Supporting Information

Facile access to *N*-formyl imide and its new applications as a *N*-formylating agent for the direct synthesis of *N*-formamides, benzimidazoles and quinazolinones

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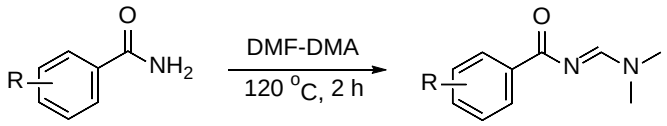
General Information:

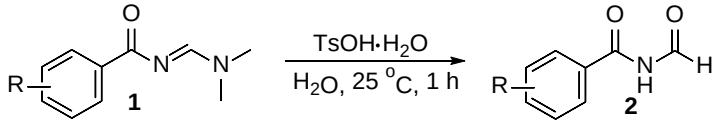
All reactions were performed under nitrogen atmosphere. ^{1}H NMR and ^{13}C NMR spectra were reported on a Varian 400 MHz NMR spectrometer with $CDCl_3$、CD_3OD、d_6-DMSO、d_6-Acetone as the solvent. Chemical shifts were reported in parts per million (ppm) relative to residual solvent peak ($CDCl_3$ δ H = 7.26 ppm, δ C = 77.00 ppm; CD_3OD δ H = 3.31 ppm, δ C = 49.15 ppm; d_6-DMSO δ H = 2.49 ppm, δ C = 39.15 ppm; d_6-Acetone δ H = 2.05 ppm, δ C = 29.92 ppm). The IR spectra were measured on a Thermo Scientific Nicolet iS5 FT-IR spectrophotometer. TLC was performed on pre-coated glass plates of Silica Gel 60 F254 (0.25 mm, E. Merck); detection was performed by spraying with using a UV light, a solution of basic aqueous potassium permanganate ($KMnO_4$) and a solution of phosphomolybdic acid with heating. Flash column chromatography was carried out on Silica Gel 60 (230–400 mesh, E. Merck). High resolution mass spectrometry data were recorded on an EI spectrometer. Melting points were measured by Electrothermo (UK) " Mel- Temp 1101D" type melting point apparatus and are uncorrected. The following abbreviations were used to indicate multiplicities: s = singlet, d = doublet, t = triplet, q = quartet, quin. = quintet, dd = doublet of doublets, dt = doublet of triplets, td = triplet of doublets, br = broad, m = multiplet.

General Procedure I by Literature:[1]



To a solution of the organic amide (1.00 equiv) in *N*,*N*-dimethylformamide dimethyl acetal (DMF-DMA) (3.00 equiv) were stirred at 120 °C under nitrogen atmosphere for two hours. After that, the resulting reaction mixture was cooled to room temperature and the reaction was completed as indicated by TLC. After the reaction was completed, the mixture was removed the excess reagents under reduced pressure to a high vacuum pump to afford the desired amidine products **1a** – **1j.**

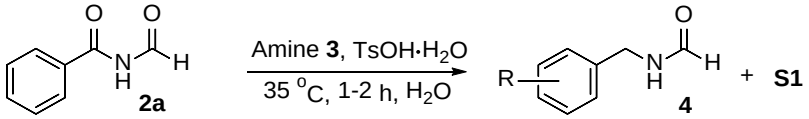
General Procedure II:



To a solution of the amidine (1.00 equiv) in H_2O (0.1 M) was added *p*-toluenesulfonic acid monohydrate (1.00 equiv) stirred at 25 °C under nitrogen atmosphere. Then the resulting reaction mixture was stirred for one hour. After the reaction was completed, the mixture was extracted with ethyl acetate for three times and the combined organic

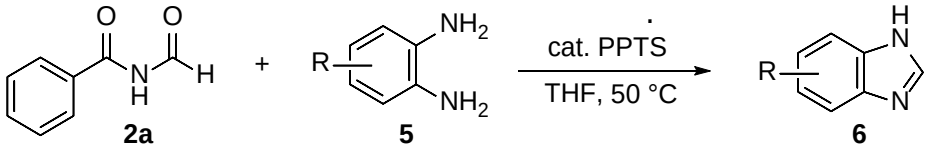
phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography to afford the desired products **2a** – **2j.**

General Procedure III:



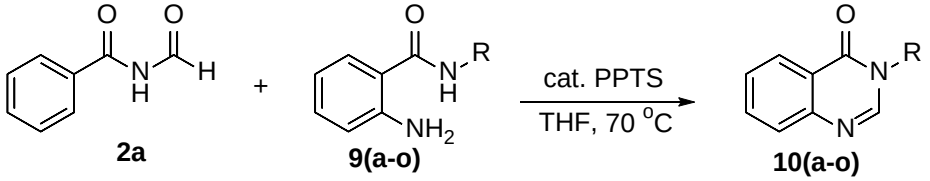
To a solution of the *N*-formyl imide **2a** (1.00 equiv) in H_2O (0.1 M) was added *p*-toluenesulfonic acid monohydrate (0.20 equiv) and amine **3** (2.00-5.00 equiv). Then the resulting reaction mixture was stirred at 35 °C for 1-2 hours under nitrogen atmosphere. After the reaction was completed, the mixture was extracted with ethyl acetate for three times and the combined organic phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography to afford the desired products **4a – 4r**.

General Procedure IV: For benzimidazole synthesis



To a solution of the *N*-formyl imide **2a** (1.00 equiv) in THF (0.3 M) was added pyridinium *p*-toluenesulfonate (0.20 equiv) and diamine **5** (1 equiv). Then the resulting reaction mixture was stirred at 50 °C for 7-24 hours under nitrogen atmosphere. After the reaction was completed, the mixture was removed the solvent under reduced pressure to a high vacuum pump and extracted with ethyl acetate for three times. The combined organic phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography to afford the desired products **6a – 6p**.

General Procedure V: For quinazolinone synthesis



To a solution of the *N*-formyl imide **2a** (1.00 equiv) in THF (0.3 M) was added pyridinium *p*-toluenesulfonate (0.20 equiv) and 2-aminobenzamide **9** (1 equiv). Then the resulting reaction mixture was stirred at 70 °C for 8-24 hours under nitrogen

atmosphere. After the reaction was completed, the mixture was removed the solvent under reduced pressure to a high vacuum pump and extracted with ethyl acetate for three times. The combined organic phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography to afford the desired products **10a – 10o**.

(*E*)-*N*-((Dimethylamino)methylene)benzamide (1a)[2]: Following General Procedure I, using the benzamide (1000 mg, 8.25 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (1967 mg, 16.5 mmol). Compound **1a** was obtained (1309 mg, 90% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.63 (s, 1H), 8.25 (d, *J* = 8.0 Hz, 2H), 7.47 (t, *J* = 7.8 Hz, 1H), 7.39 (t, *J* = 7.7 Hz, 2H), 3.20 (s, 3H), 3.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.6, 160.6, 136.6, 131.7, 129.6, 127.8, 41.2, 35.1.

(*E*)-*N*-((Dimethylamino)methylene)-4-methoxybenzamide (1b)[3]: Following General Procedure I, using the 4-methoxybenzamide (82.0 mg, 0.54 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (130.0 mg, 1.08 mmol). Compound **1b** was obtained (106.0 mg, 95% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.59 (s, 1H), 8.23 (d, *J* = 8.7 Hz, 2H), 6.89 (d, *J* = 8.7 Hz, 2H), 3.83 (s, 3H), 3.17 (s, 3H), 3.14 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.2, 162.6, 160.4, 131.7, 129.3, 113.0, 55.3, 41.3, 35.1.

(*E*)-*N*-((Dimethylamino)methylene)-3-methoxybenzamide (1c): Following General Procedure I, using the 3-methoxybenzamide (50.0 mg, 0.33 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (79.0 mg, 0.66 mmol). Compound **1c** was obtained (63.0 mg, 93% yield) as a white solid, m.p. 123-125 °C. IR (KBr, cm^{-1}): 3355, 3161, 2966, 2843, 1664, 1583, 1399, 1250, 1054, 1032, 686. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.63 (s, 1H), 7.90 (d, *J* = 7.6 Hz, 1H), 7.81 (s, 1H), 7.33 (t, *J* = 7.9 Hz, 1H), 7.04 (d, *J* = 8.1 Hz, 1H), 3.86 (s, 3H), 3.21 (s, 3H), 3.19 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 177.6, 160.8, 159.4, 138.2, 128.9, 122.4, 118.4, 113.9, 55.4, 41.4, 35.3. HRMS (EI) m/z: $[M]^+$ calcd for $C_{11}H_{14}N_2O_2$ 206.1055; found 206.1049.

(*E*)-*N*-((Dimethylamino)methylene)-2-methylbenzamide (1d): Following General Procedure I, using the 2-methylbenzamide (113.0 mg, 0.84 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (199.0 mg, 1.67 mmol). Compound **1d** was obtained (131.0 mg, 82% yield) as a white solid, m.p. 118-120 °C. IR (KBr, cm^{-1}): 3293, 3276, 2980, 2950, 2864, 2843, 1663, 1620, 1454, 1398, 1056, 1031, 693. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.58 (s, 1H), 8.06 (d, *J* = 7.8 Hz, 1H), 7.31 (t, *J* = 8.1 Hz, 1H), 7.21 (t, *J* = 7.9 Hz, 2H), 3.18 (s, 3H), 3.17 (s, 3H), 2.62 (s, 3H); ^{13}C NMR (100

MHz, $CDCl_3$): δ 180.3, 160.3, 138.9, 136.7, 131.2, 130.5, 130.4, 125.2, 41.3, 35.3, 21.7. HRMS (EI) m/z: $[M]^+$ calcd for $C_{11}H_{14}N_2O$ 190.1106; found 190.1105.

(*E*)-*N*-((Dimethylamino)methylene)-4-nitrobenzamide (1e)[4]: Following General Procedure I, using the 4-nitrobenzamide (90.0 mg, 0.54 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (129.0 mg, 1.08 mmol). Compound **1e** was obtained (117.0 mg, 98% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.66 (s, 1H), 8.39 (d, *J* = 8.9 Hz, 2H), 8.23 (d, *J* = 8.9 Hz, 2H), 3.24 (s, 3H), 3.23 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 175.5, 161.2, 149.7, 142.3, 130.6, 123.0, 41.6, 35.5.

(*E*)-2-Chloro-*N*-((dimethylamino)methylene)benzamide (1f)[33]: Following General Procedure III, using the 2-chlorolbenzamide (100.0 mg, 0.64 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (153.0 mg, 1.29 mmol). Compound **1f** was obtained (80.0 mg, 60% yield) as a yellow liquid. IR (KBr, cm^{-1}): 3358, 3180, 2938, 2832, 1667, 1596, 1480, 1432, 1391, 1100, 1048, 1032, 739. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.59 (s, 1H), 7.88 (d, *J* = 7.4 Hz, 1H), 7.39 (d, *J* = 7.1 Hz, 1H), 7.33-7.25 (m, 2H), 3.19 (s, 3H), 3.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 178.4, 161.0, 137.5, 132.7, 131.2, 131.1, 130.8, 126.5, 41.8, 35.7. Quadrupole-Mass Filter,[33] m/z: $[M+H]^+$ calcd for $C_{10}H_{11}ClN_2O$ 211.1; found 211.2.

(*E*)-*N*-((Dimethylamino)methylene)thiophene-2-carboxamide (1g)[2]: Following General Procedure I, using the thiophene-2-carboxamide (75.0 mg, 0.59 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (141.0 mg, 1.18 mmol). Compound **1g** was obtained (106.0 mg, 99% yield) as a yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.61 (s, 1H), 7.88 (d, *J* = 3.7 Hz, 1H), 7.48 (d, *J* = 4.9 Hz, 1H), 7.09 (t, *J* = 4.3 Hz, 1H), 3.19 (s, 3H), 3.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 172.4, 160.4, 142.9, 131.7, 131.5, 127.7, 41.3, 35.3.

***N*-((*E*)-(Dimethylamino)methylene)cinnamamide (1h)[2]:** Following General Procedure I, using the cinnamamide (200.0 mg, 1.35 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (324.0 mg, 2.71 mmol). Compound **1h** was obtained (264.0 mg, 96% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.58 (s, 1H), 7.82 (d, *J* = 15.8 Hz, 1H), 7.56 (d, *J* = 8.0 Hz, 2H), 7.39-7.26 (m, 3H), 6.73 (d, *J* = 15.8 Hz, 1H), 3.18 (s, 3H), 3.17 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 178.2, 160.5, 142.5, 135.4, 129.5, 128.6, 128.0, 126.3, 41.4, 35.2.

(*E*)-*N*-((Dimethylamino)methylene)-4-pyridinecarboxamide (1i)[2]: Following General Procedure I, using the pyridine-4-carboxylic acid amide (150.0 mg, 1.23

mmol) in *N*,*N*-dimethylformamide dimethyl acetal (439.0 mg, 3.68 mmol). Compound **1i** was obtained (212.0 mg, 97% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.71 (dd, *J* = 4.4, 1.6 Hz, 2H), 8.66 (s, 1H), 8.03 (dd, *J* = 4.4, 1.6 Hz, 2H), 3.22 (dd, *J* = 5.7, 0.5 Hz, 2H); ^{13}C NMR (100 MHz, CD_3OD): δ 176.9, 162.9, 150.6, 146.3, 124.7, 41.9, 35.9.

(*E*)-*N*-((dimethylamino)methylene)pivalamide (1j)[2]: Following General Procedure I, using the pivalamide (200.0 mg, 1.97 mmol) in *N*,*N*-dimethylformamide dimethyl acetal (707.0 mg, 5.93 mmol). Compound **1j** was obtained (285.0 mg, 92% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.33 (s, 1H), 3.07 (d, *J* = 9.8 Hz, 6H), 1.19 (s, 9H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 159.7, 41.1, 34.9, 27.8.

***N*-Formylbenzamide (2a)**[2]**:** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)benzamide **1a** (1044 mg, 5.93 mmol) and *p*-toluenesulfonic acid (1128 mg, 5.93 mmol). The product was then purified by column chromatography (hexane/EtOAc, 1:1), compound **2a** was obtained (834.0 mg, 94% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.97 (br, 1H), 9.41 (d, *J* = 9.7 Hz, 1H), 7.98 (d, *J* = 8.4 Hz, 2H), 7.66 (t, *J* = 7.4 Hz, 1H), 7.55 (t, *J* = 7.4 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 178.2, 160.5, 142.5, 135.4, 129.5, 128.6, 128.0, 126.3, 41.4, 35.2.

***N*-Formyl-4-methoxybenzamide (2b)**[2,5]**:** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)-4-methoxybenzamide **1b** (52.0 mg, 0.25 mmol) and *p*-toluenesulfonic acid (48.0 mg, 0.25 mmol). The product was then purified by column chromatography (hexane/EtOAc, 1:1), compound **2b** was obtained (41.0 mg, 91% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.36 (d, *J* = 9.8 Hz, 1H), 9.09 (br, 1H), 7.88 (d, *J* = 8.8 Hz, 2H), 7.01 (d, *J* = 8.8 Hz, 2H), 3.90 (s, 3H). ^{13}C NMR (100 MHz, *d*$_6$-DMSO): δ 166.6, 164.6, 163.4, 130.7, 123.5, 114.0, 55.6.

***N*-Formyl-3-methoxybenzamide (2c):** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)-3-methoxybenzamide **1c** (35.0 mg, 0.17 mmol) and *p*-toluenesulfonic acid (32.0 mg, 0.17 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2c** was obtained (30.0 mg, 99% yield) as a white solid, m.p. 107-109 ^{o}C. IR (KBr, cm^{-1}): 3706, 3010, 2965, 2941, 2863, 2843, 1727, 1679, 1478, 1276, 1055, 1032, 1016, 802, 743. ^{1}H NMR (400 MHz, $CDCl_3$): δ 10.19 (br, 1H), 9.39 (d, *J* = 9.6 Hz, 1H), 7.55 (d, *J* = 9.2 Hz, 2H), 7.43 (t, *J* = 7.9 Hz, 1H), 7.18 (d, *J* = 8.2 Hz, 1H), 3.88 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.5, 164.8, 160.0, 132.2, 130.0, 120.5, 120.1, 112.6, 55.5. HRMS (EI) m/z: $[M]^+$

calcd for $C_9H_9NO_3$ 179.0582; found 179.0580.

***N*-Formyl-2-methylbenzamide (2d):** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)-2-methylbenzamide **1e** (56.0 mg, 0.29 mmol) and *p*-toluenesulfonic acid (56.0 mg, 0.29 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2e** was obtained (44.0 mg, 92% yield) as a white solid, m.p. 103-105 ºC. IR (KBr, cm^{-1}): 3705, 3401, 3238, 2972, 2939, 2922, 2843, 1728, 1690, 1499, 1470, 1374, 1257, 1055, 1032, 732. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.24 (d, *J* = 9.6 Hz, 1H), 9.19 (br, 1H), 7.51 (d, *J* = 7.4 Hz, 1H), 7.45 (t, *J* = 7.5 Hz, 1H), 7.30 (t, *J* = 7.4 Hz, 2H), 2.53 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 168.6, 163.6, 138.5, 133.2, 131.9, 131.8, 127.3, 126.1, 20.4. HRMS (EI) m/z: $[M]^+$ calcd for $C_9H_9NO_2$ 163.0633; found 163.0632.

***N*-Formyl-4-nitrobenzamide (2e)[2]:** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)-4-nitrobenzamide **1e** (54.0 mg, 0.24 mmol) and *p*-toluenesulfonic acid (46.0 mg, 0.24 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2e** was obtained (36.0 mg, 77% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.38 (d, *J* = 9.5 Hz, 1H), 9.31 (br, 1H), 8.40 (d, *J* = 8.7 Hz, 2H), 8.11 (d, *J* = 8.7 Hz, 2H); ^{13}C NMR (100 MHz, d_6-DMSO): δ 166.7, 164.7, 150.5, 137.5, 130.4, 124.1.

2-Chloro-*N*-formylbenzamide (2f): Following General Procedure II, using the (*E*)-2-chloro-*N*-((dimethylamino)methylene)benzamide **1f** (80.0 mg, 0.38 mmol) and *p*-toluenesulfonic acid (72.0 mg, 0.38 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2f** was obtained (62.0 mg, 88% yield) as a white solid, m.p. 79-80 ºC. IR (KBr, cm^{-1}): 3679, 3293, 2980, 2972, 2941, 2865, 2843, 1735, 1689, 1487, 1370, 1189, 1052, 1017, 749. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.24 (br, 1H), 8.98 (br, 1H), 7.76 (d, *J* = 7.2 Hz, 1H), 7.53-7.47 (m, 2H), 7.44-7.40 (m, 1H). ^{13}C NMR (100 MHz, $CDCl_3$): δ 166.1, 162.3, 133.4, 131.6, 131.3, 130.9, 130.8, 127.6. HRMS (EI) m/z: $[M]^+$ calcd for $C_8H_6ClNO_2$ 183.0087; found 183.0081.

***N*-Formylthiophene-2-carboxamide (2g)[2]:** Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)thiophene-2-carboxamide **1g** (56.0 mg, 0.31 mmol) and *p*-toluenesulfonic acid (58.0 mg, 0.31 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2g** was obtained (47.0 mg, 99% yield) as a yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.68 (br, 1H), 9.34 (br, 1H), 7.90 (d, *J* = 3.8 Hz, 1H), 7.74 (d, *J* = 4.9 Hz, 1H), 7.21 (t, *J* =

4.4 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ164.5, 160.9, 136.0, 134.7, 131.8, 128.6.

***N*-Formylcinnamamide (2h)[2]:** Following General Procedure II, using the *N*-((*E*)-(dimethylamino)methylene)cinnamamide **1h** (55.0 mg, 0.27 mmol) and *p*-toluenesulfonic acid (52.0 mg, 0.27 mmol). The product was then purified by column chromatography (hexane/EtOAc, 4:1), Compound **2h** was obtained (42.0 mg, 88% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.27 (d, *J* = 10.0 Hz, 1H), 8.56 (br, 1H), 7.87 (d, *J* = 15.7 Hz, 1H), 7.57 (d, *J* = 7.8 Hz, 2H), 7.45-7.40 (m, 3H), 6.47 (d, *J* = 15.7 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 165.5, 164.5, 146.5, 133.6, 131.2, 129.0, 128.5, 117.9.

***N*-formylisonicotinamide** (**2i**)[2]: Following General Procedure II, using the (*E*)-*N*-((Dimethylamino)methylene)-4-pyridinecarboxamide **1i** (50.0 mg, 0.28 mmol) and *p*-toluenesulfonic acid (53.0 mg, 0.28 mmol). The product was then purified by column chromatography (hexane/EtOAc, 1:1), Compound **2i** was obtained (24.0 mg, 56% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.87 (br, 1H), 9.40 (d, *J* = 9.5 Hz, 1H), 8.89 ((d, *J* = 4.8 Hz, 2H), 7.80 (d, *J* = 4.8 Hz, 2H); ^{13}C NMR (100 MHz, CD_3OD): δ 167.9, 164.9, 151.3, 150.9, 141.5, 123.3, 123.1.

***N*-formylpivalamide** (**2j**)[2]: Following General Procedure II, using the (*E*)-*N*-((dimethylamino)methylene)pivalamide **1j** (50.0 mg, 0.32 mmol) and *p*-toluenesulfonic acid (60.0 mg, 0.32 mmol). The product was then purified by column chromatography (hexane/EtOAc, 1:1), Compound **2j** was obtained (37.0 mg, 90% yield) as a white solid. ^{1}H NMR (400 MHz, CD_3OD): δ 9.12 (s, 1H), 1.22 (s, 9H); ^{13}C NMR (100 MHz, CD_3OD): δ 181.4, 165.4, 40.5, 26.6.

***N*-Benzylformamide (4a)[6]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (54.0 mg, 0.36 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.072 mmol) and benzylamine (78.0 mg, 0.72 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4a** was obtained (43.0 mg, 86% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.02/0.21): δ 8.28 (s, 1H) (major), 8.21 (d, *J* = 11.9 Hz, 1H) (minor), 7.37-7.27 (m, 5H), 5.80 (br, 1H), 4.50 (d, *J* = 5.9 Hz, 2H) (major), 4.44 (d, *J* = 6.6 Hz, 2H) (minor); ^{13}C NMR (100 MHz, $CDCl_3$): δ 164.7, 161.1, 137.5, 137.4, 128.8, 128.7, 127.9, 127.7, 127.7, 126.9, 45.6, 42.0.

***N*-Benzyl-*N*-methylformamide (4b)[6]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (54.0 mg, 0.36 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.072

mmol) and *N*-methylbenzylamine (87.0 mg, 0.72 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4b** was obtained (46.0 mg, 85% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.00/0.73): δ 8.29 (s, 1H) (major), 8.16 (s, 1H) (minor), 7.39-7.20 (m, 5H), 4.52 (s, 2H) (minor), 4.40 (s, 2H) (major), 2.85 (s, 3H) (minor), 2.78 (s, 3H) (major); ^{13}C NMR (100 MHz, $CDCl_3$): δ 162.7, 162.5, 135.9, 135.6, 128.8, 128.6, 128.1, 128.0, 127.5, 127.3, 53.4, 47.6, 34.0, 29.4.

***N*-(Furan-2-ylmethyl)formamide (4c)[7]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (57.0 mg, 0.38 mmol), *p*-toluenesulfonic acid (15.0 mg, 0.076 mmol) and furan-2-ylmethanamine (74.0 mg, 0.76 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4c** was obtained (38.0 mg, 79% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 2.00/0.38): δ 8.22 (s, 1H) (major), 8.18 (d, *J* = 11.9 Hz, 1H) (minor), 7.384-7.381 (m, 1H) (minor), 7.364-7.362 (m, 1H) (major), 6.33-6.32 (m, 1H), 6.26-6.23 (m, 1H), 5.87 (br, 1H), 4.48 (d, *J* = 5.7 Hz, 2H) (major), 4.37 (d, *J* = 6.4 Hz, 2H) (minor); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.1, 150.6, 142.1, 110.4, 107.5, 34.9.

***N*-Phenylformamide (4d)[8]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (61.0 mg, 0.41 mmol), *p*-toluenesulfonic acid (16.0 mg, 0.082 mmol) and aniline (77.0 mg, 0.82 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4d** was obtained (31.0 mg, 62% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.09/1.00): δ 8.69 (d, *J* = 11.4 Hz, 1H) (minor), 8.38 (s, 1H) (major), 8.20 (br, 1H), 7.54 (d, *J* = 7.6 Hz, 1H), 7.38-7.31 (m, 2H), 7.21-7.08 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 163.0, 159.5, 136.9, 136.7, 129.6, 128.9, 125.2, 124.7, 120.0, 118.7.

***N*-Dodecylformamide (4e)[9]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (51.0 mg, 0.34 mmol), *p*-toluenesulfonic acid (13.0 mg, 0.067 mmol) and dodecylamine (125.0 mg, 0.67 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4e** was obtained (67.0 mg, 93% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.00/0.40): δ 8.16 (s, 1H) (major), 8.03 (d, *J* = 12.1 Hz, 1H) (minor), 5.45 (br, 1H), 3.29 (q, *J* = 6.8 Hz, 2H) (major), 3.21 (q, *J* = 6.7 Hz, 2H) (minor), 1.54-1.49 (m, 2H), 1.35-1.21 (m, 18H), 0.88 (t, *J* = 6.6 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 164.6, 161.2, 41.8, 38.1, 31.8, 31.1, 29.5, 29.49, 29.45, 29.41, 29.3, 29.2, 29.1, 26.8, 26.3, 22.6, 14.0.

***N*-*tert*-Butylformamide (4f)**[10]**:** To a solution of the *N*-formylbenzamide **2a** (63.0 mg, 0.42 mmol) and *tert*-butylamine (154.0 mg, 2.11 mmol). was stirred at 35 ºC under nitrogen atmosphere. Then the resulting reaction mixture was stirred for 4 hours. After the reaction was completed, the mixture was extracted with ethyl acetate for three times and the combined organic phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography (DCM/Acetone, 10:1), Compound **4f** was obtained (26.0 mg, 60% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.00/1.00): δ 8.24 (d, *J* = 12.1 Hz, 1H), 8.00 (s, 1H), 6.27 (br, 1H), 5.49 (br, 1H), 1.35 (s, 9H), 1.31 (s, 9H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 163.0, 160.5, 51.3, 50.3, 30.8, 28.8.

***N*-(Prop-2-yn-1-yl)formamide (4g)**[8]**:** Following General Procedure III, using the *N*-formylbenzamide **2a** (59.0 mg, 0.39 mmol), *p*-toluenesulfonic acid (15.0 mg, 0.078 mmol) and prop-2-yn-1-amine (107.0 mg, 1.95 mmol). The product was then purified by column chromatography (DCM/Acetone, 10:1), Compound **4g** was obtained (24.0 mg, 74% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.00/0.12): δ 8.18 (s, 1H) (major), 8.13 (d, *J* = 12.0 Hz, 1H) (minor), 5.81 (br, 1H), 4.10 (q, *J* = 2.4 Hz, 2H) (major), 4.02 (q, *J* = 2.4 Hz, 2H) (minor), 2.35 (t, *J* = 2.2 Hz, 1H) (minor), 2.26 (t, *J* = 2.3 Hz, 1H) (major); ^{13}C NMR (100 MHz, $CDCl_3$): δ160.9, 78.8, 71.7, 27.7.

***N*-Cyclohexylformamide (4h)**[9]**:** Following General Procedure III, using the *N*-formylbenzamide **2a** (54.0 mg, 0.36 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.072 mmol) and cyclohexylamine (179.0 mg, 1.80 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4h** was obtained (30.0 mg, 64% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 1.00/0.43): δ 8.12 (s, 1H) (minor), 8.08 (s, 1H) (major), 5.79 (br, 1H) (minor), 5.57 (br, 1H) (major), 3.89-3.79 (m, 1H) (major), 3.34-3.25 (m, 1H) (minor), 1.93-1.86 (m, 2H), 1.73-1.68 (m, 2H), 1.62-1.58 (m, 1H), 1.40-1.14 (m, 5H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 163.5, 160.3, 50.9, 47.0, 34.6, 32.9, 25.3, 24.9, 24.6.

***N,N*-Diallylformamide (4i)**[11]**:** Following General Procedure III, using the *N*-formylbenzamide **2a** (55.0 mg, 0.37 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.073 mmol) and diallylamine (71.0 mg, 0.74 mmol). The product was then purified by column chromatography (DCM/Acetone, 10:1), Compound **4i** was obtained (20.0 mg, 44% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.12 (s, 1H), 5.78-5.65 (m, 2H),

5.25-5.13 (m, 4H), 3.93 (d, *J* = 5.9 Hz, 2H), 3.81 (d, *J* = 5.8 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ162.5, 133.0, 132.0, 118.6, 118.1, 49.2, 44.3.

***N*-isobutylformamide (4j)[6]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), *p*-toluenesulfonic acid (13.0 mg, 0.068 mmol) and isobutylamine (49.0 mg, 0.68 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **4j** was obtained (25.0 mg, 86% yield) as a liquid. ^{1}H NMR (400 MHz, d_6-Acetone; Mixture of rotamers is observed. Ratio: .1/0.34): δ 8.17 (s, 1H) (major), 7.98 (d, *J* = 11.9 Hz, 1H) (minor), 5.86 (br, 1H), 3.11 (t, *J* = 6.5 Hz, 2H) (major), 3.01 (t, *J* = 6.6 Hz, 2H) (minor), 1.83-1.67 (m, 1H), 0.91 (d, *J* = 6.7 Hz, 9H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 165.1, 161.5, 49.4, 45.5, 29.7, 28.5, 20.1, 19.7.

Morpholine-4-carbaldehyde (4k)[9]: Following General Procedure III, using the *N*-formylbenzamide **2a** (50.0 mg, 0.33 mmol), *p*-toluenesulfonic acid (13.0 mg, 0.066 mmol) and morpholine (145.0 mg, 1.66 mmol). The product was then purified by column chromatography (DCM/EtOAc, 3:1), Compound **4k** was obtained (25.0 mg, 65% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.05 (s, 1H), 3.70-3.68 (m, 2H), 3.67-3.64 (m, 2H), 3.58-3.56 (m, 2H), 3.40-3.38 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.8, 67.2, 66.4, 45.8, 40.6.

***N,N*-(1,3-Phenylenebis(methylene))diformamide (4l)[12]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (136.0 mg, 0.91 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.073 mmol) and 1,3-phenylenedimethanamine (50.0 mg, 0.37 mmol). The product was then purified by column chromatography (DCM/Acetone, 1:1), Compound **4l** was obtained (30.0 mg, 43% yield) as a white solid. ^{1}H NMR (400 MHz, d_6-Acetone; Mixture of rotamers is observed. Ratio: 2.00/0.37): δ 8.24 (s, 2H) (major), 8.19 (d, *J* = 11.8 Hz, 2H) (minor), 7.64 (br, 2H), 7.33-7.19 (m, 4H), 4.42 (d, *J* = 6.6 Hz, 4H) (minor), 4.41 (d, *J* = 6.2 Hz, 4H) (major); ^{13}C NMR (100 MHz, d_6-Acetone): δ 161.6, 140.4, 129.4, 127.5, 127.1, 41.9. HRMS (EI) m/z: $[M]^+$ calcd for $C_{10}H_{12}N_2O_2$ 192.0899; found 192.0900.

***N*-(2-Hydroxyethyl)formamide (4m)[6,13]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (56.0 mg, 0.38 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.075 mmol) and ethanolamine (114.0 mg, 1.87 mmol). The product was then purified by column chromatography (DCM/Acetone, 6:1), Compound **4m** was obtained (34.0 mg, 99% yield) as a liquid. ^{1}H NMR (400 MHz, d_6-DMSO; Mixture of rotamers is observed. Ratio: 2.00/1.29): δ 8.00 (br, 1H), 7.98 (s, 1H) (major), 7.88 (d, *J* = 11.8 Hz,

1H) (minor), 7.84 (br, 1H), 4.71 (t, *J* = 5.4 Hz, 1H) (major), 4.65 (t, *J* = 5.4 Hz, 1H) (minor), 3.38 (q, *J* = 5.8 Hz, 2H) (major), 3.37 (q, *J* = 6.1 Hz, 2H) (minor), 3.12 (q, *J* = 5.9 Hz, 2H) (major), 3.06 (q, *J* = 6.0 Hz, 2H) (minor); ^{13}C NMR (100 MHz, *d*$_6$-DMSO): δ 164.4, 61.8, 61.7, 43.3, 41.8.

***N*-(2-Aminobenzyl)formamide (4n):** Following General Procedure III, using the *N*-formylbenzamide **2a** (55.0 mg, 0.37 mmol), *p*-toluenesulfonic acid (14.0 mg, 0.074 mmol) and 2-aminobenzylamine (90.0 mg, 0.74 mmol). The product was then purified by column chromatography (DCM/Acetone, 2:1), Compound **4n** was obtained (37.0 mg, 66% yield) as a liquid. IR (KBr, cm^{-1}): 3710, 3382, 2949, 2921, 2864, 2843, 1664, 1454, 1345, 1048, 1032, 1014, 670. ^{1}H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 2.15/0.20): δ 8.10 (s, 1H), 7.10 (t, *J* = 7.6 Hz, 1H), 7.02 (d, J = 7.5 Hz, 1H), 6.71-6.64 (m, 2H), 6.29 (br, 1H), 4.35 (d, *J* = 6.3 Hz, 2H) (major), 4.24 (d, *J* = 5.9 Hz, 2H) (minor), 3.97 (br, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ161.5, 145.2, 130.5, 129.3, 121.2, 117.9, 115.8, 39.1. HRMS (EI) m/z: $[M]^+$ calcd for $C_8H_{10}N_2O$ 150.0793; found 150.0797.

Indoline-1-carbaldehyde (4p)[6]: Following General Procedure III, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), *p*-toluenesulfonic acid (13.0 mg, 0.067 mmol) and indoline (80.0 mg, 0.67 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **4p** was obtained (36.0 mg, 89% yield) as a brown solid. ^{1}H NMR (400 MHz, CDCl3; Mixture of rotamers is observed. Ratio: 0.84/0.22): δ 8.89 (s, 1H) (major), 8.48 (s, 1H) (minor), 8.04 (d, *J* = 7.5 Hz, 1H) (minor), 7.22 - 7.11 (m, 3H), 7.04 – 7.00 (m, 1H), 4.09 – 4.00 (m, 2H), 3.17 - 3.09 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 159.4, 157.7, 141.1, 132.0, 127.64, 127.63, 126.1, 124.9, 124.6, 124.3, 116.7, 109.5, 47.0, 44.7, 27.8, 27.3.

***N*-(2-(1H-indol-3-yl)ethyl)formamide (4q)[6]:** Following General Procedure III, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), *p*-toluenesulfonic acid (13.0 mg, 0.067 mmol) and tryptamine (107.0 mg, 0.67 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **4q** was obtained (29.0 mg, 70% yield) as a liquid. ^{1}H NMR (300 MHz, CDCl3; Mixture of rotamers is observed. Ratio: 0.73/0.27): δ 8.49 (br , 1H), 8.03 (s, 1H) (major), 7.82 (d, *J* = 12.0 Hz, 1H) (minor), 7.58 (d, *J* = 8 Hz, 1H) (major), 7.54(d, *J* = 8 Hz, 1H) (minor), 7.36 (d, *J* = 8.8 Hz, 1H), 7.21 (t, *J* = 8 Hz, 1H), 7.10-7.14 (m, 1H), 6.98 (d, *J* = 2 Hz, 1H), 6.94 (d, *J* = 2.2 Hz, 1H), 5.82 (br , 1H), 3.61 (q, *J* = 6.4 Hz, 2 H) (major), 3.46 (q, *J* = 6.4 Hz, 2H) (minor), 2.97 (t, *J* = 6.8 Hz, 2H) (major), 2.92 (t, *J* = 6.8 Hz, 2H) (minor); ^{13}C NMR (100 MHz, $CDCl_3$): δ 164.9, 161.6, 136.6, 136.5, 127.3, 126.9, 122.9, 122.4,

122.2, 122.1, 119.5, 119.4, 118.6, 118.4, 112.3, 111.6, 111.5, 111.3, 42.2, 38.4, 27.3, 25.2.

Ethyl formylglycinate (4r)[14]**:** To a solution of the e *N*-formylbenzamide **2a** (53.0 mg, 0.35 mmol) in H_2O (0.1 M) was added glycine *N*-ethyl ester hydrochloride (247.0 mg, 1.77 mmol), $NaHCO_3$ (148.0 mg, 1.77 mmol) and stirred at 35 ^oC under nitrogen atmosphere. Then the resulting reaction mixture was stirred for 1.5 hours. After the reaction was completed, the mixture was extracted with ethyl acetate for three times and the combined organic phase was washed with brine, dried over $MgSO_4$, filtered and concentrated. The crude products were purified by flash column chromatography (Hexane/EtOAc, 1:1), Compound **4r** was obtained (30.0 mg, 65% yield) as a liquid. ^1H NMR (400 MHz, $CDCl_3$; Mixture of rotamers is observed. Ratio: 2.00/0.12): δ 8.24 (s, 1H), 6.29 (br, 1H), 4.22 (q, *J* = 7.2 Hz, 2H), 4.07 (d, *J* = 5.3 Hz, 2H) (major), .3.98 (d, *J* = 6.2 Hz, 2H) (minor), 1.28 (t, *J* = 7.2 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 169.5, 161.0, 61.7, 39.9, 14.1.

1*H*-Benzoimidazole (6a)[15]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and *o*-Phenylenediamine (36.0 mg, 0.34 mmol). The product was then purified by column chromatography (EtOAc), Compound **6a** was obtained (36.0 mg, 90% yield) as a brown solid. ^1H NMR (400 MHz, $CDCl_3$): δ 8.10 (s, 1H), 7.68 (dd, *J* = 6.1 Hz, 3.2 Hz, 2H), 7.31 (dd, *J* = 6.1 Hz, 3.2 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 140.6, 137.5, 123.2, 115.6.

6-methyl-1*H*-benzimidazole (6b)[16]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-Methyl-*o*-phenylenediamine (41.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6b** was obtained (27.0 mg, 61% yield) as a brown solid. ^1H NMR (400 MHz, $CDCl_3$) : δ 10.10 (br, 1H), 8.12 (s, 1H), 7.58 (d, *J* = 8.2 Hz, 1H), 7.46(s, 1H), 7.12 (d, *J* = 8.2 Hz, 1H), 2.47 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 140.7, 137.6, 136.5, 132.8, 124.5, 115.5, 114.9, 21.7.

7-methyl-1*H*-benzimidazole (6c)[15]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 3-Methyl-*o*-phenylenediamine (41.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6c** was obtained (40.0 mg, 91% yield) as a pale yellow solid. ^1H NMR

(400 MHz, $CDCl_3$): δ 10.00 (br, 1H), 8.16 (s, 1H), 7.52 (d, *J* = 8.0 Hz, 1H), 7.21 (t, *J* = 7.7 Hz, 1H), 7.11 (d, *J* = 7.2Hz, 1H), 2.64 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 140.7, 137.8, 137.4, 125.9, 123.3, 122.9, 112.8, 17.4.

6-methoxy-1*H*-benzimidazole (6d)[17,18]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-Methoxy-*o*-phenylenediamine (46.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:5), Compound **6d** was obtained (45.0 mg, 91% yield) as a liquid. ^1H NMR (400 MHz, $CDCl_3$) : δ 8.04 (s, 1H), 7.55 (d, *J* = 8.8 Hz, 1H), 7.09 (d, *J* = 2.1 Hz, 1H), 6.93 (dd, *J* = 8.8, 2.3 Hz, 1H), 3.82(s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ156.7, 140.6, 137.7, 133.0, 116.5, 112.7, 97.6, 55.9.

6-fluoro-1*H*-benzimidazole (6e)[19]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-fluoro-*o*-phenylenediamine (42.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6e** was obtained (43.0 mg, 94% yield) as a brown solid. ^1H NMR (400 MHz, $CDCl_3$) : δ 8.14 (s, 1H), 7.59 (dd, *J* = 8.8, 4.7 Hz, 1H), 7.33 (dd, *J* = 8.8, 2.4 Hz, 1H), 7.06 (td, (dd, *J* = 8.8, 2.4 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.0, 158.7, 141.9, 137.9, 137.7, 134.6, 116.4, 116.3, 111.7, 111.5, 101.6, 101.3.; ^{19}F NMR (376 MHz, $CDCl_3$): δ -120.9 (m, C-*F*).

6-chloro-1*H*-benzimidazole (6f)[15]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-chloro-*o*-phenylenediamine (48.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6f** was obtained (44.0 mg, 86% yield) as a brown solid. ^1H NMR (400 MHz, $CDCl_3$) : δ 8.12(s, 1H), 7.65 (d, *J* = 1.7 Hz, 1H), 7.58 (d, *J* = 8.6 Hz, 1H), 7.27 (dd, *J* = 8.6, 1.7 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 141.6, 138.5, 136.5, 128.8, 123.8, 116.5, 115.5.

6-bromo-1*H*-benzimidazole (6g)[20]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-bromo-*o*-phenylenediamine (63.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6g** was obtained (60.0 mg, 92% yield) as a pale yellow solid. ^1H NMR (400 MHz, $CDCl_3$) : δ 10.36 (br, 1H), 8.18 (s, 1H), 7.82 (s, 1H), 7.53 (d, *J* = 8.6 Hz,

1H), 7.40 (d, *J* = 8.6 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 141.9, 139.1, 136.7, 126.3, 118.5, 116.8, 116.1.

6-nitro-1*H*-benzimidazole (6h)[15,18]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (1000 mg, 6.70 mmol), pyridinium *p*-toluenesulfonate (337.0 mg, 1.34 mmol) and 4-nitro-*o*-phenylenediamine (1026 mg, 6.70 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6h** was obtained (1022 mg, 93% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, CD_3OD) : δ 8.54 (s, 1H), 8.44 (s, 1H), 8.20 (d, *J* = 8.9 Hz, 1H), 7.74 (d, *J* = 8.9 Hz, 1H); ^{13}C NMR (100 MHz, CD_3OD): δ 147.1, 145.1, 119.3.

1*H*-benzimidazole-6-carbonitrile (6i)[20]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 3,4-Diaminobenzonitrile (45.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:2), Compound **6i** was obtained (45.0 mg, 94% yield) as a brown solid. ^{1}H NMR (400 MHz, CD_3OD) : δ 8.39 (s, 1H), 8.02 (s, 1H), 7.74 (d, *J* = 8.4 Hz, 1H), 7.56 (dd, *J* = 8.4, 1.4 Hz, 1H); ^{13}C NMR (100 MHz, CD_3OD): δ 145.9, 127.1, 120.6, 106.7.

5-Methylester-1*H*-benzoimidazole (6j)[18,21]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and Methyl 3,4-diaminobenzoate (56.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:2), Compound **6j** was obtained (56.0 mg, 96% yield) as a brown solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.42 (s, 1H), 8.19 (s, 1H), 8.03 (dd, *J* = 8.4, 1.1 Hz, 1H), 7.68(d, *J* = 8.4 Hz, 1H), 3.95 (s, 3H); ^{13}C NMR (100 MHz, CD_3OD): δ 168.9, 145.1, 125.7, 125.0, 118.9, 115.6, 52.6.

5,6-dichloro-1*H*-benzimidazole (6k)[18]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4,5-Dichloro-*o*-phenylenediamine (59.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:2), Compound **6k** was obtained (39.0 mg, 62% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, CD_3OD) : δ 8.20 (s, 1H), 7.67 (s, 1H); ^{13}C NMR (100 MHz, CD_3OD): δ 144.9, 138.4, 127.5, 117.4.

5,6-dimethyl-1*H*-benzimidazole (6l)[22]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0

mg, 0.068 mmol) and 4,5-Dimethyl-1,2-phenylenediamine (46.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **6l** was obtained (47.0 mg, 96% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 7.97 (s, 1H), 7.43 (s, 2H), 2.37 (s, 6H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 139.9, 136.3, 132.1, 115.7, 20.5.

1*H*-Naphth[2,3-*d*]imidazole (6m)[15,23]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 2,3-Diaminonaphthalene (53.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:2), Compound **6m** was obtained (38.0 mg, 67% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, CD_3OD) : δ 8.37 (s, 1H), 8.06 (s, 2H), 7.93 (dd, *J* = 6.4, 3.2 Hz, 2H), 7.36 (dd, *J* = 6.4, 3.2 Hz, 2H); ^{13}C NMR (100 MHz, CD_3OD): δ 146.9, 131.9, 128.9, 124.9.

1-methyl-1*H*-benzoimidazole (6n)[19]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (227.0 mg, 1.52 mmol), pyridinium *p*-toluenesulfonate (76.0 mg, 0.30 mmol) and *N*-Methyl-1,2-phenylenediamine (186.0 mg, 1.52 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **6n** was obtained (161.0 mg, 81% yield) as a liquid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 7.85 (s, 1H), 7.81 (d, *J* = 8.0 Hz, 1H), 7.39 (d, *J* = 8.0 Hz, 1H), 7.34-7.26 (m, 2H), 3.83 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 143.5, 134.4, 122.8, 121.9, 120.0, 109.3, 30.9.

1-Benzyl-1*H*-benzimidazole (6o)[24,25]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.033 mmol) and 1,2-benzenediamine (33.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **6o** was obtained (22.0 mg, 62% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 7.92 (s, 1H), 7.82 (d, *J* = 8.6 Hz, 1H), 7.33-7.21 (m, 5H), 7.15 (d, *J* = 7.9 Hz, 2H), 5.31 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 143.9, 143.3, 135.5, 133.9, 129.0, 128.3, 127.1, 123.1, 122.3, 120.4, 110.1, 48.8.

1-tosyl-1*H*-benzimidazole (6p)[26]**:** Following General Procedure IV, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.033 mmol) and benzenesulfonamide (44.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **6p** was obtained (12.0 mg, 27% yield) as a pale yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.38 (s, 1H), 7.88-7.84 (m, 3H), 7.76 (d, *J* = 8.3 Hz, 1H), 7.39-7.33 (m, 2H), 7.29 (d,

J = 8.3 Hz, 2H) 2.36 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 146.3, 144.1, 141.3, 134.6, 130.8, 130.4, 127.3, 125.7, 125.7, 121.1, 112.6, 21.8.

quinazolin-4(*3H*)-one (10a)[22,27]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 2-aminobenzamide (46.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:3), Compound **10a** was obtained (40.0 mg, 83% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 10.84 (br, 1H), 8.32 (ddd, *J* = 8.0, 1.5, 0.5 Hz, 1H), 8.11 (s, 1H), 7.85-7.77 (m, 2H), 7.56 (ddd, *J* = 8.0, 6.9, 1.5 Hz,1H); ^{13}C NMR (100 MHz, CD_3OD): δ 163.3, 149.7, 146.4, 135.9, 128.4, 127.8, 127.2, 123.8.

