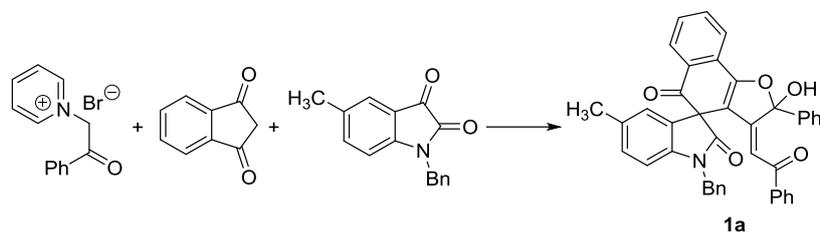


Selective synthesis of spirooxindoles via one-pot two-step reaction of *N*-phenacylpyridinium bromide, indane-1,3-dione and *N*-alkylisatins

Jing Sun, Yu Zhang, Rong-Guo Shi, Chao-Guo Yan*

Supporting Information

Table s1 optimizing reaction conditions	2
Single crystal structures (Fig. s1-s2)	3
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Table s1 optimizing reaction conditions ^a

Entry	Base	Solvent	Time (h)	T(°C)	Yield (%) ^b
1	Et ₃ N	EtOH	12	20	0
2	Et ₃ N	EtOH	12	40	0
3	Et ₃ N	EtOH	12	60	15
4	Et ₃ N	EtOH	8	reflux	22
5	Et ₃ N	EtOH	8	reflux	35 ^c
6	Et ₃ N	MeOH	10	reflux	32 ^c
7	Et ₃ N	THF	8	reflux	52 ^c
8	Et ₃ N	MeCN	4	reflux	75 ^c
9	Et ₃ N	MeCN	6	reflux	70 ^c
10	Et ₃ N	DMF	6	reflux	30 ^c
11	DBU	MeCN	6	reflux	54 ^c
12	DABCO	MeCN	5	reflux	47 ^c
13	K ₂ CO ₃	MeCN	12	reflux	20 ^c

Reaction conditions: a. pyridinium salt (0.5 mmol), 1,3-indanedione (0.5 mmol), isatin (0.5 mmol), base (1.0 mmol),

Solvent (15.0 mL); b. Isolated yields; c. Entries 5-12: pyridinium salt (1.1 mmol), 1,3-indanedione (0.6 mmol), isatin (0.5 mmol), base (1.5 mmol).

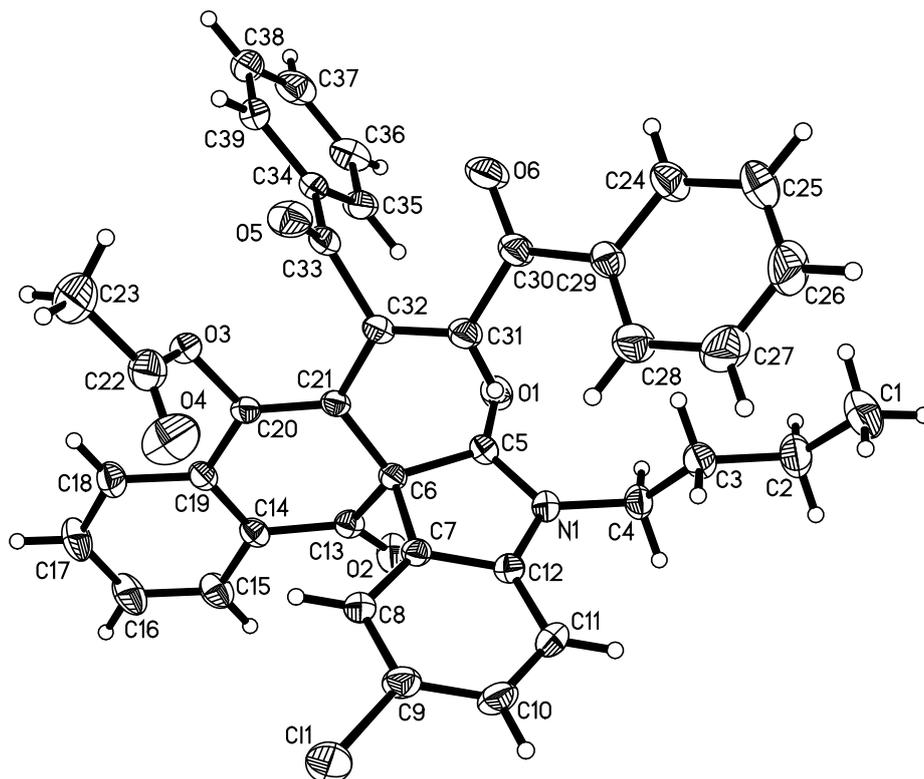


Fig. s1 ORTEP diagram of the compounds **2f**

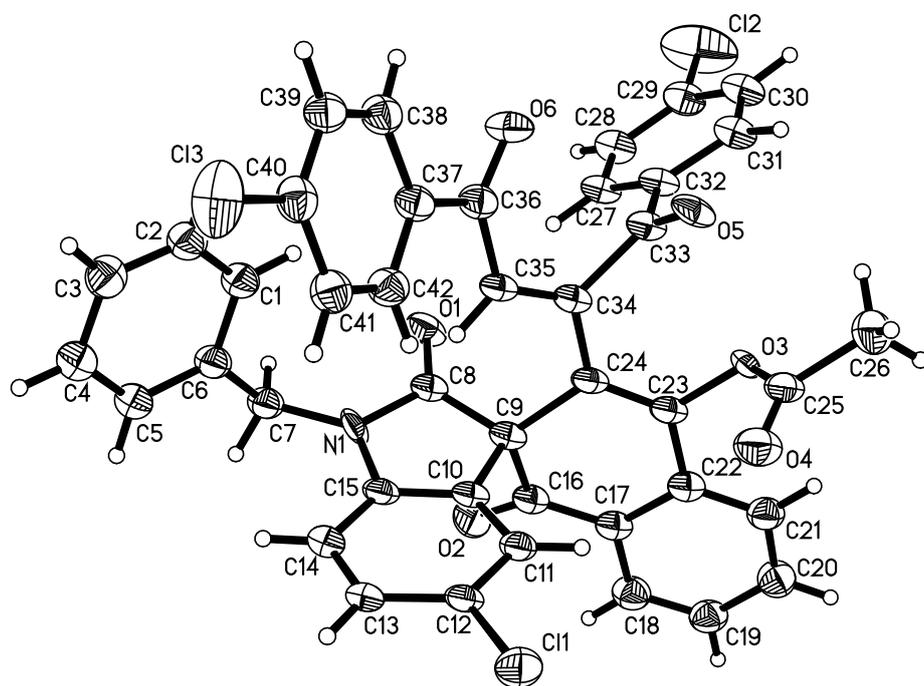
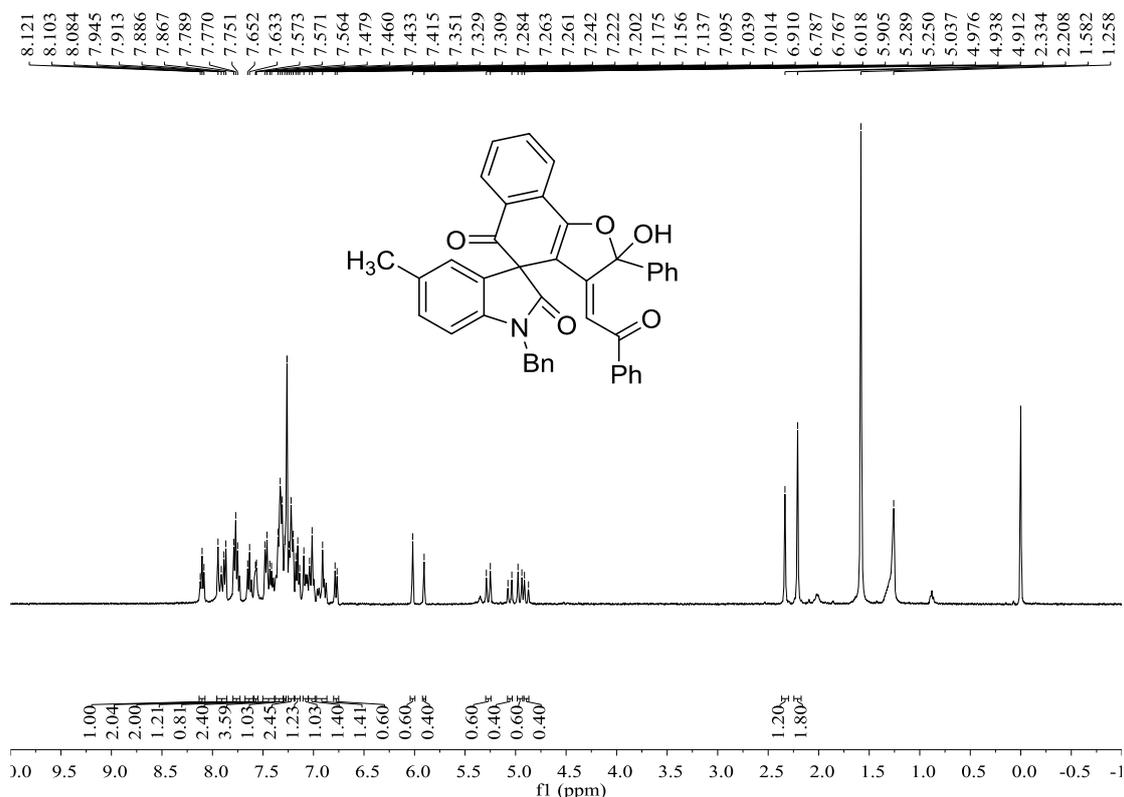
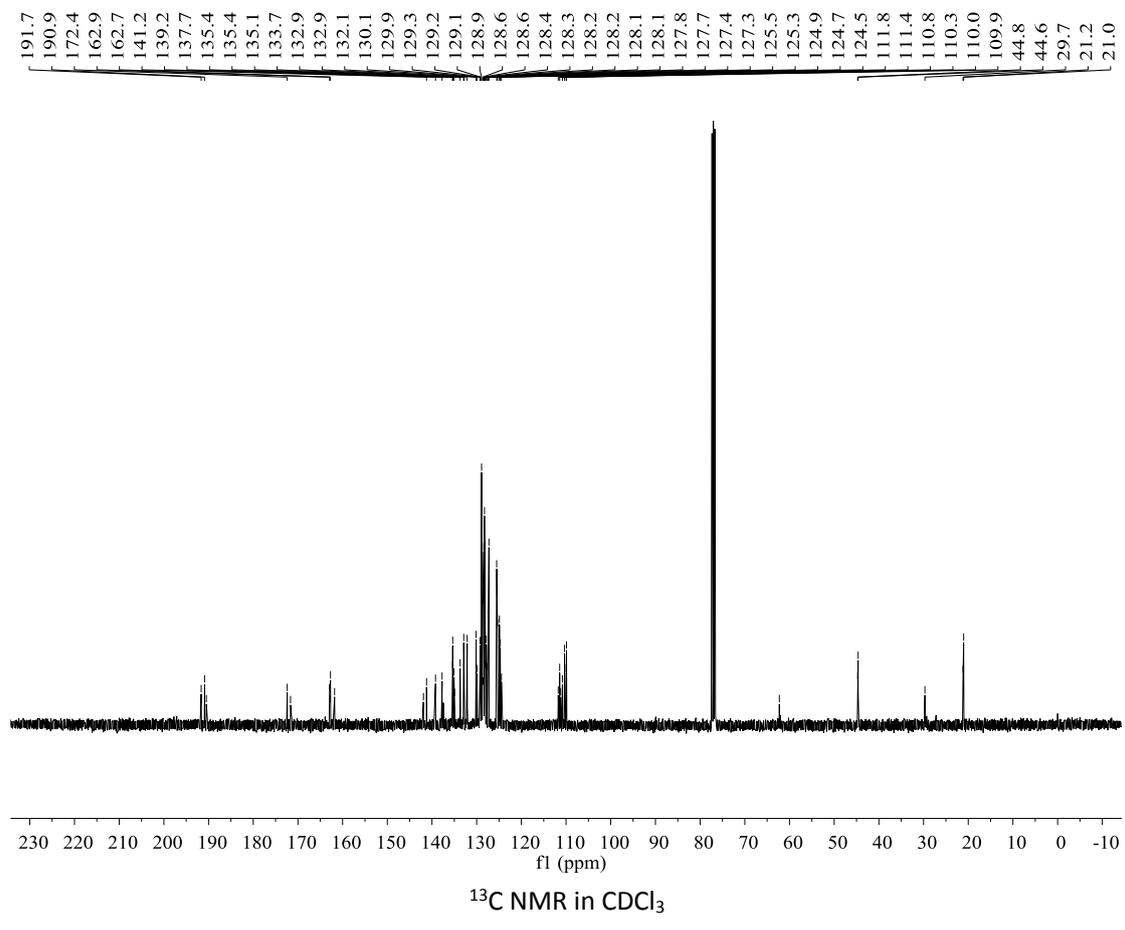
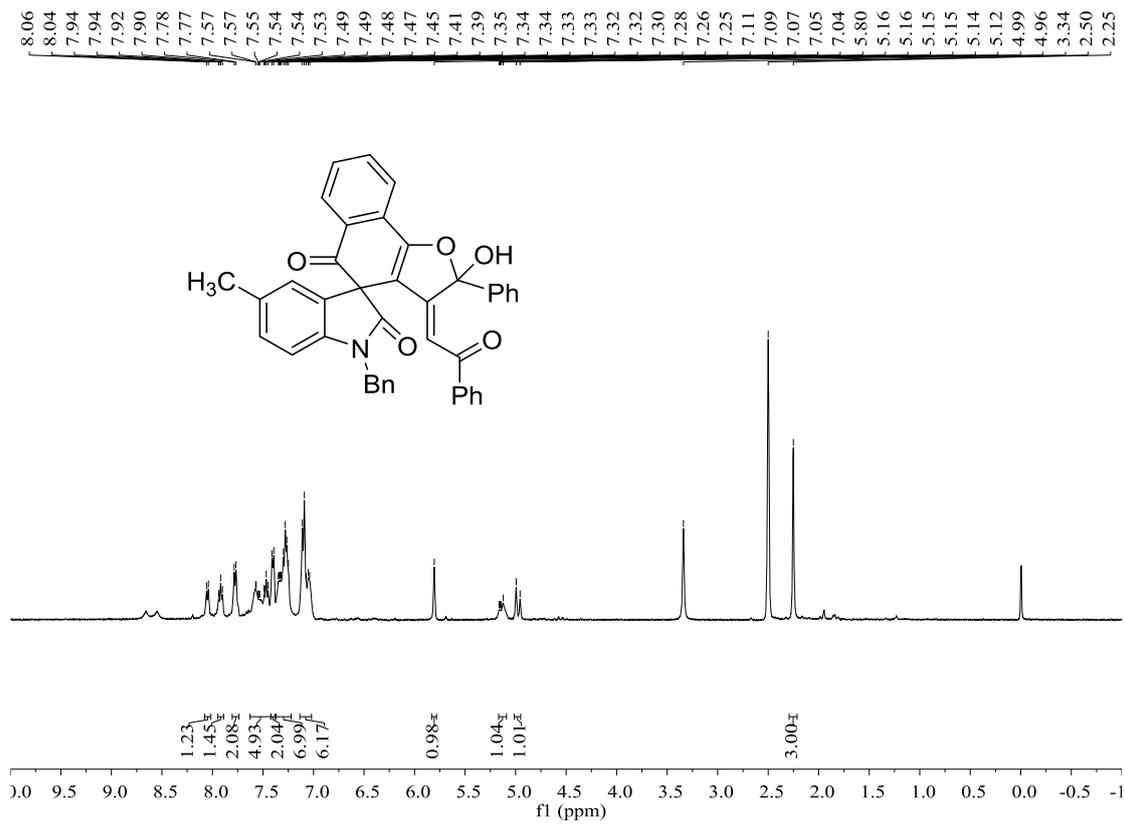


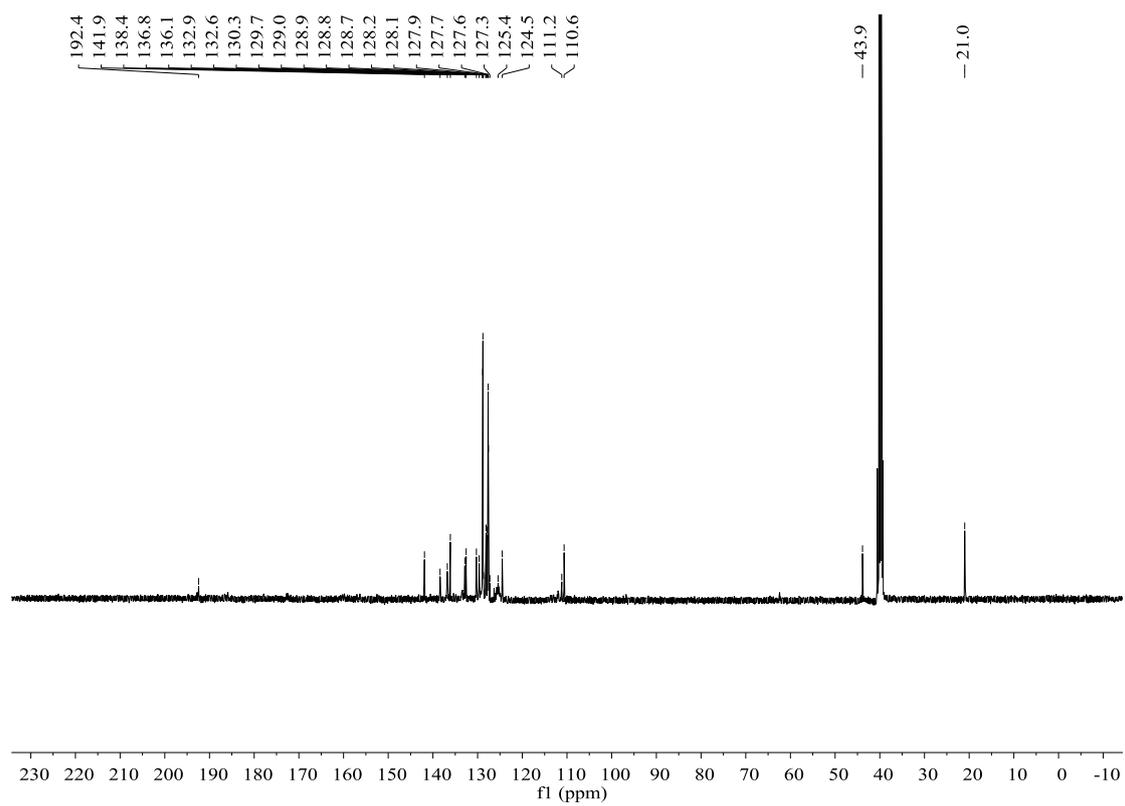
Fig. s2 ORTEP diagram of the compounds **2s**

(Z)-1-benzyl-2'-hydroxy-5-methyl-3'-(2-oxo-2-phenylethylidene)-2'-phenyl-2',3'-dihydro-5'H-spiro[indoline-3,4'-naphtho[1,2-b]furan]-2,5'-dione (**1a**): yellow solid, 75%, m.p. 163-165 °C; ¹H NMR (400 MHz, CDCl₃) δ: major: 8.12-8.08 (m, 1H, ArH), 7.95-7.87 (m, 2H, ArH, OH), 7.79-7.75 (m, 2H, ArH), 7.65-7.61 (m, 2H, ArH), 7.37-7.31 (m, 6H, ArH), 7.28 (s, 1H, ArH), 7.24-7.20 (m, 4H, ArH), 7.10-7.06 (m, 1H, ArH), 7.04-6.99 (m, 1H, ArH), 6.91-6.90 (m, 1H, ArH), 6.78 (d, *J* = 8.0 Hz, 1H, ArH), 6.02 (s, 1H, CH), 5.27 (d, *J* = 15.6 Hz, 1H, CH), 4.96 (d, *J* = 15.2 Hz, 1H, CH), 2.21 (s, 3H, CH₃); minor: 7.57-7.56 (m, 2H, ArH), 7.48-7.42 (m, 6H, ArH), 7.18-7.14 (m, 3H, ArH), 7.03-6.99 (m, 2H, ArH), 5.91 (s, 1H, CH), 5.06 (d, *J* = 15.6 Hz, 1H, CH), 4.89 (d, *J* = 16.0 Hz, 1H, CH), 2.33 (s, 3H, CH₃); ratio of major:minor = 0.6:0.4. ¹³C NMR (150 MHz, CDCl₃) δ: 191.7, 191.6, 190.9, 172.40, 162.9, 162.7, 141.2, 139.2, 137.7, 135.4, 133.7, 132.9, 132.1, 130.1, 129.9, 129.3, 129.2, 129.1, 128.9, 128.6, 128.4, 128.2, 128.1, 127.8, 127.4, 127.3, 125.5, 125.3, 124.9, 124.7, 124.5, 111.8, 111.4, 110.8, 110.3, 110.0, 109.9, 44.8, 44.6, 29.7, 21.2, 21.0; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.05 (d, *J* = 7.6 Hz, 1H, ArH), 7.94-7.90 (m, 1H, ArH), 7.78 (d, *J* = 7.2 Hz, 2H, ArH), 7.57-7.45 (m, 5H, ArH), 7.40 (d, *J* = 7.2 Hz, 2H, ArH), 7.35-7.25 (m, 7H, ArH), 7.11-7.04 (m, 6H, ArH), 5.81 (s, 1H, CH), 5.16-5.12 (m, 1H, CH), 4.97 (d, *J* = 15.6 Hz, 1H, CH), 2.25 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.4, 141.9, 138.4, 136.8, 132.9, 132.6, 130.3, 129.7, 129.0, 128.9, 128.8, 128.7, 128.2, 128.1, 127.9, 127.7, 127.6, 127.3, 125.4, 124.5, 111.2, 110.6, 43.9, 21.0; IR (KBr) ν: 3061, 2921, 1964, 1713, 1623, 1587, 1520, 1444, 1367, 1226, 1126, 1054, 970, 884, 818, 770, 700 cm⁻¹; HRMS (ESI) Calcd. for C₄₁H₂₉NNaO₅ ([M+Na]⁺): 638.1938, Found: 638.1944.



