Supporting Information

Iron-Catalyzed C-O Bond Functionalization of Butyrolactam Derivatives with Various *N*-/*C*-Nucleophiles

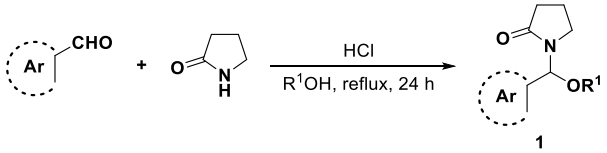
Danhua Ge, Mao-Ling Wang, Xin Wang, and Xue-Qiang Chu*

Table of Contents

1. General information

Unless otherwise stated, all reagents were purchased from commercial suppliers and used without further purification. All reactions were carried out under air atmosphere using undistilled solvent. Melting points were recorded on an Electrothermal digital melting point apparatus. IR spectra were recorded on a FT-IR spectrophotometer using KBr optics. ^{1}H, ^{19}F, and ^{13}C NMR spectra were recorded in $CDCl_3$ on Bruker Avance or Joel 400 MHz spectrometers. The chemical shifts (δ) are reported in ppm and coupling constants (J) in Hz. High resolution mass spectra (HRMS) were obtained using a commercial apparatus (ESI Source). Column chromatography was generally performed on silica gel (300-400 mesh) and reactions were monitored by thin layer chromatography (TLC) using UV light to visualize the course of the reactions.

2. General procedures for the synthesis of butyrolactam derivatives



1

According to Ji's reported method,[1-2] a solution of aldehyde (10 mmol) and pyrrolidin-2-one (2.55g, 30 mmol) in alcohol (40 mL) and 0.5N hydrochloric acid (40 mL) was stirred under nitrogen atmosphere (by 3 times' vacuum evacuation/N_2 backfill cycles) at room temperature for 24 h. The reaction was then quenched by saturated NH_4Cl solution (100 mL) and extracted with EtOAc (50 mL x 3). The organic layer was washed with saturated brine twice, dried over $MgSO_4$, filtered, and concentrated under reduced pressure. The crude product was purified by flash column chromatography (300-400 mesh) using petroleum ether/ethyl acetate as eluent to afford the butyrolactam derivatives **1**.

Representative examples:

1-(Ethoxy(4-nitrophenyl)methyl)pyrrolidin-2-one (1a):

^{1}H NMR (300 MHz, Chloroform-*d*): δ = 8.26–8.11 (m, 2H), 7.66–7.52 (m, 2H), 6.43 (s, 1H), 3.75–3.52 (m, 2H), 3.38–3.26 (m, 1H), 2.91–2.80 (m, 1H), 2.62–2.39 (m, 2H), 2.11–1.89 (m, 2H), 1.31 (t, J = 7.0 Hz, 3H).

1-(Methoxy(4-nitrophenyl)methyl)pyrrolidin-2-one (1a-MeOH):

^{1}H NMR (300 MHz, Chloroform-*d*): δ = 8.26–8.16 (m, 2H), 7.65–7.47 (m, 2H), 6.33 (s, 1H), 3.50–3.37 (m, 3H), 3.35–3.23 (m, 1H), 2.90–2.76 (m, 1H), 2.63–2.39 (m, 2H), 2.14–1.83 (m, 2H).

1-(Isopropoxy(4-nitrophenyl)methyl)pyrrolidin-2-one (1a-^{i}PrOH):

^{1}H NMR (300 MHz, Chloroform-*d*): δ = 8.21 (d, J = 8.4 Hz, 2H), 7.58 (d, J = 8.6 Hz, 2H), 6.53 (s, 1H), 3.93–3.71 (m, 1H), 3.35 (q, J = 8.1 Hz, 1H), 2.96–2.77 (m, 1H), 2.49 (d, J = 8.5 Hz, 2H), 2.10–1.85 (m, 2H), 1.27 (d, J = 6.2 Hz, 6H).

1-(Ethoxy(3-nitrophenyl)methyl)pyrrolidin-2-one (1b):

^{1}H NMR (300 MHz, Chloroform-*d*): δ = 8.60 (d, J = 21.4 Hz, 2H), 7.77–7.65 (m, 1H), 7.31 (s), 6.42 (d, J = 4.7 Hz, 1H), 3.71–3.53 (m, 2H), 3.40–3.25 (m, 1H), 2.99–2.86 (m, 1H), 2.56–2.44 (m, 2H), 2.10–1.88 (m, 2H), 1.30 (t, J = 7.0 Hz, 3H).

1-(Ethoxy(2-nitrophenyl)methyl)pyrrolidin-2-one (1c):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.81–7.73 (m, 1H), 7.67 (d, *J* = 7.7 Hz, 1H), 7.63–7.55 (m, 1H), 7.48 (t, *J* = 7.2 Hz, 1H), 6.76 (s, 1H), 3.62 (q, *J* = 6.8 Hz, 2H), 3.35–3.23 (m, 1H), 3.00–2.86 (m, 1H), 2.54–2.43 (m, 2H), 2.10–1.90 (m, 2H), 1.24 (t, *J* = 7.0 Hz, 3H).

1-(Ethoxy(4-(methylsulfonyl)phenyl)methyl)pyrrolidin-2-one (1e):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.98–7.84 (m, 2H), 7.60 (t, *J* = 9.7 Hz, 2H), 6.43 (s, 1H), 3.70–3.53 (m, 2H), 3.37–3.26 (m, 1H), 3.06 (s, 3H), 2.92–2.79 (m, 1H), 2.56–2.42 (m, 2H), 2.09–1.93 (m, 2H), 1.31 (t, *J* = 7.0 Hz, 3H).

1-((3-Chlorophenyl)(ethoxy)methyl)pyrrolidin-2-one (1g):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 8.70–8.54 (m, 1H), 7.77–7.67 (m, 1H), 7.56 (d, *J* = 7.9 Hz, 1H), 7.23 (t, *J* = 6.2 Hz, 1H), 6.38 (s, 1H), 3.76–3.59 (m, 2H), 3.43–3.29 (m, 1H), 3.08–2.94 (m, 1H), 2.50 (t, *J* = 8.1 Hz, 2H), 2.09–1.92 (m, 2H), 1.31 (t, *J* = 7.0 Hz, 3H).

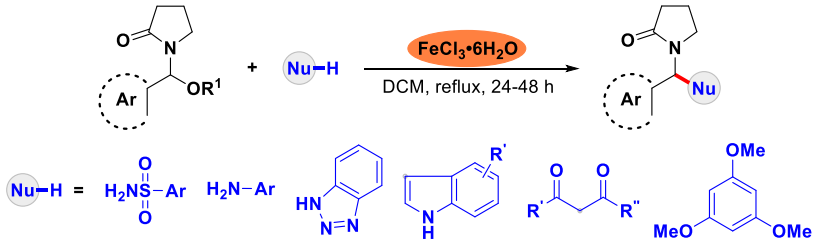
1-(Ethoxy(phenyl)methyl)pyrrolidin-2-one (1i):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.49–7.27 (m, 5H), 6.37 (s, 1H), 3.70–3.51 (m, 2H), 3.38–3.24 (m, 1H), 3.02–2.86 (m, 1H), 2.59–2.35 (m, 2H), 2.07–1.81 (m, 2H), 1.29 (t, *J* = 7.0 Hz, 3H).

1-((4-(Dimethylamino)phenyl)(ethoxy)methyl)pyrrolidin-2-one (1k):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.27 (d, *J* = 18.2 Hz, 2H), 6.70 (d, *J* = 8.2 Hz, 2H), 6.29 (s, 1H), 3.74–3.50 (m, 2H), 3.43–3.26 (m, 1H), 3.14–2.82 (m, 7H), 2.60–2.35 (m, 2H), 2.08–1.85 (m, 2H), 1.41–1.16 (m, 3H).

1-(Ethoxy(thiophen-2-yl)methyl)pyrrolidin-2-one (1n):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.32–7.20 (m, 1H), 6.98 (d, *J* = 3.2 Hz, 2H), 6.55 (s, 1H), 3.61 (q, *J* = 7.0 Hz, 2H), 3.42–3.32 (m, 1H), 3.26–3.15 (m, 1H), 2.55–2.39 (m, 2H), 2.11–1.89 (m, 2H), 1.28 (t, *J* = 7.0 Hz, 3H).

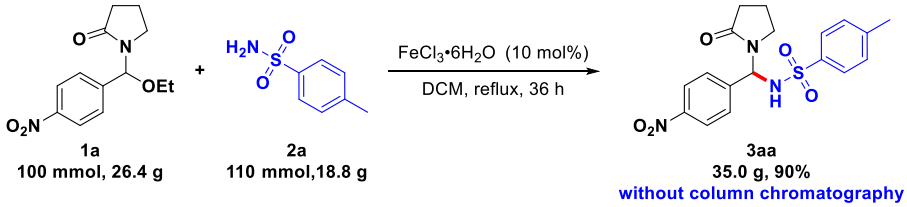
1-(Ethoxy(4-methoxyphenyl)methyl)pyrrolidin-2-one (1q):
^{1}H NMR (300 MHz, Chloroform-*d*): δ = 7.31 (d, *J* = 8.3 Hz, 2H), 6.88 (d, *J* = 8.4 Hz, 2H), 6.32 (s, 1H), 3.80 (s, 3H), 3.65–3.52 (m, 2H), 3.37–3.24 (m, 1H), 3.01–2.89 (m, 1H), 2.57–2.35 (m, 2H), 2.06–1.86 (m, 2H), 1.28 (t, *J* = 7.0 Hz, 3H).

3. General procedures for iron-catalyzed C-O bond functionalization of butyrolactam derivatives with *N*-/*C*-nucleophiles



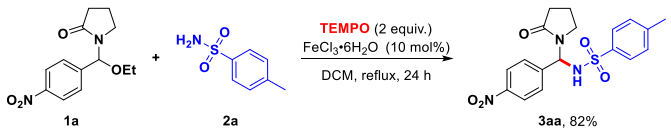
A solution of butyrolactam derivative **1** (1.2 mmol), *N*-/*C*-nucleophile (1.0 mmol), and $FeCl_3·6H_2O$ (16 mg, 0.1 mmol) in DCM (1.0 mL) was stirred under air at 40 °C for 24-48 h. Upon completion of the reaction (indicated by TLC), the free-flowing solid was filtered and washed with ethyl acetate/petroleum ether (1:4) to afford the desired products as pale white solids. The product thus obtained was recrystallized from ethanol or ethyl acetate to get pure compounds as white crystals. Once the final product can't be separated by filtration, the solvent of the resulting mixture was removed with the aid of a rotary evaporator, the pure product was obtained after purification of the residue by column chromatography (silica gel, ethyl acetate/petroleum ether).

4. General procedure for the large-scale synthesis



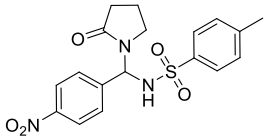
1a (100 mmol, 26.4 g), 4-methylbenzenesulfonamide **2a** (110 mmol, 18.8 g), and $FeCl_3·6H_2O$ (1.62 g, 10 mmol) in 100 mL of CH_2Cl_2 was stirred at 40 °C for 36 h. The free-flowing solid was filtered and washed with ethyl acetate/petroleum ether (1:4) to afford the desired product as pale white solid (35.0 g, 90% isolated yield).

5. Mechanistic study

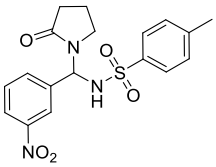


A solution of butyrolactam derivative **1a** (317 mg, 1.2 mmol), 4-methylbenzenesulfonamide (171 mg, 1.0 mmol), TEMPO (313 mg, 2 mmol), and $FeCl_3·6H_2O$ (16 mg, 0.1 mmol) in DCM (1.0 mL) was stirred under air at 40 °C for 24 h. Upon completion of the reaction (indicated by TLC), the free-flowing solid was filtered and washed with ethyl acetate/petroleum ether (1:4) to afford the desired products as pale white solids. The product thus obtained was recrystallized from ethanol or ethyl acetate to get pure compounds as white crystals (319 mg, 82% yield).

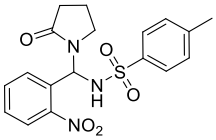
6. Characterization data for products



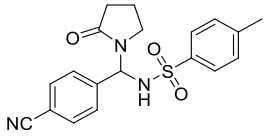
4-Methyl-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3aa): White solid. Mp = 223.7-226.5 °C. IR (KBr) ν = 3097, 2936, 2896, 1662, 1517, 1470, 1339, 1159, 1092, 702, 671, 565 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.22 (d, *J* = 10.3 Hz, 1H), 8.27 (d, *J* = 8.7 Hz, 2H), 7.66 (d, *J* = 8.1 Hz, 2H), 7.55 (d, *J* = 8.4 Hz, 2H), 7.42 (d, *J* = 8.0 Hz, 2H), 6.57 (d, *J* = 10.2 Hz, 1H), 2.85–2.77 (m, 1H), 2.74–2.66 (m, 1H), 2.40 (s, 3H), 2.21–2.07 (m, 1H), 1.71–1.58 (m, 2H), 1.12–1.00 (m, 1H). ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.8, 147.3, 143.7, 143.2, 137.7, 129.4, 127.6, 126.4, 123.9, 61.5, 40.6, 29.8, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{20}N_3O_5S$ $[M+H]^+$ 390.1124, Found 390.1117.



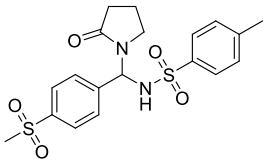
4-Methyl-*N*-((3-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ba): White solid. Mp = 192.2-193.8 °C. IR (KBr) ν = 3140, 2964, 2903, 1647, 1530, 1469, 1436, 1338, 1163, 1018, 823, 735, 702 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.27 (d, *J* = 10.3 Hz, 1H), 8.22 (d, *J* = 7.8 Hz, 1H), 8.14 (s, 1H), 7.73–7.63 (m, 4H), 7.41 (d, *J* = 7.9 Hz, 2H), 6.58 (d, *J* = 10.3 Hz, 1H), 2.86–2.77 (m, 1H), 2.74–2.65 (m, 1H), 2.40 (s, 3H), 2.19–2.07 (m, 1H), 1.07–1.58 (m, 2H), 1.12–0.99 (m, 1H)ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.9, 148.0, 143.2, 138.7, 137.7, 132.8, 130.6, 129.4, 126.4, 123.3, 120.8, 61.3, 40.5, 29.8, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{20}N_3O_5S$ $[M+H]^+$ 390.1124, Found 390.1118.



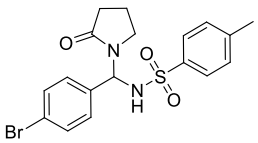
4-Methyl-*N*-((2-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ca): White solid. Mp = 165.5-167.9 °C. IR (KBr) ν = 3201, 2912, 1669, 1532, 1418, 1352, 1286, 1168, 921, 814, 784, 685, 587 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.27 (d, *J* = 10.3 Hz, 1H), 8.22 (d, *J* = 7.8 Hz, 1H), 8.14 (s, 1H), 7.73–7.63 (m, 4H), 7.41 (d, *J* = 7.9 Hz, 2H), 6.58 (d, *J* = 10.3 Hz, 1H), 2.86–2.77 (m, 1H), 2.74–2.65 (m, 1H), 2.40 (s, 3H), 2.19–2.07 (m, 1H), 1.07–1.58 (m, 2H), 1.12–0.99 (m, 1H)ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.5, 148,7, 143.1, 137.9, 132.5, 129.9, 129.6, 129.4, 128.2, 126.5, 124.4, 58.8, 41.8, 29.8, 21.0, 17.0 ppm. HRMS(EI) calcd for $C_{18}H_{20}N_3O_5S$ $[M+H]^+$ 390.1124, Found 390.1125.



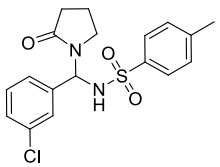
***N*-((4-Cyanophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3da):** White solid. Mp = 174.5-176.5 °C. IR (KBr) ν = 3262, 3207, 2230, 1700, 1595, 1335, 1264, 1154, 1083, 1010, 920, 860, 812, 689, 589 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.14 (d, *J* = 7.2 Hz, 1H), 7.88 (d, *J* = 8.1 Hz, 2H), 7.64 (d, *J* = 8.0 Hz, 2H), 7.43 (dd, *J* = 19.2, 8.0 Hz, 4H), 6.52 (d, *J* = 6.5 Hz, 1H), 2.85-2.75 (m, 1H), 2.74-2.65 (m, 1H), 2.21-2.07 (m, 1H), 1.71–1.58 (m, 2H), 1.14–0.97 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.9, 143.2, 141.8, 137.8, 132.8, 129.4, 127.2, 126.4, 118.5, 111.1, 61.6, 40.6, 29.8, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{19}H_{20}N_3O_3S$ $[M+H]^+$ 370.1225, Found 370.1235.



4-Methyl-*N*-((4-(methylsulfonyl)phenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ea): White solid. Mp = 172.4-173.9 °C. IR (KBr) ν = 3618, 3164, 2928, 1655, 1469, 1343, 1307, 1157, 1075, 959, 916, 818, 780, 758 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.15 (s, 1H), 7.96 (d, *J* = 8.2 Hz, 2H), 7.65 (d, *J* = 8.0 Hz, 2H), 7.53 (d, *J* = 8.1 Hz, 2H), 7.41 (d, *J* = 7.9 Hz, 2H), 6.55 (s, 1H), 3.22 (s, 3H), 2.85–2.77 (m, 1H), 2.75–2.67 (m, 1H), 2.40 (s, 3H), 2.19–2.07 (m, 1H), 171–1.59 (m, 2H), 1.12–1.00 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.8, 143.2, 142.1, 140.7, 137.8, 129.4, 127.6, 127.2, 126.4, 61.6, 43.5, 40.7, 29.9, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{19}H_{23}N_2O_5S_2$ $[M+H]^+$ 423.1048, Found 423.1048.

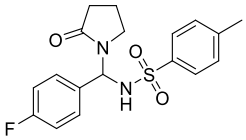


***N*-((4-Bromophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (2fa):** White solid. Mp = 182.7-184.7 °C. IR (KBr) ν = 3107, 2949, 2898, 1657, 1469, 1337, 1286, 1162, 1084, 1009, 815, 665, 562 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.03 (d, *J* = 10.5 Hz, 1H), 7.62 (dd, *J* = 14.4, 8.2 Hz, 4H), 7.40 (d, *J* = 7.9 Hz, 2H), 7.21 (d, *J* = 8.5 Hz, 2H), 6.44 (d, *J* = 10.5 Hz, 1H), 2.81–2.74 (m, 1H), 2.73–2.65 (m, 1H), 2.39 (s, 3H), 2.16–2.04 (m, 1H), 1.68–1.54 (m, 2H), 1.08–0.96 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.6, 143.1, 137.9, 135.9, 131.6, 128.3, 126.4, 121.4, 61.4, 40.5, 29.9, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{20}BrN_2O_3S$ $[M+H]^+$ 423.0378, Found 423.0378.

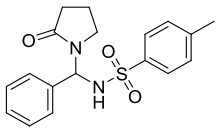


***N*-((3-Chlorophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ga):** White solid. Mp = 171.3-172.5 °C. IR (KBr) ν = 3155, 2893, 1669, 1459, 1429, 1337, 1284, 1159, 1083, 890, 812, 693, 653 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 9.06 (d, *J* = 11.0 Hz, 1H), 7.64 (d, *J* = 7.4 Hz, 2H), 7.44–7.38 (m, 4H), 7.30 (s, 1H), 7.20 (d, *J* = 6.7 Hz, 1H), 6.47 (d, *J* = 10.2 Hz, 1H), 2.83–2.76 (m, 1H), 2.75–2.67 (m, 1H), 2.40 (s, 3H), 2.18–2.07 (m, 1H), 1.70–1.57 (m, 2H), 1.10–0.97 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.7, 143.1, 138.9, 137.8, 133.4, 129.4,

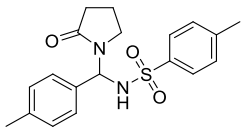
128.2, 126.3, 125.8, 124.8, 61.4, 40.6, 29.9, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{20}ClN_2O_3S$ $[M+H]^+$ 379.0883, Found 379.0888.



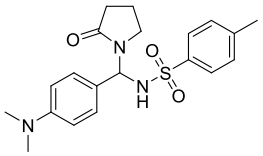
***N*-((4-Fluorophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ha):** White solid. Mp = 176.4-179.3 °C. IR (KBr) ν = 3194, 2937, 1642, 1595, 1412, 1379,1212, 1021, 911, 834, 812, 635 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): δ = 9.00 (d, *J* = 10.1 Hz, 1H), 7.64 (d, *J* = 8.1 Hz, 2H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.33–7.17 (m, 4H), 6.47 (d, *J* = 10.0 Hz, 1H), 2.83–2.67 (m, 2H), 2.40 (s, 3H), 2.18–2.04 (m, 1H), 1.70–1.54 (m, 2H), 1.11–0.94 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.7, 161.8 (d, *J* = 242.0 Hz), 143.1, 137.9, 132.6, 129.4, 128.2 (d, *J* = 7.7 Hz), 126.4, 115.6 (d, *J* = 21.6 Hz), 61.4, 41.4, 30.0, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{20}FN_2O_3S$ $[M+H]^+$ 363.1179, Found 363.1184.



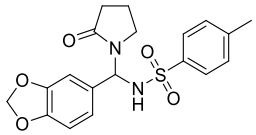
4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(phenyl)methyl)benzenesulfonamide (3ia): White solid. Mp = 169.3-171.5 °C. IR (KBr) ν = 3194, 2900, 1658, 1464, 1332, 1162, 1065, 1020, 815, 711, 687, 567 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): δ = 8.97 (d, *J* = 10.0 Hz, 1H), 7.65 (d, *J* = 8.1 Hz, 2H), 7.46–7.30 (m, 5H), 7.26 (d, *J* = 7.2 Hz, 2H), 6.49 (d, *J* = 9.9 Hz, 1H), 2.85–2.66 (m, 2H), 2.40 (s, 3H), 2.20–2.03 (m, 1H), 1.70–1.55 (m, 2H), 1.10–0.92 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 173.6, 143.0, 138.00, 136.4, 129.3, 128.7, 128.2, 126.4, 125.9, 61.8, 41.3, 30.0, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{18}H_{21}N_2O_3S$ $[M+H]^+$ 345.1273, Found 345.1271.



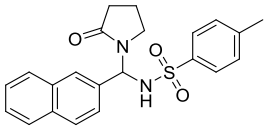
4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(*p*-tolyl)methyl)benzenesulfonamide (3ja): White solid. Mp = 153.7-155.7 °C. IR (KBr) ν = 3142, 1655, 1462, 1416, 1341, 1280, 1158, 1088, 912, 812, 681, 551 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 8.91 (d, *J* = 10.4 Hz, 1H), 7.64 (d, *J* = 8.2 Hz, 2H), 7.39 (d, *J* = 8.1 Hz, 2H), 7.20 (d, *J* = 8.0 Hz, 2H), 7.14 (d, *J* = 8.1 Hz, 2H), 6.45 (d, *J* = 10.5 Hz, 1H), 2.81–2.74 (m, 2H), 2.73–2.66 (m, 1H), 2.39 (s, 3H), 2.29 (s, 3H), 2.14–2.04 (m, 1H), 1.66–1.56 (m, 2H), 1.05–0.93 (m, 1H)ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 173.5, 142.9, 139.1, 137.4, 133.5, 129.3, 129.2, 126.4, 125.9, 61.7, 40.5, 30.0, 21.0, 20.6, 16.6 ppm. HRMS(EI) calcd for $C_{19}H_{23}N_2O_3S$ $[M+H]^+$ 359.1429, Found 359.1434.



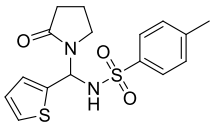
***N*-((4-(Dimethylamino)phenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ka):** Pare Yellow solid. Mp = 173.6-175.8 °C. IR (KBr) ν = 3164, 2893, 1653, 1532, 1464, 1333, 1282, 1167, 1086, 921, 812, 677, 665 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): δ = 8.81 (s, 1H), 7.78 (dd, *J* = 11.9, 8.6 Hz, 4H), 7.41 (d, *J* = 8.1 Hz, 2H), 6.79 (d, *J* = 8.9 Hz, 2H), 3.25–3.17 (m, 2H), 3.08 (s, 6H), 2.38 (s, 3H), 2.13–2.01 (m, 2H), 2.01–1.89 (m, 2H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 177.3, 169.2, 155.0, 143.6, 136.8, 127.0, 118.8, 111.6, 41.4, 29.9, 21.1, 20.4 ppm. HRMS(EI) calcd for $C_{20}H_{26}N_3O_3S$ $[M+H]^+$ 388.1695, Found 388.169.



***N*-(Benzo[*d*][1,3]dioxol-5-yl(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3la):** White solid. Mp = 166.8-167.5 °C. IR (KBr) ν = 3115, 2884, 1663, 1507, 1463, 1336, 1253, 1159, 1089, 914, 815, 691, 666 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6): δ = 8.89 (d, *J* = 6.9 Hz, 1H), 7.63 (d, *J* = 7.9 Hz, 2H), 7.39 (d, *J* = 8.0 Hz, 2H), 6.91 (d, *J* = 8.6 Hz, 1H), 6.74 (d, *J* = 6.7 Hz, 2H), 6.38 (d, *J* = 8.1 Hz, 1H), 6.02 (s, 2H), 2.84–2.67 (m, 2H), 2.39 (s, 3H), 2.16–2.03 (m, 1H), 1.67–1.55 (m, 2H), 1.08–0.92 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.5, 131.0, 130.0, 129.3, 127.5, 126.4, 119.5, 108.3, 106.3, 102.6, 101.3, 61.7, 41.4, 29.9, 21.0, 20.4, 16.7 ppm. HRMS(EI) calcd for $C_{19}H_{21}N_2O_5S$ $[M+H]^+$ 389.1171, Found 389.1178.

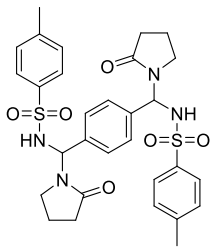


4-Methyl-*N*-(naphthalen-2-yl(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ma): White solid. Mp = 184.5-186.2 °C. IR (KBr) ν = 3434, 3161, 1167, 1459, 1337, 1285, 1166, 1087, 811, 671, 549 cm^{-1}. ^1H NMR (300 MHz, DMSO-d_6): δ = 9.11 (d, *J* = 10.3 Hz, 1H), 7.94 (d, *J* = 8.2 Hz, 3H), 7.83 (s, 1H), 7.69 (d, *J* = 8.0 Hz, 2H), 7.59–7.49 (m, 2H), 7.41 (d, *J* = 7.9 Hz, 2H), 7.34 (d, *J* = 8.5 Hz, 1H), 6.66 (d, *J* = 10.2 Hz, 1H), 2.90–2.81 (m, 1H), 2.80–2.69 (m, 1H), 2.40 (s, 3H), 2.21–2.08 (m, 1H), 1.72–1.58 (m, 2H), 1.11–0.96 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d_6): δ = 173.8, 143.1, 138.1, 134.0, 132.6, 132.5, 129.4, 128.6, 128.1, 127.5, 126.5, 125.6, 124.9, 123.9, 122.3, 62.0, 41.4, 30.1, 21.0, 16.8 ppm. HRMS(EI) calcd for C22H23N2O3S $[M+H]^+$ 395.1429, Found 395.1428.

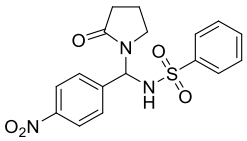


4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(thiophen-2-yl)methyl)benzenesulfonamide (3na): White solid. Mp = 175.3-176.5 °C. IR (KBr) ν = 3158, 3113, 1661, 1465, 1334, 1167, 1075, 917, 815, 724,

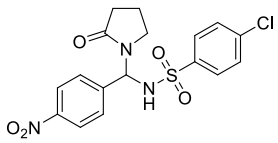
663, 546 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 9.18 (d, *J* = 9.6 Hz, 1H), 7.63 (d, *J* = 8.1 Hz, 2H), 7.53 (d, *J* = 4.8 Hz, 1H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.05–6.97 (m, 1H), 6.93 (d, *J* = 3.3 Hz, 1H), 6.60 (d, *J* = 9.3 Hz, 1H), 2.96–2.85 (m, 1H), 2.81–2.71 (m, 1H), 2.39 (s, 3H), 2.18–2.05 (m, 1H), 1.70–1.55 (m, 2H), 1.09–0.94 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): *δ* = 173.3, 143.2, 140.2, 137.8, 130.1, 129.4, 127.3, 126.4, 125.3, 59.4, 41.4, 29.9, 21.0, 16.8 ppm. HRMS(EI) calcd for $C_{16}H_{19}N_2O_3S_2$ [M+H]$^+$ 351.0837, Found 351.0843.



***N*,*N*'-(1,4-Phenylenebis((2-oxopyrrolidin-1-yl)methylene))bis(4-methylbenzenesulfonamide) (3pa):** White solid. Mp >300 °C. IR (KBr) *ν* = 3135, 2900, 1665, 1597, 1460, 1417, 1339, 1287, 1157, 1089, 921, 812, 774, 685 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 8.99 (d, *J* = 10.3 Hz, 2H), 7.64 (d, *J* = 8.2 Hz, 4H), 7.40 (d, *J* = 8.1 Hz, 4H), 7.28 (s, 4H), 6.47 (t, *J* = 5.1 Hz, 2H), 2.82–2.74 (m, 2H), 2.73–2.64 (m, 2H), 2.39 (s, 6H), 2.16–2.04 (m, 2H), 1.67–1.56 (m, 4H), 1.08–0.95 (m, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): *δ* = 173.6, 143.0, 138.0, 136.5, 129.3, 126.5, 126.4, 61.6, 40.6, 29.9, 21.0, 16.7 ppm. HRMS(EI) calcd for $C_{30}H_{35}N_4O_6S_2$ [M+H]$^+$ 611.1998, Found 611.1995.

