Supporting Information

$BF_3 \cdot Et_2O$-Promoted Dearomative Spirocyclization of Ynones for the Synthesis of Spirolactams and Spirolactones

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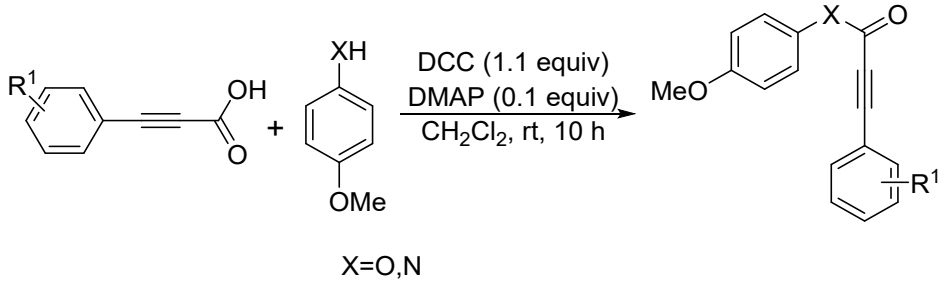
1.General Experimental Information.

All reactions were carried out under air atmosphere, unless otherwise stated. All chemicals were purchased from commercial companies or synthesized according to the literature method. ^{1}H NMR spectra were recorded on 400 MHz or 600 MHz in $CDCl_3$ or DMSO-d_6. $^{13}C\{^{1}H\}$ NMR spectra were recorded on 100 MHz or 150 MHz in $CDCl_3$ or DMSO-d_6. The data is reported as (s = singlet, d = doublet, t = triplet, q = quartet, dd = doublet of doublet, m = multiplet or unresolved, coupling constant(s) in Hz, integration, assignment).

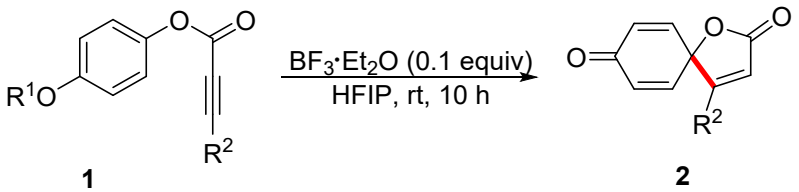
2.General procedure for synthesis of 1 and 3.



X=O,N

Following the literature procedure[1], we synthesized the substrates for our reaction, as detailed below: a mixture of 4-methoxyphenol (1.365 g, 11 mmol) and 3-phenylpropiolic acid (1.5 g, 10 mmol) in anhydrous dichloromethane (20 mL) was prepared in a round-bottom flask. The solution was cooled to 0°C in an ice bath, followed by sequential addition of dicyclohexylcarbodiimide (DCC, 2.3 g, 11 mmol) and 4-dimethylaminopyridine (DMAP, 122.2 mg, 1 mmol). The reaction was allowed to warm to room temperature gradually and stirred for 10 hours (monitored by TLC). Upon completion, the reaction mixture was quenched with distilled water (20 mL) and extracted with ethyl acetate (3 × 15 mL). The combined organic extracts were dried over anhydrous sodium sulfate, filtered, and concentrated under reduced pressure. The crude product was purified by flash column chromatography to afford the desired product. Among these, 1a[1], 1b[2], 1d[3], 1e[3], 1g[3], 1h[4], 1i[2], 1j[2], 1k[5], 1l[6], 3a[7], 3b[7], 3c[7], 3e[7], 3f[7], 3g[7], 3j[8], 3k[9], 3l[9], 3m[7], 3n[7], 3o[7], 3p[10], 3q[7], 3s[10], 3t[8] are known compounds synthesized via this method, while 1c, 1f, 3b, 3h, 3i, 3r are unknown compounds obtained by the same protocol. NMR and HRMS data for these unknown compounds have been supplemented.

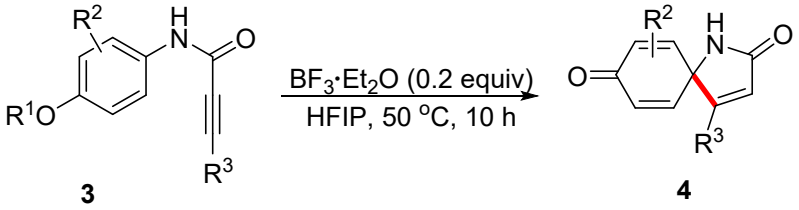
3.General procedure for synthesis of 2.



1 **2**

To a glass tube with substrate **1** (0.1 mmol, 1.0 equiv) and HFIP (1.5 mL) was added $BF_3 \cdot Et_2O$ (0.01 mmol, 0.1 equiv) under air atmosphere, and it was stirred 10 h at room temperature. The reaction mixture was purified by column chromatography on silica gel with eluent to afford the pure desired product.

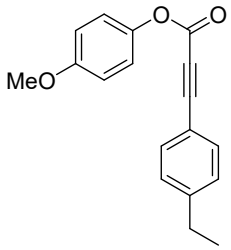
4.General procedure for synthesis of 4.



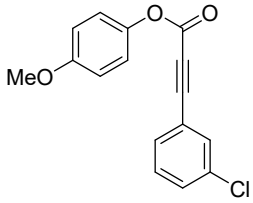
3 **4**

To a glass tube with substrate **3** (0.1 mmol, 1.0 equiv) and HFIP (1.5 mL) was added $BF_3 \cdot Et_2O$ (0.02 mmol, 0.2 equiv) under air atmosphere, and it was stirred 10 h at 50 °C. The reaction mixture was purified by column chromatography on silica gel with eluent to afford the pure desired product.

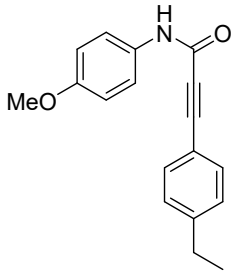
5. Characterization data



4-methoxyphenyl 3-(4-ethylphenyl) propiolate (1c): **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.55 (d, *J* = 8.0 Hz, 2H), 7.23 (d, *J* = 8.0 Hz, 2H), 7.11 (d, *J* = 8.0 Hz, 2H), 6.92 (d, *J* = 8.0 Hz, 2H), 3.81 (s, 3H), 2.69 (q, *J* = 8.0 Hz, 2H), 1.25 (t, *J* = 8.0 Hz, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 157.7, 153.1, 148.1, 143.8, 133.4, 128.4, 122.4, 116.5, 114.7, 89.4, 80.1, 55.7, 29.1, 15.2. **HRMS (ESI)** m/z: calcd. for $C_{18}H_{16}NaO_3^+$ $[M + Na]^+$ 303.0992, found 303.0997.

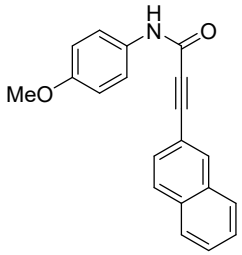


4-methoxyphenyl 3-(3-chlorophenyl) propiolate (1f): **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.61 (s, 1H), 7.52 – 7.45 (m, 2H), 7.34 (t, *J* = 8.0 Hz, 1H), 7.11 (d, *J* = 8.0 Hz, 2H), 6.92 (d, *J* = 8.0 Hz, 2H)，3.81 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 157.9, 152.6, 143.6, 134.8, 132.9, 131.4, 131.3, 130.1, 122.3, 121.2, 114.7, 86.7, 81.1, 55.8. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{11}ClNaO_3^+$ $[M + Na]^+$ 309.0289, found 303.0279.

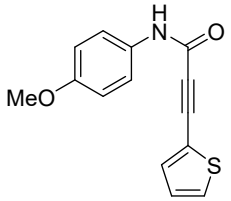


3-(4-ethylphenyl)-*N*-(4-methoxyphenyl) propiolamide (3d): **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.77 (s, 1H), 7.50 – 7.46 (m, 4H), 7.18 (d, *J* = 8.0 Hz, 2H), 6.86 (d, *J* = 8.0 Hz, 2H), 3.79 (s,

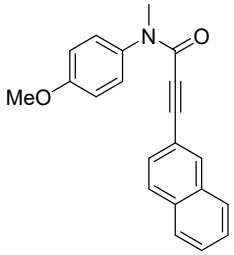
3H), 2.66 (q, J = 8.0 Hz, 2H), 1.23 (t, J = 8.0 Hz, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 156.9, 151.3, 147.1, 132.8, 130.7, 128.3, 121.9, 117.3, 114.3, 86.2, 83.2, 55.6, 29.1, 15.3. **HRMS (ESI)** m/z: calcd. for $C_{18}H_{17}NNaO_2^+$ [M + Na]$^+$ 302.1152, found 302.1140.



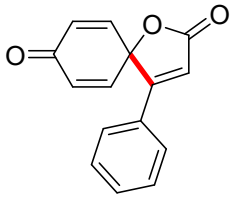
***N*-(4-methoxyphenyl)-3-(naphthalen-2-yl) propiolamide (3h)**: **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 8.08 (s, 1H), 7.98 (s, 1H), 7.82 – 7.77 (m, 3H), 7.54 – 7.51 (m, 5H), 6.86 (d, J = 8.0 Hz, 2H), 3.77 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 156.9, 151.2, 133.7, 132.8, 130.7, 128.5, 128.3, 128.2, 128.0, 127.8, 127.0, 121.9, 117.4, 114.4, 86.1, 83.9, 55.6, 34.0. **HRMS (ESI)** m/z: calcd. for $C_{20}H_{15}NNaO_2^+$ [M + Na]$^+$ 324.0995, found 324.0984.



***N*-(4-methoxyphenyl)-3-(thiophen-2-yl) propiolamide (3i)**: **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.74 (s, 1H), 7.49 – 7.41 (m, 4H), 7.04 – 7.02 (m, 1H), 6.87 (d, J = 8.0 Hz, 2H), 3.79 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 156.9, 150.8, 135.6, 130.4, 130.3, 127.5, 121.9, 119.9, 114.3, 87.5, 79.6, 55.5. **HRMS (ESI)** m/z: calcd. for $C_{14}H_{11}NNaO_2S^+$ [M + Na]$^+$ 280.0403, found 280.0405.

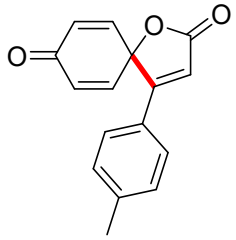


***N*-(4-methoxyphenyl)-*N*-methyl-3-(naphthalen-2-yl) propiolamide (3r)**: **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.77 – 7.68 (m, 4H), 7.51 – 7.45 (m, 2H), 7.30 (d, J = 8.0 Hz, 2H), 7.16 (d, J = 8.0 Hz, 1H), 6.98 (d, J = 8.0 Hz, 2H), 3.87 (s, 3H), 3.37 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ: 159.2, 154.7, 136.2, 133.5, 133.4, 132.6, 128.7, 128.2, 128.1, 128.0, 127.8, 127.6, 126.8, 117.8, 114.4, 91.4, 83.0, 55.7, 36.7. HRMS (ESI) m/z: calcd. for $C_{21}H_{17}NNaO_2^+$ [M + Na]$^+$ 338.1152, found 338.1142.

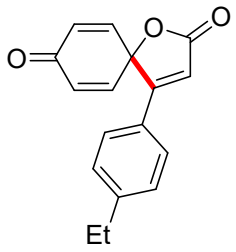


4-phenyl-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2a)[11]: white solid, (23 mg, 97% yield),

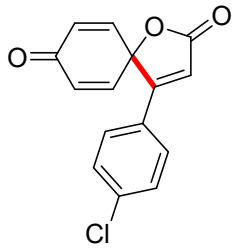
mp: 115 – 117 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.51 – 7.46 (m, 3H), 7.39 (t, *J* = 8.0 Hz, 2H), 6.71 (d, *J* = 8.0 Hz, 2H), 6.57 (s, 1H), 6.50 (d, *J* = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.2, 170.6, 165.4, 143.5, 132.2, 131.9, 129.4, 129.0, 127.3, 116.9, 81.5.



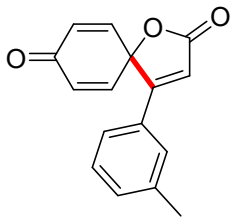
4-(*p*-tolyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2b): white solid, (20 mg, 79% yield), mp: 105 – 107 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.40 (d, *J* = 8.0 Hz, 2H), 7.19 (d, *J* = 8.0 Hz, 2H), 6.70 (d, *J* = 8.0 Hz, 2H), 6.52 – 6.48 (m, 3H), 2.37 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.2, 170.8, 165.3, 143.7, 143.1, 131.8, 130.1, 127.2, 126.1, 115.7, 81.5, 21.7. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{12}NaO_3^+$ [*M* + Na]$^+$ 275.0679, found 275.0680.



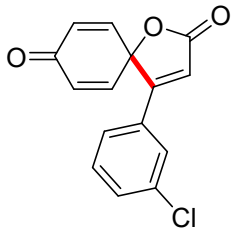
4-(4-ethylphenyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2c): yellow solid, (22 mg, 83% yield), mp: 100 – 102 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.43 (d, *J* = 8.0 Hz, 2H), 7.21 (d, *J* = 8.0 Hz, 2H), 6.71 (d, *J* = 12.0 Hz, 2H), 6.53 – 6.48 (m, 3H), 2.66 (q, *J* = 8.0 Hz, 2H), 1.22 (d, *J* = 8.0 Hz, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.3, 170.9, 165.3, 149.2, 143.7, 131.8, 129.0, 127.4, 126.3, 115.7, 81.3, 28.9, 15.2. **HRMS (ESI)** m/z: calcd. for $C_{17}H_{14}NaO_3^+$ [*M* + Na]$^+$ 289.0836, found 289.0834.



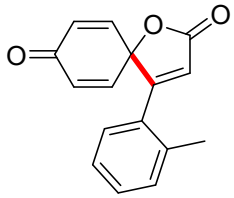
4-(4-chlorophenyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2d): white solid, (24 mg, 88% yield), mp: 117 – 119 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.44 (d, *J* = 8.0 Hz, 2H), 7.37 (d, *J* = 8.0 Hz, 2H), 6.69 (d, *J* = 8.0 Hz, 2H), 6.56 (s, 1H), 6.50 (d, *J* = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 183.9, 170.2, 164.0, 143.2, 138.5, 132.1, 129.8, 128.5, 127.3, 117.3, 81.3. **HRMS (ESI)** m/z: calcd. for $C_{15}H_9ClNaO_3^+$ [*M* + Na]$^+$ 295.0133, found 295.0131.



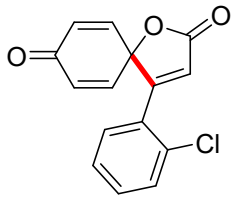
4-(*m*-tolyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2e): white solid, (21 mg, 83% yield), mp: 128 – 130 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.31 – 7.26 (m, 4H), 6.70 (d, *J* = 8.0 Hz, 2H), 6.54 (s, 1H), 6.49 (d, *J* = 8.0 Hz, 2H), 2.33 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.2, 170.7, 165.6, 143.6, 139.3, 133.0, 131.8, 129.3, 128.9, 128.0, 124.2, 116.7, 81.5, 21.5. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{12}NaO_3^+$ [*M* + Na]$^+$ 275.0679, found 275.0679.



4-(3-chlorophenyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2f): yellow solid, (23 mg, 85% yield), mp: 106 – 108 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.48 – 7.44 (m, 2H), 7.37 – 7.31 (m, 2H), 6.69 (d, *J* = 8.0 Hz, 2H), 6.57 (s, 1H), 6.51 (d, *J* = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 183.8, 170.1, 163.9, 142.9, 135.6, 132.2, 132.0, 130.7, 130.7, 127.5, 125.1, 118.3, 81.4. **HRMS (ESI)** m/z: calcd. for $C_{15}H_9ClNaO_3^+$ [*M* + Na]$^+$ 295.0133, found 295.0131.

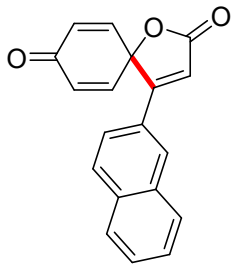


4-(*o*-tolyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2g): yellow solid, (21 mg, 83% yield), mp: 109 – 111 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.24 – 7.21 (m, 2H), 7.11 (t, *J* = 8.0 Hz, 1H), 7.03 (d, *J* = 8.0 Hz, 1H), 6.69 (d, *J* = 8.0 Hz, 2H), 6.27 (s, 1H), 2.27 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 183.7, 170.9, 166.8, 142.4, 135.8, 131.9, 131.3, 130.4, 128.7, 127.4, 125.8, 121.9, 84.7, 20.9. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{12}NaO_3^+$ [*M* + Na]$^+$ 275.0679, found 275.0678.

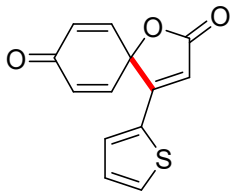


4-(2-chlorophenyl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2h): yellow solid, (24 mg, 88% yield), mp: 89 – 91 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.48 (d, *J* = 8.0 Hz, 2H), 7.37 (t, *J* = 8.0 Hz, 1H), 7.28 – 7.24 (m, 1H), 7.14 (d, *J* = 8.0 Hz, 2H), 6.48 (s, 1H), 6.37 (d, *J* = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 183.7, 170.5, 163.8,

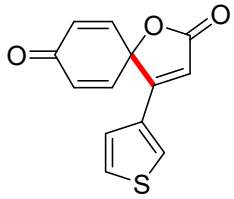
142.2, 132.7, 131.6, 131.9, 131.6, 130.6, 128.9, 128.2, 126.7, 123.6, 84.4. **HRMS (ESI)** m/z: calcd. for $C_{15}H_9ClNaO_3^+$ $[M + Na]^+$ 295.0133, found 295.0133.



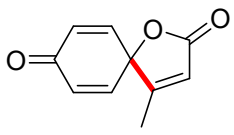
4-(naphthalen-2-yl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2i): yellow solid, (23 mg, 80% yield), mp: 143 – 145 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.98 (s, 1H), 7.85 (t, J = 8.0 Hz, 2H), 7.76 (d, J = 8.0 Hz, 1H), 7.59 – 7.53 (m, 3H), 6.78 (d, J = 8.0 Hz, 2H), 6.69 (s, 1H), 6.54 (d, J = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.7, 170.7, 165.0, 143.8, 134.6, 132.7, 131.9, 129.4, 129.2, 128.7, 127.9, 127.8, 127.5, 126.1, 123.7, 116.9, 81.4. **HRMS (ESI)** m/z: calcd. for $C_{19}H_{12}NaO_3^+$ $[M + Na]^+$ 311.0679, found 311.0678.



4-(thiophen-2-yl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2j): yellow solid, (19 mg, 78% yield), mp: 120 – 122 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.55 (d, J = 6.0 Hz, 1H), 7.32 (d, J = 6.0 Hz, 1H), 7.08 – 7.07 (m, 1H), 6.70 (d, J = 12.0 Hz, 2H), 6.51 (d, J = 12.0 Hz, 2H), 6.41 (s, 1H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.1, 170.5, 158.6, 143.5, 132.1, 131.6, 131.2, 130.2, 128.8, 113.9, 81.0. **HRMS (ESI)** m/z: calcd. for $C_{13}H_8NaO_3S^+$ $[M + Na]^+$ 267.0087, found 267.0086.

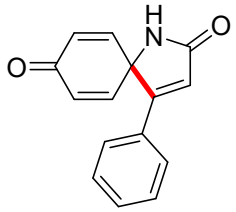


4-(thiophen-3-yl)-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2k): yellow solid, (18 mg, 74% yield), mp: 130 – 132 °C, eluent PE/EtOAc (3:1, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.54 (s, 1H), 7.40 – 7.39 (m, 1H), 7.25 (s, 1H), 6.79 (d, J = 12.0 Hz, 2H), 6.50 (d, J = 12.0 Hz, 2H), 6.41 (s, 1H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.1, 170.9, 159.6, 143.9, 131.8, 130.2, 127.9, 127.7, 126.5, 115.2, 81.0. **HRMS (ESI) m/z**: calcd. for $C_{13}H_8NaO_3S^+$ $[M + Na]^+$ 267.0087, found 267.0085.

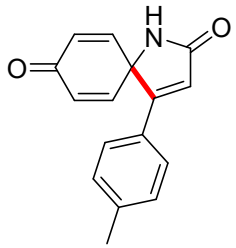


4-**methyl-1-oxaspiro[4.5]deca-3,6,9-triene-2,8-dione (2l)**[12]: yellow oil, (16 mg, 91% yield), eluent PE/EtOAc (1:1, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 6.52 (d, J = 12.0 Hz, 2H), 6.44 (d,

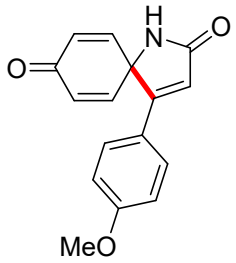
J = 12.0 Hz, 2H), 6.08 (s, 1H), 1.91 (s, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.0, 171.3, 166.4, 143.2, 131.9, 119.2, 83.9, 12.5.



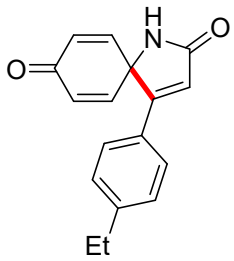
4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4a)[13]: white solid, (23 mg, 97% yield), mp: 215 – 217 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.48 (d, J = 8.0 Hz, 2H), 7.42 – 7.35 (m, 3H), 7.20 (s, 1H), 6.71 (d, J = 8.0 Hz, 2H), 6.57 (s, 1H), 6.46 (d, J = 12.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.5, 172.6, 159.1, 146.3, 131.2, 131.0, 130.8, 129.1, 126.9, 123.8, 63.3.



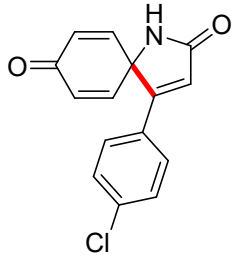
4-(*p*-tolyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4b): yellow solid, (24 mg, 96% yield), mp: 216 – 218 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, DMSO) δ: 8.64 (s, 1H), 7.45 (d, J = 8.0 Hz, 2H), 7.19 (d, J = 8.0 Hz, 2H), 6.87 (d, J = 12.0 Hz, 2H), 6.78 (s, 1H) 6.38 (d, J = 12.0 Hz, 2H), 2.28 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, DMSO) δ: 184.6, 171.5, 157.0, 184.4, 140.1, 129.8, 129.4, 128.4, 126.6, 123.8, 62.4, 20.9. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{12}NO_2^-$ [M - H]$^-$ 250.0713, found 250.0713.



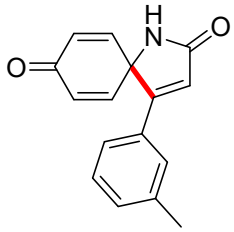
4-(4-methoxyphenyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4c): yellow solid, (21 mg, 78% yield), mp: 195 – 197 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, DMSO) δ: 8.57 (s, 1H), 7.53 (d, J = 8.0 Hz, 2H), 6.95 (d, J = 8.0 Hz, 2H), 6.87 (d, J = 8.0 Hz, 2H), 6.73 (s, 1H) 6.39 (d, J = 12.0 Hz, 2H), 3.75 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, DMSO) δ: 184.6, 171.7, 160.7, 156.6, 148.6, 129.7, 128.3, 123.5, 122.2, 114.3, 62.2, 55.3. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{13}NNaO_3^+$ [M + Na]$^+$ 290.0788, found 290.0787.



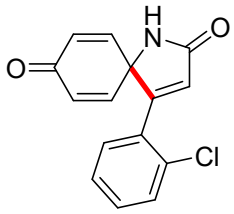
4-(4-ethylphenyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4d): yellow solid, (25 mg, 93% yield), mp: 217 – 219 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.62 (s, 1H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.16 (d, *J* = 8.0 Hz, 2H), 6.70 (d, *J* = 8.0 Hz, 2H), 6.53 (s, 1H), 6.44 (d, *J* = 8.0 Hz, 2H), 2.65 – 2.60 (m, 2H), 1.22 – 1.18 (m, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.6, 172.9, 159.0, 147.7, 146.5, 131.0, 128.6, 128.2, 126.9, 122.9, 63.2, 28.8, 15.2. **HRMS (ESI)** m/z: calcd. for $C_{17}H_{14}NO_2^-$ [*M* - H]$^-$ 264.1030, found 264.1029.



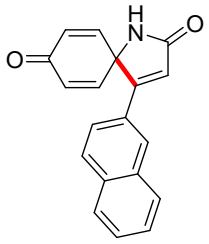
4-(4-chlorophenyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4e): white solid, (25 mg, 92% yield), mp: 240 – 242 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, DMSO) δ: 8.74 (s, 1H), 7.58 (d, *J* = 12.0 Hz, 2H), 7.48 (d, *J* = 8.0 Hz, 2H), 7.89 – 7.87 (m, 3H), 6.49 (d, *J* = 8.0 Hz, 2H). **^{13}C{^{1}H} NMR** (100 MHz, DMSO) δ: 184.5, 171.2, 155.7, 148.0, 134.9, 130.1, 130.0, 129.0, 128.5, 125.5, 62.4. **HRMS (ESI)** m/z: calcd. for $C_{15}H_9ClNO_2^-$ [*M* - H]$^-$ 270.0327, found 270.0326.



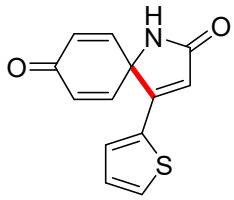
4-(*m*-tolyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4f): yellow solid, (24 mg, 96% yield), mp: 168 – 170 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.31 – 7.24 (m, 5H), 6.73 (d, *J* = 12.0 Hz, 2H), 6.57 (s, 1H), 6.47 (d, *J* = 12.0 Hz, 2H), 2.34, (s, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.5, 172.7, 159.3, 146.3, 138.9, 131.7, 131.1, 130.9, 129.0, 127.7, 123.9, 123.7, 63.4, 21.5. **HRMS (ESI)** m/z: calcd. for $C_{16}H_{12}NO_2^-$ [*M* - H]$^-$ 250.0873, found 250.0873.