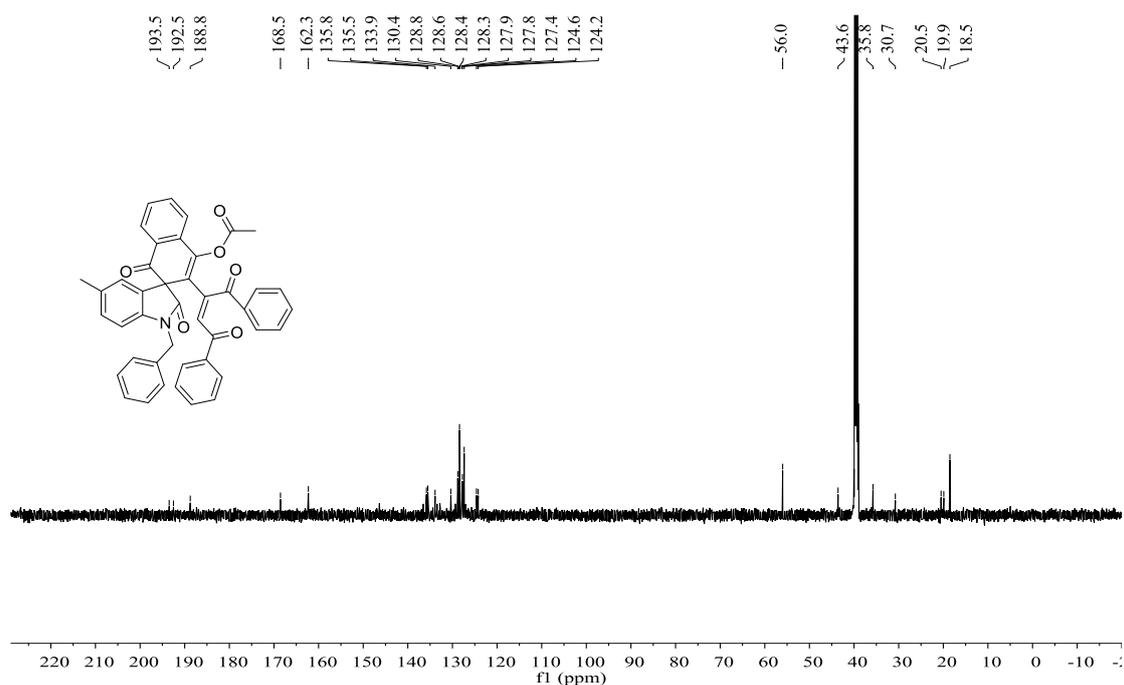
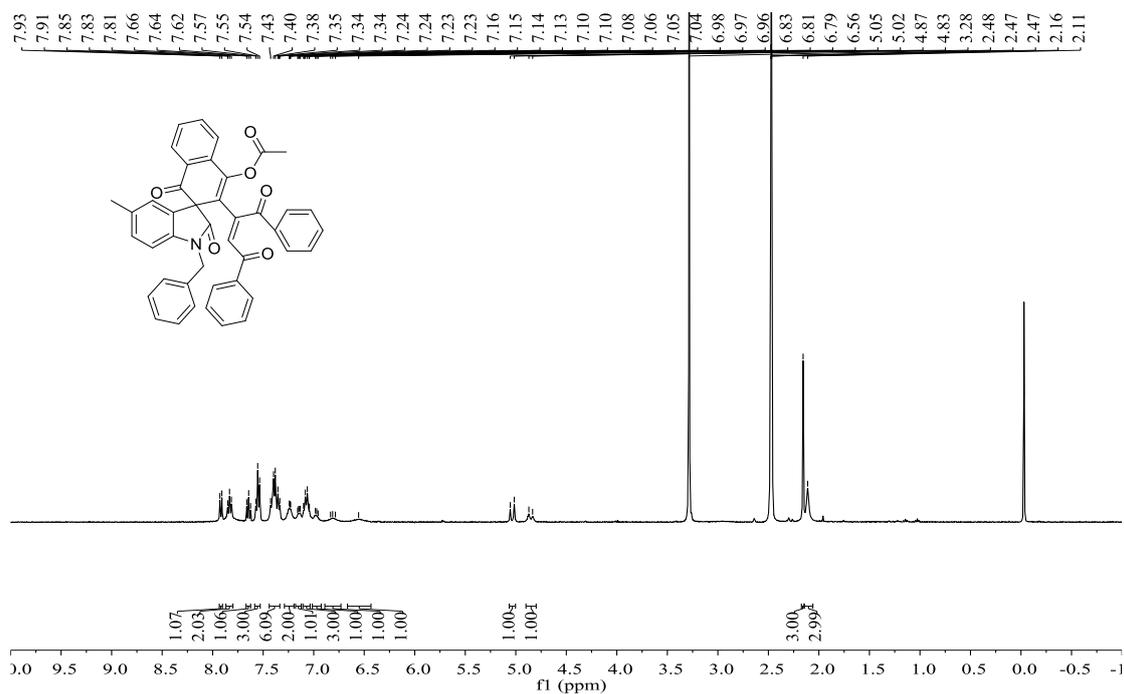
¹H NMR in CDCl₃



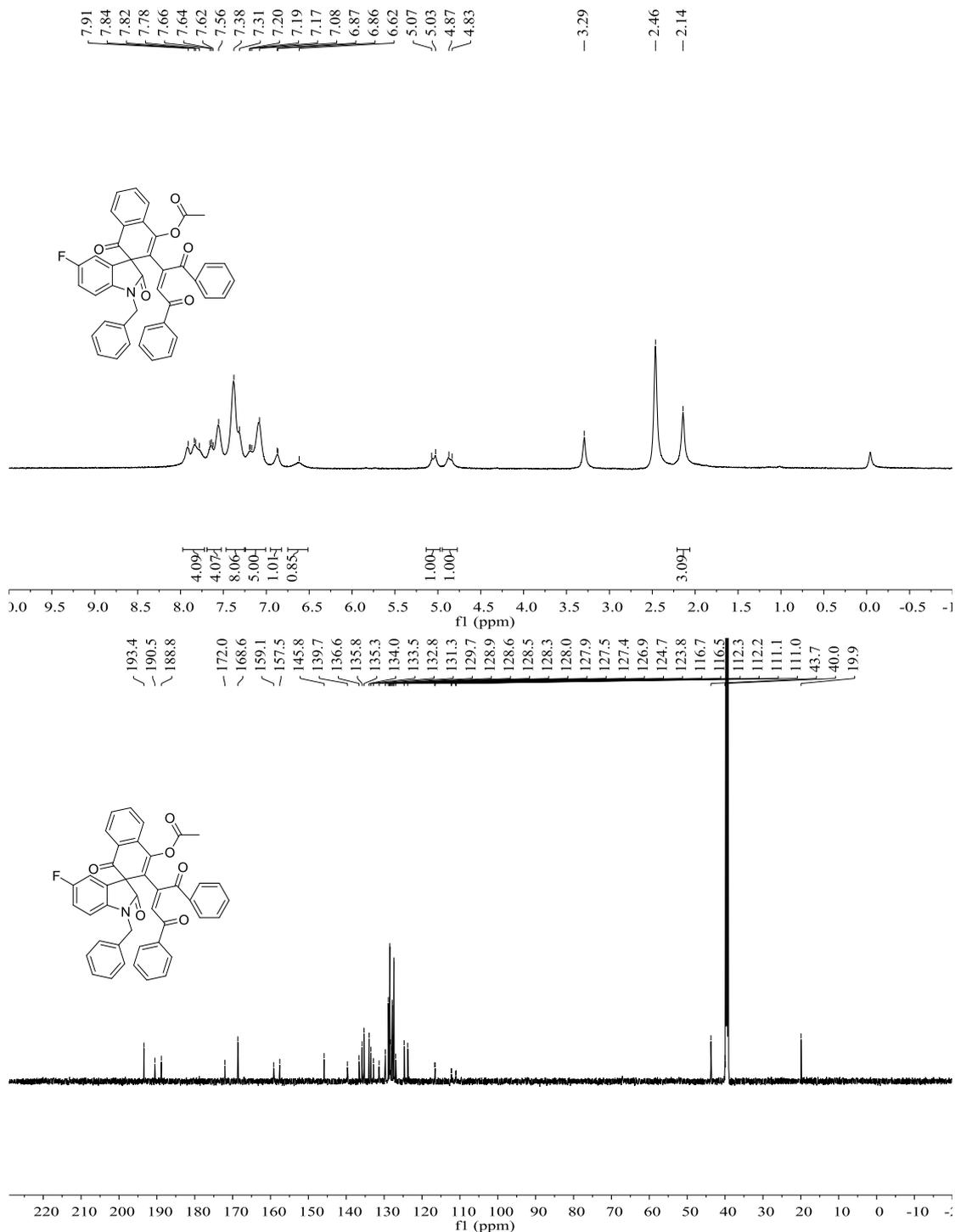


^{13}C NMR in DMSO-d_6

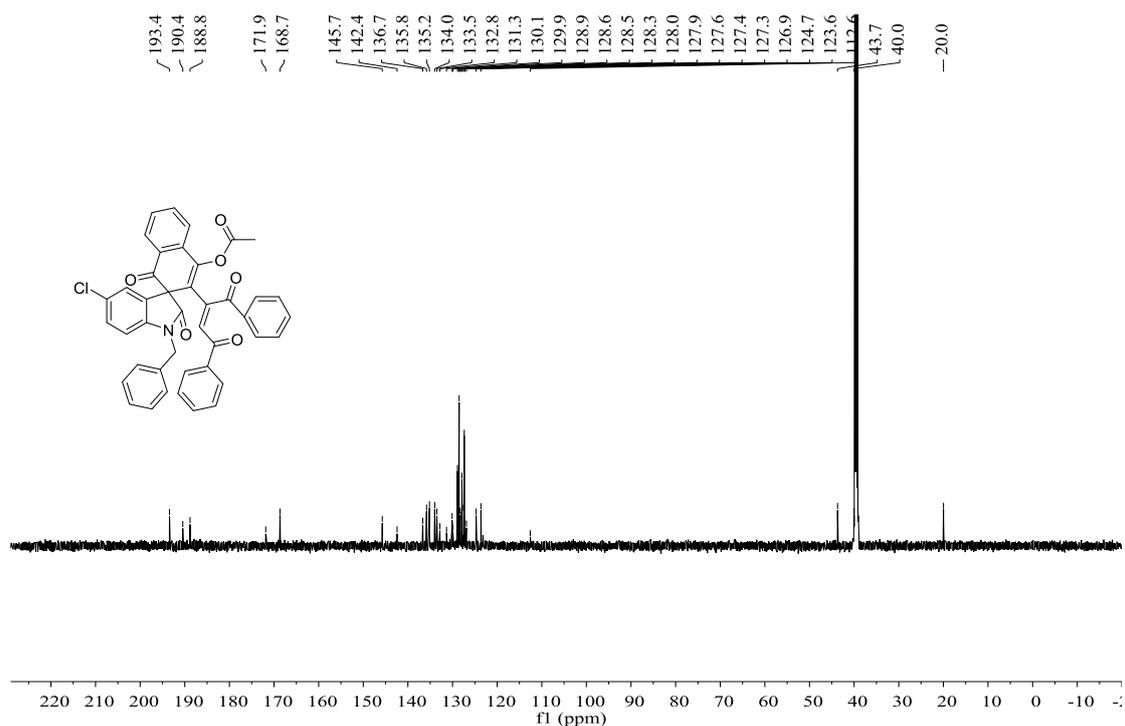
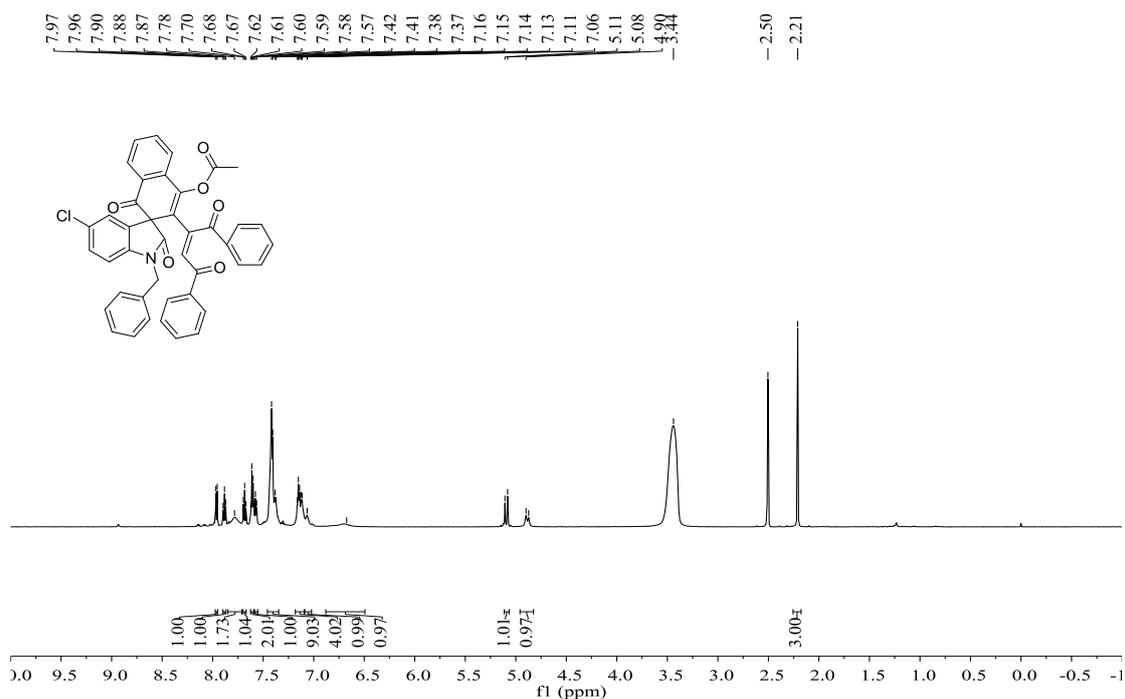
(Z)-1-benzyl-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-5-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2a): yellow solid, 73%, m.p. 159-161 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.94 (d, *J* = 7.6 Hz, 2H, ArH), 7.88-7.84 (m, 2H, ArH), 7.69-7.65 (m, 1H, ArH), 7.60-7.57 (m, 3H, ArH), 7.46-7.37 (m, 6H, ArH), 7.27-7.26 (m, 2H, ArH), 7.18-7.17 (m, 1H, ArH), 7.13-7.01 (m, 4H, ArH), 6.87-6.81 (m, 1H, ArH), 6.59 (br, 1H, CH), 5.06 (d, *J* = 15.6 Hz, 1H, CH), 4.88 (d, *J* = 15.2 Hz, 1H, CH), 2.19 (s, 3H, CH₃), 2.14 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.9, 191.4, 189.1, 174.0, 173.0, 173.0, 168.9, 146.8, 146.1, 141.3, 136.9, 136.2, 135.9, 134.3, 133.8, 133.2, 133.2, 131.7, 130.8, 129.7, 129.2, 129.0, 128.9, 128.8, 128.7, 128.3, 128.2, 127.8, 127.5, 125.0, 124.6, 123.6, 111.5, 44.0, 20.9, 20.3; IR (KBr) ν: 3053, 2912, 2902, 1772, 1730, 1689, 1425, 1376, 886, 756, 757, 668 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₃₁NNaO₆ ([M+Na]⁺): 680.2044, Found: 680.2022.



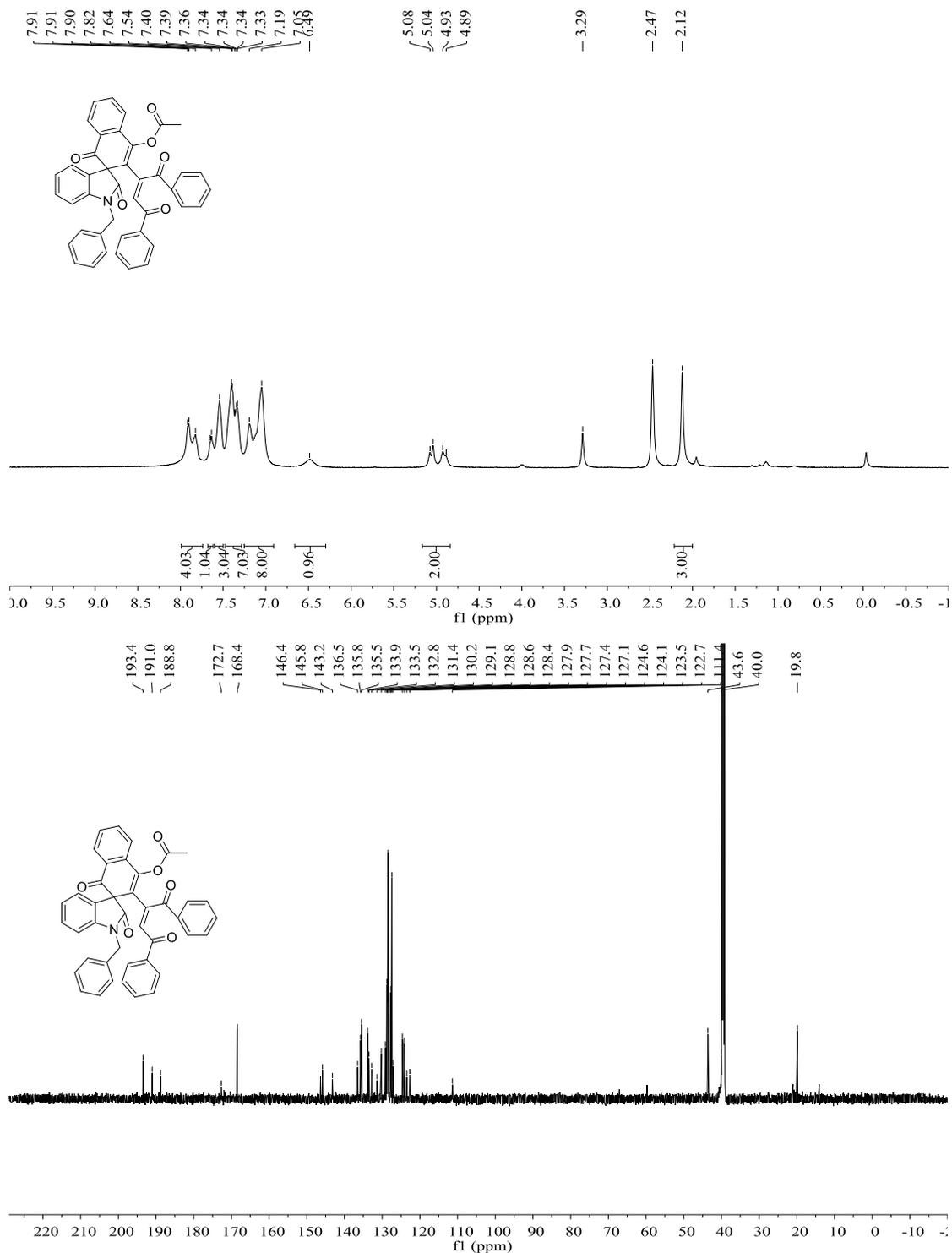
(Z)-1-benzyl-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-5-fluoro-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2b): yellow solid, 75%, m.p. 155-156 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.91-7.78 (m, 4H, ArH), 7.66-7.56 (m, 4H, ArH), 7.38-7.31 (m, 8H, ArH), 7.20-7.08 (m, 5H, ArH), 6.87-6.86 (m, 1H, ArH), 6.62 (br, 1H, CH), 5.05 (d, *J* = 10.4 Hz, 1H, CH), 4.86 (d, *J* = 11.2 Hz, 1H, CH), 2.14 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.3, 190.5, 188.8, 172.0, 168.5, 159.1, 157.5, 145.8, 139.6, 136.6, 135.8, 135.2, 133.9, 133.5, 132.8, 131.3, 129.6, 128.8, 128.5, 128.4, 128.3, 127.9, 127.8, 127.5, 127.4, 126.9, 124.7, 123.7, 116.6, 116.5, 112.2, 112.2, 111.1, 110.9, 43.7, 40.0, 19.9; IR (KBr) ν: 3015, 2942, 1717, 1694, 1662, 1487, 1371, 761, 727, 681 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₈FNNaO₆ ([M+Na]⁺): 684.1793, Found: 684.1811.



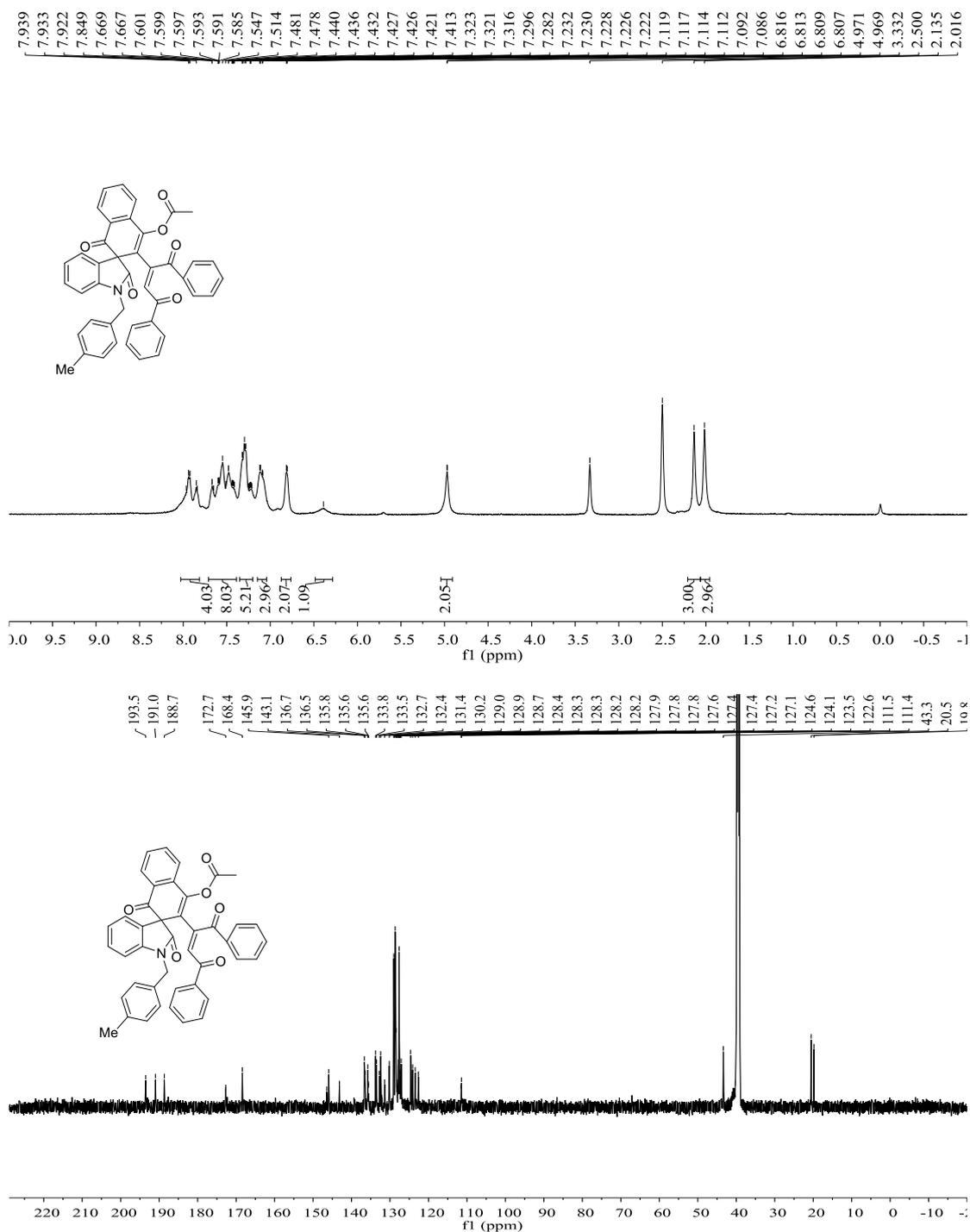
(Z)-1-benzyl-5-chloro-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2c): yellow solid, 66%, m.p. 175-177 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.96 (d, *J* = 7.8 Hz, 1H, ArH), 7.88 (t, *J* = 7.8 Hz, 1H, ArH), 7.79-7.77 (m, 2H, ArH), 7.68 (t, *J* = 7.8 Hz, 1H, ArH), 7.62-7.59 (m, 2H, ArH), 7.57 (d, *J* = 7.2 Hz, 1H, ArH), 7.42-7.37 (m, 9H, ArH), 7.16-7.12 (m, 4H, ArH), 7.06 (br, 1H, ArH), 6.67 (br, 1H, CH), 5.09 (d, *J* = 14.4 Hz, 1H, CH), 4.89 (d, *J* = 14.4 Hz, 1H, CH), 2.21 (s, 3H, CH₃), 2.11 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.4, 190.4, 188.8, 171.8, 168.6, 145.7, 142.4, 136.6, 135.8, 135.1, 133.9, 133.5, 132.8, 131.3, 130.0, 129.9, 128.8, 128.5, 128.5, 128.2, 128.0, 127.8, 127.5, 127.3, 127.2, 126.8, 124.7, 123.6, 112.5, 43.7, 40.0, 19.9; IR (KBr) ν: 3019, 2925, 2902, 1720, 1697, 1656, 1427, 1378, 828, 677 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₈ClNNaO₆ ([M+Na]⁺): 700.1497, Found: 700.1499.



