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Divergent synthesis of dual 1,4-dihydropyridines with different substituted patterns from enaminones and aldehydes through domino reactions †

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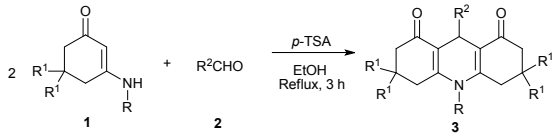
1. General Information

All compounds were fully characterized by spectroscopic data. NMR spectra were recorded on a Bruker DRX400 (^{1}H: 400 MHz, ^{13}C: 100 MHz). Chemical shifts (δ) are expressed in units of ppm, and *J* values are given in Hz. $CDCl_3$ was used as solvent. IR spectra were recorded on a FT-IR Thermo Nicolet Avatar 360 using a KBr pellet. The reactions were monitored by thin-layer chromatography (TLC) using silica gel GF254. The melting points are uncorrected and were determined on a XT-4A melting point apparatus. HRMs were performed on an Agilent LC/MSD TOF instrument and a Monoisotopic Mass instrument. All chemicals and solvents were used as received without further purification unless otherwise stated.

All chemicals and solvents were used as received without further purification unless otherwise stated. Column chromatography was performed on silica gel (200–300 mesh).

2. Synthesis and Spectral Data of Compounds 3

General procedure for the synthesis of 1,4-dihydropyridines 3



A mixture of enaminones **1** (1.0 mmol), aldehydes **2** (0.6 mmol), *p*-TSA (0.3 mmol), and EtOH (15 mL) was stirred at reflux for 3 h. After the desired product was formed as indicated by TLC, the reaction mixture was quenched with saturated NH_4Cl solution (2 mL) and extracted with ethyl acetate (40 mL). The organic phase were dried over Mg_2SO_4, and concentrated under vacuum. The residue was purified by flash chromatography (petroleum ether/ethyl acetate = 2:3) giving a white solid **3**.

Spectroscopic data of 1,4-dihydropyridines 3

9-(4-chlorophenyl)-10-(4-fluorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3a)

White solid; Mp 296–298 °C; IR (KBr): 2959, 1649, 1618, 1500, 1371, 1226 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ =7.35 (d, J = 8.4 Hz, 2H, ArH), 7.20–7.29 (m, 6H, ArH), 5.24 (s, 1H, CH), 2.03–2.22 (m, 6H, CH_2), 1.78–1.83 (m, 2H, CH_2), 0.96 (s, 6H, 2×CH_3), 0.81 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 162.3 (d, $^1J_{C–F}$ = 248.8 Hz), 162.3 (d, $^1J_{C–F}$ = 248.8 Hz), 149.7, 144.6, 134.9, 131.6, 130.9, 129.5, 129.3, 128.2, 117.3, 114.5, 50.1, 41.9, 32.4, 32.4, 29.7, 26.8; HRMS (ESI-TOF): m/z calcd for $C_{29}H_{30}ClFNO_2^+$ [$(M+H)^+$], 478.1944; found, 478.1943.

10-(4-fluorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3b)

White solid; Mp 298–299 °C; IR (KBr): 2959, 1641, 1510, 1402, 1220, 700 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.43 (d, J = 8.0 Hz, 2H, ArH), 7.24–7.30 (m, 6H, ArH), 7.10–7.13 (m, 1H, ArH), 5.29 (s, 1H, CH), 2.21 (AB, J = 16.0 Hz, 2H, CH_2), 2.14 (AB, J = 16.0 Hz, 2H, CH_2), 2.08 (AB, J = 16.0 Hz, 2H, CH_2), 1.83 (AB, J = 16.0 Hz, 2H, CH_2), 0.97 (s, 6H, 2×CH_3), 0.82 (s,

6H, 3×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 162.5 (d, $^1J_{C–F}$ = 249.0 Hz), 149.5, 146.0, 135.1, 135.0, 131.8, 128.1, 127.8, 126.0, 117.2, 114.8, 50.2, 41.9, 32.6, 32.4, 29.8, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{31}FNO_2^+$ [(M+H)$^+$], 444.2333; found, 444.2332.

10-(3-fluorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3c)

White solid; Mp 274–275 °C; IR (KBr): 2959, 1642, 1608, 1371, 1222, 693 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.58 (q, *J* = 8.0 Hz, 1H, ArH), 7.43 (d, *J* = 8.0 Hz, 2H, ArH), 7.28–7.33 (m, 3H, ArH), 7.08–7.15 (m, 2H, ArH), 7.01 (d, *J* = 8.0 Hz, 1H, ArH), 5.29 (s, 1H, CH), 2.18–2.24 (m, 4H, CH_2), 2.15 (AB, *J* = 16.0 Hz, 2H, CH_2), 1.85 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 149.0, 145.9, 140.6, 140.5, 128.1, 127.8, 126.0, 117.0, 116.8 (d, $^2J_{C–F}$ = 20.0 Hz), 50.2, 41.7, 32.7, 32.5, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{31}FNO_2^+$[(M+H)$^+$], 444.2333; found, 444.2339.

10-(3-chlorophenyl)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3d)

White solid; Mp 276–277 °C; IR (KBr): 2954, 1634, 1400, 1359, 1225, 839 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.52–7.59 (m, 2H, ArH), 7.37 (d, *J* = 8.0 Hz, 2H, ArH), 7.22–7.29 (m, 3H, ArH), 7.17 (d, *J* = 8.0 Hz, 1H, ArH), 5.24 (s, 1H, CH), 2.22 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.15 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.09 (AB, *J* = 16.0 Hz, 2H, CH_2), 1.83 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 149.2, 144.5, 140.1, 131.6, 130.0, 129.3, 128.3, 114.5, 50.1, 41.8, 32.5, 32.4, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{30}FClNO_2^+$[(M+H)$^+$], 494.1648; found, 494.1648.

10-(3-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3e)

White solid; Mp 257–258 °C; IR (KBr): 2961, 1637, 1571, 1369, 1224, 693 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.55 (t, *J* = 4.0 Hz, 2H, ArH), 7.42 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.29 (m, 2H, ArH), 7.18–7.21 (m, 1H, ArH), 7.12–7.14 (m, 1H, ArH), 5.29 (s, 1H, CH), 2.07–2.24 (m, 6H, CH_2), 1.84 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3);

^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 149.0, 145.9, 140.3, 129.9, 128.1, 127.8, 126.0, 114.9, 50.2, 41.8, 32.7, 32.5, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{31}ClNO_2^+$ [(M+H)$^+$], 460.2038; found, 460.2038.

10-(4-bromophenyl)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3f)

White solid; Mp 287–290 °C; IR (KBr): 1658, 1636, 1610, 1398, 1225, 845 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.73 (d, *J* = 12.0 Hz, 2H, ArH), 7.36 (d, *J* = 8.0 Hz, 2H, ArH), 7.23 (d, *J* = 8.0 Hz, 2H, ArH), 7.13 (d, *J* = 8.0 Hz, 2H, ArH), 5.24 (s, 1H, CH), 2.05–2.24 (m, 6H, CH_2), 1.80–1.85 (m, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 149.3, 144.5, 138.0, 131.6, 129.3, 128.3, 123.6, 114.5, 50.1, 41.9, 32.5, 32.4, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{30}BrClNO_2^+$ [(M+H)$^+$], 538.1143; found, 538.1445.

10-(4-bromophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3g)

White solid; Mp >300 °C; IR (KBr): 2955, 1632, 1593, 1363, 1224, 708 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.72 (d, *J* = 8.0 Hz, 2H, ArH), 7.42 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.29 (m, 2H, ArH), 7.12–7.16 (m, 3H, ArH), 5.29 (s, 1H, CH), 2.13-2.24 (m, 4H, CH_2), 2.08 (AB, *J* = 16.0 Hz, 2H, CH_2), 1.83 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 149.1, 145.9, 138.2, 133.4, 128.1, 127.8, 126.0, 123.5, 114.9, 50.2, 41.9, 32.6, 32.5, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{31}BrNO_2^+$ [(M+H)$^+$], 504.1533; found, 504.1535.

10-(3-bromophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3h)

White solid; Mp 262–264 °C; IR (KBr): 2955, 1642, 2510, 1361, 1222, 851 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ =7.70–7.73 (m, 1H, ArH), 7.41–7.50 (m, 4H, ArH), 7.22–7.28 (m, 3H, ArH), 7.10–7.14 (m, 1H, ArH), 5.28 (s, 1H, CH), 2.07–2.24 (m, 6H, CH_2), 1.83 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.98 (s, 6H, 2×CH_3), 0.83 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 149.1, 145.9, 140.4, 132.8, 128.1, 127.8, 126.1, 114.9, 77.4, 77.1, 76.8, 50.2, 41.8,

32.7, 32.5, 29.7, 26.9; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{31}BrNO_2^+$ [(M+H)$^+$], 504.1533; found, 504.1533.

9-(4-chlorophenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3i)

White solid; Mp 239–241 °C; IR (KBr): 2955, 1649, 1583, 1336, 1224, 736 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.57–7.60 (m, 3H, ArH), 7.39 (d, *J* = 8.4 Hz, 2H, ArH), 7.22–7.25 (m, 4H, ArH), 5.26 (s, 1H, CH), 2.07–2.24 (m, 6H, CH_2), 1.80–1.85 (m, 2H, CH_2), 0.96(s, 6H, 2×CH_3), 0.82 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 149.9, 144.8, 138.9, 131.5, 129.5, 129.3, 128.2, 50.1, 41.8, 32.4, 32.4, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for : $C_{29}H_{31}ClNO_2^+$ [(M+H)$^+$], 460.2038; found, 460.2040.

3,3,6,6-tetramethyl-9,10-diphenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3j)

White solid; Mp 255–257 °C; IR (KBr): 2957, 1642, 1593, 1365, 1220, 708 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ =7.56–7.61 (m, 3H, ArH), 7.46 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.29 (m, 4H, ArH), 7.11 (t, *J* = 8.0 Hz, 1H, ArH), 5.31 (s, 1H, CH), 2.22 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.14 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.10 (AB, *J* = 16.0 Hz, 2H, CH_2), 1.84 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.96 (s, 6H, 2×CH_3), 0.81 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 149.7, 146.2, 139.1, 130.0, 129.4, 128.1, 127.9, 125.9, 114.6, 50.2, 41.8, 32.4, 29.7, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{29}H_{32}NO_2^+$ [(M+H)$^+$], 426.2428; found, 426.2427.

9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-10-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3k)

White solid; Mp 234–235 °C; IR (KBr): 2957, 1642, 1577, 1369, 1224, 700 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.54–7.60 (m, 3H, ArH), 7.37 (t, *J* = 8.0 Hz, 2H, ArH), 7.25 (t, *J* = 8.0 Hz, 2H, ArH), 6.80 (t, *J* = 8.0 Hz, 2H, ArH), 5.23 (s, 1H, CH), 3.75 (m, 3H, OCH_3), 2.20 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.13 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.08 (AB, *J* = 16.0 Hz, 2H, CH_2), 1.82 (AB, *J* = 16.0 Hz, 2H, CH_2), 0.94 (s, 6H, 2×CH_3), 0.81 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 157.7, 149.5, 139.1, 138.8, 130.1, 129.4, 128.8, 114.8, 113.5, 55.1, 50.2, 41.8, 32.4, 31.9, 29.8, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{30}H_{34}NO_3^+$ [(M+H)$^+$],

456.2553; found, 456.2532.

3,3,6,6-tetramethyl-9-phenyl-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3l)

White solid; Mp 233–234 °C; IR (KBr): 2961, 1644, 1573, 1361, 1224, 698 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.46 (d, *J* = 8.0 Hz, 1H, ArH), 7.35 (d, *J* = 8.0 Hz, 1H, ArH), 7.26 (d, *J* = 8.0 Hz, 2H, ArH), 7.10–7.14 (m, 1H, ArH), 7.04–7.08 (m, 2H, ArH), 5.30 (s, 1H, CH), 2.50 (s, 3H, CH_3), 2.07–2.24 (m, 6H, CH_2), 1.83–1.87 (m, 2H, CH_2), 0.96 (s, 6H, 2×CH_3), 0.82 (s, 6H, 2×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 149.8, 146.2, 139.0, 130.1, 128.0, 127.9, 125.9, 114.5, 50.3, 41.8, 32.7, 32.4, 29.7, 26.8, 21.5; HRMS (ESI-TOF): *m*/*z* calcd for $C_{30}H_{34}NO_2^+$ [$(M+H)^+$], 440.2584; found, 440.2582.

10-butyl-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3m)

White solid; Mp 210–213 °C; IR (KBr): 2961, 1649, 1628, 1369, 1238, 1122 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ =7.17–7.20 (m, 2H, ArH), 7.11–7.14 (m, 2H, ArH), 5.22 (s, 1H, CH), 3.63–3.72 (m, 2H, CH_2), 2.54 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.40 (AB, *J* = 16.0 Hz, 2H, CH_2), 2.18–2.26 (m, 4H, CH_2), 1.57–1.61 (m, 2H, CH_2), 1.35–1.41 (m, 2H, CH_2), 1.09 (s, 6H, 2×CH_3), 0.99 (s, 9H, 3×CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.5, 150.4, 144.4, 131.3, 129.0, 128.0, 115.0, 115.0, 49.9, 44.6, 40.4, 33.5, 32.5, 31.6, 29.3, 27.8, 19.9, 13.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{27}H_{35}ClNO_2^+$ [$(M+H)^+$], 440.2351; found, 440.2351.

9,10-bis(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3n)

White solid; Mp 267–268 °C; IR (KBr): 2944, 1634, 1495, 1363, 1234, 728 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ =7.54 (d, *J* = 8.0 Hz, 2H, ArH), 7.33 (d, *J* = 8.0 Hz, 2H, ArH), 7.20–7.29 (m, 4H, ArH), 5.34 (s, 1H, CH), 2.33–2.40 (m, 2H, CH_2), 2.15–2.29 (m, 4H, CH_2), 2.02–2.08 (m, 2H, CH_2), 1.87–1.94 (m, 2H, CH_2), 1.75–1.84 (m, 2H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 151.3, 144.9, 137.4, 135.6, 131.7, 130.3, 129.1, 128.3, 115.4, 36.7, 31.8, 28.3, 21.1; HRMS (ESI-TOF): *m*/*z* calcd for $C_{25}H_{22}Cl_2NO_2^+$ [$(M+H)^+$], 438.1022; found, 438.1022.

10-(3-chlorophenyl)-9-(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-

dione（3o）

White solid; Mp >300 °C; IR (KBr): 1636, 1589, 1402, 1233, 1180, 957 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.48–7.54 (m, 2H, ArH), 7.32 (d, *J* = 8.0 Hz, 2H, ArH), 7.19–7.30 (m, 3H, ArH), 5.32 (s, 1H, CH), 2.32–2.39 (m, 2H, CH_2), 2.17–2.28 (m, 4H, CH_2), 2.01–2.08 (m, 2H, CH_2), 1.87–1.94 (m, 2H, CH_2), 1.74–1.83 (m, 2H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.0, 151.2, 144.9, 140.0, 131.6, 129.9, 128.3, 115.4, 77.4, 77.1, 76.8, 36.7, 31.8, 28.3, 21.1; HRMS (ESI-TOF): *m*/*z* calcd for $C_{25}H_{22}Cl_2NO_2^+$ [(M+H)$^+$], 438.1022; found, 438.1001.

10-(3-bromophenyl)-9-(4-methoxyphenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3p)

White solid; Mp >300 °C; IR (KBr): 2951, 1632, 1512, 1363, 1234, 697 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.68 (t, *J* = 8.0 Hz, 1H, ArH), 7.45 (t, *J* = 8.0 Hz, 2H, ArH), 7.32 (d, *J* = 8.0 Hz, 1H, ArH), 7.25–7.29 (m, 1H, ArH), 6.82 (d, *J* = 8.0 Hz, 2H, ArH), 5.32 (s, 1H, CH), 3.77 (m, 3H, OCH_3), 2.20–2.41 (m, 6H, CH_2), 2.02–2.08 (m, 2H, CH_2), 1.78–1.94 (m, 4H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.0, 157.9, 150.6, 140.4, 138.8, 132.7, 128.7, 116.1, 113.7, 55.2, 36.8, 31.2, 28.3, 21.2; HRMS (ESI-TOF): *m*/*z* calcd for $C_{30}H_{24}BrNNaO_3^+$ [(M+H)$^+$], 500.0832; found, 500.0826.

9-(4-chlorophenyl)-10-(p-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3q)

White solid; Mp 275–278 °C; IR (KBr): 2962, 1630, 1569, 1363, 1223, 835 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ =7.33–7.37 (m, 4H, ArH), 7.22 (d, *J* = 8.0 Hz, 2H, ArH), 7.13–7.17 (m, 2H, ArH), 5.34 (s, 1H, CH), 2.47 (s, 3H, CH_3), 2.16–2.38 (m, 6H, CH_2), 2.03–2.10 (m, 6H, CH_2), 1.86–1.92 (m, 2H, CH_2), 1.75–1.79 (m, 2H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.1, 152.1, 145.2, 139.7, 136.2, 131.5, 129.2, 128.2, 115.1, 36.8, 31.8, 28.3, 21.2, 21.1; HRMS (ESI-TOF): *m*/*z* calcd for $C_{26}H_{25}ClNO_2^+$ [(M+H)$^+$], 418.1568; found, 417.1568.

10-(4-fluorophenyl)-3,3,6,6-tetramethyl-9-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (**3r**)

White solid; Mp 260–263 °C; IR (KBr): 2957, 1634, 1510, 1385, 1224, 851 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$): δ = 7.23 (t, *J* = 10.0 Hz, 2H, ArH), 7.14–7.18 (m, 2H, ArH), 4.23 (d, *J* =

5.0 Hz, 1H, CH), 2.05–2.22 (m, 4H, 2×CH_2), 2.03 (AB, *J* = 15.0 Hz, 2H, CH_2), 1.73 (AB, *J* = 15.0 Hz, 2H, CH_2), 1.41–1.46 (m, 2H, CH_2), 1.26–1.32 (m, 2H, CH_2), 0.97 (s, 6H, 2×CH_3), 0.95 (s, 6H, 2×CH_3), 0.88 (t, *J* = 5.0 Hz, 3H, CH_3); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 196.2, 162.4 (d, $^1J_{C-F}$ = 248.8 Hz), 161.4, 150.7, 135.2, 131.4, 117.1, 114.3, 77.3, 77.1, 76.8, 50.4, 41.8, 38.1, 32.2, 30.0, 26.6, 26.0, 18.8, 14.4; HRMS (ESI-TOF): *m*/*z* calcd for $C_{26}H_{33}FN_2O^+$ [$(M+H)^+$], 410.2409; found, 410.2491.

3. Synthesis and Spectral Data of Compounds 4

General procedure for the synthesis of compounds 4

A mixture of enaminones **1** (1.0 mmol), aldehydes **2** (0.6mmol), p-TSA (0.3mmol), and CH_3CN (10 mL) was stirred at reflux for 2 h. After the desired product was formed as indicated by TLC, the reaction mixture was quenched with saturated NH_4Cl solution (2 mL) and extracted with ethyl acetate (40 mL). The organic phase were dried over Mg_2SO_4, and concentrated under vacuum. The residue was purified by flash chromatography (petroleum ether/ethyl acetate = 1:1) giving a yellow solid **4**.

Spectroscopic data of 1,4-dihydropyridines 4

(*E*)-9-(4-chlorophenyl)-10-(4-fluorophenyl)-8-((4-fluorophenyl)imino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4a)

Yellow solid; Mp 260–262 °C; IR (KBr): 2959, 1649, 1500, 1371, 1226, 846cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.41 (t, *J* = 8.0 Hz, 2H, ArH), 7.17–7.27 (m, 6H, ArH), 6.87–6.93 (m, 2H, ArH), 6.41–6.47 (m, 2H, ArH), 5.54 (s, 1H, CH), 5.25 (d, *J* = 32.4 Hz, 1H, CH), 1.92–2.24 (m, 6H, 3×CH_2), 1.67–1.83 (m, 2H, CH_2), 0.95 (s, 3H, CH_3), 0.85 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.78 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 162.4 (d, $^1J_{C–F}$ = 245.0 Hz), 158.8 (d, $^1J_{C–F}$ = 238.5 Hz), 150.3, 148.0, 145.4, 142.0, 145.4, 142.0, 135.4, 131.4, 131.1, 129.6, 127.8, 120.7 (d, $^3J_{C–F}$ = 7.5 Hz), 120.6 (d, $^3J_{C–F}$ = 7.5 Hz), 117.0 (d, $^2J_{C–F}$ = 21.5 Hz), 115.4(d, $^2J_{C–F}$ = 21.9 Hz), 115.1(d, $^2J_{C–F}$ = 21.9 Hz), 114.6, 113.1, 50.2, 41.9, 40.8, 33.2, 32.4, 31.4, 29.9, 29.6, 26.7; HRMS (ESI-TOF): *m/z* calcd for $C_{35}H_{34}ClF_2N_2O^+$ [$(M+H)^+$], 571.2322; found, 571.2323.

(*E*)-10-(4-fluorophenyl)-8-((4-fluorophenyl)imino)-3,3,6,6-tetramethyl-9-phenyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4b)

Yellow solid; Mp 233–234 °C; IR (KBr): 2957, 1644, 1579, 1367, 1267, 853 cm^{-1}; ^{1}H NMR

(400 MHz, $CDCl_3$): δ = 7.51 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.28 (m, 6H, ArH), 7.15–7.11 (m, 1H, ArH), 6.91–6.95 (m, 2H, ArH), 6.48 (d, *J* = 8.0 Hz, 2H, ArH), 5.63 (s, 1H, CH), 2.01–2.27 (m, 6H, 3×CH_2), 1.81–1.87 (m, 2H, CH_2), 0.99 (s, 3H, CH_3), 0.88 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.74 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.3 (d, $^1J_{C–F}$ = 249.0 Hz), 158.8 (d, $^1J_{C–F}$ = 238.0 Hz), 150.19, 148.21, 146.69, 141.85, 135.57, 135.53, 131.59, 128.18, 127.69, 125.57, 120.72 (d, $^3J_{C–F}$ = 8.0 Hz), 116.9 (d, $^2J_{C–F}$ = 21.3 Hz), 115.3 (d, $^2J_{C–F}$ = 22.0 Hz), 115.1 (d, $^2J_{C–F}$ = 22.0 Hz), 114.9, 113.4, 50.3, 41.9, 41.8, 40.8, 33.5, 32.4, 31.5, 30.0, 29.6, 26.8, 26.69; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{35}F_2N_2O^+$ [$(M+H)^+$], 537.2712; found,537.2713.

(*E*)-9,10-bis(4-chlorophenyl)-8-((4-chlorophenyl)imino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4c)

Yellow solid; Mp 260–262 °C; IR (KBr): 2959, 1647, 1489, 1369, 1222, 837 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.52 (d, *J* = 8.0 Hz, 2H, ArH), 7.40 (d, *J* = 8.0 Hz, 2H, ArH), 7.19 (t, *J* = 8.0 Hz, 6H, ArH), 6.42 (d, *J* = 8.0 Hz, 1H, ArH), 5.53 (s, H, CH), 1.93–2.24 (m, 6H, 3×CH_2), 1.68–1.84 (m, 2H, CH_2), 0.97 (s, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.81 (s, 3H, CH_3), 0.71 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 163.4, 150.5, 149.9, 145.3, 141.9, 137.9, 135.2, 131.1, 130.2, 129.6, 128.7, 127.8, 127.5, 120.8, 114.6, 113.2, 50.2, 41.9, 41.8, 40.8, 33.3, 32.4, 31.5, 29.9, 29.5, 26.7 ; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{34}Cl_3N_2O^+$ [$(M+H)^+$], 603.1731; found, 603.1730.

(*E*)-10-(4-chlorophenyl)-8-((4-chlorophenyl)imino)-3,3,6,6-tetramethyl-9-phenyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4d)

Yellow solid; Mp 259–262 °C; IR (KBr): 2955, 1639, 1583, 1368, 1224, 697 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.47–7.56 (m, 5H, ArH), 7.14–7.29 (m, 6H, ArH), 6.47 (d, *J* = 8.0 Hz, 2H, ArH), 5.62 (s, 1H, CH), 1.96–2.29 (m, 6H, 3×CH_2), 1.73–1.88 (m, 2H, CH_2), 1.00 (s, 3H, CH_3), 0.89 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.74 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.2, 163.6, 150.7, 150.2, 146.5, 141.9, 138.1, 135.1, 133.6, 131.2, 130.2, 129.6,

128.6, 128.5, 128.2, 127.7, 127.4, 125.7, 120.9, 115.0, 113.5, 50.2, 41.9, 41.8, 40.9, 33.5, 32.4, 31.5, 29.9, 29.5, 26.8, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{35}Cl_2N_2O$ [(M+H)$^+$], 569.2121; found, 569.2121.

(*E*)-10-(3-chlorophenyl)-9-(4-chlorophenyl)-8-((3-chlorophenyl)imino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4e)

Yellow solid; Mp 274–277 °C; IR (KBr): 2957, 1640, 1508, 1369, 1261, 706 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.51 (d, *J* = 8.0 Hz, 2H, ArH), 7.42 (d, *J* = 8.0 Hz, 2H, ArH), 7.23–7.29 (m, 3H, ArH), 7.16 (t, *J* = 8.0 Hz, 2H, ArH), 6.96 (d, *J* = 8.0 Hz, 1H, ArH), 6.53 (s, 1H, ArH), 6.40 (d, *J* = 8.0 Hz, 1H, ArH), 5.53 (s, 1H, CH), 1.97–2.27 (m, 6H, 3×CH_2), 1.84 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.74 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.00 (s, 3H, CH_3), 0.89 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.75 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.6, 153.3, 149.8, 145.1, 142.1, 140.5, 135.6, 134.2, 131.2, 130.9, 130.1, 129.8, 129.7, 129.6, 129.5, 127.9, 122.3, 119.4, 117.8, 114.5, 113.3, 50.2, 41.9, 41.8. 41.0, 33.3, 32.5, 31.6, 29.9, 29.5, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{34}Cl_3N_2O^+$ [(M+H)$^+$], 603.1731; found, 603.1729.

(*E*)-10-(3-chlorophenyl)-8-((3-chlorophenyl)imino)-3,3,6,6-tetramethyl-9-phenyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4f)

Yellow solid; Mp 209–211 °C; IR (KBr): 2957, 1644, 1584, 1371, 1230, 695 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.48–7.54 (m, 4H, ArH), 7.26–7.30 (m, 2H, ArH), 7.12–7.21 (m, 3H, ArH), 6.95 (d, *J* = 8.0 Hz, 1H, ArH), 6.54 (s, 1H, ArH), 6.41 (d, *J* = 8.0 Hz, 1H, ArH), 5.58 (s, 1H, CH), 1.73–2.26 (m, 8H, 4×CH_2), 1.00 (s, 3H, CH_3), 0.90 (s, 3H, CH_3), 0.84 (s, 3H, CH_3), 0.75 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.7, 153.5, 149.6, 146.4, 141.9, 140.7, 134.2, 129.7, 129.6, 128.4, 128.2, 128.1, 127.8, 125.7, 122.1, 119.5, 117.8, 114.9, 113.6, 50.3, 41.9, 41.8, 41.0, 33.5, 32.5, 31.6, 29.9, 29.5, 26.8, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{35}Cl_2N_2O^+$ [(M+H)$^+$], 569.2121; found, 569.2122.

(*E*)-10-(4-bromophenyl)-8-((4-bromophenyl)imino)-3,3,6,6-tetramethyl-9-phenyl-

3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4g)

Yellow solid; Mp 163–166 °C; IR (KBr): 2955, 1639, 1581, 1369, 1218, 696 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.71 (d, *J* = 8.0 Hz, 2H, ArH), 7.48 (d, *J* = 8.0 Hz, 2H, ArH), 7.32 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.29 (m, 3H, ArH), 7.13–7.17 (m, 3H, ArH), 6.42 (d, *J* = 8.0 Hz, 2H, ArH), 5.59 (s, 1H, CH), 1.95–2.27 (m, 6H, 3×CH_2), 1.72–1.99 (m, 2H, CH_2), 0.99 (s, 3H, CH_3), 0.88 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.73 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.7, 163.5, 151.2, 149.8, 146.5, 141.8, 138.6, 133.2, 131.6, 128.2, 127.7, 125.6, 123.1, 121.4, 115.0, 114.9, 113.6, 50.3, 41.9, 41.8, 40.9, 33.5, 32.4, 31.5, 29.9, 29.5, 26.8, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{35}Br_2N_2O^+$ [$(M+H)^+$], 657.1111; found, 657.1111.

(*E*)-10-(3-bromophenyl)-8-((3-bromophenyl)imino)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4h)

Yellow solid; Mp 164–167 °C; IR (KBr): 2961, 1644, 1580, 1404, 1230, 838 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.68–7.71 (m, 1H, ArH), 7.40–7.48 (m, 4H, ArH), 7.21–7.25 (m, 3H, ArH), 7.10 (d, *J* = 4.0 Hz, 2H, ArH), 6.69 (s, 1H, ArH), 6.43–6.45 (m, 1H, ArH), 5.53 (s, 1H, CH), 1.97–2.27 (m, 6H, 3×CH_2), 1.84 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.74 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.00 (s, 3H, CH_3), 0.89 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.75 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 163.6, 153.5, 149.8, 145.1, 142.1, 140.7, 132.6, 131.2, 130.1, 129.6, 127.9, 125.2, 122.4, 122.3, 118.3, 114.5, 113.3, 50.2, 41.9, 41.8, 41.0, 33.3, 32.5, 31.6, 30.0, 29.5, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{34}Br_2ClN_2O^+$ [$(M+H)^+$], 691.0721; found, 691.0721.

(*E*)-10-(3-bromophenyl)-8-((3-bromophenyl)imino)-3,3,6,6-tetramethyl-9-phenyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4i)

Yellow solid; Mp 124–126 °C; IR (KBr): 2959, 1640, 1583, 1369, 1230, 694 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.69 (d, *J* = 8.0 Hz, 1H, ArH), 7.44–7.49 (m, 4H, ArH), 7.23–7.29 (m, 3H, ArH), 7.09–7.17 (m, 3H, ArH), 6.70 (s, 1H, ArH), 6.44–6.47 (m, 1H, ArH), 5.57 (s, 1H, CH), 1.97–2.27 (m, 6H, 3×CH_2), 1.84 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.75 (AB, *J* = 16.0 Hz, 1H,

CH_2), 1.00 (s, 3H, CH_3), 0.90 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.76 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.7, 153.7, 149.7, 146.4, 141.9, 140.9, 132.5, 130.0, 128.2, 127.8, 125.7, 125.1, 122.4, 122.3, 118.3, 114.9, 113.7, 50.3, 41.9, 41.8, 41.0, 33.5, 32.5, 31.6, 30.0, 29.5, 26.8, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{35}Br_2ClN_2O^+$ [$(M+H)^+$], 657.1111; found, 657.1112.

(*E*)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-10-phenyl-8-(phenylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4j)

White solid; Mp 252–254 °C; IR (KBr): 2957, 1636, 1579, 1373, 1216, 695 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.53–7.57 (m, 3H, ArH), 7.49 (d, *J* = 8.0 Hz, 2H, ArH), 7.23–7.30 (m, 6H, ArH), 6.97–7.01 (m, 1H, ArH), 6.54 (d, *J* = 8.0 Hz, 2H, ArH), 5.62 (s, 1H, CH), 1.97–2.27 (m, 6H, 3×CH_2), 1.71–1.88 (m, 2H, CH_2), 0.98 (s, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.72 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.9, 152.2, 150.6, 145.6, 142.0, 139.5, 130.9, 129.9, 129.8, 129.5, 129.1, 128.6, 127.7, 122.2, 119.5, 114.5, 112.7, 50.3, 41.9, 41.8, 40.8, 33.4, 32.4, 31.4, 29.9, 29.6, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{36}ClN_2O^+$ [$(M+H)^+$], 535.2511; found, 535.2510.

(*E*)-3,3,6,6-tetramethyl-9,10-diphenyl-8-(phenylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4k)

Yellow solid; Mp 230–232 °C; IR (KBr): 2955, 1639, 1589, 1367, 1227,700 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.52–7.60 (m, 5H, ArH), 7.23–7.30 (m, 6H, ArH), 7.14 (t, *J* = 8.0 Hz, 1H, ArH), 6.99 (t, *J* = 8.0 Hz, 1H, ArH), 6.57 (d, *J* = 8.0 Hz, 2H, ArH), 5.68 (s, 1H, CH), 1.99–2.29 (m, 6H, 3×CH_2), 1.73–1.89 (m, 2H, CH_2), 0.99 (s, 3H, CH_3), 0.87 (s, 3H, CH_3), 0.84 (s, 3H, CH_3), 0.74 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.0, 152.4, 150.5, 146.9, 141.9, 139.6, 129.9, 129.1, 128.6, 128.3, 127.6, 125.5, 122.1, 119.6, 114.9, 113.1, 50.4, 41.9, 41.8, 40.9, 33.6, 32.4, 31.4, 30.0, 29.5, 26.8, 26.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{37}N_2O^+$ [$(M+H)^+$], 501.2900; found, 501.2903.

(*E*)-9-(4-methoxyphenyl)-3,3,6,6-tetramethyl-10-phenyl-8-(phenylimino)-

3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4l)

Yellow solid; Mp 236–238 °C; IR (KBr): 2957, 1642, 1510, 1363, 1222, 706 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.52–7.58 (m, 3H, ArH), 7.46 (d, *J* = 8.0 Hz, 2H, ArH), 7.23–7.29 (m, 5H, ArH), 6.98 (t, *J* = 8.0 Hz, 1H, ArH), 6.82 (t, *J* = 8.0 Hz, 2H, ArH), 6.56 (d, *J* = 8.0 Hz, 1H, ArH), 5.60 (s, 1H, ArH), 3.80 (s, 3H, OCH_3), 1.96–2.27 (m, 6H, 3×CH_2), 1.85 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.72 (AB, *J* = 16.0 Hz, 1H, CH_2), 1.98 (s, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.84 (s, 3H, CH_3), 0.75 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.0, 157.4, 152.4, 150.2, 141.5, 139.7, 139.5, 129.8, 129.2, 128.9, 128.6, 122.0, 119.6, 115.1, 113.3, 113.0, 55.1, 50.4, 41.9, 41.8, 40.9, 32.7, 32.4, 31.4, 29.9, 29.5, 26.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{35}H_{39}N_2O_2^+$ [$(M+Na)^+$], 531.3006; found, 531.3007.

(*E*)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-10-(p-tolyl)-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4m)

Yellow solid; Mp 163–165 °C; IR (KBr): 2955, 1639, 1504, 1362, 1218, 736 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.47 (d, *J* = 8.0 Hz, 2H, ArH), 7.34 (d, *J* = 8.0 Hz, 2H, ArH), 7.22 (d, *J* = 8.0 Hz, 2H, ArH), 7.11 (d, *J* = 8.0 Hz, 2H, ArH), 7.06 (d, *J* = 8.0 Hz, 2H, ArH), 6.45 (d, *J* = 8.0 Hz, 2H, ArH), 5.60 (s, 1H, CH), 2.50 (s, 3H, CH_3), 2.32 (s, 3H, CH_3), 1.96–2.26 (m, 6H, 3×CH_2), 1.71–1.96 (m, 2H, CH_2), 0.98 (s, 3H, CH_3), 0.86 (s, 1H, CH_3), 0.83 (s, 3H, CH_3), 0.71 (s, 1H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.8, 150.8, 149.6, 145.7, 141.9, 139.1, 136.8, 131.4, 130.8, 130.5, 129.8, 129.2, 127.7, 119.5, 114.6, 112.6, 50.3, 41.8, 41.7, 40.7, 33.3, 32.4, 31.4, 29.9, 29.6, 26.7, 26.6, 21.3, 20.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{40}ClN_2O^+$ [$(M+H)^+$], 563.2824; found, 563.2823.

(*E*)-3,3,6,6-tetramethyl-9-phenyl-10-(p-tolyl)-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4n)

Yellow solid; Mp 224–226 °C; IR (KBr): 2957, 1646, 1577, 1367, 1216, 697 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.54 (d, *J* = 8.0 Hz, 2H, ArH), 7.34 (d, *J* = 8.0 Hz, 2H, ArH), 7.24–7.29 (m, 2H, ArH), 7.08–7.16 (m, 3H, ArH), 7.05 (d, *J* = 8.0 Hz, 2H, ArH), 6.46 (d, *J* = 8.0 Hz, 3H,

ArH), 5.85 (s, 1H, CH), 2.50 (s, 3H, $ArCH_3$), 2.31 (s, 3H, $ArCH_3$), 1.67–2.27 (m, 8H, 4×CH_2), 0.98 (s, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.72 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.8, 162.9, 150.7, 149.8, 147.0, 141.7, 138.9, 137.0, 131.2, 130.4, 129.1, 128.3, 127.6, 125.4, 119.5, 115.0, 113.0, 50.4, 41.9, 41.8, 40.8, 33.6, 32.3, 31.4, 30.0, 29.5, 26.8, 26.7, 21.3, 20.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{41}N_2O^+$ [$(M+H)^+$], 529.3213; found, 529.3218.

(*E*)-3,3,6,6-tetramethyl-9,10-diphenyl-8-(phenylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4o)

Yellow solid; Mp 134–136 °C; IR (KBr):2959, 1644, 1600, 1373, 1228, 779 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.42–7.50 (m, 3H, ArH), 7.33 (d, *J* = 8.0 Hz, 1H, ArH), 7.24 (d, *J* = 8.0 Hz, 2H, ArH), 7.13 (t, *J* = 8.0 Hz, 1H, ArH), 7.04 (d, *J* = 8.0 Hz, 2H, ArH), 6.81 (d, *J* = 8.0 Hz, 1H, ArH), 6.37 (s, 1H,ArH), 6.33 (d, *J* = 8.0 Hz, 1H, ArH), 5.60 (s, 1H, CH), 2.50 (s, 3H, CH_3), 2.31 (s, 3H, CH_3), 1.97–2.23 (m, 6H, 3×CH_2), 1.72–1.89 (m, 2H, CH_2), 0.98 (s, 3H, CH_3), 0.87 (s, 3H, CH_3), 0.83 (s, 3H, CH_3), 0.73 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.7, 152.2, 150.7, 145.7, 141.9, 139.4, 138.4, 130.9, 129.9, 129.7, 128.4, 127.7, 122.9, 120.2, 116.4, 114.4, 112.6, 50.3, 41.8, 41.7, 40.8, 33.3, 32.4, 31.4, 29.9, 29.6, 26.7, 21.5; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{40}ClN_2O$ [$(M+H)^+$], 563.2824; found, 563.3142.

(*E*)-3,3,6,6-tetramethyl-9-phenyl-10-(m-tolyl)-8-(m-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4p)

Yellow solid; Mp 230–233 °C; IR (KBr): 2955, 1642, 1583, 1371, 1228, 695 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.55 (d, *J* = 8.0 Hz, 2H, ArH), 7.44 (t, *J* = 8.0 Hz, 1H, ArH), 7.26–7.34 (m, 3H, ArH), 7.11–7.16 (m, 2H, ArH), 7.04–7.08 (m, 2H, ArH), 6.79 (d, *J* = 8.0 Hz, 1H, ArH), 6.39 (s, 1H, ArH), 6.34 (d, *J* = 8.0 Hz, 1H, ArH), 5.65 (s, 1H, CH), 2.50 (s, 3H, CH_3), 2.31 (s, 3H, CH_3), 1.97–2.24 (m, 6H, 3×CH_2), 1.73–1.90 (m, 2H, CH_2), 0.99 (s, 3H, CH_3), 0.87 (s, 1H, CH_3), 0.83 (s, 3H, CH_3), 0.74 (s, 1H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.8, 152.4, 147.0, 141.8, 139.6, 138.3, 129.8, 128.4, 127.6, 125.4, 122.8, 120.2,

116.5, 114.8, 113.0, 50.4, 41.8, 41.8, 40.9, 33.6, 32.4, 31.4, 29.9, 29.5, 26.8, 26.7, 21.5; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{41}N_2O^+$ [(M+H)$^+$], 529.3213; found, 529.3214.

(*E*)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-10-(o-tolyl)-8-(o-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4q)

Yellow solid; Mp 250–253 °C; IR (KBr): 2955, 1536, 1485, 1369, 1224, 734 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.40–7.50 (m, 6H, ArH), 7.20–7.24 (m, 3H, ArH), 7.05–7.10 (m, 2H, ArH), 6.90 (t, *J* = 8.0 Hz, 1H, ArH), 6.37 (d, *J* = 8.0 Hz, 1H, ArH), 5.59 (s, 1H, CH), 2.33 (d, *J* = 24.0 Hz, 3H, $ArCH_3$), 1.80–2.26 (m, 6H, $3CH_2$), 1.80-2.26 (m, 8H, $4\times CH_2$), 2.30 (s, 3H, $ArCH_3$), 1.60 (s, 3H, $ArCH_3$), 0.98 (s, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.82 (s, 3H, CH_3), 0.75 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 162.6, 151.0, 150.0, 146.3, 141.6, 138.6, 137.3, 131.6, 130.9, 130.1, 130.0, 129.5, 129.4, 127.8, 127.6, 127.5, 126.0, 122.2, 118.1, 114.3, 112.9, 50.4, 41.3, 40.7, 40.6, 34.2, 32.4, 31.4, 29.4, 29.2, 27.2, 17.8, 17.4; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{40}ClN_2O^+$ [(M+H)$^+$], 563.2824; found, 563.2828.

(*E*)-9-(4-chlorophenyl)-10-(4-methoxyphenyl)-8-((4-methoxyphenyl)imino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4r)

Yellow solid; Mp 186–189 °C; IR (KBr): 2957, 1638, 1606, 1355, 1251, 846 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.46 (d, *J* = 8.0 Hz, 2H, ArH), 7.22 (d, *J* = 8.0 Hz, 2H, ArH), 7.14 (d, *J* = 8.0 Hz, 2H, ArH), 7.04 (d, *J* = 8.0 Hz, 2H, ArH), 6.81 (d, *J* = 8.0 Hz, 2H, ArH), 6.49 (d, *J* = 8.0 Hz, 2H, ArH), 5.60 (s, 1H, CH), 3.92 (s, 3H, OCH_3), 3.79 (s, 3H, OCH_3), 1.97–2.25 (m, 6H, $3\times CH_2$), 1.73–1.90 (m, 2H, CH_2), 0.98 (s, 3H, CH_3), 0.87 (s, 3H, CH_3), 0.82 (s, 3H, CH_3), 0.71 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 195.9, 163.2, 159.6, 155.2, 151.1, 145.7, 145.4, 142.2, 132.0, 130.9, 130.8, 129.7, 129.5, 127.7, 120.7, 114.6, 114.0, 112.7, 55.6, 55.5, 50.3, 41.9, 41.8, 40.8, 33.3, 32.3, 31.4, 30.0, 29.7, 26.8, 26.6; HRMS (ESI-TOF): *m*/*z* calcd for $C_{37}H_{40}ClN_2O_3^+$ [(M+H)$^+$], 595.2722; found, 595.2723.

(*E*)-10-(3-chlorophenyl)-9-(4-chlorophenyl)-8-((3-chlorophenyl)imino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4r)

Yellow solid; Mp 108–110 °C; IR (KBr): 2948, 1641, 1585, 1367, 1238, 961 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.46–7.50 (m, 2H, ArH), 7.39 (d, *J* = 8.0 Hz, 2H, ArH), 7.28 (s, 1H, ArH), 7.23 (d, *J* = 8.0 Hz, 2H, ArH), 7.13–7.18 (m, 2H, ArH), 6.93–6.95 (m, 1H, ArH), 6.57 (s, 1H, ArH), 6.44 (d, *J* = 4.0 Hz, 1H, ArH), 5.63 (s, 1H, CH), 2.19–2.37 (m, 3H, CH_2), 2.03–2.12 (m, 3H, CH_2), 1.90–1.99 (m, 2H, CH_2), 1.74–1.81 (m, 2H, CH_2), 1.55–1.63 (m, 2H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.1, 164.2, 153.3, 151.5, 154.5, 143.9, 140.6, 134.2, 131.3, 129.7, 129.6, 129.5, 129.5, 128.0, 122.4, 119.7, 118.1, 115.5, 114.3, 36.8, 32.6, 28.4, 28.2, 28.0, 21.4, 21.0; HRMS (ESI-TOF): *m*/*z* calcd for $C_{31}H_{26}Cl_3N_2O^+$ [$(M+H)^+$], 547.1105; found, 547.1122.

(*E*)-9-(4-chlorophenyl)-10-phenyl-8-(phenylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4s)

Yellow solid; Mp 100–102 °C; IR (KBr):2918, 1635, 1590, 1383, 1088, 990 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.48–7.53 (m, 2H, ArH), 7.45 (d, *J* = 8.0 Hz, 2H, ArH), 7.20–7.27 (m, 6H, ArH), 6.95–6.98 (m, 1H, ArH), 6.57 (d, *J* = 4.0 Hz, 2H, ArH), 5.71 (s, 1H, CH), 2.19–2.34 (m, 3H, CH_2), 2.03–2.12 (m, 3H, CH_2), 1.86–1.97 (m, 2H, CH_2), 1.70–1.78 (m, 2H, CH_2), 1.54–1.61 (m, 2H, CH_2); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.2, 163.5, 152.2, 146.0, 143.8, 139.5, 131.1, 129.6, 129.1, 128.6, 127.9, 122.4, 119.8, 115.5, 113.8, 36.9, 32.7, 28.4, 28.2, 27.9, 21.5, 21.2; HRMS (ESI-TOF): *m*/*z* calcd for $C_{31}H_{28}ClN_2O^+$ [$(M+H)^+$], 479.1885; found, 479.1900.

