Supporting Information

Geometry Engineering of Multiple Resonance Core *via* Phenyl-Embedded Strategy toward Highly Efficient Narrowband Blue OLEDs

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I. General Remarks

NMR spectra were recorded on an Agilent 400-MR DD2 spectrometer. The ^{1}H NMR (400 MHz) chemical shifts were measured relative to $CDCl_3$ or DMSO-d_6 as the internal reference (DMSO-d_6: δ = 2.50 ppm; $CDCl_3$: δ = 7.26 ppm). The ^{13}C NMR (100 MHz) chemical shifts were given using $CDCl_3$ or DMSO-d_6 as the internal standard (DMSO-d_6: δ = 39.52 ppm; $CDCl_3$: δ = 77.16 ppm). High-resolution mass spectra (HRMS) were obtained with a Shimadzu LCMS-IT-TOF (ESI). X-Ray single-crystal diffraction data were collected on a Bruker D8 VENTURE single crystal diffractometer. Absorbtion spectra were measured on a HITACHI U-2910. Fluorescence spectra and photoluminescence quantum yield were collected on a Horiba Jobin Yvon-Edison Fluoromax-3 fluorescence spectrometer with a calibrated integrating sphere system with the excitation wavelength of 365 nm. Phosphorescence spectra were collected on a HITACHI F-7100 fluorescence spectrophotometer and a Horiba Jobin Yvon-Edison Fluoromax-3 fluorescence spectrometer with the excitation wavelength of 365 nm. Transient photoluminescence decay spectra were obtained with Horiba Single Photon Counting Controller: FluoroHub and Horiba TBX Picosecond Photon Detection with the excitation wavelength of 365 nm. Thermogravimetric analysis (TGA) was carried out using DTG-60(H) at a rate of 10 ^{o}C/min under nitrogen atmosphere. Cyclic voltammogram (CV) were performed on LK2005A with a solution of tetrabutylammonium hexafluorophosphate (Bu_4NPF_6, 0.1 M) in dichloromethane (DCM) as electrolyte and ferrocene/ferrocenium (Fc/Fc^{+}) as standard. Three-electrode system (Ag/Ag^{+}, platinum wire and glassy carbon electrode as reference, counter and work electrode respectively) was used in the CV measurement. HPLC analysis was conducted on a Shimadzu Prominence Modular HPLC system. HPLC traces were performed using a Daicel analytical column in hexane and isopropyl alcohol.

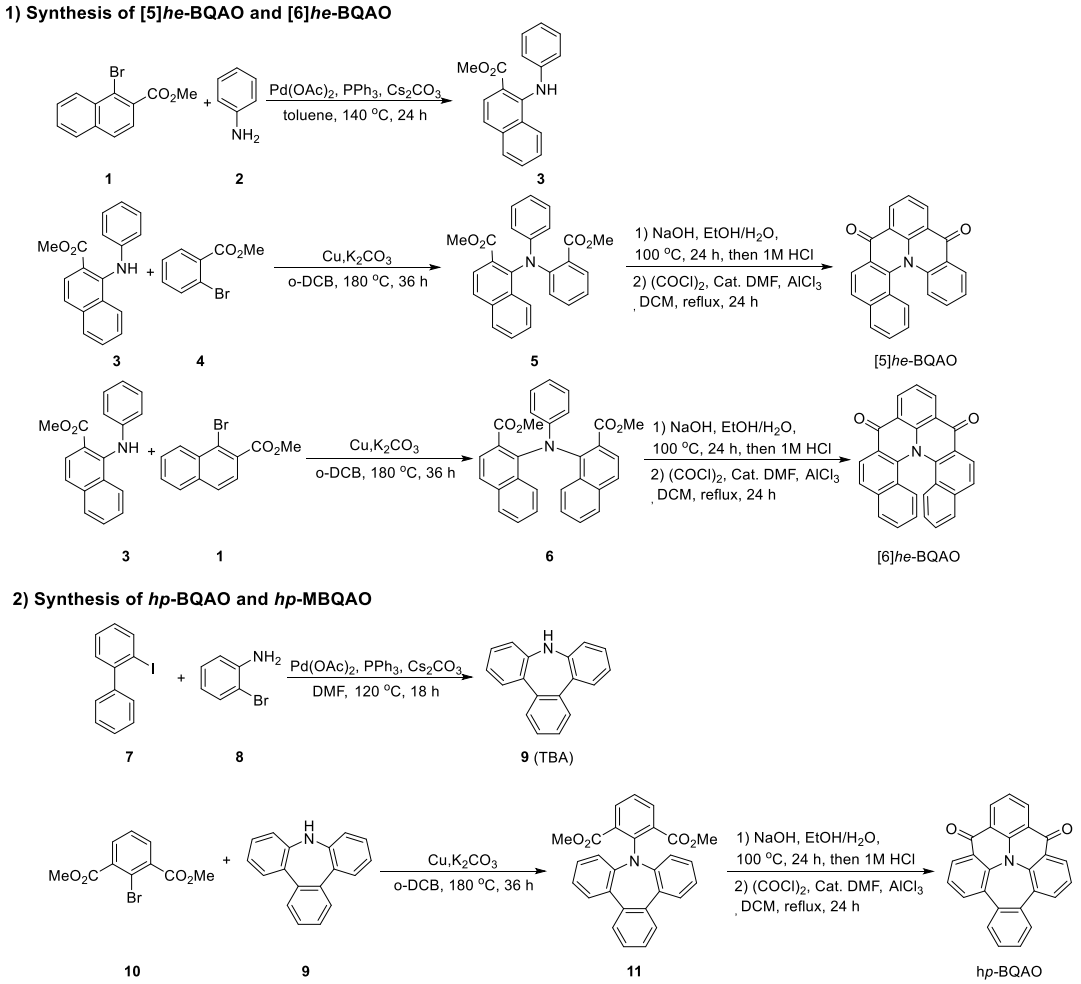
All commercially available reagents and chemicals were used as received without further purification. Unless otherwise noted, all reactions were carried out using Schlenk techniques under a nitrogen atmosphere. The solvents were dried and purified using an Innovative Technology PS-MD-5 Solvent Purification System. ***pe*-QAO**,[1] **QAO**[2] and 9*H*-tribenzo[*b,d,f*]azepine (**TBA**)[3] were prepared according to the literature procedures.

II. OLED Fabrication and Characterization

Indium-tin-oxide (ITO) coated glass with a sheet resistance of 15 Ω sq^{-1} was used as the anode substrate. Ahead of film deposition, ITO substrates were cleaned with alkaline detergent, boiled deionized water, deionized water in ultrasonic bath, dried in an oven, and finally treated with oxygen plasma for 10 min to enhance the surface work function of ITO anode. All organic layers were deposited with the rate of 0.1 $nm \cdot s^{-1}$ under high vacuum. The doped and co-doped layers were prepared by co-evaporating dopant and host material from two individual sources, and the doping concentrations were modulated by controlling the evaporation rates of dopant.

Current density-voltage-luminance (*J-V-L*) characteristics were measured by using KEYSIGHT B1500A. The luminance and electroluminescence spectra were collected with model DLM-100Z photometer and OPT2000 spectrophotometer, respectively.

III. Synthesis and Characterization



Scheme S1. Synthesis of **[5]*he*-BQAO**, **[6]*he*-BQAO** and ***hp*-BQAO**.

Synthesis of methyl 1-(phenylamino)-2-naphthoate (3)

A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **1** (1.0 mmol, 1.0 equiv), **2** (1.2 equiv), $Pd(OAc)_2$ (5.0 mol%), PPh_3 (10 mol%), Cs_2CO_3 (2.0 equiv), and toluene (2.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 140 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, diluted with 10.0 mL of DCM, filtered through a celite pad, and washed with 25.0-35.0 mL of DCM. The combined organic extracts were concentrated under reduced pressure and the resulting residue was purified *via* silica gel column chromatography (petroleum ether/DCM = 4/1, v/v) to afford the desired product **3** as a white solid (279.1 mg, 93% yield). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 3.93 (s, 3H), 6.81 (d, J = 7.6 Hz, 2H), 6.92 (t, J = 7.6 Hz, 1H), 7.15-7.20 (m, 2H), 7.28-7.33 (m, 1H), 7.50-7.54 (m, 2H), 7.82 (d, J = 8.0 Hz, 1H), 7.96 (d, J = 7.6 Hz, 1H), 8.00 (d, J = 8.8 Hz, 1H), 9.42 (s, 1H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 52.3, 115.3, 118.7, 121.4, 122.2, 125.4 126.2, 127.36, 127.43, 128.3, 128.4, 129.1, 136.8, 145.2, 146.0, 169.1 ppm. HRMS (ESI^+): calcd for $C_{18}H_{15}NNaO_2$ $[M+Na]^+$ 300.0995, found 300.0991.

Synthesis of methyl 1-((2-(methoxycarbonyl)phenyl)-(phenyl)amino)-2 -naphthoate (5)

A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **3** (1.0 mmol, 1.0 equiv), **4** (2.0 equiv), Cu (50 mol%), K_2CO_3 (2.5 equiv), and 1,2-dichlorobenzene (*o*-DCB) (2.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 180 °C in a pre-heated oil bath for 36 h. The reaction mixture was then cooled to room temperature, diluted with 10.0 mL of DCM, filtered through a celite pad, and washed with 25.0-35.0 mL of DCM. The combined organic extracts were concentrated under reduced pressure and the resulting residue was purified *via* silica gel column chromatography (petroleum ether/DCM = 1/4, v/v) to afford the desired product **5** as a white solid (377.6 mg, 87% yield). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 3.29 (s, 3H), 3.48 (s, 3H), 6.54 (d, J = 6.4 Hz, 1H), 6.83-6.88 (m, 2H), 6.99-7.09 (m, 3H), 7.16 (t, J = 7.2 Hz, 1H), 7.23 (t, J = 7.2 Hz, 1H), 7.43 (t, J = 7.6 Hz, 1H), 7.53 (t, J = 7.6 Hz, 1H), 7.66 (d, J = 6.4 Hz, 1H), 7.70 (d, J = 4.0 Hz, 1H), 7.84 (d, J = 8.4 Hz, 1H), 7.88 (d, J = 8.0 Hz, 1H), 8.47 (d, J = 8.4 Hz, 1H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ =

51.7, 52.2, 119.8, 122.1, 122.2, 122.3, 124.2, 125.2, 126.1, 126.4, 127.2, 127.6, 128.0, 128.2, 128.5, 129.26, 129.28, 130.9, 132.1, 132.5, 136.6, 143.3, 145.8, 149.2, 168.2, 168.7 ppm. HRMS (ESI^+): calcd for $C_{26}H_{21}NNaO_4$ $[M+Na]^+$ 434.1363, found 434.1365.

Synthesis of dimethyl 1,1'-(phenylazanediyl)bis(2-naphthoate) (6)

A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **3** (1.0 mmol, 1.0 equiv), **1** (2.0 equiv), Cu (50 mol%), K_2CO_3 (2.5 equiv), and *o*-DCB (2.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 180 °C in a pre-heated oil bath for 36 h. The reaction mixture was then cooled to room temperature, diluted with 10.0 mL of DCM, filtered through a celite pad, and washed with 25.0-35.0 mL of DCM. The combined organic extracts were concentrated under reduced pressure and the resulting residue was purified *via* silica gel column chromatography (petroleum ether/DCM = 1/6, v/v) to afford the desired product **6** as a white solid (280.7 mg, 58% yield). NMR spectra were difficult to interpret precisely due to large peak broadening, however, further conformation of structure can be made by HRMS and X-ray crystallography. The single crystal of **6** for X-ray crystallography was obtained by slow diffusion of hexane into a concentrated solution of **6** in DCM at room temperature. ^1H NMR (400 MHz, $CDCl_3$): δ = 3.02 (bs, 6H),6.90-6.95 (m, 3H), 7.14-7.20 (m, 4H), 7.39 (t, J = 7.6 Hz, 2H), 7.49 (bs, 2H), 7.69 (d, J = 8.4 Hz, 2H), 7.78 (d, J = 8.4 Hz, 2H), 8.02 (bs, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 51.9, 122.2, 125.8, 126.0, 127.1, 128.2, 129.1, 130.1, 136.3, 169.0 ppm. HRMS (ESI^+): calcd for $C_{30}H_{23}NNaO_4$ $[M+Na]^+$ 484.1519, found 484.1516.

Synthesis of [5]*he*-BQAO

A round bottom flask with a magnetic stirring bar was charged with **5** (1.0 mmol, 1.0 equiv), NaOH (20.0 equiv) and EtOH/H_2O = 1/1 (10.0 mL). The reaction mixture was heated at 100 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, and volatile was evaporated under reduced pressure. The remaining solid was dissolved in 20.0 mL of H_2O, and acidified with 1.0 M HCl (aq). The resulting mixture was extracted with ethyl acetate (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure. Then the residue was dissolved in 10.0 mL of DCM and 5 drops of *N*,*N*-dimethylformamide (DMF) was added. The

reaction mixture was stirred for 5 min at room temperature, and oxalyl chloride (10.0 equiv) was added in drops. After being stirred for 1 h, the reaction mixture was added with $AlCl_3$ (20.0 equiv) and refluxed for 24 h. Then the reaction mixture was cooled to room temperature and H_2O (5.0 mL) was added in drops. Subsquentlly, the resulting mixture was extracted with DCM (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure and the residue was purified *via* silica gel column chromatography (petroleum ether/DCM/ethyl acetate = 6/2/1, v/v/v) to afford the desired product **[5]*he*-BQAO** as a yellow solid (299.7 mg, 81% yield). The single crystal of **[5]*he*-BQAO** for X-ray crystallography was obtained by temperature gradient vacuum sublimation process. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.05 (d, J = 8.0 Hz, 1H), 7.26-7.42 (m, 3H), 7.49 (d, J = 8.8 Hz, 1H), 7.59 (t, J = 7.2 Hz, 1H), 7.67 (t, J = 7.6 Hz, 1H), 7.93 (d, J = 8.8 Hz, 1H), 7.97 (d, J = 7.6 Hz, 1H), 8.40-8.45 (m, 2H), 8.68-8.73 (m, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 121.6, 122.2, 124.0, 124.1, 124.4, 124.9, 125.1, 125.70, 125.76, 125.81, 127.0, 127.2, 127.6, 128.8, 129.1, 132.2, 132.4, 132.6, 136.5, 137.9, 140.7, 142.3, 178.8, 179.7 ppm. HRMS (ESI^+): calcd for $C_{24}H_{13}NNaO_2$ $[M+Na]^+$ 370.0838, found 370.0834.