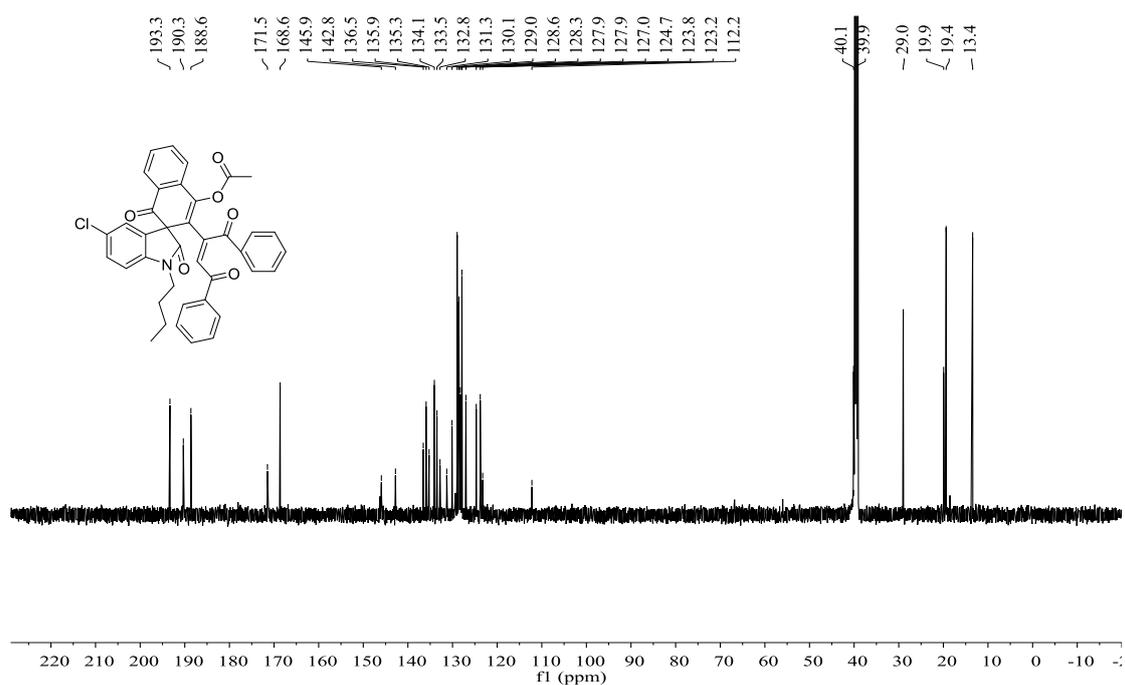
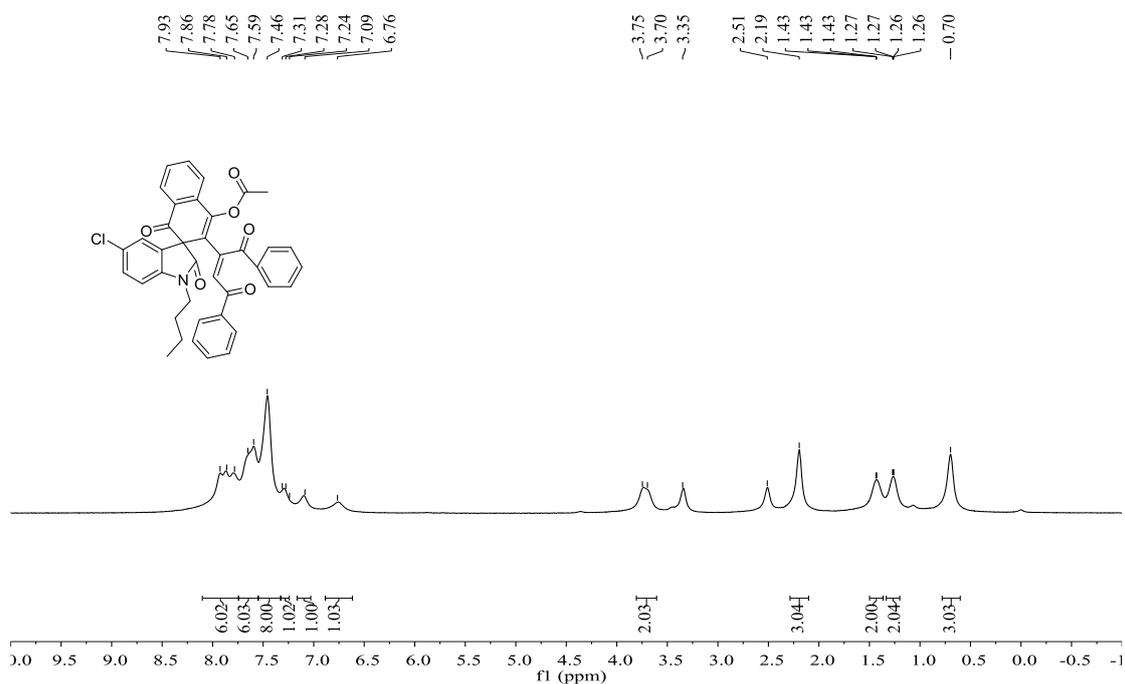
(Z)-1-benzyl-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2d): yellow solid, 71%, m.p. 151-153°C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.92-7.82 (m, 4H, ArH), 7.64 (t, *J* = 5.6 Hz, 1H, ArH), 7.55-7.54 (m, 3H, ArH), 7.40-7.33 (m, 7H, ArH), 7.19-7.05 (m, 8H, ArH), 6.49 (br, 1H, CH), 5.08-4.89 (m, 2H, CH), 2.12 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.4, 191.0, 188.7, 172.7, 168.4, 146.3, 145.8, 143.2, 136.5, 135.8, 135.4, 133.8, 133.5, 132.7, 131.3, 130.2, 129.1, 128.7, 128.6, 128.4, 127.8, 127.7, 127.4, 127.0, 124.6, 124.1, 123.4, 122.6, 111.3, 43.5, 40.0, 19.8; IR (KBr) ν: 3099, 2950, 2902, 1727, 1703, 1638, 1413, 1368, 815, 766, 717, 673 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₉N₂NaO₆ ([M+Na]⁺): 666.1887, Found: 666.1904.



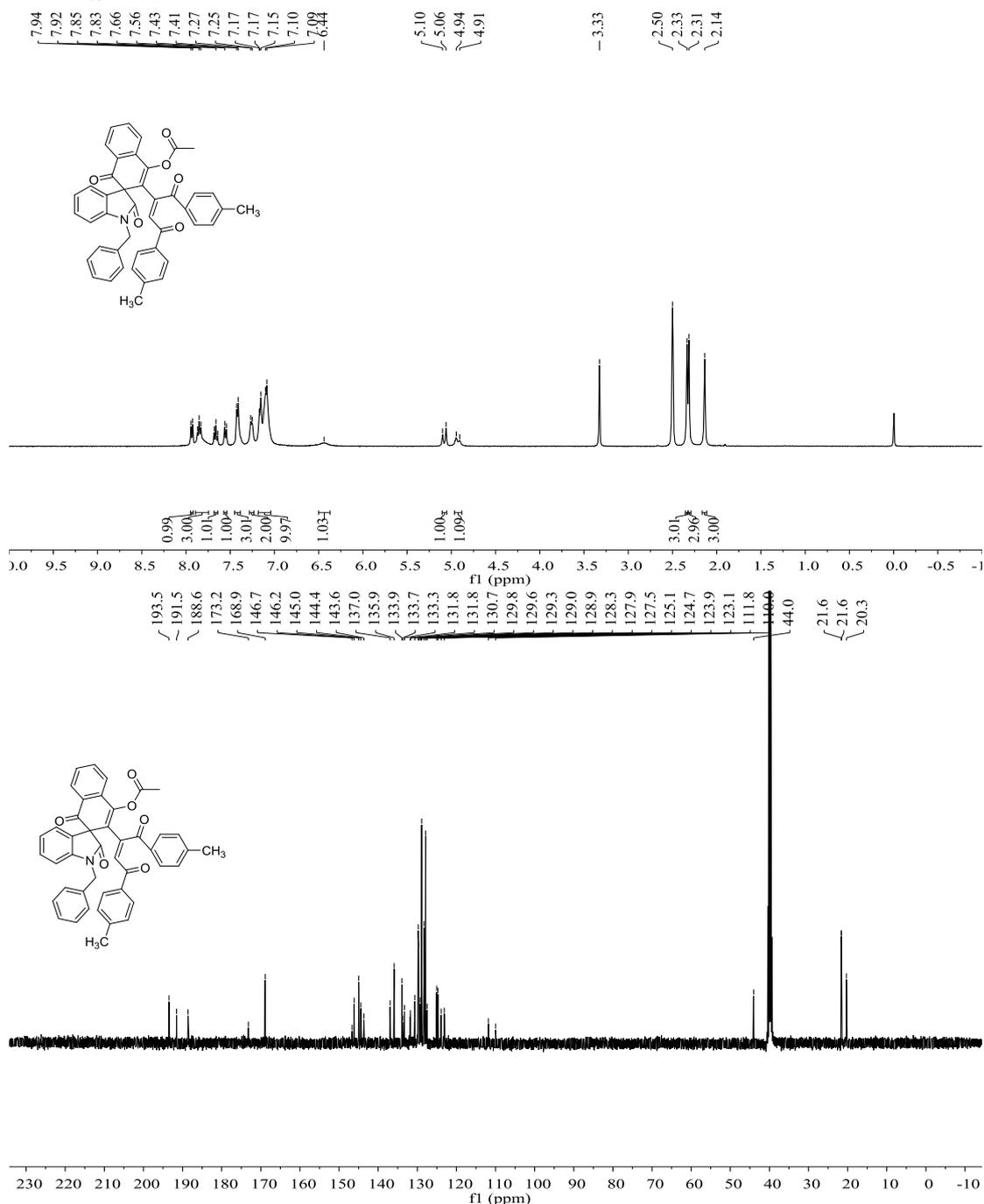
(Z)-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-1-(4-methylbenzyl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2e): yellow solid, 59%, m.p. 160-162 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.96-7.84 (m, 4H, ArH), 7.67-7.41 (m, 8H, ArH), 7.32-7.21 (m, 5H, ArH), 7.12-7.09 (m, 3H, ArH), 6.82-6.80 (m, 2H, ArH), 6.39 (br, 1H, CH), 4.97 (s, 2H, CH), 2.14 (s, 3H, CH₃), 2.02 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.5, 191.0, 188.7, 172.7, 168.4, 145.9, 143.1, 136.7, 136.5, 135.8, 135.6, 133.8, 133.5, 132.7, 132.4, 131.4, 130.2, 129.0, 128.9, 128.7, 128.4, 128.3, 128.2, 128.1, 127.9, 127.8, 127.6, 127.4, 127.3, 127.2, 127.1, 124.6, 124.1, 123.5, 122.6, 111.5, 111.4, 43.3, 20.5, 19.8; IR (KBr) ν: 3041, 3019, 2943, 2908, 2824, 1717, 1704, 1650, 1442, 1373, 841, 835, 789, 761, 714, 676 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₃₁NNaO₆ ([M+Na]⁺): 680.2044, Found: 680.2056.



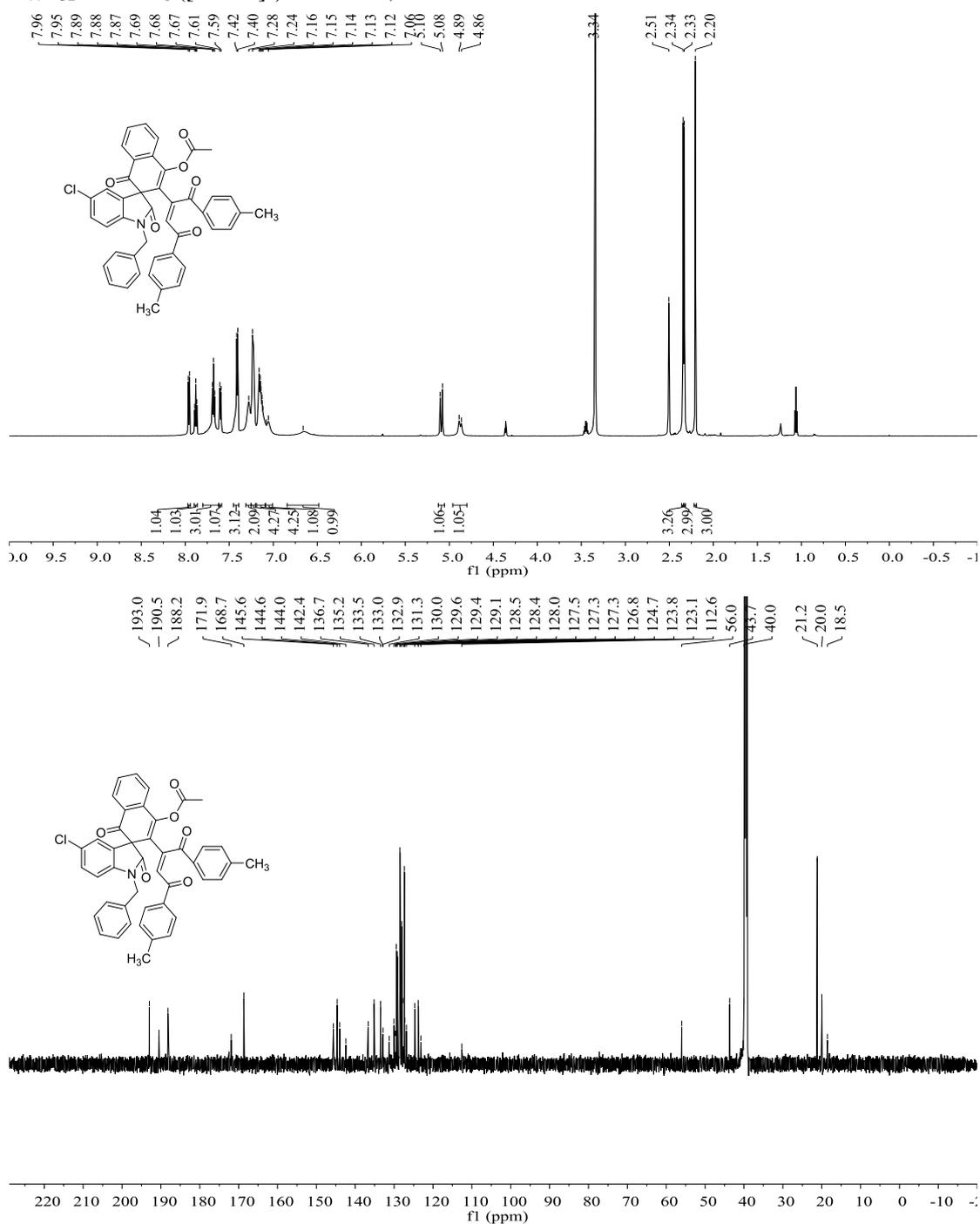
(Z)-1-butyl-5-chloro-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2f): yellow solid, 71%, m.p. 179-181 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.89 (d, *J* = 7.6 Hz, 1H, ArH), 7.85 (t, *J* = 7.6 Hz, 1H, ArH), 7.76-7.78 (m, 2H, ArH), 7.65-7.62 (m, 2H, ArH), 7.57-7.55 (m, 2H, ArH), 7.45-7.42 (m, 7H, ArH), 7.25 (d, *J* = 7.6 Hz, 1H, ArH), 7.05 (br, 1H, ArH), 6.71 (br, 1H, CH), 3.73-3.62 (m, 2H, CH), 2.15 (s, 3H, CH₃), 1.39-1.37 (m, 2H, CH), 1.23-1.22 (m, 2H, CH), 0.66 (t, *J* = 6.4 Hz, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.3, 190.3, 188.6, 171.4, 168.6, 145.9, 142.7, 136.5, 135.9, 135.2, 134.0, 133.4, 132.8, 131.2, 130.1, 128.9, 128.5, 128.2, 127.9, 127.8, 126.9, 124.6, 123.7, 123.2, 112.2, 40.1, 28.9, 19.9, 19.3, 13.4; IR (KBr) ν: 3020, 3009, 2964, 2919, 2912, 1843, 1713, 1699, 1654, 1446, 1367, 1375, 884, 856, 781, 756, 757, 679 cm⁻¹; HRMS (ESI) Calcd. for C₃₉H₃₀ClNNaO₆ ([M+Na]⁺): 666.1654, Found: 666.1662.



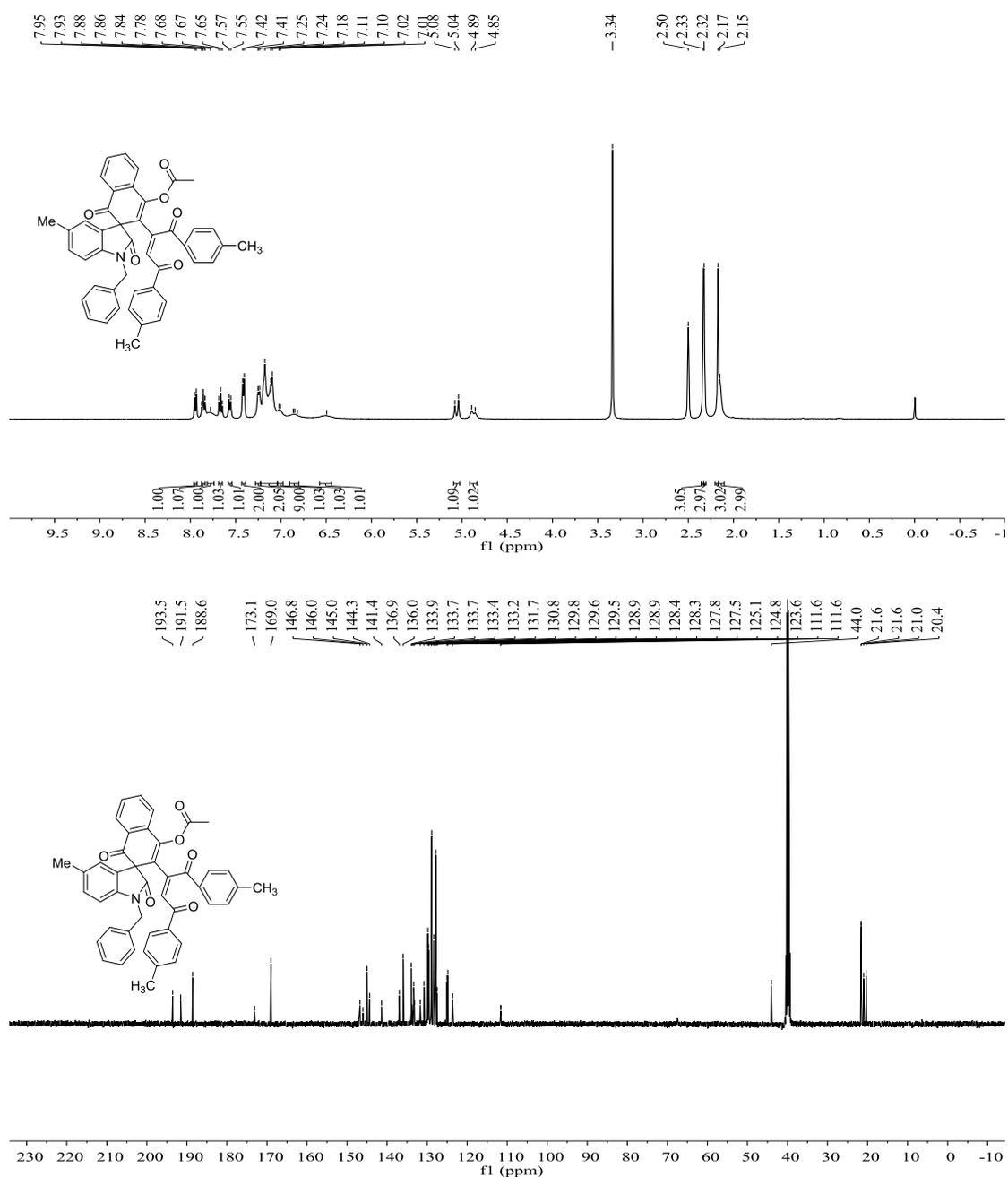
(Z)-1-benzyl-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2g): yellow solid, 57%, m.p. 171-173 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.93 (d, *J* = 7.2 Hz, 1H, ArH), 7.87-7.83 (m, 3H, ArH), 7.68-7.64 (m, 1H, ArH), 7.55 (d, *J* = 7.6 Hz, 1H, ArH), 7.43-7.41 (m, 3H, ArH), 7.27-7.25 (m, 2H, ArH), 7.17-7.07 (m, 10H, ArH), 6.44 (br, 1H, CH), 5.08 (d, *J* = 15.6 Hz, 1H, CH), 4.93 (d, *J* = 15.2 Hz, 1H, CH), 2.34 (s, 3H, CH₃), 2.31 (s, 3H, CH₃), 2.14 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.4, 191.5, 188.5, 173.1, 168.8, 146.6, 146.1, 144.9, 144.4, 143.6, 136.9, 135.9, 133.9, 133.7, 133.2, 131.7, 131.7, 130.6, 129.7, 129.6, 129.3, 128.9, 128.8, 128.2, 127.8, 127.4, 125.0, 124.7, 123.9, 123.0, 111.8, 109.9, 44.0, 21.6, 21.5, 20.3; IR (KBr) ν: 3056, 2924, 2876, 1671, 1579, 1486, 1331, 1282, 1240, 1167, 1051, 978, 877, 792, 756, 730 cm⁻¹; HRMS (ESI) Calcd. for C₄₄H₃₃NNaO₆ ([M+Na]⁺): 694.2200, Found: 694.2222.



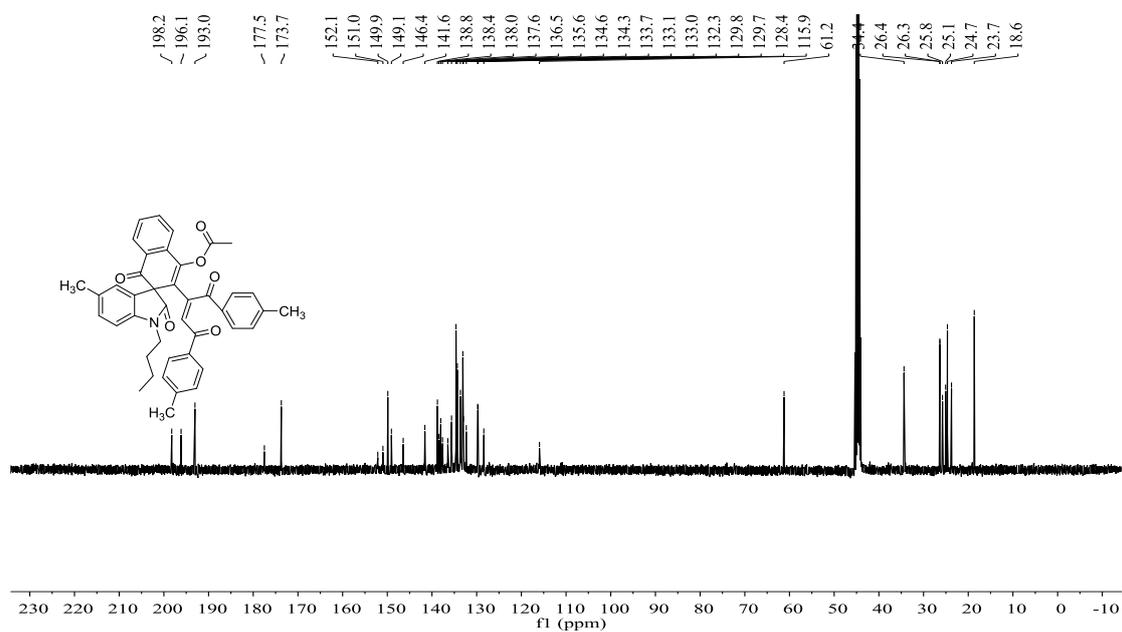
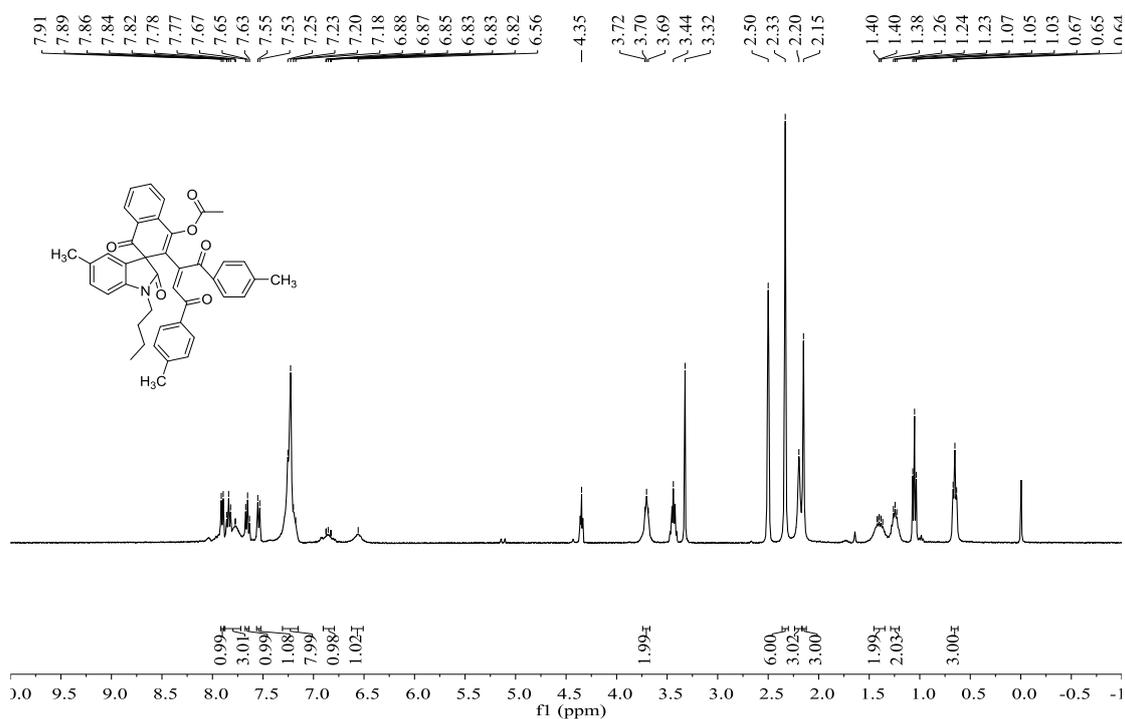
(Z)-1-benzyl-5-chloro-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2h): yellow solid, 59%, m.p. 175-176 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.96 (d, *J* = 7.8 Hz, 1H, ArH), 7.88 (t, *J* = 7.2 Hz, 1H, ArH), 7.70-7.67 (m, 3H, ArH), 7.60 (d, *J* = 7.8 Hz, 1H, ArH), 7.42-7.40 (m, 3H, ArH), 7.28-7.27 (m, 2H, ArH), 7.24-7.22 (m, 4H, ArH), 7.16-7.12 (m, 4H, ArH), 7.08-7.05 (m, 1H, ArH), 6.66 (s, 1H, CH), 5.09 (d, *J* = 15.6 Hz, 1H, CH), 4.88 (d, *J* = 15.6 Hz, 1H, CH), 2.34 (s, 3H, CH₃), 2.33 (s, 3H, CH₃), 2.20 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.9, 190.4, 188.1, 171.9, 168.6, 145.6, 144.6, 144.0, 142.4, 136.6, 135.1, 133.4, 132.9, 132.8, 131.2, 130.0, 129.6, 129.4, 129.1, 128.4, 128.3, 128.0, 127.5, 127.3, 127.2, 126.8, 124.6, 123.7, 123.1, 112.6, 55.9, 43.6, 21.1, 19.9; IR (KBr) ν: 3023, 2942, 2867, 1711, 1701, 1665, 1411, 1369, 821, 769, 724, 689 cm⁻¹; HRMS (ESI) Calcd. for C₄₄H₃₂ClNNaO₆ ([M+Na]⁺): 728.1810, Found: 728.1811.



