Supporting Information

Diastereoselective reaction to construct polycyclic spiroindolines from 3-(2-isocyanoethyl)indoles and ynones

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1. General Information

The NMR spectra were recorded on Bruker AC-500 spectrometer (500 MHz for ^{1}H NMR, 125 MHz for ^{13}C NMR) and JEOL ECX- 400 spectrometer (400 MHz for ^{1}H NMR and 101 MHz for ^{13}C NMR) with $CDCl_3$ as the solvent and TMS as internal reference. ^{1}H NMR spectral data were reported as follows: chemical shift (δ, ppm), multiplicity, integration, and coupling constant (Hz). ^{13}C NMR spectral data were reported in terms of the chemical shift. The following abbreviations were used to indicate multiplicities: s = singlet; d = doublet; t = triplet; q = quartet; m = multiplet. Low-resolution mass spectra were obtained on a Shimadzu LCMS-2010EV spectrometer in ESI mode and reported as m/z. High-resolution mass spectra (HRMS) were recorded on a Bruker Daltonics, Inc. APEXIII 7.0 TESLA FTMS instrument. Melting points were obtained on an X-4 digital melting point apparatus without correction. Purification of products was accomplished by column chromatography packed with silica gel. Unless otherwise stated, all reagents were commercially purchased and used without further purification.

2. General procedures

2.1 General procedure for the synthesis of spiroindoline 3.

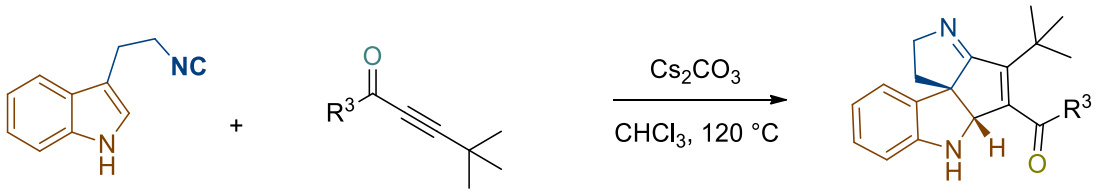
Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), ynone **2** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. After completion of the reaction, the reaction mixture was concentrated under vacuum. The residue was purified by column chromatography on silica gel to afford the desired product.

2.2 General procedure for the synthesis of spiroindoline 4.

Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with tryptamine-derived isocyanide **1** (0.2 mmol), ynone **2** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. After completion of the reaction, the reaction mixture was concentrated under vacuum. The residue was purified by column chromatography on silica gel to afford the desired product.

2.3 General procedure for the control experiments.

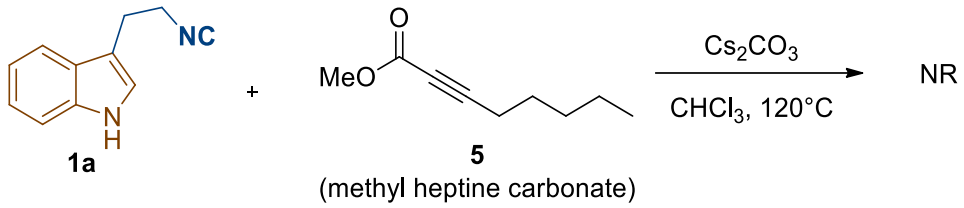
2.3.1 Reaction with dialkyl- and alkyl-aryl-substituted ynone.



R^3 = Et, **3p**, 36%
= Ph, NR

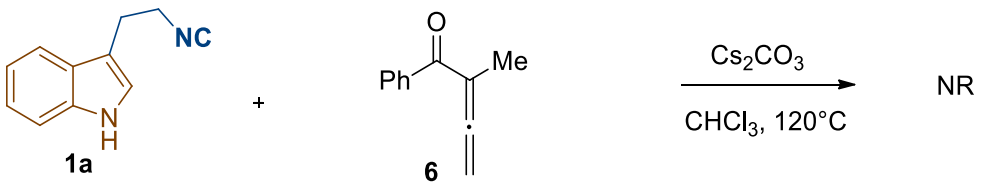
Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), ynone **2** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. After completion of the reaction, the reaction mixture was concentrated under vacuum. The residue was purified by column chromatography on silica gel to afford the desired product.

2.3.2 Reaction with methyl heptine carbonate.



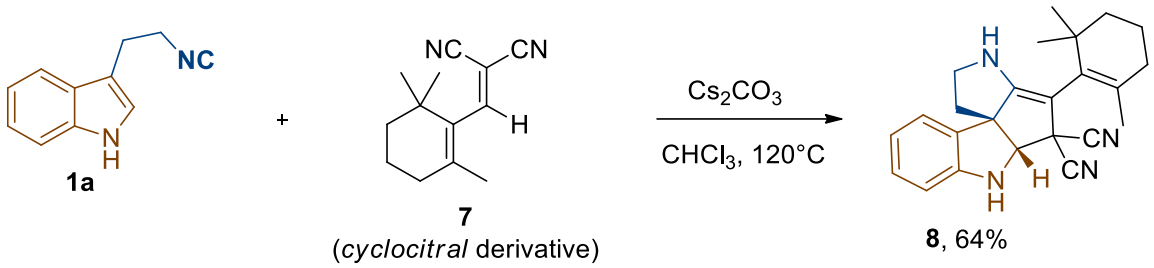
Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), methyl heptine carbonate **5** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. The analysis of TLC showed that no reaction occurred.

2.3.3 Reaction with allenic ketone.



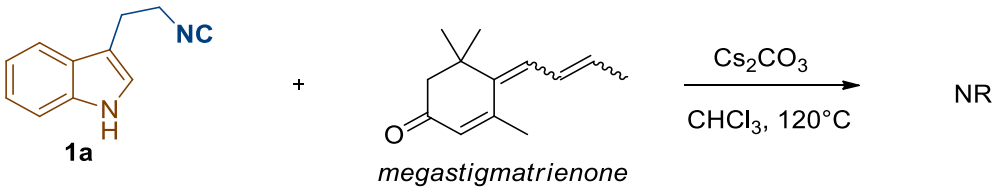
Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), allenic ketone **6** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. The analysis of TLC showed that no reaction occurred.

2.3.4 Reaction with cyclocitral-derived malononitrile.



Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), cyclocitral-derived malononitrile **7** (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. After the completion of the reaction, the organic solvent was removed under vacuum and the residue was purified by column chromatography to afford **8** (64% yield) as yellow oil.

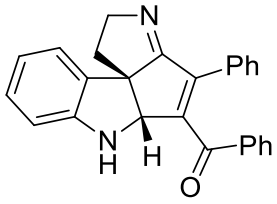
2.3.5 Reaction with megastigmatrienone.



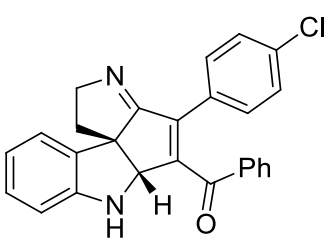
Under air atmosphere, a sealable reaction tube equipped with a magnetic stir bar was charged with 3-(2-isocyanoethyl)-1H-indole **1a** (0.2 mmol), megastigmatrienone (0.2 mmol) and 0.2 mmol Cs_2CO_3 in 2.0 mL $CHCl_3$ at room temperature. The rubber septum was then replaced by a Teflon-coated screw cap, and the reaction vessel placed in an oil bath at 120 °C for 12 hours. The analysis of TLC showed that no reaction occurred.

3. Product Characterization

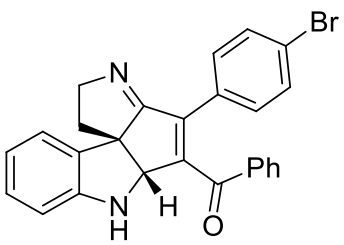
Spectroscopic Data of All Compounds



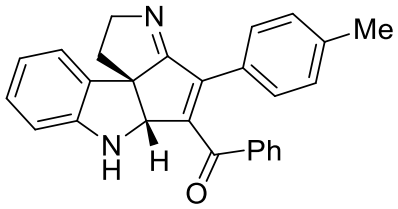
(**3a**): 60 mg, 80% yield, yellow solid. m.p. 48-50 ºC. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.55 (d, *J* = 7.8 Hz, 2H), 7.39-7.32 (m, 3H), 7.23 (d, *J* = 7.5 Hz, 1H), 7.20-7.09 (m, 6H), 6.83 (t, *J* = 7.5 Hz, 1H), 6.64 (d, *J* = 7.8 Hz, 1H), 4.92 (s, 1H), 4.76-4.64 (m, 1H), 4.55-4.45 (m, 1H), 4.40 (s, 1H), 2.42-2.34 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.13, 187.13, 152.55, 148.21, 140.35, 136.31, 133.77, 132.13, 130.66, 129.40, 129.36, 129.18, 128.55, 128.31, 123.61, 120.13, 111.17, 68.74, 68.19, 66.80, 38.52. HRMS (ESI): Calcd. for $C_{26}H_{21}N_2O$ $[M+H]^+$: 377.1654, Found: 377.1658.



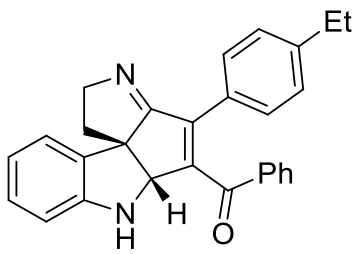
(**3b**): 57 mg, 70% yield, yellow solid. m.p. 180-182 ºC. ^{1}H NMR (400 MHz, Chloroform-d) δ 7.57-7.52 (m, 2H), 7.44-7.39 (m, 1H), 7.33-7.29 (m, 2H), 7.23-7.18 (m, 3H), 7.14-7.10 (m, 1H), 7.10-7.06 (m, 2H), 6.82 (td, *J* = 7.5, 1.0 Hz, 1H), 6.63 (dt, *J* = 7.9, 0.8 Hz, 1H), 4.89 (s, 1H), 4.72-4.62 (m, 1H), 4.48 (m, 1H), 4.39 (s, 1H), 2.42-2.31 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-d) δ 196.87, 186.83, 153.10, 148.13, 138.70, 136.09, 135.36, 134.10, 132.00, 130.64, 129.34, 129.28, 129.17, 128.74, 128.59, 123.57, 120.24, 111.26, 68.77, 68.27, 66.80, 38.52. HRMS (ESI): Calcd. for $C_{26}H_{20}ClN_2O$ $[M+H]^+$: 411.1264, Found: 411.1260.



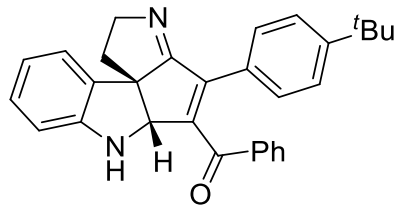
(**3c**): 39 mg, 43% yield, red solid. m.p. 189-191 ºC. ^{1}H NMR (400 MHz, DMSO-d6) δ 7.56-7.52 (m, 2H), 7.49-7.45 (m, 1H), 7.30 (t, *J* = 7.8 Hz, 2H), 7.25 (dd, *J* = 7.3, 2.4 Hz, 2H), 7.16 (dd, *J* = 3.7, 2.0 Hz, 3H), 7.14 (d, *J* = 1.6 Hz, 1H), 7.04 (dd, *J* = 8.4, 2.2 Hz, 1H), 6.49 (d, *J* = 8.2 Hz, 2H), 4.98 (d, *J* = 2.1 Hz, 1H), 4.63 (m, 1H), 4.26 (dd, *J* = 15.9, 8.3 Hz, 1H), 2.46 (d, *J* = 1.9 Hz, 2H), 2.39-2.29 (m, 1H), 2.10 (dd, *J* = 12.4, 5.5 Hz, 1H). ^{13}C NMR (101 MHz, DMSO-d6) δ 196.47, 185.83, 154.40, 148.32, 138.45, 136.43, 134.54, 134.14, 130.95, 129.68, 129.36, 129.27, 129.14, 128.78, 123.61, 121.82, 111.54, 68.98, 68.50, 66.39, 38.48. HRMS (ESI): Calcd. for $C_{26}H_{20}BrN_2O$ $[M+H]^+$: 455.0759, Found: 455.0769.



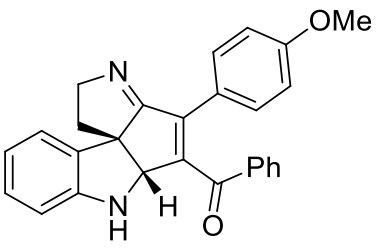
(**3d**): 44 mg, 57% yield, viscious oil. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.57 (d, *J* = 8.9 Hz, 2H), 7.40-7.36 (m, 1H), 7.27-7.18 (m, 5H), 7.11 (t, *J* = 7.7 Hz, 1H), 6.91 (d, *J* = 7.8 Hz, 2H), 6.82 (t, *J* = 7.4 Hz, 1H), 6.63 (d, *J* = 7.7 Hz, 1H), 4.90 (s, 1H), 4.75-4.62 (m, 1H), 4.55-4.44 (m, 1H), 4.39 (s, 1H), 2.39-2.33 (m, 2H), 2.18 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.31, 187.23, 151.64, 148.26, 140.17, 139.45, 136.40, 133.71, 132.22, 129.38, 129.12, 129.02, 128.57, 127.73, 123.61, 120.06, 111.13, 68.74, 68.13, 66.75, 38.51, 21.35. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O$ $[M+H]^+$: 391.1810, Found: 391.1801.



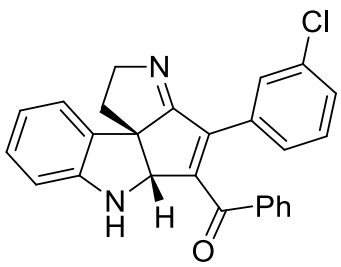
(**3e**): 62 mg, 75% yield, red solid. m.p. 161-163 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.57 (dd, *J* = 8.4, 1.3 Hz, 2H), 7.39-7.34 (m, 1H), 7.29-7.26 (m, 2H), 7.23 (d, *J* = 6.8 Hz, 1H), 7.20-7.16 (m, 2H), 7.11 (td, *J* = 7.7, 1.3 Hz, 1H), 6.93 (d, *J* = 8.1 Hz, 2H), 6.82 (td, *J* = 7.5, 1.0 Hz, 1H), 6.62 (d, *J* = 7.9 Hz, 1H), 4.90 (s, 1H), 4.73-4.63 (m, 1H), 4.53-4.39 (m, 2H), 2.46 (q, *J* = 7.6 Hz, 2H), 2.39-2.33 (m, 2H), 1.06 (t, *J* = 7.6 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.30, 187.29, 151.71, 148.28, 145.75, 140.30, 136.43, 133.67, 132.20, 129.49, 129.39, 129.13, 128.55, 127.94, 127.85, 123.61, 120.04, 111.12, 68.75, 68.12, 66.75, 38.51, 28.71, 15.40. HRMS (ESI): Calcd. for $C_{28}H_{25}N_2O$ $[M+H]^+$: 405.1967, Found: 405.1975.



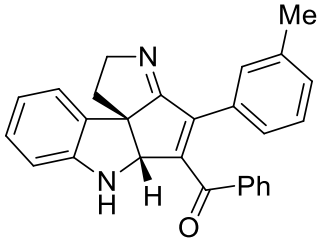
(**3f**): 45 mg, 52% yield, viscious oil. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.49-7.45 (m, 2H), 7.40-7.36 (m, 2H), 7.22 (d, *J* = 7.6 Hz, 1H), 7.14-7.09 (m, 4H), 6.98 (d, *J* = 7.2 Hz, 2H), 6.82 (t, *J* = 7.8 Hz, 1H), 6.62 (d, *J* = 7.5 Hz, 1H), 4.88 (s, 1H), 4.77-4.60 (m, 1H), 4.55-4.44 (m, 1H), 4.35 (s, 1H), 2.38-2.33 (m, 2H), 2.26 (s, 9H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.27, 187.30, 152.49, 151.72, 148.25, 140.37, 136.47, 133.59, 132.18, 129.42, 129.20, 129.12, 128.53, 127.64, 125.25, 123.61, 122.75, 122.40, 120.05, 119.75, 118.24, 111.53, 111.10, 68.74, 68.07, 66.76, 38.48, 34.64, 31.07. HRMS (ESI): Calcd. for $C_{30}H_{29}N_2O$ $[M+H]^+$: 433.2280, Found: 433.2280.



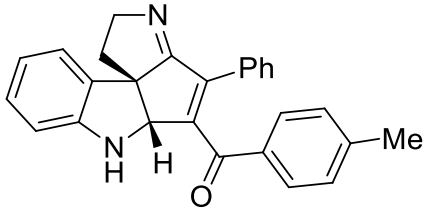
(**3g**): 32 mg, 39% yield, red solid. m.p. 175-176 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.58-7.53 (m, 2H), 7.40-7.35 (m, 1H), 7.32 (d, *J* = 2.1 Hz, 1H), 7.30 (d, *J* = 2.1 Hz, 1H), 7.19 (qd, *J* = 7.2, 6.7, 1.4 Hz, 3H), 7.10 (td, *J* = 7.7, 1.2 Hz, 1H), 6.80 (td, *J* = 7.5, 1.0 Hz, 1H), 6.65-6.59 (m, 3H), 4.88 (s, 1H), 4.72-4.62 (m, 1H), 4.47 (m, 1H), 4.39 (s, 1H), 3.66 (s, 3H), 2.39-2.30 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.40, 187.37, 160.38, 150.63, 148.30, 139.84, 136.42, 133.73, 132.20, 131.09, 129.37, 129.12, 128.60, 123.59, 123.10, 120.02, 113.77, 111.12, 68.69, 68.04, 66.74, 55.27, 38.51. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O_2$ $[M+H]^+$: 407.1760, Found: 407.1771.



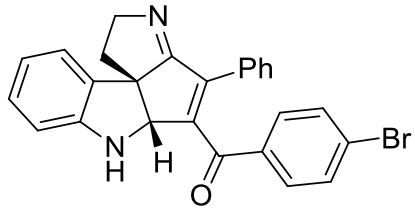
(**3h**): 49 mg, 60% yield, 120-121 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.54 (d, *J* = 7.8 Hz, 2H), 7.43-7.38 (m, 2H), 7.24-7.08 (m, 6H), 7.02 (t, *J* = 7.8 Hz, 1H), 6.84 (t, *J* = 7.4 Hz, 1H), 6.65 (d, *J* = 7.8 Hz, 1H), 4.92 (s, 1H), 4.75-4.62 (m, 1H), 4.53-4.46 (m, 1H), 4.43 (s, 1H), 2.43-2.33 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.73, 186.60, 153.78, 148.11, 138.77, 136.22, 134.27, 133.99, 132.44, 131.95, 129.53, 129.35, 129.30, 129.25, 128.68, 127.46, 123.60, 120.27, 111.24, 68.75, 68.28, 66.85, 38.53. HRMS (ESI): Calcd. for $C_{26}H_{20}ClN_2O$ $[M+H]^+$: 411.1264, Found: 411.1269.



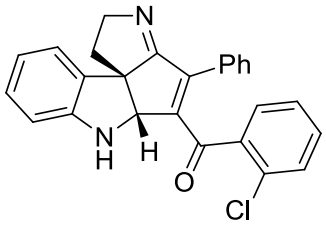
(**3i**): 42 mg, 54% yield, yellow solid. m.p. 45-46 ^{o}C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.53-7.50 (m, 2H), 7.35 (t, 1H), 7.23 (d, *J* = 6.5 Hz, 1H), 7.18-7.11 (m, 5H), 6.91 (d, *J* = 7.6 Hz, 2H), 6.81 (t, *J* = 7.6 Hz, 1H), 6.63 (d, *J* = 6.8 Hz, 1H), 4.91 (s, 1H), 4.75-4.62 (m, 1H), 4.53-4.43 (m, 1H), 4.40 (s, 1H), 2.39-2.34 (m, 2H), 2.11 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.33, 187.24, 151.62, 148.25, 140.18, 139.46, 136.38, 133.73, 132.21, 129.39, 129.13, 129.03, 128.58, 127.71, 123.62, 120.06, 111.14, 68.73, 68.11, 66.76, 38.52, 21.37. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O$ $[M+H]^+$: 391.1810, Found: 391.1815. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O$ $[M+H]^+$: 391.1810, Found: 391.1803.



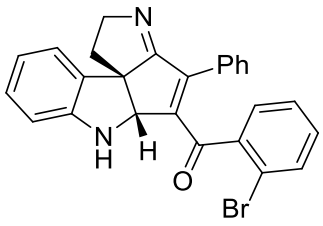
(**3j**): 47 mg, 60% yield, viscious oil. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.50-7.46 (m, 2H), 7.41-7.37 (m, 2H), 7.23 (d, *J* = 7.4 Hz, 1H), 7.15-7.10 (m, 4H), 6.99 (d, *J* = 7.9 Hz, 2H), 6.83 (t, *J* = 7.5 Hz, 1H), 6.63 (d, *J* = 7.8 Hz, 1H), 4.89 (s, 1H), 4.78-4.61 (m, 1H), 4.57-4.45 (m, 1H), 4.36 (s, 1H), 2.40-2.34 (m, 2H), 2.27 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.67, 187.12, 152.94, 148.23, 145.03, 139.42, 133.76, 132.18, 130.74, 129.57, 129.37, 129.31, 129.24, 129.15, 128.34, 123.60, 120.10, 111.15, 68.77, 68.19, 66.78, 38.44, 21.81. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O$ $[M+H]^+$: 391.1810, Found: 391.1822.



