

Metal-Free Sulfonylation of Arenes with *N*-Fluorobenzenesulfonimide via Cleavage of S-N Bonds: Expeditious Synthesis of Diarylsulfones (Supporting Information)

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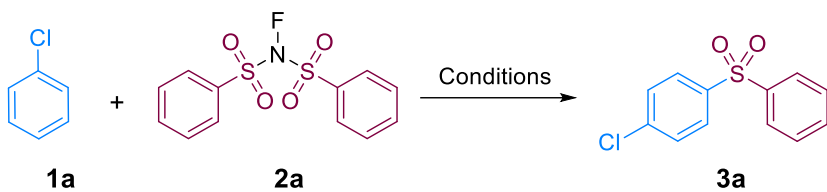
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General remarks

All manipulations were conducted with sealed tubes. ^1H -NMR spectra were recorded on a Bruker AVIII-400 spectrometers. Chemical shifts (in ppm) were calibrated with Chloroform-d. ^{13}C -NMR spectra were obtained by using the same NMR spectrometers and were calibrated with Chloroform-d. Analytical thin-layer chromatography (TLC) was conducted with TLC plates (Silica gel 60 F254, Qingdao Haiyang). Flash column chromatography was performed on silica gel 200-300 mesh with freshly distilled solvents. HRMS data were recorded on a maXis UHR-TOF mass spectrometer. Unless otherwise noted, materials obtained from commercial suppliers were used without further purification.

Table S1. Optimization of reaction conditions (Representative results).^{a, b}



1a + **2a** $\xrightarrow{\text{Conditions}}$ **3a**

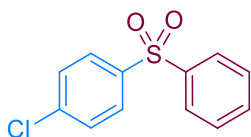
Entry	Acid (1.0 eq)	Time (h)	Yield ^b (%)
1	TfOH	48	73
2	HCl (36%)	48	37
3	HNO ₃ (65%)	48	65
4	AlCl ₃	48	32
5	—	48	—
6	TfOH	30	75
7	TfOH	36	79
8	TfOH	44	75

^a Reaction conditions: **2a** (1.0 mmol) and acid in **1a** (2 mL) at 130 °C under air.

^b Isolated yields.

Experimental procedure and characterization data

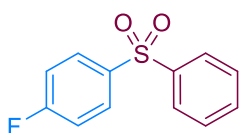
1) 1-Chloro-4-(phenylsulfonyl)benzene (3a) ^[1]



Typical procedure:

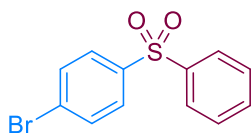
The reaction of chlorobenzene (2.5 mmol, 281.0 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 199.2 mg (79%) of **3a** as solid: ¹H NMR (400 MHz, CDCl₃) δ : 7.93 (dd, J = 7.6, 1.6 Hz, 2H), 7.89 (dd, J = 6.8, 2.0 Hz, 2H), 7.58 (d, J = 7.6 Hz, 1H), 7.52-7.47 (m, 4H). ¹³C NMR (100 MHz, CDCl₃) δ : 141.1, 140.0, 139.8, 133.4, 129.5, 129.4, 129.1, 127.6. MS (70 eV): m/z (%): [M]⁺, 252 (45).

2) 1-Fluoro-4-(phenylsulfonyl)benzene (3b) ^[2]



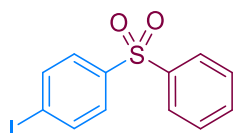
The reaction of fluorobenzene (2.5 mmol, 240.3 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 178.7 mg (76%) of **3b** as solid: ¹H NMR (400 MHz, CDCl₃) δ : 7.99-7.93 (m, 4H), 7.58 (d, J = 6.0 Hz, 1H), 7.57-7.50 (m, 2H), 7.18 (t, J = 8.8 Hz, 2H). ¹³C NMR (100 MHz, CDCl₃) δ : 165.3 (d, ¹ J_{C-F} = 254.7 Hz), 141.3, 137.5 (d, ⁴ J_{C-F} = 2.5 Hz), 133.3, 130.4 (d, ³ J_{C-F} = 9.4 Hz), 129.3, 127.5, 116.5 (d, ² J_{C-F} = 22.4 Hz). ¹⁹F NMR (376 MHz, CDCl₃): δ -104.2 ppm. MS (70 eV): m/z (%): [M]⁺, 236 (50).

3) 1-Bromo-4-(phenylsulfonyl)benzene (3c) ^[2]



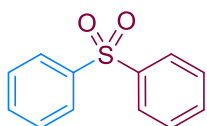
The reaction of bromobenzene (2.5 mmol, 392.5 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 203.3 mg (68%) of **3c** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.92 (t, J = 1.2 Hz, 2H), 7.81 (dd, J = 6.4, 2.0 Hz, 2H), 7.65 (dd, J = 6.8, 2.0 Hz, 2H), 7.58 (d, J = 7.2 Hz, 1H), 7.52 (dd, J = 8.0, 7.2 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3) δ : 141.0, 140.6, 133.4, 132.5, 129.4, 129.1, 128.4, 127.6. MS (70 eV): m/z (%): $[\text{M}]^+$, 296 (35). $[\text{M}]^+$, 296 (35).

4) 1-Iodo-4-(phenylsulfonyl)benzene (**3d**)^[1]



The reaction of iodobenzene (2.5 mmol, 510.0 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 218.0 mg (63%) of **3d** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.93 (dd, J = 7.6, 1.6 Hz, 2H), 7.86 (dd, J = 6.4, 2.0 Hz, 2H), 7.65 (dd, J = 6.8, 2.0 Hz, 2H), 7.58 (d, J = 7.2 Hz, 1H), 7.52 (dd, J = 8.4, 6.8 Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3) δ : 141.2, 141.0, 138.5, 133.4, 129.3, 129.0, 127.5, 101.0. MS (70 eV): m/z (%): $[\text{M}]^+$, 344 (100).

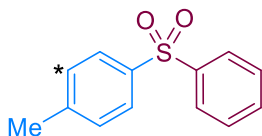
5) Sulfonyldibenzene (**3e**)^[2]



The reaction of benzene (1.25 mmol, 97.6 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.25 mmol, 78.9 mg), trifluoromethanesulfonic (150 mol %, 33 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 158.4 mg (73%) of **3e** as solid. ^1H NMR (400 MHz,

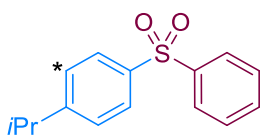
CDCl₃) δ : 7.95 (dd, J = 8.0, 1.2 Hz, 4H), 7.56 (dd, J = 4.0, 1.2 Hz, 2H), 7.51 (dd, J = 8.0, 7.2 Hz, 4H). ¹³C NMR (100 MHz, CDCl₃) δ : 141.4, 133.1, 129.2, 127.6. MS (70 eV): m/z (%): [M]⁺, 218 (25).

6) 1-Methyl-4-(phenylsulfonyl)benzene (**3f**)^[2]



The reaction of toluene (2.5 mmol, 230.4 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 185.8 mg (80%)(1.3:1) of **3f** as solid. (major): ¹H NMR (400 MHz, CDCl₃) δ : 7.93 (t, J = 1.6 Hz, 2H), 7.83 (d, J = 8.4 Hz, 2H), 7.54 (d, J = 7.6 Hz, 1H), 7.49 (t, J = 6.8 Hz, 2H), 7.30 (d, J = 8.4 Hz, 2H), 2.40 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ : 144.1, 141.9, 138.6, 133.0, 129.9, 129.2, 127.7, 127.4, 21.5. (minor): ¹H NMR (400 MHz, CDCl₃) δ : 8.22 (dd, J = 8.0, 1.6 Hz, 1H), 7.86 (t, J = 1.2 Hz, 2H), 7.57 (d, J = 7.2 Hz, 1H), 7.52-7.47 (m, 3H), 7.41 (d, J = 8.0 Hz, 1H), 7.23 (d, J = 7.6 Hz, 1H), 2.44 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ : 141.2, 138.7, 137.9, 133.6, 133.0, 132.6, 129.4, 129.0, 127.6, 126.4, 20.2. MS (70 eV): m/z (%): [M]⁺, 232 (100).

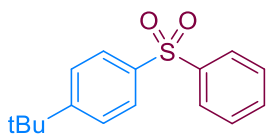
7) 1-Isopropyl-4-(phenylsulfonyl)benzene (**3g**)^[4]



The reaction of cumene (2.5 mmol, 300.4 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 256.2 mg (97%) (4:1) of **3g** as solid. (major): ¹H NMR (400 MHz, CDCl₃) δ : 7.96-7.94 (m, 2H), 7.86 (dd, J = 6.4, 1.6 Hz, 2H), 7.54 (t, J = 0.8 Hz, 1H), 7.52-7.48 (m, 4H), 7.39 (s, 1H), 7.35 (d, J = 8.4 Hz, 2H), 2.94 (d, J = 6.8 Hz, 1H), 1.25 (d, J = 7.6 Hz, 3H), 1.23 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ : 154.8, 141.9, 138.8, 133.0, 129.2, 127.8, 127.6, 127.4, 34.2, 23.6. (minor): ¹H NMR (400 MHz, CDCl₃) δ : 8.21 (dd, J = 8.0, 1.6

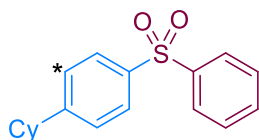
Hz, 1H), 7.86 (dd, $J = 6.8, 1.6$ Hz, 2H), 7.57-7.48 (m, 4H), 7.31 (d, $J = 7.6$ Hz, 1H), 2.86 (dd, $J = 15.2, 7.2$ Hz, 2H), 1.03 (t, $J = 7.6$ Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 144.1, 141.9, 138.4, 133.7, 132.9, 130.9, 129.5, 129.0, 127.4, 126.2, 25.5, 15.0. MS (70 eV): m/z (%): $[\text{M}]^+$, 260 (80).

8) 1-(*tert*-Butyl)-4-(phenylsulfonyl)benzene (**3h**)^[5]



The reaction of *tert*-butylbenzene (2.5 mmol, 335.6 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 251.4 mg (92%) of **3h** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.96-7.94 (m, 2H), 7.86 (dd, $J = 6.4, 1.6$ Hz, 2H), 7.54 (t, $J = 0.8$ Hz, 1H), 7.52-7.48 (m, 4H), 1.31 (s, 9H). ^{13}C NMR (100 MHz, CDCl_3) δ : 154.8, 141.9, 138.8, 133.0, 129.2, 127.8, 127.6, 127.4, 34.2, 23.6. MS (70 eV): m/z (%): $[\text{M}]^+$, 274 (25).

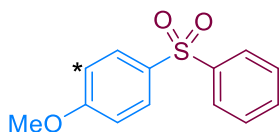
9) 1-Cyclohexyl-4-(phenylsulfonyl)benzene (**3i**)



The reaction of cyclohexylbenzene (2.5 mmol, 400.7 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 80 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 224.8 mg (75%) (4:1) of **3i** as solid. (major): ^1H NMR (400 MHz, CDCl_3) δ : 7.95 (d, $J = 7.2$ Hz, 2H), 7.85 (d, $J = 8.4$ Hz, 2H), 7.57-7.48 (m, 3H), 7.31 (t, $J = 8.4$ Hz, 2H), 2.55 (d, $J = 8.4$ Hz, 1H), 1.84-1.74 (d, $J = 6.4$ Hz, 4H), 1.43-1.22 (m, 6H). ^{13}C NMR (100 MHz, Chloroform- d) δ : 153.9, 141.9, 138.7, 133.0, 129.2, 127.8, 127.7, 127.5, 44.5, 34.0, 26.6, 26.0. (minor): ^1H NMR (400 MHz, CDCl_3) δ : 8.27 (dd, $J = 8.4, 1.6$ Hz, 1H), 7.86 (dd, $J = 7.2, 1.6$ Hz, 2H), 7.56 (t, $J = 4.4$ Hz, 2H), 7.51 (t, $J = 1.6$ Hz, 2H), 7.40-7.36 (m, 2H), 1.67 (d, $J = 6.8$ Hz, 4H), 1.23 (t, $J = 6.4$ Hz, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ : 147.8, 142.5, 138.0, 133.8, 132.8, 129.0, 128.9, 128.8, 127.4, 126.0, 39.5, 33.7, 26.7, 25.9. HRMS

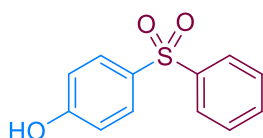
(EI), m/z calcd. for $C_{18}H_{20}O_2S$ $[M]^+$: 300.1184, found: 300.1177.

10) 1-Methoxy-4-(phenylsulfonyl)benzene (**3j**)^[1]



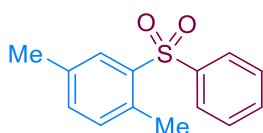
The reaction of anisole (2.5 mmol, 270.4 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 224.0 mg (90%) (1.5:1) of **3j** as solid. (major): 1H NMR (400 MHz, $CDCl_3$) δ : 7.92 (t, J = 6.8 Hz, 2H), 7.88 (dd, J = 7.2, 2.0 Hz, 2H), 7.54 (d, J = 7.2 Hz, 1H), 7.50 (t, J = 1.6 Hz, 2H), 6.97 (dd, J = 7.2, 2.4 Hz, 2H), 3.85 (s, 3H). ^{13}C NMR (100 MHz, Chloroform-*d*) δ : 163.3, 142.3, 133.0, 132.8, 129.9, 129.2, 127.3, 114.5, 55.6. (minor): 1H NMR (400 MHz, $CDCl_3$) δ : 8.17 (dd, J = 7.6, 1.6 Hz, 1H), 7.97 (t, J = 1.2 Hz, 2H), 7.56 (dd, J = 7.6, 2.0 Hz, 2H), 7.49 (t, J = 7.2 Hz, 2H), 7.12 (d, J = 7.6 Hz, 1H), 6.91 (d, J = 8.4 Hz, 1H), 3.76 (s, 3H). ^{13}C NMR (100 MHz, Chloroform-*d*) δ : 157.0, 141.1, 135.5, 132.9, 129.8, 128.9, 128.4, 128.3, 120.5, 112.4, 55.8. MS(70 eV): m/z (%): $[M]^+$, 248 (60).

11) 4-(Phenylsulfonyl)phenol (**3k**)^[3]



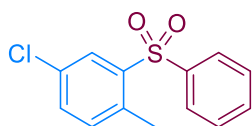
The reaction of phenol (0.8 mmol, 75.3 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.4 mmol, 126.4 mg), trifluoromethanesulfonic (500 mol %, 177 μ L), was carried out in 1.0 mL trifluoroacetic acid at 100 °C under air for 12 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 29.2 mg (16%) of **3k** as solid. 1H NMR (400 MHz, $CDCl_3$) δ : 7.94-7.90 (m, 2H), 7.86-7.82 (m, 2H), 7.57-7.47 (m, 3H), 6.93-6.89 (m, 2H), 5.62 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ : 160.6, 141.8, 133.7, 132.07, 130.0, 129.3, 127.1, 116.2. HRMS: m/z $[M+Na]^+$: calcd for $C_{12}H_{10}NaO_3S$: 257.0243; found: 257.0241.

12) 1,4-Dimethyl-2-(phenylsulfonyl)benzene (**3m**)^[1]



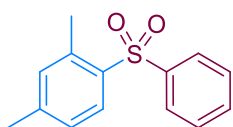
The reaction of *p*-xylene (2.5 mmol, 268.1 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 152.3 mg (62%) of **3m** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 8.05 (d, $J = 0.8$ Hz, 1H), 7.86 (t, $J = 1.6$ Hz, 2H), 7.56 (d, $J = 7.2$ Hz, 1H), 7.50 (dd, $J = 8.0, 6.8$ Hz, 2H), 7.28 (d, $J = 0.8$ Hz, 1H), 7.11 (d, $J = 8.0$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3) δ : 141.2, 138.2, 136.3, 134.6, 134.3, 132.8, 132.5, 129.6, 128.9, 127.4, 20.8, 19.6. MS (70 eV): m/z (%): $[\text{M}]^+$, 246 (90).

13) 1-Chloro-4-methyl-2-(phenylsulfonyl)benzene (**3n**)



The reaction of 1-chloro-4-methylbenzene (2.5 mmol, 316.5 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 170.0 mg (64%) of **3n** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 8.22 (d, $J = 2.0$ Hz, 1H), 7.88 (t, $J = 1.6$ Hz, 2H), 7.60 (d, $J = 7.2$ Hz, 1H), 7.53 (dd, $J = 8.0, 6.8$ Hz, 2H), 7.45 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.17 (d, $J = 8.0$ Hz, 1H), 2.40 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 140.5, 140.3, 136.3, 134.0, 133.5, 133.4, 132.5, 129.2, 129.1, 127.8, 19.6. HRMS (EI), m/z calcd. for $\text{C}_{13}\text{H}_{11}\text{ClO}_2\text{S}$ $[\text{M}]^+$: 266.0168, found: 266.0174.

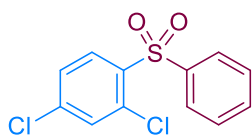
14) 2,4-Dimethyl-1-(phenylsulfonyl)benzene (**3o**)^[6]



The reaction of *m*-xylene (2.5 mmol, 265.0 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic

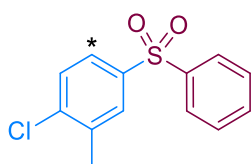
acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 208.0 mg (84%) of **3o** as solid. ¹H NMR (400 MHz, CDCl₃) δ: 8.11 (d, *J* = 0.8 Hz, 1H), 7.84 (t, *J* = 1.6 Hz, 2H), 7.60 (d, *J* = 7.6 Hz, 1H), 7.51-7.47 (m, 2H), 7.20 (d, *J* = 8.0 Hz, 1H), 7.04 (s, 1H), 2.39 (s, 3H), 2.37 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ: 144.4, 141.5, 137.7, 135.8, 133.3, 132.8, 129.6, 128.9, 127.5, 127.0, 21.3, 20.1. MS (70 eV): *m/z* (%): [M]⁺, 246 (60).

15) 2,4-Dichloro-1-(phenylsulfonyl)benzene (**3p**)



The reaction of 1,3-dichlorobenzene (2.5 mmol, 371.2 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 169.6 mg (59%) of **3p** as solid. ¹H NMR (400 MHz, CDCl₃) δ: 8.31 (d, *J* = 8.8 Hz, 1H), 7.94 (dd, *J* = 7.2, 5.6 Hz, 2H), 7.62 (d, *J* = 7.6 Hz, 1H), 7.53 (t, *J* = 7.2 Hz, 2H), 7.49-7.45 (m, 2H). ¹³C NMR (100 MHz, CDCl₃) δ: 140.6, 139.6, 137.0, 133.9, 133.7, 132.0, 131.8, 129.0, 128.5, 127.6. HRMS (EI), *m/z* calcd. for C₁₂H₈Cl₂O₂S [M]⁺ : 285.9622, found: 285.9625.

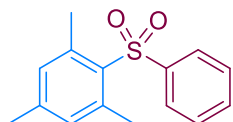
16) 2-Chloro-1-methyl-4-(phenylsulfonyl)benzene (**3q**)^[7]



The reaction of 1-chloro-2-methylbenzene (2.5 mmol, 316.5 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 225.6 mg (85%) (3.5:1) of **3q** as solid. (major): ¹H NMR (400 MHz, CDCl₃) δ: 7.96 (m, , 2H), 7.81 (d, *J* = 2.0 Hz, 1H), 7.71 (dd, *J* = 8.4, 2.0 Hz, 1H), 7.58 (d, *J* = 6.0 Hz, 1H), 7.56-7.49 (m, 2H), 7.45 (d, *J* = 8.4 Hz, 1H), 2.40

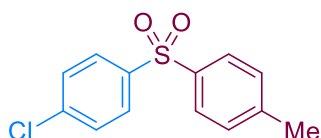
(d, $J = 2.4$ Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 141.2, 139.9, 139.7, 137.7, 133.3, 129.9, 129.7, 129.3, 127.6, 126.3, 20.1. (minor): ^1H NMR (400 MHz, CDCl_3) δ : 7.95 – 7.91 (m, 3H), 7.72 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.59–7.57 (m, 1H), 7.54–7.50 (m, 2H), 7.36 (d, $J = 8.0$ Hz, 1H), 2.41 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 142.2, 141.2, 140.5, 135.4, 133.4, 131.7, 129.4, 128.2, 127.6, 125.7, 20.4. HRMS (EI), m/z calcd. for $\text{C}_{13}\text{H}_{11}\text{ClO}_2\text{S}$ $[\text{M}]^+$: 266.0168, found: 266.0173.

17) 1,3,5-Trimethyl-2-(phenylsulfonyl)benzene (3r) ^[1]



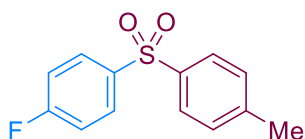
The reaction of mesitylene (2.5 mmol, 300.5 mg), *N*-fluorobenzenesulfonimide (**2a**) (0.5 mmol, 157.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 80 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 183.2 mg (70%) of **3s** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.78 (t, $J = 1.2$, Hz, 2H), 7.54 (s, 1H), 7.47 (t, $J = 1.6$, Hz, 2H), 6.94 (s, 2H), 2.59 (s, 6H), 2.30 (s, 3H) ppm. ^{13}C NMR (100 MHz, CDCl_3) δ : 143.4, 143.4, 140.0, 133.7, 132.5, 132.2, 128.8, 126.2, 22.8, 21.0 ppm. MS (70 eV): m/z (%): $[\text{M}]^+$, 260 (40).

18) 1-Chloro-4-tosylbenzene (4a) ^[8]



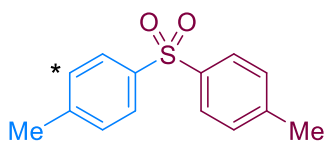
The reaction of chlorobenzene (2.5 mmol, 281.0 mg), *N*-fluoro-4-methyl-*N*-tosylbenzenesulfonamide (**2b**) (0.5 mmol, 171.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 204.3 mg (76%) of **4a** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.86 (dd, $J = 19.2, 6.8$ Hz, 4H), 7.46 (d, $J = 6.8$ Hz, 2H), 7.31 (d, $J = 8.0$ Hz, 2H), 2.41 (s, 3H) ppm. ^{13}C NMR (100 MHz, CDCl_3) δ : 144.1, 141.9, 133.0, 129.9, 129.2, 127.7, 127.5, 21.6. MS (70 eV): m/z (%): $[\text{M}]^+$, 266 (60).

19) 1-Fluoro-4-tosylbenzene (4b) ^[2]



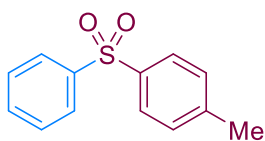
The reaction of fluorobenzene (2.5 mmol, 242.7 mg), *N*-fluoro-4-methyl-*N*-tosylbenzenesulfonamide (**4b**) (0.5 mmol, 171.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 204.7 mg (82%) of **4b** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 7.94 (dd, J = 8.8, 5.6 Hz, 2H), 7.81 (d, J = 8.4 Hz, 2H), 7.29 (d, J = 8.0 Hz, 2H), 7.16 (dd, J = 8.8, 7.6 Hz, 2H), 2.40 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 165.3 (d, $^1J_{\text{C-F}}$ = 253.6 Hz), 144.3, 138.4, 138.0 (d, $^4J_{\text{C-F}}$ = 2.5 Hz), 130.3 (d, $^3J_{\text{C-F}}$ = 9.4 Hz), 130.0, 127.6, 116.5 (d, $^2J_{\text{C-F}}$ = 21.9 Hz), 21.6. ^{19}F NMR (376 MHz, CDCl_3): δ -104.2 ppm. MS (70 eV): m/z (%): $[\text{M}]^+$, 250 (100).

20) 4,4'-Sulfonylbis(methylbenzene) (**4c**)^[8]



The reaction of toluene (2.5 mmol, 230.4 mg), *N*-fluoro-4-methyl-*N*-tosylbenzenesulfonamide (**2b**) (0.5 mmol, 171.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 201.7 mg (82%) (3:1) of **4c** as solid. (major): ^1H NMR (400 MHz, CDCl_3) δ : 7.81 (d, J = 7.6 Hz, 4H), 7.26 (d, J = 8.0 Hz, 4H), 2.35 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ : 143.8, 138.8, 129.7, 127.3, 21.3. (minor): ^1H NMR (400 MHz, CDCl_3) δ : 8.19 (dd, J = 7.6, 1.2 Hz, 1H), 7.75 (d, J = 8.0 Hz, 2H), 7.47 (td, J = 7.5, 1.6 Hz, 1H), 7.42 - 7.33 (m, 1H), 7.32 - 7.27 (m, 2H), 7.22 (dd, J = 7.4, 1.4 Hz, 1H), 2.44 (s, 3H), 2.41 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ : 143.9, 139.0, 138.1, 133.4, 132.5, 129.6, 129.2, 127.7, 126.4, 21.5, 20.1. HRMS: m/z $[\text{M}+\text{Na}]^+$: calcd for $\text{C}_{14}\text{H}_{14}\text{NaO}_2\text{S}$: 269.0607; found: 269.0605.

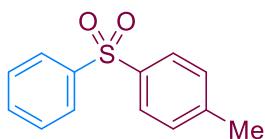
21) 1-Methyl-4-(phenylsulfonyl)benzene (**3f**)^[2]



The reaction of benzene (2.5 mmol, 195.3 mg), *N*-fluoro-4-methyl-*N*-tosylbenzenesulfonamide (**2b**) (0.5 mmol, 171.7 mg), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL

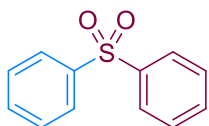
trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 80.1 mg (34%) of **3f** as solid. ¹H NMR (400 MHz, CDCl₃) δ: 7.93 (t, *J* = 1.6 Hz, 2H), 7.83 (d, *J* = 8.4 Hz, 2H), 7.54 (d, *J* = 7.6 Hz, 2H), 7.49 (t, *J* = 6.8 Hz, 2H), 7.30 (d, *J* = 8.4 Hz, 2H), 2.40 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ: 144.1, 141.9, 138.6, 133.0, 129.9, 129.2, 127.7, 127.4, 21.5. MS (70 eV): *m/z* (%): [M]⁺, 232 (100).

22) 1-Methyl-4-(phenylsulfonyl)benzene (**3f**)^[2]



The reaction of benzene (2.5 mmol, 195.3 mg), *N*-fluoro-4-methyl-*N*-(phenylsulfonyl)benzenesulfonamide (**2c**) (0.5 mmol, 164.7 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 100 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 70.1 mg (60%) of **3f** as solid. ¹H NMR (400 MHz, CDCl₃) δ: 7.93 (t, *J* = 1.6 Hz, 2H), 7.83 (d, *J* = 8.4 Hz, 2H), 7.54 (d, *J* = 7.6 Hz, 2H), 7.49 (t, *J* = 6.8 Hz, 2H), 7.30 (d, *J* = 8.4 Hz, 2H), 2.40 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ: 144.1, 141.9, 138.6, 133.0, 129.9, 129.2, 127.7, 127.4, 21.5. MS (70 eV): *m/z* (%): [M]⁺, 232 (100).

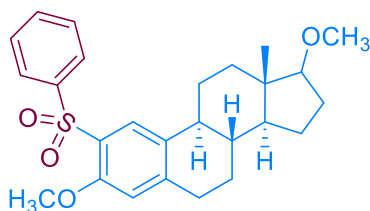
23) Sulfonyldibenzene (**3e**)^[2]



The reaction of benzene (2.5 mmol, 195.3 mg), *N*-fluoro-4-nitro-*N*-(phenylsulfonyl)benzenesulfonamide (**2d**) (0.5 mmol, 180.2 mg), trifluoromethanesulfonic (150 mol %, 66 μL), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 40.1 mg (35%) of **3e** as solid. ¹H NMR (400 MHz, CDCl₃) δ: 7.95 (dd, *J* = 8.0, 1.2 Hz, 4H), 7.56 (dd, *J* = 4.0, 1.2 Hz, 2H), 7.51 (dd, *J* = 8.0, 7.2 Hz, 4H). ¹³C NMR (100 MHz, CDCl₃) δ: 141.4, 133.1, 129.2, 127.6. MS (70 eV): *m/z* (%): [M]⁺, 218 (25).