Synthesis of [6]*he*-BQAO

A round bottom flask with a magnetic stirring bar was charged with **6** (1.0 mmol, 1.0 equiv), NaOH (20.0 equiv), and EtOH/H_2O = 1/1 (10.0 mL). The reaction mixture was heated at 100 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, and volatile was evaporated under reduced pressure. The remaining solid was dissolved in 20.0 mL of H_2O, and acidified with 1 M HCl (aq). The resulting mixture was extracted with ethyl acetate (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure. Then the residue was dissolved in 10.0 mL of DCM and 5 drops of DMF was added. The reaction mixture was stirred for 5 min at room temperature, and oxalyl chloride (10.0 equiv) was added in drops. After being stirred for 1 h, the reaction mixture was added with $AlCl_3$ (20.0 equiv) and refluxed for 24 h. Then the reaction mixture was cooled to room temperature and H_2O (5.0 mL) was added in drops. Subsquentlly, the resulting mixture was extracted with DCM (3 × 10.0 mL). The combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure and the

residue was purified *via* silica gel column chromatography (petroleum ether/DCM/ethyl acetate = 7/2/1, v/v/v) to afford the desired product **[6]*he*-BQAO** as a yellow solid (222.3 mg, 53% yield). The single crystal of **[6]*he*-BQAO** for X-ray crystallography was obtained by temperature gradient vacuum sublimation process. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 6.79 (t, J = 7.6 Hz, 2H), 6.91 (d, J = 8.8 Hz, 2H), 7.29 (t, J = 8.0 Hz, 2H), 7.74 (t, J = 7.6 Hz, 1H), 7.81 (d, J = 7.6 Hz, 2H), 7.90 (d, J = 8.4 Hz, 2H), 8.49 (d, J = 8.4 Hz, 2H), 8.75 (d, J = 7.6 Hz, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 122.1, 124.3, 124.5, 124.8, 125.1, 125.8, 125.9, 126.7, 128.4, 128.7, 132.0, 136.1, 140.9, 141.4, 179.5 ppm. HRMS (ESI^+): calcd for $C_{28}H_{15}NNaO_2$ $[M+Na]^+$ 420.0995, found 420.0991.

Synthesis of 9*H*-tribenzo[*b,d,f*]azepine (9)[3]

A flame-dried round bottom flask with a magnetic stirring bar was charged with **7** (10.0 mmol, 1.0 equiv), **8** (1.5 equiv), $Pd(OAc)_2$ (5.0 mol%), PPh_3 (10 mol%), Cs_2CO_3 (2.0 equiv), and DMF (30.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 120 °C in a pre-heated oil bath for 18 h. The reaction mixture was then cooled to room temperature, diluted with 10.0 mL of DCM, filtered through a celite pad, and washed with 40.0-60.0 mL of DCM. The combined organic extracts were concentrated under reduced pressure and the resulting residue was purified *via* silica gel column chromatography (petroleum ether/ethyl acetate = 9/1, v/v) to afford the desired product **9** as a white solid (2.23 g, 85% yield). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 5.21 (bs, 1H), 6.91 (d, J = 8.0 Hz, 2H), 7.13 (t, J = 7.2 Hz, 2H), 7.22 (t, J = 7.6 Hz, 2H), 7.43-7.50 (m, 6H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ =119.9, 124.3, 127.9, 128.6, 130.2, 130.3, 132.8, 139.5, 151.1 ppm. HRMS (ESI^+): calcd for $C_{18}H_{13}NNa$ $[M+Na]^+$ 266.0940, found 266.0938.

Synthesis of dimethyl 2-(9*H*-tribenzo[*b,d,f*]azepin-9-yl)isophthalate (11)

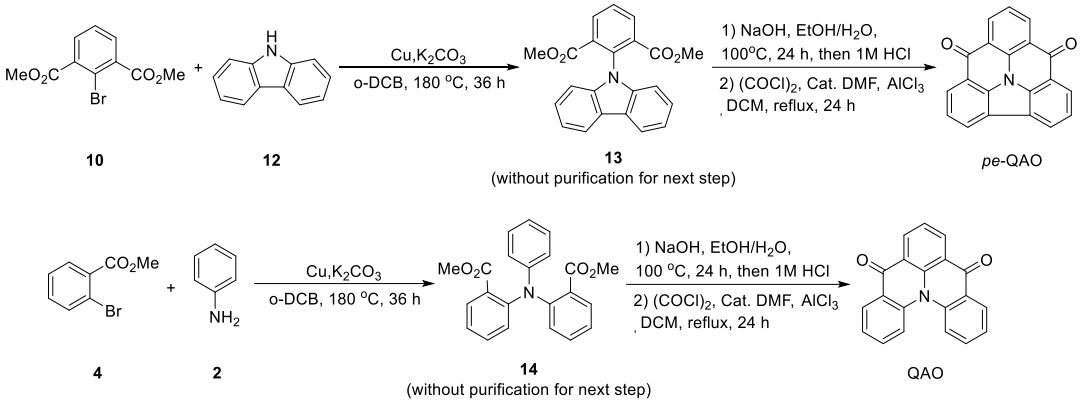
A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **10** (1.0 mmol, 1.0 equiv), **9** (2.0 equiv), Cu (50 mol%), K_2CO_3 (2.5 equiv), and *o*-DCB (2.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 180 °C in a pre-heated oil bath for 36 h. The reaction mixture was then cooled to room temperature, diluted with 10.0 mL of DCM, filtered through a celite pad, and washed with 25.0-35.0 mL of DCM. The combined organic extracts were concentrated under reduced pressure and the resulting

residue was purified *via* silica gel column chromatography (petroleum ether/ethyl acetate = 6/1, v/v) to afford the desired product **11** as a white solid (320.4 mg, 70% yield). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 3.10 (s, 6H), 6.76 (t, J = 7.6 Hz, 1H), 7.29-7.39 (m, 8H), 7.55-7.58 (m, 4H), 7.74-7.76 (m, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 51.3, 116.7, 119.7, 123.6, 126.9, 128.0, 128.4, 129.0, 130.4, 132.8, 137.2, 137.7, 144.1, 147.6, 167.8 ppm. HRMS (ESI$^+$): calcd for $C_{28}H_{21}NNaO_4$ $[M+Na]^+$ 458.1363, found 458.1366.

Synthesis of *hp*-BQAO

A round bottom flask with a magnetic stirring bar was charged with **11** (1.0 mmol, 1.0 equiv), NaOH (20.0 equiv), and EtOH/H_2O = 1/1 (10.0 mL). The reaction mixture was heated at 100 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, and volatile was evaporated under reduced pressure. The remaining solid was dissolved in 20.0 mL of H_2O, and acidified with 1 M HCl (aq). The resulting mixture was extracted with ethyl acetate (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure. Then the residue was dissolved in 10.0 mL of DCM and 5 drops of DMF was added. The reaction mixture was stirred for 5 min at room temperature, and oxalyl chloride (10.0 equiv) was added in drops. After being stirred for 1 h, the reaction mixture was added with $AlCl_3$ (20.0 equiv) and refluxed for 24 h. Then the reaction mixture was cooled to room temperature and H_2O (5.0 mL) was added in drops. Subsquentlly, the resulting mixture was extracted with DCM (3 × 10.0 mL). The combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure and the residue was purified *via* silica gel column chromatography (petroleum ether/DCM/ethyl acetate = 1/9/1, v/v/v) to afford the desired product ***hp*-BQAO** as a red solid (346.1 mg, 88% yield). The single crystal of ***hp*-BQAO** for X-ray crystallography was obtained by temperature gradient vacuum sublimation process. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 6.96-7.01 (m, 2H), 7.41-7.45 (m, 2H), 7.59 (t, J =7.6 Hz, 2H), 7.70 (t, J = 7.6 Hz, 1H), 7.85 (dd, J = 7.2 Hz, 2.0 Hz, 2H), 8.48 (dd, J = 8.0 Hz, 1.6 Hz, 2H), 8.88 (d, J = 8.0 Hz, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 123.2, 124.6, 125.5, 126.8, 127.5, 129.4, 131.2, 132.3, 134.1, 136.4, 138.5, 146.4, 175.6 ppm. HRMS (ESI$^+$): calcd for $C_{26}H_{13}NNaO_2$ $[M+Na]^+$ 394.0838, found 394.0841.

3) Synthesis of *pe*-QAO and QAO



Scheme S2. Synthesis of ***pe*-QAO** and **QAO**.[1,2]

Synthesis of dimethyl 2-(9*H*-carbazol-9-yl)isophthalate (13)

A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **10** (1.0 mmol, 1.0 equiv), **12** (2.0 equiv), Cu (50 mol%), K_2CO_3 (2.5 equiv), and *o*-DCB (2.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 180 °C in a pre-heated oil bath for 36 h. The reaction mixture was then cooled to room temperature, filtered through a short column of silica gel, and washed with DCM. The combined organic extracts were concentrated under reduced pressure to provide **13** and used in next step without further purification.

Synthesis of *pe*-QAO

A round bottom flask with a magnetic stirring bar was charged with **13** (1.0 mmol, 1.0 equiv), NaOH (20.0 equiv), and EtOH/H_2O = 1/1 (10.0 mL). The reaction mixture was heated at 100 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, and volatile was evaporated under reduced pressure. The remaining solid was dissolved in 20.0 mL of H_2O, and acidified with 1 M HCl (aq). The resulting mixture was extracted with ethyl acetate (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure. Then the residue was dissolved in 10.0 mL of DCM and 5 drops of DMF was added. The reaction mixture was stirred for 5 min at room temperature, and oxalyl chloride (10.0 equiv) was added in drops. After being stirred for 1 h, the reaction mixture was added with $AlCl_3$ (20.0 equiv) and refluxed for 24 h. Then the reaction mixture was cooled to room temperature and H_2O (5.0 mL) was added in drops. Subsquentlly, the resulting mixture was extracted with DCM (3 × 10.0 mL). The combined organic layers were dried over anhydrous

Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure and the residue was purified *via* silica gel column chromatography (petroleum ether/DCM/ethyl acetate = 6/3/1, v/v/v) to afford the desired product ***pe*-QAO** as a yellow solid (200.3 mg, 63% yield). The single crystal of ***pe*-QAO** for X-ray crystallography was obtained by temperature gradient vacuum sublimation process. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.69-7.73 (m, 3H), 8.39-8.42 (m, 4H), 8.76 (d, J = 7.6 Hz, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 120.6, 124.4, 124.5, 125.0, 125.3, 125.6, 128.3, 133.6, 134.5, 136.7, 178.7 ppm. HRMS (ESI^+): calcd for $C_{20}H_9NNaO_2$ $[M+Na]^+$ 318.0525, found 318.0529.

Synthesis of dimethyl 2,2'-(phenylazanediyl)dibenzoate (14)

A flame-dried Schlenk test tube with a magnetic stirring bar was charged with **2** (1.0 mmol, 1.0 equiv), **4** (4.0 equiv), Cu (50 mol%), K_2CO_3 (5.0 equiv), and *o*-DCB (3.0 mL). The reaction mixture was allowed to stir for 5 min at room temperature under an N_2 atmosphere, and then heated at 180 °C in a pre-heated oil bath for 36 h. The reaction mixture was then cooled to room temperature, filtered through a short column of silica gel, and washed with DCM. The combined organic extracts were concentrated under reduced pressure to provide **14** and used in next step without further purification.

Synthesis of QAO

A round bottom flask with a magnetic stirring bar was charged with **14** (1.0 mmol, 1.0 equiv), NaOH (20.0 equiv), and EtOH/H_2O = 1/1 (10.0 mL). The reaction mixture was heated at 100 °C in a pre-heated oil bath for 24 h. The reaction mixture was then cooled to room temperature, and volatile was evaporated under reduced pressure. The remaining solid was dissolved in 20.0 mL of H_2O, and acidified with 1 M HCl (aq). The resulting mixture was extracted with ethyl acetate (3 × 10.0 mL) and the combined organic layers were dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure. Then the residue was dissolved in 10.0 mL of DCM and 5 drops of DMF was added. The reaction mixture was allowed to stir for 5 min at room temperature, and then oxalyl chloride (10.0 equiv) was added in drops. The reaction mixture was stirred for 5 min at room temperature, and oxalyl chloride (10.0 equiv) was added in drops. After being stirred for 1 h, the reaction mixture was added with $AlCl_3$ (20.0 equiv) and refluxed for 24 h. Then the resulting mixture was extracted with DCM (3 × 10.0 mL). The combined organic layers were

dried over anhydrous Na_2SO_4. After filtration, the solvent was evaporated under reduced pressure and the residue was purified *via* silica gel column chromatography (petroleum ether/DCM/ethyl acetate = 4/2/1, v/v/v) to afford the desired product **QAO** as a yellow solid (236.8 mg, 74% yield). The single crystal of **QAO** for X-ray crystallography was obtained by temperature gradient vacuum sublimation process. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.47 (t, J = 7.2 Hz, 2H), 7.63 (t, J = 7.6 Hz, 1H), 7.68 (t, J = 7.2 Hz, 2H), 8.11 (d, J = 4.4 Hz, 2H), 8.46 (d, J = 6.4 Hz, 2H), 8.71 (d, J = 7.6 Hz, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 120.4, 123.5, 123.7, 125.3, 126.5, 127.9, 132.8, 133.0, 139.3, 139.8, 178.7 ppm. HRMS (ESI^+): calcd for $C_{20}H_{11}NNaO_2$ $[M+Na]^+$ 320.0682, found 320.0686.

IV. Method of Theoretical Calculations

All theoretical calculations were performed using Gaussian 09 serials software. The HOMO/LUMO distributions were calculated on the basis of crystal structures at the B3LYP/ 6-31g(d) level and visualized using Gaussview 5.0 software.

The singlet radiative decay rate (k_R^S), the intersystem crossing rate (k_{ISC}), the reverse intersystem crossing rate (k_{RISC}) and the triplet non-radiative decay rate (k_{NR}^T) could be estimated using the following equations.[3,4]

$$\Phi_P = C_1\Phi_{PL}$$

$$\Phi_d = C_2\Phi_{PL}$$

$$k_R^S = \Phi_P/\tau_p = \Phi_{PL}C_1/\tau_p$$

$$k_{ISC} = (1-\Phi_P)/\tau_p = (1-\Phi_{PL}C_1)/\tau_p$$

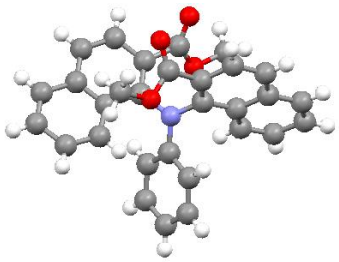
$$k_{RISC} = \Phi_d/(k_{ISC}\tau_p\tau_d\Phi_p) = C_2/[C_1\tau_d(1-\Phi_{PL}C_1)]$$

$$k_{NR}^T = 1/\tau_d-\Phi_P\, k_{RISC} = (1-\Phi_{PL})/[\tau_d(1-\Phi_{PL}C_1)]$$

Where Φ_p and Φ_d represent prompt and delayed fluorescence components and can be distinguished from the total Φ_{PL} by comparing the integrated intensities of prompt (C_1) and delayed components (C_2) in the transient PL spectra.

