

## Supplementary Information

### **1,4-dihydroxyanthraquinone-copper(II) supported on superparamagnetic Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>: An efficient catalyst for *N*-Arylation of nitrogen heterocycles and alkylamines with aryl halides and click synthesis of 1-aryl-1,2,3-triazole derivatives**

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**1-Phenyl-1H-imidazole (3a)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 7.14-7.45 (m, 7 H), 7.80 (s, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 118.2, 121.4, 127.5, 129.9, 130.4, 135.5, 137.4.

**2-Imidazol-1-ylpyridine (3g)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 7.14-7.20 (m, 2 H), 7.28-7.34 (m, 1 H), 7.25-7.34 (m, 1 H), 7.58 (s, 1 H), 7.73-7.80 (m, 1 H), 8.28 (s, 1 H), 8.41-8.43 (m, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 112.3, 116.7, 122.0, 130.6, 132.2, 134.9, 139.0, 149.1; Anal. Calcd for C<sub>8</sub>H<sub>7</sub>N<sub>3</sub>: C, 66.19; H, 4.86; N, 28.95%. Found: C, 65.89; H, 4.97; N, 29.14%.

**1-Phenyl-1H-indole (3h)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 6.59-6.63 (m, 1 H), 7.06-7.17 (m, 2 H), 7.25-7.30 (m, 2 H), 7.41-7.52 (m, 5 H), 7.59-7.64 (m, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 103.6, 110.5, 120.4, 121.1, 122.4, 124.4, 126.4, 128.0, 129.3, 129.6, 135.8, 139.8; Anal. Calcd for C<sub>14</sub>H<sub>11</sub>N: C, 87.01; H, 5.75; N 7.25%. Found: C, 86.91; H, 5.86; N, 7.23%.

**1-p-Tolyl-1H-indole (3i)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 2.35 (s, 3 H), 6.58 (dd, *J*= 4.2 Hz, 1 H), 7.04-7.32 (m, 7 H), 7.43-7.47 (m, 1 H), 7.58-7.62 (m, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 21.1, 103.2, 110.5, 120.1, 121.1, 122.2, 124.3, 128.1, 129.2, 130.1, 136.3, 137.3; Anal. Calcd for C<sub>15</sub>H<sub>13</sub>N: C, 86.92; H, 6.32; N, 6.76%. Found: C, 86.57; H, 6.55; N, 6.88%.

**1-Pyridin-2-yl-1H-indole (3j)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 6.56 (d, *J*= 3.5 Hz, 1 H), 6.97 (t, *J*= 6.0 Hz, 1 H), 7.00-7.25 (m, 3 H), 7.49-7.56 (m, 3 H), 8.09 (d, *J*= 8.2 Hz, 1 H), 8.36-8.39 (m, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 105.7, 113.3, 114.6, 120.2, 121.2, 121.5, 123.3, 126.1, 130.6, 135.2, 138.5, 149.0, 152.5; Anal. Calcd for C<sub>13</sub>H<sub>10</sub>N<sub>2</sub>: C, 80.39; H, 5.19; N, 14.42%. Found: C, 80.57; H, 5.09; N, 14.34%.

**2-Methyl-1-phenyl-1H-indole (3k)**

Dark brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 2.22 (s, 3 H), 6.32 (s, 1 H), 7.00-7.07 (m, 3 H), 7.25-7.29 (m, 2 H), 7.36-7.51 (m, 4 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 13.4, 101.3, 110.0, 119.6, 120.0, 121.0, 127.7, 128.0, 128.2, 129.4, 137.0, 138.2; Anal. Calcd for C<sub>15</sub>H<sub>13</sub>N: C, 86.92; H, 6.32; N, 6.76%. Found: C, 86.72; H, 6.45; N, 6.83%.

**1-Phenyl-1H-benzimidazole (3l)**

Brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 7.32-7.36 (m, 2 H), 7.44-7.61 (m, 6 H), 7.89-7.90 (m, 1 H), 8.17 (s, 1 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 110.5, 120.6, 122.8, 123.7, 124.0, 128.0, 130.0, 136.3, 142.8, 144.2; Anal. Calcd for C<sub>13</sub>H<sub>10</sub>N<sub>2</sub>: C, 80.39; H, 5.19; N, 14.42%. Found: C, 80.51; H, 5.01; N, 14.48%.

**1-Pyridin-2-yl-1H-benzimidazole (3n)**

Brown oil, <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>): δ= 7.26-7.39 (m, 3 H), 7.58 (d, *J*= 8.2 Hz, 1 H), 7.86-7.93 (m, 2 H), 8.06 (d, *J*= 7.8 Hz, 1 H), 8.58-8.62 (m, 2 H); <sup>13</sup>C NMR (62.9 MHz, CDCl<sub>3</sub>): δ= 112.6, 114.3, 120.6, 121.8, 123.3, 124.2, 132.1, 138.9, 141.3, 144.7, 149.5, 149.9; Anal. Calcd for C<sub>12</sub>H<sub>9</sub>N<sub>3</sub>: C, 73.83; H, 4.65; N, 21.52%. Found: C, 73.62; H, 4.76; N, 21.62%.

**1-Phenyl-1H-pyrrole (3o)**

Dark brown oil,  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 6.27 (t,  $J$ = 4.5 Hz, 2 H), 7.00 (t,  $J$ = 4.5 Hz, 2 H), 7.12-7.18 (m, 1 H), 7.28-7.33 (m, 4 H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 110.6, 119.4, 120.6, 125.7, 129.7, 140.9; Anal. Calcd for  $\text{C}_{10}\text{H}_9\text{N}$ : C, 83.88; H, 6.34; N, 9.78%. Found: C, 83.64; H, 6.43; N, 9.93%.

### 1,4-Diphenylpiperazine (3p)

Dark brown solid, mp 149-150 °C,  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 3.36 (s, 8 H), 6.88-7.02 (m, 6 H), 7.28-7.34 (m, 4 H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 49.5, 116.4, 120.1, 129.2, 151.3; Anal. Calcd for  $\text{C}_{16}\text{H}_{18}\text{N}_2$ : C, 80.63; H, 7.61; N, 11.75%. Found: C, 80.89; H, 7.52; N, 11.59%.

### 1-Phenyl-4-pyridin-2-ylpiperazine (3r)

Light brown solid, mp 100-101 °C,  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 3.24 (dd,  $J^1$ = 7.0 Hz,  $J^2$ = 5.0 Hz, 4 H), 3.64 (t,  $J$ = 5.2 Hz, 4 H), 6.56-6.65 (m, 2 H), 6.79-6.93 (m, 3H), 7.18-7.25 (m, 2 H), 7.40-7.47 (m, 1 H), 8.14-8.16 (m, 1 H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 45.3, 49.2, 107.2, 113.6, 116.3, 120.1, 129.2, 137.6, 148.0, 151.3, 159.4; Anal. Calcd for  $\text{C}_{15}\text{H}_{17}\text{N}_3$ : C, 75.28; H, 7.16; N, 17.56%. Found: C, 75.38; H, 7.27; N, 17.35%.

### N,N-Dibutylaniline (3s)

Black oil,  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 0.88 (t,  $J$ = 7.5 Hz, 6 H), 1.20-1.32 (m, 4 H), 1.46-1.55 (m, 4 H), 3.18 (t,  $J$ = 7.5 Hz, 4 H), 6.54-6.58 (m, 3 H), 7.09-7.17 (m, 2 H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 14.0, 20.4, 29.4, 50.8, 111.6, 115.0, 129.2, 148.2;

### N,N-Diethylaniline (3t)

Dark brown oil,  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 1.18 (t, 6 H), 3.37 (q, 4 H), 6.63-6.73 (m, 3 H), 7.20-7.27 (m, 2 H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ ):  $\delta$ = 12.6, 44.3, 111.8, 115.3, 129.3, 147.8; Anal. Calcd for  $\text{C}_{10}\text{H}_{15}\text{N}$ : C, 80.48; H, 10.13; N, 9.39%. Found: C, 80.25; H, 10.03; N, 9.72%.

**1,4-Diphenyl-1H-1,2,3-triazole (7a)**: White solid; mp: 174-175 °C (175-177 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.26 (d,  $J$ = 1.6 Hz, 1H), 7.38 (t,  $J$ = 7.6 Hz, 1H), 7.47 (t,  $J$ =7.6 Hz, 2H), 7.56 (t,  $J$ = 8.0 Hz, 2H), 7.81 (d,  $J$ = 8.0 Hz, 2H), 7.93 (d,  $J$ = 7.6 Hz, 2H), 8.20 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 117.6, 120.6, 125.9, 128.4, 128.8, 128.9, 129.8, 130.3, 137.1, 148.5; IR (KBr,  $\text{Cm}^{-1}$ ): 3118, 2919, 2356, 1729, 1463, 1236, 1070.

**1-(4-Hydroxyphenyl)-4-phenyl-1H-1,2,3-triazole (7b)**: White solid; mp: 207-208 °C (206-208 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CD}_3\text{OD}$ )  $\delta$ : 6.96 (d,  $J$ = 9.2 Hz, 2H), 7.37 (t,  $J$ = 6.0 Hz, 1H), 7.46 (t,  $J$ = 7.6 Hz, 2H), 7.68 (d,  $J$ = 6.4 Hz, 2H), 7.91 (d,  $J$ = 7.2 Hz, 2H), 8.75 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{DMSO}-d_6$ )  $\delta$ : 116.0, 119.5, 121.9, 125.3, 128.1, 128.8, 128.9, 130.4, 146.9, 157.8; IR (KBr,  $\text{Cm}^{-1}$ ): 3135, 1603, 1520, 1229, 1055.

**1-(4-Methoxyphenyl)-4-phenyl-1H-1,2,3-triazole (7c)**: White solid; mp: 156-157 °C (155-159 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 3.89, (s, 3H), 7.06 (d,  $J$ = 7.2 Hz, 2H), 7.35 (t,  $J$ = 7.2 Hz, 1H), 7.46 (t,  $J$ = 7.4 Hz, 2H), 7.71 (d,  $J$ = 7.4 Hz, 2H), 7.92 (d,  $J$ = 7.2 Hz), 8.11 (s, 1H); IR (KBr,  $\text{Cm}^{-1}$ ): 3133, 2956, 2356, 1722, 1517, 1229, 1041.

