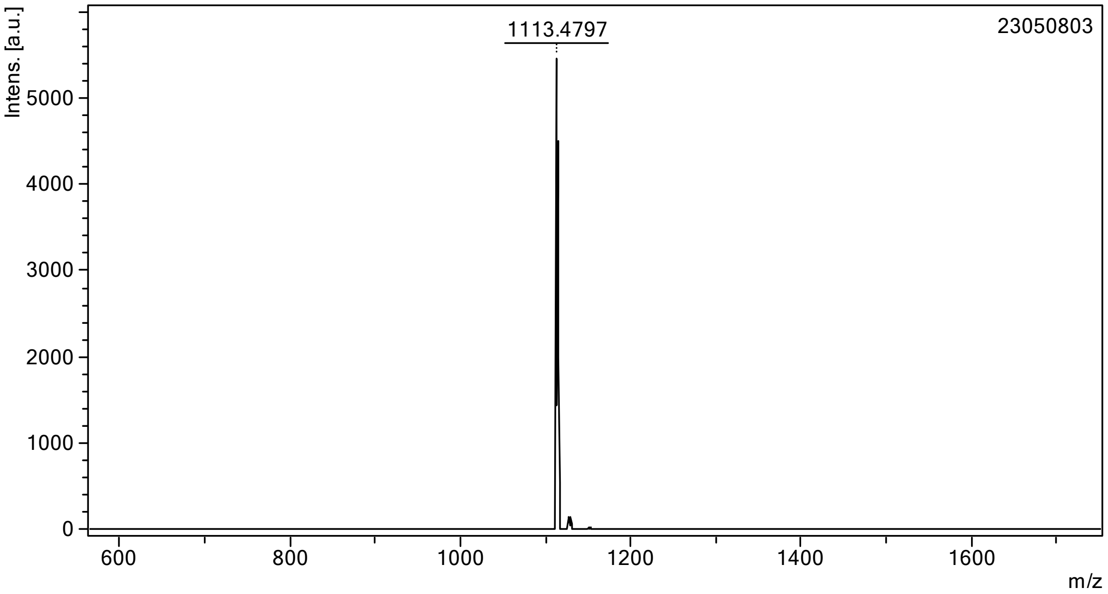


Figure S10. HR-MS of TCBZ-27

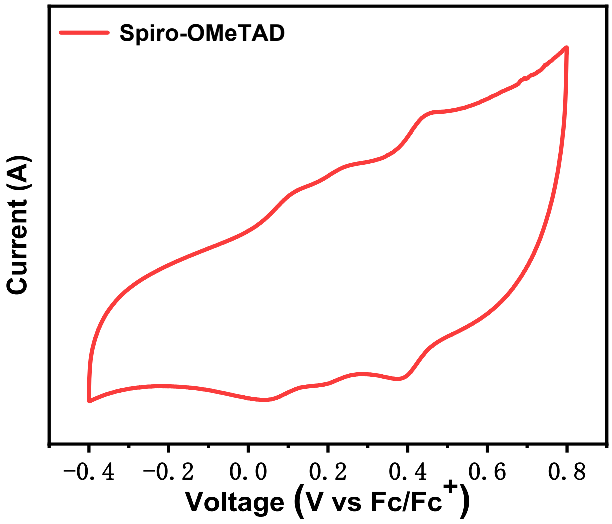


Figure S11. Cyclic voltammetry of Spiro-OMeTAD in CH_2Cl_2 solution.

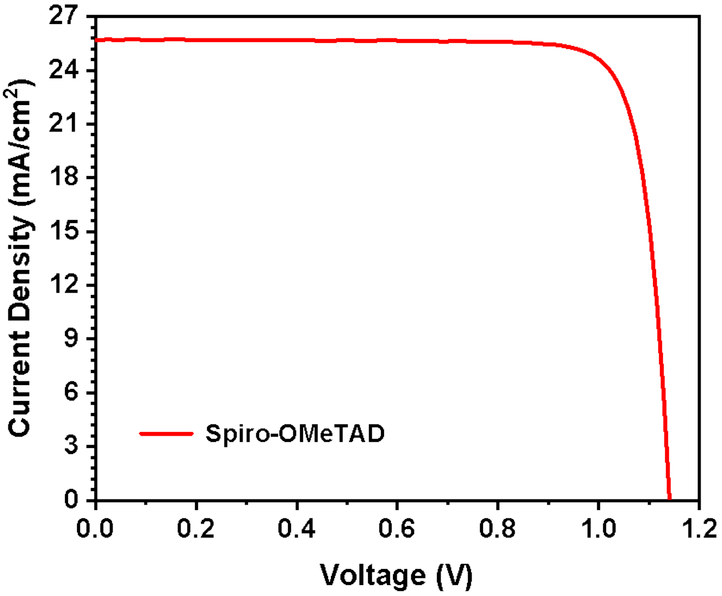


Figure S12. Current-voltage (*J*-*V*) curves for PSCs based on Spiro-OMeTAD.

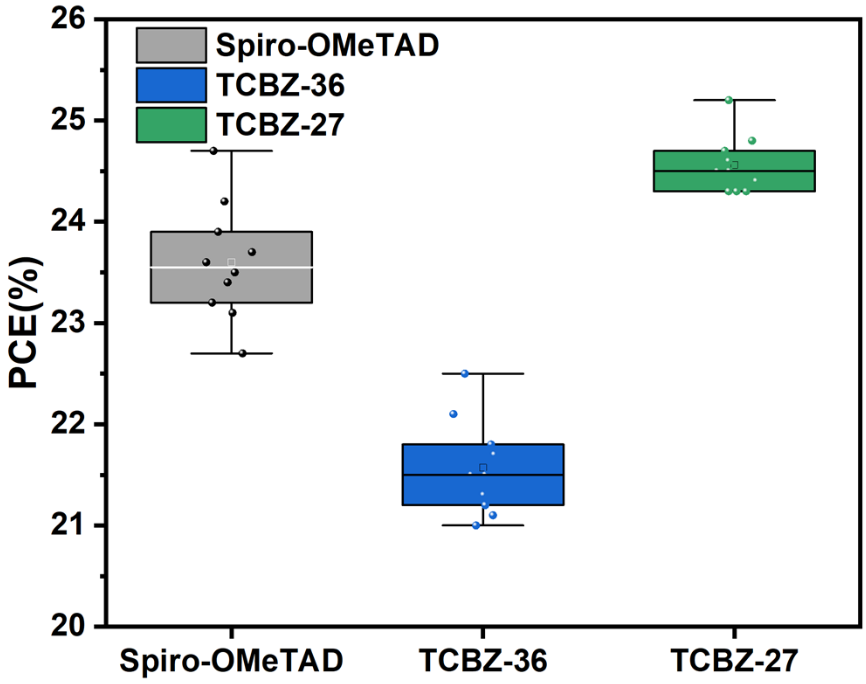


Figure S13. PCE based on at least 10 devices.

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