SUPPORTING INFORMATION

I_2/DMSO-mediated substrate selective oxidation of tetrahydro indole2,4-dione towards 4-hydroxy isatine and 5,6-dihydro-1H-indole-2,4-dione derivatives.

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The characterization data of all synthesized compounds (3a-3ad) are given below:

1-(4-methoxyphenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3a) (Table 2 Entry 1): Yellow solid, Yield: 208 mg, 81%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 145-147 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.07 (d, *J*=8.8Hz, 2H), 6.91 (d, *J*=8.8Hz, 2H), 3.76 (s, 3H), 3.31-3.30 (m, 2H), 2.37-2.32 (m, 4H), 2.03-2.02 (m, 2H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.4, 176.1, 163.2, 159.6, 128.3, 125.7,114.7, 113.2, 55.4, 36.5, 33.3, 23.0, 21.9 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_{16}NO_3]$: 258.1130; found: 258.1137.

1-(p-tolyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3b) (Table 2 Entry 2): Yellow solid, Yield: 220 mg, 91%; R_f : 0.3 (15 % Ethyl acetate in Petroleum ether); m.p. = 139-142°C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.26 (d, *J*=8Hz, 2H), 7.08 (d, *J*=8.4Hz, 2H), 3.38 (m, 2H), 2.46-2.39 (m, 4H), 2.37 (s, 3H), 2.12-2.05 (m, 2H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ =192.6, 176.1, 163.1, 139.1, 130.7, 130.3, 127.0, 113.6, 36.7, 33.6, 23.3, 22.2, 21.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_{16}NO_2]$: 242.1181; found: 242.1185.

1-(4-chlorophenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3c)(Table 2 Entry 3): Yellow solid, Yield: 241 mg, 92%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 135-138 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42 (d, *J*=8.8 Hz,2H), 7.14 (d, *J* = 8.8 Hz, 2H), 3.36 (t, *J* = 2.8 Hz, 2H), 2.44-2.38 (m, 4H), 2.11-2.07 (m, 2H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.6, 175.6, 162.1, 134.6, 131.8, 129.8, 128.4, 113.9, 36.6, 33.5, 23.2, 22.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{13}ClNO_2]$: 262.0635; found: 262.0639.

1-(4-bromophenyl)-6,7-dihydro-1H-indole-2,4(3H,5H)-dione(3d) (Table 2 Entry 4): Yellow solid, Yield: 272 mg, 89%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 145-147 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.61 (d, *J*=8.7Hz, 2H), 7.11 (d, *J*=8.4Hz, 2H), 3.41 (t,*J* = 2.7 Hz, 2H), 2.49-2.43 (m, 4H), 2.16-2.10 (m, 2H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.6, 175.6, 162.0, 133.0, 132.5, 128.8, 122.8, 114.2, 36.7, 33.7, 23.4, 22.3 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{13}BrNO_2]$: 306.0130; found: 306.0137.

1-(3-chloro-4-methoxyphenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3e) (Table 2 Entry 5): Yellow solid, Yield: 248 mg, 85%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 154-158 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.19 (d, J=2.4, 1H), 7.05 (dd, J_1=8.8Hz, J_2=2.4Hz, 1H), 6.94 (d, J=8.8Hz, 1H), 3.84 (s, 3H), 3.28 (s, 2H), 2.37-2.34 (m, 4H),2.06-2.02(m, 2H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.7, 175.8, 162.8, 155.2, 128.9, 126.8, 125.9, 122.9, 113.4, 112.2, 56.3, 36.4, 33.2, 22.9, 21.9 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_{15}ClNO_3]$: 292.0740; found: 292.0747.

1-(2-hydroxyphenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3f) (Table 2 Entry 6): Red solid, Yield: 197 mg, 81%; R_f : 0.2 (25 % Ethyl acetate in Petroleum ether); m.p. = 158-161 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.19 (t, J =7.8 Hz, 1H), 7.04 (d, J = 7.8 Hz, 1H), 6.94 (d, J = 8.4 Hz, 1H), 6.87 (t, J=7.5Hz, 1H), 5.25 (brs, 1H), 3.32 (s, 2H), 2.45-2.30 (m, 4H), 2.09-2.00 (m, 2H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 194.0, 176.9, 171.5, 166.4, 153.0, 130.8, 129.2, 120.3, 117.5, 113.2,36.3, 33.4, 22.6, 21.8 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{14}NO_3]$: 244.0974; found: 244.0979.

1-(3-bromophenyl)-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3g) (Table 2 Entry 7): Yellow solid, Yield: 263 mg, 86%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 146-148 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.56 (d,J = 7.8 Hz, 1H), 7.42-7.34 (m, 2H), 7.19 (d, J = 8.4 Hz, 1H) 3.42-3.40 (m, 2H), 2.50-2.43 (m, 4H), 2.17-2.11(m, 2H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.7, 175.5, 162.0, 134.8, 132.1, 131.0, 130.4, 126.0, 123.1, 114.2, 36.7, 33.7, 23.4, 22.3 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{13}BrNO_2]$: 306.0130; found: 306.0138.

1-(3-nitrophenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3h)(Table 2 Entry 8): Yellow solid, Yield: 231 mg, 85%; R_f : 0.2 (25 % Ethyl acetate in Petroleum ether); m.p. = 156-158 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.29-8.26 (m, 1H), 7.72-7.68 (m, 2H), 7.65-7.62 (m, 1H), 3.46 (s, 2H), 2.55-2.50(m, 4H), 2.18-2.14(m, 2H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.9, 175.3, 161.3, 143.6, 139.7, 133.2, 130.7, 122.2, 119.9, 114.7, 36.6, 33.6, 23.5, 22.3 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{13}N_2O_4]$: 273.0875; found: 273.0879.

1-(benzo[d][1,3]dioxol-4-yl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3i)(Table 2 Entry 9): Black solid, Yield: 222 mg, 82%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 158-160 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 6.85 (d, J=8.1, 1H), 6.67-6.62 (m, 2H), 6.01 (s, 2H), 3.37-3.35 (m, 2H), 2.45-2.38 (m, 4H), 2.13-2.04 (m, 2H) ppm.^{13}C {^{1}H} NMR (75

MHz, $CDCl_3$) d: δ = 192.6, 176.2, 163.1, 148.5, 148.1, 126.9, 121.0, 113.5, 108.7, 108.4, 102.1, 36.6, 33.5, 23.2, 22.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_{14}NO_4]$: 272.0923; found: 272.0929.

1-(3-methoxyphenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3j) (Table 2 Entry 10): Yellow solid,Yield: 226 mg, 88%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 145-147 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.34 (t, *J*=8.1Hz, 1H), 6.92 (dd, J_1=8.4Hz, J_2=1.5Hz, 1H), 6.77-6.72 (m, 2H), 3.78 (s, 3H), 3.37-3.35 (m, 2H), 2.44-2.39 (m, 4H), 2.11-2.02 (m, 2H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.6, 175.7, 162.8, 160.4,134.3, 130.3, 119.2, 114.4, 113.6, 113.1, 55.5, 36.6, 33.5, 23.2, 22.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_{16}NO_3]$: 258.1130; found: 258.1139.

1-(2-iodophenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3k) (Table 2 Entry 11): Yellow solid, Yield: 283 mg, 80%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 141-143 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.95-7.93 (m, 1H), 7.47 (t, *J*=7.6Hz, 1H), 7.27-7.24 (m, 1H), 7.18 (t, *J*=7.6Hz, 1H), 3.42 (s, 2H), 2.46 (t, *J*=3.2Hz,2H), 2.24 (t, *J*=3.2Hz, 2H), 2.13-2.10 (m, 2H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.7, 175.3, 162.5, 140.1, 136.7, 131.4, 129.9, 129.8, 114.0, 99.3, 36.7, 33.7, 23.1, 22.0 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{13}NO_2\,I]$: 353.9991; found: 353.9999.

1-cyclohexyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3l) (Table 2 Entry 12): Black solid, Yield: 210mg, 90%; R_f : 0.3 (15 % Ethyl acetate in Petroleum ether); m.p. = 126-128 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.69-3.61 (m, 1H), 3.09-3.07 (m, 2H), 2.57 (t, *J*=3Hz,2H), 2.31-2.26 (m, 2H), 2.05 (t, *J*=6.3Hz, 2H), 1.98-1.84 (m, 2H), 1.78-1.72 (m, 2H), 1.64-1.56 (m, 3H) 1.28-1.06 (m, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.1, 176.7, 163.4, 113.2, 53.6, 35.9, 33.7, 30.3, 25.9, 24.9, 23.3, 22.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_{20}NO_2]$: 234.1494; found: 234.1499.

1-(4-chlorophenyl)-6-phenyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3m) (Table 2 Entry 13): Yellow solid, Yield: 304 mg, 90%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.41 (d,*J* = 8.4 Hz, 2H), 7.33-7.29 (m, 2H), 7.24-7.19 (m, 3H), 7.16 (d, *J*=8.4Hz, 2H), 3.51-3.44 (m, 3H), 2.75-2.67 (m, 3H), 2.59-2.53 (m, 1H) ppm ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 191.6, 175.6, 161.3, 142.0, 134.8, 131.7, 130.0, 129.0, 128.4, 127.5, 126.7, 113.7, 43.7, 40.9, 33.6, 31.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{20}H_{17}ClNO_2]$: 338.0948; found: 338.0956.

1-(3-bromophenyl)-6-phenyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3n) (Table 2 Entry 14): Yellow solid, Yield: 339 mg, 89%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 150-152 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.55-7.52 (m, 1H), 7.42-7.41 (m,1H), 7.36-7.32 (m, 3H), 7.29-7.27 (m, 1H), 7.24-7.22 (m, 2H), 7.18-7.16 (m, 1H), 3.55-3.49 (m, 1H), 3.47-3.46 (m, 2H), 2.77-2.69 (m, 3H), 2.61-2.56 (m, 1H) ppm ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 191.5, 175.5, 161.0, 142.1, 134.5, 132.2, 131.0, 130.4, 129.1, 127.6, 126.8, 125.9, 123.1, 113.9, 43.8, 41.0, 33.6, 31.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{20}H_{17}BrNO_2]$: 382.0443; found: 382.0449.

1-(3-nitrophenyl)-6-phenyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3o) (Table 2 Entry 15): Yellow solid, Yield: 296 mg, 85%; R_f : 0.2 (25 % Ethyl acetate in Petroleum ether); m.p. = 167-168 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.25 (d,*J*=8.4Hz, 1H), 8.13 (t, *J* = 2 Hz, 1H), 7.67 (t, *J* = 8 Hz, 1H), 7.62-7.59 (m, 1H), 7.35-7.31 (m, 3H), 7.28-7.22 (m, 2H), 3.52-3.50 (m, 2H), 2.81-2.71 (m, 4H), 2.62-2.57 (m, 1H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 191.8, 175.3, 160.3, 148.9, 141.8, 134.4, 133.1, 130.7, 129.1, 127.7, 126.8, 123.6, 122.3, 114.4,43.7, 41.0, 33.6, 31.4 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{20}H_{17}N_2O_4]$: 349.1188; found: 349.1193.

1-(2-iodophenyl)-6-phenyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3p) (Table 2 Entry 16): Yellow solid, Yield: 339 mg, 79%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 165-168 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.95-7.91 (m, 1H), 7.48-7.43 (m, 1H), 7.35-7.27 (m, 3H), 7.27-7.16(m, 4H), 3.62-3.57 (m, 1H), 3.50-3.48 (m, 2H), 2.79-2.69 (m, 2H), 2.58-2.34 (m, 2H) ppm.^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 191.6, 175.2, 161.7, 142.2, 140.1, 136.5, 131.4, 130.0, 129.9, 129.0, 127.4, 126.8, 113.8, 99.3, 43.9, 40.9, 33.7, 31.1ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{20}H_{17}INO_2]$: 430.0304; found: 430.0309.

6-phenyl-1-(o-tolyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3q)(Table 2 Entry 17): Yellow solid, Yield: 244mg, 77%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 155-157 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ =7.34-7.31 (m, 5H), 7.22-7.20 (m, 4H), 3.51-3.49 (m, 4H), 2.77-2.72(m, 3H), 2.21 (s, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 191.7, 162.8, 142.1, 136.4, 131.6, 131.5, 129.9, 129.8, 128.9, 128.1, 127.4, 127.3, 126.8, 113.5, 43.9, 41.0, 33.5, 30.8, 17.9 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{21}H_{20}NO_2]$: 318.1494; found: 318.1498.

1-(2-methoxyphenyl)-6-methyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3r) (Table 2 Entry 18): Yellow solid, Yield: 211 mg, 78%; R_f : 0.3 (25 % Ethyl acetate in Petroleum

ether); m.p. = 148-150 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.45-7.40 (m, 1H), 7.22-7.14 (m, 1H), 7.08-7.02 (m,2H), 3.82 (s, 2H), 3.42-3.39 (m, 2H), 2.47-2.13 (m, 4H), 1.07(d, *J*=6.3Hz, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.8, 176.4, 164.3, 155.3, 131.0, 130.9, 130.0, 129.7, 121.2, 112.2, 55.8, 45.2, 33.5, 30.8, 30.3, 21.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{18}NO_3]$: 272.1287; found: 272.1294.

1-(4-methoxyphenyl)-6-methyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3s) (Table 2 Entry 19): Yellow solid, Yield: 225 mg, 83%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 145-147 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.12 (d, *J*=9Hz, 2H), 6.97 (d, *J*=9Hz, 2H), 3.82 (s, 3H), 3.37 (s, 2H), 2.50-2.44 (m, 1H), 2.39-2.33 (m, 2H), 2.18-2.13 (m, 2H), 1.07(d, *J*=6Hz, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.5, 176.4, 162.8, 159.8, 128.5, 126.0, 114.9, 113.1, 55.6, 45.1, 33.5, 31.2, 30.3, 21.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{18}NO_3]$: 272.1287; found: 272.1295.

6-methyl-1-(3-nitrophenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3t)(Table 2 Entry 20): Yellow solid, Yield: 235mg, 82%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 158-160 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.25 (d, *J*=8.4Hz, 1H), 8.11 (t, *J*=2.1Hz, 1H), 7.69 (t,*J*=8.1Hz, 1H), 7.62-7.59 (m,1H), 3.41 (s, 2H), 2.54-2.21 (m, 5H), 1.10 (d, *J*=5.7Hz, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.7, 175.4, 160.7, 148.8, 134.6, 133.2, 130.6, 123.4, 122.2, 114.2, 45.0, 33.6, 31.4, 30.5, 21.2 ppm. HRMS (ESI/TOF-Q) m/z:$[M+H]^+$calcd for $[C_{15}H_{15}N_2O_4]$: 287.1032 ; found: 287.1038.

6,6-dimethyl-1-(p-tolyl)-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3u) (Table 2 Entry 21): Yellow solid, Yield: 245 mg, 91%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 157-158 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.30 (d,*J* = 8.1 Hz, 2H), 7.08 (d,*J* = 8.1 Hz, 2H), 3.44-3.42 (m, 2H), 2.41 (s, 3H), 2.34 (s, 2H), 2.28-2.26 (m, 2H), 1.10 (s, 6H)ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d:δ = 192.2, 176.4, 161.6, 139.2, 130.8, 130.4, 127.2, 112.3, 50.9, 37.2, 34.6, 33.6, 28.7, 21.3 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{17}H_{20}NO_2]$: 270.1494; found: 270.1499.

1-(4-bromophenyl)-6,6-dimethyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3v) (Table 2 Entry 22): Yellow solid, Yield: 290 mg, 87%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.63 (d, *J* = 8.4 Hz, 2H), 7.10 (d, *J* = 8.7 Hz, 2H), 3.44-3.42 (m, 2H), 2.35 (s, 2H), 2.28 (t, *J*=2.7Hz, 2H), 1.11 (s, 6H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.3, 175.8, 160.6, 133.0, 131.0, 128.8, 122.9,

112.8, 50.9, 37.3, 34.7, 33.6, 28.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{17}BrNO_2]$: 334.0437; found: 334.0448.

1-(4-methoxyphenyl)-6,6-dimethyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3w) (Table 2 Entry 23): Yellow solid, Yield: 256 mg, 90%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 153-155 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.05 (d, *J*=7.2Hz, 2H), 6.92 (d, *J*=7.2Hz, 2H), 3.75 (s, 3H), 3.2 (t, *J*=2.4Hz, 2H), 2.25 (t, *J*=2Hz, 2H), 2.20 (s, 2H), 1.02(s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.0, 176.2, 161.7, 159.6, 128.3, 125.7, 114.8, 111.8, 55.4, 50.6, 36.8, 34.2, 33.2, 28.5 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{17}H_{20}NO_3]$: 286.1443; found: 286.1449 .

1-(3-chloro-4-methoxyphenyl)-6,6-dimethyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3x) (Table 2 Entry 24): Yellow solid, Yield: 284 mg, 89%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.24 (d, *J* = 2.7 Hz, 1H), 7.09 (dd, J_1=8.7Hz, J_2=2.4Hz, 1H), 7.01 (d, *J*=8.7Hz, 1H), 3.93 (s, 3H), 3.41-3.39 (m, 2H), 2.33 (s, 2H), 2.26 (t, *J* = 3 Hz, 2H), 1.10 (s, 6H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.3, 176.2, 161.2, 155.6, 129.3, 127.0, 126.3, 123.5, 112.5, 112.4, 56.6, 50.8, 37.1, 34.6, 33.4, 28.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{17}H_{19}ClNO_3]$: 320.1053; found: 320.1059.

1-(3,4-dimethoxyphenyl)-6,6-dimethyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3y) (Table 2 Entry 25): Red solid, Yield: 290 mg, 92%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 6.90 (d, *J*=8.4, 1H), 6.72-6.67 (m, 2H), 3.87 (s, 3H), 3.84 (s, 3H), 3.37 (s, 2H), 2.29(s, 2H), 2.23 (s, 2H), 1.06 (s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.2, 176.4, 161.8, 149.6, 149.5, 126.0, 119.7, 112.1, 111.4, 110.7, 56.1, 56.1, 50.8, 36.9, 34.4, 33.4, 28.6 ppmHRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{18}H_{22}NO_4]$: 316.1549; found: 316.1558.

1-(3-chlorophenyl)-6,6-dimethyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3z) (Table 2 Entry 26): Yellow solid, Yield: 243 mg, 84%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.43-7.41 (m, 2H), 7.24-7.23 (m, 1H), 7.12-7.09(m, 1H),3.43-3.41 (m, 2H), 2.34 (s, 2H), 2.29 (t, *J*=3Hz, 2H), 1.10(s, 6H) ppm.^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 192.3, 175.7, 160.4, 135.4, 134.6, 130.7, 129.2, 127.6, 125.5, 112.8, 50.9, 37.2, 34.7, 33.5, 28.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{17}ClNO_2]$: 290.0948; found: 290.0956.

6,6-dimethyl-1-(4-nitrophenyl)-3,5,6,7-tetrahydro-1H-indole-2,4-dione(3aa)(Table 2 Entry 27): Yellow solid, Yield: 267 mg, 89%; R_f : 0.2 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.37 (d, *J*=8.8Hz, 2H), 7.44 (d, *J* = 8.8 Hz, 2H), 3.47 (s, 2H), 2.38-2.37 (m, 4H), 1.12 (s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.4, 175.3, 159.2, 147.1, 139.1, 127.6, 126.5, 125.1, 50.8, 37.6, 35.0, 33.6, 28.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{17}N_2O_4]$: 301.1188; found: 301.1191.

6,6-dimethyl-1-(o-tolyl)-6,7-dihydro-1H-indole-2,4(3H,5H)-dione (3ab) (Table 2 Entry 28): Yellow solid, Yield: 218 mg, 81%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.38-7.29 (m, 3H), 7.07 (d,*J* = 7.2 Hz, 1H), 3.46 (s, 2H), 2.39-2.32 (m, 2H), 2.29-2.23 (m, 1H), 2.18 (s, 3H), 1.99-1.93 (m, 1H), 1.10 (s, 3H), 1.08 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.1, 176.0, 162.0, 136.6, 132.6, 131.6, 129.9, 128.5, 127.5, 112.5, 51.0, 36.8, 34.5, 33.5, 29.3, 28.2, 17.8 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{17}H_{20}NO_2]$: 270.1494; found: 270.1499.

1-(3-bromophenyl)-6,6-dimethyl-6,7-dihydro-1H-indole-2,4(3H,5H)-dione(3ac) (Table 2 Entry 29): Yellow solid, Yield: 287 mg, 86%; R_f : 0.3 (20 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57 (d,*J*=8Hz, 1H), 7.40-7.36 (m,2H), 7.16 (d, *J*=7.6Hz, 1H),3.43-3.42 (m, 2H), 2.35 (s, 2H), 2.29-2.28 (m, 2H), 1.11 (s,6H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 192.3, 175.8, 160.4, 134.7, 132.1, 131.0, 130.5, 126.0, 123.2, 112.9, 50.9, 37.2, 34.7, 33.6, 28.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{17}BrNO_2]$: 334.0443; found: 334.0449.

1-cyclohexyl-6,6-dimethyl-3,5,6,7-tetrahydro-1H-indole-2,4-dione (3ad) (Table 2 Entry 30): Black solid, Yield: 230 mg, 88%; R_f : 0.3 (15 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 3.62-3.55 (m, 1H), 3.02 (t, *J*=3.2Hz, 2H), 2.37 (t, *J* = 3.2 Hz, 2H), 2.10 (s, 2H), 1.87-1.83 (m, 2H), 1.71-1.67 (m, 2H), 1.57-1.53 (m, 2H), 1.21-1.03 (m, 4H), 0.99 (s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 191.3, 176.7, 161.7, 111.6, 53.3, 49.9, 37.0, 34.1, 33.4, 30.2, 28.4, 25.7, 24.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{24}NO_2]$: 262.1807; found: 262.1818.

The characterization data of all synthesized compounds (4a-4q) are given below:

4-hydroxy-1-(4-methoxyphenyl)indoline-2,3-dione (4a) (Table 4 Entry 1): Red solid, Yield: 226 mg, 84%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 158-160 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ = 11.37 (brs, 1H), 7.37-7.34 (m, 3H), 7.10 (d,*J* = 8.4 Hz, 2H), 6.58 (d,*J* = 8.7 Hz,1H), 6.08 (d,*J* = 7.5 Hz, 1H), 3.81 (s, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, DMSO-d_6) d: δ = 179.6, 159.0, 158.1, 157.5, 151.8, 139.5, 128.3, 126.0, 114.8, 112.6, 104.8, 101.1, 55.5ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{15}H_{11}NO_4Na]$: 292.0586; found: 292.0594.

4-hydroxy-1-(p-tolyl)indoline-2,3-dione (4b) (Table 4 Entry 2): Red solid, Yield: 215 mg, 85%; R_f : 0.4 (25 % Ethyl acetate in Petroleum ether); m.p. = 140-145 °C ; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.38 (brs, 1H), 7.37-7.30 (m, 5H), 6.59 (d, *J* = 8.4 Hz, 1H), 6.11 (d, *J* = 7.6 Hz, 1H), 2.37 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 179.5, 158.0, 157.6, 151.5, 139.5, 138.0, 130.9, 130.1, 126.6, 112.7, 104.9, 101.1, 20.8 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{15}H_{11}NO_3Na]$: 276.0637; found: 276.0645.

1-(4-chlorophenyl)-4-hydroxyindoline-2,3-dione (4c) (Table 4 Entry 3): Red solid, Yield: 234 mg, 85%; R_f : 0.4 (25 % Ethyl acetate in Petroleum ether); m.p. = 142-144 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.40 (brs, 1H), 7.64 (d, *J*=8.8Hz, 2H), 7.48 (d,*J*=8.8Hz, 2H), 7.37 (t, *J* = 8 Hz, 1H), 6.60 (d, *J* = 8.4 Hz, 1H), 6.17 (d, *J* = 7.6 Hz, 1H) ppm. ^{13}C {^{1}H} NMR (100 MHz, DMSO-d_6) d: δ = 179.1, 157.9, 157.7, 151.0, 139.5, 132.7, 132.5, 129.7, 128.7, 112.9, 105.0, 101.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{14}H_8ClNO_3Na]$: 296.0090; found: 296.0105.

1-(3-chloro-4-methoxyphenyl)-4-hydroxyindoline-2,3-dione (4d) (Table 4 Entry 4): Red solid, Yield: 252 mg, 83%; R_f : 0.4 (35 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.40 (s, 1H), 7.56 (d, *J* = 2.4 Hz, 1H), 7.43-7.32 (m, 3H), 6.59 (d, *J* = 8.4 Hz, 1H), 6.11 (d, *J* = 7.6 Hz, 1H), 3.93 (s, 3H), ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 179.3, 158.1, 157.6, 154.5, 151.5, 139.6, 128.6, 127.3, 126.5, 121.4, 113.3, 112.7, 104.9, 101.0, 56.5 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{15}H_{10}NO_4ClNa]$: 326.0196; found: 326.0204.

4-hydroxy-1-(2-hydroxyphenyl)indoline-2,3-dione (4e) (Table 4 Entry 5): Red solid, Yield: 199 mg, 78%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.34 (brs, 1H), 9.77(s, 1H), 7.36-7.32 (m, 2H), 7.28 (d, *J*=7.6Hz, 1H), 7.03 (d, *J* = 8 Hz, 1H), 6.95 (t, *J*=7.6Hz, 1H), 6.57 (d, *J*=8.4, 1H), 5.87 (d, *J*=7.6,1H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 179.8, 158.4, 157.5, 153.7, 151.7,

139.6, 130.6, 129.6, 120.1, 119.8, 117.3, 112.5, 105.1, 101.3 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{14}H_9NO_4Na]$: 278.0429; found: 278.0435.

4-hydroxy-1-(3-nitrophenyl)indoline-2,3-dione (4f) (Table 4 Entry 6): Red solid, Yield: 227 mg, 80%; R_f : 0.3 (30 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.45 (brs, 1H), 8.34-8.31 (m, 2H), 7.95 (d, *J*=8.4Hz, 1H), 7.87(t, *J*=8Hz, 1H) 7.40 (t, *J* = 8 Hz, 1H), 6.64 (d, *J* = 8.4 Hz, 1H), 6.28 (d, *J*=7.6 Hz, 1H), ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 178.8, 157.9, 157.8, 150.6, 148.4, 139.5, 134.7, 133.5, 131.1, 123.1, 121.8, 113.1, 105.1, 101.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{14}H_8N_2O_5Na]$: 307.0331; found: 307.0338.

1-(benzo[d][1,3]dioxol-4-yl)-4-hydroxyindoline-2,3-dione(4g) (Table 4 Entry 7): Red solid, Yield: 229 mg, 81%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.36 (brs, 1H), 7.38-7.34 (m, 1H), 7.07 (d, *J*=8Hz, 1H), 7.03(d, *J*=2Hz, 1H), 6.92-6.89 (m, 1H), 6.58 (d, *J* = 8.4 Hz, 1H), 6.12 (s, 2H), 6.11 (d, J=7.6,1H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 179.5, 158.1, 157.5, 151.8, 148.0, 147.3, 139.6, 127.1, 120.8, 112.6, 108.7, 108.1, 104.8, 101.9, 101.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{15}H_9NO_5Na]$: 306.0378; found: 306.0385.

4-hydroxy-1-(3-methoxyphenyl)indoline-2,3-dione(4h) (Table 4 Entry 8): Red solid, Yield: 213 mg, 79%; R_f : 0.2 (20 % Ethyl acetate in Petroleum ether); m.p. = 145-147 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.45-7.38 (m, 2H), 6.99-6.96 (m, 2H), 6.93-6.92 (m, 1H), 6.62 (d, *J*=8.4Hz, 1H), 6.39 (d, *J*=7.6Hz, 1H), 3.83 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 183.3, 160.8, 158.2, 157.8, 150.0, 141.2, 134.1, 130.7, 118.1, 114.6, 113.3, 111.9, 105.0, 103.2, 55.7 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{15}H_{11}NO_4Na]$: 292.0586; found: 292.0597.

1-cyclohexyl-4-hydroxyindoline-2,3-dione (4i) (Table 4 Entry 9): Red solid, Yield: 203 mg, 83%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 154-156 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ = 11.25 (brs, 1H), 7.40 (t, *J* = 8.1 Hz, 1H), 6.69 (d,*J* = 7.8 Hz, 1H), 6.53 (d, *J* = 8.4 Hz, 1H), 3.96-3.87 (m, 1H), 2.06-1.98 (m, 2H), 1.78-1.68 (m, 5H), 1.38-1.25 (m, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, DMSO-d_6) d: δ = 180.1, 158.2, 157.6, 150.6, 139.6, 112.1, 104.8, 101.9, 52.3, 28.7, 25.4, 24.8 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{14}H_{15}NO_3Na]$: 268.0950; found: 268.0959.

4-hydroxy-1-(2-iodophenyl)indoline-2,3-dione (4j) (Table 4 Entry 10): Red solid, Yield: 292 mg, 80%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR

(400 MHz, DMSO-d_6): δ = 11.55(brs, 1H), 7.07 (d, *J*=8Hz, 2H), 7.63-7.54 (m, 2H), 7.39-7.31 (m, 2H), 6.62 (d, *J*=8.8Hz, 1H), 5.79 (d, *J*=7.6Hz, 1H) ppm. ^{13}C {^{1}H} NMR (100 MHz, DMSO-d_6) d: δ = 179.1, 157.9, 157.4, 150.9, 140.0, 139.8, 136.5, 131.4, 130.1, 130.0, 112.9, 104.7, 101.4, 99.5 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{14}H_8INO_3Na]$: 387.9447; found: 387.9453.

4-hydroxy-1-(2-methoxyphenyl)-6-methylindoline-2,3-dion(4k) (Table 4 Entry 11): Red solid, Yield: 215 mg, 76%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 168-170 °C;^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84(brs, 1H), 7.47-7.43 (m, 1H), 7.28 (d, *J*=6.4Hz, 1H), 7.10-7.07 (m, 2H), 6.41 (s, 1H), 5.87 (s, 1H), 3.80 (s, 3H), 2.26 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 182.5, 159.2, 157.6, 155.4, 154.2, 150.7, 131.0, 129.3, 121.6, 121.3, 112.7, 112.6, 104.2, 103.6, 56.0, 23.4 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{16}H_{13}NO_4Na]$: 306.0742; found: 306.0749.

4-hydroxy-6-methyl-1-(3-nitrophenyl) indoline-2,3-dione(4l) (Table 4 Entry 12): Red solid, Yield: 223 mg, 75%; R_f : 0.4 (35 % Ethyl acetate in Petroleum ether); m.p. = 178-180 °C;^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.37 (brs, 1H), 8.33-8.30 (m, 2H), 7.94 (d, *J*=7.6, 1H), 7.87 (t, *J*=8, 1H), 6.43 (s, 1H), 6.13 (s, 1H), 2.21 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, DMSO-d_6) d: δ = 178.0, 158.5, 157.8, 151.6, 150.6, 148.5, 134.7, 133.7, 131.1, 123.1, 122.1, 112.9, 103.3, 102.4, 22.4 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{15}H_{10}N_2O_5Na]$: 321.0487; found: 321.0495.

4-hydroxy-1-(4-methoxyphenyl)-6-methylindoline-2,3-dione(4m) (Table 4 Entry 13): Red solid, Yield: 224 mg, 79%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 165-167 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.76 (brs, 1H), 7.29 (d, *J*=8.7Hz, 2H), 7.04 (d, *J*=9Hz, 2H), 6.42 (s, 1H), 6.11 (s, 1H), 3.86 (s, 3H), 2.29 (s, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 182.6, 159.8, 159.3, 157.7, 154.3, 150.5, 127.7, 125.7, 115.2, 112.9, 104.3, 103.3, 55.7, 23.5 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{16}H_{13}NO_4Na]$: 306.0742; found: 306.0749.

4-hydroxy-1-(3-nitrophenyl)-6-phenylindoline-2,3-dione(4n) (Table 4 Entry 14): Red solid, Yield: 277 mg, 77%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 215-217 °C;^{1}H NMR (300 MHz, DMSO-d_6): δ = 11.57 (brs, 1H), 8.38 (s, 1H), 8.33 (d, *J*=8.4Hz, 1H), 8.01 (d, *J*=7.8Hz, H), 7.88 (t, *J*=8.1, 1H), 7.59-7.55 (m, 2H), 7.46-7.43 (m, 3H), 6.86 (s, 1H), 6.47 (s, 1H) ppm. ^{13}C {^{1}H} NMR (75 MHz, DMSO-d_6) d: δ = 178.2, 158.4, 157.9, 151.2, 148.6, 138.8, 134.6, 133.7, 131.2, 129.3, 129.1, 128.9, 127.0, 123.2, 122.2, 111.0, 104.3, 99.9

ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{20}H_{12}N_2O_5Na]$: 383.0644; found: 383.0653.

4-hydroxy-6-phenyl-1-(o-tolyl)indoline-2,3-dione (4o) (Table 4 Entry 15): Red solid, Yield: 240 mg, 73%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 188-190 °C ; ^{1}H NMR (300 MHz, DMSO-d_6): δ = 11.61 (brs, 1H), 7.47-7.40 (m,9H), 6.82 (s, 1H), 6.00 (s, 1H), 2.18 (s, 3H) ppm. ^{13}C {^{1}H} NMR (75 MHz, DMSO-d_6) d: δ = 178.8, 158.3, 157.8, 152.0, 151.4, 138.8, 136.3, 132.3, 131.3, 129.4, 129.3, 129.2, 128.3, 127.3, 126.8, 110.8, 104.2, 99.8, 17.4 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{21}H_{15}NO_3Na]$: 352.0950; found: 352.1003.

1-(3-bromophenyl)-4-hydroxy-6-phenylindoline-2,3-dione (4p) (Table 4 Entry 16): Red solid, Yield: 292 mg, 74%; R_f : 0.4 (35 % Ethyl acetate in Petroleum ether); m.p. = 206-208 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.90(brs, 1H), 7.62-7.59 (m, 2H), 7.54-7.51 (m, 2H), 7.47-7.41 (m, 5H), 6.86 (s, 1H), 6.56 (s, 1H) ppm. ^{13}C {^{1}H} NMR (75 MHz, $CDCl_3$) d: δ = 182.0, 158.6, 158.0, 155.1, 149.7, 139.2, 134.4, 132.2, 131.3, 129.9, 129.4, 129.3, 127.4, 125.0, 123.4, 111.8, 104.1, 102.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{20}H_{12}BrNO_3Na]$: 415.9898; found: 415.9899.

4-hydroxy-1-(2-iodophenyl)-6-phenylindoline-2,3-dione (4q) (Table 4 Entry 17): Red solid, Yield: 318 mg, 72%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 210-212 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ = 11.74 (brs, 1H), 8.08 (d,J = 8.1 Hz, 1H), 7.63-7.61 (m, 2H), 7.49-7.43 (m, 5H), 7.35-7.30 (m, 1H), 6.85 (s, 1H), 5.98 (s, 1H) ppm. ^{13}C {^{1}H} NMR (75 MHz, DMSO-d_6) d: δ = 178.8, 158.1, 157.9, 151.6, 151.4, 139.8, 138.6, 136.4, 131.4, 130.2, 130.1, 129.4, 129.2, 126.8, 110.9, 103.9, 100.1, 99.5ppm. HRMS (ESI/TOF-Q) m/z: $[M+Na]^+$calcd for $[C_{20}H_{12}INO_3Na]$: 463.9760; found: 463.9771.

Methyl(Z)-2-(4-hydroxy-1-(3-methoxyphenyl)-2-oxoindolin-3-ylidene)acetate(4ha): Yellow solid, Yield: 254 mg, 78%; R_f : 0.3 (35 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ =11.80 (brs, 1H), 7.44-7.40 (m, 1H), 7.18 (t, J = 8 Hz, 1H), 6.99-6.93 (m, 2H), 6.89-6.88 (m, 2H), 6.59 (d, J=8.4Hz, 1H), 6.20 (d, J=8Hz, 1H), 3.94 (s, 3H), 3.82 (s, 3H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 170.8, 166.7, 160.7, 157.9, 147.7, 137.0, 135.6, 135.1, 130.6, 119.3, 117.5, 114.7, 114.5, 112.7, 106.9, 101.6, 55.6, 53.8 ppm. HRMS (ESI/TOF-Q) m/z:$[M+H]^+$calcd for $[C_{18}H_{16}NO_5]$: 326.1028; found: 326.1037.

The characterization data of all synthesized compounds (5a-5d) are given below:

1-(4-methoxyphenyl)-6,6-dimethyl-5,6-dihydro-1H-indole-2,4-dione (5a) (Table 5 Entry 1): Yellow solid, Yield: 215 mg, 76%; R_f : 0.3 (15 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.24 (d, *J*=8.8Hz, 2H), 7.01 (d, *J*=8.8Hz, 2H), 6.62-6.61 (m, 1H), 5.74-5.73 (m, 1H), 3.84 (s, 3H), 2.64 (d, J=2Hz, 2H), 1.22 (s, 6H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 194.4, 168.4, 159.3, 139.7, 137.8, 128.5, 126.4, 123.6, 120.0, 114.9, 55.7, 55.0, 37.6, 31.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{17}H_{18}NO_3]$: 284.1287; found: 284.1294.

1-(4-bromophenyl)-6,6-dimethyl-5,6-dihydro-1H-indole-2,4-dione(5b) (Table 5 Entry 2): Yellow solid, Yield: 249 mg, 75%; R_f : 0.4 (15 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ;^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.56-7.52 (m, 2H), 7.39 (t, *J* = 8 Hz, 1H), 7.31-7.28 (m, 1H), 6.63 (d, *J* = 2 Hz, 1H), 5.81 (d, J=2, 1H), 2.67 (S, 2H), 1.26 (S, 6H) ppm. ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 194.1, 167.9, 138.8, 138.2, 135.2, 131.3, 130.9, 130.3, 125.9, 123.9, 123.0, 119.8, 54.9, 37.8, 31.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{15}NO_2Br]$: 332.0286; found: 332.0295.

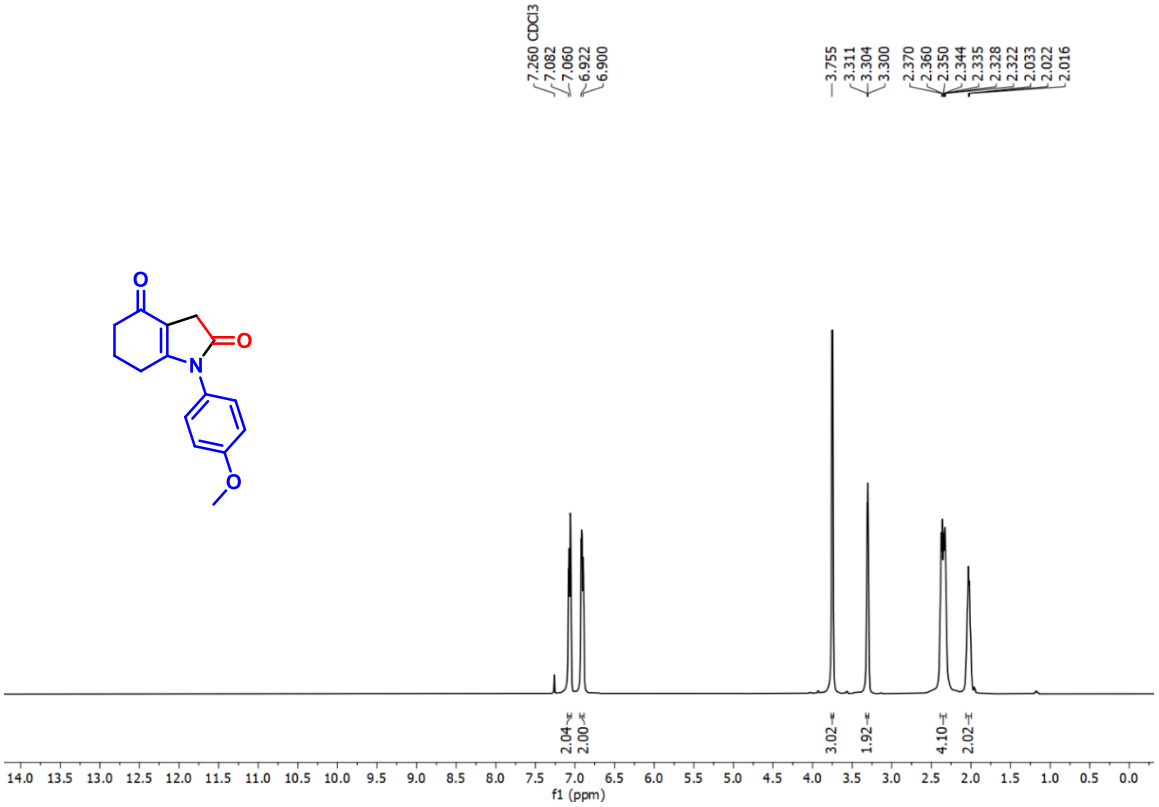
1-(3,4-dimethoxyphenyl)-6,6-dimethyl-5,6-dihydro-1H-indole-2,4-dione(5c) (Table 5 Entry 3): Yellow solid, Yield: 244 mg, 78%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ;^{1}H NMR (400 MHz, $CDCl_3$): δ = 6.94 (d, J=8.4, 1H), 6.85-6.82 (m, 2H), 6.58 (d, *J* = 2 Hz, 1H), 5.74 (d, *J* = 2 Hz, 1H), 3.89 (s, 3H), 3,86 (s, 3H), 2.63 (s, 2H), 1.21 (s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 194.3, 168.4, 149.6, 149.0, 139.7, 137.7, 126.5, 123.8, 119.8, 119.7, 111.4, 110.9, 56.2, 54.9, 37.6, 31.0 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{18}H_{20}NO_4]$: 314.1392; found: 314.1401.

1-(3-bromophenyl)-6,6-dimethyl-5,6-dihydro-1H-indole-2,4-dione(5d) (Table 5 Entry 4): Yellow solid, Yield: 239 mg, 72%; R_f : 0.3 (15 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.56-7.52 (m, 2H), 7.39 (t, *J* = 8 Hz, 1H), 7.31-7.28 (m, 1H), 6.63 (d, *J* = 2 Hz, 1H), 5.81 (d, *J*=2, 1H), 2.67 (s, 2H), 1.26 (s, 6H) ppm.^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 194.1, 167.9, 138.8, 138.2, 135.2, 131.3, 130.9, 130.3, 125.9, 123.9, 123.0, 119.8, 54.9, 37.8, 31.1 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{16}H_{15}NO_2Br]$: 332.0286; found: 332.0294.

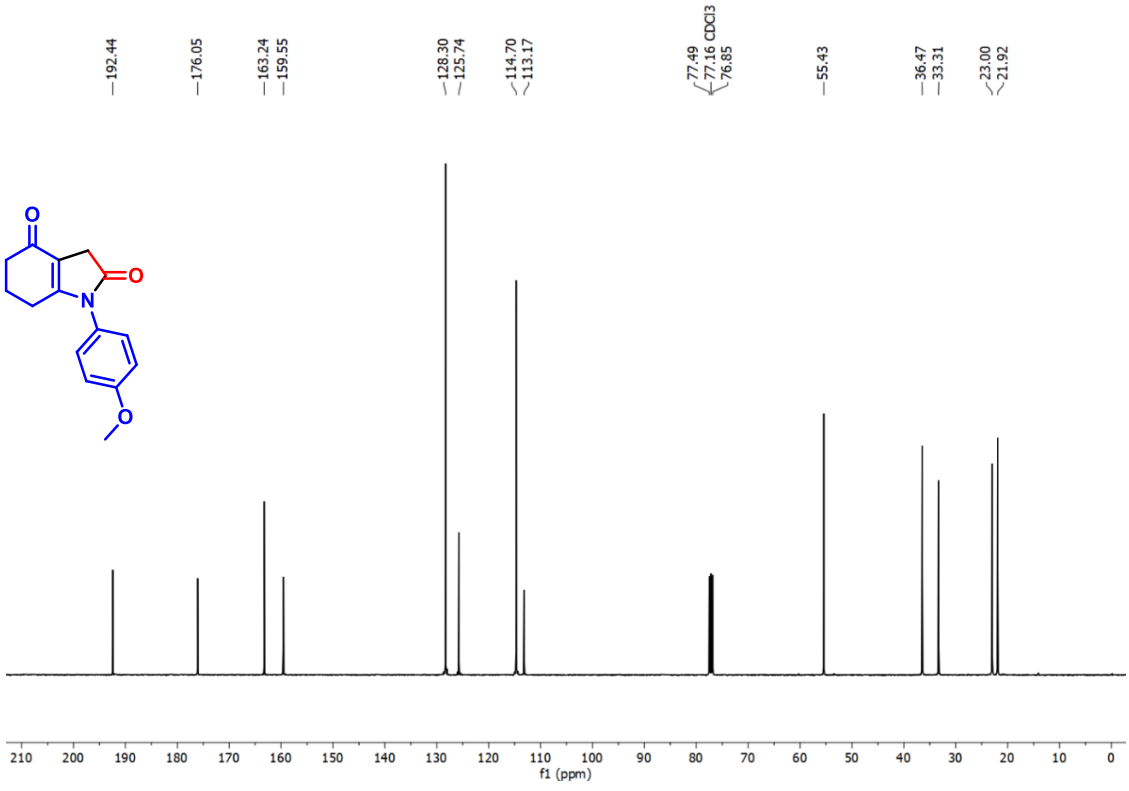
4-methoxy-1-(4-methoxyphenyl)-6,6-dimethyl-1,6-dihydro-2H-indol-2-one(6a): White solid, Yield: 244 mg, 82%; R_f : 0.3 (25 % Ethyl acetate in Petroleum ether); m.p. = 250-251 °C ; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.25 (d, *J*=8.8Hz, 2H), 7.00 (d, *J*=8.8Hz, 2H), 6.05 (d, J=1.6, 1H), 5.56 (m, 1H), 5.25 (d, *J*=1.6, 1H), 3.84 (s, 3H), 3.73 (s, 3H), 1.24 (s, 6H) ppm.

^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) d: δ = 171.0, 158.8, 146.8, 138.8, 137.5, 128.5, 127.6, 119.6, 116.0, 114.7, 109.2, 55.7, 54.8, 39.5, 29.2 ppm. HRMS (ESI/TOF-Q) m/z: $[M+H]^+$calcd for $[C_{18}H_{20}NO_3]$: 298.1443; found: 298.1449.

