Supporting Information

Metal-free efficient synthesis of aryl sulfonamides from N-hydroxy sulfonamide and amines

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Table of contents

1. Experimental Section

1.1. General Information

All starting materials and commercial reagent were purchased from Alfa Aesar, Sigma Aldrich, Avra, Spectrochem, TCI. Thin Layer Chromatography plates were visualizedby exposure to ultraviolet light (UV) with 254 nm of wavelength and then further analyzed byusing iodine chamber. Thin-layer chromatography was performed usingpre-coated plates. Column chromatography was performed in 120 to 200 mesh size silica gel.The reactions were carried out inround bottom flask and sealed tube. and all NMR spectra were recorded by Bruker Avance 400 spectrometer (^{1}H at 400 MHz and ^{13}C at 100 MHz). Chemical shifts for ^{1}H NMR spectra have been reported in parts per million (ppm) from tetramethylsilane with the solvent resonance as the internal standard ($CDCl_3$: δ 7.26 ppm). Simillarly,^{13}C NMR spectra have been reported in parts per million (ppm) from tetramethylsilane with the solvent as the internalstandard ($CDCl_3$: δ 77.0 ppm). The ^{1}H NMRand ^{13}C NMR of the known products were compared with literature reports.

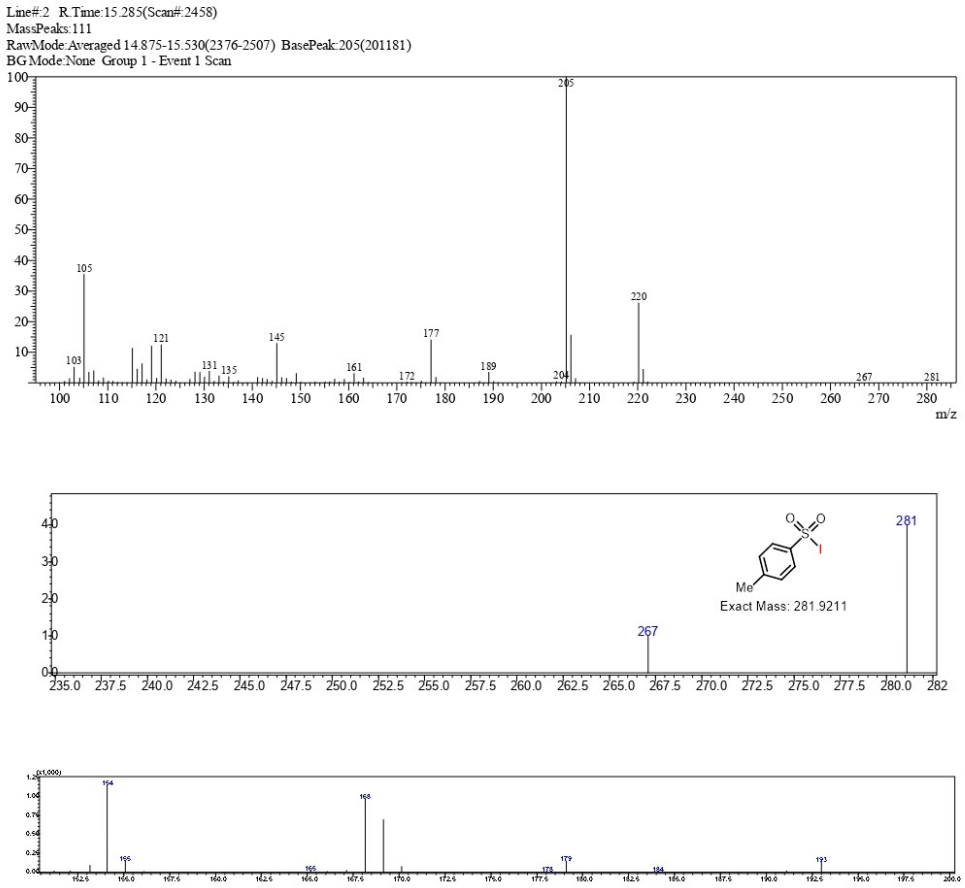
1.2. Synthesis of N-hydroxybenzenesulfonamide (1a):

In 100 mL round bottom flux, a solution of potassium carbonate (10 mmol, 1.38 g) in water was added dropwise to a solution of hydroxyl amine hydrochloride (10 mmol, 0.695 g) in water: methanol (3:2; ~5-6 mL) at 0 ℃ with vigorous magnetic starring for 1 hour. Then addition of ice-cold methanol was happened in single portion to this mixture followed by dropwise addition of toluene sulphonyl chloride (10 mmol, 1.90 g) over 1 hour. After 18 hours of stirring the mixture was filtered and methanol was removed under vacuums, the water residue extracted with ethyl acetate.

1.3 Synthesis of 1-(phenylsulfonyl)piperidine (3a):

In a 25 mL round bottom flask, N-hydroxybenzenesulfonamide (**1a**, 290 mg, 1 mmol), iodine (254 mg, 1 equiv.) and *tert*-butyl hydroperoxide (360 mg, 4 equiv.) was added in 2 mL 2-MeTHF solvent at normal temperature. After five minutes, piperidine (**2a**, 2 mmol, 170 mg) mixed into it. The resulting mixture was stirred at 70 ℃ for 6h. After the reaction completed, monitoring by TLC, 10mL of cold water was added to the mixture, then extracted with EtOAc three times (3 × 15 mL). The extract was washed with 10% Na_2SO_3 solution (w/w), dried over anhydrous Na_2SO_4 and evaporation. The residue was purified by column chromatography on silica gel (petroleum ether/EtOAc) to yield the desired product **3a** as a pale-yellow solid.

1.4. GC-MS spectra and data:



2. ^{1}H and ^{13}C data of compound:

1-(phenylsulfonyl)piperidine (3a):[1] Yield: 87%, 195 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.84 (dd, *J* = 7.0, 1.5 Hz, 2H), 7.58 (dd, *J* = 5.0, 3.6 Hz, 1H), 7.56 – 7.50 (m, 2H), 3.25 (t, *J* = 6.8 Hz, 4H), 1.80 – 1.68 (m, 4H), 1.64 – 1.53 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 137.01, 132.55, 129.00, 127.50, 47.93, 25.24, 23.50.

1-(phenylsulfonyl)pyrrolidine (3b):[1] Yield: 76%, 160 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 – 7.76 (m, 2H), 7.55 (dd, *J* = 16.1, 10.0 Hz, 3H), 3.25 (ddd, *J* = 6.8, 4.4, 2.7 Hz, 4H), 1.87 – 1.68 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 137.01, 132.49, 129.00, 127.56, 48.19, 25.03.

4-(phenylsulfonyl)morpholine (3c):[1] Yield: 83%, 188 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (dd, *J* = 8.4, 3.4 Hz, 2H), 7.63 (dd, *J* = 9.8, 3.7 Hz, 1H), 7.56 (dd, *J* = 7.3, 5.9 Hz, 2H), 3.83 – 3.66 (m, 4H), 3.10 – 2.87 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 135.13, 133.09, 129.15, 127.85, 66.11, 46.00.

2-(1-(phenylsulfonyl)piperidin-4-yl)-1H-benzo[d]imidazole (3d): Yield: 73%, 248 mg, gummy mass; ^{1}H NMR (400 MHz, DMSO) δ 7.90 – 7.68 (m, 5H), 7.54 (dd, *J* = 5.9, 3.2 Hz, 2H), 7.20 (dd, *J* = 6.0, 3.2 Hz, 2H), 3.77 (d, *J* = 12.0 Hz, 2H), 2.97 (tt, *J* = 11.2, 3.7 Hz, 1H), 2.53 (dd, *J* = 11.7, 2.3 Hz, 2H), 2.19 (dd, *J* = 13.4, 2.9 Hz, 2H), 2.01 – 1.81 (m, 2H). ^{13}C NMR (101 MHz, DMSO) δ 157.14, 136.09, 134.86, 133.63, 129.91, 127.90, 122.10 (d, *J* = 9.2 Hz), 46.02, 34.81, 29.90. HRMS (ESI) calcd for $C_{19}H_{19}N_3O_2S$ $[M+H]^+$ 342.1271; found 342.1292.

1-(2-methoxyphenyl)-4-(phenylsulfonyl)piperazine (3e): Yield: 71%, 235 mg, gummy mass; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88 – 7.72 (m, 2H), 7.57 (dt, *J* = 8.6, 7.2 Hz, 3H), 7.10 – 6.96 (m, 1H), 6.94 – 6.88 (m, 2H), 6.84 (d, *J* = 8.2 Hz, 1H), 3.80 (s, 3H), 3.17 (dt, *J* = 9.5, 4.4 Hz, 8H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 152.08, 140.27, 135.40, 132.92, 129.11, 127.92, 123.72, 121.07, 118.49, 111.13, 55.36, 50.03, 46.35. HRMS (ESI) calcd for $C_{18}H_{20}N_2O_3S$ $[M+H]^+$ 333.1267; found 333.1278.

1-(phenylsulfonyl)-1H-imidazole (3f):[3] Yield: 78%, 162 mg; ^{1}H NMR (400 MHz, DMSO) δ 9.10 (s, 1H), 7.69 (d, *J* = 1.2 Hz, 2H), 7.63 (dd, *J* = 6.6, 3.1 Hz, 2H), 7.39 – 7.25 (m, 3H). ^{13}C NMR (101 MHz, DMSO) δ 148.66, 134.87, 129.02, 128.17, 125.94, 119.79.

N-benzylbenzenesulfonamide (3g):[1] Yield: 75%, 185 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87 (dd, *J* = 5.3, 3.3 Hz, 2H), 7.58 (d, *J* = 7.3 Hz, 1H), 7.55 – 7.48 (m, 2H), 7.30 – 7.23 (m, 3H), 7.18 (dd, *J* = 7.3, 2.0 Hz, 2H), 4.77 (s, 1H), 4.14 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 139.94), 136.17, 132.73, 129.16, 128.74, 127.93 (d, *J* = 11.5 Hz), 127.13, 47.32.

N-(2-fluorobenzyl)benzenesulfonamide (3h): Yield: 69%, 182 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86 – 7.78 (m, 2H), 7.51 (dd, *J* = 4.9, 3.7 Hz, 1H), 7.44 (t, *J* = 7.6 Hz, 2H), 7.22 (ddd, *J* = 15.7, 12.4, 6.3 Hz, 2H), 7.06 – 6.97 (m, 1H), 6.96 – 6.88 (m, 1H), 5.16 (t, *J* = 6.2 Hz, 1H), 4.22 (d, *J* = 6.4 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 161.92, 159.47, 139.93, 132.65, 130.12 (d, *J* = 4.0 Hz), 129.75 (d, *J* = 8.2 Hz), 129.05, 127.02, 124.30 (d, *J* = 3.6 Hz), 123.44 (d, *J* = 14.4 Hz), 115.43, 115.22, 41.32 (d, *J* = 3.9 Hz). HRMS (ESI) calcd for $C_{14}H_{12}FNO_2S$ $[M+H]^+$ 266.0646; found 266.0659.

N-(4-chlorobenzyl)benzenesulfonamide (3i):[1] Yield: 70%, 196 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.84 (dd, *J* = 5.2, 3.3 Hz, 2H), 7.59 (ddd, *J* = 6.6, 4.4, 1.2 Hz, 1H), 7.54 – 7.46 (m, 2H), 7.25 – 7.19 (m, 2H), 7.12 (d, *J* = 8.5 Hz, 2H), 4.99 (t, *J* = 6.0 Hz, 1H), 4.11 (d, *J* = 6.3 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 139.85, 134.83, 133.75, 132.81, 129.21 (d, *J* = 2.9 Hz), 128.81, 127.06, 46.56.

N-propylbenzenesulfonamide (3j):[4] Yield: 66%, 161 mg**;** ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.92 – 7.86 (m, 2H), 7.62 – 7.48 (m, 3H), 5.05 (t, *J* = 5.6 Hz, 1H), 2.91 (dd, *J* = 13.4, 7.0 Hz, 2H), 1.48 (dd, *J* = 14.5, 7.3 Hz, 2H), 0.86 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 140.03, 132.56, 129.09, 127.02, 44.98, 22.89, 11.09**.**

N-butylbenzenesulfonamide (3k):[1] Yield: 63%, 134 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88 (dd, *J* = 7.1, 1.5 Hz, 2H), 7.62 – 7.46 (m, 3H), 4.95 (s, 1H), 2.94 (dd, *J* = 12.0, 6.8 Hz, 2H), 1.52 – 1.39 (m, 2H), 1.31 – 1.24 (m, 2H), 0.83 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 140.00, 132.55, 129.08, 127.13, 42.94, 31.72, 19.57, 13.64.

N,N-diethylbenzenesulfonamide (3l):[5] Yield: 68%, 144 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (dd, *J* = 7.5, 2.1 Hz, 2H), 7.66 – 7.41 (m, 3H), 3.25 (q, *J* = 7.2 Hz, 4H), 1.13 (t, *J* = 7.2 Hz, 6H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 140.42, 132.24, 129.00, 126.97, 42.03, 14.12.

1-tosylpiperidine (3m):[2] Yield: 75%, 179 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (d, *J* = 8.3 Hz, 2H), 7.32 (d, *J* = 8.0 Hz, 2H), 3.10 – 2.80 (m, 4H), 2.43 (s, 3H), 1.64 (dt, *J* = 11.3, 5.9 Hz, 4H), 1.47 – 1.35 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.28, 133.34, 129.54, 127.73, 46.94, 25.18, 23.54, 21.51.

1-tosylpyrrolidine (3n):[2] Yield: 71%, 159 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (d, *J* = 8.2 Hz, 2H), 7.32 (d, *J* = 8.0 Hz, 2H), 3.23 (t, *J* = 6.8 Hz, 4H), 2.43 (s, 3H), 1.81 – 1.68 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.28, 133.98, 129.71, 127.58, 47.91, 25.02, 21.52.

4-tosylmorpholine (3o):[1] Yield: 73%, 175mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (d, *J* = 8.3 Hz, 2H), 7.35 (d, *J* = 8.0 Hz, 2H), 3.82 – 3.63 (m, 4H), 3.06 – 2.93 (m, 4H), 2.45 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.37, 133.34, 129.54, 127.84, 66.43, 46.94, 21.59.

2-(1-tosylpiperidin-4-yl)-1H-benzo[d]imidazole (3p): Yield: 62%, 220 mg, gummy mass; ^{1}H NMR (400 MHz, DMSO) δ 7.72 (d, *J* = 8.2 Hz, 2H), 7.52 (dd, *J* = 8.9, 3.4 Hz, 4H), 7.18 (dd, *J* = 6.0, 3.2 Hz, 2H), 3.73 (d, *J* = 11.8 Hz, 2H), 2.94 (dd, *J* = 13.1, 9.4 Hz, 1H), 2.55 – 2.48 (m, 2H), 2.47 (s, 3H), 2.18 (dd, *J* = 23.5, 10.1 Hz, 2H), 1.92 (dd, *J* = 18.1, 5.1 Hz, 2H). ^{13}C NMR (101 MHz, DMSO) δ 157.19, 143.98, 138.66 (d, *J* = 13.3 Hz), 133.14, 130.34, 127.97, 121.96, 115.01 (d, *J* = 14.4 Hz), 46.03, 34.87, 29.93, 21.49. HRMS (ESI) calcd for $C_{20}H_{21}N_3O_2S$ $[M+H]^+$ 356.1427; found 356.1433.

1-(2-methoxyphenyl)-4-tosylpiperazine (3q): Yield: 64%, 221 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.67 (d, *J* = 8.2 Hz, 2H), 7.34 (d, *J* = 8.0 Hz, 2H), 7.02 (ddd, *J* = 8.1, 5.9, 3.3 Hz,

1H), 6.94 – 6.87 (m, 2H), 6.84 (d, *J* = 8.0 Hz, 1H), 3.81 (s, 3H), 3.13 (dd, *J* = 15.4, 10.5 Hz, 8H), 2.44 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 152.09, 143.75, 140.33, 132.32, 129.69, 127.98, 123.67, 121.06, 118.48, 111.13 (s), 55.35, 50.02, 46.35, 21.51. HRMS (ESI) calcd for $C_{19}H_{22}N_2O_3S$ $[M+H]^+$ 347.1424; found 356.1434.

1-tosyl-1H-imidazole (3r):[3] Yield: 70%, 155 mg**;** ^{1}H NMR (400 MHz, DMSO) δ 9.10 (s, 1H), 7.69 (s, 2H), 7.51 (d, *J* = 8.1 Hz, 2H), 7.13 (d, *J* = 7.9 Hz, 2H), 2.29 (s, 3H). ^{13}C NMR (101 MHz, DMSO) δ 145.78, 138.31, 134.86, 128.62, 125.96, 119.79, 21.88 (d, *J* = 128.3 Hz).

N-benzyl-4-methylbenzenesulfonamide (3s):[2] Yield: 73%, 190 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.79 – 7.73 (m, 2H), 7.34 – 7.25 (m, 5H), 7.19 (dd, *J* = 7.6, 1.9 Hz, 2H), 4.65 (s, 1H), 4.12 (d, *J* = 6.2 Hz, 2H), 2.44 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.56, 136.88, 136.26, 129.77, 128.73, 127.92 (d, *J* = 7.1 Hz), 127.21, 47.31. 21.37.

N-(2-fluorobenzyl)-4-methylbenzenesulfonamide (3t):[6] Yield: 84%, 166 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (d, *J* = 8.3 Hz, 2H), 7.22 (dd, *J* = 19.5, 8.0 Hz, 4H), 7.03 (t, *J* = 7.5 Hz, 1H), 6.99 – 6.83 (m, 1H), 4.99 (s, 1H), 4.20 (s, 2H), 2.40 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 161.93, 159.48, 143.48, 136.89, 130.13 (d, *J* = 4.0 Hz), 129.81 – 129.53 (m), 127.11, 124.30 (d, *J* = 3.6 Hz), 123.57 (d, *J* = 14.4 Hz), 115.42, 115.21, 41.29 (d, *J* = 3.9 Hz), 21.51.

N-(4-chlorobenzyl)-4-methylbenzenesulfonamide (3u):[2] Yield: 69%, 203 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (d, *J* = 8.3 Hz, 2H), 7.29 (d, *J* = 8.0 Hz, 2H), 7.23 (d, *J* = 8.5 Hz, 2H), 7.13 (d, *J* = 8.5 Hz, 2H), 4.94 (s, 1H), 4.09 (s, 2H), 2.44 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.69, 136.82, 134.91, 133.73, 129.78, 129.23, 128.79, 127.15, 46.56, 21.55.

N-butyl-4-methylbenzenesulfonamide (3v):[2] Yield: 65%, 147 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, *J* = 8.3 Hz, 2H), 7.23 (d, *J* = 8.0 Hz, 2H), 4.72 (t, *J* = 5.7 Hz, 1H), 2.84 (dd, *J* = 13.3, 6.8 Hz, 2H), 2.35 (s, 3H), 1.41 – 1.31 (m, 2H), 1.21 (dq, *J* = 14.2, 7.3 Hz, 2H), 0.76 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 142.27, 135.98, 128.65, 126.08, 41.89, 30.53, 20.48, 18.72, 12.50.

1-((4-bromophenyl)sulfonyl)piperidine (3w):[3] Yield: 72%, 217 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.74 – 7.55 (m, 2H), 3.05 – 2.90 (m, 2H), 1.64 (t, *J* = 11.4 Hz, 2H), 1.50 – 1.36 (m, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 135.49, 132.26, 129.16, 127.59, 46.92, 25.14, 23.46.

4-((4-nitrophenyl)sulfonyl)morpholine (3x):[5] Yield: 62%, 168 mg; ^{1}H NMR (400 MHz, DMSO) δ 8.57 – 8.36 (m, 2H), 8.09 – 7.93 (m, 2H), 3.67 – 3.62 (m, 4H), 3.02 – 2.88 (m, 4H). ^{13}C NMR (101 MHz, DMSO) δ 150.68, 140.78, 129.67, 125.23, 65.76, 46.25.

4-tosylthiomorpholine (3y):[7] Yield: 68%, 174 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.62 (d, *J* = 8.3 Hz, 2H), 7.33 (d, *J* = 8.0 Hz, 2H), 3.42 – 3.25 (m, 4H), 2.76 – 2.64 (m, 4H), 2.44 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 143.78, 133.74, 129.81, 127.44, 47.86, 27.32, 21.53.

4-((4-bromophenyl)sulfonyl)thiomorpholine (3z):[7] Yield: 65%, 208 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.69 (d, *J* = 8.7 Hz, 1H), 7.60 (d, *J* = 8.7 Hz, 1H), 3.41 – 3.25 (m, 2H), 2.78 – 2.64 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 135.96, 132.54, 128.88, 128.02, 47.85, 27.31.

4-methyl-N,N-dipropylbenzenesulfonamide (3aa):[7] Yield: 56%, 142 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.75 (d, *J* = 8.3 Hz, 2H), 7.25 (dd, *J* = 8.5, 0.5 Hz, 2H), 3.40 – 3.32 (m, 2H), 3.28 – 3.24 (m, 2H), 2.40 (s, 3H), 1.67 – 1.54 (m, 4H), 0.94 – 0.85 (m, 6H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 142.24, 139.79, 129.27, 126.33, 54.26, 21.90, 19.96, 11.19.

4-((2,4-difluorophenyl)sulfonyl)morpholine (3ab): Yield: 63%, 165 mg, gummy mass; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.86 (td, *J* = 8.2, 6.2 Hz, 1H), 7.08 – 6.95 (m, 2H), 3.78 – 3.73 (m, 4H), 3.18 (dd, *J* = 4.8, 3.7 Hz, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 167.20 (d, *J* = 11.4 Hz), 164.63 (d, *J* = 11.4 Hz), 161.17 (d, *J* = 12.8 Hz), 158.60 (d, *J* = 12.8 Hz), 133.14 (dd, *J* = 10.5, 2.1 Hz), 121.22 (dd, *J* = 15.0, 4.0 Hz), 112.10 (dd, *J* = 21.8, 3.8 Hz), 106.16, 105.90, 105.64, 66.31, 45.76. HRMS (ESI) calcd for $C_{10}H_{12}F_2NO_3S$ $[M+H]^+$ 264.2663; found 264.2671.

4-((2-bromophenyl)sulfonyl)morpholine (3ac):[8] Yield: 60%, 182 mg; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (dd, *J* = 7.9, 1.7 Hz, 1H), 7.68 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.45 (td, *J* = 7.7, 1.2 Hz, 1H), 7.39 – 7.34 (m, 1H), 3.81 – 3.75 (m, 4H), 3.61 – 3.55 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 140.39, 134.96, 133.27, 130.67, 127.71, 120.28, 65.80, 44.70.