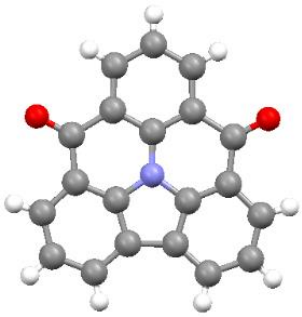
V. Crystal data

Table S1. Crystal Data for 6 [CCDC 2249621]



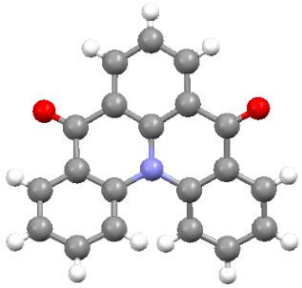
Identification code	**6**
Empirical formula	$C_{30}H_{23}NO_4$
Formula weight	461.49
Temperature/K	197.0
Crystal system	monoclinic
Space group	C2/c
Unit cell dimensions	a = 14.7721(8) Å, α = 90 deg. b = 14.0494(8) Å, β = 120.406(3) deg. c =12.9393(7) Å, γ = 90 deg.
Volume	2316.1(2) $Å^3$
Z	4
$\rho_{calc}mg/mm^3$	1.324
μ/mm^{-1}	0.088
F(000)	968.0
Crystal size/mm^3	0.41 × 0.3 × 0.21
Radiation	Mo Kα (λ = 0.71073)
2Θ range for data collection	4.316 to 55.042°
Index ranges	–19 ≤ h ≤ 19, –18 ≤ k ≤ 18, –16 ≤ l ≤ 16
Reflections collected	27302
Independent reflections	2655 [R_{int} = 0.0638, R_{sigma} = 0.0329]
Data/restraints/parameters	2655/0/161
Goodness–of–fit on F^2	1.048
Final R indexes [I>=2σ (I)]	R_1 = 0.0392, wR_2 = 0.0939
Final R indexes [all data]	R_1 = 0.0586, wR_2 = 0.1024
Largest diff. peak/hole / e $Å^{-3}$	0.17/–0.19

Table S2. Crystal Data for *pe*-QAO [CCDC 2249111]



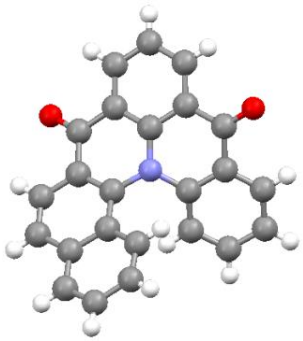
Identification code	***pe*-QAO**
Empirical formula	$C_{20}H_9NO_2$
Formula weight	295.28
Temperature/K	273.15
Crystal system	orthorhombic
Space group	$P2_12_12_1$
Unit cell dimensions	a = 3.7973(3) Å, α = 90 deg. b = 17.7163(14) Å, β = 90 deg. c =19.1364(15) Å, γ = 90 deg.
Volume	1287.38(18) $Å^3$
Z	4
$\rho_{calc}mg/mm^3$	1.523
μ/mm^{-1}	0.099
F(000)	608.0
Crystal size/mm^3	0.23 × 0.06 × 0.06
Radiation	Mo Kα (λ = 0.71073)
2Θ range for data collection	4.256 to 55.088°
Index ranges	$-4 \leq h \leq 4$, $-22 \leq k \leq 23$, $-24 \leq l \leq 24$
Reflections collected	7439
Independent reflections	2840 [R_{int} = 0.0758, R_{sigma} = 0.0896]
Data/restraints/parameters	2840/0/208
Goodness–of–fit on F^2	1.019
Final R indexes [I>=2σ (I)]	R_1 = 0.0590, wR_2 = 0.1039
Final R indexes [all data]	R_1 = 0.1299, wR_2 = 0.1322
Largest diff. peak/hole / e $Å^{-3}$	0.15/–0.21

Table S3. Crystal Data for QAO [CCDC 2248788]



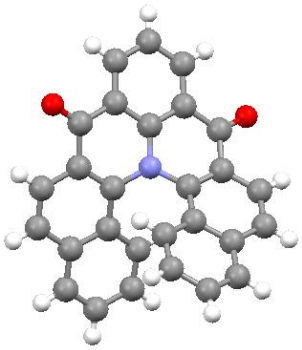
Identification code	**QAO**
Empirical formula	$C_{20}H_{11}NO_2$
Formula weight	297.30
Temperature/K	292.26(10)
Crystal system	monoclinic
Space group	C2/c
Unit cell dimensions	a = 9.1959(6) Å, α = 90 deg. b = 13.4702(10) Å, β = 91.216(5) deg. c =10.7519(6) Å, γ = 90 deg.
Volume	1331.54(15) $Å^3$
Z	4
ρcalcmg/mm^3	1.483
μ/mm^{-1}	0.777
F(000)	616.0
Crystal size/mm^3	0.35 × 0.3 × 0.2
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection	11.654 to 143.472°
Index ranges	–10 ≤ h ≤ 11, –15 ≤ k ≤ 16, –13 ≤ l ≤ 10
Reflections collected	3083
Independent reflections	1274 [R_{int} = 0.0286, R_{sigma} = 0.0307]
Data/restraints/parameters	1274/0/106
Goodness–of–fit on F^2	1.080
Final R indexes [I>=2σ (I)]	R_1 = 0.0719, wR_2 = 0.1623
Final R indexes [all data]	R_1 = 0.0742, wR_2 = 0.1673
Largest diff. peak/hole / e $Å^{-3}$	0.47/–0.55

Table S4. Crystal Data for [5]*he*-BQAO [CCDC 2248789]



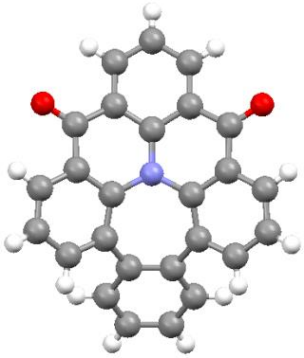
Identification code	**[5]*he*-BQAO**
Empirical formula	$C_{24}H_{13}NO_2$
Formula weight	347.35
Temperature/K	308.0
Crystal system	triclinic
Space group	P-1
Unit cell dimensions	a = 8.0822(16) Å, α = 61.646(6) deg. b = 11.0340(19) Å, β = 69.236(4) deg. c =11.143(2) Å, γ = 83.968(6) deg.
Volume	815.3(3) $Å^3$
Z	2
ρcalcmg/mm^3	1.415
μ/mm^{-1}	0.090
F(000)	360.0
Crystal size/mm^3	0.41 × 0.4 × 0.21
Radiation	Mo Kα (λ = 0.71073)
2Θ range for data collection	4.208 to 54.962°
Index ranges	–10 ≤ h ≤ 10, –14 ≤ k ≤ 14, –14 ≤ l ≤ 14
Reflections collected	22848
Independent reflections	3742 [R_{int} = 0.0624, R_{sigma} = 0.0465]
Data/restraints/parameters	3742/0/244
Goodness–of–fit on F^2	1.030
Final R indexes [I>=2σ (I)]	R_1 = 0.0429, wR_2 = 0.1031
Final R indexes [all data]	R_1 = 0.0650, wR_2 = 0.1158
Largest diff. peak/hole / e $Å^{-3}$	0.17/–0.17

Table S5. Crystal Data for [6]*he*-BQAO [CCDC 2248790]



Identification code	**[6]*he*-BQAO**
Empirical formula	$C_{28}H_{15}NO_2$
Formula weight	397.41
Temperature/K	293.9(2)
Crystal system	monoclinic
Space group	I2/a
Unit cell dimensions	a = 15.1670(4) Å, α = 90 deg. b = 10.9238(4) Å, β = 94.397(3) deg. c =11.5354(4) Å, γ = 90 deg.
Volume	1905.56(10) $Å^3$
Z	4
ρcalcmg/mm^3	1.385
μ/mm^{-1}	0.694
F(000)	824.0
Crystal size/mm^3	0.6 × 0.5 × 0.3
Radiation	Cu Kα (λ = 1.54184)
2Θ range for data collection	9.988 to 142.72°
Index ranges	–18 ≤ h ≤ 15, –10 ≤ k ≤ 13, –12 ≤ l ≤ 14
Reflections collected	5024
Independent reflections	1823 [R_{int} = 0.0347, R_{sigma} = 0.0250]
Data/restraints/parameters	1823/0/142
Goodness–of–fit on F^2	1.081
Final R indexes [I>=2σ (I)]	R_1 = 0.0606, wR_2 = 0.1541
Final R indexes [all data]	R_1 = 0.0637, wR_2 = 0.1589
Largest diff. peak/hole / e $Å^{-3}$	0.24/–0.41

Table S6. Crystal Data for *hp*-BQAO [CCDC 2249112]



Identification code	***hp*-BQAO**
Empirical formula	$C_{26}H_{13}NO_2$
Formula weight	371.37
Temperature/K	200.0
Crystal system	orthorhombic
Space group	$Pmn2_1$
Unit cell dimensions	a = 16.208(4) Å, α = 90 deg. b = 4.0870(10) Å, β = 90 deg. c =12.278(2) Å, γ = 90 deg.
Volume	813.3(3) $Å^3$
Z	2
$\rho_{calc}mg/mm^3$	1.516
μ/mm^{-1}	0.096
F(000)	384.0
Crystal size/mm^3	0.39 × 0.1 × 0.03
Radiation	Mo Kα (λ = 0.71073)
2Θ range for data collection	4.162 to 55.042°
Index ranges	–21 ≤ h ≤ 16, –4 ≤ k ≤ 5, –15 ≤ l ≤ 14
Reflections collected	4870
Independent reflections	1841 [R_{int} = 0.0506, R_{sigma} = 0.0556]
Data/restraints/parameters	1841/1/136
Goodness–of–fit on F^2	1.059
Final R indexes [I>=2σ (I)]	R_1 = 0.0448, wR_2 = 0.0901
Final R indexes [all data]	R_1 = 0.0632, wR_2 = 0.0994
Largest diff. peak/hole / e $Å^{-3}$	0.19/–0.24

VI. Additional Spectra and Data

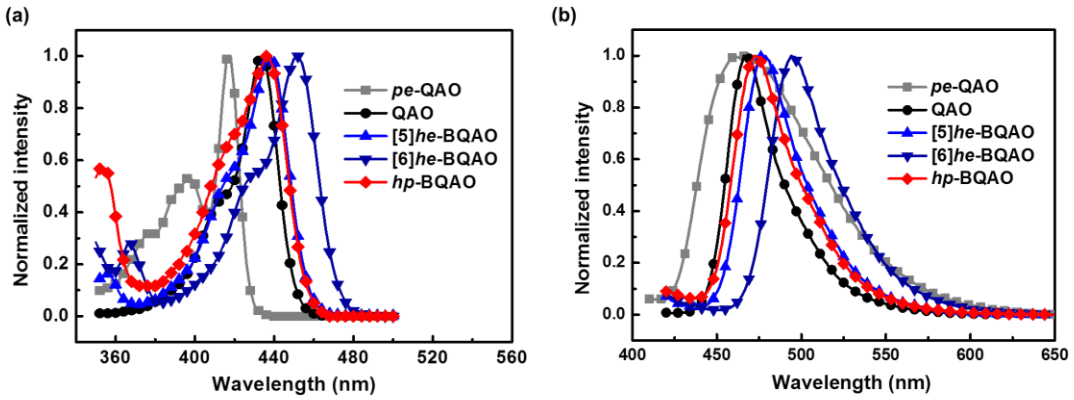


Fig. S1. (a) Absorption (Abs.) spectra at 298 K in toluene solutions at 1.0×10^{-5} M. (b) Photoluminescence spectra of the 3 wt% DCz-BTP host blended films at 298 K.

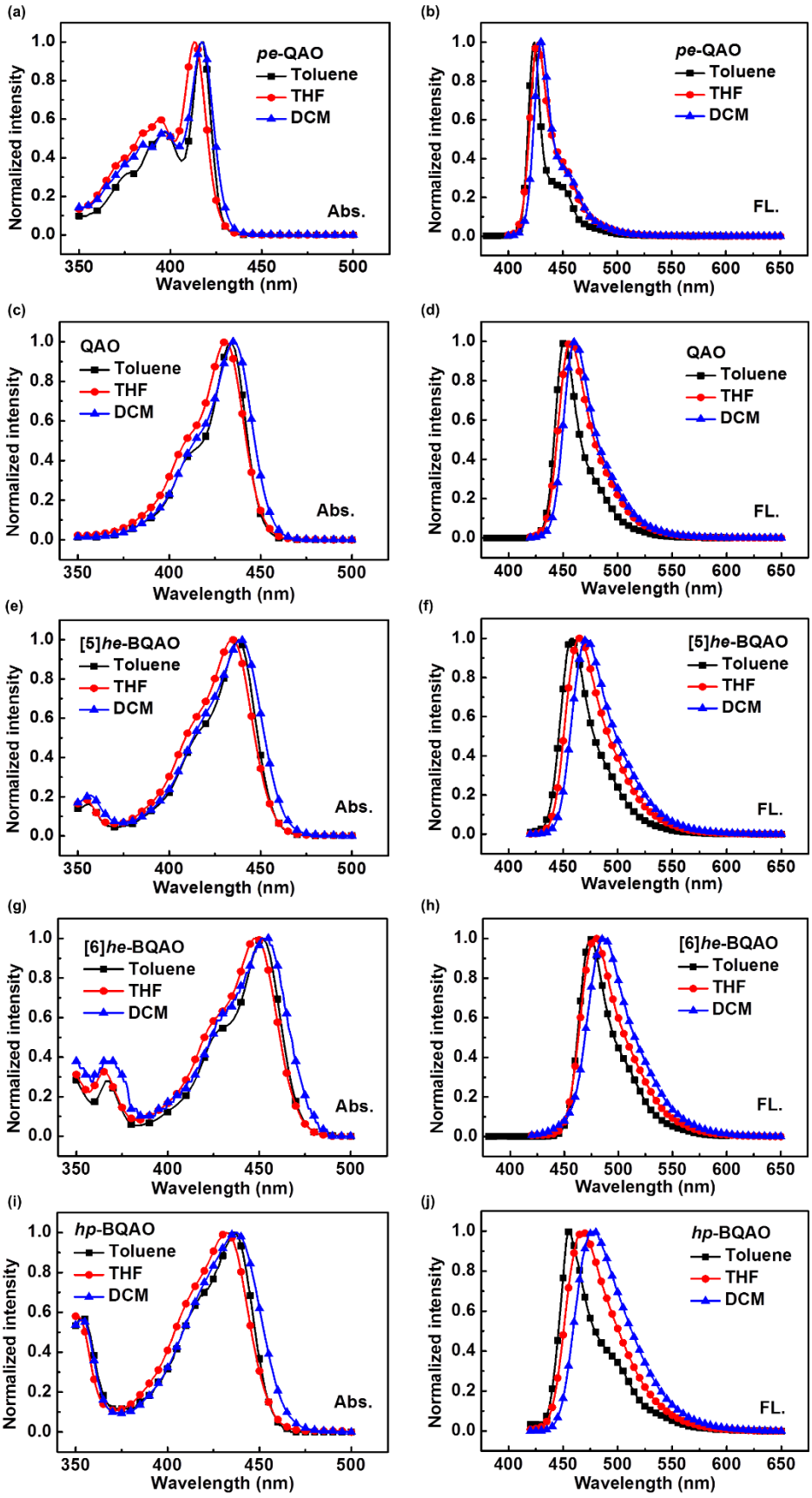


Fig. S2. Absorption spectra (Abs.) and fluorescence spectra (FL.) measured in the solvents with different polarities at 298 K.