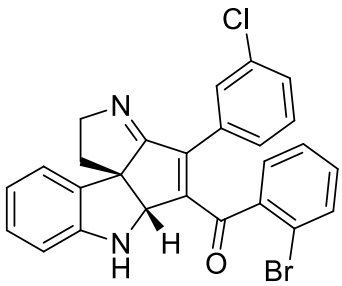
(**3k**): 65 mg, 72% yield, 135-136 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.38 (d, *J* = 8.6 Hz, 2H), 7.34-7.28 (m, 4H), 7.24 (d, *J* = 7.6 Hz, 1H), 7.17-7.11 (m, 4H), 6.84 (t, *J* = 7.4 Hz, 1H), 6.64 (d, *J* = 7.9 Hz, 1H), 4.93 (s, 1H), 4.76-4.63 (m, 1H), 4.55-4.44 (m, 1H), 4.39 (s, 1H), 2.41-2.33 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.18, 186.97, 152.08, 148.08, 140.60, 135.13, 132.21, 131.89, 130.73, 130.44, 129.63, 129.40, 129.26, 129.10, 128.48, 123.64, 120.33, 111.35, 68.78, 68.36, 66.82, 38.55. HRMS (ESI): Calcd. for $C_{26}H_{20}BrN_2O$ $[M+H]^+$: 455.0759, Found: 455.0769.



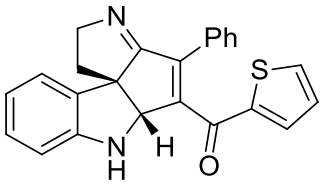
(**3l**): 61 mg, 75% yield, yellow solid: 58-60 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.19-7.11 (m, 4H), 7.09-7.02 (m, 6H), 6.95 -6.89 (m, 1H), 6.78 (t, *J* = 7.4 Hz, 1H), 6.68 (d, *J* = 7.8 Hz, 1H), 4.97 (s, 1H), 4.71 (s, 1H), 4.69-4.58 (m, 1H), 4.52-4.41 (m, 1H), 2.44-2.33 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 195.44, 187.74, 151.74, 148.68, 145.50, 137.63, 132.15, 132.08, 131.36, 130.49, 130.45, 130.21, 129.32, 129.19, 127.84, 126.38, 123.62, 119.70, 110.64, 67.94, 67.44, 66.81, 38.70. HRMS (ESI): Calcd. for $C_{26}H_{20}ClN_2O$ $[M+H]^+$: 411.1264, Found: 411.1270.



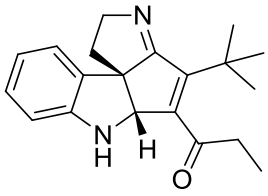
(**3m**): 65 mg, 72% yield, yellow solid. m.p. 180-181 ºC. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.25-7.21 (m, 1H), 7.17-7.10 (m, 4H), 7.06 -6.93 (m, 6H), 6.77 (t, *J* = 7.4 Hz, 1H), 6.67 (d, *J* = 7.8 Hz, 1H), 4.96 (s, 1H), 4.71 (s, 1H), 4.67-4.57 (m, 1H), 4.52-4.40 (m, 1H), 2.42-2.32 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 195.99, 187.82, 151.29, 148.78, 145.92, 139.39, 133.50, 132.09, 131.29, 130.63, 130.45, 129.32, 129.20, 127.82, 126.91, 123.63, 120.44, 119.67, 110.60, 67.97, 67.32, 66.81, 38.74. HRMS (ESI): Calcd. for $C_{26}H_{20}BrN_2O$ $[M+H]^+$: 455.0759, Found: 455.0760.



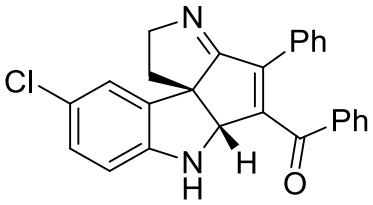
(**3n**) 80 mg, 82% yield, red solid. m.p. 195-197 ºC. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.26 (d, *J* = 7.7 Hz, 1H), 7.18-7.11 (m, 2H), 7.03 (m, 7H), 6.78 (t, *J* = 7.4 Hz, 1H), 6.68 (d, *J* = 7.9 Hz, 1H), 4.97 (s, 1H), 4.75 (s, 1H), 4.67-4.57 (m, 1H), 4.46 (dd, *J* = 16.1, 7.4 Hz, 1H), 2.44 -2.32 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 195.68, 187.40, 152.04, 148.71, 144.40, 139.31, 133.78, 133.48, 132.31, 132.20, 131.03, 130.47, 129.30, 129.13, 127.28, 127.11, 123.62, 120.19, 119.76, 110.62, 67.88, 67.28, 66.87, 38.77. HRMS (ESI): Calcd. for $C_{26}H_{19}BrCl_1N_2O$ $[M+H]^+$: 489.0369, Found: 489.0358.



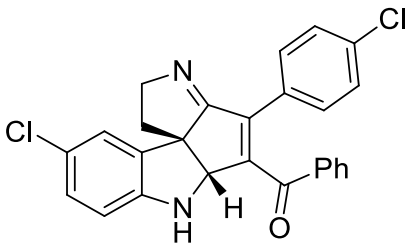
(**3o**): 17 mg, 22% yield, red solid. m.p. 156-157 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.52 (d, *J* = 5.2 Hz, 1H), 7.45 (dd, *J* = 6.7, 3.0 Hz, 2H), 7.23-7.16 (m, 4H), 7.10 (t, *J* = 7.7 Hz, 1H), 7.03 (d, *J* = 3.8 Hz, 1H), 6.80 (t, *J* = 7.4 Hz, 1H), 6.75 (t, *J* = 4.4 Hz, 1H), 6.63 (d, *J* = 7.9 Hz, 1H), 4.88 (s, 1H), 4.73-4.57 (m, 1H), 4.47 (m, 2H), 2.35 (dd, *J* = 9.3, 5.9 Hz, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 188.62, 186.96, 152.67, 148.14, 143.31, 139.60, 135.65, 135.50, 132.10, 130.85, 129.41, 129.28, 129.16, 128.50, 128.39, 123.56, 120.13, 111.20, 68.60, 68.34, 66.78, 38.39. HRMS (ESI): Calcd. for $C_{24}H_{19}N_2OS$ $[M+H]^+$: 383.1218, Found: 383.1220.



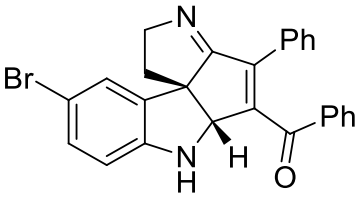
(**3p**): 22 mg, 36% yield, viscious oil. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.13-7.09 (m, 1H), 7.03 (d, *J* = 7.4 Hz, 1H), 6.78-6.72 (m, 2H), 4.56-4.32 (m, 3H), 4.20 (brs, 1H), 2.79-2.62 (m, 2H), 2.19-2.15 (m, 2H), 1.20 (s, 9H), 1.14 (t, *J* = 7.6 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 207.80, 186.94, 152.77, 147.78, 144.32, 132.89, 128.87, 123.08, 120.44, 111.88, 68.56, 67.85, 66.36, 38.53, 37.79, 33.69, 29.31, 7.35. HRMS (ESI): Calcd. for $C_{20}H_{25}N_2O$ $[M+H]^+$: 309.1967, Found: 309.1960.



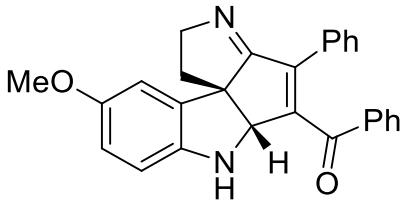
(**4a**): 62 mg, 75% yield, red solid. m.p. 200-202 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.56-7.53 (m, 2H), 7.43-7.32 (m, 4H), 7.30 (d, *J* = 2.0 Hz, 1H), 7.23-7.18 (m, 3H), 7.14-7.10 (m, 3H), 6.51 (d, *J* = 8.3 Hz, 1H), 4.93 (s, 1H), 4.72-4.60 (m, 1H), 4.56-4.43 (m, 2H), 2.40-2.34 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.86, 186.43, 152.08, 147.33, 140.60, 136.18, 134.16, 133.88, 131.99, 130.43, 129.53, 129.47, 129.33, 128.61, 128.36, 126.64, 112.39, 111.41, 68.97, 68.04, 66.79, 38.46. HRMS (ESI): Calcd. for $C_{26}H_{20}ClN_2O$ $[M+H]^+$: 411.1264, Found: 411.1265.



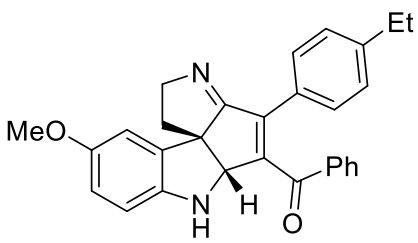
(**4b**): 52mg, 58% yield, red solid. m.p. 185-187 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.59-7.51 (m, 2H), 7.44 (t, *J* = 7.4 Hz, 1H), 7.32 (d, *J* = 8.5 Hz, 1H), 7.23 (t, *J* = 7.8 Hz, 2H), 7.15 (d, *J* = 2.1 Hz, 1H), 7.12-7.06 (m, 5H), 6.56 (d, *J* = 8.4 Hz, 1H), 4.92 (s, 1H), 4.72-4.59 (m, 1H), 4.50 (m, 1H), 4.42 (s, 1H), 2.43-2.31 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.62, 186.12, 152.67, 146.78, 138.91, 135.95, 135.57, 134.21, 133.59, 130.69, 129.31, 129.25, 128.93, 128.80, 128.65, 124.58, 123.81, 112.01, 69.10, 68.18, 66.81, 38.41. HRMS (ESI): Calcd. for $C_{26}H_{19}Cl_2N_2O$ $[M+H]^+$: 445.0874, Found: 445.0879.



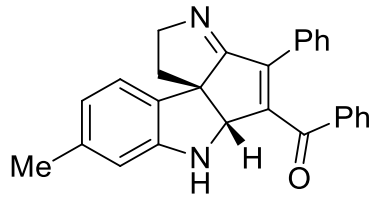
(**4c**): 48 mg, 46% yield, red solid. m.p. 172-174 °C. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.57-7.52 (m, 2H), 7.40-7.33 (m, 3H), 7.30 (d, *J* = 2.0 Hz, 1H), 7.22-7.16 (m, 3H), 7.14-7.08 (m, 3H), 6.51 (d, *J* = 8.3 Hz, 1H), 4.93 (s, 1H), 4.71-4.60 (m, 1H), 4.54-4.42 (m, 2H), 2.42-2.33 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 196.85, 186.41, 152.03, 147.32, 140.62, 136.18, 134.16, 133.87, 131.99, 130.43, 129.53, 129.46, 129.33, 128.61, 128.36, 126.65, 112.39, 111.42, 68.97, 68.03, 66.80, 38.47. HRMS (ESI): Calcd. for $C_{26}H_{20}BrN_2O$ $[M+H]^+$: 455.0759, Found: 455.0763.



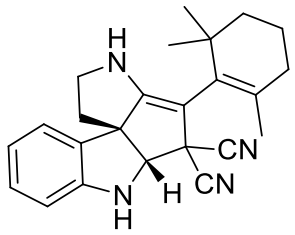
(**4d**): 45 mg, 56% yield, red solid. m.p. 140-141 °C.^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.58-7.49 (m, 2H), 7.40-7.29 (m, 3H), 7.21-7.07 (m, 5H), 7.09 (dd, *J* = 5.4, 3.6 Hz, 4H), 6.81 (d, *J* = 2.6 Hz, 1H), 6.70 (dd, *J* = 8.5, 2.5 Hz, 1H), 6.59 (d, *J* = 8.5 Hz, 1H), 4.91 (s, 1H), 4.66 (m, 1H), 4.47 (m, 1H), 4.23-4.04 (m, 1H), 3.77 (s, 3H), 2.42-2.29 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.21, 186.97, 154.58, 153.31, 142.10, 139.80, 136.35, 134.22, 133.75, 130.64, 129.36, 129.30, 128.52, 128.31, 114.35, 112.59, 109.81, 69.63, 69.03, 66.79, 55.97, 38.08. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O_2$ $[M+H]^+$: 407.1760, Found: 407.1770.



(**4e**): 56 mg, 64% yield, red solid. m.p. 142-143 ºC .^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.55 (dd, *J* = 8.2, 1.3 Hz, 2H), 7.37-7.32 (m, 1H), 7.28 (s, 1H), 7.26 (s, 1H), 7.17 (t, *J* = 7.8 Hz, 2H), 6.92 (d, *J* = 8.2 Hz, 2H), 6.81 (d, *J* = 2.5 Hz, 1H), 6.69 (dd, J = 8.5, 2.6 Hz, 1H), 6.57 (d, *J* = 8.5 Hz, 1H), 4.89 (s, 1H), 4.66 (m, 1H), 4.46 (dd, *J* = 14.9, 6.5 Hz, 1H), 4.17 (s, 1H), 3.76 (s, 3H), 2.45 (q, *J* = 7.6 Hz, 2H), 2.33 (t, *J* = 8.1 Hz, 2H), 1.05 (t, *J* = 7.6 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.40, 187.13, 154.52, 152.45, 145.71, 142.17, 139.73, 136.45, 134.29, 133.66, 129.44, 128.53, 127.87, 114.31, 112.54, 109.83, 69.63, 68.95, 66.75, 55.97, 38.07, 28.70, 15.39.



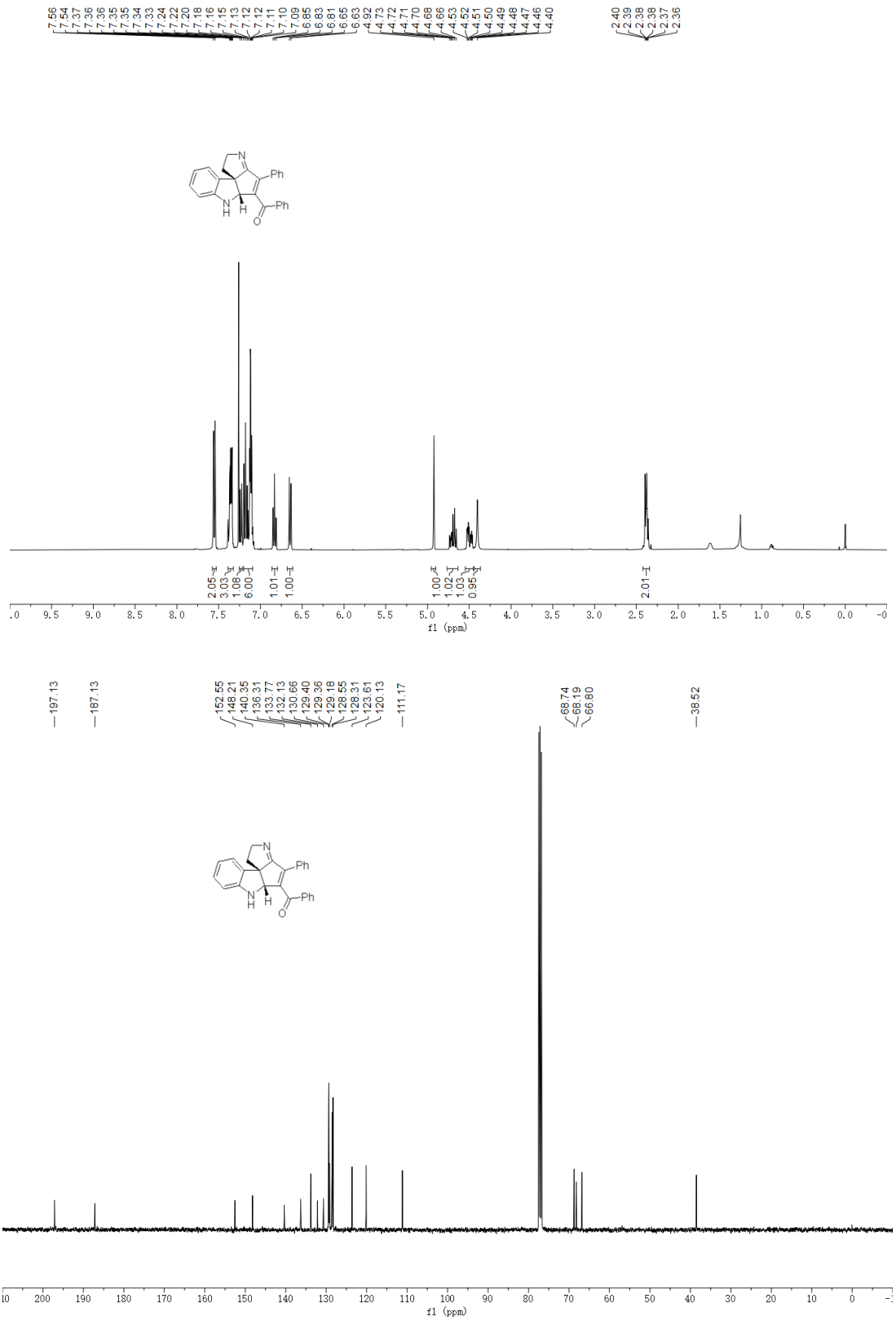
(**4f**): 41 mg, 53% yield, red solid. m.p. 153-155 ºC. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.56 (dd, *J* = 8.3, 1.4 Hz, 2H), 7.38-7.32 (m, 3H), 7.19-7.15 (m, 2H), 7.12-7.07 (m, 4H), 6.63 (d, *J* = 7.0 Hz, 1H), 6.45 (s, 1H), 4.90 (s, 1H), 4.66 (dt, *J* = 15.9, 8.1 Hz, 1H), 4.46 (dt, *J* = 15.8, 5.0 Hz, 1H), 4.37 (s, 1H), 2.37-2.31 (m, 2H), 2.25 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 197.13, 187.31, 152.62, 148.51, 140.25, 139.23, 136.33, 133.74, 130.75, 129.42, 129.40, 129.35, 129.29, 128.54, 128.28, 123.26, 120.91, 111.85, 68.98, 67.91, 66.68, 38.60, 21.66. HRMS (ESI): Calcd. for $C_{27}H_{23}N_2O$ $[M+H]^+$: 391.1810, Found: 391.1809.



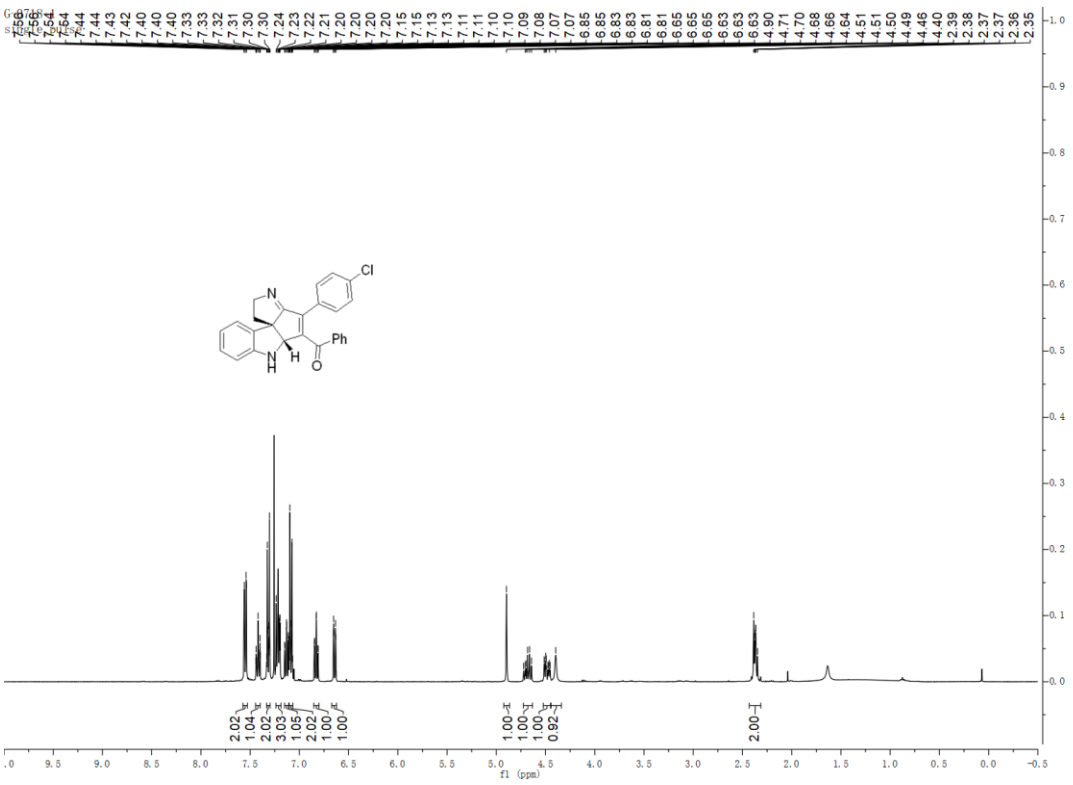
(**8**): 47 mg, 64% yield, viscious oil. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.20 (m, 2H), 6.86 (td, *J* = 7.5, 1.0 Hz, 1H), 6.82 (dd, J = 8.1, 1.0 Hz, 1H), 4.79 (s, 1H), 4.65 (s, 1H), 3.91 (td, *J* = 10.5, 5.8 Hz, 2H), 3.81-3.74 (m, 1H), 2.26-2.17 (m, 1H), 2.12 (m, 1H), 2.08 – 2.01 (m, 2H), 1.79-1.69 (m, 1H), 1.66 (s, 3H), 1.65-1.61 (m, 1H), 1.53 (m, 1H), 1.41 (dd, *J* = 12.2, 3.2 Hz, 1H), 1.12 (s, 3H), 0.89 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.07, 147.60, 135.87, 131.89, 129.78, 129.56, 122.51, 120.76, 115.79, 114.58, 110.62, 92.01, 77.68, 63.78, 57.03, 48.89, 39.88, 37.87, 34.94, 32.33, 29.48, 28.72, 21.40, 18.86. HRMS (ESI): Calcd. for $C_{24}H_{27}N_4$ $[M+H]^+$: 371.2236, Found: 371.2239.

