

## Supporting Information

### Synthesis of Novel Azole Fused-Quinazolines *via* One Pot Sequential Ullmann type Coupling and Intramolecular Dehydrogenative C–N Bonding

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## 1. General information

Melting points were determined in open capillary tubes on a EZ-Melt Automated melting point apparatus and are uncorrected. Reactions were monitored by using thin layer chromatography (TLC) on 0.2 mm silica gel F<sub>254</sub> plates (Merck). The chemical structures of final products were determined by nuclear magnetic resonance spectra (<sup>1</sup>H NMR, <sup>13</sup>C NMR) determined on a Bruker AV 400 MHz and Varian 300 MHz spectrometer. <sup>13</sup>C NMR spectra are fully decoupled. Chemical shifts were reported in parts per million (ppm) using deuterated solvent peak or Tetramethylsilane (internal) as the standard. High resolution mass spectra (HRMS-ESI) were carried out using a quadrupole time of-flight (Q-TOF) mass spectrometer (Applied Biosystem). All chemicals were obtained from the commercial suppliers and used without further purification.

**2. 1. Experimental procedure for synthesis of 1a:** An oven-dried 10 mL RB flask was charged with benzil (1 mmol), 2-bromobenzaldehyde (1 mmol), NH<sub>4</sub>OAc (4 mmol), L-proline (20 mol %) and MeOH (3 mL). The resulting solution was refluxed for 12 h. On completion of the reaction as indicated by TLC, MeOH was evaporated. The residue was diluted with water (10 mL) and extracted in EtOAc (2 × 10 mL). The combined organic layers were dried over anhydrous Na<sub>2</sub>SO<sub>4</sub> and evaporated to dryness. The crude solid obtained was washed with hexane twice to remove impurity to afford **1a** (79% yield); mp 203-205 °C.

**2.2. Experimental procedure for synthesis of 1m:** An oven dried 10 ml RB was charged with 1,2-phenylenediamine (1 mmol), 2-bromobenzaldehyde (1 mmol), H<sub>2</sub>O<sub>2</sub> (30%, 4 mmol, 0.4 mL), and ceric ammonium nitrate (CAN) (0.1 mmol). The reaction mixture was stirred at room temperature for 5 min. On completion of the reaction as indicated by TLC, the reaction mass was allowed to cool to ambient temperature, dissolved in EtOH and poured into ice water. Solid was filtered and washed with cooled water and dried to get **1m** in 69% yield; mp 210-213 °C.

**3. Experimental procedure for synthesis of 2,3-diphenyldiimidazo[1,2-*a*:1',2'-*c*]quinazoline (3a):** A clean oven dried 10 mL round bottom flask was charged with **1a** (1 mmol), imidazole **2a** (1.2 mmol), K<sub>2</sub>CO<sub>3</sub> (2 mmol), CuI (0.20 mmol) and DMF (3 mL). The resulting solution was stirred at 150 °C for 1 h. On completion of first step Pd(OAc)<sub>2</sub> (0.05 mmol) and Cu(OAc)<sub>2</sub> (2 mmol) was added in same flask without isolating intermediate and again the reaction mass was stirred at 150 °C for 2 h. Reaction progress was monitored by TLC. After completion, the reaction mass was allowed to cool to ambient temperature, diluted with water and extracted with EtOAc (2 × 10 mL). The combined organic layer was dried with anhydrous Na<sub>2</sub>SO<sub>4</sub> and evaporated to dryness. The crude material was purified by column chromatography to obtain desired tandem product **3a** in 71% yield (255 mg).

#### 4. Characterization data for 3a and 5a-o

**2-(2-(1*H*-imidazol-1-yl)phenyl)-4,5-diphenyl-1*H*-imidazole (3a):** Yield 81%, Off-white solid; mp 144-147 °C; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.68 (d, *J* = 7.1 Hz, 1H), 7.71 – 7.54 (m, 8H), 7.53 – 7.45 (m, 4H), 7.32 – 7.19 (m, 4H), 7.14 (d, *J* = 1.4 Hz, 1H); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 141.40, 139.67, 137.16, 133.70, 131.86, 130.38, 130.05, 129.98, 129.02, 128.84, 128.29, 128.26, 127.89, 127.36, 126.04, 125.24, 124.83, 115.18, 114.64, 110.02.

**2,3-Diphenyldiimidazo[1,2-*a*:1',2'-*c*]quinazoline (5a):** Yield 71%; Off-white solid; mp 231-234 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.73 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.74 (d, *J* = 7.9 Hz, 1H), 7.69 (d, *J* = 1.6 Hz, 1H), 7.68 – 7.57 (m, 6H), 7.56 – 7.51 (m, 3H), 7.33 – 7.29 (m, 1H), 7.28 – 7.23 (m, 2H), 7.18 (d, *J* = 1.6 Hz, 1H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 141.38, 139.68, 137.19, 133.74, 131.88, 130.40, 130.05, 129.05, 128.92, 128.28, 127.89, 127.36, 126.03, 125.26, 124.83, 115.19, 114.65, 110.00; HRMS (ESI) calcd for C<sub>24</sub>H<sub>17</sub>N<sub>4</sub> 361.1448 found 361.1443 [M+H]<sup>+</sup>.

**5,6-Diphenylimidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5b):** Yield 62%; Off-white solid; mp 260-261 °C; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.69 (d, *J* = 7.8 Hz, 1H), 8.25 (d, *J* = 8.2 Hz, 1H), 8.01 (s, 1H), 7.72 (t, *J* = 7.4 Hz, 1H), 7.68 – 7.60 (m, 3H), 7.60 – 7.47 (m, 5H), 7.31 – 7.22 (m, 3H); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 151.24, 143.82, 142.08, 140.40, 133.20, 131.79, 130.80, 130.73, 129.48, 129.02, 128.47, 128.35, 127.91, 127.74, 127.09, 124.72, 115.55, 114.58; HRMS (ESI) calcd for C<sub>23</sub>H<sub>16</sub>N<sub>5</sub> 362.1400 found 362.1403 [M+H]<sup>+</sup>.

**2-Phenyldiimidazo[1,2-*a*:1',2'-*c*]quinazoline (5c):** Yield 58%; Off-white solid; mp 188-190 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.55 (d, *J* = 7.6 Hz, 1H), 8.24 (s, 1H), 7.99 (d, *J* = 7.3 Hz, 2H), 7.66 (d, *J* = 1.3 Hz, 1H), 7.62 (d, *J* = 7.9 Hz, 1H), 7.60 – 7.55 (m, 1H), 7.51 – 7.44 (m, 3H), 7.38 – 7.33 (m, 1H), 7.31 (d, *J* = 1.3 Hz, 1H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 144.65, 140.22, 136.63, 133.10, 130.32, 130.02, 128.78, 128.70, 127.94, 126.06, 125.77, 125.10, 114.85, 114.72, 110.76, 108.03; HRMS (ESI) calcd for C<sub>18</sub>H<sub>13</sub>N<sub>4</sub> 285.1135 found 285.1131 [M+H]<sup>+</sup>.

**6-Phenylimidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5d):** Yield 52%; Brown solid; mp 203-205 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.54 (d, *J* = 8.0 Hz, 1H), 8.17 (d, *J* = 8.2 Hz, 1H), 8.14 (s, 1H), 8.11 (s, 1H), 7.93 – 7.88 (m, 1H), 7.68 – 7.62 (m, 1H), 7.56 – 7.51 (m, 1H), 7.38 (t, *J* = 7.6 Hz, 2H), 7.28 (t, *J* = 7.4 Hz, 1H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 151.29, 145.73, 143.24, 141.19, 132.57, 130.85, 130.83, 128.86, 128.38, 127.19, 125.96, 124.77, 115.62, 114.38, 108.25; HRMS (ESI) calcd for C<sub>17</sub>H<sub>12</sub>N<sub>5</sub> 286.1087 found 286.1094 [M+H]<sup>+</sup>.

**2,3-Bis(4-fluorophenyl)diimidazo[1,2-*a*:1',2'-*c*]quinazoline (5e):** Yield 81%; Off-white solid; mp 250-253 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.71 (d, *J* = 7.9 Hz, 1H), 7.78 – 7.66 (m, 3H), 7.62 – 7.52 (m, 5H), 7.26 – 7.15 (m, 3H), 7.05 – 6.96 (m, 2H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 140.77, 139.78, 133.80, 133.72, 131.87, 130.41, 130.25, 129.63, 129.55, 128.83, 128.27, 126.14, 125.78, 125.22, 115.74, 115.52, 115.41, 115.20, 115.03, 114.70, 110.14; HRMS (ESI) calcd for C<sub>24</sub>H<sub>15</sub>F<sub>2</sub>N<sub>4</sub> 397.1259 found 397.1256 [M+H]<sup>+</sup>.

**5,6-Bis(4-fluorophenyl)imidazo[1,2-*c*]benzimidazolo[1,5-*a*]quinazoline (5f):** Yield 65%; Off-white solid; mp 307 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.77 (dd, *J* = 7.9, 1.4 Hz, 1H), 8.44 (d, *J* = 8.4 Hz, 1H), 8.22 (dd, *J* = 7.2, 1.7 Hz, 1H), 7.83 – 7.60 (m, 1H), 7.72 – 7.67 (m, 1H), 7.65 – 7.55 (m, 5H), 7.48 – 7.39 (m, 2H), 7.28 – 7.19 (m, 2H), 7.07 – 6.96 (m, 2H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 142.01, 134.01, 133.93, 132.84, 130.65, 129.74, 129.66, 129.54, 125.48, 125.42, 124.15, 123.32, 120.79, 115.55, 115.46, 115.33, 115.25, 114.86, 114.83, 112.89, 112.85, 112.82; HRMS (ESI) calcd for C<sub>28</sub>H<sub>17</sub>F<sub>2</sub>N<sub>4</sub> 447.1416 found 447.1421 [M+H]<sup>+</sup>.

**5,6-Bis(4-fluorophenyl)imidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5g):** Yield 79%; Off-white solid; mp 264-266 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.72 (dd, *J* = 8.0, 0.9 Hz, 1H), 8.31 (d, *J* = 7.9 Hz, 1H), 8.06 (s, 1H), 7.83 – 7.75 (m, 1H), 7.70 – 7.60 (m, 3H), 7.59 – 7.53 (m, 2H), 7.27 – 7.21 (m, 2H), 7.06 – 7.01 (m, 2H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 164.22 (d, *J* = 95.3 Hz), 161.74 (d, *J* = 93.1 Hz), 151.17, 143.66, 141.49, 140.52, 133.72 (d, *J* = 8.4 Hz), 130.96, 130.82, 129.65 (d, *J* = 8.1 Hz), 129.15 (d, *J* = 2.9 Hz), 127.22, 124.77, 124.76 (d, *J* = 3.6 Hz), 123.36, 115.91 (d, *J* = 21.8 Hz), 115.61, 115.45 (d, *J* = 21.5 Hz), 114.42; HRMS (ESI) calcd for C<sub>23</sub>H<sub>14</sub>F<sub>2</sub>N<sub>5</sub> 398.1212 found 398.1206 [M+H]<sup>+</sup>.

**2,3-Di(p-tolyl)diimidazo[1,2-*a*:1',2'-*c*]quinazoline (5h):** Yield 56%; Brown solid; mp 224 °C; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.70 (d, *J* = 7.8 Hz, 1H), 7.72 – 7.58 (m, 3H), 7.58 – 7.44 (m, 5H), 7.31 (d, *J* = 7.8 Hz, 2H), 7.18 (s, 1H), 7.11 (d, *J* = 7.9 Hz, 2H), 2.49 (s, 3H), 2.34 (s, 3H); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 141.26, 139.42, 138.73, 137.19, 137.05, 131.65, 130.82, 130.29, 129.95, 129.13, 128.99, 128.87, 127.77, 126.95, 125.98, 125.22, 124.55, 115.10, 114.60, 109.95, 21.74, 21.29; HRMS (ESI) calcd for C<sub>26</sub>H<sub>21</sub>N<sub>4</sub> 389.1761 found 389.1764 [M+H]<sup>+</sup>.