Structural modification of β -estradiol derivative **5**

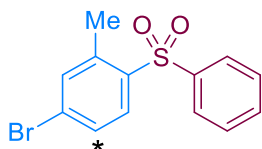
(8*R*,9*S*,13*S*,14*S*)-3,17-Dimethoxy-13-methyl-2-(phenylsulfonyl)-7,8,9,11,12,13,14,15,16,17-decahydro-6H-cyclopenta[*a*]phenanthrene (**5**)



The reaction of (8*R*,9*S*,13*S*,14*S*)-3,17-dimethoxy-13-methyl-7,8,9,11,12,13,14,15,16,17-decahydro-6H-cyclopenta[*a*]phenanthrene (0.25 mmol, 75.0 mg), *N*-fluoro-*N*-(phenylsulfonyl)benzenesulfonamide (**2a**) (0.0625 mmol, 19.8 mg), trifluoromethanesulfonic (600 mol %, 33 μ L), was carried out in 0.5 mL trifluoroacetic acid at 30 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 30.2 mg (55%) of **5** as solid. ^1H NMR (400 MHz, CDCl_3) δ : 8.04 (s, 1H), 7.97-7.95 (m, 2H), 7.56-7.52 (m, 1H), 7.46 (dd, $J = 14.8, 7.6$ Hz, 2H), 6.58 (s, 1H), 3.69 (s, 3H), 3.47 (s, 3H), 3.32 (dd, $J = 16.8, 8.4$ Hz, 1H), 2.85 (dd, $J = 8.8, 4.2$ Hz, 2H), 2.42 (dt, $J = 12.9, 3.5$ Hz, 1H), 2.27- 2.14 (m, 1H), 2.08 (ddd, $J = 13.8, 9.5, 3.5$ Hz, 2H), 1.88 (ddt, $J = 12.3, 4.4, 2.5$ Hz, 1H), 1.75-1.22 (m, 8H). ^{13}C NMR (100 MHz, CDCl_3) δ : 154.7, 145.4, 141.9, 133.1, 132.6, 128.4, 128.2, 126.9, 126.1, 112.7, 90.6, 76.7, 57.9, 55.8, 50.2, 43.7, 43.2, 38.3, 37.8, 30.1, 27.7, 26.7, 26.4, 23.0, 11.5. HRMS (EI), m/z calcd. for $\text{C}_{26}\text{H}_{32}\text{O}_4\text{S}$ $[\text{M}]^+$: 440.2021, found: 440.2028.

Synthesis of the intermediate **6** of an inhibitor of Farnesyl-protein transferase

4-Bromo-2-methyl-1-(phenylsulfonyl)benzene (**6**)^[9]

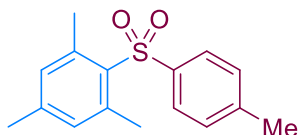


The reaction of 1-bromo-3-methylbenzene (1.25 mmol, 427.6 mg), *N*-fluoro-*N*-(phenylsulfonyl)benzenesulfonamide (**2a**) (0.5 mmol, 158.0 mg), trifluoromethanesulfonic (0.75 mmol, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 196.0 mg (63%) of **6** as solid. (major): ^1H NMR (400

MHz, CDCl₃) δ : 8.08 (d, J = 8.4 Hz, 1H), 7.85 (m, 2H), 7.59 (d, J = 3.6 Hz, 1H), 7.54-7.50 (m, 3H), 7.40 (d, J = 1.2 Hz, 1H), 2.41 (s, 3H) ppm. ¹³C NMR (100 MHz, CDCl₃) δ : 140.8, 139.9, 138.0, 135.4, 133.2, 130.9, 129.7, 129.1, 128.5, 127.6, 20.0 ppm. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 56.1 mg (18%) of **6** (minor) as solid. (minor): ¹H NMR (400 MHz, CDCl₃) δ : 8.29 (d, J = 8.0 Hz, 1H), 7.94 (d, J = 7.6 Hz, 2H), 7.58 (d, J = 3.6 Hz, 1H), 7.52-7.48 (m, 3H), 7.33 (d, J = 8.0 Hz, 1H), 2.39 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ : 146.0, 140.2, 136.9, 136.0, 133.2, 131.3, 128.7, 128.5, 128.4, 120.9, 21.0. HRMS: m/z [M+Na]⁺: calcd for C₁₃H₁₁BrNaO₂S : 332.9555; found: 332.9553.

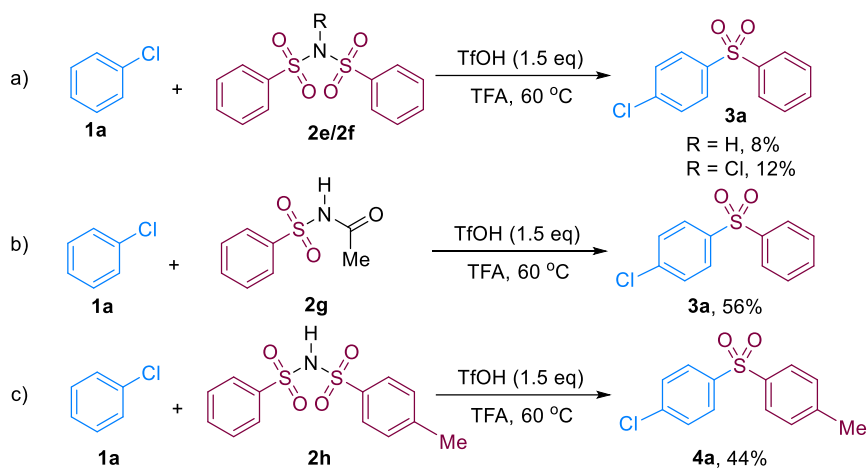
Gram-scale synthesis of EPAC2 antagonist **8**

1,3,5-Trimethyl-2-tosylbenzene (**8**)^[6]



The reaction of mesitylene (23.1 mmol, 2.78 g), *N*-fluoro-4-methyl-*N*-tosylbenzenesulfonamide (**2b**) (4.62 mmol, 1.59 g), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL CH₃CN at 130 °C under air for 36 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford 2.38 g (94%) of **8** as solid. ¹H NMR (400 MHz, CDCl₃) δ : 7.67 (d, J = 8.4 Hz, 2H), 7.26 (t, J = 5.6 Hz, 2H), 6.93 (s, 2H), 2.59 (s, 6H), 2.40 (s, 3H), 2.29 (s, 3H) ppm. ¹³C NMR (100 MHz, CDCl₃) δ : 143.3, 143.1, 140.5, 139.9, 134.1, 132.1, 129.4, 126.2, 22.8, 21.5, 21.0 ppm. HRMS: m/z [M+Na]⁺: calcd for C₁₆H₁₈NaO₂S : 297.0920; found: 297.0917.

Scheme S1 Control Experiments



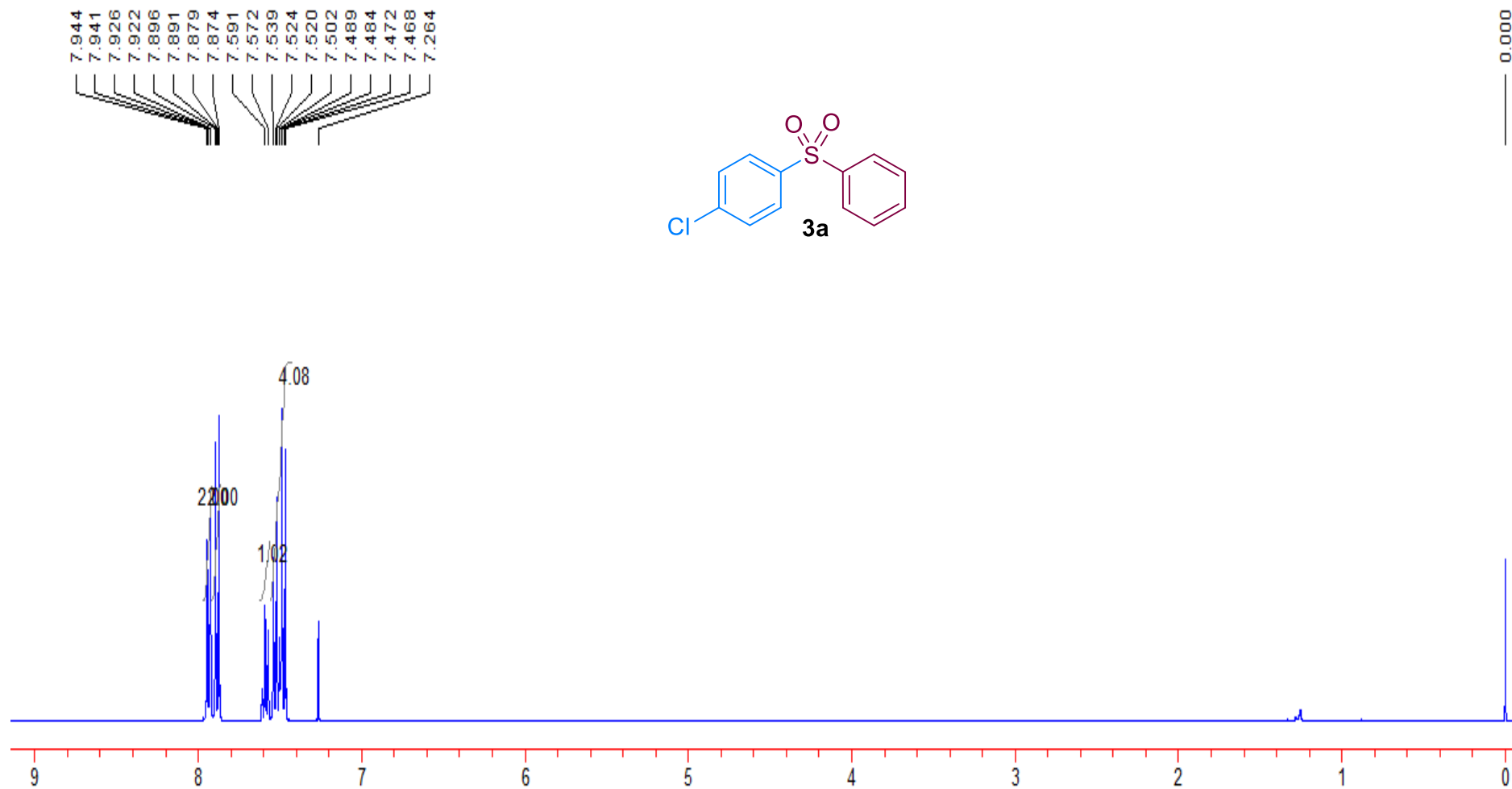
Typical procedure:

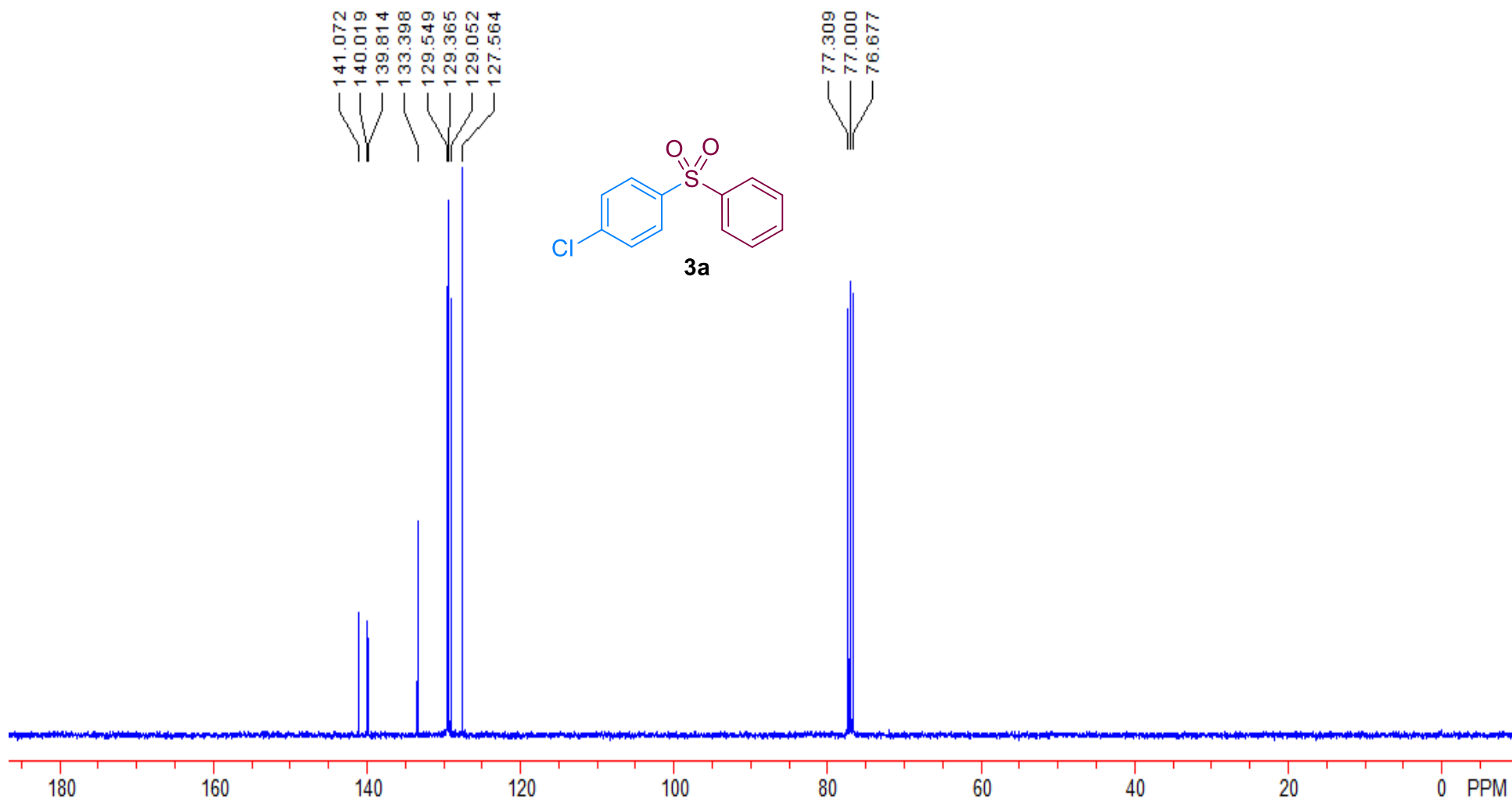
The reaction of chlorobenzene **1a** (2.5 mmol, 281.0 mg), **2** (0.5 mmol), trifluoromethanesulfonic (150 mol %, 66 μ L), was carried out in 0.5 mL trifluoroacetic acid at 60 °C under air for 24 h as monitored by TLC. The resulting mixture was concentrated and purified by flash chromatography on silica gel to afford the product.

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- [9] H. Zhu, Y. Shen, D. Wen, Z.-G. Le, T. Tu. *Org. Lett.* **2019**, *21*, 974.

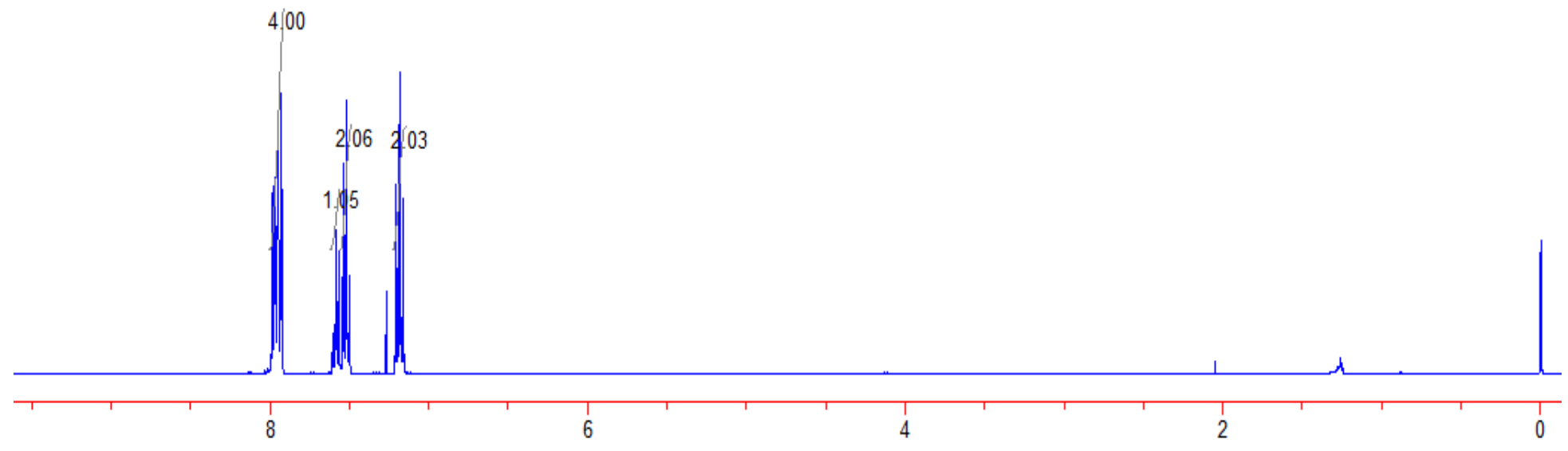
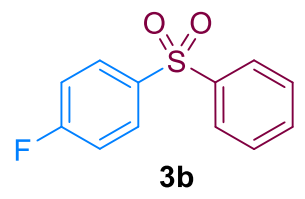
^1H NMR, ^{13}C NMR and ^{19}F NMR spectra for products

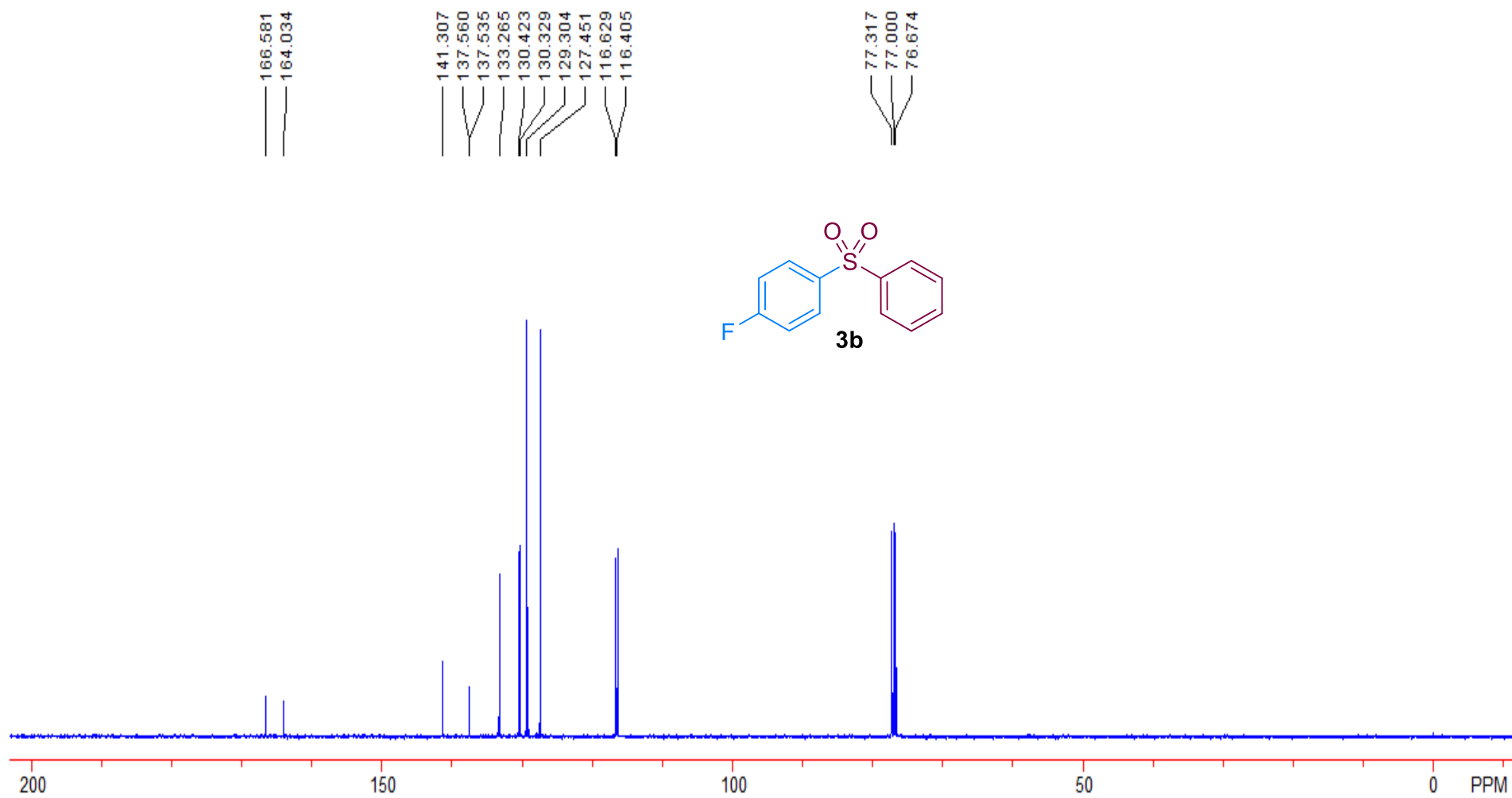


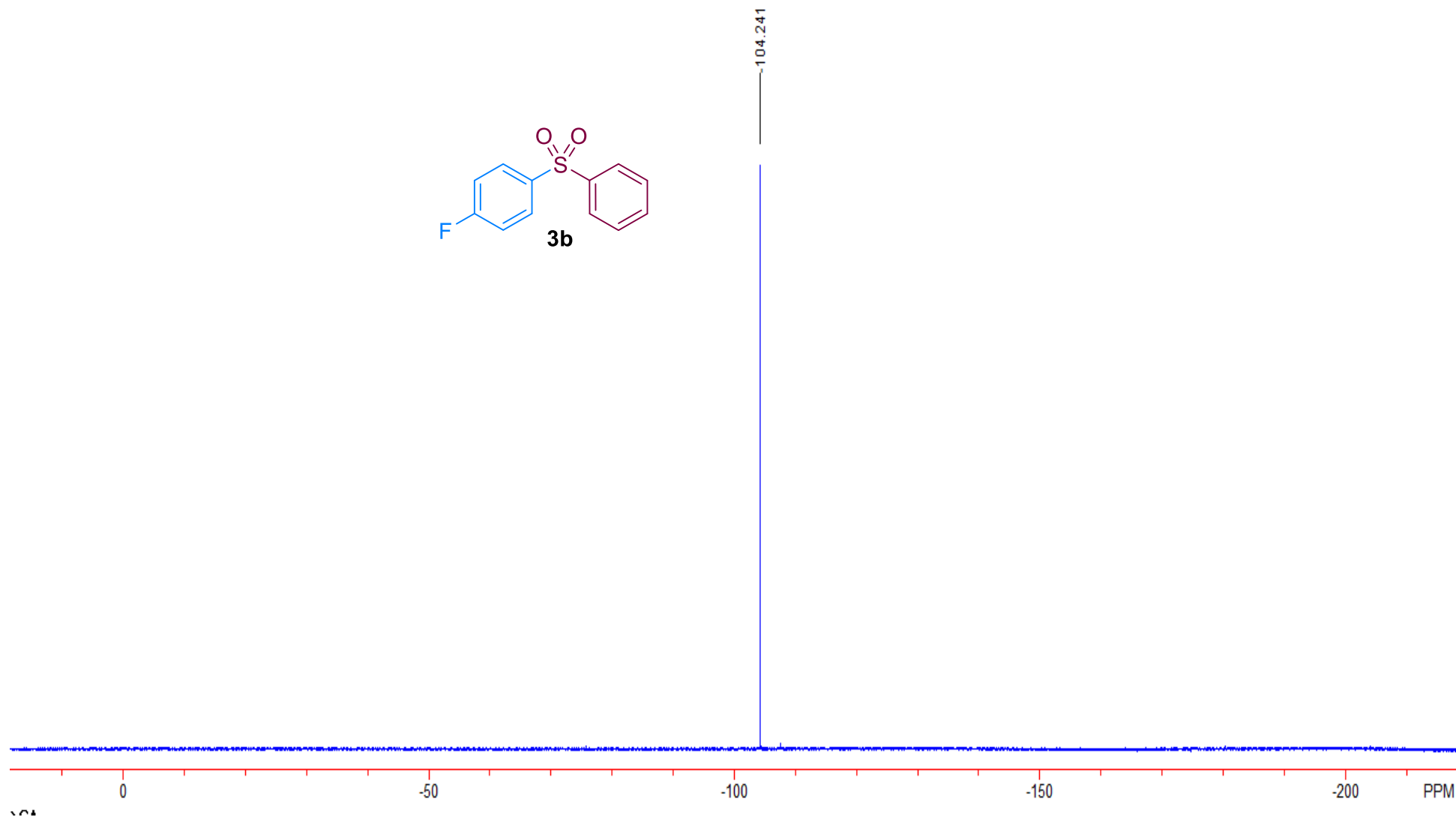
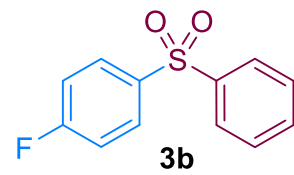


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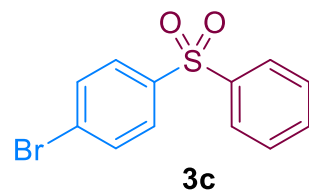
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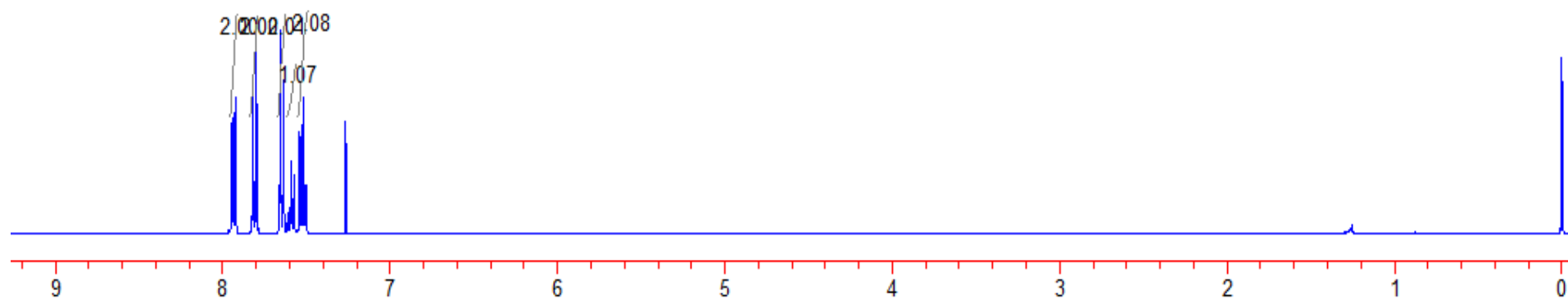


