

## Supporting Information

### **Cross-Dehydrogenative Coupling of Acetanilides with Aromatic Aldehydes**

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## 1. General information

Solvents, nickel catalyst, TBHP oxidant, aromatic aldehyde and aniline derivatives were purchased from Merck and Sigma. Other reagents were purchased from commercial distributors and used without further purification. Acetanilide derivatives were synthesized in accordance with the literatures. Analytical thin layer chromatography (TLC) was performed with pre-coated silica gel 60 F254 plates. The products were purified by flash column chromatography on silica gel (0.063–0.200 mm; Merck).  $^1\text{H}$  and  $^{13}\text{C}$  NMR Spectra were recorded with a Bruker 500 Advance instrument in  $\text{CDCl}_3$  and  $\text{DMSO-d}_6$ . Mass spectrometry was obtained with an Agilent 5975C VL MSD (Ion source: EI+, 70 eV, 230 °C). Gc-MS analysis was performed on an Agilent Gc 7890A (Column: Rtx 5 MS, length=30m, I.D=0.250 mm, Film thickness=25  $\mu\text{m}$ ) and 5975C VL MSD (Ion source: EI+, 70 eV, 230 °C). Temperature program: initial temperature=40 °C, initial time=2 min, program rate=5 °C/min, final temperature=270 °C, final time=2 min, split ratio=50 mL/min and flow rate=1 mL/min. Conditions: 1. injection port temperature: 230°C, 2. ion source temperature: 230°C, 3. carrier gas: He 99.999%, 4. sample volume: 0.5  $\mu\text{L}$

## 2. Typical procedure for the synthesis of N-Acetyl-N-phenylbenzamides.

A 10 mL microwave vial was charged with acetanilide derivatives (1 equiv, 0.5 mmol), benzaldehyde derivatives (3 equiv, 1.5 mmol), KBr (1.0 equiv., 60 mg), TBHP (70% aq. solution, 1.5 mmol, 193 mg) and DCM (2.0 mL). The vial was then stirred at room temperature, for 12 h. After this time the reaction mixture was diluted with water (5 mL) and the aqueous phase was extracted with dichloromethane (3  $\times$  5 mL). The organic extracts were combined and washed with saturated solution of  $\text{NaHCO}_3$  (3  $\times$  5 mL). Next, the organic phase was dried over sodium sulfate and filtered. Concentration of the solution by rotary evaporation under reduced pressure gave a residue which was purified by using flash column chromatography (*n*-hexane: EtOAc, 10:1) to yield the desired products.

### 3. Spectral data

#### 3.1. N-Acetyl-N-phenylbenzamide (3a):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), benzaldehyde (1.5 mmol, 160 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3a** (110 mg, 92 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>): δ 7.96 (d, J = 7.1 Hz, 2H), 7.78 (d, J = 8.2 Hz, 2H), 7.61 – 7.52 (m, 3H), 7.35 (t, J = 7.8 Hz, 2H), 7.10 (t, J = 7.4 Hz, 1H), 2.50 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>): δ 173.3, 173.1, 139.6, 135.5, 132.0, 129.3, 129.1, 129.1, 128.8, 128.1, 25.8. MS (EI): 239 (5) [M]<sup>+</sup>, 197 (99), 167 (70), 149 (99), 105 (100), 84 (44), 77 (98), 71 (56), 66 (40), 57 (80), 55 (50), 43 (72), 41 (54). C<sub>15</sub>H<sub>13</sub>NO<sub>2</sub> (239.27): calcd. C 75.30, H 5.48, N 5.85; found C 75.64, H 5.51, N 5.83

#### 3.2. N-Acetyl-4-methyl-N-phenylbenzamide (3b):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3b** (116 mg, 92 % yield) as a yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ 7.53 (d, J = 7.9 Hz, 2H), 7.33 (t, J = 7.6 Hz, 1H), 7.15 (d, J = 7.7 Hz, 2H), 7.10 (d, J = 8.1 Hz, 2H), 2.41 (s, 3H), 2.32 (s, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>): δ 173.5, 172.8, 143.0, 139.3, 131.8, 129.6, 129.3, 129.0, 128.4, 128.0, 25.5, 21.6. MS (EI): m/z (%) = 253(4) [M]<sup>+</sup>, 211(70), 167(20), 149(51), 119(100), 91(83), 65(53), 57(25), 43(31). C<sub>16</sub>H<sub>15</sub>NO<sub>2</sub> (253.30): calcd. C 75.87, H 5.97, N 5.53; found C 75.51, H 5.94, N 5.59

#### 3.3. N-Acetyl-4-methyl-N-(*p*-tolyl)benzamide (3c):

The general procedure was followed by using 4-methylacetanilide (0.5 mmol, 75 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3c** (130 mg, 97 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ 7.53 (d, J = 8.1 Hz, 2H), 7.14-7.10 (m, 4H), 7.02 (d, J = 8.3 Hz, 2H), 2.38 (s, 3H), 2.32 (s, 3H), 2.30 (s, 3H). <sup>13</sup>C-NMR (126 MHz, CDCl<sub>3</sub>): δ 173.6, 172.9, 142.9, 137.9, 131.9, 130.0, 129.5, 129.4, 129.0, 128.2, 25.5, 21.6, 21.1. MS (EI): m/z (%) = 267(5) [M]<sup>+</sup>, 177(20), 133(57), 119(25), 89(89), 87(47), 73(22), 45(100), 43(21). C<sub>17</sub>H<sub>17</sub>NO<sub>2</sub> (267.32): calcd. C 76.38, H 6.41, N 5.24; found C 76.12, H 6.45, N 5.28

#### 3.4. N-Acetyl-N-(4-methoxyphenyl)-4-methylbenzamide (3d):

The general procedure was followed by using 4-methoxyacetanilide (0.5 mmol, 83 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane :

EtOAc, 10:1) gave final product **3d** (124 mg, 88 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ 7.52 (d, J = 8.3 Hz, 2H), 7.11 (d, J = 7.9 Hz, 2H), 7.06 (d, J = 8.9 Hz, 2H), 6.84 (d, J = 8.9 Hz, 2H), 3.76 (s, 3H), 2.39 (s, 3H), 2.32 (s, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>): δ 173.8, 172.9, 159.0, 142.8, 132.0, 129.5, 129.5, 129.4, 129.0, 114.6, 55.4, 25.5, 21.6. MS (EI): m/z (%) = 283(12) [M]<sup>+</sup>, 241(43), 119(100), 91(55), 65(17), 43(19). C<sub>17</sub>H<sub>17</sub>NO<sub>3</sub> (283.32): calcd. C 72.07, H 6.05, N 4.94; found C 72.41, H 6.02, N 4.98

### 3.5. N-Acetyl-N-(4-bromophenyl)-4-methylbenzamide (3e):

The general procedure was followed by using 4-bromoacetanilide (0.5 mmol, 106 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3e** (129 mg, 78 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ 7.50 (d, J = 7.8 Hz, 2H), 7.44 (d, J = 8.2 Hz, 2H), 7.11 (d, J = 8.0 Hz, 2H), 7.01 (d, J = 8.4 Hz, 2H), 2.42 (s, 3H), 2.33 (s, 3H). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ 173.2, 172.4, 143.4, 138.3, 132.5, 131.9, 129.9, 129.7, 129.2, 127.1, 25.5, 21.6. MS (EI): m/z (%) = 334(3) [M+2], 332(3) [M]<sup>+</sup>, 291(18), 289(18), 120(15), 119(100), 91(52), 65(18), 43(19). C<sub>16</sub>H<sub>14</sub>BrNO<sub>2</sub> (332.19): calcd. C 57.85, H 4.25, N 4.22; found C 57.49, H 4.21, N 4.24