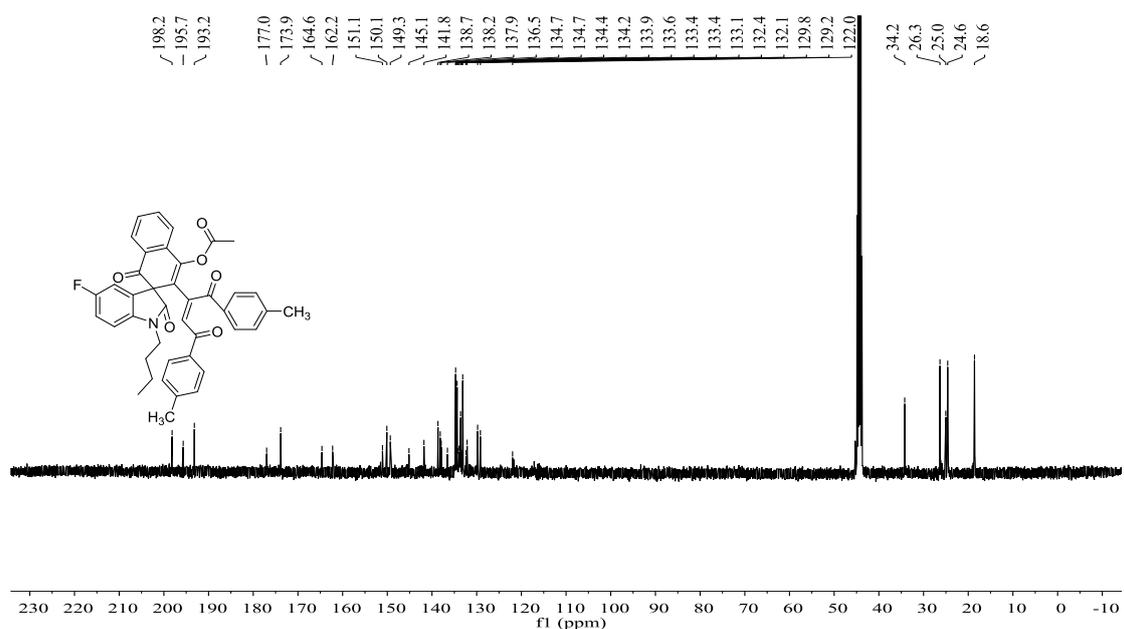
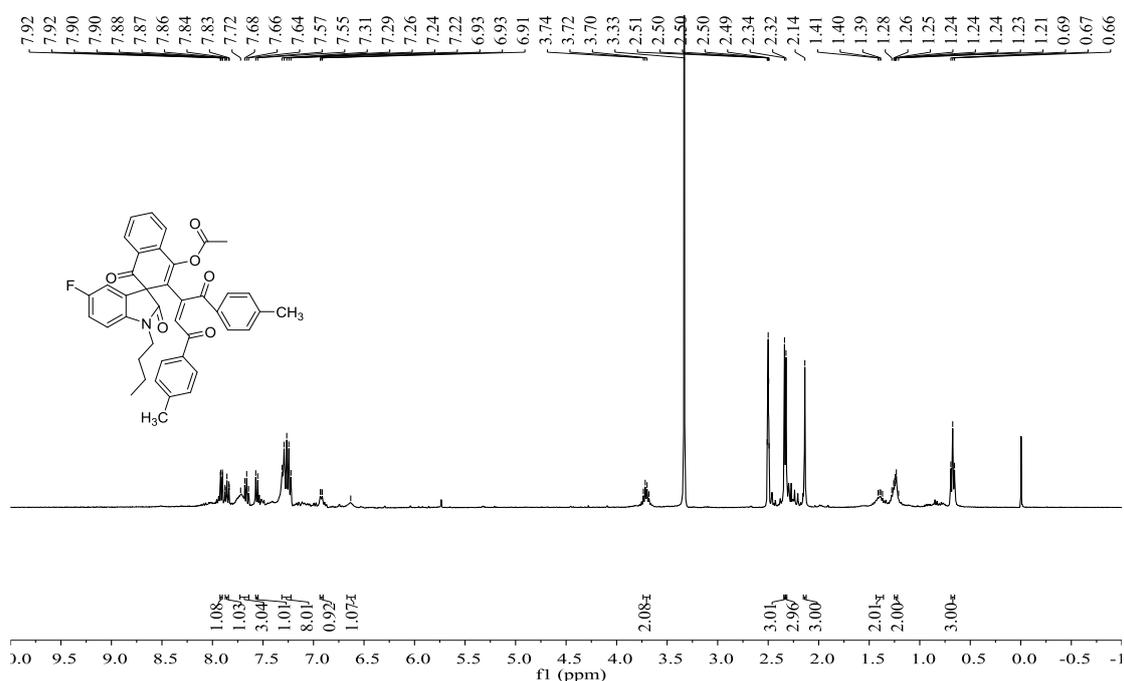
(Z)-1-benzyl-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-5-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2i): yellow solid, 64%, m.p. 170-171 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.94 (d, *J* = 7.6 Hz, 1H, ArH), 7.86 (t, *J* = 7.6 Hz, 1H, ArH), 7.78-7.77 (m, 1H, ArH), 7.67 (t, *J* = 7.6 Hz, 1H, ArH), 7.56 (d, *J* = 8.0 Hz, 1H, ArH), 7.41 (d, *J* = 7.2 Hz, 2H, ArH), 7.24 (d, *J* = 6.8 Hz, 2H, ArH), 7.18-7.10 (m, 9H, ArH), 7.02-7.00 (m, 1H, ArH), 6.86-6.82 (m, 1H, ArH), 6.49 (br, 1H, CH), 5.06 (d, *J* = 11.6 Hz, 1H, CH), 4.87 (d, *J* = 11.6 Hz, 1H, CH), 2.33 (s, 3H, CH₃), 2.32 (s, 3H, CH₃), 2.17 (s, 3H, CH₃), 2.15 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 193.5, 191.5, 188.5, 173.0, 168.9, 146.7, 146.0, 144.9, 144.3, 141.3, 136.9, 135.9, 133.9, 133.6, 133.3, 133.1, 131.7, 130.7, 129.7, 129.5, 129.4, 128.9, 128.8, 128.3, 128.3, 127.8, 127.4, 125.0, 124.8, 123.6, 111.6, 111.5, 44.0, 21.6, 21.5, 20.9, 20.3; IR (KBr) ν: 3011, 2985, 2920, 1731, 1708, 1692, 1413, 1376, 763, 725 cm⁻¹; HRMS (ESI) Calcd. for C₄₅H₃₅NNaO₆ ([M+Na]⁺): 708.2357, Found: 708.2379.



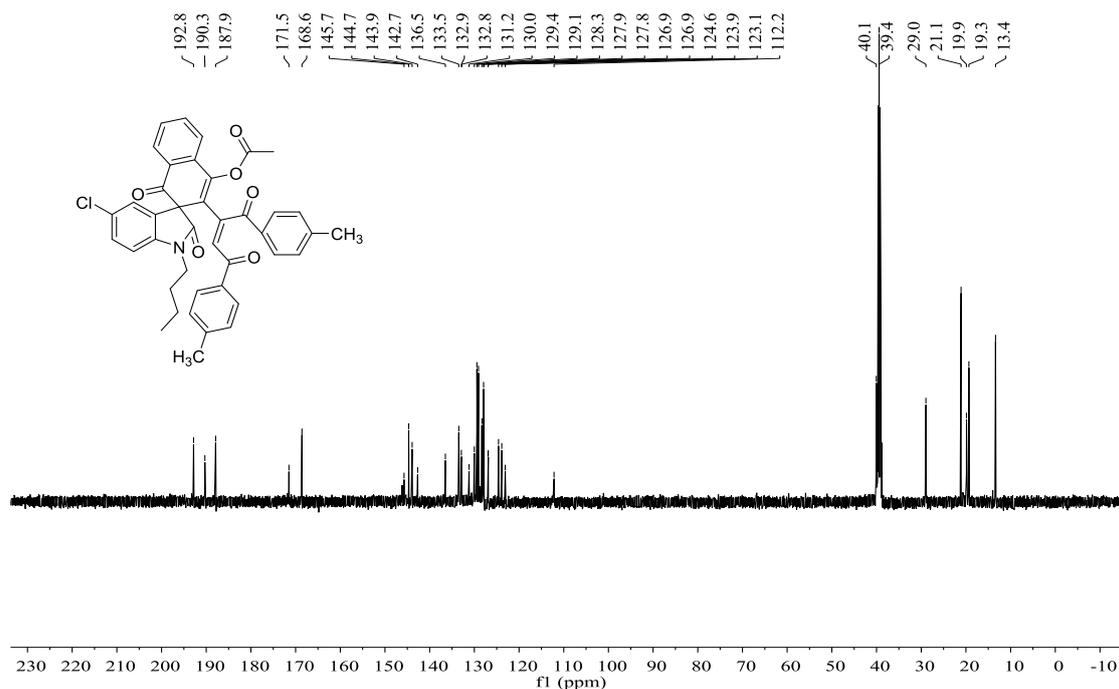
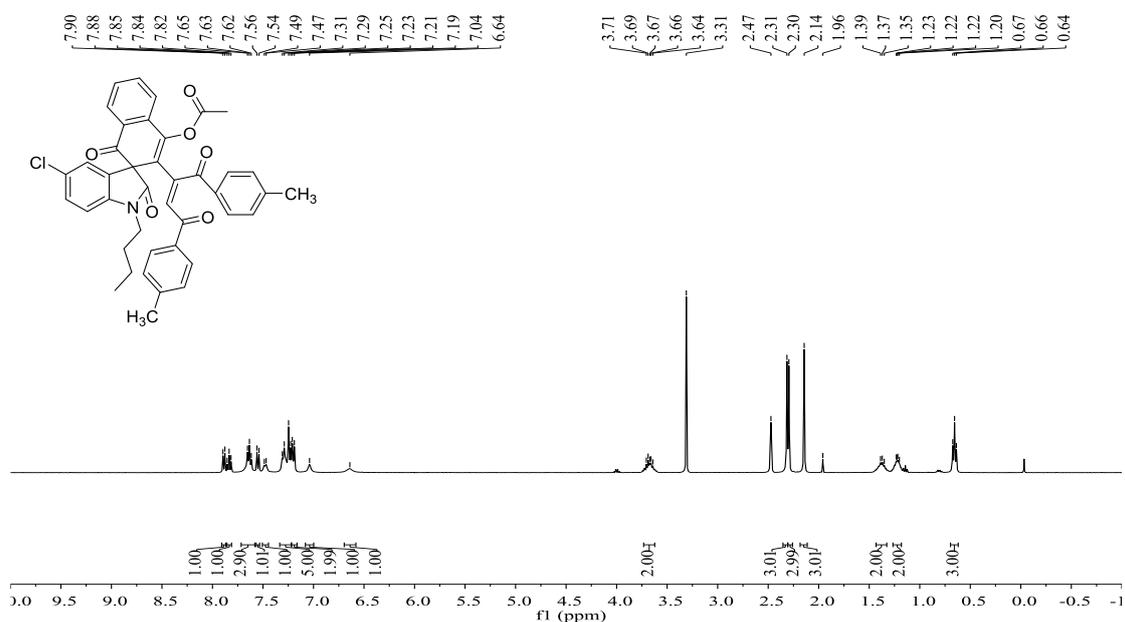
(Z)-1-butyl-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-5-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2j): yellow solid, 63%, m.p. 165-167 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.90 (d, *J* = 7.2 Hz, 1H, ArH), 7.86-7.77 (m, 3H, ArH), 7.67-7.63 (m, 1H, ArH), 7.54 (d, *J* = 7.6 Hz, 1H, ArH), 7.25-7.18 (m, 8H, ArH), 6.88-6.82 (m, 1H, ArH), 6.56 (br, 1H, CH), 3.72-3.69 (m, 2H, ArH), 2.33 (s, 3H, CH₃), 2.20 (s, 3H, CH₃), 2.15 (s, 3H, CH₃), 1.41-1.37 (m, 2H, CH), 1.26-1.23 (m, 2H, CH), 0.65 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 198.2, 196.1, 193.0, 177.5, 173.7, 152.1, 150.9, 149.8, 149.0, 146.4, 141.5, 138.7, 138.4, 138.0, 137.6, 136.4, 135.6, 134.6, 134.3, 133.6, 133.0, 132.9, 132.3, 129.7, 129.6, 128.4, 115.9, 61.2, 34.3, 26.3, 26.3, 25.7, 25.0, 24.6, 23.7, 18.6; IR (KBr) ν: 3063, 2959, 2960, 1731, 1708, 1689, 1638, 1517, 1432, 1390, 1322, 1276, 1128, 1034, 927, 860, 825, 762 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₃₇NNaO₆ ([M+Na]⁺): 674.2513, Found: 674.2532.



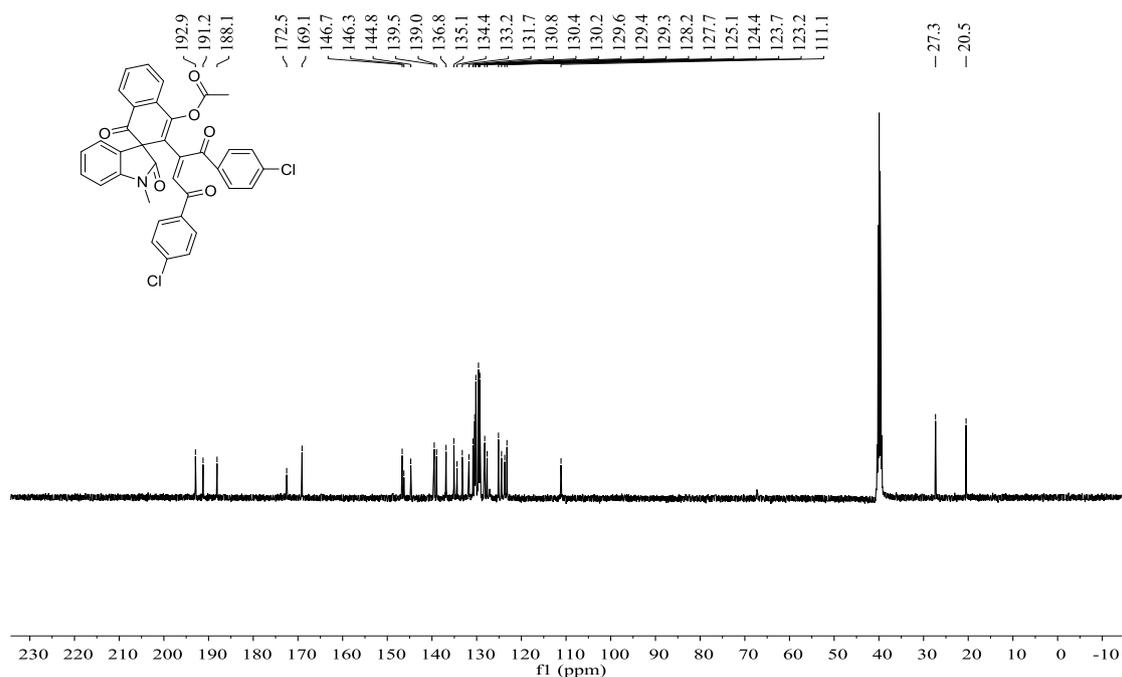
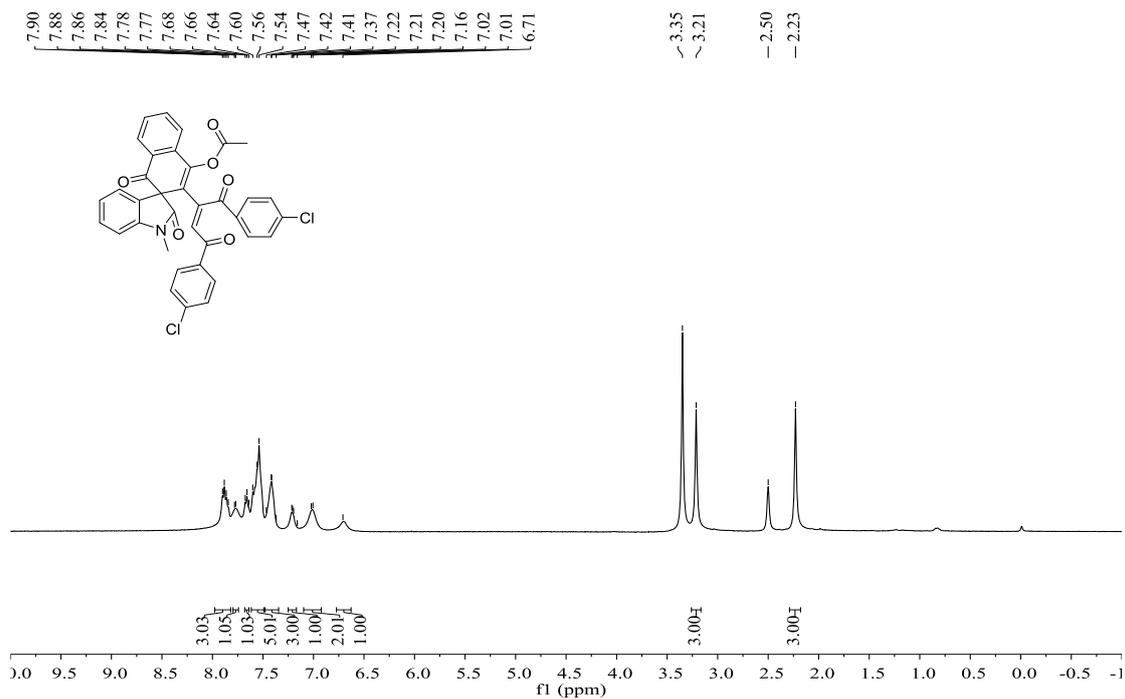
(Z)-1-butyl-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-5-fluoro-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2k): yellow solid, 66%, m.p. 179-181 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.92-7.90 (m, 1H, ArH), 7.88-7.83 (m, 1H, ArH), 7.72-7.64 (m, 3H, ArH), 7.57-7.55 (m, 1H, ArH), 7.31-7.22 (m, 8H, ArH), 6.93-6.91 (m, 1H, ArH), 6.63 (br, 1H, CH), 3.74-3.68 (m, 2H, ArH), 2.34 (s, 3H, CH₃), 2.32 (s, 3H, CH₃), 2.14 (s, 3H, CH₃), 1.41-1.37 (m, 2H, CH), 1.25-1.23 (m, 2H, CH), 0.67 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 198.1, 195.6, 193.1, 177.0, 173.8, 164.6, 162.2, 151.0, 150.1, 149.3, 145.1, 141.7, 138.6, 138.1, 137.9, 136.5, 134.7, 134.6, 134.3, 134.2, 133.9, 133.5, 133.3, 133.3, 133.1, 132.4, 132.1, 129.7, 129.1, 121.9, 121.7, 117.1, 34.2, 26.3, 25.0, 24.5, 18.5; IR (KBr) ν: 3020, 2963, 1777, 1724, 1679, 1602, 1570, 1492, 1350, 1270, 1180, 962, 878, 812, 719 cm⁻¹; HRMS (ESI) Calcd. for C₄₁H₃₄FNNO₆ ([M+Na]⁺): 678.2262, Found: 678.2269.



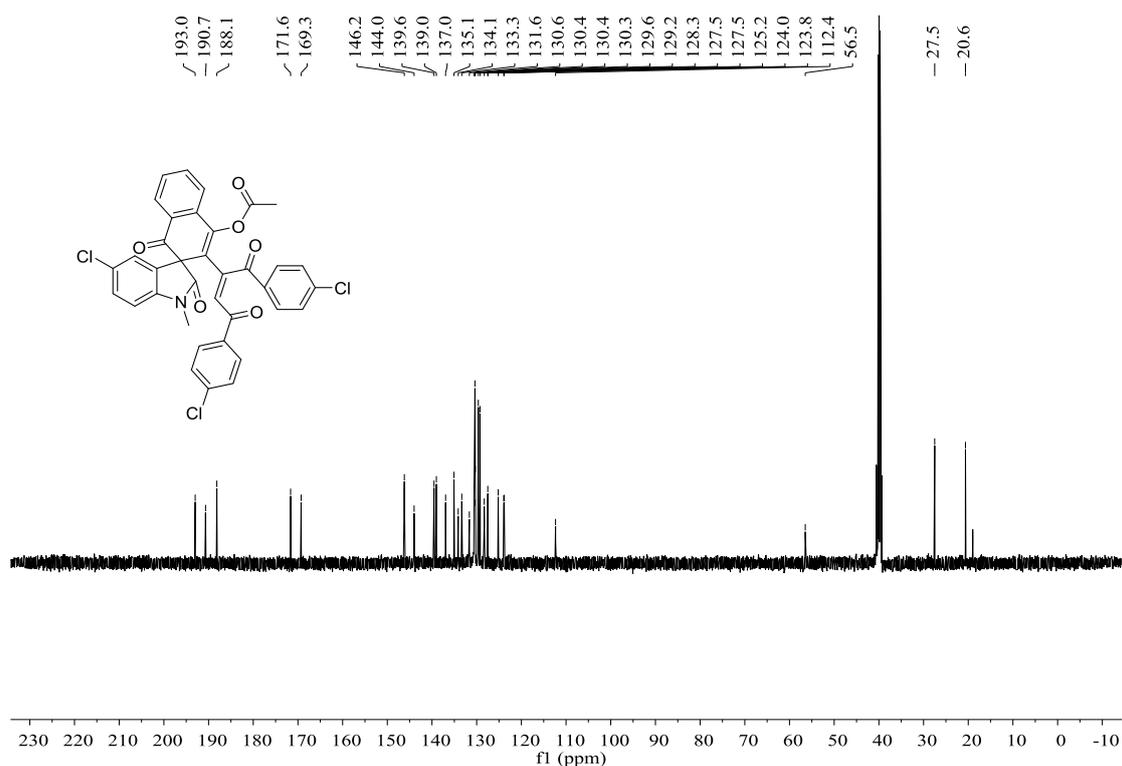
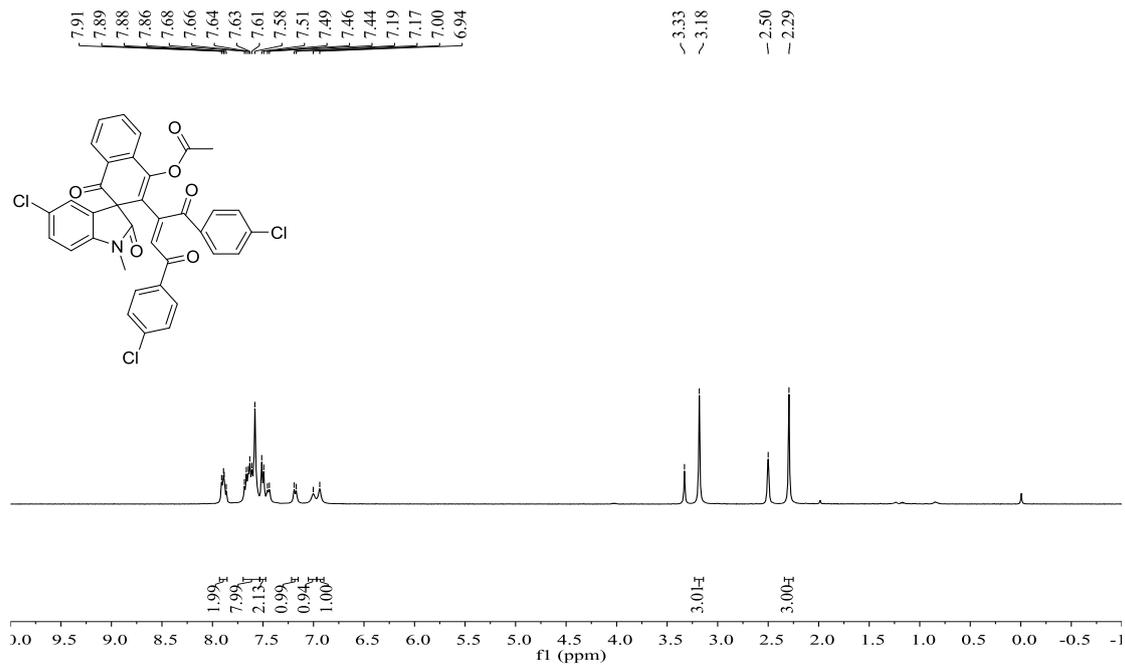
(Z)-1-butyl-5-chloro-3'-(1,4-dioxo-1,4-di-p-tolylbut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2): yellow solid, 67%, m.p. 163-165 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.89 (d, *J* = 7.6 Hz, 1H, ArH), 7.84 (t, *J* = 8.0 Hz, 1H, ArH), 7.66-7.61 (m, 3H, ArH), 7.55 (d, *J* = 7.6 Hz, 1H, ArH), 7.48 (d, *J* = 7.6 Hz, 1H, ArH), 7.31-7.23 (m, 5H, ArH), 7.20 (d, *J* = 7.6 Hz, 2H, ArH), 7.04 (br, 1H, ArH), 6.64 (br, 1H, CH), 3.71-3.64 (m, 2H, CH), 2.31 (s, 3H, CH₃), 2.30 (s, 3H, CH₃), 2.14 (s, 3H, CH₃), 1.39-1.35 (m, 2H, CH), 1.23-1.20 (m, 2H, CH), 0.66 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.8, 190.3, 187.8, 171.4, 168.5, 145.7, 144.6, 143.9, 142.6, 136.4, 133.4, 132.9, 132.7, 131.1, 130.0, 129.4, 129.0, 128.3, 127.9, 127.8, 126.9, 126.8, 124.5, 123.8, 123.0, 112.1, 40.0, 28.9, 21.0, 19.8, 19.3, 13.3; IR (KBr) ν: 3011, 2987, 2920, 2831, 1724, 1705, 1691, 1589, 1416, 1375, 761, 720 cm⁻¹; HRMS (ESI) Calcd. for C₄₁H₃₄ClNNaO₆ ([M+Na]⁺): 694.1967, Found: 694.1979.



