

## **Non-metal-mediated *N*-Oxyl radical (TEMPO)-induced acceptorless dehydrogenation of *N*-heterocycles via electrocatalysis**

Huiqing Hou,<sup>†a</sup> Xinhua Ma,<sup>†a</sup> Yaling Ye,<sup>a</sup> Mei Wu,<sup>a</sup> Sunjie Shi,<sup>a</sup> Wenhe Zheng,<sup>b</sup> Mei Lin,<sup>a</sup> Weiming Sun<sup>\*a</sup> and Fang Ke<sup>\*a</sup>

- <sup>a.</sup> *Institute of Materia Medica, School of Pharmacy, Fujian Provincial Key Laboratory of Natural Medicine Pharmacology, Fujian Medical University, Fuzhou 350122, China Tel.: +86-0591-22862016; fax: +86-0591-22862016; E-mail: kefang@mail.fjmu.edu.cn*
- <sup>b.</sup> *The First Affiliated Hospital of Fujian Medical University, Fuzhou 350004, China.*

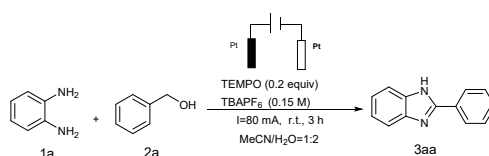
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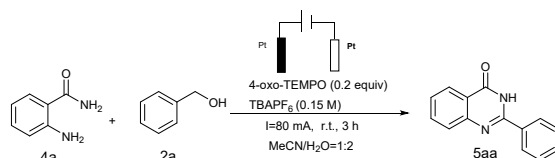
## 1. General information

All reagents were purchased from commercial sources and used without further purification. All solvents were dried in a standard manner. Reactions were monitored by TLC on silica gel plates. Column chromatography was performed over silica gel (200-300 mesh) and petroleum ether/ethyl acetate. Shanghai chenhua CHI600E electrochemical workstation was used in the standard configuration as delivered, including proprietary software. Beijing Perfectlight PCX50C Discover was used in the reaction system. All products were characterized by NMR.  $^1\text{H}$  NMR spectra were recorded at 400 MHz and  $^{13}\text{C}$  NMR spectra were recorded at 101 MHz (Bruker DPX) with  $\text{CDCl}_3$  and  $\text{DMSO-d}_6$  as solvent. Chemical shifts are reported in ppm using TMS as internal standard. NMR by the services provided at the Shandong Liaocheng University. HPLC were recorded on an SHIMADZU LC-20A instrument with a HP5-MS 30 m x 0.25 mm capillary apolar columns.

## 2. General procedure for the catalytic reactions



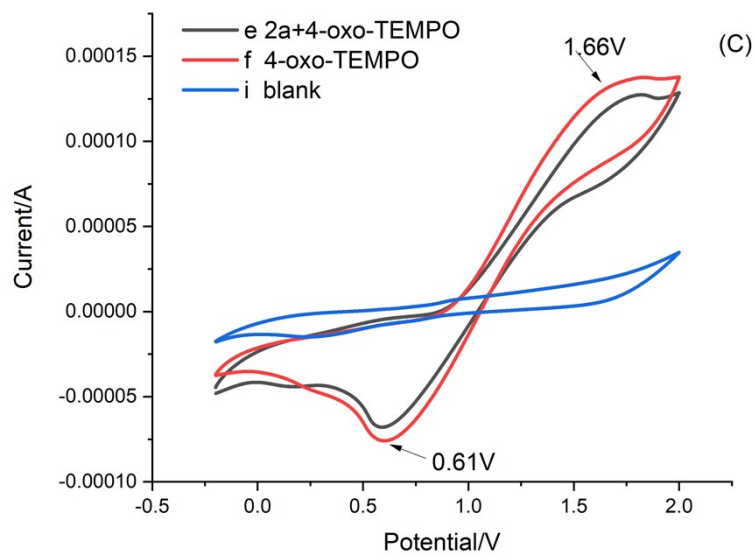
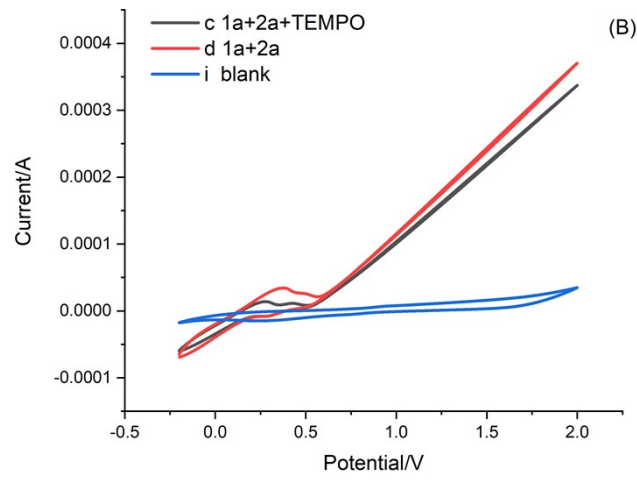
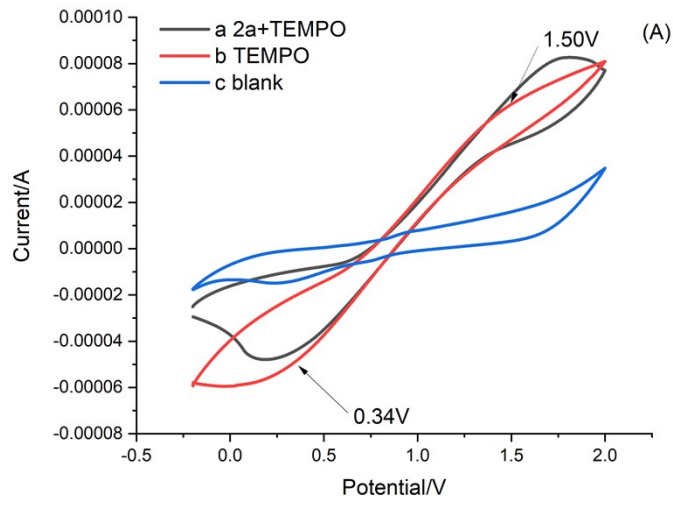
*o*-phenylenediamine **1a** (54.1 mg, 0.5 mmol), benzyl alcohol **2a** (86.5 mg, 0.8 mmol), TEMPO (0.1 mmol), TBAPF<sub>6</sub> (0.15 M), MeCN/H<sub>2</sub>O (v/v=1:2, 3 ml) were added into a 25-mL three-necked flask equipped with a platinum anode and cathode (plate, 1.0 cm × 1.0 cm). The reaction mixture were stirred for 3 h at a constant current of 80 mA at ambient temperature. After completion of the reaction (monitored by TLC), the reaction mixture was purified by column chromatography on silica gel (PE/DCM) to afford pure product **3aa**.



*o*-aminobenzamide **4a** (68.0 mg, 0.5 mmol), benzyl alcohol **2a** (86.5 mg, 0.8 mmol), 4-oxo-TEMPO (0.1 mmol), TBAPF<sub>6</sub> (0.15 M), MeCN/H<sub>2</sub>O (v/v=1:2, 3 ml) were added into a 25-mL three-necked flask equipped with a platinum anode and cathode (plate, 1.0 cm × 1.0 cm). The reaction mixture were stirred for 3 h at a constant current of 80 mA at ambient temperature. After completion of the reaction (monitored by TLC), the reaction mixture was purified by column chromatography on silica gel (PE/DCM) to afford pure product **5aa**.

## 3. Cyclic voltammetry experiment

Cyclic voltammograms were measured using Shanghai chenhua CHI600E electrochemical workstation with electrochemical analysis software, using a conventional three-electrode cell. The working electrode was a Pt disk working electrode, the counter and reference electrodes consisted of a Pt wire and a SCE, respectively. The Pt disk working electrode was polished with a polishing cloth before each measurement. The concentration of all tested compounds was 1 mmol L<sup>-1</sup>. The scan rate was 0.1 V/s.



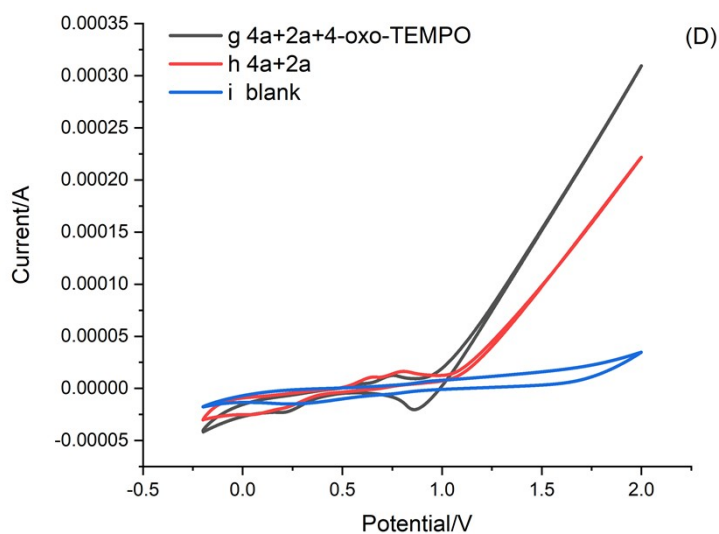
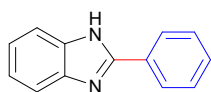


Figure1. Cyclic voltammograms

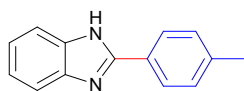
#### 4. Characterization data

##### 2-phenyl-1H-benzo[d]imidazole (3aa)<sup>1</sup>



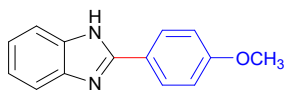
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  13.10 (s, 1H), 8.21 – 8.15 (m, 2H), 7.80 – 7.45 (m, 6H), 7.23 (d,  $J$  = 8.5 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  153.2, 143.0, 130.7, 130.2, 129.5, 127.1, 123.0, 120.5, 118.4, 114.5, 112.1. MS [EI, m/z]: 194 [M<sup>+</sup>].

##### 2-(p-tolyl)-1H-benzo[d]imidazole (3ab)<sup>4</sup>



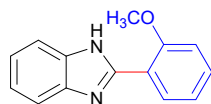
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  12.78 (s, 1H), 8.20 – 8.15 (m, 2H), 7.58 – 7.45 (m, 4H), 7.39 (s, 1H), 7.03 (dd,  $J$  = 8.1, 1.6 Hz, 1H), 2.44 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  151.4, 131.8, 130.8, 130.1, 129.4, 126.8, 124.0, 21.8. MS [EI, m/z]: 208 [M<sup>+</sup>].

##### 2-(4-methoxyphenyl)-1H-benzo[d]imidazole (3ac)<sup>1</sup>



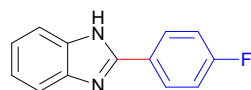
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  12.74 (s, 1H), 8.14 (d,  $J$  = 8.4 Hz, 2H), 7.69 – 7.46 (m, 2H), 7.15 (dd,  $J$  = 24.4, 7.5 Hz, 4H), 3.85 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  161.1, 151.9, 144.4, 135.5, 128.5, 123.2, 122.5, 121.9, 119.0, 114.9, 111.5, 55.8. MS [EI, m/z]: 224 [M<sup>+</sup>].

##### 2-(2-methoxyphenyl)-1H-benzo[d]imidazole (3ad)<sup>5</sup>



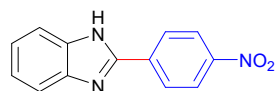
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.52 (s, 1H), 8.17 (dd,  $J = 8.0, 1.6$  Hz, 1H), 7.87 – 7.78 (m, 2H), 7.76 (dt,  $J = 5.0, 1.9$  Hz, 2H), 7.53 (t,  $J = 1.0$  Hz, 1H), 7.46 (t,  $J = 8.0$  Hz, 1H), 7.17 (d,  $J = 1.7$  Hz, 1H), 3.87 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO- $d_6$ )  $\delta$  159.8, 152.5, 149.1, 135.1, 134.5, 130.2, 128.0, 127.1, 126.3, 121.5, 120.6, 118.1, 113.0, 55.9. **MS** [EI,  $m/z$ ]: 224 [ $\text{M}^+$ ].

#### 2-(4-fluorophenyl)-1H-benzo[d]imidazole (3ae)<sup>6</sup>



$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.92 (s, 1H), 8.27 – 8.20 (m, 2H), 7.71 – 7.64 (m, 1H), 7.57 – 7.50 (m, 1H), 7.45 – 7.38 (m, 2H), 7.21 (td,  $J = 7.1, 1.7$  Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  164.8, 162.3, 150.9, 144.2, 135.5, 129.2, 127.3, 123.0, 122.2, 119.3, 116.6, 116.4, 111.8. **MS** [EI,  $m/z$ ]: 212 [ $\text{M}^+$ ].

#### 2-(4-nitrophenyl)-1H-benzo[d]imidazole (3af)<sup>2</sup>



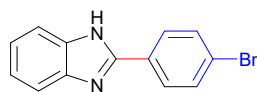
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.29 (s, 1H), 8.47 – 8.36 (m, 4H), 7.73 (d,  $J = 7.5$  Hz, 1H), 7.59 (d,  $J = 7.8$  Hz, 1H), 7.27 (qd,  $J = 7.6, 3.6$  Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  149.5, 148.3, 144.3, 136.5, 135.7, 129.4, 127.9, 124.7, 124.0, 123.7, 122.8, 119.9, 112.3. **MS** [EI,  $m/z$ ]: 239 [ $\text{M}^+$ ].

#### 2-(4-chlorophenyl)-1H-benzo[d]imidazole (3ag)<sup>1</sup>



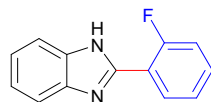
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.99 (s, 1H), 8.20 (d,  $J = 8.2$  Hz, 2H), 7.63 (d,  $J = 8.3$  Hz, 4H), 7.23 (d,  $J = 6.4$  Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  150.6, 144.2, 135.0, 129.5, 128.6, 123.6, 119.5, 113.7. **MS** [EI,  $m/z$ ]: 228 [ $\text{M}^+$ ].

#### 2-(4-bromophenyl)-1H-benzo[d]imidazole (3ah)<sup>3</sup>



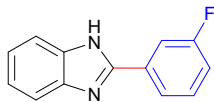
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.09 (d,  $J = 12.4$  Hz, 1H), 8.21 – 8.15 (m, 2H), 7.70 (s, 1H), 7.65 – 7.49 (m, 4H), 7.35 (d,  $J = 7.8$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  153.1, 145.8, 143.4, 130.7, 130.2, 129.5, 127.1, 125.7, 125.2, 121.7, 121.0, 114.4, 113.6. **MS** [EI,  $m/z$ ]: 272 [ $\text{M}^+$ ].

#### 2-(2-fluorophenyl)-1H-benzo[d]imidazole (3ai)<sup>7</sup>



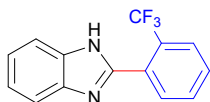
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.58 (s, 1H), 8.26 (td,  $J = 7.8, 1.9$  Hz, 1H), 7.66 (dq,  $J = 6.8, 3.4$  Hz, 2H), 7.57 (dddd,  $J = 8.7, 7.2, 5.3, 1.8$  Hz, 1H), 7.47 – 7.38 (m, 2H), 7.24 (dp,  $J = 8.0, 4.0$  Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  161.2, 158.7, 146.9, 132.4, 130.7, 125.6, 122.8, 118.6, 117.1, 116.9. **MS** [EI,  $m/z$ ]: 212 [ $\text{M}^+$ ].

#### 2-(3-fluorophenyl)-1H-benzo[d]imidazole (3aj)<sup>7</sup>



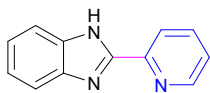
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.11 (s, 1H), 8.07 (d,  $J$  = 7.8 Hz, 1H), 8.03 – 7.99 (m, 1H), 7.64 (dt,  $J$  = 6.0, 2.9 Hz, 2H), 7.59 (dd,  $J$  = 8.0, 6.0 Hz, 1H), 7.33 (td,  $J$  = 8.6, 2.6 Hz, 1H), 7.24 (dt,  $J$  = 7.2, 3.6 Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  164.17, 161.75, 150.50, 150.47, 133.05, 132.97, 131.6, 131.5, 123.0, 122.9, 117.1, 116.9, 113.7, 113.4. **MS** [EI,  $m/z$ ]: 212 [ $\text{M}^+$ ].

**2-(2-(trifluoromethyl)phenyl)-1H-benzo[d]imidazole (3ak)<sup>8</sup>**



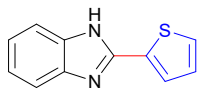
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.81 (s, 1H), 8.10 – 8.04 (m, 2H), 7.67 – 7.60 (m, 1H), 7.51 (d,  $J$  = 7.4 Hz, 1H), 7.39 – 7.33 (m, 2H), 7.19 (dd,  $J$  = 7.7, 4.3 Hz, 2H), 2.38 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  151.3, 139.5, 129.5, 127.4, 126.4, 122.3, 118.7, 111.1, 20.9. **MS** [EI,  $m/z$ ]: 262 [ $\text{M}^+$ ].

**2-(pyridin-2-yl)-1H-benzo[d]imidazole (3al)<sup>3</sup>**



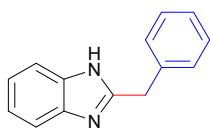
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.09 (s, 1H), 9.37 (dd,  $J$  = 2.4, 0.8 Hz, 1H), 8.69 (dd,  $J$  = 4.8, 1.7 Hz, 1H), 8.51 (dt,  $J$  = 8.0, 2.0 Hz, 1H), 7.72 (d,  $J$  = 7.7 Hz, 1H), 7.62 – 7.54 (m, 2H), 7.24 (h,  $J$  = 7.0, 6.5 Hz, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  151.0, 149.3, 148.0, 144.2, 135.5, 134.2, 126.7, 124.5, 123.5, 122.4, 119.57, 112.0. **MS** [EI,  $m/z$ ]: 195 [ $\text{M}^+$ ].

**2-(thiophen-2-yl)-1H-benzo[d]imidazole (3am)<sup>1</sup>**



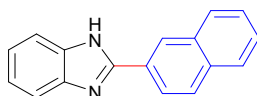
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.93 (s, 1H), 7.84 (dd,  $J$  = 3.7, 1.2 Hz, 1H), 7.72 (dd,  $J$  = 5.0, 1.2 Hz, 1H), 7.62 (d,  $J$  = 7.5 Hz, 1H), 7.51 (d,  $J$  = 7.6 Hz, 1H), 7.27 – 7.15 (m, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  147.5, 144.1, 135.2, 134.2, 129.2, 128.7, 127.2, 123.1, 122.2, 119.0, 111.6. **MS** [EI,  $m/z$ ]: 200 [ $\text{M}^+$ ].

**2-benzyl-1H-benzo[d]imidazole (3an)<sup>2</sup>**



$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.27 (s, 1H), 7.59 – 7.38 (m, 2H), 7.36 – 7.29 (m, 4H), 7.23 (ddd,  $J$  = 8.6, 5.3, 2.4 Hz, 1H), 7.15 – 7.08 (m, 2H), 4.17 (s, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  154.0, 138.1, 129.2, 128.9, 127.0, 121.7, 35.4. **MS** [EI,  $m/z$ ]: 208 [ $\text{M}^+$ ].

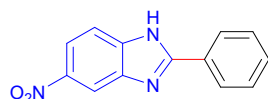
**2-(naphthalen-2-yl)-1H-benzo[d]imidazole (3ao)<sup>3</sup>**



$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.08 (s, 1H), 8.75 (d,  $J$  = 1.6 Hz, 1H), 8.33 (dd,  $J$  = 8.6, 1.8 Hz, 1H), 8.11 – 8.04 (m, 2H), 8.01 – 7.98 (m, 1H), 7.68 (s, 1H), 7.64 – 7.58 (m, 3H), 7.24 (dt,  $J$  = 6.6, 3.2 Hz,

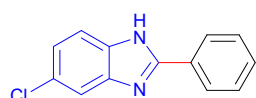
2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  151.7, 133.9, 133.3, 129.0, 128.9, 128.3, 128.1, 127.6, 127.4, 126.3, 124.4, 119.6. MS [EI, m/z]: 244 [ $\text{M}^+$ ].

**5-nitro-2-phenyl-1H-benzo[d]imidazole (3ap)<sup>9</sup>**



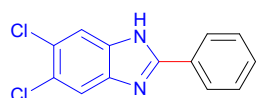
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.55 (s, 1H), 8.51 (s, 1H), 8.24 – 8.17 (m, 2H), 8.14 – 8.06 (m, 1H), 7.74 (d,  $J = 18.9$  Hz, 1H), 7.64 – 7.52 (m, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  155.9, 143.2, 140.3, 131.4, 129.6, 129.5, 127.5, 118.6, 115.3, 112.2, 108.4. MS [EI, m/z]: 239 [ $\text{M}^+$ ].

**5-chloro-2-phenyl-1H-benzo[d]imidazole (3aq)<sup>5</sup>**



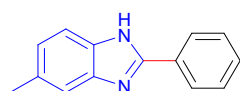
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.09 (s, 1H), 8.22 – 8.13 (m, 2H), 7.70 (d,  $J = 20.4$  Hz, 1H), 7.62 – 7.46 (m, 4H), 7.23 (d,  $J = 8.3$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  153.1, 134.2, 130.7, 130.2, 129.5, 127.1, 123.2, 120.6, 118.8, 112.9, 111.5. MS [EI, m/z]: 228 [ $\text{M}^+$ ].

**5,6-dichloro-2-phenyl-1H-benzo[d]imidazole (3ar)<sup>4</sup>**



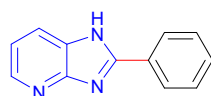
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.24 (s, 1H), 8.17 (d,  $J = 7.3$  Hz, 2H), 7.84 (s, 2H), 7.56 (q,  $J = 8.0$ , 7.5 Hz, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO- $d_6$ )  $\delta$  154.3, 131.0, 129.8, 129.5, 127.2, 125.0, 120.5, 113.6. MS [EI, m/z]: 262 [ $\text{M}^+$ ].

**5-methyl-2-phenyl-1H-benzo[d]imidazole (3as)<sup>2</sup>**



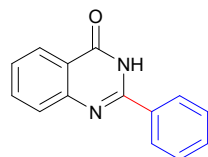
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.75 (d,  $J = 13.5$  Hz, 1H), 8.20 – 8.14 (m, 2H), 7.57 – 7.45 (m, 4H), 7.33 (s, 1H), 7.03 (s, 1H), 2.44 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  151.6, 142.5, 135.7, 132.3, 130.8, 130.1, 129.4, 126.8, 124.4, 123.7, 119.1, 111.5, 21.8. MS [EI, m/z]: 208 [ $\text{M}^+$ ].

**2-phenyl-1H-imidazo[4,5-b]pyridine (3at)<sup>10</sup>**



$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  13.55 (s, 1H), 8.34 (s, 1H), 8.25 (d,  $J = 7.0$  Hz, 2H), 8.06 (s, 1H), 7.63 – 7.50 (m, 3H), 7.25 (dd,  $J = 8.0, 4.8$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  152.9, 149.9, 144.3, 131.0, 130.2, 129.5, 127.2, 126.8, 119.6, 118.6. MS [EI, m/z]: 195 [ $\text{M}^+$ ].

**2-phenylquinazolin-4(3H)-one (5aa)<sup>11</sup>**

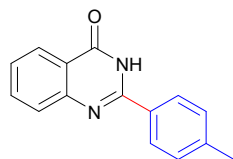


$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.49 (s, 1H), 8.17 (t,  $J = 8.1$  Hz, 3H), 7.82 (t,  $J = 7.6$  Hz, 1H), 7.74 (d,  $J = 8.1$  Hz, 1H), 7.55 (q,  $J = 11.6, 10.7$  Hz, 4H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.8, 152.8, 149.2,



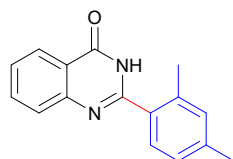
135.0, 133.2, 131.8, 129.1, 128.2, 127.9, 127.9, 127.0, 126.3, 121.4. **MS** [EI, m/z]: 222 [M<sup>+</sup>].

**2-(p-tolyl)quinazolin-4(3H)-one (5ab)<sup>11</sup>**



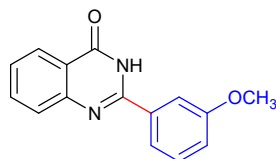
<sup>1</sup>H NMR (400 MHz, DMSO) δ 12.44 (s, 1H), 8.15 (dd, *J* = 8.0, 1.6 Hz, 1H), 8.12 – 8.08 (m, 2H), 7.83 (ddd, *J* = 8.5, 7.1, 1.6 Hz, 1H), 7.73 (dd, *J* = 8.3, 1.6 Hz, 1H), 7.51 (ddd, *J* = 8.1, 7.1, 1.3 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 2H), 2.40 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO) δ 162.7, 152.7, 149.3, 141.9, 135.1, 130.4, 129.7, 128.2, 127.9, 126.9, 126.3, 121.4, 21.5. **MS** [EI, m/z]: 236 [M<sup>+</sup>].

**2-(2,4-dimethylphenyl)quinazolin-4(3H)-one (5ac)<sup>12</sup>**



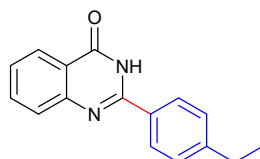
<sup>1</sup>H NMR (400 MHz, DMSO) δ 12.36 (s, 1H), 8.17 (dd, *J* = 7.9, 1.7 Hz, 1H), 7.86 – 7.80 (m, 1H), 7.68 (d, *J* = 7.1 Hz, 1H), 7.56 – 7.50 (m, 1H), 7.41 (d, *J* = 7.8 Hz, 1H), 7.15 (d, *J* = 13.9 Hz, 2H), 2.37 (s, 3H), 2.35 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO) δ 162.3, 154.9, 149.3, 139.9, 136.5, 134.9, 131.9, 131.7, 129.6, 127.8, 127.0, 126.7, 126.2, 121.4, 21.3, 20.1. **MS** [EI, m/z]: 250 [M<sup>+</sup>].

**2-(3-methoxyphenyl)quinazolin-4(3H)-one (5ad)<sup>13</sup>**



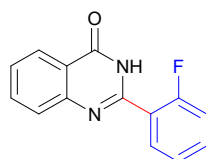
<sup>1</sup>H NMR (400 MHz, DMSO) δ 12.52 (s, 1H), 8.17 (dd, *J* = 7.9, 1.6 Hz, 1H), 7.87 – 7.73 (m, 4H), 7.56 – 7.50 (m, 1H), 7.47 (t, *J* = 8.0 Hz, 1H), 7.16 (ddd, *J* = 8.3, 2.6, 0.9 Hz, 1H), 3.87 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO) δ 162.7, 159.8, 152.5, 149.2, 135.1, 134.5, 130.2, 128.0, 127.1, 126.3, 121.5, 120.6, 118.1, 113.0, 55.9. **MS** [EI, m/z]: 252 [M<sup>+</sup>].

**2-(4-ethylphenyl)quinazolin-4(3H)-one (5ae)<sup>14</sup>**



<sup>1</sup>H NMR (400 MHz, DMSO) δ 12.45 (s, 1H), 8.17 – 8.11 (m, 3H), 7.84 (ddd, *J* = 8.5, 7.1, 1.6 Hz, 1H), 7.73 (dt, *J* = 7.9, 1.0 Hz, 1H), 7.52 (ddd, *J* = 8.1, 7.0, 1.2 Hz, 1H), 7.42 – 7.37 (m, 2H), 2.70 (q, *J* = 7.6 Hz, 2H), 1.23 (t, *J* = 7.6 Hz, 3H). <sup>13</sup>C NMR (101 MHz, DMSO) δ 162.7, 152.7, 149.3, 148.1, 135.1, 130.6, 128.5, 128.3, 127.9, 126.9, 126.3, 121.4, 28.5, 15.7. **MS** [EI, m/z]: 250 [M<sup>+</sup>].

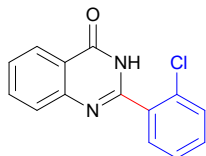
**2-(2-fluorophenyl)quinazolin-4(3H)-one (5af)<sup>15</sup>**



<sup>1</sup>H NMR (400 MHz, DMSO) δ 12.55 (s, 1H), 8.18 (dd, *J* = 8.0, 1.6 Hz, 1H), 7.86 (ddd, *J* = 8.5, 7.1, 1.6 Hz, 1H), 7.79 (td, *J* = 7.6, 1.8 Hz, 1H), 7.74 (dd, *J* = 8.3, 1.4 Hz, 1H), 7.66 – 7.55 (m, 2H), 7.43 – 7.35

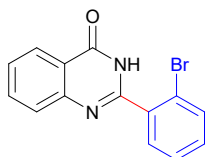
(m, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.0, 161.3, 158.8, 150.4, 149.1, 135.1, 133.4, 133.3, 131.5, 131.5, 128.0, 127.5, 126.3, 125.1, 125.1, 122.8, 122.7, 121.6, 116.8, 116.5. **MS** [EI, m/z]: 240 [ $\text{M}^+$ ].

**2-(2-chlorophenyl)quinazolin-4(3H)-one (5ag)<sup>15</sup>**



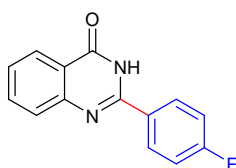
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.82 (s, 1H), 8.12 (d,  $J = 2.5$  Hz, 1H), 7.89 (dd,  $J = 8.8, 2.5$  Hz, 1H), 7.75 (d,  $J = 8.6$  Hz, 1H), 7.71 – 7.55 (m, 4H), 7.51 (td,  $J = 7.4, 1.5$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  160.9, 153.2, 147.8, 135.2, 134.0, 132.2, 131.9, 131.8, 131.4, 130.2, 130.1, 127.7, 125.4, 123.0. **MS** [EI, m/z]: 256 [ $\text{M}^+$ ].

**2-(2-bromophenyl)quinazolin-4(3H)-one (5ah)<sup>15</sup>**



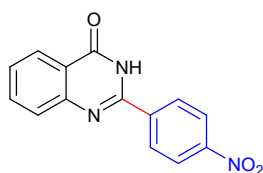
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.60 (s, 1H), 8.19 (dd,  $J = 8.0, 1.6$  Hz, 1H), 7.86 (ddd,  $J = 8.5, 7.1, 1.6$  Hz, 1H), 7.78 (dd,  $J = 7.9, 1.4$  Hz, 1H), 7.72 (d,  $J = 7.1$  Hz, 1H), 7.65 (dd,  $J = 7.5, 1.9$  Hz, 1H), 7.53 (dq,  $J = 23.3, 7.9, 1.6$  Hz, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  161.9, 153.8, 149.0, 136.4, 135.1, 133.1, 132.1, 131.2, 128.1, 127.9, 127.5, 126.3, 121.8, 121.4. **MS** [EI, m/z]: 300 [ $\text{M}^+$ ].

**2-(4-fluorophenyl)quinazolin-4(3H)-one (5ai)<sup>15</sup>**



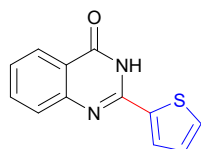
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.55 (s, 1H), 8.29 – 8.23 (m, 2H), 8.16 (dd,  $J = 7.9, 1.5$  Hz, 1H), 7.84 (ddd,  $J = 8.5, 7.1, 1.6$  Hz, 1H), 7.74 (dd,  $J = 8.2, 1.1$  Hz, 1H), 7.53 (ddd,  $J = 8.1, 7.0, 1.2$  Hz, 1H), 7.43 – 7.36 (m, 2H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  165.8, 163.3, 162.8, 152.0, 149.1, 135.1, 130.9, 130.8, 129.8, 127.8, 127.1, 126.3, 121.4, 116.2, 116.0. **MS** [EI, m/z]: 240 [ $\text{M}^+$ ].

**2-(4-nitrophenyl)quinazolin-4(3H)-one (5aj)<sup>16</sup>**



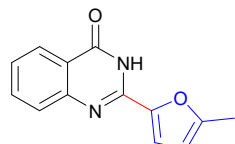
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  8.40 (dd,  $J = 9.1, 6.4$  Hz, 4H), 8.19 (dd,  $J = 7.9, 1.6$  Hz, 1H), 7.92 – 7.86 (m, 1H), 7.81 (dd,  $J = 8.2, 1.2$  Hz, 1H), 7.59 (ddd,  $J = 8.1, 7.0, 1.3$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.5, 151.3, 149.4, 139.0, 135.2, 129.7, 128.2, 127.8, 126.3, 124.1, 121.6. **MS** [EI, m/z]: 267 [ $\text{M}^+$ ].

**2-(thiophen-2-yl)quinazolin-4(3H)-one (5ak)<sup>17</sup>**



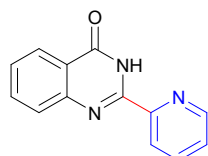
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.44 (s, 1H), 8.60 (dd,  $J = 2.9, 1.4$  Hz, 1H), 8.14 (dd,  $J = 7.9, 1.7$  Hz, 1H), 7.88 (dd,  $J = 5.1, 1.4$  Hz, 1H), 7.84 – 7.78 (m, 1H), 7.73 – 7.67 (m, 2H), 7.52 – 7.47 (m, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.5, 149.4, 148.8, 135.9, 135.0, 129.1, 127.8, 127.7, 127.5, 126.8, 126.3, 121.5. **MS** [EI,  $m/z$ ]: 228 [ $\text{M}^+$ ].

#### 2-(5-methylfuran-2-yl)quinazolin-4(3H)-one (5al)<sup>18</sup>



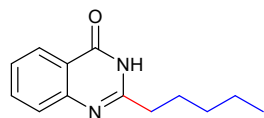
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.35 (s, 1H), 8.11 (d,  $J = 6.3$  Hz, 1H), 7.83 – 7.78 (m, 1H), 7.69 (d,  $J = 8.3$  Hz, 1H), 7.55 (d,  $J = 3.4$  Hz, 1H), 7.48 (t,  $J = 7.5$  Hz, 1H), 6.38 (d,  $J = 3.5$  Hz, 1H), 2.42 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.1, 156.6, 149.3, 144.9, 144.4, 135.1, 127.6, 126.7, 126.4, 121.5, 116.3, 109.4, 14.0. **MS** [EI,  $m/z$ ]: 226 [ $\text{M}^+$ ].

#### 2-(pyridin-2-yl)quinazolin-4(3H)-one (5am)<sup>11</sup>



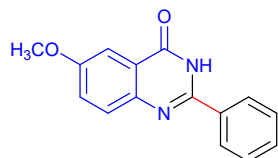
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  11.56 (s, 1H), 8.74 – 8.70 (m, 1H), 8.47 (d,  $J = 8.0$  Hz, 1H), 8.18 (dd,  $J = 7.9, 1.7$  Hz, 1H), 8.02 (t,  $J = 7.8$  Hz, 1H), 7.84 – 7.75 (m, 2H), 7.60 (ddd,  $J = 7.7, 4.7, 1.4$  Hz, 1H), 7.52 (tt,  $J = 8.0, 1.3$  Hz, 1H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  161.3, 150.1, 149.2, 149.0, 148.8, 138.2, 134.9, 128.0, 127.5, 126.8, 126.5, 122.5. **MS** [EI,  $m/z$ ]: 223 [ $\text{M}^+$ ].

#### 2-pentylquinazolin-4(3H)-one (5an)<sup>12</sup>



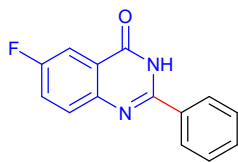
$^1\text{H}$  NMR (400 MHz, DMSO)  $\delta$  12.14 (s, 1H), 8.08 (dd,  $J = 8.0, 1.6$  Hz, 1H), 7.77 (ddd,  $J = 8.4, 7.1, 1.6$  Hz, 1H), 7.61 – 7.57 (m, 1H), 7.45 (ddd,  $J = 8.2, 7.1, 1.2$  Hz, 1H), 2.62 – 2.57 (m, 2H), 1.77 – 1.69 (m, 2H), 1.31 (ddt,  $J = 6.3, 4.9, 2.3$  Hz, 4H), 0.88 (td,  $J = 7.3, 1.8$  Hz, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO)  $\delta$  162.3, 158.0, 149.4, 134.7, 127.3, 126.4, 126.1, 121.2, 34.9, 31.2, 26.9, 22.3, 14.3. **MS** [EI,  $m/z$ ]: 216 [ $\text{M}^+$ ].

#### 6-methoxy-2-phenylquinazolin-4(3H)-one (5ao)<sup>19</sup>



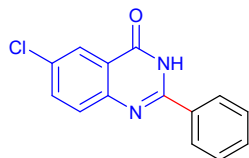
$^1\text{H}$  NMR (400 MHz, DMSO+ $\text{CDCl}_3$ )  $\delta$  12.34 – 12.18 (m, 1H), 8.16 – 8.11 (m, 2H), 7.87 (d,  $J = 6.1$  Hz, 1H), 7.63 (dd,  $J = 8.9, 5.6$  Hz, 1H), 7.55 (t,  $J = 3.0$  Hz, 1H), 7.46 (td,  $J = 5.4, 5.0, 1.9$  Hz, 3H), 7.31 (dt,  $J = 8.9, 2.9$  Hz, 1H), 3.87 (d,  $J = 5.7$  Hz, 3H).  $^{13}\text{C}$  NMR (101 MHz, DMSO+ $\text{CDCl}_3$ )  $\delta$  162.8, 158.1, 150.4, 143.8, 133.3, 131.0, 129.4, 128.7, 127.7, 124.3, 122.2, 106.0, 55.8. **MS** [EI,  $m/z$ ]: 252 [ $\text{M}^+$ ].