### 3.6. N-Acetyl-N-(4-chlorophenyl)-4-methylbenzamide (3f):

The general procedure was followed by using 4-chloroacetanilide (0.5 mmol, 85 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3f** (96 mg, 67 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ 7.50 (d, J = 8.2 Hz, 2H), 7.29 (d, J = 8.8 Hz, 2H), 7.12 (d, J = 8.9 Hz, 2H), 7.07 (d, J = 8.6 Hz, 2H), 2.42 (s, 3H), 2.33 (s, 3H). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ 173.3, 172.5, 143.4, 137.8, 133.8, 131.4, 129.5, 129.4, 129.2, 129.0, 25.5, 21.6. MS (EI): m/z (%) = 289(1) [M+2], 287(3) [M]<sup>+</sup>, 247(18), 245(53), 120(30), 119(100), 91(92), 65(46), 63(20), 43(32). C<sub>16</sub>H<sub>14</sub>ClNO<sub>2</sub> (287.74): calcd. C 66.79, H 4.90, N 4.87; found C 66.38, H 4.86, N 4.90

### 3.7. N-Acetyl-4-chloro-N-phenylbenzamide (3g):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 4-chlorobenzaldehyde (1.5 mmol, 210 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3g** (99 mg, 73 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>): δ 7.66 (d, J = 8.5 Hz, 2H), 7.44 (d, J = 8.5 Hz, 2H), 7.40 – 7.37 (m, 2H), 7.34–7.30 (m, 3H), 2.27 (s, 3H). <sup>13</sup>C NMR (125 MHz, DMSO-d<sub>6</sub>) δ 173.3, 172.0, 139.5, 136.9, 134.6, 131.0, 129.7, 129.4, 128.8, 128.6, 25.8. MS (EI): m/z (%) = 275(3) [M+2], 273(9) [M]<sup>+</sup>, 233(35), 231(86), 141(66), 139(100), 113(23), 111(67), 77(17), 75(37), 43(76). C<sub>15</sub>H<sub>12</sub>ClNO<sub>2</sub> (273.71): calcd. C 65.82, H 4.42, N 5.12; found C 65.59, H 4.37, N 5.09

### 3.8. N-Acetyl-4-chloro-N-(2-chlorophenyl)benzamide (3h):

The general procedure was followed by using 2-chloroacetanilide (0.5 mmol, 85 mg), 4-chlorobenzaldehyde (1.5 mmol, 210 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3h** (95 mg, 62 % yield) as a yellow oil. <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>): δ 8.26 (d, *J* = 8.1 Hz, 2H), 8.04-8.00 (m, 3H), 7.81 (d, *J* = 8.0 Hz, 1H), 7.64 (t, *J* = 8.1 Hz, 1H), 7.50 (t, *J* = 7.7 Hz, 1H), 2.43 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>): δ 173.2, 171.1, 141.9, 137.5, 137.3, 133.6, 131.9, 131.8, 131.2, 129.8, 128.7, 128.6, 25.8. MS (EI): *m/z* (%) = 312(1) [*M*+4], 310(5) [*M*+2], 308(9) [*M*]<sup>+</sup>, 269(13), 267(51), 265(85), 141(68), 139(100), 113(23), 111(73), 43(55). C<sub>15</sub>H<sub>11</sub>Cl<sub>2</sub>NO<sub>2</sub> (308.16): calcd. C 58.46, H 3.60, N 4.55; found C 58.72, H 3.63, N 4.59

### 3.9. N-Acetyl-N-(2-chloro-5-methylphenyl)-4-methylbenzamide (3i):

The general procedure was followed by using 2-chloro-5-methylacetanilide (0.5 mmol, 92 mg), 4-methylbenzaldehyde (1.5 mmol, 180 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3i** (117 mg, 78 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.50 (d, *J* = 7.9 Hz, 2H), 7.22-7.11 (m, 4H), 7.11 (s, 1H), 2.35 (s, 6H), 2.22 (s, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>): δ 173.7, 172.9, 142.8, 136.0, 132.3, 131.5, 130.3, 129.3, 129.0, 129.0, 128.8, 127.6, 25.7, 21.7, 18.2. MS (EI): *m/z* (%) = 303(12) [*M*+2], 301(40) [*M*]<sup>+</sup>, 119(100), 91(90), 89(35), 65(78), 63(25), 43(98). C<sub>17</sub>H<sub>16</sub>ClNO<sub>2</sub> (301.77): calcd. C 67.66, H 5.34, N 4.64; found C 67.93, H 5.37, N 4.67

### 3.10. N-Acetyl-3,4-dimethoxy-N-phenylbenzamide (3j):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 3,4-dimethoxybenzaldehyde (1.5 mmol, 250 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3j** (124 mg, 83 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>): δ 7.56 (d, *J* = 7.7 Hz, 3H), 7.39 (s, 1H), 7.28 (t, *J* = 7.4 Hz, 2H), 7.18 (d, *J* = 8.2 Hz, 1H), 7.01 (t, *J* = 7.4, 1H), 3.87 (s, 3H), 3.83 (s, 3H), 2.33 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>): δ 172.3, 171.1, 155.6, 150.6, 140.7, 131.0, 130.1, 127.5, 124.3, 120.4, 112.7, 110.8, 57.3, 56.9, 25.4. MS (EI): *m/z* (%) = 299(10) [*M*]<sup>+</sup>, 257(55), 165(100), 137(65), 65(22), 43(33). C<sub>17</sub>H<sub>17</sub>NO<sub>4</sub> (299.32): calcd. C 68.22, H 5.72, N 4.68; found C 68.36, H 5.68, N 4.71

### 3.11. N-Acetyl-N-(2-chlorophenyl)benzamide (3k):

The general procedure was followed by using 2-chloroacetanilide (0.5 mmol, 85 mg), benzaldehyde (1.5 mmol, 160 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3k** (100 mg, 73 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>): δ 7.96 (d, *J* = 8.7 Hz, 1H), 7.63 (d, *J* = 8.7 Hz, 2H), 7.51 – 7.46 (m, 2H), 7.40-7.36 (m, 4H), 2.31 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>): δ 173.0, 172.1, 137.2, 135.4, 133.3, 132.2, 130.8, 130.5, 129.7, 129.0, 128.7, 128.6, 25.7. MS (EI): *m/z* (%) = 275(2) [*M*+2], 273(5) [*M*]<sup>+</sup>,

233(29), 231(81), 196(23), 122(37), 105(100), 84(91), 77(95), 66(97), 43(56). C<sub>15</sub>H<sub>12</sub>ClNO<sub>2</sub> (273.71): calcd. C 65.82, H 4.42, N 5.12; found C 65.66, H 4.37, N 5.18

### 3.12. N-Acetyl-2-chloro-N-phenylbenzamide (3l):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 2-chlorobenzaldehyde (1.5 mmol, 210 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3l** (99 mg, 73 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>): δ 8.20 (d, J = 7.6 Hz, 1H), 8.03 (t, J = 7.1 Hz, 1H), 7.96 (d, J = 8.0 Hz, 1H), 7.90 – 7.85 (m, 3H), 7.60 (t, J = 7.8 Hz, 2H), 7.34 (t, J = 7.2 Hz, 1H), 2.36 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>) δ = 171.3, 170.1, 141.1, 138.1, 137.6, 133.9, 132.6, 131.5, 130.4, 129.7, 128.6, 127.8, 25.8. MS (EI): m/z (%) = 275(5) [M+2], 273(15) [M]<sup>+</sup>, 233(33), 231(82), 141(46), 139(100), 113(20), 111(58), 43(53). C<sub>15</sub>H<sub>12</sub>ClNO<sub>2</sub> (273.71): calcd. C 65.82, H 4.42, N 5.12; found C 66.44, H 4.39, N 5.14.