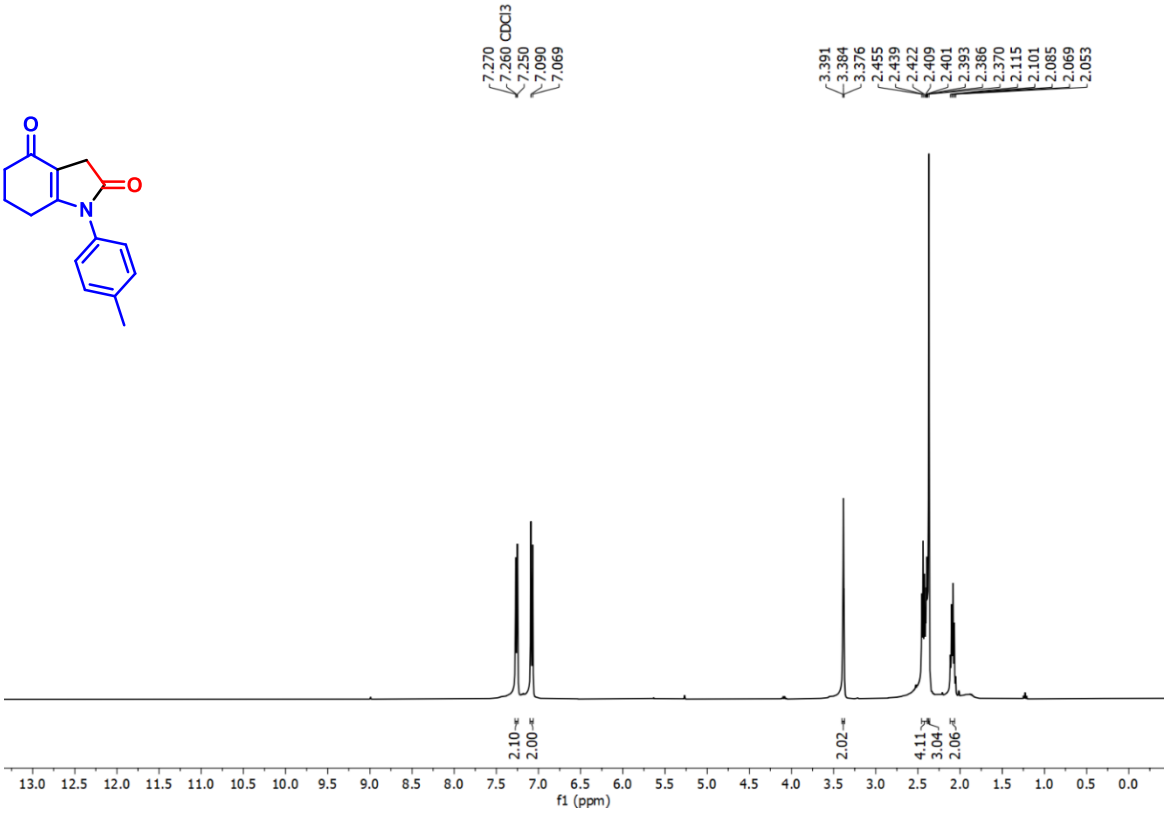
^{1}H NMR and ^{13}C NMR Spectra of compounds



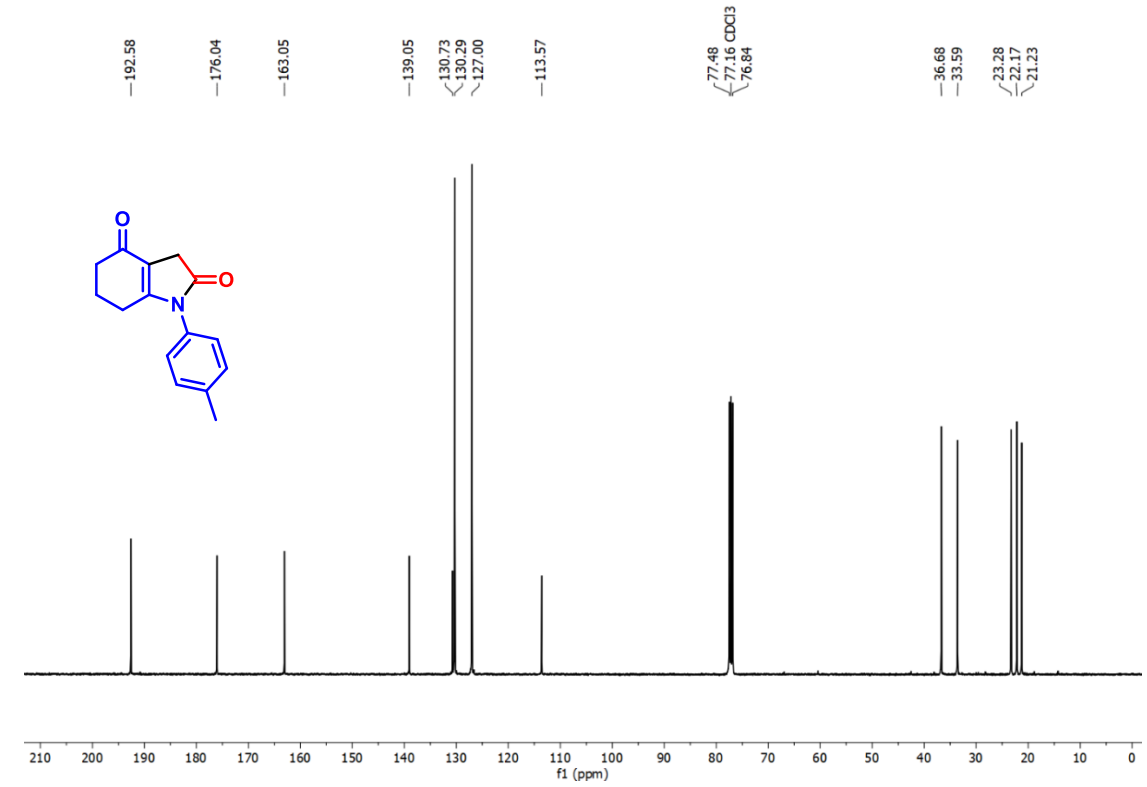
^{1}H NMR Spectrum of Compound **3a** (400 MHz, $CDCl_3$)



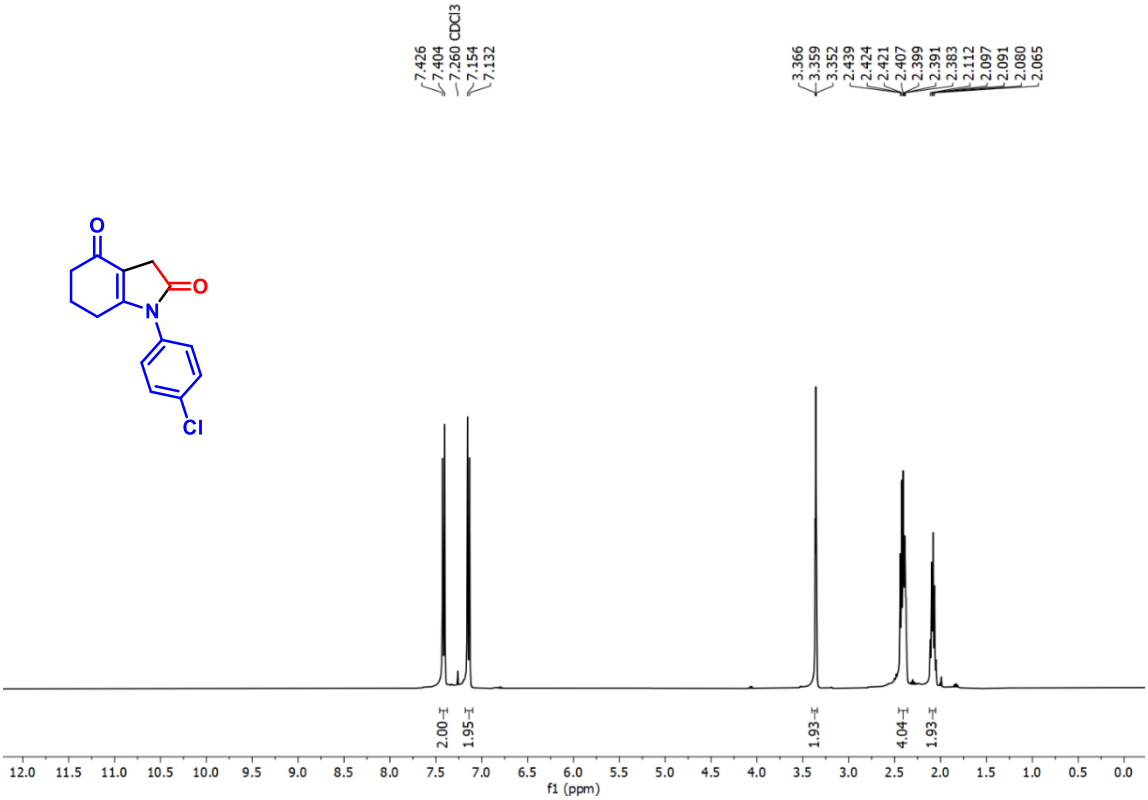
^{13}C {^{1}H} NMR Spectrum of Compound **3a** (100 MHz, $CDCl_3$)



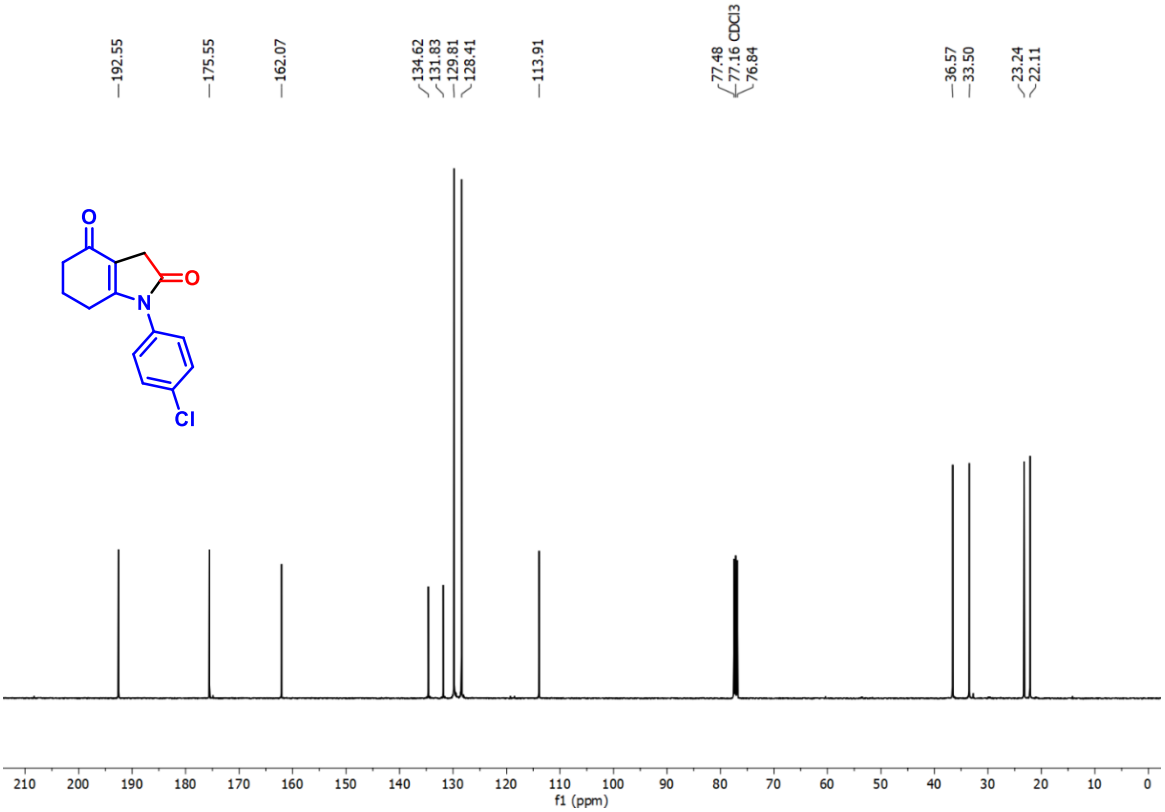
^{1}H NMR Spectrum of Compound **3b** (400 MHz, $CDCl_3$)



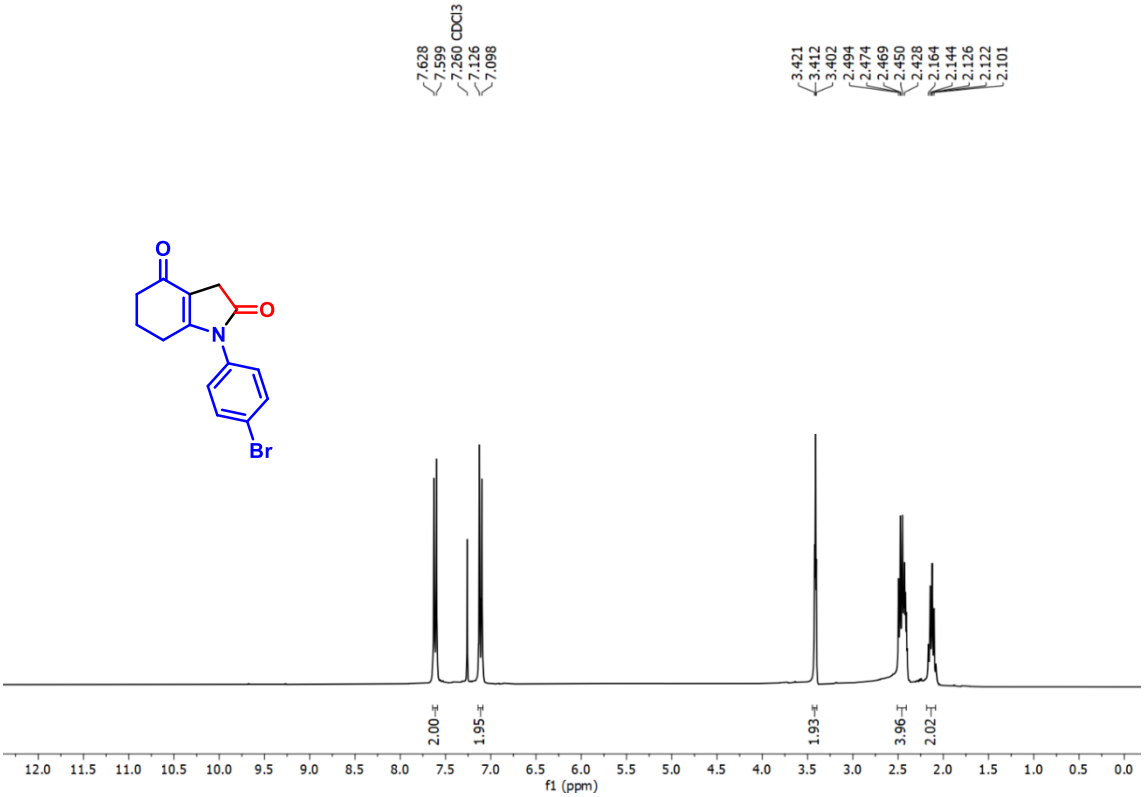
^{13}C {^{1}H} NMR Spectrum of Compound **3b** (100 MHz, $CDCl_3$)



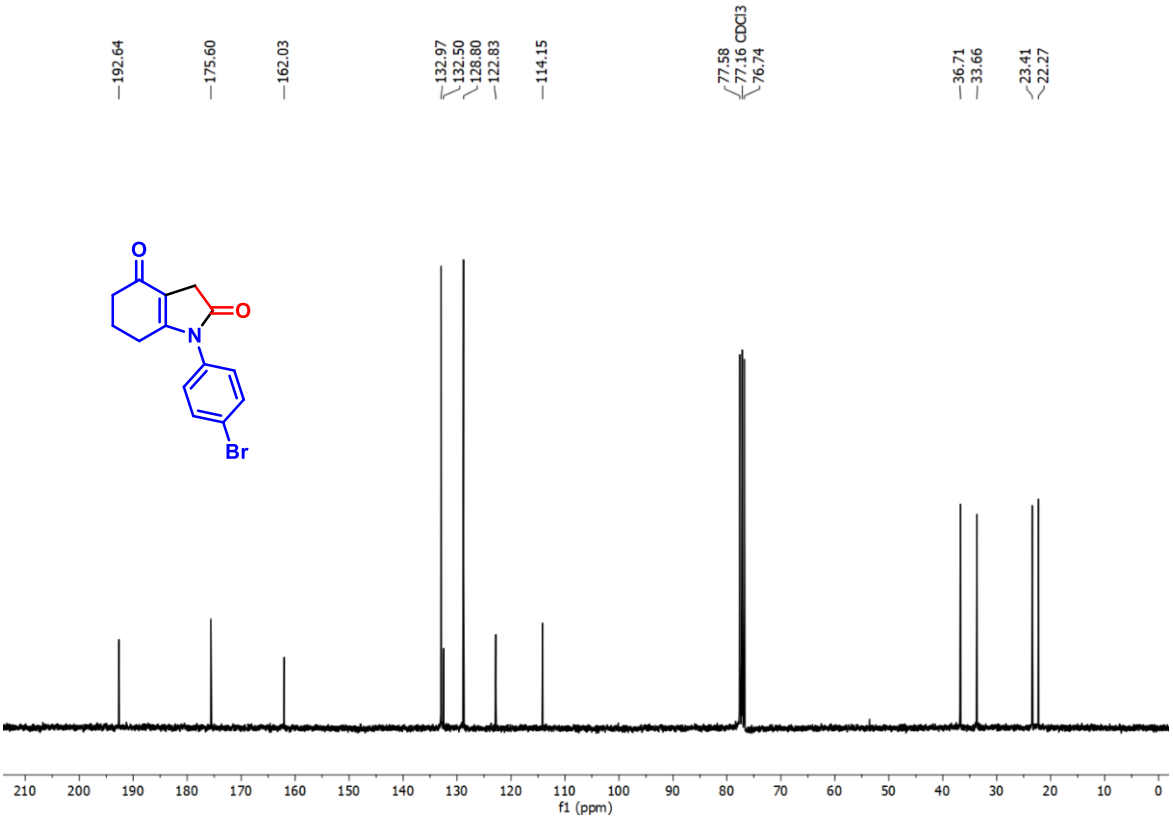
^{1}H NMR Spectrum of Compound **3c** (400 MHz, $CDCl_3$)



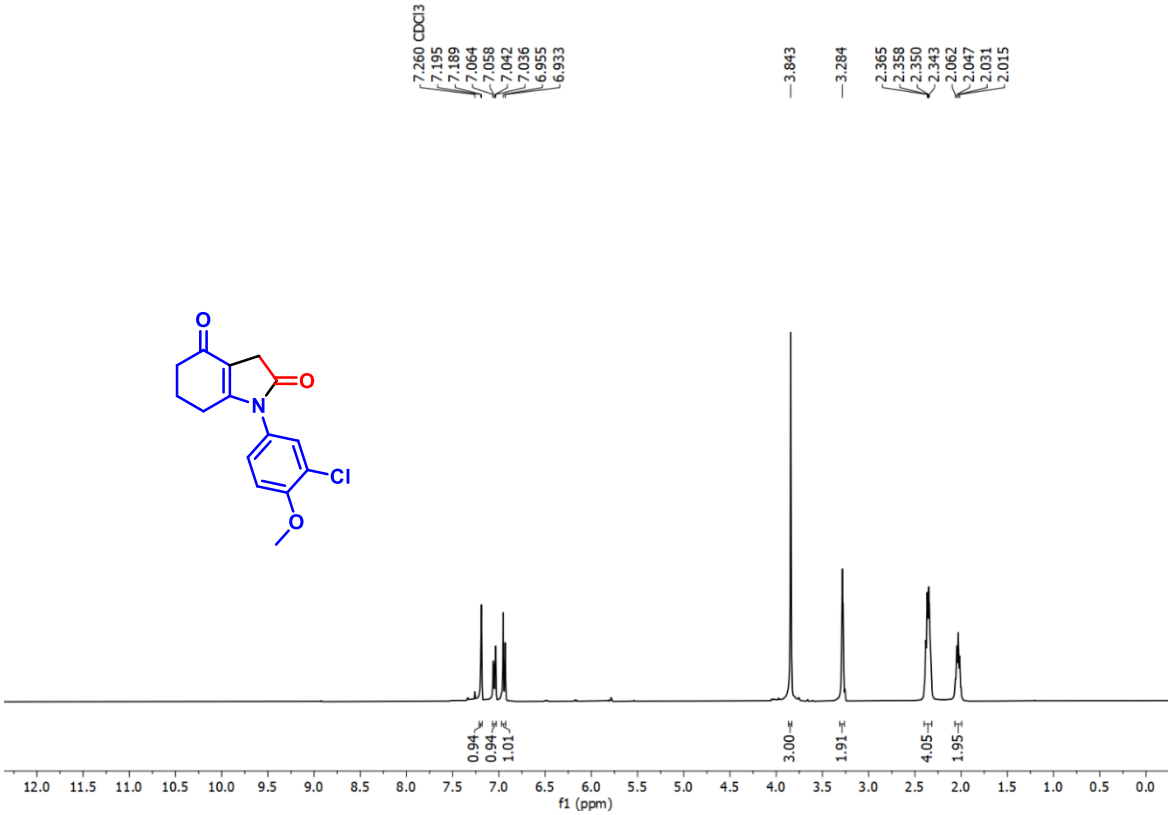
^{13}C {^{1}H} Spectrum of Compound **3c** (100 MHz, $CDCl_3$)



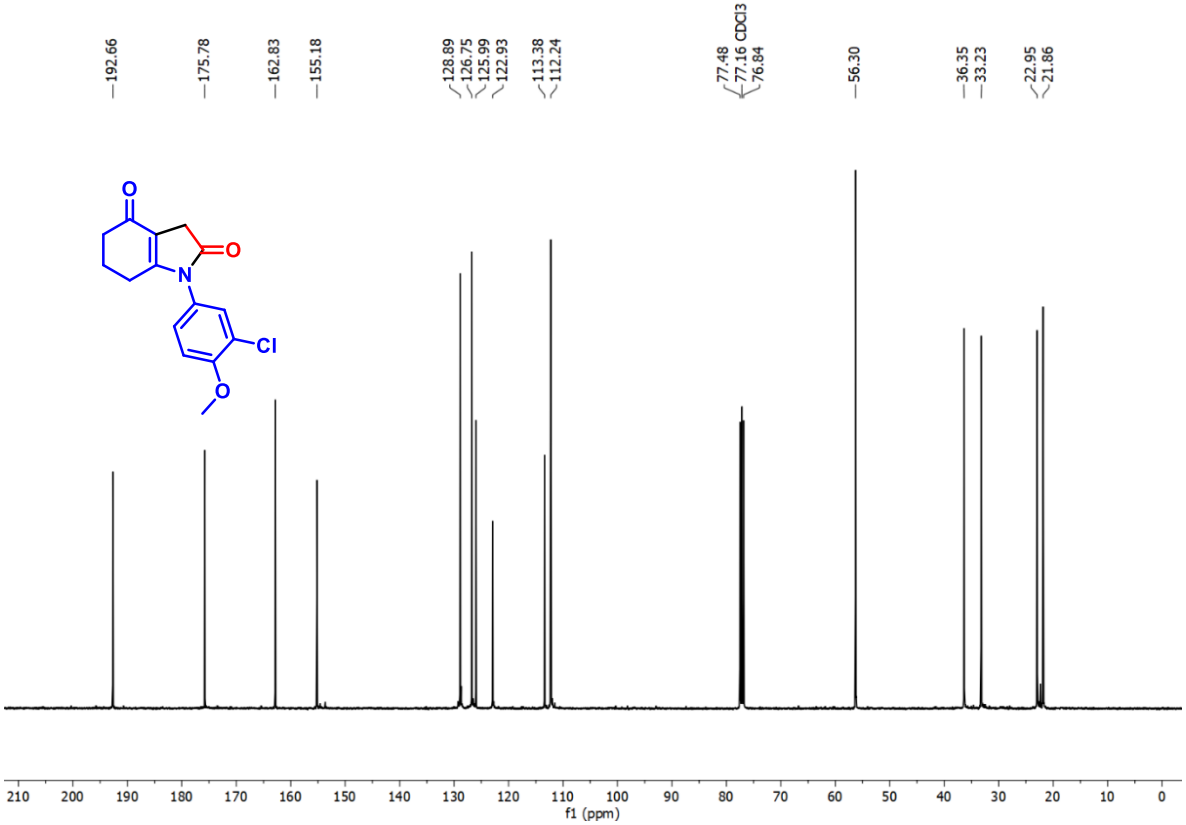
^{1}H NMR Spectrum of Compound **3d** (300 MHz, $CDCl_3$)



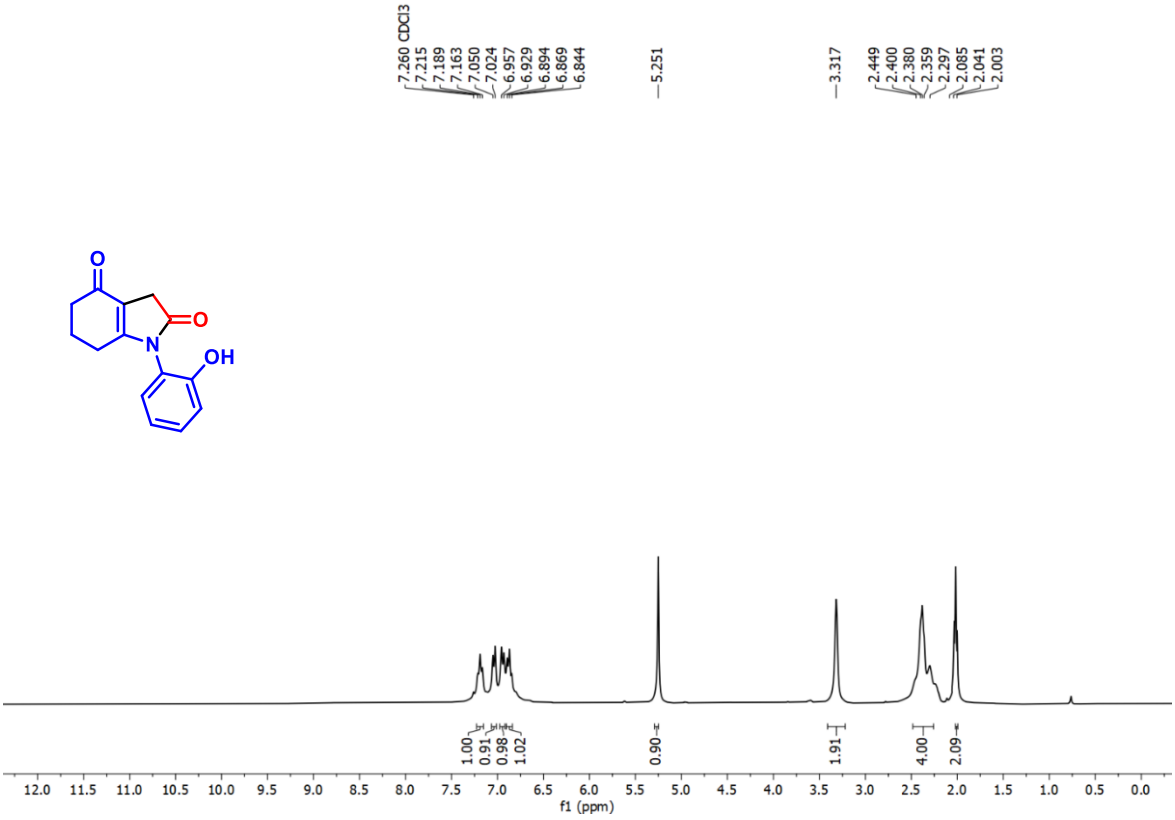
^{13}C {^{1}H}NMR Spectrum of Compound **3d** (75 MHz, $CDCl_3$)



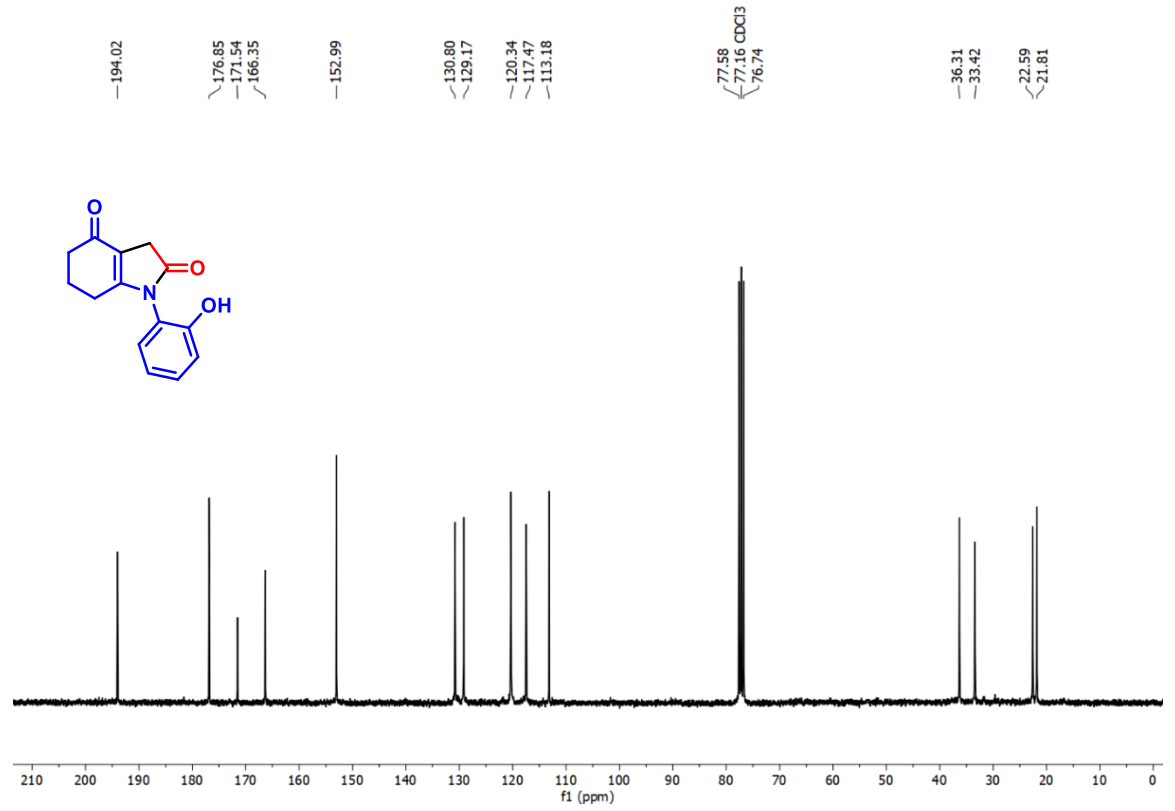
^{1}H NMR Spectrum of Compound **3e** (400 MHz, $CDCl_3$)



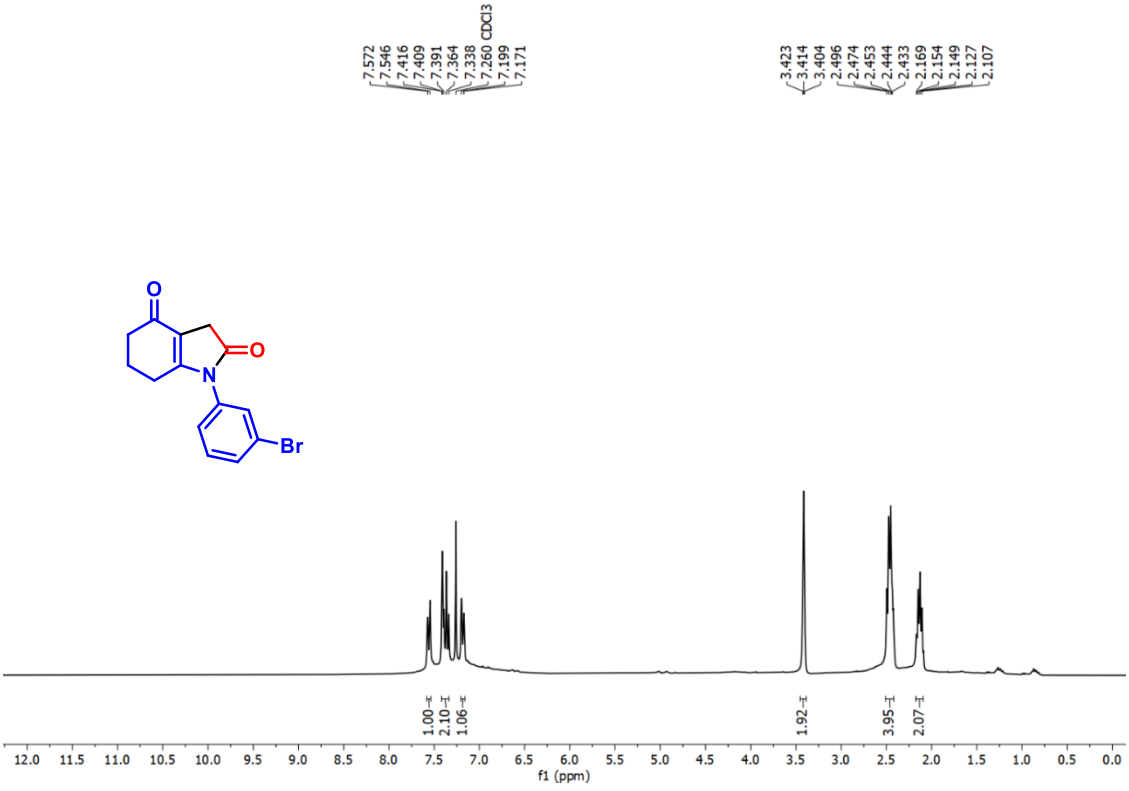
^{13}C {^{1}H}NMR Spectrum of Compound **3e** (100 MHz, $CDCl_3$)



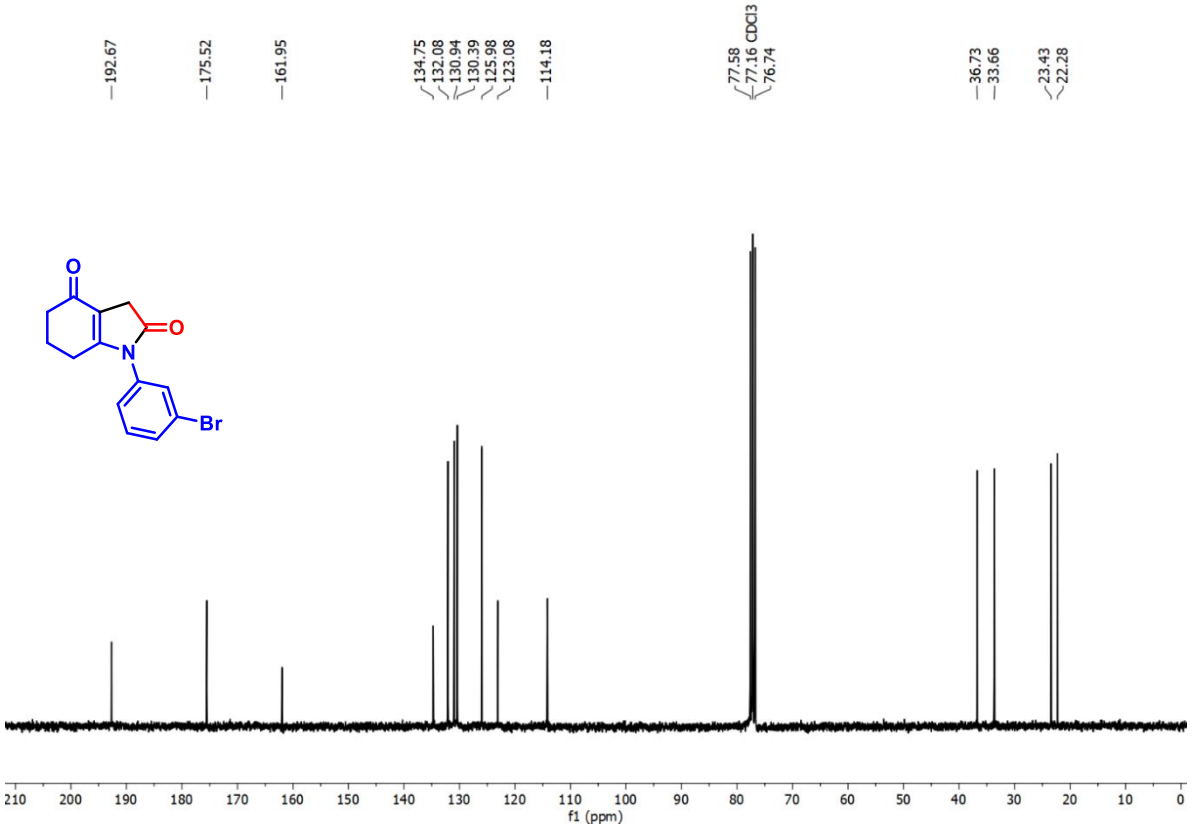
^{1}H NMR Spectrum of Compound **3f** (300 MHz, $CDCl_3$)



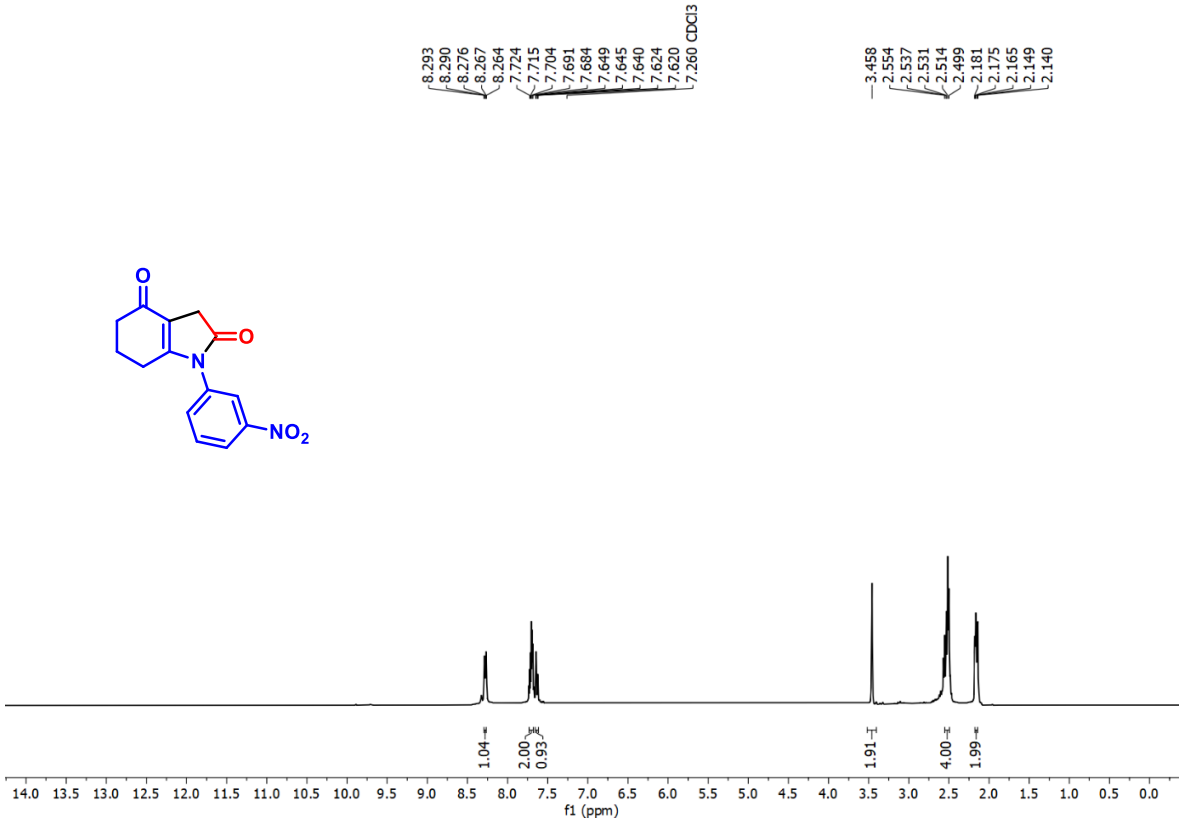
^{13}C {^{1}H}NMR Spectrum of Compound **3f** (75 MHz, $CDCl_3$)



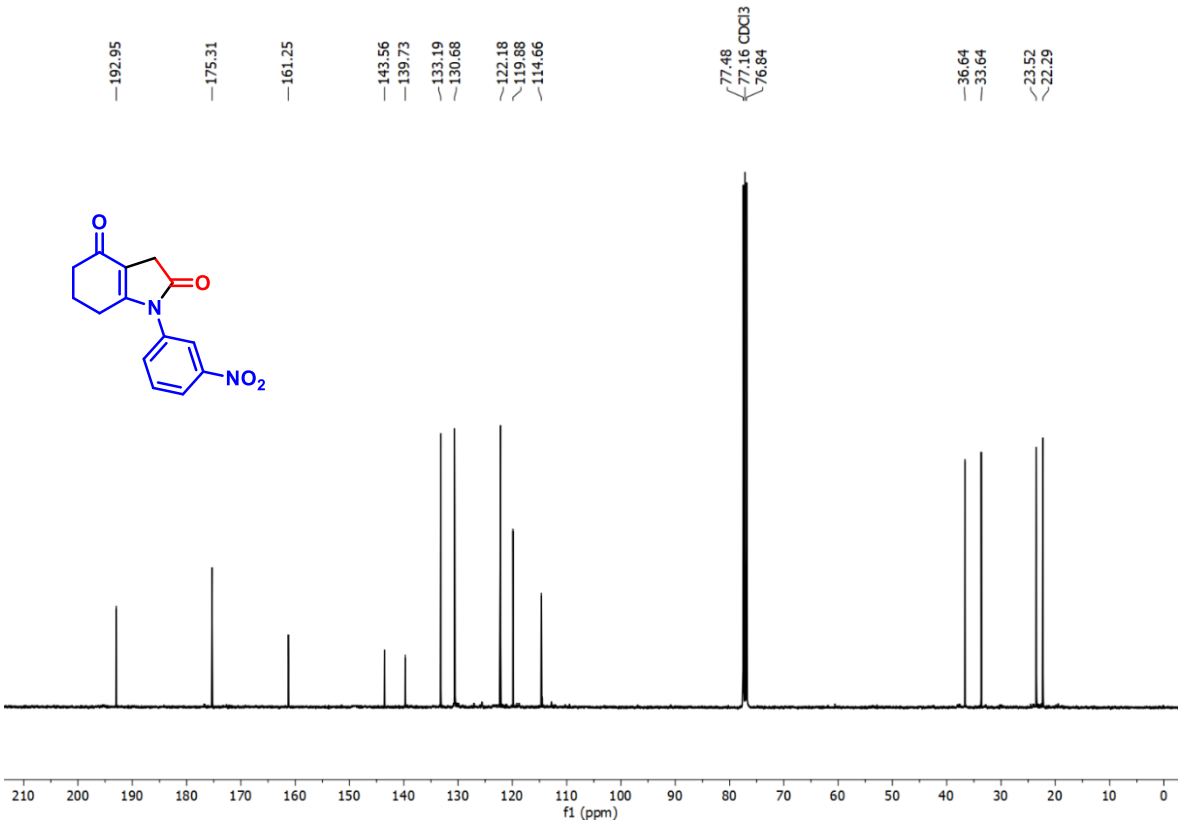
^{1}H NMR Spectrum of Compound **3g** (300 MHz, $CDCl_3$)



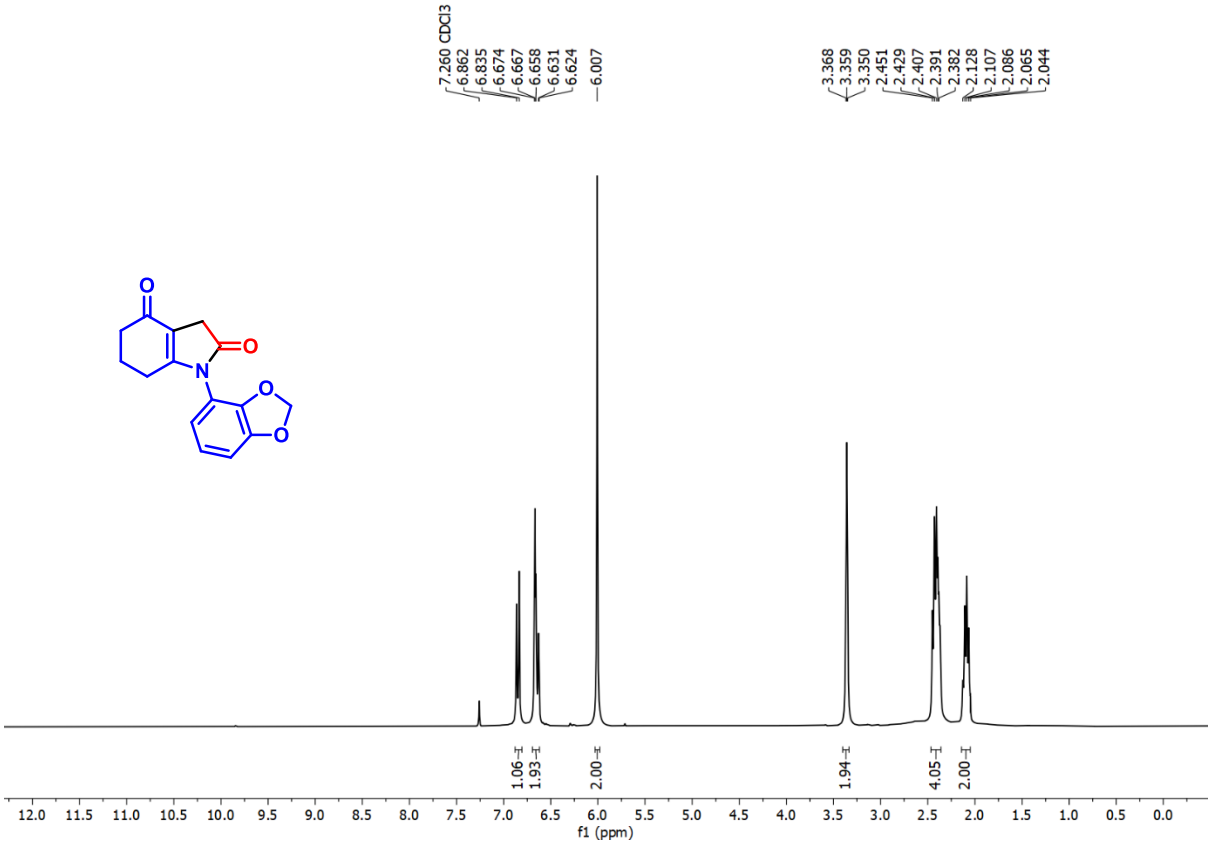
^{13}C {^{1}H}NMR Spectrum of Compound **3g** (75 MHz, $CDCl_3$)



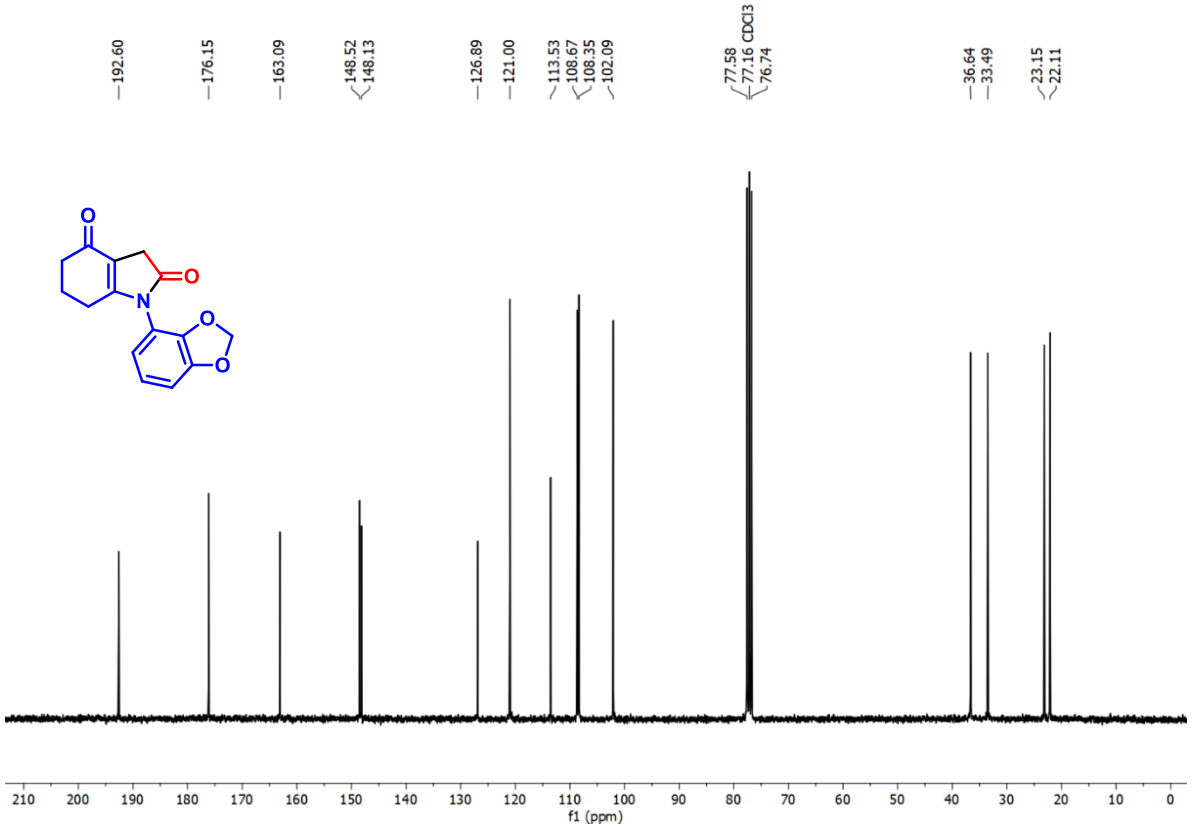
^{1}H NMR Spectrum of Compound **3h** (400 MHz, $CDCl_3$)



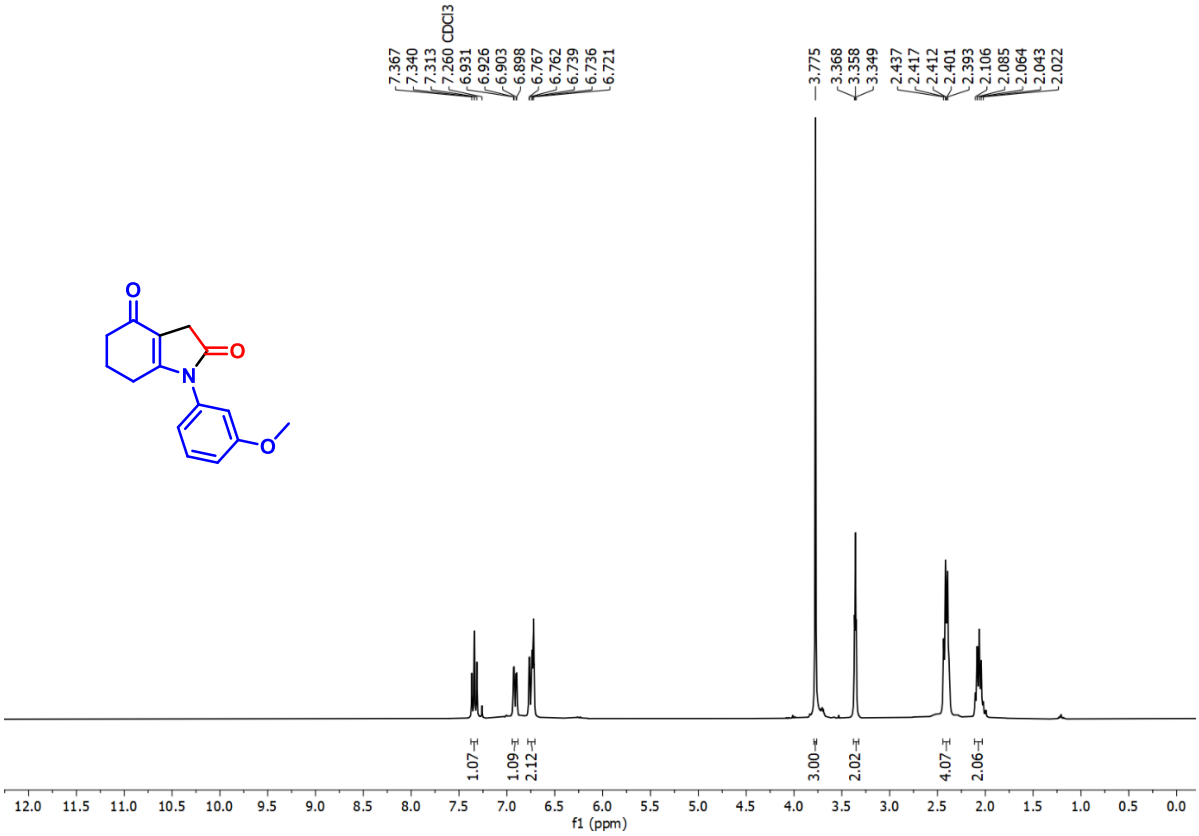
^{13}C {^{1}H} NMR Spectrum of Compound **3h** (100 MHz, $CDCl_3$)