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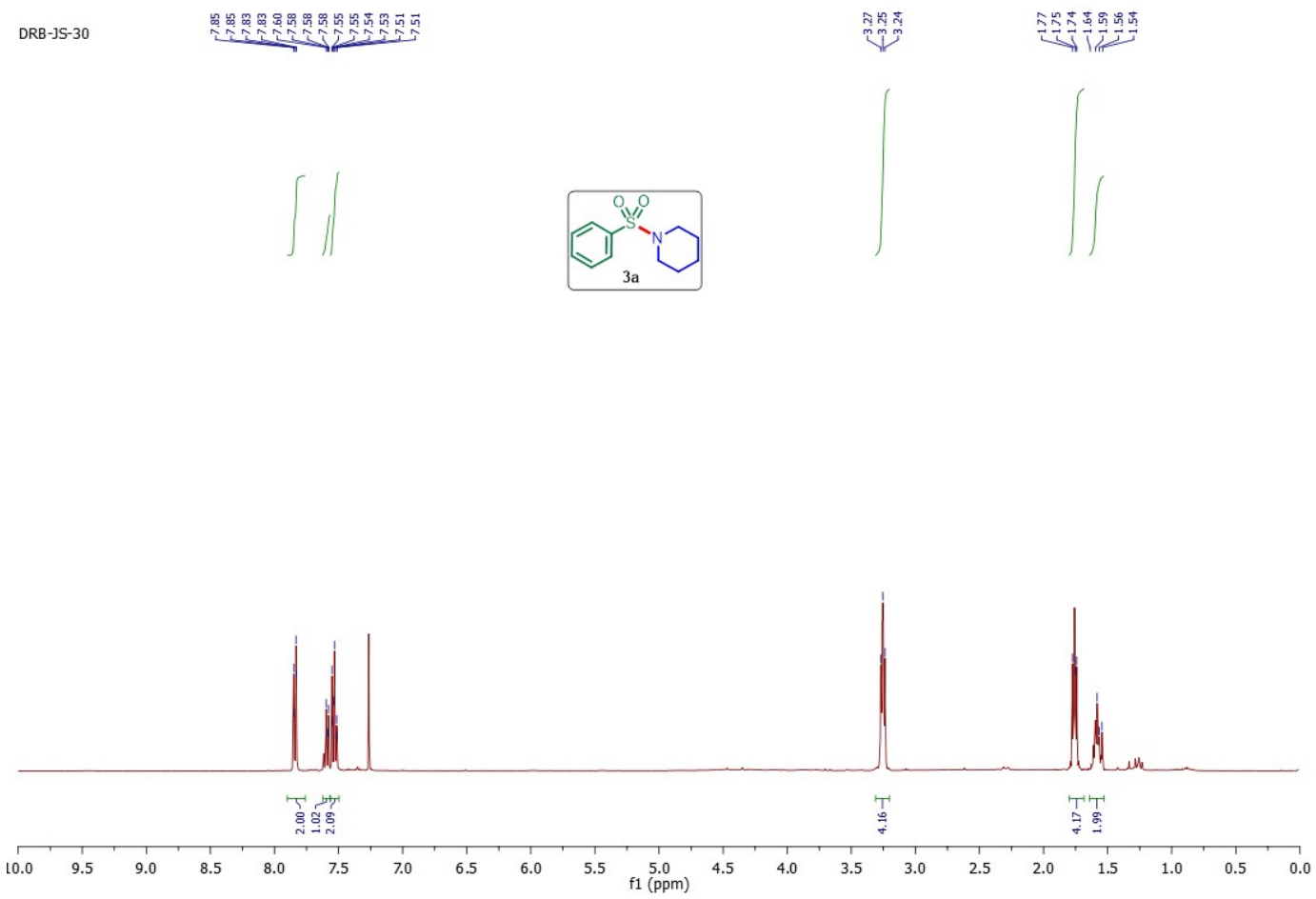
^{1}H and ^{13}C NMR Spectra of Compounds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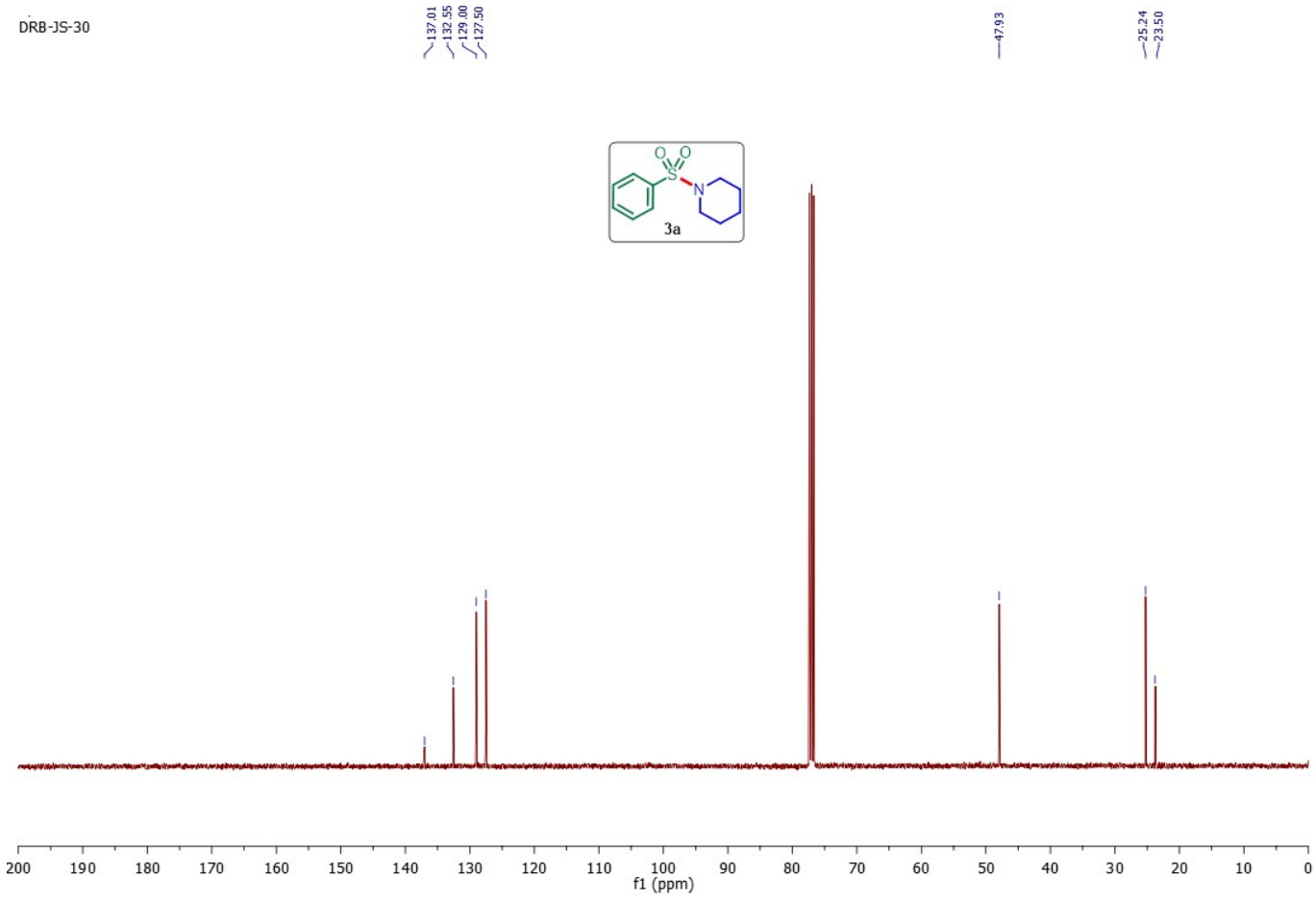
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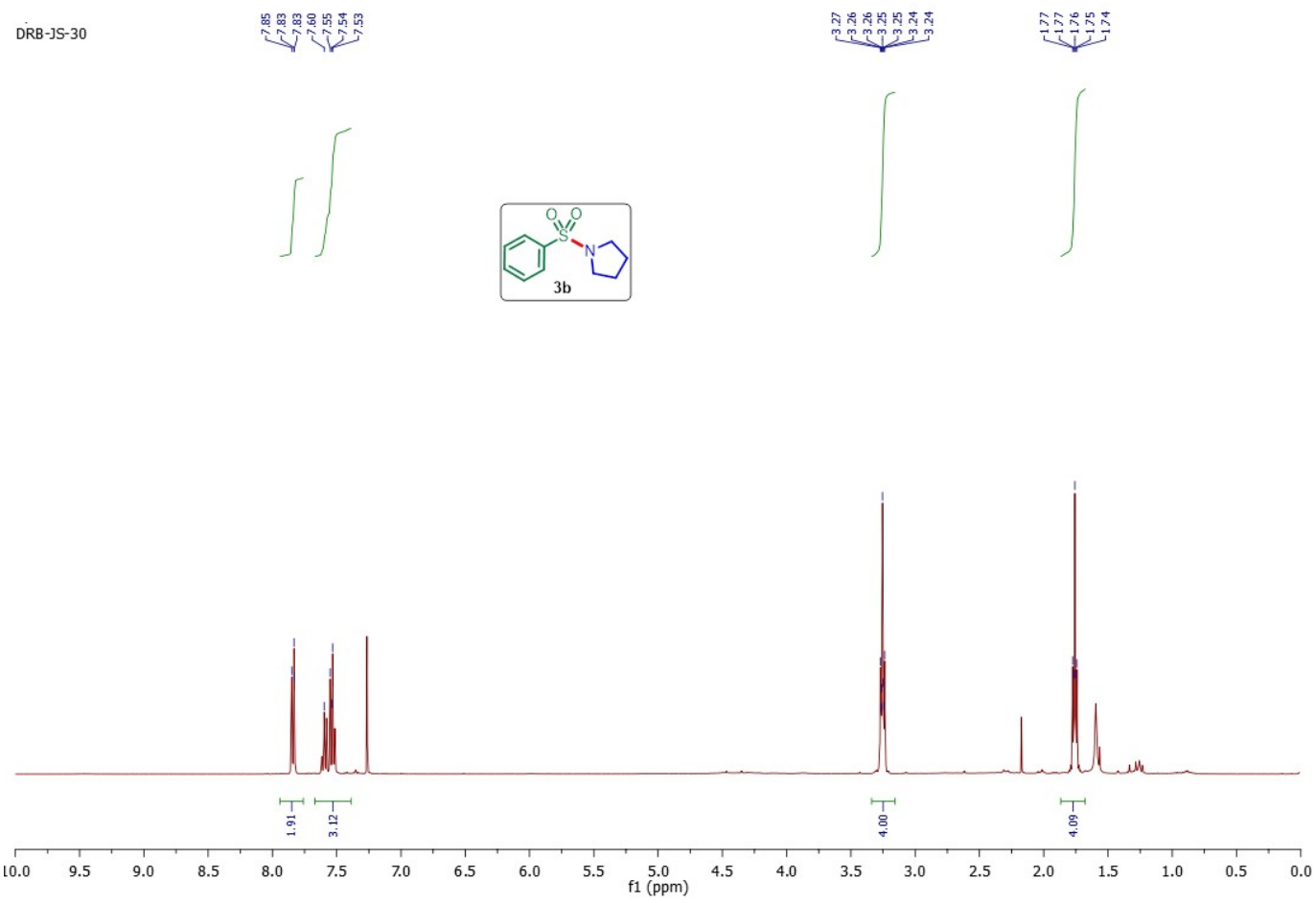
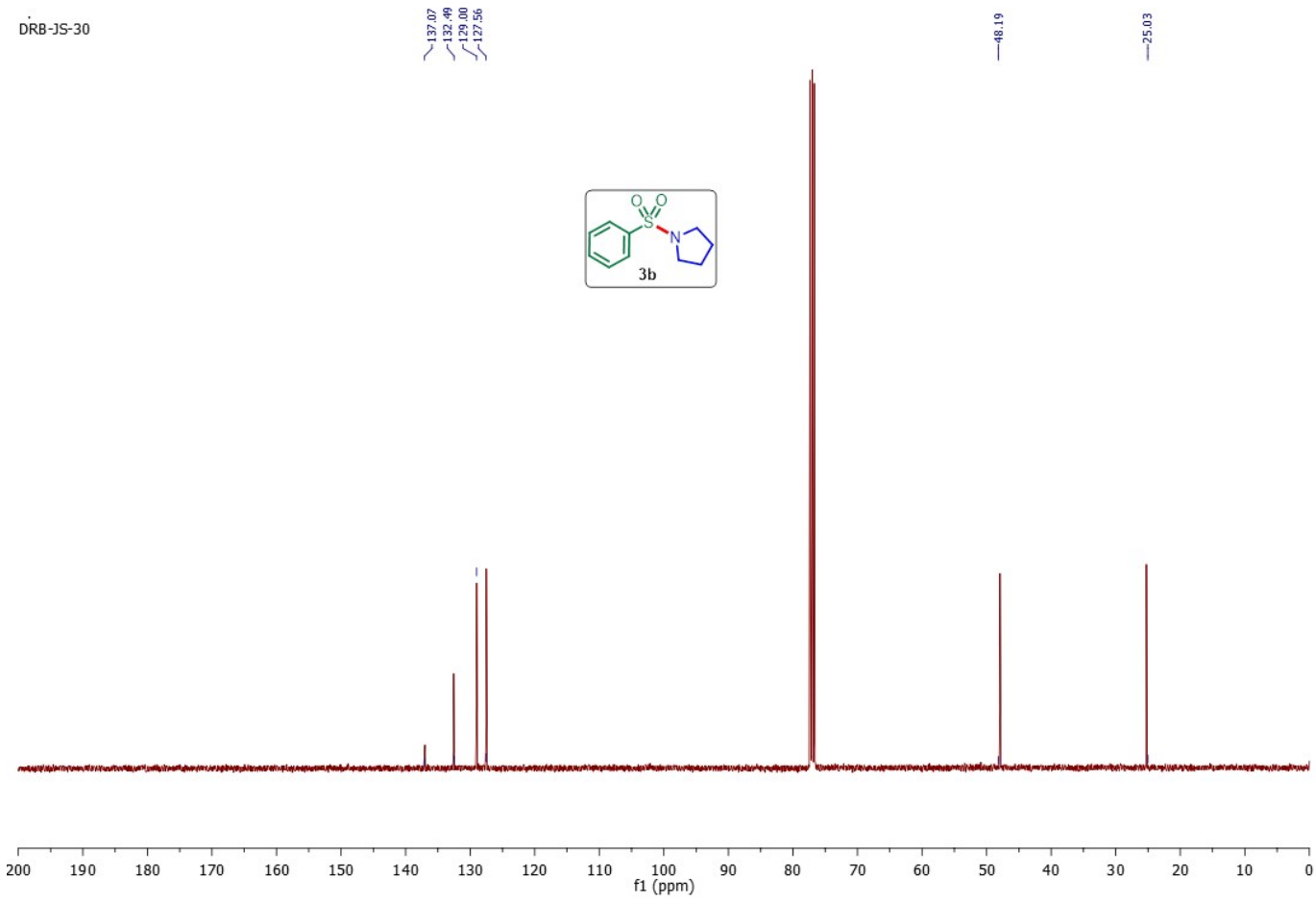
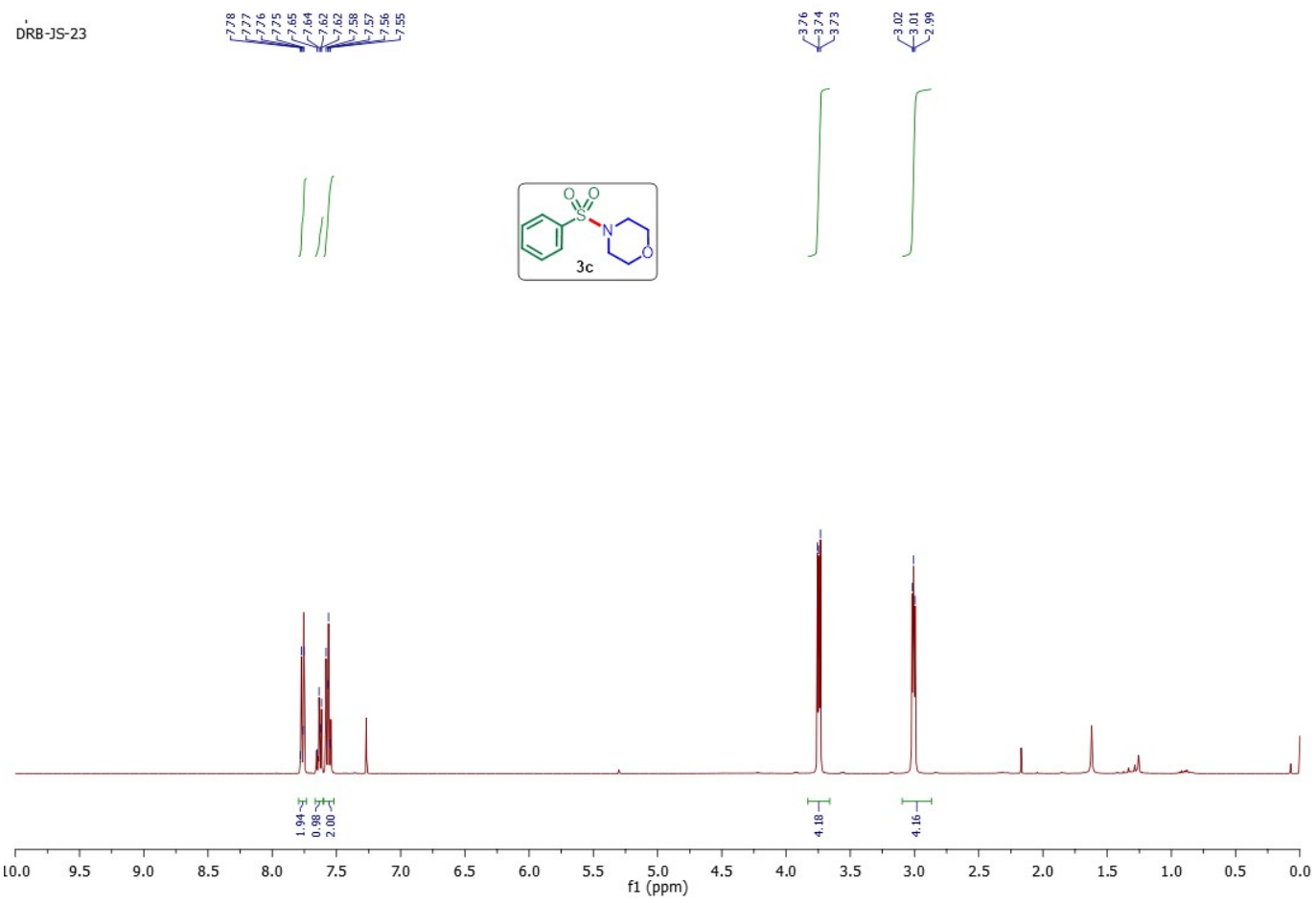
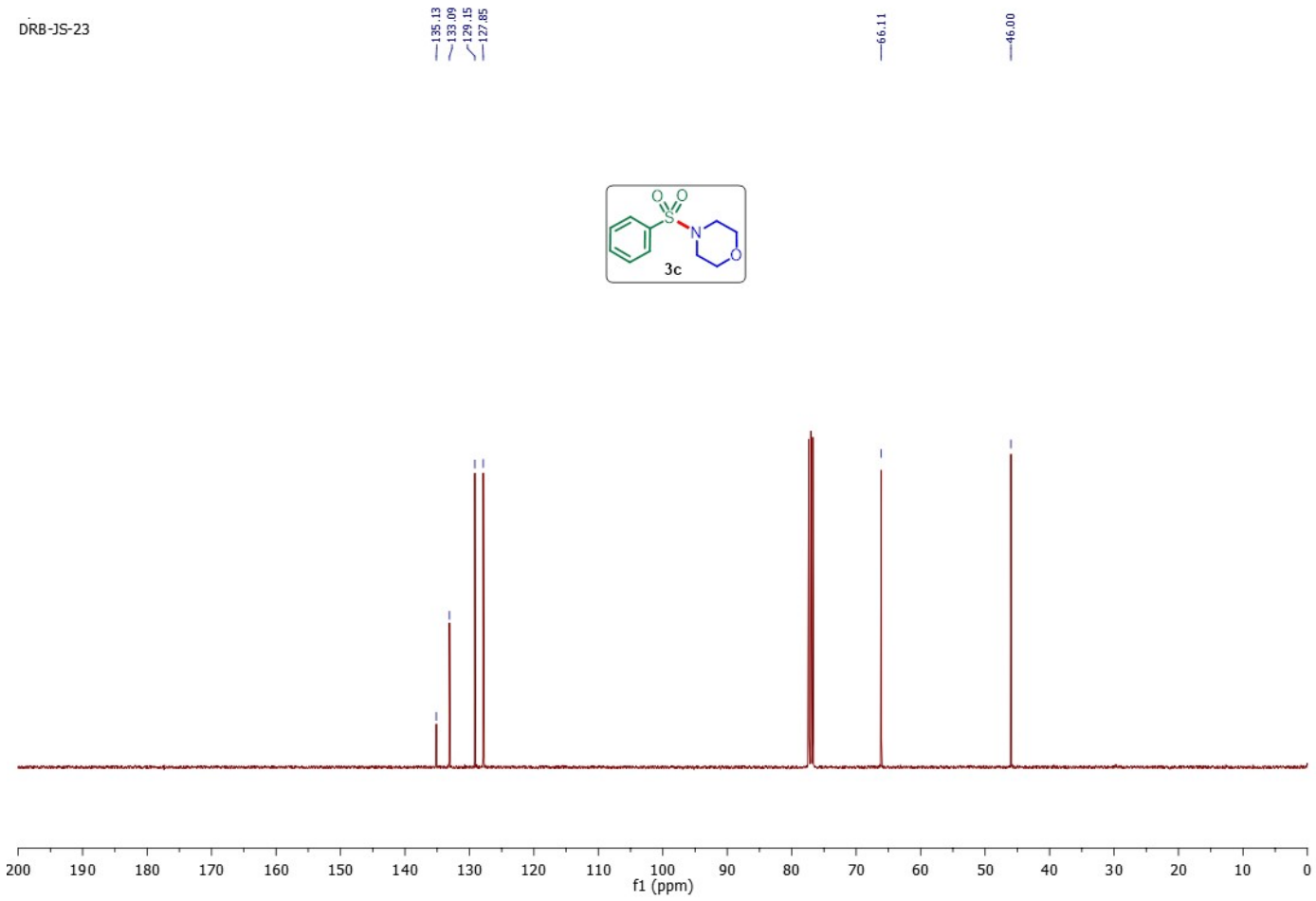
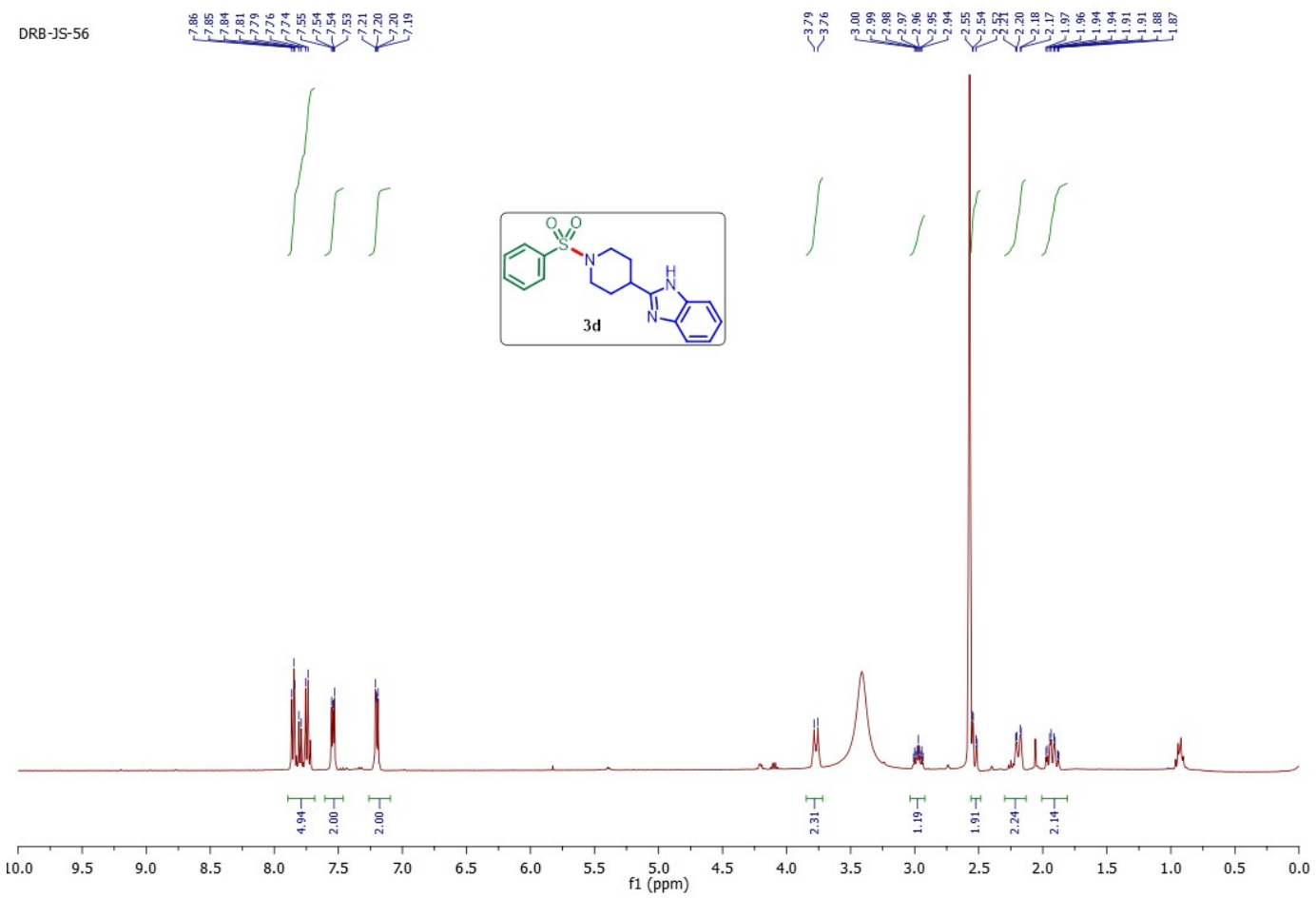
0
f1 (ppm)

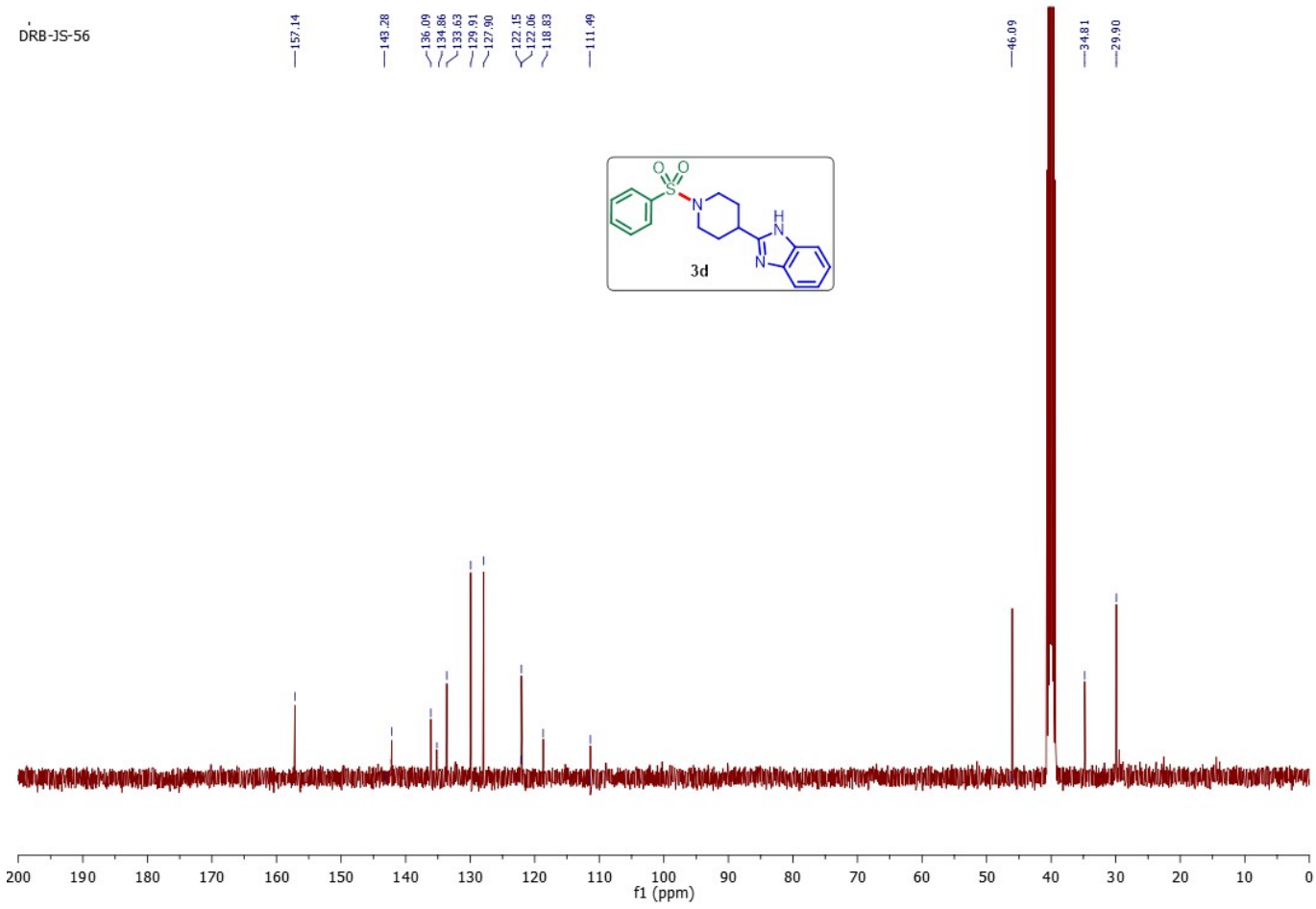

DRB-JS-30
7.85
7.83
7.83
7.60
7.55
7.54
7.53
3.27
3.26
3.26
3.25
3.25
3.24
3.24
1.77
1.77
1.76
1.75
1.74
3b
1.91
3.12
4.00
4.09
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

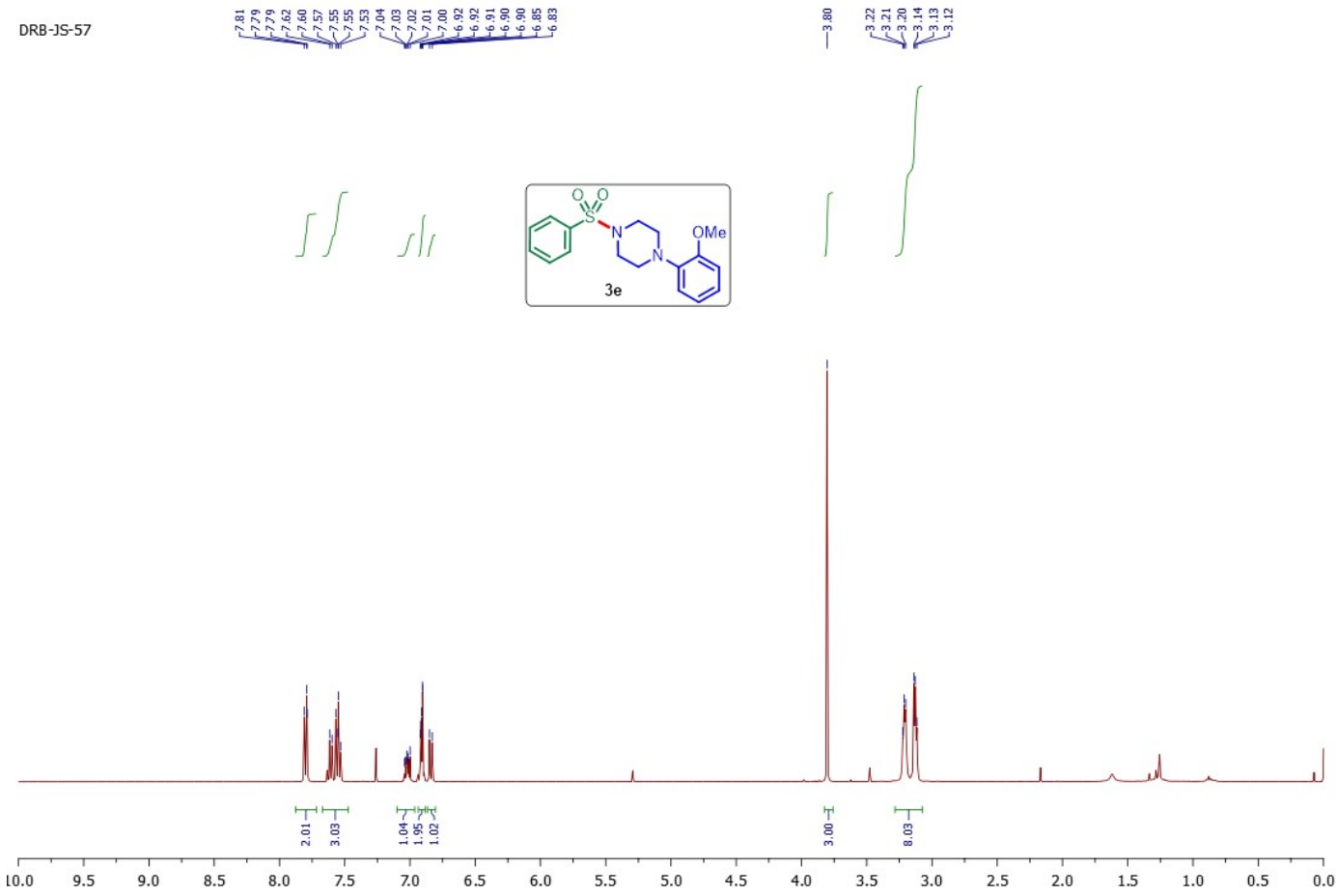

DRB-JS-30
137.07
132.49
129.00
127.56
48.19
25.03
3b
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

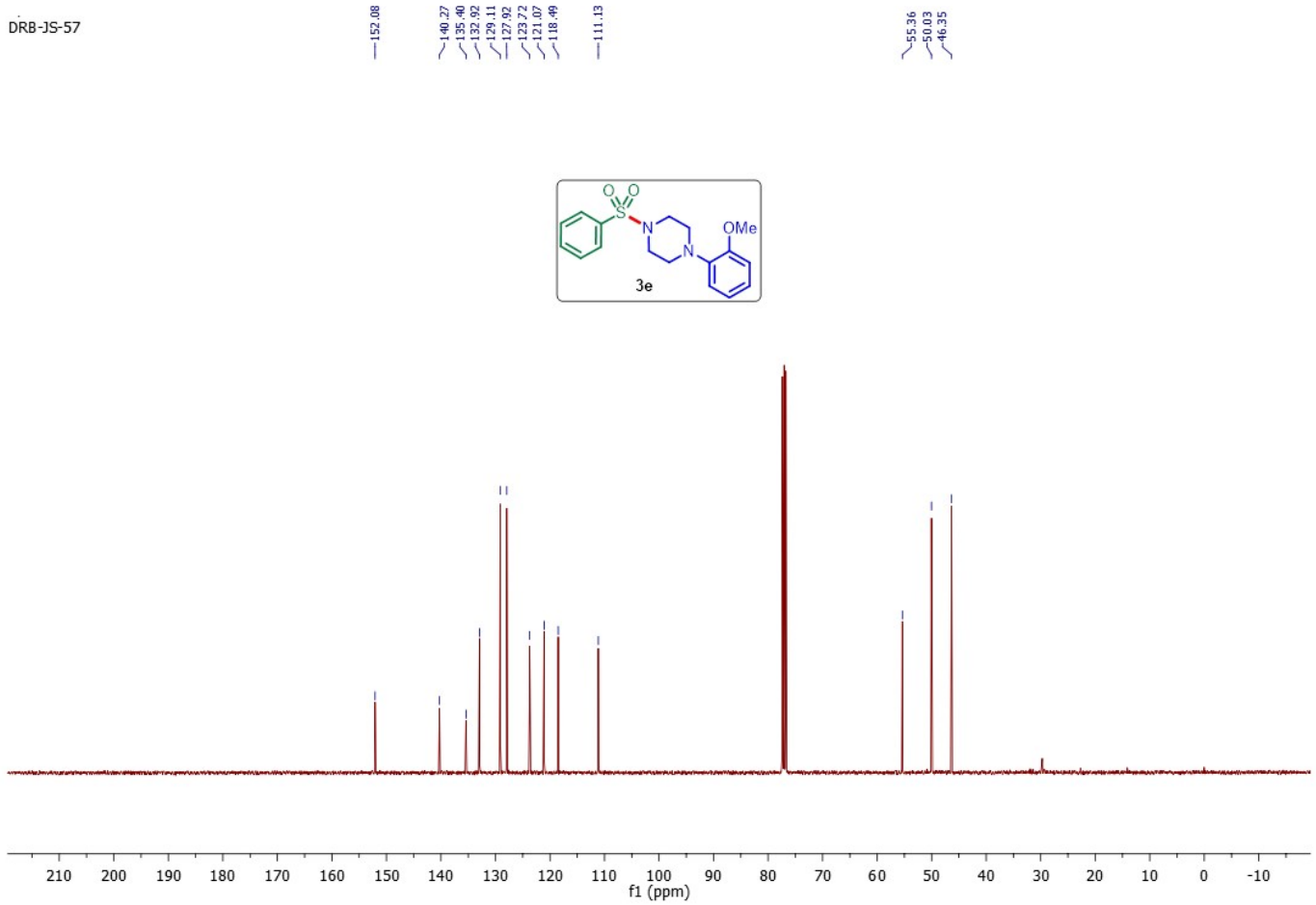

DRB-JS-23
7.78
7.77
7.76
7.75
7.65
7.64
7.62
7.62
7.58
7.57
7.56
7.55
3.76
3.74
3.73
3.02
3.01
2.99
3c
1.94
0.98
2.00
4.18
4.16
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

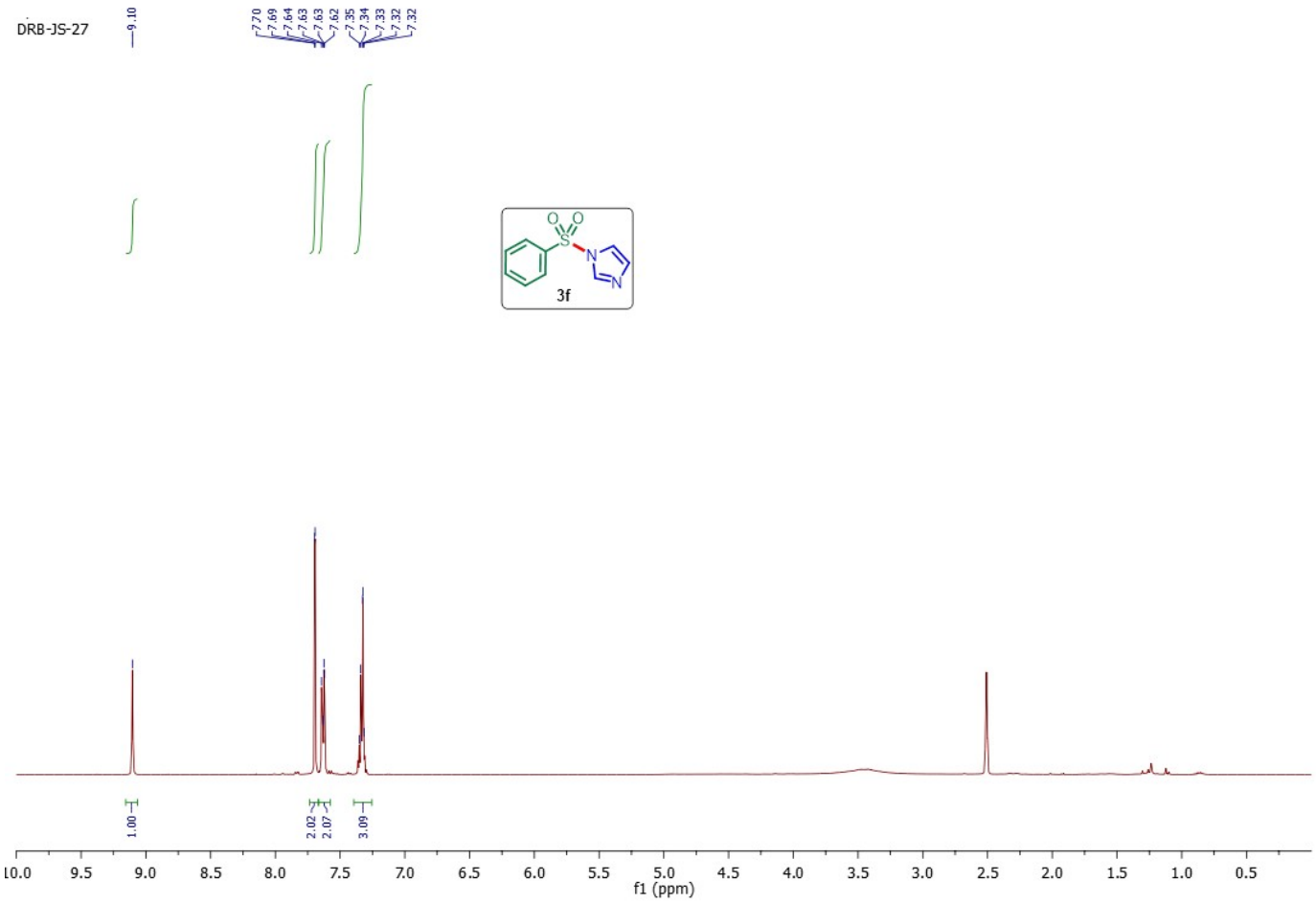

DRB-JS-23
135.13
133.09
129.15
127.85
66.11
46.00
3c
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


