Supporting Information

Construction of Fluorescence-tunable Pyrido-Fused Benzimidazoles *via* Direct Intramolecular C-H amination under Transition-Metal-Free Conditions

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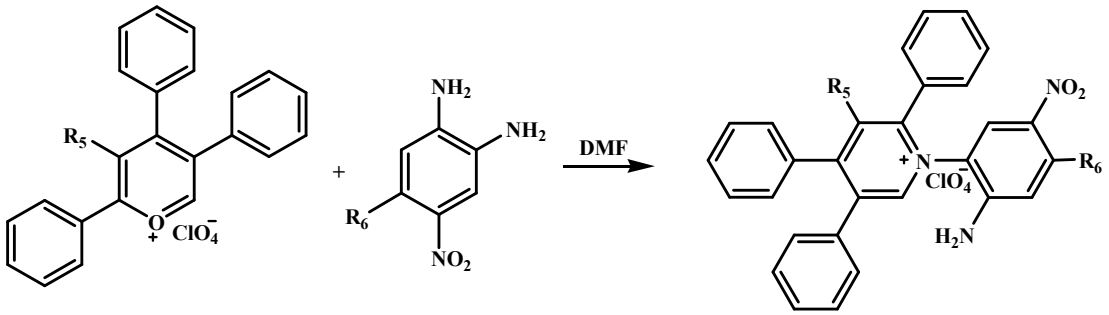
General Information: ^{1}H NMR and ^{13}C NMR spectra were measured on a VARIAN INOVA-400 and AVANCE II 400 spectrometer with chemical shifts reported as ppm (in CH_3CN- d^3 and $CDCl_3$, TMS as internal standard). Mass spectrometric data were obtained on a Q-Tof MS spectrometry. Absorption spectra were recorded on a HITACHI U-4100 spectrometer. Fluorescence spectra were taken on a JASCO FP-6300 spectrometer. TLC analysis was performed on silica gel plates and column chromatography was conducted over silica gel (mesh 200-300).

General Procedure for the Synthesis of nitrophenylpyridinium perchlorate derivatives (1e, 1j and 1m)[1]:



1e: R_5=H, R_6=H;
1j: R_5=Ph, R_6=H;
1m: R_5=H, R_6=NO_2

2,4,5-triphenylpyrylium perchlorate (0.2g, 0.49mmol) and 4-Nitro-o-phenylenediamine (0.090g, 0.59mmol) was stirred in 5mL DMF at 78℃. After 4h, the solvent was concentrated in vacuo, diluted with CH_3CN (2~3ml), then 30mL diethyl ether was slowly poured into the solution, and the deep yellow precipitate was formed. The crude product was washed with diethyl ether (3×10mL) and dried to give pure target 2,4,5-triphenylpyridinium **1e**.

1-(5-nitrophenyl)-2,4,5-triphenylpyridinium perchlorate (1e): ^{1}H NMR (400 MHz, DMSO) δ 9.31 (s, 1H), 8.55 (s, 1H), 8.41 (s, 1H), 8.13 – 7.87 (m, 1H), 7.69 – 7.53 (m, 3H), 7.47 (dd, J = 16.9, 9.3 Hz, 10H), 7.38 – 7.20 (m, 3H), 6.78 (d, J = 9.2 Hz, 1H); ^{13}C NMR (DMSO, 100 MHz) δ: 157.36, 154.88, 149.87, 148.57, 139.07, 135.65, 135.58, 134.08, 131.61, 131.52, 131.43, 130.89, 130.21, 130.02, 129.75, 129.60, 129.41, 129.36, 128.91, 127.72, 126.74, 124.42, 116.02; HRMS calcd for $C_{29}H_{22}N_3O_2$ (M^+) 444.1712, found 444.1726. Yield: 57%.

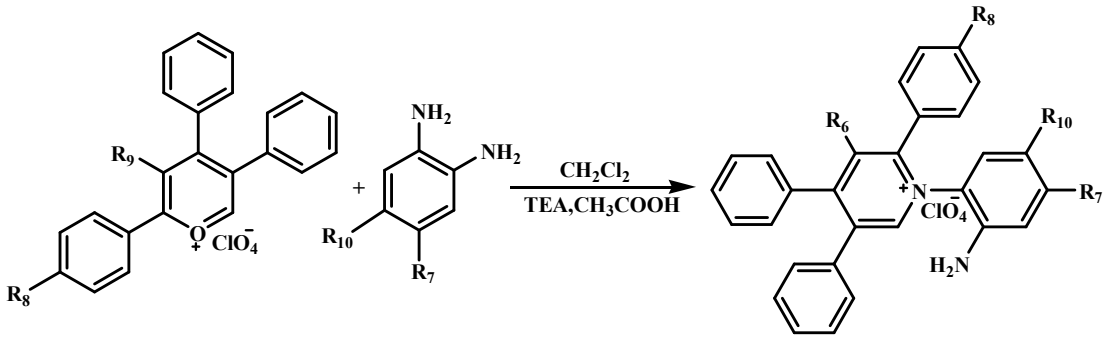
1-(5-nitrophenyl)-1,2,4,5-tetraphenylpyridinium perchlorate (1j): ^{1}H NMR (400 MHz, DMSO) δ 9.43 (s, 1H), 8.57 (d, *J* = 16.8 Hz, 1H), 7.94 (d, *J* = 2.6 Hz, 1H), 7.41 – 7.30 (m, 5H), 7.27 (d, *J* = 3.6 Hz, 4H), 7.22 – 7.09 (m, 6H), 7.09 – 6.87 (m, 7H), 6.74 (d, *J* = 23.8 Hz, 1H), 3.39 (s, 3H); ^{13}C NMR (DMSO, 100 MHz) *δ*: 154.37, 154.88, 149.06, 146.00, 142.28, 140.13, 134.70, 133.78, 130.58, 130.21, 130.01, 129.69, 129.12, 128.43, 127.89, 127.44, 127.33, 124.57, 115.26; HRMS calcd for $C_{35}H_{26}N_3O_2$ (M^+) 520.2020, found 520.2017. Yield: 56%.

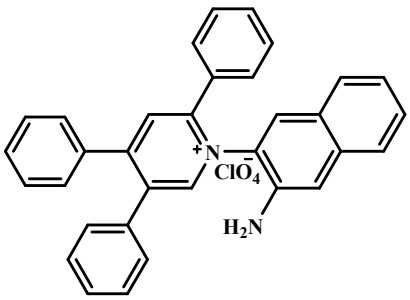
1-(4,5-dinitrophenyl)-2,4,5-triphenylpyridinium perchlorate (1m): ^{1}H NMR (500 MHz, DMSO) δ 9.33 (d, *J* = 18.2 Hz, 1H), 8.76 (s, 1H), 8.46 (s, 1H), 8.28 – 6.70 (m, 17H), 5.75 (s, 1H). ^{13}C NMR(100 MHz, DMSO): *δ*=157.293, 154.237, 149.757, 147.930, 146.656, 138.559, 134.912, 133.502, 131.296, 131.091, 130.690, 130.568, 129.605, 129.545, 129.355, 129.249, 128.984, 128.620, 128.370, 125.921, 124.109, 110.690; HRMS calcd for $C_{29}H_{21}N_4O_4$ (M^+) 489.1563, found 489.1561. Yield: 57%.

Safety note:

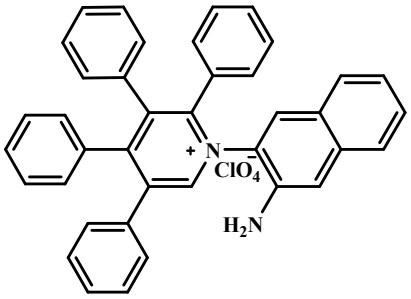
Organic perchlorates are unstable and highly explosive! Only we can synthesis in simal scal, and these should be handled with great cautions.

General Procedure for the Synthesis of pyridinium perchlorate derivatives (1a,1b,1c,1d,1f,1g,1h,1i,1j,1k,1l,1n,1o and 1p):

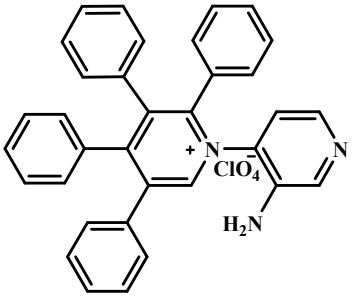




1n



1o



1p

1a: R_9=H, R_7=H, R_8=H, R_{10}=H;
1b: R_9=H, R_7=CH_3, R_8=H, R_{10}=H;
1c: R_9=H, R_7=OCH_3, R_8=H, R_{10}=H;
1d: R_9=H, R_7=Cl, R_8=H, R_{10}=H;
1f: R_9=H, R_7=H, R_8=Ph, R_{10}=H;
1g: R_9=Ph, R_7=H, R_8=H, R_{10}=H;
1h: R_9=Ph, R_7=CH_3, R_8=H, R_{10}=H;
1i: R_9=Ph, R_7=Cl, R_8=H, R_{10}=H;
1k:R_9=H, R_7=H, R_8=H, R_{10}=COOMe;
1l: R_9=Ph, R_7=Br, R_8=H, R_{10}=H

2,4,5-triphenylpyrylium perchlorate (0.2g, 0.49mmol), o-phenylenediamine (0.064g, 0.59mmol) and triethylamine (50mg, 0.49mmol) was stirred in dichloromethane (15ml) at room temperature. After 20 minutes, acetic acid (59mg, 0.98mmol) was added into the solution. After 2h, dichloromethane was concentrated under reduced pressure to 2~3mL. 30mL diethyl ether was poured into the solution, and the yellow precipitate was formed. The filtered precipitate was washed by diethyl ether (3×10mL) and dried to give pure target 2,4,5-triphenylpyridinium **1a**.

1-(2-aminophenyl)-2,4,5-triphenylpyridinium perchlorate (1a). ^{1}H NMR(400MHz, CD_3CN) δ 8.71 (s, 1H), 8.21 (s, 1H), 7.29-7.58 (m, 15H), 7.22 (m, 1H), 7.16 (d, *J*=8Hz, 1H), 6.81 (d, *J*=8Hz, 1H), 6.68 (m, 1H), 4.62 (s, 2H); ^{13}C NMR (100MHz, CD_3CN) δ: 159.2, 155.9, 148.1, 143.3, 140.7, 136.5, 134.7, 132.8, 132.7, 132.5, 132.0, 131.4, 130.8, 130.6, 130.4, 130.3, 129.9, 129.8, 129.5, 128.6, 127.6, 117.9. HRMS (ESI) m/z calcd for $C_{29}H_{23}N_2$ $[M]^+$, 399.1816; found, 399.1846. Yield: 87%.

1-(4-methyl-2-aminophenyl)-2,4,5-triphenylpyridinium perchlorate (1b): ^{1}H NMR (400 MHz, DMSO): δ 9.06 (d, J = 8.5 Hz, 1H), 9.06 (d, J = 8.5 Hz, 1H), 8.36 (s, 1H), 7.62 – 7.53 (m, 2H), 7.67 – 7.53 (m, 2H), 7.45 (dt, J = 17.7, 5.1 Hz, 11H), 7.32 (dd, J = 6.6, 2.6 Hz, 2H), 7.10 (d, J = 8.2 Hz, 1H), 6.98 (d, J = 8.4 Hz, 1H), 6.67 (d, J = 8.3 Hz, 1H), 6.57 (s, 1H), 6.36 (d, J = 8.2 Hz, 1H), 2.11 (d, J = 36.0 Hz, 3H).; ^{13}C NMR (CD_3CN, 100 MHz) δ: 156.66, 154.94, 154.70, 148.45, 148.24, 143.14, 138.80, 135.73, 134.12, 131.14, 130.69, 130.27, 130.02, 129.78, 129.73, 129.49, 129.31, 129.26, 128.76, 128.72, 117.28, 116.91, 21.39; HRMS (ESI) m/z calcd for $C_{30}H_{25}N_2$ $[M]^+$, 413.2012; found, 413.2032. Yield: 72%.

1-(4-methoxyl-2-aminophenyl)-2,4,5-triphenylpyridinium perchlorate (1c): ^{1}H NMR (400 MHz, CD_3CN) δ 8.70 (d, *J* = 14.3 Hz, 1H), 8.19 (d, *J* = 8.5 Hz, 1H), 7.65 – 7.47 (m, 4H), 7.47 – 7.35 (m, 9H), 7.32 (dd, *J* = 8.0, 1.3 Hz, 2H), 7.06 (d, *J* = 8.9 Hz, 1H), 6.32 (d, *J* = 2.6 Hz, 1H), 6.23 (dd, *J* = 8.9, 2.6 Hz, 1H), 3.71 (s, 3H); ^{13}C NMR (100 MHz, DMSO) *δ*: 162.03, 157.94, 155.23, 147.65, 143.56, 143.51, 139.57, 135.48, 133.70, 131.66, 131.60, 130.98, 130.37, 129.81, 129.54, 129.41, 129.35, 129.27, 128.87, 128.85, 128.62, 128.52, 120.34, 117.37, 104.09, 100.33, 55.17; HRMS calcd for $C_{30}H_{25}N_2O$ (M^+) 429.1961, found 429.1970. Yield: 77%.

1-(4-chloro-2-aminophenyl)-2,4,5-triphenylpyridinium perchlorate (1d): ^{1}H NMR (400 MHz, DMSO) δ 9.17 (d, *J* = 10.9 Hz, 1H), 8.38 (s, 1H), 7.59 (t, *J* = 6.3 Hz, 2H), 7.54 – 7.38 (m, 11H), 7.32 (d, *J* = 8.4 Hz, 3H), 6.78 (dd, *J* = 17.3, 5.5 Hz, 1H), 6.58 (d, *J* = 10.7 Hz, 1H), 6.03 (d, *J* = 55.3 Hz, 2H); ^{13}C NMR (100 MHz, DMSO) *δ*: 156.56, 154.37, 154.27, 144.48, 138.50, 135.48, 135.18, 133.59, 131.45, 130.26, 129.77, 129.58, 129.54, 129.22, 129.03, 128.83, 128.78, 128.35, 124.80, 118.21, 117.63, 115.20, 115.16; HRMS calcd for $C_{29}H_{22}ClN_2$ (M^+) 433.1466, found 433.1453. Yield: 52%.

1-(2-aminophenyl)-2-(biphenyl)-4,5-diphenylpyridinium perchlorate (1f): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.44 (s, 1H), 8.09 (s, 1H), 7.65 – 7.51 (m, 6H), 7.39 (ddt, *J* = 31.0, 23.4, 7.6 Hz, 13H), 7.26 (s, 2H), 7.19 (t, *J* = 7.8 Hz, 1H), 7.05 (d, *J* = 8.0 Hz, 1H), 6.77 – 6.60 (m, 2H); ^{13}C NMR (100 MHz, CD_3CN) *δ*:159.19, 154.48, 146.37, 143.42, 142.17, 140.18, 139.45, 135.47, 133.26, 132.01, 130.02, 129.90, 129.80, 128.98, 128.79, 127.21, 127.14, 117.49; HRMS calcd for $C_{35}H_{27}N_2$ (M^+) 475.2169, found 475.2168. Yield: 80%.

1-(2-aminophenyl)-1,2,4,5-tetraphenylpyridinium perchlorate (1g): ^{1}H NMR (400 MHz, DMSO) δ 9.24 (s, 1H), 7.53 – 7.20 (m, 10H), 7.07 (ddd, *J* = 32.1, 14.8, 6.9 Hz, 15H), 6.67 (d, *J* = 8.2 Hz, 1H), 6.49 (t, *J* = 7.6 Hz, 1H), 5.80 (s, 2H); ^{13}C NMR (100 MHz, CD_3CN) *δ*: 157.21, 154.76, 146.09, 143.07, 142.82, 140.48, 135.51, 134.43, 131.60, 130.63, 130.32, 128.85, 128.42, 128.28, 128.10, 127.87, 127.41, 116.54, 115.69; HRMS calcd for $C_{35}H_{27}N_2$ (M^+) 475.2169, found 475.2163. Yield: 89%.

1-(4-methyl-2-aminophenyl)-2,3,4,5-tetraphenylpyridinium perchlorate (1h): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.52 (d, *J* = 7.3 Hz, 1H), 8.02 (s, 1H), 7.52 (d, *J* = 7.7 Hz, 1H), 7.35 (d, *J* = 5.8 Hz, 3H), 7.30 – 7.16 (m, 6H), 7.16 – 6.67 (m, 12H), 6.55 (d, *J* = 8.6 Hz, 1H), 6.44 (s, 1H), 6.36 (d, *J* = 8.0 Hz, 1H), 5.30 (s, 1H), 2.14 (d, *J* = 21.1 Hz, 3H); ^{13}C NMR (100 MHz, CD_3CN): *δ*=144.68,

142.22, 141.72, 135.29, 132.06, 132.06, 131.33, 131.11, 130.90, 130.04, 129.89, 128.58, 127.67, 118.29, 117.27, 21.48; HRMS calcd for $C_{36}H_{29}N_2$ (M^+) 489.2325, found 489.2317. Yield: 70%.

1-(4-chloro-2-aminophenyl)-2,3,4,5-tetraphenylpyridinium perchlorate (1i): ^{1}H NMR (400 MHz, DMSO) δ 9.31 (d, *J* = 6.5 Hz, 1H), 7.52 (d, *J* = 2.4 Hz, 1H), 7.44 – 7.28 (m, 6H), 7.28 – 7.21 (m, 2H), 7.21 – 7.06 (m, 6H), 7.06 – 6.92 (m, 7H), 6.73 – 6.62 (m, 1H), 6.55 (dd, *J* = 8.5, 2.2 Hz, 1H), 6.06 (d, *J* = 47.9 Hz, 2H); ^{13}C NMR(100 MHz, CD_3CN): *δ*=157.12, 154.43, 145.80, 144.02, 142.25, 139.83, 135.11, 133.74, 130.84, 129.98, 129.70, 129.16, 128.35, 127.79, 127.37, 125.31, 114.70; HRMS (ESI) calcd. for $C_{35}H_{25}NCl$ (M^+) 509.1779; found 509.1796. Yield: 52%.

1-(5-ester-2-aminophenyl)-2, 4,5-triphenylpyridinium perchlorate (1k): ^{1}H NMR (500 MHz, DMSO) δ 9.20 (s, 1H), 8.37 (s, 1H), 8.01 (d, *J* = 1.6 Hz, 1H), 7.70 (dd, *J* = 8.7, 1.9 Hz, 1H), 7.56 (d, *J* = 7.3 Hz, 2H), 7.46 (dt, *J* = 14.4, 9.8 Hz, 11H), 7.38 – 7.26 (m, 2H), 6.76 (d, *J* = 8.7 Hz, 1H), 6.60 (s, 2H). ^{13}C NMR(100 MHz, CD_3CN): *δ*=165.299, 156.550, 154.306, 148.021, 147.354, 138.551, 135.254, 133.654, 132.244, 131.410, 131.046, 130.818, 130.341, 130.273, 129.787, 129.522, 129.211, 129.029, 128.878, 128.787, 128.339, 125.078, 116.080, 115.648, 51.676; HRMS calcd for $C_{31}H_{25}N_2O_2$ (M^+) 457.1916, found 457.1928. Yield: 68%.

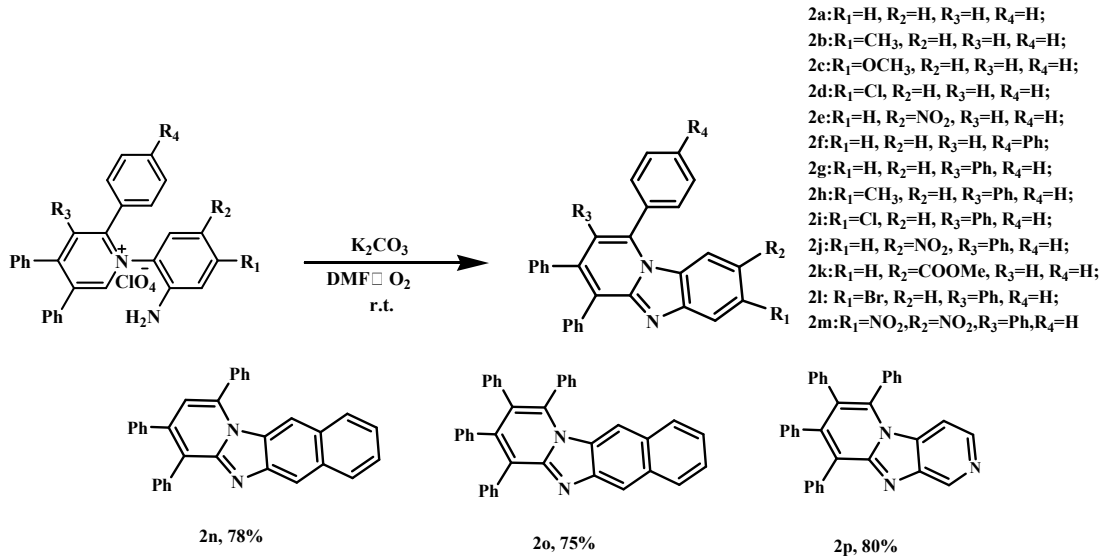
1-(4-bromo-2-aminophenyl)-2, 3,4,5-tetraphenylpyridinium perchlorate (1l): ^{1}H NMR (500 MHz, DMSO) δ 9.30 (d, *J* = 8.0 Hz, 1H), 7.67 – 7.53 (m, 1H), 7.39 – 7.23 (m, 8H), 7.19 – 7.09 (m, 6H), 7.06 – 6.93 (m, 7H), 6.86 (d, *J* = 1.8 Hz, 1H), 6.72 – 6.58 (m, 1H), 6.05 (d, *J* = 40.3 Hz, 2H). ^{13}C NMR(100 MHz, DMSO): *δ*=157.058,154.435, 145.814, 144.275, 142.357, 140.075, 134.920, 134.556, 133.866, 130.947, 130.318, 130.091, 129.818, 129.666, 129.257, 128.718, 128.423, 127.869, 127.596, 127.452, 127.285, 125.974, 123.904, 117.877, 117.649; HRMS calcd for $C_{35}H_{26}N_2Br$ (M^+) 553.1279, found 553.1226. Yield: 55%.

1-(2-aminonathphyl)-2,4,5-triphenylpyridinium perchlorate (1n): ^{1}H NMR (500 MHz, DMSO) δ 9.26 (s, 1H), 8.44 (s, 1H), 7.99 (s, 1H), 7.62 (dd, *J* = 14.5, 7.7 Hz, 4H), 7.56 – 7.49 (m, 1H), 7.47 (s, 4H), 7.43 (d, *J* = 4.3 Hz, 3H), 7.38 (t, *J* = 9.4 Hz, 6H), 7.18 (t, *J* = 7.4 Hz, 1H), 7.09 (s, 1H), 5.82 (d, *J* = 63.8 Hz, 2H); ^{13}C NMR(100 MHz, CD_3CN): *δ*=156.60, 154.22, 147.77, 140.83, 135.22, 133.60, 129.81, 129.58, 129.46, 129.40, 128.86, 128.78, 128.31, 128.05, 127.64, 125.29, 122.59, 109.32; HRMS (ESI) calcd. for $C_{33}H_{25}N_2$ (M^+) 449.2012; found 449.2023. Yield: 82%.

1-(2-aminonathphyl)-2,3,4,5-tetraphenylpyridinium perchlorate (1o): ^{1}H NMR (400 MHz, DMSO) δ 9.39 (s, 1H), 7.98 (d, *J* = 21.9 Hz, 1H), 7.62 (d, *J* = 8.2 Hz, 1H), 7.55 (d, *J* = 8.4 Hz, 1H), 7.43 (d, *J* = 7.4 Hz, 1H), 7.35 (t, *J* = 8.2 Hz, 5H), 7.28 (d, *J* = 3.7 Hz, 2H), 7.17 (d, *J* = 3.1 Hz, 4H), 7.11 – 6.88 (m, 11H), 5.93 (s, 2H); ^{13}C NMR(100 MHz, CD_3CN): *δ*=157.16, 154.42, 148.24, 146.04, 142.57, 140.57, 139.53, 134.91, 134.46, 133.73, 131.23, 130.25, 129.75, 129.21, 128.35, 127.81, 127.38, 125.32, 124.59, 122.82; HRMS (ESI) calcd. for $C_{39}H_{29}N_2$ (M^+) 525.2325; found 525.2326. Yield: 82%.

1-(2-aminopyridyl)-2,3,4,5-tetraphenylpyridinium perchlorate (1p): ^{1}H NMR (500 MHz, DMSO) δ 9.37 (s, 1H), 8.31 (s, 1H), 7.92 (d, *J* = 5.7 Hz, 1H), 7.40 – 7.21 (m, 6H), 7.14 (t, *J* = 7.4 Hz, 5H), 7.09 – 6.92 (m, 6H), 6.75 (s, 2H), 6.55 (d, *J* = 5.7 Hz, 1H). ^{13}C NMR(100 MHz, DMSO): *δ*= 138.87, 136.37, 131.83, 129.92, 129.47, 127.79, 123.97, 121.77, 116.51, 116.15, 115.49, 112.54, 111.94, 111.74, 111.49, 111.36, 110.93, 110.44, 110.13, 109.58, 109.51, 109.32, 109.15, 109.09, 106.15, 91.91; HRMS calcd for $C_{34}H_{26}N_3$ (M^+) 476.2127, found 476.2109. Yield: 70%.

General Procedure for the Synthesis of pyrido [1,2-a]benzimidazole derivatives (2a,2b,2c,2d,2e,2f,2g,2h,2i,2j,2k,2l,2m,2n,2o and 2p)[2]:



A solution of 2,4,5-triphenylpyridinium perchlorate **1a** (0.20g, 0.40mmol) was dissolved in 5mL DMF and equivalent of K_2CO_3 was added. The solution was stirred at room temperature for 12hours under the presence of air. Then, the solvent was diluted with CH_2Cl_2 (50ml), and washed with water (3×20ml). The organic layer was seperated, dryed and concentrated in vacuo. The mixture was purified by flash column chromatography with CH_2Cl_2, to afford the corresponding product **2a**. All the targeted compounds synthesized and purified with the same procedure as 2a. (using dichloromethane as the eluent in column chromatography for all synthesized compounds)

1,3,4-triphenyl-pyrido[1,2-*a*]benzimidazole (2a): ^{1}H NMR(400 MHz, $CDCl_3$) δ: 7.93 (d, J=8 Hz, 1H), 7.54-7.74 (m, 5H), 7.47 (m, 2H), 7.30-7.42 (m, 4H), 7.23 (s, 5H), 6.97 (m, 1H), 6.85 (s, 1H), 6.64 (d, J=8 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ: 140.2, 139.8, 131.5, 130.2, 129.9, 129.3, 128.3, 128.2, 127.9, 127.6, 125.0, 120.5, 115.6, 114.7. HRMS (ESI) m/z calcd for $C_{29}H_{21}N_2$ $[M+H]^+$, 397.1750; found, 397.1712. Yield: 65%.

7-methyl-1,3,4-triphenyl-pyrido[1,2-*a*]benzimidazole (2b): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (s, 1H), 7.69 – 7.55 (m, 6H), 7.46 (dd, J = 8.0, 1.4 Hz, 3H), 7.36 (t, J = 1.8 Hz, 1H), 7.34 (s, 1H), 7.31 (dd, J = 3.9, 2.3 Hz, 2H), 7.29 (t, J = 3.1 Hz, 1H), 7.27 – 7.24 (m, 2H), 7.24 – 7.16 (m, 6H), 6.86 – 6.76 (m, 2H), 6.51 (d, J = 8.6 Hz, 1H), 2.45 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 149.93, 146.04, 139.81, 139.45, 135.59, 134.88, 134.59, 134.54, 131.37, 130.01, 129.77, 129.20, 128.15, 128.09, 127.65, 127.60, 127.35, 126.82, 122.17, 119.74, 115.19, 114.35, 114.00, 21.68; HRMS (ESI) m/z calcd for $C_{30}H_{23}N_2$ $[M+H]^+$, 411.1783; found, 411.1863. Yield: 62%.

7-methoxyl-1,3,4-triphenyl-pyrido[1,2-*a*]benzimidazole (2c): ^{1}H NMR (400 MHz, $CDCl_3$) δ

7.72 – 7.55 (m, 5H), 7.50 – 7.40 (m, 2H), 7.36 (dd, *J* = 5.6, 2.2 Hz, 2H), 7.31 (ddd, *J* = 10.0, 6.4, 4.0 Hz, 2H), 7.26 (s, 2H), 7.21 (d, *J* = 7.3 Hz, 5H), 6.84 (s, 1H), 6.61 (dd, *J* = 9.2, 2.5 Hz, 1H), 6.51 (d, *J* = 9.2 Hz, 1H), 3.83 (s, 3H);^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 152.4, 150.7, 146.3, 140.2, 139.3, 131.2, 130.2, 129.7, 129.3, 129.1, 128.4, 128.1, 127.6, 115.1, 55.7;MS *m/z* (%): 427.2 ([M+H]$^+$, 100). HRMS (ESI) m/z calcd for $C_{30}H_{23}N_2O$ [M+H]$^+$, 427.1732; found, 427.1824. Yield: 66%.

7-chloro-1,3,4-triphenyl-pyrido[1,2-*a*]benzimidazole (2d)： ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (s, 1H), 7.68 – 7.55 (m, 6H), 7.50 – 7.40 (m, 3H), 7.31 (ddd, *J* = 10.7, 10.1, 5.2 Hz, 4H), 7.26 (s, 1H), 7.21 (d, *J* = 8.6 Hz, 5H), 6.89 – 6.74 (m, 2H), 6.51 (d, *J* = 8.6 Hz, 1H);^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 151.1, 147.3, 144.2, 138.2, 137.9, 133.1, 129.8, 129.3, 127.6, 126.7, 126.3, 124.7, 124.0, 122.1, 118.8, 116.0; HRMS (ESI) m/z calcd for $C_{29}H_{20}ClN_2$ [M+H]$^+$, 431.1237; found, 431.1321. Yield: 72%.

8-nitro-1,3,4-triphenyl-pyrido[1,2-*a*]benzimidazole (2e)： ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.29 (dd, *J* = 9.1, 2.2 Hz, 1H), 7.93 (d, *J* = 9.1 Hz, 1H), 7.81 – 7.75 (m, 1H), 7.75 – 7.63 (m, 4H), 7.59 (d, *J* = 2.1 Hz, 1H), 7.46 (dd, *J* = 7.9, 1.6 Hz, 2H), 7.42 – 7.31 (m, 3H), 7.31 – 7.19 (m, 6H), 7.06 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 153.2, 149.4, 143.2, 140.8, 140.3, 138.5, 134.4, 132.9, 131.2, 131.1, 130.9, 129.9, 129.6, 129.4, 129.2, 128.7, 128.4, 128.3, 128.2, 128.0, 127.8, 127.5, 120.8, 119.7, 117.0, 112.0;HRMS (ESI) m/z calcd for $C_{29}H_{21}N_2$ [M+H]$^+$, 442.1556; found, 441.1549. Yield: 86%.

1-biphenyl-3,4-diphenyl-pyrido[1,2-*a*]benzimidazole (2f): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 (d, *J* = 8.2 Hz, 1H), 7.86 (d, *J* = 8.0 Hz, 2H), 7.74 (t, *J* = 8.4 Hz, 4H), 7.53 (t, *J* = 7.6 Hz, 2H), 7.50 – 7.26 (m, 7H), 7.23 (s, 5H), 7.00 (t, *J* = 7.8 Hz, 1H), 6.90 (s, 1H), 6.85 (d, *J* = 8.5 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 143.78, 142.93, 140.30, 140.04, 139.40, 131.32, 129.75, 129.58, 129.07, 128.19, 128.12, 127.68, 127.45, 127.23, 120.49, 114.64; HRMS (ESI) m/z calcd for $C_{35}H_{25}N_2$ [M+H]$^+$, 473.1939; found, 473.2029. Yield: 63%.

1,2,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2g)： ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.96 (d, *J* = 8.1 Hz, 2H), 7.38 (dd, *J* = 22.3, 7.4 Hz, 9H), 7.26 (dt, *J* = 14.0, 7.2 Hz, 4H), 6.90 (d, *J* = 18.1 Hz, 10H), 6.11 (d, *J* = 8.5 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 133.93, 131.69, 131.11, 130.78, 130.41, 128.92, 127.91, 127.05, 120.68, 119.62; HRMS (ESI) m/z calcd for $C_{35}H_{25}N_2$ [M+H]$^+$, 473.1939; found, 473.2014. Yield: 60%.

7-methyl-1,2,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2h)： ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.69 (s, 1H), 7.50 – 7.28 (m, 7H), 7.28 – 7.21 (m, 3H), 7.18 (t, *J* = 7.3 Hz, 1H), 6.94 – 6.76 (m, 9H), 6.70 (d, *J* = 10.0 Hz, 1H), 5.95 (d, *J* = 8.7 Hz, 1H), 2.40 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 147.91, 145.00, 140.73, 137.17, 136.53, 135.80, 135.06, 133.68, 132.95, 130.70, 130.14, 129.82, 129.37, 128.18, 127.81, 127.18, 127.15, 126.75, 126.21, 125.95, 125.19, 125.10, 124.65, 121.10, 118.62, 113.12, 108.79, 20.66; HRMS (ESI) m/z calcd for $C_{36}H_{27}N_2$ [M+H]$^+$, 487.2096; found, 487.2189. Yield: 65%.

7-chloro-1,2,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2i)： ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.91 (d, *J* = 1.7 Hz, 1H), 7.49 – 7.33 (m, 7H), 7.25 (tt, *J* = 14.3, 7.1 Hz, 4H), 6.96 – 6.80 (m, 11H), 5.99 (d, *J* = 9.0 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 142.94, 131.60, 131.07, 130.73, 130.28, 129.55, 129.08, 127.92, 127.12, 127.10, 126.47, 126.37, 120.93, 119.45, 115.49; HRMS (ESI) m/z calcd for $C_{35}H_{24}ClN_2$ [M+H]$^+$, 507.1550; found, 507.1631. Yield: 70%.

8-nitro-1,2,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2j)： ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.23 (dd, *J* = 9.1, 1.9 Hz, 1H), 7.88 (d, *J* = 9.1 Hz, 1H), 7.53 (dt, *J* = 19.5, 7.0 Hz, 3H), 7.40 (d, *J*

= 7.7 Hz, 4H), 7.34 – 7.10 (m, 4H), 7.02 (d, *J* = 1.2 Hz, 1H), 6.95 (s, 8H), 6.89 (d, *J* = 2.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 152.26, 149.48, 144.97, 131.33, 130.91, 130.46, 130.17, 129.63, 129.59, 127.89, 127.68, 127.21, 127.15, 126.69, 126.61, 120.44, 119.46, 112.35; HRMS (ESI) m/z calcd for $C_{35}H_{24}N_3O_2$ $[M+H]^+$, 518.1790; found, 518.1881. Yield: 88%.