**5,6-Di(p-tolyl)imidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5i):** Yield 61%; Off-white solid; mp 266-268 °C; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.72 (d, *J* = 7.8 Hz, 1H), 8.26 (d, *J* = 8.2 Hz, 1H), 8.05 (s, 1H), 7.74 (t, *J* = 7.7 Hz, 1H), 7.63 (t, *J* = 7.6 Hz, 1H), 7.55 (d, *J* = 8.1 Hz, 2H),

7.48 (d,  $J = 7.9$  Hz, 2H), 7.35 (d,  $J = 7.9$  Hz, 2H), 7.12 (d,  $J = 8.0$  Hz, 2H), 2.51 (s, 3H), 2.35 (s, 3H);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ )  $\delta$  151.26, 143.83, 142.00, 140.18, 139.31, 137.51, 131.58, 130.71, 130.65, 130.31, 129.32, 129.07, 127.78, 127.08, 125.97, 124.73, 124.45, 115.51, 114.54, 21.76, 21.30; HRMS (ESI) calcd for  $\text{C}_{25}\text{H}_{20}\text{N}_5$  390.1713 found 390.1719  $[\text{M}+\text{H}]^+$ .

**2,3-Bis(4-methoxyphenyl)diimidazo[1,2-*a*:1',2'-*c*]quinazoline (5j):** Yield 56%; Brown solid; mp 223 °C;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  8.69 (d,  $J = 7.8$  Hz, 1H), 7.71 – 7.63 (m, 2H), 7.63 – 7.48 (m, 6H), 7.18 (s, 1H), 7.03 (d,  $J = 7.8$  Hz, 2H), 6.84 (d,  $J = 8.0$  Hz, 2H), 3.91 (s, 3H), 3.81 (s, 3H);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ )  $\delta$  159.92, 158.93, 141.03, 139.28, 137.21, 133.13, 130.26, 129.92, 129.09, 128.84, 126.30, 125.99, 125.17, 123.80, 122.09, 115.06, 114.61, 113.81, 113.72, 109.95, 55.23, 55.20; HRMS (ESI) calcd for  $\text{C}_{26}\text{H}_{21}\text{N}_4\text{O}_2$  421.1659 found 421.1658  $[\text{M}+\text{H}]^+$ .

**5,6-Di(*p*-tolyl)imidazo[1,2-*c*]benzimidazolo[1,5-*a*]quinazoline (5k):** Yield 52%; Brown solid; mp 253-255 °C;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  8.77 (d,  $J = 7.6$  Hz, 1H), 8.37 (d,  $J = 8.2$  Hz, 1H), 8.16 (d,  $J = 7.1$  Hz, 1H), 7.71 (dd,  $J = 16.1, 7.8$  Hz, 2H), 7.57 (dd,  $J = 19.1, 8.3$  Hz, 5H), 7.43 – 7.33 (m, 2H), 7.05 (d,  $J = 8.1$  Hz, 2H), 6.85 (d,  $J = 8.3$  Hz, 2H), 3.96 (s, 3H), 3.82 (s, 3H);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ )  $\delta$  159.97, 159.07, 142.09, 141.35, 140.31, 133.38, 132.69, 132.27, 130.39, 130.12, 129.23, 125.94, 125.50, 125.32, 124.68, 123.97, 123.13, 122.05, 120.81, 115.01, 114.74, 113.75, 113.66, 112.80, 55.32, 55.25; HRMS (ESI) calcd for  $\text{C}_{30}\text{H}_{23}\text{N}_4\text{O}_2$  471.1816 found 471.1816  $[\text{M}+\text{H}]^+$ .

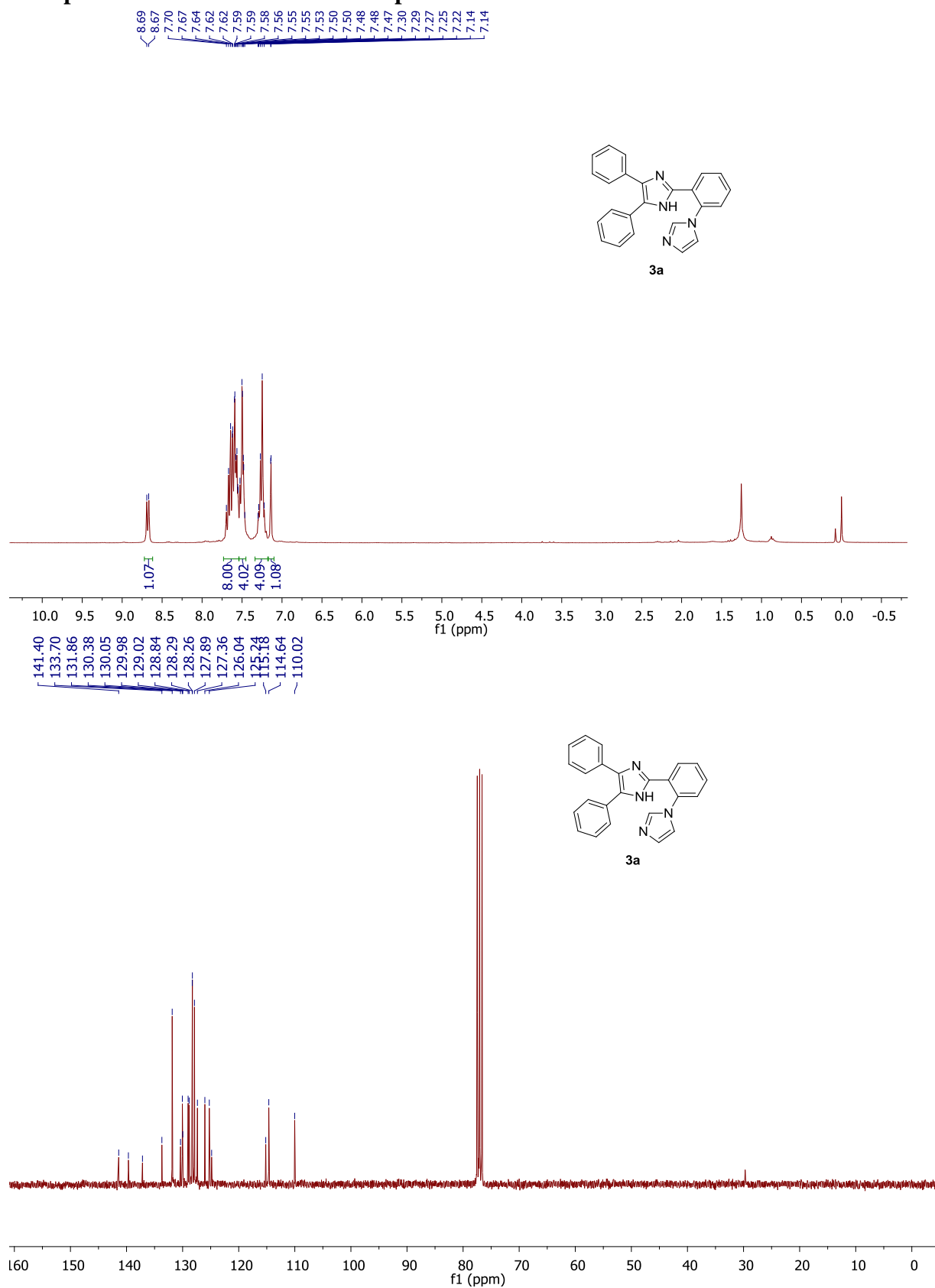
**5,6-Bis(4-methoxyphenyl)imidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5l):** Yield 58%; Brown solid; mp 226 °C;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  8.69 (d,  $J = 7.8$  Hz, 1H), 8.26 (d,  $J = 8.1$  Hz, 1H), 8.05 (s, 1H), 7.73 (t,  $J = 7.7$  Hz, 1H), 7.66 – 7.56 (m, 3H), 7.50 (d,  $J = 8.3$  Hz, 2H), 7.06 (d,  $J = 8.3$  Hz, 2H), 6.85 (d,  $J = 8.4$  Hz, 2H), 3.93 (s, 3H), 3.82 (s, 3H);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ )  $\delta$  160.25, 159.19, 151.24, 143.85, 141.82, 140.07, 133.08, 130.68, 130.58, 129.11, 127.06, 125.90, 124.64, 123.68, 121.07, 115.51, 114.55, 114.00, 113.80, 55.25; HRMS (ESI) calcd for  $\text{C}_{25}\text{H}_{20}\text{N}_5\text{O}_2$  422.1612 found 422.1619  $[\text{M}+\text{H}]^+$ .