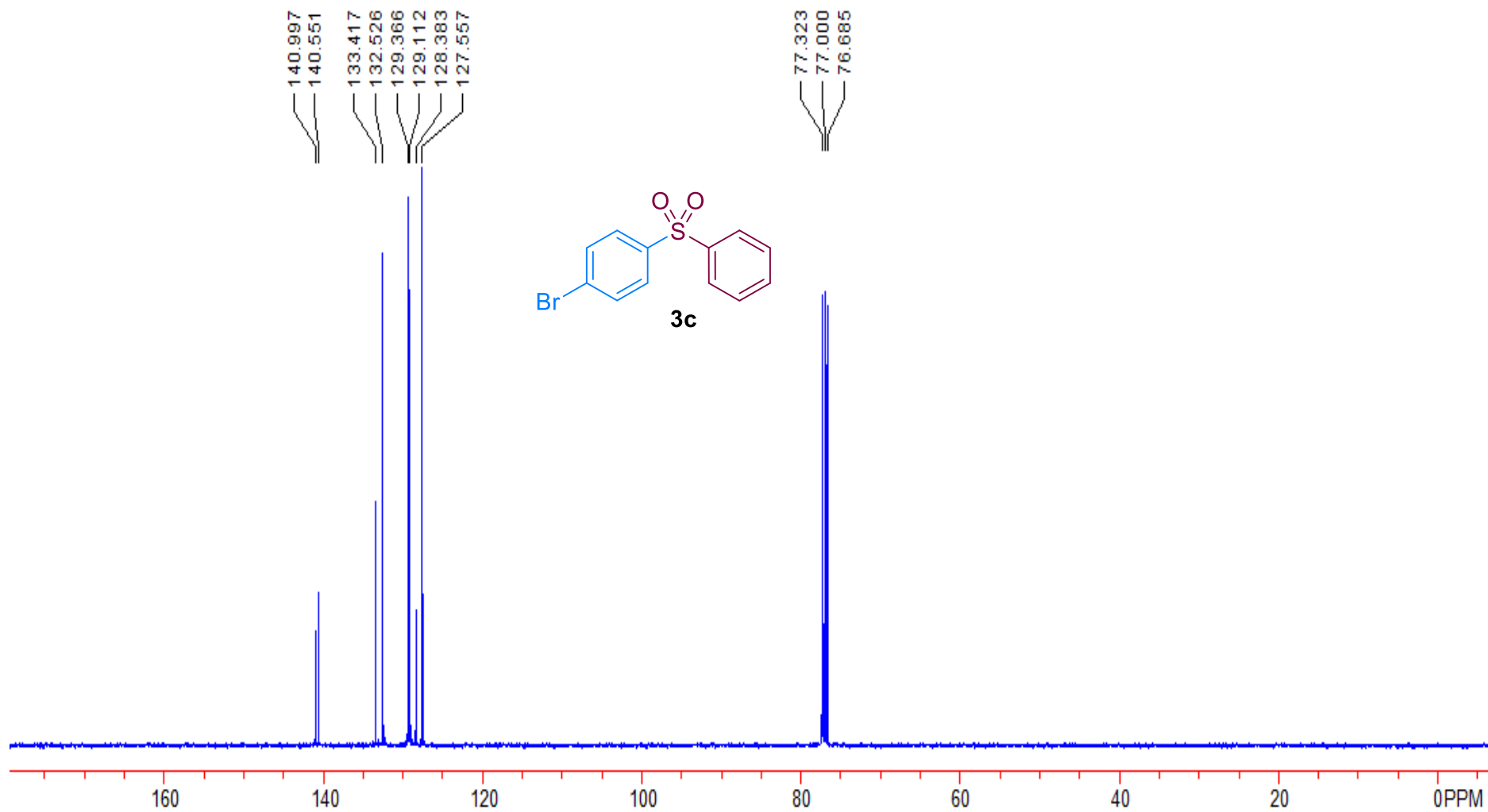


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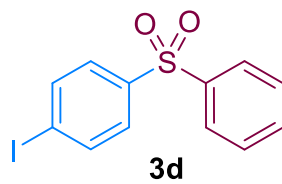


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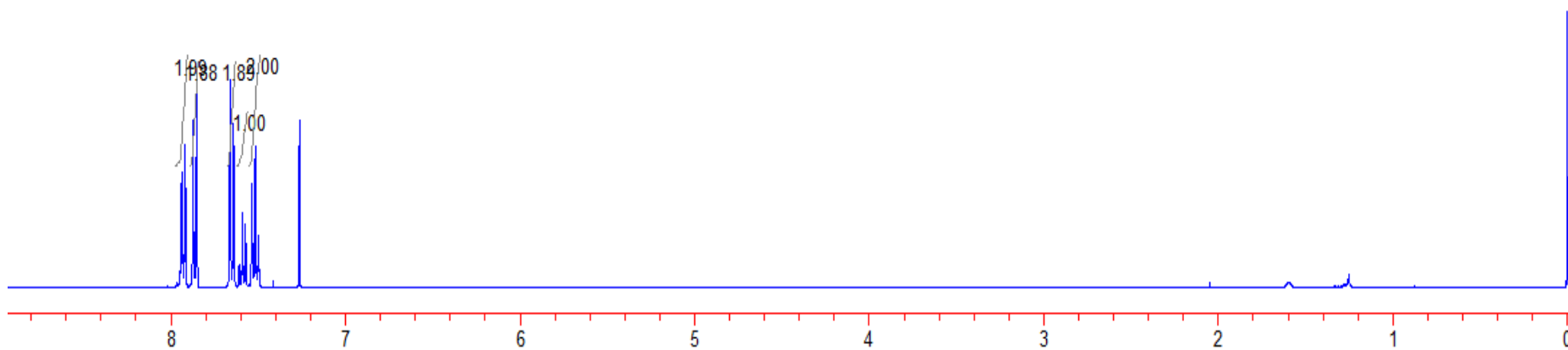


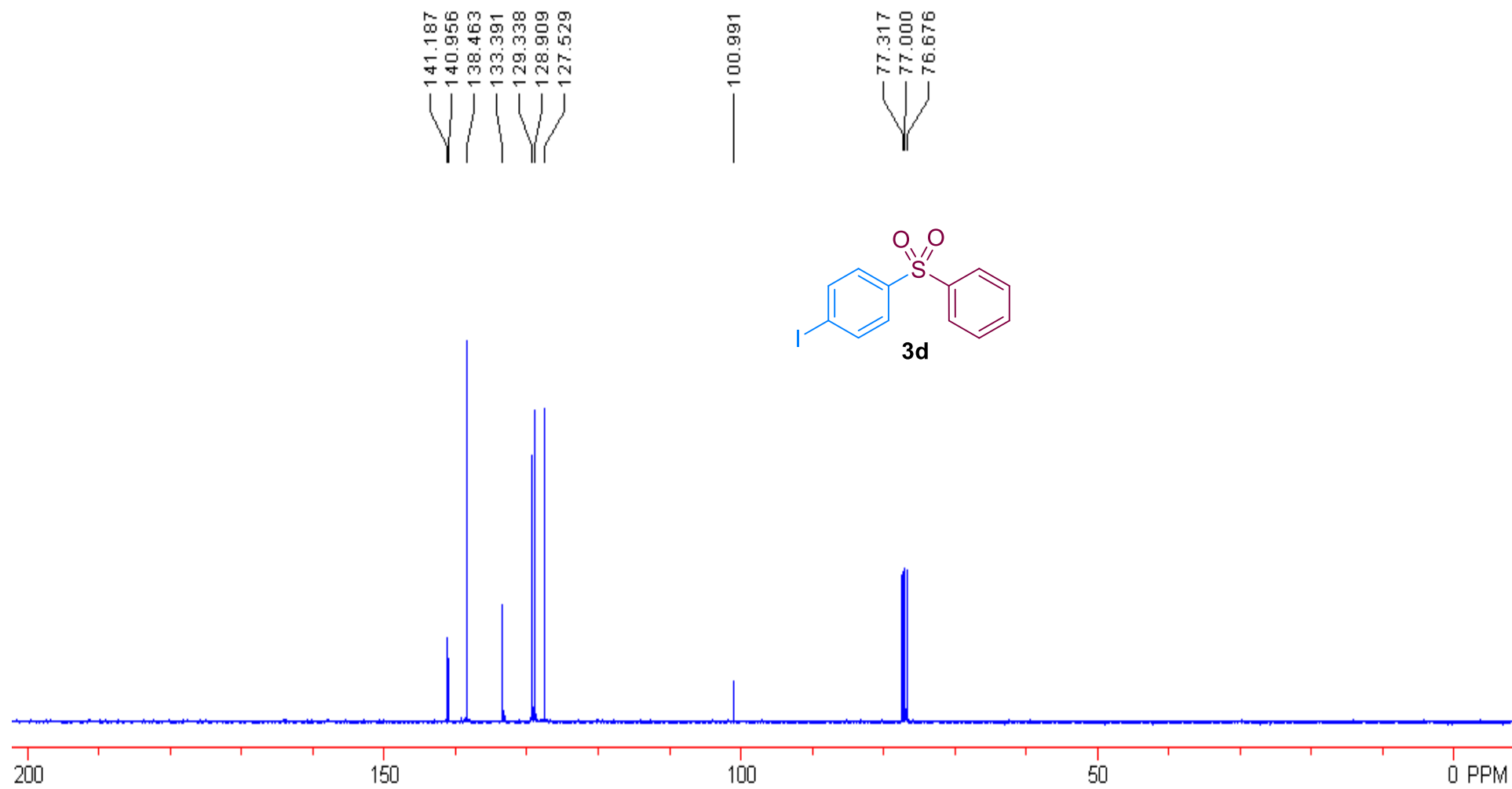


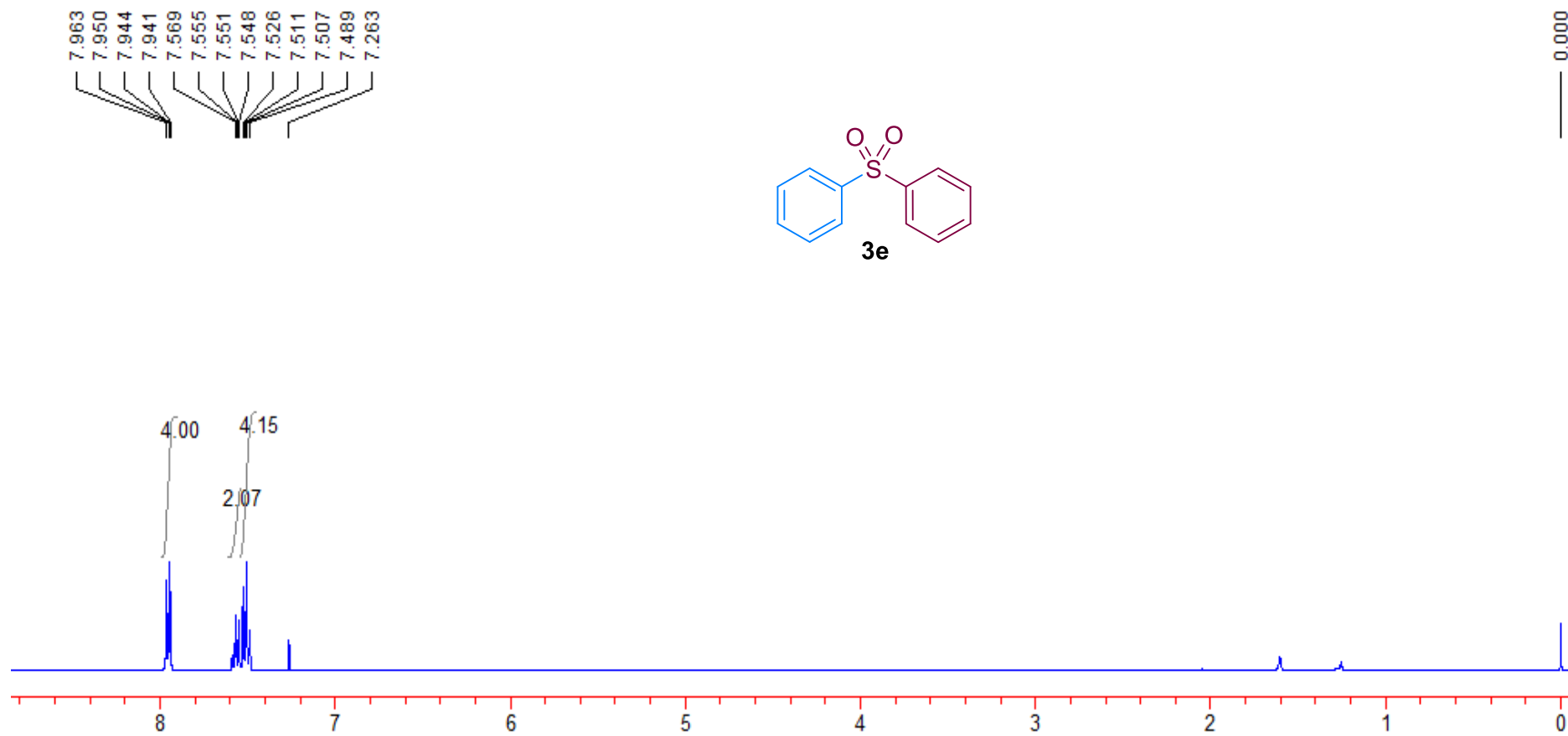
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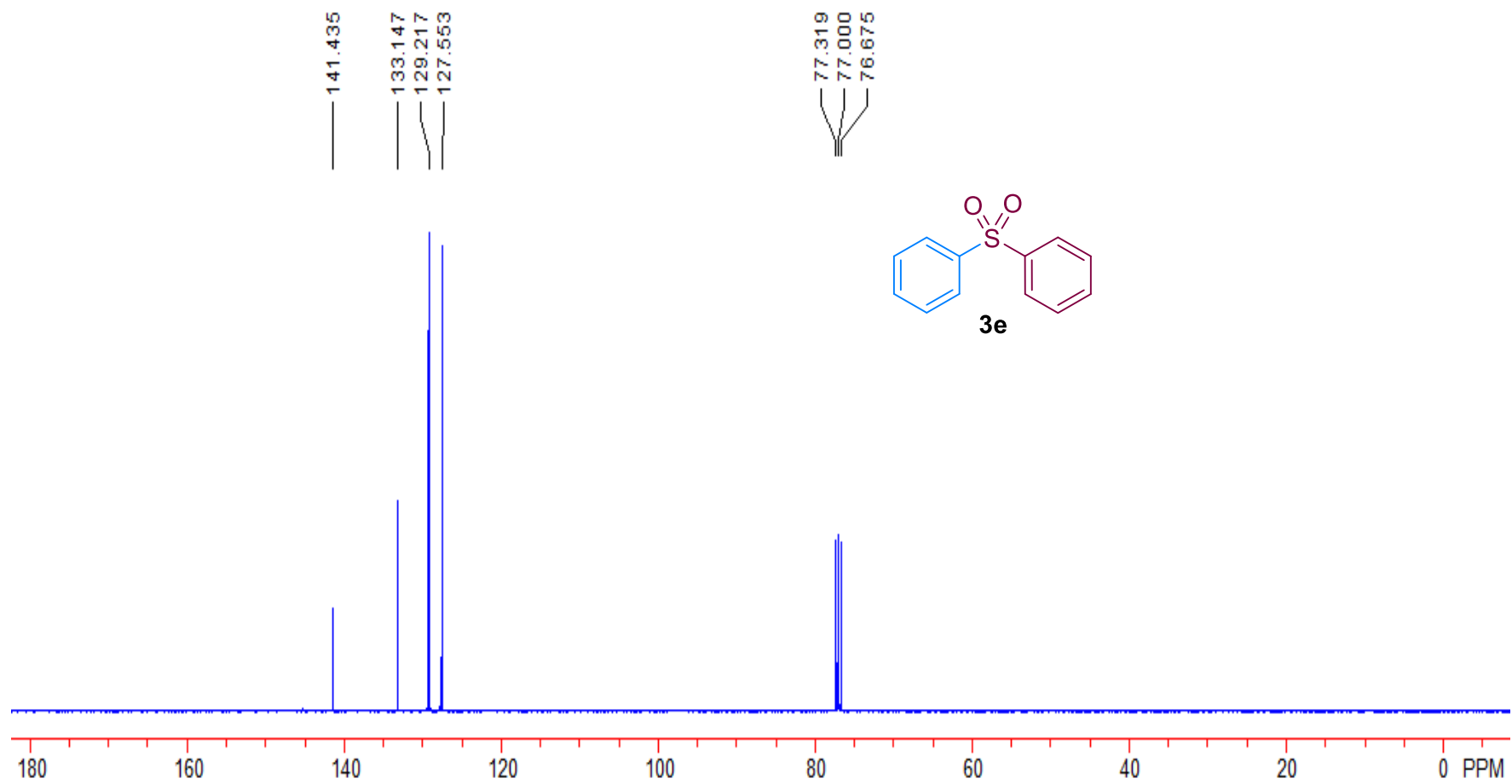


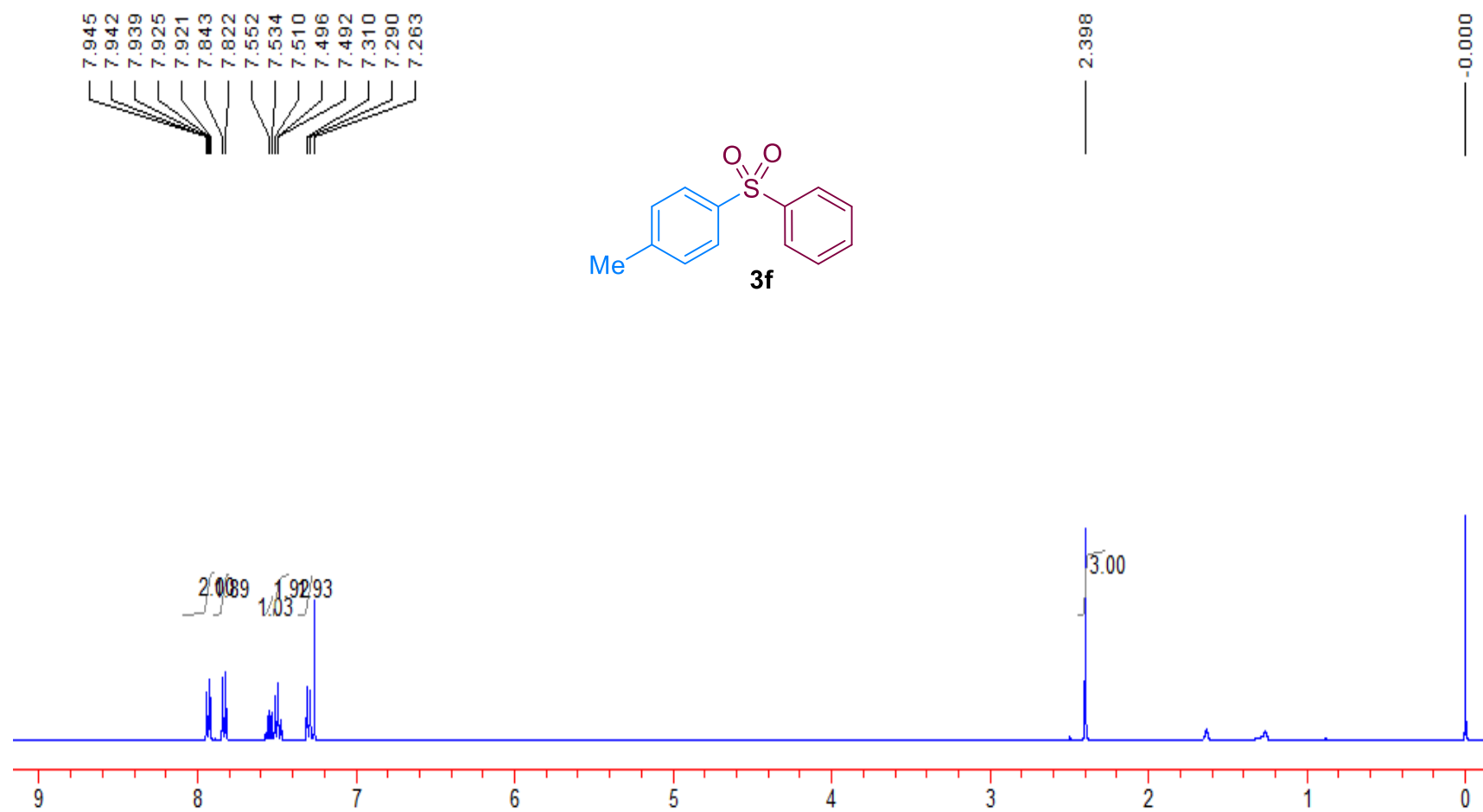
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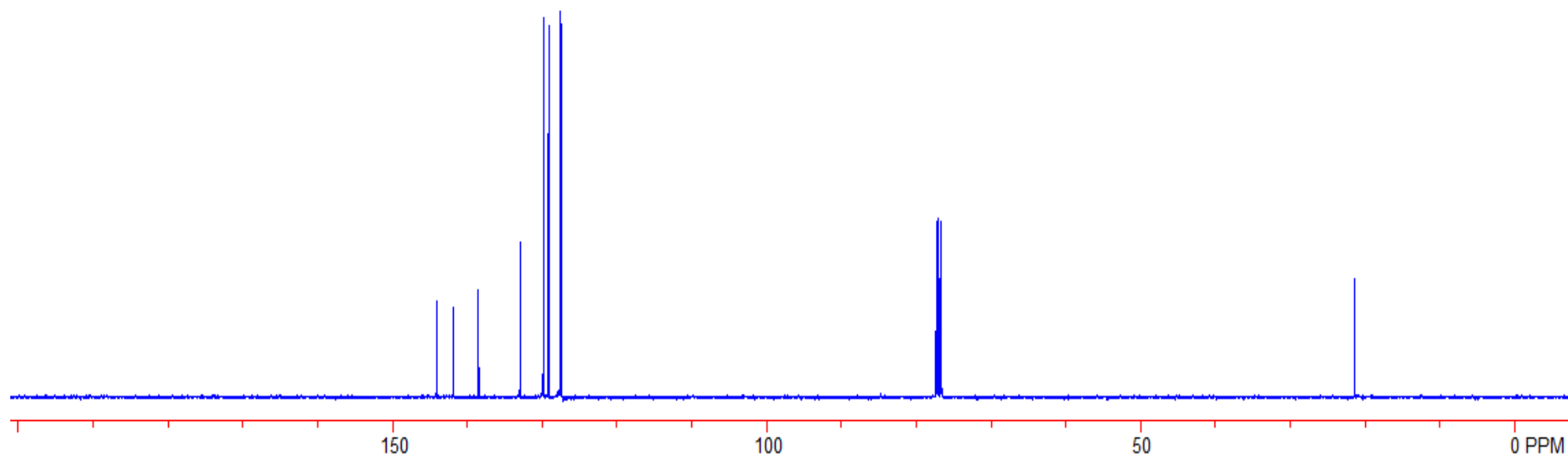
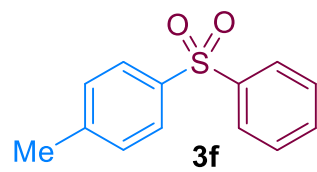


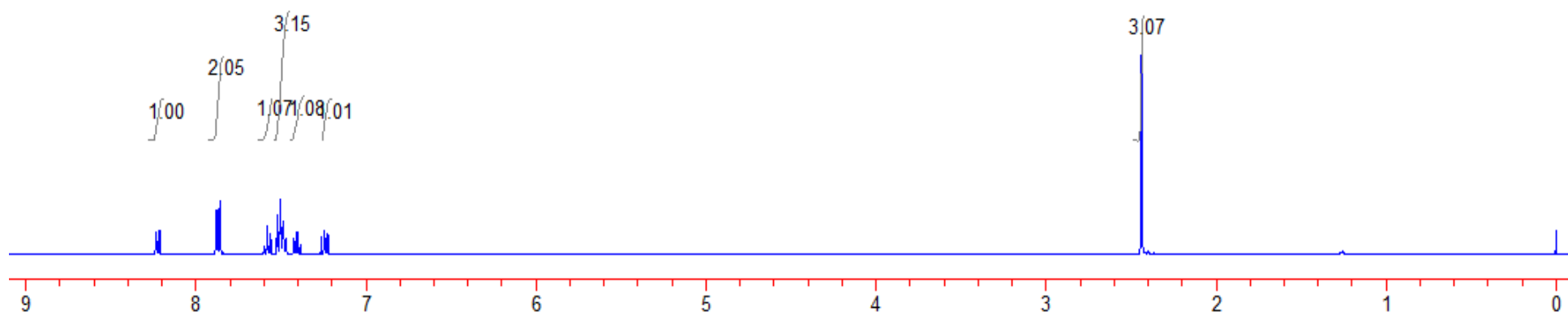
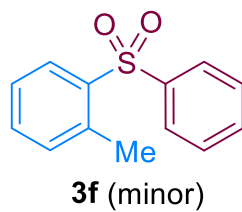
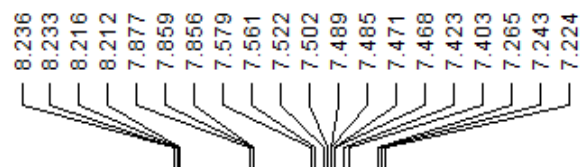


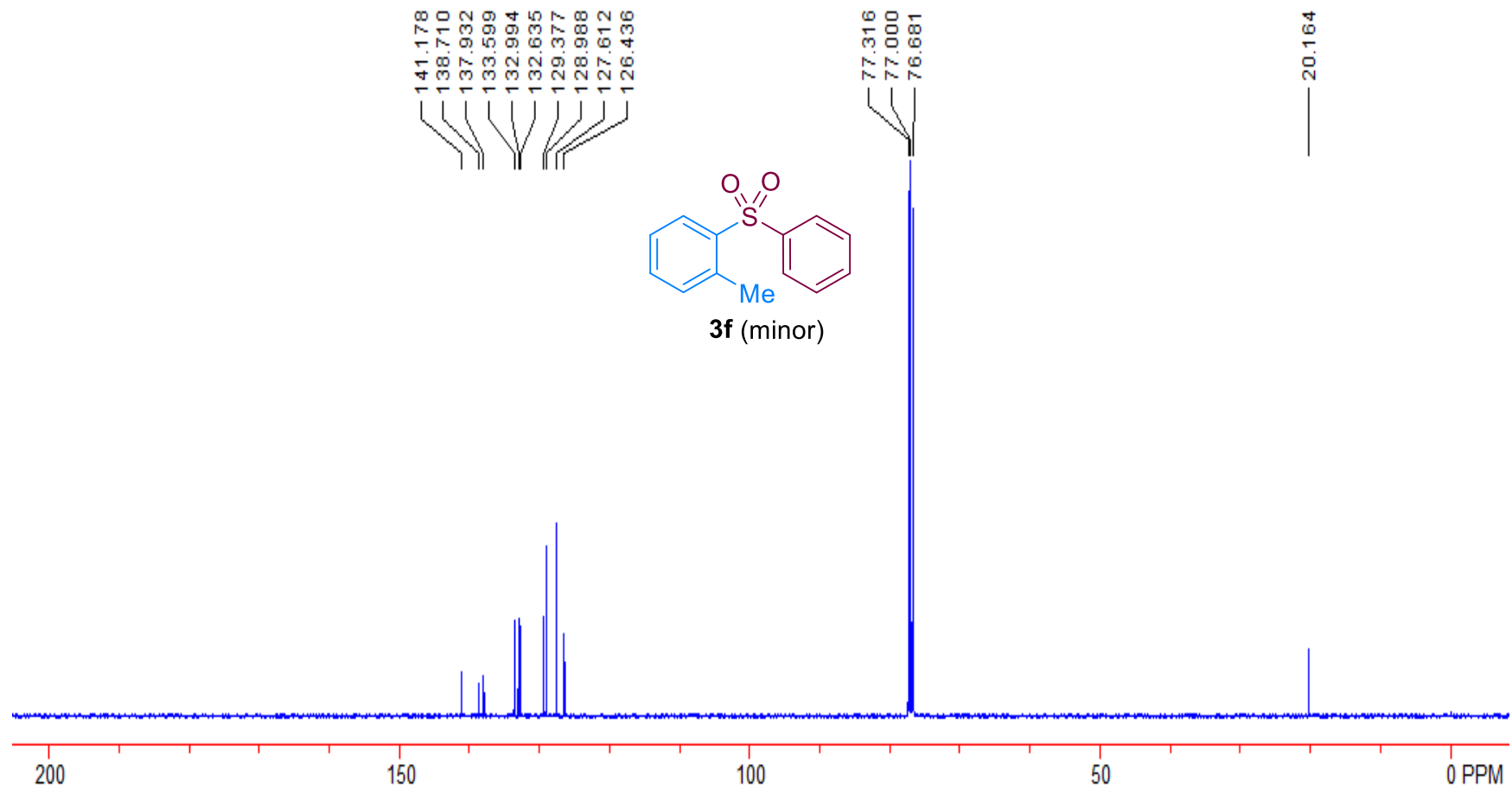
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127.572
127.347