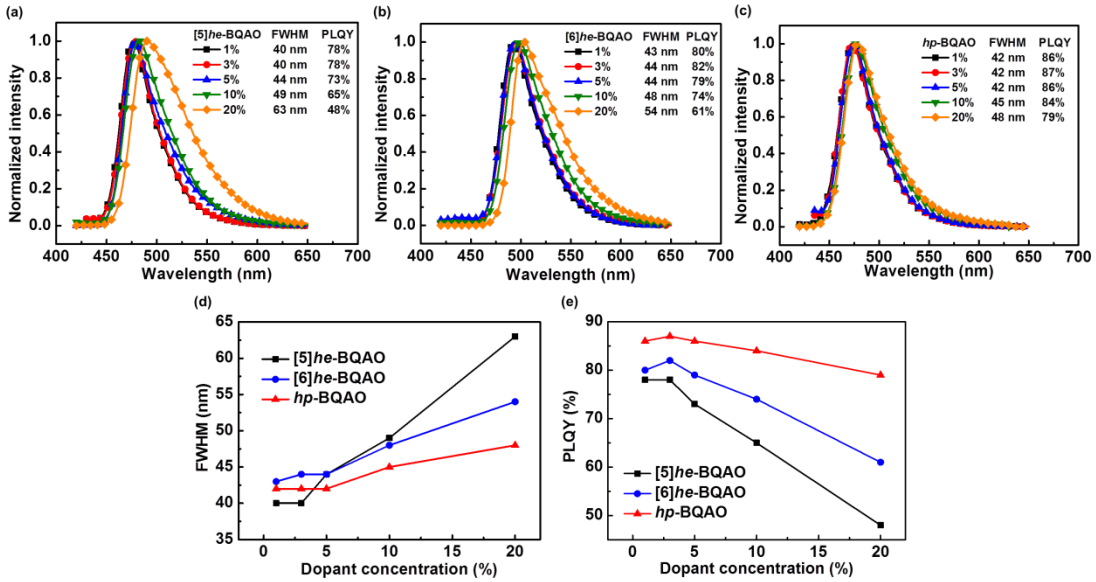


Fig. S3. Normalized photoluminescence spectra (PL), full-width at half-maximum (FWHM) and PL quantum yields (PLQY) of **[5]*he*-BQAO**, **[6]*he*-BQAO** and ***hp*-BQAO** doped in DCz-BTP films with different dopant concentrations of 1%, 3%, 5%, 10% and 20% at 298 K.

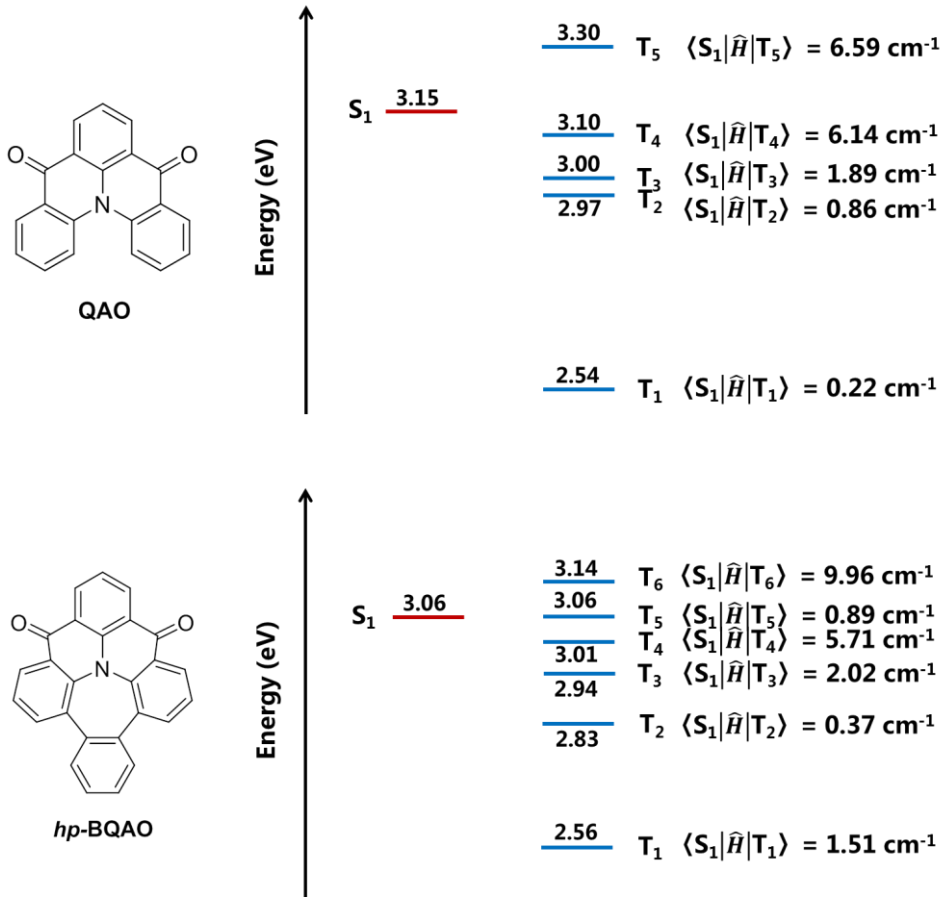


Fig. S4. Spin orbit coupling (SOC) analysis of **QAO** and ***hp*-BQAO**

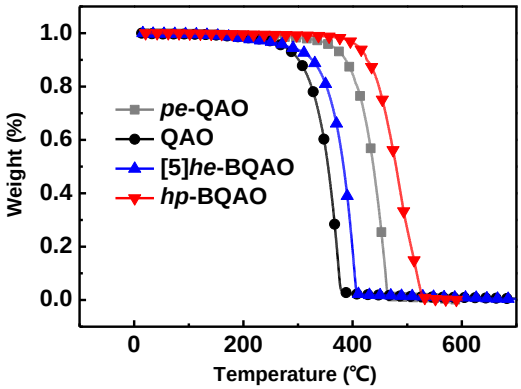


Fig. S5. TGA thermograms measured at a heating rate of 10 ºC/min.

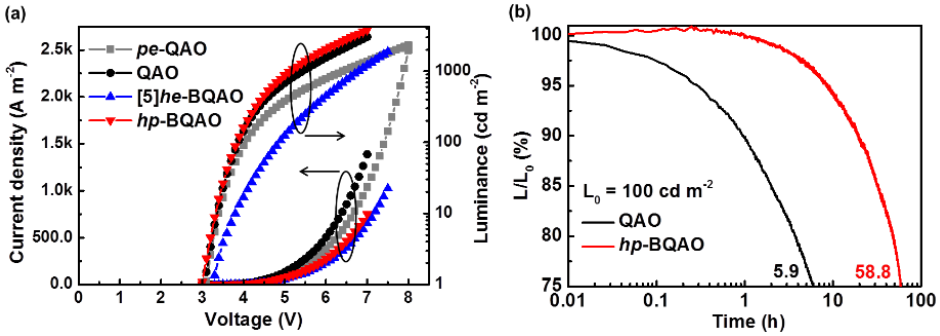


Fig. S6. (a) Current density-voltage-luminance curves of OLEDs. (b) Luminance (L)-deterioration curves of **QAO** and ***hp*-BQAO**-based OLEDs at the initial luminance (L_0) of 100 cd m^{-2}.

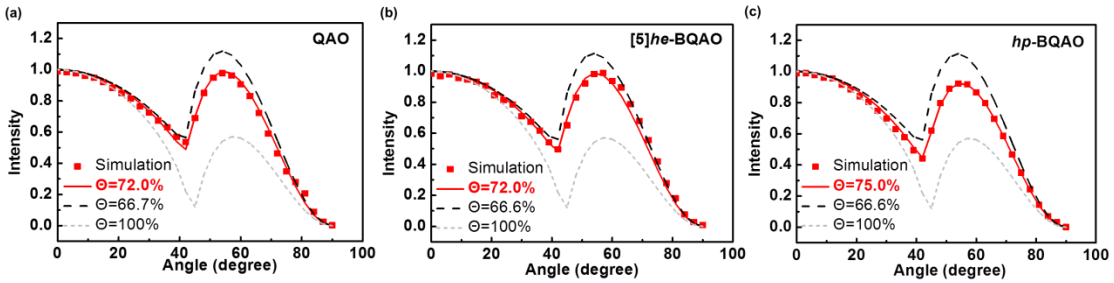


Fig. S7. Photoluminescence spectra intensity-emission angle curves with simulated horizontal dipole ratios (Θ) values in (a) **QAO**, (b) **[5]*he*-BQAO** and (c) ***hp*-BQAO**-doped DCz-BTP films (3 wt%).

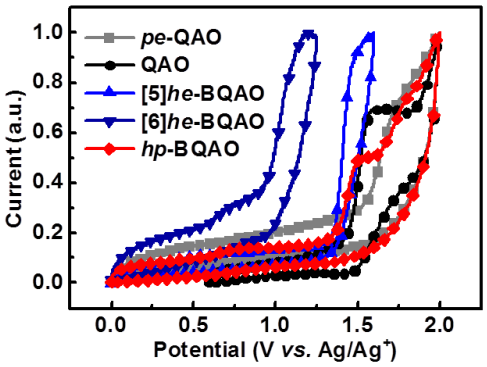


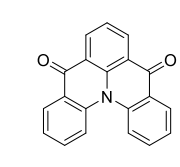
Fig. S8. Cyclic voltammograms measured in dry DCM containing 0.1 M of Bu_4NPF_6.

Table S7. Summary of photoluminescence spectra data in different polar solvents.

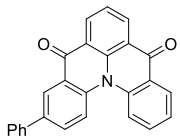
Compound	In toluene		In THF		In DCM	
	λ_{em} [nm]	FWHM [nm]	λ_{em} [nm]	FWHM [nm]	λ_{em} [nm]	FWHM [nm]
Pe-QAO	424	13	427	22	430	19
QAO	452	25	458	34	461	33
[5]*he*-BQAO	458	32	465	41	471	43
[6]*he*-BQAO	474	34	477	44	486	46
***hp*-BQAO**	456	34	469	50	478	53

Table S8. Summary of molecular structures, photophysical properties and OLED performances for blue N-CO MR-emitters.

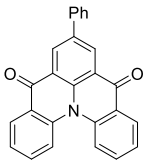
1. Molecular structures



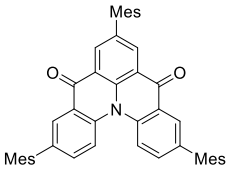
QAO



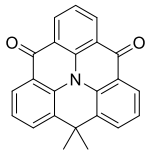
3-PhQAD



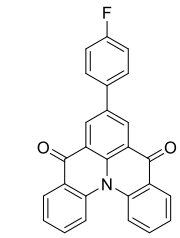
7-PhQAD



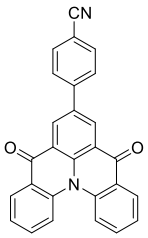
Mes_3DiKTa



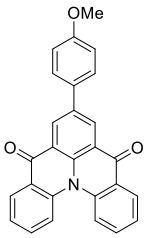
DQAO



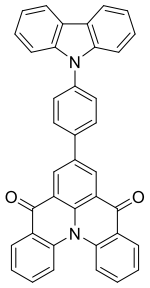
QA-PF



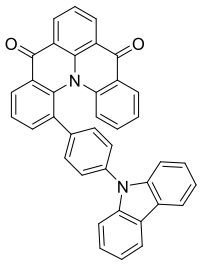
QA-PCN



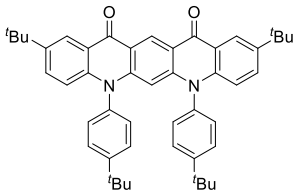
QA-PMO



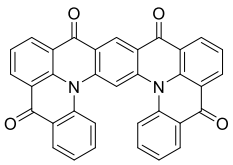
QA-PCZ



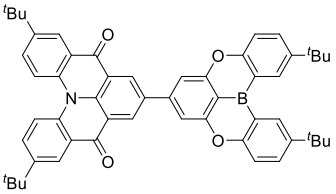
QAO-PhCZ



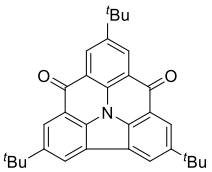
QA-1



QA-2



BOQAO



CZ2CO

2. Photophysical properties and OLED performances

Emitters	In solution			In film		In device			
	λ_{em} [nm]	FWHM [nm]	Φ_{PL} [%]	k_R^S [10^7 s^{-1}]	k_{RISC} [10^4 s^{-1}]	EL_{peak} [nm]	FWHM [nm]	EQE [%]	Ref.
***hp*-BQAO**	456	34	87	6.8	25.4	471	41	24.1	This work
QAO	466	32	72	-	-	468	39	19.4	5
3-PhQAD	466	30	73	4.0	1.2	480	44	19.1	6
7-PhQAD	464	22	68	3.6	0.6	472	34	18.7	6
Mes_3DiKTa	468	29	80	5.4[a]	3.1[a]	480	36	21.1	2

DQAO	465	33	59	-	-	472	34	15.2	7
QA-PF	465	23			4.9	474	27	16.8	8
QA-PCN	462	25			16.1	473	30	16.9	8
QA-PMO	475	27			2.2	484	27	15.0	8
QA-PCZ	471	29	89	9.6	5.0	482	29	17.5	8
QAO-PhCZ	461	29	68	4.3	-	467	36	14.0	9
QA-1	434	31	66	8.6	1.5	455	49	17.1	10
QA-2	444	22	97	1.5	85.0	463	37	19.0	10
BOQAO	474	28	99	3.2	2.4	484	32	21.8	11
CZ2CO	440	16	84	-	-	445/445[b]	23/26[b]	13.0/25.6[b]	1

[a] Measured in solution. [b] With TADF sensitizers.