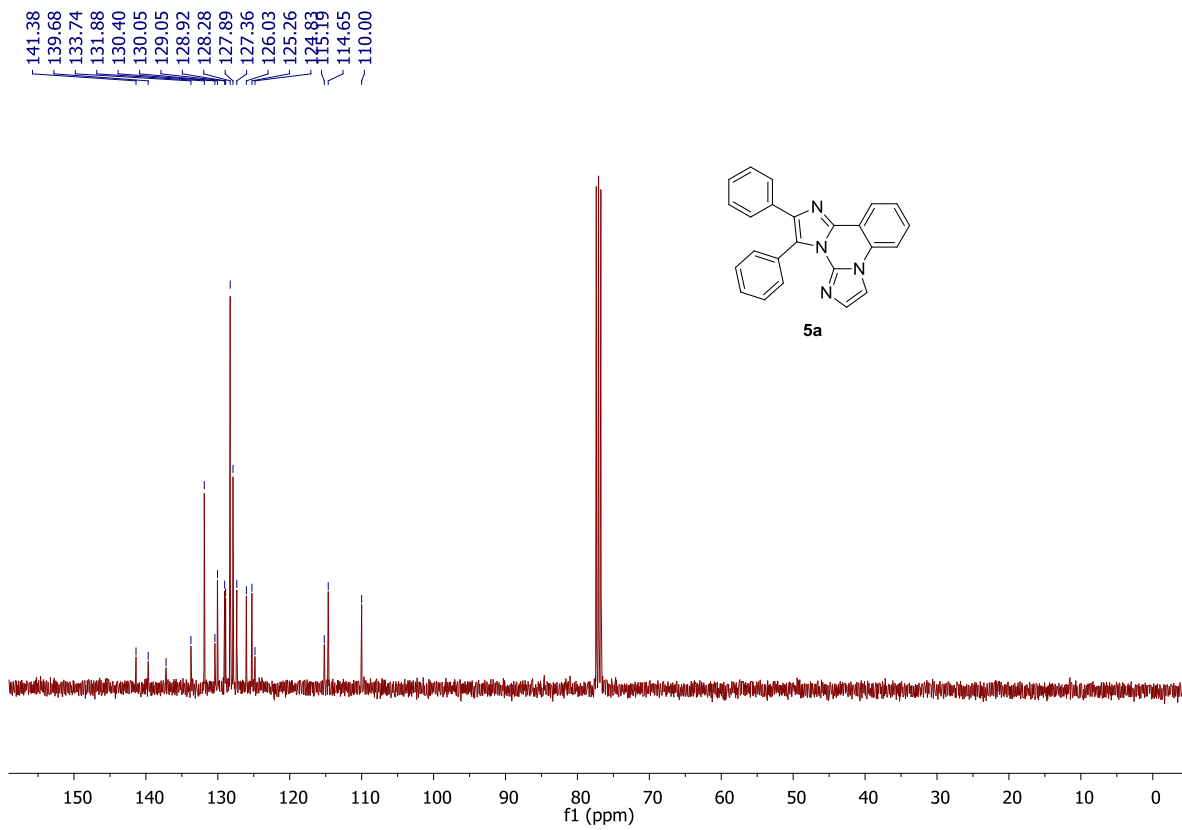
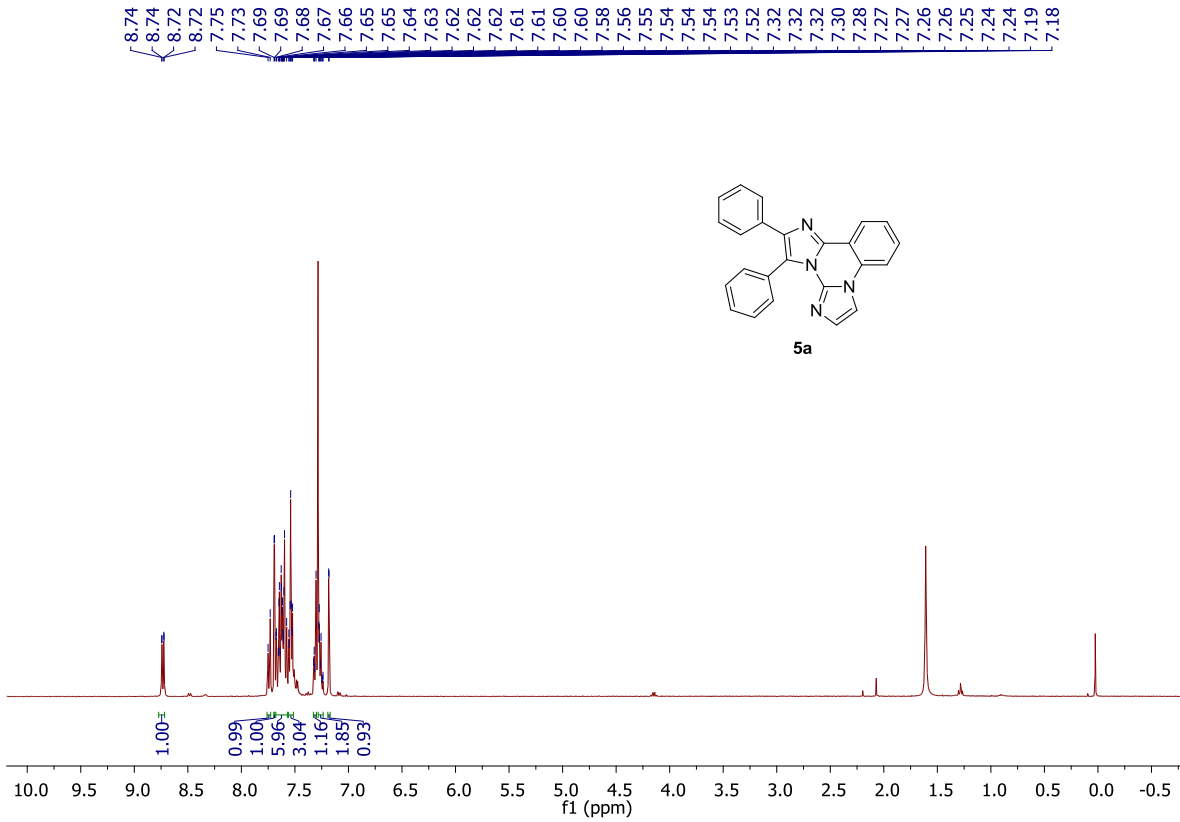
**Benzo[4,5]imidazo[1,2-*c*]imidazo[1,2-*a*]quinazoline (5m):** Yield 62%; Brown solid; mp 192-195 °C;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  8.60 (dd,  $J = 14.9, 5.6$  Hz, 2H), 7.95 (d,  $J = 5.5$  Hz, 1H), 7.67 – 7.59 (m, 3H), 7.55 – 7.43 (m, 3H), 7.36 (s, 1H);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ )  $\delta$  143.82, 143.25, 137.53, 131.87, 131.79, 129.69, 128.93, 126.38, 126.11, 125.03, 124.17, 119.55, 114.84, 114.43, 114.23, 110.09; HRMS (ESI) calcd for  $\text{C}_{16}\text{H}_{11}\text{N}_4$  259.0978 found 259.0984  $[\text{M}+\text{H}]^+$ .

**Benzo[4,5]imidazo[1,2-*a*]benzo[4,5]imidazo[1,2-*c*]quinazoline (5n):** Yield 68%; Off-white solid; mp 221 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.76 – 8.70 (m, 1H), 8.67 (dd, *J* = 7.9, 1.4 Hz, 1H), 8.23 (d, *J* = 8.4 Hz, 1H), 8.01 (d, *J* = 7.7 Hz, 1H), 7.97 – 7.91 (m, 1H), 7.84 (dd, *J* = 7.8, 0.9 Hz, 1H), 7.74 – 7.68 (m, 1H), 7.56 – 7.51 (m, 2H), 7.50 – 7.45 (m, 1H), 7.42 – 7.32 (m, 2H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 144.52, 143.77, 142.50, 141.62, 134.07, 132.00, 130.32, 130.05, 126.57, 125.35, 125.33, 124.39, 124.25, 123.08, 120.03, 119.61, 114.99, 114.91, 114.46, 112.64; HRMS (ESI) calcd for C<sub>20</sub>H<sub>13</sub>N<sub>4</sub> 309.1135 found 309.1132 [M+H]<sup>+</sup>.

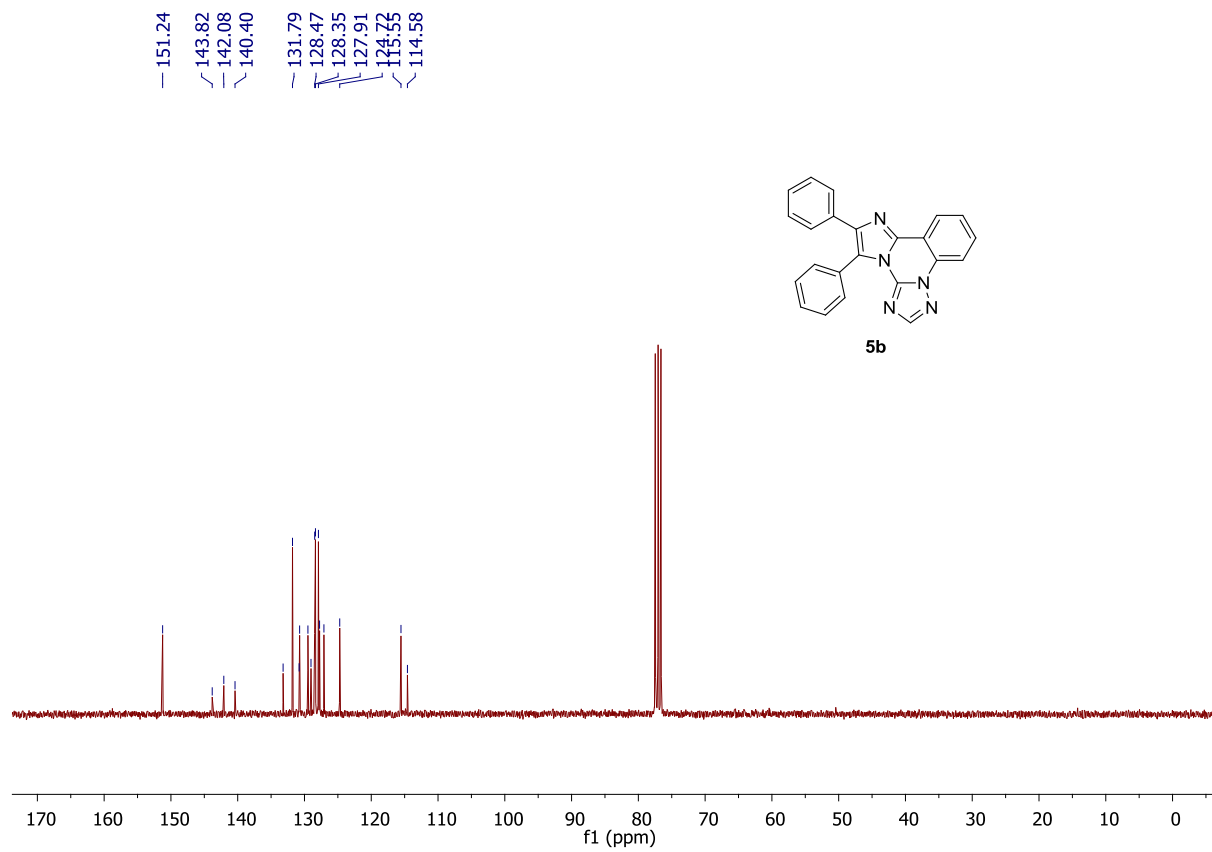
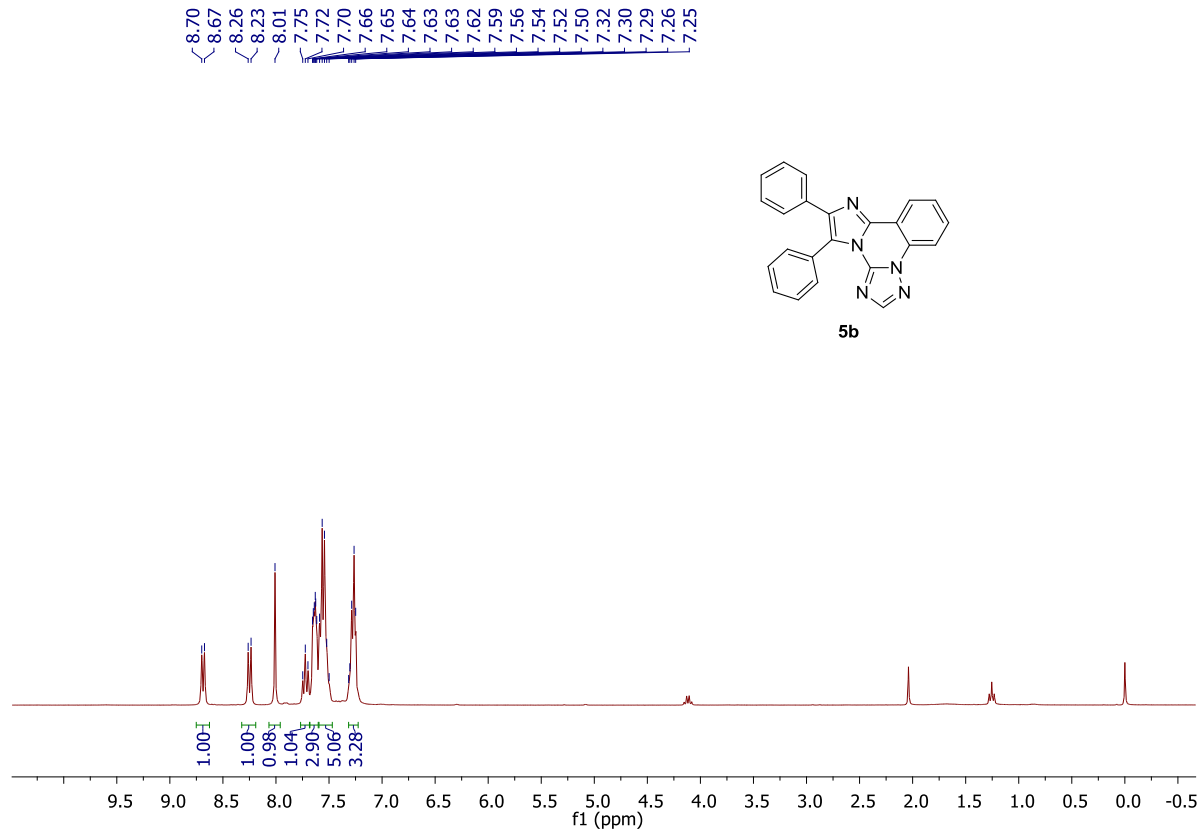
**Benzo[4,5]imidazo[1,2-*c*][1,2,4]triazolo[1,5-*a*]quinazoline (5o):** Yield 61%; Off-white solid; mp 170-172 °C; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.60 (d, *J* = 7.9 Hz, 1H), 8.42 – 8.34 (m, 1H), 8.19 (d, *J* = 7.3 Hz, 2H), 7.94 – 7.87 (m, 1H), 7.75 (t, *J* = 7.8 Hz, 1H), 7.57 (t, *J* = 7.7 Hz, 1H), 7.52 – 7.46 (m, 2H); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 166.64, 151.52, 144.15, 143.88, 143.54, 132.32, 132.13, 129.21, 127.11, 125.84, 125.58, 124.60, 119.97, 115.62, 113.98, 113.61; HRMS (ESI) calcd for C<sub>15</sub>H<sub>10</sub>N<sub>5</sub> 260.0931 found 260.0936 [M+H]<sup>+</sup>.

