AN EFFICIENT SOLVENT- AND CATALYST-FREE SYNTHESIS OF BICYCLLIC PYRIDONES WITH HIGH MOLECULAR DIVERSITY VIA CASCADE REACTION

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Contents

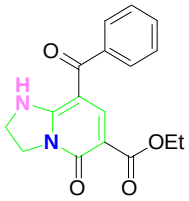
Spectroscopic Data of Bicyclic 2-Pyridones 1

^{1}H NMR and ^{13}C NMR Spectra for Bicyclic Pyridones 10

Reference 35

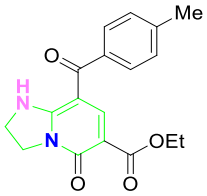
Spectroscopic Data of Bicyclic 2-Pyridones

8-Benzoyl-7-oxo-1,2,3,7-tetrahydro-imidazo[1,2-a]pyridine-6-carboxylic acid ethyl ester



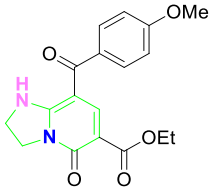
4aa: White needle-like crystals; Mp 226-228 ^{o}C (Ref. 234-236 ^{o}C[1]); IR (KBr) (ν_{max}, cm^{-1}) 3438, 3298, 2979, 1725, 1652, 1610, 1560, 1267, 1220, 1166, 868, 794 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.75 (br, 1H, NH), 8.51 (s, 1H, CH), 7.49-7.58 (m, 5H, ArH), 4.26-4.33 (m, 4H, CH_2CH_3 and NCH_2), 4.06 (t, J = 9.64, 2H, $NHCH_2$), 1.32 (t, J = 7.04, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.9, 165.0, 158.4, 158.0, 149.7, 138.2, 131.4, 128.6, 128.3, 107.1, 97.6, 60.7, 43.6, 43.2, 14.3; HRMS (ESI-TOF^{+}): *m/z* Calcd for $C_{17}H_{16}O_4N_2Na$ [(M+Na)$^{+}$] 335.1002; found, 335.0995.

8-(4-Methyl-benzoyl)-7-oxo-1,2,3,7-tetrahydro-imidazo[1,2-*a*]pyridine-6-carboxylic acid ethyl ester



4ba: White needle-like crystals; Mp 203-205 ^{o}C (Ref. 203-205 ^{o}C[1]); IR (KBr) (ν_{max}, cm^{-1}) 3429, 2930, 1727, 1648, 1611, 1552, 1267, 1159, 1074, 861 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.79 (br, 1H, NH), 8.54 (s, 1H, CH), 7.49 (d, J = 7.84, 2H, ArH), 7.30 (d, J = 8.40, 2H, ArH), 4.28-4.31 (m, 4H, CH_2CH_3 and $CONCH_2$), 4.02-4.06 (m, 2H, $NHCH_2$), 2.44 (s, 3H, $ArCH_3$), 1.34 (t, J = 7.04, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 192.8, 165.1, 158.3, 158.0, 149.8, 142.1, 135.4, 129.2, 128.5, 106.8, 97.8, 60.7, 43.6, 43.1, 21.6, 14.4; HRMS (ESI-TOF^{+}): *m/z* Calcd for $C_{18}H_{18}O_4N_2Na$ [(M+Na)$^{+}$] 349.1159; found, 349.1156.

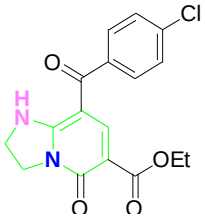
8-(4-Methoxy-benzoyl)-7-oxo-1,2,3,7-tetrahydro-imidazo[1,2-*a*]pyridine-6-carboxylic acid ethyl ester



4ca: White needle-like crystals; Mp 213-215 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3452, 3340, 2984, 1727, 1669, 1609, 1561, 1256, 1230, 1174, 842 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.64 (br, 1H, NH), 8.49 (s, 1H, CH), 7.51 (d, J = 8.40, 2H, ArH), 6.91 (d, J = 8.40, 2H, ArH), 4.20-4.25 (m, 4H, CH_2CH_3 and $CONCH_2$), 3.97 (t, J = 9.56, 2H,

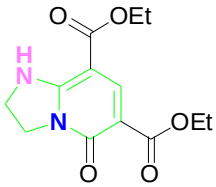
NH*CH*$_2$), 3.81 (s, 3H, OCH_3), 1.26 (t, *J* = 7.08, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 190.8, 164.1, 161.4, 157.4, 148.7, 129.7, 129.6, 112.8, 105.6, 96.7, 59.7, 54.5, 42.6, 42.1, 13.4; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{18}H_{18}O_5N_2Na$ [(M+Na)$^+$] 365.1108; found, 365.1107.