4. ^{1}H NMR and ^{13}C NMR Spectra of All Compounds

Compound **3a**



Compound **3b**


Cl
N
Ph
O
H
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2.02
3.03
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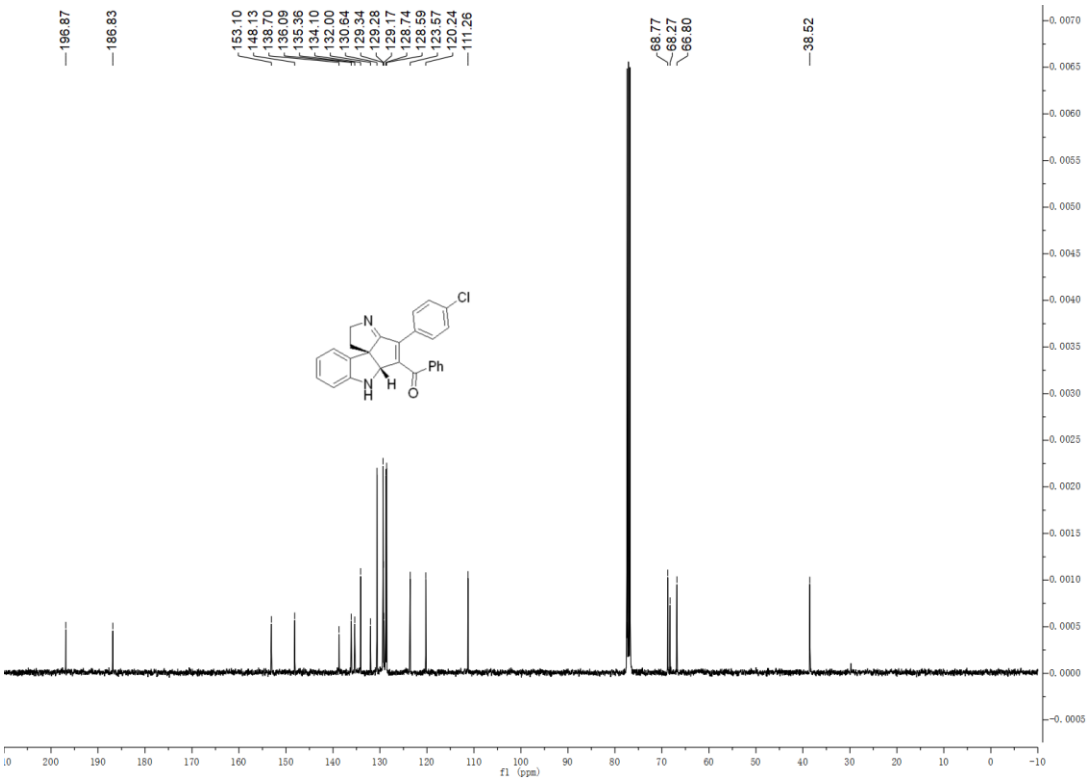
2.02
1.00
1.00
1.00
1.00
0.92
2.00
fl (ppm)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186.83
153.10
148.13
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134.10
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129.28
129.17
128.74
128.59
123.57
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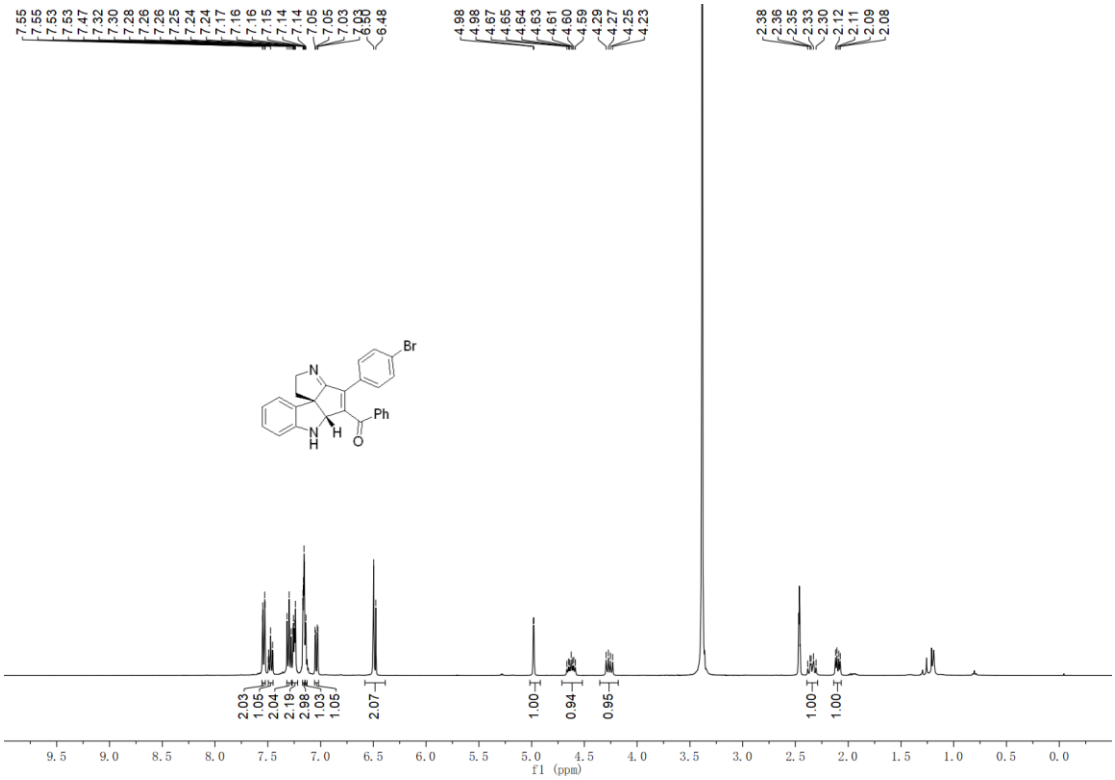
68.77
68.27
66.80
38.52
Cl
N
Ph
O
H
fl (ppm)

Compound **3c**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4.25
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2.35
2.33
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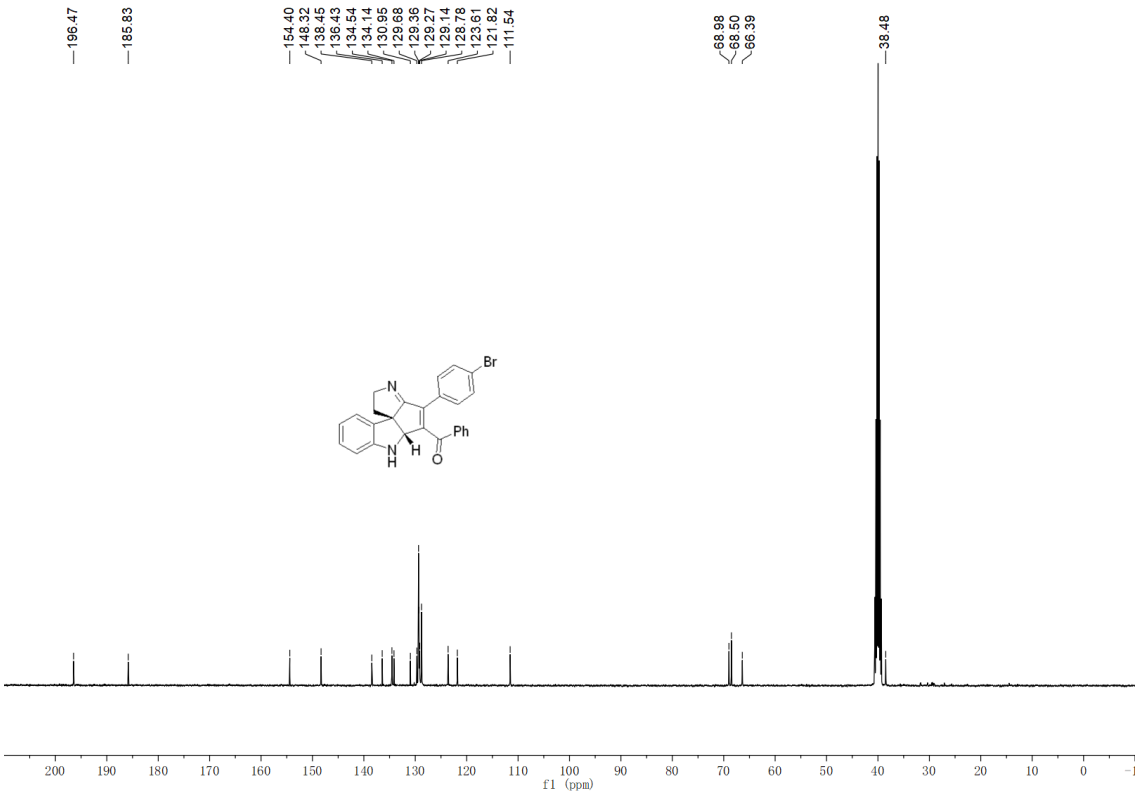
2.11
2.09
2.08
Br
N
Ph
O
N
H
2.03
1.05
2.04
2.19
2.98
1.03
1.05
2.07
1.00
0.94
0.95
1.00
1.00
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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129.36
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128.78
123.61
121.82
111.54
68.98
68.50
66.39
38.48
Br
N
Ph
O
N
H
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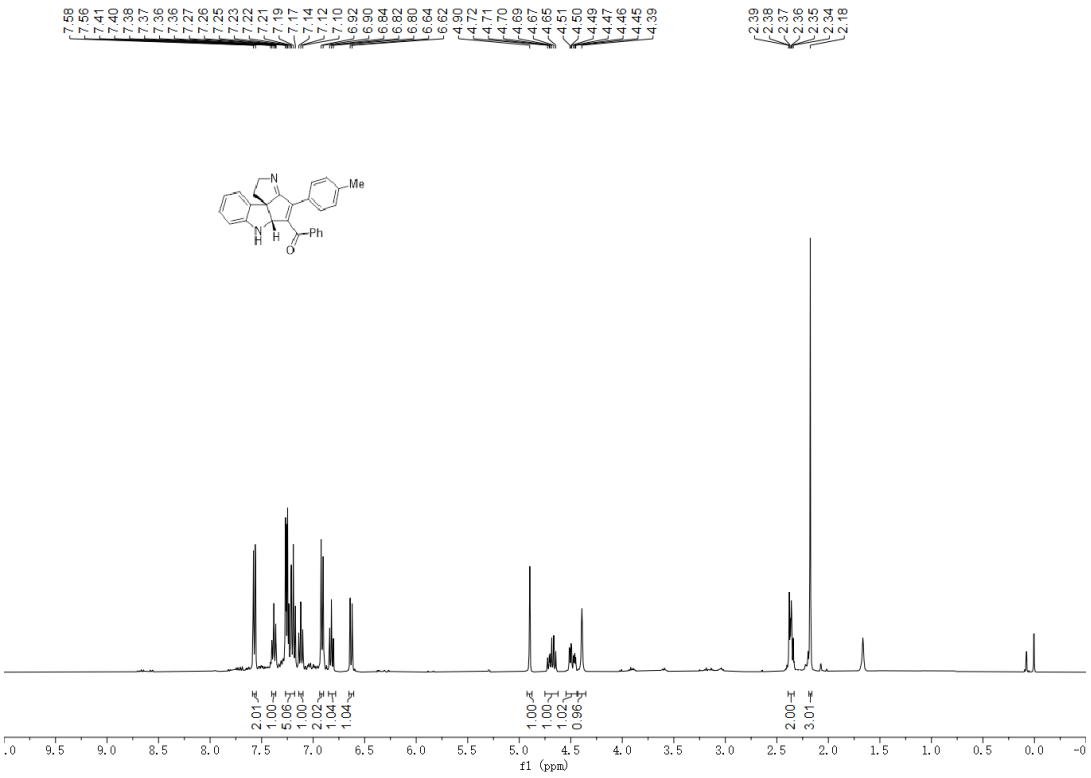
f1 (ppm)

Compound **3d**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Me
Ph
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1.00
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1.04
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1.00
1.00
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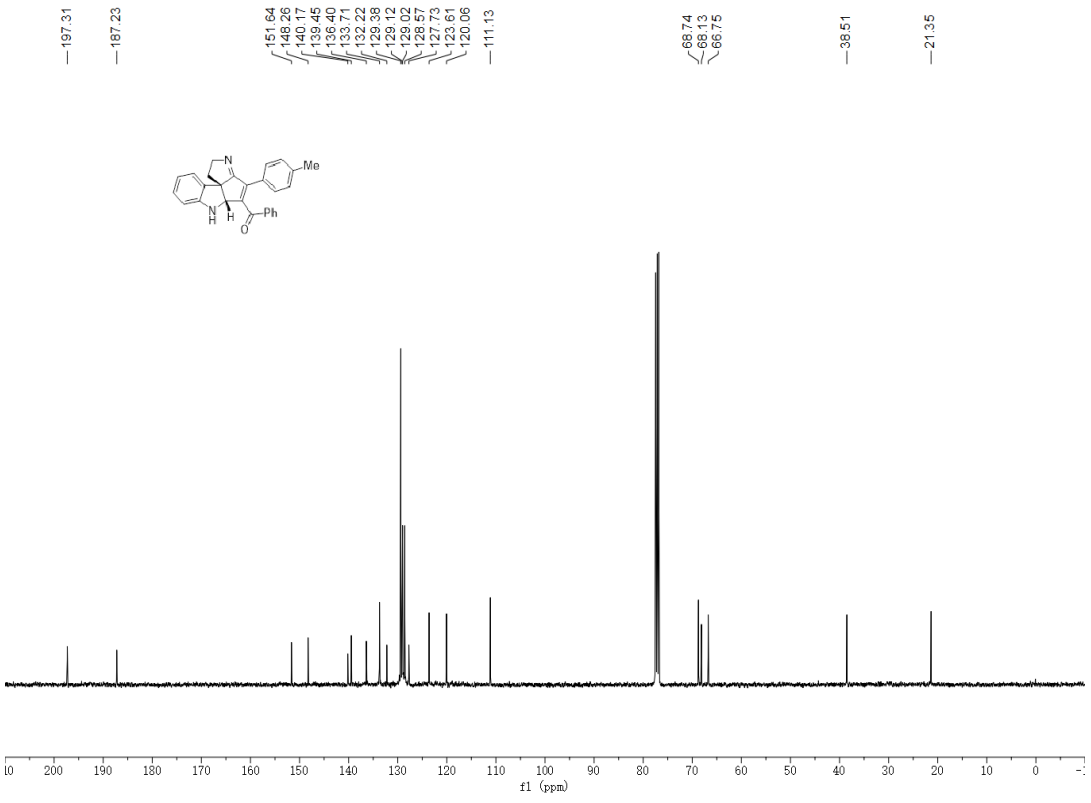
0.96
2.00
3.01
f1 (ppm)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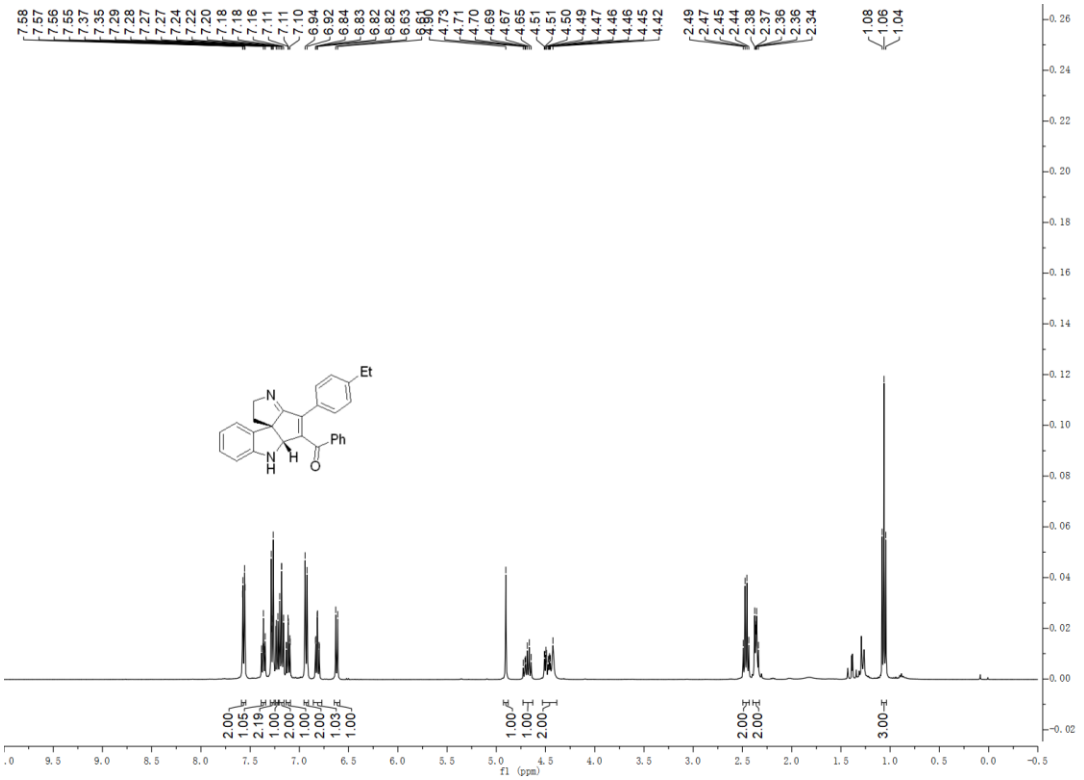
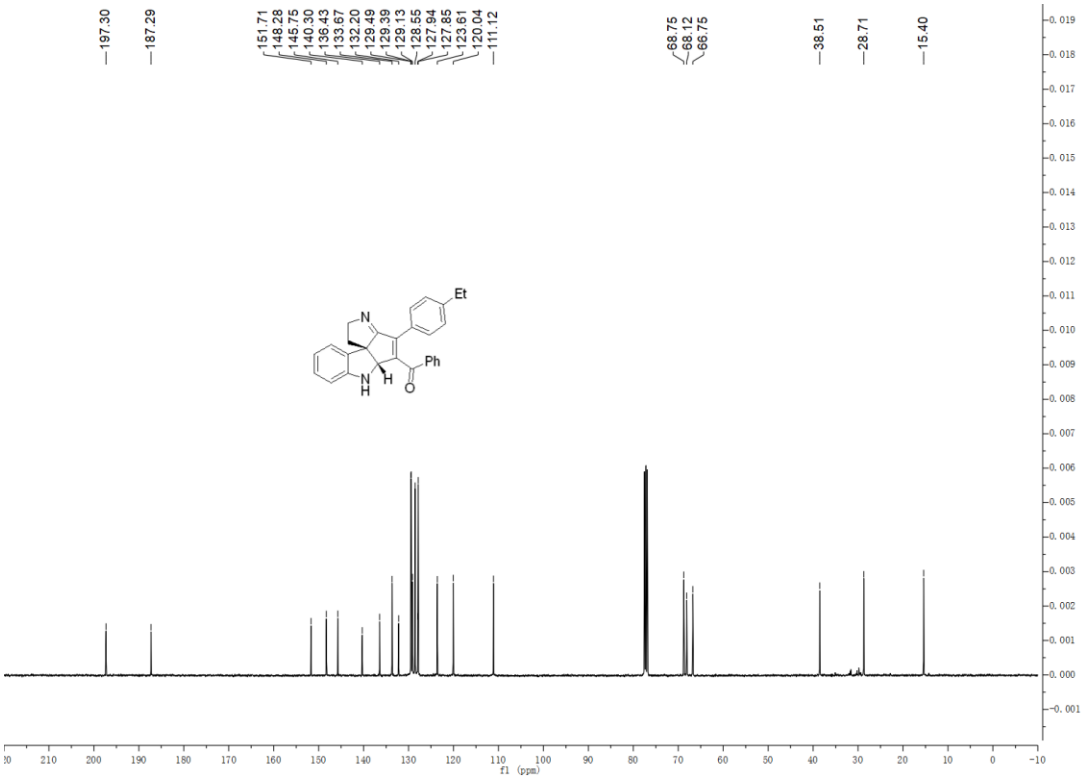
187.23
151.64
148.26
140.17
139.45
136.40
133.71
132.22
129.38
129.12
129.02
128.57
127.73
123.61
120.06
111.13
68.74
68.13
66.75
38.51
21.35
Me
Ph
f1 (ppm)

Compound **3e**


7.58 7.57 7.56 7.55 7.37 7.35 7.29 7.28 7.27 7.27 7.24 7.22 7.20 7.18 7.18 7.16 7.11 7.11 7.10 6.94 6.92 6.84 6.83 6.82 6.82 6.63 6.61 4.90 4.73 4.71 4.70 4.69 4.67 4.65 4.51 4.51 4.50 4.49 4.47 4.46 4.46 4.45 4.42 2.49 2.47 2.45 2.44 2.38 2.37 2.36 2.36 2.34 1.08 1.06 1.04
Et
N
Ph
N
H
O
2.00 1.05 2.19 1.00 2.00 1.00 2.00 1.03 1.00 1.00 1.00 2.00 2.00 2.00 3.00
f1 (ppm)


197.30 187.29 151.71 148.28 145.75 140.30 136.43 133.67 132.20 129.49 129.39 129.13 128.55 127.94 127.85 123.61 120.04 111.12 68.75 68.12 66.75 38.51 28.71 15.40
Et
N
Ph
N
H
O
f1 (ppm)