**1-(4-Phenoxy-phenyl)-4-phenyl-1H-1,2,3-triazole (7d)**: White solid; mp: 170-171 °C (171-173 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.08 (d,  $J$ = 8.5 Hz, 2H), 7.16 (m, 3H), 7.34 (m, 3H), 7.44 (t,  $J$ = 7.7 Hz, 2H), 7.74 (d,  $J$ = 8.9 Hz, 2H), 7.92 (d,  $J$ = 8.3 Hz, 2H), 8.15 (s, 1H);

$^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 117.8, 119.3, 119.5, 122.3, 124.2, 125.9, 128.4, 128.9, 130.0, 132.3, 148.4, 156.4, 158.0; IR (KBr,  $\text{Cm}^{-1}$ ): 3125, 2923, 2356, 1585, 1511, 1247.

**1-(3-Formylphenyl)-4-phenyl-1H-1,2,3-triazole (7e)**: White solid; mp: 164-166 °C (162-165 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.41 (t,  $J$ = 4.0 Hz, 1H), 7.50 (t,  $J$ = 5.6 Hz, 2H), 7.77 (t,  $J$ = 8.0 Hz, 1H), 7.94 (d,  $J$ = 7.2 Hz, 2H), 7.99 (d,  $J$ = 7.6 Hz, 1H), 8.19 (d,  $J$ = 7.2 Hz, 1H), 8.30 (d,  $J$ = 7.61 Hz, 2H), 10.13 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 117.4, 120.1, 125.9, 128.7, 129.0, 129.9, 130.6, 130.8, 137.7, 137.8, 148.9, 190.8; IR (KBr,  $\text{Cm}^{-1}$ ): 3137, 2817, 1703, 1597, 1238, 1011.

**1-Thiophen-3-yl-4-phenyl-1H-1,2,3-triazole (7f)**: White solid; mp: 166-167 °C (164-166 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.32-7.38 (m, 1H), 7.44 (m, 1H), 7.48-7.50 (m, 1H), 7.52-7.53 (m, 1H), 7.61-7.62 (q,  $J$ = 1.2, 3.2 Hz, 1H), 7.68 (m, 1H), 7.90 (s, 1H), 7.92 (d,  $J$ = 1.6 Hz, 1H), 8.11 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 114.1, 118.0, 120.9, 125.9, 127.3, 128.5, 128.9, 130.2, 135.9, 148.0; IR (KBr,  $\text{Cm}^{-1}$ ): 3107, 1559, 1445, 1229, 1072.

**1-(4-Hydroxyphenyl)-4-cyclohexyl-1H-1,2,3-triazole (7g)**: White solid; mp: 169-170 °C (167-169 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 1.23-1.35 (m, 2H), 1.38-1.50 (m, 3H), 1.76 (m, 1H), 1.85 (m, 2H), 2.14 (m, 2H), 2.86 (m, 1H), 6.25 (brs, OH), 7.03 (d,  $J$ = 8.8 Hz, 2H), 7.56-7.60 (m, 3H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 26.0, 26.1, 33.0, 35.2, 116.6, 118.1, 122.2, 130.0, 154.0, 157.4; IR (KBr,  $\text{Cm}^{-1}$ ): 3141, 2928, 2358, 1601, 1517, 1225, 1063.

**1-(4-Methoxyphenyl)-4-cyclohexyl-1H-1,2,3-triazole (7h)**: White solid; mp: 91-92 °C (90-93 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 1.26-1.30 (m, 1H), 1.40 (m, 4H), 1.80-1.90 (m, 3H), 2.18 (m, 2H), 2.82 (m, 1H), 3.82 (s, 3H), 6.98 (d,  $J$ = 6.8 Hz, 2H), 7.62 (m, 3H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 26.2, 29.4, 33.1, 35.8, 55.0, 114.7, 117.2, 121.8, 132.5, 154.2, 159.6; IR (KBr,  $\text{Cm}^{-1}$ ): 3130, 2924, 2361, 1725, 1519, 1254, 1046.

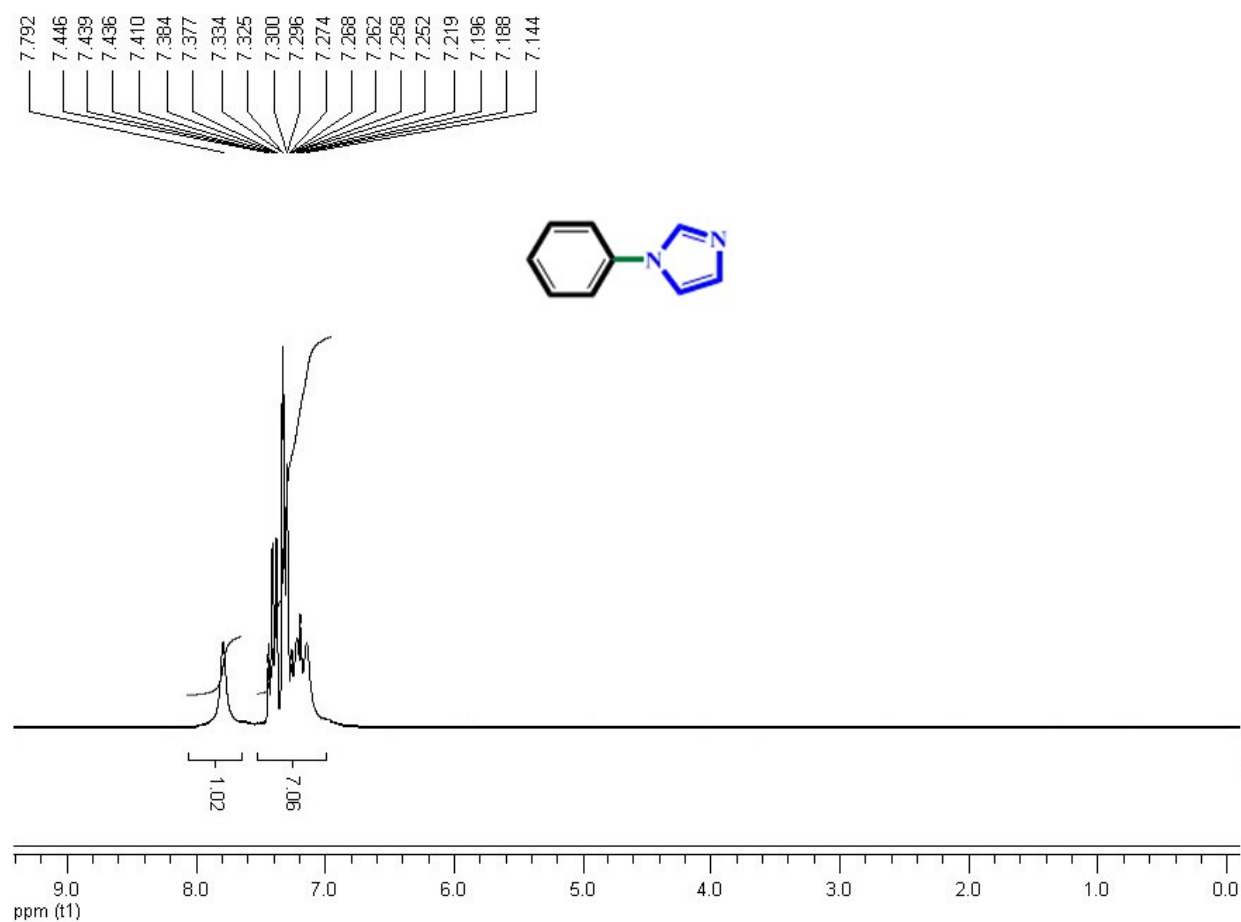
**1-(Naphth-2-yl)-4-cyclohexyl-1H-1,2,3-triazole (7i)**: White solid; mp: 141-142 °C (139-141 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 1.22-1.36 (m, 1H), 1.39-1.55 (m, 4H), 1.79 (m, 1H), 1.88 (m, 2H), 2.16 (m, 2H), 2.90-2.92 (m, 1H), 7.51-7.58 (m, 2H), 7.81 (s, 1H), 7.87-7.92 (m, 3H), 8.00 (d,  $J$ = 8.8 Hz, 1H), 8.15 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 26.1, 26.2, 29.7, 33.0, 35.4, 117.7, 118.1, 119.0, 126.8, 127.3, 127.9, 128.2, 129.9, 132.7, 133.3, 134.8, 154.5; IR (KBr,  $\text{Cm}^{-1}$ ): 3128, 2926, 1566, 1501, 1239, 1038.

**1-(Indol-5-yl)-4-cyclohexyl-1H-1,2,3-triazole (7j)**: Light pink solid; mp: 138-139 °C (136-139 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 1.21-1.37 (m, 2H), 1.36-1.51 (m, 3H), 1.77 (m, 1H), 1.86 (m, 2H), 2.16 (m, 2H), 2.87 (m, 1H), 6.63 (t,  $J$ = 2.4 Hz, 1H), 7.33 (t,  $J$ = 2.8 Hz, 1H), 7.47-7.56 (m, 2H), 7.68 (s, 1H), 7.91 (d,  $J$ = 1.6 Hz, 1H), 8.48 (s, 1H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 26.1, 26.2, 33.1, 35.4, 103.3, 111.8, 113.2, 115.9, 118.5, 126.2, 128.0, 130.9, 135.4, 154.0; IR (KBr,  $\text{Cm}^{-1}$ ): 3349, 2921, 2360, 1444, 1223, 1046.

**1-(4-Hydroxyphenyl)-4-octyl-1H-1,2,3-triazole (7k)**: Light pink solid; mp: 99-101 °C (98-100 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 0.88 (t,  $J$ = 6.8 Hz, 3H), 1.26-1.39 (m, 10H), 1.72-1.76 (m, 2H), 2.79 (t,  $J$ = 7.6 Hz, 2H), 7.06 (d,  $J$ = 7.2 Hz, 2H), 7.58 (d,  $J$ = 6.8 Hz, 2H), 7.64 (s, 1H); 14.1, 22.7, 25.5, 29.2 (2 C), 29.3, 29.4, 31.9, 116.7, 119.5, 122.3, 129.9, 148.8, 157.8; IR (KBr,  $\text{Cm}^{-1}$ ): 3122, 2920, 1600, 1236, 1057.