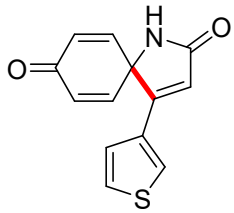
4-(2-chlorophenyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4g): white solid, (24 mg, 89% yield), mp: 158 – 160 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.60 (s, 1H), 7.42 (d, *J* = 6.0 Hz, 1H), 7.29 (t, *J* = 6.0 Hz, 1H), 7.21 – 7.18 (m, 1H), 7.10 (d, *J* = 6.0 Hz, 1H), 6.75 (d, *J* = 12.0 Hz, 2H), 6.40 (s, 1H), 6.30 (d, *J* = 12.0 Hz, 2H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.1, 172.7, 157.3, 144.8, 133.0, 121.6, 130.6, 130.4, 129.8, 129.7, 129.6, 126.4, 66.2. **HRMS (ESI)** m/z: calcd. for $C_{15}H_9ClNO_2^-$ [*M* - H]$^-$ 270.0327, found 270.0326.



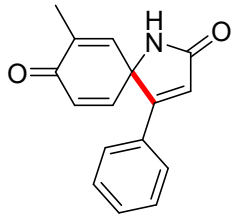
4-(naphthalen-2-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4h): yellow solid, (25 mg, 87% yield), mp: 192 – 194 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, DMSO) δ: 8.74 (s, 1H), 8.02 (s, 1H), 7.93 – 7.89 (m, 2H), 7.79 – 7.74 (m, 2H), 7.57 – 7.51 (m, 2H), 6.97 – 6.94 (m, 3H), 6.44 (d, *J* = 12.0 Hz, 2H). **^{13}C{^{1}H} NMR** (150 MHz, DMSO) δ: 184.5, 171.3, 156.7, 148.2, 133.2, 132.2, 129.9, 128.6, 128.4, 128.3, 127.5, 127.4, 126.9, 126.0, 125.2, 124.2, 62.5. **HRMS (ESI)** m/z: calcd. for $C_{19}H_{12}NO_2^-$ [*M* - H]$^-$ 286.0873, found 286.0871.



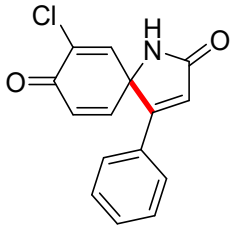
4-(thiophen-2-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4i): yellow solid, (23 mg, 95% yield), mp: 128 – 130 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.43 (d, *J* = 6.0 Hz, 1H), 7.24 (d, *J* = 6.0 Hz, 1H), 7.02 – 7.01 (m, 2H), 6.68 (d, *J* = 12.0 Hz, 2H), 6.48 (d, *J* = 6.0 Hz, 2H), 6.44 (s, 1H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.5, 172.9, 153.6, 146.7, 131.9, 131.2, 127.1, 126.4, 125.8, 122.2, 62.9. **HRMS (ESI)** m/z: calcd. for $C_{13}H_8NO_2S^-$ [*M* - H]$^-$ 242.0281, found 242.0281.



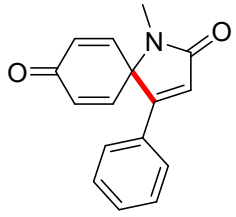
4-(thiophen-3-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4j): yellow solid, (23 mg, 95% yield), mp: 195 – 197 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.47 (s, 1H), 7.37 – 7.35 (m, 1H), 7.28 – 7.27 (m, 1H), 7.17 (s, 1H), 6.71 (d, *J* = 8.0 Hz, 2H), 6.50 – 6.46 (m, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.4, 172.6, 152.7, 146.1, 133.1, 131.6, 129.6, 128.4, 128.4, 121.2, 63.0. **HRMS (ESI)** m/z: calcd. for $C_{13}H_8NO_2S^-$ [*M* - H]$^-$ 242.0281, found 242.0279.



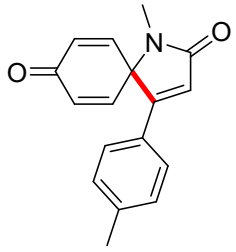
7-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4k): white solid, (21 mg, 84% yield), mp: 176 – 178 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.46 (d, *J* = 6.0 Hz, 2H), 7.39 (t, *J* = 6.0 Hz, 1H), 6.44 (d, *J* = 6.0 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 185.3, 172.5, 159.3, 146.1, 141.2, 138.2, 131.0, 130.8, 129.1, 126.9, 123.5, 63.7, 16.1 **HRMS (ESI)** m/z: calcd. for $C_{16}H_{13}NNaO_2^+$ [*M* + Na]$^+$ 274.0839, found 274.0839.



7-chloro-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4l): white solid, (24 mg, 89% yield), mp: 196 – 198 °C, eluent PE/EtOAc (1:2, v/v).**^{1}H NMR** (400 MHz, DMSO) δ: 8.69 (s, 1H), 7.55 – 7.53 (m, 2H), 7.42 – 7.41 (m, 3H), 7.35 (d, *J* = 6.0 Hz, 1H), 7.02 – 6.98 (m, 1H), 6.88 (s, 1H), 6.53 (d, *J* = 12.0 Hz, 1H). **^{13}C{^{1}H} NMR** (150 MHz, DMSO) δ: 177.6, 171.0, 156.2, 148.9, 144.2, 132.5, 130.8, 130.2, 128.9, 128.6, 126.6, 125.0, 64.3. **HRMS (ESI)** m/z: calcd. for $C_{15}H_{10}ClNNaO_2^+$ [*M* + Na]$^+$ 294.0293, found 294.0292.

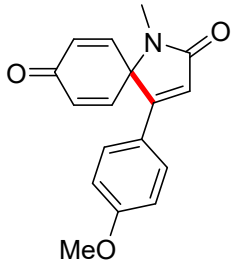


1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4m)[14]: white solid, (24 mg, 96% yield), mp: 125 – 127 °C, eluent PE/EtOAc (1:2, v/v).**^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.47 (d, *J* = 8.0 Hz, 2H), 7.38 – 7.31 (m, 3H), 6.65 (s, 1H), 6.59 – 6.54(m, 4H), 2.82 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.3, 170.1, 156.1, 146.0, 132.9, 130.8, 130.7, 129.1, 126.6, 124.1, 66.9, 25.1.

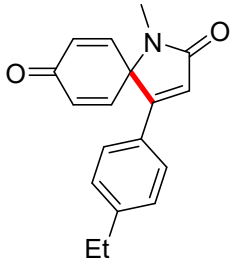


1-methyl-4-(p-tolyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4n): white solid, (25 mg, 94% yield), mp: 109 – 111 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.38 – 7.35 (m, 2H), 7.14 – 7.11 (m, 2H), 6.60 – 6.54(m, 5H), 2.81 (s, 3H), 2.33 (s, 3H). **^{13}C{^{1}H}**

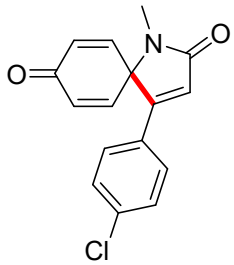
NMR (150 MHz, $CDCl_3$) δ: 184.3, 170.3, 156.1, 146.2, 141.2, 132.8, 129.8, 128.1, 126.6, 123.2, 67.0, 25.1, 21.4. **HRMS (ESI)** m/z: calcd. for $C_{17}H_{15}NNaO_2^+$ $[M + Na]^+$ 288.0995, found 288.0995.



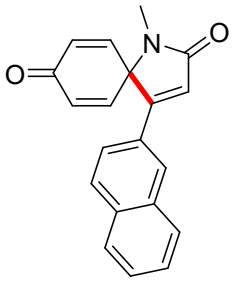
4-(4-methoxyphenyl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4o)[15]: white solid, (28 mg, 98% yield), mp: 105 – 107 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (400 MHz, $CDCl_3$) δ: 7.44 – 7.41 (m, 2H), 6.84 – 6.81 (m, 2H), 6.58 – 6.52 (m, 5H), 3.79 (s, 3H), 2.79 (s, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.3, 170.4, 161.6, 155.6, 146.4, 132.7, 128.2, 123.4, 121.8, 114.5, 66.8, 55.5, 25.1.



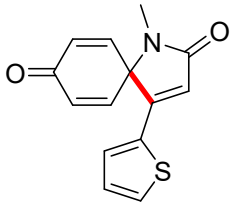
4-(4-ethylphenyl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4p): white solid, (26 mg, 93% yield), mp: 100 – 102 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.40 (d, J = 12.0 Hz, 2H), 7.16 (d, J = 12.0 Hz, 2H), 6.61 (s, 1H), 6.58 – 6.53 (m, 4H), 2.81 (s, 3H), 2.65 – 2.61 (m, 2H), 1.22 – 1.19 (m, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.3, 170.3, 156.1, 147.5, 146.2, 132.8, 128.6, 128.3, 126.7, 123.2, 66.9, 28.8, 25.1, 15.2. **HRMS (ESI)** m/z: calcd. for $C_{18}H_{17}NNaO_2^+$ $[M + Na]^+$ 302.1152, found 302.1152.



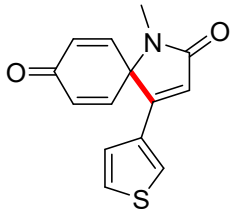
4-(4-chlorophenyl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4q)[15]: white solid, (26 mg, 91% yield), mp: 136 – 138 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.41 (d, J = 12.0 Hz, 2H), 7.32 (d, J = 12.0 Hz, 2H), 6.64 (s, 1H), 6.59 (d, J = 6.0 Hz, 2H), 6.53 (d, J = 12.0 Hz, 2H), 2.83 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.0, 169.8, 154.8, 145.7, 136.9, 133.1, 129.4, 129.2, 127.9, 124.6, 66.9, 25.2.



1-methyl-4-(naphthalen-2-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4r)[15]: white solid, (29 mg, 96% yield), mp: 160 – 162 °C, eluent PE/EtOAc (1:2, v/v)^{1}H NMR (600 MHz, $CDCl_3$) δ: 7.94 (s, 1H), 7.81 – 7.79 (m, 2H), 7.73 (d, *J* = 12.0 Hz, 1H), 7.57 (d, *J* = 12.0 Hz, 1H), 7.53 – 7.49 (m, 2H), 6.77 (s, 1H), 6.62 – 6.61 (m, 4H), 2.85 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.3, 170.1, 155.8, 146.3, 134.0, 132.9, 132.8, 129.0, 128.8, 128.1, 127.8, 127.8, 127.1, 126.4, 124.3, 123.8, 66.9, 25.1.



1-methyl-4-(thiophen-2-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4s): white solid, (25 mg, 97% yield), mp: 169 – 171 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.39 (d, *J* = 6.0 Hz, 1H), 7.22 (d, *J* = 6.0 Hz, 1H), 7.00 – 6.99 (m, 1H), 6.59 (d, *J* = 6.0 Hz, 2H), 6.54 (d, *J* = 12.0 Hz, 2H),6.50 (s, 1H), 2.83 (s, 3H). **^{13}C{^{1}H} NMR** (100 MHz, $CDCl_3$) δ: 184.2, 170.0, 149.7, 145.9, 133.3, 133.2, 129.0, 128.3, 127.8, 121.6, 66.7, 25.3. **HRMS (ESI)** m/z: calcd. for $C_{14}H_{14}NO_2S^+$ $[M + H]^+$ 258.0584, found 258.0584.



1-methyl-4-(thiophen-3-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (4t)[15]: white solid, (24 mg, 93% yield), mp: 214 – 216 °C, eluent PE/EtOAc (1:2, v/v). **^{1}H NMR** (600 MHz, $CDCl_3$) δ: 7.44 – 7.43 (m, 1H), 7.35 – 7.33 (m, 1H), 7.25 – 7.23 (m, 1H), 6.59 (d, *J* = 12.0 Hz, 2H), 6.55 (d, *J* = 12.0 Hz, 2H), 6.51 (s, 1H), 2.83 (s, 3H). **^{13}C{^{1}H} NMR** (150 MHz, $CDCl_3$) δ: 184.2, 170.4, 150.8, 146.4, 132.9, 132.0, 127.0, 126.2, 125.0, 122.6, 66.7, 25.2.

6.Reference

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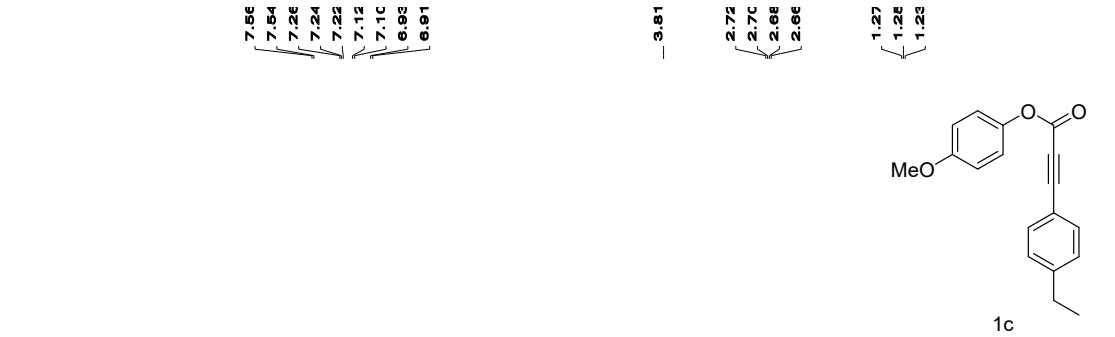
7.Copies of ^{1}H and $^{13}C\{^{1}H\}$ NMR Spectra



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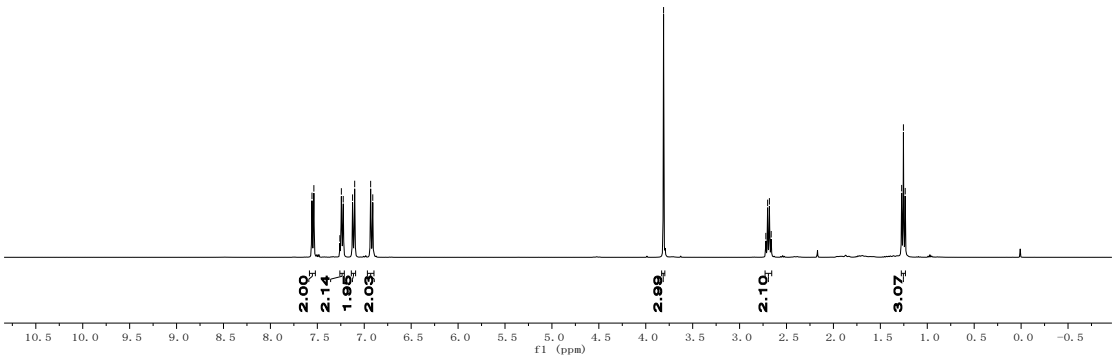

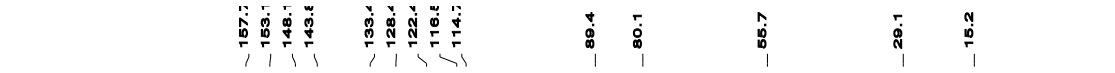

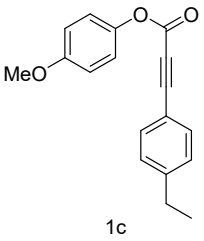

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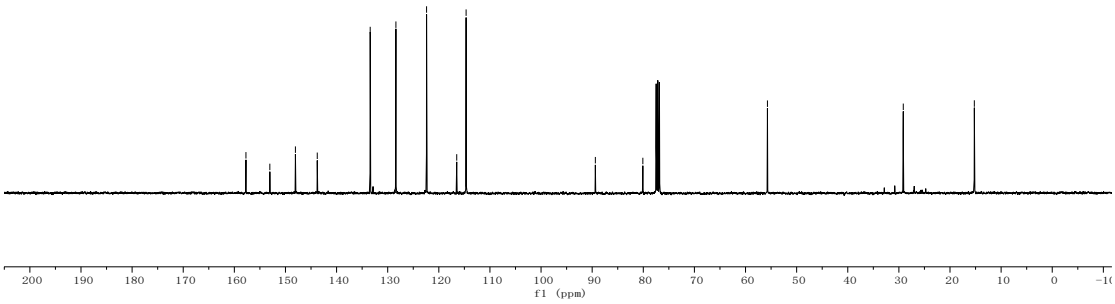

