

## Electronic Supporting Information

### *N*-Alkylation of Substituted Alcohols through a Self-Supported Mesoporous CuMn<sub>2</sub>O<sub>4</sub>

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**Figure S1.** (a) Small angle XRD pattern, (b) TEM image, (c) Nitrogen sorption isotherms, and (c inset) its corresponding pore size distribution curve of the mesoporous silica KIT-6.

**Figure S2.** (a) Wide-angle XRD pattern of self-supported CuMn<sub>2</sub>O<sub>4</sub>, (b) Wide-angle XRD patterns of CuMn<sub>2</sub>O<sub>4</sub> without using KIT-6, (c) TEM image of CuMn<sub>2</sub>O<sub>4</sub> without using KIT-6.

**Figure S3.** Pore size distribution curve of mesoporous CuMn<sub>2</sub>O<sub>4</sub>.

**Figure S4.** The <sup>1</sup>H NMR spectrum of mixtures containing 4-methyl-*N*-(4-methylbenzyl)-benzenesulfonamide and *N*-benzyl-4-methylbenzenesulfonamide.

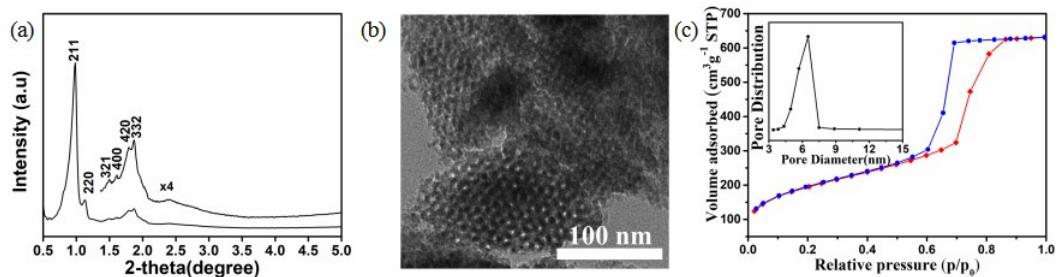
**Figure S5.** Wide-angle XRD pattern of self-supported CuMn<sub>2</sub>O<sub>4</sub> after 5 cycle experiments.

**Figure S6.** TEM image of self-supported CuMn<sub>2</sub>O<sub>4</sub> after 5 cycle experiments.

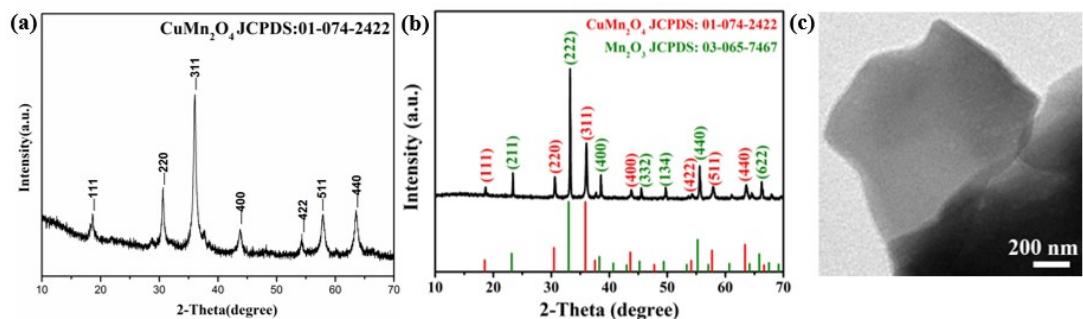
**Table S1.** Optimization of reaction conditions.

**Table S2.** Comparison of the *N*-alkylation of *p*-toluenesulfonamide and benzyl alcohol for mesoporous CuMn<sub>2</sub>O<sub>4</sub> in our study and some catalysts reported in the literature with the optimized conditions.

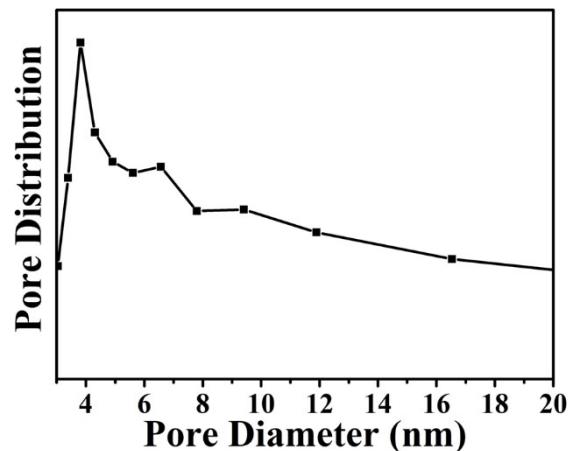
## Characterization of the products



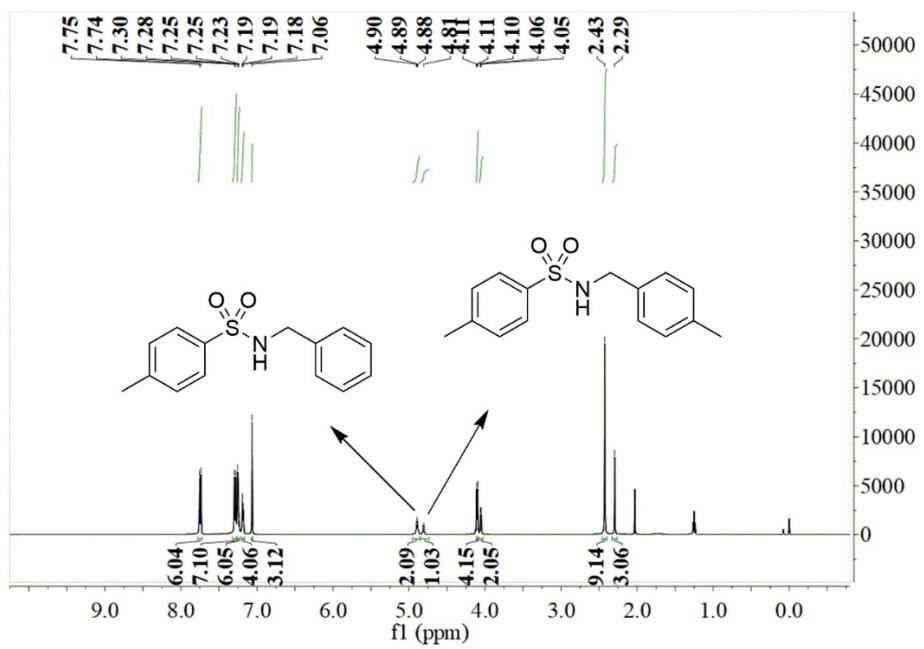
**Figure S1.** (a) Small angle XRD pattern, (b) TEM image, (c) Nitrogen sorption isotherms, and (c inset) its corresponding pore size distribution curve of the mesoporous silica KIT-6.



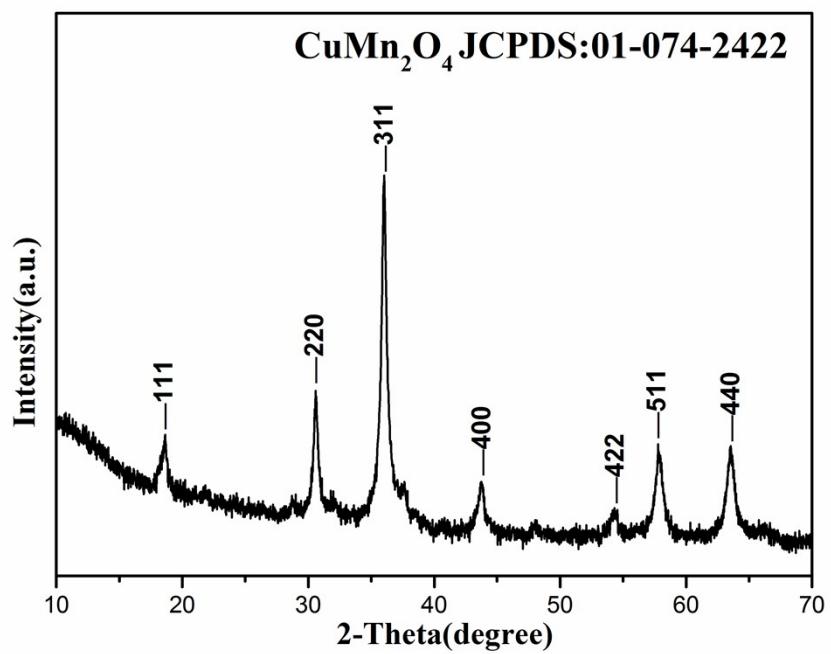
**Figure S2.** (a) Wide-angle XRD pattern of self-supported  $\text{CuMn}_2\text{O}_4$ , (b) Wide-angle XRD patterns of  $\text{CuMn}_2\text{O}_4$  without using KIT-6, (c) TEM image of  $\text{CuMn}_2\text{O}_4$  without using KIT-6.



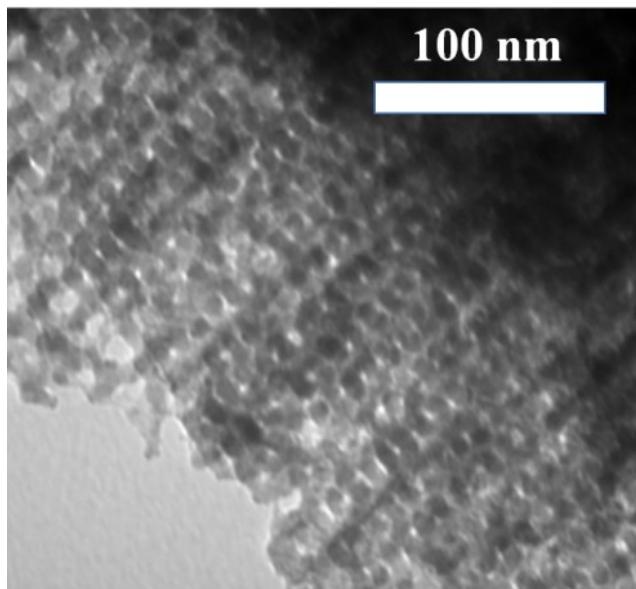
**Figure S3.** Pore size distribution curve of mesoporous  $\text{CuMn}_2\text{O}_4$ .



**Figure S4.** The  $^1\text{H}$  NMR spectrum of mixtures containing 4-methyl-N-(4-methylbenzyl)-benzenesulfonamide and *N*-benzyl-4-methylbenzenesulfonamide.

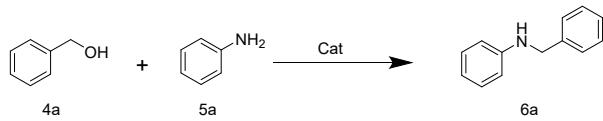


**Figure S5.** Wide-angle XRD pattern of self-supported  $\text{CuMn}_2\text{O}_4$  after 5 cycle experiments.



**Figure S6.** TEM image of self-supported CuMn<sub>2</sub>O<sub>4</sub> after 5 cycle experiments.

**Table S1.** Optimization of reaction conditions.



Entry <sup>a</sup>	a(mmol)	b(mmol)	Temperature (°C)	Time (h)	<i>t</i> -BuOK (mol%)	CuMn <sub>2</sub> O <sub>4</sub> (mol%)	Yield (%)
1	0.6	0.4	100	2	10	3	80
2	0.4	0.4	100	2	10	3	65
3	0.4	0.6	100	2	10	3	44
4	0.6	0.4	120	2	10	3	86
5	0.6	0.4	140	2	10	3	85
6	0.6	0.4	120	4	10	3	93
7	0.6	0.4	120	6	10	3	92
8	0.6	0.4	120	4	0	3	<5%
9 <sup>b</sup>	0.6	0.4	120	4	20	3	99
10	0.6	0.4	120	8	20	0	trace
11	0.6	0.4	120	4	20	2	85

<sup>a</sup>The yields were determined by high performance liquid chromatography (HPLC).

<sup>b</sup>The isolated yield was 95%.

**Table S2.** Comparison of the *N*-alkylation of *p*-toluenesulfonamide and benzyl alcohol for mesoporous CuMn<sub>2</sub>O<sub>4</sub> in our study and some catalysts reported in the literature with the optimized conditions.

Entry	Cat.	Cat. (mol%)	base	Temperature (°C)	Time (h)	Yield (%)	Ref.
1	Pd(OAc) <sub>2</sub>	0.01	K <sub>2</sub> CO <sub>3</sub>	150	8	88	<sup>1</sup>
2	FeCl <sub>2</sub>	5	K <sub>2</sub> CO <sub>3</sub>	135	20	98	<sup>2</sup>
3	FeCl <sub>3</sub> ·6H <sub>2</sub> O	1	K <sub>2</sub> CO <sub>3</sub>	135	12	10 (con.)	<sup>2</sup>
4	Fe <sub>3</sub> (CO)₁₂	1	K <sub>2</sub> CO <sub>3</sub>	135	12	19 (con.)	<sup>2</sup>
5	Fe(acac) <sub>3</sub>	1	K <sub>2</sub> CO <sub>3</sub>	135	12	25 (con.)	<sup>2</sup>
6	Ru/Fe <sub>3</sub> O <sub>4</sub>	40 mg	K <sub>2</sub> CO <sub>3</sub>	150	12	97	<sup>3</sup>
7	Pd(COD)ClSnCl <sub>3</sub>	3	<i>o</i> -xylene	140	24	75	<sup>4</sup>
8	AgOTf	5	-	100	8	74	<sup>5</sup>
9	Ni/SiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub>	10	K <sub>2</sub> CO <sub>3</sub>	175	60	63	<sup>6</sup>
10	Cu(AcO) <sub>2</sub>	1	<i>t</i> -BuOK	150	6	40	<sup>7</sup>
<b>11</b>	<b>CuMn<sub>2</sub>O<sub>4</sub></b>	<b>2.5</b>	<b>K<sub>2</sub>CO<sub>3</sub></b>	<b>135</b>	<b>5</b>	<b>99</b>	<b>This work</b>

## Characterization of the products

***N*-Benzyl-4-methylbenzenesulfonamide** (3a)<sup>8</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.73 (d, *J* = 8.2 Hz, 2H), 7.36 – 7.20 (m, 5H), 7.20 – 7.09 (m, 2H), 5.06 (s, 1H), 4.09 (d, *J* = 6.2 Hz, 2H), 2.41 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 143.50 (s), 136.88 (s), 136.40 (s), 129.75 (s), 128.66 (s), 127.90 (s), 127.83 (s), 127.20 (s), 47.23 (s), 21.56 (s).

***N*-(3-Chlorobenzyl)-4-methylbenzenesulfonamide** (3b)<sup>9</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.71 (d, *J* = 8.2 Hz, 2H), 7.27 (d, *J* = 8.0 Hz, 2H), 7.22 – 7.14 (m, 2H), 7.12 (s, 1H), 7.08 (dd, *J* = 4.8, 0.8 Hz, 1H), 5.29 (t, 1H), 4.07 (d, *J* = 6.4 Hz, 2H), 2.42 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 143.72 (s), 138.48 (s), 136.73 (s), 134.41 (s), 129.90 (s), 129.80 (s), 127.92 (d, *J* = 3.0 Hz), 127.13 (s), 125.98 (s), 46.58 (s), 21.57 (s).

**4-Methyl-*N*-(4-methylbenzyl)benzenesulfonamide** (3c)<sup>10</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.73 (d, *J* = 8.3 Hz, 2H), 7.27 (d, *J* = 8.1 Hz, 2H), 7.08 – 7.02 (m, 4H), 5.02 (d, *J* = 5.7 Hz, 1H), 4.03 (d, *J* = 6.2 Hz, 2H), 2.41 (s, 3H), 2.28 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 143.44 (s), 137.56 (s), 136.87 (s), 133.34 (s), 129.73 (s), 129.32 (s), 127.90 (s), 127.22 (s), 47.01 (s), 21.57 (s), 21.12 (s).

***N*-(2-Methoxybenzyl)-4-methylbenzenesulfonamide** (3d)<sup>10</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.66 (d, *J* = 8.2 Hz, 2H), 7.22 – 7.16 (m, 3H), 7.06 (dd, 1H), 6.79 (t, *J* = 7.4 Hz, 1H), 6.72 (d, *J* = 8.2 Hz, 1H), 5.17 (t, *J* = 5.9 Hz, 1H), 4.13 (d, *J* = 6.3 Hz, 2H), 3.73 (s, 3H), 2.38 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 157.22 (s), 143.05 (s), 137.25 (s), 129.79 (s), 129.40 (s), 129.22 (s), 127.08 (s), 124.33 (s), 120.54 (s), 110.11 (s), 55.18 (s), 43.99 (s), 21.49 (s).

**N-(4-Methoxybenzyl)-4-methylbenzenesulfonamide** (3e)<sup>9</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.74 (d, *J* = 8.2 Hz, 2H), 7.29 (d, *J* = 8.1 Hz, 2H), 7.09 (d, *J* = 8.6 Hz, 2H), 6.78 (d, *J* = 8.6 Hz, 2H), 4.85 (t, *J* = 5.8 Hz, 1H), 4.03 (d, *J* = 6.1 Hz, 2H), 3.76 (s, 3H), 2.43 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 159.26 (s), 143.46 (s), 136.89 (s), 129.74 (s), 129.29 (s), 128.33 (s), 127.20 (s), 114.04 (s), 55.30 (s), 46.77 (s), 21.56 (s).

**N-(3-Bromobenzyl)-4-methylbenzenesulfonamide** (3f)<sup>11</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.70 (d, *J* = 8.2 Hz, 2H), 7.32 (d, *J* = 7.2 Hz, 1H), 7.26 (d, *J* = 8.5 Hz, 3H), 7.13 – 7.08 (m, 2H), 5.32 (t, *J* = 6.3 Hz, 1H), 4.07 (d, *J* = 6.4 Hz, 2H), 2.42 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 143.72 (s), 138.76 (s), 136.73 (s), 130.83 (d, *J* = 2.4 Hz), 130.18 (s), 129.82 (s), 127.12 (s), 126.47 (s), 122.59 (s), 46.53 (s), 21.60 (s).

**N-(4-Bromobenzyl)-4-methylbenzenesulfonamide** (3g)<sup>10</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.70 (d, *J* = 7.8 Hz, 2H), 7.36 (d, *J* = 7.9 Hz, 2H), 7.27 (d, *J* = 8.0 Hz, 2H), 7.06 (d, *J* = 8.0 Hz, 2H), 5.19 (t, *J* = 6.2 Hz, 1H), 4.05 (d, *J* = 6.3 Hz, 2H), 2.43 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 143.70 (s), 136.74 (s), 135.47 (s), 131.71 (s), 129.79 (s), 129.58 (s), 127.13 (s), 121.75 (s), 46.57 (s), 21.59 (s).

**N-Benzyl-4-bromobenzenesulfonamide** (3h)<sup>10</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.67 (d, *J* = 8.6 Hz, 2H), 7.59 (d, *J* = 8.6 Hz, 2H), 7.30 – 7.22 (m, 3H), 7.16 (dd, *J* = 6.8, 2.5 Hz, 2H), 5.17 (t, *J* = 6.1 Hz, 1H), 4.12 (d, *J* = 6.2 Hz, 2H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 139.02 (s), 135.96 (s), 132.38 (s), 128.75 (s), 128.67 (s), 128.02 (s), 127.91 (s), 127.64 (s), 47.26 (s).

**4-Bromo-N-(3-chlorobenzyl)benzenesulfonamide** (3i)<sup>12</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.67 (d, *J* = 8.6 Hz, 2H), 7.62 (d, *J* = 8.5 Hz, 2H), 7.25 – 7.17 (m, 2H), 7.14 (s, 1H), 7.07 (d, *J* = 7.0 Hz, 1H), 5.15 (t, *J* = 6.1 Hz, 1H), 4.12 (d, *J* = 6.3 Hz, 2H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 138.93 (s), 137.99 (s), 134.60 (s), 132.46 (s), 130.03 (s), 128.60 (s), 128.17 (s), 127.98 (s), 127.89 (s), 125.95 (s), 46.66 (s).

**4-Bromo-N-(4-methylbenzyl)benzenesulfonamide** (3j)<sup>11</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.67 (d, *J* = 8.6 Hz, 2H), 7.59 (d, *J* = 8.6 Hz, 2H), 7.09 – 7.02 (m, 4H), 5.02 (t, *J* = 6.0 Hz, 1H), 4.08 (d, *J* = 6.1 Hz, 2H), 2.30 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 139.10 (s), 137.85 (s), 132.87 (s), 132.34 (s), 129.40 (s), 128.69 (s), 127.89 (s), 127.57 (s), 47.07 (s), 21.11 (s).

**4-Bromo-N-(2-methoxybenzyl)benzenesulfonamide** (3k), <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.58 (d, *J* = 8.5 Hz, 2H), 7.48 (d, *J* = 8.5 Hz, 2H), 7.20 (td, *J* = 8.1, 1.5 Hz, 1H), 7.03 (dd, *J* = 7.3, 1.2 Hz, 1H), 6.79 (t, *J* = 7.4 Hz, 1H), 6.70 (d, *J* = 8.2 Hz, 1H), 5.35 (t, *J* = 6.3 Hz, 1H), 4.16 (d, *J* = 6.4 Hz, 2H), 3.72 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 157.15 (s), 139.47 (s), 131.92 (s), 129.87 (s), 129.46 (s), 128.55 (s), 127.13 (s), 123.89 (s), 120.56 (s), 110.15 (s), 55.19 (s), 44.16 (s).

**4-Bromo-N-(4-methoxybenzyl)benzenesulfonamide** (3l)<sup>13</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.68 (d, *J* = 8.6 Hz, 2H), 7.61 (d, *J* = 8.6 Hz, 2H), 7.08 (d, *J* = 8.6 Hz, 2H), 6.78 (d, *J* = 8.6 Hz, 2H), 4.90 (t, *J* = 5.8 Hz, 1H), 4.07 (d, *J* = 6.0 Hz, 2H), 3.77 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 159.42 (s), 139.19 (s), 132.34 (s), 129.29 (s), 128.68 (s), 127.92 (s), 127.55 (s), 114.13 (s), 55.32 (s), 46.83 (s).