3-phenylquinazolin-4(3*H*)-one (10b)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-aminobenzanilide (36.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 1:1), Compound **10b** was obtained (33.0 mg, 89% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.38 (d, *J* = 8.0 Hz, 1H), 8.13 (s, 1H), 7.83-7.76 (m, 2H), 7.58-7.54 (m, 3H), 7.51-7.48 (m, 1H), 7.44-7.42 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.9, 147.9, 146.2, 137.6, 134.7, 129.8, 129.2, 127.8, 127.7, 127.3, 127.1, 122.5.

3-(p-tolyl)quinazolin-4(3*H*)-one (10c)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (50.0 mg, 0.34 mmol), pyridinium *p*-toluenesulfonate (17.0 mg, 0.068 mmol) and 4-methylanthranilanilide (76.0 mg, 0.34 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10c** was obtained (75.0 mg, 95% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.35 (dd, *J* = 8.2, 0.8 Hz, 1H), 8.10 (s, 1H), 7.80-7.73 (m, 2H), 7.53 (ddd, *J* = 8.2, 6.9, 1.6 Hz, 1H), 7.35-7.28 (m, 4H), 2.42 (s, 3H) ; ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.9, 147.9, 146.3, 139.2, 134.9, 134.5, 130.2, 127.6, 127.2, 126.8, 122.4, 21.2.

3-(4-methoxyphenyl)quinazolin-4(3*H*)-one (10d)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (1000 mg, 6.7 mmol), pyridinium *p*-toluenesulfonate (337.0 mg, 1.34 mmol) and 2-amino-*N*-(4-methoxyphenyl)benzamide (1624 mg, 6.7 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10d** was obtained (1573 mg, 93% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.36 (dd, *J* = 8.0, 0.8 Hz, 1H), 8.11 (s, 1H), 7.79-7.75 (m, 2H), 7.54 (ddd, *J* = 8.0, 6.9, 1.4 Hz, 1H), 7.35-7.31 (m, 2H), 7.06-7.02 (m, 2H), 3.87 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.1, 160.0, 148.0, 146.6, 134.6, 130.3, 128.3,

127.7, 127.3, 122.5, 114.9, 55.7.

3-(4-Fluorophenyl)quinazolin-4(3*H*)-one (10e)[28]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-(4-fluorophenyl)benzamide (39.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10e** was obtained (38.0 mg, 95% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.35 (dd, *J* = 8.0, 1.4 Hz, 1H), 8.09 (s, 1H), 7.83-7.75 (m, 2H), 7.56 (ddd, *J* = 8.0, 6.9, 1.4 Hz, 1H), 7.44-7.39 (m, 2H), 7.27-7.21 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 163.9, 161.5, 160.9, 147.9, 145.9, 134.8, 133.5, 133.4, 129.1, 128.9, 127.9, 127.2, 122.3, 116.9, 116.7.; ^{19}F NMR (376 MHz, $CDCl_3$): δ -113.0 (m, C-*F*).

3-(4-Chlororophenyl)quinazolin-4(3*H*)-one (10f)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-(4-chlorophenyl)benzamide (41.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10f** was obtained (42.0 mg, 97% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.37 (ddd, *J* = 8.0, 1.5, 0.6 Hz, 1H), 8.09 (s, 1H), 7.84-7.63 (m, 2H), 7.59-7.51 (m, 3H), 7.40-7.37 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.7, 147.9, 145.7, 135.9, 135.2, 134.9, 129.9, 128.4, 127.9, 127.8, 127.3, 122.3.

3-(4-Bromophenyl)quinazolin-4(3*H*)-one (10g)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (13.0 mg, 0.084 mmol), pyridinium *p*-toluenesulfonate (4.0 mg, 0.0167 mmol) and 2-amino-*N*-(4-bromophenyl)benzamide (25.0 mg, 0.084 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10g** was obtained (24.0 mg, 94% yield) as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.35(ddd, *J* = 8.0, 1.4, 0.6 Hz, 1H), 8.08 (s, 1H), 7.83-7.75 (m, 2H), 7.70-7.67 (m, 2H), 7.56 (ddd, *J* = 8.0, 6.8, 1.4 Hz, 1H), 7.34-7.31 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.7, 147.9, 145.6, 136.5, 134.9, 132.9, 128.7, 127.9, 127.8, 127.3, 123.3, 122.3.

3-(4-Iodophenyl)quinazolin-4(3*H*)-one (10h)[29]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-(4-iodophenyl)benzamide (57.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10h** was obtained (55.0 mg, 95% yield) as a pale

yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.35 (ddd, *J* = 8.0, 1.4, 0.6 Hz, 1H), 8.08 (s, 1H), 7.90-7.87 (m, 2H), 7.83-7.75 (m, 2H), 7.56 (ddd, *J* = 8.0, 7.0, 1.4 Hz, 1H), 7.20-7.17 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.6, 147.8, 145.5, 138.9, 137.2, 134.9, 128.9, 127.9, 127.8, 127.3, 122.3, 94.8.

3-Propylquinazolin-4(3*H*)-one (10i)[22]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-propylbenzamide (30.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10i** was obtained (28.0 mg, 88% yield) as white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.30 (ddd, *J* = 8.0, 1.5, 0.6 Hz, 1H), 8.02 (s, 1H), 7.74-7.68 (m, 2H), 7.49 (ddd, *J* = 8.0, 6.9, 1.4 Hz, 1H), 3.98-3.94 (m, 2H), 1.87-1.78 (m, 2H), 0.99 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.1, 148.2, 146.7, 134.2, 127.5, 127.3, 126.8, 122.3, 48.7, 22.7, 11.2.

3-Butylquinazolin-4(3*H*)-one (10j)[22,30]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-butylbenzamide (32.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10j** was obtained (29.0 mg, 84% yield) as white solid. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.31 (dd, *J* = 8.0, 1.3 Hz, 1H), 8.02 (s, 1H), 7.74-7.70 (m, 2H), 7.49 (ddd, *J* = 8.1, 6.9, 1.3 Hz, 1H), 4.00 (t, *J* = 7.4 Hz, 2H), 1.81-1.74 (m, 2H), 1.41 (sext, *J* = 7.4 Hz, 2H), 0.97 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.2, 148.2, 146.7, 134.2, 127.5, 127.3, 126.8, 122.3, 46.9, 31.5, 20.0, 13.8.

3-Isobutylquinazolin-4(3*H*)-one (10k): Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-(2-methylpropyl)benzamide (32.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10k** was obtained (30.0 mg, 90% yield) as Colorless oil. IR (KBr, cm^{-1}): 2951, 2915, 2849, 1659, 1608, 1563, 1471, 1372, 1108, 768, 695. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.28 (ddd, *J* = 8.0, 1.4, 0.6 Hz, 1H), 7.97 (s, 1H), 7.75-7.66 (m, 2H), 7.49-7.45 (m, 1H), 3.78 (d, *J* = 7.4 Hz, 2H), 2.23-2.13 (m, 1H), 0.96 (d, *J* = 6.7 Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.3, 148.2, 147.0, 134.3, 127.5, 127.3, 126.9, 122.3, 54.2, 28.3, 20.0. HRMS (EI) m/z: $[M]^+$ calcd for $C_{11}H_{14}N_2O_2$ 202.1106; found 202.1099.

3-Dodecylquinazolin-4(3*H*)-one (10l): Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-dodecylbenzamide (51.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10l** was obtained (49.0 mg, 93% yield) as White solid, m.p. 57-61 ^{o}C. IR (KBr, cm^{-1}): 2950, 2915, 2849, 1656, 1608, 1471, 1371, 1108, 767, 695. ^{1}H NMR (400 MHz, $CDCl_3$) : δ 8.32 (dd, *J* = 8.0, 1.4 Hz, 1H), 8.02 (s, 1H), 7.75-7.71 (m, 2H), 7.50 (ddd, *J* = 8.0, 6.9, 1.4 Hz, 1H), 4.01-3.97 (m, 2H), 1.82-1.74 (m, 2H), 1.41 (m, 18H), 0.87 (t, *J* = 6.9 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.2, 148.2, 146.7, 134.2, 127.5, 127.3, 126.8, 122.3, 47.2, 32.0, 29.7, 29.7, 29.6, 29.5, 29.5, 29.3, 26.8, 22.8, 14.2. HRMS (EI) m/z: $[M]^+$ calcd for $C_{11}H_{14}N_2O_2$ 314.2358; found 314.2363.

3-Cyclohexylquinazolin-4(3*H*)-one (10m)[31]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-cyclohexylbenzamide (37.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10m** was obtained (27.0 mg, 71% yield) as liquid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.31 (d, *J* = 8.0 Hz, 1H), 8.12 (s, 1H), 7.76-7.68 (m, 2H), 7.49 (t, 7.4 Hz, 1H), 4.85-4.79 (m, 1H), 2.02-1.93 (m, 4H), 1.81-1.47 (m, 5H), 1.31-1.21 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.8, 147.6, 144.0, 134.2, 127.4, 127.2, 127.1, 122.1, 53.5, 32.8, 26.0, 25.4.

3-Cyclopentylquinazolin-4(3*H*)-one (10n)[32]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-cyclopentylbenzamide (34.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10n** was obtained (27.0 mg, 75% yield) as pale yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.30 (ddd, *J* = 8.0, 1.4, 0.6 Hz, 1H), 7,75-7.67 (m, 2H), 7.48 (ddd, *J* = 8.0, 6.9, 1.4 Hz, 1H), 5.22-5.14 (m, 1H), 2.26-2.17 (m, 2H), 1.94-1.88 (m, 2H), 1.85-1.74 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.3, 147.7, 144.5, 134.2, 127.4, 127.3, 126.9, 121.9, 55.8, 32.3, 24.7.

3-Benzylquinazolin-4(3*H*)-one (10o)[28]**:** Following General Procedure V, using the *N*-formylbenzamide **2a** (25.0 mg, 0.17 mmol), pyridinium *p*-toluenesulfonate (8.0 mg, 0.034 mmol) and 2-amino-*N*-(phenylmethyl)benzamide (38.0 mg, 0.17 mmol). The product was then purified by column chromatography (Hexane/EtOAc, 2:1), Compound **10o** was obtained (28.0 mg, 71% yield) as white solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.33 (d, *J* = 8.0 Hz, 1H), 8.11 (s, 1H), 7.77-7.69 (m, 2H), 7.53-7.49

(m, 1H), 7.36-7.30 (m, 5H), 5.20 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 161.2, 148.2, 146.5, 135.8, 134.4, 129.2, 128.4, 128.1, 127.6, 127.5, 127.0, 122.3, 49.7.

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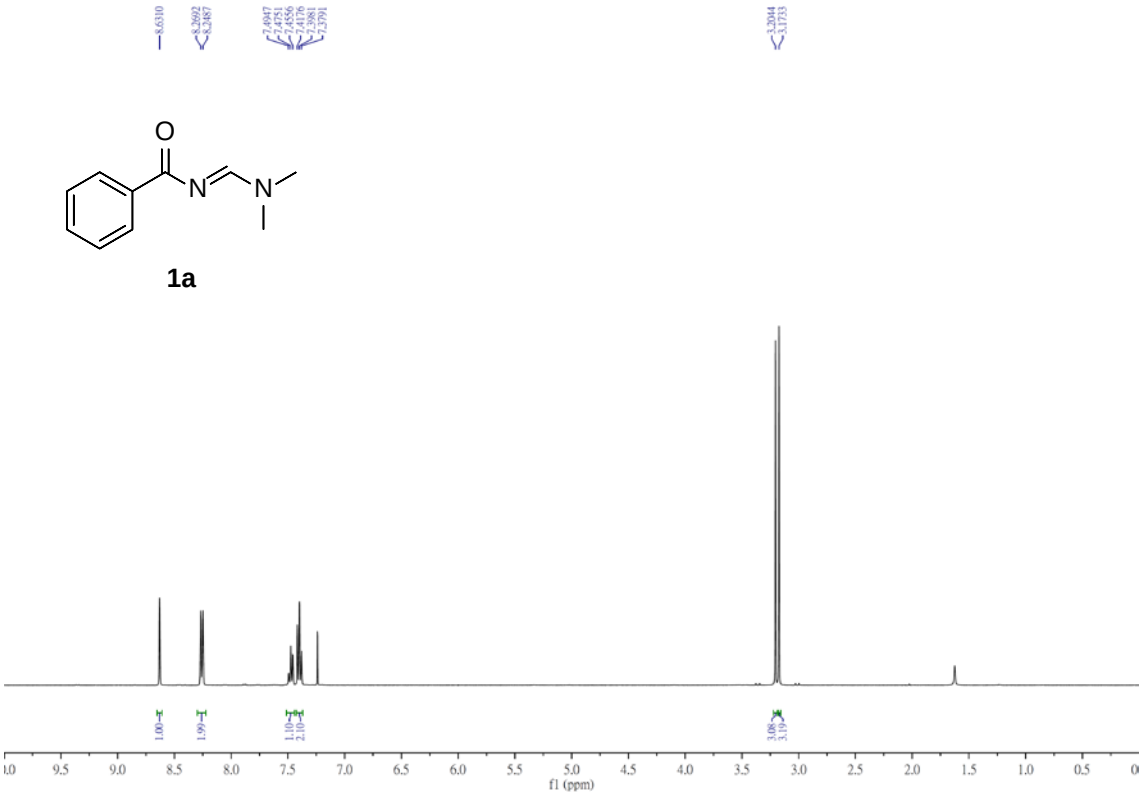
33. Compound **1f** cannot be detected by EI or ESI. Therefore, the mass of compound **1f** was detected by Quadrupole-Mass Filter method. Reference: Y. H. Chen, I. C. Lu, *Rapid Commun. Mass Spectrom*., 2019,1-5.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8.2487
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7.4751
7.4556
7.4176
7.3981
7.3791
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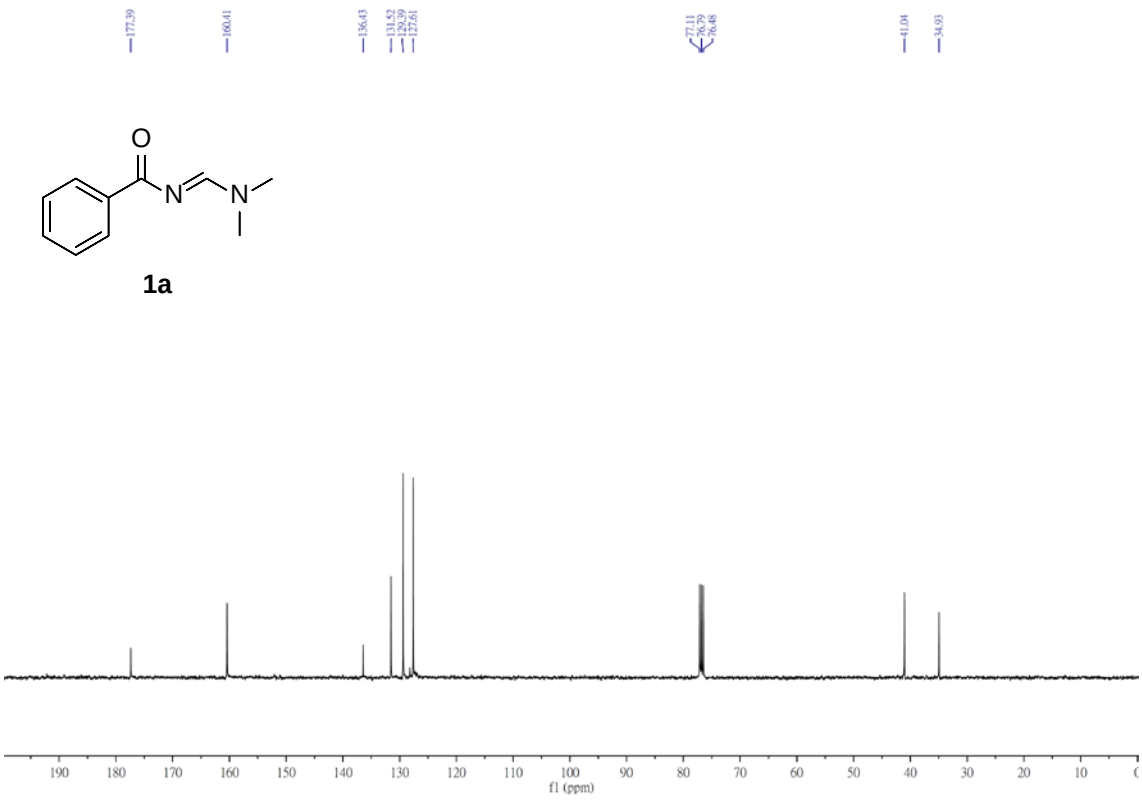
3.1733
1.00
1.99
1.10
2.10
3.08
3.19
f1 (ppm)

1a


177.39
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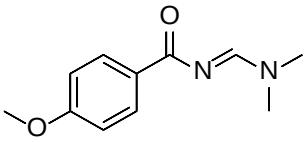
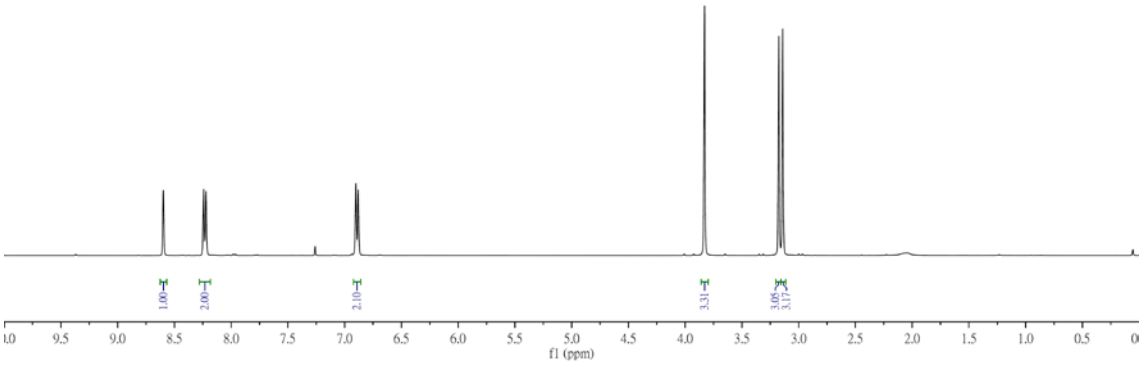
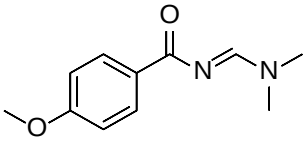
136.43
131.52
129.39
127.61
77.11
76.79
76.48
41.04
34.93
f1 (ppm)

1a



1b





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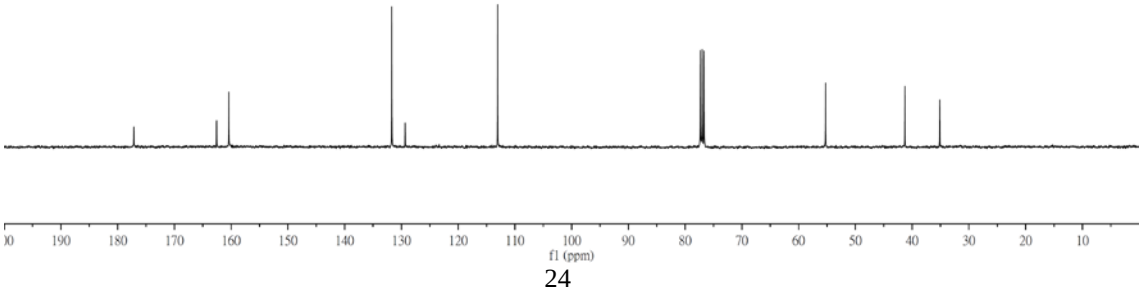

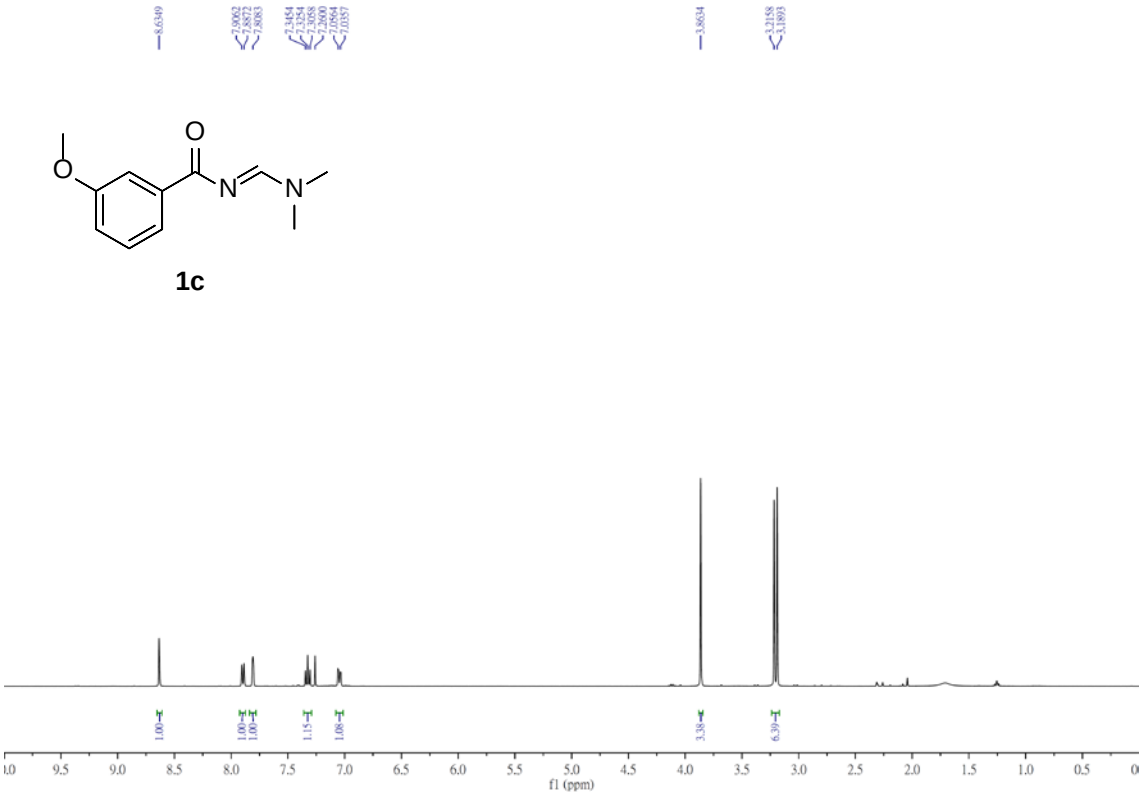
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7.8083
7.3454
7.3254
7.3058
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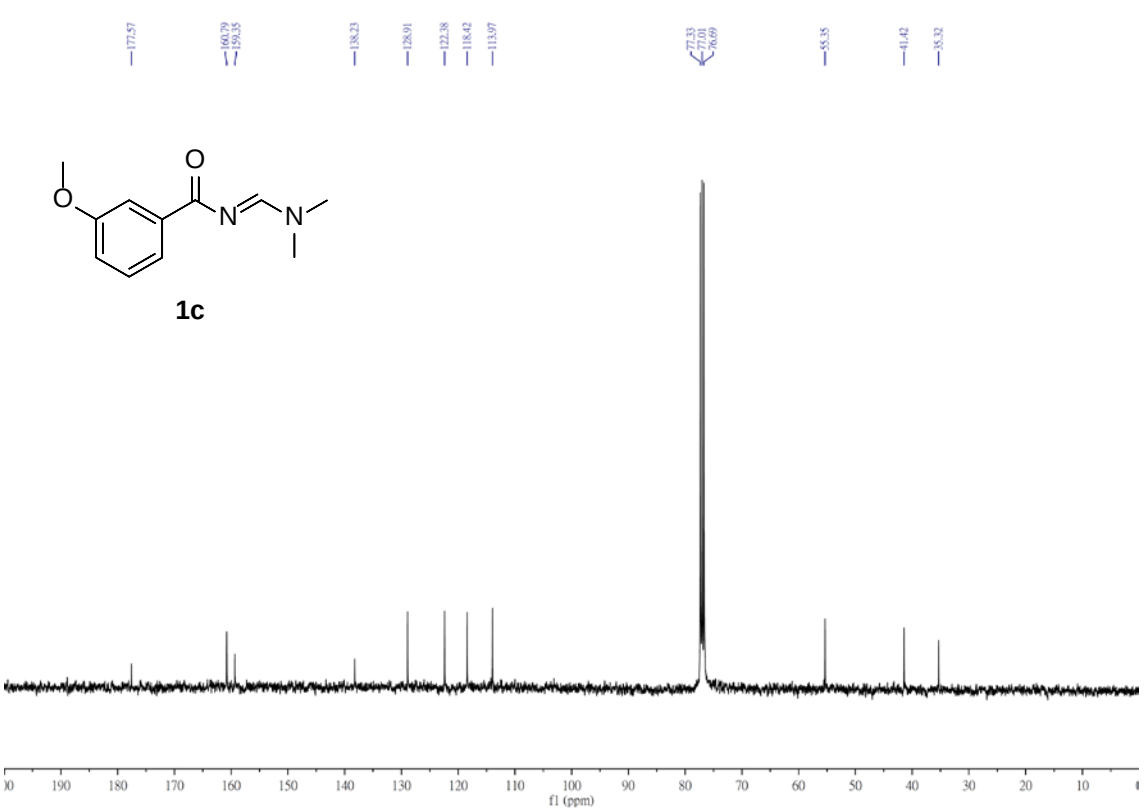
7.0564
7.0357
3.8634
3.2158
3.1893
O
O
N
N
1.00
1.00
1.00
1.15
1.08
3.38
6.39
f1 (ppm)

1c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138.23
128.91
122.38
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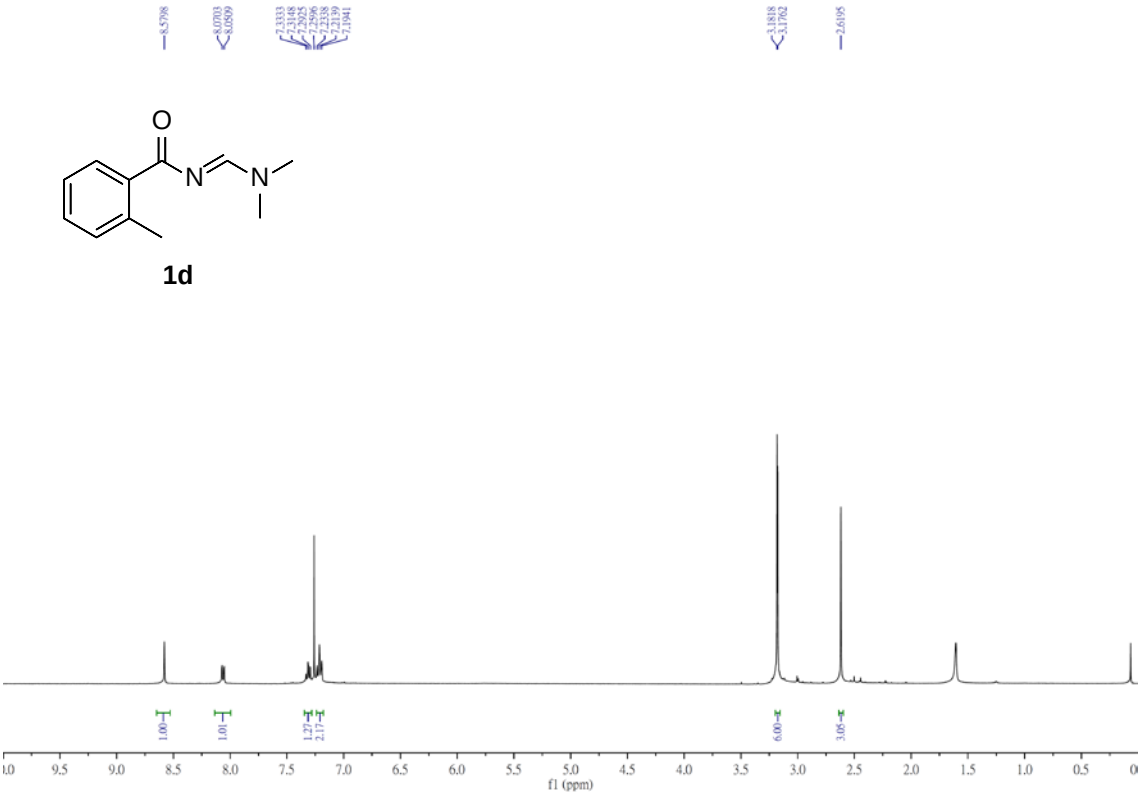
77.01
76.69
55.35
41.42
35.32
O
O
N
N
f1 (ppm)

1c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7.2139
7.1941
3.1838
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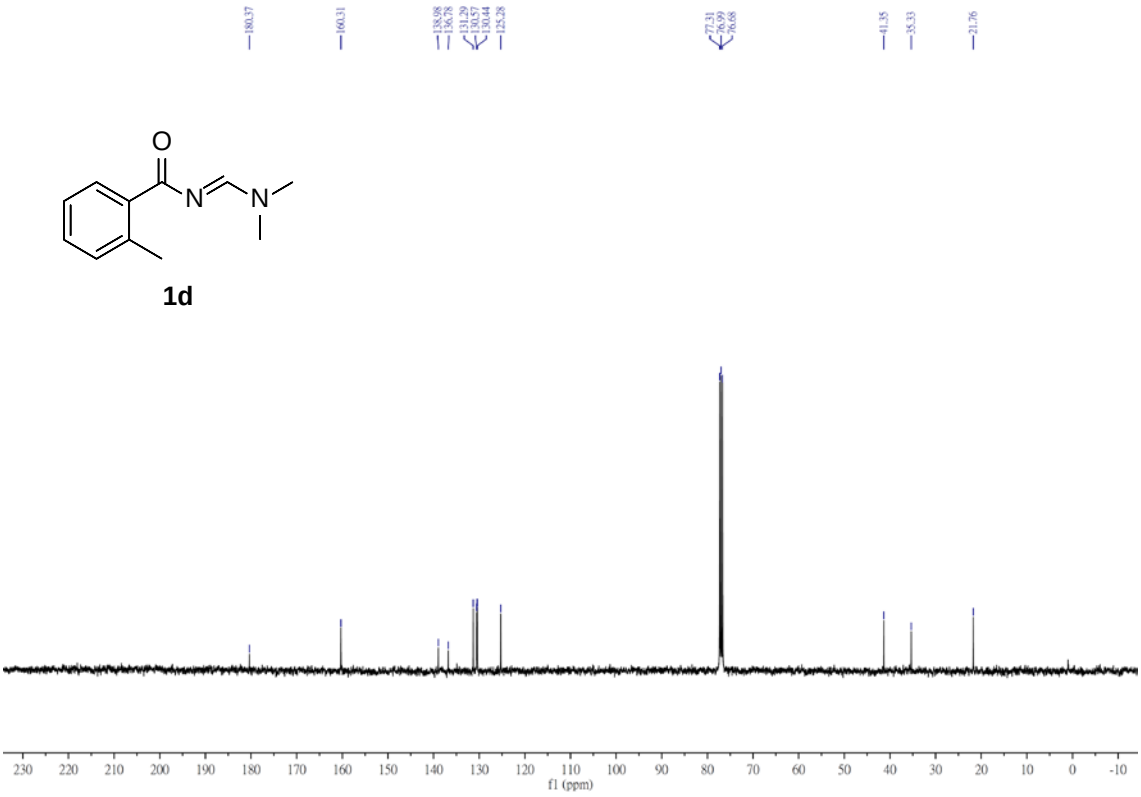
O
N
N
1.00
1.01
1.27
2.17
6.00
3.05
f1 (ppm)

1d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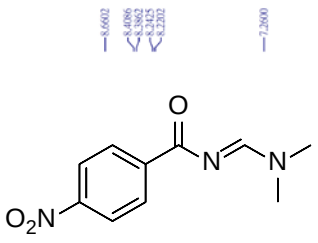
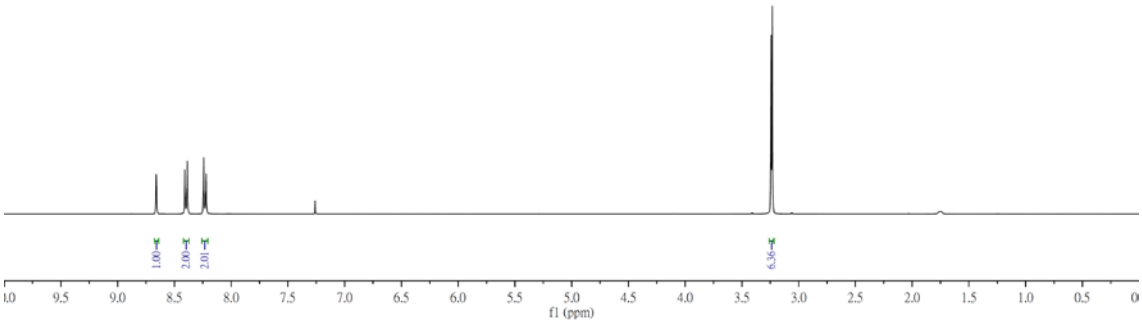
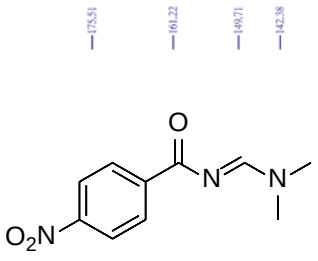
41.35
35.33
21.76
O
N
N
f1 (ppm)

1d



1e





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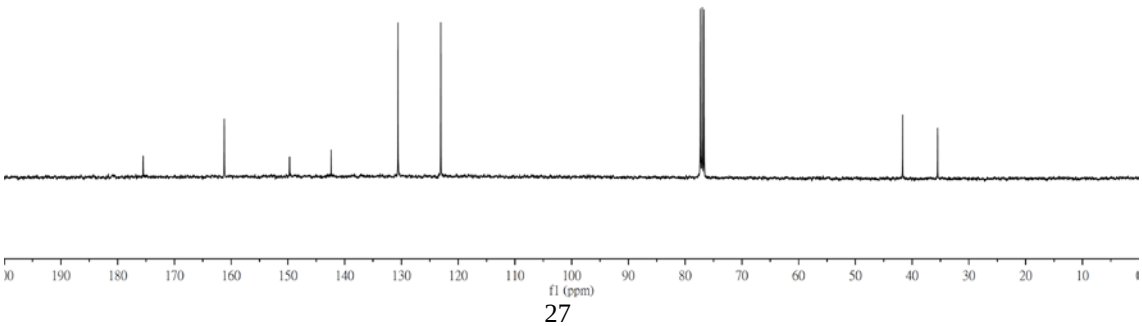
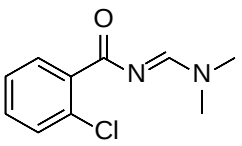
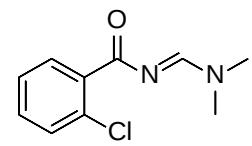




1f



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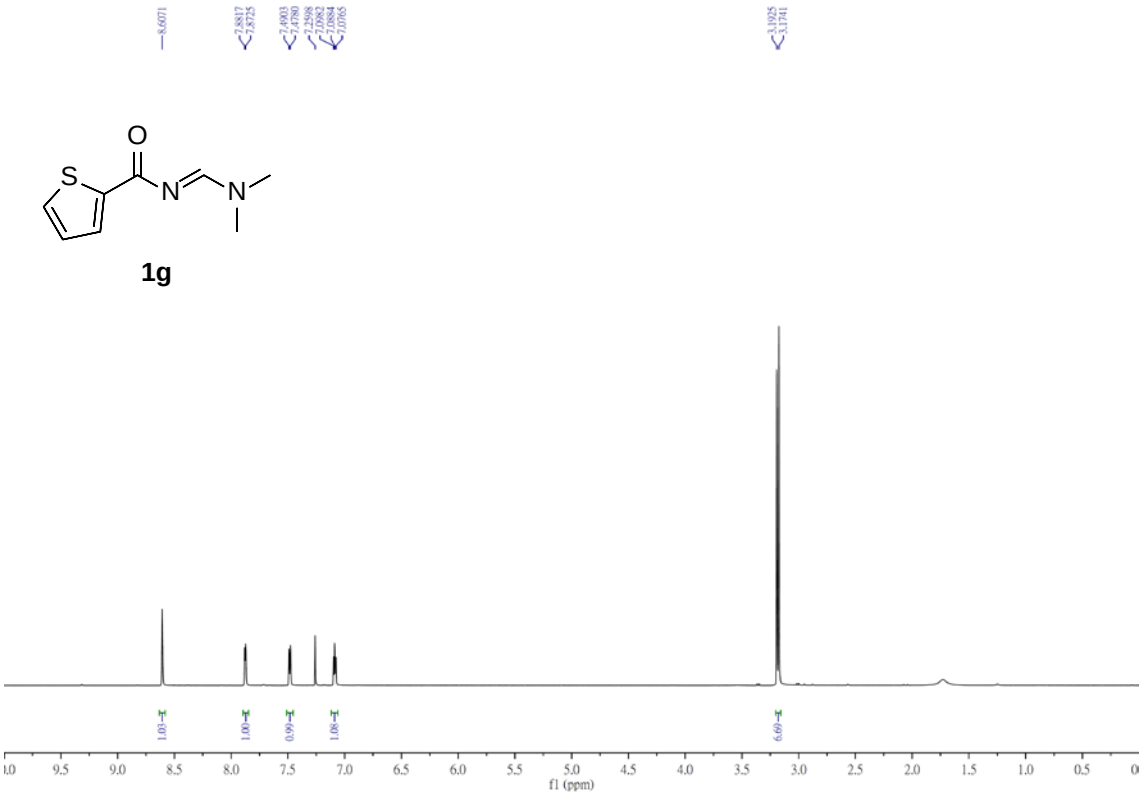
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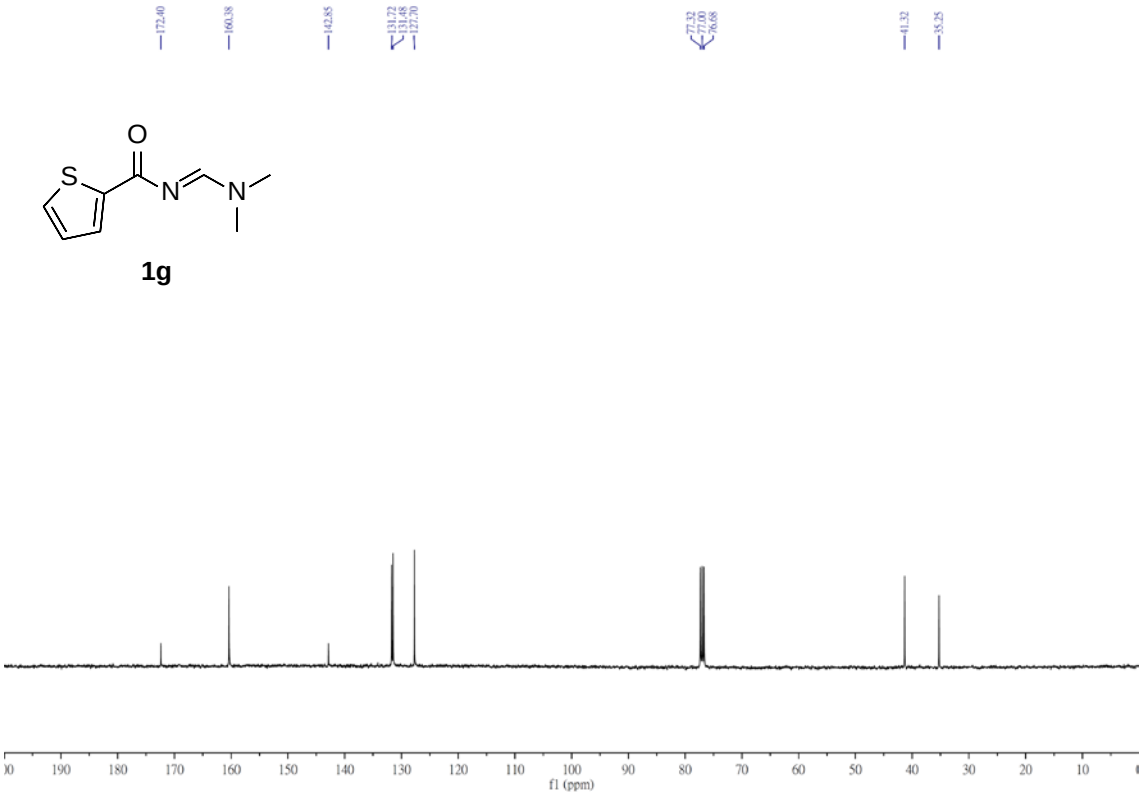
O
S
N
N
1.03
1.00
0.99
1.08
6.69
f1 (ppm)

1g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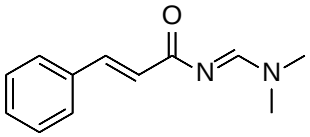
77.00
76.68
41.32
35.25
O
S
N
N
f1 (ppm)

1g



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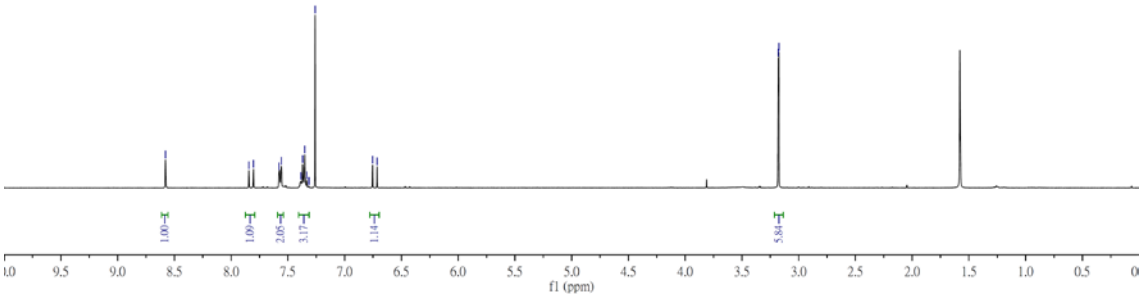

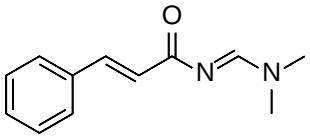

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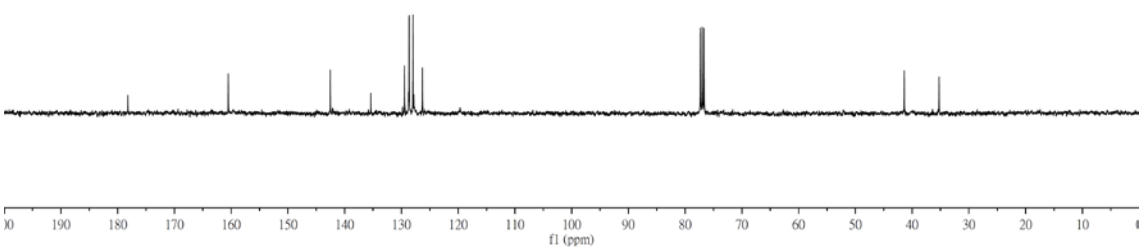

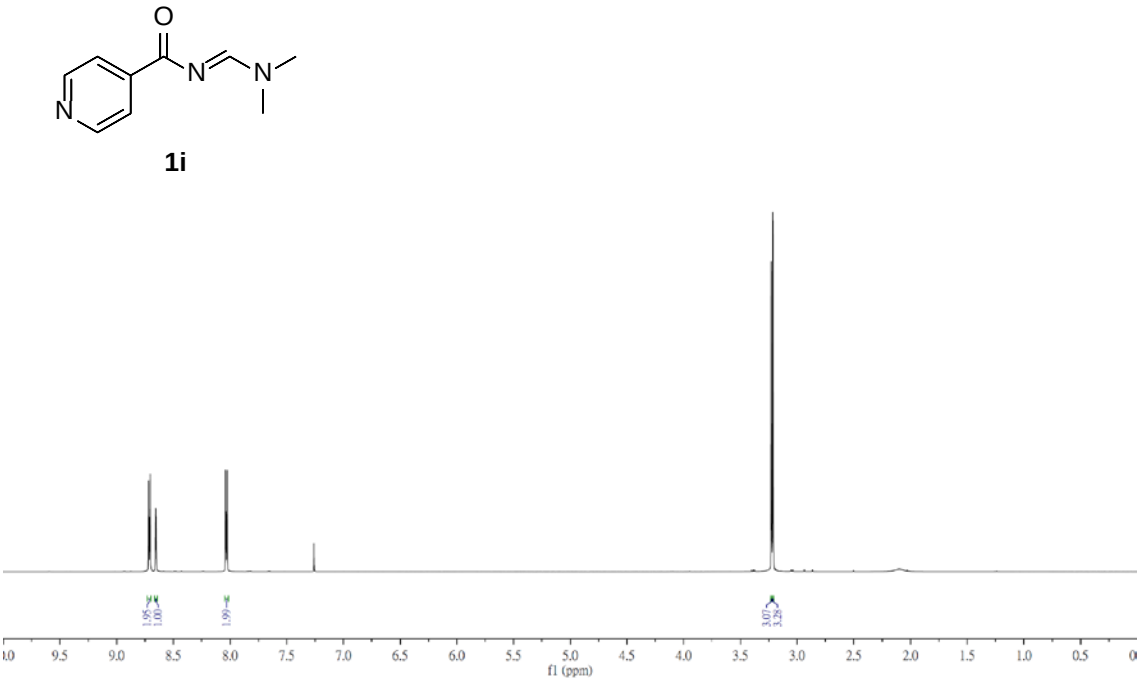
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8.0362
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8.0252
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3.2133
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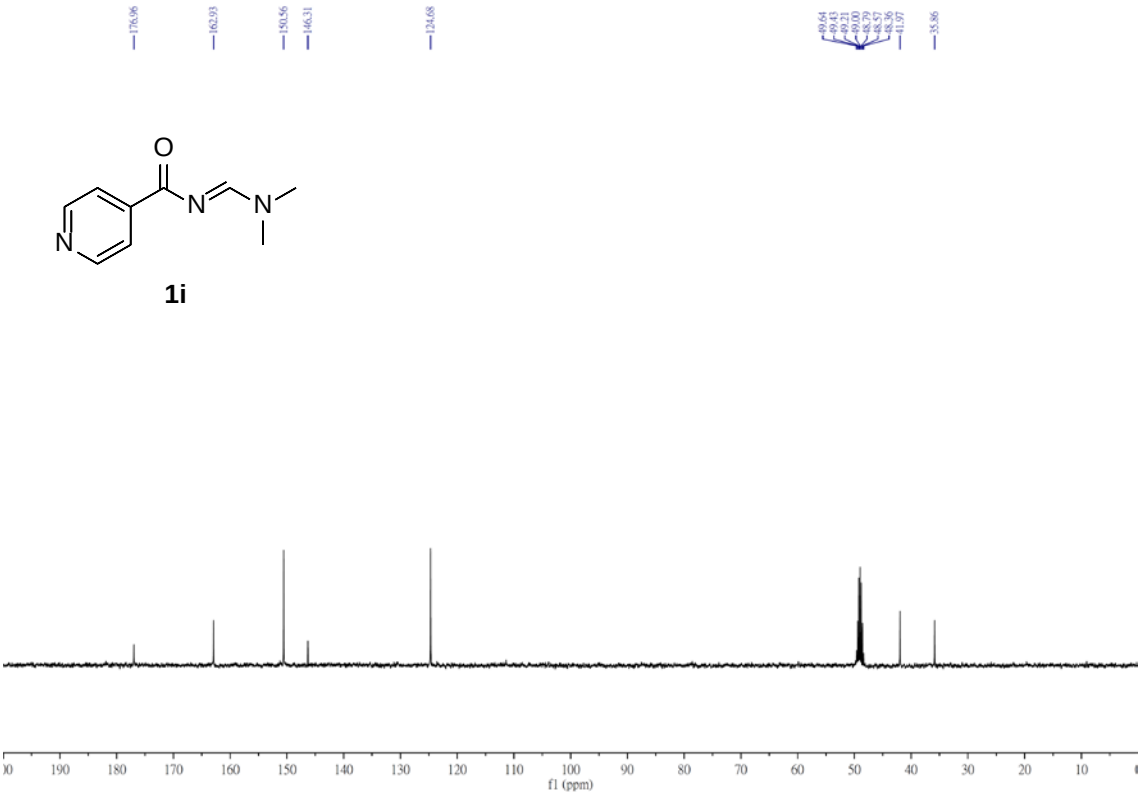
1.95
1.00
1.99
3.07
3.28
f1 (ppm)