8-ester-1,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2k)： ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.06 (dd, *J* = 8.7, 1.5 Hz, 1H), 7.90 (d, *J* = 8.7 Hz, 1H), 7.76 – 7.63 (m, 5H), 7.50 – 7.43 (m, 2H), 7.43 – 7.29 (m, 4H), 7.25 (d, *J* = 6.3 Hz, 5H), 6.96 (s, 1H), 3.81 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 152.006, 148.941, 141.904, 140.303, 139.103, 135.187, 133.908, 131.411, 130.602, 129.838, 129.642, 129.337, 129.078, 128.391, 128.337, 128.050, 127.846, 127.286, 126.208, 121.974, 119.636, 117.457, 116.206; HRMS (ESI) m/z calcd for $C_{31}H_{23}N_2O_2$ $[M+H]^+$, 455.1760; found, 455.1754. Yield: 78%.

7-bromo-1,2,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2l)： ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.06 (d, *J* = 1.7 Hz, 1H), 7.54 – 7.33 (m, 8H), 7.24 (dt, *J* = 14.6, 6.0 Hz, 4H), 7.05 – 6.77 (m, 11H), 5.94 (d, *J* = 9.0 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 149.44, 146.58, 142.98, 137.81, 137.66, 136.40, 135.55, 133.48, 131.58, 131.05, 130.70, 130.25, 129.98, 129.53, 129.06, 128.98, 128.90, 128.53, 128.40, 127.89, 127.51, 127.10, 127.08, 126.63, 126.45, 126.35, 123.49, 122.61, 118.43, 115.82; HRMS (ESI) m/z calcd for $C_{35}H_{24}N_2Br$ $[M+H]^+$, 551.1123; found, 551.1138. Yield: 67%.

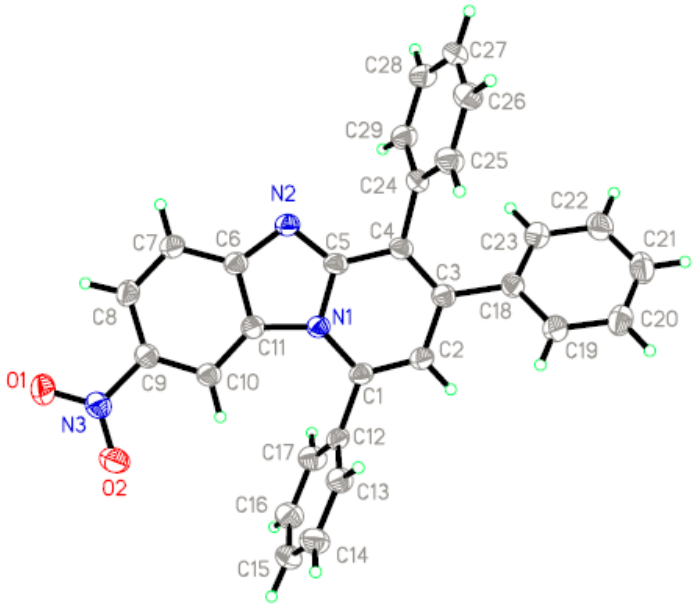
7,8-dinitro-1,3,4-tetraphenyl-pyrido[1,2-*a*]benzimidazole (2m)： ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.26 (d, *J* = 23.2 Hz, 1H), 7.78 – 7.70 (m, 3H), 7.65 (dd, *J* = 8.1, 1.3 Hz, 2H), 7.46 – 7.35 (m, 5H), 7.31 – 7.23 (m, 7H), 7.15 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 154.72, 147.04, 144.06, 142.01, 140.07, 138.03, 134.72, 133.98, 132.31, 131.50, 131.10, 130.14, 129.60, 129.10, 128.69, 128.45, 128.42, 128.37, 127.91, 118.06, 116.35, 113.35; HRMS (ESI) m/z calcd for $C_{29}H_{19}N_4O_4$ $[M+H]^+$, 487.1406; found, 487.1418. Yield: 88%.

1,3,4-triphenyl-Naphth[2,3-*d*]imidazole (2n): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.38 (s, 1H), 7.96 (d, *J* = 8.3 Hz, 1H), 7.69 (dt, *J* = 13.3, 6.8 Hz, 5H), 7.57 (d, *J* = 8.2 Hz, 1H), 7.51 (d, *J* = 7.1 Hz, 2H), 7.44 – 7.14 (m, 10H), 7.06 (s, 1H), 6.85 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 152.78, 144.89, 142.16, 140.79, 139.21, 131.35, 130.25, 129.68, 129.22, 129.20, 128.34, 128.25, 128.16, 127.82, 127.65, 124.53, 116.06, 114.77, 111.83; HRMS (ESI) m/z calcd for $C_{33}H_{23}N_2$ $[M+H]^+$, 447.1783; found, 447.1866. Yield: 78%.

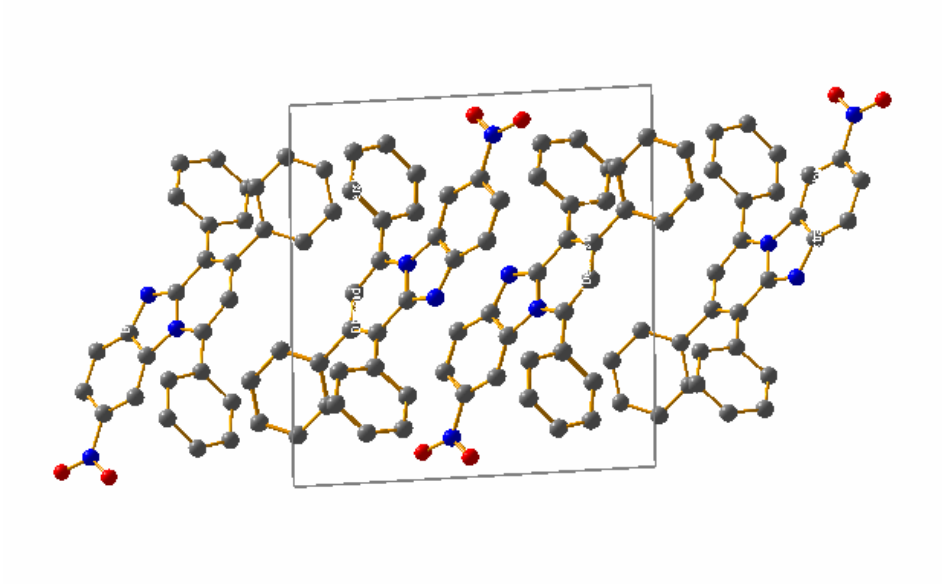
1,2,3,4-tetrahenyl-Naphth[2,3-*d*]imidazole (2o): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.37 (s, 1H), 7.94 (d, *J* = 8.5 Hz, 1H), 7.72 – 7.09 (m, 15H), 7.09 – 6.62 (m, 11H), 6.49 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 171.16, 138.91, 131.75, 131.13, 130.65, 130.42, 129.03, 128.46, 127.12, 127.09,112.32;HRMS (ESI) m/z calcd for $C_{39}H_{27}N_2$ $[M+H]^+$, 523.2096; found, 523.2165. Yield: 75%.

1,2,3,4-tetrapyridyl-pyrido[1,2-*a*]benzimidazole (2p)： ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.42 (d, *J* = 5.8 Hz, 1H), 7.78 (d, *J* = 5.8 Hz, 1H), 7.50 – 7.44 (m, 3H), 7.41 (dd, *J* = 10.4, 5.8 Hz, 4H), 7.32 – 7.21 (m, 4H), 6.99 – 6.92 (m, 5H), 6.90 (dd, *J* = 7.1, 2.5 Hz, 2H), 6.87 (dd, *J* = 6.6, 3.0 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$) *δ*: 150.760, 150.191, 144.856, 143.574, 138.584, 137.770, 137.706, 136.255, 135.519, 133.622, 131.625, 131.170, 130.747, 130.156, 129.951, 129.733, 128.810, 128.678, 128.105, 127.786, 127.368, 127.327, 126.813, 126.708, 114.510; HRMS (ESI) m/z calcd for $C_{34}H_{24}N_3$ $[M+H]^+$, 474.1970; found, 474.1956. Yield: 80%.

Crystal Structure Data



ORTEP views (ellipsoid at 30% probability level) of compound **2e**



Crystal packing of **2e**

Table 1 Crystallographic data and structure refinements for **2e**

Identification code	**2e**
Empirical formula	$C_{29}H_{19}ClN_3O_2$
Formula weight	441.47
Temperature (K)	273(2)
Wavelength (Å)	0.71073
Crystal system	Triclinic
space group	*P*-1
a (Å)	7.9933(15)

b (Å)	11.481(2)
c (Å)	12.651(2)
α (º)	86.659(14)
β (º)	72.316(13)
γ (º)	86.751(14)
Volume (Å^3)	1103.3(4)
Z	2
D_{Calc} (mg/m^{-3})	1.329
μ (mm^{-1})	0.085
$F_{(000)}$	460
Data / restraints / parameters	4083 / 0 / 307
GOF on F^2	0.989
R_1 [I>2σ(I)] [a]	0.0574
wR_2 [I>2σ(I)] [a]	0.1517
R_1 (all data) [a]	0.0914
wR_2 (all data) [a]	0.1823

* $R_1 = \Sigma||F_o| - |F_c|| / \Sigma|F_o|$; $wR_2 = \{\Sigma[w(F_o^2 - F_c^2)^2] / \Sigma[w(F_o^2)]^2\}^{1/2}$

Table 2 Selected bond lengths (Å) for compound **2e**

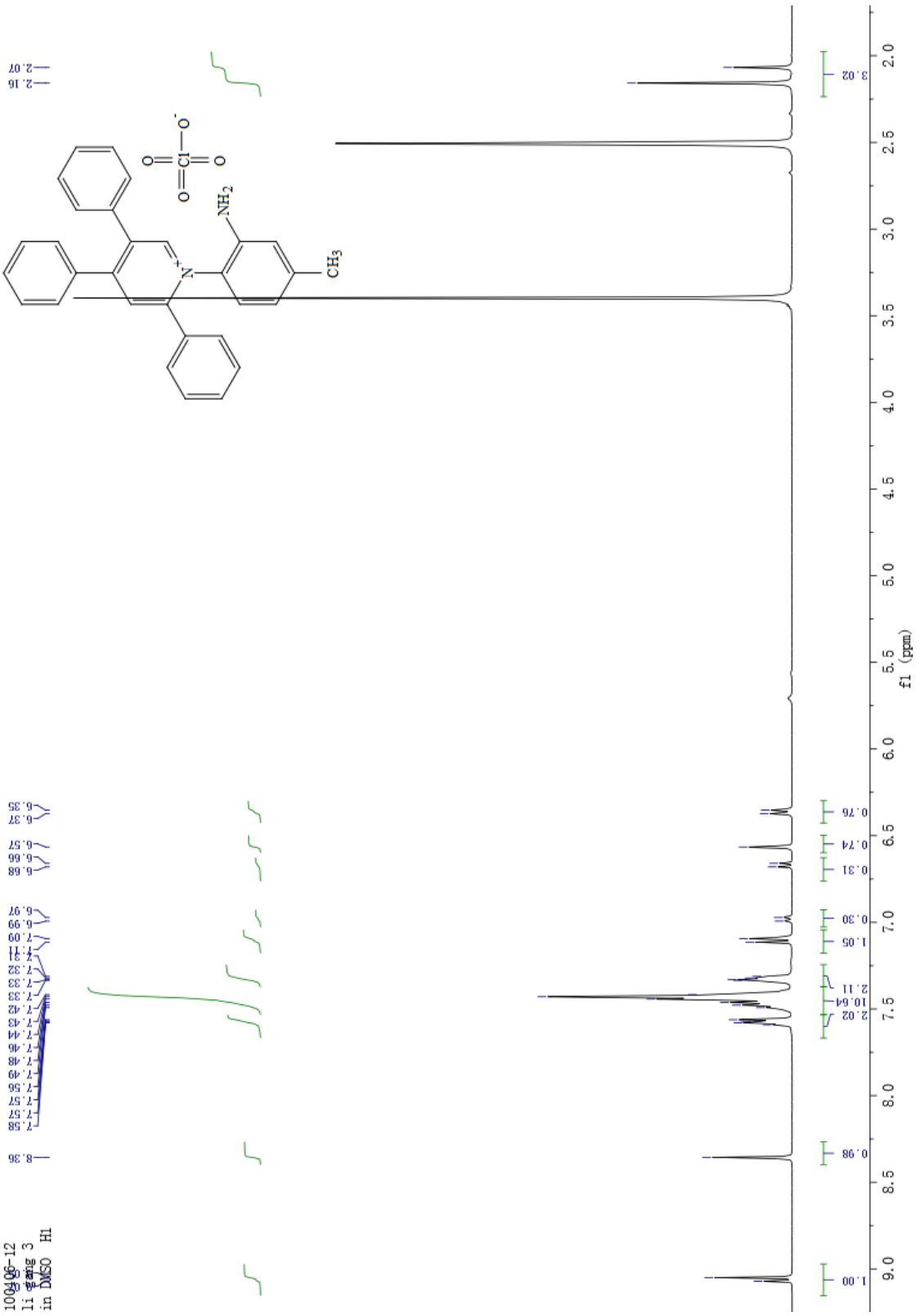
C(1)-C(2)	1.346(3)	C(1)-N(1)	1.384(3)
C(3)-C(4)	1.374(3)	C(2)-C(3)	1.433(3)
C(1)-C(12)	1.490(3)	N(3)-O(1)	1.216(3)
C(9)-N(3)	1.466(3)	C(11)-N(1)	1.399(3)
C(4)-C(5)	1.431(3)	C(3)-C(18)	1.503(3)
C(5)-N(1)	1.401(3)	C(5)-N(2)	1.328(3)
C(6)-N(2)	1.371(3)	C(4)-C(24)	1.494(3)
C(9)-C(10)	1.380(3)	C(10)-C(11)	1.384(3)
C(8)-C(9)	1.392(3)	C(7)-C(8)	1.374(3)
C(6)-C(11)	1.412(3)	C(6)-C(7)	1.399(3)

Table 3 Selected angles (°) for compound **2e**

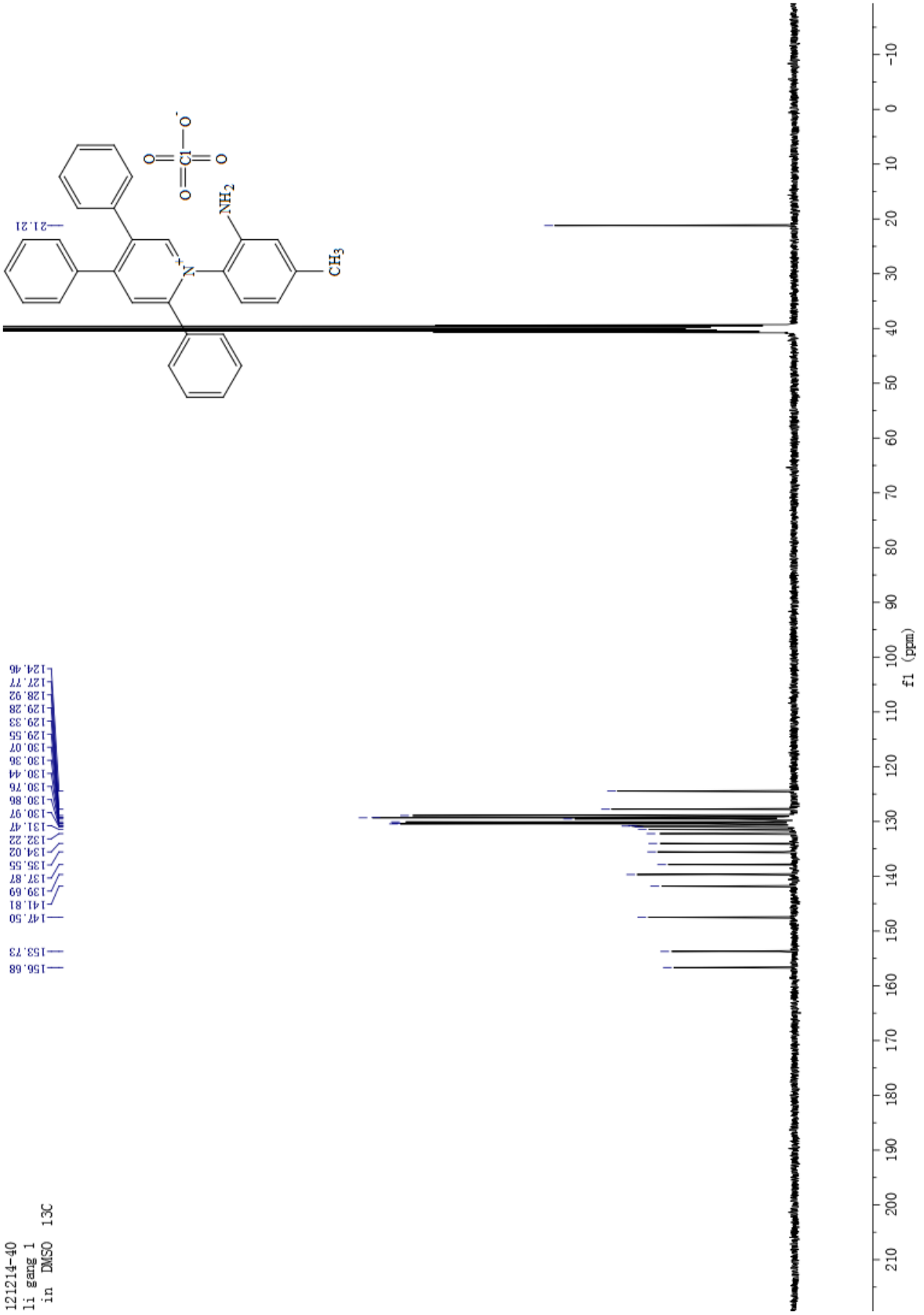
C(2)-C(1)-N(1)	117.80(18)	C(1)-C(2)-C(3)	123.0(2)
C(4)-C(3)-C(2)	119.1(2)	C(3)-C(4)-C(5)	118.71(18)
N(2)-C(5)-N(1)	113.12(18)	N(1)-C(5)-C(4)	118.98(19)
N(2)-C(6)-C(11)	111.86(19)	C(7)-C(6)-C(11)	119.64(19)
C(8)-C(7)-C(6)	118.7(2)	C(7)-C(8)-C(9)	119.8(2)
C(10)-C(9)-C(8)	123.8(2)	C(9)-C(10)-C(11)	115.7(2)
C(10)-C(11)-C(6)	122.2(2)	N(1)-C(11)-C(6)	104.61(17)
C(1)-N(1)-C(5)	122.24(17)	C(5)-N(2)-C(6)	104.60(17)
C(11)-N(1)-C(5)	105.75(17)	O(1)-N(3)-O(2)	122.7(2)

Spectrum Chart

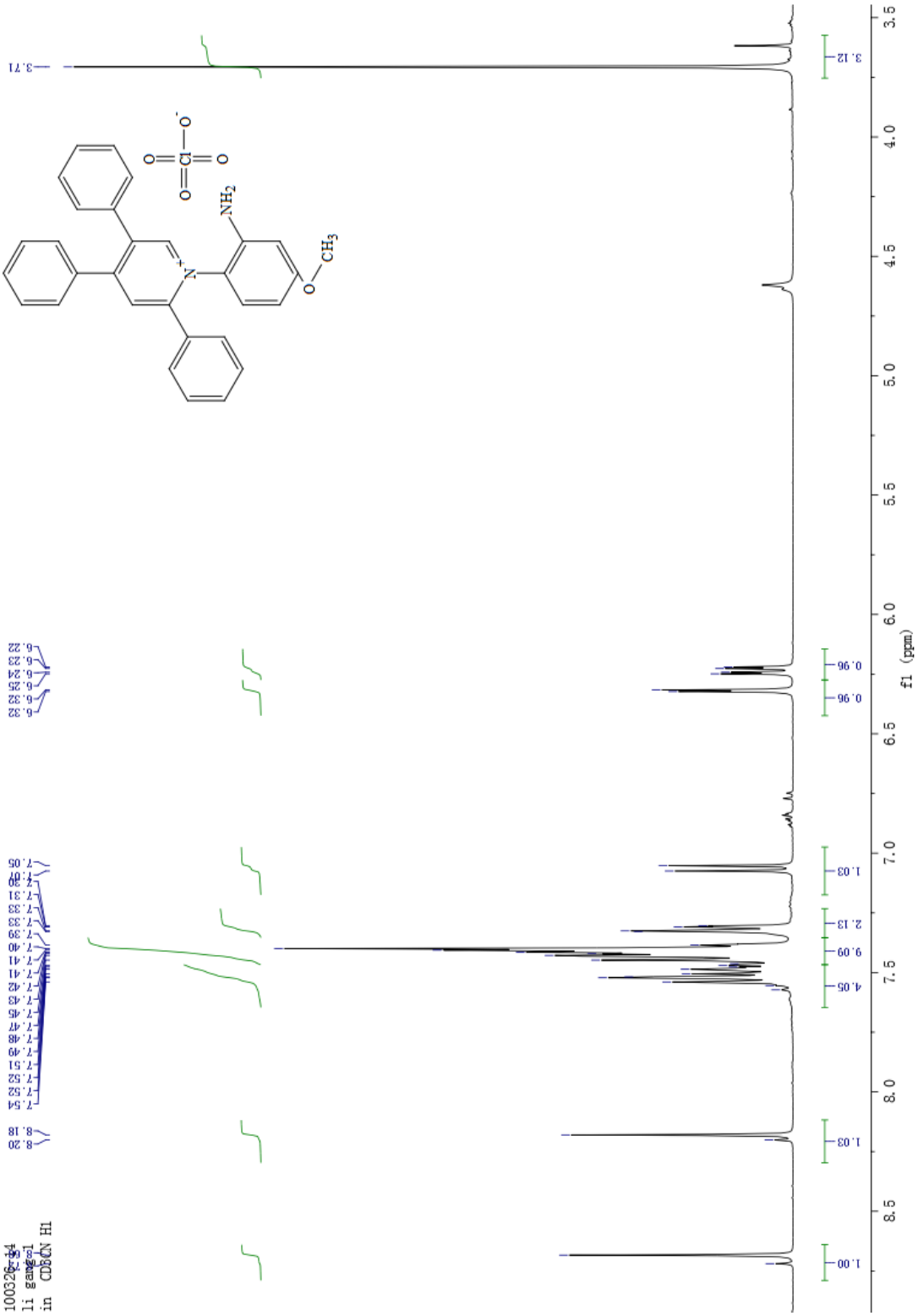
Compound 1b:



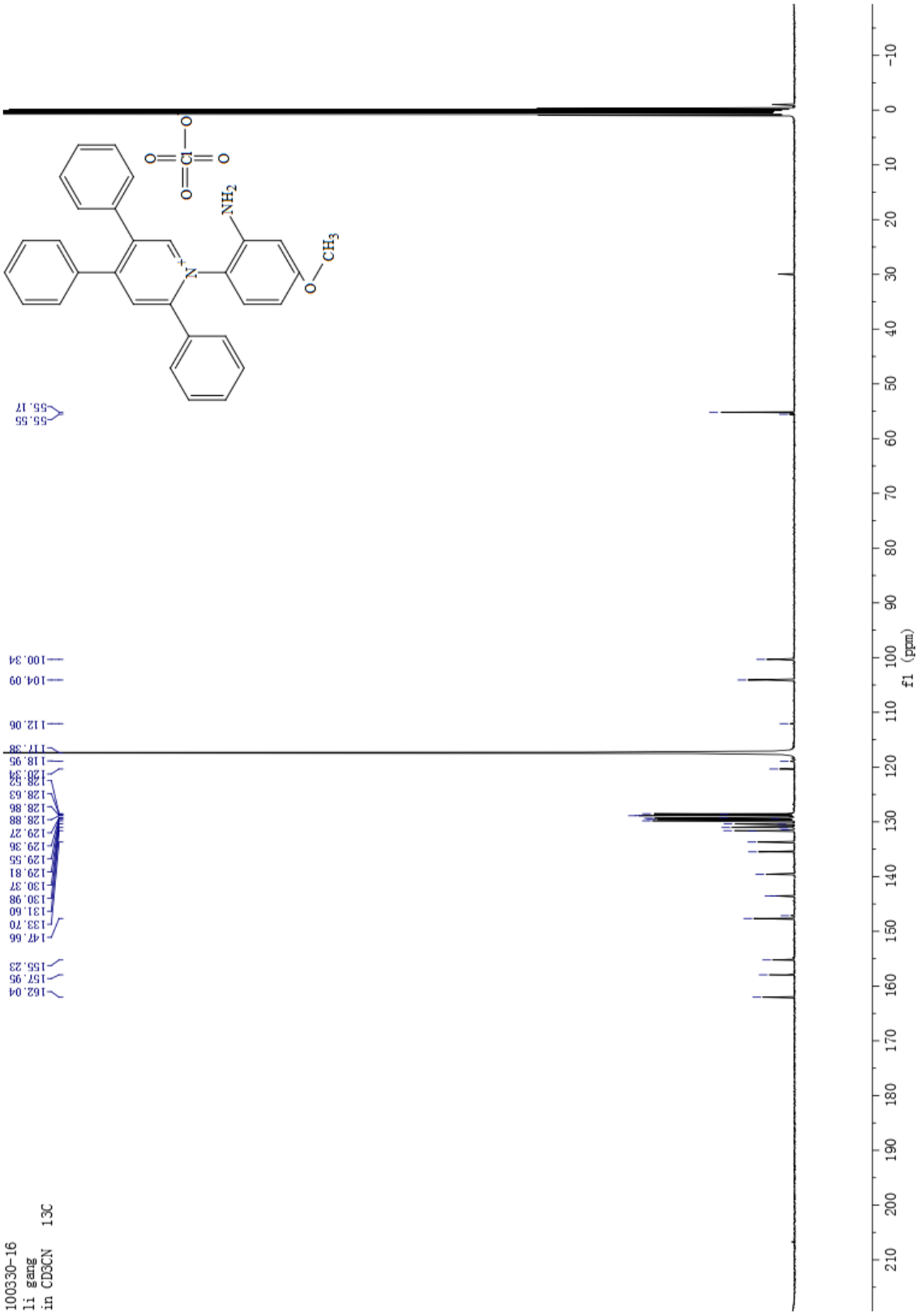
Compound 1b:


121214-40
li gang 1
in DMSO 13C
21.21
124.46
127.77
128.92
129.28
129.33
129.55
130.07
130.36
130.44
130.76
130.86
130.97
131.47
132.22
134.02
135.55
137.87
139.69
141.81
147.50
153.73
156.68
f1 (ppm)

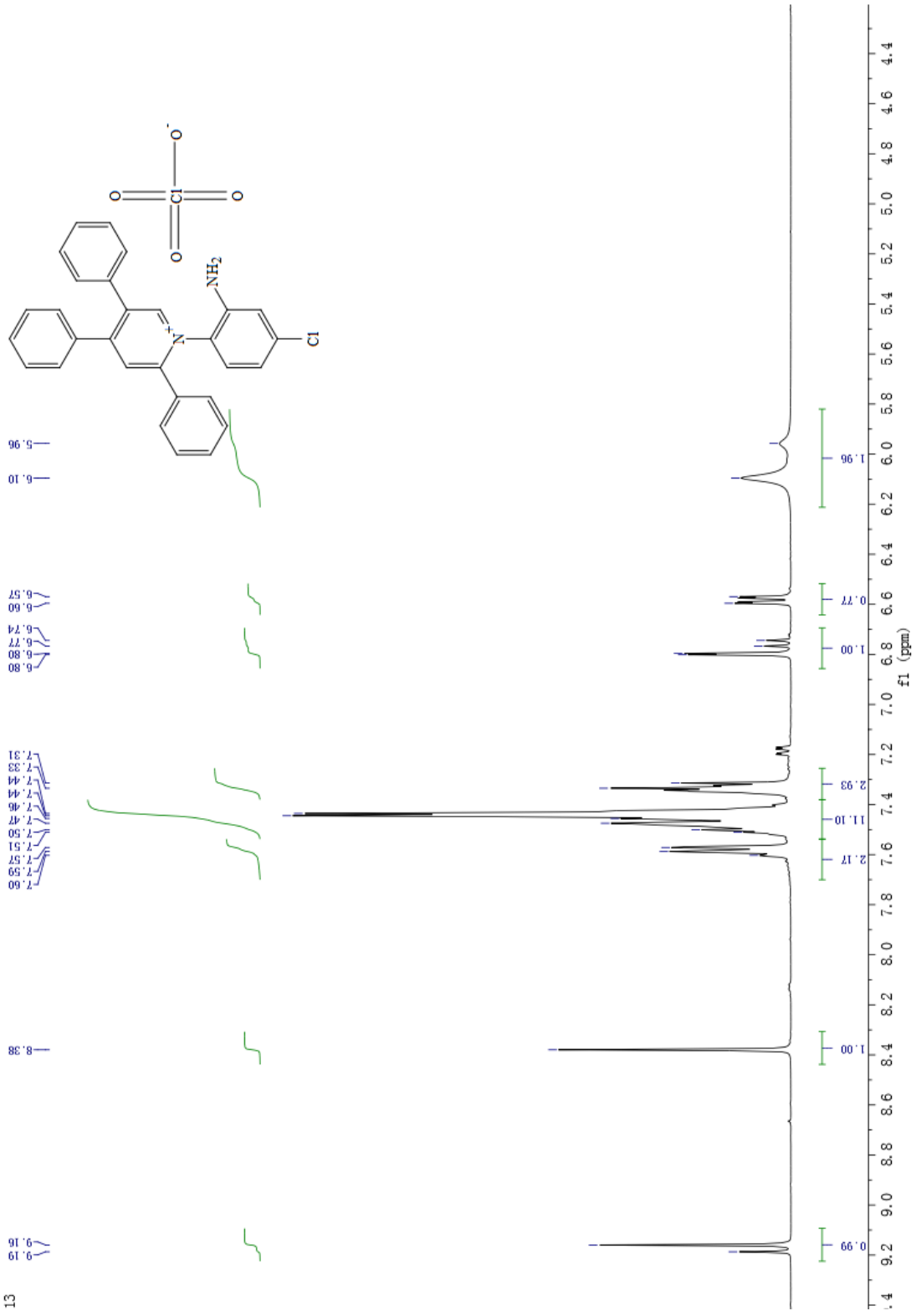
Compound 1c:



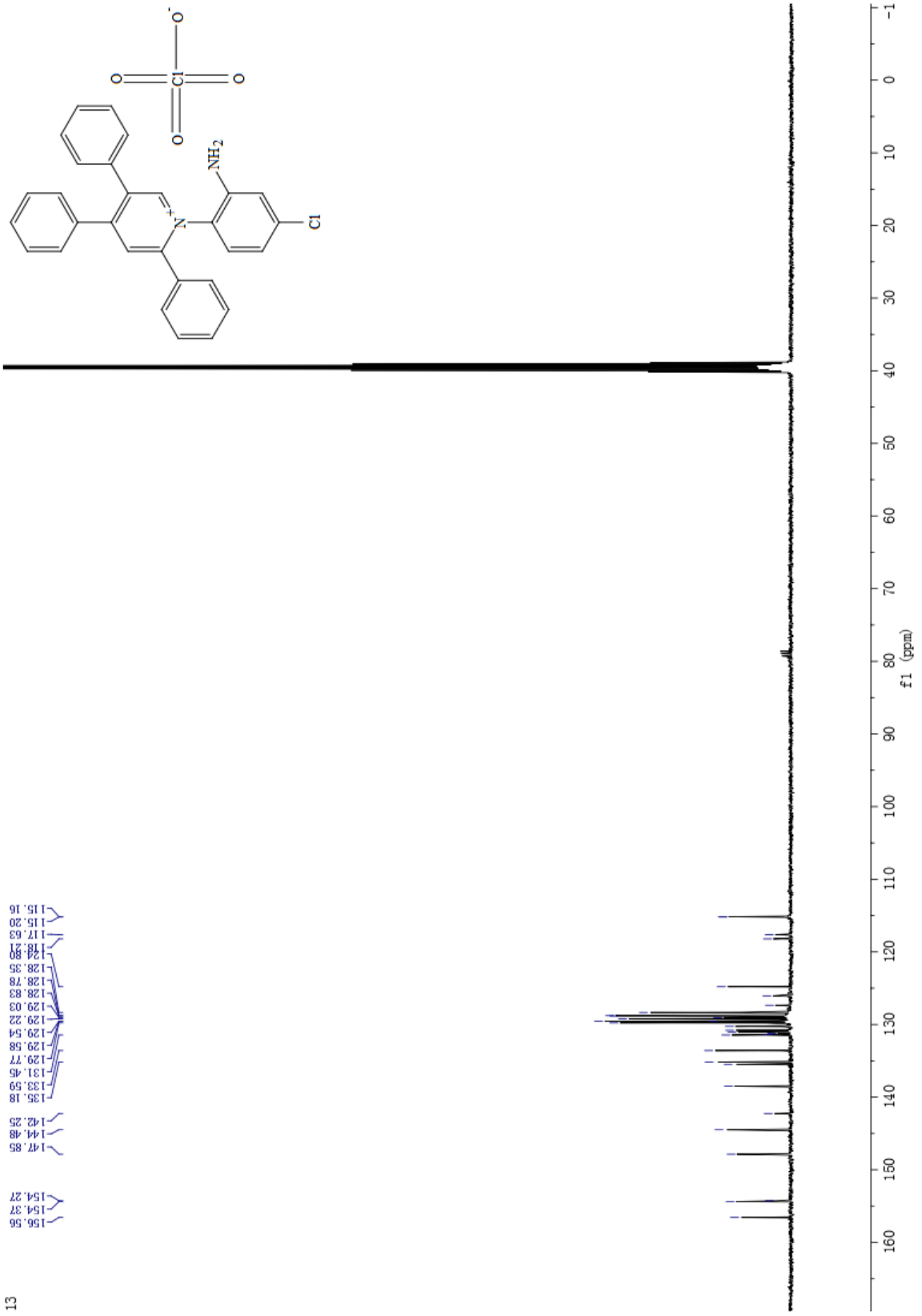
Compound 1c:



Compound 1d:



Compound 1d:


Cl
O
O
O
O
NH2
N+
Cl
13
156.56
154.37
154.27
147.85
144.48
142.25
135.18
133.59
131.45
129.77
129.58
129.54
129.22
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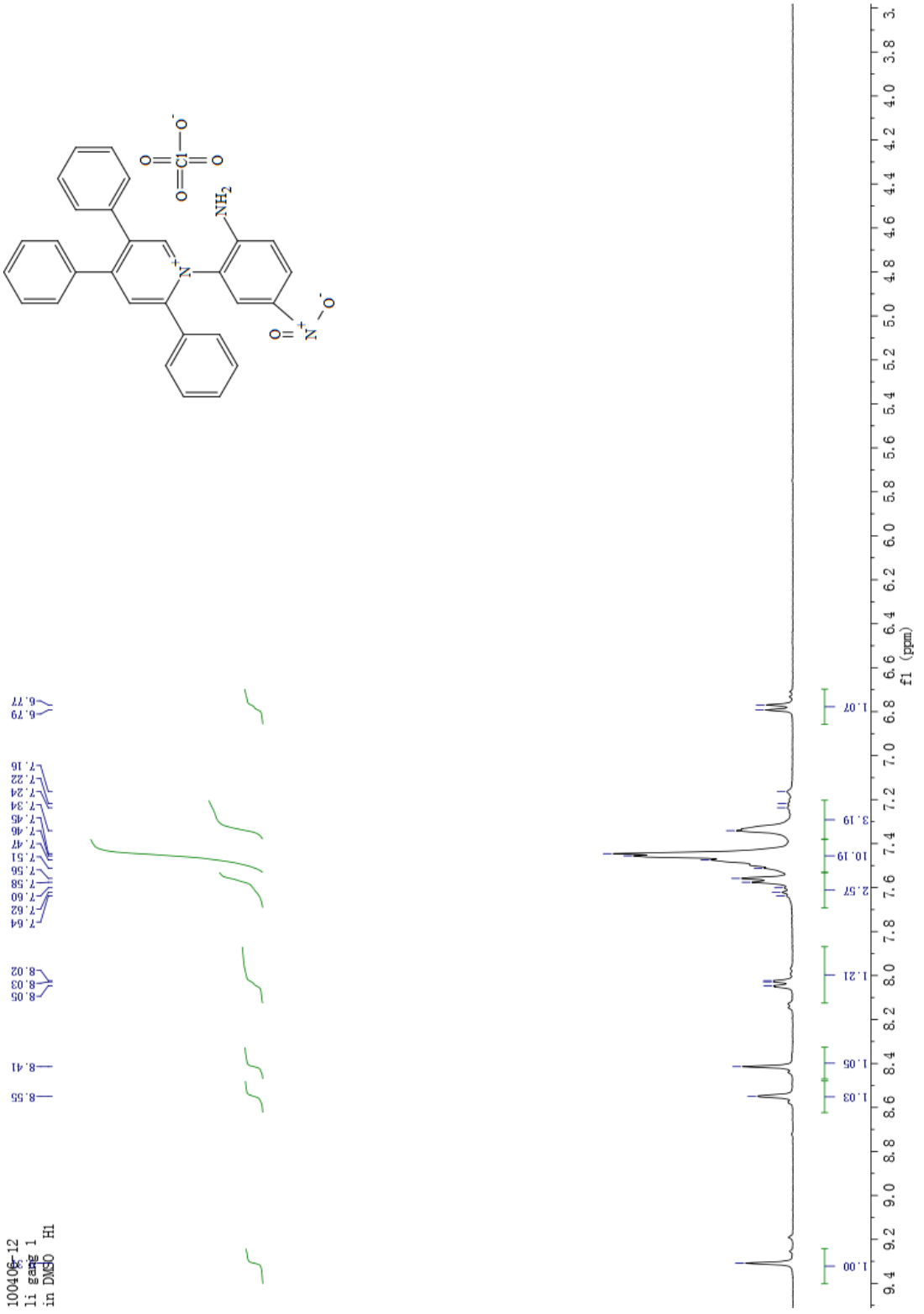
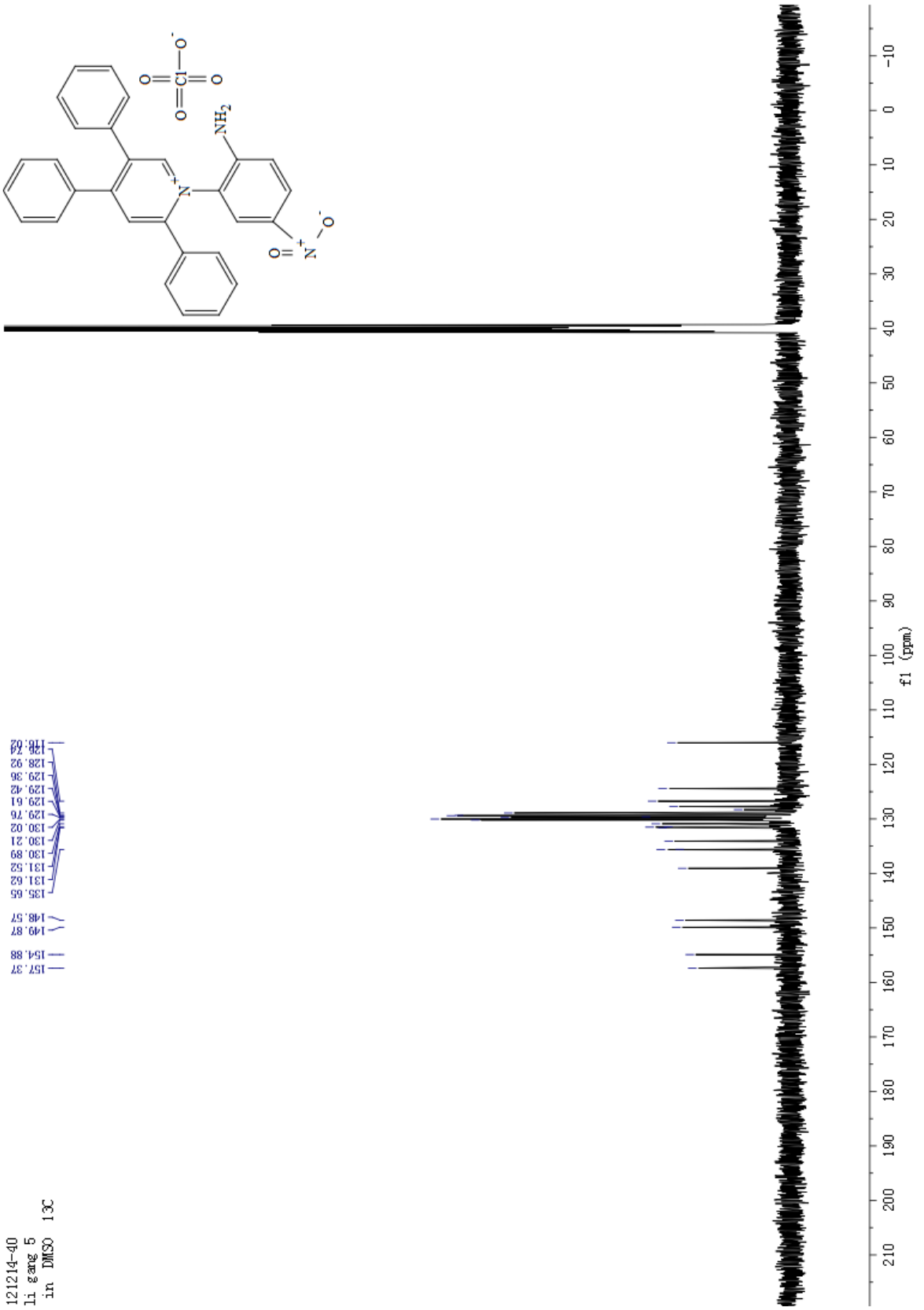
118.21
117.63
115.20
115.16
f1 (ppm)

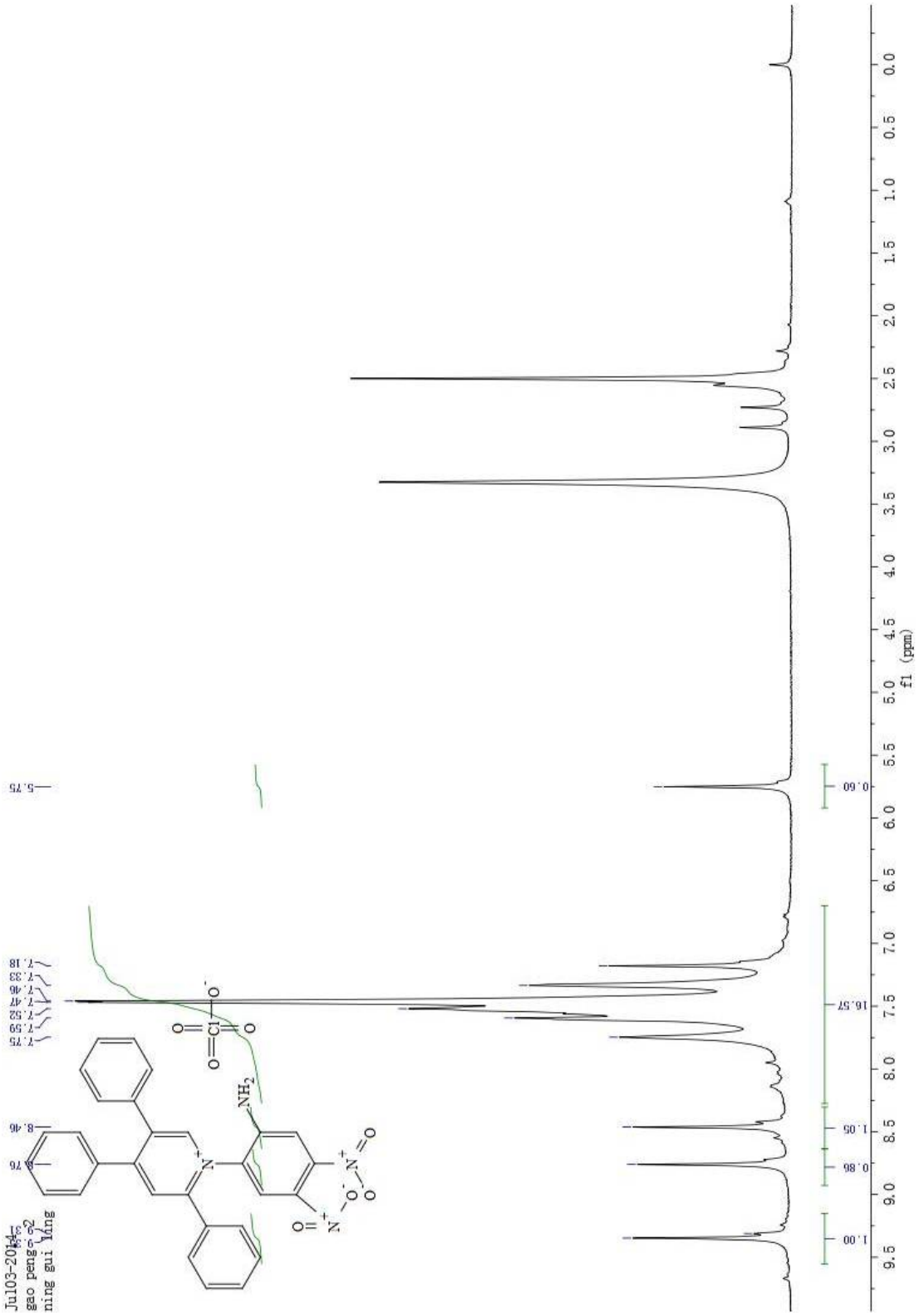
Compound 1e:



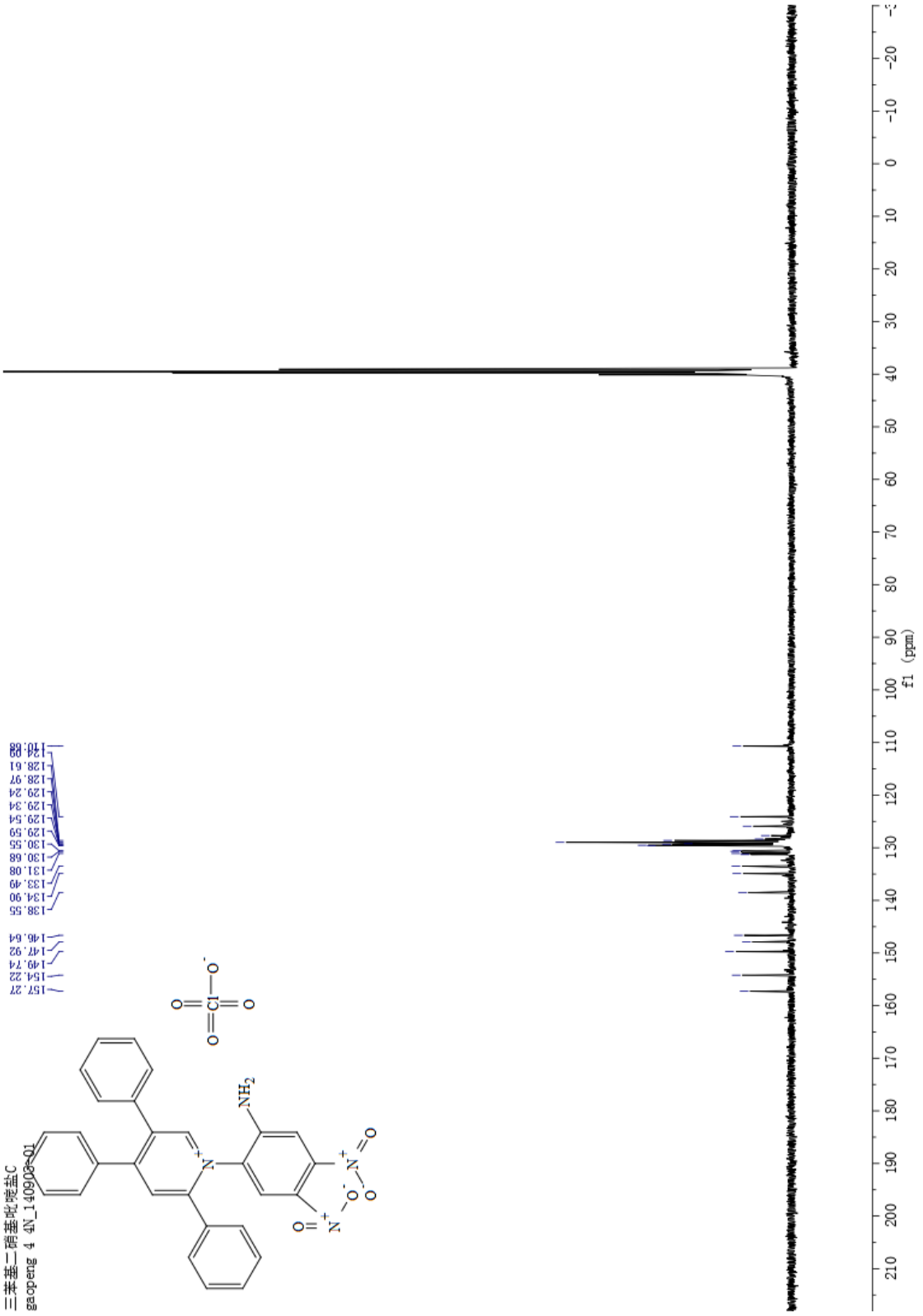
Compound 1e:


121214-40
li gang 5
in DMSO 13C
157.37
154.88
149.87
148.57
135.65
131.62
131.52
130.89
130.21
130.02
129.76
129.61
129.42
129.36
128.92
128.69
126.62
f1 (ppm)

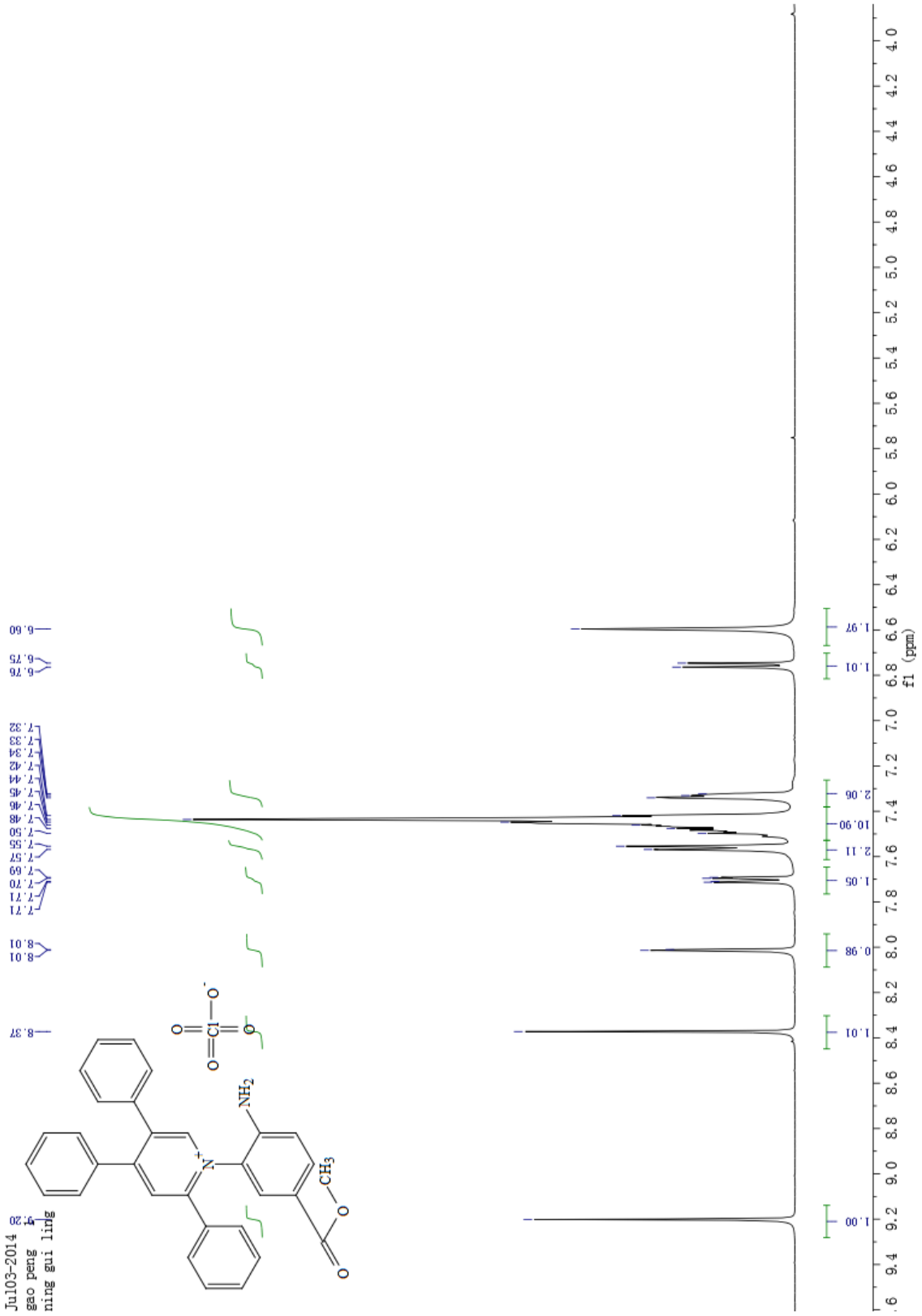
Compound 1m:



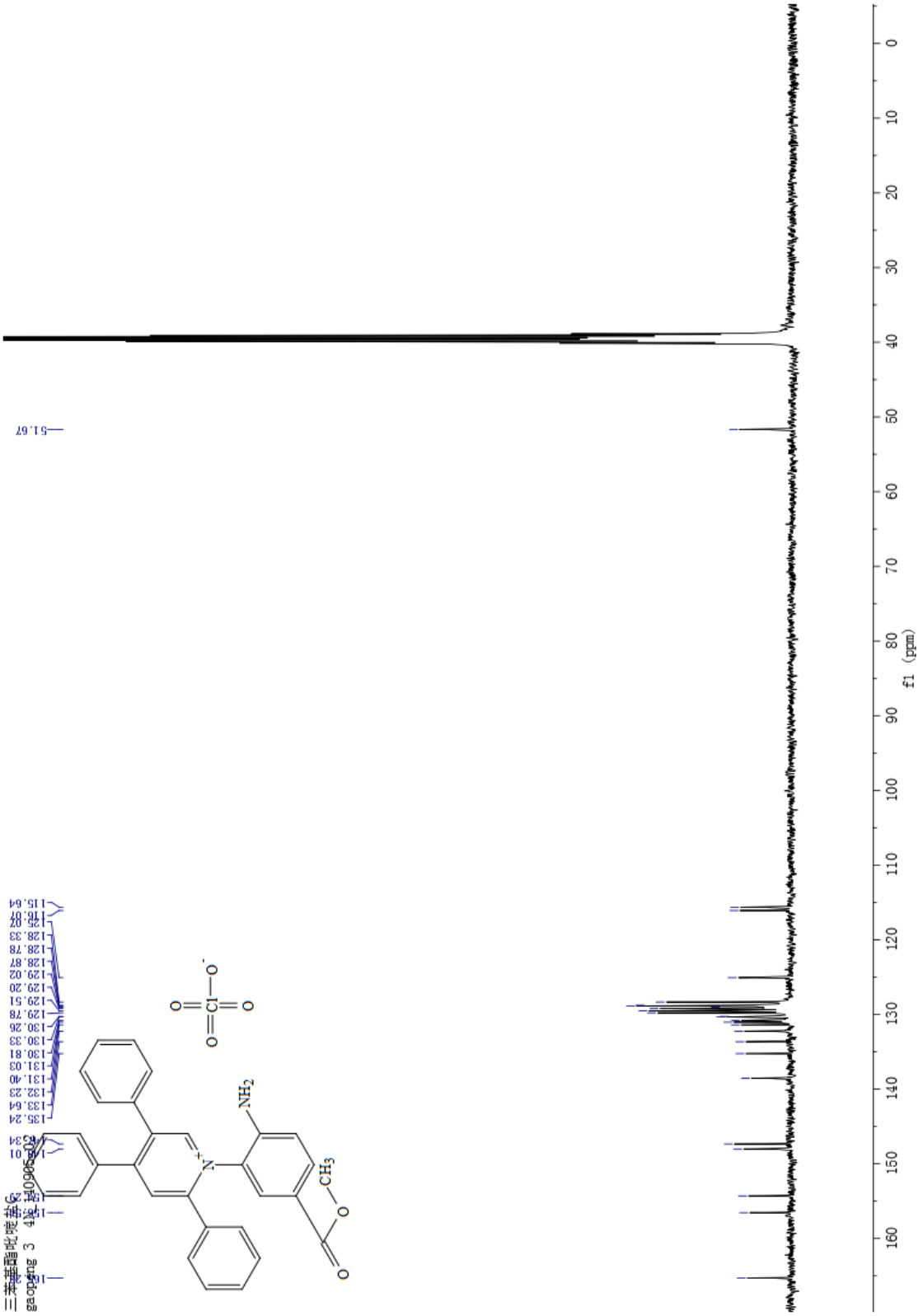
Compound 1m:


三苯基二硝基吡啶盐C
gaopeng 4 4N_140903-01
157.27
154.22
149.74
147.92
146.64
138.55
134.90
133.49
131.08
130.68
130.55
129.59
129.54
129.34
129.24
128.97
128.61
124.09
110.68
f1 (ppm)

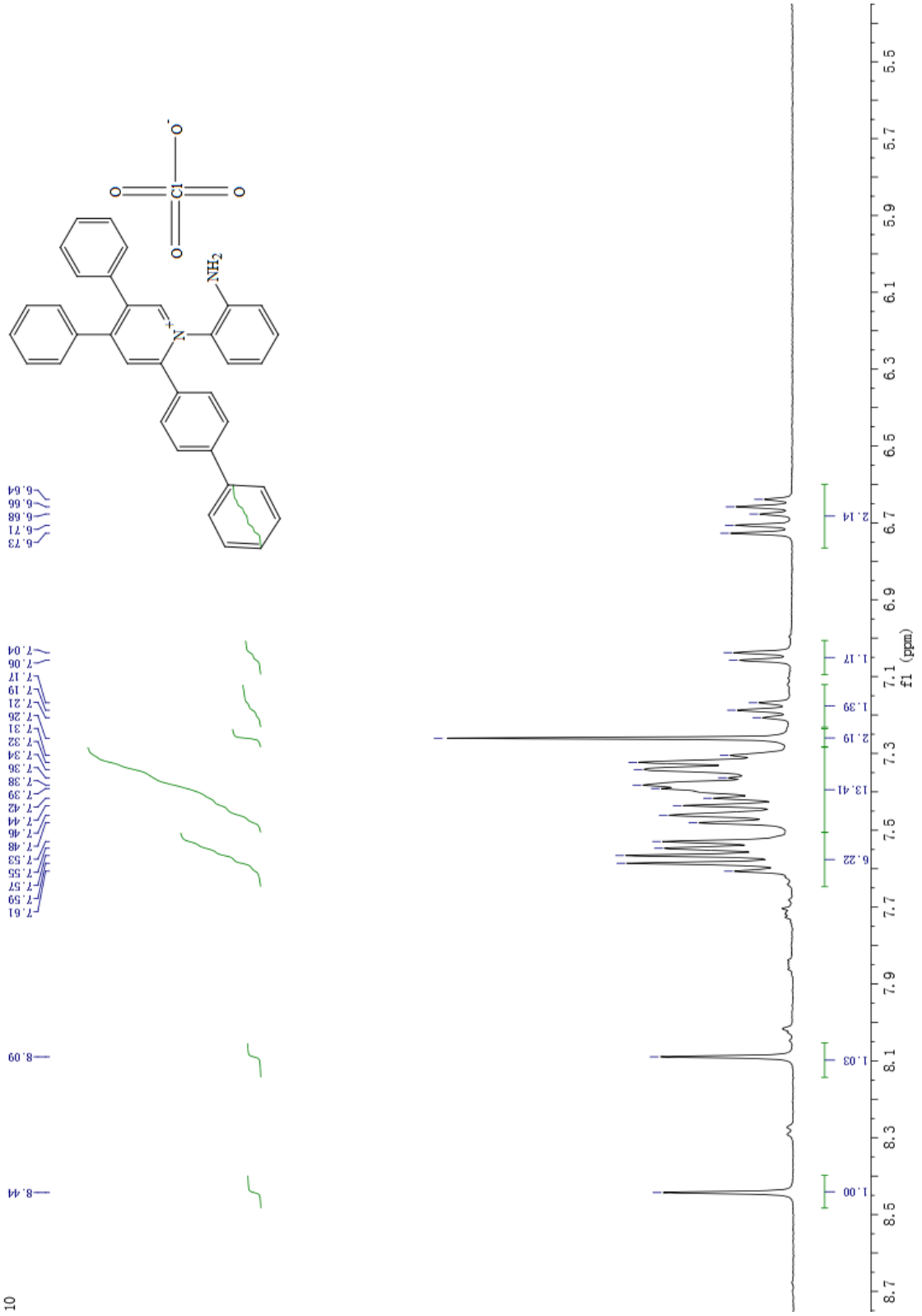
Compound 1k:



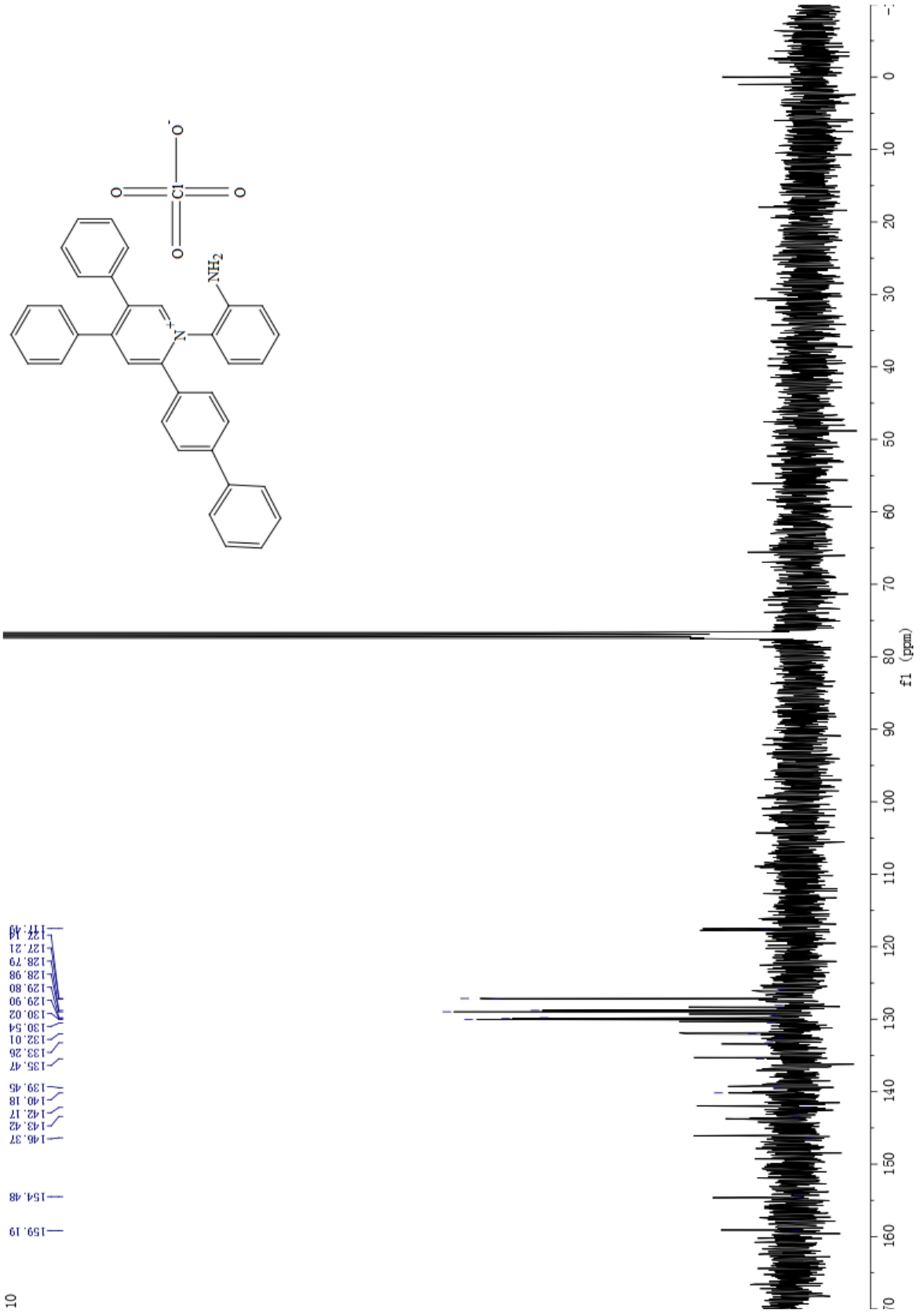
Compound 1k:



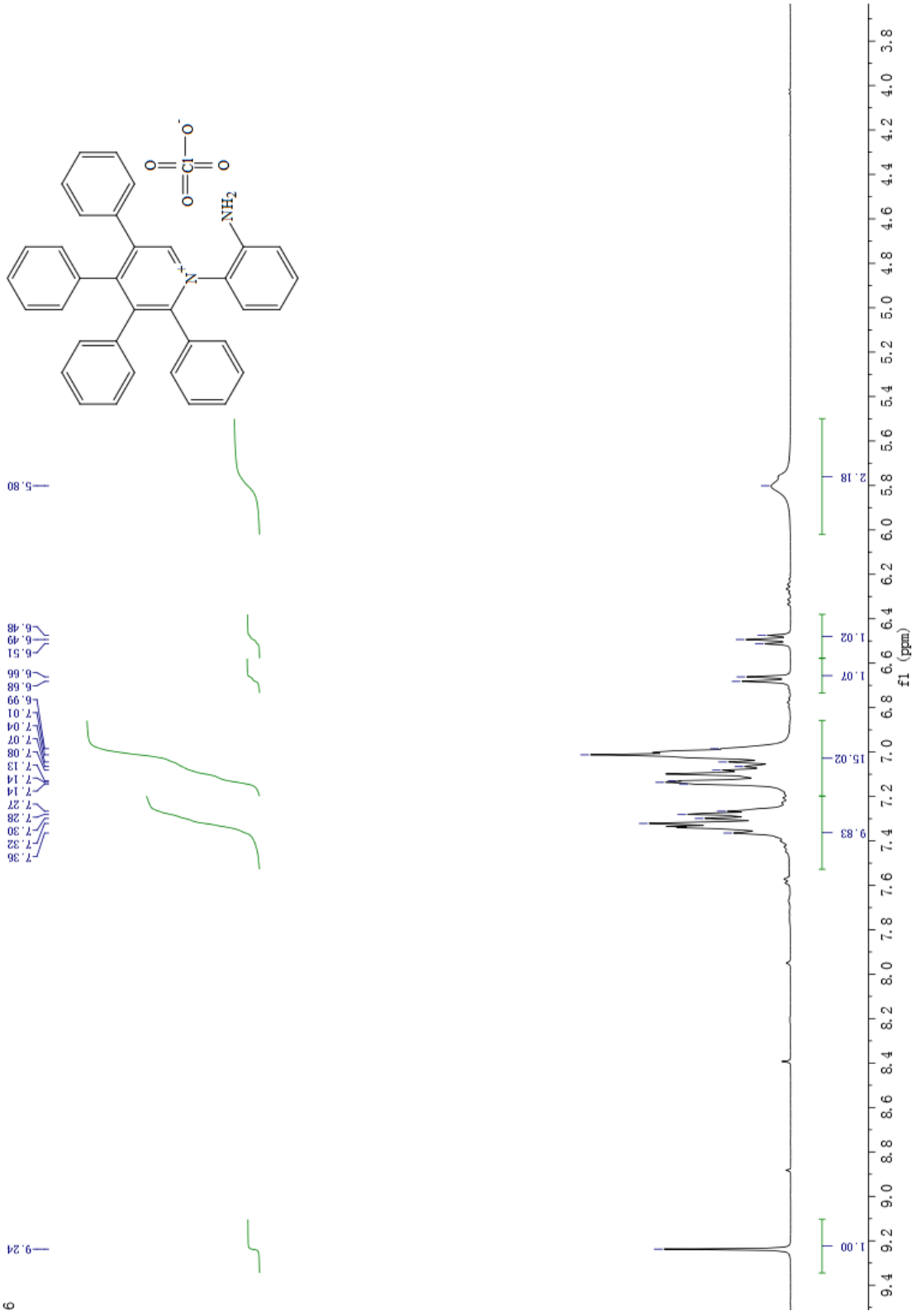
Compound 1f:



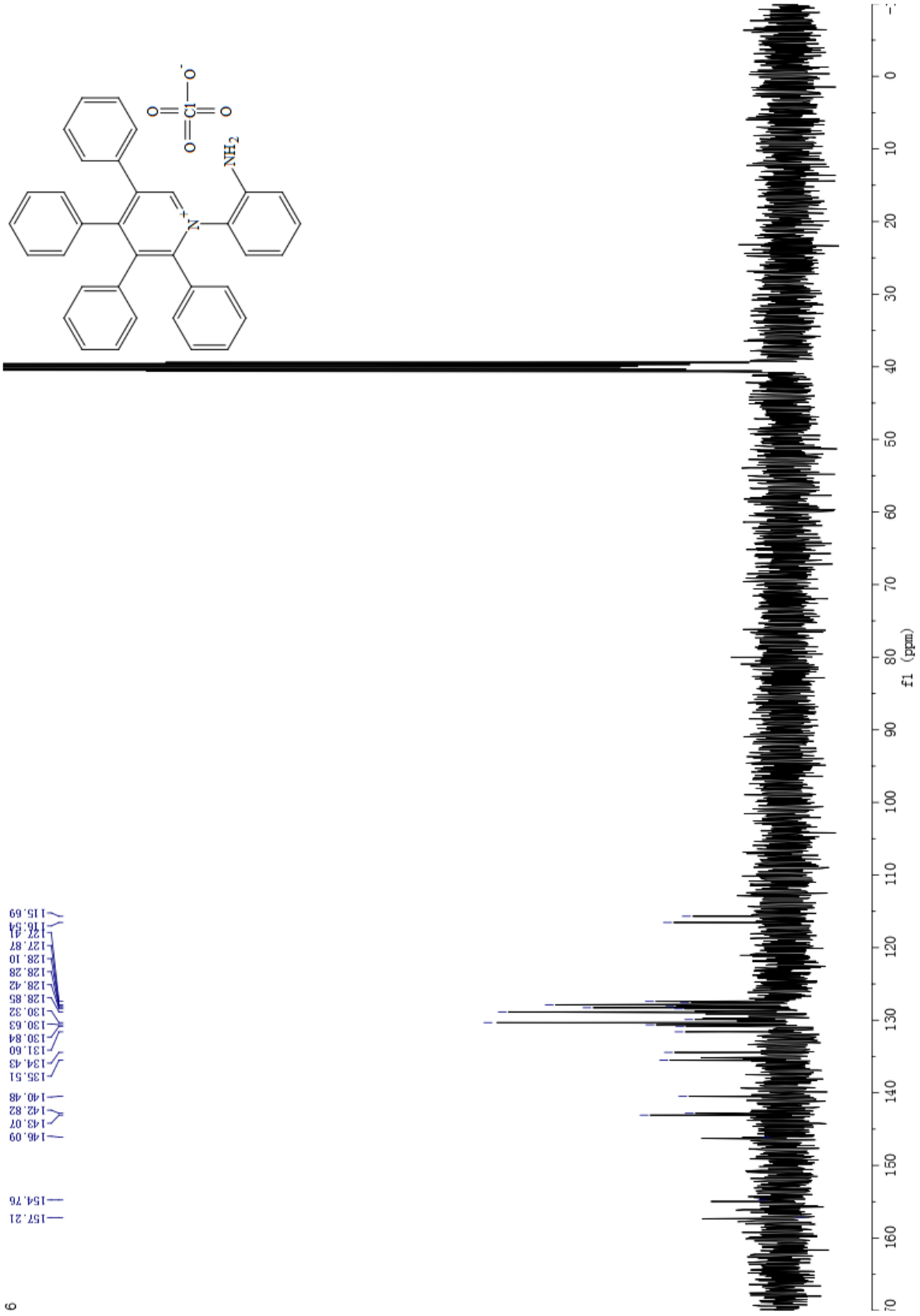
Compound 1f:



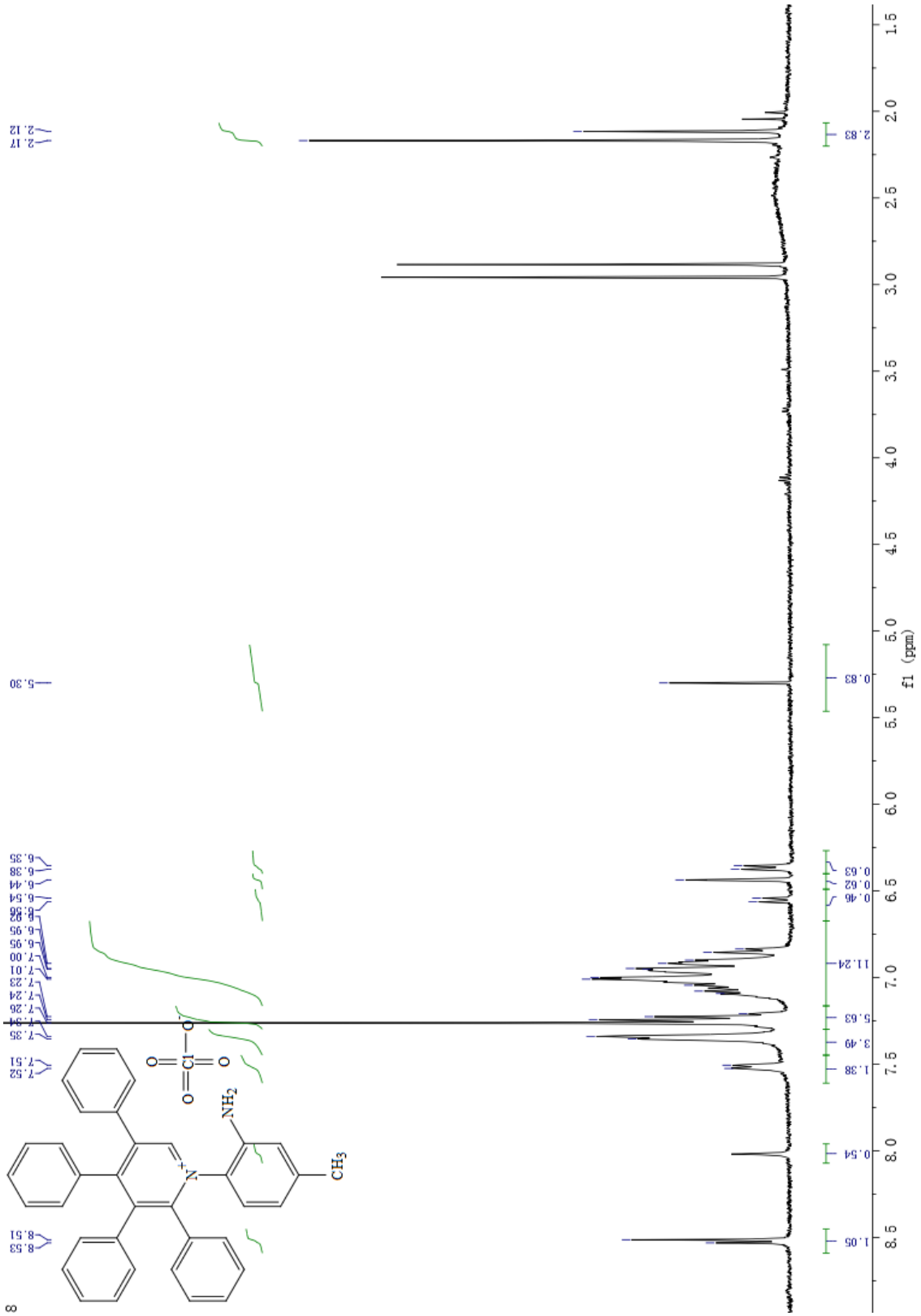
Compound 1g:



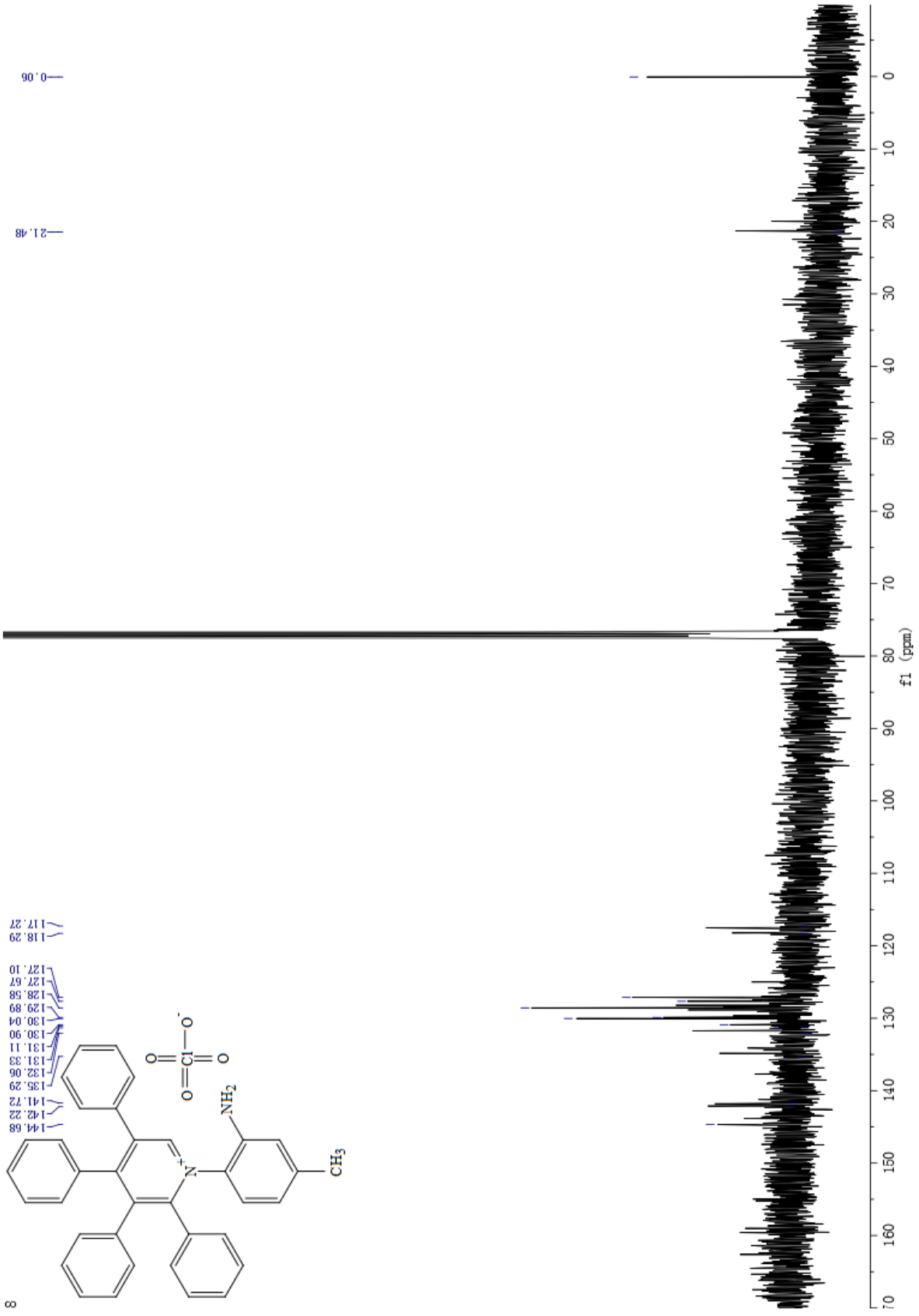
Compound 1g:



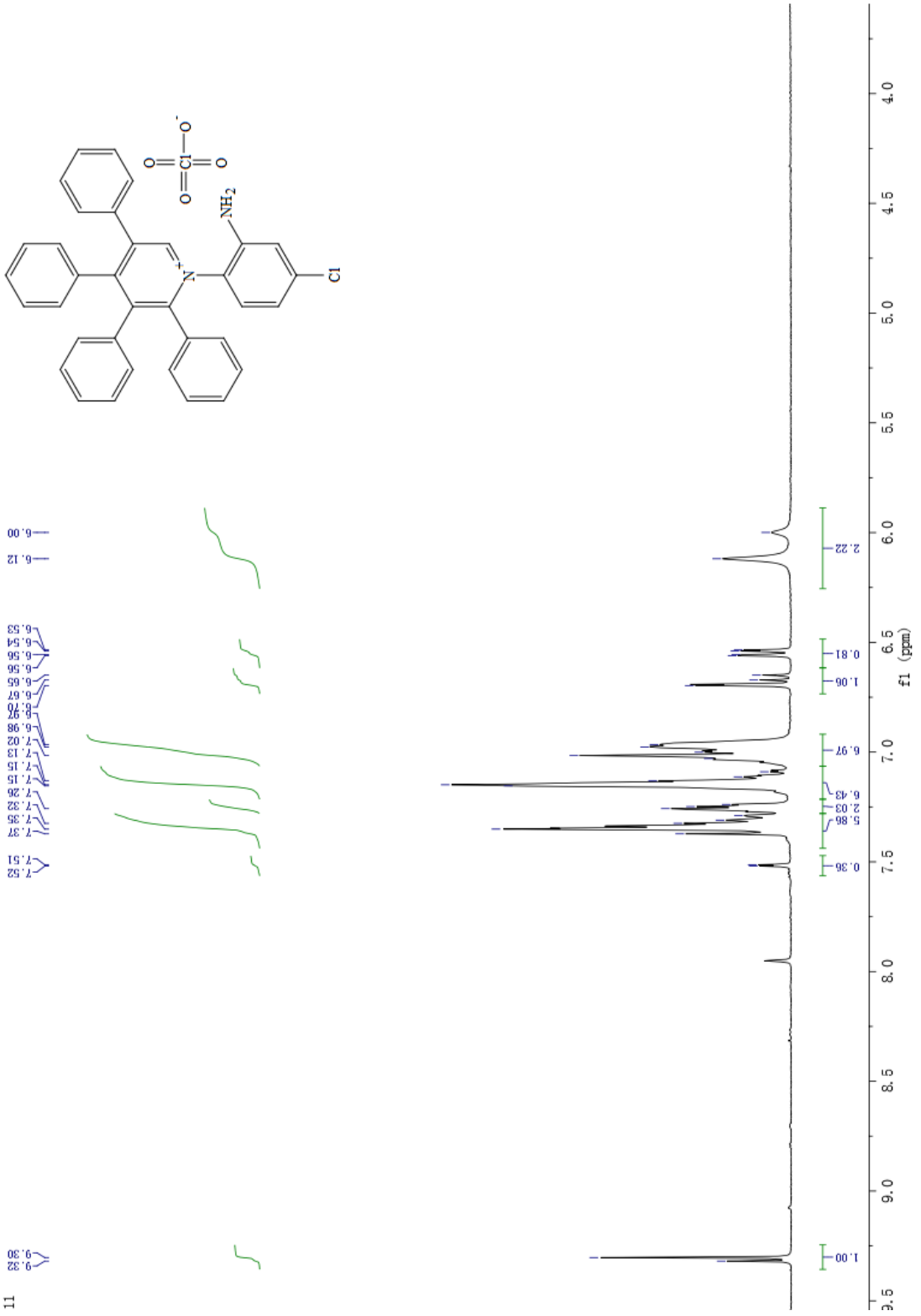
Compound 1h:



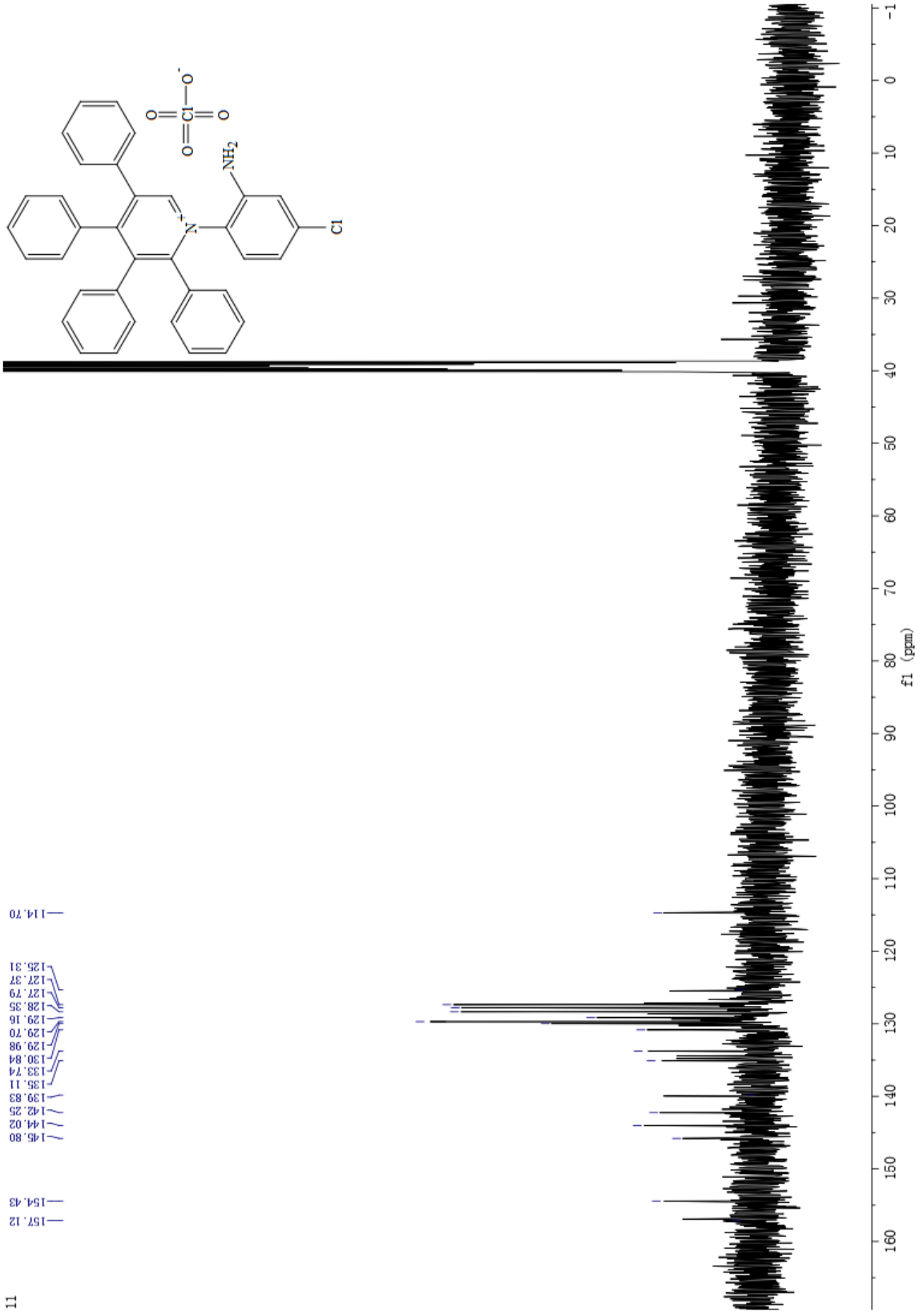
Compound 1h:


0.06
21.48
117.27
118.29
127.10
127.67
128.58
129.89
130.04
130.90
131.11
131.33
132.06
135.29
141.72
142.22
144.68
NH2
CH3
f1 (ppm)

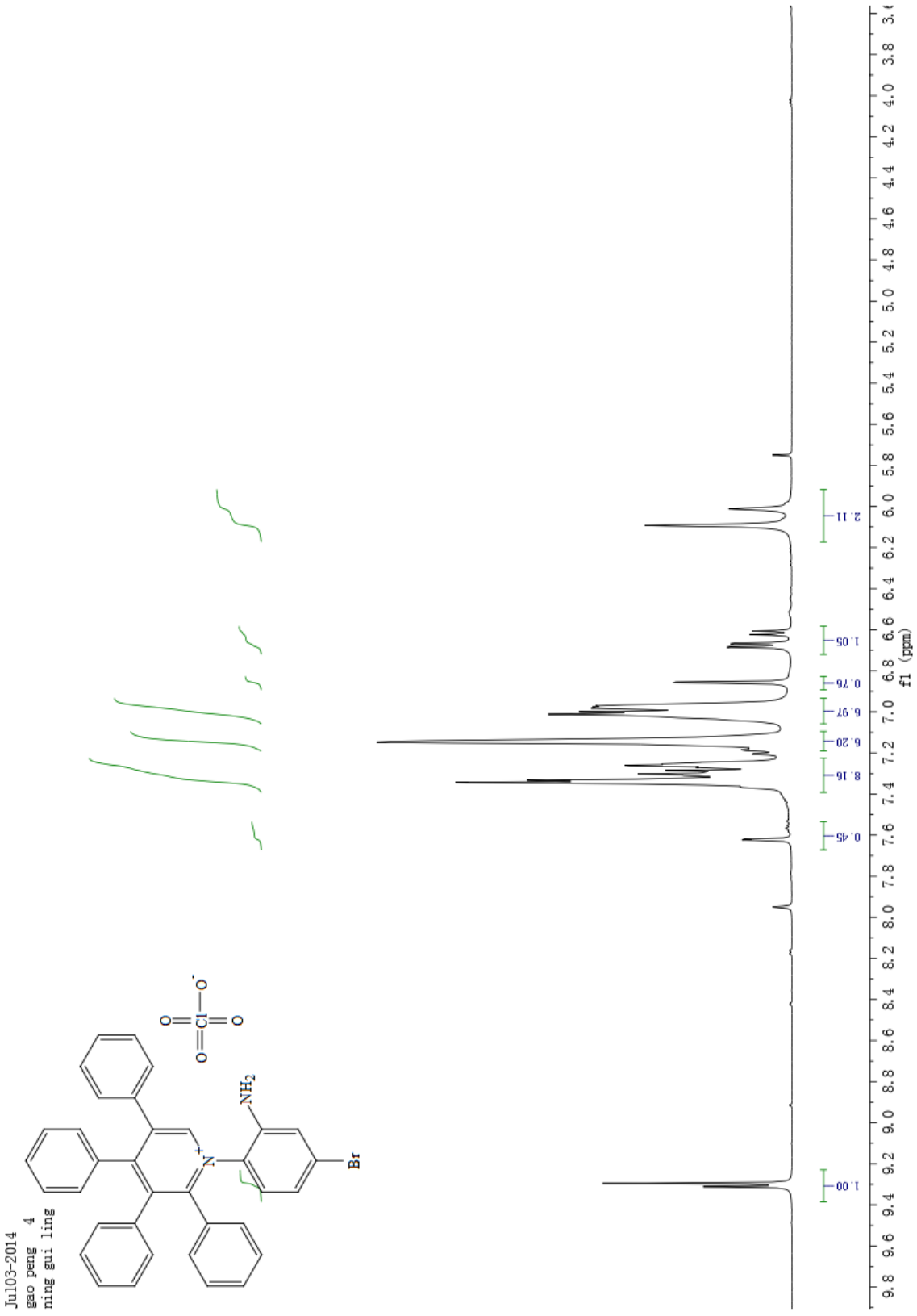
Compound 1i:



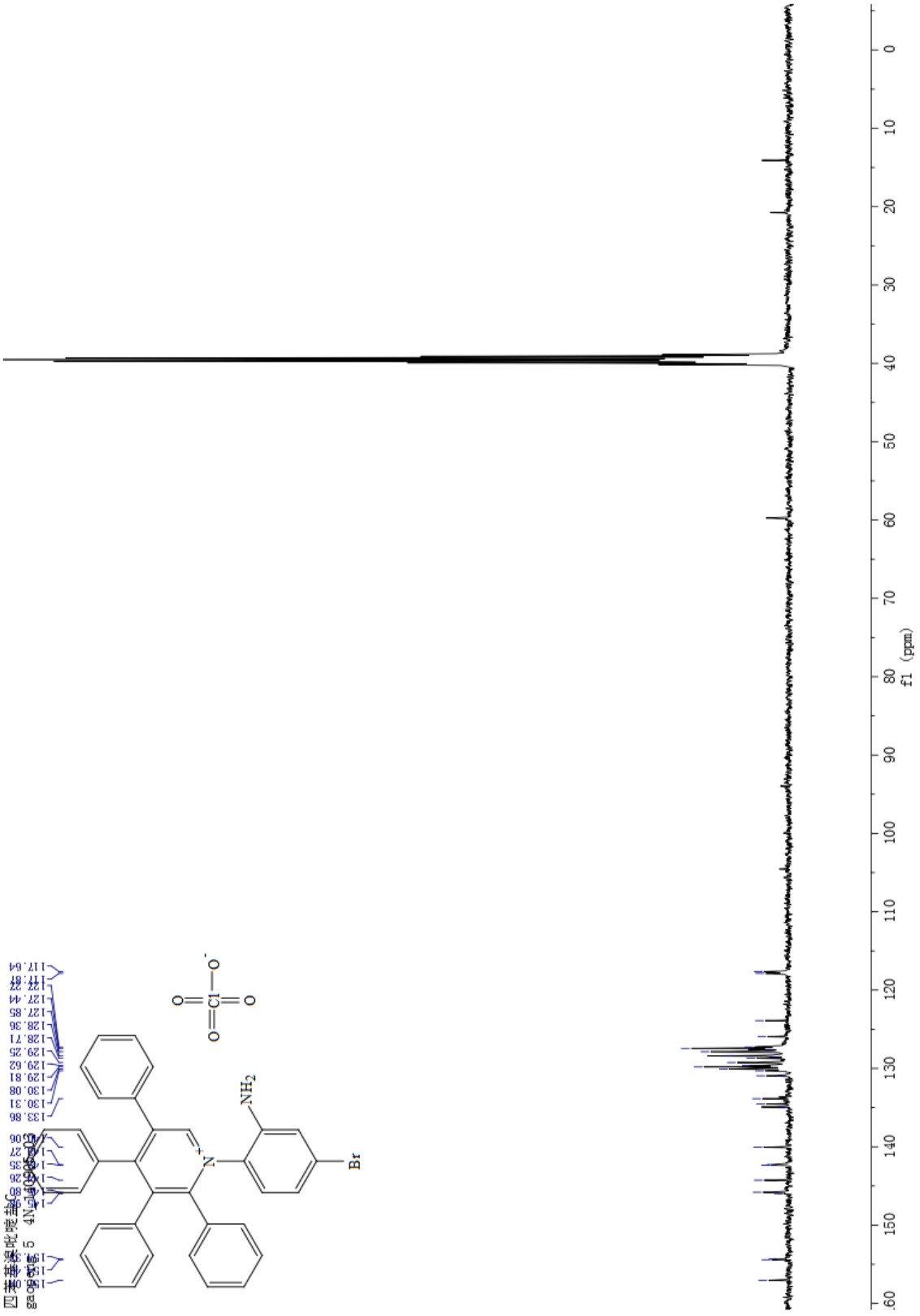
Compound 1i:


NH2
Cl
Cl
O
O
O
O
N+
11
114.70
125.31
127.37
127.79
128.35
129.16
129.70
129.98
130.84
133.74
135.11
139.83
142.25
144.02
145.80
154.43
157.12
f1 (ppm)

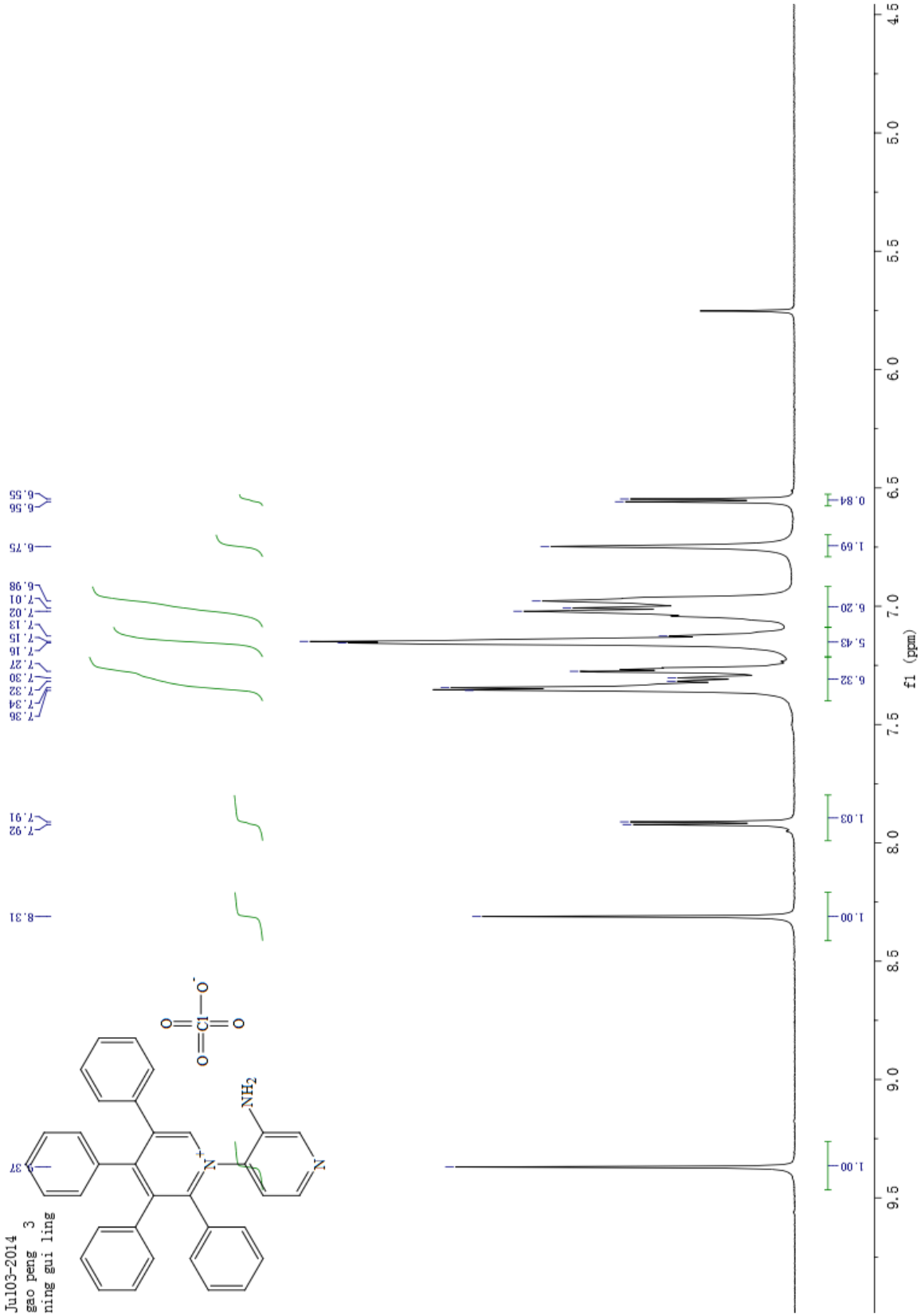
Compound 1l:


Ju103-2014
gao peng 4
ning gui ling
NH2
Br
f1 (ppm)
1.00
0.45
8.16
6.20
6.97
0.76
1.05
2.11

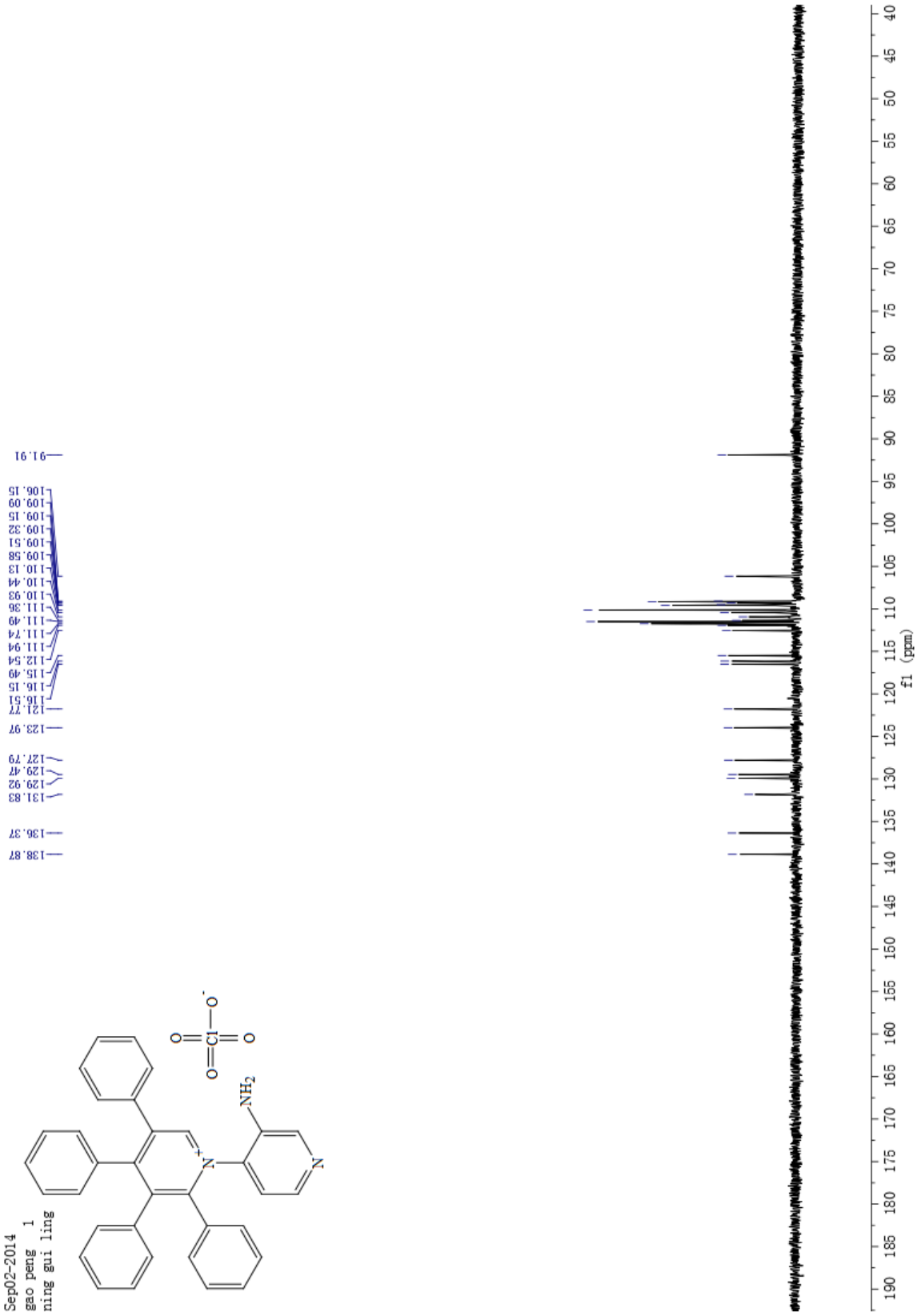
Compound 1l:



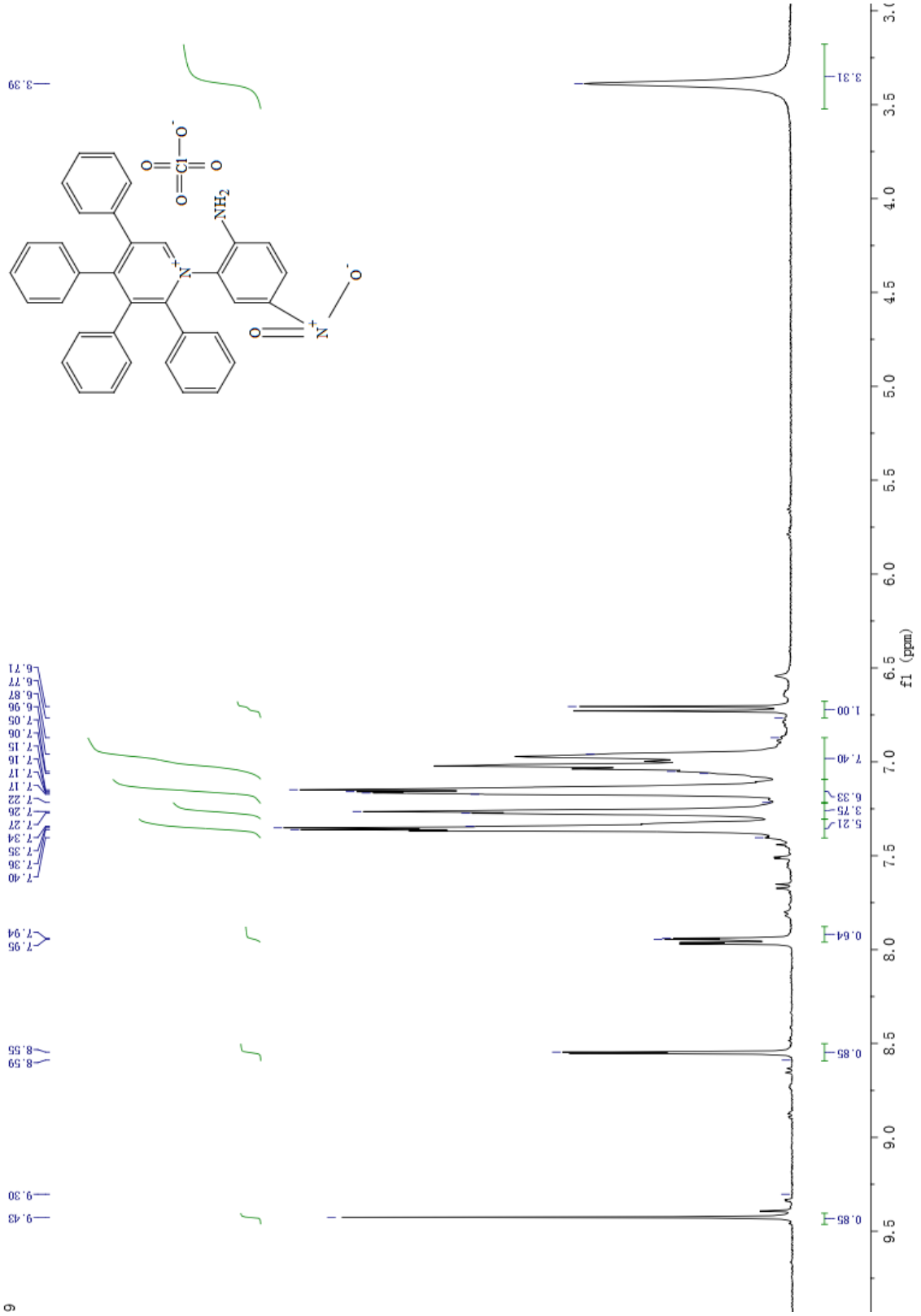
Compound 1p:



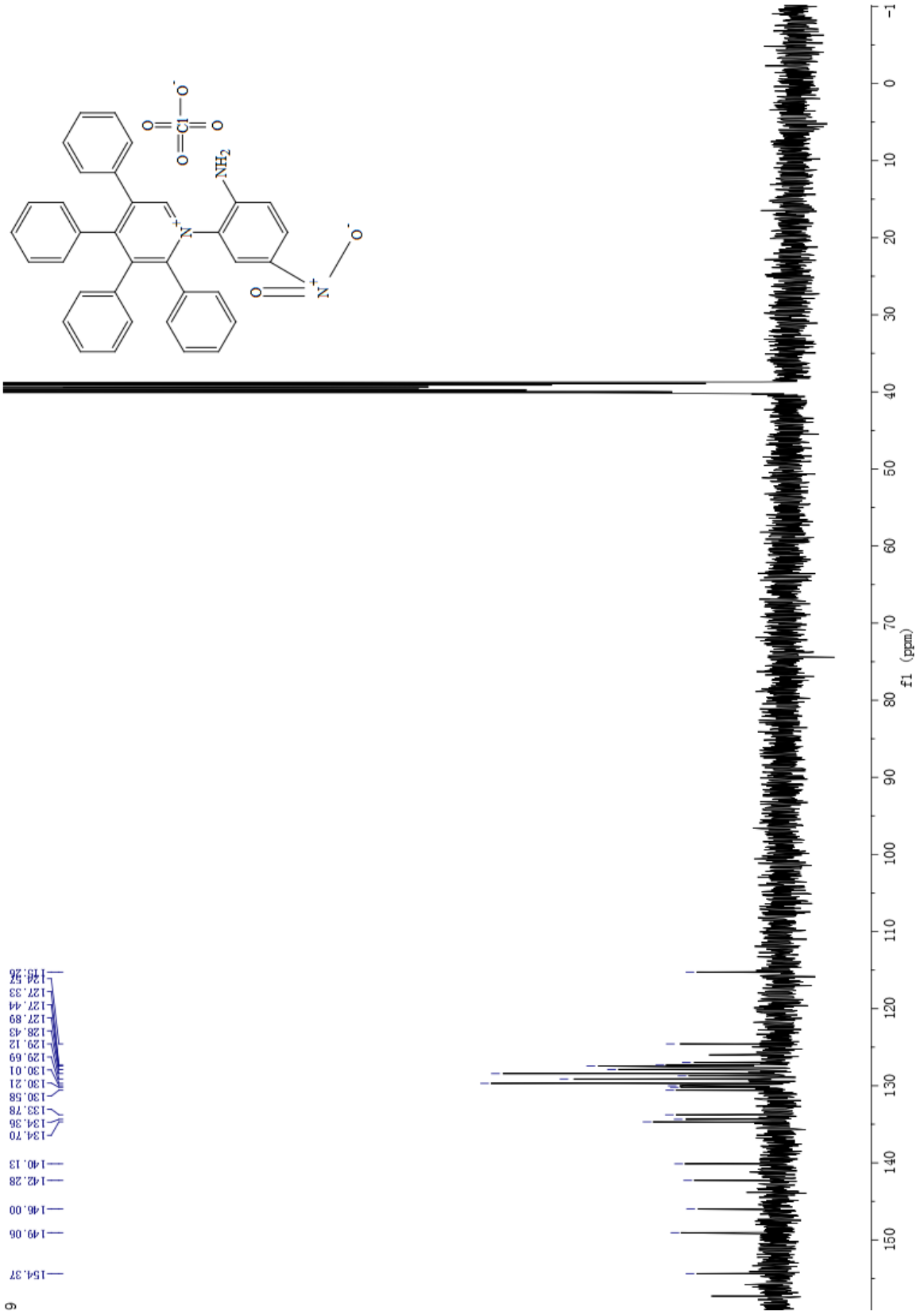
Compound 1p:



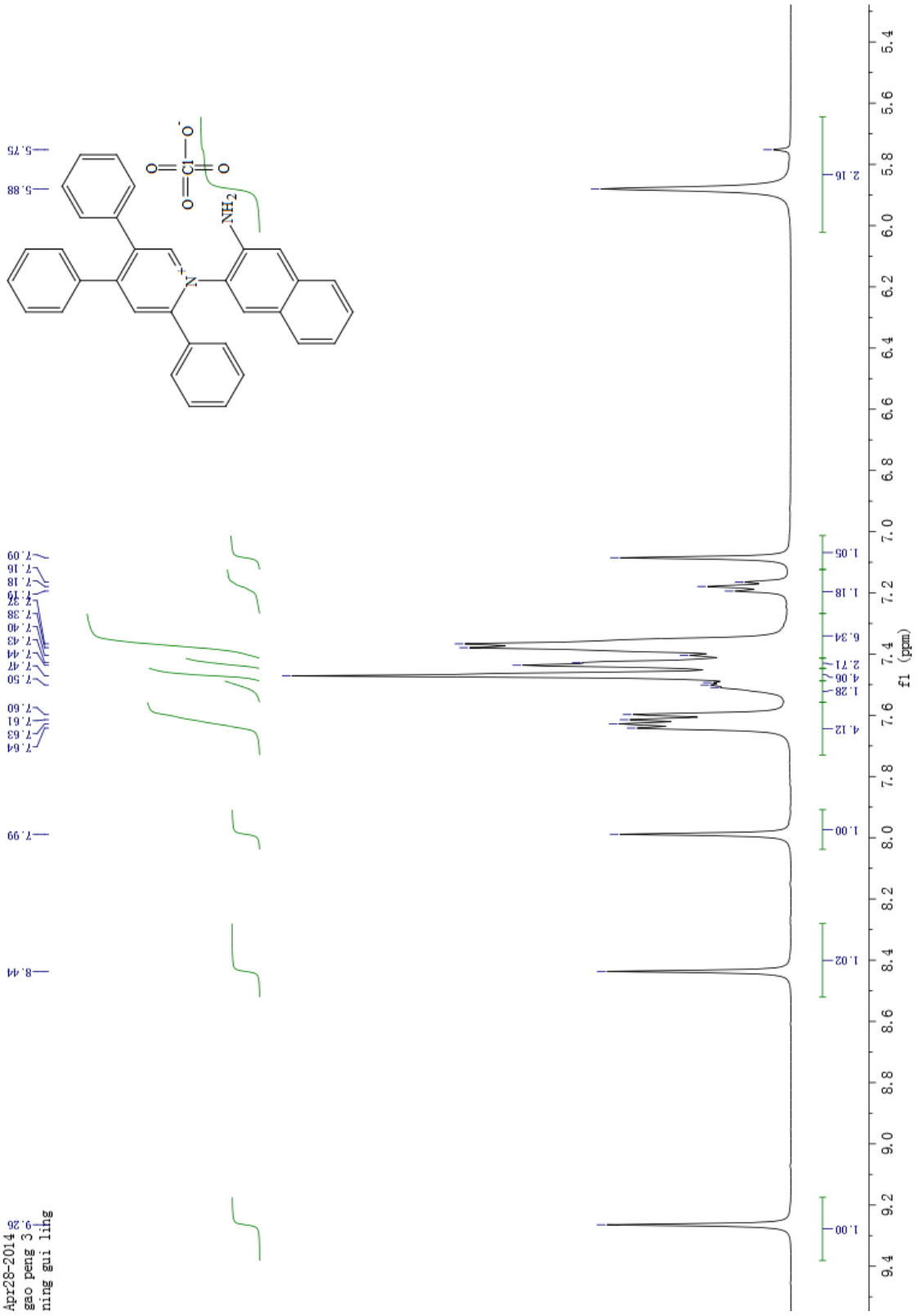
Compound 1j:



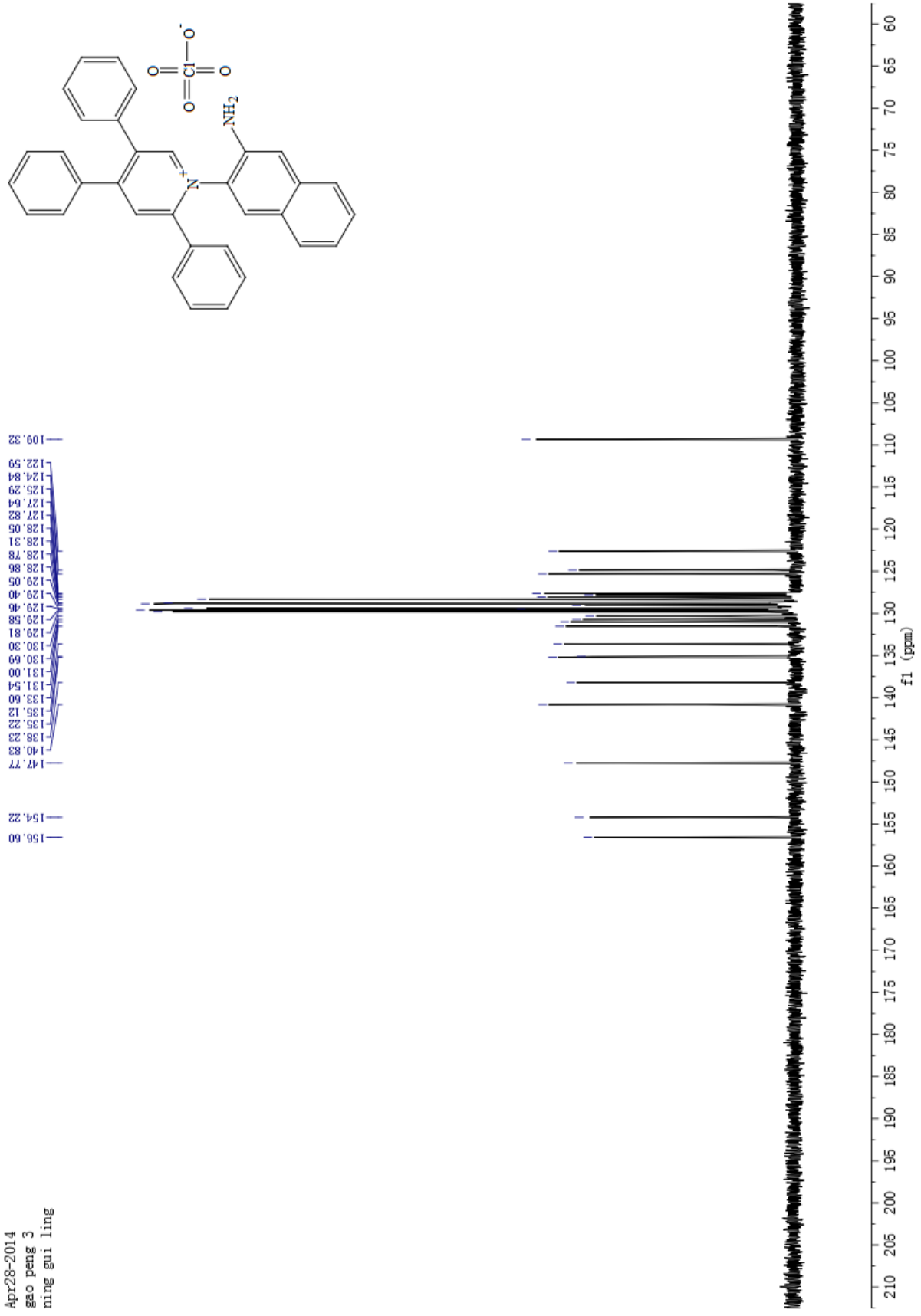
Compound 1j:



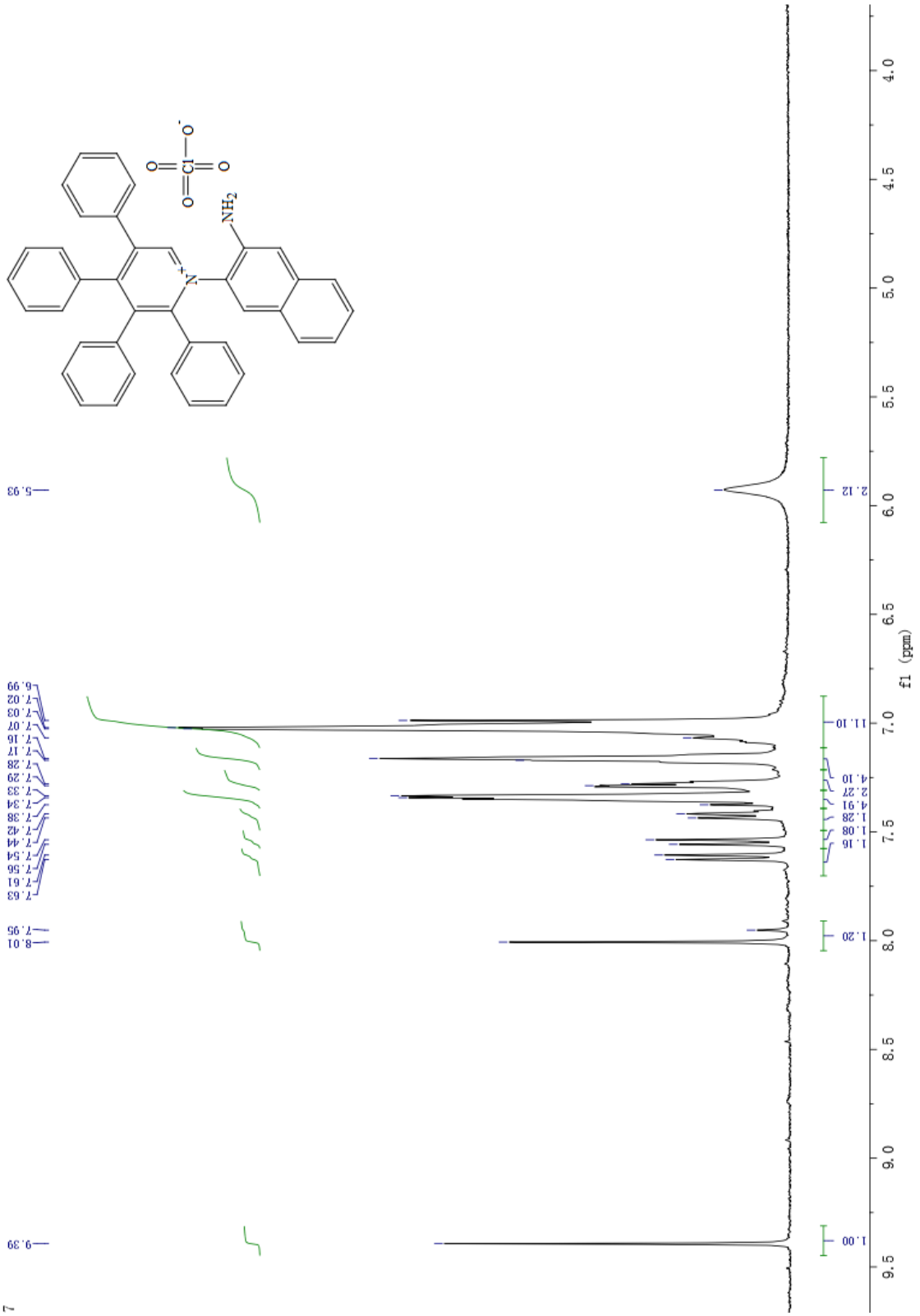
Compound 1n:



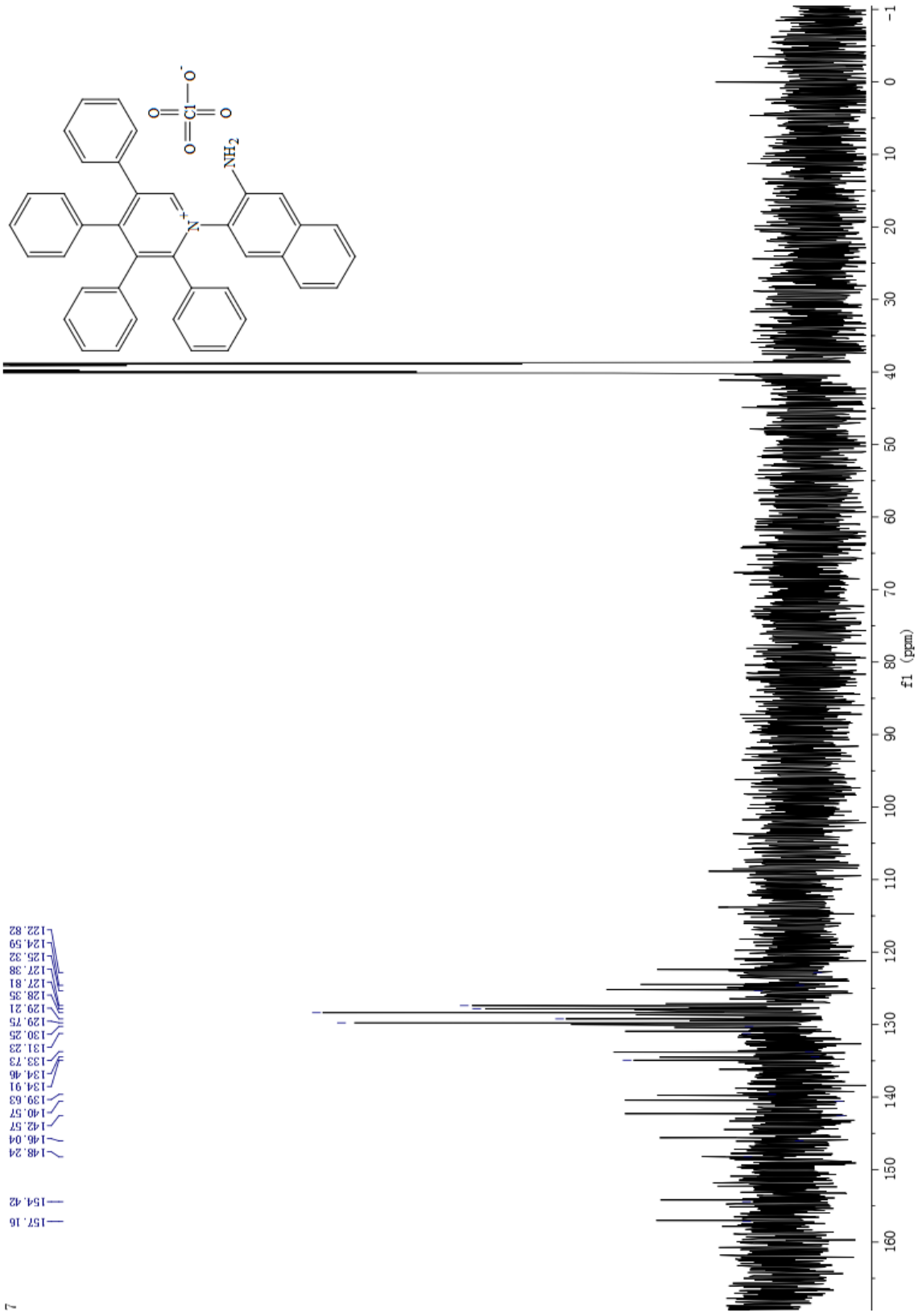
Compound 1n:



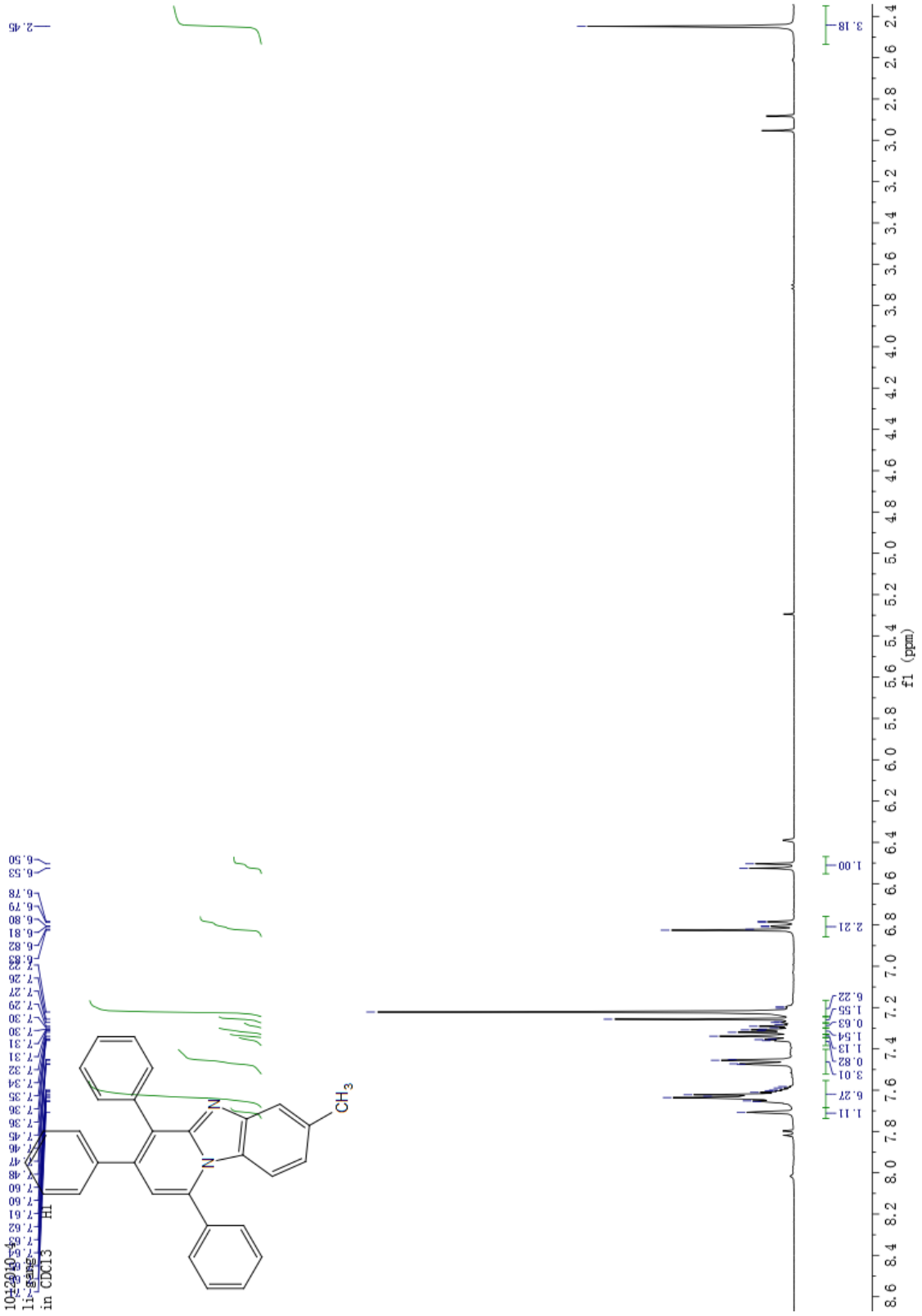
Compound 1o:



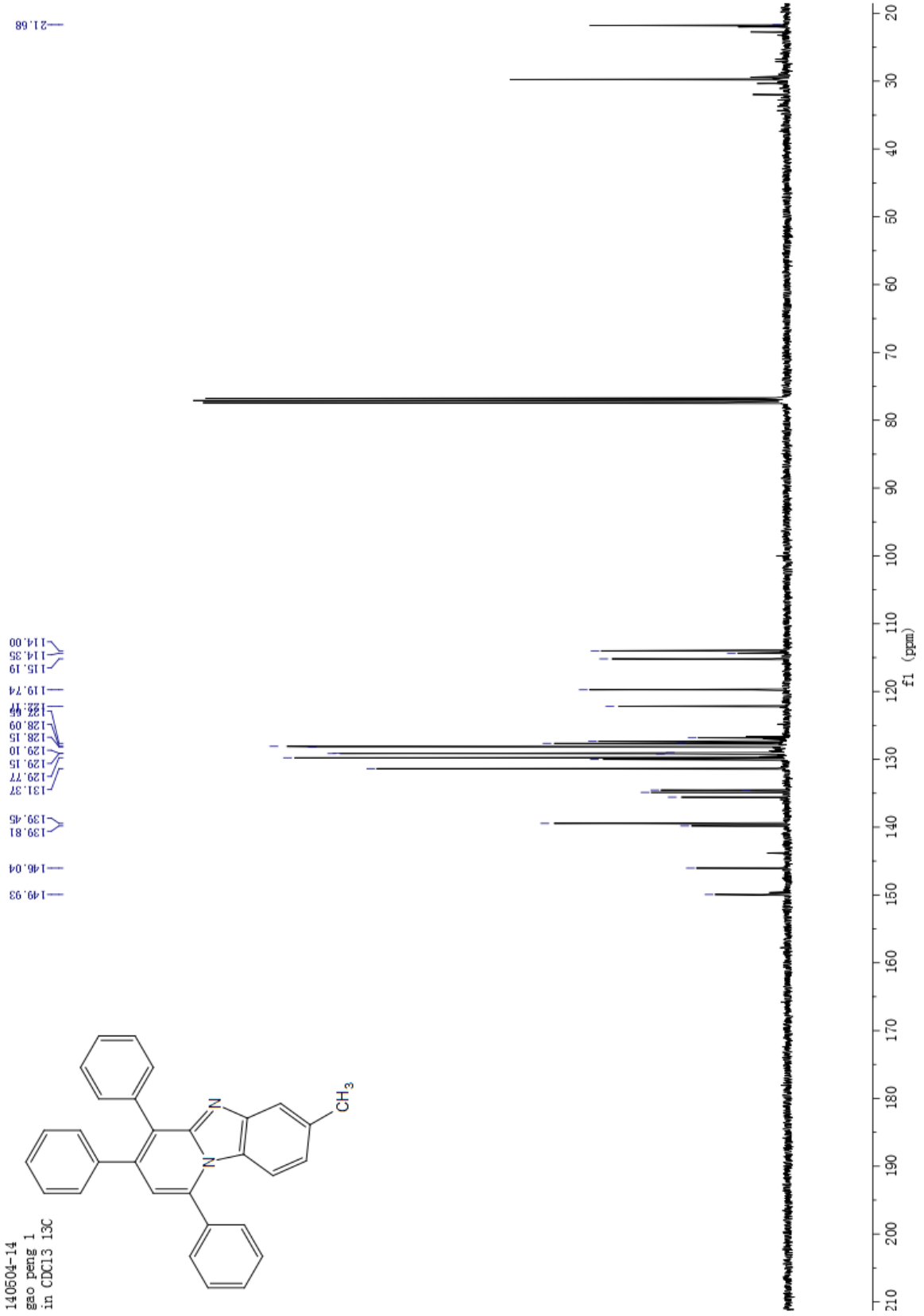
Compound 1o:


NH2
N+
Cl
O
O
O
O-
157.16
154.42
148.24
146.04
142.57
140.57
139.63
134.91
134.46
133.73
131.25
130.25
129.75
129.21
128.35
127.81
127.38
125.32
124.59
122.82
f1 (ppm)

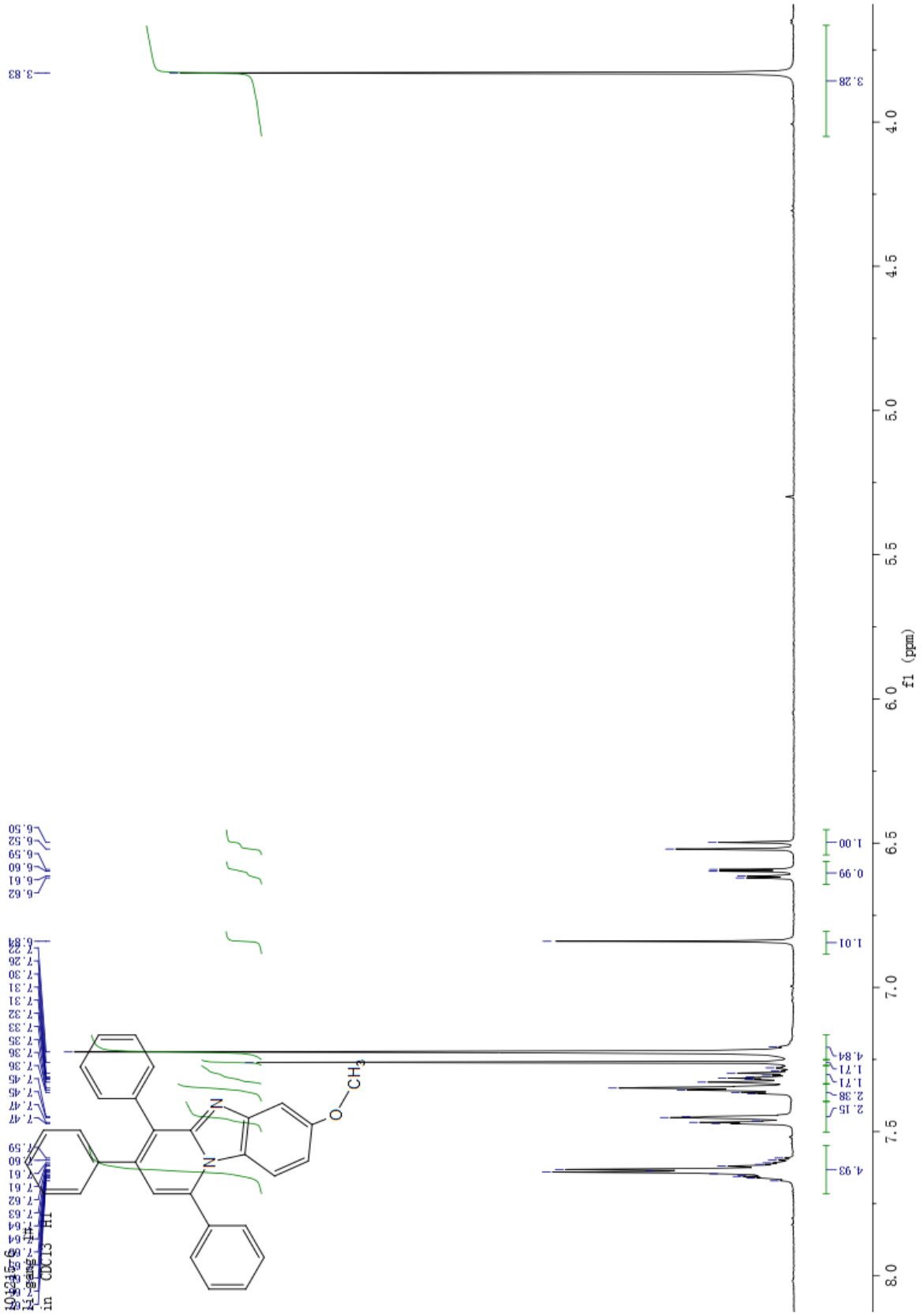
Compound 2b:



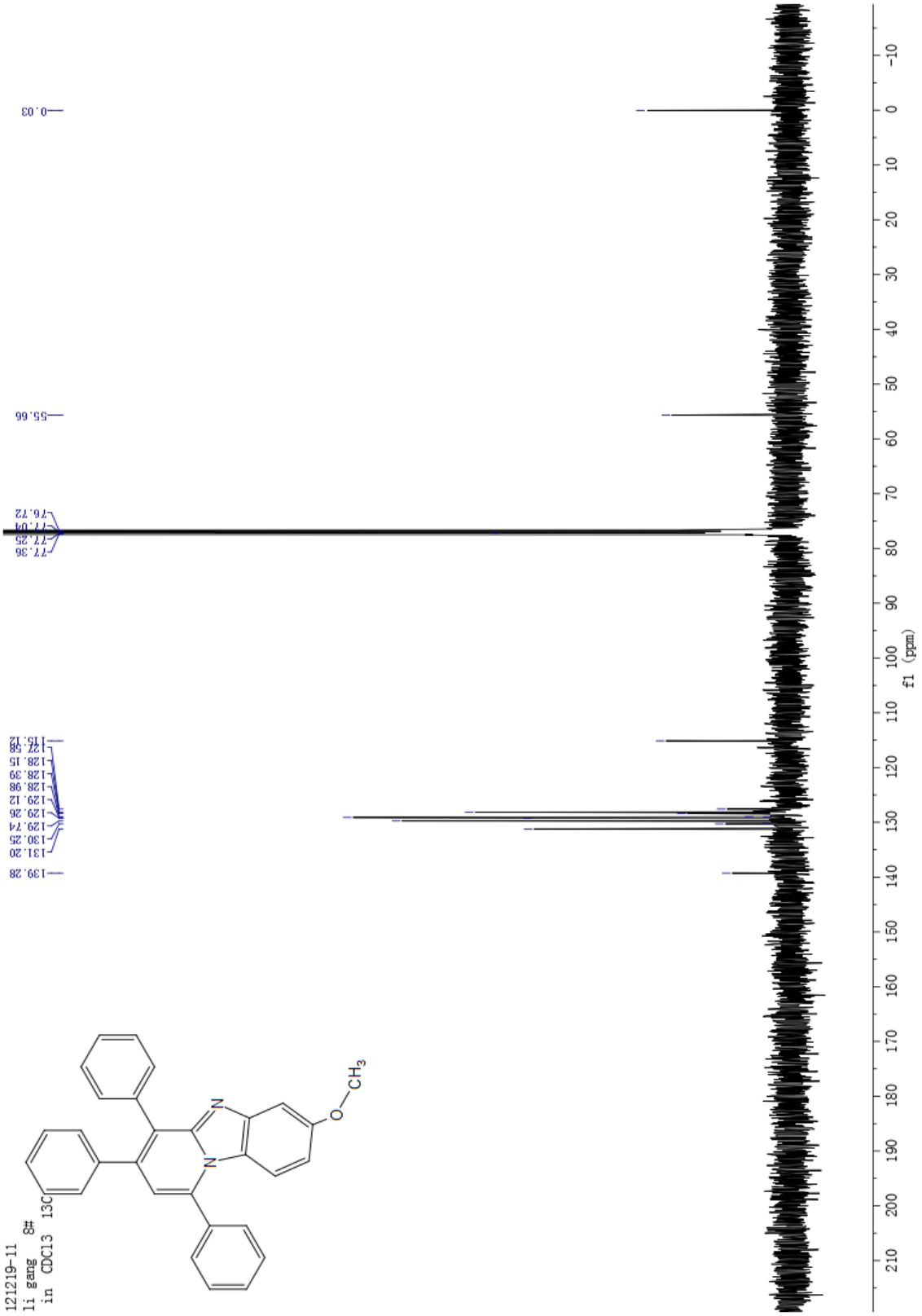
Compound 2b:


21.68
114.00
114.35
115.19
119.74
122.46
122.47
128.09
128.15
129.10
129.15
129.77
131.37
139.45
139.81
146.04
149.93
f1 (ppm)
140504-14
gao peng 1
in CDCl3 13C
CH3
N
N

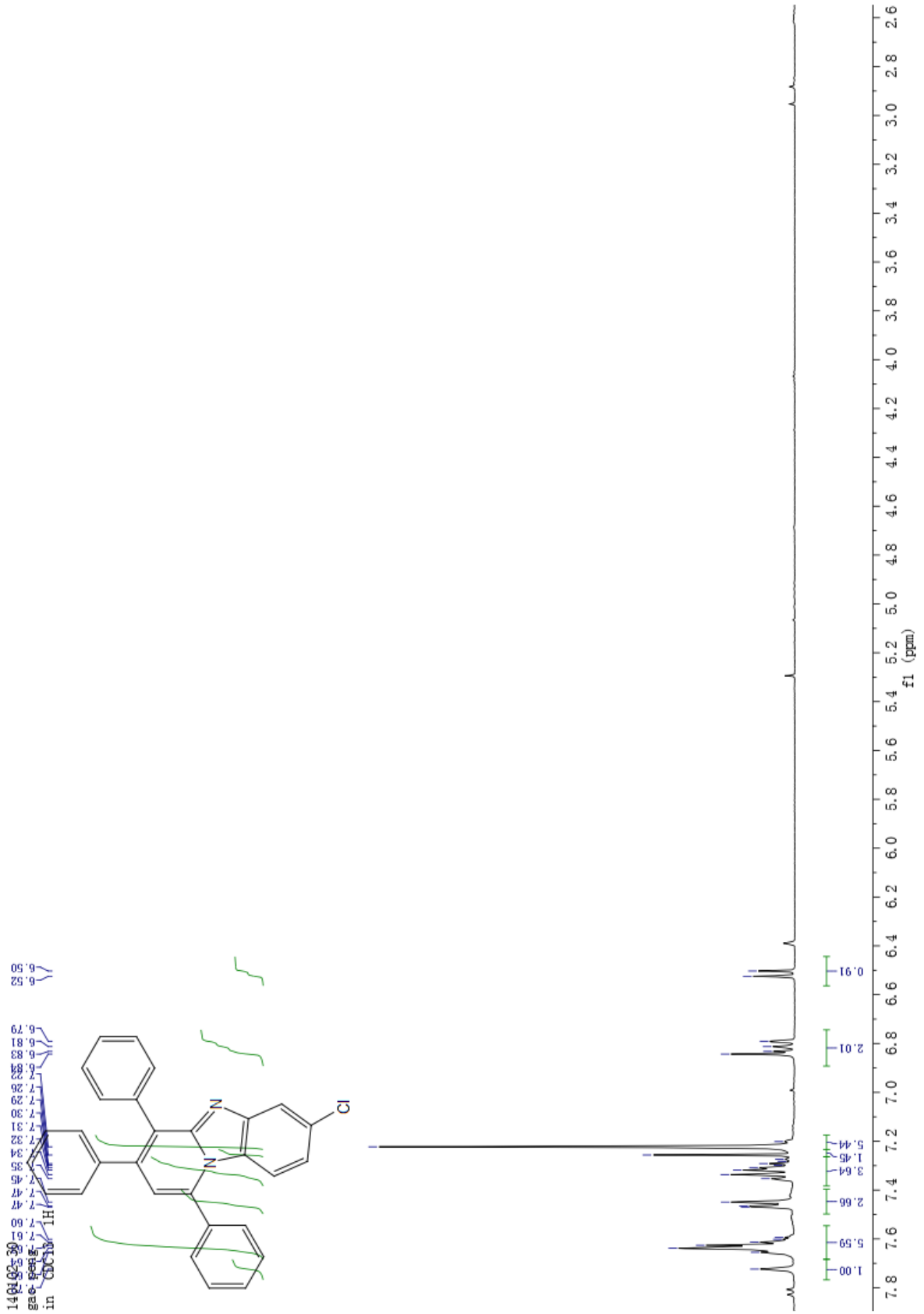
Compound 2c:



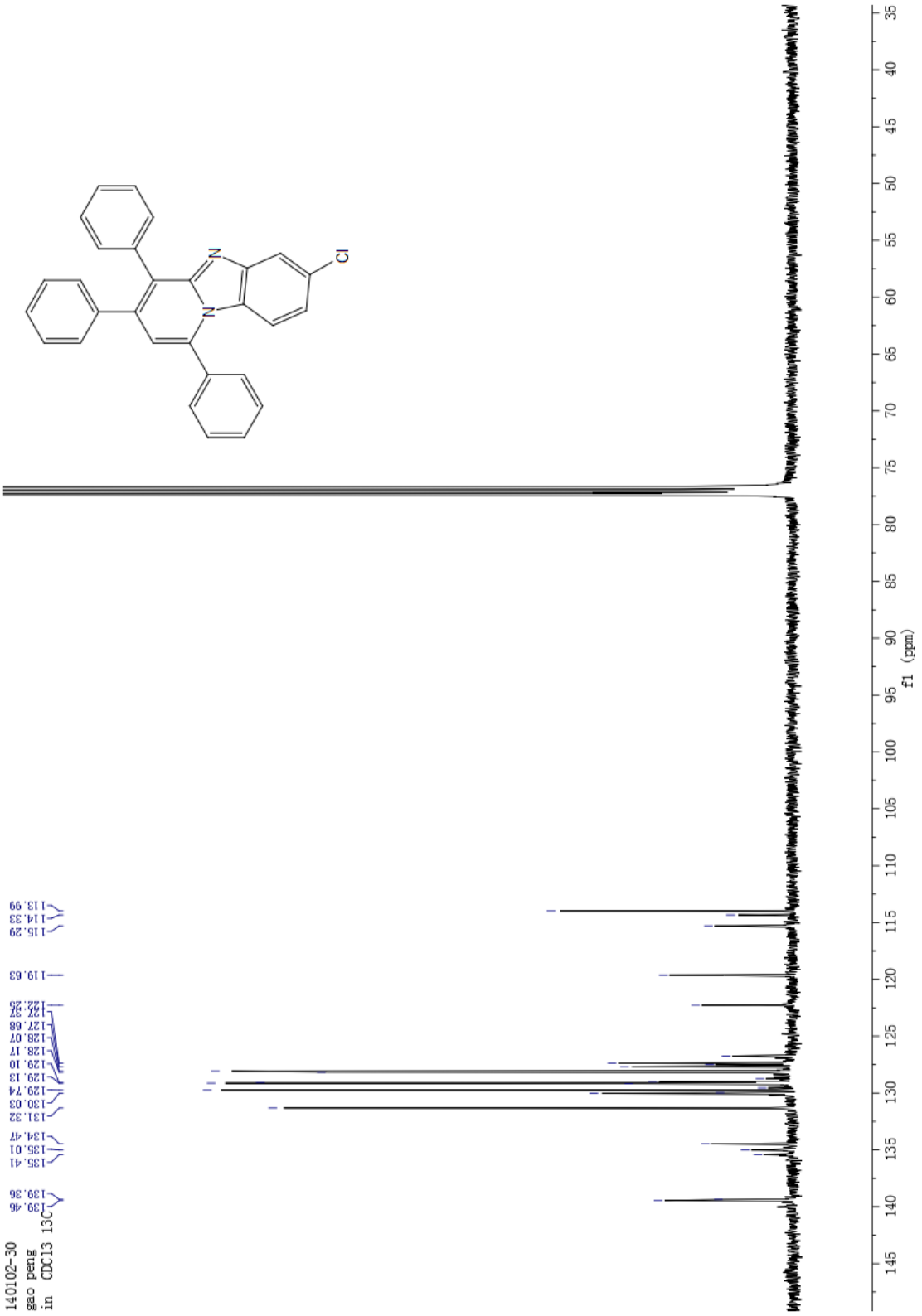
Compound 2c:


121219-11
li gang 8#
in CDCl3 13C
O
N
N
CH3
139.28
131.20
130.25
129.74
129.26
129.12
128.98
128.39
128.15
127.98
115.12
77.36
77.25
77.04
76.72
55.66
0.03
f1 (ppm)

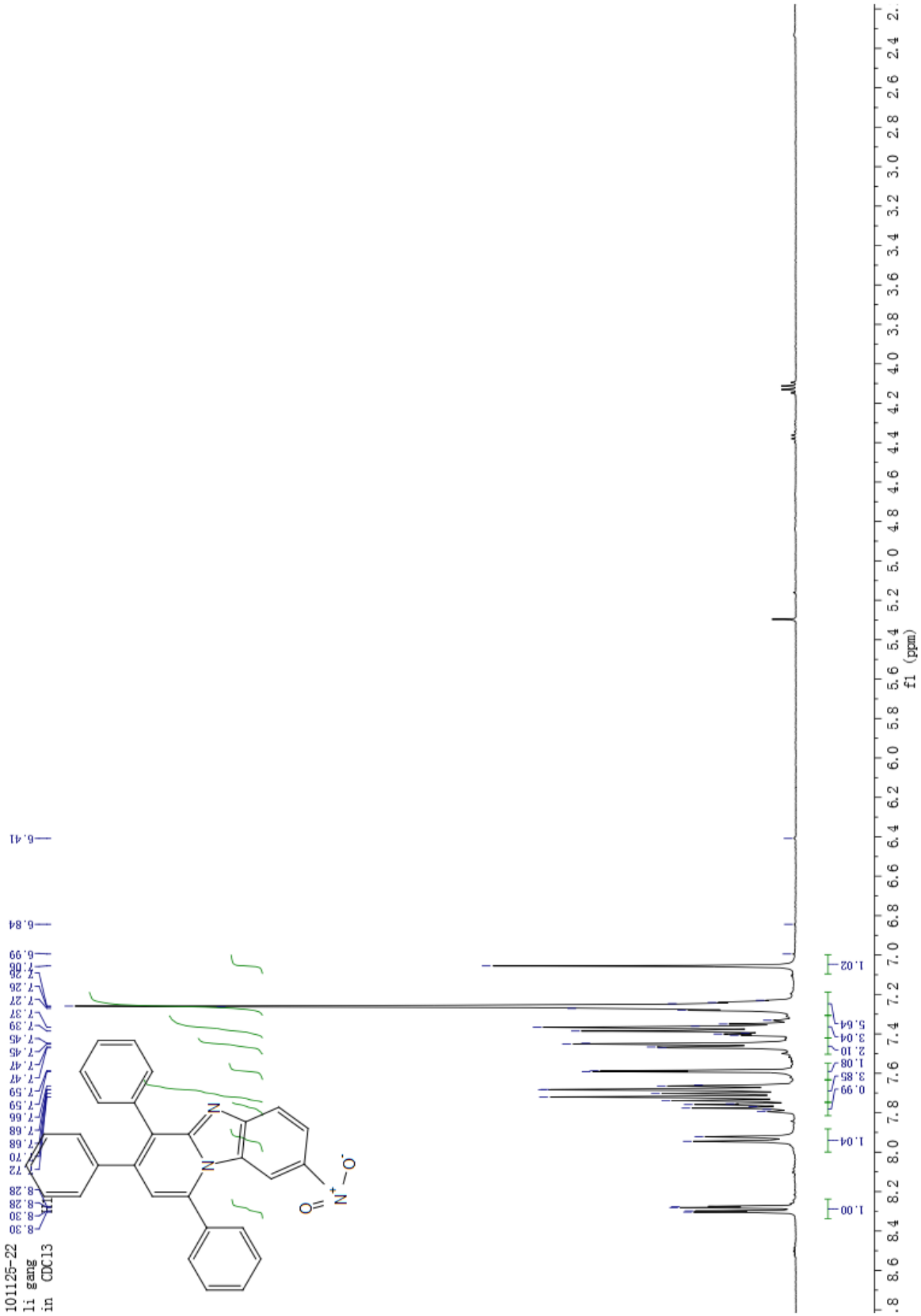
Compound 2d:



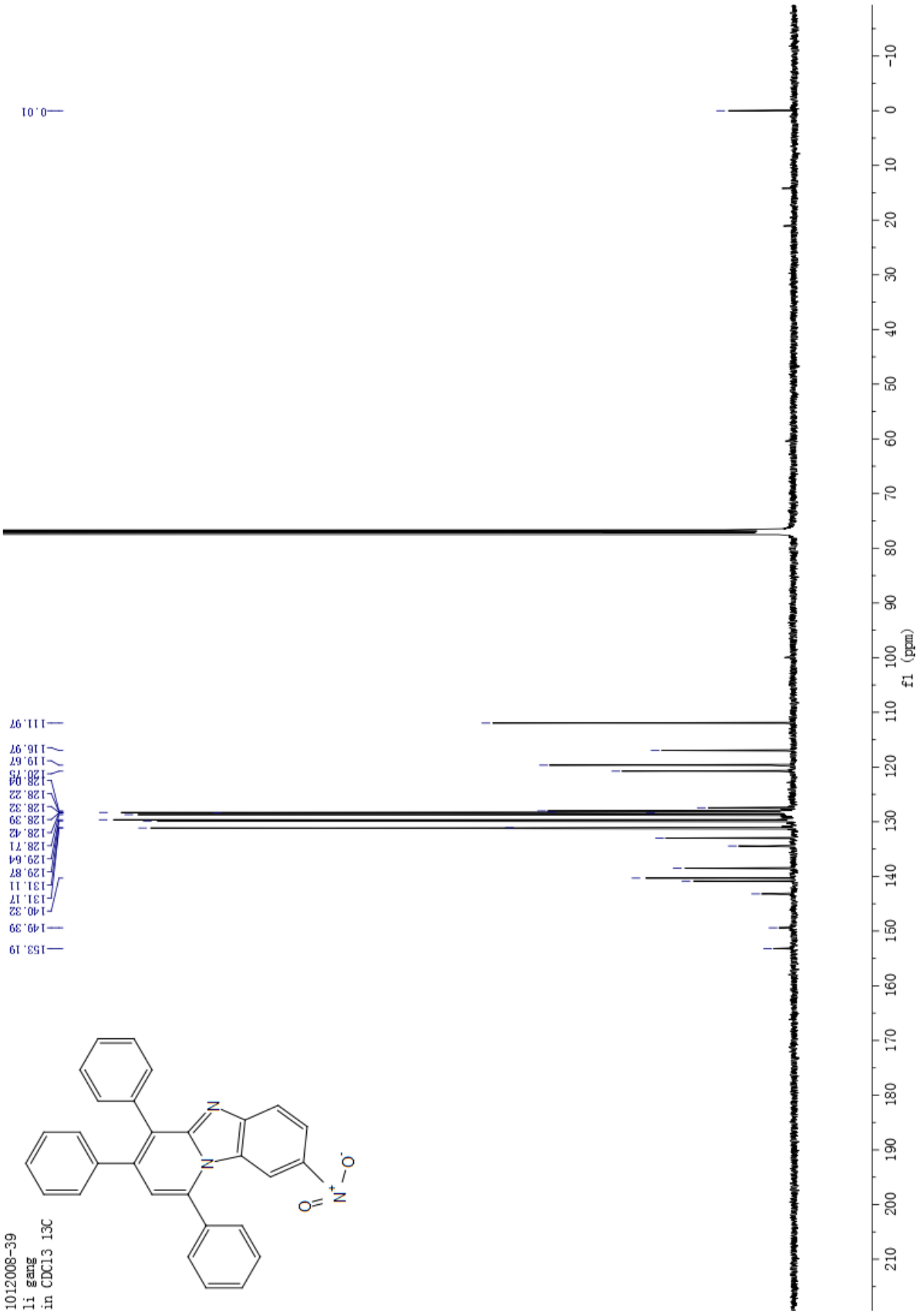
Compound 2d:



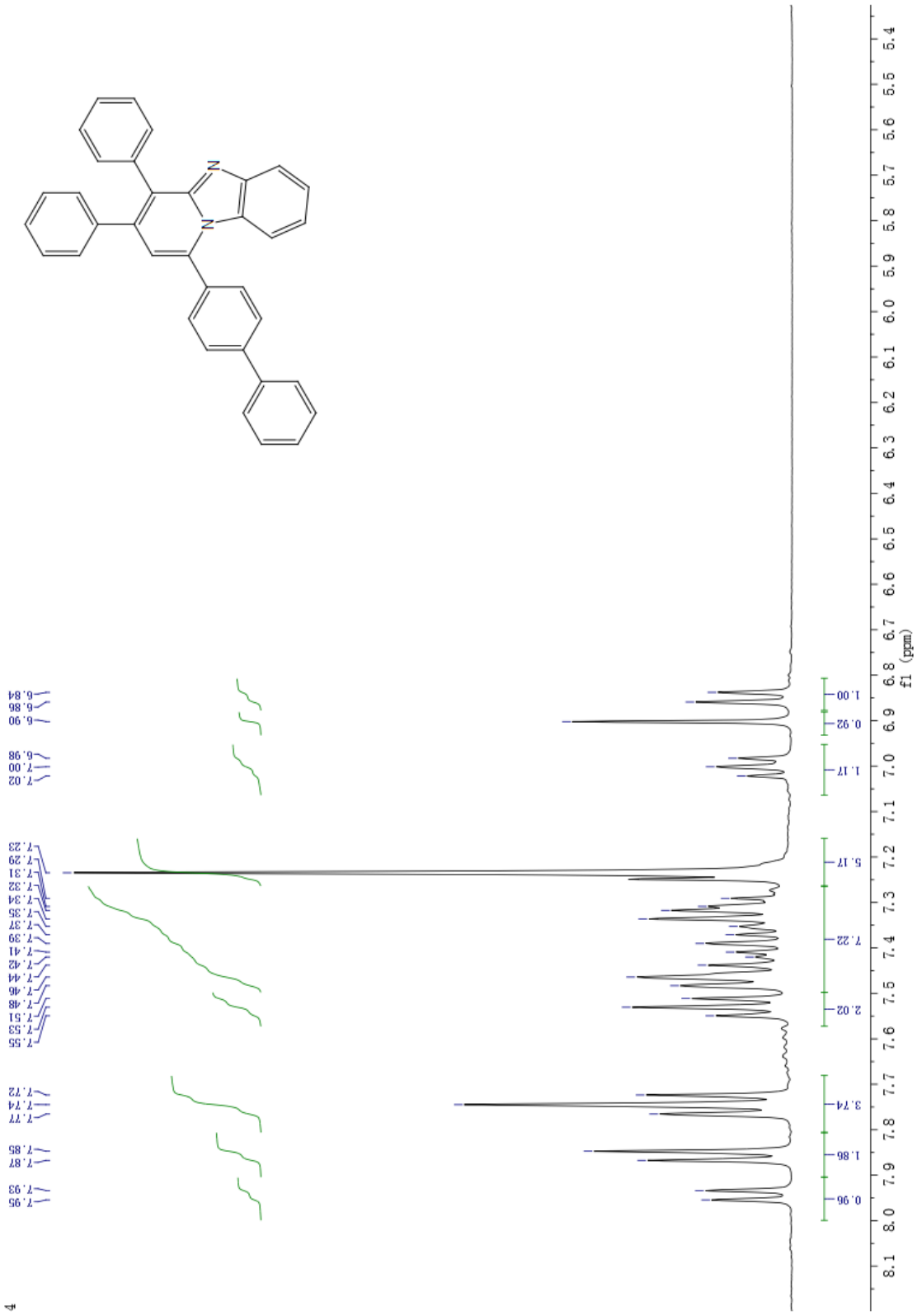
Compound 2e:



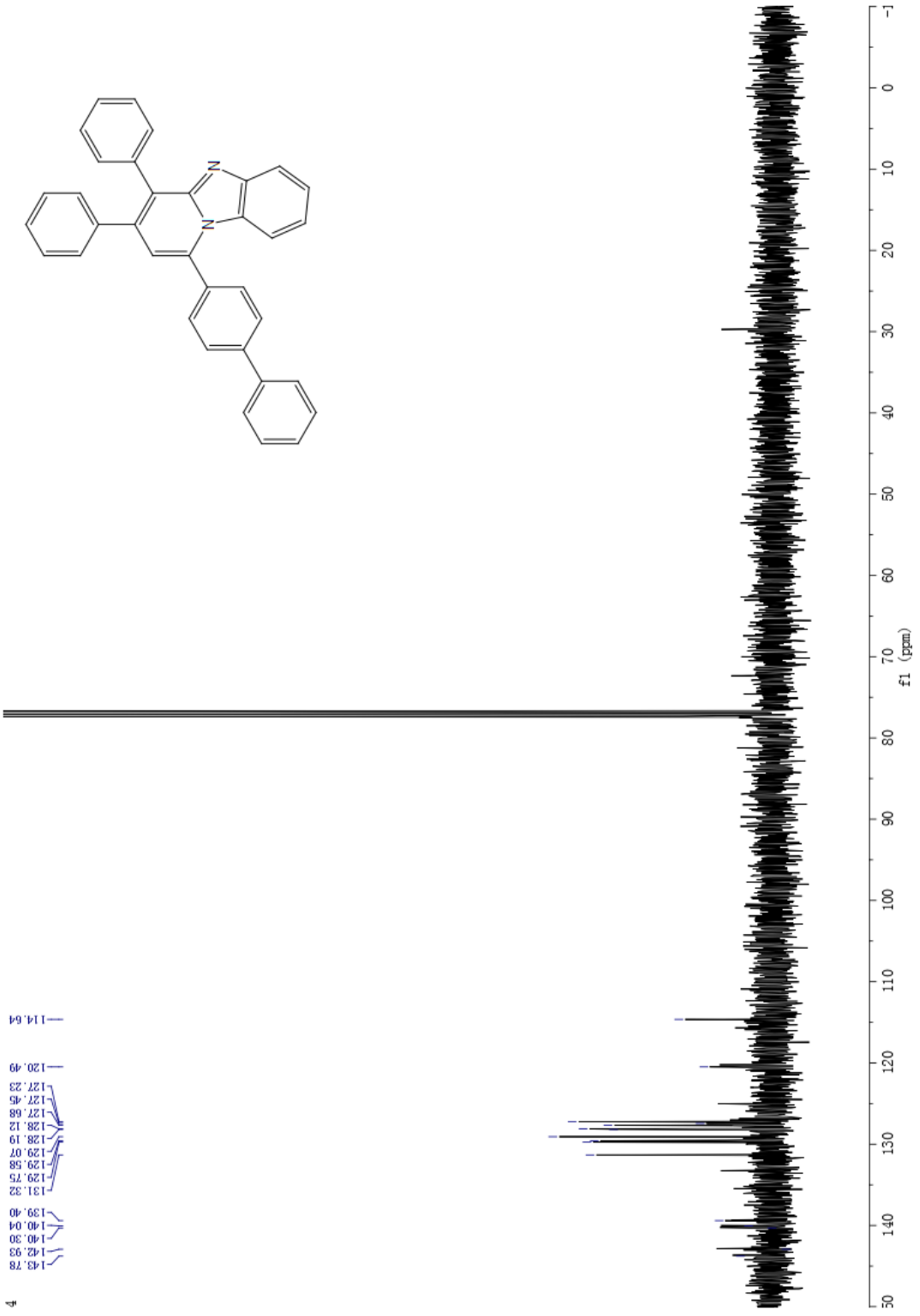
Compound 2e:



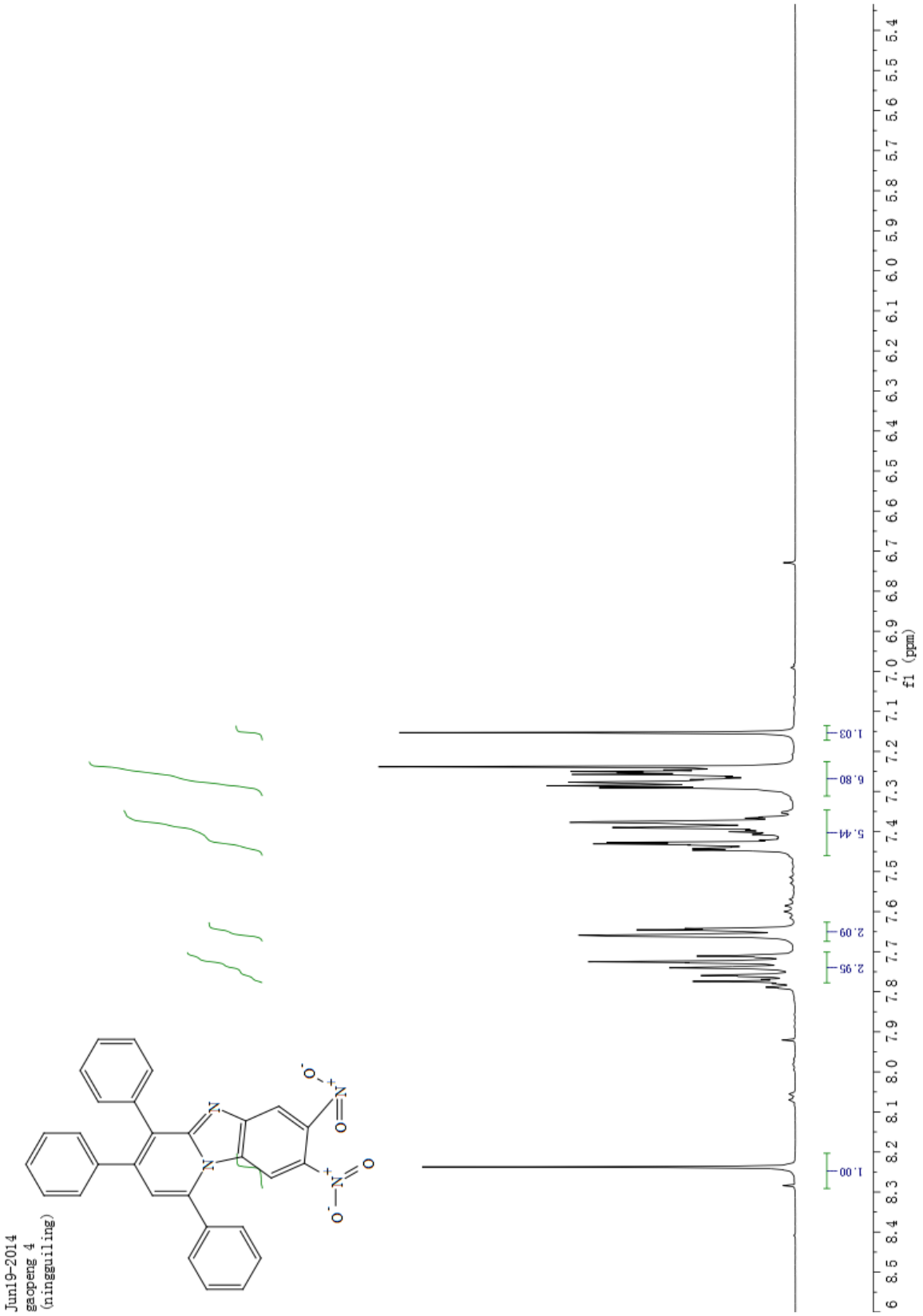
Compound 2f:



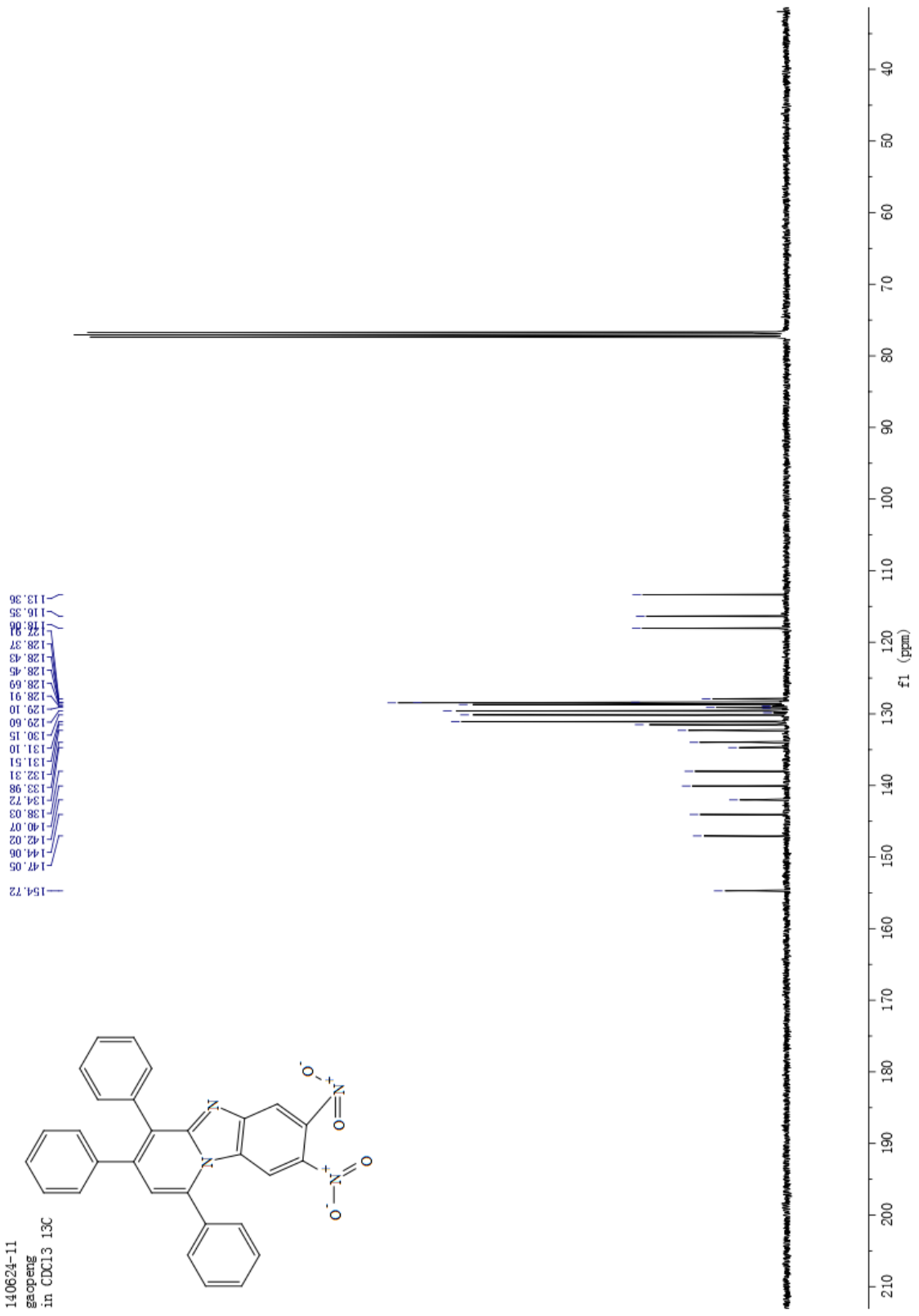
Compound 2f:


N
N
114.64
120.49
127.23
127.45
127.68
128.12
128.19
129.07
129.58
129.75
131.32
139.40
140.04
140.30
142.93
143.78
f1 (ppm)