**6-fluoro-2-phenylquinazolin-4(3H)-one (5ap)**<sup>20</sup>



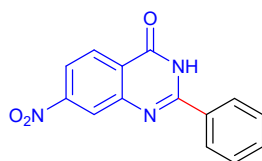
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  12.60 (s, 1H), 8.20 – 8.16 (m, 2H), 7.80 (dt,  $J$  = 8.7, 4.1 Hz, 2H), 7.68 – 7.63 (m, 1H), 7.58 – 7.49 (m, 3H). <sup>13</sup>C NMR (101 MHz, DMSO+CDCl<sub>3</sub>)  $\delta$  162.2, 161.7, 159.2, 146.1, 133.1, 131.7, 130.7, 130.6, 128.9, 128.2, 123.4, 123.2, 122.7, 122.7, 111.0, 110.8. **MS** [EI,  $m/z$ ]: 240 [M<sup>+</sup>].

**6-chloro-2-phenylquinazolin-4(3H)-one (5aq)**<sup>20</sup>



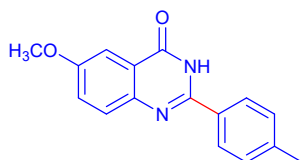
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  12.69 (s, 1H), 8.18 (dt,  $J$  = 8.0, 1.4 Hz, 2H), 8.10 (d,  $J$  = 2.5 Hz, 1H), 7.87 (ddd,  $J$  = 8.7, 2.5, 0.9 Hz, 1H), 7.77 (d,  $J$  = 8.7 Hz, 1H), 7.63 – 7.52 (m, 3H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  161.8, 153.4, 148.0, 135.2, 132.9, 132.1, 131.3, 130.2, 129.1, 128.3, 125.4, 122.7. **MS** [EI,  $m/z$ ]: 256 [M<sup>+</sup>].

**7-nitro-2-phenylquinazolin-4(3H)-one (5ar)**<sup>21</sup>



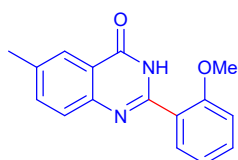
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  8.44 (d,  $J$  = 2.2 Hz, 1H), 8.37 (d,  $J$  = 8.7 Hz, 1H), 8.22 (ddd,  $J$  = 8.7, 5.9, 2.0 Hz, 3H), 7.65 – 7.56 (m, 3H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  161.9, 155.1, 151.8, 149.9, 132.6, 132.5, 129.2, 128.7, 128.5, 125.8, 122.8, 120.51. **MS** [EI,  $m/z$ ]: 267 [M<sup>+</sup>].

**6-methoxy-2-(p-tolyl)quinazolin-4(3H)-one (5as)**<sup>22</sup>



<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  7.80 – 7.75 (m, 2H), 7.51 (d,  $J$  = 8.7 Hz, 1H), 7.25 (d,  $J$  = 7.9 Hz, 2H), 6.98 – 6.90 (m, 2H), 3.78 (s, 3H), 2.35 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  169.2, 168.3, 158.8, 141.5, 140.5, 134.1, 132.0, 129.2, 128.0, 117.0, 114.6, 109.3, 56.1, 21.4. **MS** [EI,  $m/z$ ]: 266 [M<sup>+</sup>].

**2-(2-methoxyphenyl)-6-methylquinazolin-4(3H)-one (5at)**<sup>22</sup>



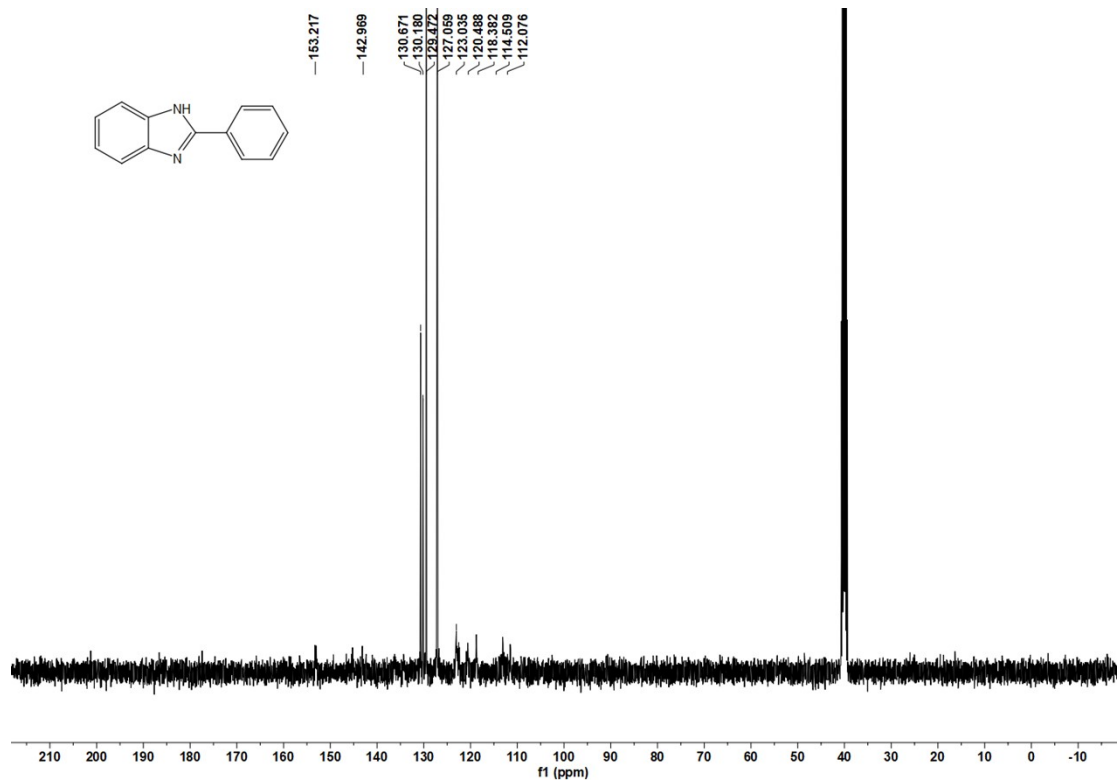
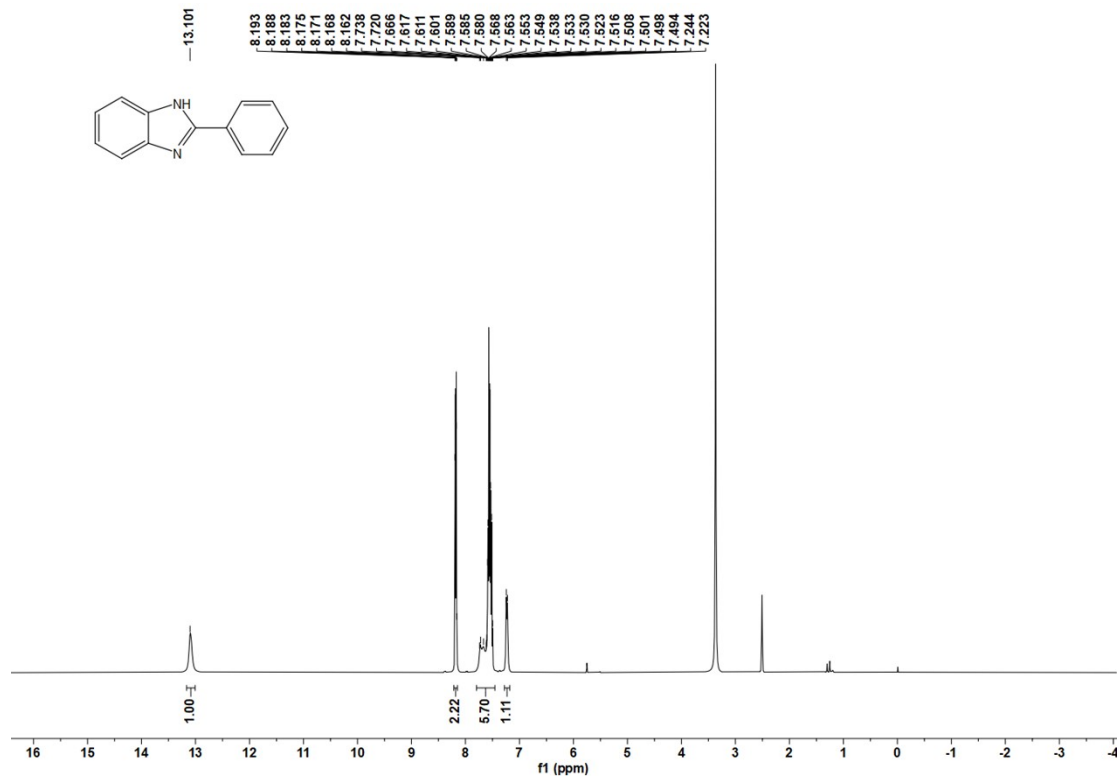
<sup>1</sup>H NMR (400 MHz, DMSO)  $\delta$  12.49 (s, 1H), 8.01 (s, 1H), 7.85 – 7.78 (m, 2H), 7.71 (d,  $J$  = 1.6 Hz, 2H), 7.50 (t,  $J$  = 8.0 Hz, 1H), 7.19 (dd,  $J$  = 8.3, 3.6 Hz, 1H), 3.92 (s, 3H), 2.52 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO)  $\delta$  161.6, 158.8, 150.7, 146.1, 135.8, 135.4, 133.5, 129.2, 126.9, 124.7, 120.2, 119.5, 116.9, 111.9, 54.8, 35.0, 20.3. **MS** [EI,  $m/z$ ]: 266 [M<sup>+</sup>].

## 5. References

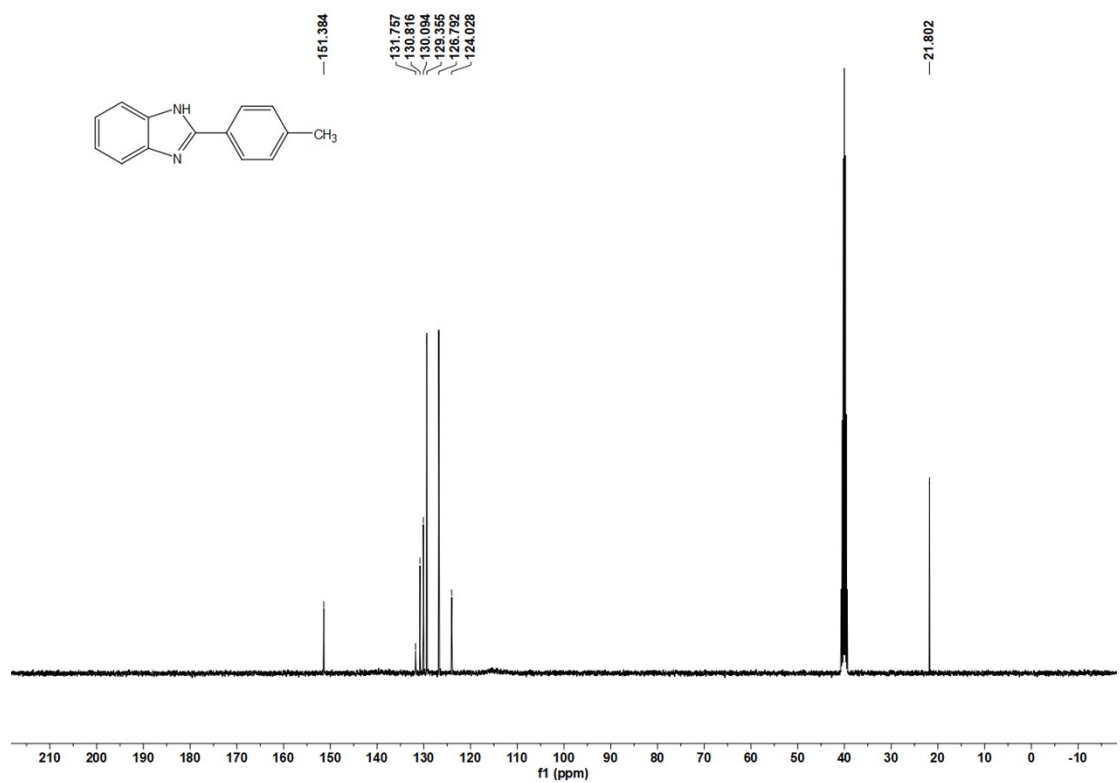
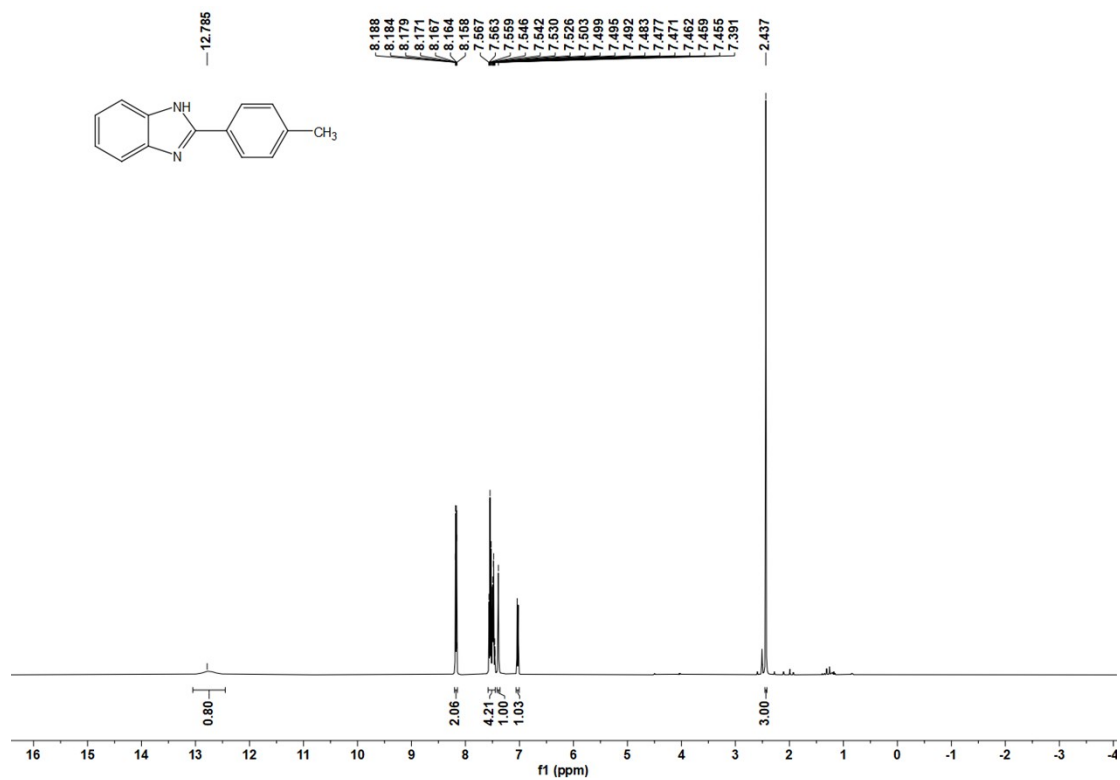
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## 6. $^1\text{H}$ NMR and $^{13}\text{C}$ NMR spectra for the products

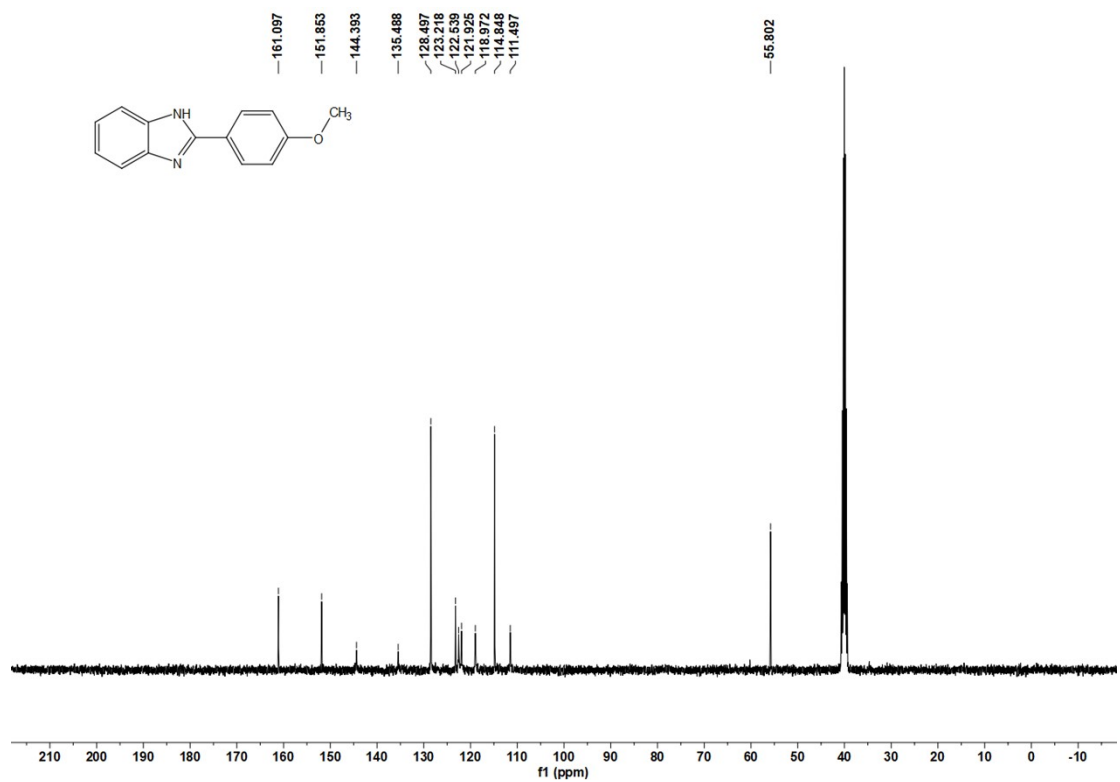
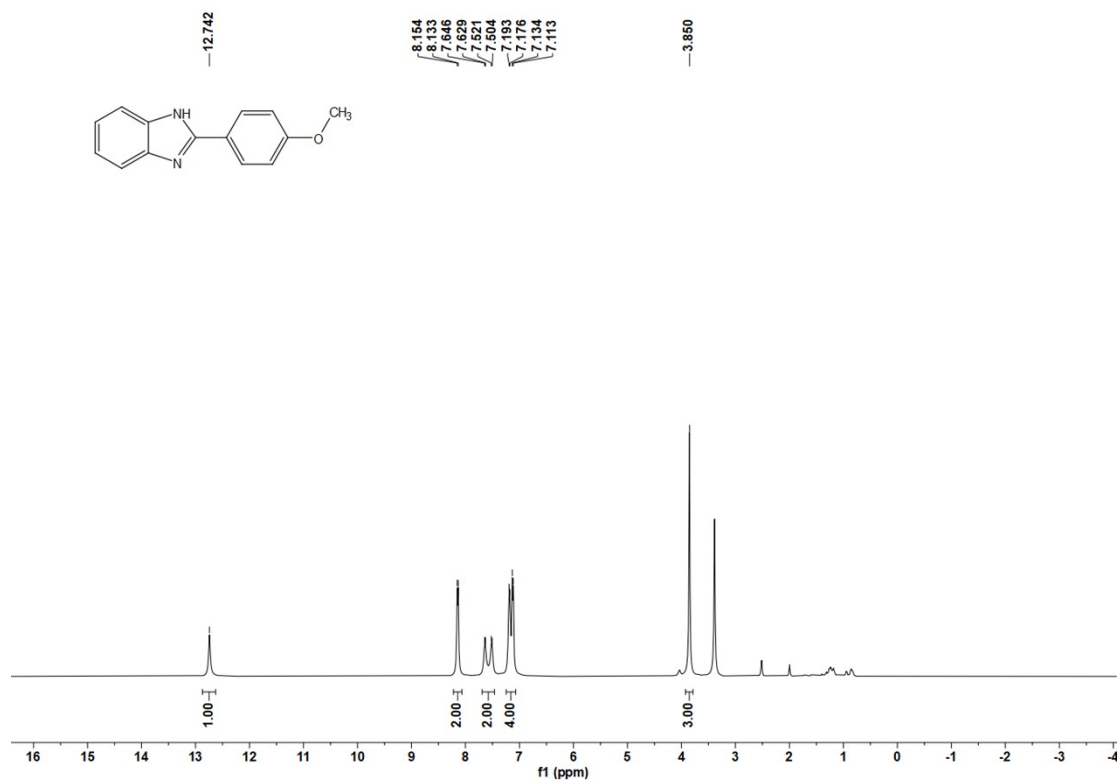
### 2-phenyl-1H-benzo[d]imidazole (3a)



### 2-(p-tolyl)-1H-benzo[d]imidazole (3ab)

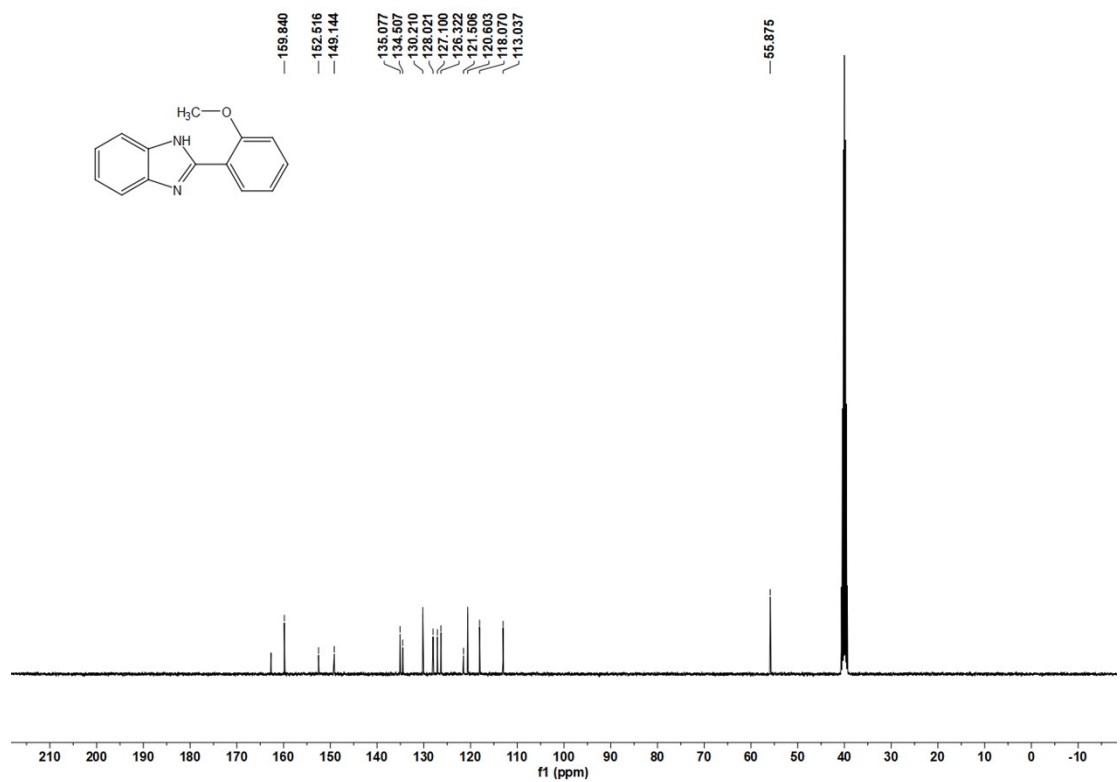
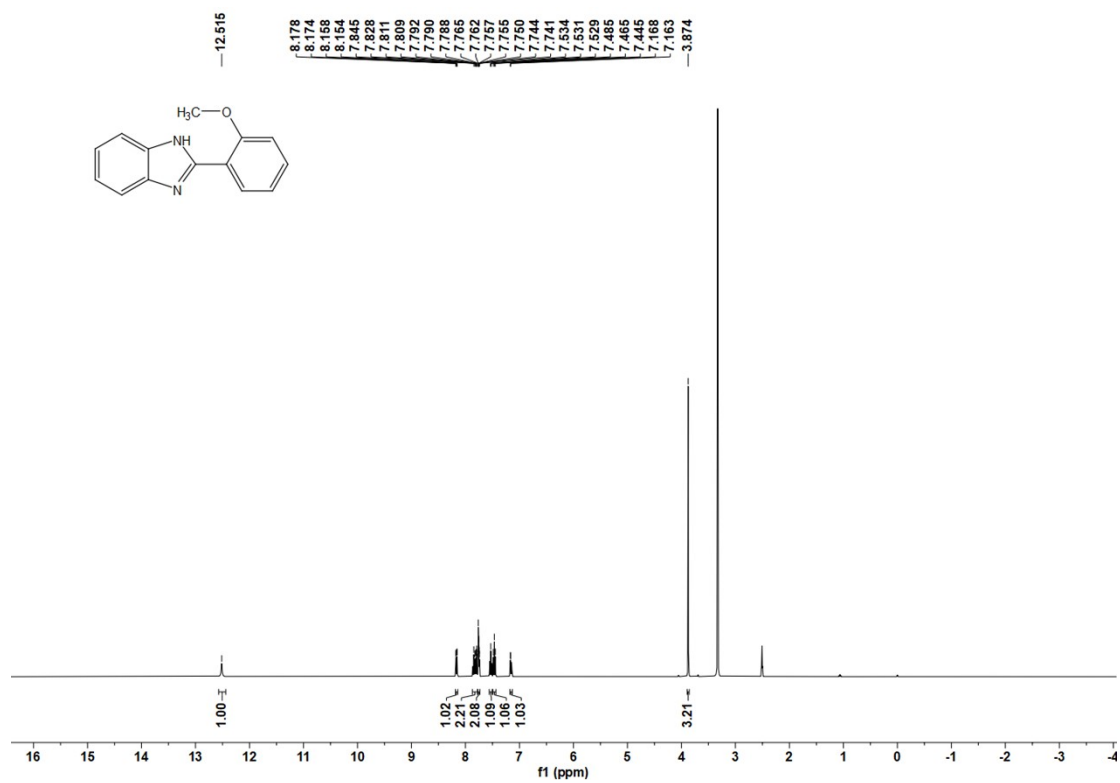


## 2-(4-methoxyphenyl)-1H-benzo[d]imidazole (3ac)

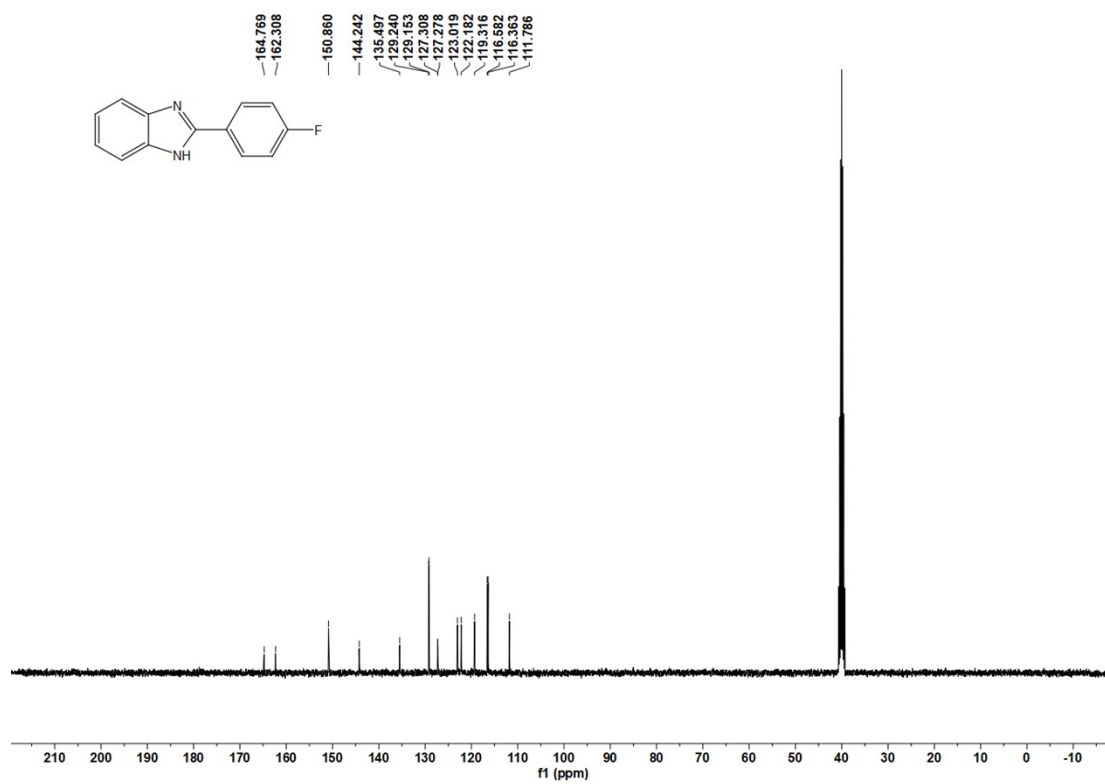
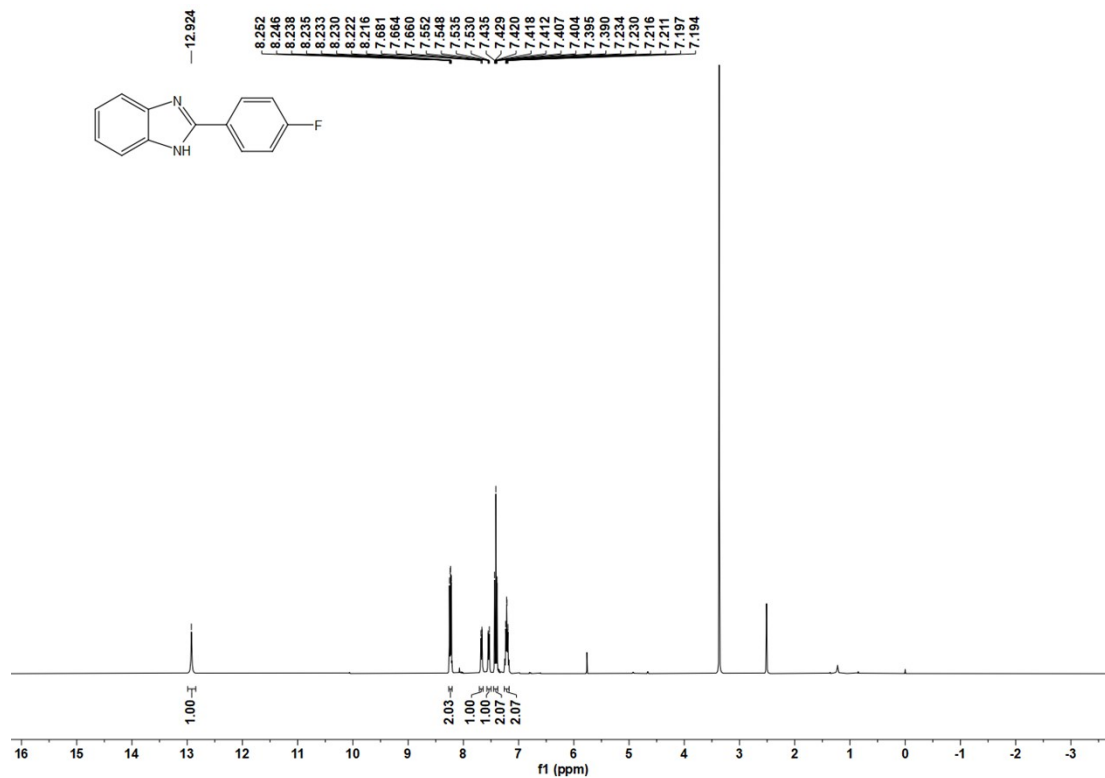




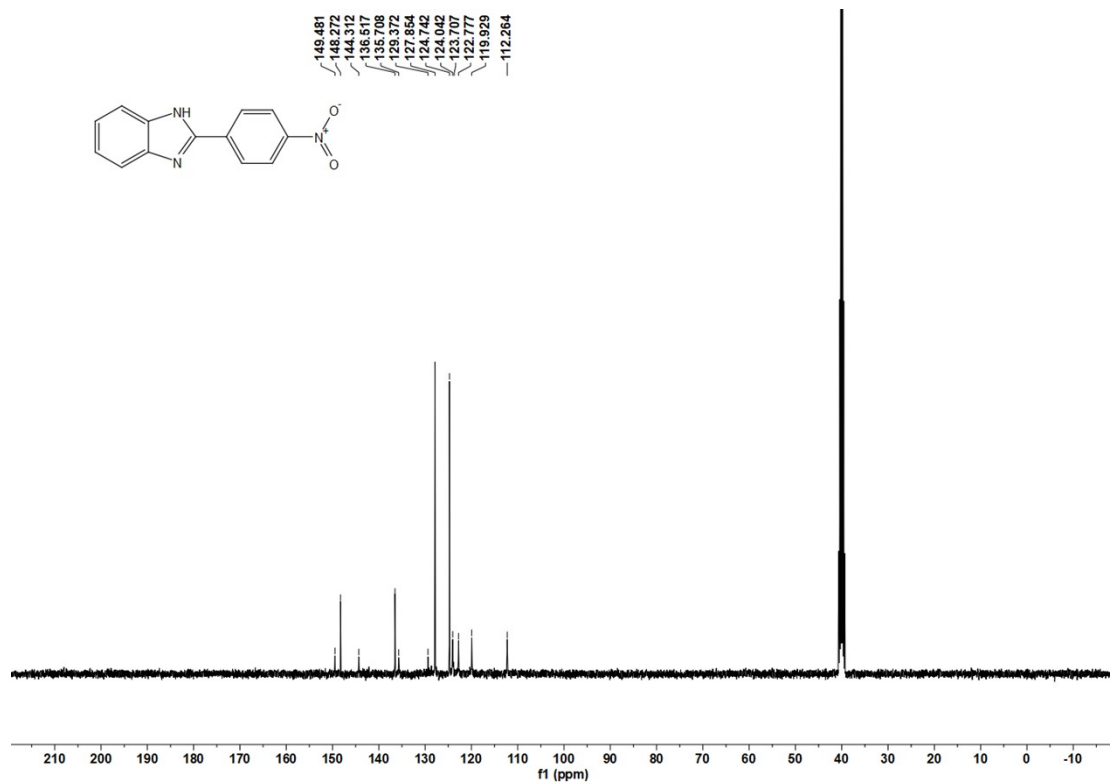
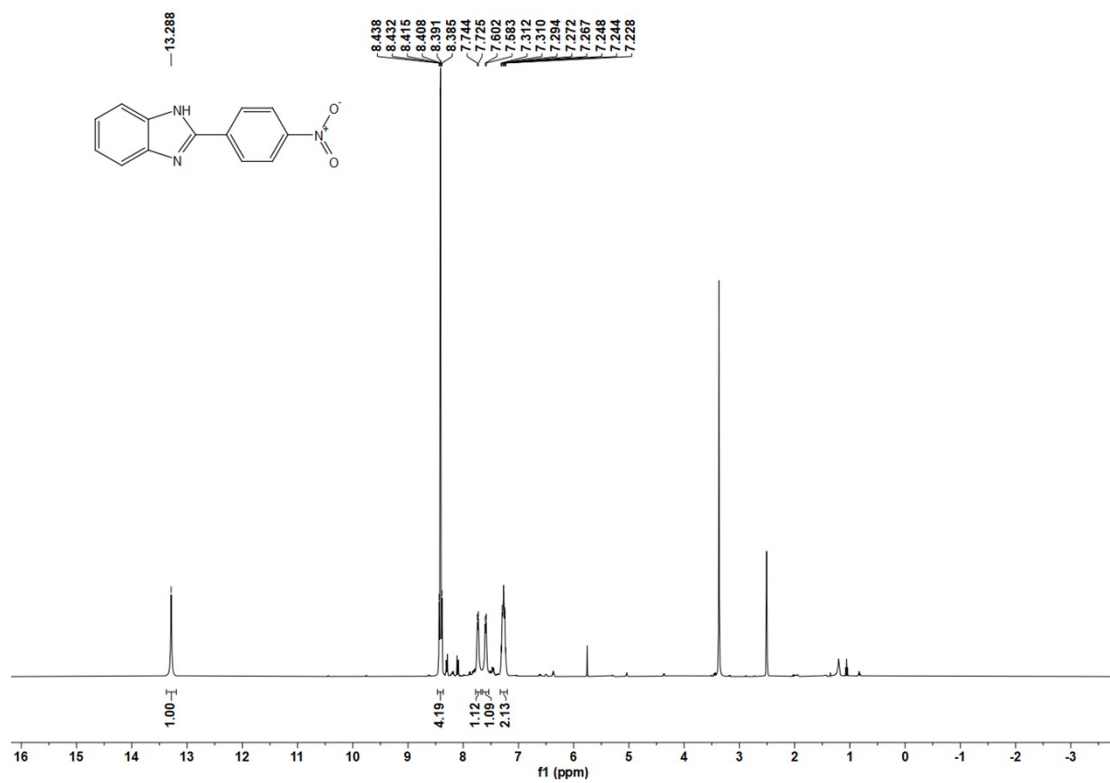
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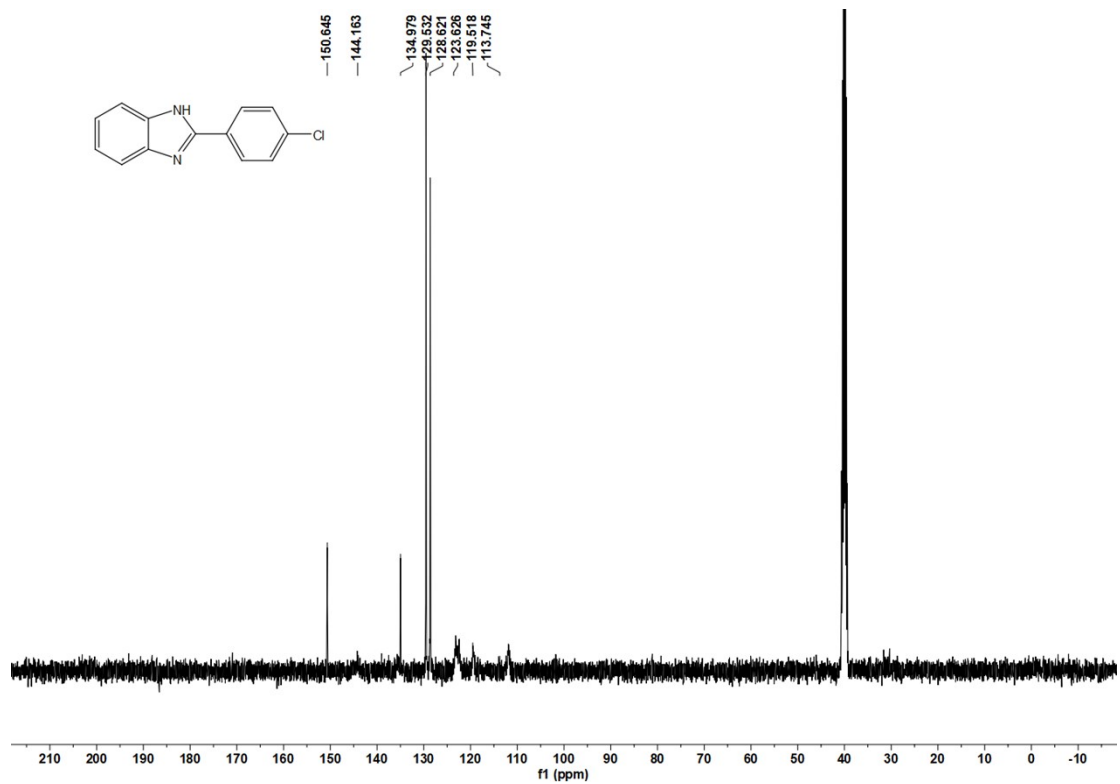
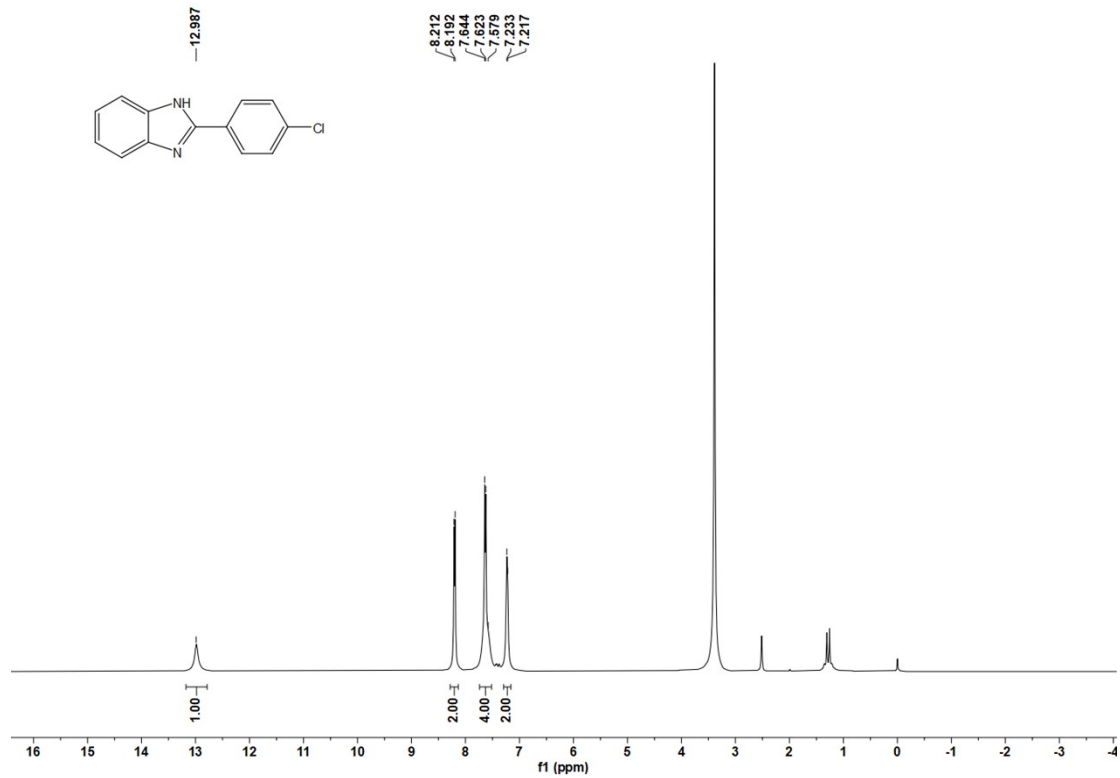
## 2-(4-fluorophenyl)-1H-benzo[d]imidazole (3ac)