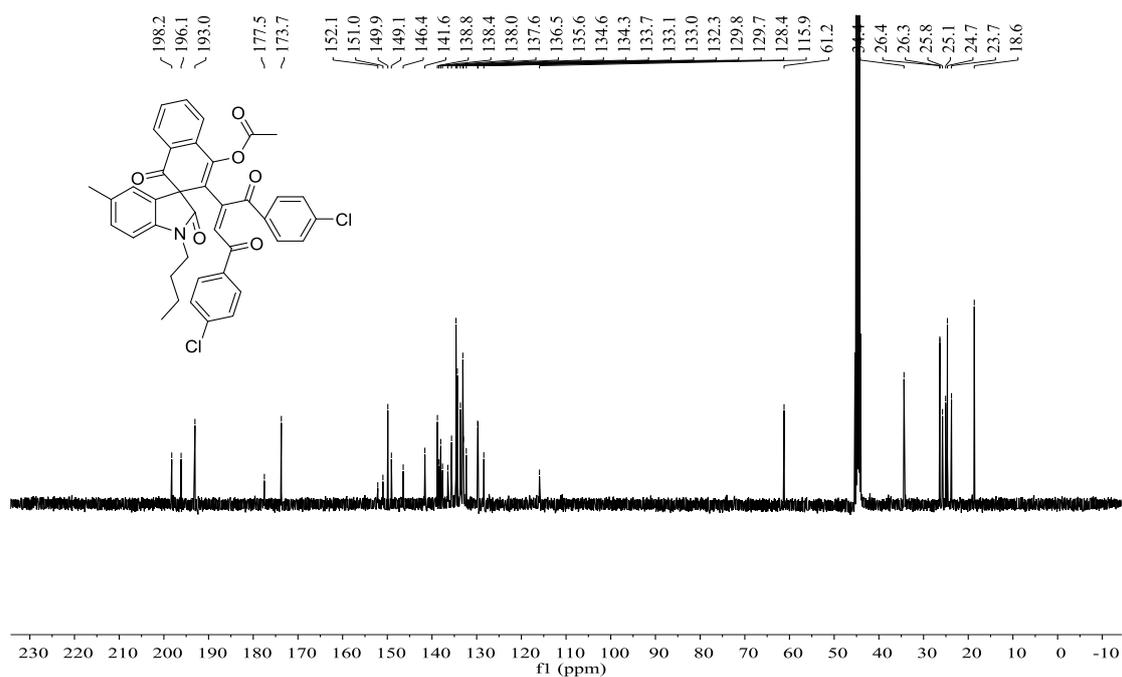
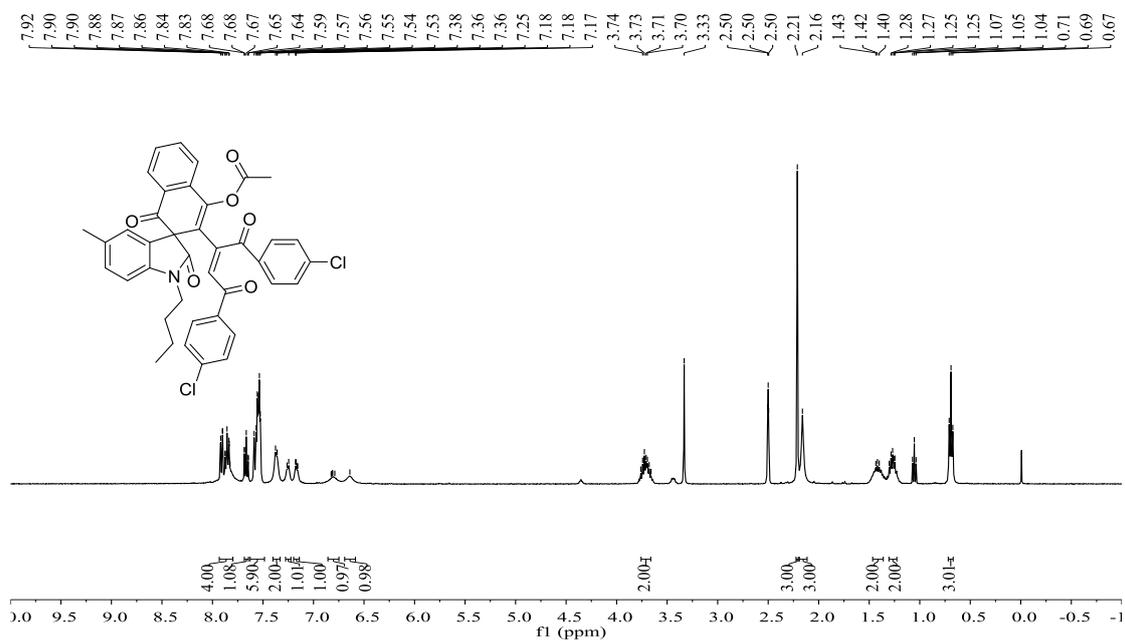
(Z)-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-1-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2m): yellow solid, 72%, m.p. 170-171 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.90-7.84 (m, 3H, ArH), 7.78-7.76 (m, 1H, ArH), 7.66 (t, *J* = 7.6 Hz, 1H, ArH), 7.60-7.54 (m, 5H, ArH), 7.47-7.37 (m, 3H, ArH), 7.22-7.20 (m, 1H, ArH), 7.02-7.00 (m, 2H, ArH), 6.71 (br, 1H, CH), 3.21 (s, 3H, CH₃), 2.30 (s, 3H, CH₃), 2.23 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.9, 191.2, 188.0, 172.5, 169.0, 146.6, 146.2, 144.7, 139.4, 138.9, 136.8, 135.1, 134.3, 133.2, 131.7, 130.8, 130.4, 130.1, 129.5, 129.4, 129.2, 128.2, 127.6, 125.1, 124.4, 123.7, 123.2, 111.1, 27.3, 20.4; IR (KBr) ν: 3009, 2968, 2825, 1731, 1704, 1688, 1593, 1416, 1378, 1375, 777, 715 cm⁻¹; HRMS (ESI) Calcd. for C₃₆H₂₃Cl₂NNaO₆ ([M+Na]⁺): 658.0795, Found: 658.0798.



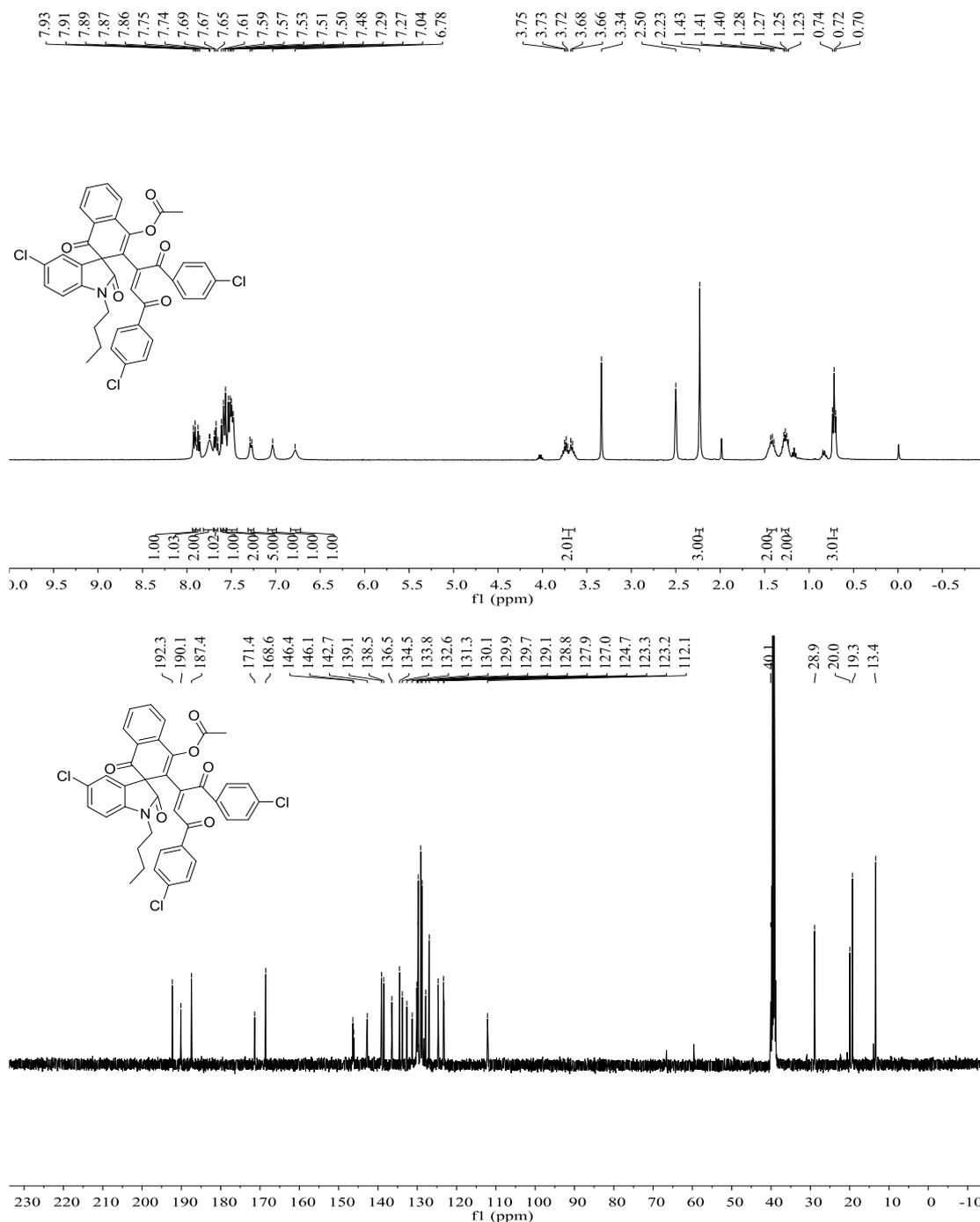
(Z)-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-5-chloro-1-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2n): yellow solid, 74%, m.p. 195-197 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.91-7.86 (m, 2H, ArH), 7.68-7.58 (m, 8H, ArH), 7.50 (d, *J* = 8.0 Hz, 1H, ArH), 7.18 (d, *J* = 8.0 Hz, 1H, ArH), 7.00 (br, 1H, ArH), 6.94 (br, 1H, CH), 3.18 (s, 3H, CH₃), 2.29 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.9, 190.6, 188.1, 171.6, 169.2, 146.1, 144.0, 139.5, 139.0, 136.9, 135.0, 134.1, 133.3, 131.6, 130.5, 130.4, 130.3, 130.3, 129.6, 129.2, 128.3, 127.5, 127.4, 125.1, 123.9, 123.8, 112.3, 56.4, 27.5, 20.6; IR (KBr) ν: 3039, 2896, 2805, 1778, 1717, 1677, 1589, 1486, 1425, 1344, 1265, 1174, 1063, 964, 813, 765 cm⁻¹; HRMS (ESI) Calcd. for C₃₆H₂₂Cl₃NNaO₆ ([M+Na]⁺): 692.0405, Found: 692.0430.



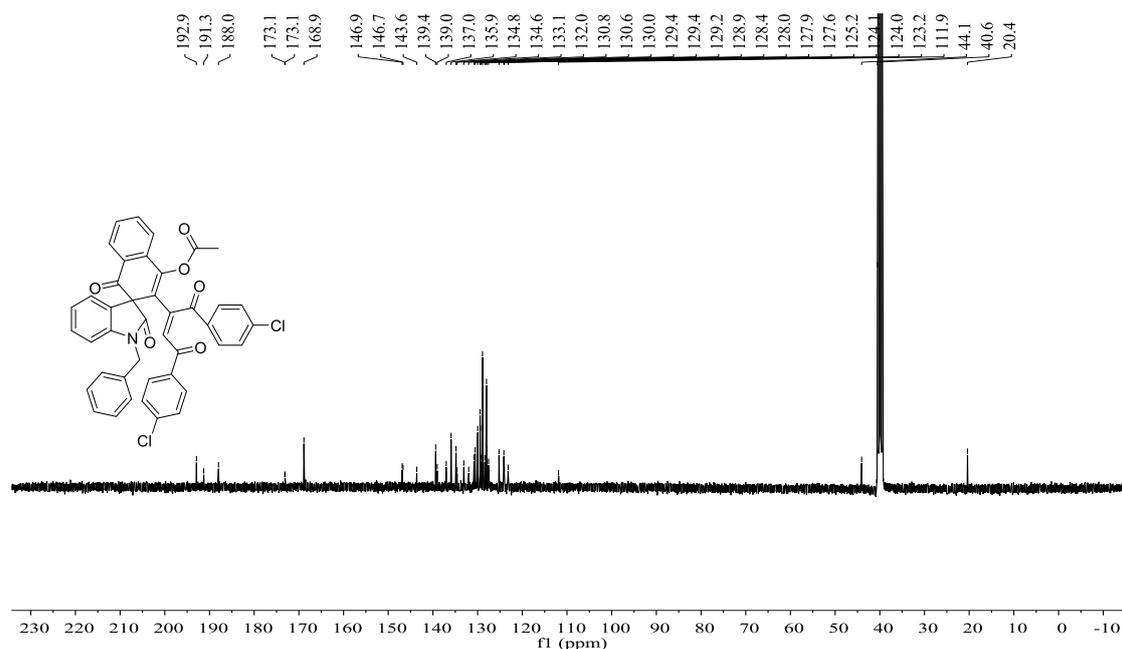
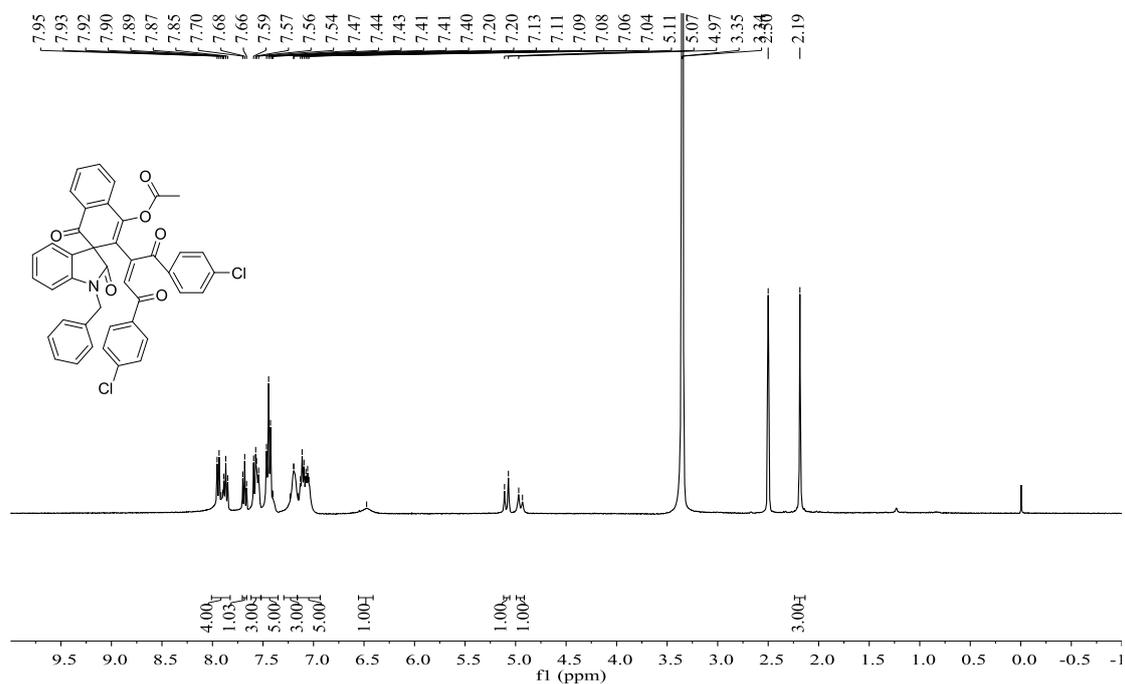
(Z)-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-1-butyl-5-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2o): yellow solid, 65%, m.p. 159-161 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.92-7.83 (m, 4H, ArH), 7.68-7.65 (m, 1H, ArH), 7.59-7.53 (m, 6H, ArH), 7.38-7.36 (m, 2H, ArH), 7.27-7.25 (m, 1H, ArH), 7.18-7.16 (m, 1H, ArH), 6.83-6.79 (m, 1H, ArH), 6.64 (br, 1H, CH), 3.76-3.66 (m, 2H, CH), 2.21 (s, 3H, CH₃), 2.16 (s, 3H, CH₃), 1.43-1.40 (m, 2H, CH), 1.30-1.25 (m, 2H, CH), 0.69 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.8, 191.1, 187.8, 172.5, 168.9, 141.7, 139.5, 138.9, 136.8, 135.0, 134.5, 133.1, 132.9, 131.8, 131.0, 130.4, 130.1, 129.6, 129.3, 128.2, 127.6, 125.1, 124.4, 123.7, 111.2, 111.2, 29.6, 20.9, 20.4, 19.8, 13.9; IR (KBr) ν: 3067, 2957, 2869, 1783, 1722, 1680, 1667, 1591, 1491, 1402, 1365, 1270, 1221, 1177, 1158, 1093, 965, 841, 756 cm⁻¹; HRMS (ESI) Calcd. for C₄₀H₃₁Cl₂NNaO₆ ([M+Na]⁺): 714.1421, Found: 714.1452.



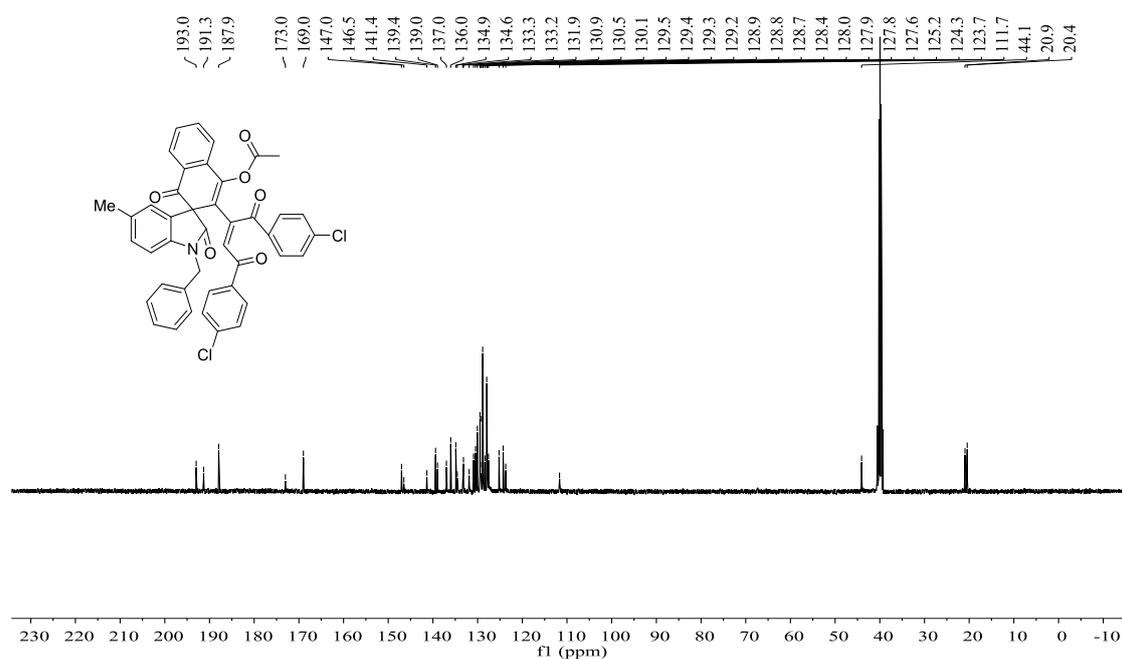
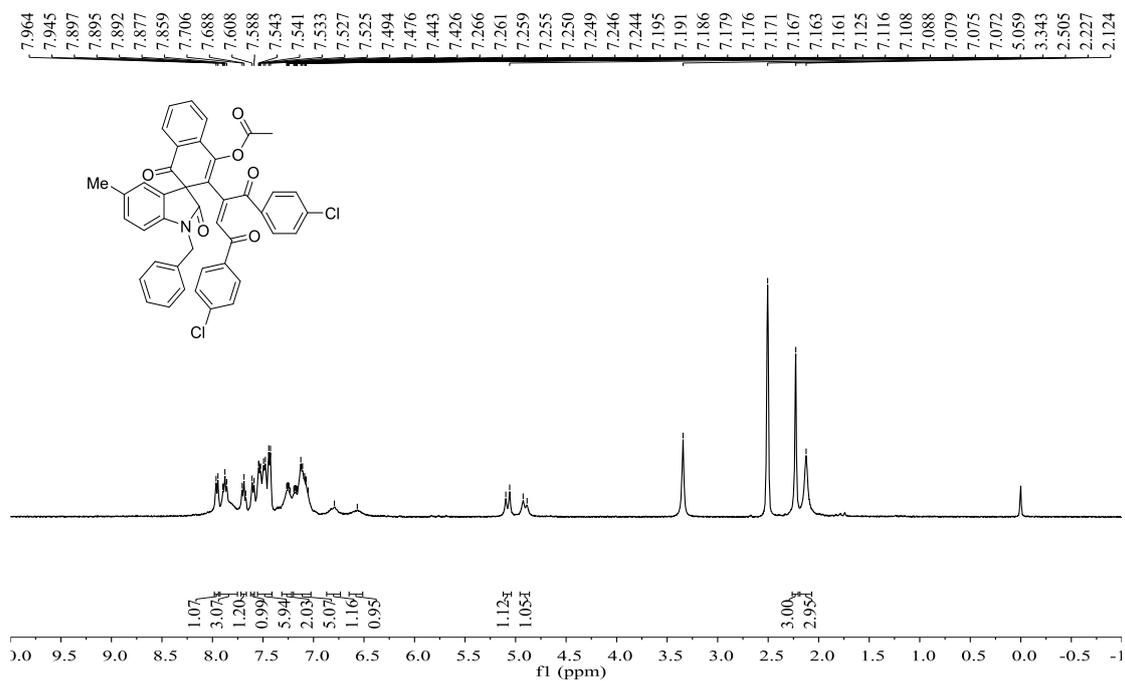
(Z)-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-1-butyl-5-chloro-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2p): yellow solid, 68%, m.p. 186-188 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.91 (d, *J* = 7.6 Hz, 1H, ArH), 7.88 (t, *J* = 8.0 Hz, 1H, ArH), 7.75-7.74 (m, 2H, ArH), 7.67 (t, *J* = 7.6 Hz, 1H, ArH), 7.60 (d, *J* = 7.6 Hz, 1H, ArH), 7.58 (d, *J* = 8.8 Hz, 2H, ArH), 7.54-7.48 (m, 5H, ArH), 7.28 (d, *J* = 8.0 Hz, 1H, ArH), 7.04 (s, 1H, ArH), 6.78 (s, 1H, CH), 3.76-3.66 (m, 2H, CH), 2.23 (s, 3H, CH₃), 1.44-1.39 (m, 2H, CH), 1.27-1.23 (m, 2H, CH), 0.72 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.2, 190.1, 187.4, 171.3, 168.5, 146.3, 146.1, 142.7, 139.0, 138.5, 136.4, 134.4, 133.8, 132.6, 131.2, 130.1, 129.8, 129.7, 129.1, 128.7, 127.8, 126.9, 124.7, 123.3, 123.2, 112.1, 40.0, 28.9, 19.9, 19.2, 13.3; IR (KBr) ν: 3018, 2954, 2911, 2827, 1726, 1710, 1694, 1379, 1375, 837, 756, 721 cm⁻¹; HRMS (ESI) Calcd. for C₃₉H₂₈Cl₃NNaO₆ ([M+Na]⁺): 734.0874, Found: 734.0885.