VII. References

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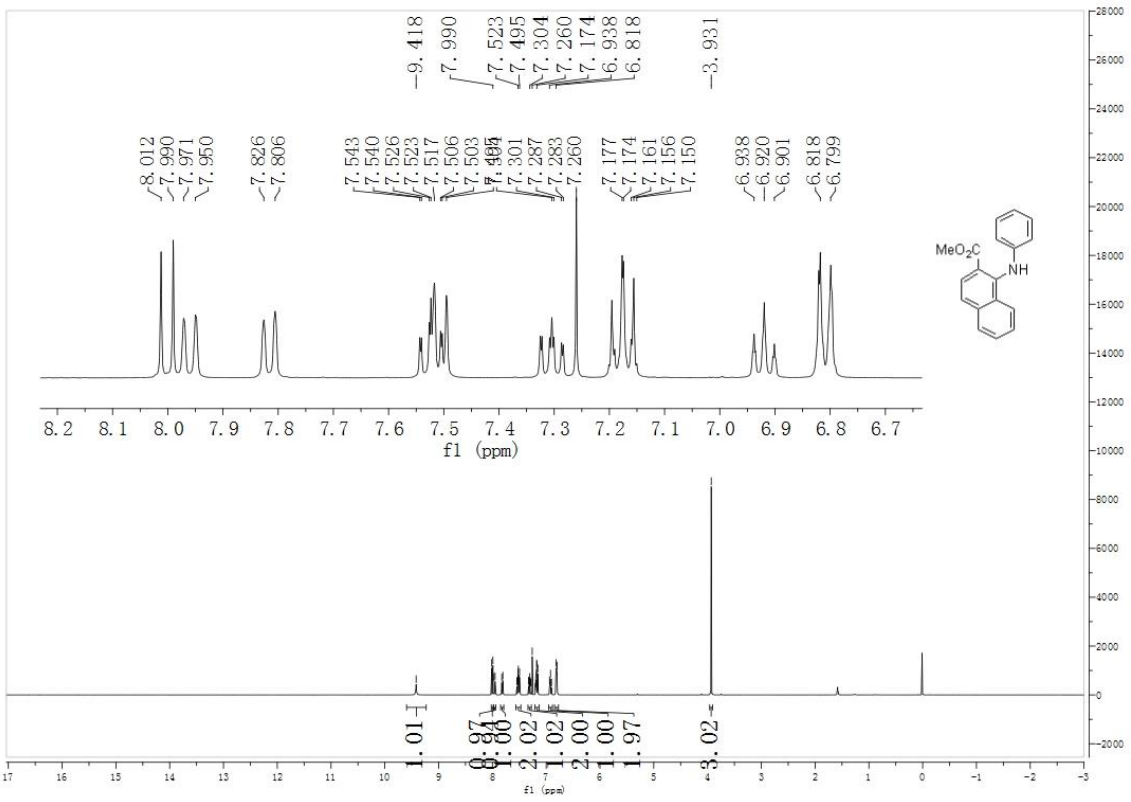
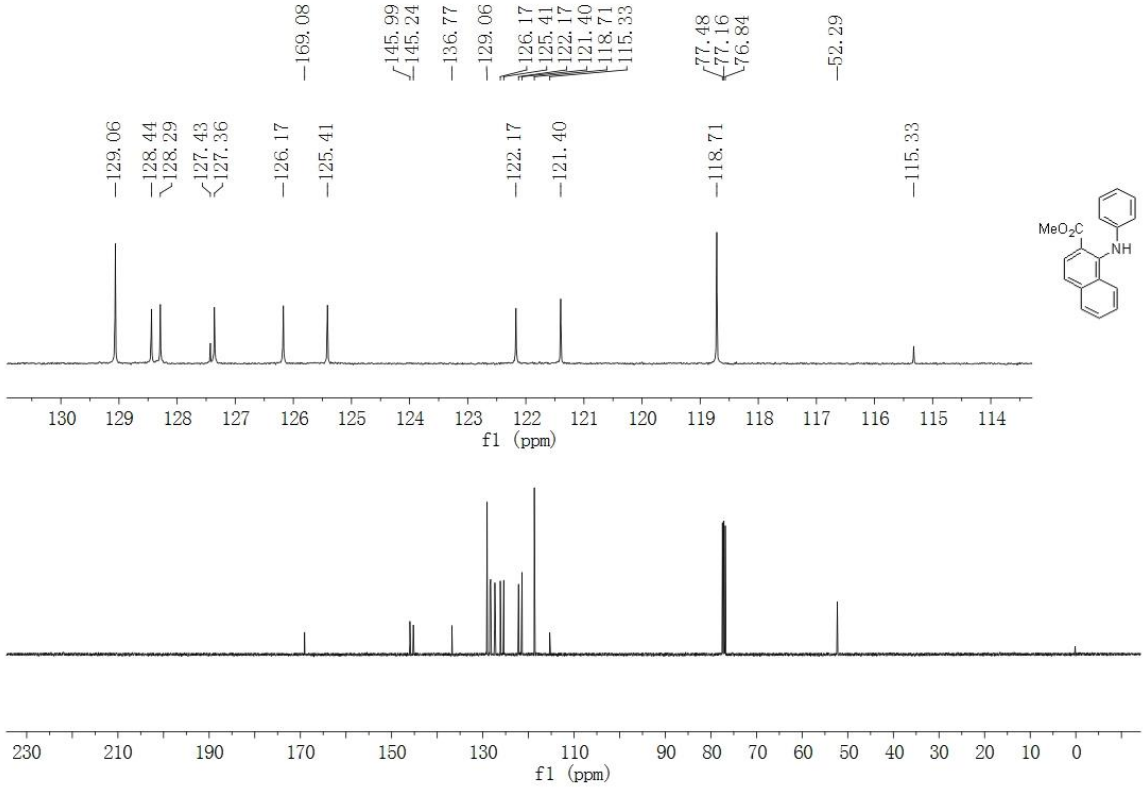
VIII. Copies of ^{1}H and ^{13}C NMR spectra

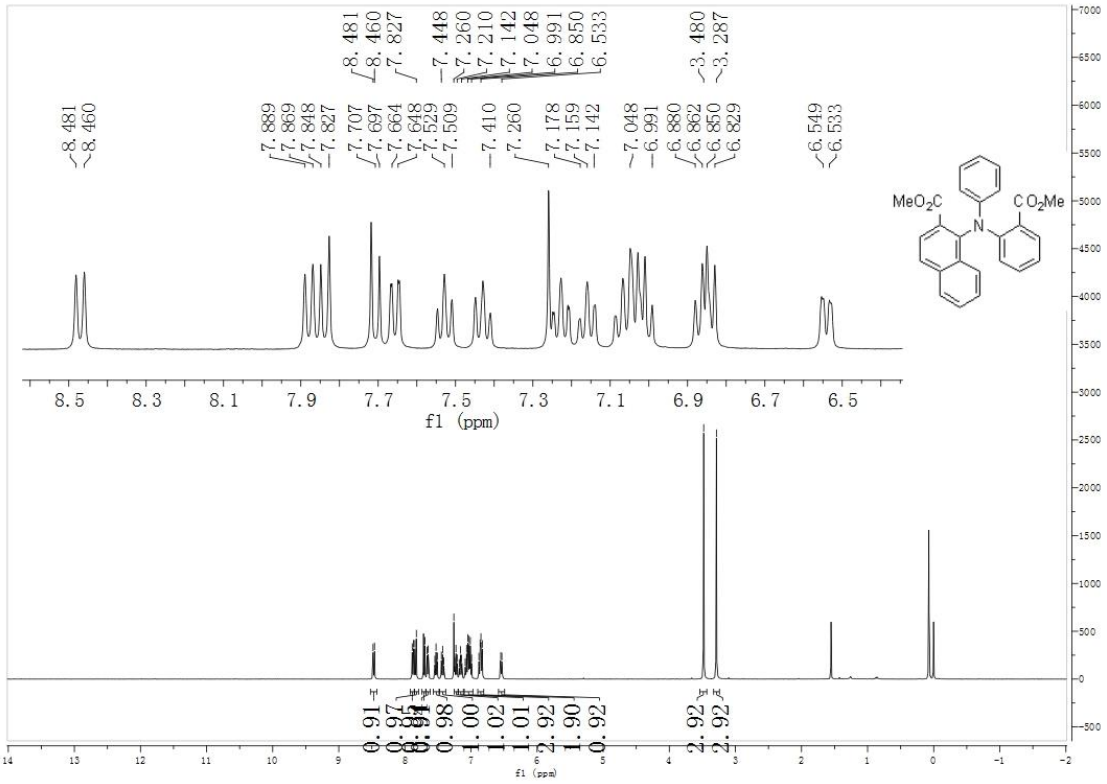
^{1}H NMR spectrum of **3** ($CDCl_3$)



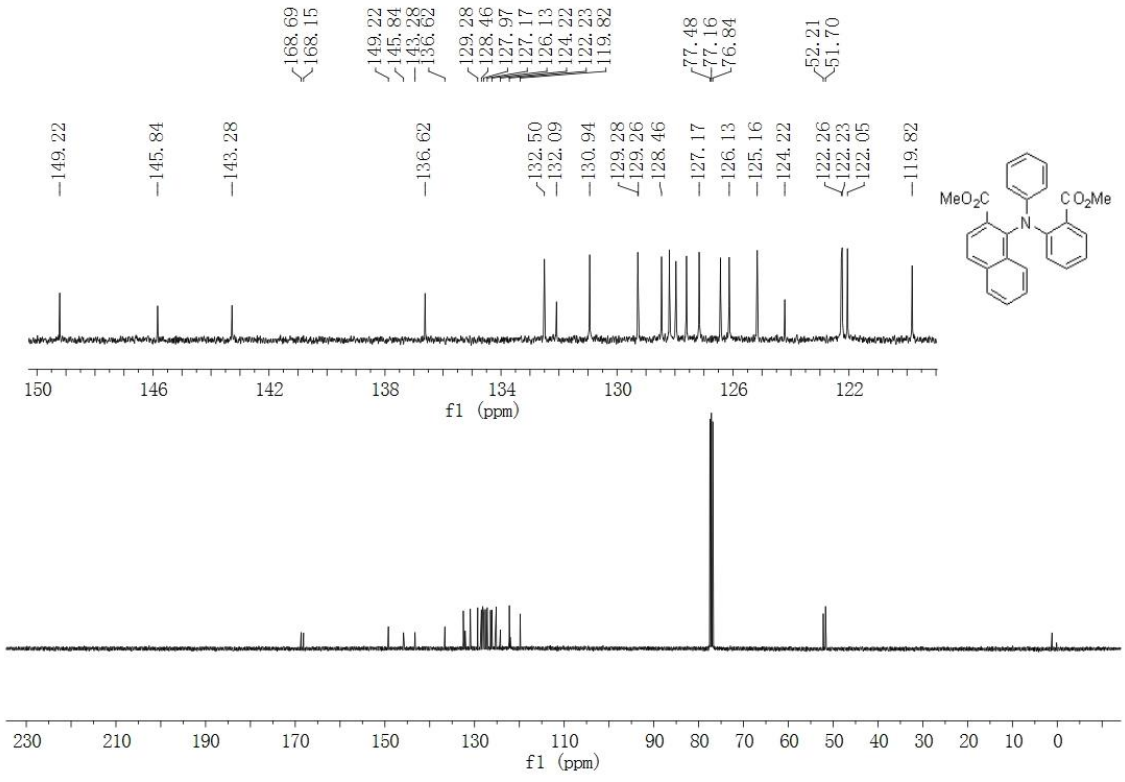
^{13}C NMR spectrum of **3** ($CDCl_3$)



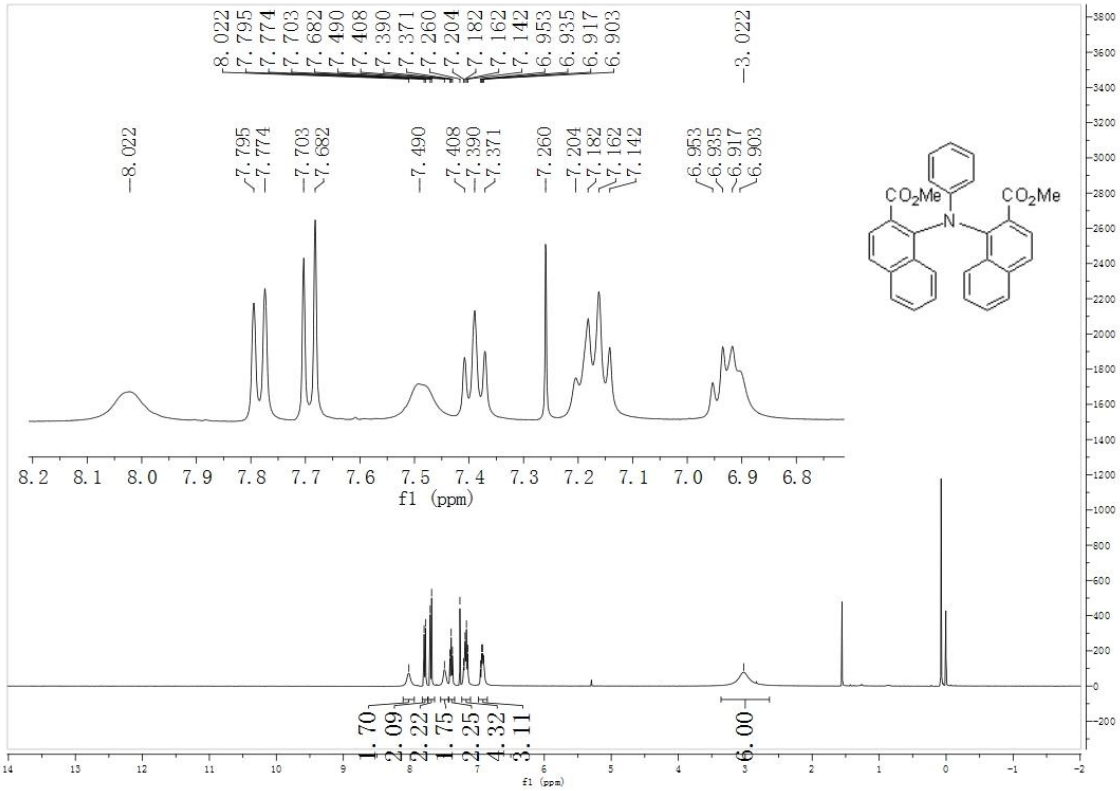
^{1}H NMR spectrum of **5** ($CDCl_3$)



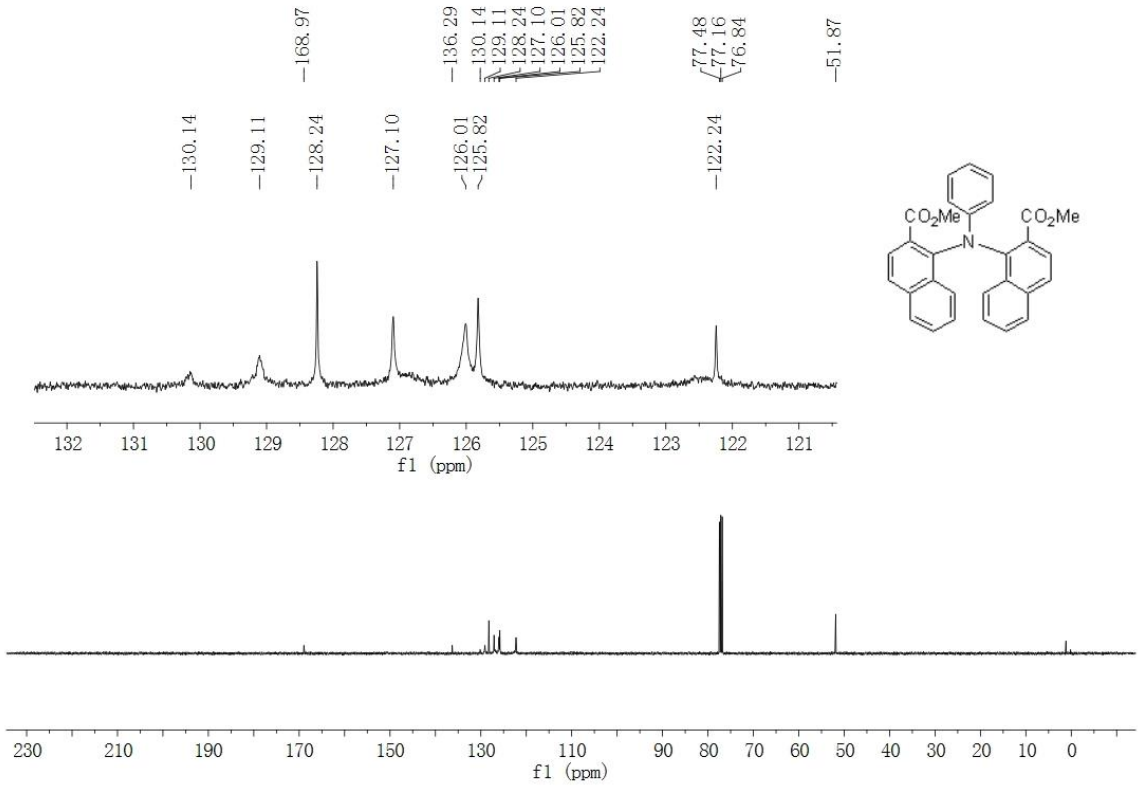
^{13}C NMR spectrum of **5** ($CDCl_3$)