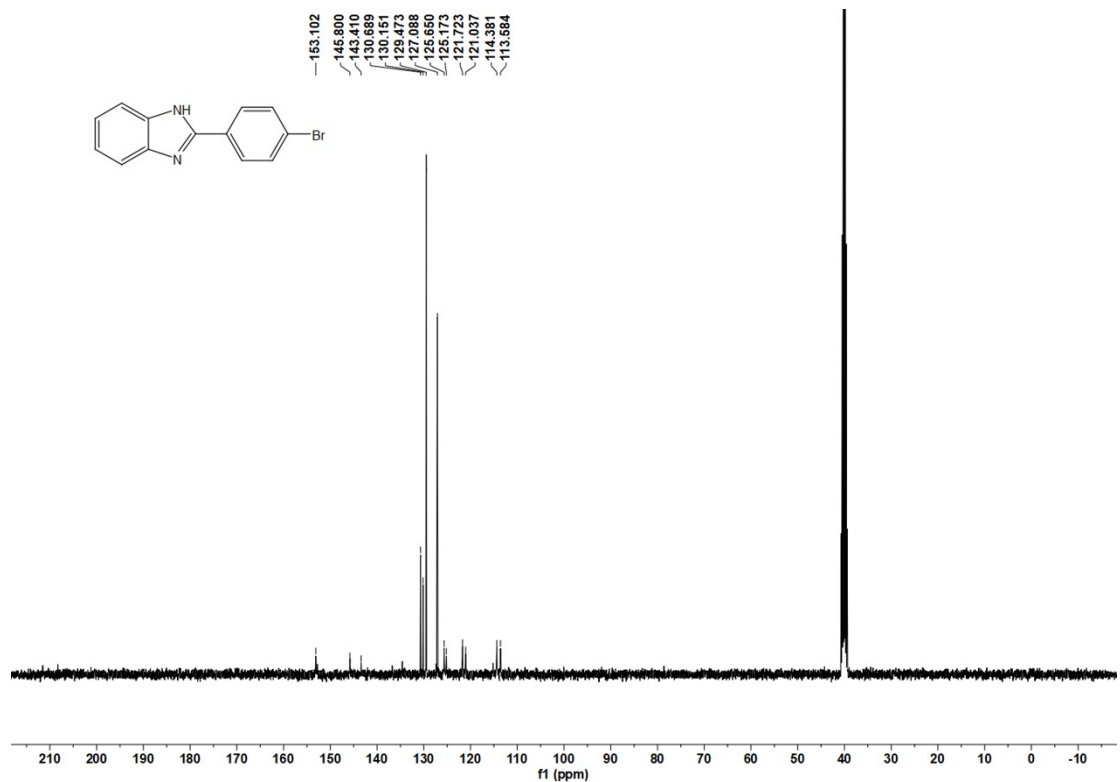
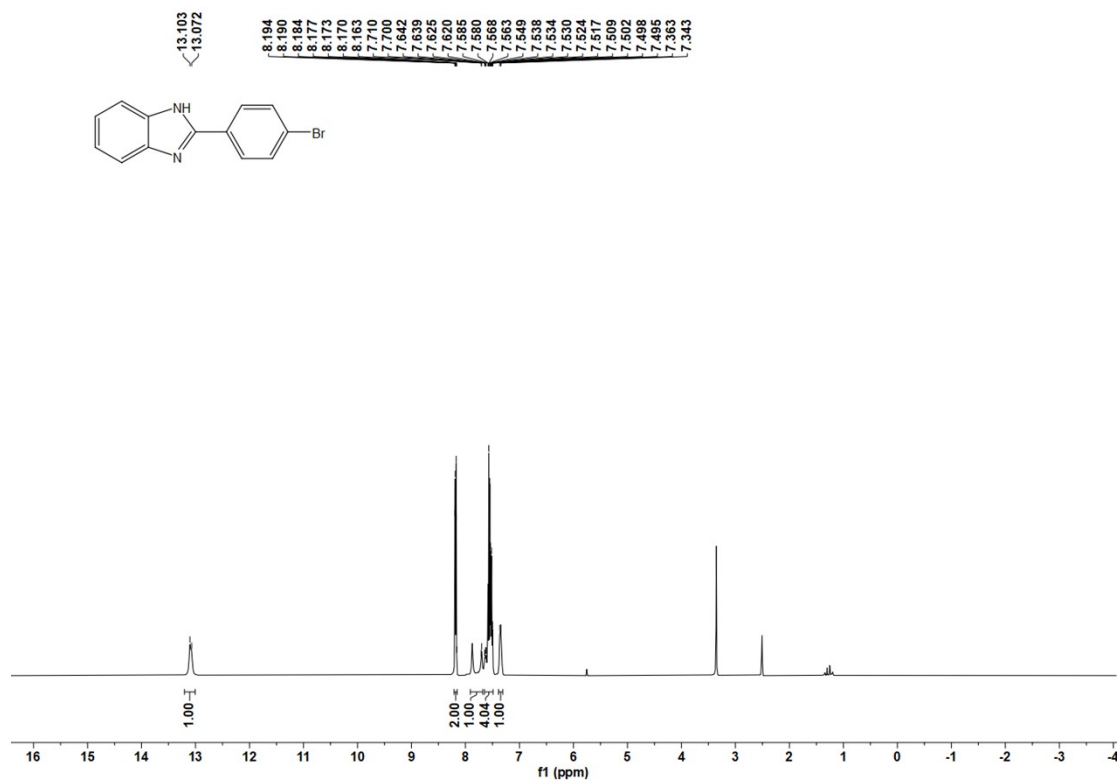
### 2-(4-nitrophenyl)-1H-benzo[d]imidazole (3af)



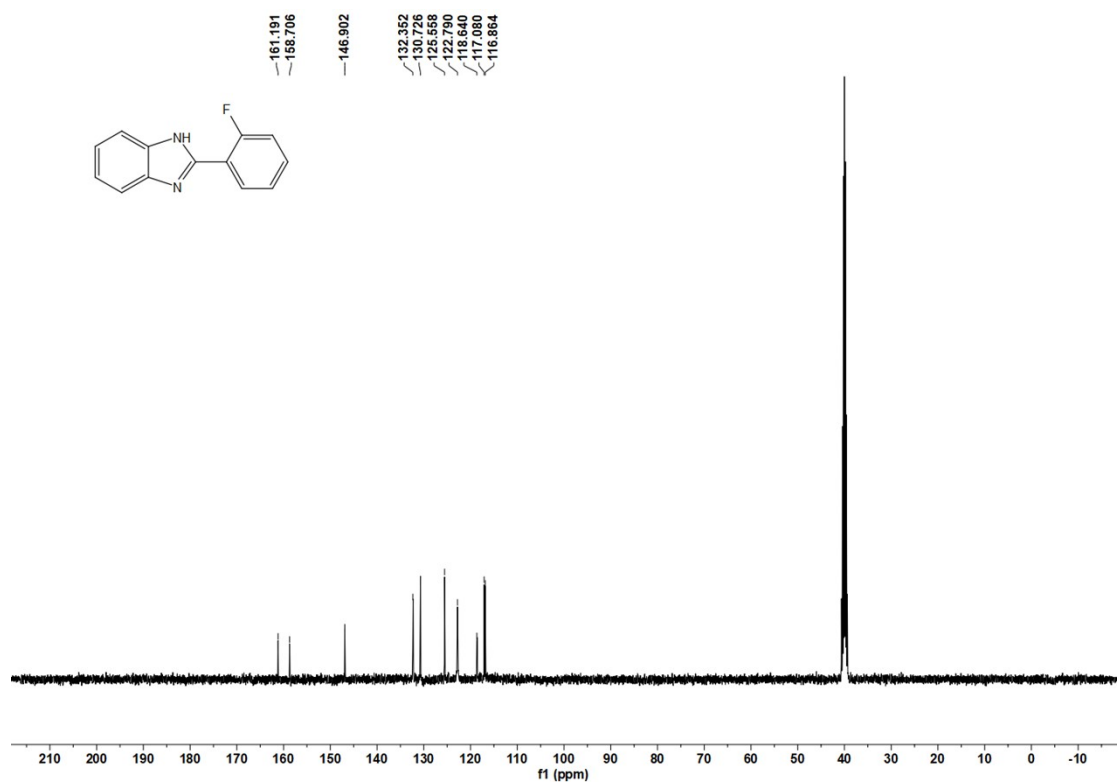
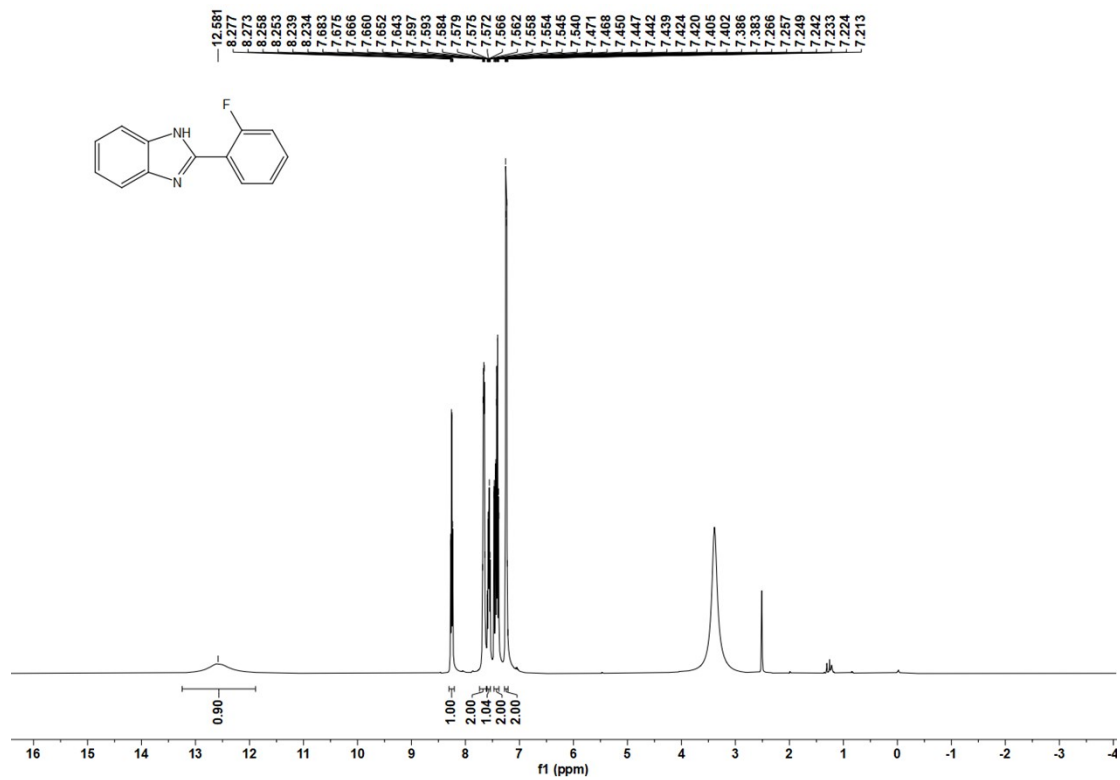
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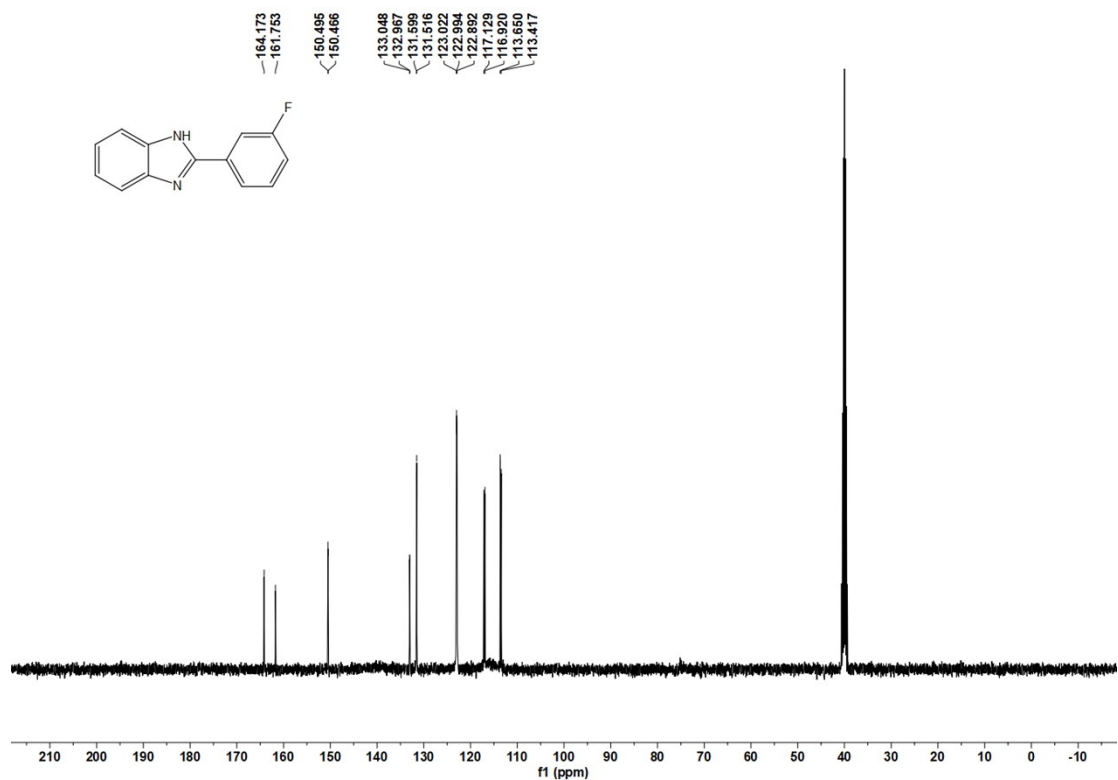
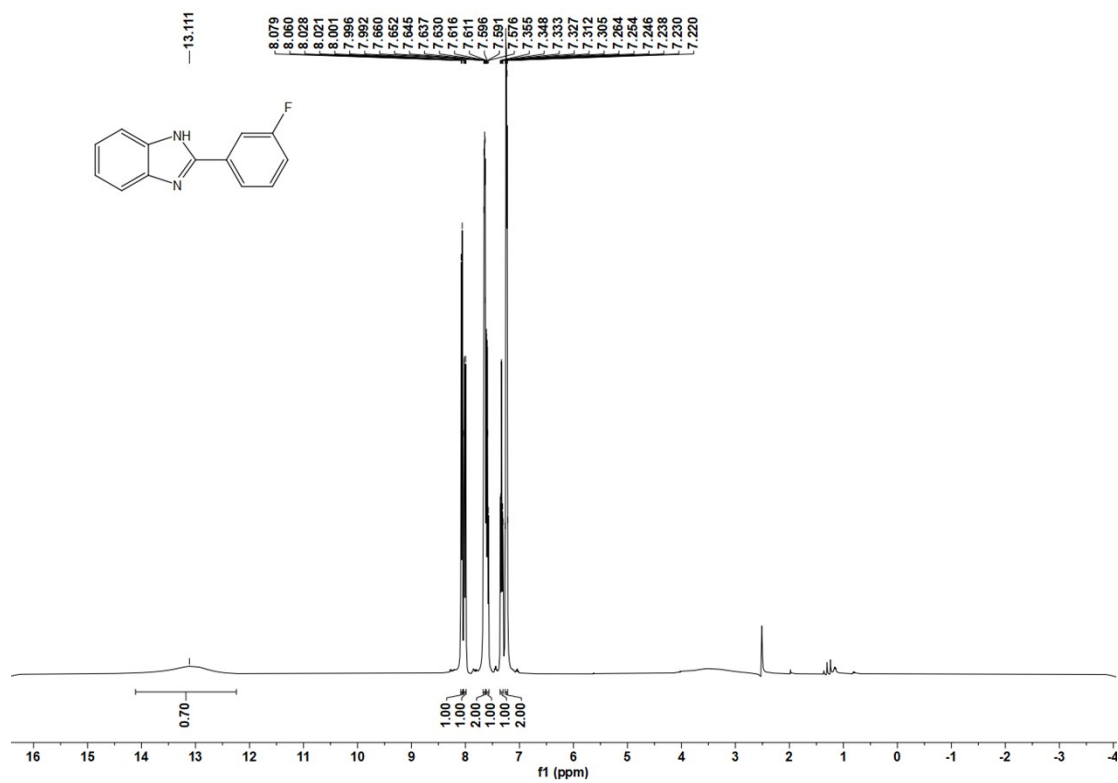
## 2-(4-bromophenyl)-1H-benzo[d]imidazole (3ah)



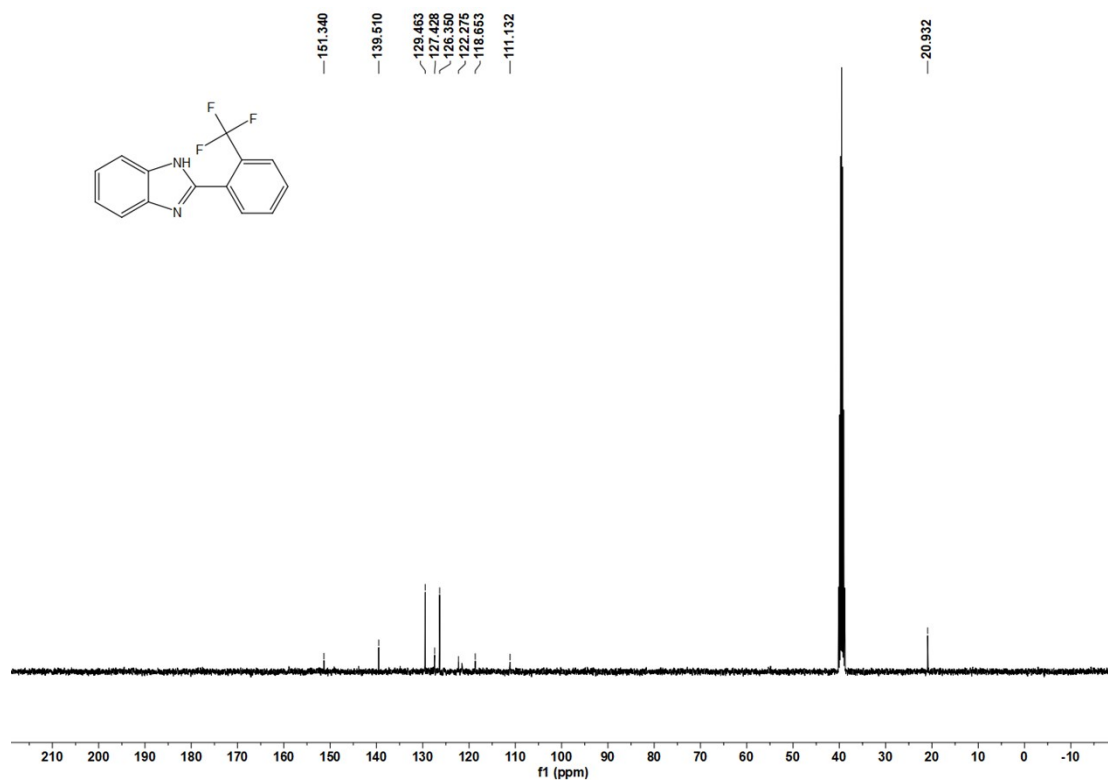
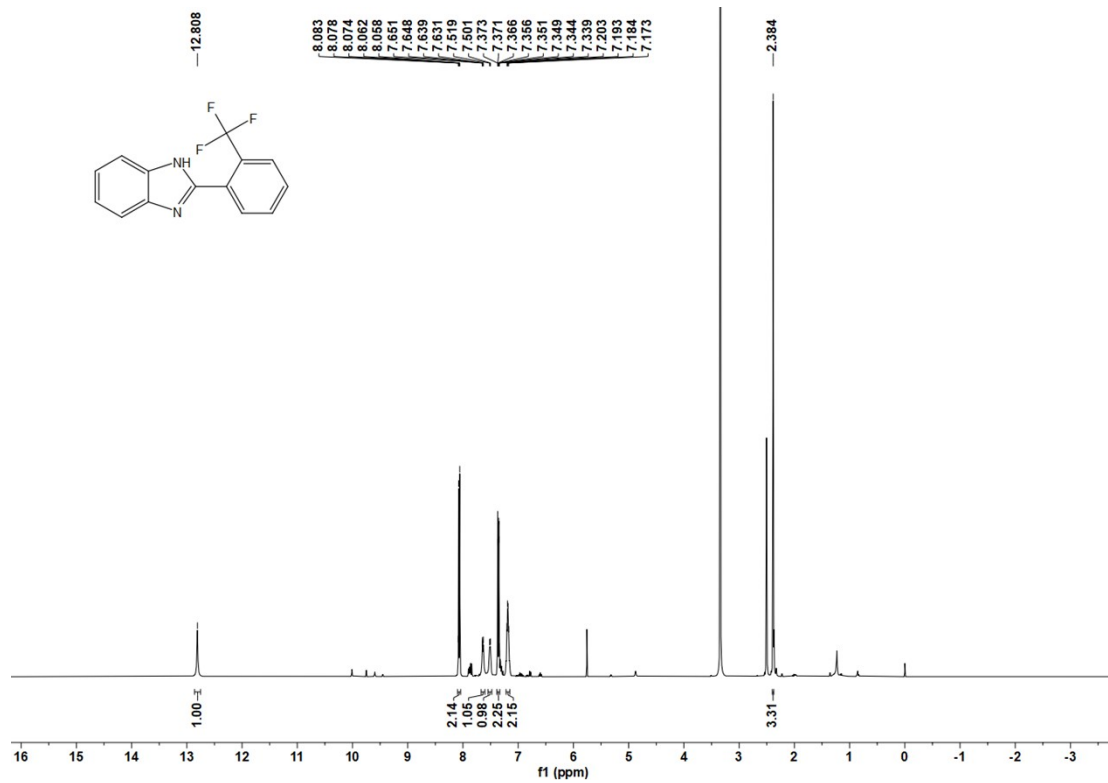
2-(2-fluorophenyl)-1H-benzo[d]imidazole (3ai)



### 2-(3-fluorophenyl)-1H-benzo[d]imidazole (3aj)

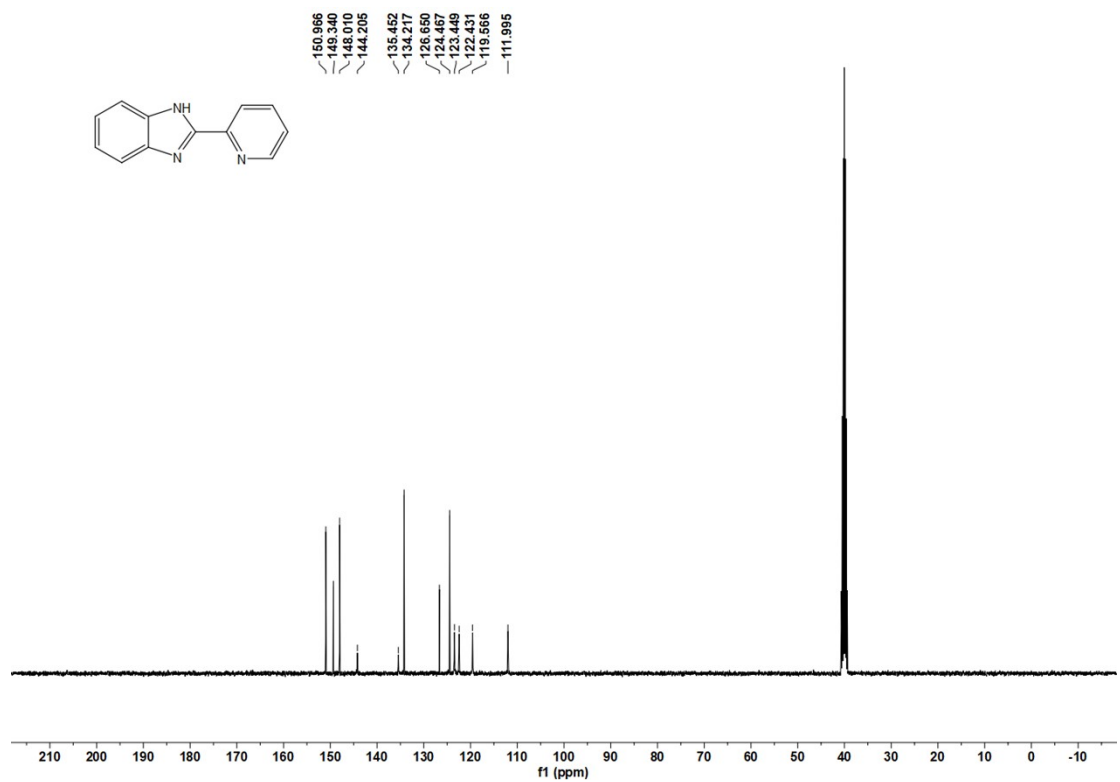
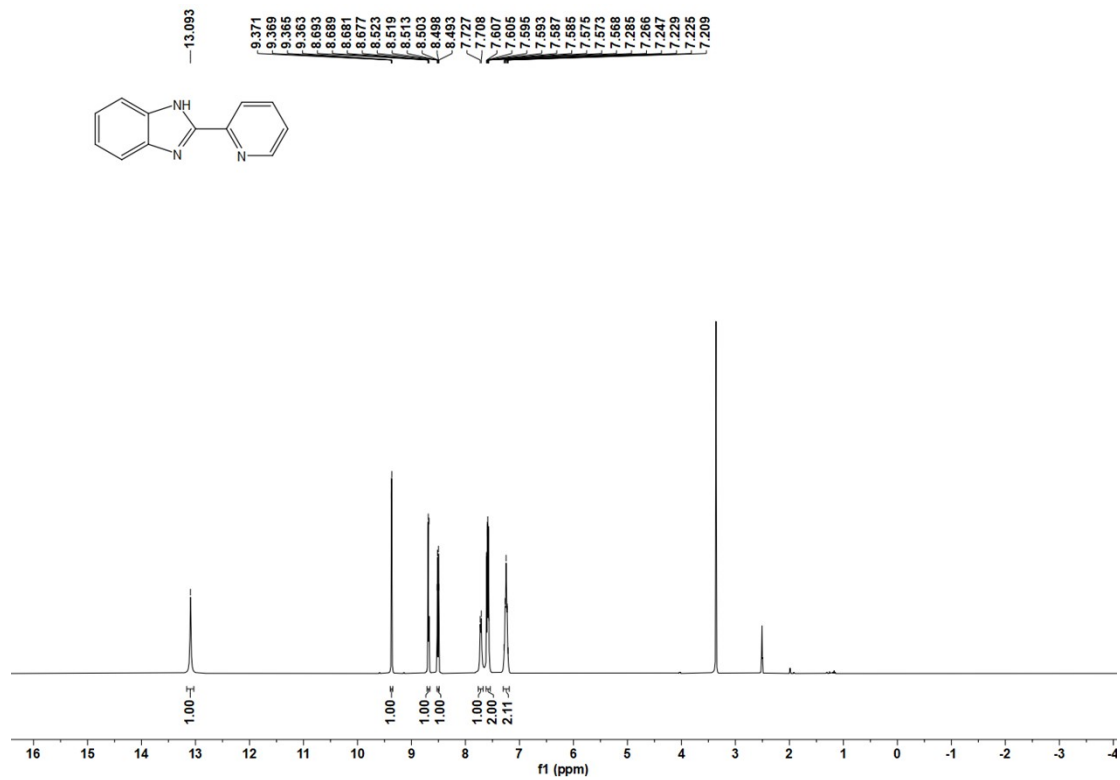


### 2-(2-(trifluoromethyl)phenyl)-1H-benzo[d]imidazole (3ak)

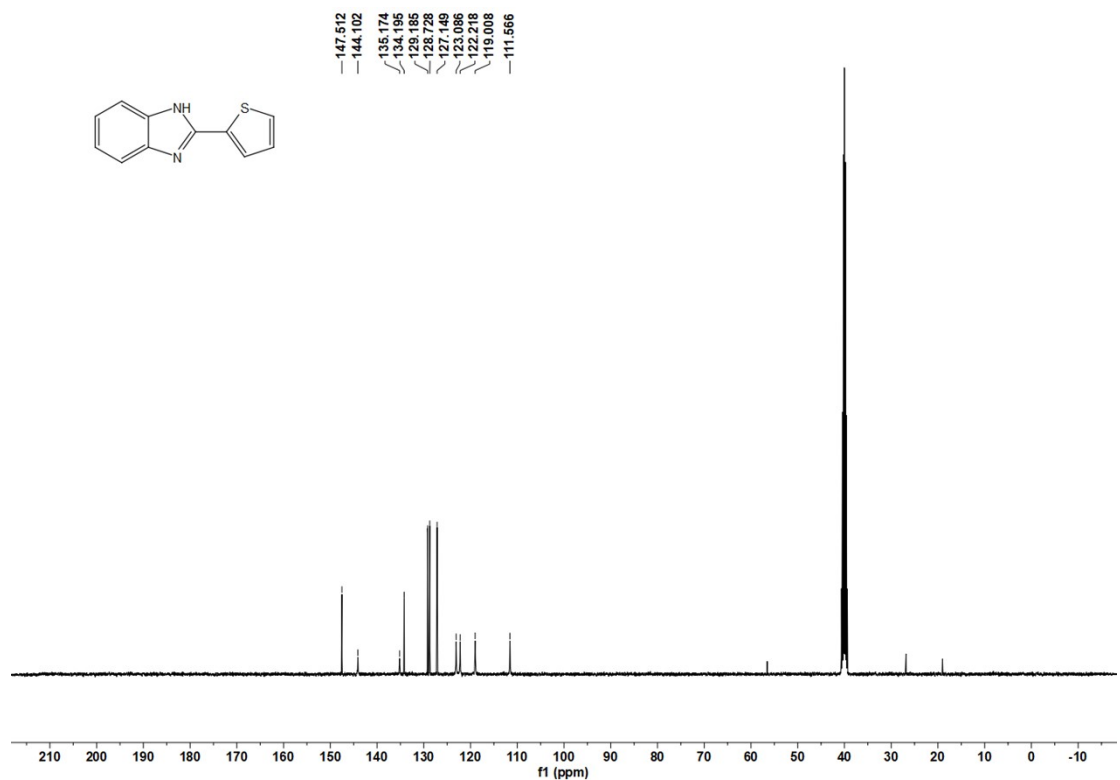
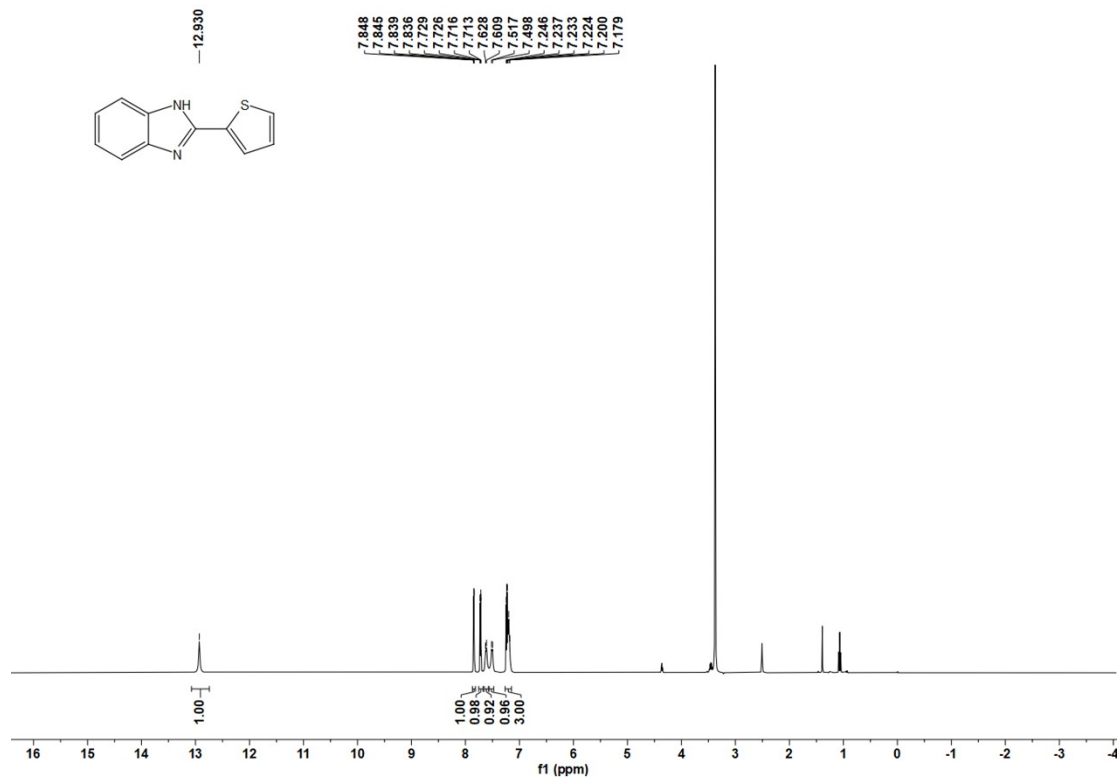