Compound **3f**


7.48
7.47
7.46
7.46
7.40
7.39
7.38
7.38
7.37
7.35
7.22
7.21
7.14
7.13
7.12
7.11
7.11
7.09
7.09
6.99
6.97
6.83
6.81
6.81
6.63
6.61
4.88
4.72
4.70
4.68
4.66
4.64
4.51
4.50
4.49
4.47
4.46
4.45
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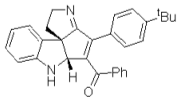
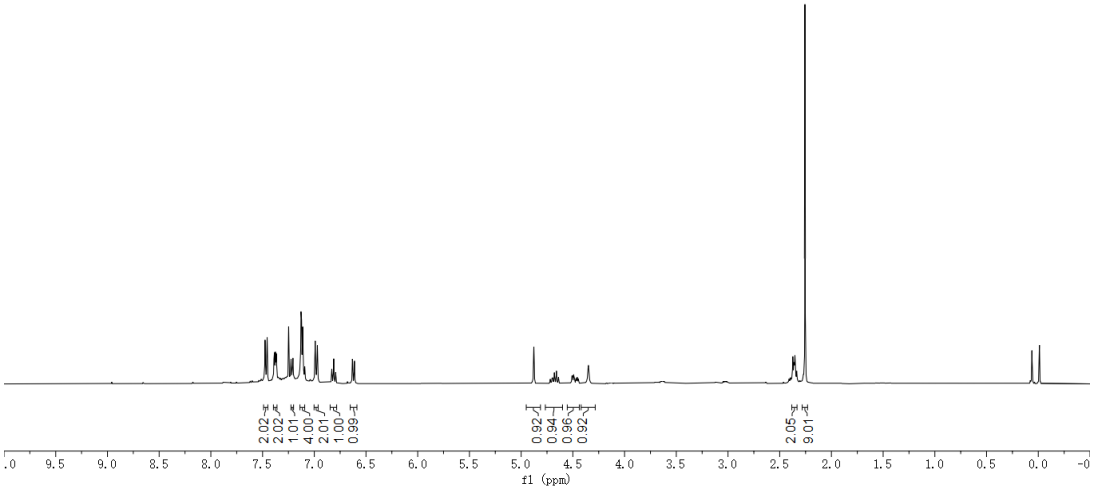
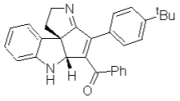
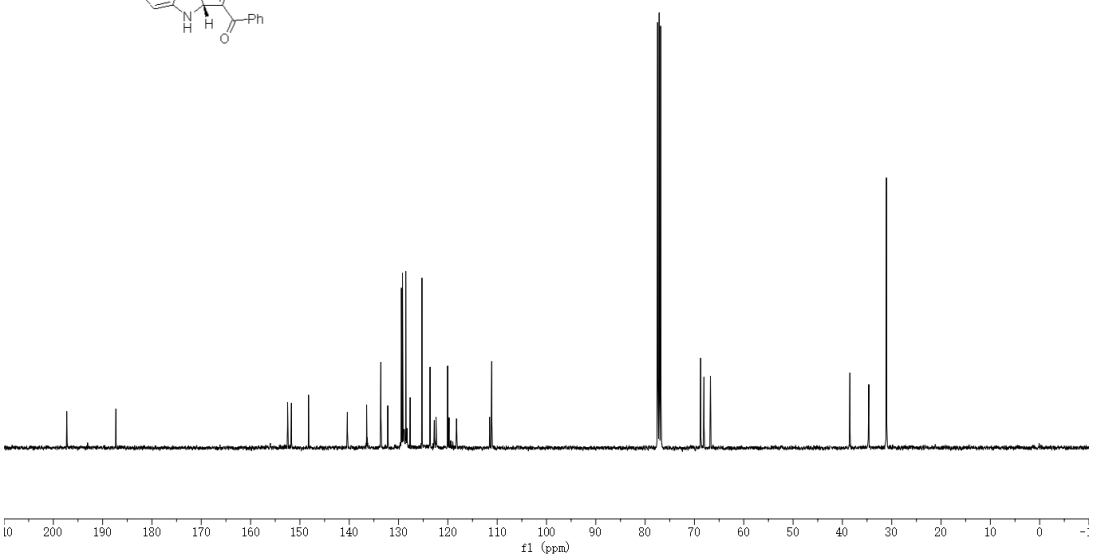
2.36
2.36
2.35
2.35
2.34
2.26


tBu
N
H
H
Ph
O

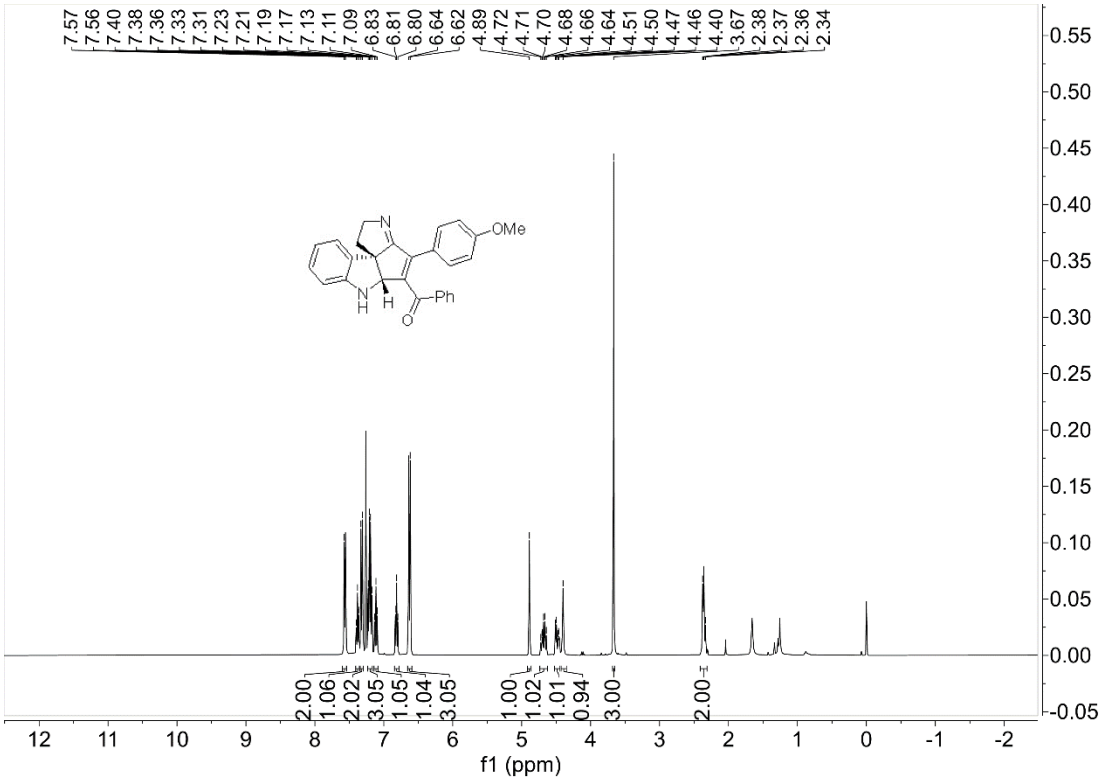

2.02
2.02
1.01
4.00
2.01
1.00
0.99
0.92
0.94
0.96
0.92
2.05
9.01
f1 (ppm)

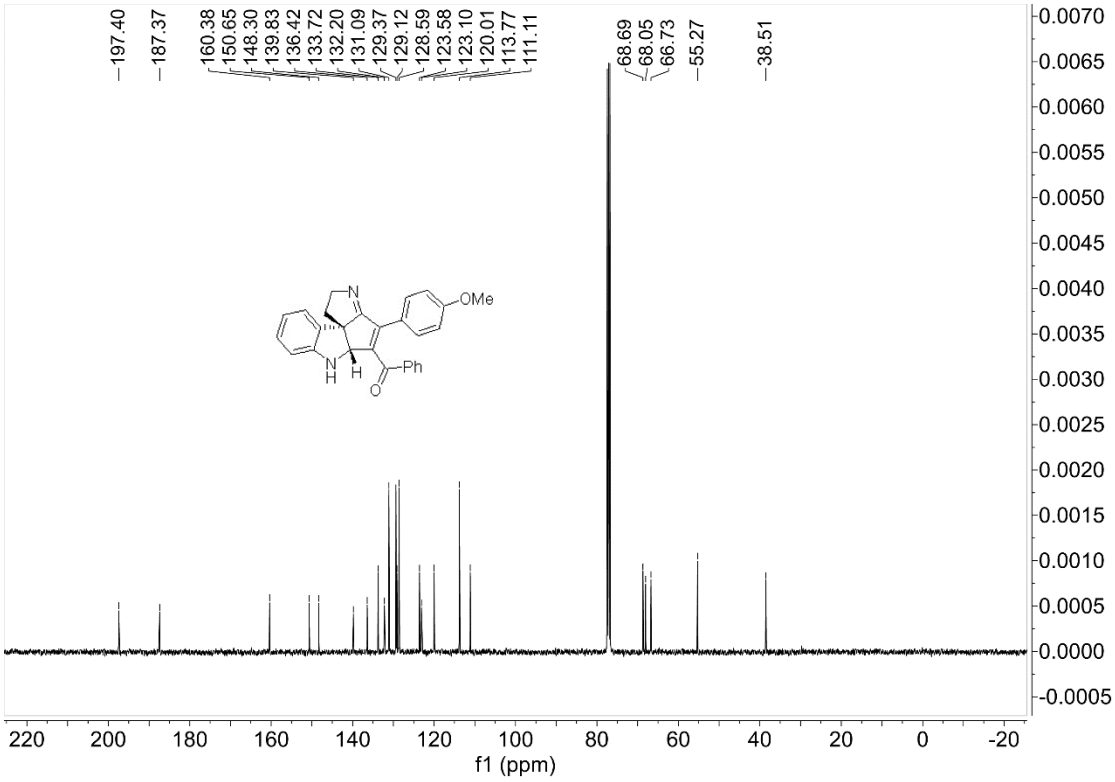

197.27
187.30
152.49
151.72
148.25
140.37
136.47
133.59
132.18
129.42
129.20
129.12
128.53
127.64
125.25
123.61
122.75
122.40
120.05
119.75
118.24
111.53
111.10
68.74
68.07
66.76
38.48
34.64
31.07


tBu
N
H
H
Ph
O

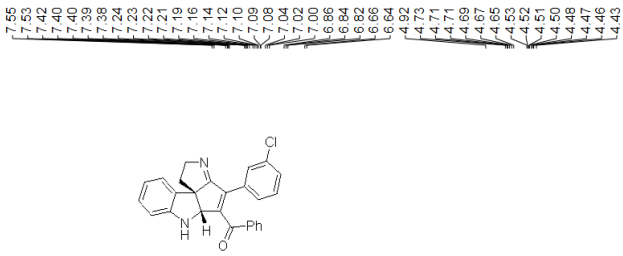

f1 (ppm)

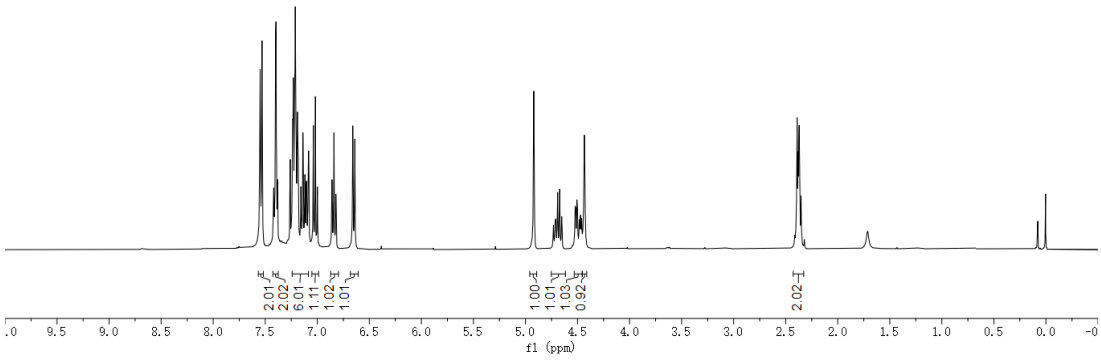
Compound **3g**

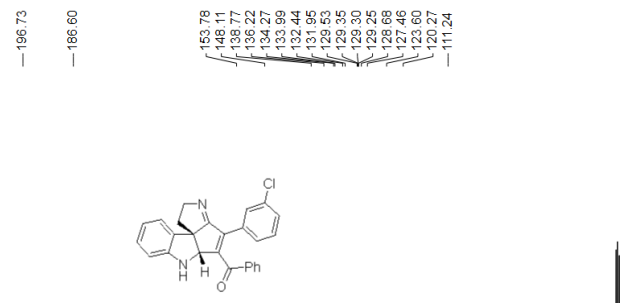

7.57
7.56
7.40
7.38
7.36
7.33
7.31
7.23
7.21
7.19
7.17
7.13
7.11
7.09
6.83
6.81
6.80
6.64
6.62
4.89
4.72
4.71
4.70
4.68
4.66
4.64
4.51
4.50
4.47
4.46
4.40
3.67
2.38
2.37
2.36
2.34
OMe
Ph
2.00
1.06
2.02
3.05
1.05
1.04
3.05
1.00
1.02
1.01
0.94
3.00
2.00
f1 (ppm)

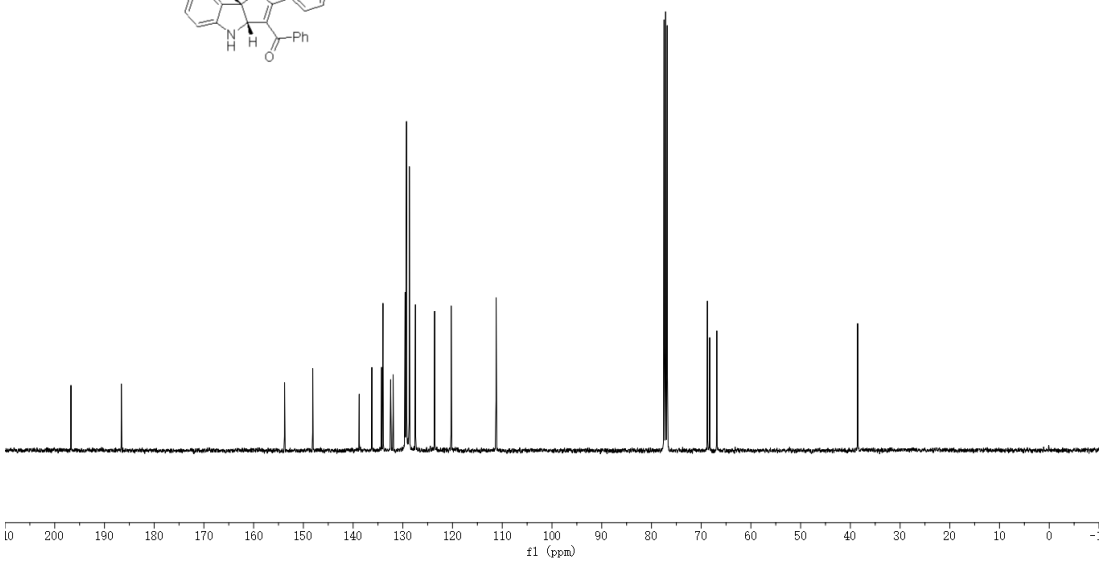

197.40
187.37
160.38
150.65
148.30
139.83
136.42
133.72
132.20
131.09
129.37
129.12
128.59
123.58
123.10
120.01
113.77
111.11
68.69
68.05
66.73
55.27
38.51
OMe
Ph
f1 (ppm)

Compound **3h**

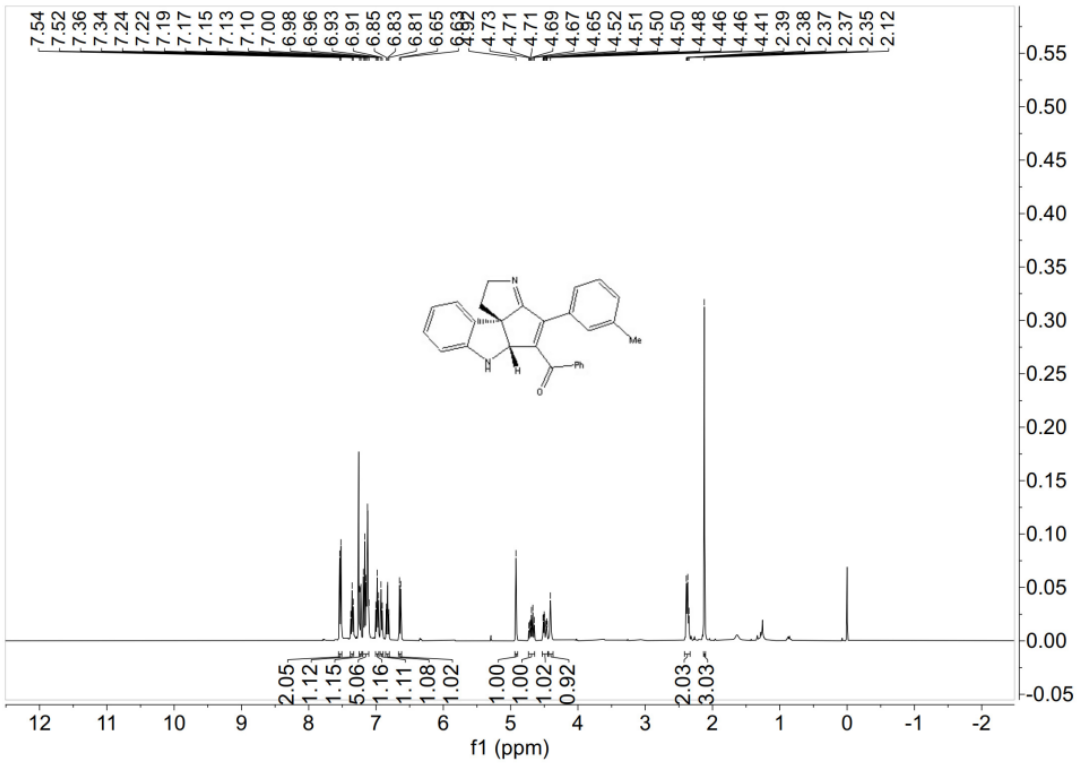

7.55
7.53
7.42
7.40
7.40
7.39
7.38
7.24
7.23
7.22
7.21
7.19
7.16
7.14
7.12
7.10
7.09
7.08
7.04
7.02
7.00
6.86
6.84
6.82
6.66
6.64
4.92
4.73
4.71
4.71
4.69
4.67
4.65
4.53
4.52
4.51
4.50
4.48
4.47
4.46
4.43
2.40
2.39
2.38
2.38
2.37
2.35
Cl
N
H
H
O
Ph

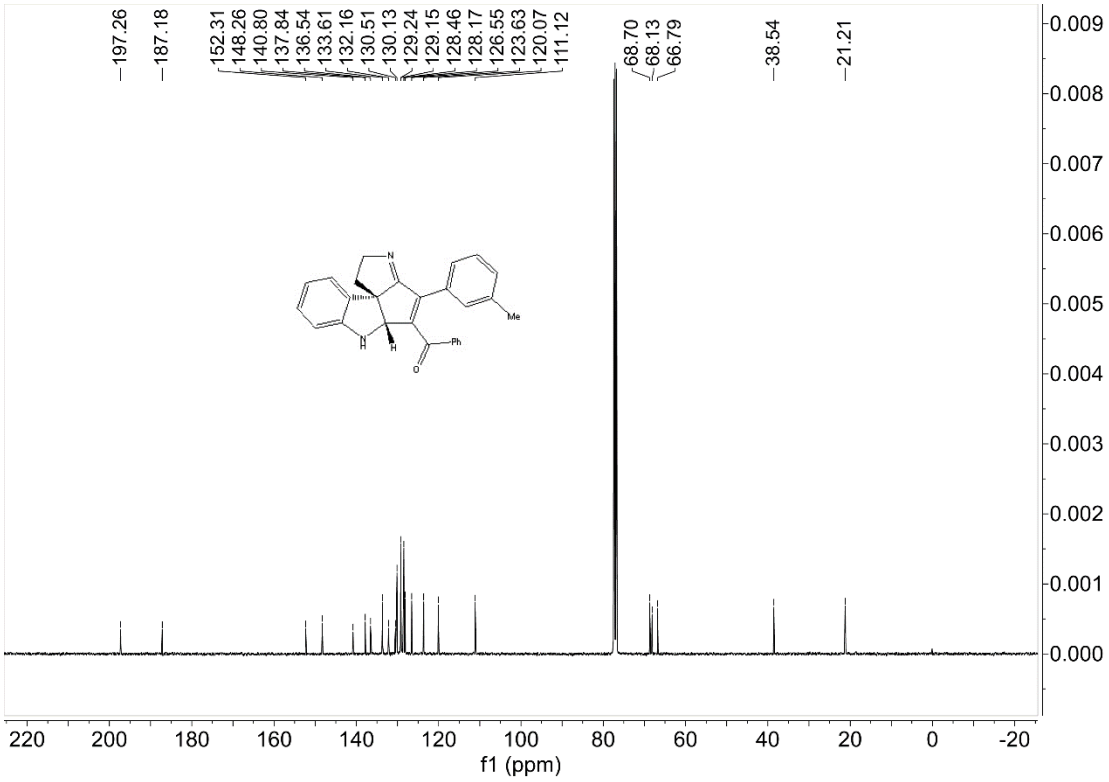

2.01
2.02
6.01
1.11
1.02
1.01
1.00
1.01
1.03
0.92
2.02
f1 (ppm)


196.73
186.60
153.78
148.11
138.77
136.22
134.27
133.99
132.44
131.95
129.53
129.35
129.30
129.25
128.68
127.46
123.60
120.27
111.24
68.75
68.28
66.85
38.53
Cl
N
H
H
O
Ph