### 3.13. N-Acetyl-2-methoxy-N-phenylbenzamide (3m):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 2-methoxybenzaldehyde (1.5 mmol, 204 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3m** (105 mg, 78 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>): δ = 8.02-7.96 (m, 2H), 7.89 (d, J = 8.2 Hz, 2H), 7.59 (t, J = 7.4 Hz, 2H), 7.54 (d, J = 8.4 Hz, 1H), 7.39 (t, J = 7.4 Hz, 1H), 7.33 (t, J = 7.4 Hz, 1H), 4.23 (s, 3H), 2.35 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>): δ 171.5, 169.9, 163.1, 141.0, 138.1, 130.2, 129.3, 125.7, 124.6, 122.2, 120.6, 114.3, 57.5, 25.6. MS (EI): m/z (%) = 269(12) [M]<sup>+</sup>, 226(72), 135(100), 107(85), 65(51), 43(73). C<sub>16</sub>H<sub>15</sub>NO<sub>3</sub> (269.30): calcd. C 71.36, H 5.61, N 5.20; found C 71.58, H 5.57, N 5.23

### 3.14. N-Acetyl-3-chloro-N-phenylbenzamide (3n):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), 3-chlorobenzaldehyde (1.5 mmol, 210 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, *n*-hexane : EtOAc, 10:1) gave final product **3n** (98 mg, 72 % yield) as a colorless oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>) δ = 7.71 (s, 1H), 7.59 (d, J = 7.8 Hz, 1H), 7.50 (d, J = 7.9 Hz, 1H), 7.41 – 7.30 (m, 6H), 2.29 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>) δ = 173.3, 171.5, 139.4, 138.0, 133.4, 131.7, 130.5, 129.7, 129.5, 128.7, 128.6, 127.5, 25.9. MS (EI): m/z (%) = 275(4) [M+2], 273(13) [M]<sup>+</sup>, 233(69), 231(100), 141(88), 139(100), 113(34), 111(89), 86(76), 85(97), 84(89), 68(21), 66(71), 43(42). C<sub>15</sub>H<sub>12</sub>ClNO<sub>2</sub> (273.71): calcd. C 65.82, H 4.42, N 5.12; found C 65.39, H 4.45, N 5.17

### 3.15. N-Acetyl-N-phenylfuran-2-carboxamide (3o):

The general procedure was followed by using acetanilide (0.5 mmol, 68 mg), furfuraldehyde (1.5 mmol, 144 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL).

Purification by flash column chromatography (silica gel, n-hexane : EtOAc, 10:1) gave final product **3o** (86 mg, 75 % yield) as a dark brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>) δ 8.05 – 8.04 (m, 1H), 7.60 (dt, *J*=8.6, 1.4 Hz, 2H), 7.49 – 7.48 (m, 1H), 7.28 – 7.24 (m, 2H), 7.01 – 6.98 (m, 1H), 6.73 – 6.72 (m, 1H), 2.35 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>) δ 173.5, 171.8, 153.0, 149.5, 139.8, 129.0, 123.4, 123.2, 119.5, 113.2, 24.3. MS (EI): *m/z* (%) = 229(6) [M<sup>+</sup>], 187(70), 139(43), 95(100), 77(100), 67(33), 43(25). C<sub>13</sub>H<sub>11</sub>NO<sub>3</sub> (229.23): calcd. C 68.11, H 4.84, N 6.11; found C 68.41, H 4.81, N 6.17

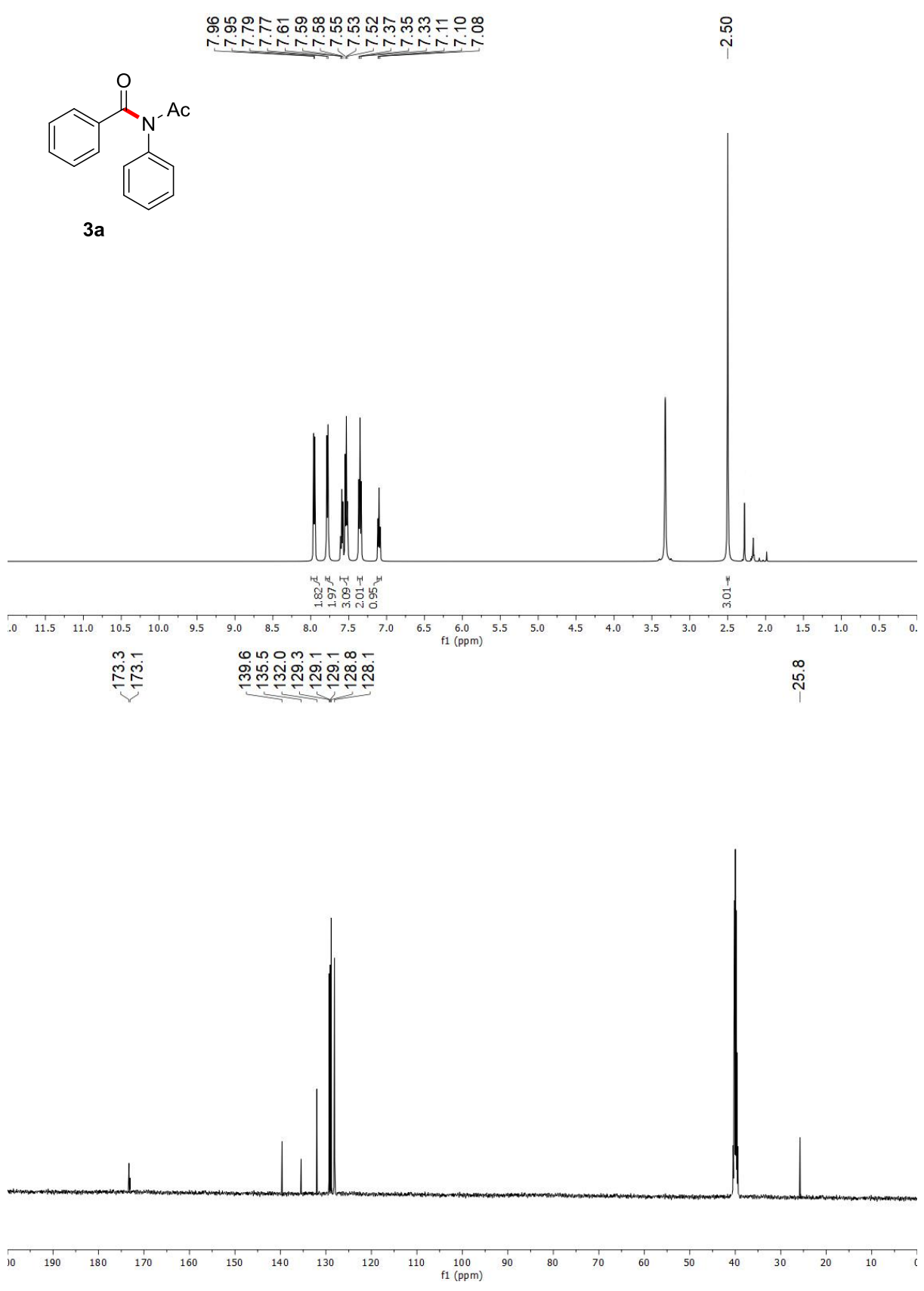
### 3.16. N-Acetyl-N-(p-tolyl)furan-2-carboxamide (3p):

The general procedure was followed by using 4-methylacetanilide (0.5 mmol, 75 mg), furfuraldehyde (1.5 mmol, 144 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, n-hexane : EtOAc, 10:1) gave final product **3p** (100 mg, 82 % yield) as a dark brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>) δ 8.06 (d, *J* = 1.8 Hz, 1H), 7.50 – 7.47 (m, 3H), 7.06 (d, *J* = 8.0 Hz, 2H), 6.74 – 6.73 (m, 1H), 2.41 (s, 3H), 2.23 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>) δ 173.5, 172.3, 153.0, 149.5, 137.3, 132.3, 129.4, 123.3, 119.5, 113.3, 24.3, 20.8. MS (EI): *m/z* (%) = 243(11) [M<sup>+</sup>], 153(20), 133(62), 95(82), 87(45), 45(100), 43(22). C<sub>14</sub>H<sub>13</sub>NO<sub>3</sub> (243.26): calcd. C 69.12, H 5.39, N 5.76; found C 69.53, H 5.34, N 5.84