**4-Bromo-N-(3-bromobenzyl)benzenesulfonamide** (3m), <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.65 (d, *J* = 8.6 Hz, 2H), 7.60 (d, *J* = 8.6 Hz, 2H), 7.36 (dd, *J* = 3.8, 2.5 Hz, 1H), 7.27 (s, 1H), 7.16 – 7.08 (m, 2H), 5.33 (t, *J* = 6.2 Hz, 1H), 4.11 (d, *J* = 6.3 Hz, 2H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 138.90 (s), 138.28 (s), 132.46 (s), 131.04 (s), 130.89 (s), 130.28 (s), 128.58 (s), 127.90 (s), 126.45 (s), 122.71 (s), 46.57 (s).

**N-Benzylidene-p-toluenesulfonamide** (product of **Scheme 4b**)<sup>11</sup>, <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 9.03 (s, 1H), 7.91 (dd, *J* = 17.2, 8.0 Hz, 4H), 7.61 (t, *J* = 7.4 Hz, 1H), 7.49 (t, *J* = 7.6 Hz, 2H), 7.35 (d, *J* = 8.0 Hz, 2H), 2.44 (s, 3H); <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 170.15 (s), 144.63 (s), 135.16 (s), 134.95 (s), 132.41 (s), 131.33 (s), 129.83 (s), 129.16 (s), 128.12 (s), 21.67 (s).

**N-Benzylaniline** (6a)<sup>14</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.35 (d, *J* = 7.5 Hz, 2H), 7.31 (t, *J* = 7.5 Hz, 2H), 7.22 (d, *J* = 7.2 Hz, 1H), 7.02 (t, *J* = 7.8 Hz, 2H), 6.56 (d, *J* = 7.9 Hz, 2H), 6.50 (t, *J* = 7.2 Hz, 1H), 6.20 (s, 1H), 4.25 (d, *J* = 6.0 Hz, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 149.14 (s), 140.79 (s), 129.27 (s), 128.73 (s), 127.64 (s), 127.05 (s), 116.18 (s), 112.74 (s), 46.92 (s).

**N-Benzyl-4-bromoaniline** (6b)<sup>15</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.37 – 7.28 (m, 4H), 7.22 (t, *J* = 6.6 Hz, 1H), 7.16 (d, *J* = 8.8 Hz, 2H), 6.53 (d, *J* = 8.8 Hz, 2H), 4.24 (s, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 148.29 (s), 140.17 (s), 131.79 (s), 128.79 (s), 127.64 (s), 127.19 (s), 114.74 (s), 106.89 (s), 46.82 (s).

**N-Benzyl-3-chloroaniline** (6c)<sup>16</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.38 – 7.28 (m, 4H), 7.26 – 7.21 (m, 1H), 7.03 (t, *J* = 8.0 Hz, 1H), 6.57 (t, *J* = 2.0 Hz, 2H), 6.54 – 6.48 (m, 2H), 4.26 (d, *J* = 5.5 Hz, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 150.63 (s), 140.12 (s), 134.02 (s), 130.78 (s), 128.82 (s), 127.64 (s), 127.23 (s), 115.57 (s), 111.85 (s), 111.43 (s), 46.64 (s).

**N-Benzyl-3-methylaniline** (6d)<sup>16</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.36 – 7.28 (m, 4H), 7.21 (t, *J* = 7.2 Hz, 1H), 6.90 (t, *J* = 7.7 Hz, 1H), 6.40 (s, 1H), 6.34 (q, *J* = 15.7, 7.7 Hz, 2H), 6.10 (t, *J* = 6.0 Hz, 1H), 4.24 (d, *J* = 6.1 Hz, 2H), 2.14 (s, 3H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 149.16 (s), 140.91 (s), 138.18 (s), 129.15 (s), 128.70 (s), 127.61 (s), 127.01 (s), 117.16 (s), 113.44 (s), 109.97 (s), 46.91 (s), 21.84 (s).

**N-Benzyl-3-methoxyaniline** (6e)<sup>15</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.44 – 7.27 (m, 4H), 7.21 (t, *J* = 7.1 Hz, 1H), 6.92 (t, *J* = 8.0 Hz, 1H), 6.23 (s, 1H), 6.17 (dd, *J* = 8.1, 1.4 Hz, 1H), 6.13 – 6.05 (m, 2H), 4.23 (d, *J* = 4.2 Hz, 2H), 3.62 (s, 3H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 160.71 (s), 150.47 (s), 140.78 (s), 129.96 (s), 128.72 (s), 127.62 (s), 127.05 (s), 105.92 (s), 101.63 (s), 98.57 (s), 55.02 (s), 46.94 (s).

**N-Phenyl-4-methoxybenzylamine** (6f)<sup>17</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.27 (d, *J* = 8.7 Hz, 2H), 7.02 (dd, *J* = 8.4, 7.3 Hz, 2H), 6.87 (d, *J* = 8.7 Hz, 2H), 6.56 (dd, *J* = 8.5, 0.8 Hz, 2H), 6.49 (t, *J* = 7.3 Hz, 1H), 6.10 (t, *J* = 5.9 Hz, 1H), 4.17 (d, *J* = 6.0 Hz, 2H), 3.71 (s, 3H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 158.55 (s), 149.17 (s), 132.51 (s), 129.24 (s), 128.87 (s), 116.11 (s), 114.15 (s), 112.77 (s), 55.47 (s), 46.38 (s).

**N-Benzyl-4-methoxyanilne** (6g)<sup>14</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.37 (d, *J* = 7.1 Hz, 2H), 7.32 (t, *J* = 7.6 Hz, 2H), 7.22 (t, *J* = 7.3 Hz, 1H), 6.70 (d, *J* = 8.9 Hz, 2H), 6.56 (d, *J* = 8.9 Hz, 2H), 5.80 (s, 1H), 4.22 (s, 2H), 3.62 (s, 3H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 151.19 (s), 143.39 (s), 141.04 (s), 128.68 (s), 127.70 (s), 127.00 (s), 115.01 (s), 113.81 (s), 55.73 (s), 47.79 (s).

**4-N-[(4-Fluorophenyl)methyl]-2-nitrobenzene-1,4-diamine** (6h), <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.40 (t, *J* = 8.0, 5.8 Hz, 2H), 7.15 (t, *J* = 8.7 Hz, 2H), 7.07 – 6.96 (m, 3H), 6.93 (d, *J* = 2.3 Hz, 1H), 6.88 (d, *J* = 9.0 Hz, 1H), 6.04 (s, 1H), 4.20 (d, *J* = 5.7 Hz, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 140.24 (s), 139.42 (s), 130.48 (s), 129.63 (d, *J* = 8.1 Hz), 127.21 (s), 120.75 (s), 115.56 (s), 115.39 (s), 102.84 (s), 46.83 (s); <sup>19</sup>F NMR (471 MHz, DMSO) δ -116.34 (s).

**N-Benzyl-2-nitroaniline** (6i)<sup>18</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 8.66 (s, 1H), 8.09 (d, 1H), 7.44 (t, *J* = 7.7 Hz, 1H), 7.36 (dt, *J* = 15.0, 7.5 Hz, 4H), 7.27 (t, *J* = 7.0 Hz, 1H), 6.92 (d, *J* = 8.7 Hz, 1H), 6.68 (t, 1H), 4.64 (d, *J* = 6.1 Hz, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 145.39 (s), 138.99 (s), 136.87 (s), 131.79 (s), 129.06 (s), 127.48 (d, *J* = 16.3 Hz), 126.69 (s), 115.90 (s), 115.39 (s), 46.18 (s).

**N-Benzylanthranilic Acid** (6j)<sup>19</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.80 (d, *J* = 7.9 Hz, 1H), 7.31 (M, 6H), 6.67 (d, *J* = 8.5 Hz, 1H), 6.56 (S, 1H), 4.46 (s, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 170.46 (s), 151.13 (s), 139.84 (s), 134.84 (s), 132.15 (s), 129.00 (s), 127.45 (d, *J* = 15.8 Hz), 114.97 (s), 112.15 (s), 110.75 (s), 46.31 (s).

**1H-Indole** (8)<sup>20</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 11.06 (s, 1H), 7.53 (d, *J* = 7.8 Hz, 1H), 7.39 (dd, *J* = 8.1, 0.7 Hz, 1H), 7.32 (t, *J* = 2.7 Hz, 1H), 7.10 – 6.94 (m, 2H), 6.43 – 6.37 (m, 1H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 136.32 (s), 128.08 (s), 125.64 (s), 121.33 (s), 120.46 (s), 119.21 (s), 111.84 (s), 101.43 (s).

**Indoline** (9)<sup>20</sup>, <sup>1</sup>H NMR (500 MHz, DMSO) δ 7.00 (d, *J* = 7.2 Hz, 1H), 6.88 (t, 1H), 6.53 – 6.47 (m, 2H), 5.40 (s, 1H), 3.38 (t, *J* = 8.4 Hz, 2H), 2.87 (t, *J* = 8.5 Hz, 2H); <sup>13</sup>C NMR (126 MHz, DMSO) δ 153.01 (s), 129.14 (s), 127.34 (s), 124.63 (s), 117.27 (s), 108.82 (s), 46.89 (s), 29.72 (s).

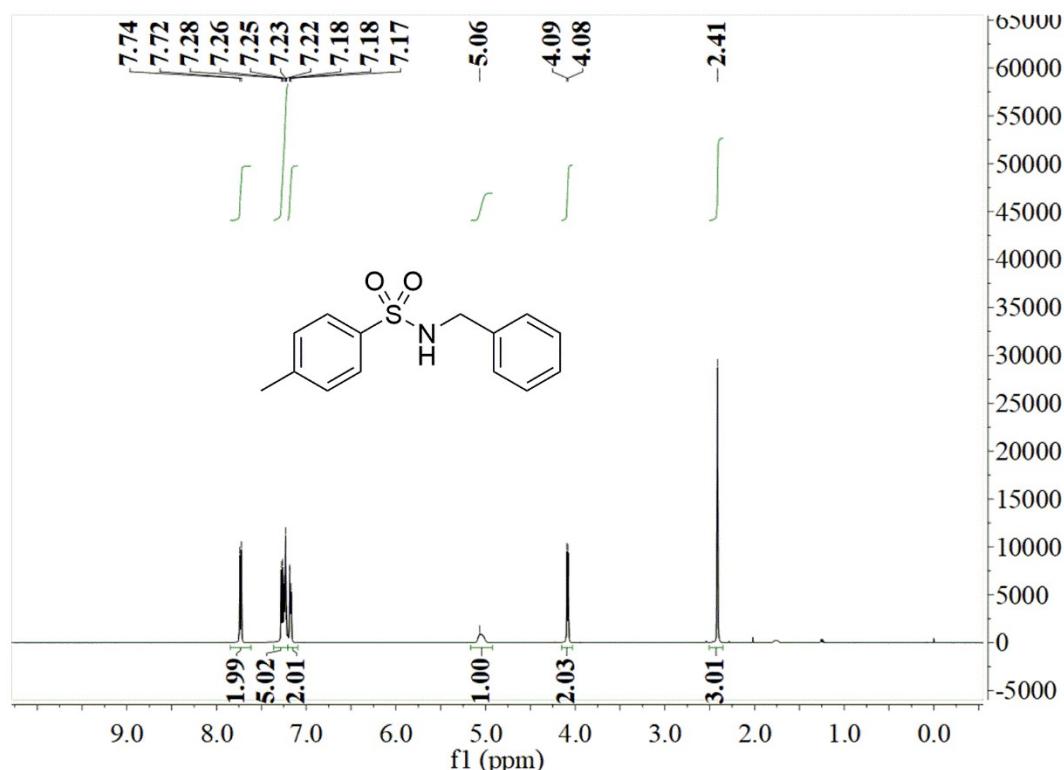


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-4-methylbenzenesulfonamide (3a).

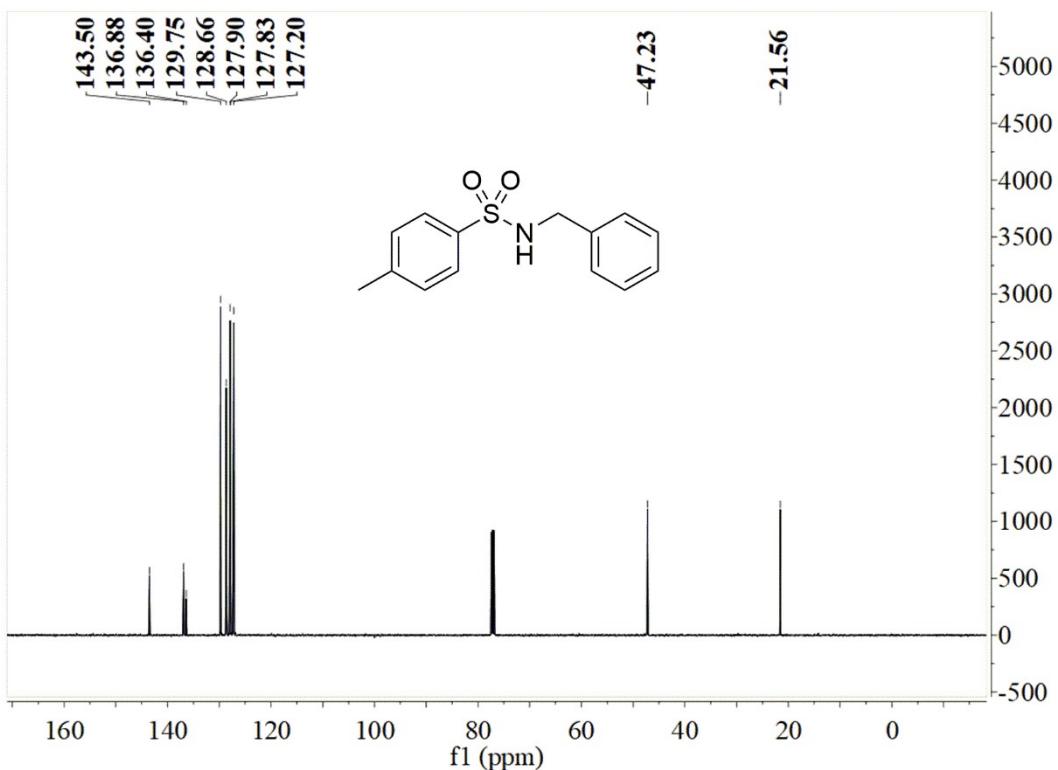


Figure  $^{13}\text{C}$  NMR spectrum of *N*-benzyl-4-methylbenzenesulfonamide (3a).

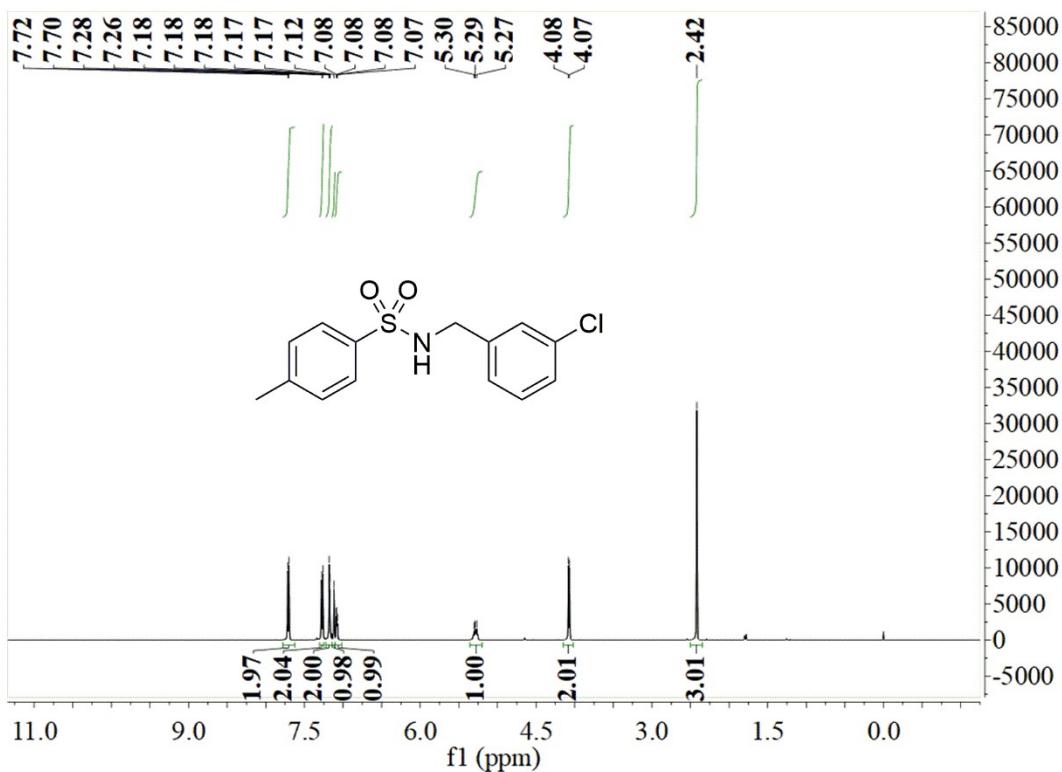


Figure  $^1\text{H}$  NMR spectrum of *N*-(3-chlorobenzyl)-4-methylbenzenesulfonamide (3b).

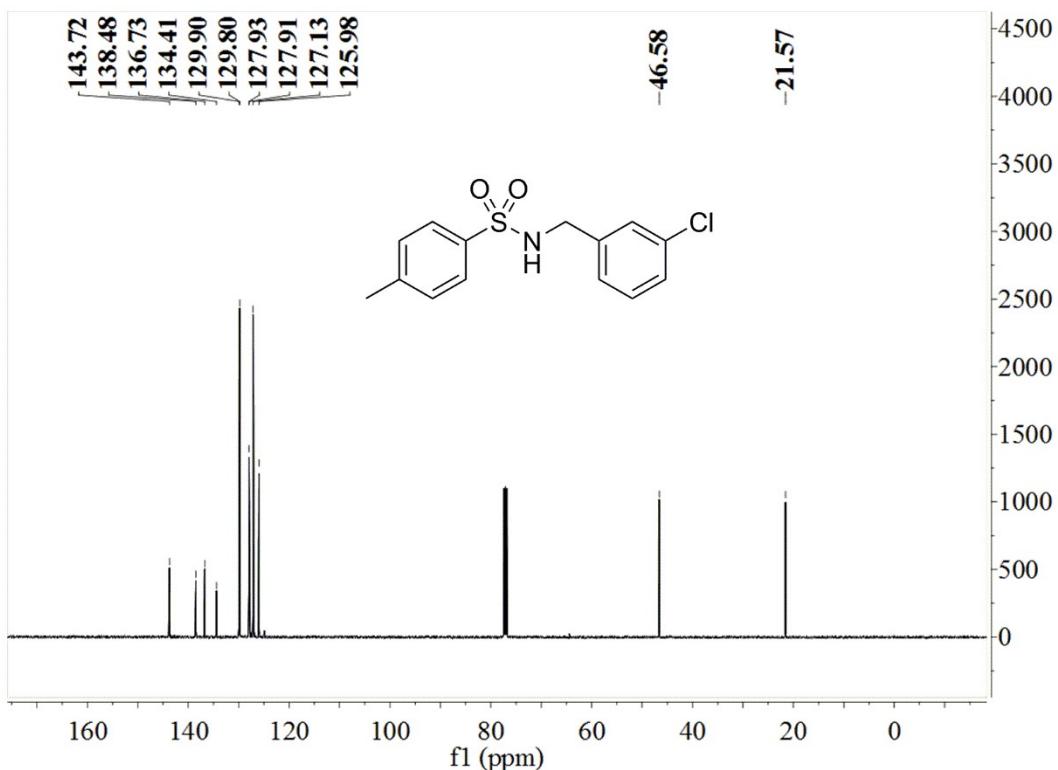


Figure  $^{13}\text{C}$  NMR spectrum of  $N$ -(3-chlorobenzyl)-4-methylbenzenesulfonamide (3b).

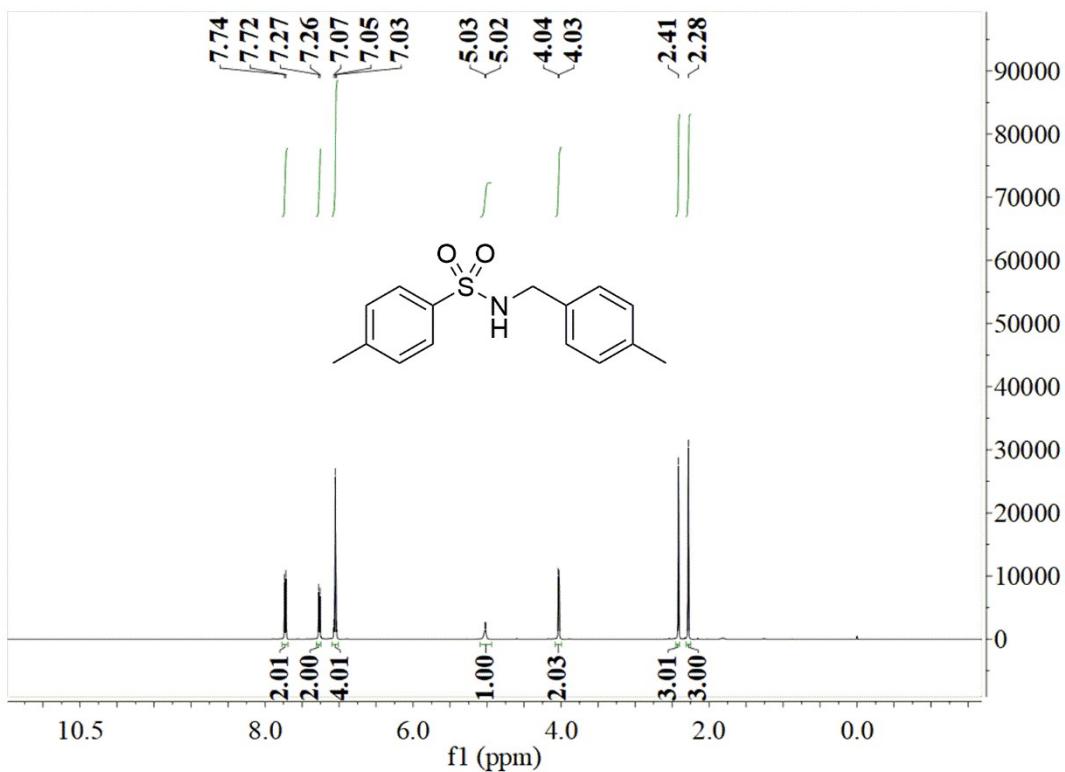


Figure  $^1\text{H}$  NMR spectrum of 4-methyl- $N$ -(4-methylbenzyl)benzenesulfonamide (3c).