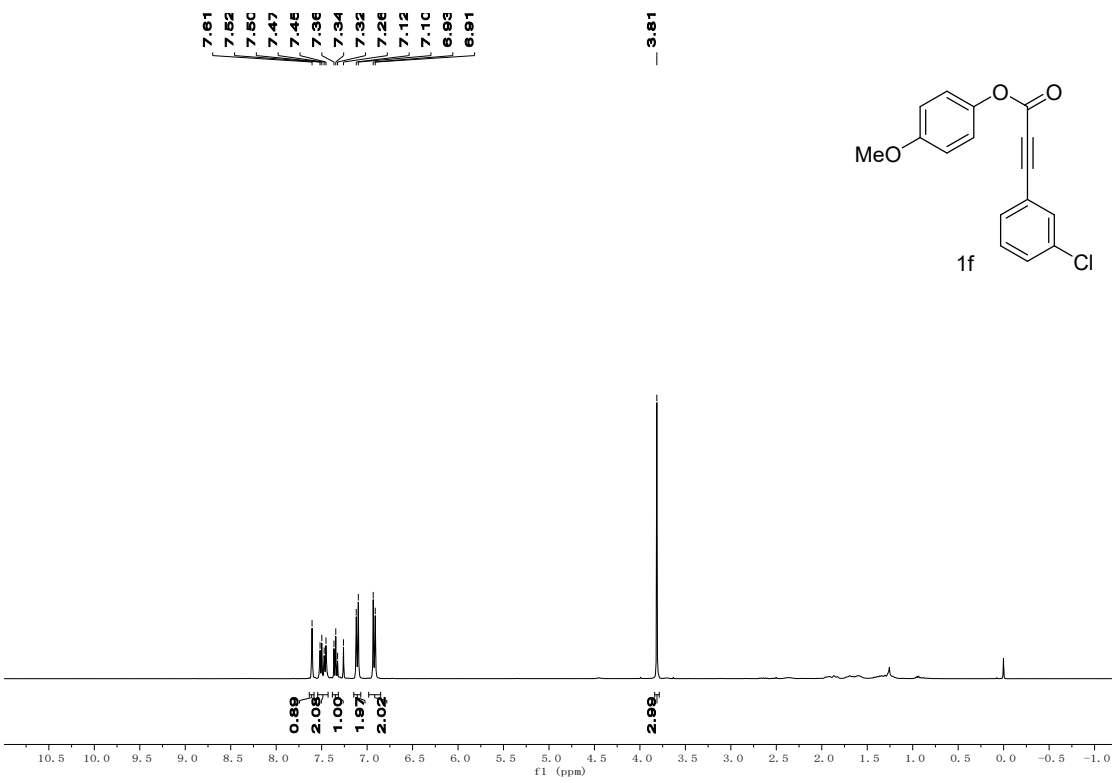
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7.10
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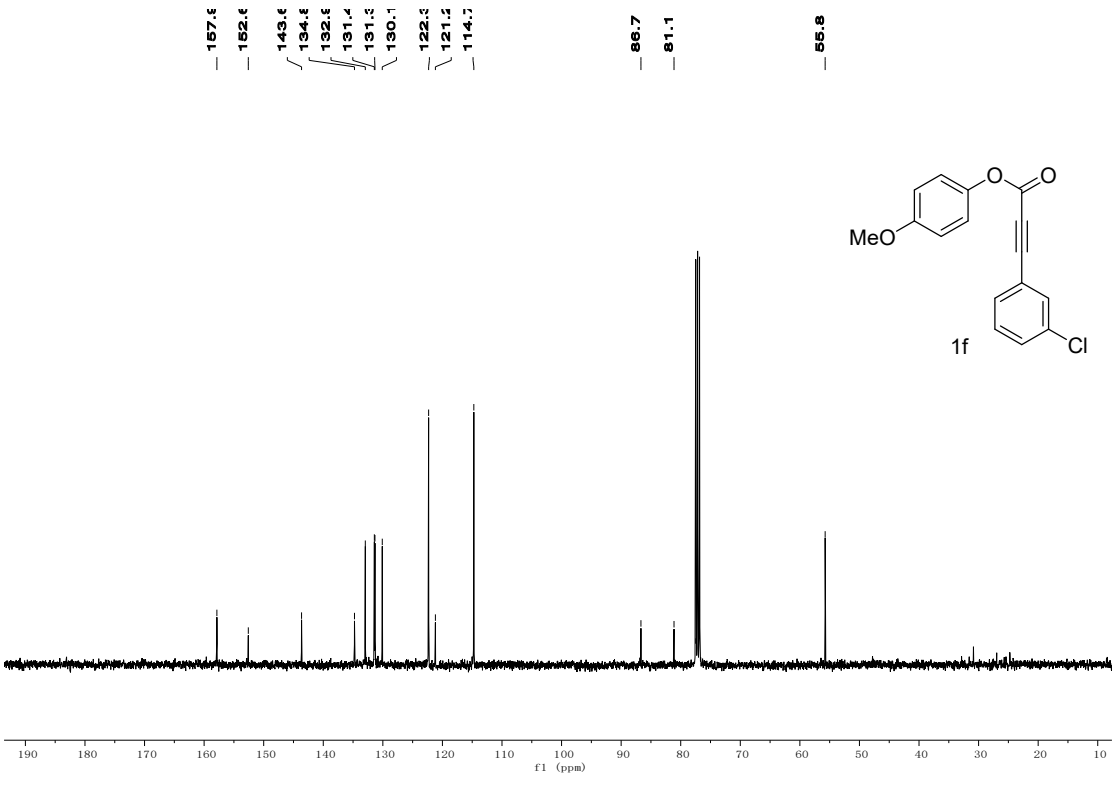
6.91
3.81
MeO
O
O
Cl
1f
0.89
2.05
1.00
1.97
2.04
2.99
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5 -1.0
f1 (ppm)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131.4
131.3
130.1
122.3
121.2
114.7
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81.1
55.8
MeO
O
O
Cl
1f
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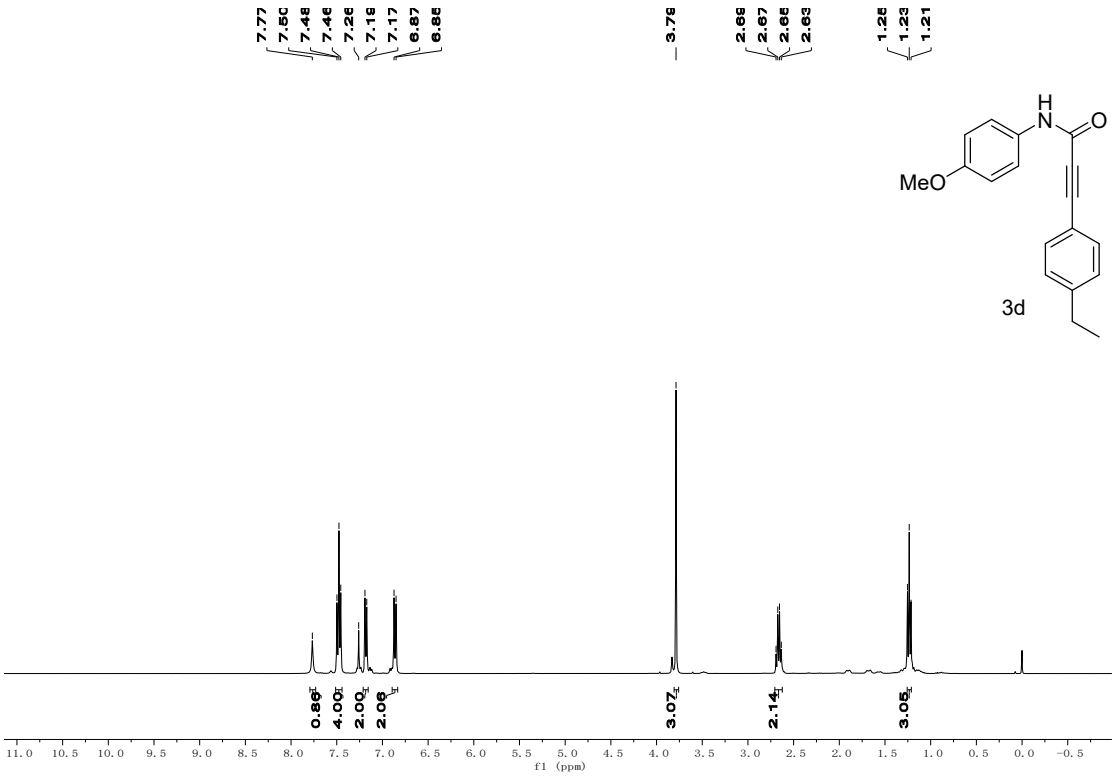
f1 (ppm)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2.63
1.25
1.23
1.21
H
N
O
MeO
3d
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2.00
2.08
3.07
2.14
3.05
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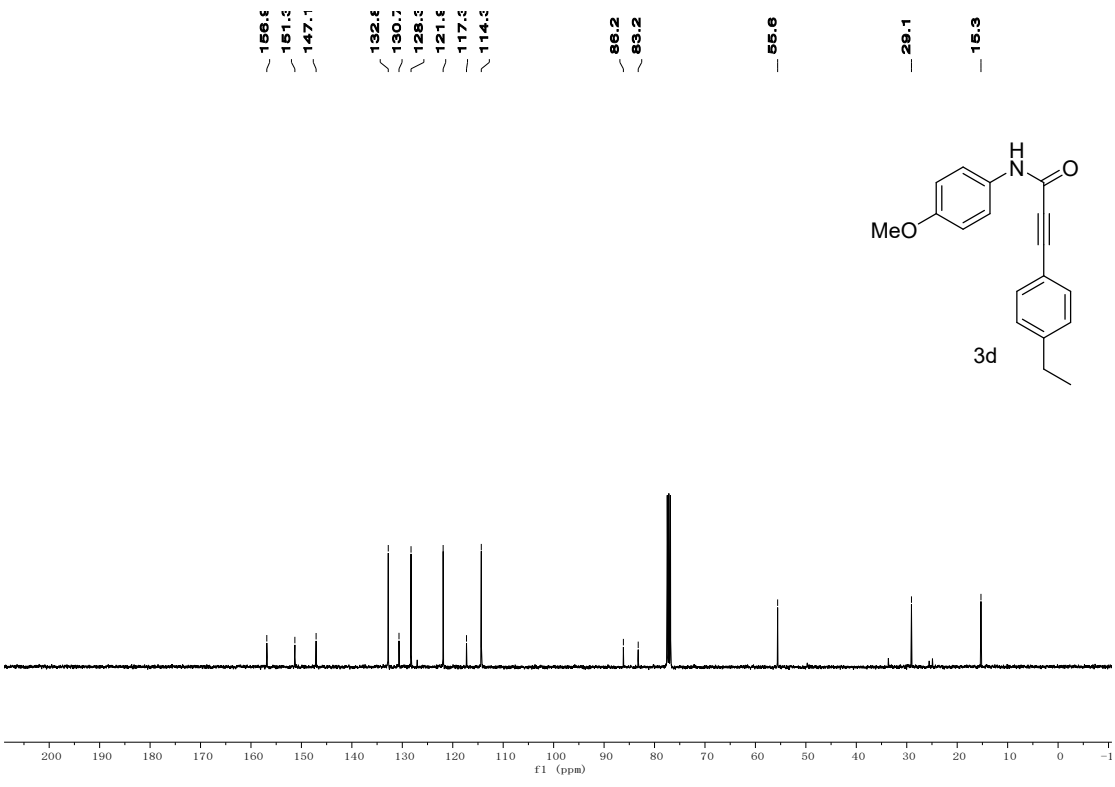
f1 (ppm)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N
O
MeO
3d
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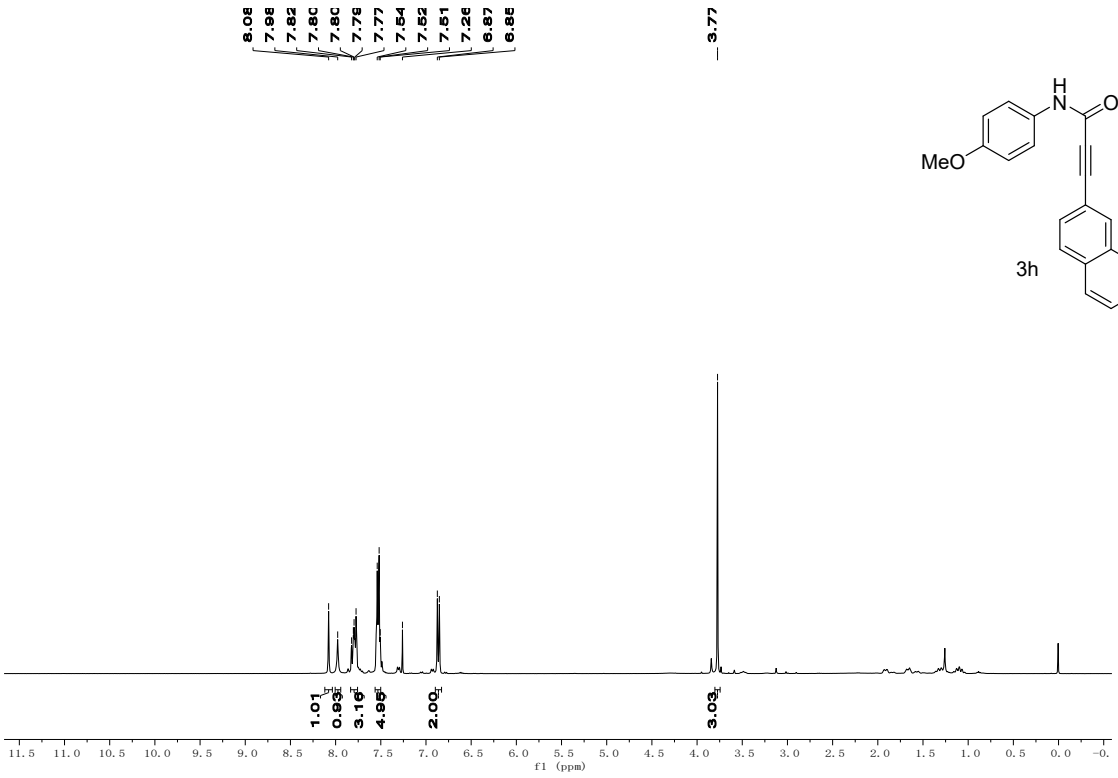
f1 (ppm)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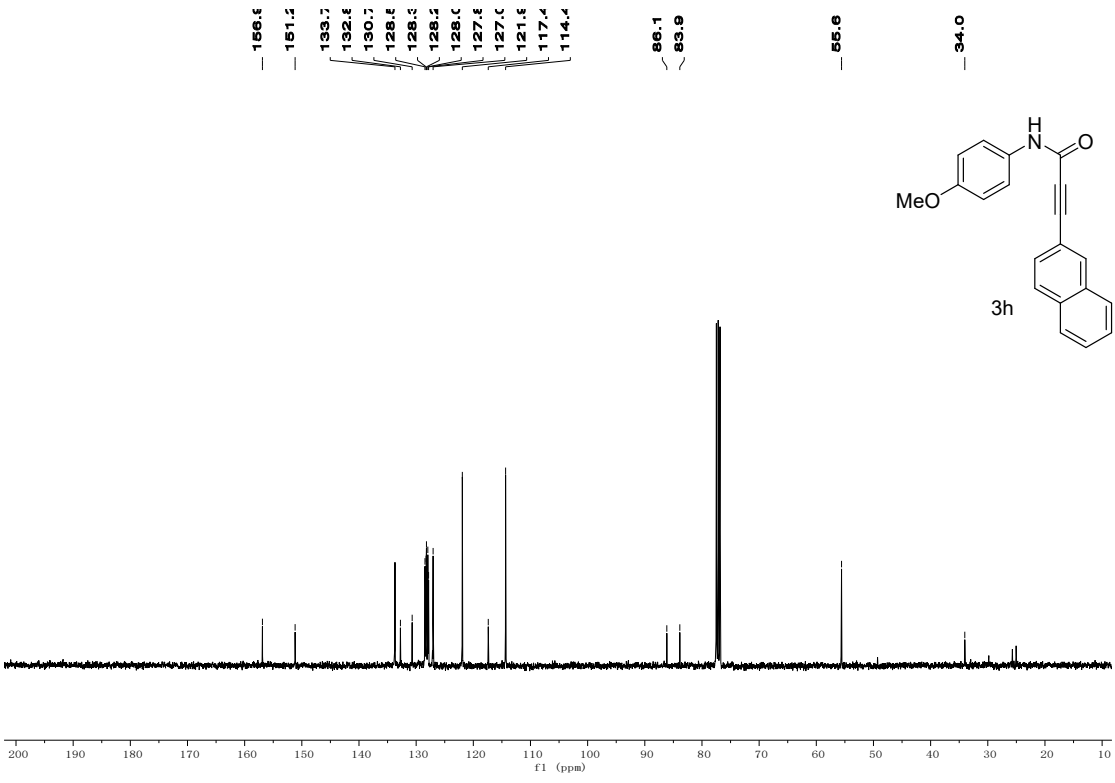
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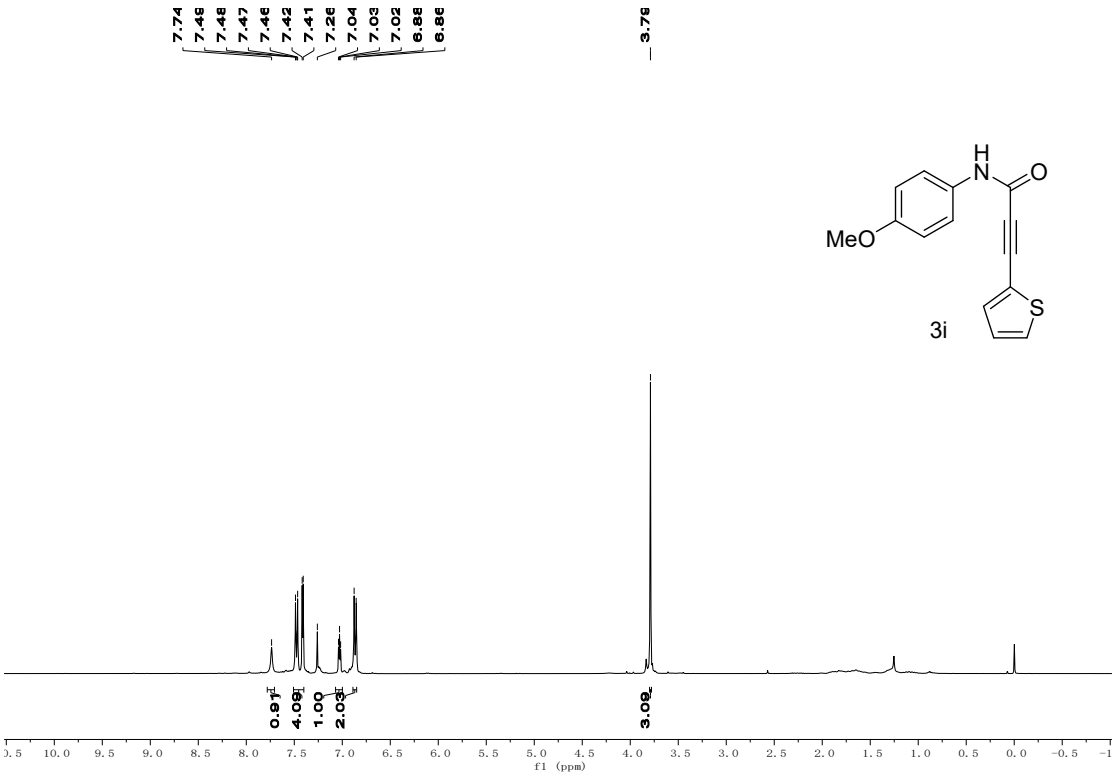
3h
f1 (ppm)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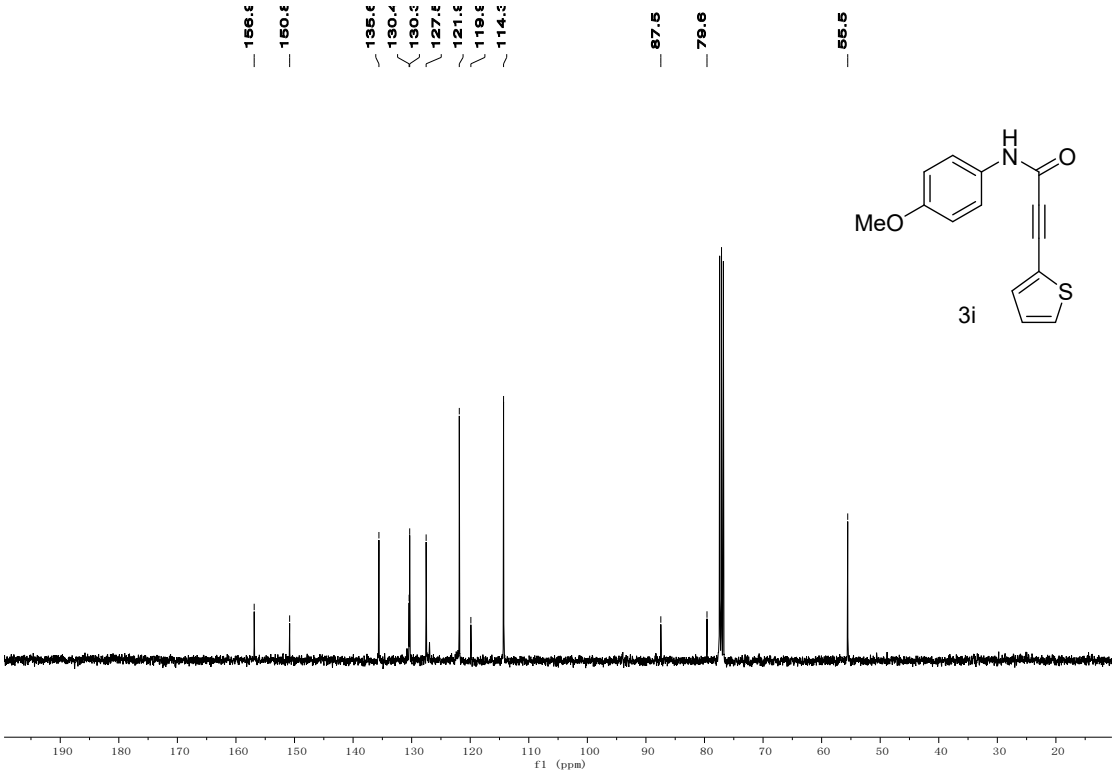
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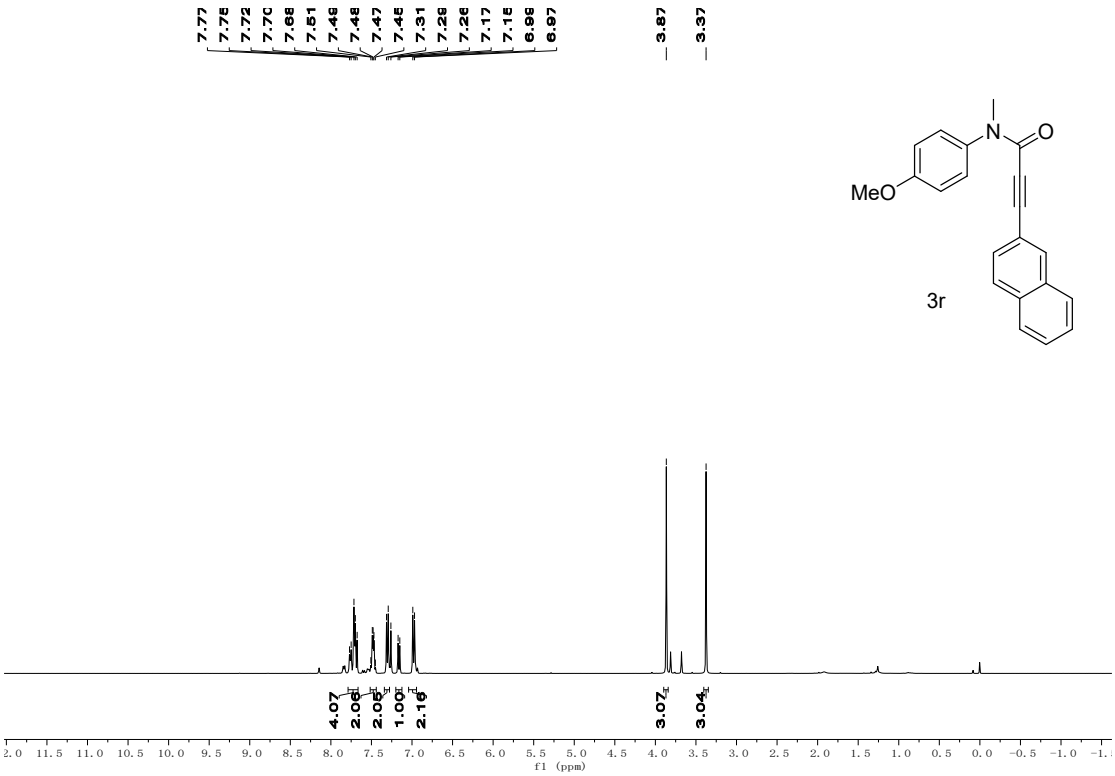
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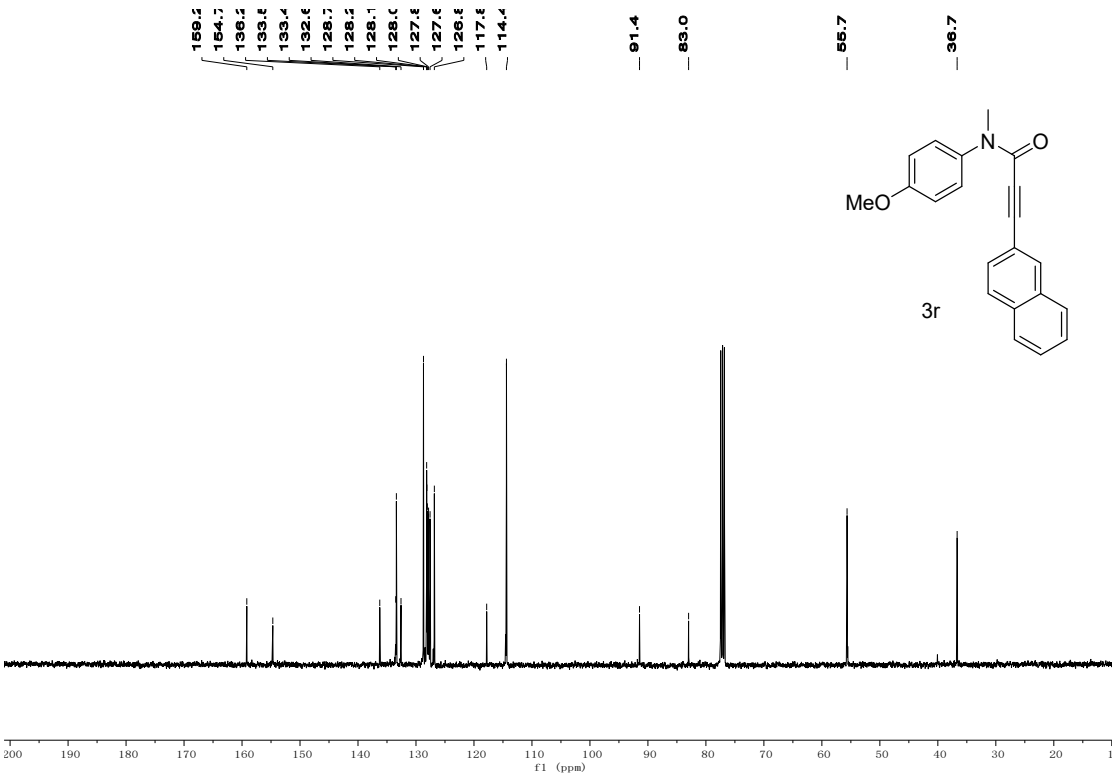
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117.8
114.4
91.4
83.0
55.7
36.7
MeO
N
O
3r
f1 (ppm)


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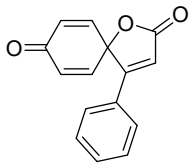

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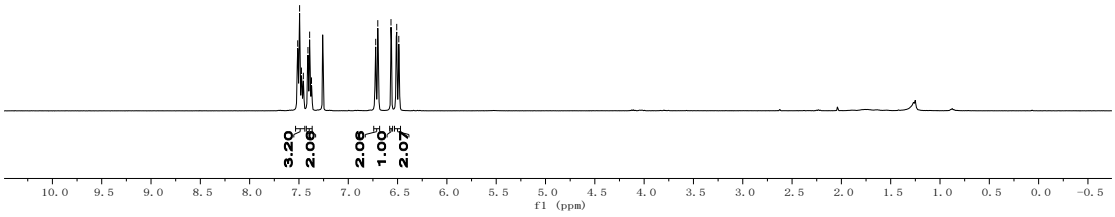
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f1 (ppm)


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165.4
143.8
132.2
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128.4
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127.3
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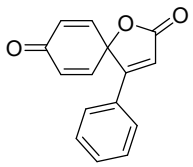
81.5



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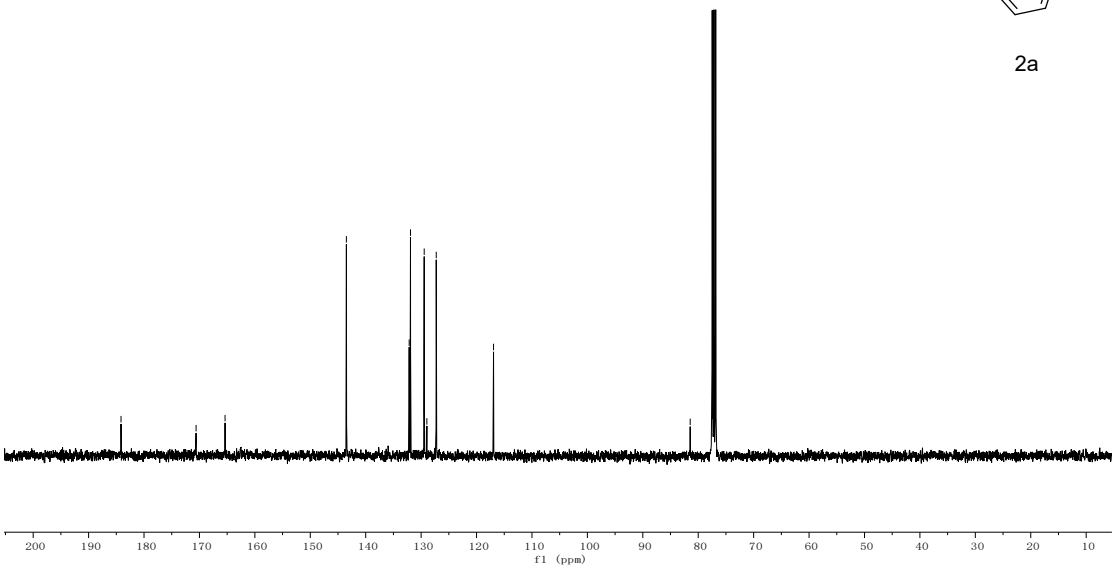
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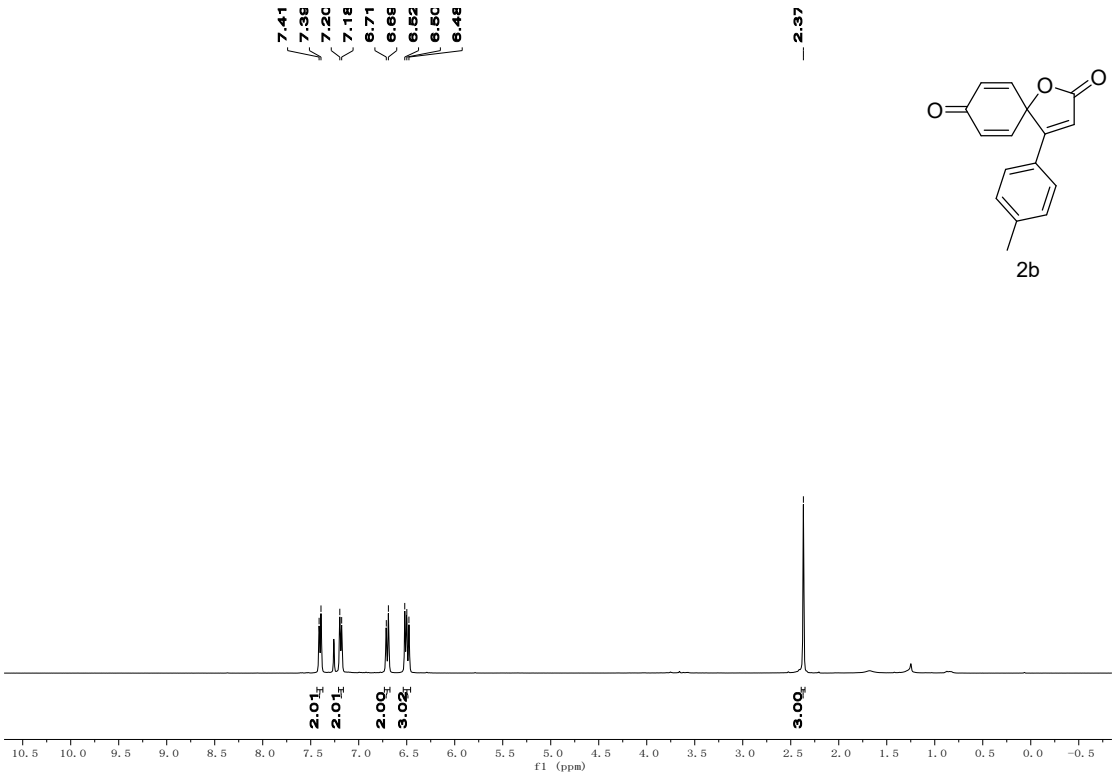
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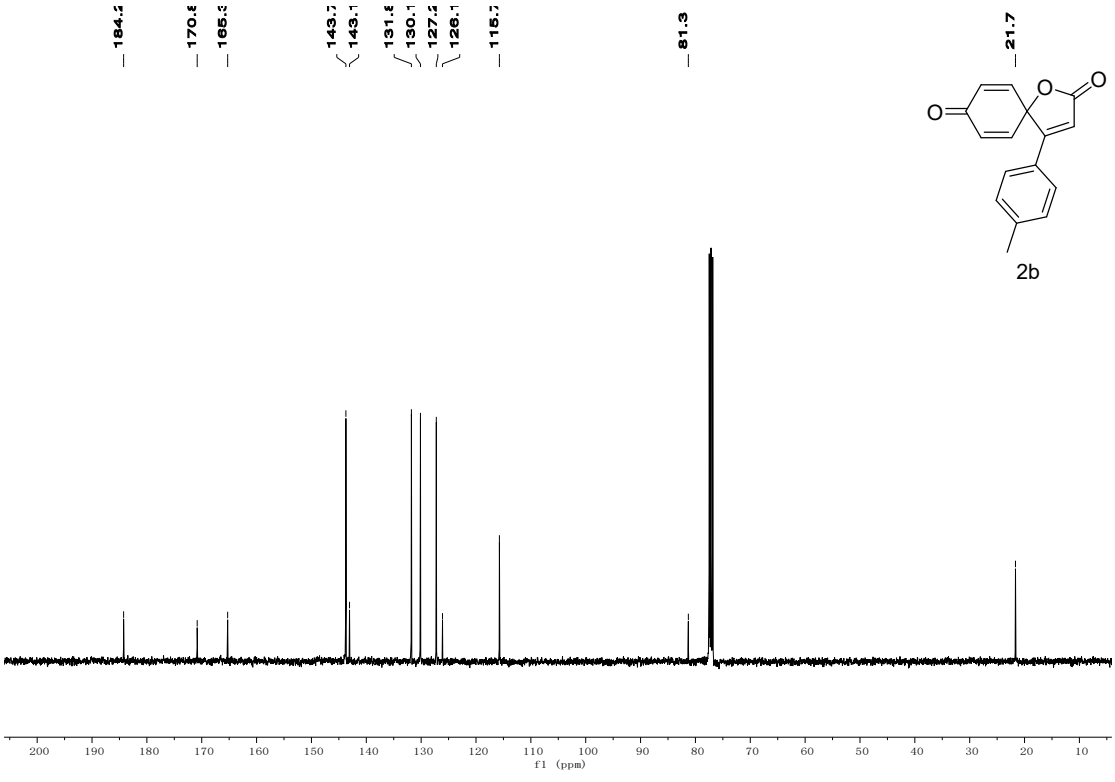
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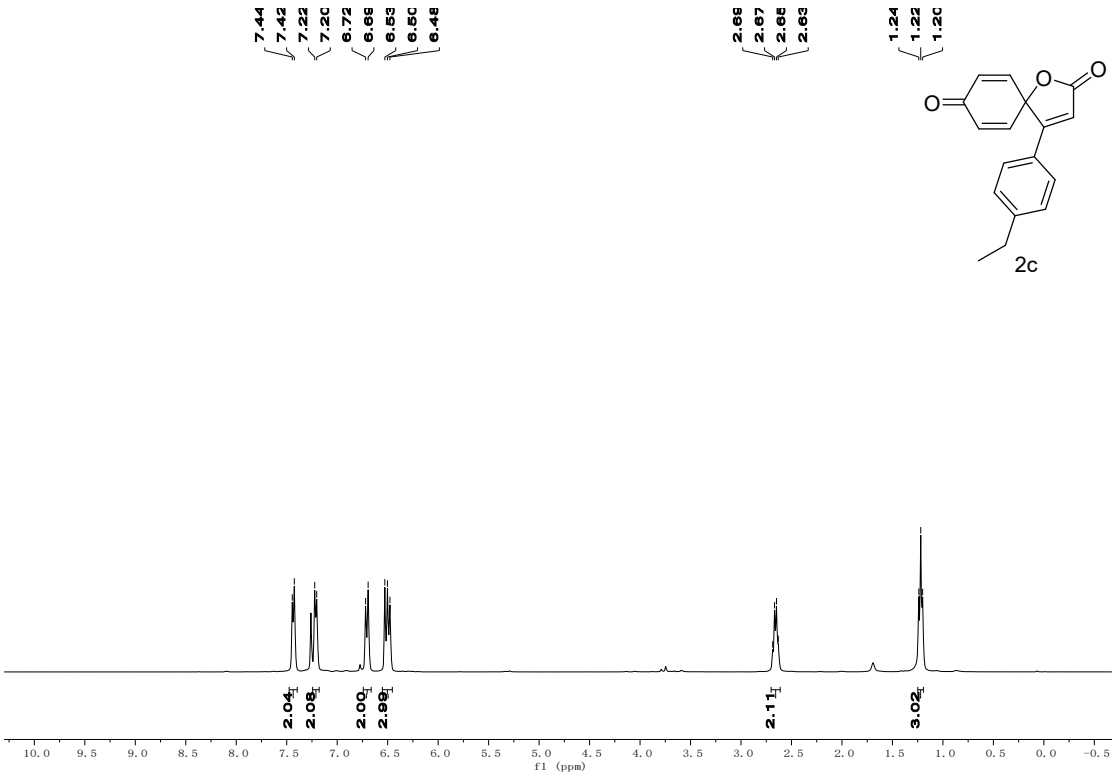
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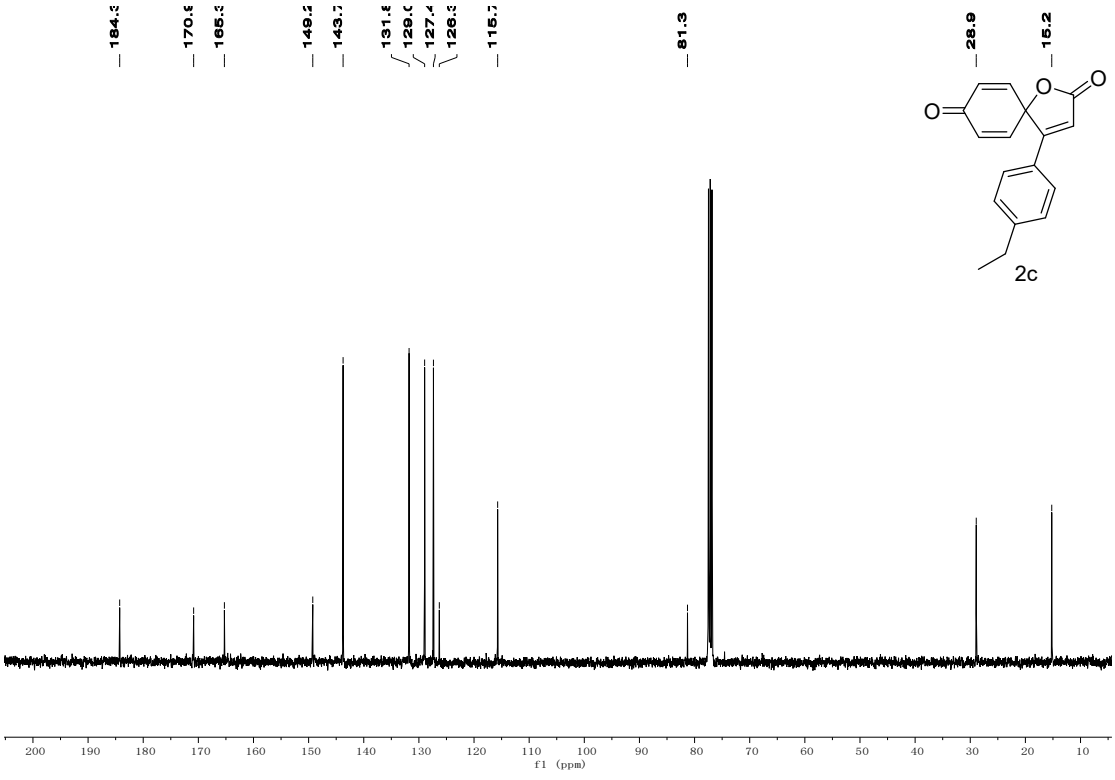
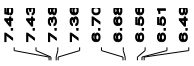
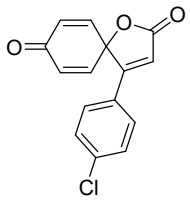
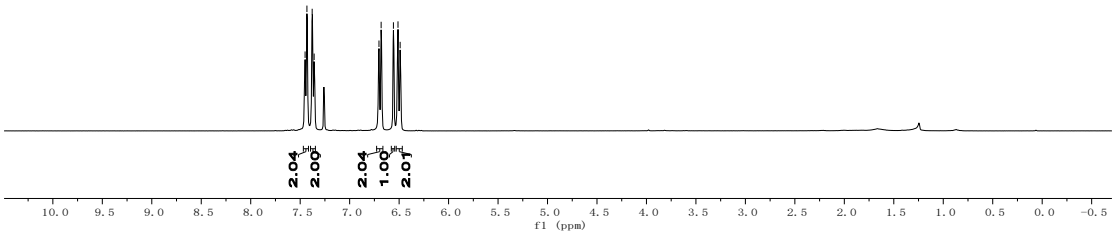
6.68
6.53
6.50
6.48
2.68
2.67
2.65
2.63
1.24
1.22
1.20
O
O
O
2c
2.04
2.08
2.00
2.99
2.11
3.02
f1 (ppm)