DRB-JS-56
7.86
7.85
7.84
7.81
7.79
7.76
7.74
7.55
7.54
7.54
7.53
7.21
7.20
7.20
7.19
3.79
3.76
3.00
2.99
2.98
2.97
2.96
2.95
2.94
2.55
2.54
2.52
2.21
2.20
2.18
2.17
1.97
1.96
1.94
1.94
1.91
1.91
1.88
1.87
3d
4.94
2.00
2.00
2.31
1.19
1.91
2.24
2.14
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

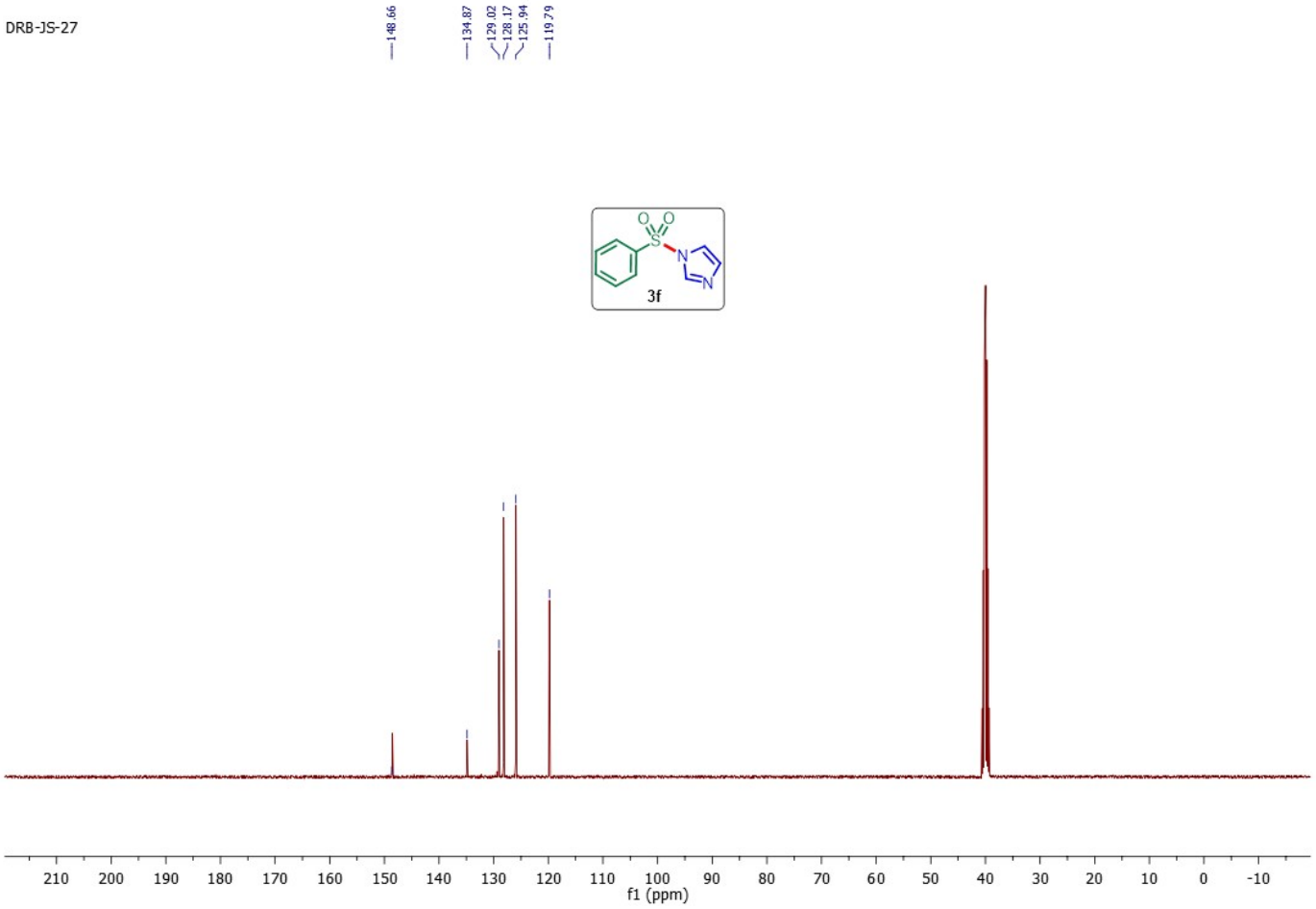

DRB-JS-56
157.14
143.28
136.09
134.86
133.63
129.91
127.90
122.15
122.06
118.83
111.49
46.09
34.81
29.90
3d
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

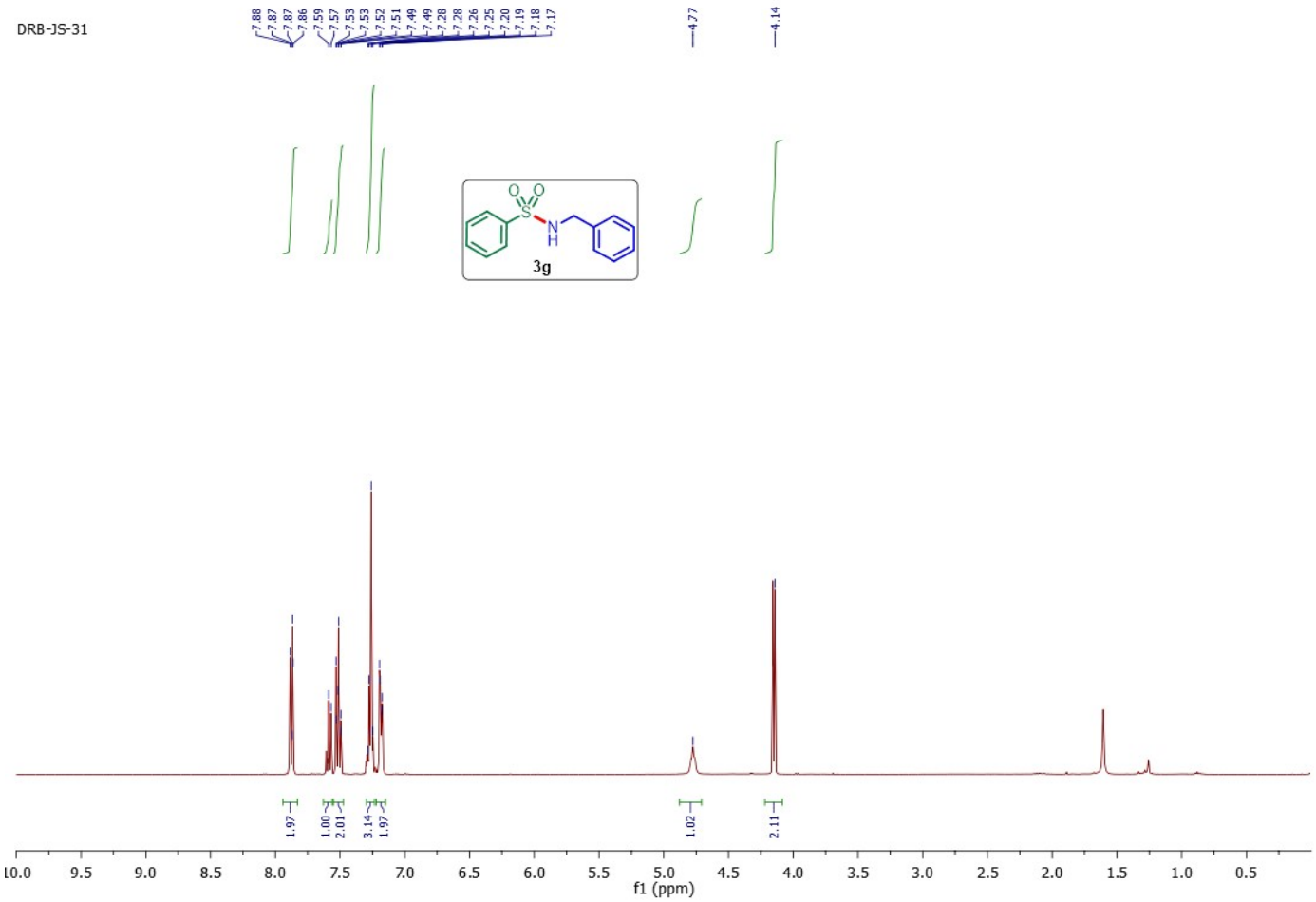

DRB-JS-57
7.81
7.79
7.79
7.62
7.60
7.57
7.55
7.55
7.53
7.04
7.03
7.02
7.01
7.00
6.92
6.92
6.91
6.90
6.90
6.85
6.83
3.80
3.22
3.21
3.20
3.14
3.13
3.12
OMe
3e
2.01
3.03
1.04
1.95
1.02
3.00
8.03
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0

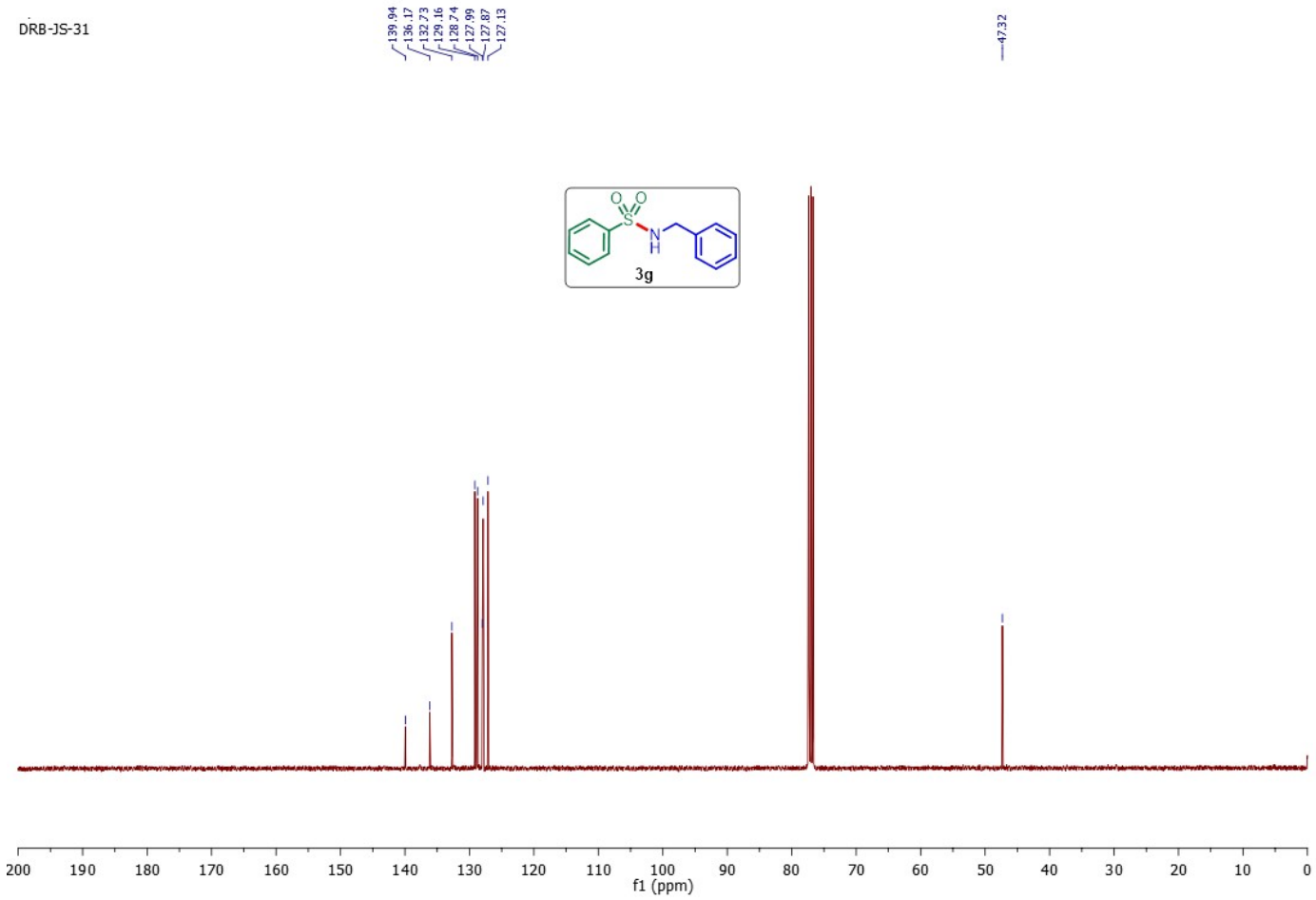

DRB-JS-57
152.08
140.27
135.40
132.92
129.11
127.92
123.72
121.07
118.49
111.13
55.36
50.03
46.35
OMe
3e
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

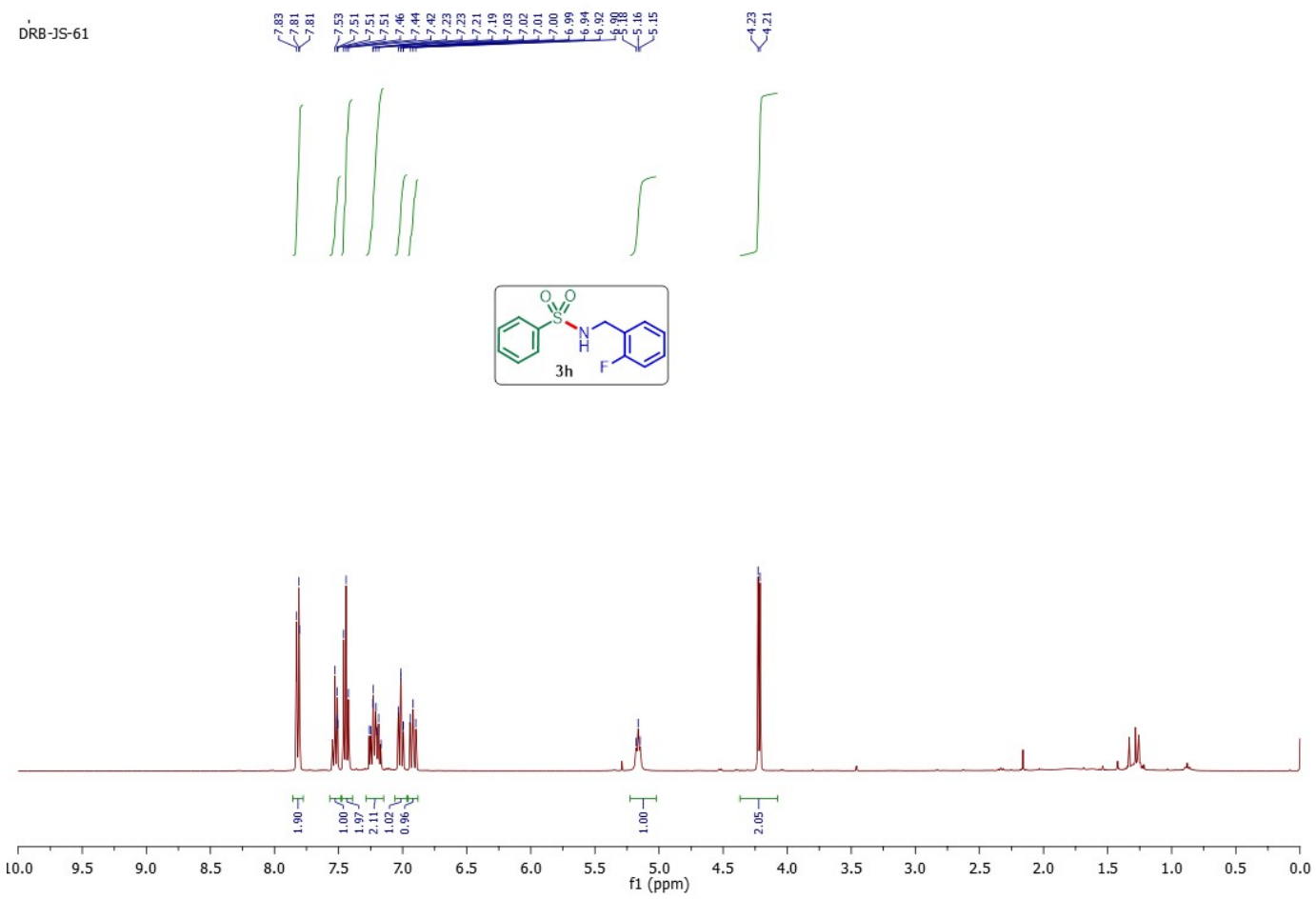

DRB-JS-27
9.10
7.70
7.69
7.64
7.63
7.63
7.62
7.35
7.34
7.33
7.32
7.32
3f
1.00
2.02
2.07
3.09
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

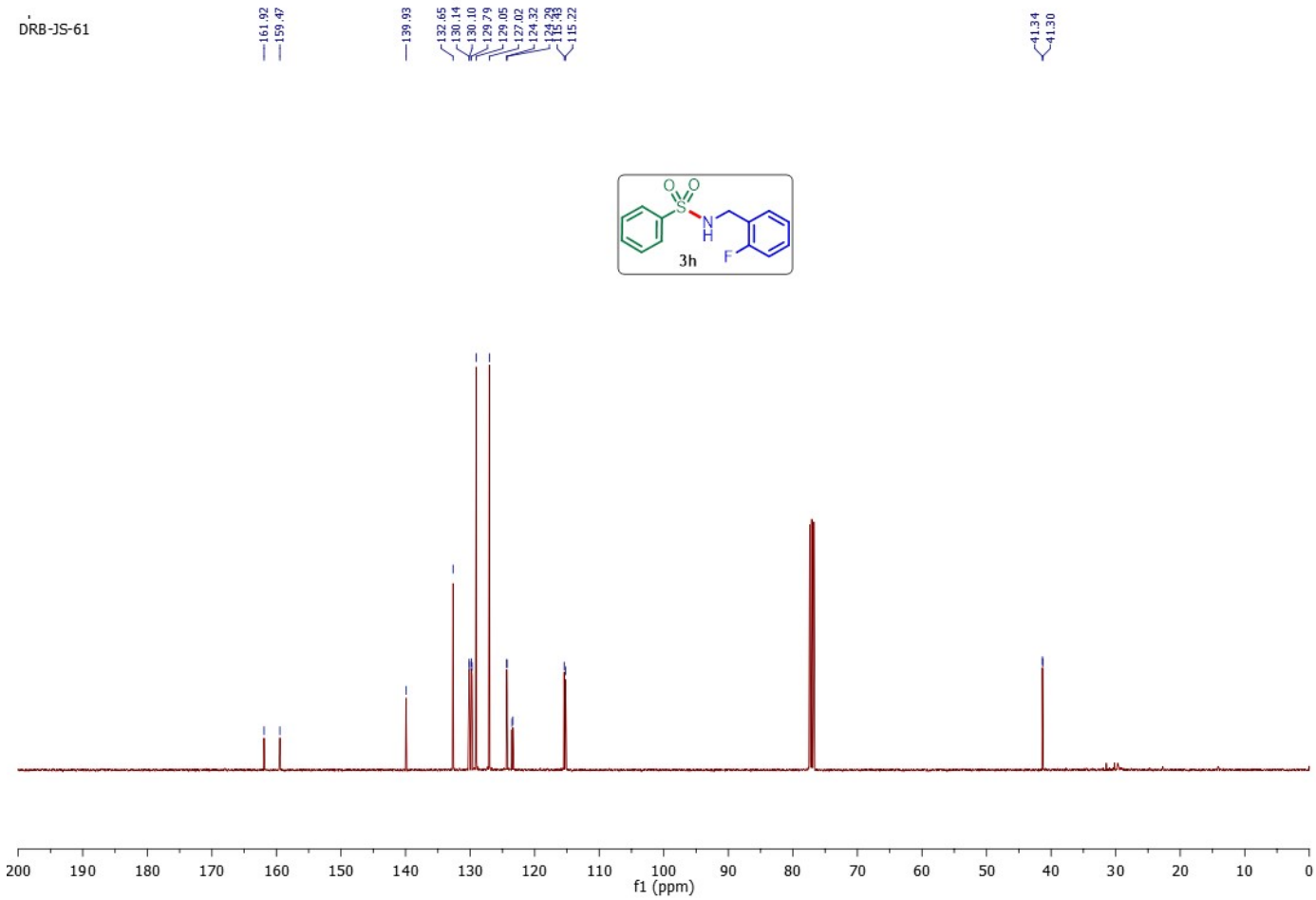
DRB-JS-27

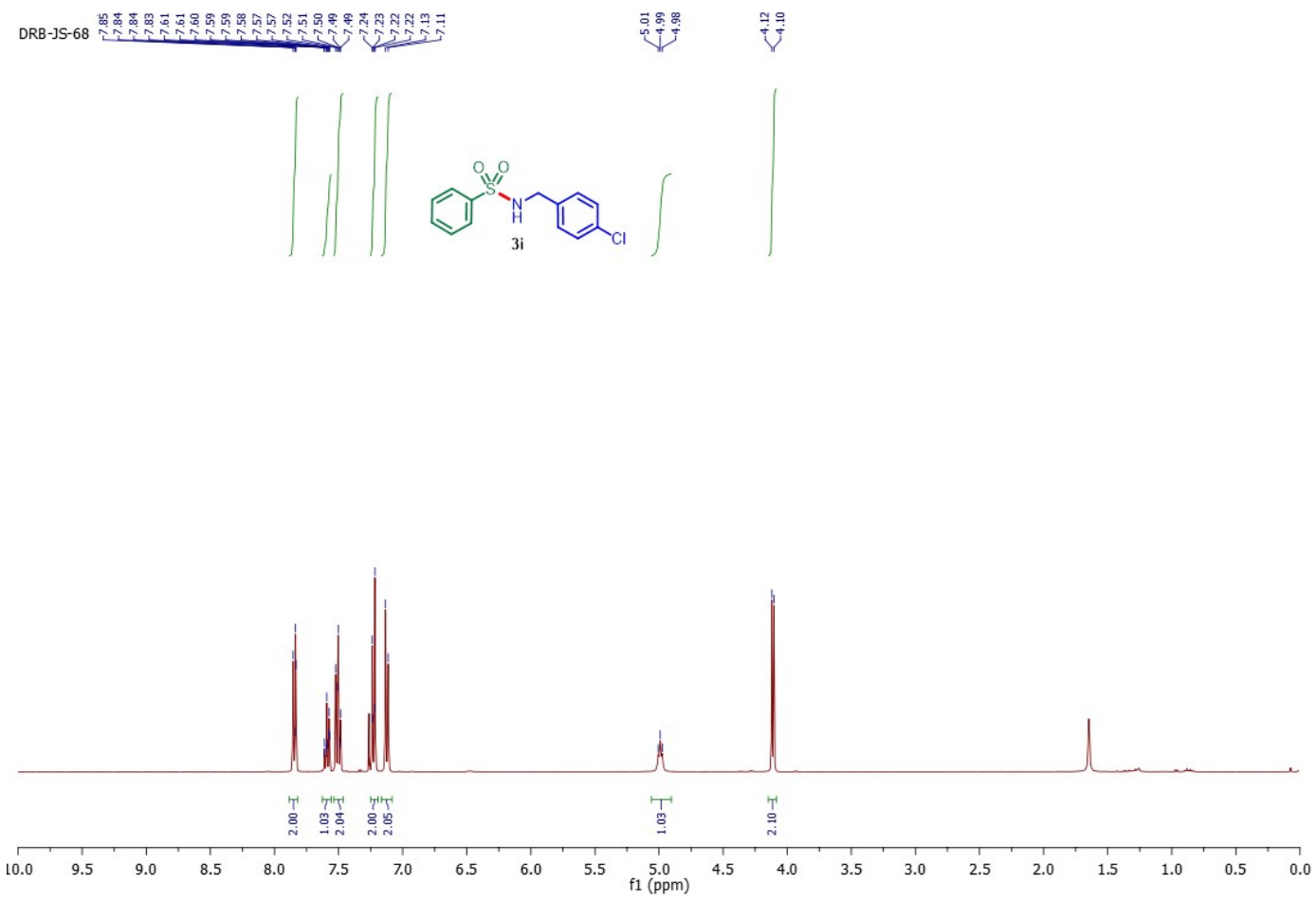

148.66
134.87
129.02
128.17
125.94
119.79
3f
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

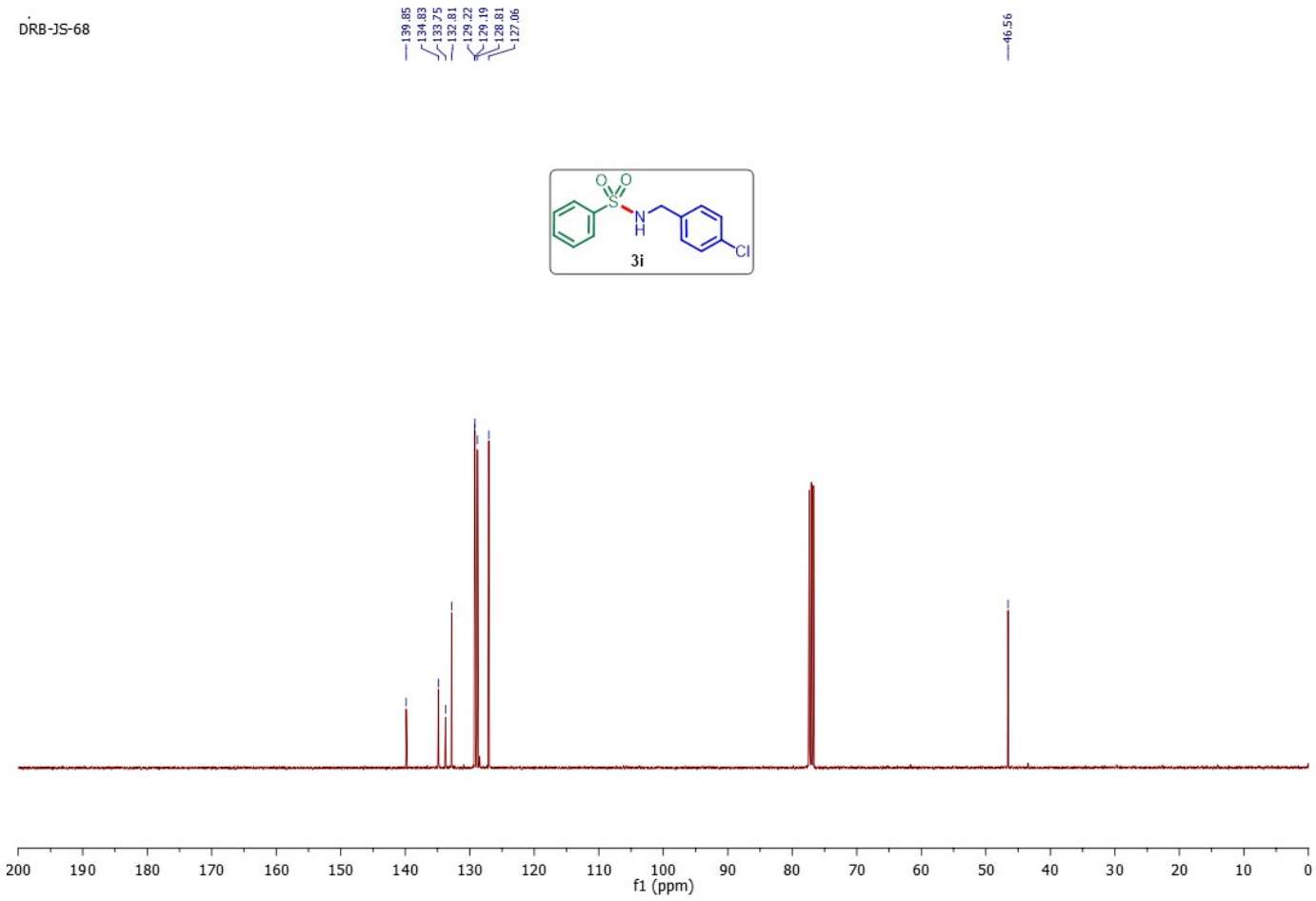

DRB-JS-31
7.88
7.87
7.87
7.86
7.59
7.57
7.53
7.53
7.52
7.51
7.49
7.49
7.28
7.28
7.26
7.25
7.20
7.19
7.18
7.17
4.77
4.14
3g
1.97
1.00
2.01
3.14
1.97
1.02
2.11
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