### 3. Copies of $^1\text{H}$ NMR and $^{13}\text{C}$ NMR spectra of 3a and 5a-o

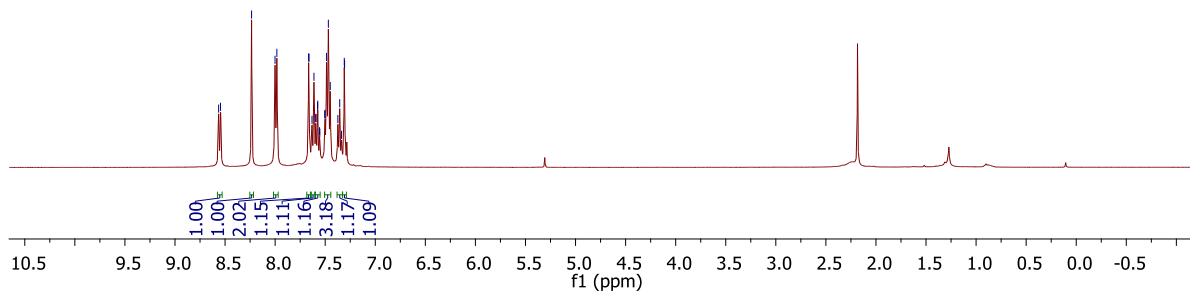
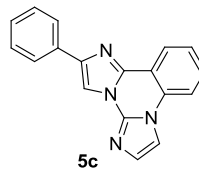




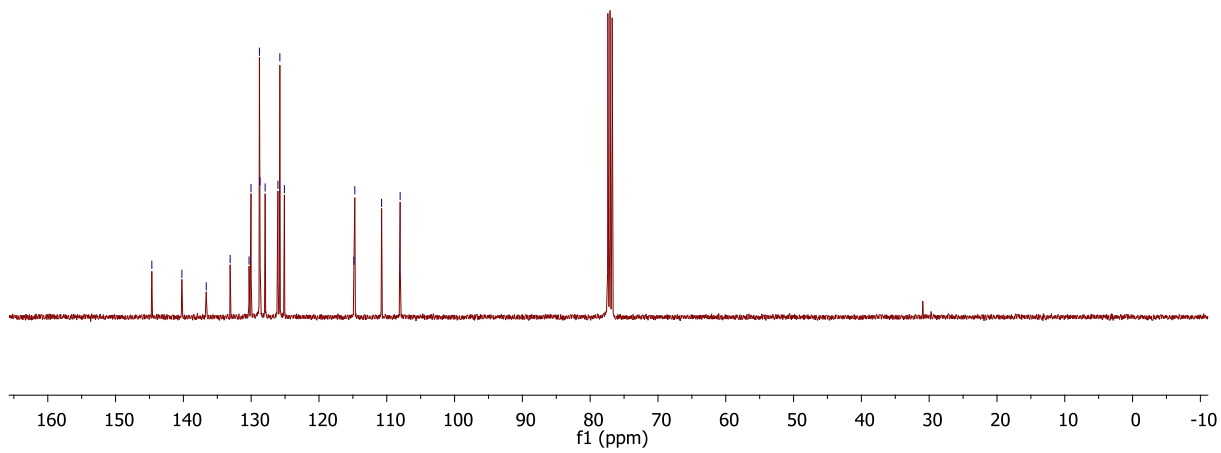
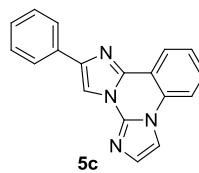