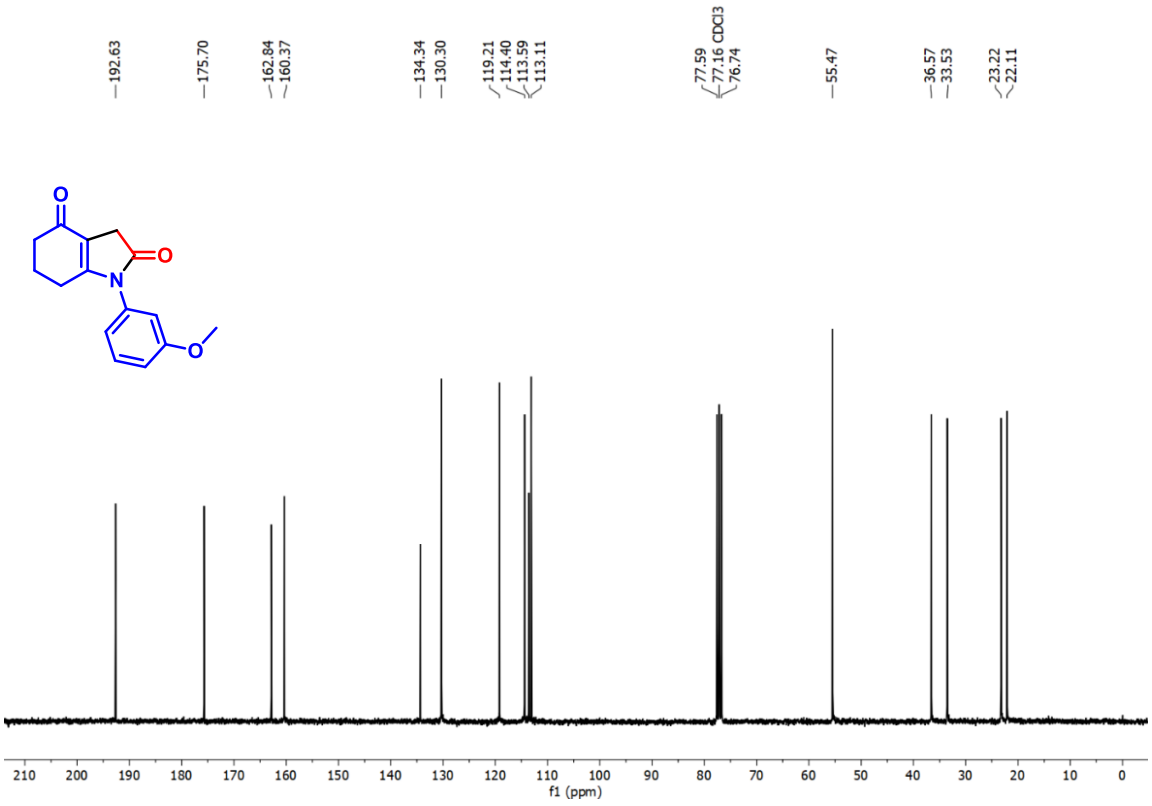
^{1}H NMR Spectrum of Compound **3i** (300 MHz, $CDCl_3$)



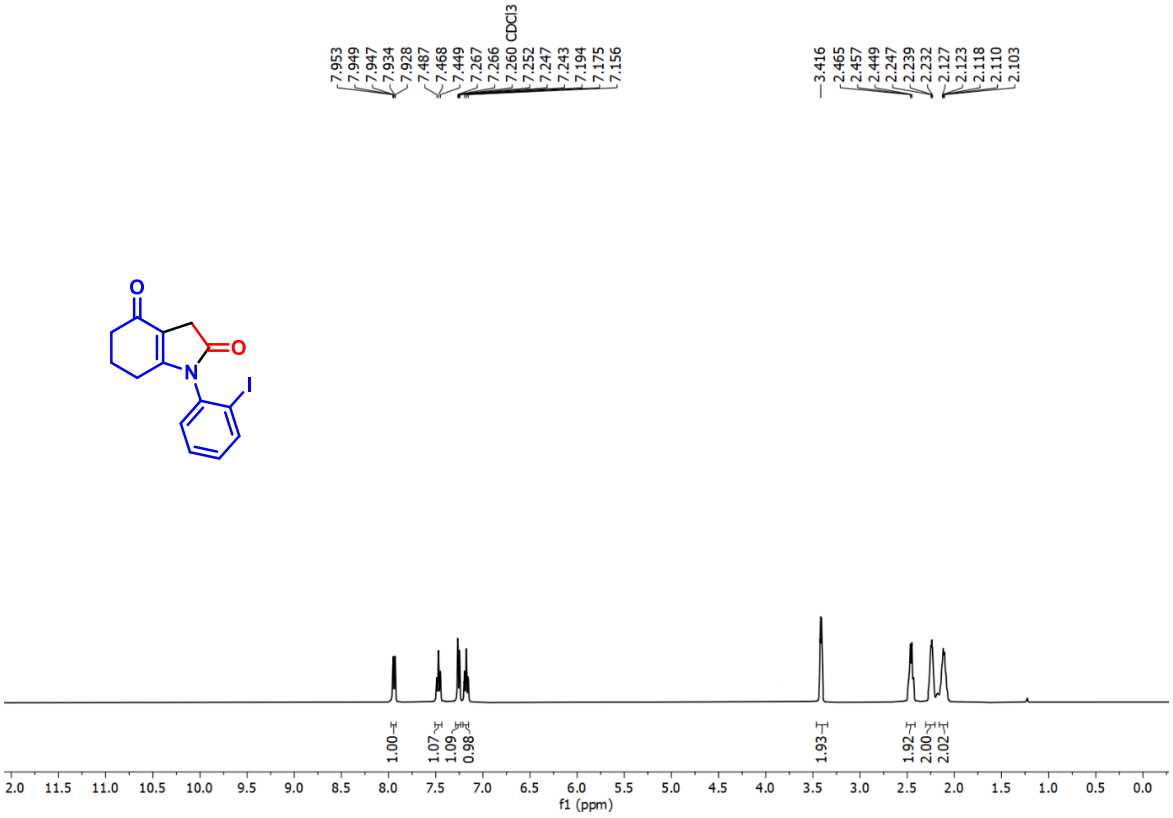
^{13}C {^{1}H}NMR Spectrum of Compound **3i** (75 MHz, $CDCl_3$)



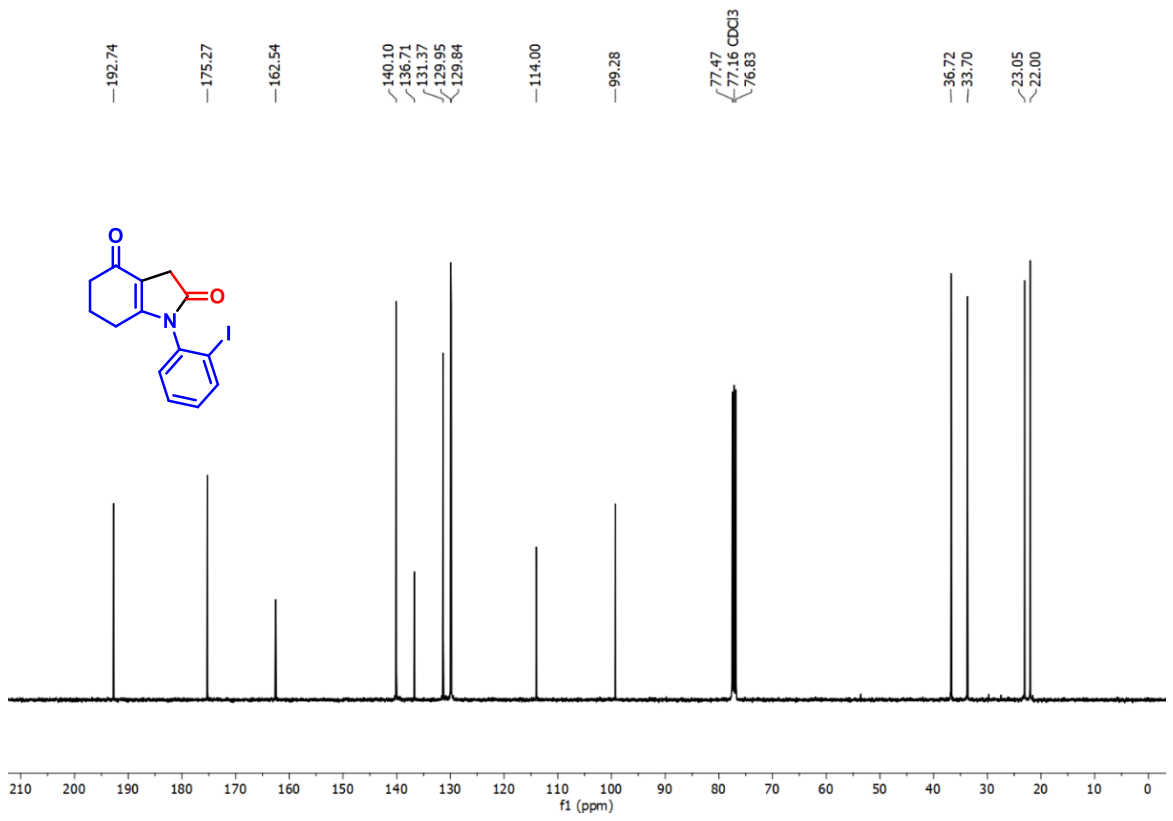
^{1}H NMR Spectrum of Compound **3j** (300 MHz, $CDCl_3$)



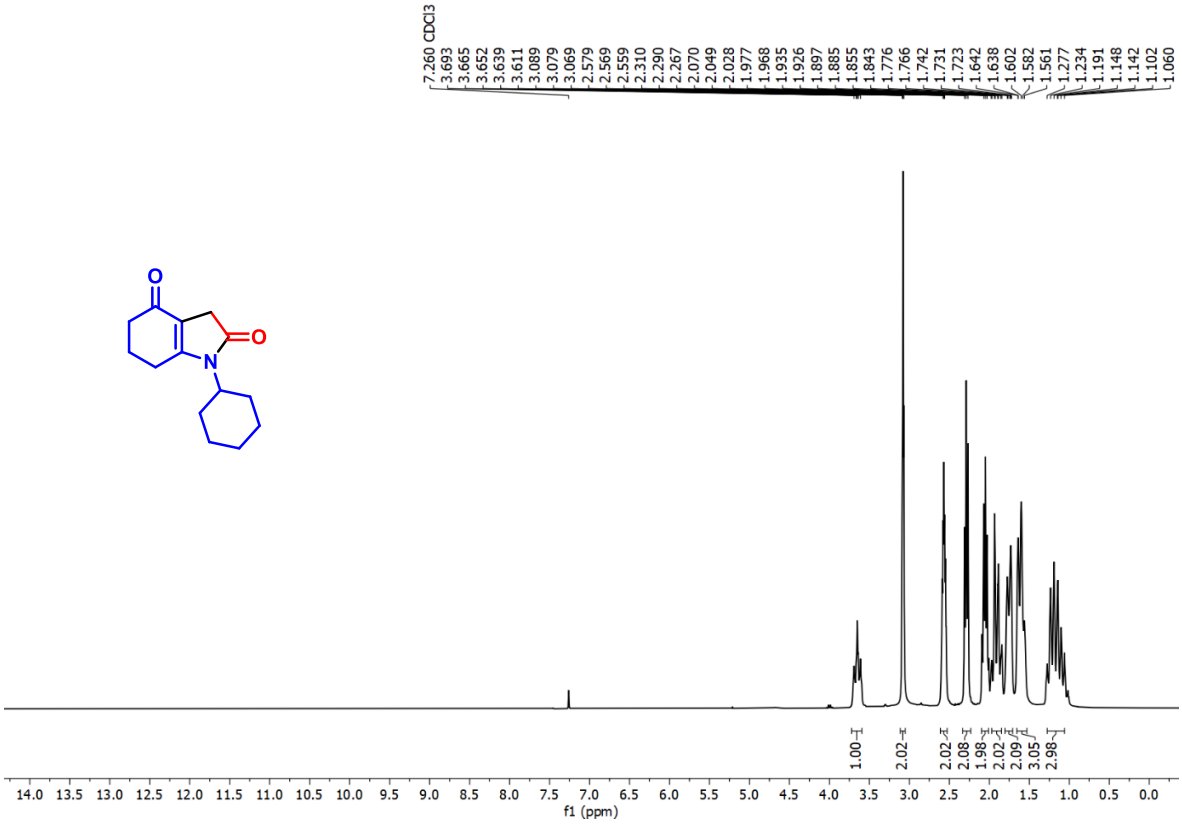
^{13}C {^{1}H} NMR Spectrum of Compound **3j** (75 MHz, $CDCl_3$)



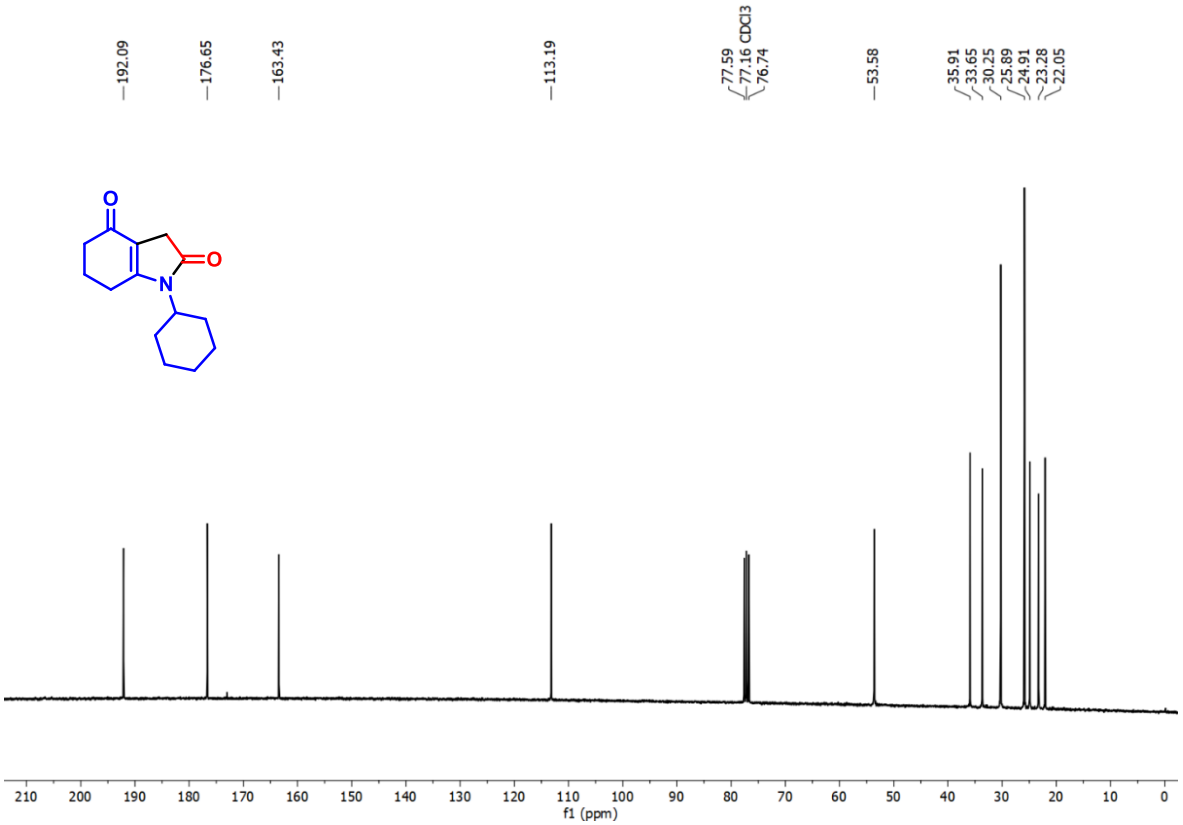
^{1}H NMR Spectrum of Compound **3k** (400 MHz, $CDCl_3$)



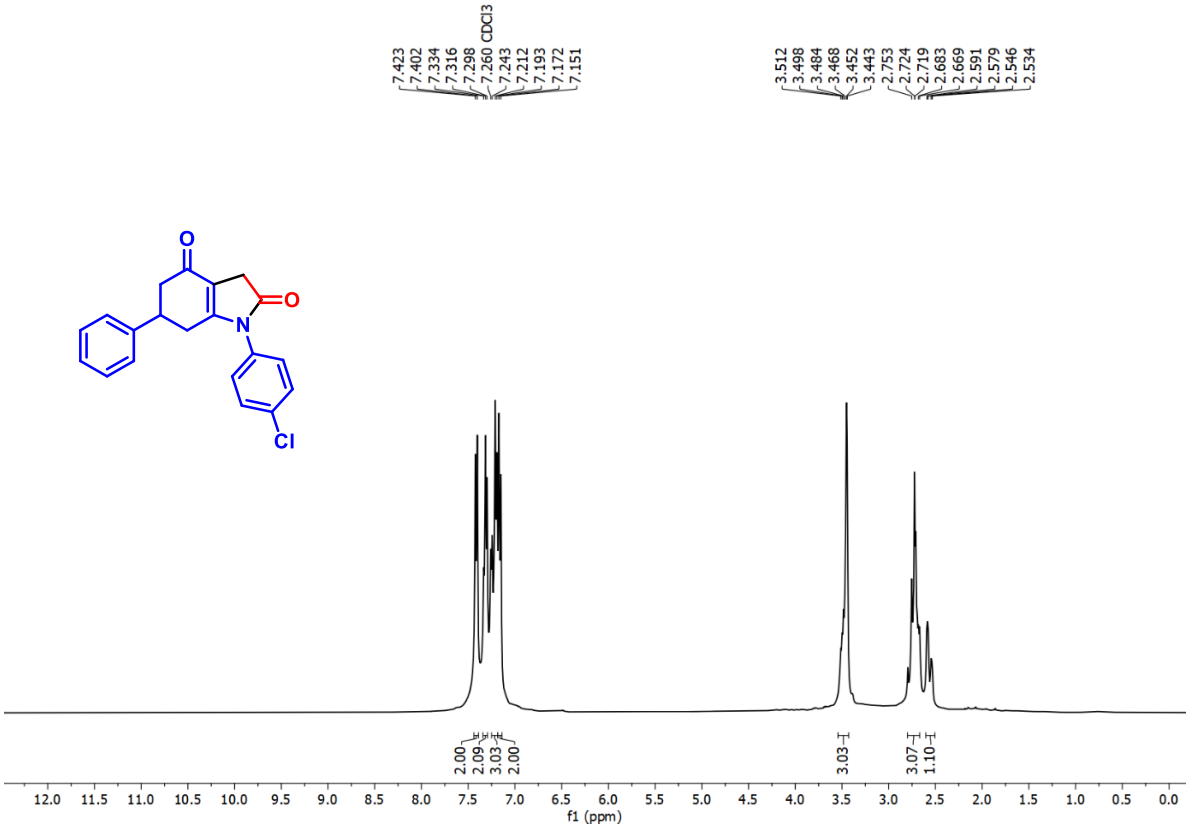
^{13}C {^{1}H} NMR Spectrum of Compound **3k** (100 MHz, $CDCl_3$)



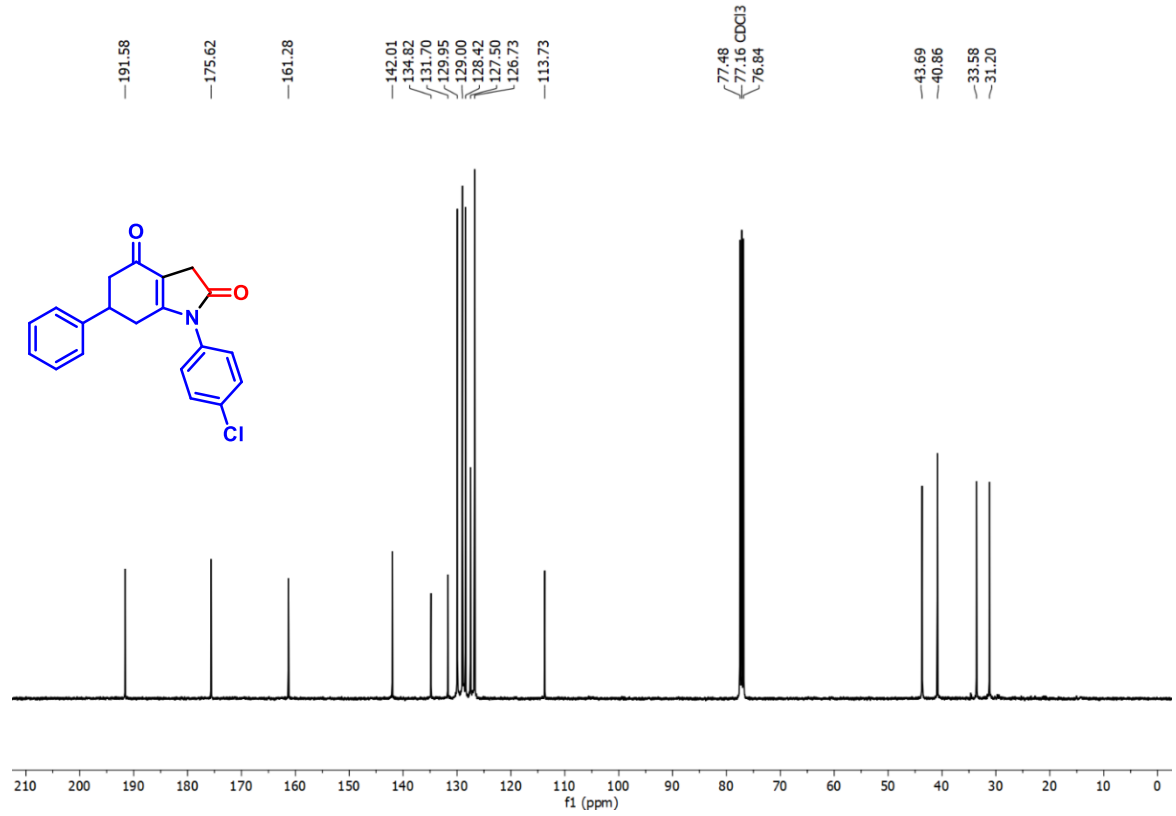
^{1}H NMR Spectrum of Compound **3l** (300 MHz, $CDCl_3$)



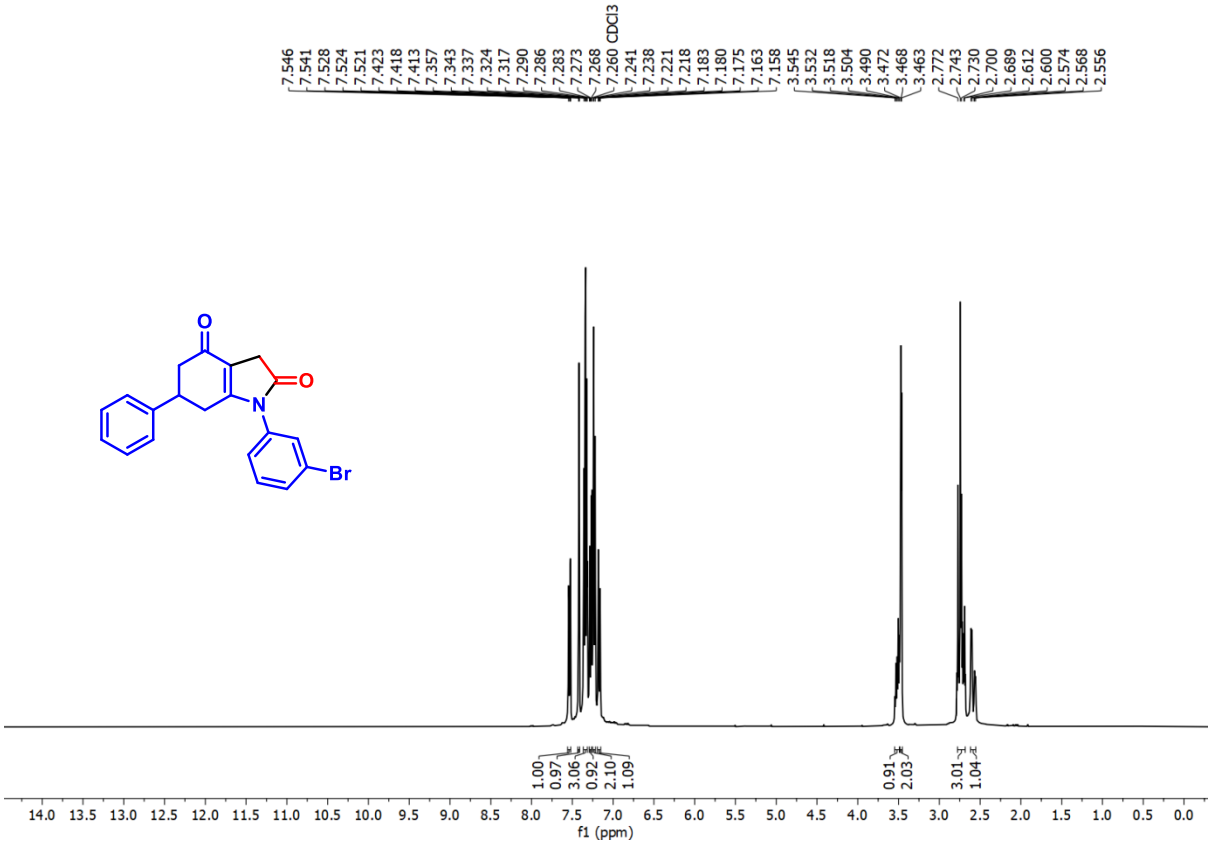
^{13}C {^{1}H} NMR Spectrum of Compound **3l** (75 MHz, $CDCl_3$)



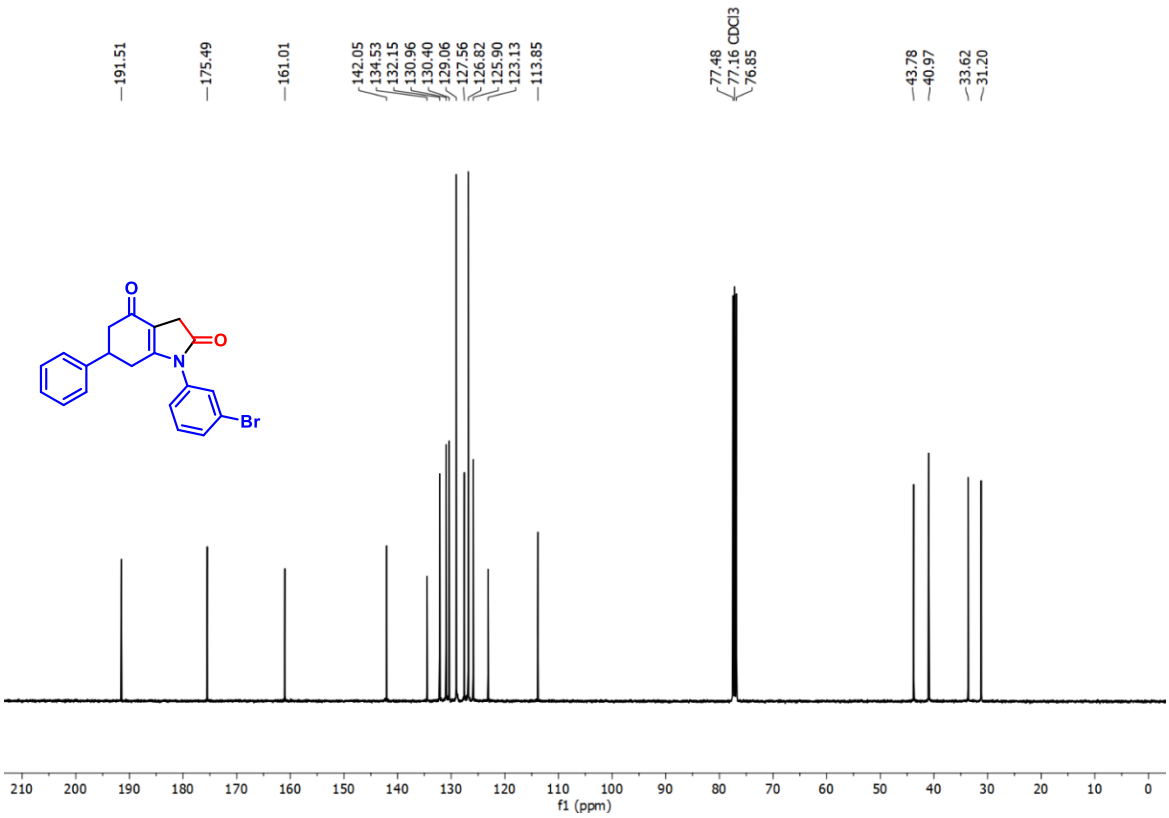
^{1}H NMR Spectrum of Compound **3m** (400 MHz, $CDCl_3$)



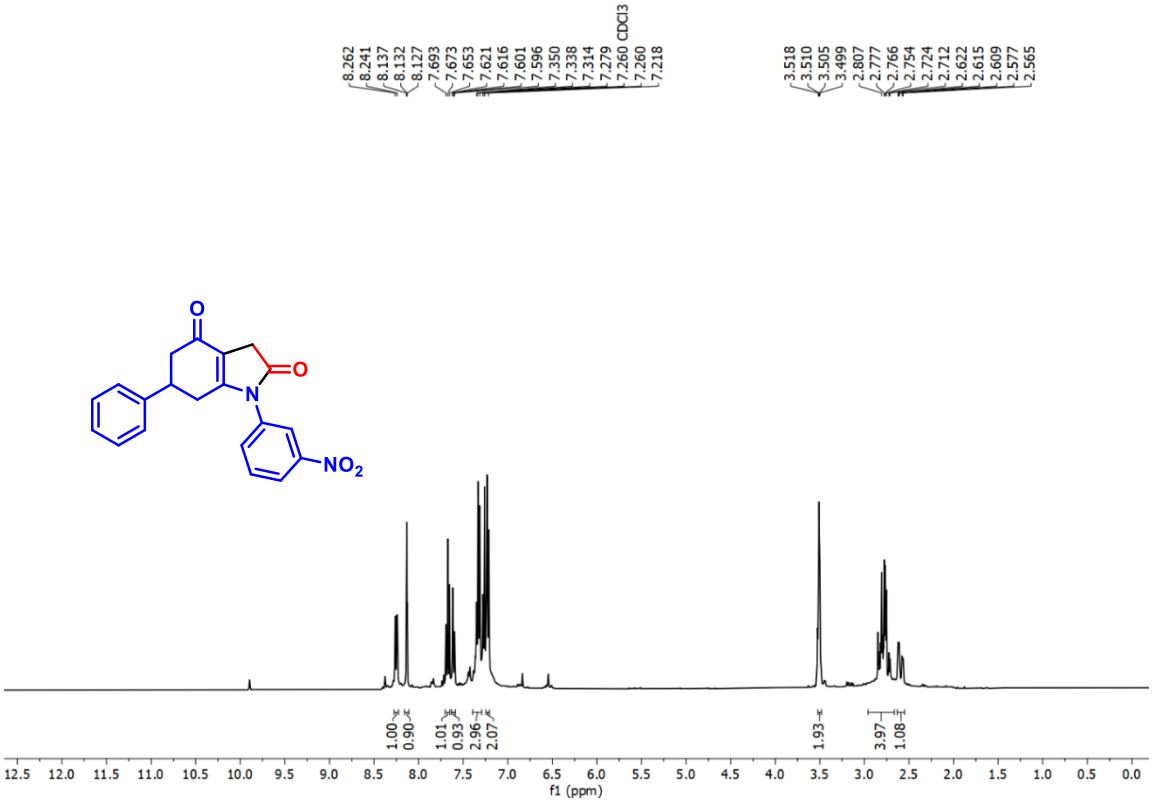
^{13}C {^{1}H} NMR Spectrum of Compound **3m** (100 MHz, $CDCl_3$)



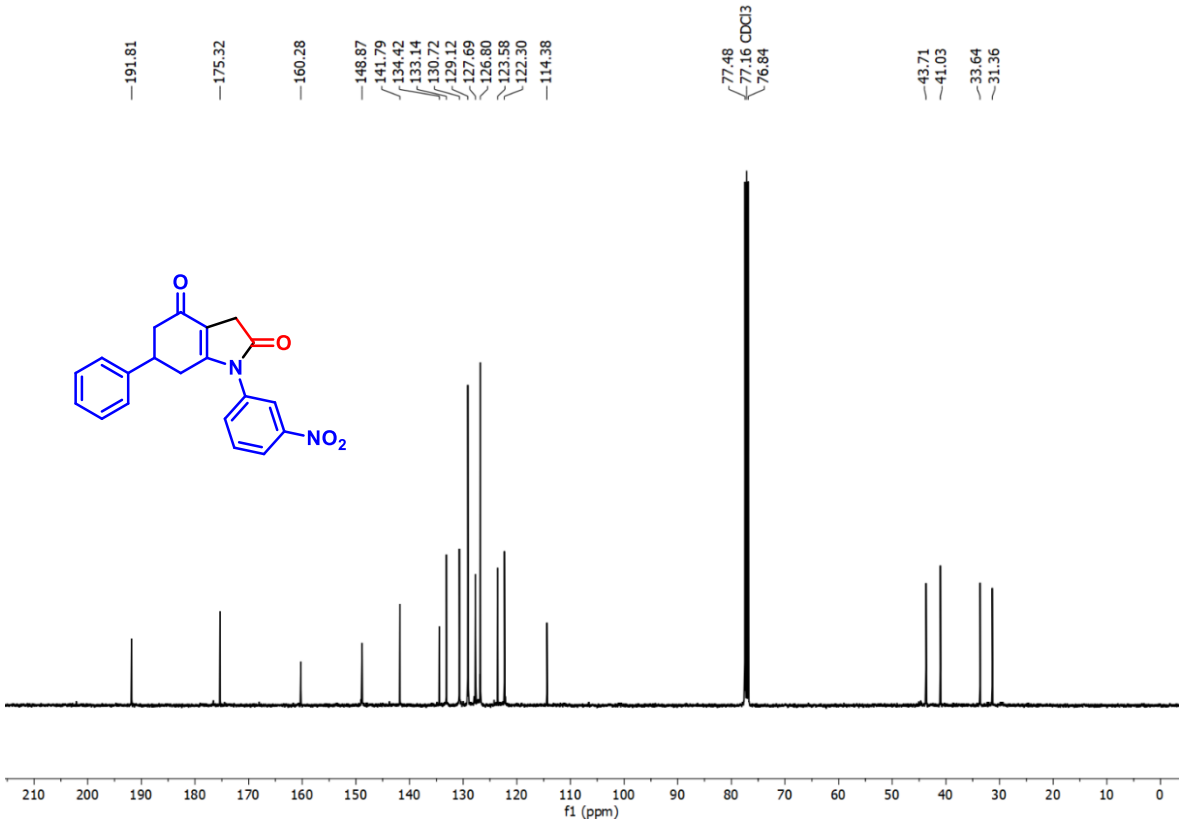
^{1}H NMR Spectrum of Compound **3n** (400 MHz, $CDCl_3$)



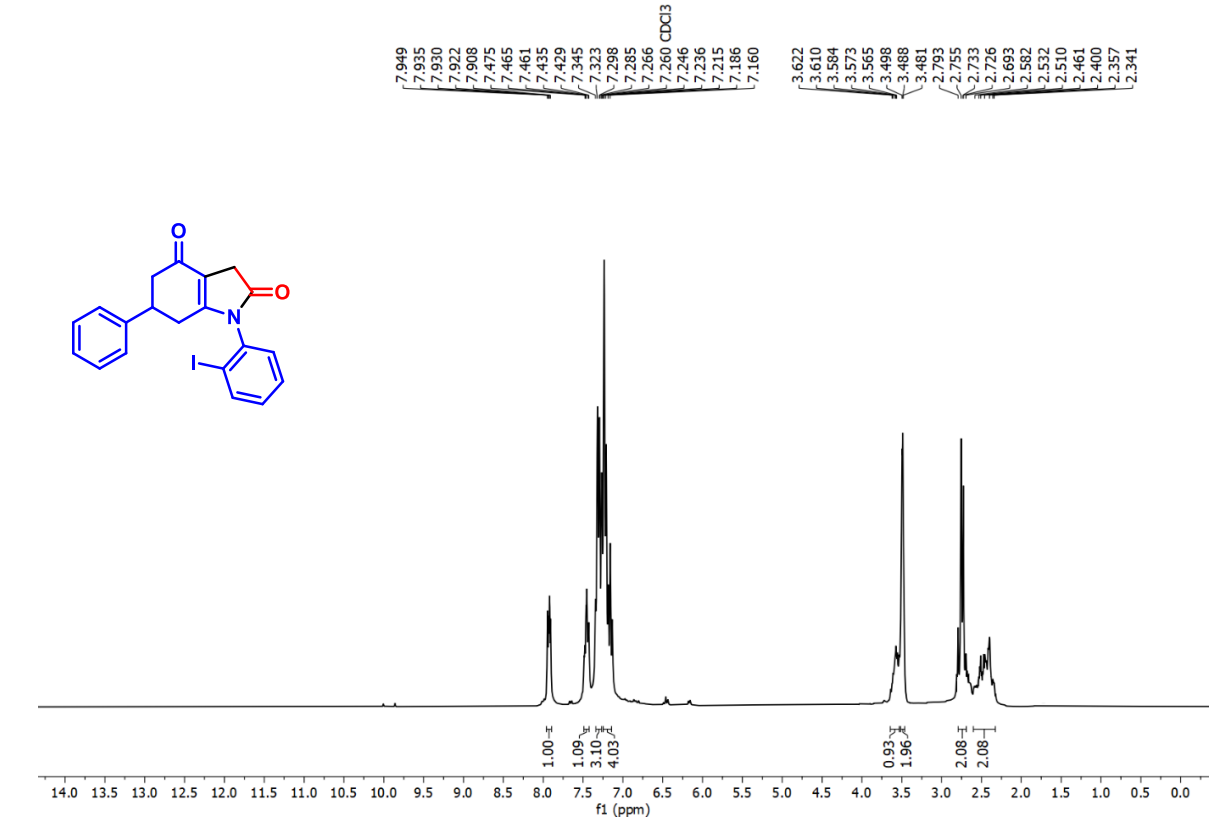
^{13}C {^{1}H} NMR Spectrum of Compound **3n** (75 MHz, $CDCl_3$)



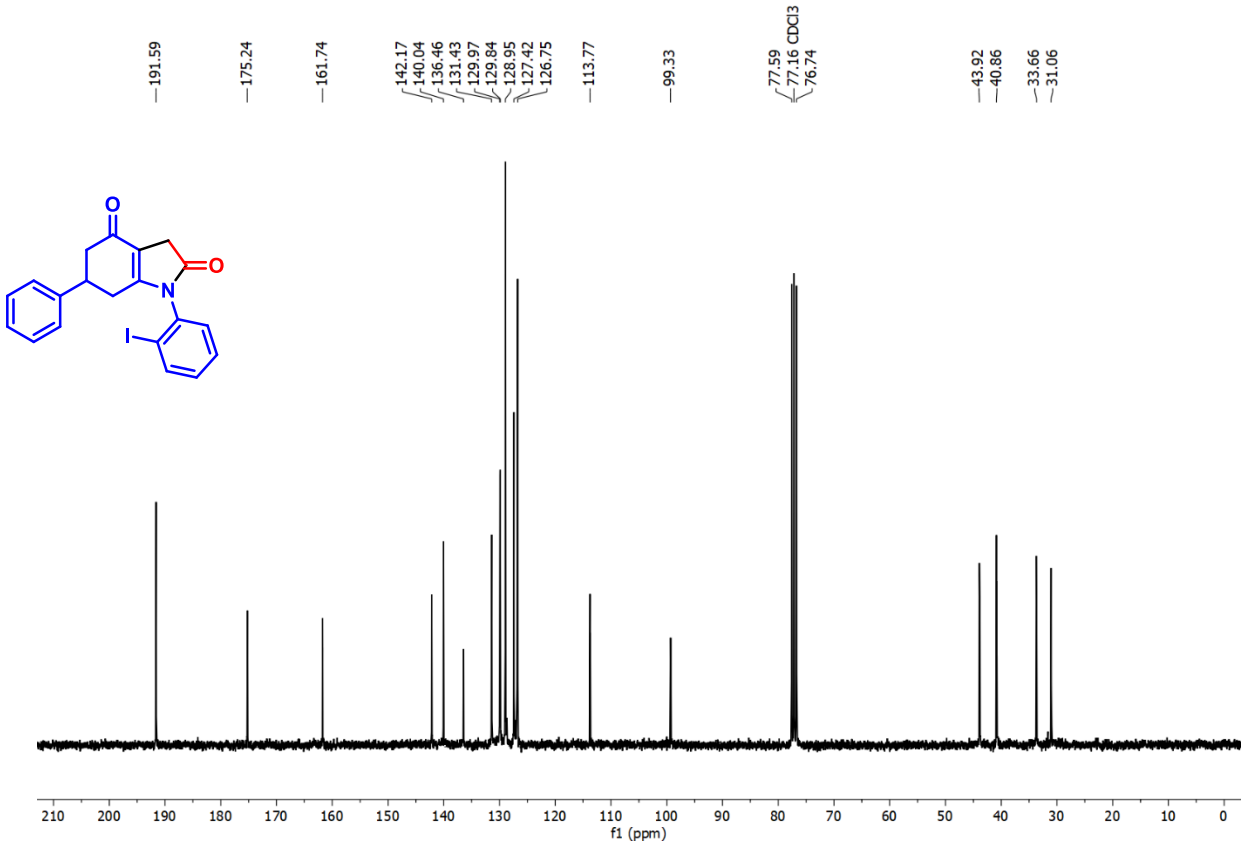
^{1}H NMR Spectrum of Compound **3o** (400 MHz, $CDCl_3$)



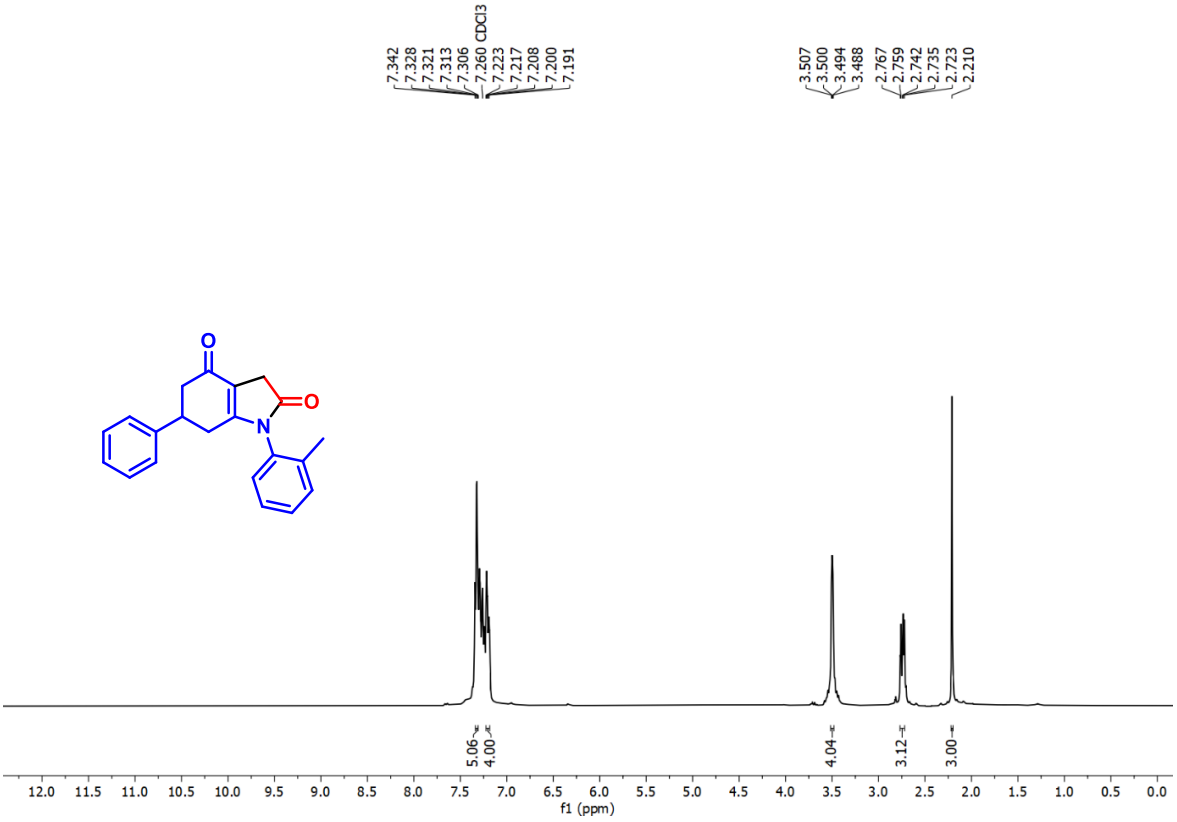
^{13}C {^{1}H} NMR Spectrum of Compound **3o** (100 MHz, $CDCl_3$)



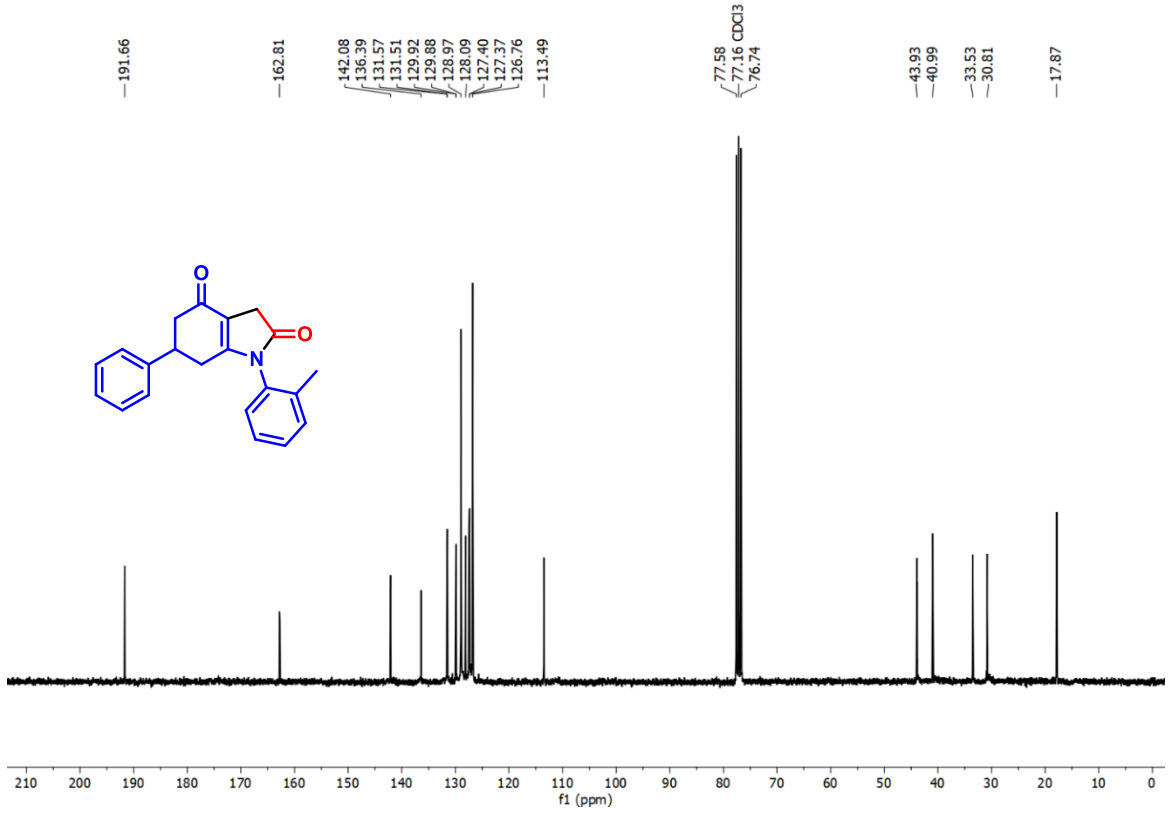
^{1}H NMR Spectrum of Compound **3p** (300 MHz, $CDCl_3$)



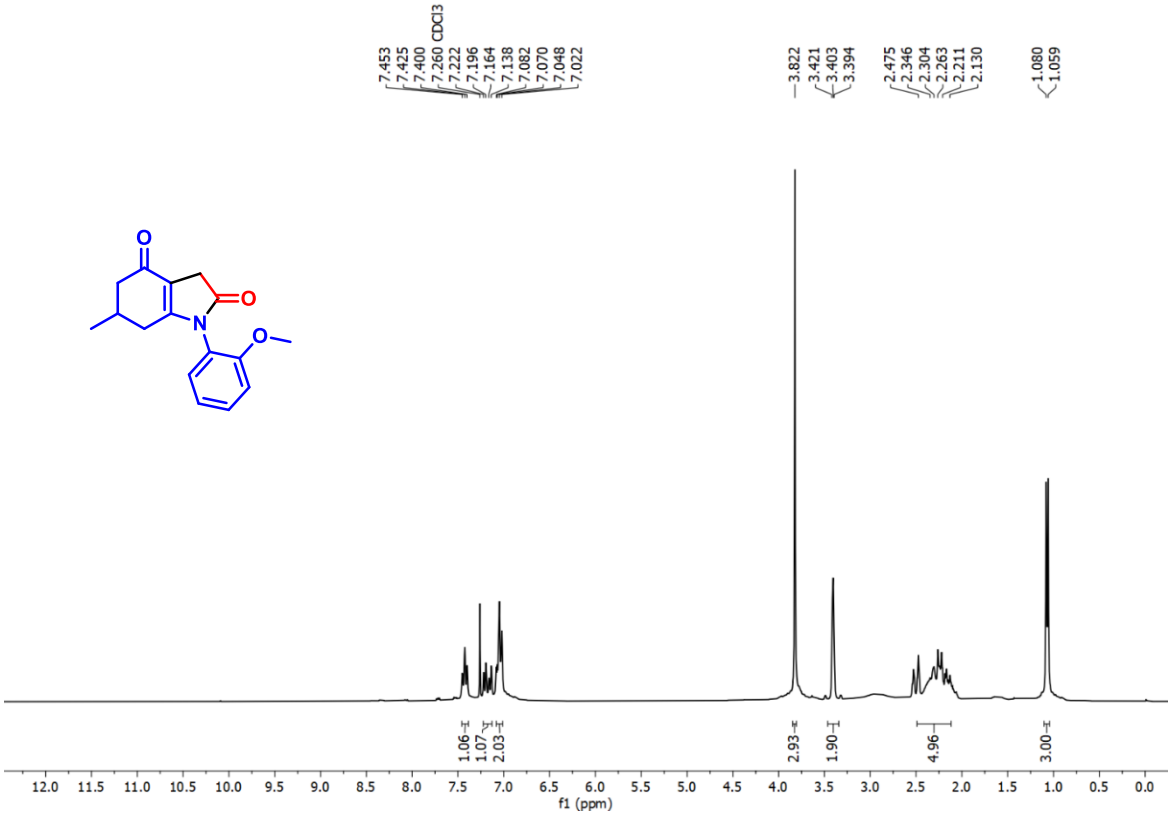
^{13}C {^{1}H} NMR Spectrum of Compound **3p** (75 MHz, $CDCl_3$)