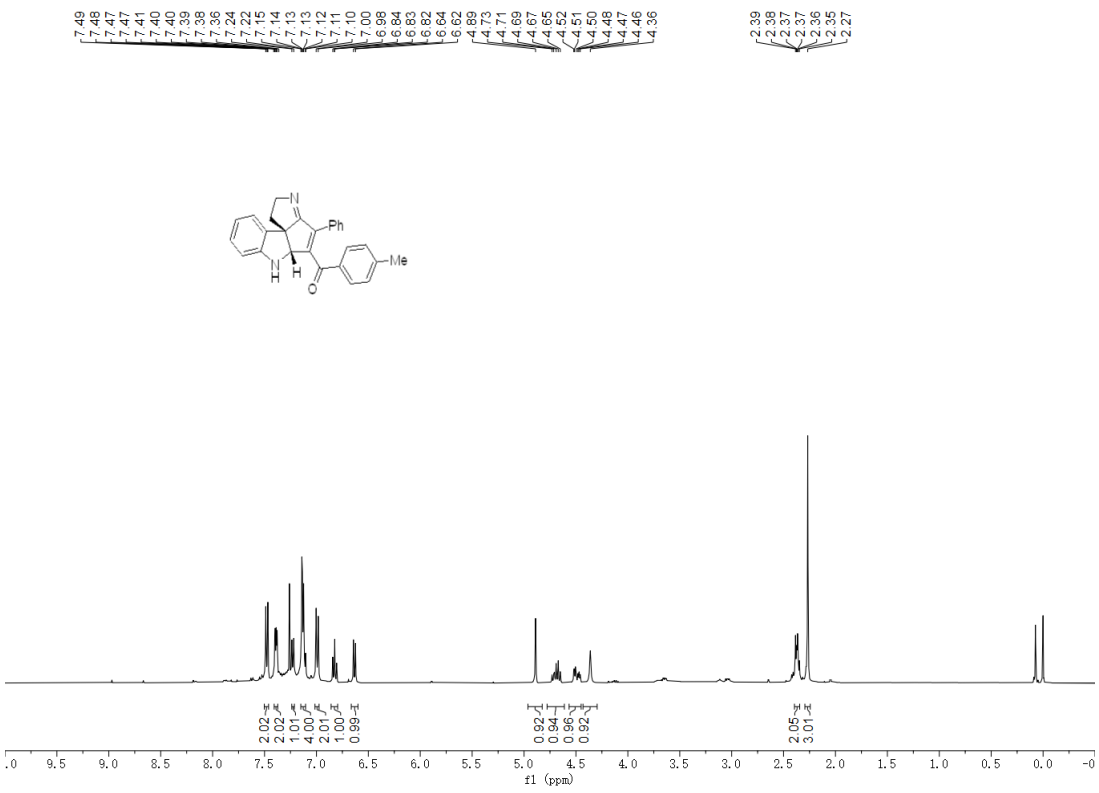

f1 (ppm)

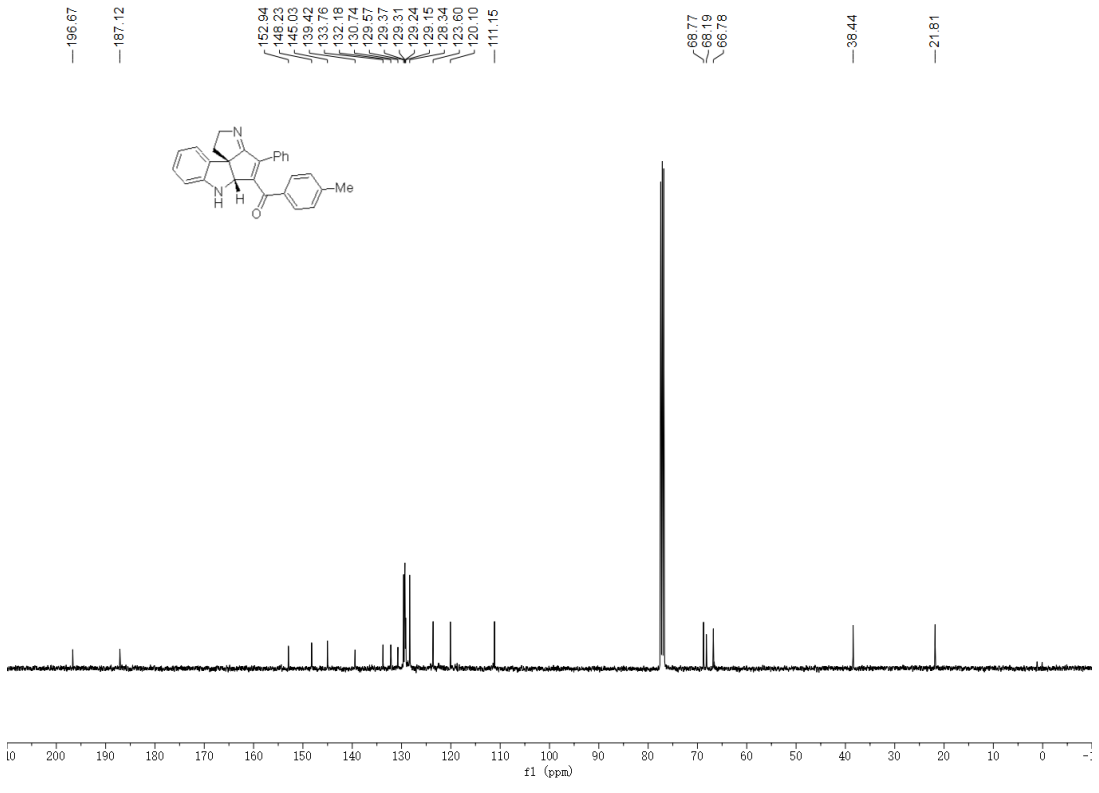
Compound **3i**


7.54
7.52
7.36
7.34
7.24
7.22
7.19
7.17
7.15
7.13
7.10
7.00
6.98
6.96
6.93
6.91
6.85
6.83
6.81
6.65
4.73
4.71
4.71
4.69
4.67
4.65
4.52
4.51
4.50
4.50
4.48
4.46
4.46
4.41
2.39
2.38
2.37
2.37
2.35
2.12
2.05
1.12
1.15
5.06
1.16
1.11
1.08
1.02
1.00
1.00
1.02
0.92
2.03
3.03
f1 (ppm)

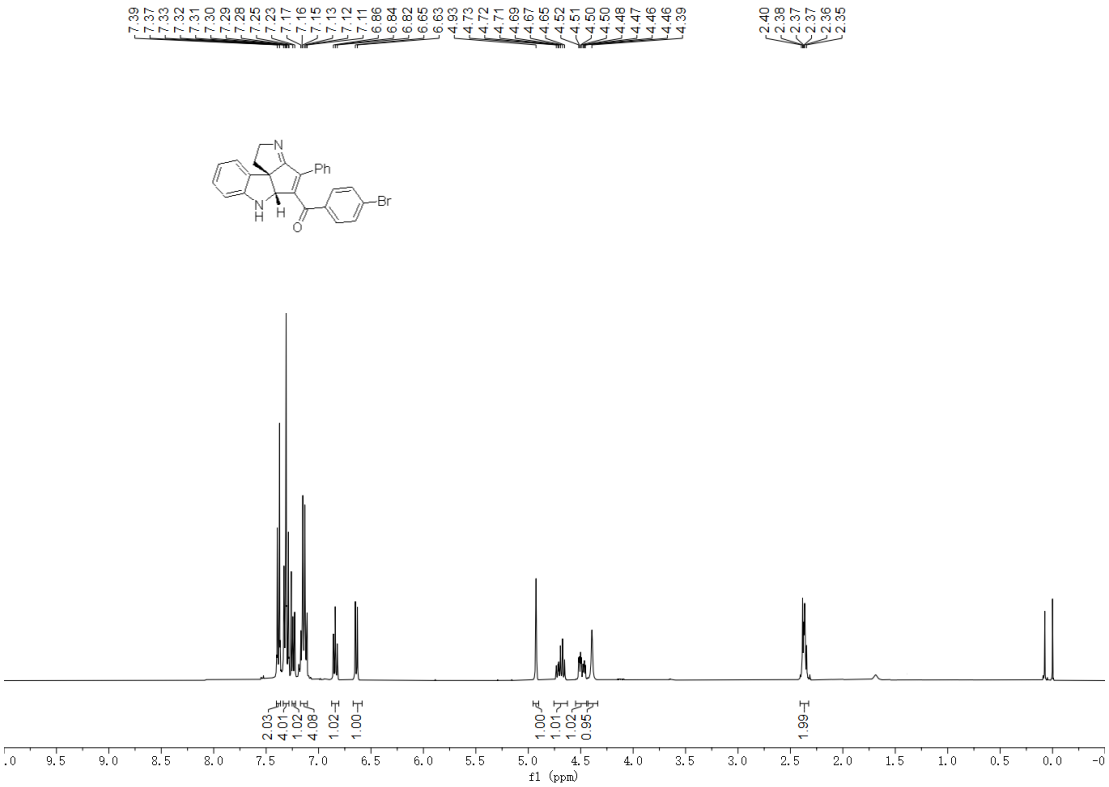

197.26
187.18
152.31
148.26
140.80
137.84
136.54
133.61
132.16
130.51
130.13
129.24
129.15
128.46
128.17
126.55
123.63
120.07
111.12
68.70
68.13
66.79
38.54
21.21
f1 (ppm)

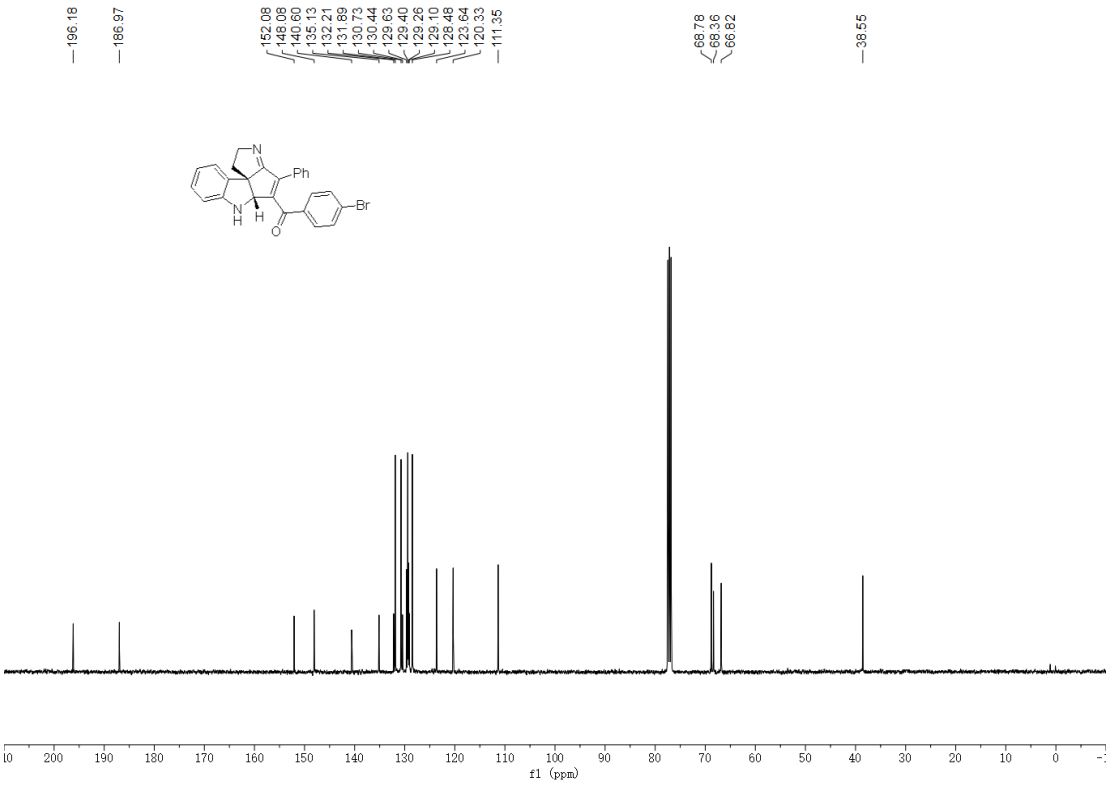
Compound **3j**


7.49
7.48
7.47
7.47
7.41
7.40
7.40
7.39
7.38
7.36
7.24
7.22
7.15
7.14
7.13
7.13
7.12
7.11
7.10
7.00
6.98
6.84
6.83
6.82
6.64
6.62
4.89
4.73
4.71
4.69
4.67
4.65
4.52
4.51
4.50
4.48
4.47
4.46
4.36
2.39
2.38
2.37
2.37
2.36
2.35
2.27
Ph
Me
N
H
O
2.02
2.02
1.01
4.00
2.01
1.00
0.99
0.92
0.94
0.96
0.92
2.05
3.01
f1 (ppm)

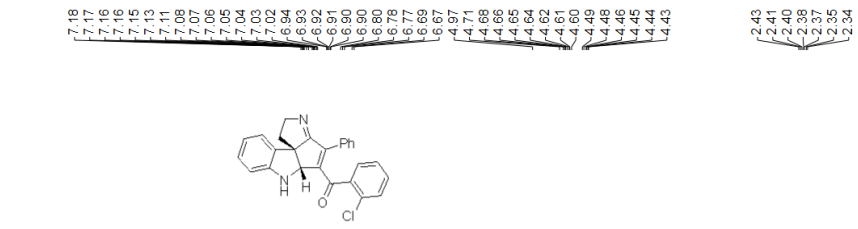

196.67
187.12
152.94
148.23
145.03
139.42
133.76
132.18
130.74
129.57
129.37
129.31
129.24
129.15
128.34
123.60
120.10
111.15
68.77
68.19
66.78
38.44
21.81
Ph
Me
N
H
O
f1 (ppm)

Compound **3k**


7.39
7.37
7.33
7.32
7.31
7.30
7.29
7.28
7.25
7.23
7.17
7.16
7.15
7.13
7.12
7.11
6.86
6.84
6.82
6.65
6.63
4.93
4.73
4.72
4.71
4.69
4.67
4.65
4.52
4.51
4.50
4.50
4.48
4.47
4.46
4.46
4.39
2.40
2.38
2.37
2.37
2.36
2.35
Ph
Br
2.03
4.01
1.02
4.08
1.02
1.00
1.00
1.01
1.02
0.95
1.99
f1 (ppm)


196.18
186.97
152.08
148.08
140.60
135.13
132.21
131.89
130.73
130.44
129.63
129.40
129.26
129.10
128.48
123.64
120.33
111.35
68.78
68.36
66.82
38.55
Ph
Br
f1 (ppm)

Compound **3l**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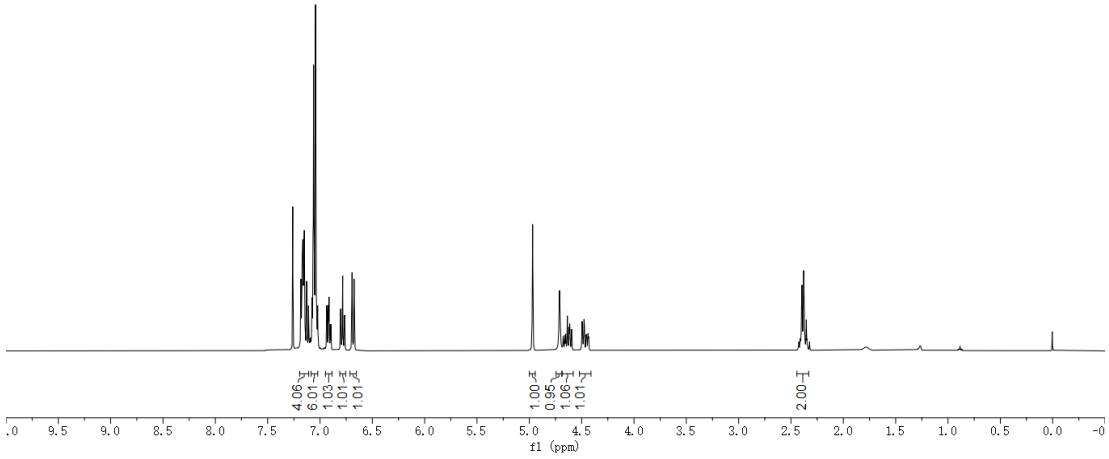
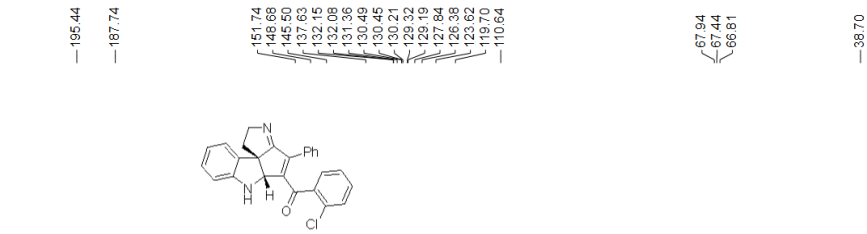
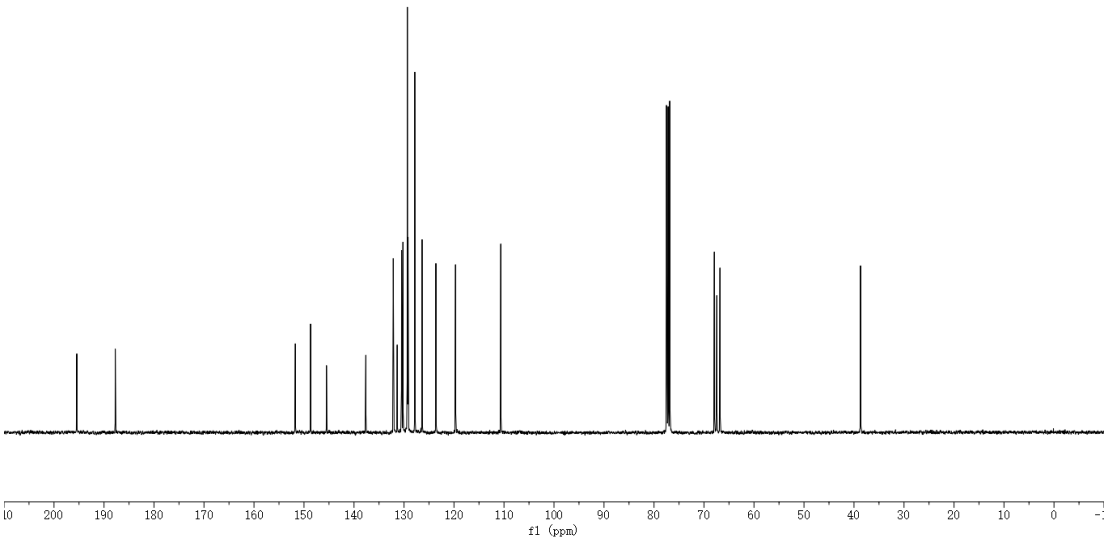
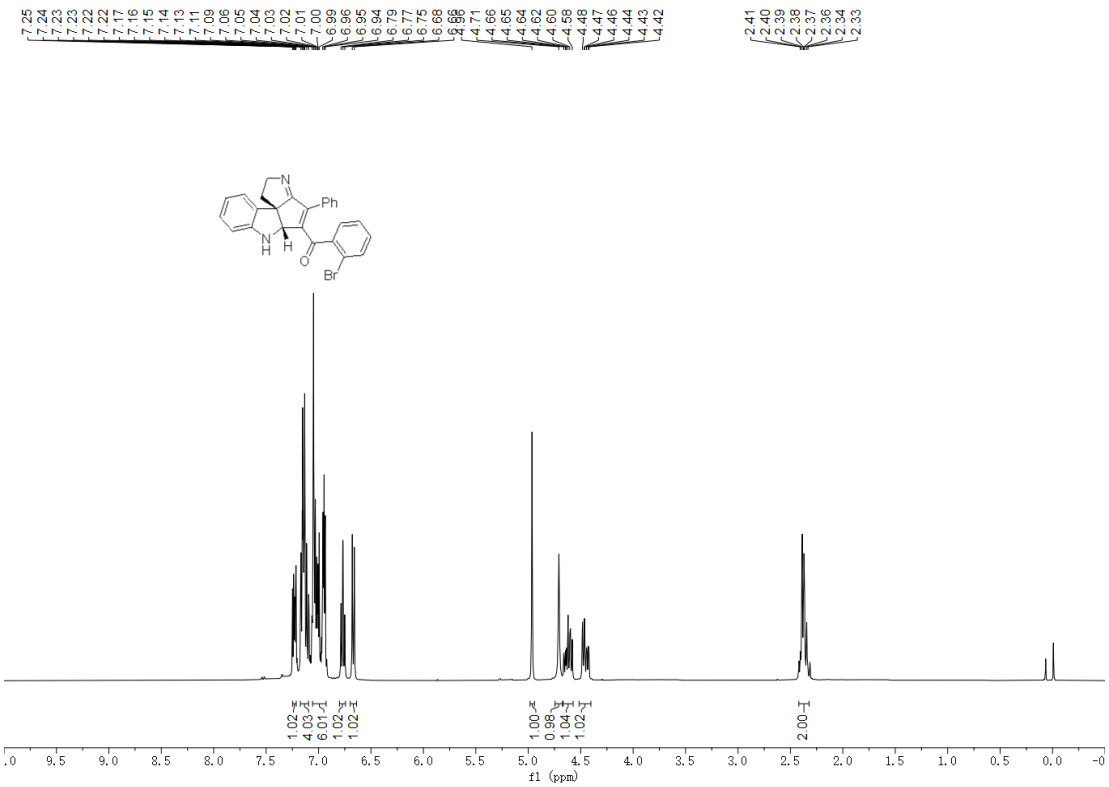
7.17
7.16
7.16
7.15
7.13
7.11
7.08
7.07
7.06
7.05
7.04
7.03
7.02
6.94
6.93
6.92
6.91
6.90
6.90
6.80
6.78
6.77
6.69
6.67
4.97
4.71
4.68
4.66
4.65
4.64
4.62
4.61
4.60
4.49
4.48
4.46
4.45
4.44
4.43
2.43
2.41
2.40
2.38
2.37
2.35
2.34
N
Ph
N
H
O
Cl