1i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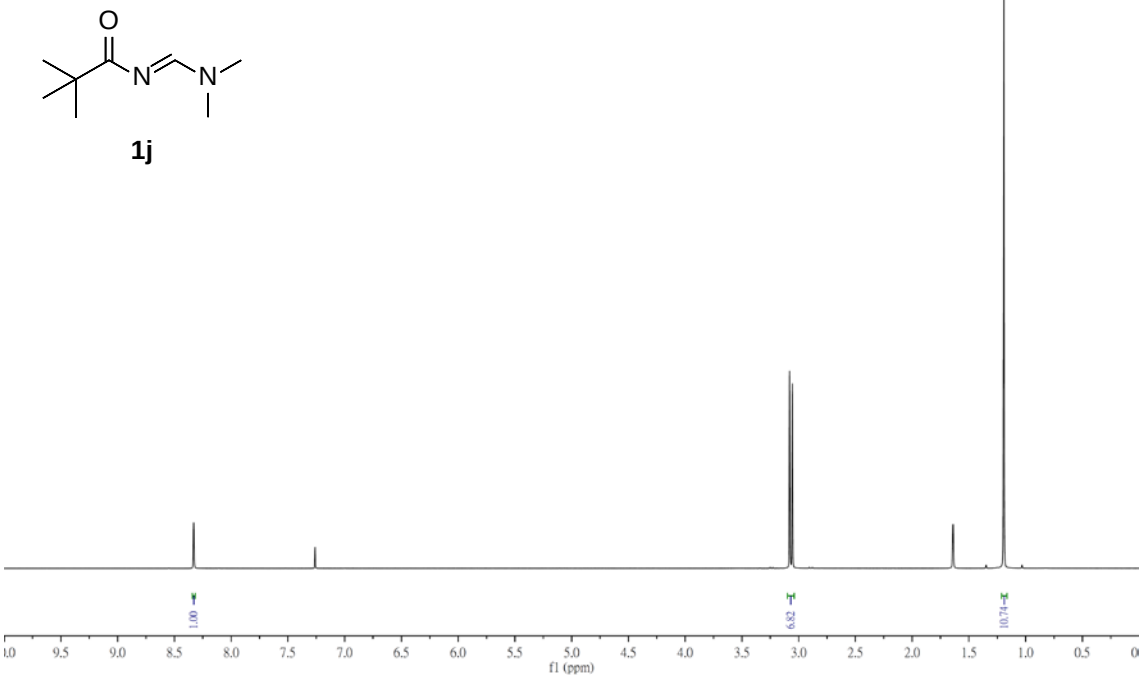
35.86
f1 (ppm)

1i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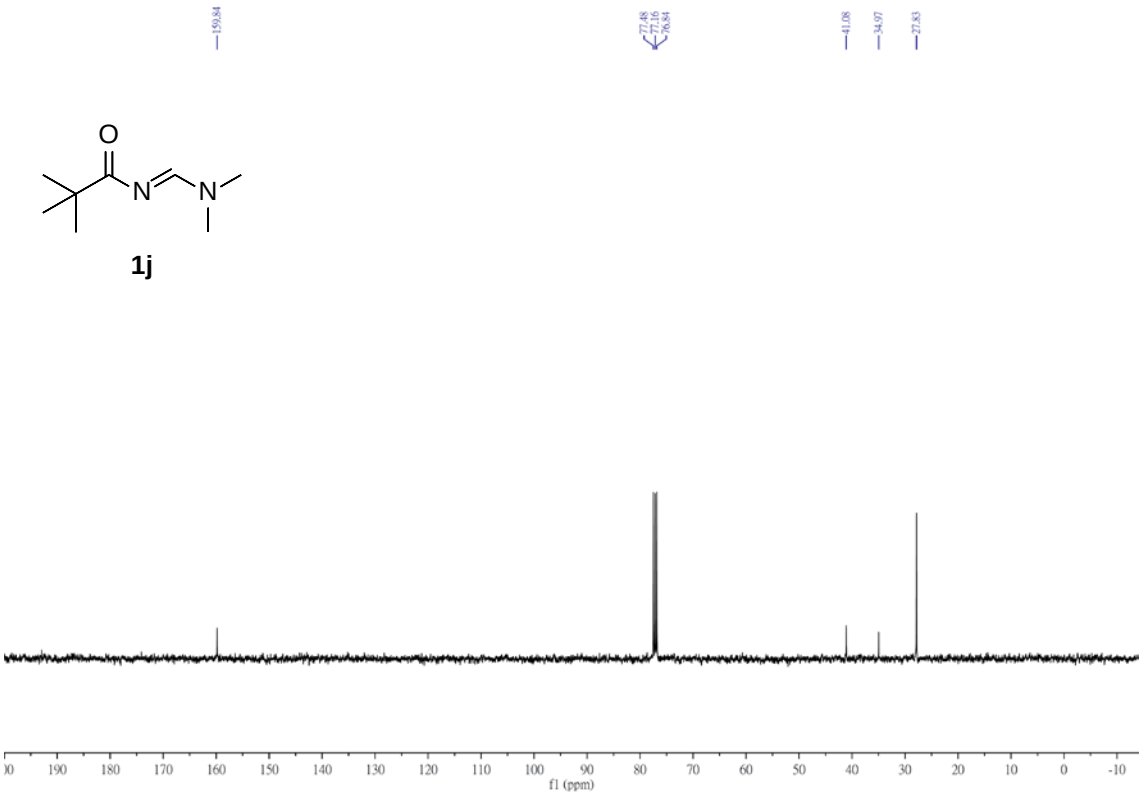
10.74
f1 (ppm)

1j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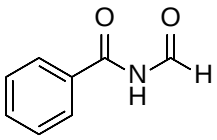
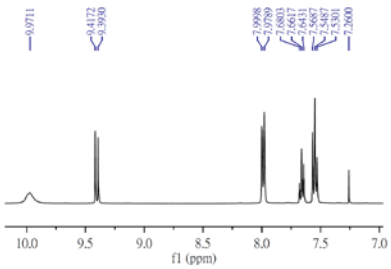
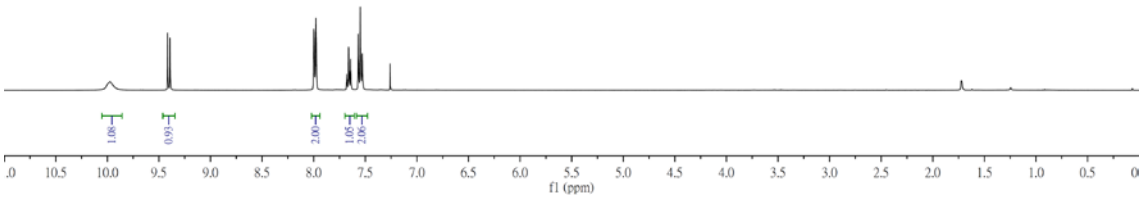
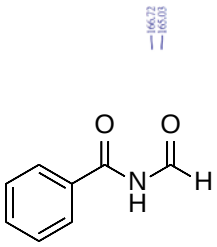
1j



2a







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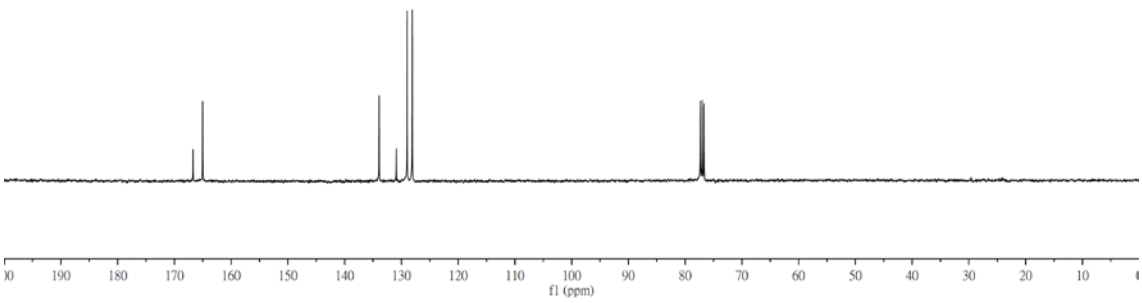

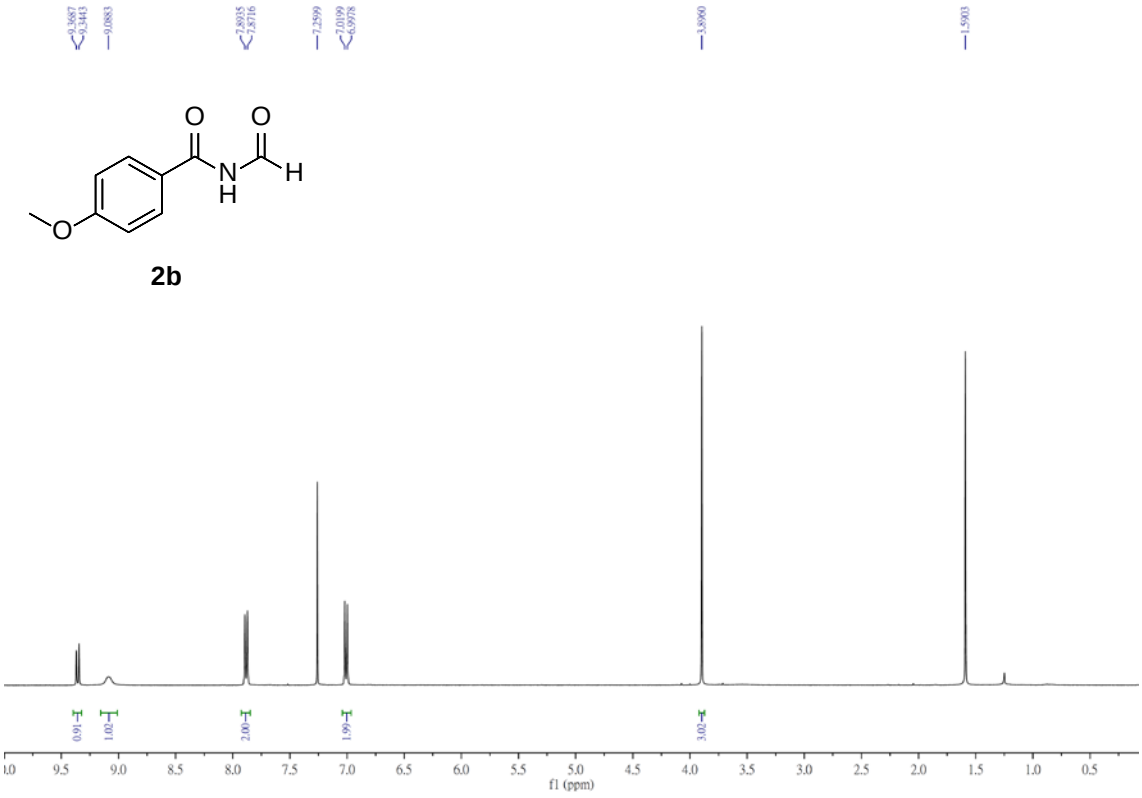
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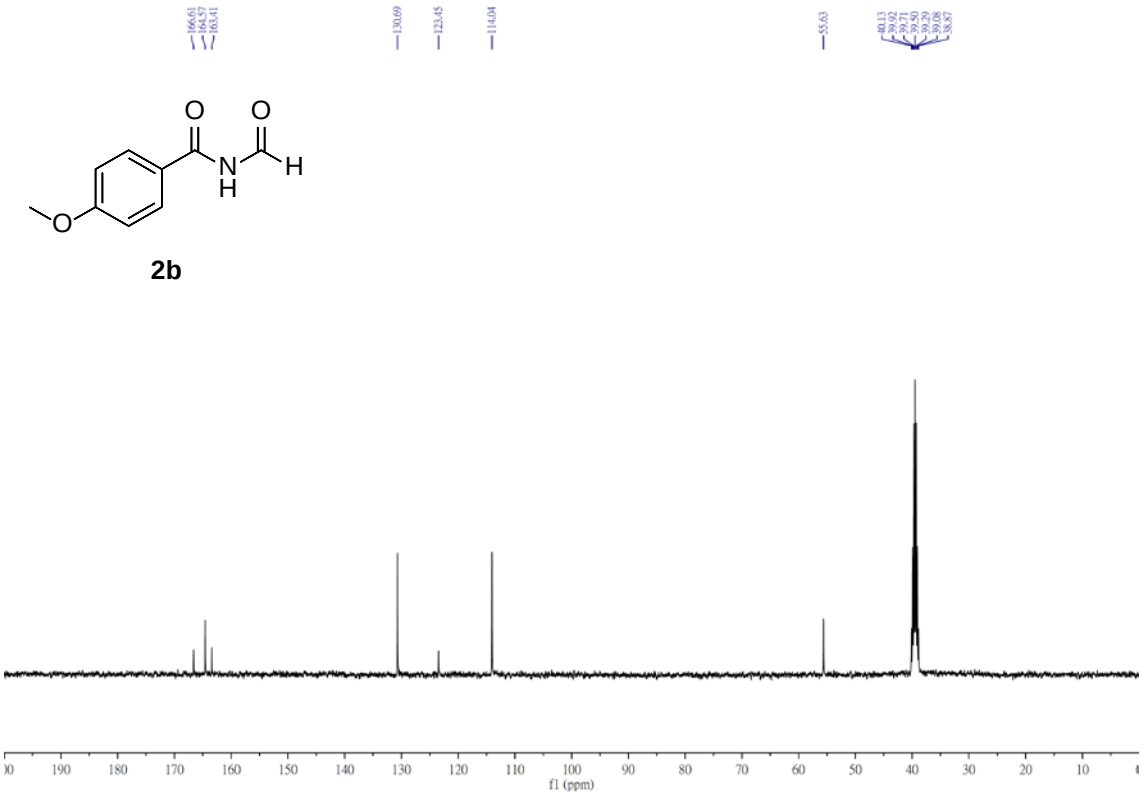
O
N
H
H
O
0.91
1.02
2.00
1.99
3.02
f1 (ppm)

2b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55.63
40.13
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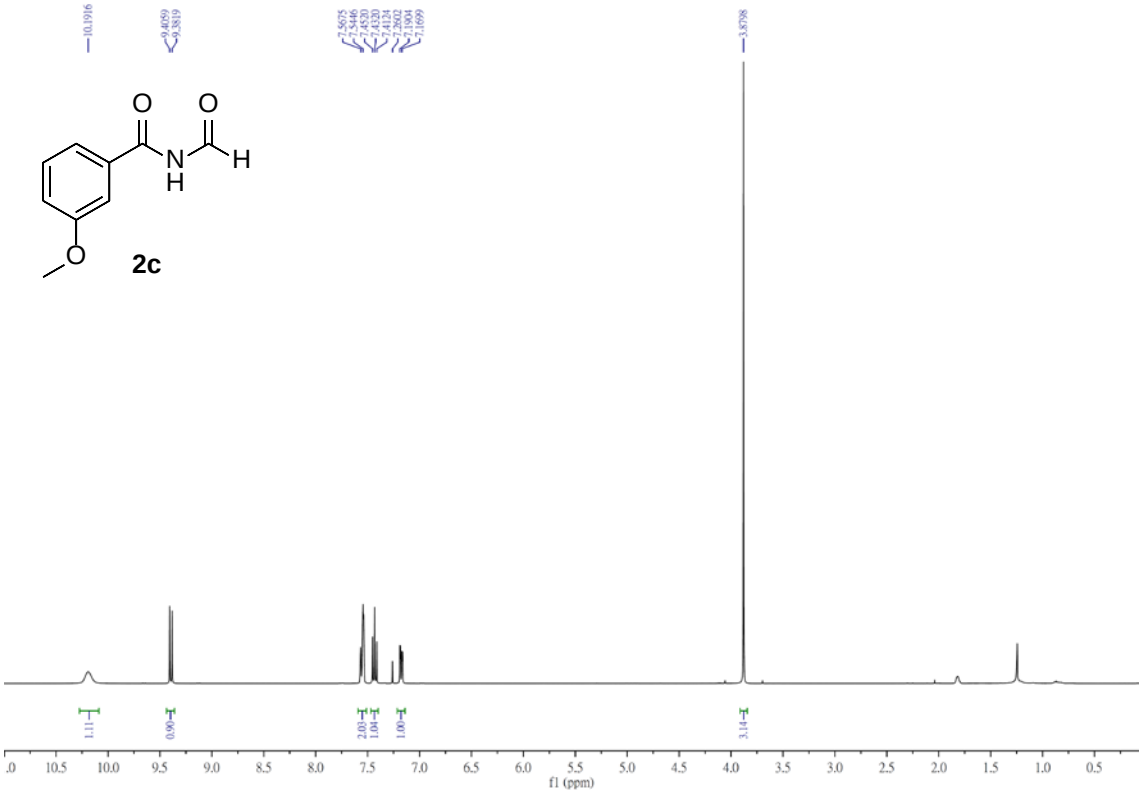
39.50
39.29
39.08
38.87
O
O
N
H
H
O
f1 (ppm)

2b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7.2802
7.1994
7.1699
3.8798
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O
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H
H
O
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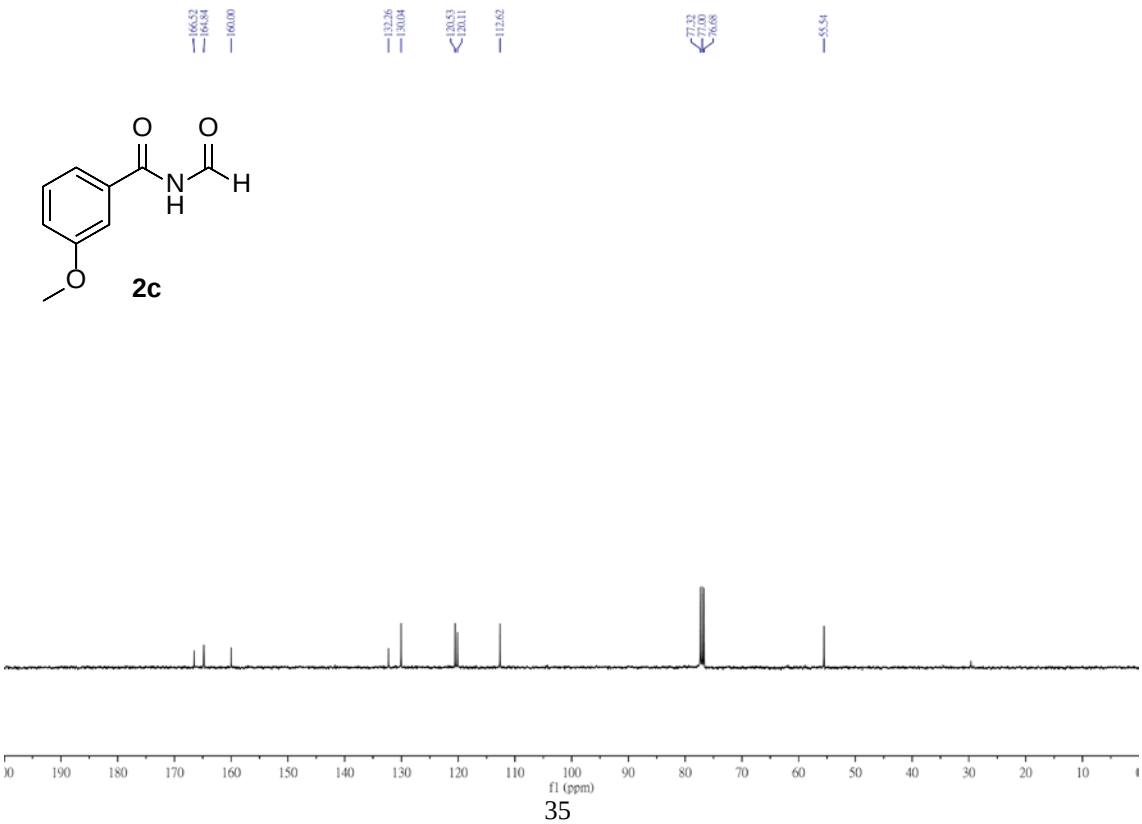
1.04
1.00
3.14
f1 (ppm)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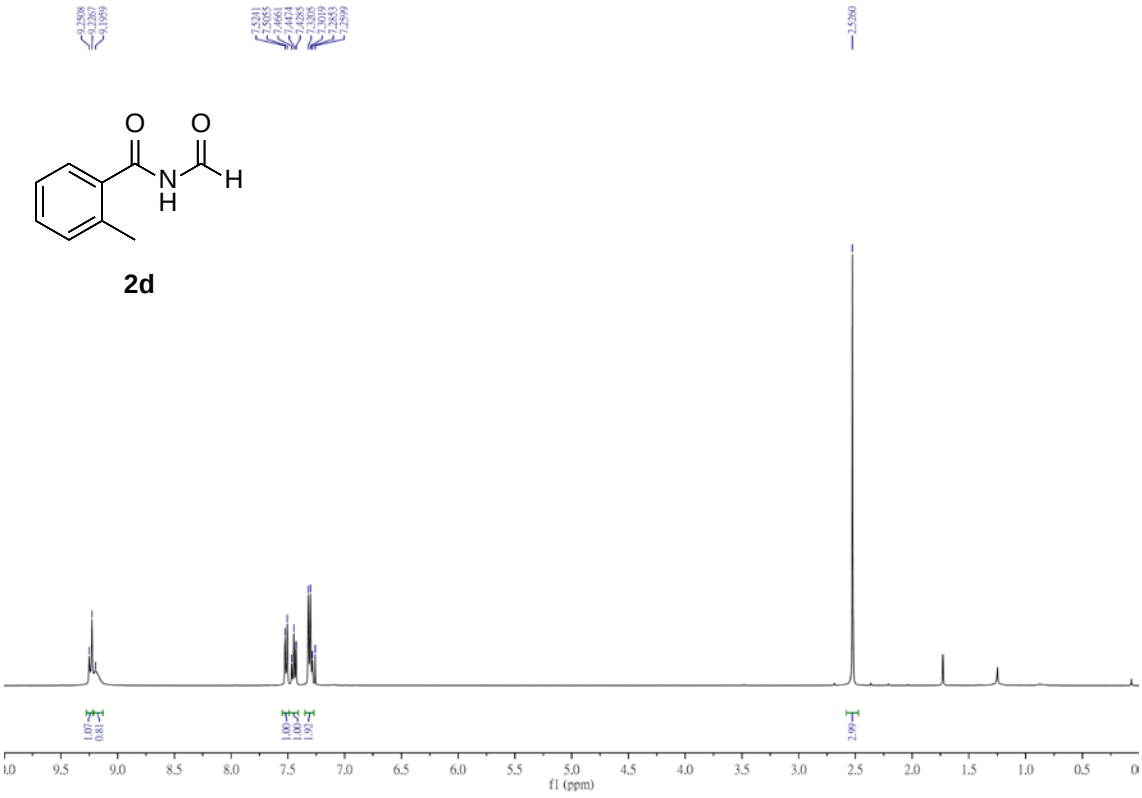
O
N
H
H
O
2c
f1 (ppm)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7.5695
7.4661
7.4414
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7.3039
7.2853
7.2599
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O
O
N
H
H
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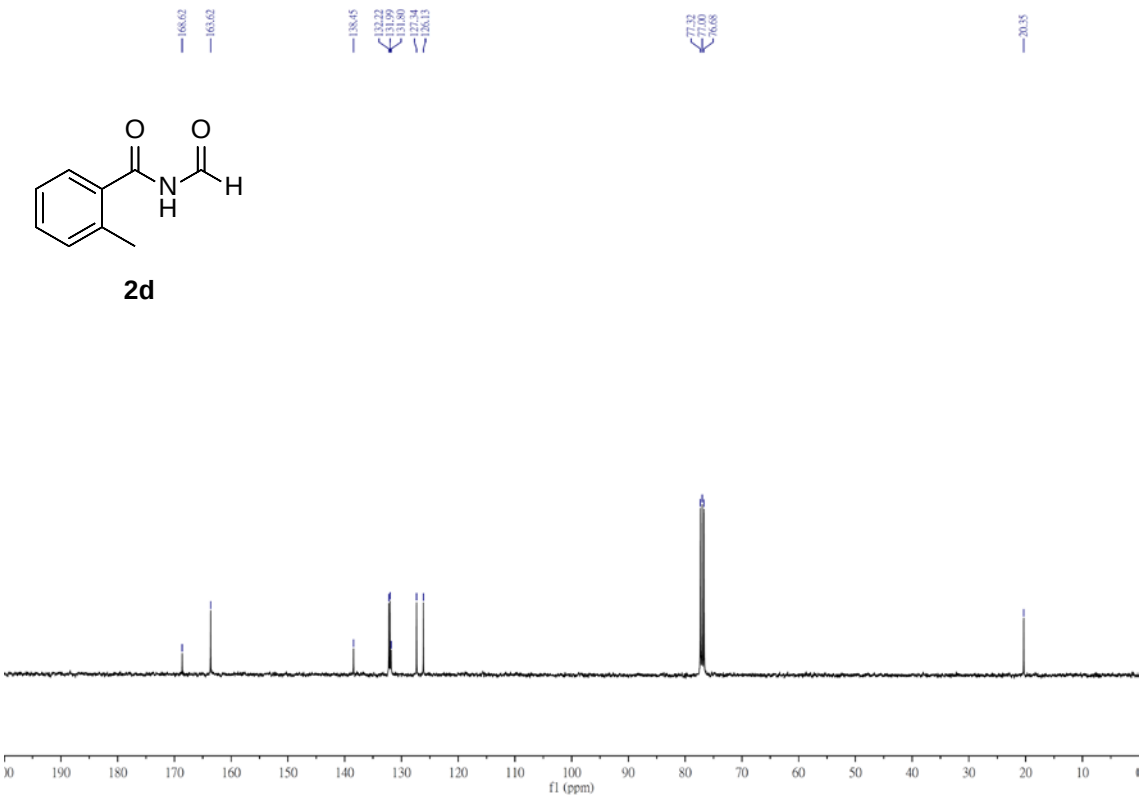
1.00
1.02
2.99
f1 (ppm)

2d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76.68
20.35
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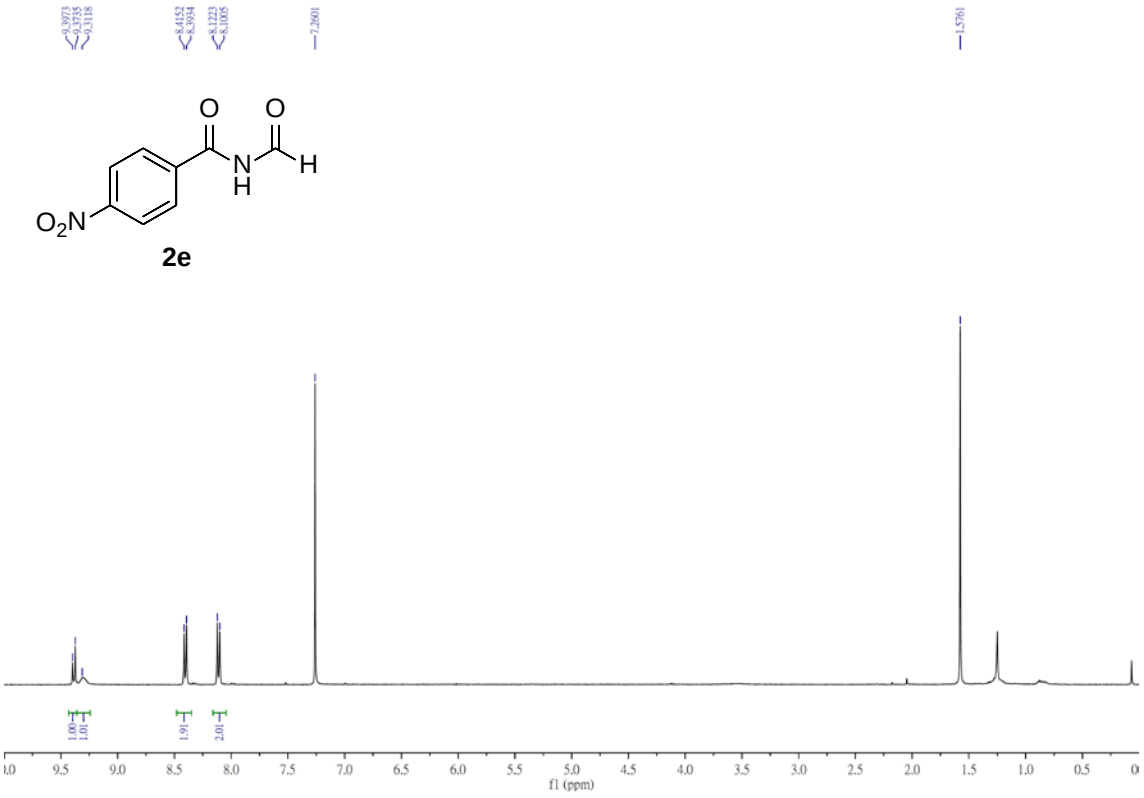
O
N
H
H
f1 (ppm)

2d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H
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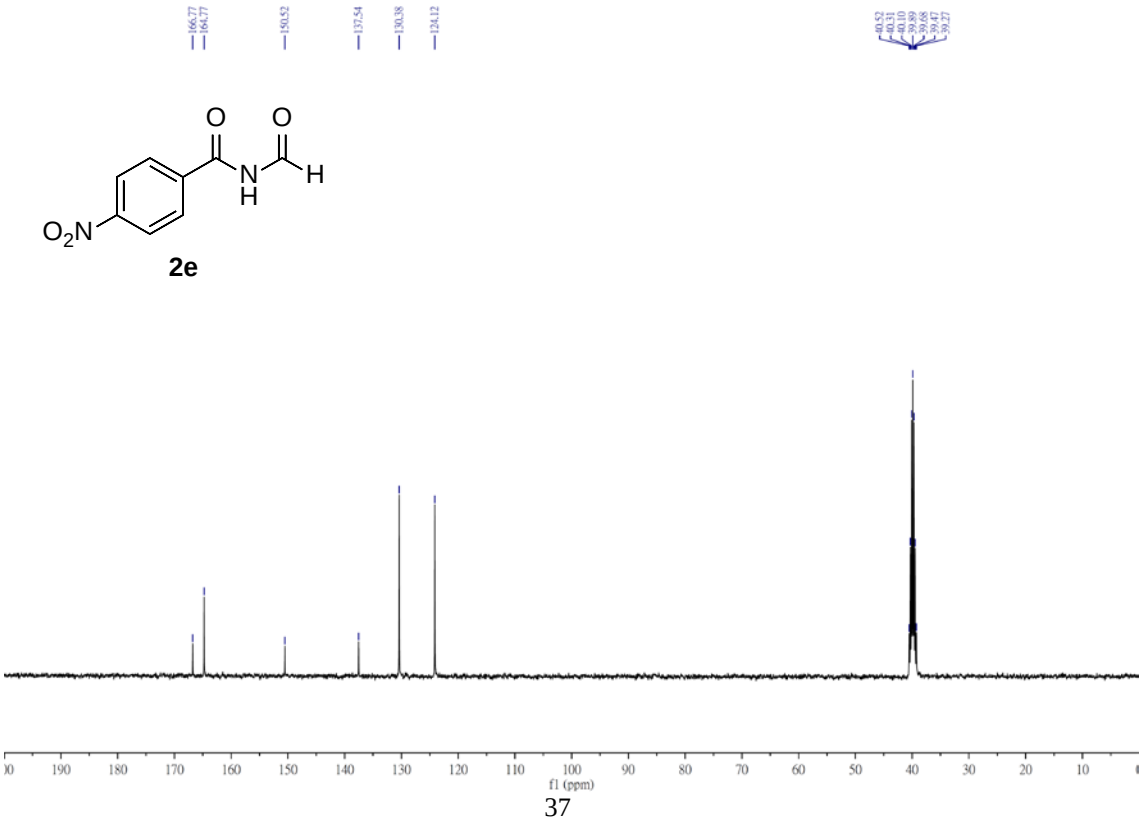
1.91
2.01
f1 (ppm)

2e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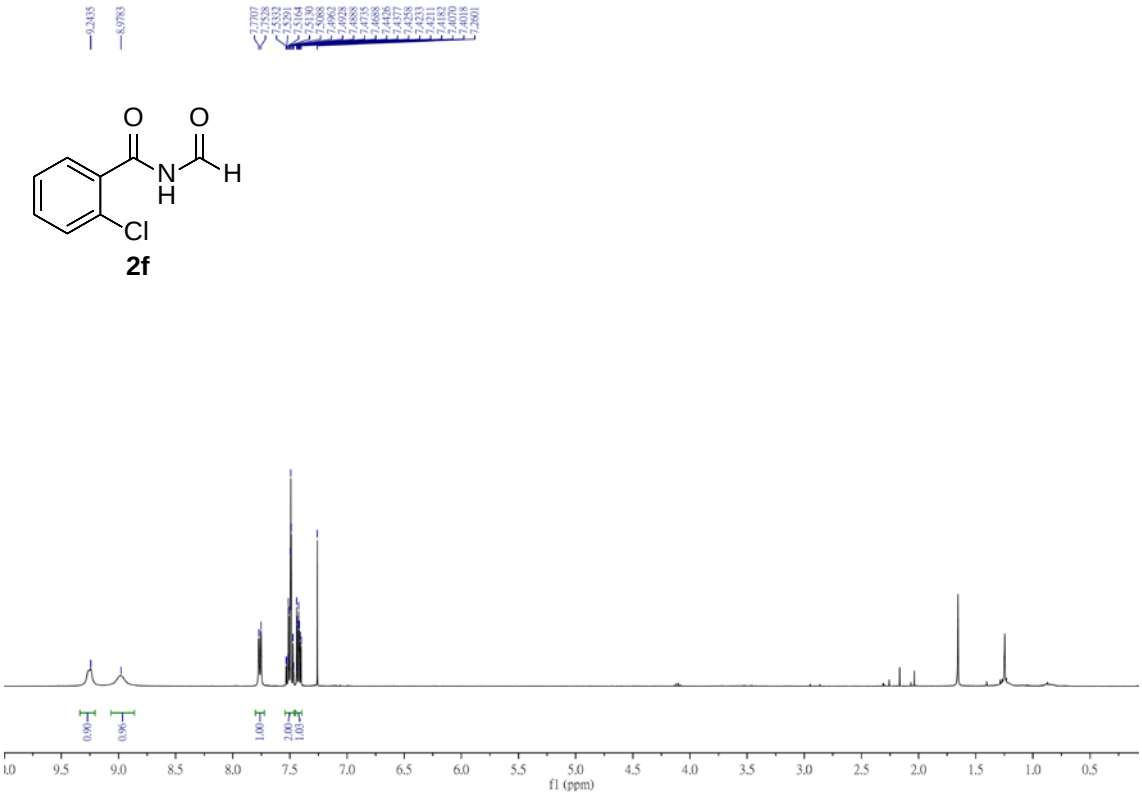
O
O
N
H
H
O_2N
f1 (ppm)

2e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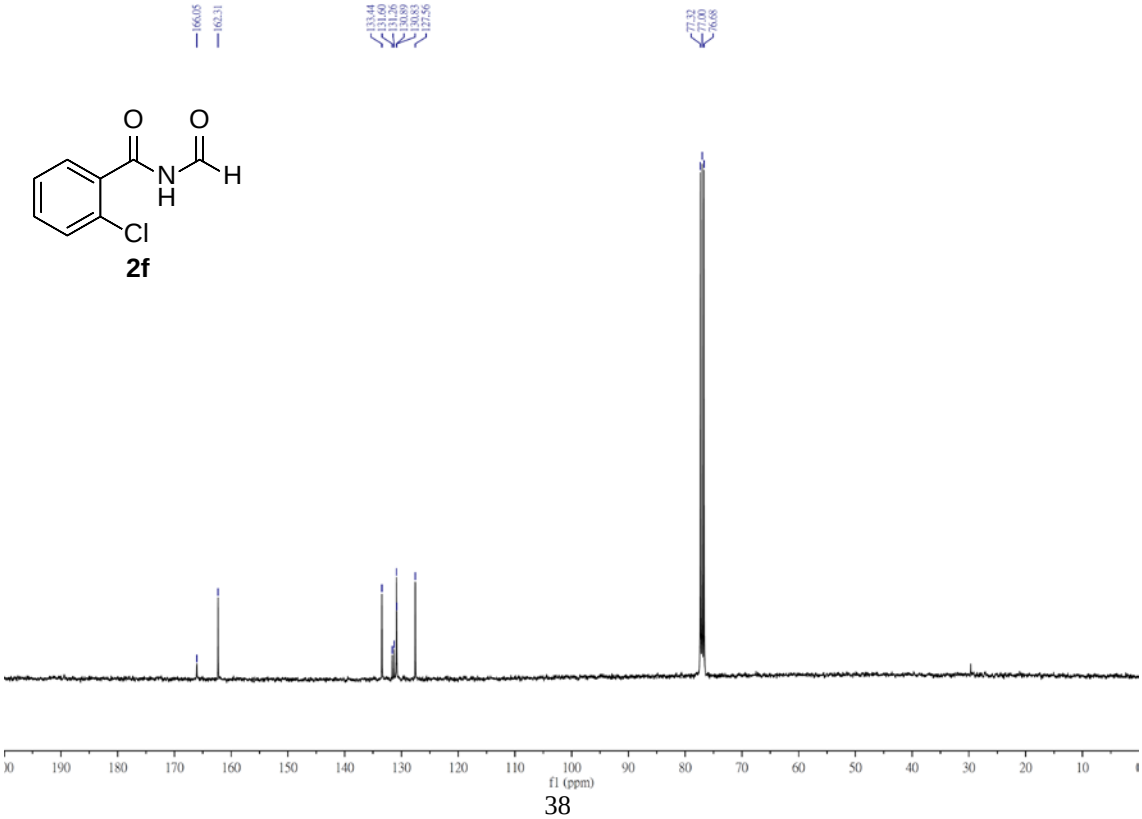
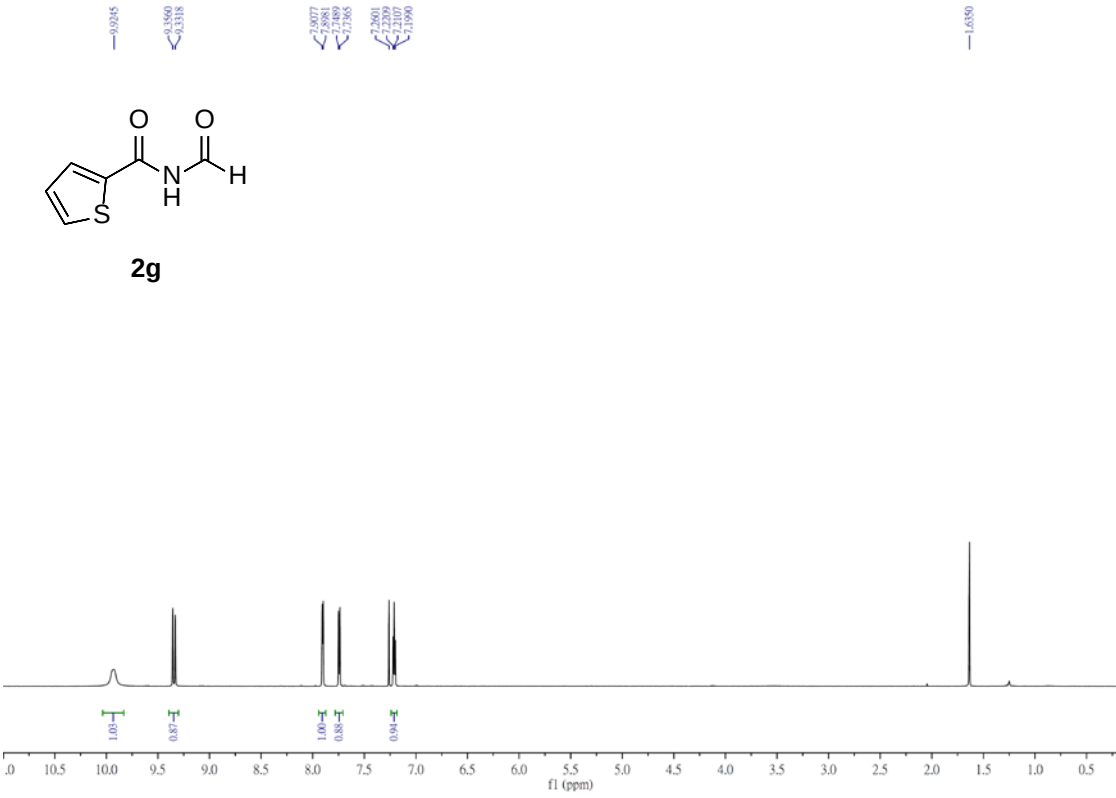
7.5164
7.5139
7.5088
7.4962
7.4928
7.4888
7.4775
7.4688
7.4426
7.4377
7.4338
7.4233
7.4211
7.4182
7.4070
7.4038
7.2601
O
O
N
H
H
Cl
0.90
0.96
1.00
2.00
1.03
f1 (ppm)

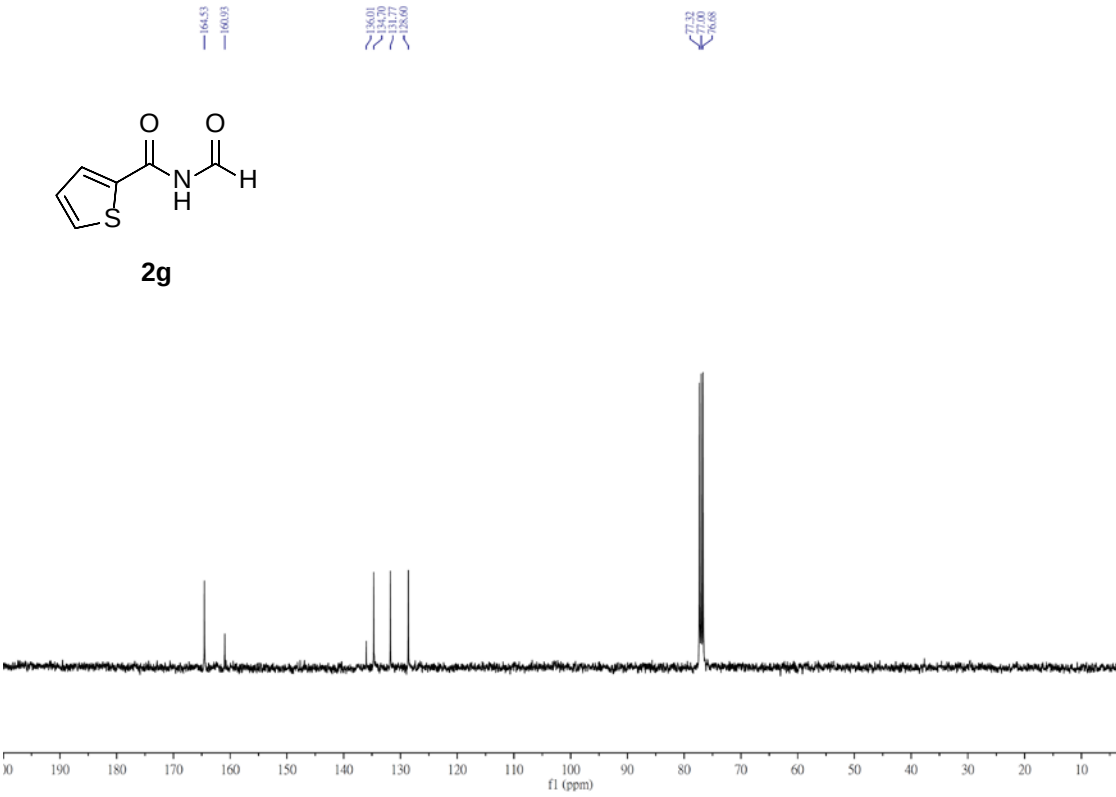
2f


166.05
162.31
133.44
131.60
131.26
130.89
130.83
127.56
77.32
77.00
76.68
O
O
N
H
H
Cl
f1 (ppm)