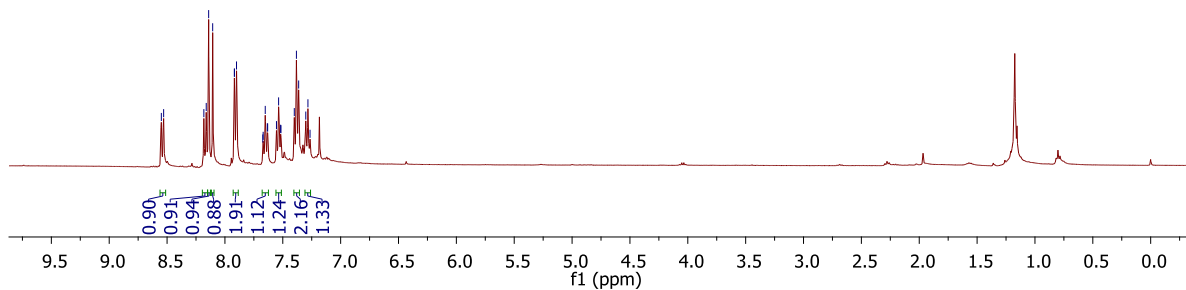
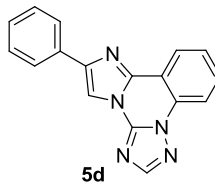
8.56  
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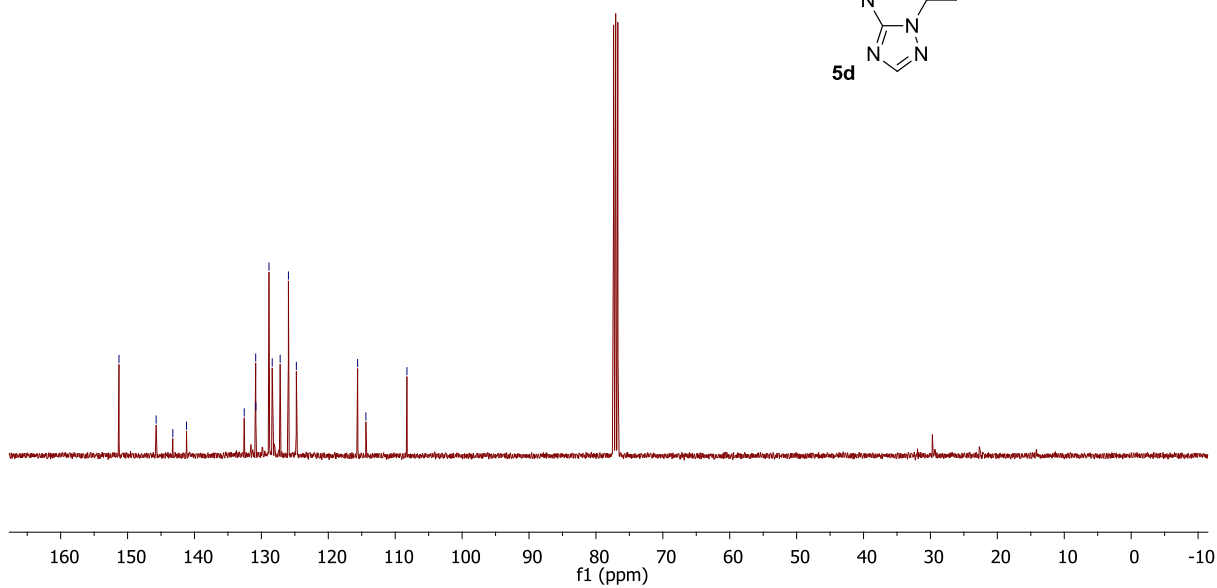
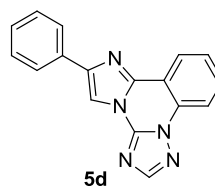
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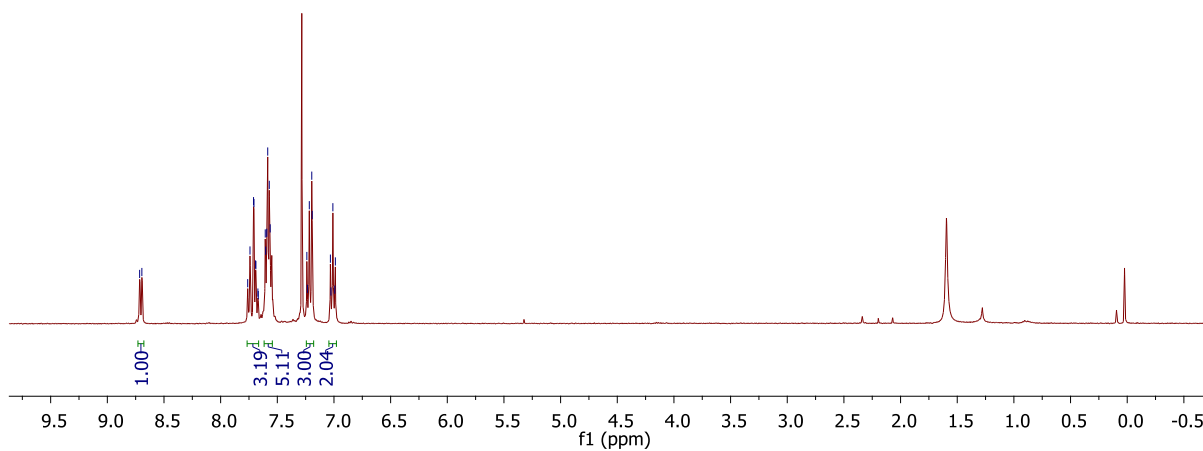
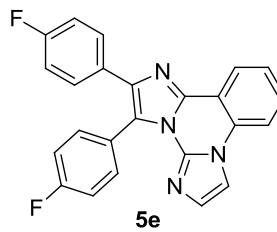
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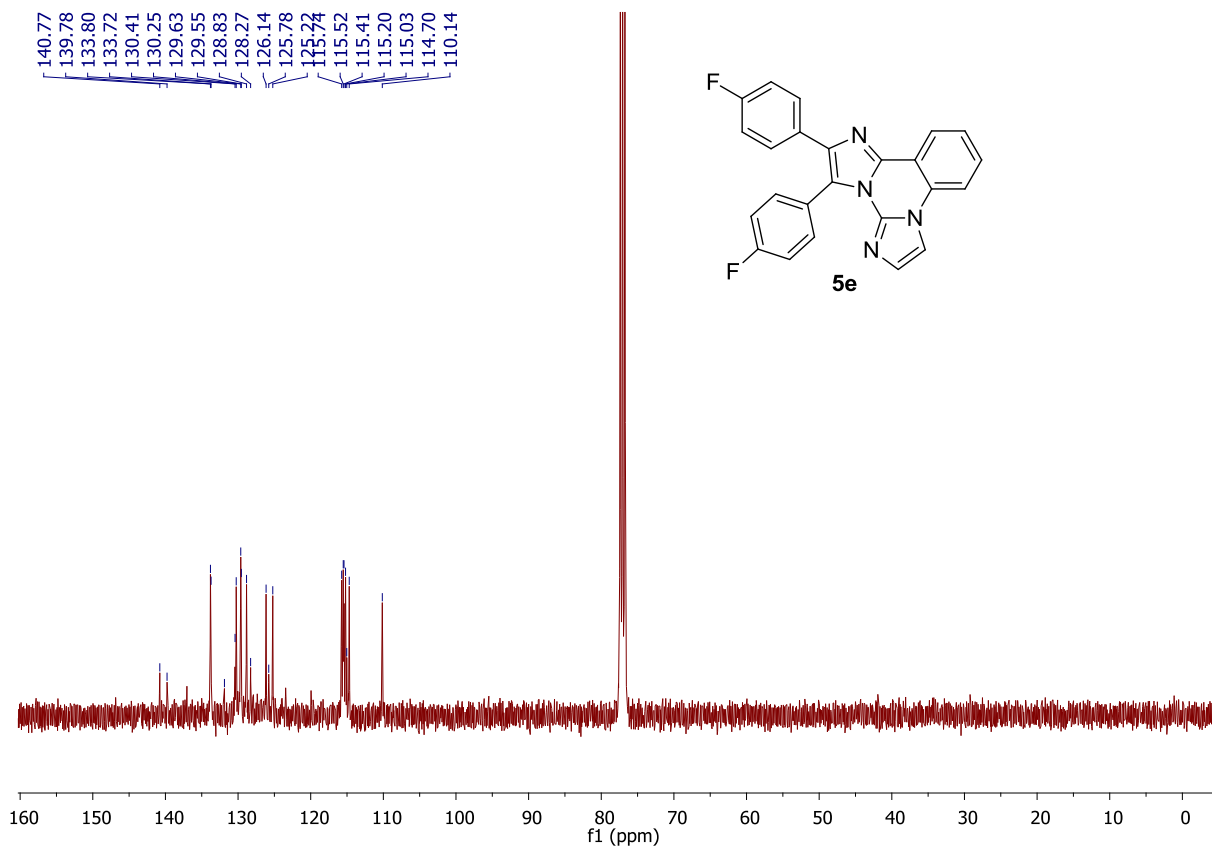
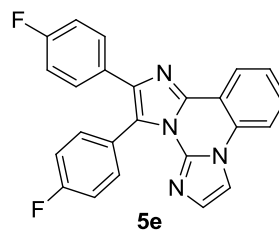
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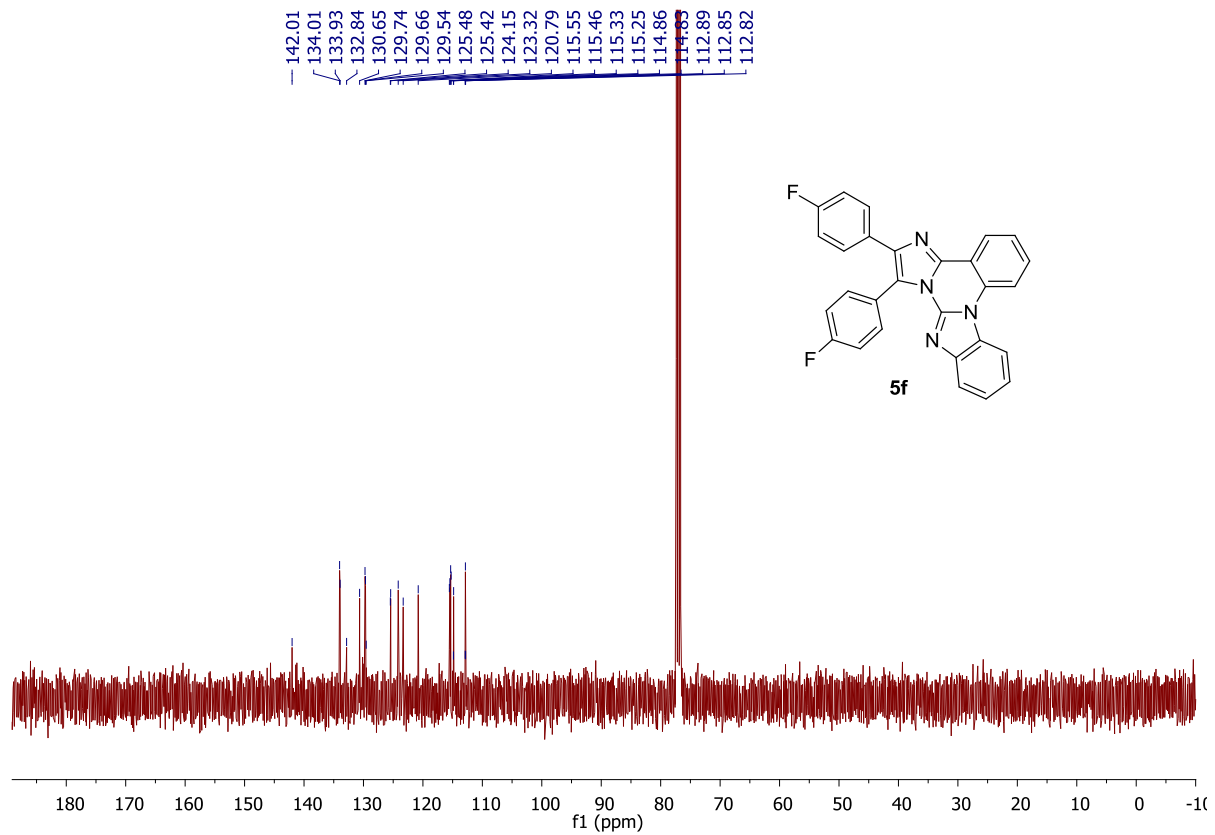
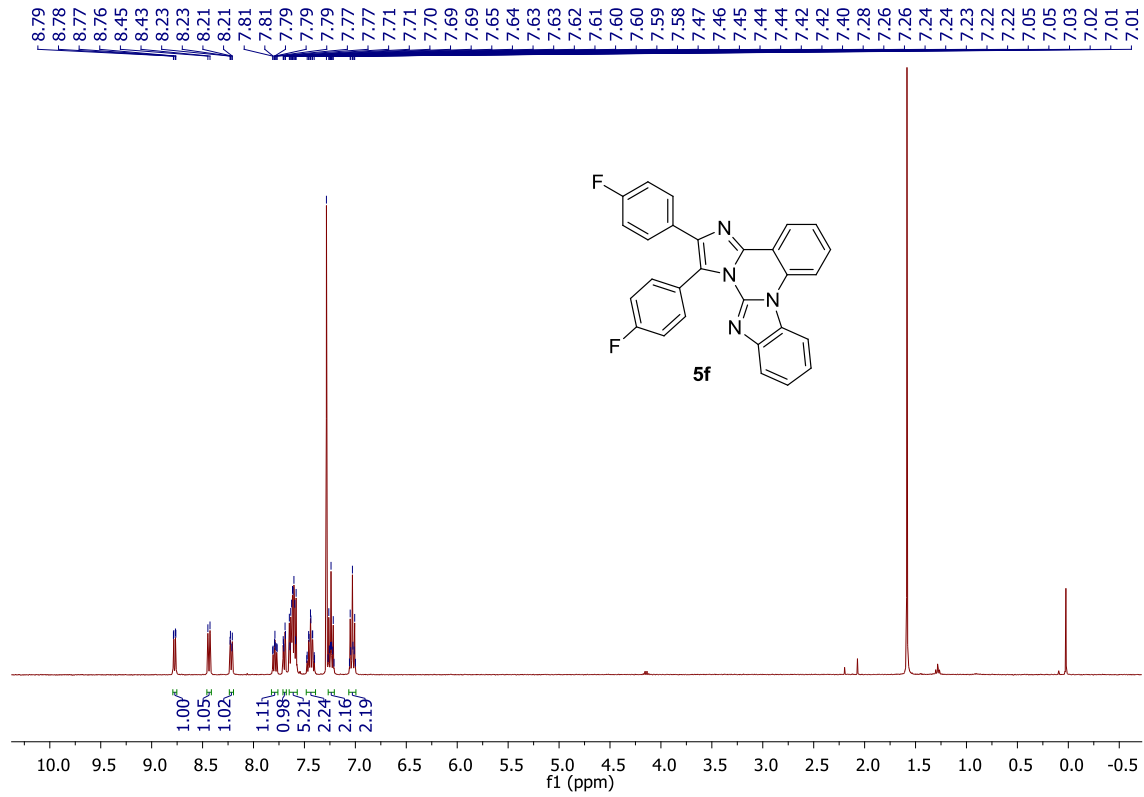


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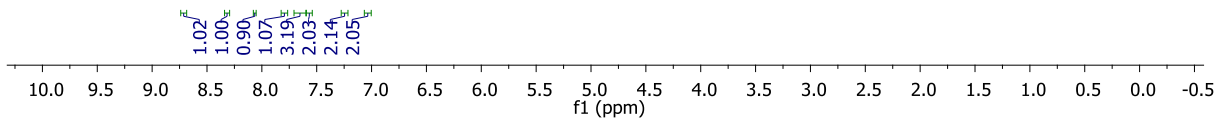
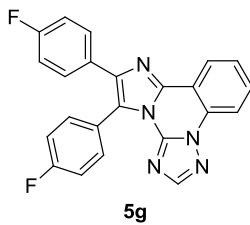


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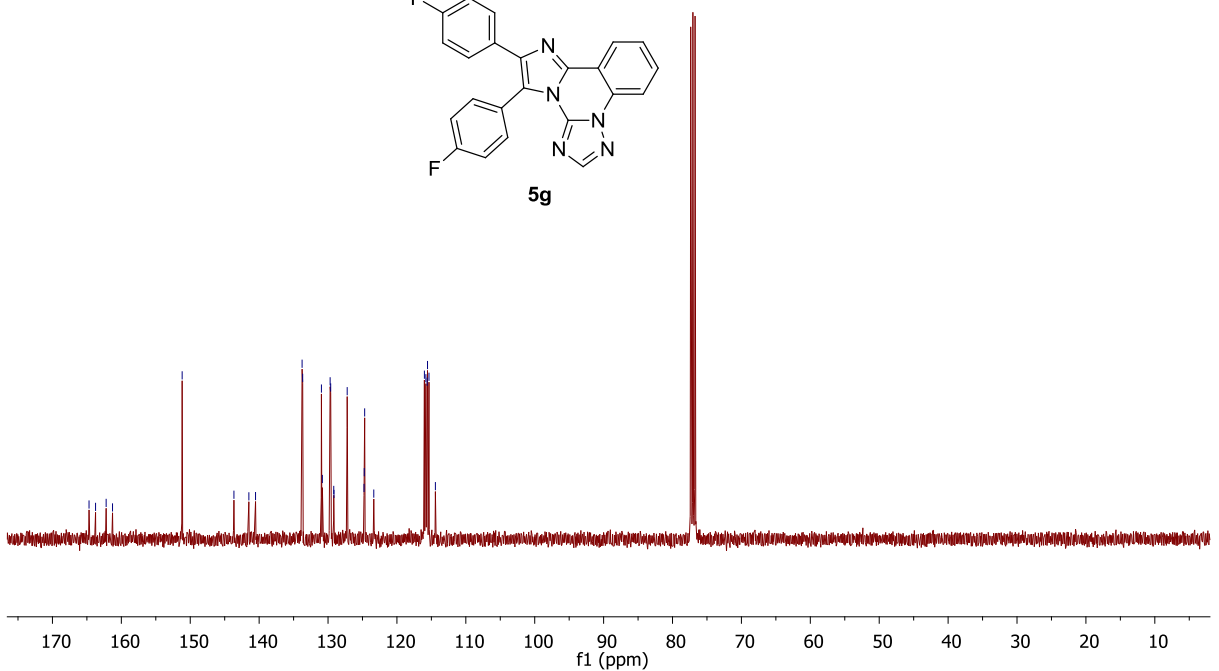
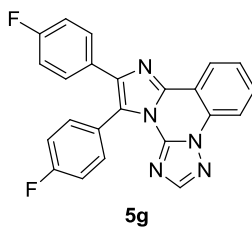