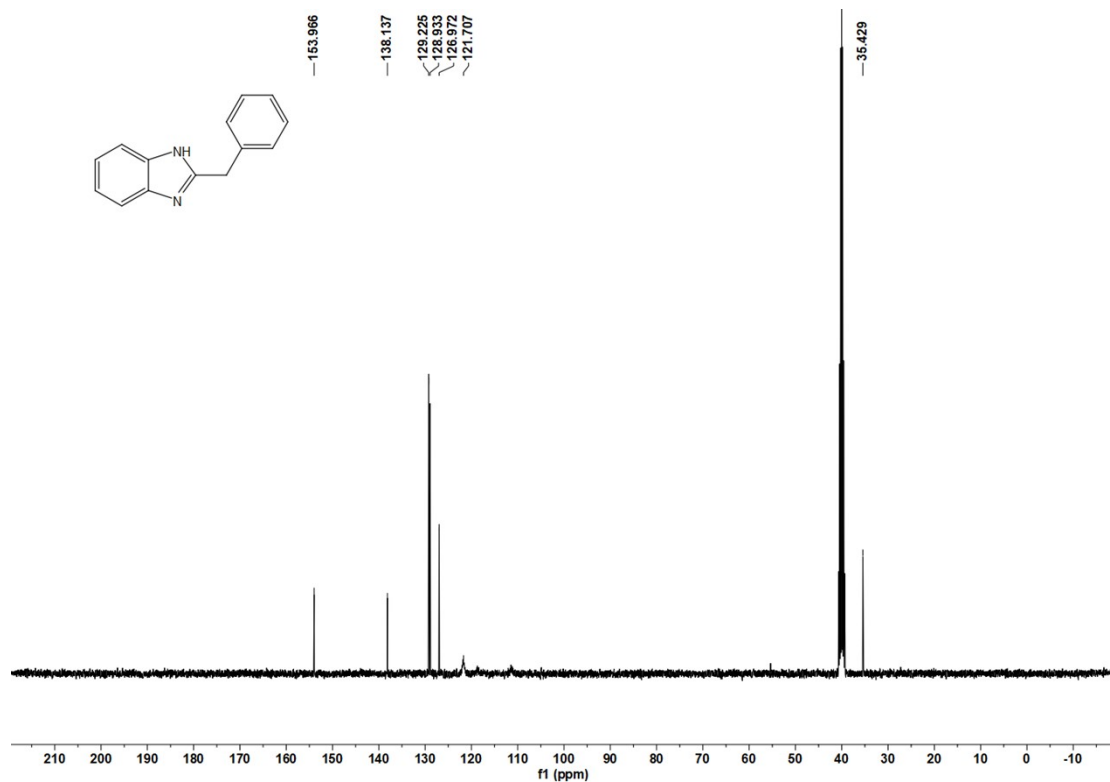
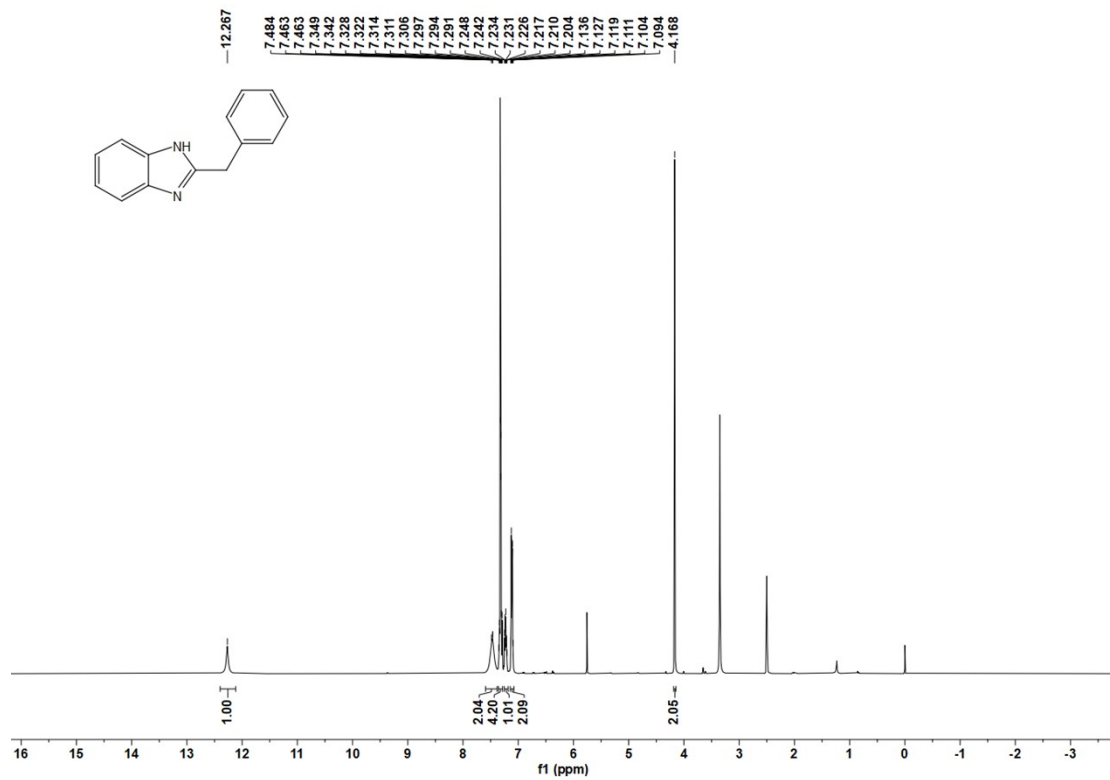
### 2-(pyridin-2-yl)-1H-benzo[d]imidazole (3a)



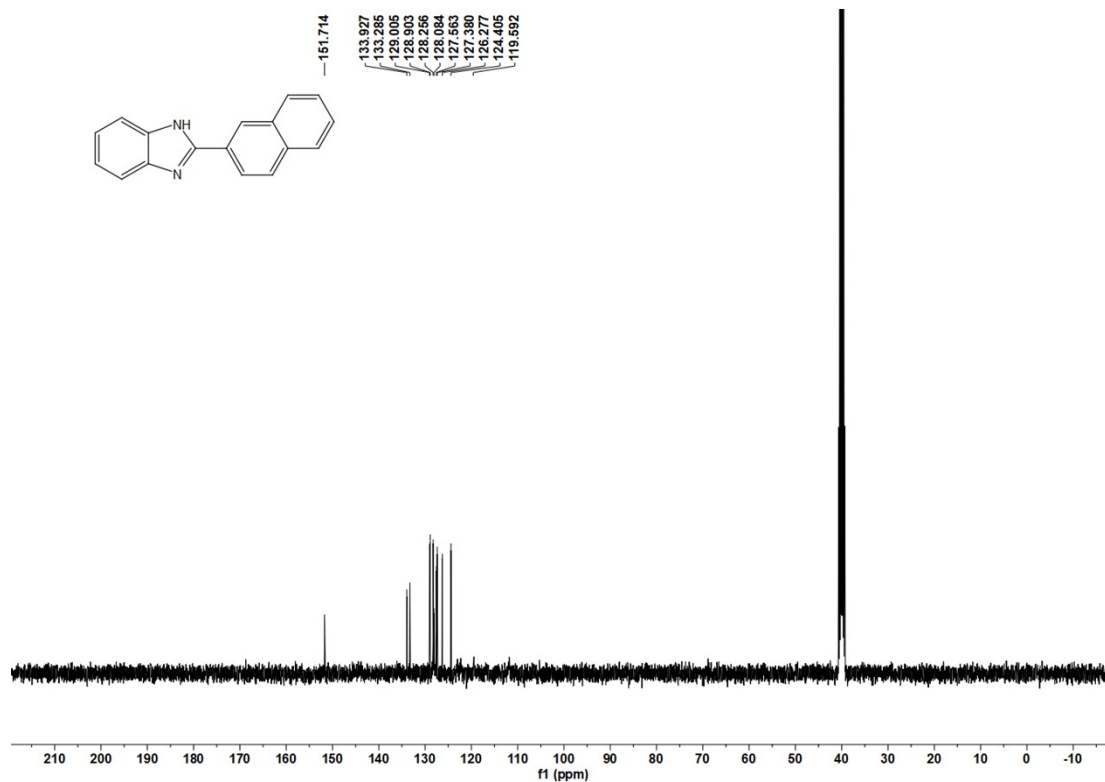
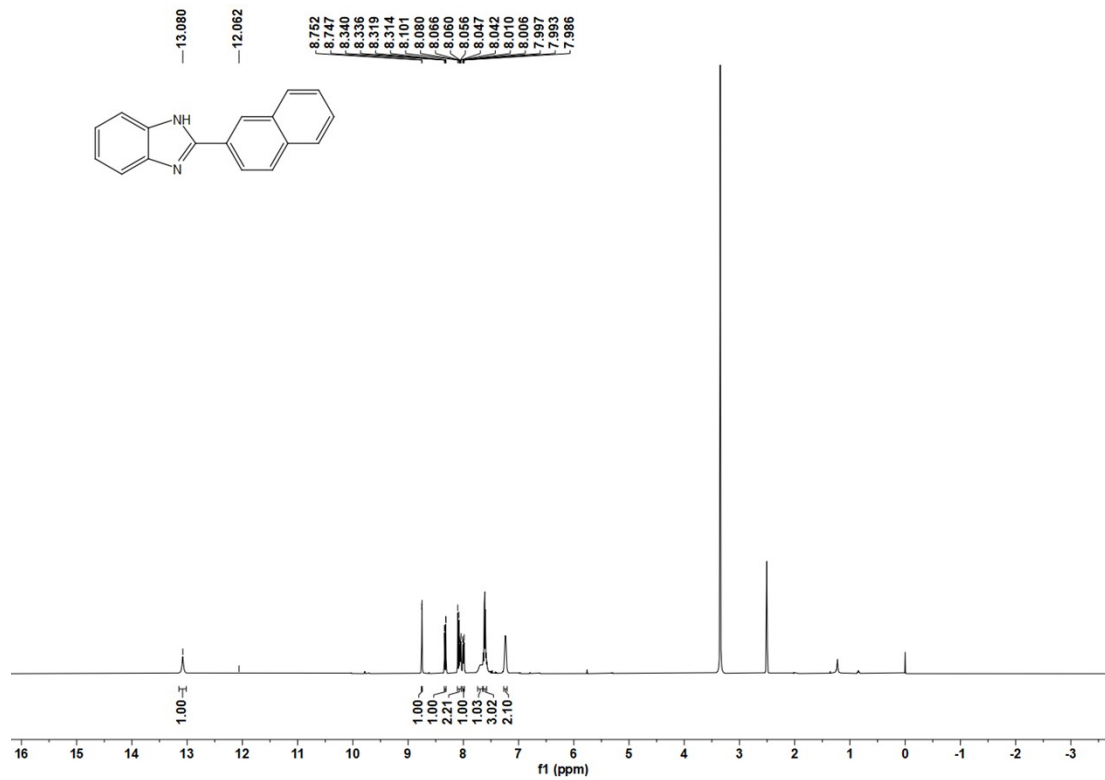
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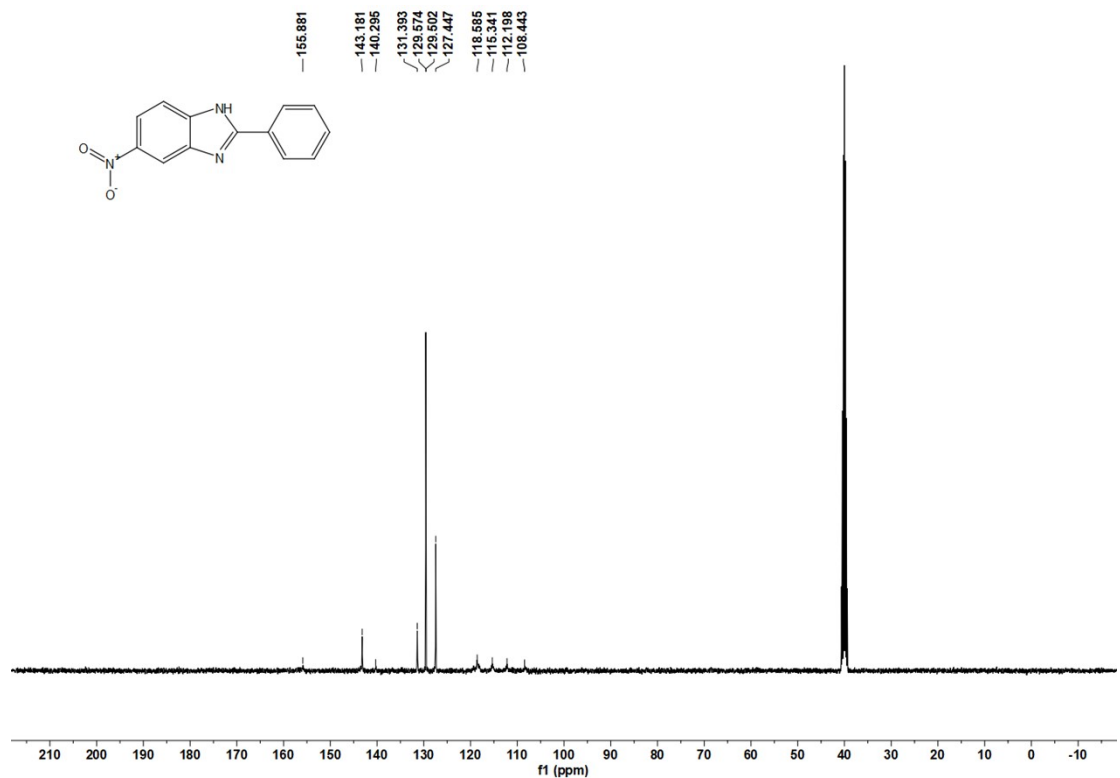
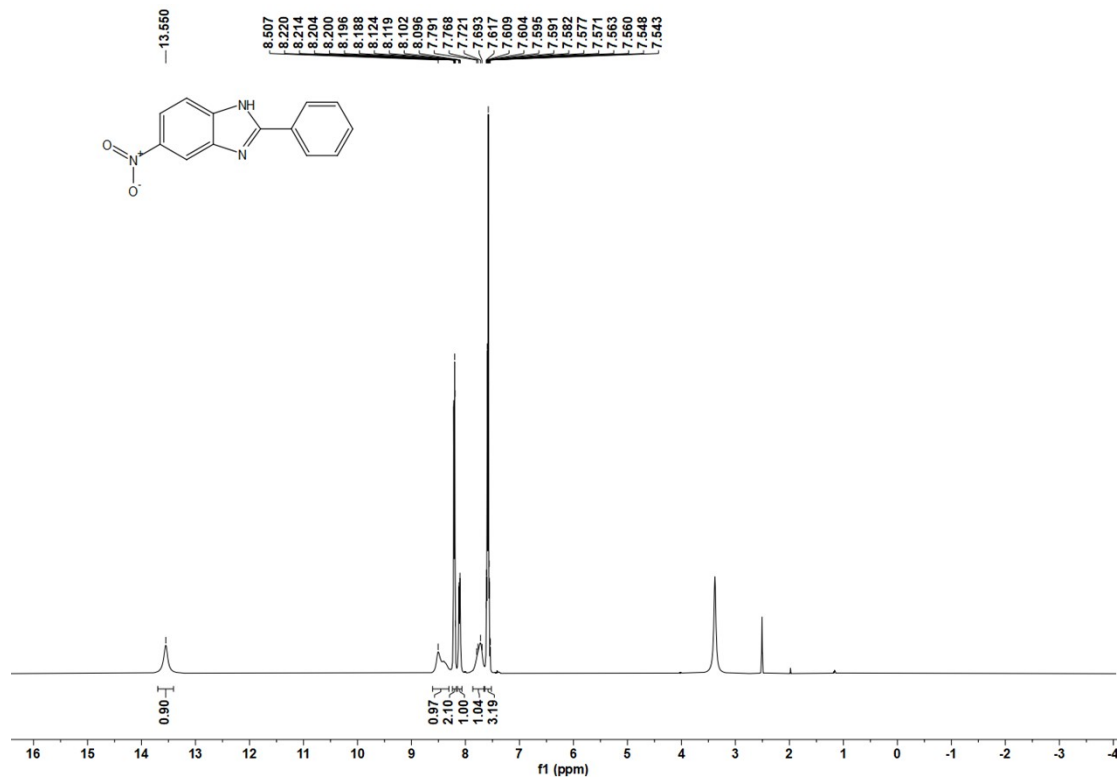
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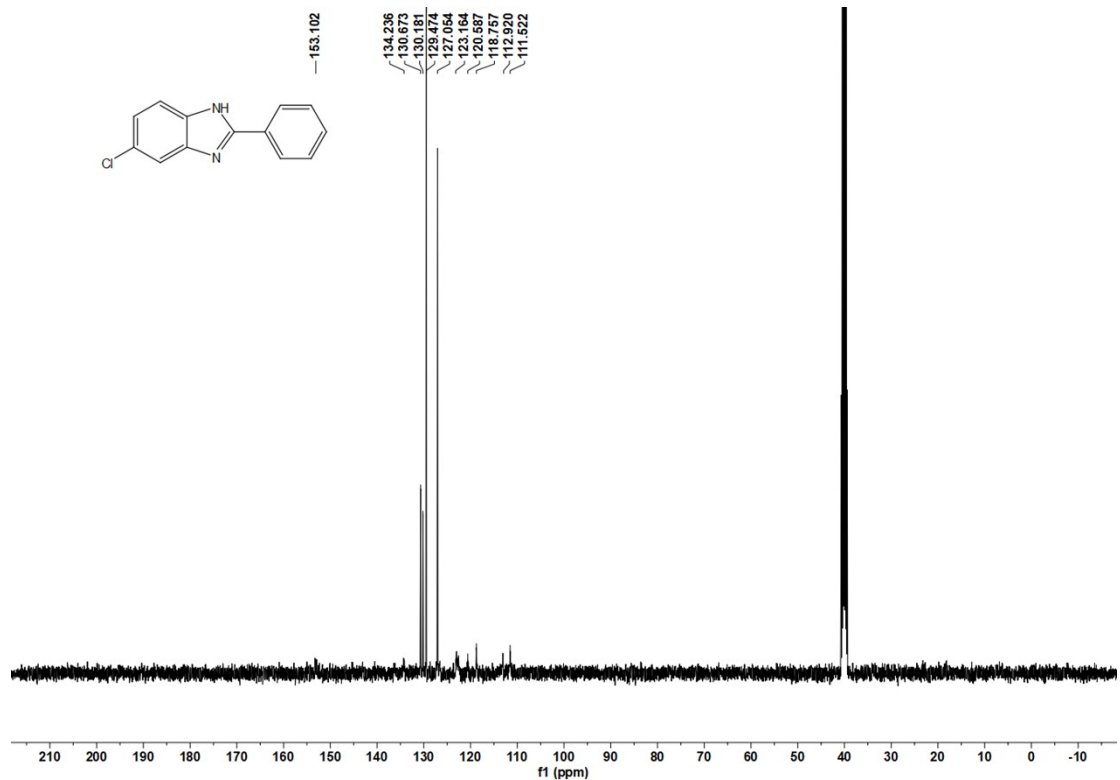
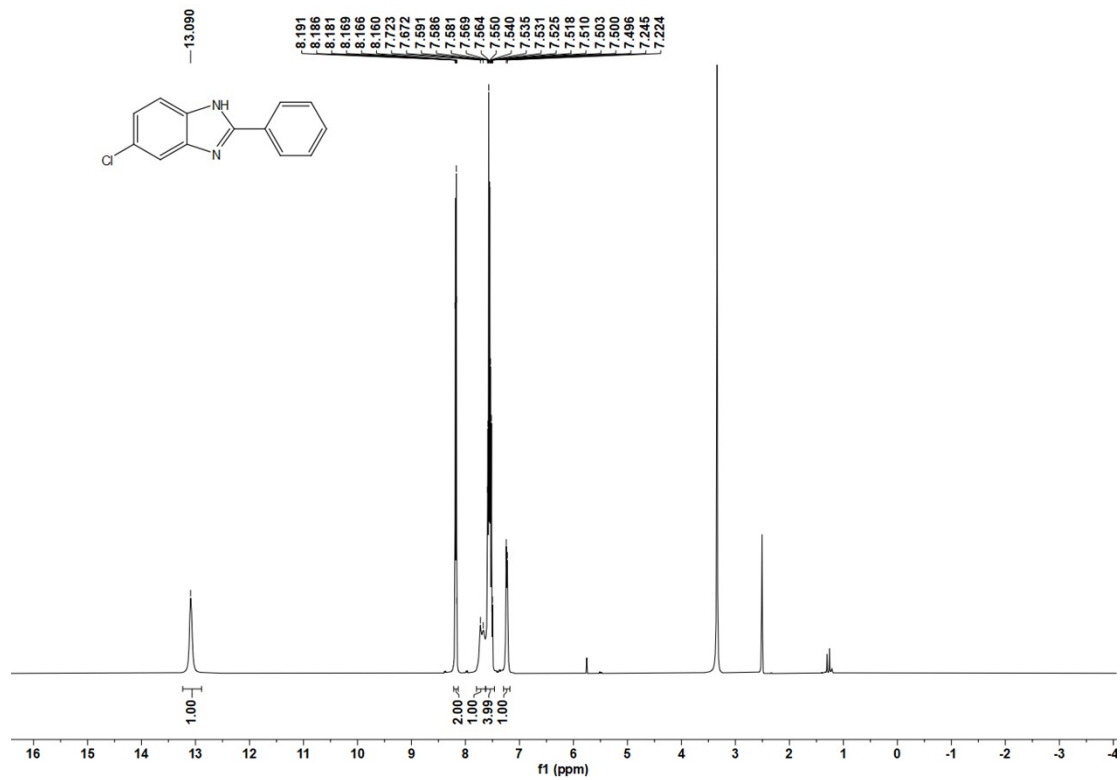
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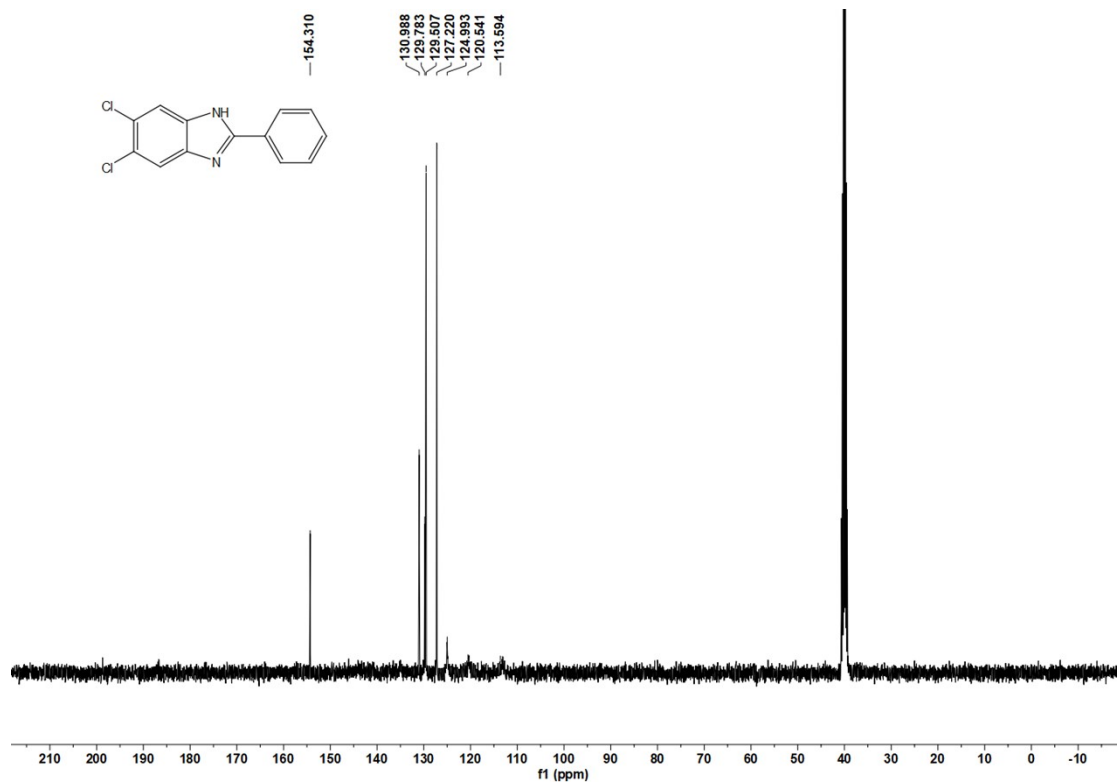
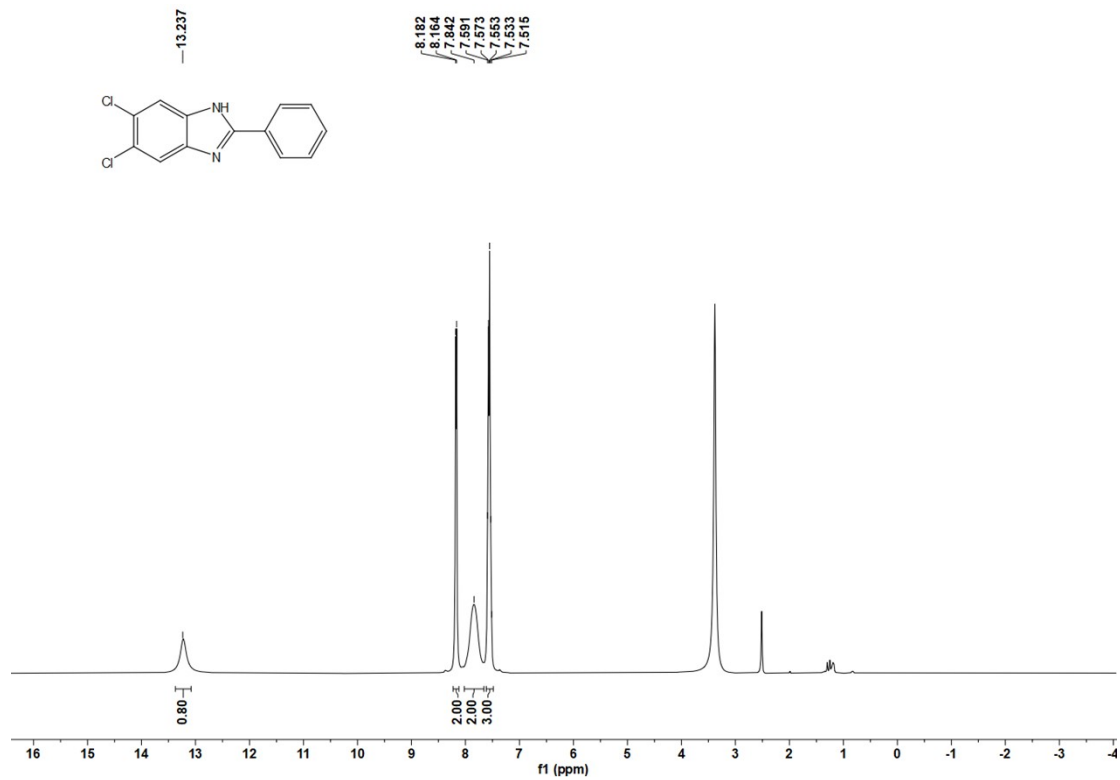
# 5-nitro-2-phenyl-1H-benzo[d]imidazole (3ap)



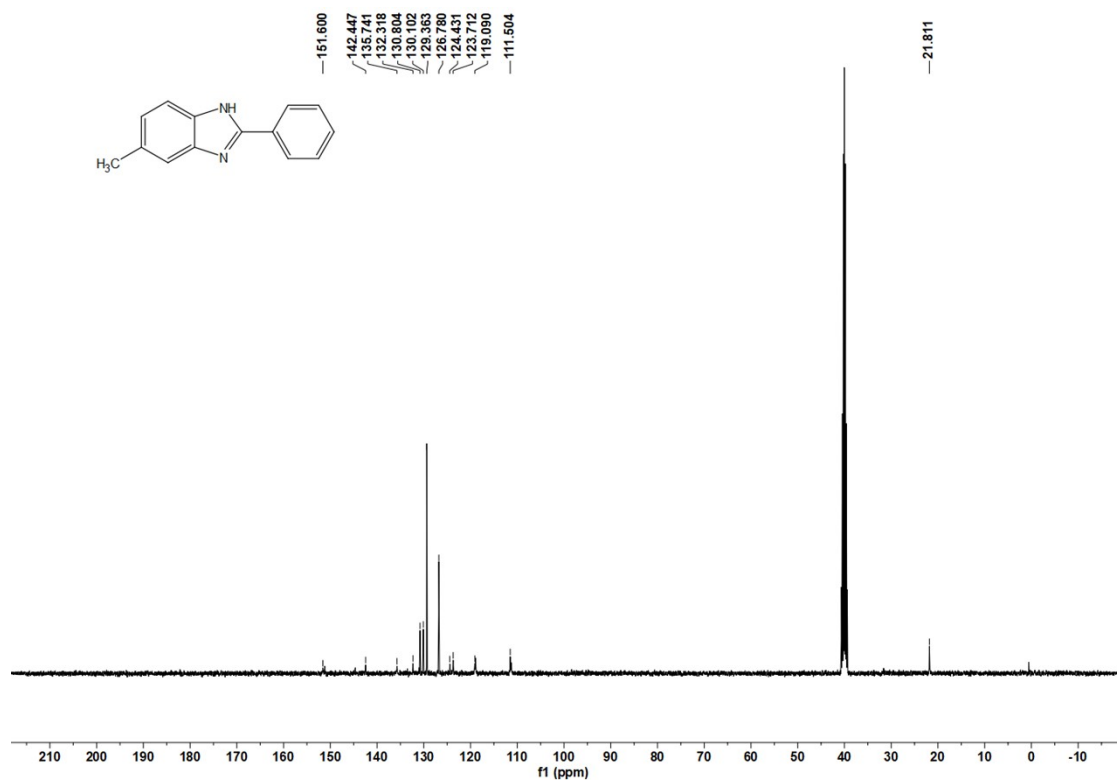
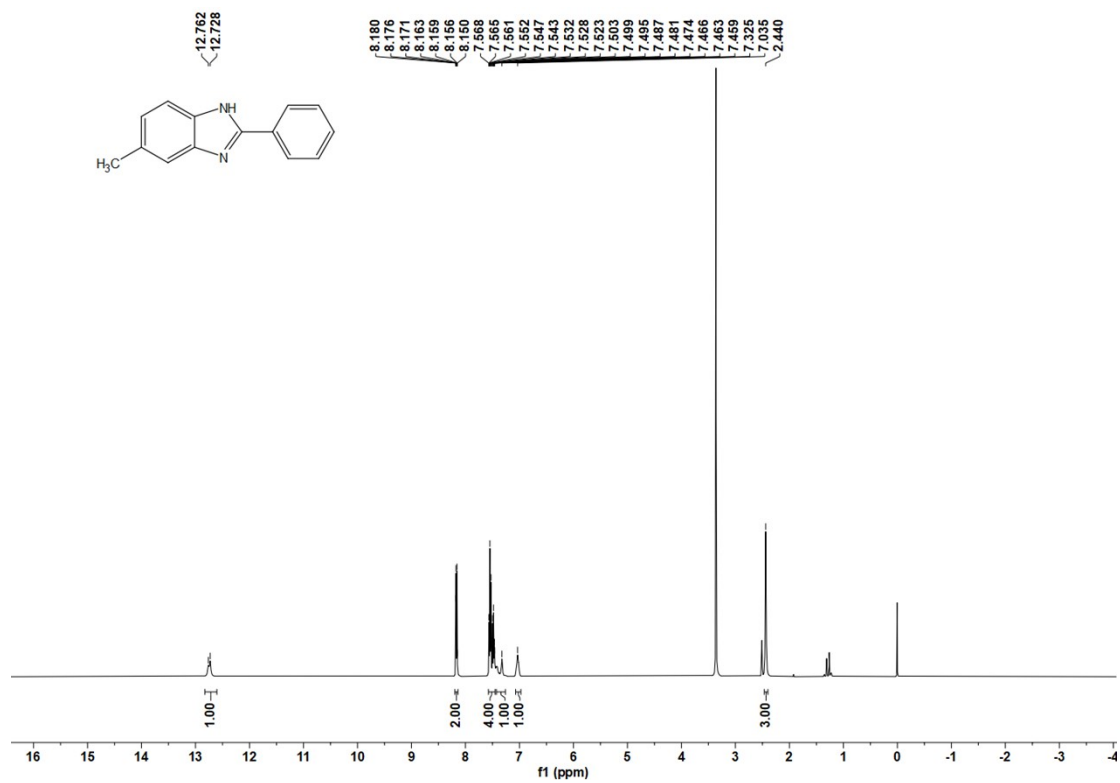
### 5-chloro-2-phenyl-1H-benzo[d]imidazole (3aq)



### 5,6-dichloro-2-phenyl-1H-benzo[d]imidazole (3ar)

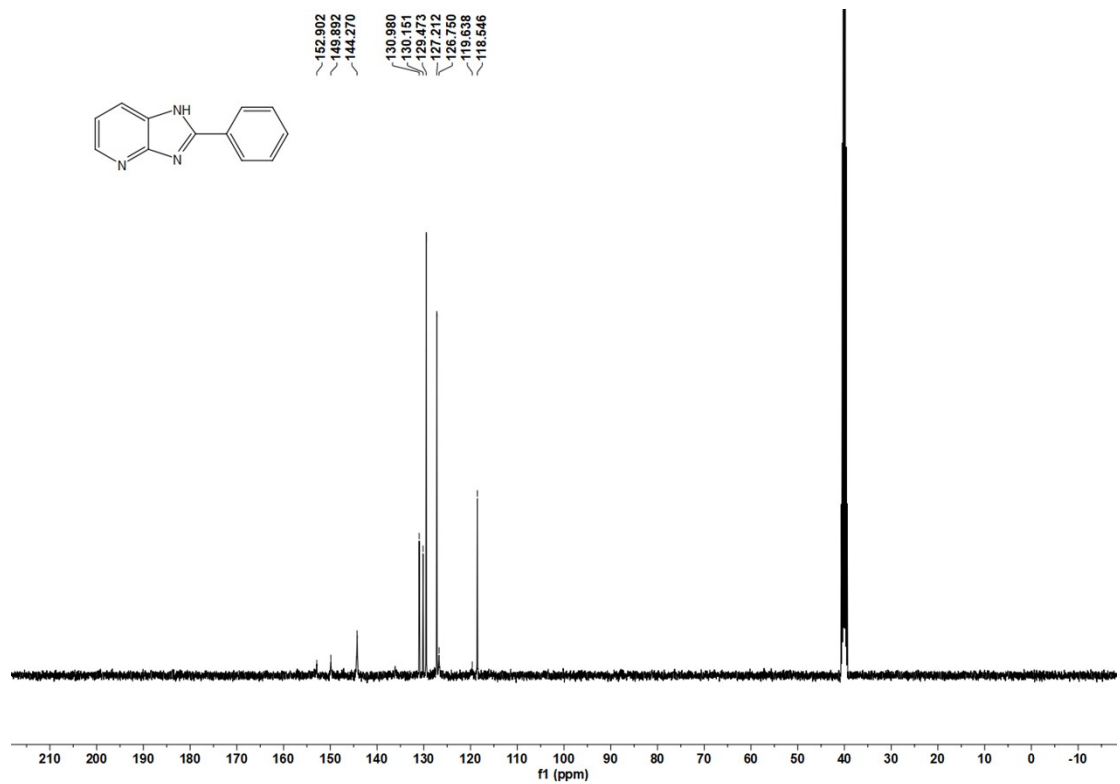
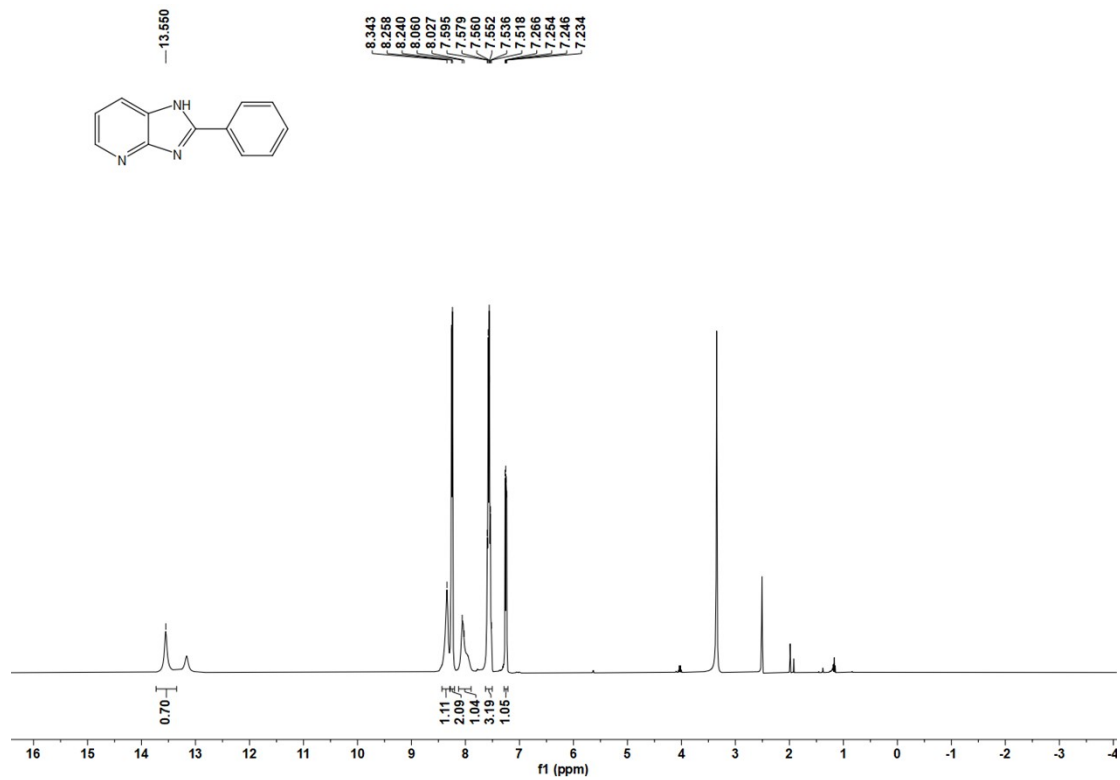