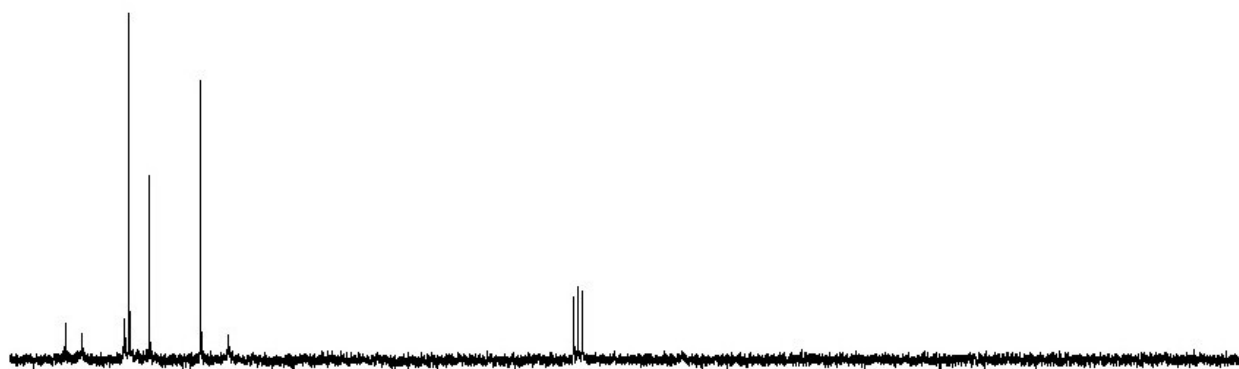
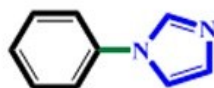
**1-(4-Methoxyphenyl)-4-octyl-1H-1,2,3-triazole (7l)**: Light yellow solid; mp: 54-56 °C (53-55 °C)<sup>58</sup>;  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ )  $\delta$ : 0.94 (t,  $J$ = 6.4 Hz, 3H), 1.27-1.43 (m, 10H), 1.72

(m, 2H), 2.82 (t,  $J=7.6$  Hz, 2H), 3.89 (s, 3H), 7.03 (d,  $J=6.8$  Hz, 2H), 7.61-7.71 (m, 3H);  $^{13}\text{C}$  NMR (62.9 MHz,  $\text{CDCl}_3$ )  $\delta$ : 14.1, 22.7, 25.7, 29.4, 29.5, 29.7, 30.4, 31.9, 55.6, 114.7, 119.0, 122.4, 