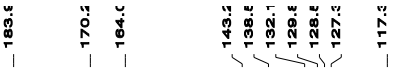

184.3
170.6
165.3
148.4
143.7
131.6
129.0
127.4
128.3
115.7
81.3
28.9
15.2
O
O
O
2c
f1 (ppm)

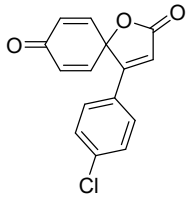

7.45
7.43
7.38
7.36
6.70
6.68
6.56
6.51
6.48


O
O
O
Cl

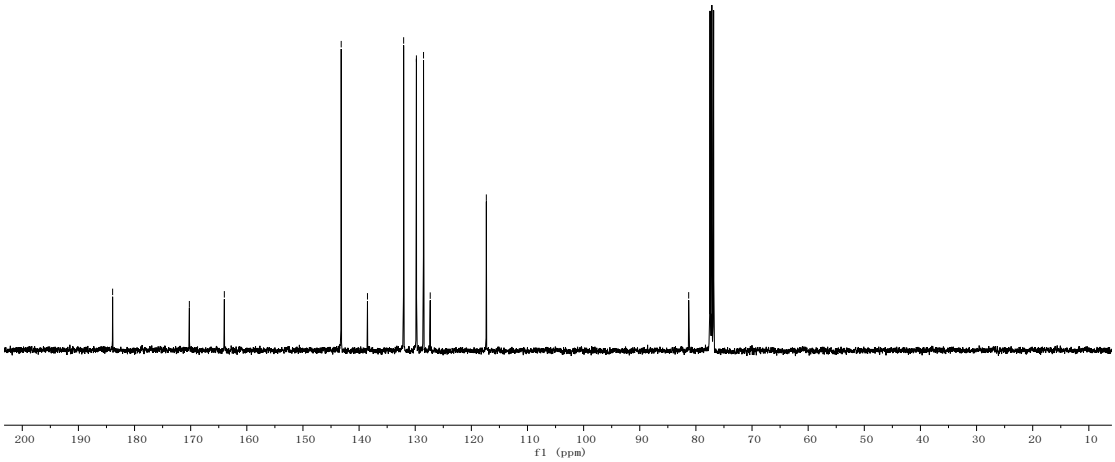
2d

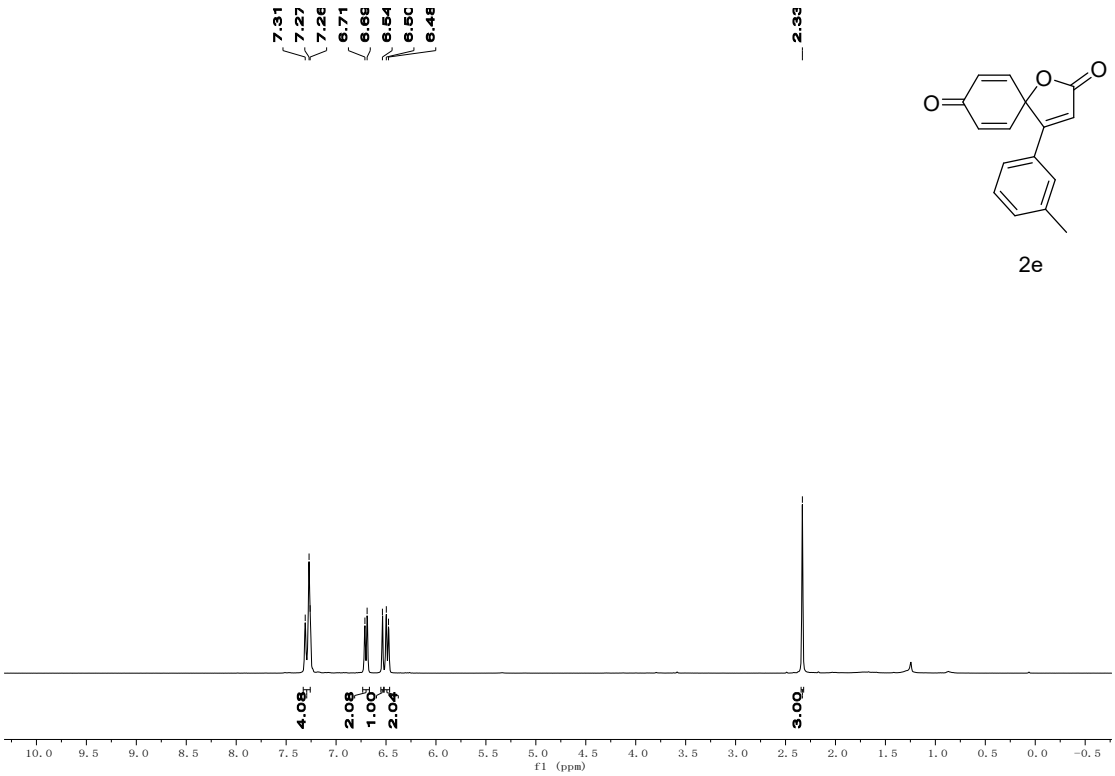

2.04
2.00
2.04
1.00
2.01
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)


183.6
170.2
164.0
143.2
138.8
132.1
129.8
128.8
127.3
117.3
81.3

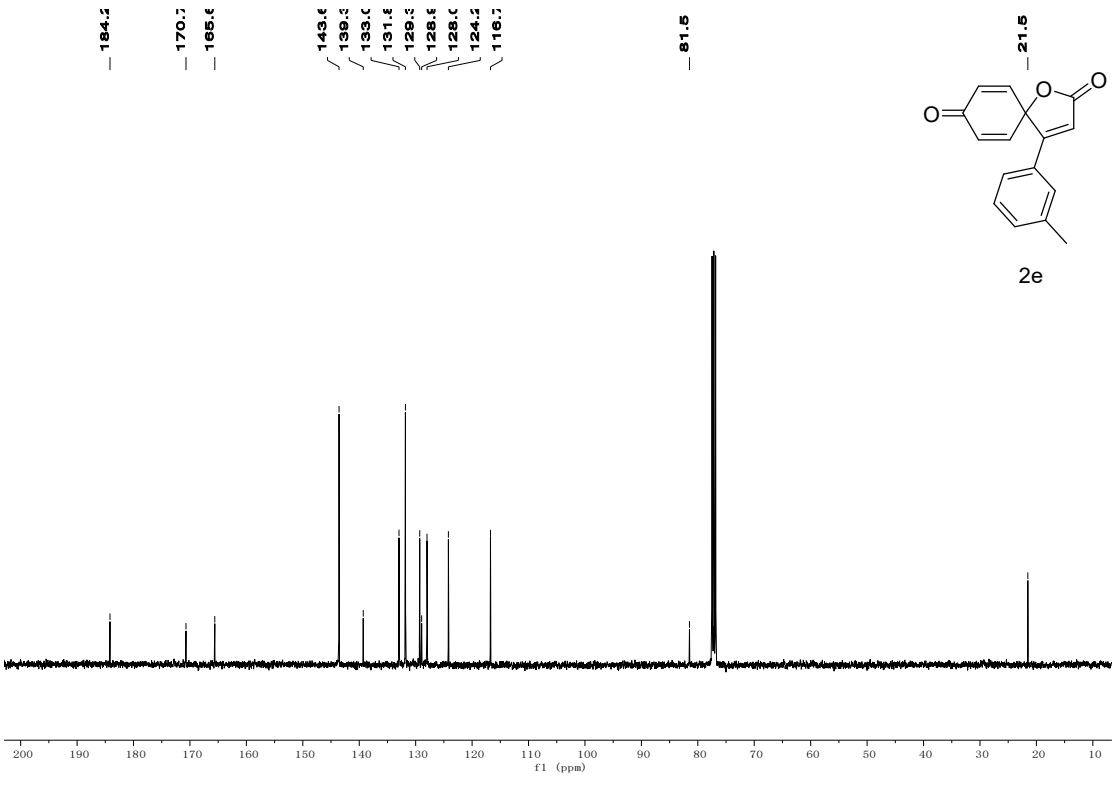

O
O
O
Cl

2d


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

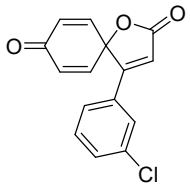

7.31
7.27
7.26
6.71
6.68
6.54
6.50
6.48
2.33
O
O
O
4.08
2.08
1.00
2.04
3.00
f1 (ppm)

2e

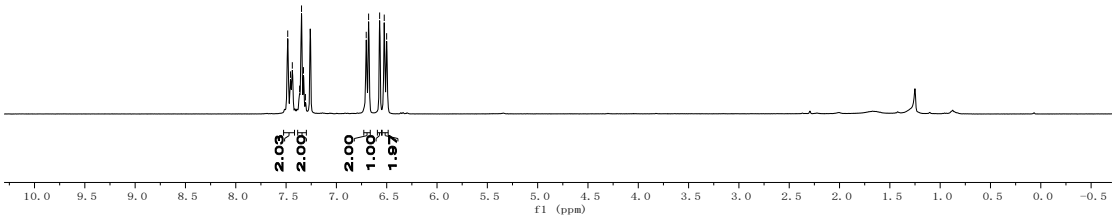

184.2
170.7
165.6
143.6
139.3
133.0
131.8
129.3
128.6
128.0
124.2
116.7
81.5
21.5
O
O
O
f1 (ppm)

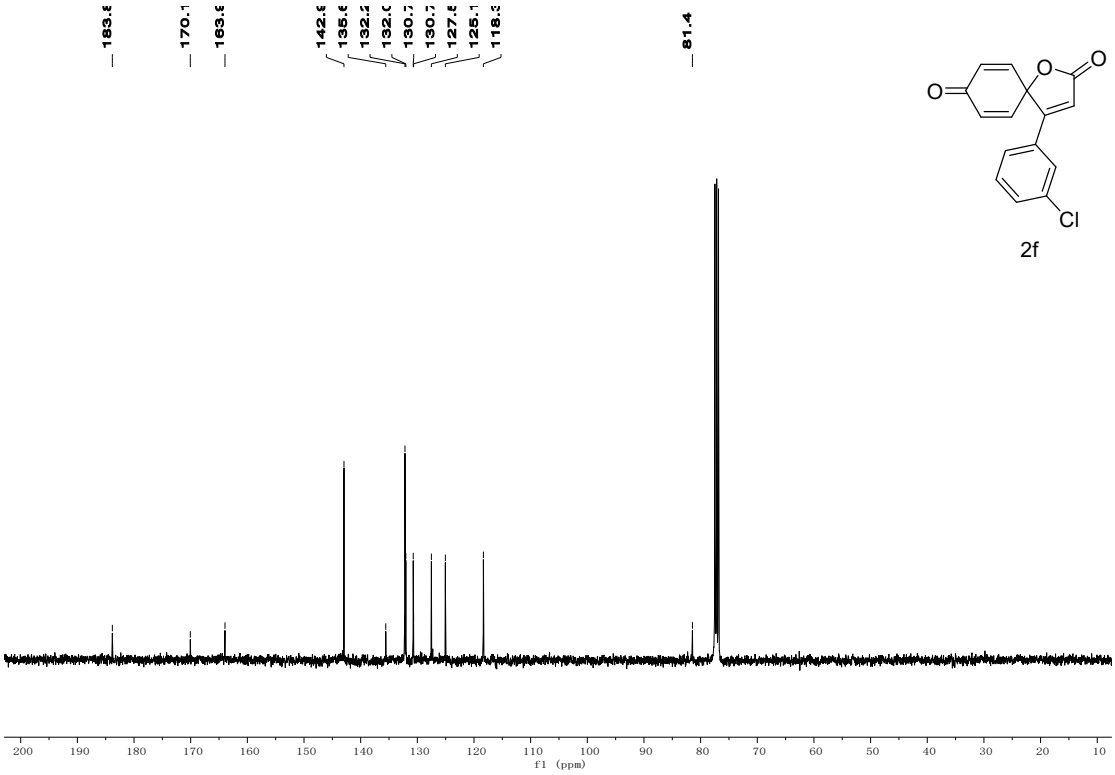
2e


7.48
7.46
7.44
7.37
7.35
7.33
7.31
6.70
6.68
6.57
6.52
6.50


O
O
O
Cl

2f


2.03
2.00
2.00
1.00
1.97
10.0 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 -0.5
f1 (ppm)

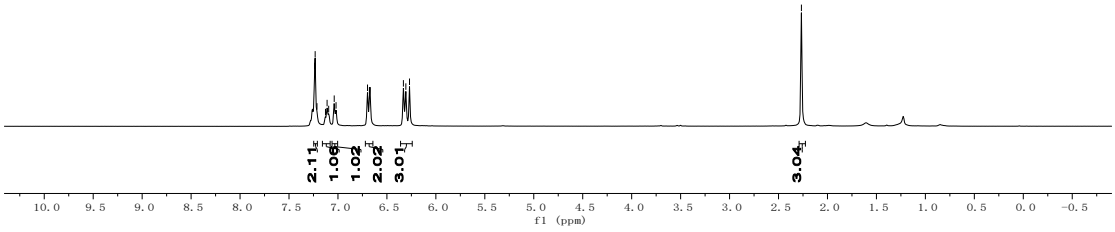

183.8
170.1
163.8
142.6
135.6
132.4
132.0
130.7
130.2
127.8
125.1
118.3
81.4
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

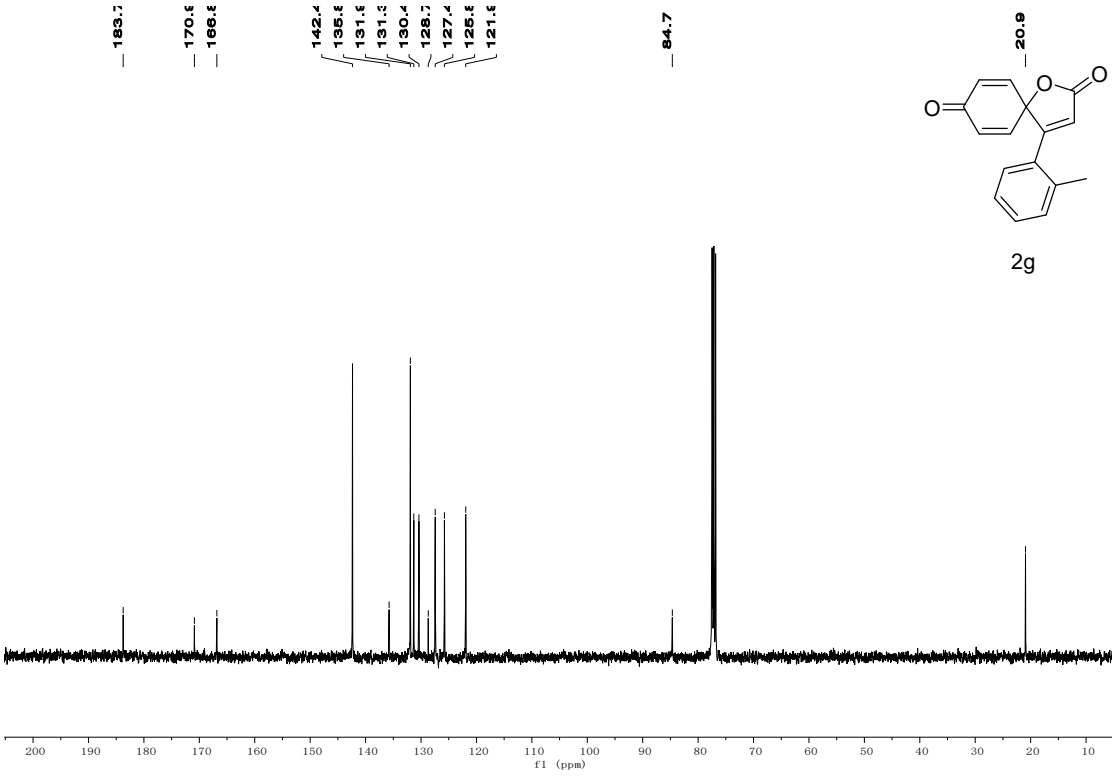
O
O
O
Cl

2f

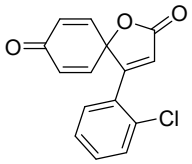

7.24
7.23
7.21
7.13
7.11
7.08
7.04
7.02
6.70
6.68
6.33
6.31
6.27
2.27
O
O
O

2g

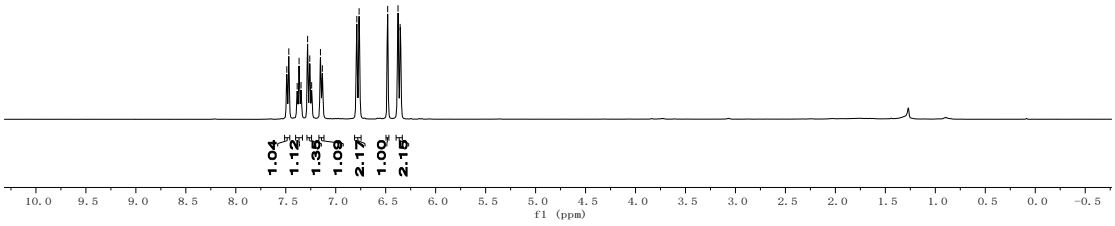

2.11
1.06
1.02
2.02
3.01
3.04
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)

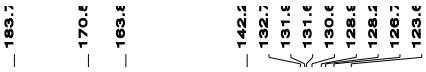

183.7
170.5
166.8
142.4
135.8
131.5
131.3
130.4
128.7
127.4
125.8
121.6
84.7
20.9
O
O
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

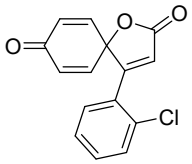
2g


O
O
O
Cl

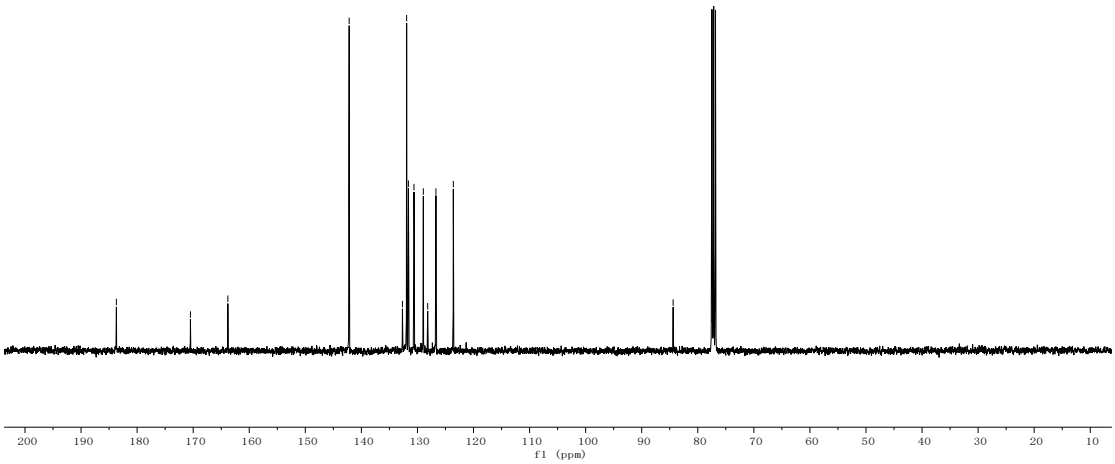
2h


1.04
1.13
1.35
1.09
2.17
1.00
2.15
10.0 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 −0.5
f1 (ppm)