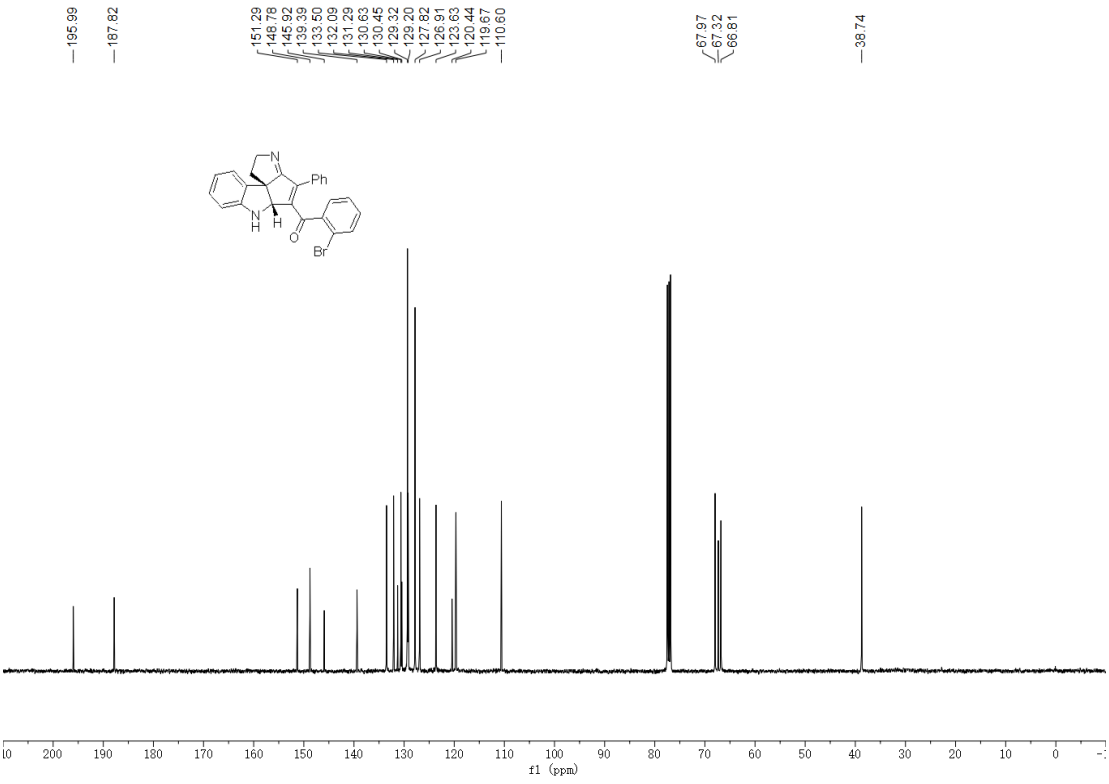

4.06
6.01
1.03
1.01
1.01
1.00
0.95
1.06
1.01
2.00
f1 (ppm)


195.44
187.74
151.74
148.68
145.50
137.63
132.15
132.08
131.36
130.49
130.45
130.21
129.32
129.19
127.84
126.38
123.62
119.70
110.64
67.94
67.44
66.81
38.70
N
Ph
N
H
O
Cl

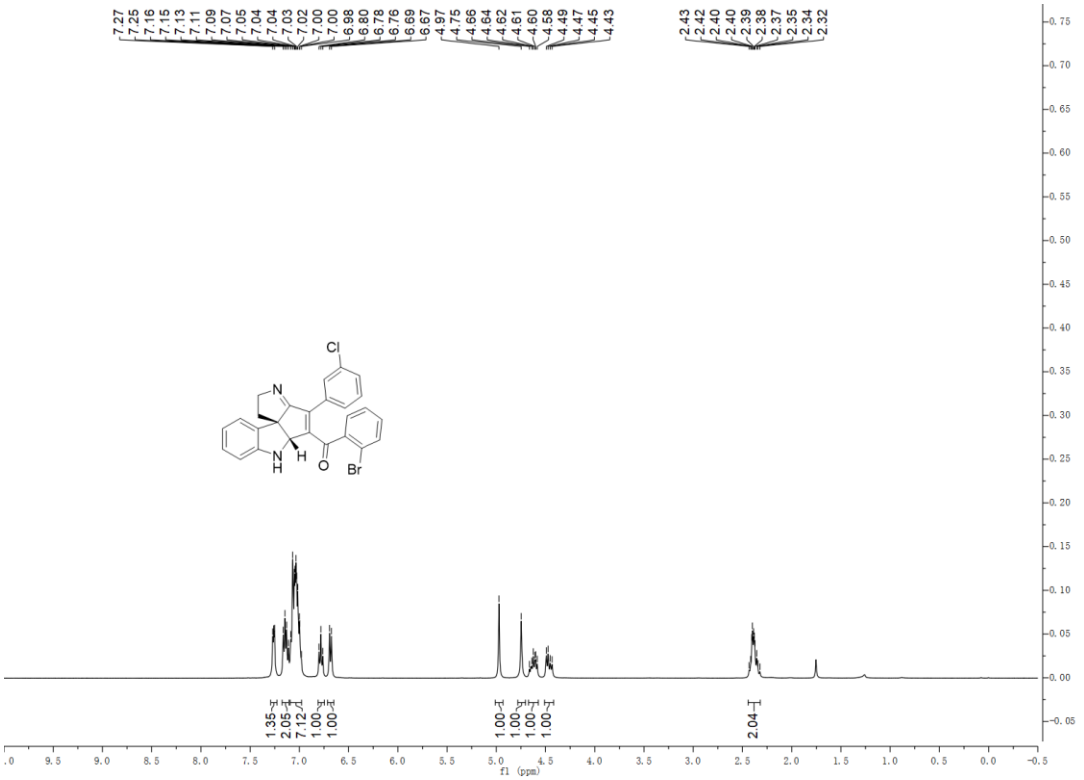

f1 (ppm)

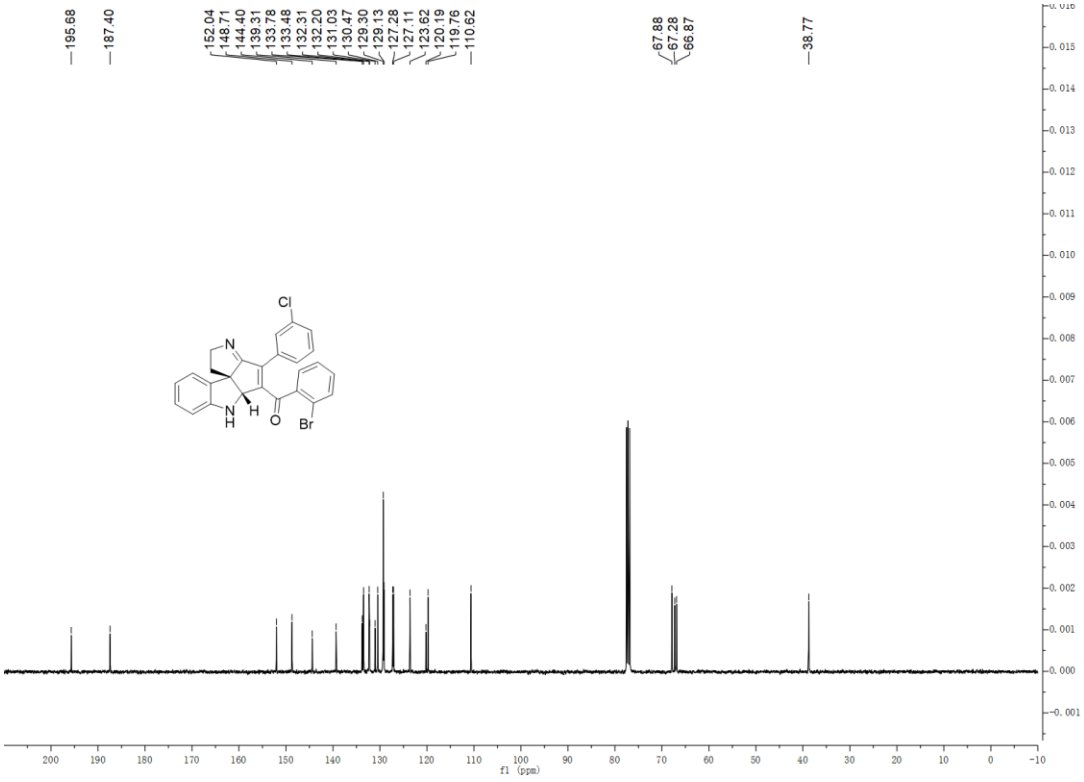
Compound **3m**


7.25
7.24
7.23
7.23
7.22
7.22
7.17
7.16
7.15
7.14
7.13
7.11
7.09
7.06
7.05
7.04
7.03
7.02
7.01
7.00
6.99
6.96
6.95
6.94
6.79
6.77
6.75
6.68
4.71
4.66
4.65
4.64
4.62
4.60
4.58
4.48
4.47
4.46
4.44
4.43
4.42
2.41
2.40
2.39
2.38
2.37
2.36
2.34
2.33
Ph
Br
1.02
4.03
6.01
1.02
1.02
1.00
0.98
1.04
1.02
2.00
f1 (ppm)

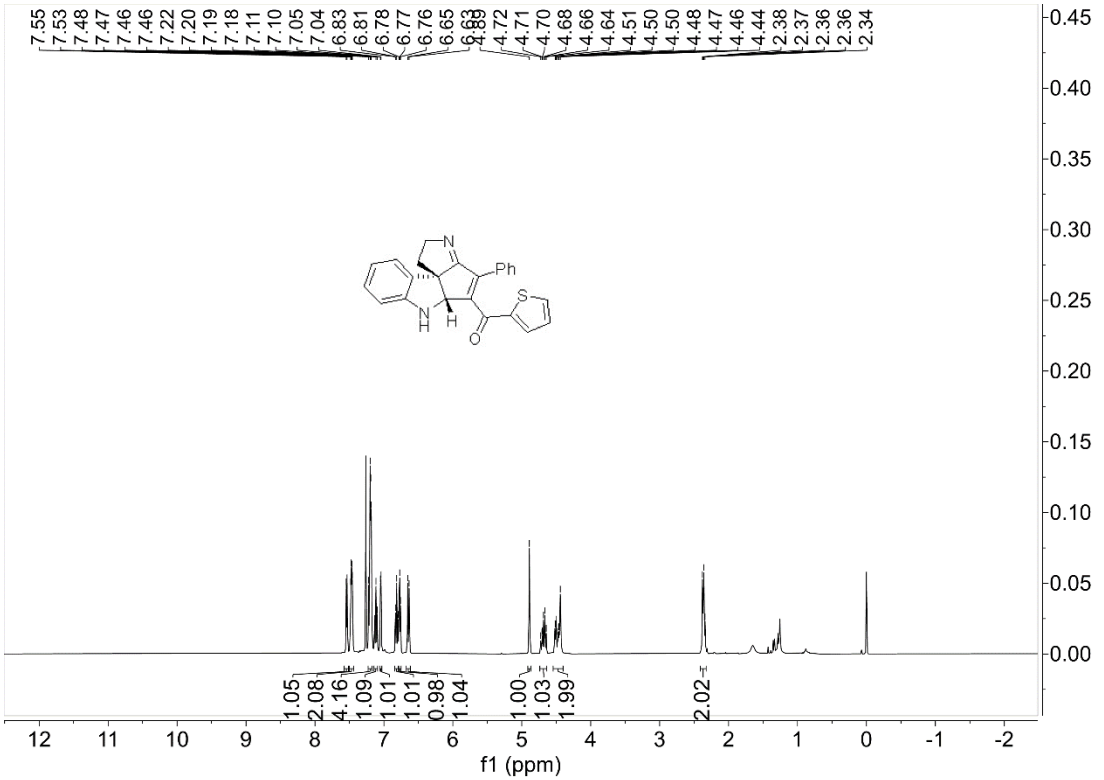

195.99
187.82
151.29
148.78
145.92
139.39
133.50
132.09
131.29
130.63
130.45
129.32
129.20
127.82
126.91
123.63
120.44
119.67
110.60
67.97
67.32
66.81
38.74
Ph
Br
f1 (ppm)

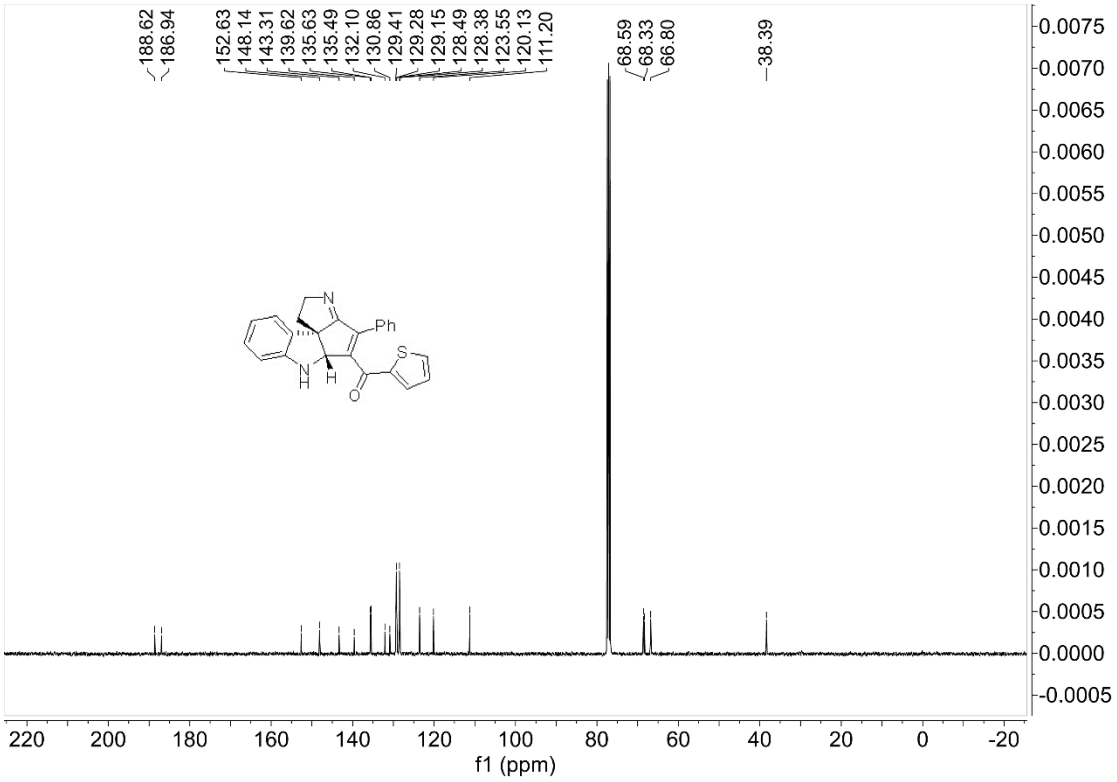
Compound **3n**


7.27
7.25
7.16
7.15
7.13
7.11
7.09
7.07
7.05
7.04
7.04
7.03
7.02
7.00
7.00
6.98
6.80
6.78
6.76
6.69
6.67
4.97
4.75
4.66
4.64
4.62
4.61
4.60
4.58
4.49
4.47
4.45
4.43
2.43
2.42
2.40
2.40
2.39
2.38
2.37
2.35
2.34
2.32
Cl
N
N
H
O
Br
1.35
2.05
7.12
1.00
1.00
1.00
1.00
1.00
2.04
f1 (ppm)

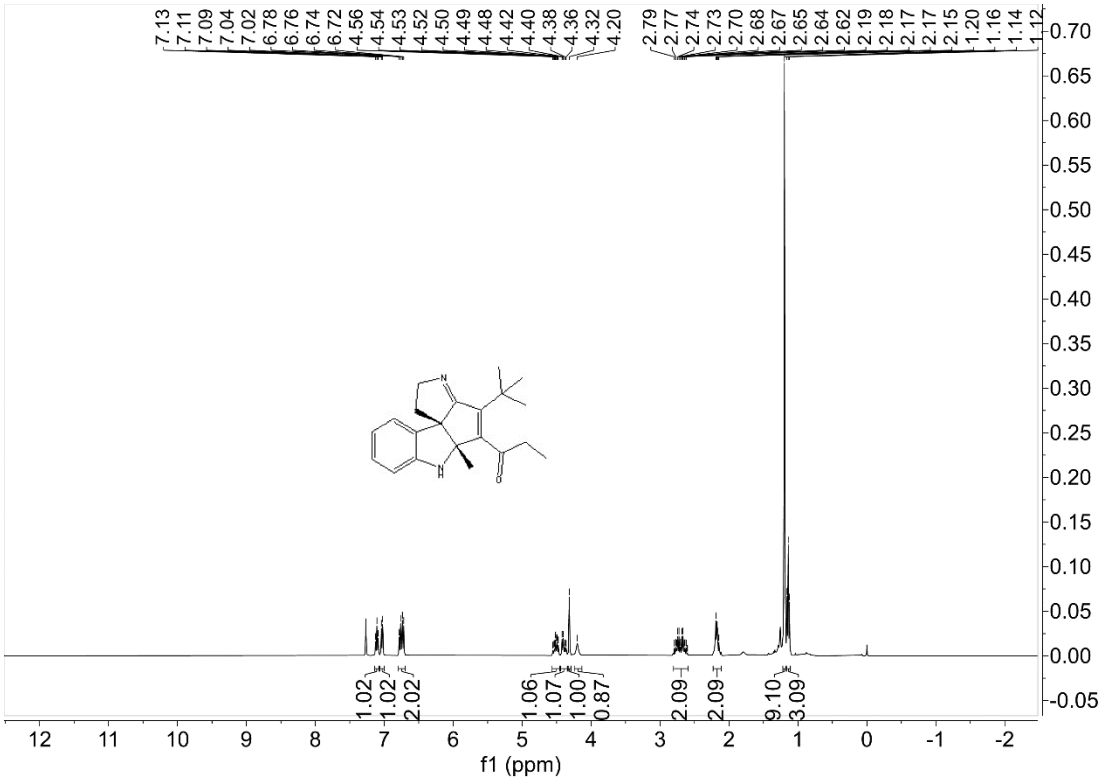

195.68
187.40
152.04
148.71
144.40
139.31
133.78
133.48
132.31
132.20
131.03
130.47
129.30
129.13
127.28
127.11
123.62
120.19
119.76
110.62
67.88
67.28
66.87
38.77
Cl
N
N
H
O
Br
f1 (ppm)

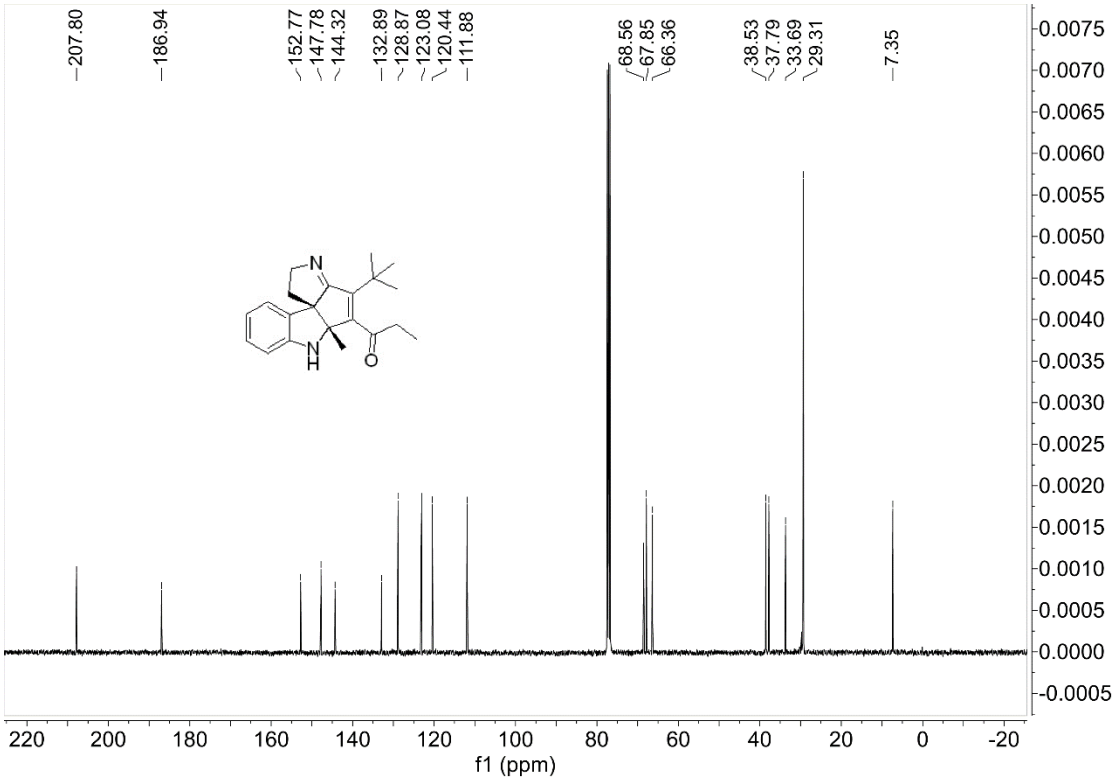
Compound **3o**


7.55
7.53
7.48
7.47
7.46
7.46
7.22
7.20
7.19
7.18
7.11
7.10
7.05
7.04
6.83
6.81
6.78
6.77
6.76
6.65
4.72
4.71
4.70
4.68
4.66
4.64
4.51
4.50
4.50
4.48
4.47
4.46
4.44
2.38
2.37
2.36
2.36
2.34
1.05
2.08
4.16
1.09
1.01
1.01
0.98
1.04
1.00
1.03
1.99
2.02
f1 (ppm)