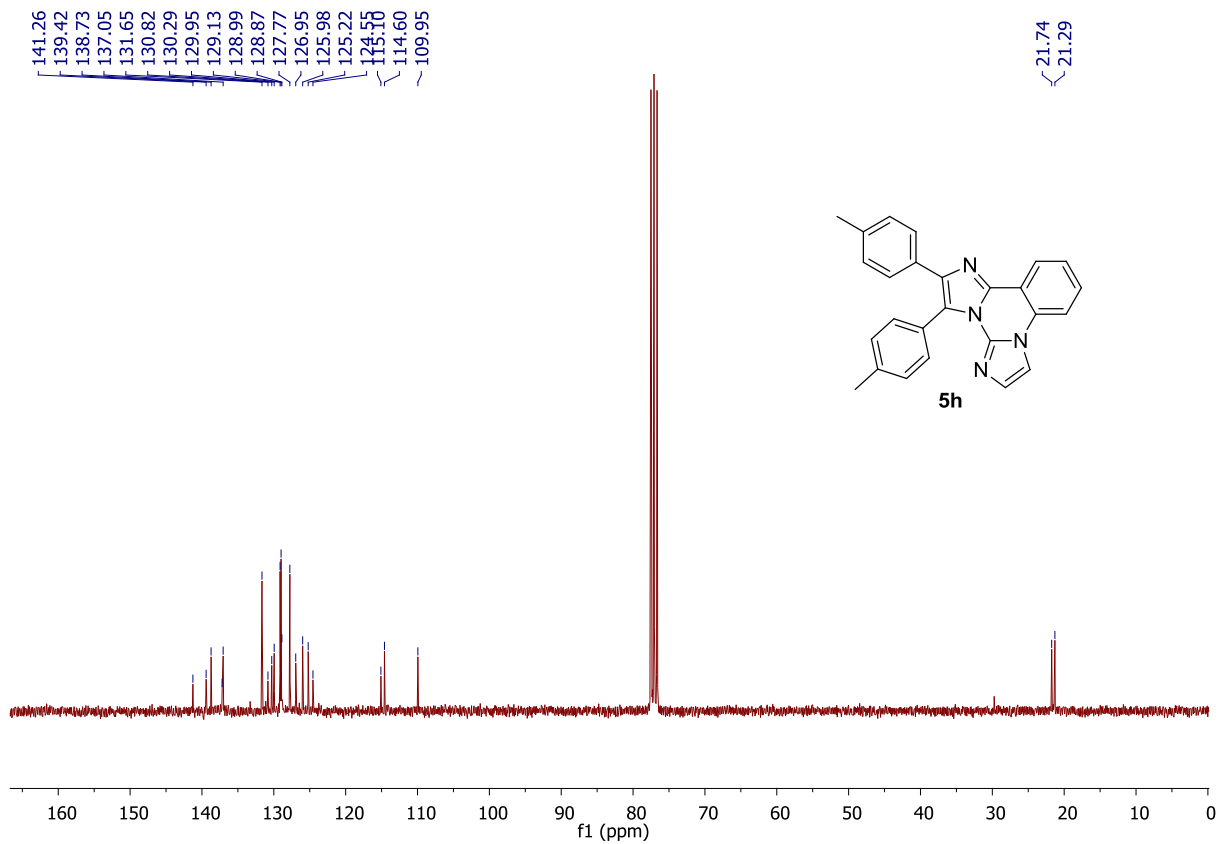
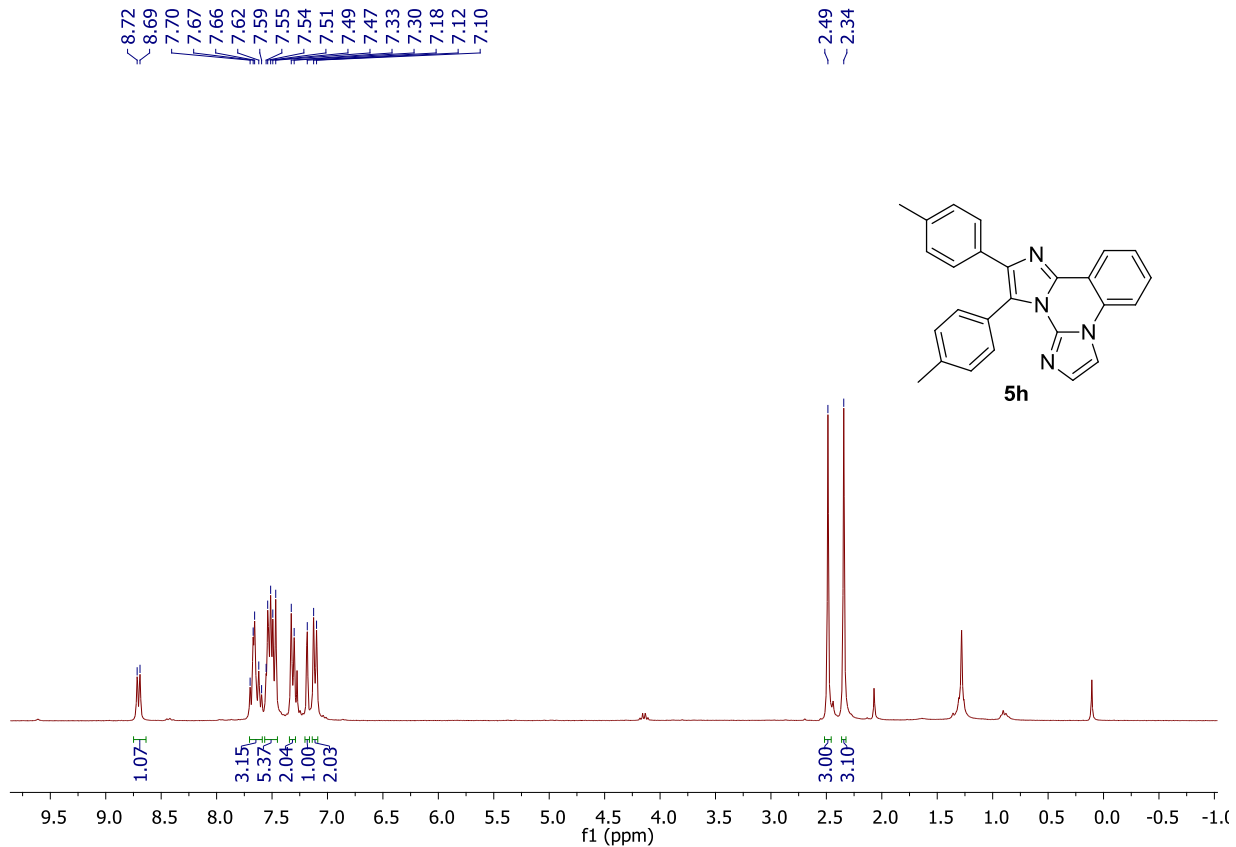