8-(4-Chloro-benzoyl)-7-oxo-1,2,3,7-tetrahydro-imidazo[1,2-*a*]pyridine-6-carboxylic acid ethyl ester



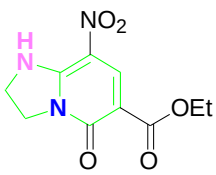
4da: White needle-like crystals; Mp 278-279 ^{o}C (Ref. 280-281 ^{o}C[1]); IR (KBr) (ν_{max}, cm^{-1}) 3441, 3138, 2826, 1725, 1653, 1609, 1577, 1561, 1263, 1222, 1199, 1122, 864, 695 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.79 (br, 1H, NH), 8.42 (s, 1H, CH), 7.51 -7.45 (m, 4H, ArH), 4.25-4.32 (m, 4H, *CH*$_2$CH$_3$ and CONCH$_2$), 4.04 (t, *J* = 9.96, 2H, NH*CH*$_2$), 1.32 (t, *J* = 7.08, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 191.4, 164.8, 158.3, 157.9, 149.1, 137.7, 136.5, 129.7, 128.9, 107.1, 97.4, 60.8, 43.6, 43.2, 14.4; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{17}H_{15}O_4N_2ClNa$ [(M+Na)$^+$] 369.0613; found, 369.0617.

7-Oxo-1,2,3,7-tetrahydro-imidazo[1,2-*a*]pyridine-6,8-dicarboxylic acid diethyl ester



4ea: White needle-like crystals; Mp 211-213 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3390, 2986, 2318, 1728, 1667, 1644, 1567, 1238, 1176, 1150, 861, 783, 695 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.62 (br, 1H, NH), 7.56 (s, 1H, NH), 4.23-4.35 (m, 6H, *CH*$_2$CH$_3$, and CONCH$_2$), 3.97 (t, *J* = 9.48, 2H, NCH$_2$), 1.36 (m, 6H, CH$_2$*CH*$_3$); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 165.5, 165.0, 158.5, 157.4, 147.4, 107.2, 88.7, 60.6, 60.6, 44.0, 43.0, 14.4; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{13}H_{16}O_5N_2Na$ [(M+Na)$^+$] 303.0951; found, 303.0950.

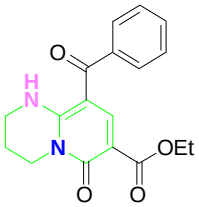
8-Nitro-7-oxo-1,2,3,7-tetrahydro-imidazo[1,2-*a*]pyridine-6-carboxylic acid ethyl ester



4fa: Red needle-like crystals; Mp 274-275 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3331, 3082, 2987,

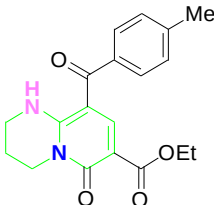
1734, 1652, 1618, 1571, 1276, 1161, 1103, 863, 764, 702 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 9.76 (br, 1H, NH), 8.61 (s, 1H, CH), 4.19 (q, J = 7.04, 2H, CH_2CH_3), 4.09 (t, J = 9.00, 2H, $CONCH_2$), 3.88 (t, J = 10.20, 2H, NCH_2), 1.25 (q, J = 7.04, 3H, CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 168.2, 162.0, 157.2, 145.6, 118.1, 111.8, 65.4, 49.3, 48.8, 19.4; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{10}H_{11}O_5N_3Na$ $[(M+Na)^+]$ 276.0591; found, 276.0591.

9-Benzoyl-8-oxo-1,3,4,8-tetrahydro-2*H*-pyrido[1,2-*a*]pyrimidine-7-carboxylic acid ethyl ester



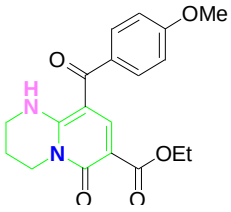
4ga: White needle-like crystals; Mp 178-180^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3442, 3112, 2972, 1735, 1697, 1613, 1589, 1252, 1179, 1136, 864, 760, 698 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 11.18 (br, 1H, NH), 8.41 (s, 1H, CH), 7.45 (d, J = 3.24, 5H, ArH), 4.23 (t, J = 7.00, 2H, CH_2CH_3), 4.05 (d, J = 4.56, 2H, $CONCH_2$), 3.51 (s, 2H, $NHCH_2$), 2.09 (s, 2H, CH_2), 1.26 (t, J = 7.04, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.8, 165.3, 158.6, 155.8, 149.6, 139.1, 131.0, 128.4, 128.4, 103.8, 98.2, 60.6, 39.4, 38.6, 19.1, 14.3; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{18}H_{18}O_4N_2Na$ $[(M+Na)^+]$ 349.1159; found, 349.1155.