(*E*)-9-(4-chlorophenyl)-10-(p-tolyl)-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4t)

Yellow solid; Mp 98–99 °C; IR (KBr):2946, 1628, 1504, 1402, 1365, 834 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.44 (d, *J* = 8.0 Hz, 2H, ArH), 7.29 (d, *J* = 4.0 Hz, 2H, ArH), 7.20 (d, *J* = 4.0 Hz, 2H, ArH), 7.11 (d, *J* = 8.0 Hz, 2H, ArH), 7.03 (d, *J* = 4.0 Hz, 2H, ArH), 6.48 (d, *J* = 8.0 Hz, 2H, ArH), 5.69 (s, 1H, CH), 2.44 (s, 3H, CH_3), 2.28 (s, 3H, CH_3), 2.18–2.38 (m, 5H, CH_2), 2.05–2.11 (m, 3H, CH_2), 1.86–1.98 (m, 2H, CH_2), 1.68–1.77 (m, 2H, CH_2); ^{13}C NMR

(100 MHz, $CDCl_3$): δ = 196.2, 163.5, 152.6, 149.4, 146.1, 143.8, 139.2, 136.8, 131.6, 131.0, 129.9, 129.8, 129.6, 129.2, 127.9, 119.8, 115.5, 113.7, 36.9, 32.7, 28.4, 28.2, 27.9, 21.5, 21.2, 20.8; HRMS (ESI-TOF): *m*/*z* calcd for $C_{33}H_{32}ClN_2O^+$ [$(M+H)^+$], 507.2198; found, 507.2215.

(*E*)-10-(4-fluorophenyl)-8-((4-fluorophenyl)imino)-3,3,6,6-tetramethyl-9-propyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4v)

Yellow solid; Mp 120–123 °C; IR (KBr): 2961, 1640, 1512, 1381, 1234, 853 cm^{-1}; ^1H NMR (500 MHz, $CDCl_3$): δ = 7.19 (t, *J* = 10.0 Hz, 1H, ArH), 7.11–7.17 (m, 2H, ArH), 6.96 (t, *J* = 8.0 Hz, 2H, ArH), 6.58–6.63 (m, 2H, ArH), 4.50 ((t, *J* = 5.0 Hz, 1H, CH), 1.24–2.28 (m, 12H, 6×CH_2), 0.98 (s, 3H, CH_3), 0.97 (s, 3H, CH_3), 0.91 (t, *J* = 5.0 Hz, 3H, CH_3), 0.86 (s, 3H, CH_3), 0.85 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.3, 163.6 (d, $^1J_{C–F}$ = 249.8 Hz), 158.8 (d, $^1J_{C–F}$ = 238.8 Hz), 151.2, 148.5, 148.4, 142.6, 135.7, 131.5, 127.0, 126.9, 126.7 (d, $^3J_{C–F}$ = 8.8 Hz), 120.8, 120.7, 116.7 (d, $^2J_{C–F}$ = 22.5 Hz), 116.0 (d, $^2J_{C–F}$ = 22.5 Hz), 115.8 (d, $^2J_{C–F}$ = 22.5 Hz), 115.3, 115.2, 114.7, 112.8, 50.5, 41.9, 41.8, 41.0, 38.0, 32.2, 31.3. 30.2, 29.9, 27.0, 26.6, 26.5, 19.2, 14.7; HRMS (ESI-TOF): *m*/*z* calcd for $C_{32}H_{37}F_2N_2O^+$ [$(M+H)^+$], 503.2868; found, 503.2865.

(*E*)-9-cyclohexyl-10-(4-fluorophenyl)-8-((4-fluorophenyl)imino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2*H*)-one (4w)

Yellow solid; Mp 257–259 °C; IR (KBr): 3412, 1642, 1616, 1383, 1244, 618 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.13–7.21 (m, 4H, ArH), 6.94–6.98 (m, 2H, ArH), 6.57–6.61(m, 2H, ArH), 4.54 (s, 1H, CH), 2.17–2.27 (m, 3H, CH_2), 2.05 (AB, *J* = 15.0 Hz, 1H, CH_2), 1.90–1.95 (m, 2H, CH_2), 1.72–1.82 (m, 5H, CH_2+CH), 1.58-1.66 (m, 4H, 2×CH_2), 1.06-1.21 (m, 3H, CH_2), 1.03 (s, 3H, CH_3), 0.98 (s, 3H, CH_3), 0.88 (s, 3H, CH_3), 0.86 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 196.6, 162.3 (d, $^1J_{C–F}$ = 248.8 Hz), 158.8 (d, $^1J_{C–F}$ = 238.8 Hz), 151.7, 148.6, 148.5, 143.0, 135.6, 131.4, 120.7, 120.6, 116.8, 116.6, 115.3, 115.1, 113.2, 111.0, 77.3, 77.0, 76.8, 50.6, 45.1, 42.1, 41.1, 31.9, 31.3, 31.2, 30.5, 27.0, 26.9, 26.8, 26.7, 26.6.