77.314
77.000
76.684

21.451





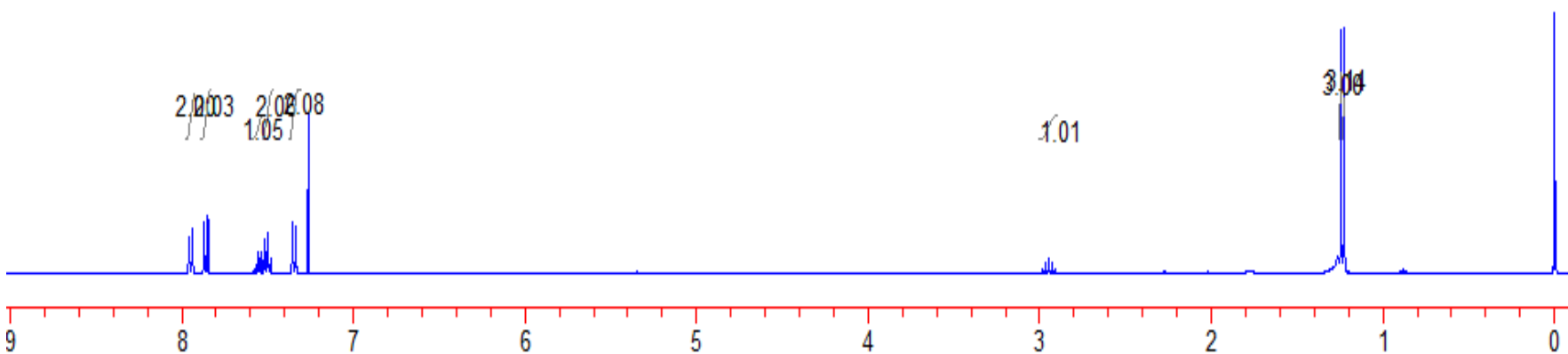
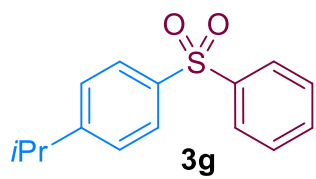


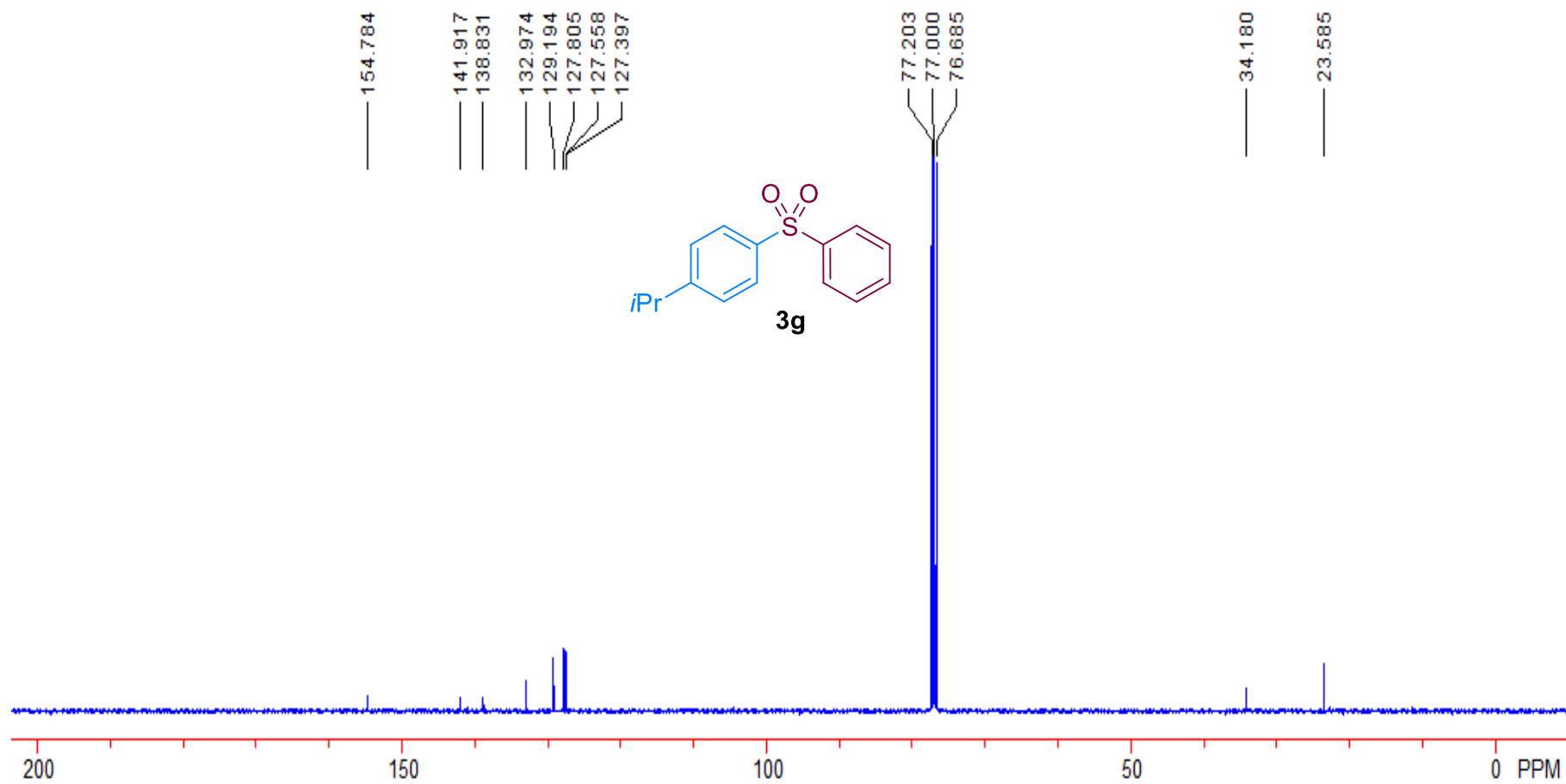
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7.954
7.940
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7.852
7.848
7.558
7.543
7.541
7.519
7.515
7.504
7.500
7.483
7.356
7.335
7.263

2.947
2.930

1.264
1.245
1.228

-0.000





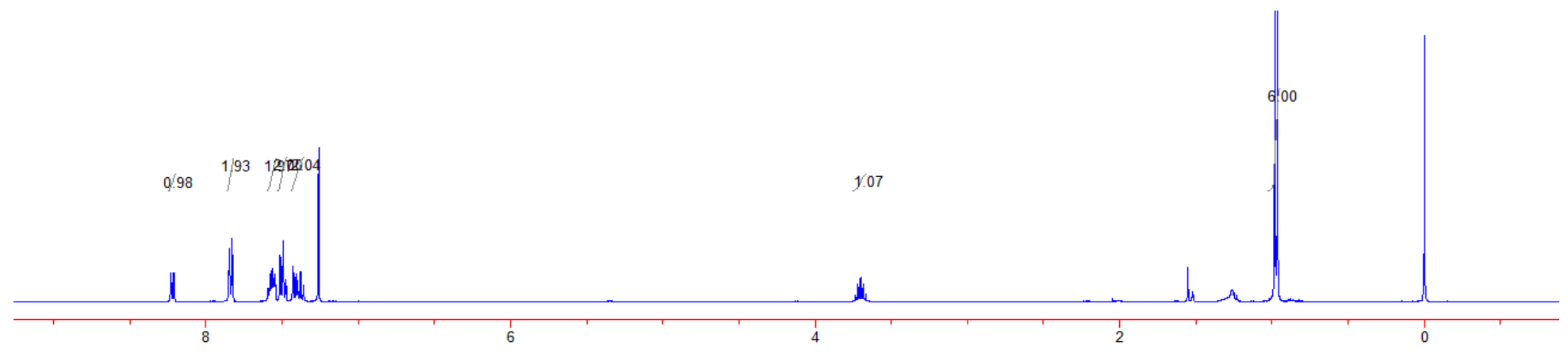
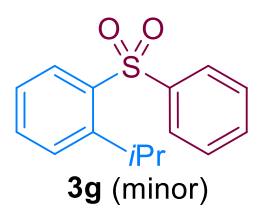
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8.228
8.211
8.208
7.846
7.827
7.824
7.571
7.563
7.556
7.553
7.548
7.545
7.511
7.495
7.492
7.474
7.426
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7.398
7.378
7.260

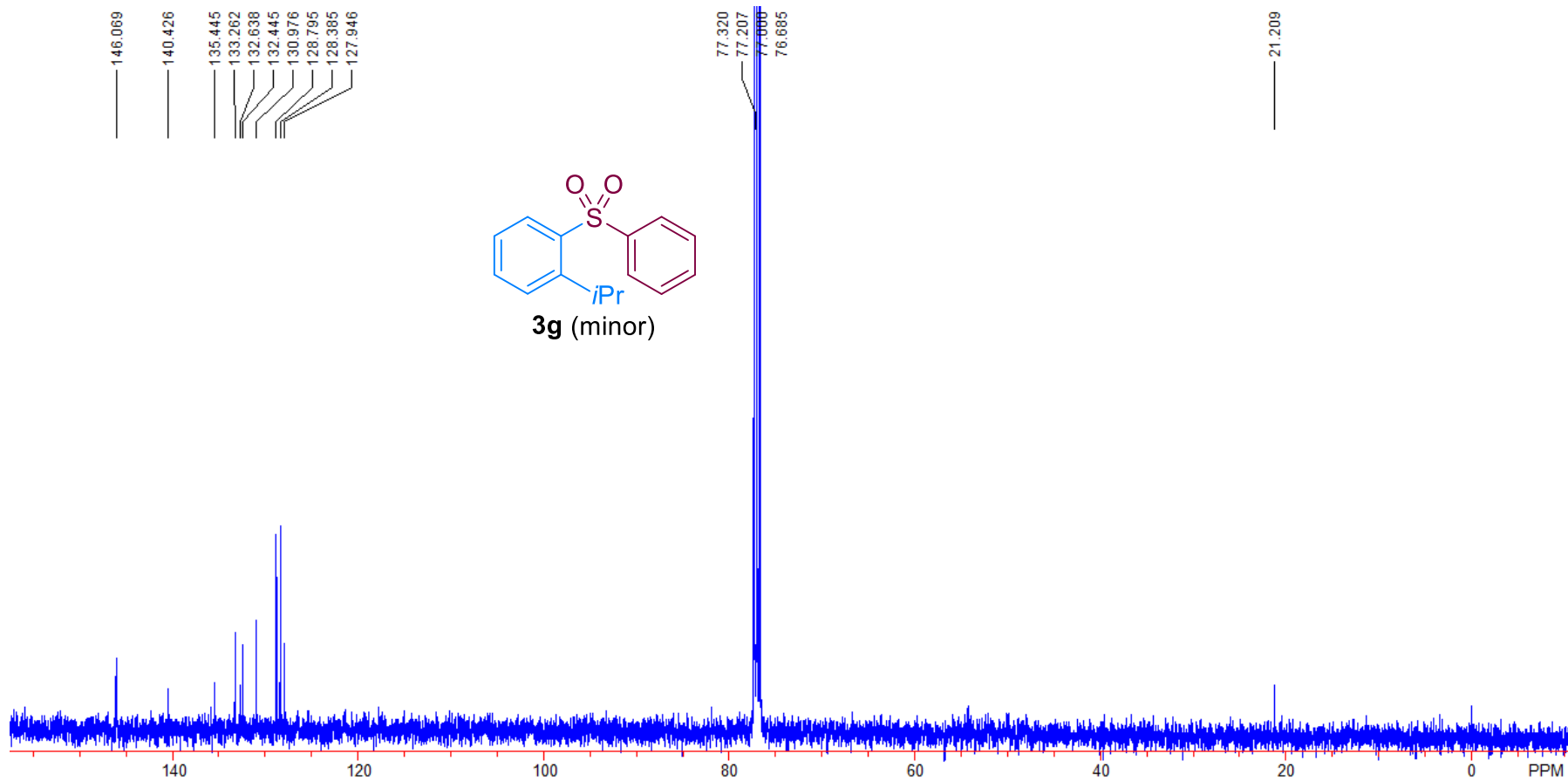
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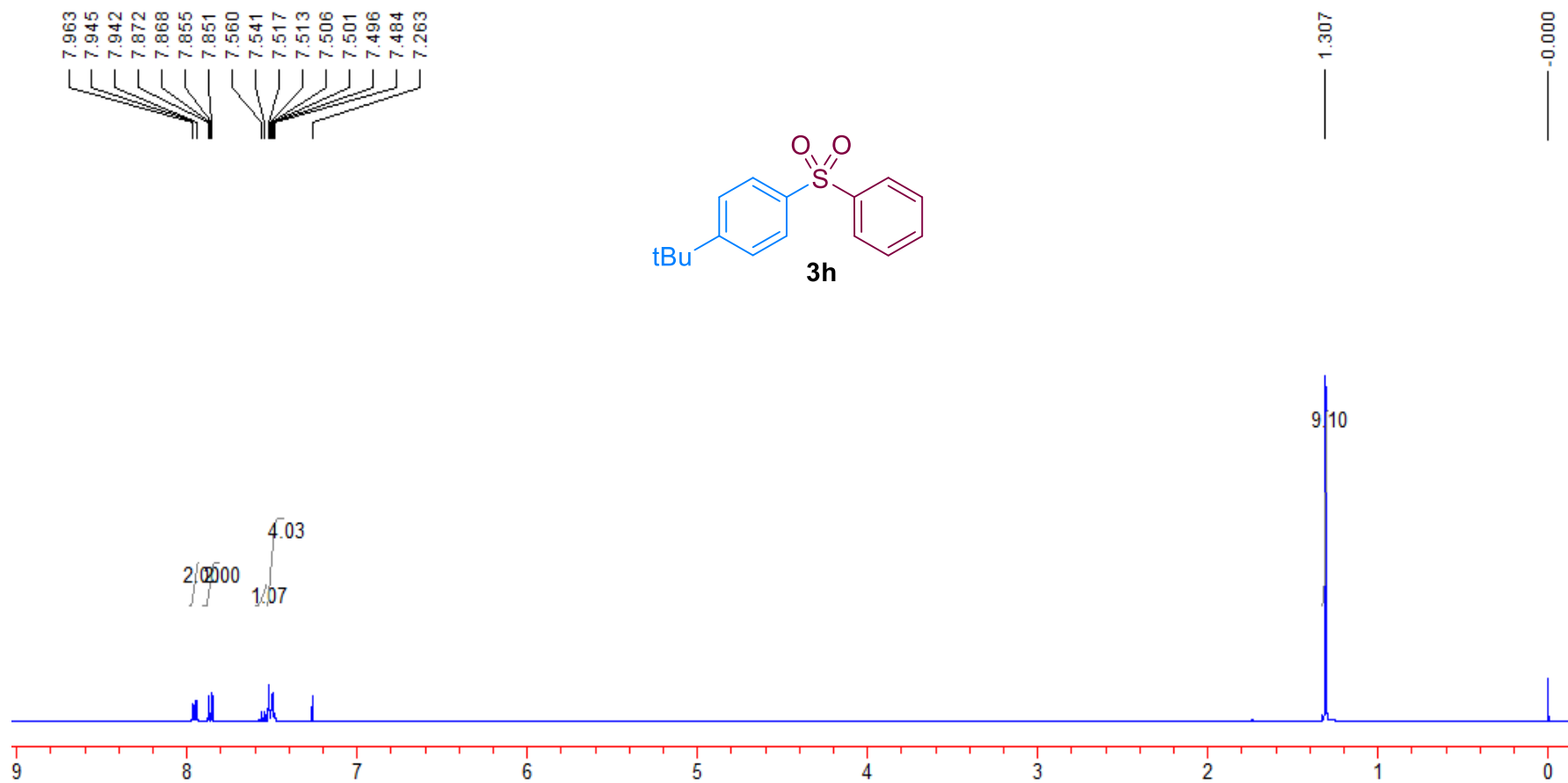
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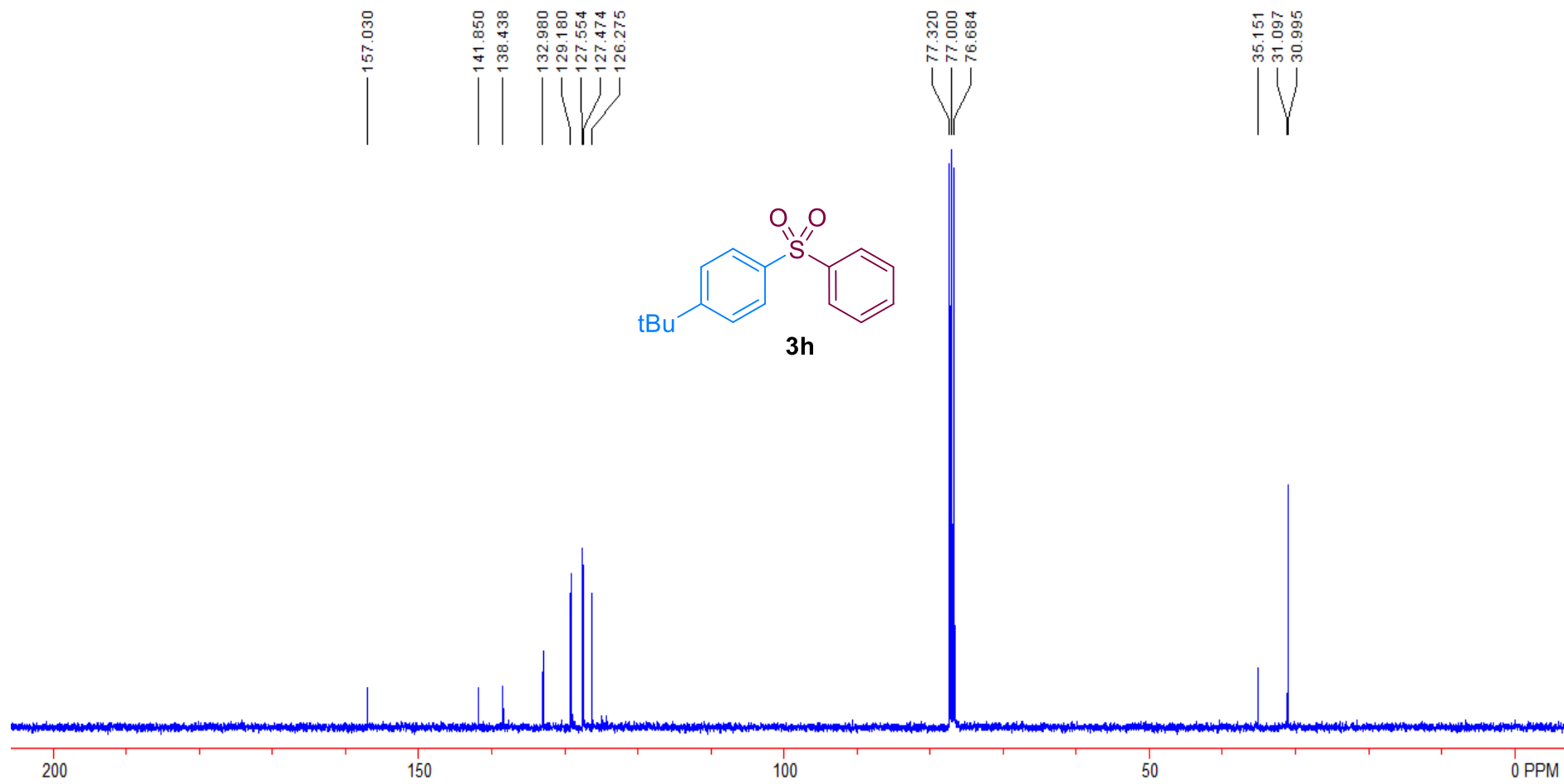
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0.963

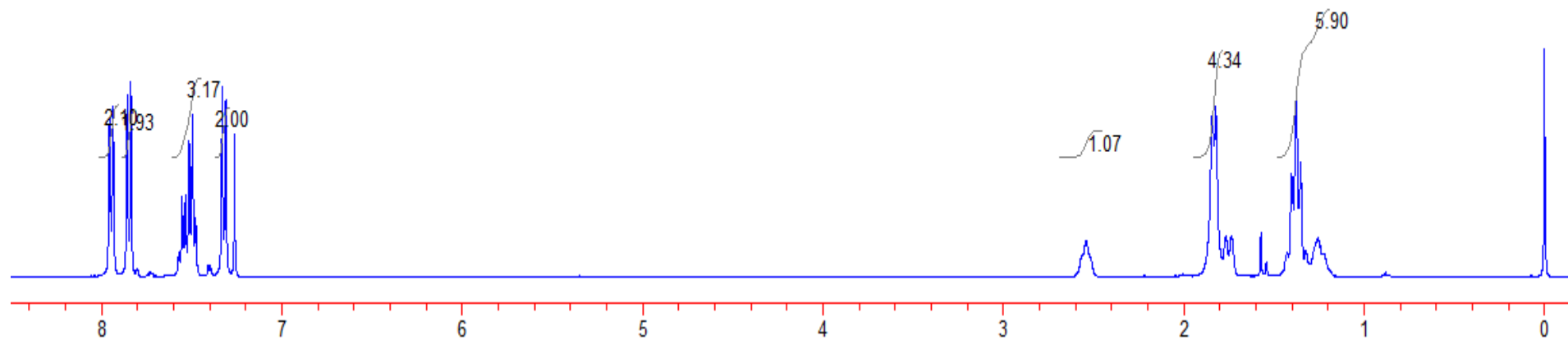
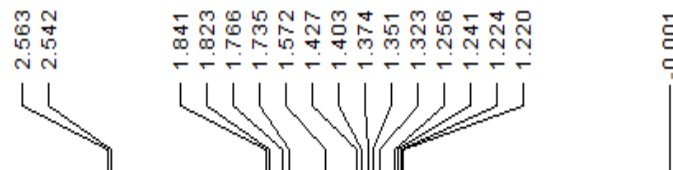
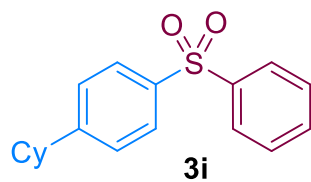
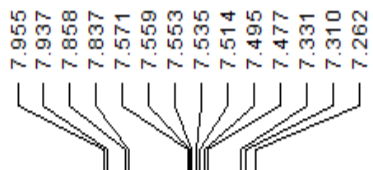
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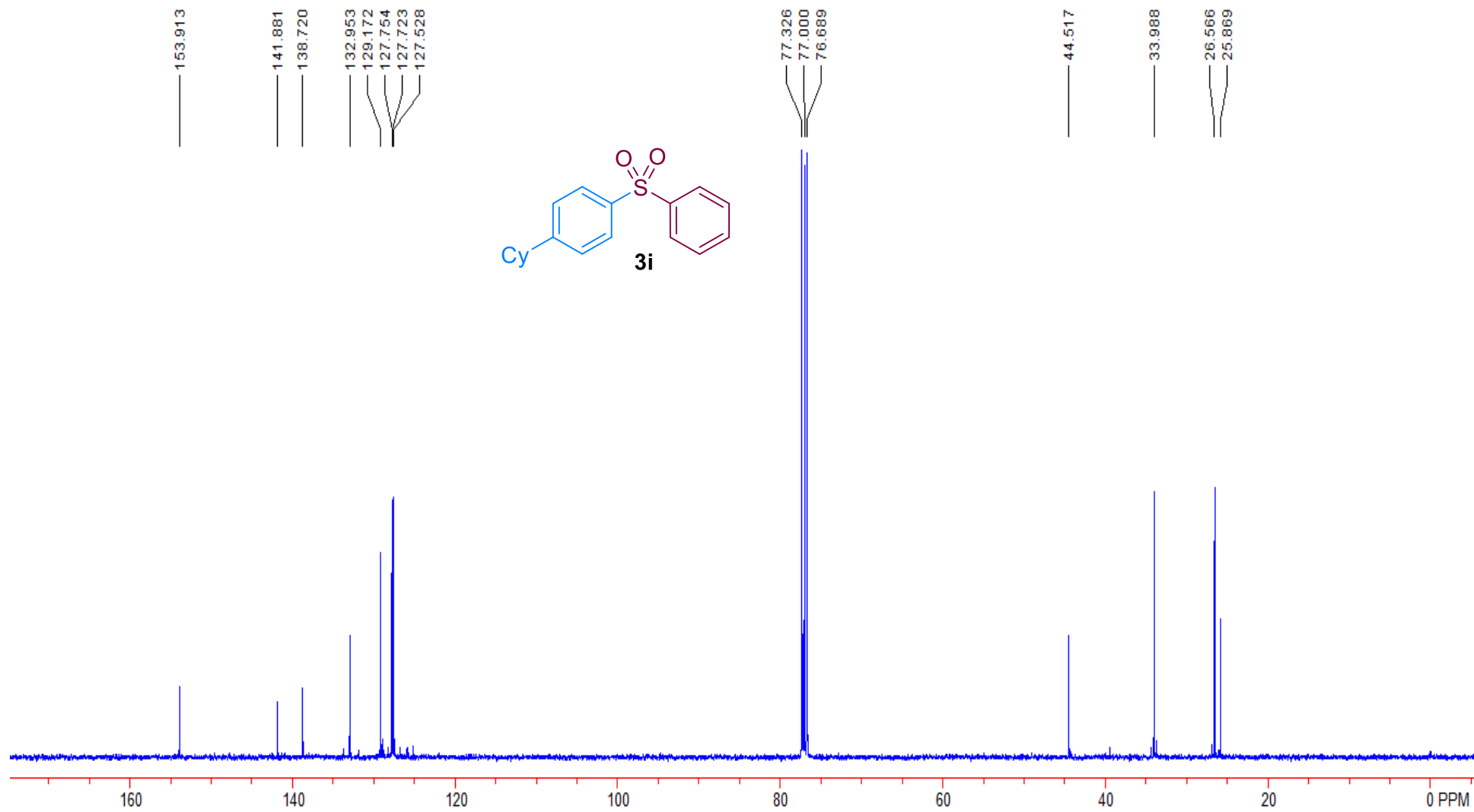