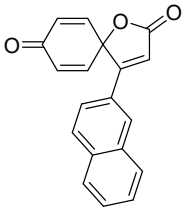

84.4


O
O
O
Cl

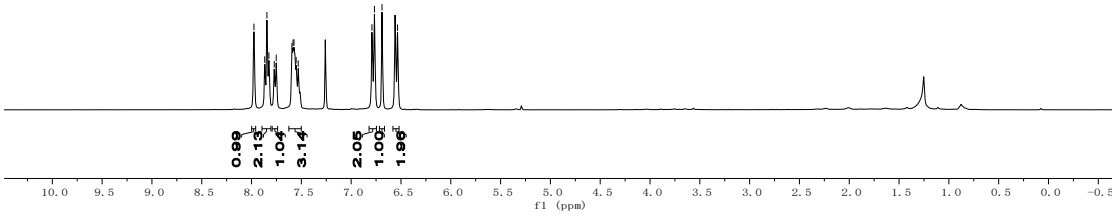
2h

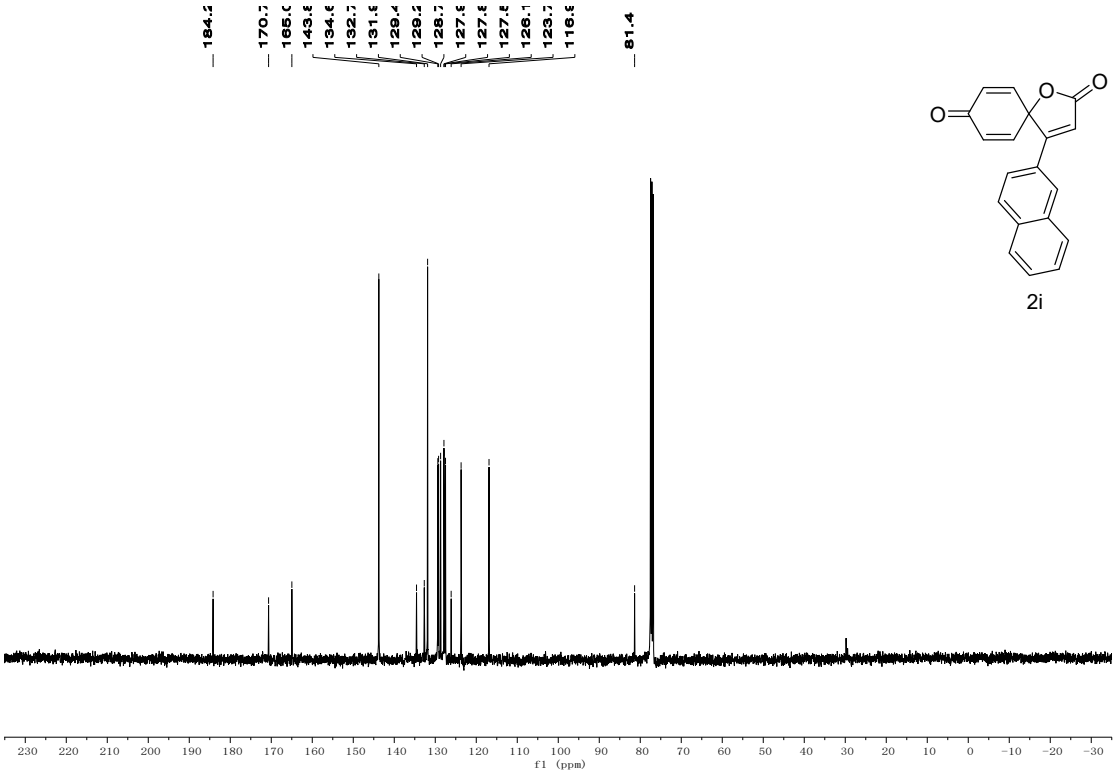

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.98
7.87
7.85
7.83
7.77
7.75
7.56
7.58
7.57
7.55
7.53
6.78
6.77
6.66
6.55
6.53

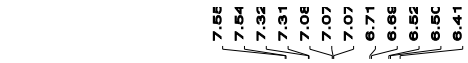


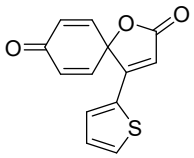
2i


0.99
2.13
1.04
3.14
2.05
1.05
1.96
f1 (ppm)

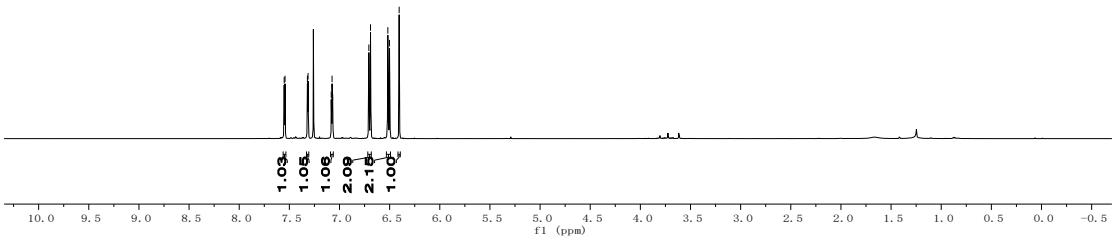

81.4
f1 (ppm)

2i

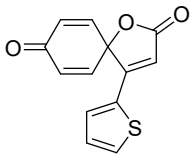

7.55
7.54
7.32
7.31
7.08
7.07
7.07
6.71
6.68
6.52
6.50
6.41


O
O
O
S

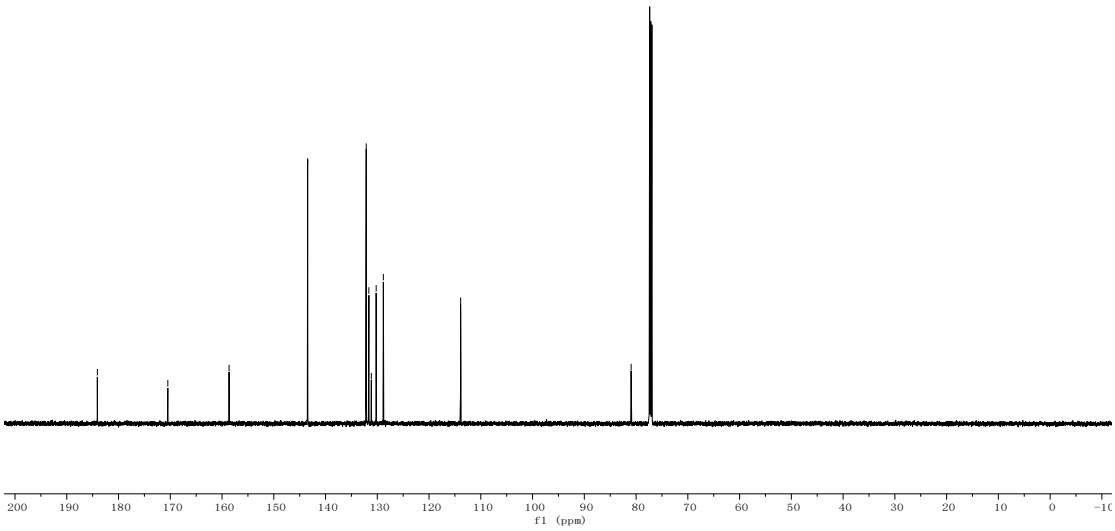
2j

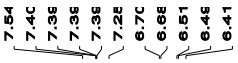

1.03
1.05
1.06
2.09
2.10
1.00
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)

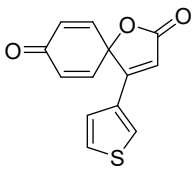

184.1
170.8
158.6
143.8
132.1
131.6
131.2
130.4
128.8
113.8
81.0


O
O
O
S

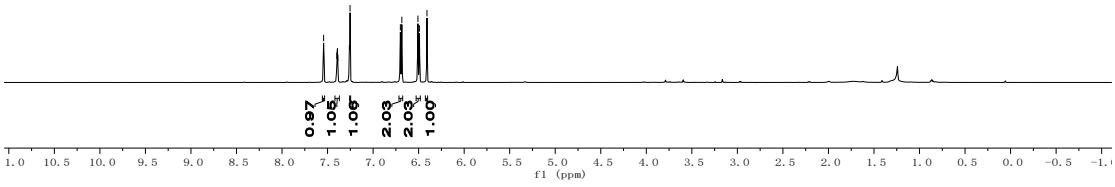
2j

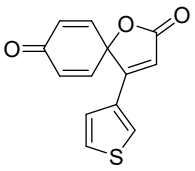

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 −10
f1 (ppm)



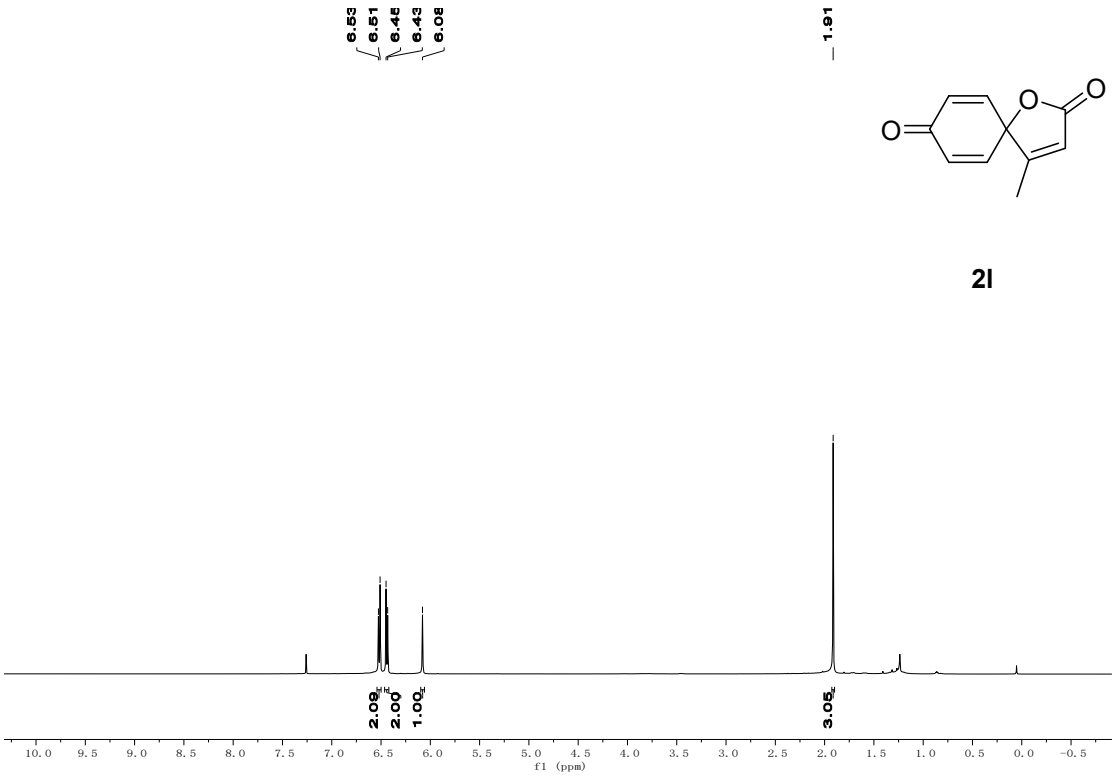


2k

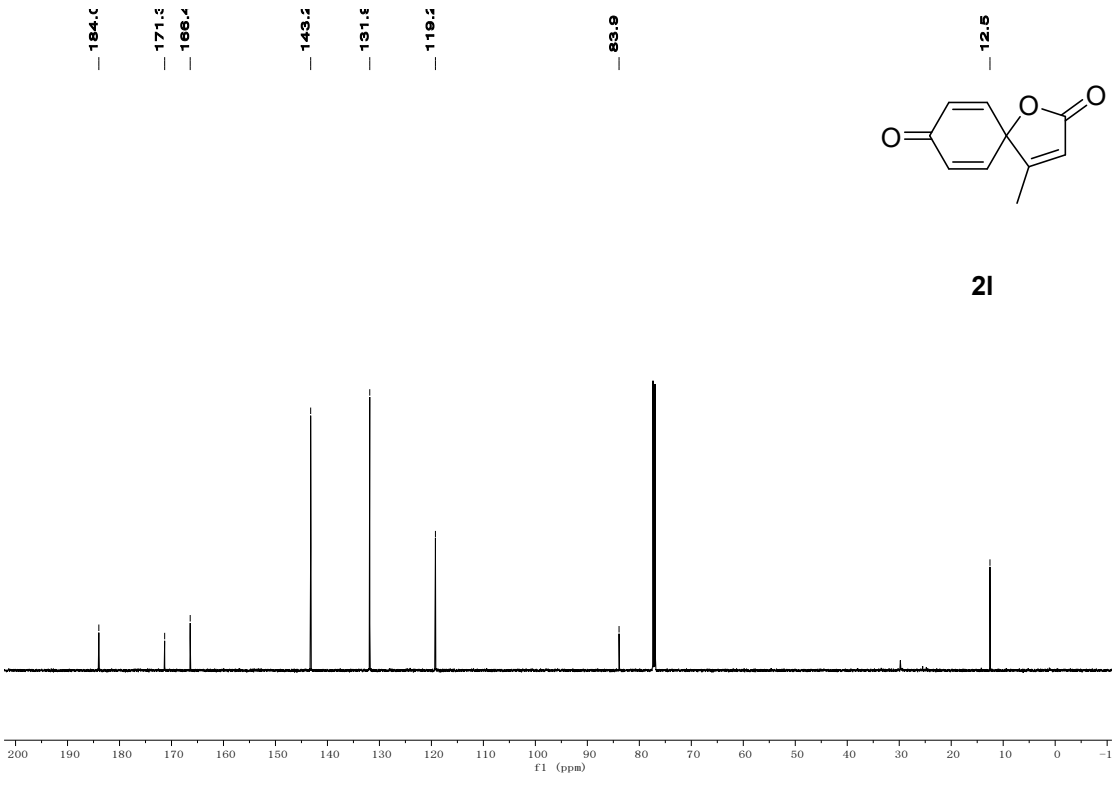




2k

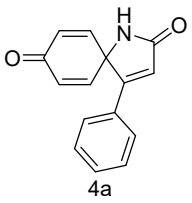

6.53
6.51
6.45
6.43
6.08
1.91
O
O
O
2.09
2.00
1.00
3.05
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)

2l

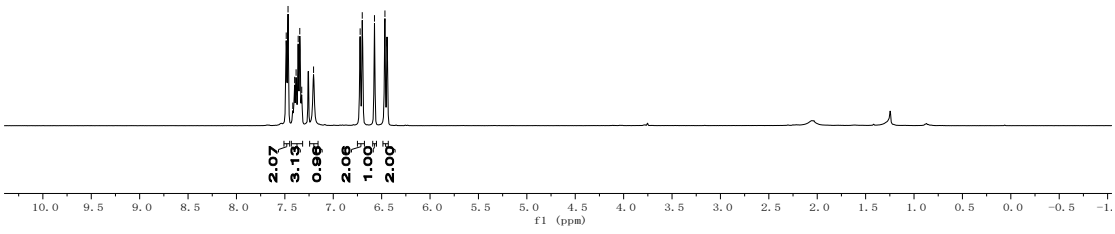

184.0
171.3
166.4
143.2
131.6
119.2
83.9
12.5
O
O
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 −1
f1 (ppm)

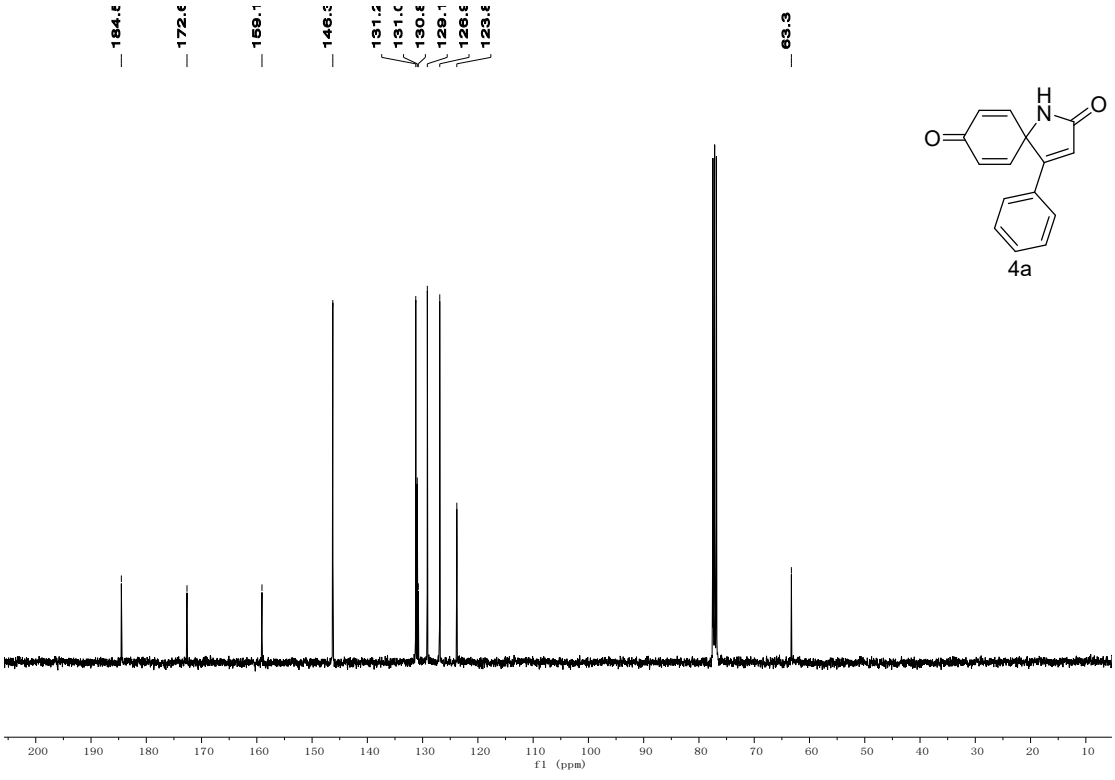
2l


7.46
7.47
7.42
7.40
7.38
7.36
7.35
7.33
7.20
6.72
6.70
6.57
6.47
6.45


H
N
O
O

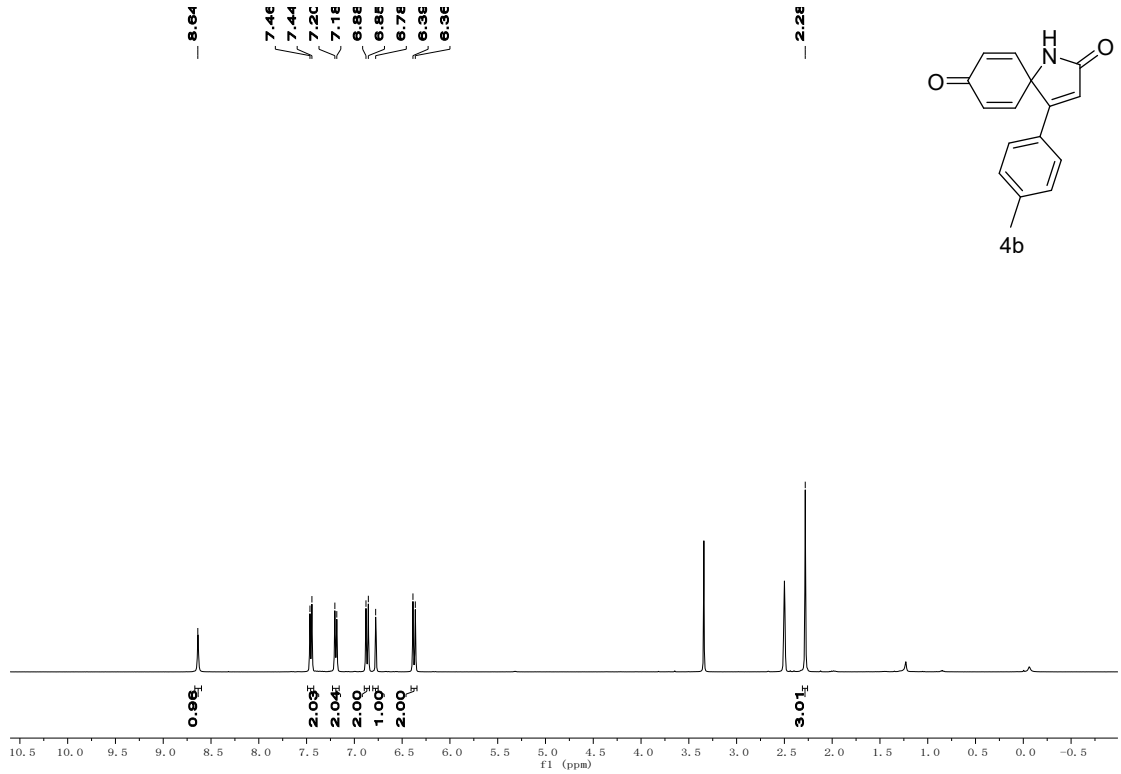
4a

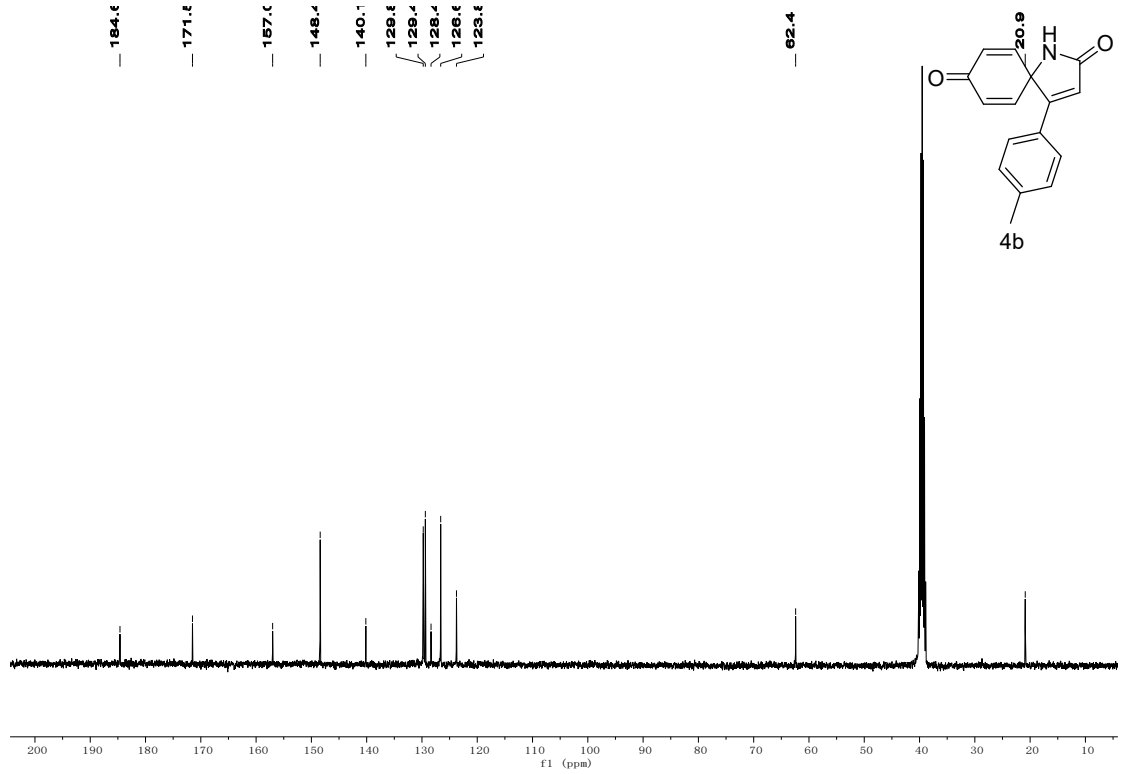

2.07
3.15
0.96
2.06
1.00
2.00
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)

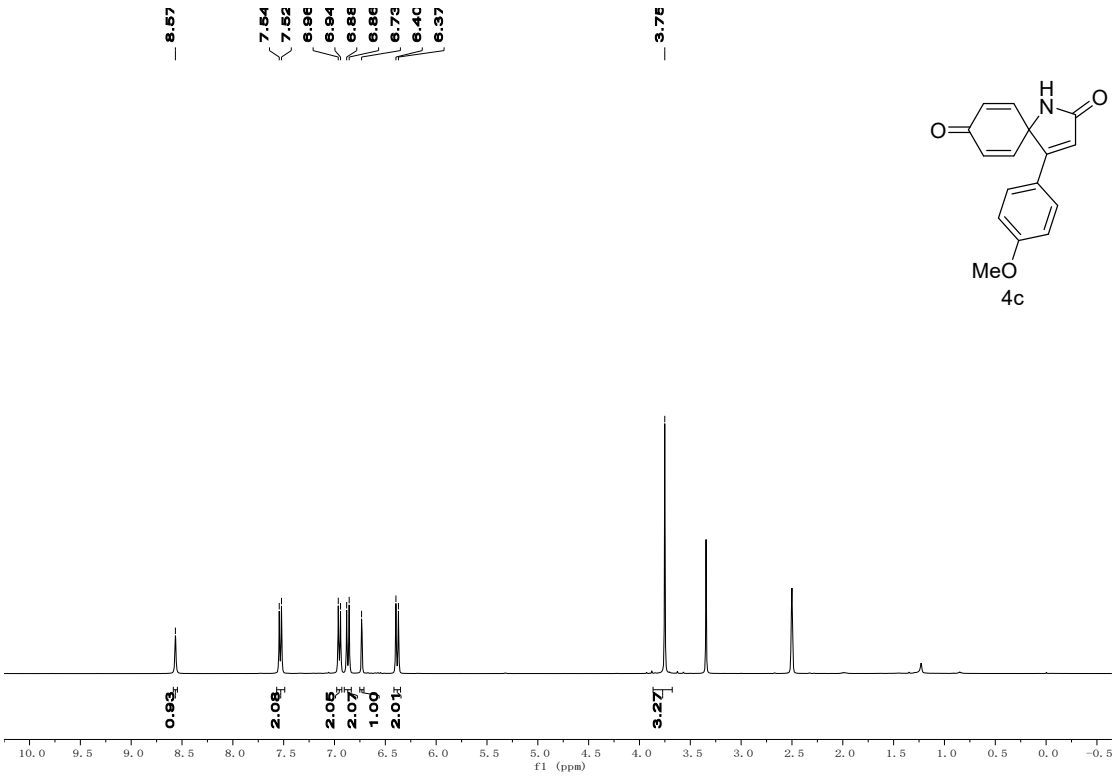

184.8
172.6
158.1
146.3
131.2
131.0
130.8
129.1
128.6
123.8
63.3
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