### 5-methyl-2-phenyl-1H-benzo[d]imidazole (3as)

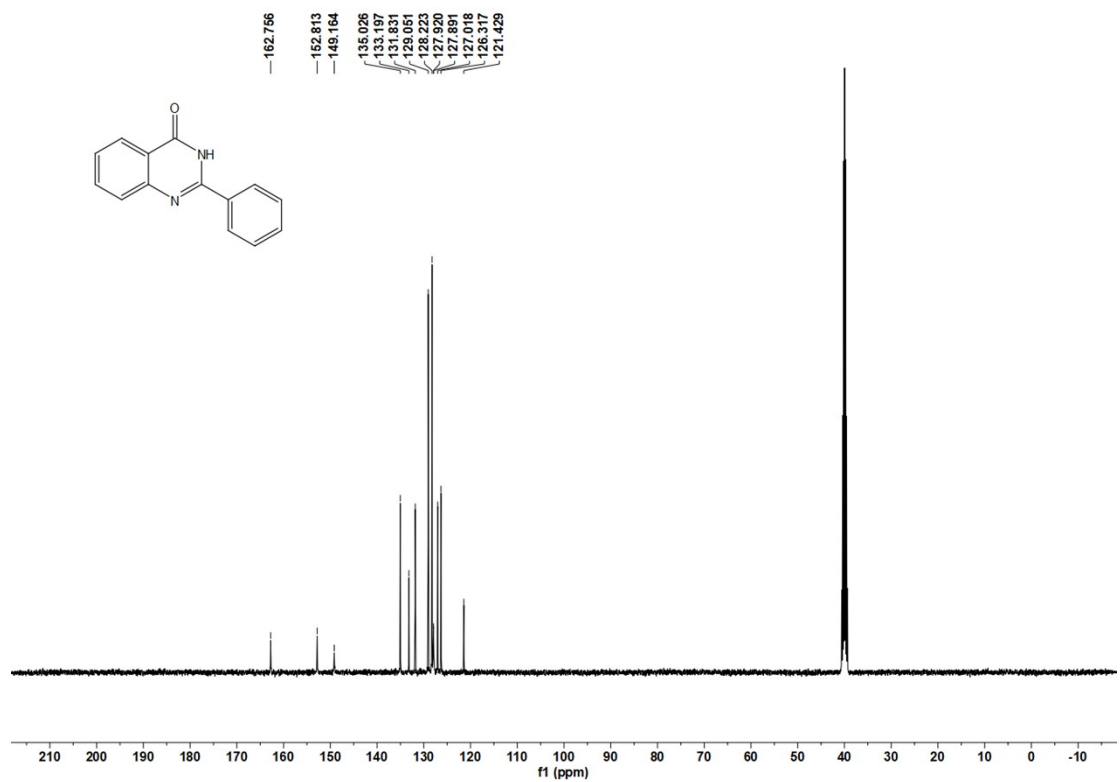
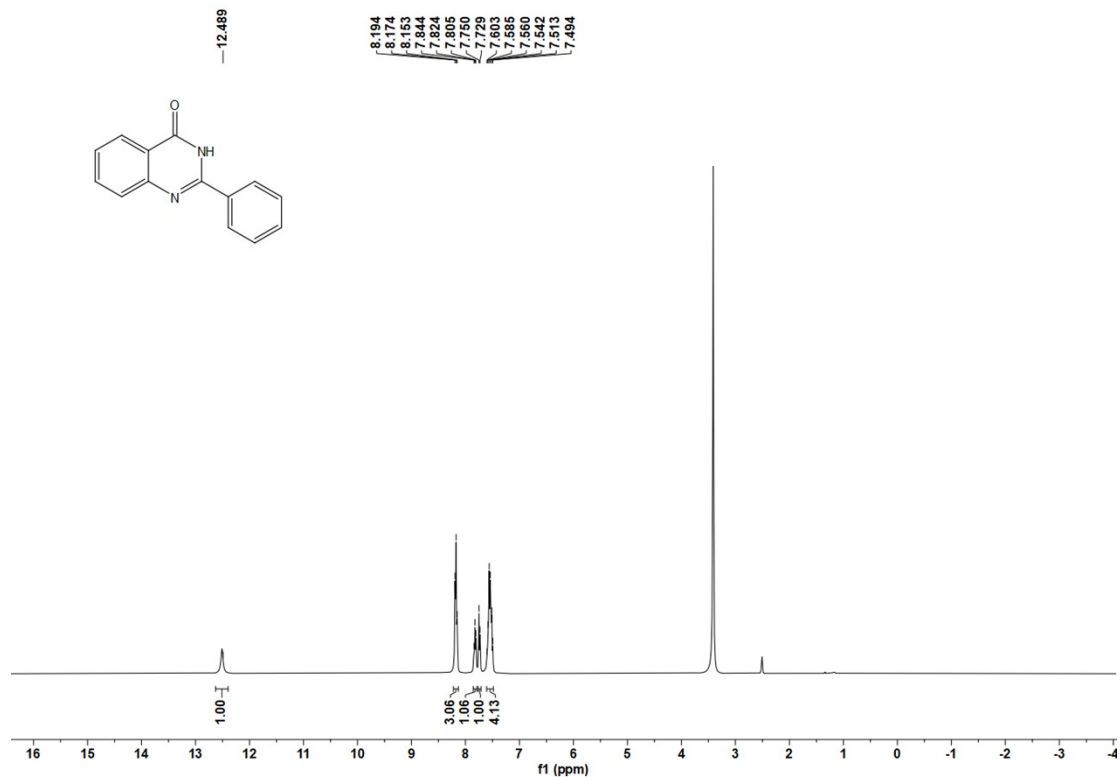




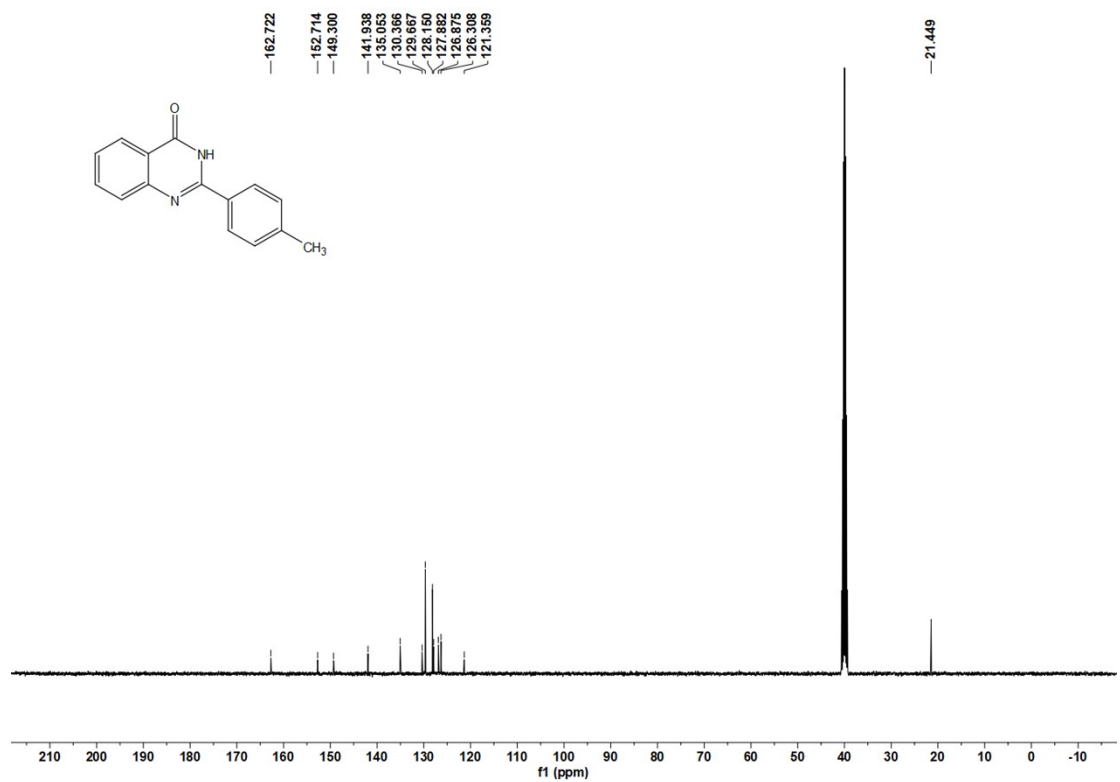
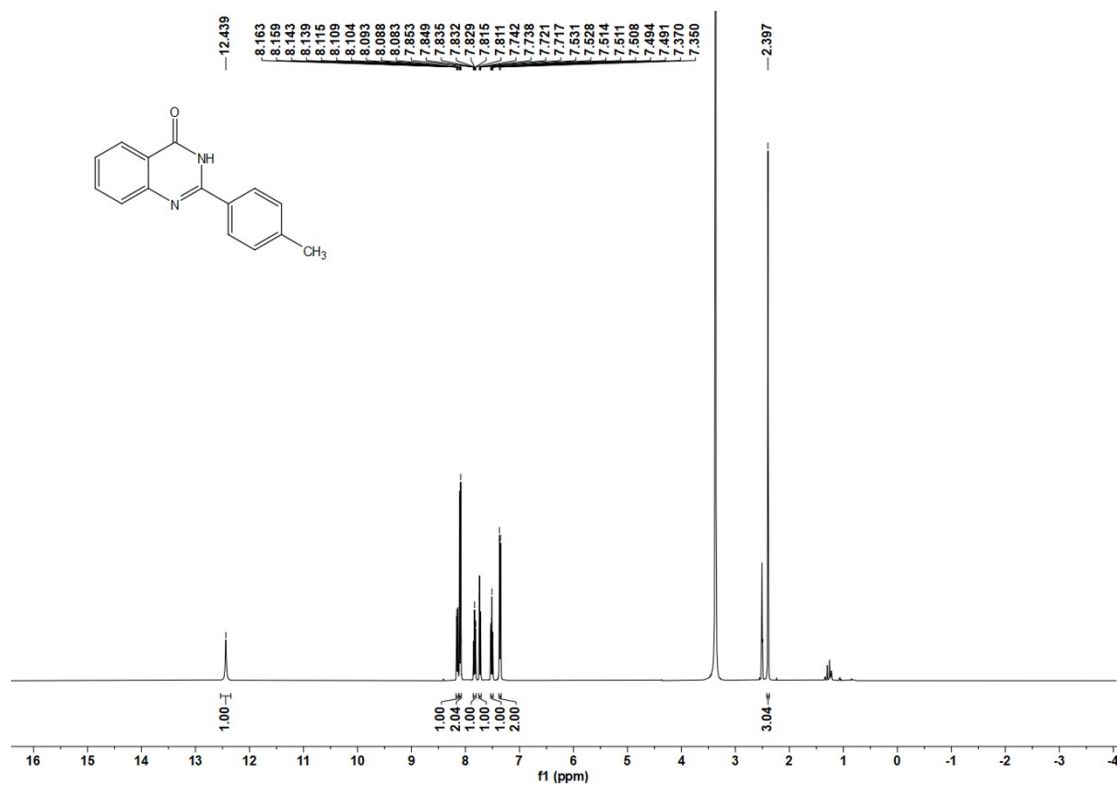
# 2-phenyl-1H-imidazo[4,5-b]pyridine (3at)



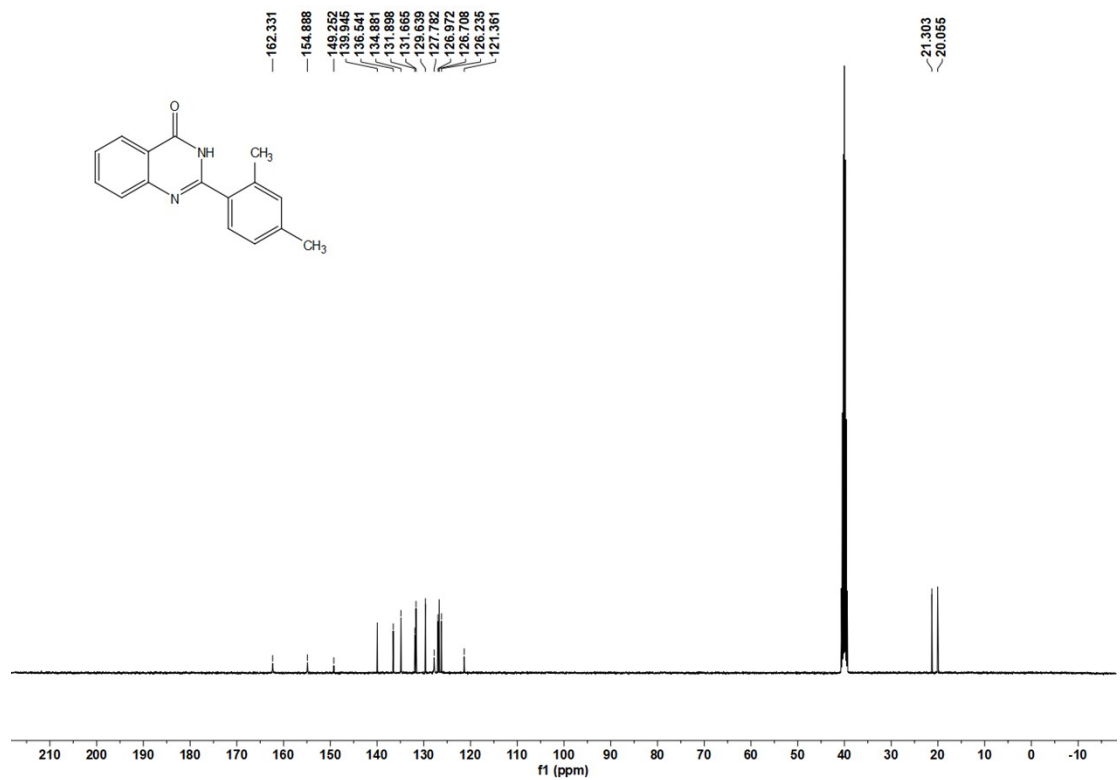
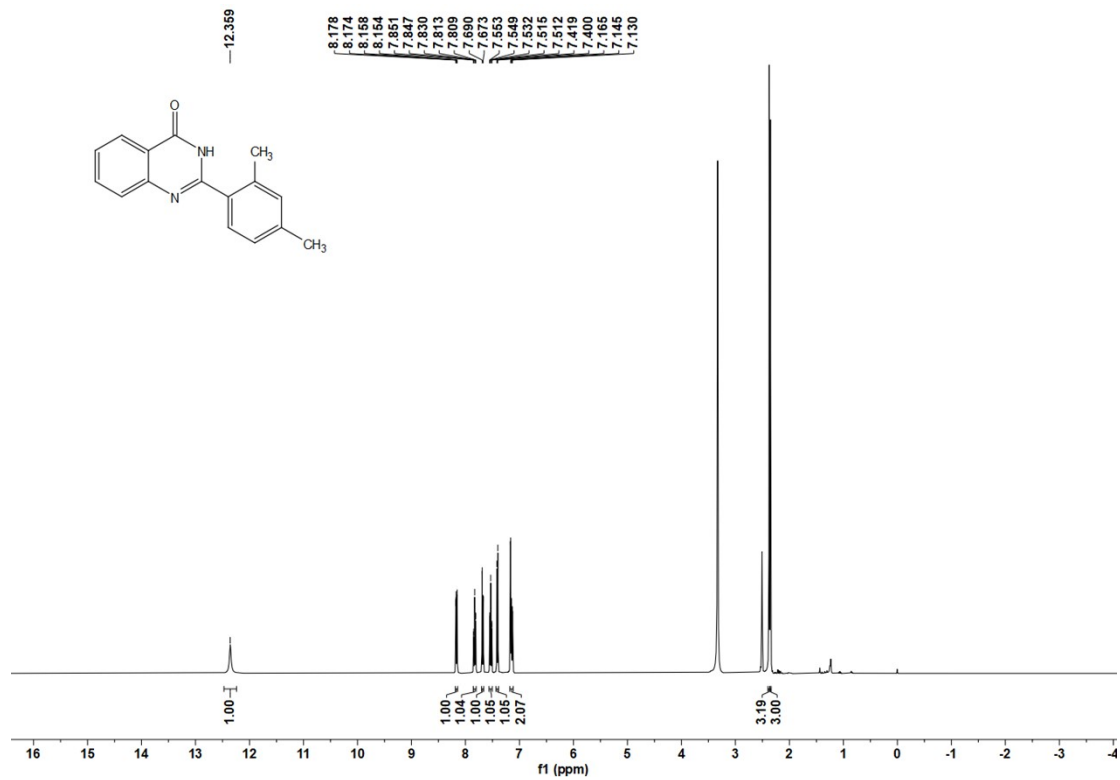
### 2-phenylquinazolin-4(3H)-one (5aa)



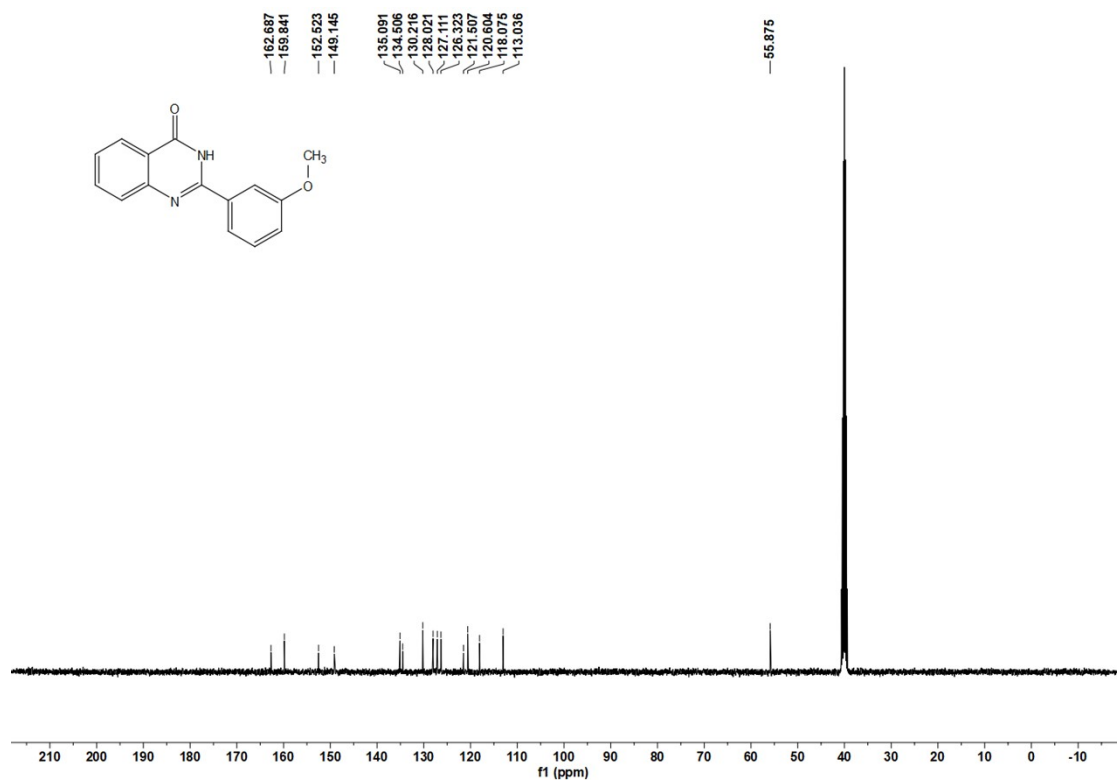
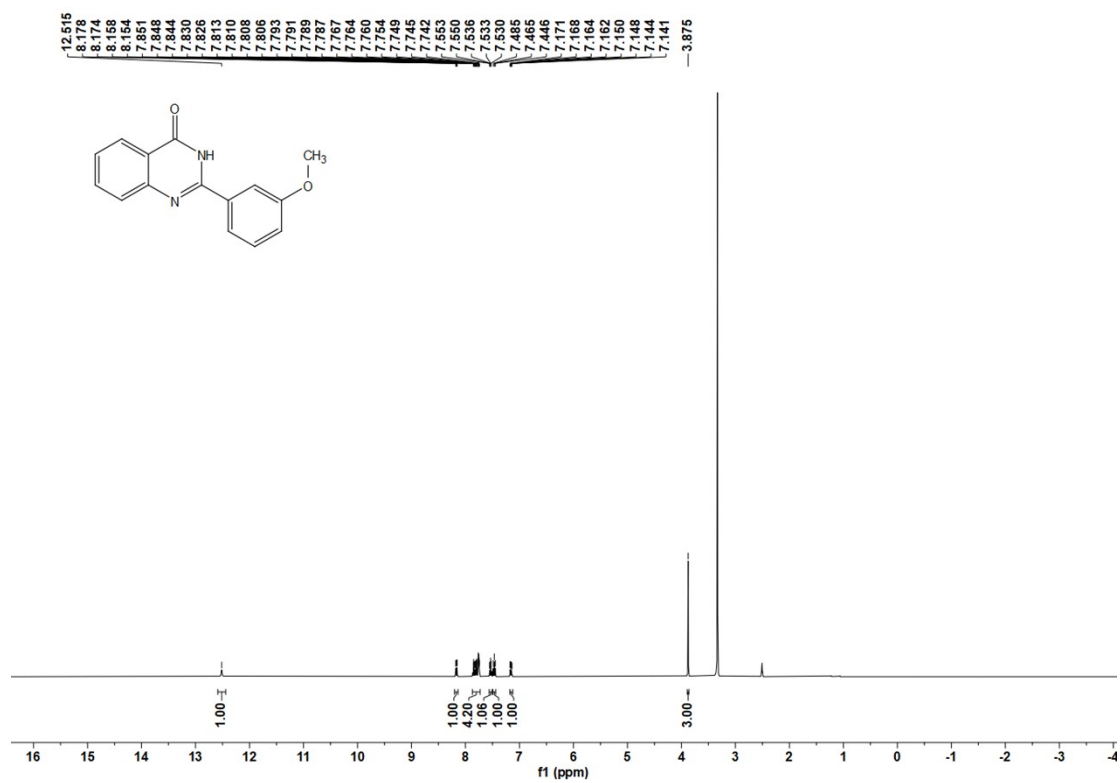
## 2-(p-tolyl)quinazolin-4(3H)-one (5ab)



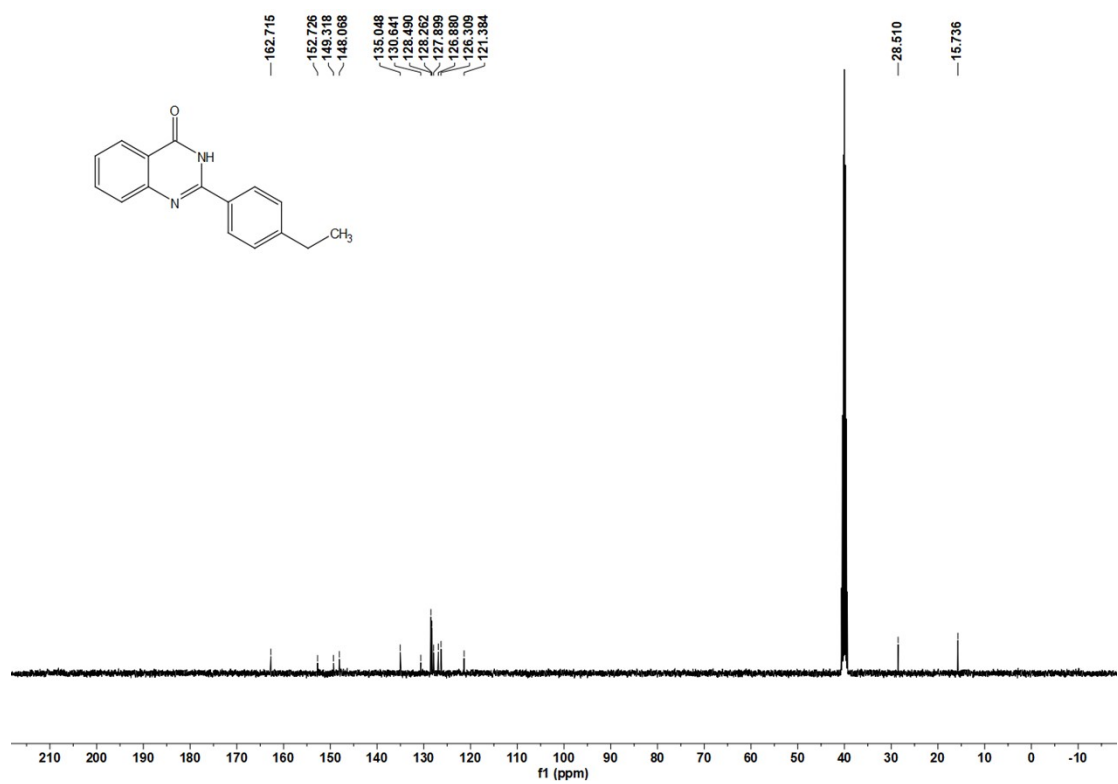
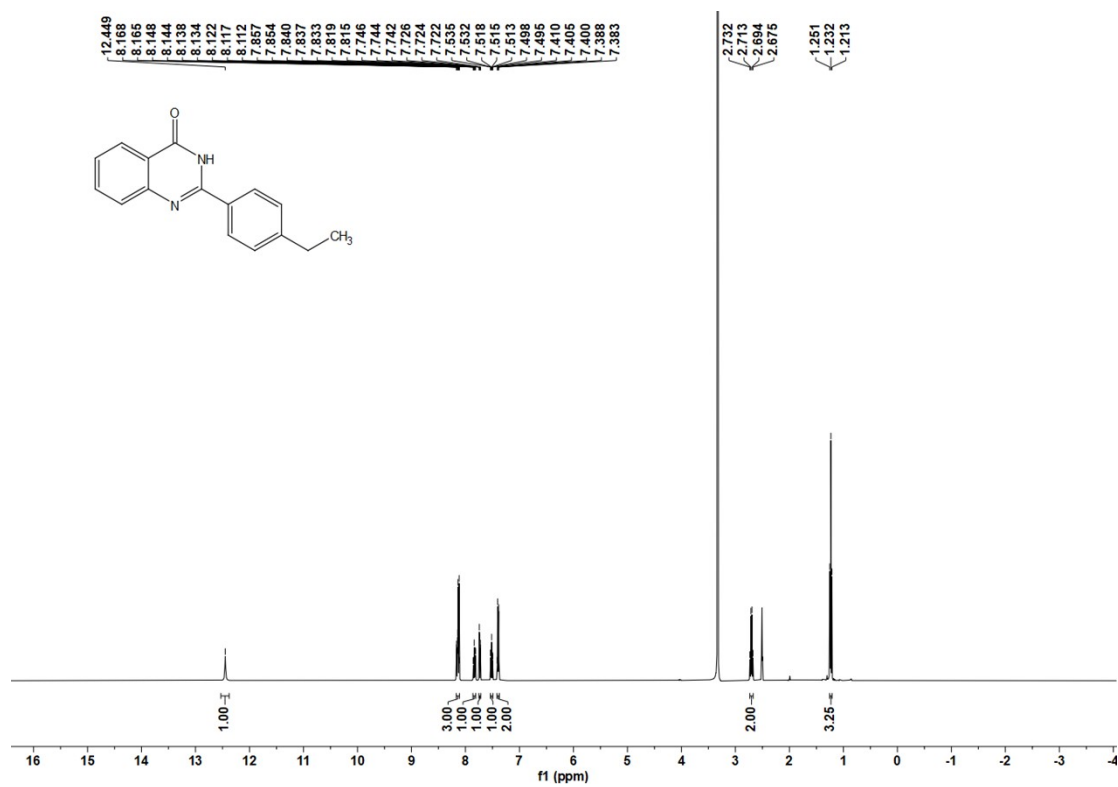
### 2-(2,4-dimethylphenyl)quinazolin-4(3H)-one (5ac)



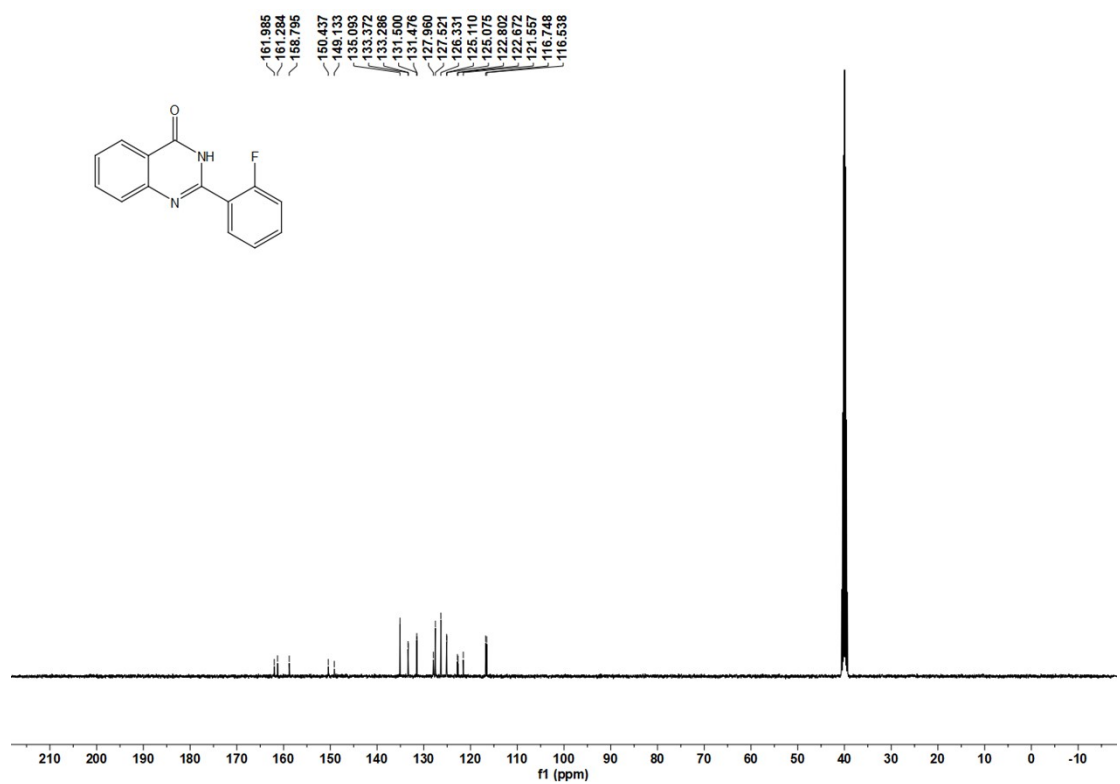
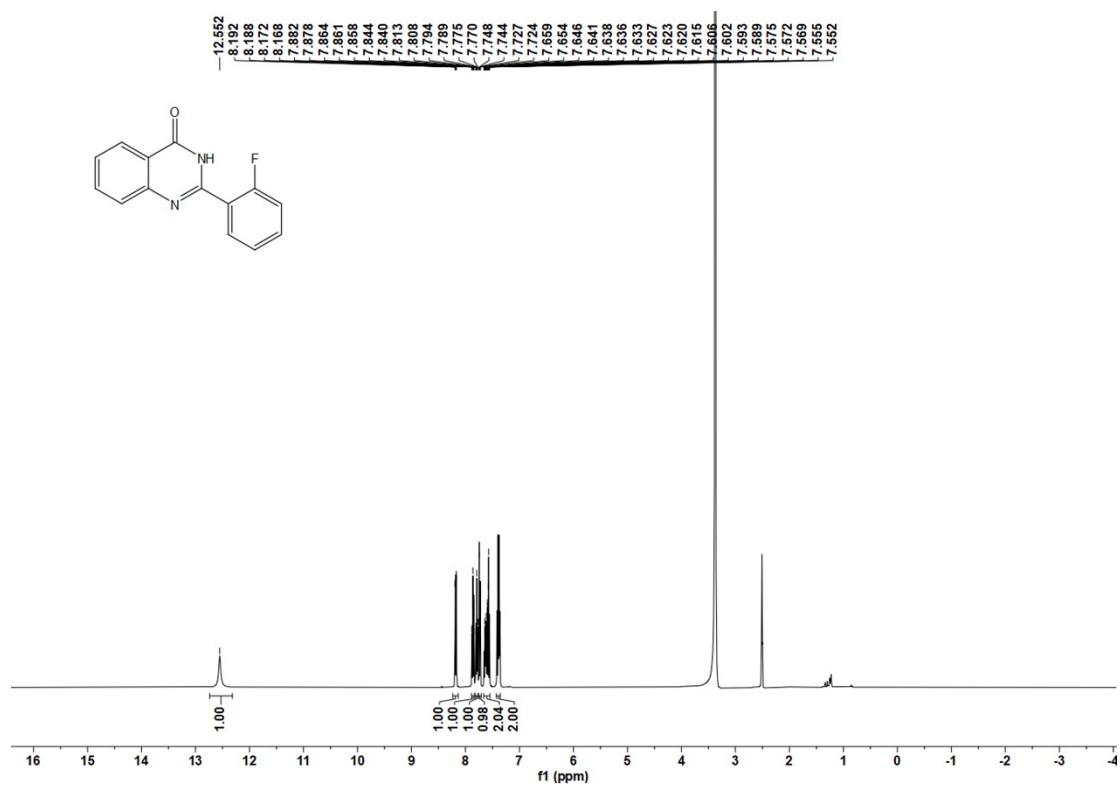
## 2-(3-methoxyphenyl)quinazolin-4(3H)-one (5ad)



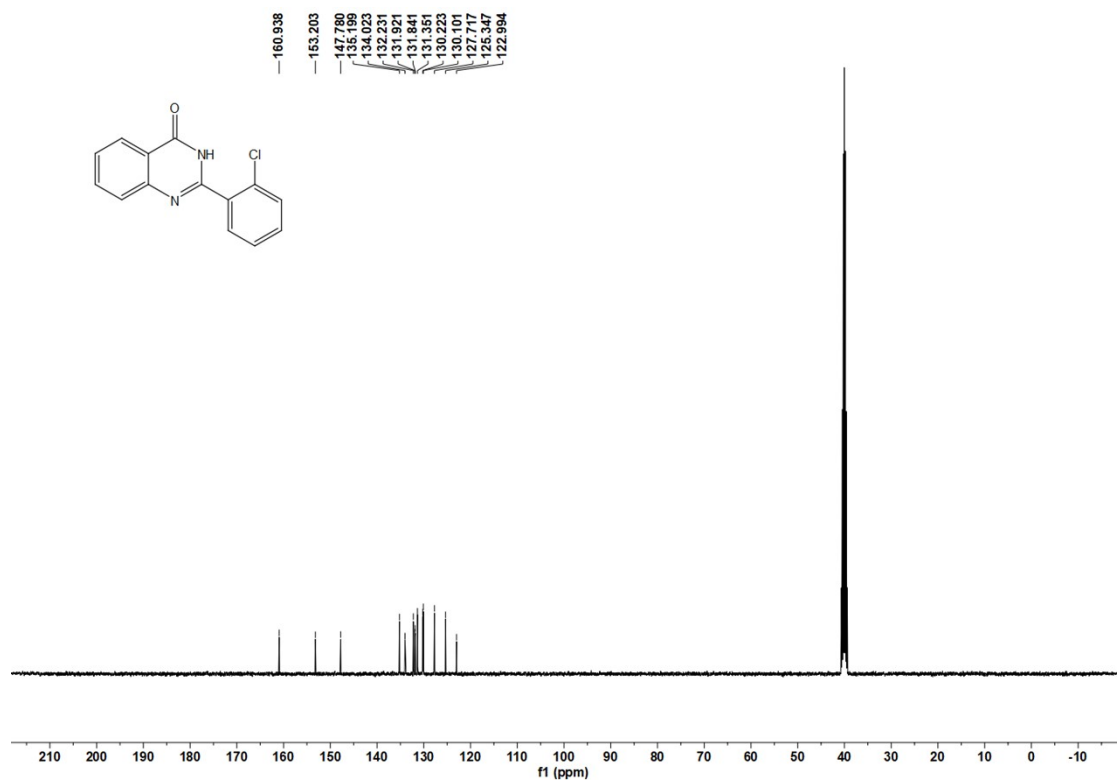
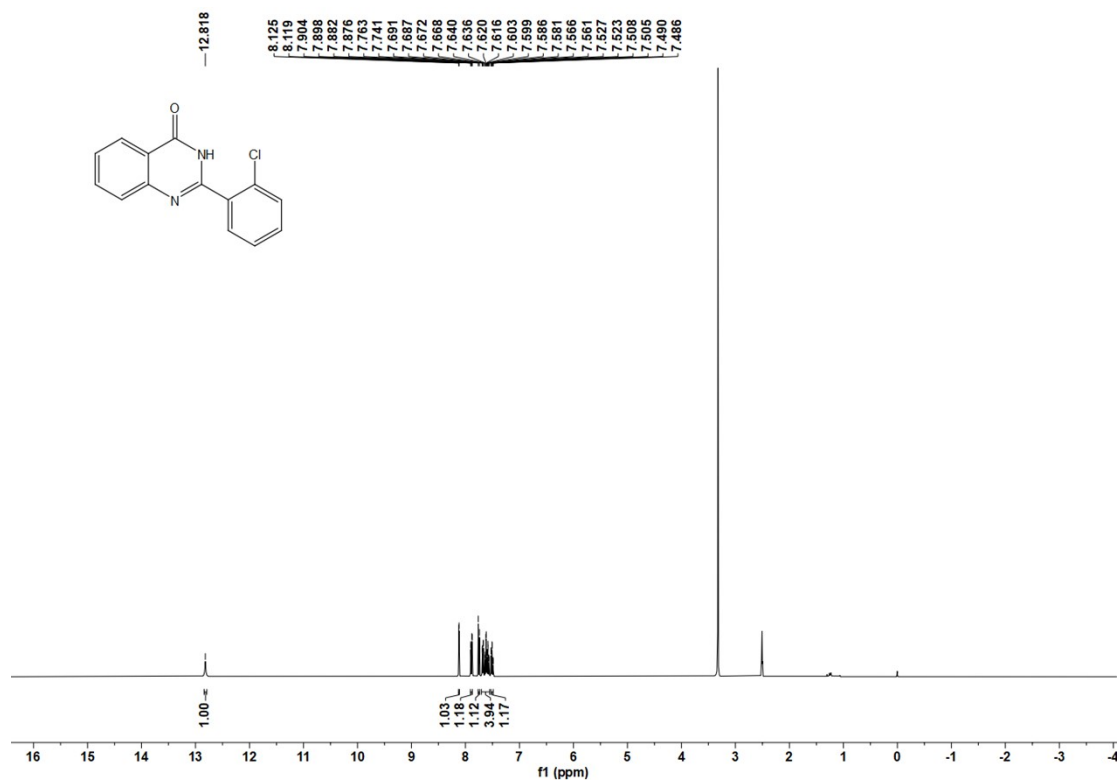
2-(4-ethylphenyl)quinazolin-4(3H)-one (5ac)