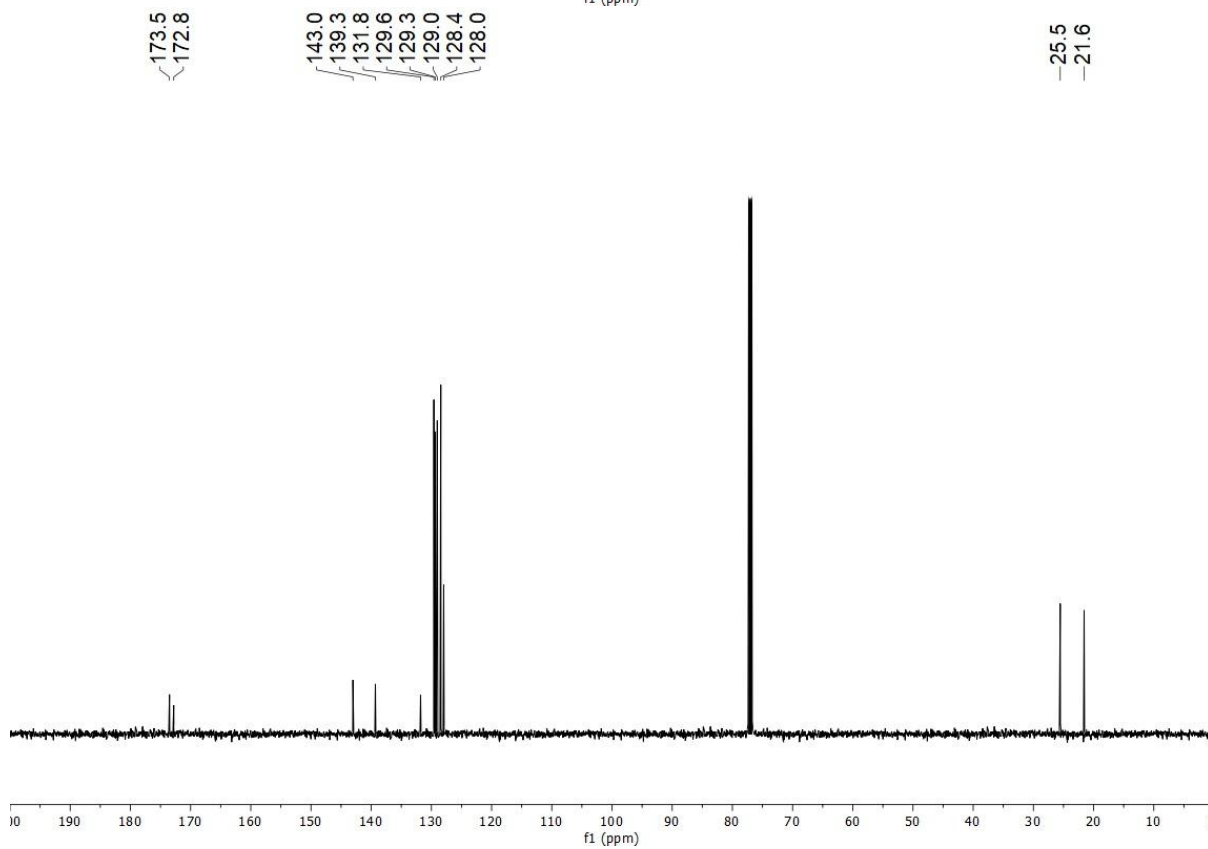
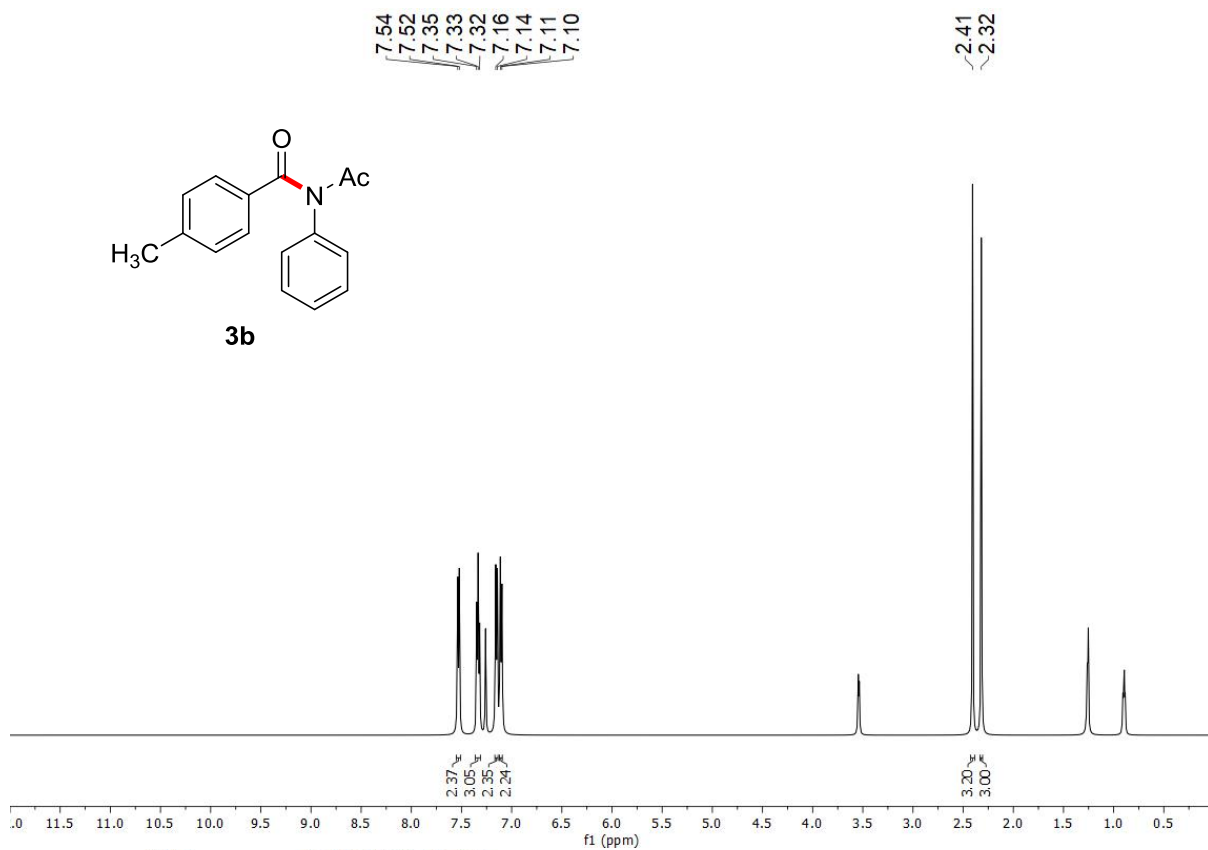
### 3.17. N-Acetyl-N-(p-tolyl)picolinamide (3q):