132.5, 149.0, 159; IR (KBr,  $\text{Cm}^{-1}$ ): 3127, 2919, 2357, 1725, 1521, 1256, 1048.



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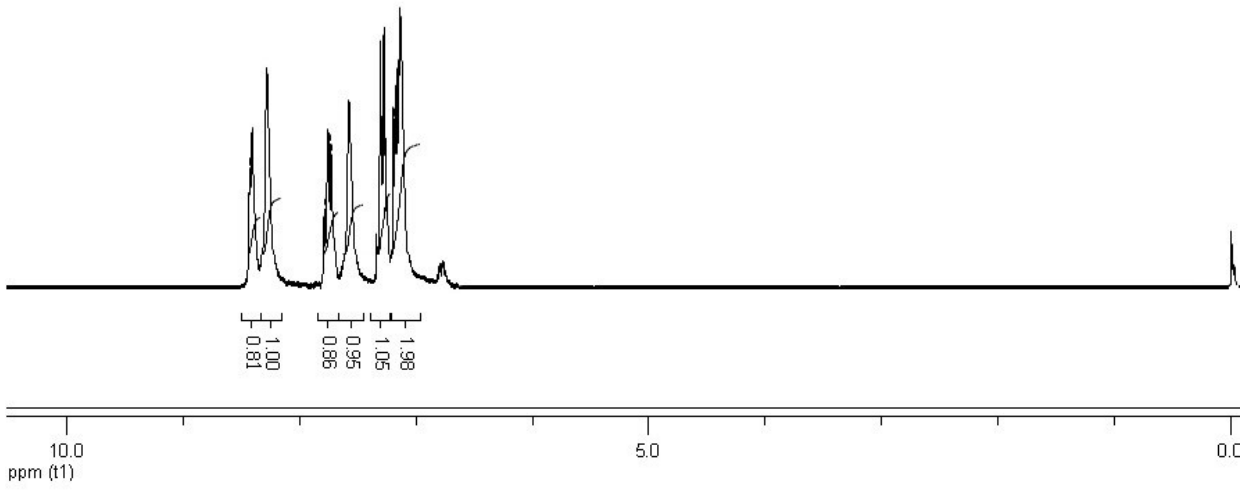
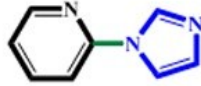
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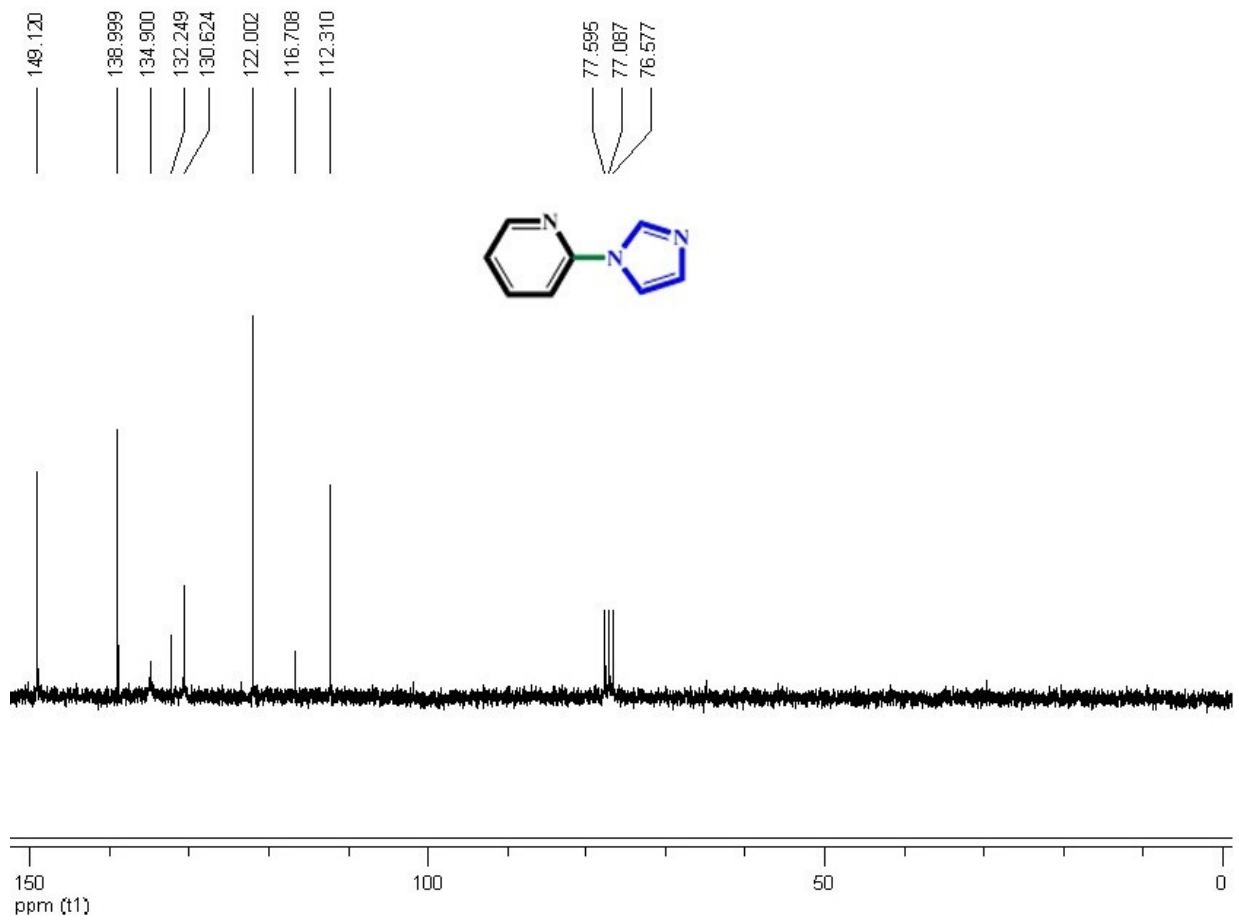
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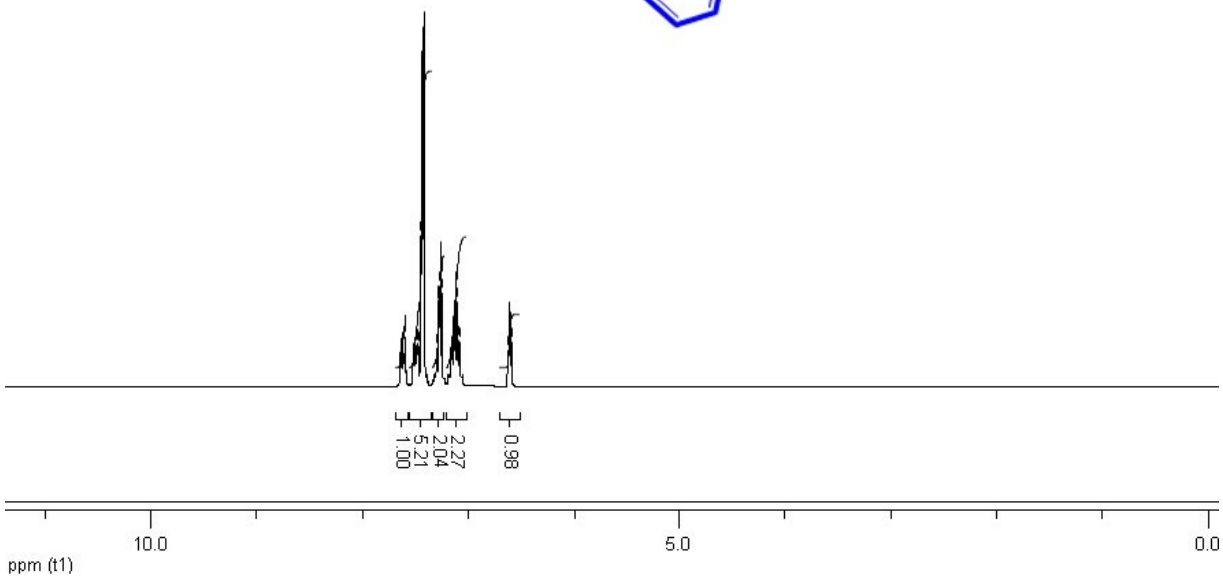
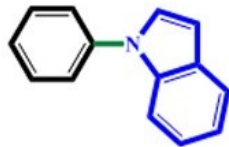
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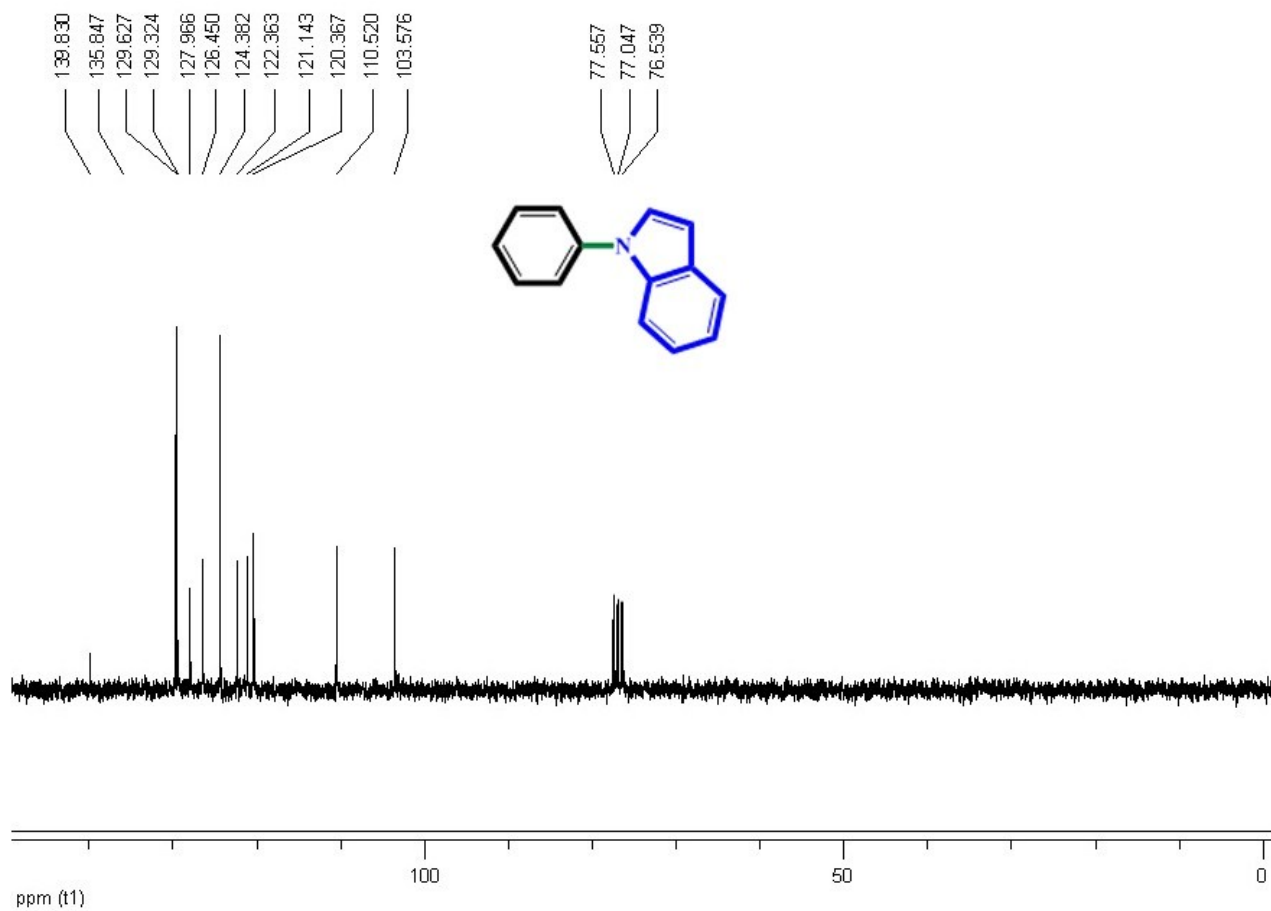