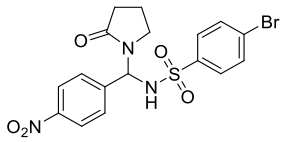


***N*-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ab):** White solid. Mp = 208.3-210.2 °C. IR (KBr) *ν* = 3166, 2896, 1653, 1518, 1352, 1287, 1156, 1086, 1022, 913, 867, 722, 690, 606 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 9.33 (d, *J* = 10.3 Hz, 1H), 8.27 (d, *J* = 8.6 Hz, 2H), 7.78 (d, *J* = 7.3 Hz, 2H), 7.73–7.53 (m, 5H), 6.60 (d, *J* = 10.3 Hz, 1H), 2.83–2.65 (m, 2H), 2.20–2.04 (m, 1H), 1.70–1.52 (m, 2H), 1.08–0.92 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): *δ* = 173.8, 147.4, 143.6, 140.6, 132.8, 129.1, 127.6, 126.3, 124.0, 61.5, 40.6, 29.8, 16.7 ppm. HRMS(EI) calcd for $C_{17}H_{18}N_3O_5S$ [M+H]$^+$ 376.0967, Found 376.0964.

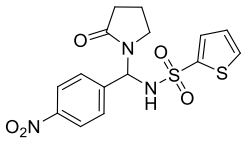


4-Chloro-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ac): White solid. Mp = 196.8-198.0 °C. IR (KBr) *ν* = 3098, 2893, 1662, 1517, 1465, 1348, 1279, 1160, 1093, 826, 755, 624 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 9.46 (d, *J* = 7.9 Hz, 1H), 8.27 (d, *J* = 8.8 Hz, 2H), 7.77 (d, *J* = 8.7 Hz, 2H), 7.72 (d, *J* = 8.6 Hz, 2H), 7.56 (d, *J* = 8.8 Hz, 2H), 6.58 (d, *J* = 6.6 Hz, 1H), 2.90–2.82 (m, 1H), 2.77–2.69 (m, 1H), 2.23–2.13 (m, 1H), 1.76–1.63 (m, 2H), 1.20–1.09 (m, 1H).ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): *δ* = 173.9, 147.4, 143.4, 139.5, 137.8, 129.2, 128.3, 127.6, 124.0, 61.5, 40.7, 29.7, 16.8 ppm. HRMS(EI) calcd for $C_{17}H_{17}ClN_3O_5S$ [M+H]$^+$ 410.0577,

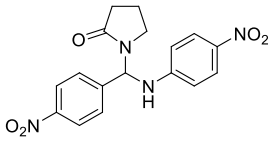
Found 410.0572.



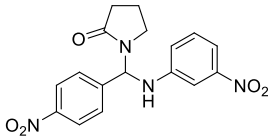
4-Bromo-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ad): White solid. Mp = 209.5-210.1 °C. IR (KBr) ν = 3095, 2889, 1655, 1514, 1466, 1341, 1278, 1166, 1090, 917, 870, 743, 614 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 9.45 (d, *J* = 10.2 Hz, 1H), 8.27 (d, *J* = 8.8 Hz, 2H), 7.86 (d, *J* = 8.6 Hz, 2H), 7.69 (d, *J* = 8.6 Hz, 2H), 7.56 (d, *J* = 8.6 Hz, 2H), 6.58 (d, *J* = 10.2 Hz, 1H), 2.90–2.81 (m, 1H), 2.77–2.69 (m, 1H), 2.24–2.13 (m, 1H), 1.76–1.62 (m, 2H), 1.20–1.07 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 173.9, 147.4, 143.3, 139.8, 132.2, 128.4, 127.6, 126.7, 124.0, 61.5, 40.7, 29.7, 16.8 ppm. HRMS(EI) calcd for $C_{17}H_{17}BrN_3O_5S$ $[M+H]^+$ 454.0072, Found 454.0078.



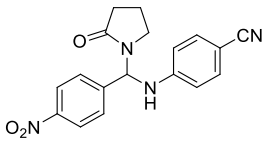
***N*-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)thiophene-2-sulfonamide (3af):** White solid. Mp = 197.3-198.1 °C. IR (KBr) ν = 3146, 2914, 1638, 1514, 1457, 1341, 1285, 1151, 1018, 909, 856, 729 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 9.53 (d, *J* = 10.1 Hz, 1H), 8.27 (d, *J* = 8.8 Hz, 2H), 7.99 (dd, *J* = 5.0, 1.3 Hz, 1H), 7.56 (d, *J* = 8.5 Hz, 3H), 7.53 (dd, *J* = 3.7, 1.4 Hz, 1H), 7.20 (dd, *J* = 5.0, 3.8 Hz, 1H), 6.63 (d, *J* = 10.0 Hz, 1H), 3.03–2.94 (m, 1H), 2.83–2.75 (m, 1H), 2.26–2.17 (m, 1H), 1.91–1.81 (m, 1H), 1.80–1.71 (m, 1H), 1.40–1.28 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 174.0, 147.4, 143.6, 141.3, 133.4, 132.1, 127.6, 127.5, 124.0, 61.7, 41.0, 29.9, 17.1 ppm. HRMS(EI) calcd for $C_{15}H_{16}N_3O_5S_2$ $[M+H]^+$ 382.0531, Found 382.0536.



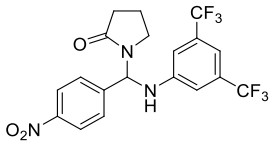
1-((4-Nitrophenyl)(4-nitrophenylamino)methyl)pyrrolidin-2-one (3ag): Yellow solid. Mp = 273.8-275.8 °C. IR (KBr) ν = 3252, 3082, 1649, 1593, 1584, 1431, 1318, 1113, 842, 751, 705 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 8.31 (d, *J* = 8.6 Hz, 2H), 8.09 (d, *J* = 9.0 Hz, 3H), 7.72 (d, *J* = 8.6 Hz, 2H), 6.86 (d, *J* = 9.1 Hz, 2H), 6.76 (d, *J* = 8.4 Hz, 1H), 3.21–3.07 (m, 2H), 2.45–2.29 (m, 2H), 2.04–1.85 (m, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 175.1, 152.2, 147.5, 144.2, 138.0, 128.1, 126.1, 123.9, 112.5, 61.3, 41.7, 30.4, 17.5 ppm. HRMS(EI) calcd for $C_{17}H_{17}N_4O_5$ $[M+H]^+$ 357.1199, Found 357.1204.



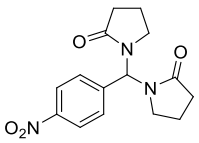
1-((4-Nitrophenyl)(3-nitrophenylamino)methyl)pyrrolidin-2-one (3ah): Yellow solid. Mp = 231.6-233.4 ºC. IR (KBr) ν = 3276, 1655, 1533, 1420, 1356, 1285, 1138, 1109, 733 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 8.31 (d, *J* = 8.6 Hz, 2H), 7.74 (d, *J* = 8.6 Hz, 2H), 7.64 (s, 1H), 7.56–7.42 (m, 3H), 7.17 (d, *J* = 8.2 Hz, 1H), 6.72 (d, *J* = 9.0 Hz, 1H), 3.18–3.05 (m, 2H), 2.45–2.26 (m, 2H), 2.02–1.91 (m, 1H), 1.90–1.80 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d$_6$): δ = 175.2, 151.0, 148.8, 147.1, 144.6, 130.4, 128.1, 123.8, 119.4, 112.2, 107.1, 61.5, 41.4, 30.5, 17.5 ppm. HRMS(EI) calcd for $C_{17}H_{17}N_4O_5$ $[M+H]^+$ 357.1199, Found 357.1208.



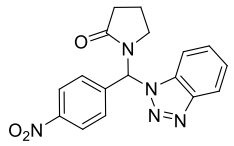
4-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methylamino)benzonitrile (3ai): Pare Yellow solid. Mp = 258.4-260.5 ºC. IR (KBr) ν = 3266, 3172, 3087, 2221, 1653, 1594, 1430, 1335, 1289, 1181, 1102, 827, 779, 694, 588 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 8.30 (d, *J* = 8.6 Hz, 2H), 7.74-7.65 (m, 3H), 7.59 (d, *J* = 8.4 Hz, 2H), 6.85 (d, *J* = 8.5 Hz, 2H), 6.68 (d, *J* = 8.6 Hz, 1H), 3.18–3.06 (m, 2H), 2.44–2.28 (m, 2H), 2.03–1.80 (m, 2H) ppm. ^{13}C NMR (75 MHz, DMSO-d$_6$): δ = 175.4, 150.0, 147.6, 144.6, 133.8, 128.2, 124.0, 120.2, 113.5, 98.9, 61.4, 41.7, 30.6, 17.7 ppm. HRMS(EI) calcd for $C_{18}H_{17}N_4O_3$ $[M+H]^+$ 337.1301, Found 337.1309.



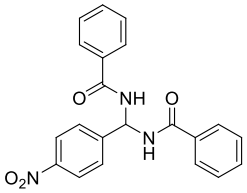
1-((3,5-Bis(trifluoromethyl)phenylamino)(4-nitrophenyl)methyl)pyrrolidin-2-one (3aj): White solid. Mp = 195.1-196.7 ºC. IR (KBr) ν = 3305, 2920, 1667, 1623, 1519, 1477, 1391, 1350, 1276, 1128, 846, 704, 682 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 8.32 (d, *J* = 8.8 Hz, 2H), 7.75 (d, *J* = 8.8 Hz, 3H), 7.36 (s, 2H), 7.29 (s, 1H), 6.79 (d, *J* = 8.9 Hz, 1H), 3.15–3.04 (m, 2H), 2.46–2.37 (m, 1H), 2.32–2.22 (m, 1H), 2.02–1.92 (m, 1H), 1.91–1.82 (m, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 175.3, 147.4, 145.8 (d, *J* = 325.4 Hz), 131.1 (q, *J* = 31.9 Hz), 128.0, 123.8, 123.5 (d, *J* = 271.1 Hz), 113.1, 109.9, 61.1, 41.2, 30.3, 17.5 ppm. HRMS(EI) calcd for $C_{19}H_{16}F_6N_3O_3$ $[M+H]^+$ 448.1096, Found 448.1097.



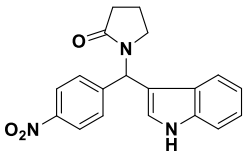
1,1'-((4-Nitrophenyl)methylene)dipyrrolidin-2-one (3ak): White soild. Mp = 160.6-162.4 ºC. IR (KBr) ν = 2975, 2937, 2879, 1696, 1511, 1407, 1344, 1266, 1209, 1103, 876, 702, 635, 530 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 8.23 (d, *J* = 8.7 Hz, 2H), 7.47 (d, *J* = 8.5 Hz, 2H), 6.88 (s, 1H), 3.32–3.27 (m, 4H), 2.42–2.30 (m, 4H), 2.09–1.97 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 174.9, 147.1, 143.4, 127.8, 123.9, 61.0, 44.7, 30.1, 18.1 ppm. HRMS(EI) calcd for $C_{15}H_{18}N_3O_4$ $[M+H]^+$ 304.1297, Found 304.1299.



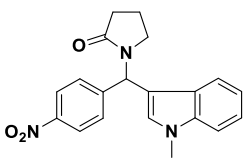
1-((1*H*-Benzo[*d*][1,2,3]triazol-1-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (3al): White solid. Mp = 162.6-164.8 °C. IR (KBr) ν = 3481, 2939, 1697, 1525, 1490, 1409, 1352, 1265, 1060, 834, 778, 750 cm^{-1}. ^{1}H NMR (400 MHz, DMSO-d$_6$): δ = 8.35 (s, 1H), 8.29 (d, *J* = 8.6 Hz, 2H), 8.16 (d, *J* = 8.3 Hz, 1H), 7.82 (d, *J* = 8.3 Hz, 1H), 7.61 (t, *J* = 7.6 Hz, 1H), 7.51–7.44 (m, 3H), 3.44–3.36 (m, 1H), 3.29–3.22 (m, 1H), 2.49–2.40 (m, 1H), 2.39–2.28 (m, 1H), 2.13-2.01 (m, 1H), 1.99–1.88 (m, 1H) ppm. ^{13}C NMR (75 MHz, DMSO-d$_6$): δ = 175.7, 147.8, 145.0, 141.5, 128.9, 128.4, 124.9, 124.1, 119.6, 111.0, 65.1, 44.0, 30.1, 17.8 ppm. HRMS(EI) calcd for $C_{17}H_{16}N_5O_3$ [M+H]$^+$ 338.1253, Found 338.1253.



***N*,*N*'-((4-Nitrophenyl)methylene)dibenzamide (3am):** White solid. Mp = 258.3-261.5 °C. IR (KBr) ν = 3271, 1650, 1548, 1506, 1345, 1227, 1055, 851, 794, 720, 695, 676 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 9.21 (d, *J* = 7.4 Hz, 2H), 8.26 (d, *J* = 8.6 Hz, 2H), 7.93 (d, *J* = 7.2 Hz, 4H), 7.75 (d, *J* = 8.6 Hz, 2H), 7.61–7.45 (m, 6H), 7.08 (t, *J* = 7.3 Hz, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 165.9, 147.6, 147.1, 133.5, 131.8, 128.4, 128.1, 127.6, 123.5 ppm. HRMS(EI) calcd for $C_{21}H_{18}N_3O_4$ [M+H]$^+$ 376.1297, Found 376.1299.

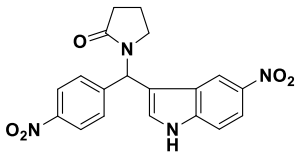


1-((1*H*-Indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5aa): Yellow solid. M.p. = 206–208 °C. IR (KBr) ν = 3437, 3189, 3079, 2978, 1653, 1518, 1348, 743 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.36 (s, 1H), 8.21 (d, *J* = 8.7 Hz, 2H), 7.46 (dd, *J* = 19.3, 8.3 Hz, 3H), 7.30 – 7.23 (m, 2H), 7.09 (t, *J* = 7.5 Hz, 1H), 6.99 – 6.88 (m, 2H), 3.38 (td, *J* = 9.4, 9.0, 4.7 Hz, 1H), 3.12 (q, *J* = 8.0 Hz, 1H), 2.51 (dq, *J* = 17.4, 9.0, 8.0 Hz, 2H), 2.14 – 1.88 (m, 2H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 175.3, 147.2, 146.9, 136.4, 128.1, 126.3, 124.6, 123.7, 122.8, 120.3, 119.2, 112.8, 111.6, 51.5, 44.6, 31.1, 18.1 ppm. HRMS: Calcd for $C_{19}H_{17}N_3O_3$: [M]$^+$ 335.1270; Found, 335.1275.

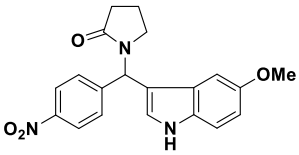


1-((1-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ab): Yellow solid. M.p. = 140.1–142.1 °C. IR (KBr) ν = 3442, 3056, 2938, 2877, 1681, 1518, 1343, 744 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.21 (d, *J* = 8.6 Hz, 2H), 7.49 (d, *J* = 8.4 Hz, 2H), 7.35 (d, *J* = 8.5 Hz, 1H),

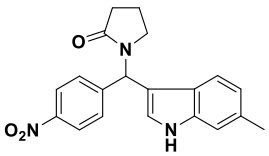
7.29 (s, 2H), 7.08 (t, J = 7.6 Hz, 1H), 6.90 (s, 1H), 6.81 (s, 1H), 3.78 (s, 3H), 3.38 (td, J = 9.1, 4.5 Hz, 1H), 3.12 (q, J = 8.3 Hz, 1H), 2.64 – 2.37 (m, 2H), 2.21 – 1.87 (m, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 175.2, 147.1, 147.0, 137.2, 129.1, 128.1, 126.8, 123.7, 122.4, 119.9, 119.4, 111.2, 109.6, 51.5, 44.6, 32.9, 31.1, 18.1 ppm. HRMS: Calcd for $C_{20}H_{19}N_3O_3$: $[M]^+$ 349.1426; Found, 349.1425.



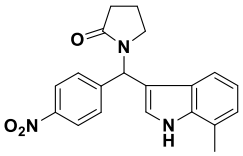
1-((5-Nitro-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ac): Yellow solid. M.p. = 103.3–105.3 °C. IR (KBr) ν = 3430, 3126, 2898, 1655, 1514, 1335, 1097, 737 cm^{-1}. ^{1}H NMR (300 MHz, DMSO-d_6): δ = 12.0 (s, 1H), 8.32 – 8.23 (m, 2H), 8.19 (d, J = 2.3 Hz, 1H), 8.04 (dd, J = 9.0, 2.3 Hz, 1H), 7.60 (dd, J = 8.8, 4.3 Hz, 3H), 7.45 (s, 1H), 6.85 (s, 1H), 3.29 (d, J = 9.0 Hz, 1H), 3.01 (q, J = 8.1 Hz, 1H), 2.41 – 2.36 (m, 2H), 2.03 – 1.84 (m, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 174.3, 146.9, 146.8, 140.8, 139.6, 139.4, 129.4, 129.2, 128.7, 125.5, 123.7, 117.1, 115.4, 114.4, 112.5, 50.5, 50.5, 44.1, 30.5, 17.7 ppm, HRMS: Calcd for $C_{19}H_{16}N_4O_5$: $[M]^+$ 380.1121; Found, 380.1121.



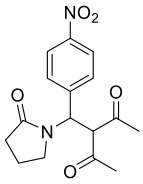
1-((5-Methoxy-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ad): Yellow solid. M.p. = 150.2–152.0 °C. IR (KBr) ν = 3441, 3188, 2893, 1658, 1516, 1347, 1047, 727 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.22 (d, J = 8.4 Hz, 3H), 7.50 (d, J = 8.3 Hz, 2H), 7.35 – 7.28 (m, 1H), 6.89 (s, 3H), 6.77 (s, 1H), 3.79 – 3.67 (m, 3H), 3.37 (s, 1H), 3.10 (d, J = 8.6 Hz, 1H), 2.53 (q, J = 8.4 Hz, 2H), 2.05 (d, J = 29.5 Hz, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 175.3, 154.5, 146.8, 131.4, 128.1, 126.9, 125.1, 123.7, 113.0, 112.8, 112.3, 100.8, 55.8, 51.5, 44.5, 31.1, 18.1 ppm. HRMS: Calcd for $C_{20}H_{19}N_3O_4$: $[M]^+$ 365.1376; Found, 365.1371.



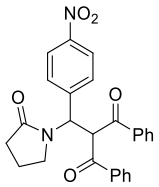
1-((6-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ae): Yellow solid. M.p. = 166.3–168.3 °C. IR (KBr) ν = 3437, 3221, 1657, 1518, 1349, 1111, 792, 725 cm^{-1}. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.20 (d, J = 7.9 Hz, 3H), 7.48 (d, J = 8.4 Hz, 2H), 7.21 (s, 1H), 7.16 (d, J = 8.2 Hz, 1H), 6.91 (d, J = 12.9 Hz, 3H), 3.36 (d, J = 10.3 Hz, 1H), 3.21 – 3.05 (m, 1H), 2.59 – 2.47 (m, 2H), 2.46 (s, 3H), 2.15 – 1.83 (m, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 175.2, 147.2, 146.9, 136.9, 132.8, 128.1, 124.2, 124.0, 123.7, 122.1, 118.9, 112.7, 111.5, 51.6, 44.6, 31.1, 21.6, 18.1 ppm. HRMS: Calcd for $C_{20}H_{19}N_3O_3$: $[M]^+$ 349.1426; Found, 349.1422.



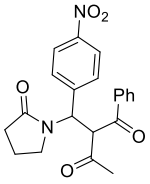
1-((7-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5af): White solid. M.p. = 123.9–125.6 °C. IR (KBr) ν = 3448, 3225, 2983, 1646, 1517, 1346, 1129, 826 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$): δ = 11.21 (s, 1H), 8.23 (d, J = 8.5 Hz, 2H), 7.54 (d, J = 8.4 Hz, 2H), 7.09 (s, 1H), 6.99 (d, J = 7.4 Hz, 1H), 6.95 – 6.83 (m, 2H), 6.69 (s, 1H), 2.98 (q, J = 8.0 Hz, 1H), 2.75 (t, J = 6.5 Hz, 1H), 2.41 – 2.34 (m, 2H), 2.03 – 1.85 (m, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): 174.8, 148.6, 147.3, 145.6, 129.1, 126.5, 124.3, 122.7, 121.8, 120.1, 116.6, 112.5, 51.8, 44.9, 31.2, 18.3, 17.4 ppm. HRMS: Calcd for $C_{20}H_{19}N_3O_3$: $[M]^+$ 349.1426; Found, 349.1427.



3-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)pentane-2,4-dione (7aa): White solid. M.p. = 123.2-124.2 °C. IR (KBr) ν = 3409, 3111, 2966, 2866, 1687, 1519, 1419, 1354, 1265, 1168, 807,704 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.29 – 8.09 (m, 2H), 7.64 – 7.47 (m, 2H), 5.69 (d, J = 11.9 Hz, 1H), 5.19 (d, J = 11.9 Hz, 1H), 3.34 (ddd, J = 9.2, 8.0, 5.6 Hz, 1H), 3.22 (dt, J = 9.2, 7.4 Hz, 1H), 2.36 – 2.28 (m, 5H), 2.12 (s, 3H), 1.98 – 1.89 (m, 2H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 200.5, 200.1, 175.5, 147.6, 144.1, 128.9, 124.1, 69.5, 55.5, 45.9, 31.2, 31.1, 28.3, 18.1 ppm. HRMS: Calcd for $C_{16}H_{18}N_2O_5$: [M]+ 318.1216; Found, 318.1302.

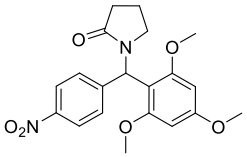


2-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)-1,3-diphenylpropane-1,3-dione (7ab): White solid. M.p. = 86.4-87.1 °C. IR (KBr) ν = 3441, 3054, 2920, 1670, 1520, 1278, 1104, 1005, 857, 695 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ= 8.59 (s, 1H), 8.05 (t, J = 7.9 Hz, 4H), 7.83 (d, J = 7.8 Hz, 2H), 7.70 (d, J = 8.3 Hz, 2H), 7.54 – 7.30 (m, 7H), 5.56 (d, J = 11.0 Hz, 1H), 3.59 (q, J = 8.1, 7.6 Hz, 1H), 3.48 (q, J = 8.0 Hz, 1H), 2.27 – 2.19 (m, 2H), 1.97 – 1.81 (m, 2H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 193.3, 176.2, 147.4, 145.1, 136.2, 135.8, 134.0, 133.8, 129.6, 128.9, 128.8, 128.8, 123.7, 60.3, 57.5, 49.8, 31.9, 29.7, 18.4 ppm. HRMS: Calcd for $C_{26}H_{22}N_2O_5$: [M]+ 442.1529; Found, 442.1414.



2-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)-1-phenylbutane-1,3-dione (7ac): White solid.

M.p. = 164.5-165.3 °C. IR (KBr) ν = 3428, 3065, 2958, 1674, 1514, 1424, 1267, 1175, 857 cm^{-1}. Characterized as a 5:2 diastereomeric mixture. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.10 (d, J = 8.7 Hz, 2H), 7.96 (d, J = 8.7 Hz, 2H), 7.65–7.45 (m, 5H), 6.10 (d, J = 11.4 Hz, 1H), 5.88 (d, J = 11.4 Hz, 1H), 3.49–3.43 (m, 1H), 3.22–3.16 (m, 1H), 2.39–2.34 (m, 2H), 2.23 (s, 3H), 1.97–1.92 (m, 2H)ppm. Isomer ^1H NMR (400 MHz, $CDCl_3$): δ = 8.21 (d, J = 8.6 Hz, 2H), 8.13 (d, J = 8.6 Hz, 2H), 7.76 (d, J = 8.2 Hz, 2H), 7.67–7.62 (m, 4H), 6.47 (d, J = 11.4 Hz, 1H), 5.46 (d, J = 11.4 Hz, 1H), 3.46–3.43 (m, 1H), 3.35–3.34 (m, 1H), 2.19–2.17 (m, 2H), 1.97 (s, 3H), 1.92–1.86 (m, 2H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 201.2, 192.7, 175.6, 147.5, 144.3, 135.8, 134.5, 129.1, 128.2, 123.9, 63.9, 55.4, 45.3, 31.2, 27.6, 18.3 ppm. Isomer ^{13}C NMR (75 MHz, $CDCl_3$): δ = 201.2, 192.8, 175.6, 147.7, 145.0, 136.2, 134.2, 128.8, 128.6, 126.9, 121.7, 58.3, 48.7, 31.6, 30.5, 29.6, 18.2 ppm. HRMS: Calcd for $C_{21}H_{20}N_2O_5$: [M]+ 380.1372; Found, 380.1264.



1-((4-Nitrophenyl)(2,4,6-trimethoxyphenyl)methyl)pyrrolidin-2-one (9): White solid. M.p. = 159.4-160.1 °C. IR (KBr) ν = 3290, 2955, 1664, 1422, 1105, 779 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$): δ = 8.10 (d, J = 8.5 Hz, 2H), 7.15 (d, J = 8.5 Hz, 2H), 7.05 (s, 1H), 6.13 (s, 2H), 3.84 (s, 3H), 3.65–3.55 (m, 7H), 3.11 (q, J = 8.3 Hz, 1H), 2.62–2.37 (m, 2H), 2.21–2.00 (m, 2H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 175.3, 161.8, 159.8, 150.4, 146.1, 126.0, 123.2, 106.4, 91.0, 55.5, 55.3, 49.3, 44.9, 30.9, 18.2 ppm. HRMS: Calcd for $C_{20}H_{23}N_2O_6$: [M+H]+ 387.1551; Found, 387.1556.

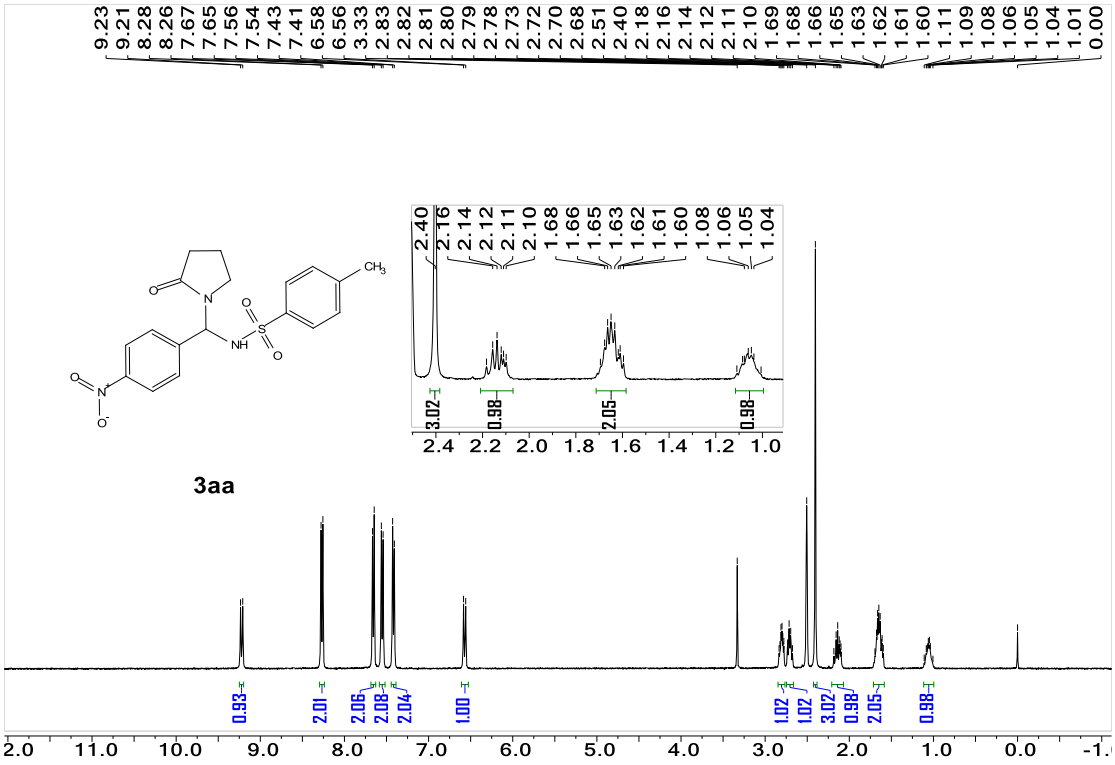
7. References

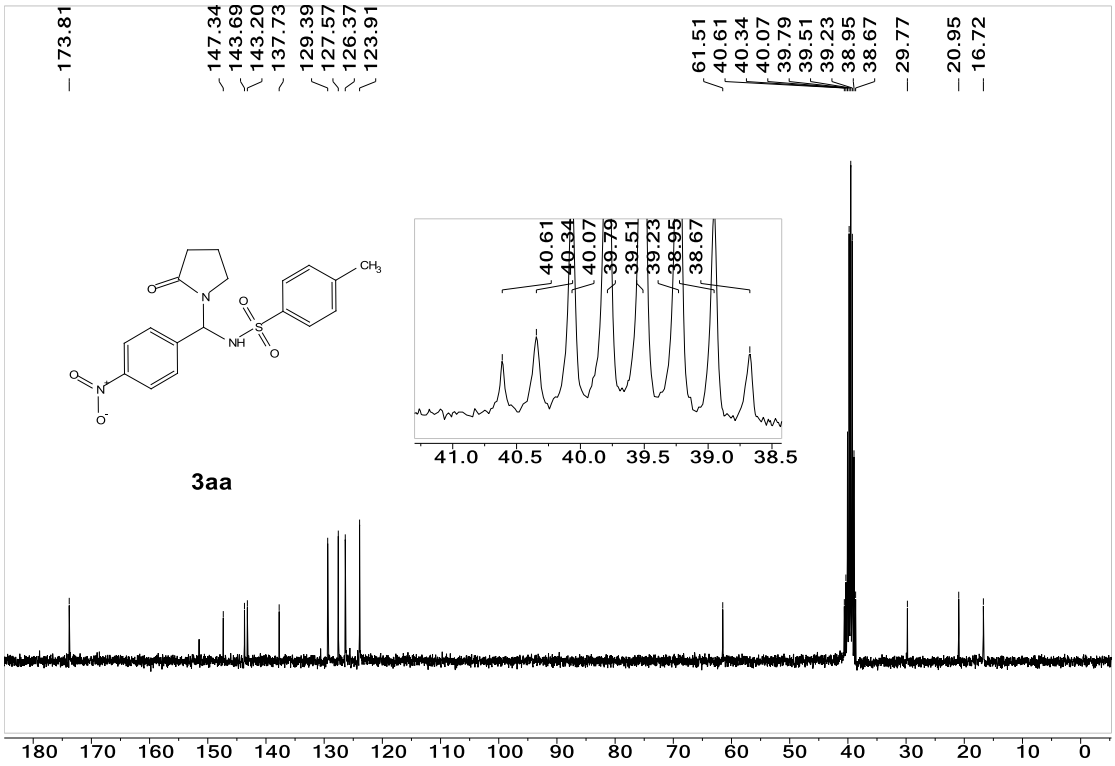
[1] H.-Y. Xu, X.-P. Xu, S.-Y. Wang, S.-J. Ji, *Eur. J. Org. Chem.* **2012**, 5440–5445.