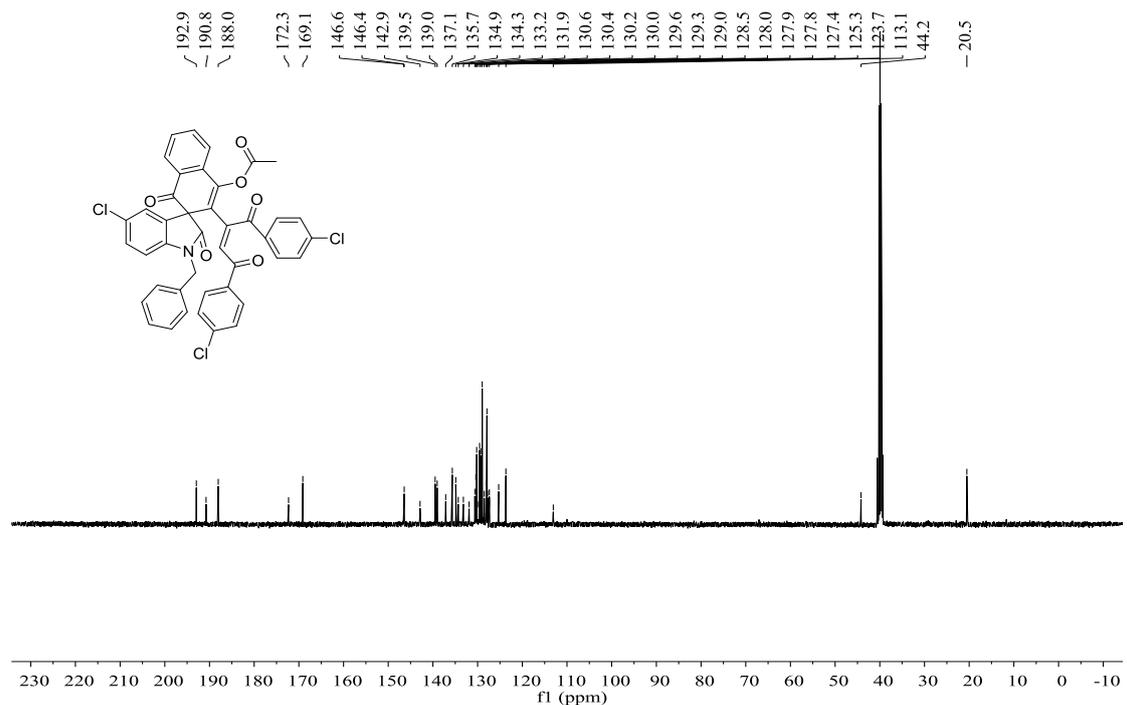
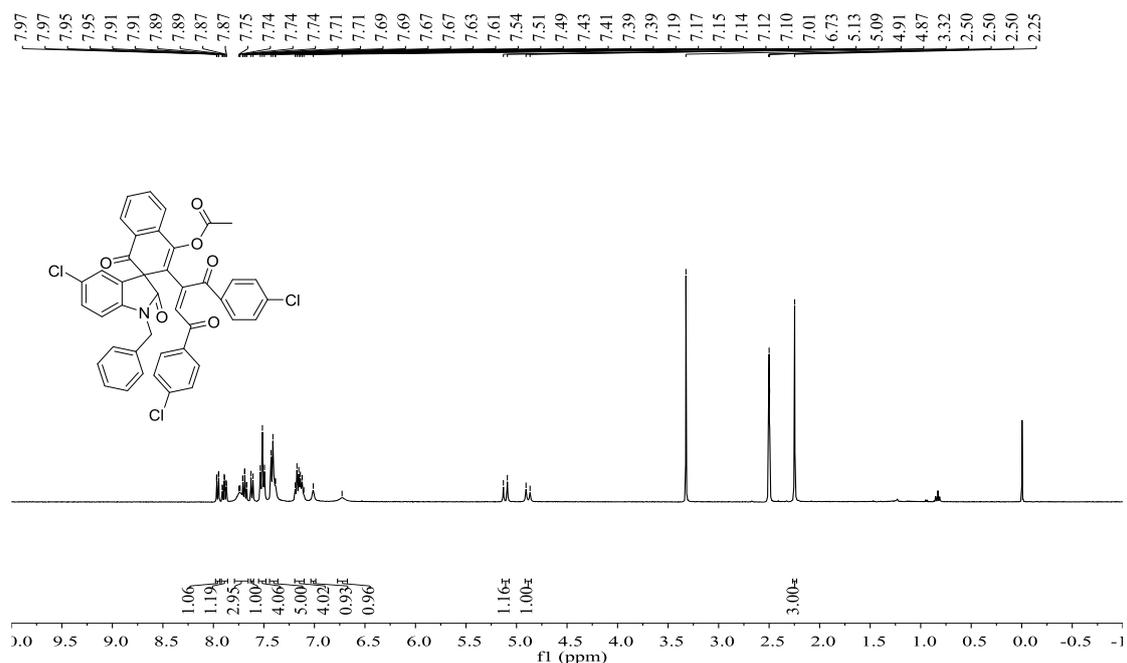
(Z)-1-benzyl-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2q): yellow solid, 70%, m.p. 180-182 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.95-7.85 (m, 4H, ArH), 7.68 (t, *J* = 7.6 Hz, 1H, ArH), 7.59-7.54 (m, 3H, ArH), 7.46-7.40 (m, 5H, ArH), 7.23-7.15 (m, 3H, ArH), 7.13-7.04 (m, 5H, ArH), 6.47 (br, 1H, CH), 5.09 (d, *J* = 15.6 Hz, 1H, CH), 4.95 (d, *J* = 14.8 Hz, 1H, CH), 2.19 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.9, 191.2, 187.9, 173.1, 173.1, 168.8, 146.8, 146.6, 143.6, 139.3, 139.0, 137.0, 135.9, 134.8, 134.5, 133.0, 131.9, 130.7, 130.5, 129.9, 129.4, 129.3, 129.1, 128.9, 128.3, 128.0, 127.9, 127.5, 125.2, 124.1, 124.0, 123.1, 111.9, 44.0, 40.5, 20.3; IR (KBr) ν: 3022, 2987, 2824, 1730, 1712, 1697, 1371, 763, 742 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₇Cl₂NNaO₆ ([M+Na]⁺): 734.1108, Found: 734.1115.



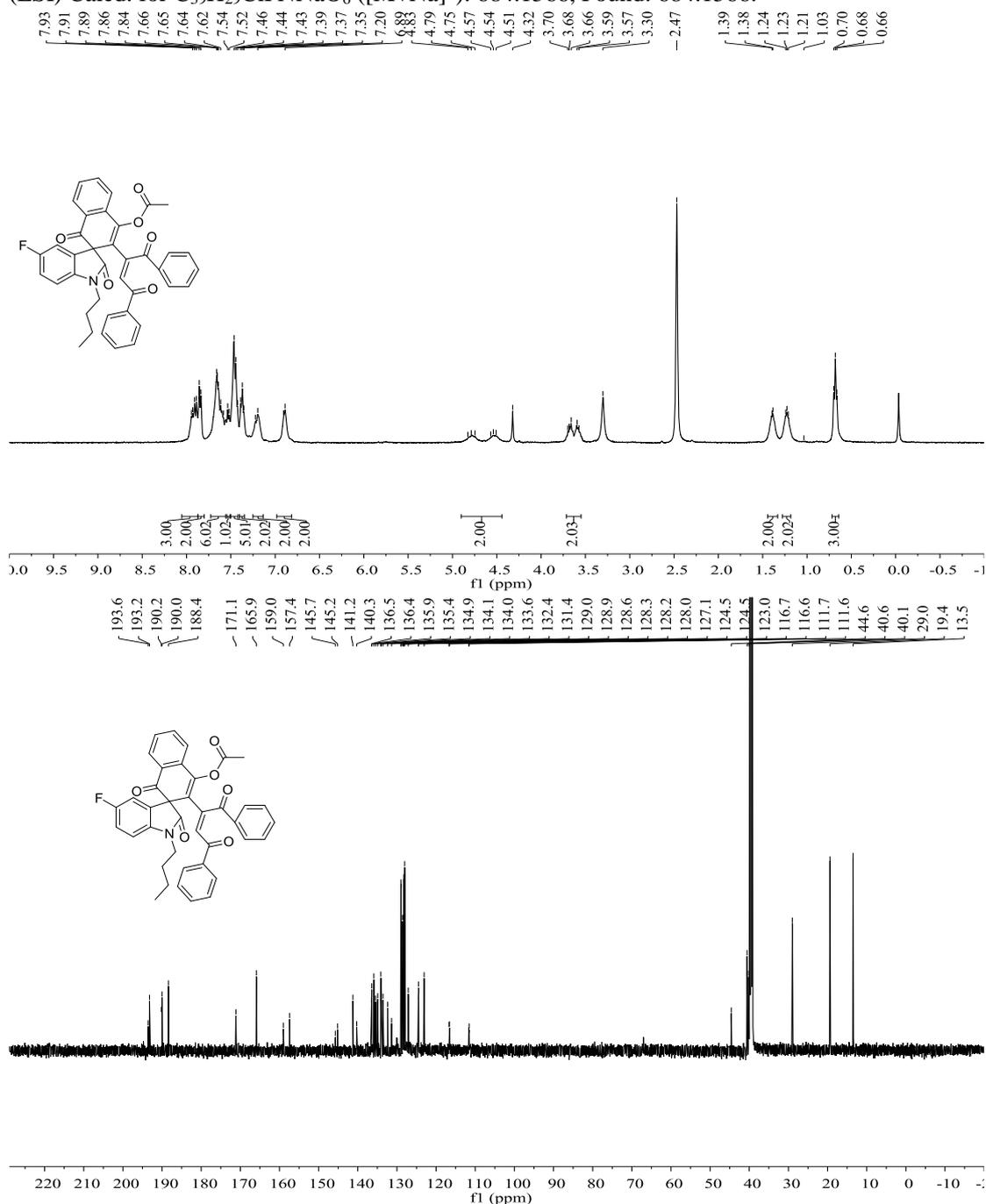
(Z)-1-benzyl-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-5-methyl-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2r): yellow solid, 66%, m.p. 189-191 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.95 (d, *J* = 7.6 Hz, 1H, ArH), 7.90-7.86 (m, 3H, ArH), 7.71-7.67 (m, 1H, ArH), 7.60 (d, *J* = 8.0 Hz, 1H, ArH), 7.54-7.43 (m, 6H, ArH), 7.27-7.23 (m, 2H, ArH), 7.20-7.05 (m, 5H, ArH), 6.79 (brs, 1H, ArH), 6.58 (brs, 1H, CH), 5.08 (d, *J* = 15.2 Hz, 1H, CH), 4.91 (d, *J* = 15.6 Hz, 1H, CH), 2.23 (s, 3H, CH₃), 2.12 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 193.0, 191.3, 187.9, 173.0, 169.0, 147.0, 146.5, 141.4, 139.4, 139.0, 137.0, 136.0, 134.9, 134.6, 133.3, 133.2, 131.9, 130.9, 130.5, 130.1, 129.5, 129.4, 129.3, 129.2, 128.9, 128.8, 128.7, 128.4, 128.0, 127.9, 127.8, 127.6, 125.2, 124.3, 123.7, 111.7, 44.1, 20.9, 20.4; IR (KBr) ν: 3016, 2971, 2831, 1719, 1703, 1687, 1375, 760, 722 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₂₉Cl₂NNaO₆ ([M+Na]⁺): 748.1264, Found: 748.1279.



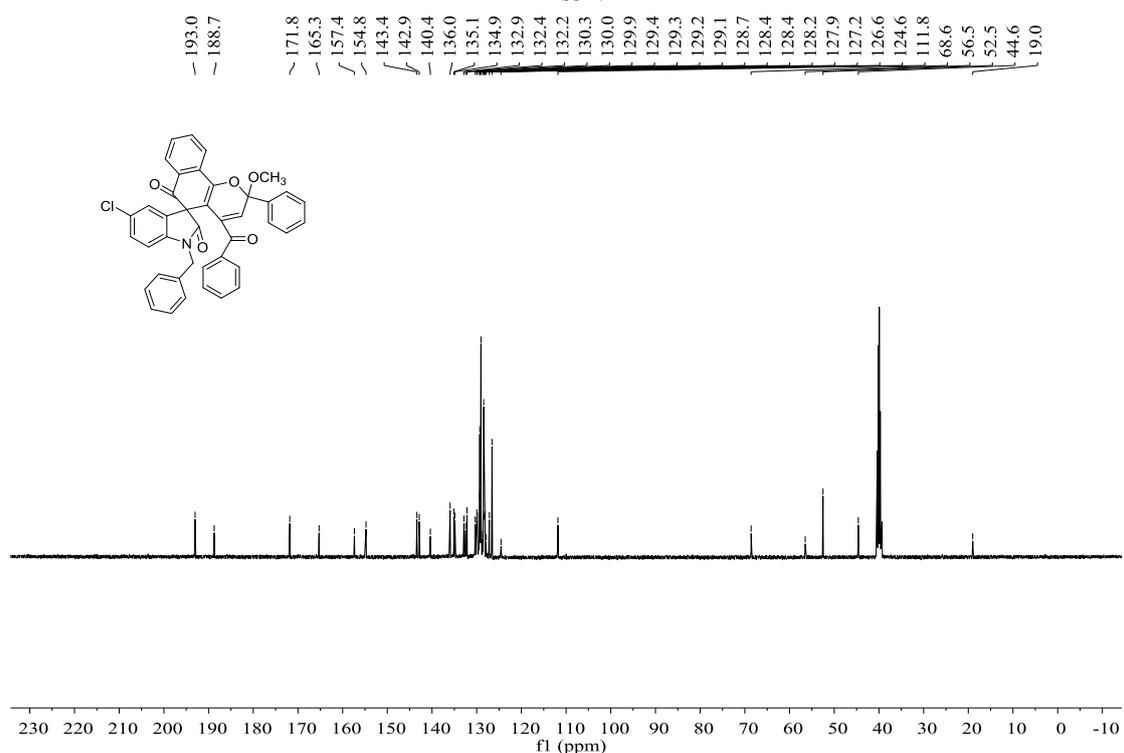
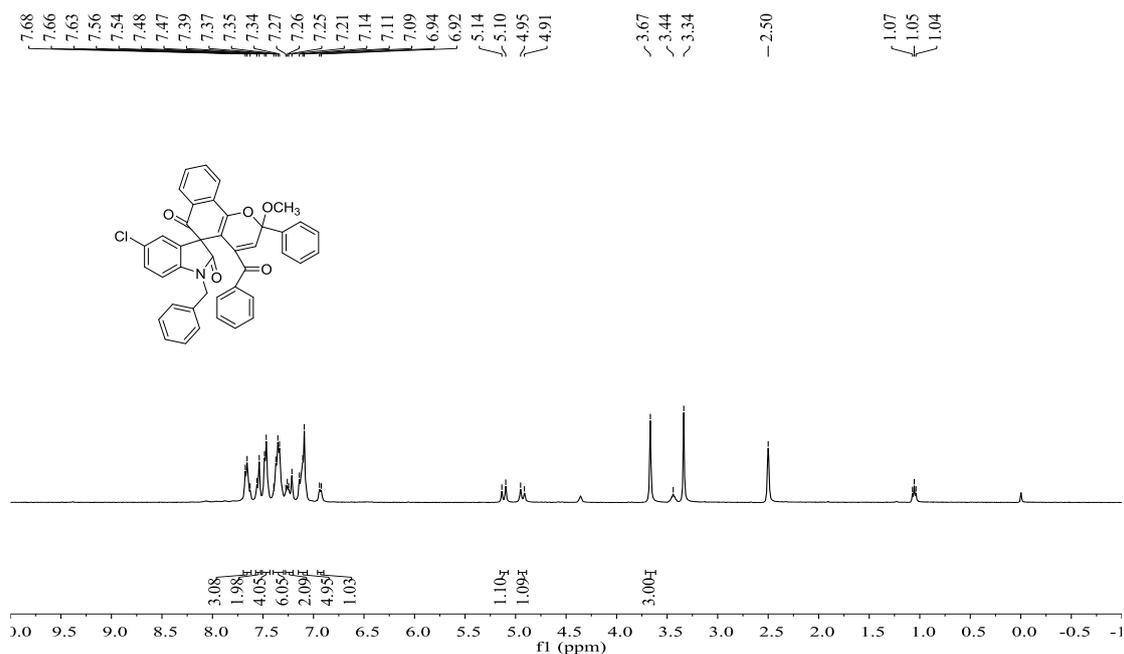
(Z)-1-benzyl-3'-(1,4-bis(4-chlorophenyl)-1,4-dioxobut-2-en-2-yl)-5-chloro-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2s): yellow solid, 71%, m.p. 187-189 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.97-7.95 (m, 1H, ArH), 7.91-7.87 (m, 1H, ArH), 7.75-7.67 (m, 3H, ArH), 7.62 (d, *J* = 8.0 Hz, 1H, ArH), 7.54-7.50 (m, 4H, ArH), 7.43-7.39 (m, 5H, ArH), 7.19-7.11 (m, 4H, ArH), 7.01 (br, 1H, ArH), 6.73 (br, 1H, CH), 5.11 (d, *J* = 15.6 Hz, 1H, CH), 4.89 (d, *J* = 16.0 Hz, 1H, CH), 2.25 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.9, 190.7, 188.0, 172.3, 169.1, 146.5, 146.4, 142.8, 139.4, 139.0, 137.1, 135.6, 134.8, 134.3, 133.1, 131.9, 130.6, 130.4, 130.2, 130.0, 129.5, 129.3, 128.9, 128.5, 128.0, 127.9, 127.7, 127.3, 125.2, 123.6, 113.0, 44.1, 20.5; IR (KBr) ν: 3033, 2961, 2813, 1778, 1724, 1680, 1602, 1483, 1363, 1271, 1228, 751, 704 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₆Cl₃NNaO₆ ([M+Na]⁺): 768.0718, Found: 768.0731.



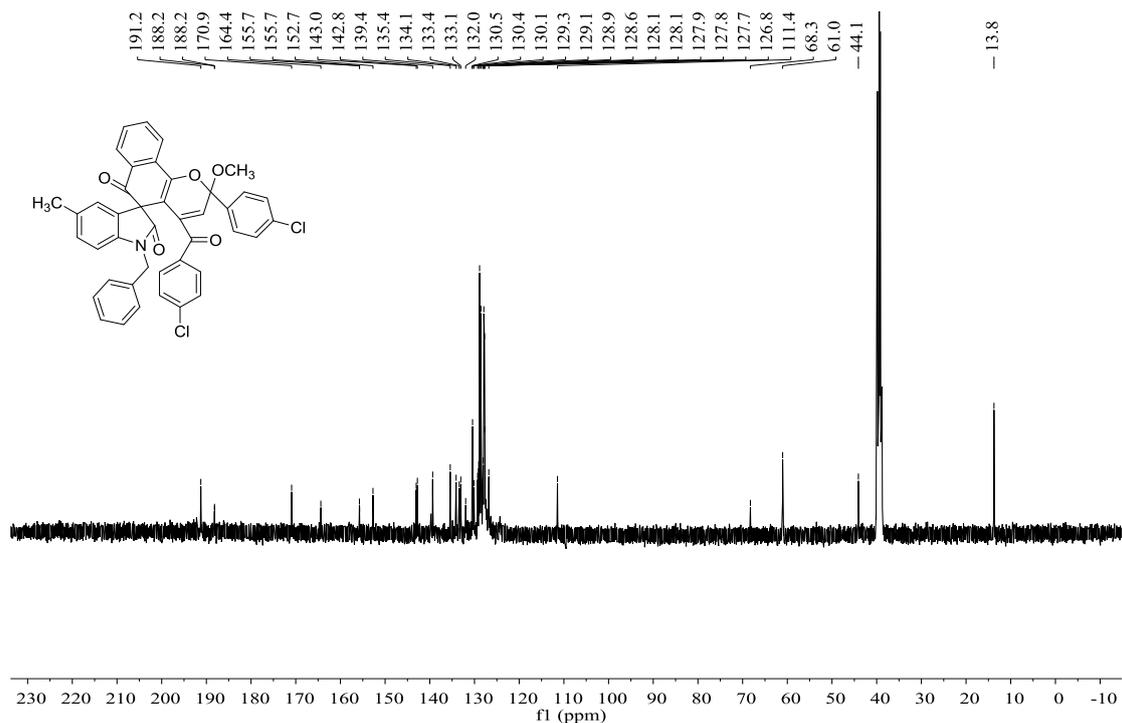
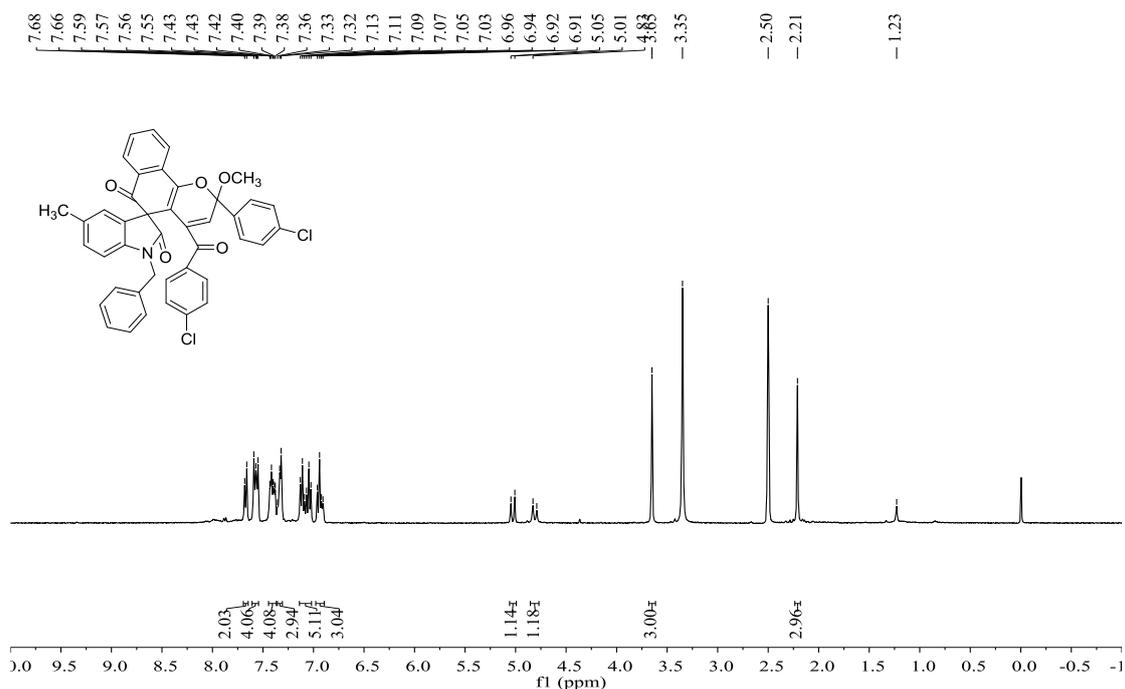
(Z)-1-butyl-3'-(1,4-dioxo-1,4-diphenylbut-2-en-2-yl)-5-fluoro-1',2-dioxo-1'H-spiro[indoline-3,2'-naphthalen]-4'-yl acetate (2t): yellow solid, 67%, m.p. 168-169 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.94-7.89 (m, 3H, ArH), 7.85 (d, *J* = 8.0 Hz, 2H, ArH), 7.66-7.58 (m, 6H, ArH), 7.53 (d, *J* = 7.6 Hz, 1H, ArH), 7.46-7.42 (m, 5H, ArH), 7.37 (t, *J* = 7.6 Hz, 2H, ArH), 7.22-7.19 (m, 2H, ArH), 6.90-6.88 (m, 2H, ArH, CH), 4.82-4.51 (m, 2H, CH), 3.70-3.57 (m, 2H, CH), 1.40-1.38 (m, 2H, CH), 1.24-1.21 (m, 2H, CH), 0.68 (t, *J* = 6.8 Hz, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 193.5, 193.2, 190.1, 189.9, 188.3, 171.1, 165.8, 159.0, 157.4, 145.7, 145.1, 141.2, 140.2, 136.4, 136.3, 135.8, 135.4, 134.9, 134.1, 134.0, 133.5, 132.3, 131.3, 128.9, 128.9, 128.5, 128.2, 128.2, 128.0, 127.1, 124.5, 124.4, 123.0, 116.7, 116.5, 111.6, 111.5, 44.6, 40.5, 40.1, 29.9, 19.3, 13.4; IR (KBr) ν: 3015, 2969, 2937, 2865, 1712, 1694, 1640, 1413, 1377, 818, 756, 729, 673 cm⁻¹; HRMS (ESI) Calcd. for C₃₉H₂₉ClFNNaO₆ ([M+Na]⁺): 684.1566, Found: 684.1566.