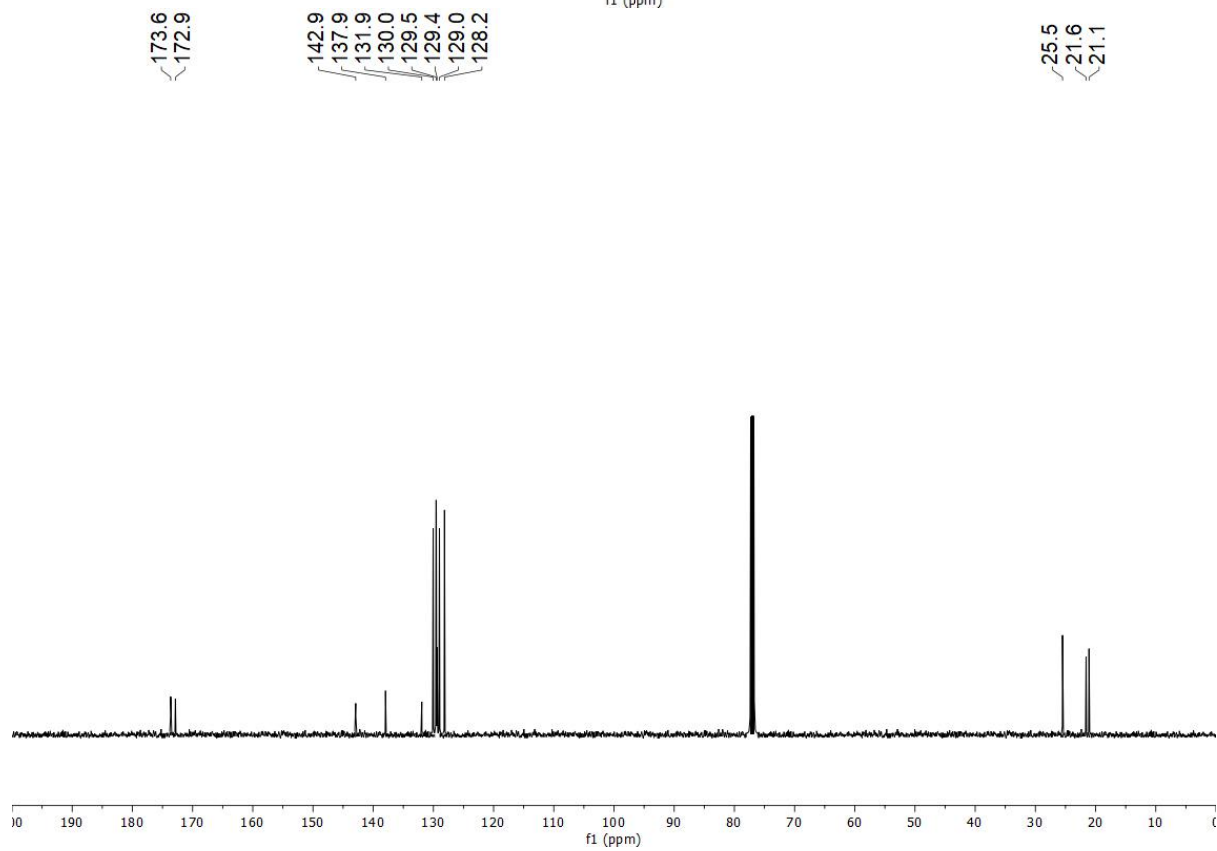
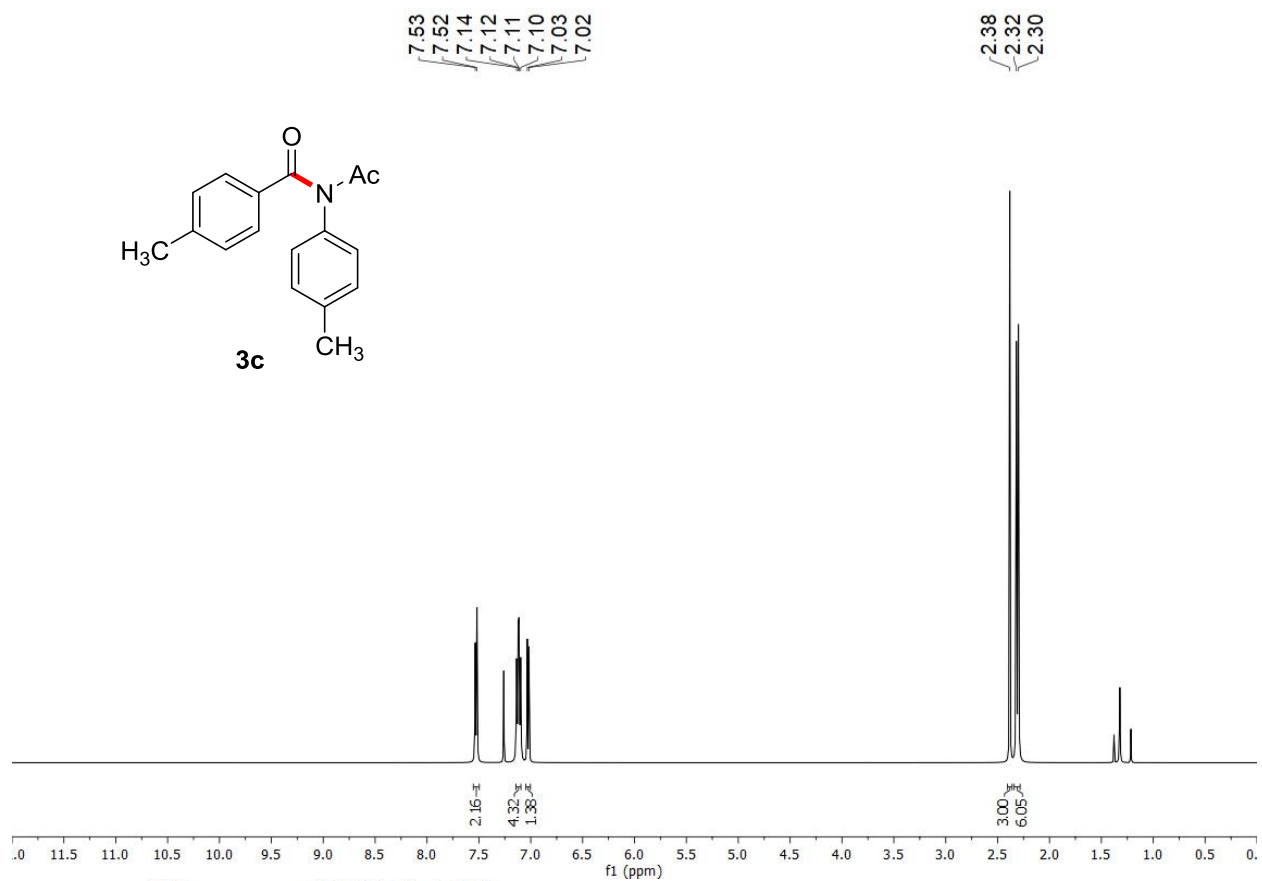
The general procedure was followed by using 4-methylacetanilide (0.5 mmol, 75 mg), picolinaldehyde (1.5 mmol, 160 mg), TBHP (1.5 mmol, 193 mg), KBr (1.0 equiv., 60 mg), and DCM (2.0 mL). Purification by flash column chromatography (silica gel, n-hexane : EtOAc, 10:1) gave final product **3q** (105 mg, 83 % yield) as a brown oil. <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>) δ 8.80 – 8.79 (m, 1H), 8.00 (td, *J* = 7.6, 1.5 Hz, 1H), 7.91 (dd, *J* = 7.8, 1.3 Hz, 1H), 7.67 – 7.64 (m, 1H), 7.48 (d, *J* = 8.2 Hz, 2H), 7.06 (d, *J* = 8.1 Hz, 2H), 2.41 (s, 3H), 2.23 (s, 3H). <sup>13</sup>C NMR (126 MHz, DMSO-d<sub>6</sub>) δ 171.3, 170.5, 152.8, 150.6, 138.0, 137.3, 132.2, 129.4, 128.8, 122.0, 119.5, 24.3, 20.8. MS (EI): *m/z* (%) = 254(5) [M<sup>+</sup>], 177(20), 164(23), 133(55), 107(100), 87(35), 45(44), 43(33). C<sub>15</sub>H<sub>14</sub>N<sub>2</sub>O<sub>2</sub> (254.28): calcd. C 70.85, H 5.55, N 11.02; found C 70.39, H 5.45, N 11.17