[2] H.-Y. Xu, S.-Y. Wang, R. Jiang, X.-P. Xu, X.-Q. Chu, S.-J. Ji, *Tetrahedron* **2012**, *68*, 8340–8346.

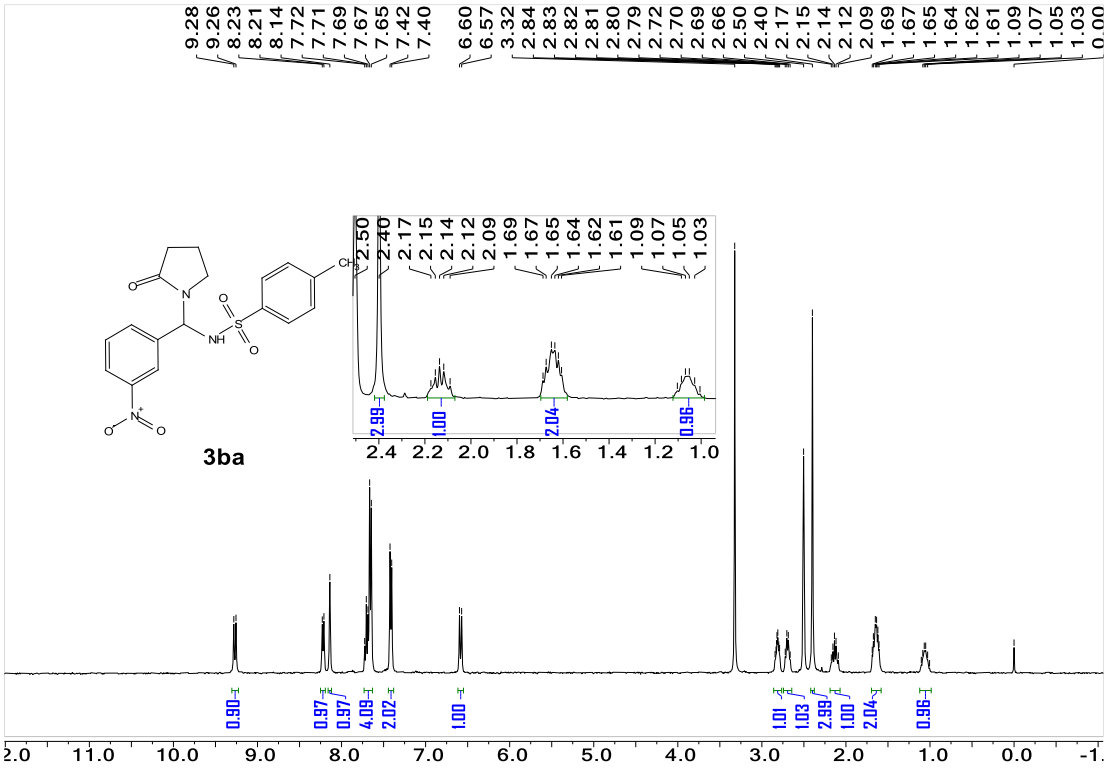
8. The ^{1}H , ^{19}F, and ^{13}C NMR spectra of products

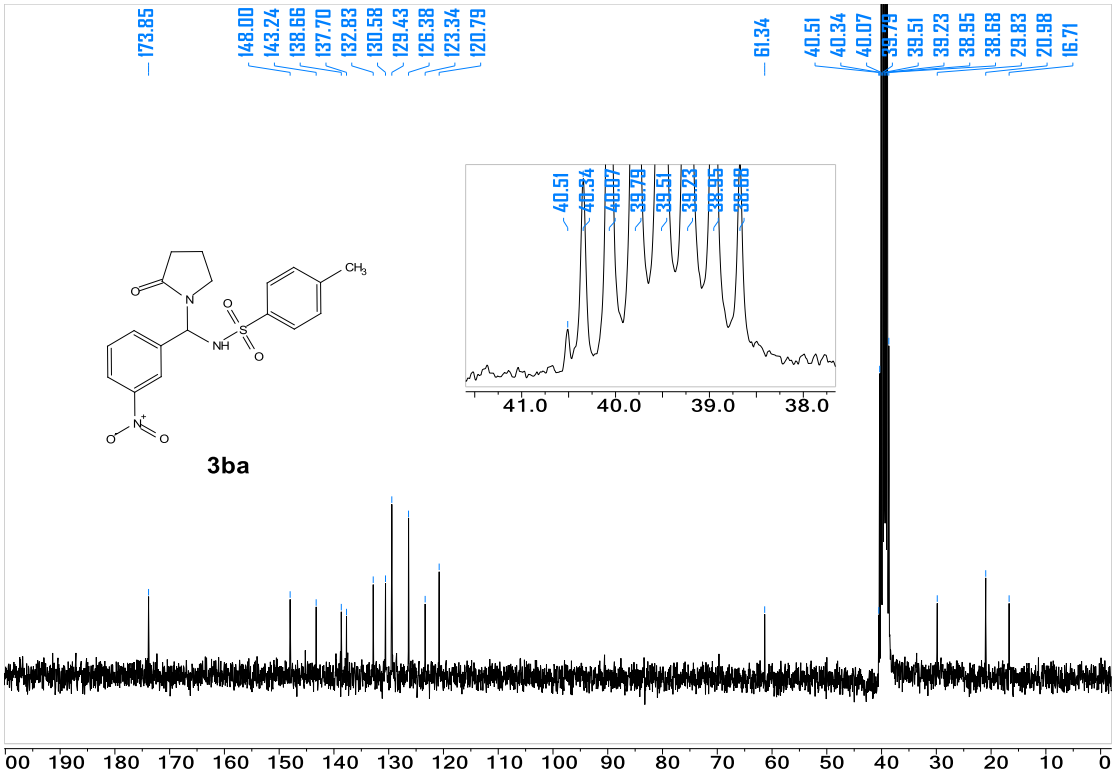
4-Methyl-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3aa):



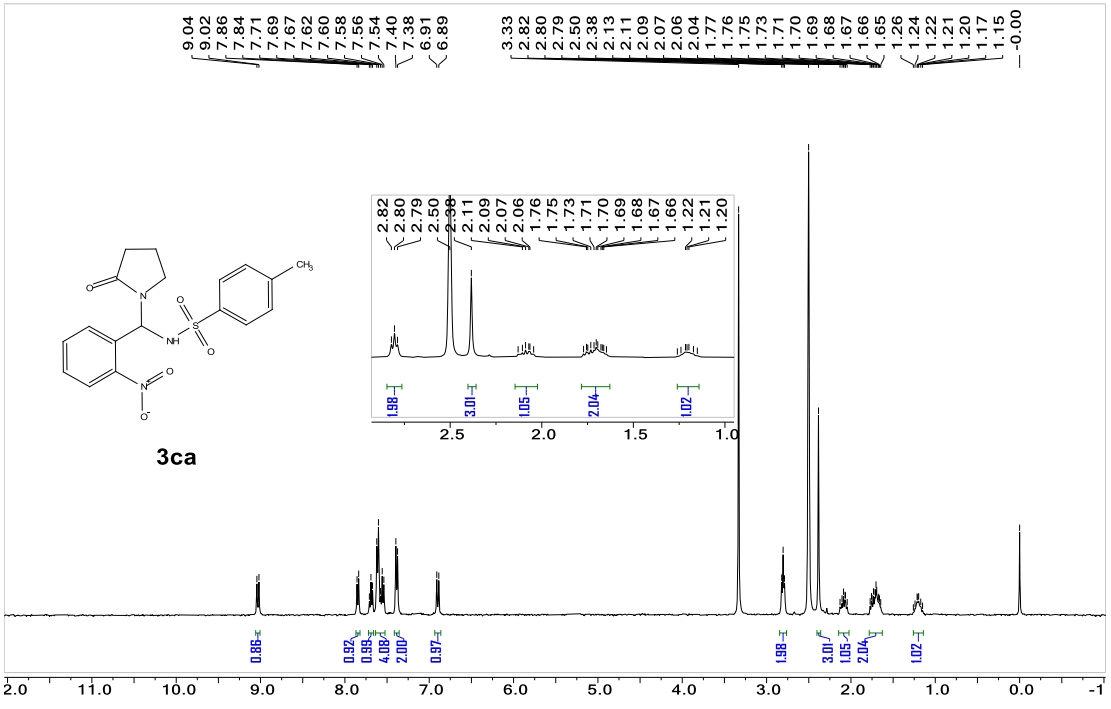


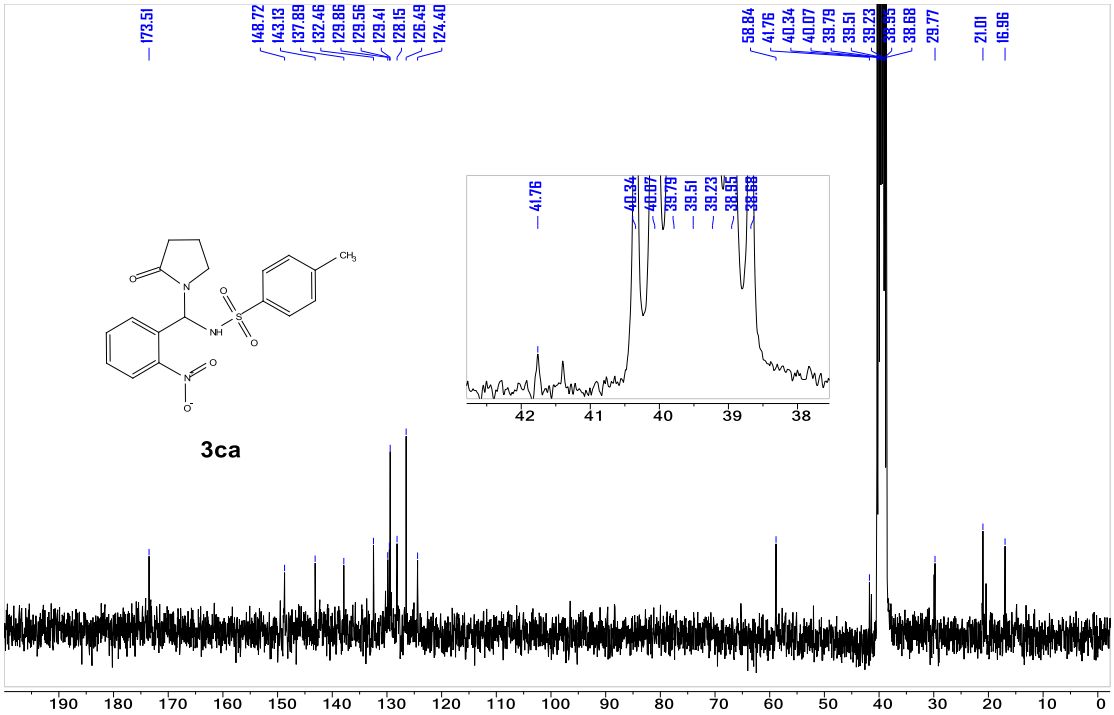
4-Methyl-*N*-((3-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ba):



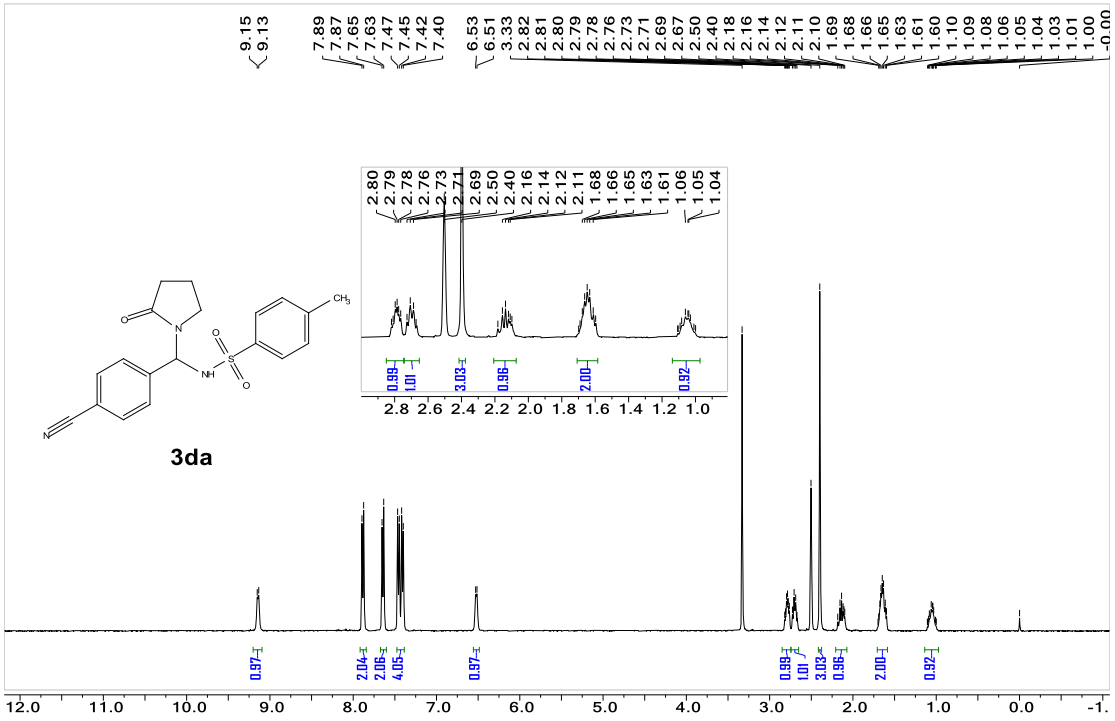


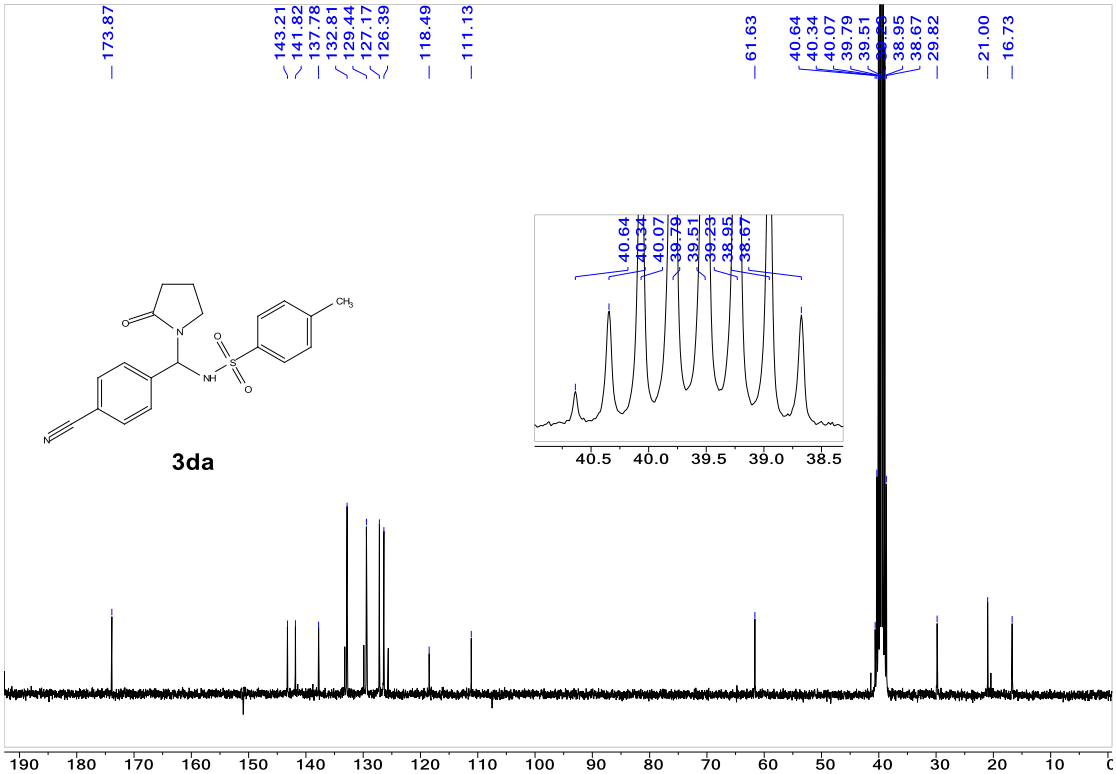
4-Methyl-*N*-((2-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ca):



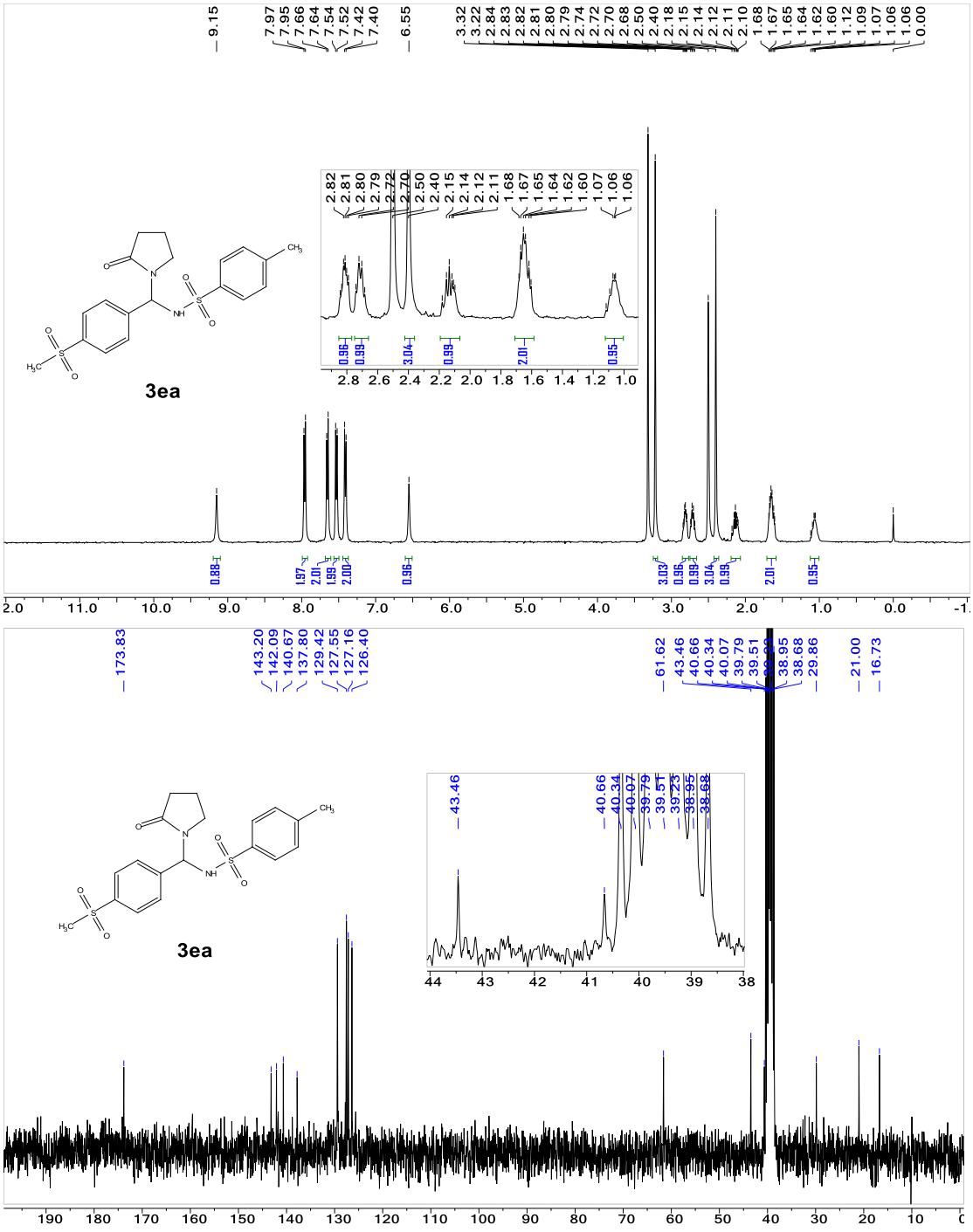


***N*-((4-Cyanophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3da):**

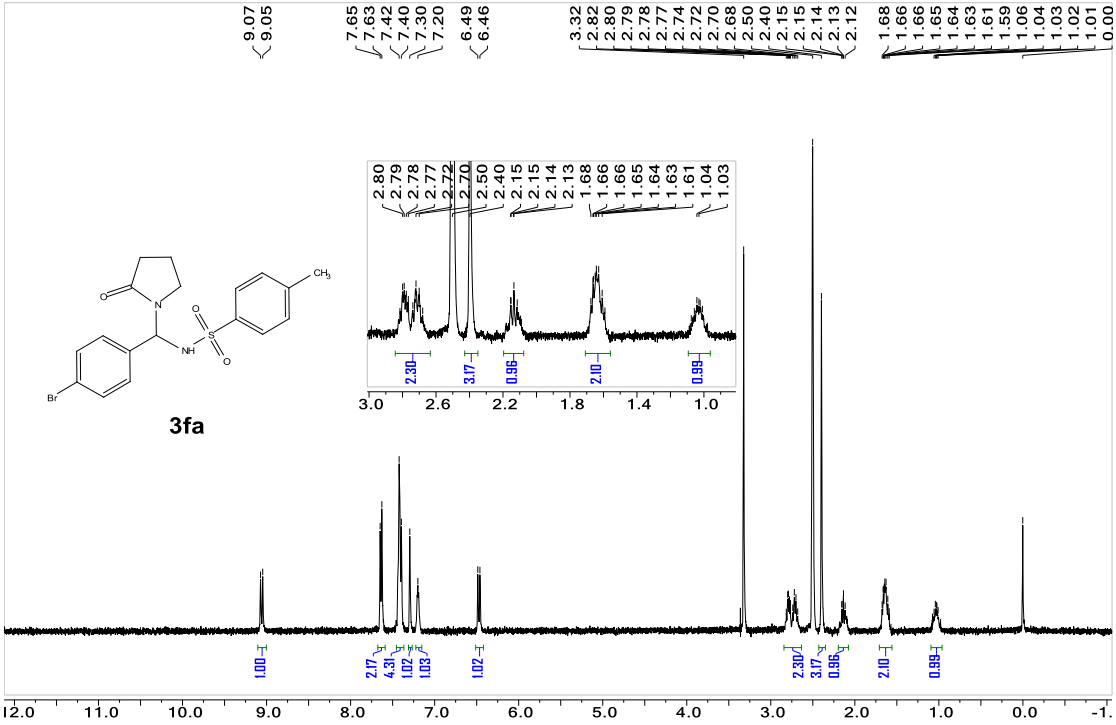


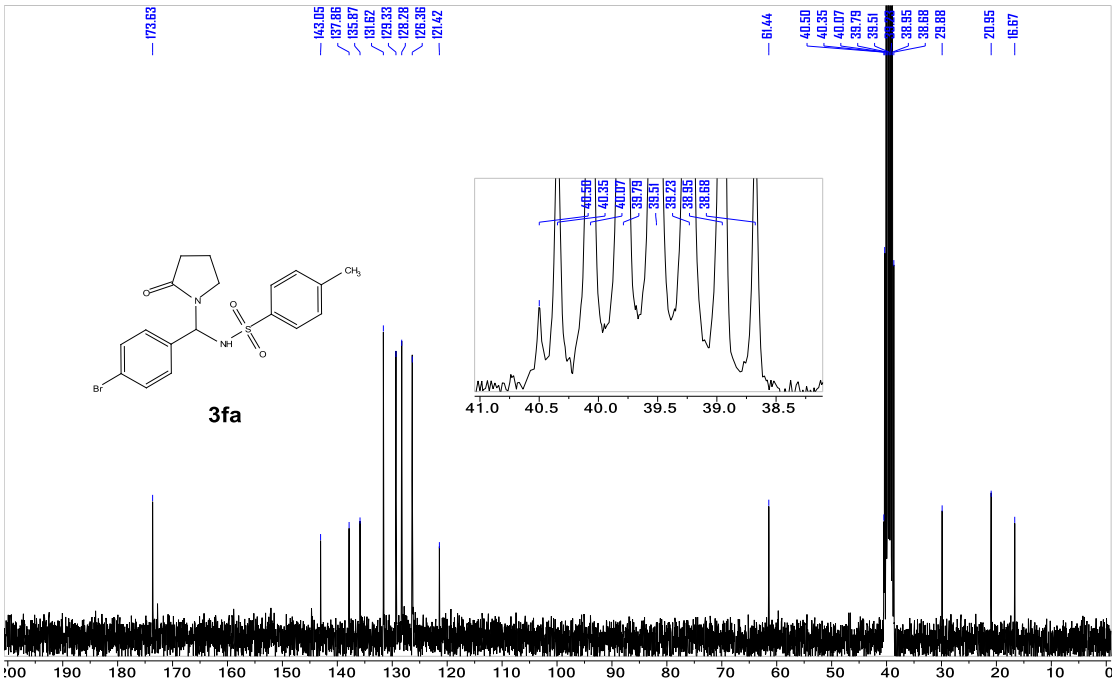


4-Methyl-*N*-((4-(methylsulfonyl)phenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ea):