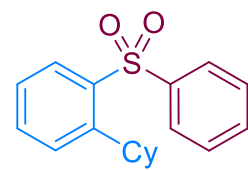




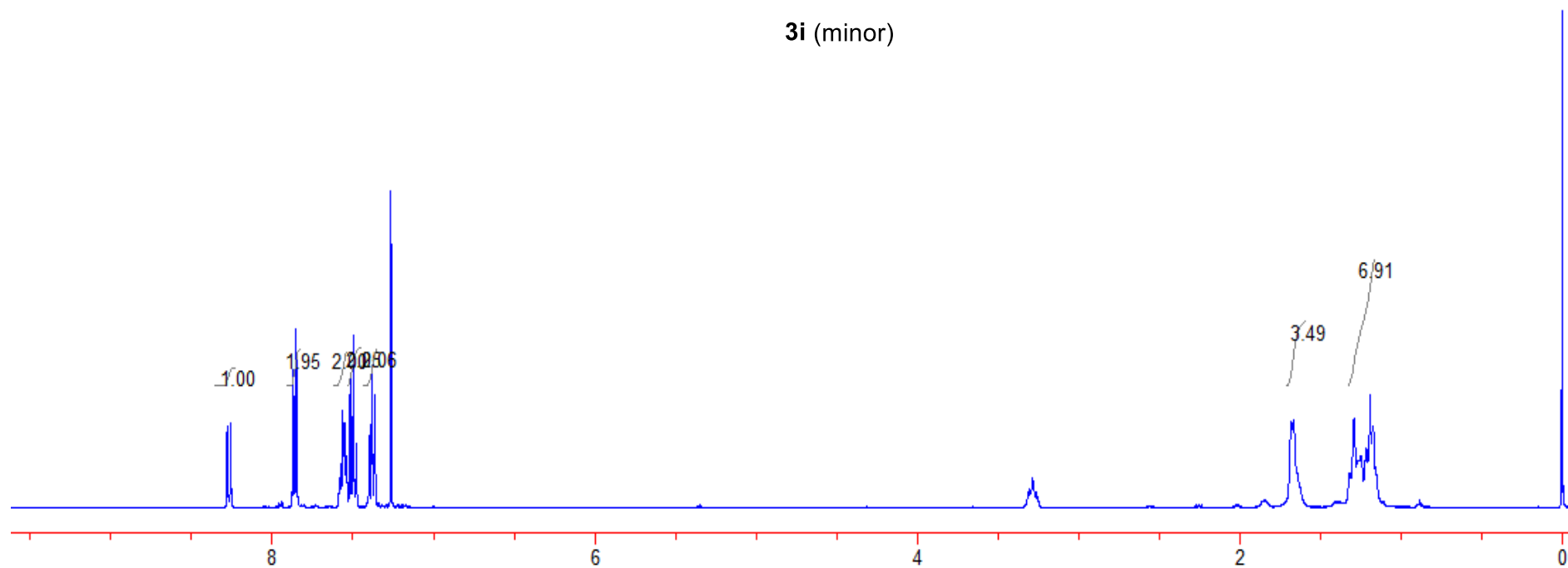


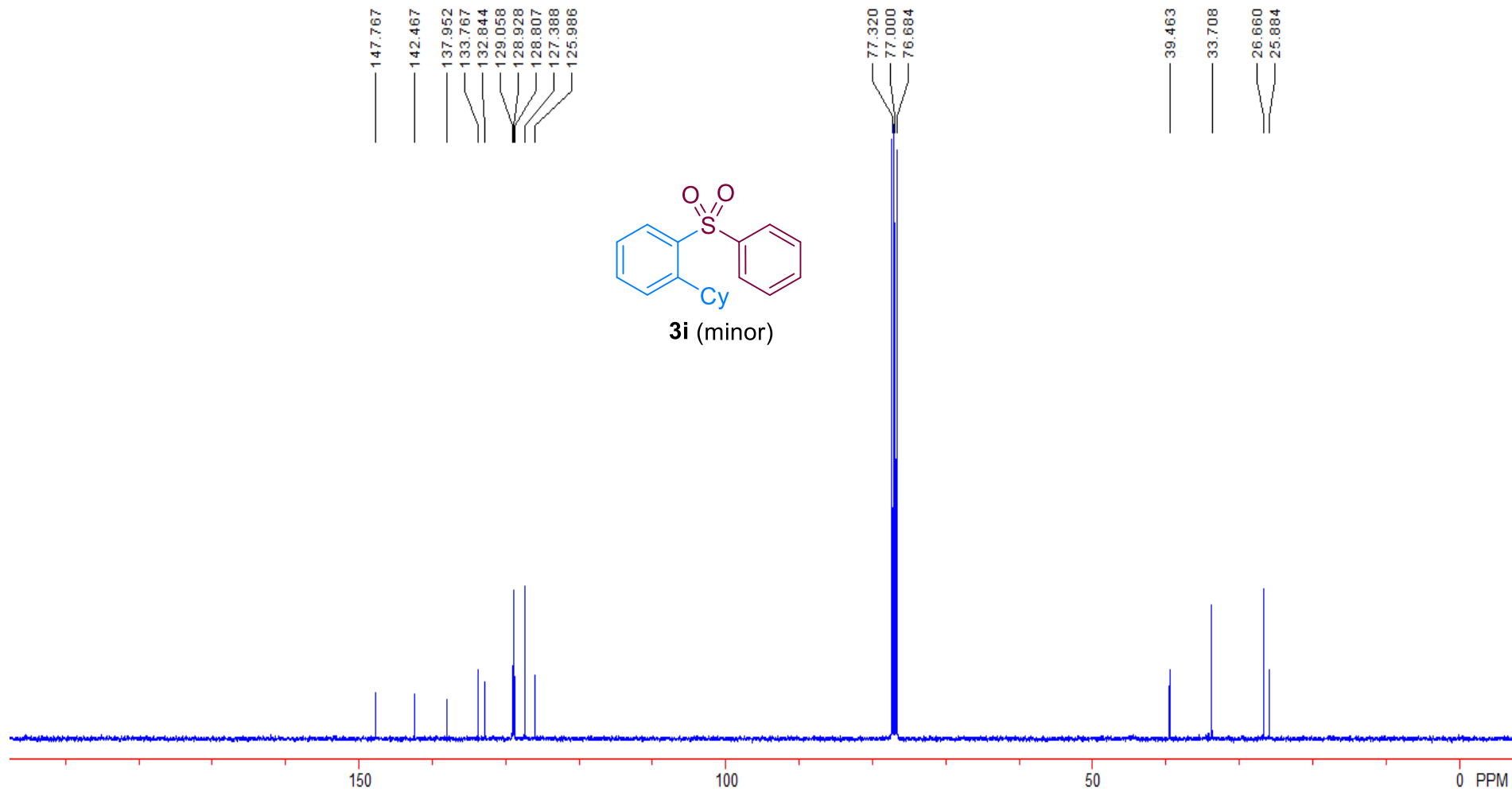
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8.277
8.260
8.256
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7.865
7.851
7.847
7.567
7.559
7.548
7.516
7.501
7.497
7.398
7.387
7.383
7.379
7.362
7.263

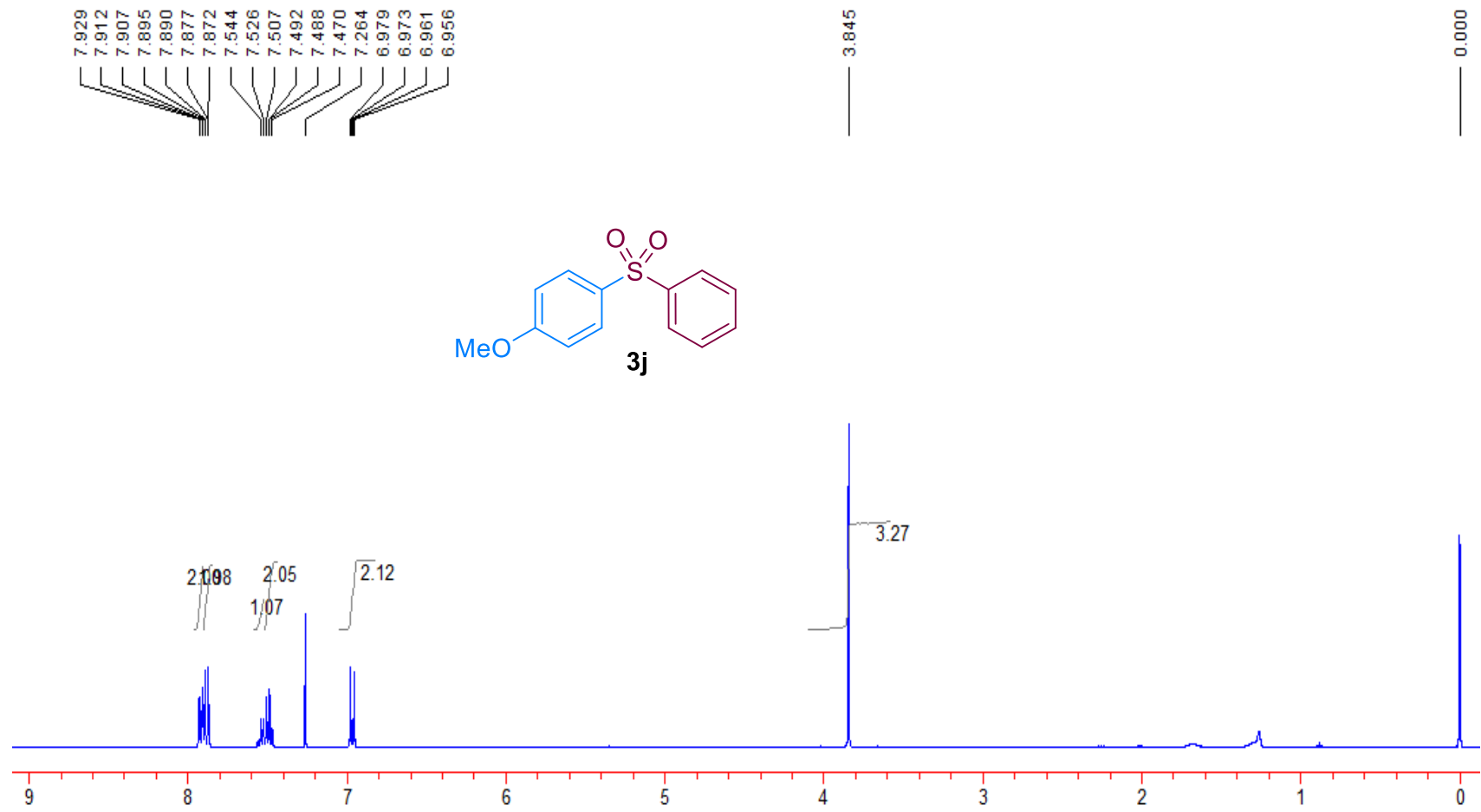
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1.664
1.289
1.190
1.174
0.000

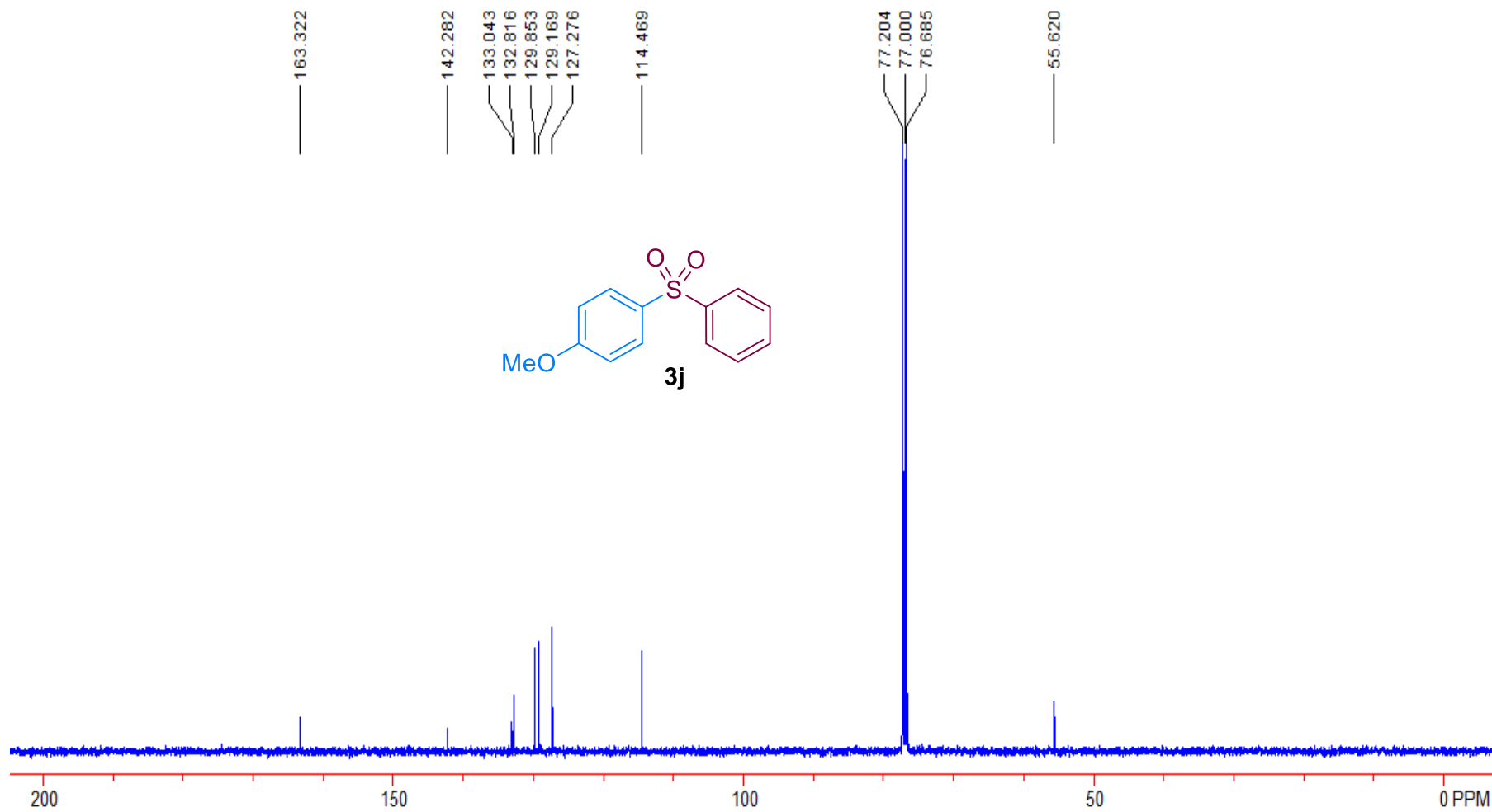


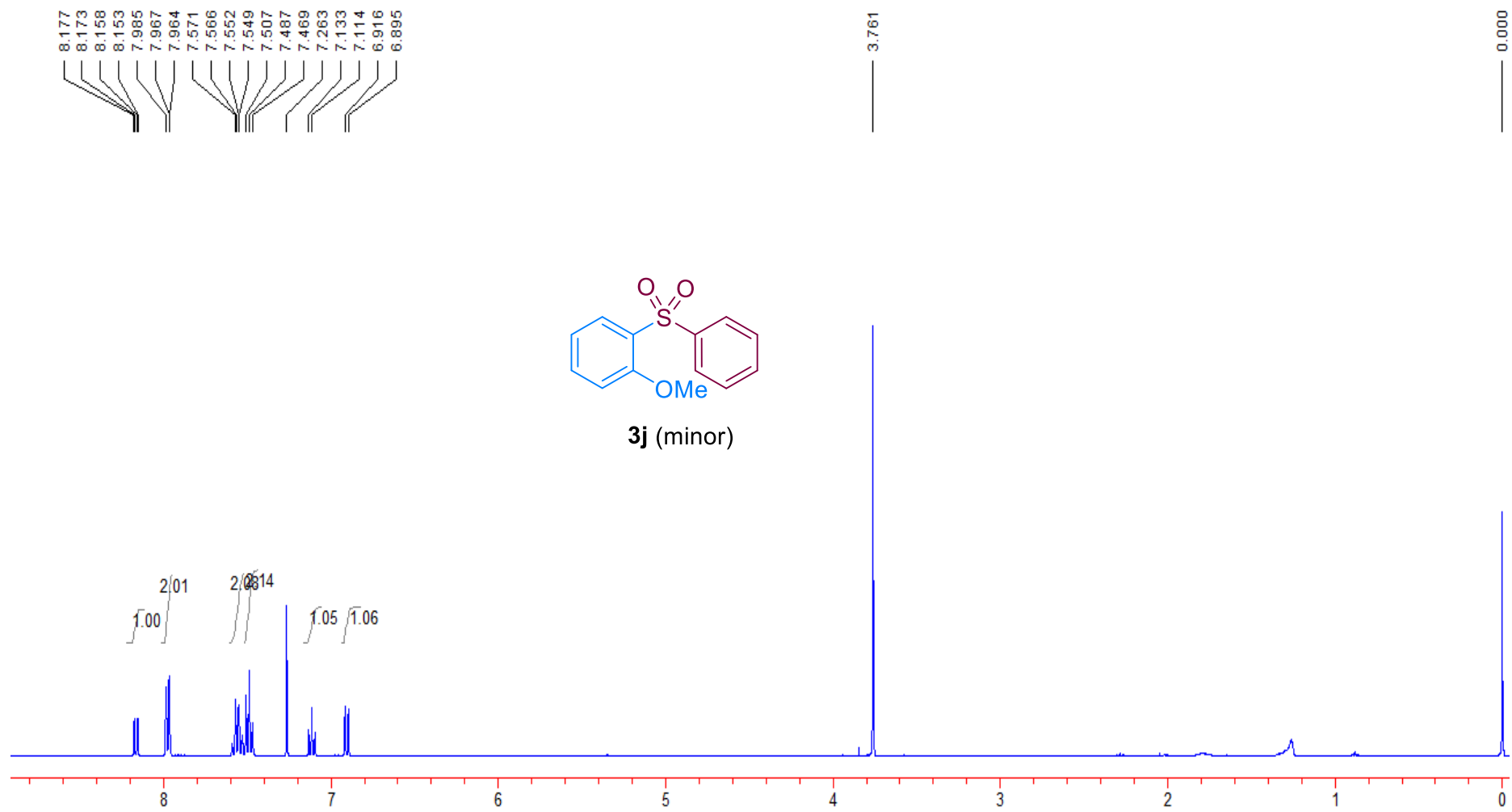
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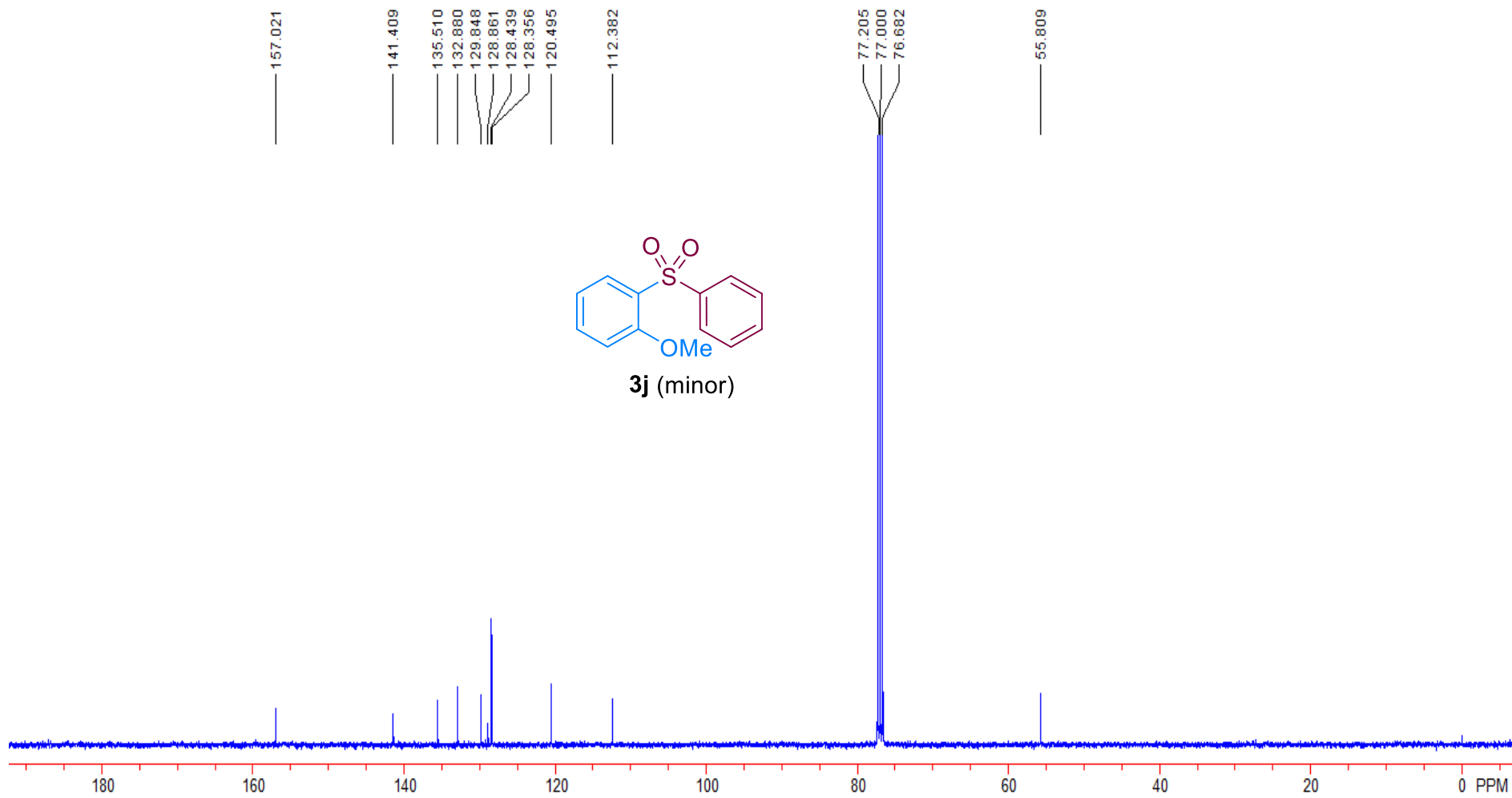


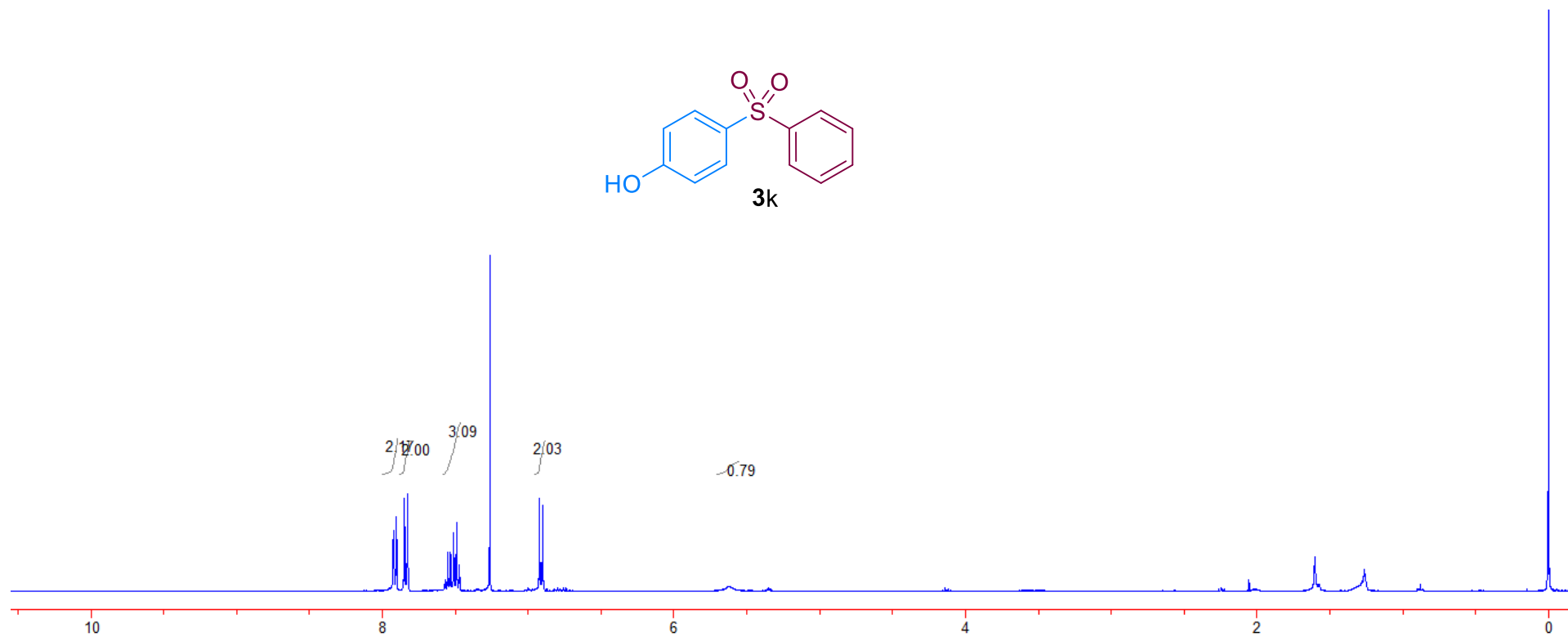
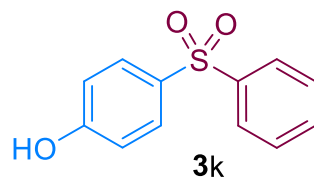
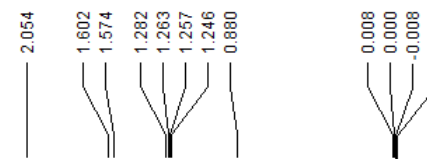
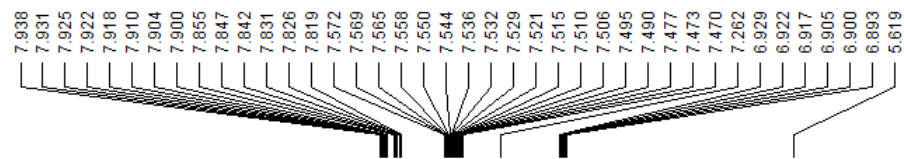