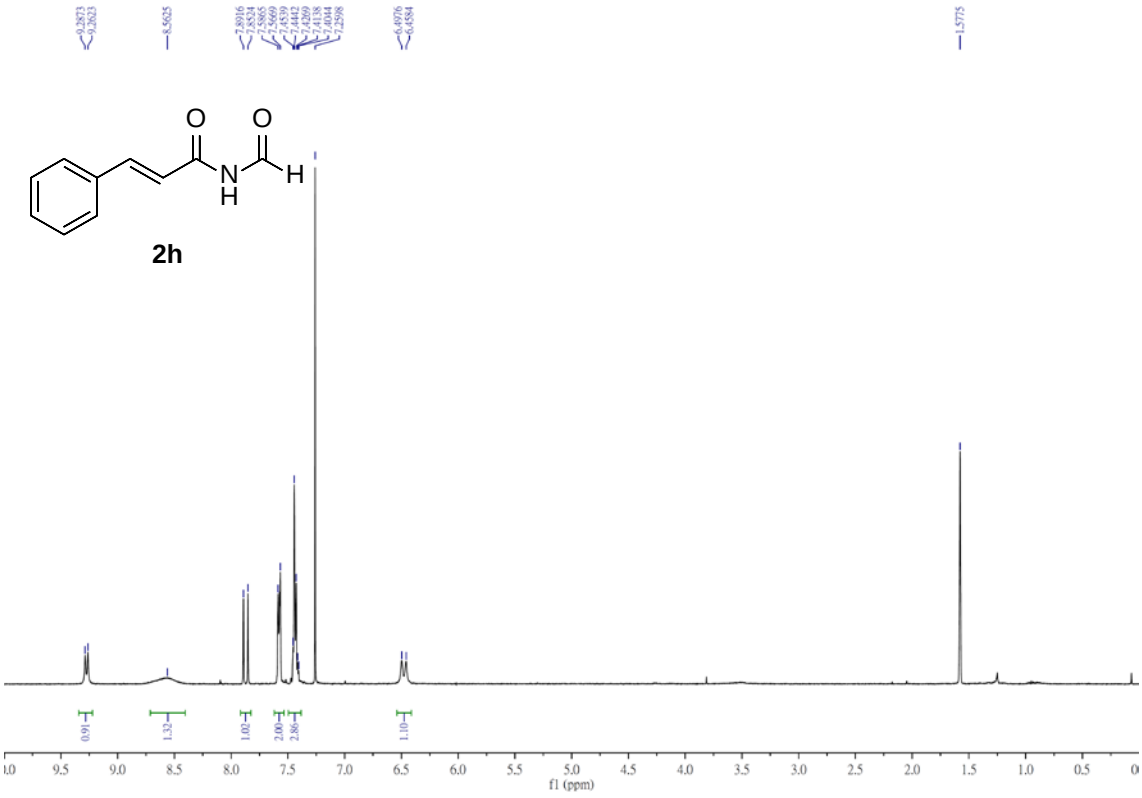
2f

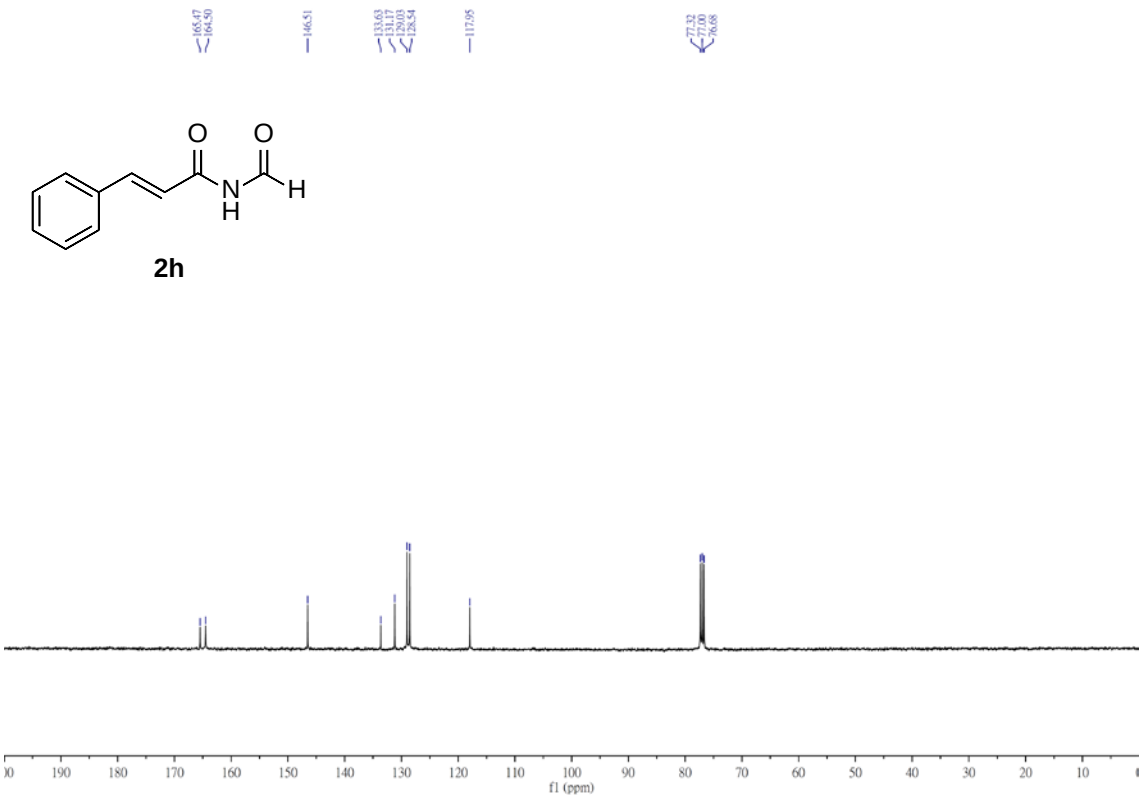

9.9245
8.3560
8.3318
7.9077
7.8881
7.7369
7.7265
7.2601
7.2309
7.2107
7.1990
1.6350
O
O
N
H
H
S
1.03
0.87
1.00
0.88
0.94
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

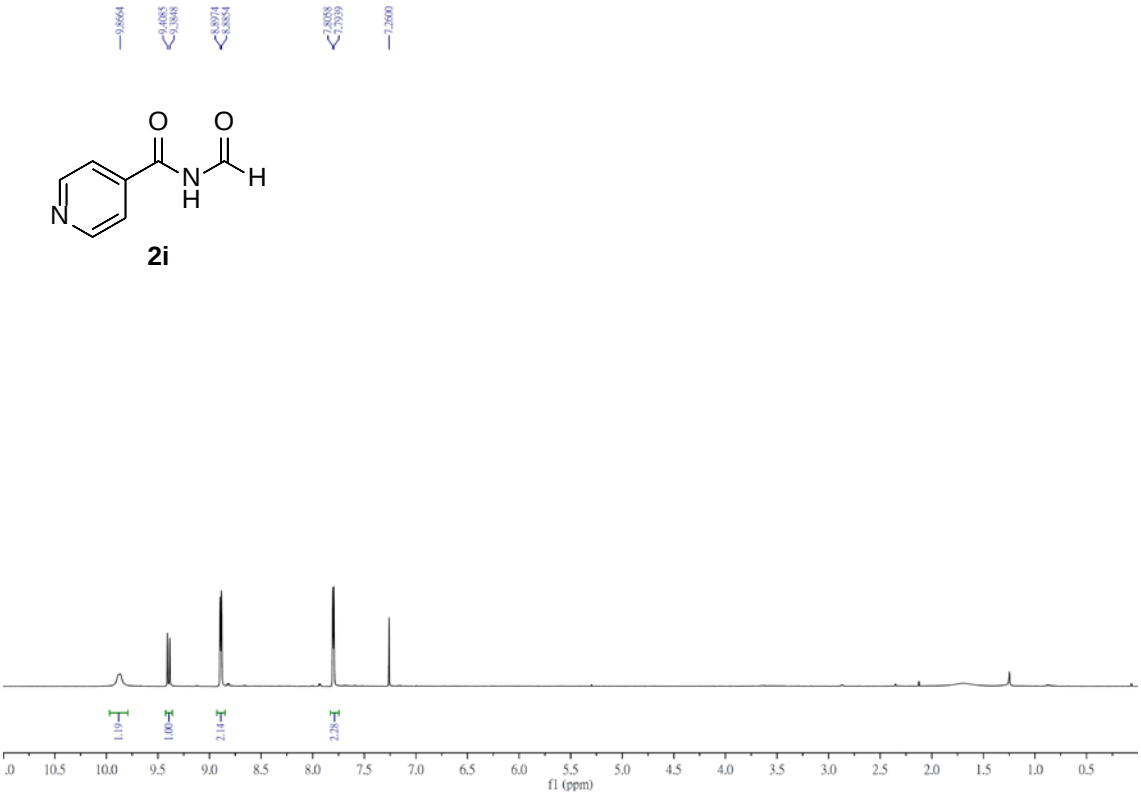
2g


164.53
160.03
136.01
134.20
131.77
128.60
77.32
77.00
76.68
O
O
N
H
H
S
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

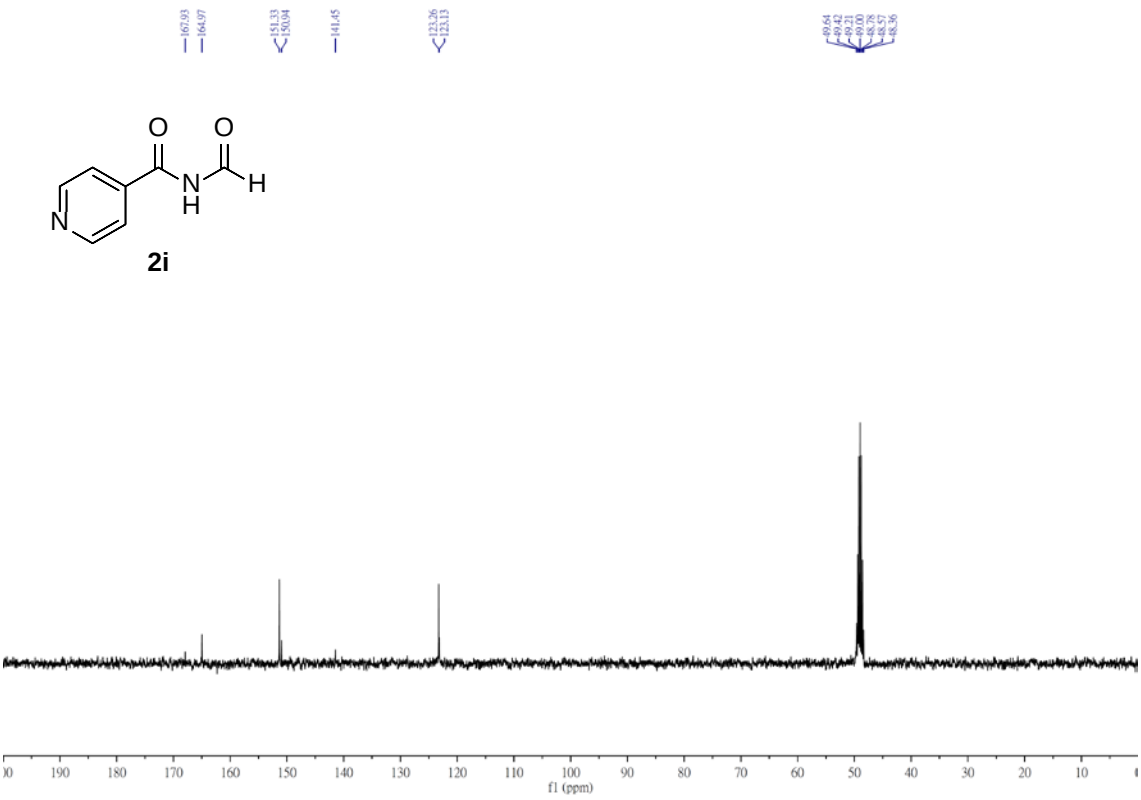
2g


9.2873
9.2623
8.5625
7.8016
7.7824
7.5865
7.5669
7.4339
7.4147
7.4138
7.4044
7.2598
6.4976
6.4784
1.5775
O
O
N
H
H
2h
0.91
1.33
1.02
2.00
2.86
1.10
f1 (ppm)

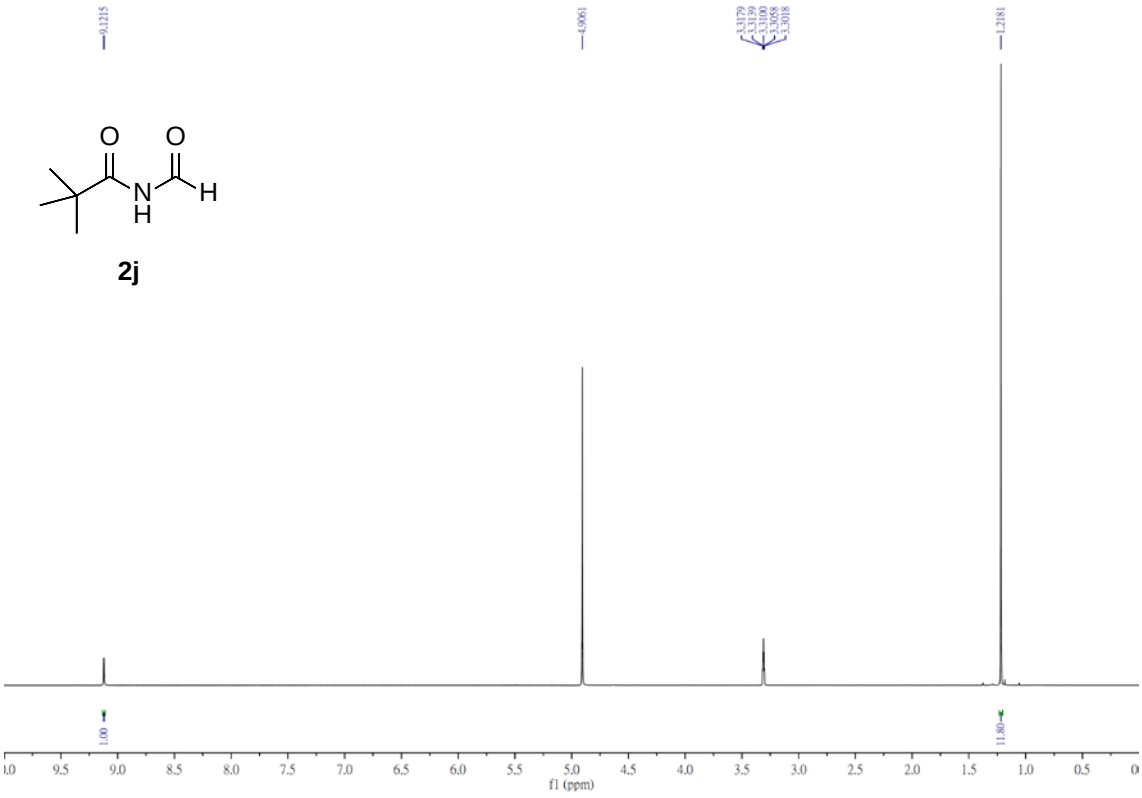

165.47
164.50
146.51
133.63
131.17
129.03
128.54
117.95
77.32
77.00
76.68
O
O
N
H
H
2h
f1 (ppm)


9.8664
9.4085
9.3848
8.8974
8.8854
7.8058
7.7939
7.2600
O
O
N
H
H
N
1.19
1.00
2.14
2.28
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

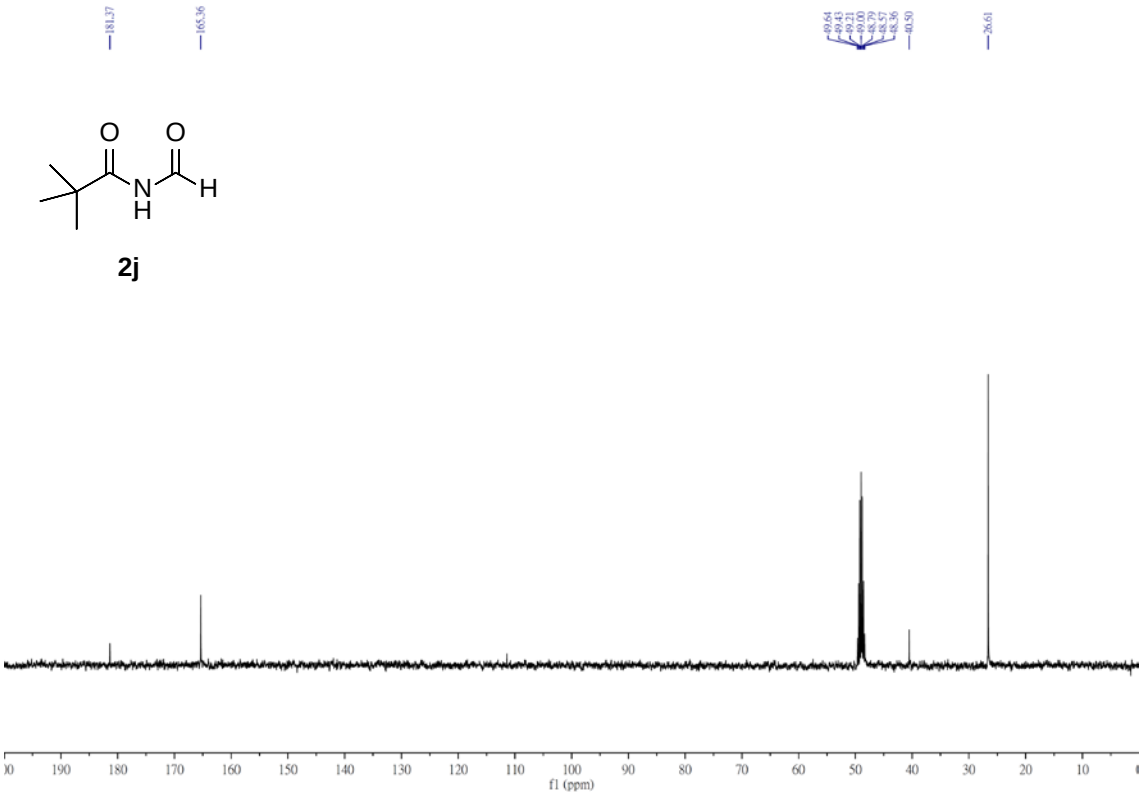
2i


167.93
164.97
151.33
150.04
141.45
123.26
123.13
49.64
49.42
49.21
49.00
48.78
48.57
48.36
O
O
N
H
H
N
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

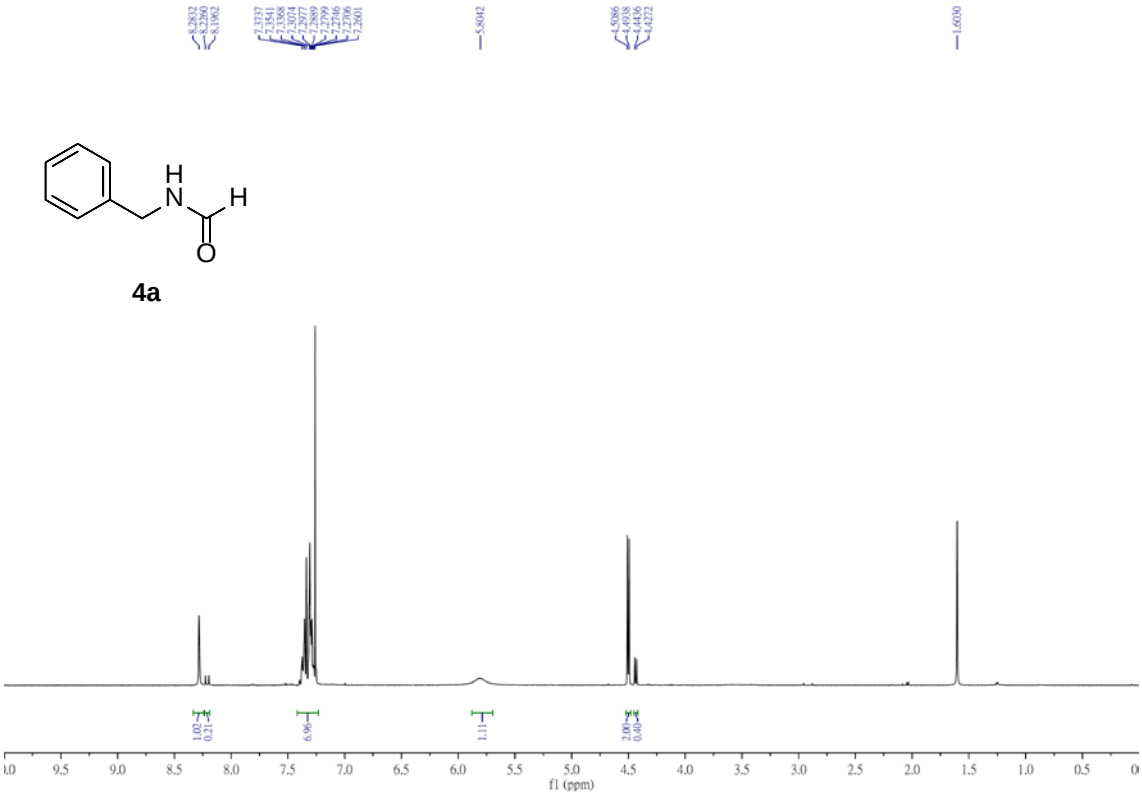
2i


9.1215
4.9061
3.3179
3.3139
3.3100
3.3058
3.3018
1.2181
O
O
N
H
H
1.00
11.80
f1 (ppm)

2j

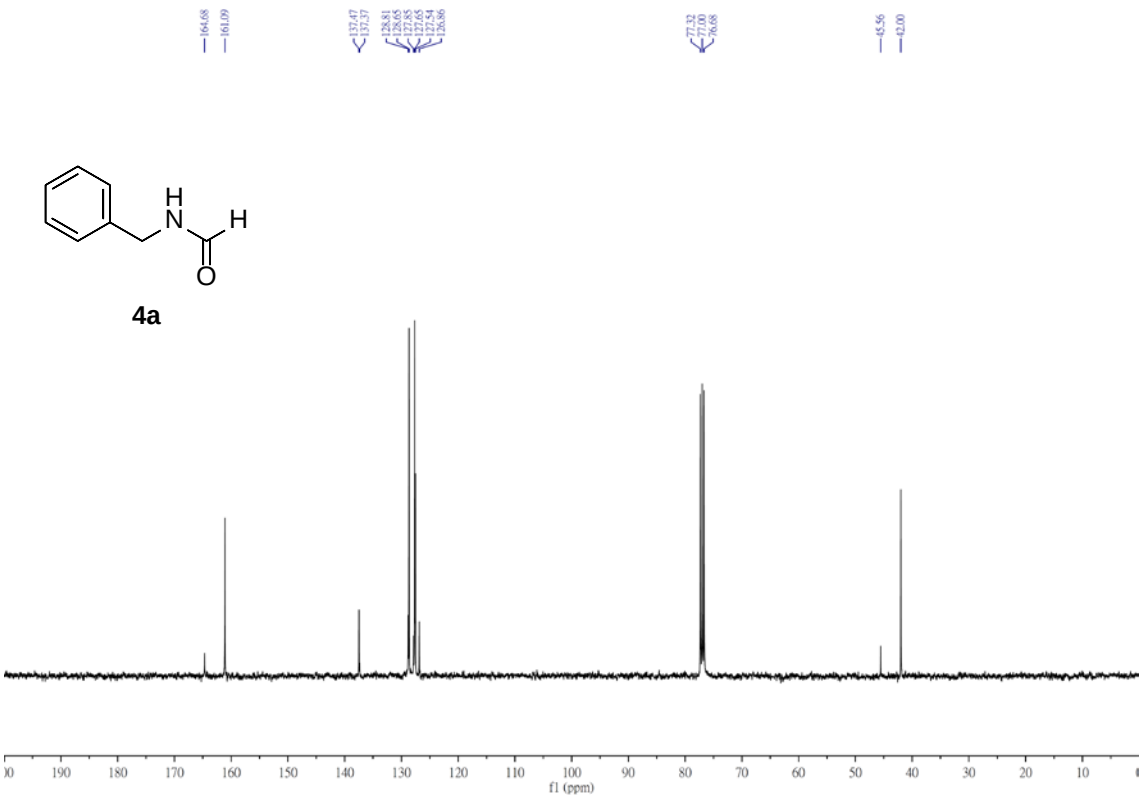

181.37
165.36
49.64
49.43
49.21
49.00
48.79
48.57
48.36
40.50
26.61
O
O
N
H
H
f1 (ppm)

2j


8.2832
8.2260
8.1962
7.3737
7.3541
7.3388
7.3074
7.2977
7.2889
7.2799
7.2706
7.2601
5.8042
4.5086
4.4938
4.4436
4.4272
1.6030
1.02
0.21
6.96
1.11
2.00
0.40
f1 (ppm)

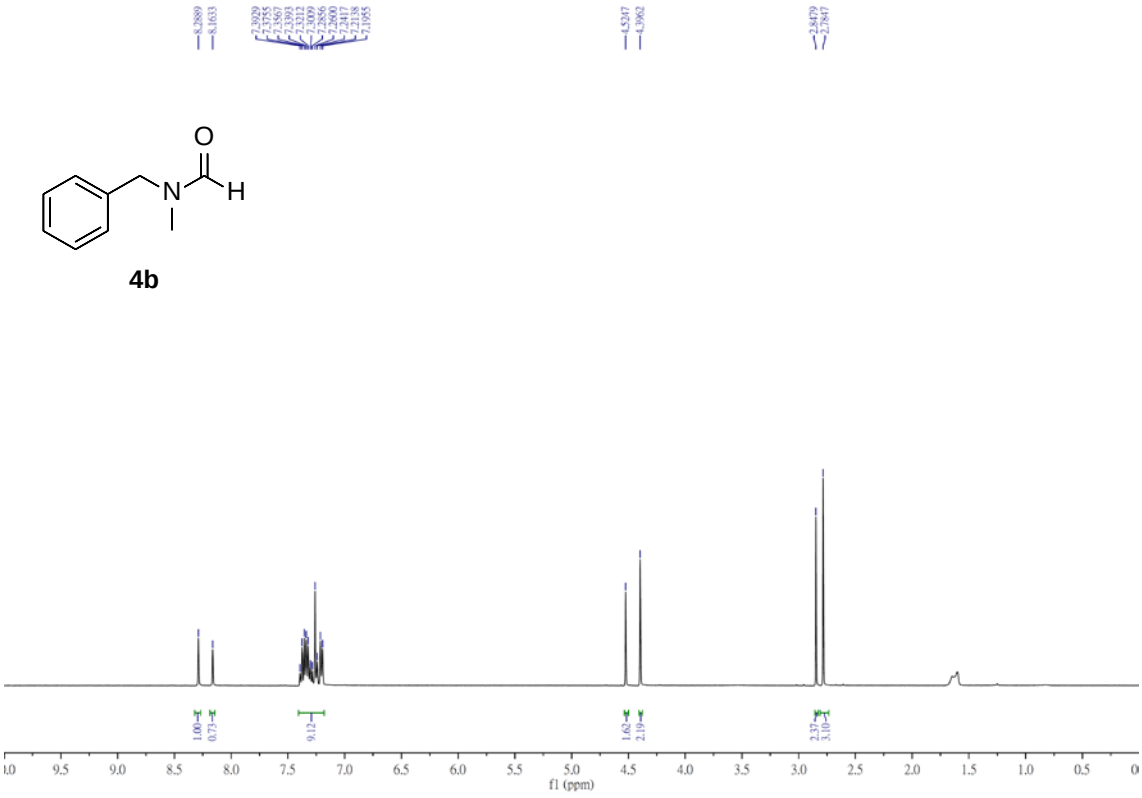
H
N
H
O

4a

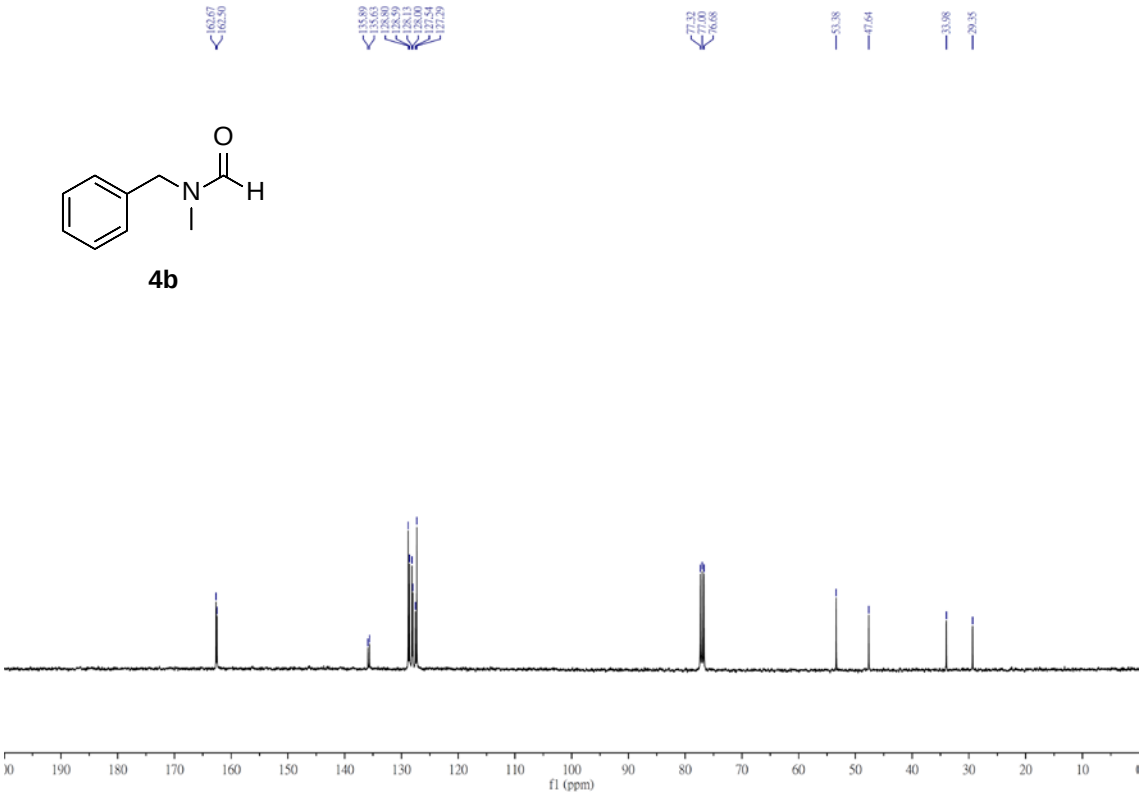

164.68
161.09
137.47
137.37
128.81
128.65
128.15
127.65
127.54
126.86
77.32
77.00
76.68
45.56
42.00
f1 (ppm)

H
N
H
O

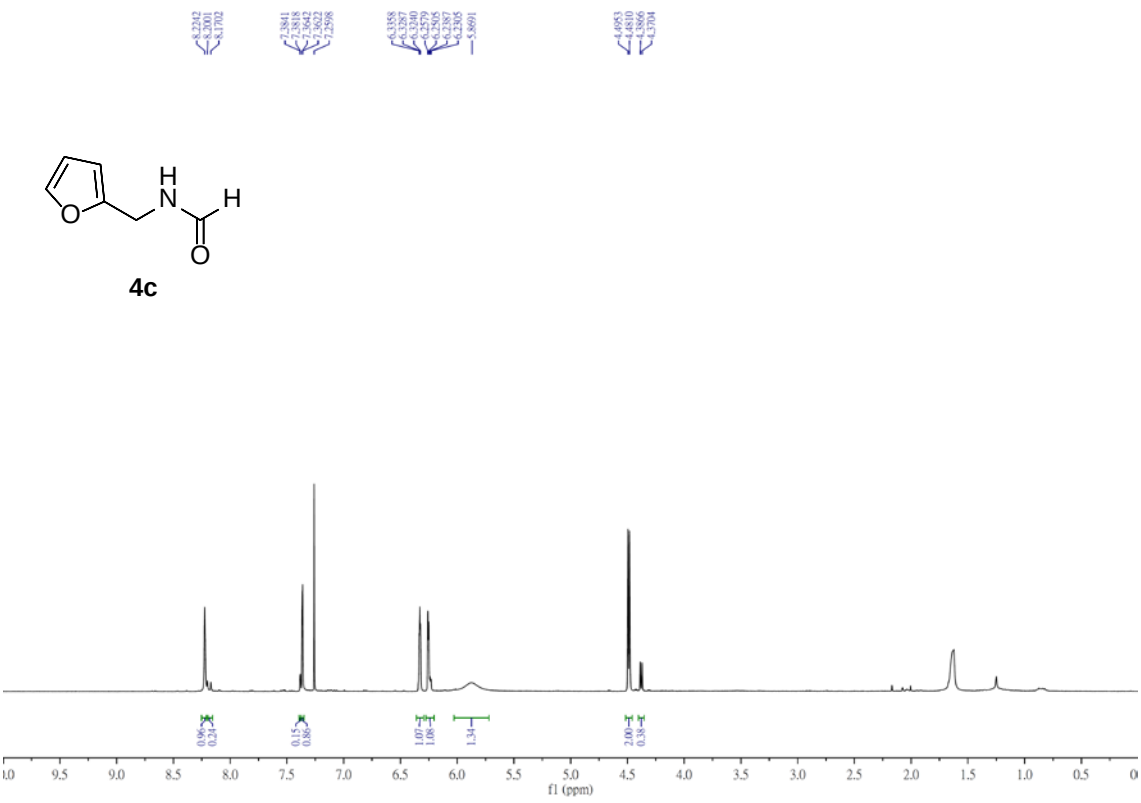
4a


8.2889
8.1633
7.3025
7.2795
7.2567
7.2393
7.2312
7.2209
7.2096
7.2000
7.2417
7.2138
7.1955
4.5347
4.3962
2.8479
2.7847
O
N
H
1.00
0.73
9.12
1.62
2.19
2.37
3.10
f1 (ppm)

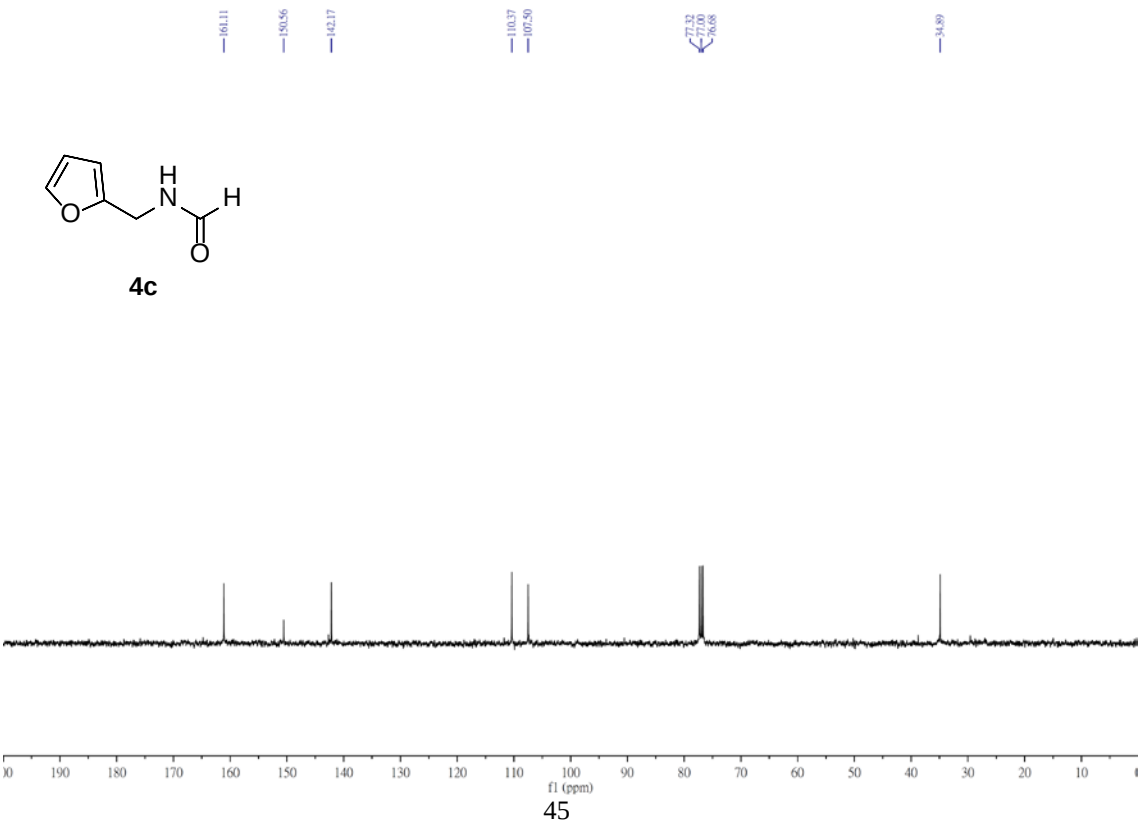
4b


162.67
162.50
135.89
135.63
128.90
128.59
128.31
128.00
127.54
127.29
77.32
77.00
76.68
53.38
47.64
33.98
29.35
O
N
H
f1 (ppm)

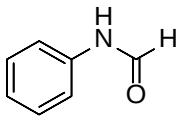
4b


8.3242
8.3201
8.1702
7.3841
7.3818
7.3642
7.2622
7.2598
6.3358
6.3287
6.3240
6.2579
6.2496
6.2287
6.2305
5.8691
4.0953
4.4810
4.3866
4.3704
0.96
0.24
0.15
0.86
1.07
1.08
1.34
2.00
0.38
f1 (ppm)

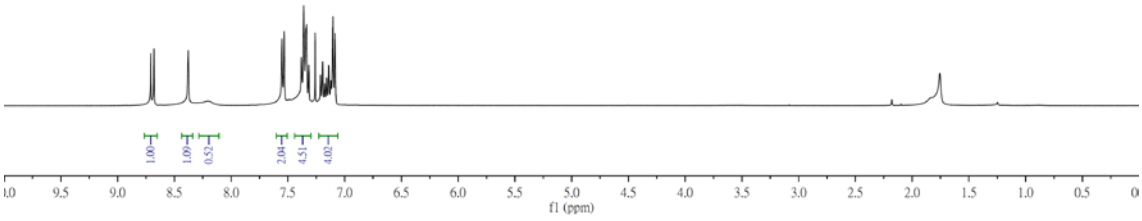
4c

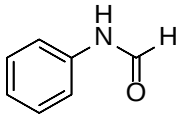

161.11
150.56
142.17
110.37
107.50
77.32
77.00
76.68
34.89
f1 (ppm)