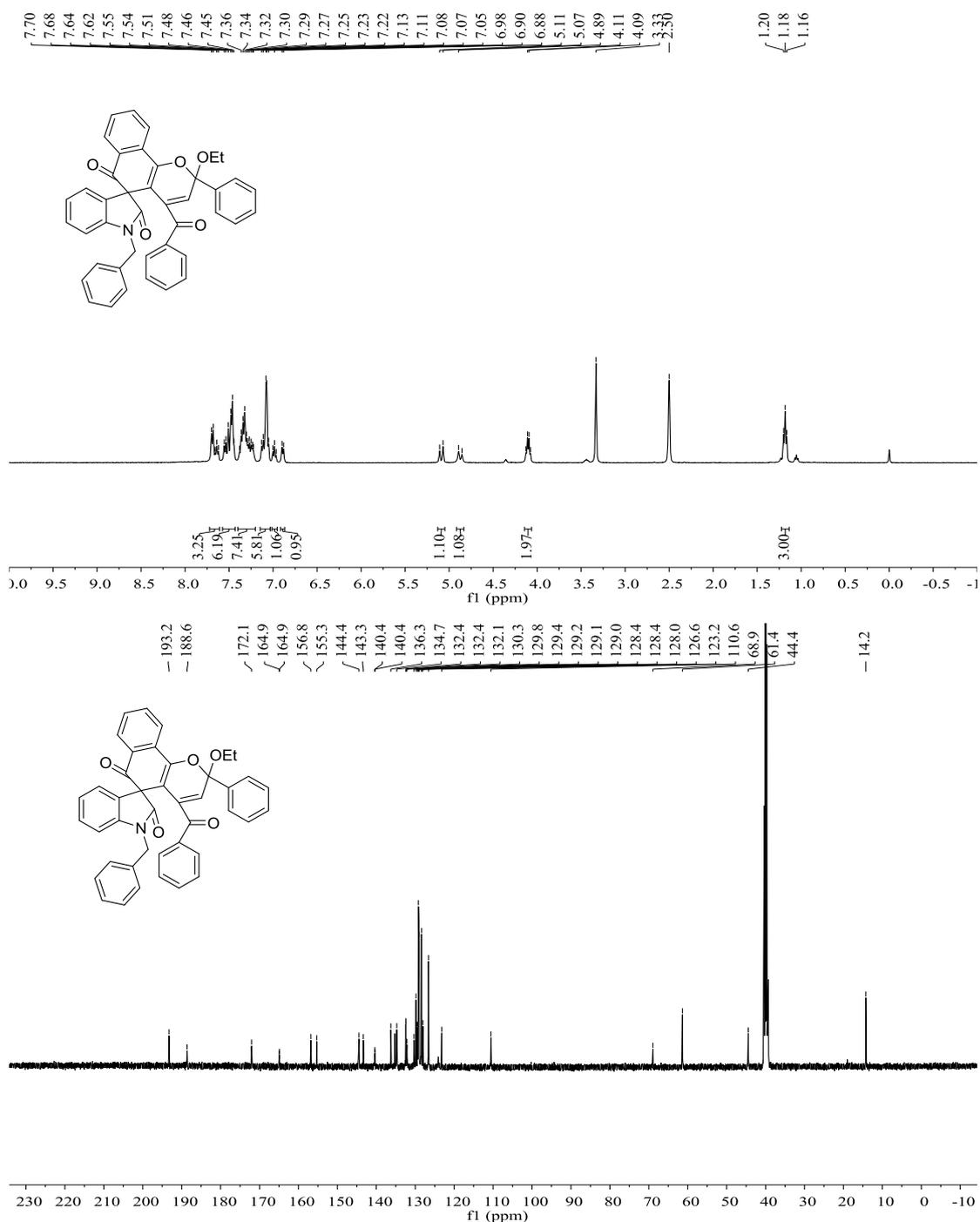
4-benzoyl-1'-benzyl-5'-chloro-2-methoxy-2-phenylspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3a): yellow solid, 67%, m.p. 173-175 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.68-7.63 (m, 3H, ArH), 7.56-7.54 (m, 2H, ArH), 7.49-7.46 (m, 4H, ArH), 7.39-7.34 (m, 6H, ArH), 7.27-7.21 (m, 2H, ArH), 7.14-7.09 (m, 5H, ArH), 6.93 (d, *J* = 6.8 Hz, 1H, CH), 5.12 (d, *J* = 15.6 Hz, 1H, CH), 4.93 (d, *J* = 14.8 Hz, 1H, CH), 3.67 (s, 3H, OCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 193.0, 188.7, 171.8, 165.2, 157.3, 154.7, 143.3, 142.8, 140.3, 135.9, 135.1, 134.8, 132.8, 132.4, 132.1, 130.3, 129.9, 129.8, 129.3, 129.3, 129.2, 129.0, 128.6, 128.4, 128.3, 128.1, 127.9, 127.1, 126.5, 124.5, 111.8, 68.5, 56.4, 52.5, 44.6, 19.0; IR (KBr) ν: 3063, 2924, 2858, 1965, 1717, 1651, 1487, 1448, 1333, 1273, 1165, 1084, 1017, 920, 864, 802, 707 cm⁻¹; HRMS (ESI) Calcd. for C₄₁H₂₈ClNaNO₅ ([M+Na]⁺): 672.1548, Found: 672.1559.



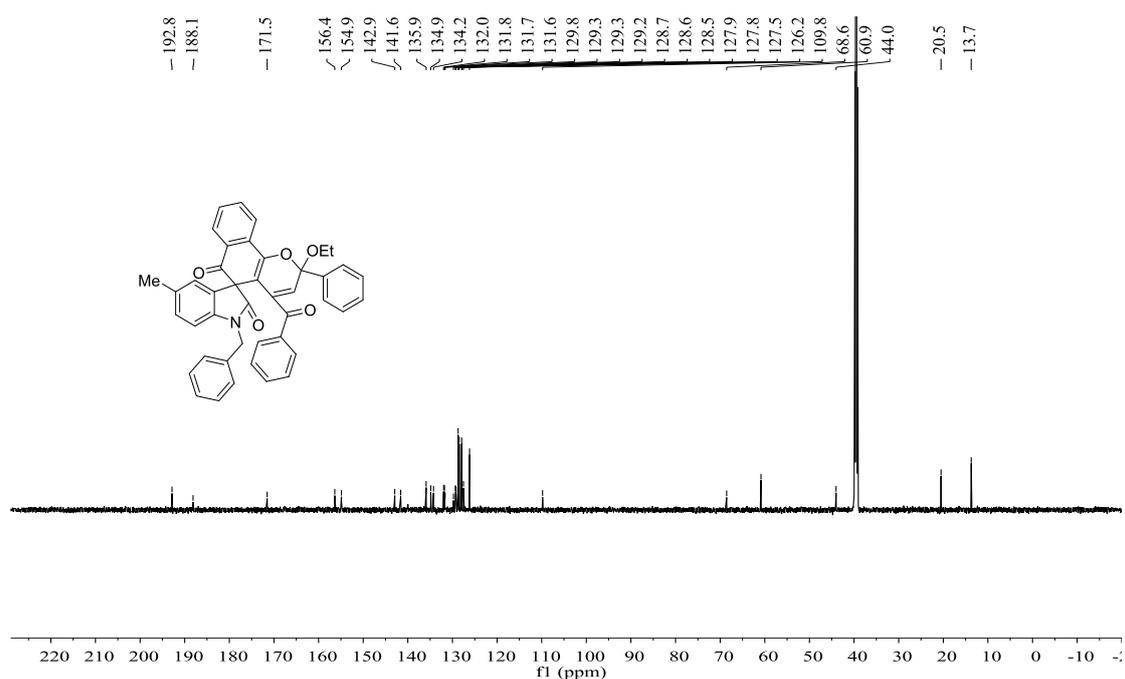
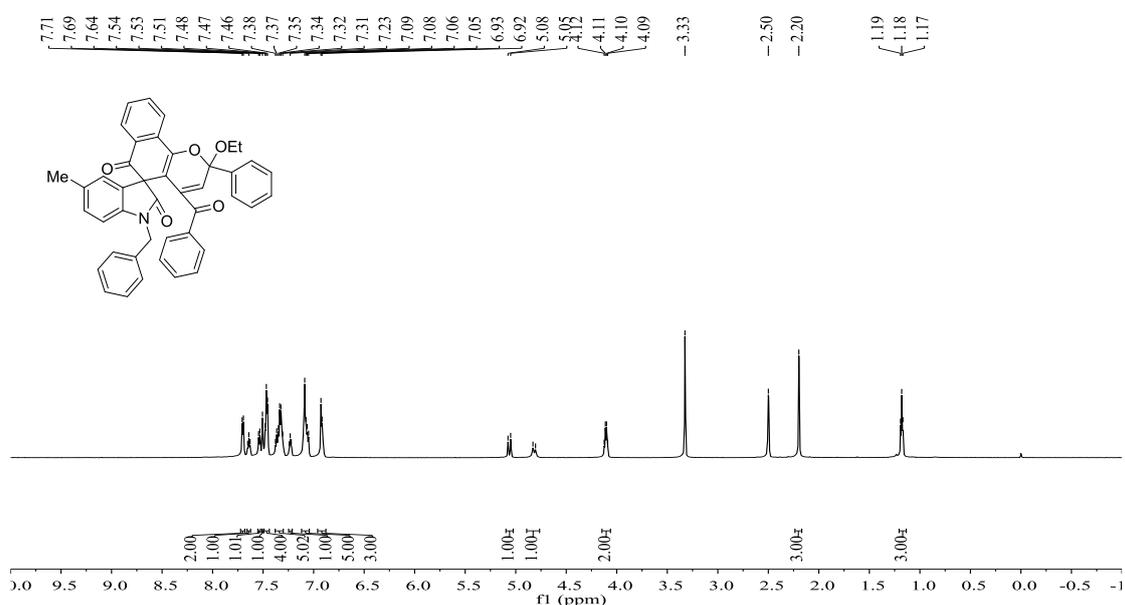
1'-benzyl-4-(4-chlorobenzoyl)-2-(4-chlorophenyl)-2-methoxy-5'-methylspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3b): yellow solid, 71%, m.p. 175-177 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.67 (d, *J* = 8.0 Hz, 2H, ArH), 7.59-7.55 (m, 4H, ArH), 7.43-7.38 (m, 4H, ArH), 7.36-7.32 (m, 3H, ArH), 7.13-7.03 (m, 5H, ArH), 6.96-6.91 (m, 3H, ArH), 5.03 (d, *J* = 15.6 Hz, 1H, CH), 4.81 (d, *J* = 15.2 Hz, 1H, CH), 3.65 (s, 3H, OCH₃), 2.21 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 191.2, 188.1, 188.1, 170.9, 164.3, 155.7, 155.7, 152.7, 143.0, 142.7, 139.3, 135.4, 134.1, 133.4, 133.0, 131.9, 130.5, 130.4, 130.1, 129.3, 129.0, 128.8, 128.5, 128.1, 128.0, 127.9, 127.7, 127.7, 126.7, 111.4, 68.2, 61.0, 44.0, 13.7; IR (KBr) ν: 3403, 2978, 2920, 1711, 1643, 1599, 1485, 1345, 1290, 1176, 1125, 1023, 972, 913, 857, 725 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₉Cl₂NaNO₅ ([M+Na]⁺): 720.1315, Found: 720.1328.



4-benzoyl-1'-benzyl-2-ethoxy-2-phenylspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (**3c**): yellow solid, 62%, m.p. 171-173 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.70-7.62 (m, 3H, ArH), 7.55-7.45 (m, 6H, ArH), 7.38-7.22 (m, 7H, ArH), 7.13-7.05 (m, 6H, ArH), 7.00-6.97 (m, 1H, ArH), 6.89 (d, *J* = 7.6 Hz, 1H, CH), 5.09 (d, *J* = 15.6 Hz, 1H, CH), 4.87 (d, *J* = 15.2 Hz, 1H, CH), 4.13-4.08 (m, 2H, CH), 1.18 (t, *J* = 6.8 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 193.2, 188.6, 172.0, 164.9, 164.9, 156.8, 155.2, 144.4, 143.3, 140.3, 140.3, 136.2, 134.7, 132.4, 132.3, 132.1, 130.3, 129.7, 129.4, 129.1, 129.0, 128.9, 128.3, 128.3, 128.0, 126.6, 123.2, 110.5, 68.9, 61.3, 44.4, 14.2; IR (KBr) ν: 3062, 2928, 2866, 1780, 1713, 1595, 1493, 1448, 1337, 1281, 1189, 1090, 1022, 919, 865, 804, 713 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₃₁NaNO₅ ([M+Na]⁺): 652.2094, Found: 652.2103.

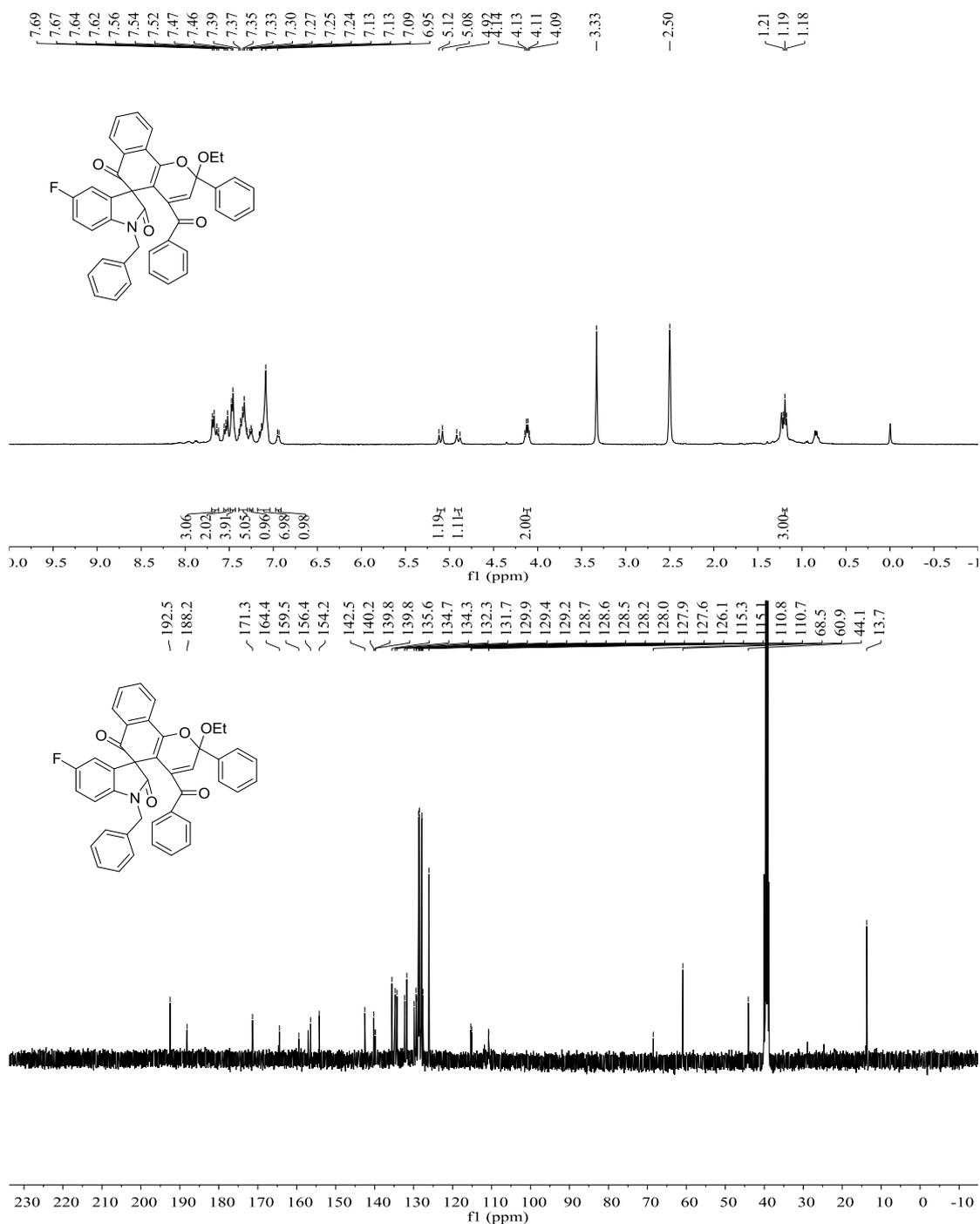


(Z)-1-benzyl-2'-ethoxy-5-methyl-3'-(2-oxo-2-phenylethylidene)-2'-phenyl-2',3'-dihydro-5'H-spiro[indoline-3,4'-naphtho[1,2-*b*]furan]-2,5'-dione (3d): yellow solid, 63%, m.p. 173-175 °C; ¹H NMR (600 MHz, DMSO-*d*₆) δ: 7.70 (d, *J* = 7.2 Hz, 2H, ArH), 7.64 (t, *J* = 7.2 Hz, 1H, ArH), 7.54 (d, *J* = 7.8 Hz, 1H, ArH), 7.51 (s, 1H, ArH), 7.48-7.45 (m, 4H, ArH), 7.38-7.31 (m, 5H, ArH), 7.23 (t, *J* = 6.6 Hz, 1H, ArH), 7.09-7.05 (m, 5H, ArH), 6.93-6.92 (m, 3H, 2ArH, CH), 5.06 (d, *J* = 12.0 Hz, 1H, CH), 4.82 (d, *J* = 12.0 Hz, 1H, CH), 4.12-4.09 (m, 2H, CH), 2.20 (s, 3H, CH₃), 1.18 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.8, 188.1, 171.5, 156.3, 154.8, 142.9, 141.5, 135.8, 134.8, 134.2, 131.9, 131.7, 131.7, 131.6, 129.7, 129.3, 129.2, 129.1, 128.7, 128.6, 128.4, 127.9, 127.8, 127.5, 126.1, 109.7, 68.5, 60.8, 44.0, 20.5, 13.7; IR (KBr) ν: 3021, 2987, 2925, 1701, 1688, 1413, 1369, 808, 761, 724, 677 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₃₃NNaO₆ ([M+Na]⁺): 666.2251, Found: 666.2266.

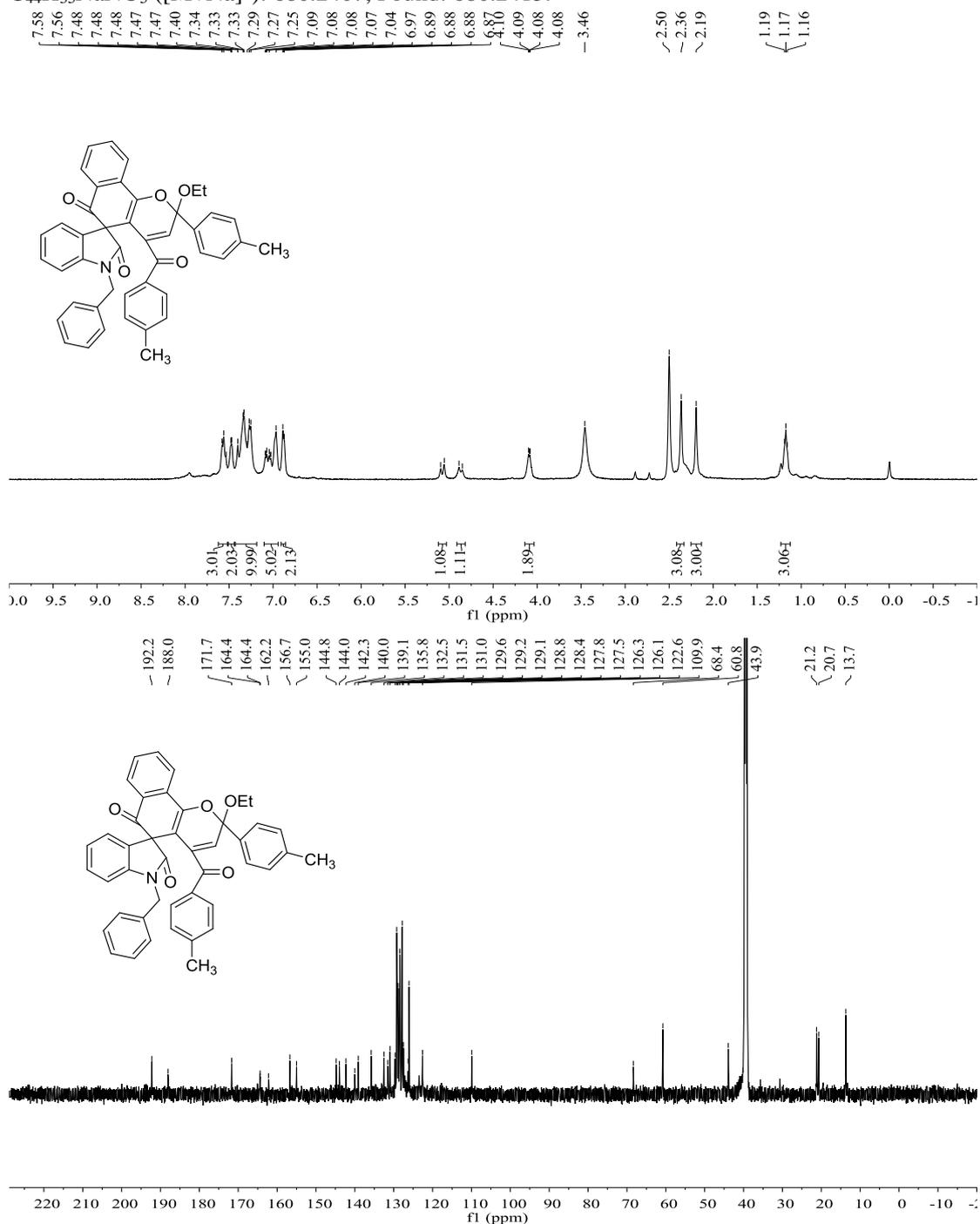


4-benzoyl-1'-benzyl-2-ethoxy-5'-fluoro-2-phenylspiro[benzo[h]chromene-5,3'-indoline]-

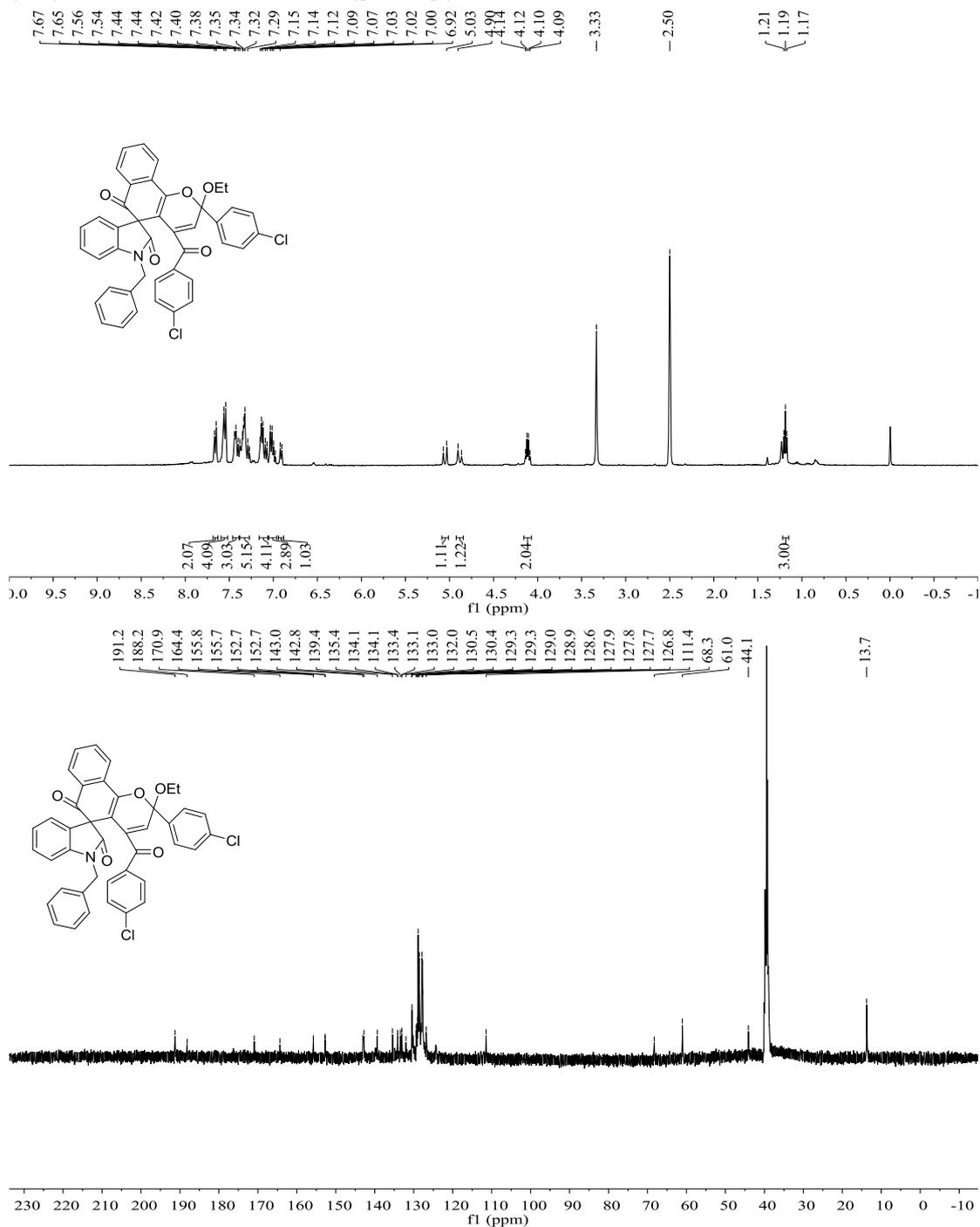
2',6(2H)-dione (3e): yellow solid, 66%, m.p. 177-179 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.69-7.62 (m, 3H, ArH), 7.56-7.52 (m, 2H, ArH), 7.48-7.46 (m, 4H, ArH), 7.39-7.30 (m, 5H, ArH), 7.27-7.24 (m, 1H, ArH), 7.16-7.08 (m, 7H, ArH), 6.94 (d, *J* = 6.4 Hz, 1H, ArH), 5.10 (d, *J* = 16.0 Hz, 1H, CH), 4.90 (d, *J* = 15.2 Hz, 1H, CH), 4.14-4.09 (m, 2H, CH), 1.19 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 192.4, 188.1, 171.3, 164.4, 159.4, 156.4, 154.2, 142.5, 140.2, 139.8, 139.8, 135.5, 134.7, 134.2, 132.2, 131.7, 129.8, 129.3, 129.2, 128.7, 128.5, 128.5, 128.1, 127.9, 127.8, 127.5, 126.0, 115.3, 115.0, 110.7, 110.6, 68.5, 60.8, 44.0, 13.6.; IR (KBr) ν: 3063, 2926, 1790, 1717, 1630, 1586, 1526, 1485, 1408, 1336, 1245, 1166, 1086, 1013, 967, 884, 817 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₃₀NaFNO₅ ([M+Na]⁺): 670.2000, Found: 670.2005.