4. Copies of Original ^{1}H and ^{13}C NMR Spectra

^{1}H NMR and ^{13}C NMR spectra for compound 3

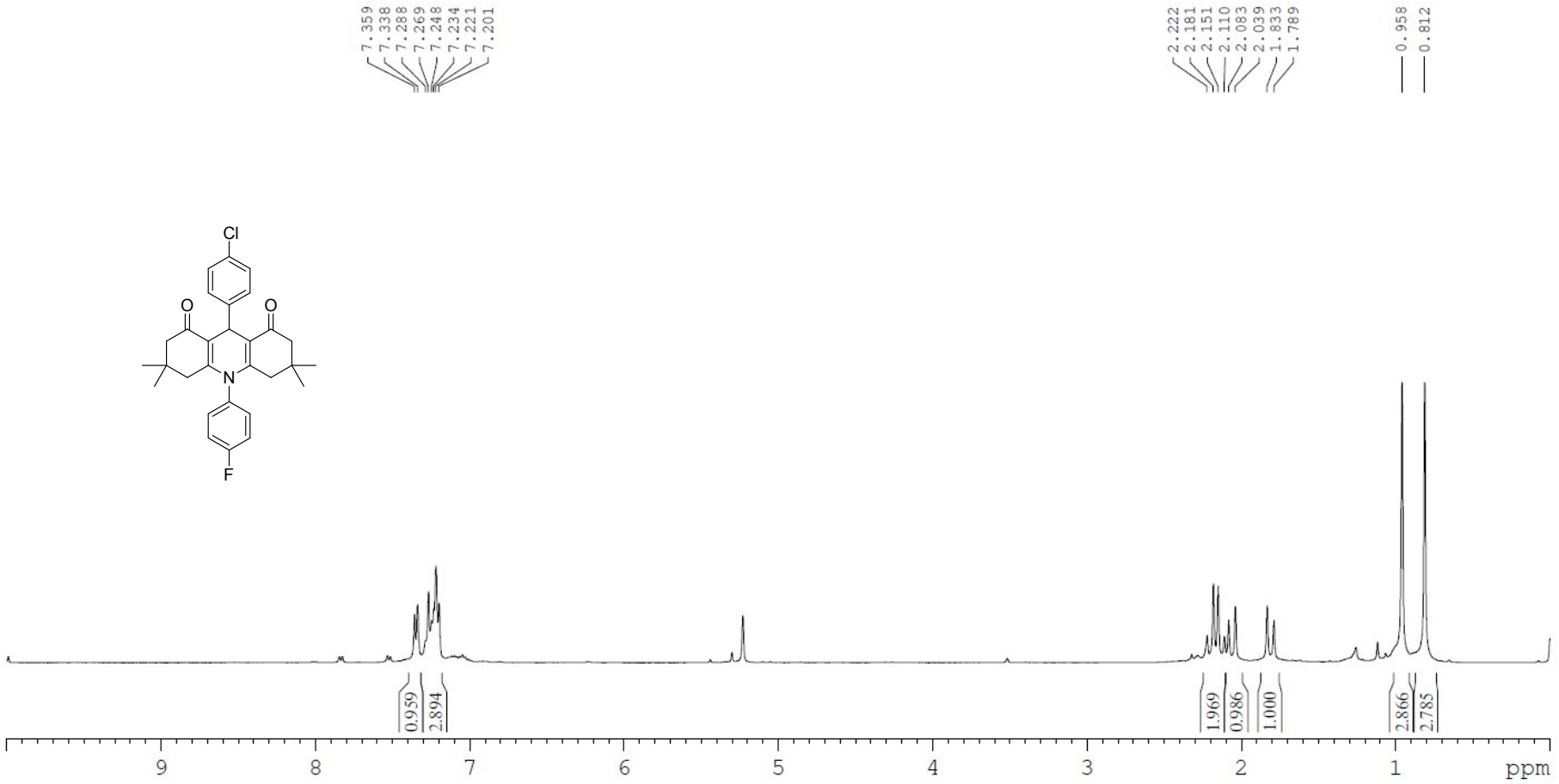


Figure 1. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3a**

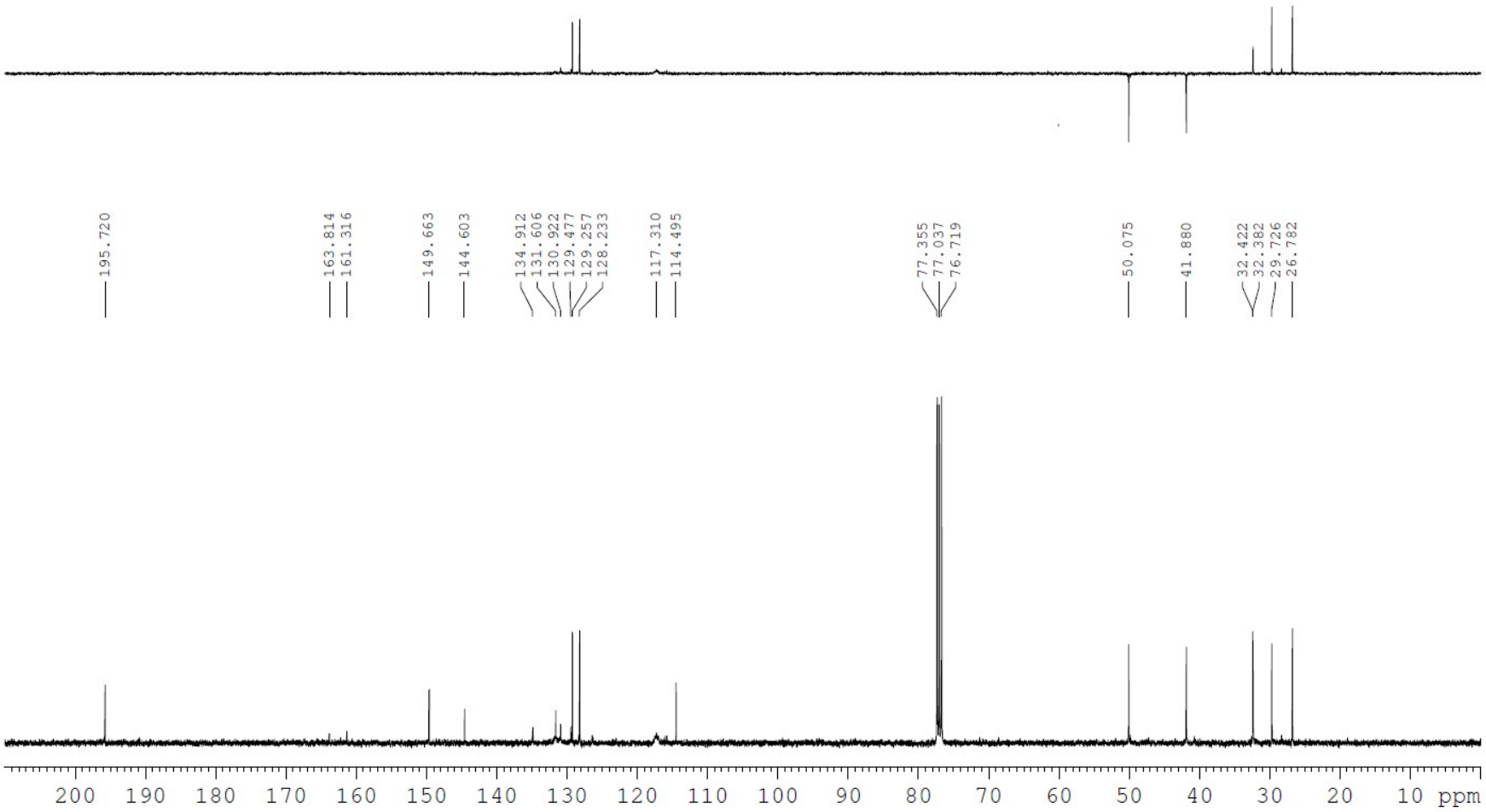


Figure 2. ^{13}C NMR (400 MHz, $CDCl_3$) spectra of compound **3a**

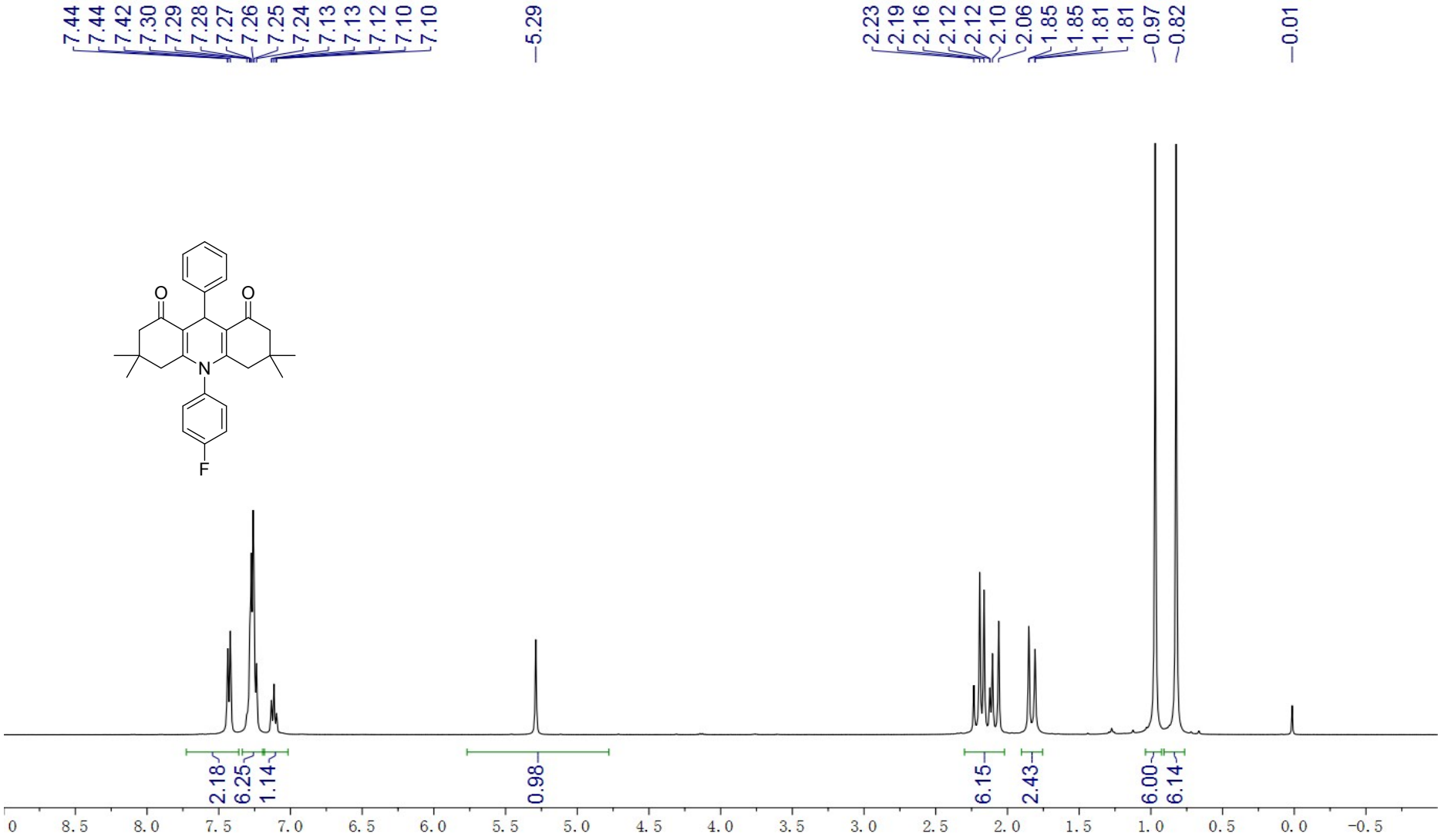


Figure 3. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3b**

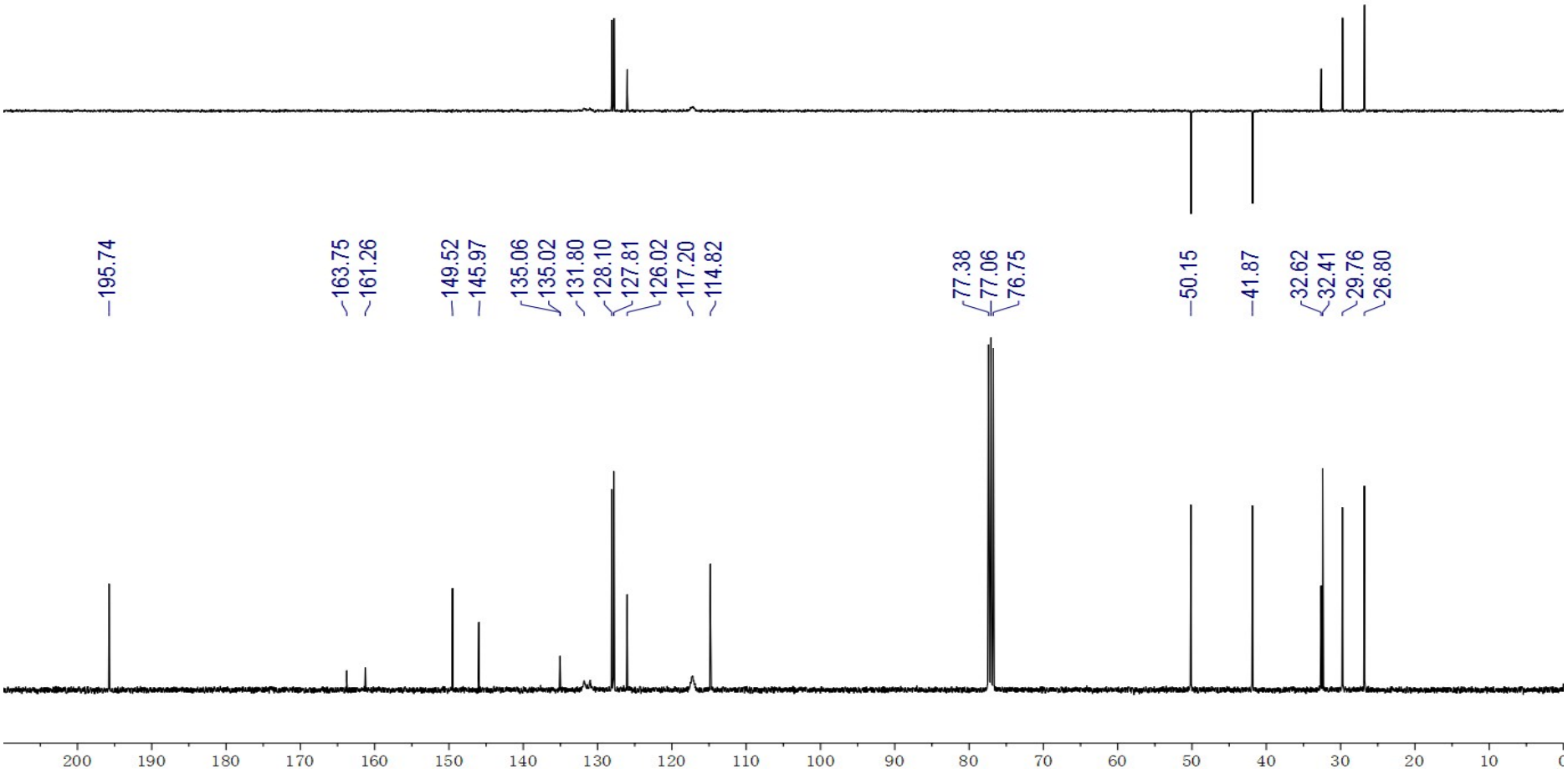


Figure 4. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3b**

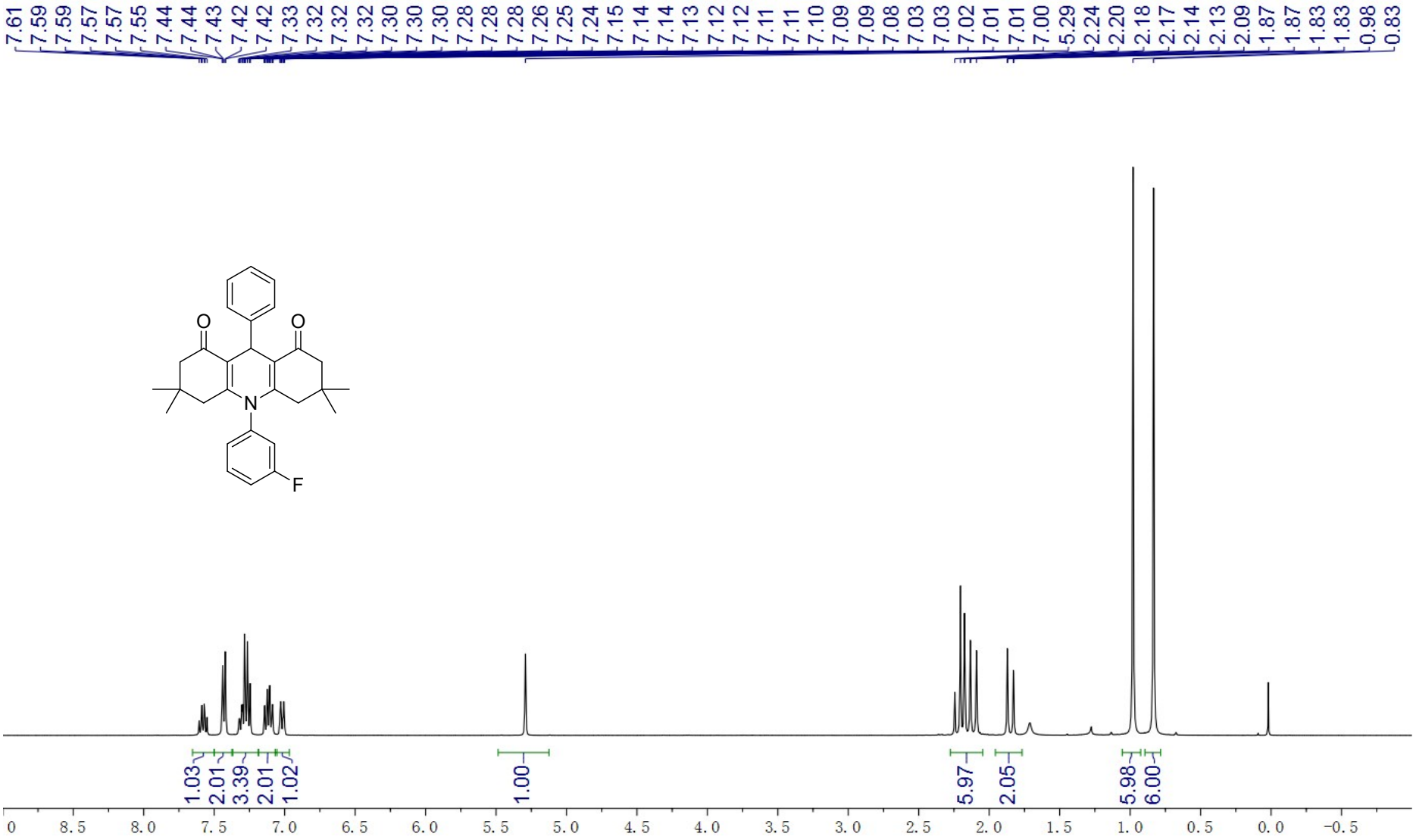


Figure 5. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3c**

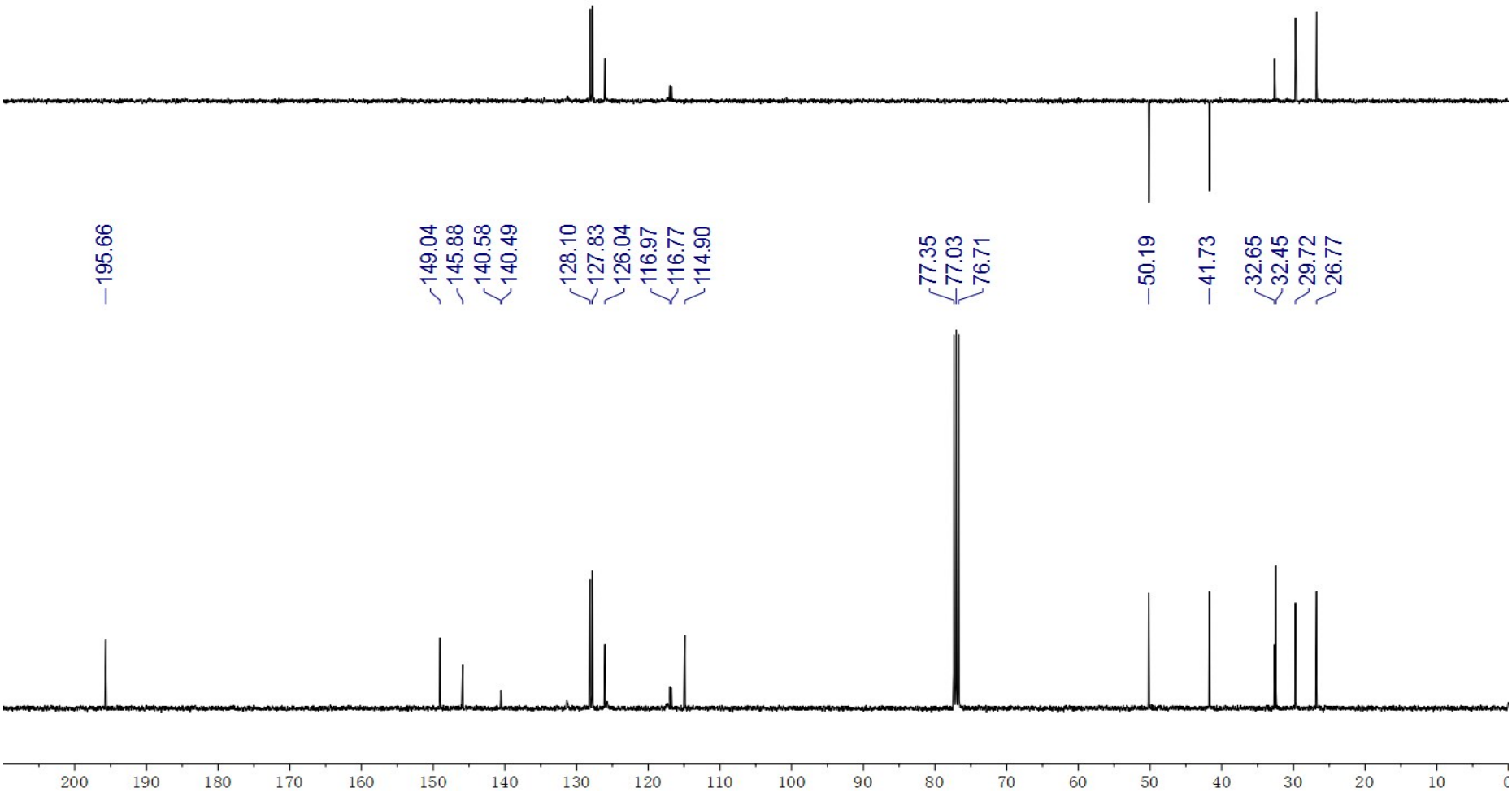


Figure 6. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3c**

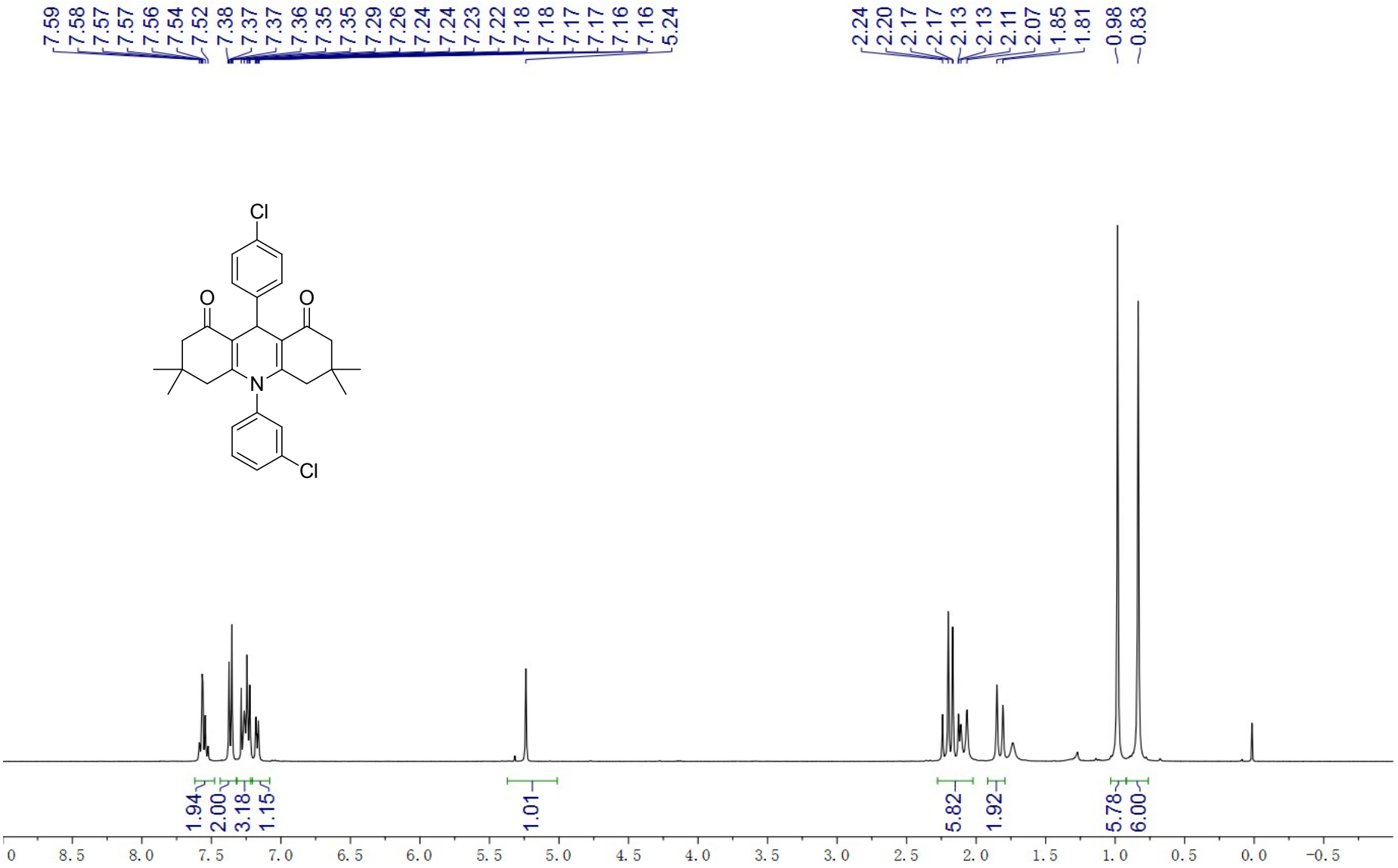


Figure 7. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3d**

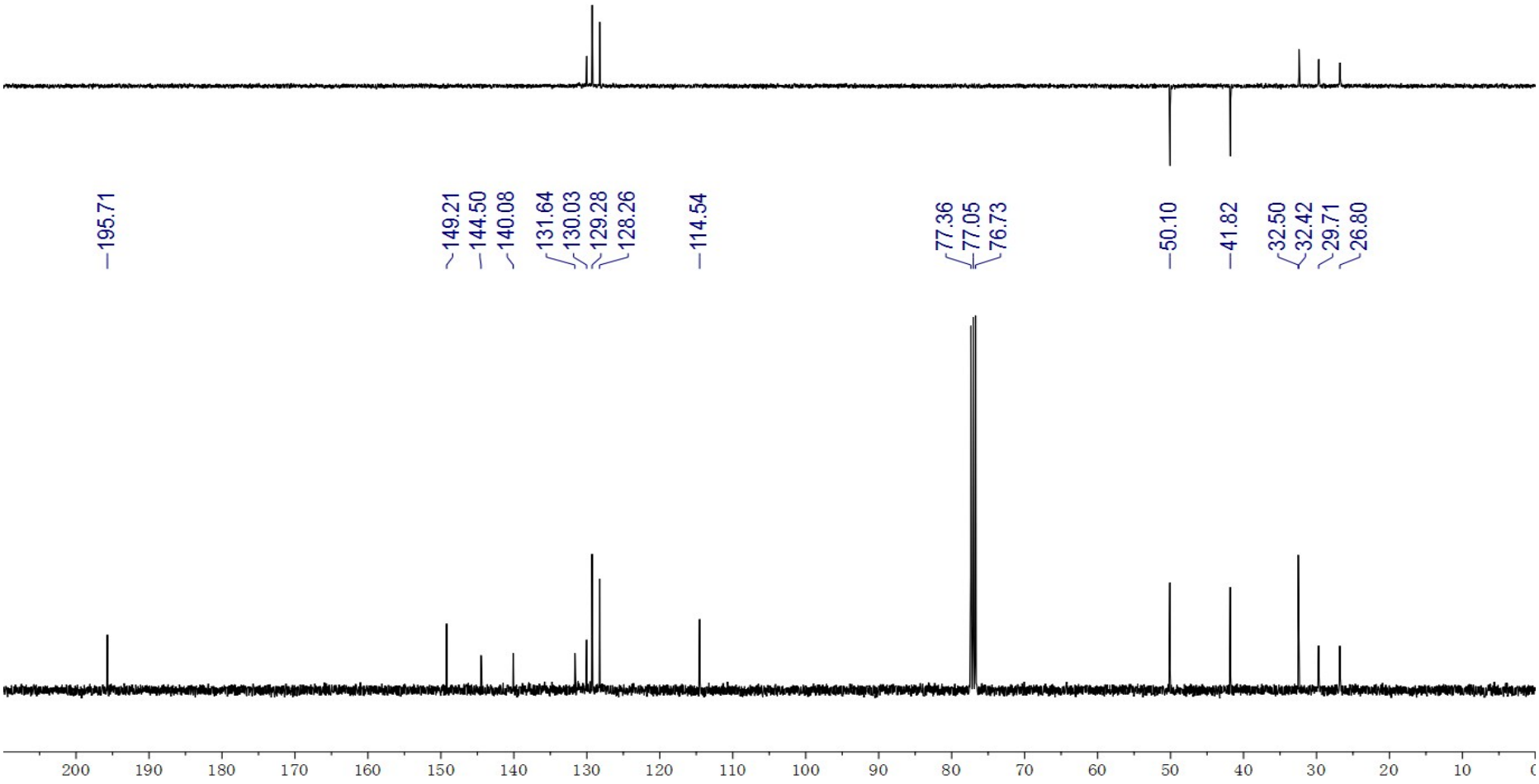


Figure 8. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3d**

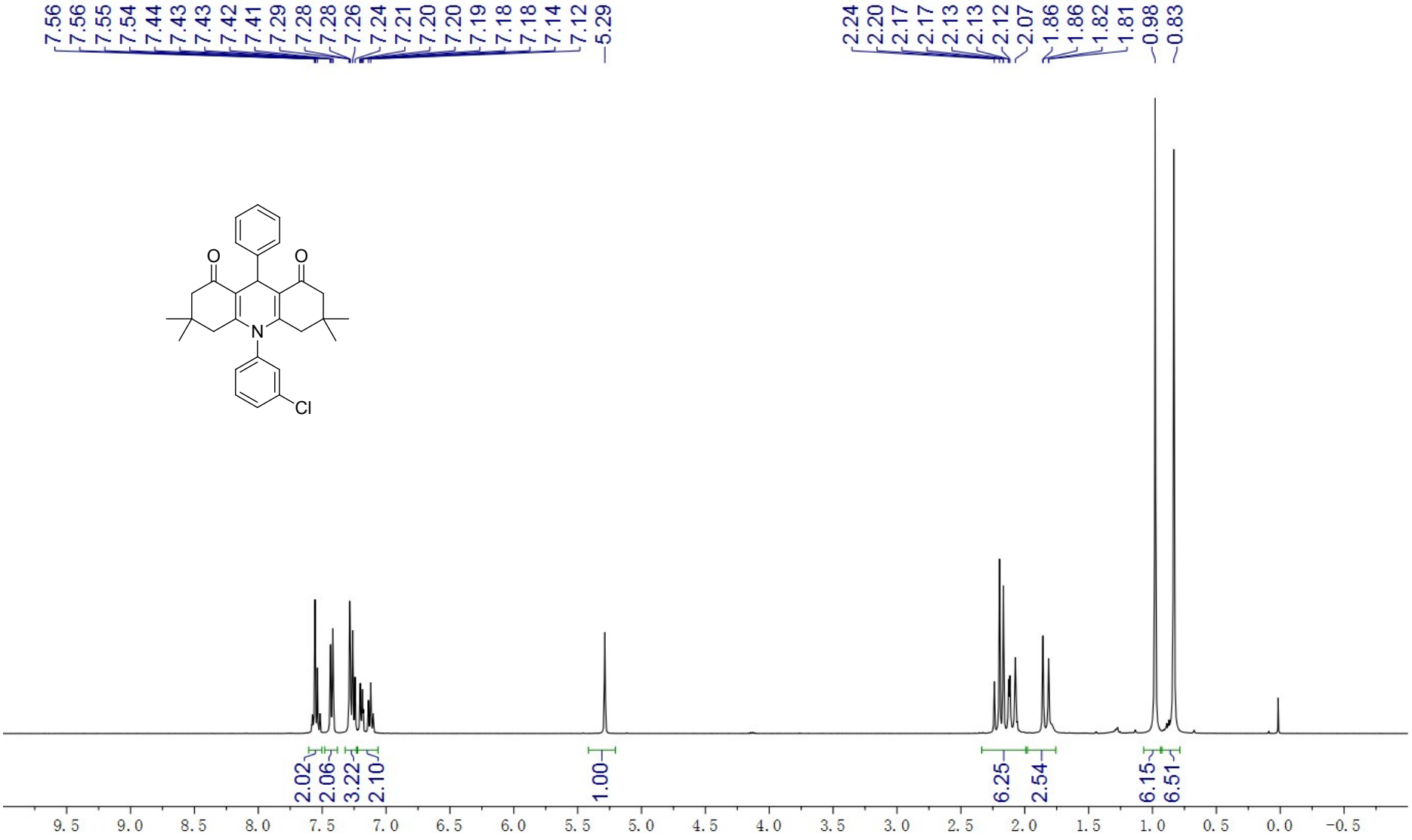


Figure 9. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3e**

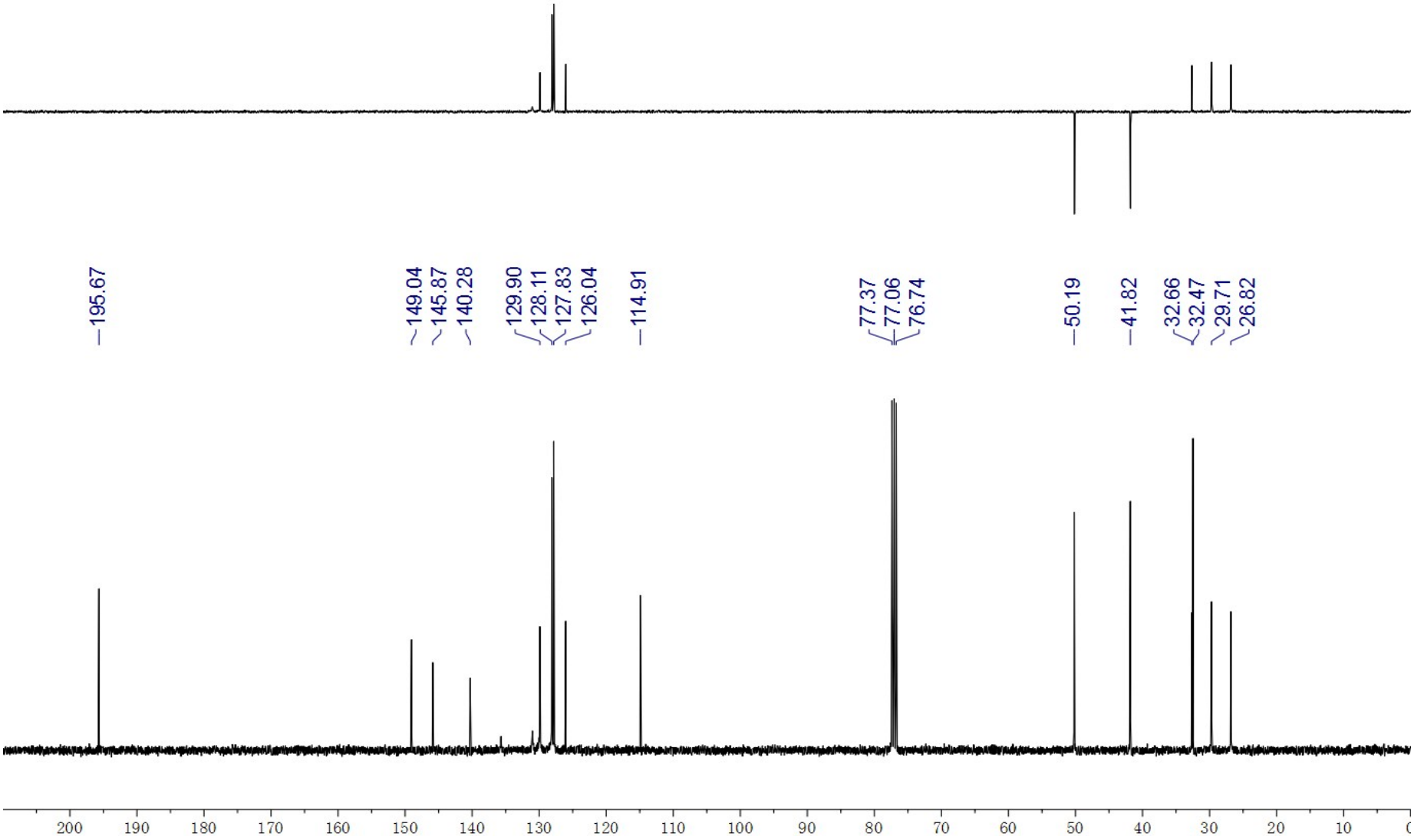


Figure 10. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3e**

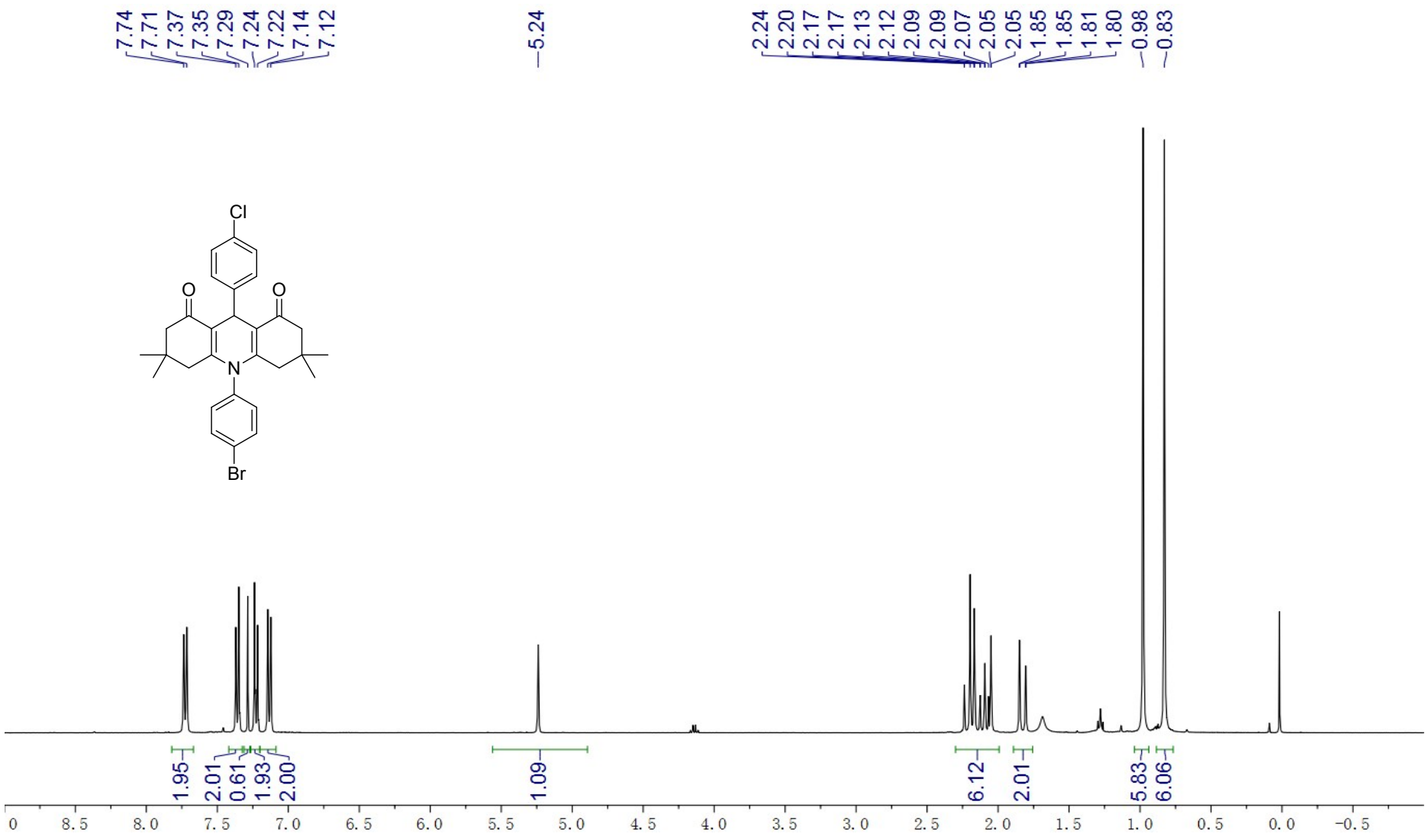


Figure 11. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3f**

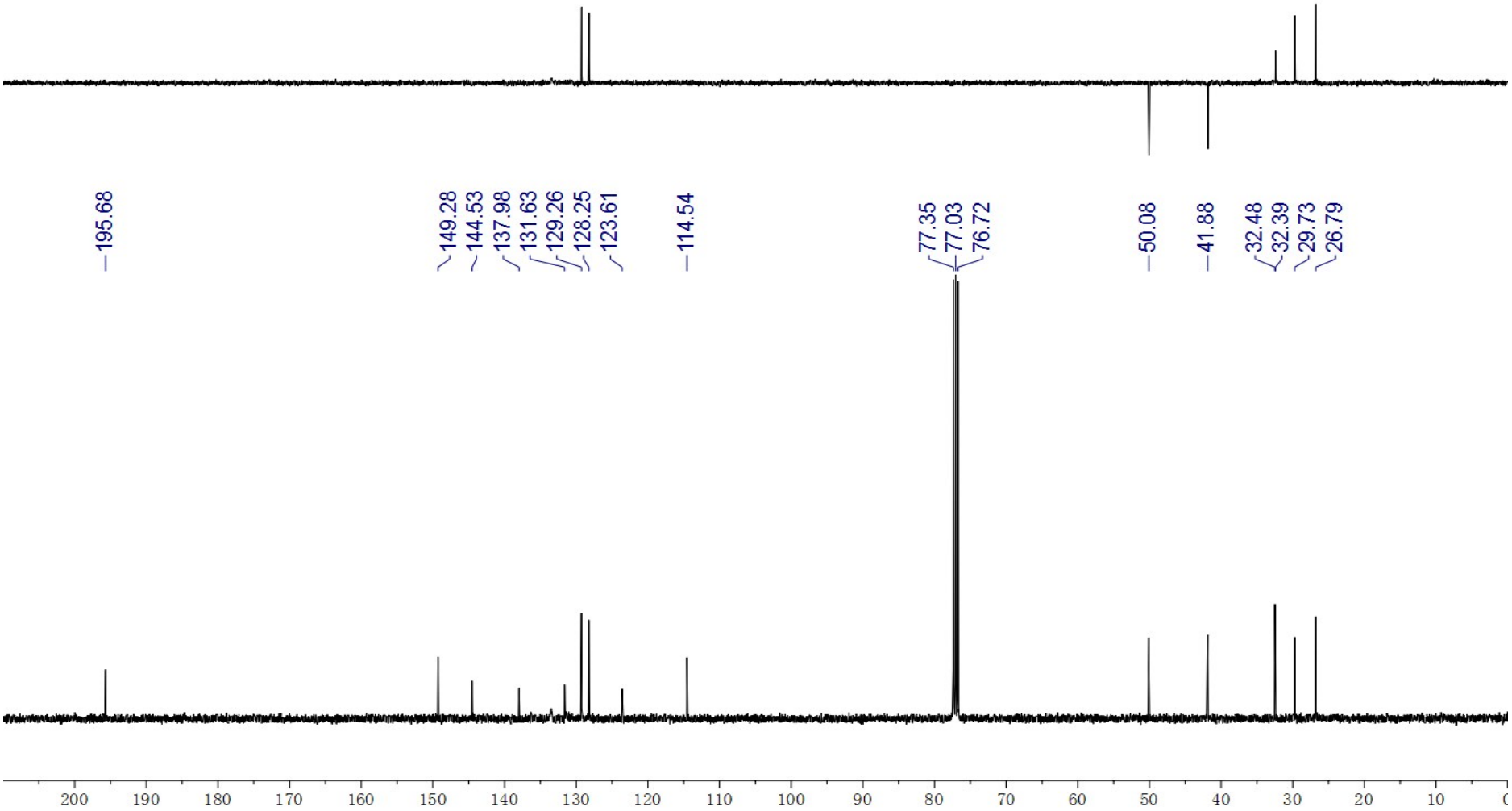


Figure 12. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3f**

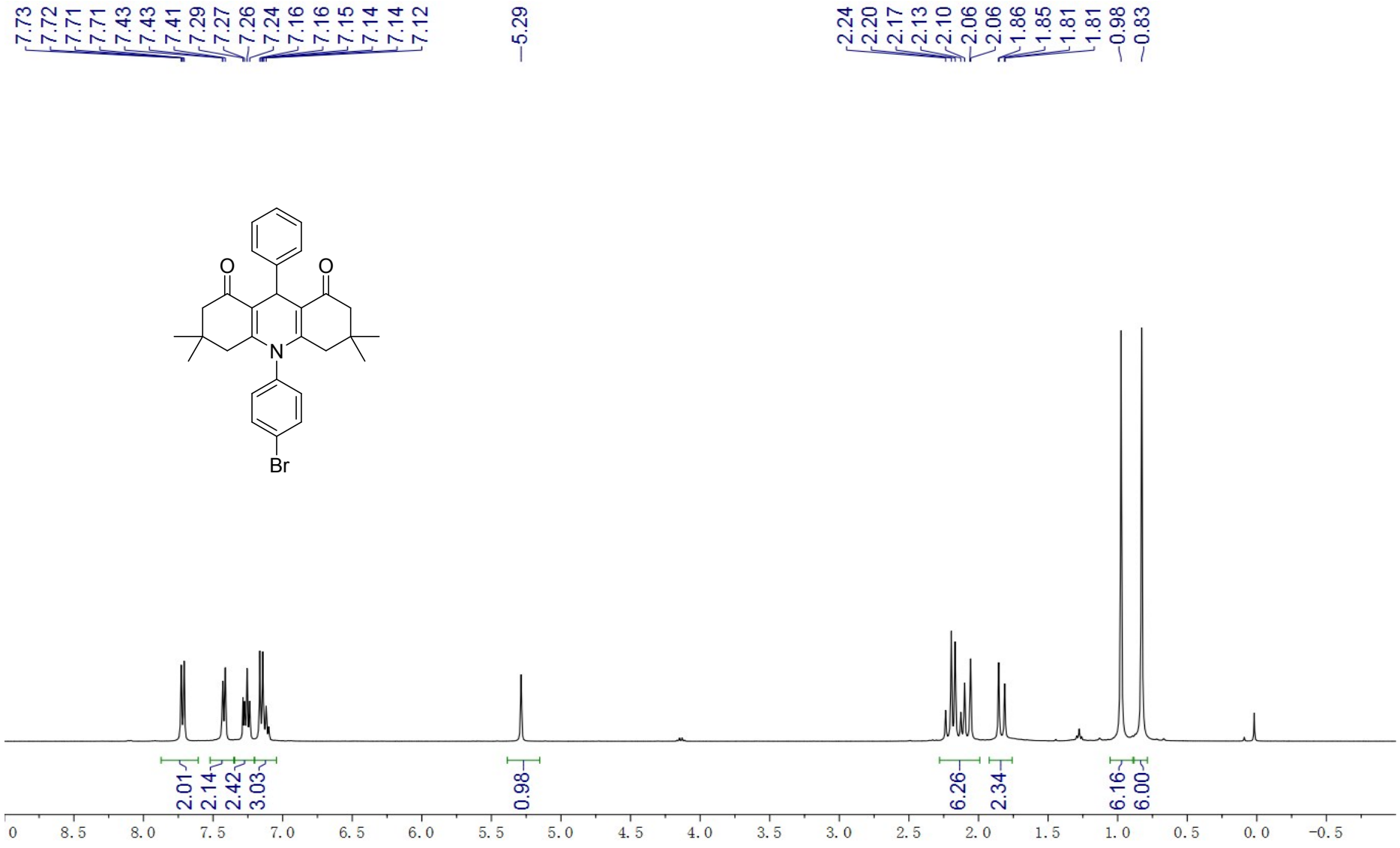


Figure 13. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3g**

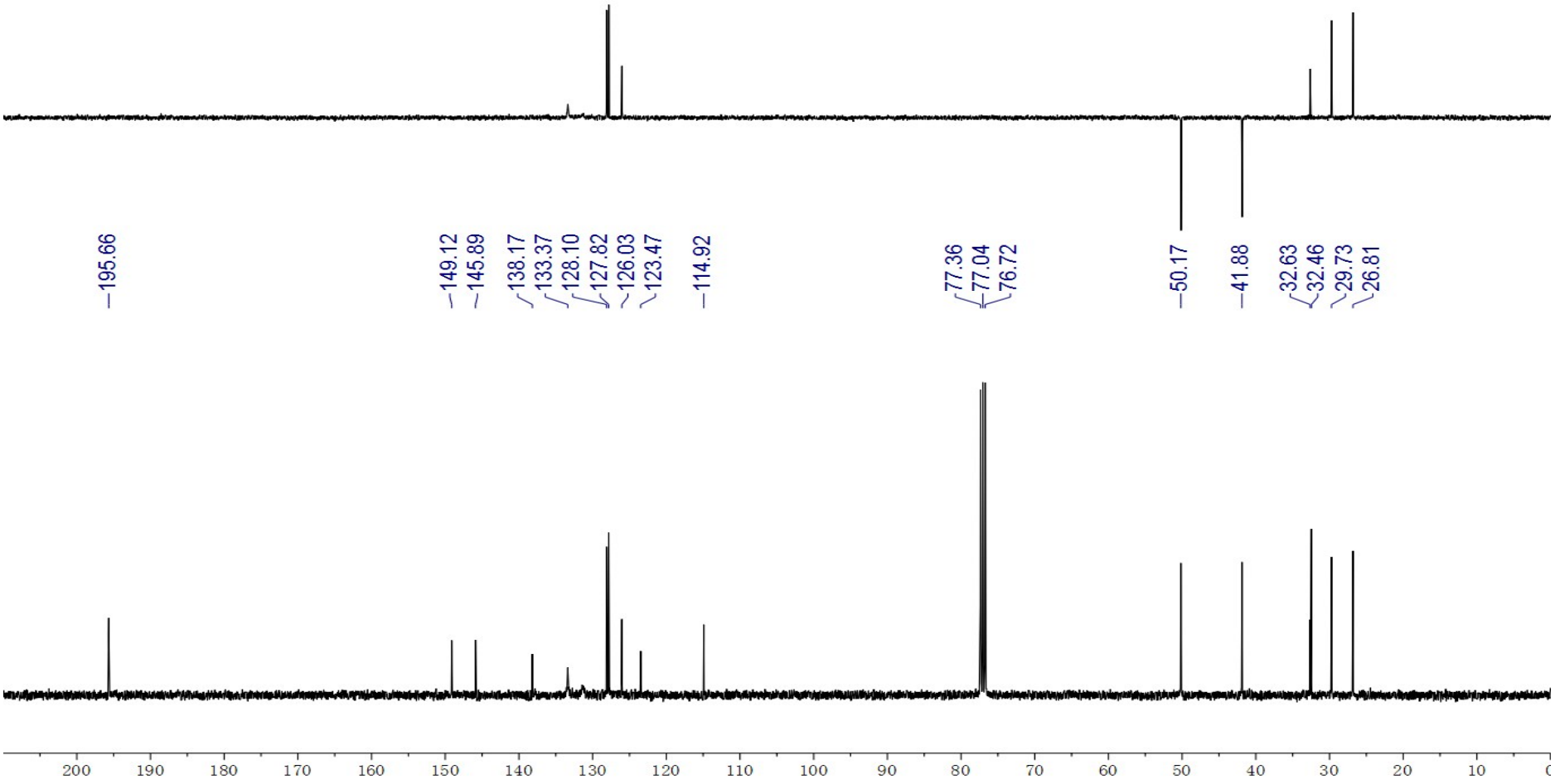


Figure 14. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3g**

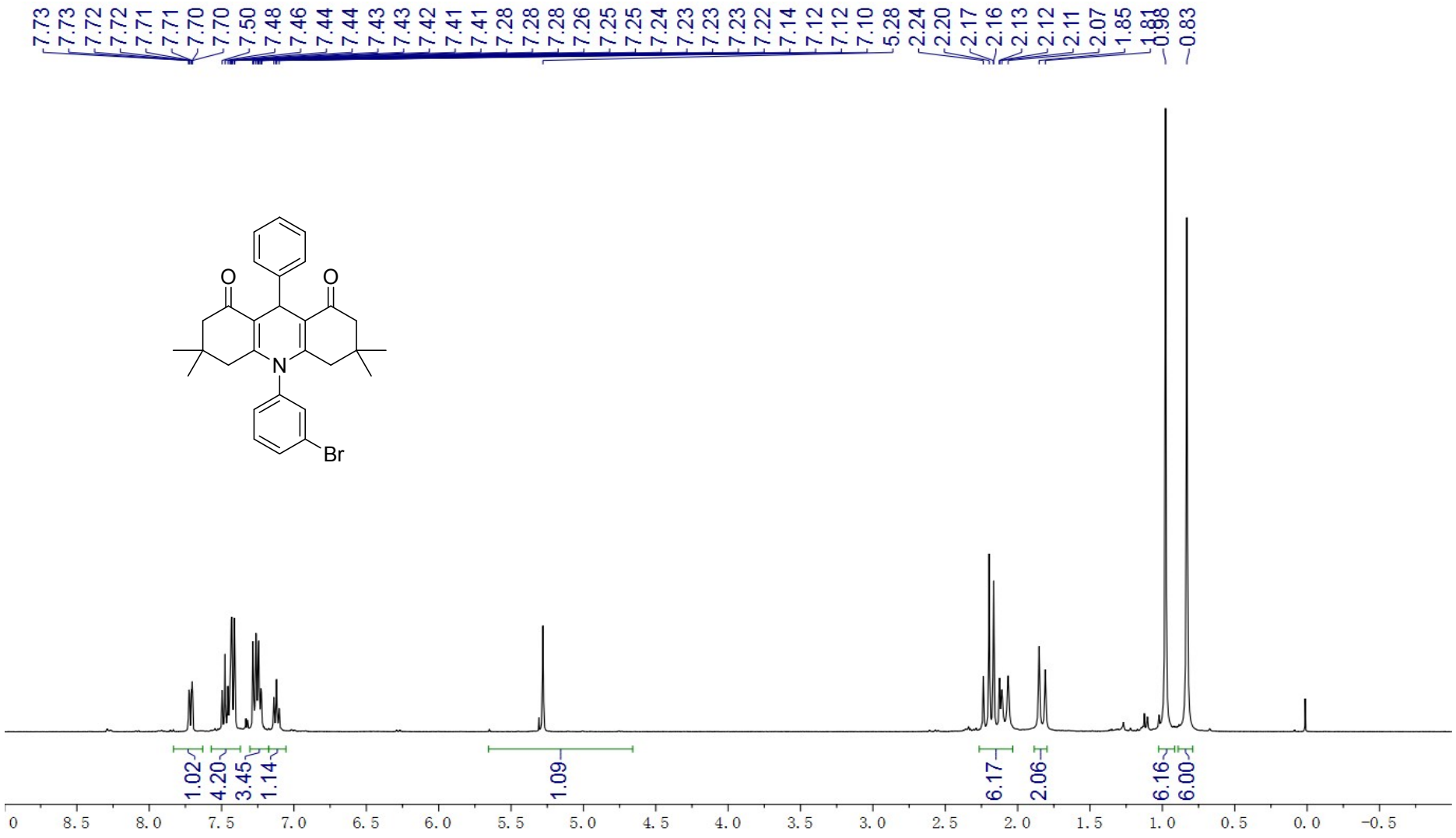


Figure 15. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3h**

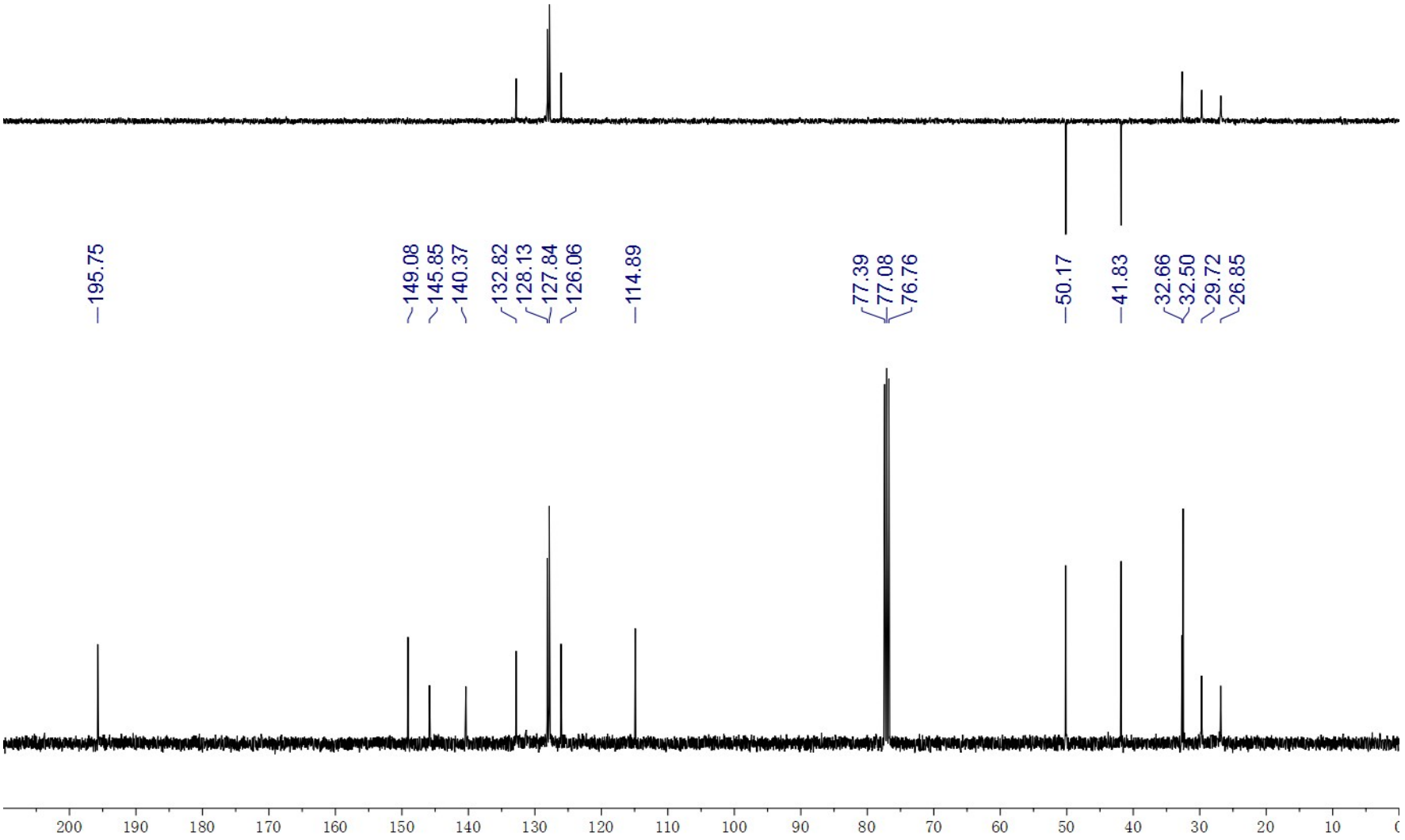