# 2-(2-fluorophenyl)quinazolin-4(3H)-one (5af)

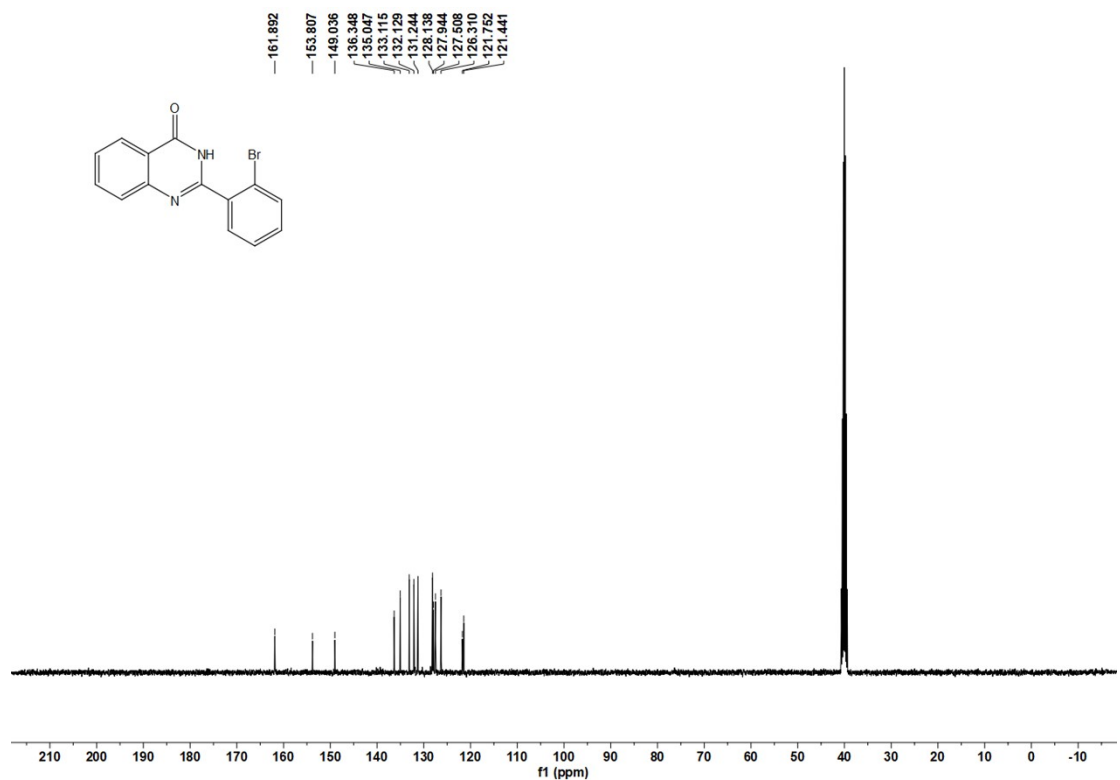
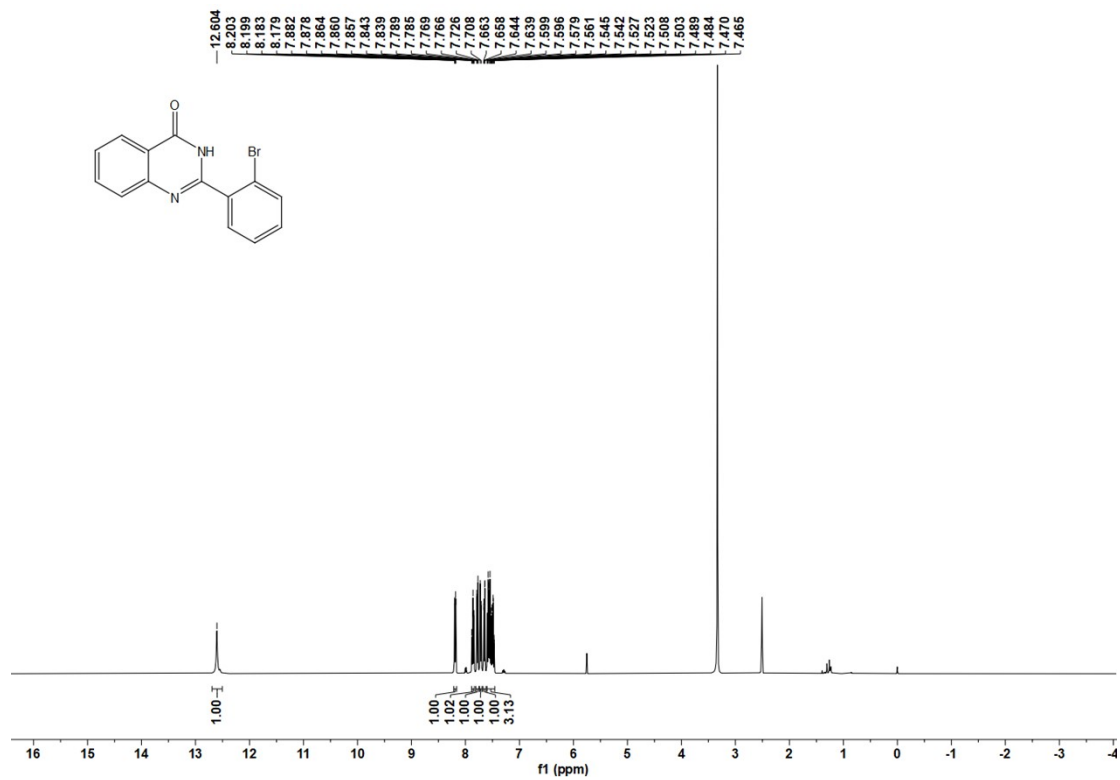


## 2-(2-chlorophenyl)quinazolin-4(3H)-one (5ag)

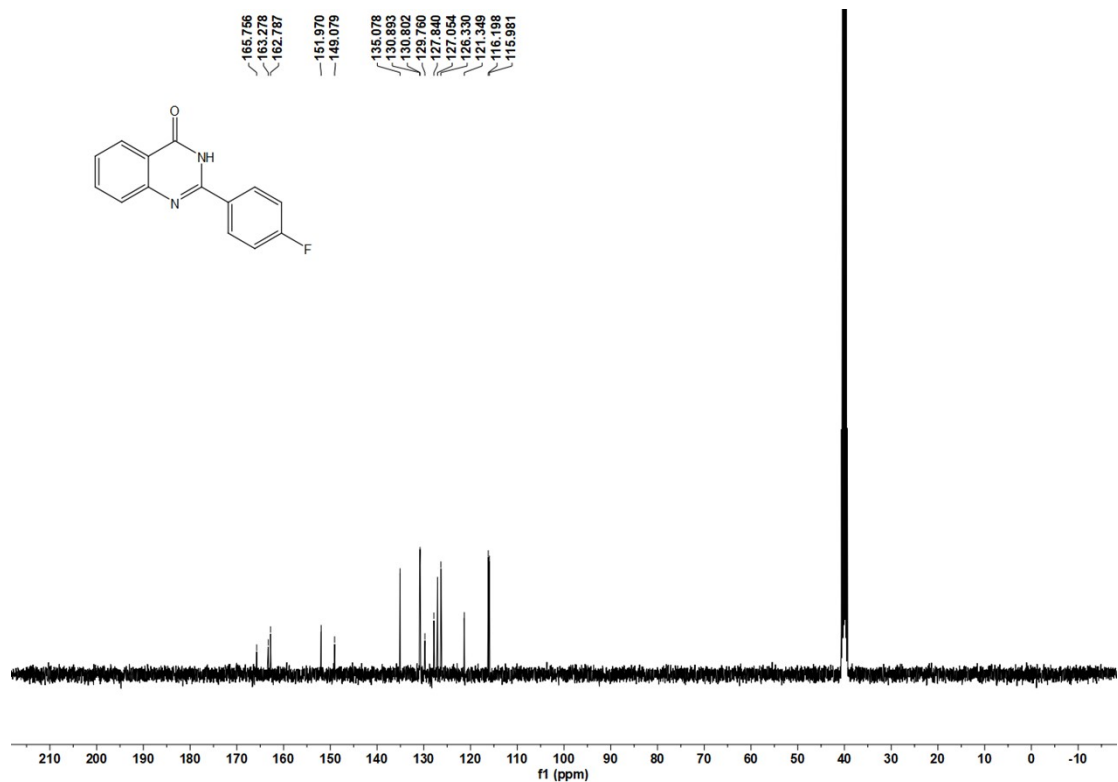
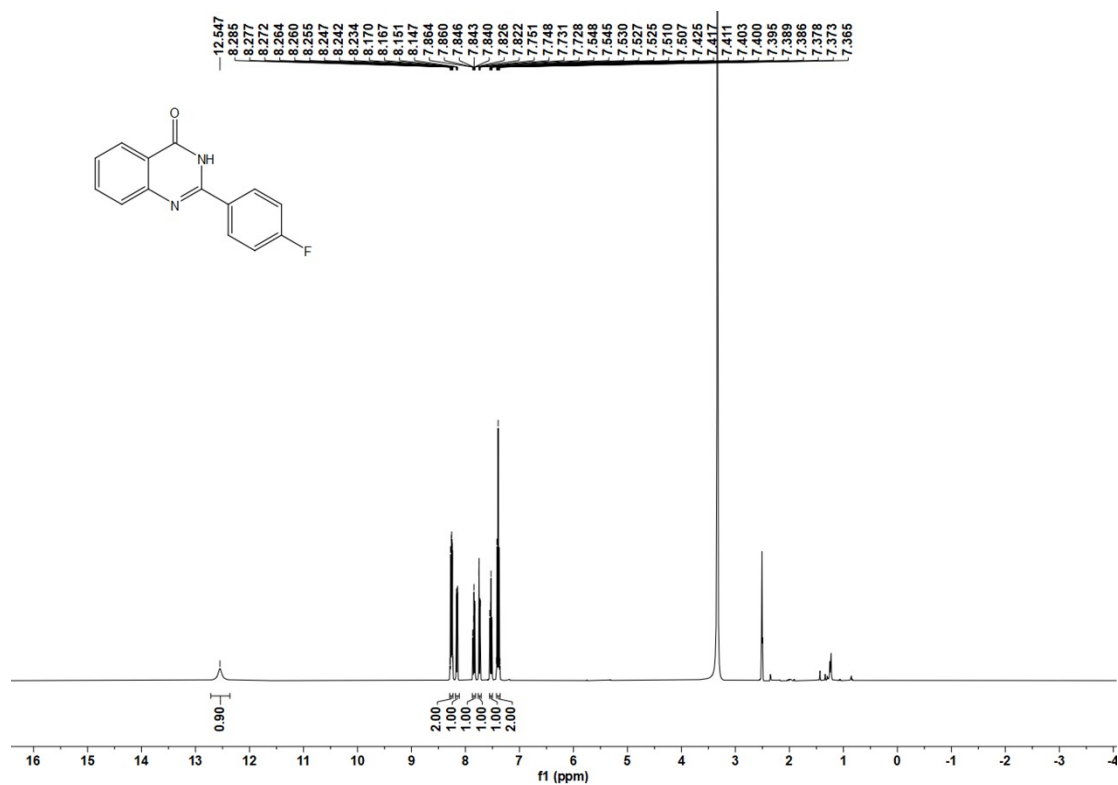




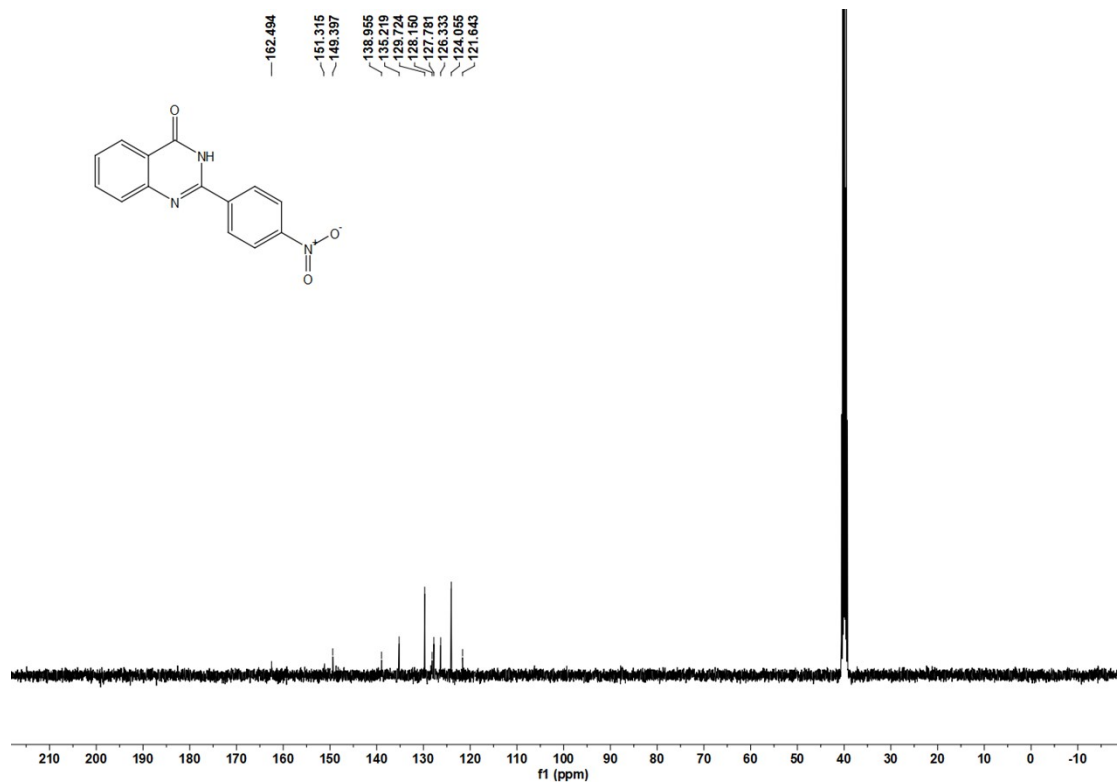
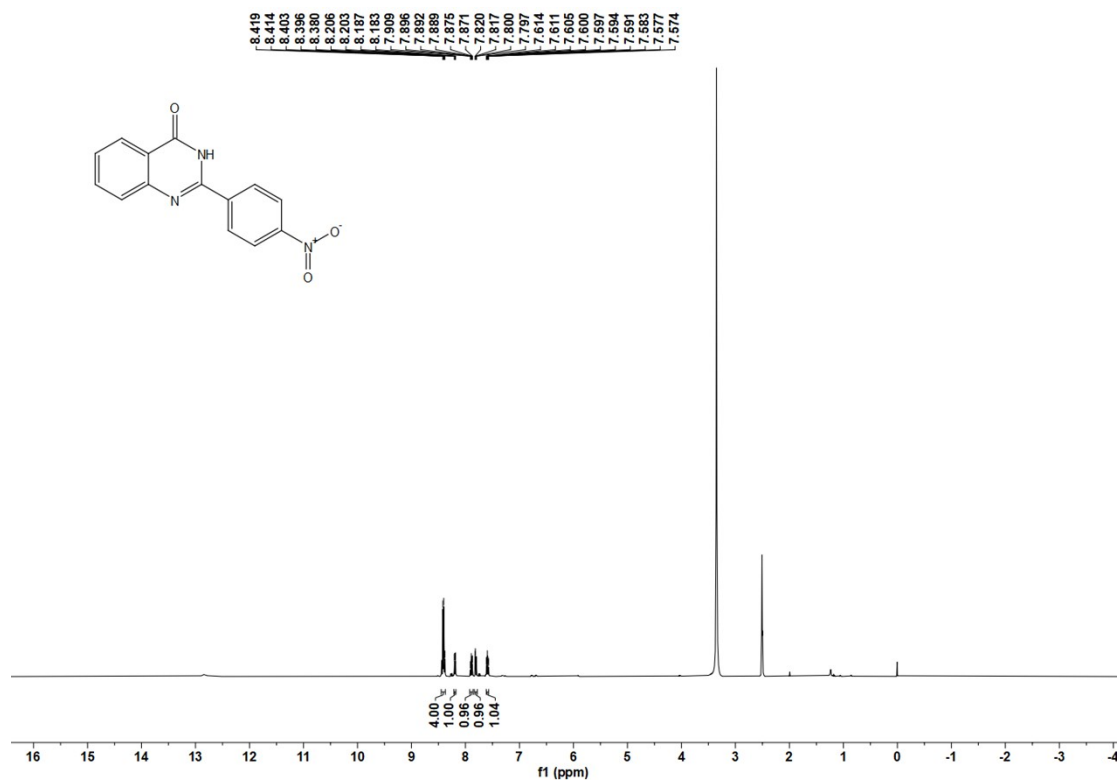
2-(2-bromophenyl)quinazolin-4(3H)-one (5ah)



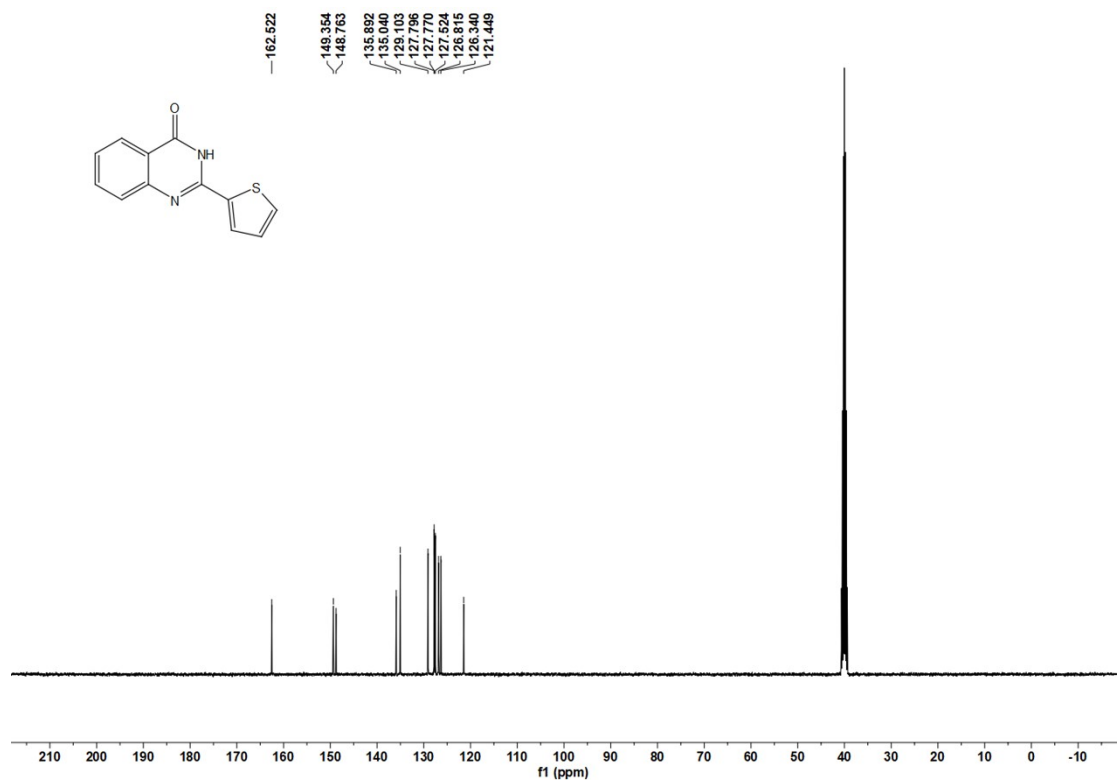
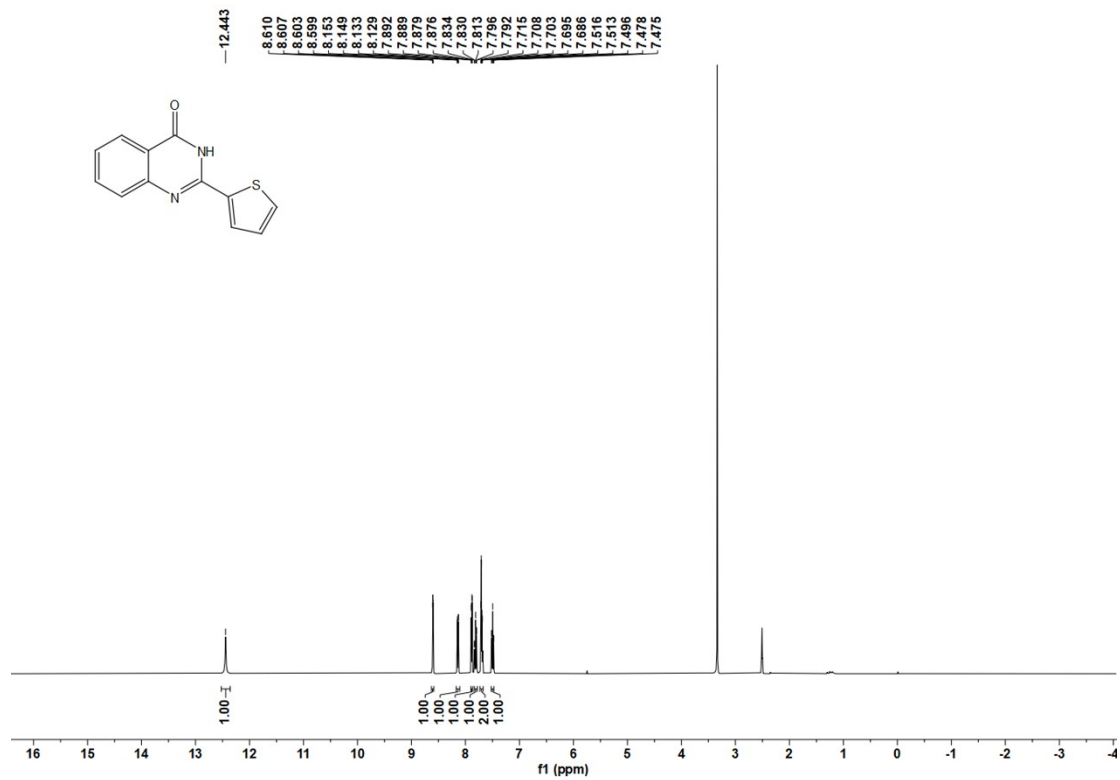
2-(4-fluorophenyl)quinazolin-4(3H)-one (5ai)



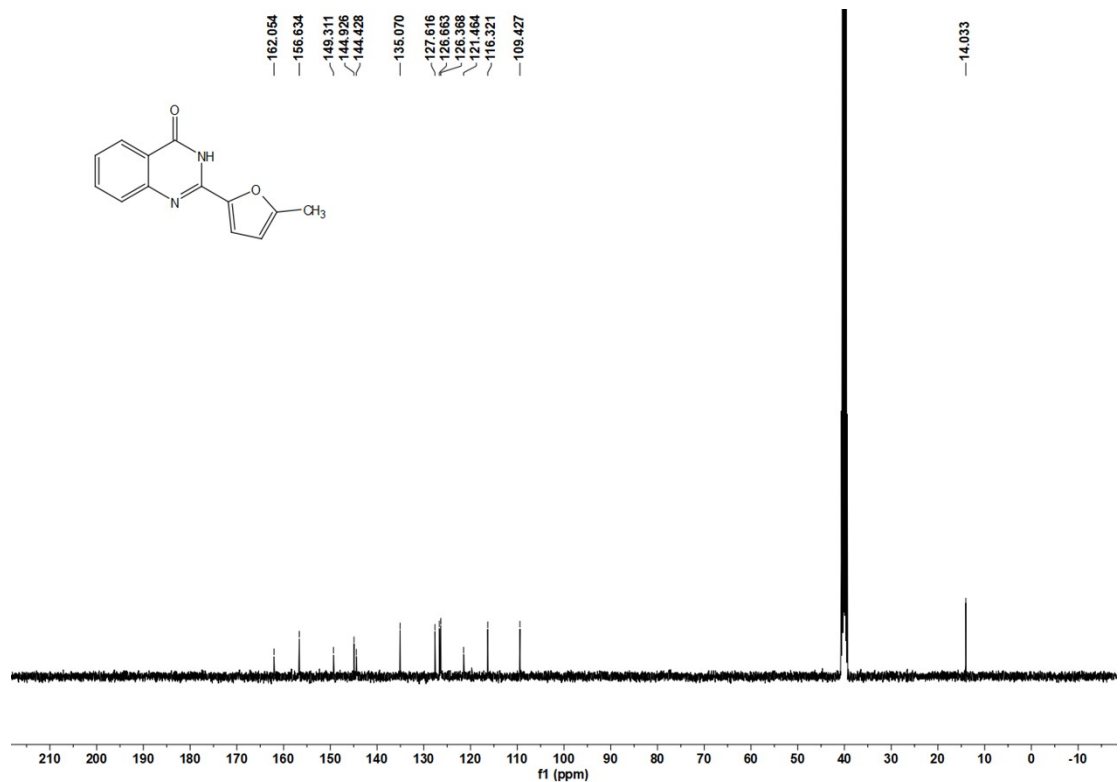
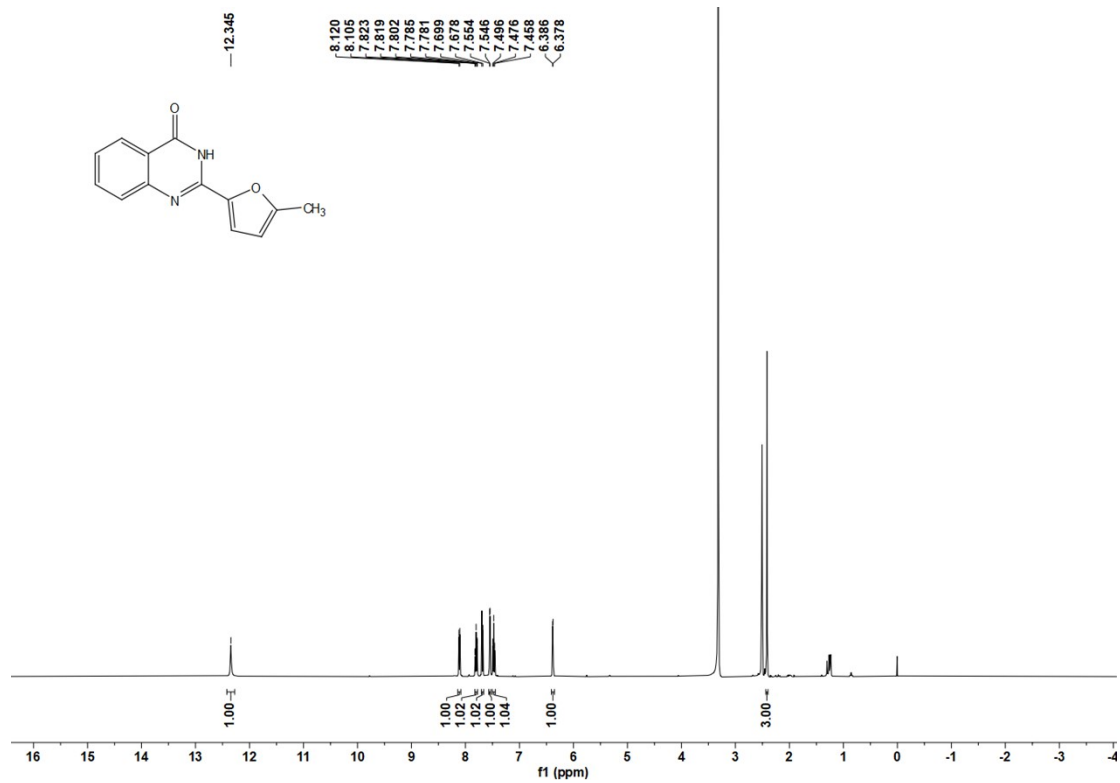
## 2-(4-nitrophenyl)quinazolin-4(3H)-one (5aj)



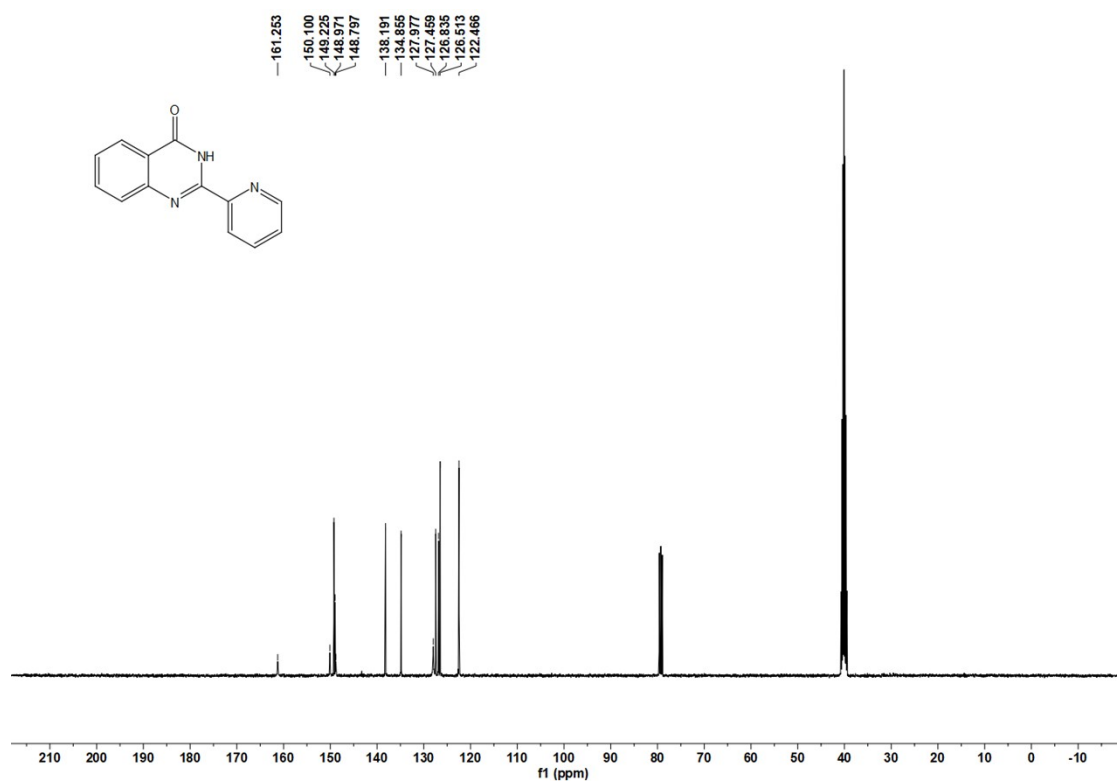
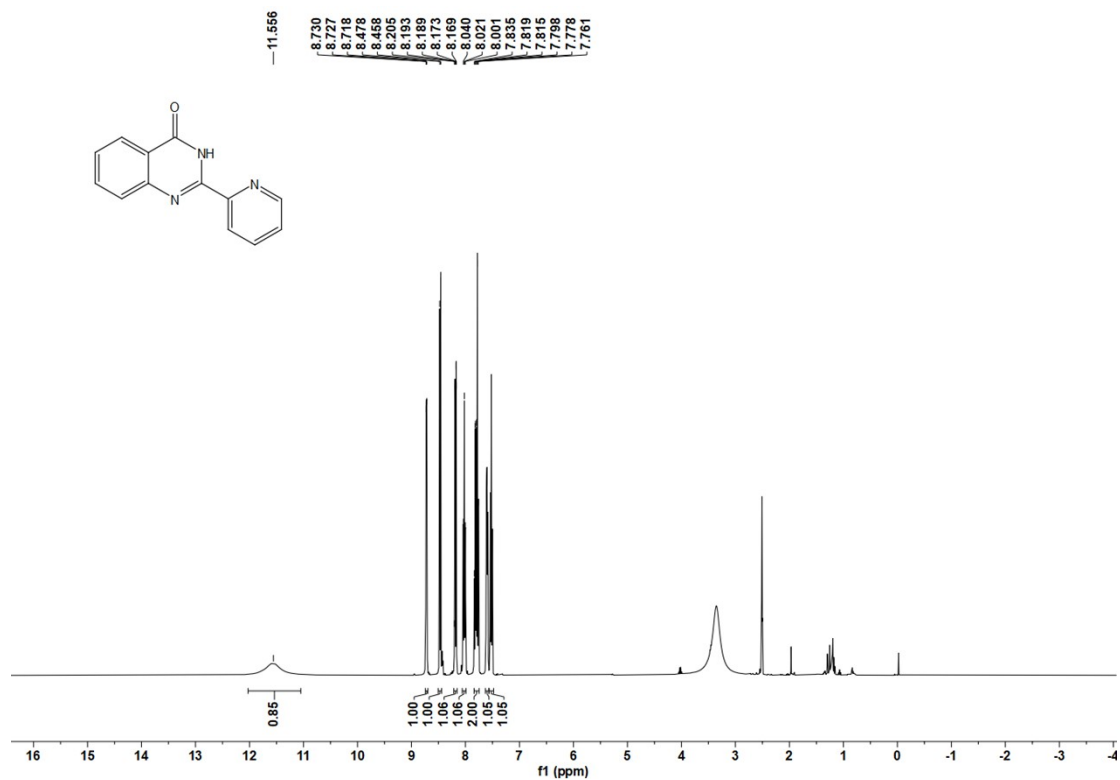
2-(thiophen-2-yl)quinazolin-4(3H)-one (5ak)



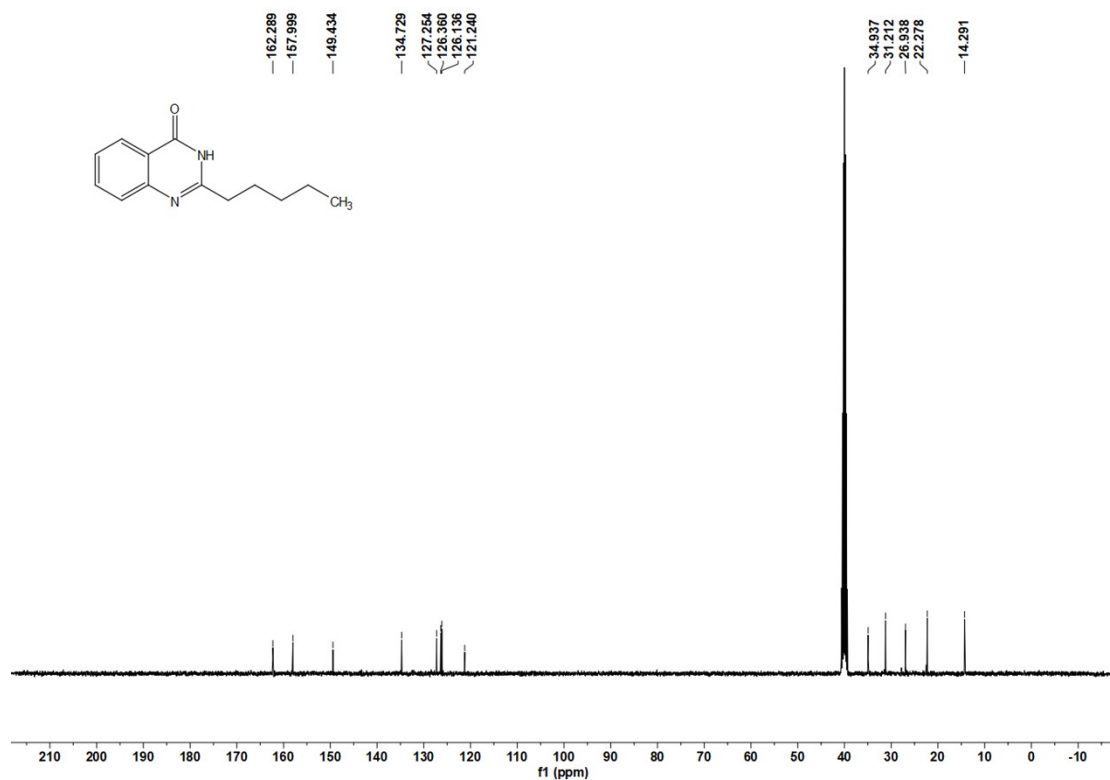
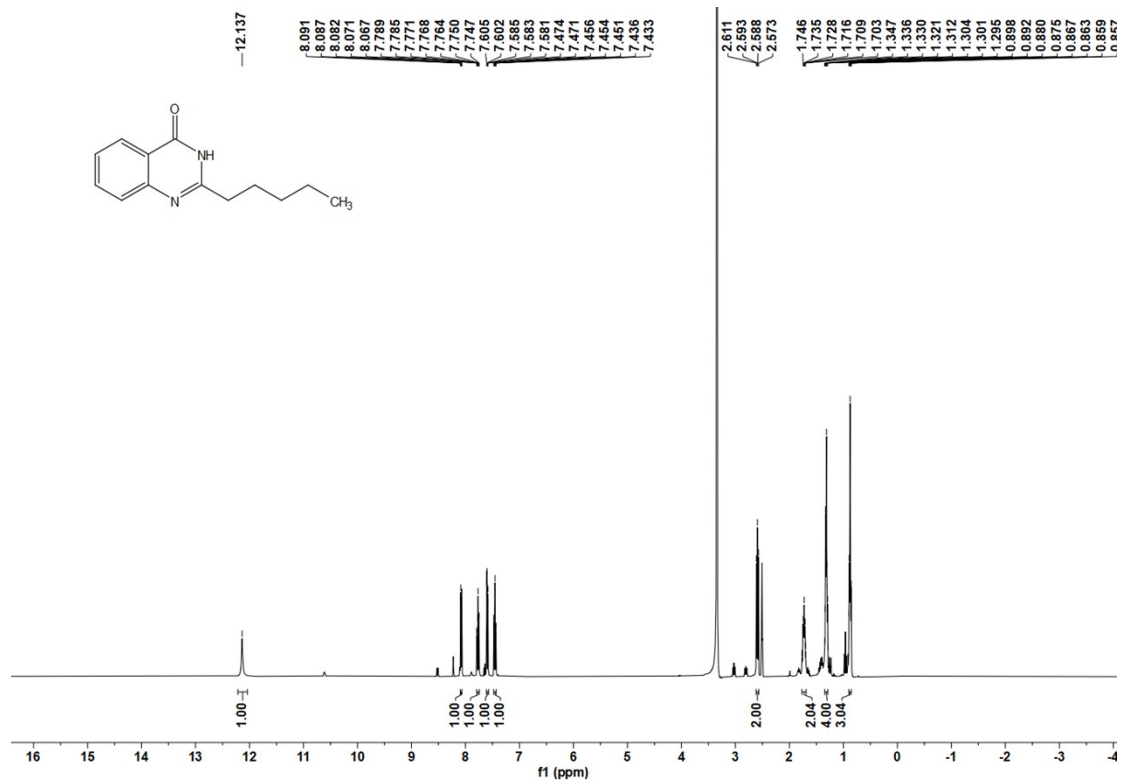
### 2-(5-methylfuran-2-yl)quinazolin-4(3H)-one (5a)