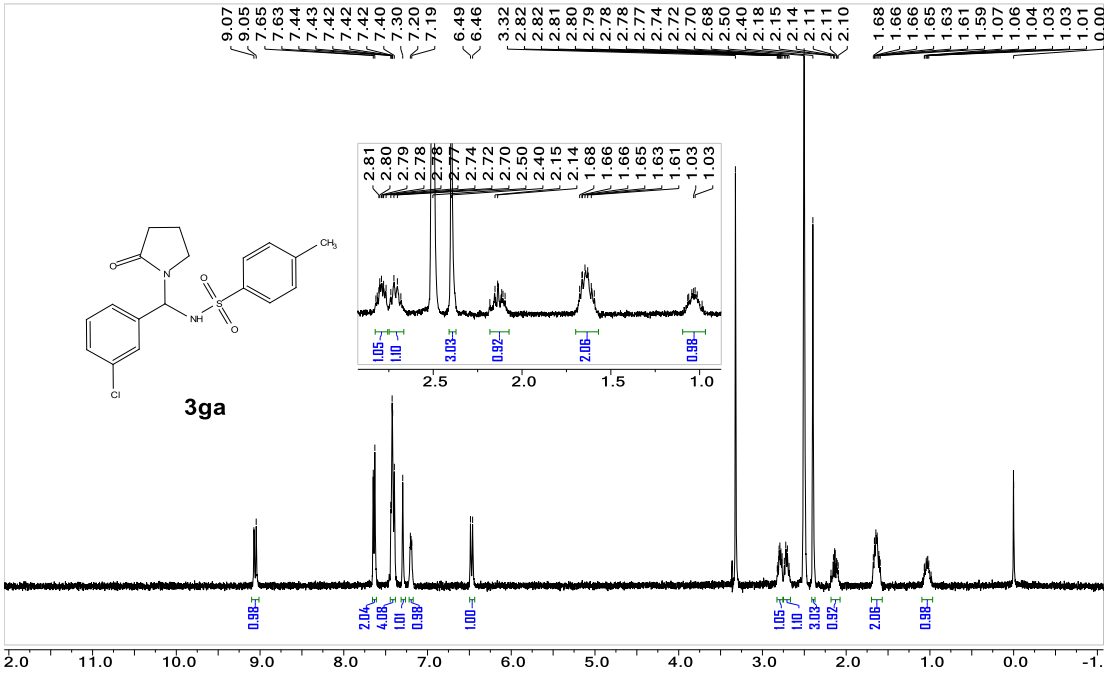


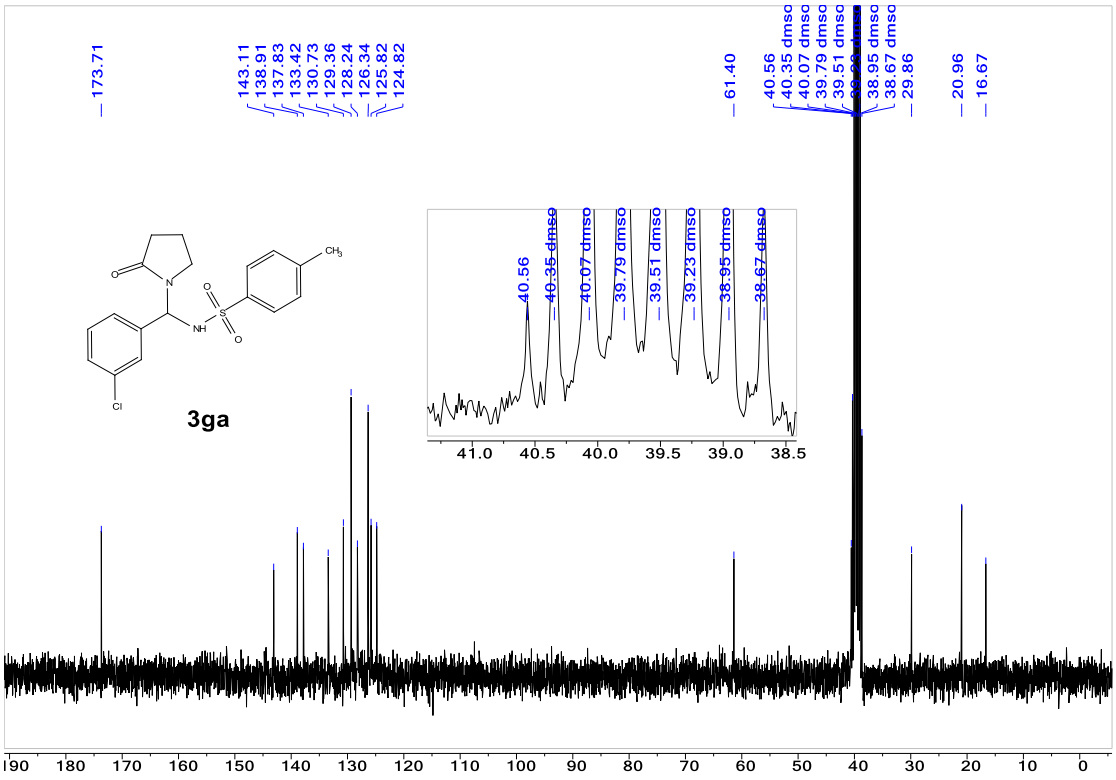
***N*-((4-Bromophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3fa):**



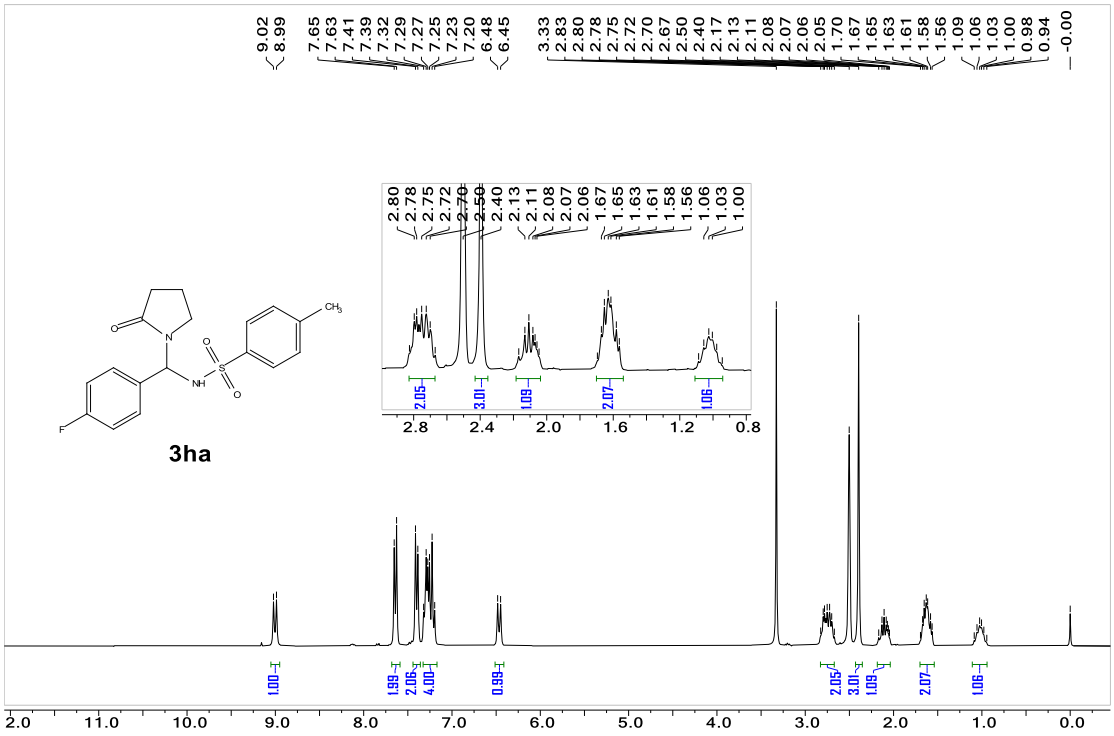


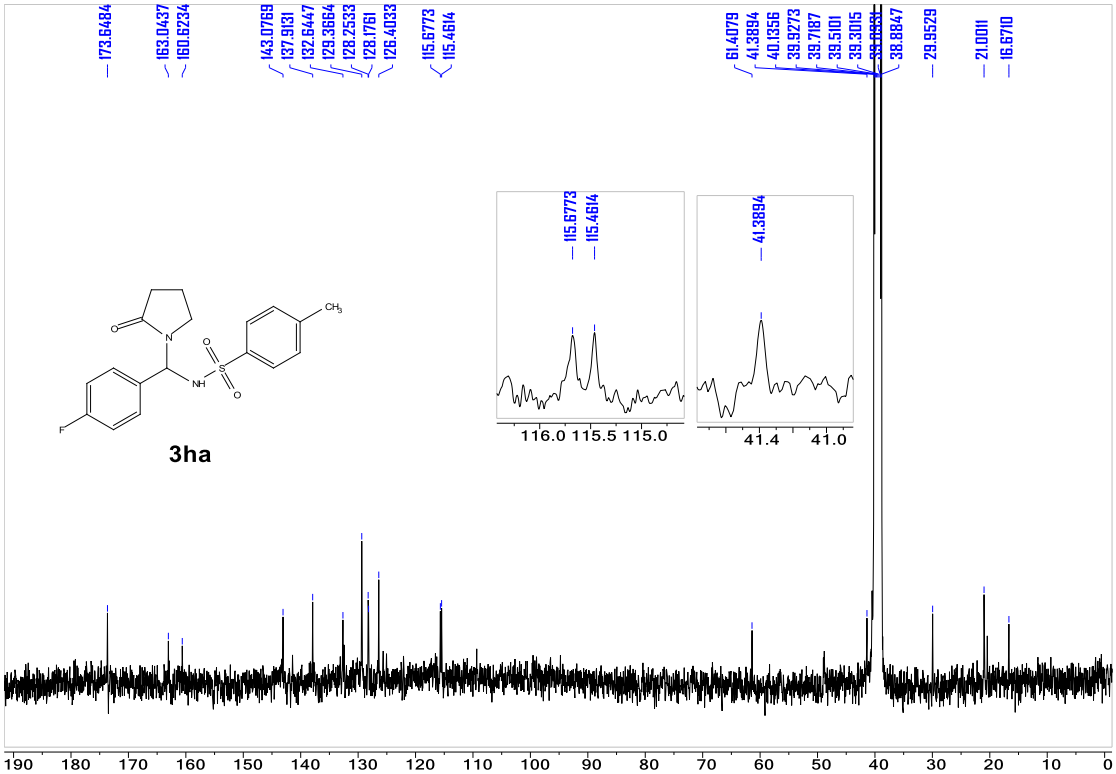
***N*-((3-Chlorophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ga):**



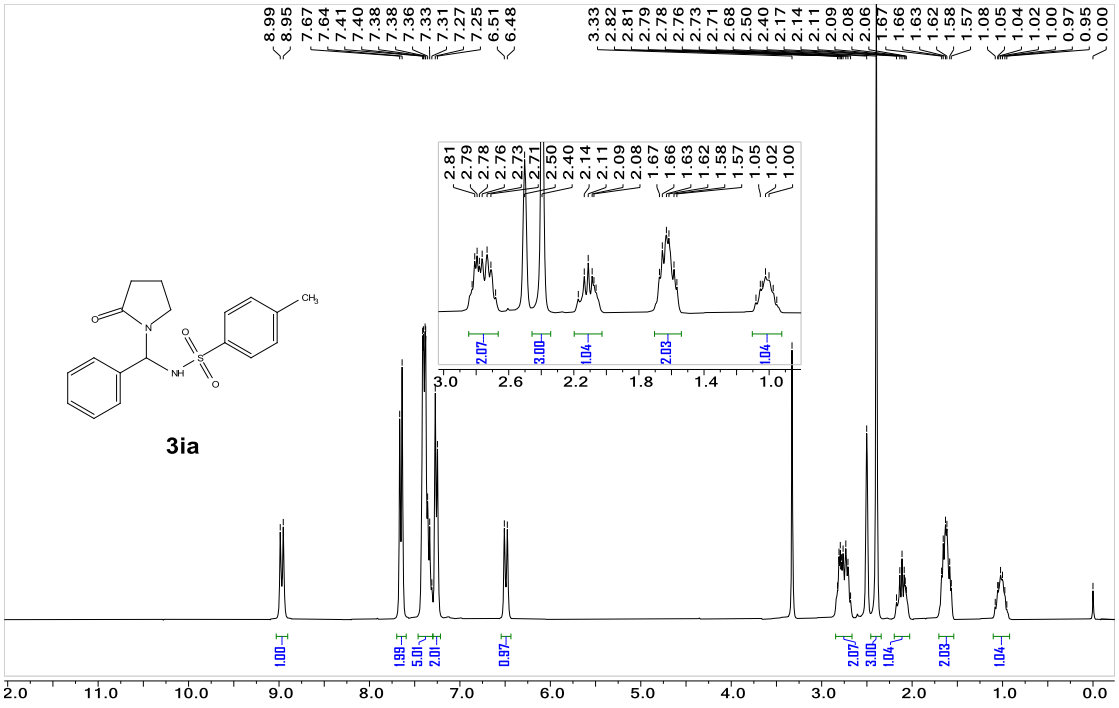


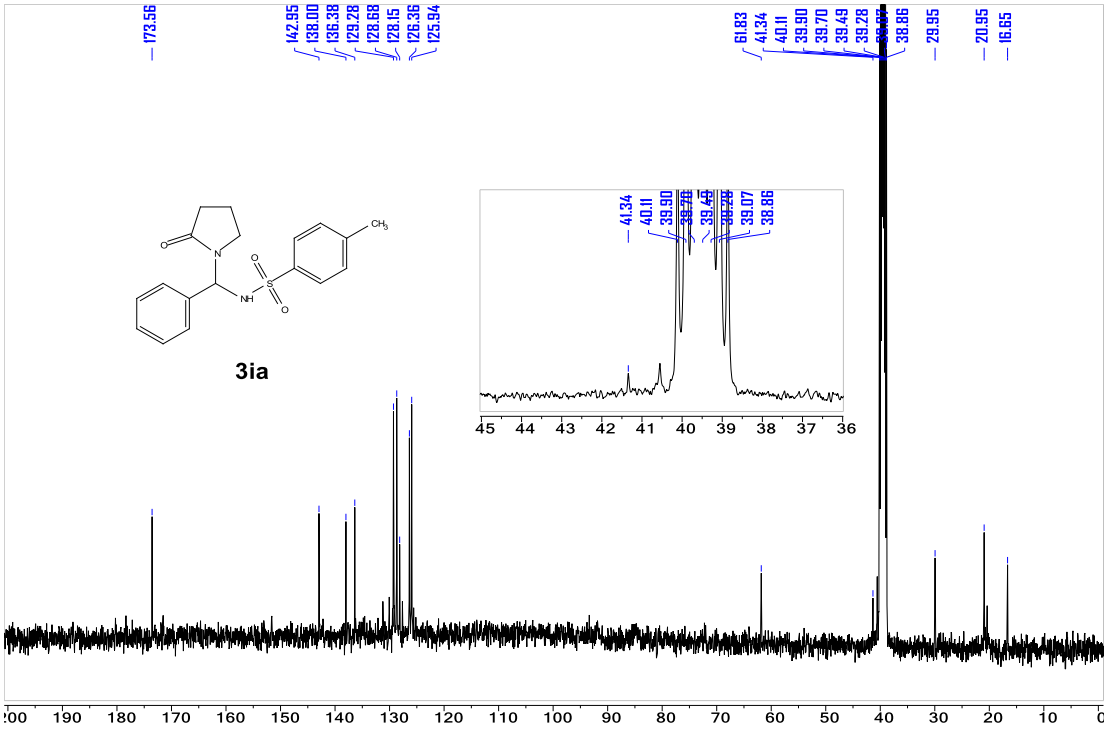
***N*-((4-Fluorophenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ha):**



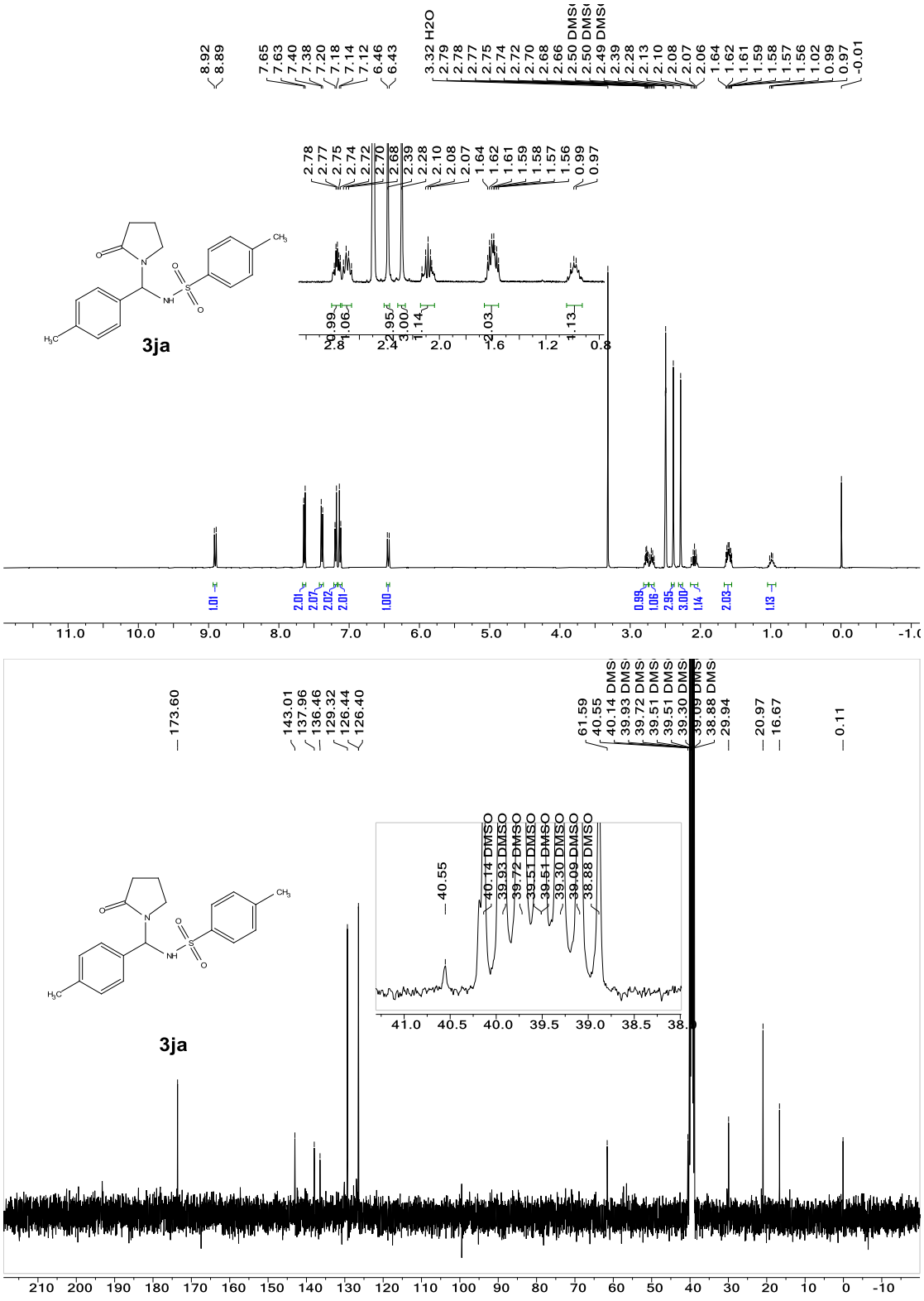


4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(phenyl)methyl)benzenesulfonamide (3ia):

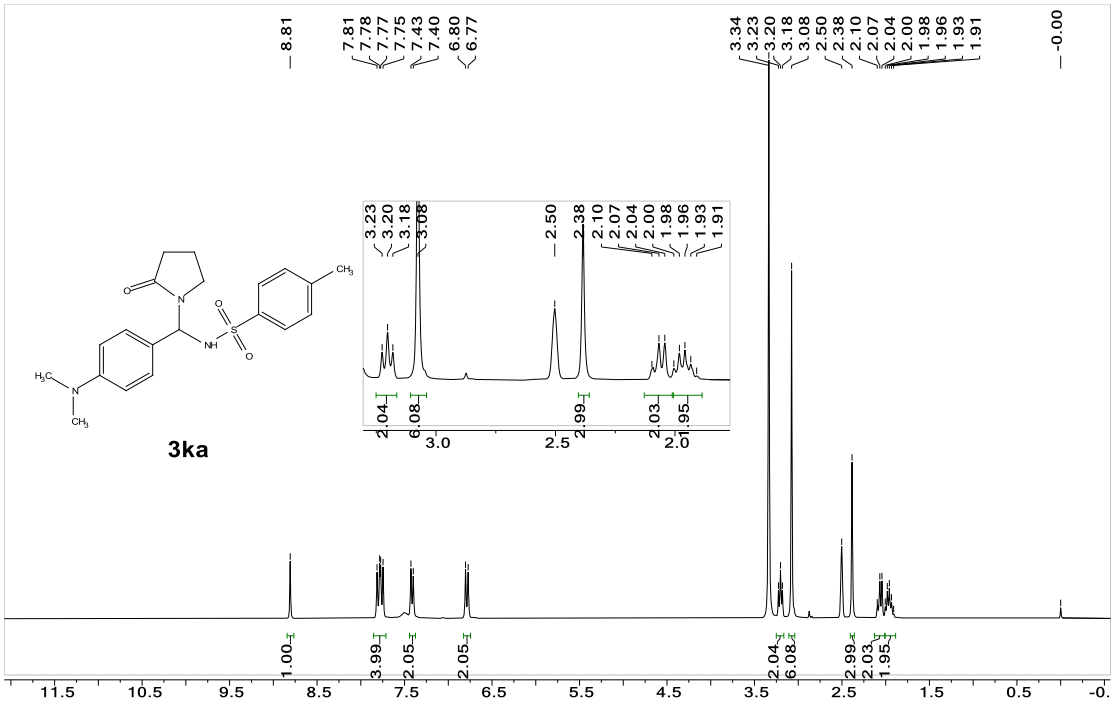


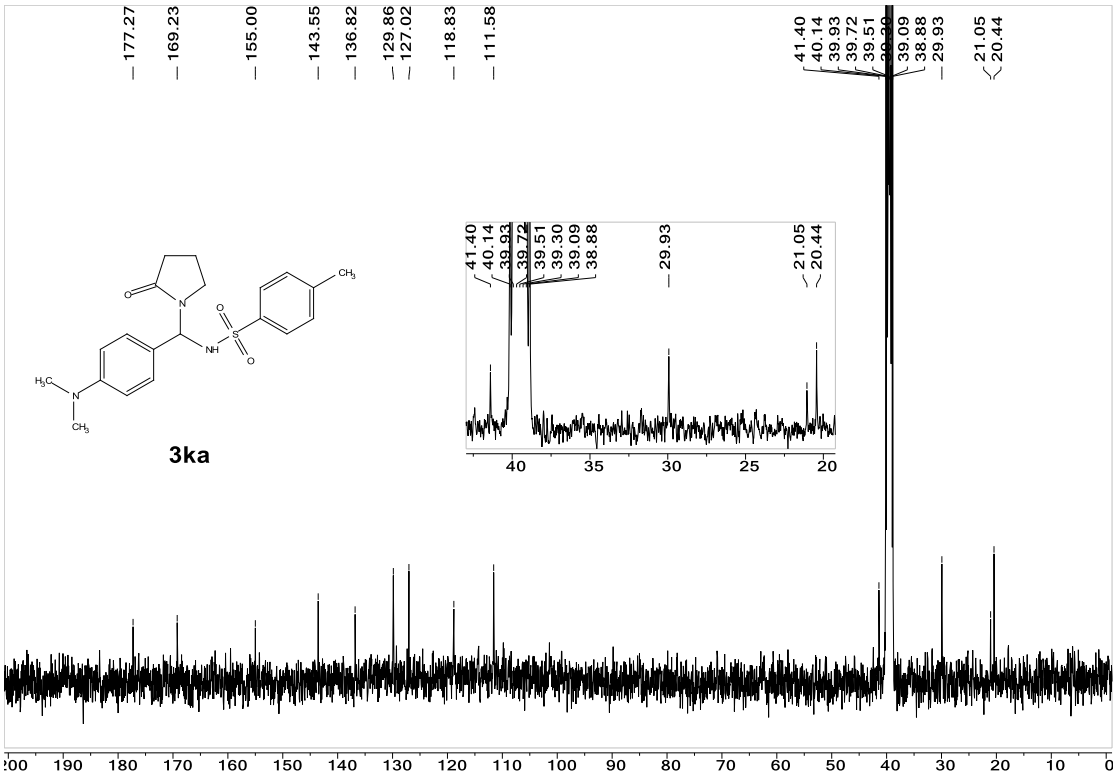


4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(*p*-tolyl)methyl)benzenesulfonamide (3ja):

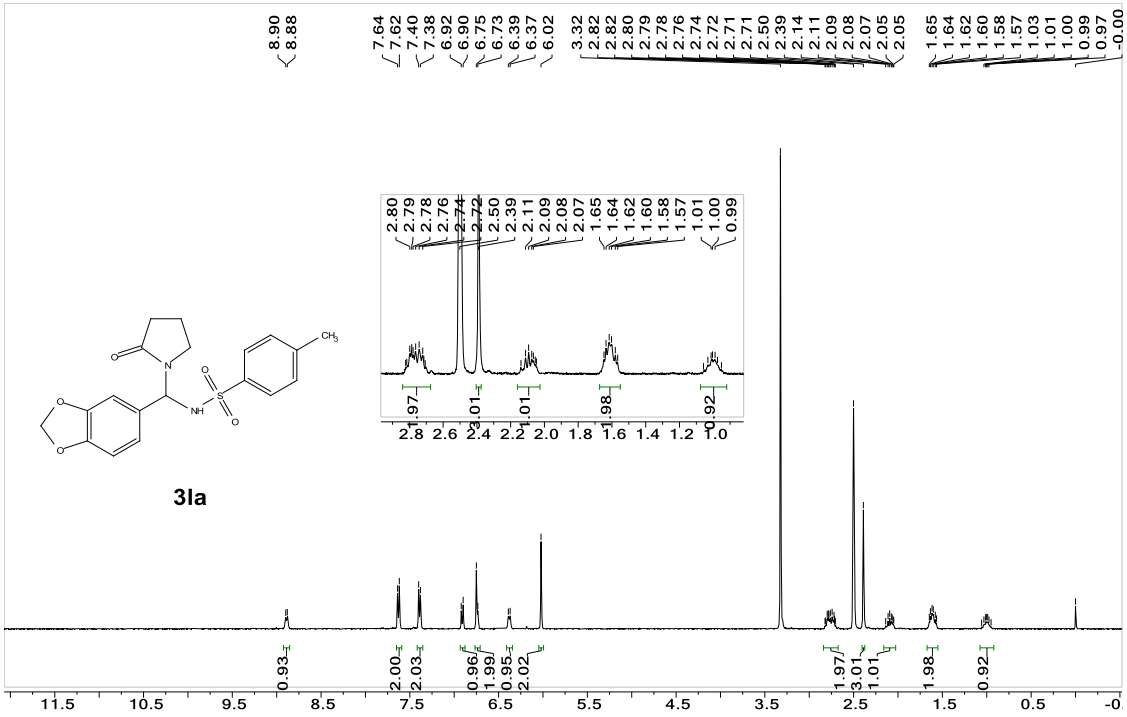


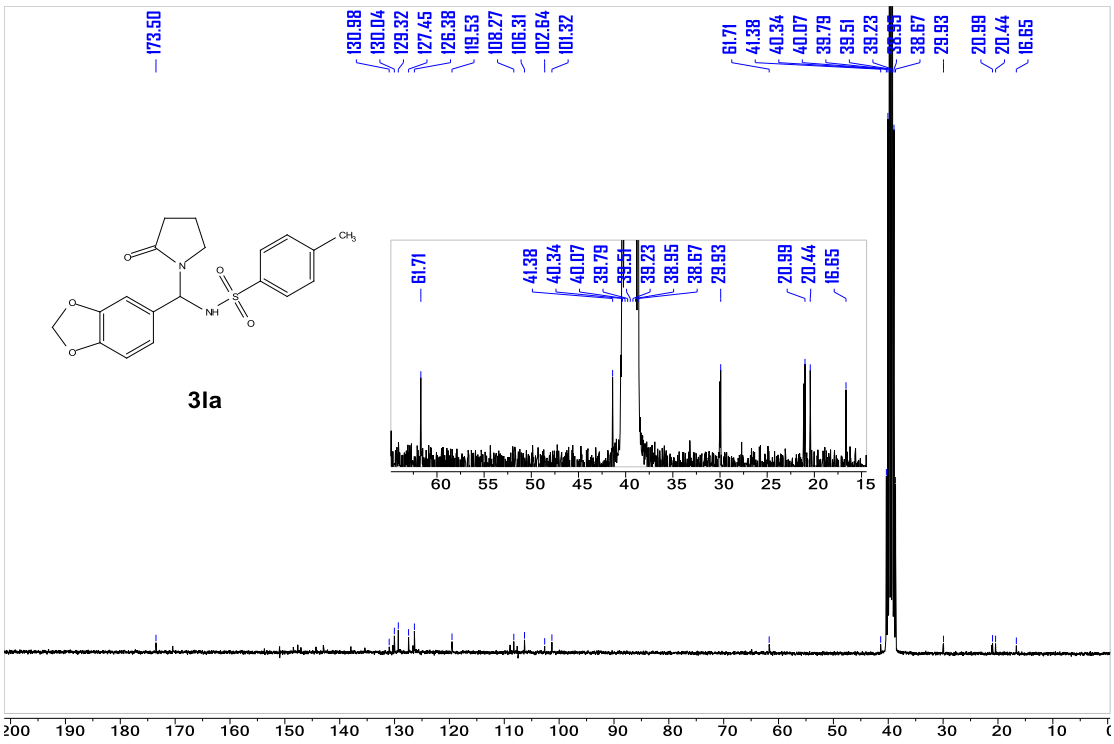
***N*-((4-(Dimethylamino)phenyl)(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3ka):**