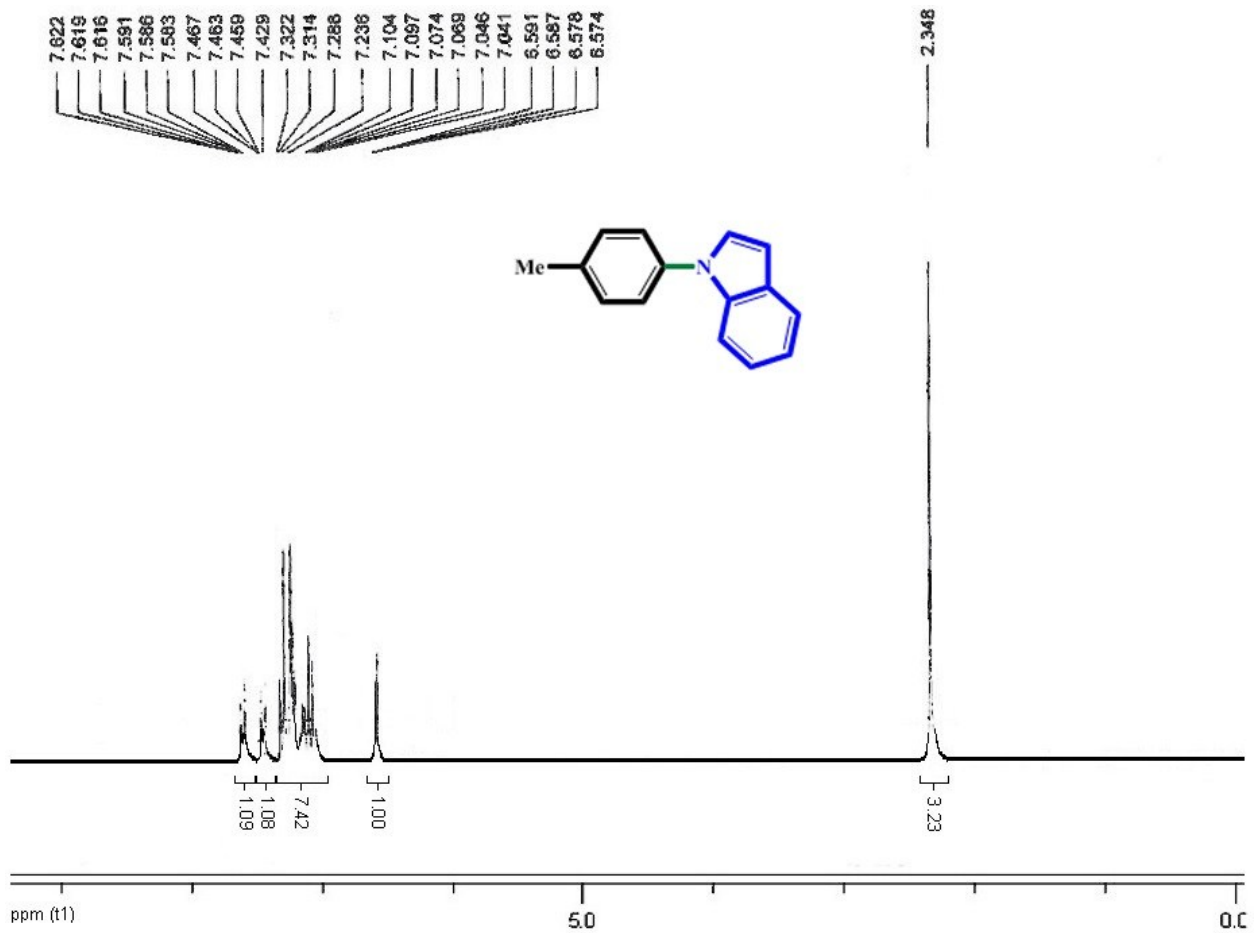


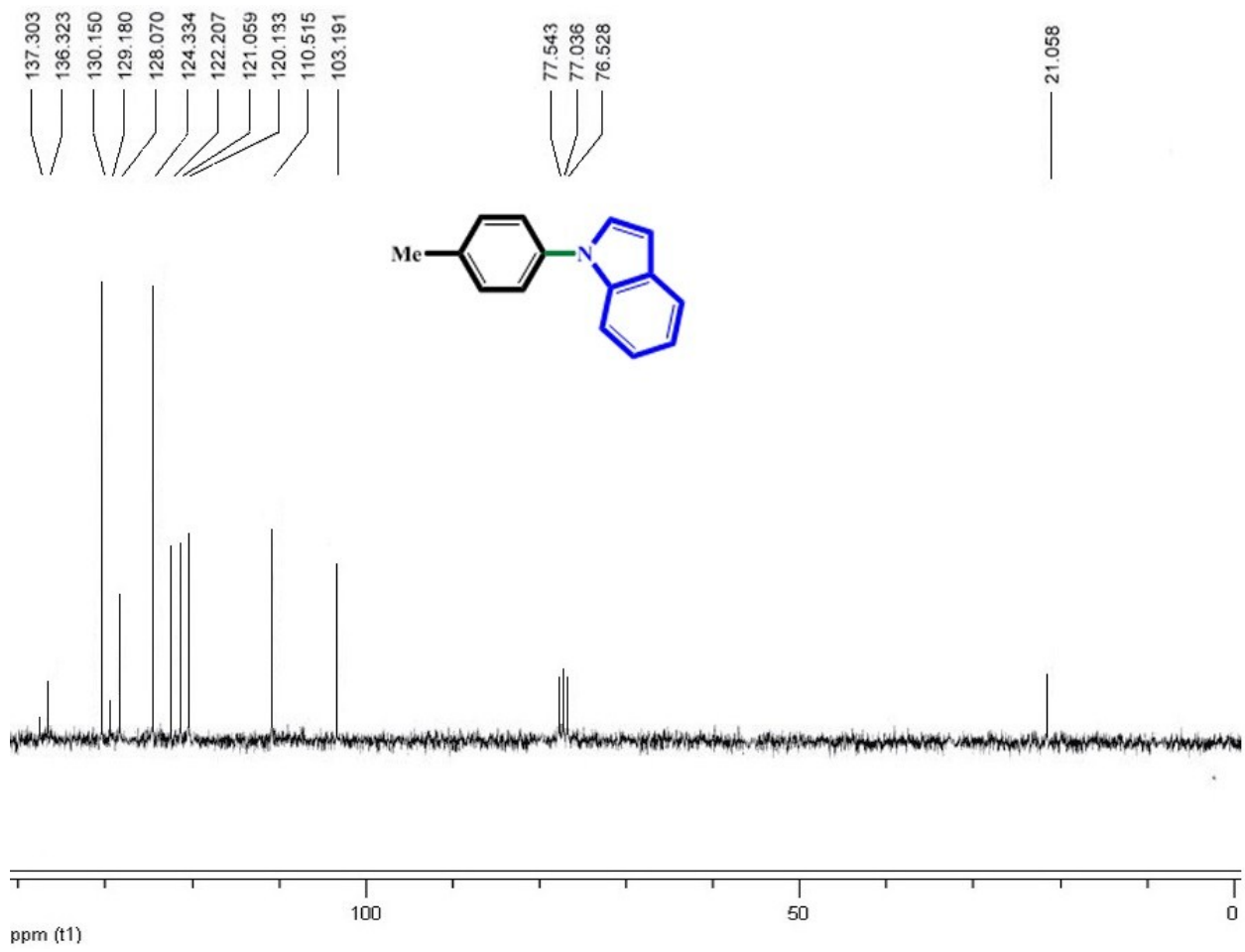


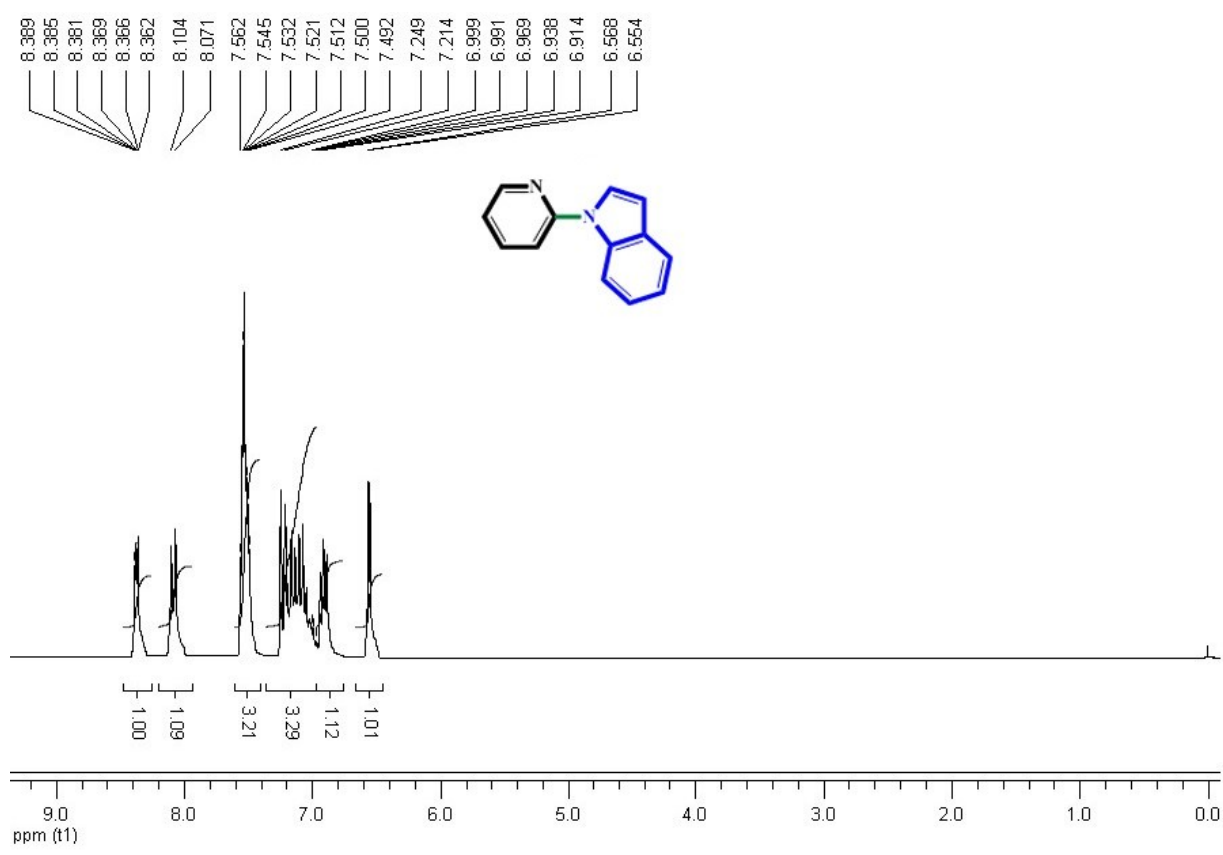
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7.280  
7.272  
7.263  
7.250  
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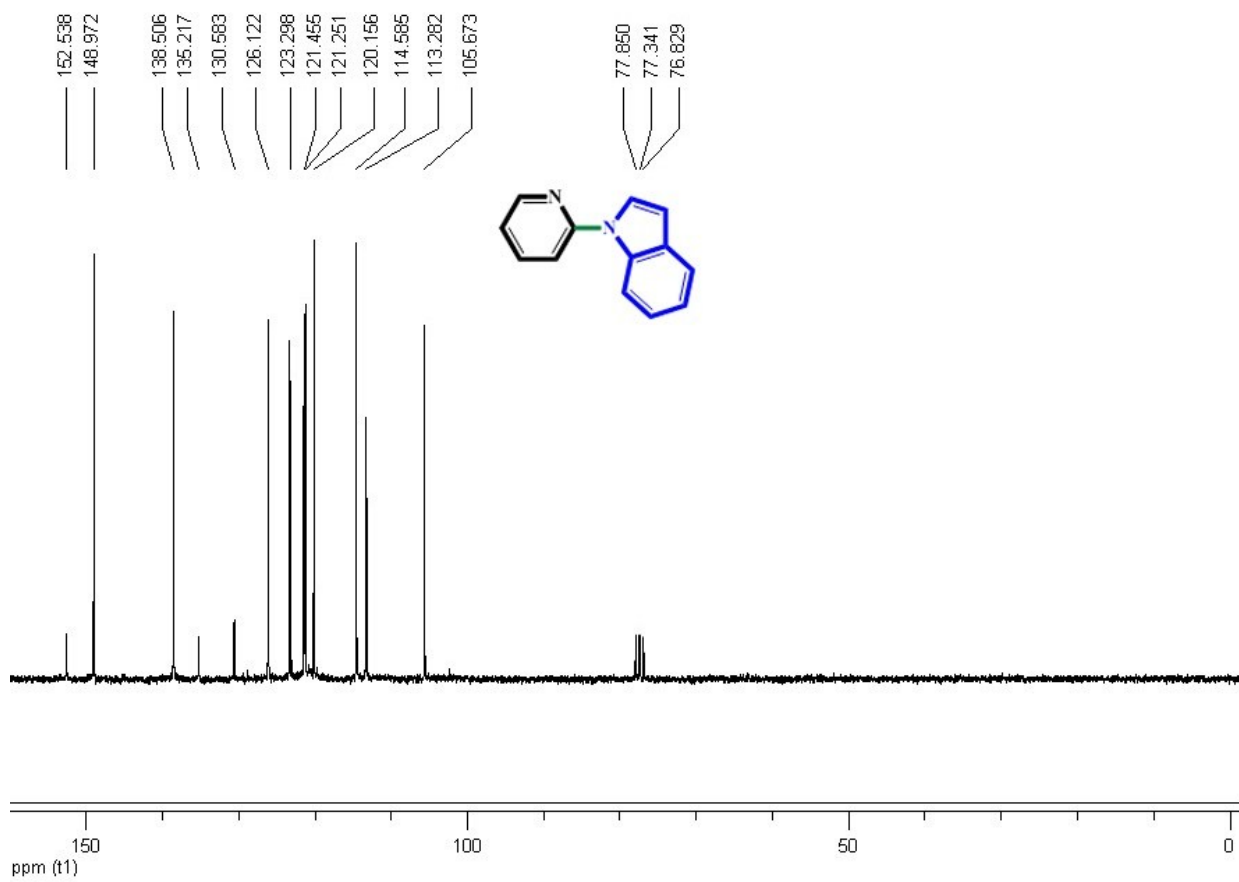