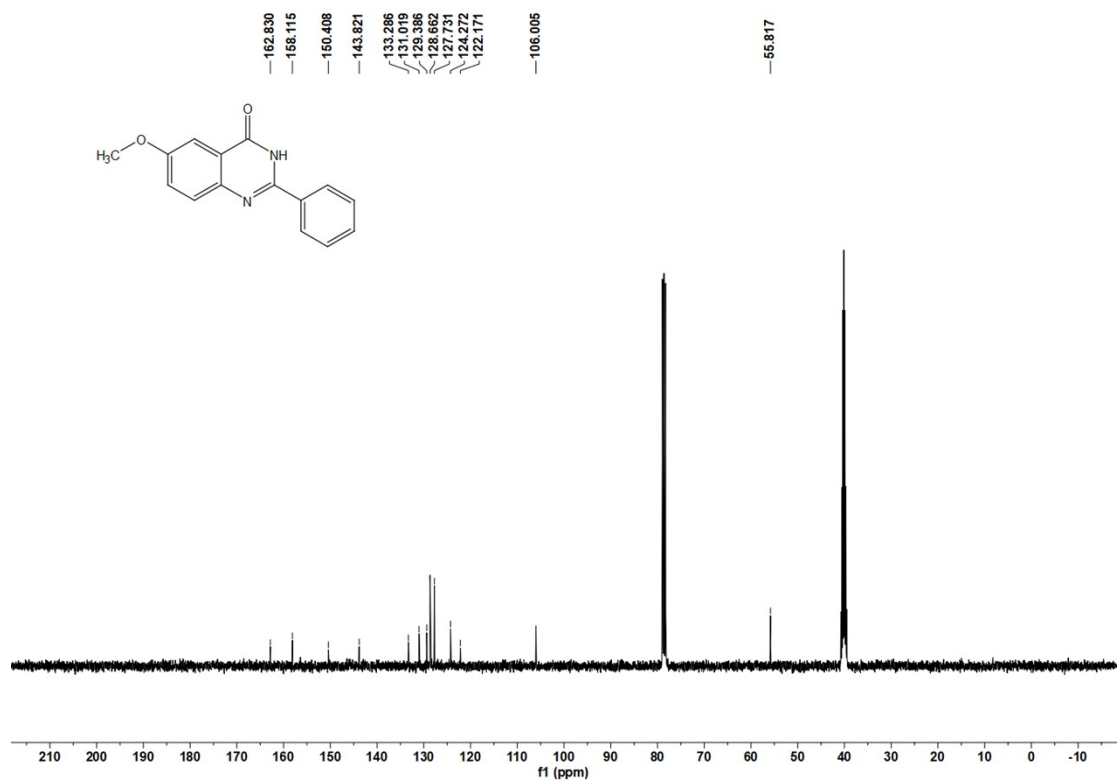
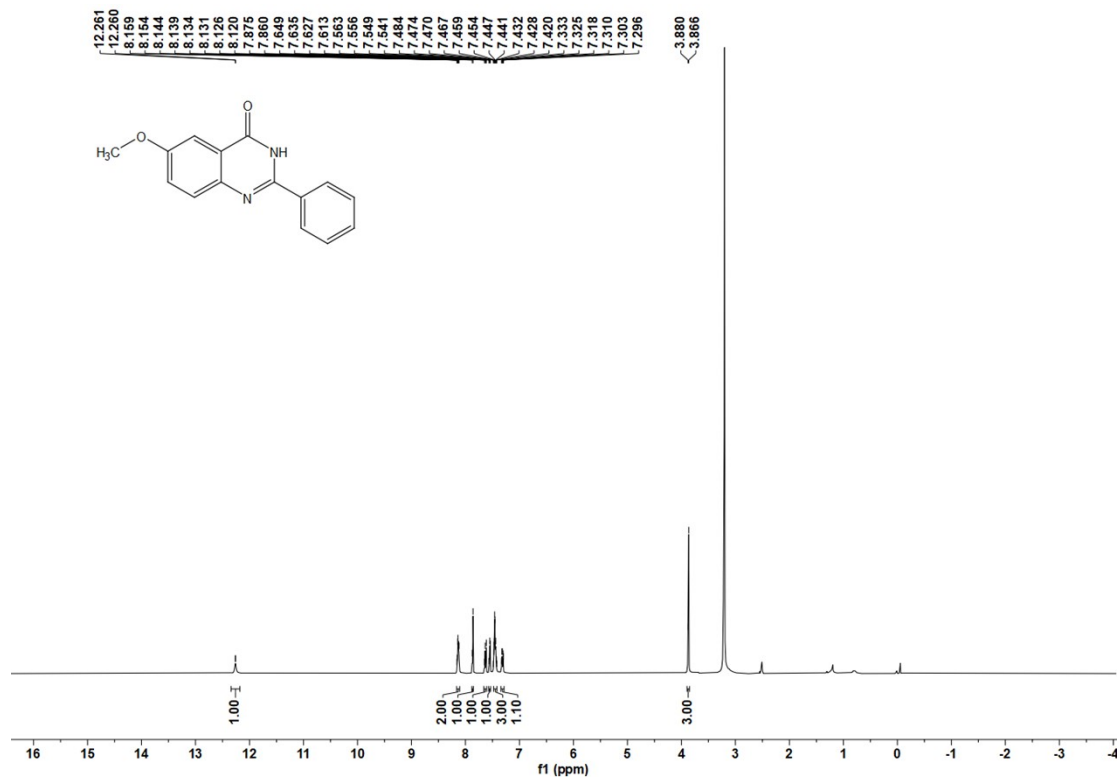
# 2-(pyridin-2-yl)quinazolin-4(3H)-one (5am)



## 2-pentylquinazolin-4(3H)-one (5an)

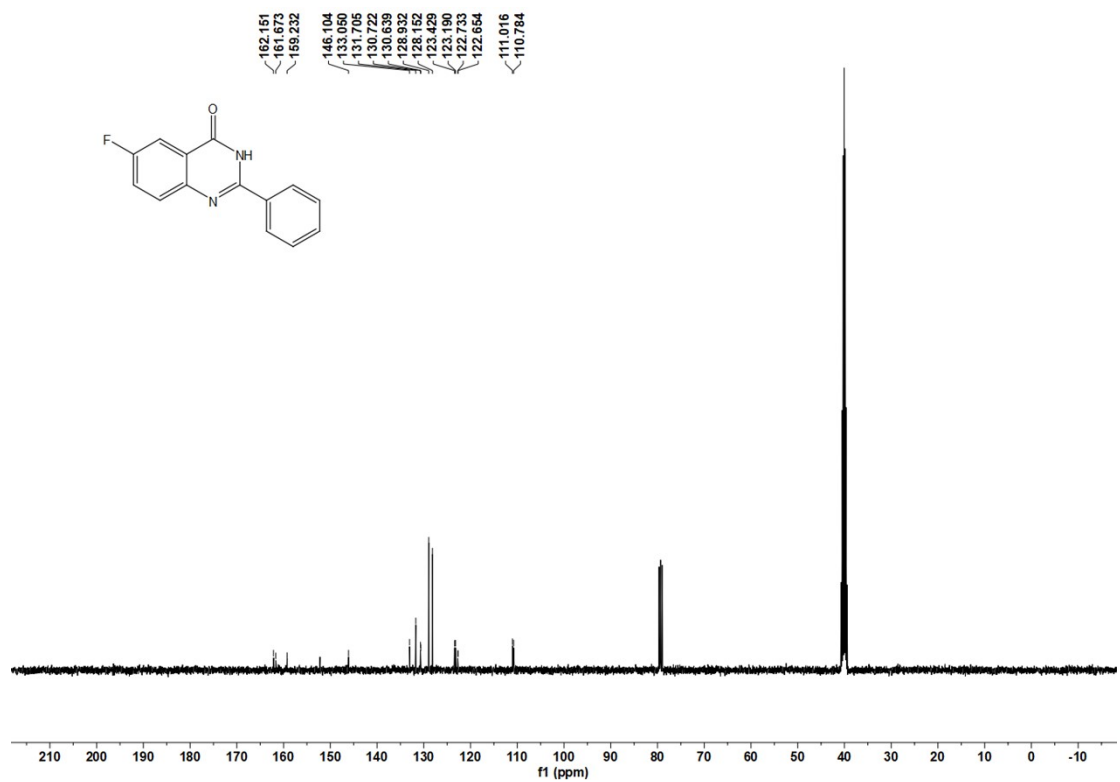
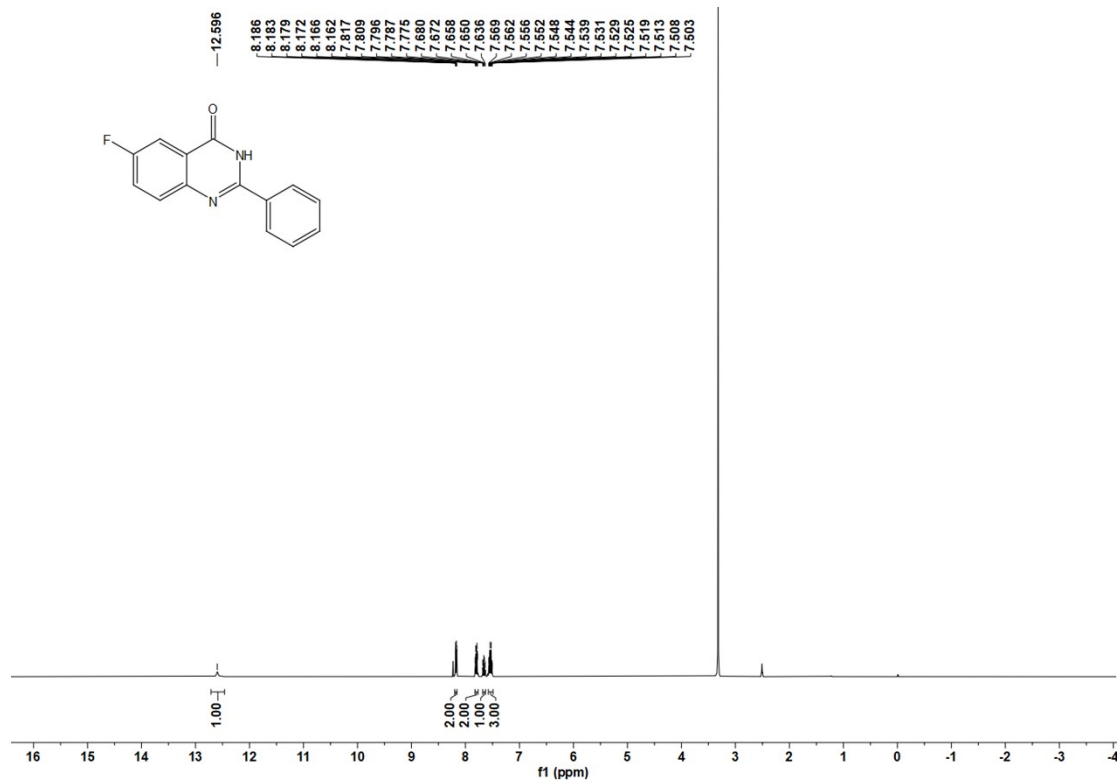


**6-methoxy-2-phenylquinazolin-4(3H)-one (5ao)**

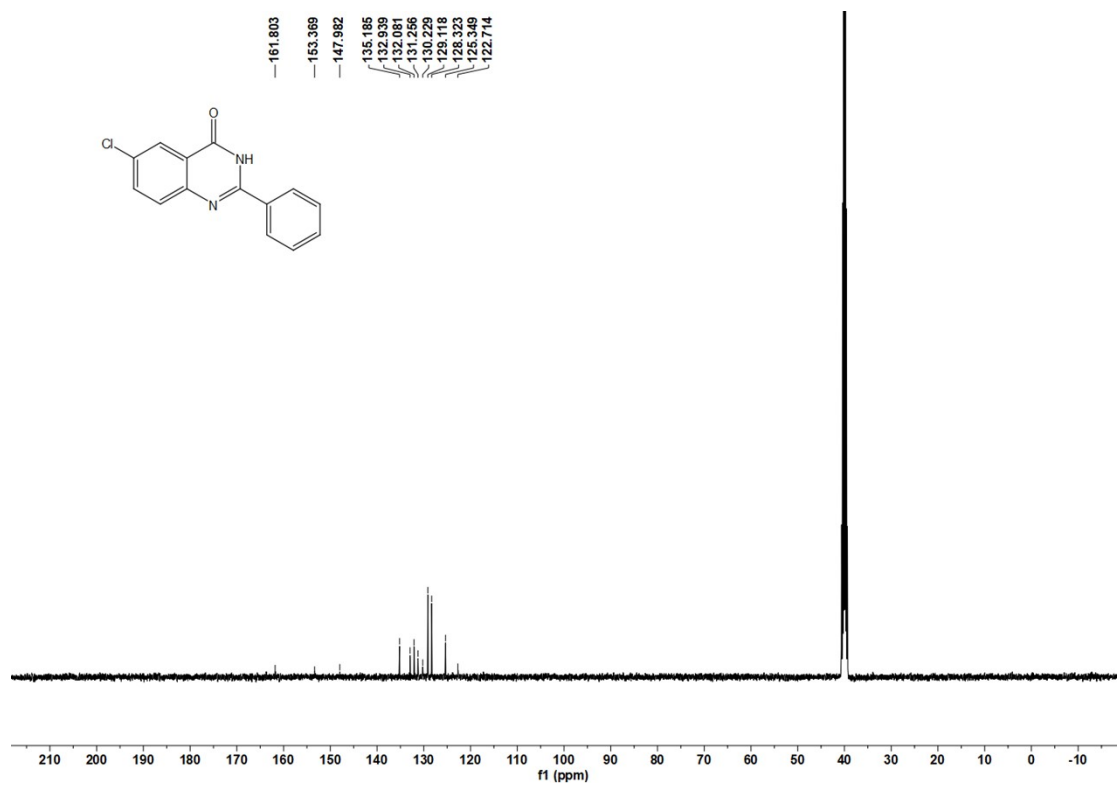
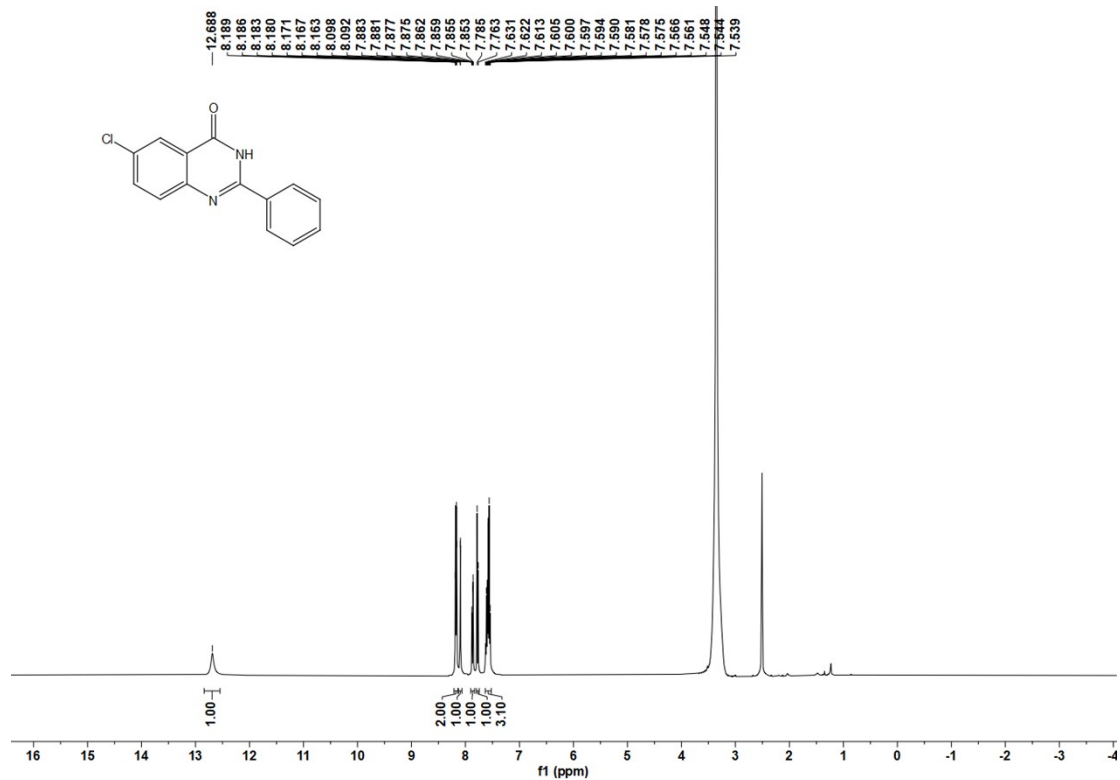




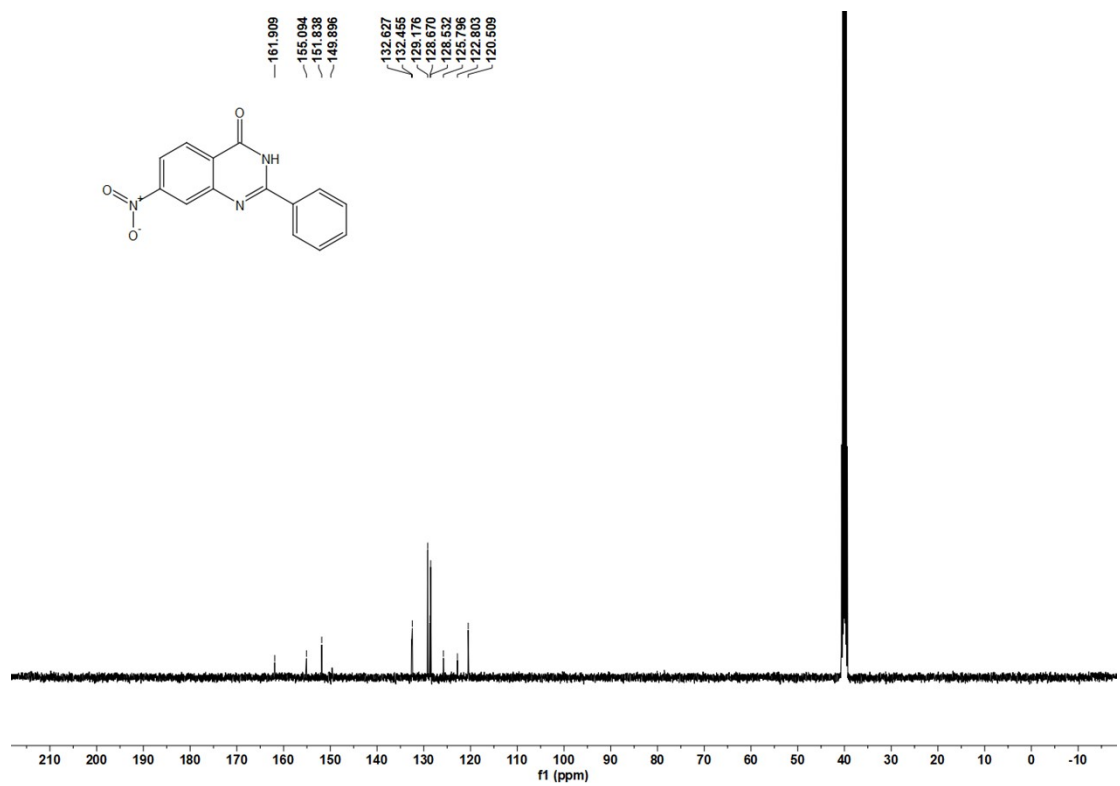
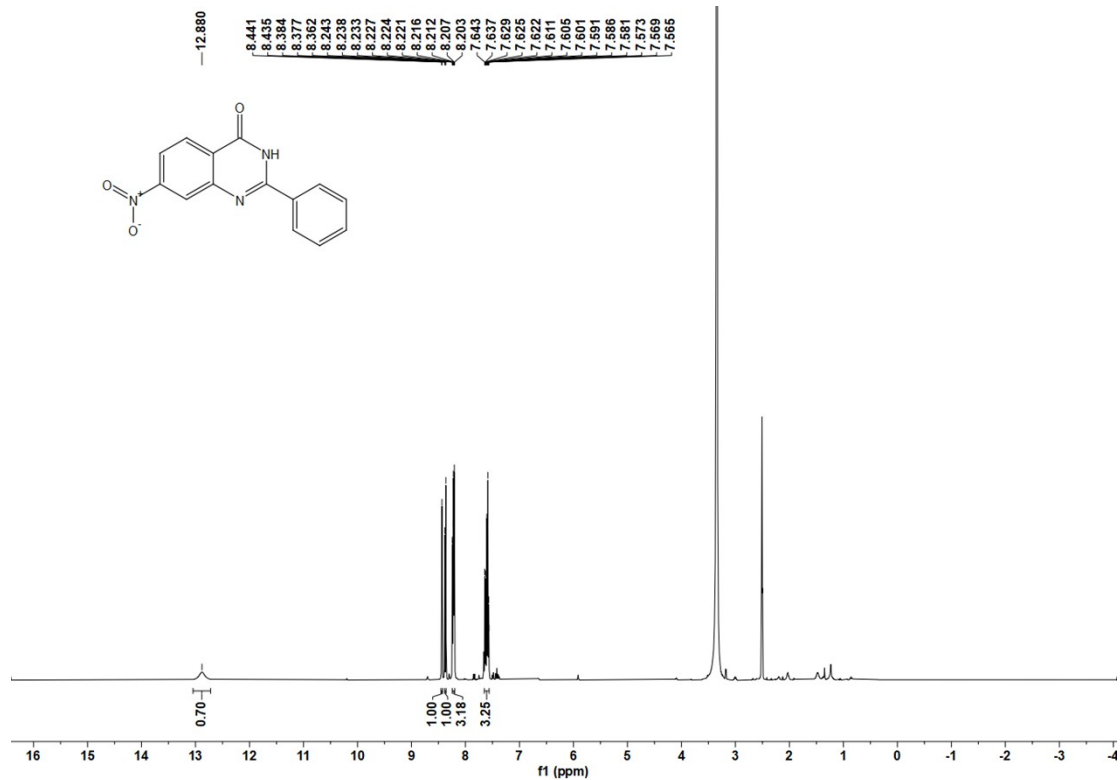
# 6-fluoro-2-phenylquinazolin-4(3H)-one (5ap)



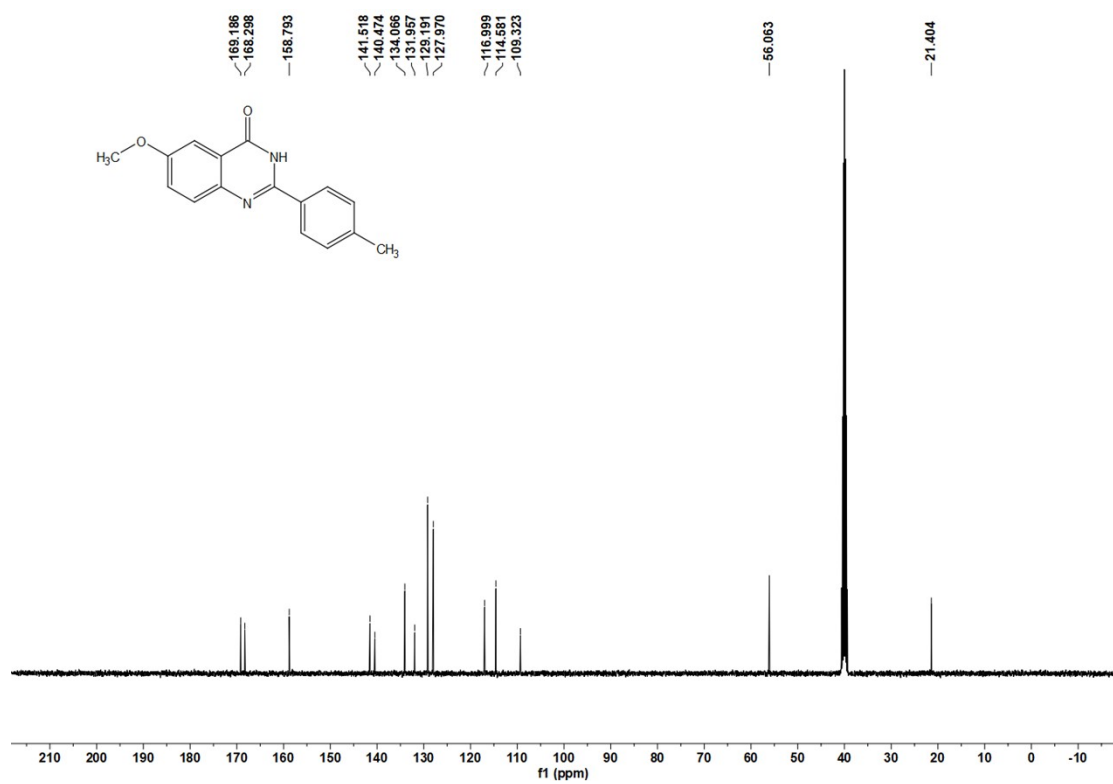
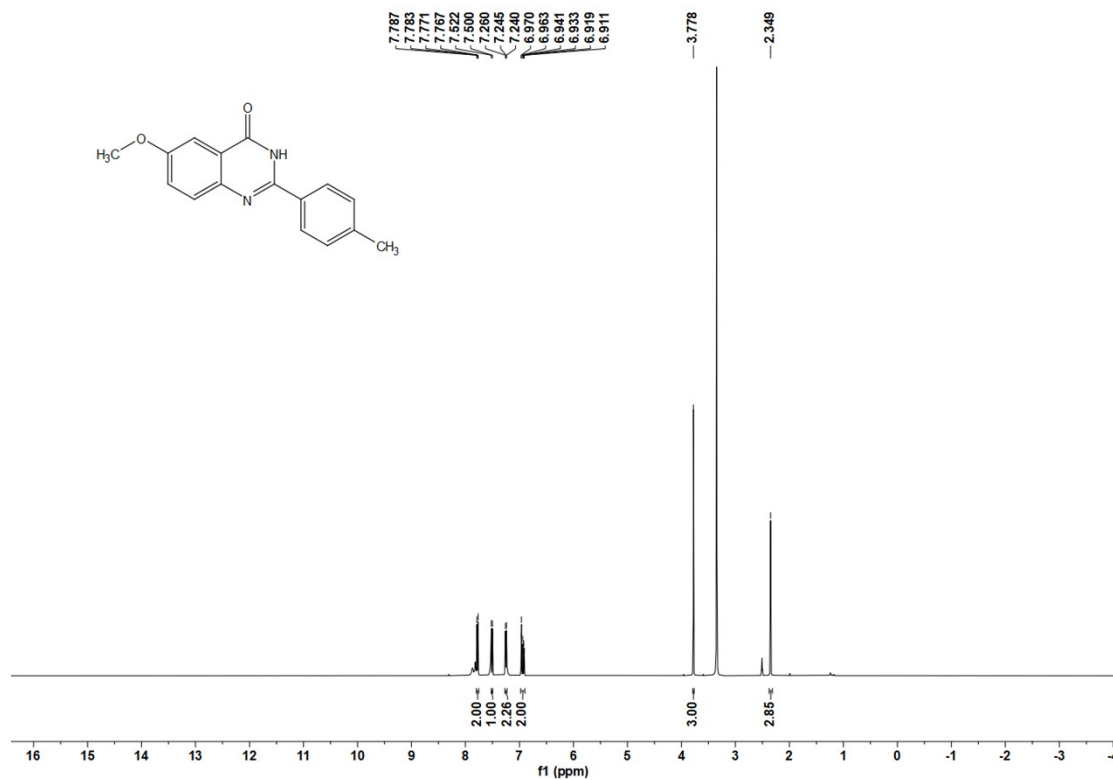
### 6-chloro-2-phenylquinazolin-4(3H)-one (5aq)



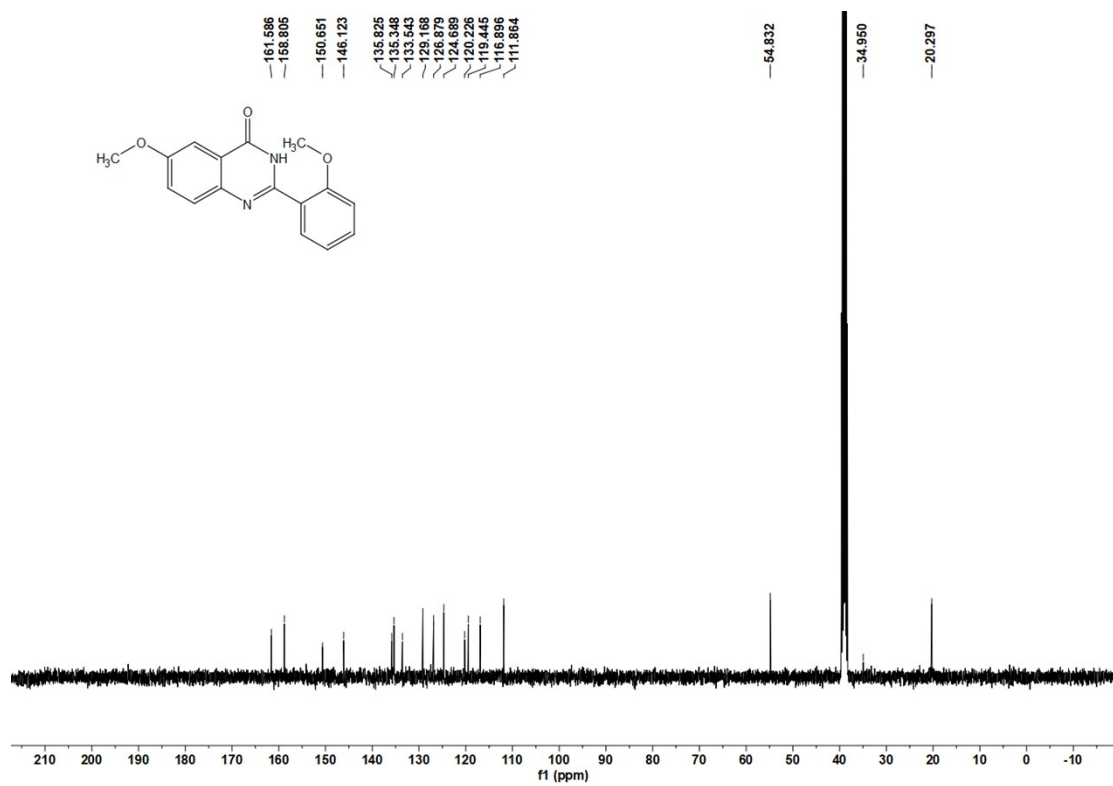
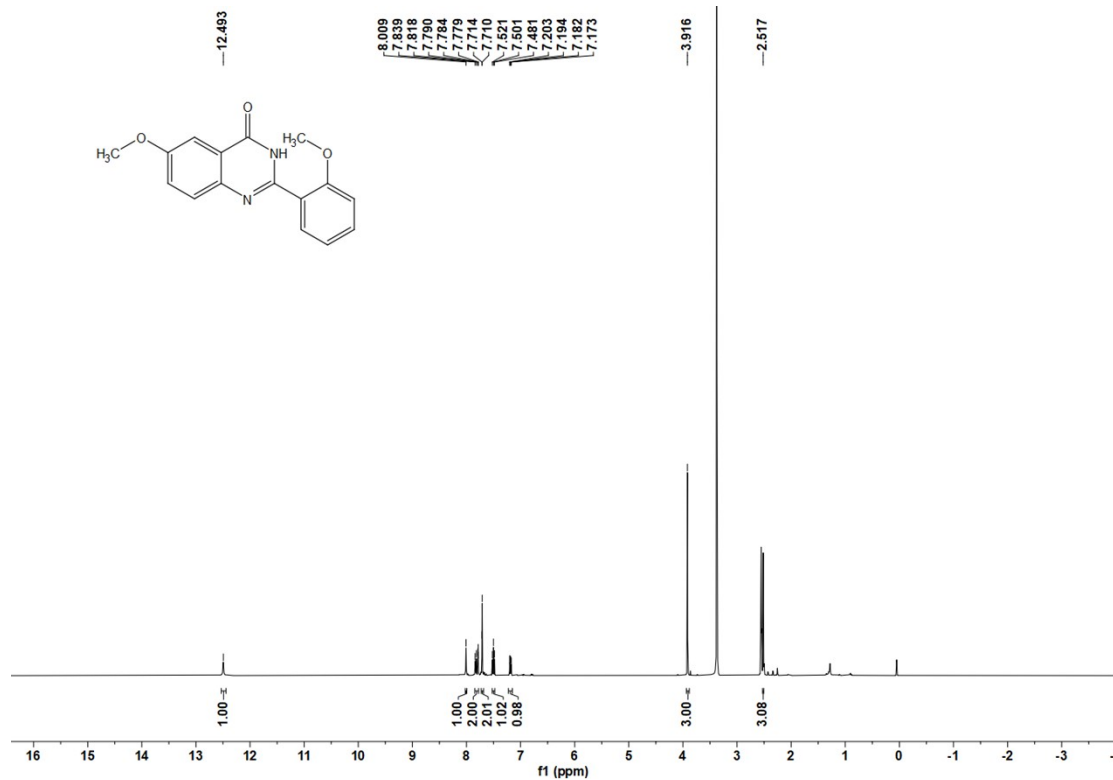
### 7-nitro-2-phenylquinazolin-4(3H)-one (5ar)



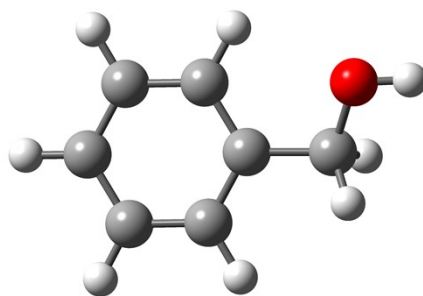
### 6-methoxy-2-(p-tolyl)quinazolin-4(3H)-one (5as)



### 2-(2-methoxyphenyl)-6-methylquinazolin-4(3H)-one (5at)



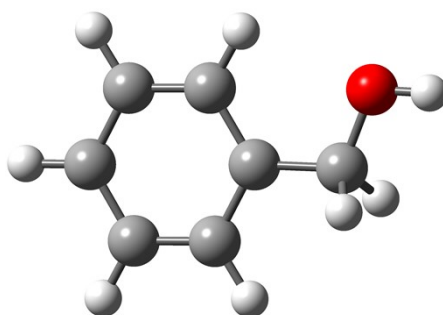
## 7. Optimized Structures and Cartesian Coordinates



2a / C<sub>6</sub>H<sub>5</sub>CH<sub>2</sub>OH

Energies= -346.893050 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | -2.31173600 | -0.31320300 | -0.04210600 |
| C | -1.37600200 | -1.34799300 | 0.03678400  |
| C | -0.00830200 | -1.06271900 | 0.09262600  |
| C | 0.43851200  | 0.26389600  | 0.06579600  |
| C | -0.50436200 | 1.29723800  | -0.02318600 |
| C | -1.87106000 | 1.01347000  | -0.07171400 |
| H | -3.37460600 | -0.53736000 | -0.08587000 |
| H | -1.70984200 | -2.38275300 | 0.05348500  |
| H | 0.72000700  | -1.86537100 | 0.14570400  |
| H | -0.16837500 | 2.33242200  | -0.05809100 |
| H | -2.58956500 | 1.82650300  | -0.14181500 |
| C | 1.90909900  | 0.60134600  | 0.17378100  |
| H | 2.15189500  | 0.85398500  | 1.21897200  |
| H | 2.12921400  | 1.48769900  | -0.44014300 |
| O | 2.69464300  | -0.51286100 | -0.25182000 |
| H | 3.62723700  | -0.32444900 | -0.06956600 |

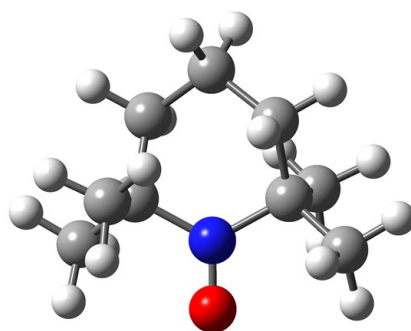


A / C<sub>7</sub>H<sub>8</sub>O<sup>+</sup>

Energies=-346.659827 a.u

|   |             |            |             |
|---|-------------|------------|-------------|
| C | -2.29058200 | 0.32512300 | -0.00005200 |
| C | -1.33448100 | 1.38784700 | -0.00000900 |
| C | 0.01179600  | 1.10378700 | 0.00015500  |

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 0.43914100  | -0.25619800 | 0.00011600  |
| C | -0.53305200 | -1.32134000 | 0.00001800  |
| C | -1.87707700 | -1.03019000 | -0.00013500 |
| H | -3.35084000 | 0.56366400  | 0.00003500  |
| H | -1.68305500 | 2.41611600  | -0.00009200 |
| H | 0.76099500  | 1.88758900  | 0.00023300  |
| H | -0.18917700 | -2.35270300 | 0.00007500  |
| H | -2.61830900 | -1.82283300 | -0.00025700 |
| C | 1.87681700  | -0.61839900 | 0.00014200  |
| H | 2.06172800  | -1.27568800 | -0.87732100 |
| H | 2.06170400  | -1.27509600 | 0.87809000  |
| O | 2.69596800  | 0.50752500  | -0.00017100 |
| H | 3.63383300  | 0.25498000  | -0.00080400 |