4c

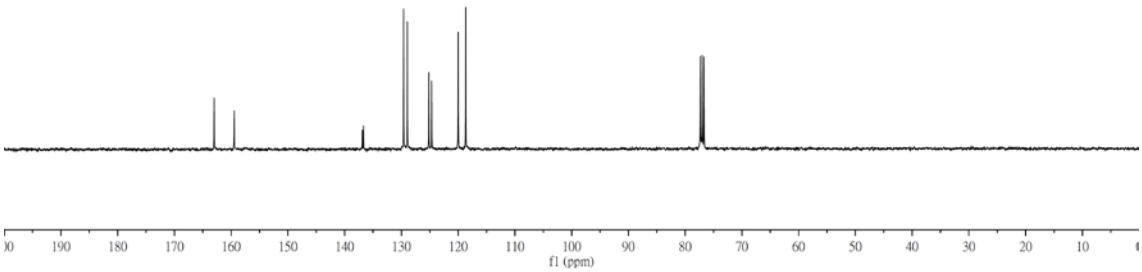


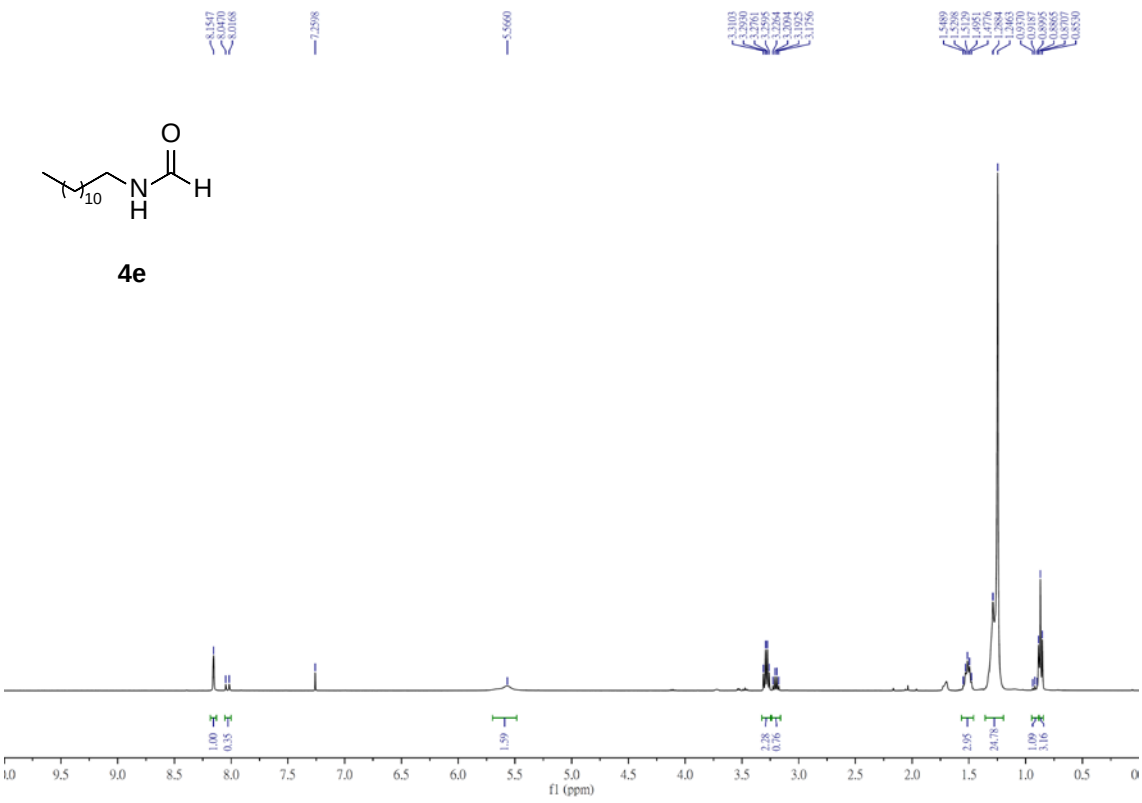
4d



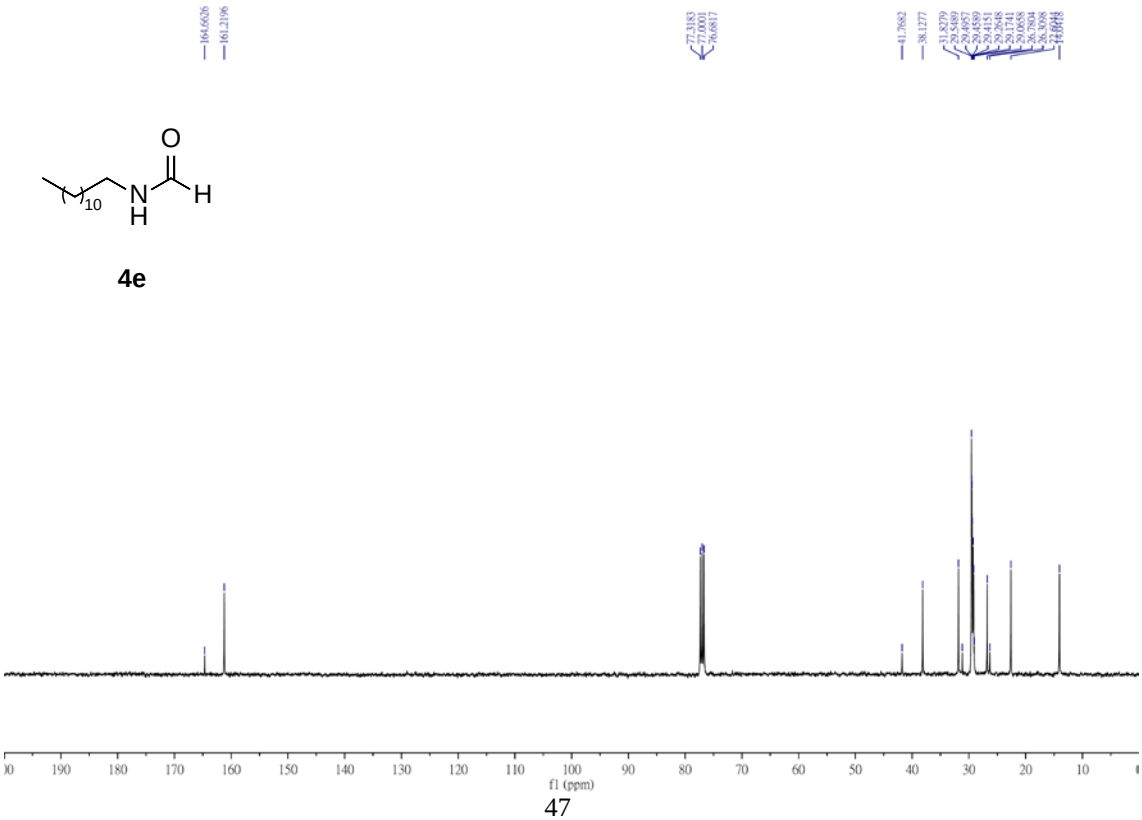


4d

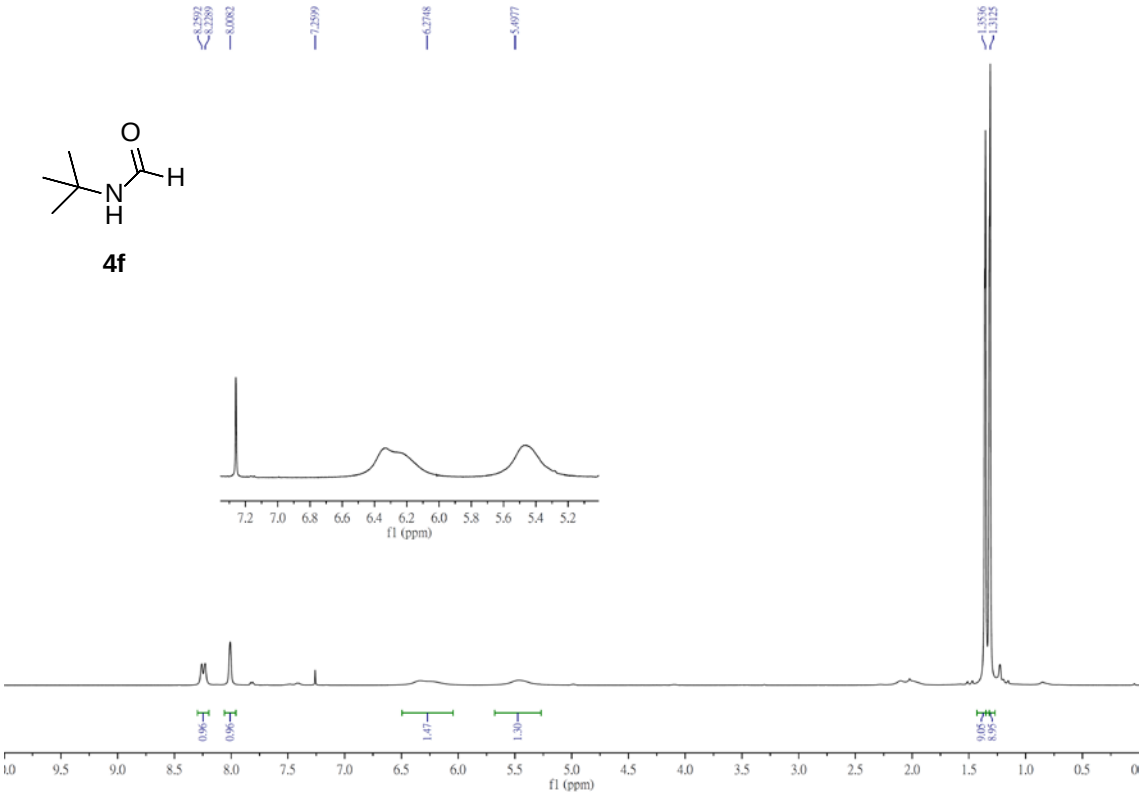




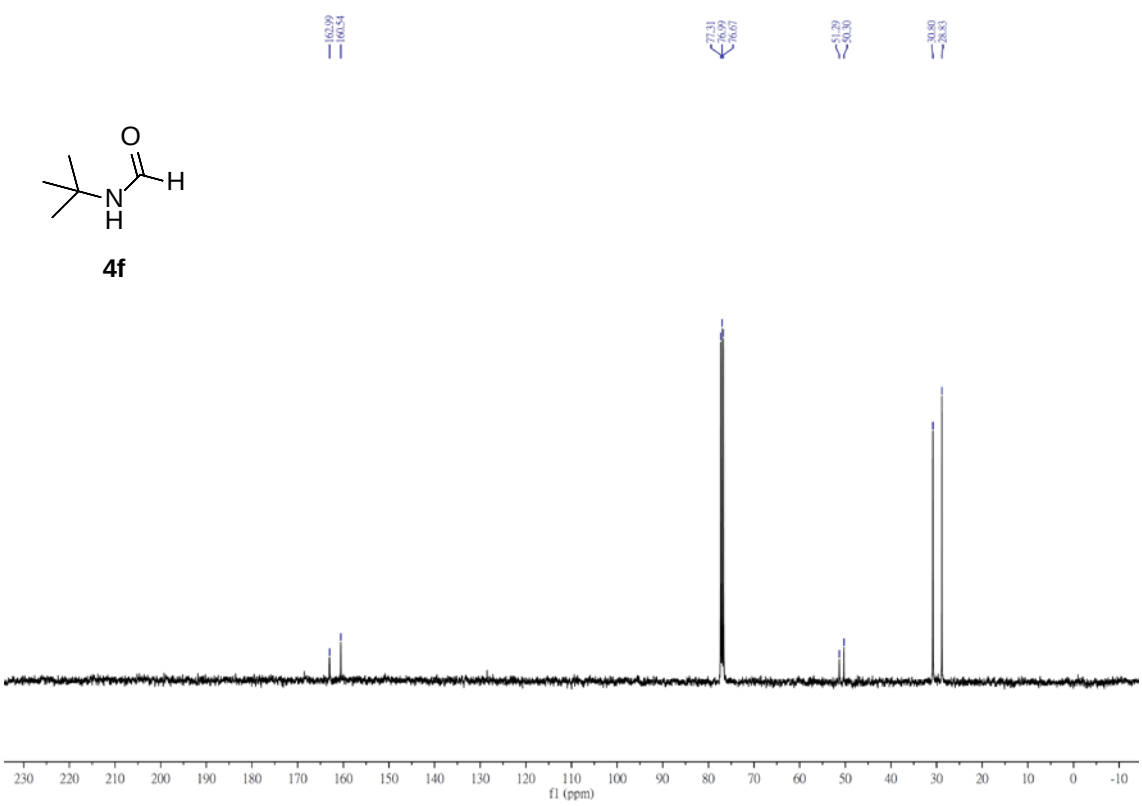
4e



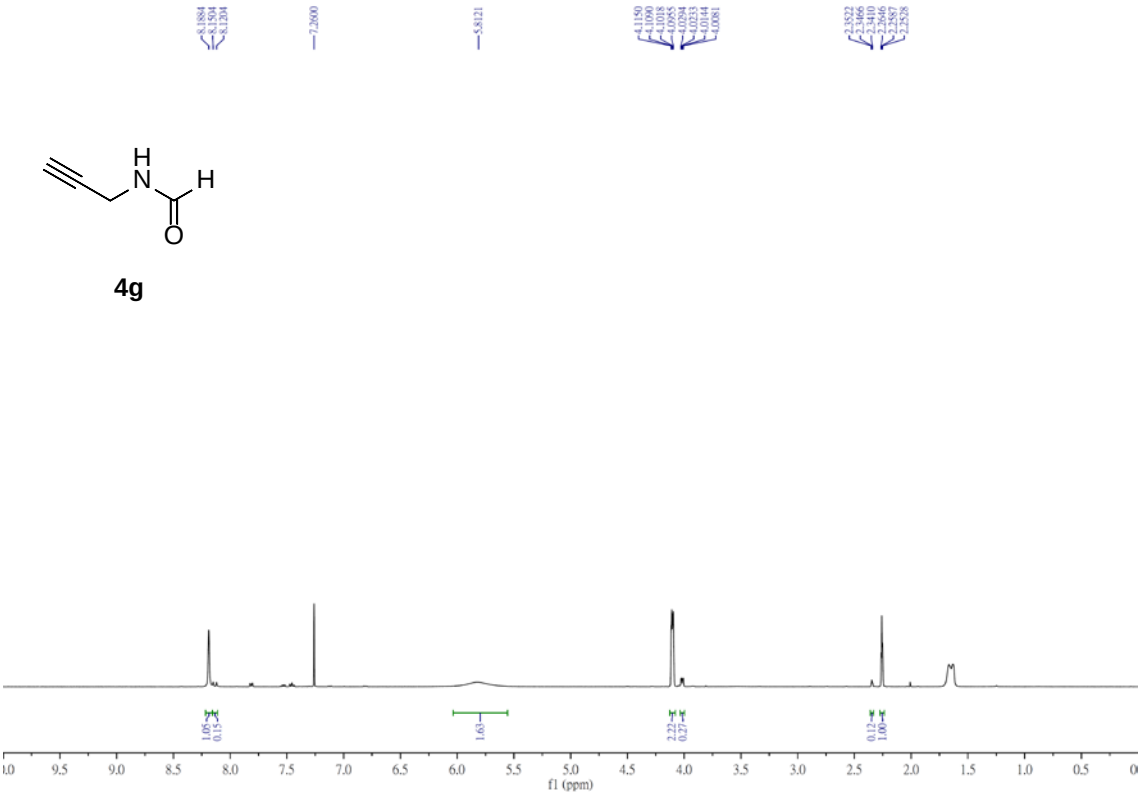
4e


8.2592
8.2269
8.0082
7.2599
6.3748
5.4977
1.3536
1.3125
7.2 7.0 6.8 6.6 6.4 6.2 6.0 5.8 5.6 5.4 5.2
f1 (ppm)
0.96
0.96
1.47
1.30
9.05
8.95
f1 (ppm)

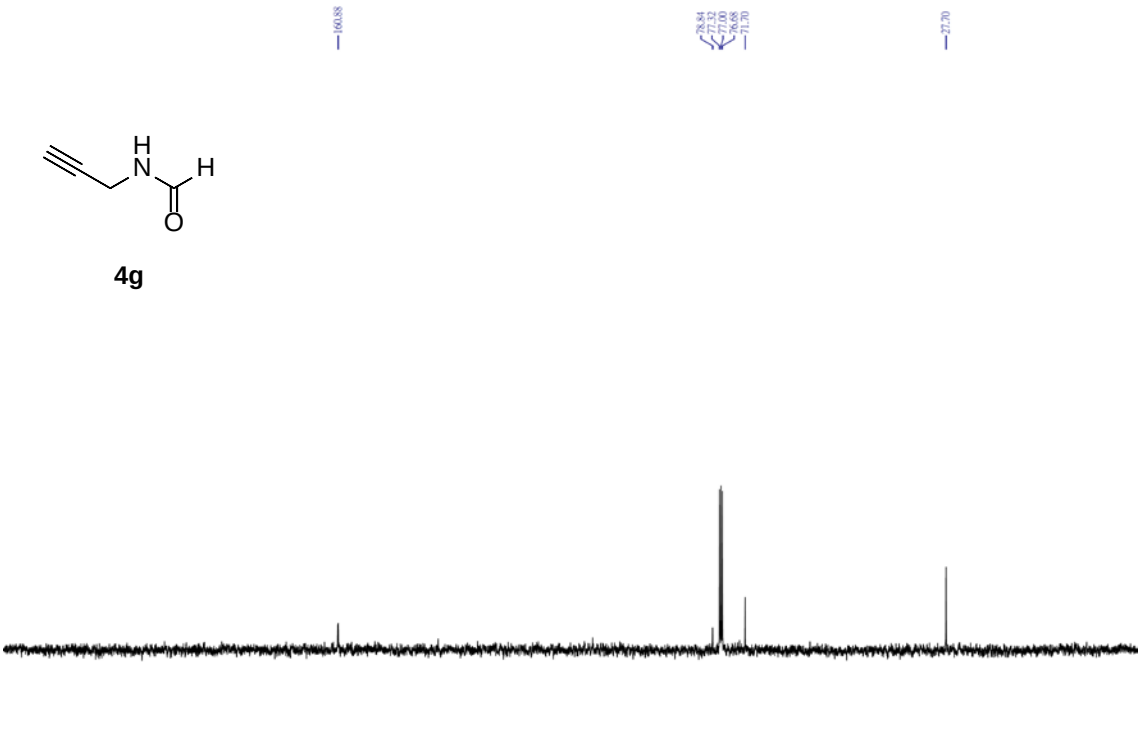
4f


162.99
160.54
77.31
76.99
76.67
51.29
50.30
30.80
28.83
f1 (ppm)

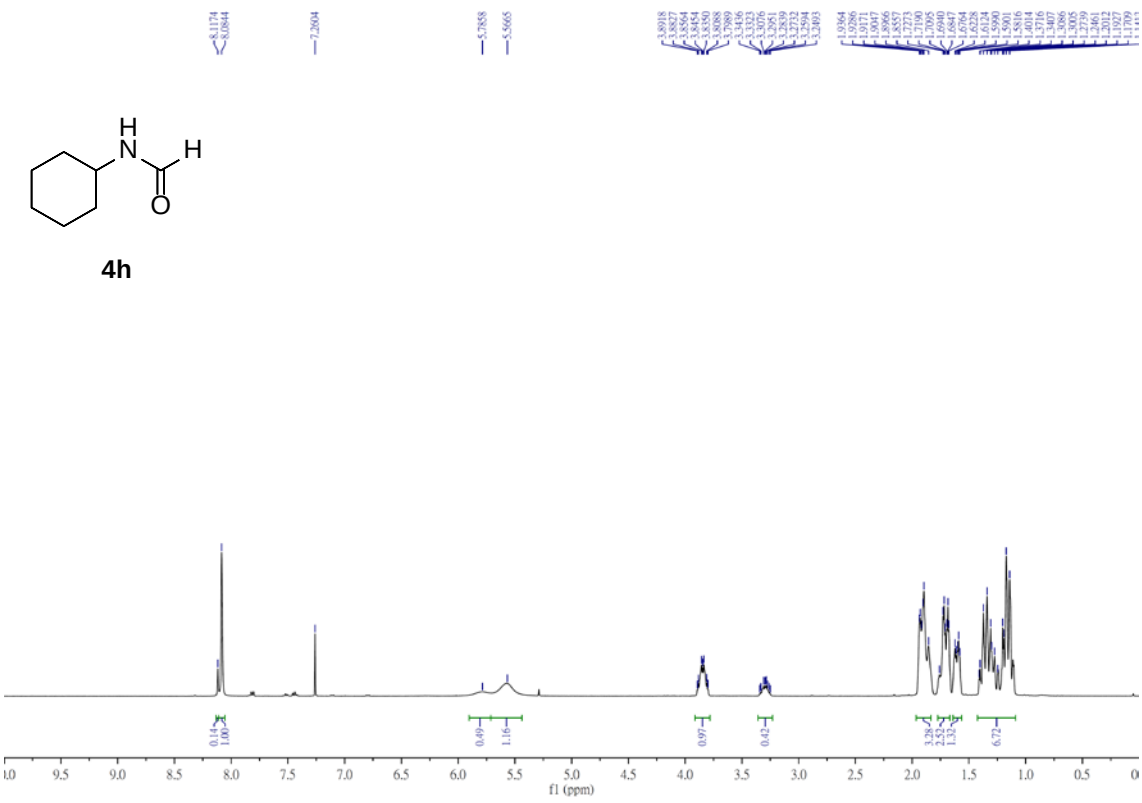
4f



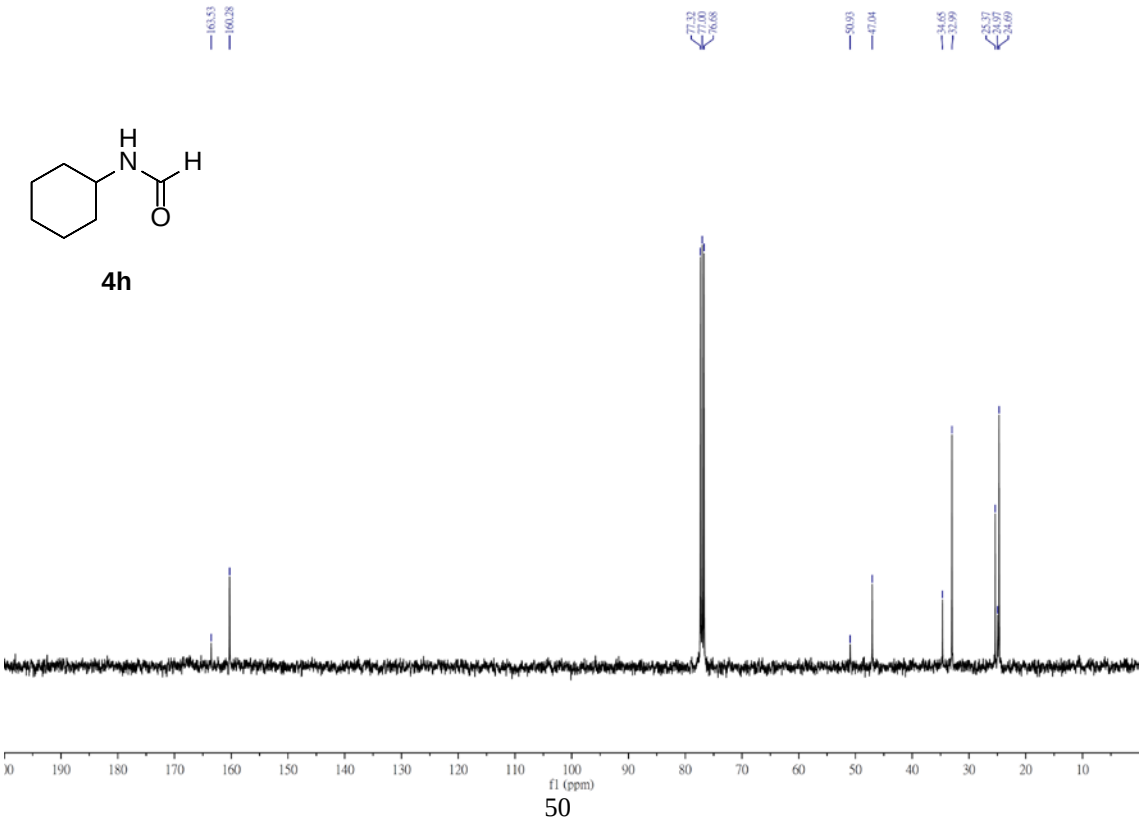
4g



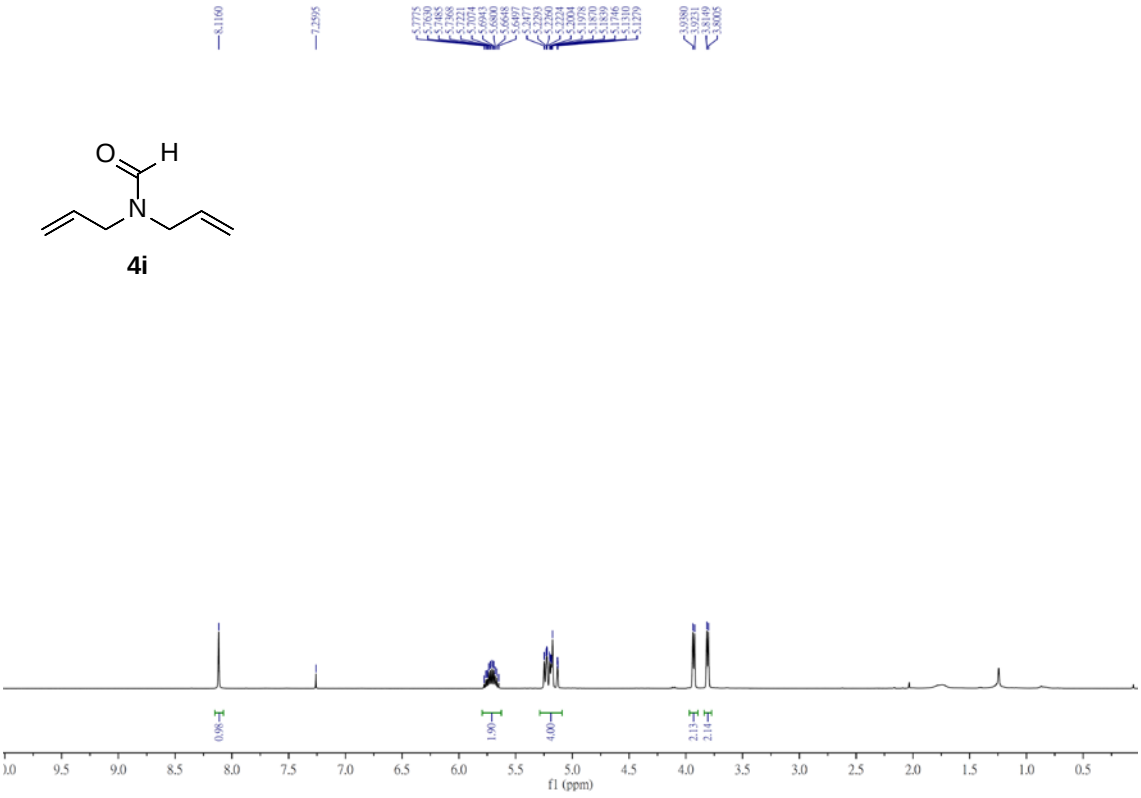
4g



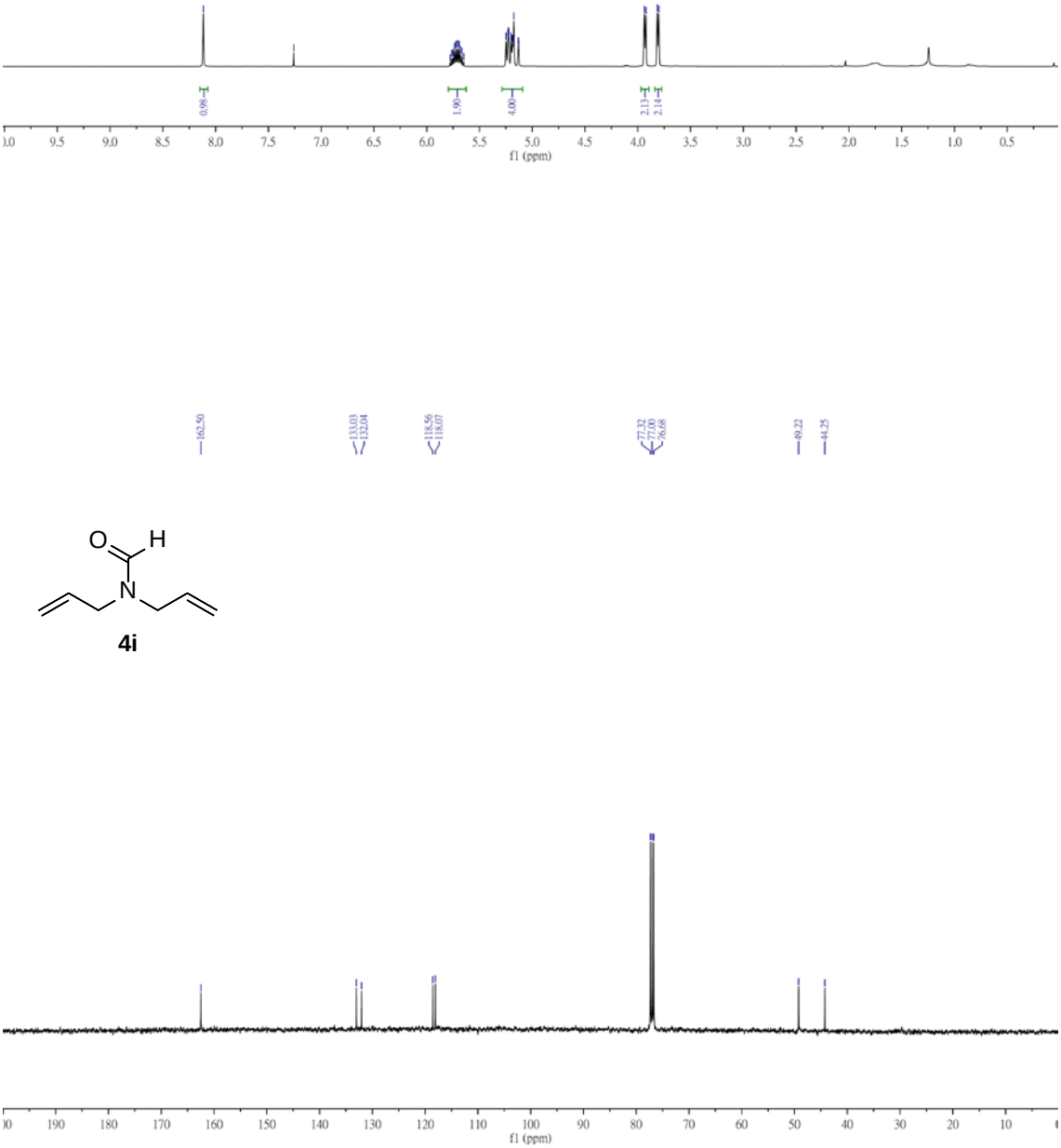
4h



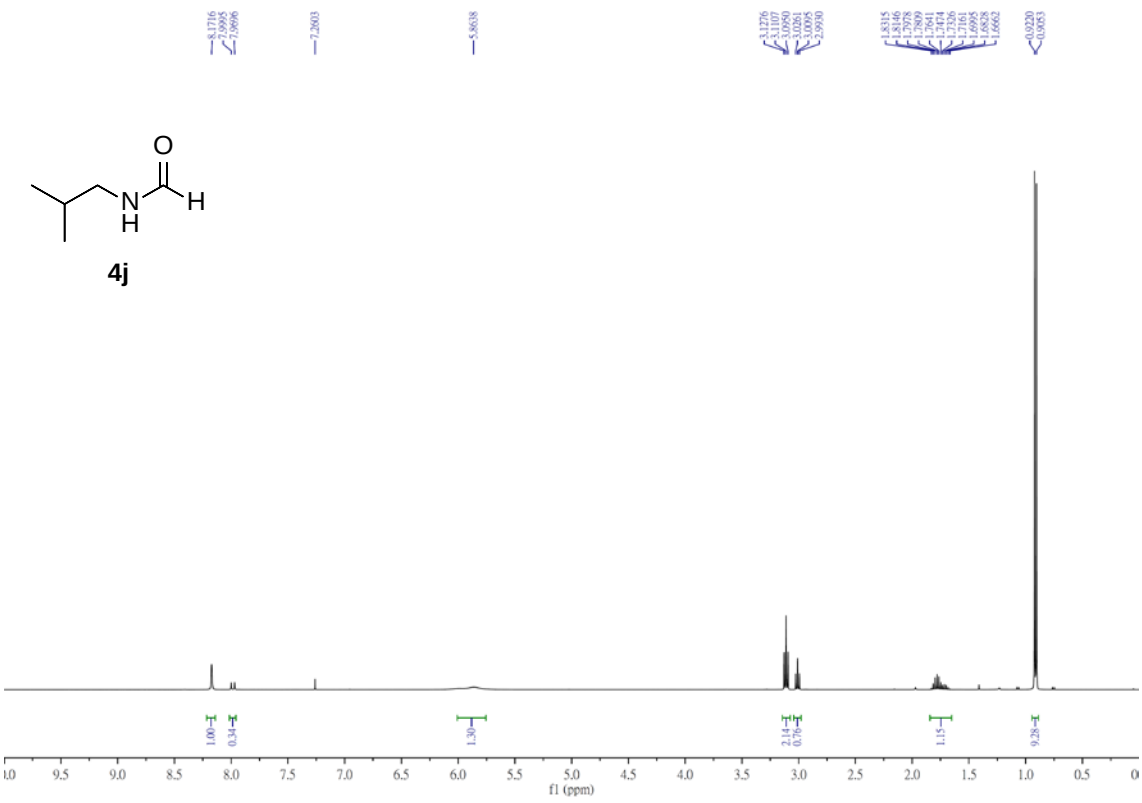
4h


8.1160
7.2595
5.7775
5.7630
5.7485
5.7336
5.7191
5.7074
5.6943
5.6800
5.6648
5.6497
5.3477
5.3293
5.3260
5.3224
5.3004
5.1978
5.1870
5.1759
5.1706
5.1330
5.1279
3.9380
3.9231
3.8149
3.8005
0.98
1.90
4.00
2.13
2.14
f1 (ppm)

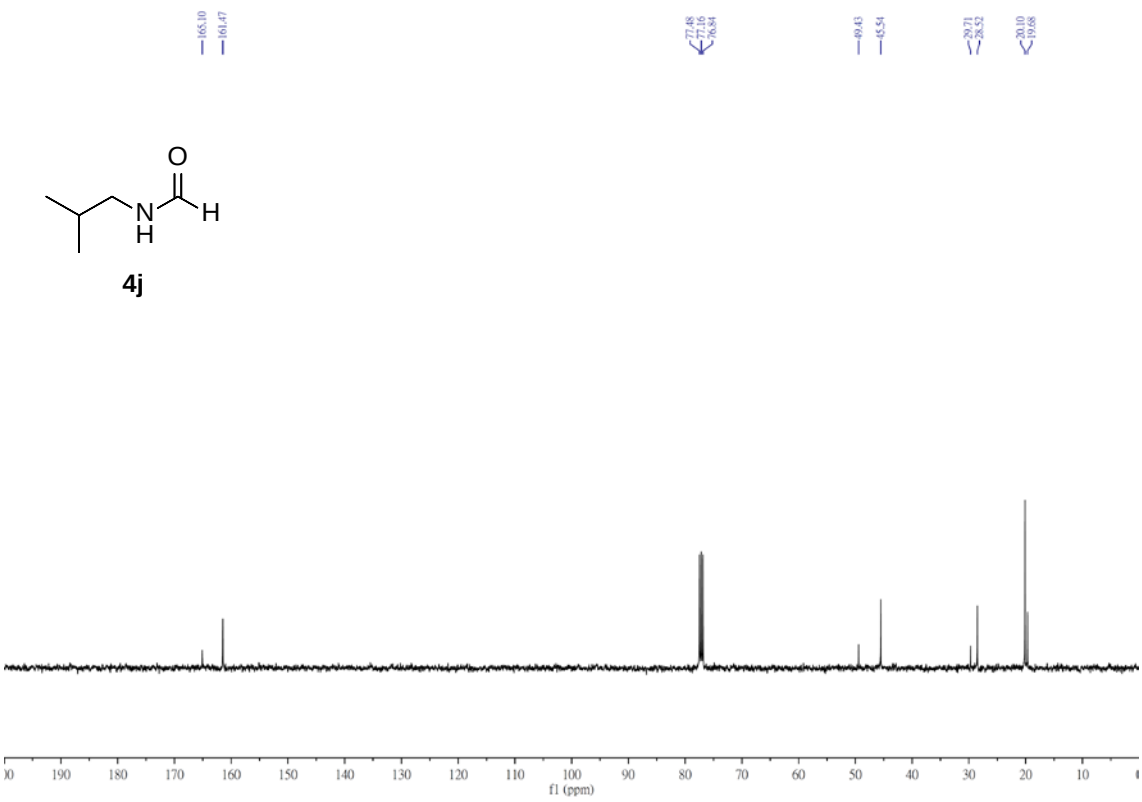
4i


162.50
133.03
132.04
118.56
118.07
77.32
77.00
76.68
49.22
44.25
f1 (ppm)

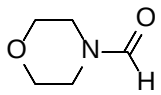
4i


8.1716
7.9995
7.9996
7.2603
5.8638
3.1276
3.1107
3.1090
3.1081
3.0095
2.9930
1.8315
1.8146
1.8146
1.7978
1.7809
1.7641
1.7474
1.7326
1.7161
1.6995
1.6828
1.6662
0.9220
10.0053
1.00
0.34
1.30
2.14
0.76
1.15
9.28
f1 (ppm)

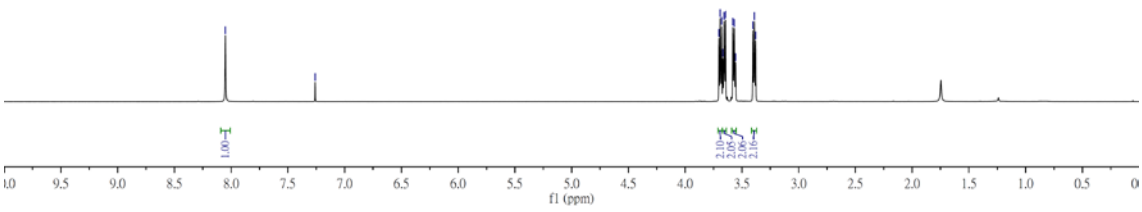
4j

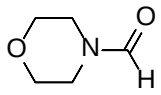

165.10
161.47
77.48
77.16
76.84
49.43
45.54
29.71
28.52
20.10
19.68
f1 (ppm)

4j

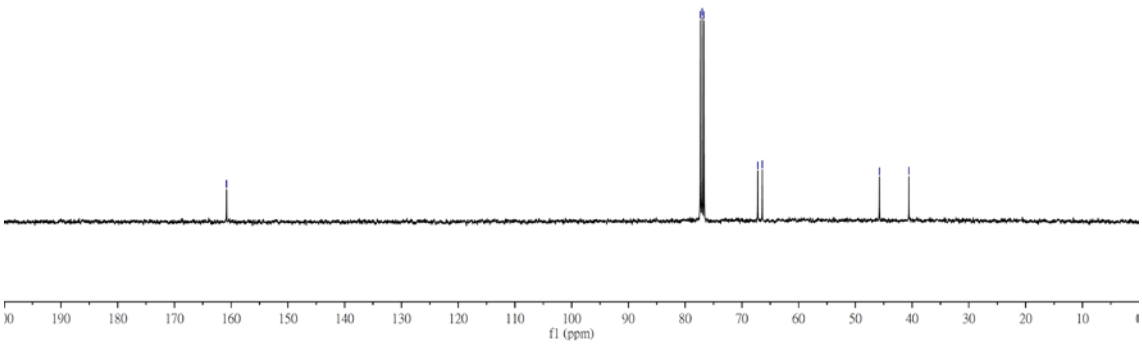


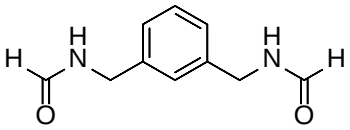
4k



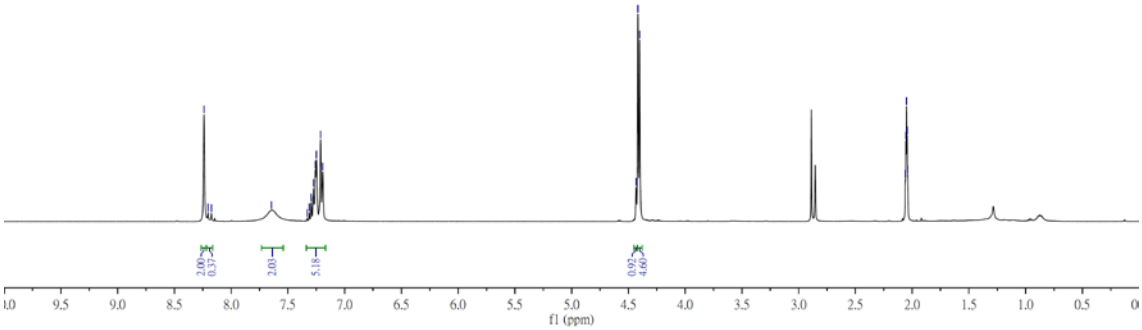


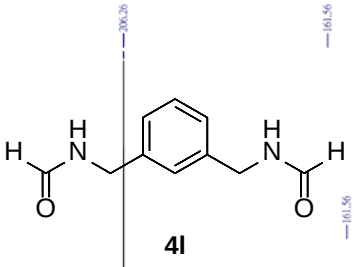
4k



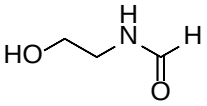


4l

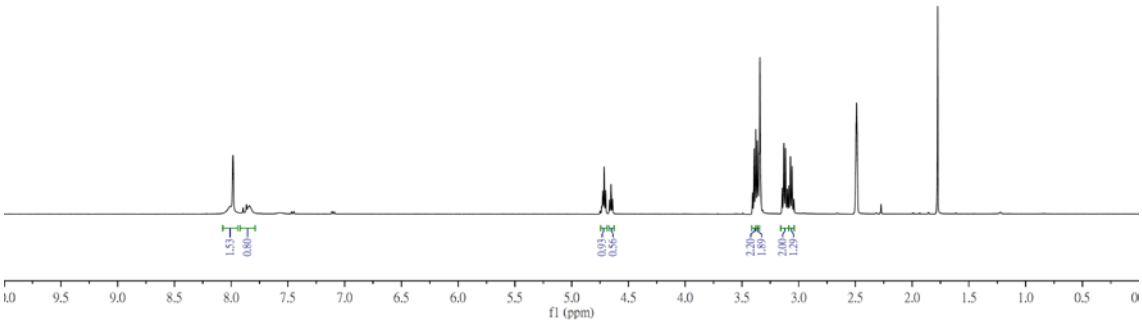


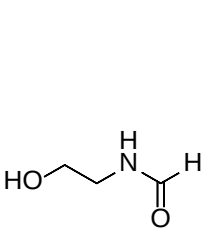


4l

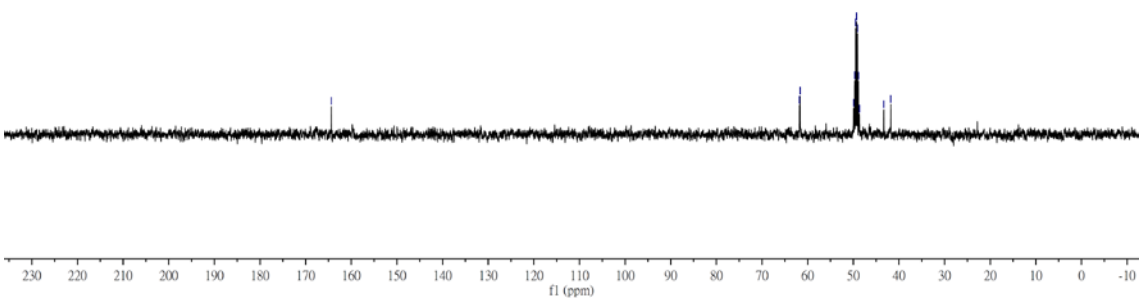


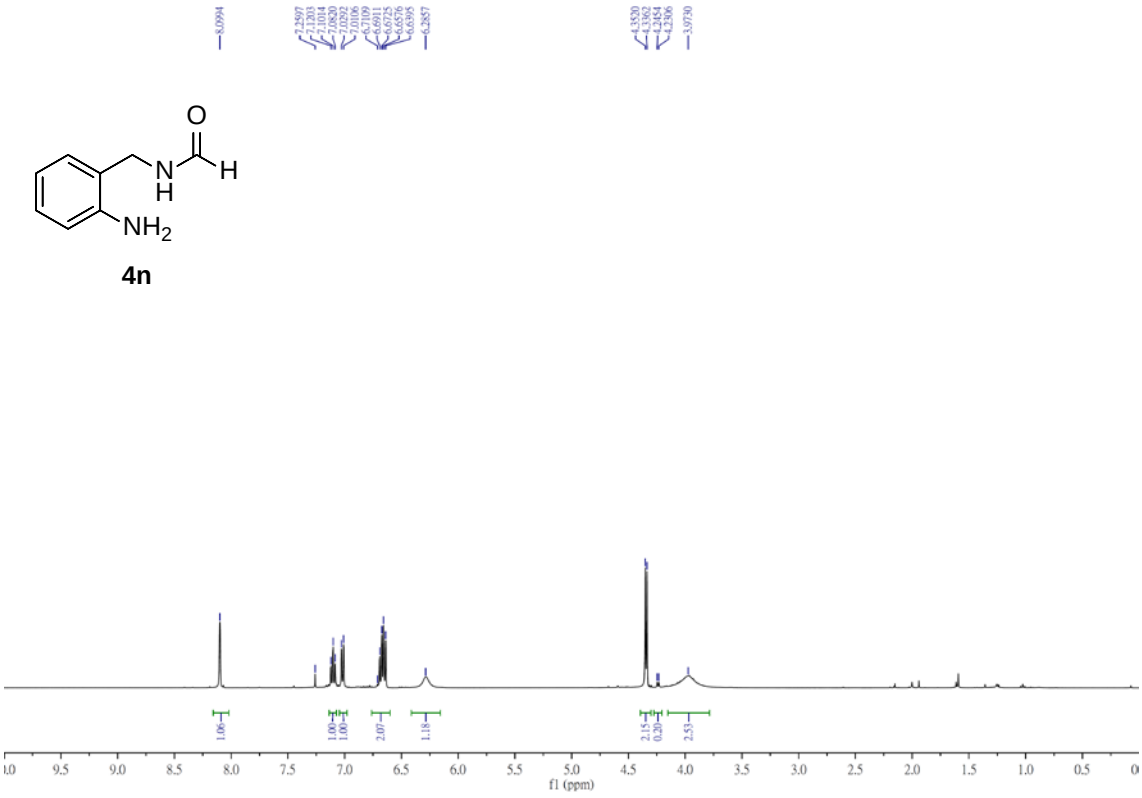
4m



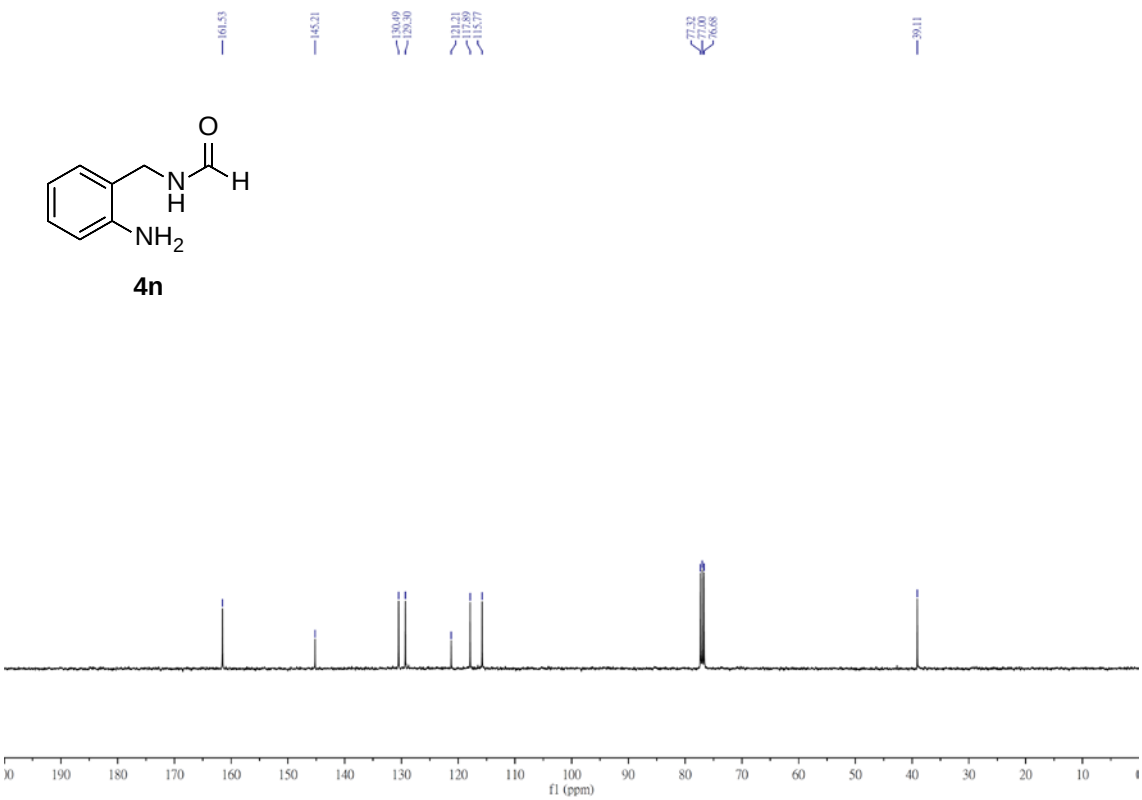


4m

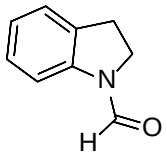



8.0994
7.2597
7.1703
7.1104
7.0820
7.0292
7.0106
7.0099
6.6911
6.6725
6.6576
6.6395
6.2857
4.3830
4.3548
4.2484
4.2306
3.9730
O
N
H
H
NH2
1.06
1.00
1.00
2.07
1.18
2.15
0.20
2.53
f1 (ppm)

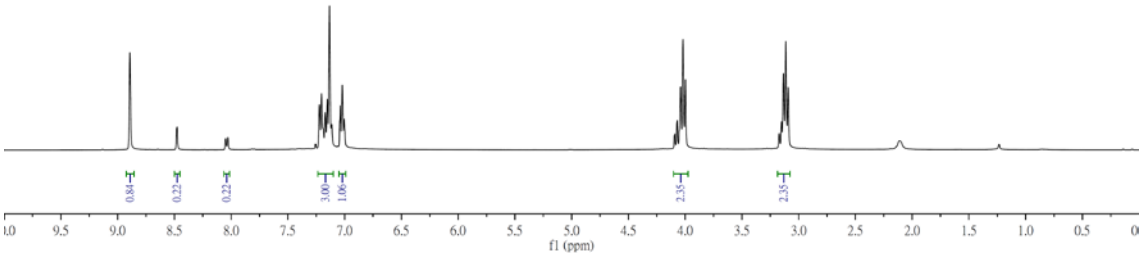
4n

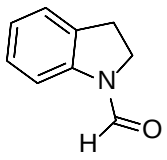

161.53
145.21
130.49
129.30
121.21
117.89
115.77
77.32
77.00
76.68
39.11
O
N
H
H
NH2
f1 (ppm)