Figure 16. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3h**

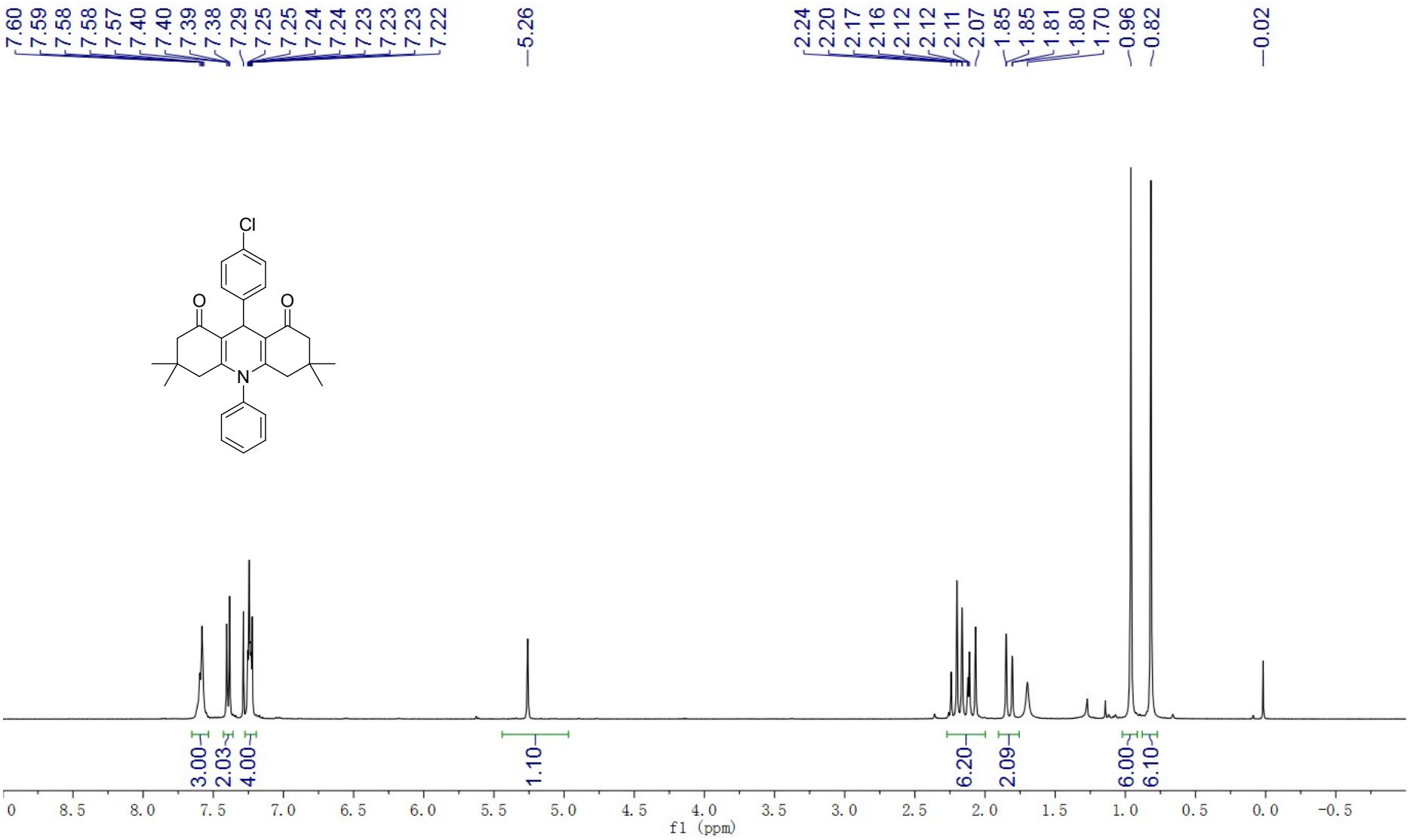


Figure 17. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3i**

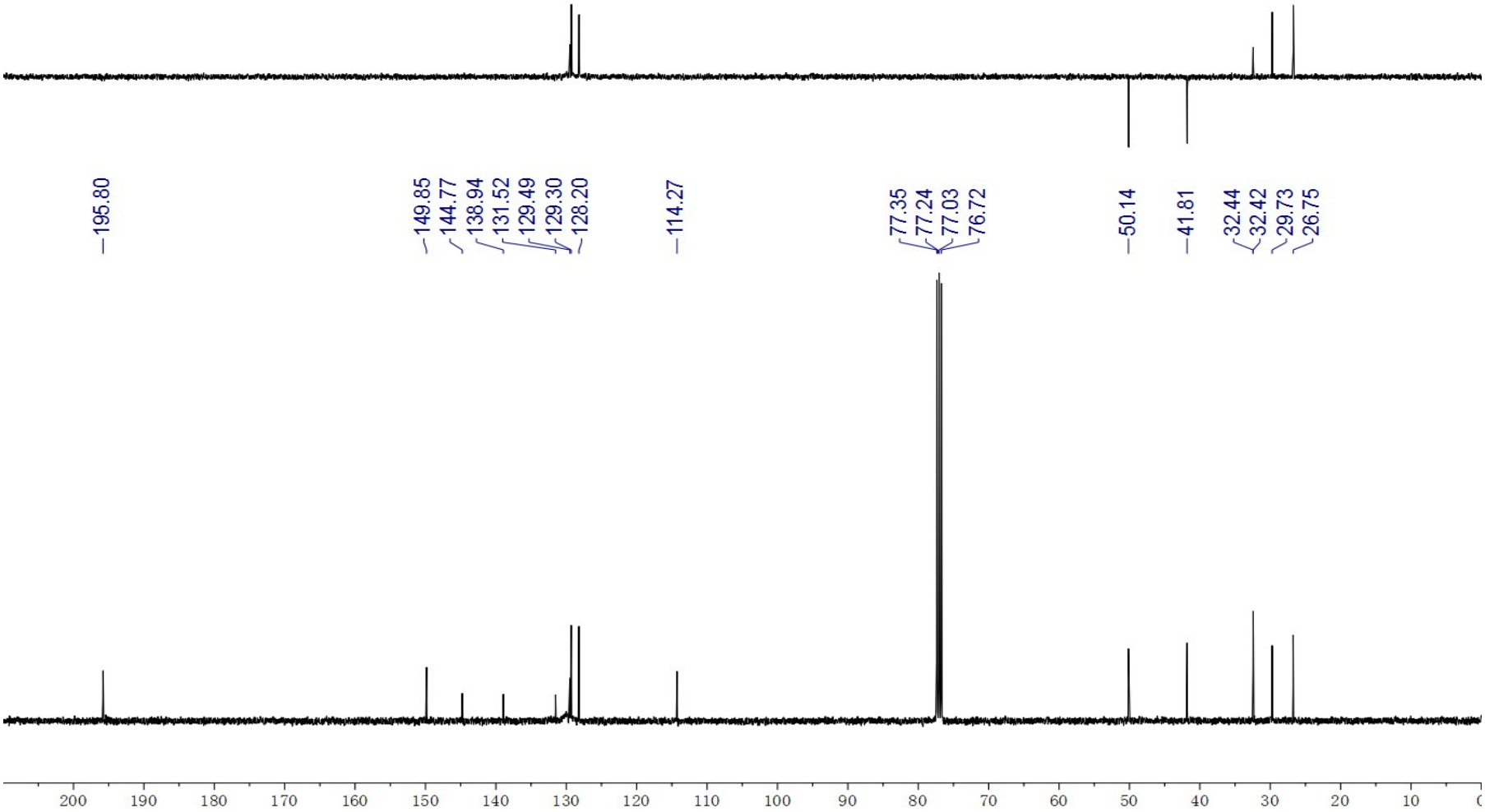


Figure 18. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3i**

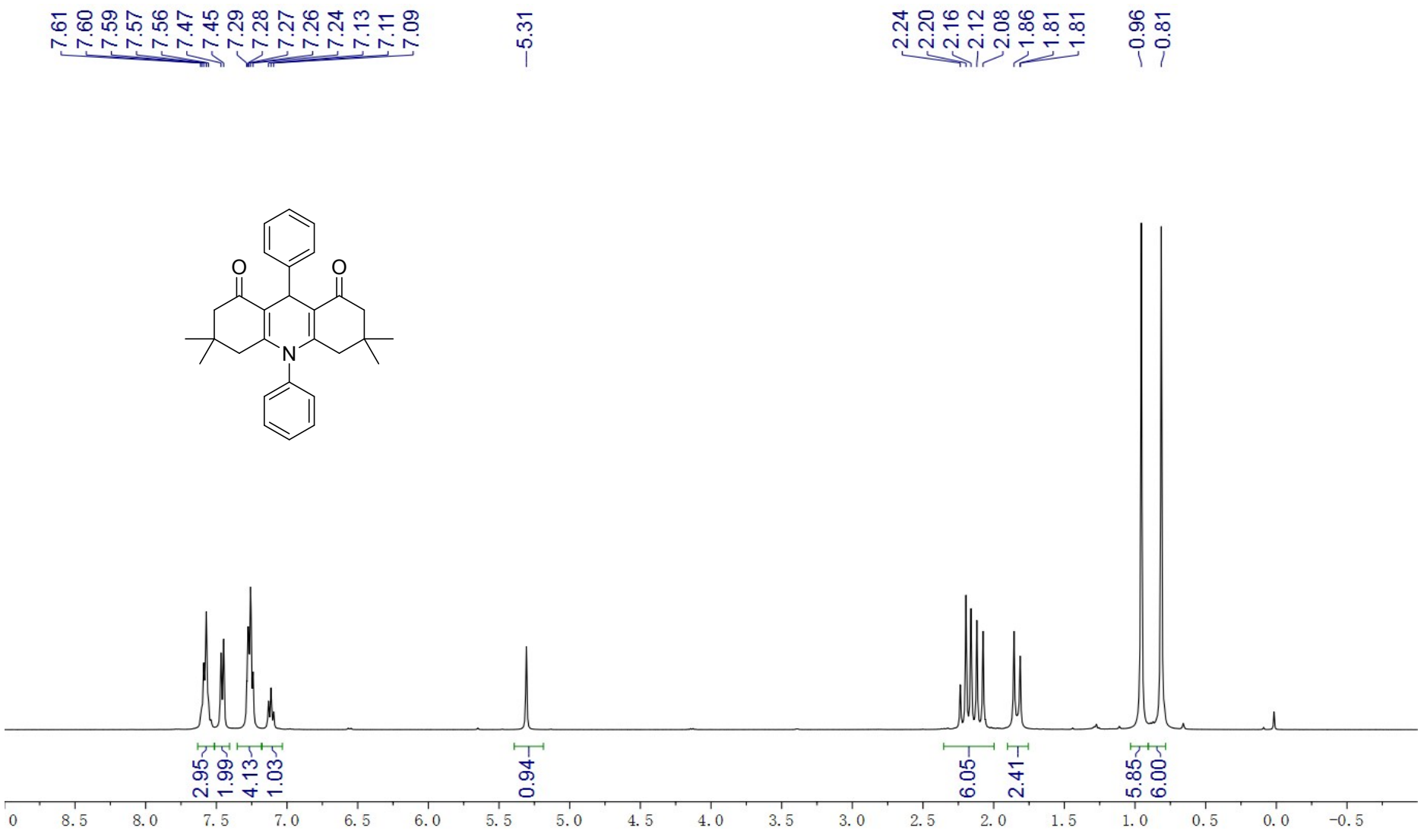


Figure 19. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3j**

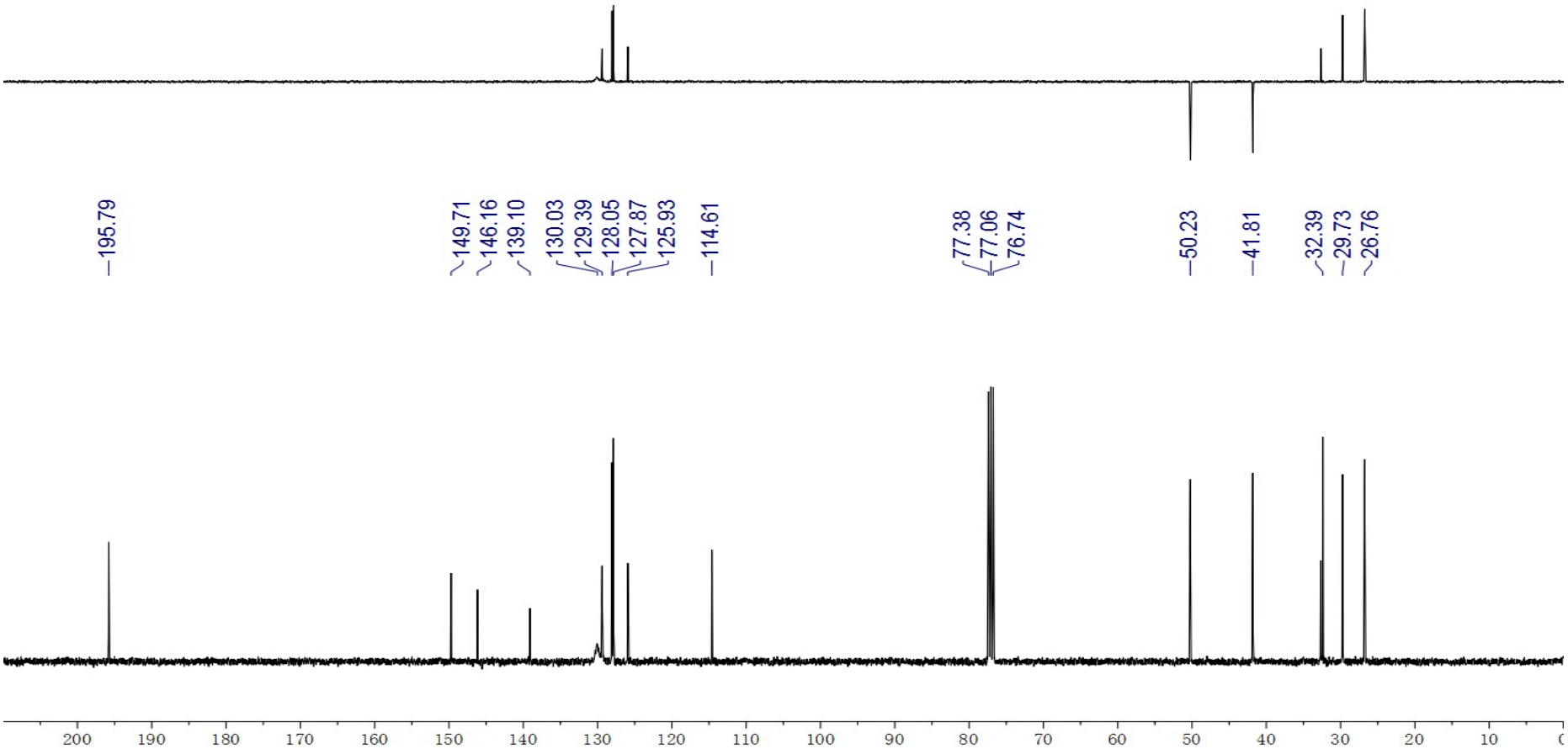


Figure 20. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3j**

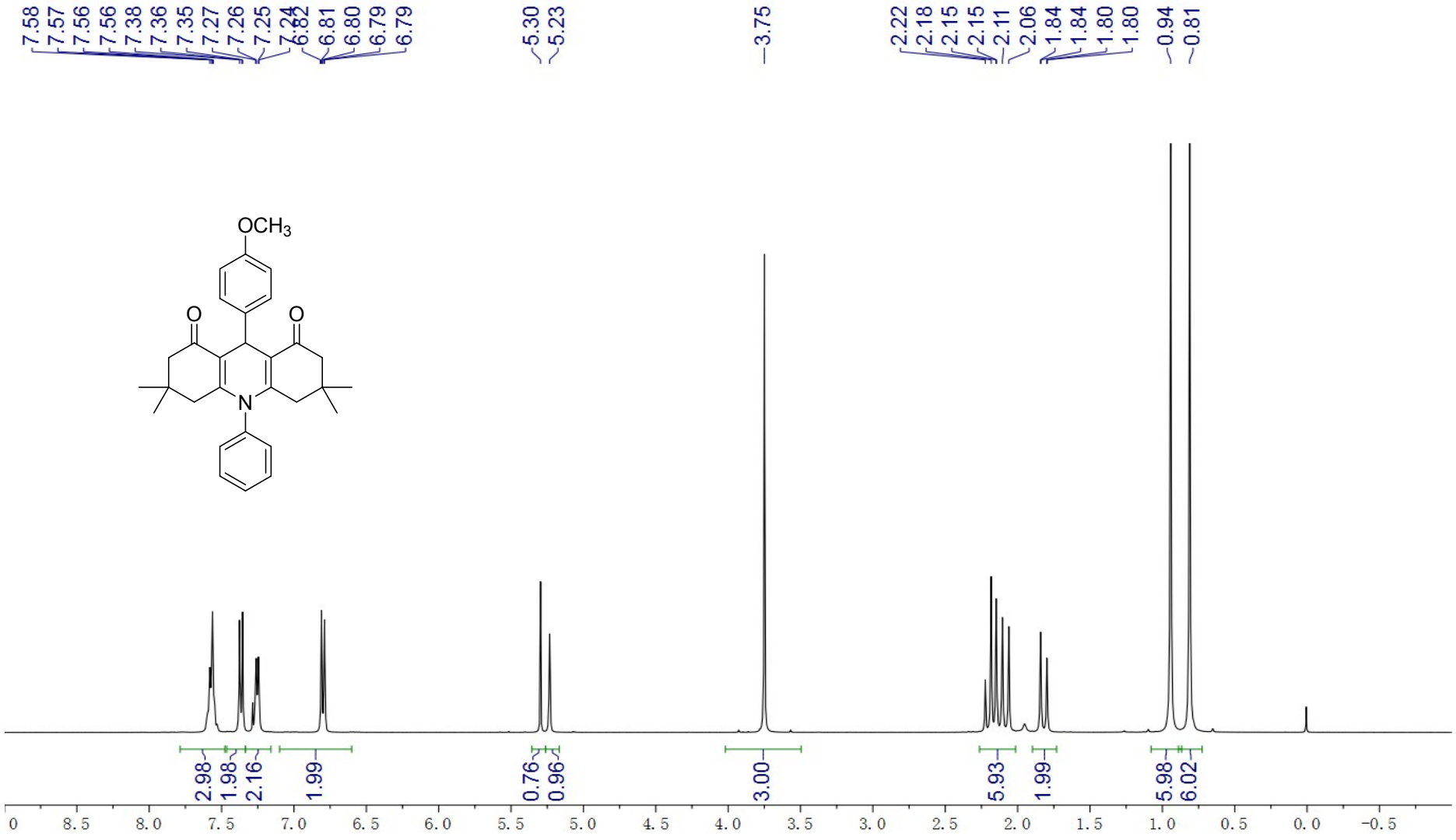


Figure 21. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3k**

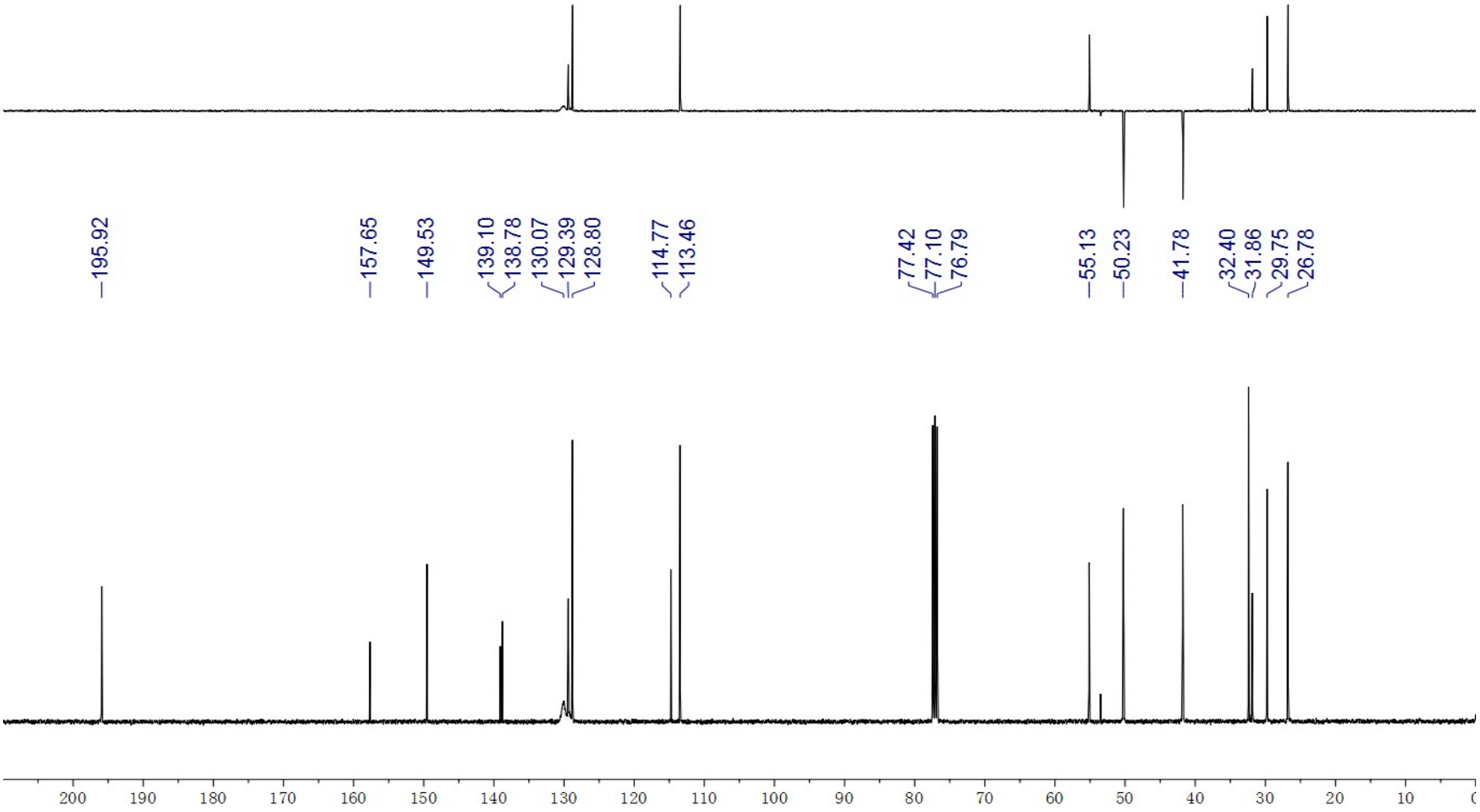


Figure 22. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3k**

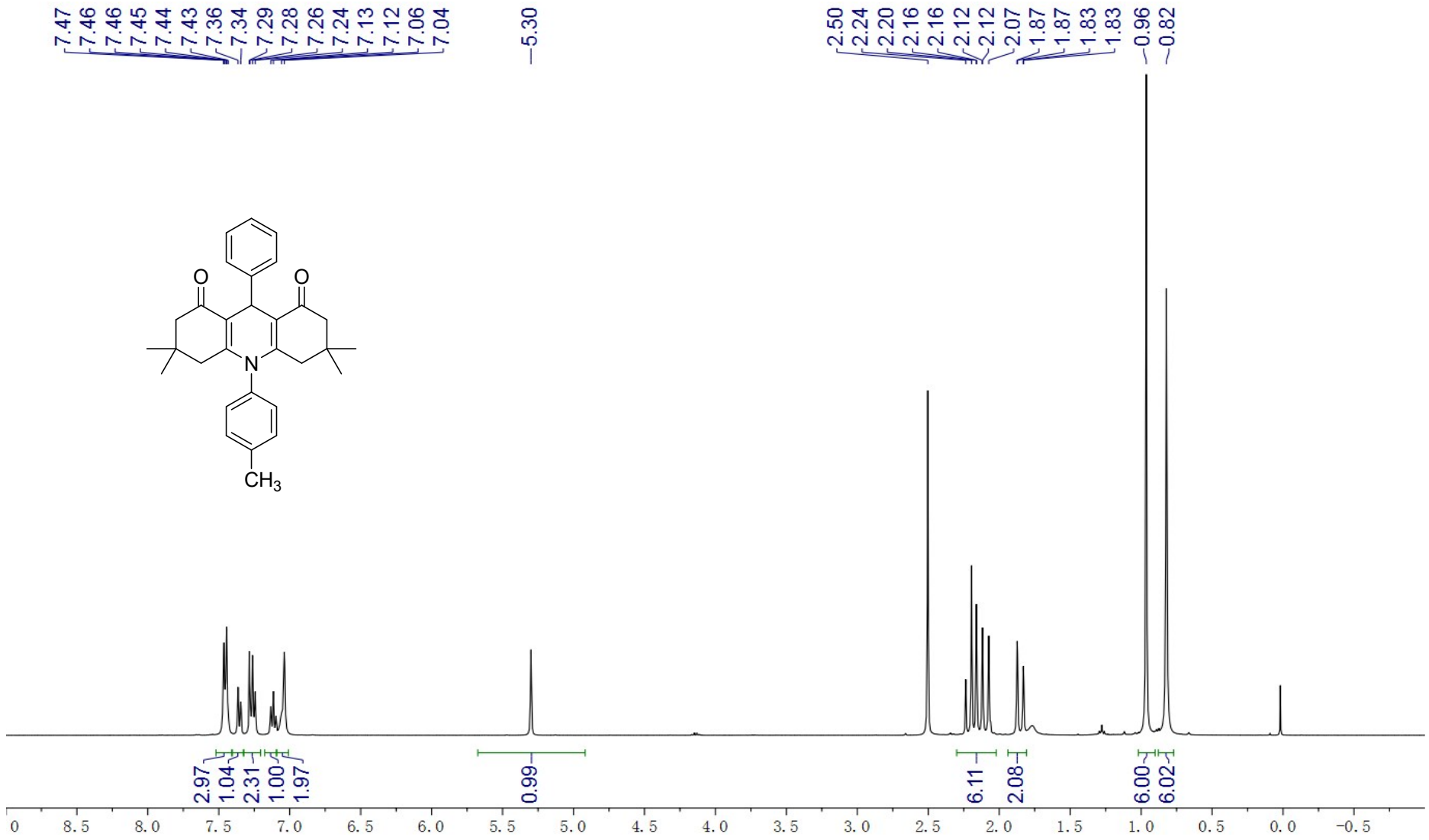


Figure 23. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **3l**

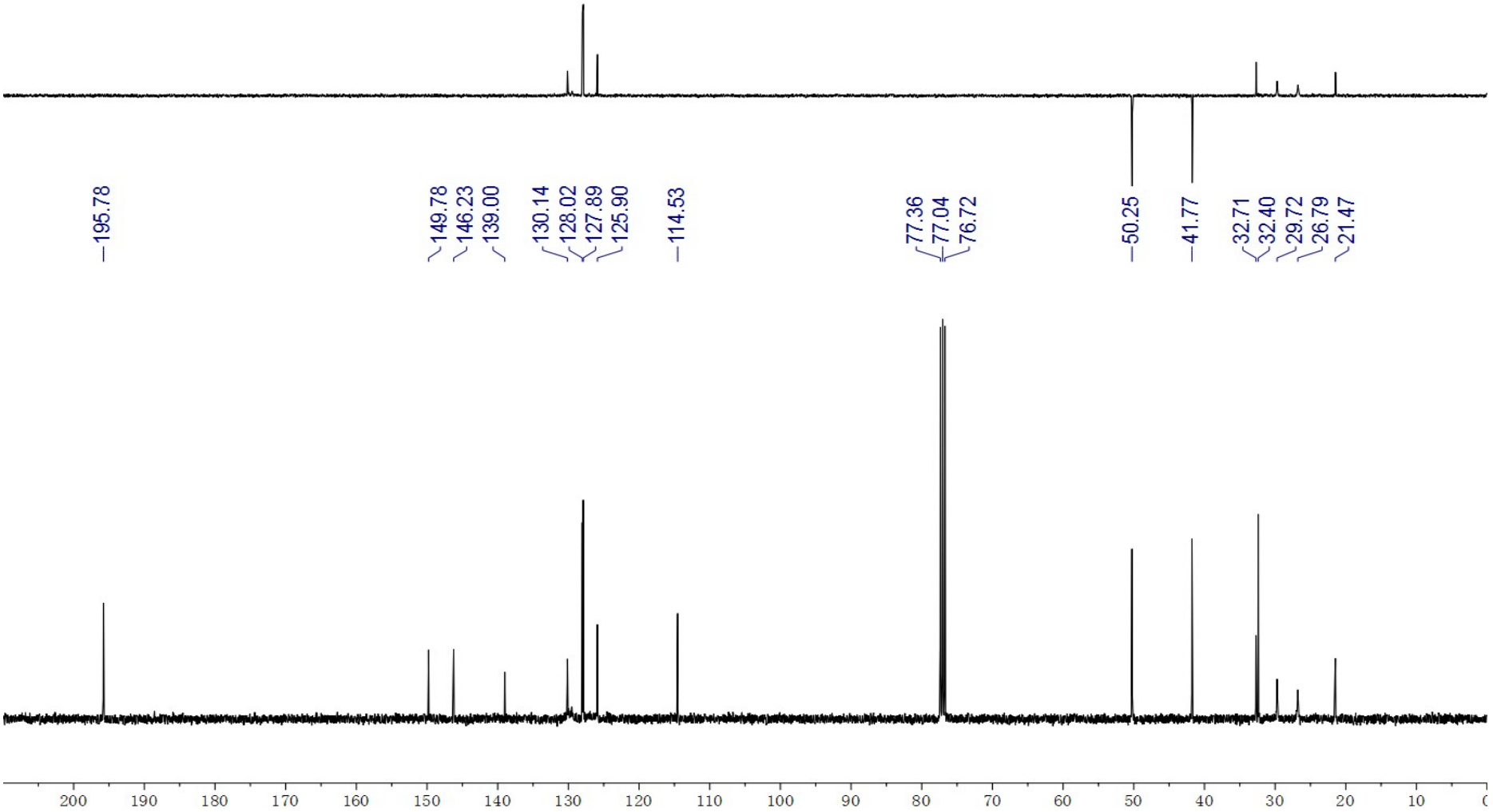


Figure 24. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **3l**

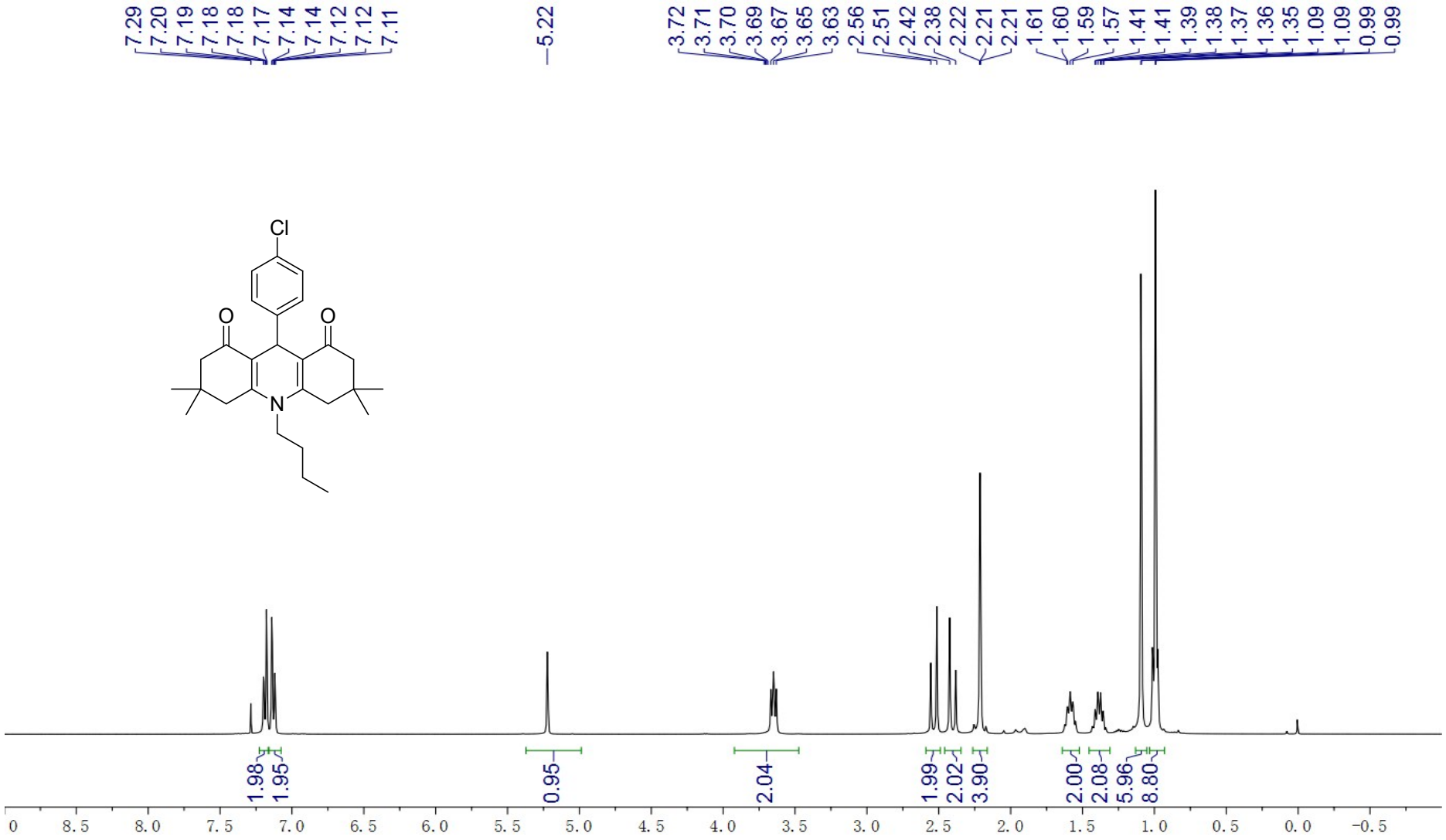


Figure 25. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3m**

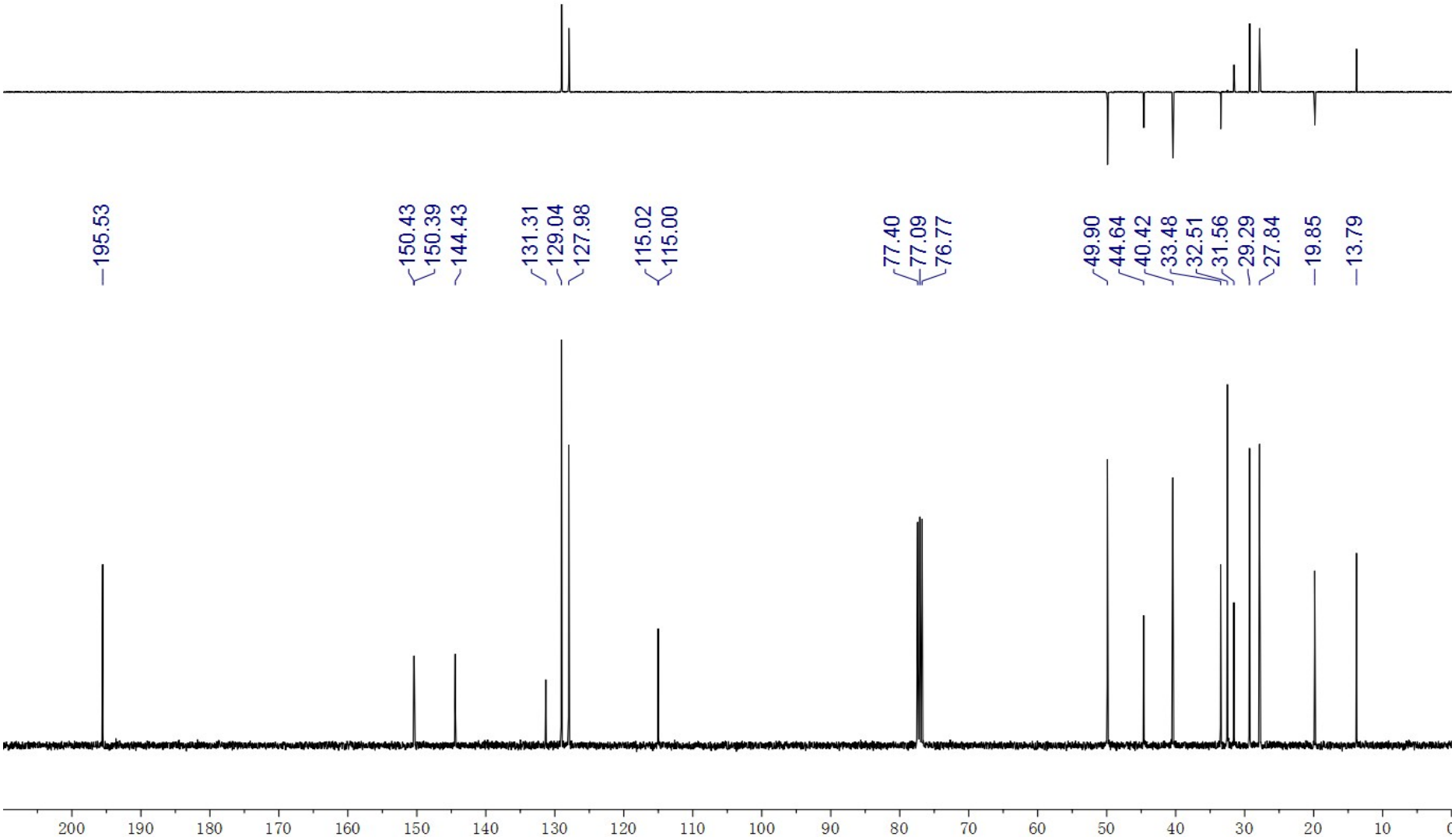


Figure 26. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3m**

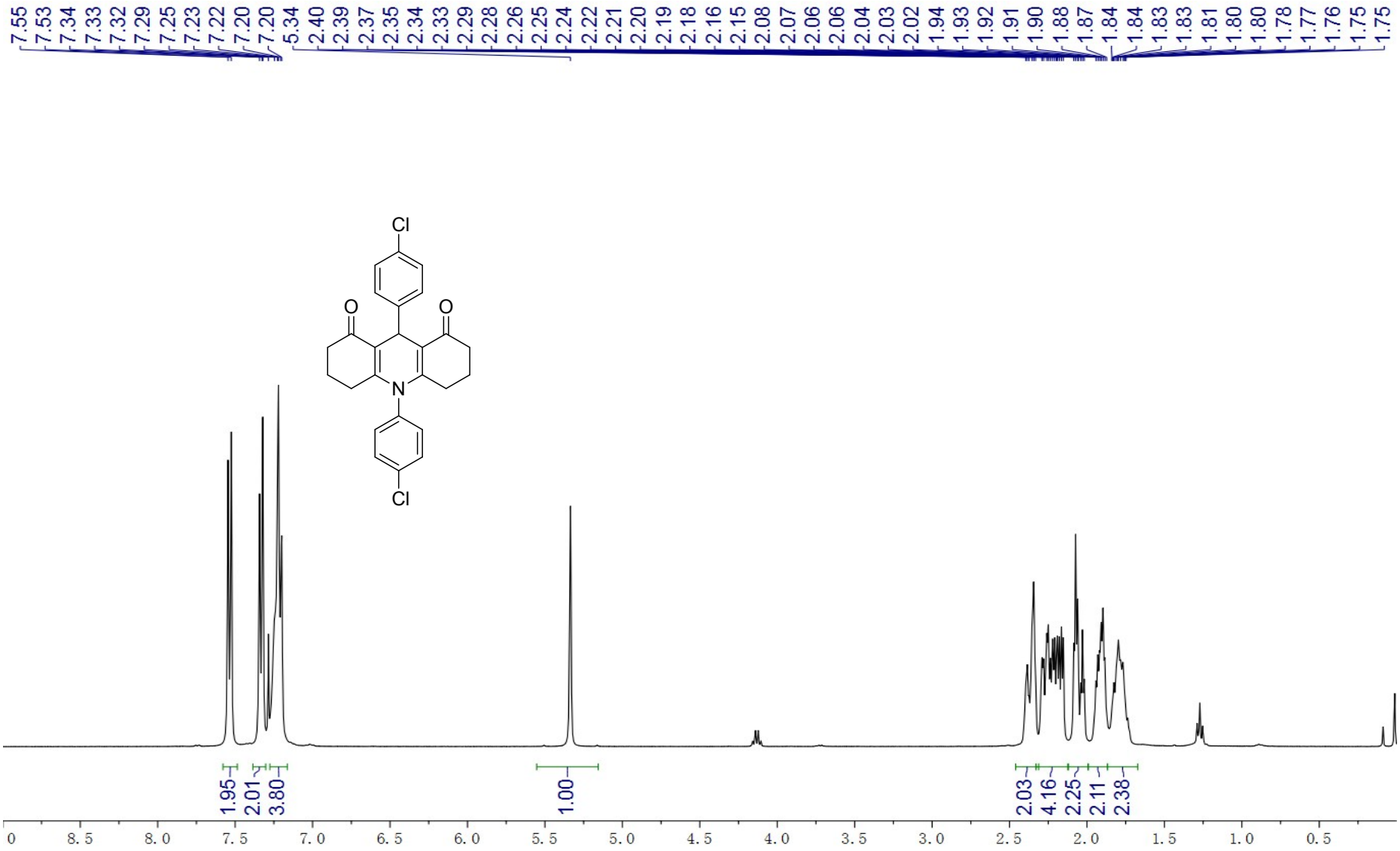


Figure 27. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3n**

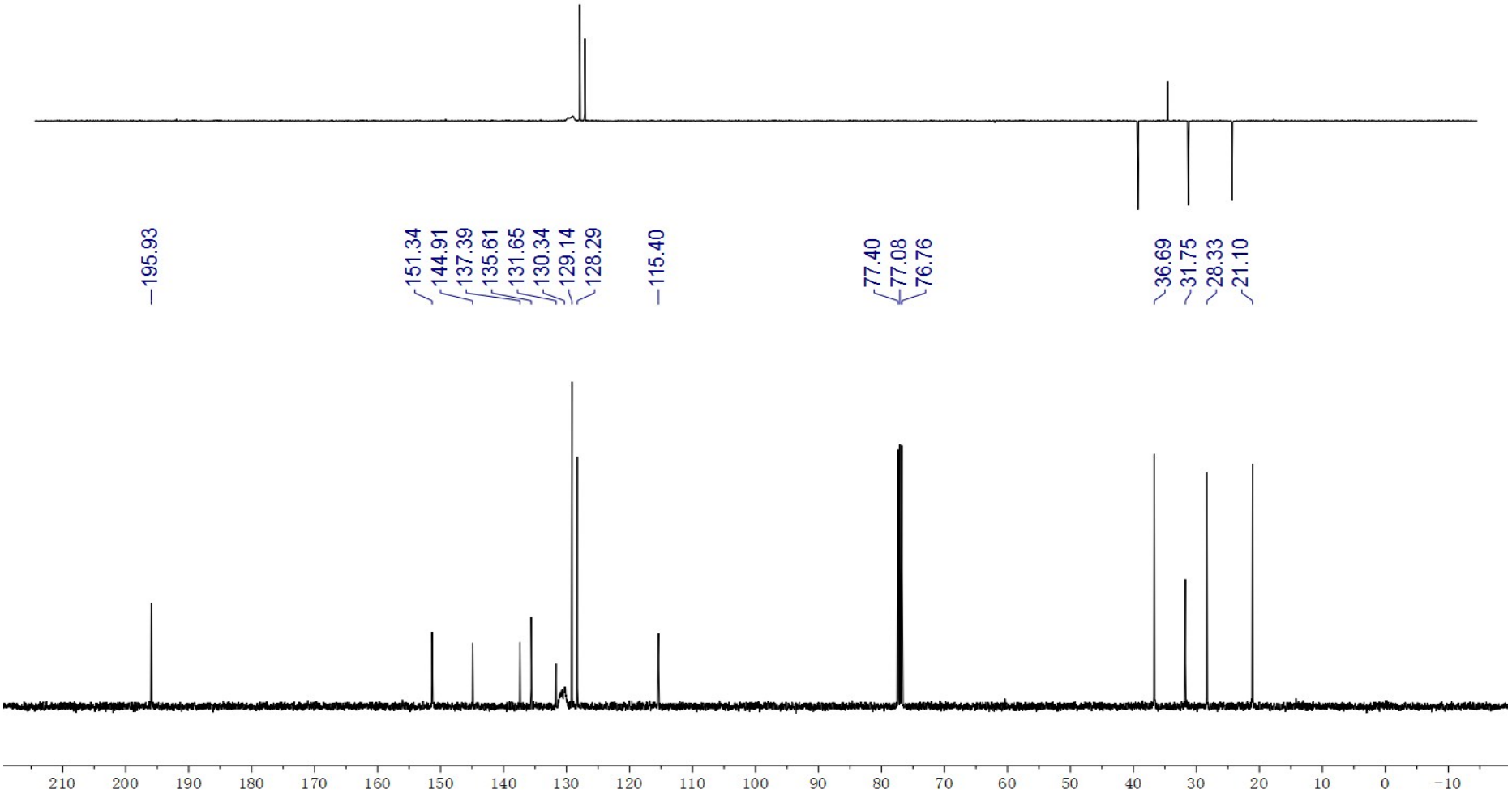


Figure 28. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3n**

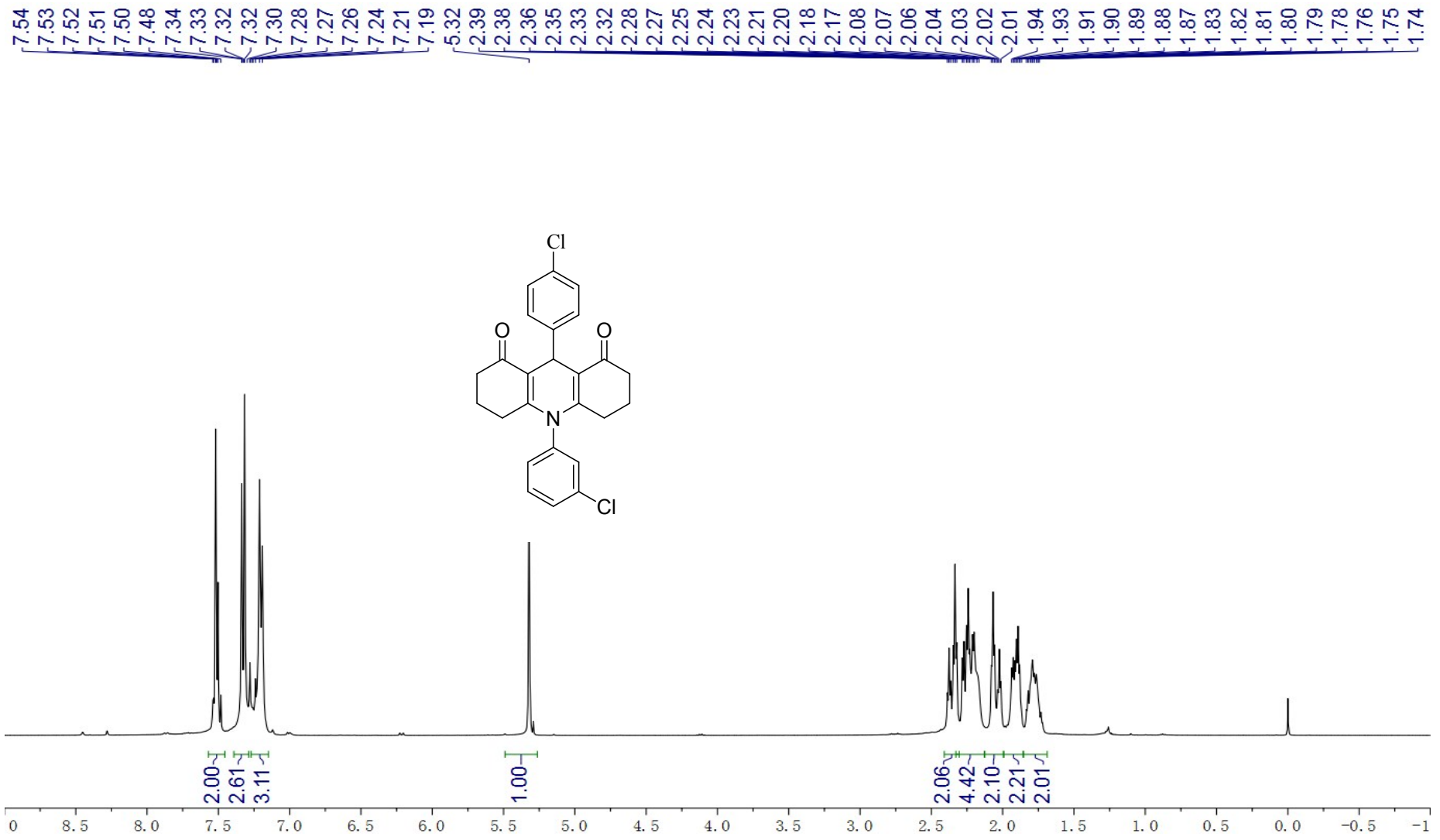


Figure 29. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3o**

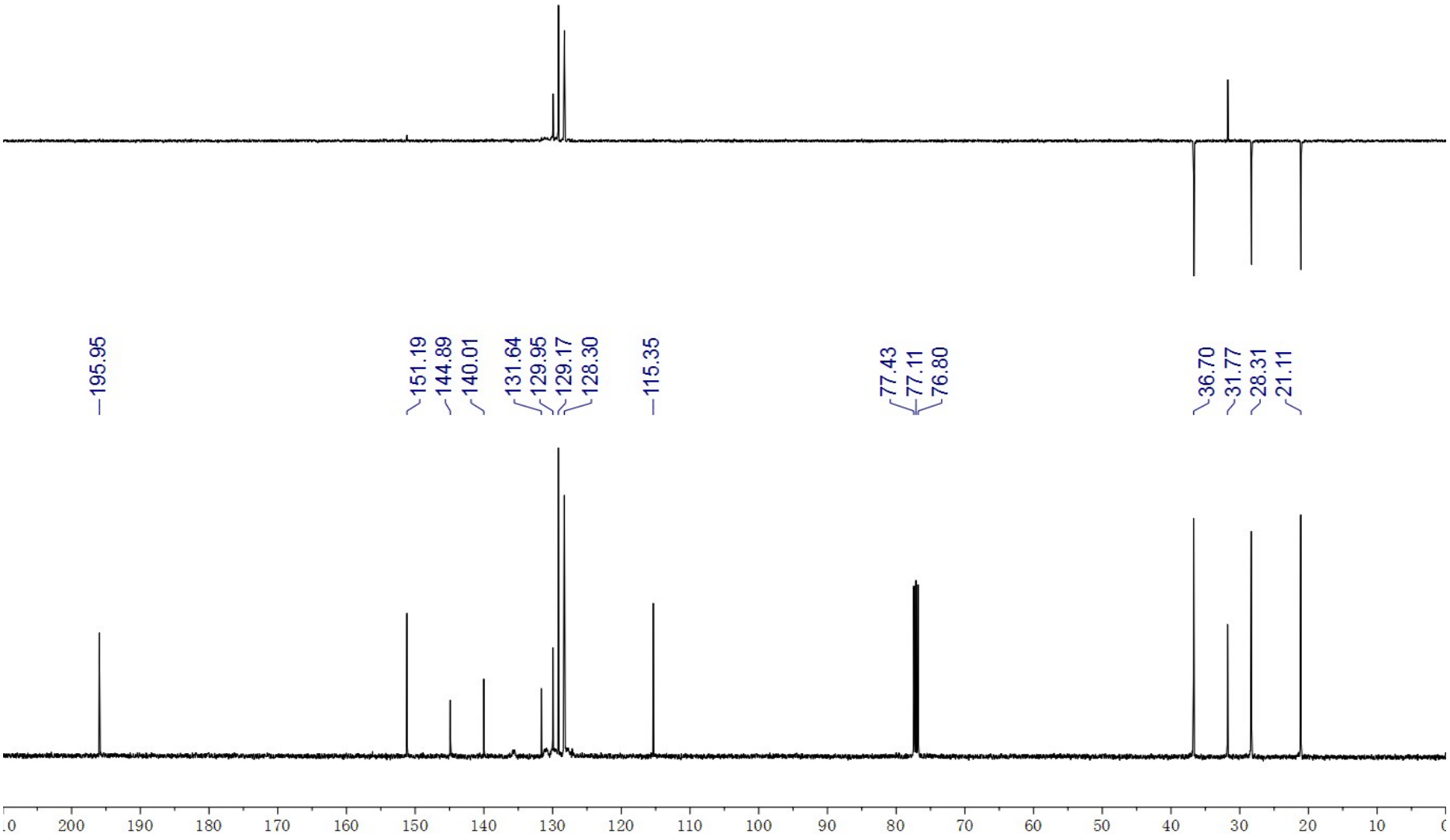


Figure 30. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3o**

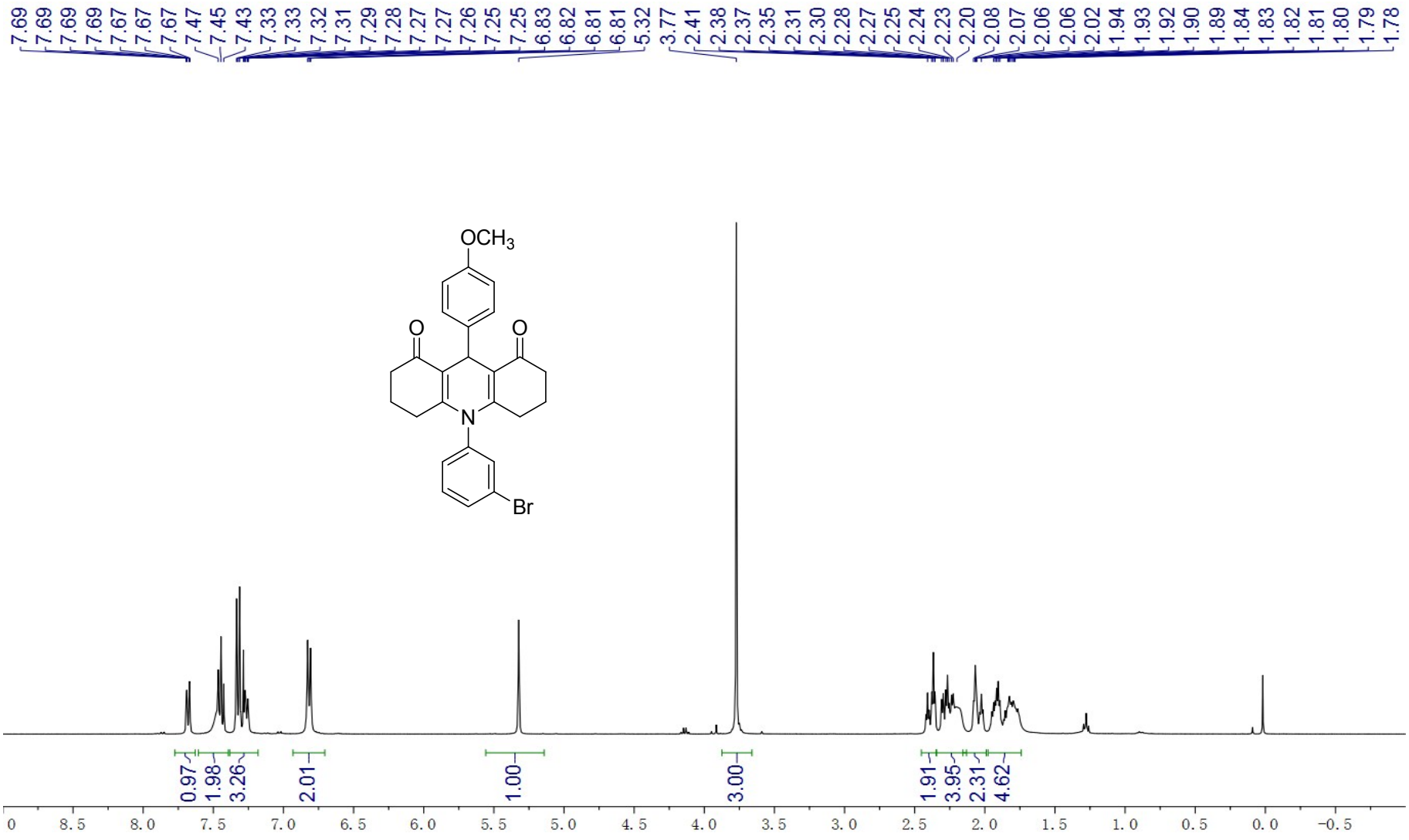


Figure 31. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3p**

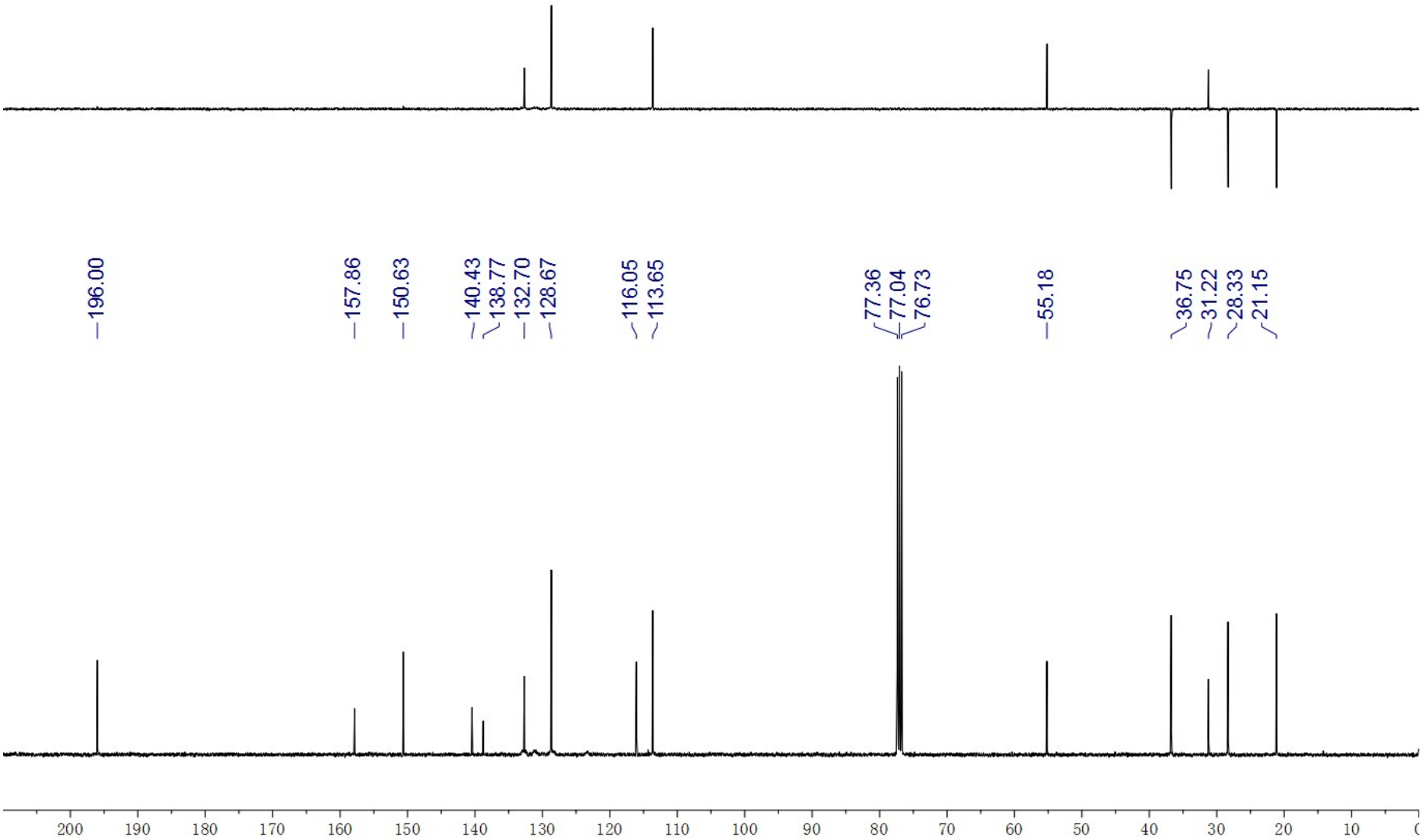


Figure 32. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3p**