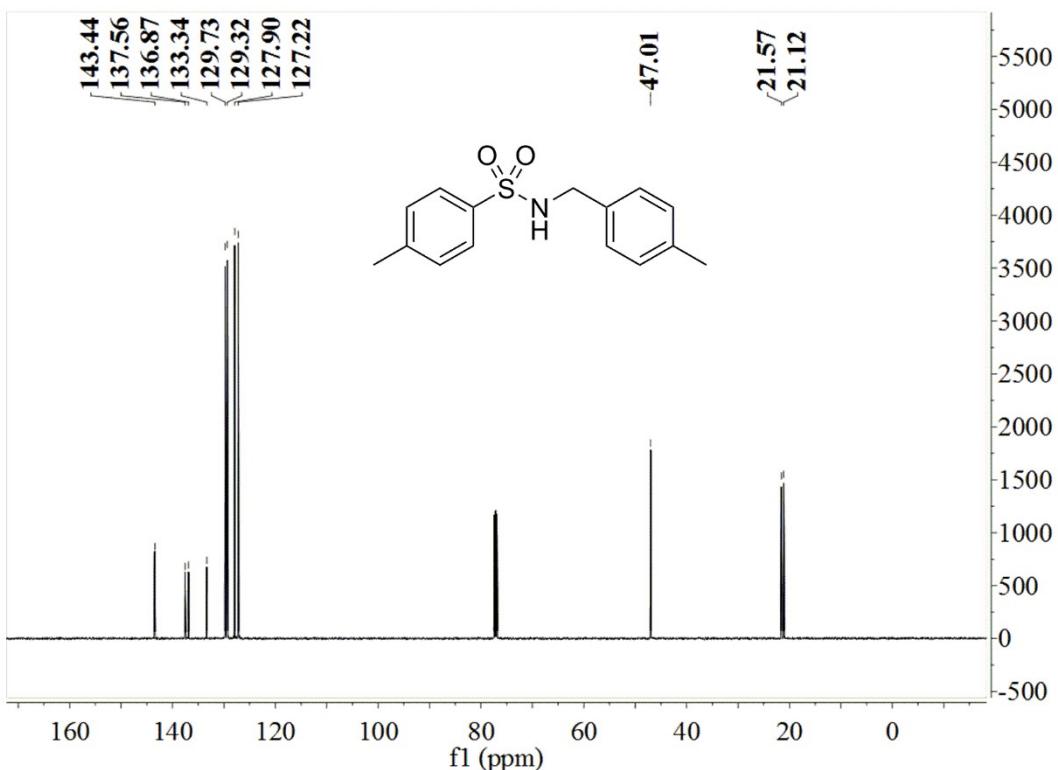


Figure  $^{13}\text{C}$  NMR spectrum of 4-methyl- $N$ -(4-methylbenzyl)benzenesulfonamide (3c).

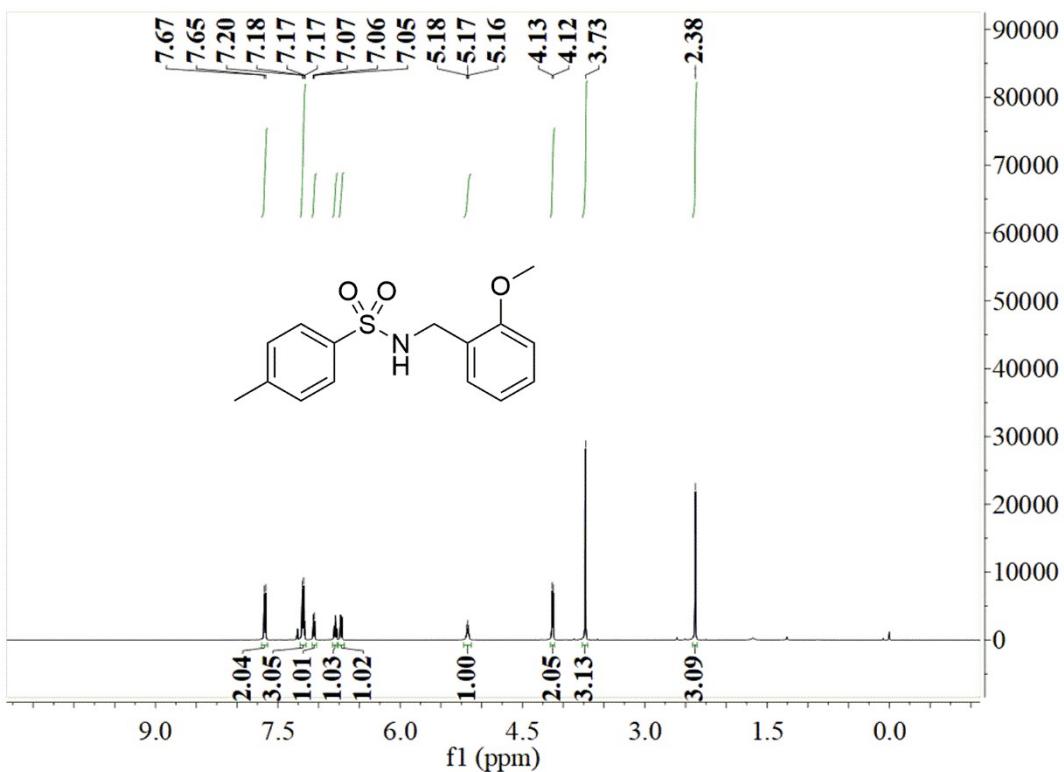


Figure  $^1\text{H}$  NMR spectrum of  $N$ -(2-methoxybenzyl)-4-methylbenzenesulfonamide (3d).

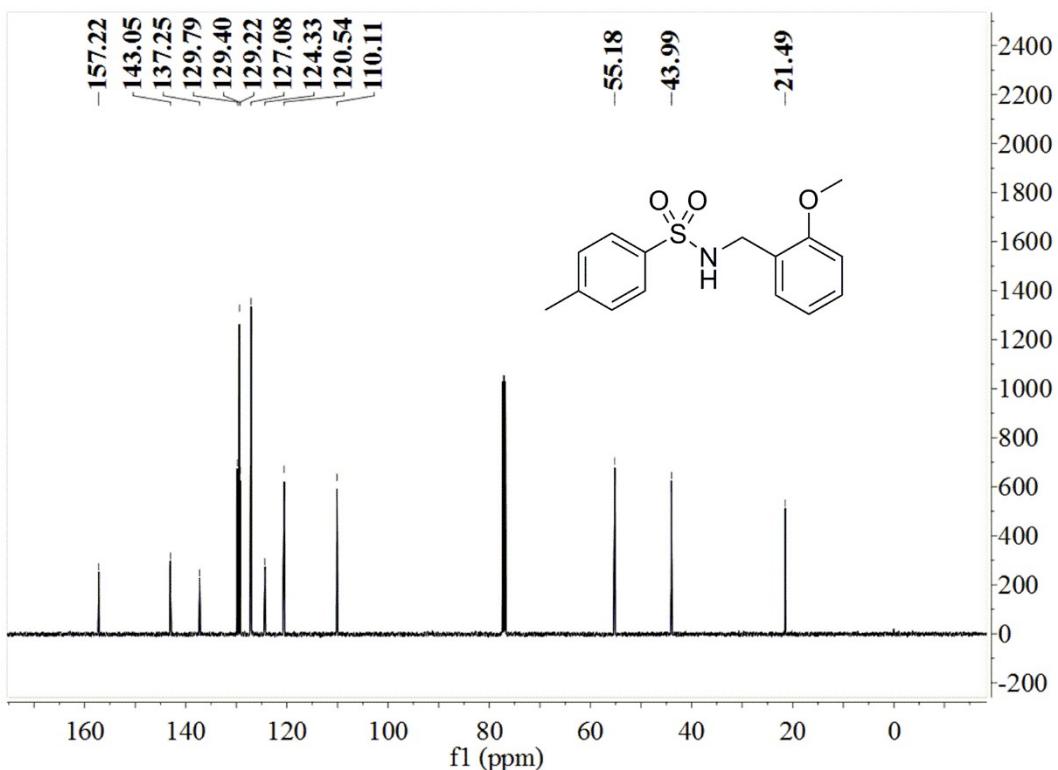


Figure <sup>13</sup>C NMR spectrum of *N*-(2-methoxybenzyl)-4-methylbenzenesulfonamide (3d).

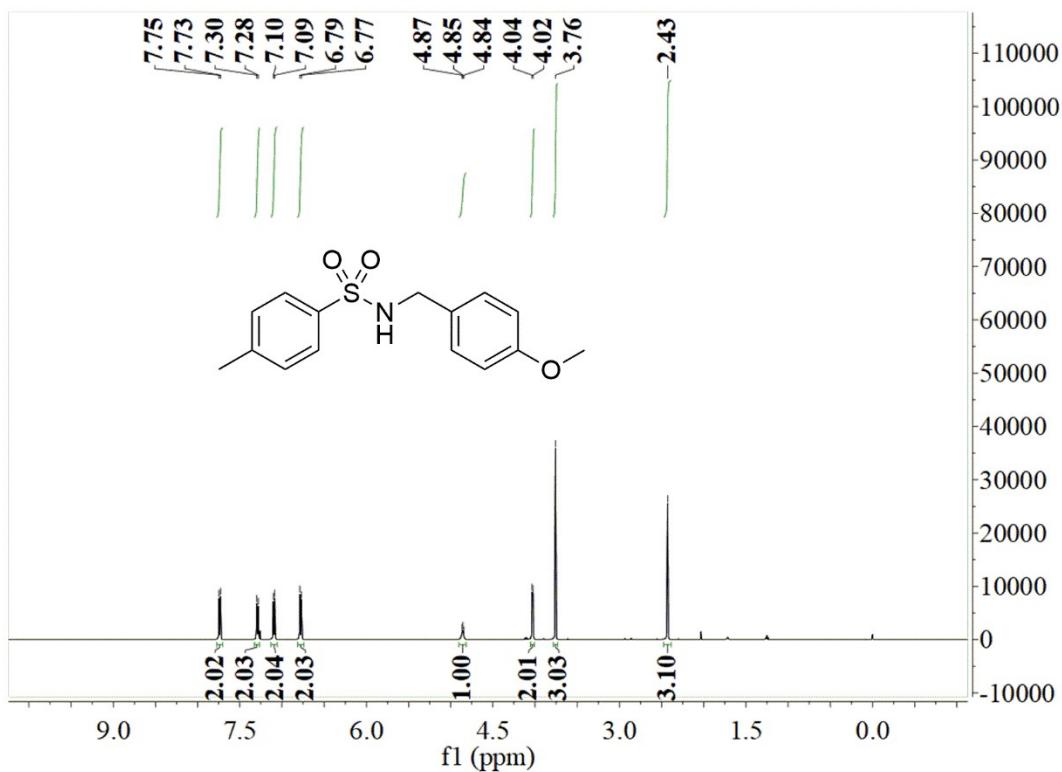


Figure <sup>1</sup>H NMR spectrum of *N*-(4-methoxybenzyl)-4-methylbenzenesulfonamide (3e).

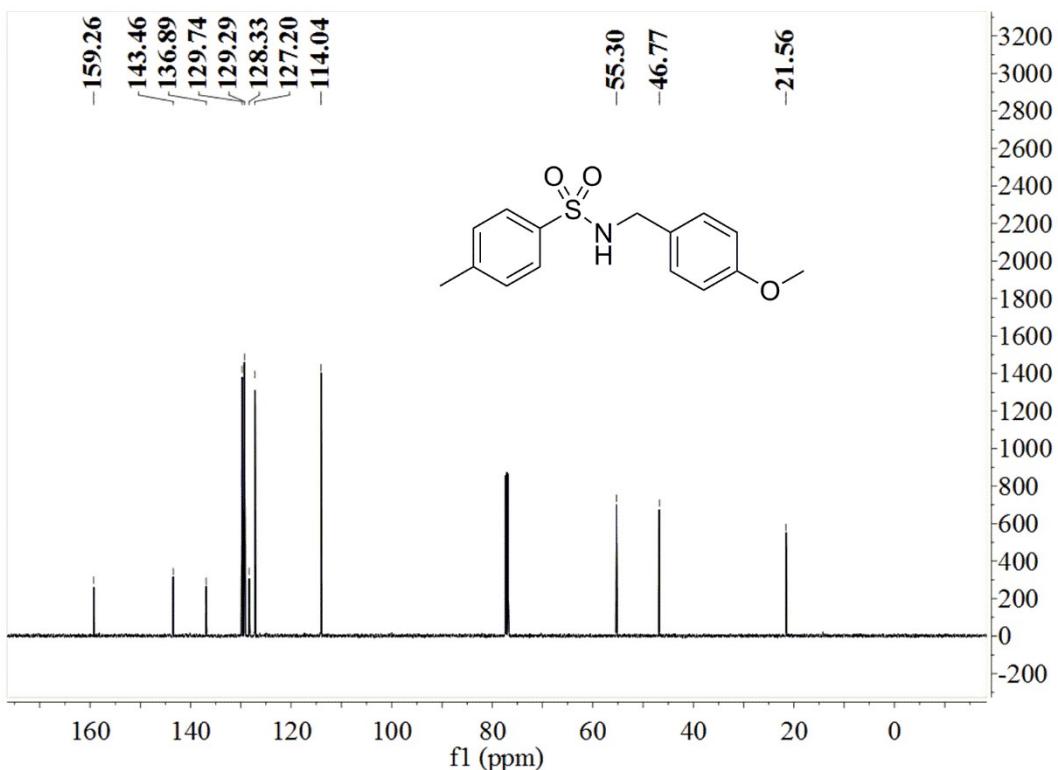


Figure  $^{13}\text{C}$  NMR spectrum of  $N$ -(4-methoxybenzyl)-4-methylbenzenesulfonamide ( $3\text{e}$ ).

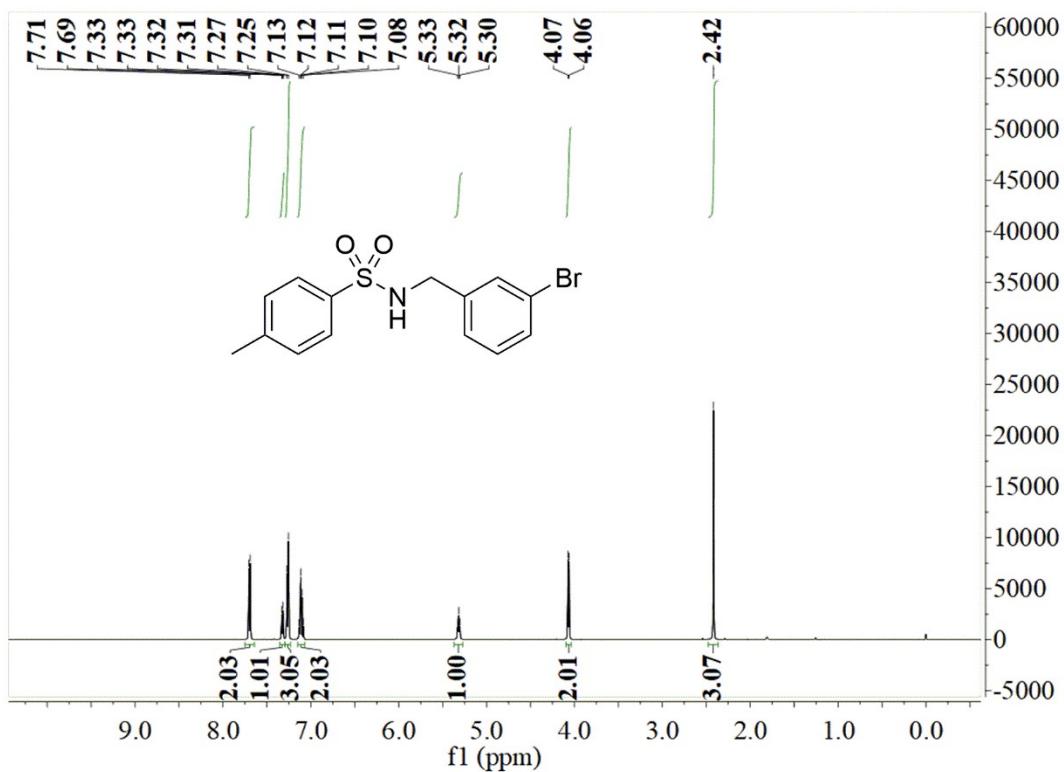


Figure  $^1\text{H}$  NMR spectrum of  $N$ -(3-bromobenzyl)-4-methylbenzenesulfonamide ( $3\text{f}$ ).

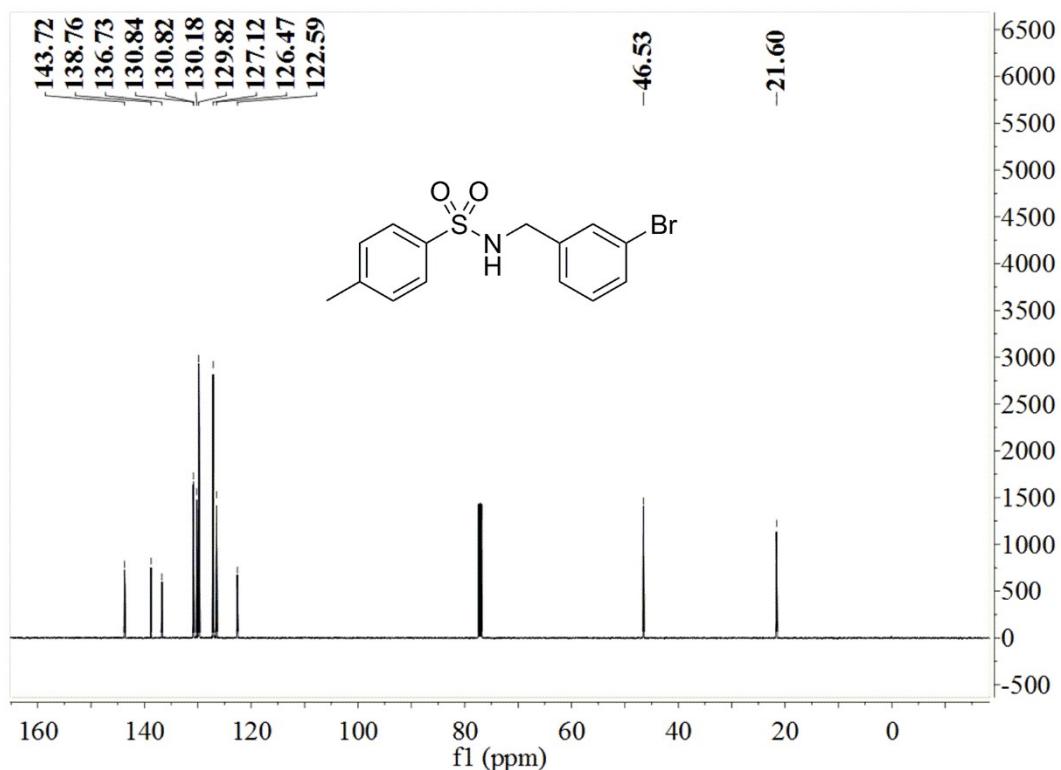


Figure <sup>13</sup>C NMR spectrum of *N*-(3-bromobenzyl)-4-methylbenzenesulfonamide (3f).

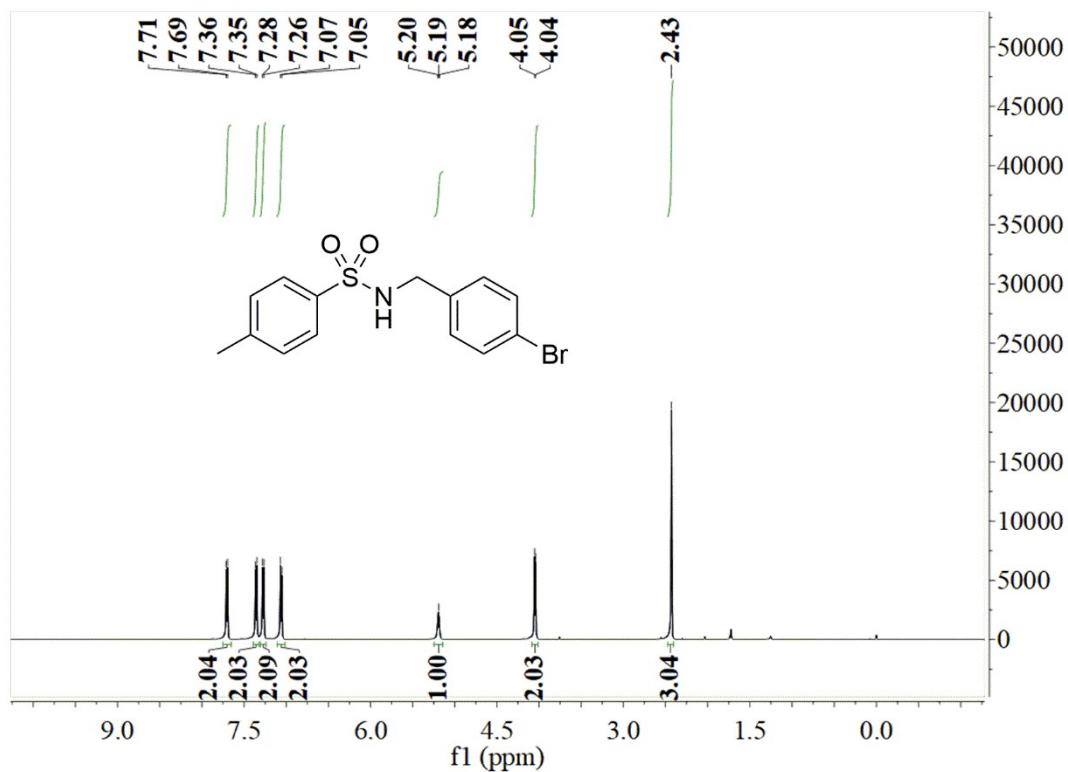


Figure <sup>1</sup>H NMR spectrum of *N*-(4-bromobenzyl)-4-methylbenzenesulfonamide (3g).