TEMPO<sup>+</sup>

Energies= -483.691648 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| N | 0.00000400  | -0.80712900 | 0.00000000  |
| O | 0.00001800  | -2.00375700 | -0.00022800 |
| C | 2.33527900  | -0.92047300 | 0.82349800  |
| H | 1.98171100  | -1.07949900 | 1.84743000  |
| H | 2.52845600  | -1.88966000 | 0.35956300  |
| H | 3.27960500  | -0.37057600 | 0.87315600  |
| C | -1.77730200 | -0.01731900 | 1.47965100  |
| H | -1.86122700 | -1.02367600 | 1.89946700  |
| H | -1.10893700 | 0.58099000  | 2.10145400  |
| H | -2.76714900 | 0.44980700  | 1.50010500  |
| C | 1.34401300  | -0.07491400 | 0.01242400  |
| C | 1.77760100  | -0.01752100 | -1.47953100 |
| H | 2.76748600  | 0.44953200  | -1.49987000 |
| H | 1.86154800  | -1.02394300 | -1.89918900 |
| H | 1.10941900  | 0.58073900  | -2.10156600 |
| C | -1.34400300 | -0.07495600 | -0.01236800 |
| C | -2.33539300 | -0.92067400 | -0.82313500 |
| H | -2.52841700 | -1.88980900 | -0.35902600 |
| H | -3.27976100 | -0.37084000 | -0.87267800 |

|   |             |             |             |
|---|-------------|-------------|-------------|
| H | -1.98202800 | -1.07982800 | -1.84711900 |
| C | -1.13198700 | 1.31805500  | -0.62860500 |
| H | -2.08850500 | 1.84329900  | -0.53757700 |
| H | -0.96826300 | 1.19201800  | -1.70432000 |
| C | -0.00007100 | 2.16185900  | -0.00020400 |
| H | -0.40453100 | 2.82124900  | 0.77355300  |
| H | 0.40437600  | 2.82107400  | -0.77411700 |
| C | 1.13182400  | 1.31817200  | 0.62842900  |
| H | 2.08833300  | 1.84344000  | 0.53745800  |
| H | 0.96795500  | 1.19227300  | 1.70414400  |



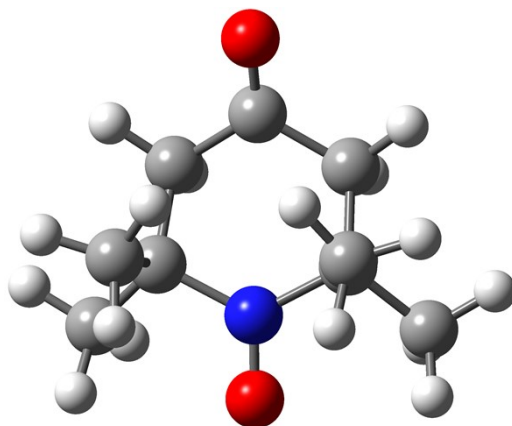
TEMPO

Energies= -483.869372 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| N | -0.00006100 | -0.79299700 | 0.00002200  |
| O | 0.00021600  | -2.07806200 | -0.00033100 |
| C | 2.30313800  | -0.89326200 | 0.86384600  |
| H | 1.91231900  | -1.03301000 | 1.87746900  |
| H | 2.48482300  | -1.88013500 | 0.43307900  |
| H | 3.25530900  | -0.35373600 | 0.92928100  |
| C | -1.85443600 | -0.00254000 | 1.44371200  |
| H | -1.90118400 | -1.00631500 | 1.87872100  |
| H | -1.22443300 | 0.62072800  | 2.08590700  |
| H | -2.86566600 | 0.42256700  | 1.44645900  |
| C | 1.31927600  | -0.08344600 | 0.00314900  |
| C | 1.85485100  | -0.00335100 | -1.44343500 |
| H | 2.86610800  | 0.42171000  | -1.44637500 |
| H | 1.90159400  | -1.00747900 | -1.87761900 |
| H | 1.22498900  | 0.61942100  | -2.08623800 |
| C | -1.31911200 | -0.08374300 | -0.00296800 |
| C | -2.30347300 | -0.89384600 | -0.86290300 |
| H | -2.48549600 | -1.88030300 | -0.43132500 |
| H | -3.25540200 | -0.35396000 | -0.92861300 |
| H | -1.91282800 | -1.03461900 | -1.87645400 |
| C | -1.12564300 | 1.31185800  | -0.62962800 |
| H | -2.08132800 | 1.84712300  | -0.56976900 |



|   |             |            |             |
|---|-------------|------------|-------------|
| H | -0.92651900 | 1.17050700 | -1.69894600 |
| C | -0.00018700 | 2.16206600 | -0.00051600 |
| H | -0.41070900 | 2.82338700 | 0.77185300  |
| H | 0.41010000  | 2.82327900 | -0.77310400 |
| C | 1.12547700  | 1.31239900 | 0.62893700  |
| H | 2.08108900  | 1.84776800 | 0.56861700  |
| H | 0.92657900  | 1.17173400 | 1.69838800  |



4-oxo-TEMPO<sup>+</sup>

Energies= -557.725227 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| N | 0.00017700  | -0.99648400 | -0.07211200 |
| O | 0.00032900  | -2.18894500 | -0.14137800 |
| C | -2.40802400 | -1.21084200 | -0.65368000 |
| H | -2.12694400 | -1.47952100 | -1.67702800 |
| H | -2.56842900 | -2.12467600 | -0.07752900 |
| H | -3.35447000 | -0.66324800 | -0.69492100 |
| C | 2.40837000  | -1.20994600 | -0.65392500 |
| H | 2.56899500  | -2.12397800 | -0.07814600 |
| H | 2.12738100  | -1.47830000 | -1.67738600 |
| H | 3.35468500  | -0.66211600 | -0.69494400 |
| C | -1.37478100 | -0.28666500 | -0.00336400 |
| C | -1.68527000 | -0.09935500 | 1.50653300  |
| H | -2.73091200 | 0.21860200  | 1.56836900  |
| H | -1.58445100 | -1.04049200 | 2.05453800  |
| H | -1.07508600 | 0.67270200  | 1.97708500  |
| C | 1.37487400  | -0.28628100 | -0.00330700 |
| C | 1.68548800  | -0.09916200 | 1.50656000  |
| H | 1.58523700  | -1.04051100 | 2.05430900  |
| H | 2.73098200  | 0.21930000  | 1.56831800  |
| H | 1.07499800  | 0.67245000  | 1.97743600  |
| C | 1.27177900  | 1.07281300  | -0.73651700 |
| H | 1.30237600  | 0.90449600  | -1.82152700 |
| H | 2.14395000  | 1.67295900  | -0.46362900 |
| C | -0.00035600 | 1.83245300  | -0.39317900 |

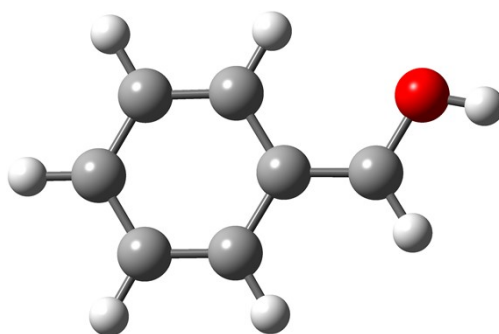
|   |             |            |             |
|---|-------------|------------|-------------|
| C | -1.27213500 | 1.07230700 | -0.73668300 |
| H | -2.14456900 | 1.67216600 | -0.46401800 |
| H | -1.30242600 | 0.90393500 | -1.82170600 |
| O | -0.00060700 | 2.93140500 | 0.11399600  |



4-oxo-TEMPO

Energies= -557.914124 a.u

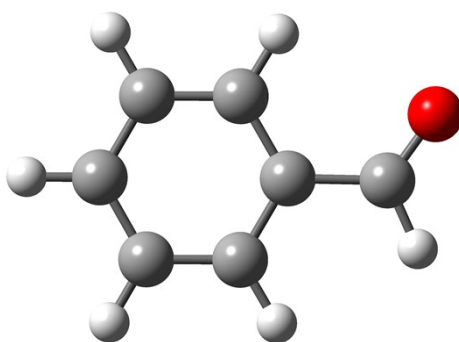
|   |             |             |             |
|---|-------------|-------------|-------------|
| N | 0.00054000  | -0.98258200 | -0.15726800 |
| O | 0.00120700  | -2.24413600 | 0.08901000  |
| C | -2.36934900 | -1.15112300 | -0.77590500 |
| H | -2.06039200 | -1.32316100 | -1.81269300 |
| H | -2.49669700 | -2.12136500 | -0.29122300 |
| H | -3.33239000 | -0.62805100 | -0.78122900 |
| C | 2.37042300  | -1.14830800 | -0.77654200 |
| H | 2.49865900  | -2.11876700 | -0.29254700 |
| H | 2.06148300  | -1.31985700 | -1.81339800 |
| H | 3.33299400  | -0.62439000 | -0.78162300 |
| C | -1.33283200 | -0.29526500 | -0.03046100 |
| C | -1.72821300 | -0.18461000 | 1.45807000  |
| H | -2.77011800 | 0.14620500  | 1.53814100  |
| H | -1.63783300 | -1.16316700 | 1.93940900  |
| H | -1.11037500 | 0.53551500  | 2.00268600  |
| C | 1.33321300  | -0.29389200 | -0.03041500 |
| C | 1.72877700  | -0.18375200 | 1.45809300  |
| H | 1.63977000  | -1.16279300 | 1.93878600  |
| H | 2.77027600  | 0.14836300  | 1.53816700  |
| H | 1.11005900  | 0.53514600  | 2.00330900  |
| C | 1.26308700  | 1.11084400  | -0.68165600 |
| H | 1.27895300  | 0.99548200  | -1.77478900 |
| H | 2.13754800  | 1.69842200  | -0.38785200 |
| C | -0.00111700 | 1.85845200  | -0.31505600 |
| C | -1.26411400 | 1.10918400  | -0.68245600 |
| H | -2.13954400 | 1.69579000  | -0.38959200 |
| H | -1.27898200 | 0.99320400  | -1.77553700 |
| O | -0.00201300 | 2.95192600  | 0.22334300  |



B / C<sub>7</sub>H<sub>7</sub>O<sup>•</sup>

Energies= -346.254893 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | -2.28190800 | -0.30465100 | -0.00023300 |
| C | -1.34400200 | -1.34853200 | 0.00006000  |
| C | 0.01980100  | -1.08315700 | 0.00031200  |
| C | 0.49429200  | 0.26127800  | 0.00023300  |
| C | -0.47241800 | 1.30991200  | 0.00010700  |
| C | -1.83030600 | 1.02571900  | -0.00014800 |
| H | -3.34640000 | -0.52225700 | -0.00060600 |
| H | -1.68608400 | -2.38096500 | 0.00013900  |
| H | 0.73872100  | -1.89606400 | 0.00060300  |
| H | -0.13361800 | 2.34405200  | 0.00028800  |
| H | -2.54881300 | 1.84200300  | -0.00025400 |
| C | 1.86732100  | 0.57140500  | 0.00019400  |
| H | 2.23680500  | 1.59260800  | 0.00036600  |
| O | 2.79200800  | -0.43780700 | -0.00042000 |
| H | 3.68664800  | -0.06876000 | -0.00033000 |

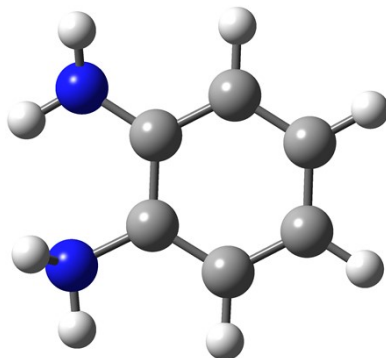


6a/C<sub>6</sub>H<sub>5</sub>CHO

Energies= -345.687506 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 2.22213400  | -0.24368300 | 0.00003700  |
| C | 1.33545600  | -1.32958900 | -0.00005500 |
| C | -0.03918100 | -1.10745200 | 0.00003100  |
| C | -0.53574200 | 0.20705400  | -0.00000100 |
| C | 0.35571400  | 1.29013400  | -0.00012600 |
| C | 1.73367000  | 1.06629100  | 0.00006600  |
| H | 3.29469200  | -0.42095300 | 0.00004500  |

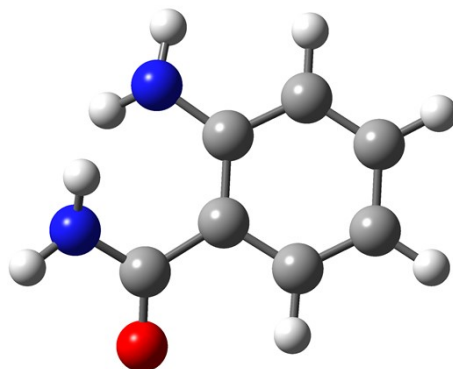
|   |             |             |             |
|---|-------------|-------------|-------------|
| H | 1.72203300  | -2.34534100 | -0.00009300 |
| H | -0.74451700 | -1.93342500 | 0.00006200  |
| H | -0.03418200 | 2.30640400  | -0.00018800 |
| H | 2.42315100  | 1.90623400  | 0.00011600  |
| C | -1.99351400 | 0.46622000  | 0.00003000  |
| H | -2.27653900 | 1.54138100  | 0.00025600  |
| O | -2.85698300 | -0.39351800 | -0.00001100 |



1a/C<sub>6</sub>H<sub>8</sub>N<sub>2</sub>

Energies= -343.082718 a.u

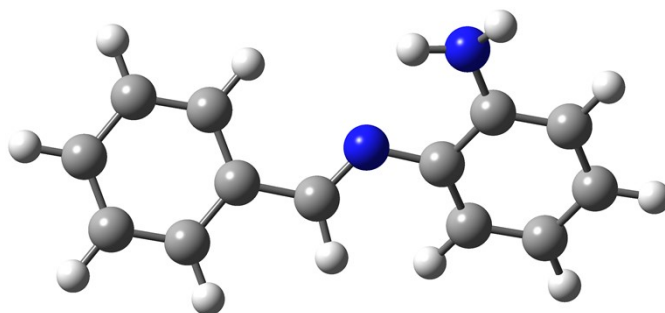
|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 0.69301300  | -1.39242400 | -0.01772400 |
| C | -0.51774100 | -0.69427000 | -0.00199400 |
| C | -0.50305600 | 0.72084400  | -0.00942400 |
| C | 0.73175600  | 1.38589500  | 0.02463600  |
| C | 1.93311200  | 0.67475500  | 0.02675900  |
| C | 1.91961800  | -0.72118300 | -0.01225100 |
| H | 0.67059600  | -2.48118600 | -0.01739100 |
| H | 0.74246300  | 2.47408200  | 0.01528200  |
| H | 2.87592700  | 1.21531700  | 0.04485300  |
| H | 2.84811500  | -1.28517900 | -0.02356100 |
| N | -1.69881500 | 1.43299000  | -0.10648800 |
| H | -2.55809400 | 0.90333700  | -0.08958900 |
| H | -1.74904200 | 2.33971500  | 0.33576400  |
| N | -1.78084000 | -1.35555700 | -0.03389400 |
| H | -1.68117900 | -2.35957000 | -0.15108800 |
| H | -2.33141600 | -1.19025600 | 0.80839500  |



4a/ C<sub>7</sub>H<sub>8</sub>N<sub>2</sub>O

Energies= -456.473590 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 2.61483200  | -0.19679900 | 0.07833900  |
| C | 1.88412800  | 0.98242000  | -0.02059700 |
| C | 0.47770100  | 0.96791900  | -0.07676500 |
| C | -0.19139800 | -0.28033900 | -0.01783000 |
| C | 0.57049000  | -1.45984000 | 0.04923000  |
| C | 1.95933000  | -1.43343600 | 0.10626600  |
| H | 3.70021400  | -0.14965600 | 0.12078100  |
| H | 2.40196200  | 1.93877700  | -0.06530000 |
| H | 0.02977200  | -2.40087400 | 0.06251200  |
| H | 2.52425900  | -2.35869000 | 0.17104600  |
| N | -0.21051600 | 2.19110500  | -0.14679900 |
| H | -1.05844200 | 2.17897600  | -0.70483800 |
| H | 0.38218600  | 2.97339000  | -0.40178800 |
| C | -1.68115300 | -0.46525000 | -0.01943300 |
| O | -2.20509600 | -1.51973500 | -0.36835500 |
| N | -2.45558900 | 0.60815800  | 0.38047700  |
| H | -3.42847000 | 0.37786800  | 0.54835800  |
| H | -2.05155000 | 1.31520100  | 0.98507100  |

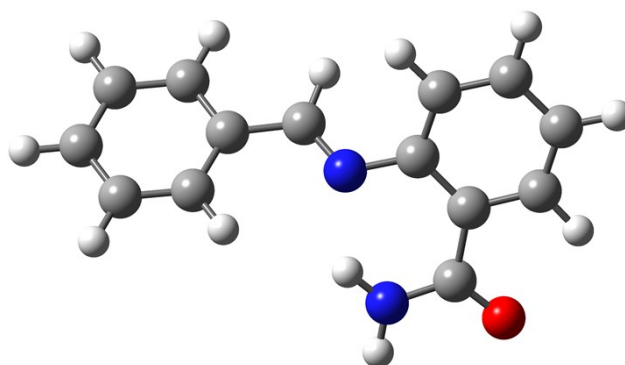


D1/ C<sub>13</sub>H<sub>12</sub>N<sub>2</sub>

Energies= -612.303197 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 2.17230100  | -1.38463200 | -0.35333600 |
| C | 1.67364700  | -0.09505800 | -0.09822400 |
| C | 2.58884800  | 0.96287300  | 0.15754400  |
| C | 3.96454500  | 0.67627000  | 0.19920800  |
| C | 4.43380400  | -0.61464200 | -0.03120000 |
| C | 3.53927300  | -1.65269800 | -0.32092900 |
| H | 1.47647000  | -2.17821800 | -0.61264100 |
| H | 4.66575200  | 1.48414500  | 0.39865300  |
| H | 5.50335000  | -0.80742000 | -0.00335700 |
| H | 3.90464700  | -2.65316900 | -0.53501900 |
| N | 2.12087500  | 2.25781700  | 0.32436700  |
| H | 1.12639400  | 2.34016000  | 0.49116900  |
| H | 2.69236200  | 2.88642500  | 0.87235600  |
| N | 0.31308500  | 0.25126800  | -0.12790600 |
| C | -0.60486300 | -0.60429700 | 0.14280000  |

|   |             |             |             |
|---|-------------|-------------|-------------|
| H | -0.36257500 | -1.62586000 | 0.47150000  |
| C | -2.03693300 | -0.29448500 | 0.05629000  |
| C | -2.50353700 | 0.93992400  | -0.43472600 |
| C | -2.97248700 | -1.25816100 | 0.47121900  |
| C | -3.86968200 | 1.20016000  | -0.49751800 |
| H | -1.77930000 | 1.67661100  | -0.76839500 |
| C | -4.34221800 | -0.99591600 | 0.40781000  |
| H | -2.62090700 | -2.21690000 | 0.84760900  |
| C | -4.79424800 | 0.23463200  | -0.07587100 |
| H | -4.22035300 | 2.15536200  | -0.88037400 |
| H | -5.05411900 | -1.74986200 | 0.73374100  |
| H | -5.86014300 | 0.44132500  | -0.12887400 |

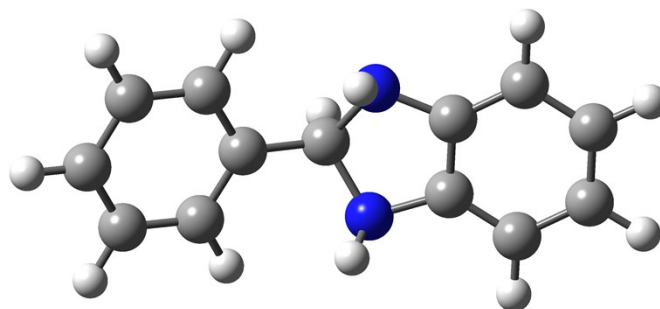


D2/ C<sub>14</sub>H<sub>12</sub>N<sub>2</sub>O

Energies= -725.689710 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 2.93464100  | -2.41850700 | -0.34218500 |
| C | 1.63325100  | -1.92547200 | -0.40137600 |
| C | 1.36083700  | -0.55793700 | -0.20185100 |
| C | 2.44009700  | 0.33363600  | 0.03293700  |
| C | 3.73820900  | -0.19139900 | 0.10601000  |
| C | 3.99514000  | -1.54840300 | -0.07818300 |
| H | 3.11757100  | -3.47673100 | -0.51160900 |
| H | 0.81163200  | -2.59354800 | -0.64450100 |
| H | 4.54306400  | 0.50728800  | 0.30924600  |
| H | 5.01471300  | -1.92069400 | -0.02929300 |
| N | 0.03548700  | -0.08439100 | -0.29967800 |
| C | 2.34196000  | 1.83330700  | 0.23474400  |
| O | 3.33232100  | 2.48689400  | 0.56714700  |
| N | 1.13721400  | 2.42297300  | 0.01647600  |
| H | 1.08256700  | 3.42736400  | 0.11730700  |
| H | 0.33388000  | 1.87516800  | -0.27383900 |
| C | -0.92346000 | -0.74265200 | 0.24301300  |
| H | -0.72066000 | -1.62867200 | 0.86156500  |
| C | -2.33615300 | -0.37054300 | 0.10774800  |
| C | -2.76056000 | 0.64748900  | -0.76685600 |
| C | -3.29517600 | -1.05815200 | 0.87132000  |

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | -4.11074200 | 0.97314600  | -0.86185000 |
| H | -2.02336500 | 1.16365400  | -1.37432300 |
| C | -4.64827600 | -0.72971200 | 0.77621700  |
| H | -2.97501000 | -1.85021900 | 1.54513200  |
| C | -5.05810000 | 0.28769600  | -0.08952500 |
| H | -4.43065200 | 1.75829700  | -1.54188600 |
| H | -5.37982900 | -1.26622200 | 1.37441100  |
| H | -6.11139700 | 0.54411400  | -0.16794700 |

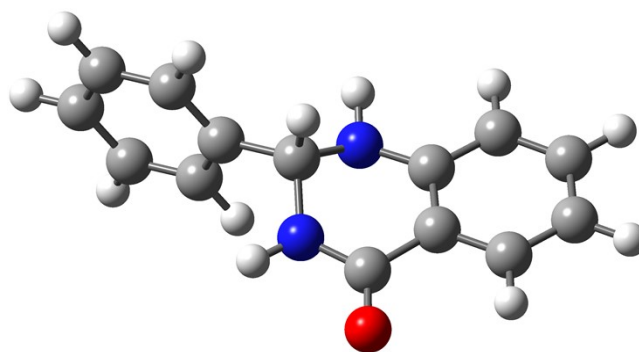


E1/ C<sub>13</sub>H<sub>12</sub>N<sub>2</sub>

Energies= -612.288698 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | -2.99916500 | -1.41915500 | -0.09900300 |
| C | -1.81974400 | -0.70230500 | 0.06397200  |
| C | -1.81980600 | 0.70235200  | 0.06414600  |
| C | -2.99933500 | 1.41909000  | -0.09864600 |
| C | -4.19198000 | 0.69947300  | -0.27376200 |
| C | -4.19190100 | -0.69964600 | -0.27396800 |
| H | -2.99480700 | -2.50589500 | -0.09289900 |
| H | -2.99516800 | 2.50583100  | -0.09226400 |
| H | -5.12774000 | 1.23817000  | -0.39795900 |
| H | -5.12760400 | -1.23841000 | -0.39830100 |
| N | -0.48883300 | 1.19847700  | 0.22746200  |
| H | -0.12580900 | 1.58437800  | -0.64626200 |
| C | 0.27163600  | 0.00008100  | 0.67022200  |
| H | 0.20033400  | -0.00022400 | 1.76796500  |
| C | 1.72955200  | 0.00009600  | 0.27053500  |
| C | 2.42688400  | -1.20718200 | 0.11132400  |
| C | 2.42687200  | 1.20725700  | 0.11083700  |
| C | 3.78284200  | -1.20687500 | -0.22608500 |
| H | 1.91283400  | -2.15153100 | 0.26791500  |
| C | 3.78286400  | 1.20680200  | -0.22657000 |
| H | 1.91291300  | 2.15173200  | 0.26699800  |
| C | 4.46491500  | -0.00006400 | -0.40129700 |
| H | 4.30602800  | -2.15201700 | -0.34788400 |
| H | 4.30605300  | 2.15189200  | -0.34874800 |
| H | 5.51954300  | -0.00010700 | -0.66432500 |
| N | -0.48870200 | -1.19840200 | 0.22715400  |

H            -0.12564400   -1.58389200   -0.64677600

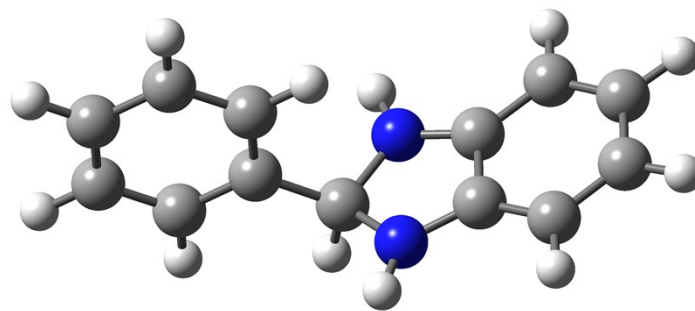


E2/ C<sub>14</sub>H<sub>12</sub>N<sub>2</sub>O

Energies= -725.698012 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 3.84701200  | -1.76068200 | 0.04379600  |
| C | 2.46588600  | -1.93783900 | 0.07253700  |
| C | 1.61352000  | -0.82098700 | 0.03955300  |
| C | 2.17540300  | 0.47419700  | -0.01128600 |
| C | 3.56677700  | 0.63247600  | -0.03788200 |
| C | 4.40778100  | -0.47645400 | -0.01607500 |
| H | 4.49386700  | -2.63443300 | 0.06387500  |
| H | 2.04124300  | -2.93846500 | 0.12035800  |
| H | 3.96202100  | 1.64346100  | -0.06591300 |
| H | 5.48610700  | -0.34937400 | -0.03874100 |
| C | 1.28829100  | 1.66670700  | 0.03540000  |
| O | 1.69528900  | 2.82103400  | 0.14797400  |
| N | -0.06113400 | 1.37377300  | 0.00160500  |
| H | -0.66965800 | 2.18118400  | -0.07744100 |
| C | -0.56245600 | 0.10320900  | -0.52186200 |
| H | -0.41963900 | 0.07497300  | -1.61986200 |
| C | -2.04094400 | -0.06344400 | -0.22747200 |
| C | -2.50186900 | -0.11154900 | 1.09649700  |
| C | -2.95907100 | -0.16384300 | -1.27849500 |
| C | -3.86421600 | -0.25797400 | 1.35997000  |
| H | -1.78713700 | -0.03370800 | 1.91072300  |
| C | -4.32523000 | -0.30993300 | -1.01518800 |
| H | -2.60627500 | -0.12579400 | -2.30723500 |
| C | -4.77869200 | -0.35709700 | 0.30457000  |
| H | -4.21444000 | -0.29364000 | 2.38837400  |
| H | -5.02993600 | -0.38573000 | -1.83915600 |
| H | -5.83966400 | -0.46987800 | 0.51239200  |
| N | 0.22591100  | -0.95290200 | 0.11126100  |
| H | -0.12539800 | -1.88369000 | -0.08561900 |

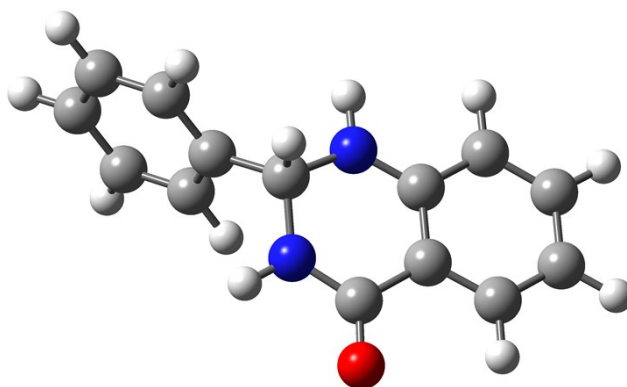




F1/ C<sub>13</sub>H<sub>12</sub>N<sub>2</sub><sup>+</sup>

Energies= -612.131995 a.u

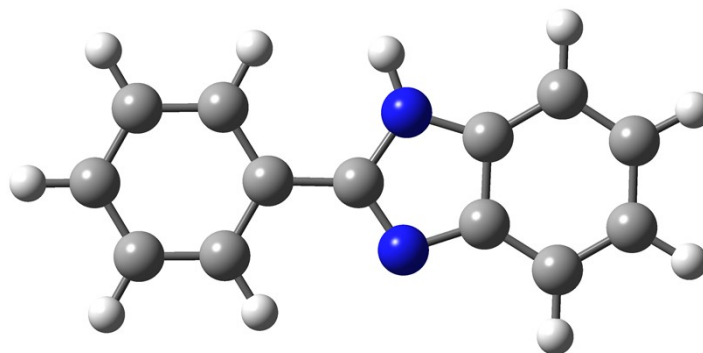
|   |             |             |             |
|---|-------------|-------------|-------------|
| C | -2.86411100 | -0.01379900 | -1.45531000 |
| C | -1.73623900 | -0.34529200 | -0.68573400 |
| C | -1.73707400 | -0.17784500 | 0.74642000  |
| C | -2.86587800 | 0.32189400  | 1.41736600  |
| C | -3.96803600 | 0.64056000  | 0.64190500  |
| C | -3.96718000 | 0.47497200  | -0.77567600 |
| H | -2.86734700 | -0.13864400 | -2.53321000 |
| H | -2.87070700 | 0.44867500  | 2.49503900  |
| H | -4.86209000 | 1.02655700  | 1.12147900  |
| H | -4.86052600 | 0.74007800  | -1.33242600 |
| N | -0.52297900 | -0.56331900 | 1.19782300  |
| H | -0.21066500 | -0.51997200 | 2.16056600  |
| C | 0.35582300  | -1.04142300 | 0.12292900  |
| H | 0.51656700  | -2.12367300 | 0.24929200  |
| C | 1.70355200  | -0.34905700 | 0.04207800  |
| C | 2.86949400  | -1.11911900 | 0.12420100  |
| C | 1.78940300  | 1.04299700  | -0.11350800 |
| C | 4.12117900  | -0.50064000 | 0.05116800  |
| H | 2.80617000  | -2.19857300 | 0.24480100  |
| C | 3.03946400  | 1.65566100  | -0.18562400 |
| H | 0.88717400  | 1.64703500  | -0.17780900 |
| C | 4.20541700  | 0.88423100  | -0.10341800 |
| H | 5.02402000  | -1.10067800 | 0.11530200  |
| H | 3.10665400  | 2.73303400  | -0.30557000 |
| H | 5.17745300  | 1.36560000  | -0.16003900 |
| N | -0.52175300 | -0.82432100 | -1.03457500 |
| H | -0.20846000 | -1.00481200 | -1.98094700 |



F2/ C<sub>14</sub>H<sub>12</sub>N<sub>2</sub>O<sup>+</sup>

Energies= -725.500178 a.u

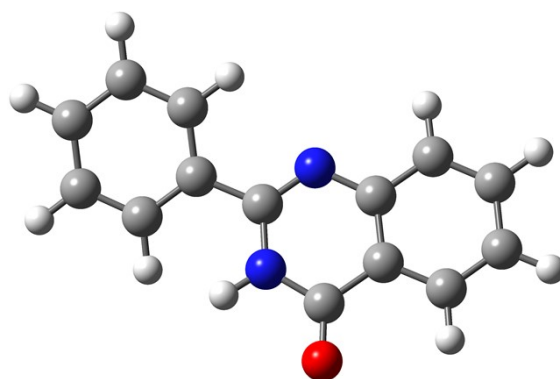
|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 3.66562700  | -1.90043100 | 0.21292200  |
| C | 2.31895300  | -2.00370600 | -0.06190200 |
| C | 1.54432100  | -0.80889400 | -0.20368400 |
| C | 2.15395500  | 0.47714300  | -0.02620100 |
| C | 3.50649500  | 0.54952100  | 0.24592400  |
| C | 4.26925100  | -0.62922600 | 0.36136200  |
| H | 4.26779800  | -2.79809100 | 0.31223700  |
| H | 1.84258000  | -2.97261400 | -0.18631200 |
| H | 3.95746500  | 1.52787900  | 0.38090400  |
| H | 5.33146000  | -0.56180300 | 0.57610700  |
| C | 1.31851700  | 1.71644800  | -0.07378300 |
| O | 1.77922600  | 2.82481600  | 0.13197900  |
| N | -0.02546200 | 1.48750700  | -0.27958100 |
| H | -0.60550800 | 2.32164300  | -0.30420100 |
| C | -0.62888100 | 0.28245200  | -0.81360700 |
| H | -0.67239400 | 0.32920200  | -1.91640800 |
| C | -2.03175700 | 0.03453000  | -0.28807100 |
| C | -2.28383200 | 0.03664900  | 1.09299500  |
| C | -3.07122800 | -0.21332700 | -1.19268100 |
| C | -3.57428700 | -0.21085500 | 1.56070600  |
| H | -1.48121100 | 0.24639900  | 1.79578500  |
| C | -4.36292500 | -0.45936600 | -0.71861000 |
| H | -2.88145600 | -0.20220300 | -2.26406600 |
| C | -4.61298000 | -0.45955900 | 0.65596100  |
| H | -3.77133900 | -0.20472800 | 2.62872000  |
| H | -5.17033300 | -0.64054600 | -1.42182700 |
| H | -5.61774000 | -0.64658700 | 1.02367000  |
| N | 0.23923700  | -0.87096300 | -0.51210700 |
| H | -0.19692700 | -1.78115000 | -0.64660300 |



3aa/ C<sub>13</sub>H<sub>10</sub>N<sub>2</sub>

Energies= -611.121214 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 3.06354900  | 1.41212200  | -0.03895800 |
| C | 1.85550700  | 0.71076000  | -0.01737700 |
| C | 1.79550100  | -0.70484600 | 0.01992000  |
| C | 2.98210500  | -1.45248800 | 0.03848200  |
| C | 4.19082200  | -0.76134500 | 0.01756600  |
| C | 4.23131600  | 0.64960500  | -0.02093200 |
| H | 3.10020500  | 2.49808500  | -0.06786000 |
| H | 2.94557100  | -2.53758000 | 0.06786400  |
| H | 5.12544700  | -1.31573700 | 0.03097300  |
| H | 5.19397700  | 1.15365500  | -0.03652100 |
| N | 0.47907400  | -1.12823900 | 0.03230200  |
| C | -0.25128600 | -0.03014400 | 0.00453900  |
| C | -1.71908100 | -0.00133200 | 0.00361600  |
| C | -2.44912900 | 1.19884100  | 0.04744600  |
| C | -2.42161100 | -1.21927100 | -0.04211600 |
| C | -3.84486200 | 1.18328200  | 0.04213400  |
| H | -1.94107700 | 2.15924400  | 0.09369400  |
| C | -3.81456800 | -1.23129500 | -0.04646300 |
| H | -1.85480900 | -2.14387700 | -0.07424000 |
| C | -4.53352500 | -0.03130200 | -0.00535300 |
| H | -4.39256000 | 2.12141800  | 0.07773200  |
| H | -4.34258300 | -2.18071200 | -0.08295700 |
| H | -5.62027600 | -0.04317100 | -0.00922600 |
| N | 0.53177100  | 1.11231700  | -0.02437800 |
| H | 0.20175900  | 2.06460600  | -0.06995800 |



5aa/ C<sub>14</sub>H<sub>10</sub>N<sub>2</sub>O

Energies= -724.504861 a.u

|   |             |             |             |
|---|-------------|-------------|-------------|
| C | 3.79661600  | -1.78923600 | 0.16145300  |
| C | 2.41618600  | -1.92819400 | 0.16561200  |
| C | 1.58679000  | -0.79055300 | 0.07001600  |
| C | 2.19450400  | 0.48687200  | -0.02860600 |
| C | 3.59342000  | 0.61464000  | -0.03175900 |
| C | 4.39212900  | -0.51649900 | 0.06335600  |
| H | 4.42556700  | -2.67285700 | 0.23591700  |
| H | 1.94402500  | -2.90289500 | 0.24393300  |
| H | 4.02265900  | 1.60884500  | -0.11073400 |
| H | 5.47444500  | -0.42177400 | 0.06266500  |
| C | 1.33649800  | 1.66884100  | -0.14428800 |
| O | 1.70362600  | 2.83384200  | -0.25936300 |
| N | -0.02824300 | 1.35527500  | -0.12712000 |
| H | -0.64565900 | 2.14681900  | -0.26928900 |
| C | -0.54588500 | 0.07997600  | -0.00655300 |
| C | -2.02319100 | -0.06331400 | 0.01027900  |
| C | -2.87300700 | 0.98884100  | 0.39139800  |
| C | -2.58946100 | -1.29592700 | -0.35710600 |
| C | -4.25842100 | 0.81528200  | 0.39363100  |
| H | -2.46705600 | 1.94007200  | 0.72674100  |
| C | -3.97261600 | -1.46437300 | -0.35865500 |
| H | -1.92865000 | -2.10830300 | -0.63990800 |
| C | -4.81263900 | -0.40930300 | 0.01379100  |
| H | -4.90185600 | 1.63541700  | 0.70053300  |
| H | -4.39712800 | -2.42055500 | -0.65311600 |
| H | -5.89124600 | -0.54259600 | 0.01262300  |
| N | 0.21258000  | -0.97173500 | 0.08571000  |

## 8. Computational methods

Density functional theory (DFT) calculations were carried out to calculate the structures and the reaction mechanisms by using Gaussian 16 program<sup>1</sup>. All structures were optimized by using the

combination of Becke's hybrid 3-parameter exchange functional<sup>2</sup> and Lee-Yang-Parr's correlation functional<sup>3</sup> known as B3LYP method in conjunction with 6-31+G\* basis set to ensure these structure without imaginary frequencies. Then, the energy of the reaction mechanism is calculated at the B3LYP/6-311++G\*\* level based on the optimized structures, and the solvation model based on electron density (SMD)<sup>4</sup> with CH<sub>3</sub>CN/H<sub>2</sub>O (1:2,v:v) mixed solvent attached was used throughout. Dimensional plots of molecular configurations were generated with the GaussView program<sup>5</sup>.

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