4n

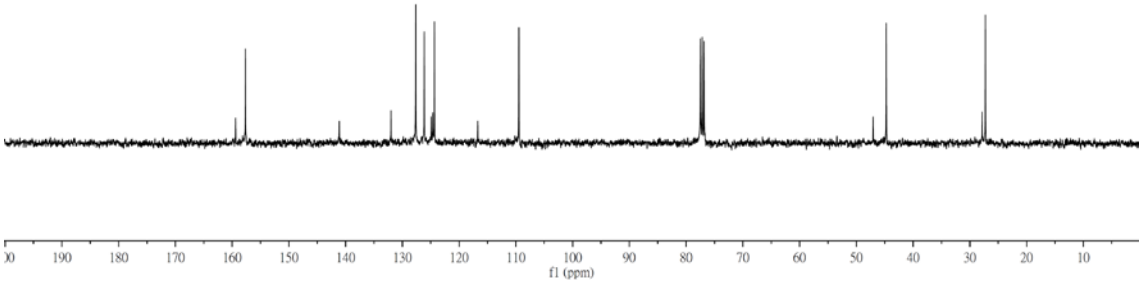


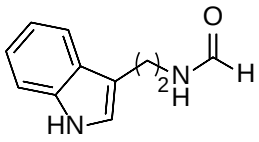
4p



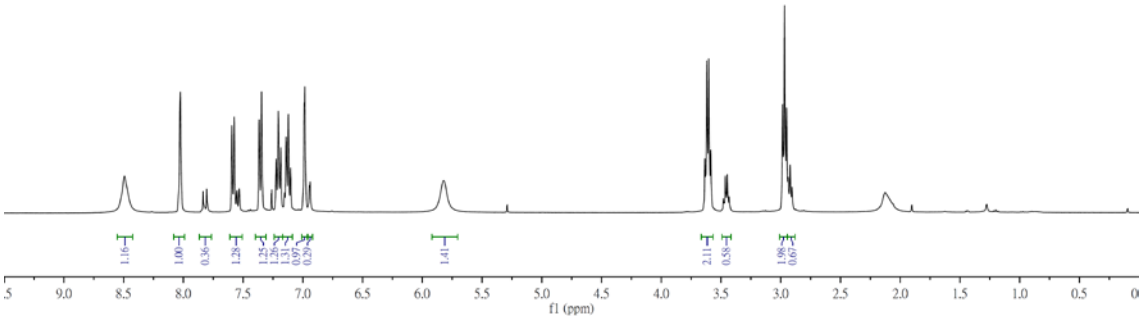


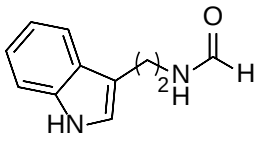
4p



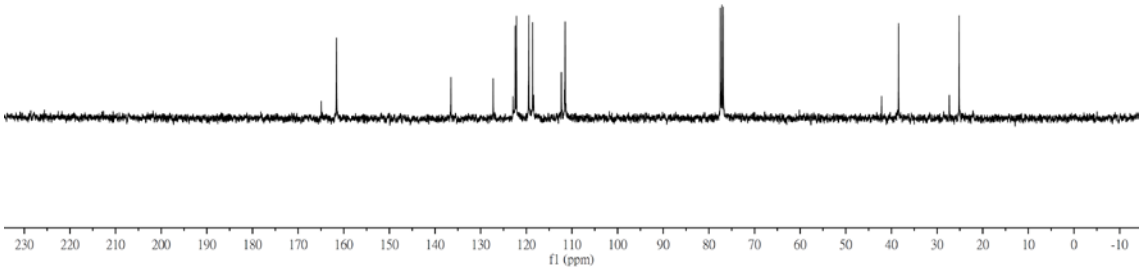


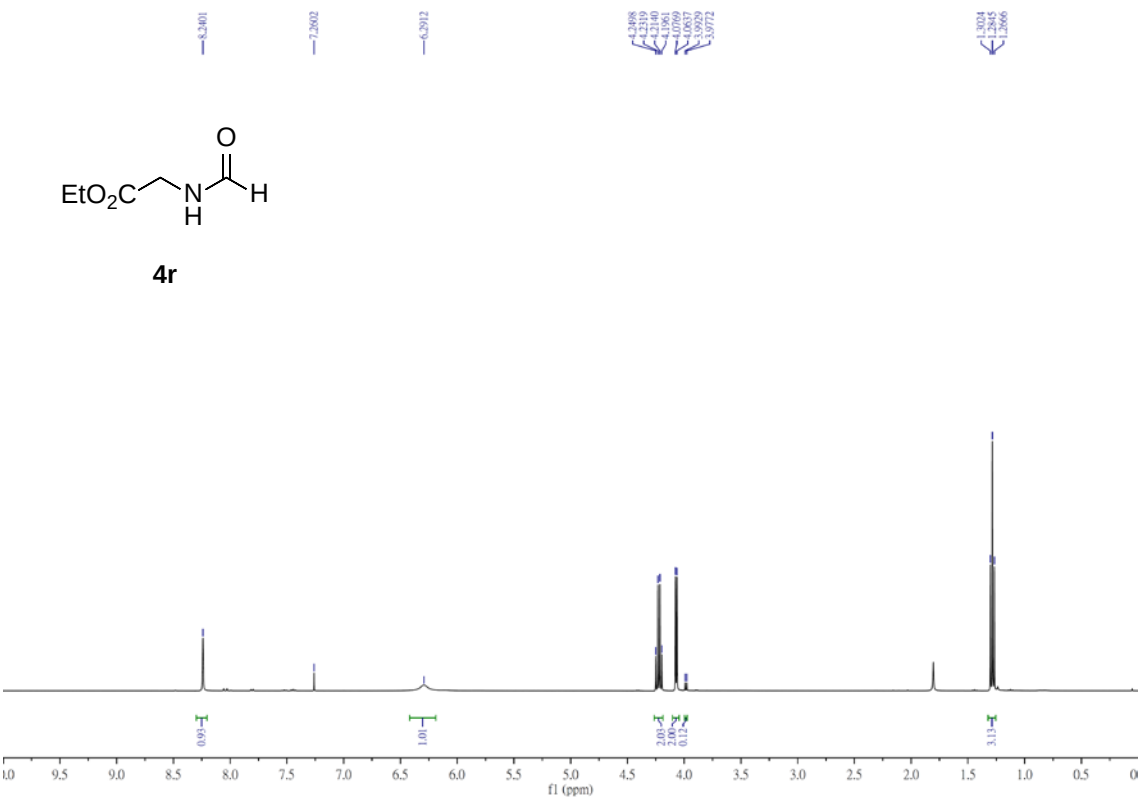
4q



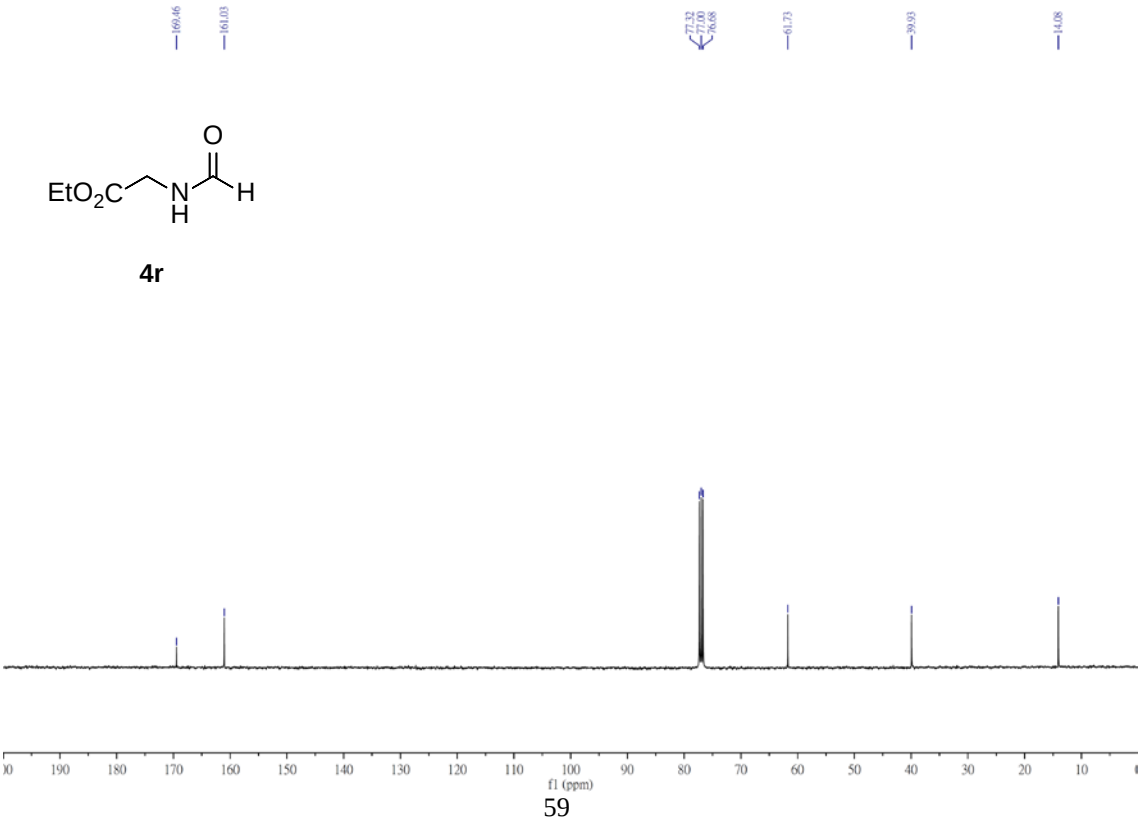


4q

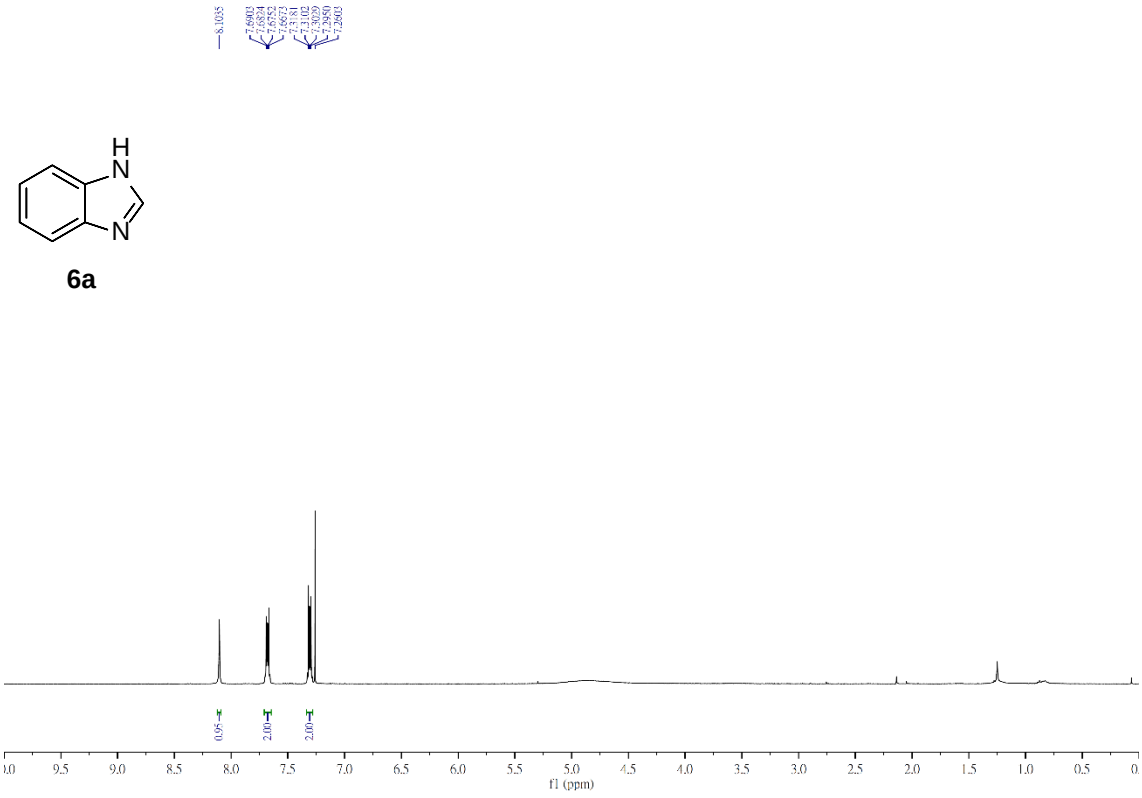



8.2401
7.2602
6.2912
4.2498
4.2339
4.2180
4.1961
4.0769
4.0597
3.9929
3.9772
1.3024
1.2845
1.2666
EtO2C
N
H
O
H
0.93
1.01
2.03
2.00
0.12
3.13
f1 (ppm)

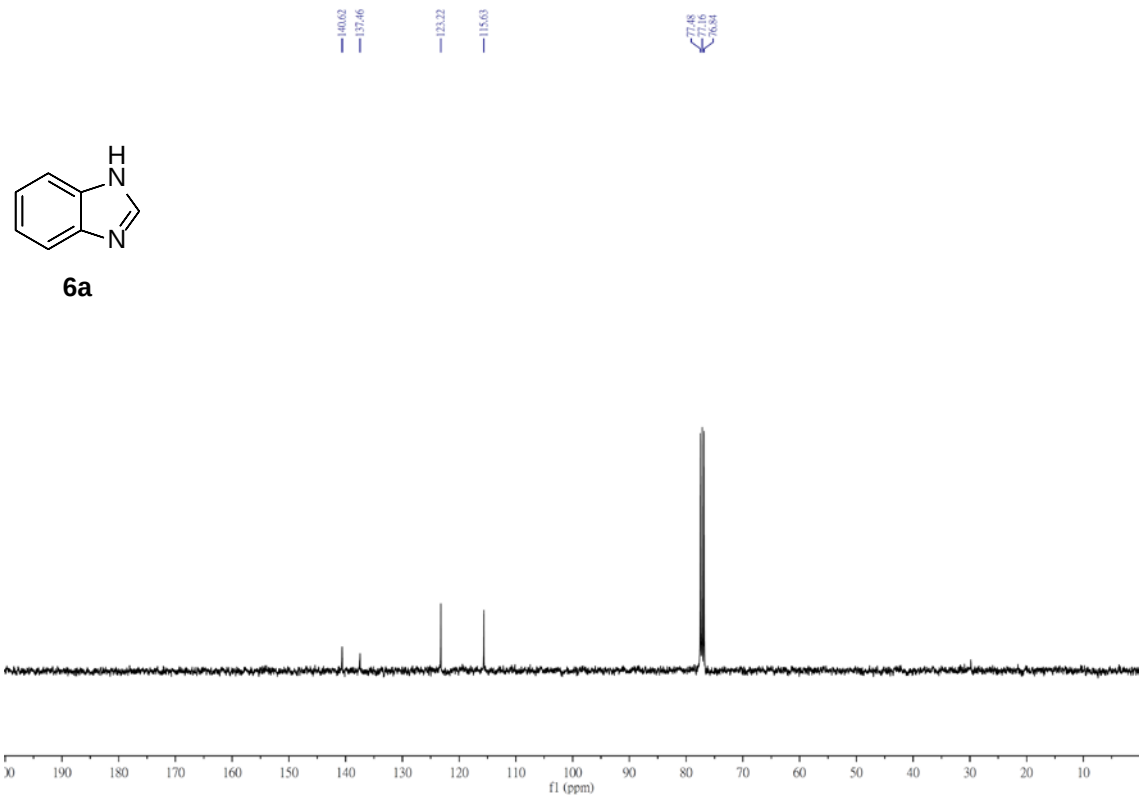
4r


169.46
161.03
77.32
77.00
76.68
61.73
39.93
14.08
EtO2C
N
H
O
H
f1 (ppm)

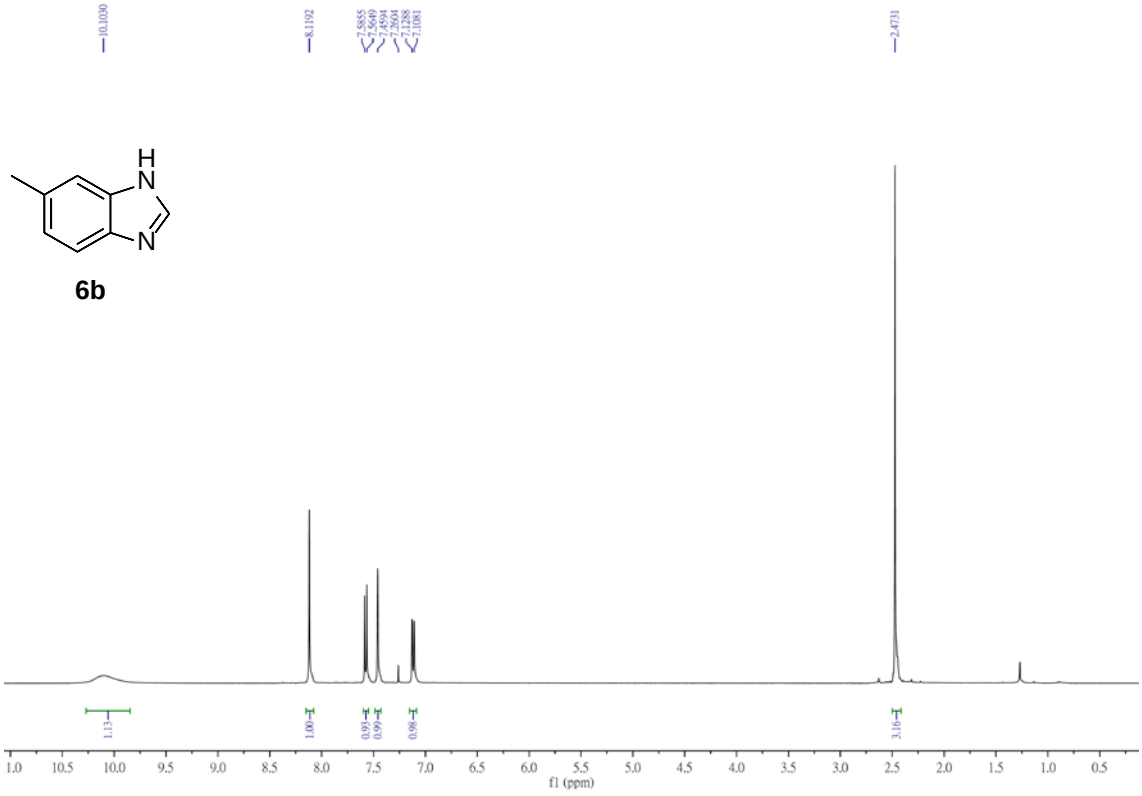
4r


8.1035
7.6903
7.6834
7.6752
7.6683
7.3153
7.3134
7.3102
7.3029
7.2490
7.2465
0.95
2.00
2.00
f1 (ppm)

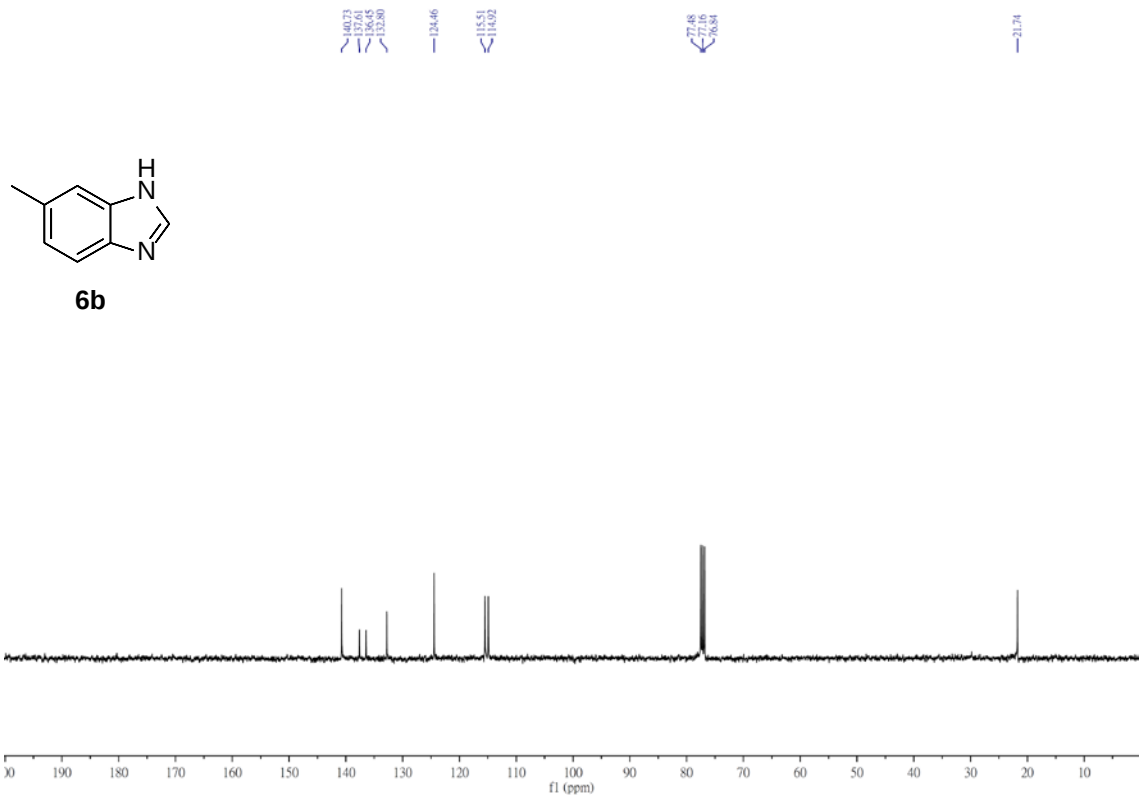
6a


140.62
137.46
123.22
115.63
77.48
77.16
76.84
f1 (ppm)

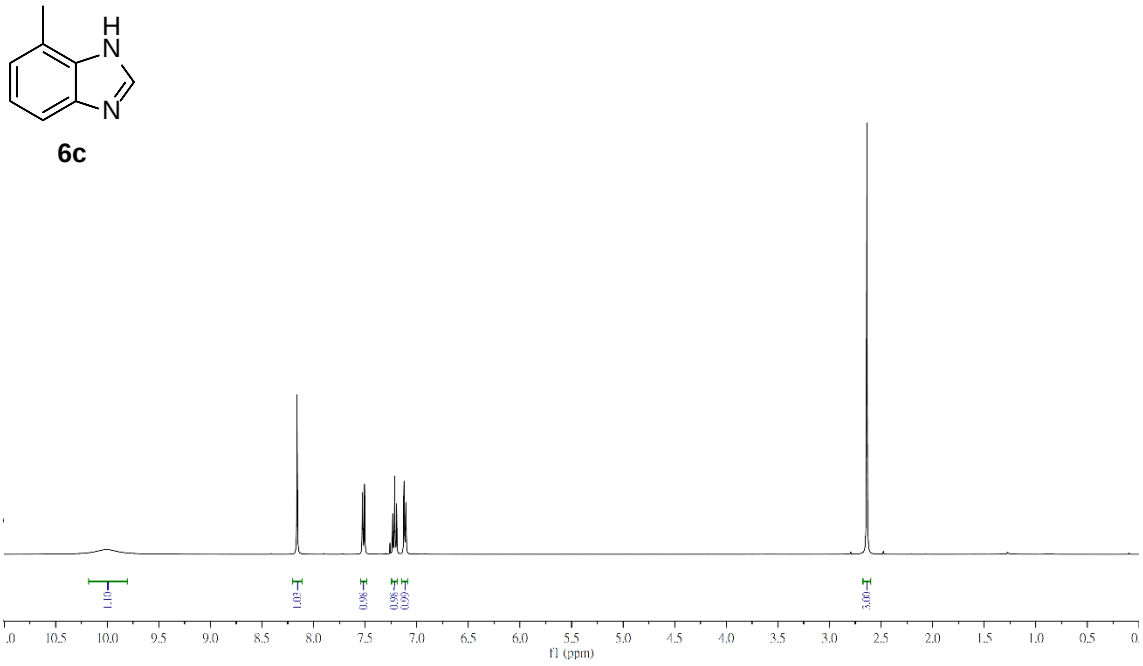
6a


10.1030
8.1192
7.5855
7.5649
7.4594
7.4364
7.1268
7.1081
2.4731
1.13
1.00
0.91
0.99
0.98
3.16
f1 (ppm)

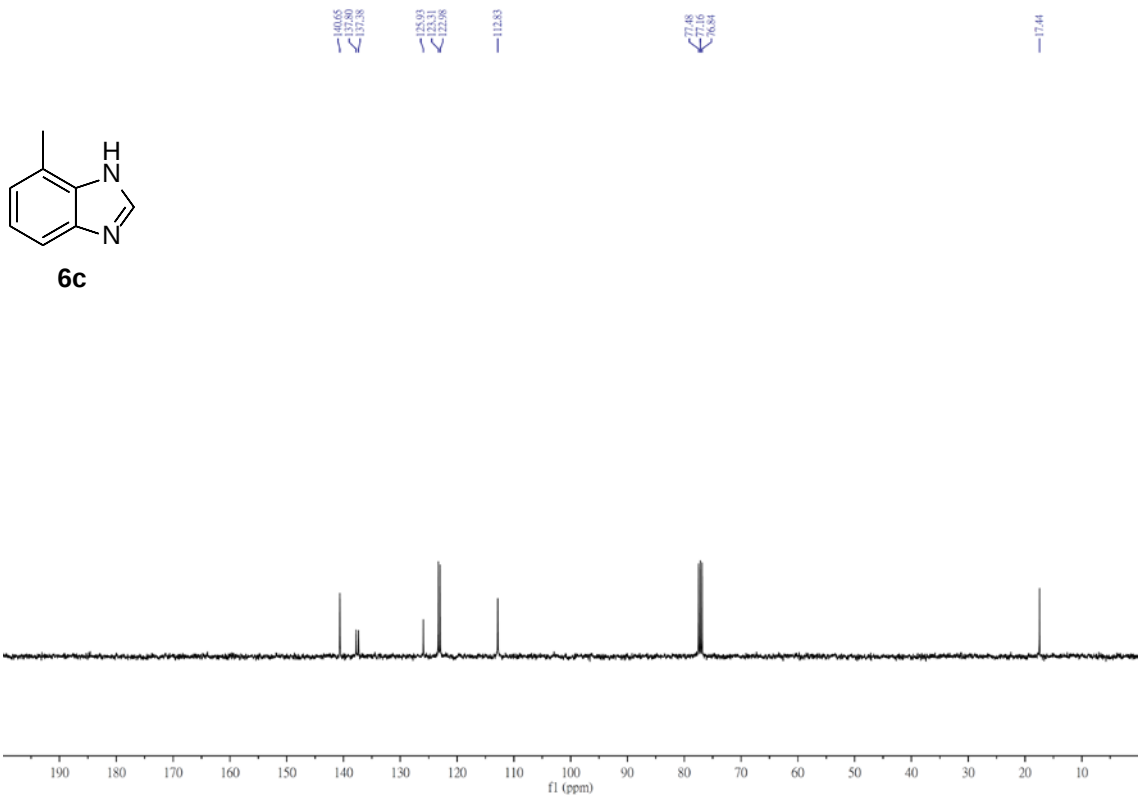
6b


140.73
137.61
136.45
132.80
124.46
115.51
114.92
77.48
77.16
76.84
21.74
f1 (ppm)

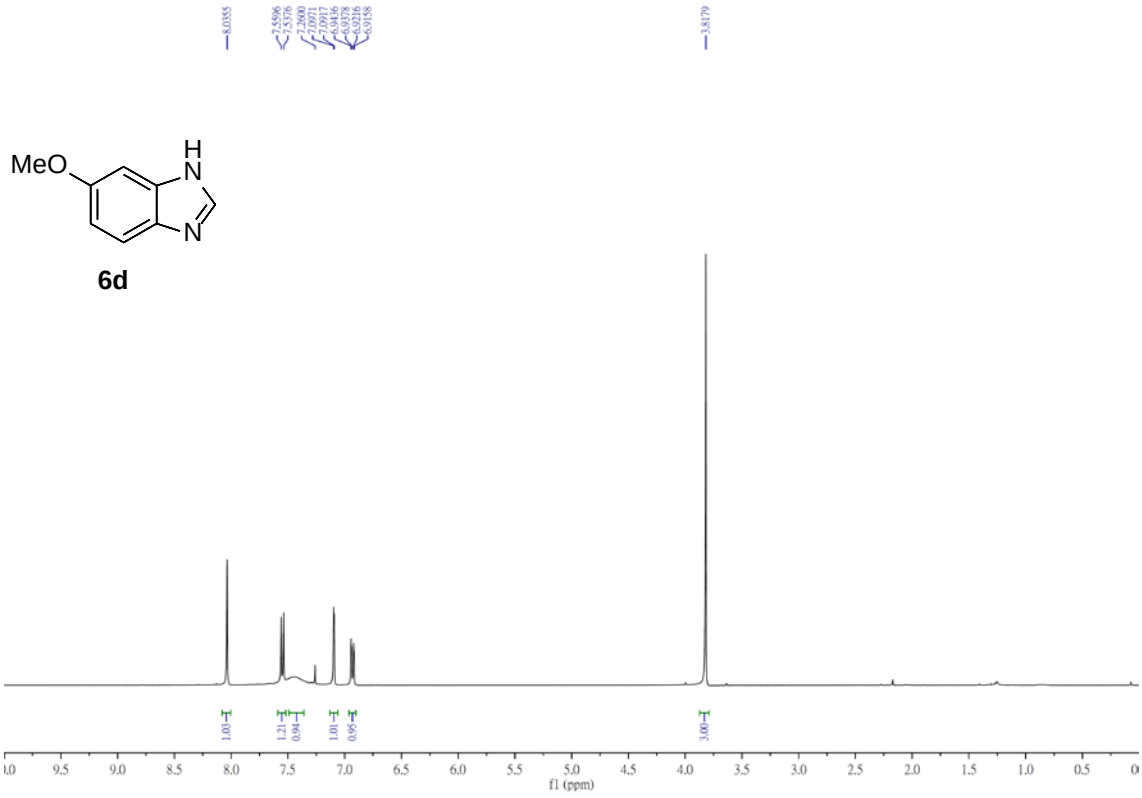
6b


10.0013
8.1601
7.5264
7.5065
7.3941
7.3835
7.2147
7.1092
7.1036
7.1035
2.6375
1.10
1.03
0.98
0.98
0.99
3.00
f1 (ppm)

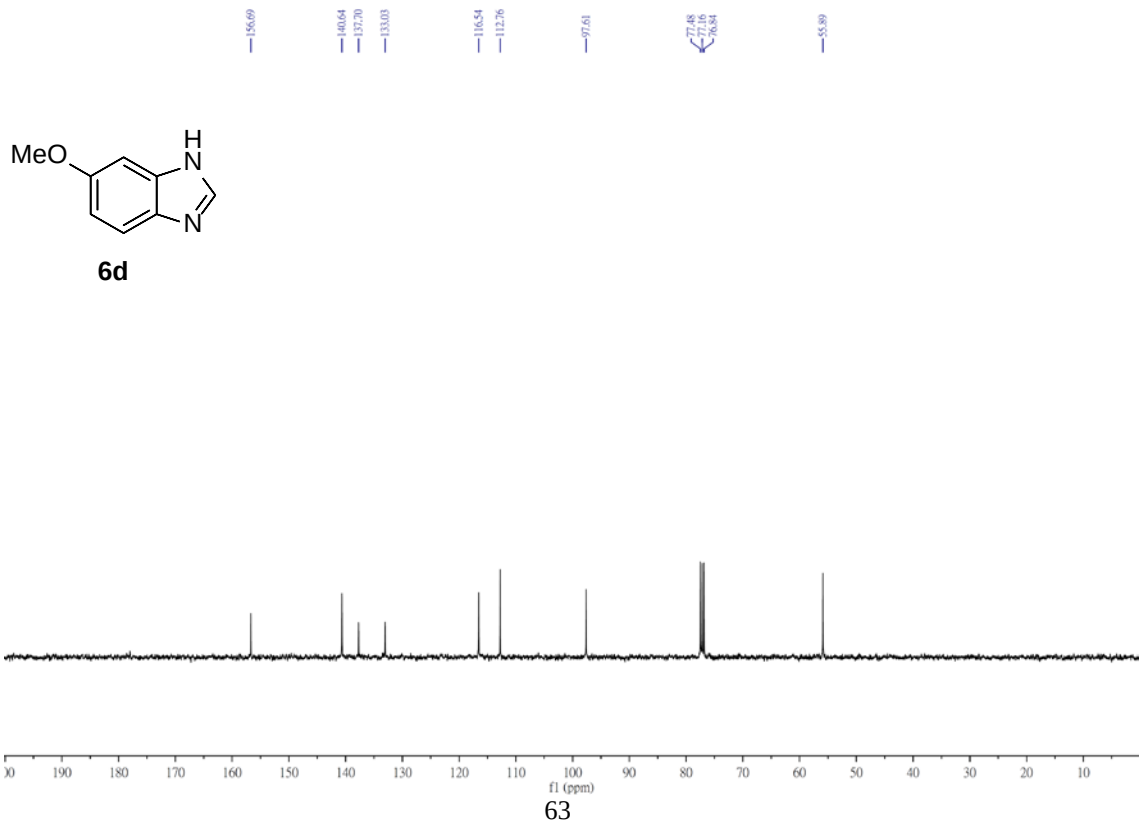
6c


140.65
137.90
137.38
125.03
123.31
122.98
112.83
77.48
77.16
76.84
17.44
f1 (ppm)

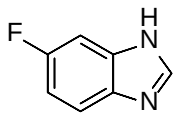
6c


8.0355
7.5596
7.5376
7.2600
7.0971
7.0757
6.9436
6.9378
6.9216
6.9158
3.8179
MeO
H
N
N
1.03
1.21
0.94
1.01
0.95
3.00
f1 (ppm)

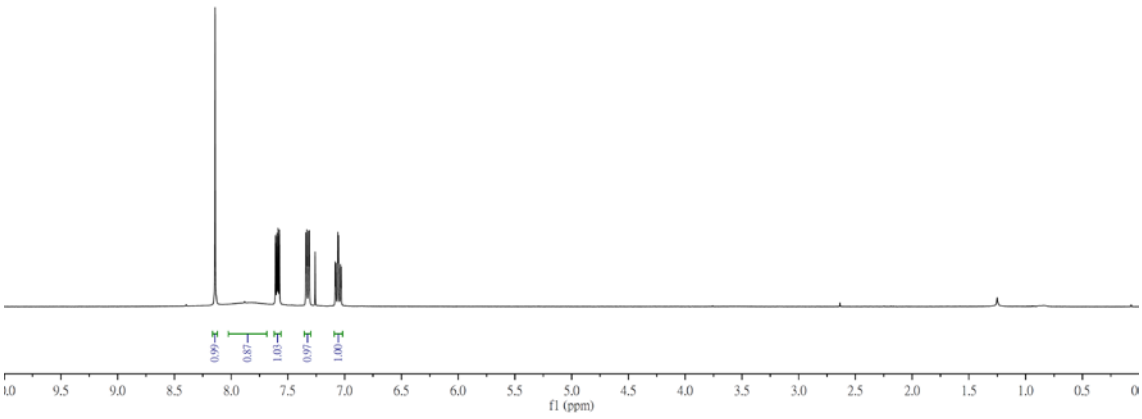
6d

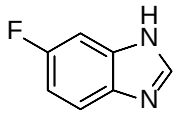

156.69
140.64
137.20
133.03
116.54
112.76
97.61
77.48
77.16
76.84
55.89
MeO
H
N
N
f1 (ppm)

6d

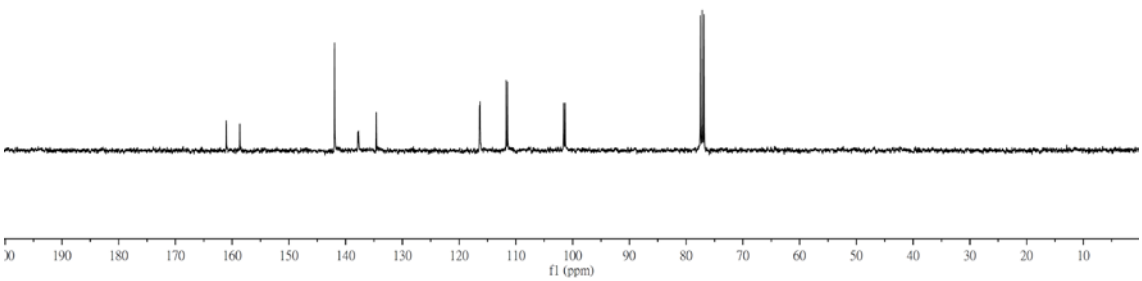


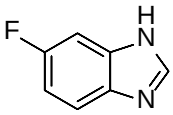
6e



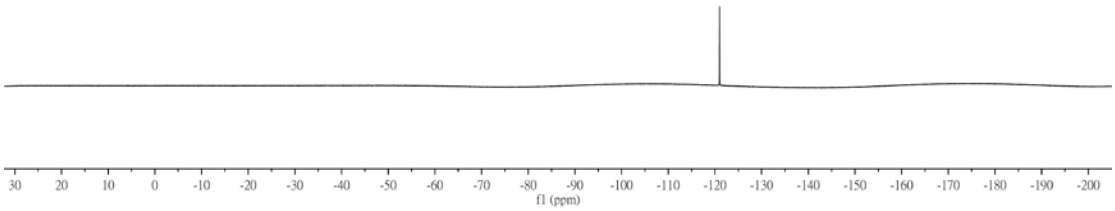


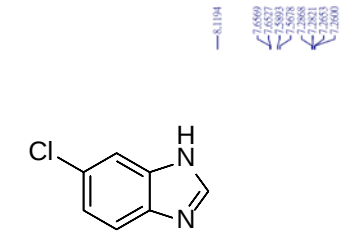
6e




F
H
N
N

6e


30 20 10 0 -10 -20 -30 -40 -50 -60 -70 -80 -90 -100 -110 -120 -130 -140 -150 -160 -170 -180 -190 -200
f1 (ppm)


8.1194
7.6569
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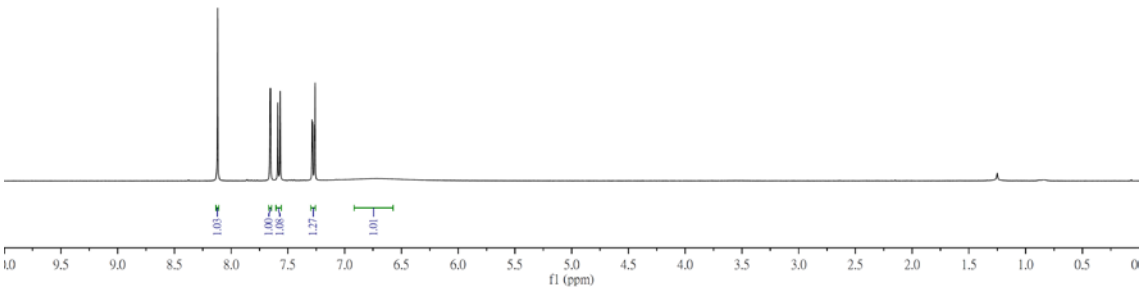
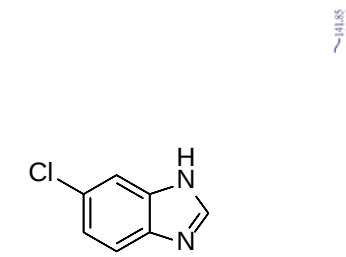
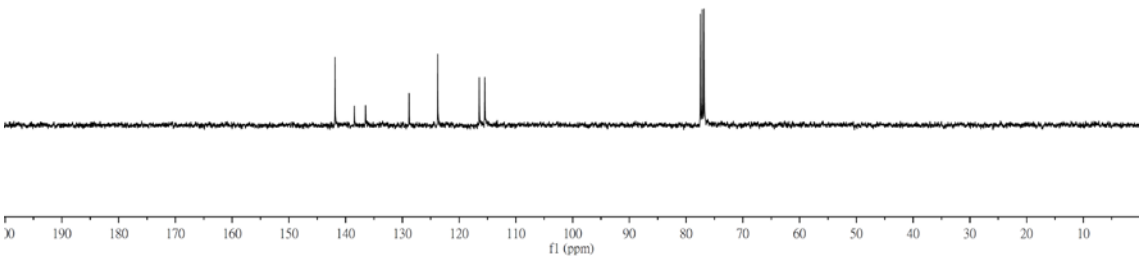
7.5893
7.5678
7.2868
7.2821
7.2655
7.2600
Cl
H
N
N

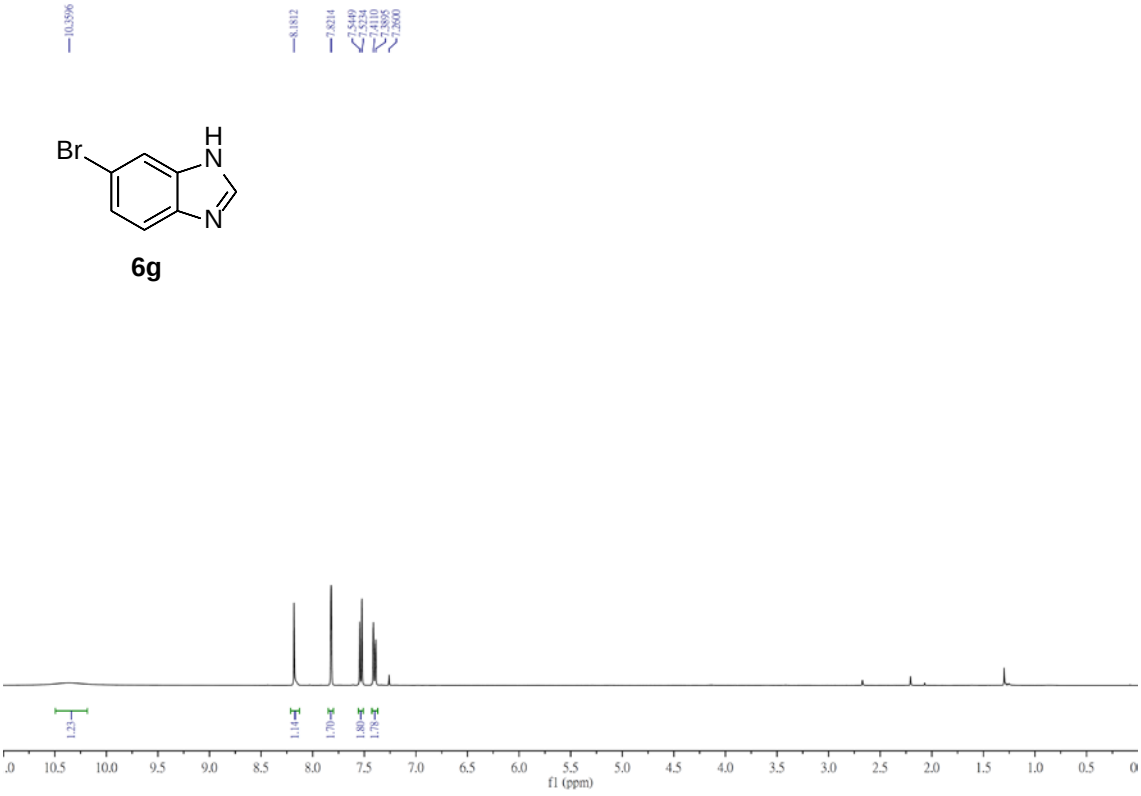
6f


1.03
1.00
1.08
1.27
1.01
f1 (ppm)

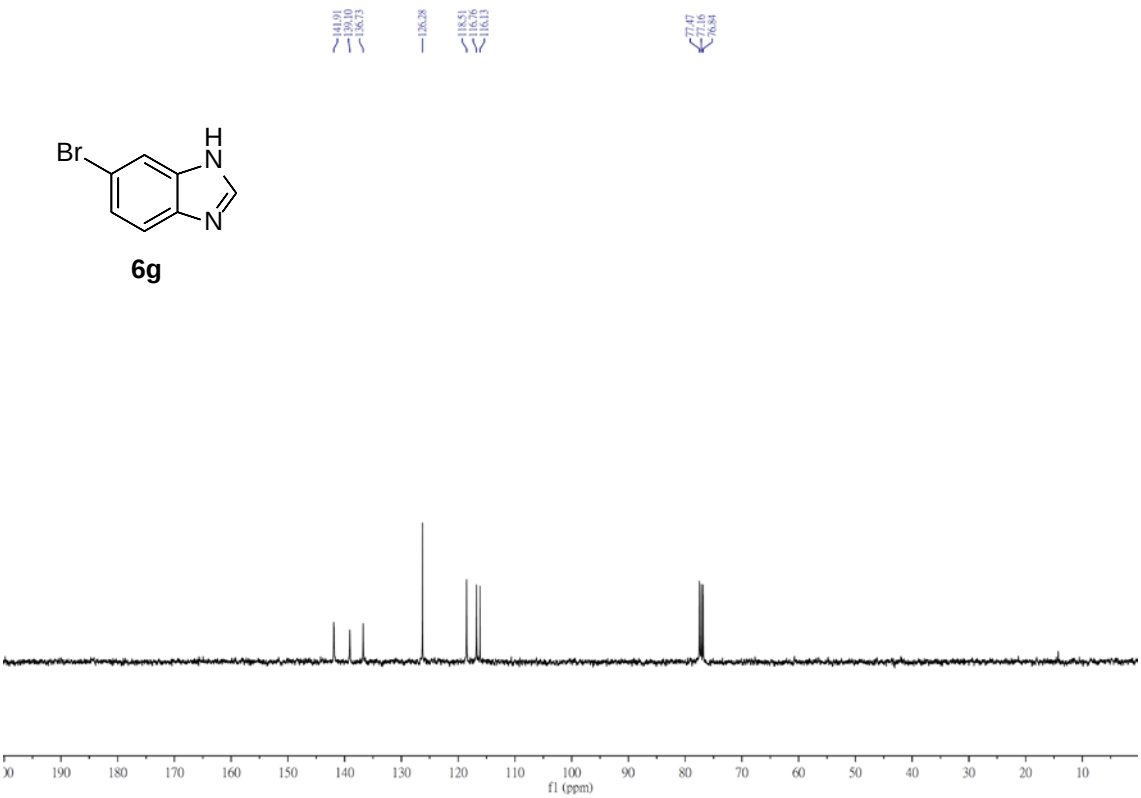

141.85
138.45
136.48
128.61
123.77
116.46
115.47
77.48
77.16
76.84
Cl
H
N
N

6f

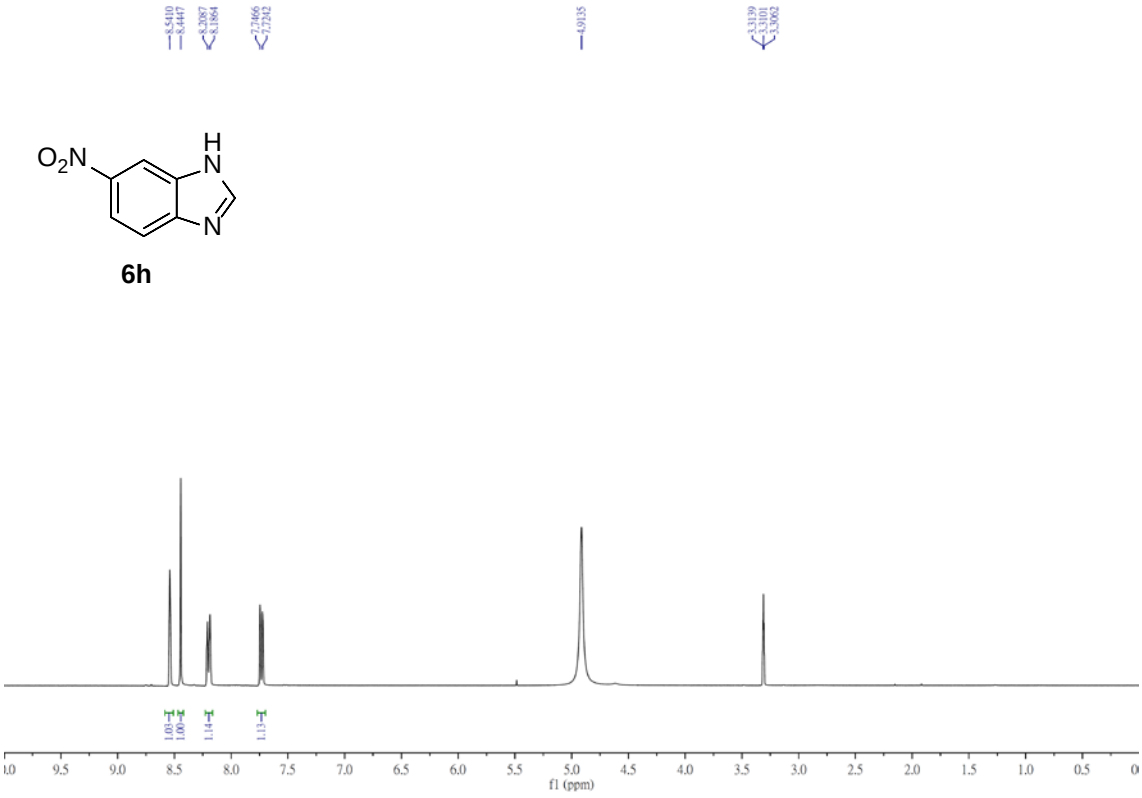

f1 (ppm)