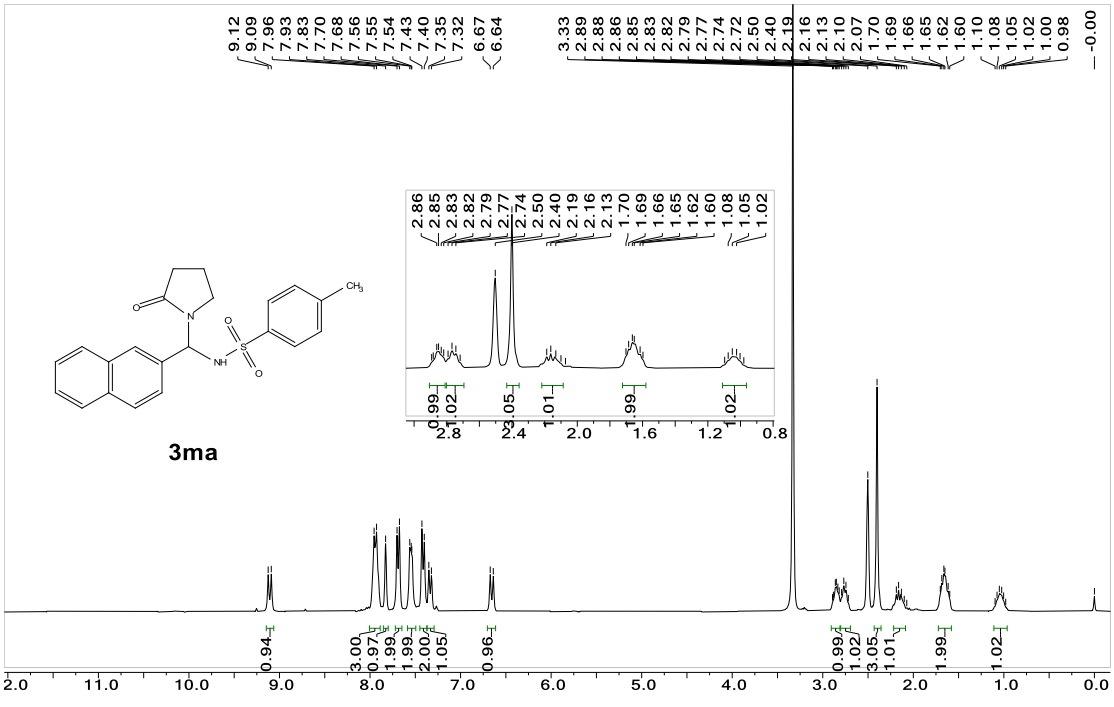


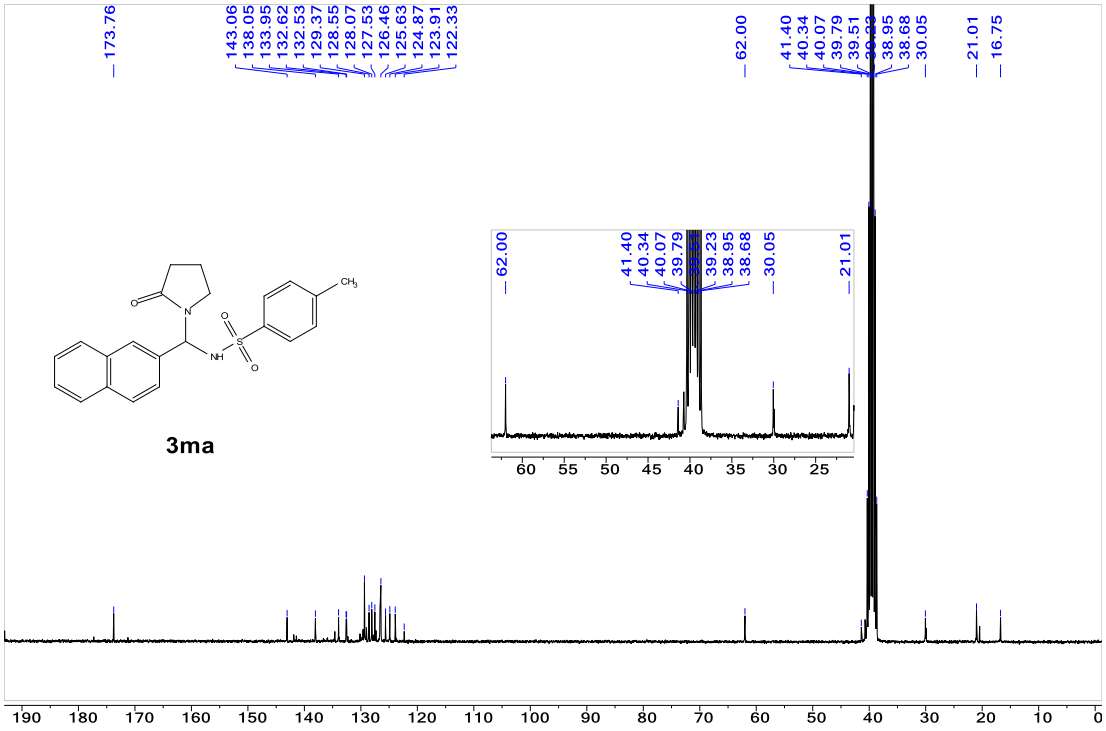
***N*-(Benzo[*d*][1,3]dioxol-5-yl(2-oxopyrrolidin-1-yl)methyl)-4-methylbenzenesulfonamide (3la):**



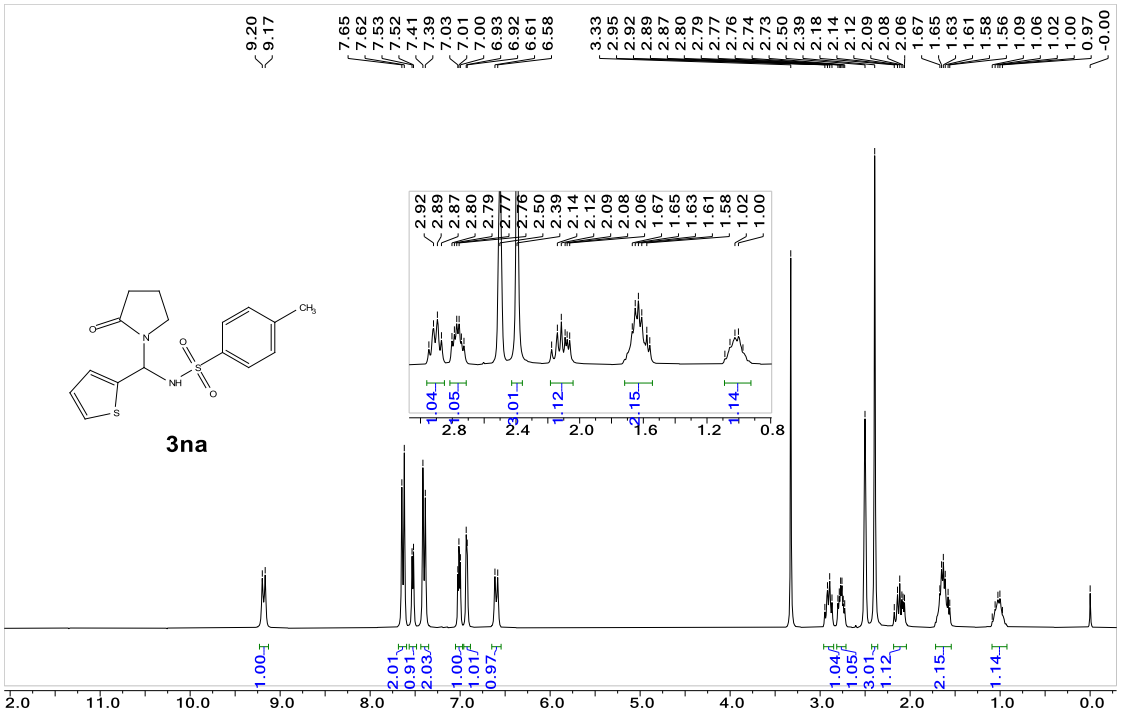


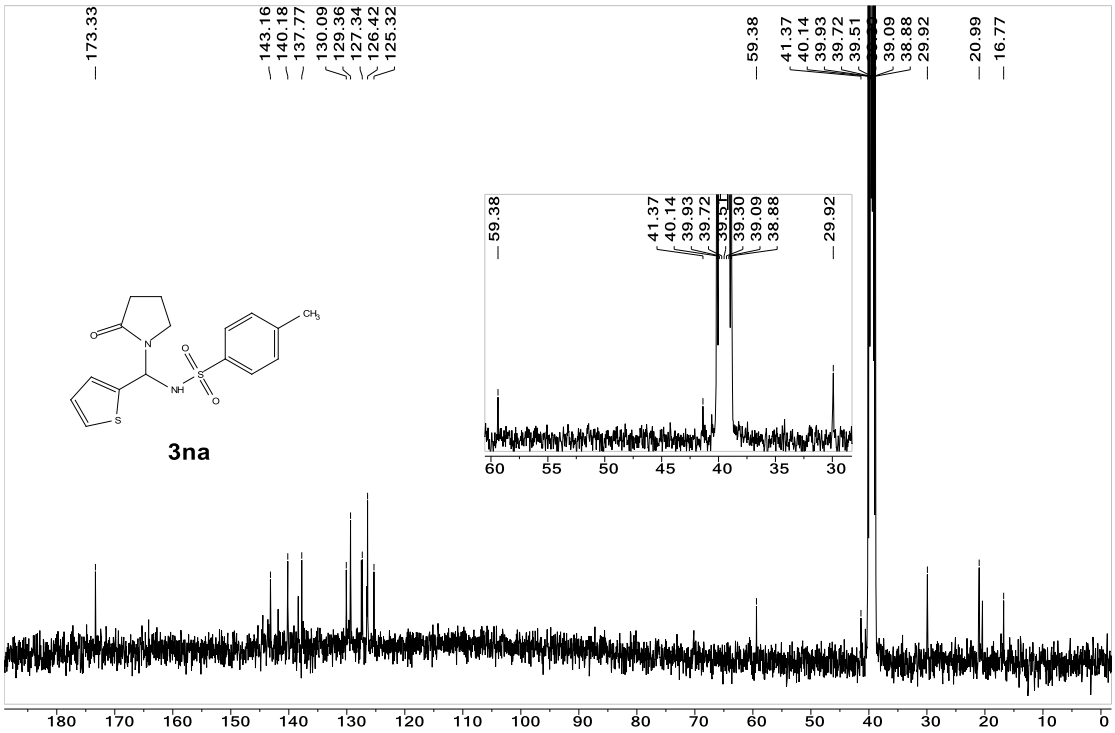
4-Methyl-*N*-(naphthalen-2-yl(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ma):



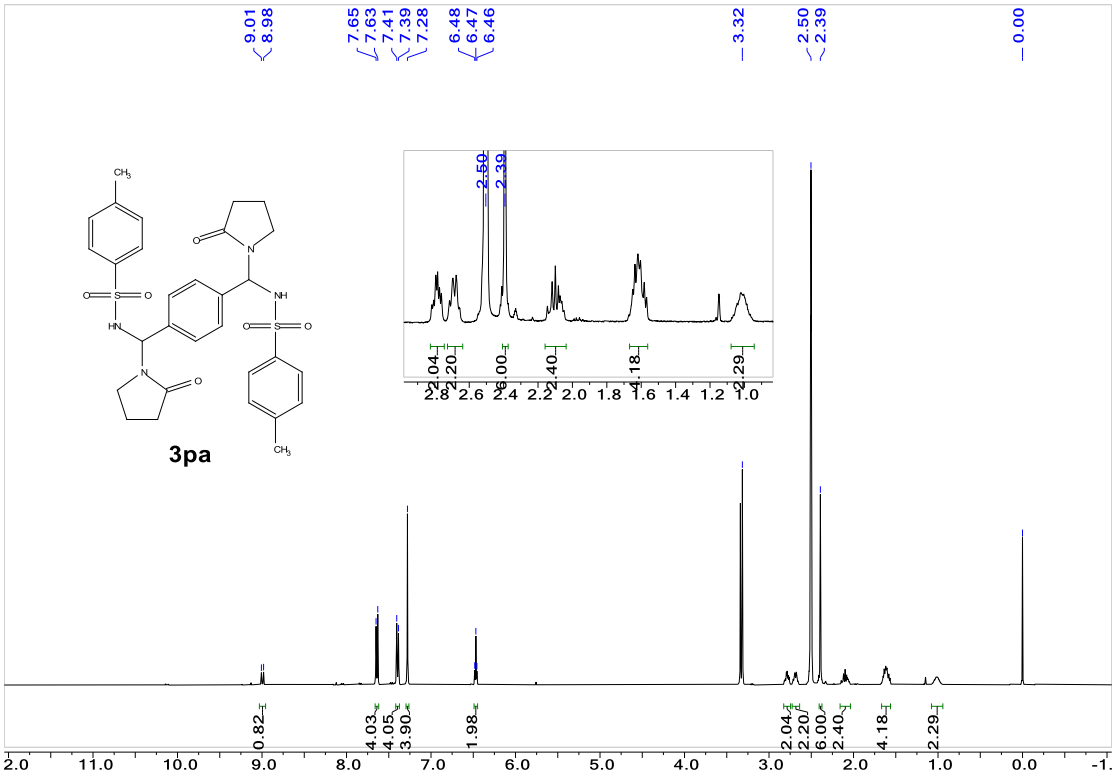


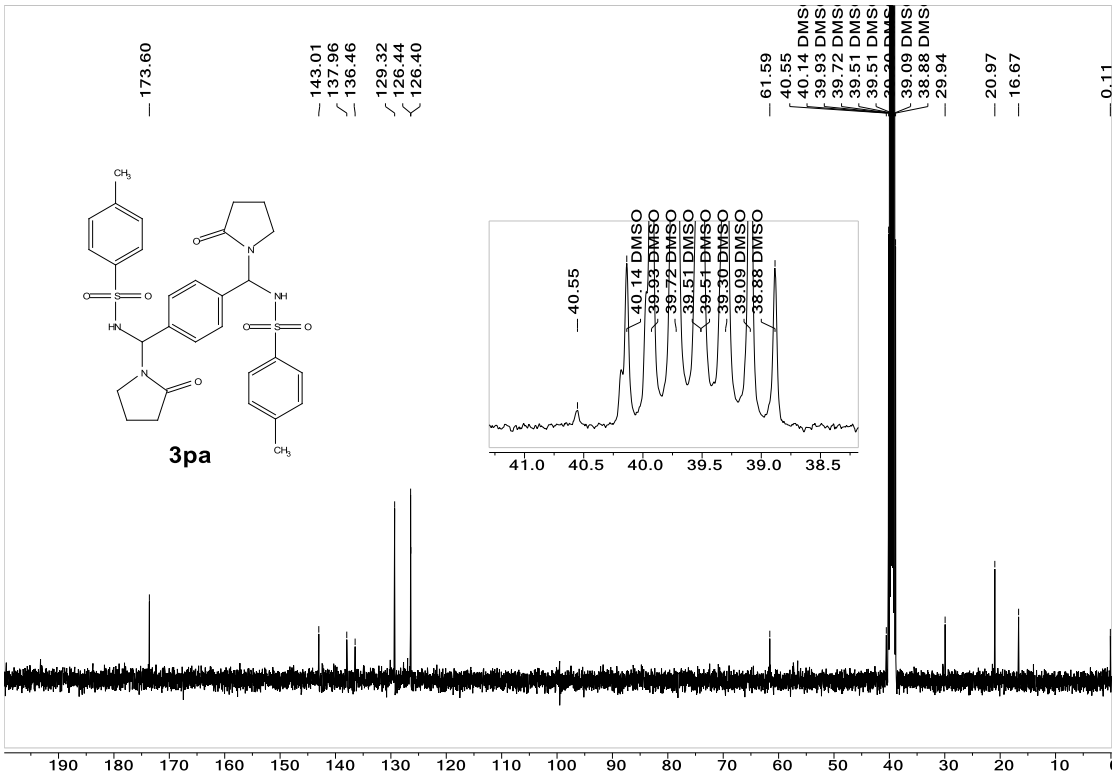
4-Methyl-*N*-((2-oxopyrrolidin-1-yl)(thiophen-2-yl)methyl)benzenesulfonamide (3na):



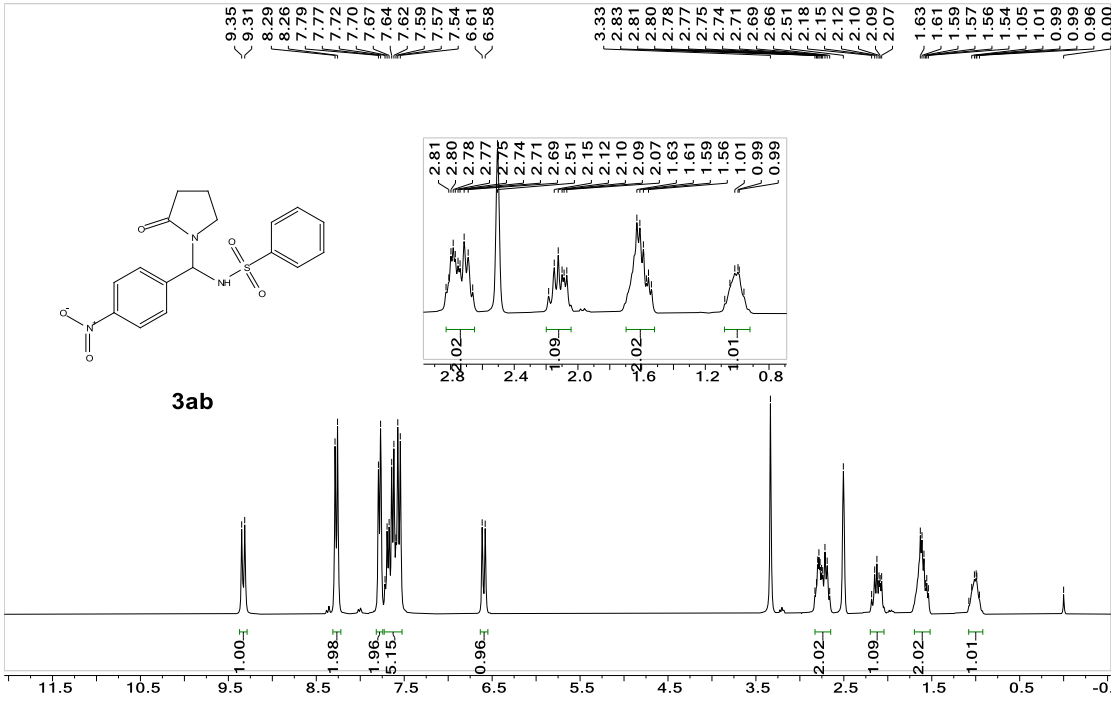


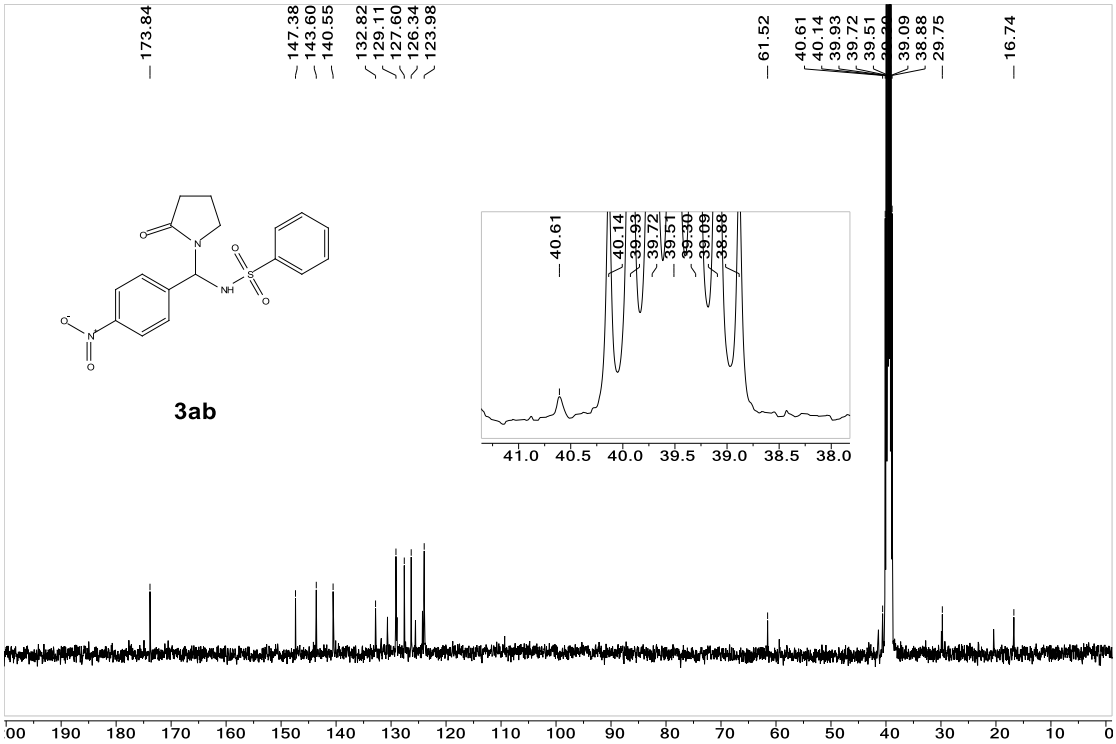
***N*,*N*'-(1,4-Phenylenebis((2-oxopyrrolidin-1-yl)methylene))bis(4-methylbenzenesulfonamide) (3pa):**



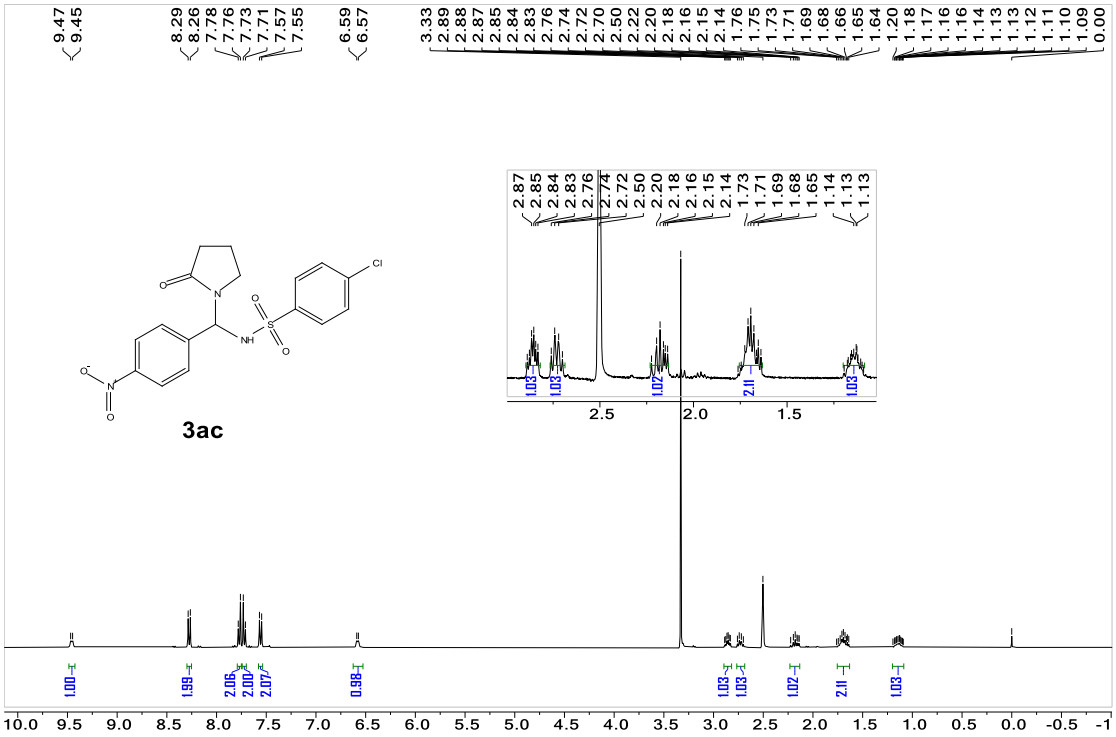


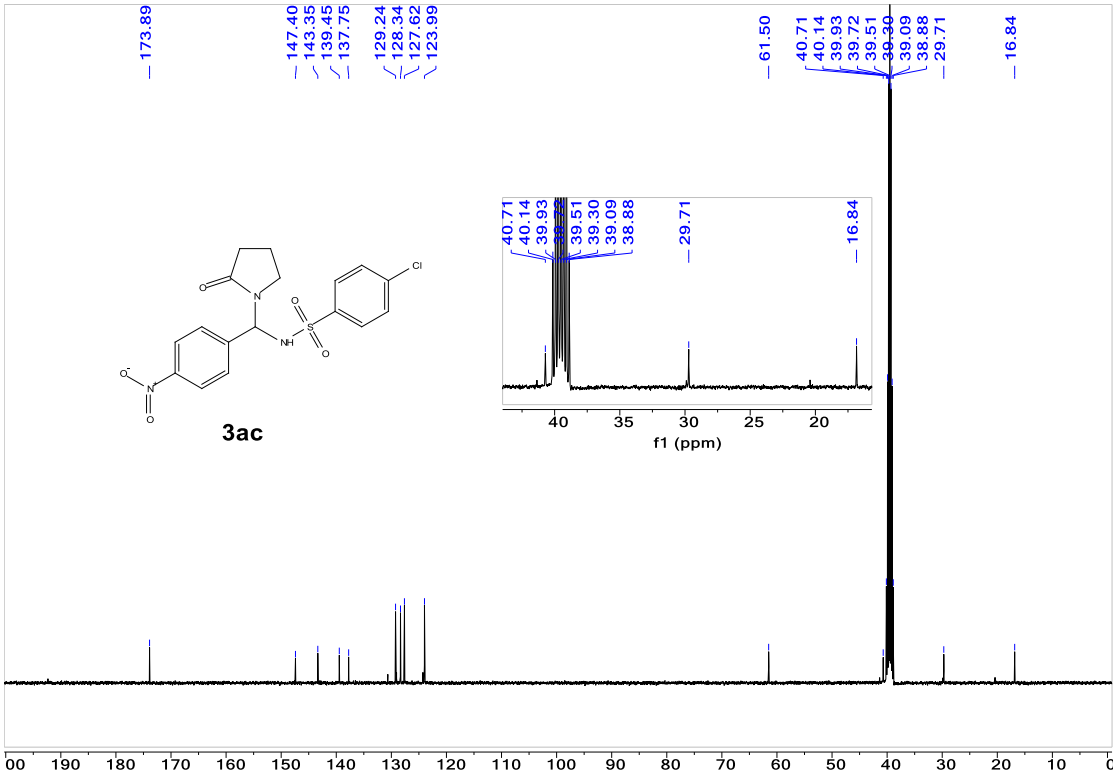
***N*-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ab):**



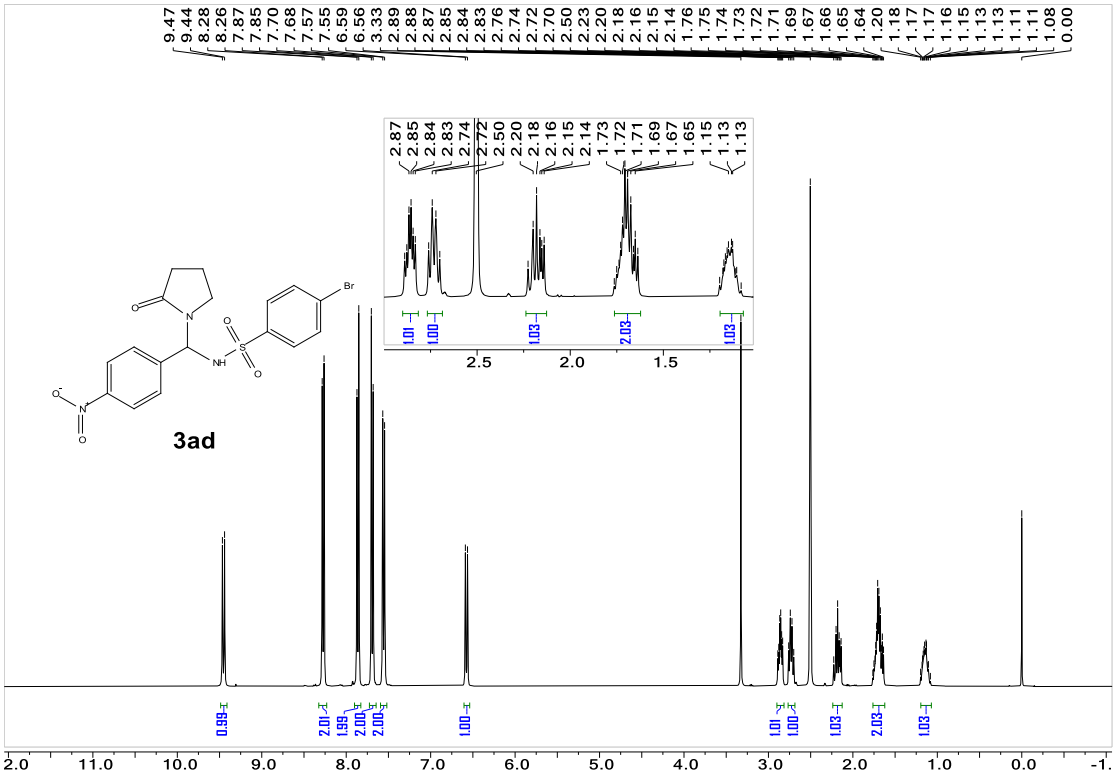


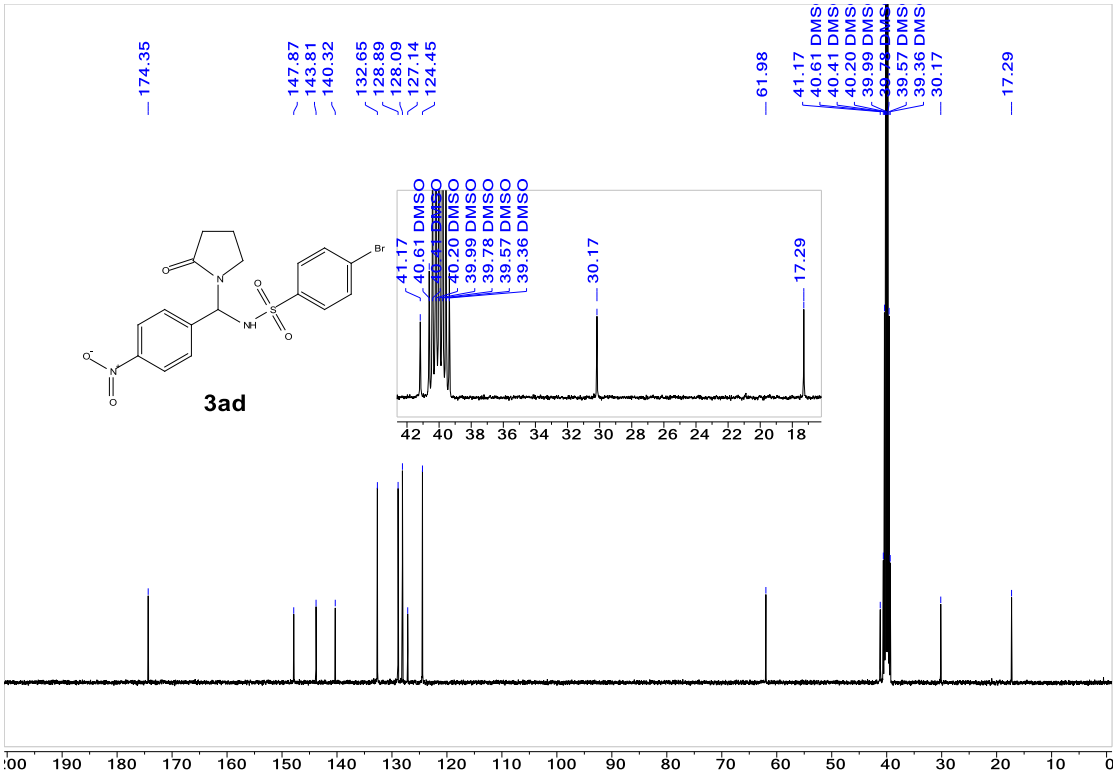
4-Chloro-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ac):