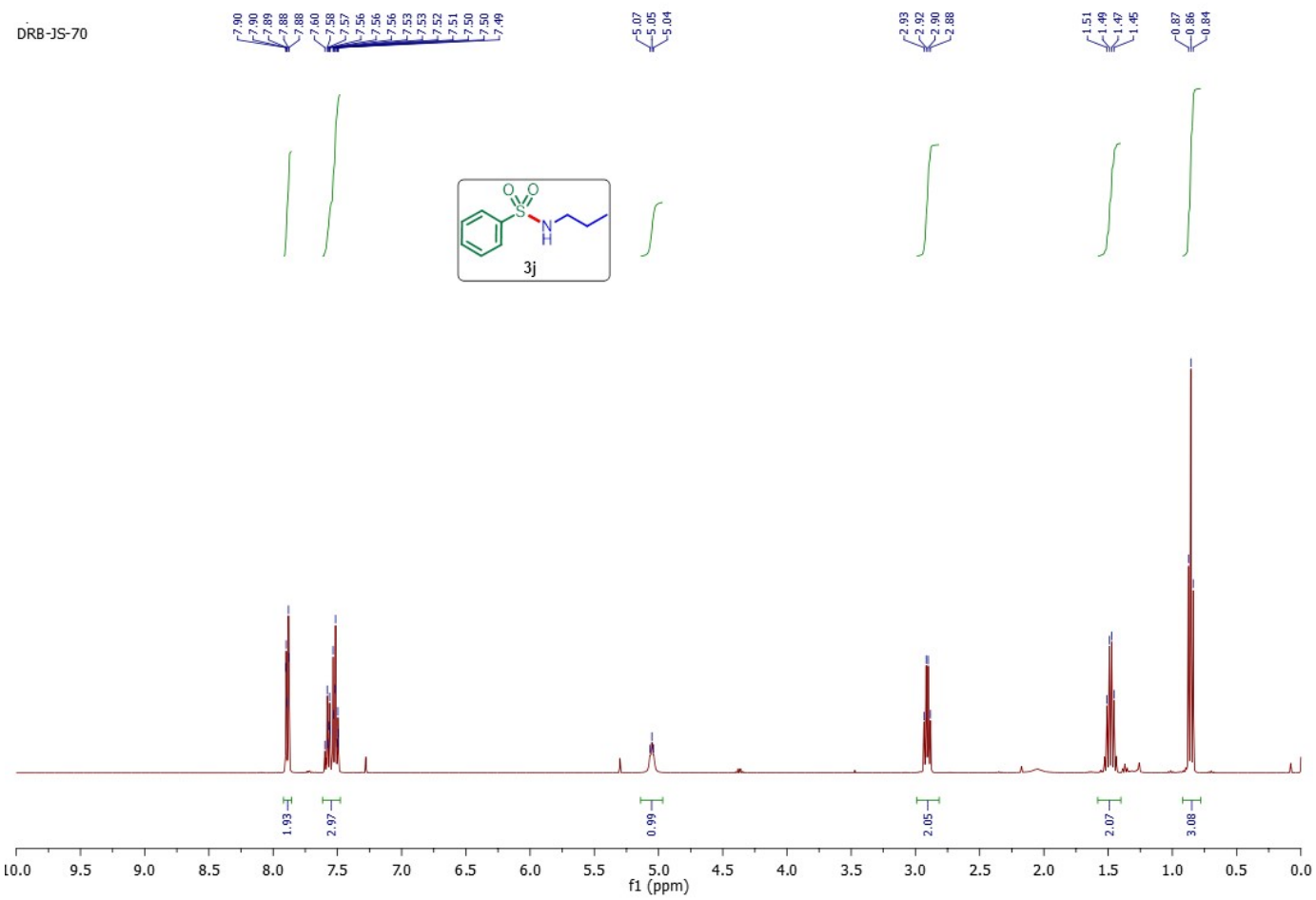

DRB-JS-31
139.94
136.17
132.73
129.16
128.74
127.99
127.87
127.13
47.32
3g
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

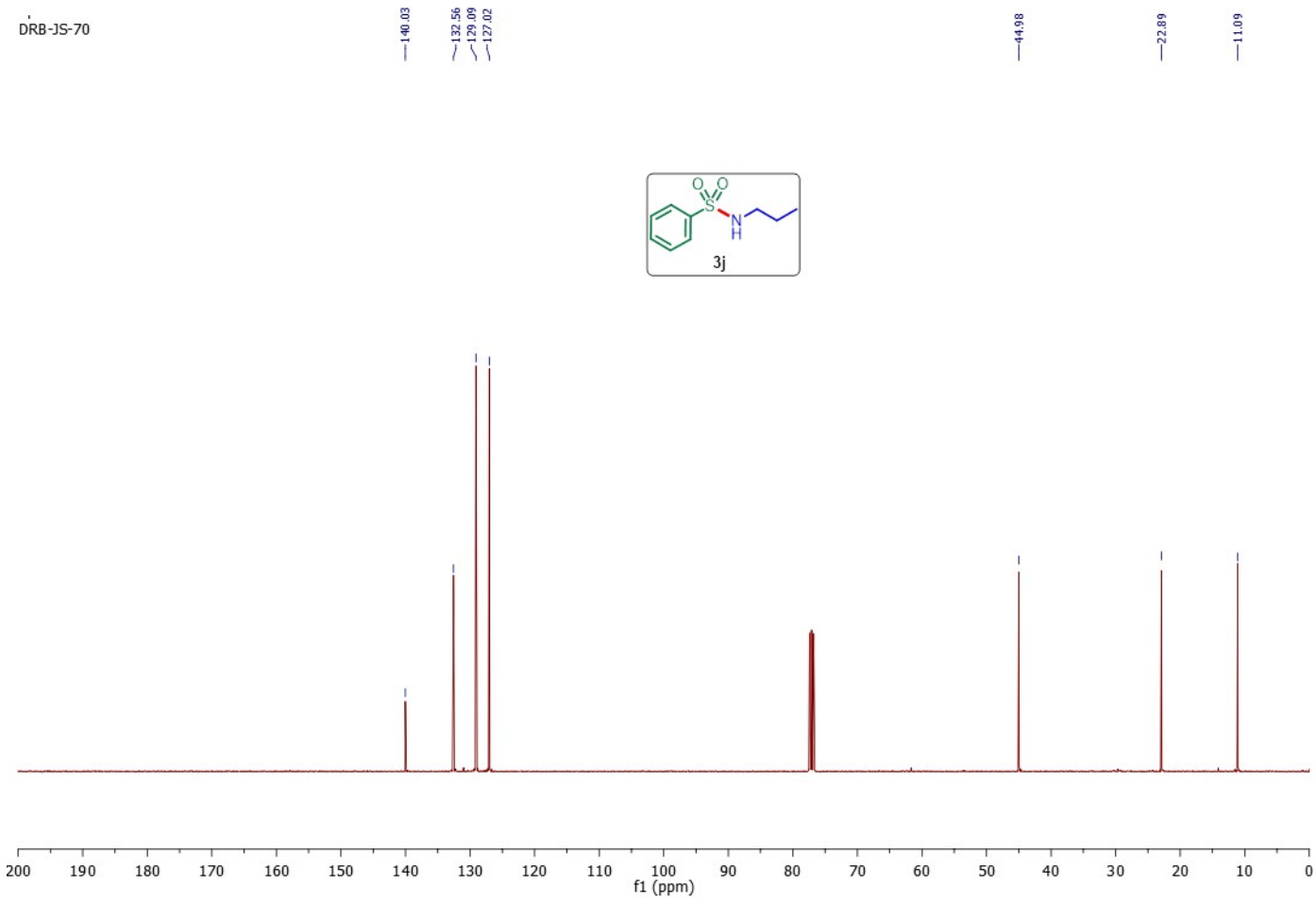

DRB-JS-61
7.83
7.81
7.81
7.53
7.51
7.51
7.51
7.46
7.44
7.42
7.23
7.23
7.21
7.19
7.03
7.02
7.01
7.00
6.99
6.94
6.92
6.90
5.18
5.16
5.15
4.23
4.21
3h
1.90
1.00
1.97
2.11
1.02
0.96
1.00
2.05
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

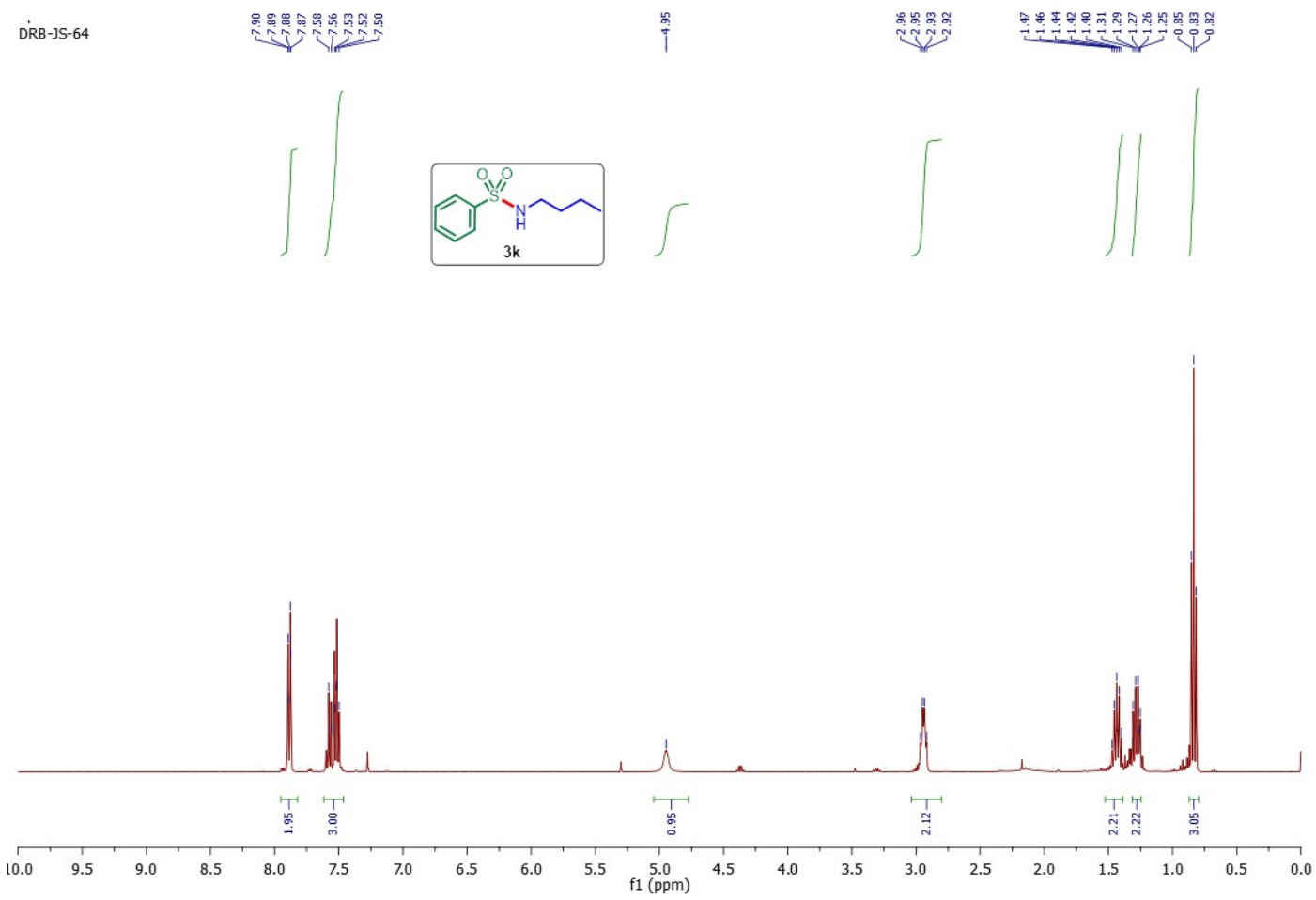

DRB-JS-61
161.92
159.47
139.93
132.65
130.14
130.10
129.79
129.05
127.02
124.32
115.43
115.22
41.34
41.30
3h
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

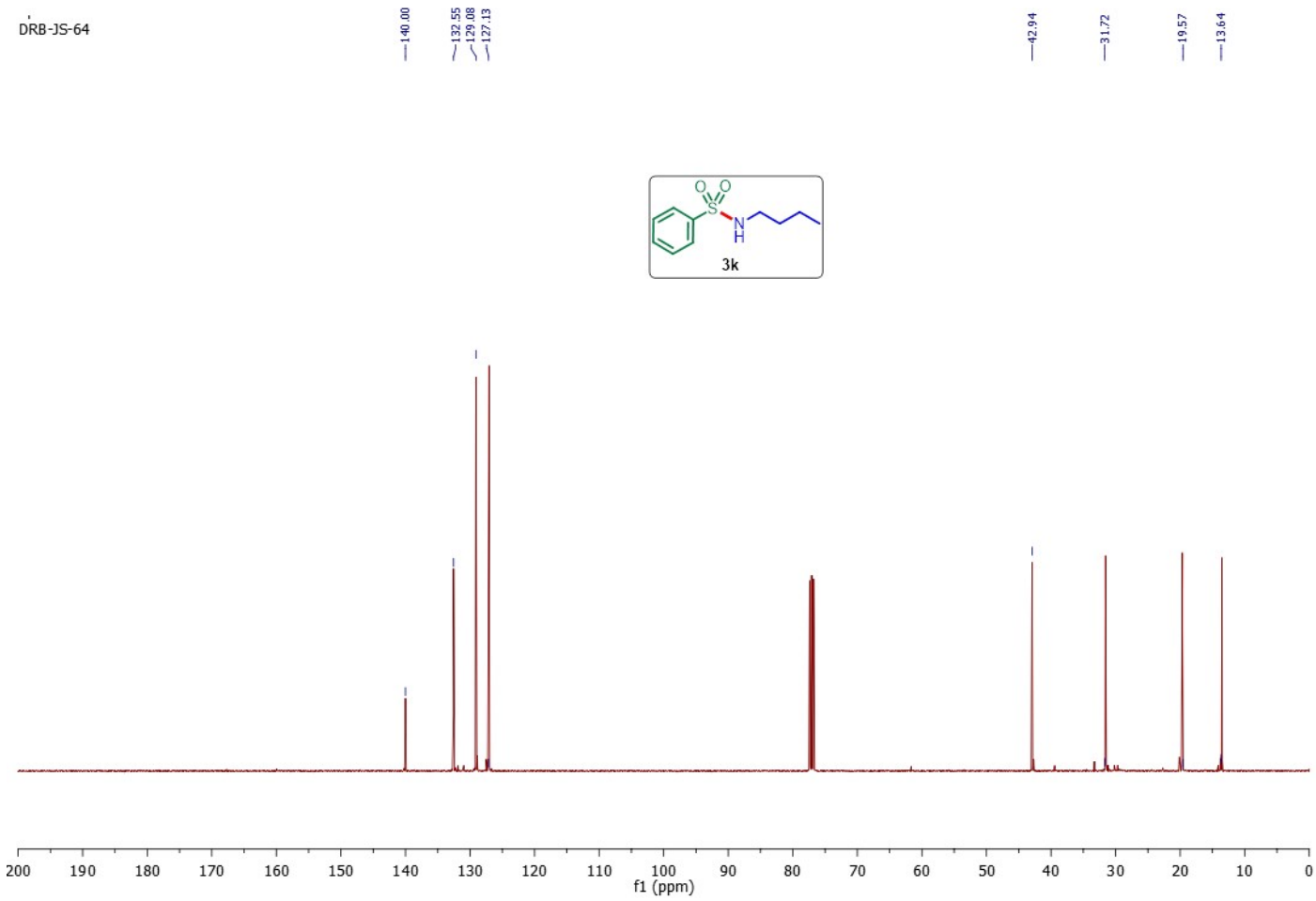

DRB-JS-68
7.85
7.84
7.84
7.83
7.61
7.61
7.60
7.59
7.59
7.58
7.57
7.57
7.52
7.51
7.50
7.49
7.49
7.24
7.23
7.22
7.22
7.13
7.11
5.01
4.99
4.98
4.12
4.10
O
O
S
N
H
Cl
3i
2.00
1.03
2.04
2.00
2.05
1.03
2.10
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

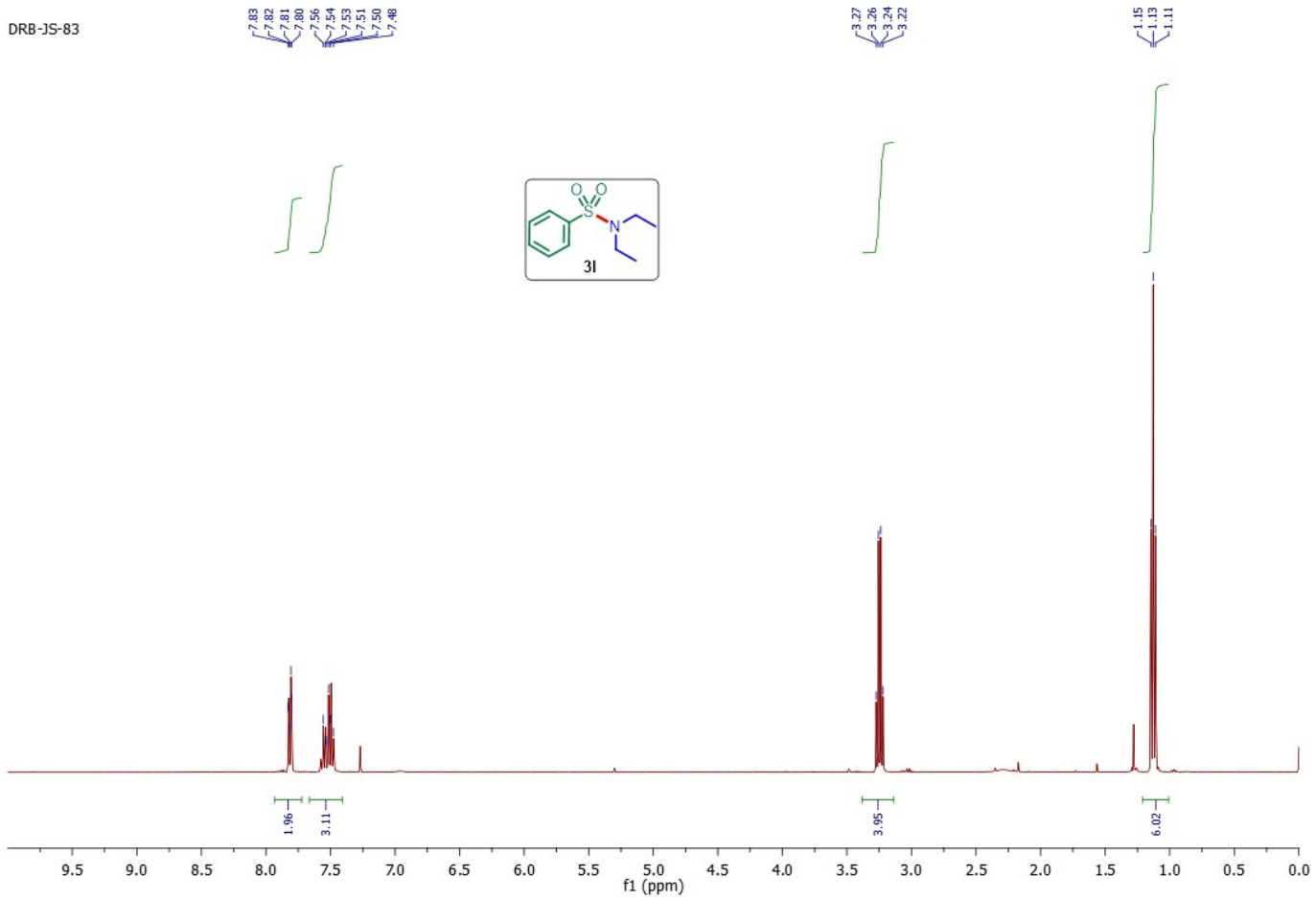

DRB-JS-68
139.85
134.83
133.75
132.81
129.22
129.19
128.81
127.06
46.56
3i
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


DRB-JS-70
7.90
7.90
7.89
7.88
7.88
7.60
7.58
7.57
7.56
7.56
7.56
7.53
7.53
7.52
7.51
7.50
7.50
7.49
5.07
5.05
5.04
2.93
2.92
2.90
2.88
1.51
1.49
1.47
1.45
0.87
0.86
0.84
3j
1.93
2.97
0.99
2.05
2.07
3.08
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

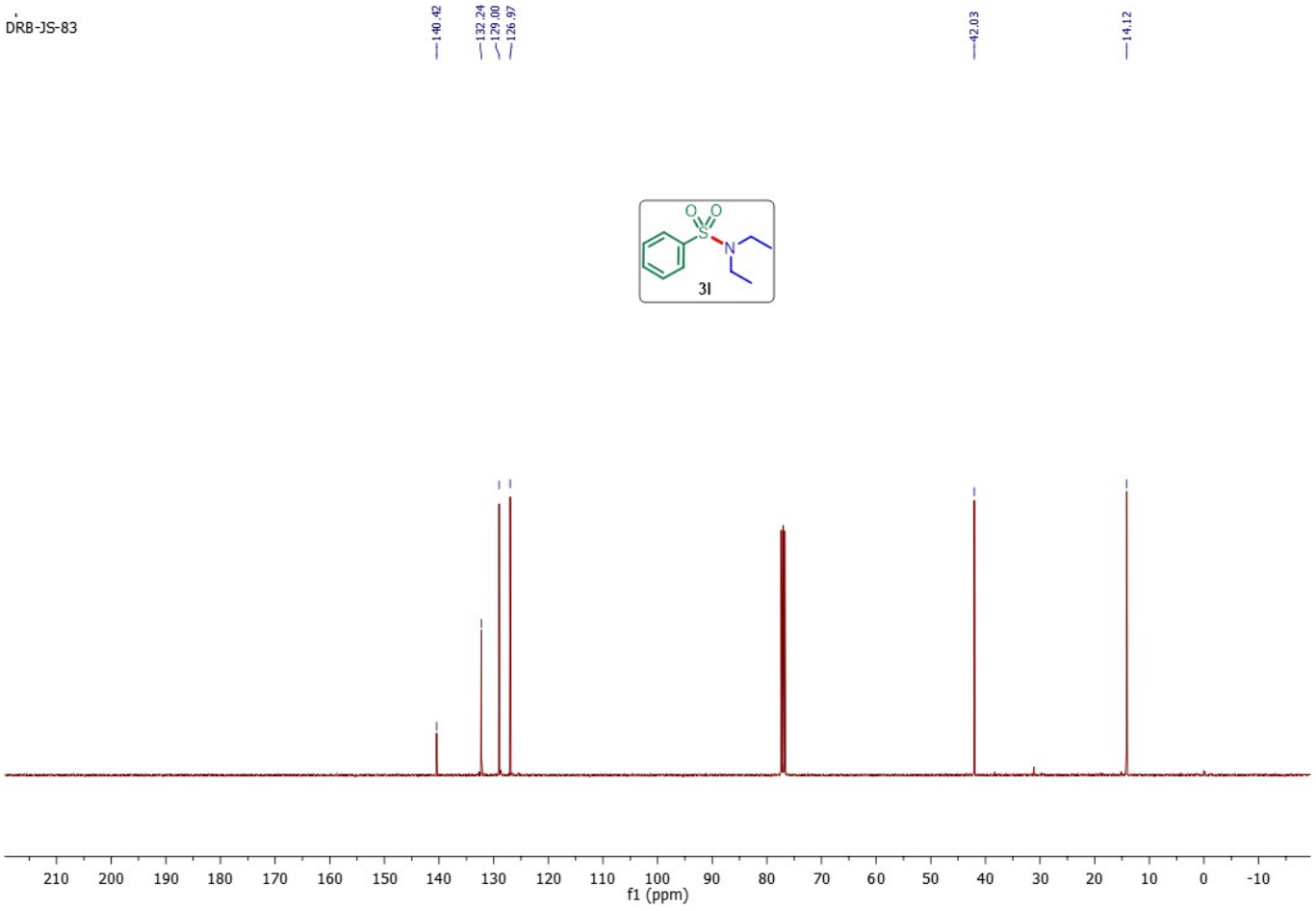

DRB-JS-70
140.03
132.56
129.09
127.02
44.98
22.89
11.09
3j
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

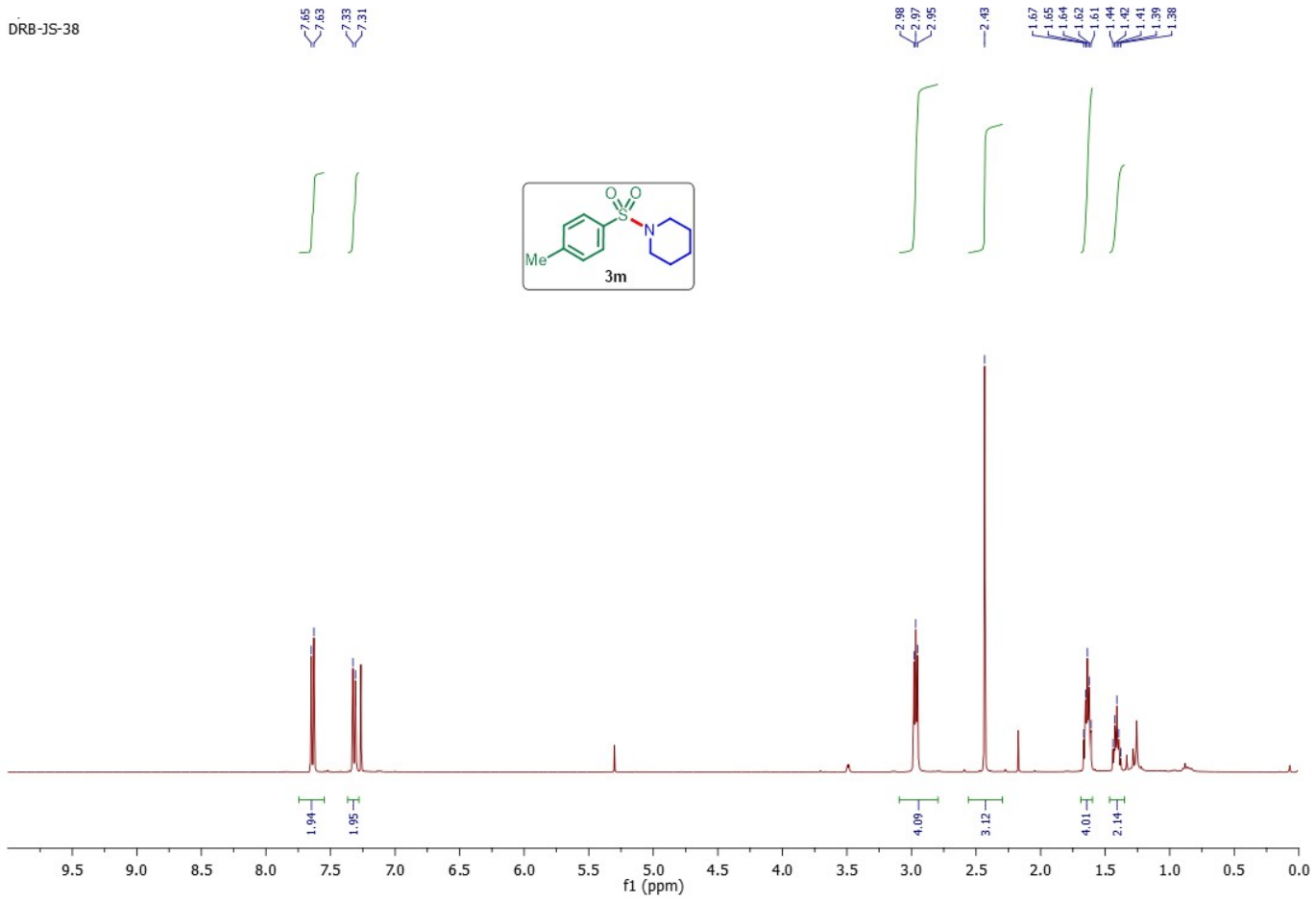

DRB-JS-64
7.90
7.89
7.88
7.87
7.58
7.56
7.53
7.52
7.50
4.95
2.96
2.95
2.93
2.92
1.47
1.46
1.44
1.42
1.40
1.31
1.29
1.27
1.26
1.25
0.85
0.83
0.82
3k
1.95
3.00
0.95
2.12
2.21
2.22
3.05
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

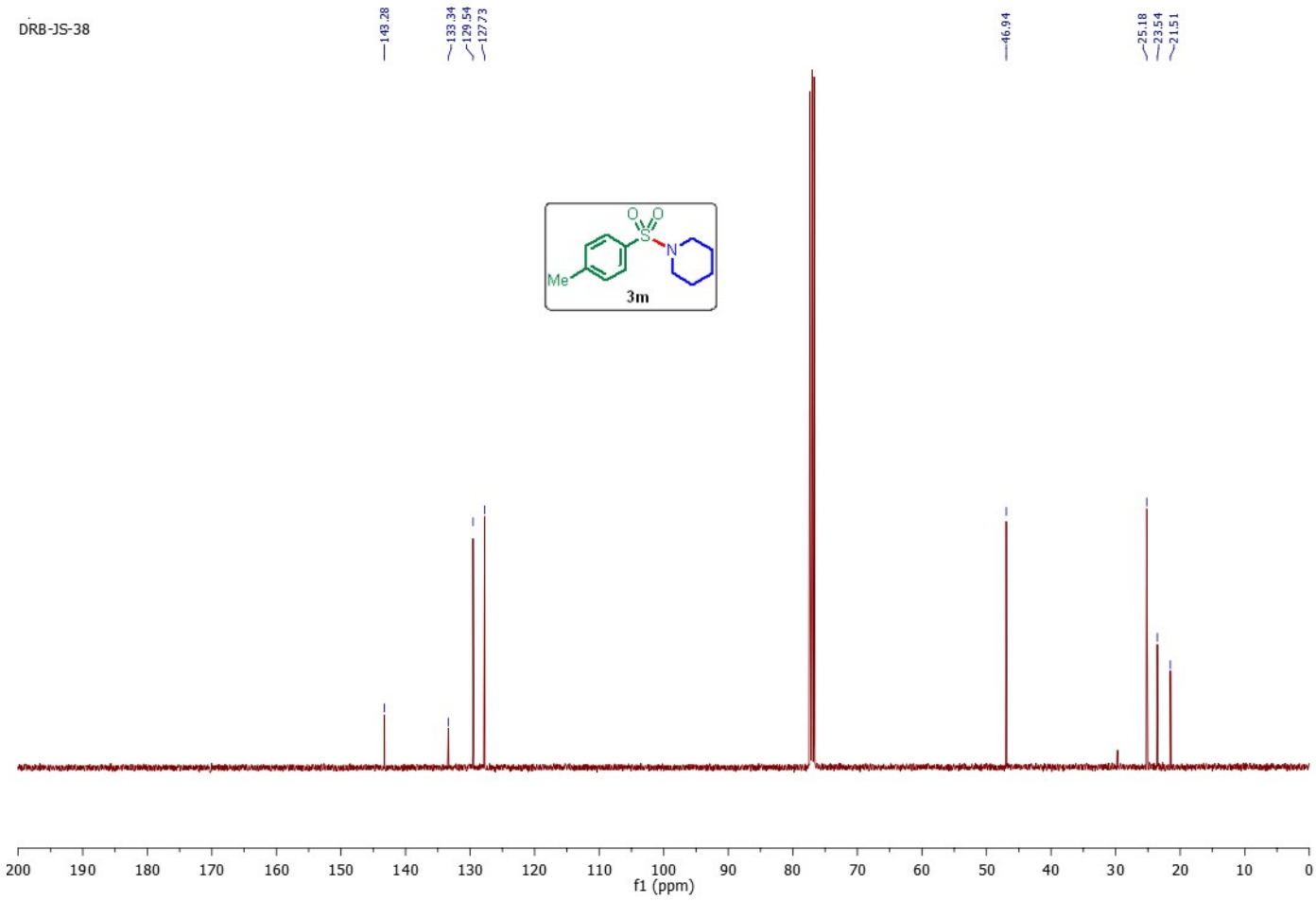

DRB-JS-64
140.00
132.55
129.08
127.13
42.94
31.72
19.57
13.64
3k
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