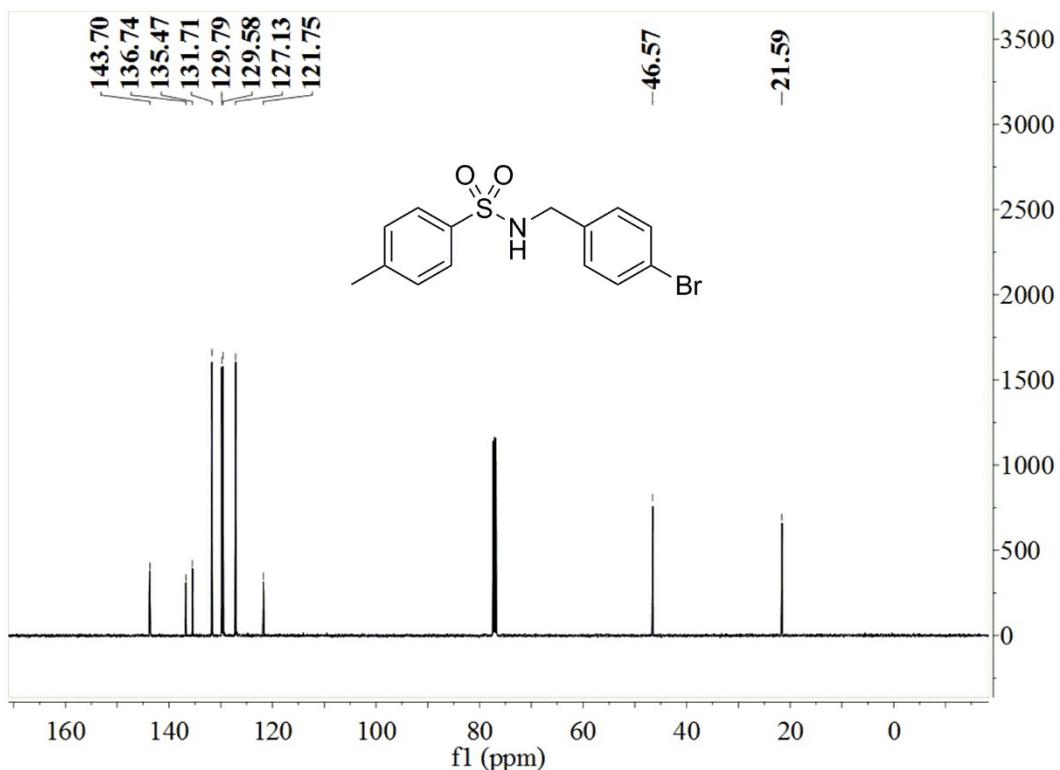


Figure  $^{13}\text{C}$  NMR spectrum of *N*-(4-bromobenzyl)-4-methylbenzenesulfonamide (3g).

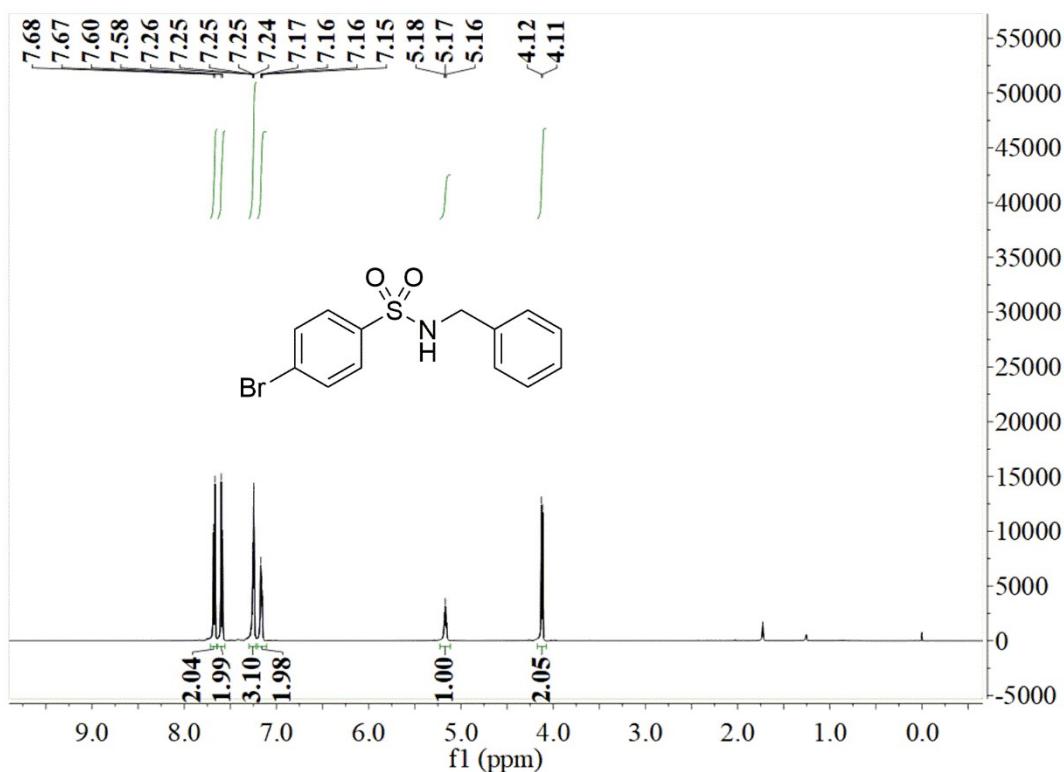


Figure  $^1\text{H}$  NMR spectrum of *N*-benzyl-4-bromobenzenesulfonamide (3h).

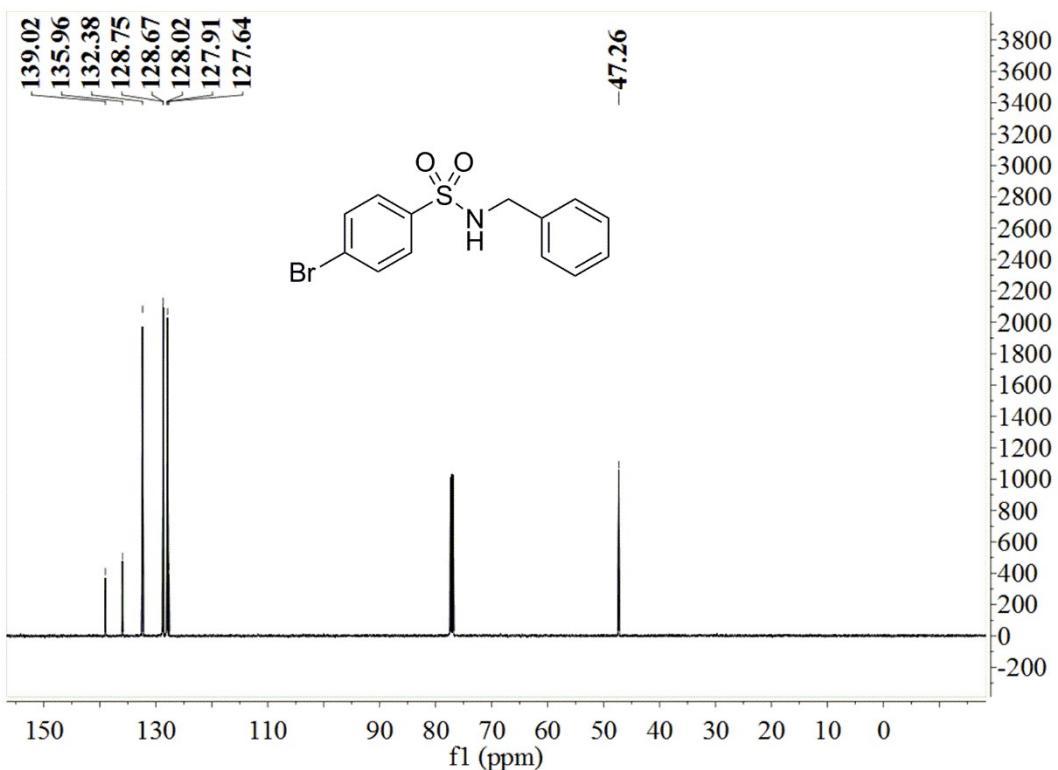


Figure  $^{13}\text{C}$  NMR spectrum of *N*-benzyl-4-bromobenzenesulfonamide (3h).

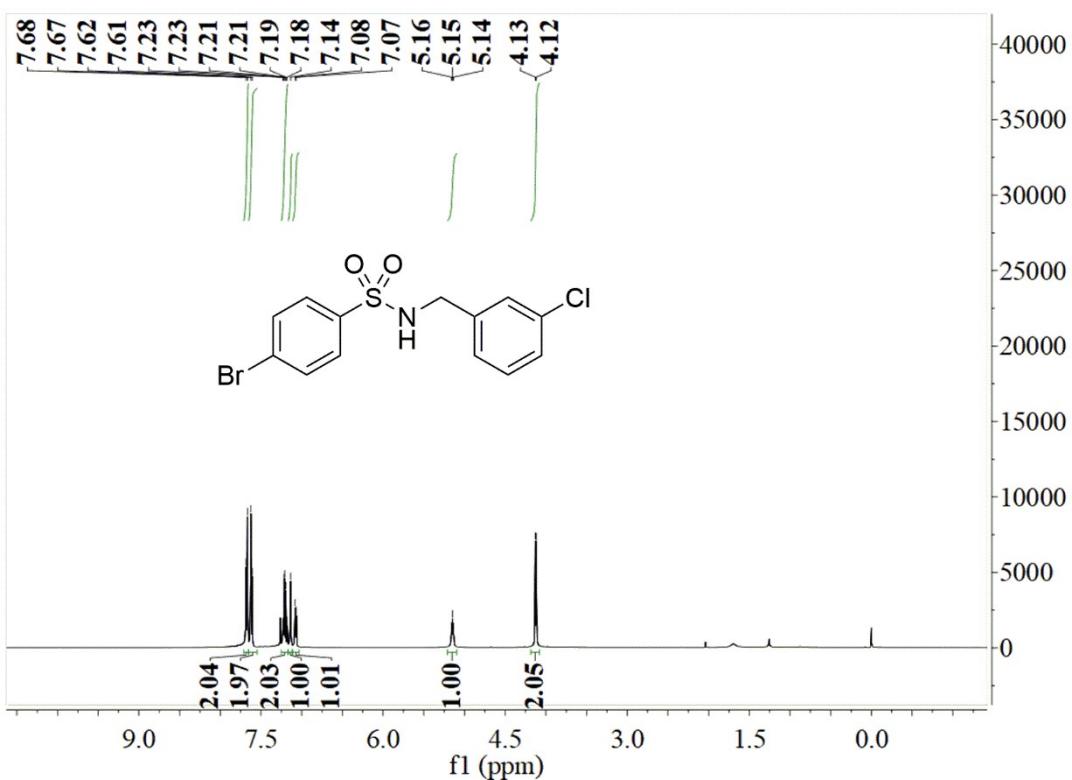


Figure  $^1\text{H}$  NMR spectrum of 4-bromo-*N*-(3-chlorobenzyl)benzenesulfonamide (3i).

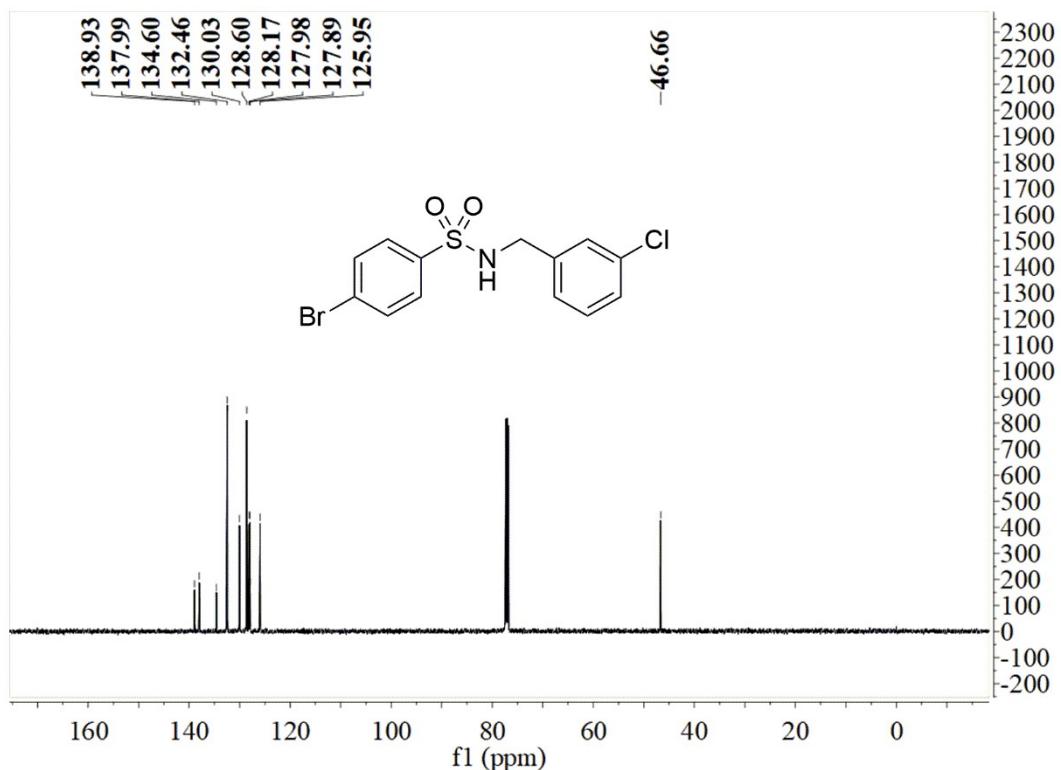


Figure  $^{13}\text{C}$  NMR spectrum of 4-bromo- $N$ -(3-chlorobenzyl)benzenesulfonamide (3i).

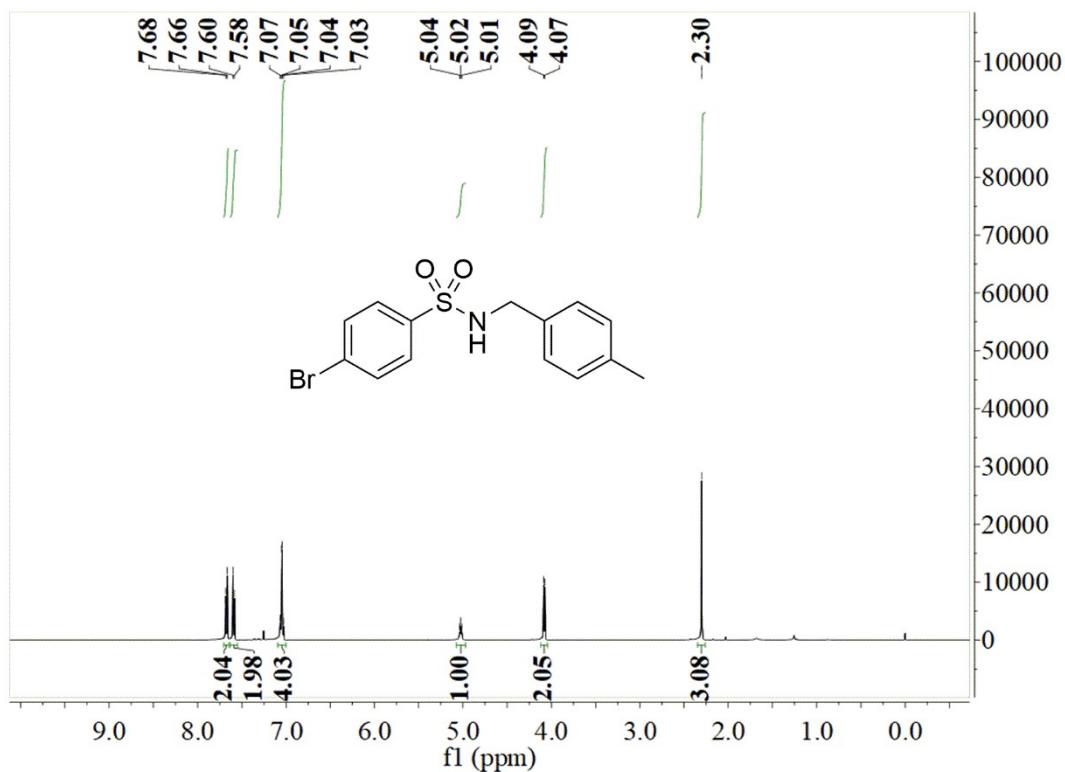


Figure  $^1\text{H}$  NMR spectrum of 4-bromo- $N$ -(4-methylbenzyl)benzenesulfonamide (3j).

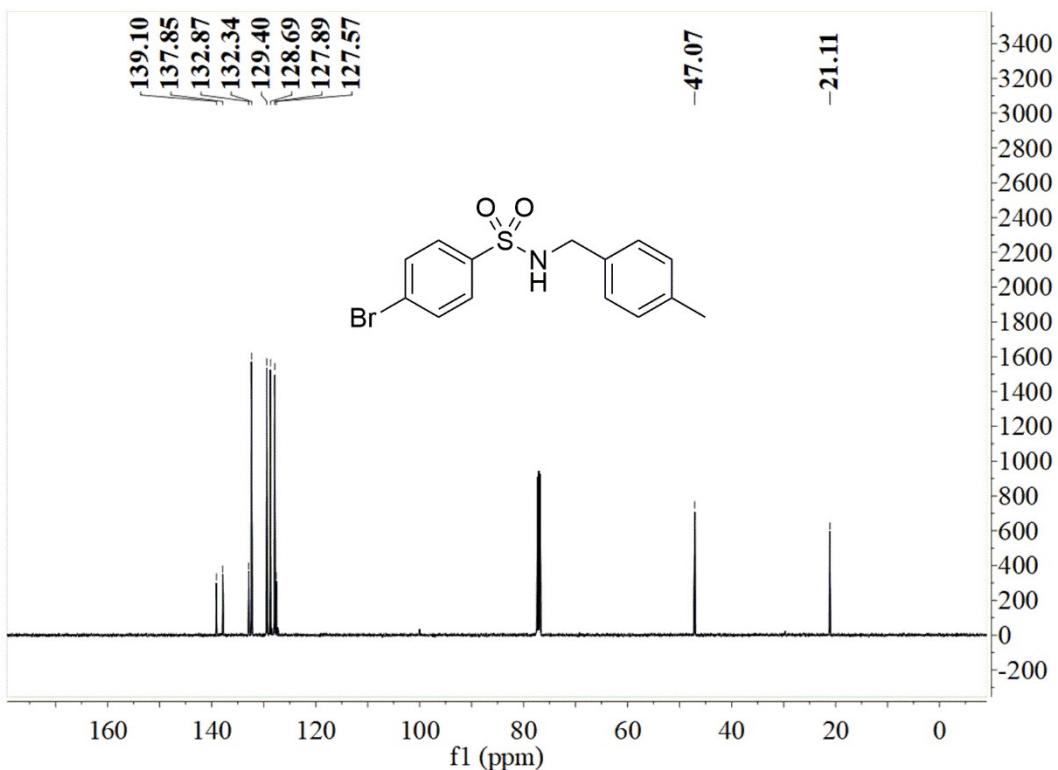


Figure <sup>13</sup>C NMR spectrum of 4-bromo-*N*-(4-methylbenzyl)benzenesulfonamide (3j).

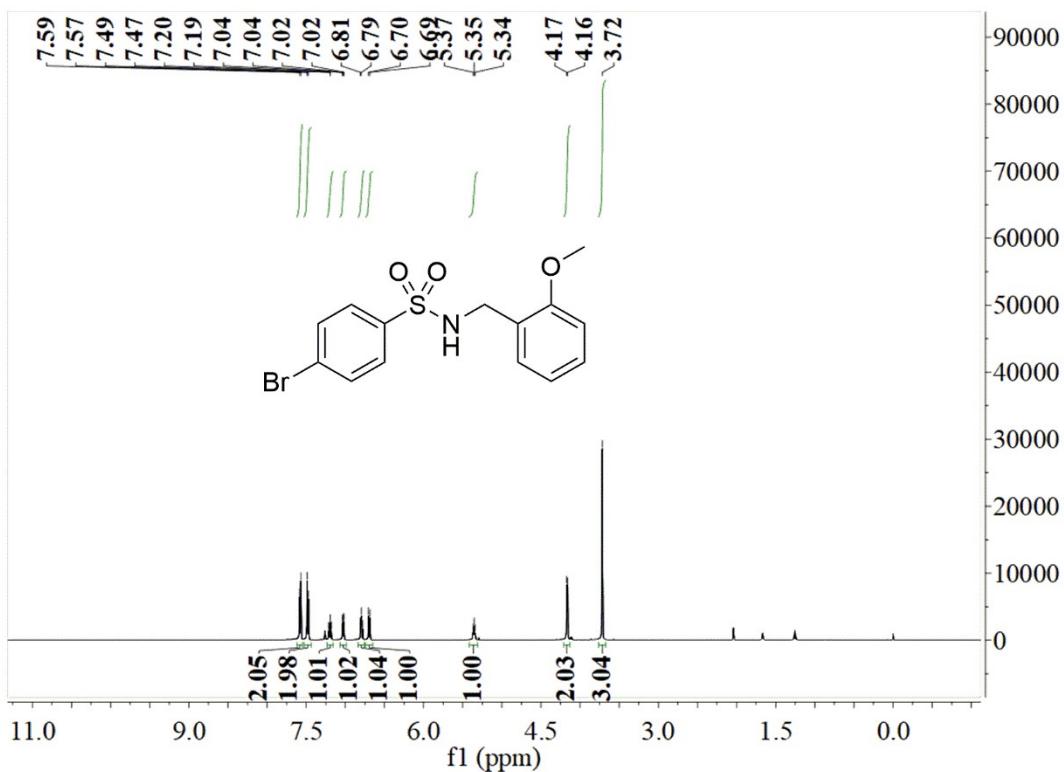


Figure <sup>1</sup>H NMR spectrum of 4-bromo-*N*-(2-methoxybenzyl)benzenesulfonamide (3k).