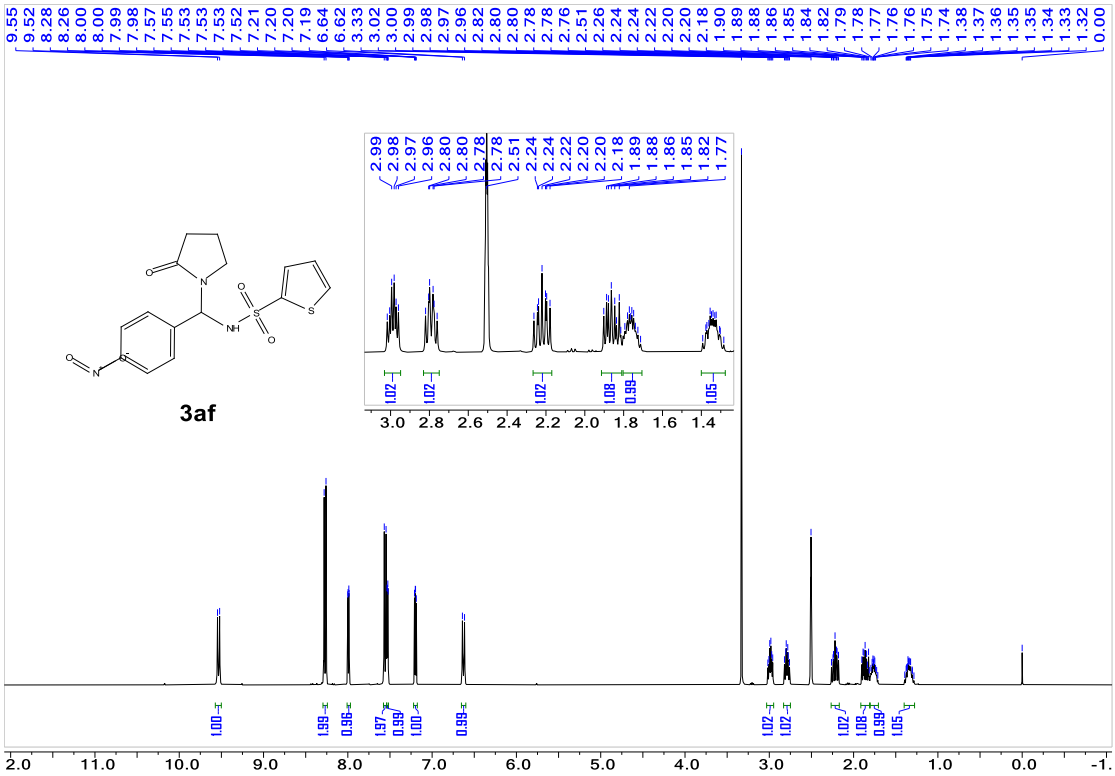


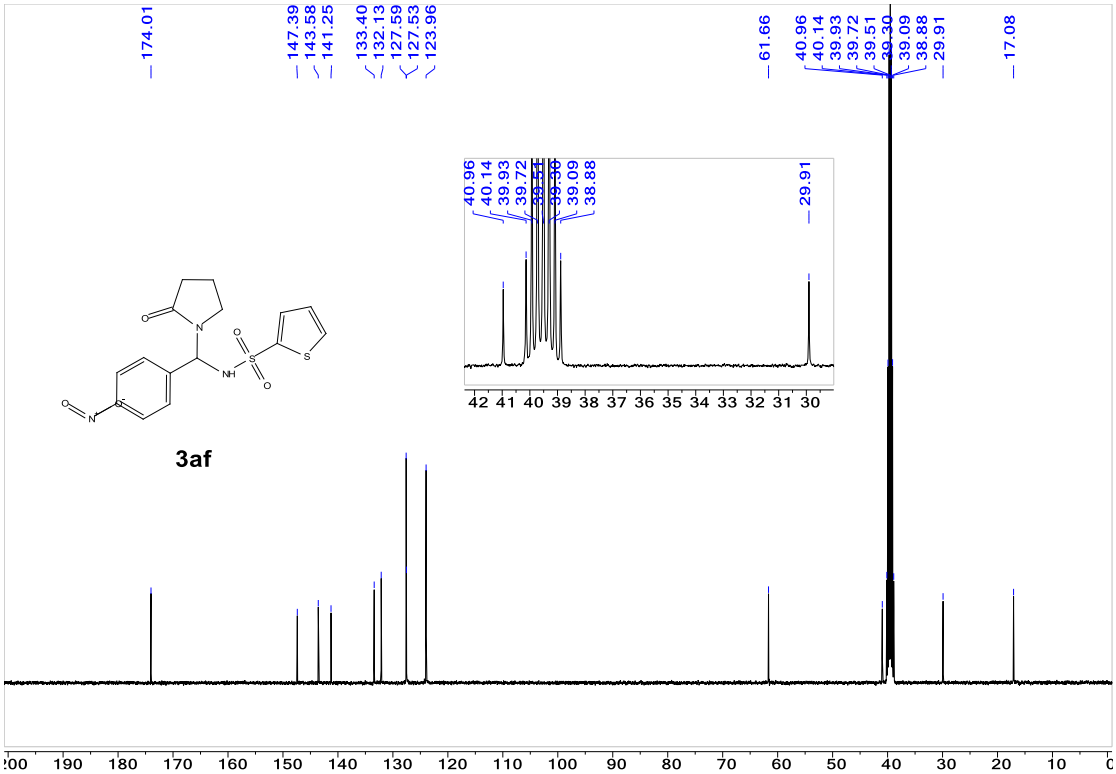
4-Bromo-*N*-((4-nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)benzenesulfonamide (3ad):



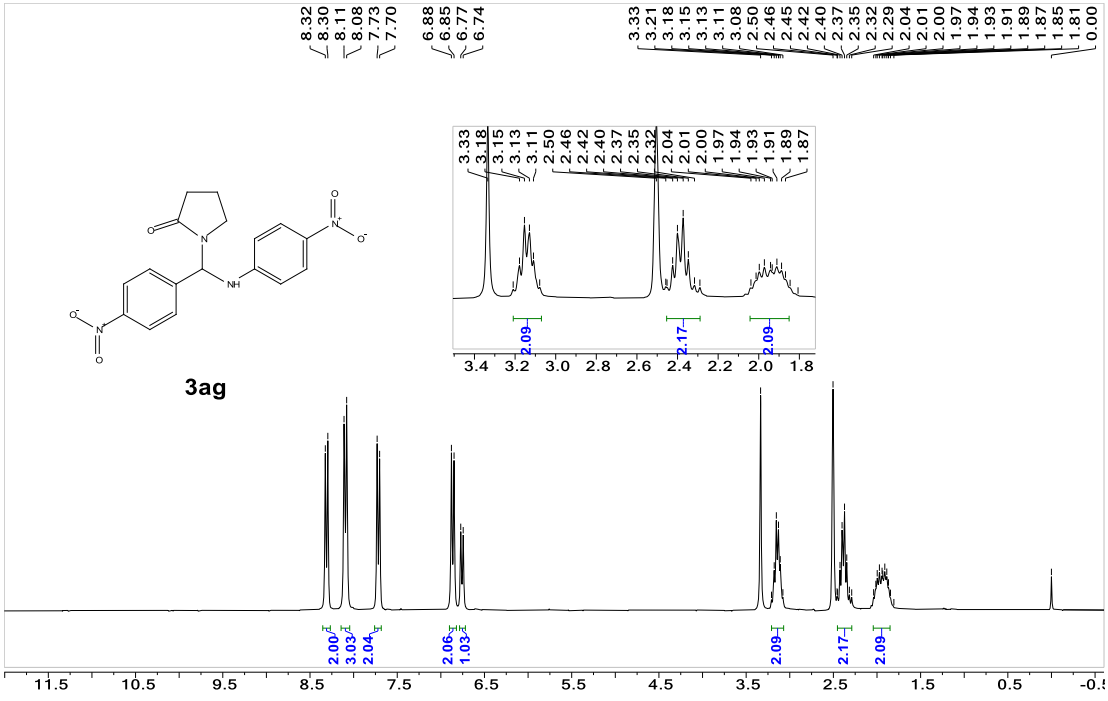


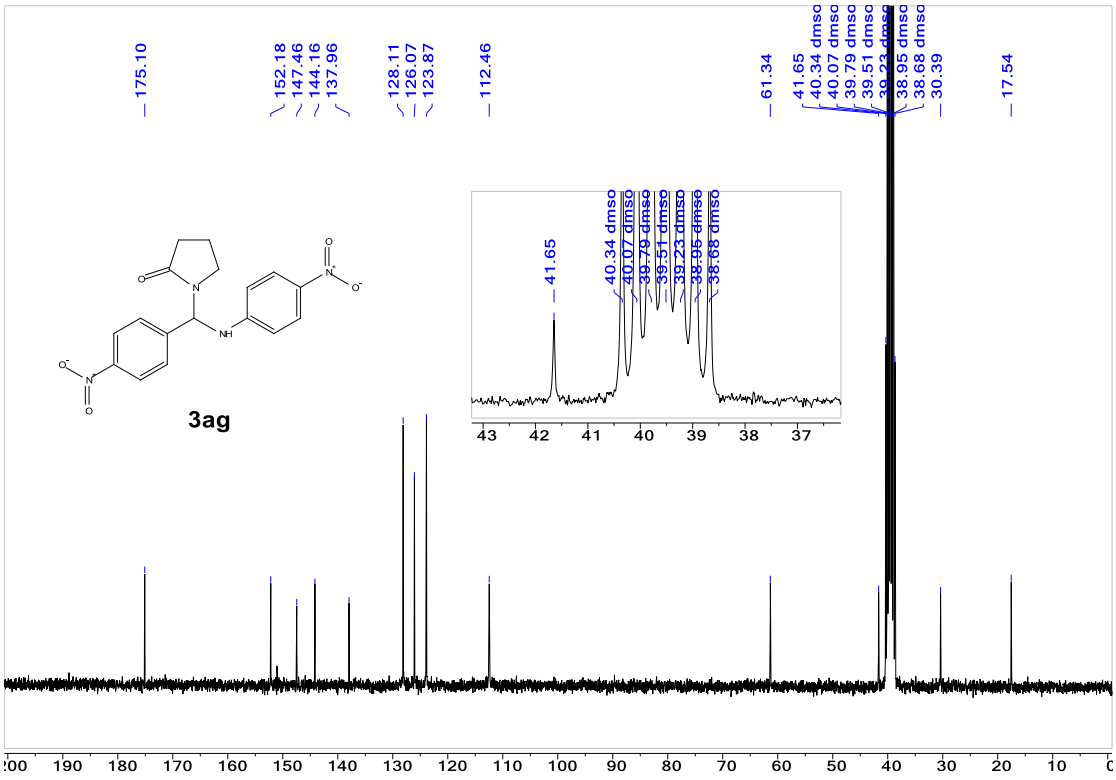
***N*-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)thiophene-2-sulfonamide (3af):**



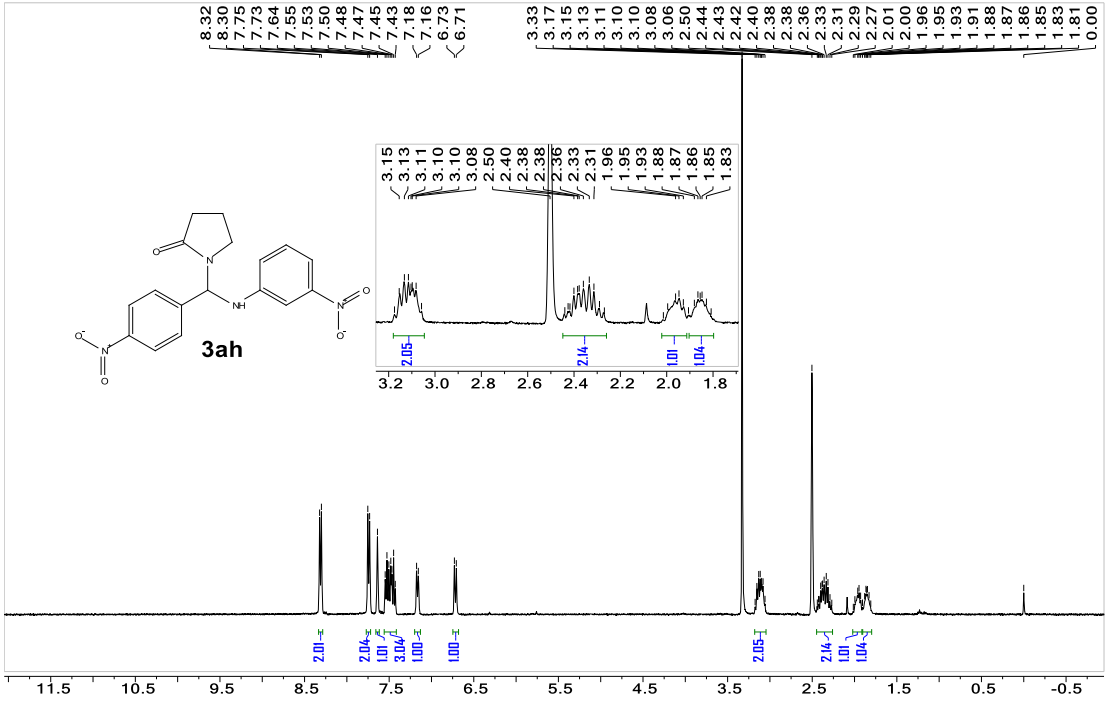


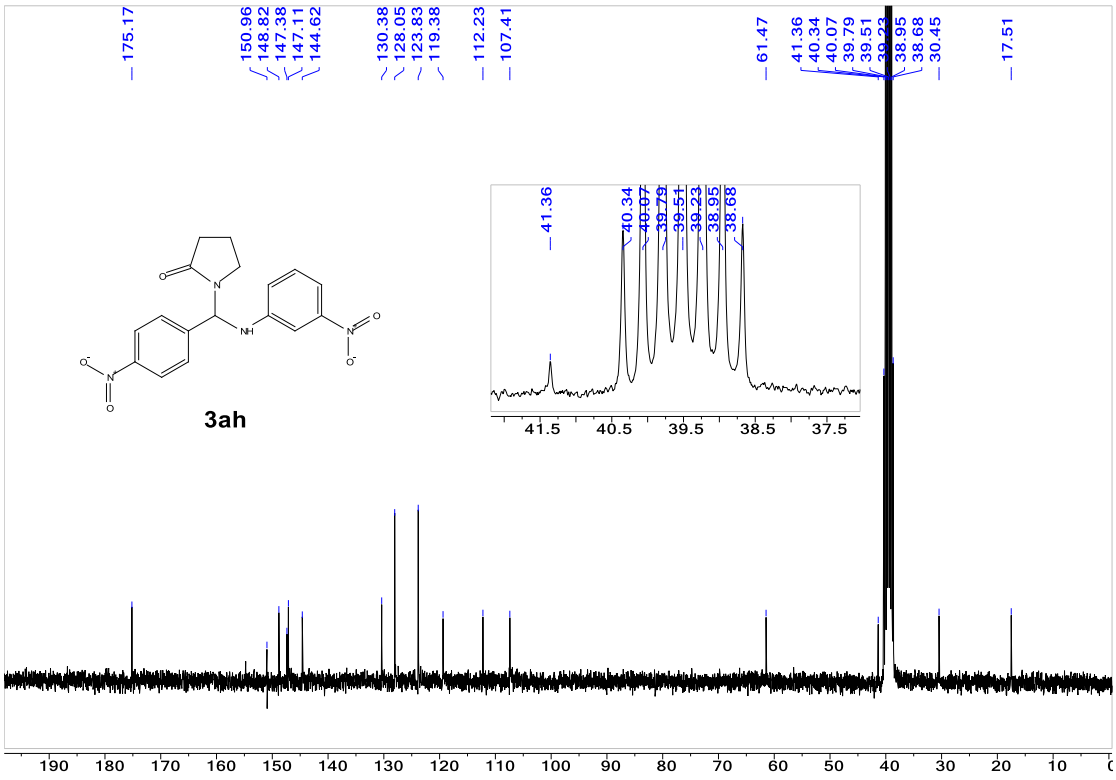
1-((4-Nitrophenyl)(4-nitrophenylamino)methyl)pyrrolidin-2-one (3ag):



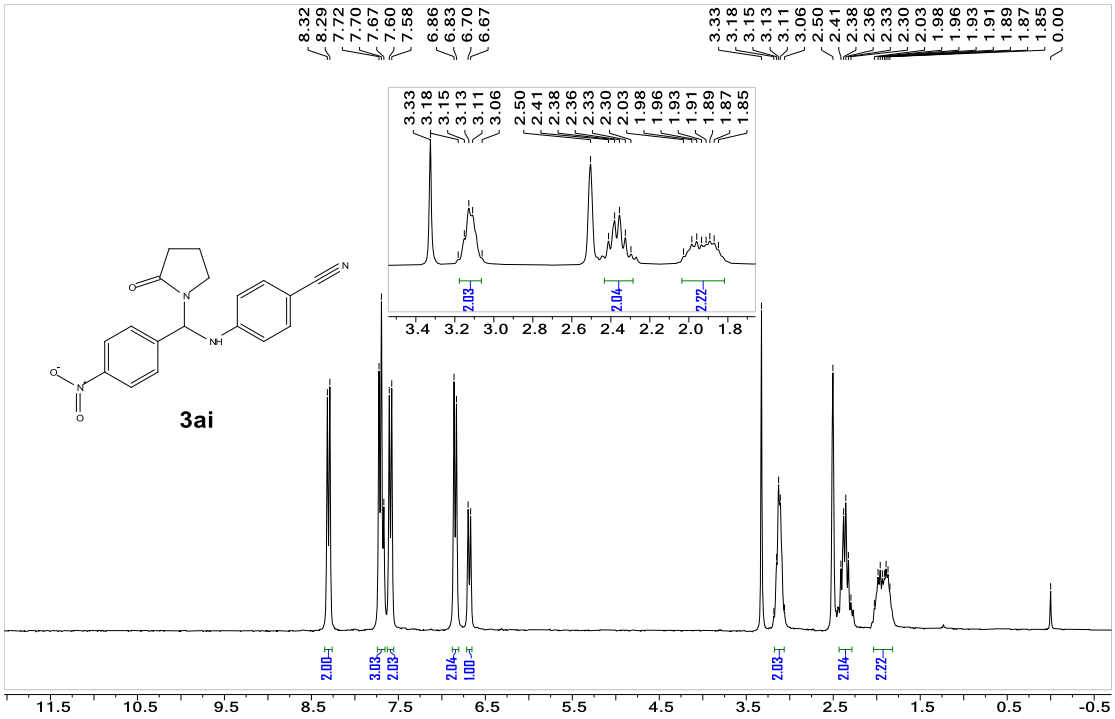


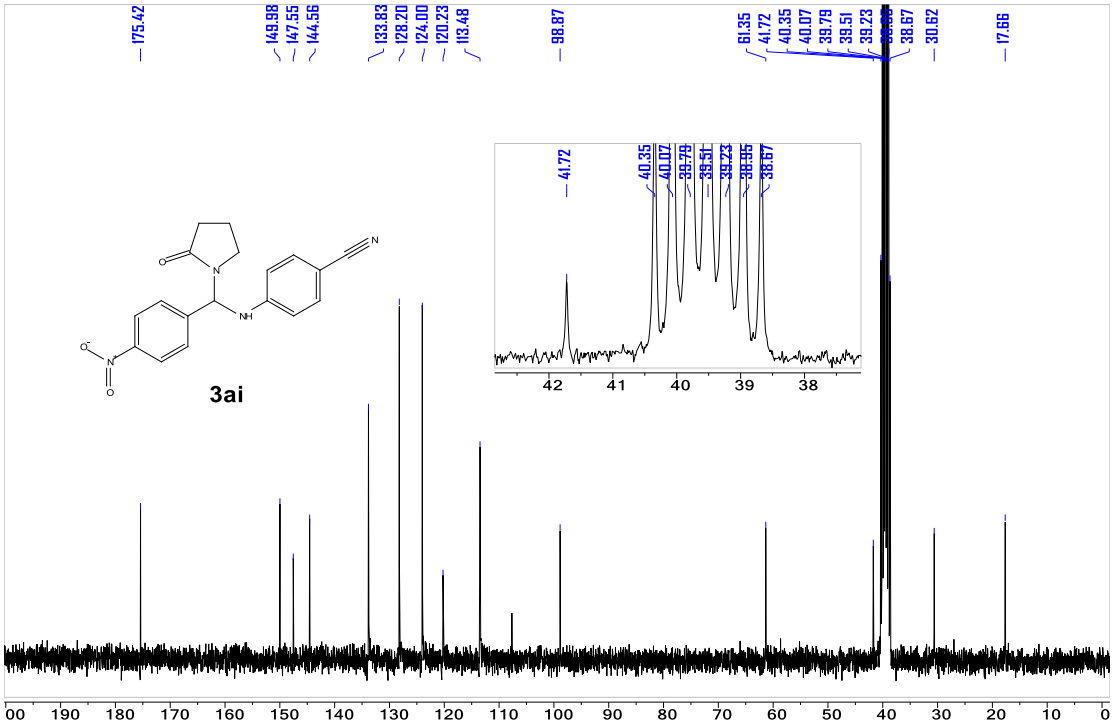
1-((4-Nitrophenyl)(3-nitrophenylamino)methyl)pyrrolidin-2-one (3ah):



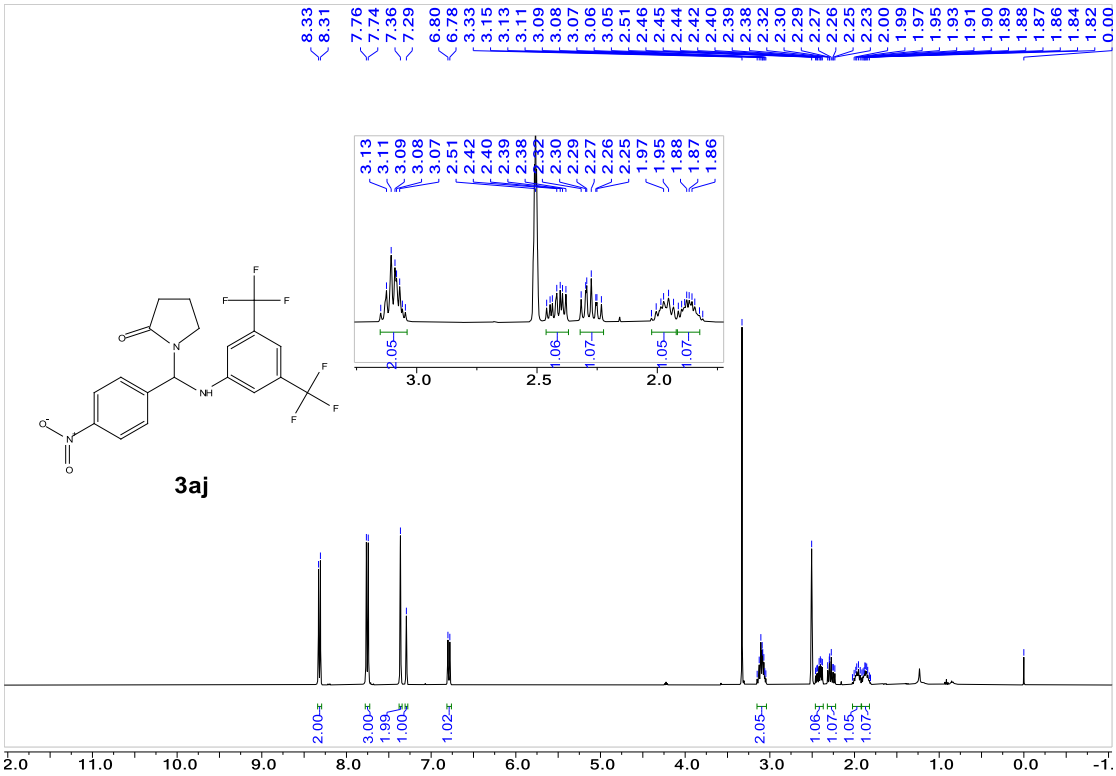


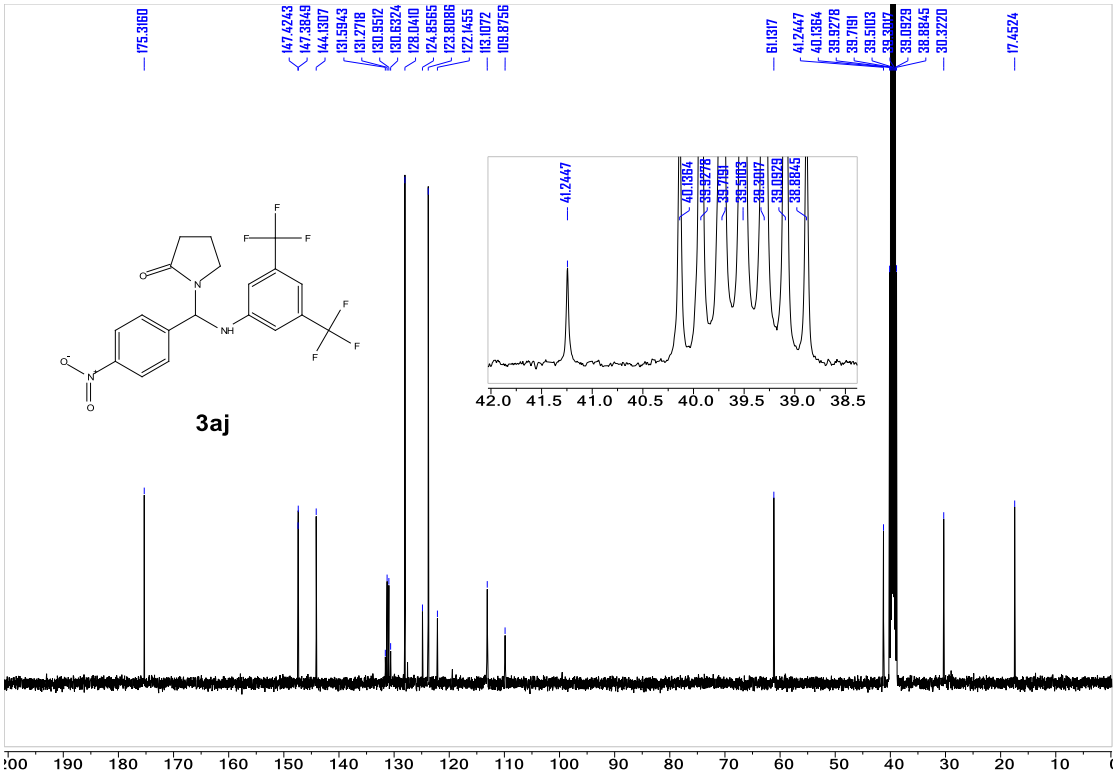
4-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methylamino)benzonitrile (3ai):