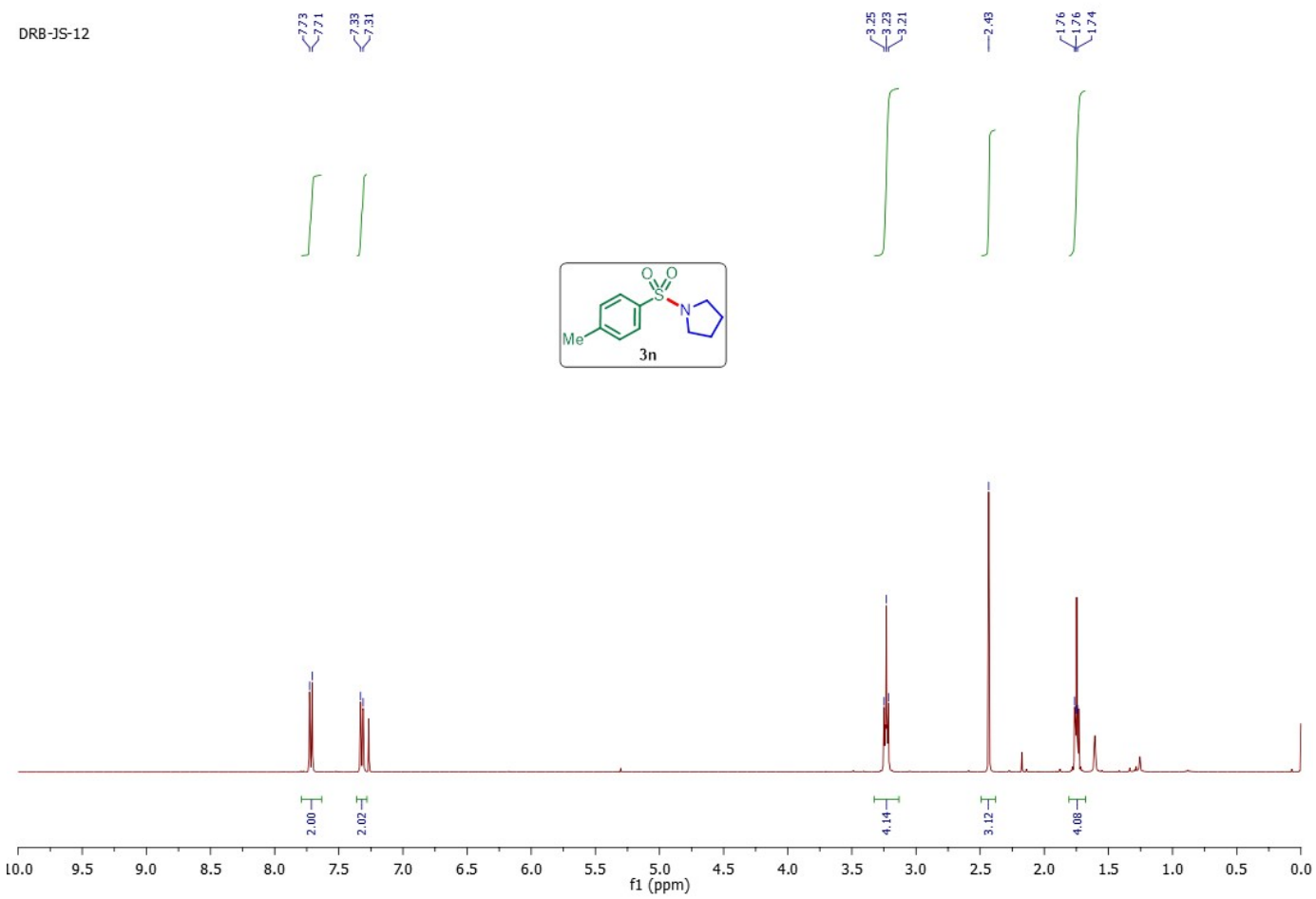

DRB-JS-83
7.83
7.82
7.81
7.80
7.56
7.54
7.53
7.51
7.50
7.48
3.27
3.26
3.24
3.22
1.15
1.13
1.11
3l
1.96
3.11
3.95
6.02
f1 (ppm)

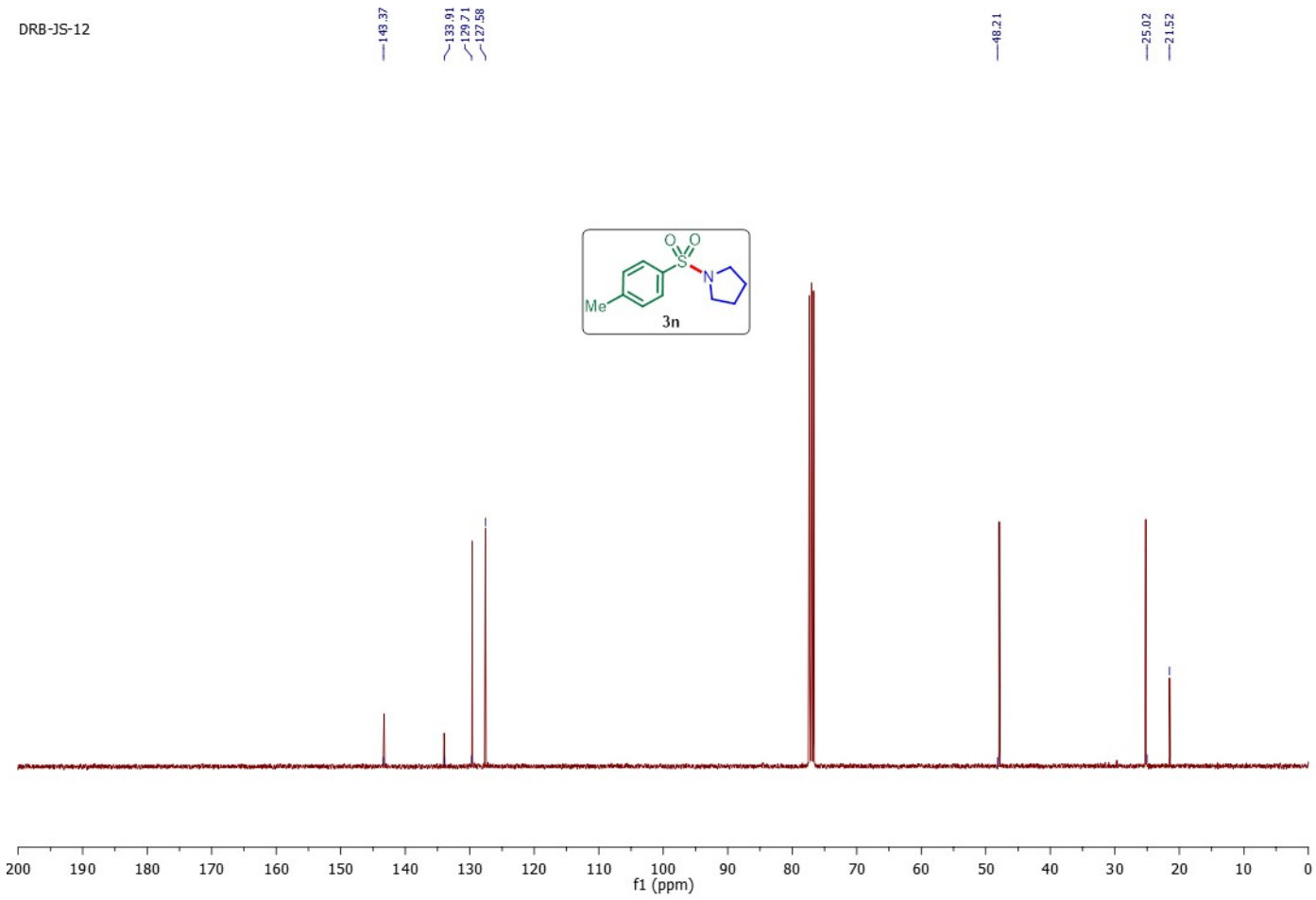
DRB-JS-83

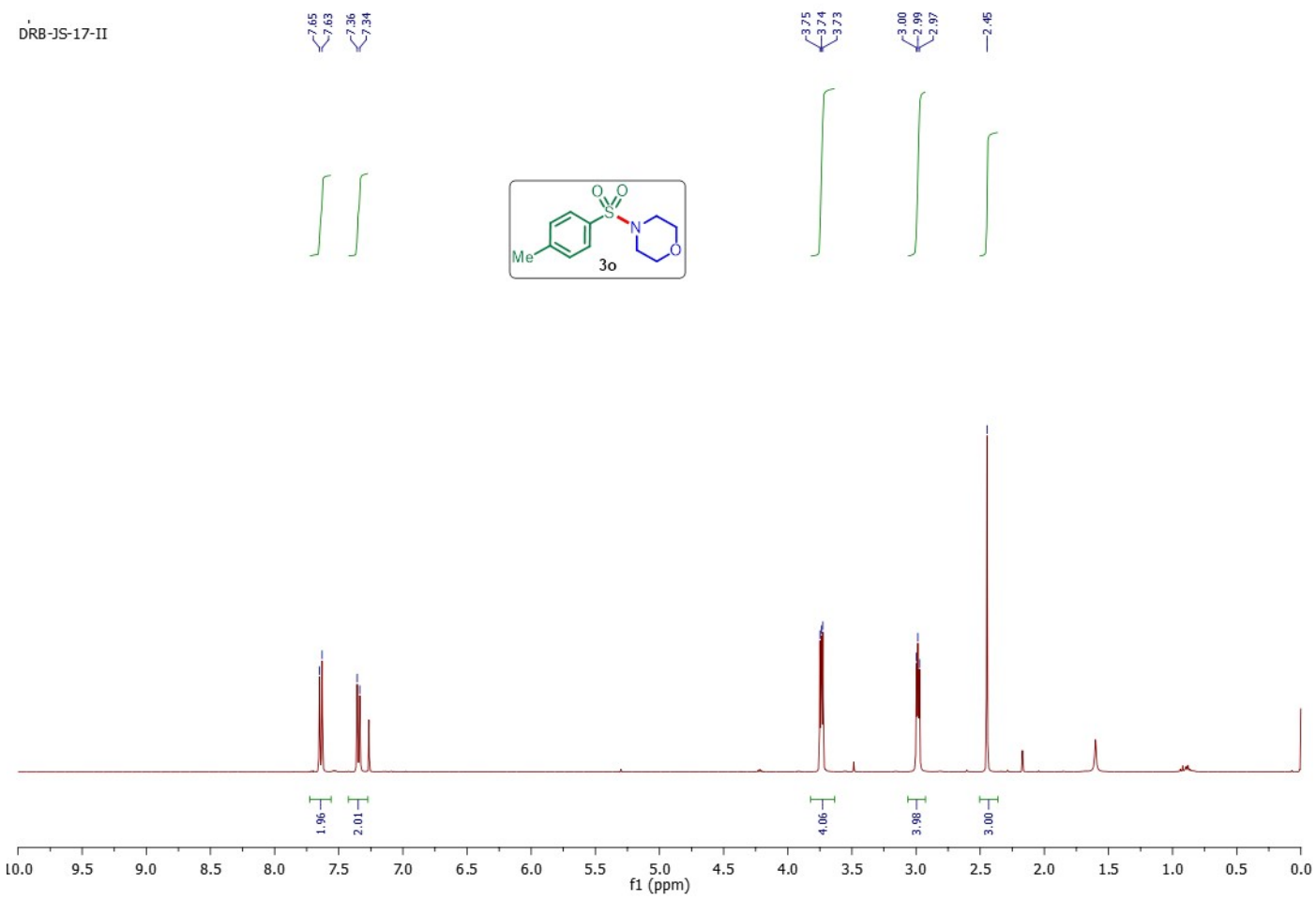

140.42
132.24
129.00
126.97
42.03
14.12
3l
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

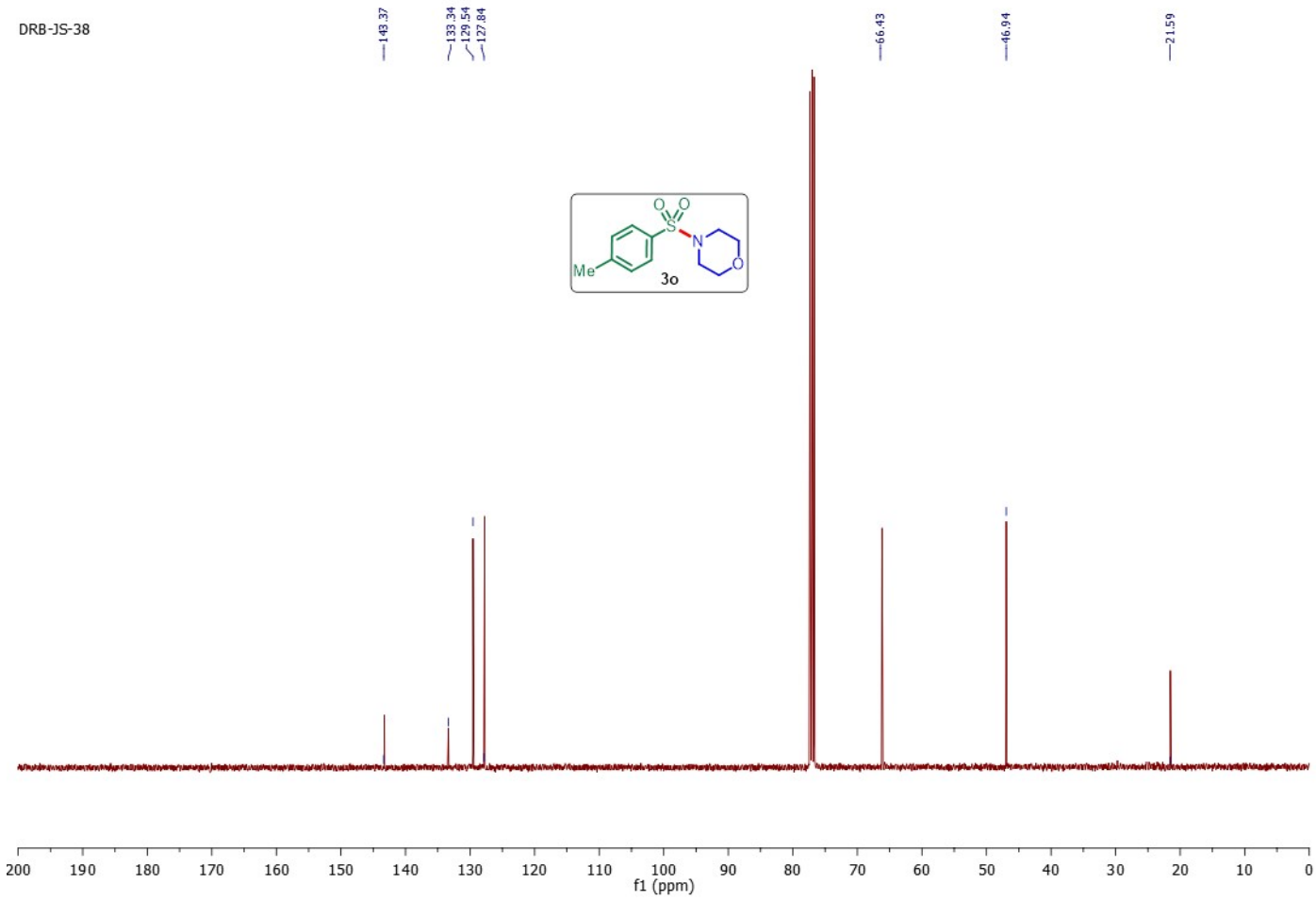

DRB-JS-38
7.65
7.63
7.33
7.31
2.98
2.97
2.95
2.43
1.67
1.65
1.64
1.62
1.61
1.44
1.42
1.41
1.39
1.38
Me
3m
1.94
1.95
4.09
3.12
4.01
2.14
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

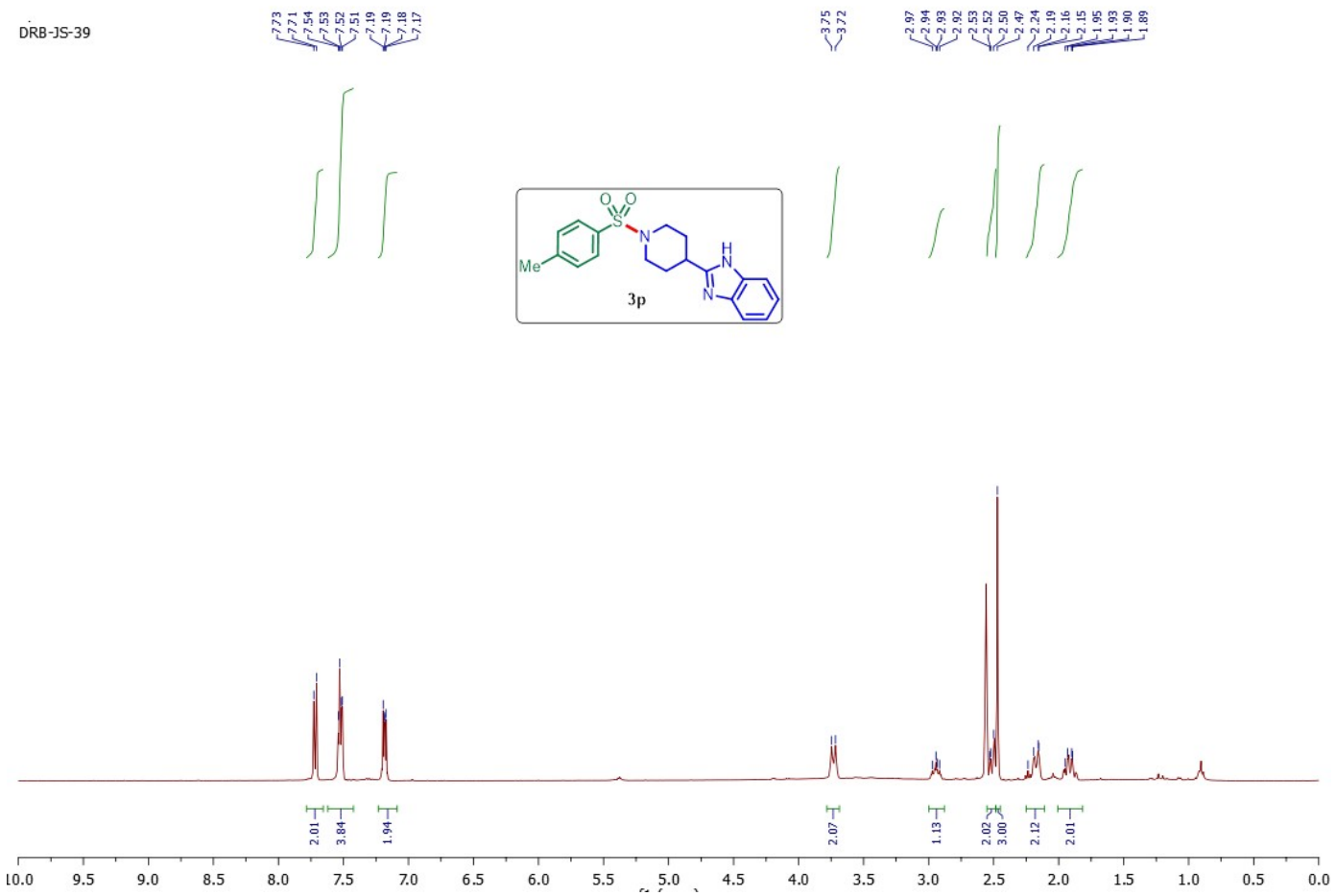

DRB-JS-38
143.28
133.34
129.54
127.73
46.94
25.18
23.54
21.51
Me
3m
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

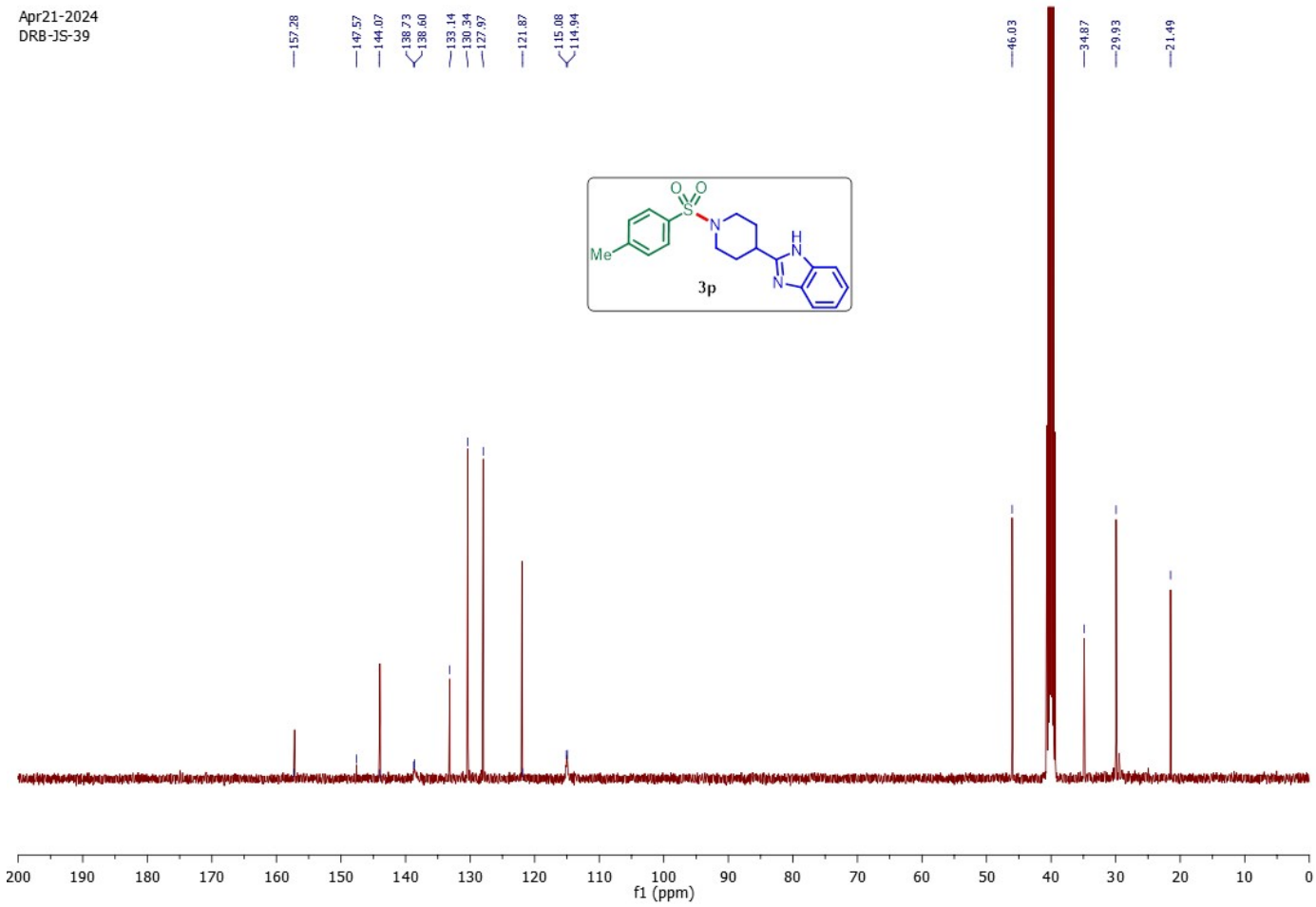

DRB-JS-12
7.73
7.71
7.33
7.31
3.25
3.23
3.21
2.43
1.76
1.76
1.74
Me
3n
2.00
2.02
4.14
3.12
4.08
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

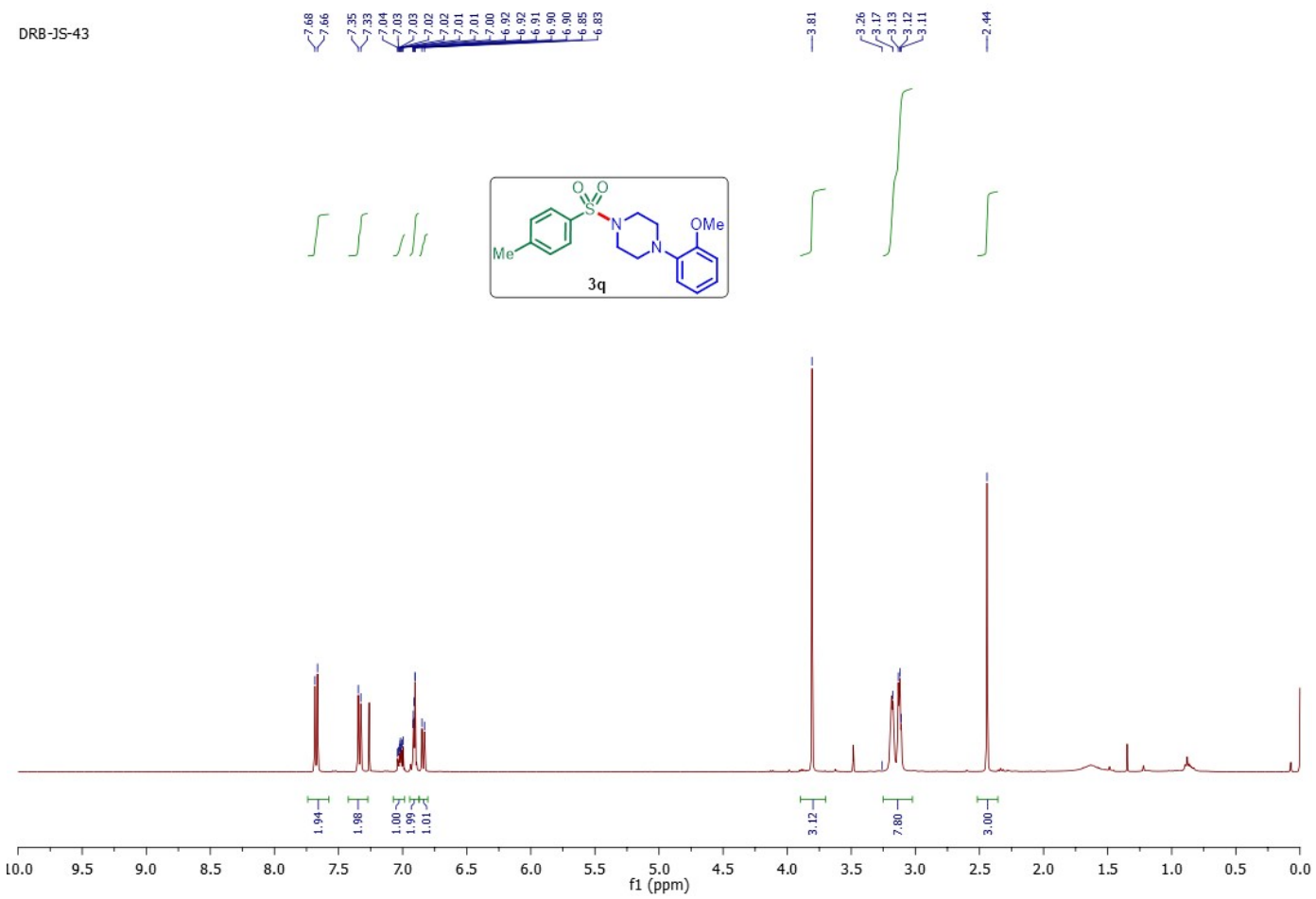

DRB-JS-12
143.37
133.91
129.71
127.58
48.21
25.02
21.52
Me
3n
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

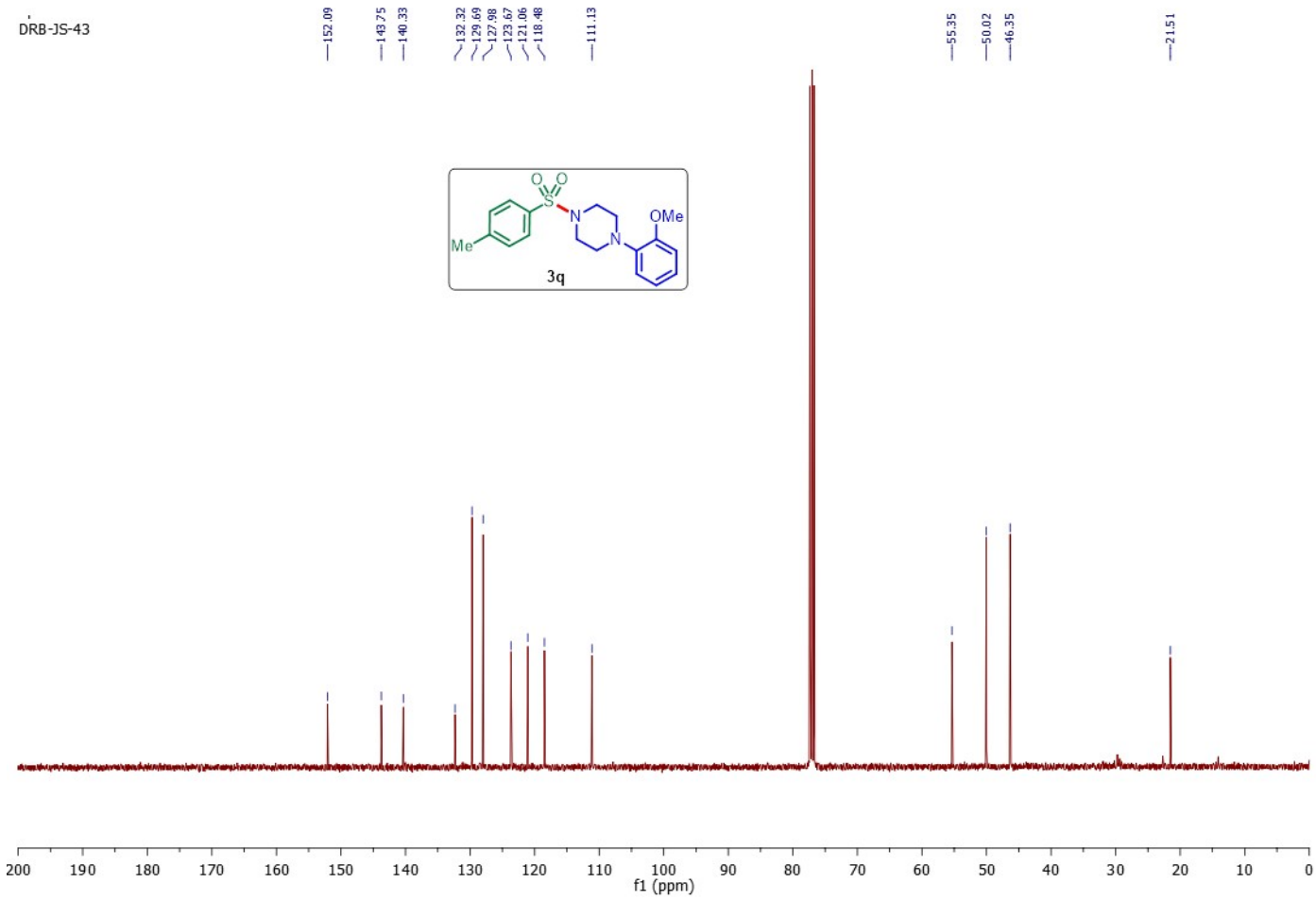

DRB-JS-17-II
7.65
7.63
7.36
7.34
3.75
3.74
3.73
3.00
2.99
2.97
2.45
Me
3o
1.96
2.01
4.06
3.98
3.00
f1 (ppm)