1'-benzyl-2-ethoxy-4-(4-methylbenzoyl)-2-(p-tolyl)spiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3f): yellow solid, 61%, m.p. 174-176 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.58-7.53 (m, 3H, ArH), 7.48-7.47 (m, 2H, ArH), 7.40-7.25 (m, 10H, ArH), 7.09-6.97 (m, 5H, ArH), 6.89-6.87 (m, 2H, ArH), 5.08 (d, *J* = 16.8 Hz, 1H, CH), 4.86 (d, *J* = 15.8 Hz, 1H, CH), 4.10-4.08 (m, 2H, CH), 2.36 (s, 3H, CH₃), 2.19 (s, 3H, CH₃), 1.17 (t, *J* = 6.0 Hz, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.2, 188.0, 171.6, 164.4, 164.3, 162.1, 156.7, 155.0, 144.8, 143.9, 142.2, 140.0, 139.1, 135.7, 132.5, 131.5, 130.9, 129.6, 129.2, 129.0, 128.8, 128.4, 127.8, 127.4, 126.2, 126.0, 122.5, 109.9, 68.3, 60.7, 43.9, 21.2, 20.6, 13.6; IR (KBr) ν: 3065, 2922, 2855, 1718, 1652, 1589, 1488, 1439, 1335, 1284, 1182, 1134, 1089, 1011, 920, 821, 755 cm⁻¹; HRMS (ESI) Calcd. for C₄₄H₃₅NaNO₅ ([M+Na]⁺): 680.2407, Found: 680.2415.

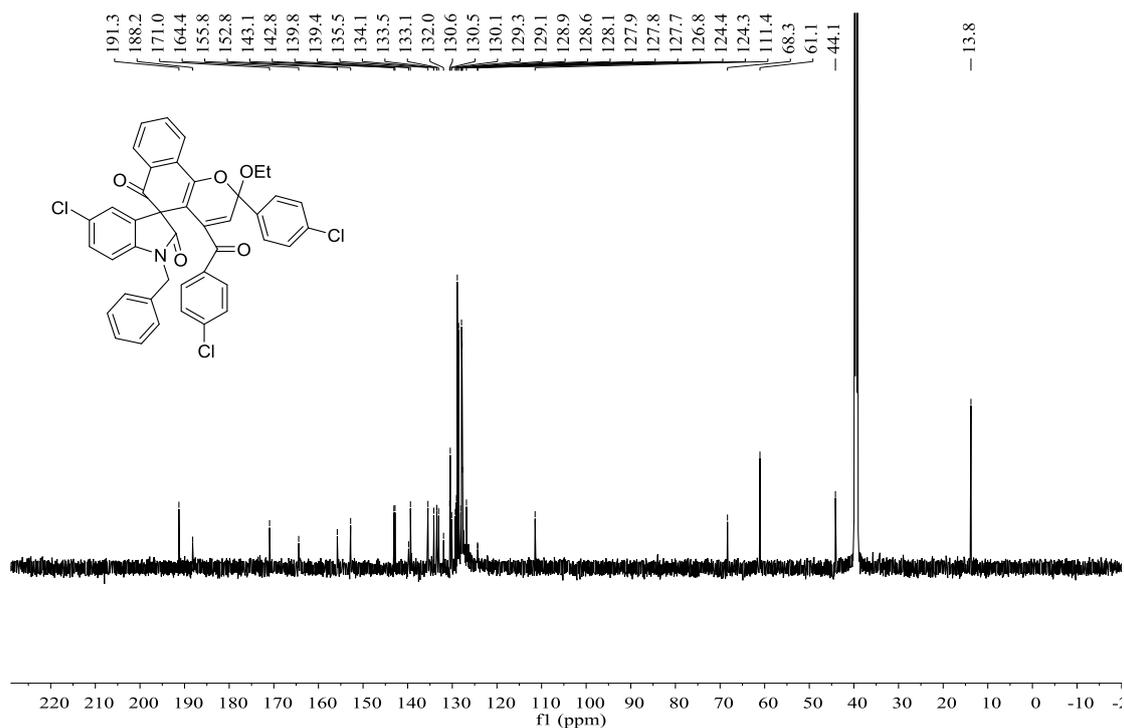
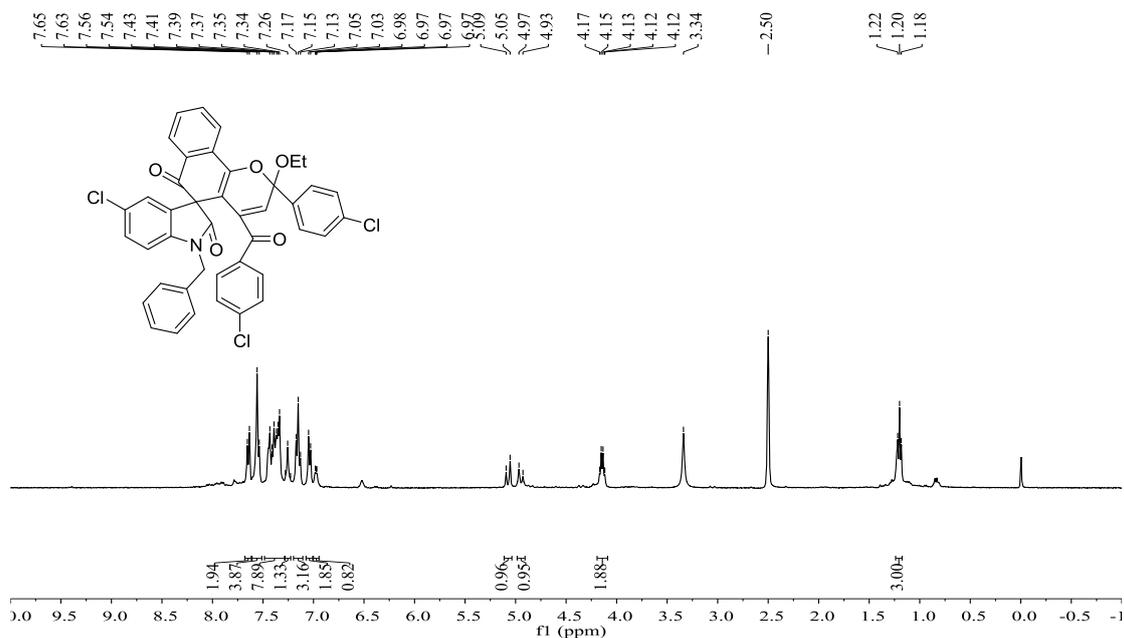


1'-benzyl-4-(4-chlorobenzoyl)-2-(4-chlorophenyl)-2-ethoxyspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3g): yellow solid, 67%, m.p. 180-182 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.66 (d, *J* = 7.6 Hz, 2H, ArH), 7.56-7.54 (m, 4H, ArH), 7.44-7.40 (m, 3H, ArH), 7.38-7.27 (m, 5H, ArH), 7.15-7.07 (m, 4H, ArH), 7.04-7.98 (m, 3H, ArH), 6.91 (d, *J* = 7.2 Hz, 1H, ArH), 5.05 (d, *J* = 15.2 Hz, 1H, CH), 4.88 (d, *J* = 16.4 Hz, 1H, CH), 4.14-4.09 (m, 2H, CH), 1.19 (t, *J* = 6.8 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 191.2, 188.1, 170.9, 164.3, 155.7, 155.7, 152.7, 152.7, 143.0, 142.7, 139.3, 135.4, 134.1, 134.0, 133.4, 133.0, 133.0, 131.9, 130.5, 130.4, 129.3, 129.2, 129.0, 128.8, 128.5, 127.9, 127.7, 127.6, 126.8, 111.4, 68.2, 61.0, 44.0, 13.7; IR (KBr) ν: 3060, 1966, 1713, 1643, 1482, 1435, 1330, 1292, 1237, 1179, 1079, 1016, 913, 852, 807, 711 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₉NaCl₂NO₅ ([M+Na]⁺): 720.1315, Found: 720.1324.

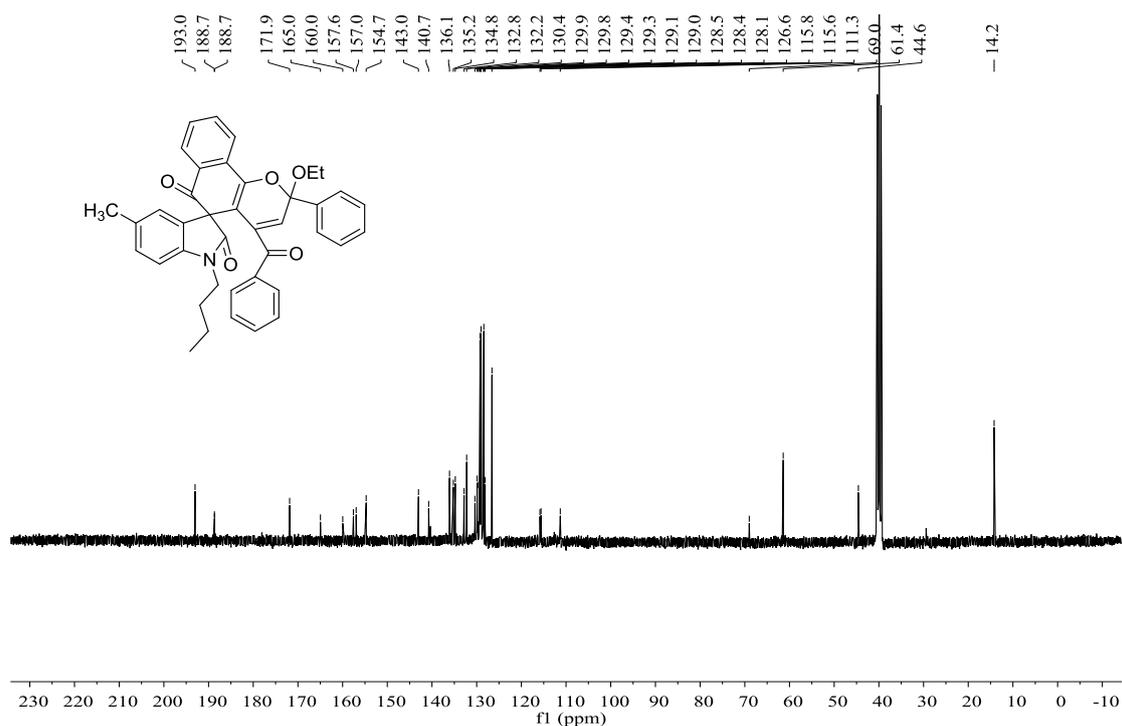
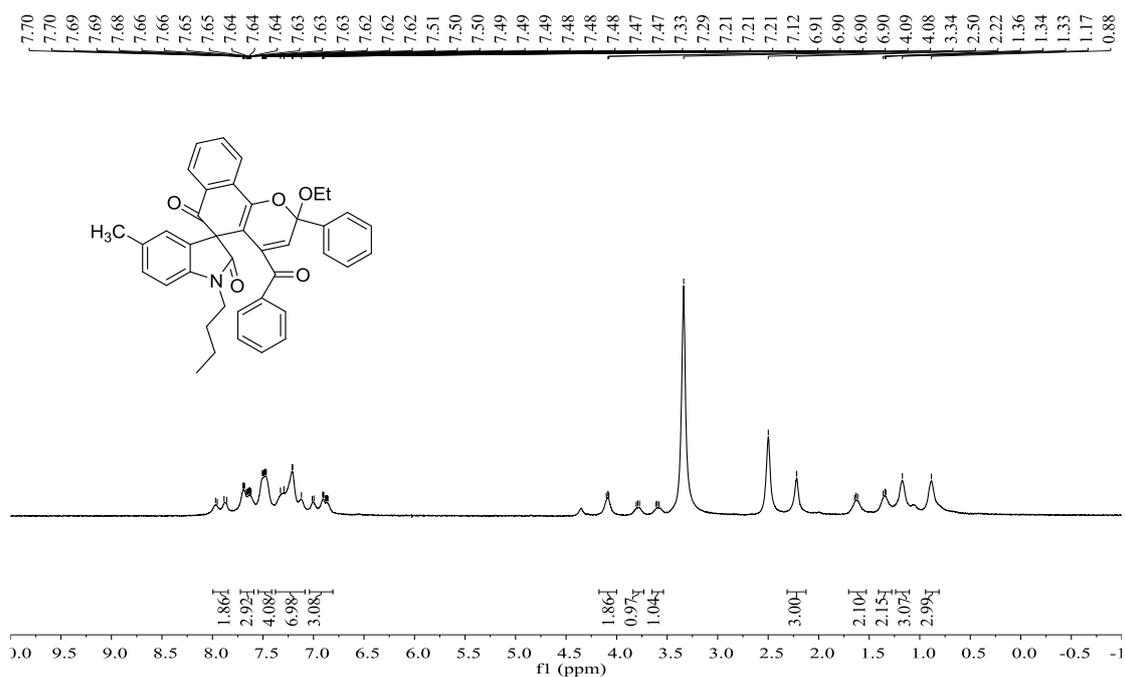


1'-benzyl-5'-chloro-4-(4-chlorobenzoyl)-2-(4-chlorophenyl)-2-

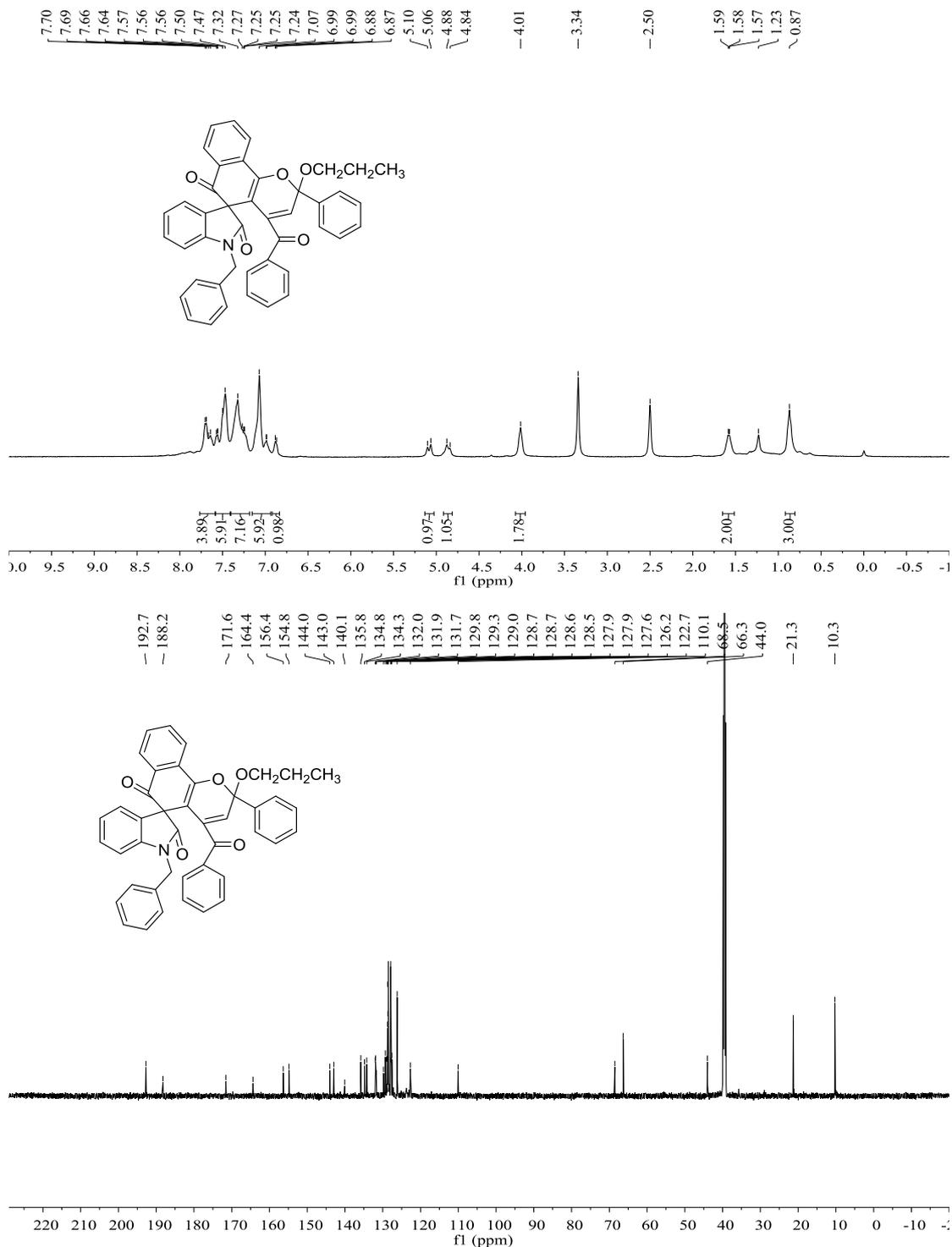
ethoxyspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3h): yellow solid, 73%, m.p. 183-185 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.64 (d, *J* = 8.4 Hz, 2H, ArH), 7.56-7.54 (m, 4H, ArH), 7.43-7.34 (m, 8H, ArH), 7.28-7.23 (m, 1H, ArH), 7.17-7.13 (m, 3H, ArH), 7.04 (d, *J* = 8.0 Hz, 2H, ArH), 6.98-6.97 (m, 1H, ArH), 5.07 (d, *J* = 15.6 Hz, 1H, CH), 4.95 (d, *J* = 16.0 Hz, 1H, CH), 4.17-4.12 (m, 2H, CH), 1.20 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 191.2, 188.1, 170.9, 164.4, 155.7, 152.7, 143.0, 142.8, 139.7, 139.3, 135.4, 134.1, 133.4, 133.0, 131.9, 130.5, 130.4, 130.1, 129.3, 129.0, 128.8, 128.5, 128.0, 127.9, 127.8, 127.7, 126.8, 124.3, 124.2, 111.4, 68.3, 61.0, 44.1, 13.7; IR (KBr) ν: 3065, 2926, 1717, 1656, 1594, 1482, 1341, 1280, 1174, 1090, 1011, 914, 838, 749, 706 cm⁻¹; HRMS (ESI) Calcd. for C₄₂H₂₉NaCl₂NO₅ ([M+Na]⁺): 754.0925, Found: 754.0962.



4-benzoyl-1'-butyl-2-ethoxy-5'-methyl-2-phenylspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3i): yellow solid, 64%, m.p. 171-173 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.97-7.86 (m, 2H, ArH), 7.70-7.62 (m, 3H, ArH), 7.51-7.47 (m, 4H, ArH), 7.33-7.12 (m, 7H, ArH), 7.01-6.86 (m, 3H, ArH), 4.10-4.08 (m, 2H, CH), 3.81-3.77 (m, 1H, CH), 3.61-3.58 (m, 1H, CH), 2.22 (s, 3H, CH₃), 1.65-1.61 (m, 2H, CH), 1.36-1.34 (m, 2H, CH), 1.17 (s, 3H, CH₃), 0.88 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 193.0, 188.6, 188.6, 171.8, 164.9, 159.9, 157.5, 156.9, 154.7, 143.0, 140.7, 136.0, 135.2, 134.7, 132.8, 132.2, 130.3, 129.8, 129.7, 129.4, 129.2, 129.0, 129.0, 128.4, 128.4, 128.1, 126.5, 115.8, 115.6, 111.2, 69.0, 61.4, 44.5, 14.2; IR (KBr) ν: 3061, 2921, 1964, 1713, 1623, 1587, 1520, 1444, 1367, 1226, 1126, 1054, 970, 884, 818, 707 cm⁻¹; HRMS (ESI) Calcd. for C₄₀H₃₅NaNO₅ ([M+Na]⁺): 632.2407, Found: 632.2420.



4-benzoyl-1'-benzyl-2-phenyl-2-propoxyspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3j): yellow solid, 62%, m.p. 185-187 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.70-7.64 (m, 4H, ArH), 7.57-7.47 (m, 6H, ArH), 7.32-7.24 (m, 7H, ArH), 7.07-6.99 (m, 6H, ArH), 6.88-6.87 (m, 1H, ArH), 5.08 (d, *J* = 14.8 Hz, 1H, CH), 4.86 (d, *J* = 15.2 Hz, 1H, CH), 1.59-1.57 (m, 2H, CH), 0.87 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.7, 188.1, 171.5, 164.3, 156.3, 154.8, 144.0, 142.9, 140.0, 135.8, 134.8, 134.2, 131.9, 131.9, 131.7, 129.7, 129.2, 128.9, 128.7, 128.6, 128.6, 128.5, 127.9, 127.8, 127.5, 126.1, 122.7, 110.0, 68.5, 66.3, 44.0, 21.2, 10.2; IR (KBr) ν: 3060, 2924, 2857, 1959, 1716, 1653, 1600, 1480, 1339, 1275, 1172, 1083, 1021, 966, 913, 855, 702 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₃₃N₂O₅ ([M+Na]⁺): 666.2251, Found: 666.2257.



4-benzoyl-1'-benzyl-5'-chloro-2-phenyl-2-propoxyspiro[benzo[h]chromene-5,3'-indoline]-2',6(2H)-dione (3k): yellow solid, 66%, m.p. 189-191 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 7.68-7.63 (m, 2H, ArH), 7.60-7.46 (m, 6H, ArH), 7.40-7.21 (m, 7H, ArH), 7.18-7.08 (m, 6H, ArH), 6.92-6.91 (m, 2H, ArH), 5.10 (d, *J* = 15.2 Hz, 1H, CH), 4.93 (d, *J* = 15.6 Hz, 1H, CH), 4.06-4.02 (m, 2H, CH), 1.63-1.58 (m, 2H, CH), 0.88 (s, 3H, CH₃); ¹³C NMR (150 MHz, DMSO-*d*₆) δ: 192.4, 188.3, 171.3, 164.4, 156.4, 154.1, 142.9, 142.6, 140.0, 135.4, 134.7, 134.3, 132.3, 131.8, 131.7, 129.9, 129.4, 129.2, 128.8, 128.7, 128.6, 128.5, 127.9, 127.6, 126.6, 126.1, 111.3, 68.2, 66.3, 44.1, 21.3, 10.2; IR (KBr) ν: 3068, 2966, 1965, 1712, 1648, 1596, 1530, 1482, 1443, 1336, 1274, 1170, 1079, 1019, 918, 816 cm⁻¹; HRMS (ESI) Calcd. for C₄₃H₃₂NaClO₅ ([M+Na]⁺): 700.1861, Found: 700.1872.