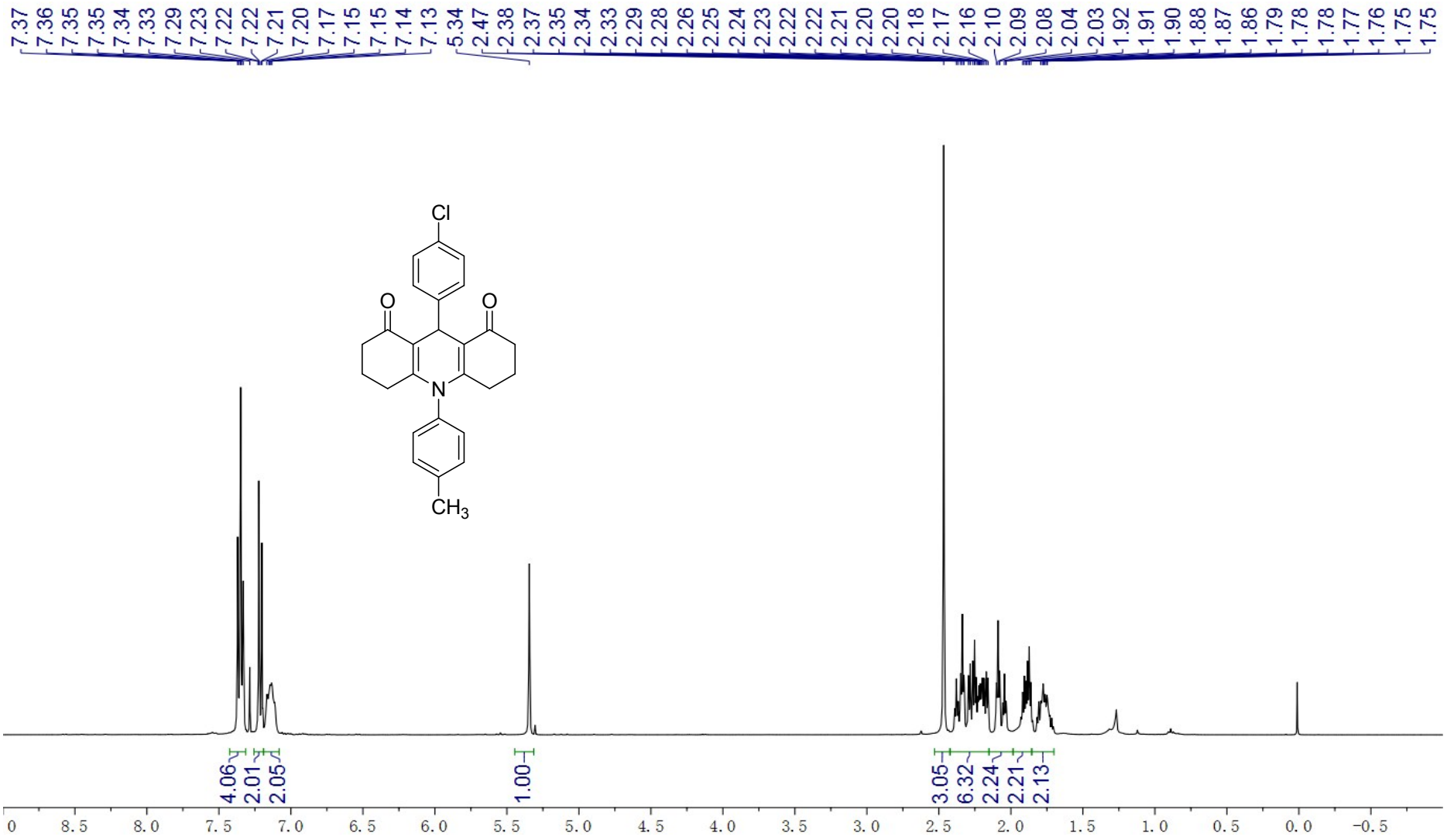


Figure 33. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **3q**

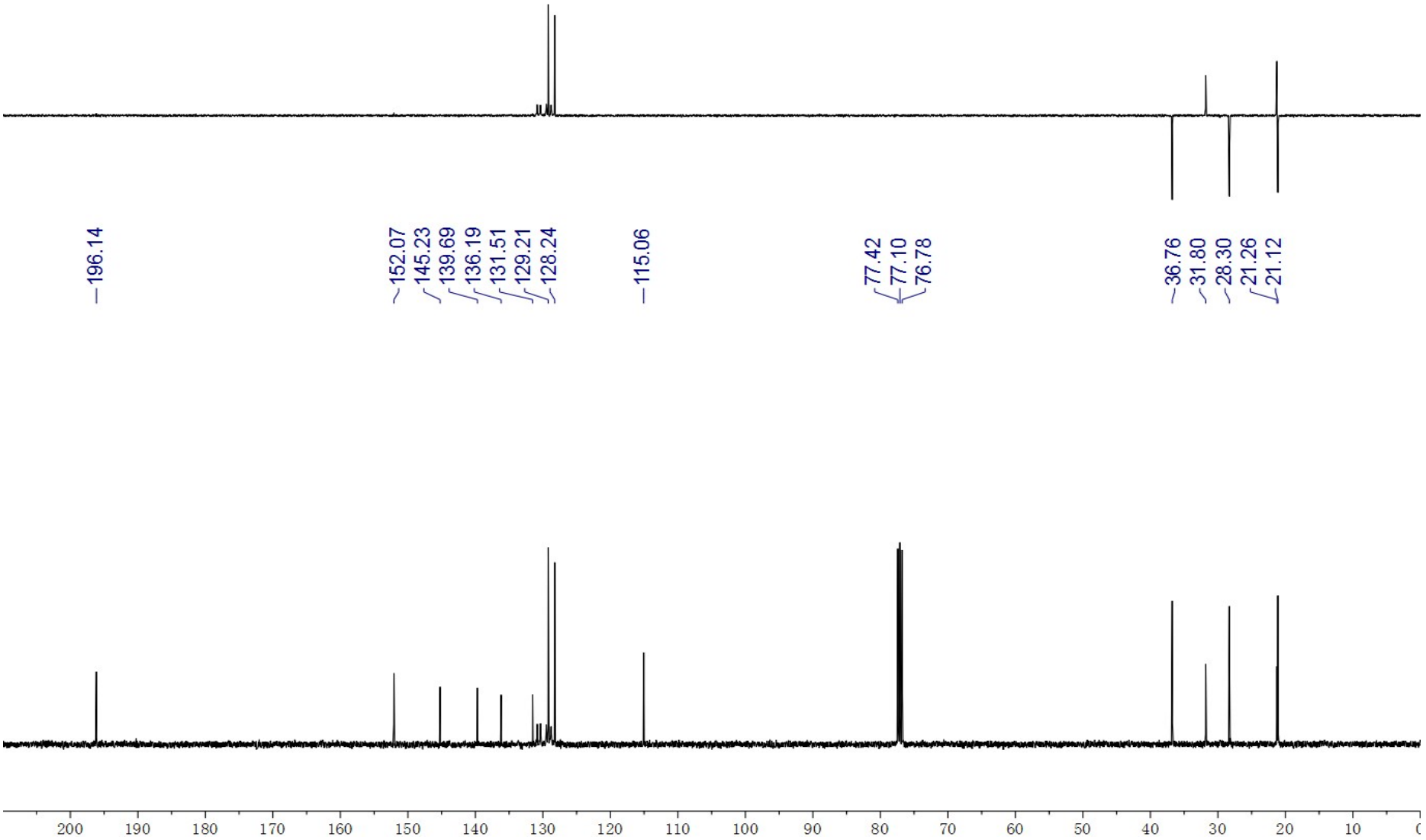


Figure 34. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **3q**

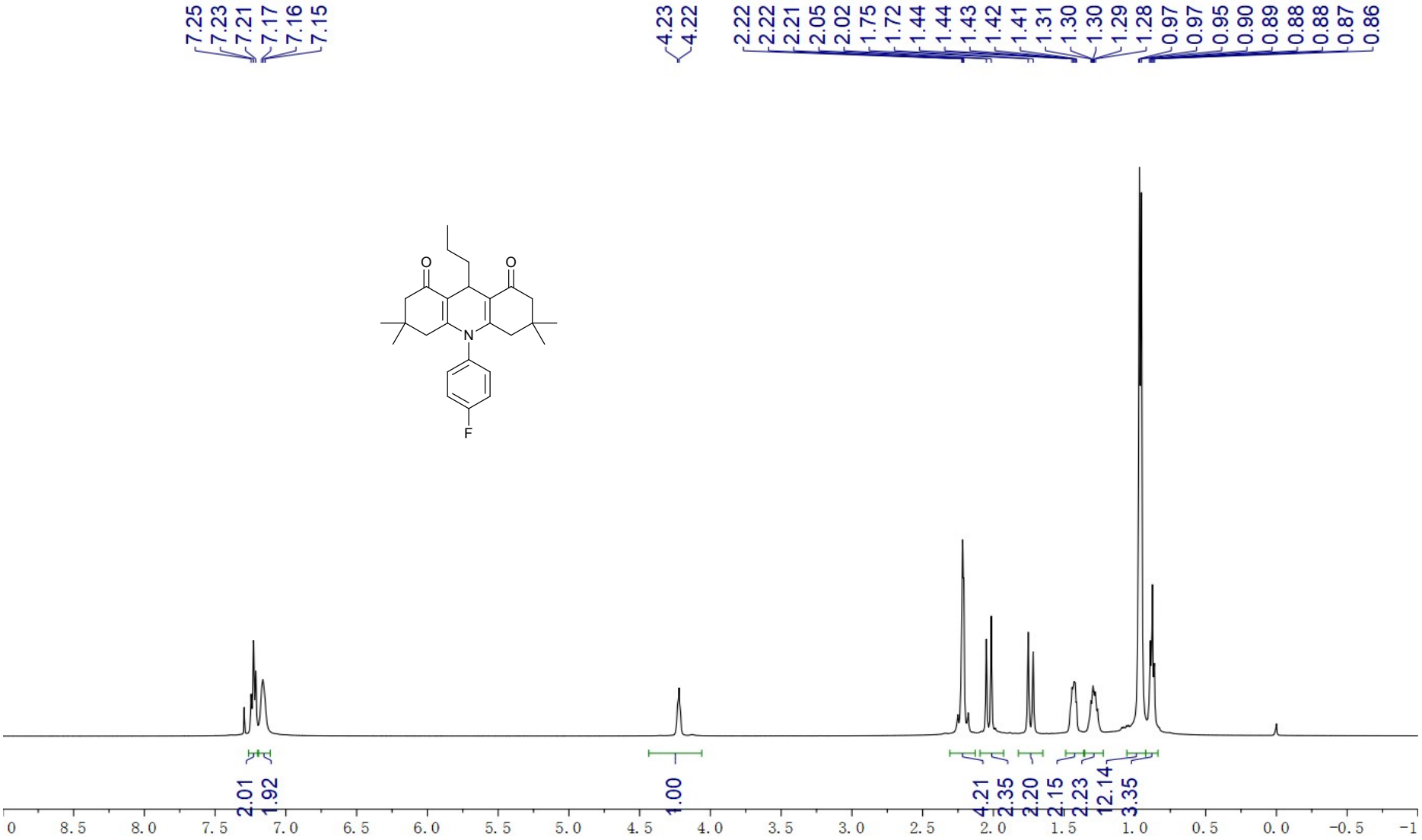


Figure 35. ^{1}H NMR (500 MHz, $CDCl_3$) spectra of compound **3r**

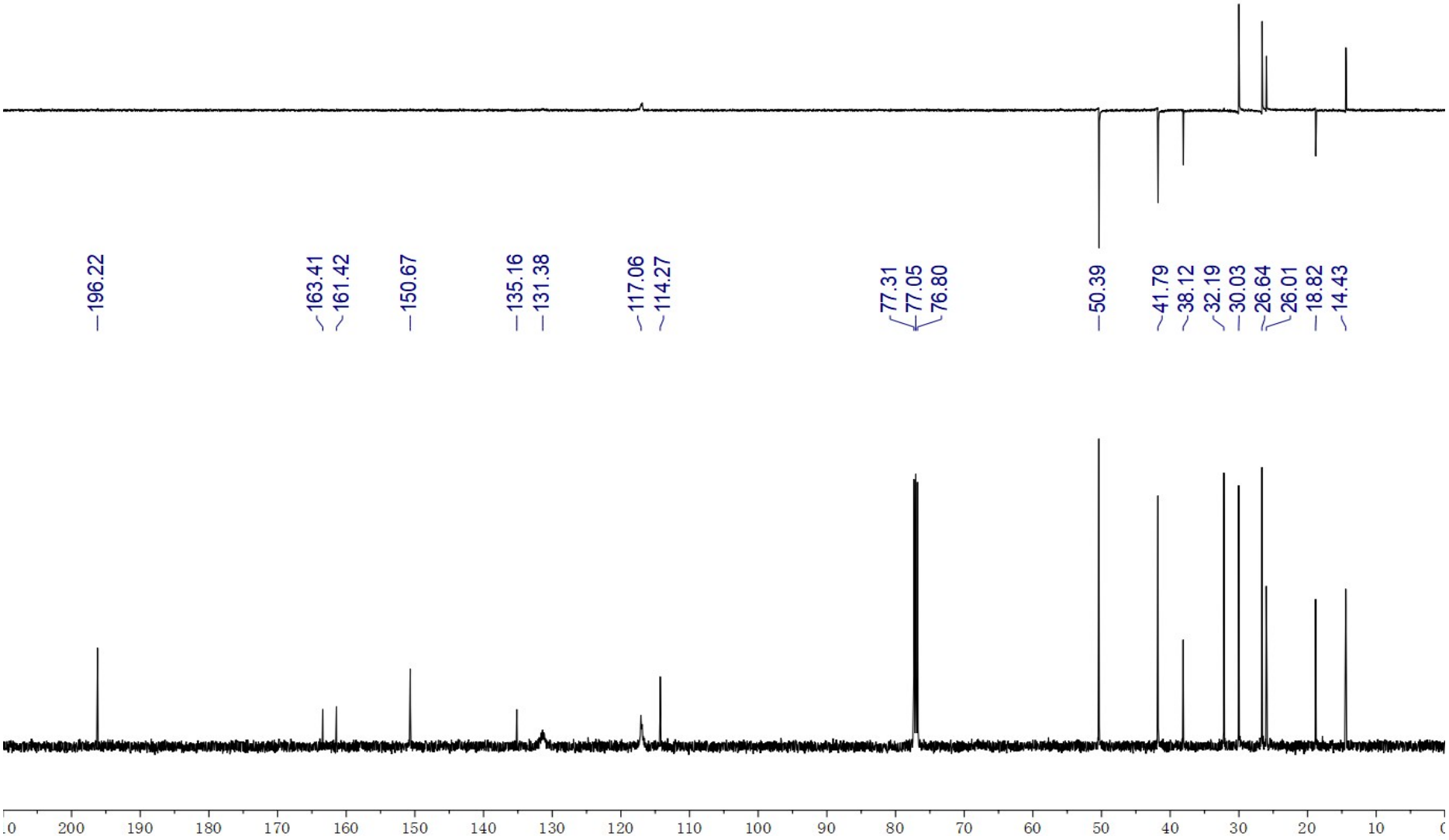


Figure 36. ^{1}H NMR (125 MHz, $CDCl_3$) spectra of compound **3r**

^{1}H NMR and ^{13}C NMR spectra for compound **4**

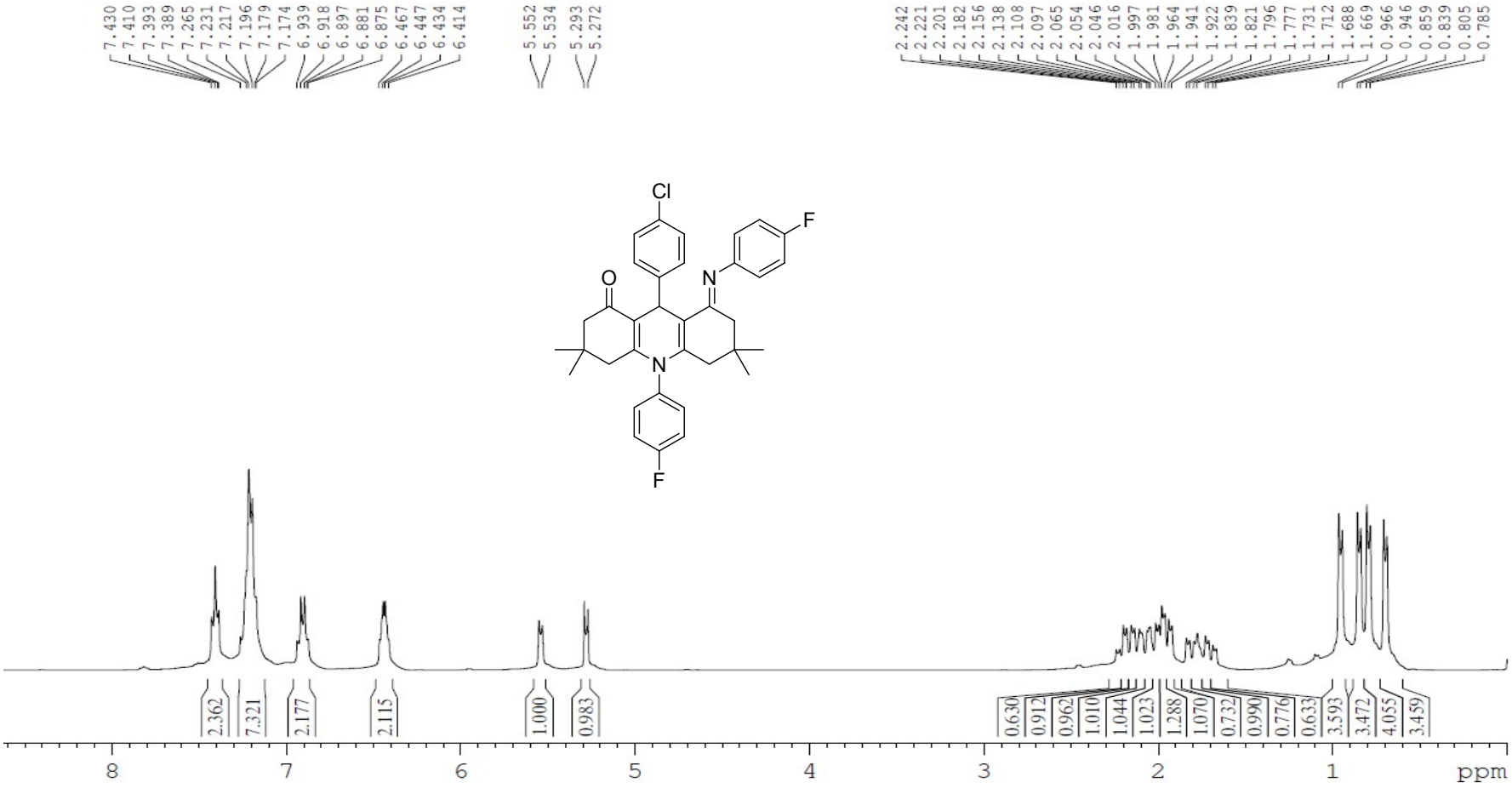


Figure 37. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4a**

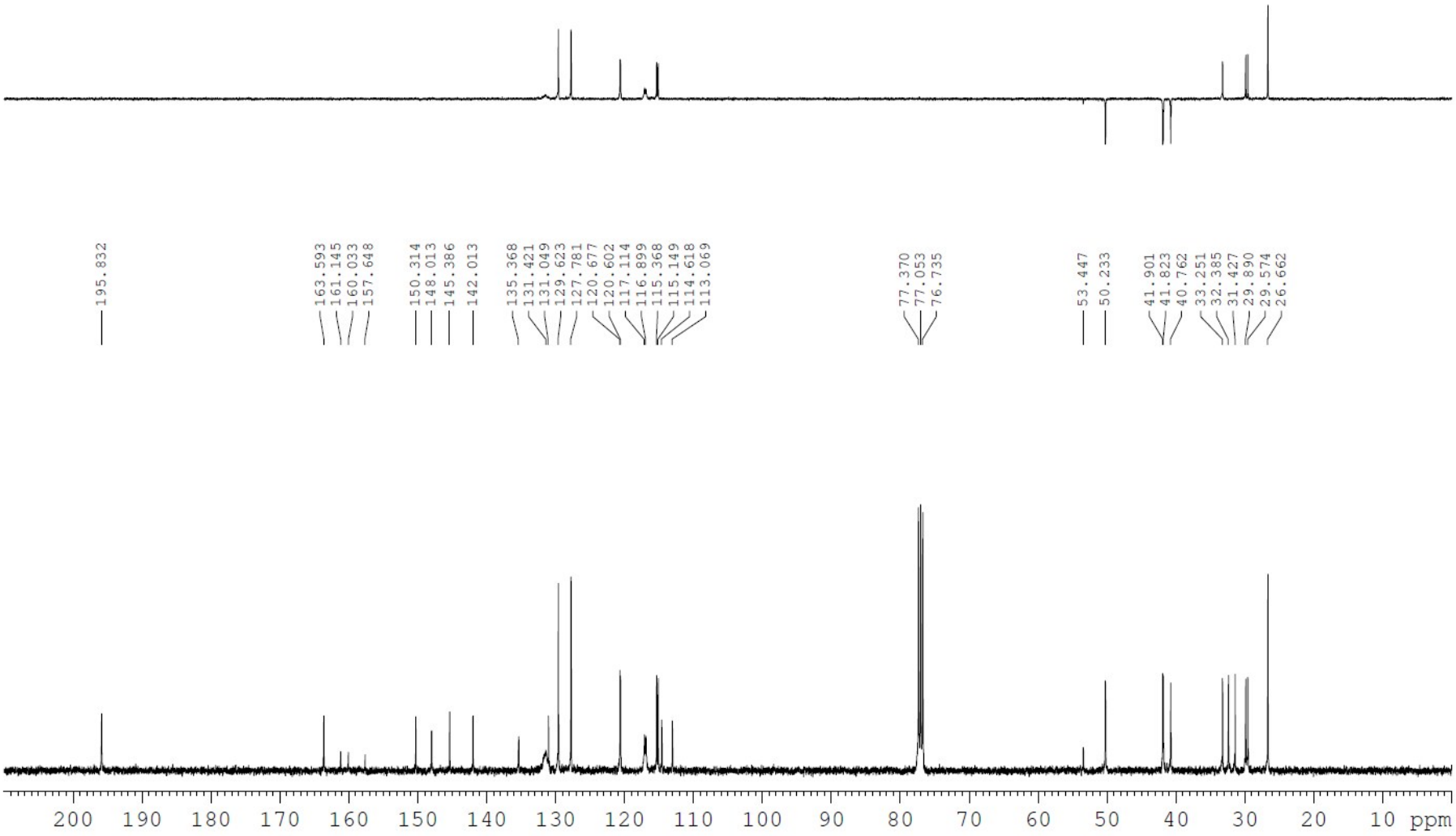


Figure 38. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4a**

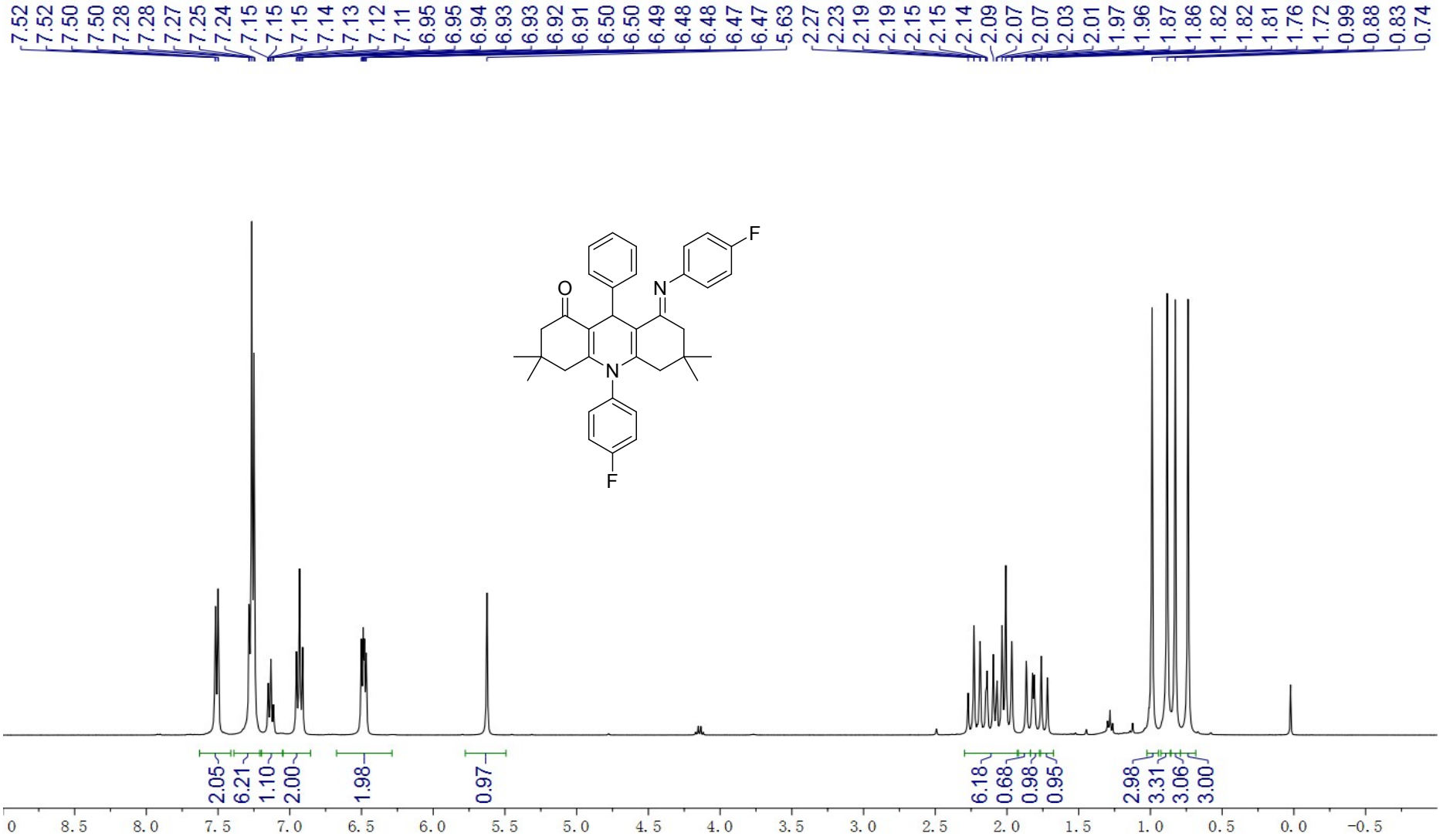


Figure 39. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4b**

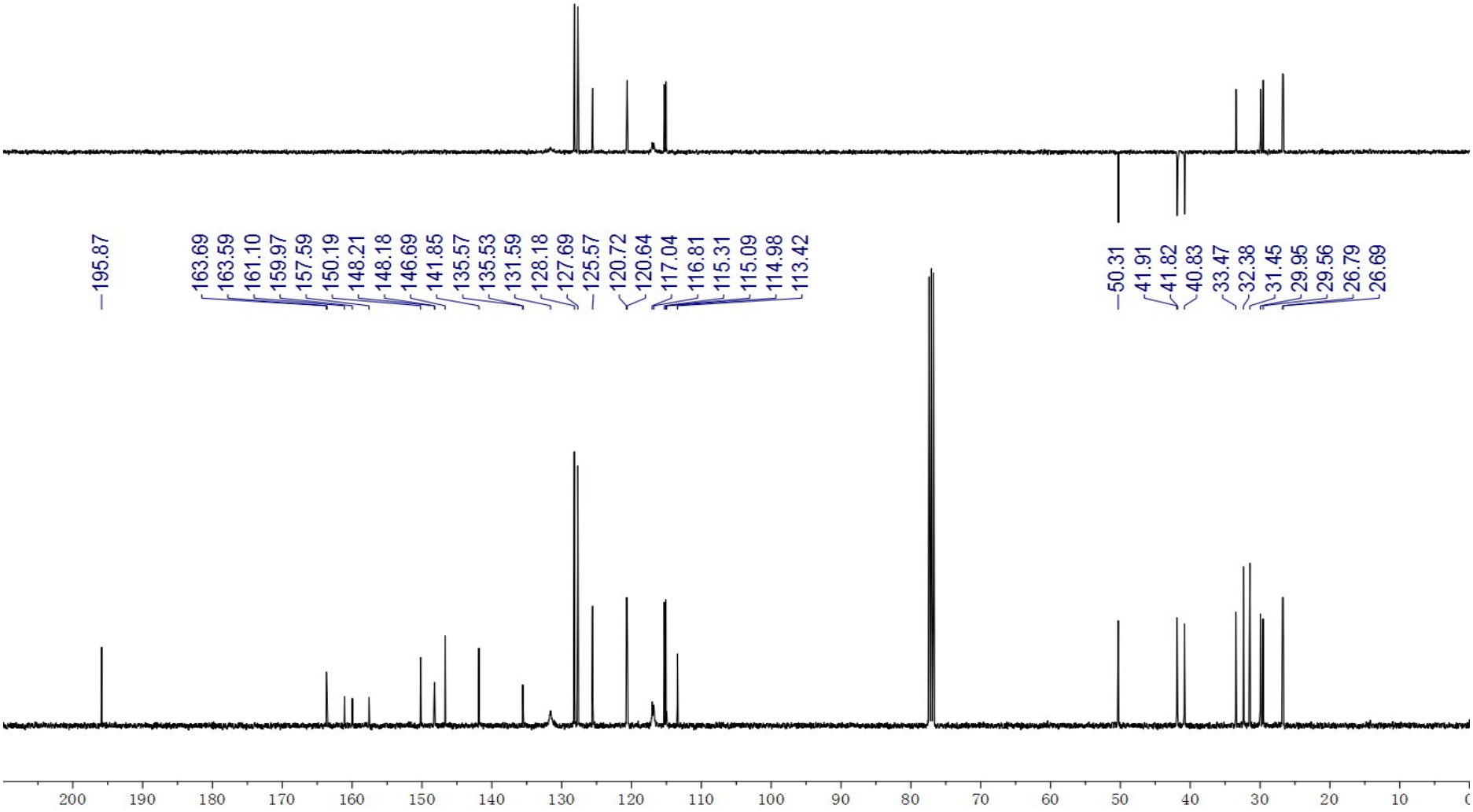


Figure 40. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4b**

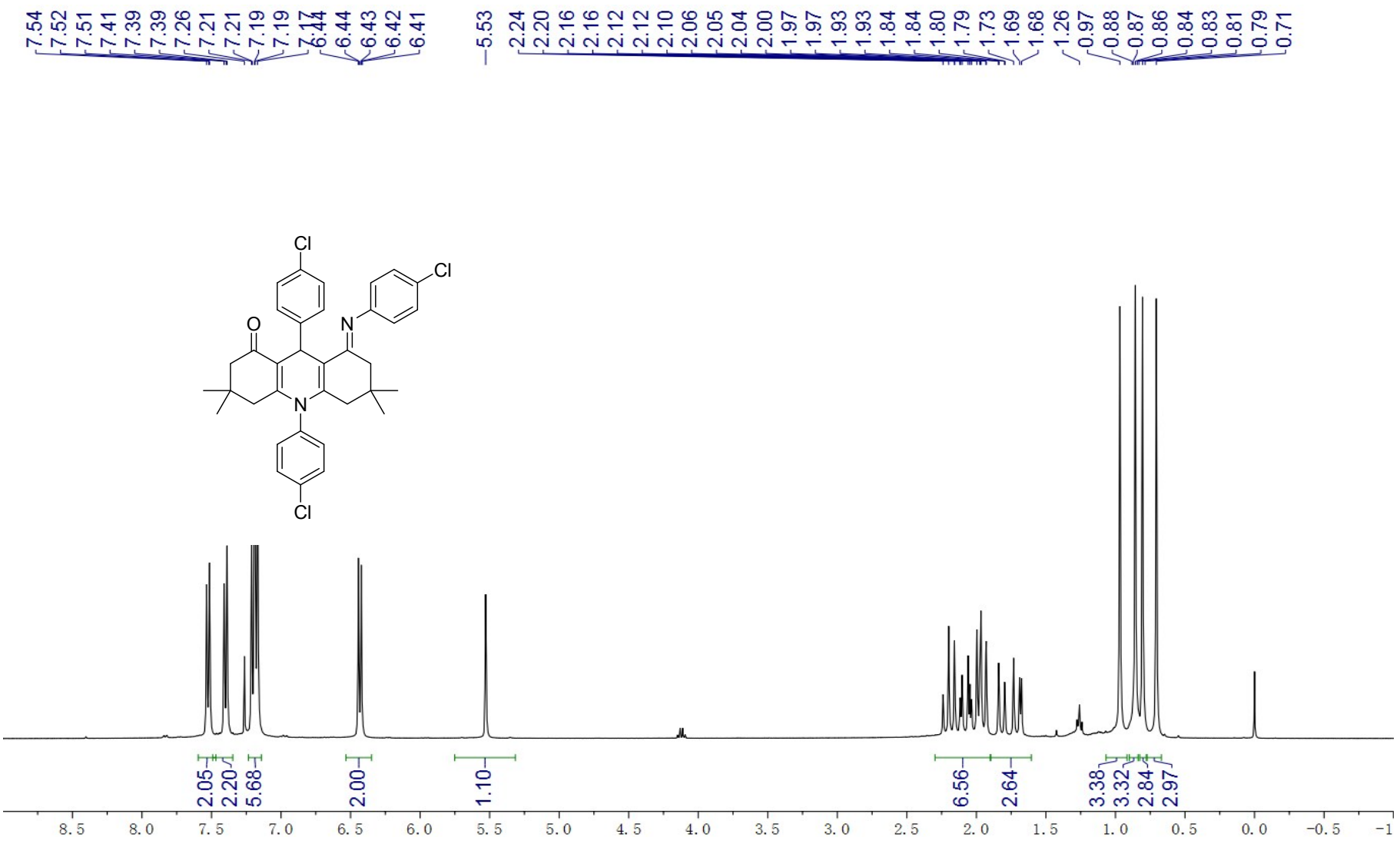


Figure 41. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4c**

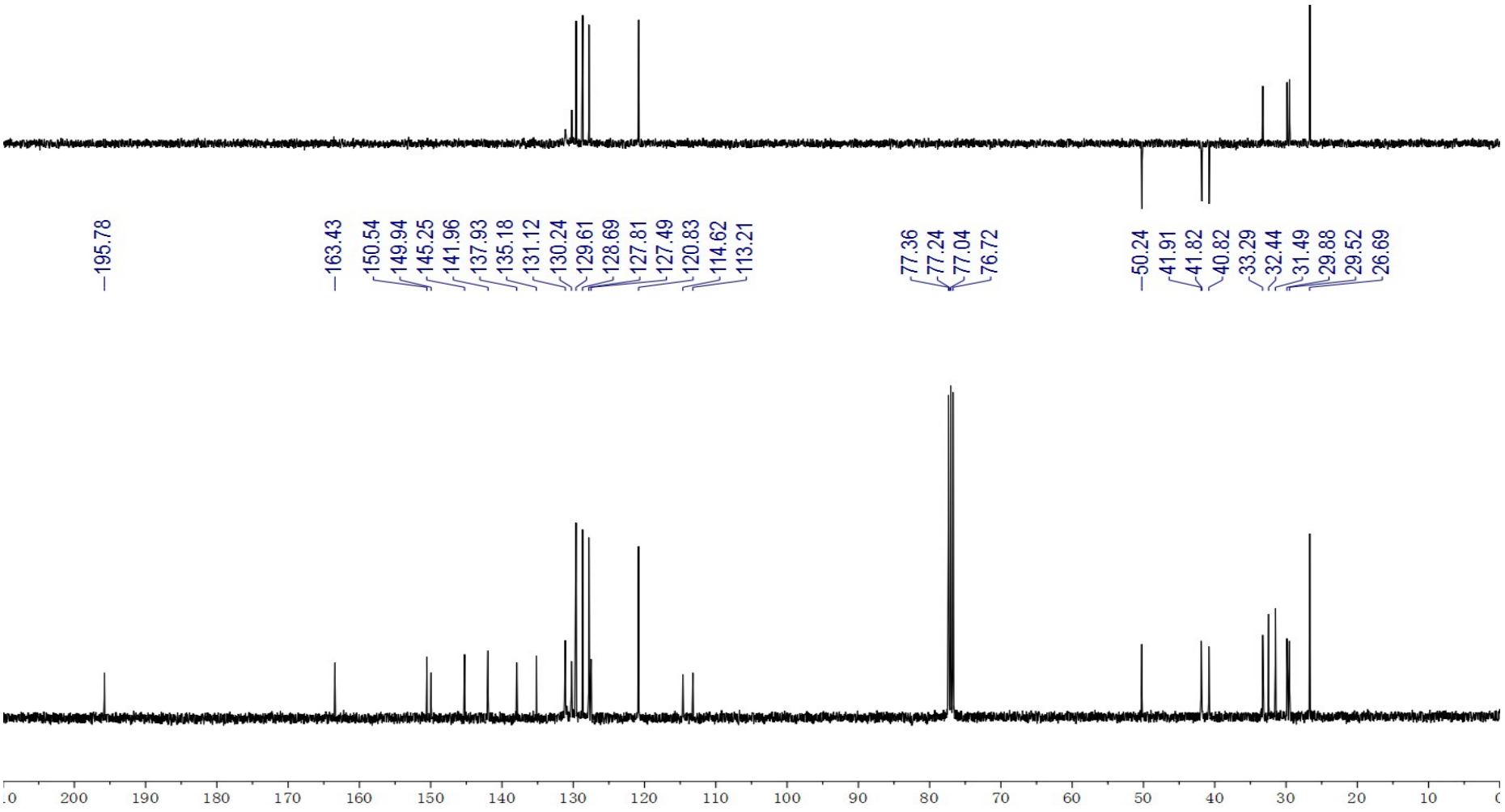


Figure 42. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4c**

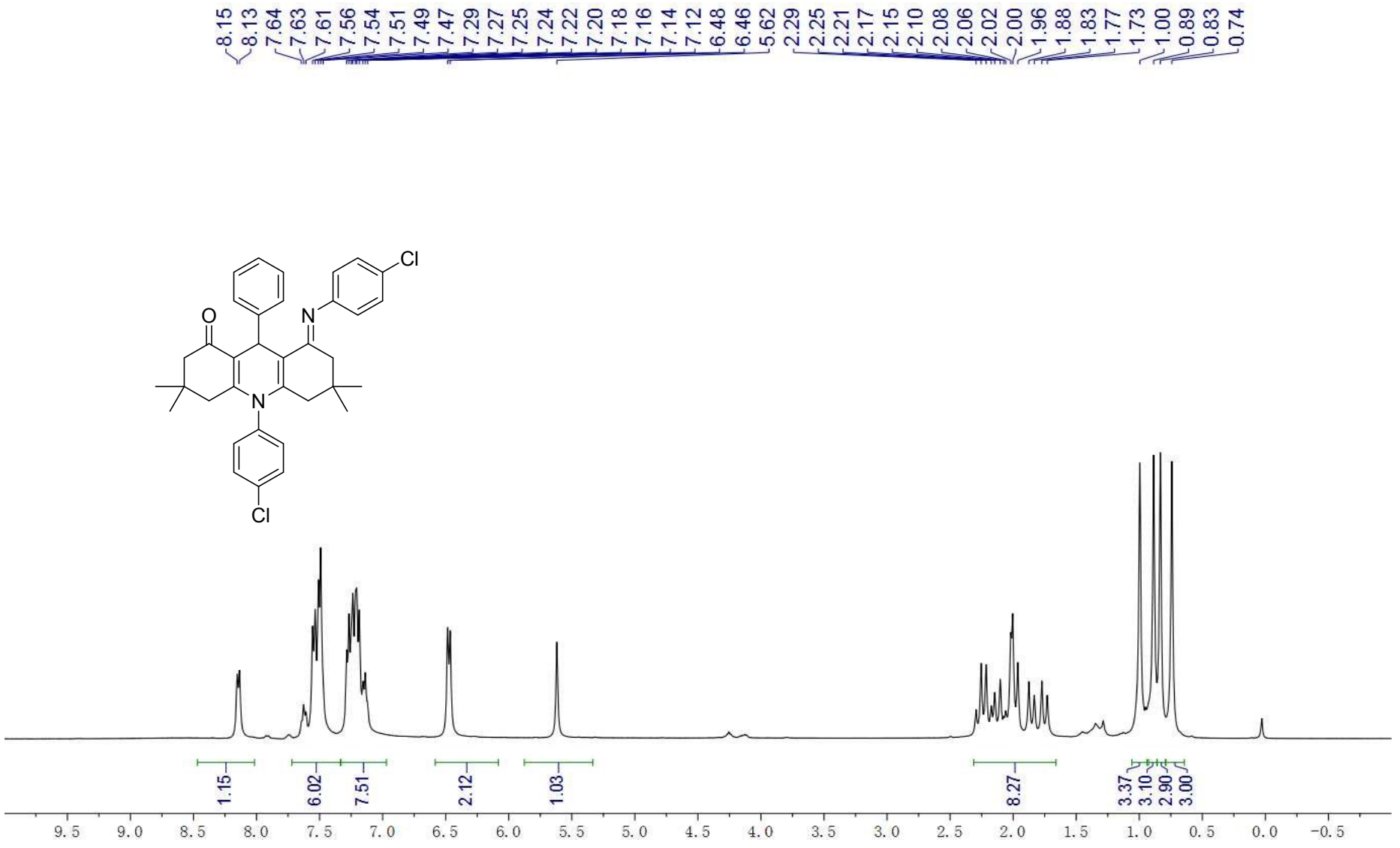


Figure 43. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4d**

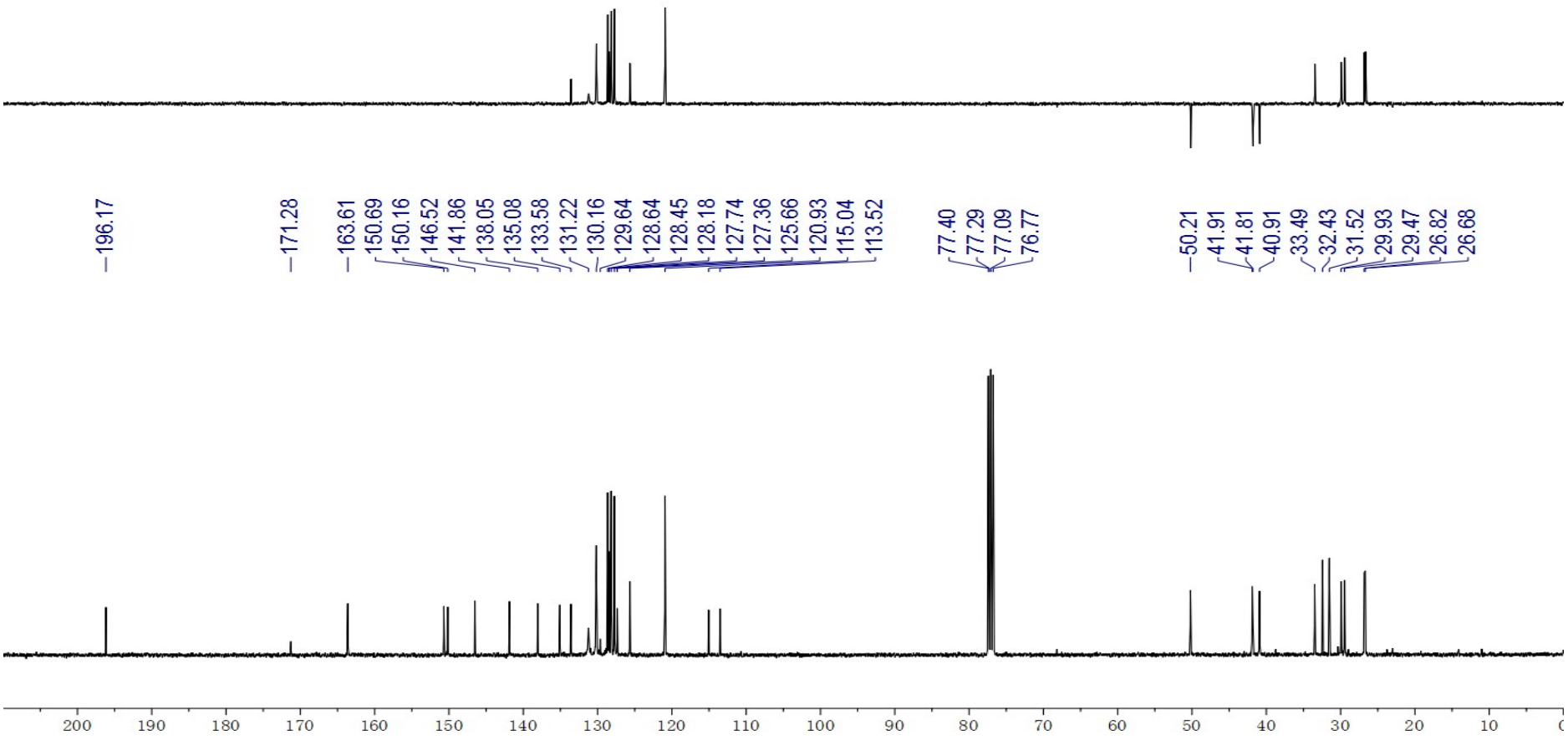


Figure 44. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4d**

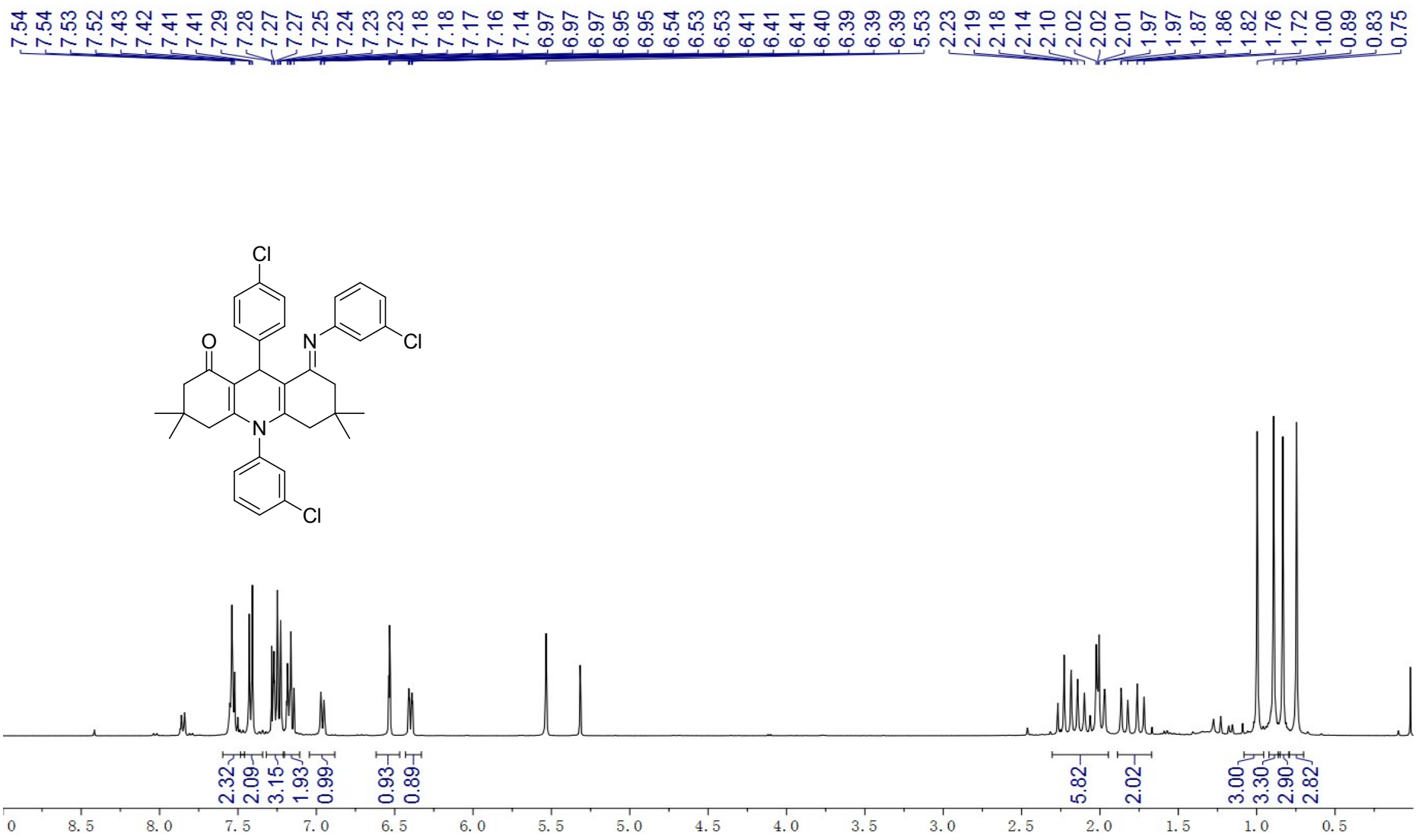


Figure 45. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4e**

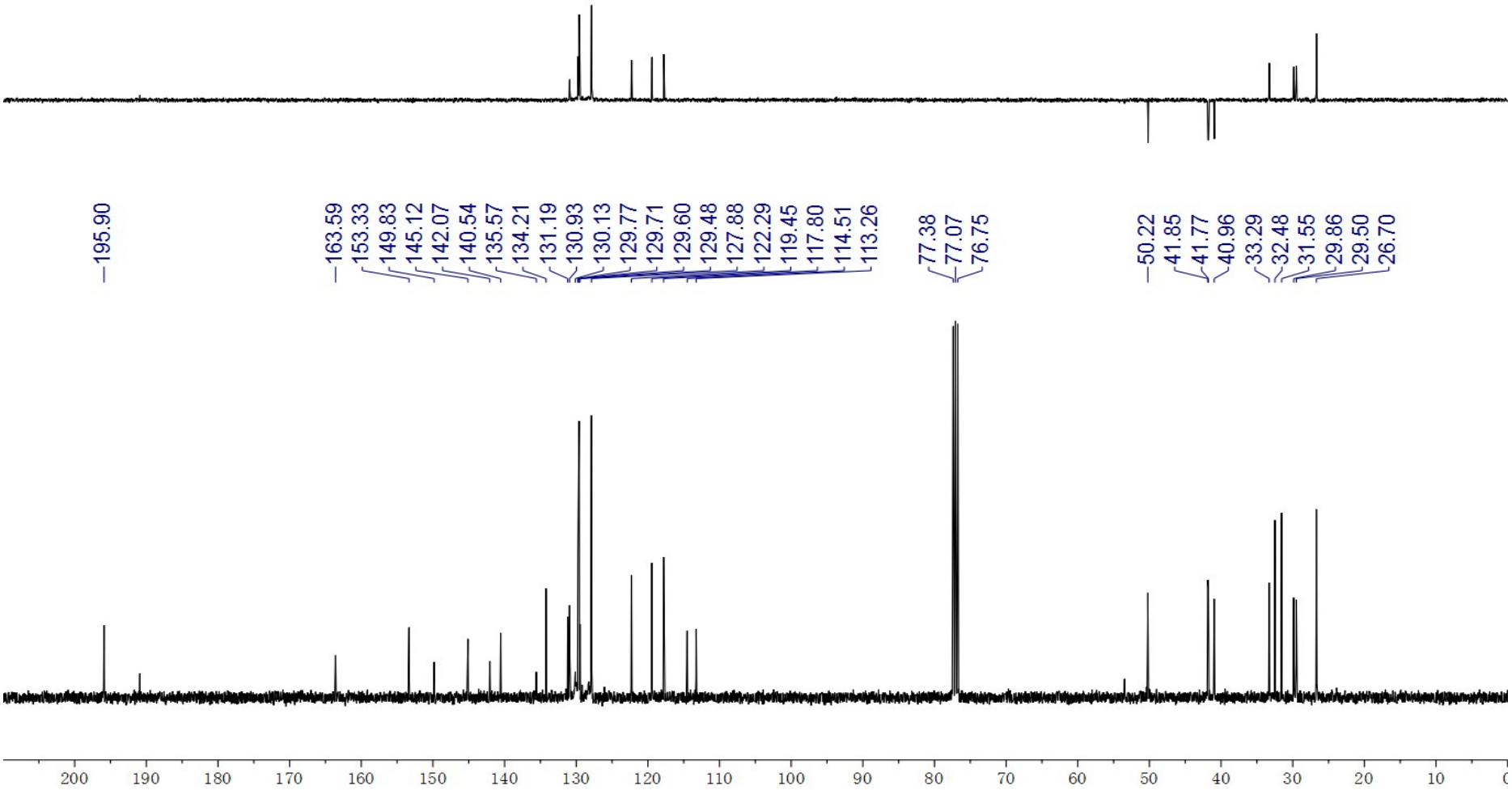


Figure 46. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4e**

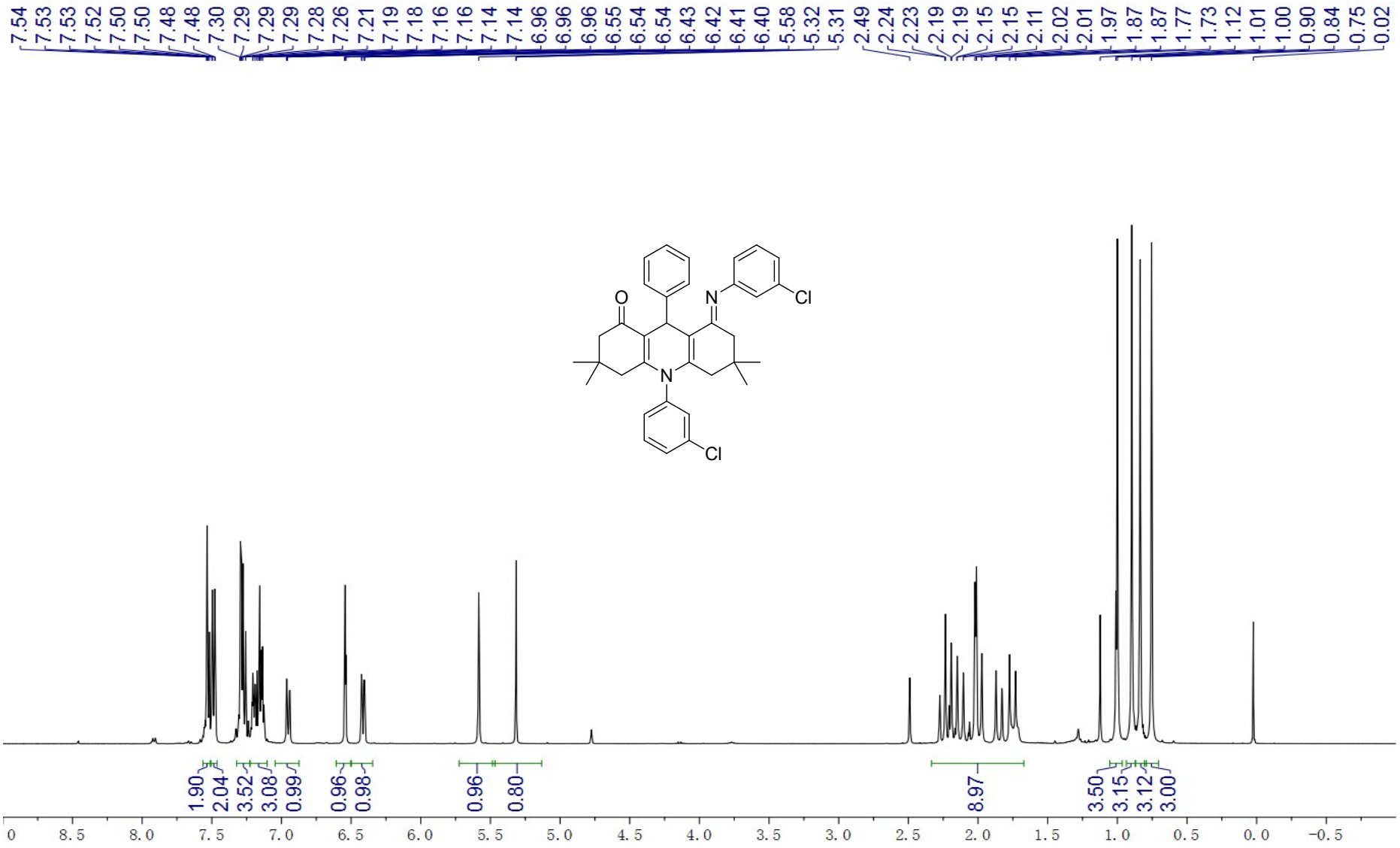


Figure 47. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4f**

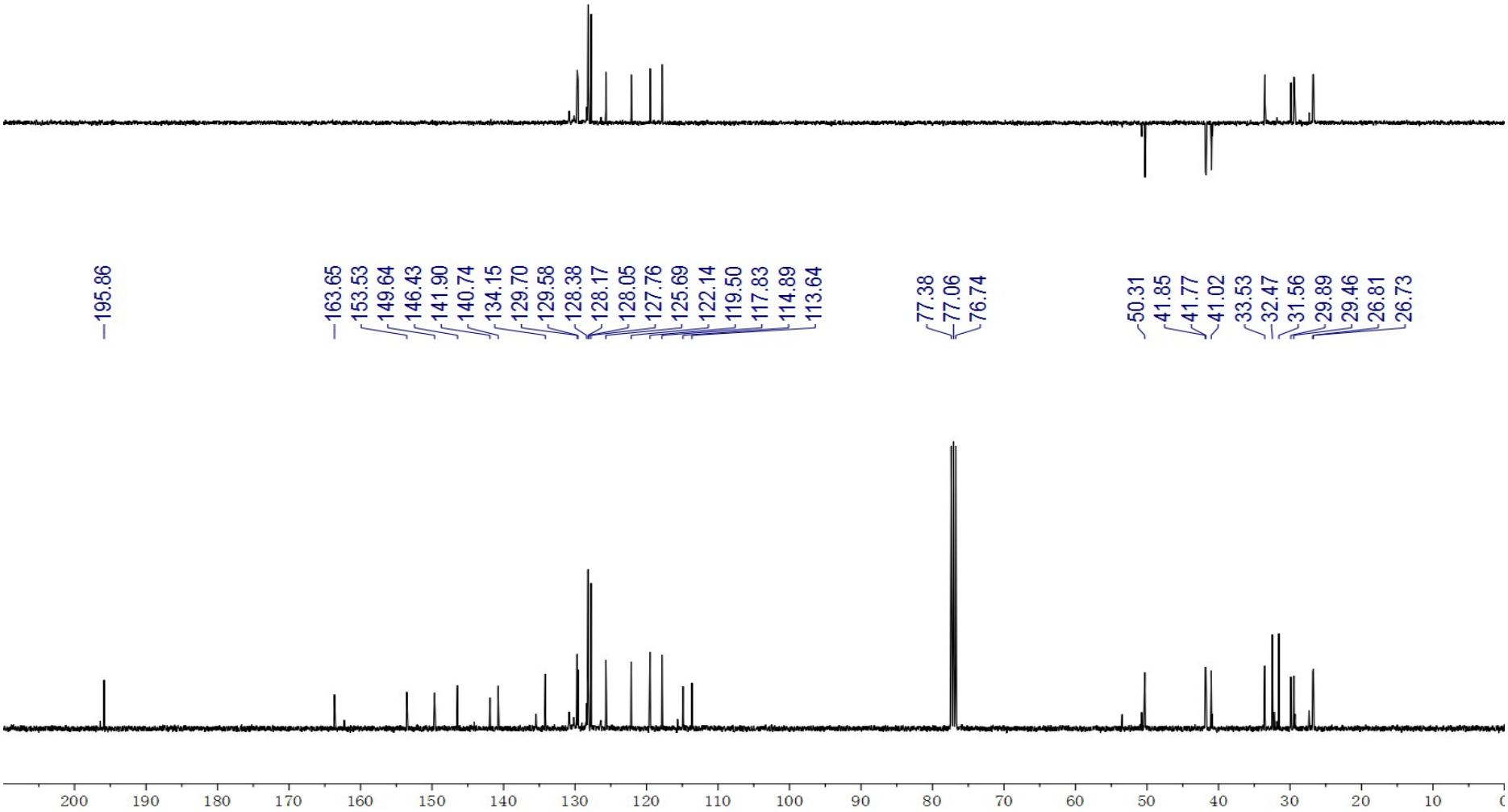