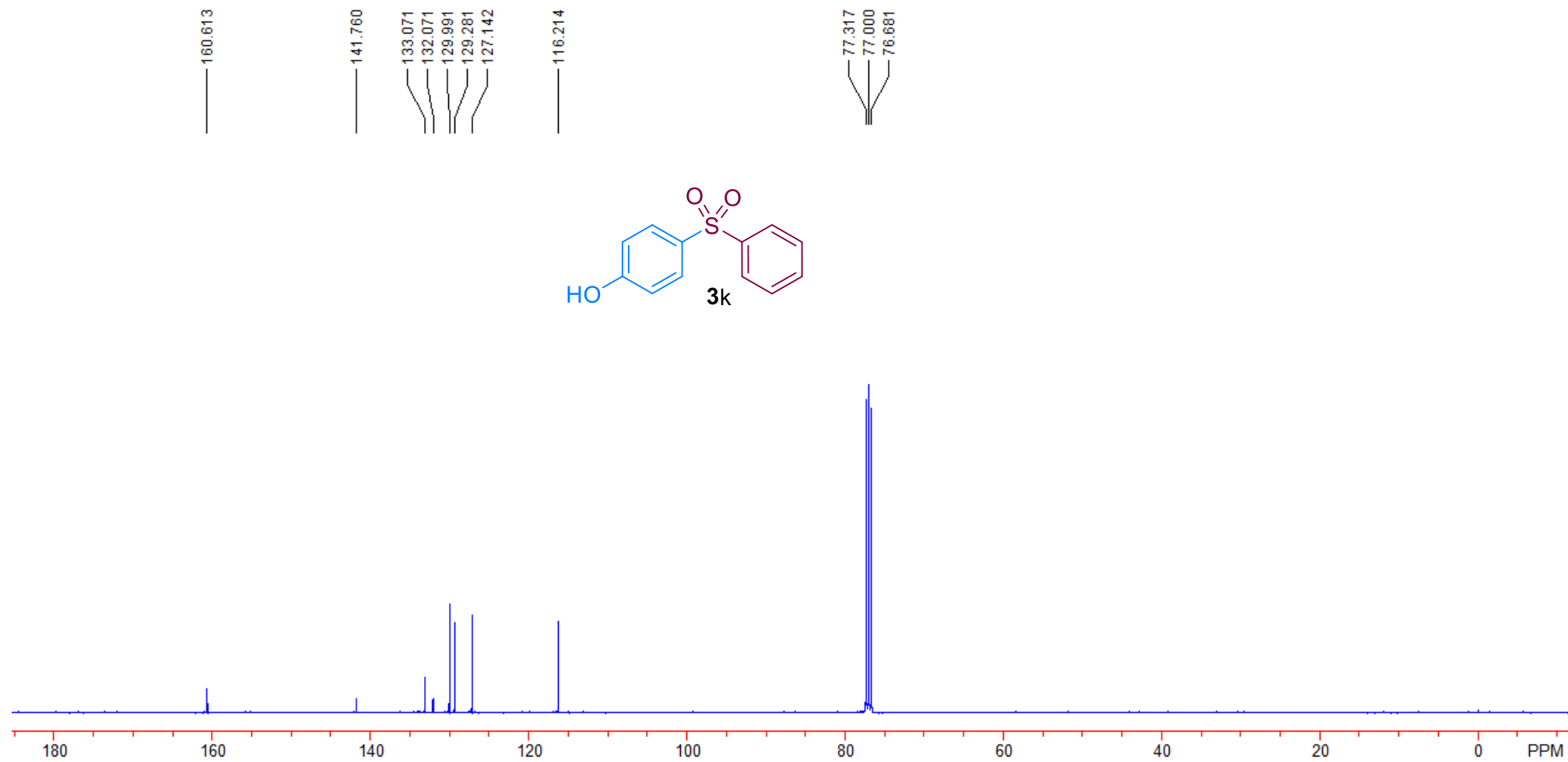


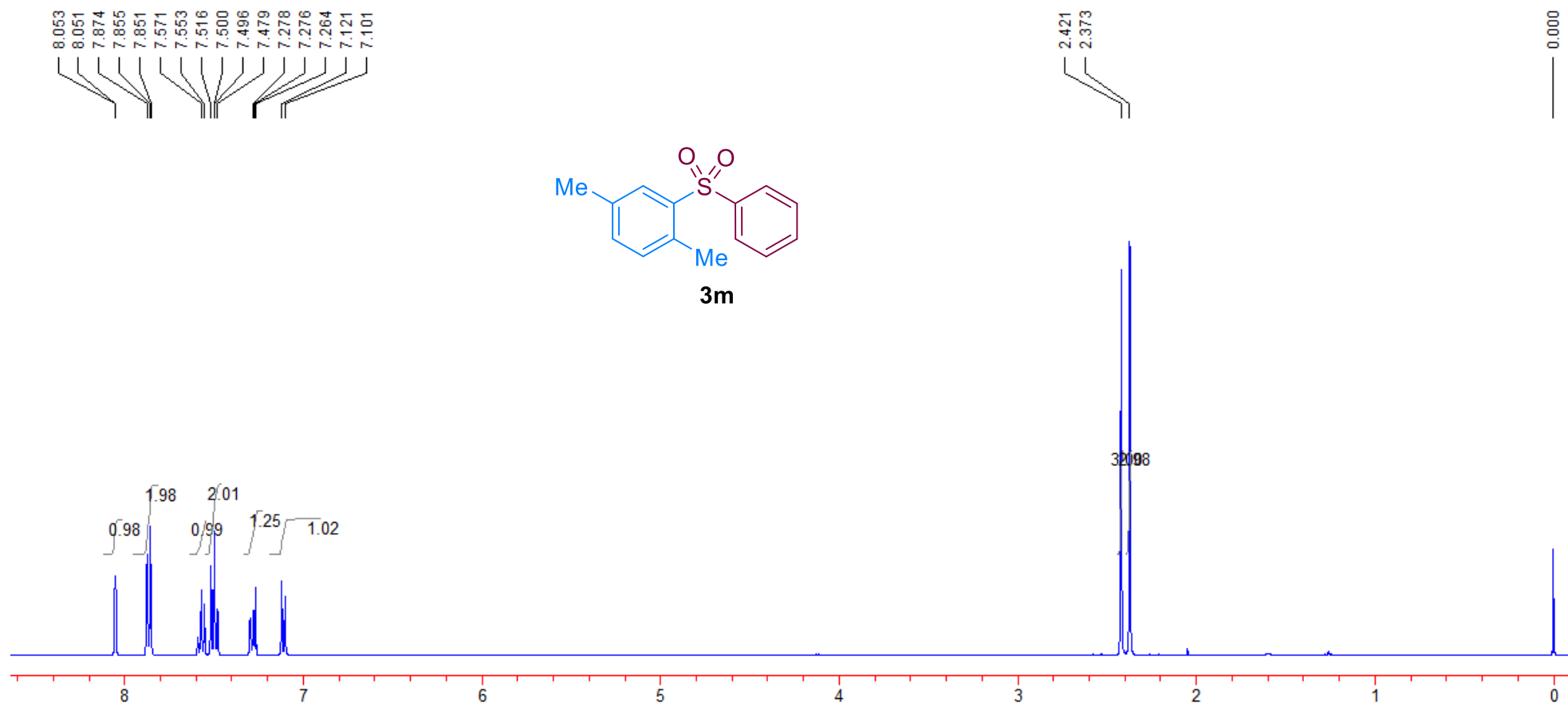


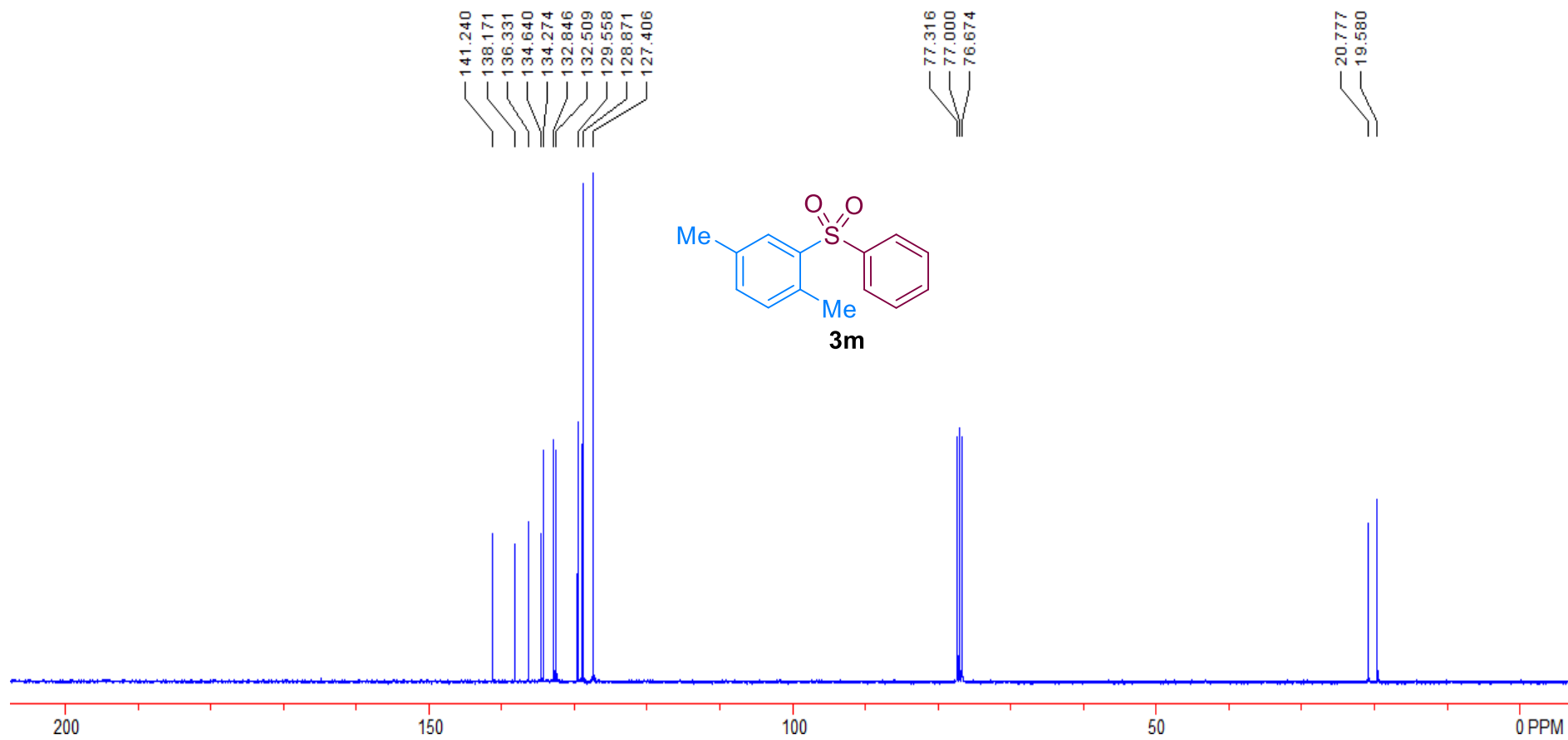








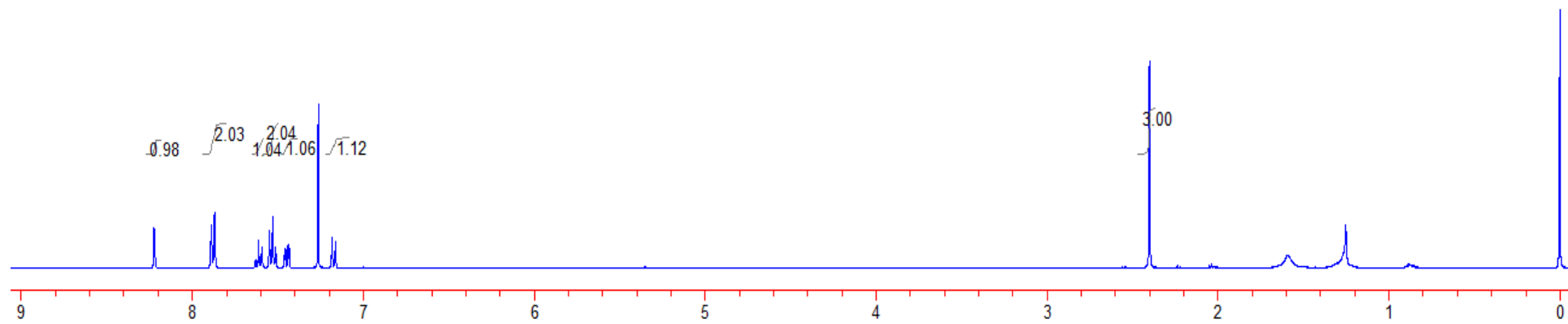
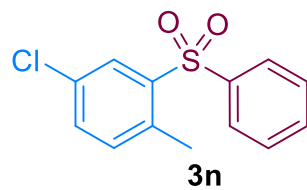


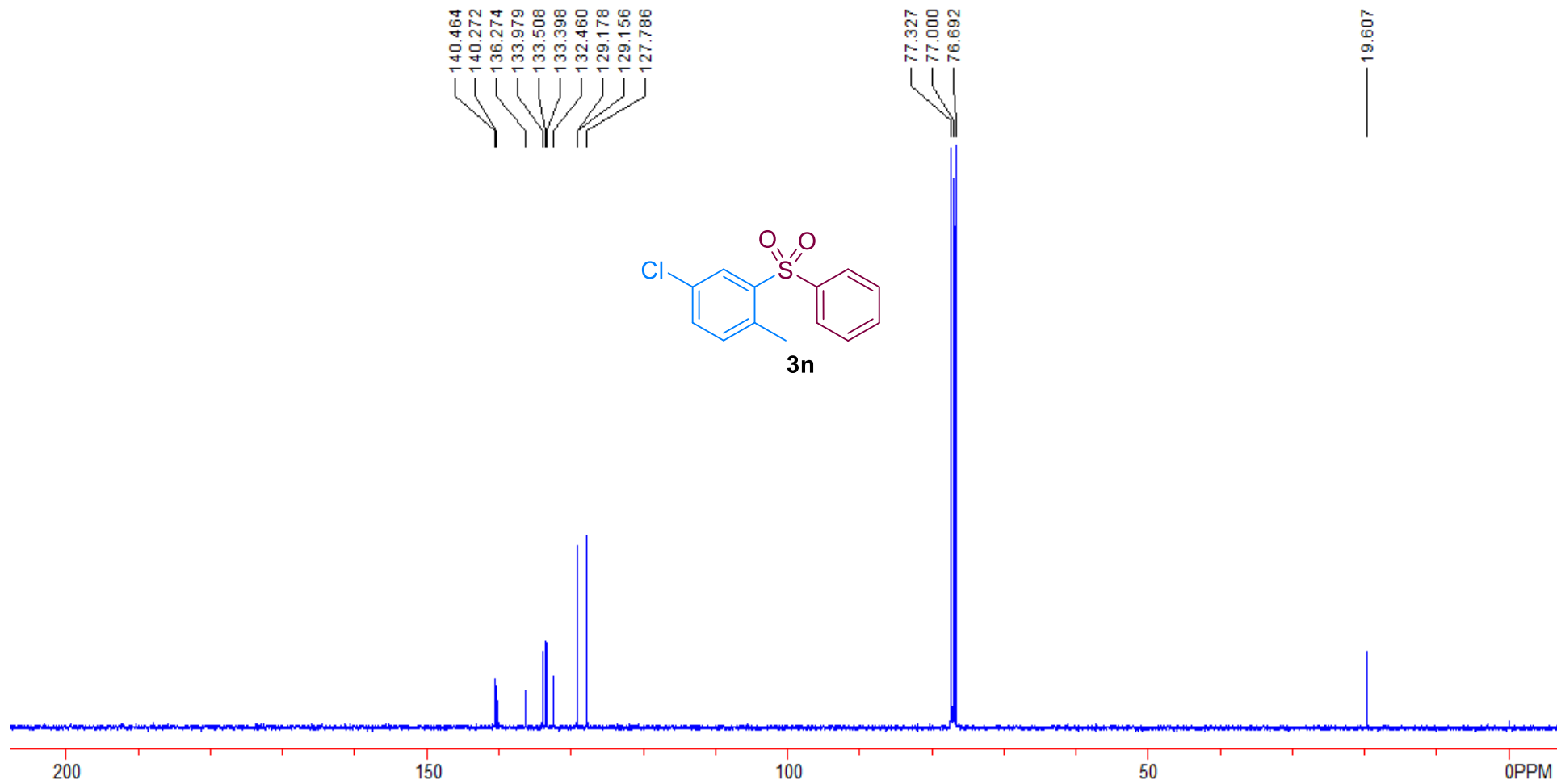


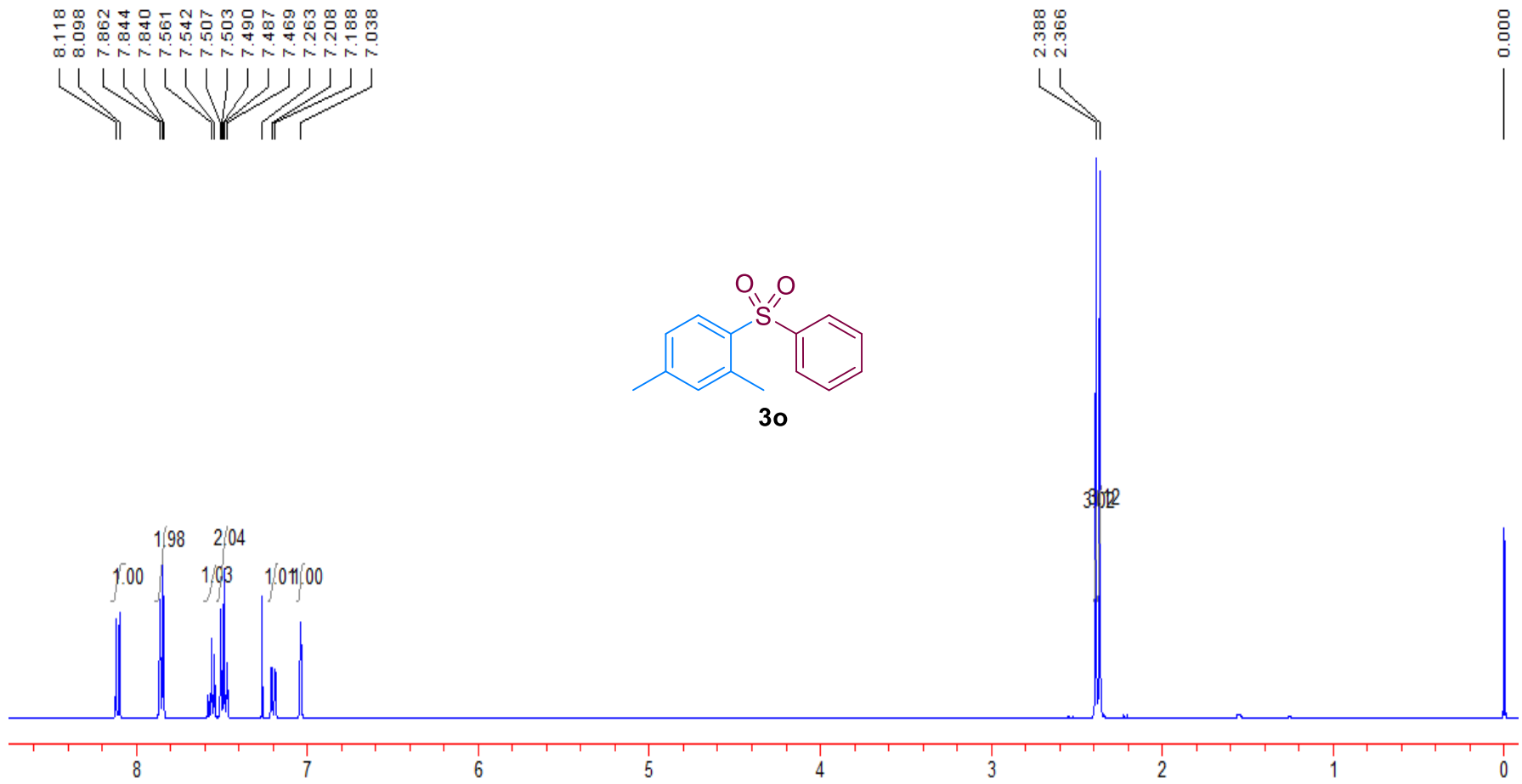
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8.219
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7.611
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7.550
7.533
7.530
7.512
7.460
7.454
7.440
7.434
7.263
7.183
7.163

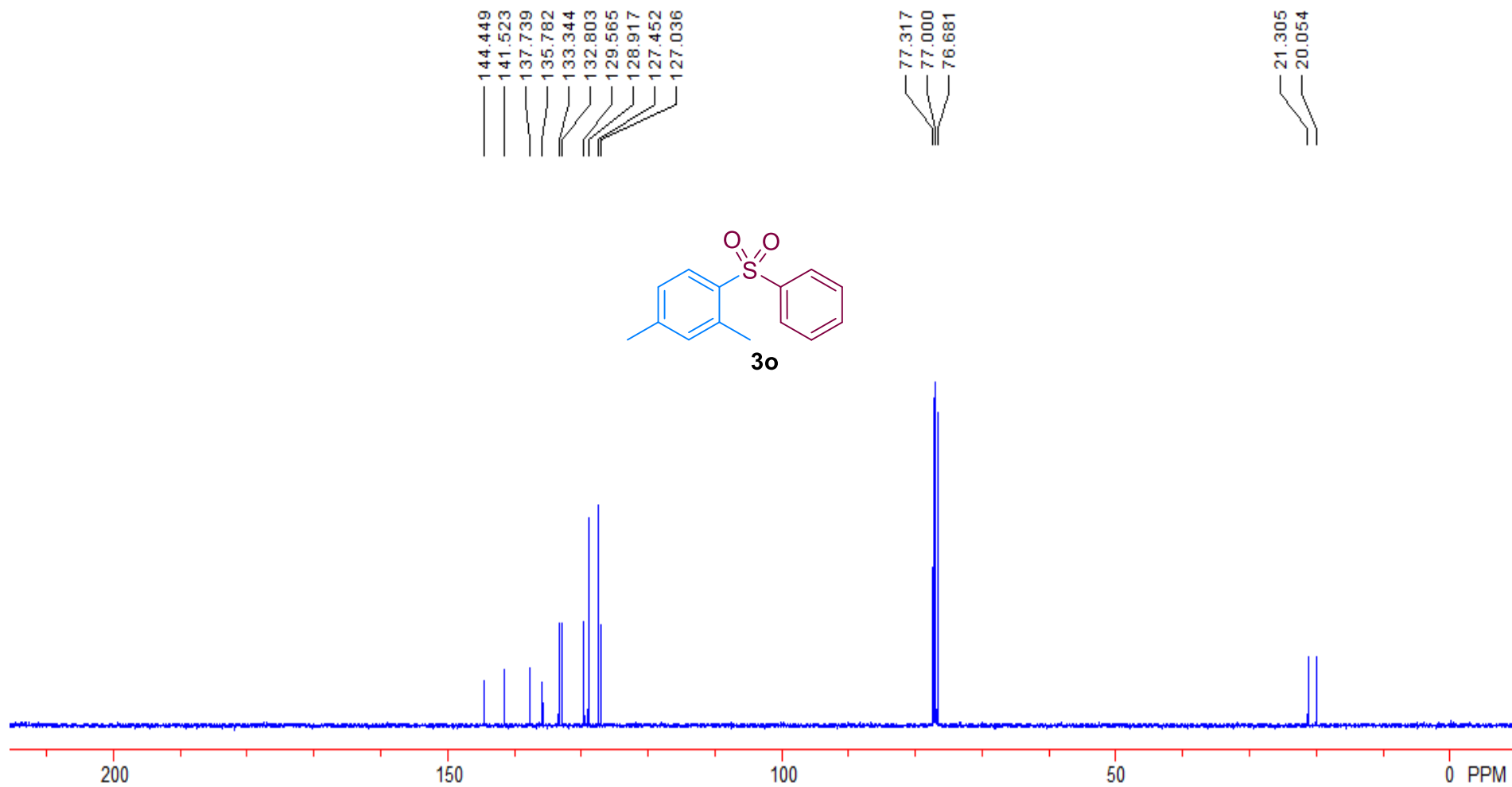
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0.000

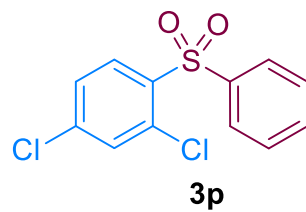




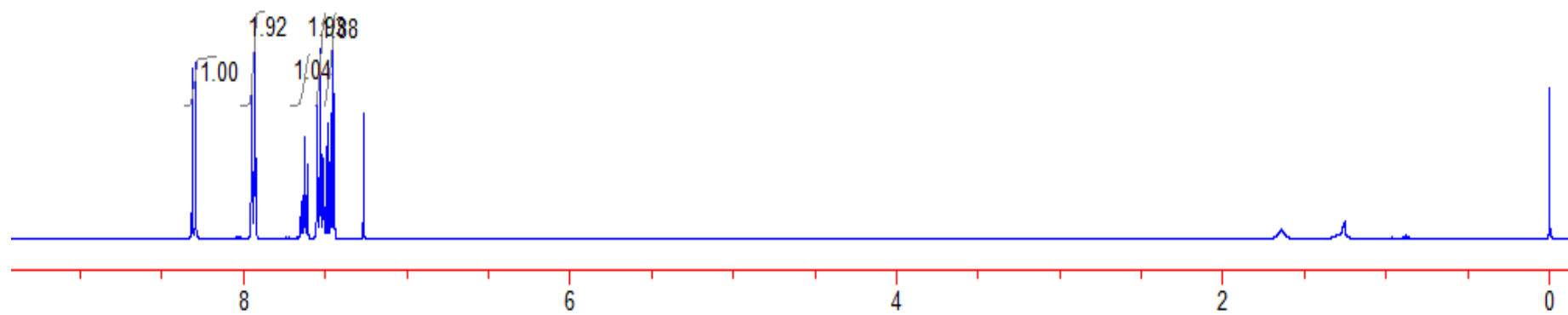


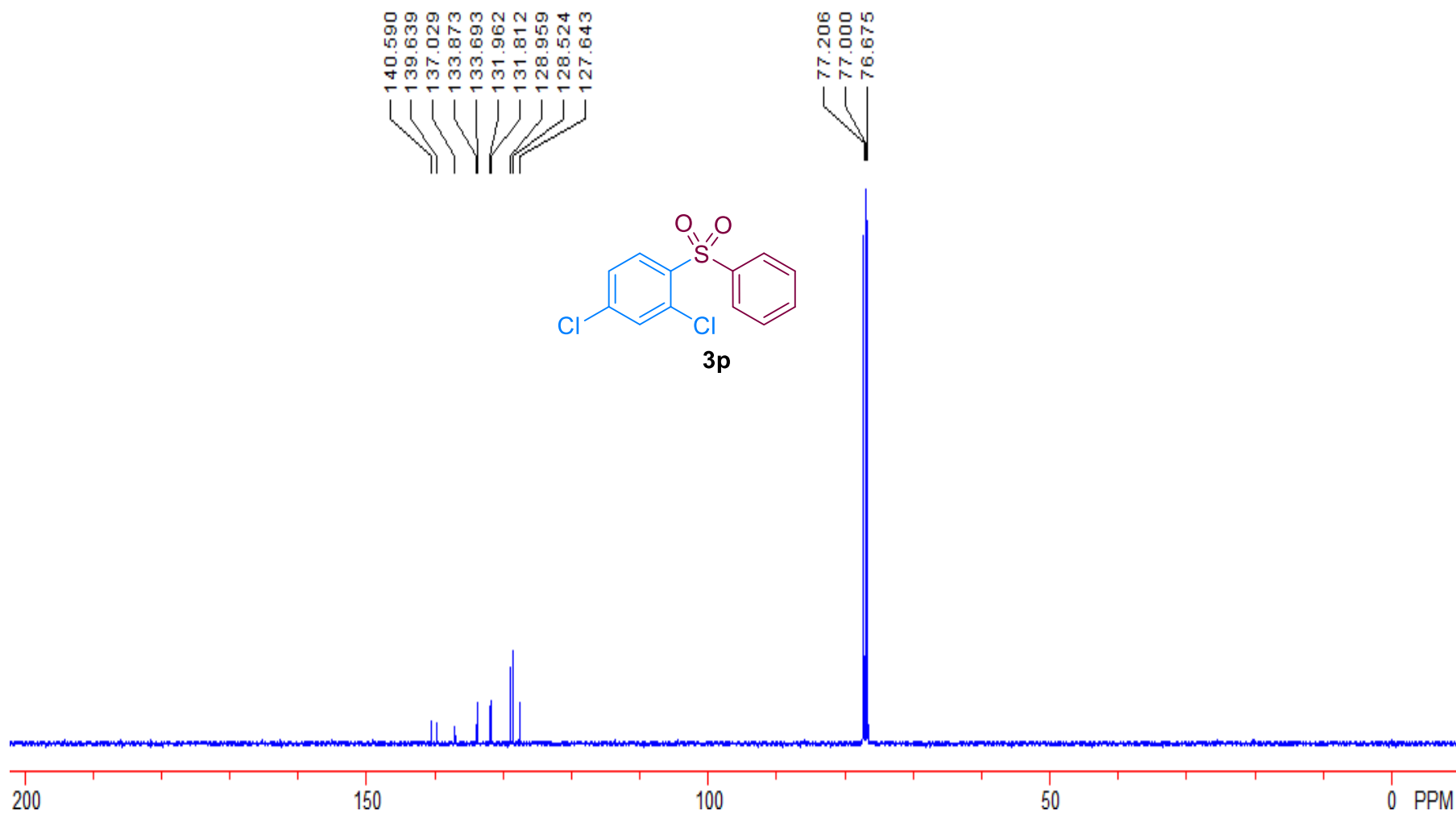


8.316
8.294
7.950
7.936
7.932
7.928
7.629
7.610
7.548
7.528
7.510
7.491
7.486
7.470
7.465
7.450
7.445
7.265