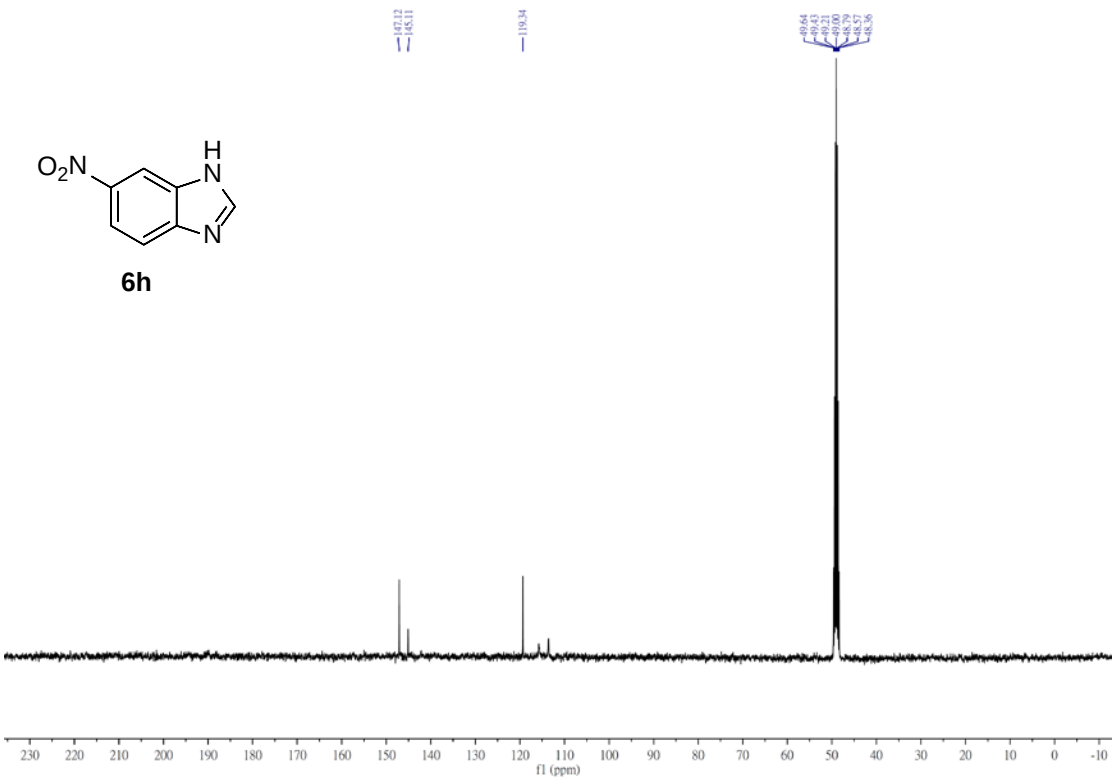

10.3596
8.1812
7.8214
7.5449
7.5234
7.4110
7.3895
7.2600
Br
H
N
N
1.23
1.14
1.70
1.80
1.78
f1 (ppm)

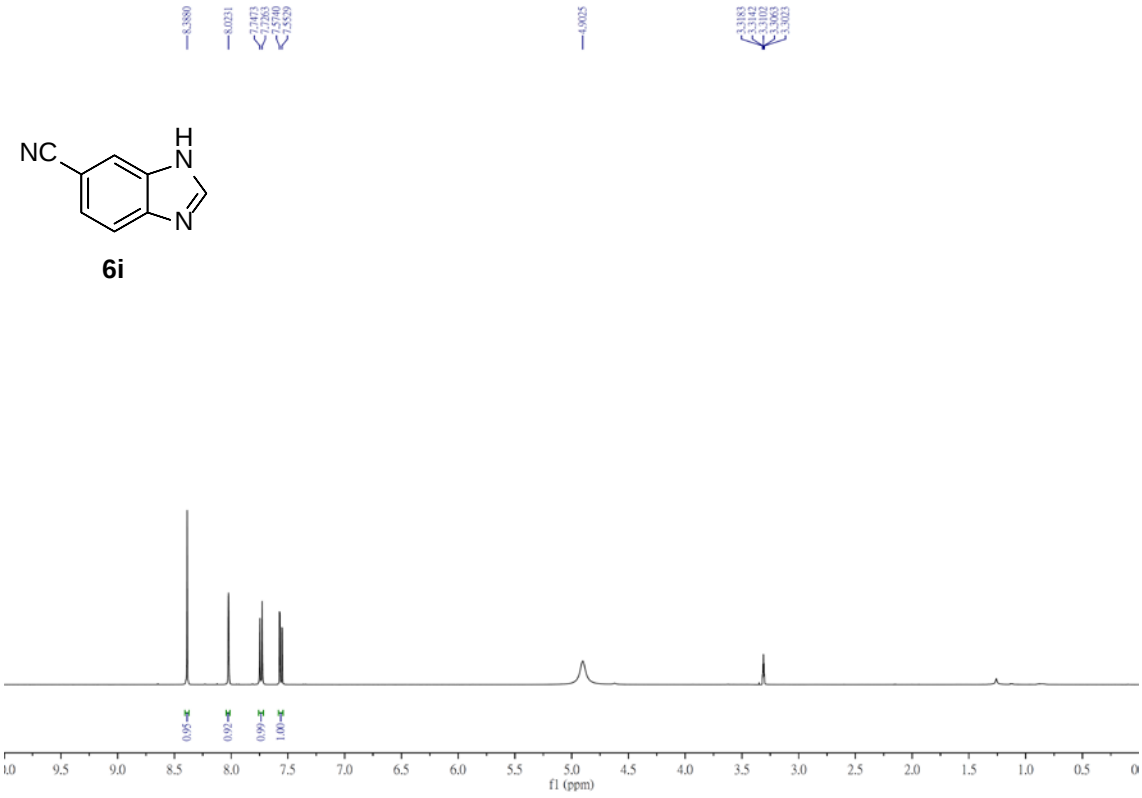
6g


141.91
139.10
136.73
126.28
118.51
116.76
116.13
77.47
77.16
76.84
Br
H
N
N
f1 (ppm)

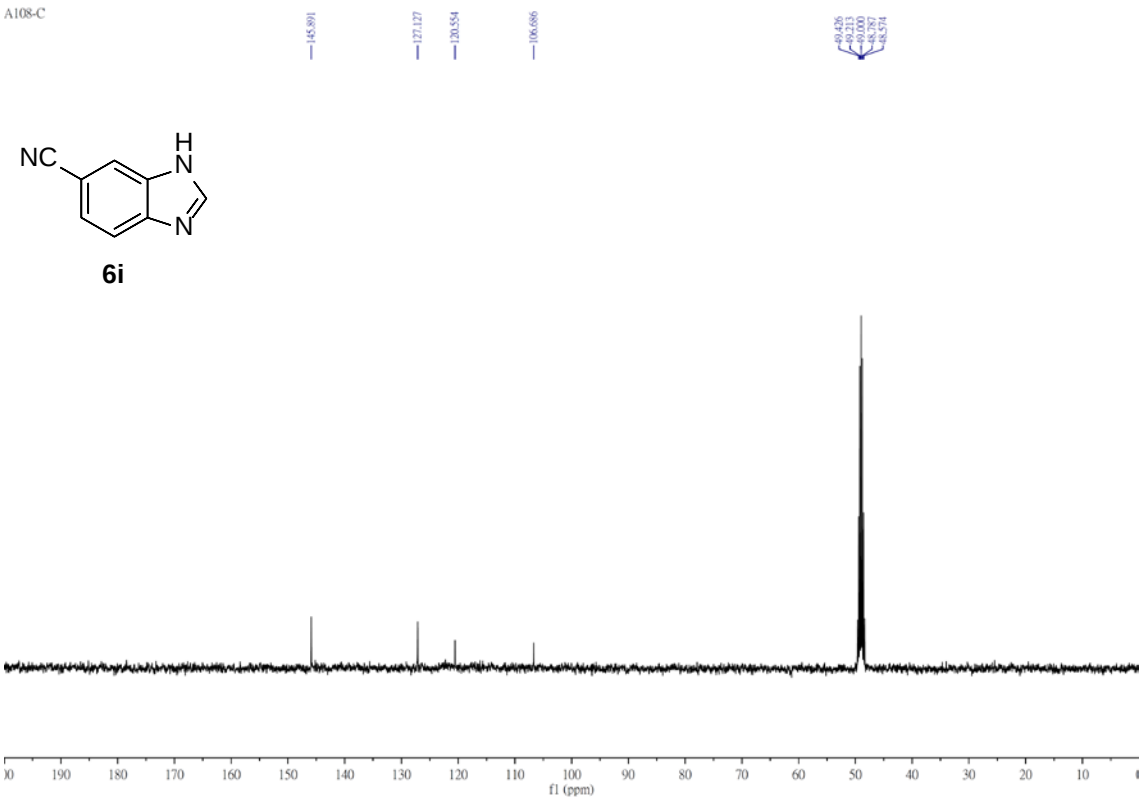
6g


8.5410
8.4447
8.2087
8.1864
7.2866
7.2528
4.9135
3.3139
3.3101
3.3062
O2N
H
N
N
6h
1.03
1.00
1.14
1.13
f1 (ppm)

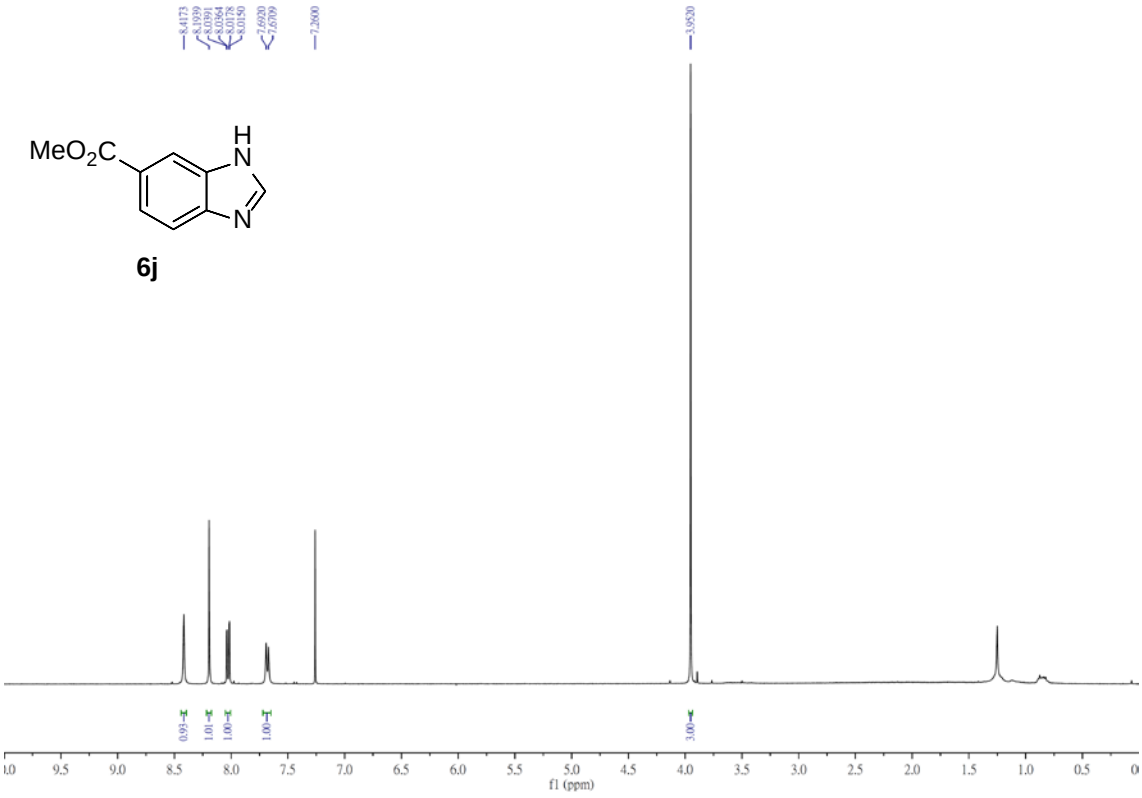

147.12
145.11
119.34
49.64
49.43
49.21
49.00
48.79
48.57
48.36
O2N
H
N
N
6h
f1 (ppm)


8.3880
8.0231
7.7473
7.7263
7.5740
7.5559
4.9025
3.3183
3.3142
3.3102
3.3063
3.3023
NC
H
N
N
0.95
0.92
0.99
1.00
f1 (ppm)

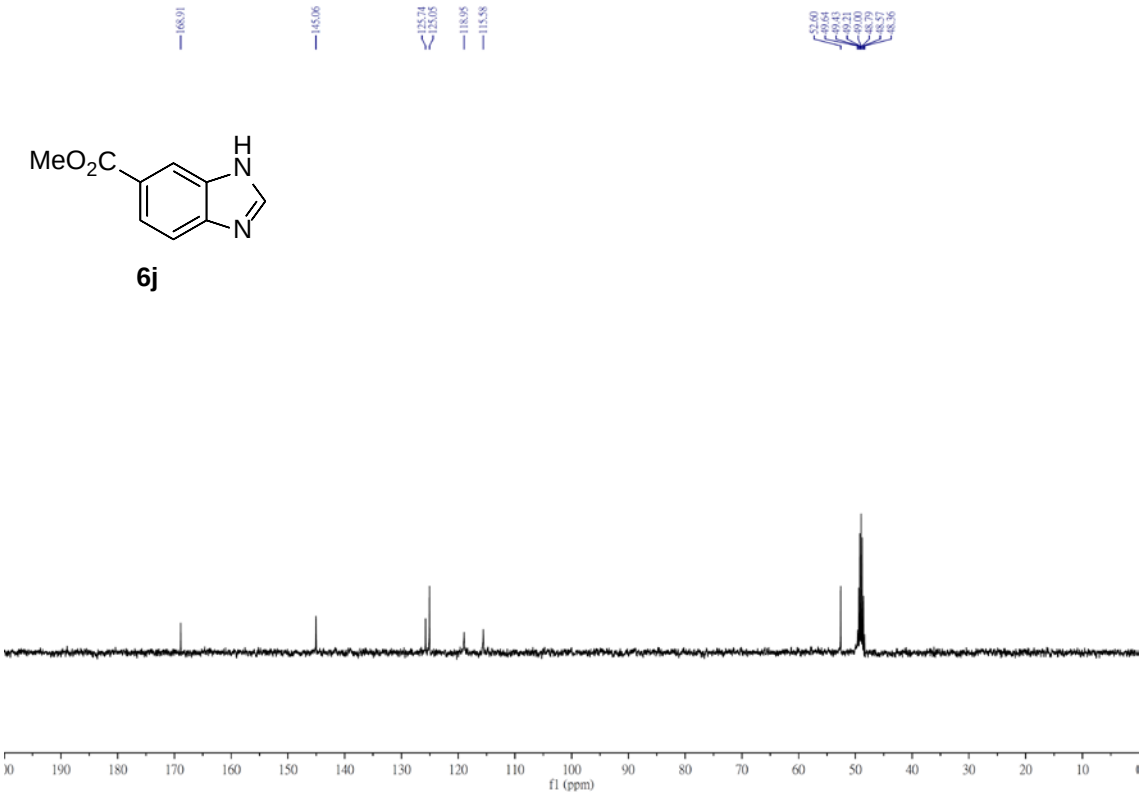
6i


A108-C
145.891
127.127
120.554
106.686
49.426
49.213
49.000
48.787
48.574
NC
H
N
N
f1 (ppm)

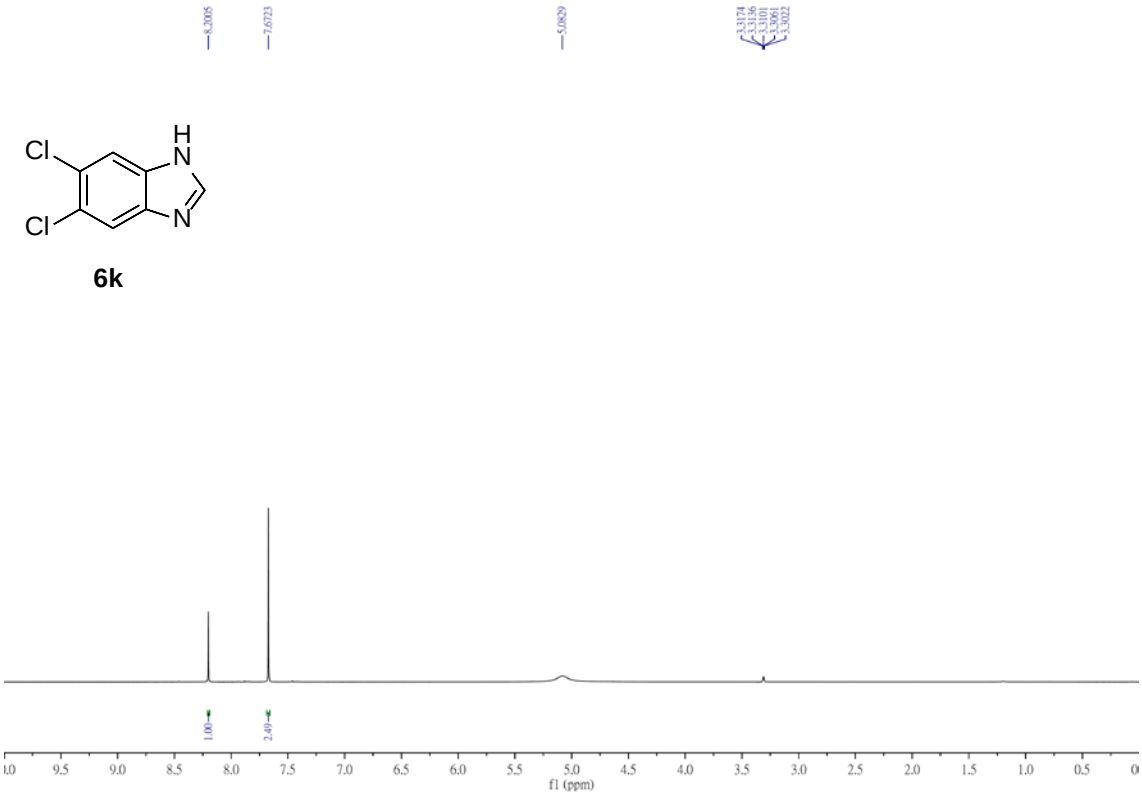
6i


8.4173
8.3199
8.3091
8.0364
8.0178
8.0150
7.6920
7.6709
7.2600
3.9520
MeO2C
H
N
N
0.93
1.01
1.00
1.00
3.00
f1 (ppm)

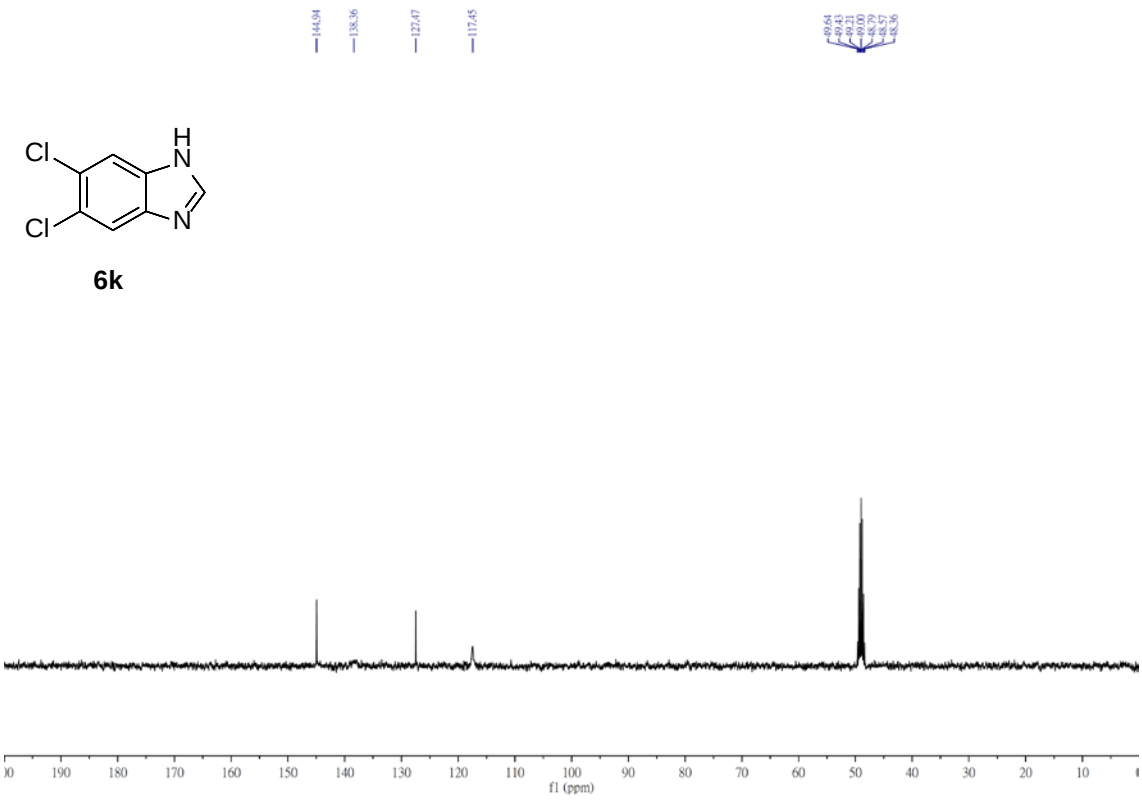
6j


168.91
145.06
125.74
125.05
118.95
115.58
52.60
49.64
49.43
49.21
49.00
48.79
48.57
48.36
MeO2C
H
N
N
f1 (ppm)

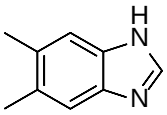
6j


8.2005
7.6723
5.0829
3.3174
3.3136
3.3101
3.3061
3.3022
Cl
Cl
H
N
N
1.00
2.49
f1 (ppm)

6k

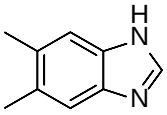

144.94
138.36
127.47
117.45
49.64
49.43
49.21
49.00
48.79
48.57
48.36
Cl
Cl
H
N
N
f1 (ppm)

6k



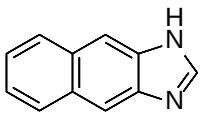
6l

7.9664
7.4250
7.2600
2.3713
1.00
1.96
6.07
f1 (ppm)

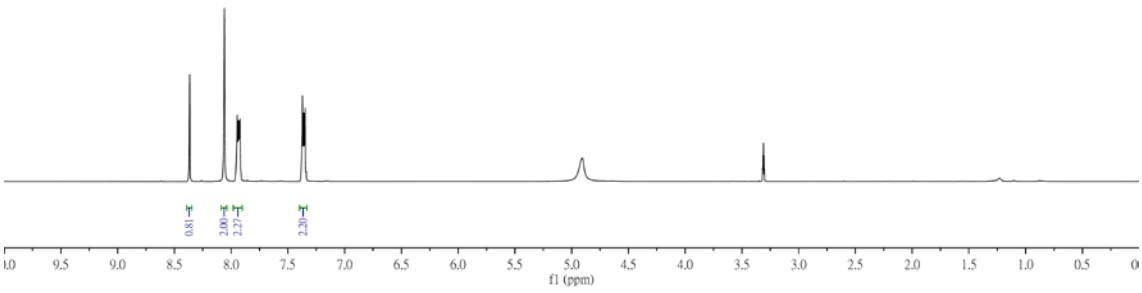


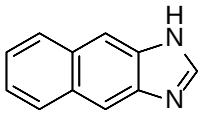
6l

139.87
136.28
132.08
115.66
77.48
77.16
76.84
20.52
f1 (ppm)

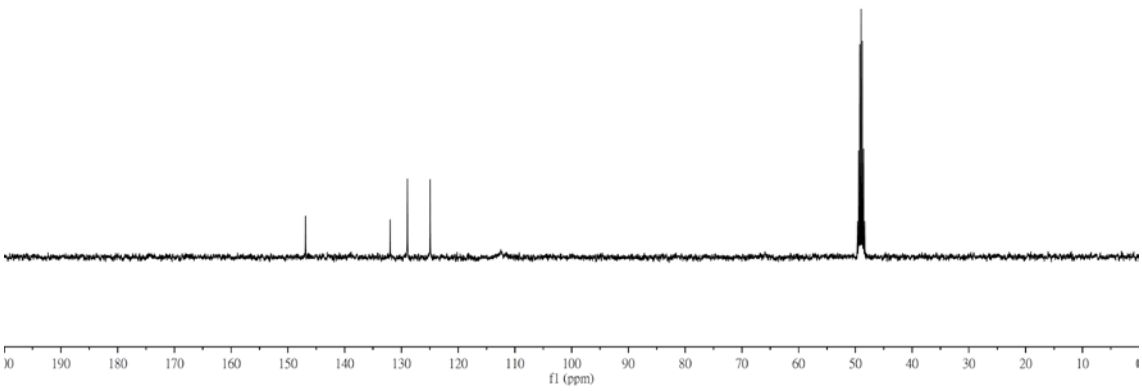


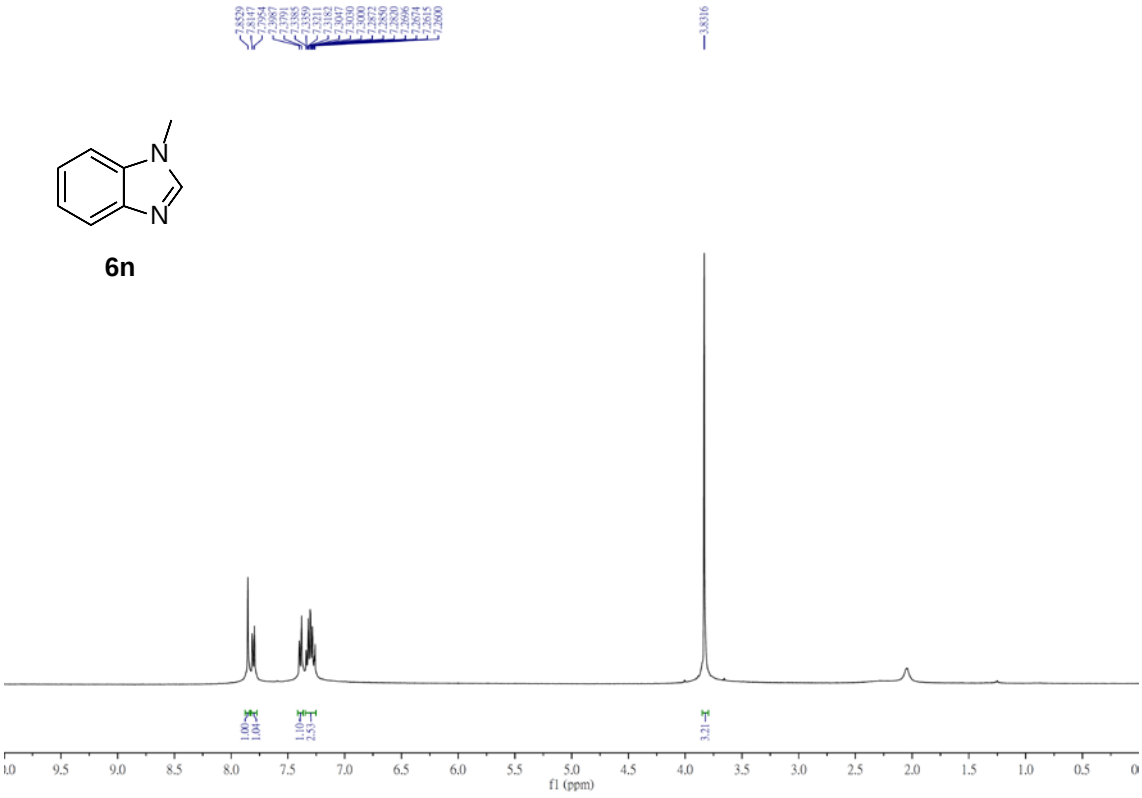
6m





6m





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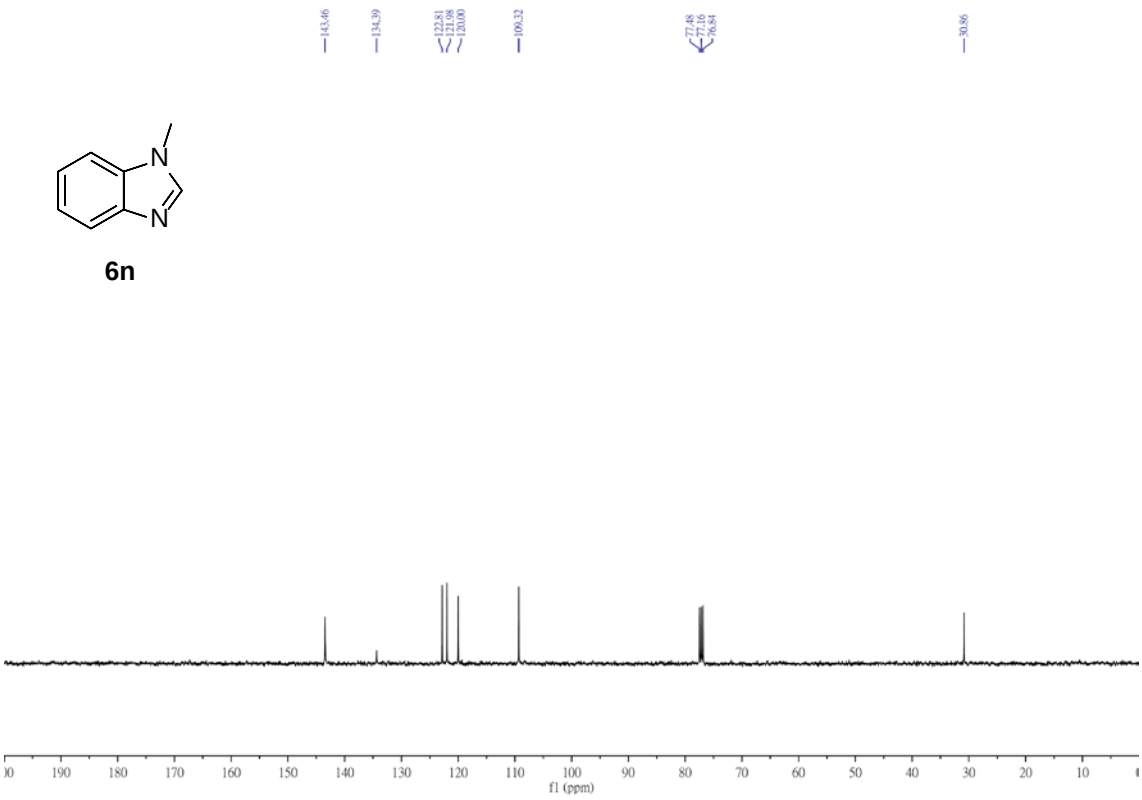

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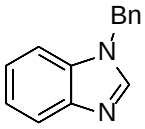

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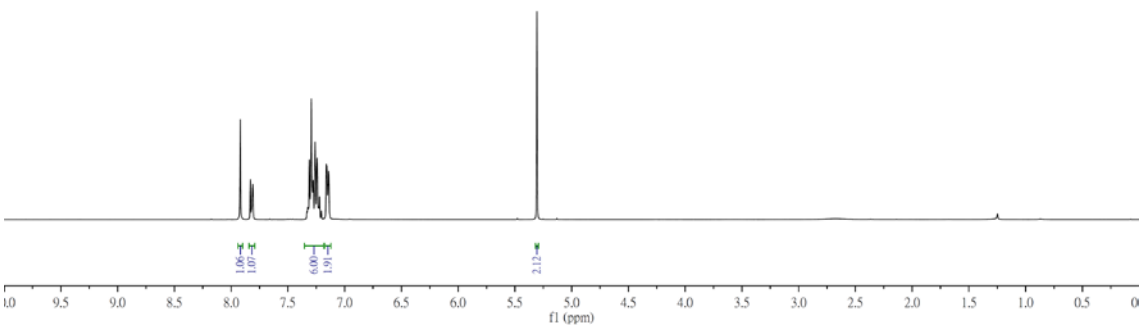
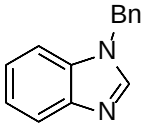
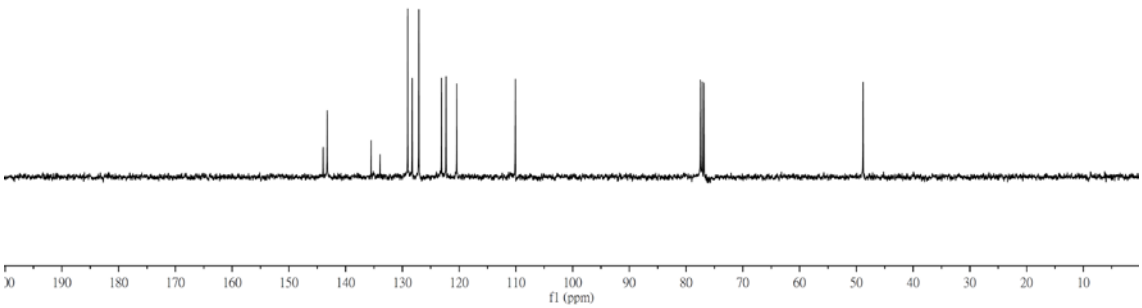
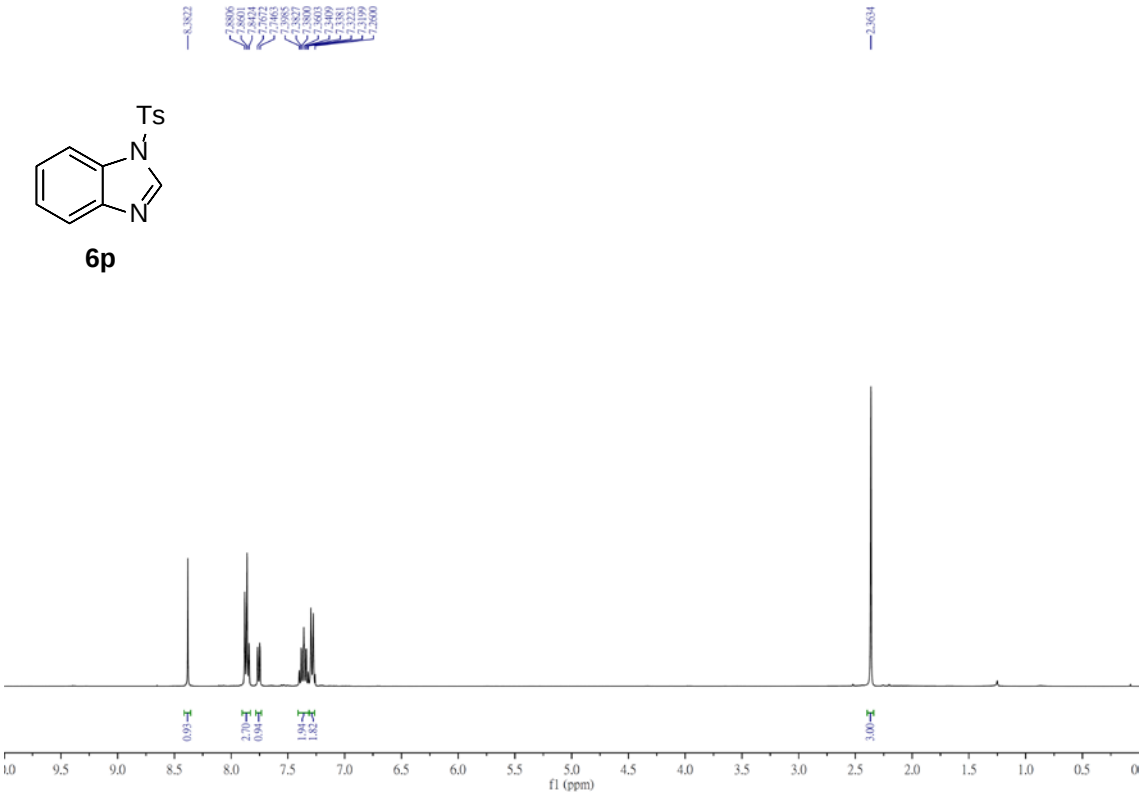




6o

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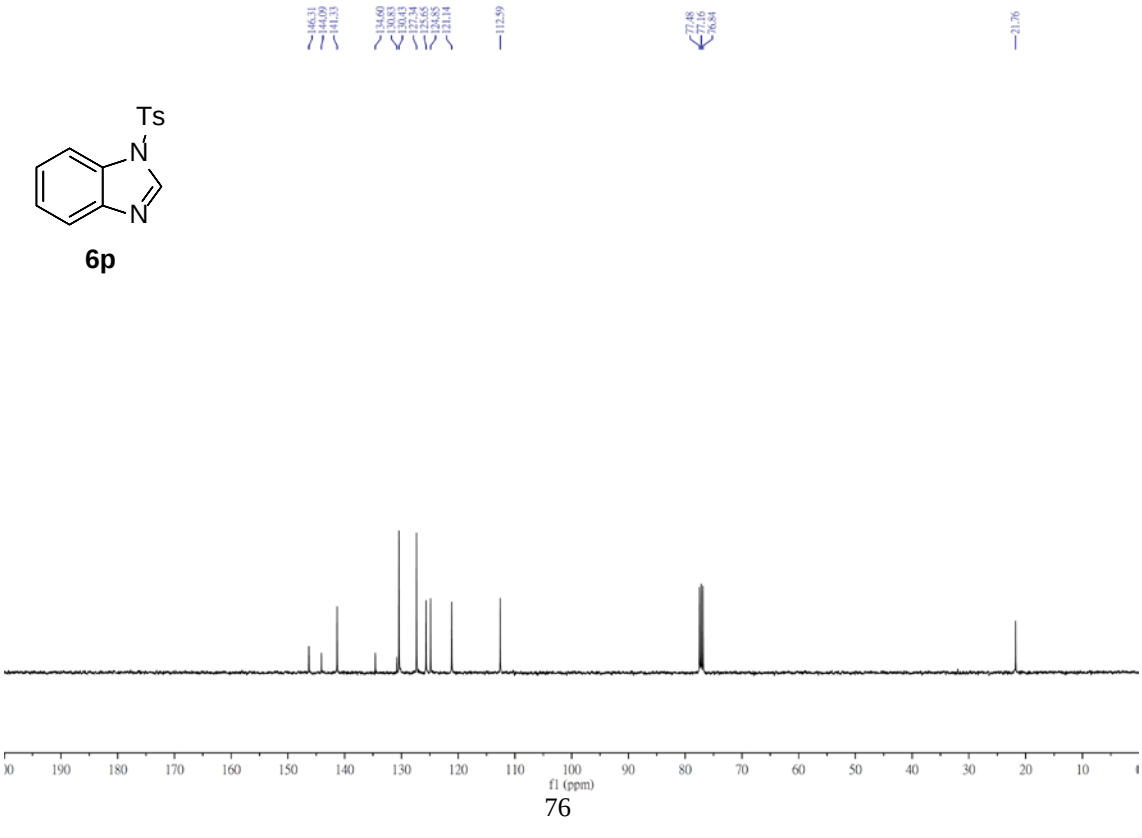
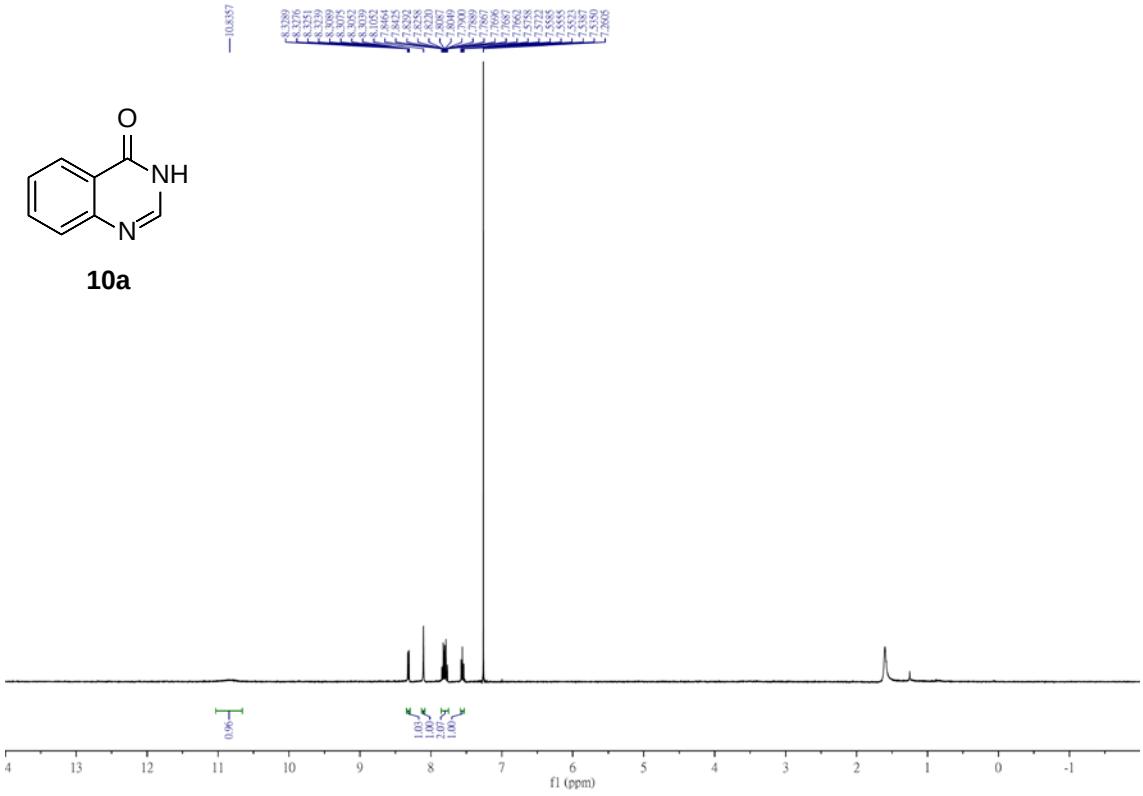
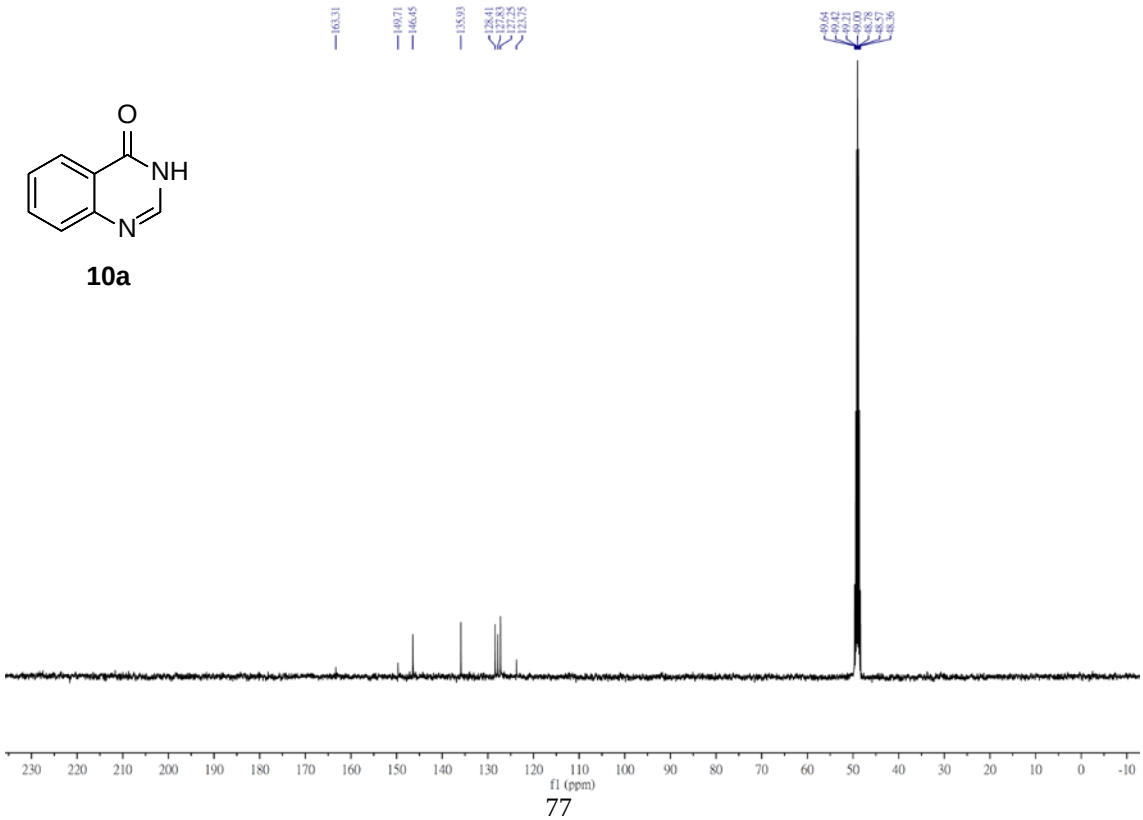
7.8806
7.8601
7.8444
7.7672
7.7463
7.3985
7.3837
7.3800
7.3603
7.3409
7.3381
7.3233
7.3199
7.2800
2.3634
Ts
N
N
6p
0.93
2.70
0.94
1.94
1.82
3.00
f1 (ppm)

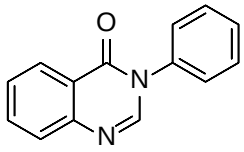

146.31
144.09
141.33
134.60
130.85
130.43
127.34
125.65
124.35
121.14
112.59
77.48
77.16
76.84
21.76
Ts
N
N
6p
f1 (ppm)