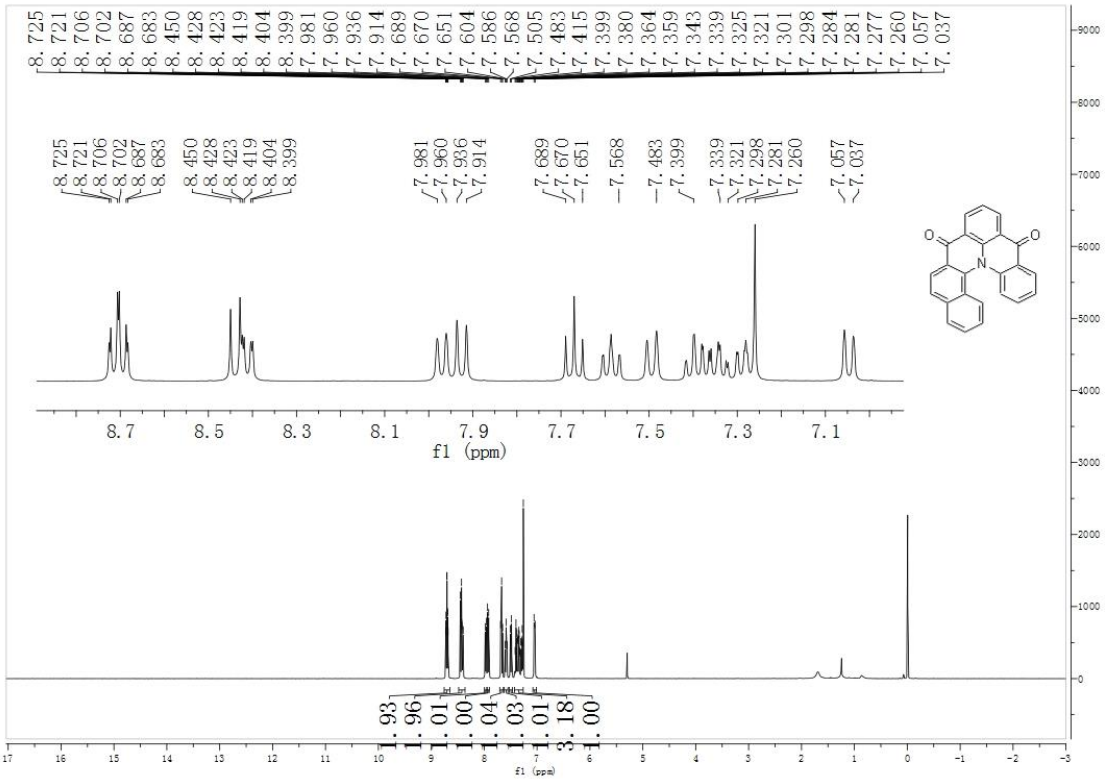
^{1}H NMR spectrum of **6** ($CDCl_3$)



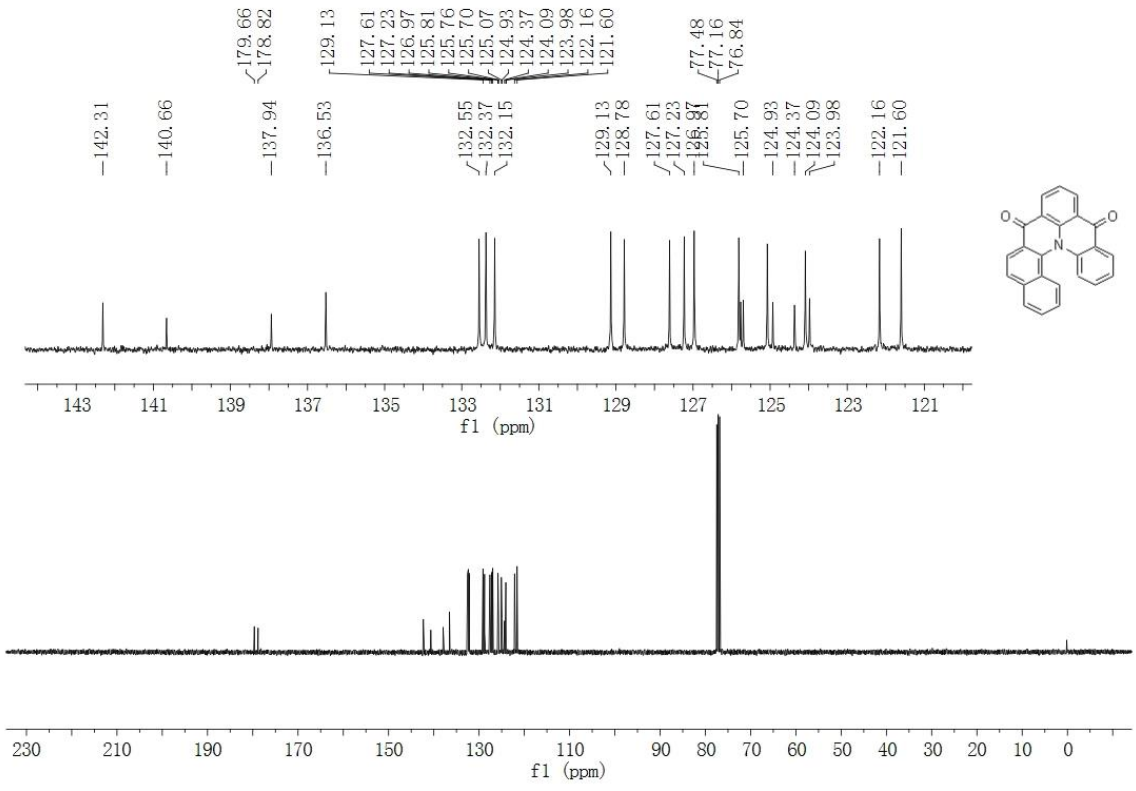
^{13}C NMR spectrum of **6** ($CDCl_3$)



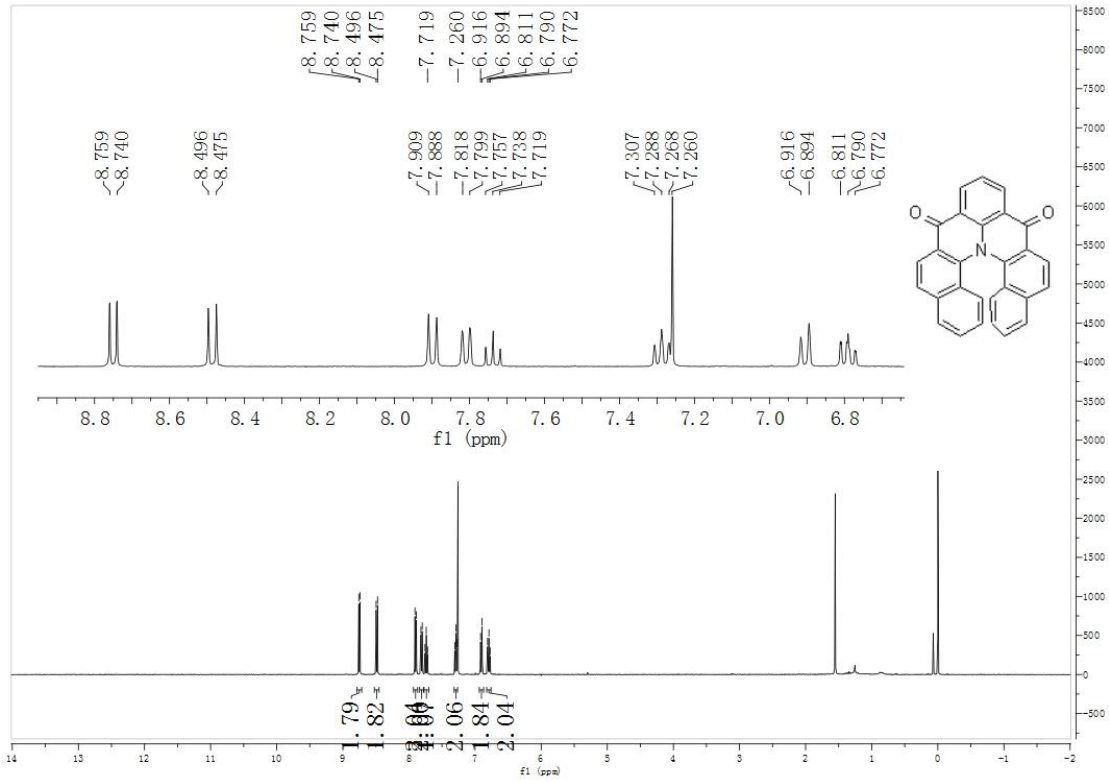
^{1}H NMR spectrum of **[5]*he*-BQAO** ($CDCl_3$)



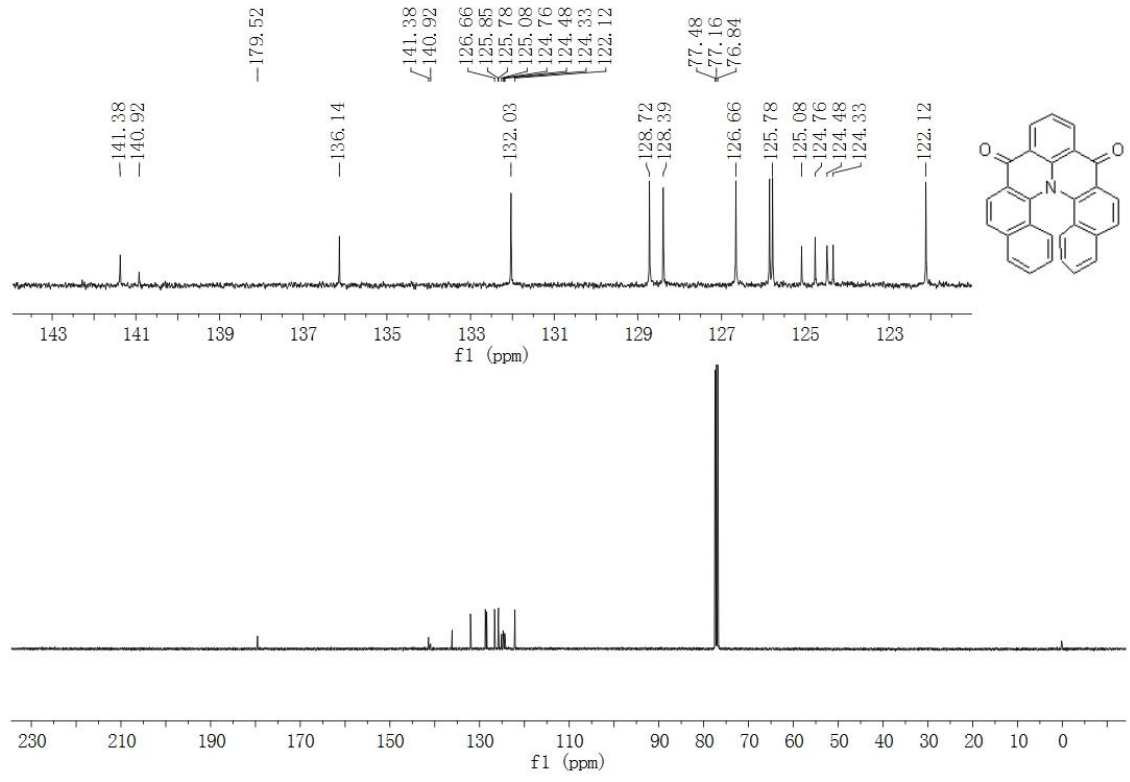
^{13}C NMR spectrum of **[5]*he*-BQAO** ($CDCl_3$)



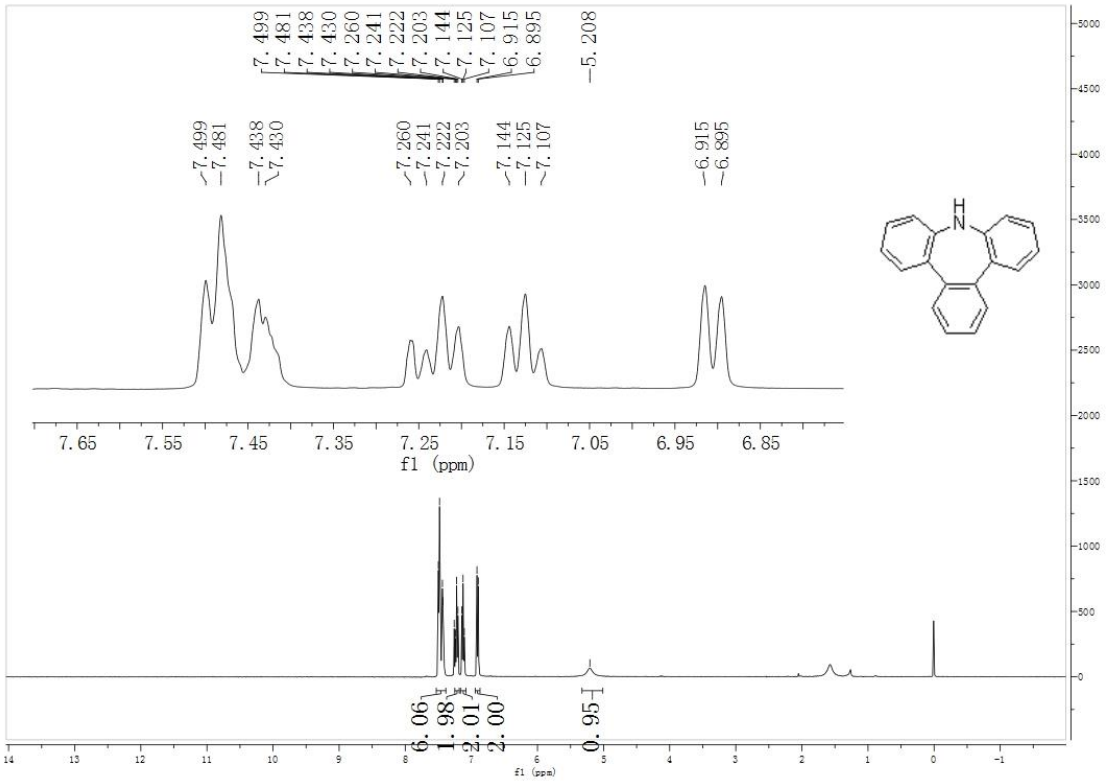
^{1}H NMR spectrum of **[6]*he*-BQAO** ($CDCl_3$)