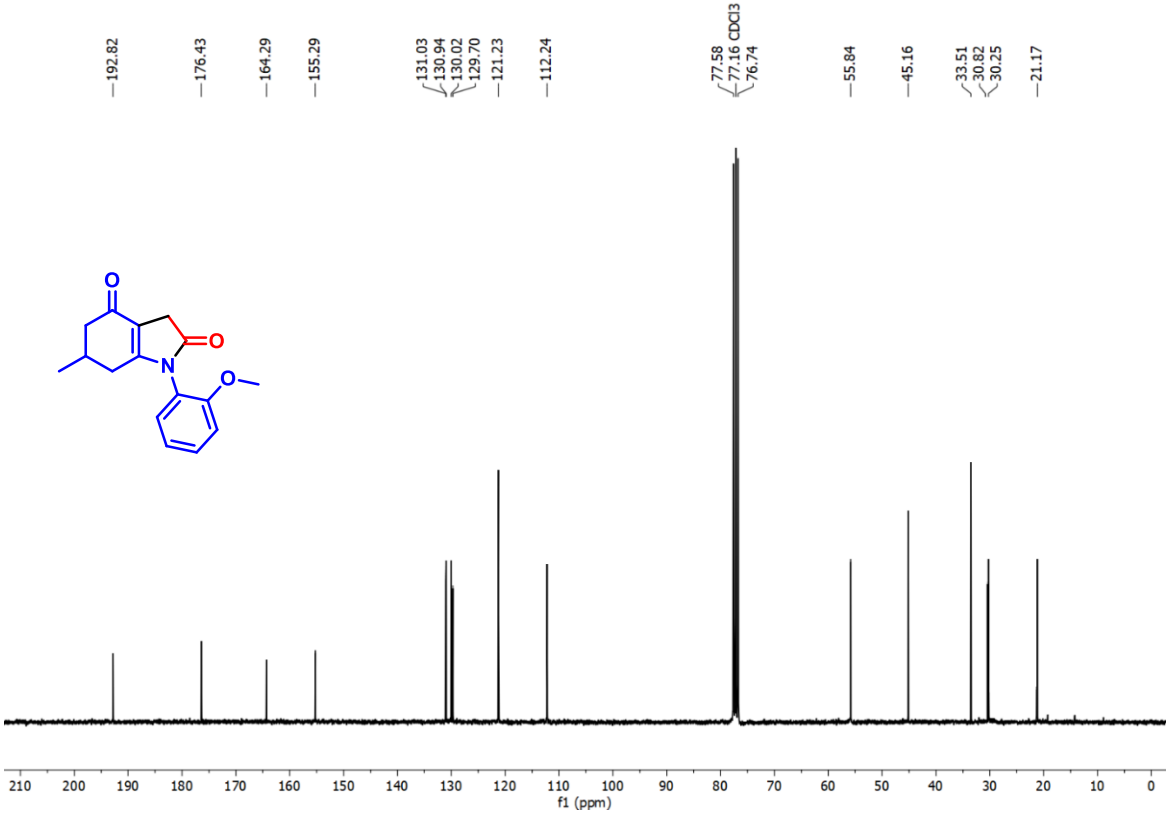
^{1}H NMR Spectrum of Compound **3q** (300 MHz, $CDCl_3$)



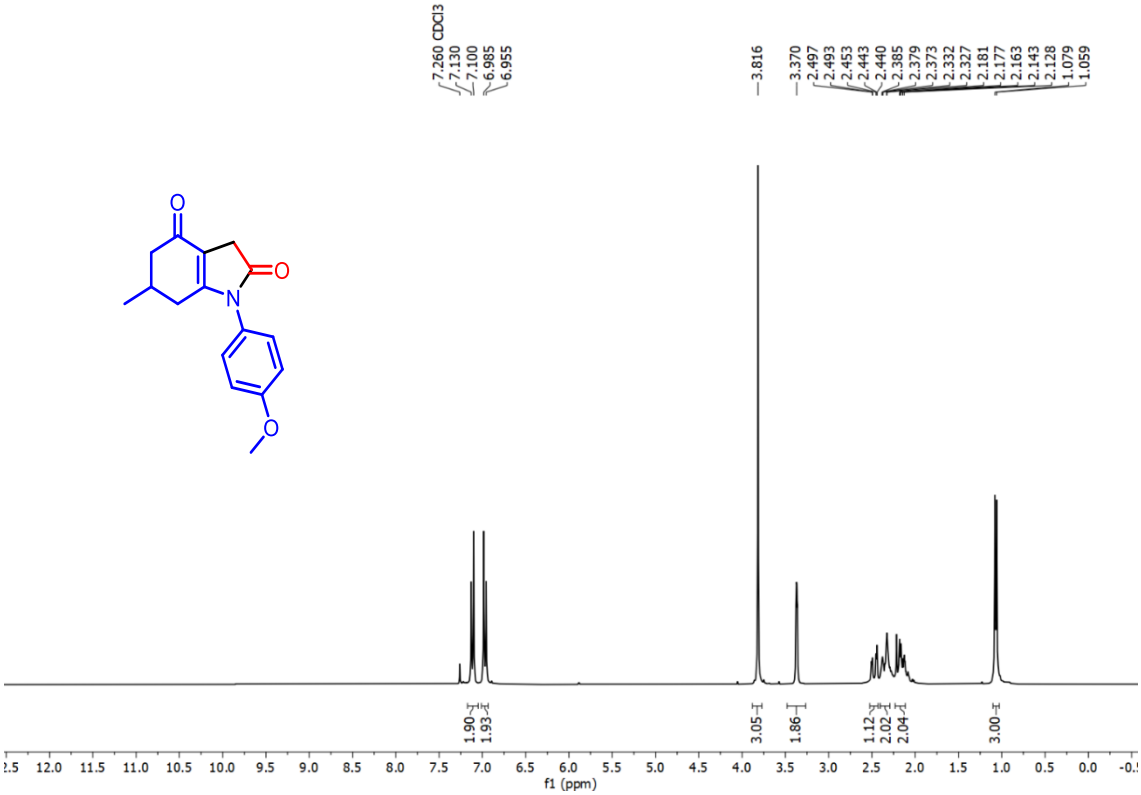
^{13}C {^{1}H} NMR Spectrum of Compound **3q** (75 MHz, $CDCl_3$)



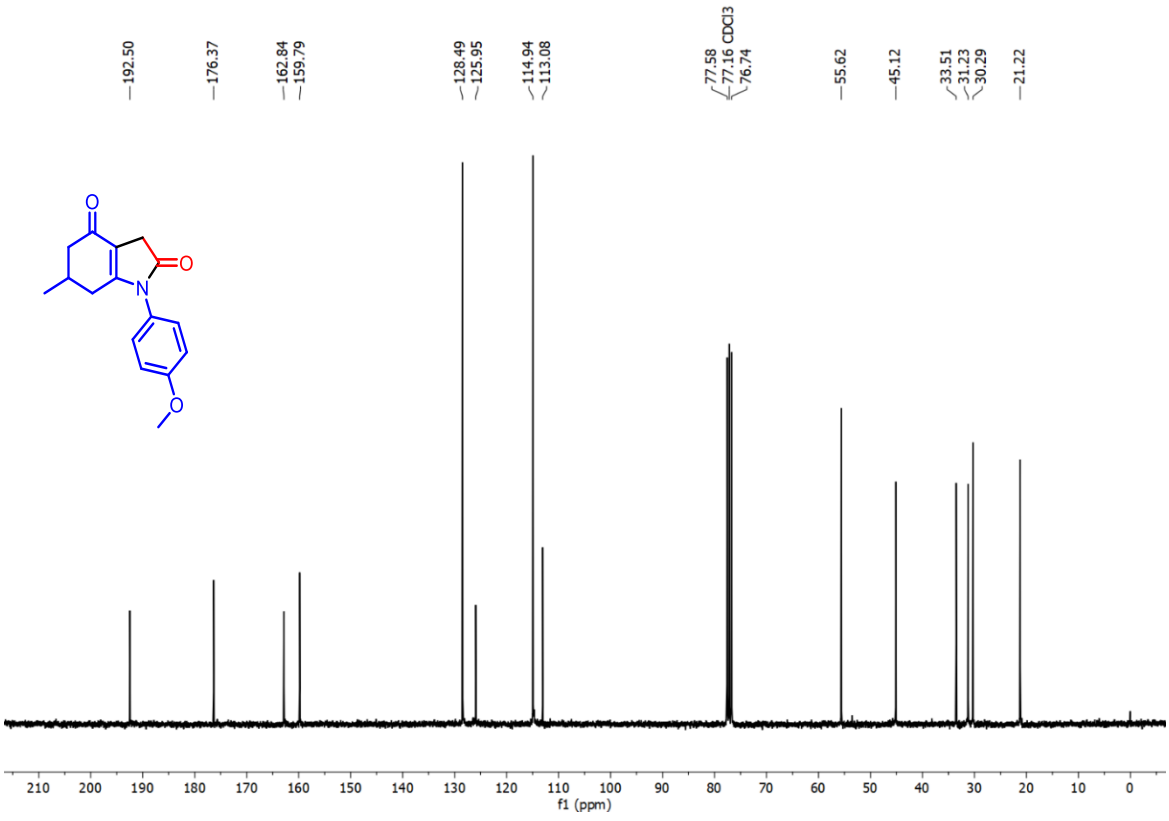
^{1}H NMR Spectrum of Compound **3r** (300 MHz, $CDCl_3$)



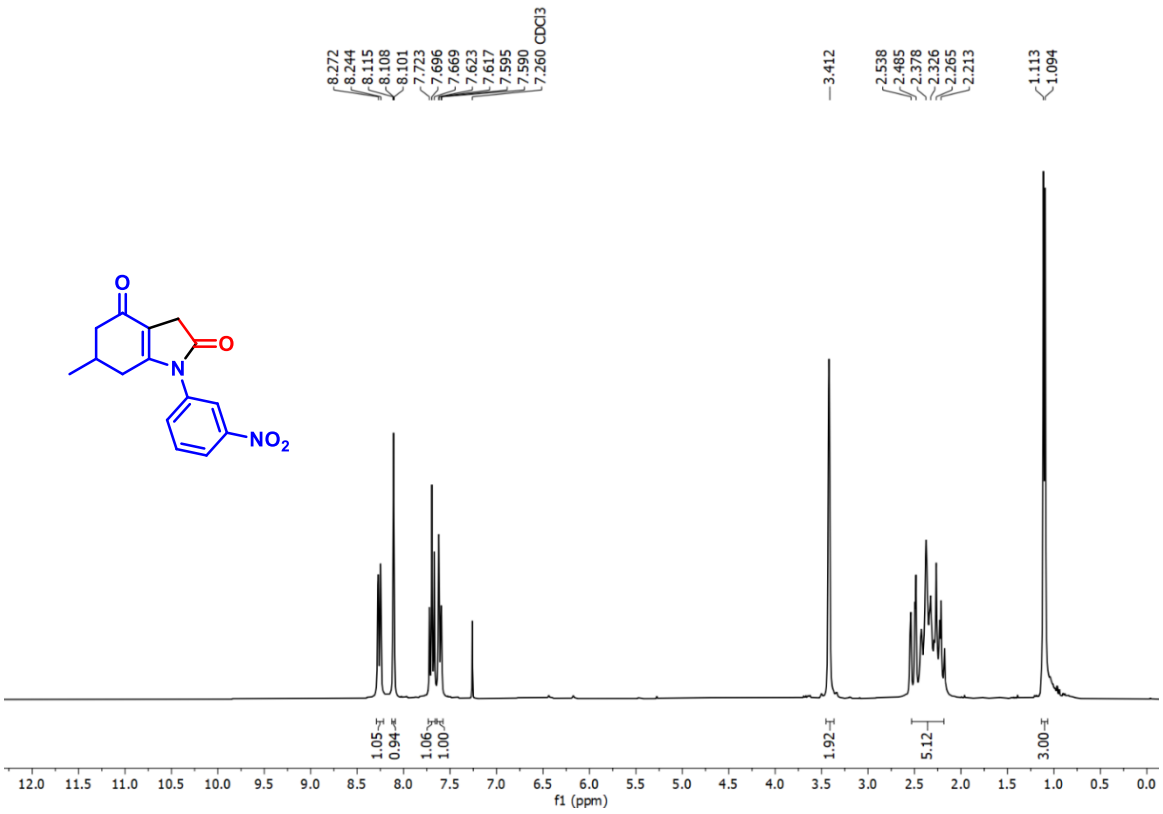
^{13}C {^{1}H}NMR Spectrum of Compound **3r** (75 MHz, $CDCl_3$



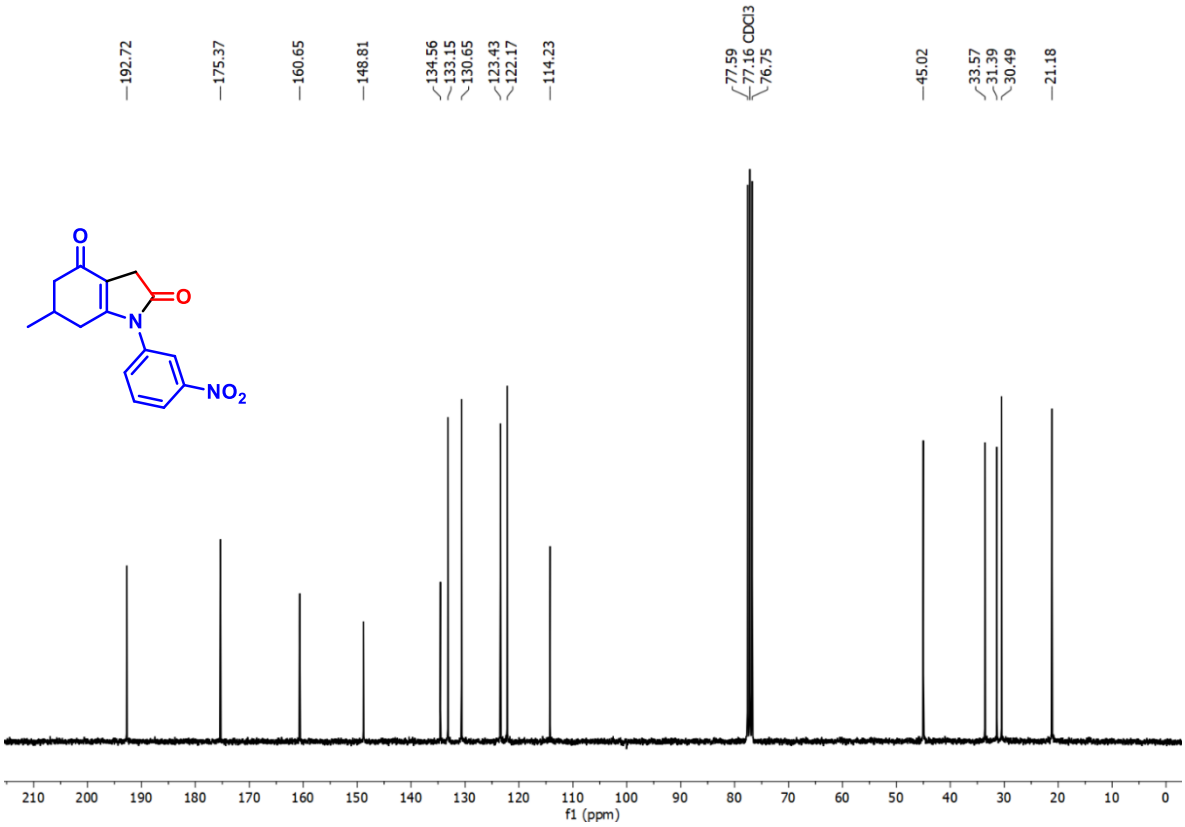
^{1}H NMR Spectrum of Compound **3s** (300 MHz, $CDCl_3$)



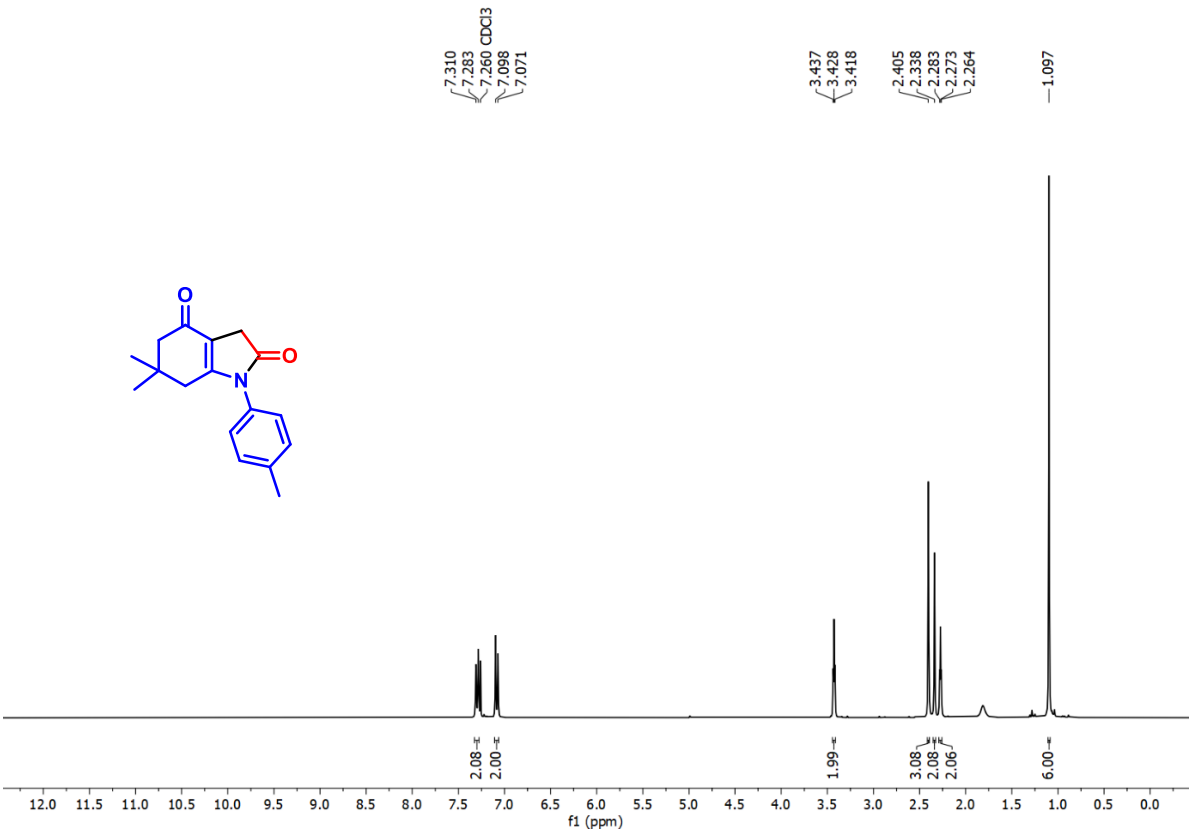
^{13}C {^{1}H} NMR Spectrum of Compound **3s** (75 MHz, $CDCl_3$)



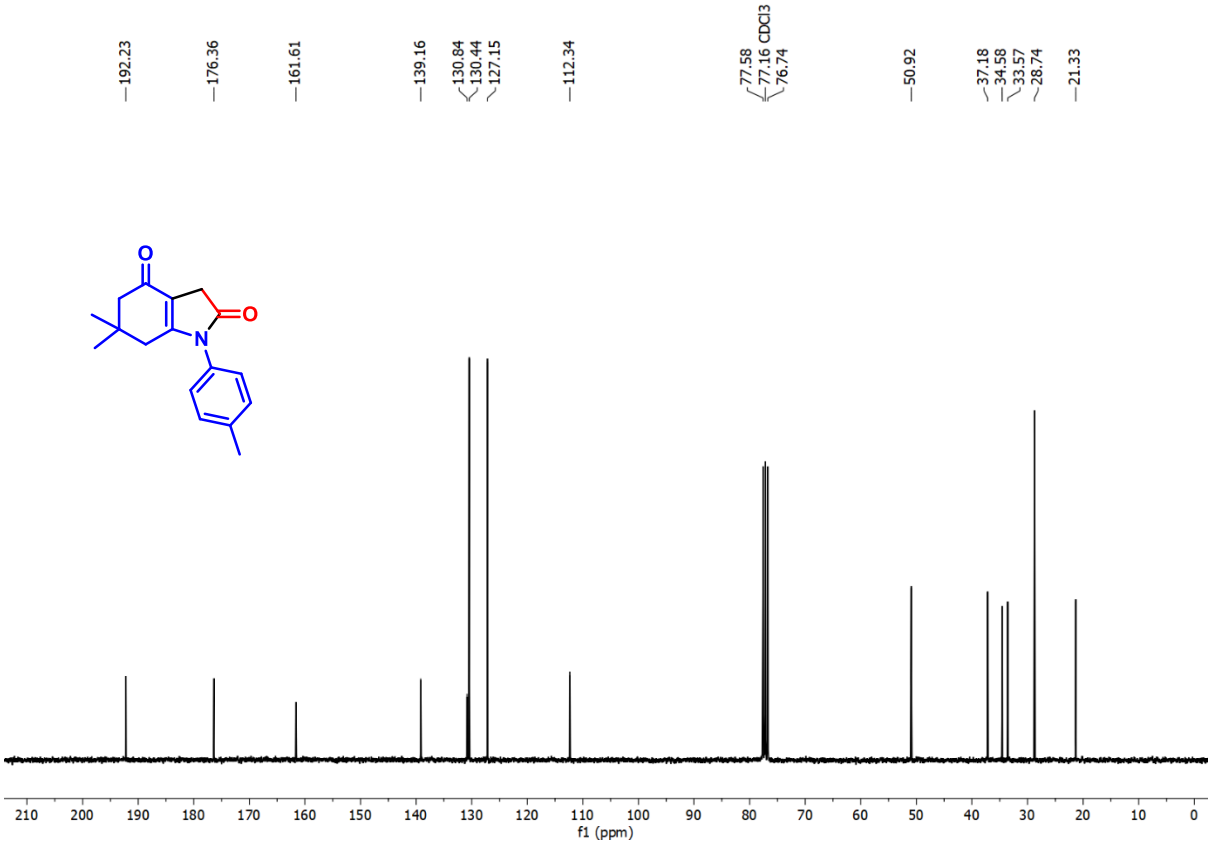
^{1}H NMR Spectrum of Compound **3t** (300 MHz, $CDCl_3$)



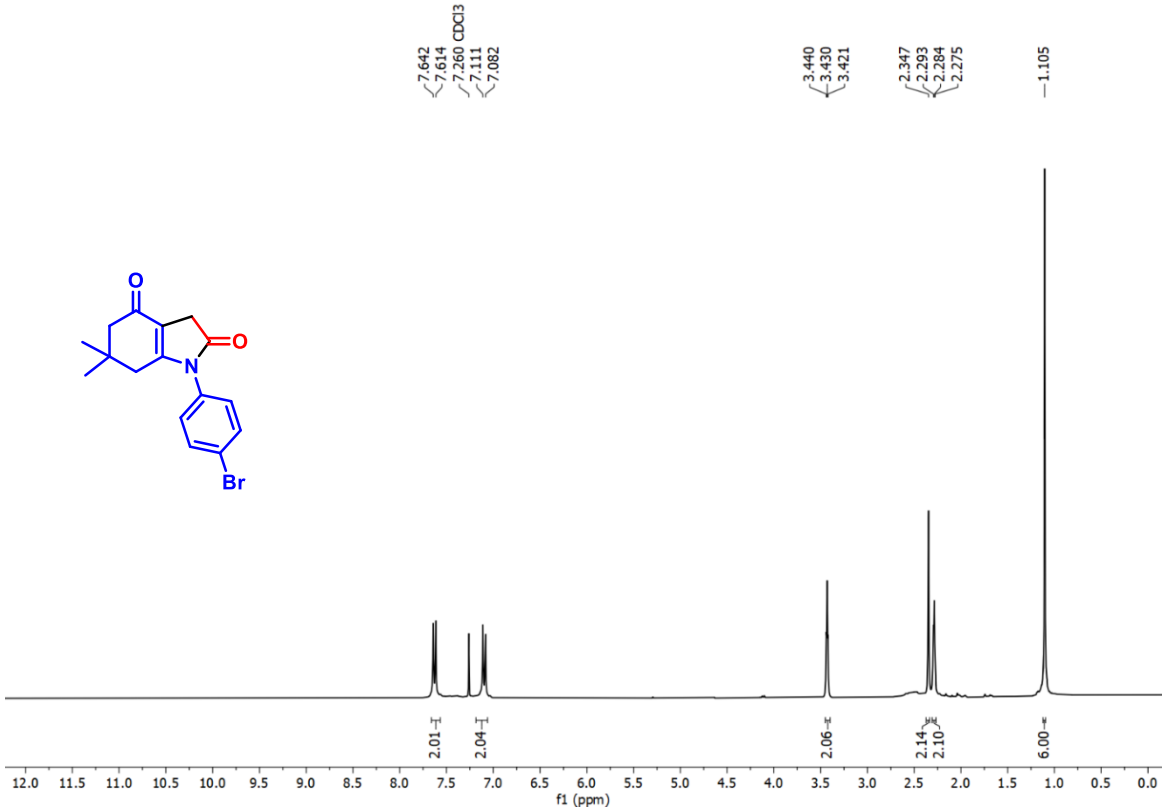
^{13}C {^{1}H} NMR Spectrum of Compound **3t** (75 MHz, $CDCl_3$)



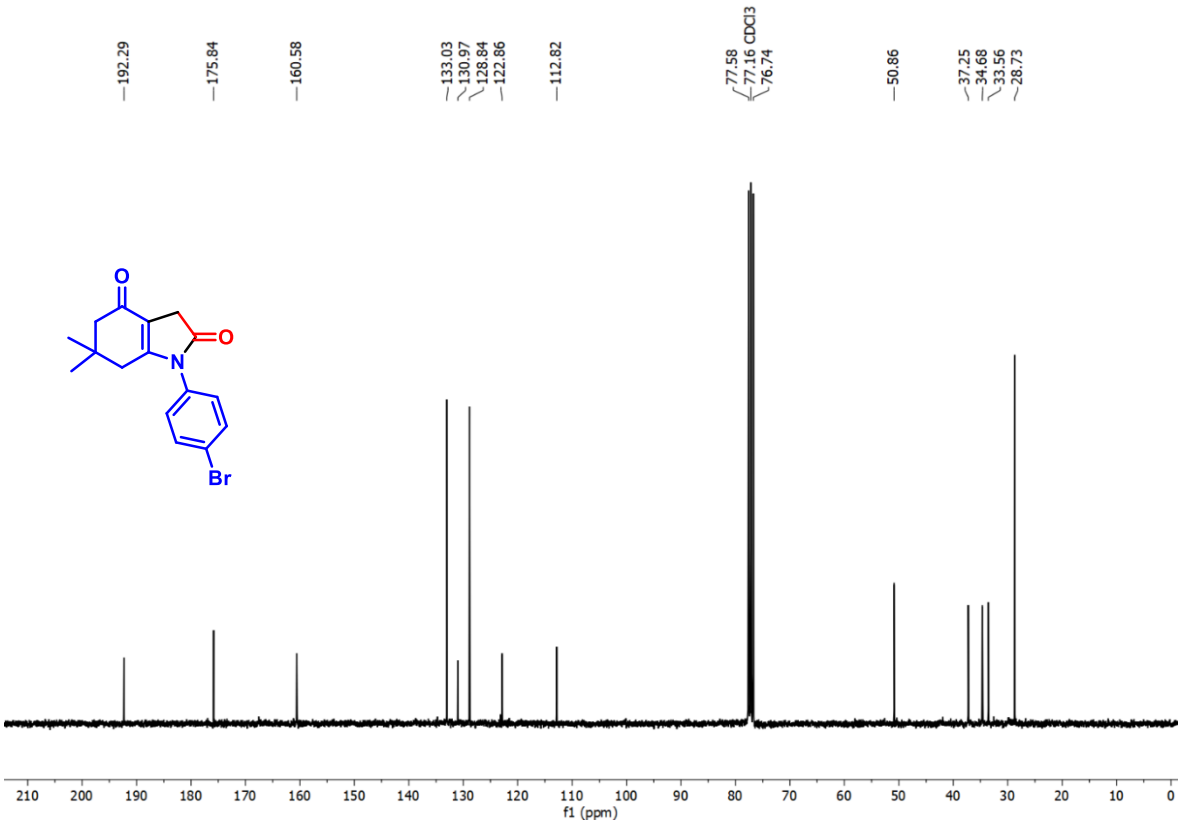
^{1}H NMR Spectrum of Compound **3u** (300 MHz, $CDCl_3$)



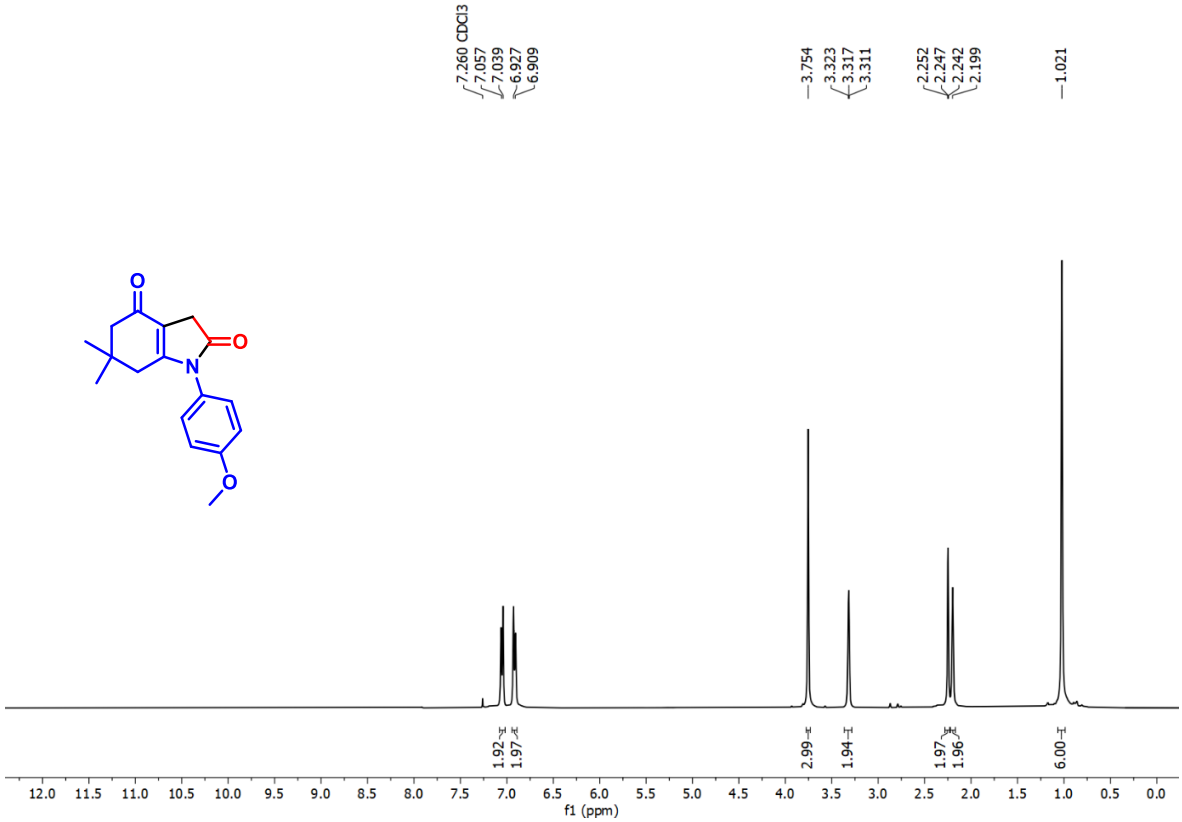
^{13}C {^{1}H} NMR Spectrum of Compound **3u** (75 MHz, $CDCl_3$)



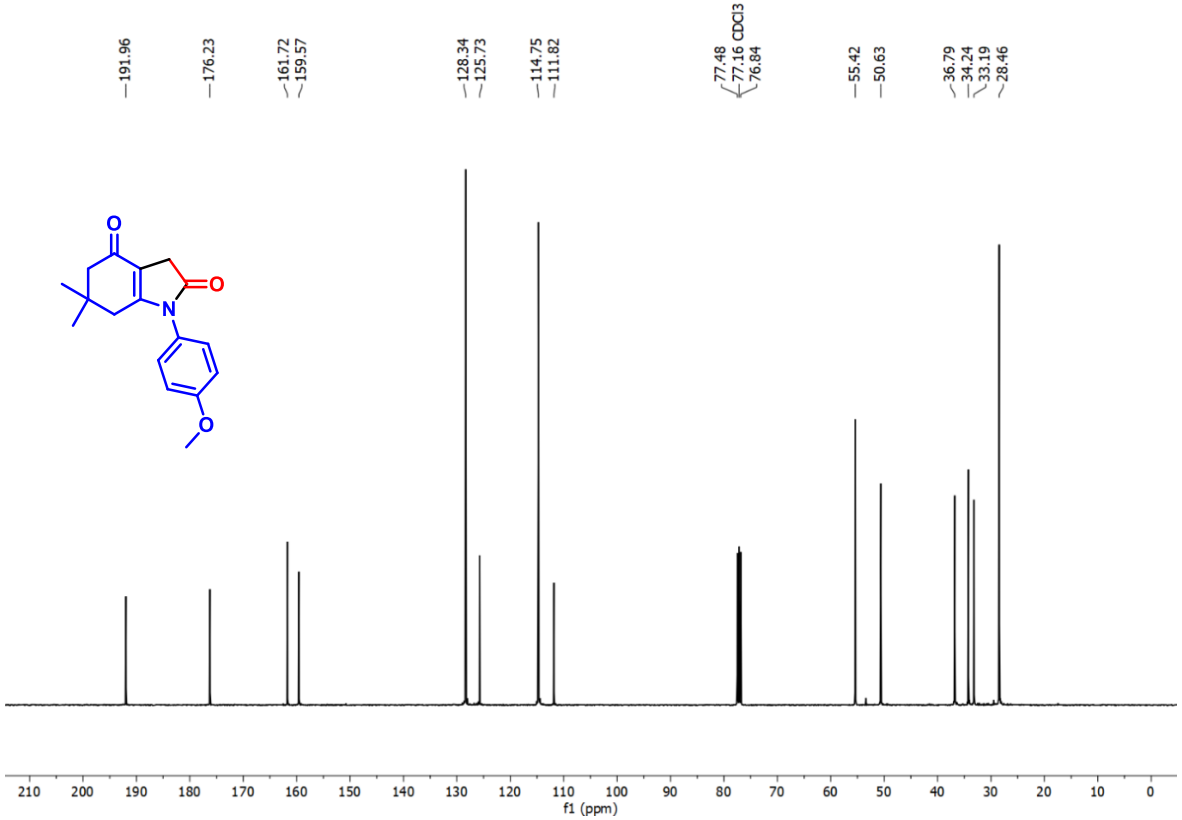
^{1}H NMR Spectrum of Compound **3v** (300 MHz, $CDCl_3$)



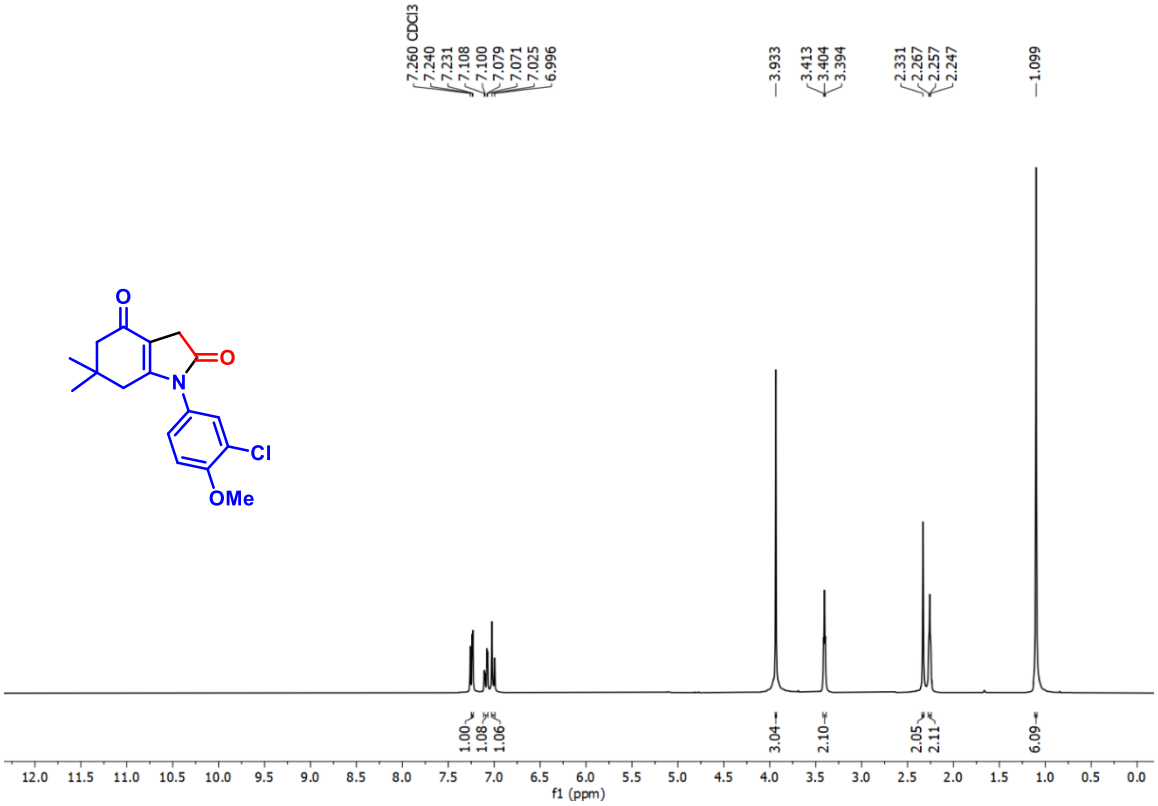
^{13}C {^{1}H} NMR Spectrum of Compound **3v** (75 MHz, $CDCl_3$)



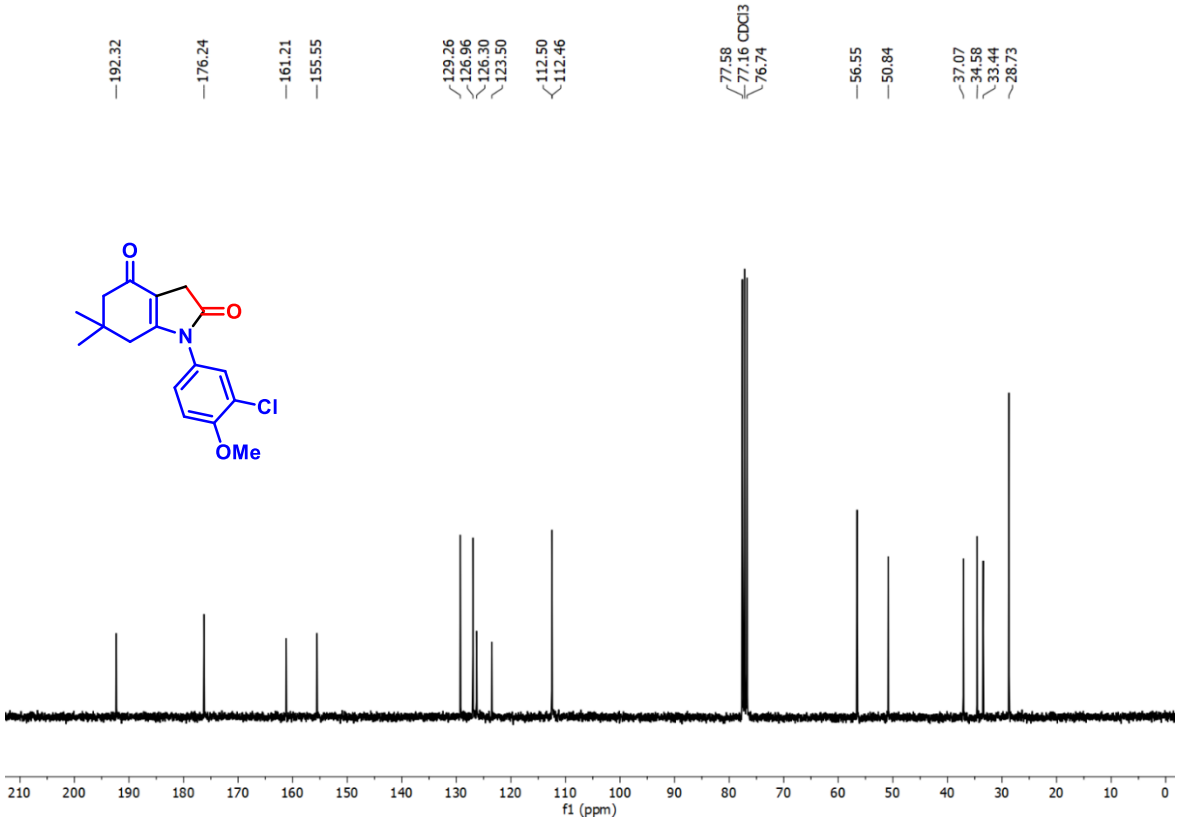
^{1}H NMR Spectrum of Compound **3w** (400 MHz, $CDCl_3$)



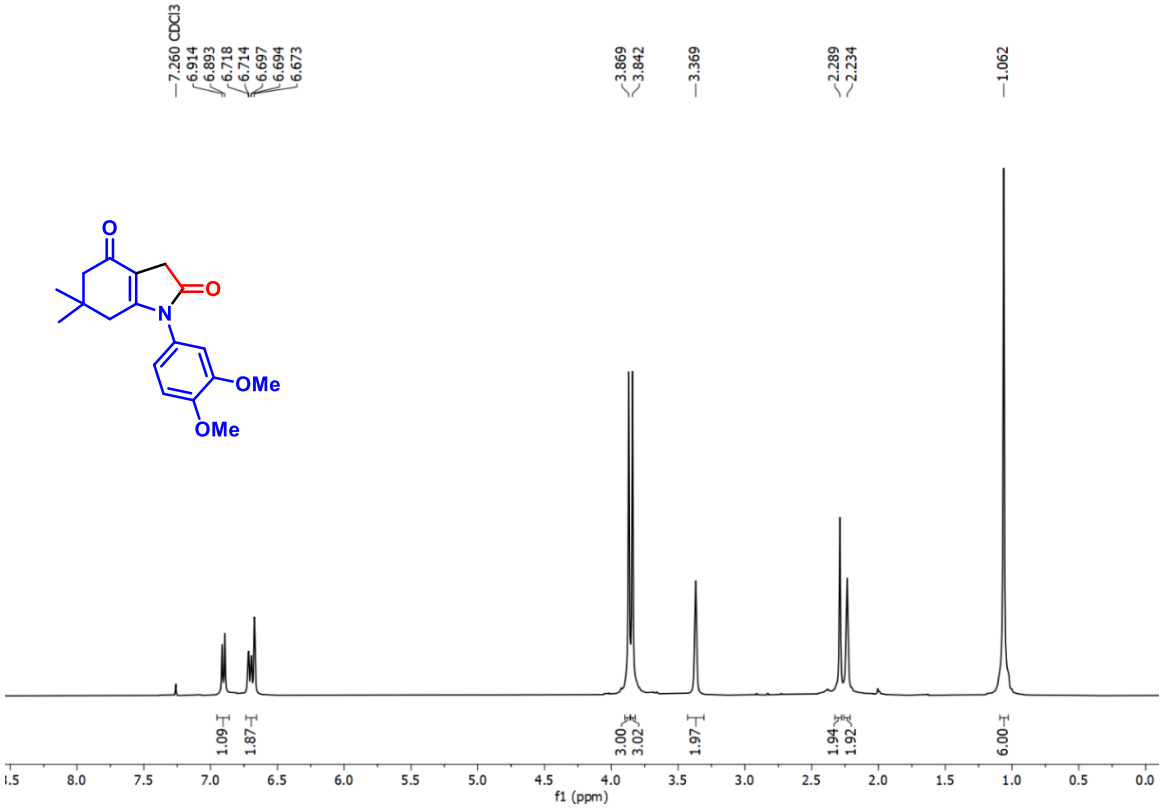
^{13}C {^{1}H} NMR Spectrum of Compound **3w** (100 MHz, $CDCl_3$)



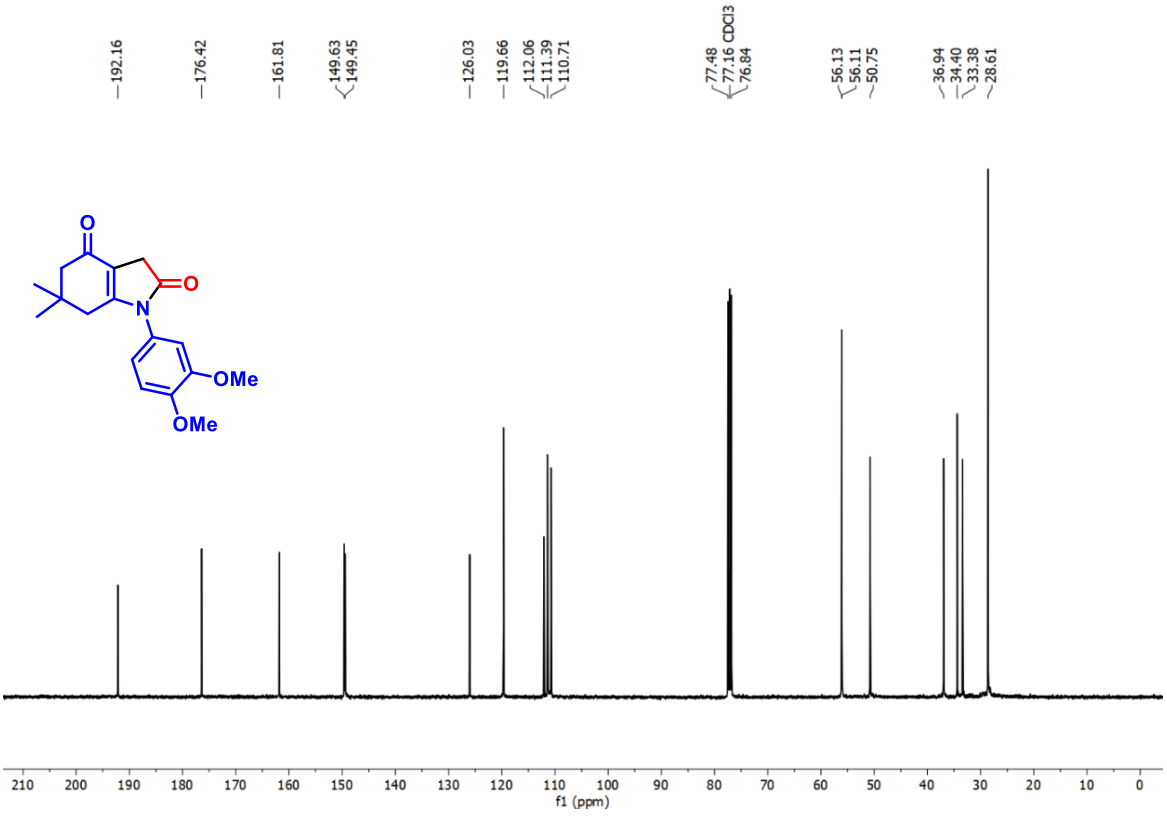
^{1}H NMR Spectrum of Compound **3x** (300 MHz, $CDCl_3$)



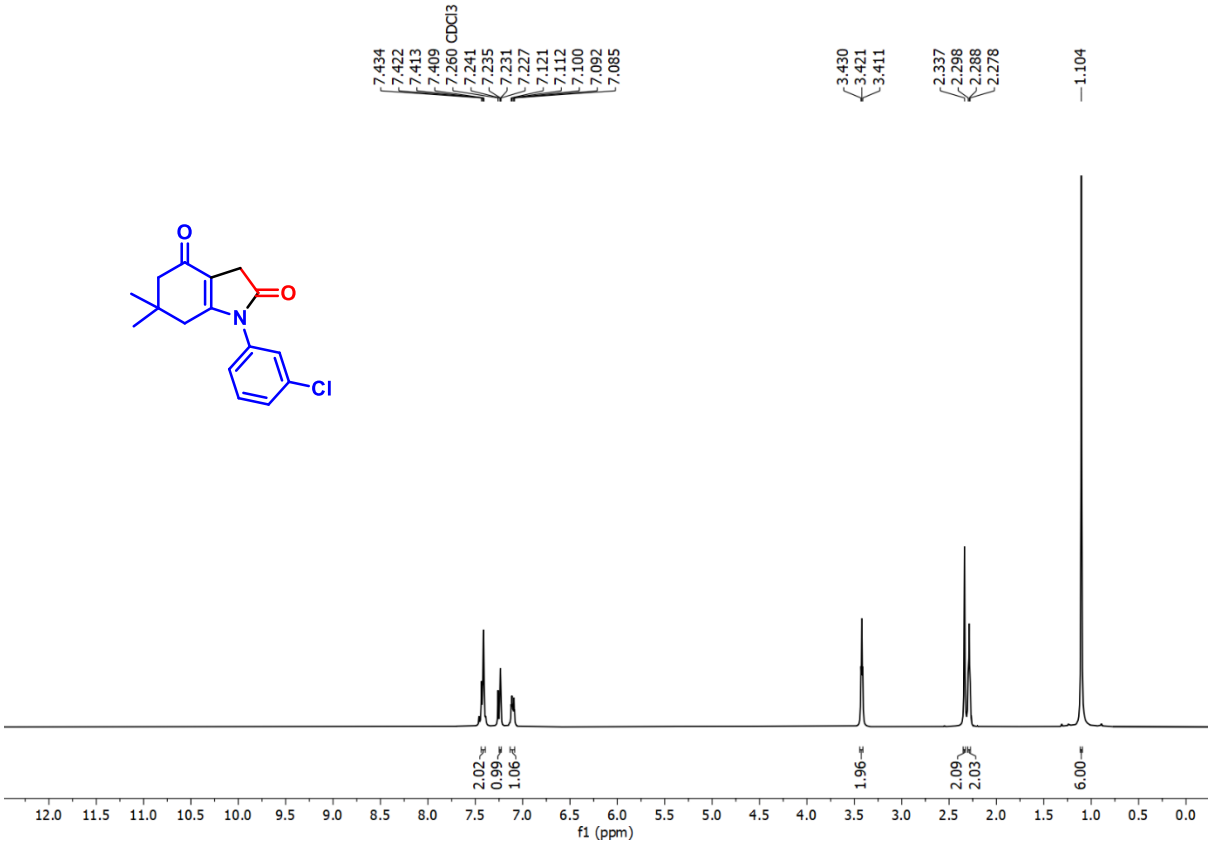
^{13}C {^{1}H} NMR Spectrum of Compound **3x** (75 MHz, $CDCl_3$)