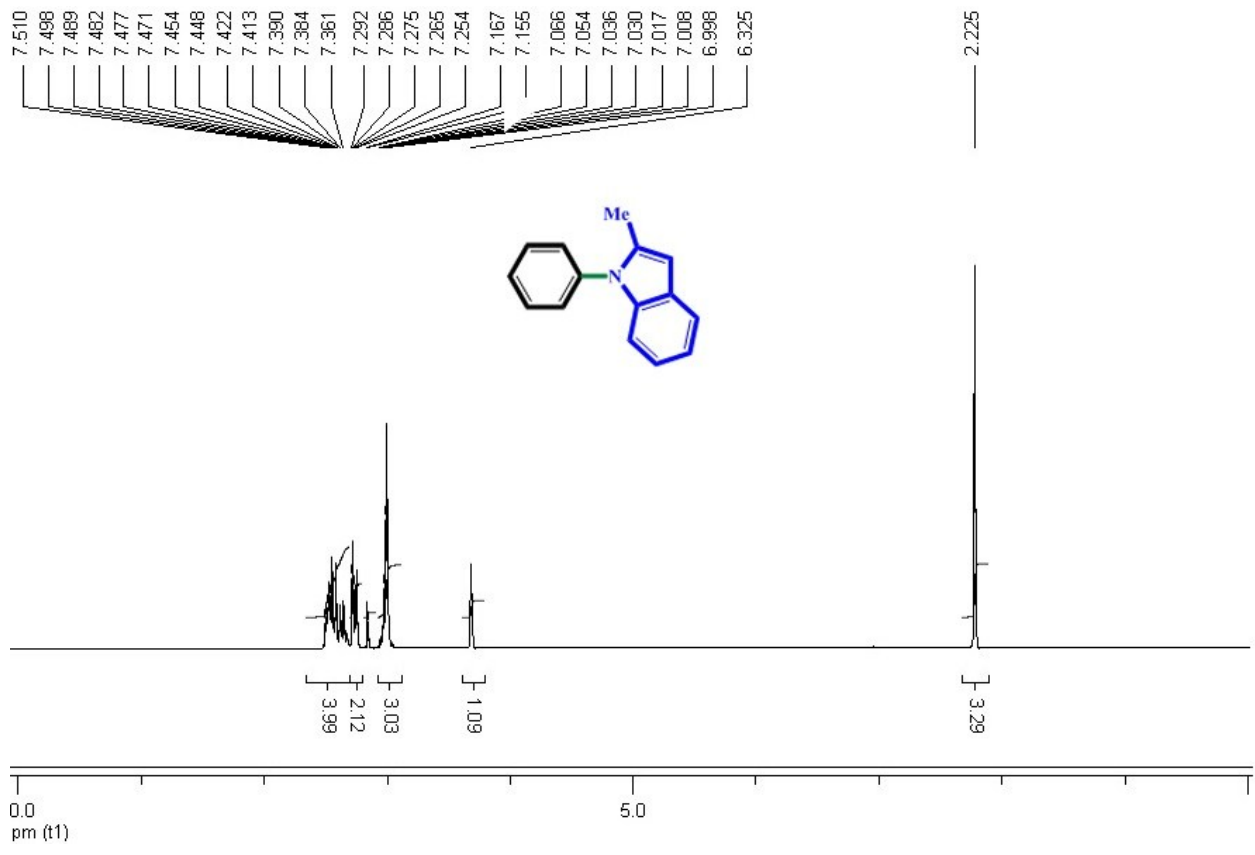


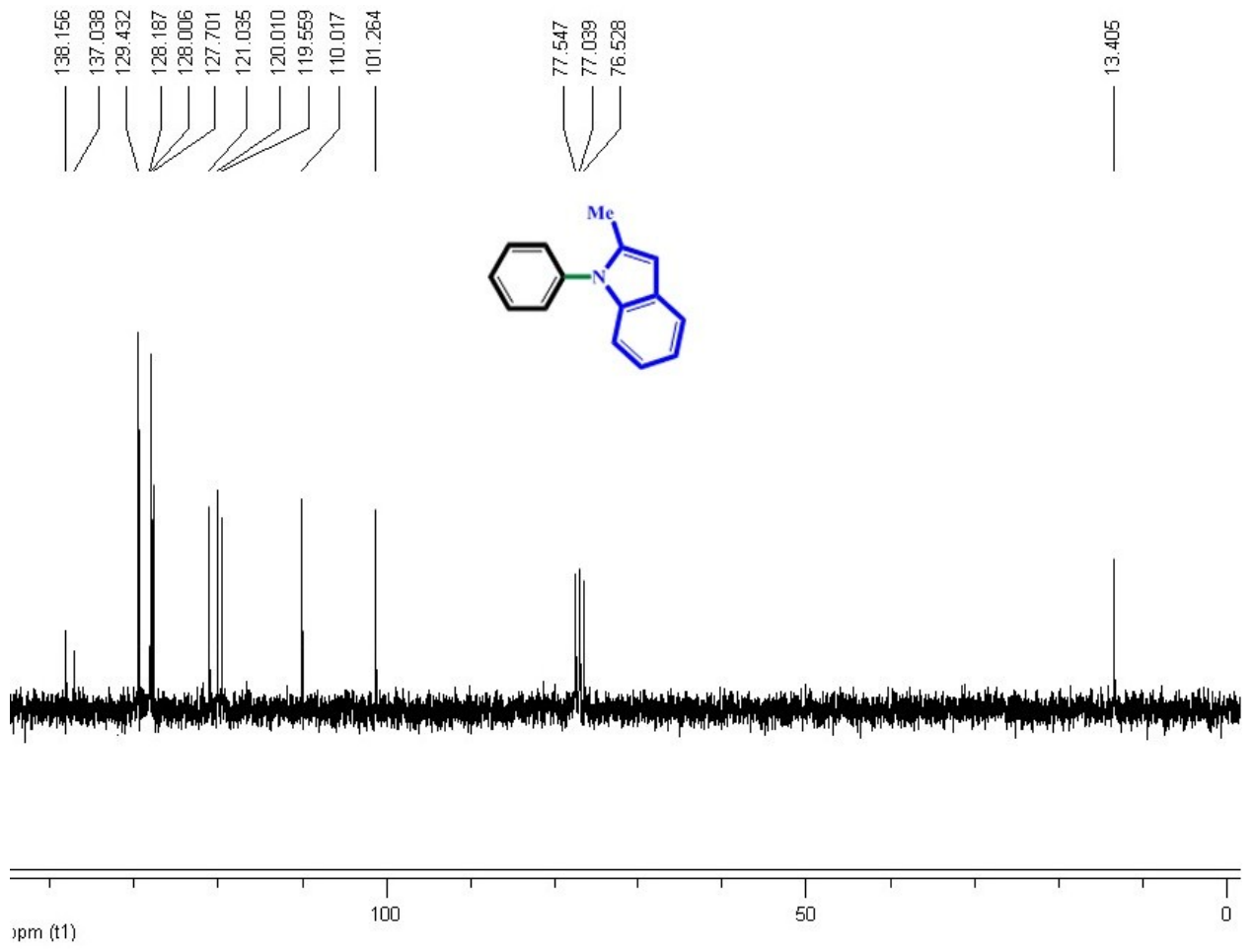




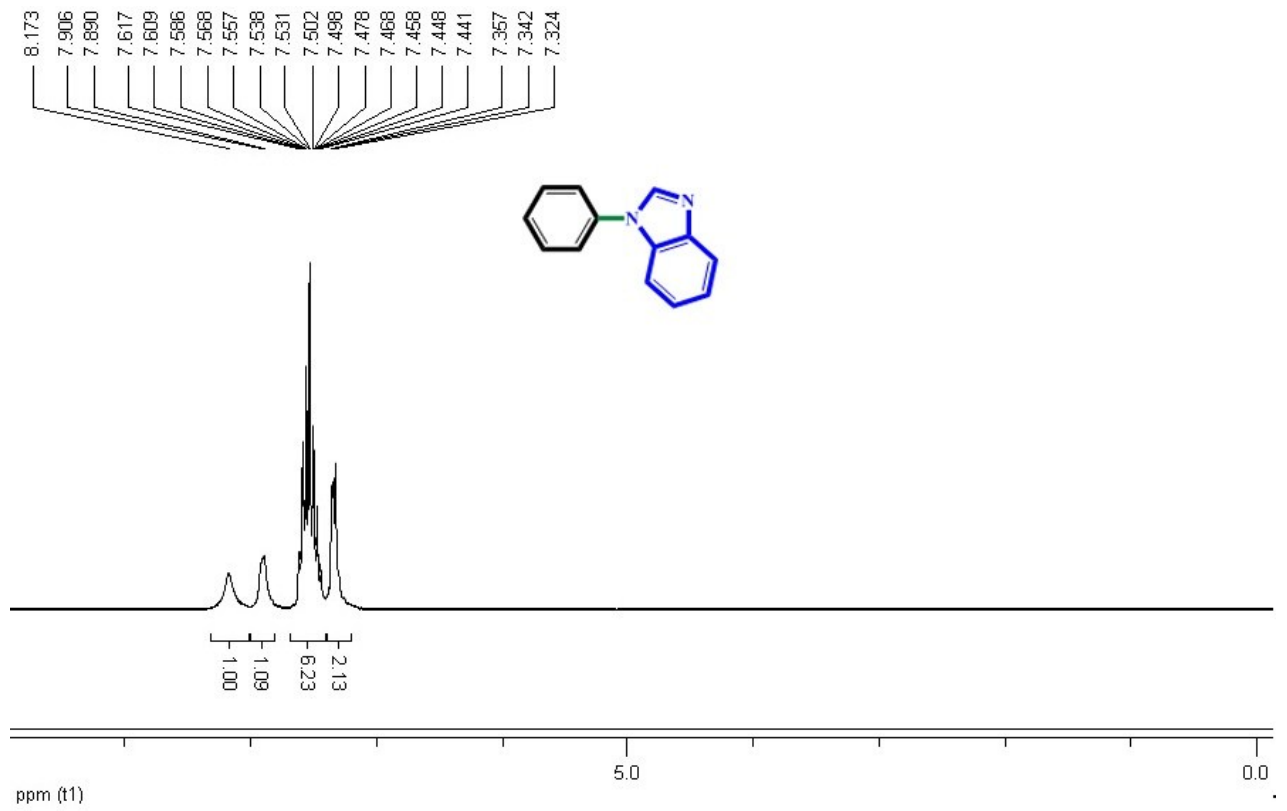


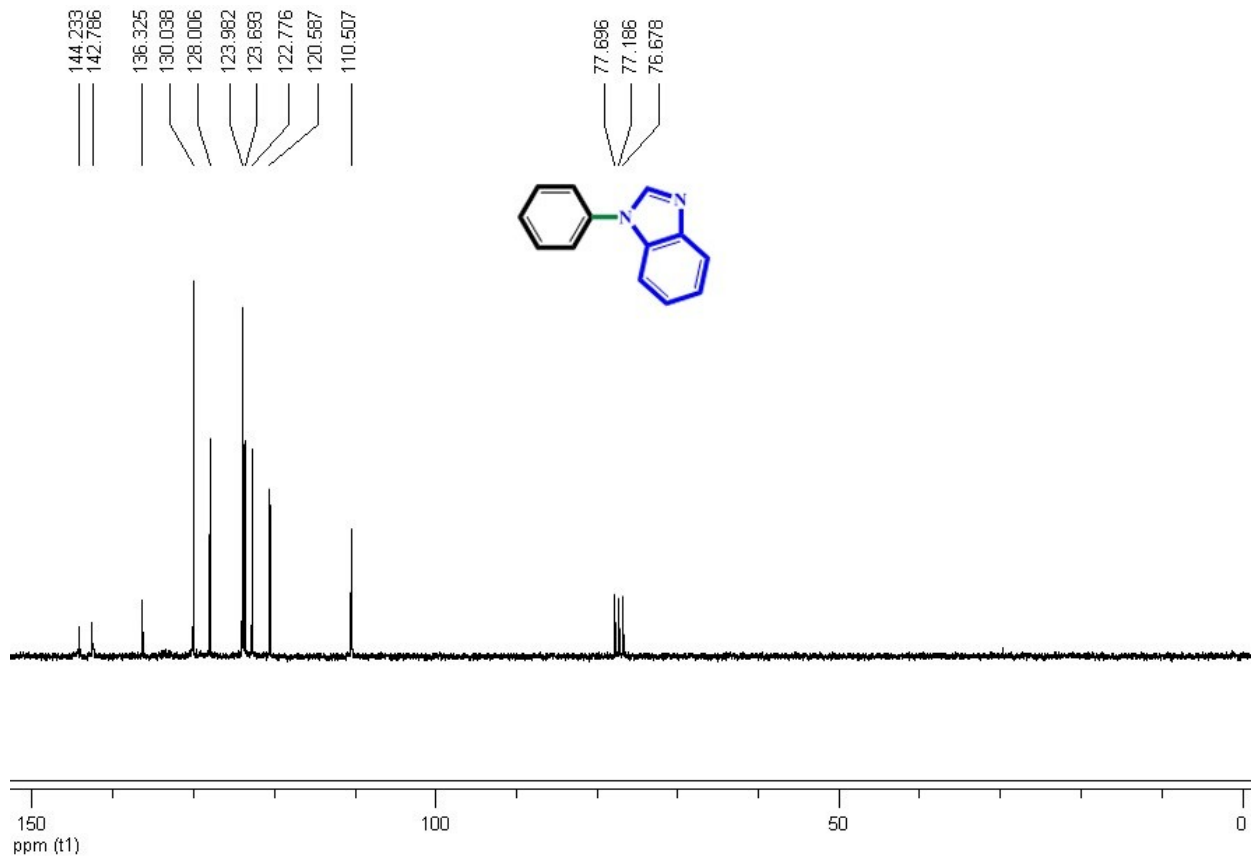