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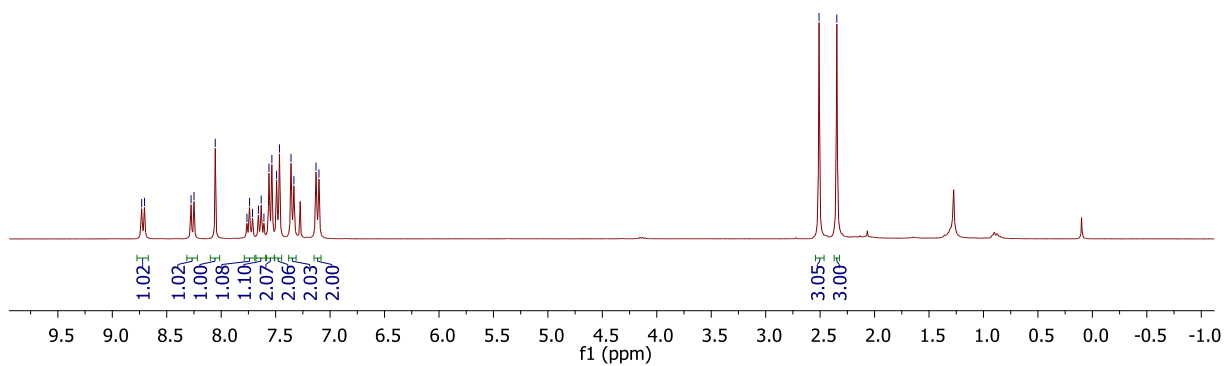
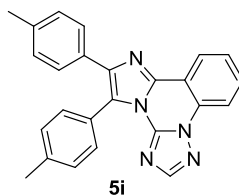
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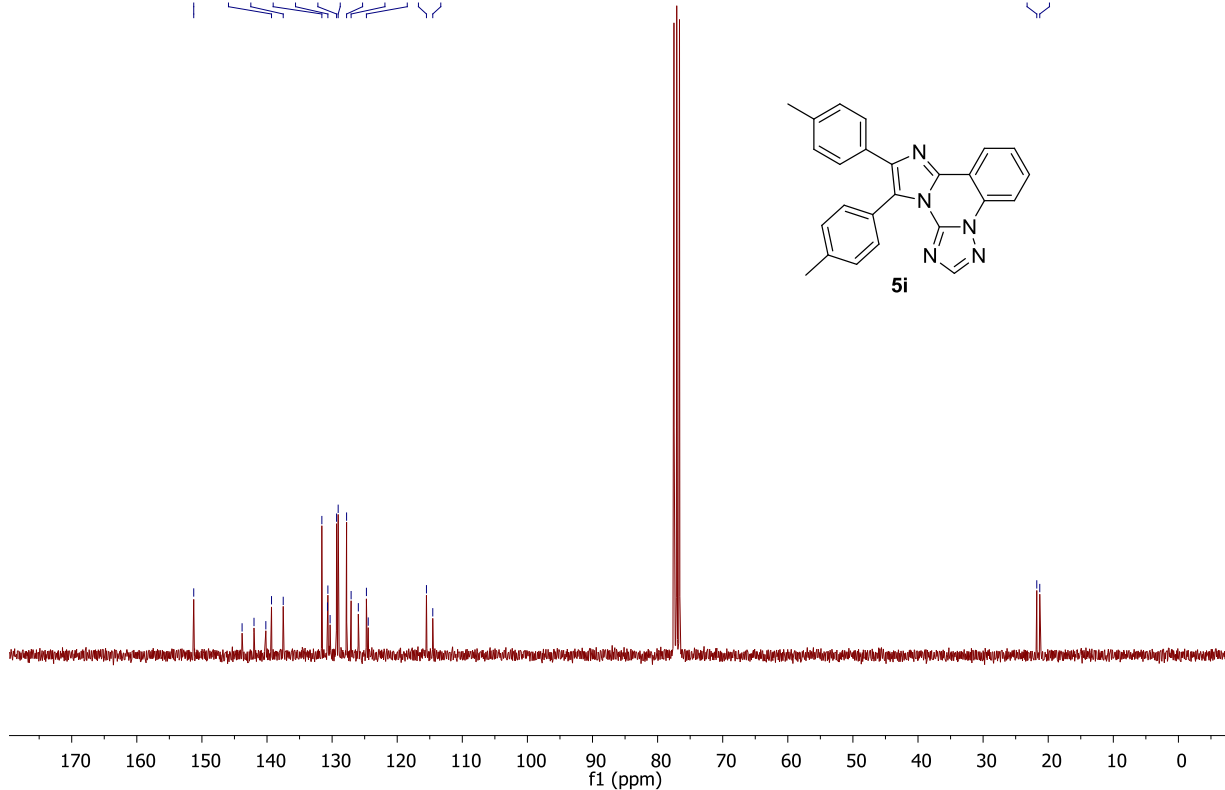
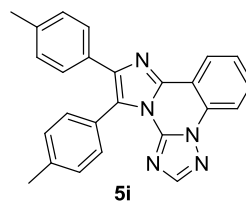
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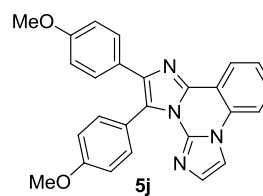
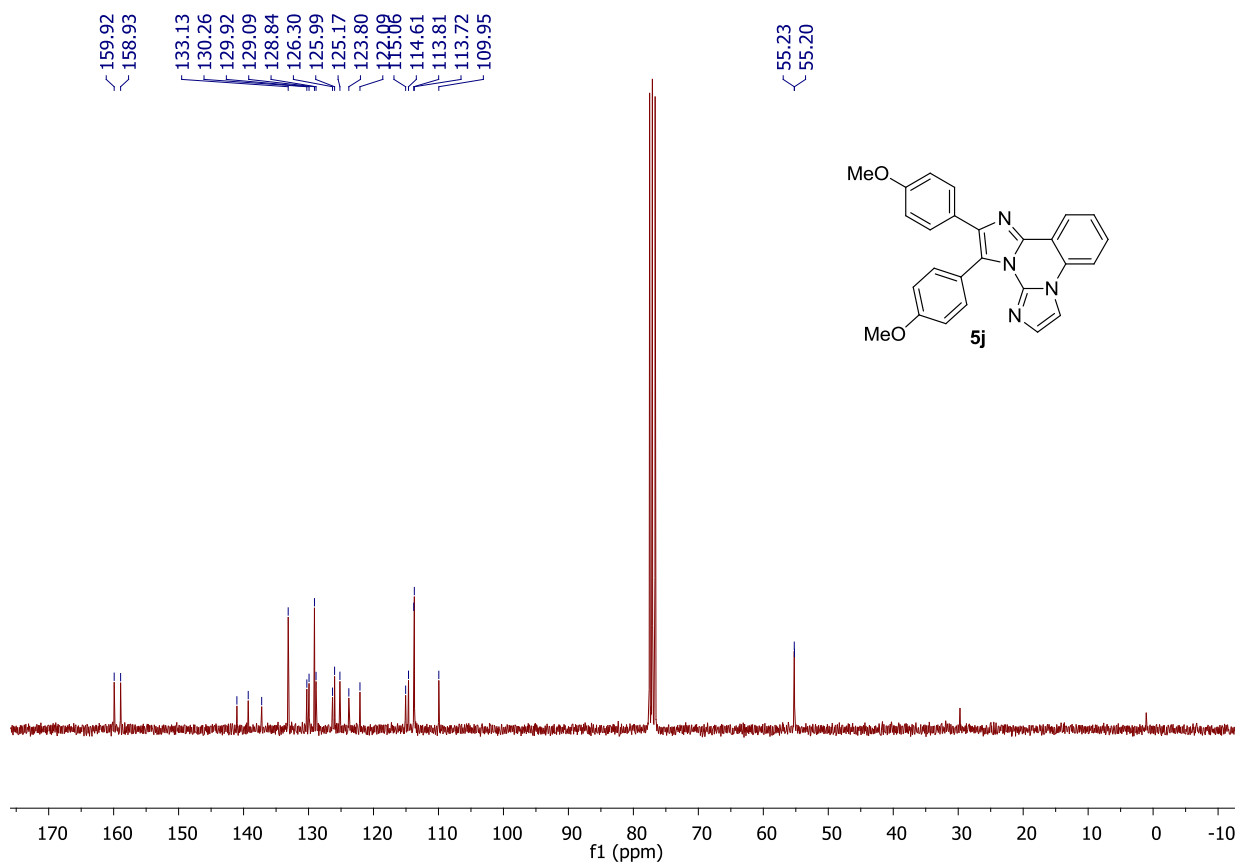
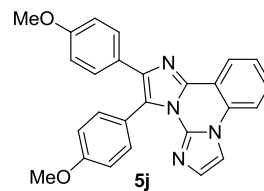
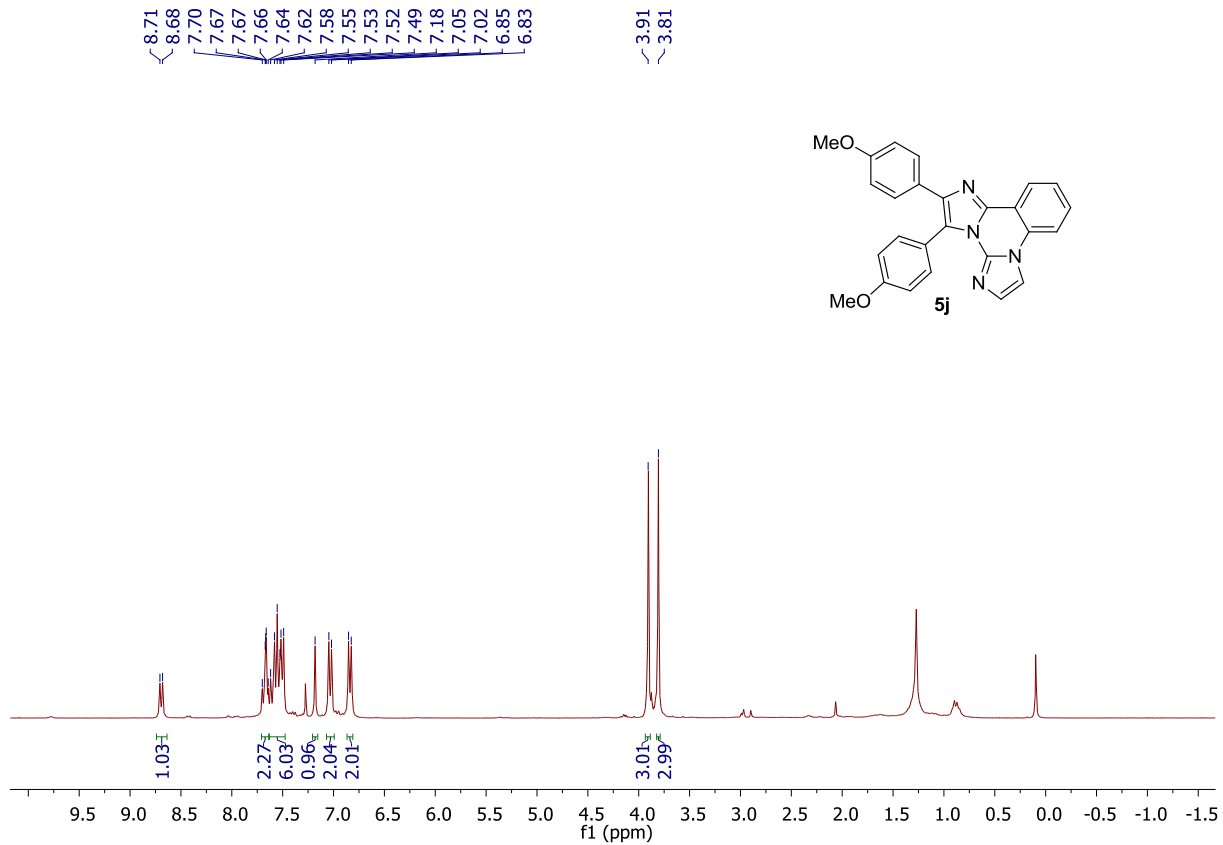