0.000

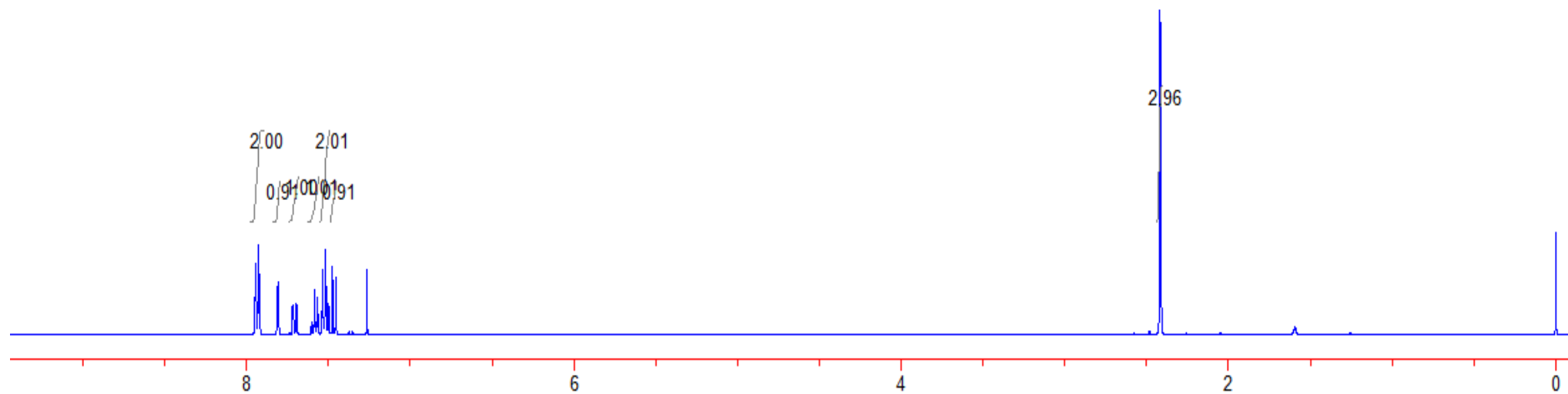
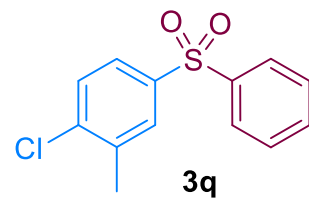




7.945
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7.927
7.923
7.812
7.806
7.719
7.713
7.698
7.693
7.585
7.570
7.566
7.563
7.537
7.520
7.517
7.499
7.497
7.475
7.454
7.264

2.418
2.412

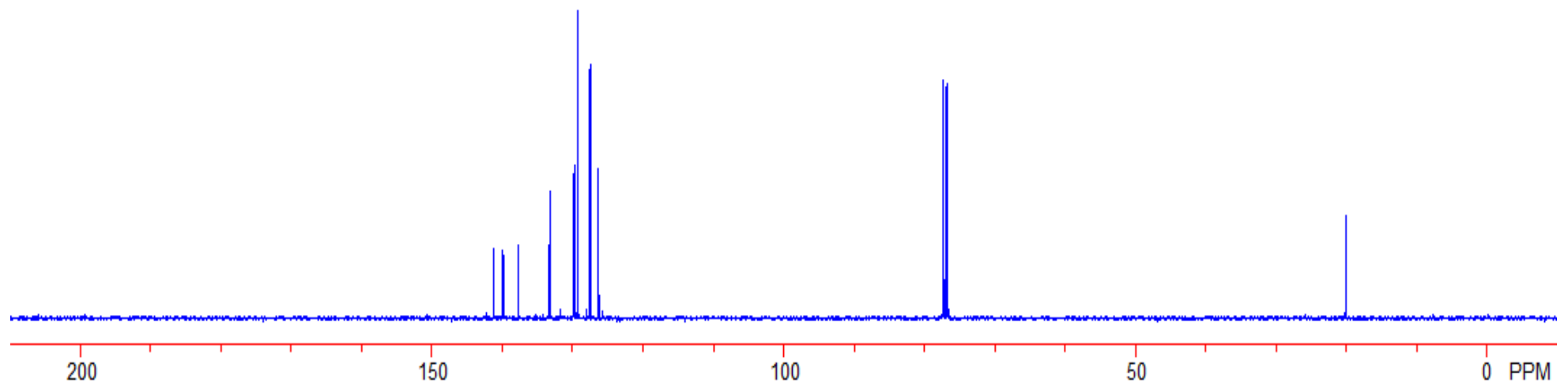
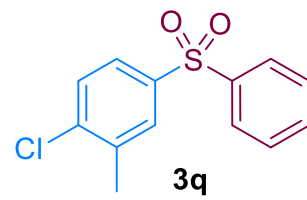
-0.000



141.264
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139.780
137.733
133.278
129.942
129.747
129.309
127.560
127.510
126.297

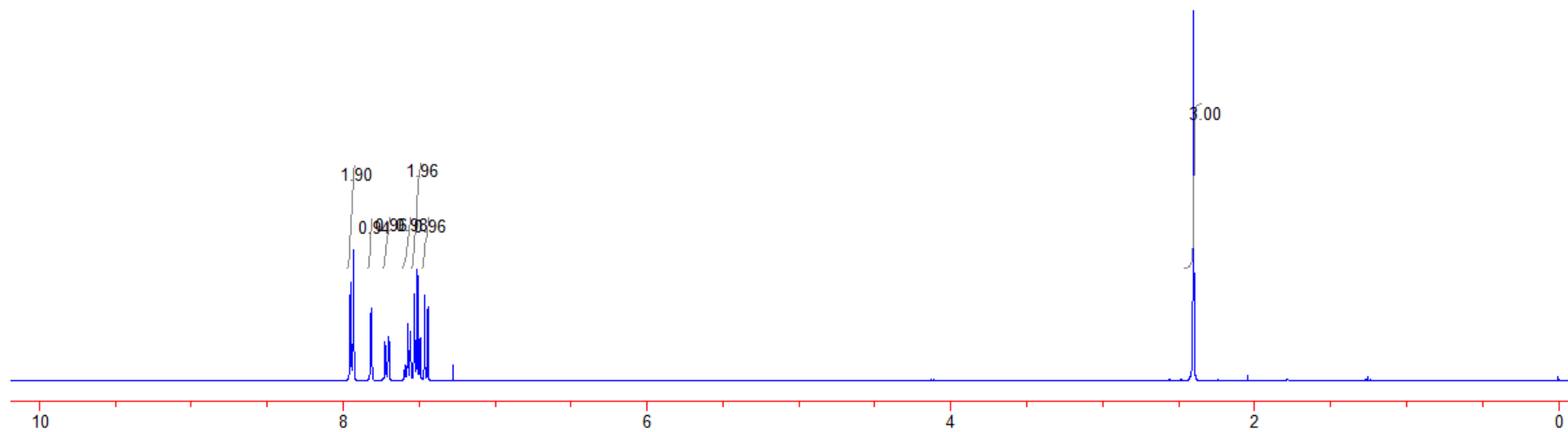
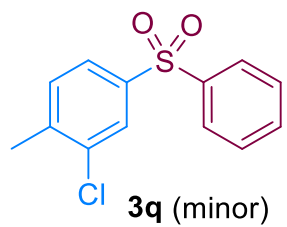
77.326
77.000
76.690

20.155



7.959
7.954
7.951
7.946
7.938
7.933
7.929
7.818
7.813
7.724
7.719
7.703
7.702
7.698
7.697
7.594
7.576
7.570
7.561
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7.510
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7.493
7.490
7.463
7.442
7.277

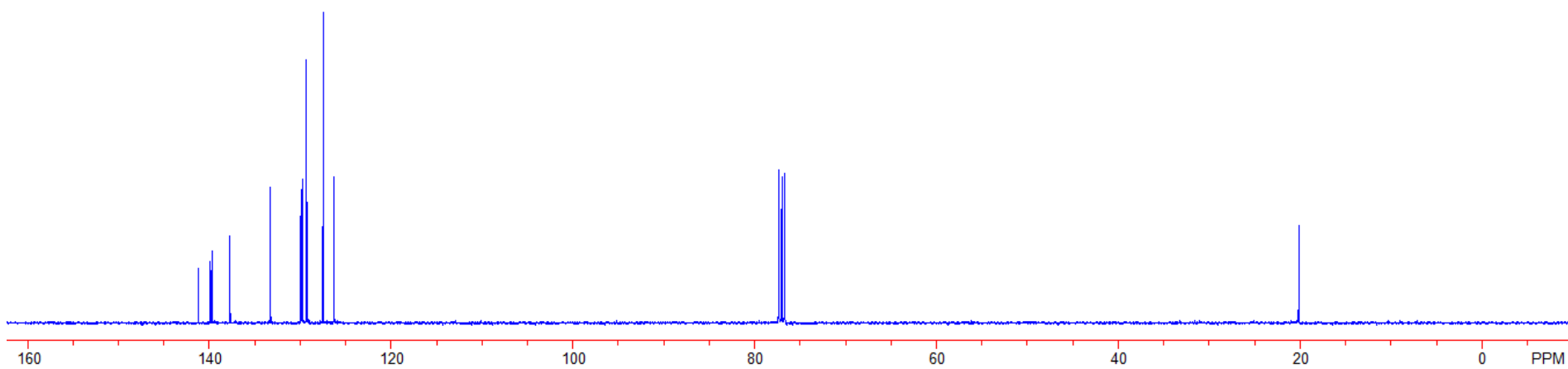
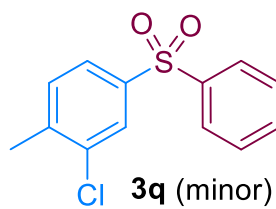
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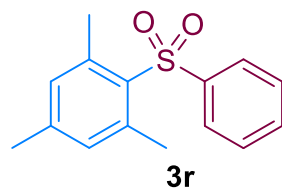
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139.909
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137.716
133.275
129.913
129.717
129.292
127.487
126.269

77.326
77.000
76.690

20.136

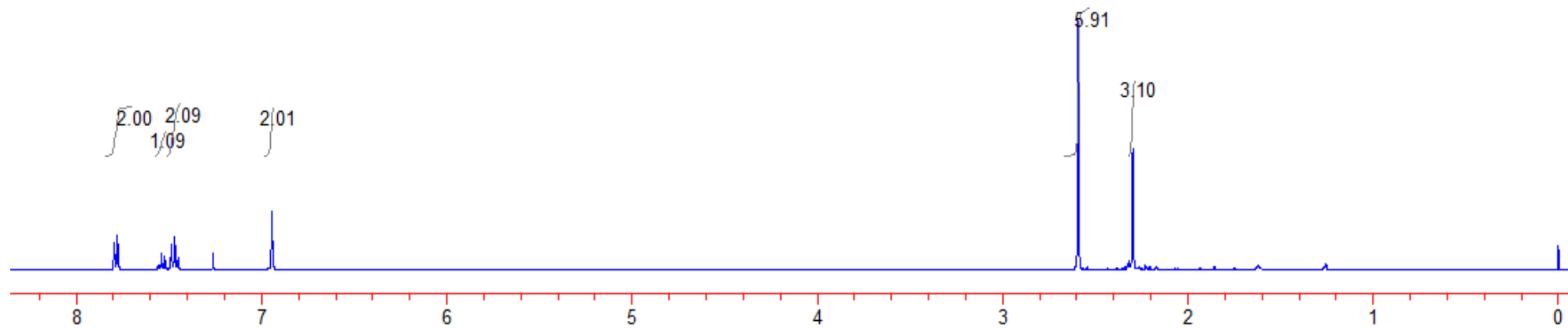


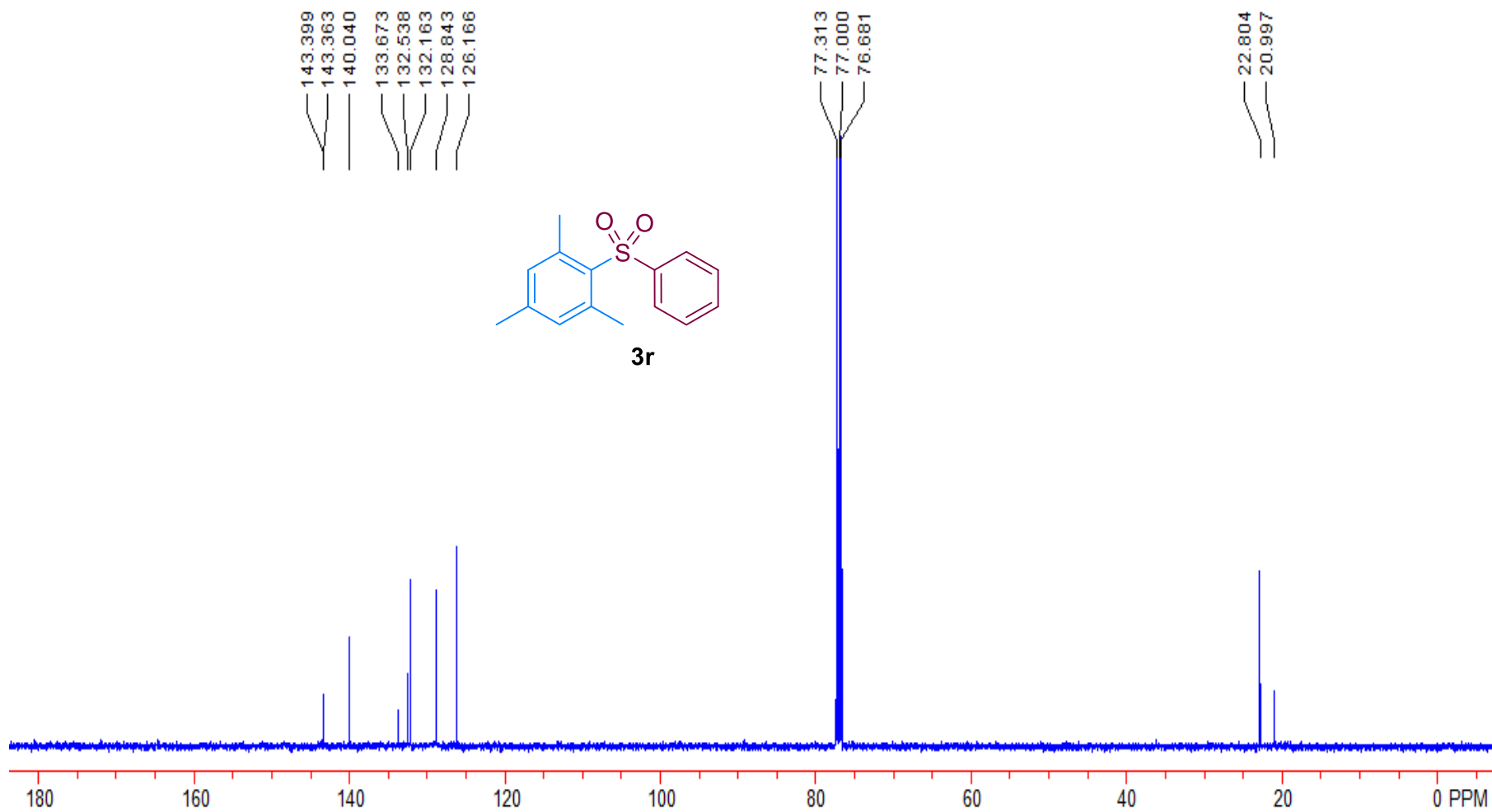
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7.777
7.774
7.540
7.487
7.471
7.467
7.261
6.943



2.592
2.297

-0.000



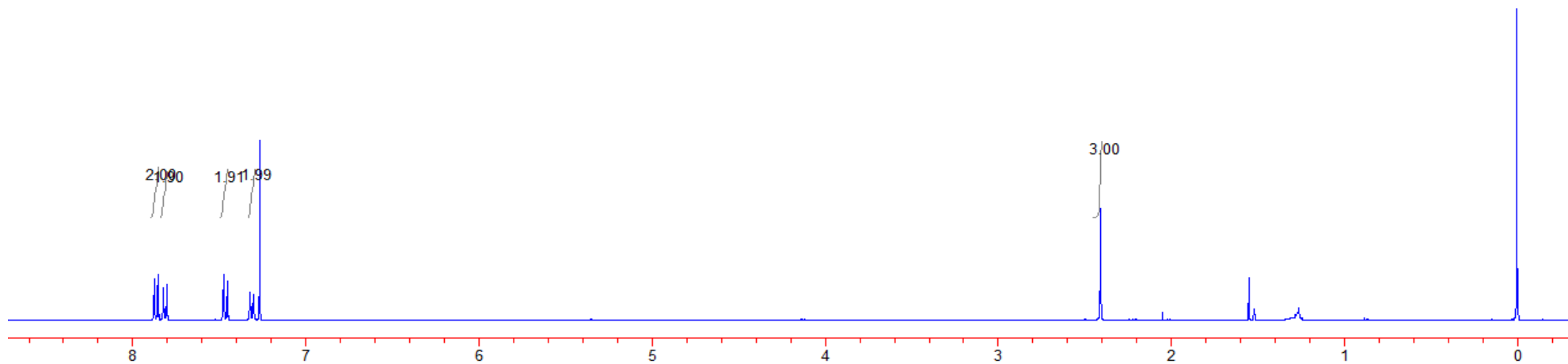
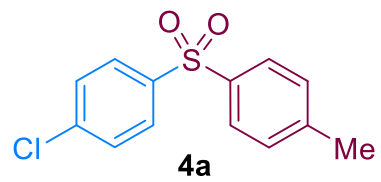


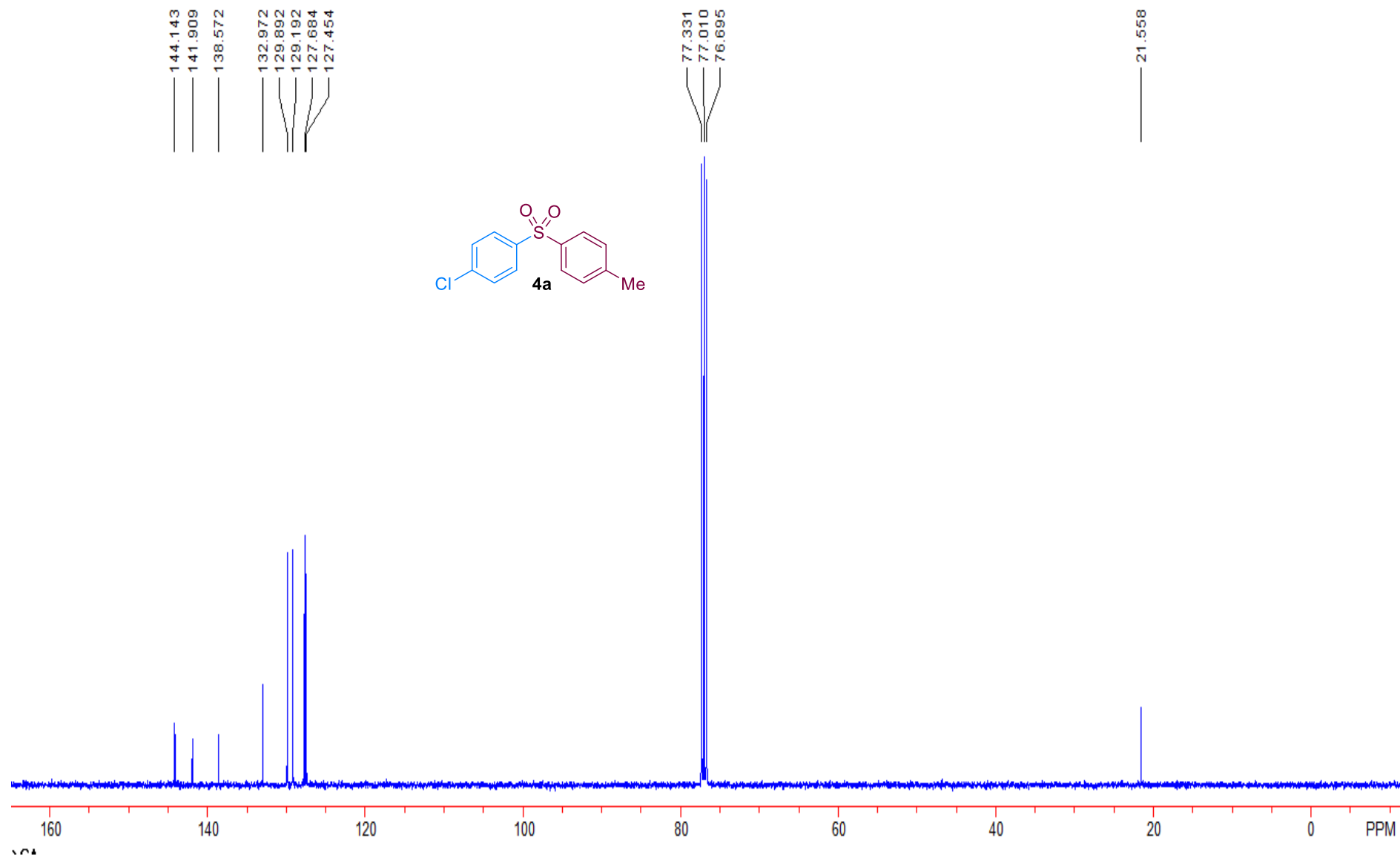
7.871
7.866
7.854
7.849
7.818
7.798
7.471
7.465
7.454
7.448
7.317
7.297
7.261

2.405

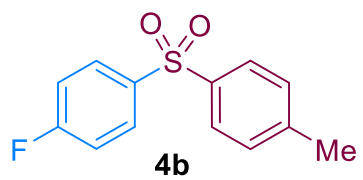
1.548

-0.000



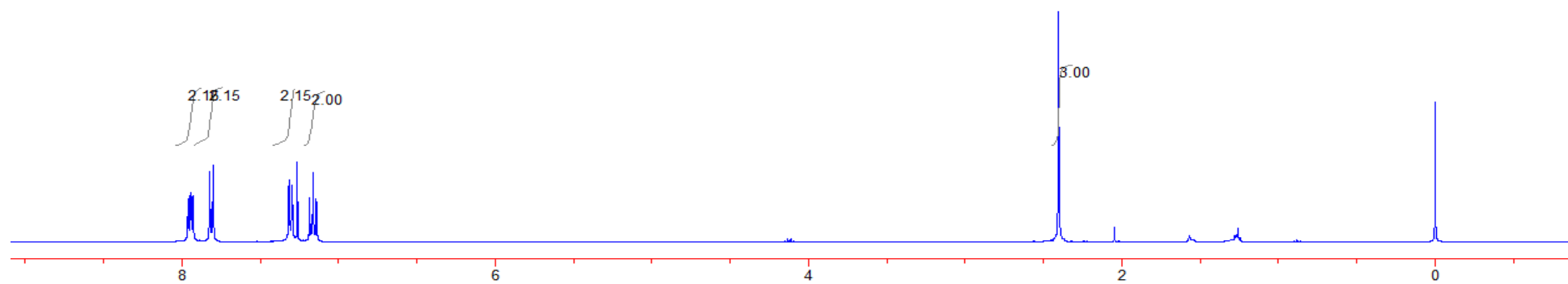


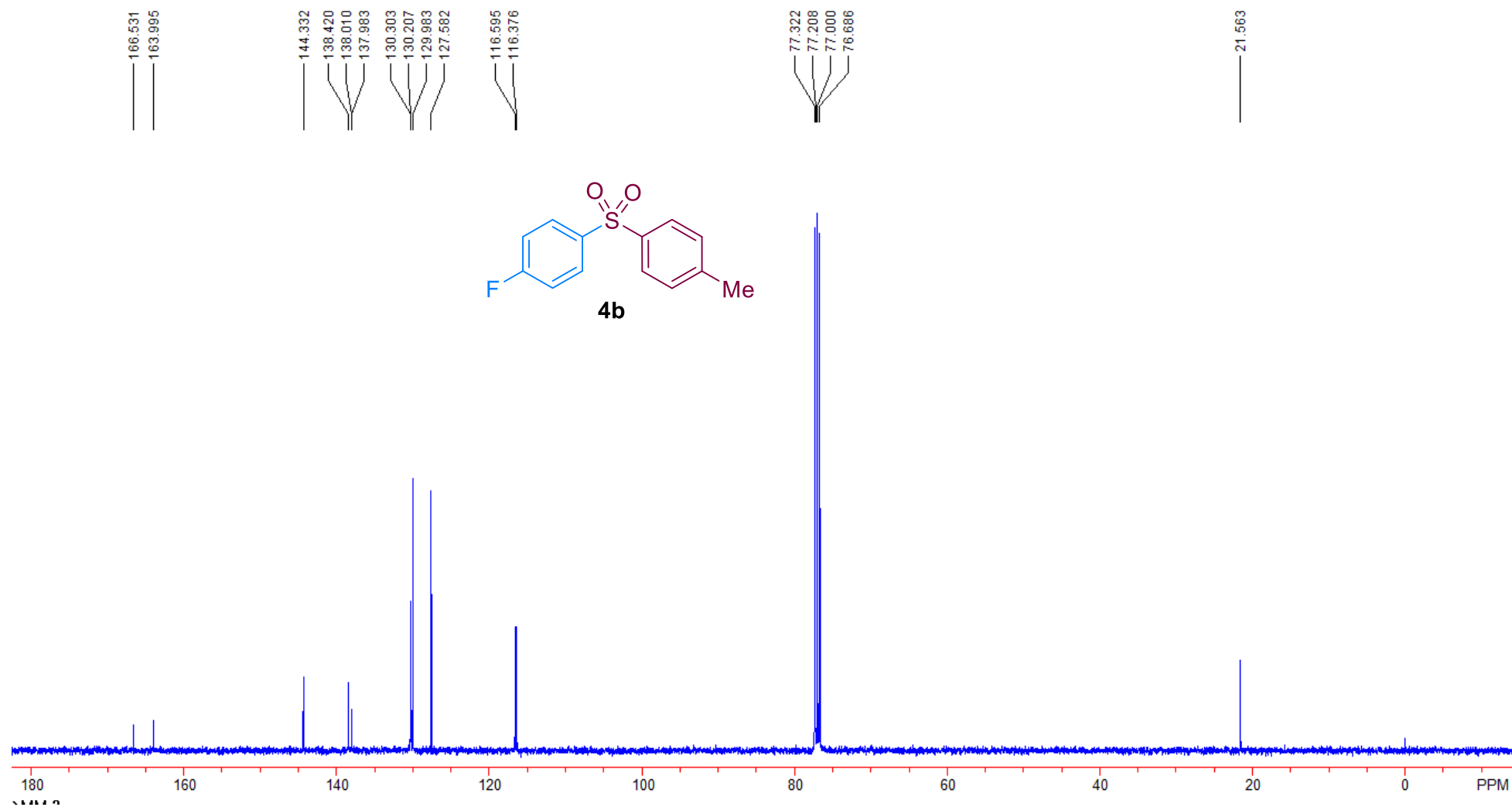
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7.927
7.822
7.801
7.316
7.296
7.283
7.184
7.163
7.142