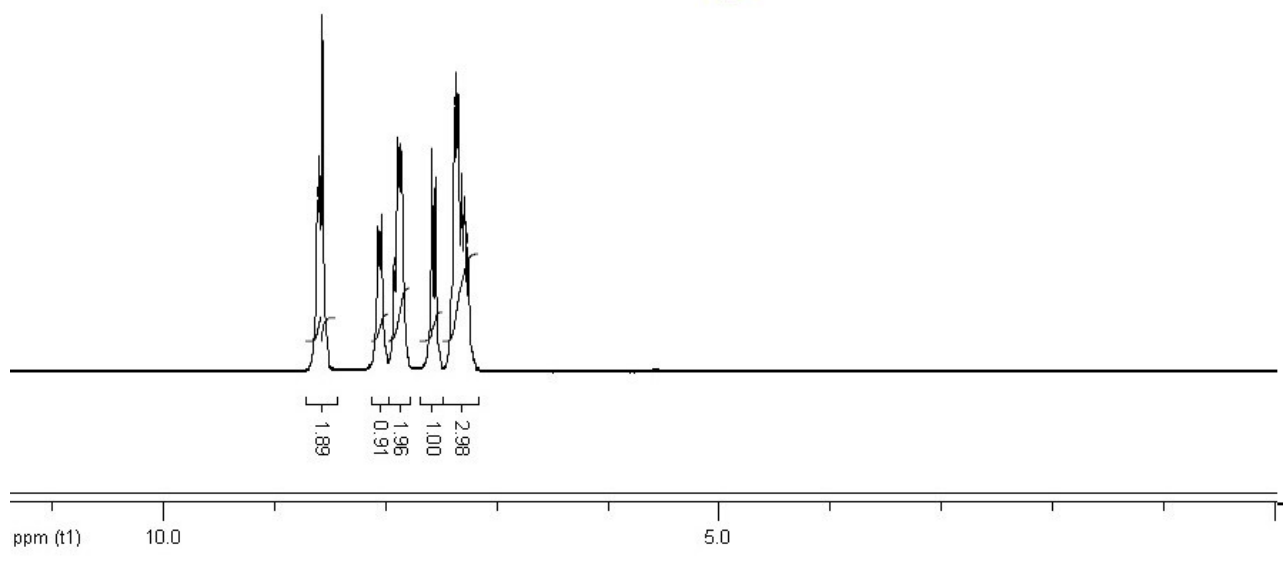
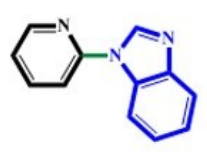


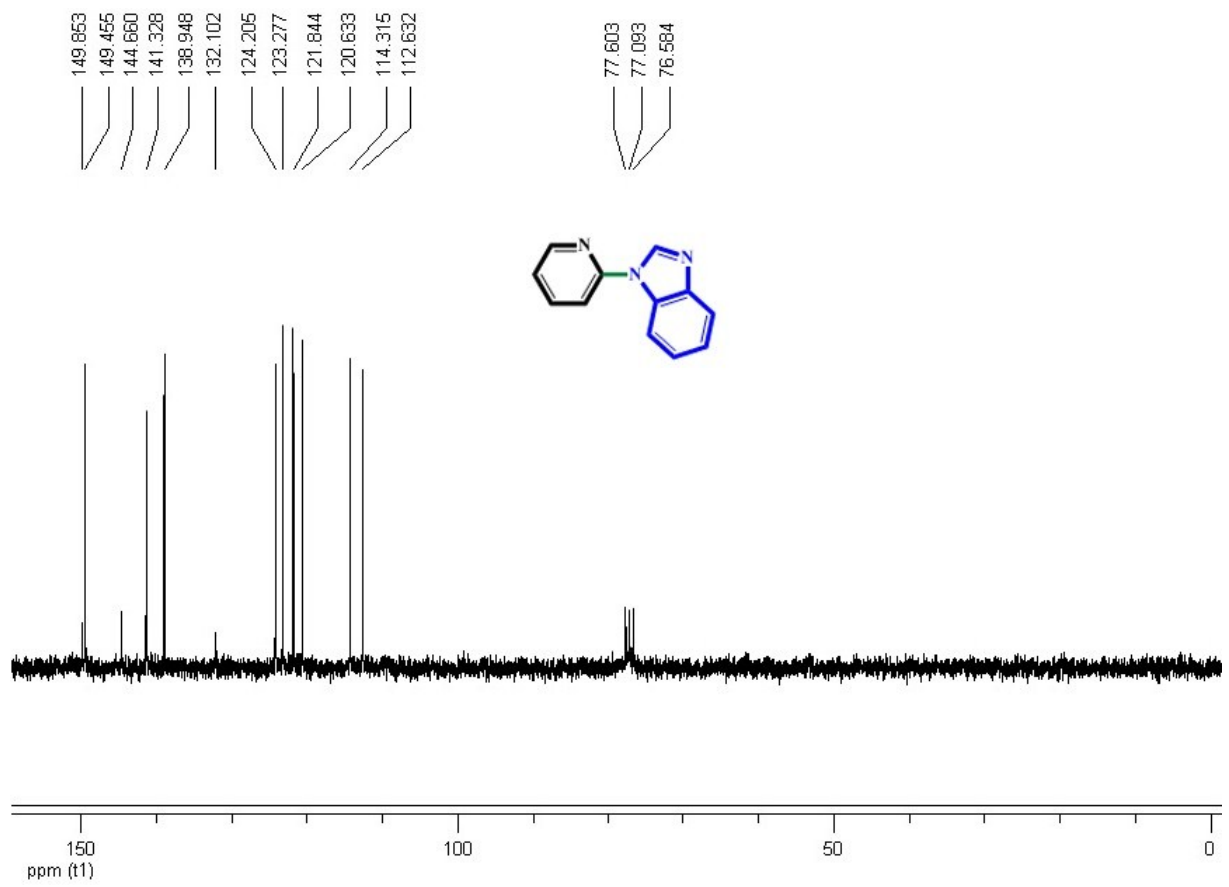




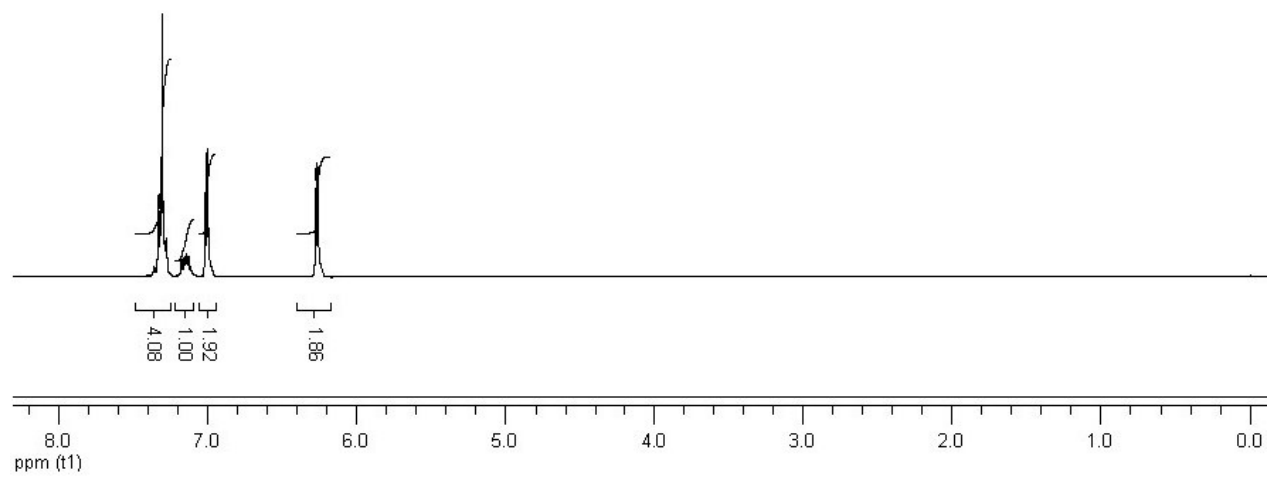
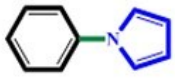