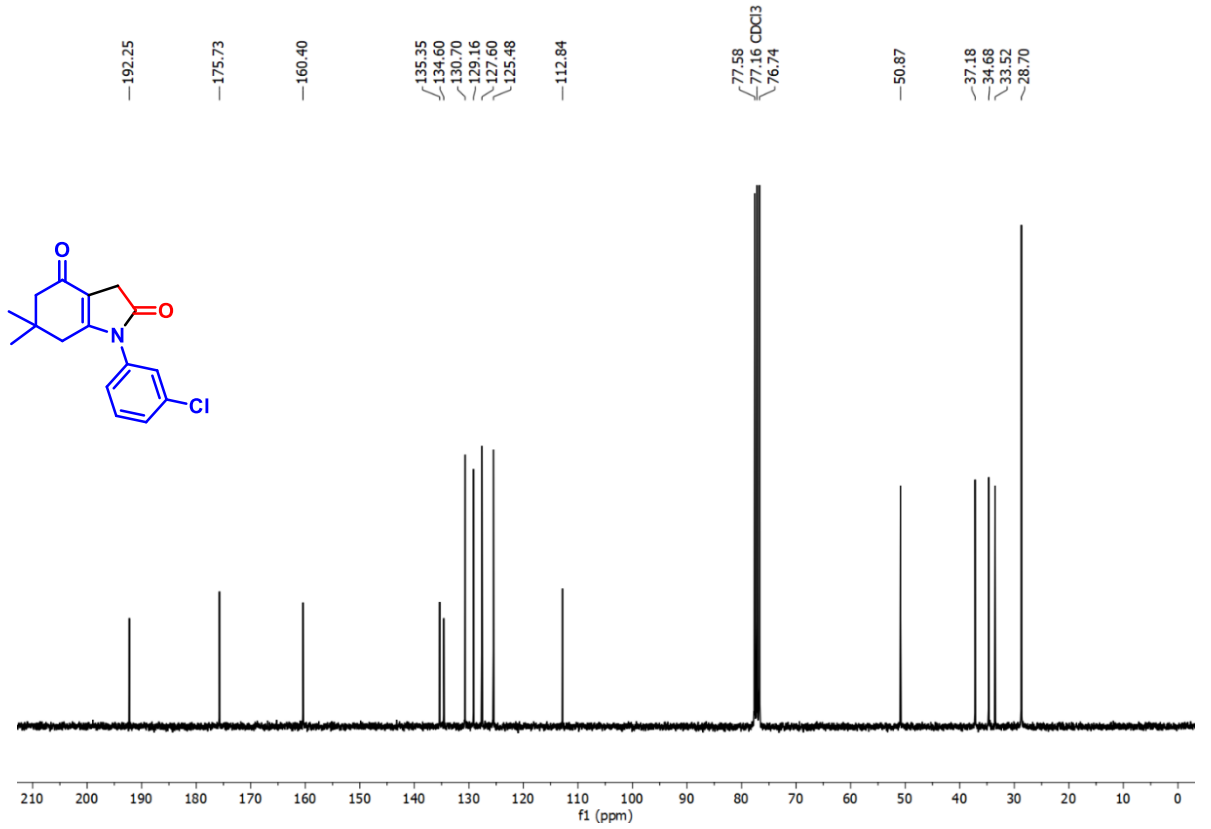
^{1}H NMR Spectrum of Compound **3y** (400 MHz, $CDCl_3$)



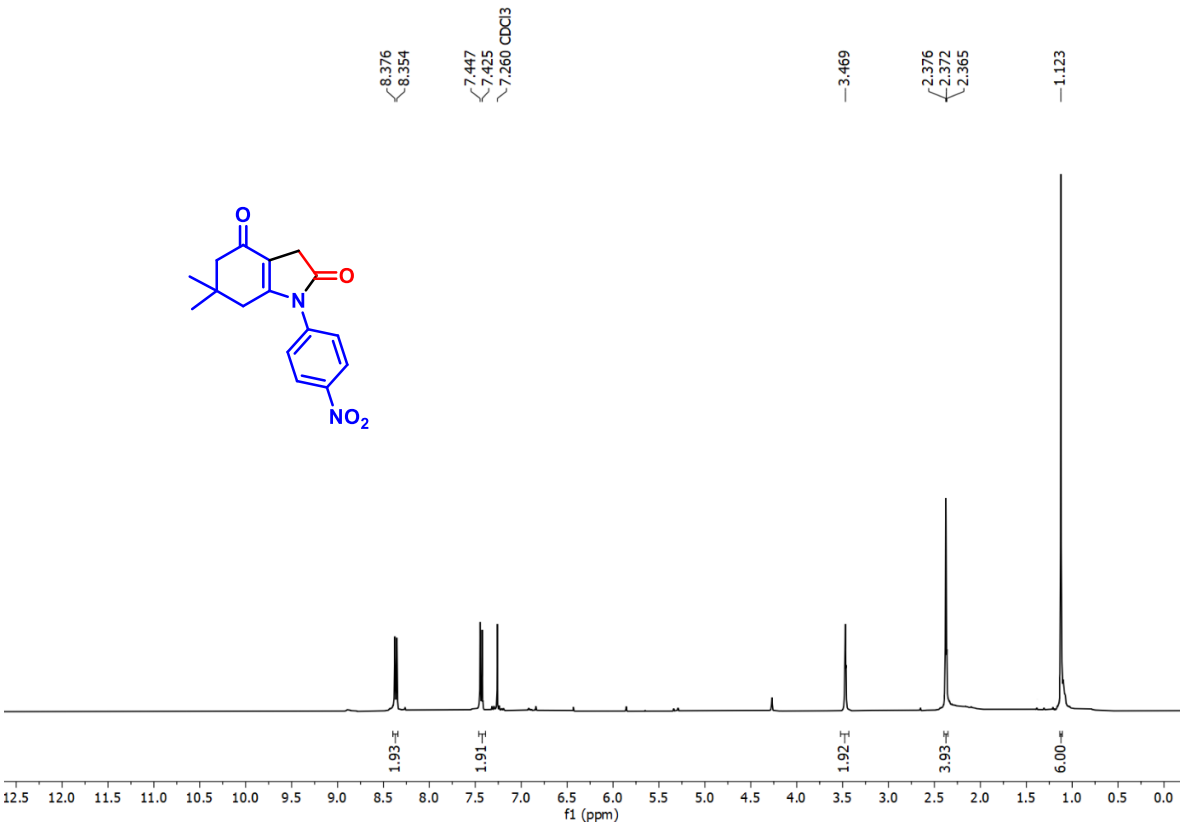
^{13}C {^{1}H} NMR Spectrum of Compound **3y** (100 MHz, $CDCl_3$)



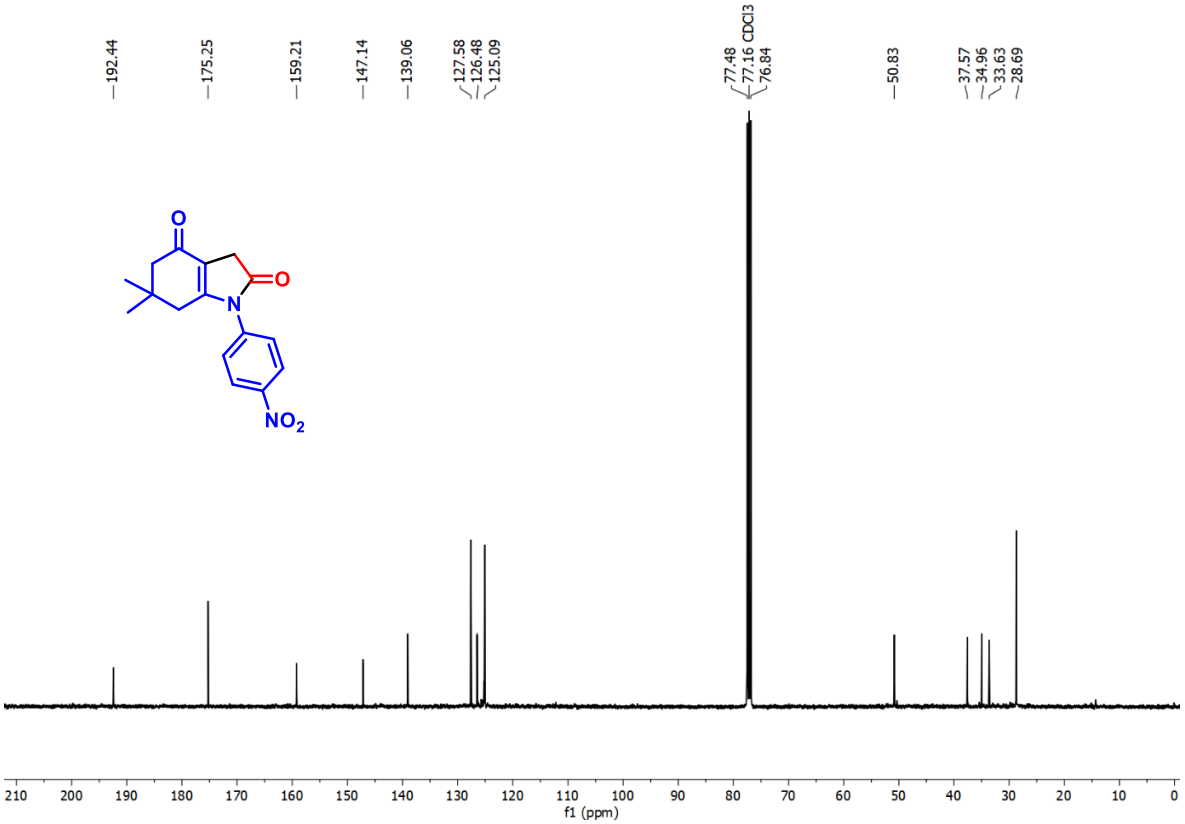
^{1}H NMR Spectrum of Compound **3z** (300 MHz, $CDCl_3$)



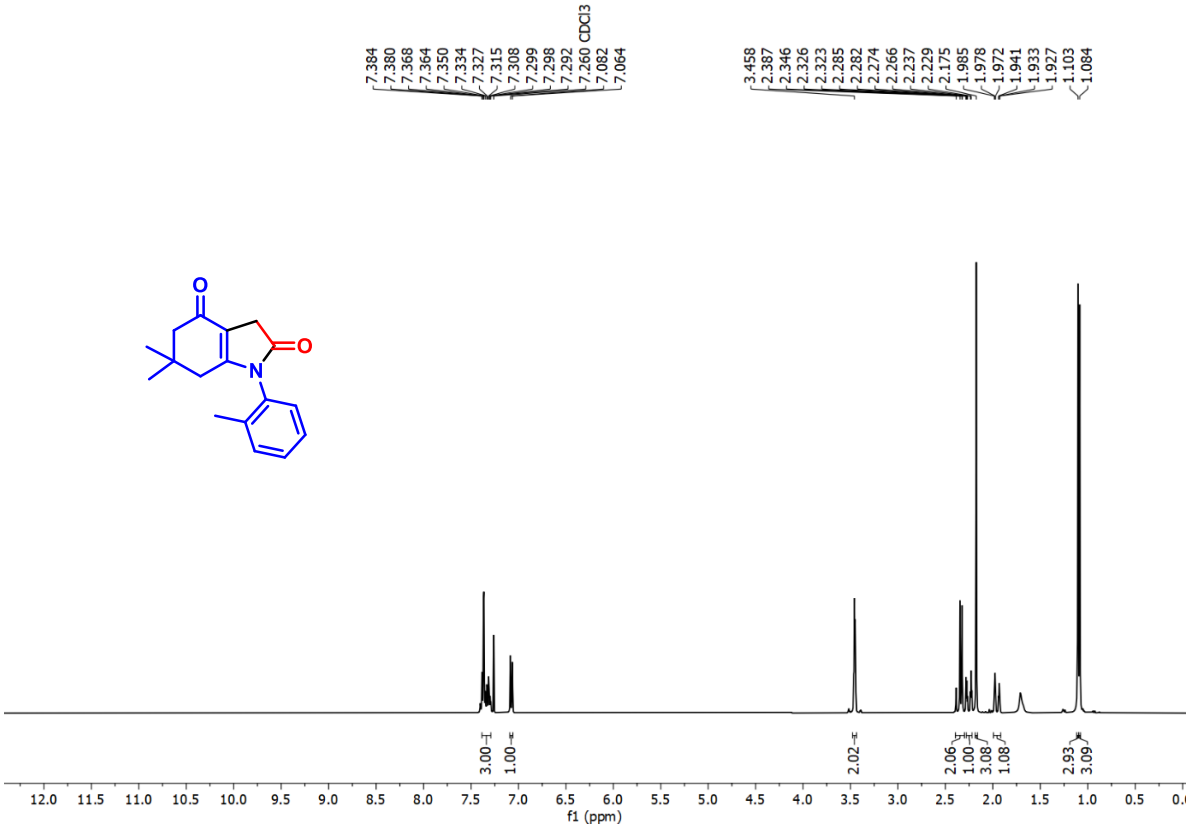
^{13}C {^{1}H} NMR Spectrum of Compound **3z** (75 MHz, $CDCl_3$)



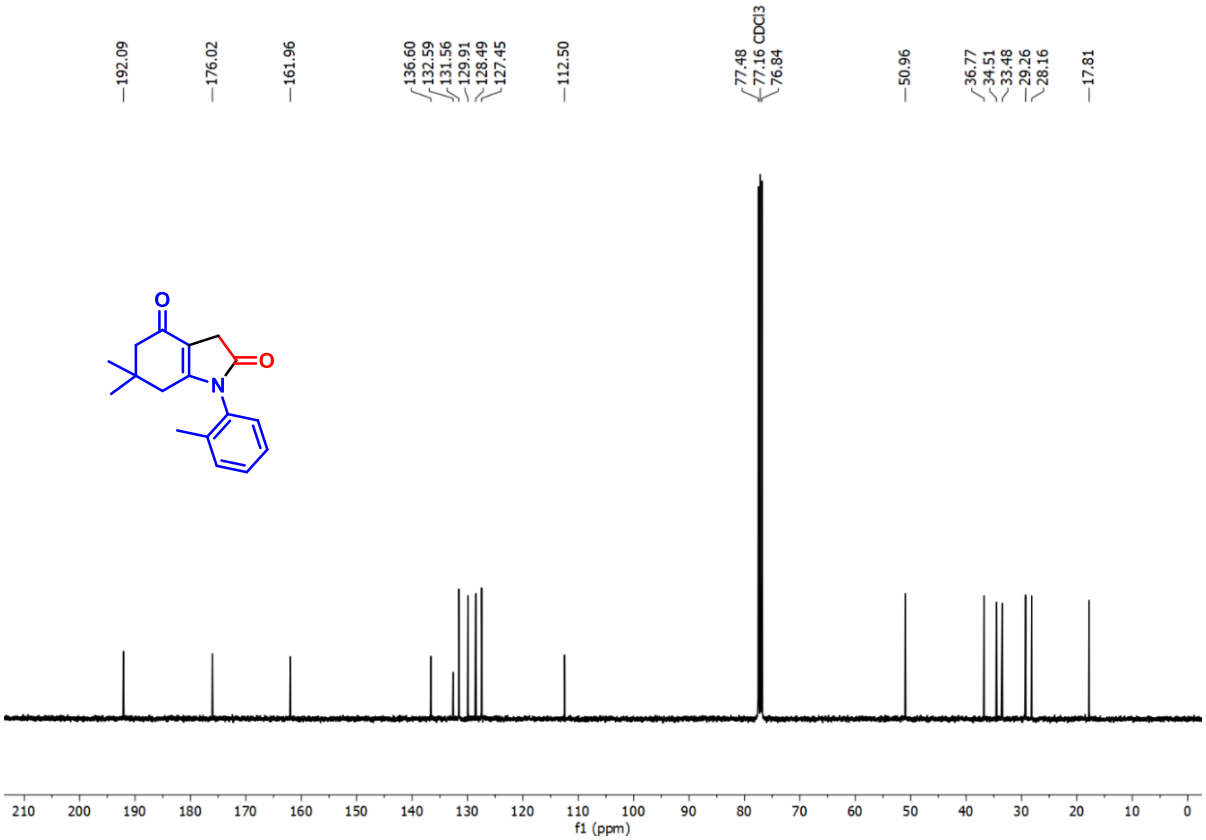
^{1}H NMR Spectrum of Compound **3aa** (400 MHz, $CDCl_3$)



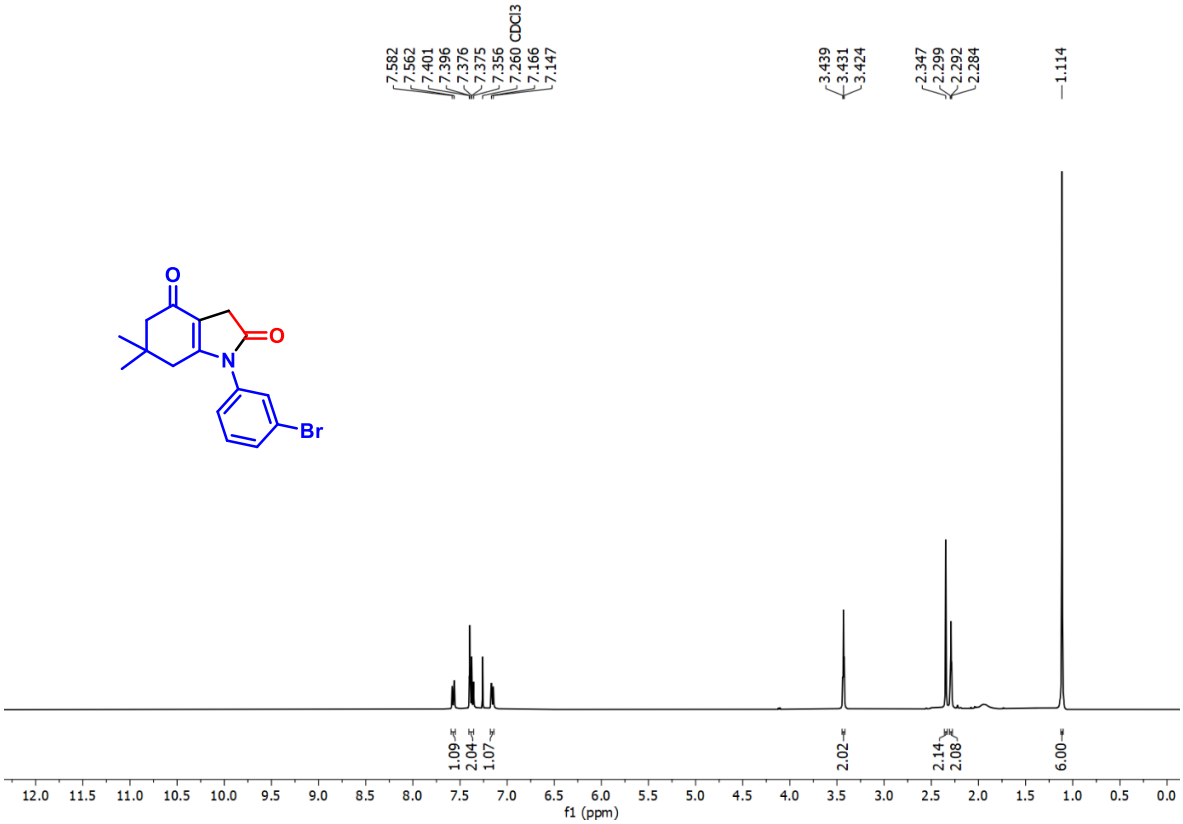
^{13}C {^{1}H} NMR Spectrum of Compound **3aa** (100 MHz, $CDCl_3$)



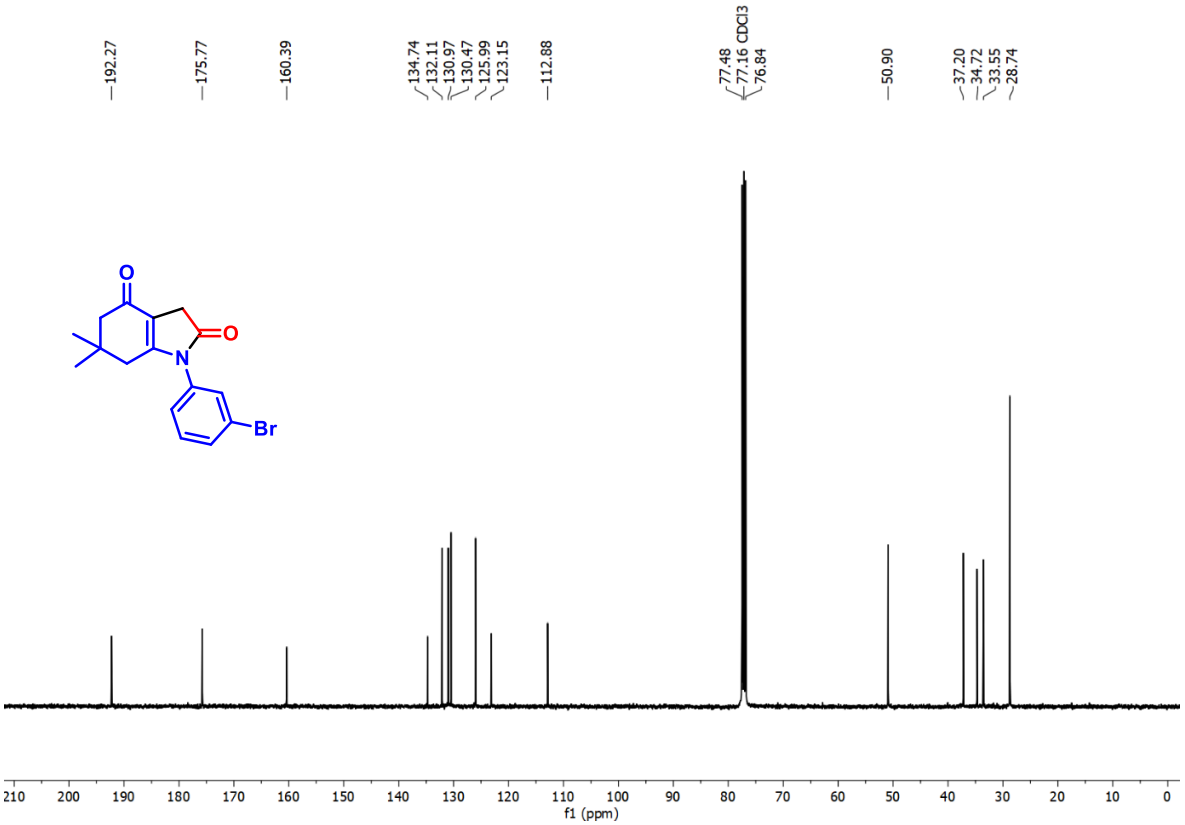
^{1}H NMR Spectrum of Compound **3ab** (400 MHz, $CDCl_3$)



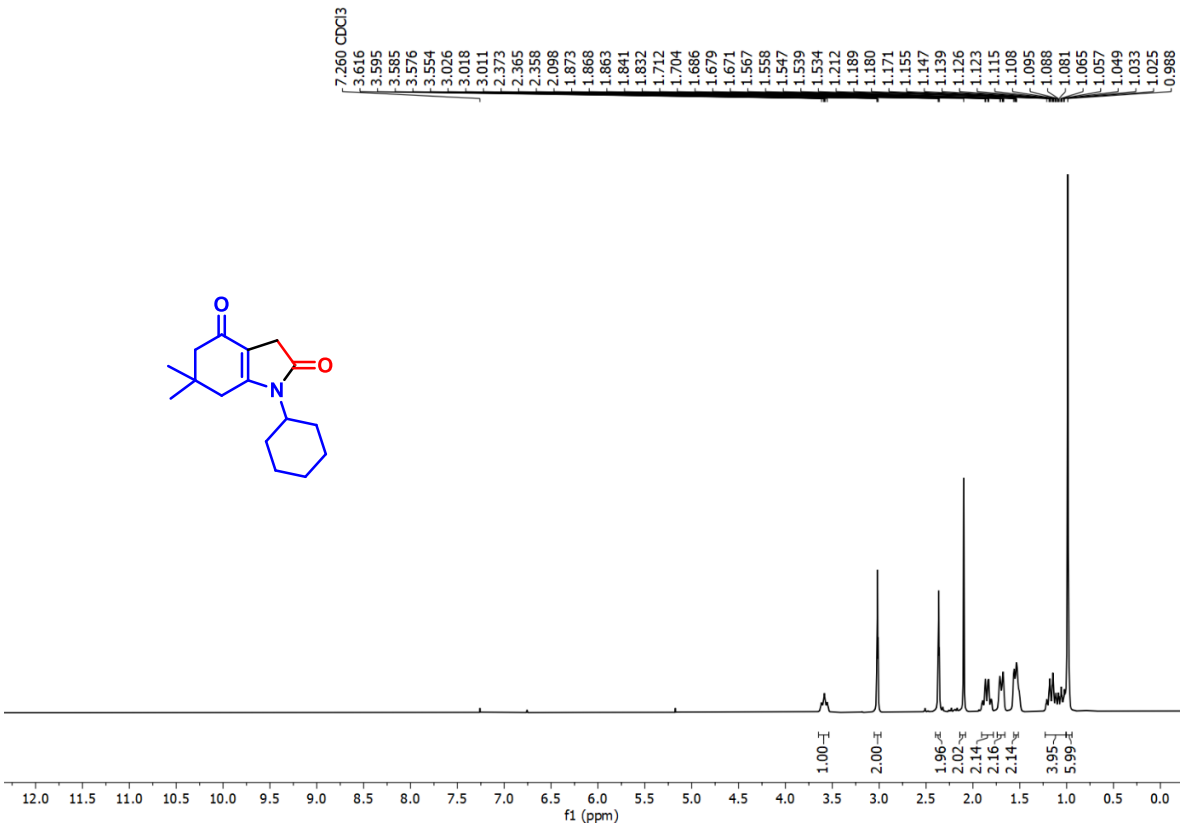
^{13}C {^{1}H} NMR Spectrum of Compound **3ab** (100 MHz, $CDCl_3$)



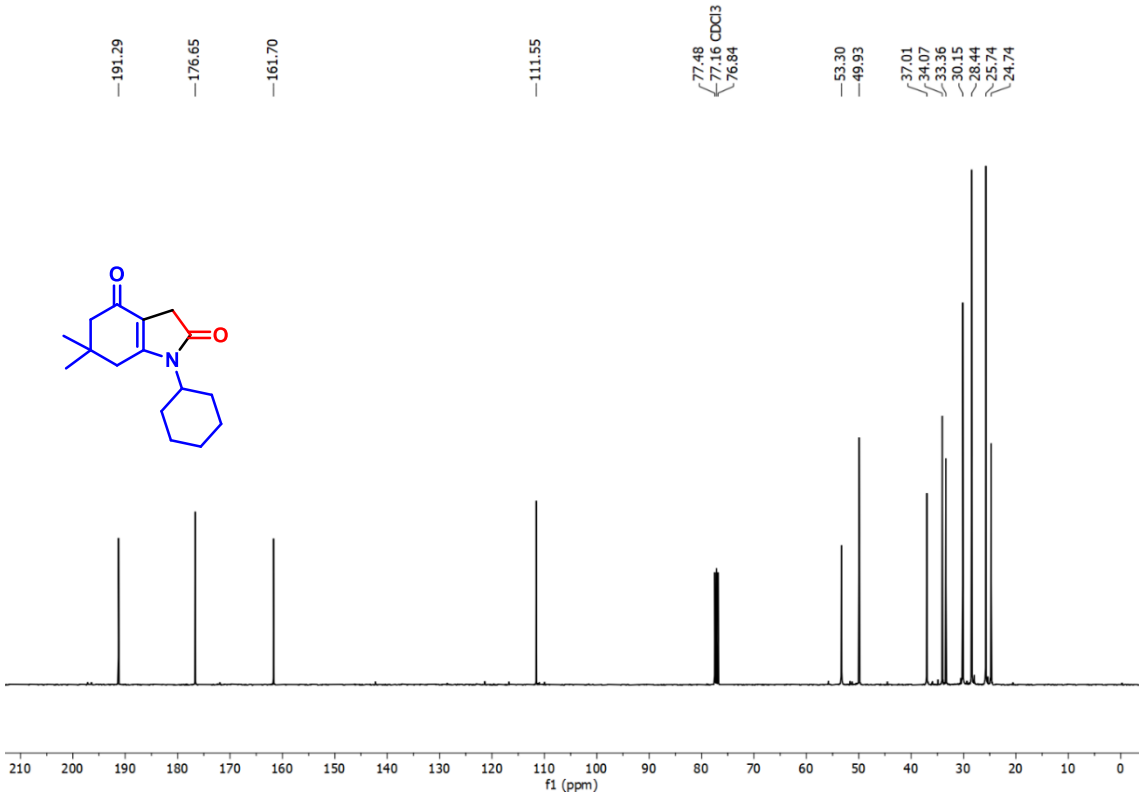
^{1}H NMR Spectrum of Compound **3ac** (400 MHz, $CDCl_3$)



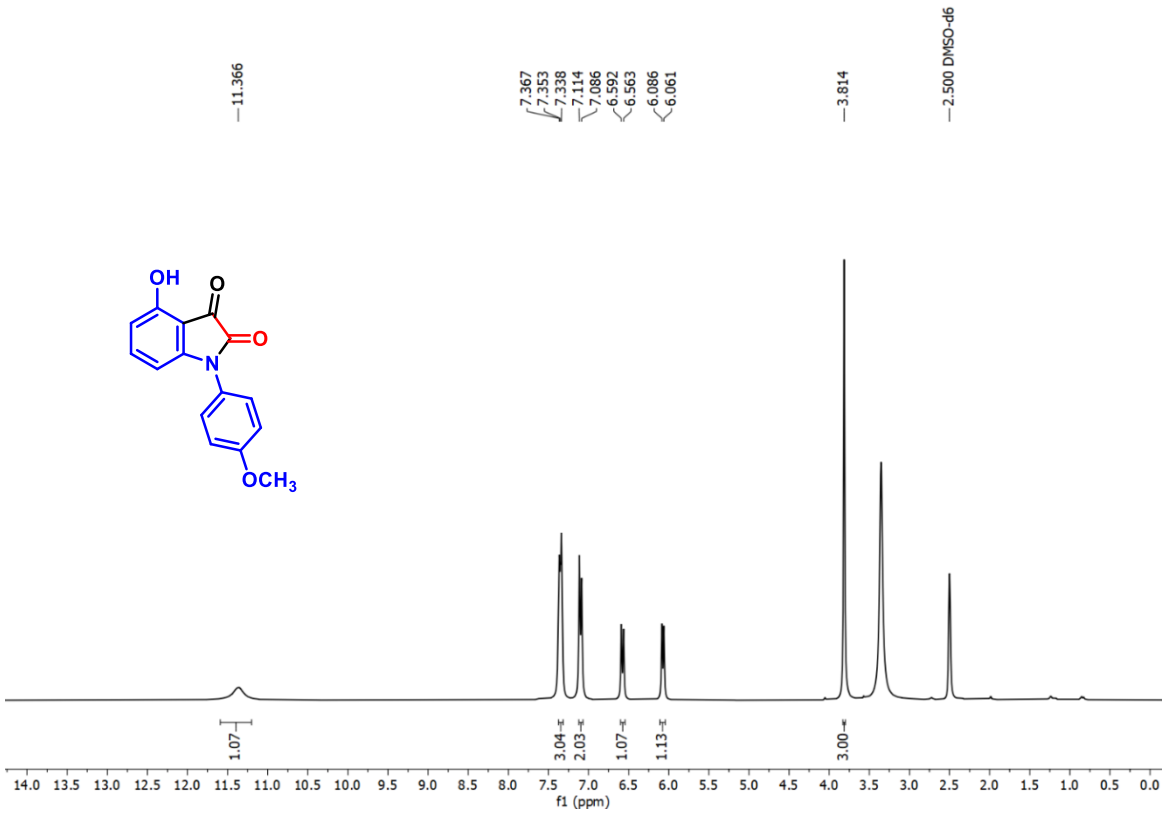
^{13}C {^{1}H} NMR Spectrum of Compound **3ac** (100 MHz, $CDCl_3$)



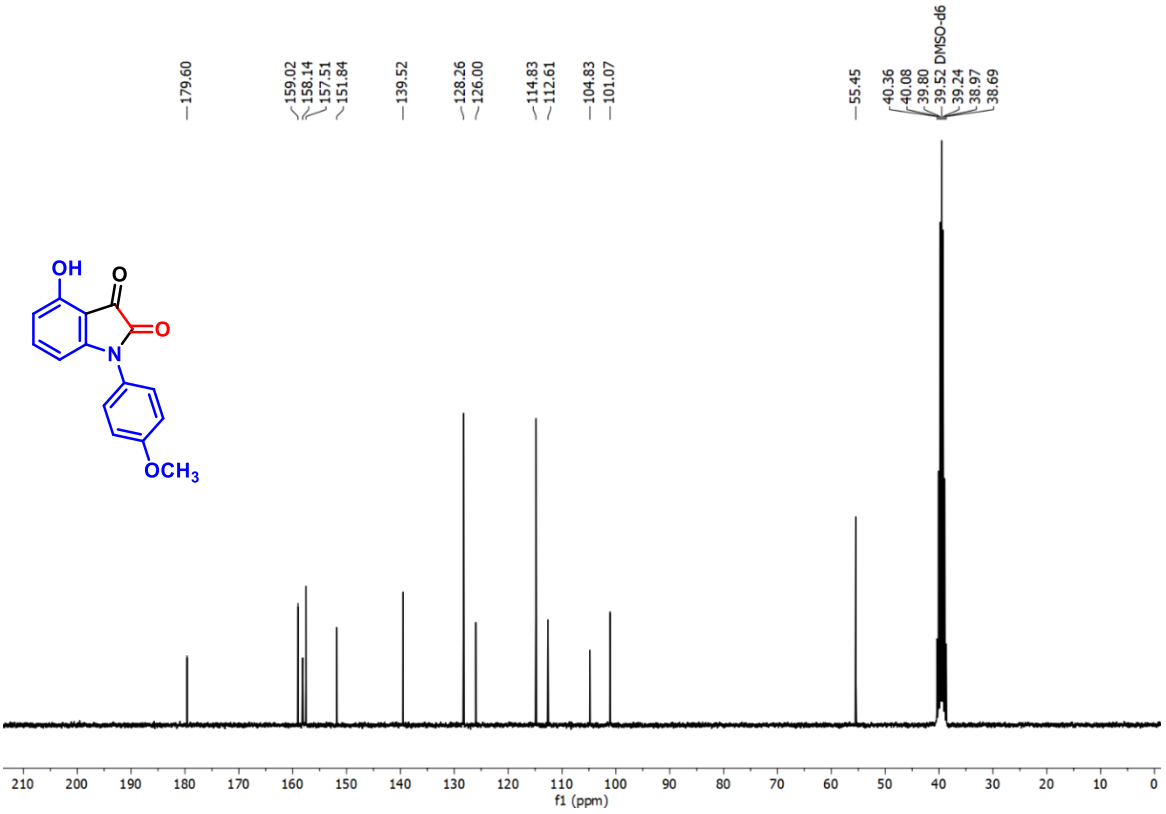
^{1}H NMR Spectrum of Compound **3ad** (400 MHz, $CDCl_3$)



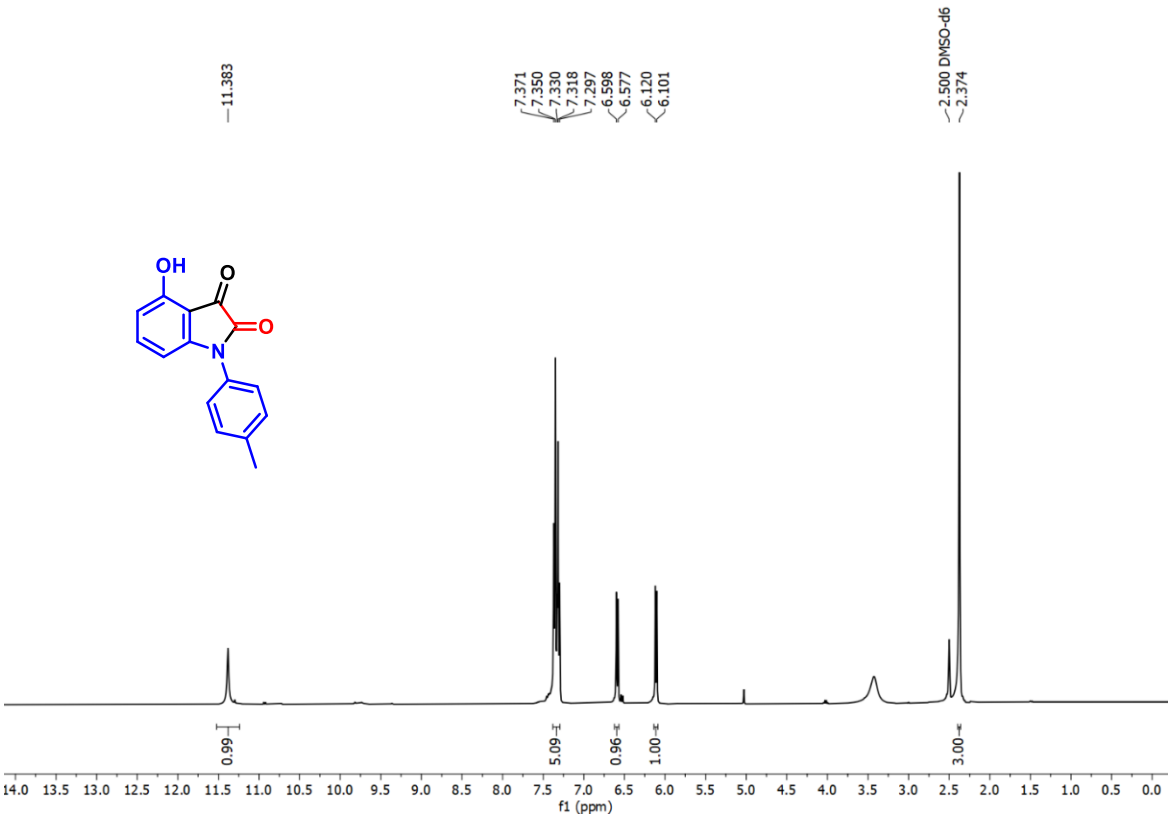
^{13}C {^{1}H}NMR Spectrum of Compound **3ad** (100 MHz, $CDCl_3$)



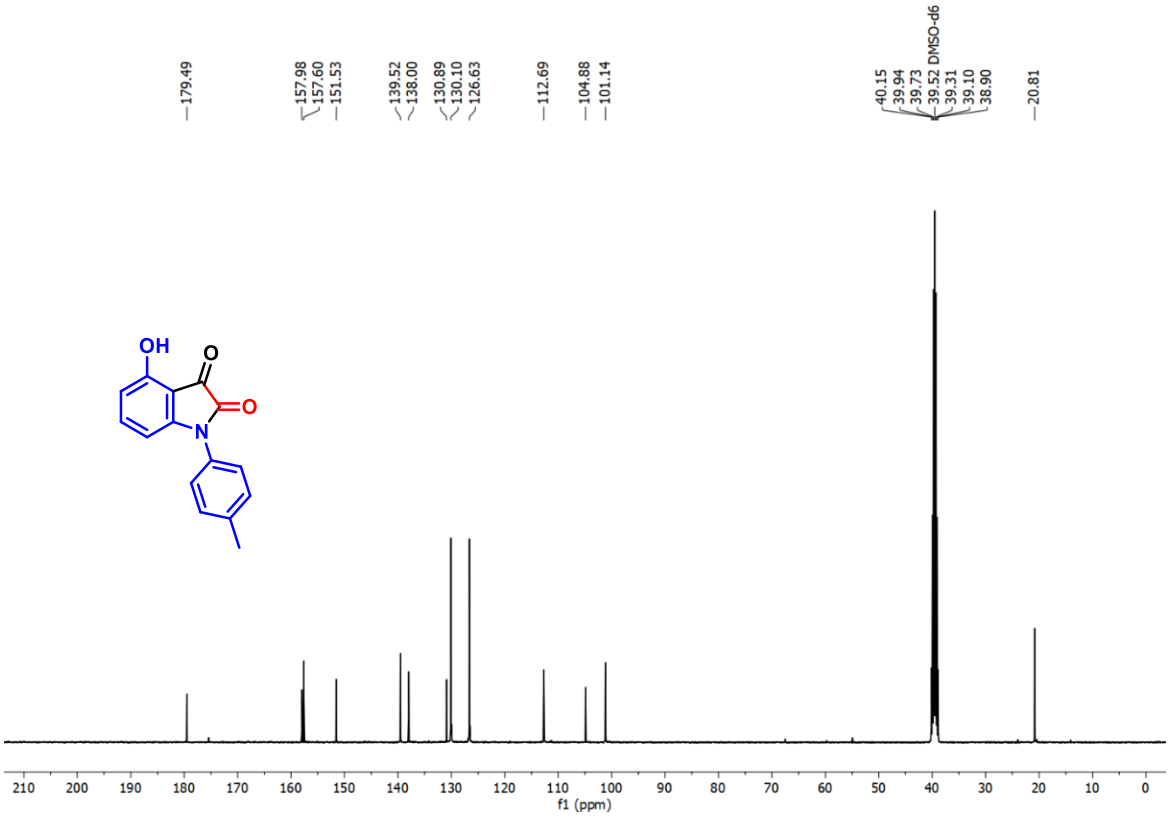
^{1}H NMR Spectrum of Compound **4a** (300 MHz, DMSO-d$_6$)



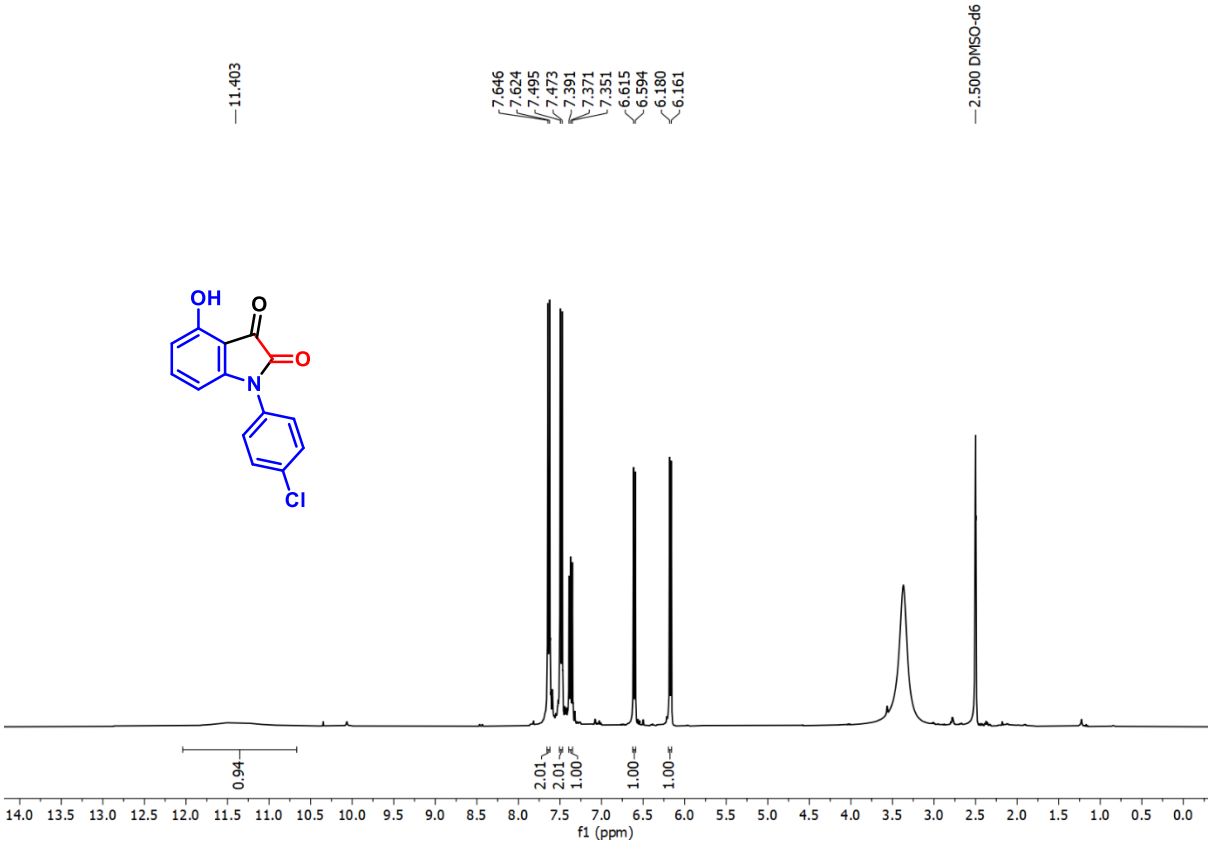
^{13}C {^{1}H} NMR Spectrum of Compound **4a** (75 MHz, DMSO-d$_6$)



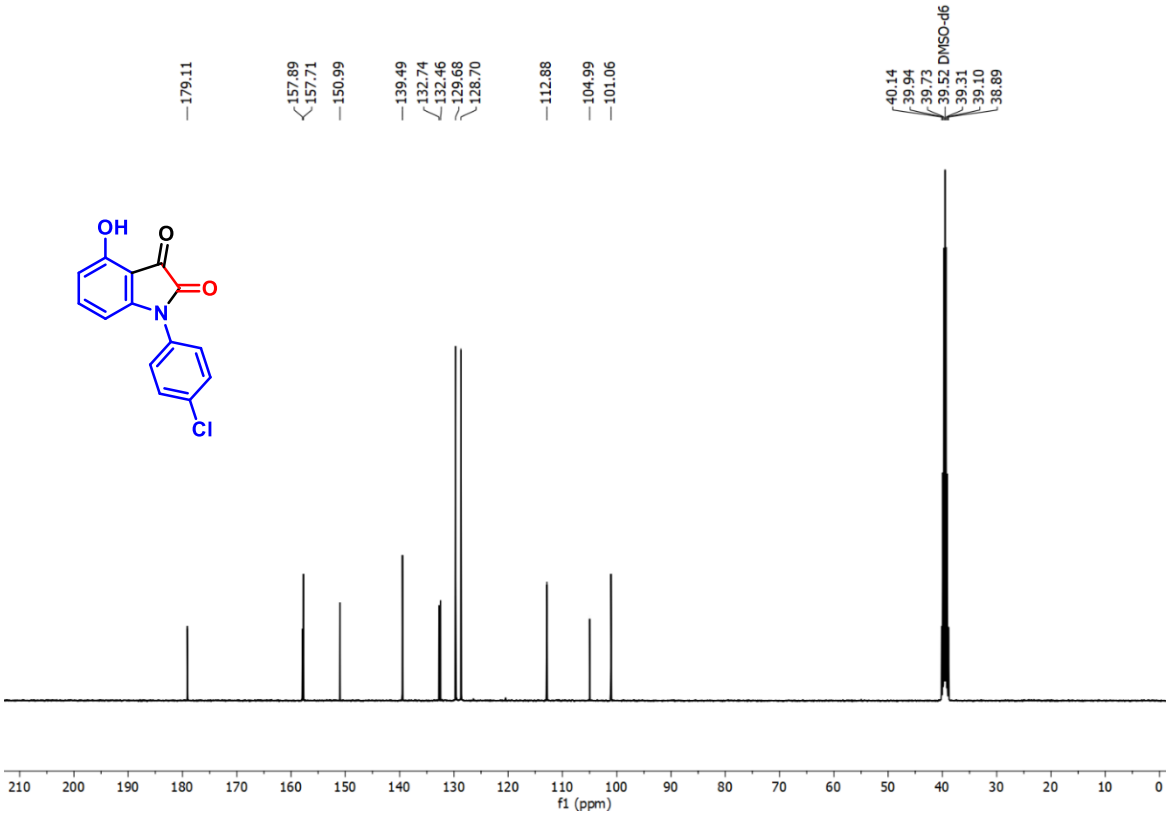
^{1}H NMR Spectrum of Compound **4b** (400 MHz, DMSO-d_6)



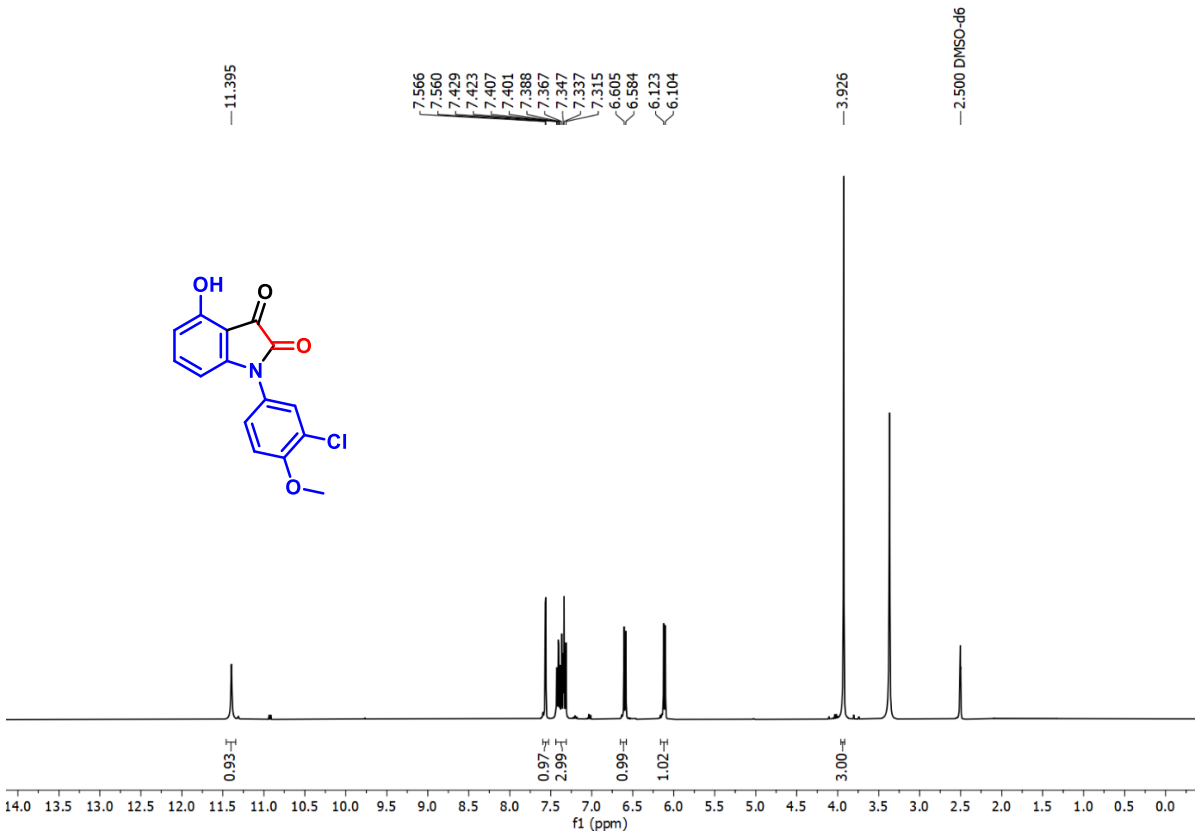
^{13}C {^{1}H}NMR Spectrum of Compound **4b** (100 MHz, DMSO-d_6)