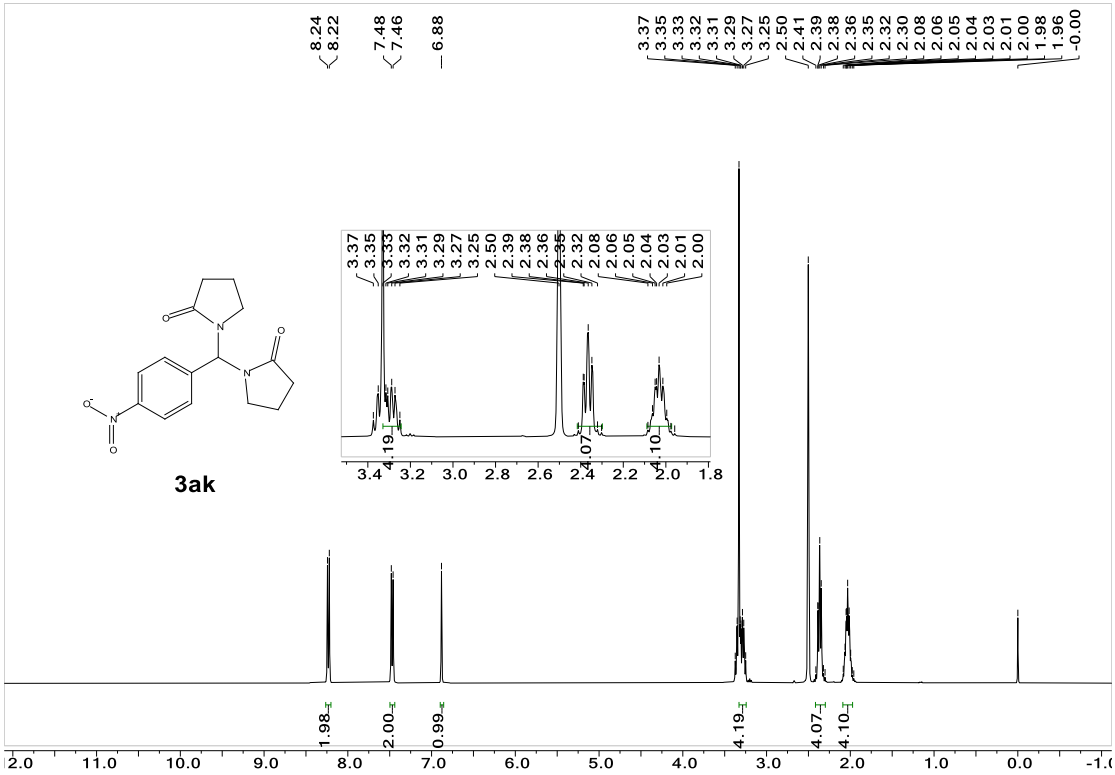


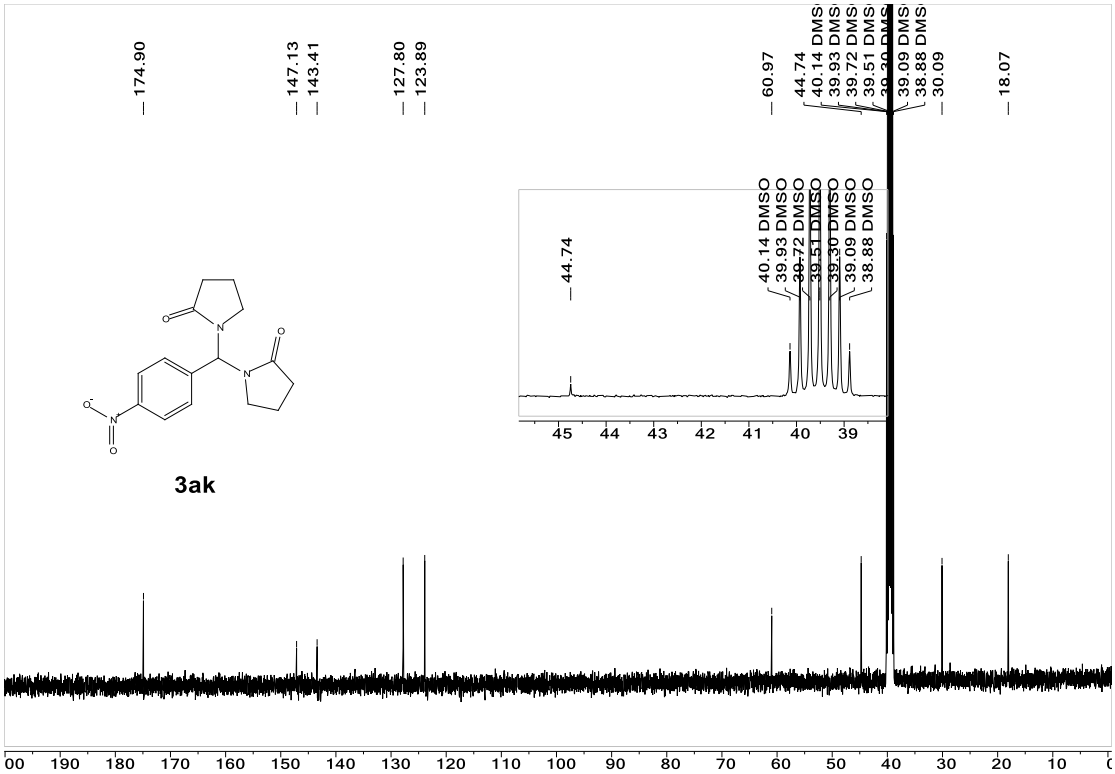
1-((3,5-Bis(trifluoromethyl)phenylamino)(4-nitrophenyl)methyl)pyrrolidin-2-one (3aj):



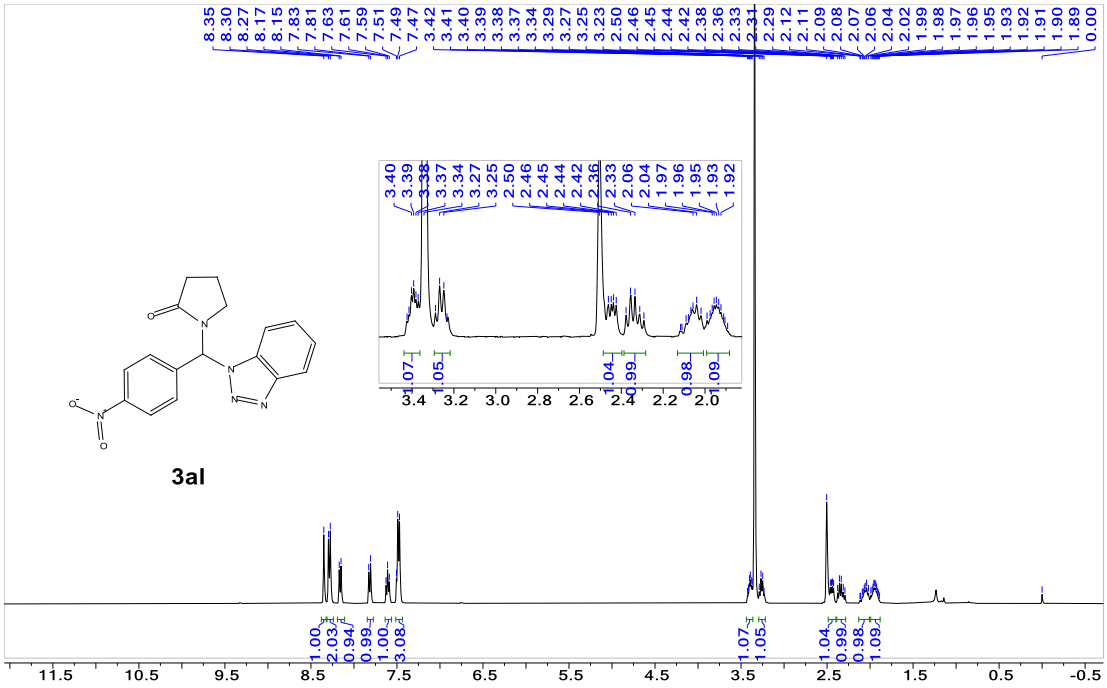


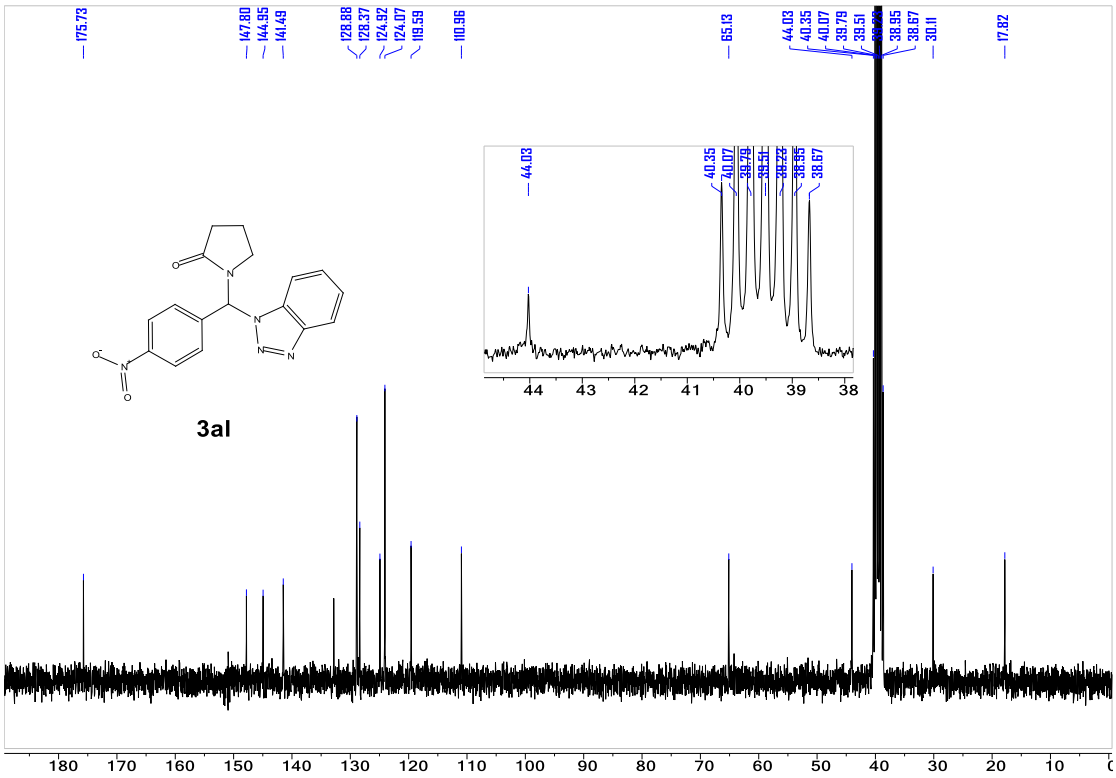
1,1'-((4-Nitrophenyl)methylene)dipyrrolidin-2-one (3ak):



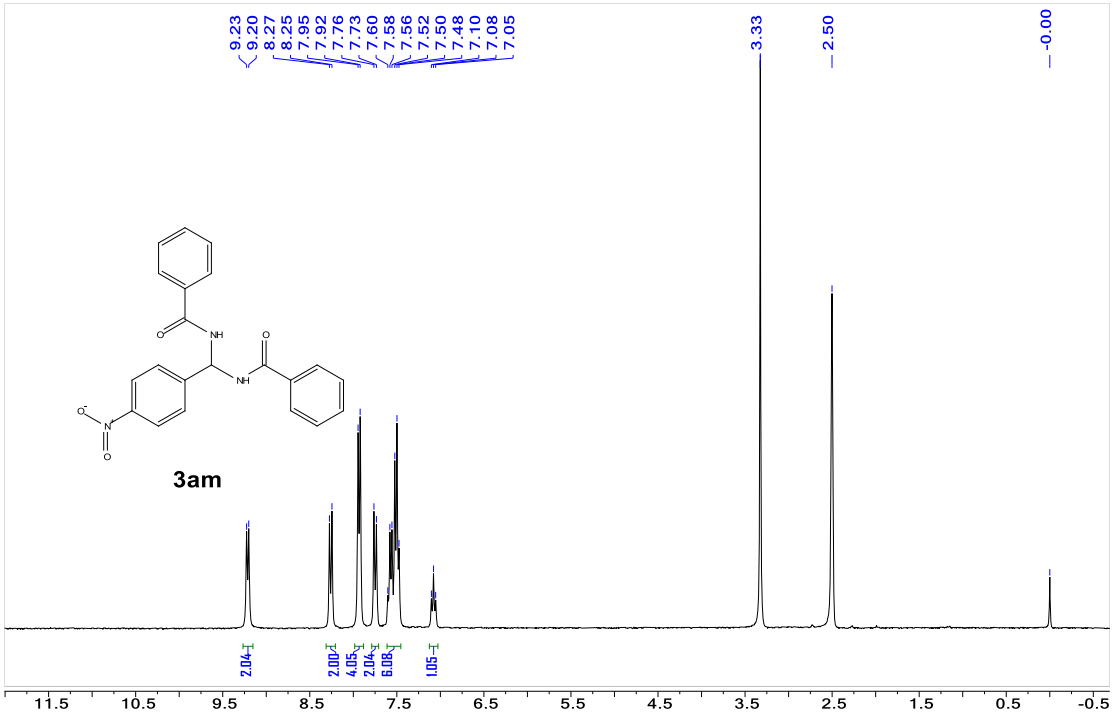


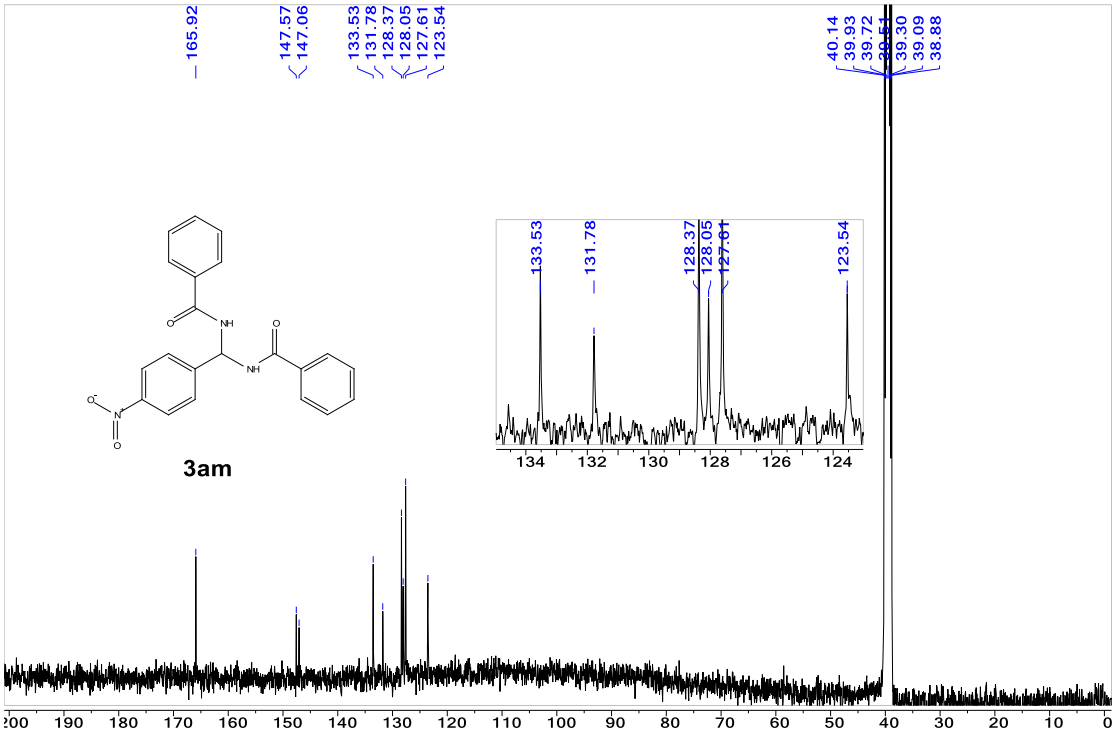
1-((1*H*-Benzo[*d*][1,2,3]triazol-1-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (3al):



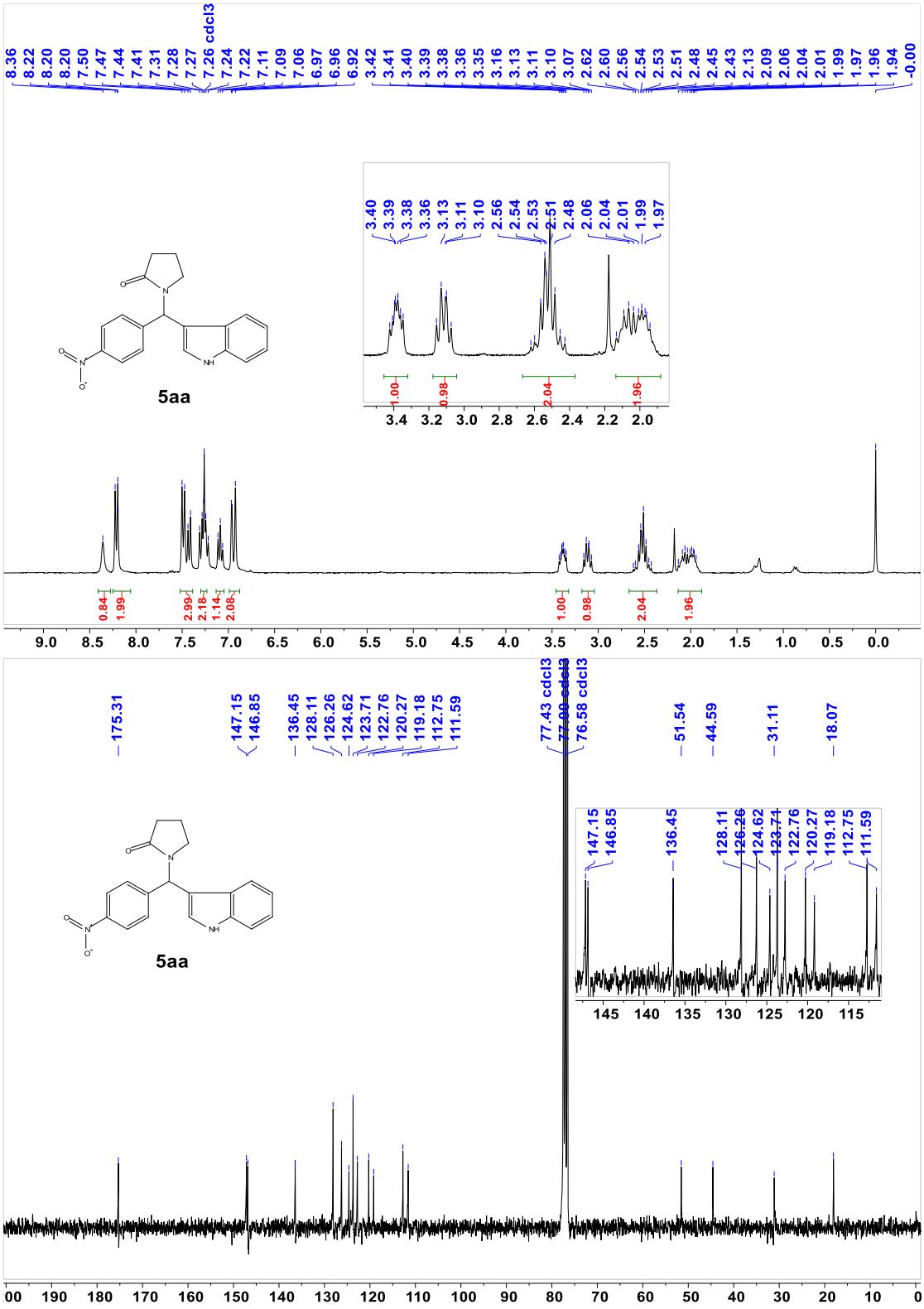


***N*,*N*'-((4-Nitrophenyl)methylene)dibenzamide (3am):**

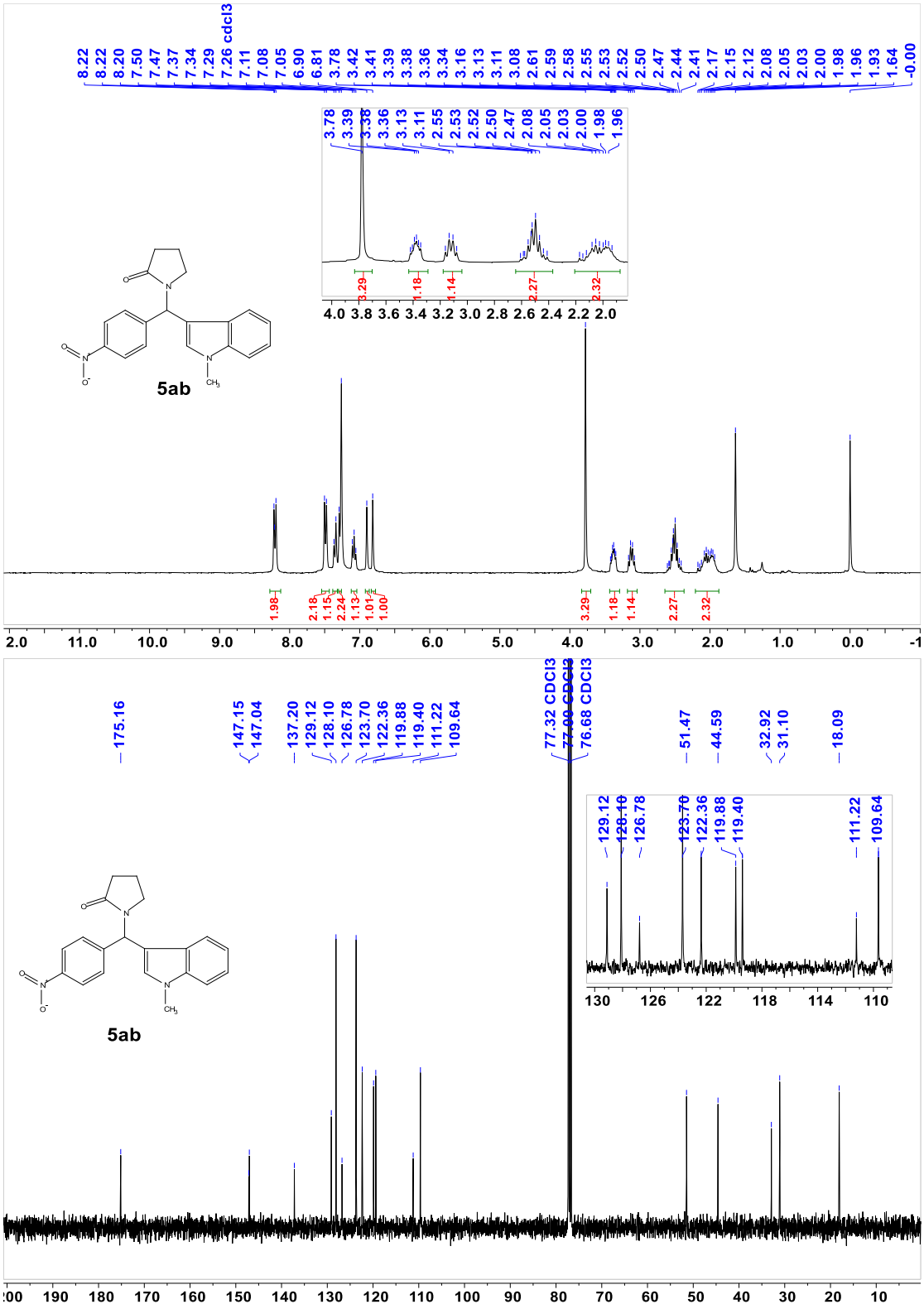




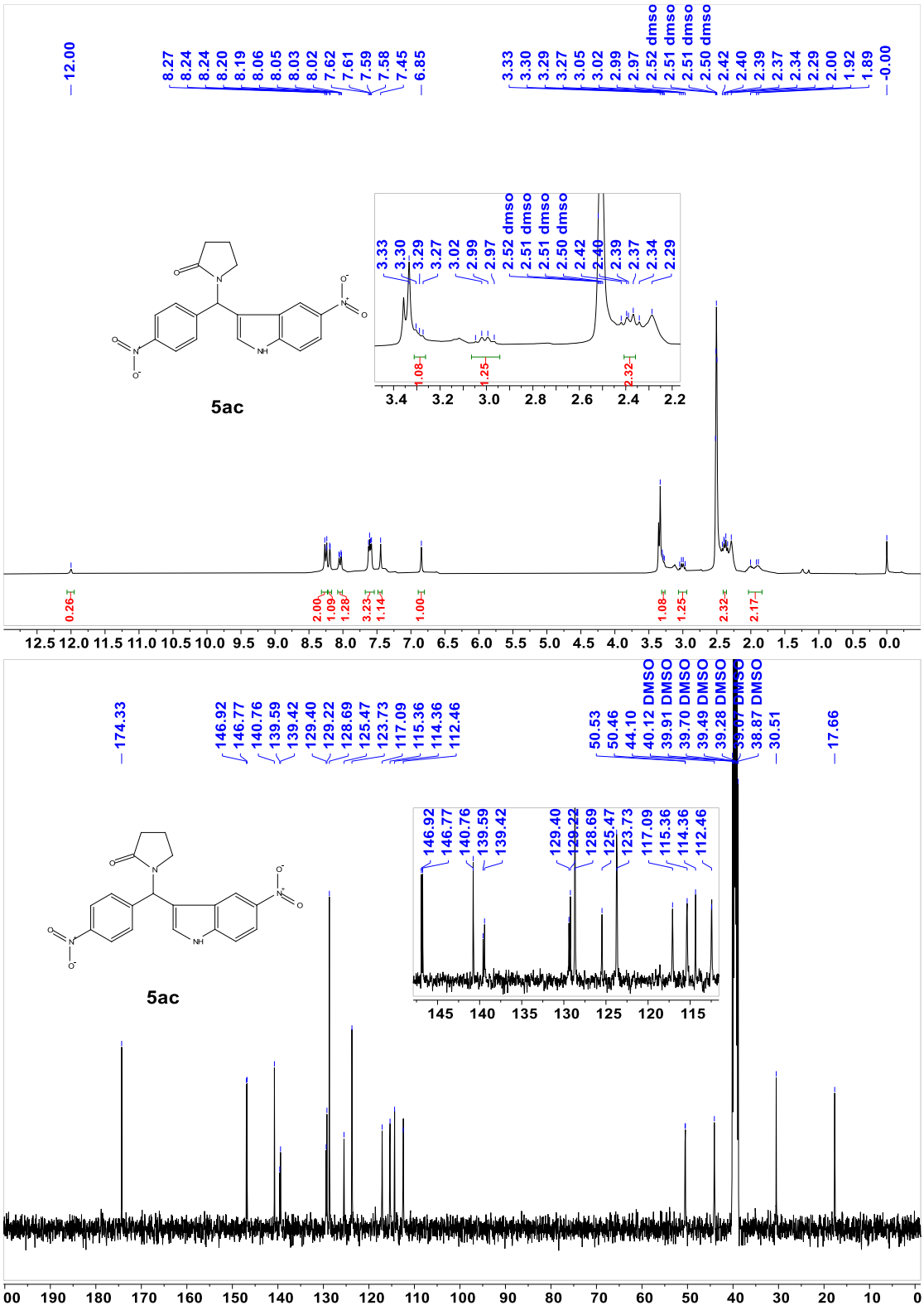
1-((1*H*-Indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5aa):



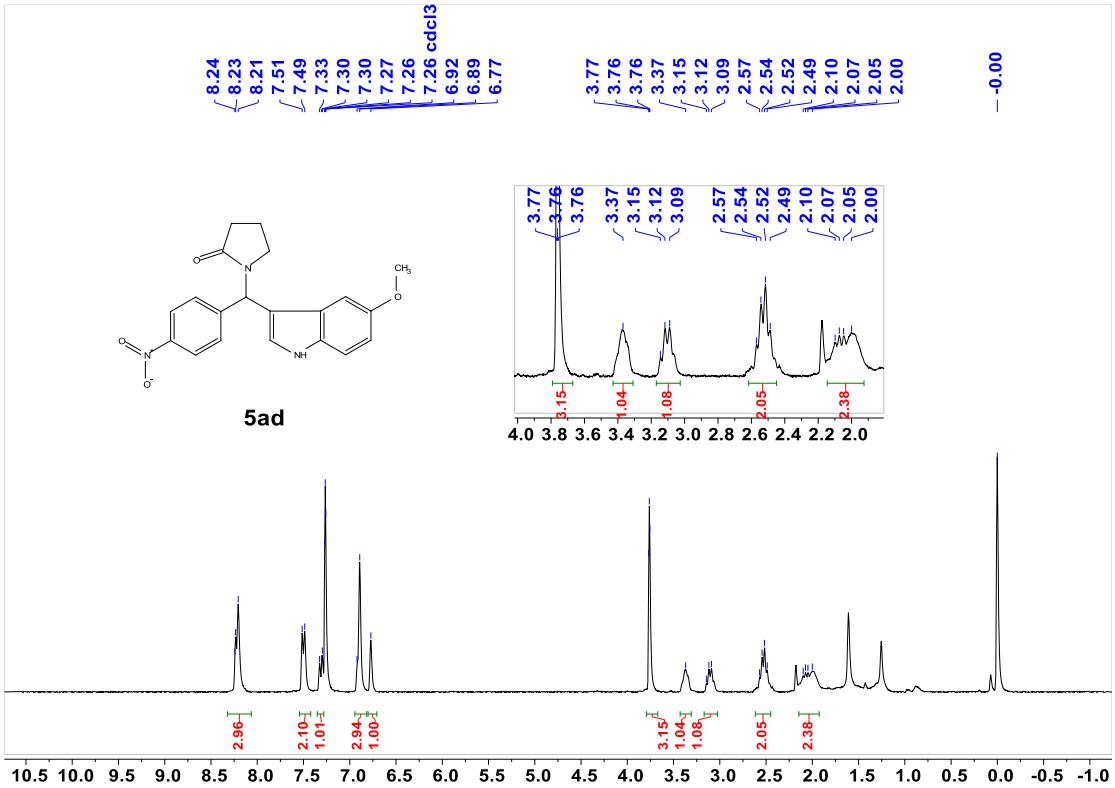
1-((1-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ab):