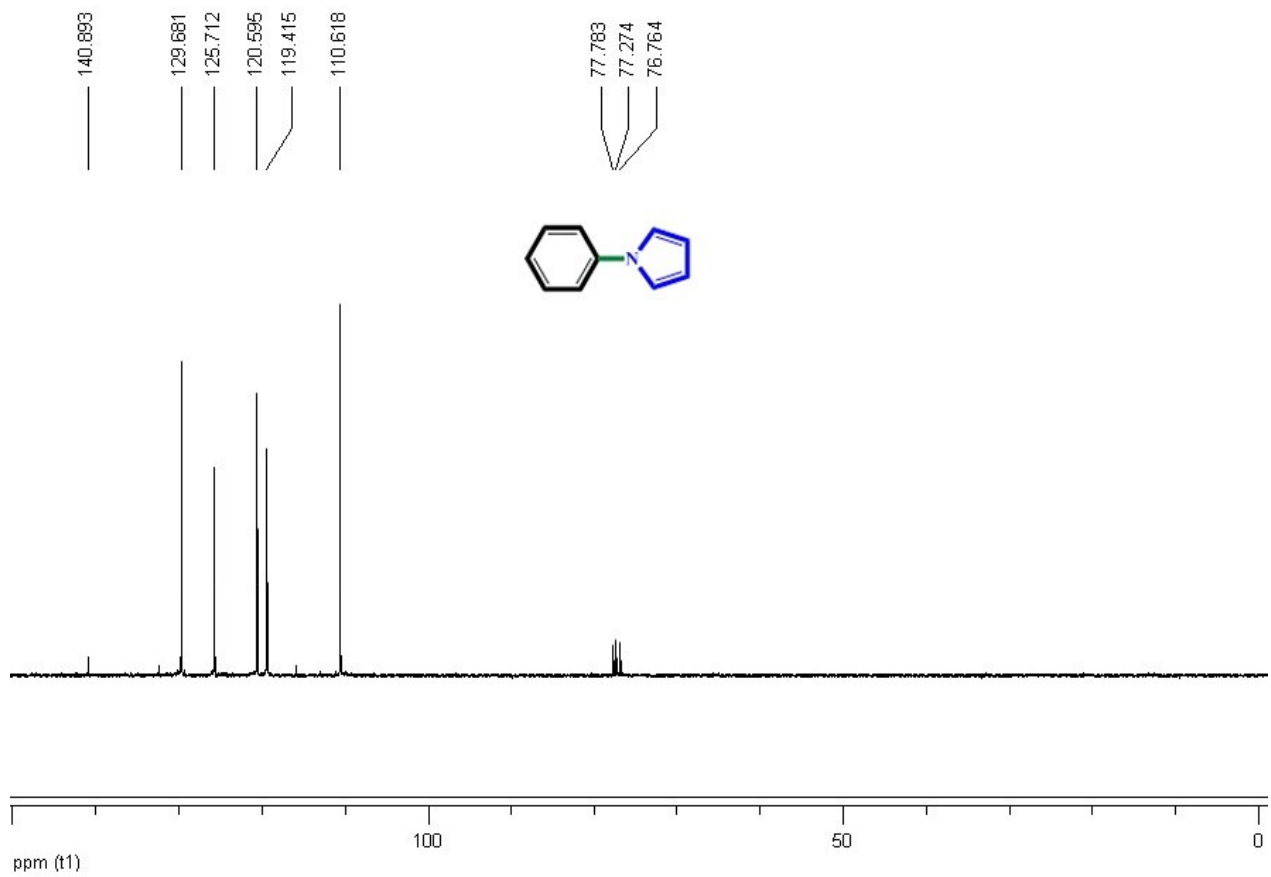
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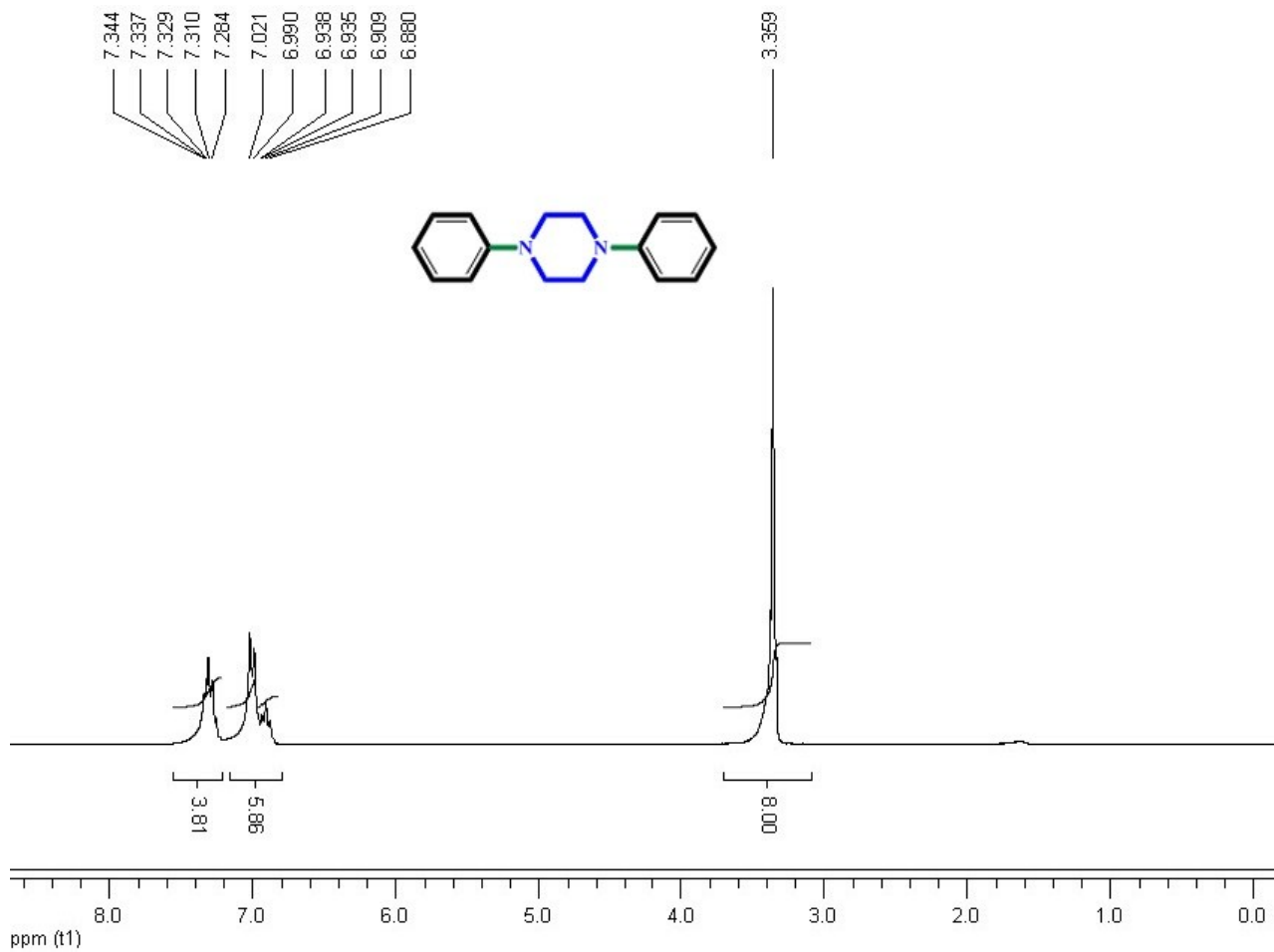


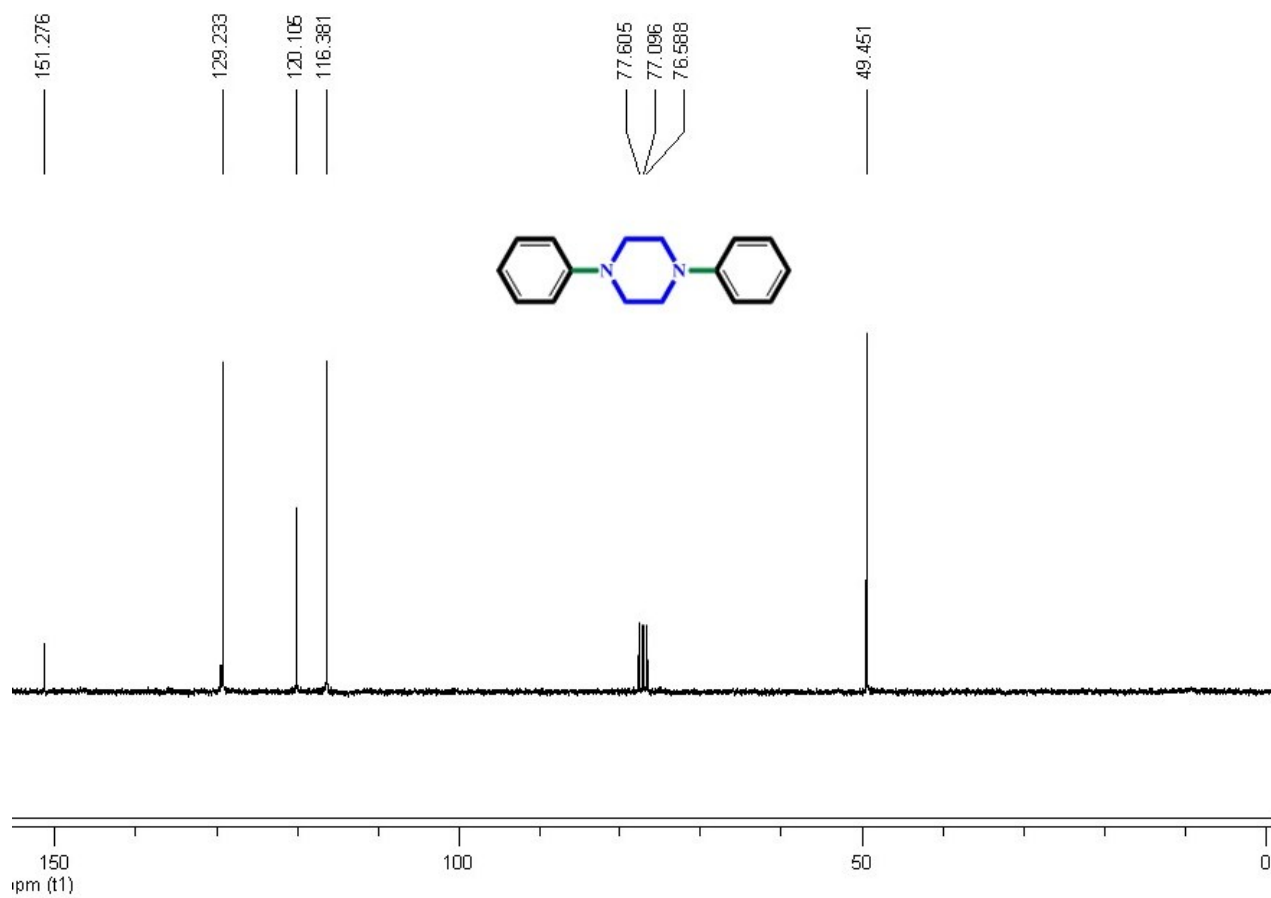


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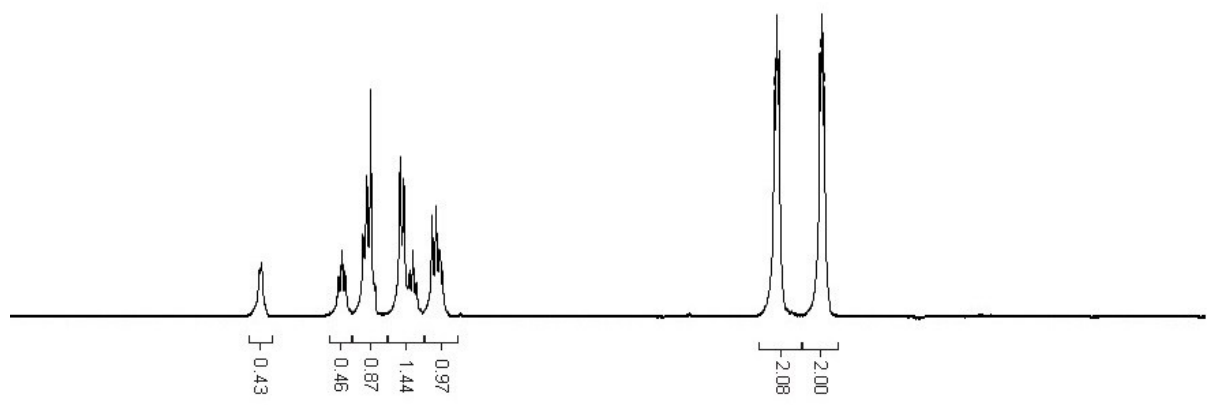
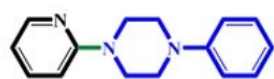








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