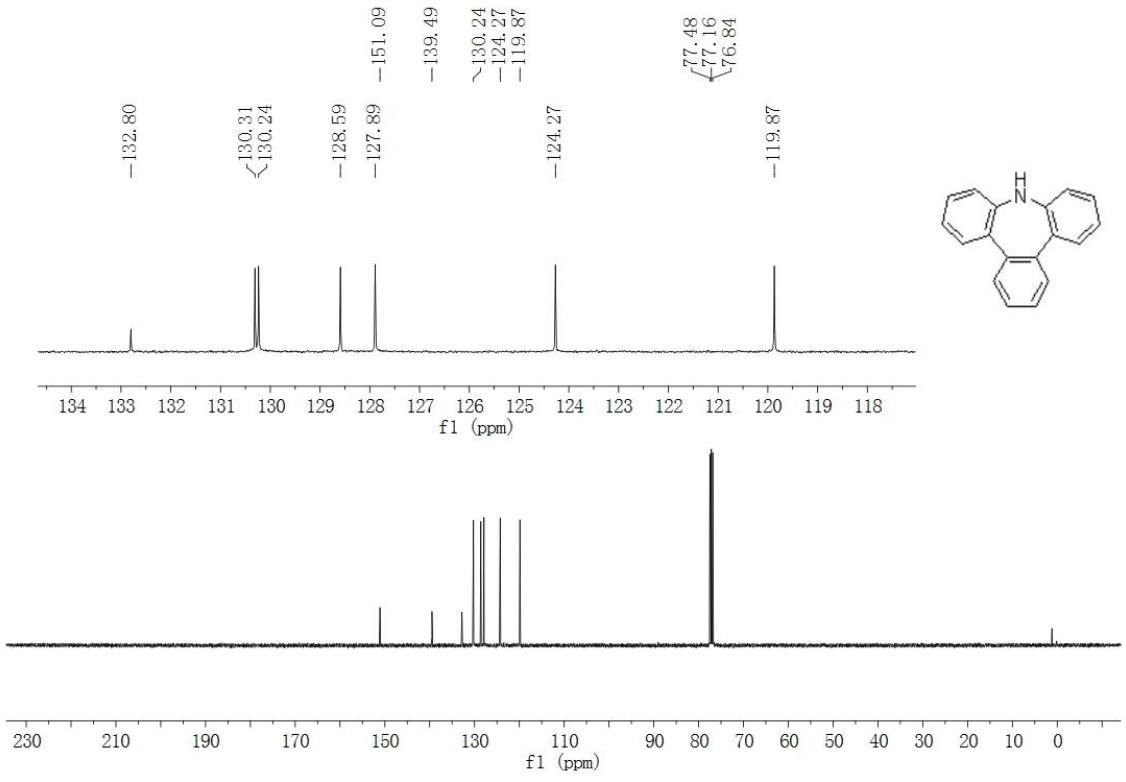
^{13}C NMR spectrum of **[6]*he*-BQAO** ($CDCl_3$)



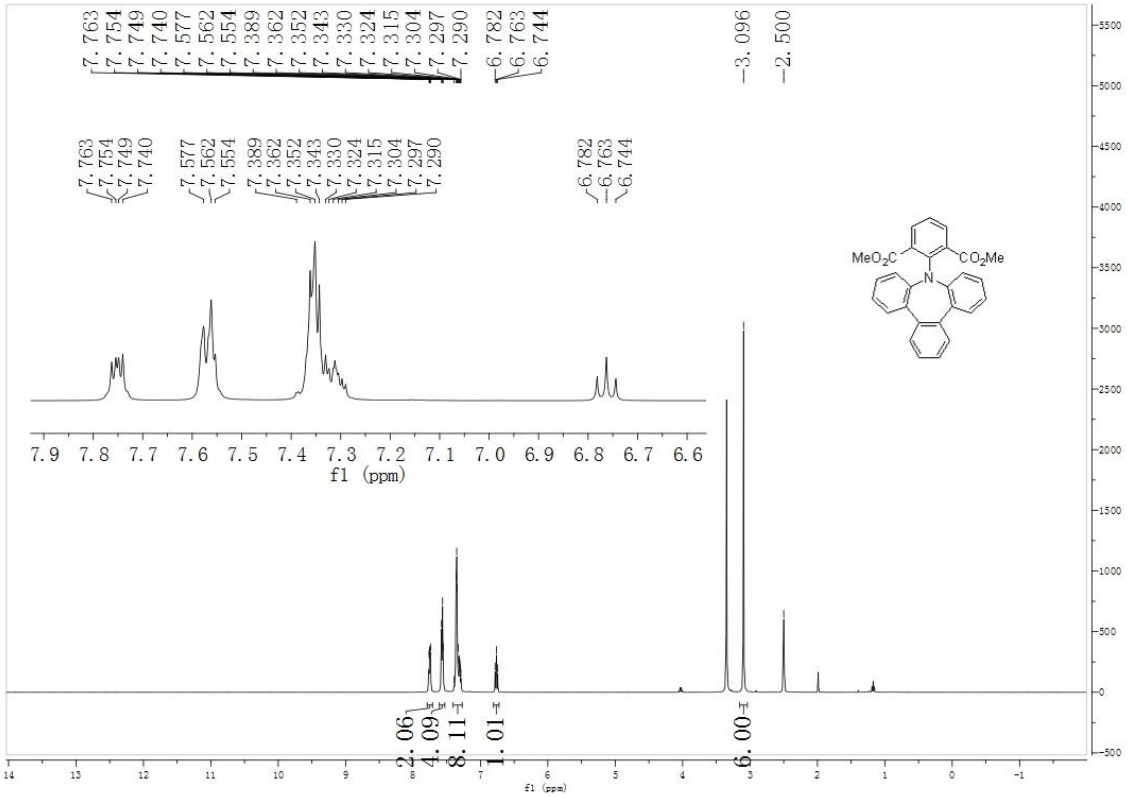
^{1}H NMR spectrum of **9** ($CDCl_3$)



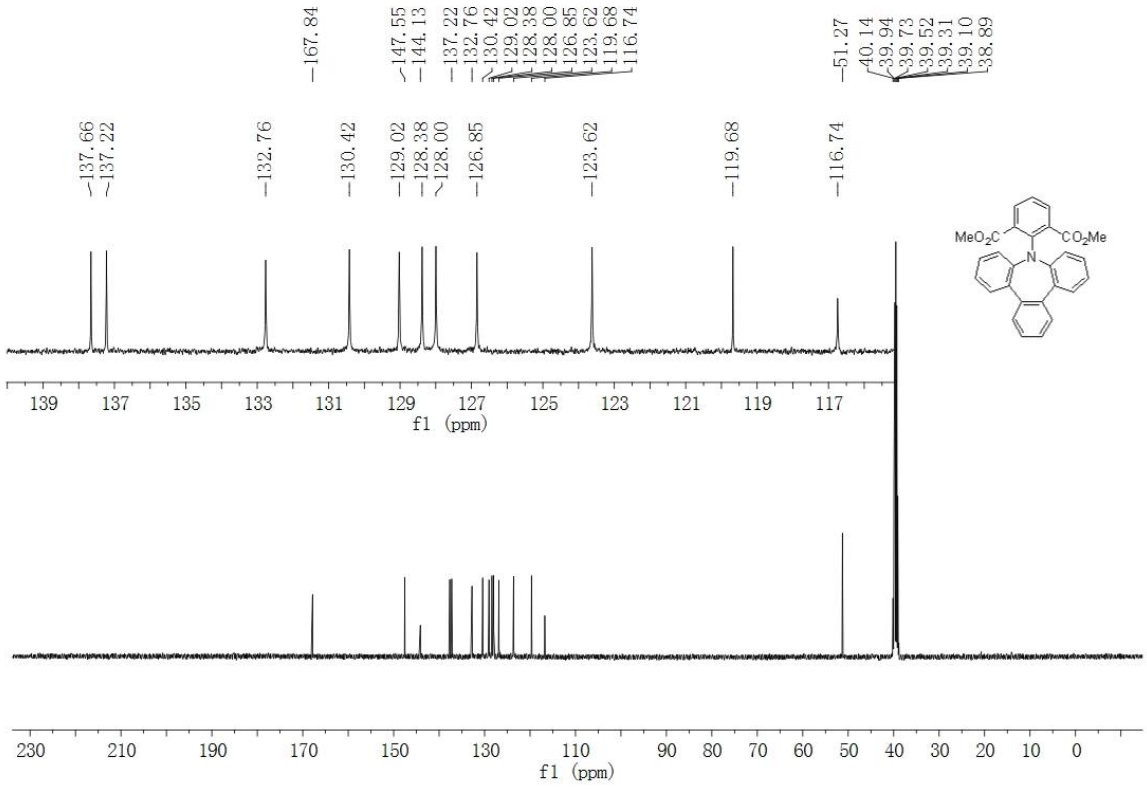
^{13}C NMR spectrum of **9** ($CDCl_3$)



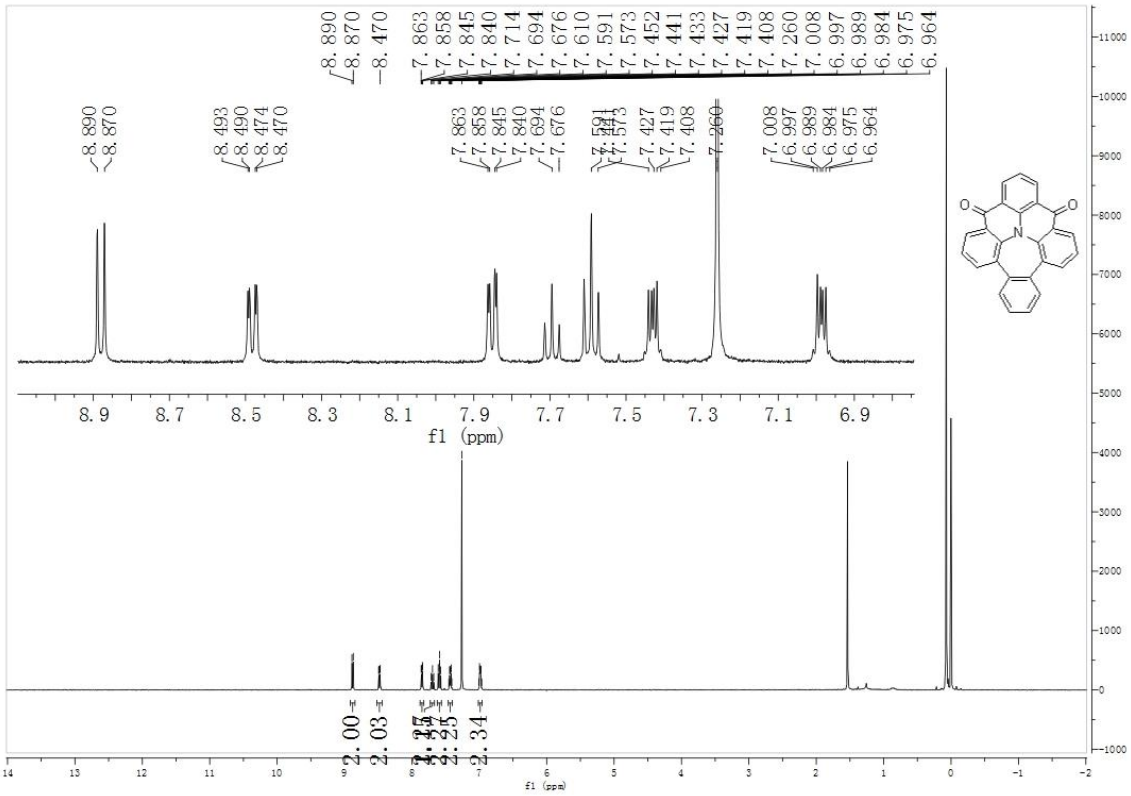
^{1}H NMR spectrum of **11** (DMSO-d_6)



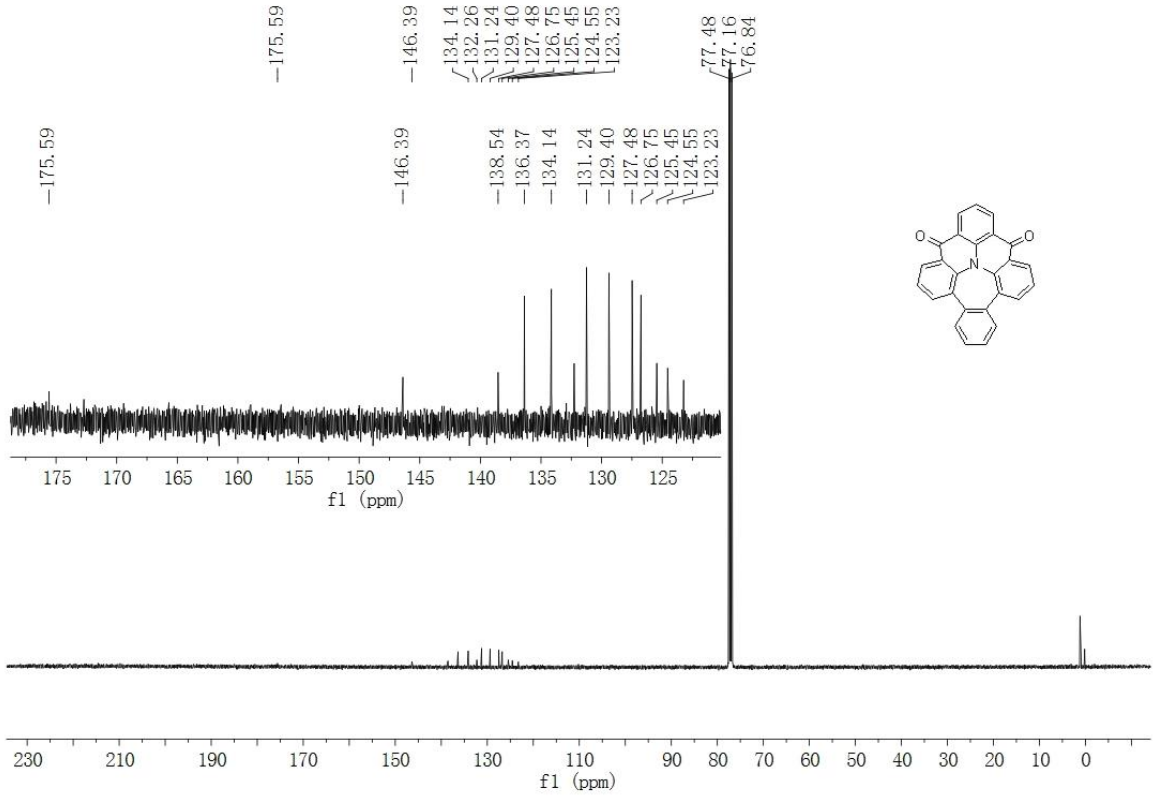
^{13}C NMR spectrum of **11** (DMSO-d_6)



^{1}H NMR spectrum of ***hp*-BQAO** ($CDCl_3$)

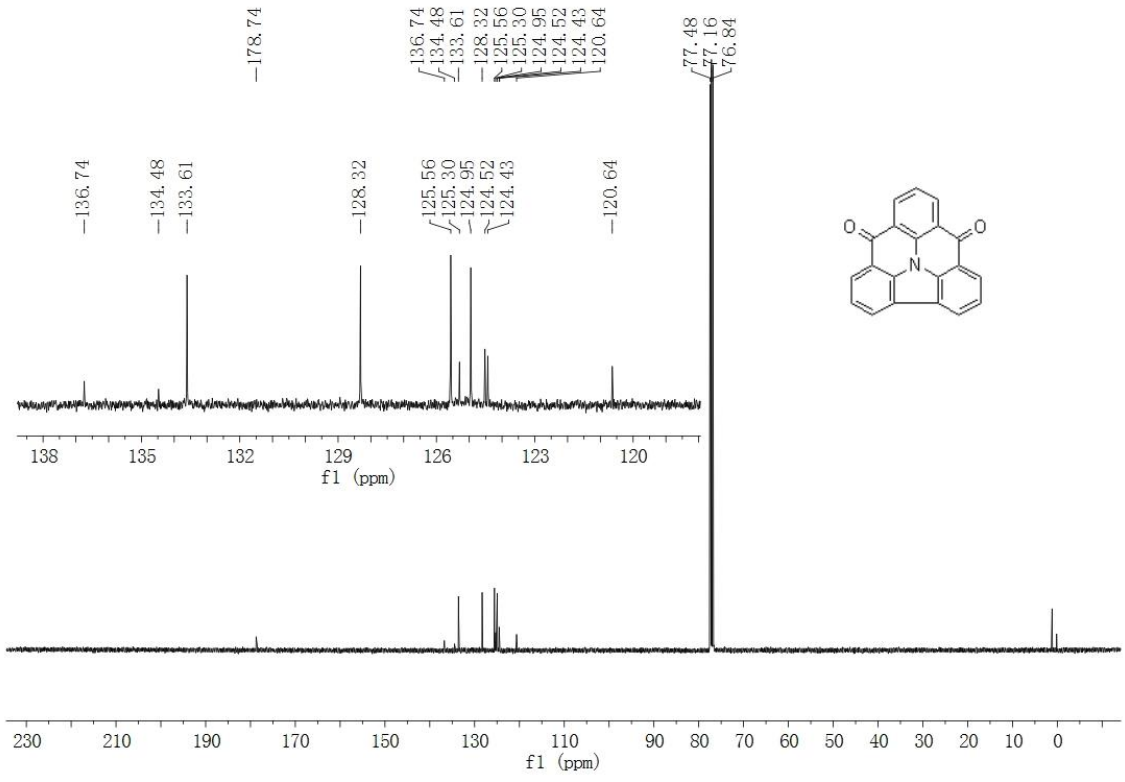


^{13}C NMR spectrum of ***hp*-BQAO** ($CDCl_3$)