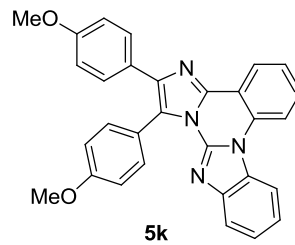
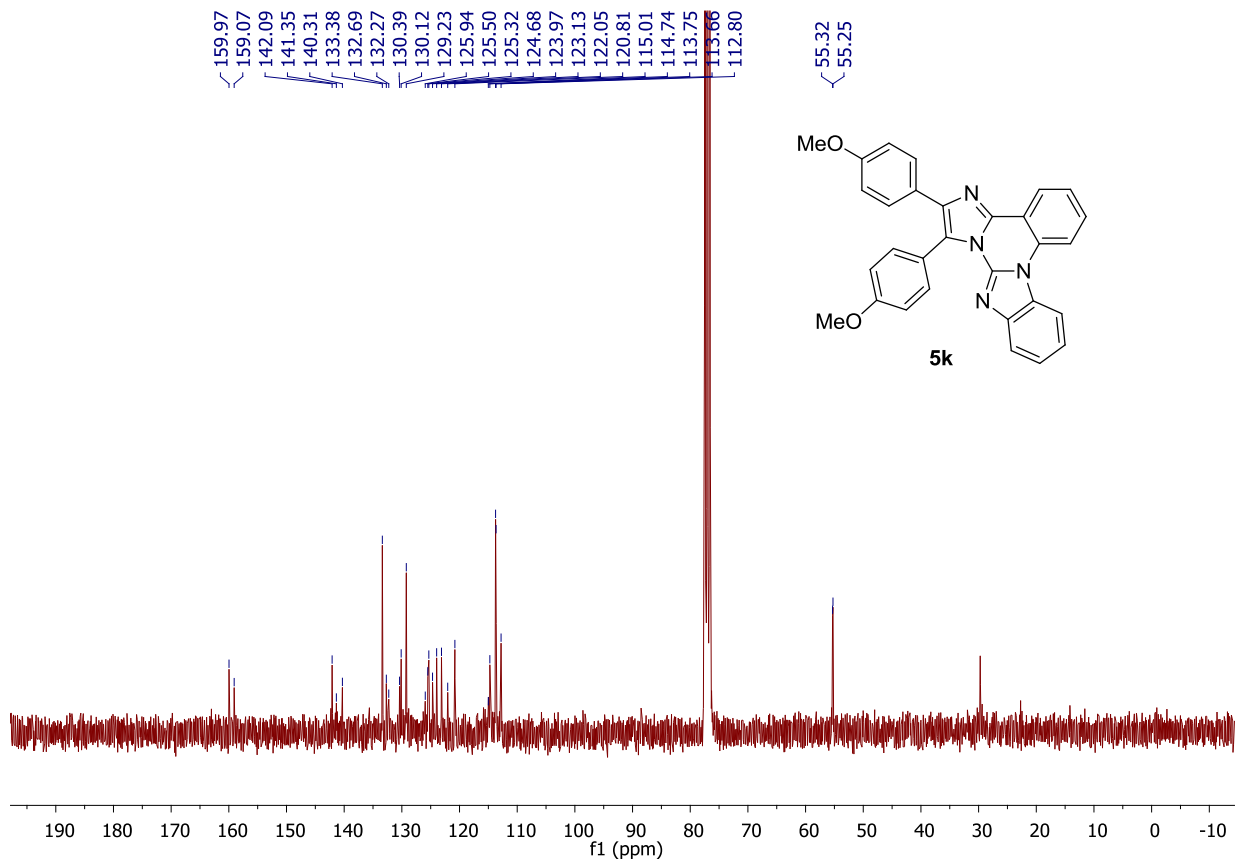
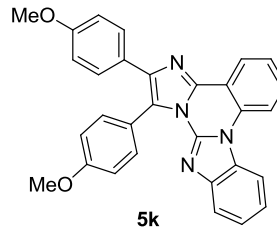
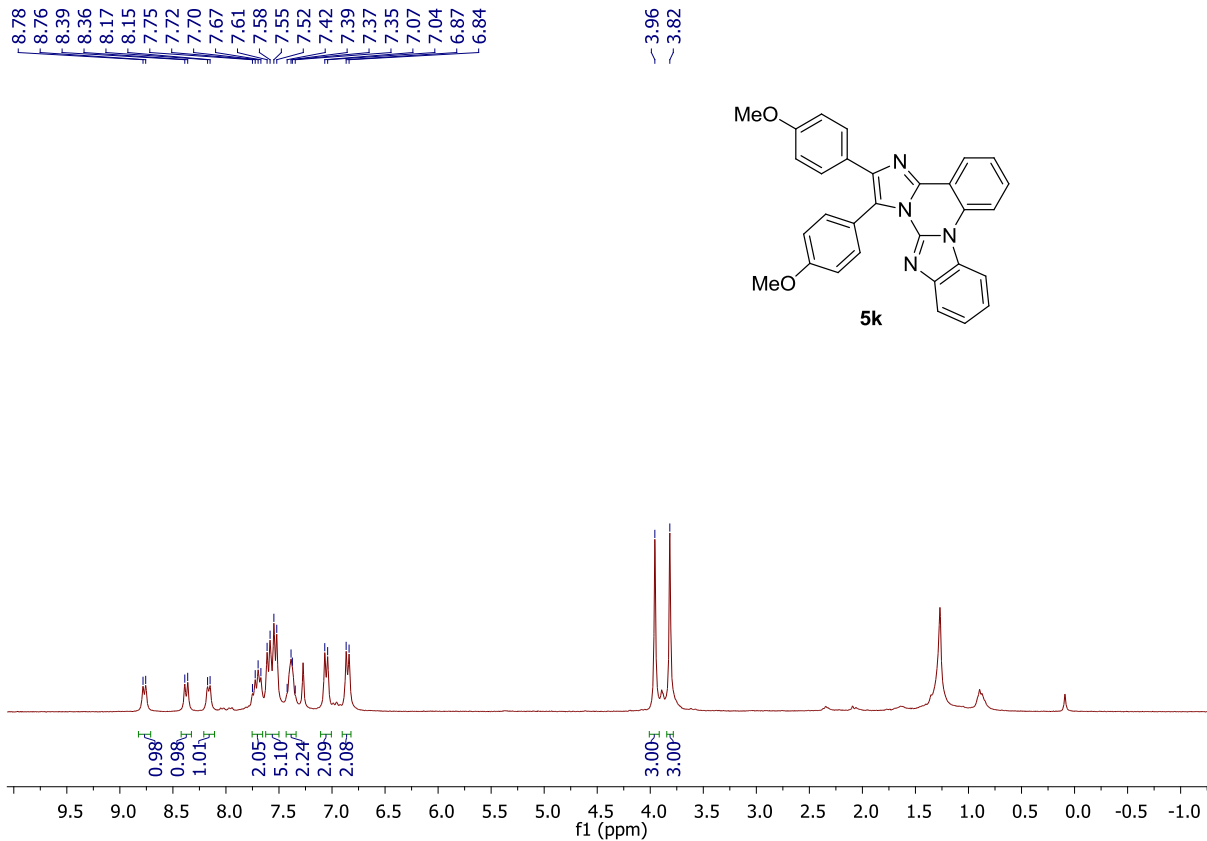
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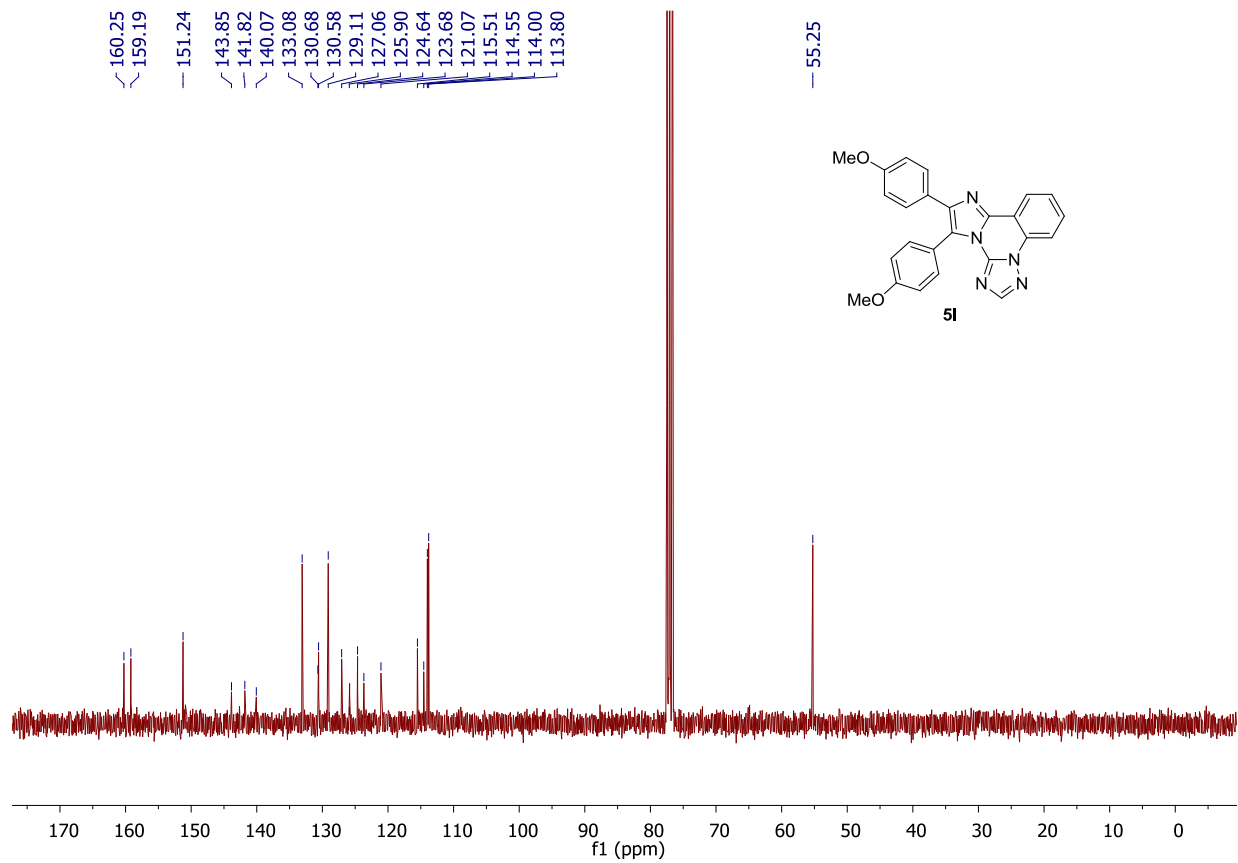
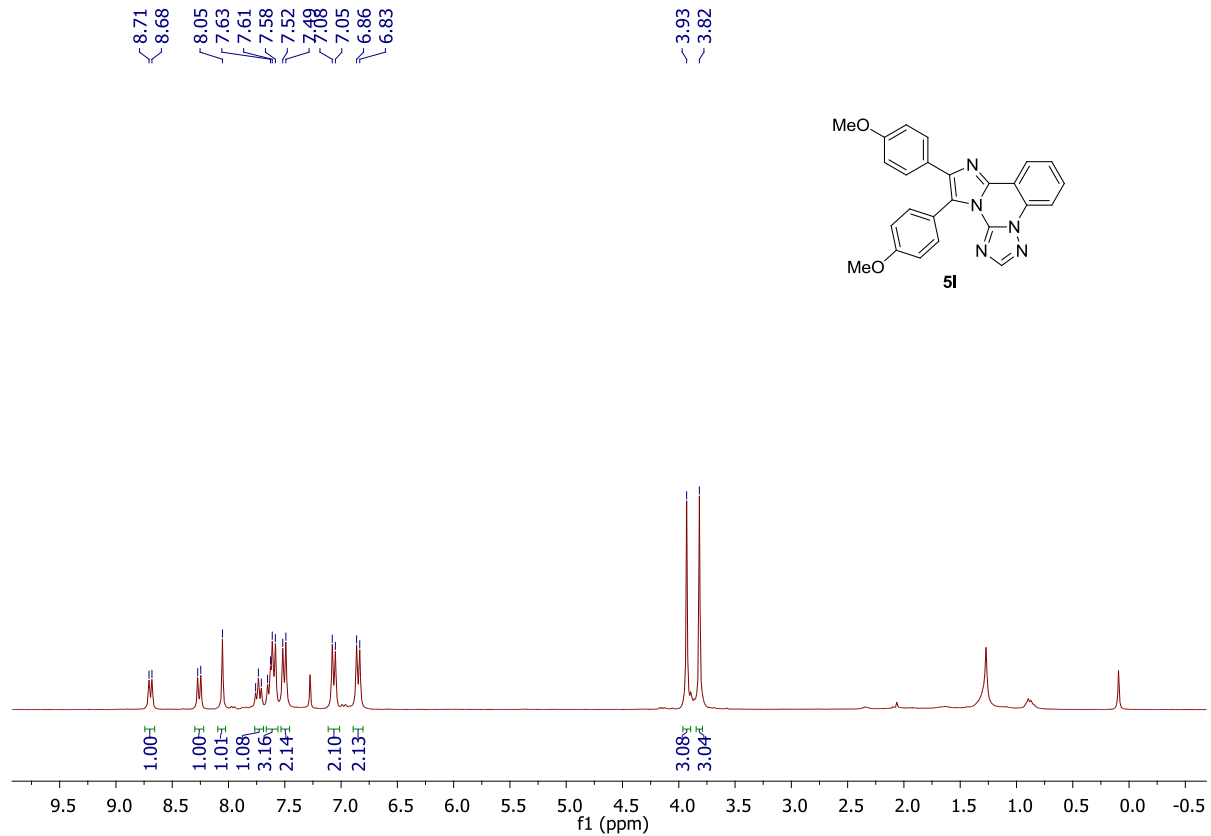
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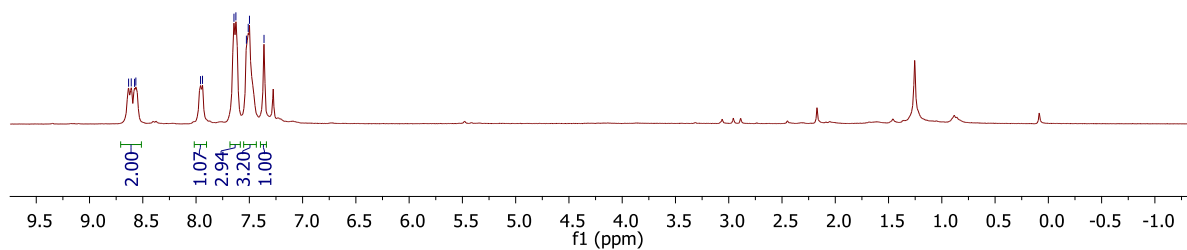
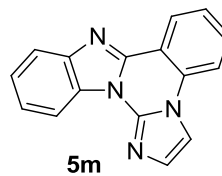








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