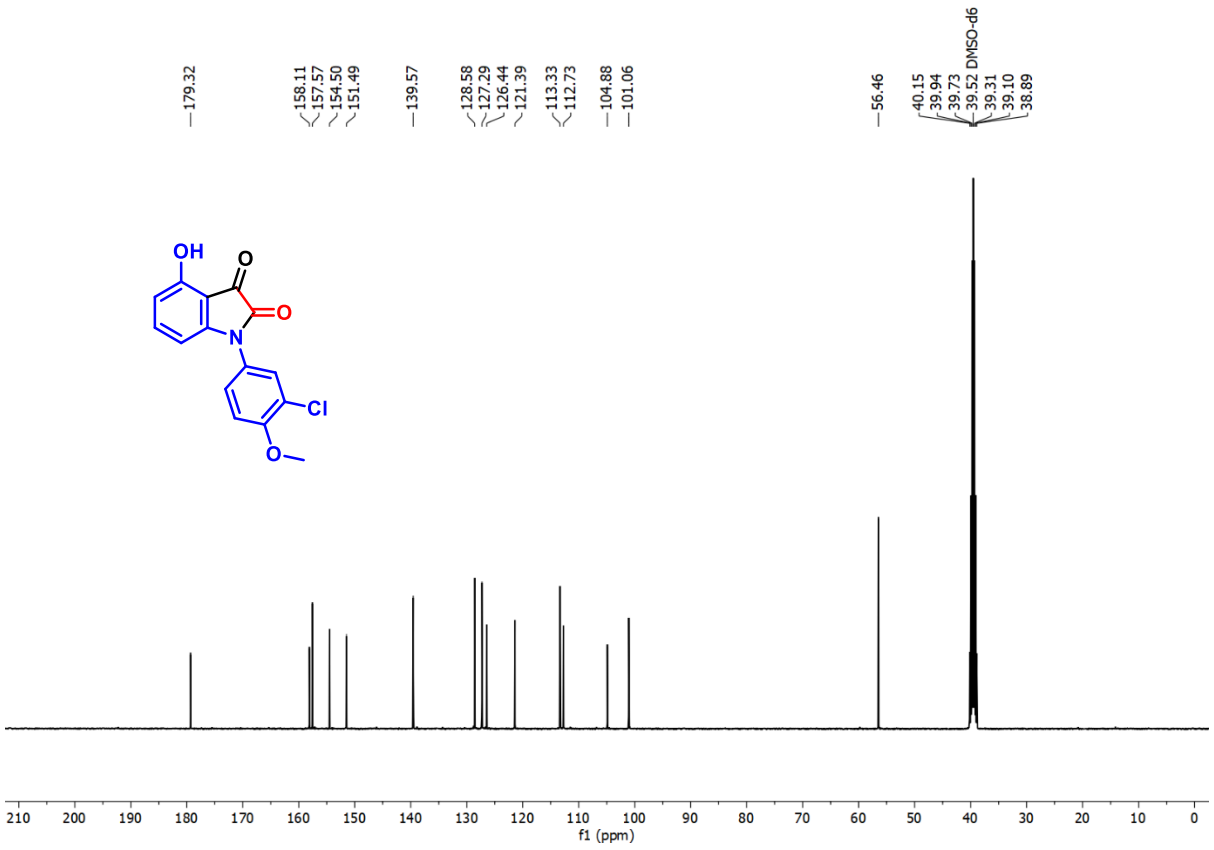
^{1}H NMR Spectrum of Compound **4c** (400 MHz, DMSO-d_6)



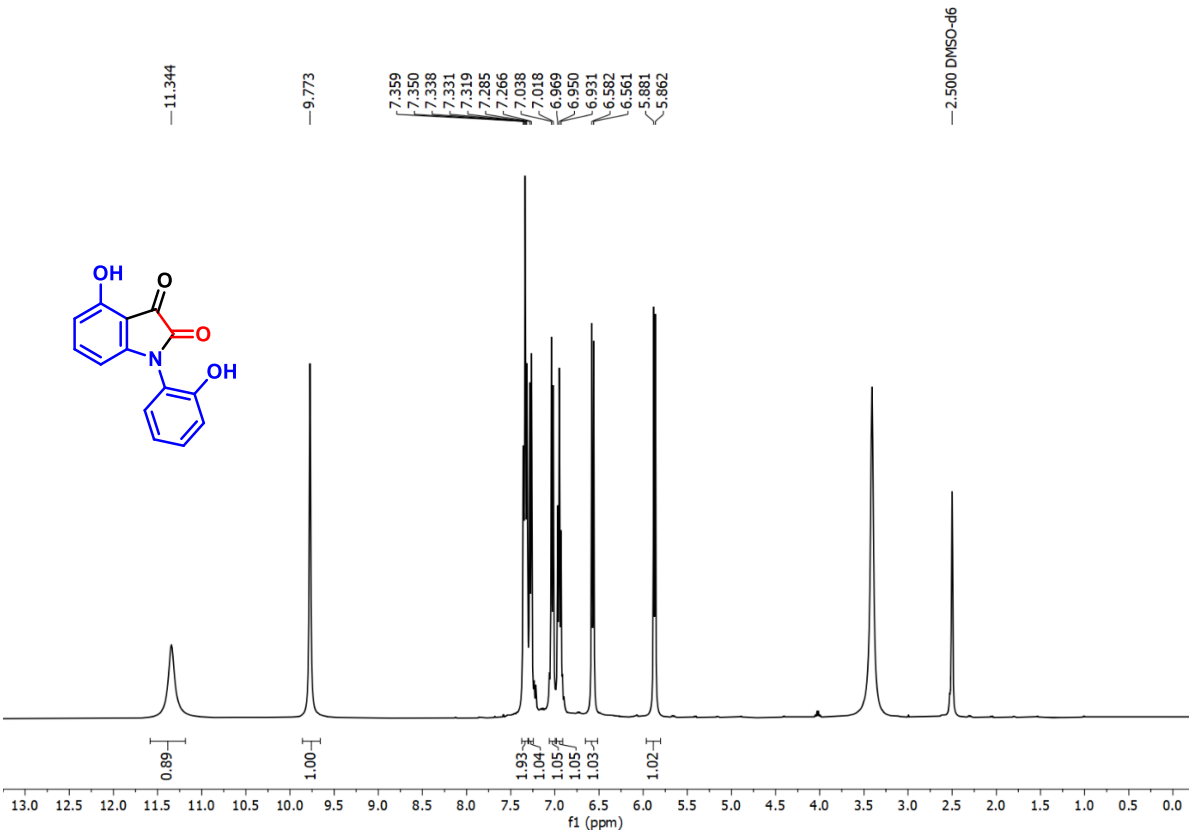
^{13}C {^{1}H} NMR Spectrum of Compound **4c** (100 MHz, DMSO-d_6)



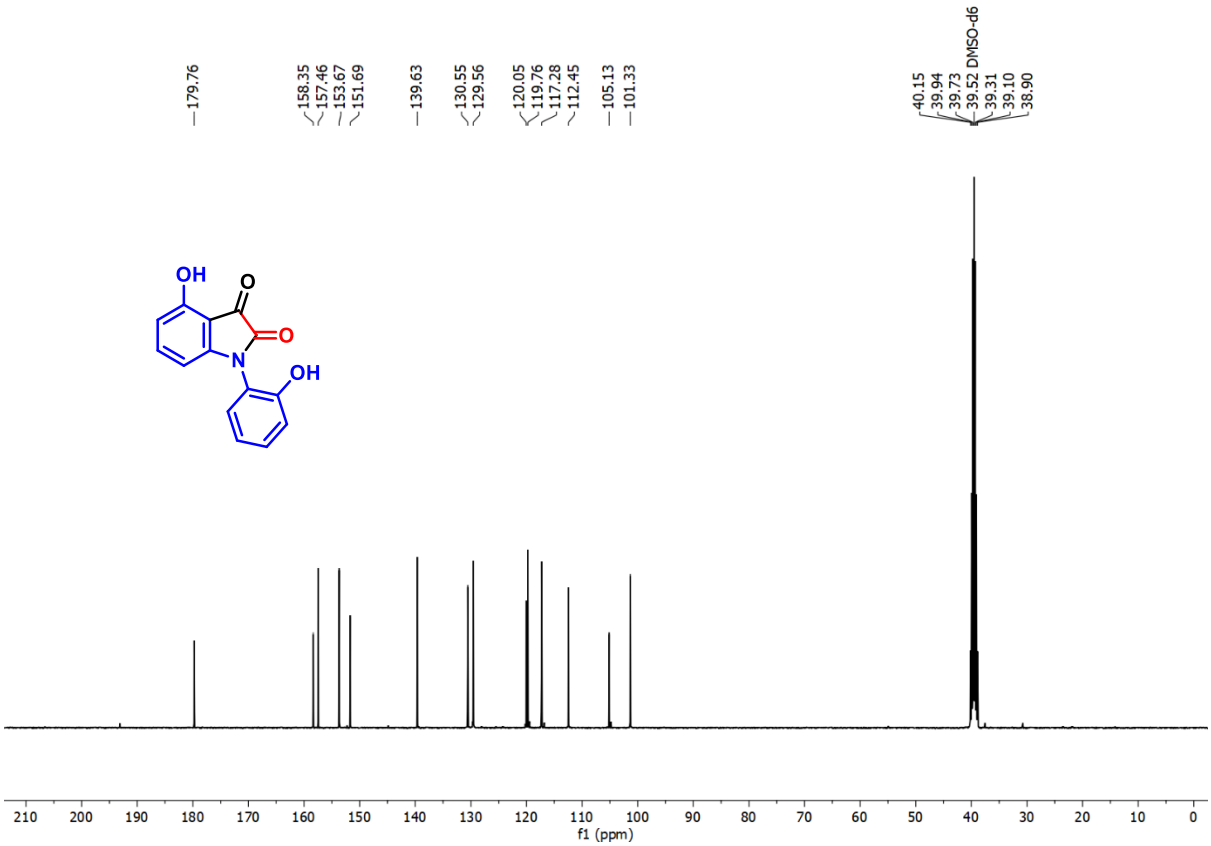
^{1}H NMR Spectrum of Compound **4d** (400 MHz, DMSO-d_6)



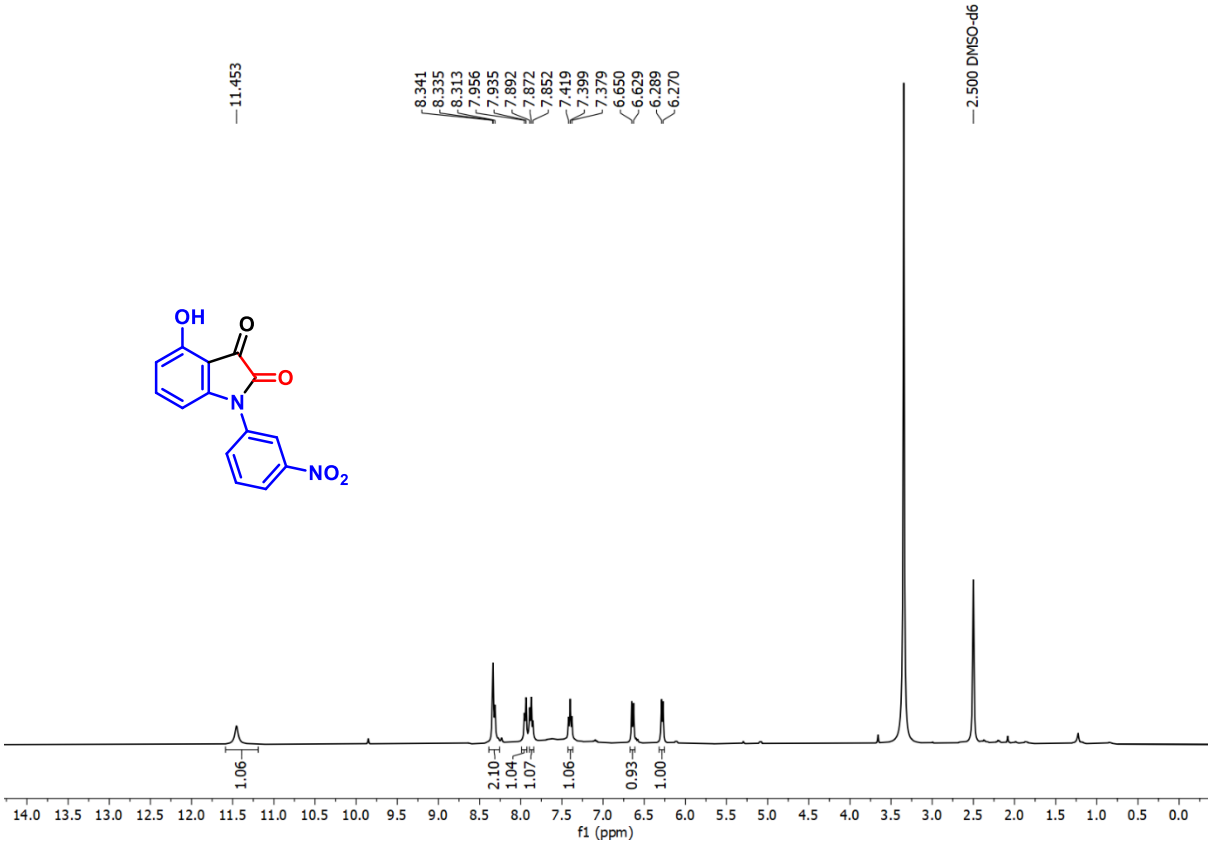
^{13}C {^{1}H} NMR Spectrum of Compound **4d** (100 MHz, DMSO-d_6)



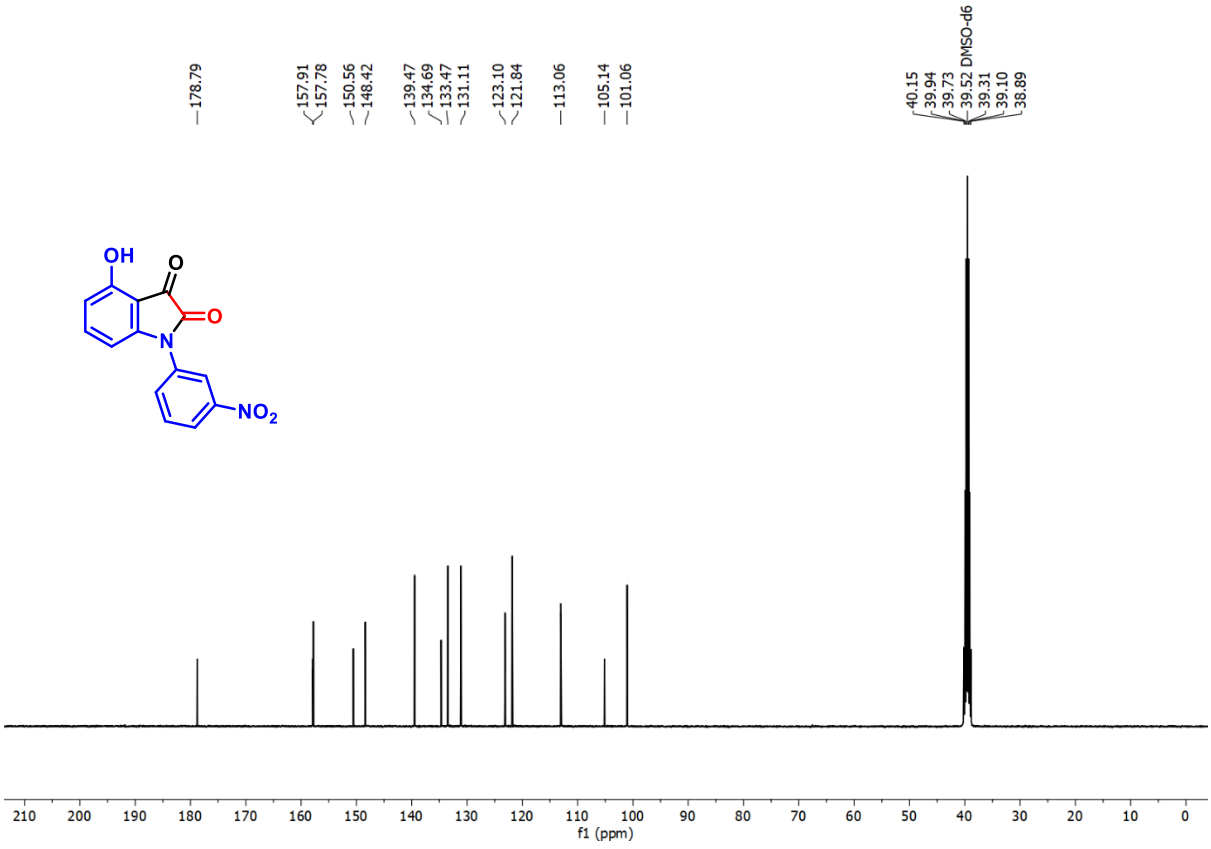
^{1}H NMR Spectrum of Compound **4e** (400 MHz, DMSO-d_6)



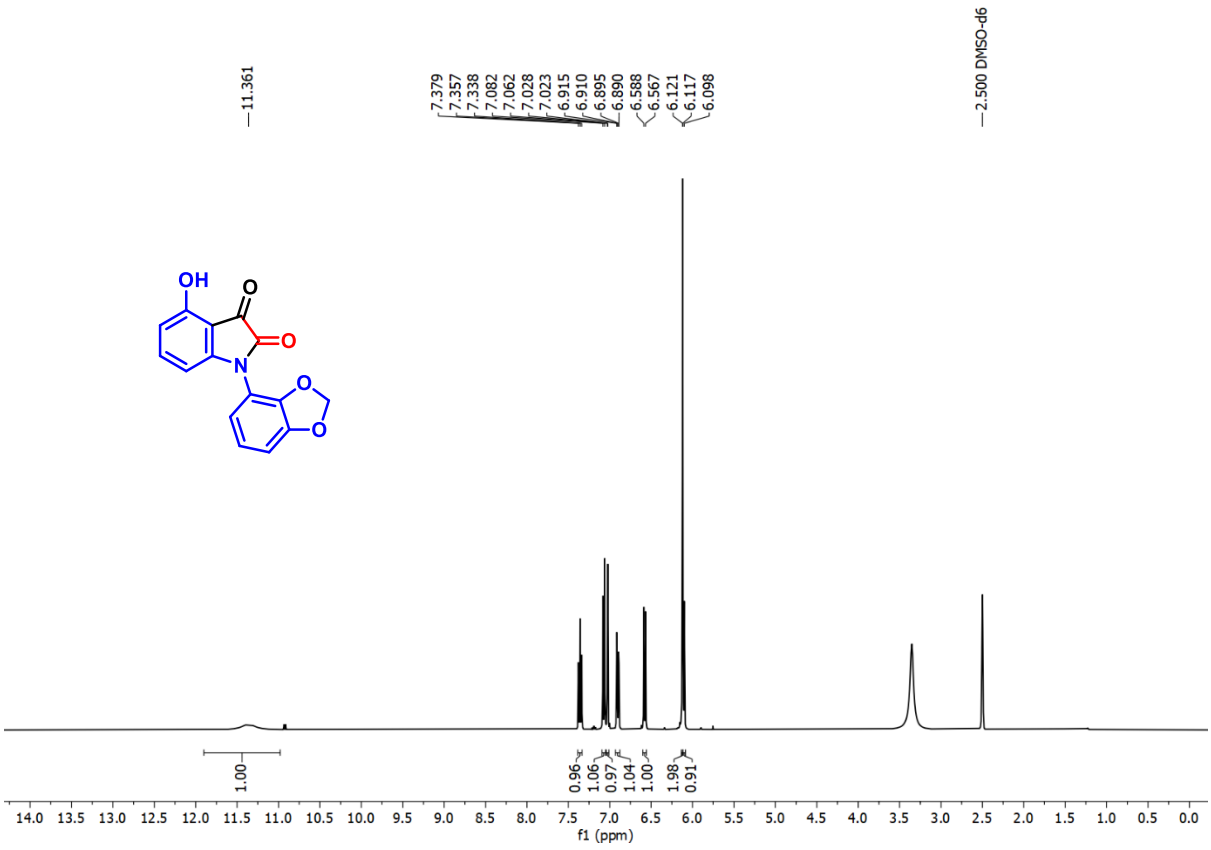
^{13}C {^{1}H} NMR Spectrum of Compound **4e** (100 MHz, DMSO-d_6)



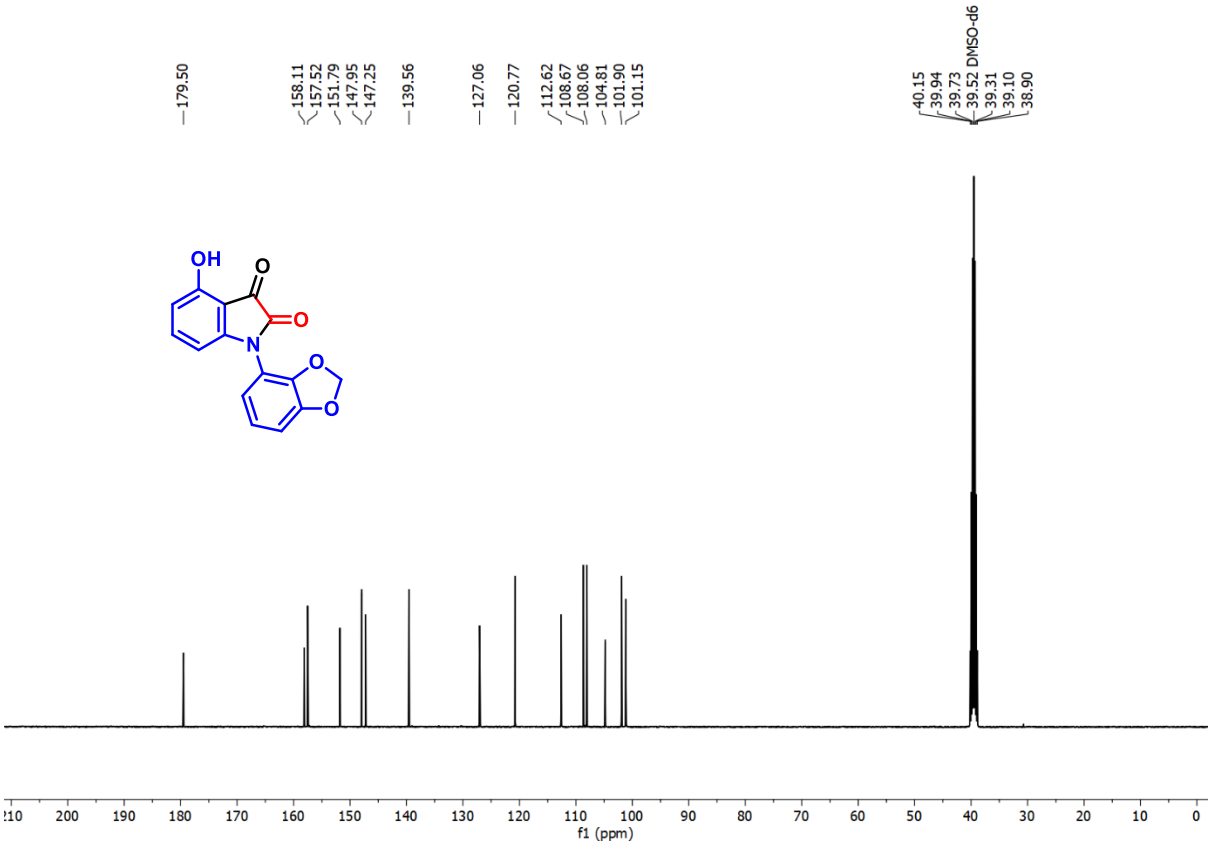
^{1}H NMR Spectrum of Compound **4f** (400 MHz, DMSO-d_6)



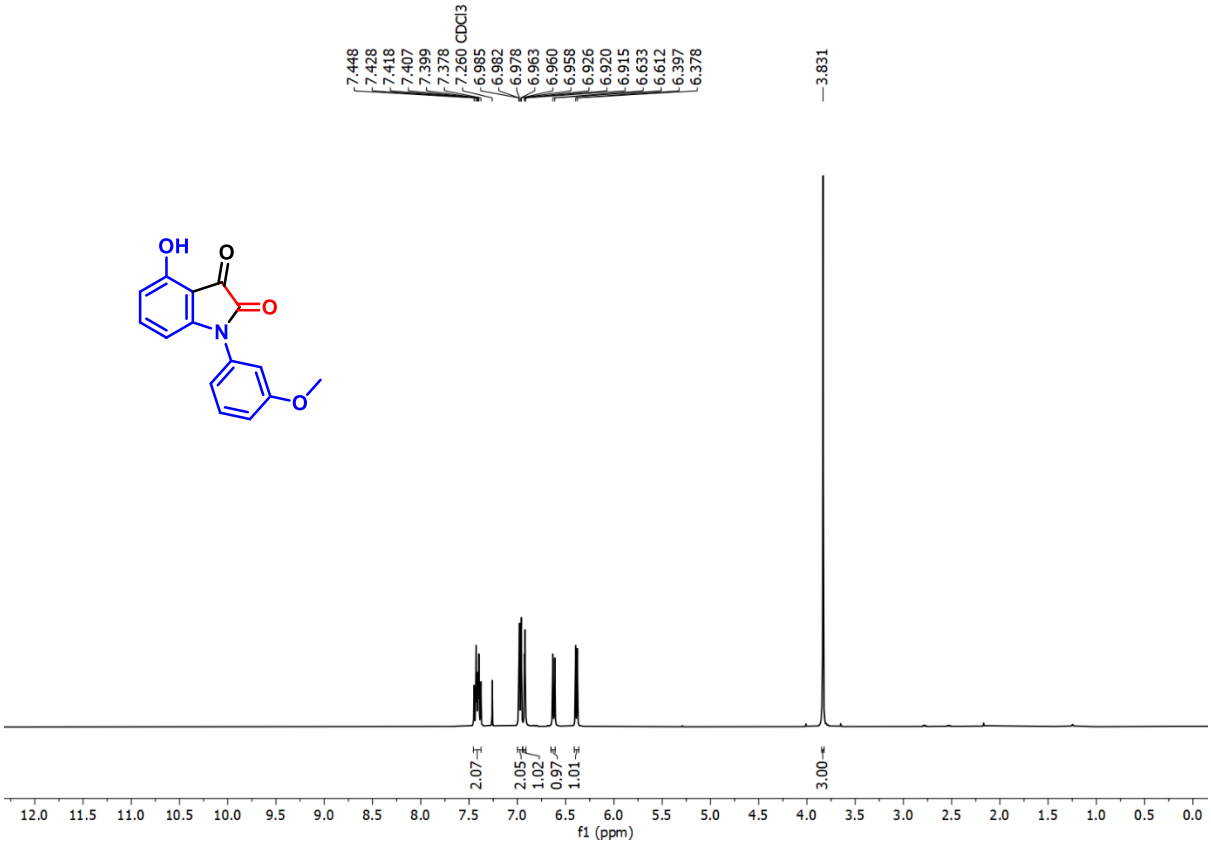
^{13}C {^{1}H} NMR Spectrum of Compound **4f** (100 MHz, DMSO-d_6)



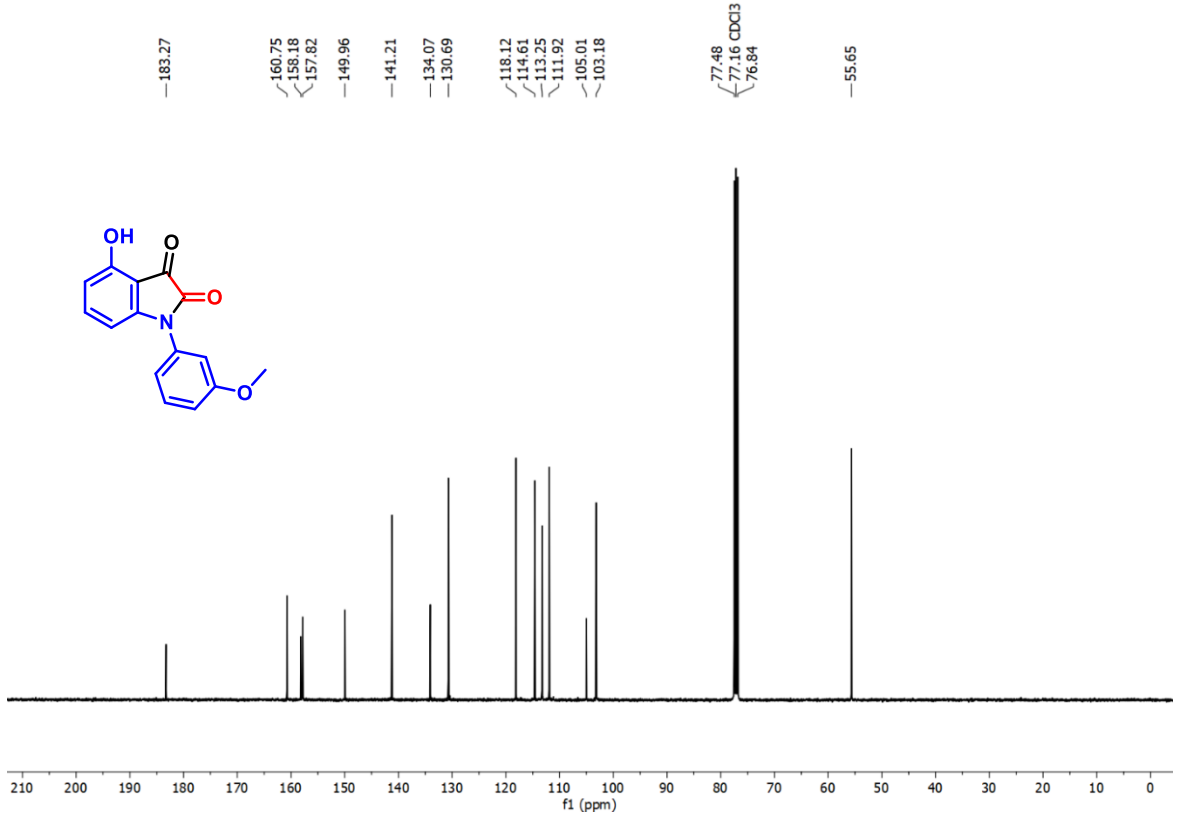
^{1}H NMR Spectrum of Compound **4g** (400 MHz, DMSO-d_6)



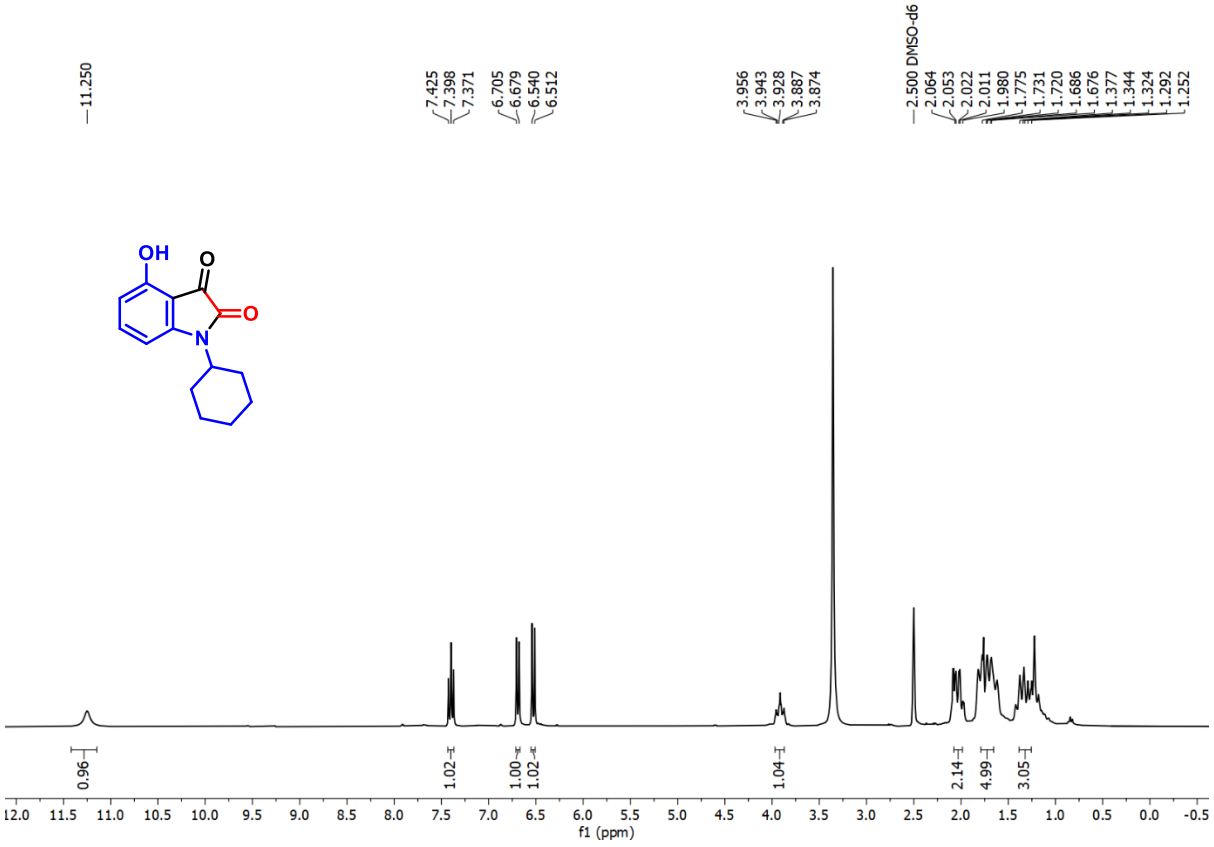
^{13}C NMR Spectrum of Compound **4g** (100 MHz, DMSO-d_6)



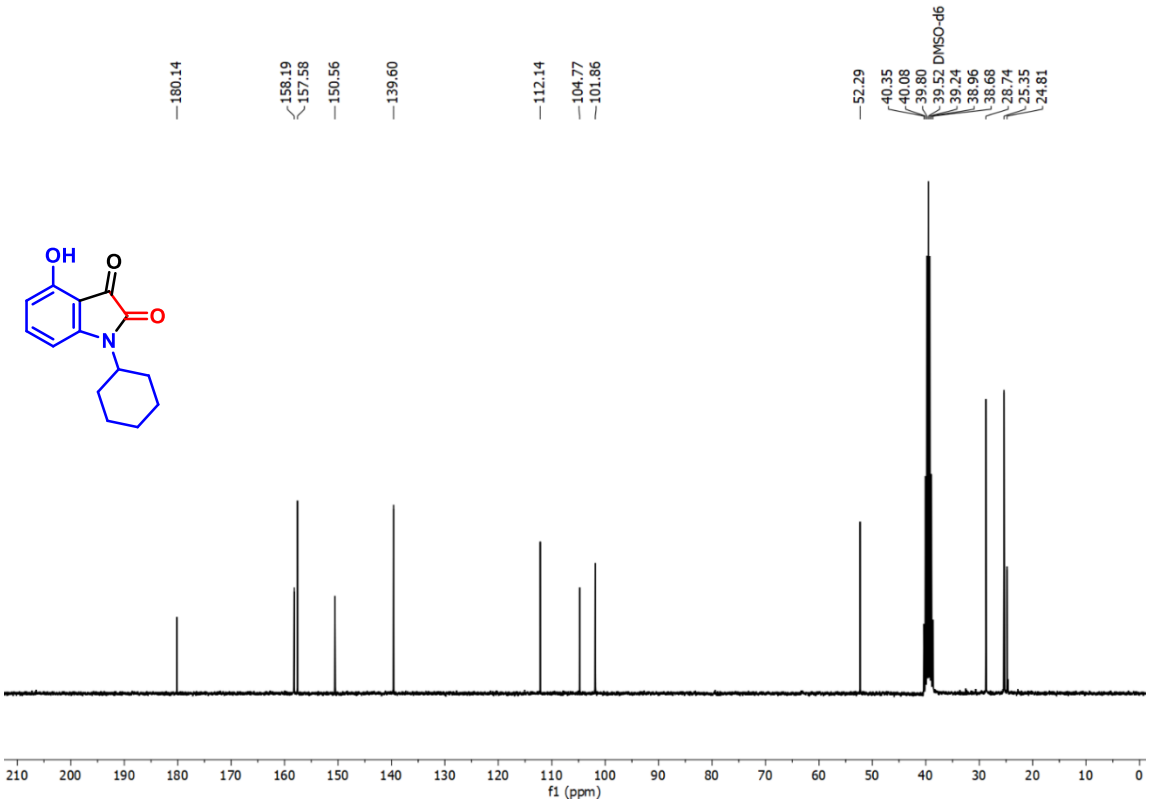
^{1}H NMR Spectrum of Compound **4h** (400 MHz, $CDCl_3$)



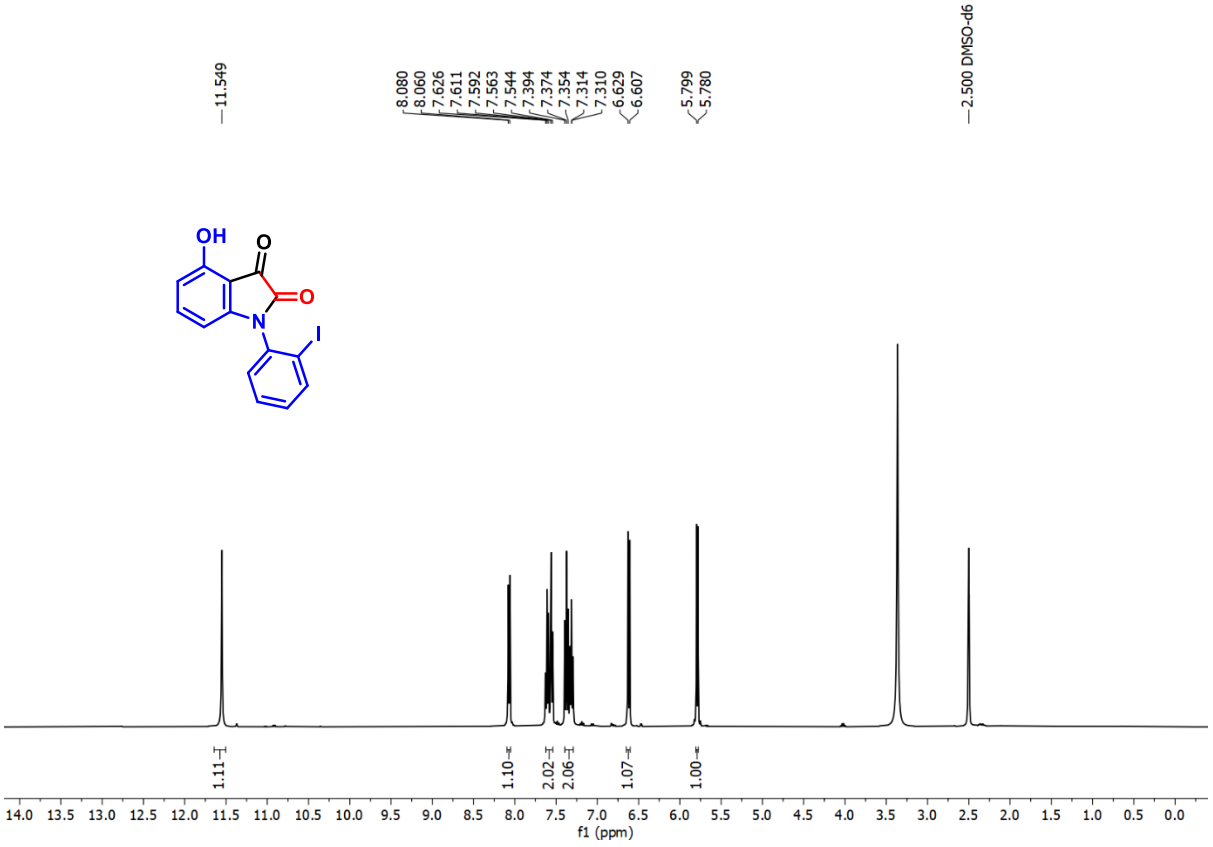
^{13}C {^{1}H} NMR Spectrum of Compound **4h** (100 MHz, $CDCl_3$)



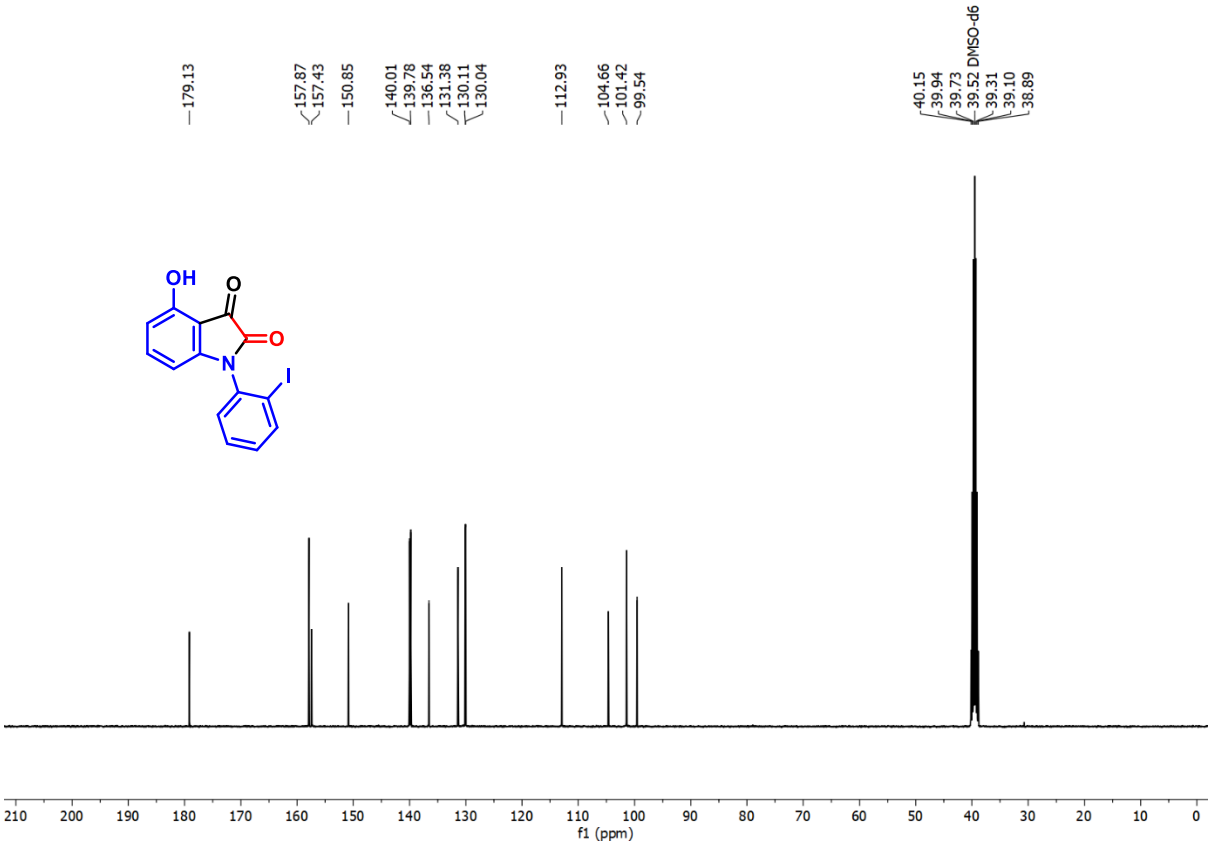
^{1}H NMR Spectrum of Compound **4i** (300 MHz, DMSO-d_6)



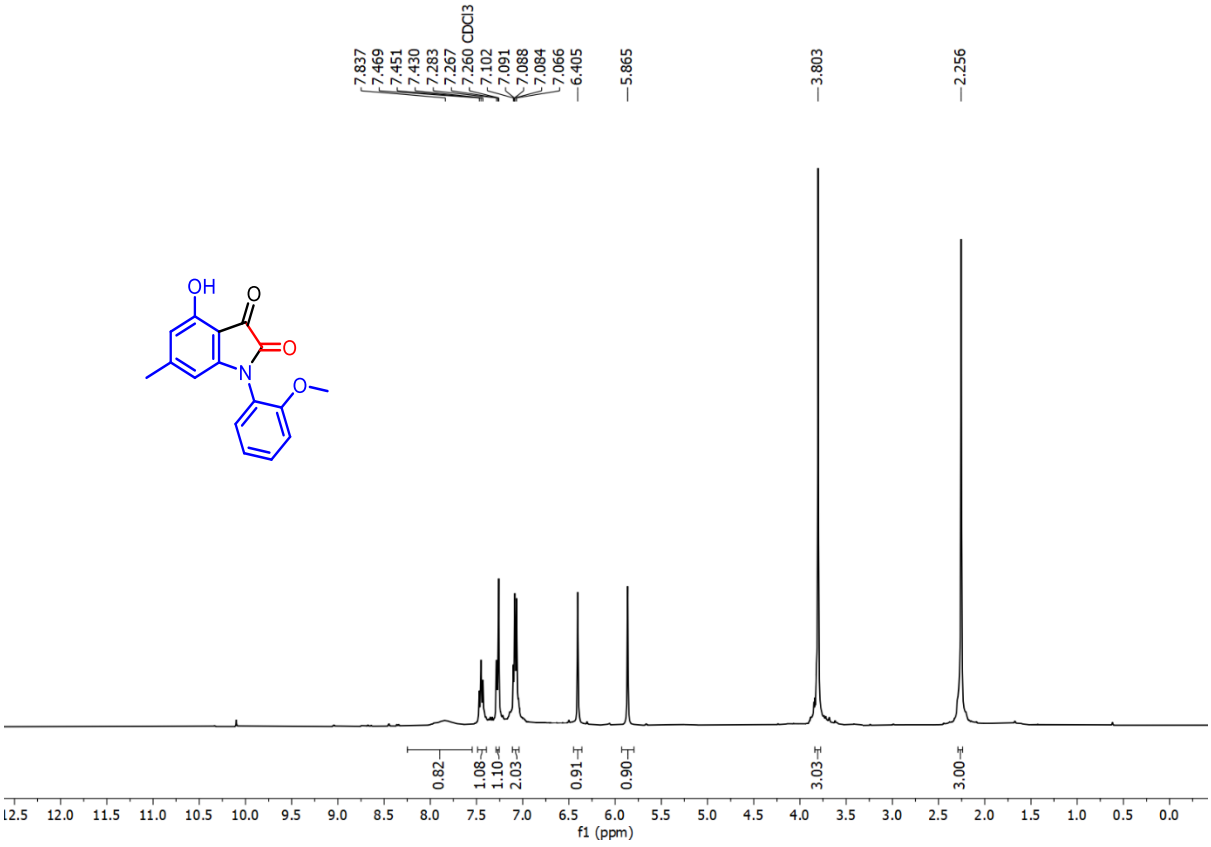
^{13}C {^{1}H} NMR Spectrum of Compound **4i** (75 MHz, DMSO-d_6)



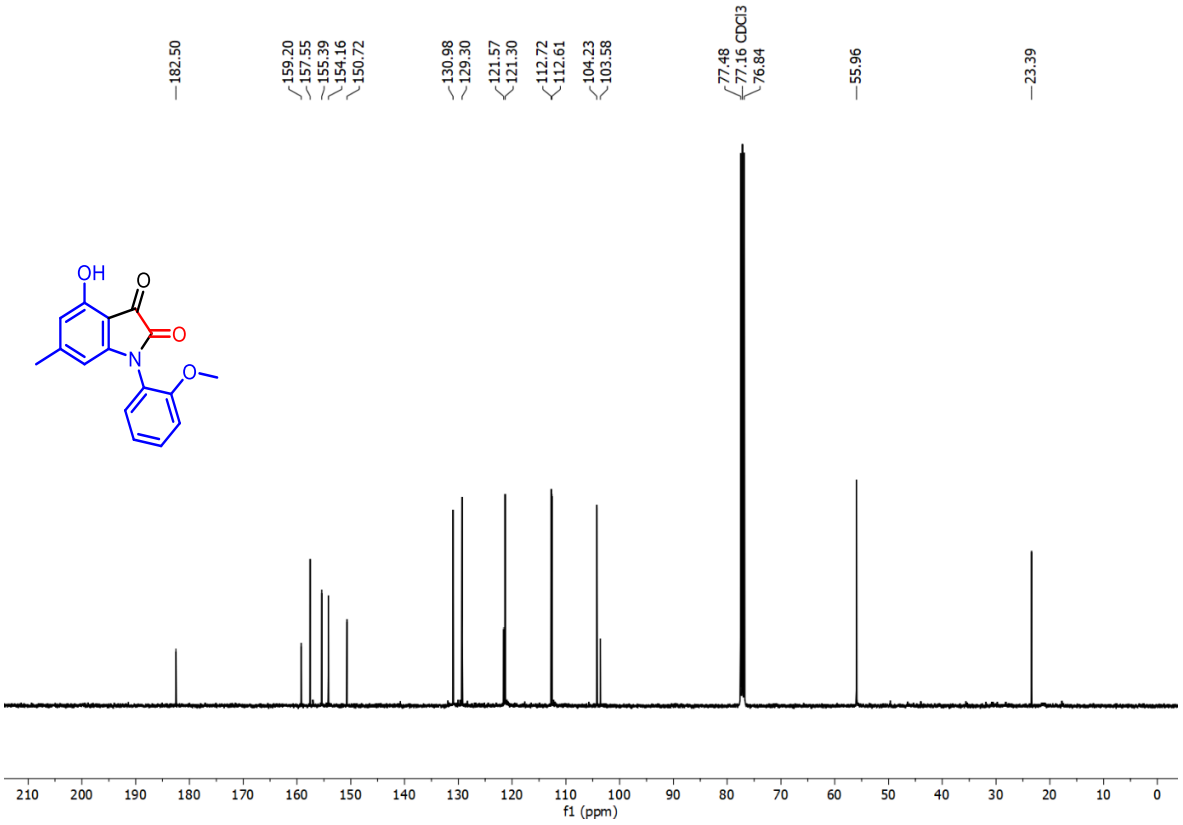
^{1}H NMR Spectrum of Compound **4j** (400 MHz, DMSO-d$_6$)