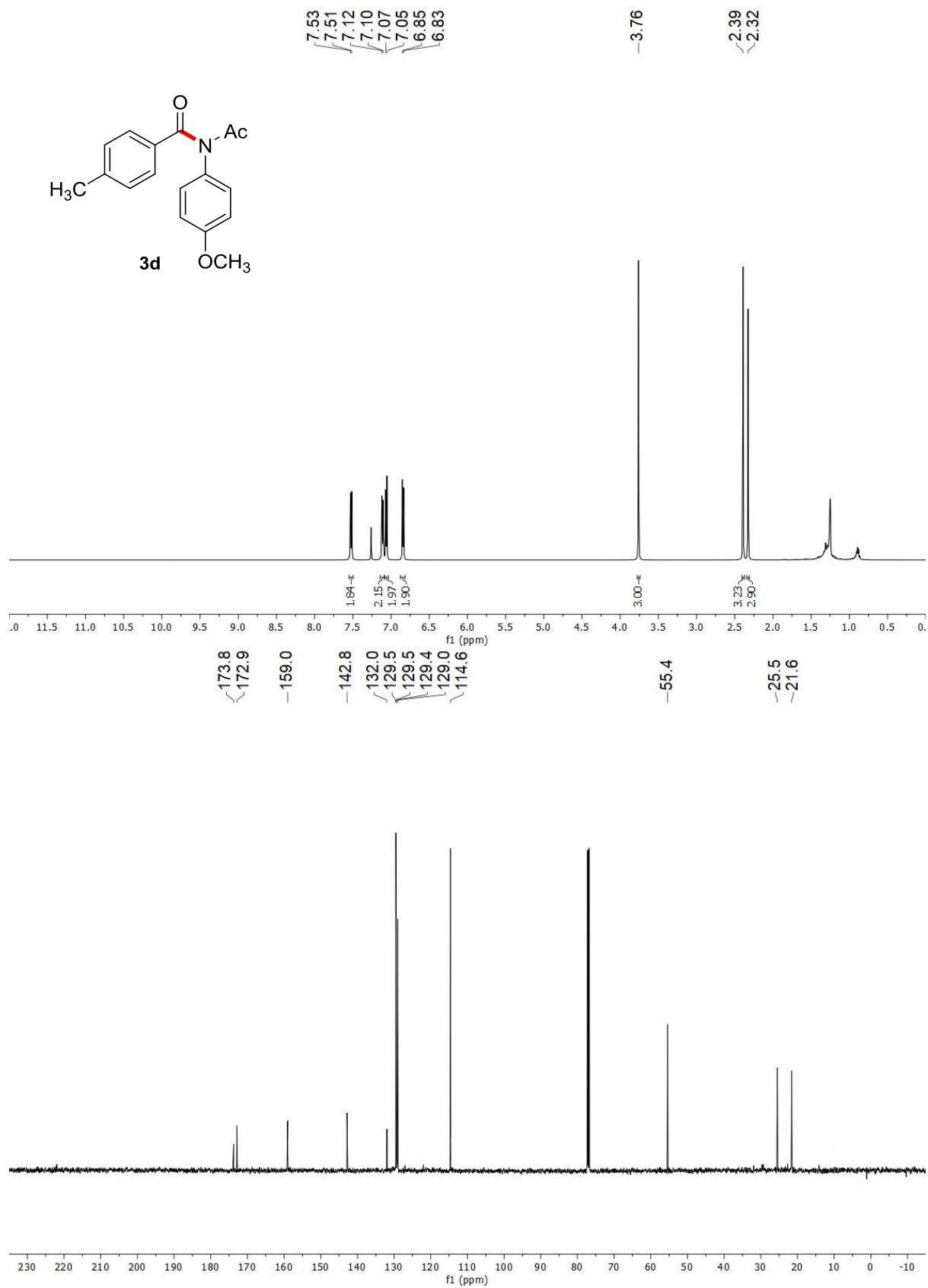
# 1. $^1\text{H}$ and $^{13}\text{C}$ NMR spectra

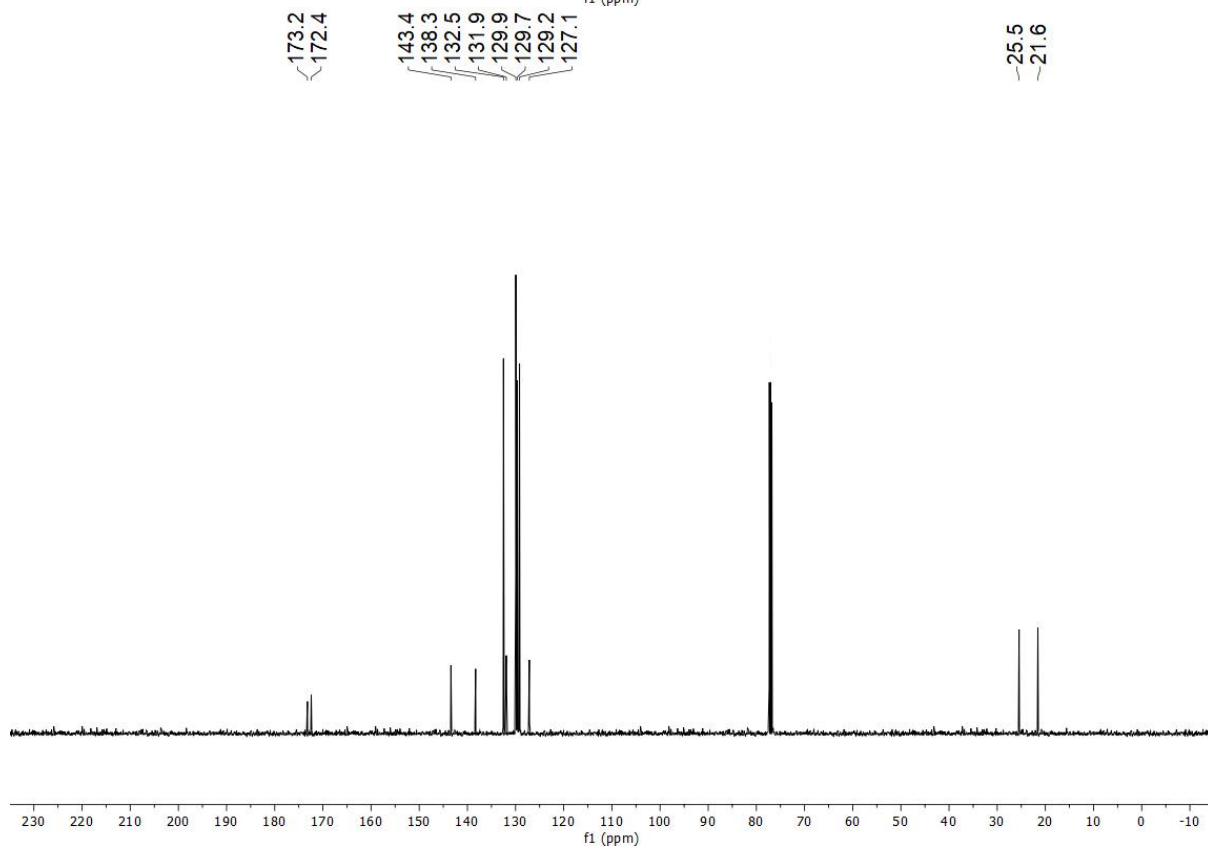
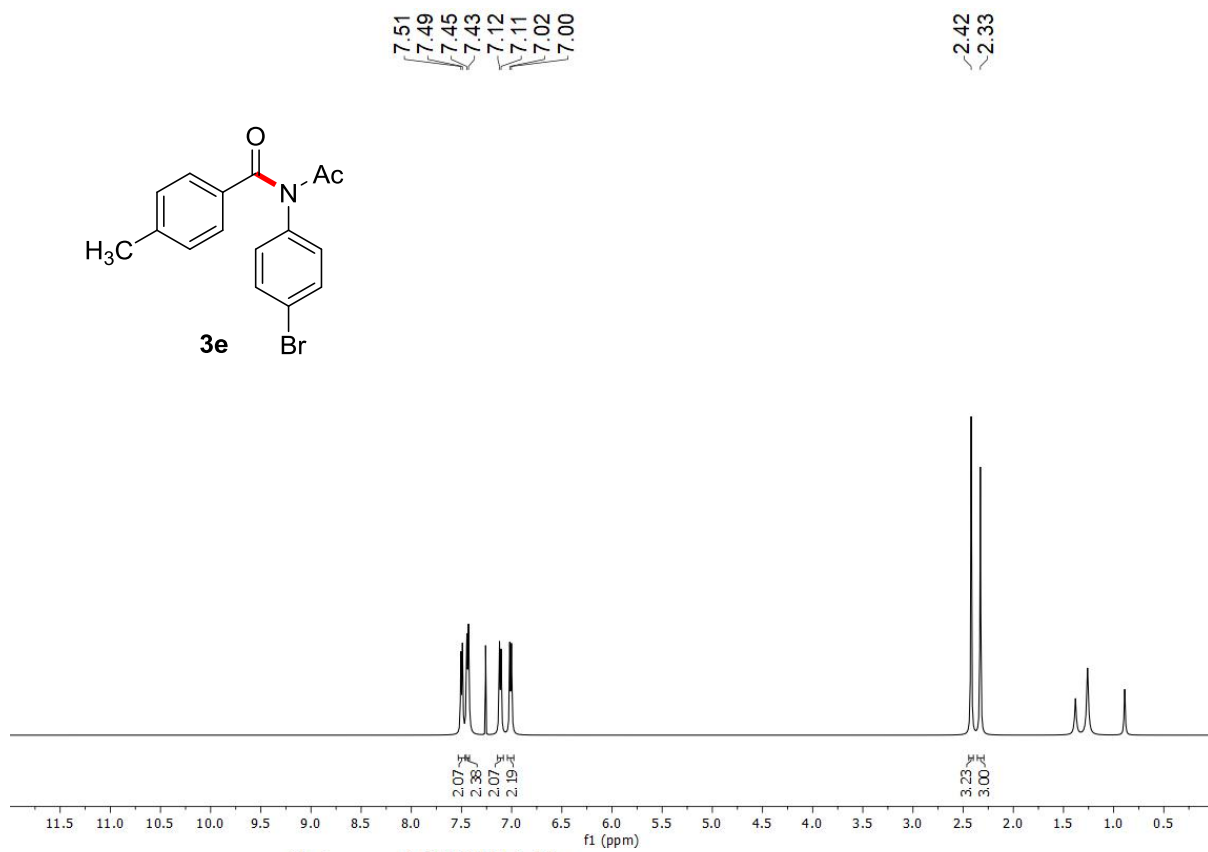
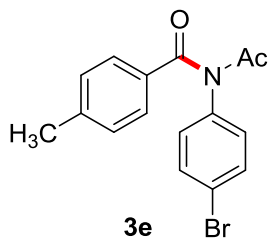