Figure 48. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4f**

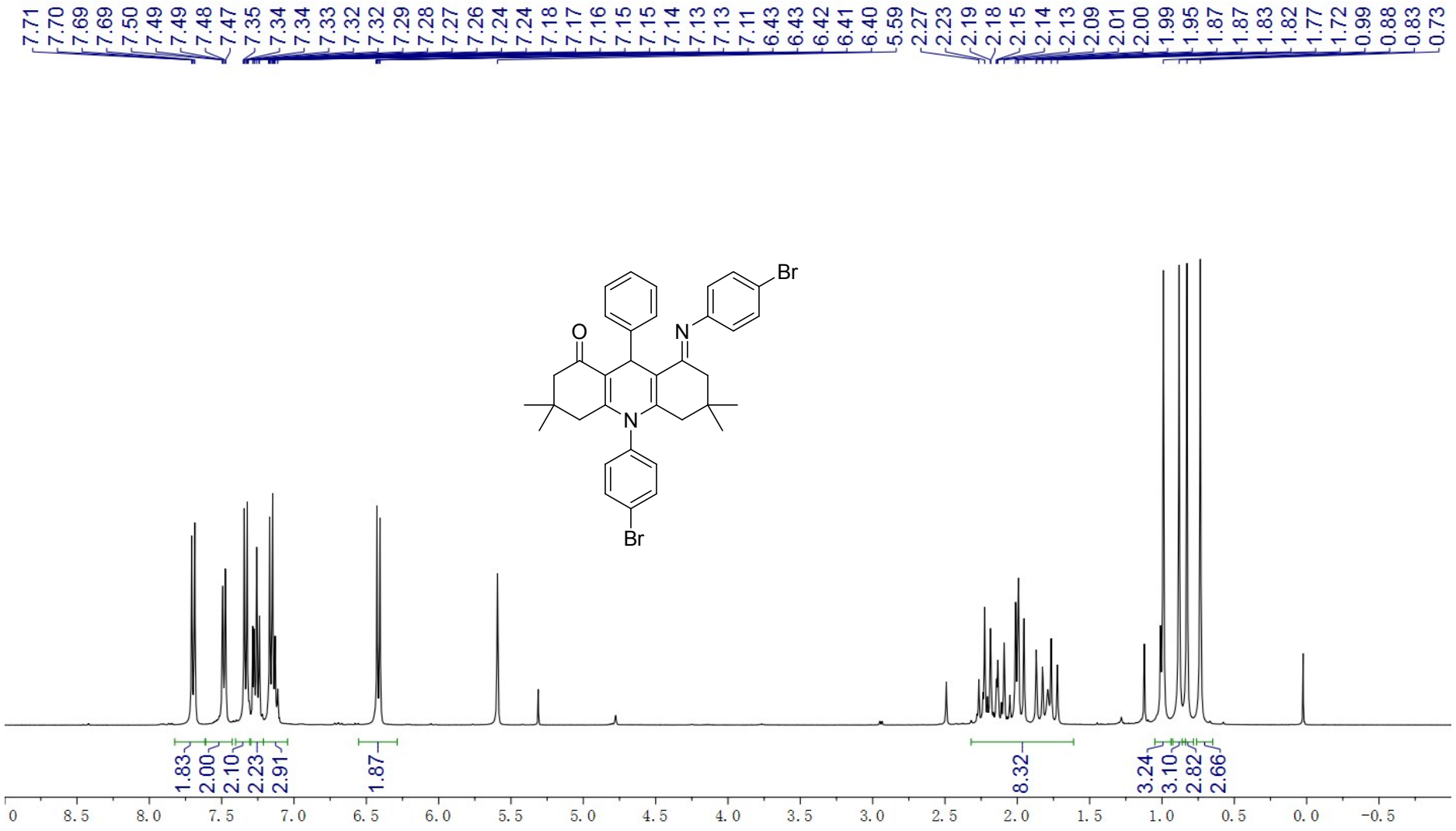


Figure 49. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4g**

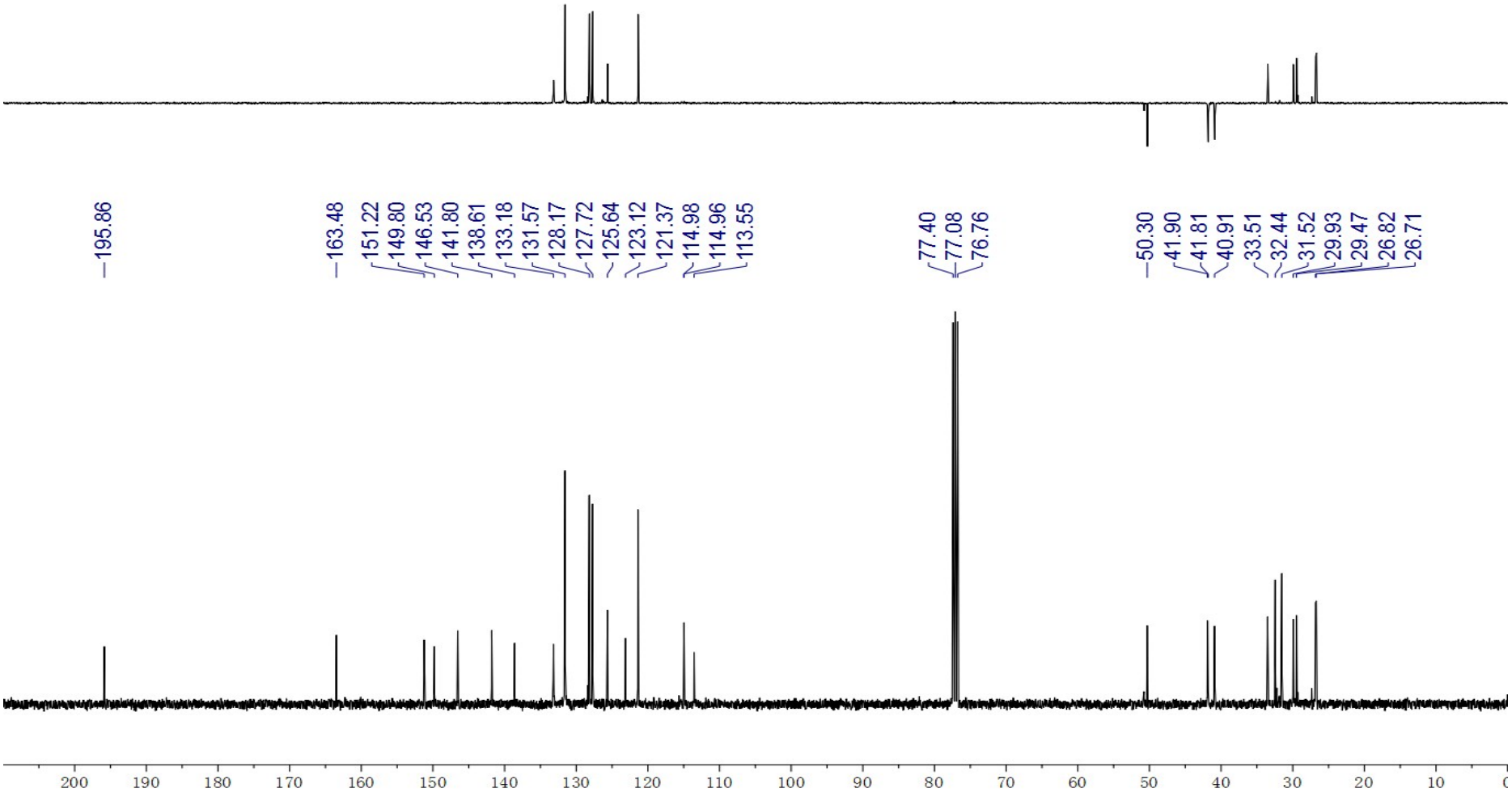


Figure 50. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4g**

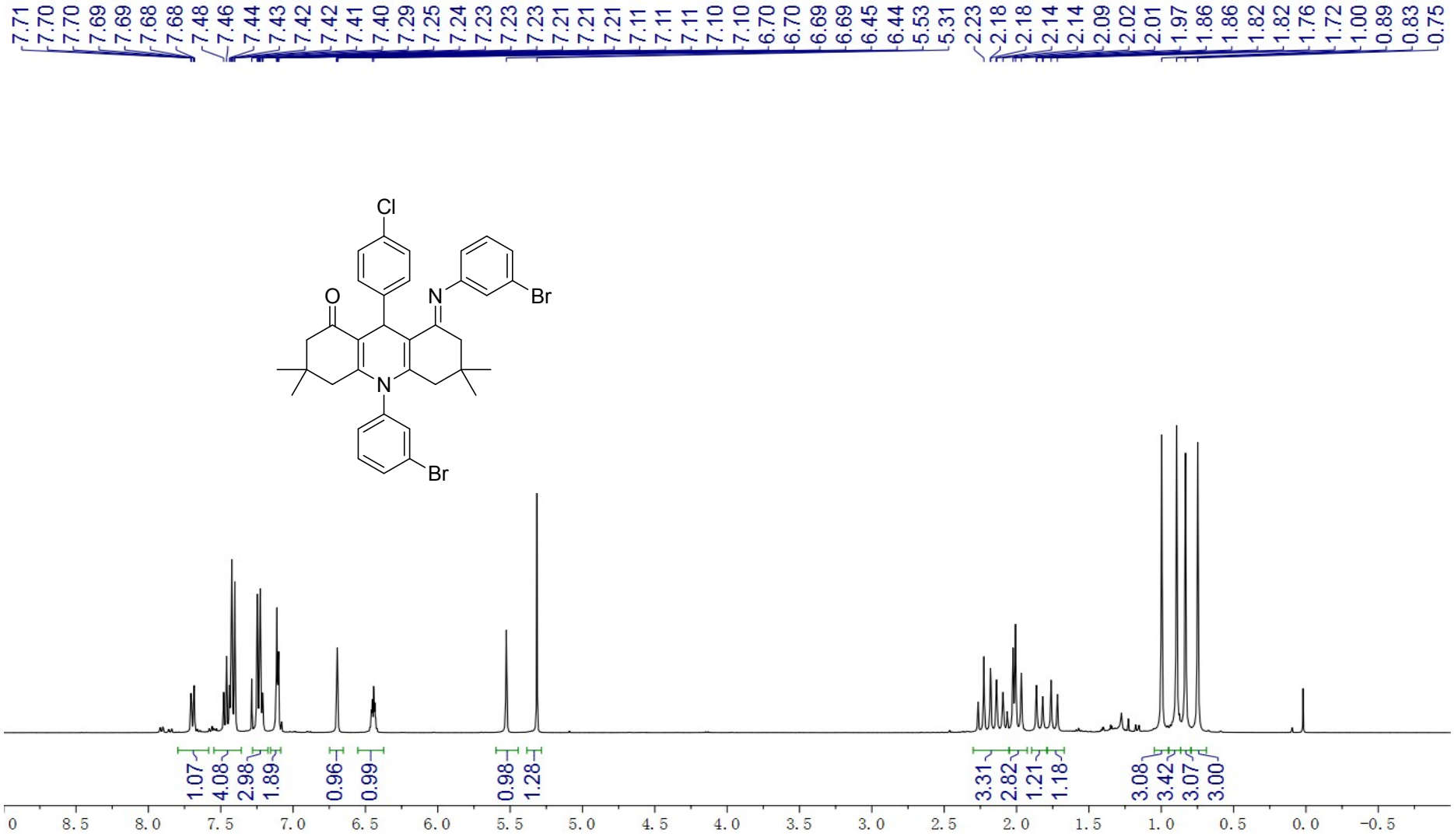


Figure 51. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4h**

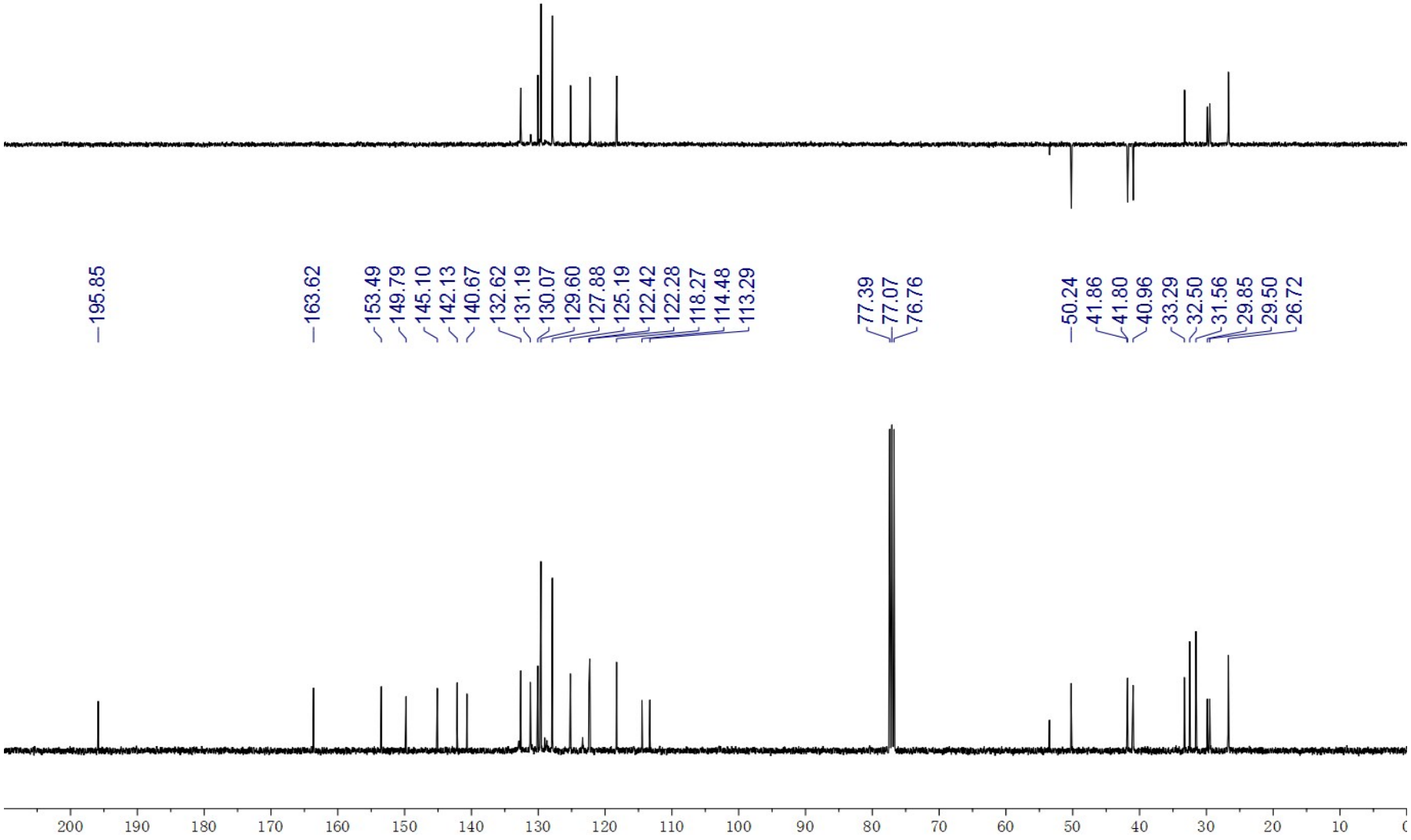


Figure 52. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4h**

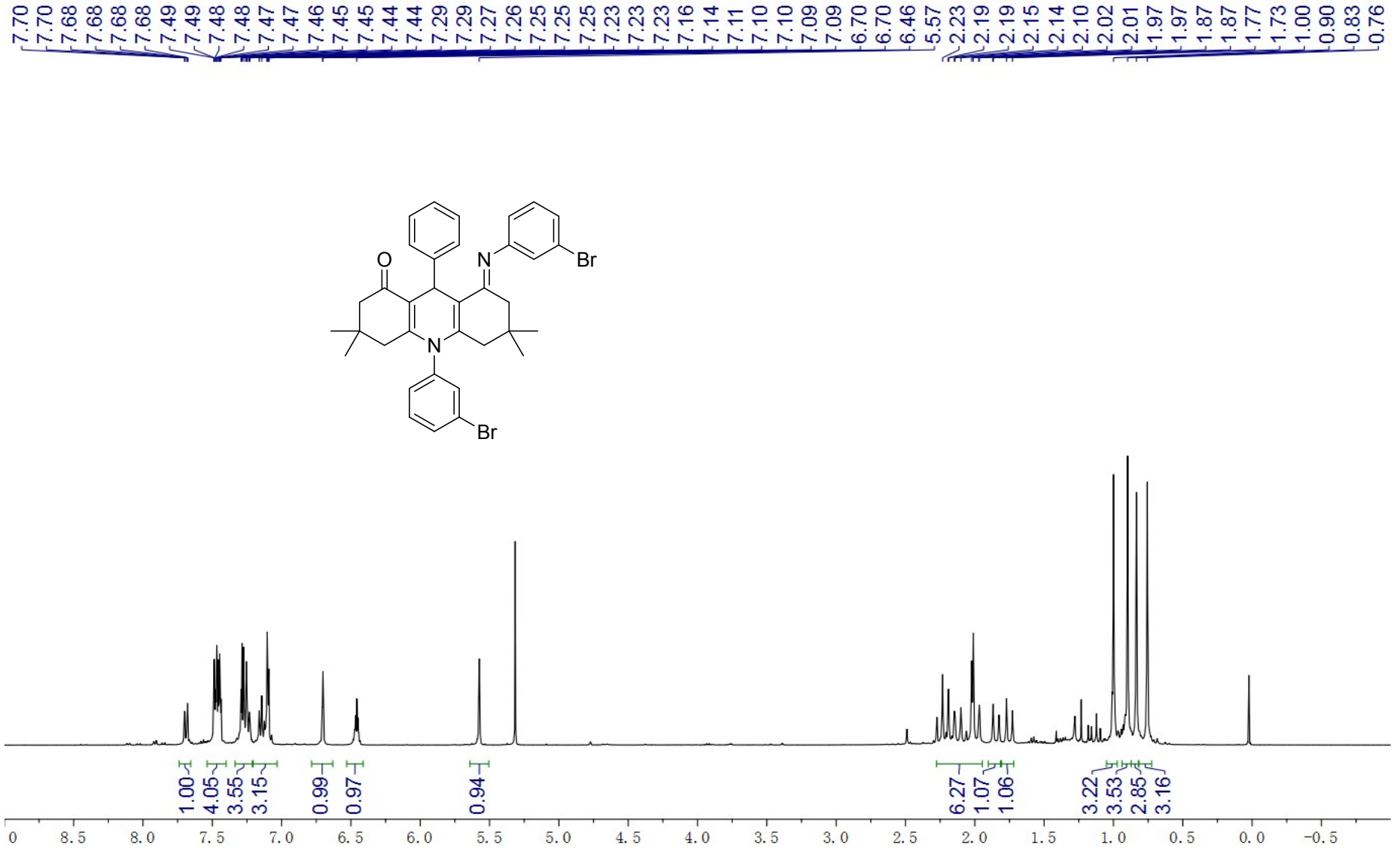


Figure 53. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4i**

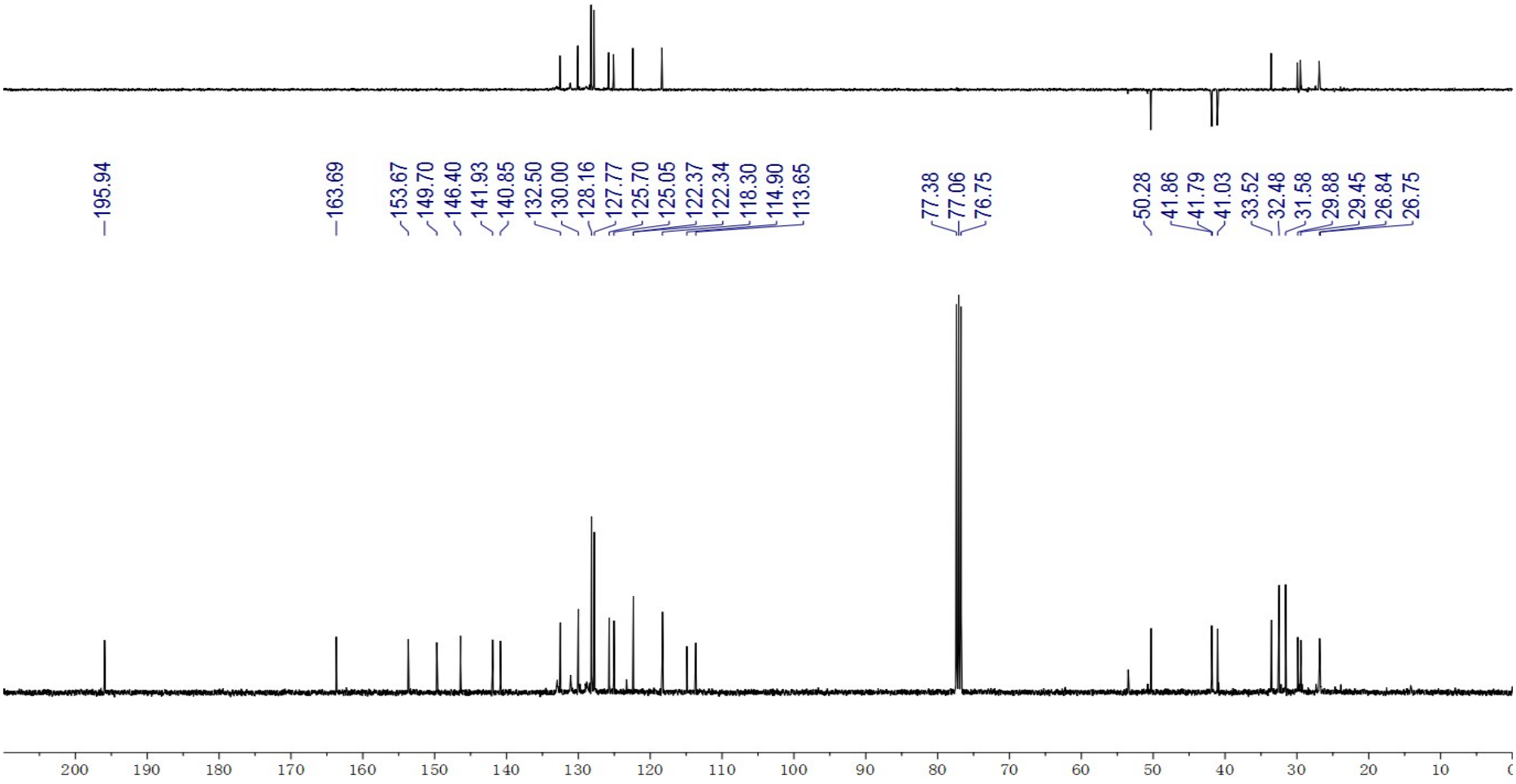


Figure 54. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4i**

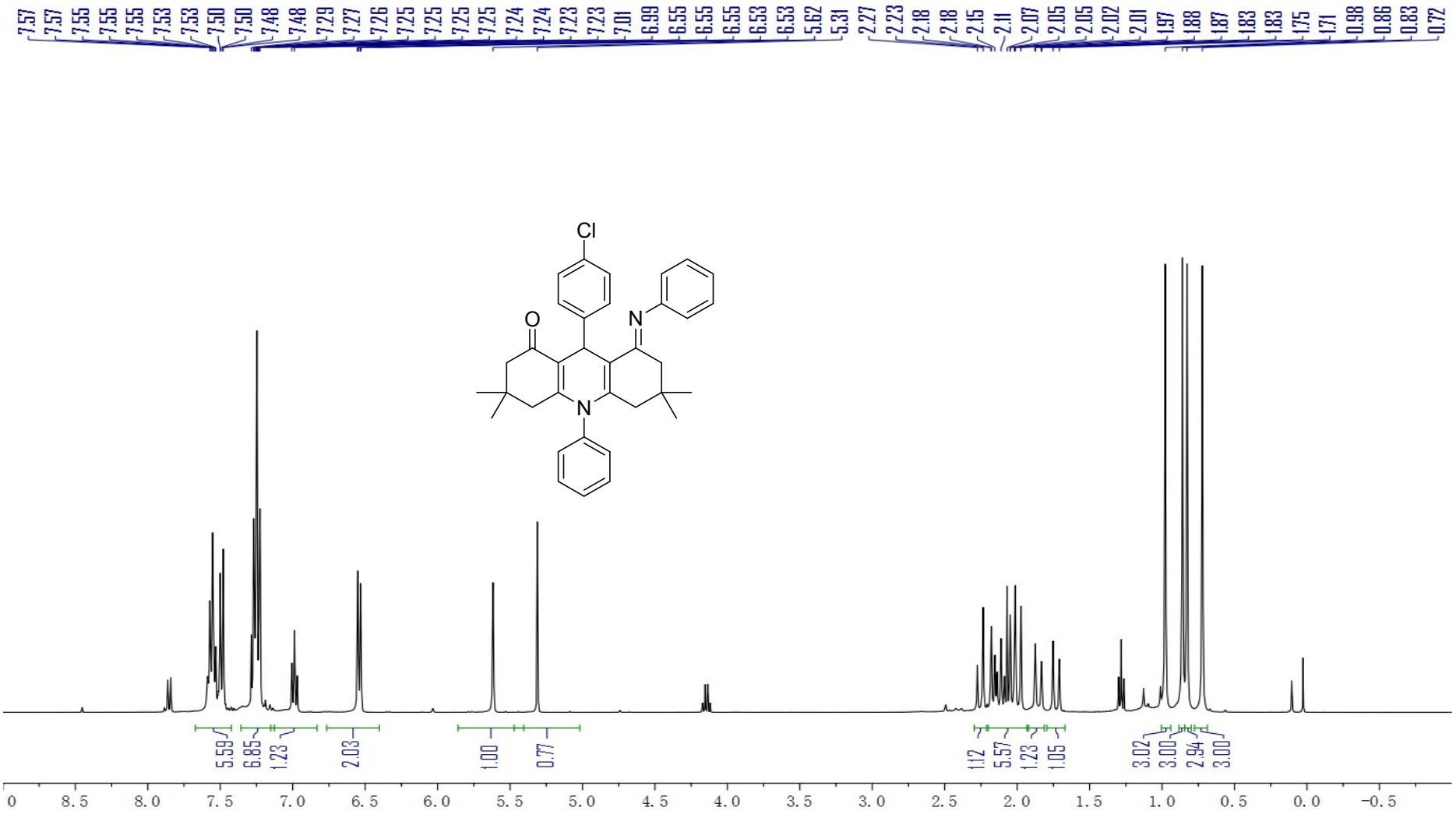


Figure 55. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4j**

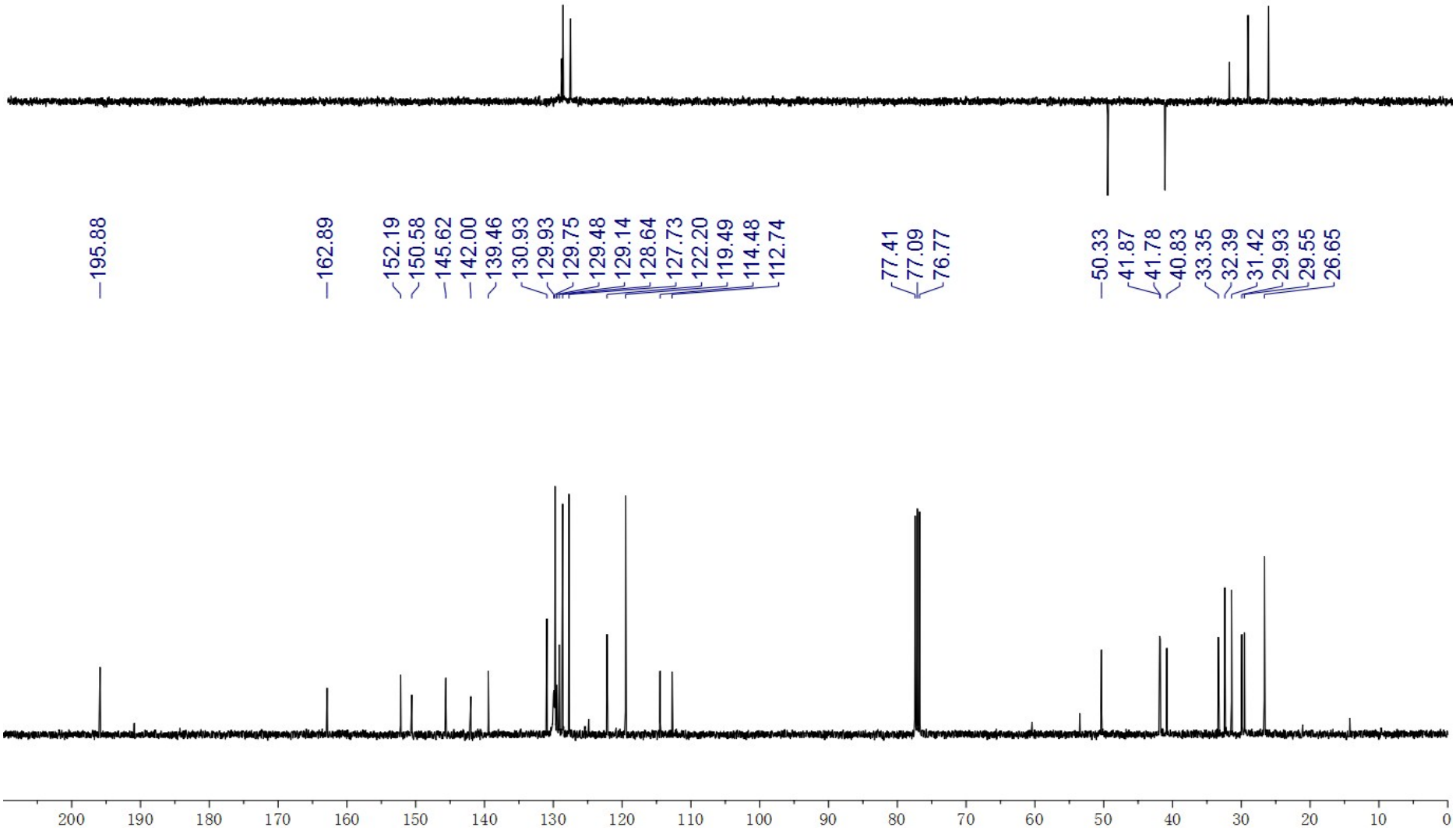


Figure 56. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4j**

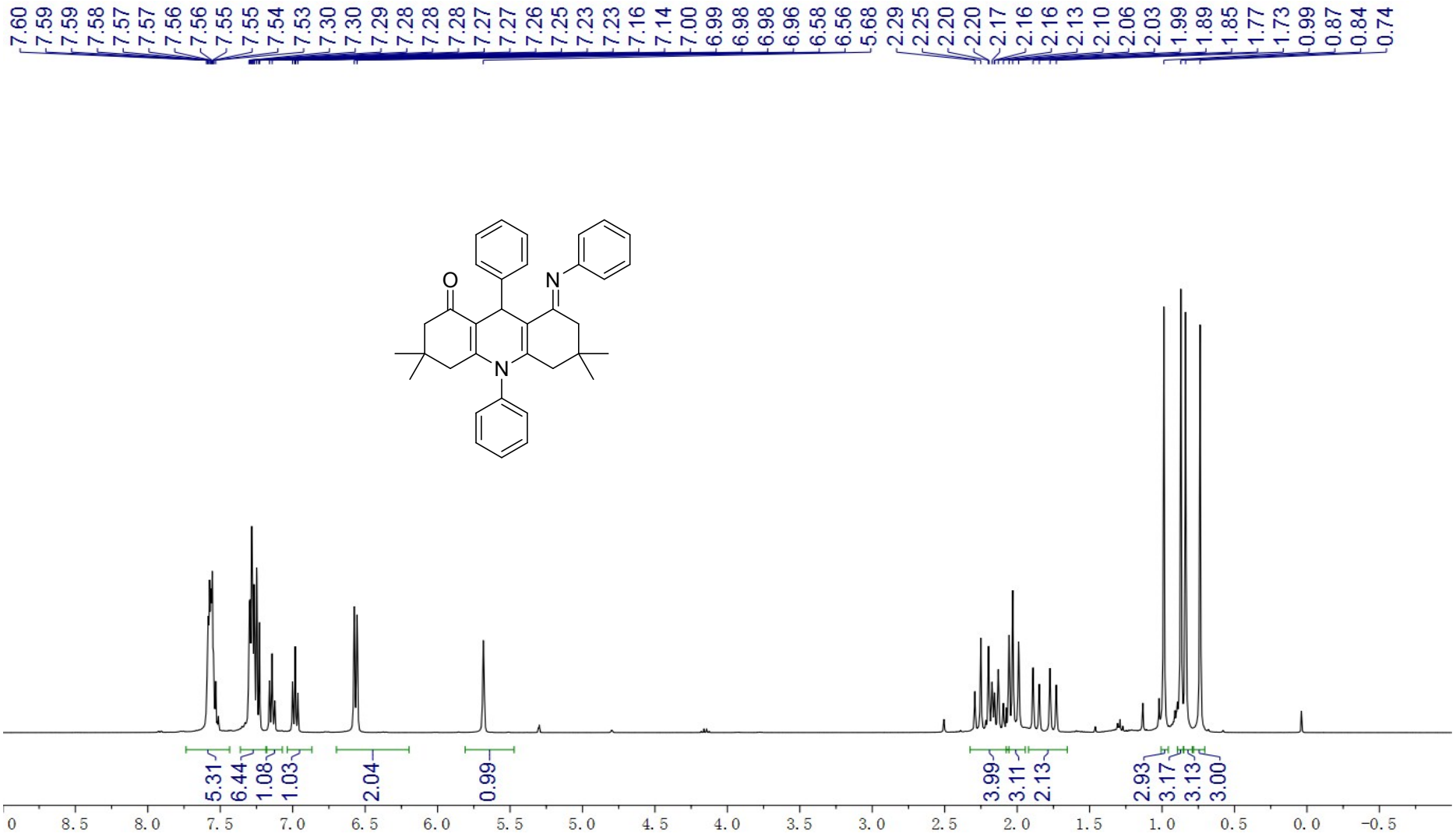


Figure 57. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4k**

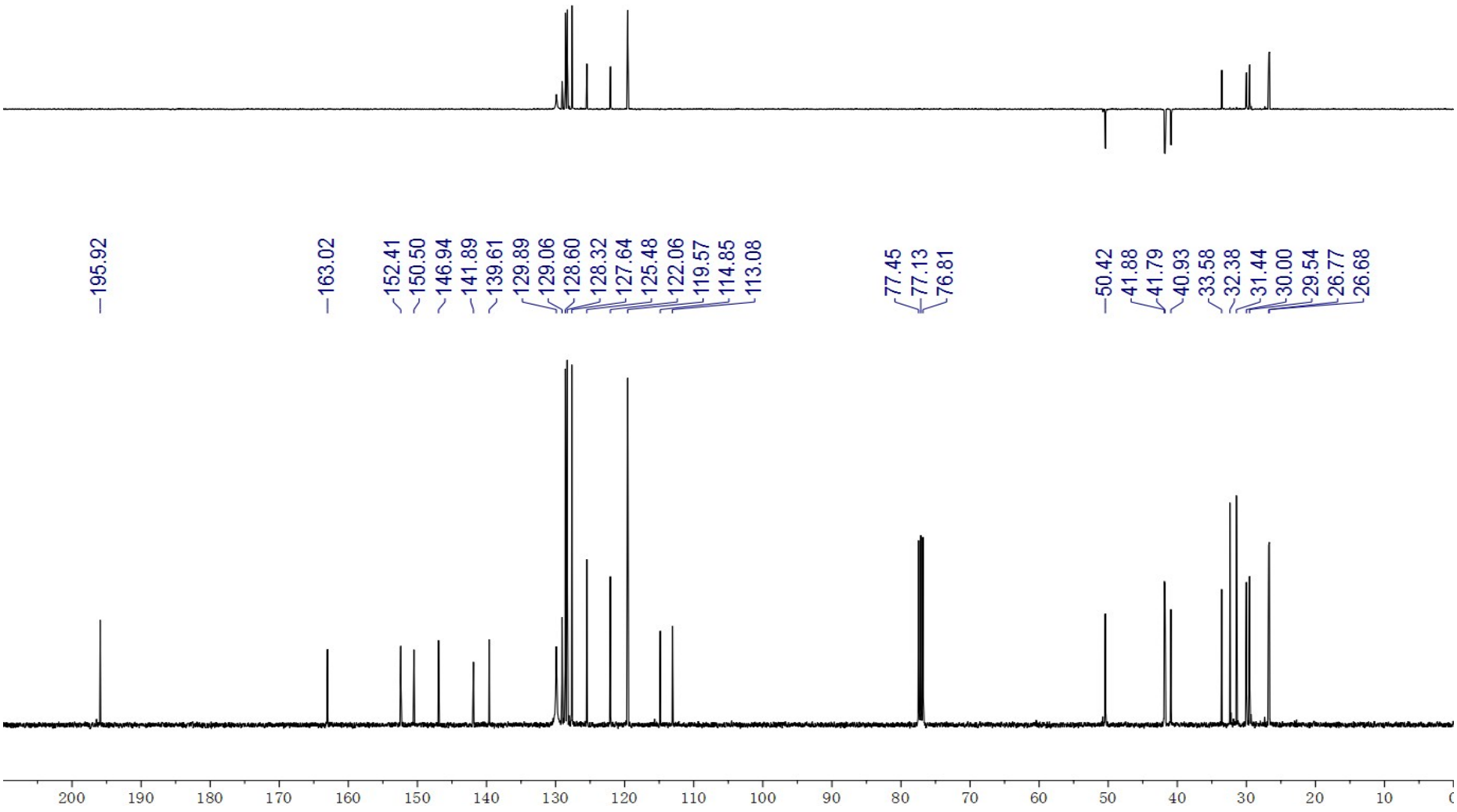


Figure 58. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4k**

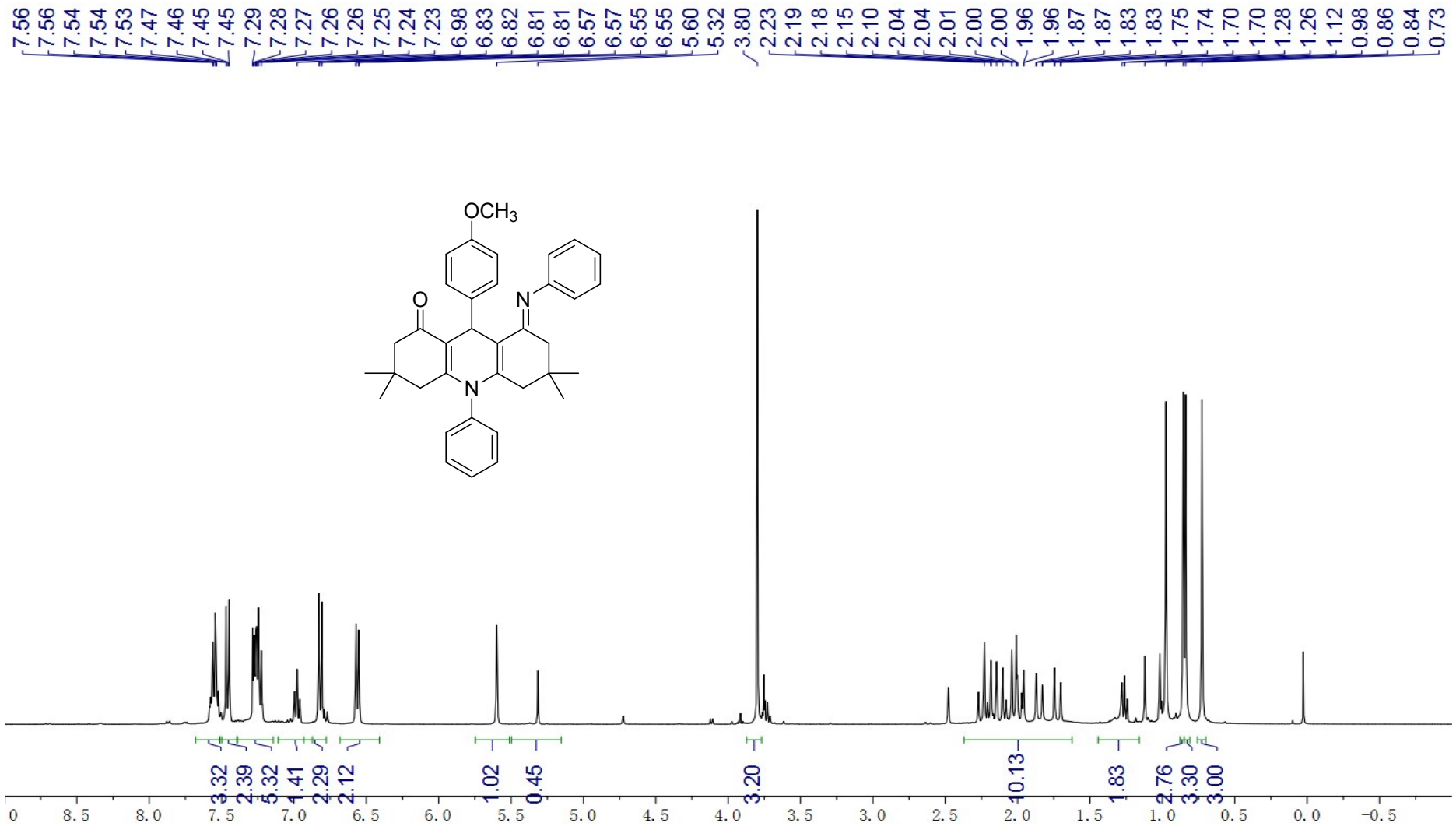


Figure 59. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4l**

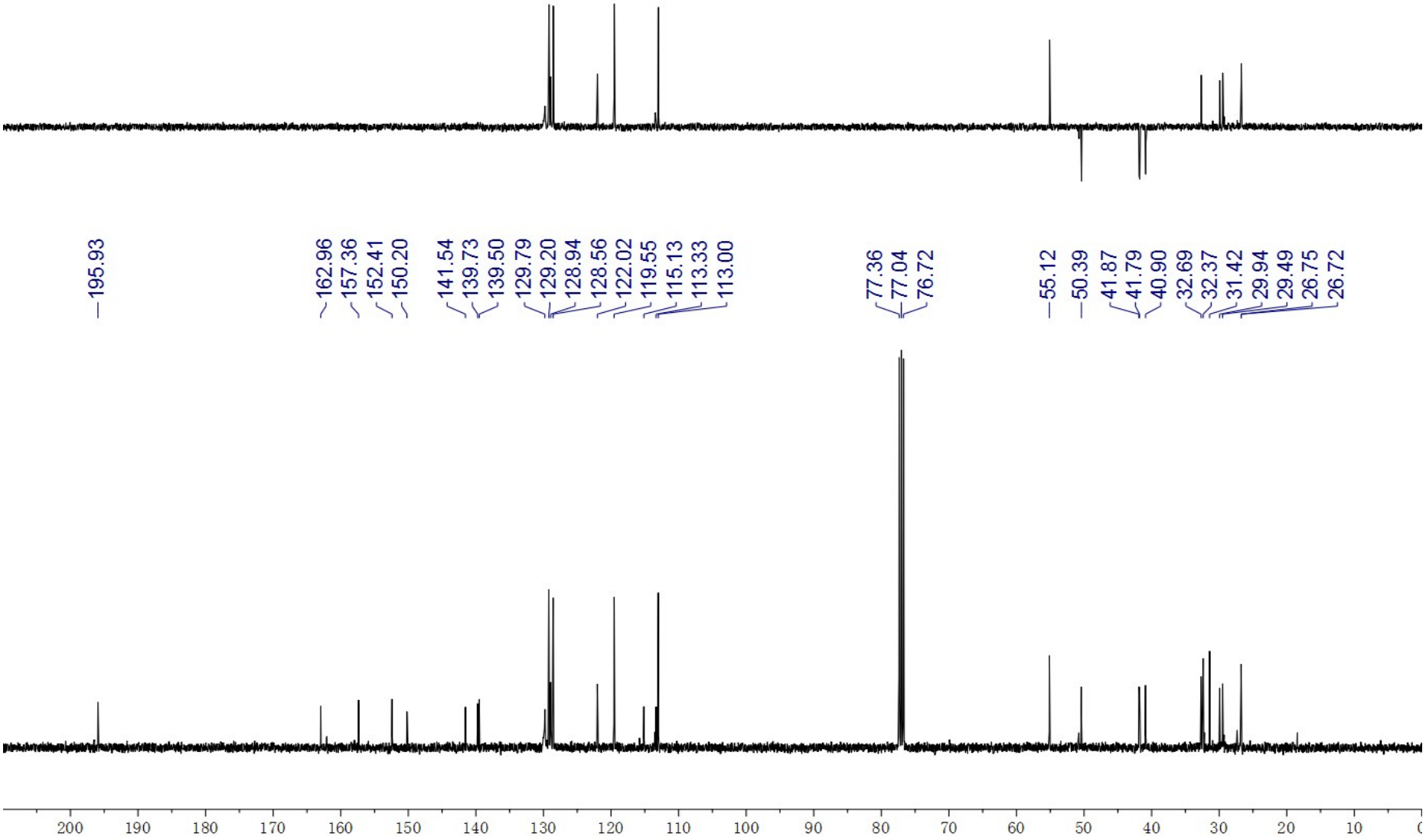


Figure 60. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4l**

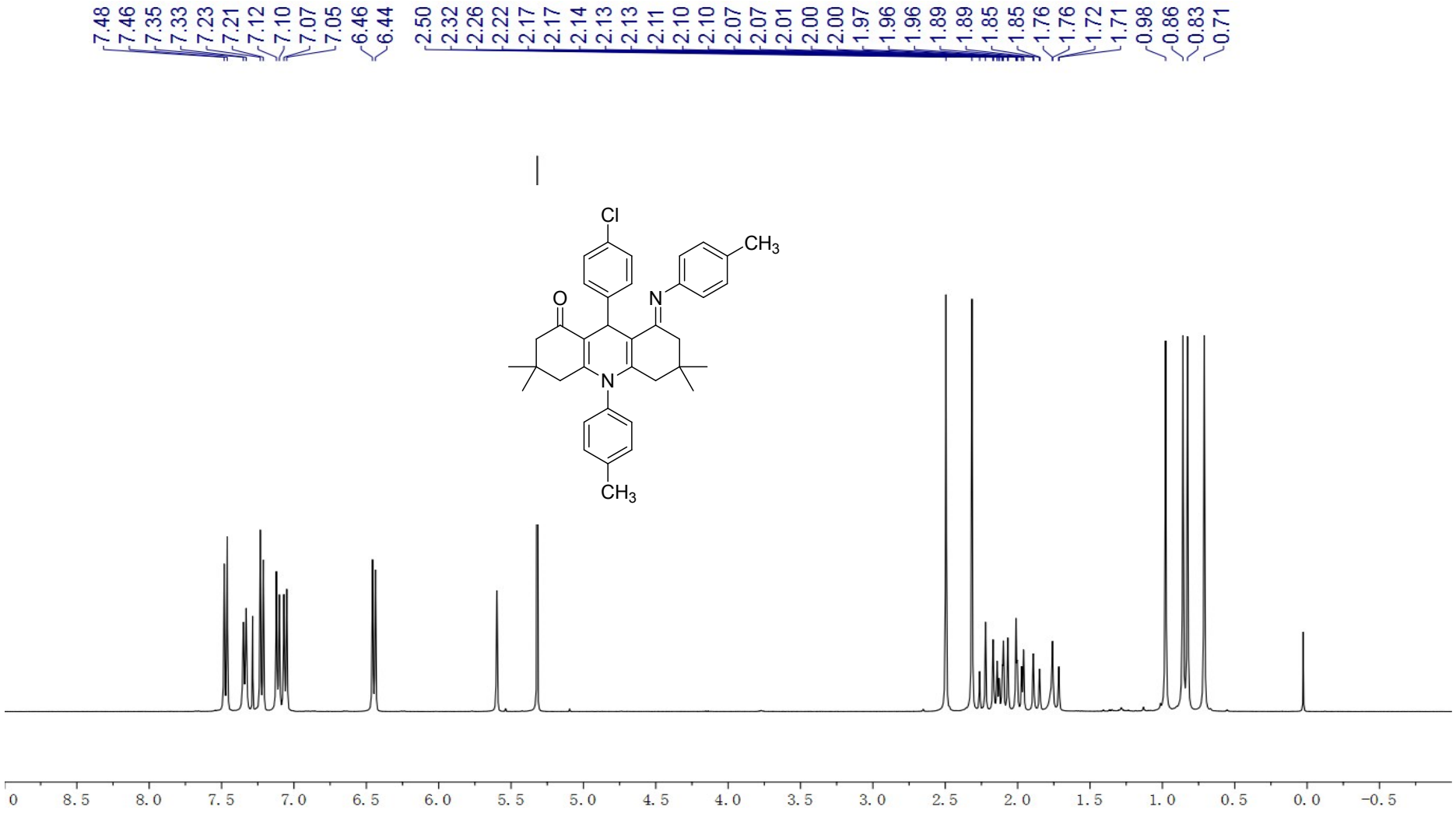


Figure 61. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4m**

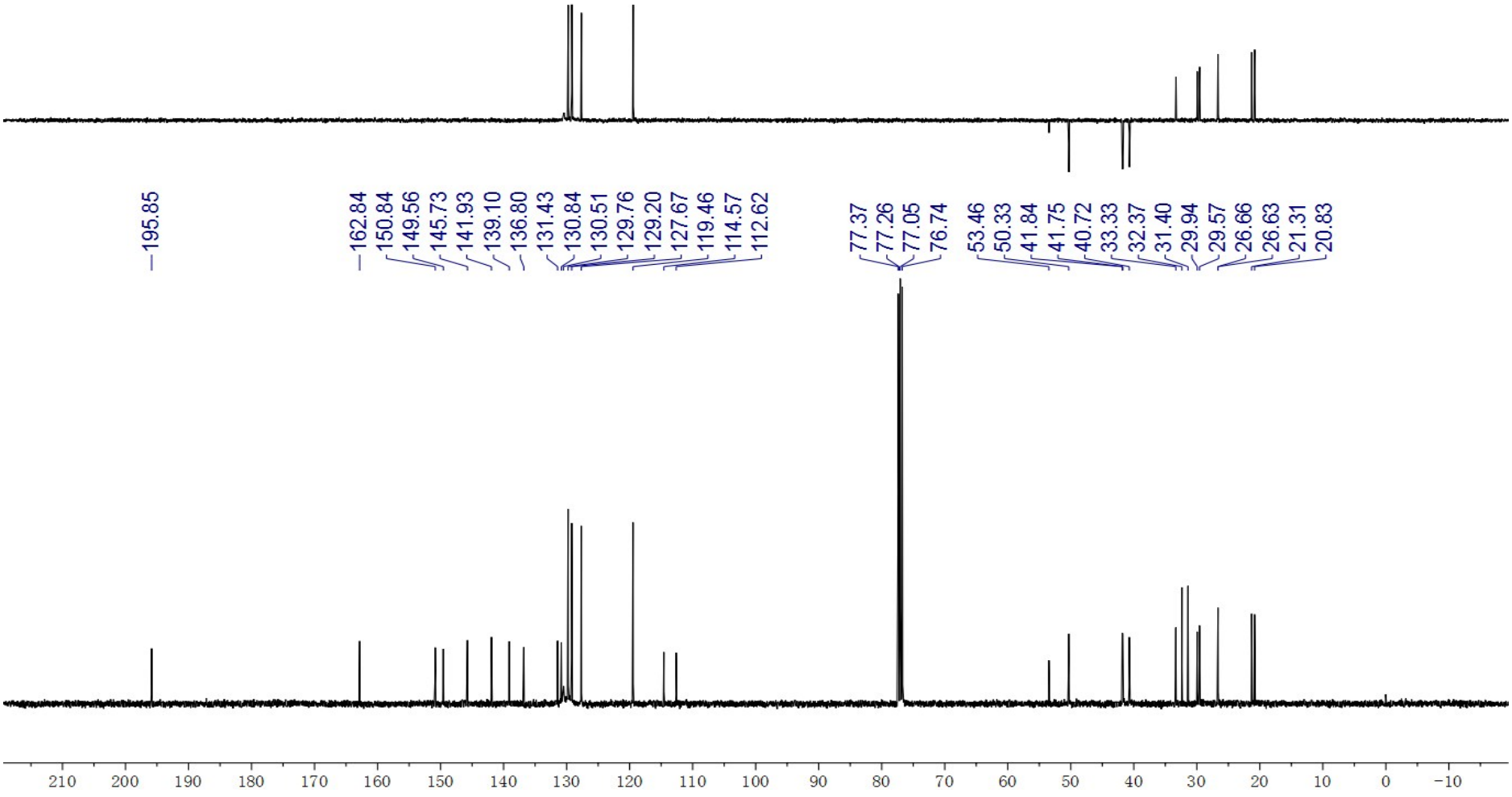


Figure 62. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4m**

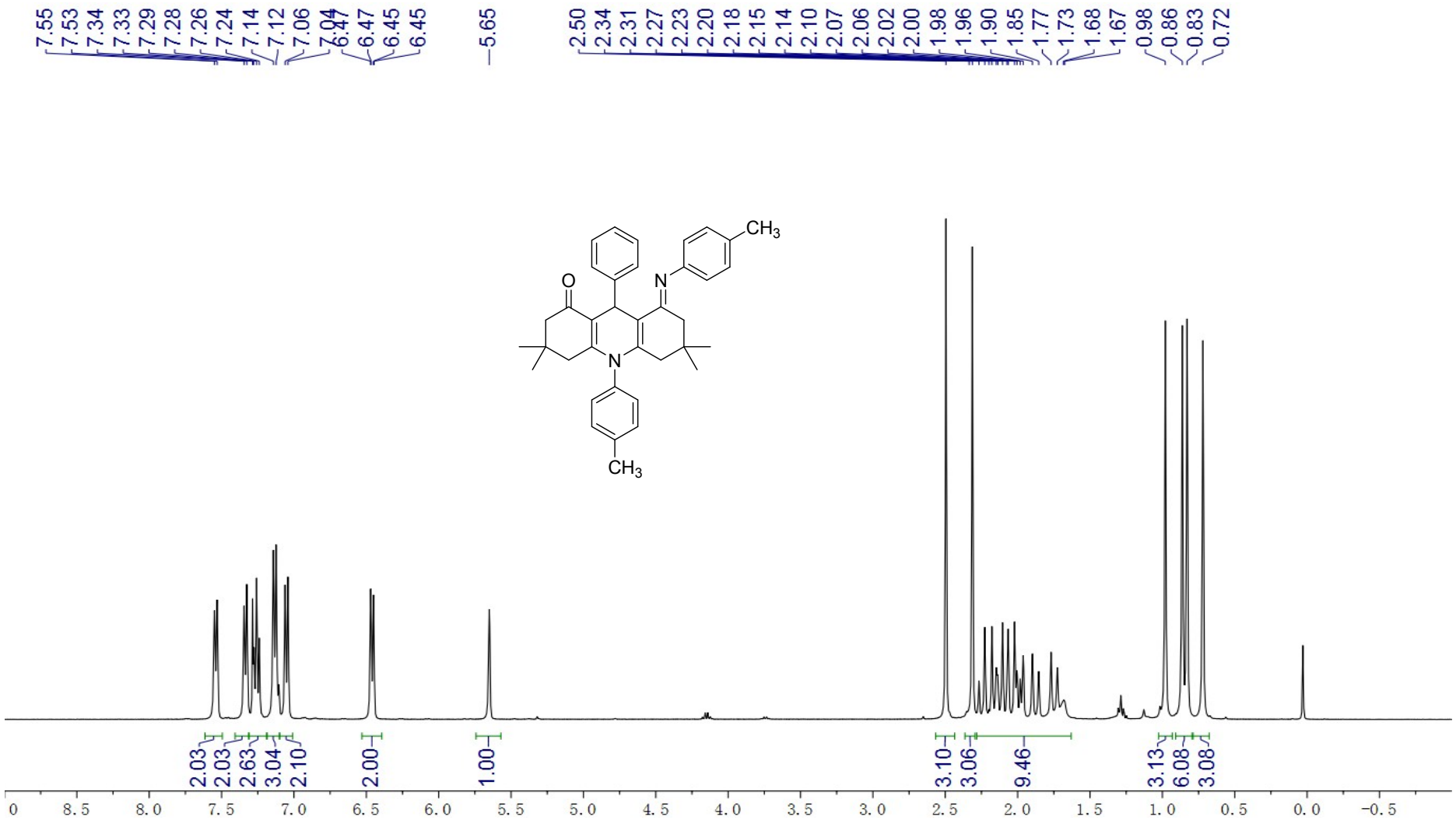


Figure 63. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4n**

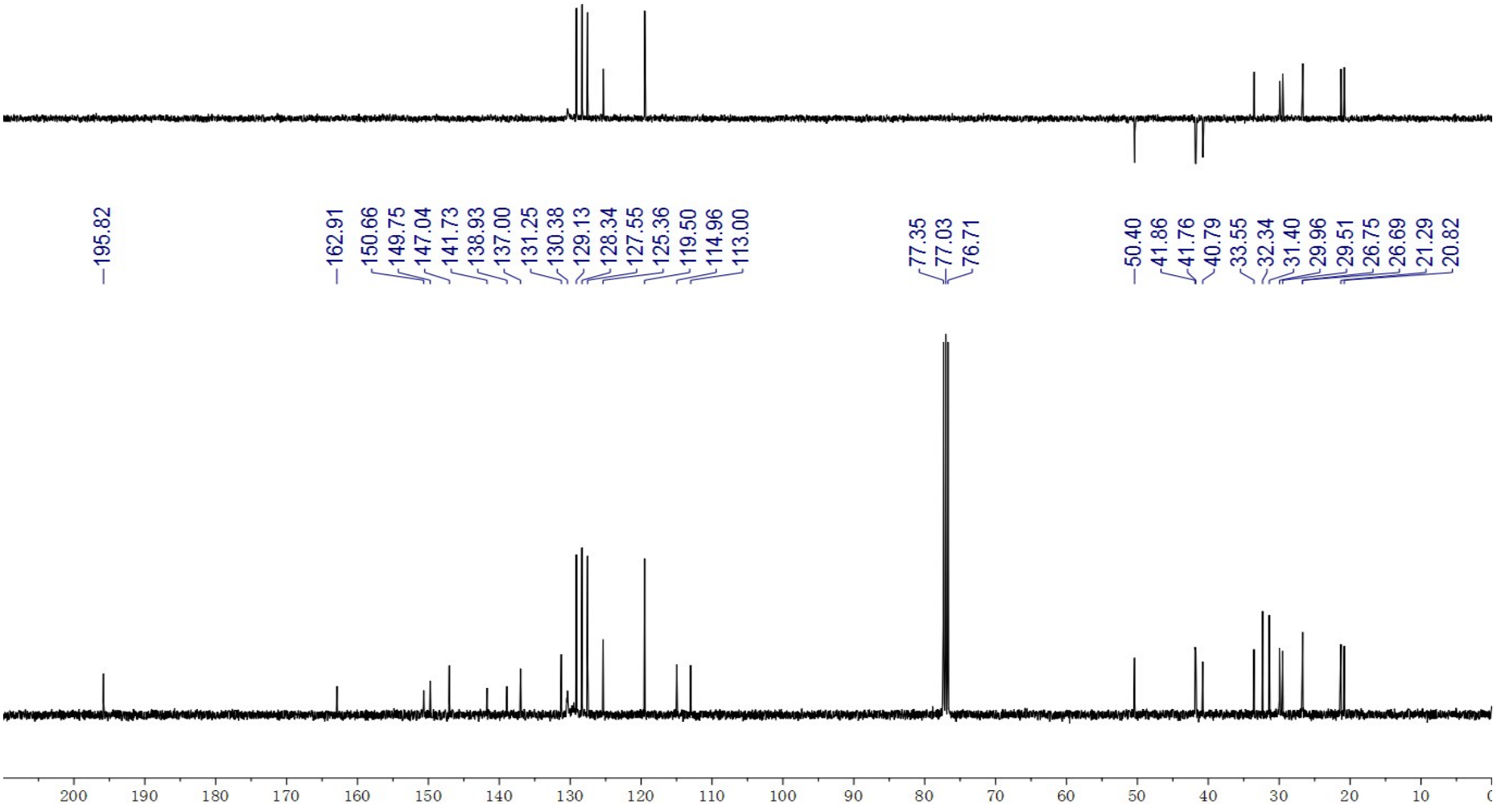


Figure 64. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4n**