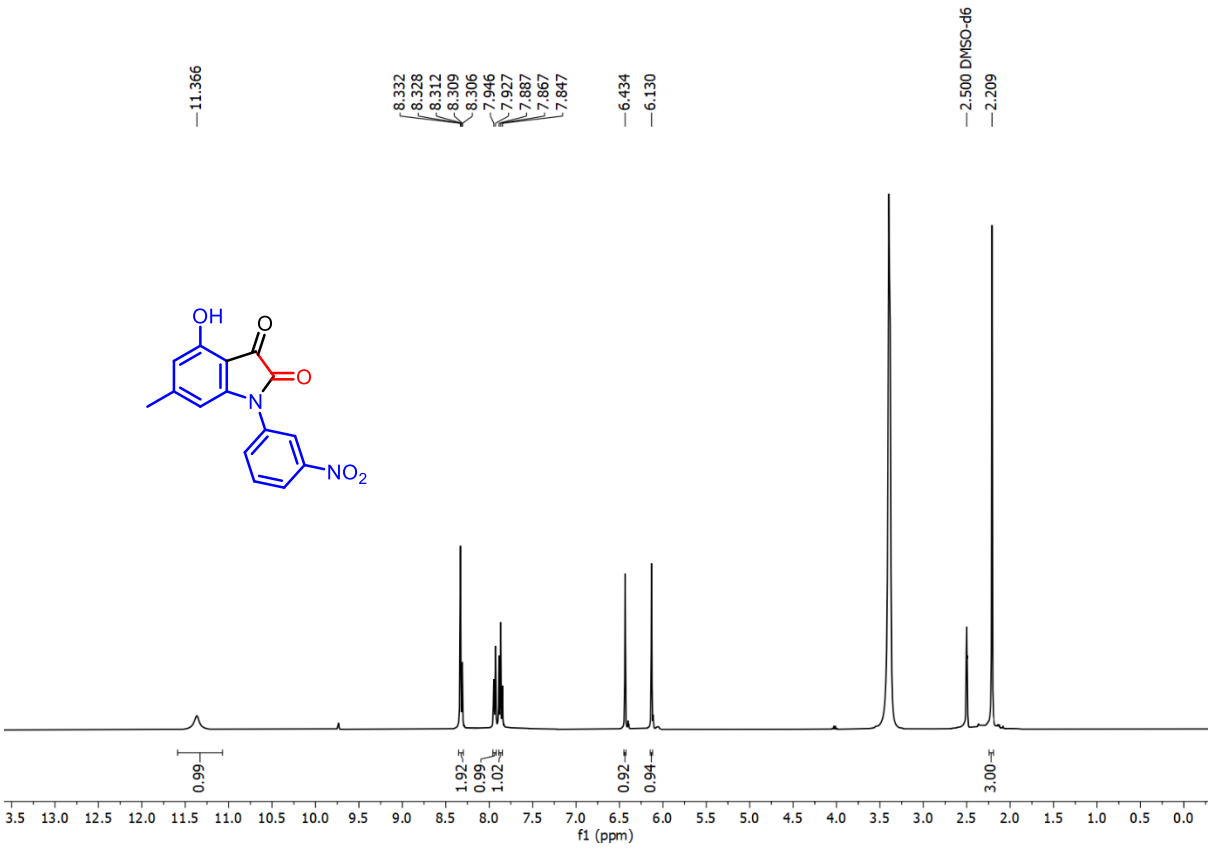
^{13}C {^{1}H} NMR Spectrum of Compound **4j** (100 MHz, DMSO-d$_6$)



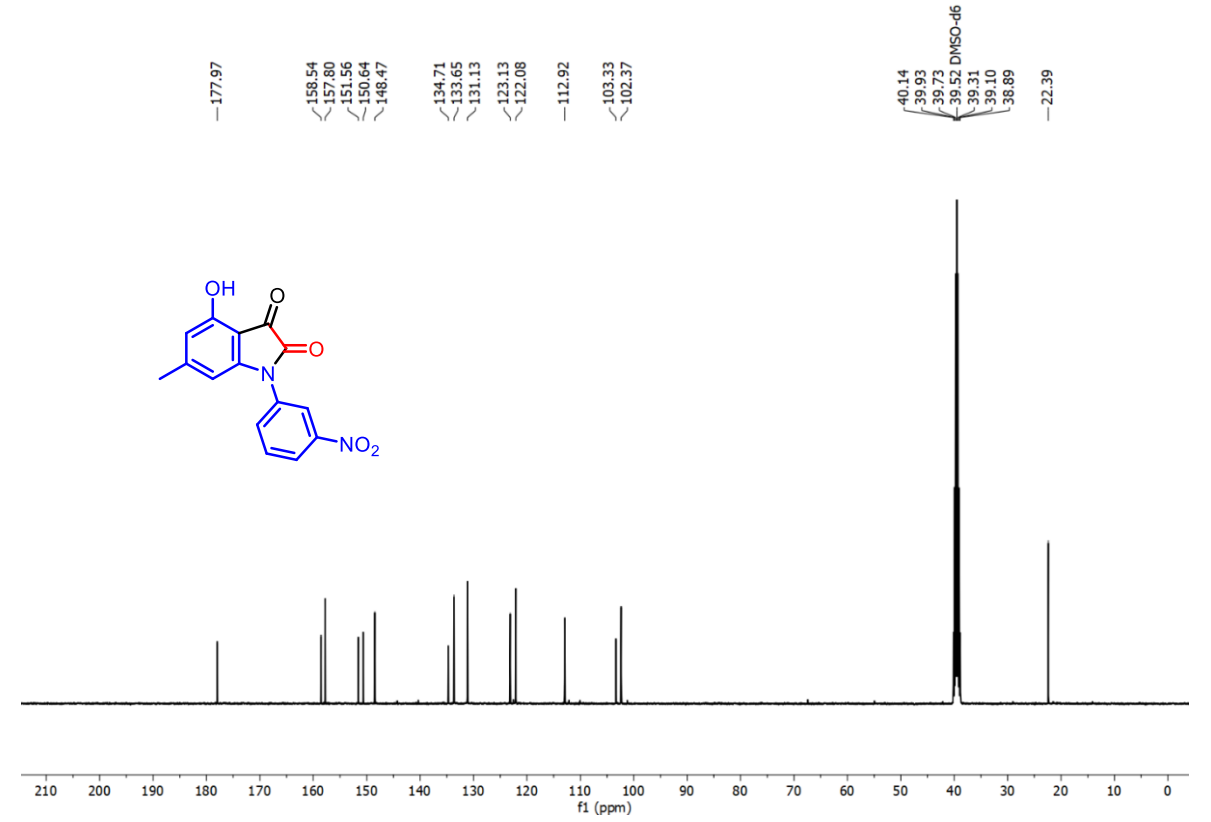
^{1}H NMR Spectrum of Compound **4k** (400 MHz, $CDCl_3$)



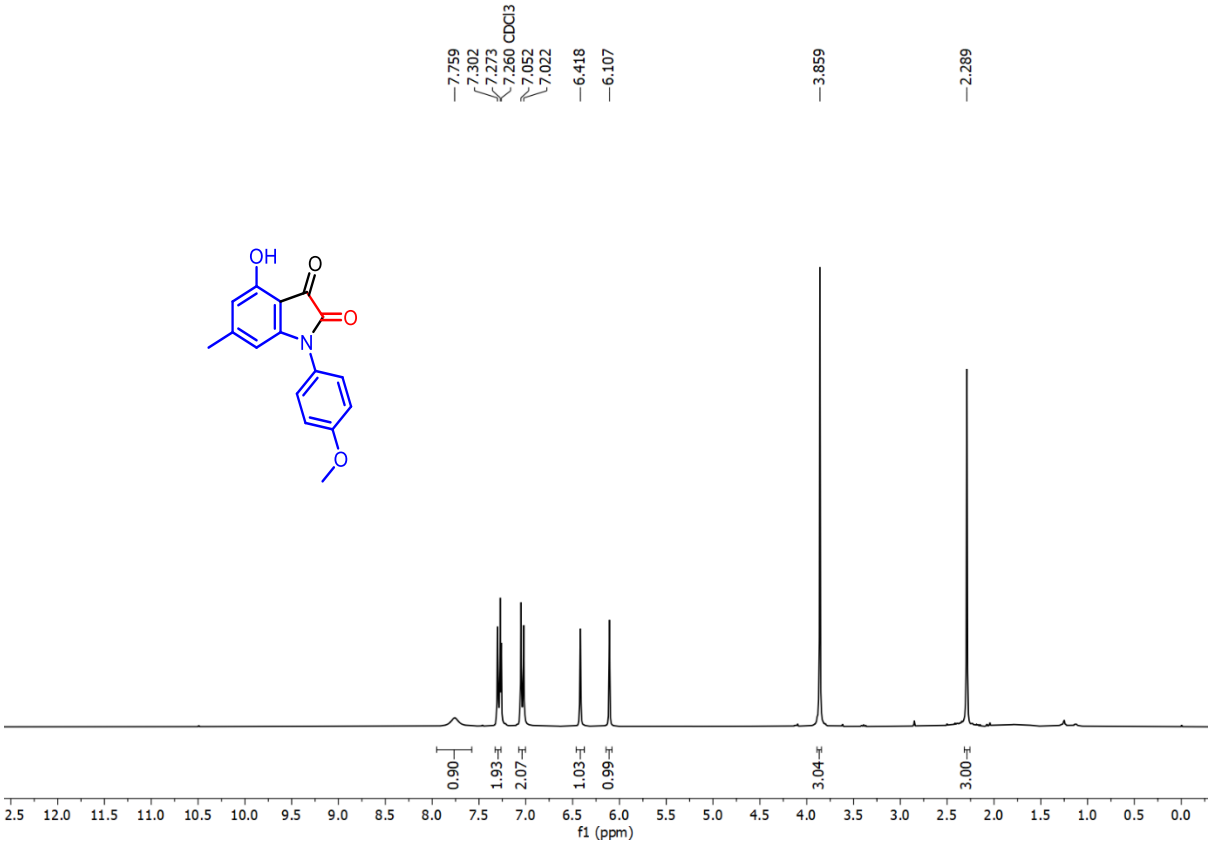
^{13}C {^{1}H} NMR Spectrum of Compound **4k** (100 MHz, $CDCl_3$)



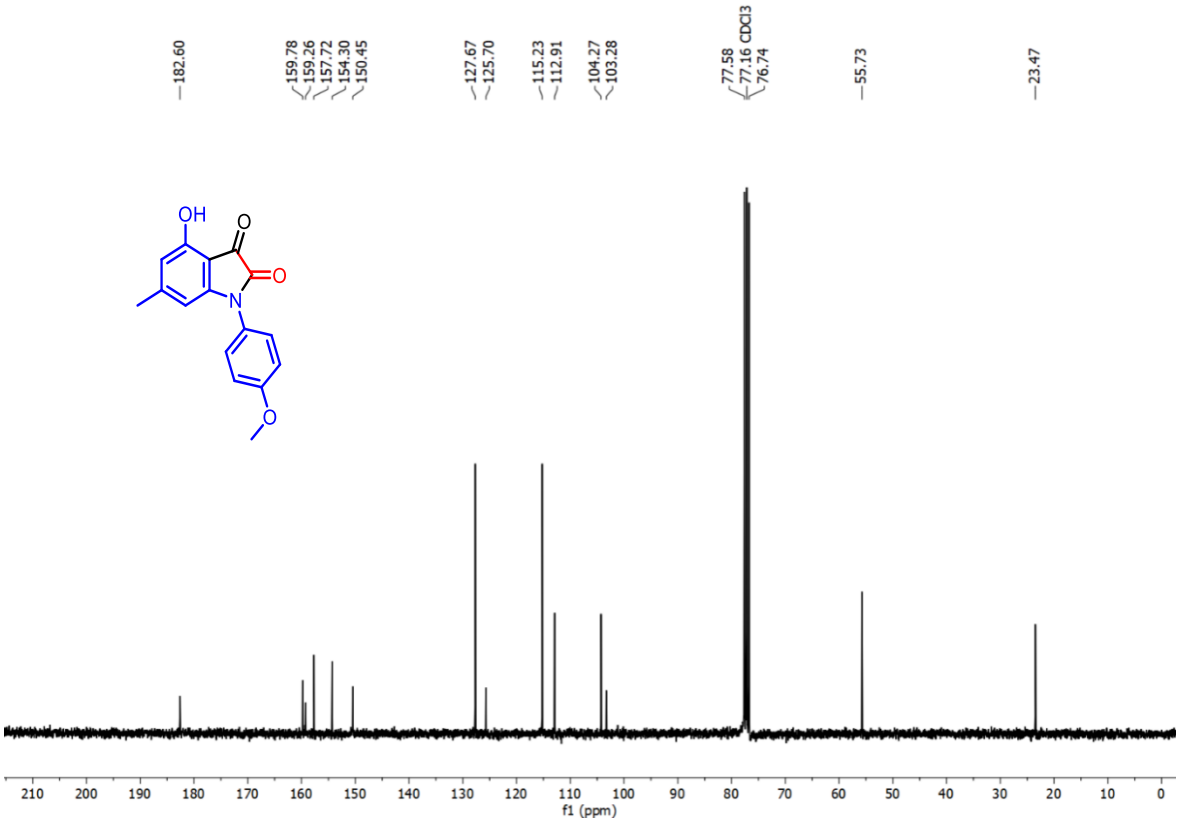
^{1}H NMR Spectrum of Compound **4l** (400 MHz, DMSO-d_6)



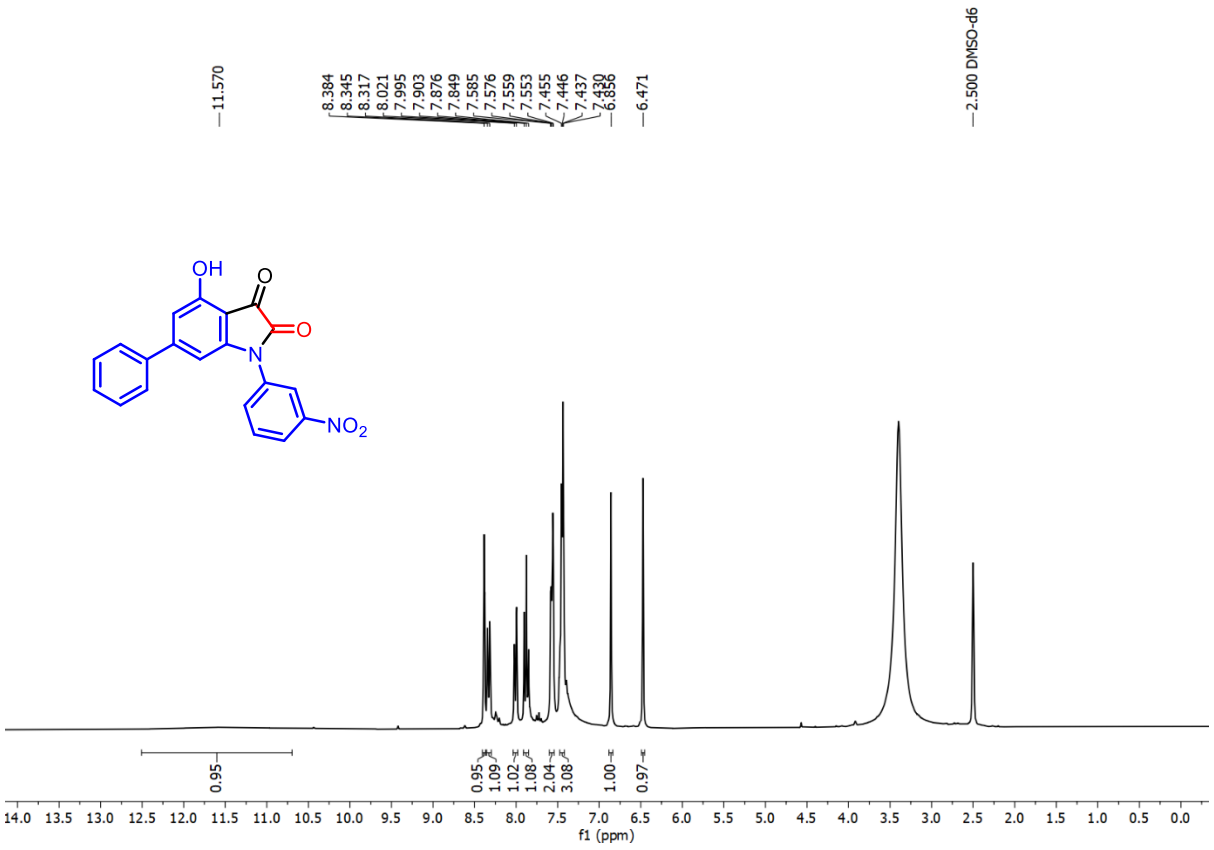
^{13}C {^{1}H} NMR Spectrum of Compound **4l** (100 MHz, DMSO-d_6)



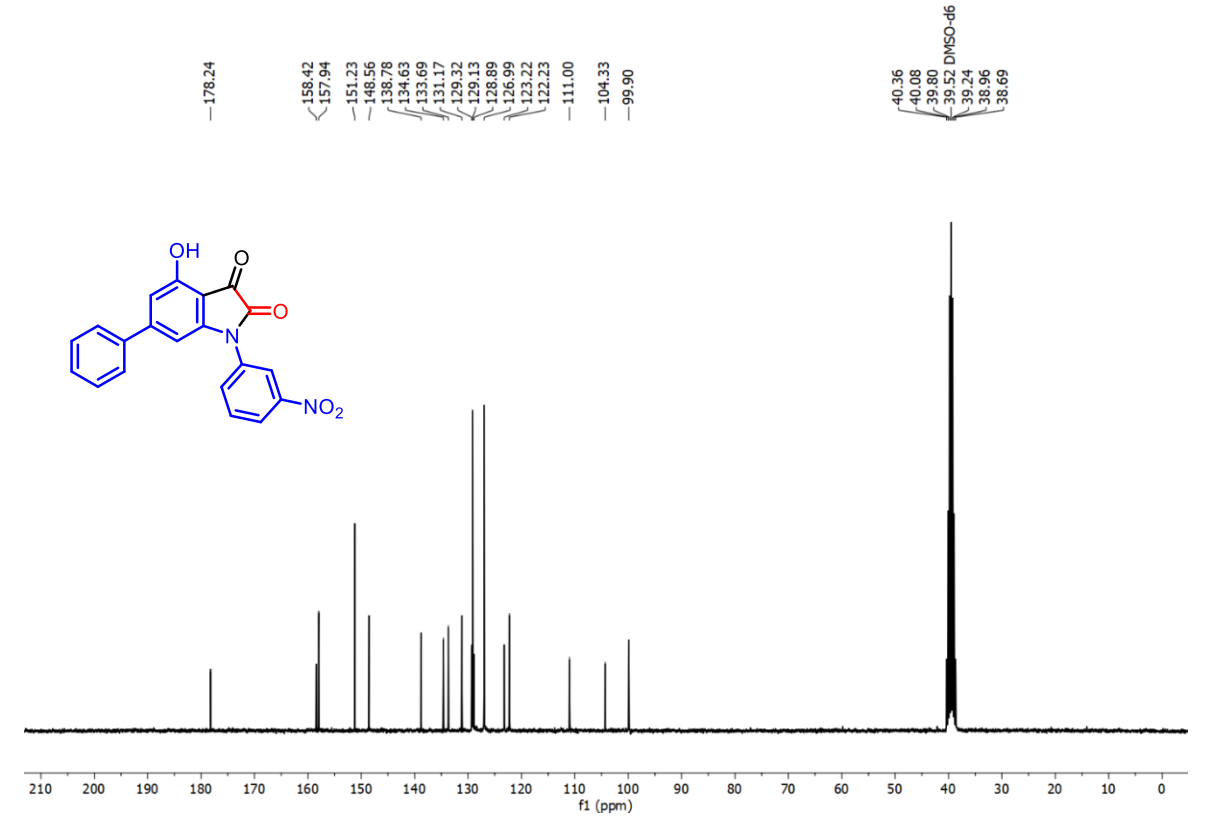
^{1}H NMR Spectrum of Compound **4m** (300 MHz, $CDCl_3$)



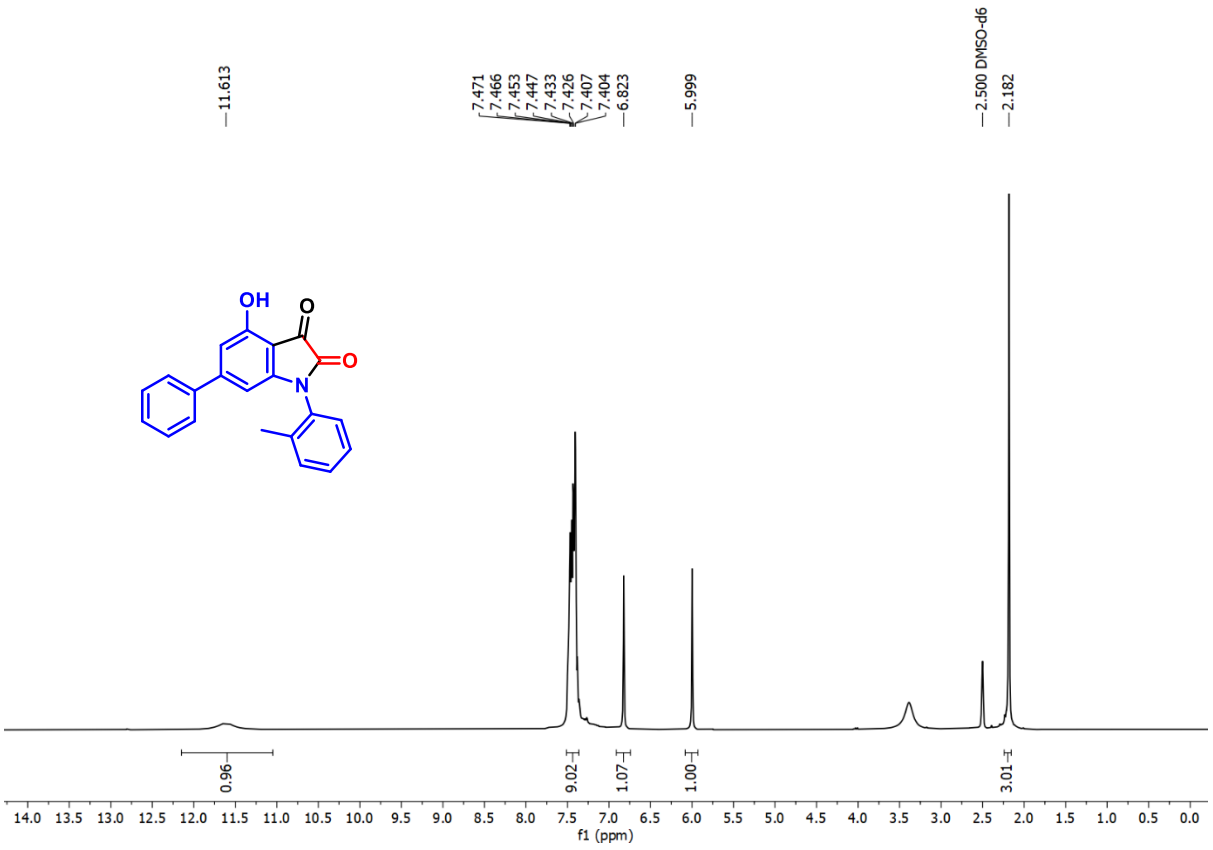
^{13}C {^{1}H} NMR Spectrum of Compound **4m** (75 MHz, $CDCl_3$)



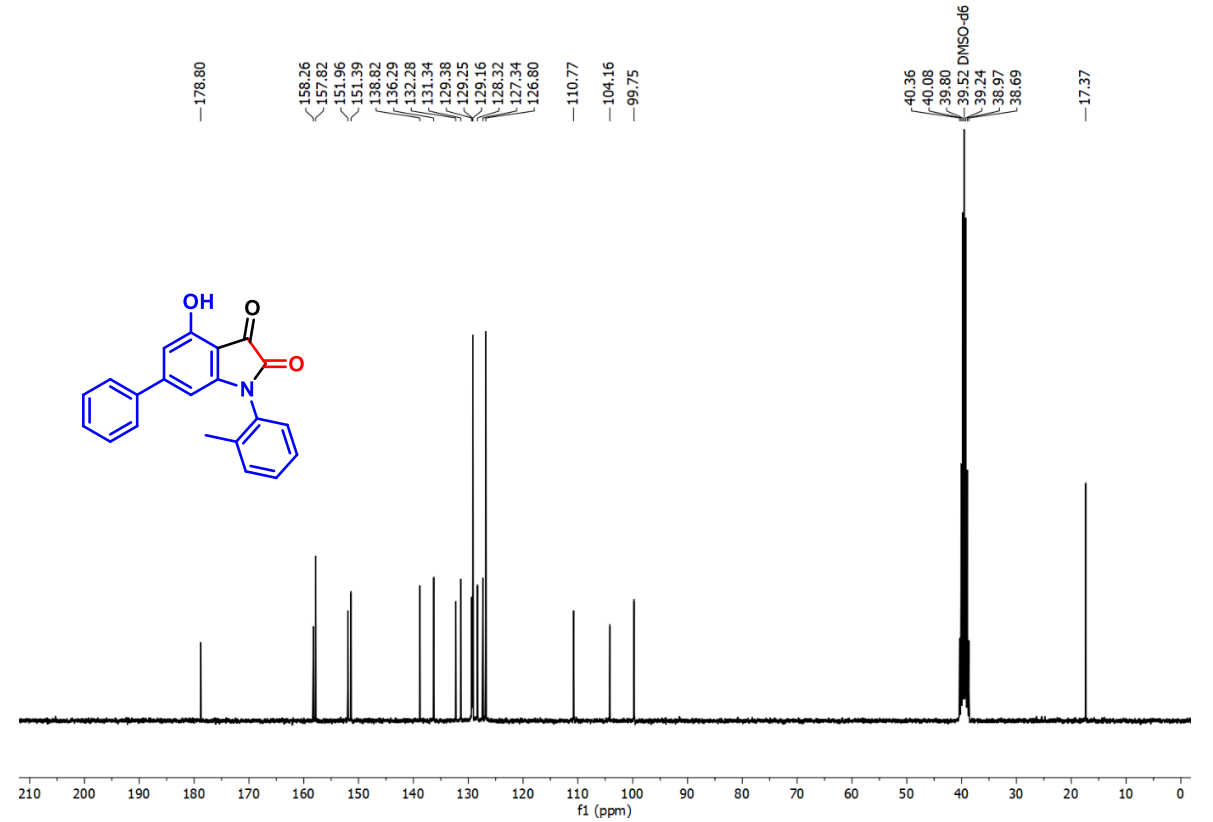
^{1}H NMR Spectrum of Compound **4n** (300 MHz, DMSO-d_6)



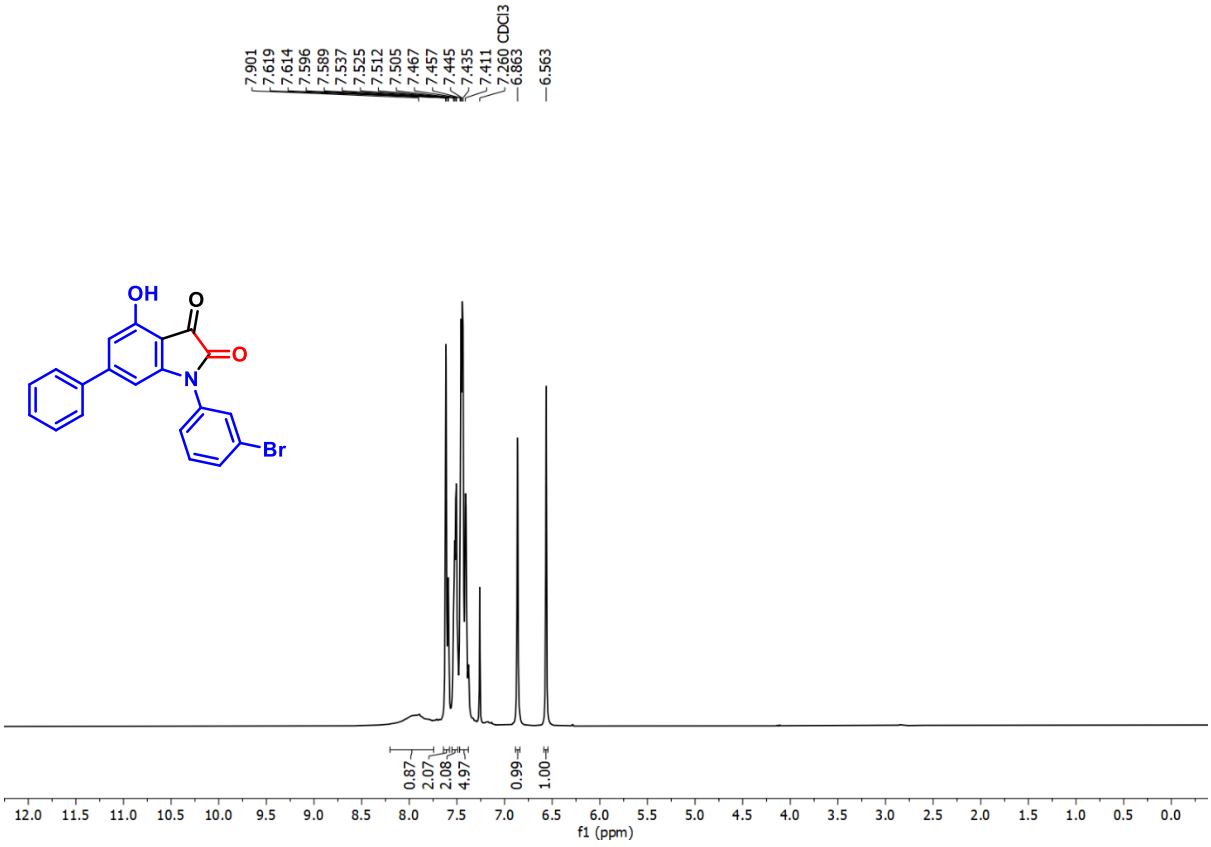
^{13}C {^{1}H} NMR Spectrum of Compound **4n** (75 MHz, DMSO-d_6)



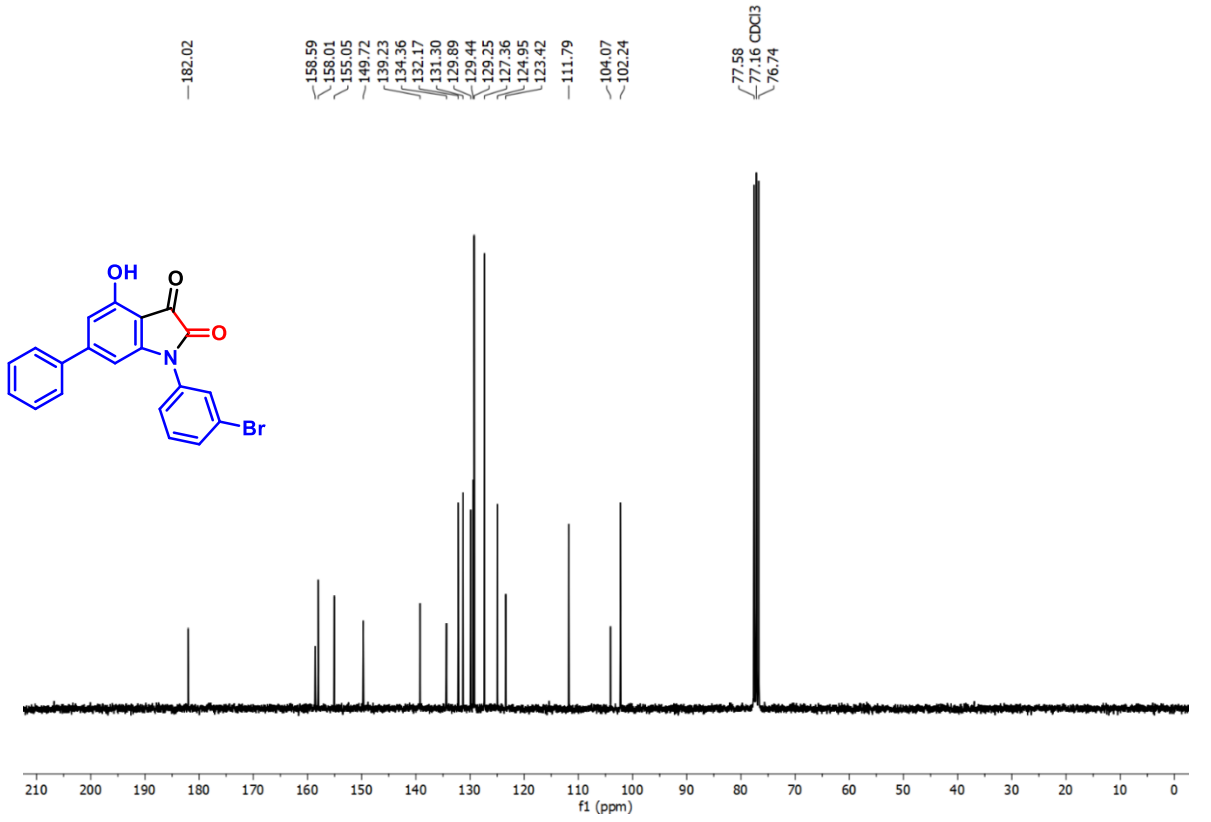
^{1}H NMR Spectrum of Compound **4o** (300 MHz, DMSO-d$_6$)



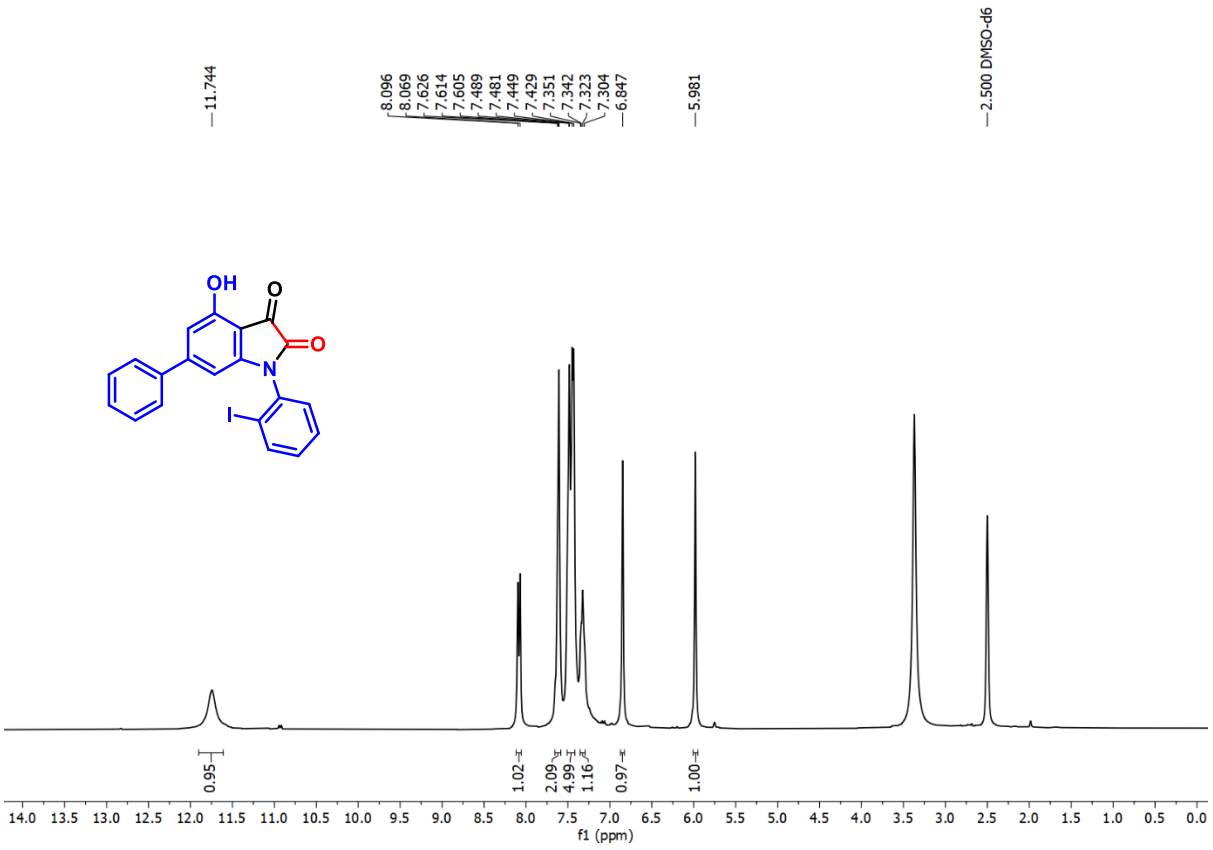
^{13}C {^{1}H} NMR Spectrum of Compound **4o** (75 MHz, DMSO-d$_6$)



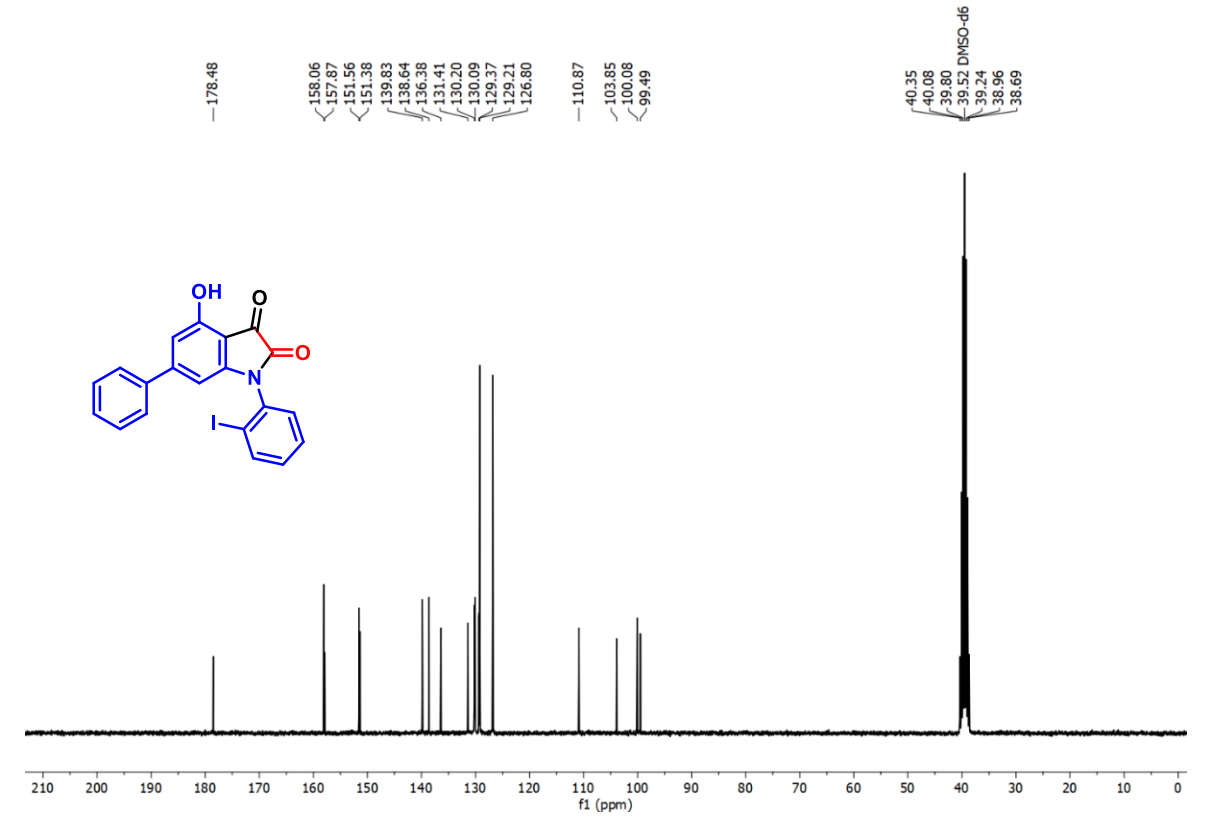
^{1}H NMR Spectrum of Compound **4p** (300 MHz, $CDCl_3$)



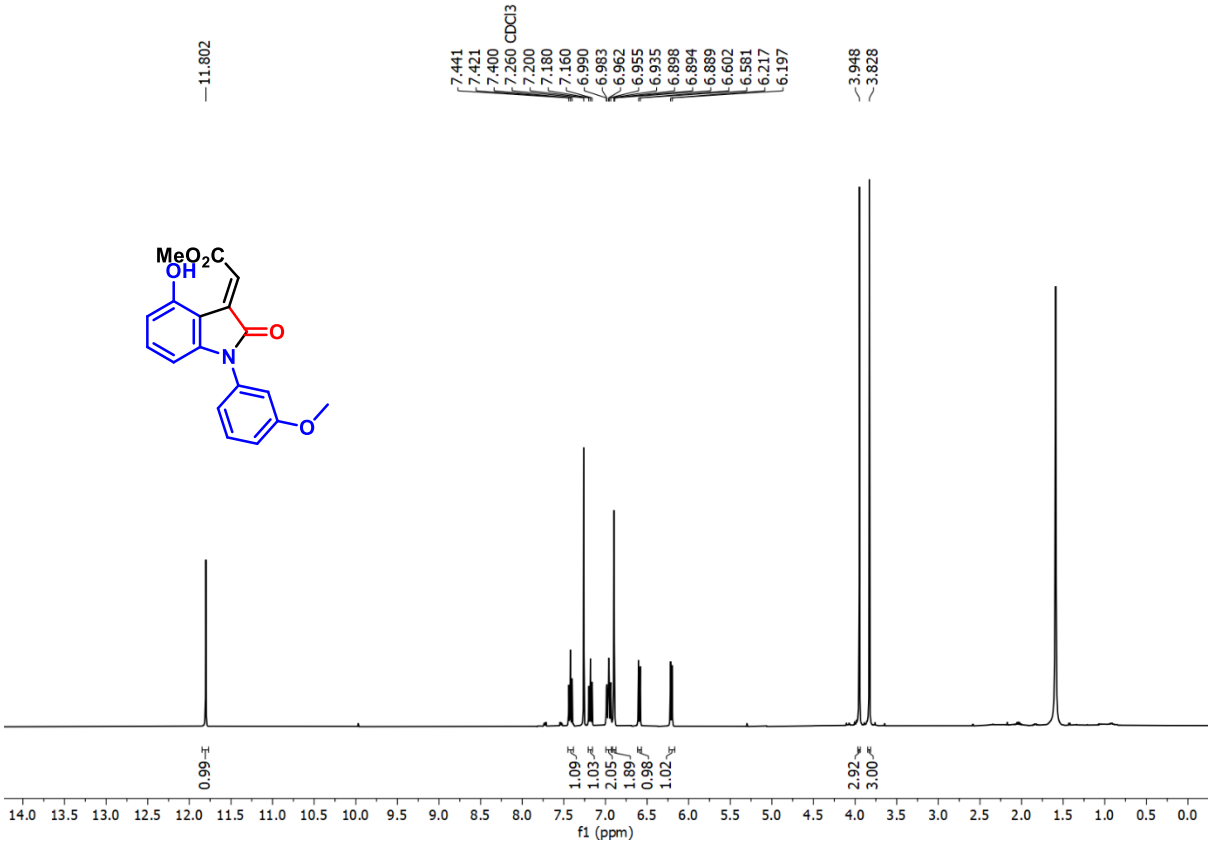
^{13}C {^{1}H} NMR Spectrum of Compound **4p** (75 MHz, $CDCl_3$)



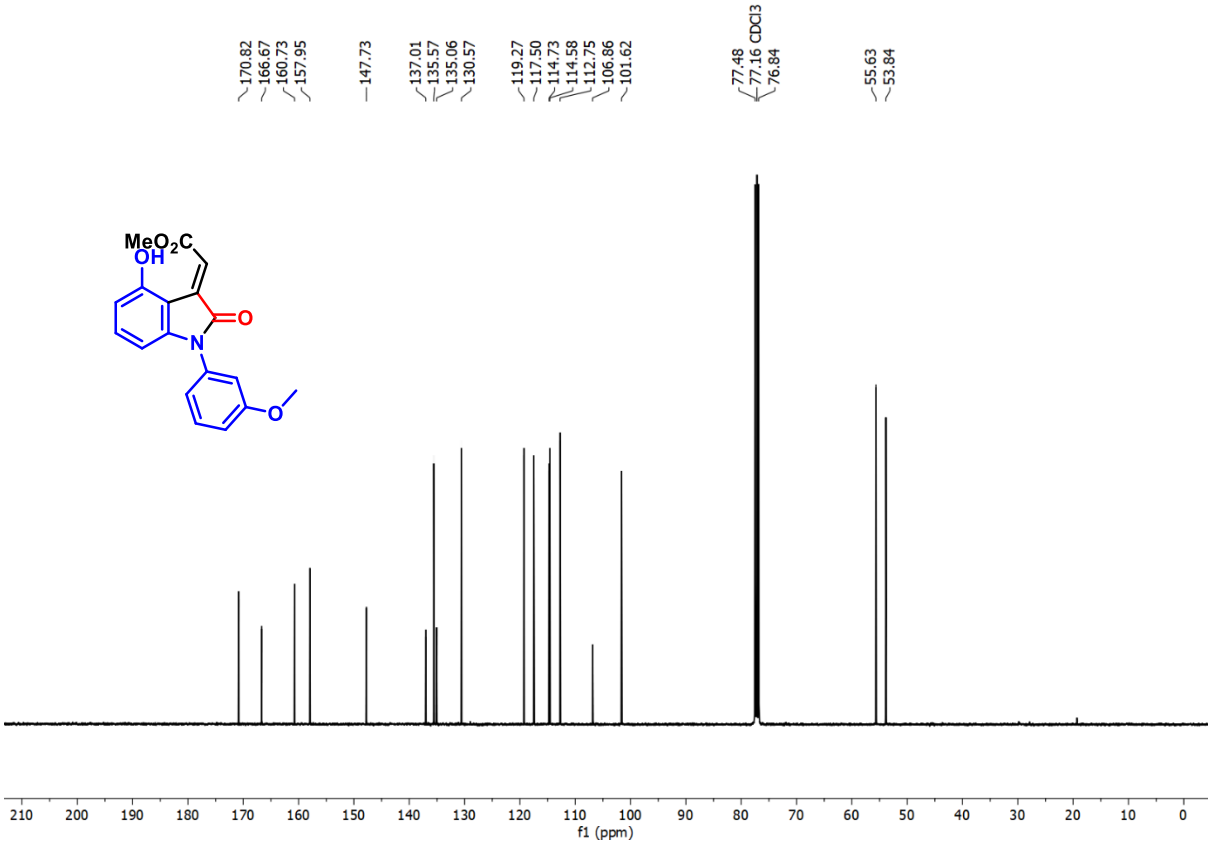
^{1}H NMR Spectrum of Compound **4q** (300 MHz, DMSO-d$_6$)



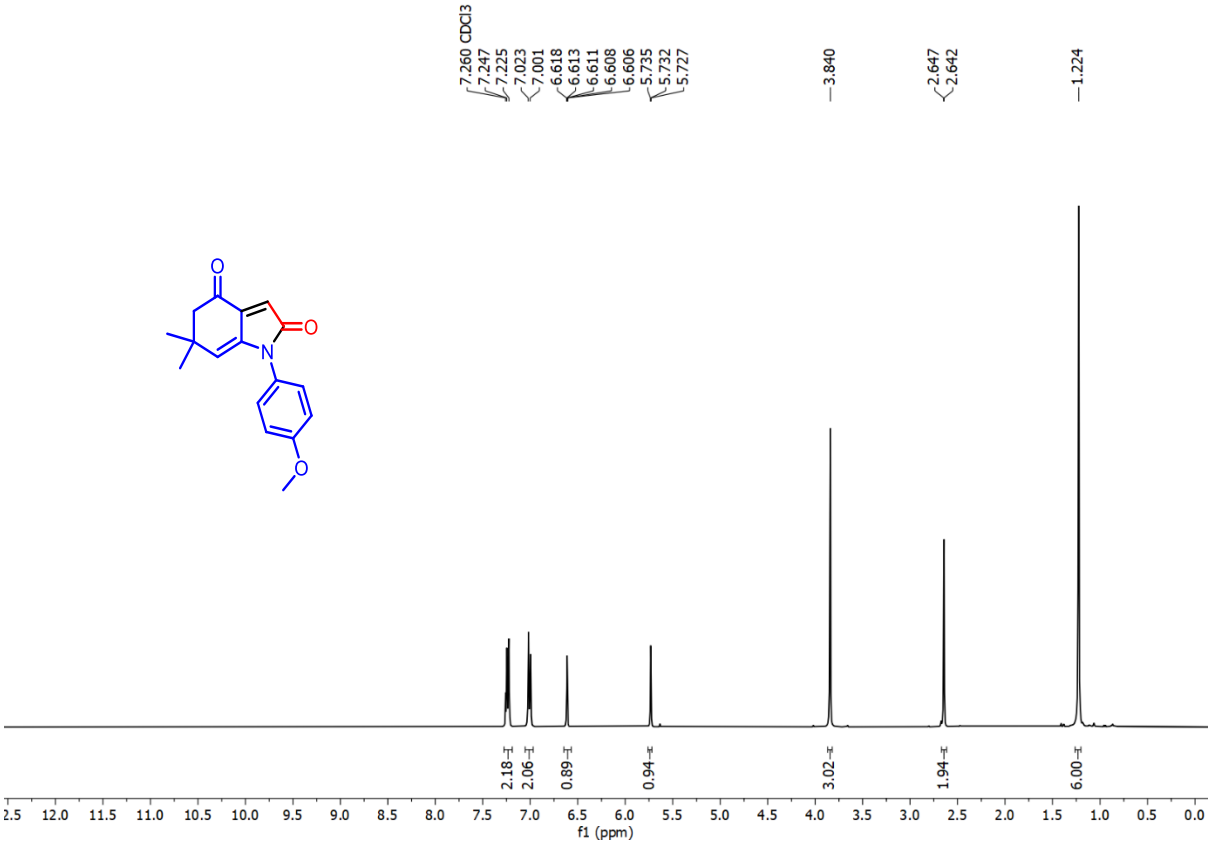
^{13}C {^{1}H} NMR Spectrum of Compound **4q** (75 MHz, DMSO-d$_6$)