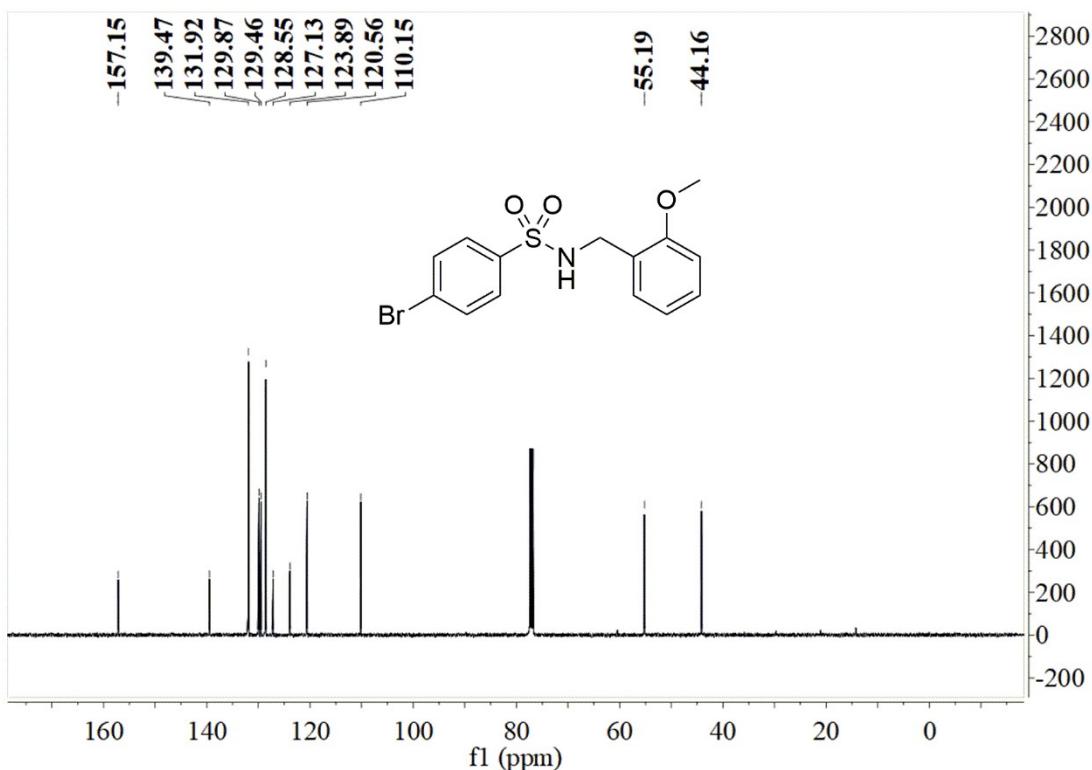


Figure  $^{13}\text{C}$  NMR spectrum of 4-bromo- $N$ -(2-methoxybenzyl)benzenesulfonamide (3k).

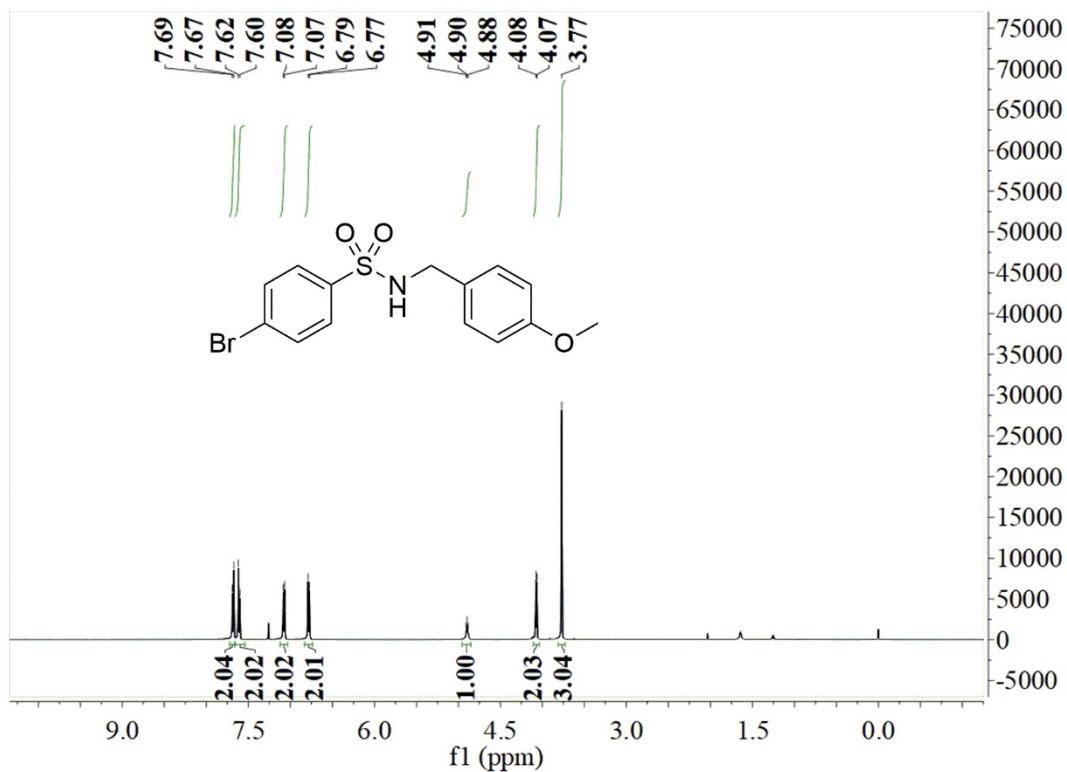


Figure  $^1\text{H}$  NMR spectrum of 4-bromo- $N$ -(4-methoxybenzyl)benzenesulfonamide (3l).

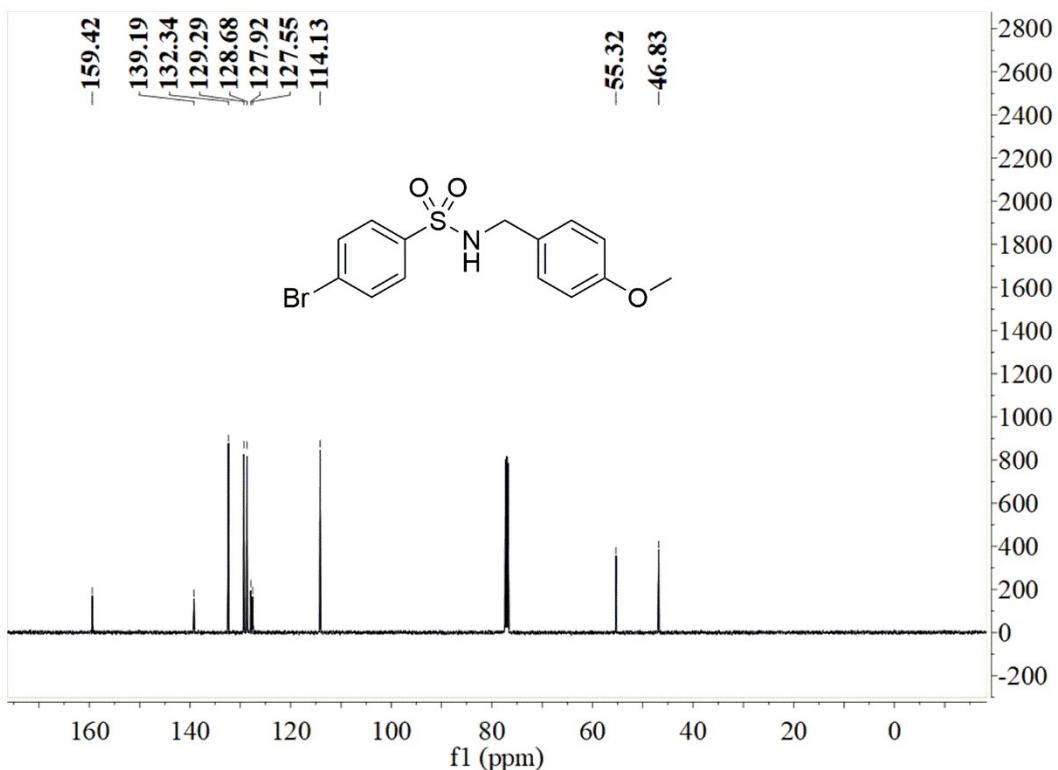


Figure <sup>13</sup>C NMR spectrum of 4-bromo-N-(4-methoxybenzyl)benzenesulfonamide (3l).

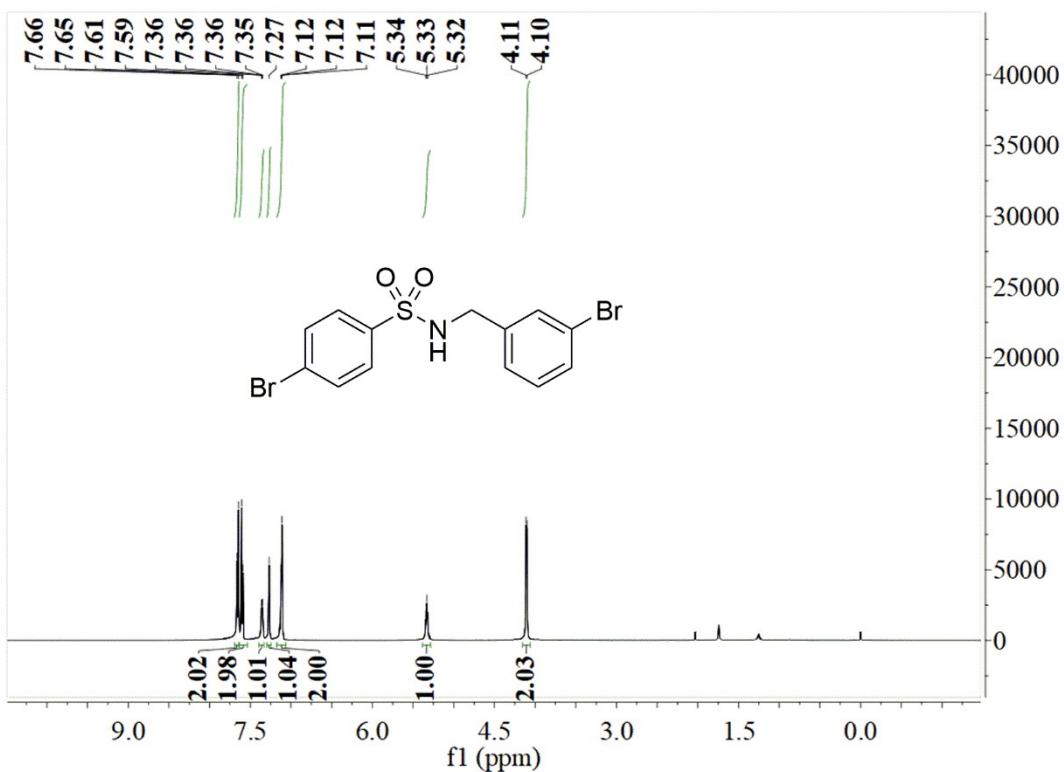


Figure <sup>1</sup>H NMR spectrum of 4-bromo-N-(3-bromobenzyl)benzenesulfonamide (3m).

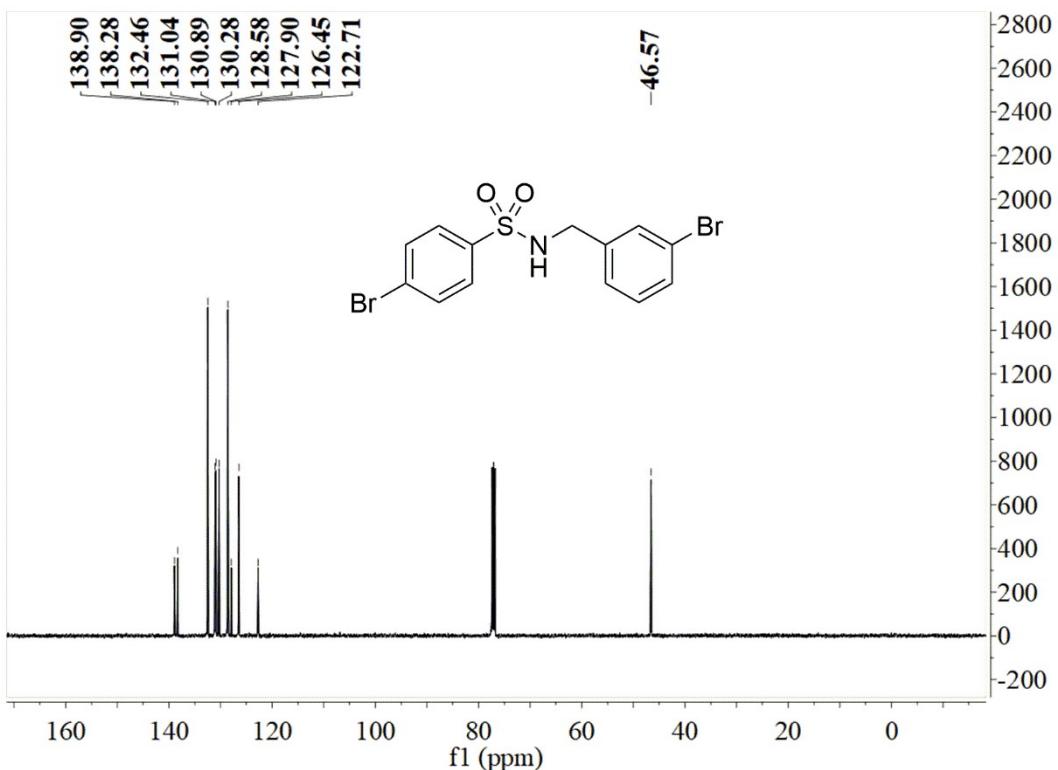


Figure <sup>13</sup>C NMR spectrum of 4-bromo-N-(3-bromobenzyl)benzenesulfonamide (3m).

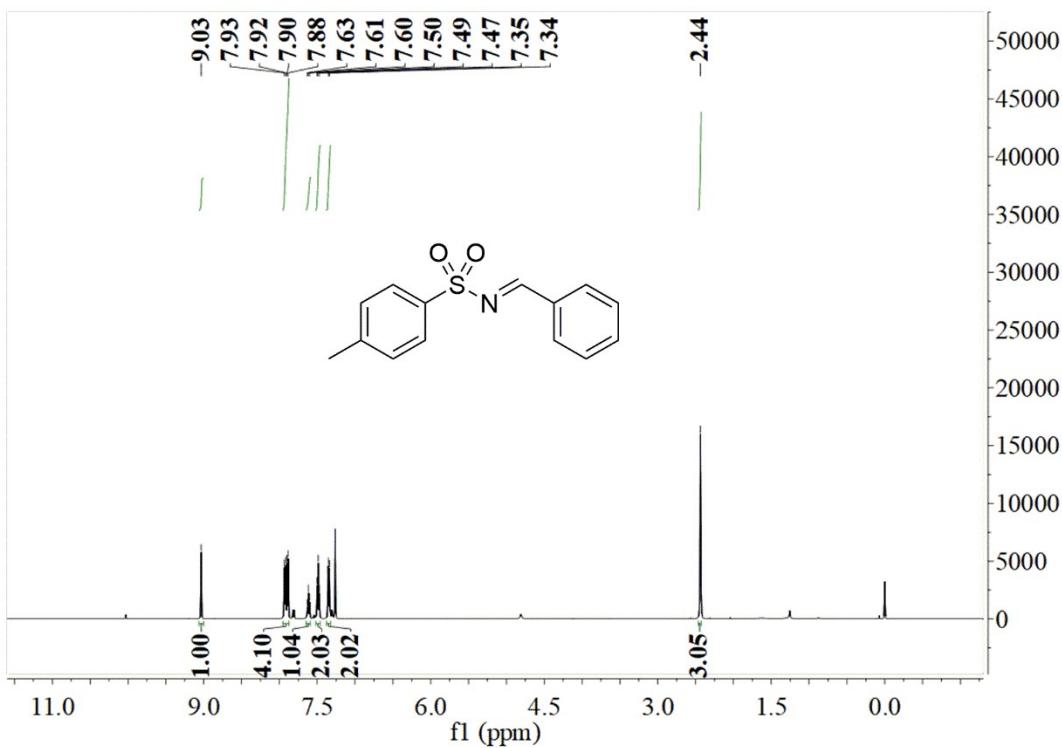


Figure <sup>1</sup>H NMR spectrum of *N*-benzylidene-*p*-toluenesulfonamide (product of **Scheme 4b**).

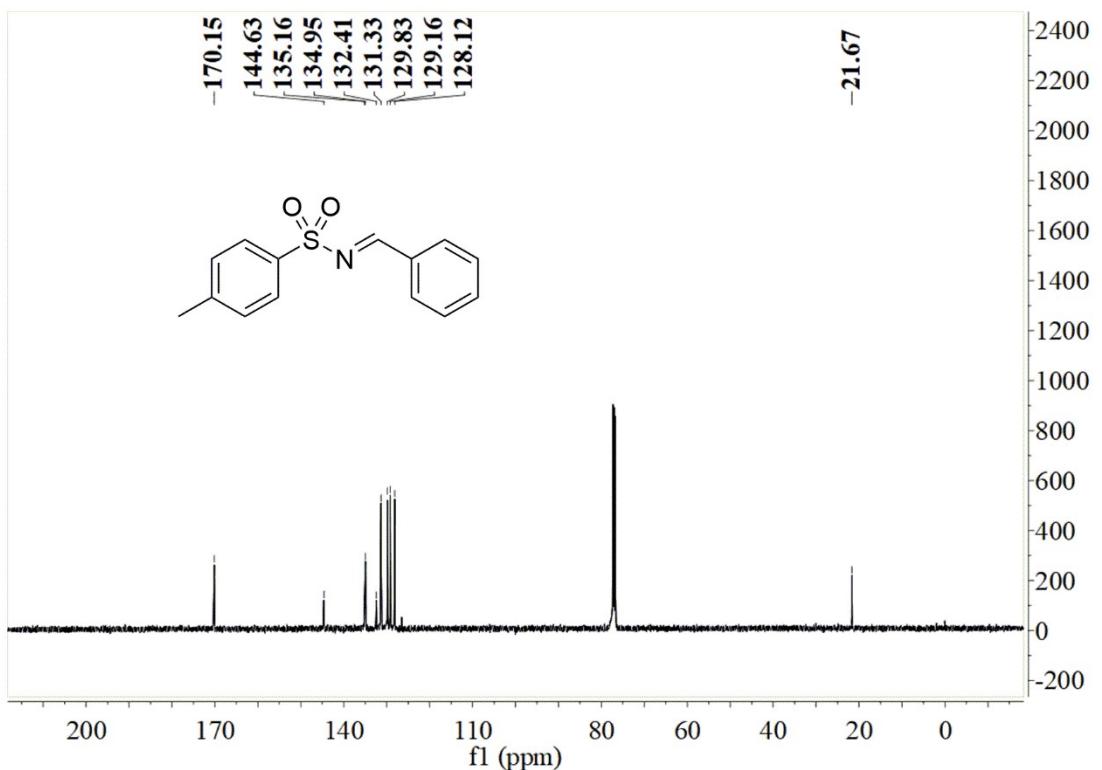


Figure  $^{13}\text{C}$  NMR spectrum of *N*-benzylidene-*p*-toluenesulfonamide (product of **Scheme 4b**).

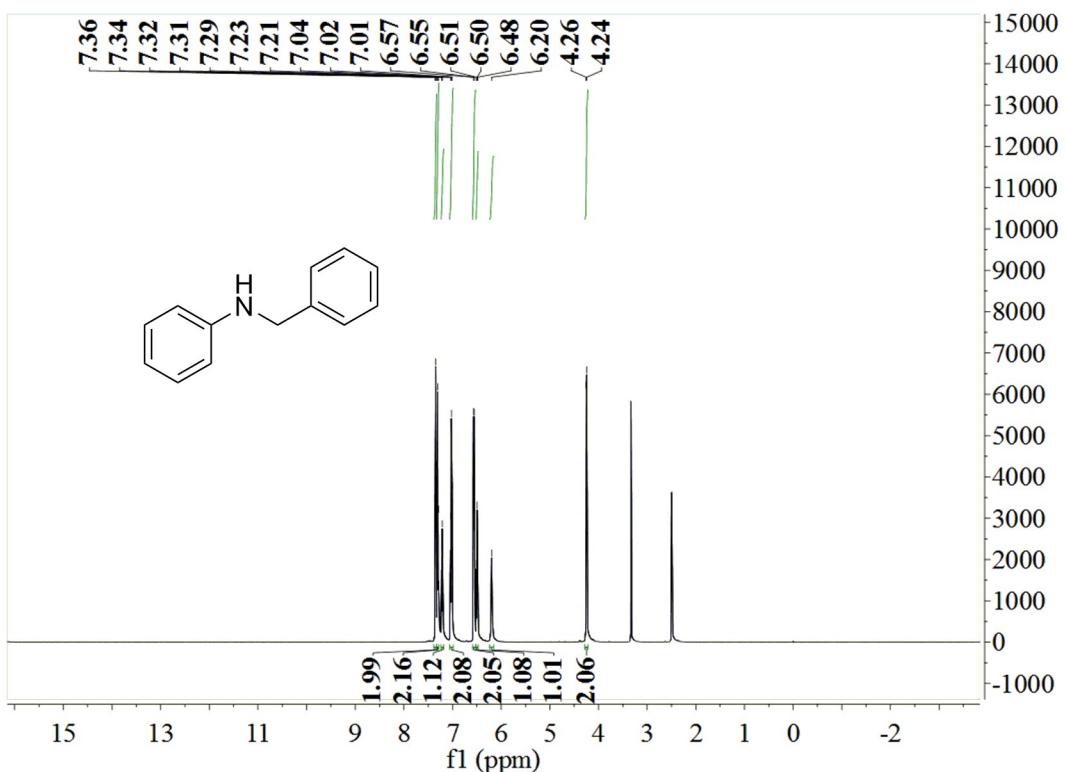


Figure <sup>1</sup>H NMR spectrum of *N*-benzyylaniline (6a).