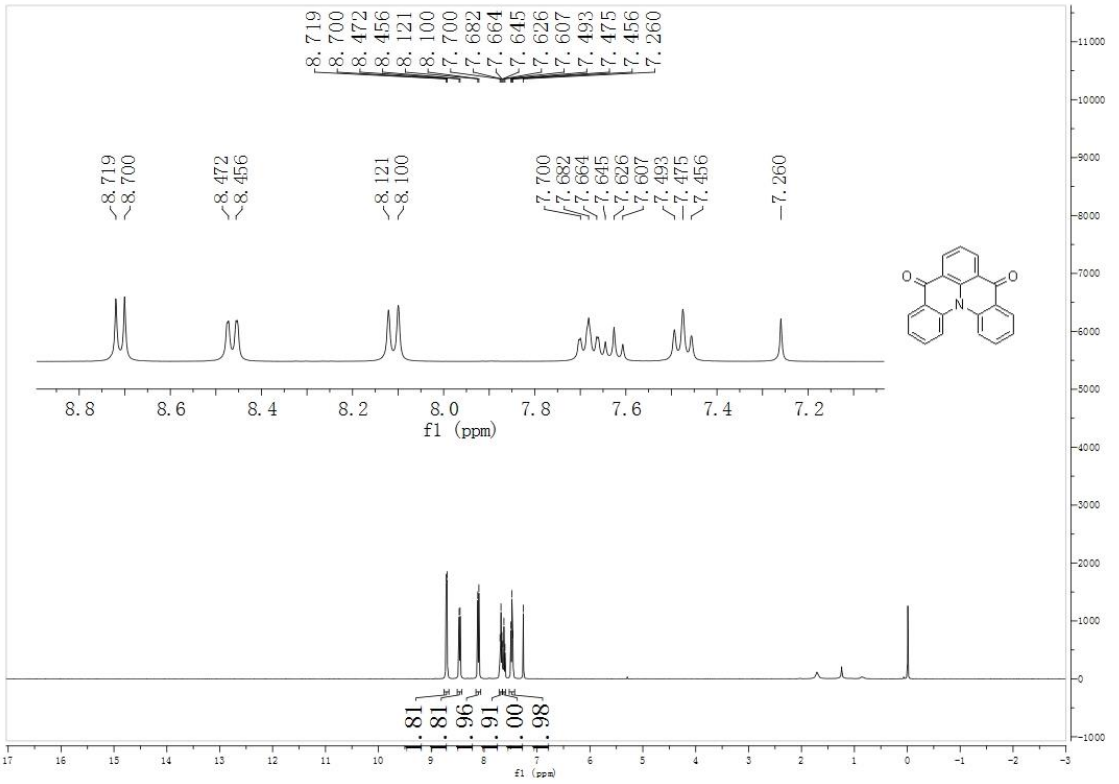


^{1}H NMR spectrum of ***pe*-QAO** ($CDCl_3$)

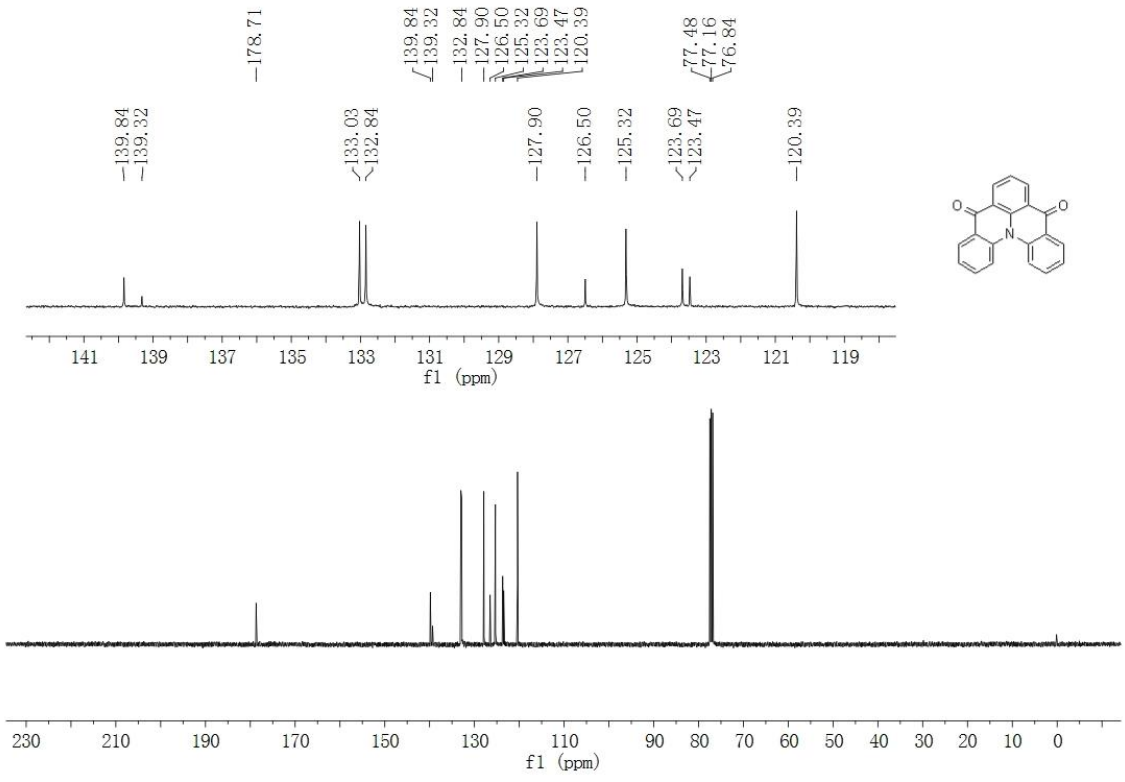
^{13}C NMR spectrum of ***pe*-QAO** ($CDCl_3$)



^{1}H NMR spectrum of **QAO** ($CDCl_3$)



^{13}C NMR spectrum of **QAO** ($CDCl_3$)



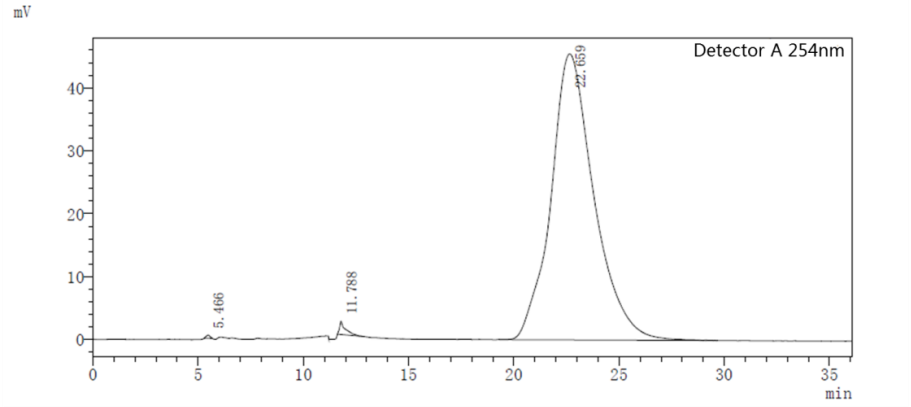
IX. Copies of HPLC spectra

HPLC trace of ***pe*-QAO**

<Sample Information>

Sample Name : *pe*-QAO
Method Filename : 4%-0.6 mL/min
Injection Volume : 10 μL

<Chromatogram>



***pe*-QAO**

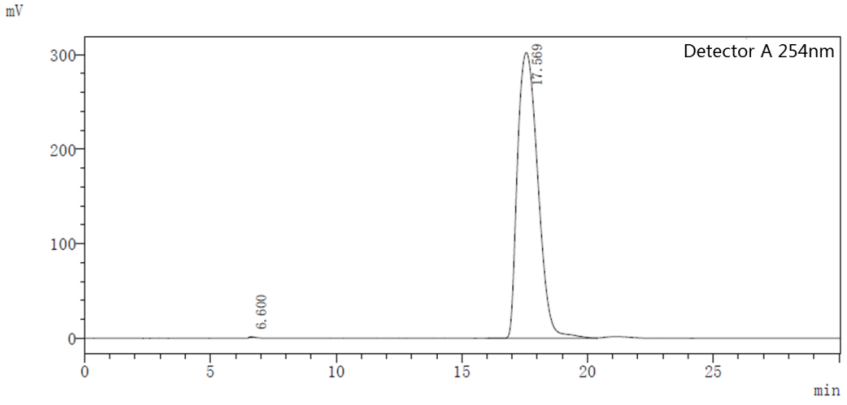
Peak#	Ret. Time	Area	Height	Area%
1	5.466	6040	484	0.093
2	11.788	41061	2085	0.629
3	22.659	6475731	45451	99.278
Total		6522832	48020	

HPLC trace of **QAO**

<Sample Information>

Sample Name : QAO
Method Filename : 3%-0.7 mL/min
Injection Volume : 6 μL

<Chromatogram>



QAO

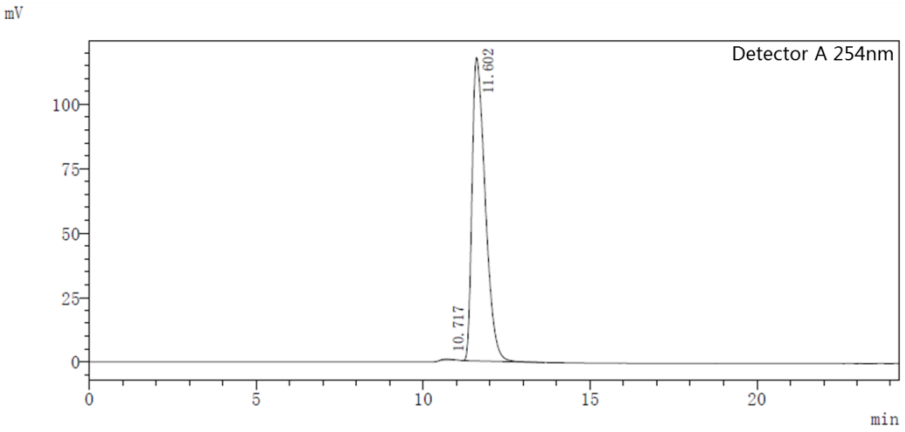
Peak#	Ret. Time	Area	Height	Area%
1	6.600	4285	595	0.025
2	17.569	16981683	302197	99.975
Total		16985968	302792	

HPLC trace of **[5]*he*-BQAO**

<Sample Information>

Sample Name : [5]*he*-BQAO
Method Filename : 3%-0.7 mL/min
Injection Volume : 4 μL

<Chromatogram>



[5]*he*-BQAO

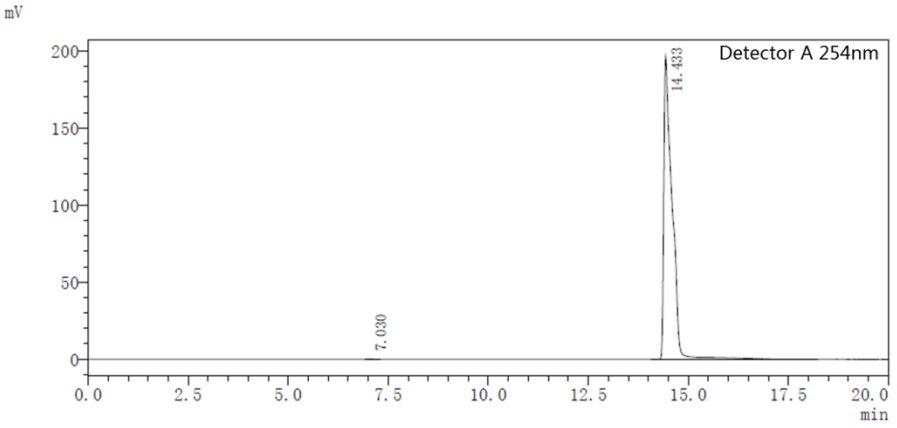
Peak#	Ret. Time	Area	Height	Area%
1	10.717	4806	323	0.148
2	11.602	3234697	117698	99.852
Total		3239503	118021	

HPLC trace of **[6]*he*-BQAO**

<Sample Information>

Sample Name : [6]*he*-BQAO
Method Filename : 3%-0.7 mL/min
Injection Volume : 6 μL

<Chromatogram>



[6]*he*-BQAO

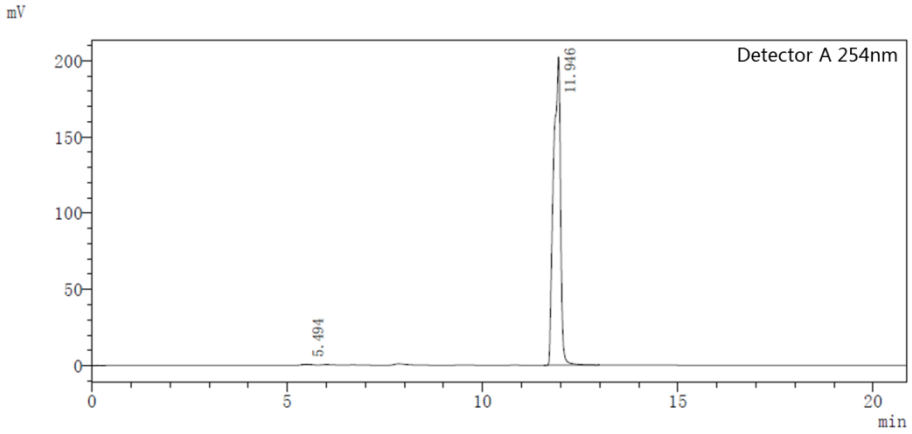
Peak#	Ret. Time	Area	Height	Area%
1	7.030	1469	115	0.052
2	14.433	2845481	196606	99.948
Total		2846950	196721	

HPLC trace of ***hp*-BQAO**

<Sample Information>

Sample Name : *hp*-BQAO
Method Filename : 4%-0.7 mL/min
Injection Volume : 6 μL

<Chromatogram>



***hp*-BQAO**

Peak#	Ret. Time	Area	Height	Area%
1	5.494	1946	216	0.077
2	11.946	2517664	201854	99.923
Total		2519610	202070	