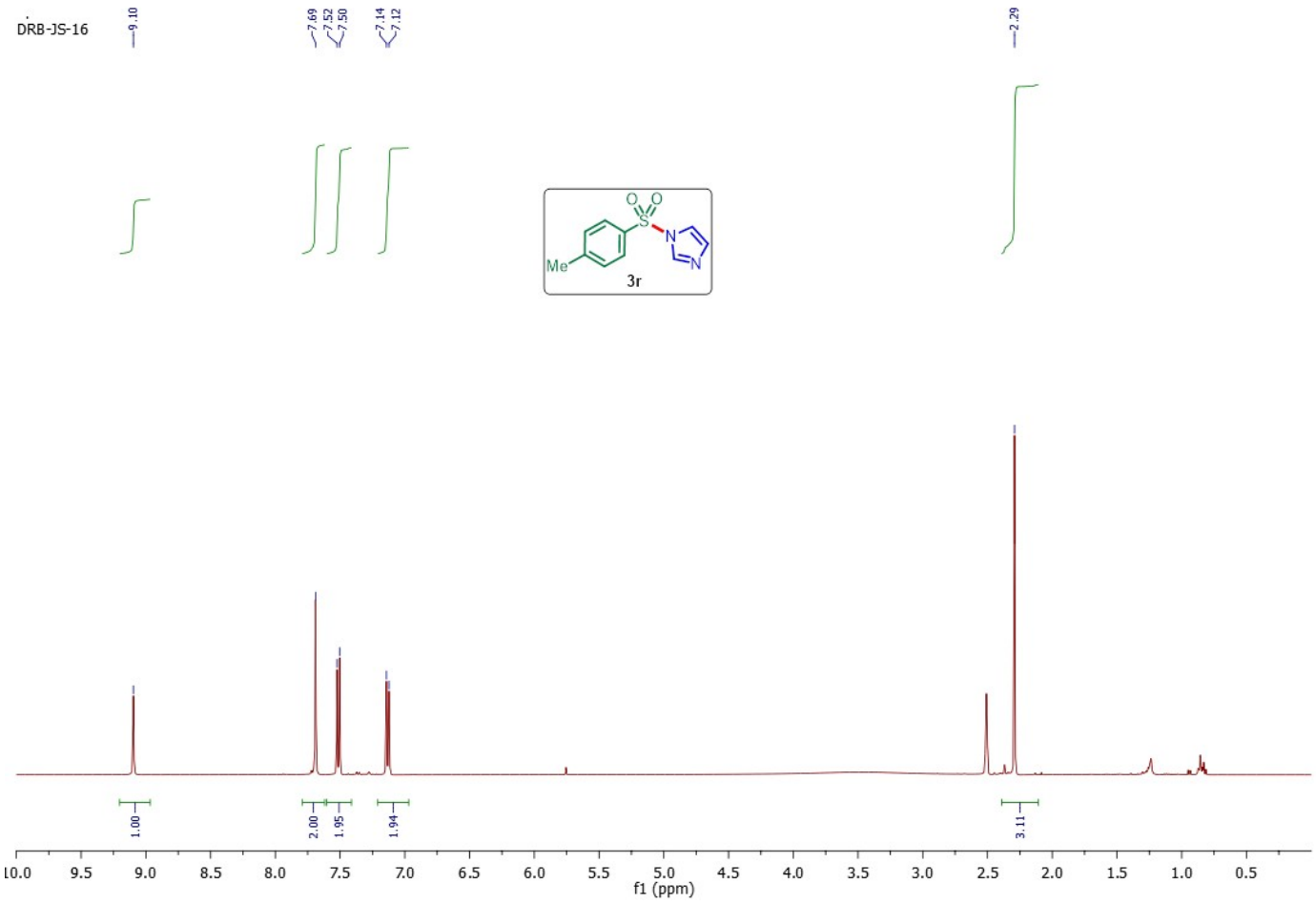

DRB-JS-38
143.37
133.34
129.54
127.84
66.43
46.94
21.59
Me
3o
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

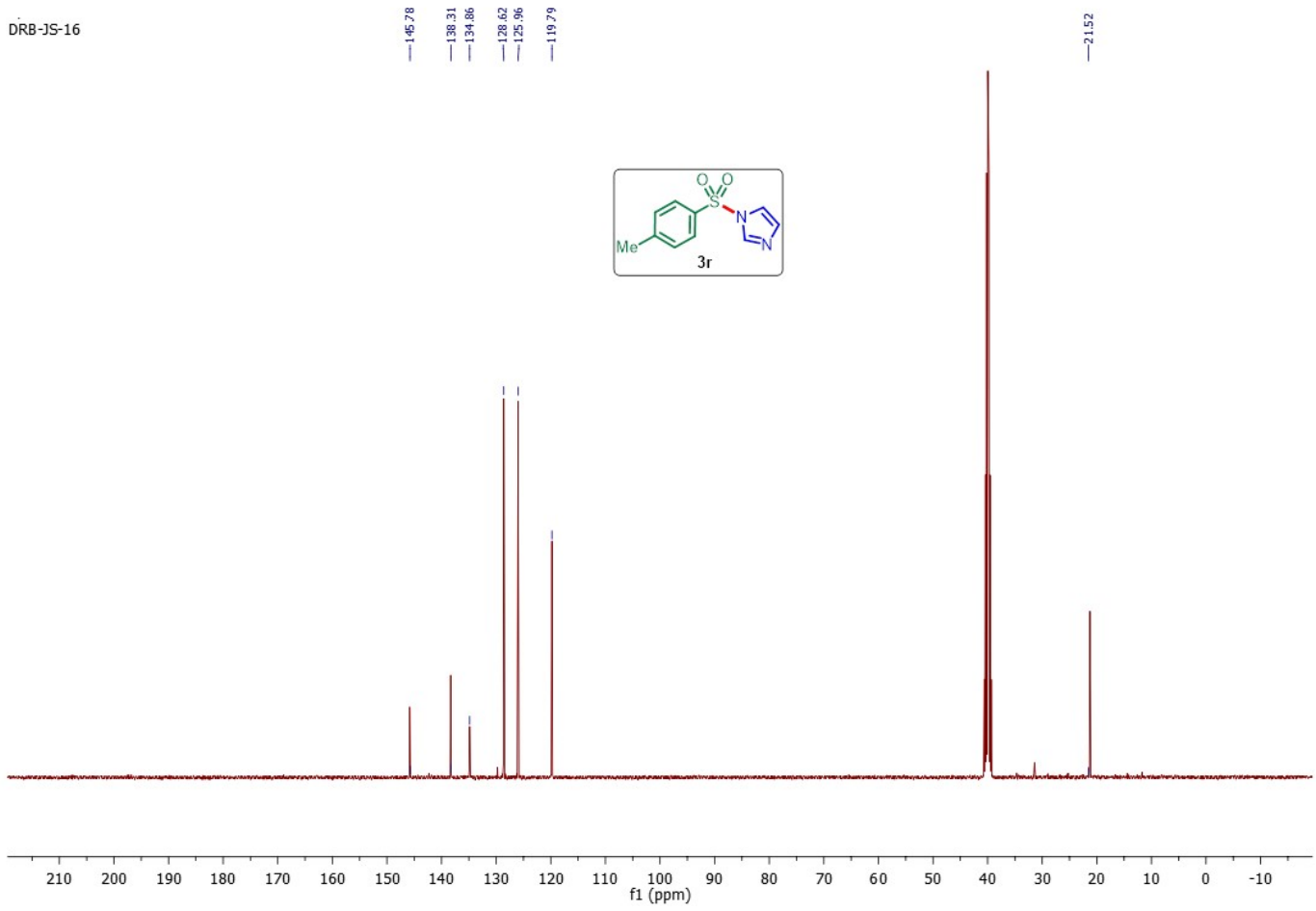

DRB-JS-39
7.73
7.71
7.54
7.53
7.52
7.51
7.19
7.19
7.18
7.17
3.75
3.72
2.97
2.94
2.93
2.92
2.53
2.52
2.50
2.47
2.24
2.19
2.16
2.15
1.95
1.93
1.90
1.89
Me
3p
2.01
3.84
1.94
2.07
1.13
2.02
3.00
2.12
2.01
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0

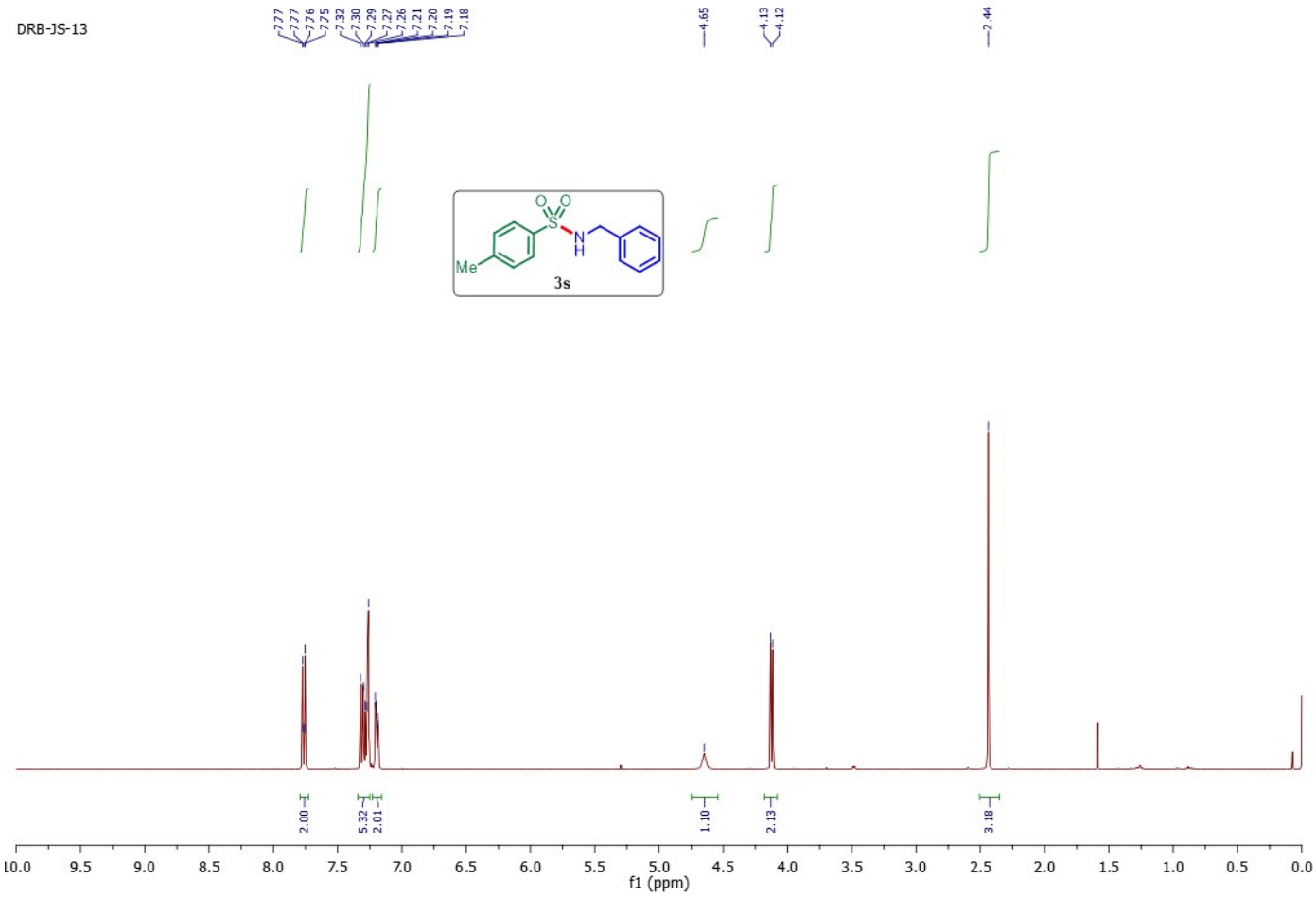

Apr21-2024
DRB-JS-39
157.28
147.57
144.07
138.73
138.60
133.14
130.34
127.97
121.87
115.08
114.94
46.03
34.87
29.93
21.49
Me
3p
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

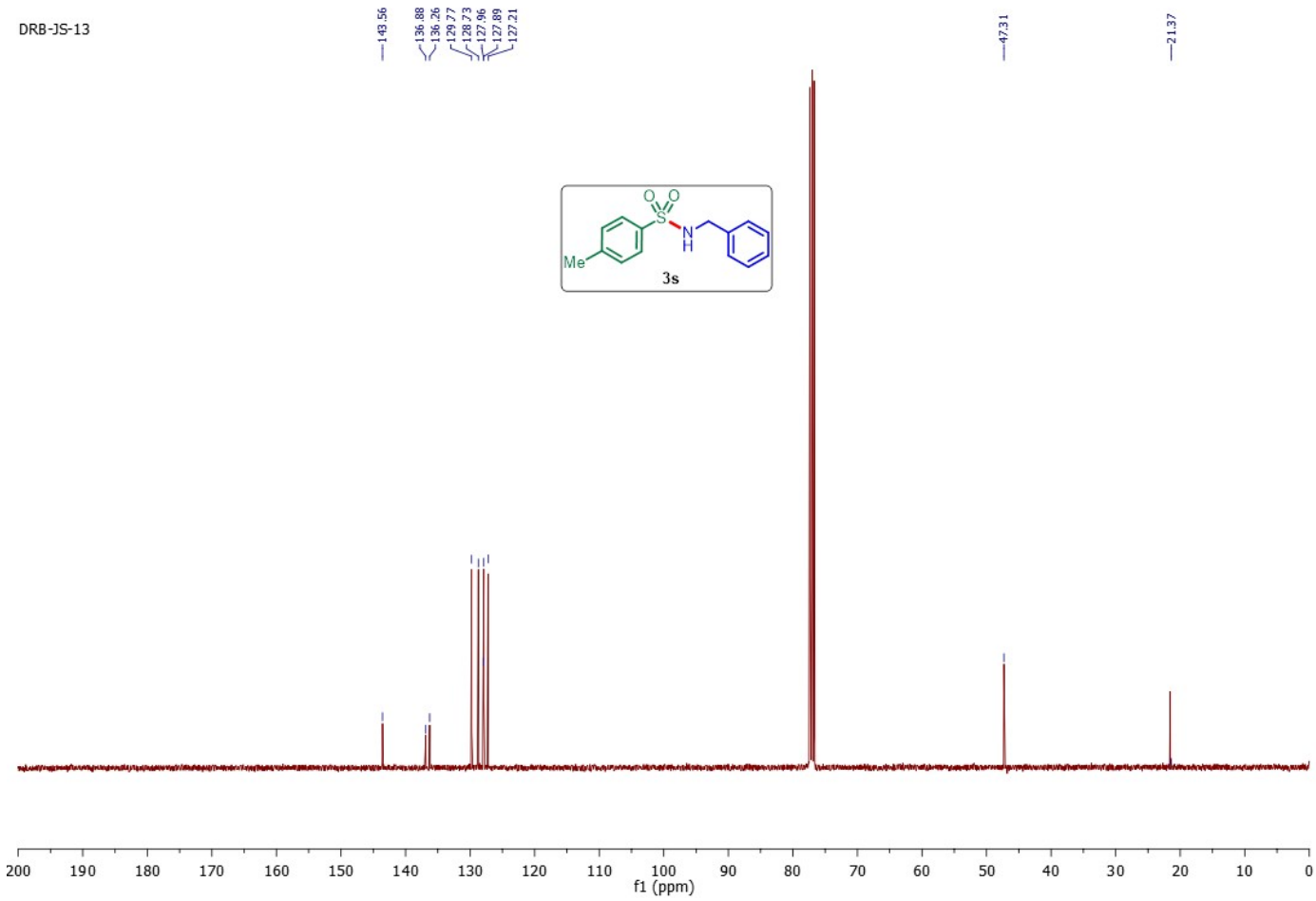

DRB-JS-43
7.68
7.66
7.35
7.33
7.04
7.03
7.03
7.02
7.02
7.01
7.01
7.00
6.92
6.92
6.91
6.90
6.90
6.85
6.83
3.81
3.26
3.17
3.13
3.12
3.11
2.44
O
O
S
N
N
Me
OMe
3q
1.94
1.98
1.00
1.99
1.01
3.12
7.80
3.00
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

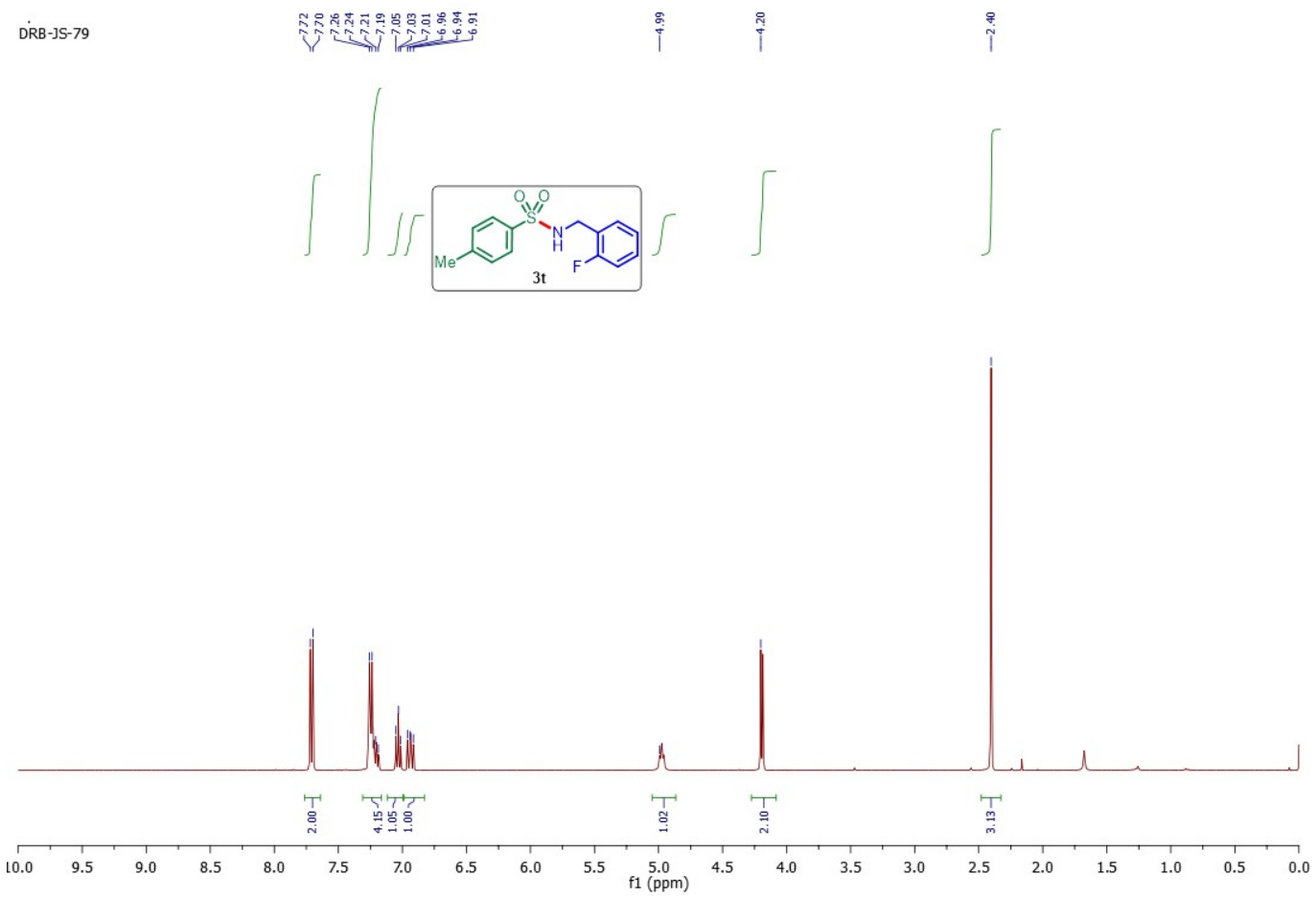

DRB-JS-43
152.09
143.75
140.33
132.32
129.69
127.98
123.67
121.06
118.48
111.13
55.35
50.02
46.35
21.51
Me
OMe
3q
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

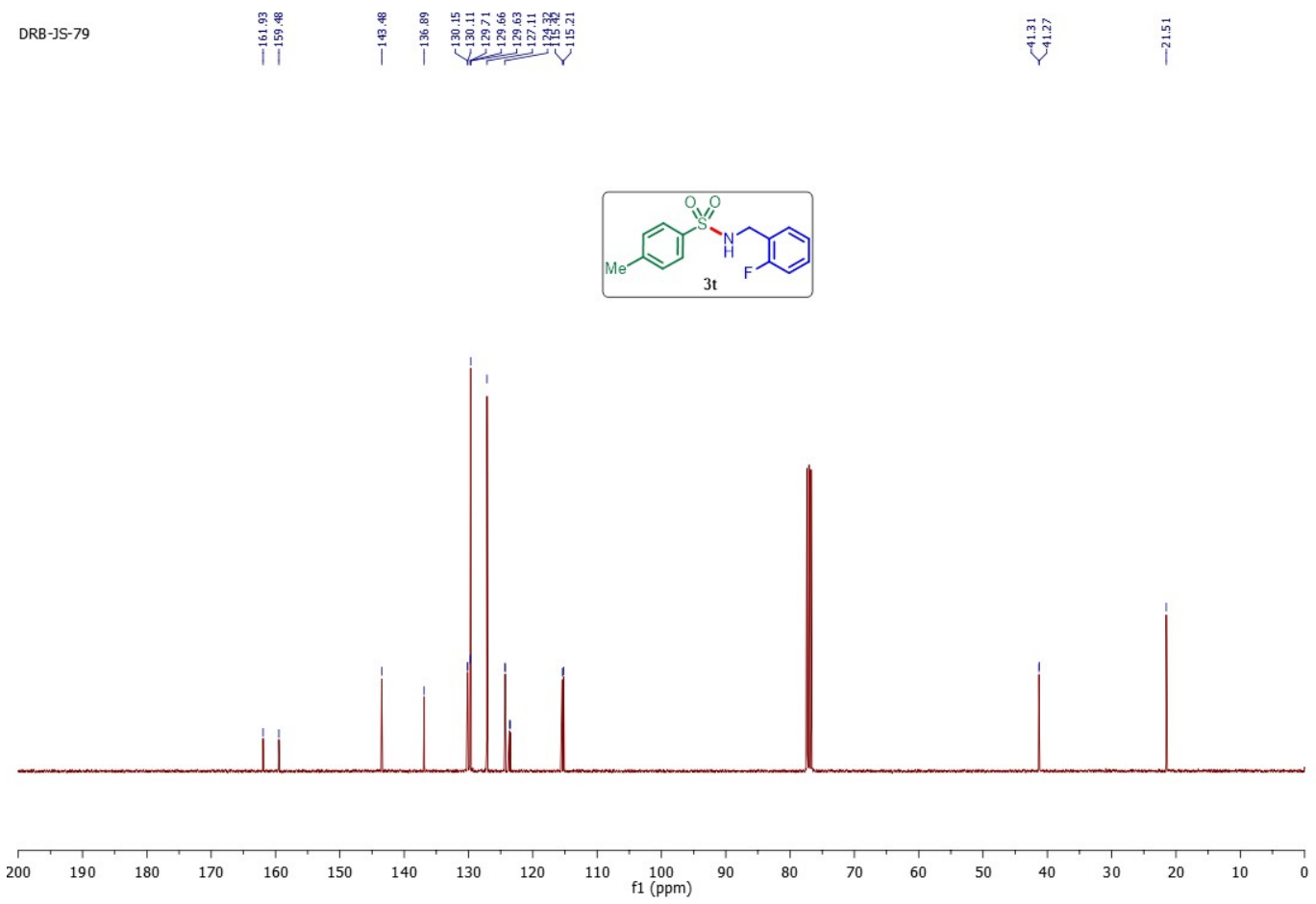

DRB-JS-16
9.10
7.69
7.52
7.50
7.14
7.12
2.29
Me
3r
1.00
2.00
1.95
1.94
3.11
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

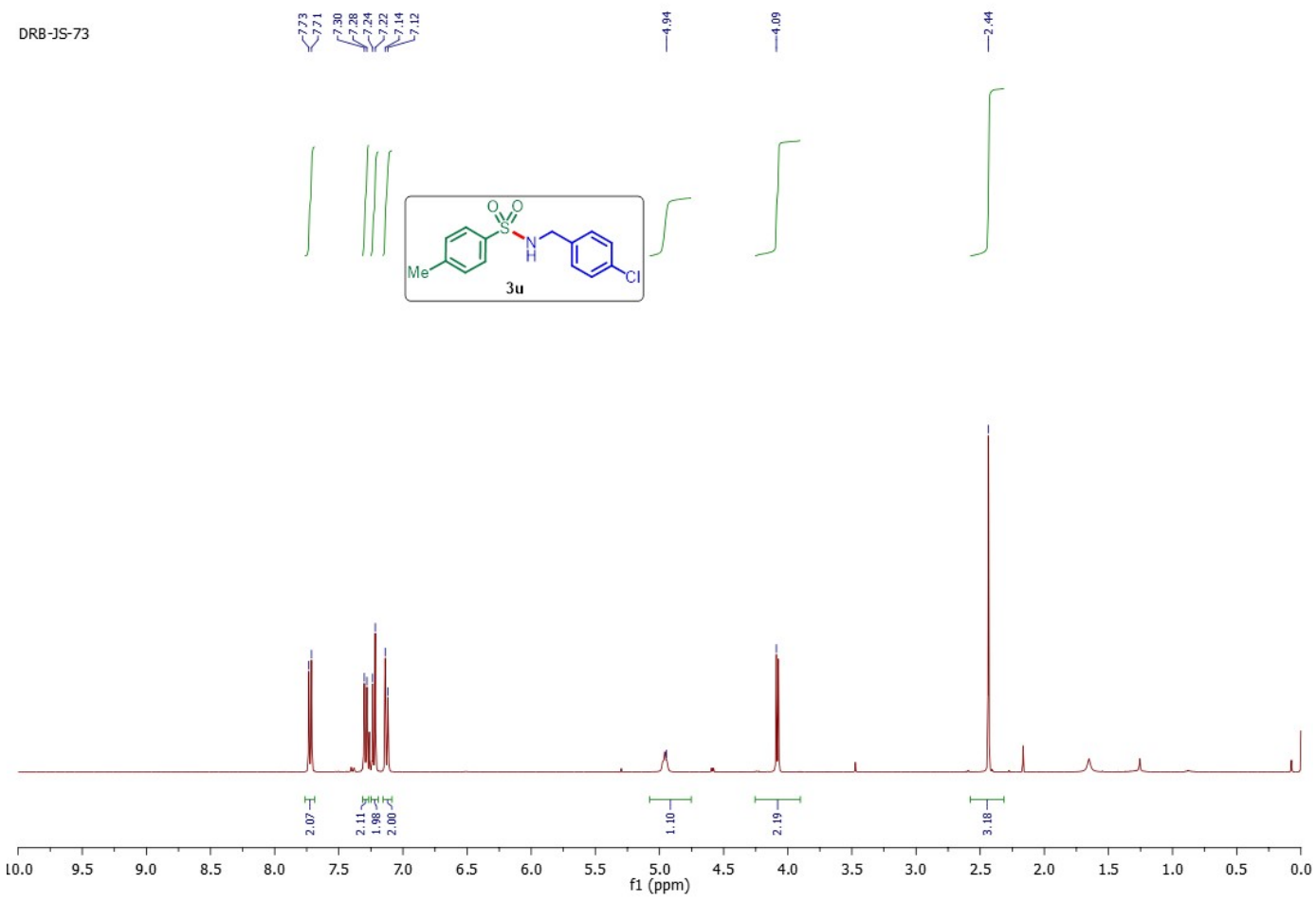

DRB-JS-16
145.78
138.31
134.86
128.62
125.96
119.79
21.52
Me
3r
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


DRB-JS-13
7.77
7.77
7.76
7.75
7.32
7.30
7.29
7.27
7.26
7.21
7.20
7.19
7.18
4.65
4.13
4.12
2.44
Me
3s
2.00
5.32
2.01
1.10
2.13
3.18
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


DRB-JS-13
143.56
136.88
136.26
129.77
128.73
127.96
127.89
127.21
47.31
21.37
Me
3s
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


DRB-JS-79
7.72
7.70
7.26
7.24
7.21
7.19
7.05
7.03
7.01
6.96
6.94
6.91
4.99
4.20
2.40
O
O
S
N
H
Me
F
3t
2.00
4.15
1.05
1.00
1.02
2.10
3.13
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

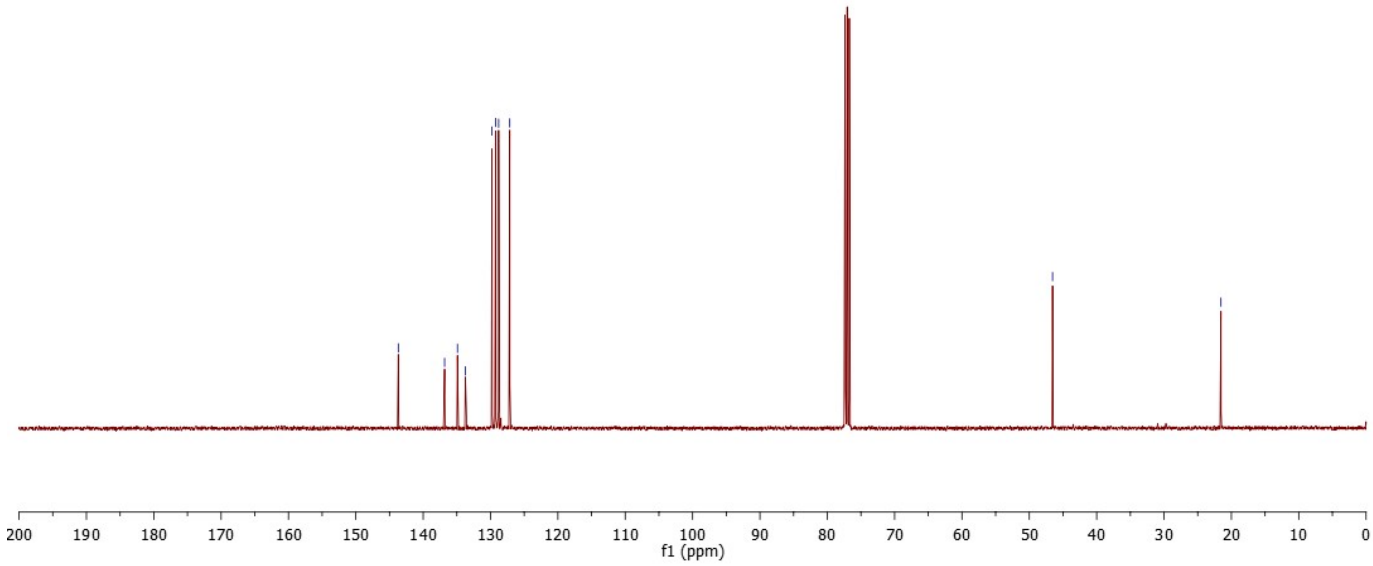

DRB-JS-79
161.93
159.48
143.48
136.89
130.15
130.11
129.71
129.66
129.63
127.11
124.32
115.42
115.21
41.31
41.27
21.51
Me
F
3t
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