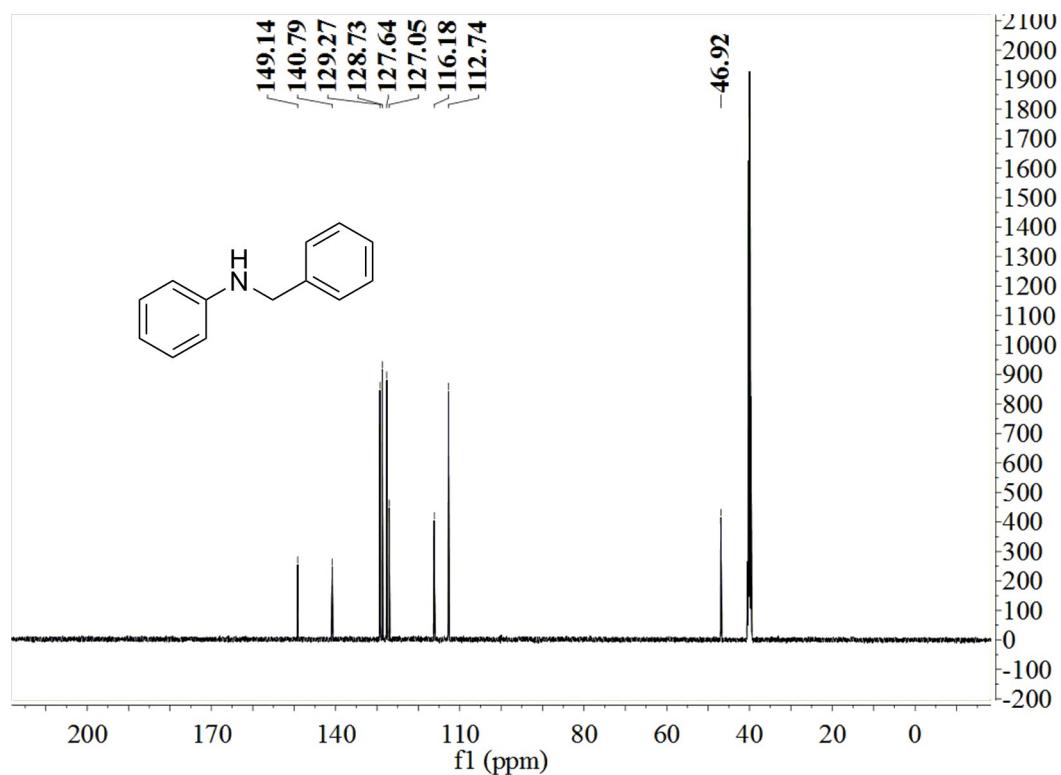


Figure <sup>13</sup>C NMR spectrum of *N*-benzyylaniline (6a).

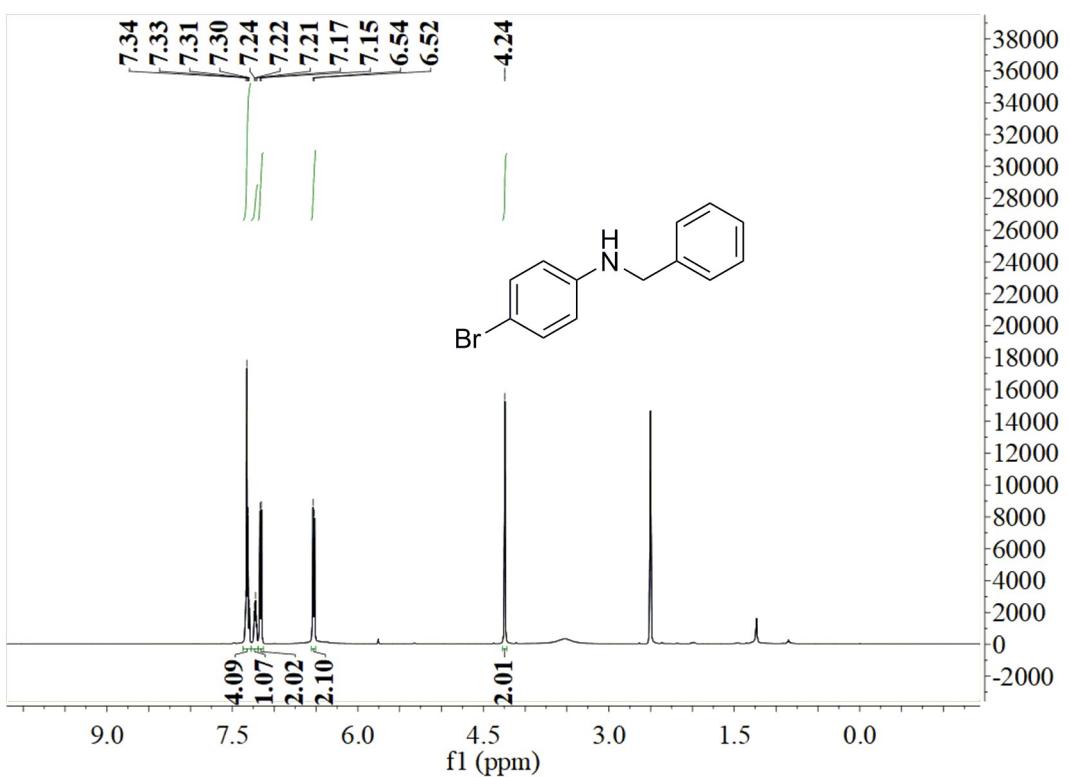


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-4-bromoaniline (6b).

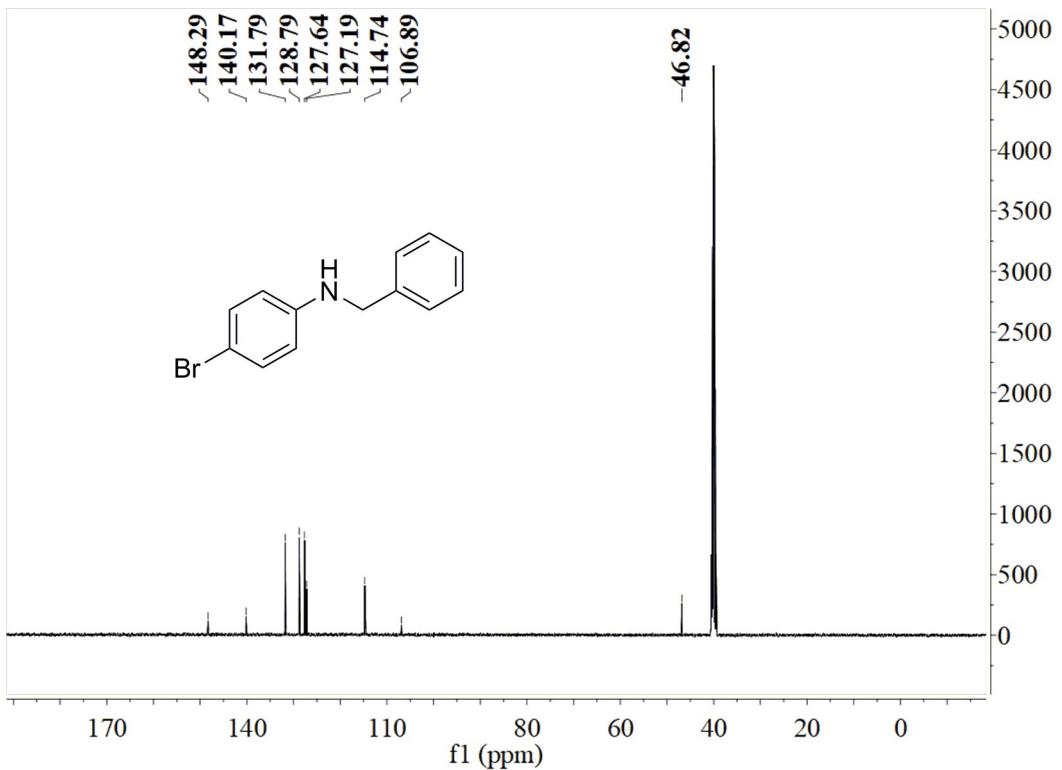


Figure <sup>13</sup>C NMR spectrum of *N*-benzyl-4-bromoaniline (6b).

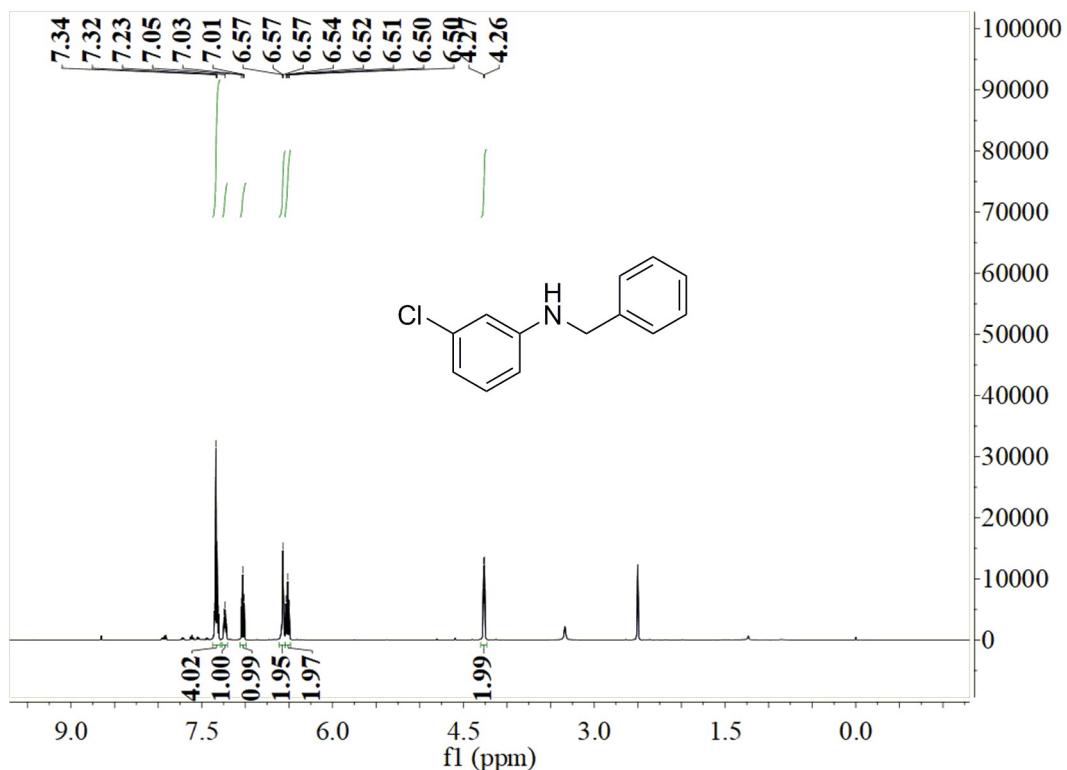


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-3-chloroaniline (6c).

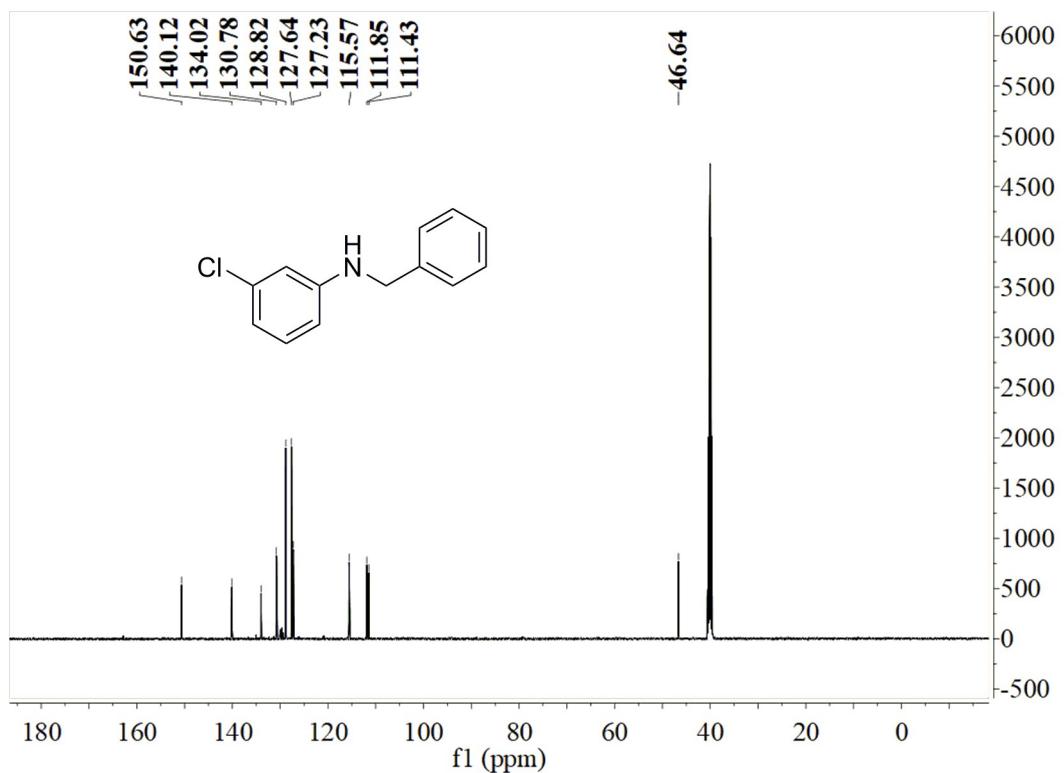


Figure <sup>13</sup>C NMR spectrum of *N*-benzyl-3-chloroaniline (6c).

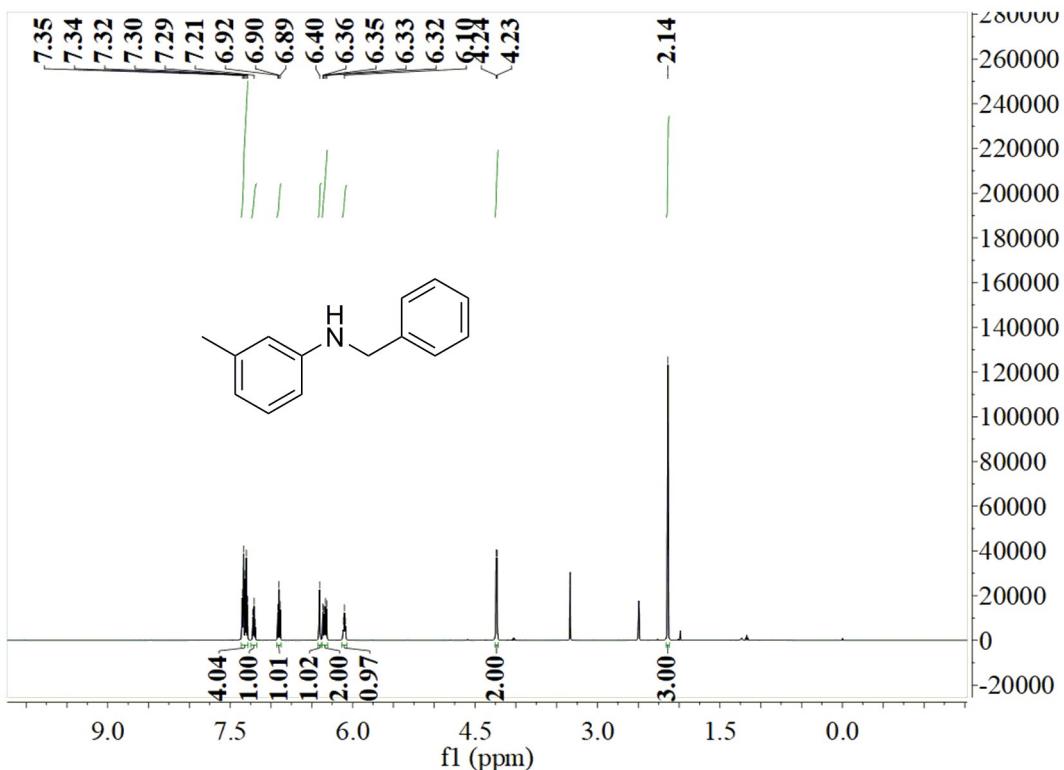


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-3-methylaniline (6d).

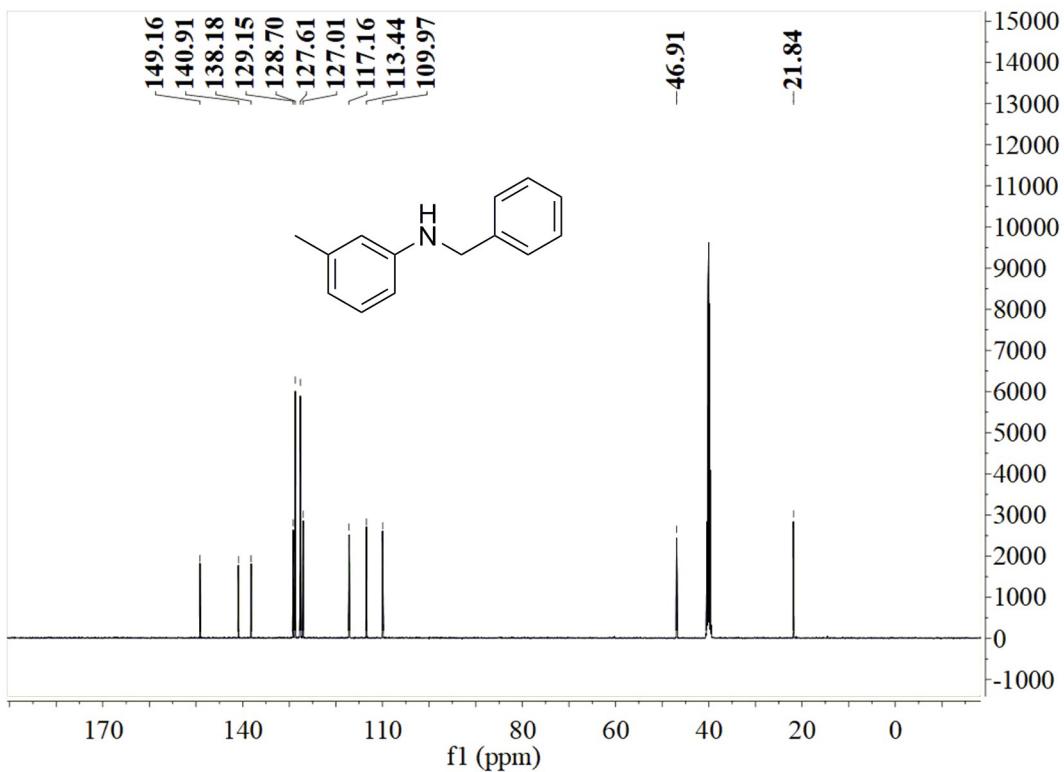


Figure <sup>13</sup>C NMR spectrum of *N*-benzyl-3-methylaniline (6d).

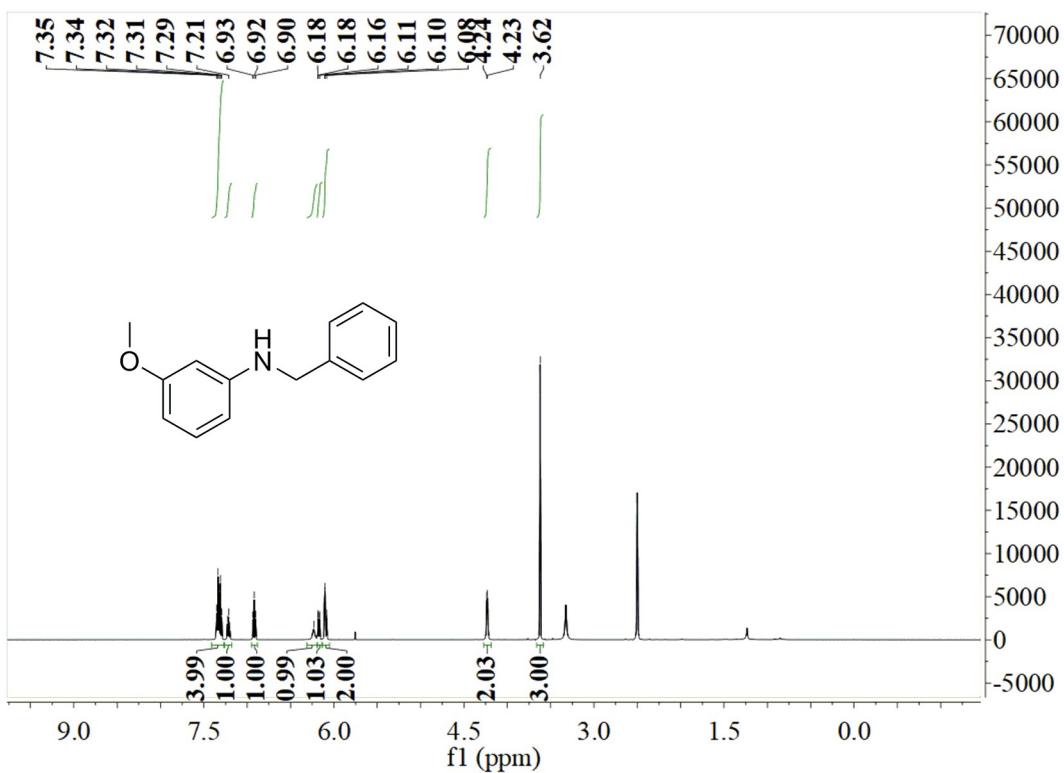


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-3-methoxyaniline (6e).