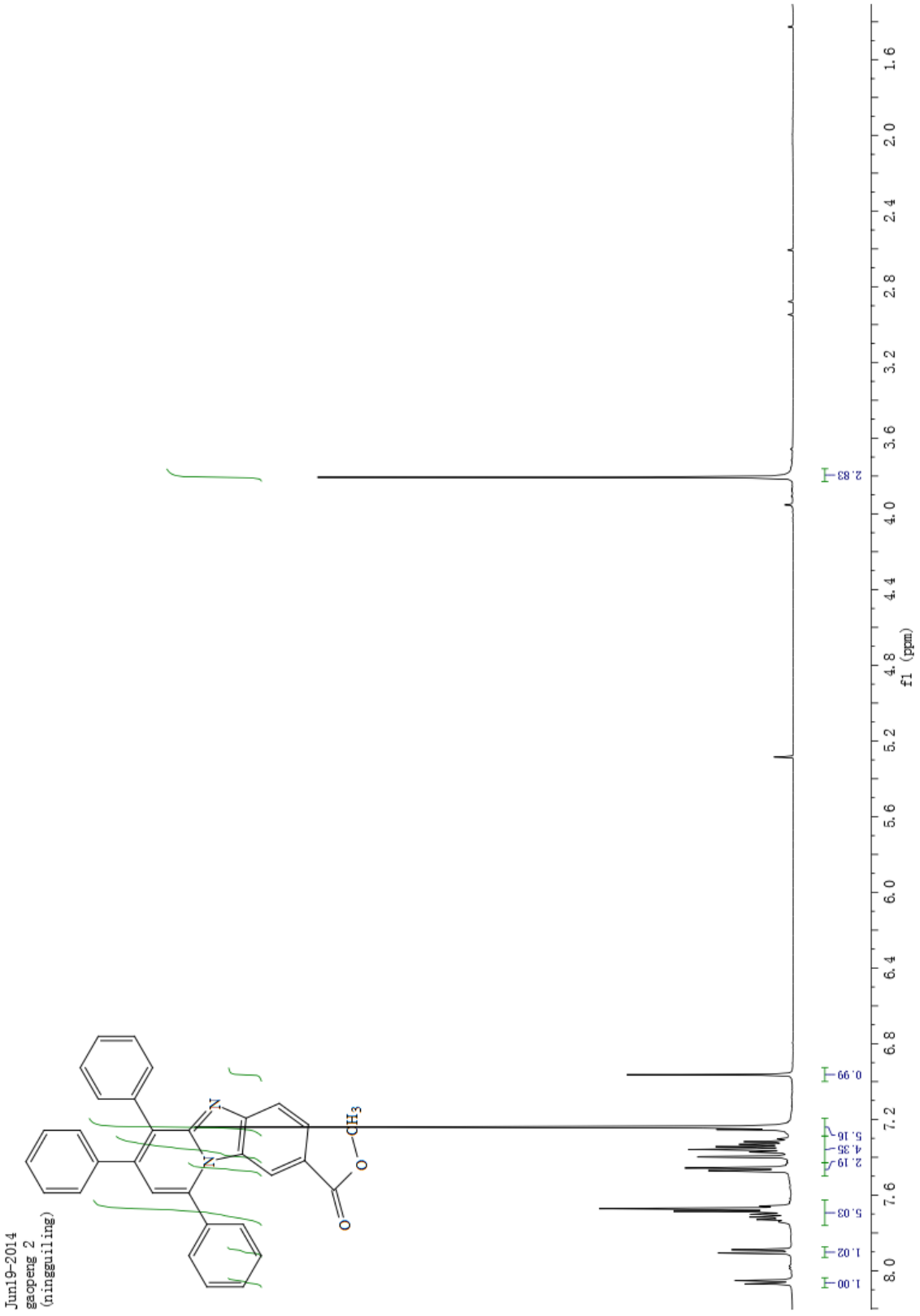
Compound 2m:


Jun19-2014
gaopeng 4
(ningguiling)
1.00
2.09
2.95
5.44
6.80
1.03
f1 (ppm)

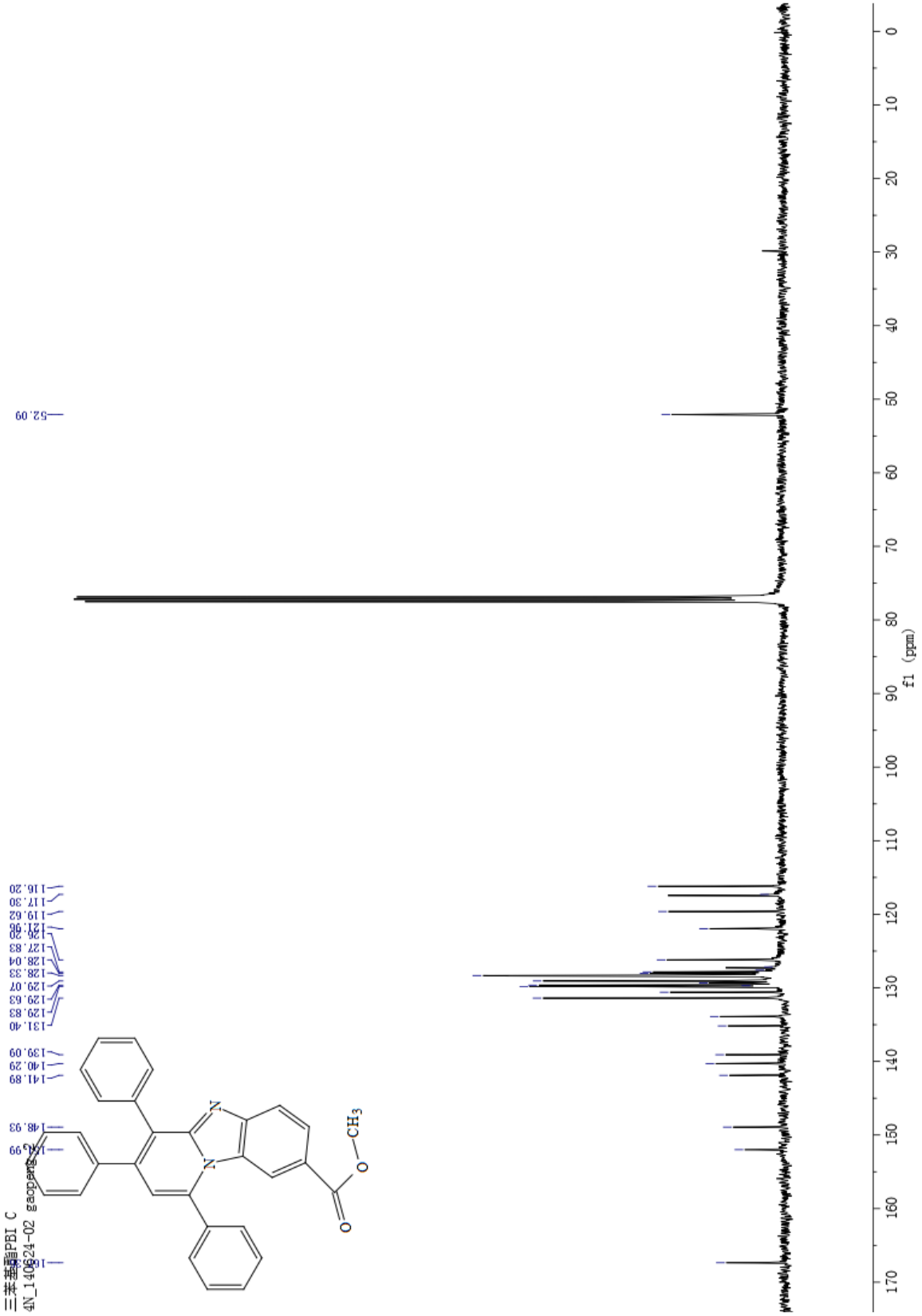
Compound 2m:


140624-11
gaopeng
in CDCl3 13C
154.72
147.05
144.06
142.02
140.07
138.03
134.72
133.98
132.31
131.51
131.10
130.15
129.60
129.10
128.91
128.69
128.46
128.37
122.91
118.06
116.35
113.36
f1 (ppm)

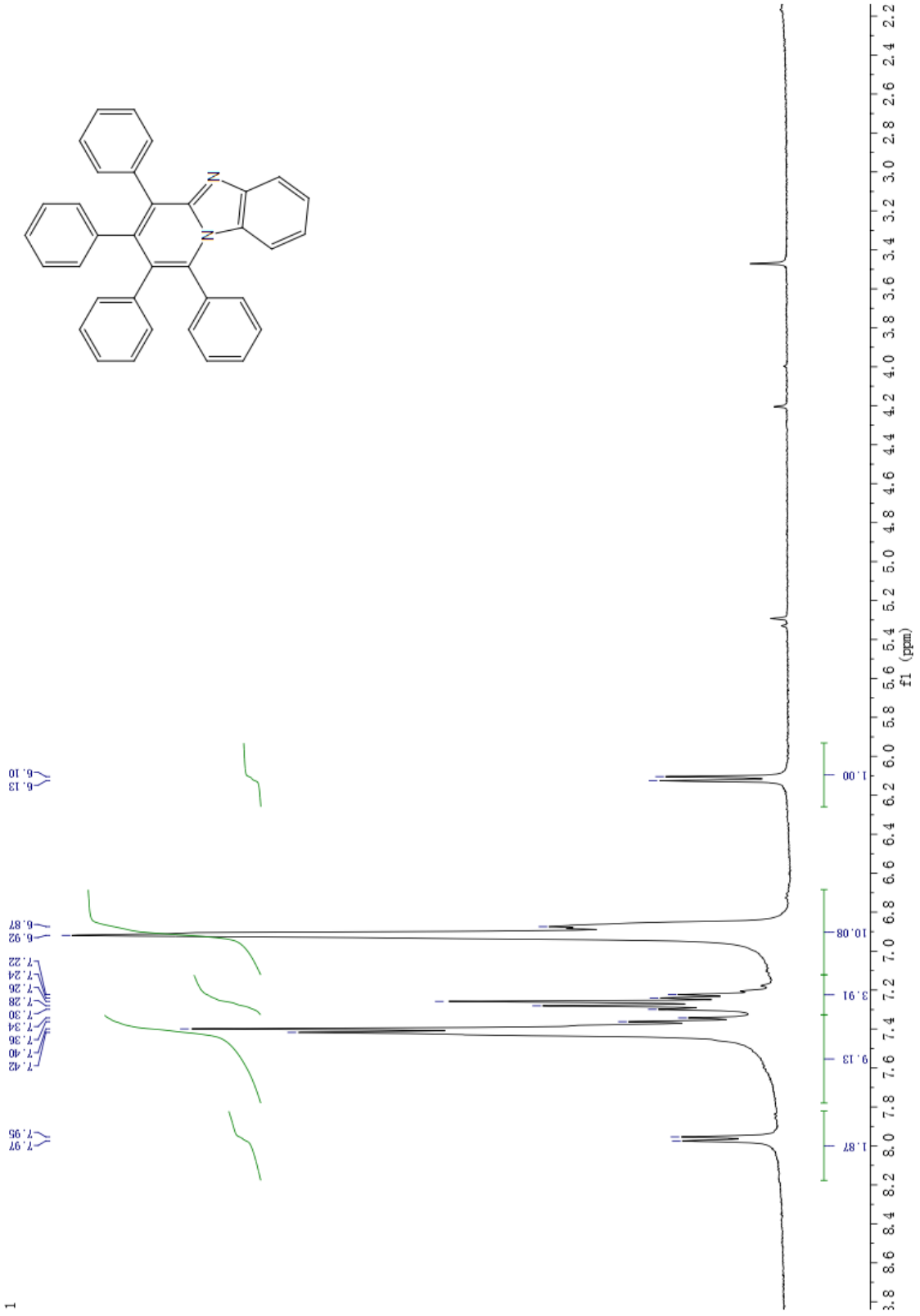
Compound 2k:


Jun19-2014
gaopeng 2
(ningguiling)
N
N
O
O
CH3
2.83
0.99
5.16
4.35
2.19
5.03
1.02
1.00
8.0
7.6
7.2
6.8
6.4
6.0
5.6
5.2
4.8
4.4
4.0
3.6
3.2
2.8
2.4
2.0
1.6
f1 (ppm)

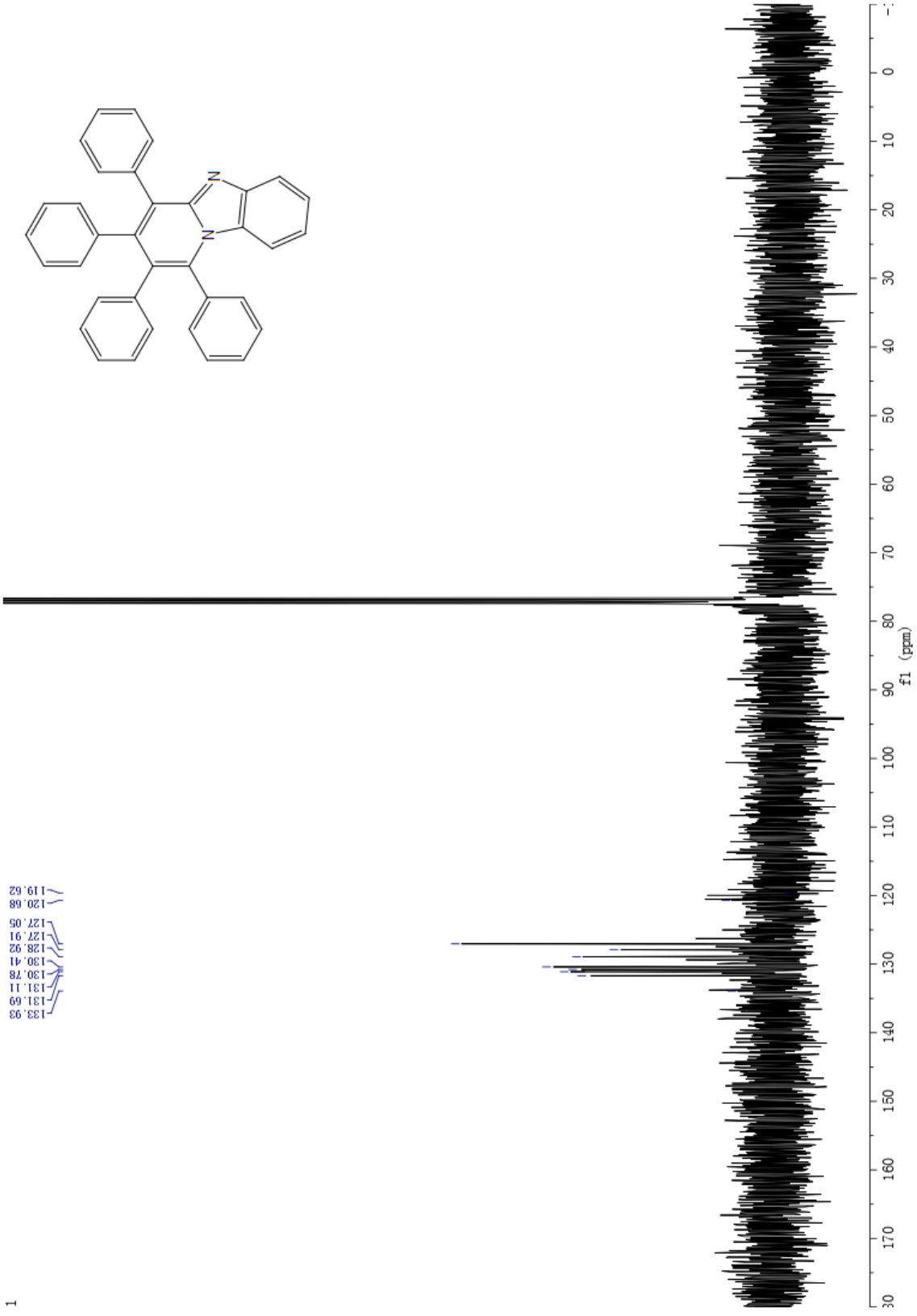
Compound 2k:


三苯基PBI C
4N_140624-02 gaopeng
52.09
116.20
117.30
119.62
121.96
126.20
127.83
128.04
128.33
129.07
129.63
129.83
131.40
139.09
140.29
141.89
148.93
f1 (ppm)

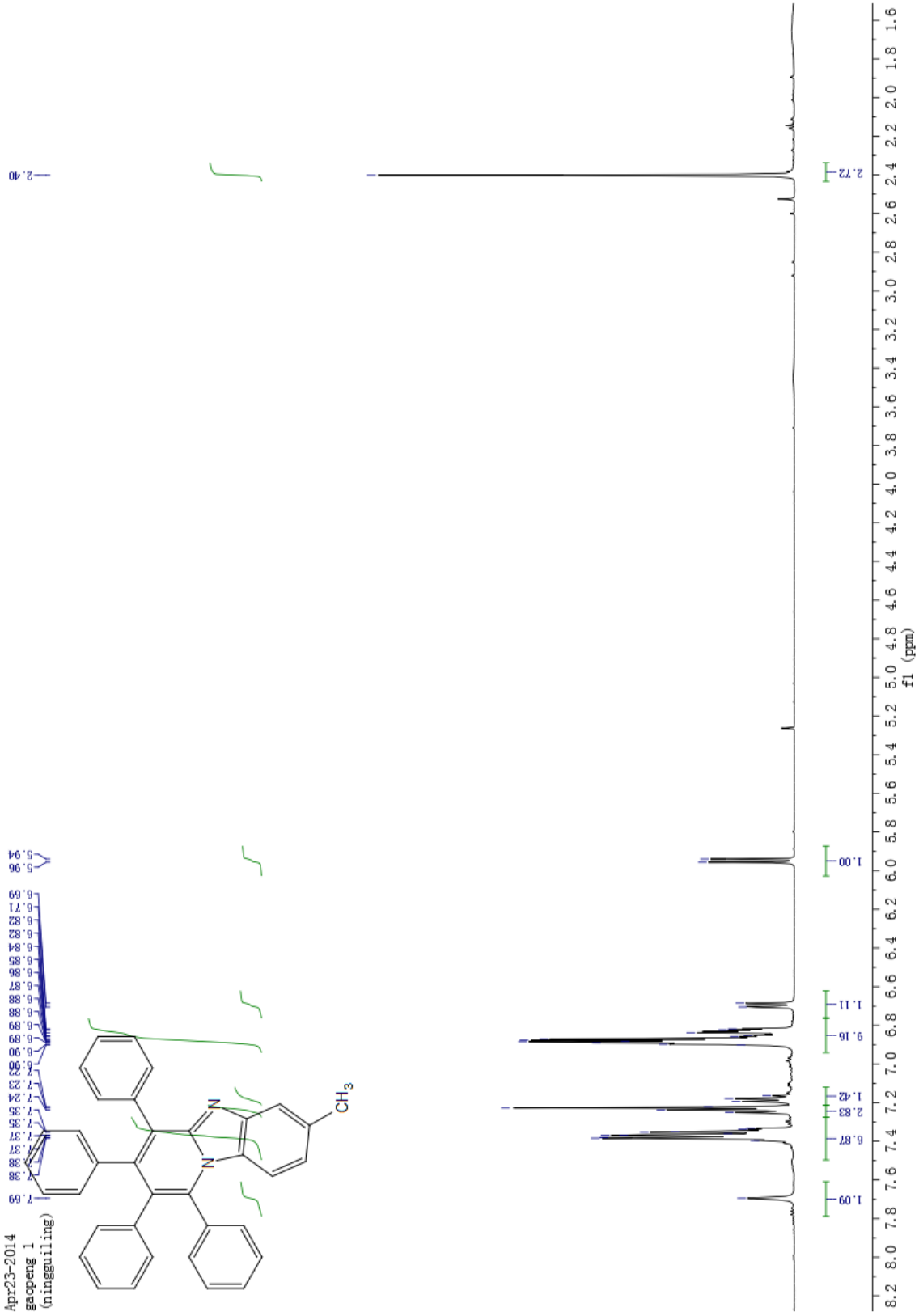
Compound 2g:


N
N
6.10
6.13
6.87
6.92
7.22
7.24
7.26
7.28
7.30
7.34
7.36
7.40
7.42
7.95
7.97
1.00
10.08
3.91
9.13
1.87
f1 (ppm)

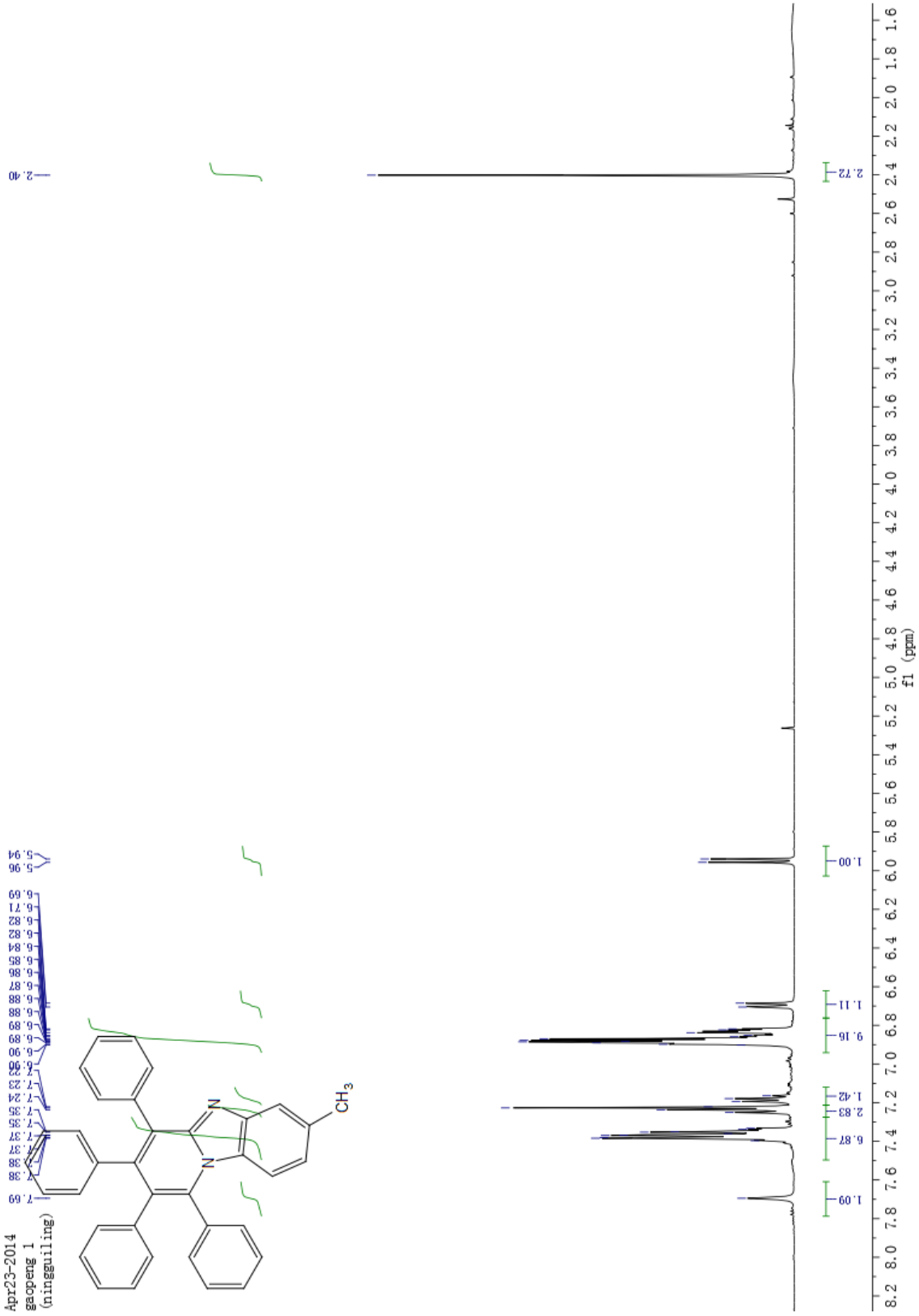
Compound 2g:


N
N
133.93
131.69
131.11
130.78
130.41
128.92
127.91
127.05
120.68
119.62
1
f1 (ppm)

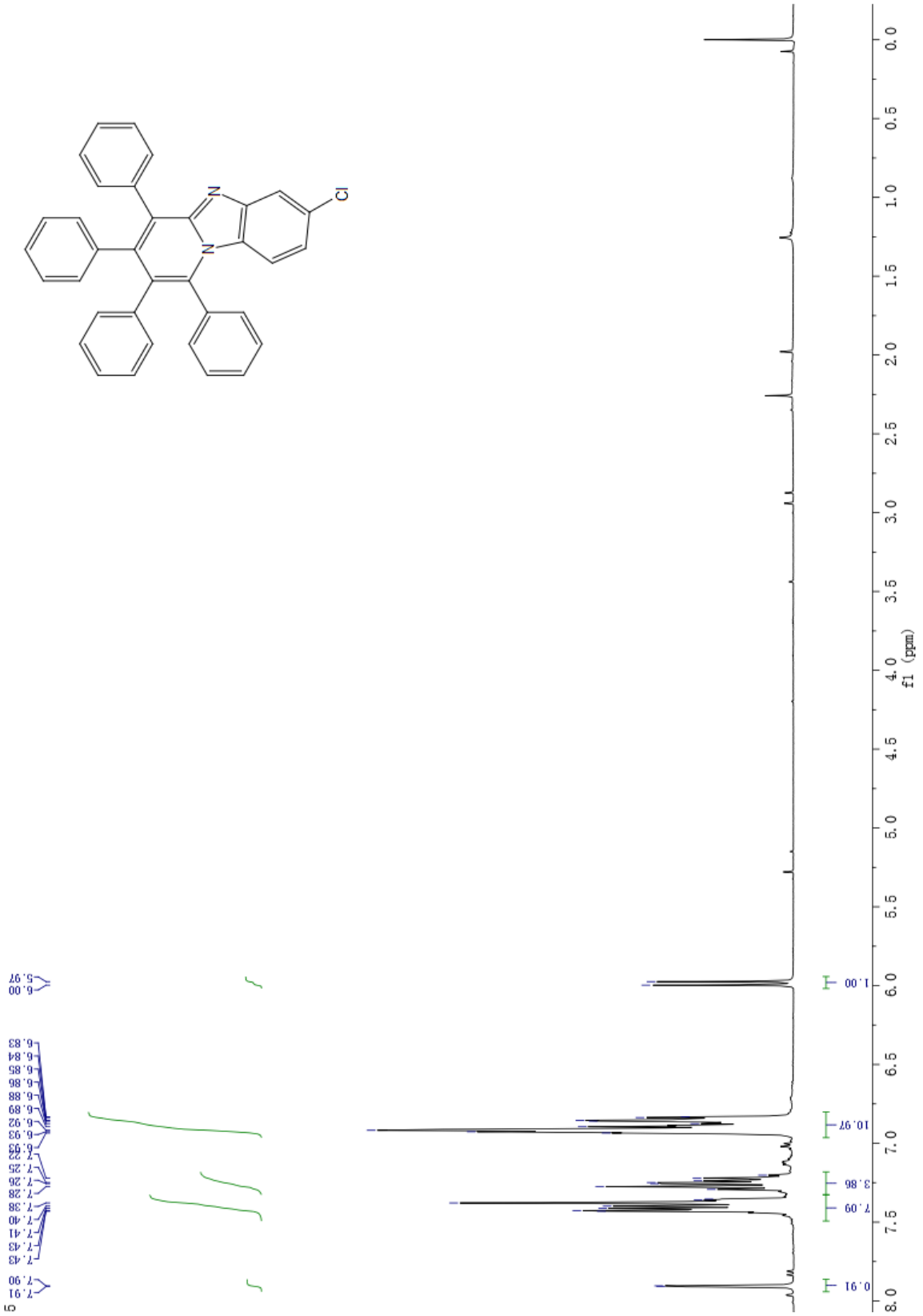
Compound 2h:



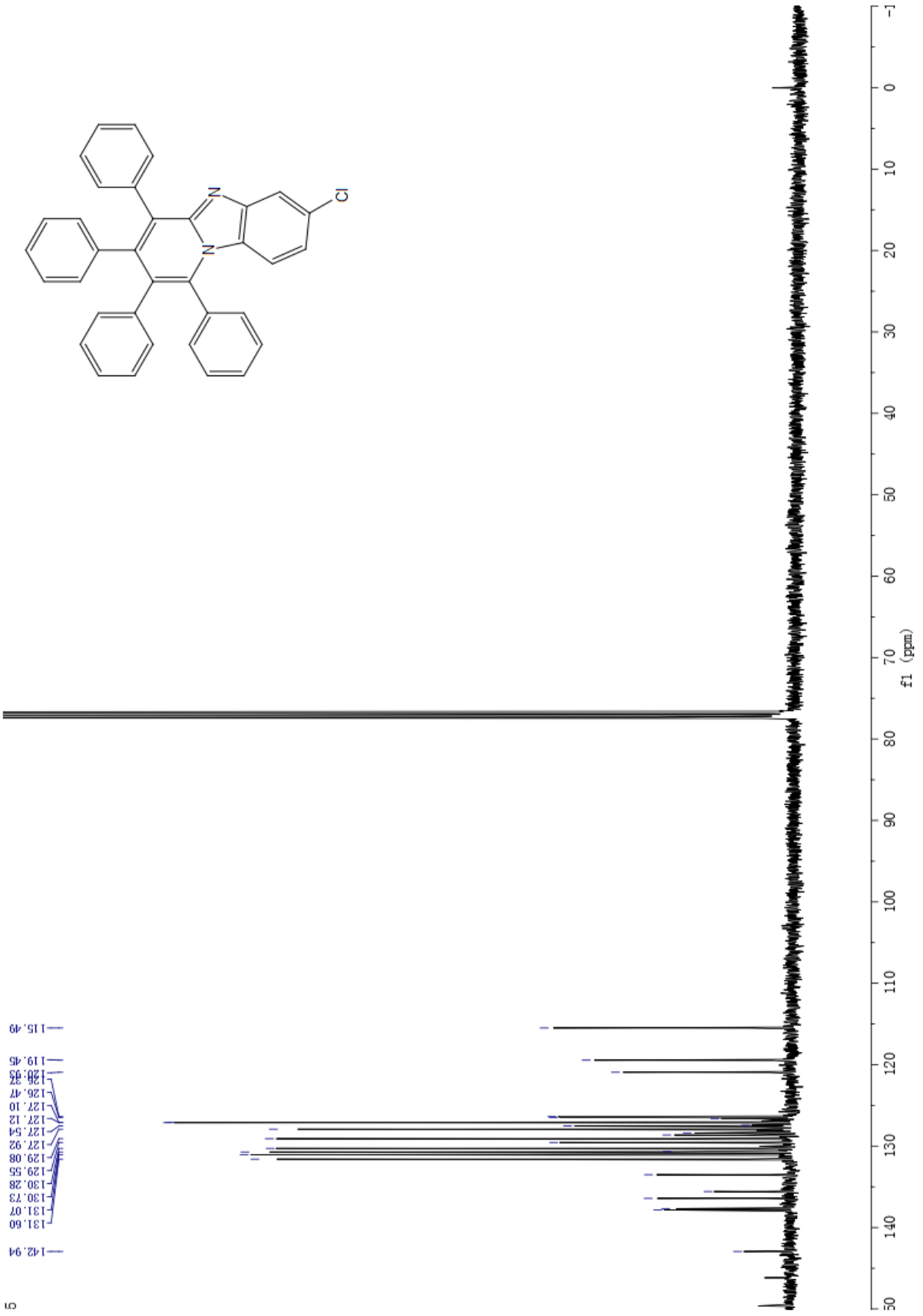
Compound 2h:



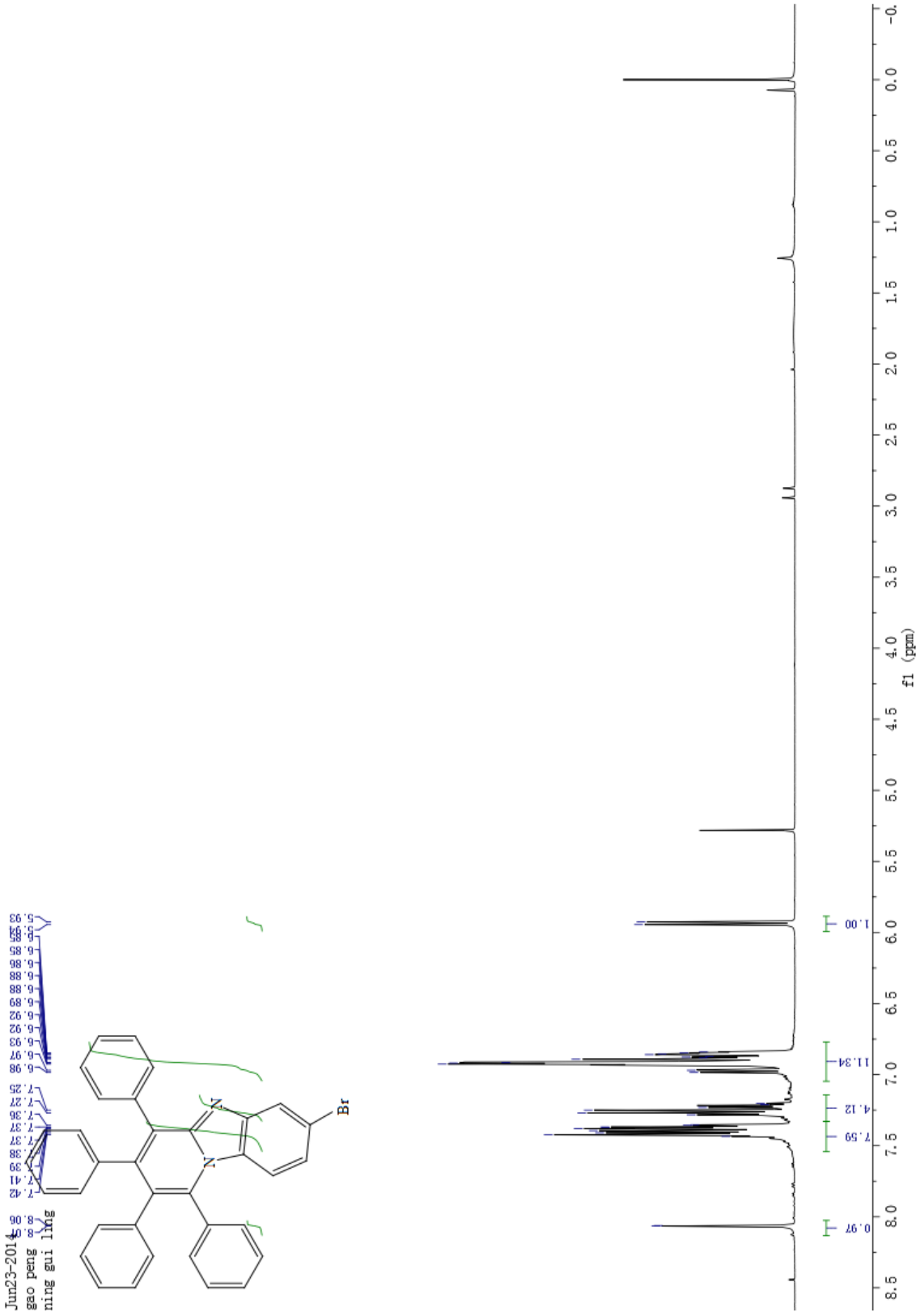
Compound 2i:



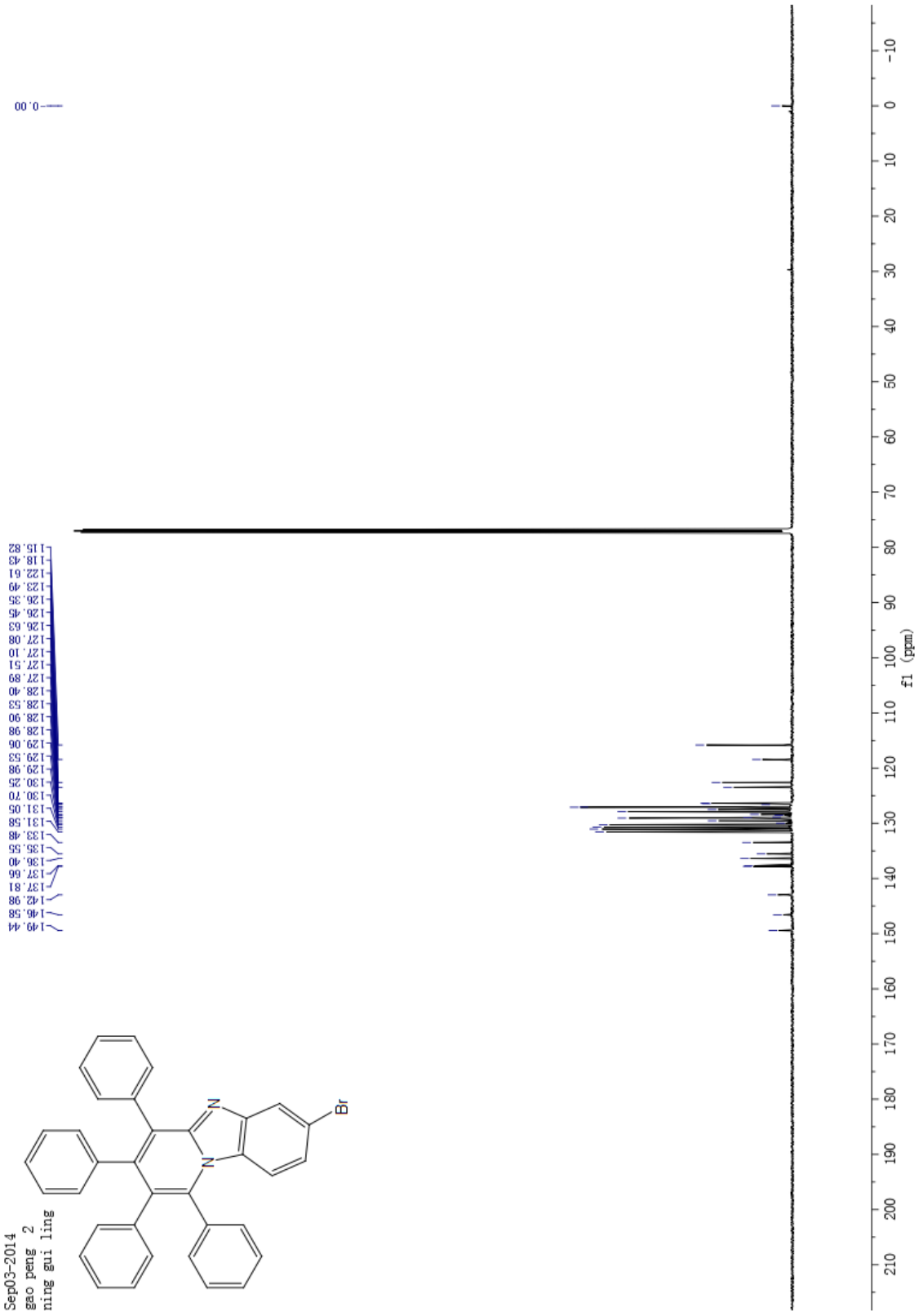
Compound 2i:



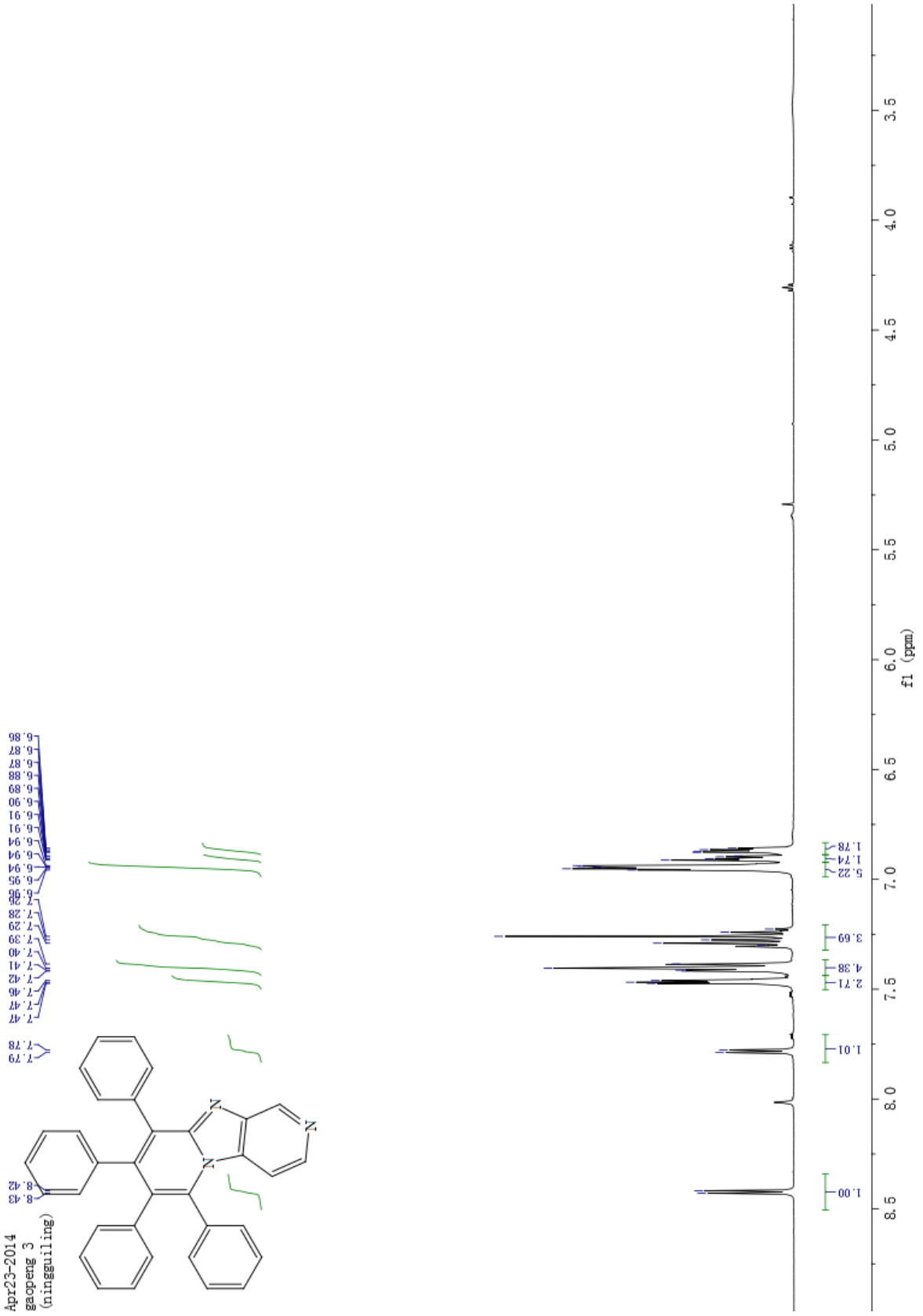
Compound 2l:


Jun23-2014
gao peng
ning gui ling
Br
N
N
1.00
11.34
4.12
7.59
0.97
f1 (ppm)

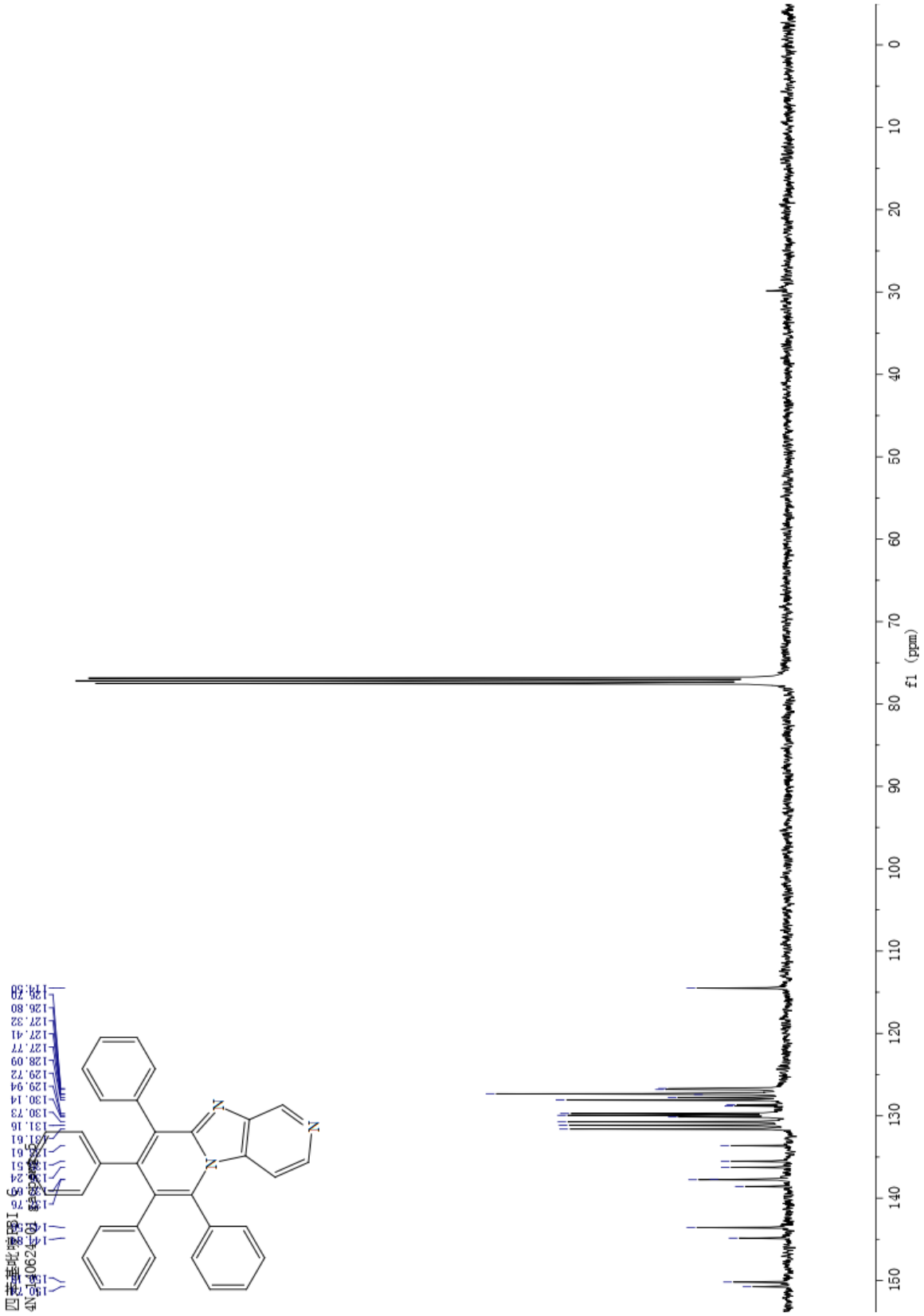
Compound 2l:



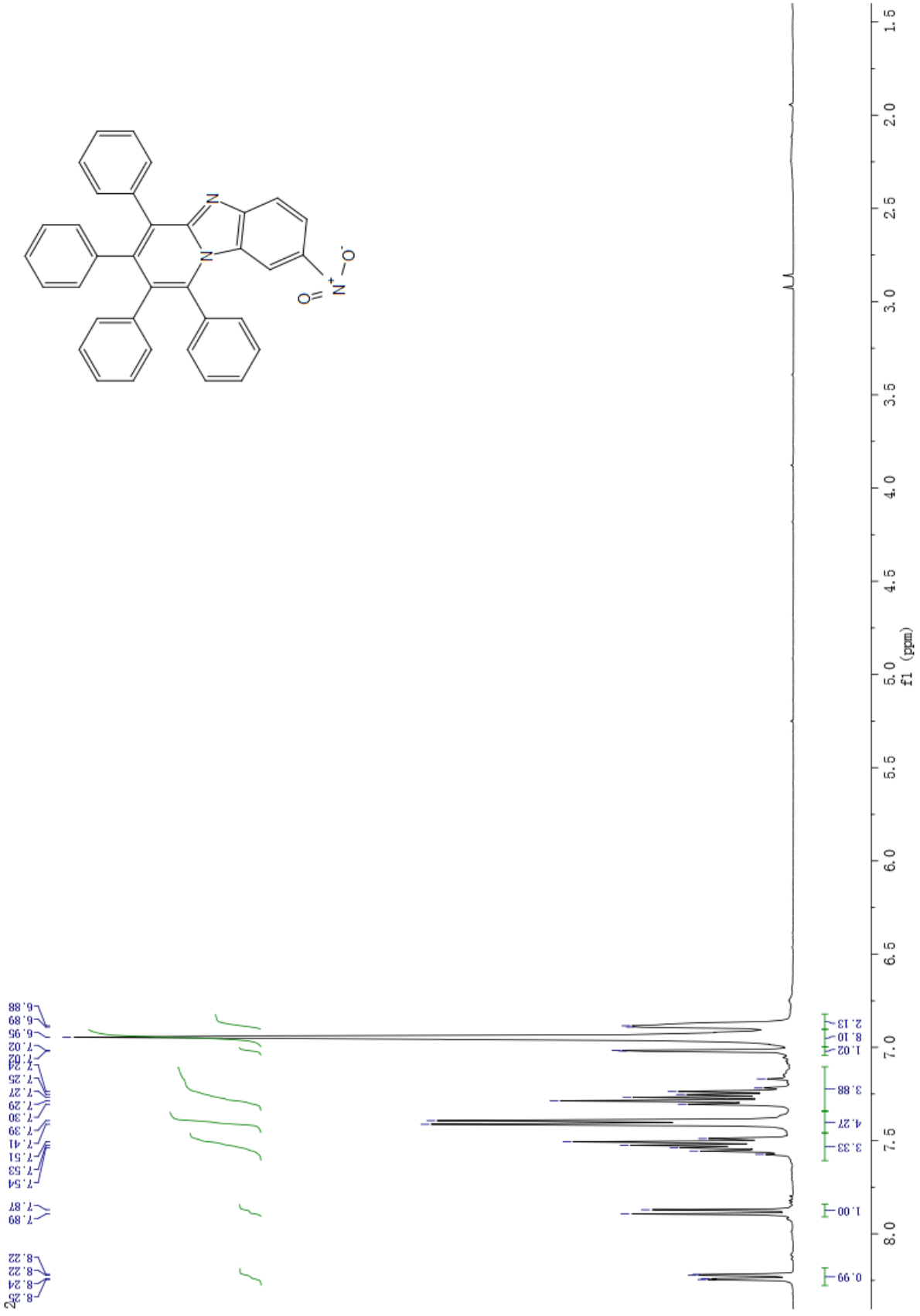
Compound 2p:



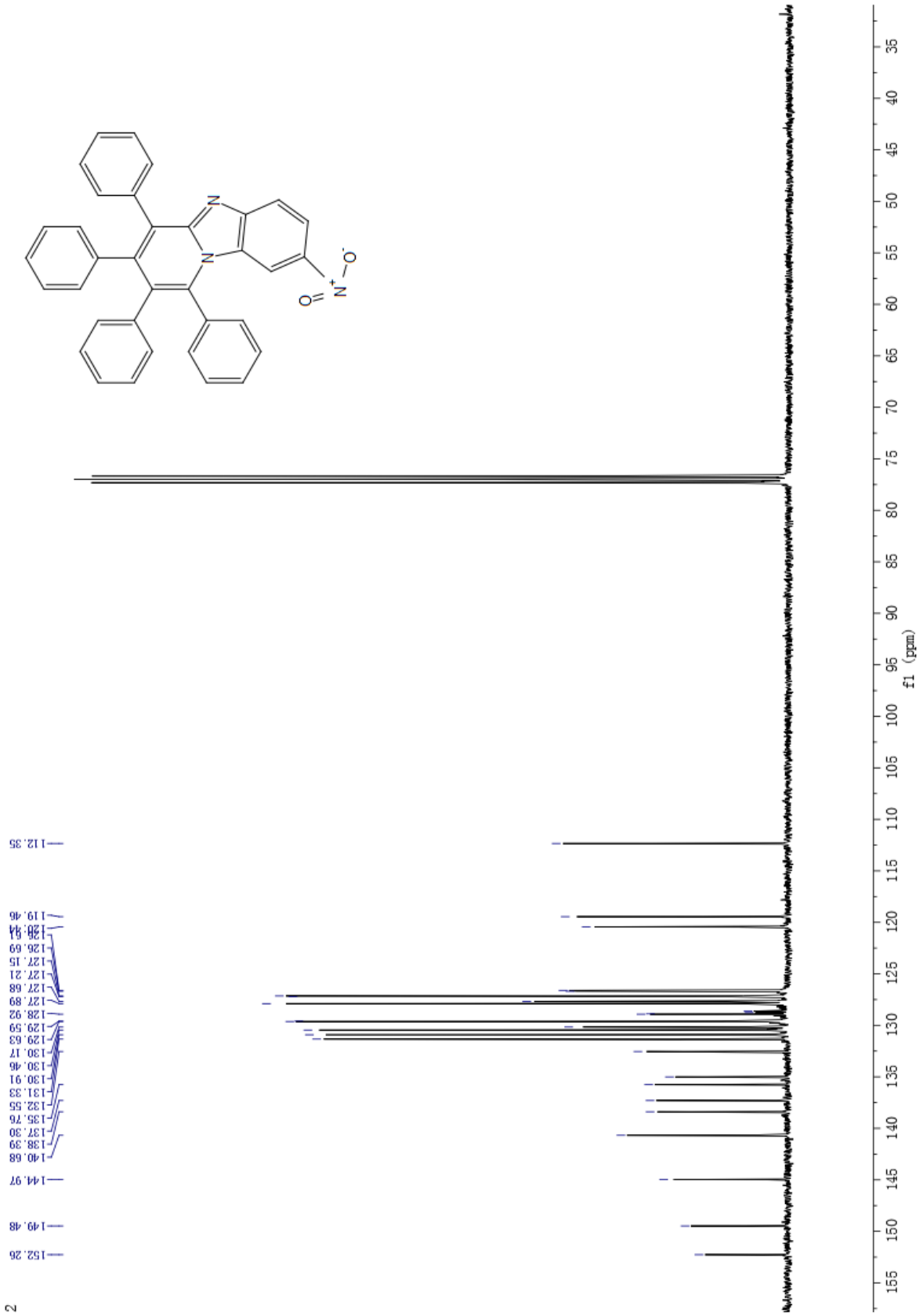
Compound 2p:



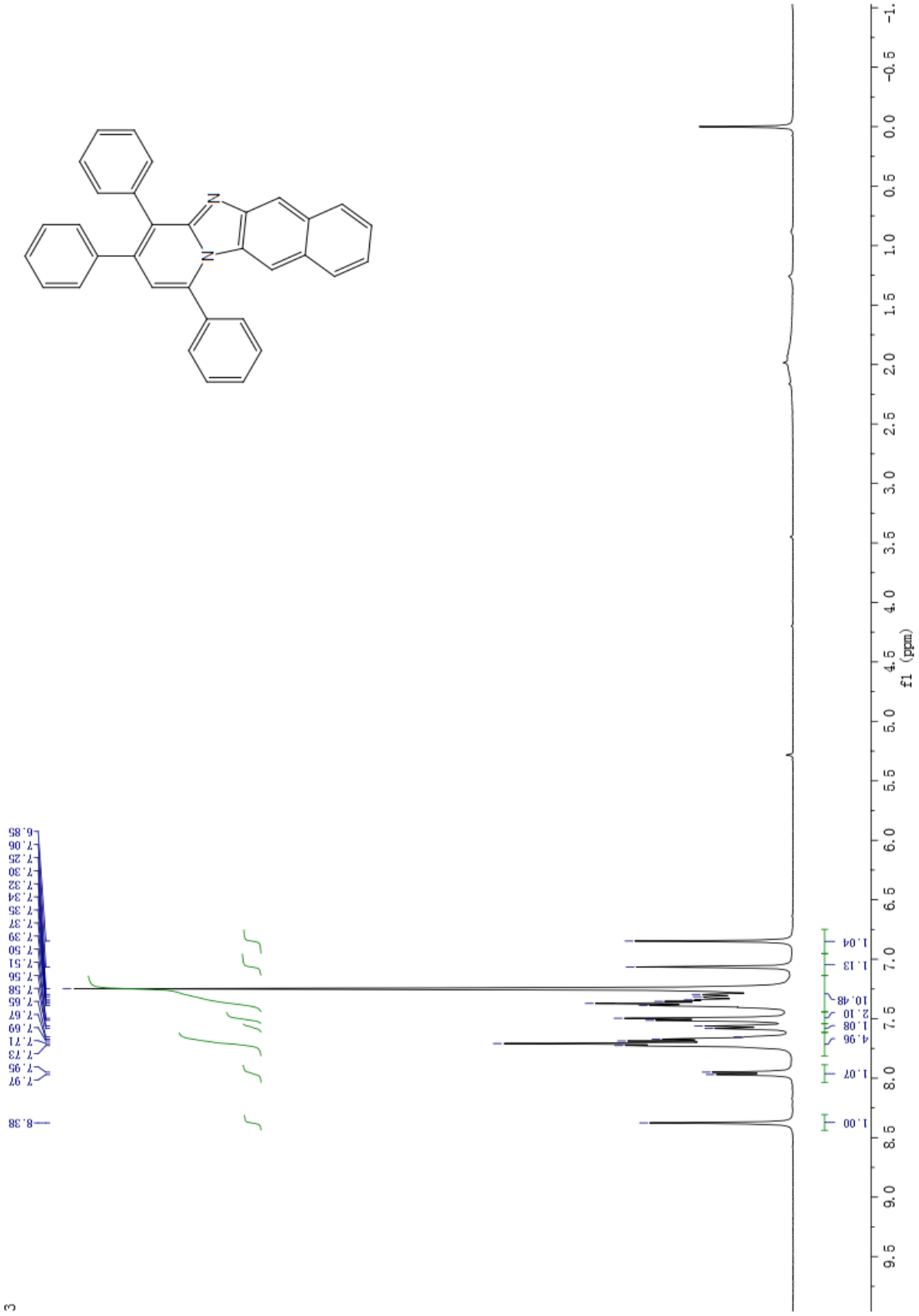
Compound 2j:



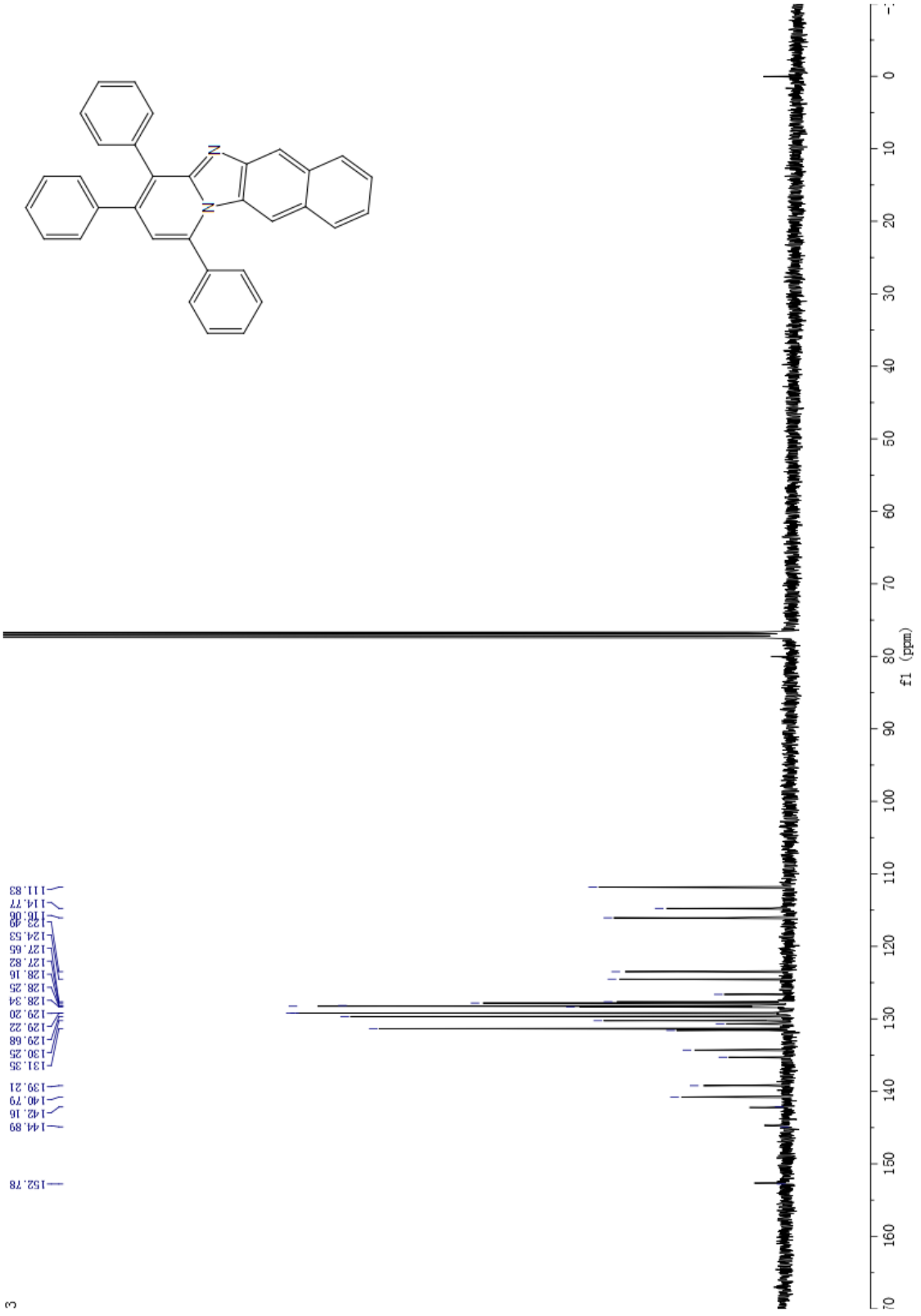
Compound 2j:



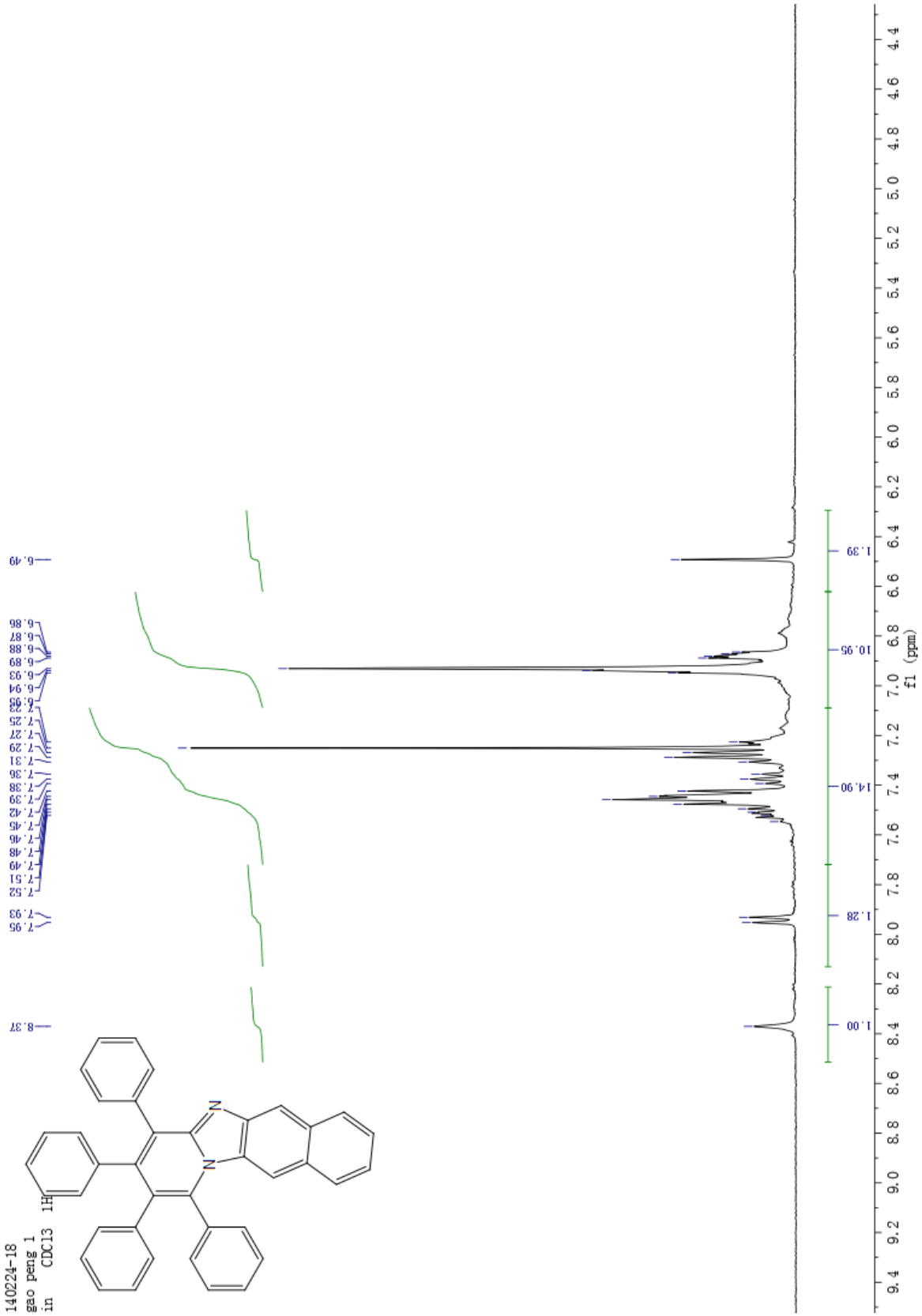
Compound 2n:



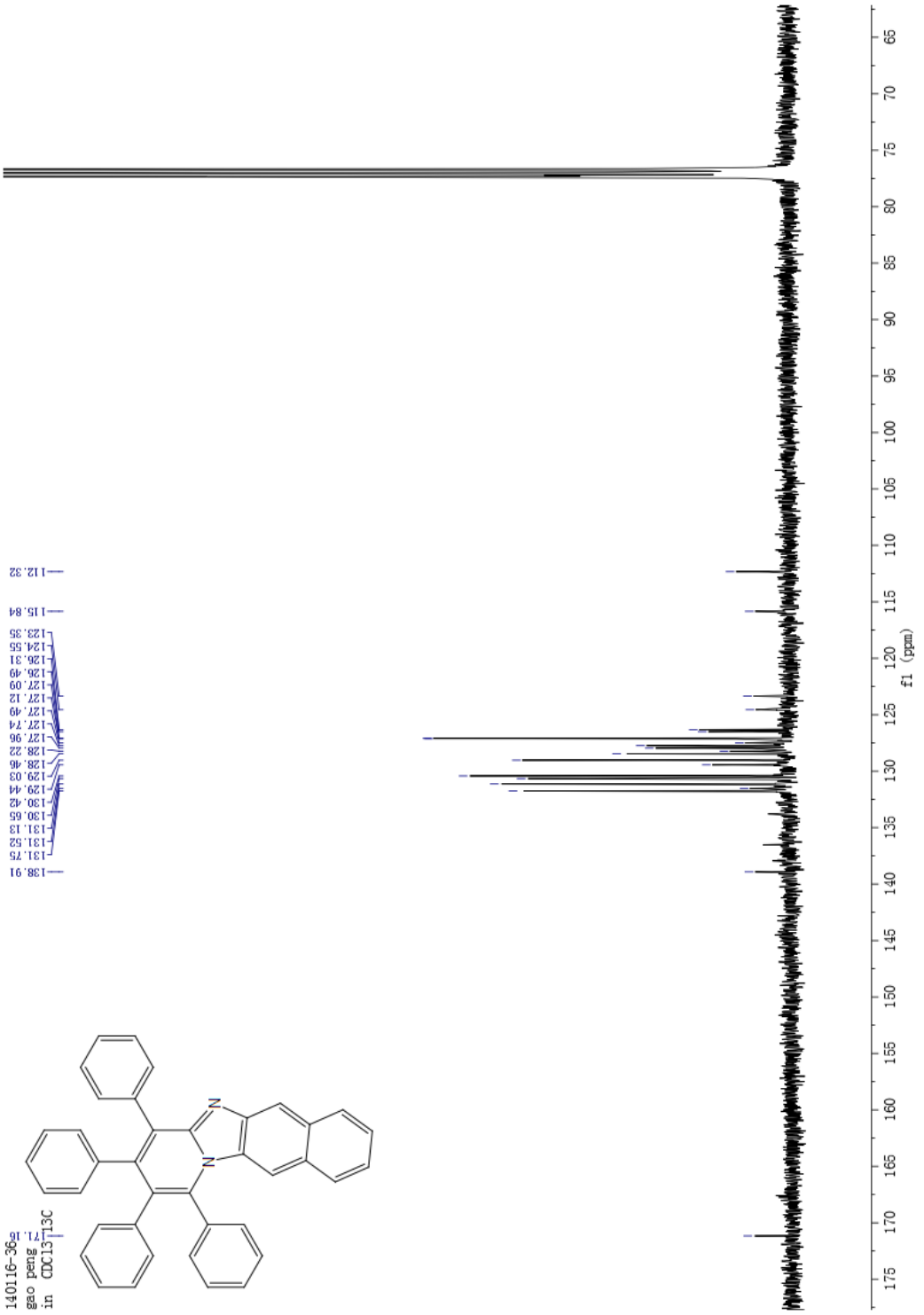
Compound 2n:


N
N
111.83
114.77
116.06
123.49
124.53
127.65
127.82
128.16
128.25
128.34
129.20
129.22
129.68
130.25
131.35
139.21
140.79
142.16
144.89
152.78
3
f1 (ppm)

Compound 2o:



Compound 2o:



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