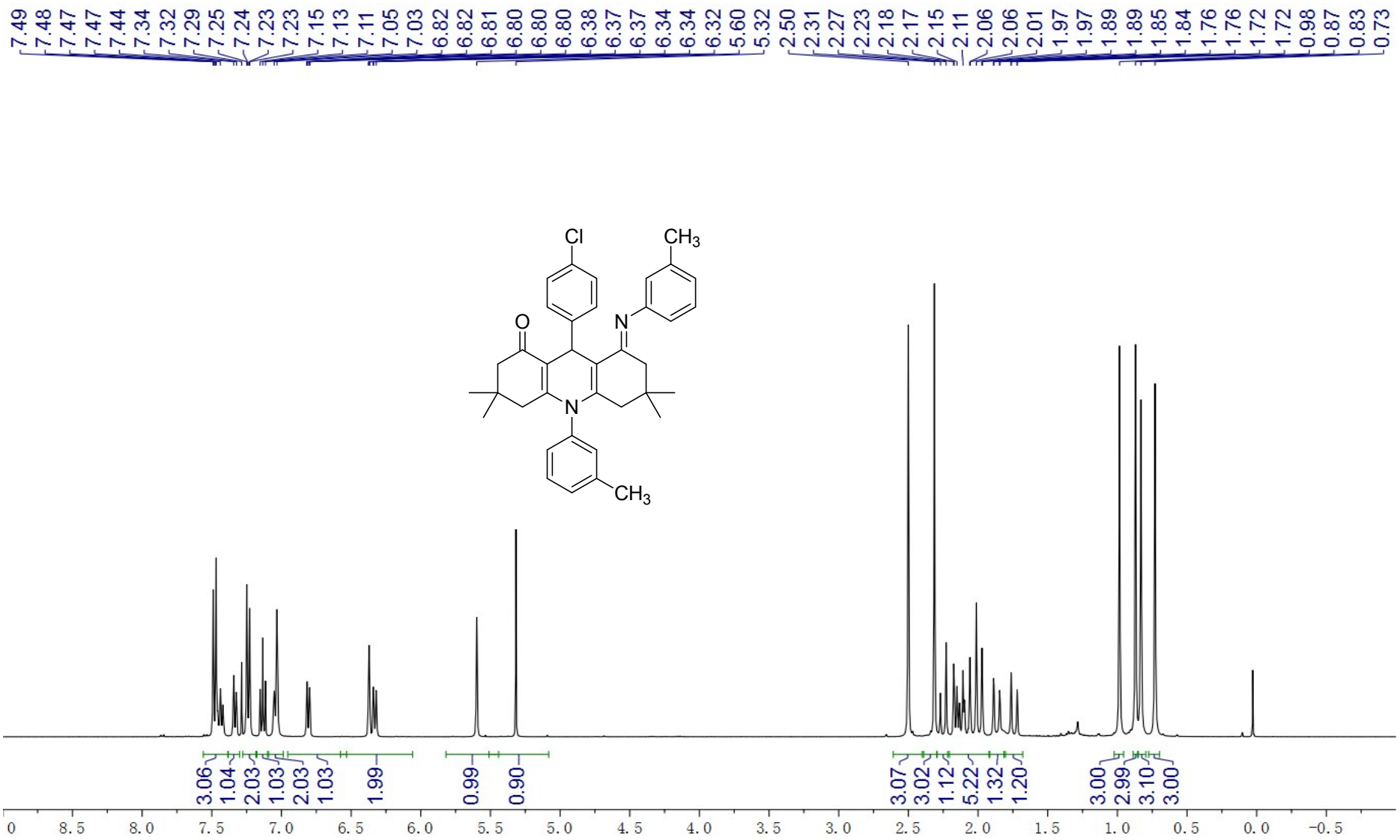


Figure 65. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4o**

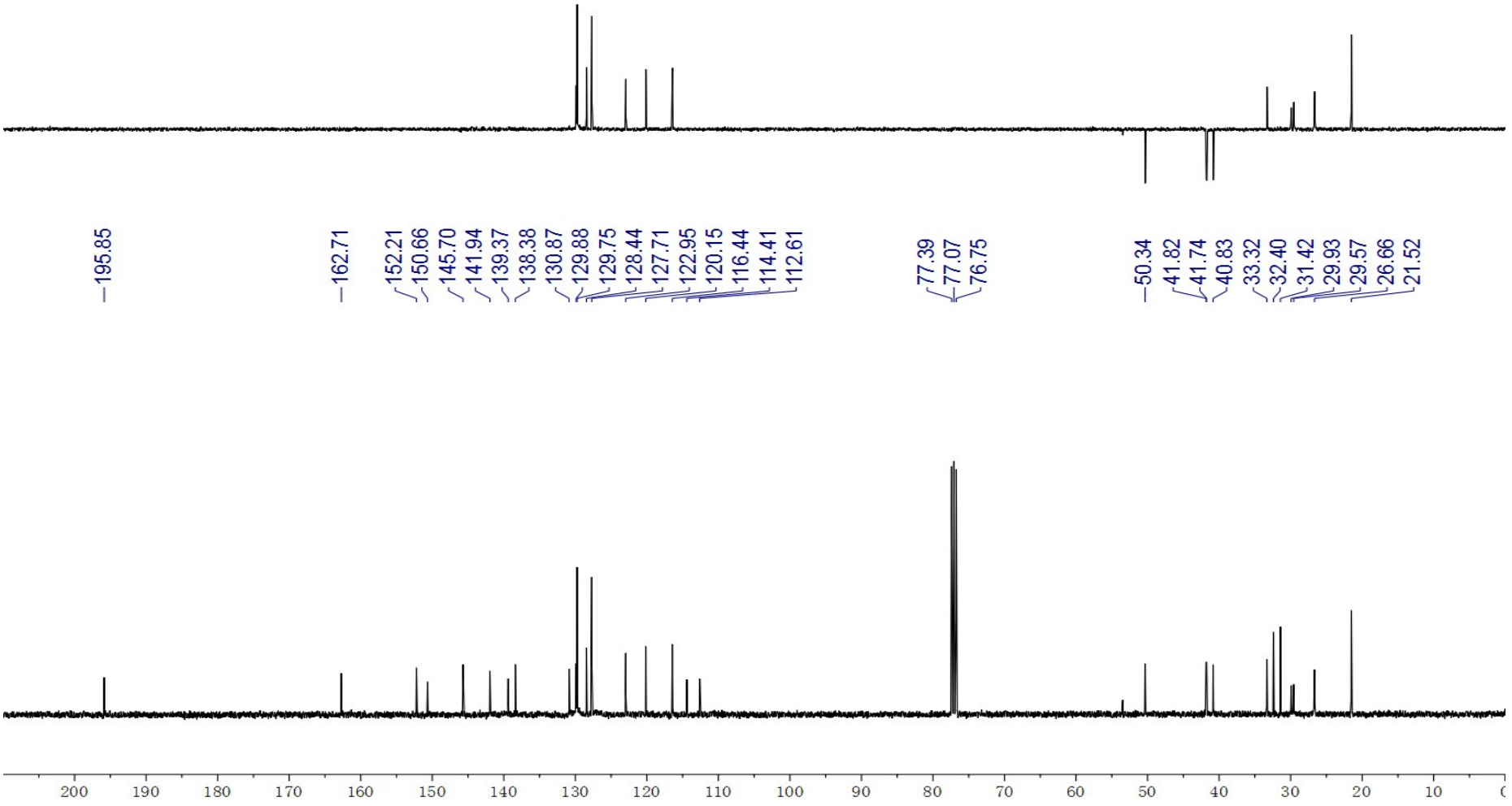


Figure 66. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4o**

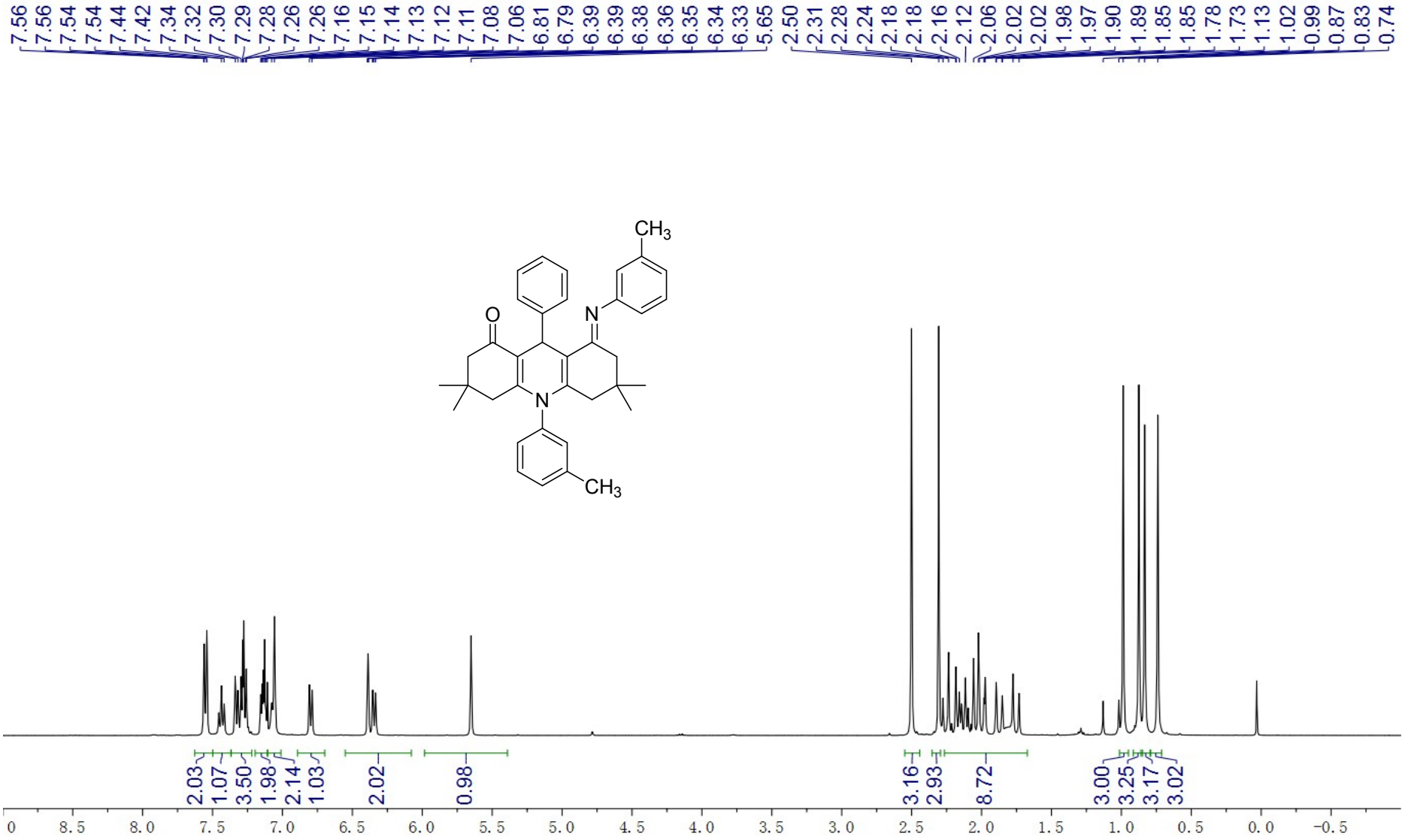


Figure 67. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4p**

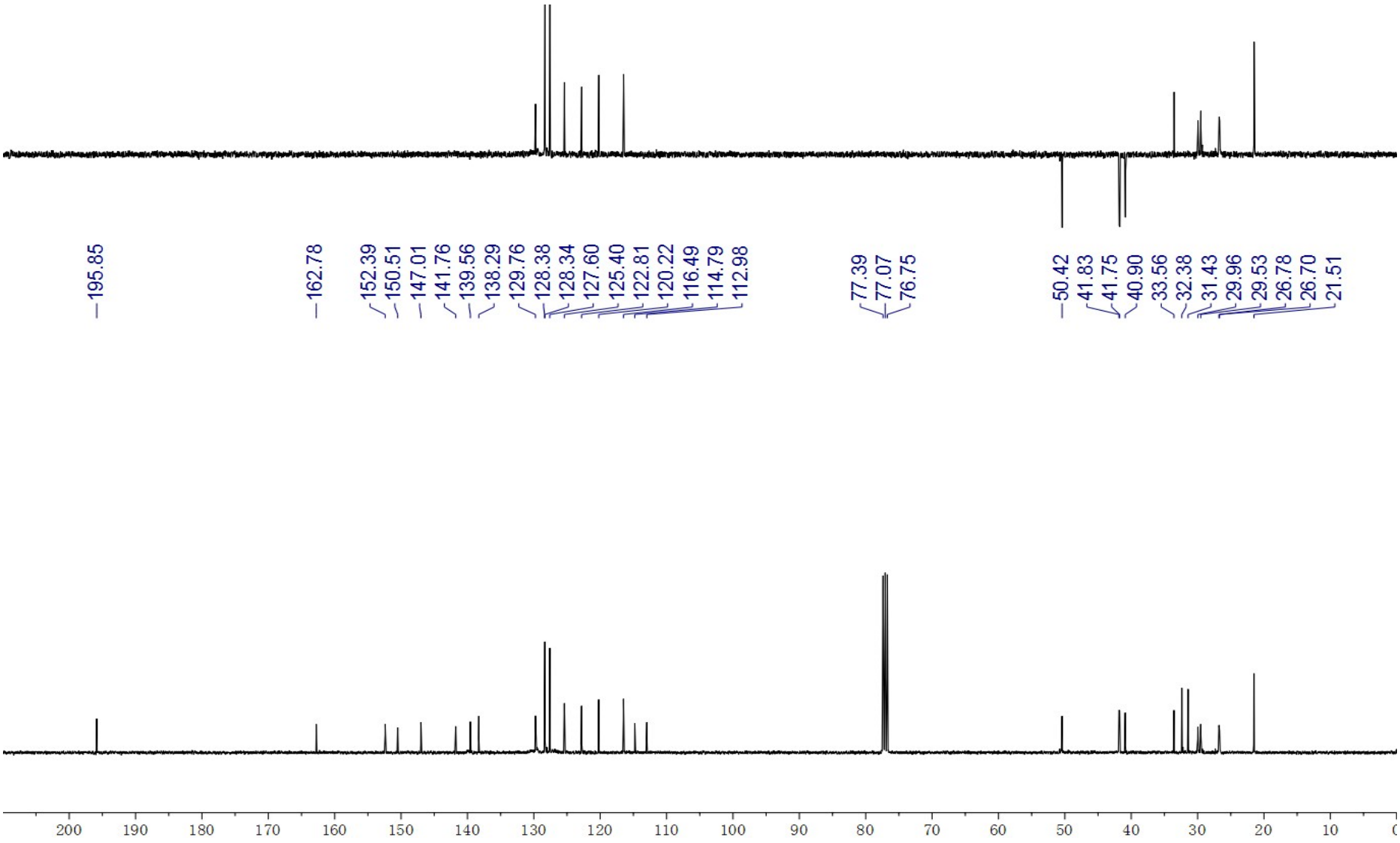


Figure 68. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4p**

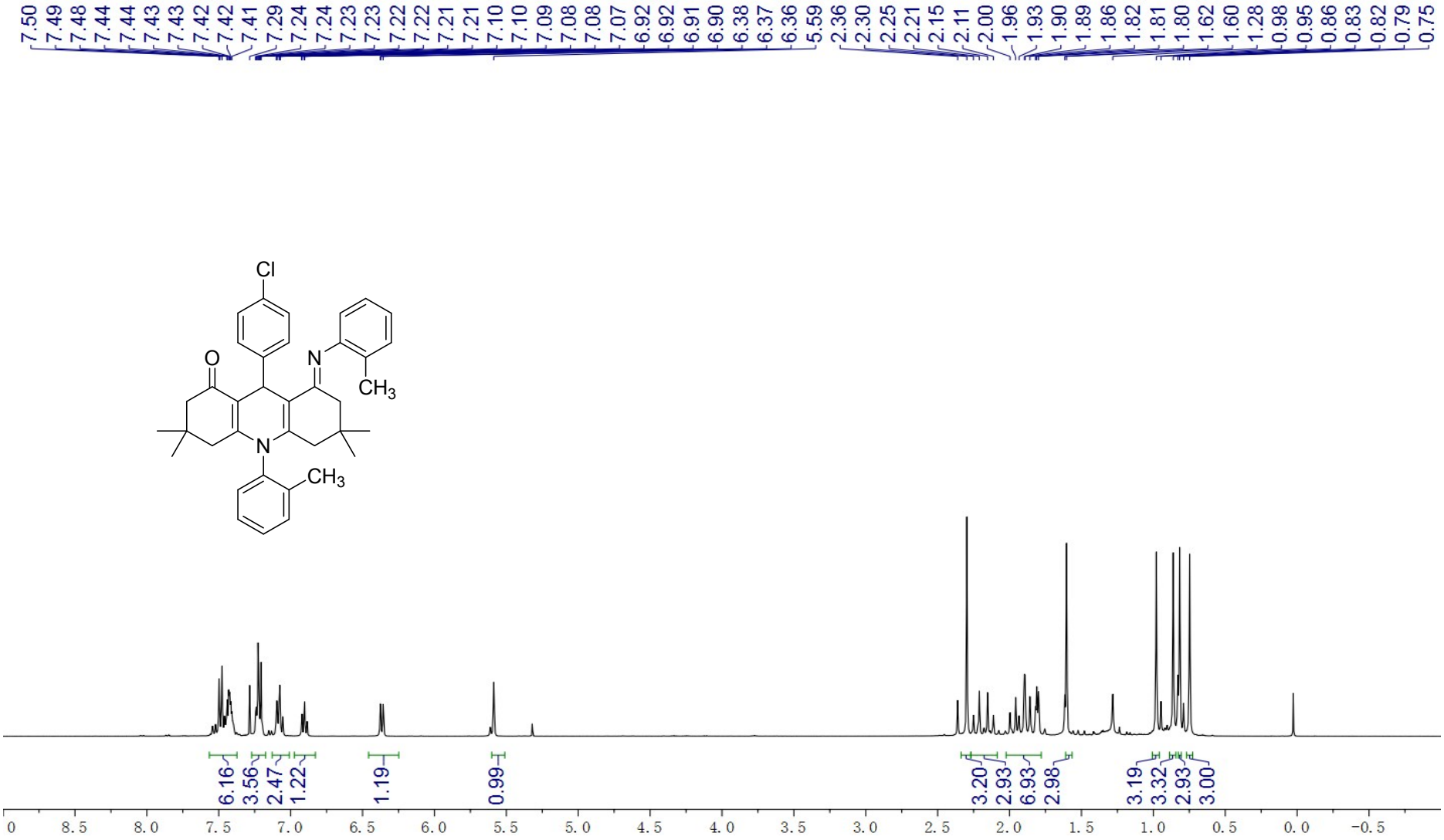


Figure 69. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4q**

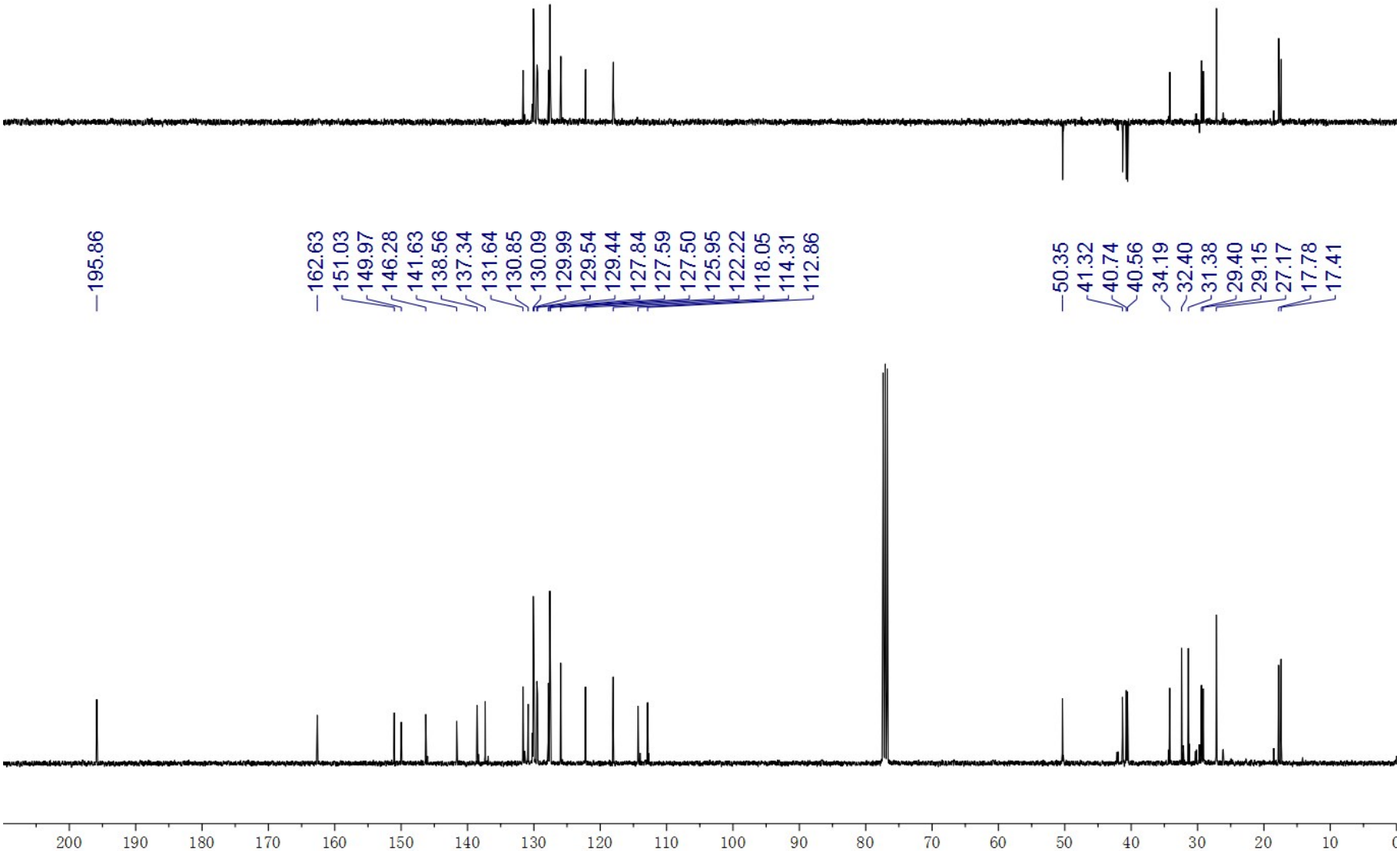


Figure 70. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4q**

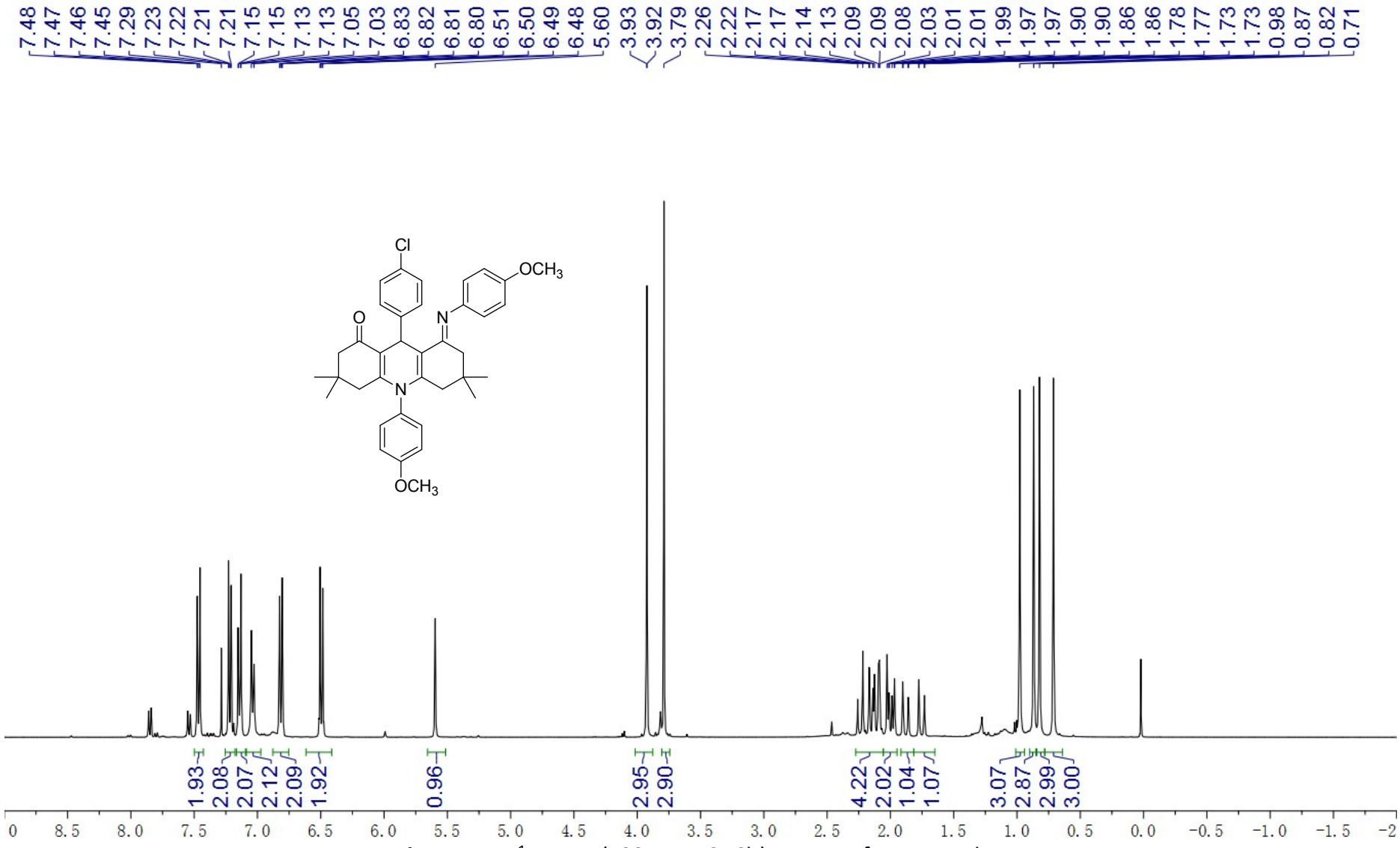


Figure 71. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4r**

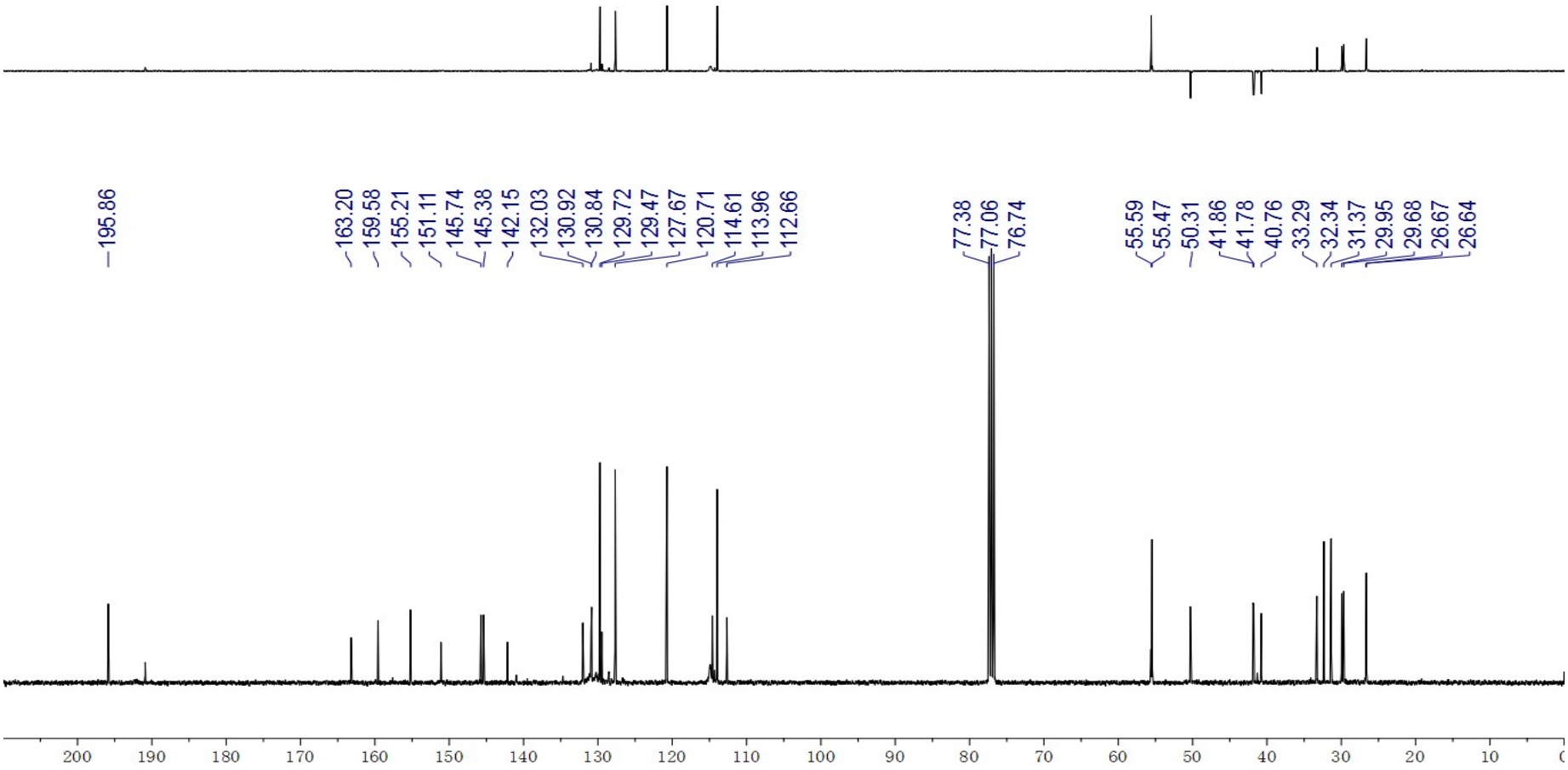


Figure 72. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4r**

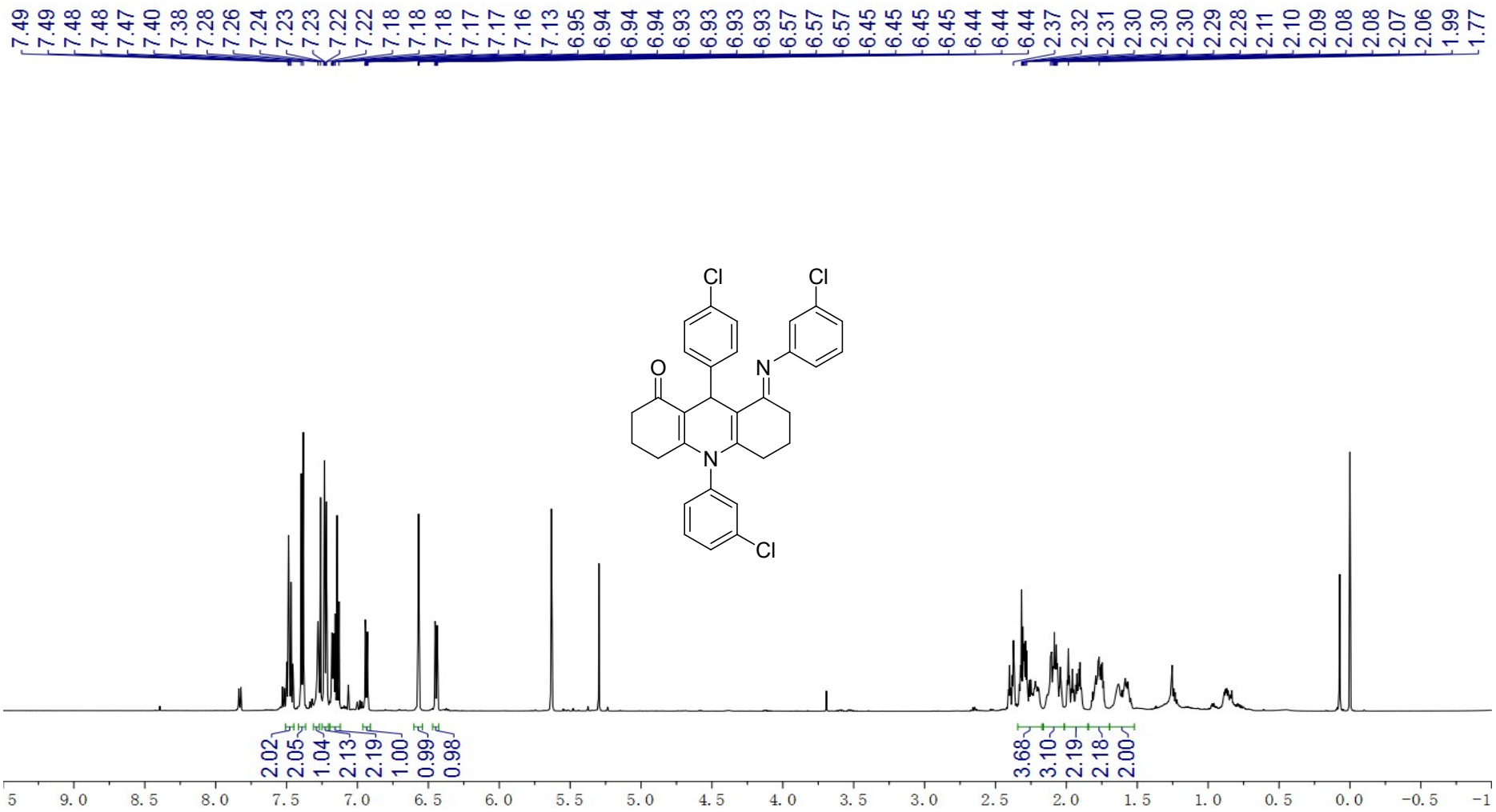


Figure 73. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4s**

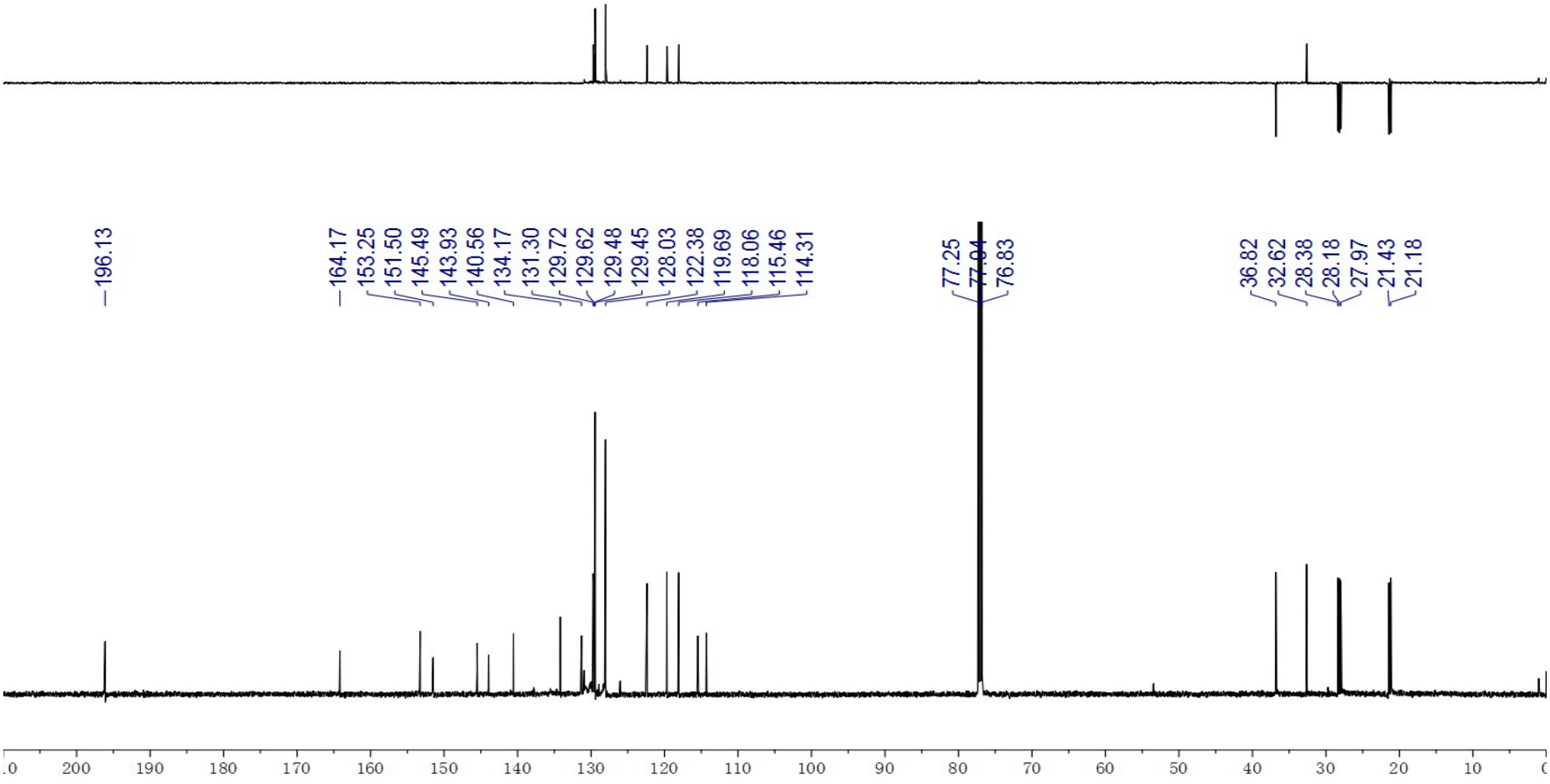


Figure 74. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4s**

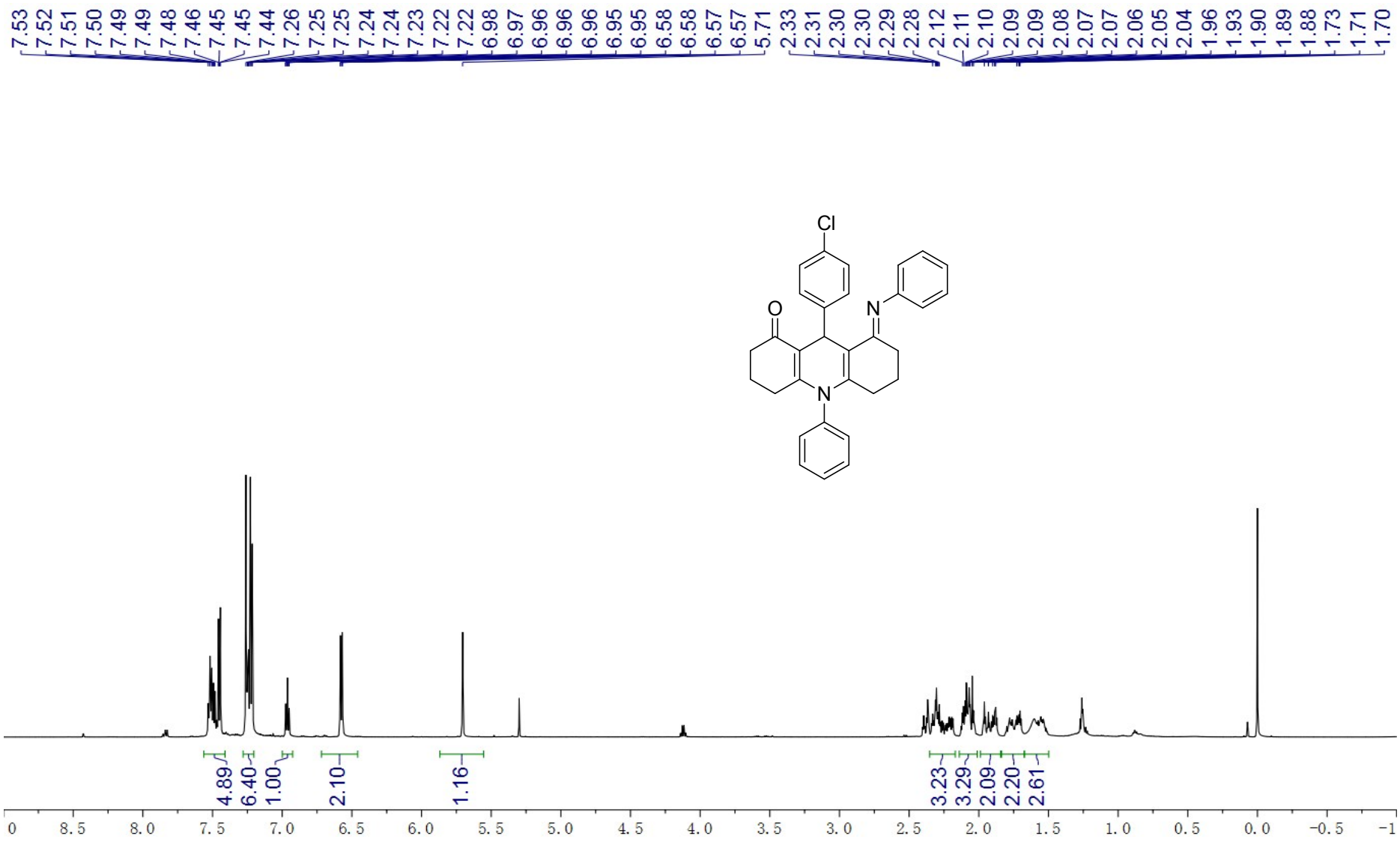


Figure 75. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4t**

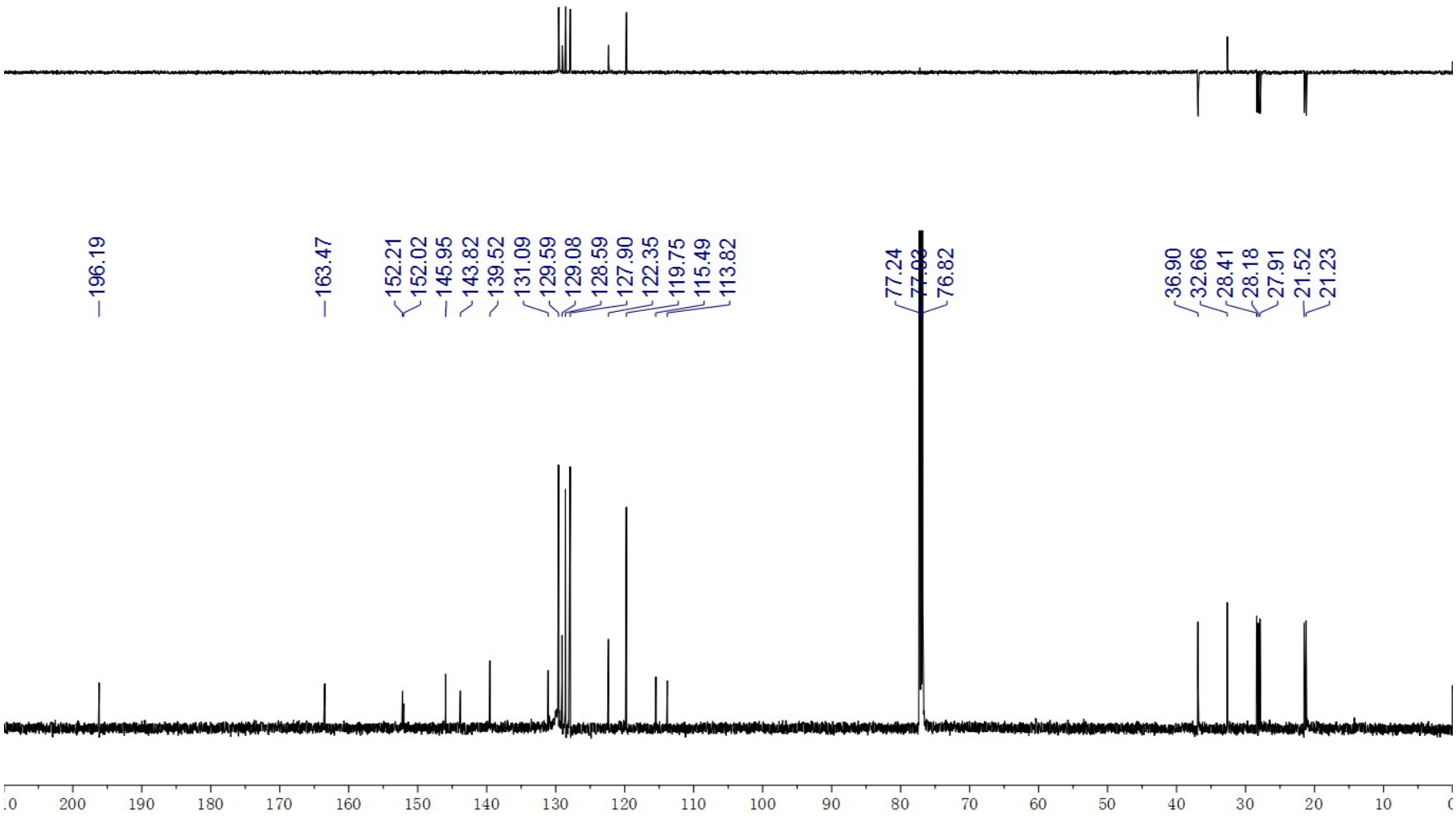


Figure 76. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4t**

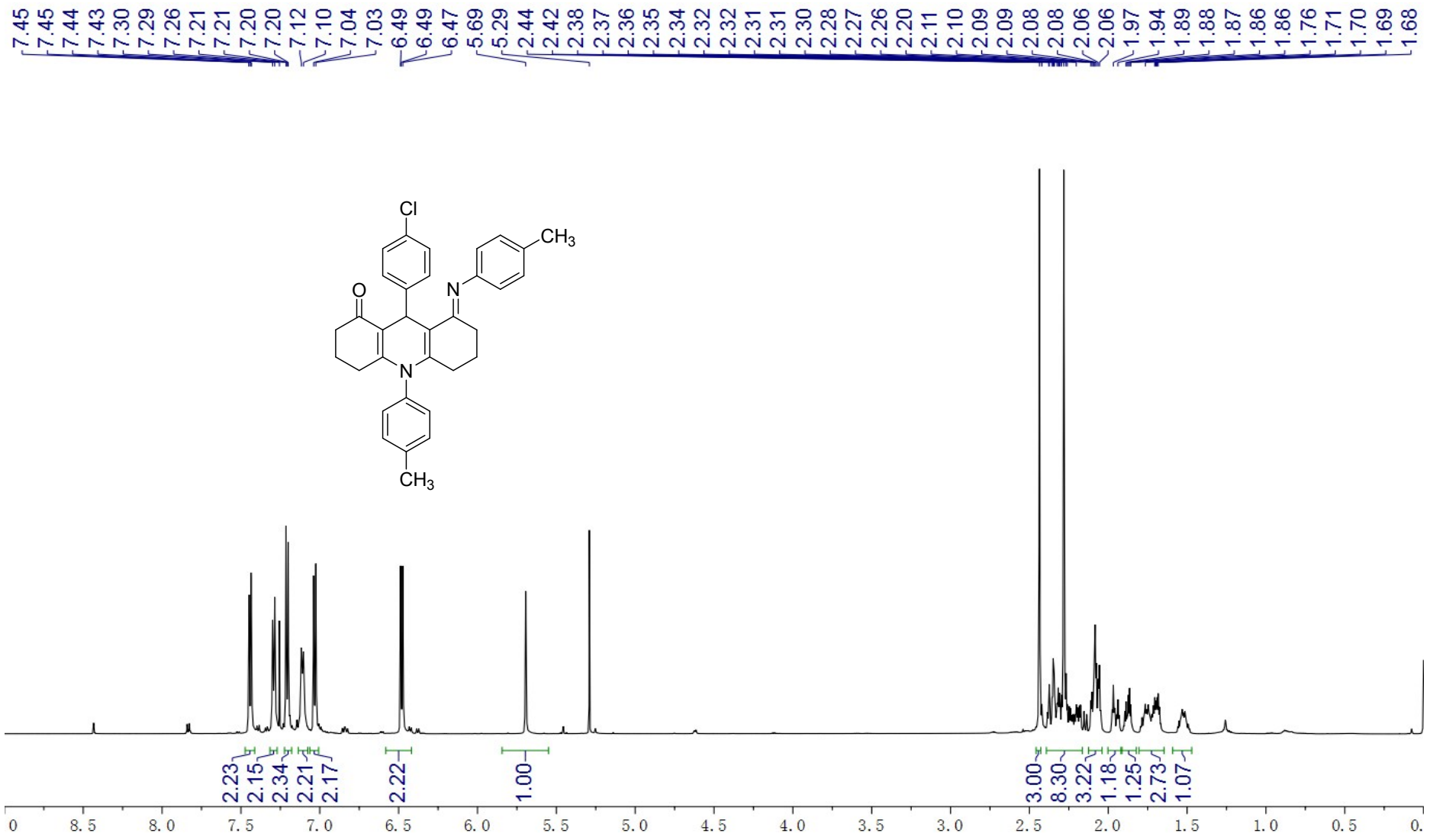


Figure 77. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4u**

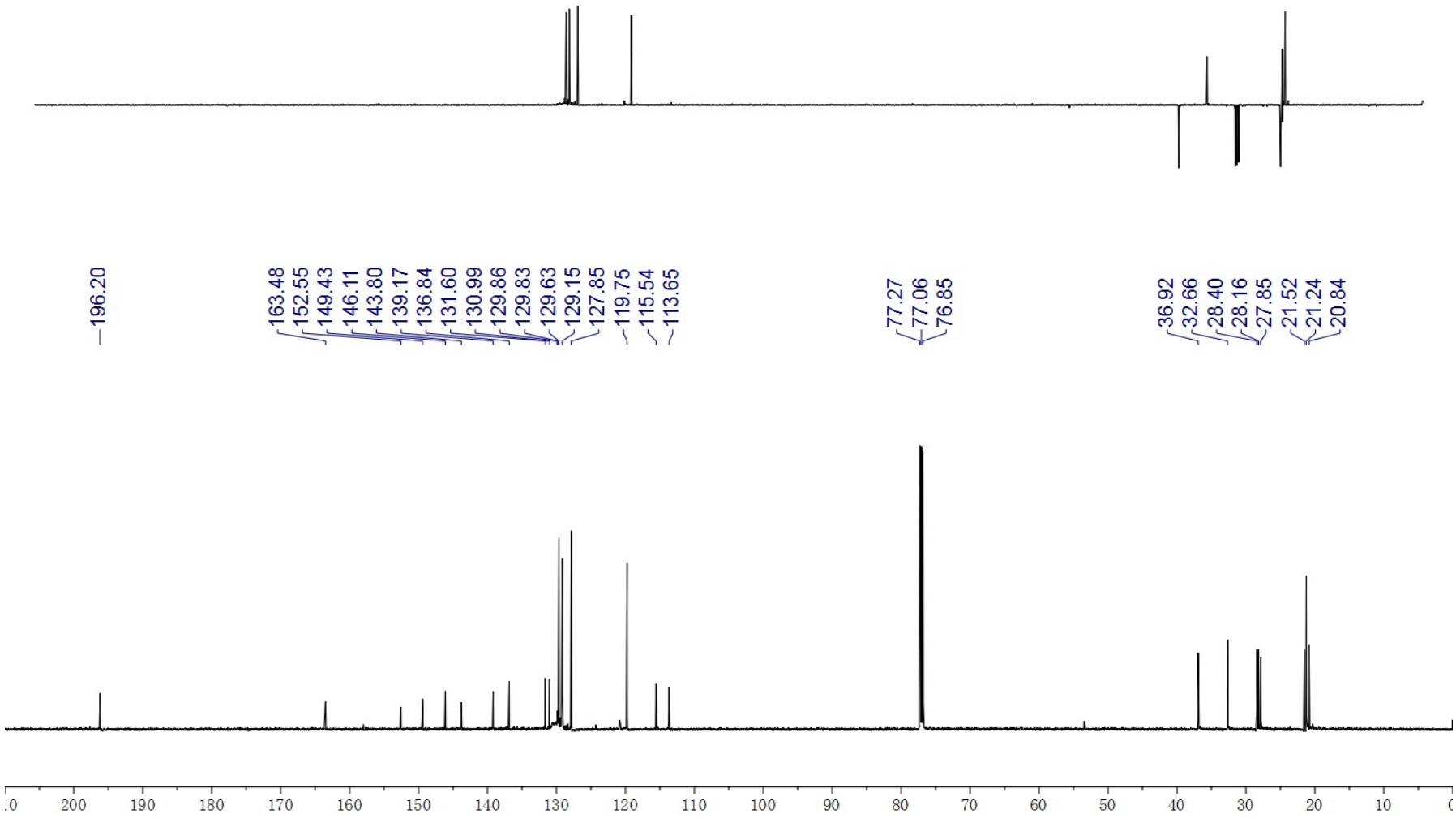


Figure 78. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4u**

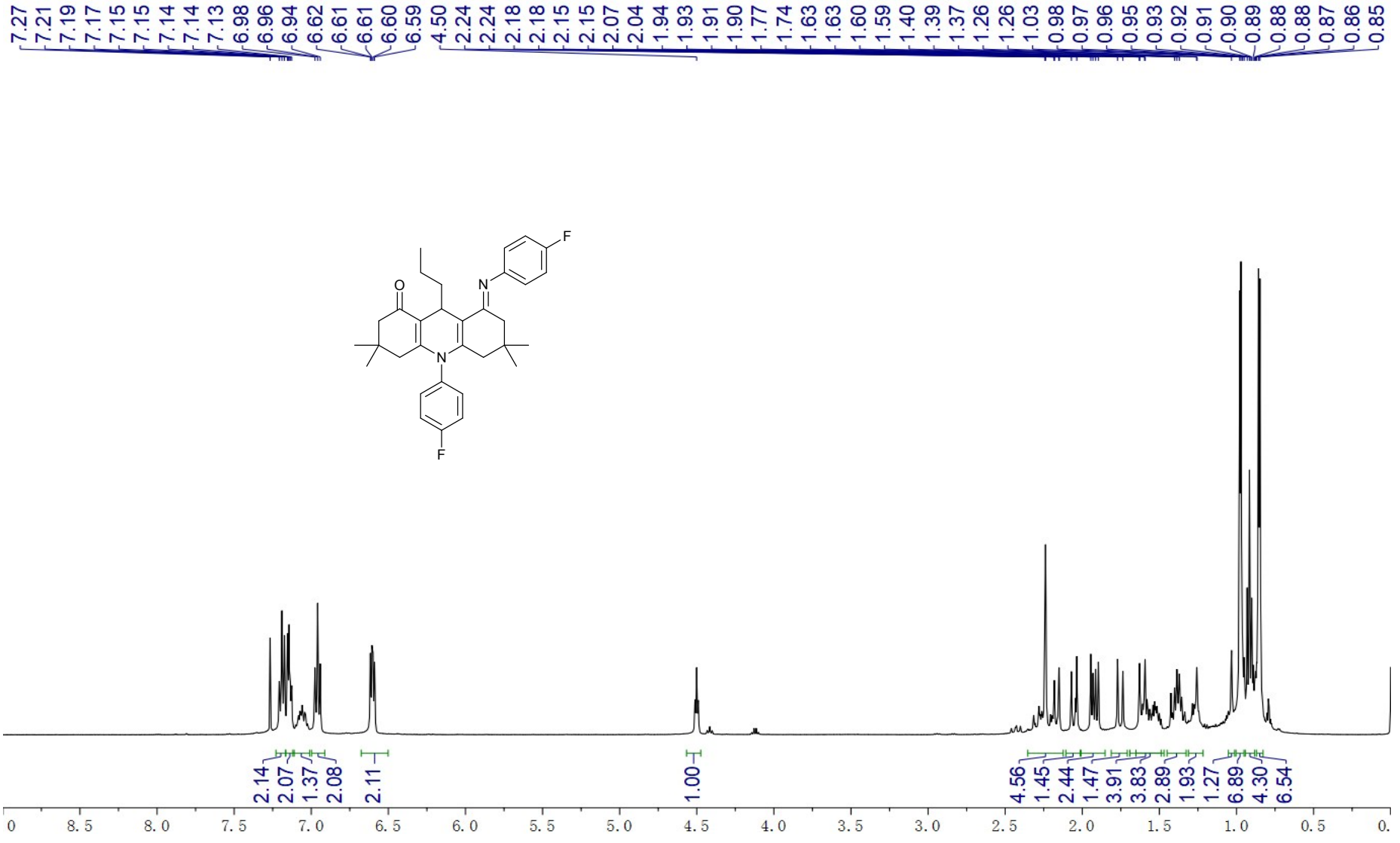


Figure 79. ^{1}H NMR (500 MHz, $CDCl_3$) spectra of compound **4v**

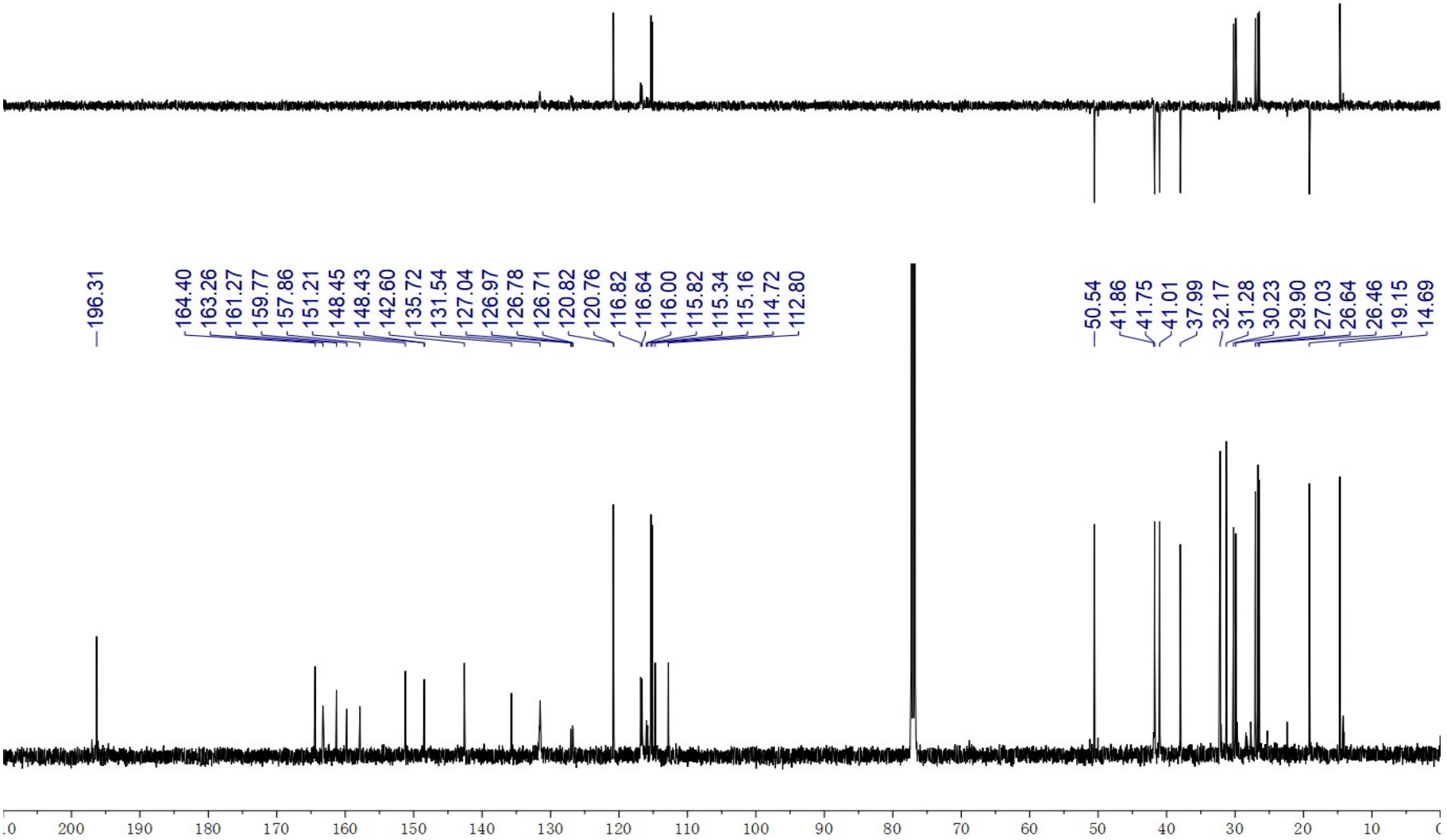


Figure 80. ^{13}C NMR (125 MHz, $CDCl_3$) spectra of compound **4v**

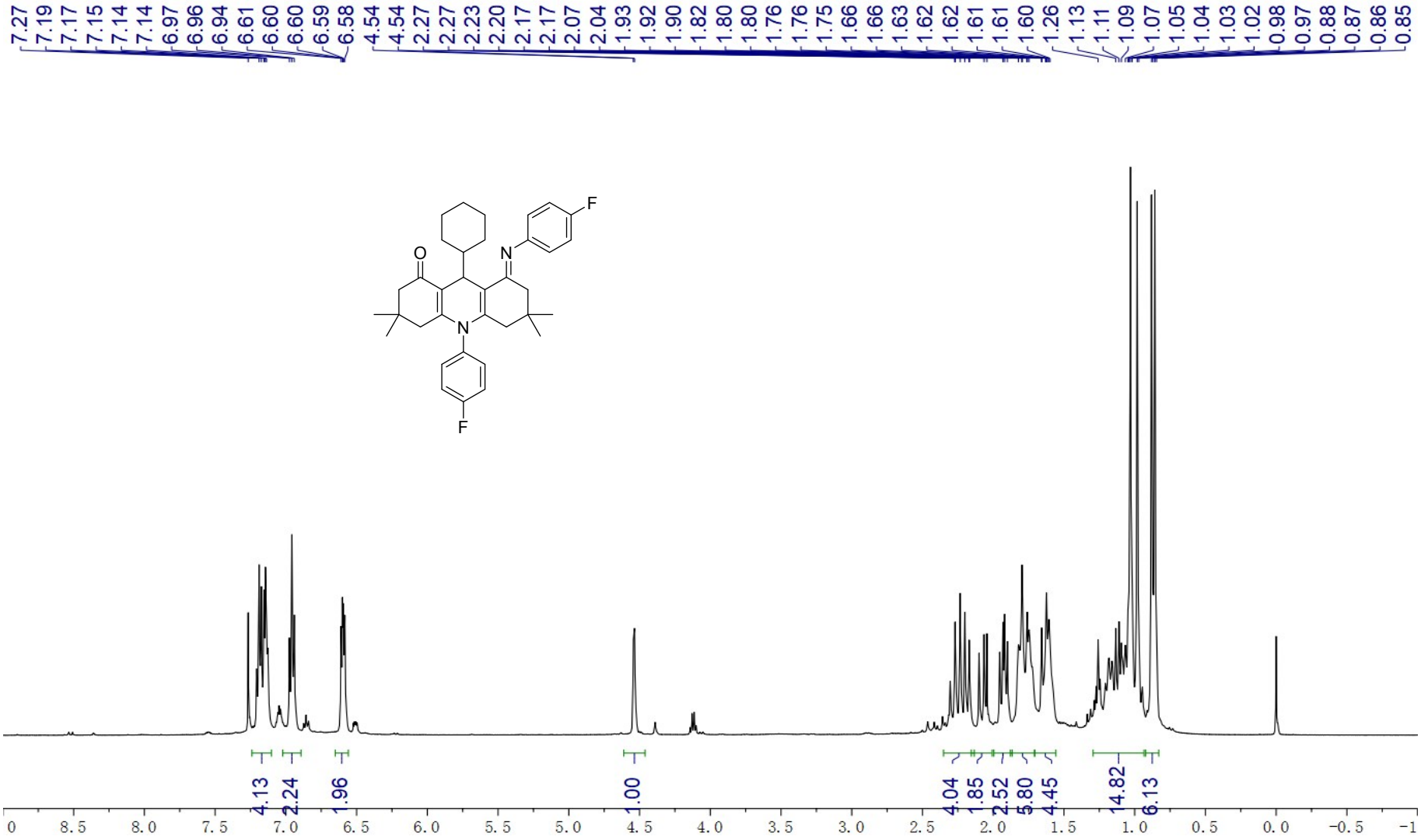