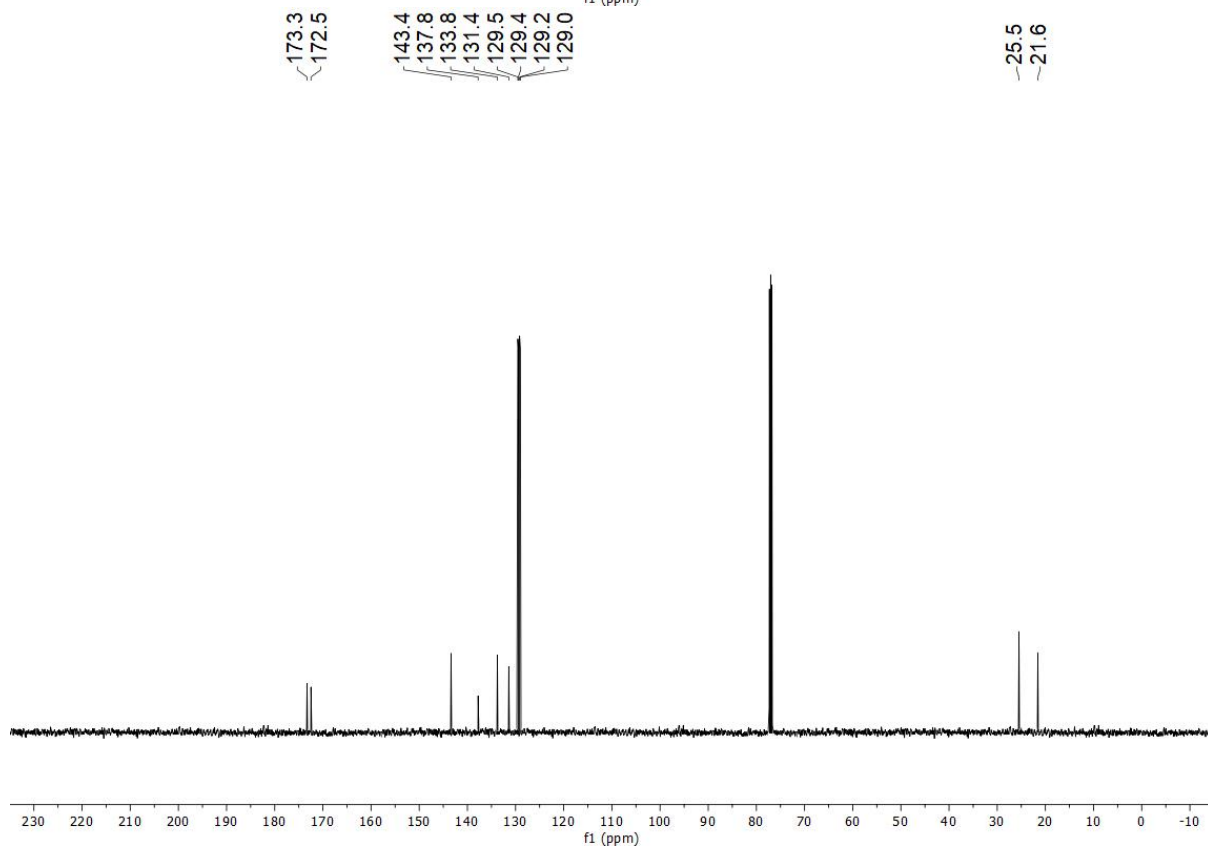
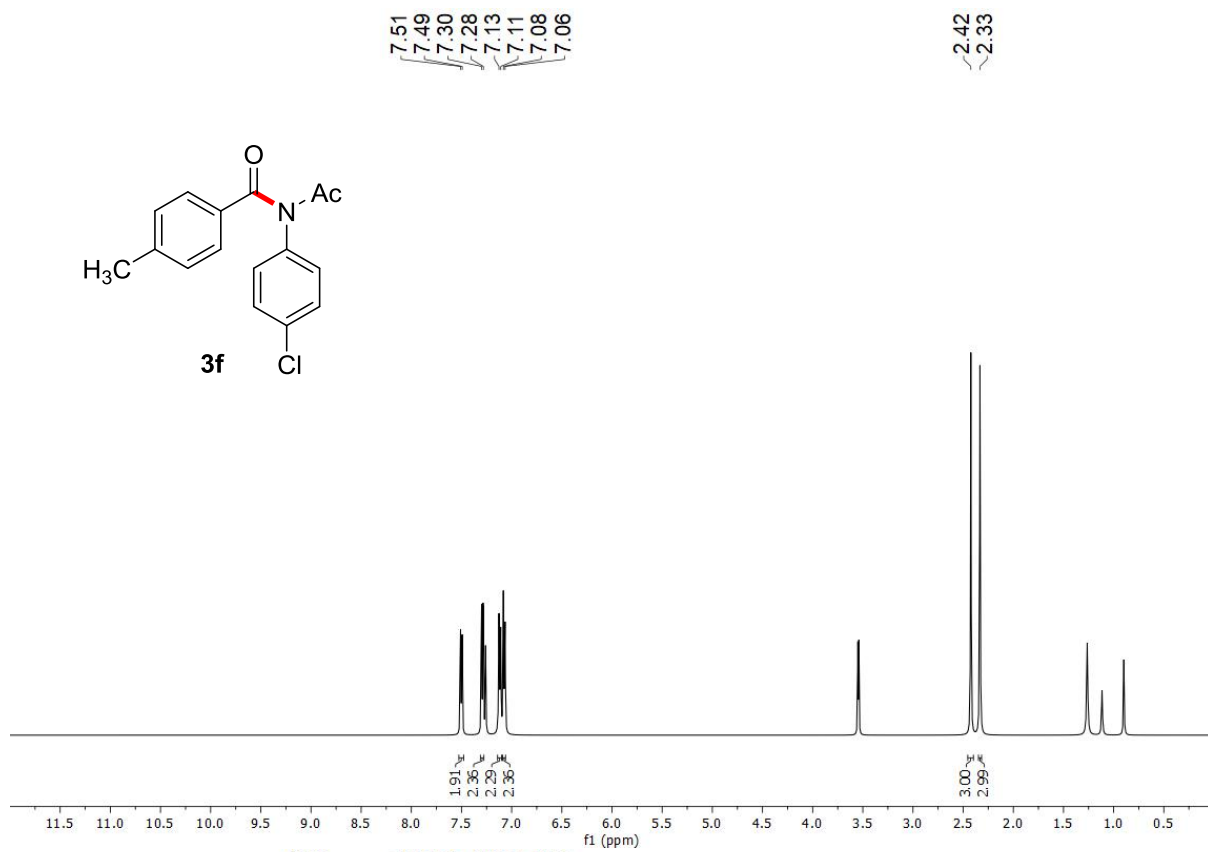