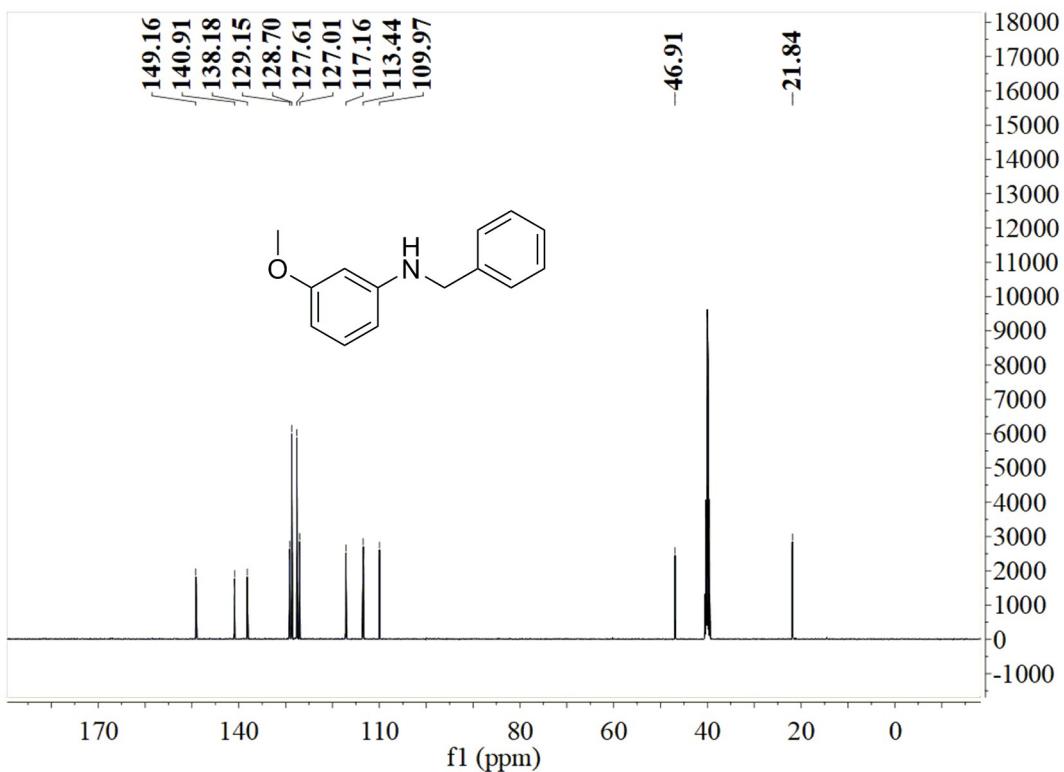


Figure <sup>13</sup>C NMR spectrum of *N*-benzyl-3-methoxyaniline (6e).

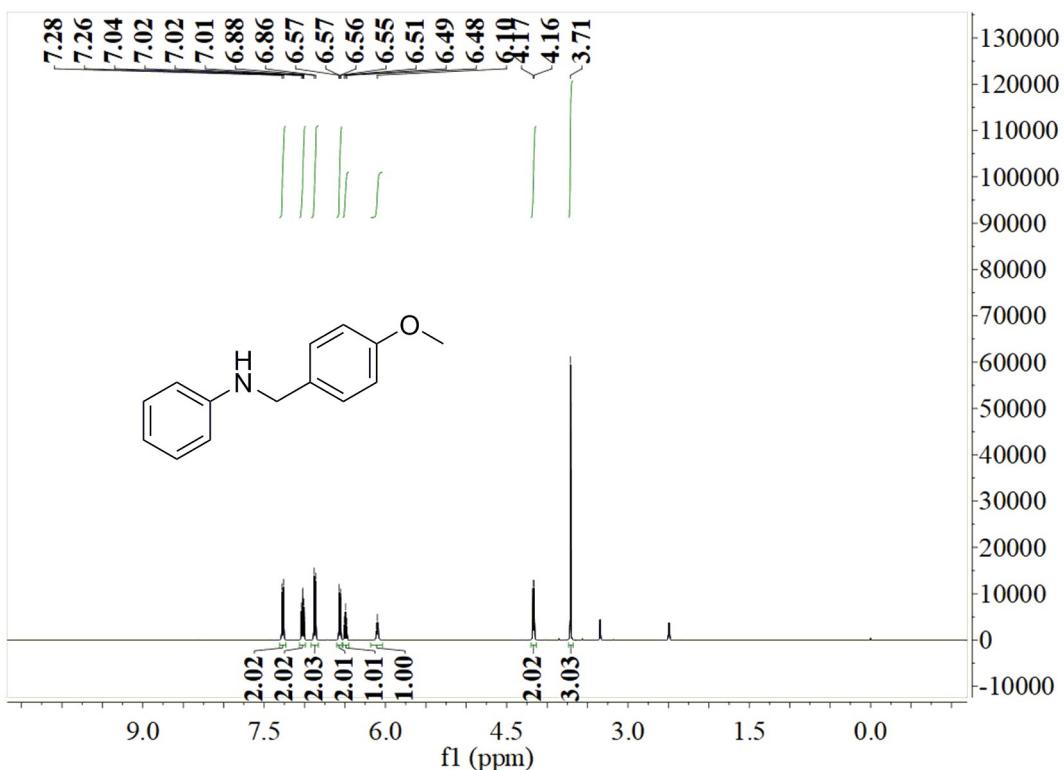


Figure <sup>1</sup>H NMR spectrum of *N*-phenyl-4-methoxybenzylamine (6f).

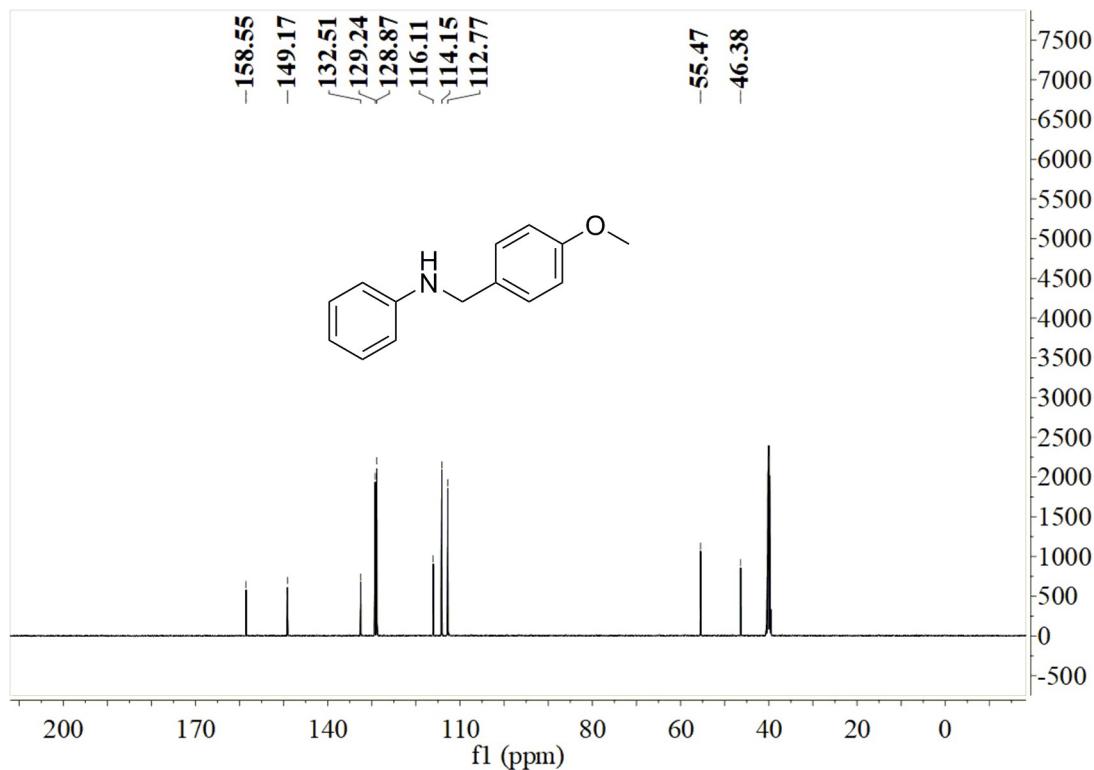


Figure <sup>13</sup>C NMR spectrum of *N*-phenyl-4-methoxybenzylamine (6f).

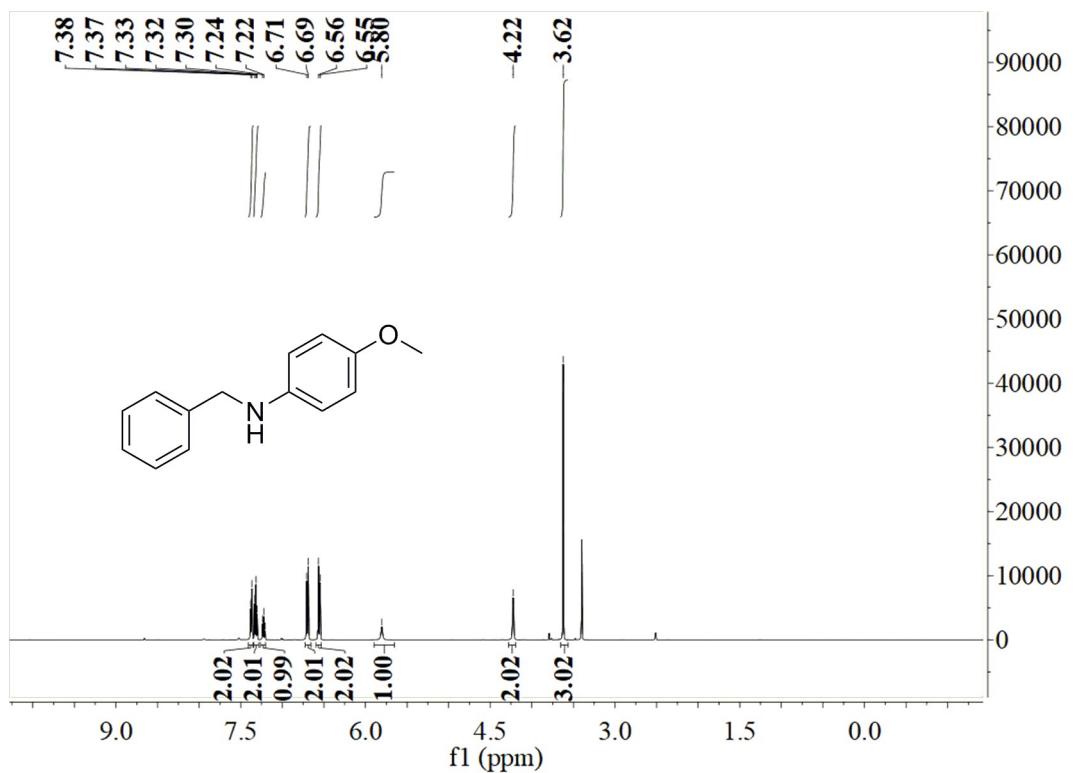


Figure <sup>1</sup>H NMR spectrum of *N*-benzyl-4-methoxyaniline (6g).

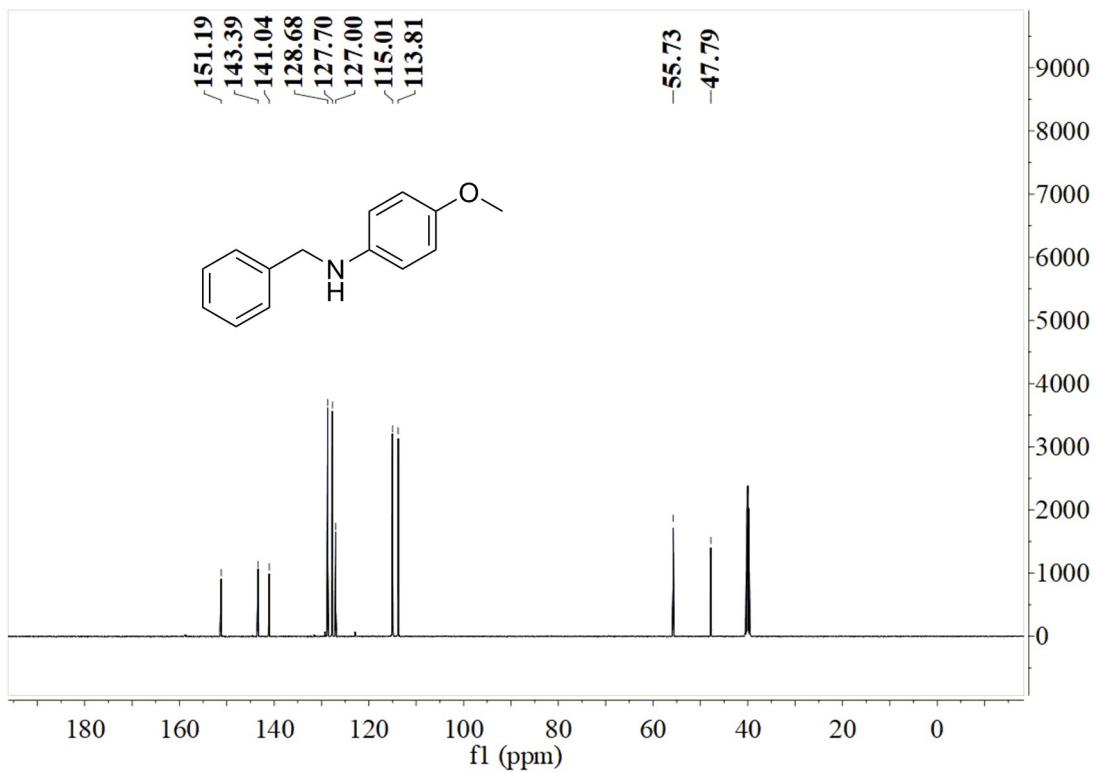


Figure <sup>13</sup>C NMR spectrum of *N*-benzyl-4-methoxyaniline (6g).

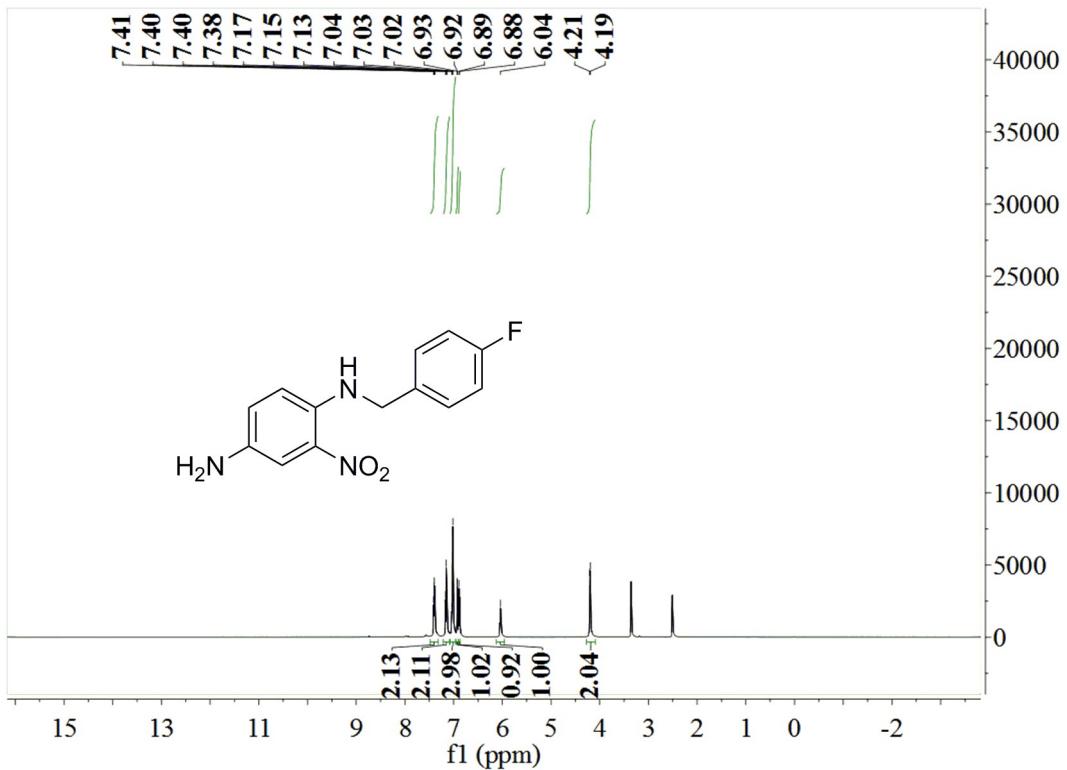


Figure <sup>1</sup>H NMR spectrum of 4-N-[(4-fluorophenyl)methyl]-2-nitrobenzene-1,4-diamine (6h).

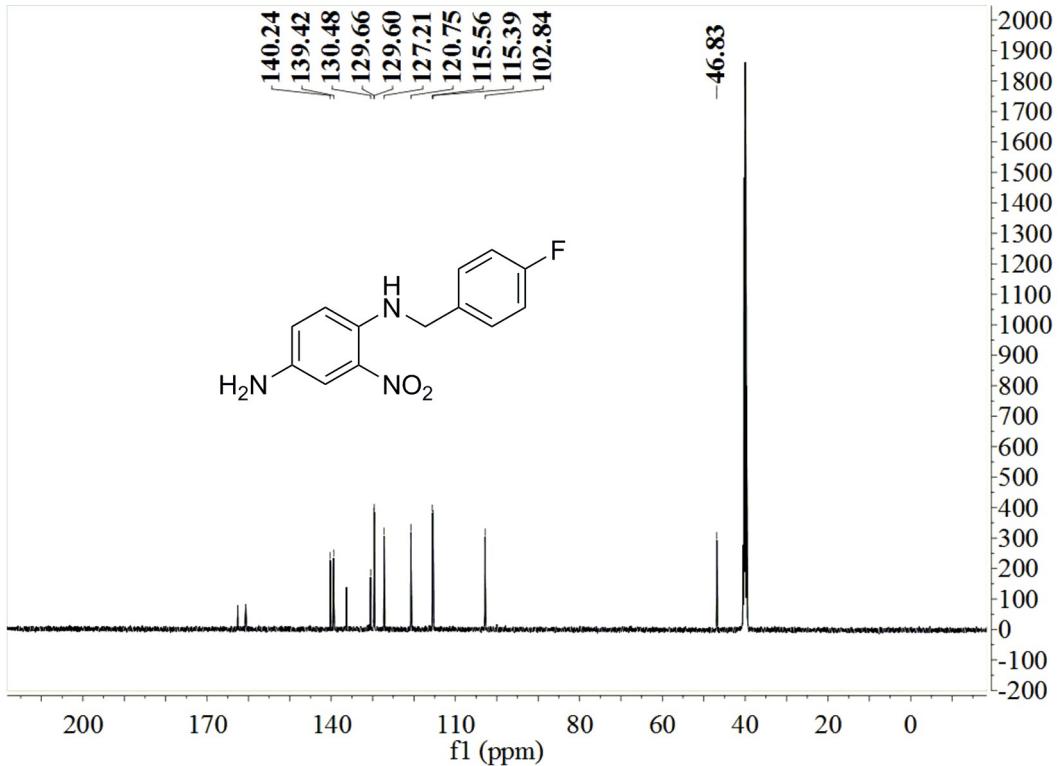


Figure <sup>13</sup>C NMR spectrum of 4-N-[(4-fluorophenyl)methyl]-2-nitrobenzene-1,4-diamine (6h).

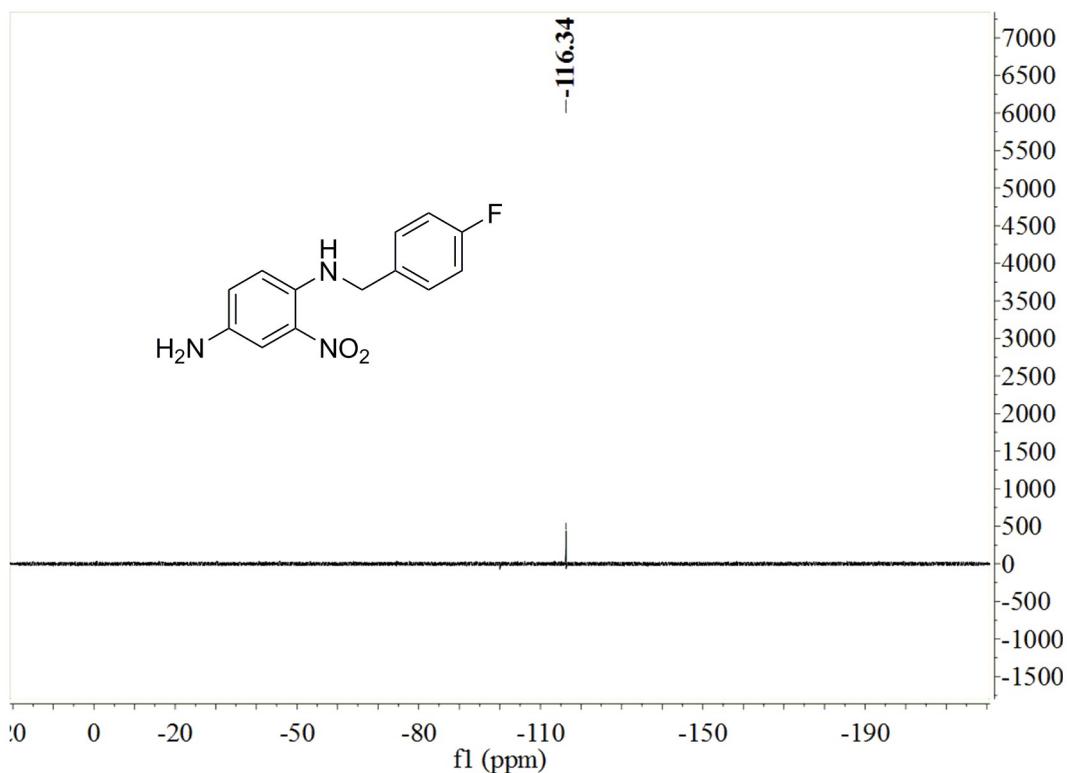


Figure  ${}^{19}\text{F}$  NMR spectrum of 4-N-[(4-fluorophenyl)methyl]-2-nitrobenzene-1,4-diamine (6h).