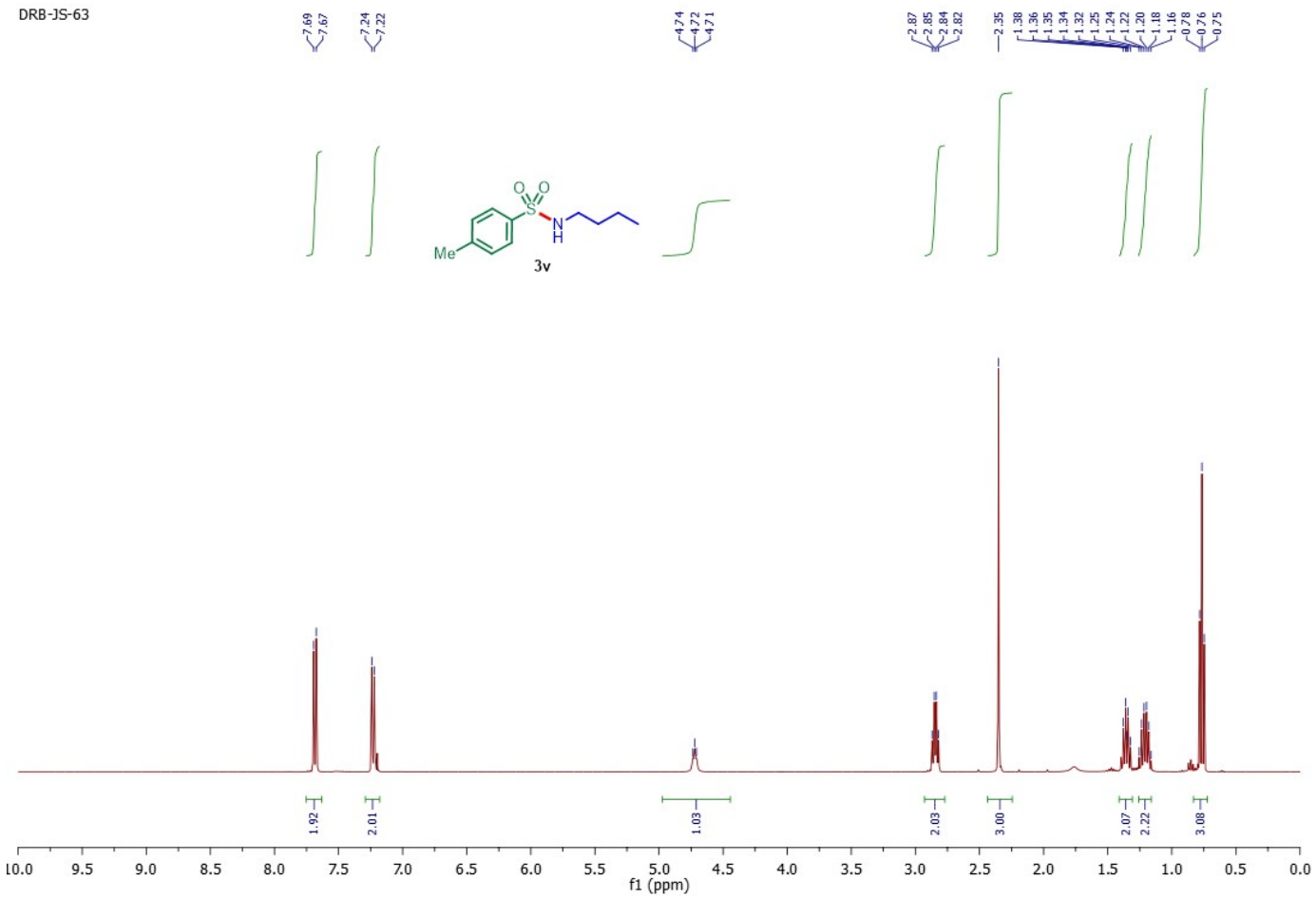

DRB-JS-73
7.73
7.71
7.30
7.28
7.24
7.22
7.14
7.12
4.94
4.09
2.44
Me
Cl
3u
2.07
2.11
1.98
2.00
1.10
2.19
3.18
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

Apr25-2024
DRB-JS-73

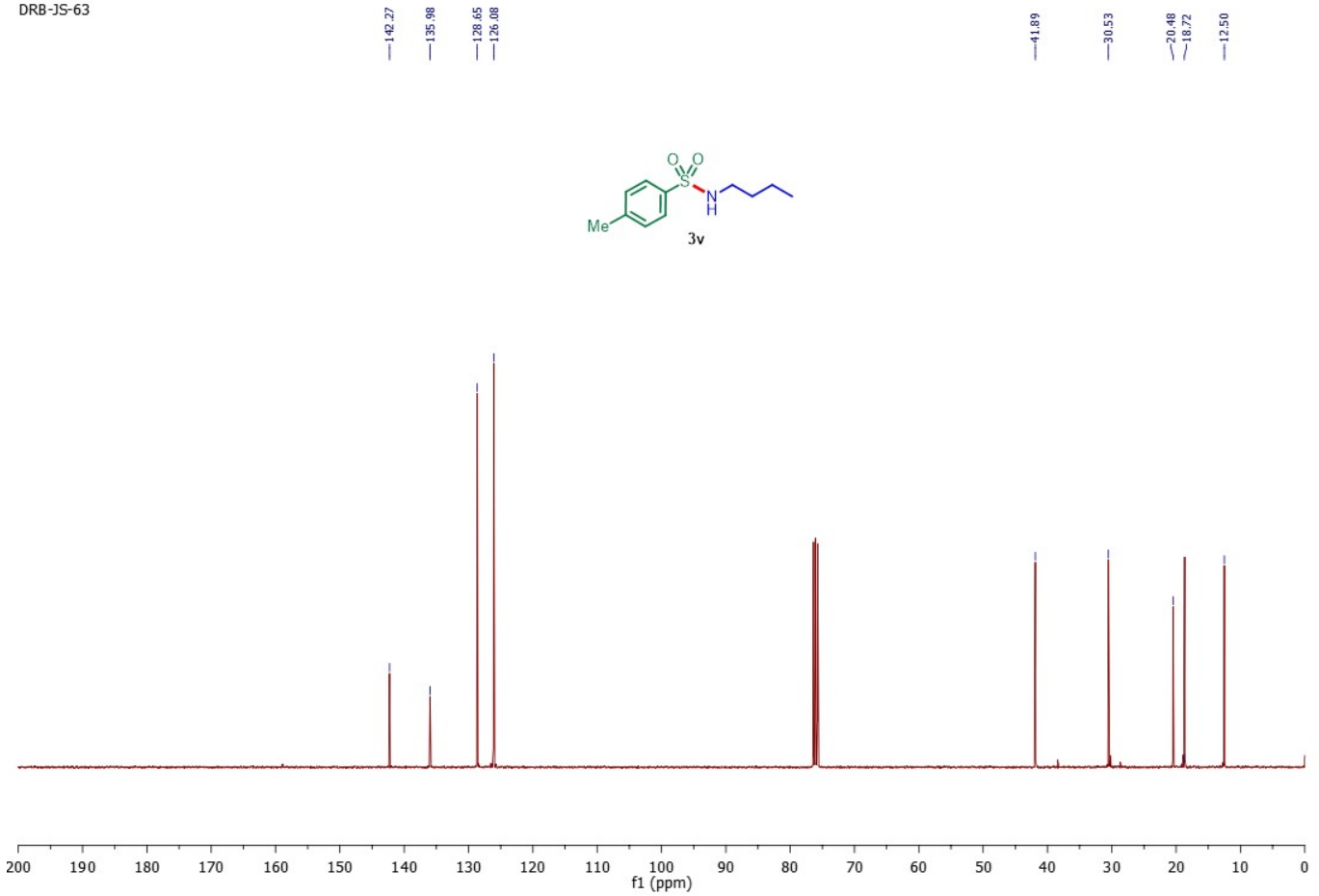
143.69
136.82
134.91
133.73
129.78
129.23
128.79
127.15
46.56
21.55

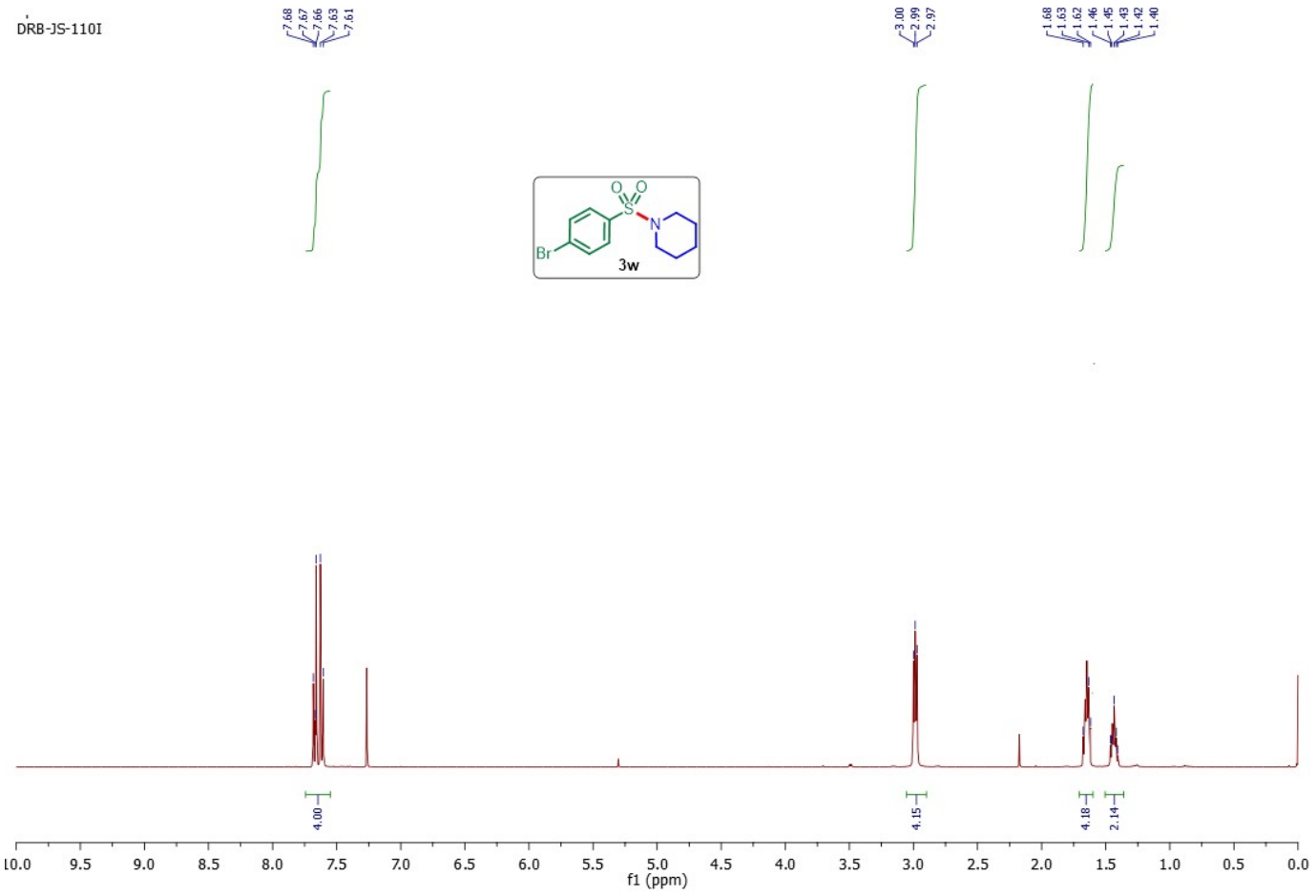
3u

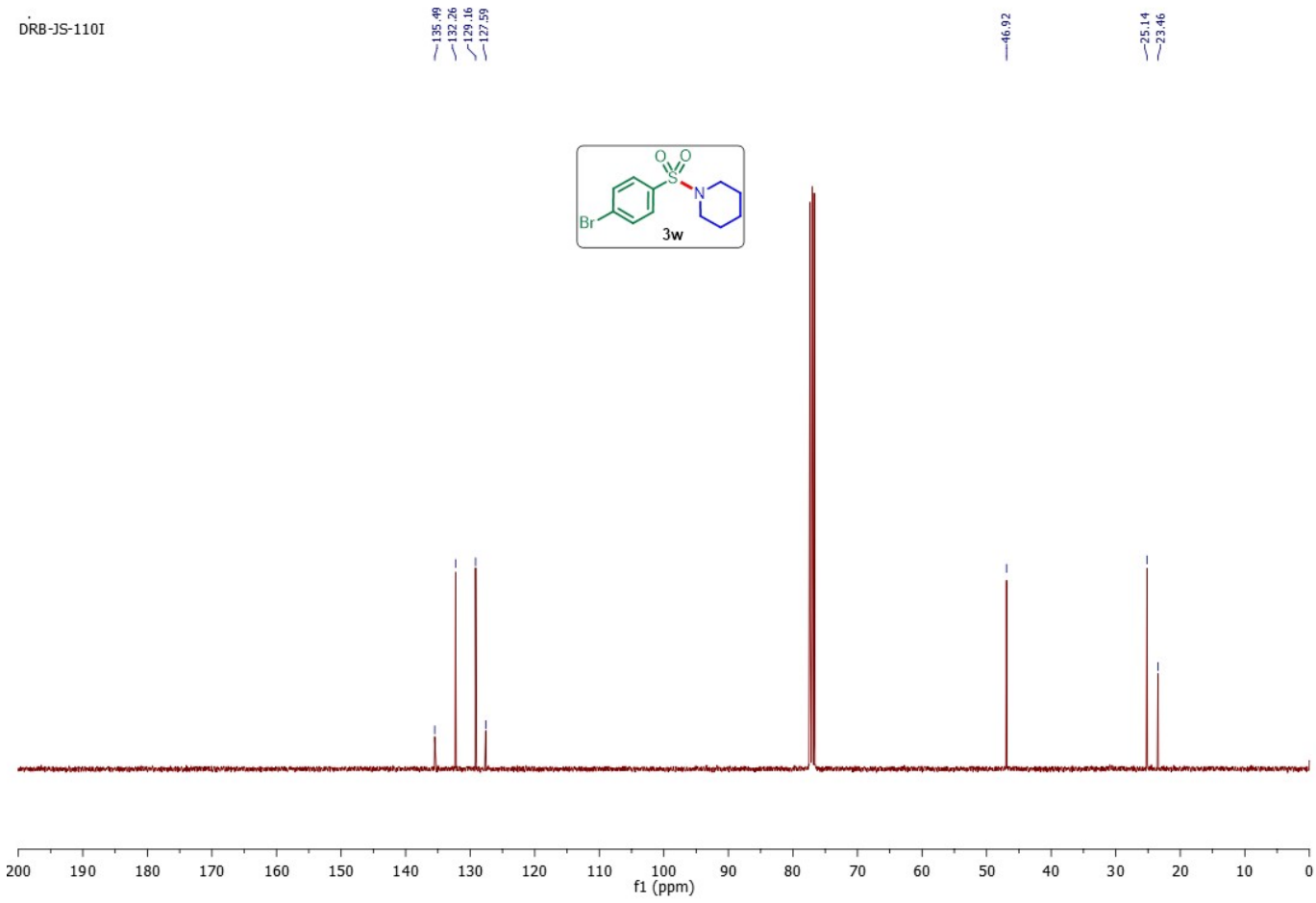


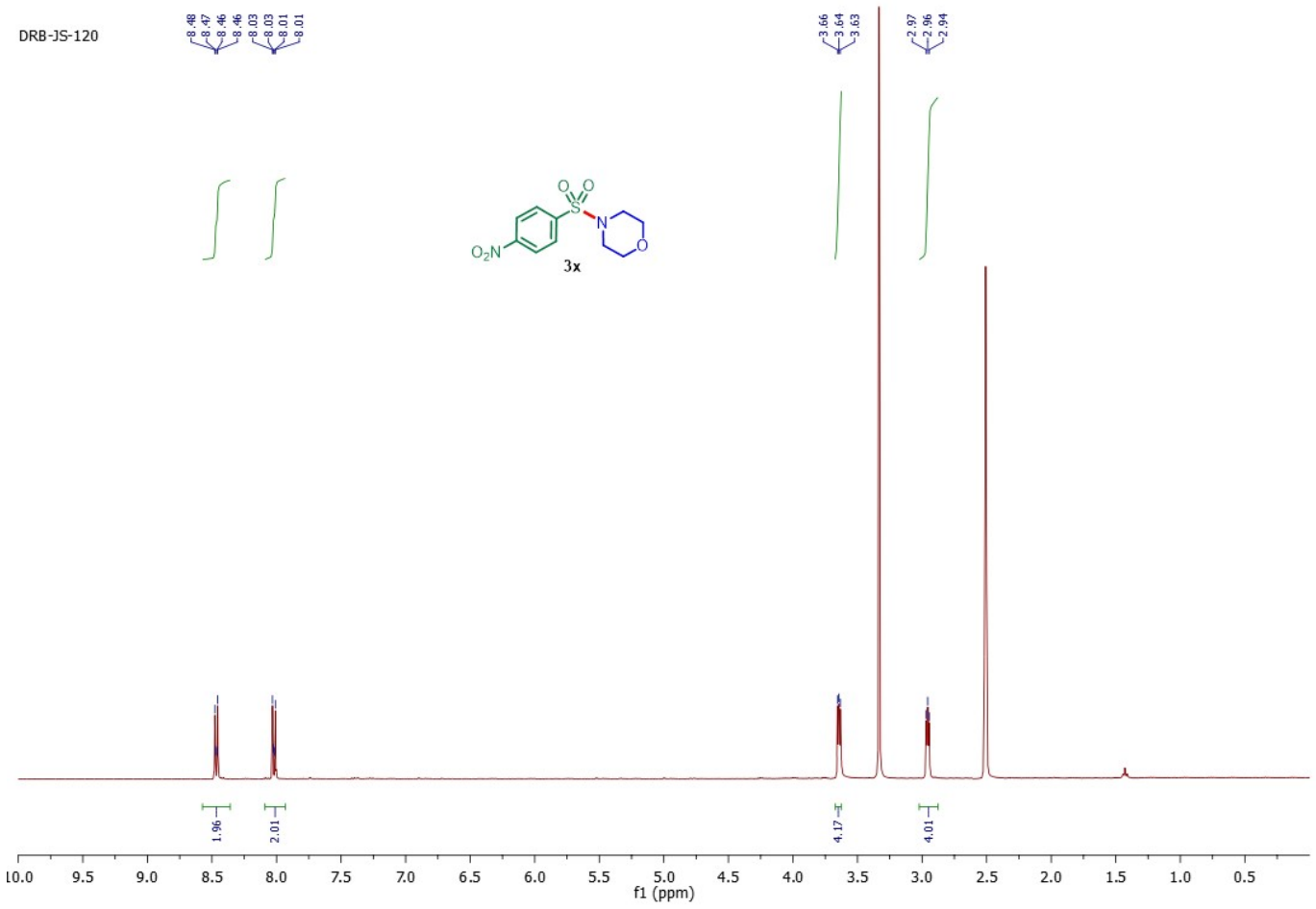

DRB-JS-63
7.69
7.67
7.24
7.22
4.74
4.72
4.71
2.87
2.85
2.84
2.82
2.35
1.38
1.36
1.35
1.34
1.32
1.25
1.24
1.22
1.20
1.18
1.16
0.78
0.76
0.75
Me
3v
1.92
2.01
1.03
2.03
3.00
2.07
2.22
3.08
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

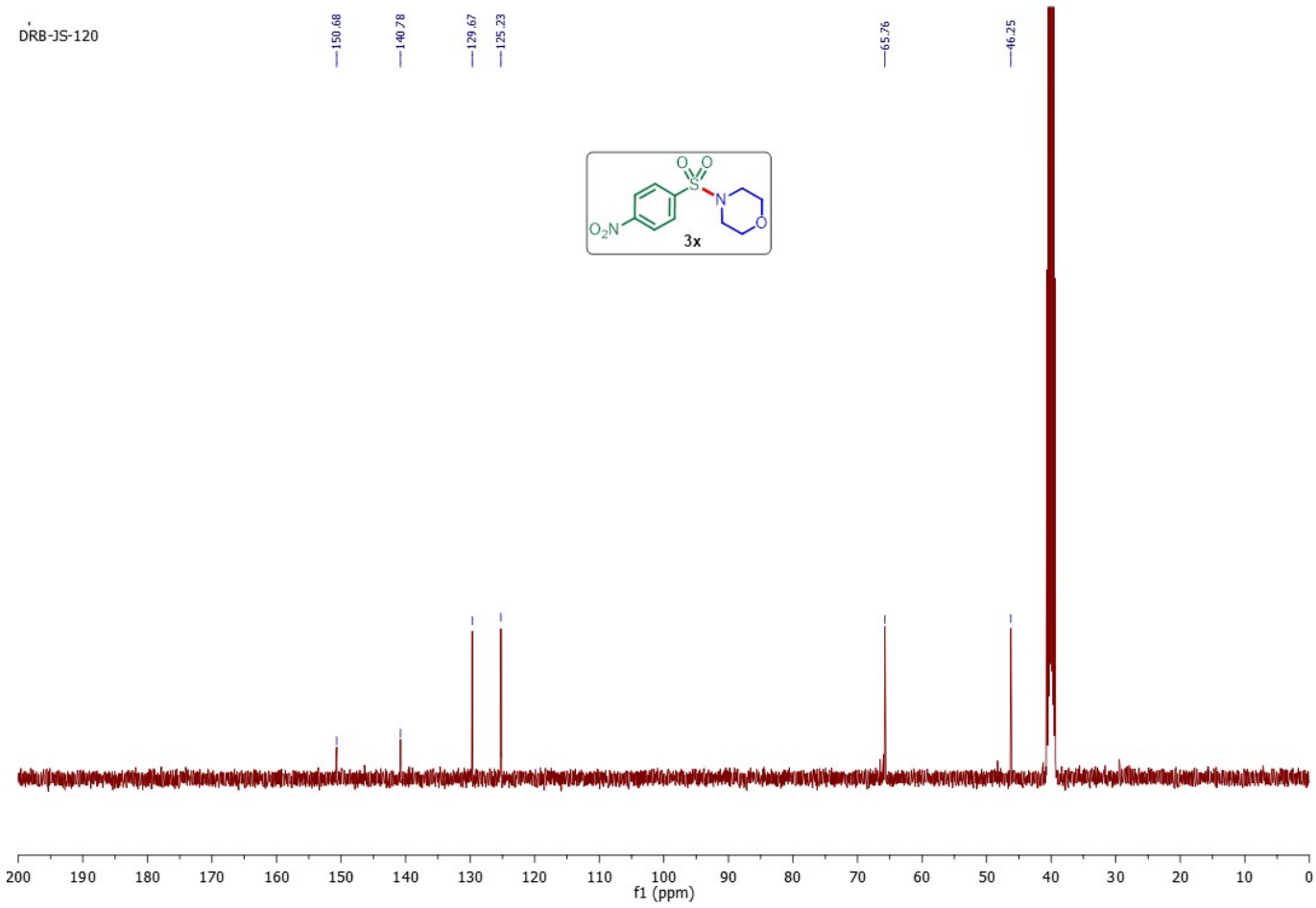
DRB-JS-63

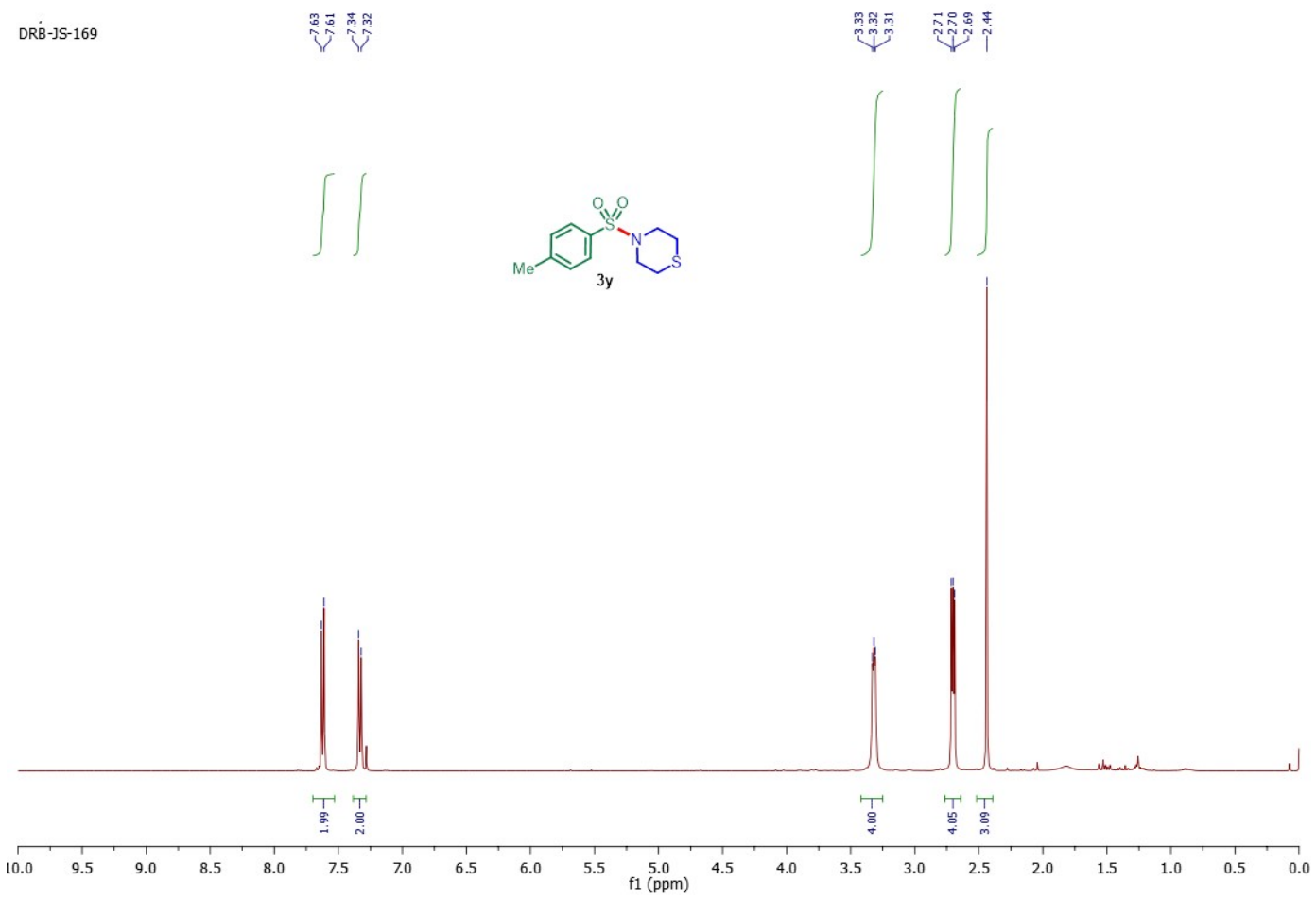

142.27
135.98
128.65
126.08
41.89
30.53
20.48
18.72
12.50
O
O
S
N
H
Me
3v
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

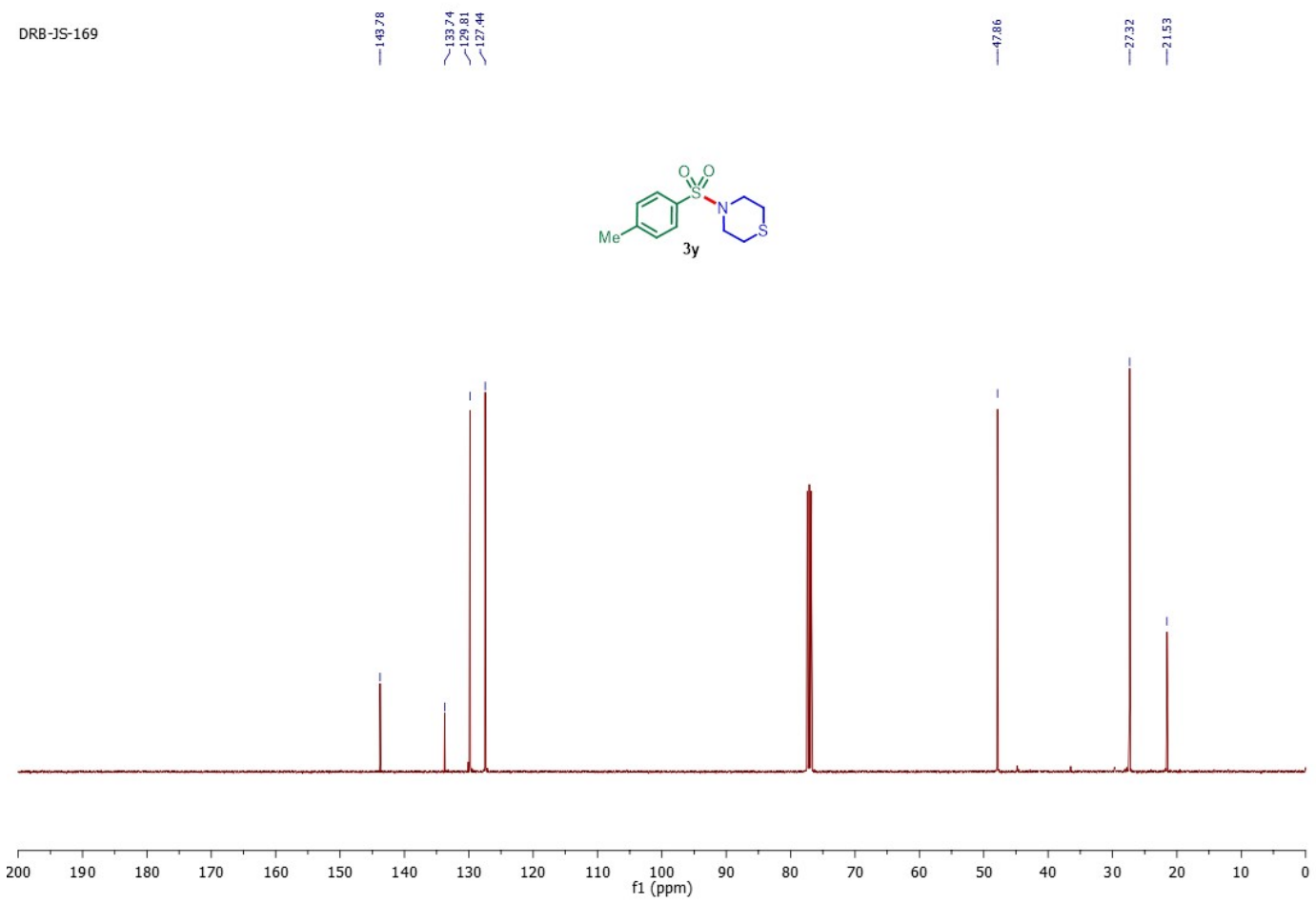

DRB-JS-110I
7.68
7.67
7.66
7.63
7.61
3.00
2.99
2.97
1.68
1.63
1.62
1.46
1.45
1.43
1.42
1.40
Br
3w
4.00
4.15
4.18
2.14
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