188.62
186.94
152.63
148.14
143.31
139.62
135.63
135.49
132.10
130.86
129.41
129.28
129.15
128.49
128.38
123.55
120.13
111.20
68.59
68.33
66.80
38.39
f1 (ppm)

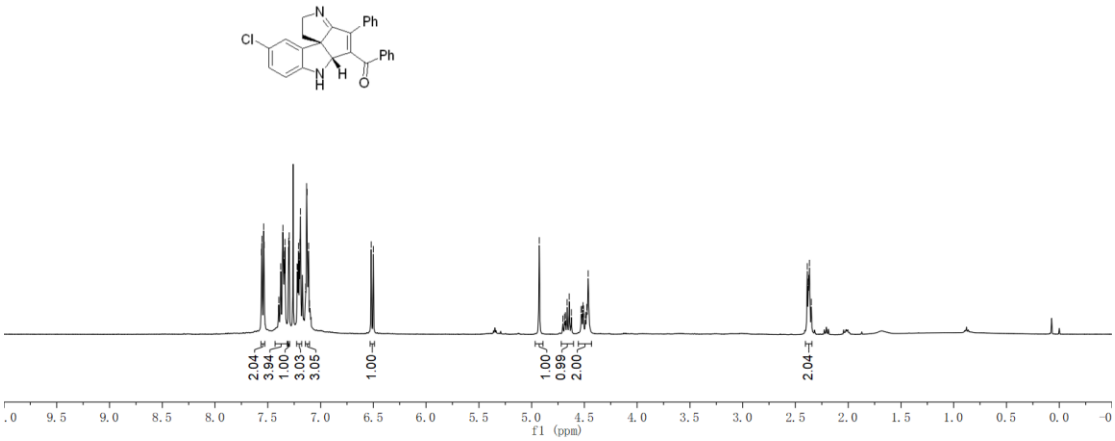
Compound **3p**

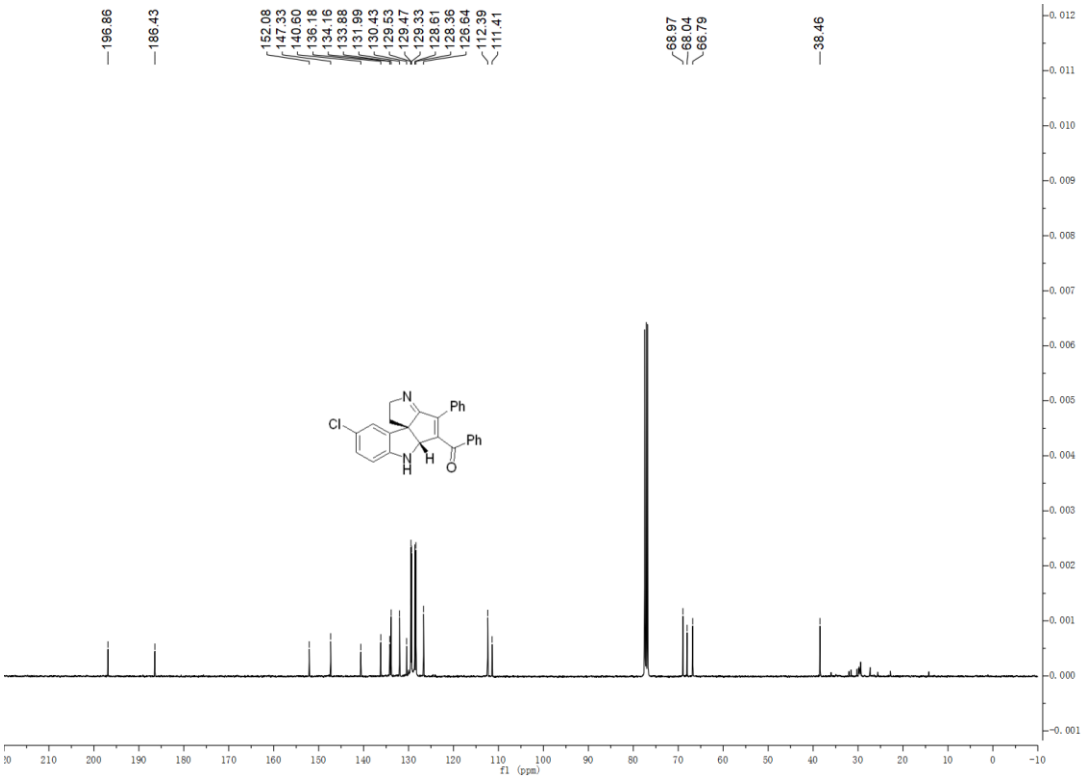

7.13
7.11
7.09
7.04
7.02
6.78
6.76
6.74
6.72
4.56
4.54
4.53
4.52
4.50
4.49
4.48
4.42
4.40
4.38
4.36
4.32
4.20
2.79
2.77
2.74
2.73
2.70
2.68
2.67
2.65
2.64
2.62
2.19
2.18
2.17
2.17
2.15
1.20
1.16
1.14
1.12
1.02
1.02
2.02
1.06
1.07
1.00
0.87
2.09
2.09
9.10
3.09
f1 (ppm)


207.80
186.94
152.77
147.78
144.32
132.89
128.87
123.08
120.44
111.88
68.56
67.85
66.36
38.53
37.79
33.69
29.31
7.35
f1 (ppm)

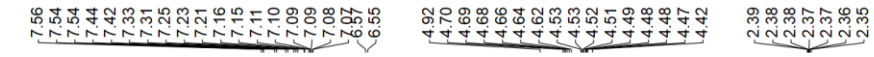
Compound **4a**

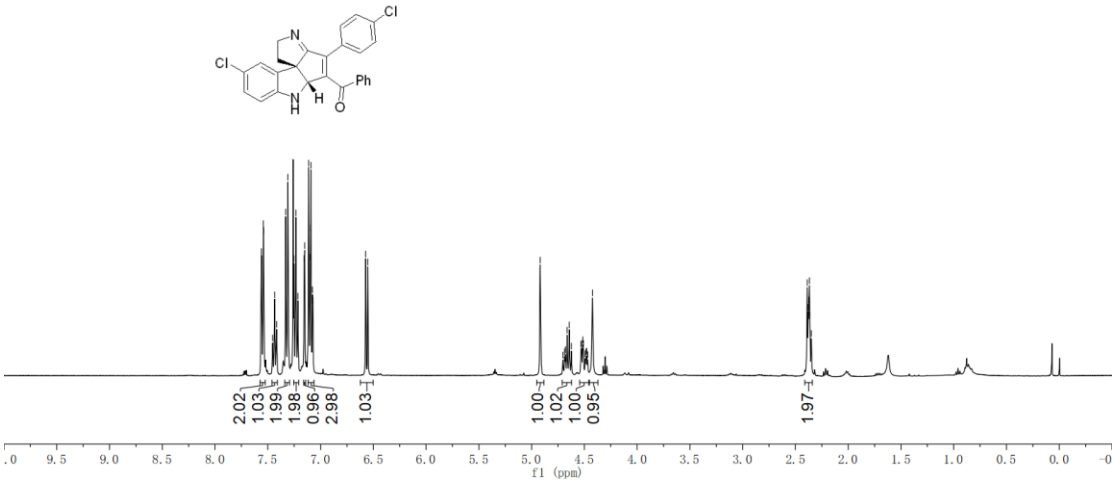

7.56
7.56
7.54
7.53
7.40
7.38
7.36
7.36
7.34
7.34
7.30
7.30
7.22
7.22
7.21
7.20
7.20
7.19
7.14
7.13
7.11
6.52
6.50
4.93
4.70
4.69
4.68
4.66
4.64
4.62
4.53
4.52
4.51
4.51
4.49
4.48
4.46
2.39
2.38
2.37
2.37
2.35

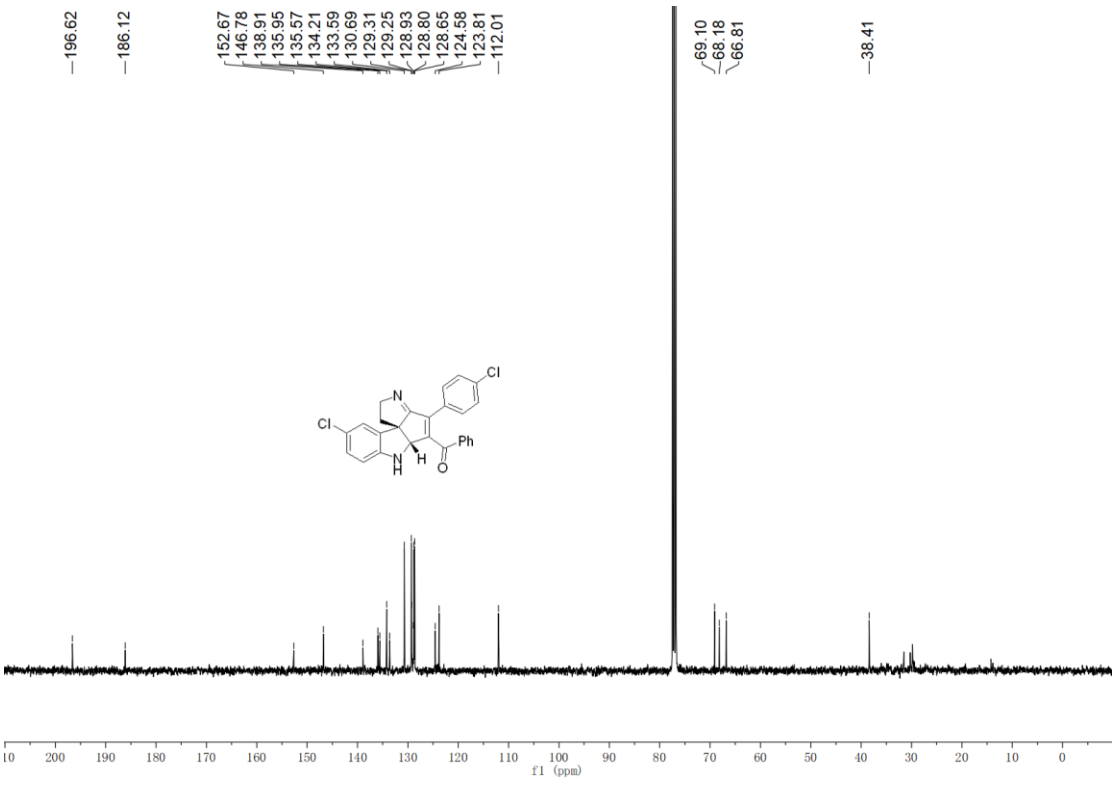

Cl
N
Ph
Ph
H
N
O
2.04
3.94
1.00
3.03
3.05
1.00
1.00
0.98
2.00
2.04
f1 (ppm)


196.86
186.43
152.08
147.33
140.60
136.18
134.16
133.88
131.99
130.43
129.53
129.47
129.33
128.61
128.36
126.64
112.39
111.41
68.97
68.04
66.79
38.46
Cl
N
Ph
Ph
H
N
O
f1 (ppm)

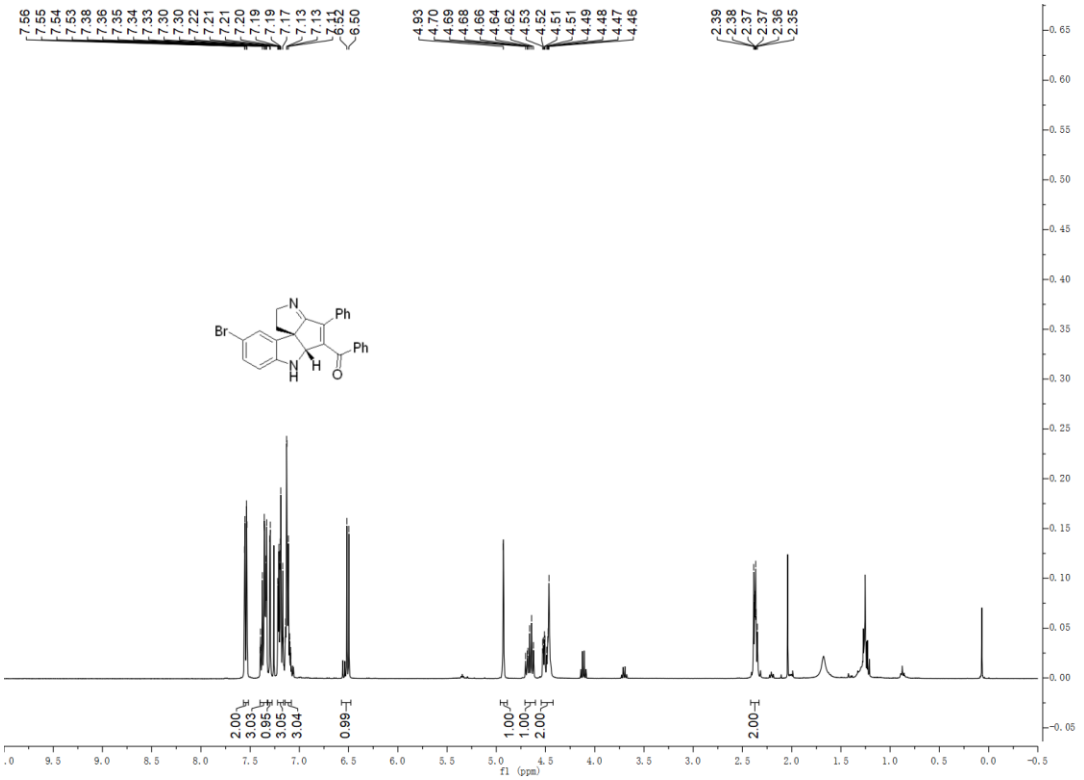
Compound **4b**

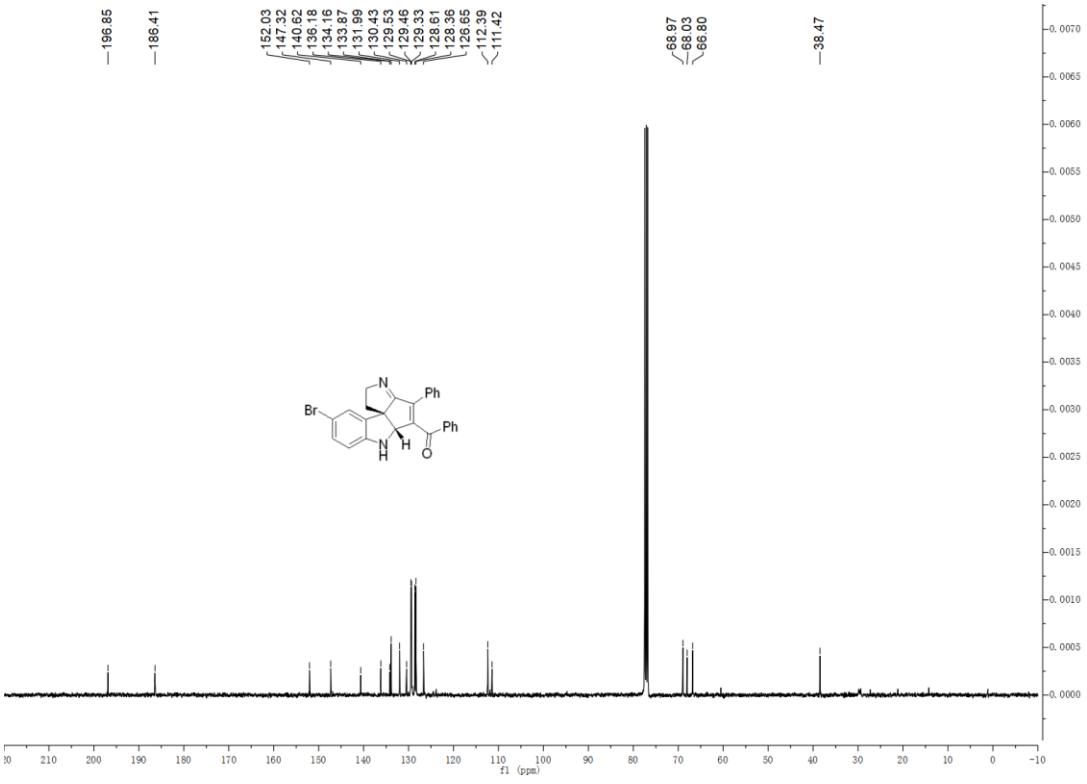

7.56
7.54
7.54
7.44
7.42
7.33
7.31
7.25
7.23
7.21
7.16
7.15
7.11
7.10
7.09
7.09
7.08
7.07
6.57
6.55
4.92
4.70
4.69
4.68
4.66
4.64
4.62
4.53
4.53
4.52
4.51
4.49
4.48
4.48
4.47
4.42
2.39
2.38
2.38
2.37
2.37
2.36
2.35


Cl
N
Cl
Ph
H
O
2.02
1.03
1.99
1.98
0.96
2.98
1.03
1.00
1.02
1.00
0.95
1.97
f1 (ppm)

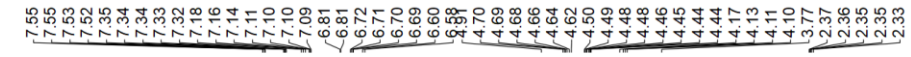

196.62
186.12
152.67
146.78
138.91
135.95
135.57
134.21
133.59
130.69
129.31
129.25
128.93
128.80
128.65
124.58
123.81
112.01
69.10
68.18
66.81
38.41
Cl
N
Cl
Ph
H
O
f1 (ppm)

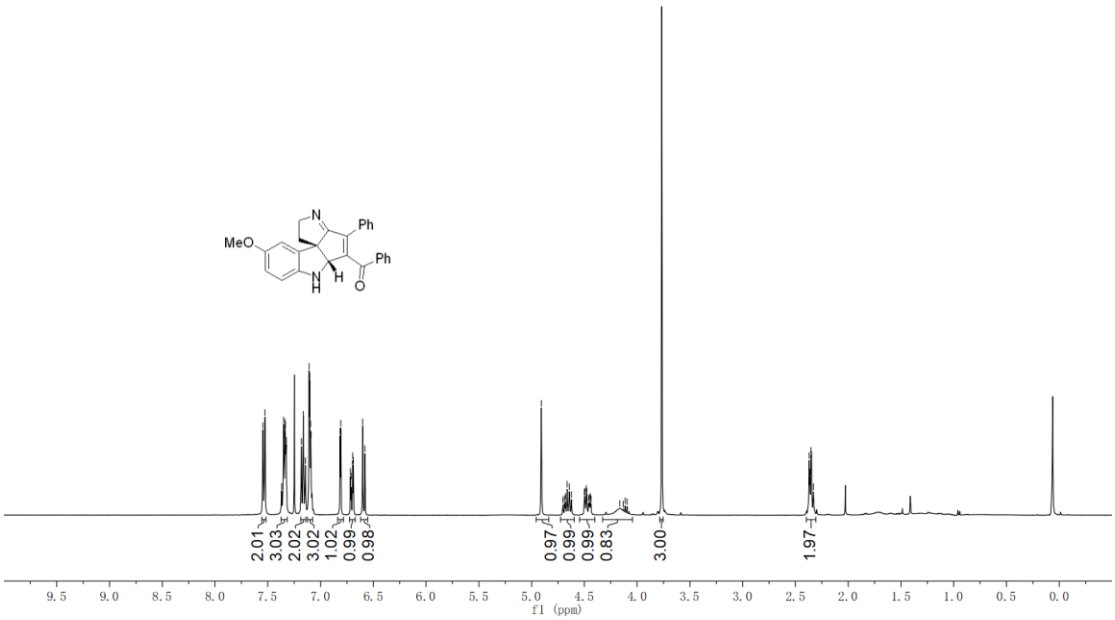
Compound **4c**

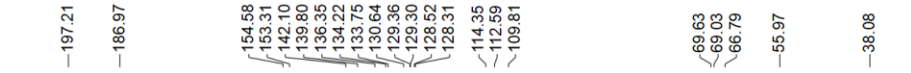

7.56
7.55
7.54
7.53
7.38
7.36
7.35
7.34
7.33
7.30
7.30
7.22
7.21
7.21
7.20
7.19
7.17
7.13
7.11
6.52
6.50
4.93
4.70
4.69
4.68
4.66
4.64
4.62
4.53
4.52
4.51
4.51
4.49
4.48
4.47
4.46
2.39
2.38
2.37
2.37
2.36
2.35
Br
N
Ph
Ph
H
O
2.00
3.03
0.95
3.05
3.04
0.99
1.00
1.00
2.00
2.00
f1 (ppm)