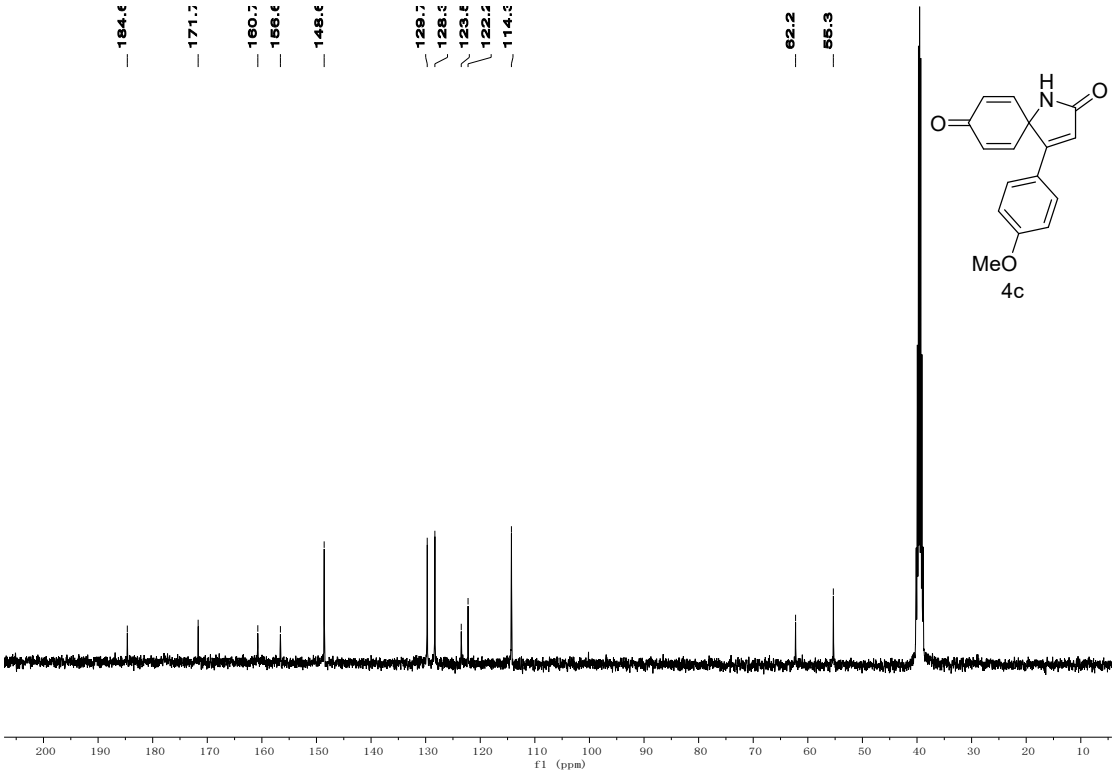
H
N
O
O

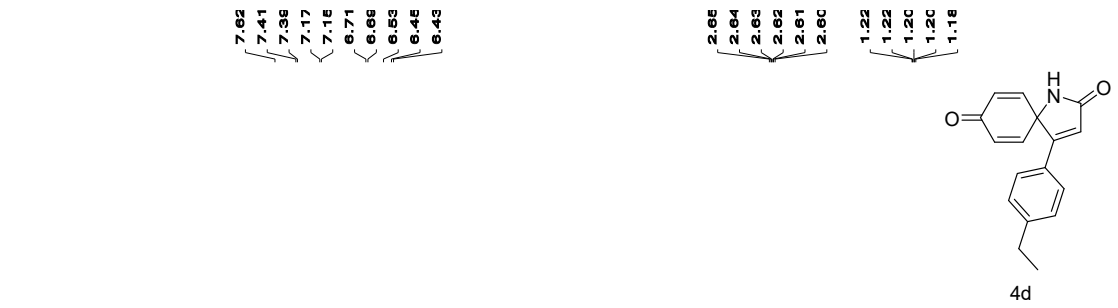
4a


8.64
7.46
7.44
7.20
7.18
6.88
6.85
6.78
6.36
6.36
2.28
0.96
2.03
2.04
2.00
1.00
2.00
3.01
f1 (ppm)
4b

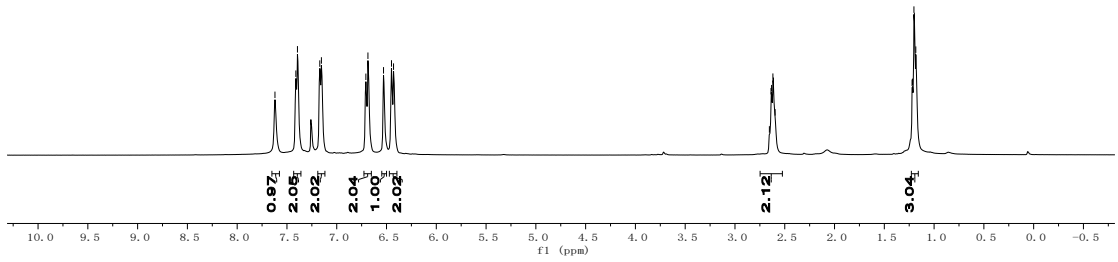

184.6
171.8
157.0
148.4
140.1
129.8
129.4
128.4
126.6
123.8
62.4
20.9
f1 (ppm)
4b

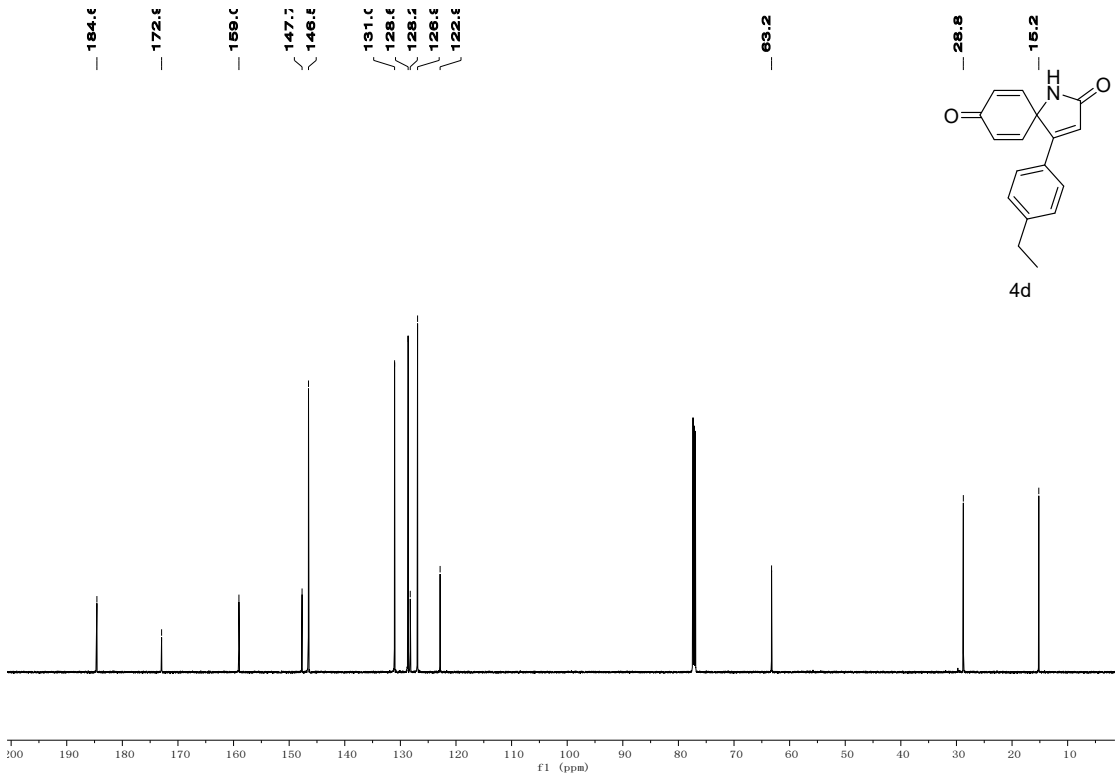

8.57
7.54
7.52
6.96
6.94
6.88
6.86
6.73
6.40
6.37
3.75
H
N
O
O
MeO
4c
0.93
2.08
2.05
2.07
1.00
2.01
3.27
f1 (ppm)


184.6
171.7
160.7
156.6
148.6
129.7
128.3
123.8
122.5
114.3
62.2
55.3
H
N
O
O
MeO
4c
f1 (ppm)

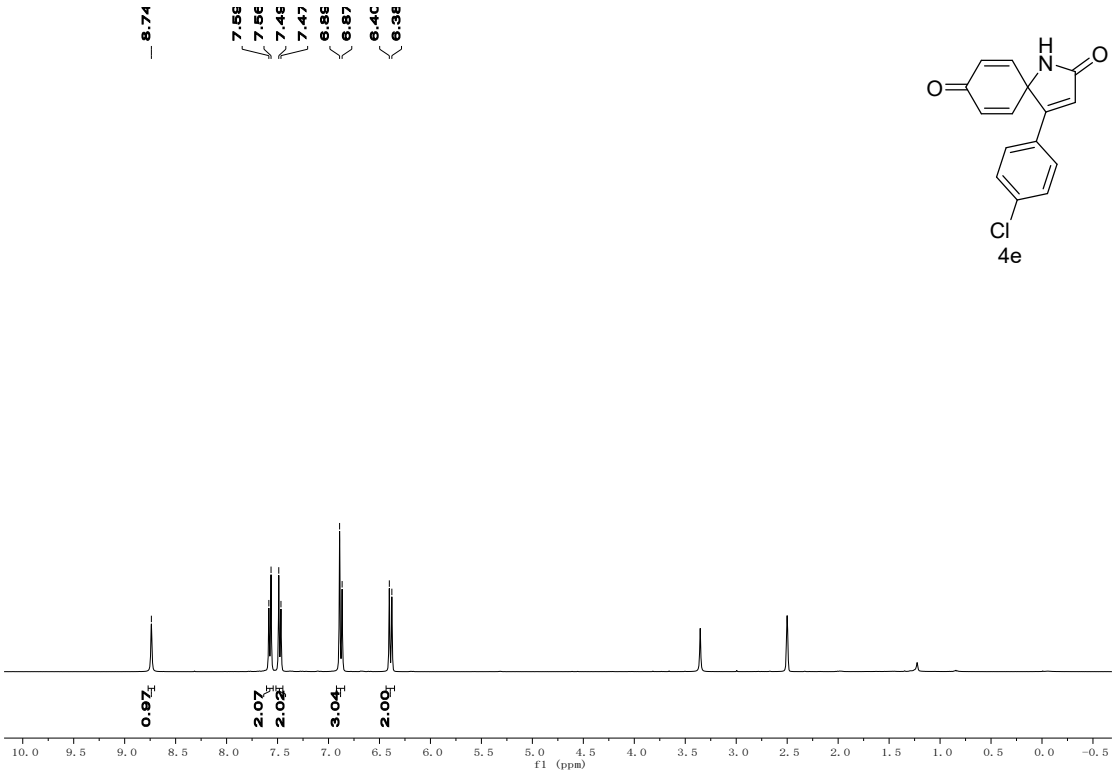

7.62
7.41
7.39
7.17
7.15
6.71
6.68
6.53
6.45
6.43
2.65
2.64
2.63
2.62
2.61
2.60
1.22
1.22
1.20
1.20
1.18
H
N
O
O

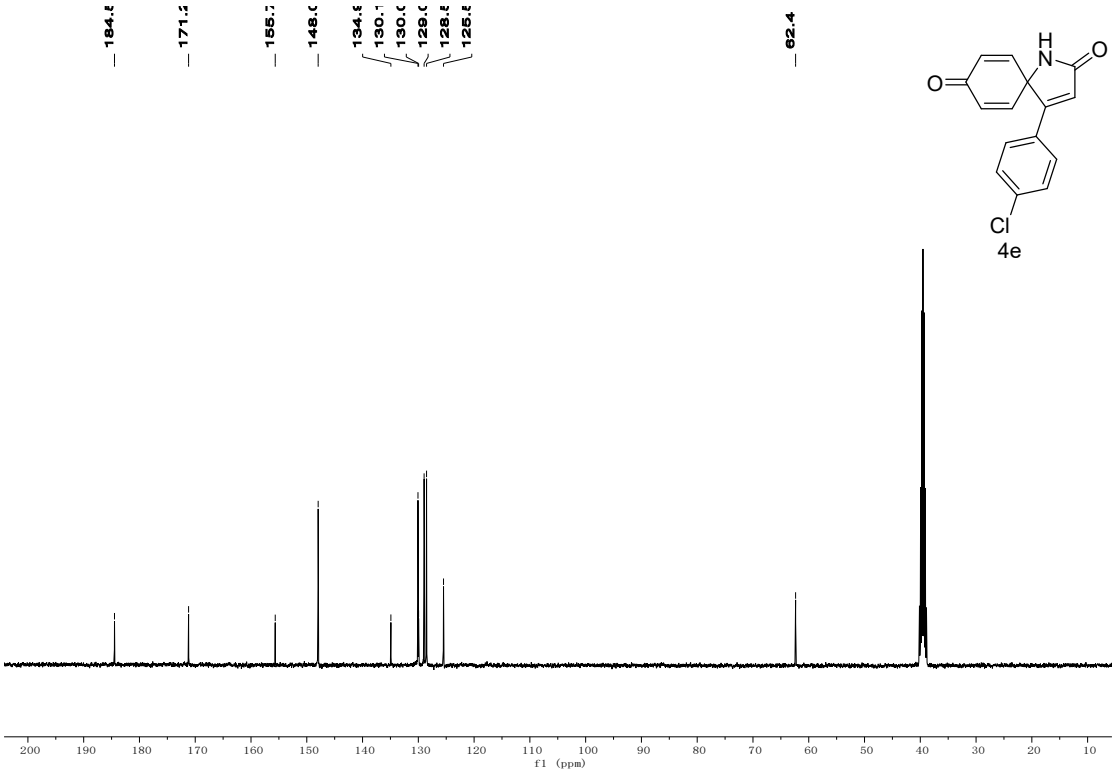
4d


0.97
2.05
2.02
2.04
1.00
2.04
2.12
3.04
10.0 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 -0.5
f1 (ppm)


184.6
172.6
159.0
147.7
146.8
131.0
128.6
128.2
126.6
122.6
63.2
28.8
15.2
H
N
O
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

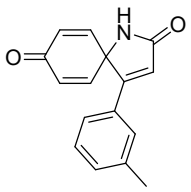
4d


8.74
7.58
7.56
7.46
7.47
6.88
6.87
6.40
6.38
H
N
O
O
Cl
4e
0.97
2.07
2.03
3.04
2.00
10.0 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 -0.5
f1 (ppm)

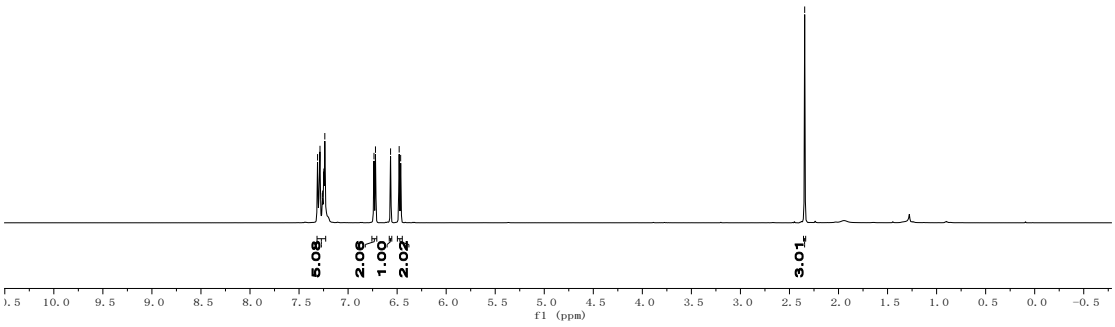

184.8
171.2
155.7
148.0
134.6
130.1
130.0
129.0
128.8
125.8
62.4
H
N
O
O
Cl
4e
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.31
7.26
7.26
7.26
7.25
7.25
7.24
6.74
6.72
6.57
6.48
6.46


2.34


H
N
O
O

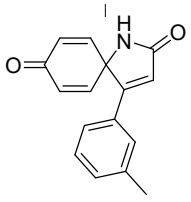
4f


5.08
2.06
1.00
2.02
3.01
f1 (ppm)

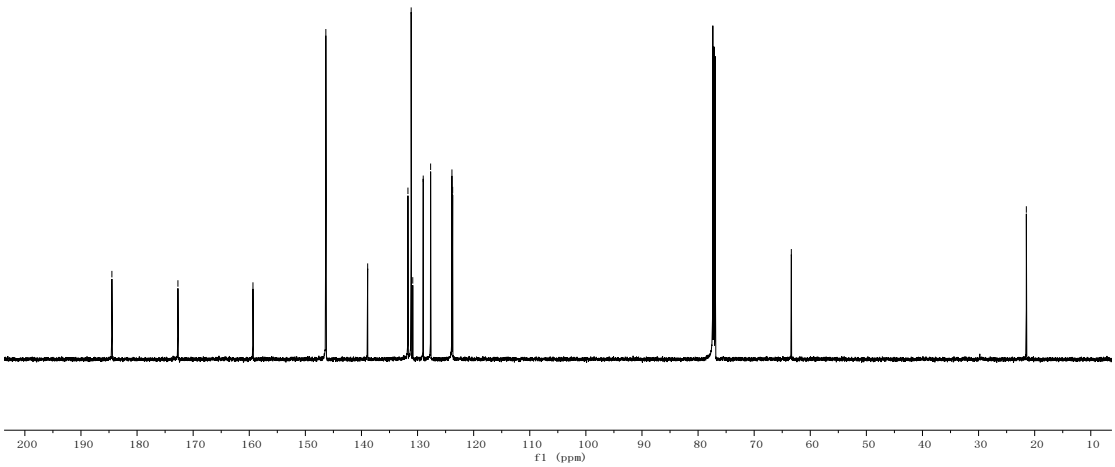

184.5
172.7
159.3
146.3
138.5
131.7
131.1
130.5
129.0
127.7
123.5
123.2

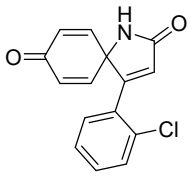

63.4


21.5

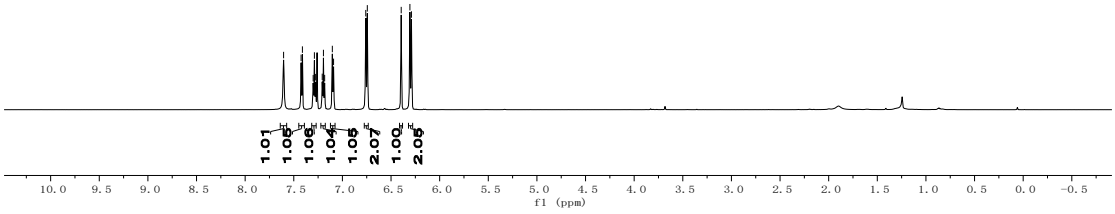

H
N
O
O

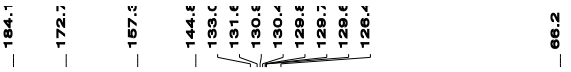
4f

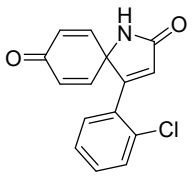

f1 (ppm)



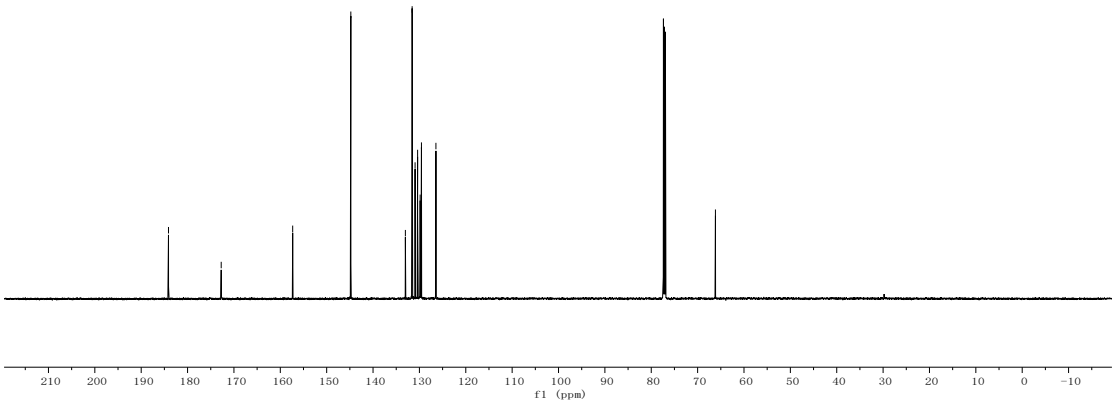
4g



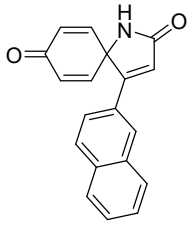




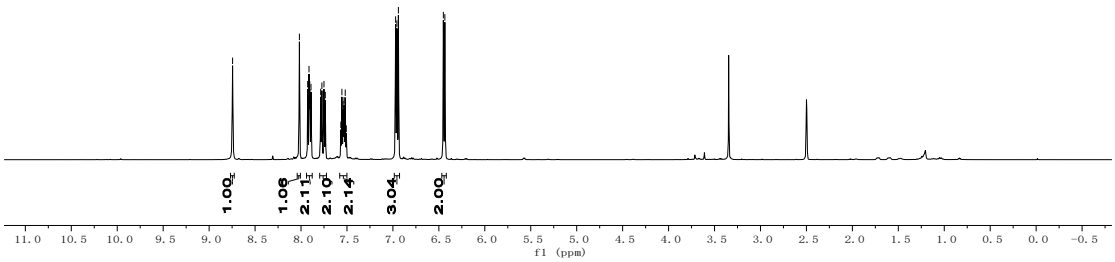
4g

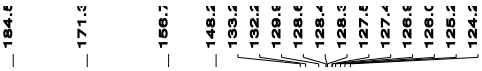


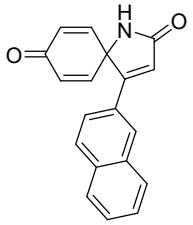

8.74
8.02
7.93
7.91
7.91
7.86
7.78
7.77
7.75
7.74
7.57
7.56
7.54
7.53
7.52
7.51
6.97
6.96
6.94
6.45
6.43


H
N
O
O

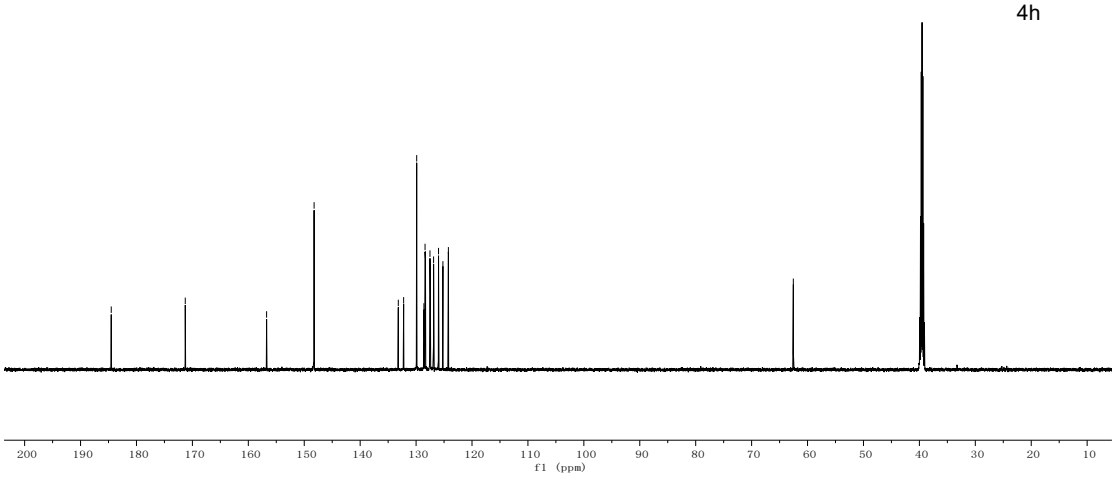
4h

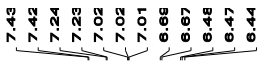

1.00
1.08
2.11
2.10
2.14
3.04
2.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)

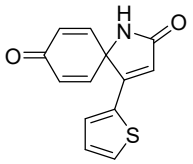

184.5
171.3
156.7
148.4
133.2
132.4
129.5
128.6
128.4
128.3
127.8
127.4
126.6
126.0
125.4
124.4
62.5


H
N
O
O

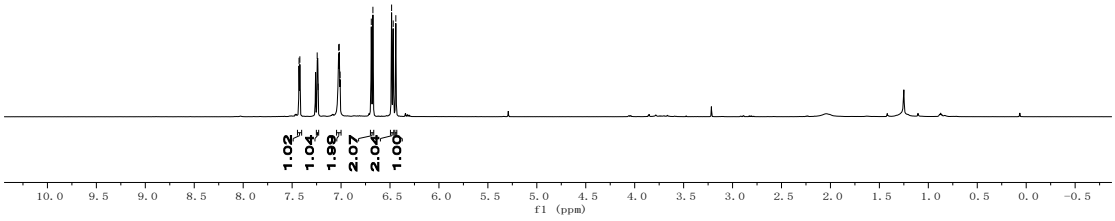
4h

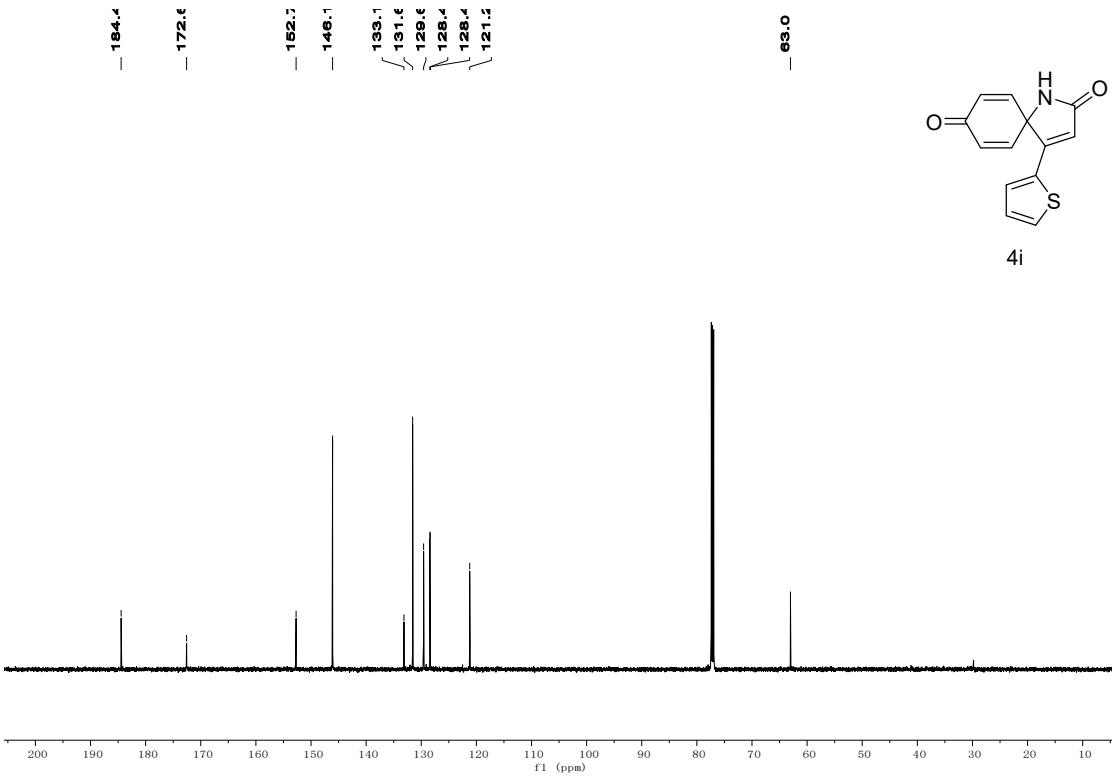

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.43
7.42
7.24
7.23
7.02
7.02
7.01
6.68
6.67
6.48
6.47
6.44