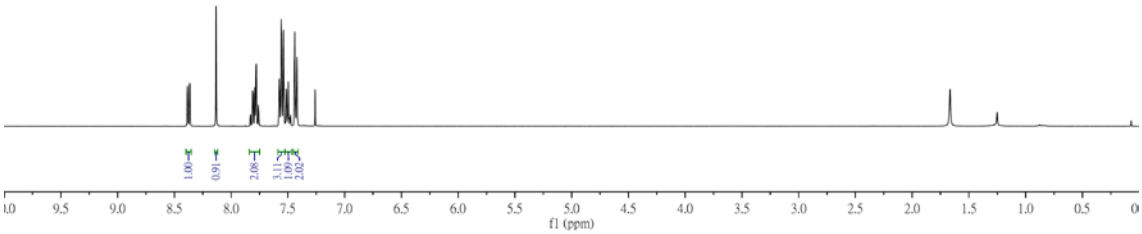
10a

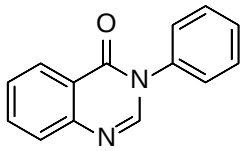


10a



10b





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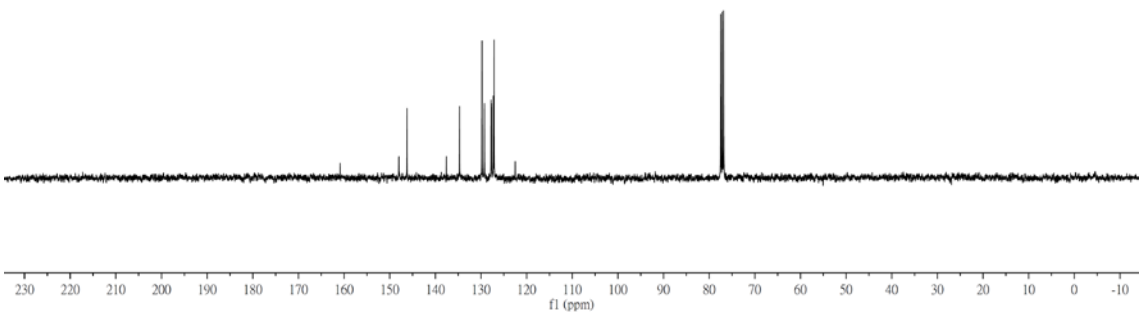
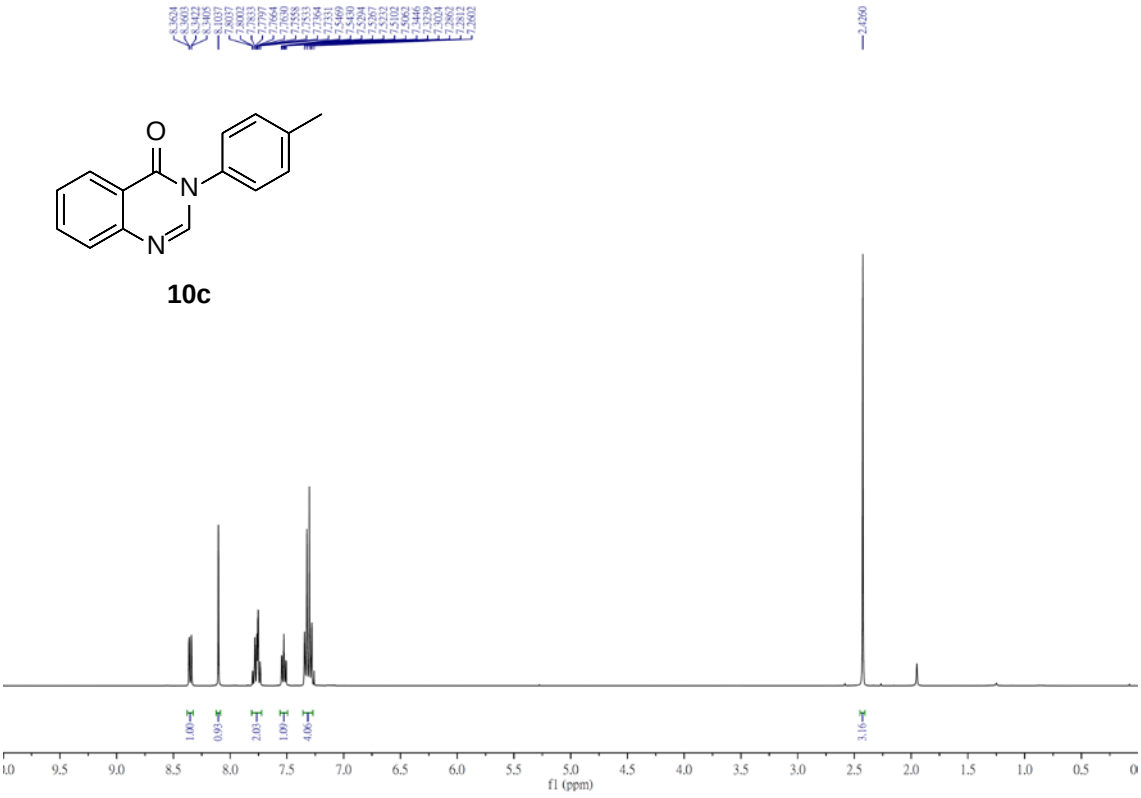
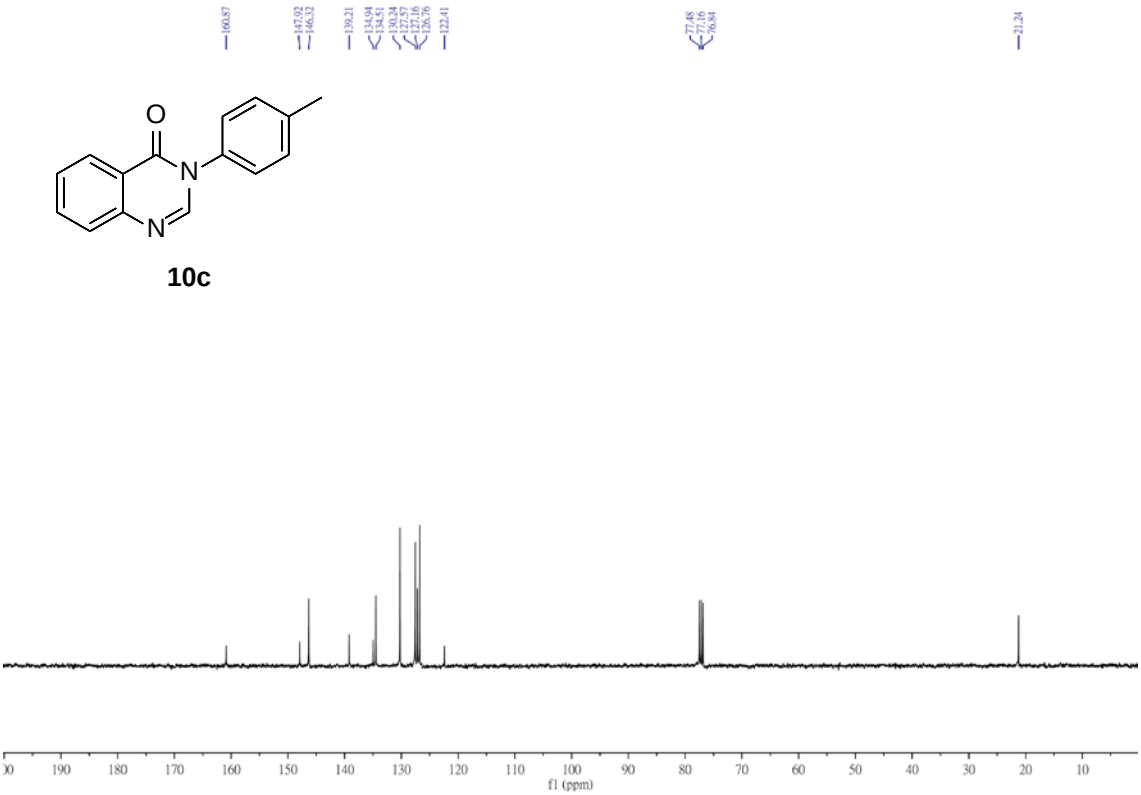



O
N
N
10c
1.00
0.93
2.03
1.09
4.06
3.16
f1 (ppm)


O
N
N
10c
160.67
147.02
146.32
139.21
134.94
134.51
130.24
127.57
127.16
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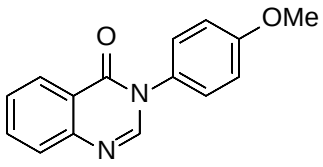
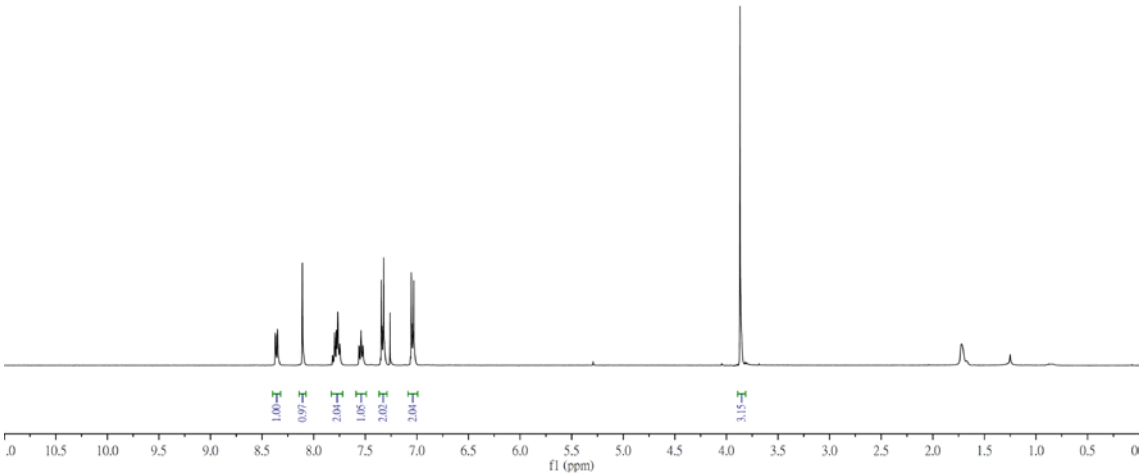
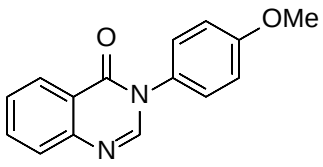
77.48
77.16
76.84
21.24
f1 (ppm)

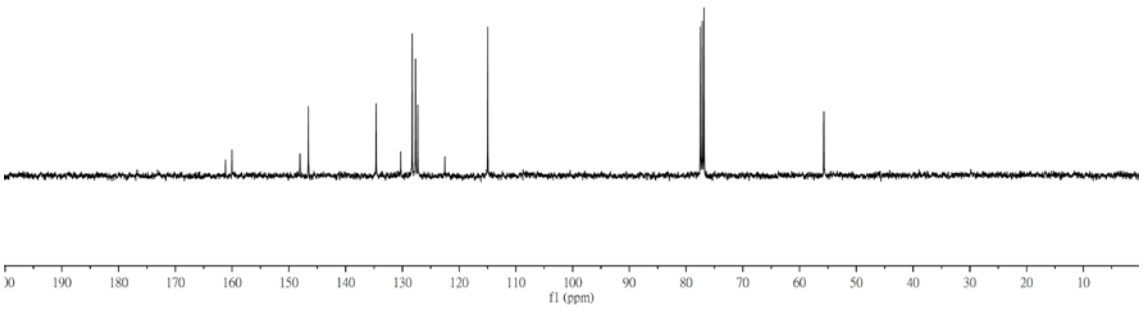


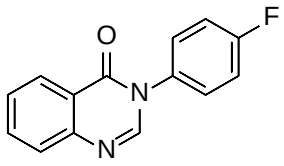
10d



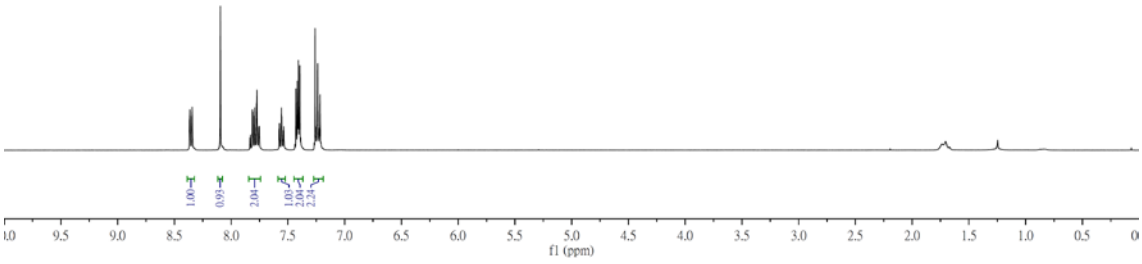


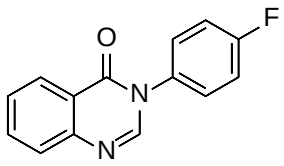
10d





10e





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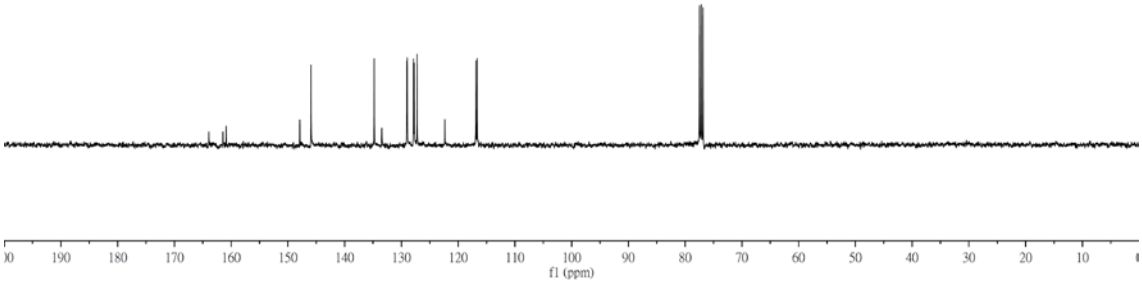
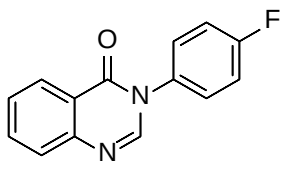
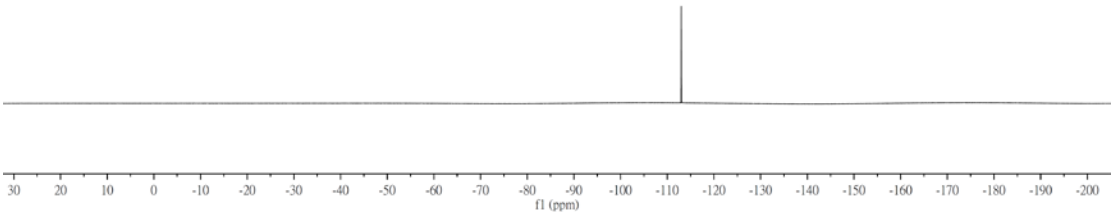
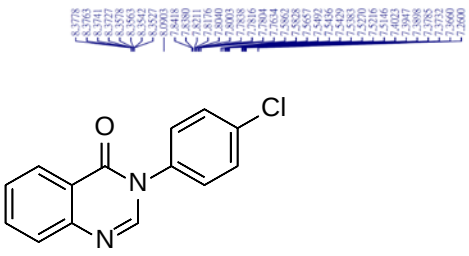


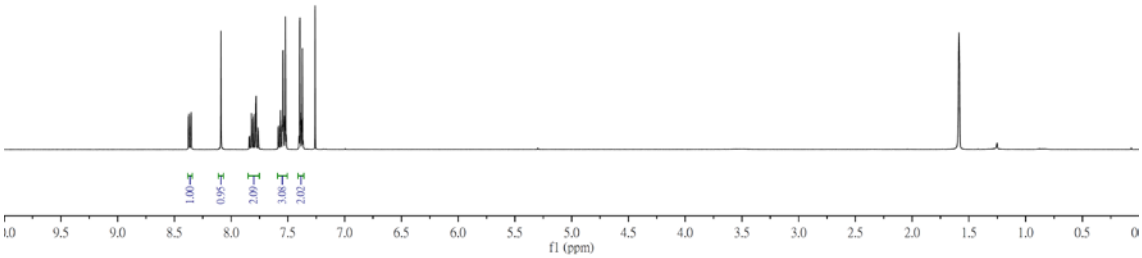

O
N
N
F

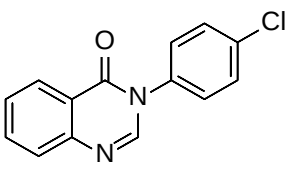
10e


30 20 10 0 -10 -20 -30 -40 -50 -60 -70 -80 -90 -100 -110 -120 -130 -140 -150 -160 -170 -180 -190 -200
f1 (ppm)

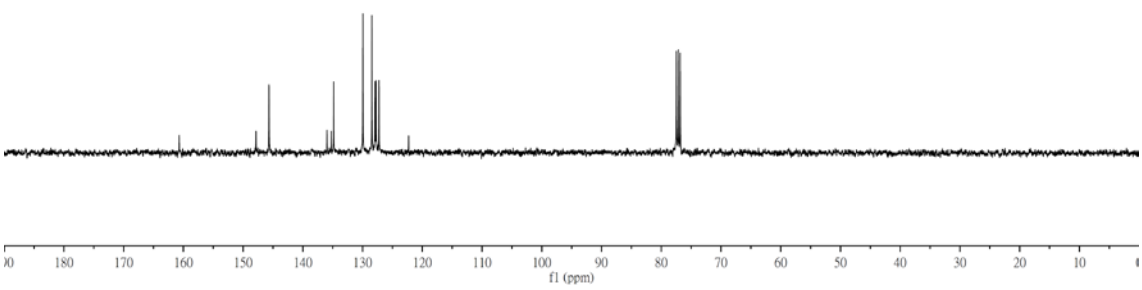


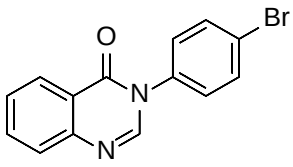
10f



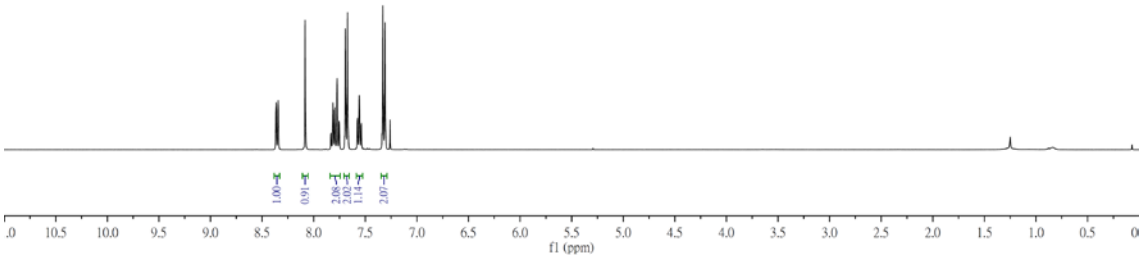


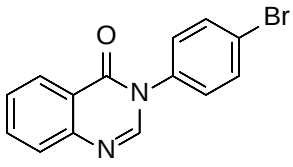
10f



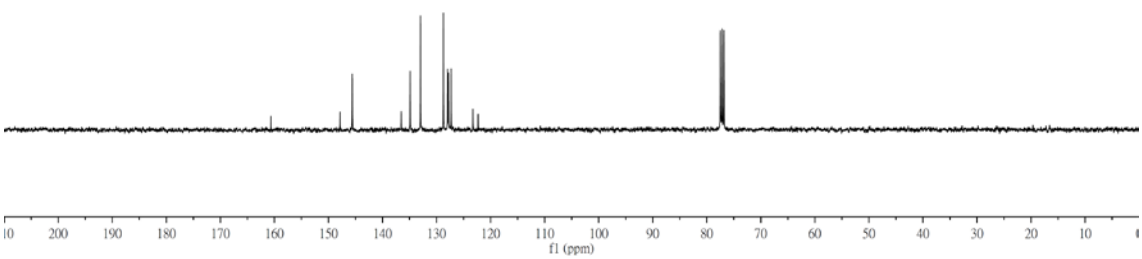


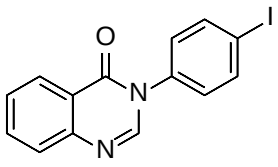
10g



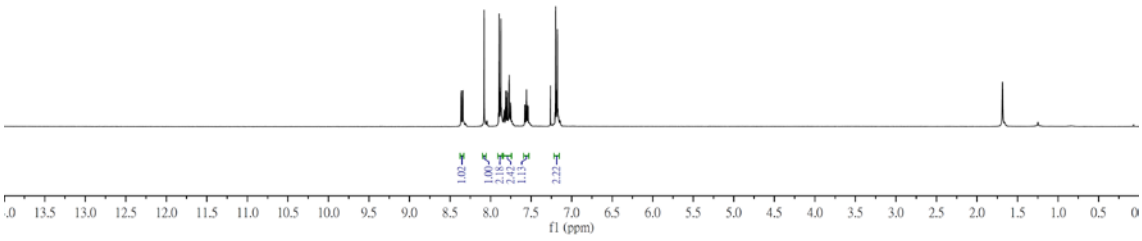


10g

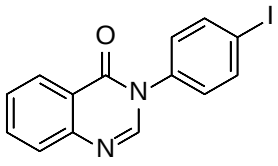



O
N
N
I

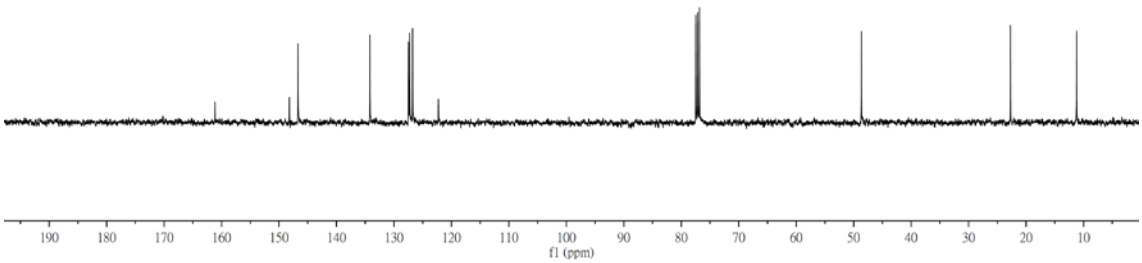
10h

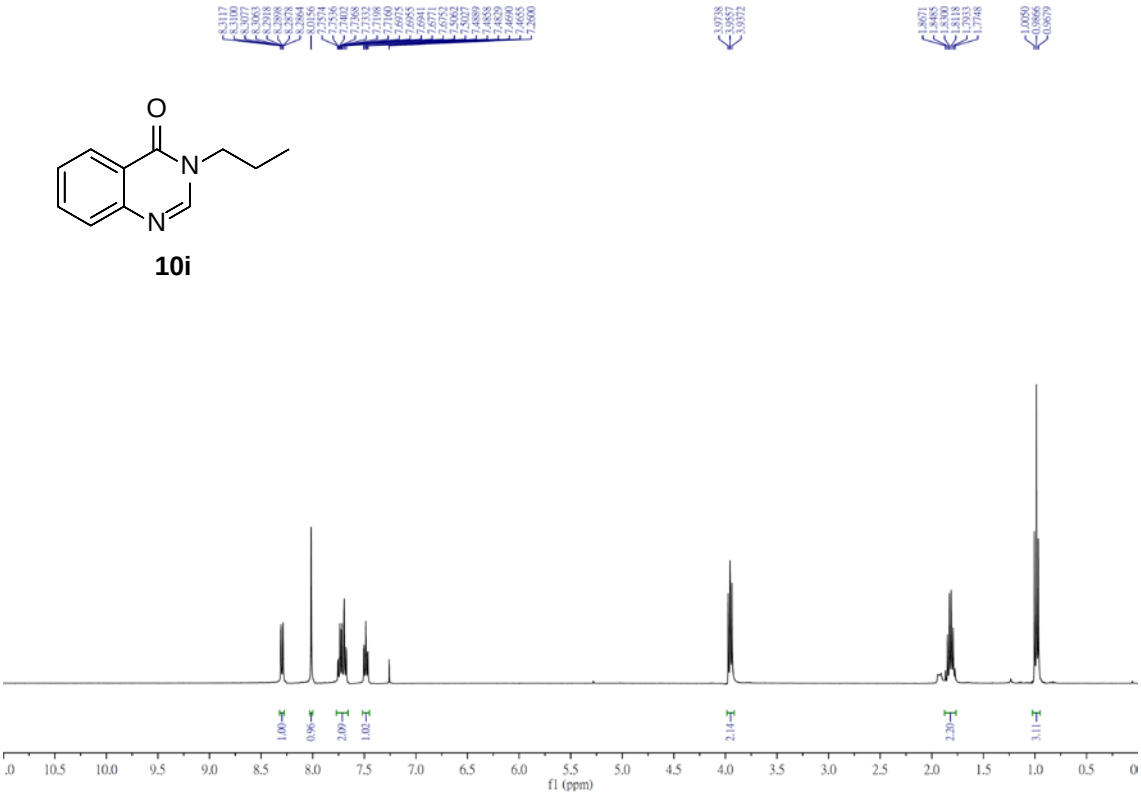

1.02
1.00
2.18
2.42
1.13
2.22
f1 (ppm)


161.13
148.20
146.70
134.19
127.47
127.27
126.76
122.26
77.48
77.16
76.84
68.66
22.73
11.21


O
N
N
I

10h


f1 (ppm)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8.3077
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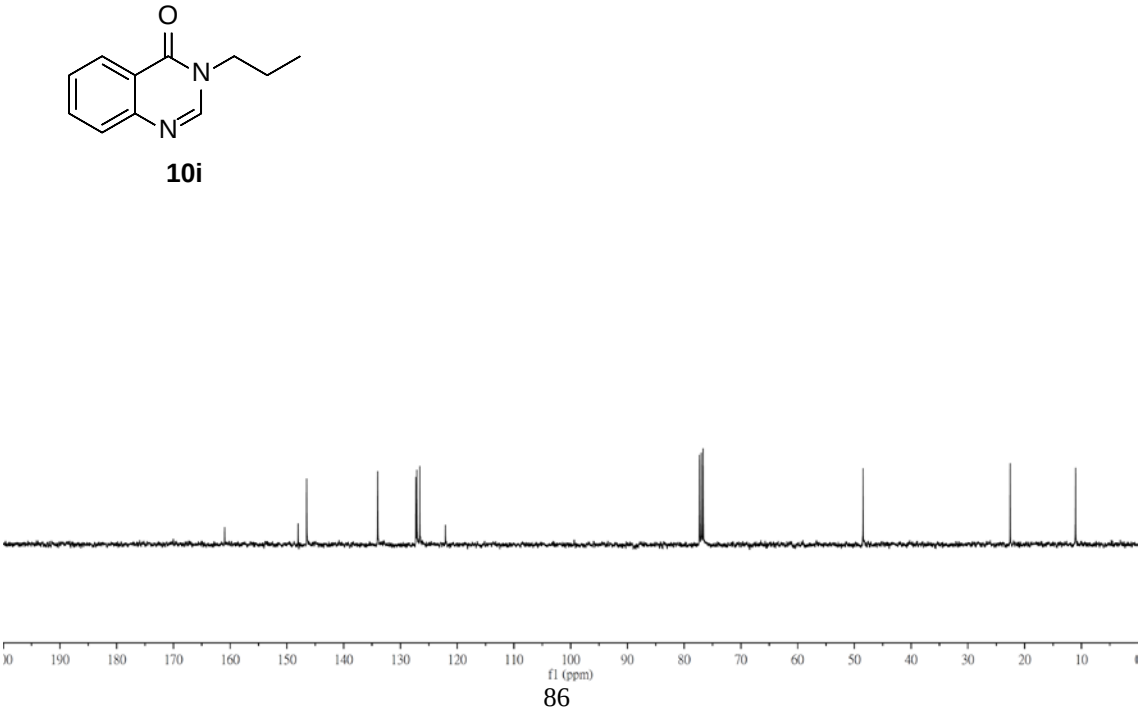
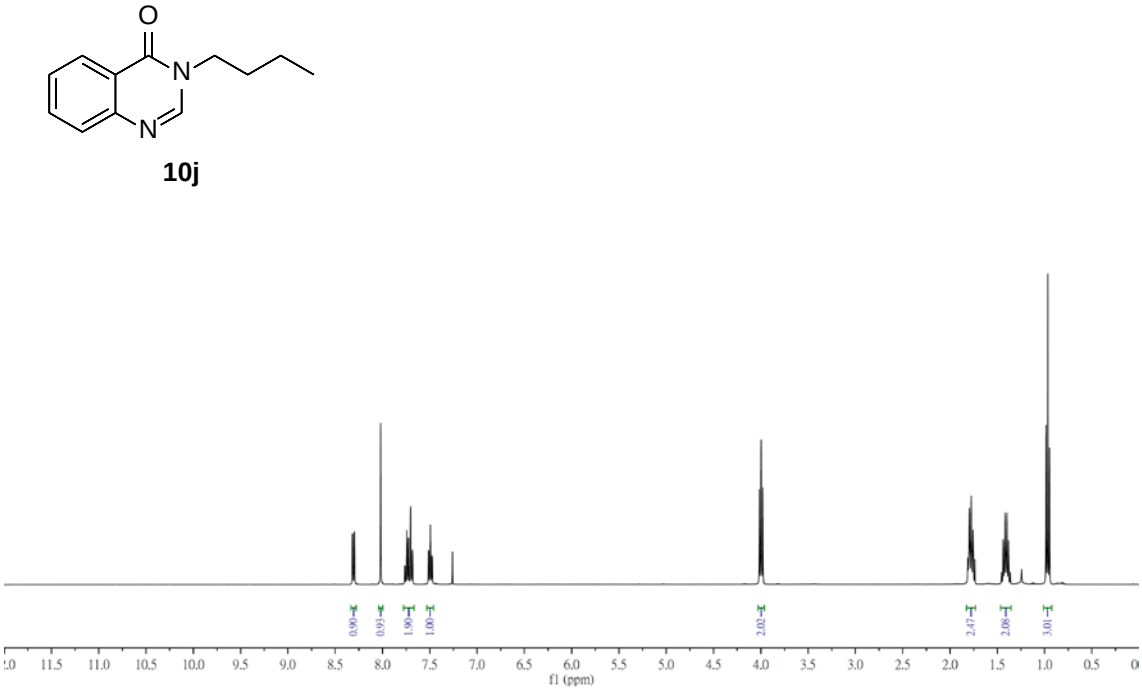
8.2938
8.2698
8.2678
8.2664
8.0156
8.0134
7.7536
7.7402
7.7388
7.7353
7.7338
7.7160
7.6975
7.6955
7.6641
7.6571
7.6752
7.5062
7.5027
7.4889
7.4858
7.4829
7.4690
7.4655
7.2600
3.9738
3.9557
3.9372
1.8671
1.8485
1.8300
1.8118
1.7933
1.7748
1.0950
1.0096
0.9879
1.00
0.96
2.09
1.02
2.14
2.20
3.11
f1 (ppm)

10i


160.05
148.02
146.52
134.01
127.29
127.09
126.59
122.08
77.30
77.06
76.66
68.48
22.55
11.03
f1 (ppm)

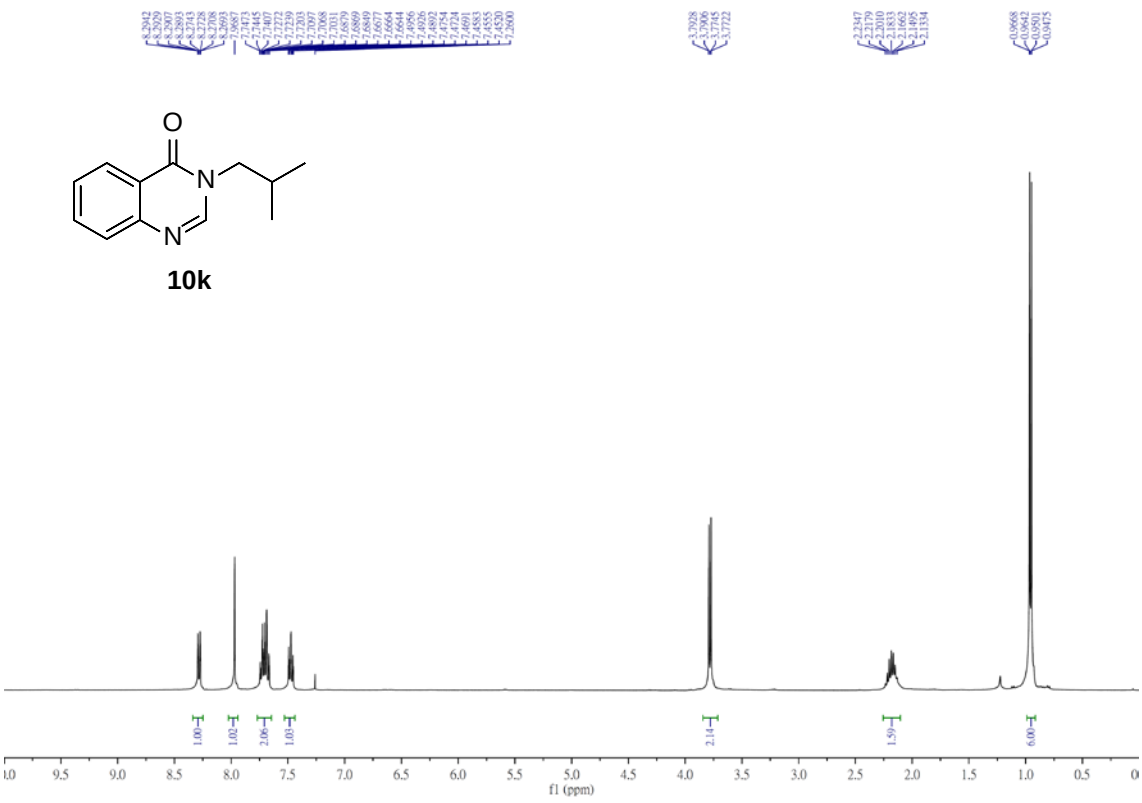
10i

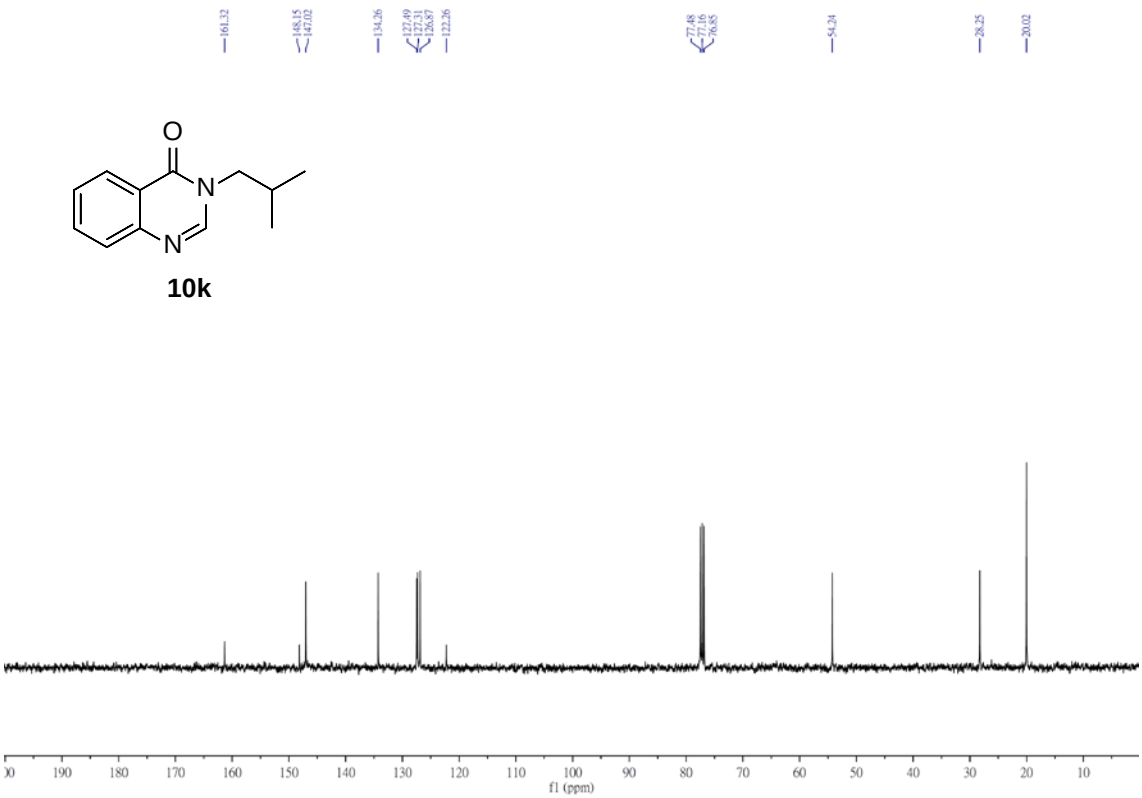

8.3180
8.3147
8.2680
8.2647
7.7644
7.7610
7.7438
7.7406
7.7206
7.7233
7.7021
7.6845
7.5138
7.5107
7.4965
7.4937
7.4699
7.4496
7.4434
7.2800
4.0149
3.9966
3.9781
1.8126
1.7942
1.7753
1.7694
1.7567
1.7375
1.4370
1.4181
1.0889
1.0657
0.9473
0.90
0.93
1.90
1.00
2.02
2.47
2.08
3.01
f1 (ppm)

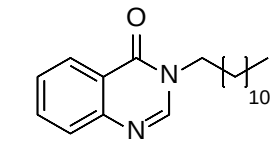
10j

161.15
148.22
146.68
134.21
127.49
127.29
126.78
122.28
77.48
77.16
76.84
46.90
31.52
20.00
13.75
f1 (ppm)

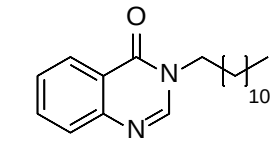
10j


O
N
N
10k
f1 (ppm)

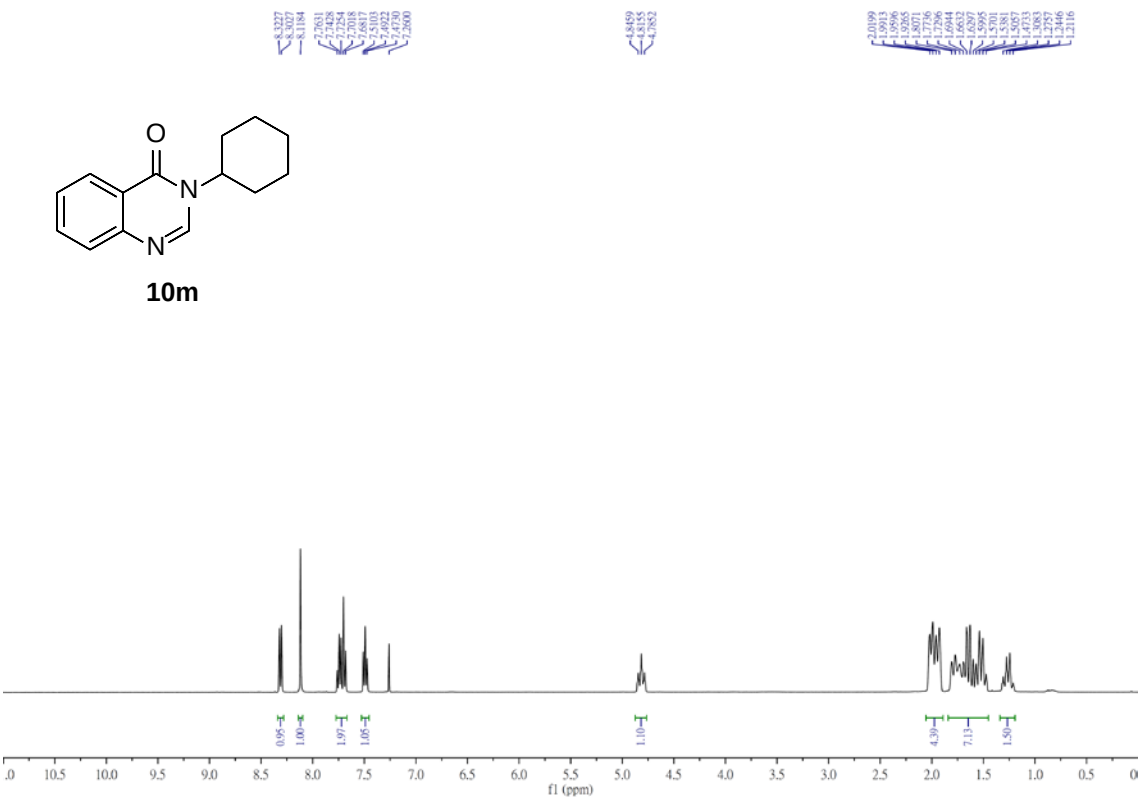

O
N
N
10k
f1 (ppm)

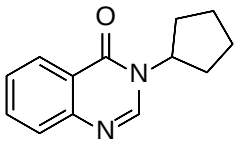


10l

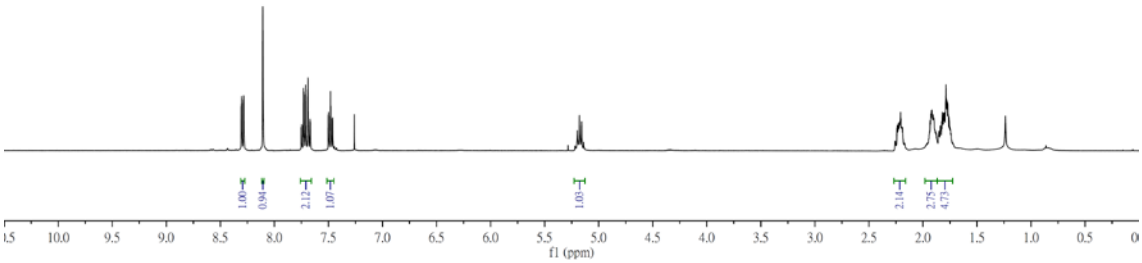


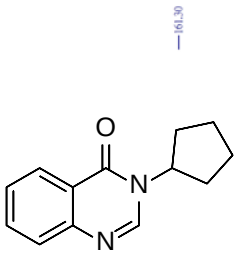
10l


O
N
N
10m
f1 (ppm)
O
N
N
10m
f1 (ppm)



10n





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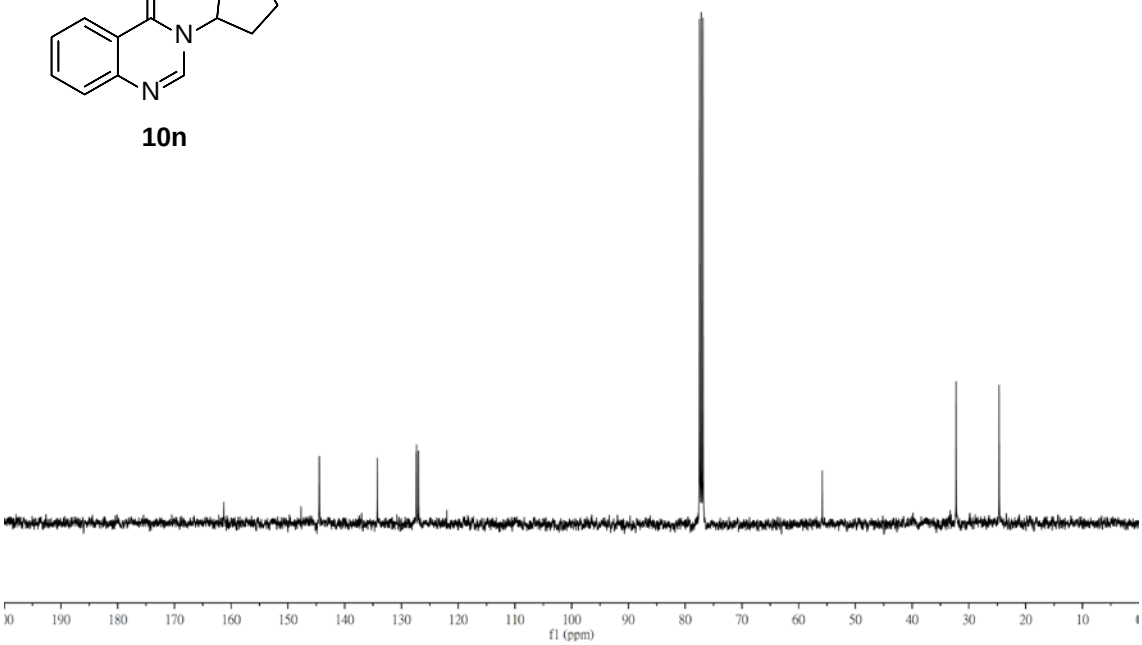

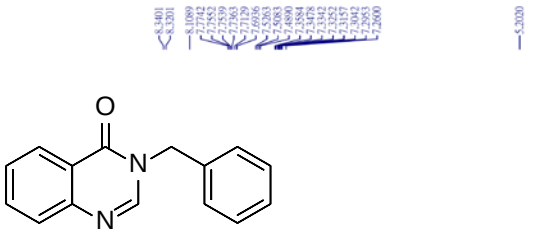

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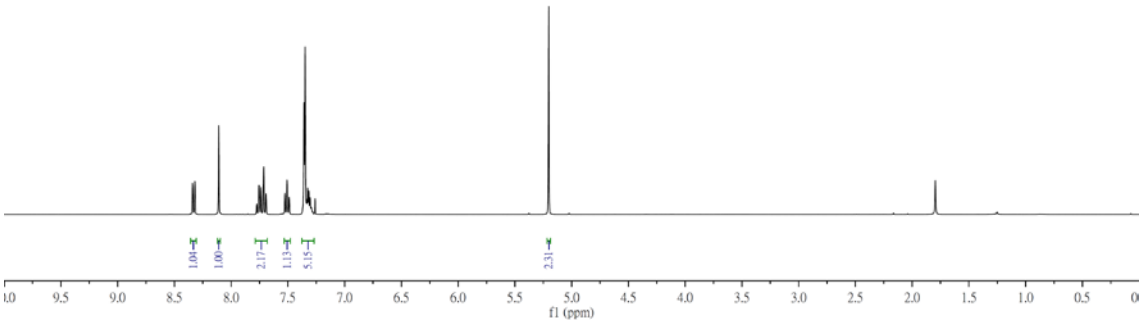

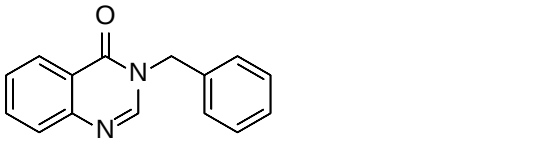

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