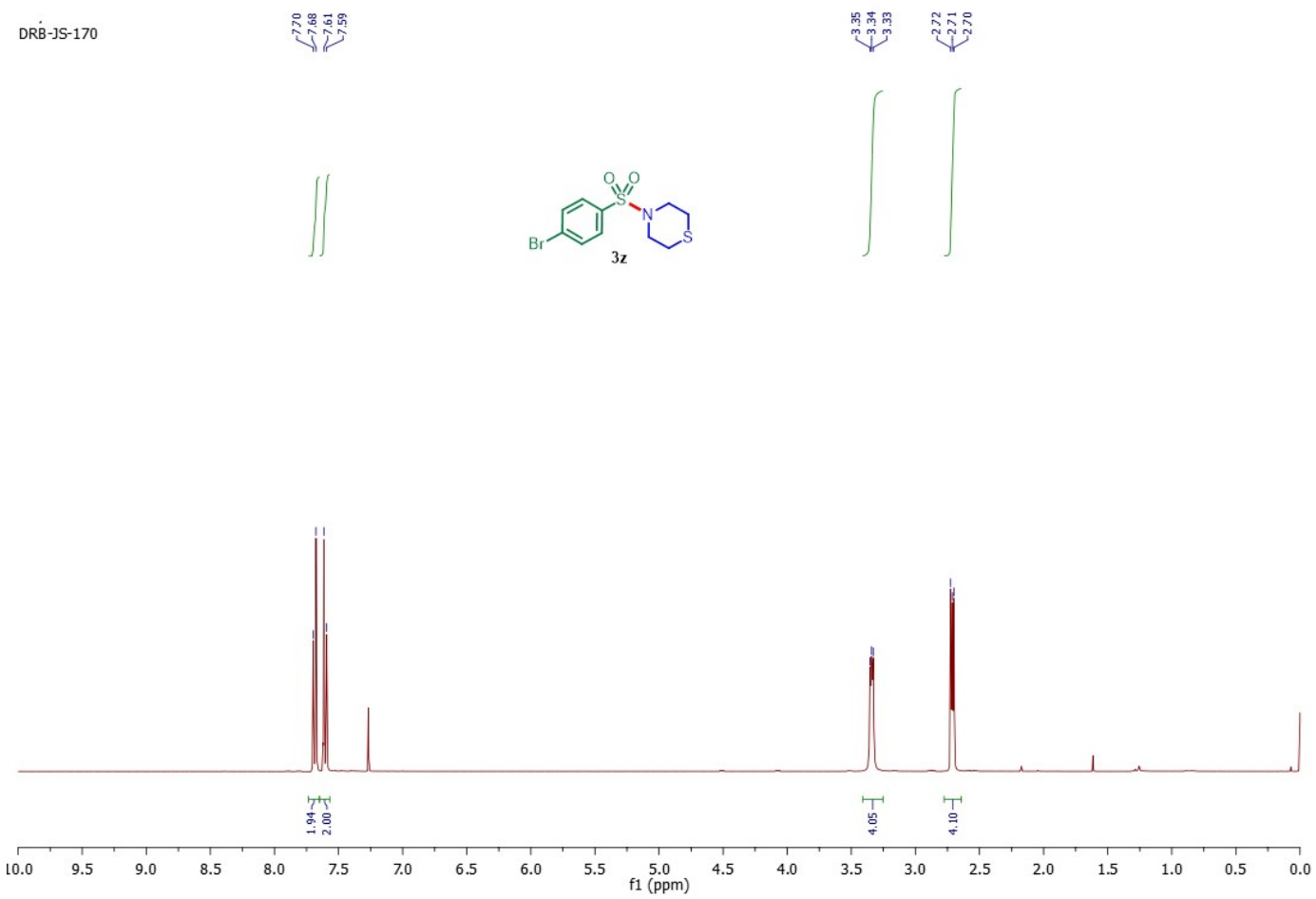

DRB-JS-110I
135.49
132.26
129.16
127.59
46.92
25.14
23.46
O
O
S
N
Br
3w
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

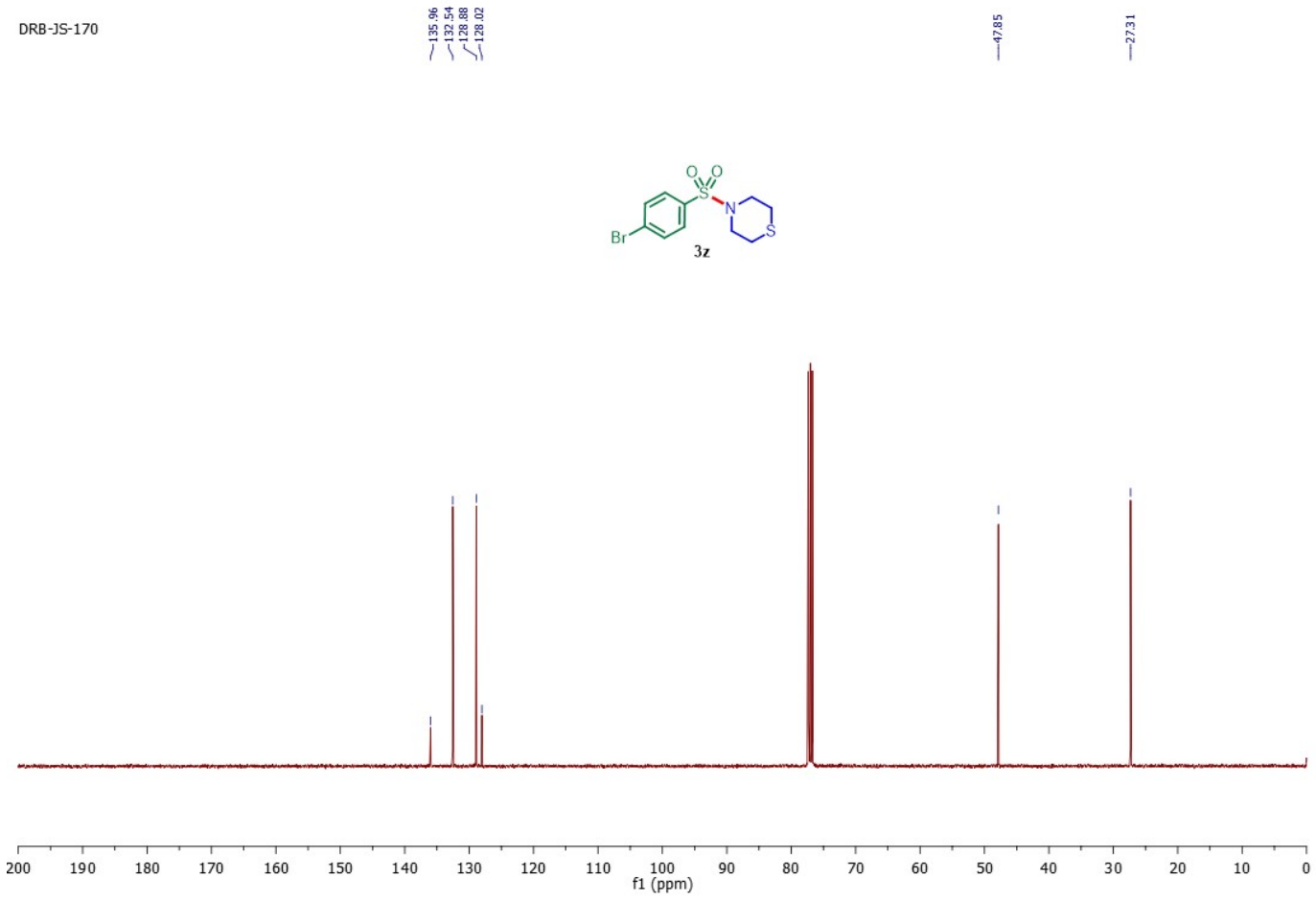

DRB-JS-120
8.48
8.47
8.46
8.46
8.03
8.03
8.01
8.01
3.66
3.64
3.63
2.97
2.96
2.94
3x
1.96
2.01
4.17
4.01
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

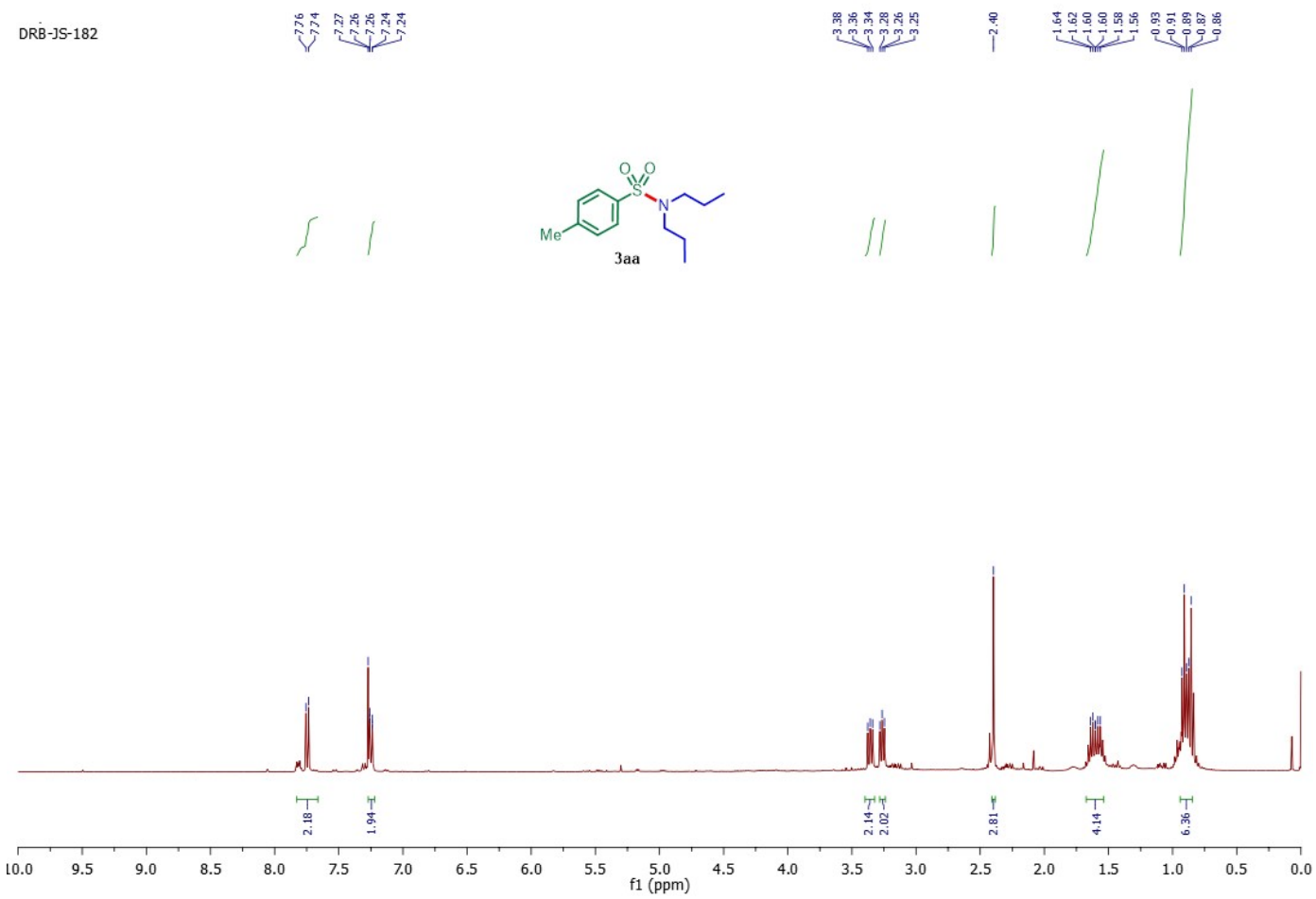

DRB-JS-120
150.68
140.78
129.67
125.23
65.76
46.25
3x
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

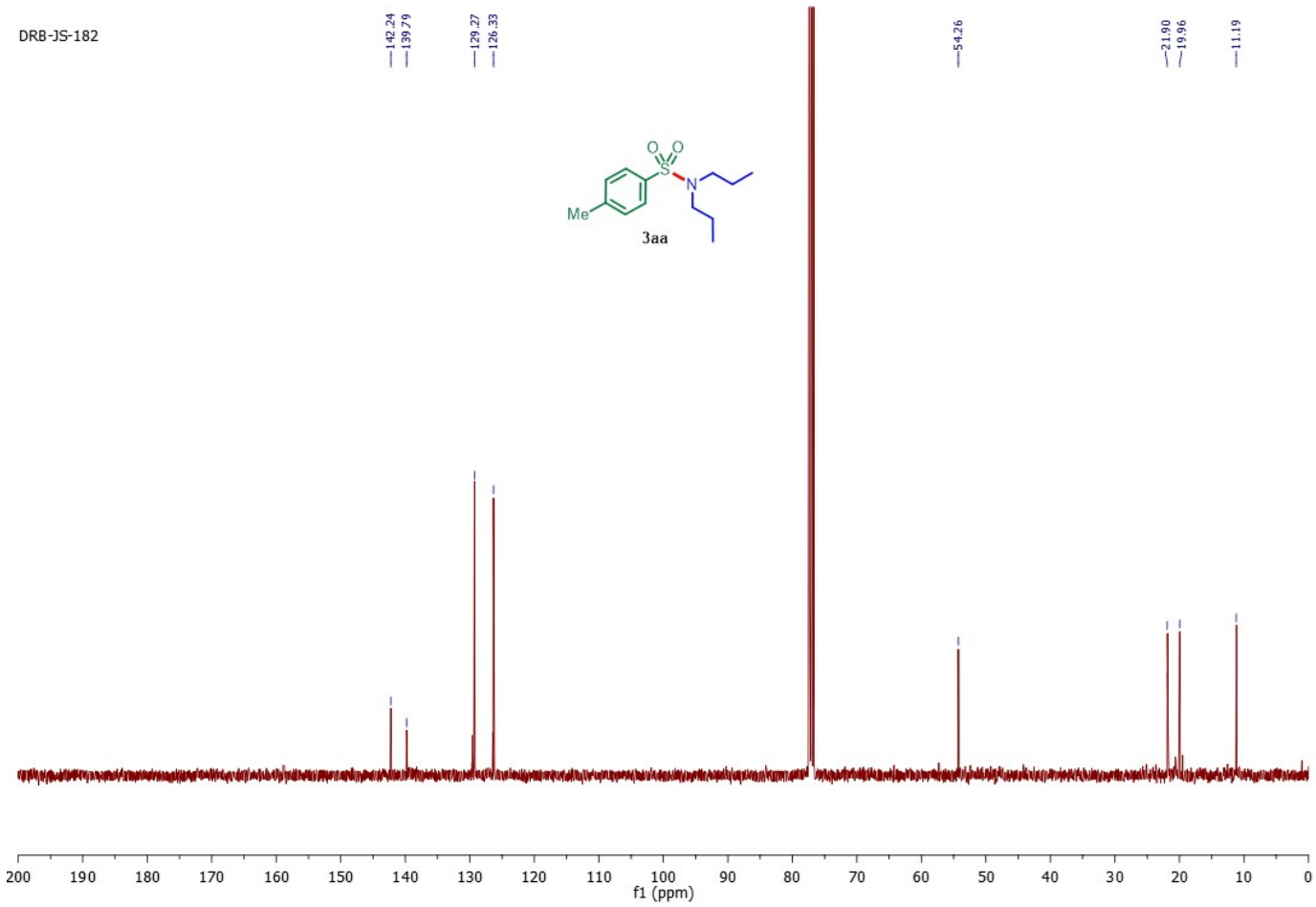

DRB-JS-169
7.63
7.61
7.34
7.32
3.33
3.32
3.31
2.71
2.70
2.69
2.44
Me
3y
1.99
2.00
4.00
4.05
3.09
f1 (ppm)

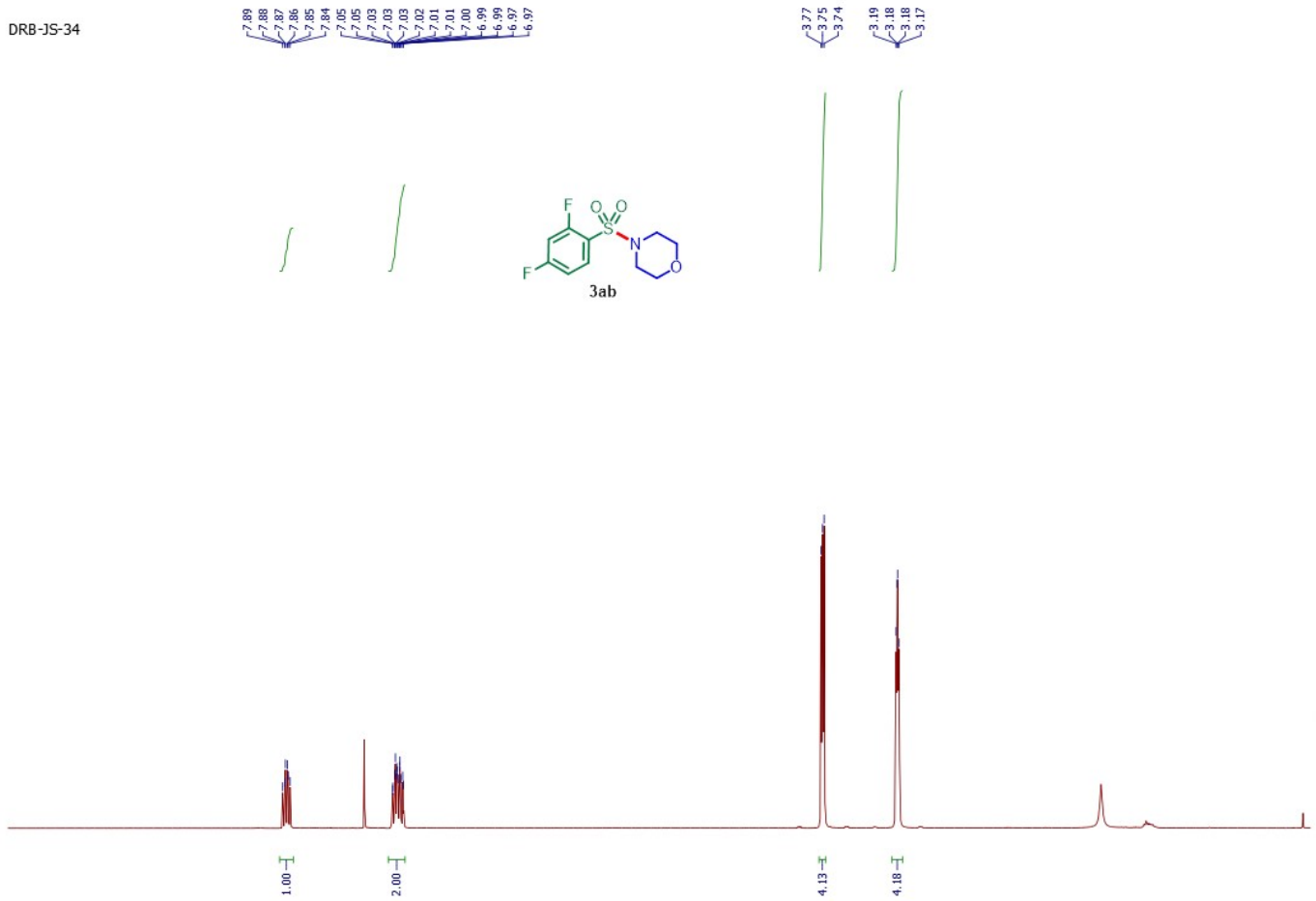

DRB-JS-169
143.78
133.74
129.81
127.44
47.86
27.32
21.53
Me
3y
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

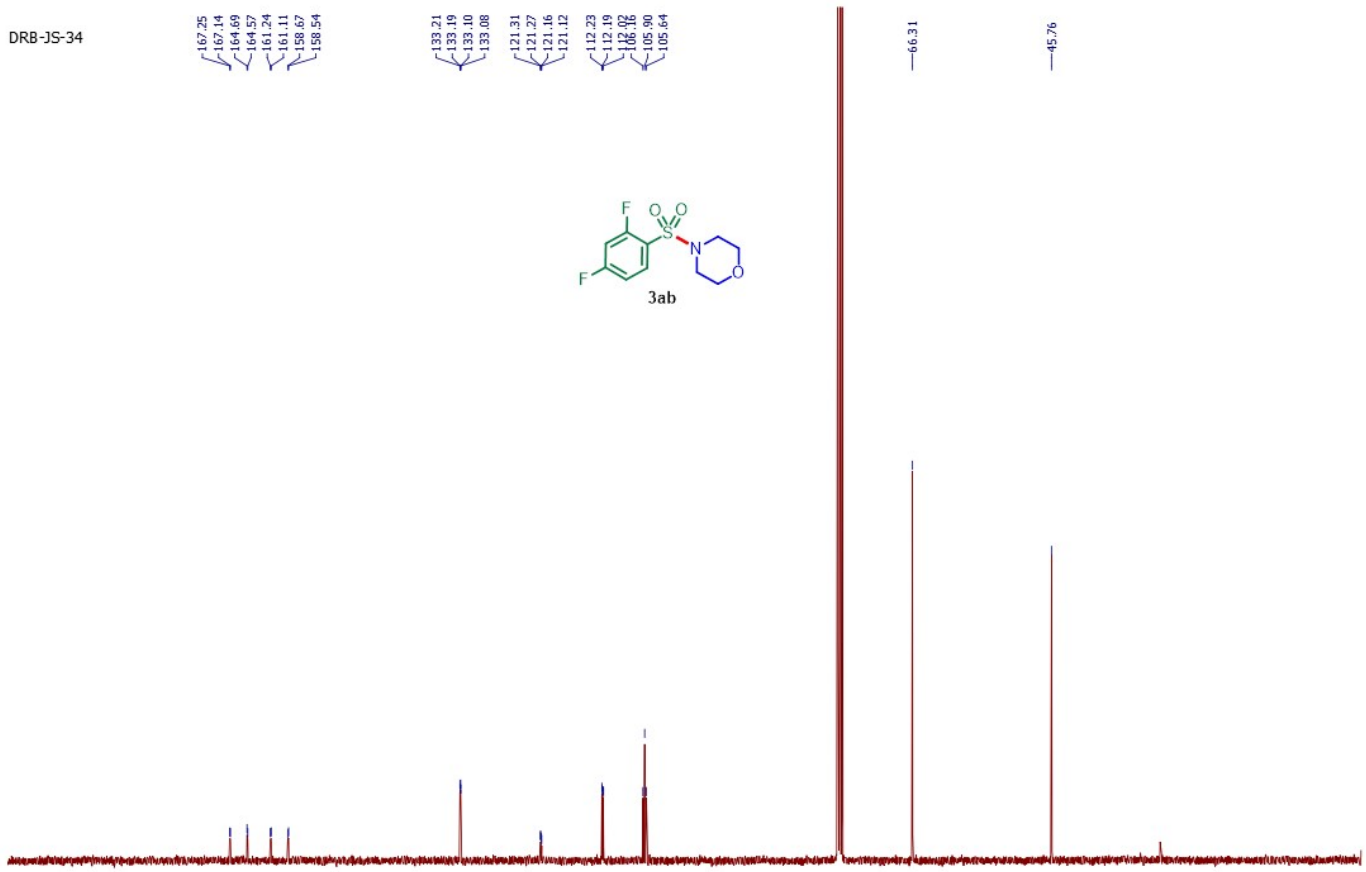

DRB-JS-170
7.70
7.68
7.61
7.59
3.35
3.34
3.33
2.72
2.71
2.70
Br
3z
1.94
2.00
4.05
4.10
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

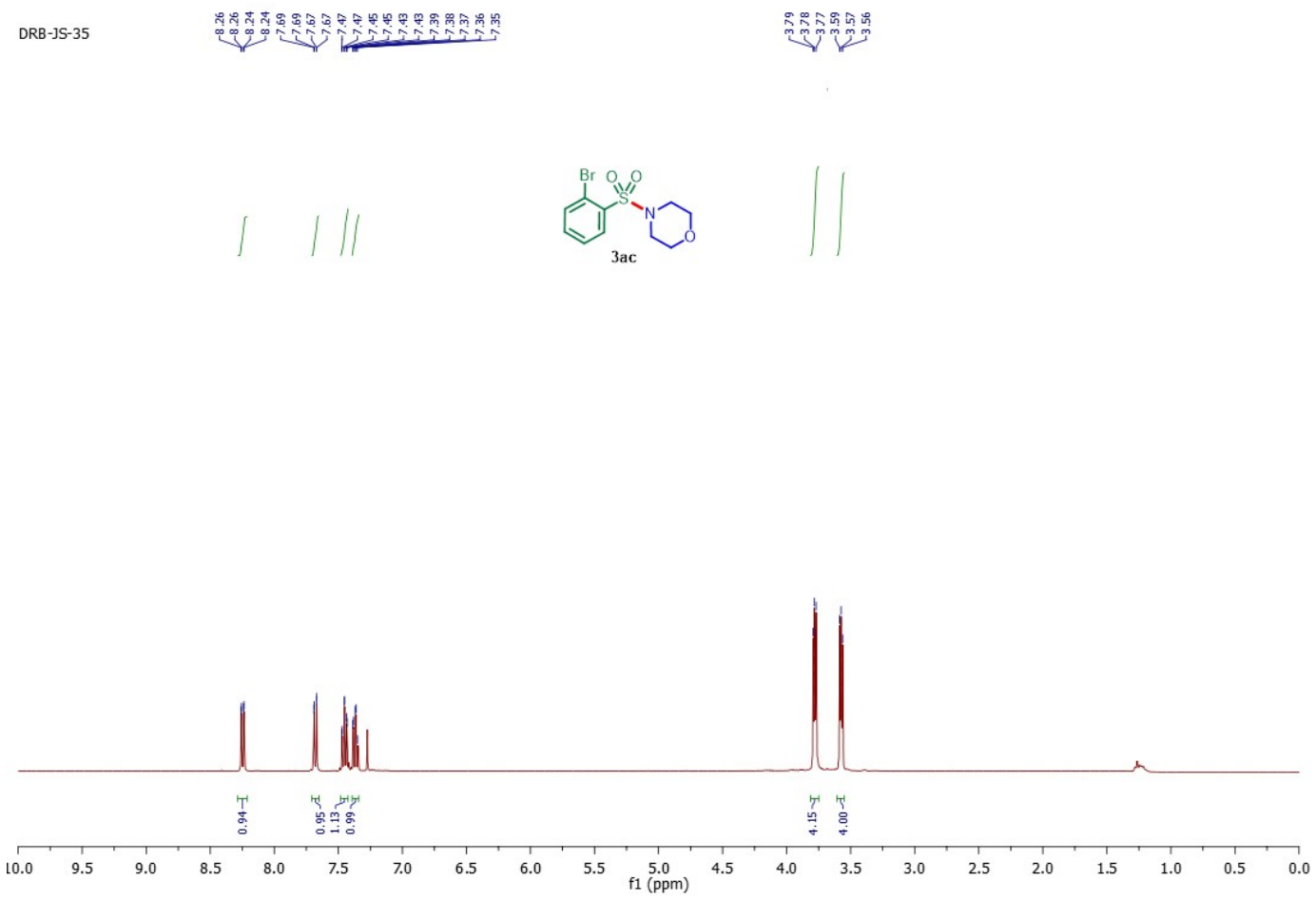

DRB-JS-170
135.96
132.54
128.88
128.02
47.85
27.31
O
O
S
N
S
Br
3z
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

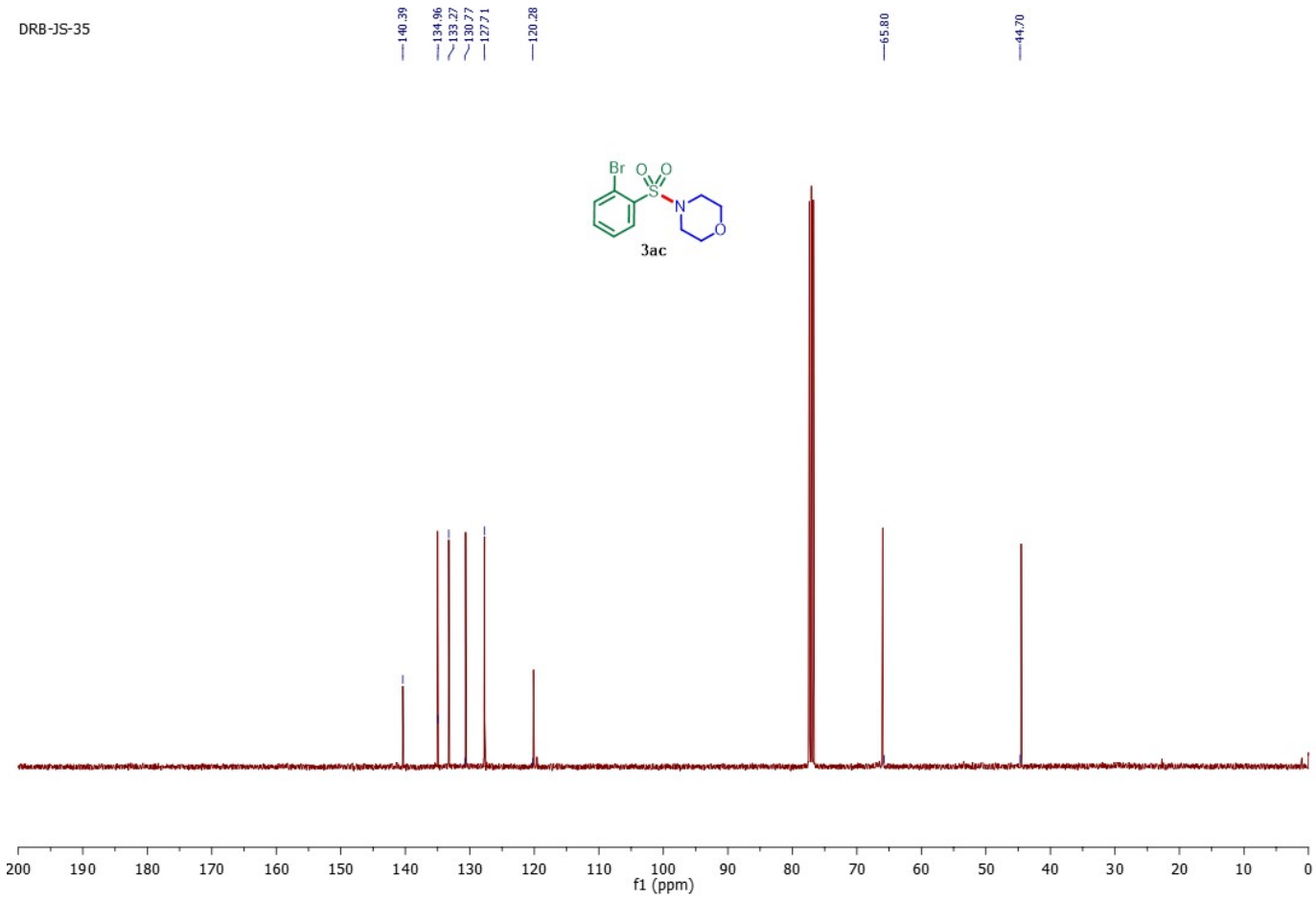

DRB-JS-182
7.76
7.74
7.27
7.26
7.26
7.24
7.24
3.38
3.36
3.34
3.28
3.26
3.25
2.40
1.64
1.62
1.60
1.60
1.58
1.56
0.93
0.91
0.89
0.87
0.86
Me
3aa
2.18
1.94
2.14
2.02
2.81
4.14
6.36
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


DRB-JS-182
142.24
139.79
129.27
126.33
54.26
21.90
19.96
11.19
Me
3aa
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


DRB-JS-34
7.89
7.88
7.87
7.86
7.85
7.84
7.05
7.05
7.03
7.03
7.03
7.02
7.01
7.01
7.00
6.99
6.99
6.97
6.97
3.77
3.75
3.74
3.19
3.18
3.18
3.17
F
O
O
S
N
O
F
3ab
1.00
2.00
4.13
4.18


DRB-JS-34
167.25
167.14
164.69
164.57
161.24
161.11
158.67
158.54
133.21
133.19
133.10
133.08
121.31
121.27
121.16
121.12
112.23
112.19
105.90
105.64
66.31
45.76
F
O
O
S
N
O
F
3ab


DRB-JS-35
8.26
8.26
8.24
8.24
7.69
7.67
7.67
7.47
7.47
7.45
7.45
7.43
7.43
7.39
7.38
7.37
7.36
7.35
3.79
3.78
3.77
3.59
3.57
3.56
Br
O
O
S
N
O
3ac
0.94
0.95
1.13
0.99
4.15
4.00
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


DRB-JS-35
140.39
134.96
133.27
130.77
127.71
120.28
65.80
44.70
Br
O
O
S
N
O
3ac
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)