H
N
O
O
S

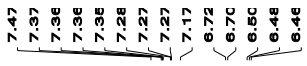
4i

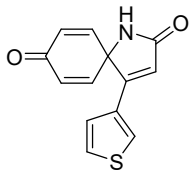

1.02
1.04
1.99
2.07
2.04
1.00
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.5
f1 (ppm)


184.4
172.6
152.7
146.1
133.1
131.6
129.6
128.4
128.4
121.5
63.0
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

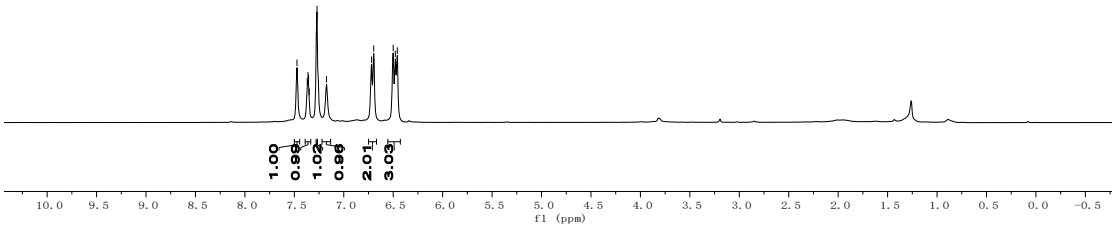
H
N
O
O
S

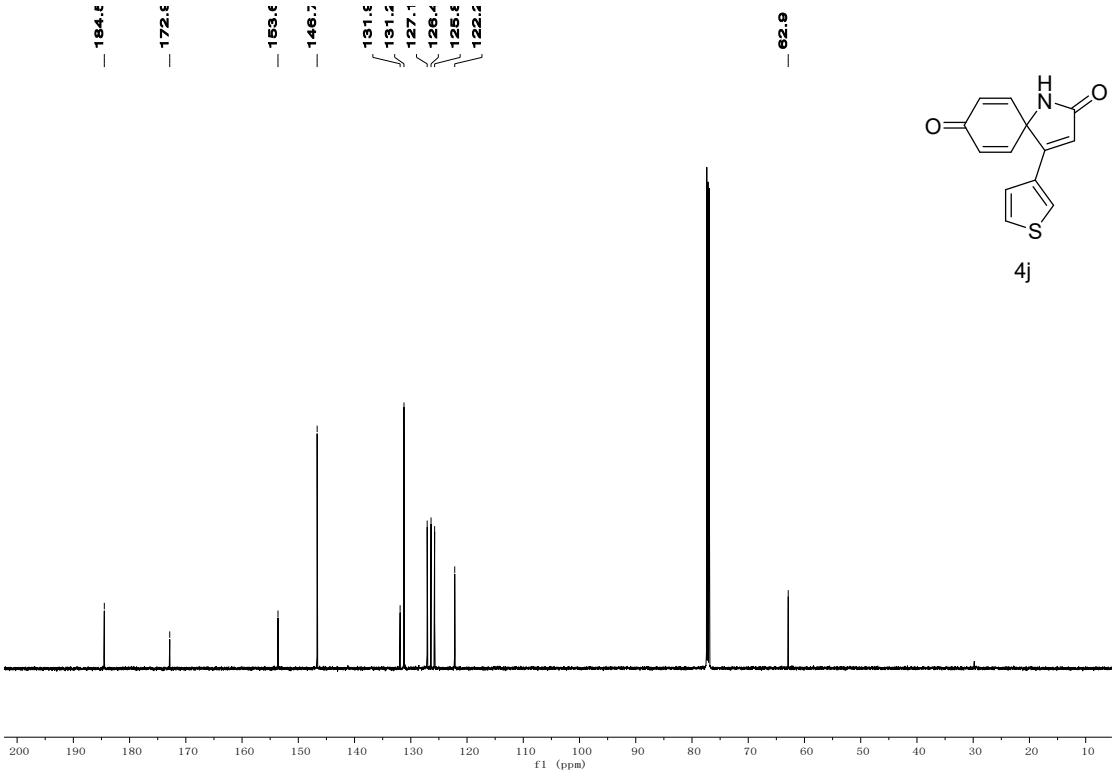
4i


7.47
7.37
7.36
7.36
7.35
7.28
7.27
7.27
7.17
6.72
6.70
6.50
6.48
6.46


H
N
O
O
S

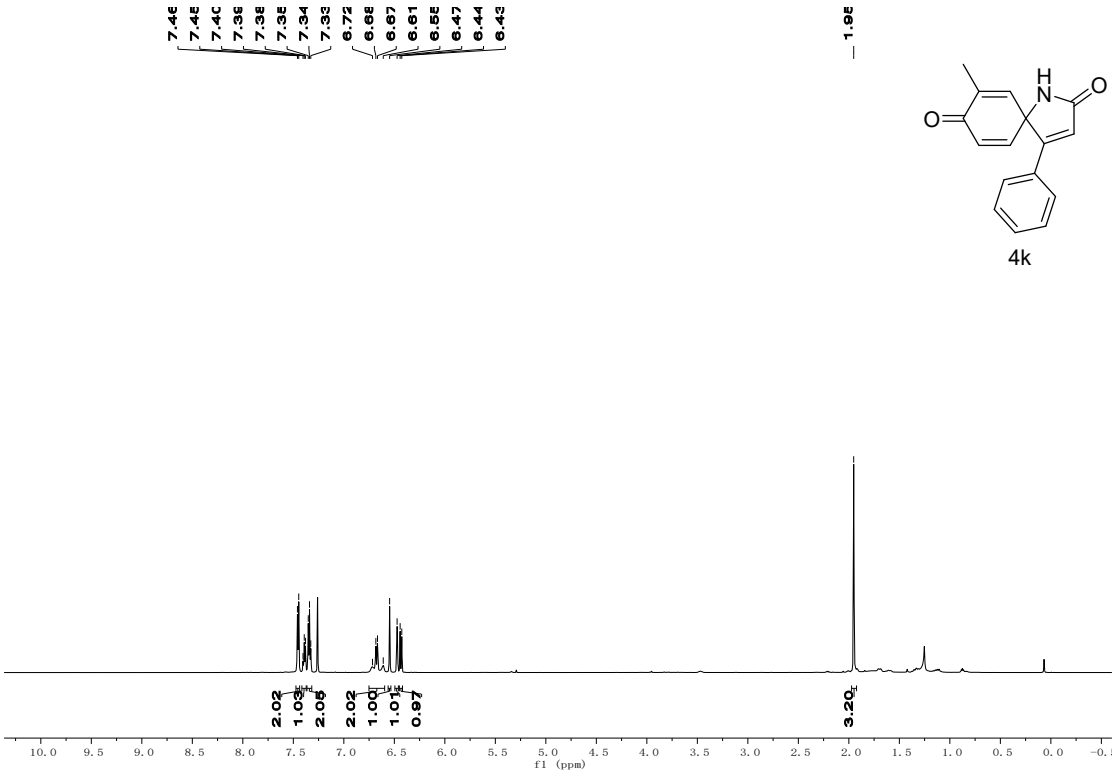
4j

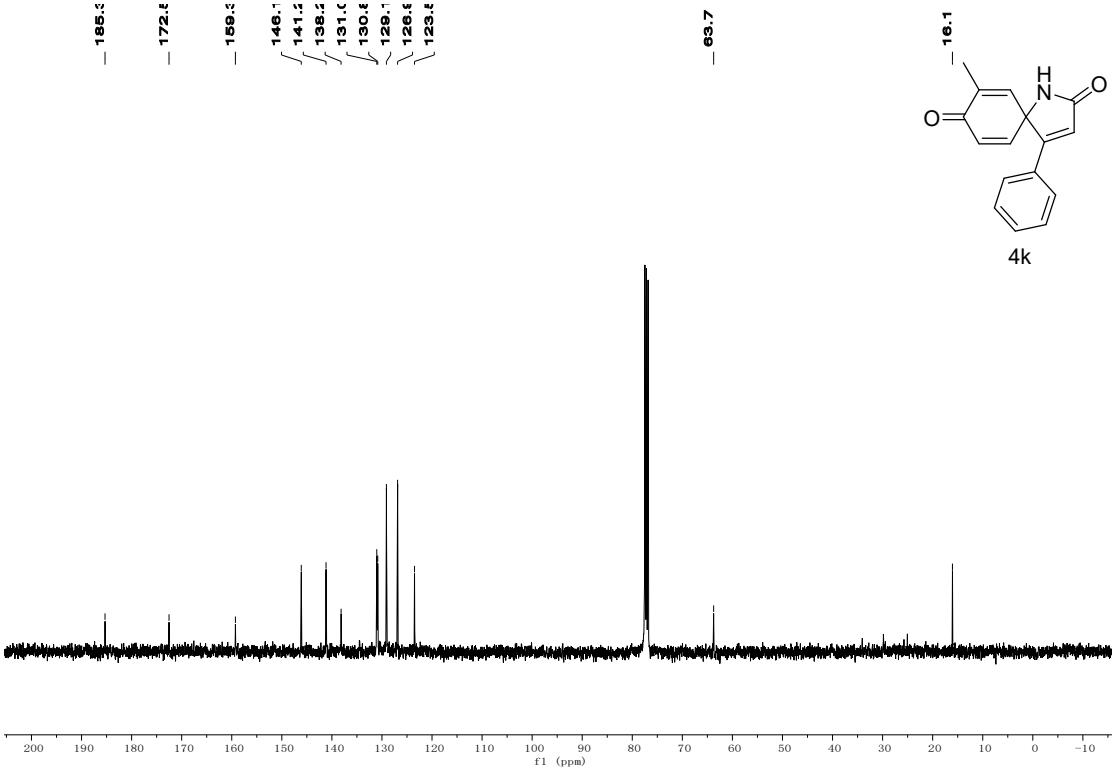

1.00
0.98
1.03
0.96
2.01
3.03
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)


184.6
172.6
153.6
146.7
131.6
131.2
127.1
126.4
125.8
122.4
62.9
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

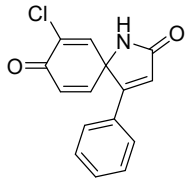
H
N
O
O
S

4j

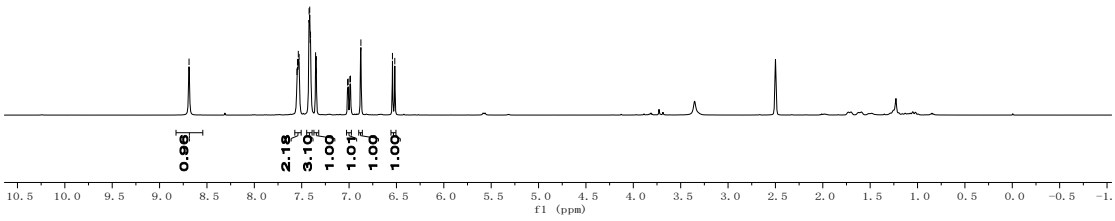

7.46
7.45
7.40
7.36
7.35
7.35
7.34
7.33
6.72
6.68
6.67
6.61
6.55
6.47
6.44
6.43
1.95
H
N
O
O
4k
2.02
1.03
2.05
2.02
1.00
1.07
0.97
3.20
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


185.3
172.8
159.3
146.1
141.2
138.2
131.0
130.8
129.1
126.8
123.8
63.7
18.1
H
N
O
O
4k
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

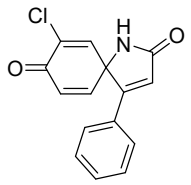

8.69
7.55
7.54
7.54
7.53
7.42
7.42
7.41
7.35
7.34
7.02
7.01
6.98
6.98
6.88
6.54
6.52


Cl
H
N
O
O

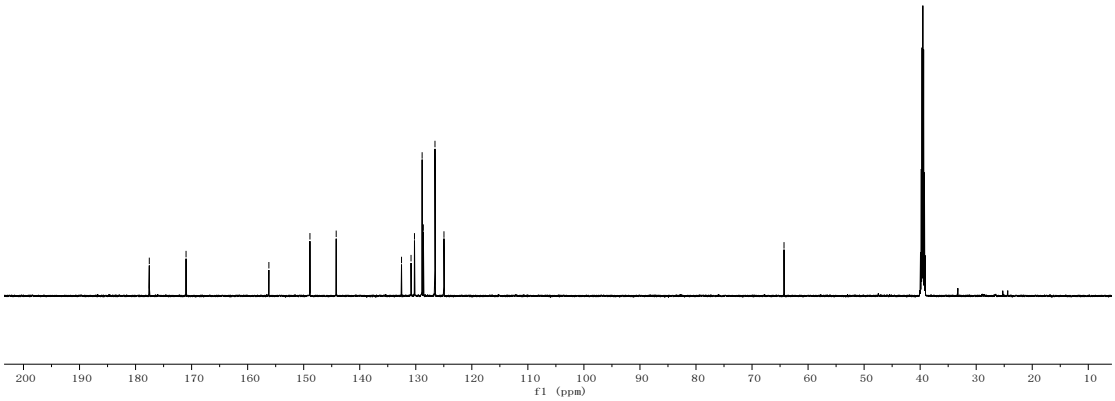
4l


0.98
2.18
3.10
1.00
1.01
1.00
1.00
10.5 10.0 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 −0.5 −1.
f1 (ppm)

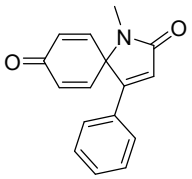

177.6
171.0
156.2
148.6
144.2
132.8
130.8
130.2
128.8
128.6
126.6
125.0
64.3


Cl
H
N
O
O

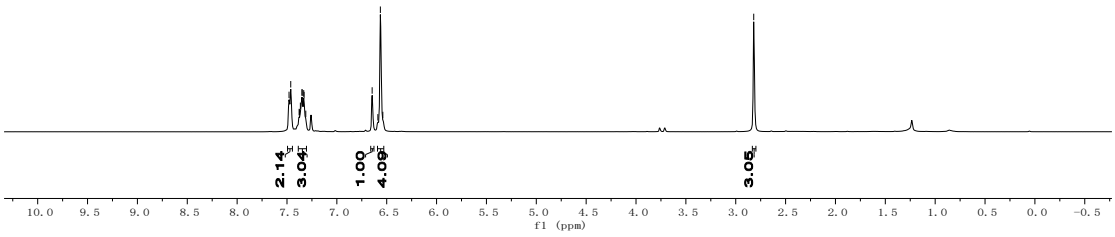
4l

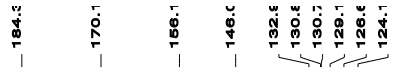

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

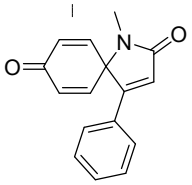

7.48
7.46
7.38
7.38
7.37
7.35
7.35
7.33
7.33
7.31
6.65
6.58
6.56
6.54
2.82



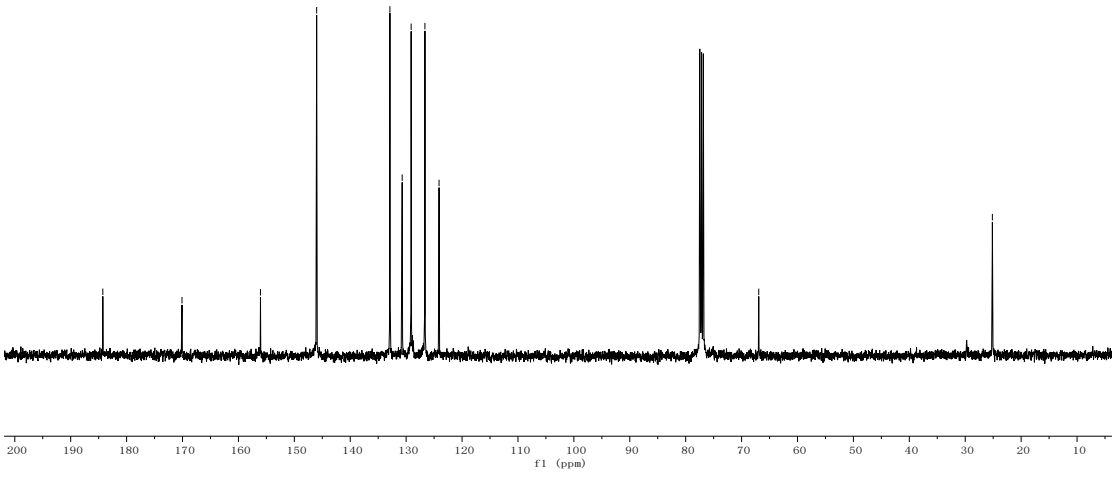
4m


2.14
3.04
1.00
4.09
3.05
10.0 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 −0.5
f1 (ppm)

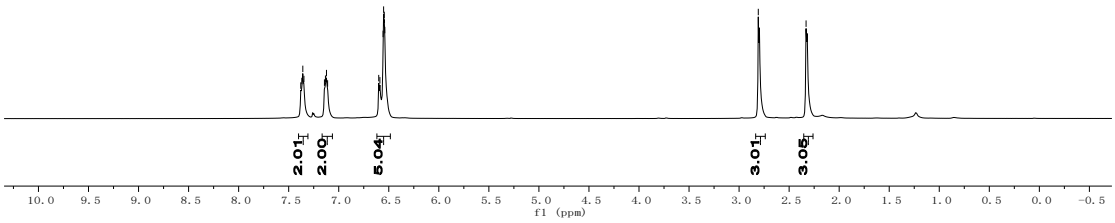

184.3
170.1
158.1
148.0
132.8
130.8
130.2
129.1
128.6
124.1
66.9
25.1

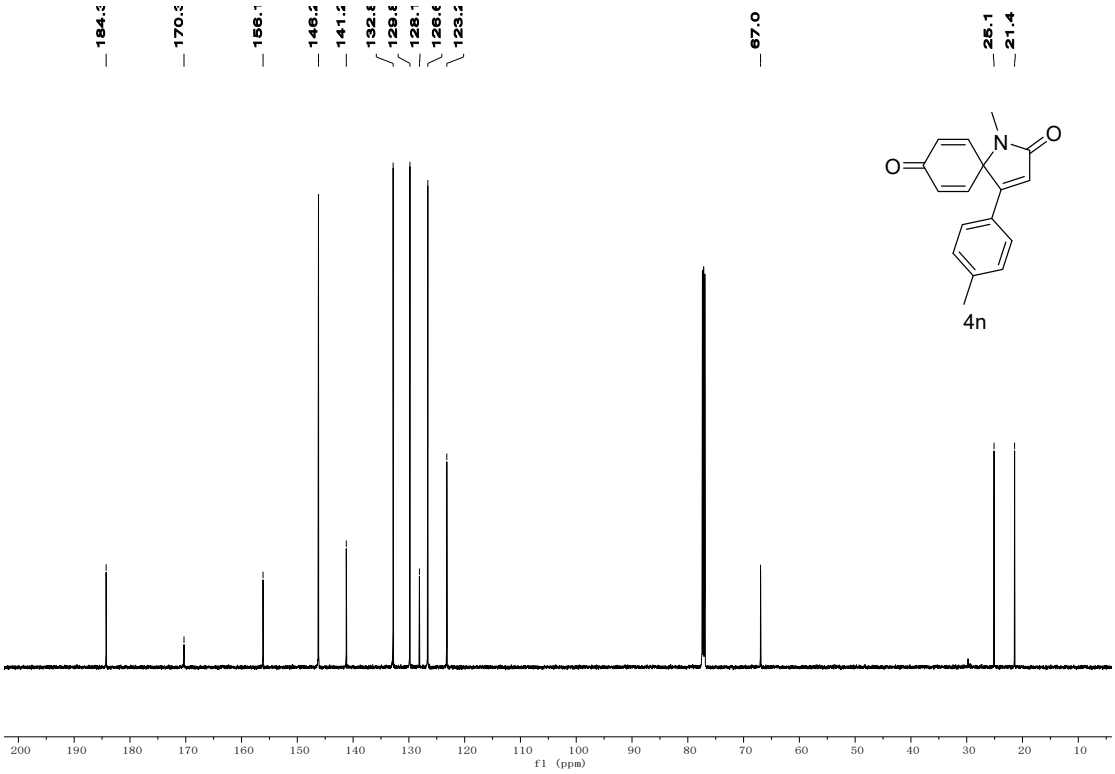


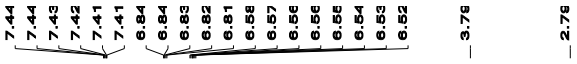
4m

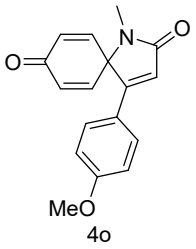

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.38
7.37
7.36
7.35
7.14
7.13
7.12
7.11
6.60
6.55
6.55
6.55
6.55
6.54
2.81
2.33
O
N
O
4n

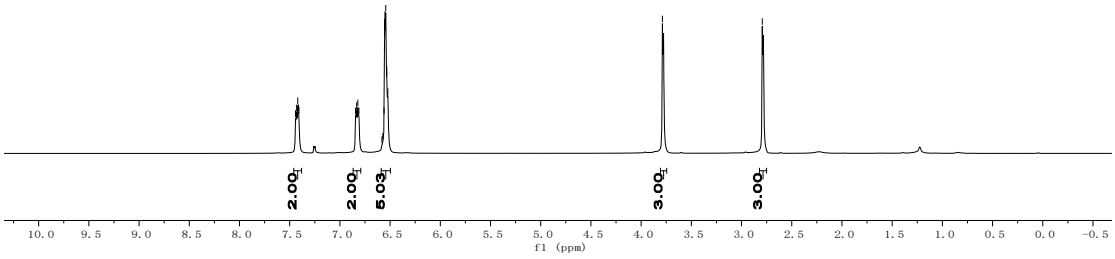

2.01
2.00
5.04
3.01
3.05
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 −0.5
f1 (ppm)

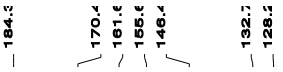

184.3
170.3
156.1
146.2
141.2
132.8
129.8
128.7
126.6
123.2
67.0
26.1
21.4
O
N
O
4n
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.44
7.44
7.43
7.42
7.41
7.41
6.84
6.84
6.83
6.82
6.81
6.58
6.57
6.56
6.56
6.55
6.54
6.53
6.52
3.76
2.76


O
N
O
MeO

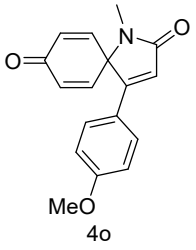
4o


2.00
2.00
5.03
3.00
3.00
10.0 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 -0.5
f1 (ppm)