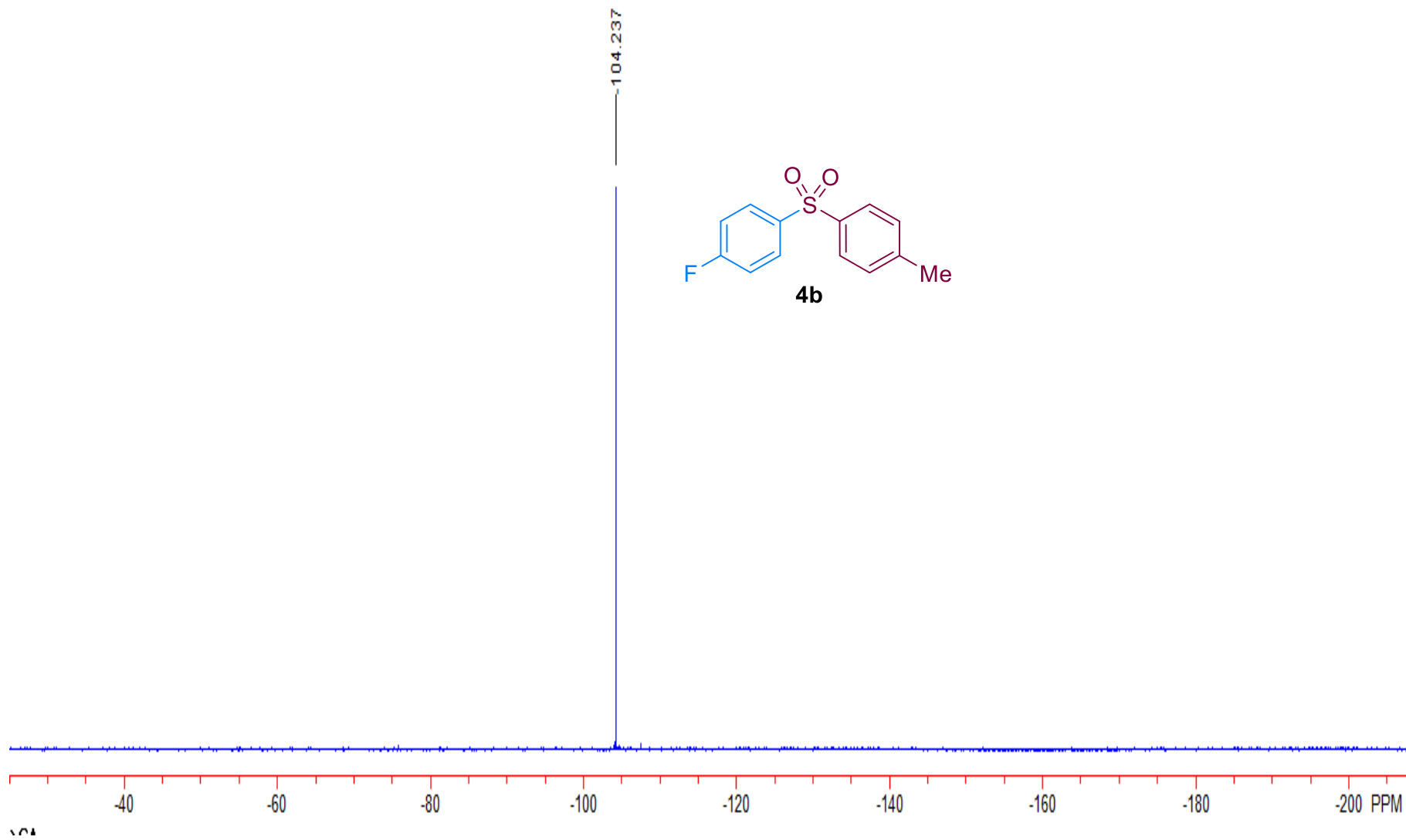


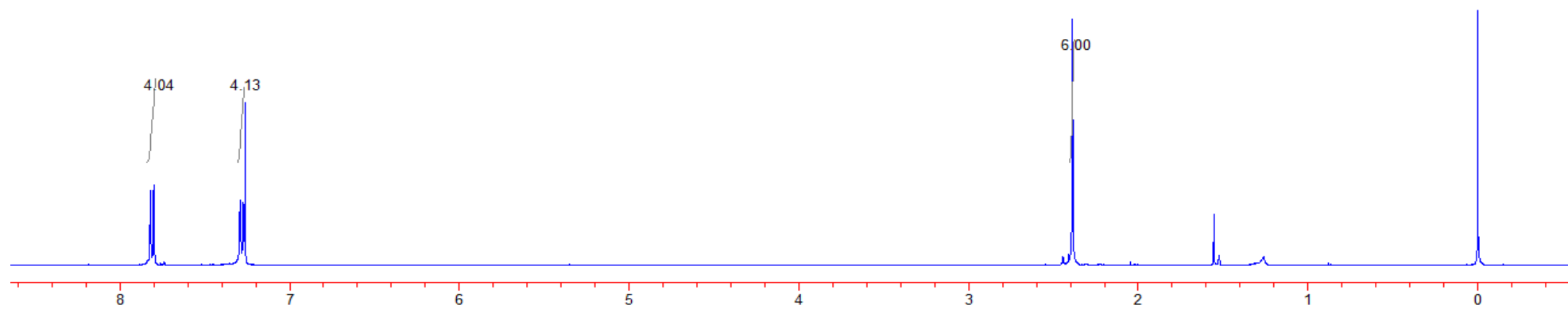
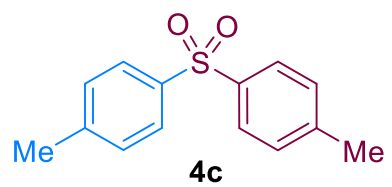
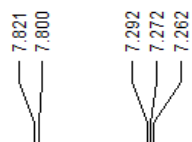
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-0.000





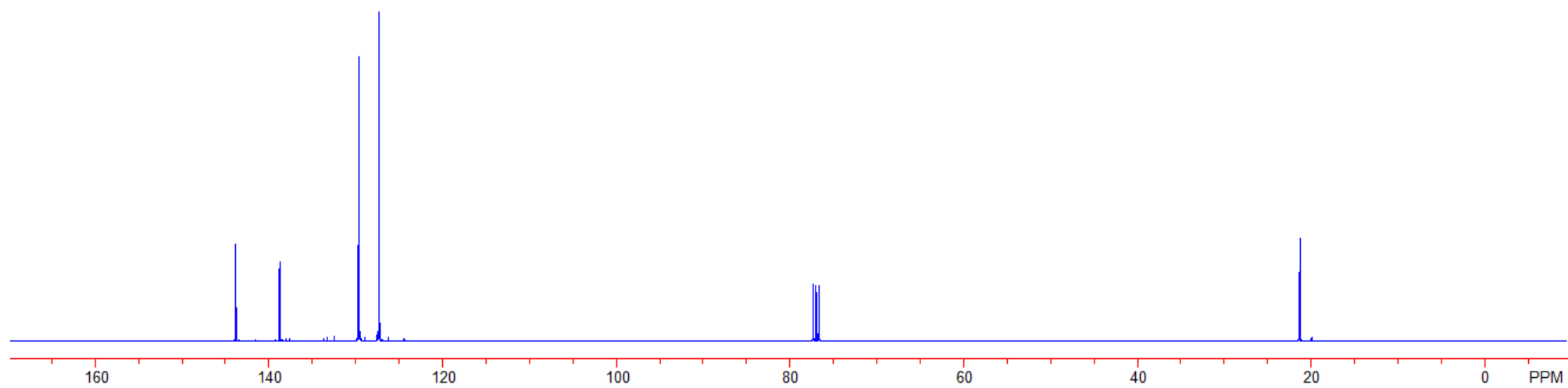
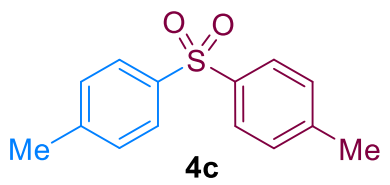




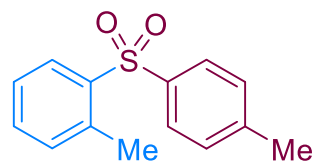
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129.669
127.283

77.325
77.000
76.687

21.322



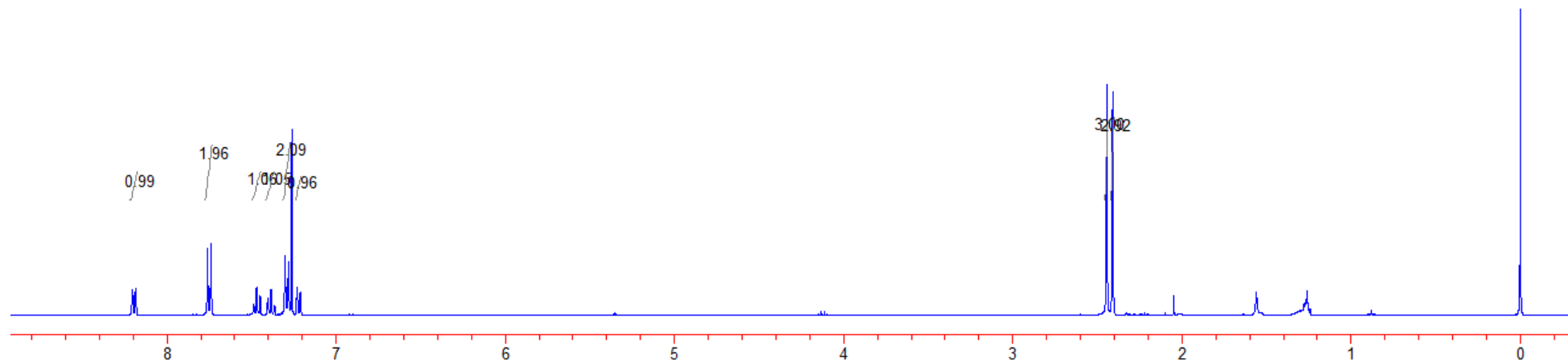
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8.202
8.186
8.183
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7.466
7.384
7.302
7.282
7.262
7.230



4c (minor)

2.444
2.410

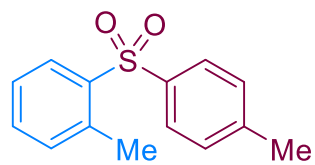
-0.000



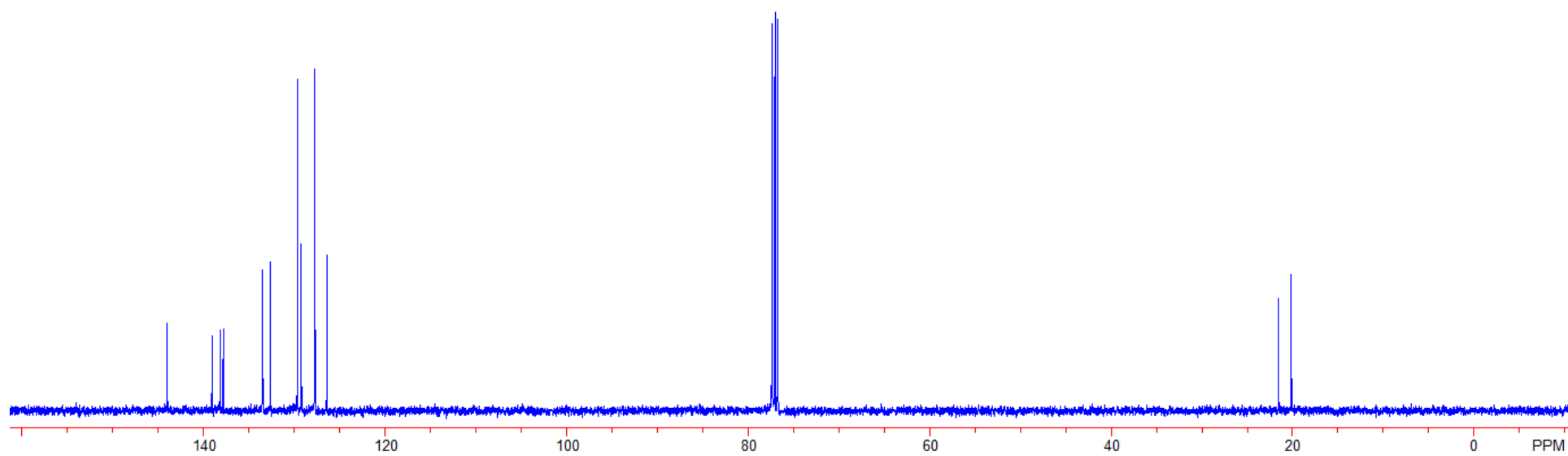
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127.657
126.352

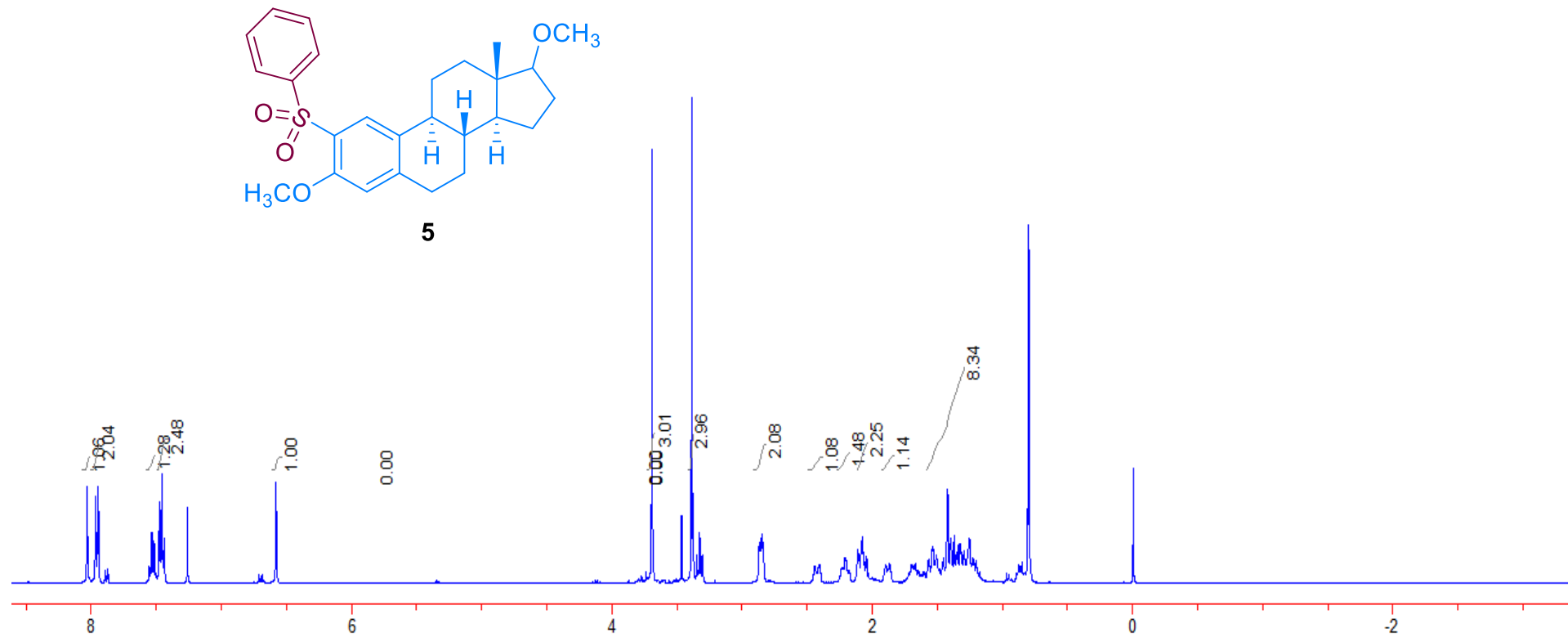
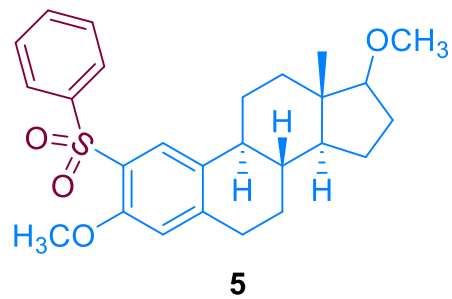
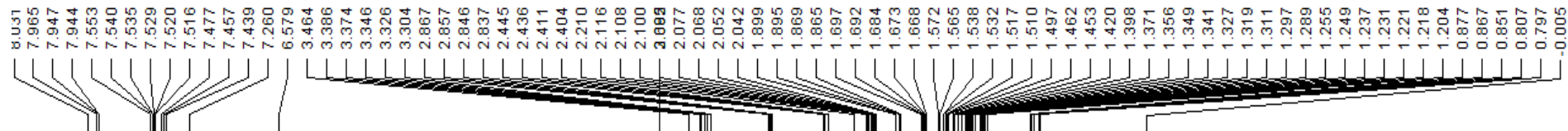
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77.000
76.681

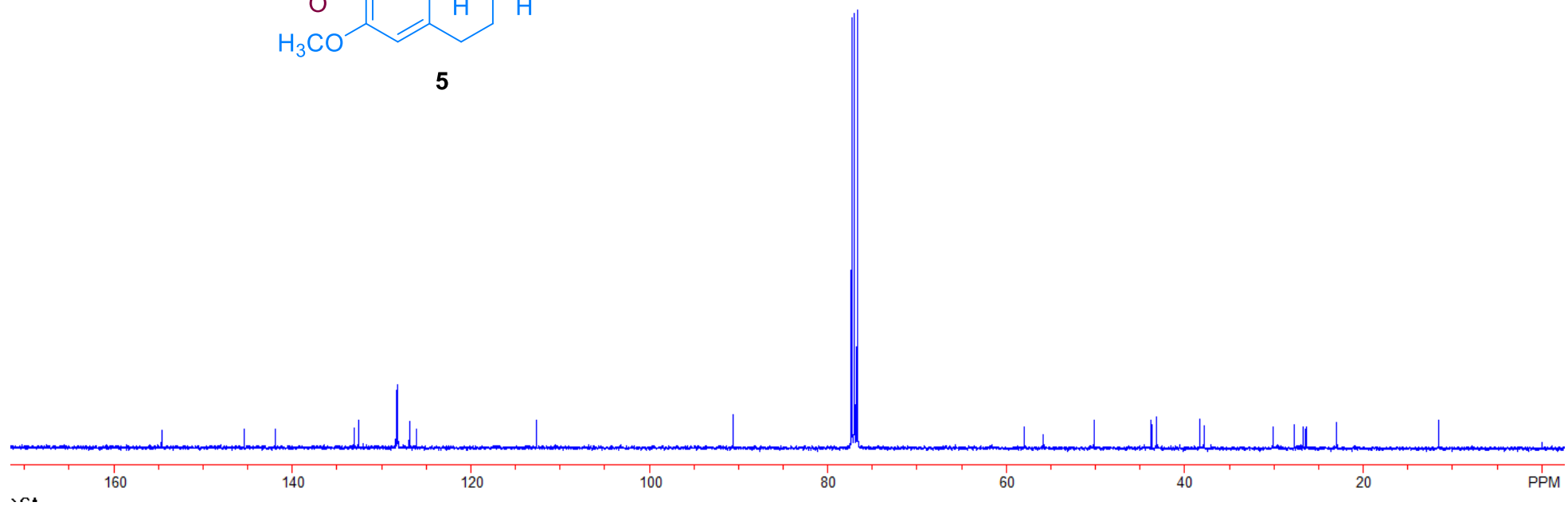
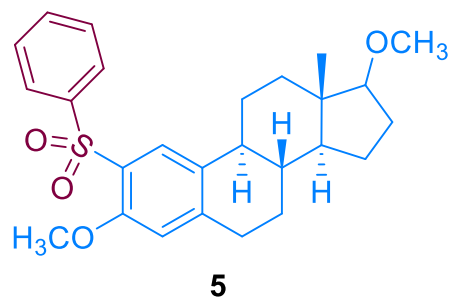
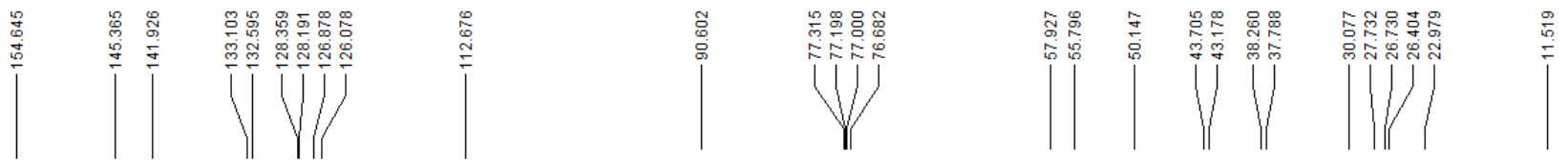
21.521
20.129



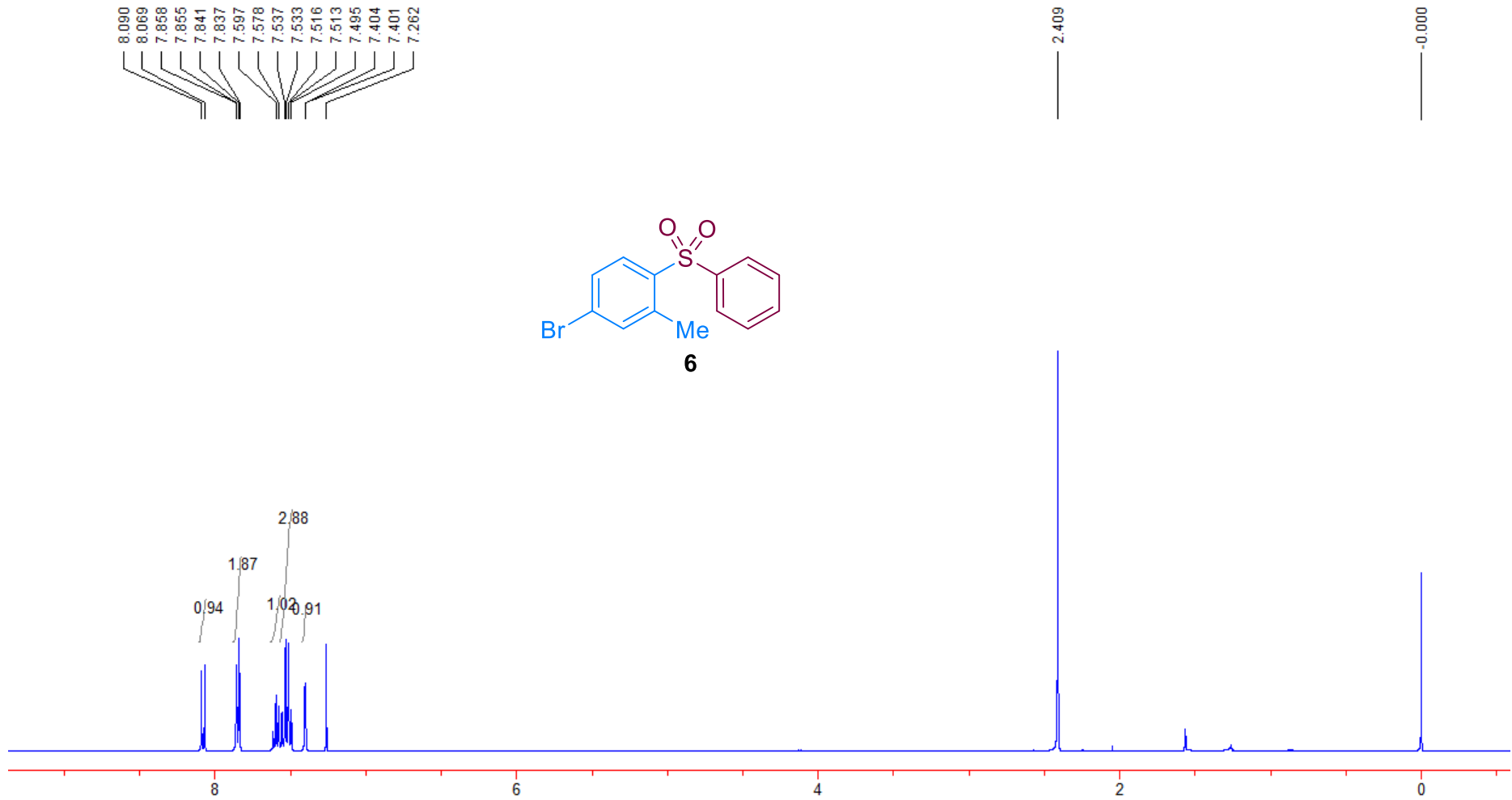
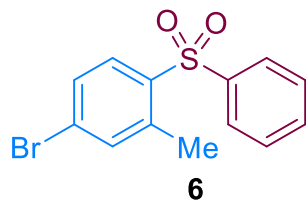
4c (minor)







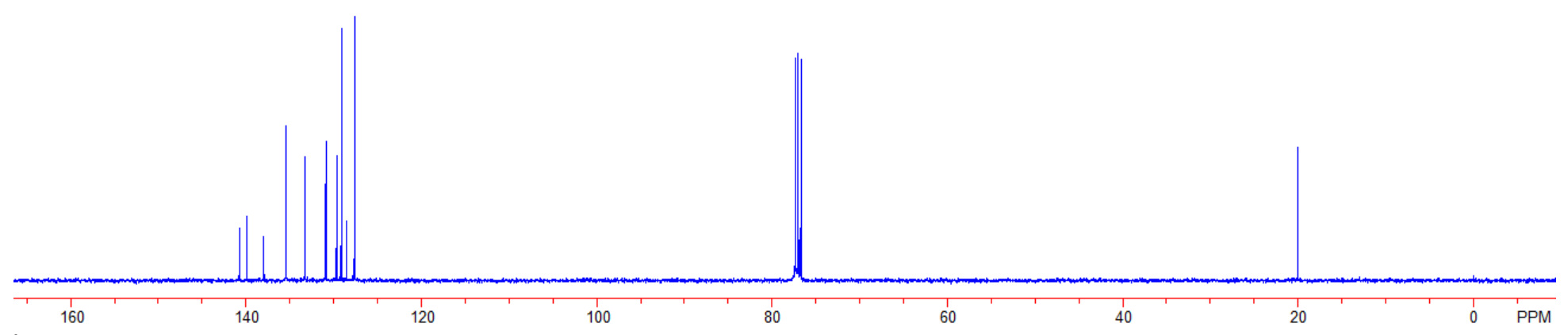
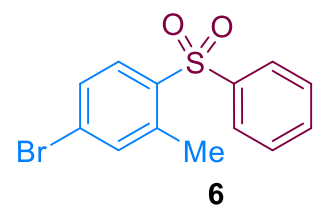
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8.069
7.858
7.855
7.841
7.837
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7.578
7.537
7.533
7.516
7.513
7.495
7.404
7.401
7.262

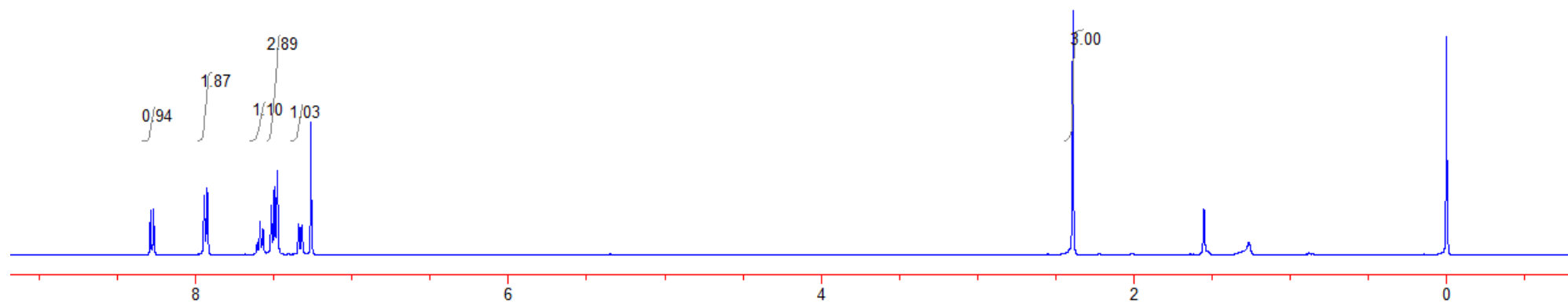
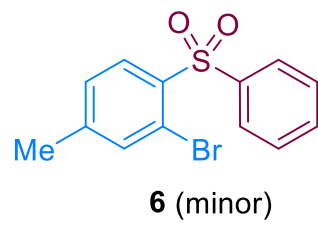
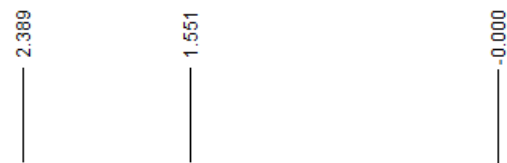
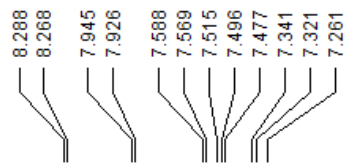


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137.960
135.421
133.249
130.890
129.653
129.112
128.497
127.611

77.318
77.000
76.682

19.379





145.974
140.178
136.884
136.000
133.182
131.334
128.726
128.519
128.430
120.882

77.318
77.000
76.883

21.041