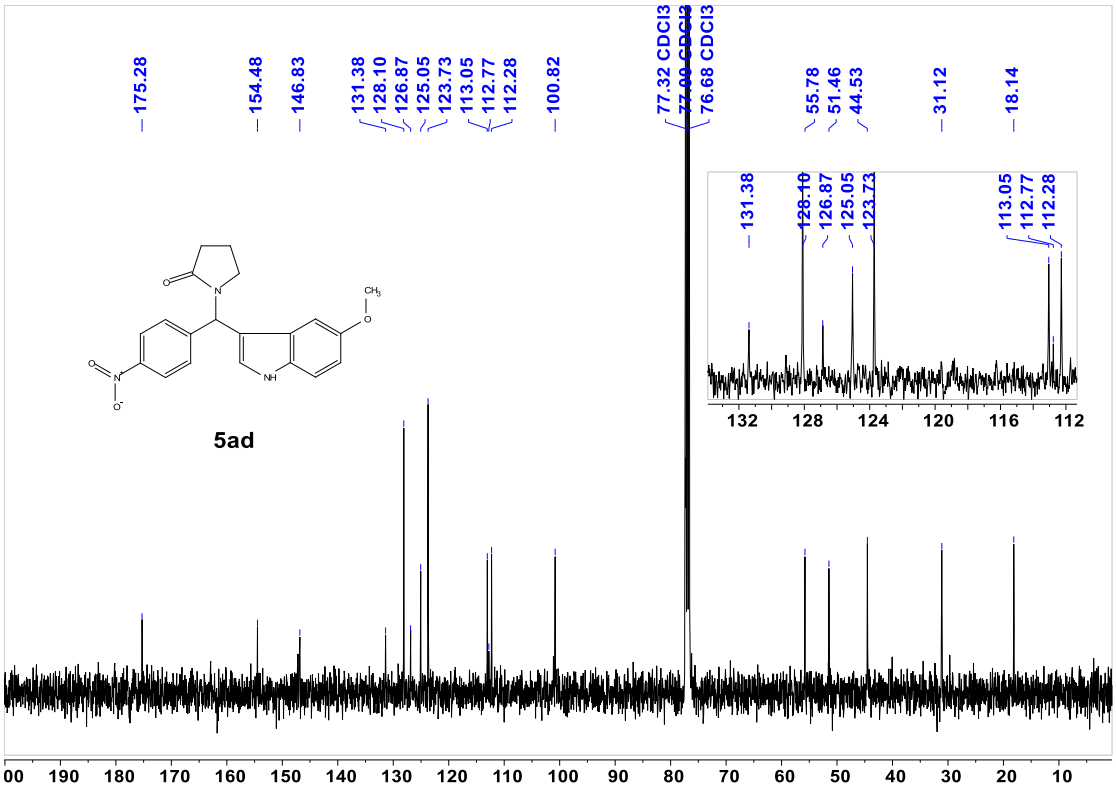


1-((5-Nitro-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ac):

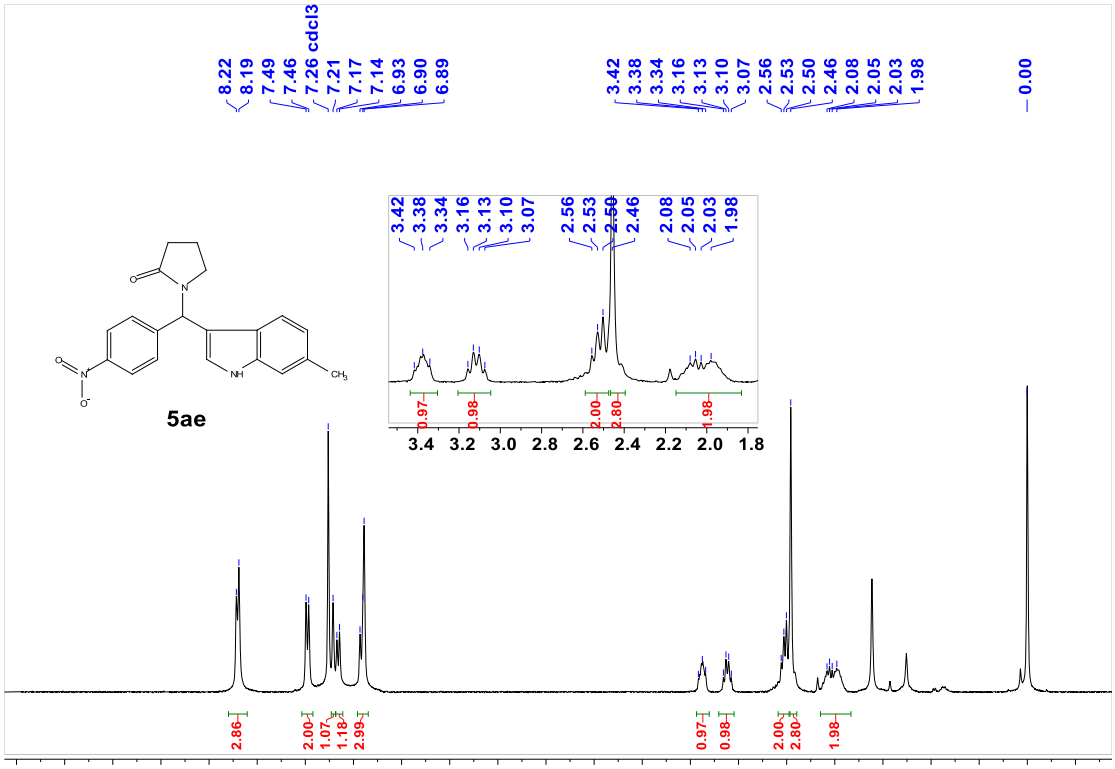


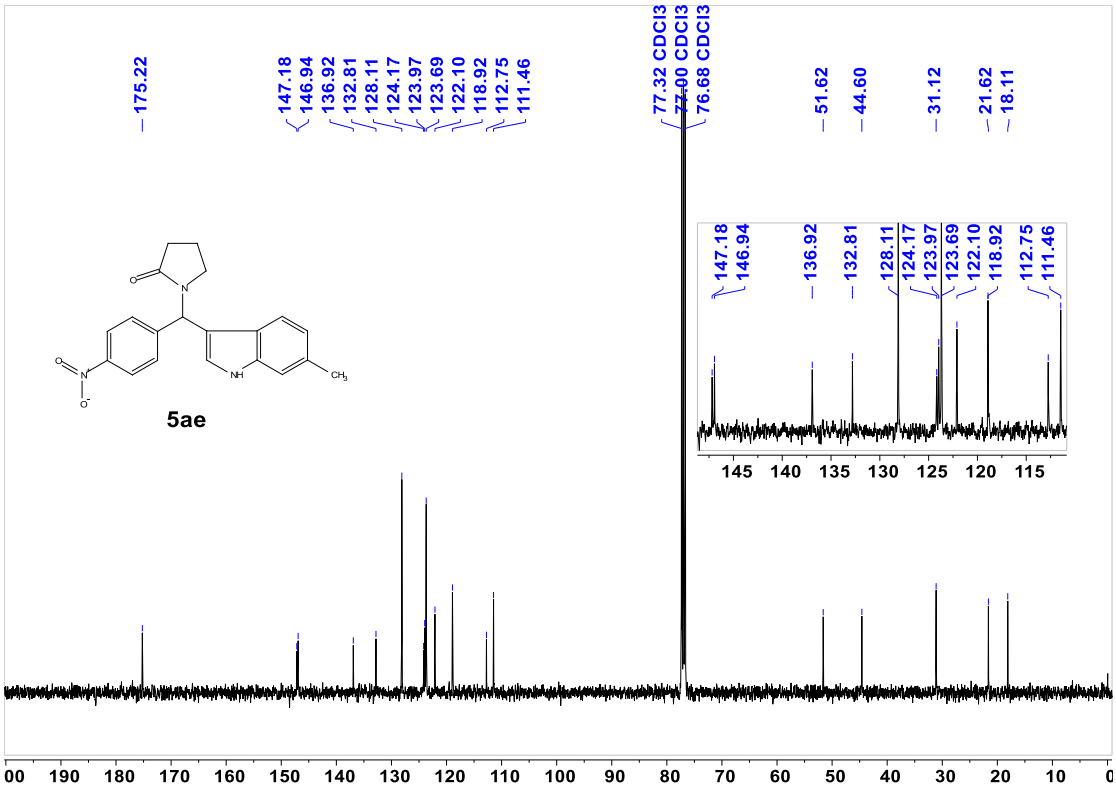
1-((5-Methoxy-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ad):



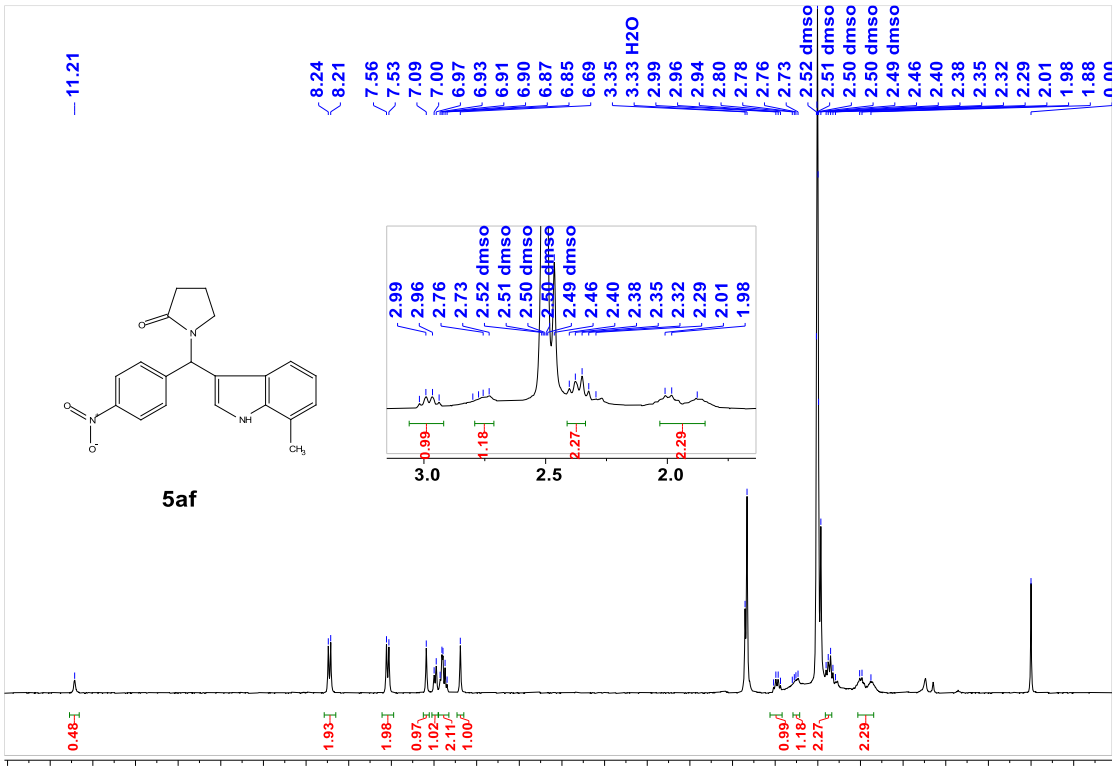


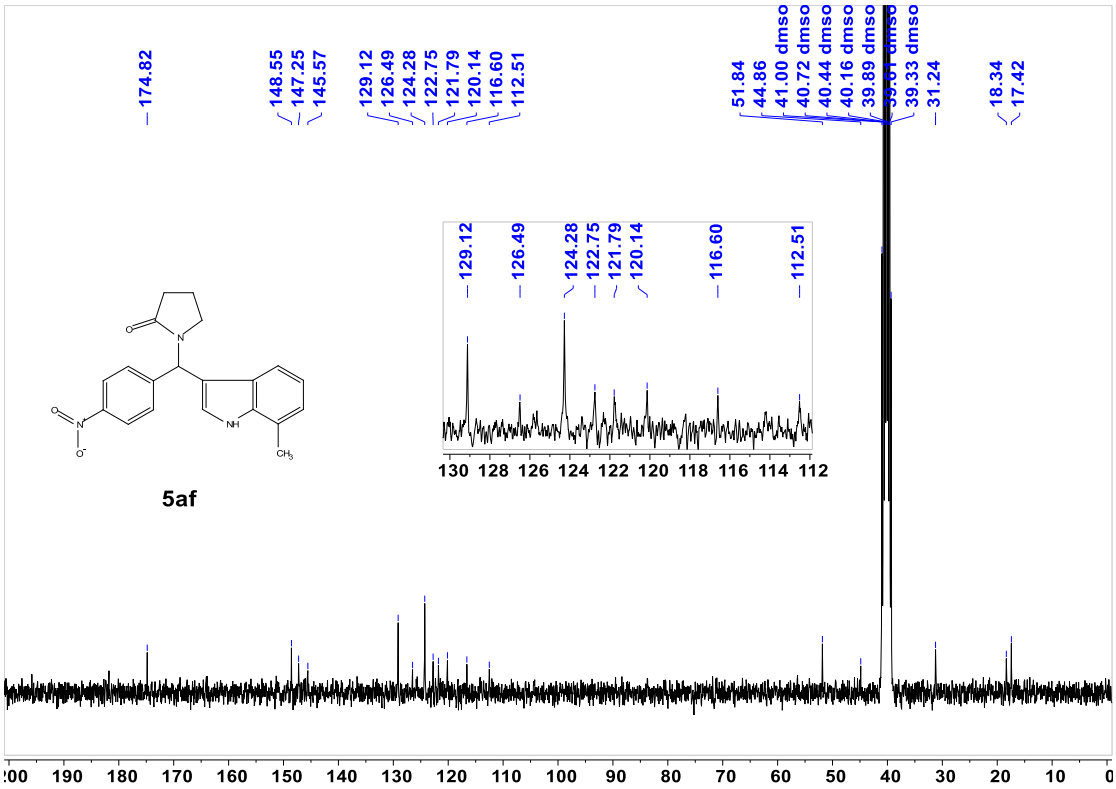
1-((6-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5ae):



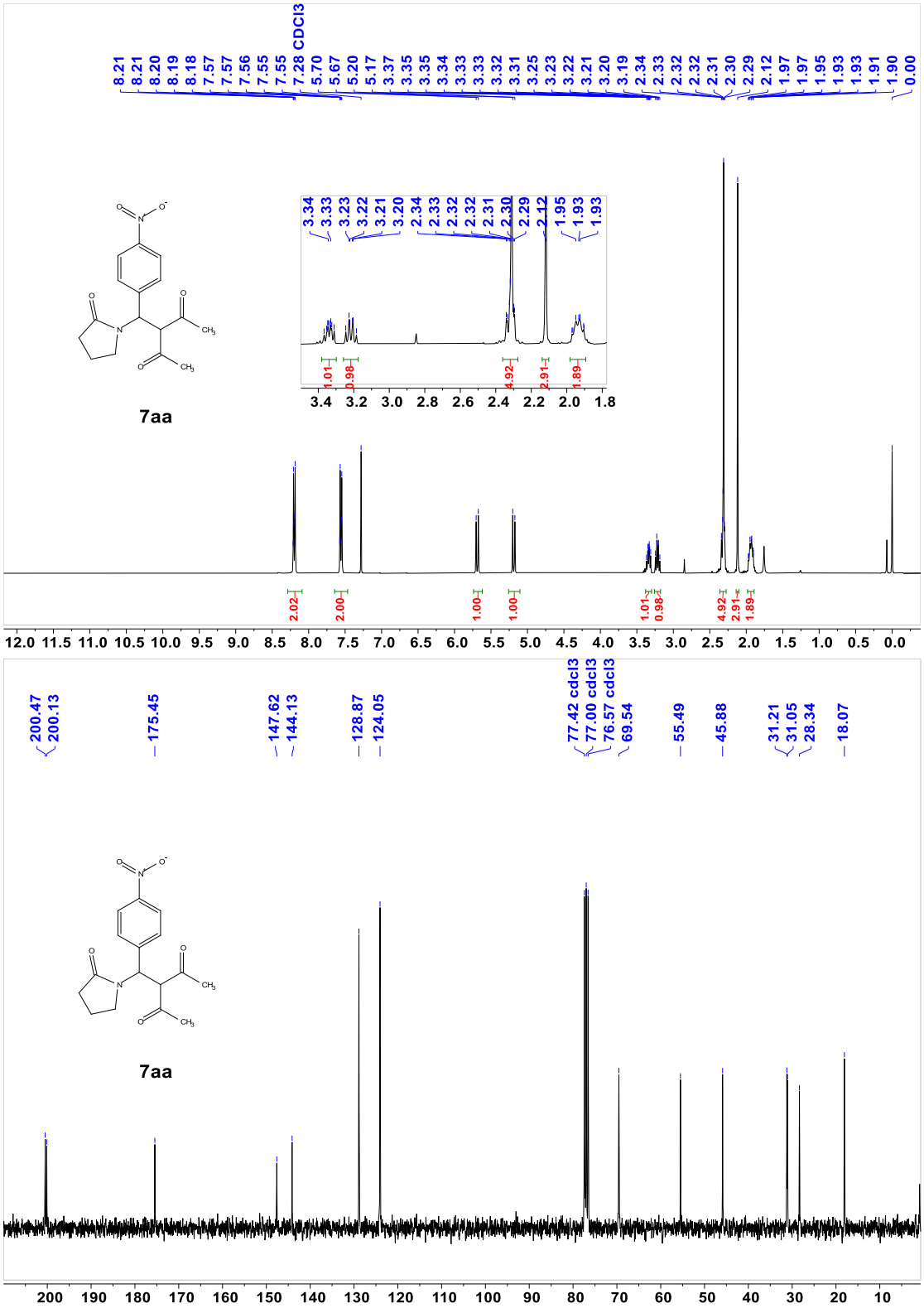


1-((7-Methyl-1*H*-indol-3-yl)(4-nitrophenyl)methyl)pyrrolidin-2-one (5af):

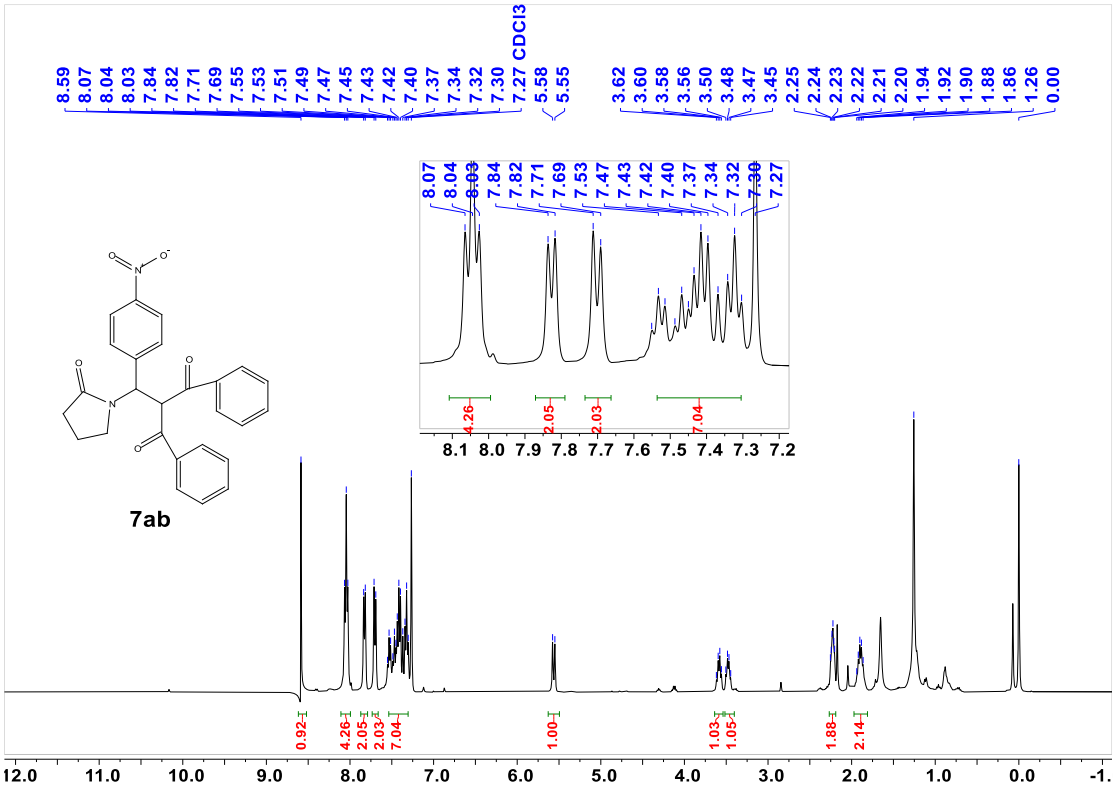


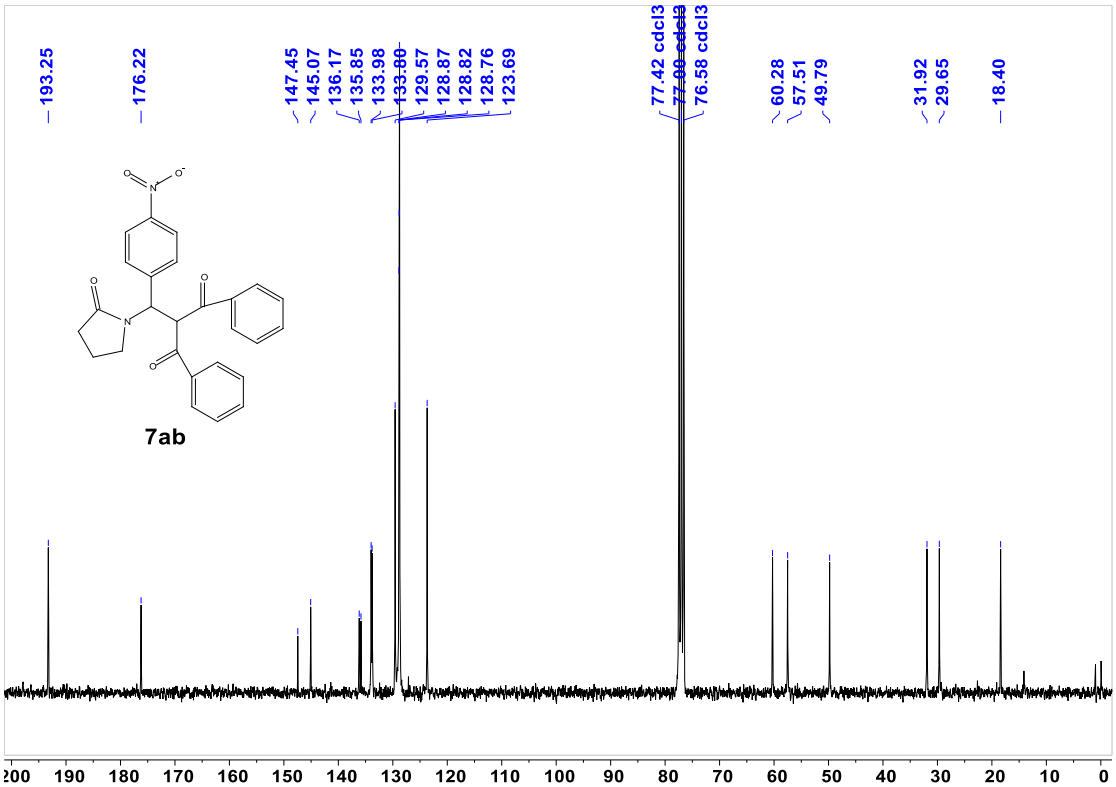


3-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)pentane-2,4-dione (7aa):

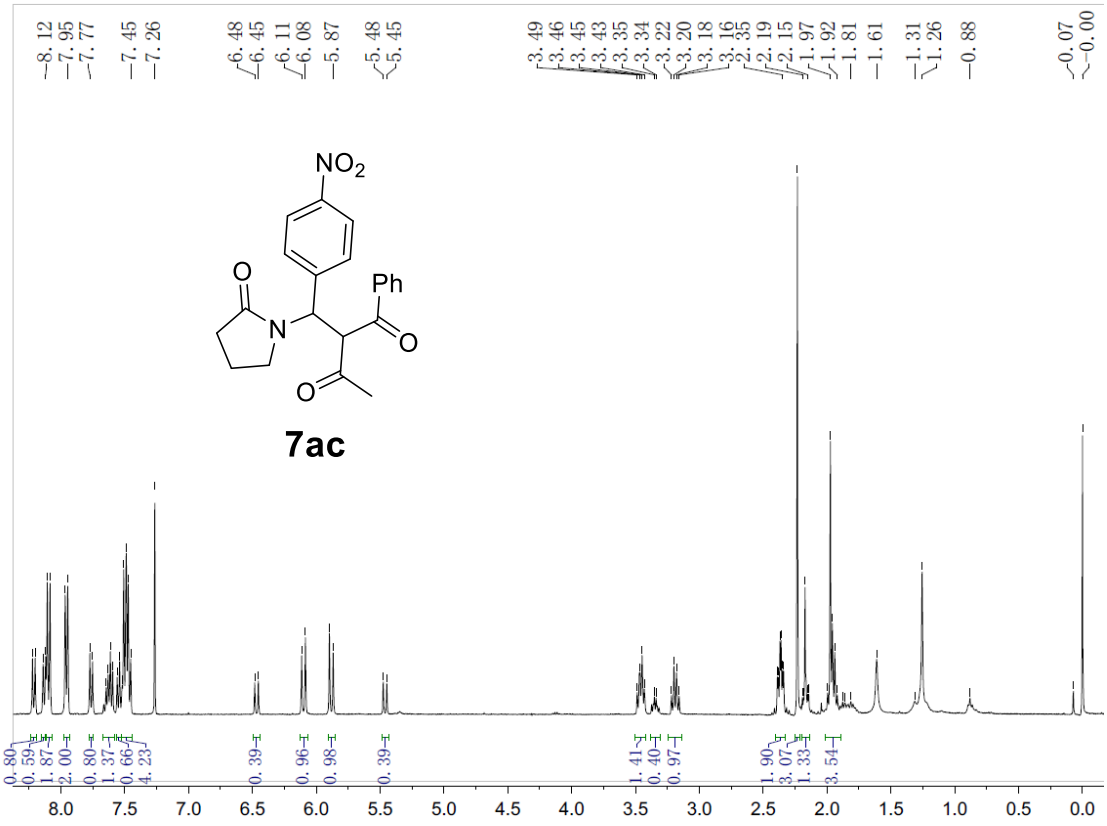


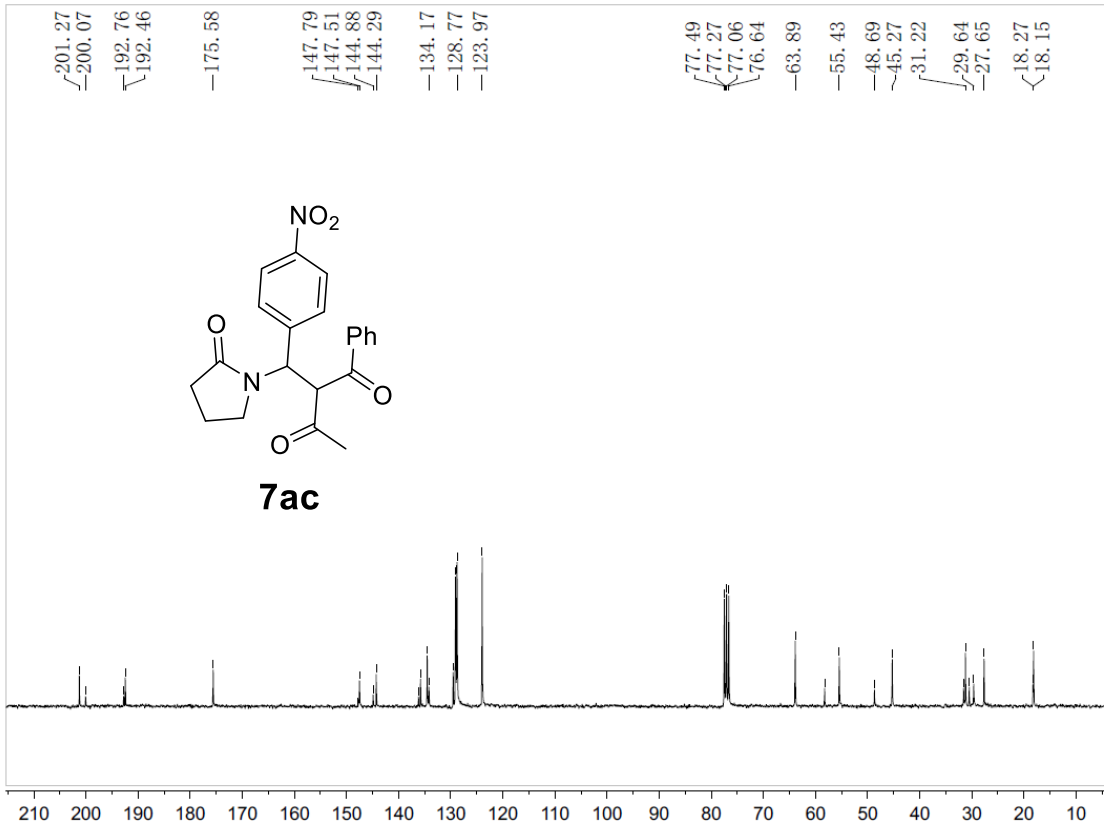
2-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)-1,3-diphenylpropane-1,3-dione (7ab):





2-((4-Nitrophenyl)(2-oxopyrrolidin-1-yl)methyl)-1-phenylbutane-1,3-dione (7ac):





1-((4-Nitrophenyl)(2,4,6-trimethoxyphenyl)methyl)pyrrolidin-2-one (9):