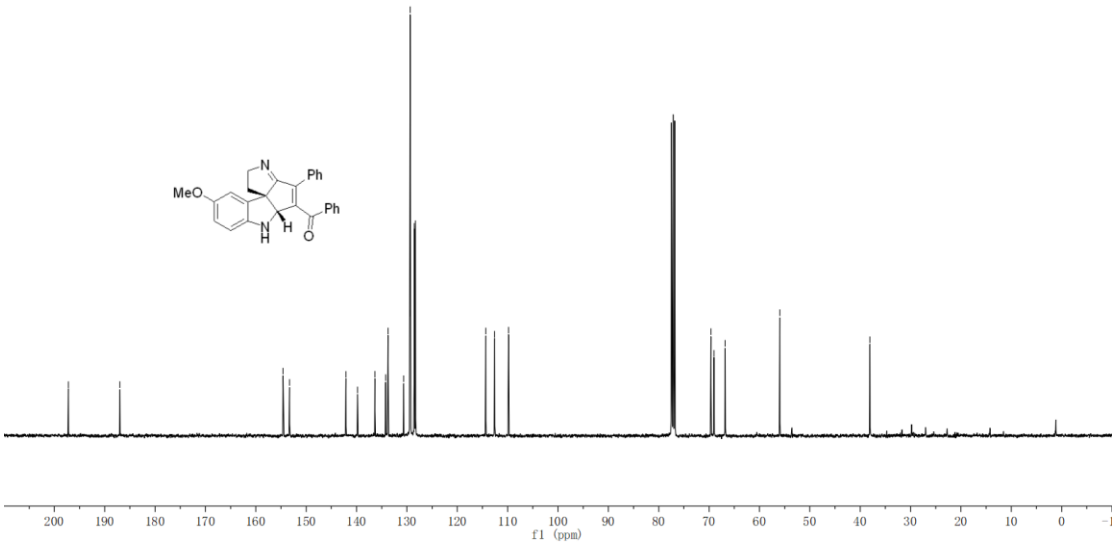

196.85
186.41
152.03
147.32
140.62
136.18
134.16
133.87
131.99
130.43
129.53
129.46
129.33
128.61
128.36
128.65
112.39
111.42
68.97
68.03
66.80
38.47
Br
N
Ph
Ph
H
O
f1 (ppm)

Compound **4d**

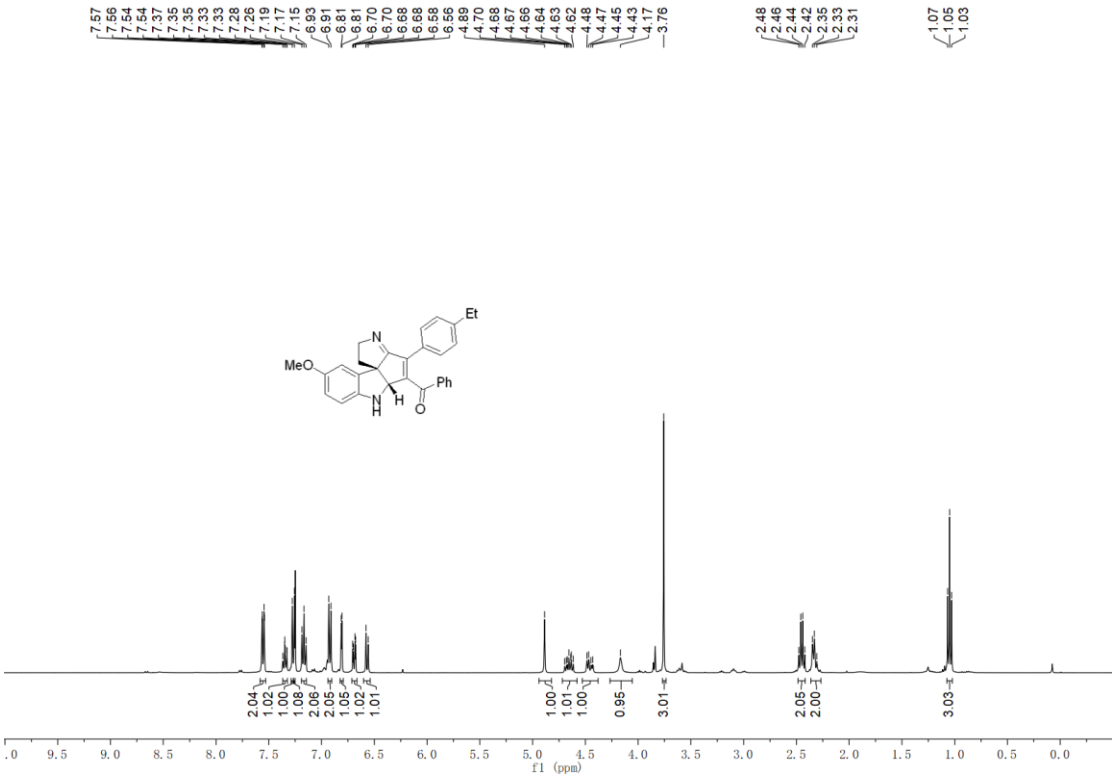

7.55
7.55
7.53
7.52
7.35
7.34
7.34
7.33
7.32
7.18
7.16
7.14
7.11
7.10
7.10
7.09
6.81
6.81
6.72
6.71
6.70
6.69
6.60
6.58
4.70
4.69
4.68
4.66
4.64
4.62
4.50
4.49
4.48
4.48
4.46
4.45
4.44
4.44
4.17
4.13
4.11
4.10
3.77
2.37
2.36
2.35
2.35
2.33

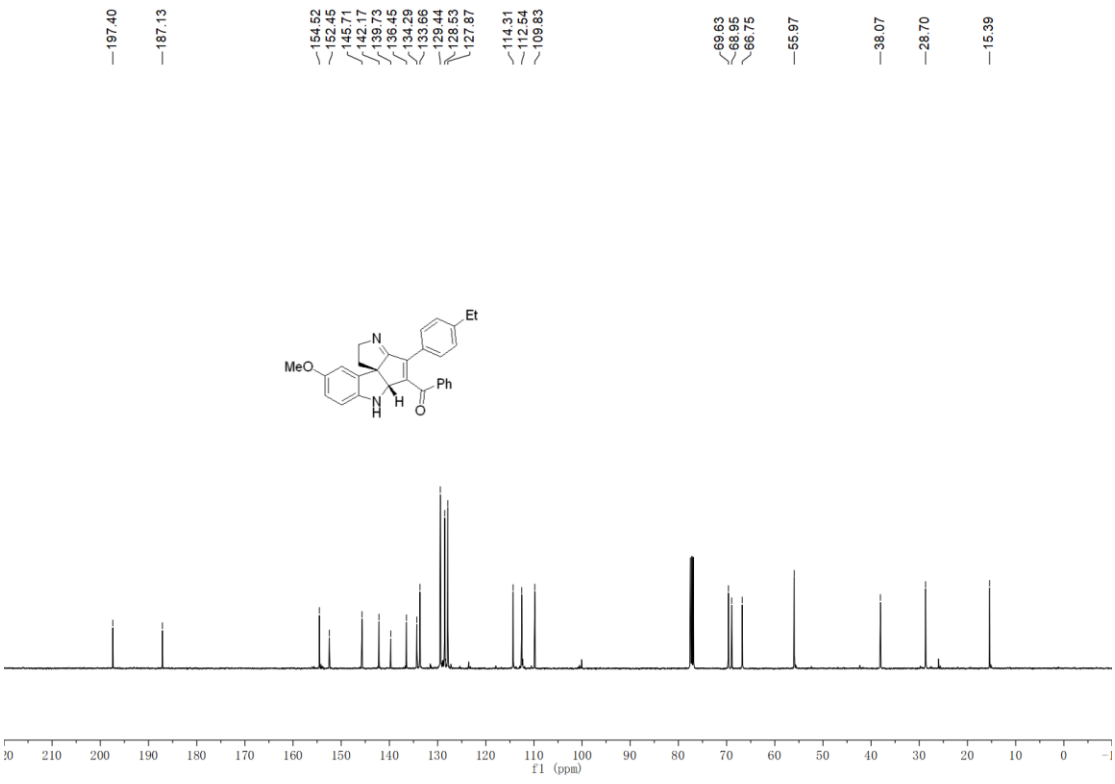

MeO
N
Ph
Ph
N
H
O
2.01
3.03
2.02
3.02
1.02
0.99
0.98
0.97
0.99
0.99
0.83
3.00
1.97
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


197.21
186.97
154.58
153.31
142.10
139.80
136.35
134.22
133.75
130.64
129.36
129.30
128.52
128.31
114.35
112.59
109.81
69.63
69.03
66.79
55.97
38.08

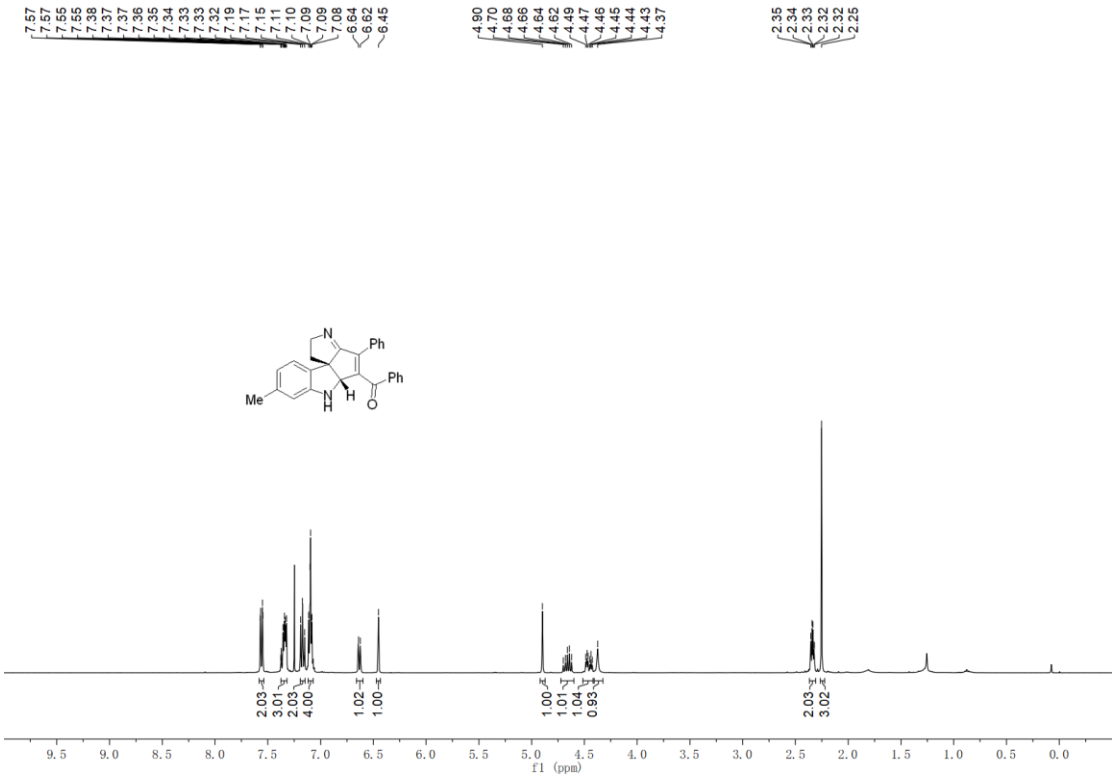

MeO
N
Ph
Ph
N
H
O
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

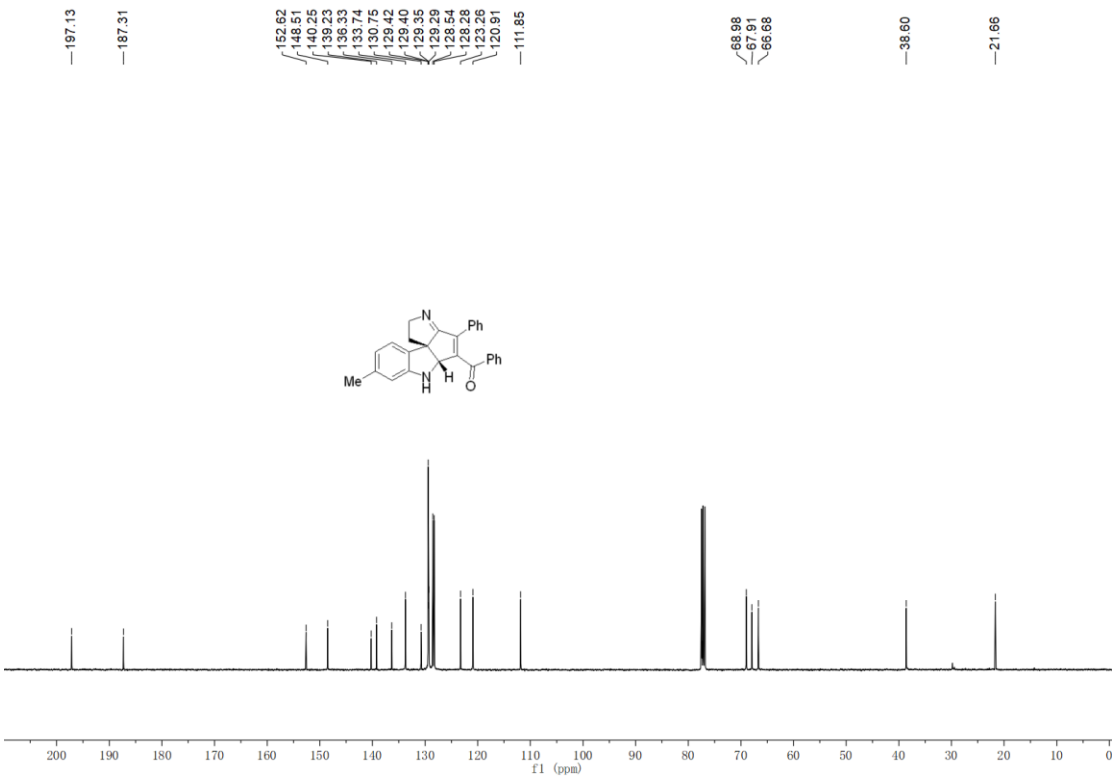
Compound **4e**


7.57
7.56
7.54
7.54
7.37
7.35
7.35
7.33
7.33
7.28
7.26
7.19
7.17
7.15
6.93
6.91
6.81
6.81
6.70
6.70
6.68
6.68
6.58
6.56
4.89
4.70
4.68
4.67
4.66
4.64
4.63
4.62
4.48
4.47
4.45
4.43
4.17
3.76
2.48
2.46
2.44
2.42
2.35
2.33
2.31
1.07
1.05
1.03
Et
N
MeO
Ph
N
H
H
O
2.04
1.02
1.00
1.08
2.06
2.05
1.05
1.02
1.01
1.00
1.01
1.00
0.95
3.01
2.05
2.00
3.03
f1 (ppm)

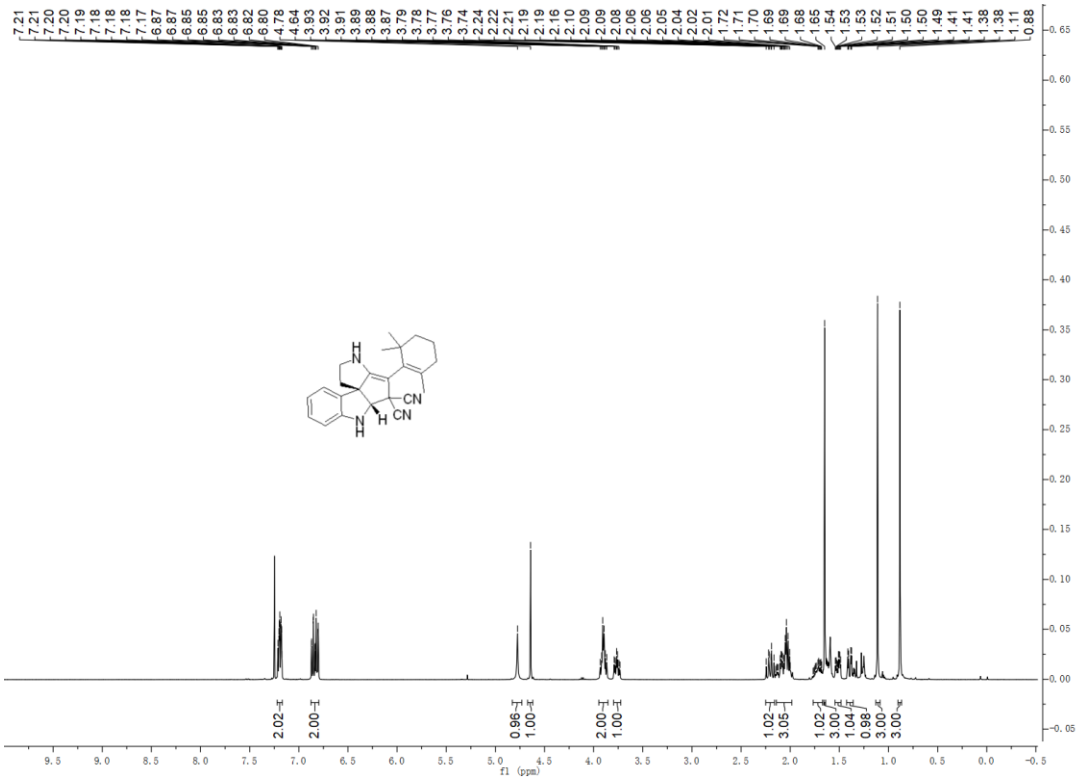

197.40
187.13
154.52
152.45
145.71
142.17
139.73
136.45
134.29
133.66
129.44
128.53
127.87
114.31
112.54
109.83
69.63
68.95
66.75
55.97
38.07
28.70
15.39
Et
N
MeO
Ph
N
H
H
O
f1 (ppm)

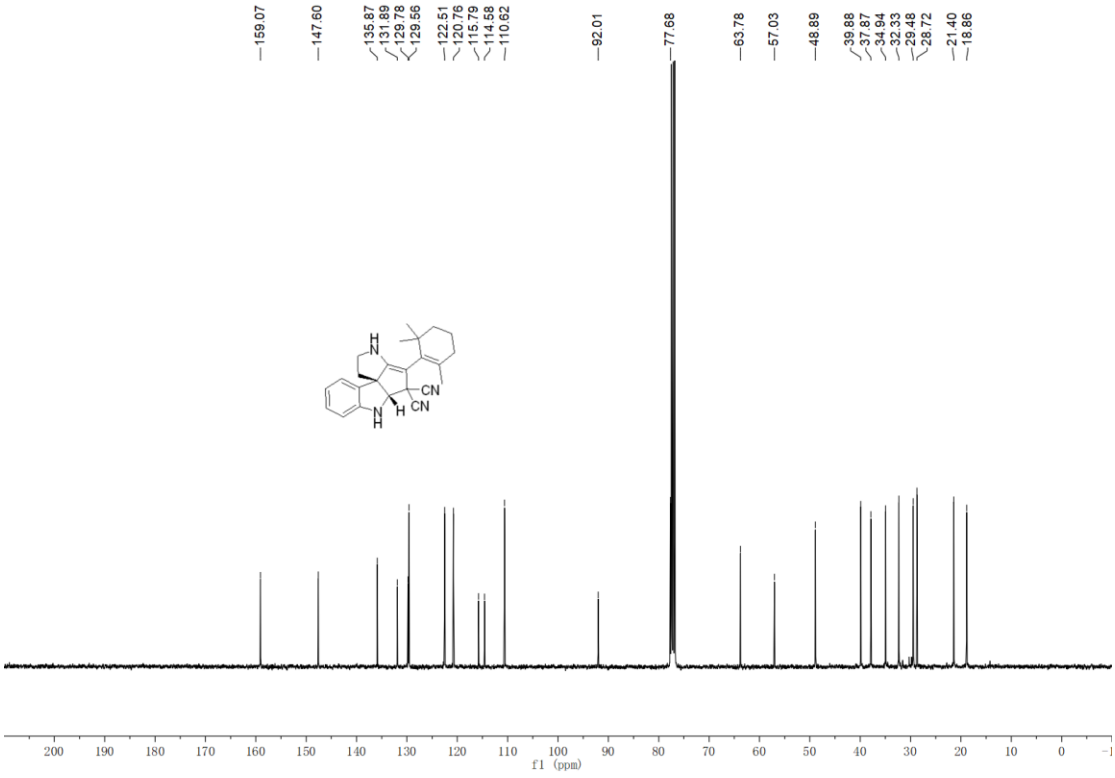
Compound **4f**


7.57
7.57
7.55
7.55
7.38
7.37
7.37
7.36
7.35
7.34
7.33
7.33
7.32
7.19
7.17
7.15
7.11
7.10
7.09
7.09
7.08
6.64
6.62
6.45
4.90
4.70
4.68
4.66
4.64
4.62
4.49
4.47
4.46
4.45
4.44
4.43
4.37
2.35
2.34
2.33
2.32
2.32
2.25
N
Ph
Ph
Me
N
H
O
2.03
3.01
2.03
4.00
1.02
1.00
1.00
1.01
1.04
0.93
2.03
3.02
f1 (ppm)


197.13
187.31
152.62
148.51
140.25
139.23
136.33
133.74
130.75
129.42
129.40
129.35
129.29
128.54
128.28
123.26
120.91
111.85
68.98
67.91
66.68
38.60
21.66
N
Ph
Ph
Me
N
H
O
f1 (ppm)

Compound **8**


7.21
7.21
7.20
7.20
7.19
7.18
7.18
7.18
7.17
6.87
6.87
6.85
6.85
6.83
6.83
6.82
6.80
4.78
4.64
3.93
3.92
3.91
3.89
3.88
3.87
3.79
3.78
3.77
3.76
3.74
2.24
2.22
2.21
2.19
2.19
2.16
2.10
2.09
2.09
2.08
2.06
2.06
2.05
2.04
2.02
2.01
1.72
1.71
1.70
1.69
1.69
1.68
1.65
1.54
1.53
1.53
1.52
1.51
1.50
1.50
1.49
1.41
1.41
1.38
1.38
1.11
0.88
CN
CN
H
N
H
2.02
2.00
0.96
1.00
2.00
1.00
1.02
3.05
1.02
3.00
1.04
0.98
3.00
3.00
f1 (ppm)


159.07
147.60
135.87
131.89
129.78
129.56
122.51
120.76
115.79
114.58
110.62
92.01
77.68
63.78
57.03
48.89
39.88
37.87
34.94
32.33
29.48
28.72
21.40
18.86
CN
CN
H
N
H
f1 (ppm)