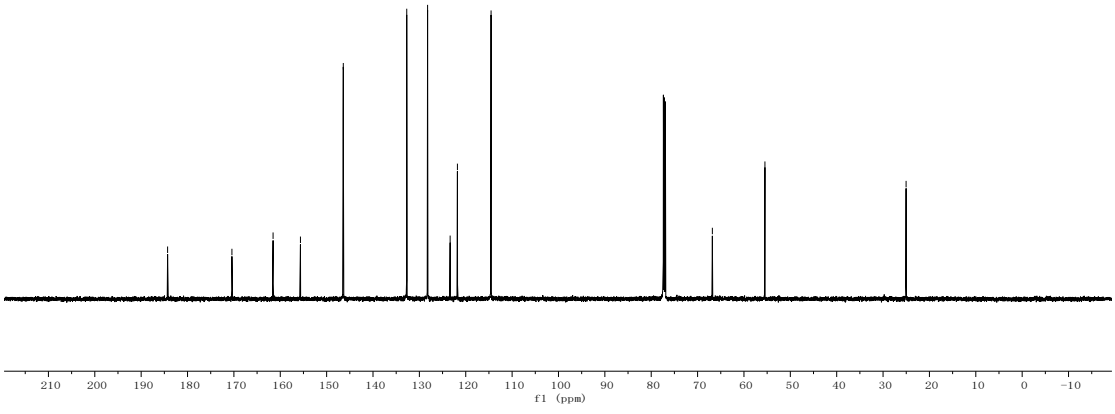

184.3
170.4
161.6
155.6
146.4
132.7
128.4
123.4
121.6
114.6

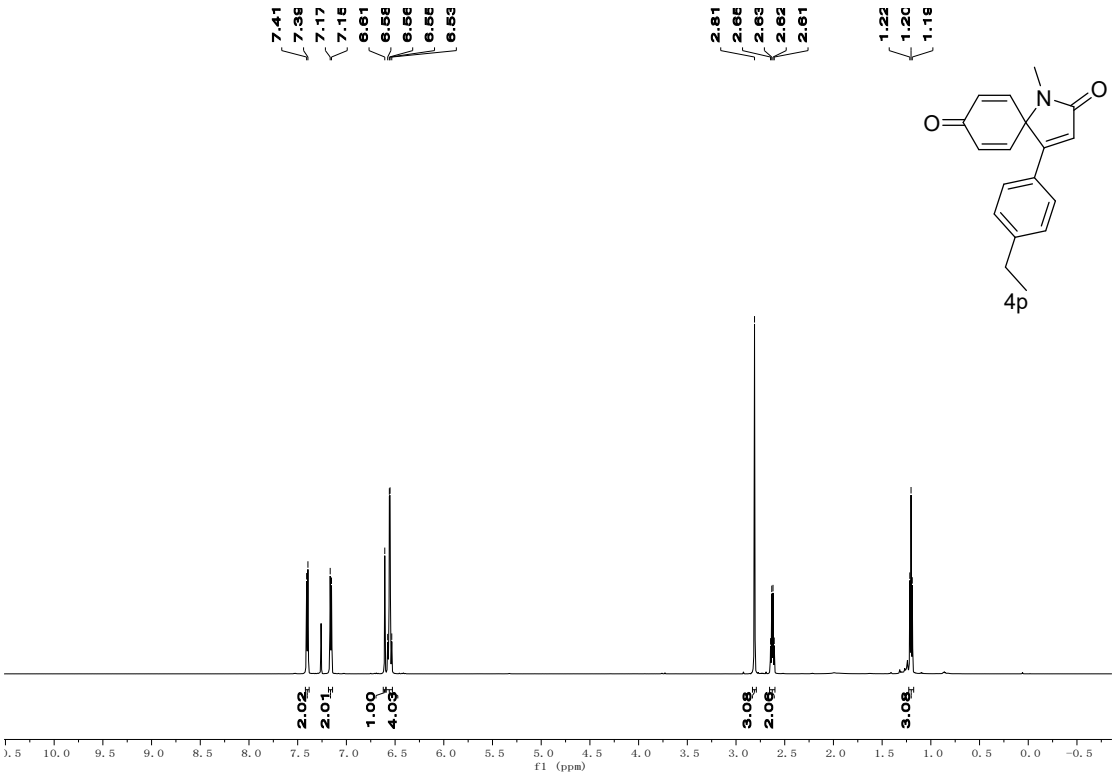

66.8
55.5

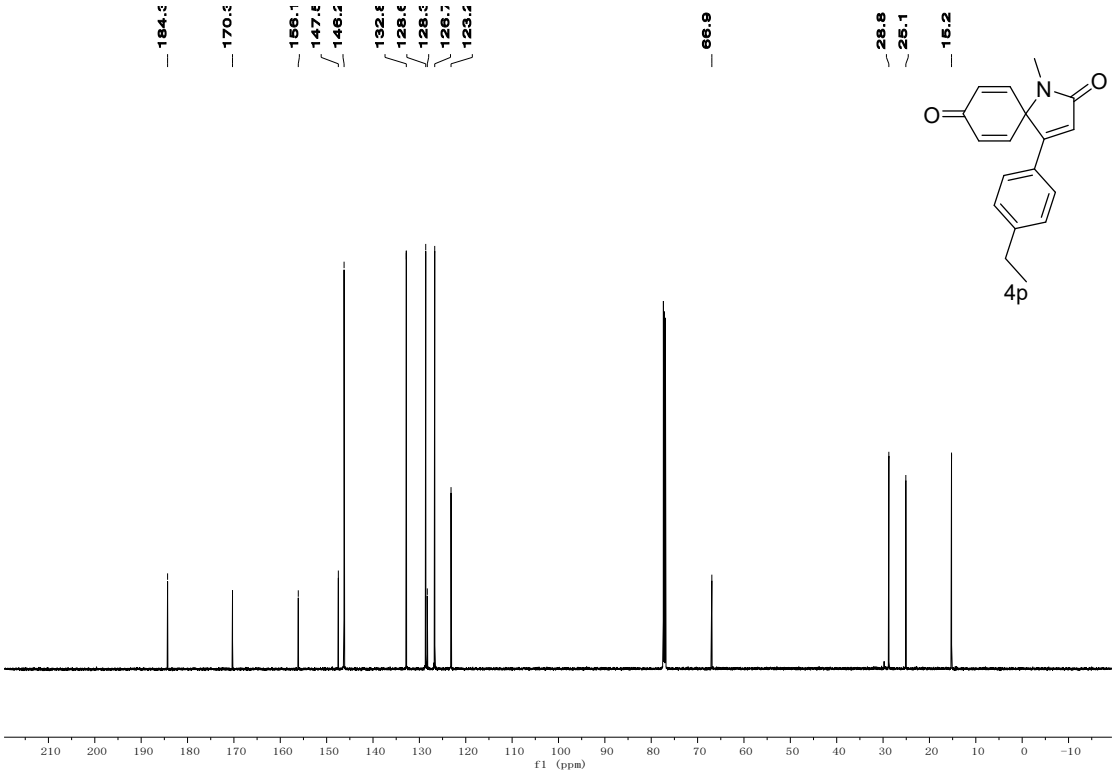

25.0

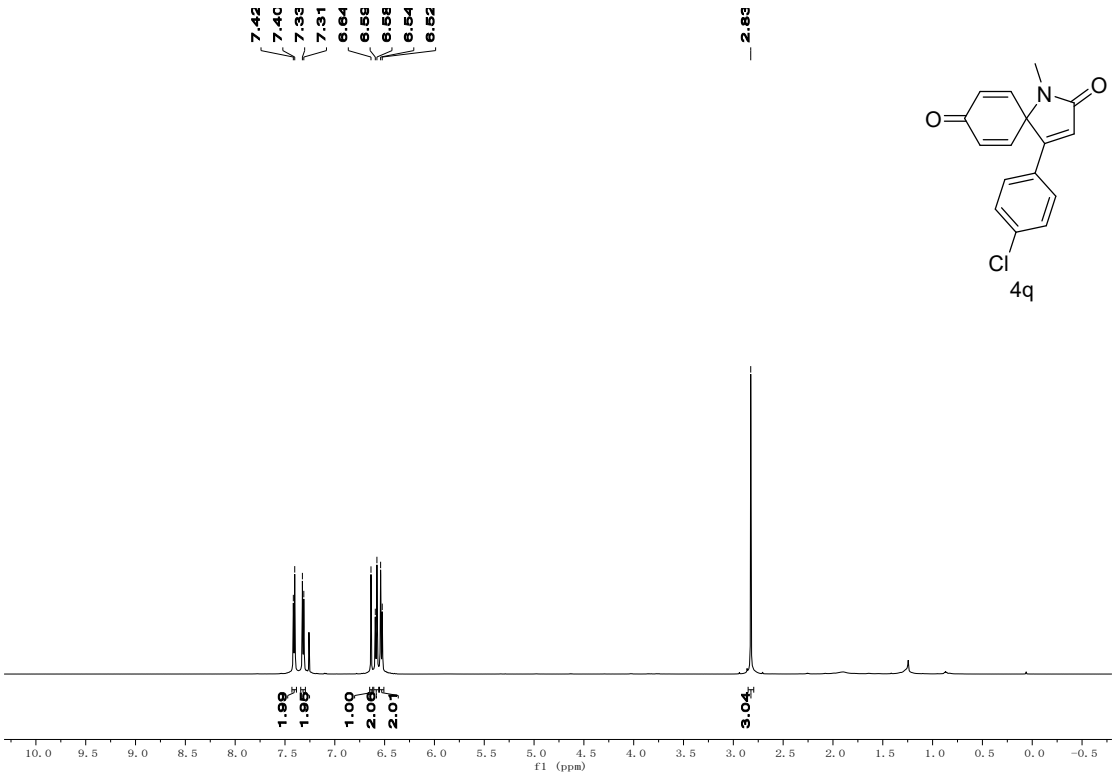

O
N
O
MeO

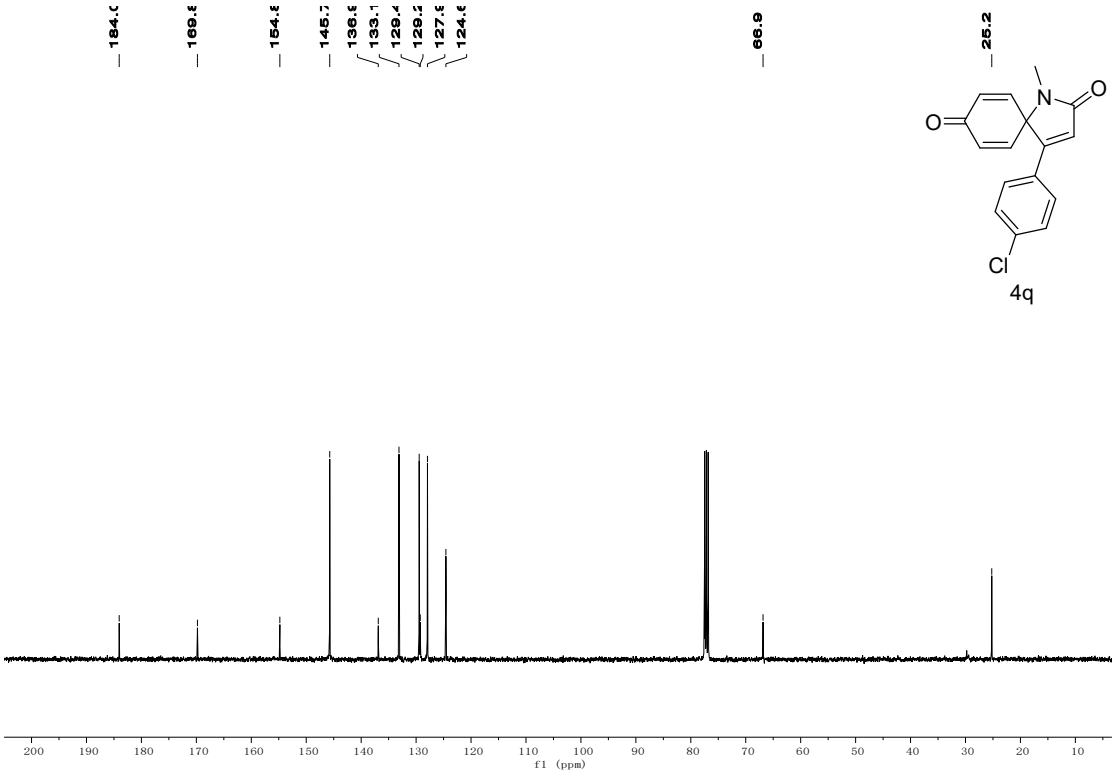
4o

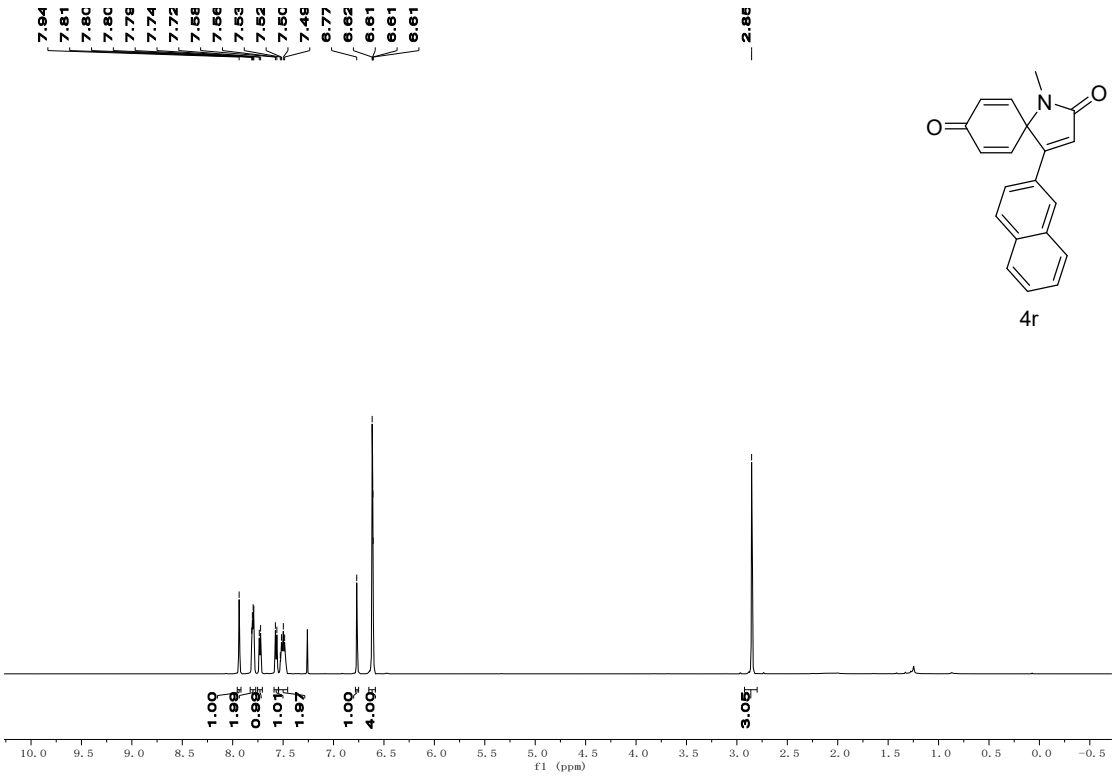

210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


7.41
7.38
7.17
7.15
6.61
6.58
6.56
6.55
6.53
2.81
2.65
2.63
2.62
2.61
1.22
1.20
1.18
O
N
O
4p
2.02
2.01
1.00
4.03
3.02
2.06
3.08
f1 (ppm)

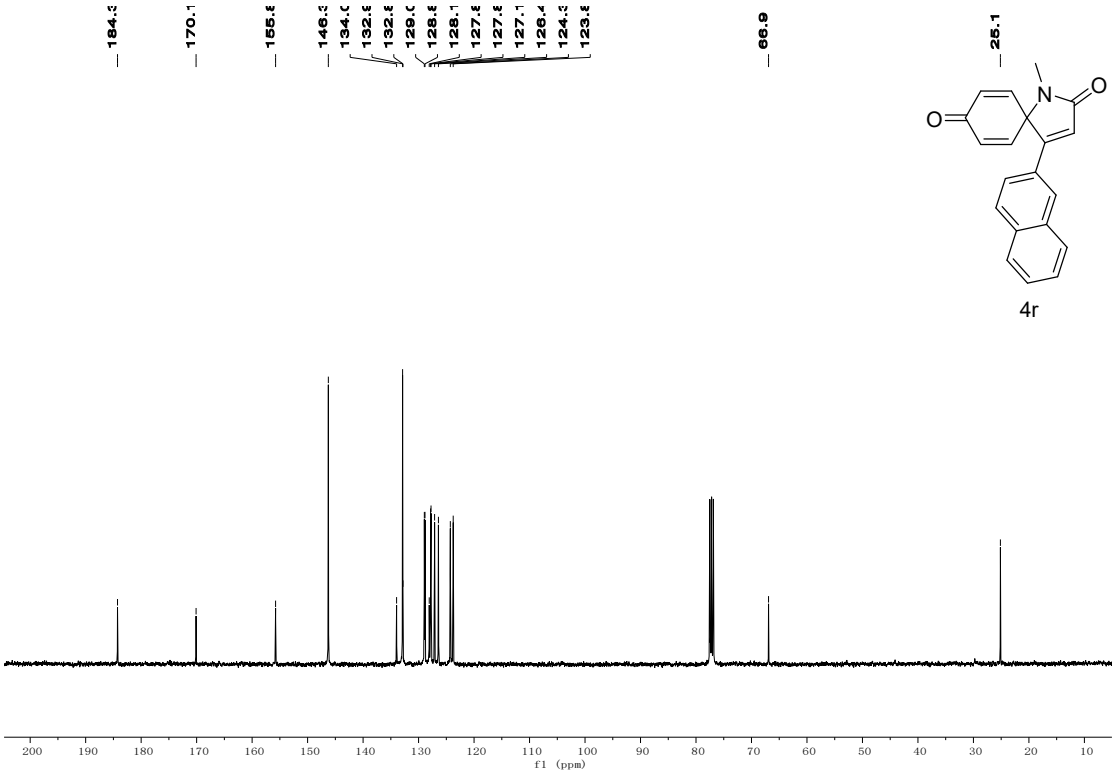

184.3
170.3
156.1
147.5
146.4
132.8
128.6
128.3
126.7
123.4
66.9
28.8
25.1
15.2
O
N
O
4p
f1 (ppm)


7.42
7.40
7.33
7.31
6.64
6.59
6.58
6.54
6.52
2.83
O
N
O
Cl
4q
1.98
1.95
1.00
2.00
2.01
3.04
10.0 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 -0.5
f1 (ppm)


184.0
169.8
154.8
145.7
136.6
133.1
129.4
128.4
127.5
124.6
66.9
25.2
O
N
O
Cl
4q
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


7.84
7.81
7.80
7.80
7.78
7.74
7.72
7.58
7.56
7.53
7.52
7.50
7.46
6.77
6.62
6.61
6.61
6.61
2.85
1.00
1.98
0.98
1.01
1.97
1.00
4.00
3.05
10.0 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 −0.5
f1 (ppm)

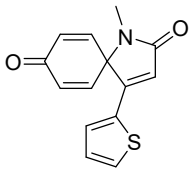
4r


184.3
170.1
155.8
146.3
134.0
132.6
132.5
129.0
128.6
128.1
127.8
127.5
127.1
126.4
124.5
123.6
66.9
25.1
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

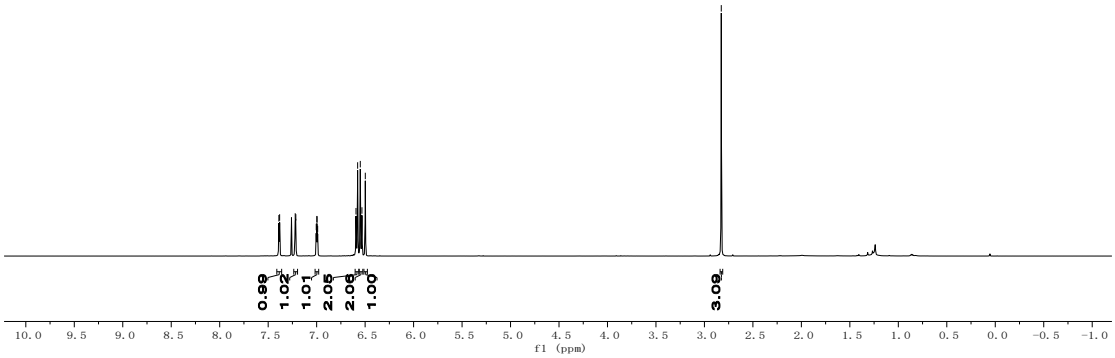
4r

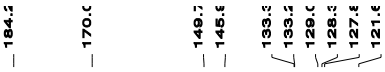

7.38
7.38
7.22
7.21
7.00
7.00
7.00
6.98
6.56
6.56
6.55
6.53
6.50

2.83


N
O
O
S

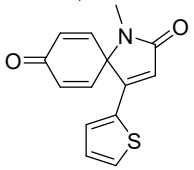
4s


0.99
1.02
1.01
2.05
2.00
1.00
3.09
10.0 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 -0.5 -1.0
f1 (ppm)

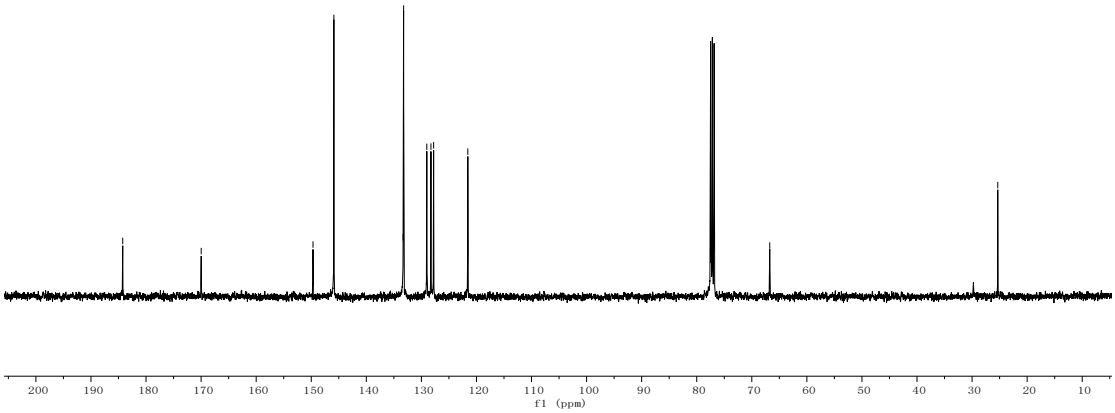

184.2
170.0
149.7
145.6
133.3
133.2
129.0
128.3
127.8
121.6

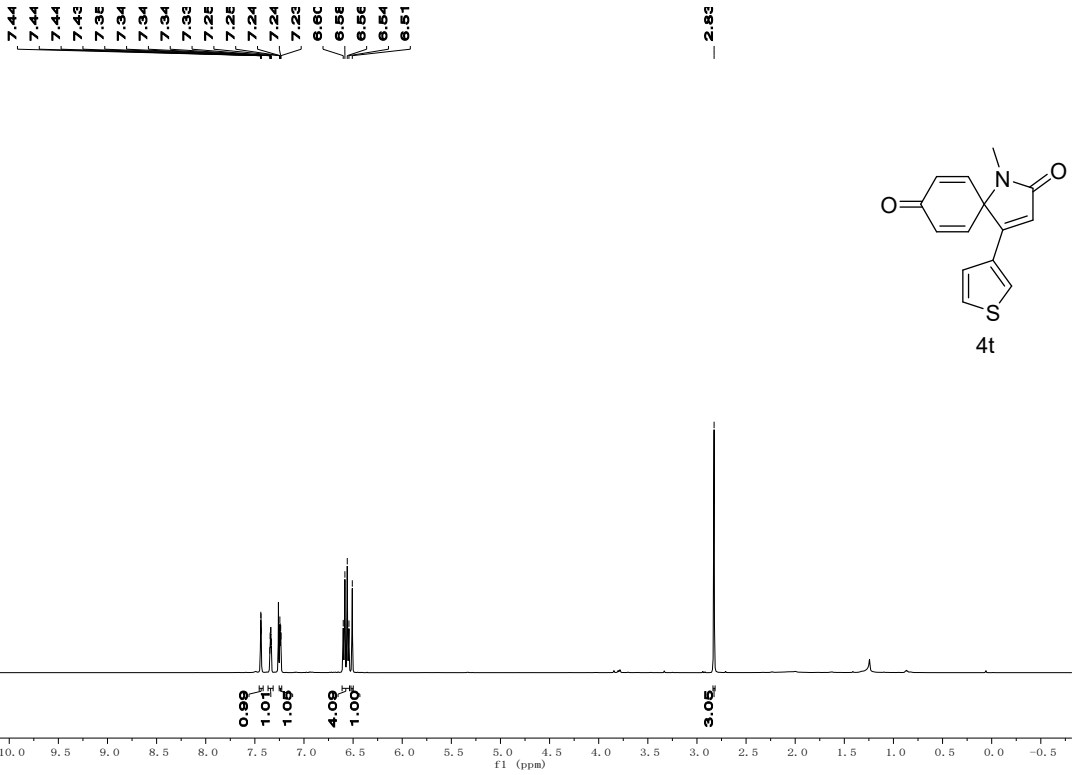

66.7

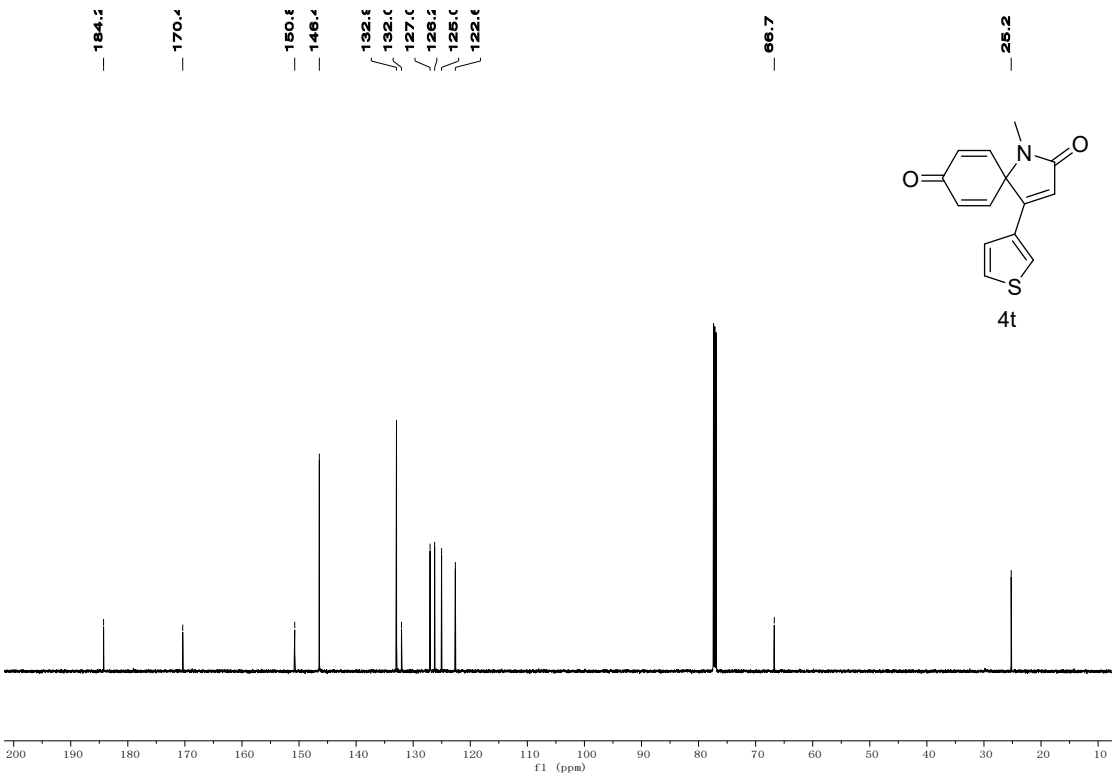
25.3


N
O
O
S

4s


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


7.44
7.44
7.44
7.43
7.35
7.34
7.34
7.34
7.33
7.25
7.25
7.24
7.24
7.23
6.60
6.58
6.56
6.54
6.51
2.83
O
N
O
S
4t
0.99
1.01
1.05
4.09
1.00
3.05
10.0 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 -0.5
f1 (ppm)


184.2
170.4
150.8
146.4
132.6
132.0
127.0
126.2
125.0
122.6
66.7
25.2
O
N
O
S
4t
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)