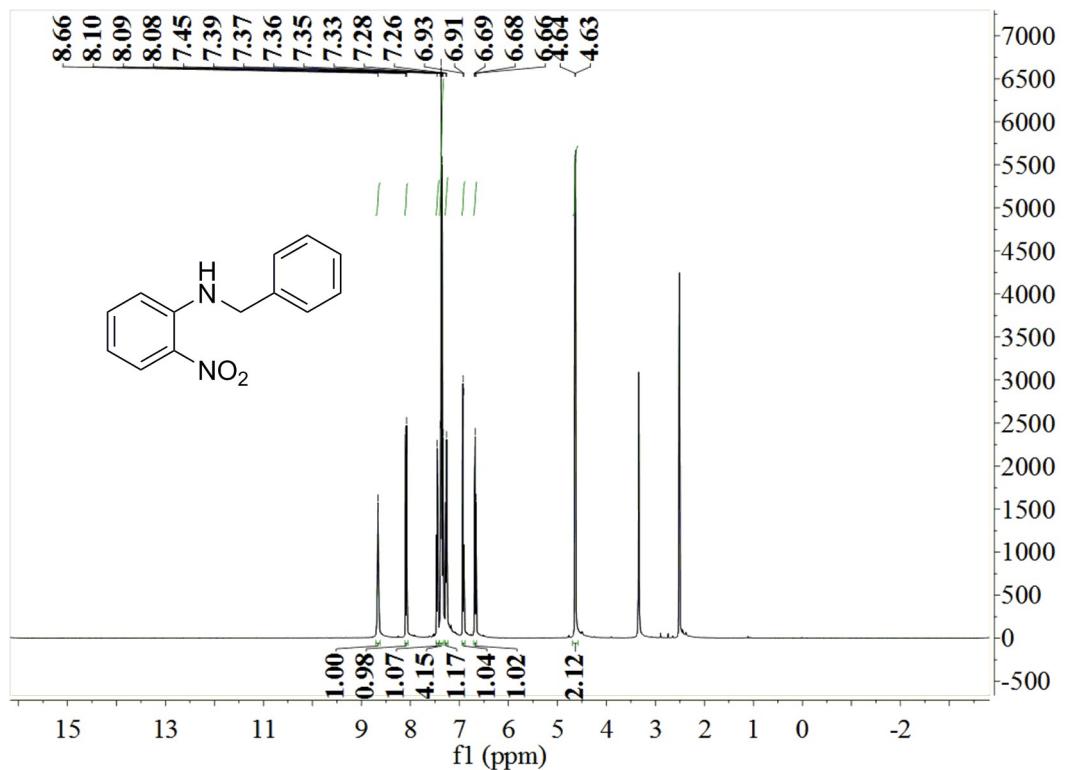


Figure  ${}^1\text{H}$  NMR spectrum of *N*-benzyl-2-nitroaniline (6i).

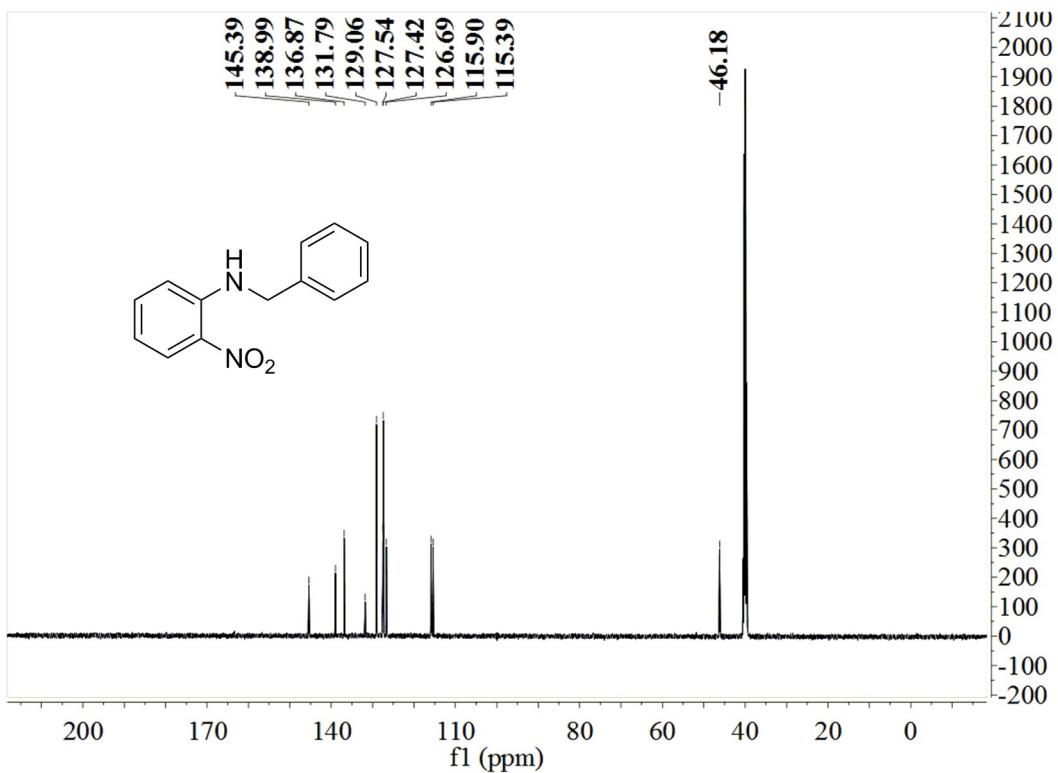


Figure  $^{13}\text{C}$  NMR spectrum of *N*-benzyl-2-nitroaniline (6i).

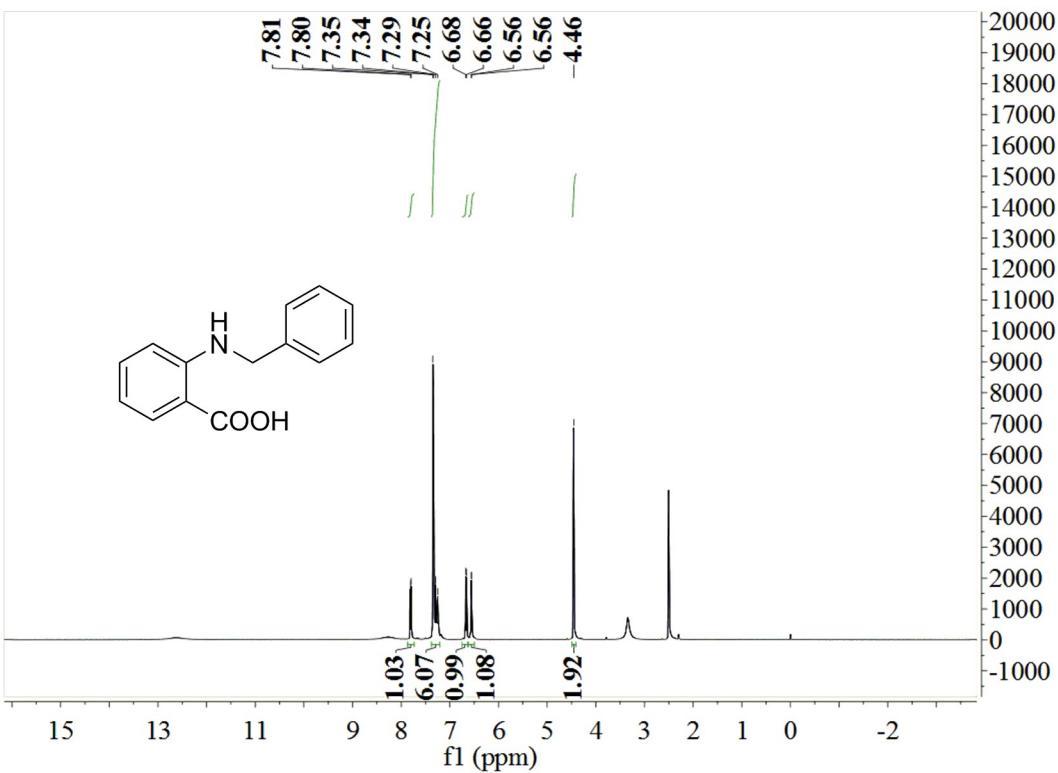


Figure  $^1\text{H}$  NMR spectrum of *N*-benzylanthranilic Acid (6j).

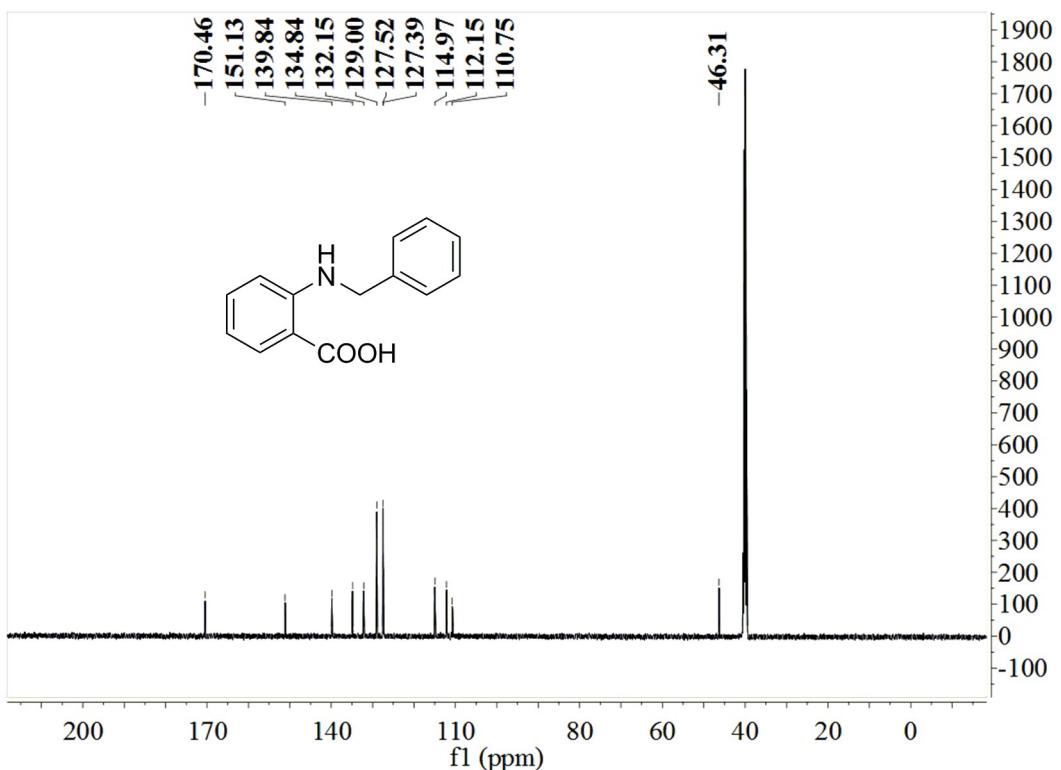


Figure  $^{13}\text{C}$  NMR spectrum of *N*-benzylanthranilic Acid (6j).

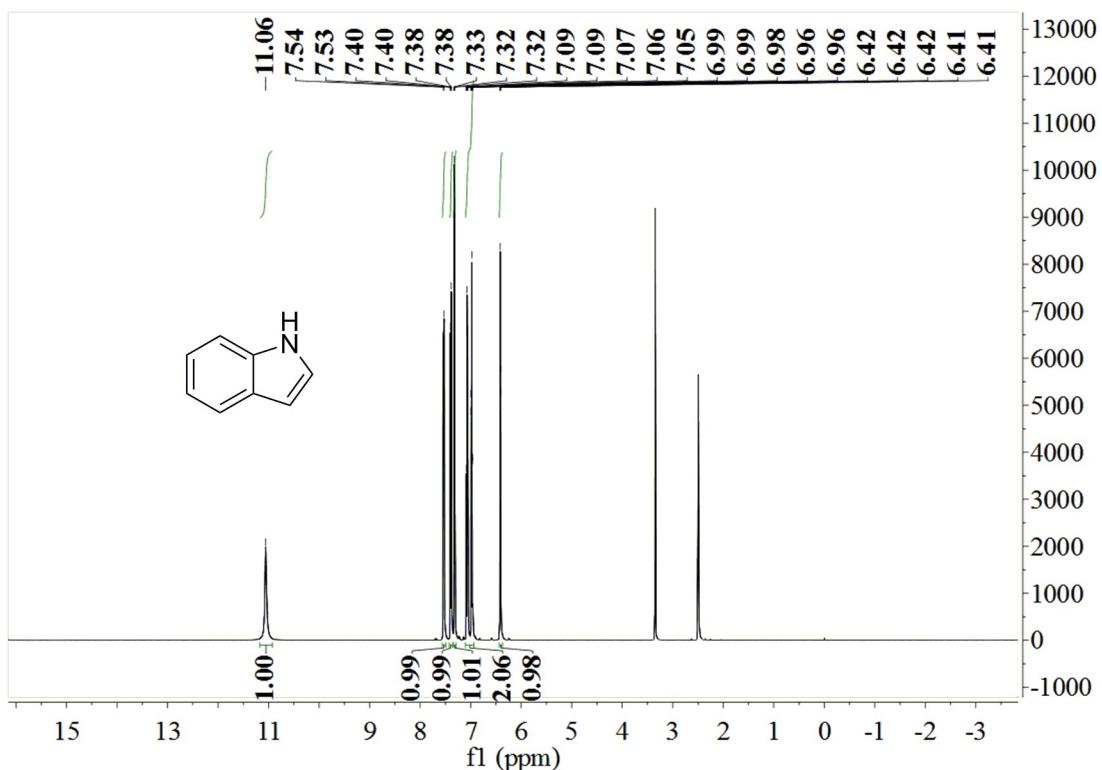


Figure  $^1\text{H}$  NMR spectrum of 1*H*-indole (8).

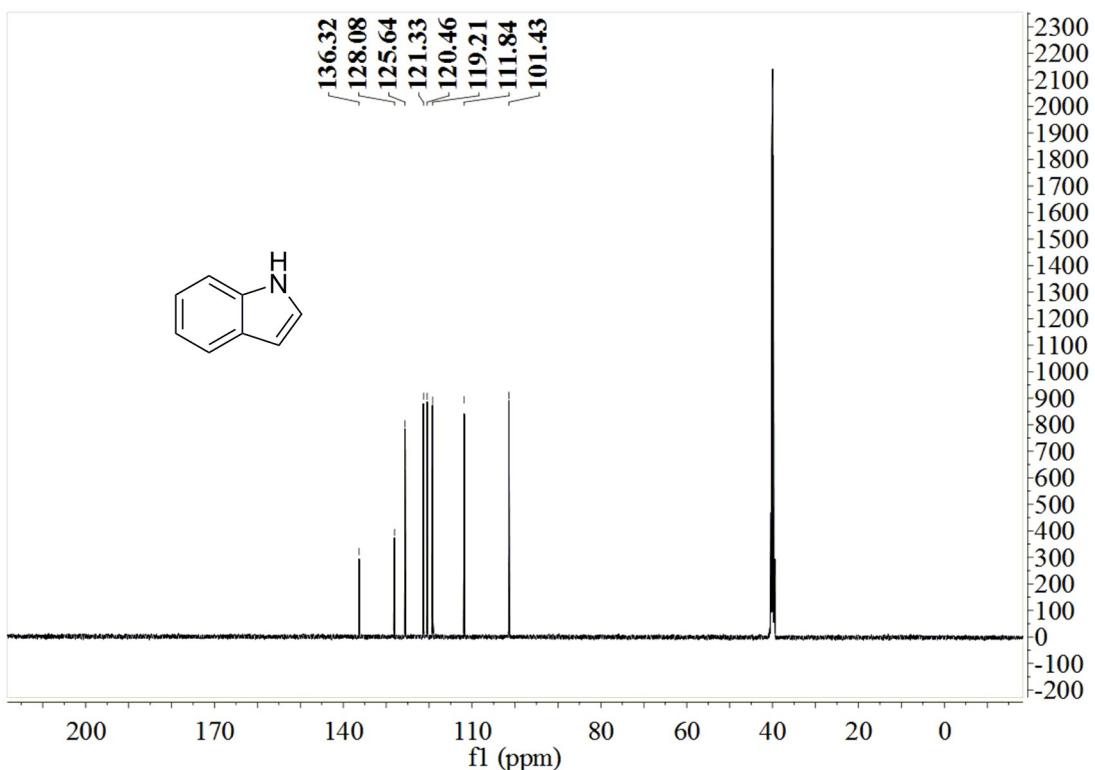


Figure  $^{13}\text{C}$  NMR spectrum of 1H-indole (8).

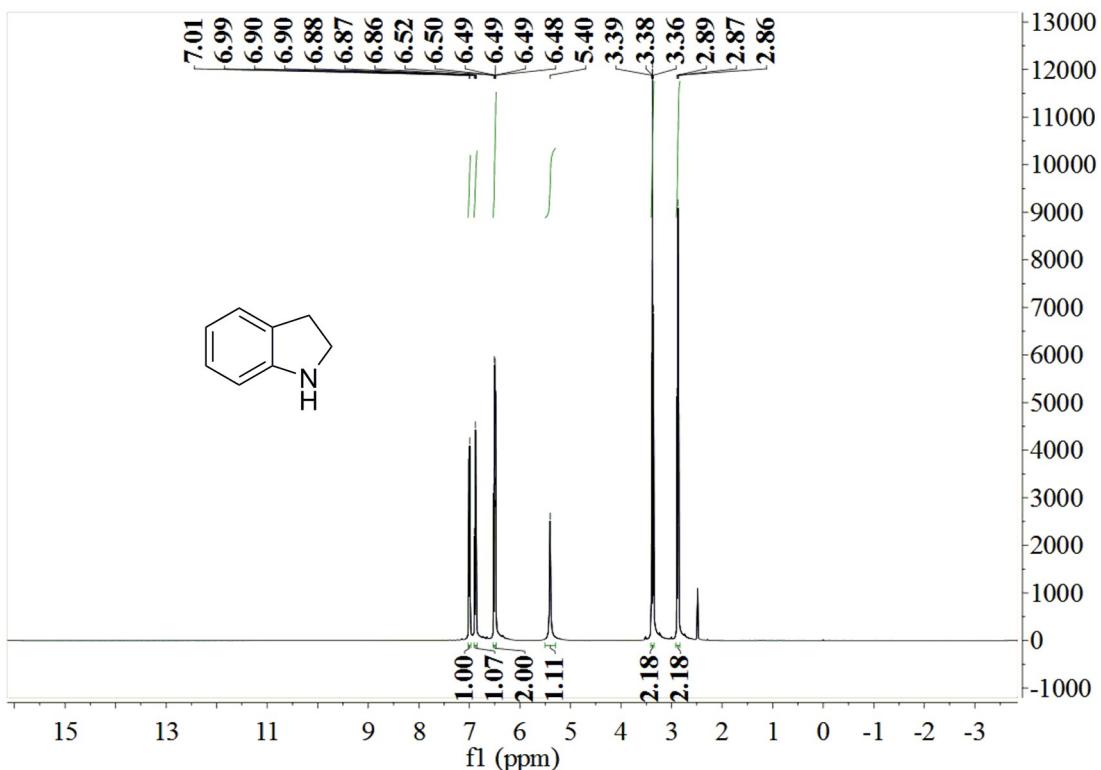


Figure  $^1\text{H}$  NMR spectrum of indoline (9).

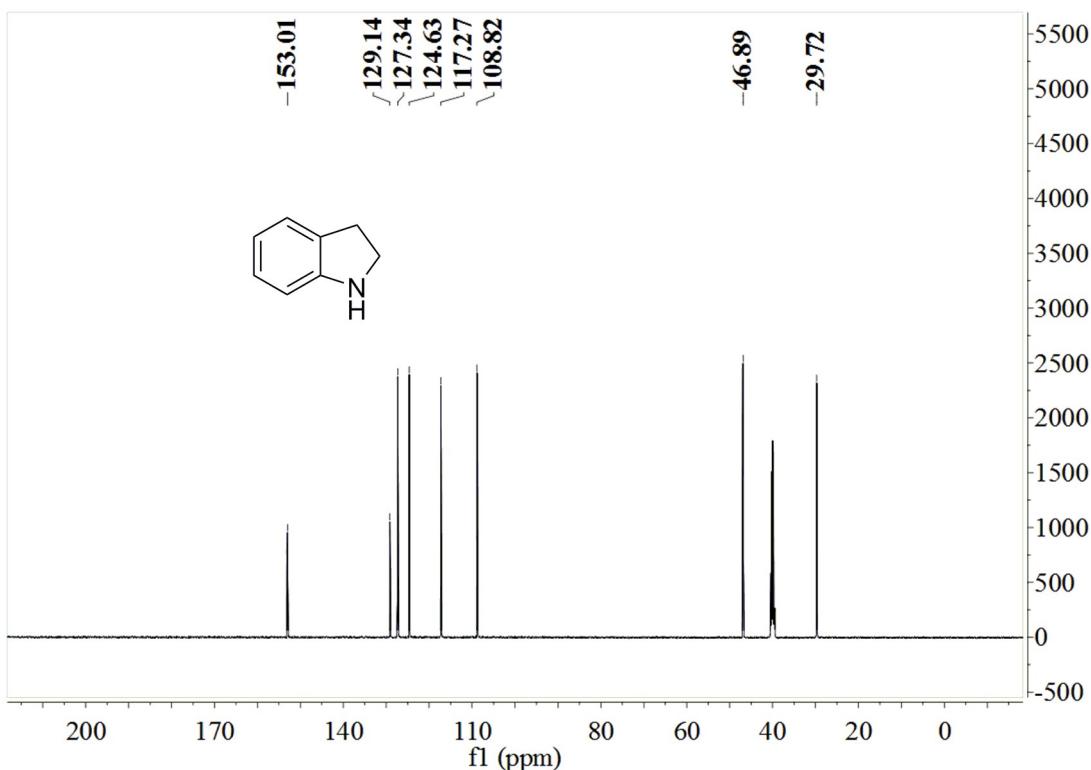


Figure  $^{13}\text{C}$  NMR spectrum of indoline (9).

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