Figure 81. ^{1}H NMR (500 MHz, $CDCl_3$) spectra of compound **4w**

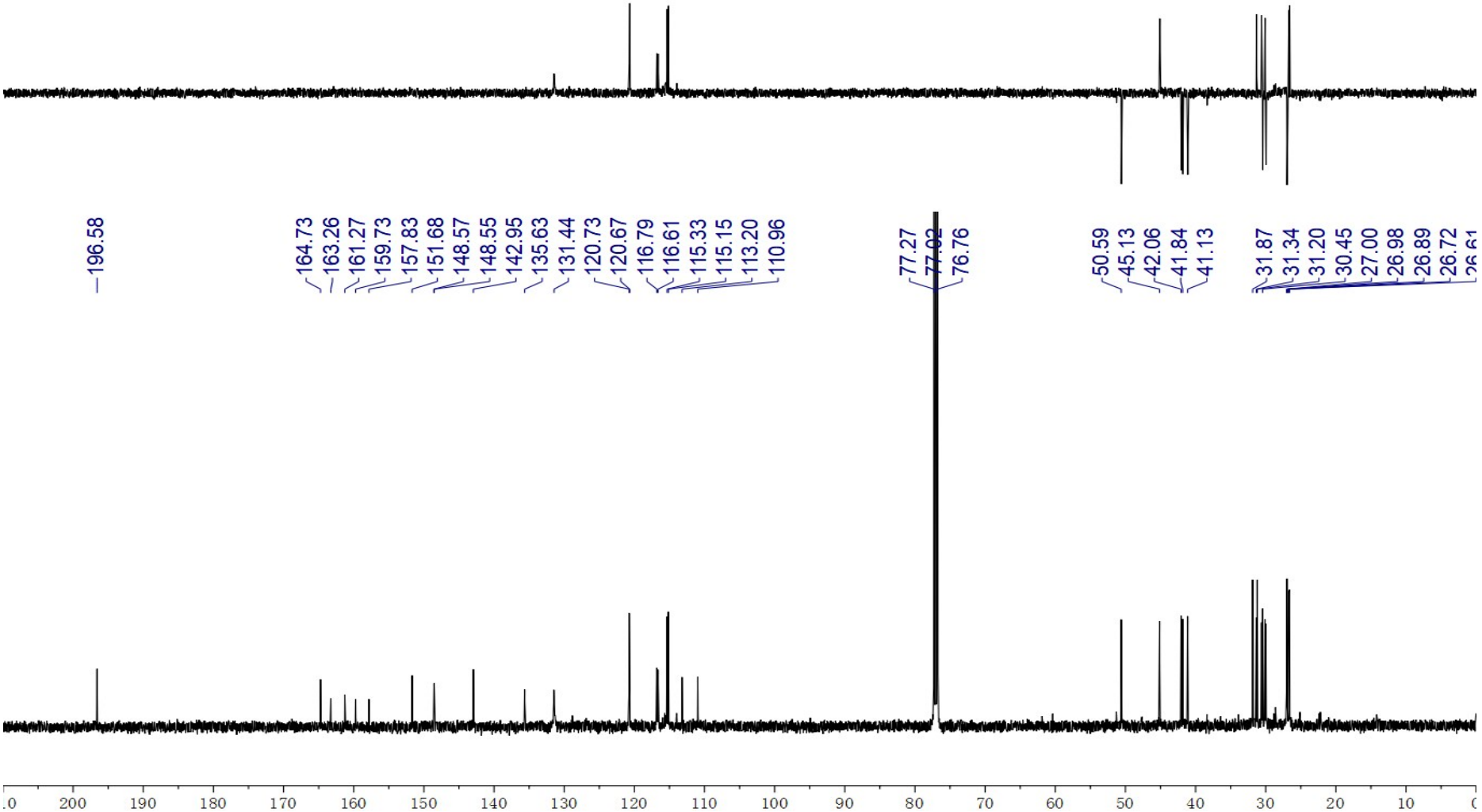
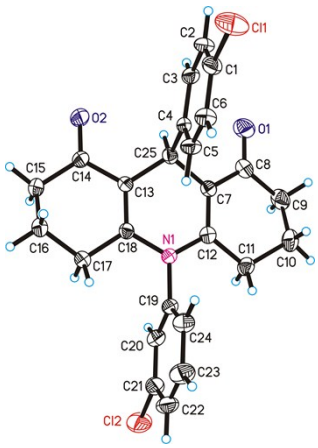


Figure 82. ^{13}C NMR (125 MHz, $CDCl_3$) spectra of compound **4w**

5. Crystal X-ray Structures of Compound 3n[1]



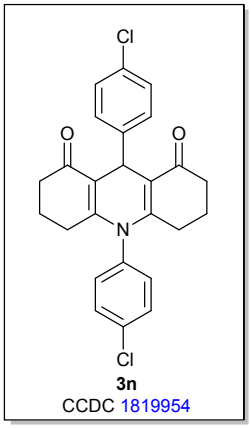


Figure S77 ORTEP view of the molecular structure of compound **3n**, thermal ellipsoids are drawn at 30% probability

Table S1 Crystal data, data collection, and structure refinement for compound **3n**

9,10-bis(4-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (3n)	
Empirical formula	$C_{25}\ H_{21}\ Cl_2\ N\ O_2$
Formula weight	438.33
Temperature	293(2) K
Wavelength	0.71073 Å
Crystal system, space group	Monoclinic, P21
Unit cell dimensions	a = 8.7539(14) A alpha = 90 deg. b = 10.6930(17) A beta = 102.694(2) deg. c = 11.5682(18) A gamma = 90 deg.
Volume	1056.4(3) A^3
Z, Calculated density	2, 1.340 Mg/m^3
Absorption coefficient	0.330 mm^-1
F(000)	456
Crystal size	0.36 x 0.30 x 0.25 mm
Theta range for data collection	1.804 to 24.998 deg.
Limiting indices	-9<=h<=10, -12<=k<=12, -12<=l<=13
Reflection collected/unique	5931 / 3277 [R(int) = 0.0254]
Completeness to theta = 28.40	97.2 %
Max. and min. transmission	0.922 and 0.890
Refinement method	Full-matrix least-squares on F^2
Data/restraints/parameters	3277 / 1 / 272
Goodness-of-fit on F^2	0.953
Final R indices [I>2sigma(I)]	R1 = 0.0387, wR2 = 0.1013
R indices (all data)	R1 = 0.0518, wR2 = 0.1103
Largest diff. peak and hole	0.247 and -0.229 e.A^-3

6. Notes and References

(1) CCDC 1819954 which containing in the supporting information (SI) for crystallographic data of compounds **3n**. This material is available free of charge from The Cambridge Crystallographic Data Center *via* the Internet at www.ccdc.cam.ac.uk/data_request/cif.