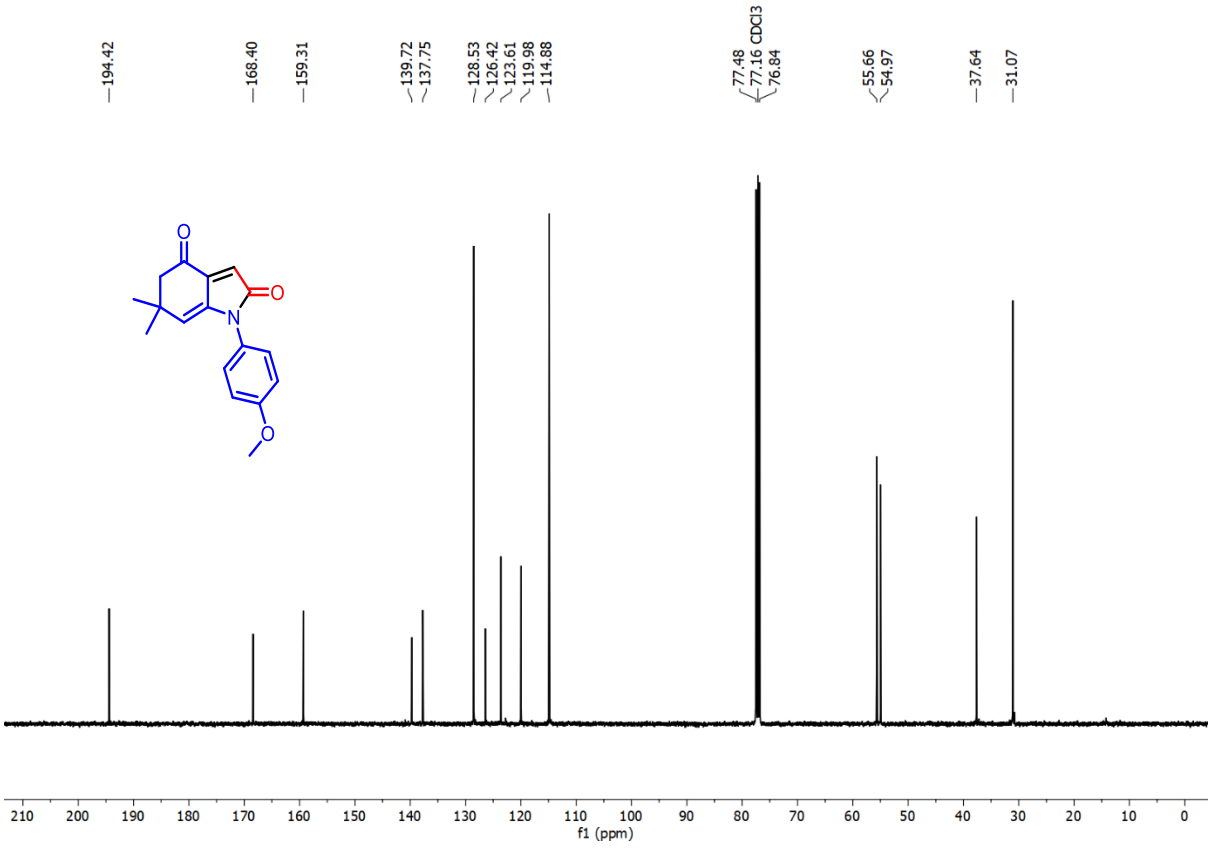
^{1}H NMR Spectrum of Compound **4ha** (400 MHz, $CDCl_3$)



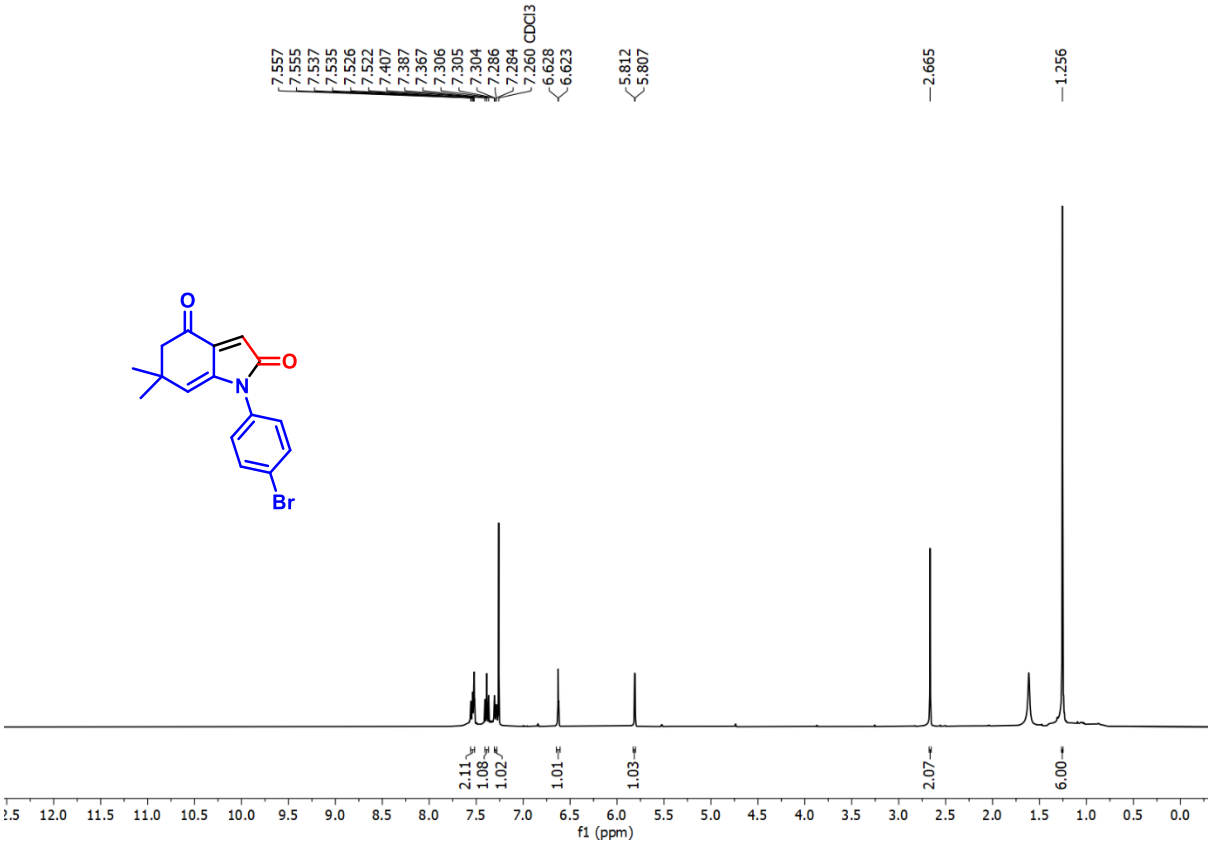
^{13}C {^{1}H} NMR Spectrum of Compound **4ha** (100 MHz, $CDCl_3$)



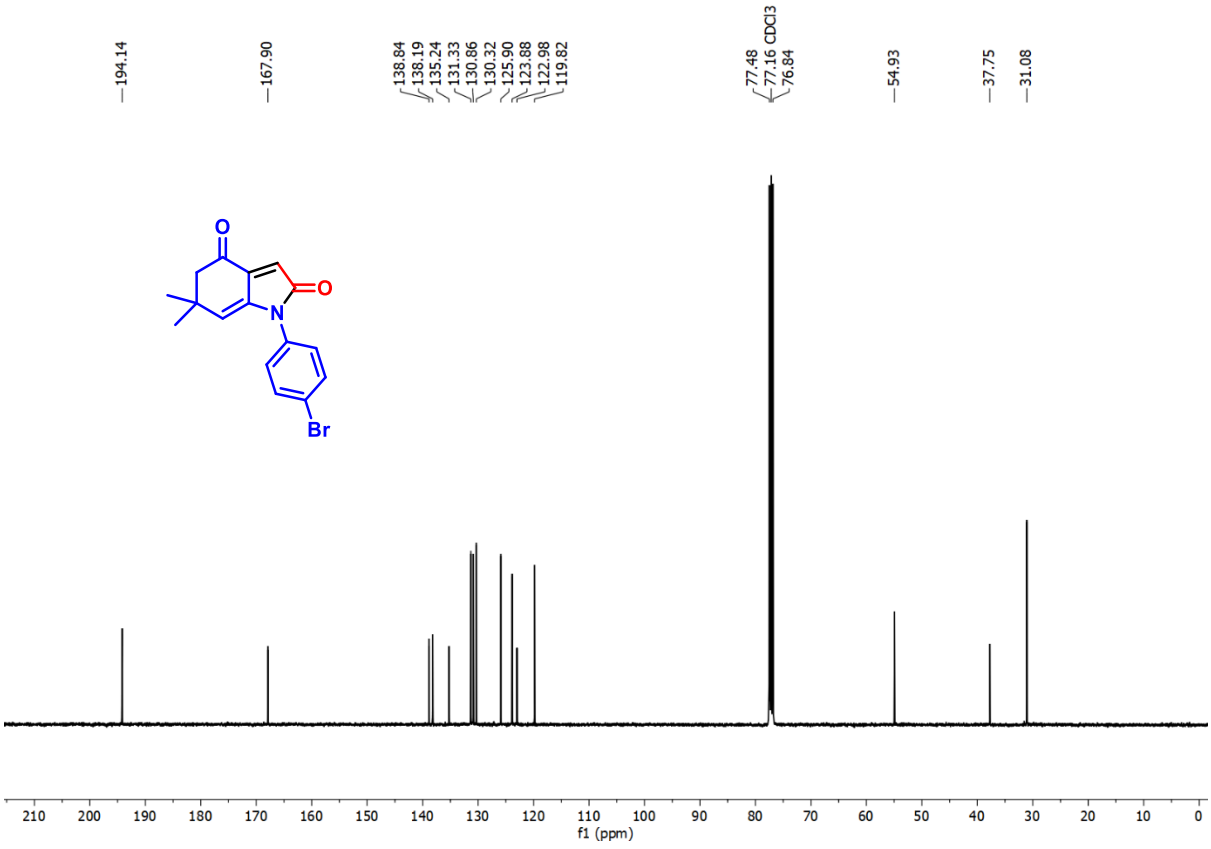
^{1}H NMR Spectrum of Compound **5a** (400 MHz, $CDCl_3$)



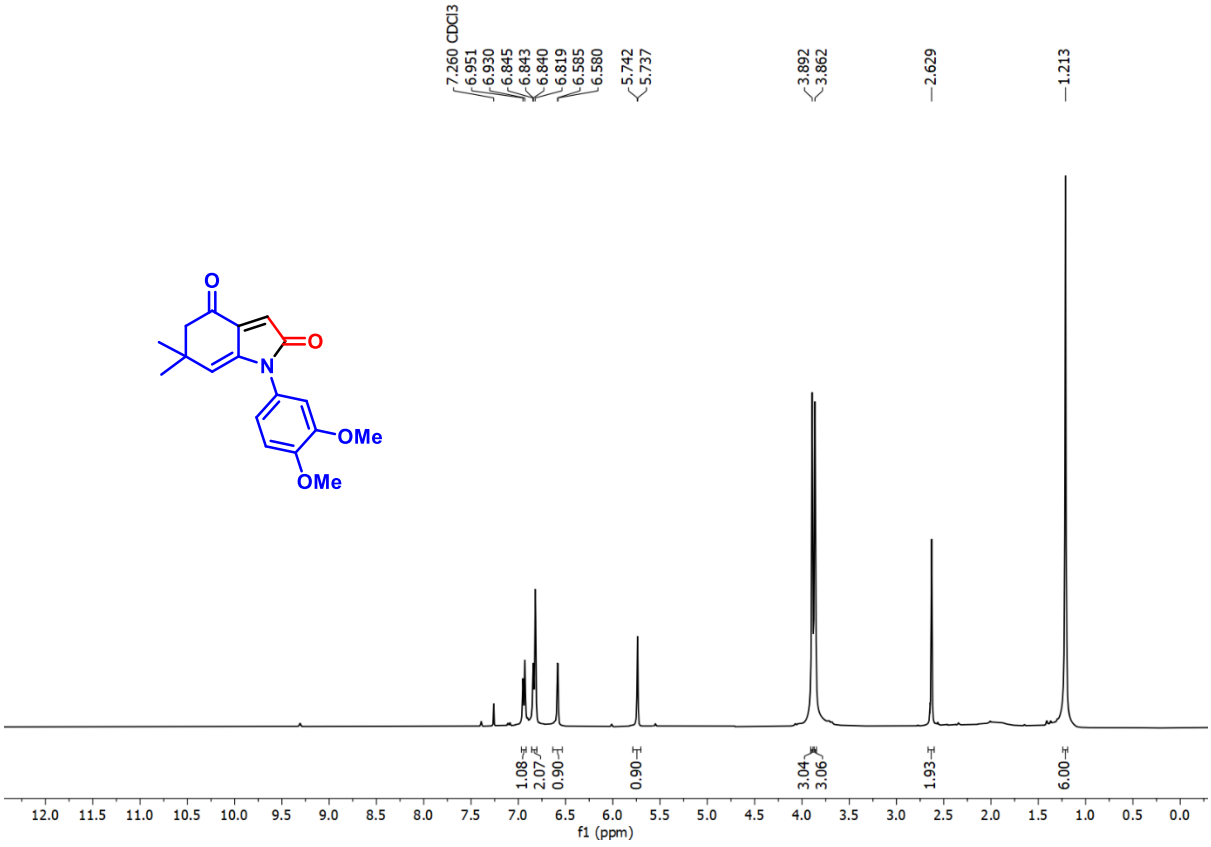
^{13}C {^{1}H} NMR Spectrum of Compound **5a** (100 MHz, $CDCl_3$)



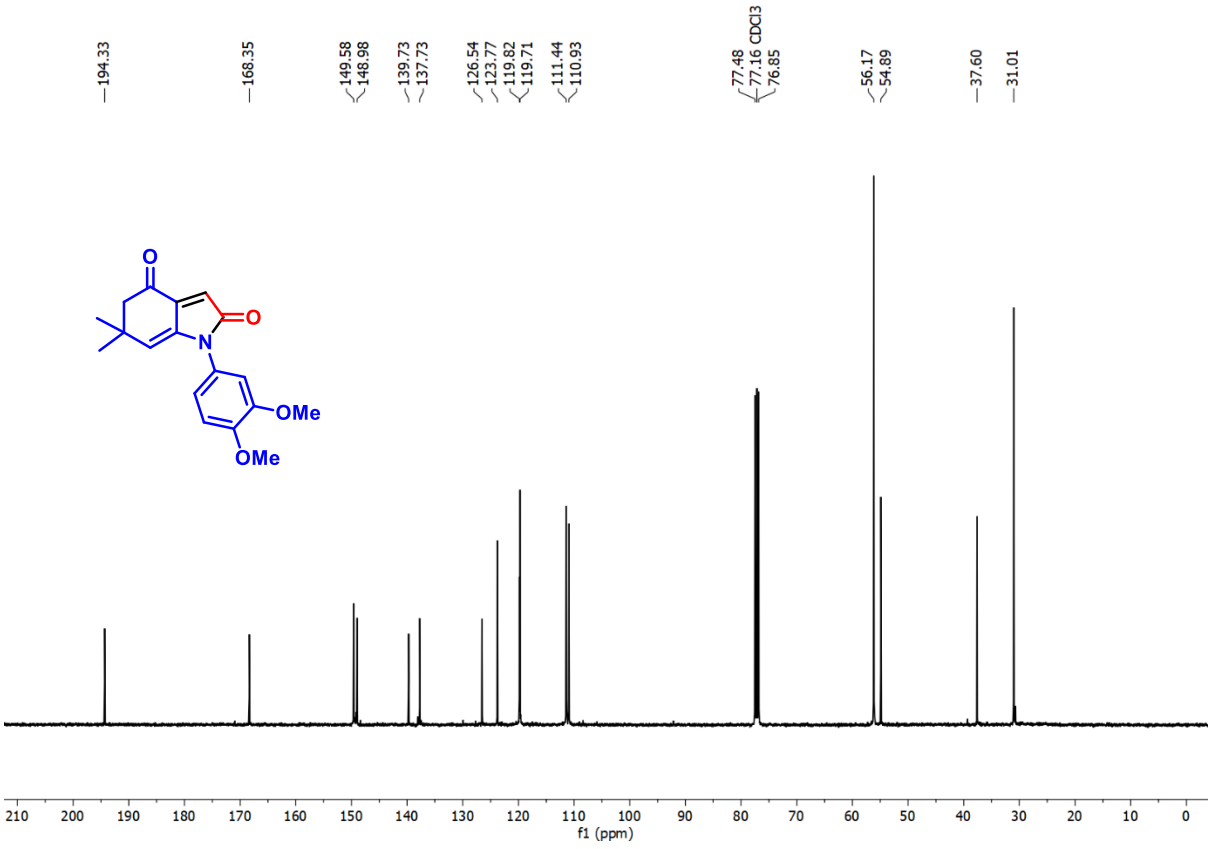
^{1}H NMR Spectrum of Compound **5b** (400 MHz, $CDCl_3$)



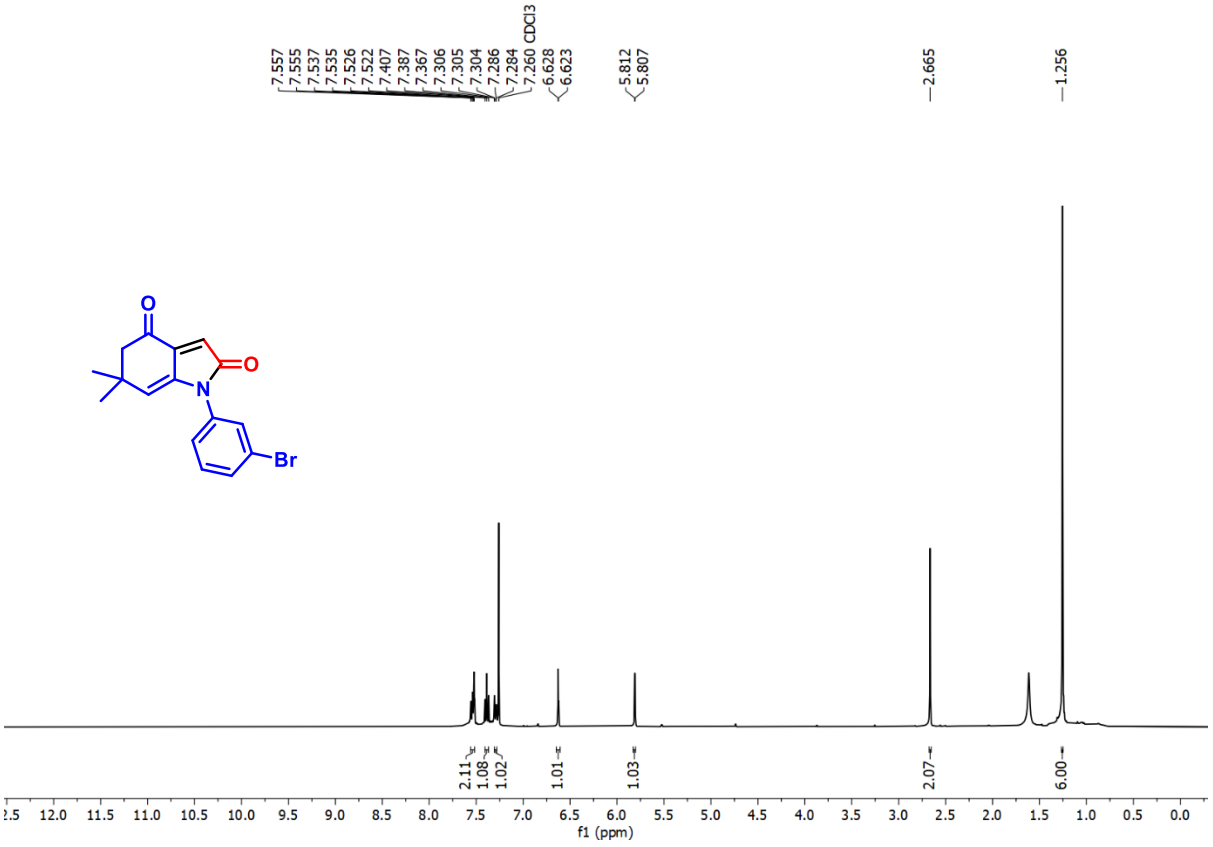
^{13}C {^{1}H} NMR Spectrum of Compound **5b** (100 MHz, $CDCl_3$)



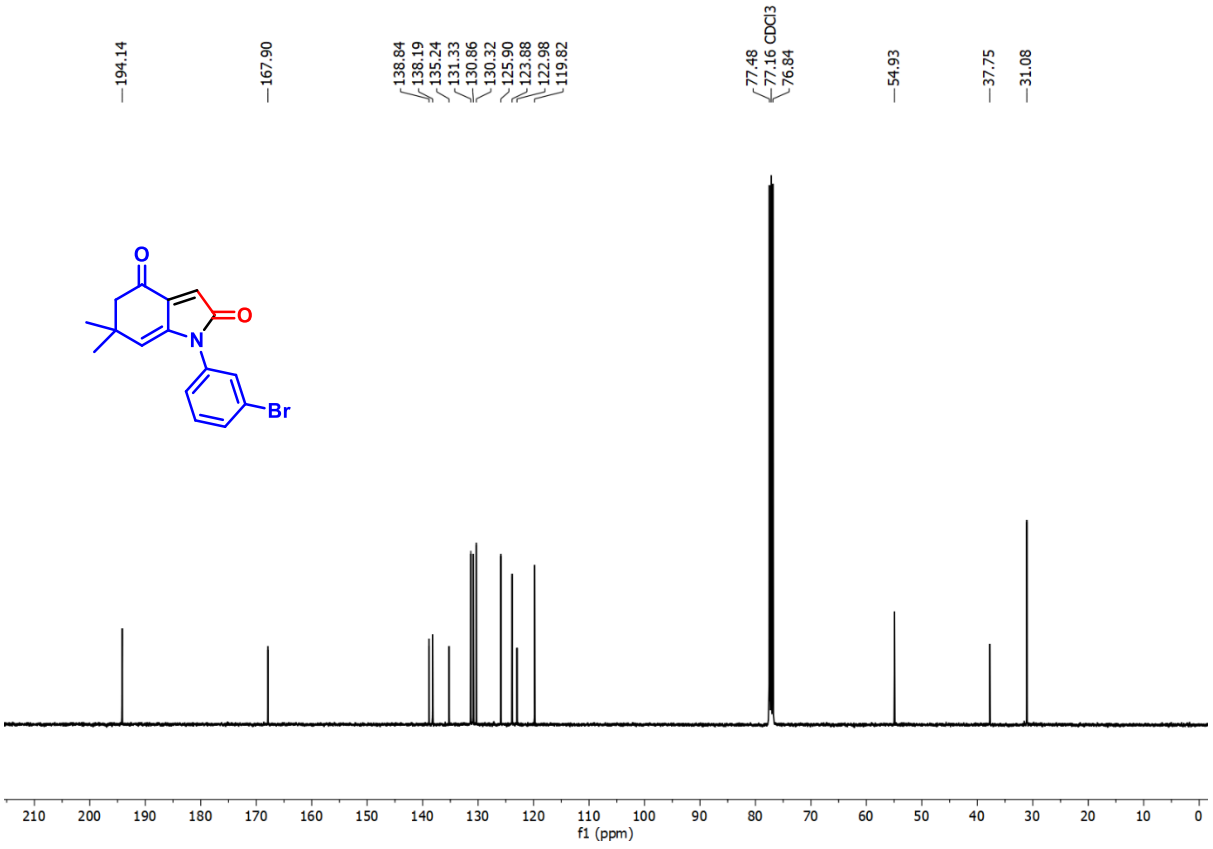
^{1}H NMR Spectrum of Compound **5c** (400 MHz, $CDCl_3$)



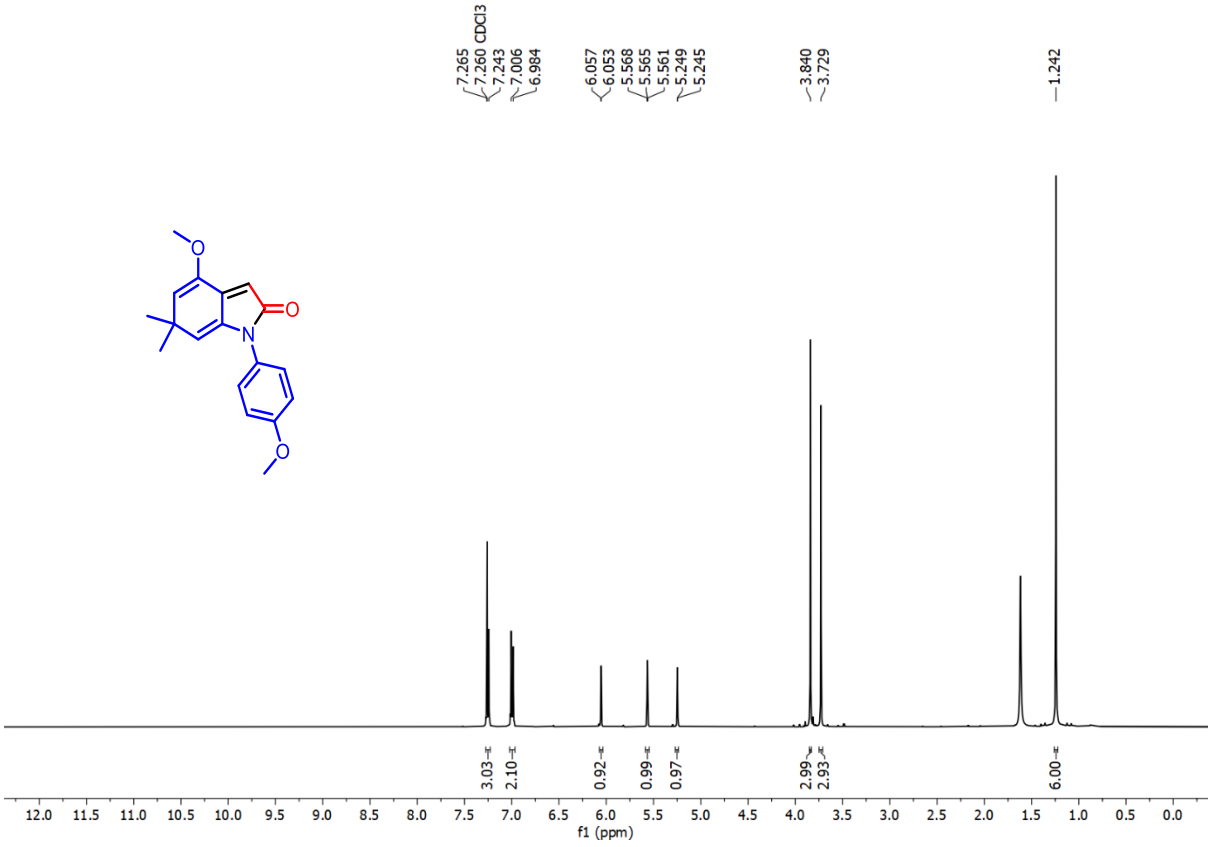
^{13}C {^{1}H} NMR Spectrum of Compound **5c** (100 MHz, $CDCl_3$)



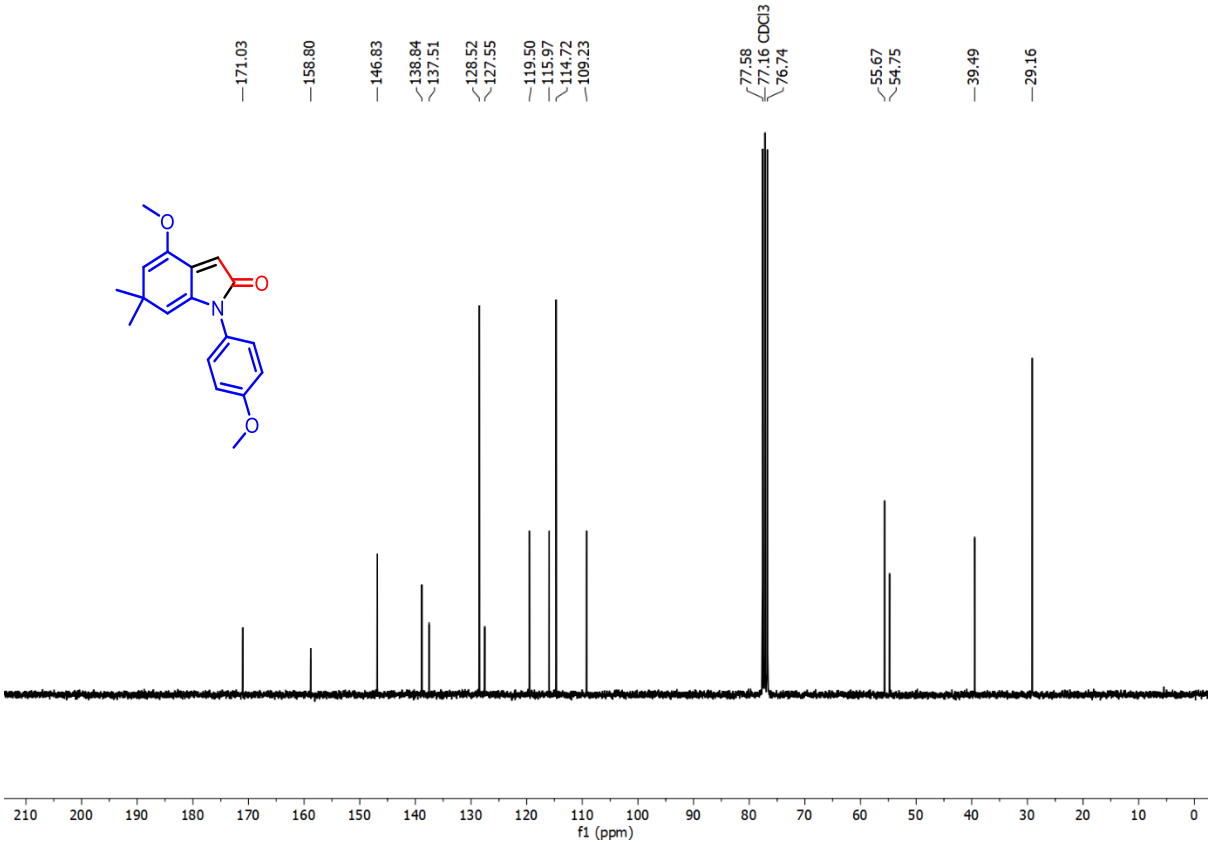
^{1}H NMR Spectrum of Compound **5d** (400 MHz, $CDCl_3$)



^{13}C {^{1}H} NMR Spectrum of Compound **5d** (100 MHz, $CDCl_3$)



^{1}H NMR Spectrum of Compound **6a** (400 MHz, $CDCl_3$)



^{13}C {^{1}H} NMR Spectrum of Compound **6a** (100 MHz, $CDCl_3$)

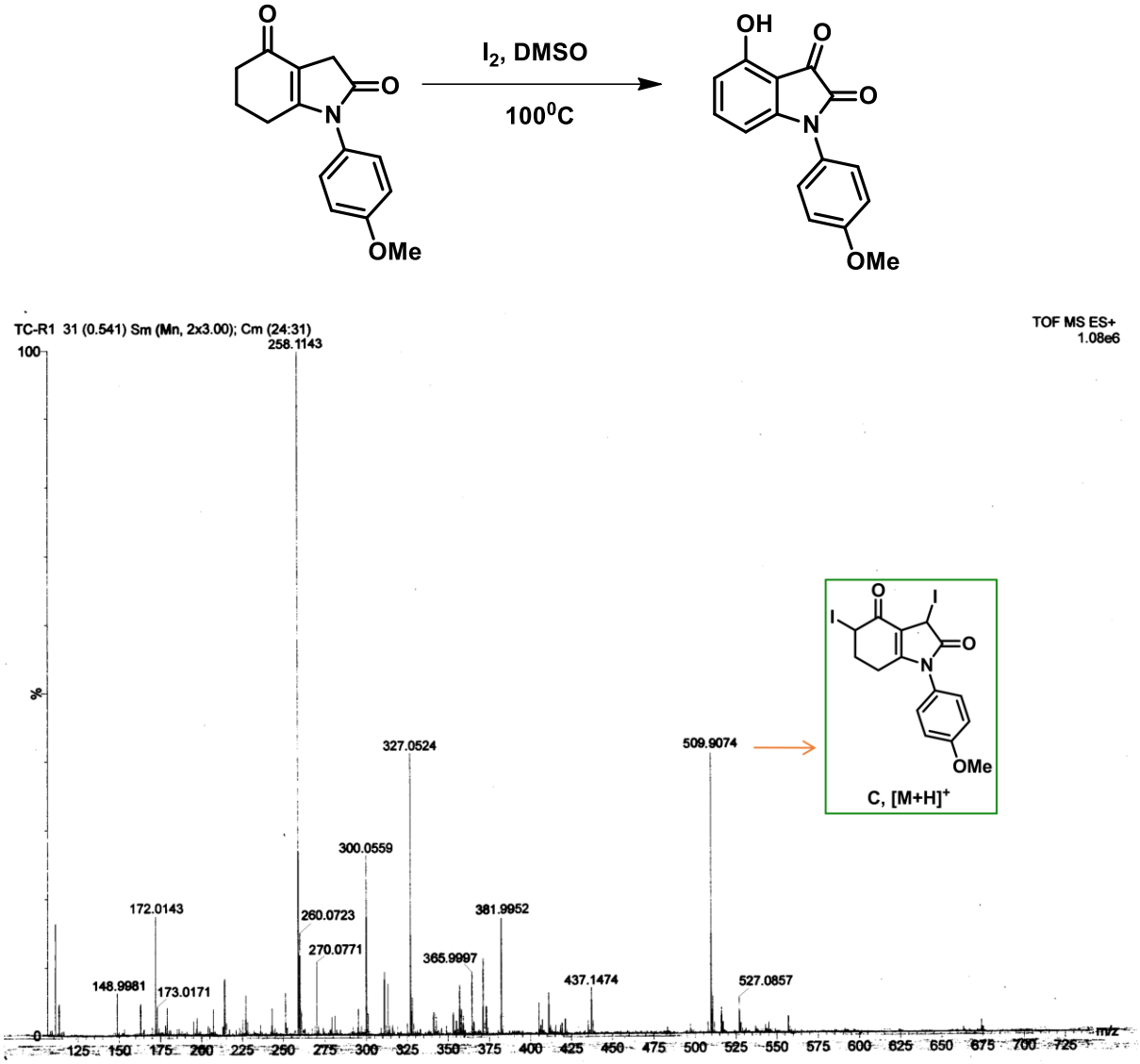
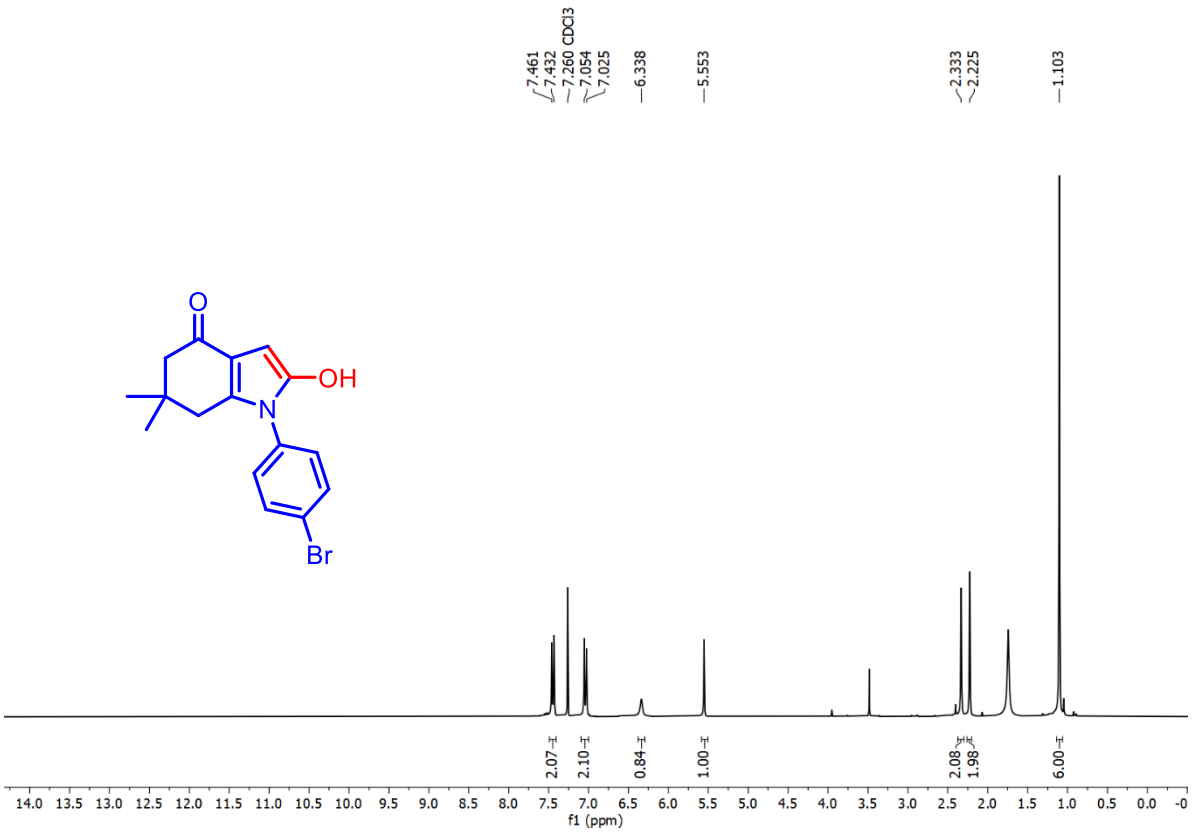
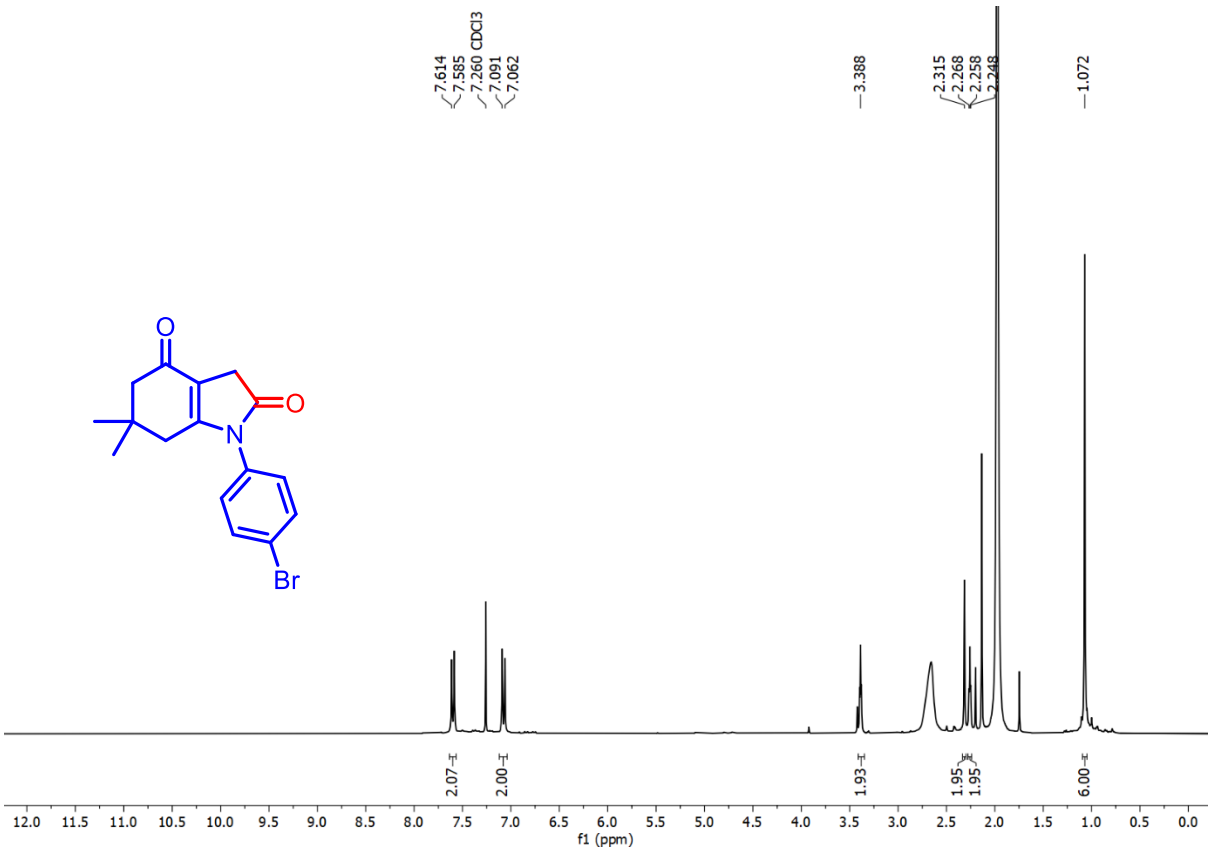


Figure S1. Crude mass of reaction mixture



Crude ^{1}H NMR Spectrum of Compound **D(Intermediate) (enol form)** (300 MHz, $CDCl_3$)



Crude ^{1}H NMR Spectrum of Compound **3**, after completion of the reaction (**keto form**) (300 MHz, $CDCl_3$)

General procedure for sample preparation for crystal growth 3ac:

The crystals were grown from MeOH solvent. The methodology is as follows:

25 mg of sample was dissolved in 0.5 mL MeOH in a vial and kept it for 4 to 5 days for growing of the crystal in open air till single crystals appeared.

General procedure for sample preparation for crystal growth 4p:

The crystals were grown from DMSO solvent. The methodology is as follows:

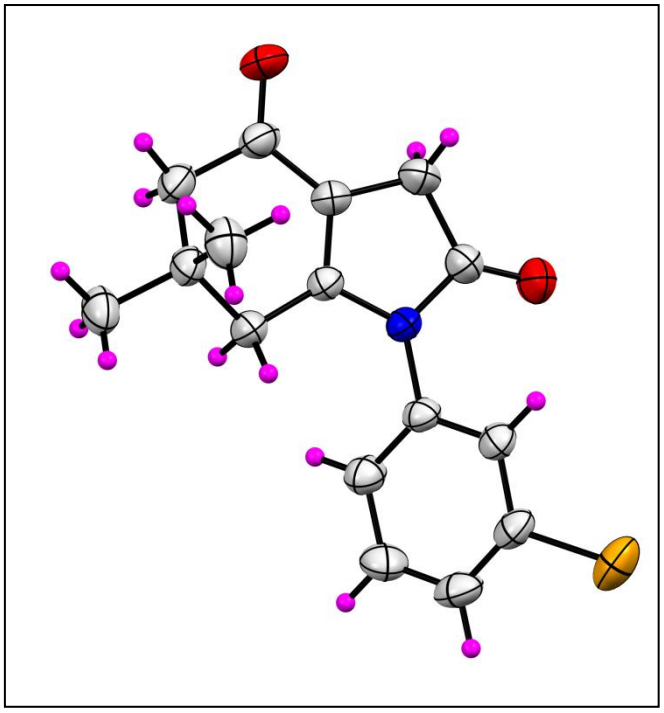
25 mg of sample was dissolved in 0.5 mL DMSO in a vial and kept it for 3 to 4 days for growing of the crystal in open air till single crystals appeared.

General procedure for sample preparation for crystal growth 6a:

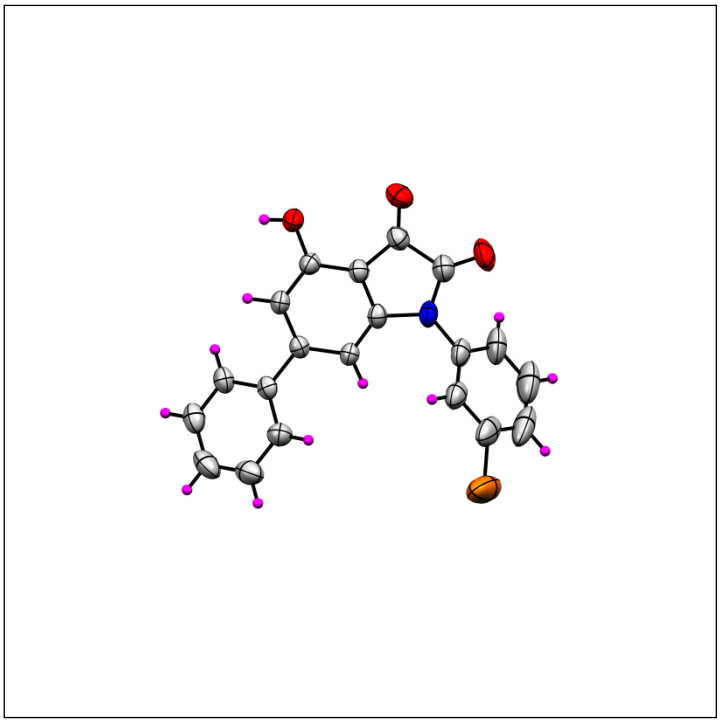
The crystals were grown from MeOH solvent. The methodology is as follows:

25 mg of sample was dissolved in 0.5 mL MeOH in a vial and kept it for 3 to 4 days for growing of the crystal in open air till single crystals appeared.

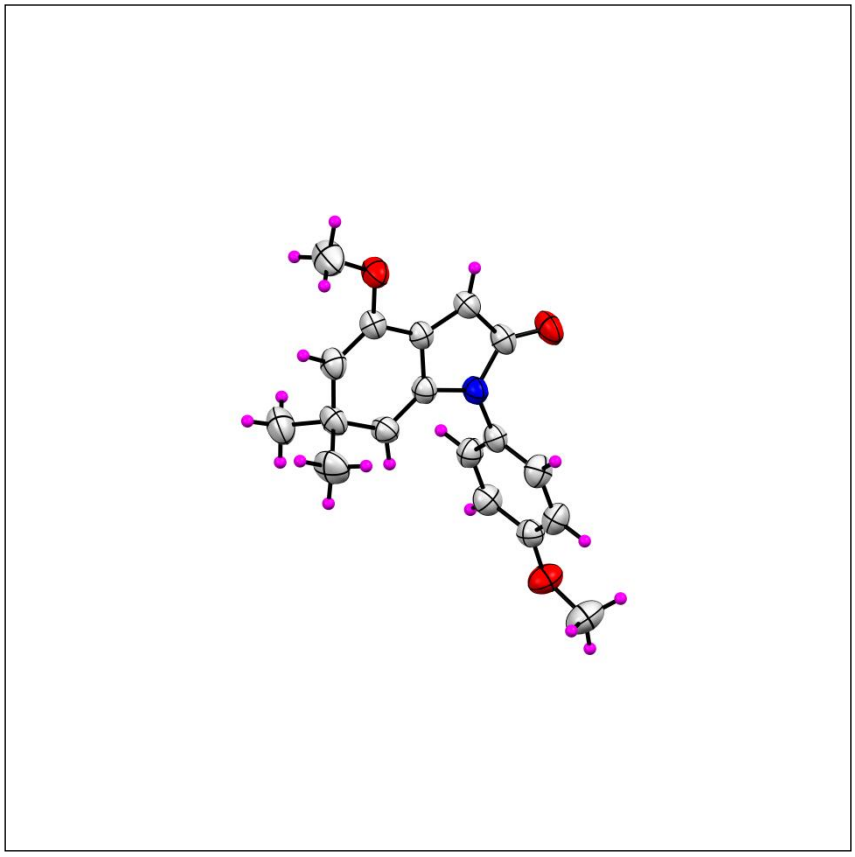
ORTEP Diagram of crystals



ORTEP diagram of product **3ac (CCDC NO. 2306270)**



ORTEP diagram of product **4p (CCDC NO. 2306269)**



ORTEP diagram of product **6a (CCDC NO. 2306506)**

Detailed analysis data of the X-ray crystal structures of compound3ac, 4p and compound6a

Complexes	**3ac**	**4p**	**6a**
CCDC	2306270	2306269	2306506
Formula	$C_{16}H_{16}BrNO_2$	$C_{20}H_{12}BrNO_3$	$C_{18}H_{19}NO_3$
Fw	334.21	394.227	297.34
crystal color	Clear light orange	metallic light brown	clear light yellow
crystal system	monoclinic	monoclinic	monoclinic
space group	P 1 21/c 1	P 1 21/c 1	C 1 2/c 1
a (Å)	11.3095(2)	16.47(3)	22.7753(7)
b (Å)	10.9116(2)	15.27(2)	9.6966(3)
c(Å)	13.1608(3)	6.904(10)	17.8720(9)
α (°)	90	90	90
β (°)	114.8270(10)	100.65(7)	126.1900(10)
γ (°)	90	90	90
V (Å^3)	1474.00(5)	1706(4)	3185.4(2)
Z	4	4	8
T (K)	273.15	299.00	273.15
2θ range	5.06 to 50.26	5.066 to 50.278	4.81 to 51.448
calcd (g cm^{-3})	1.506	1.535	1.240
ReflectionsCollected	3816	3751	3024
Radiation [Å]	MoK\a	Mo K\a	MoK\a
reflection (I>2σ(I))	2894	2081	2301
λ (Å)/μ (mm^{-1})	0.71073/2.789	0.71073/ 2.428	0.71073/0.084
F(000)	680	791.684	1264
R1[a][I>2σ(I)]/GOF [b]	0.0324 /1.027	0.0680/1.0367	0.0638/1.117
R1[a] (all data)	0.0512	0.1277	0.0864
*w*R2[c] (I>2σ (I))	0.0730	0.1914	0.1277
no. of para-meters/restr.	183/0	227/0	203/0
h, k, l (max)	15, 14, 17	20, 19, 8	27, 11, 21