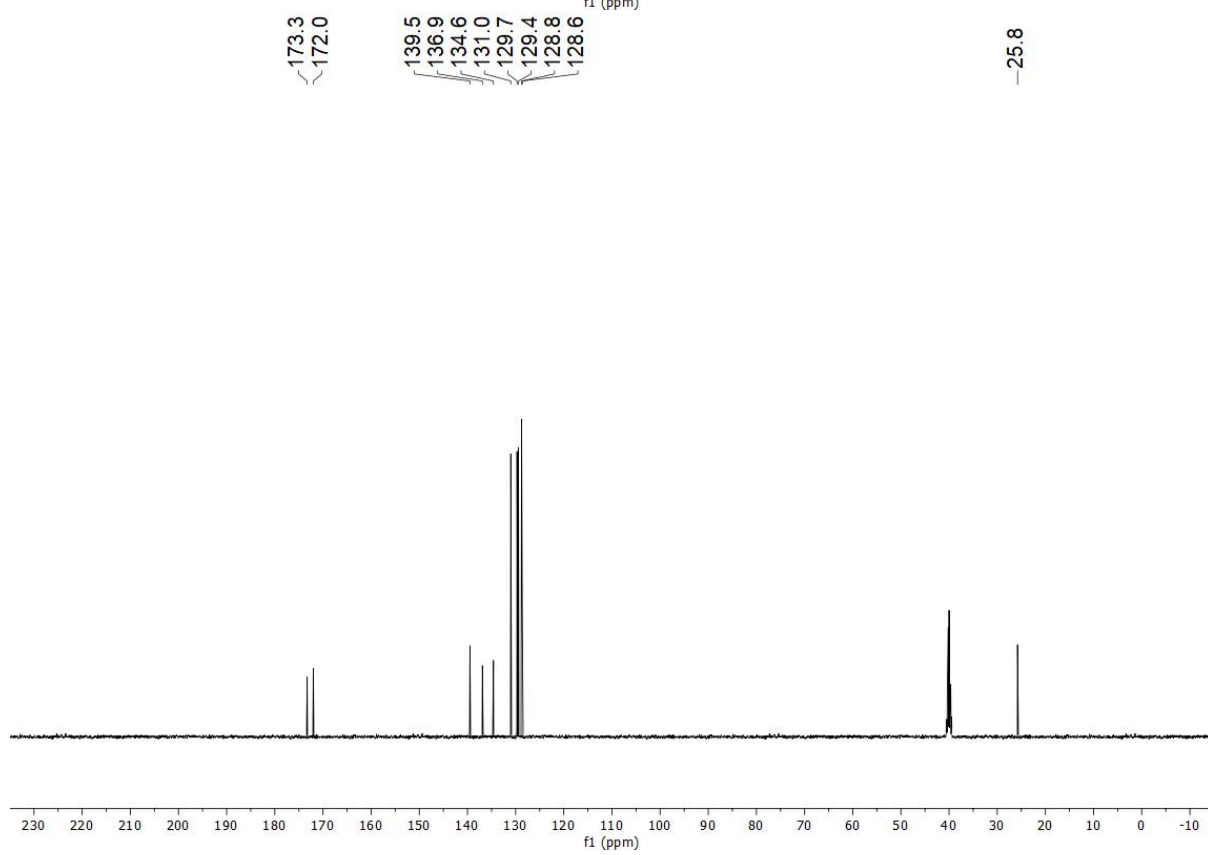
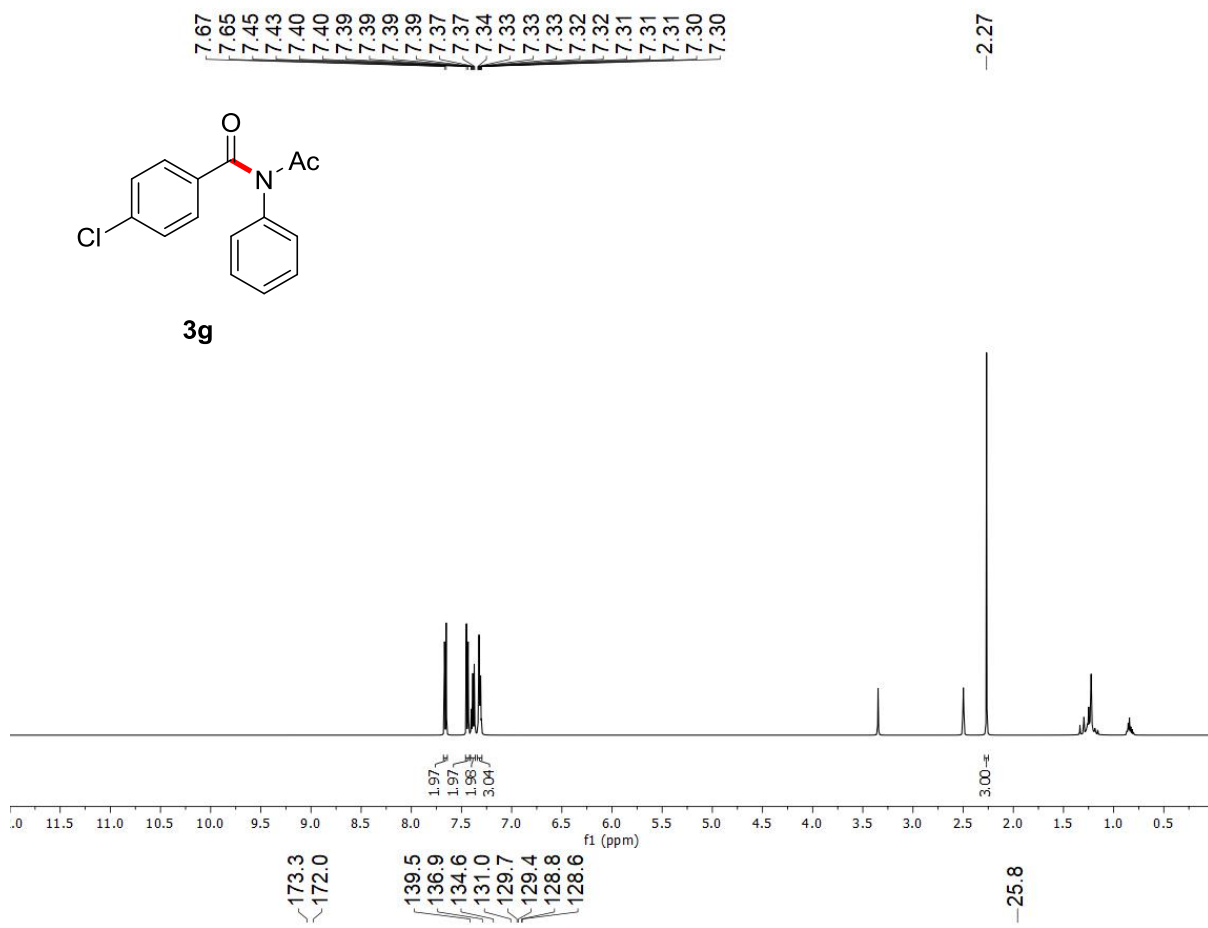


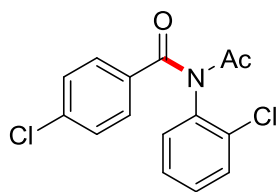












**3h**