9-(4-Methyl-benzoyl)-8-oxo-1,3,4,8-tetrahydro-2*H*-pyrido[1,2-*a*]pyrimidine-7-carboxylic acid ethyl ester



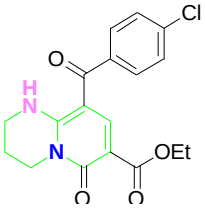
4ha: White needle-like crystals; mp 183-185 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3442, 3123, 2974, 1697, 1611, 1589, 1180, 1137, 1067, 1016, 865, 779, 738 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 11.24 (br, 1H, NH), 8.50 (s, 1H, CH), 7.43 (d, J = 7.64, 2H, COArH), 7.28 (d, J = 7.08, 2H, ArH), 4.29 (q, J = 7.08, 2H, CH_2CH_3), 4.11 (t, J = 5.76, 2H, $CONCH_2$), 3.56 (s, 2H, $NHCH_2$), 2.43 (s, 3H, $ArCH_3$), 2.15 (t, J = 5.32, 2H, CH_2), 1.32 (t, J = 7.08, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.8, 165.5, 158.6, 155.8, 149.7, 141.5, 136.4, 129.1, 128.6, 103.6, 98.3, 60.6, 39.3, 38.6, 21.5, 19.2, 14.4; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{19}H_{20}O_4N_2Na$ $[(M+Na)^+]$ 363.1315; found, 363.1313.

9-(4-Methoxy-benzoyl)-8-oxo-1,3,4,8-tetrahydro-2*H*-pyrido[1,2-*a*]pyrimidine-7-carboxylic acid ethyl ester



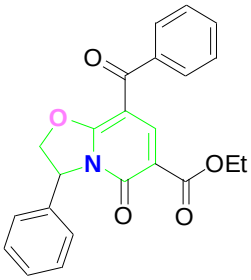
4ia: White needle-like crystals; mp 150-152 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3439, 3378, 2875, 1732, 1654, 1598, 1215, 1173, 1114, 865, 781, 734 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 11.21 (br, 1H, NH), 8.54 (s, 1H, CH), 7.54 (d, J = 7.96, 2H, COArH), 6.99 (d, J = 8.04, 2H, ArH), 4.31 (q, J =7.16, 2H, CH_2CH_3), 4.13 (t, J = 5.72, 2H, $CONCH_2$), 3.90 (s, 3H, $ArOCH_3$), 3.57 (s, 2H, $NHCH_2$), 2.16 (t, J = 5.52, 2H, CH_2), 1.34 (t, J = 7.04, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 193.9, 165.5, 162.1, 158.6, 155.8, 149.6, 131.6, 130.7, 113.7, 103.4, 98.3, 60.6, 55.5, 39.4, 38.6, 19.2, 14.4; HRMS (ESI-TOF^{+}): *m/z* Calcd for $C_{19}H_{20}O_5N_2Na$ [(M+Na)$^{+}$] 379.1264; found, 379.1261.

9-(4-Chloro-benzoyl)-8-oxo-1,3,4,8-tetrahydro-2*H*-pyrido[1,2-*a*]pyrimidine-7-carboxylic acid ethyl ester



4ja: White needle-like crystals; Mp 278-279 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3441, 3138, 2826, 1725, 1653, 1609, 1577,1263, 1199, 1122, 864, 695 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 10.80 (br, 1H, NH), 8.12 (s, 1H, CH), 7.60 (d, J = 8.20, 2H, COArH), 7.51(d, J = 8.24, 2H, ArH), 4.09(q, J = 7.00, 2H, $COOCH_2$), 3.90(t, J = 5.08, 2H, $CONCH_2$), 3.51(s, 2H, $NHCH_2$), 2.01(s, 2H, CH_2), 1.15(t, J = 7.04, 3H, CH_3) ; ^{13}C NMR (100 MHz, DMSO-d_6): δ = 192.3, 164.8, 157.9, 155.6, 148.7, 138.6, 135.9, 130.4, 129.0, 102.7, 98.1, 60.0, 39.8, 38.9, 18.8, 14.7; HRMS (ESI-TOF^{+}): *m/z* Calcd for $C_{18}H_{17}O_4N_2ClNa$ [(M+Na)$^{+}$] 383.0769; found, 383.0764.

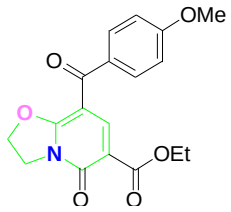
8-Benzoyl-7-oxo-3-phenyl-2,3-dihydro-7*H*-oxazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



4ka: White needle-like crystals; Mp 171-172 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3382, 3135, 1735, 1669, 1644, 1593, 1235, 1168, 1138, 863, 794, 756, 700 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ= 8.68 (s, 1H, CH), 7.73 (d, J= 7.60, 1H, COArH), 7.59 (t, J= 7.56, 1H, COArH), 7.50 (t, J= 7.44, 2H, COArH), 7.50 (t, J= 7.44, 2H, COArH), 7.36-7.42

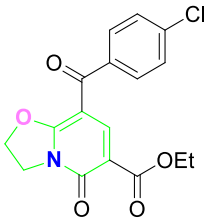
(m, 3H, ArH), 7.32 (d, *J*= 8.24, 2H, ArH), 5.82 (dd, J_1= 8.80, J_2= 3.04, 1H, OCH_2), 5.11 (t, *J*= 9.16, 1H, ArCHN), 4.85 (dd, J_1= 9.20, J_2= 3.08, 1H, OCH_2), 4.23-4.34 (m, 2H, CH_2CH_3), 1.30 (t, *J*= 7.09, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ= 189.9, 164.1, 161.2, 156.3, 149.7, 138.1, 136.7, 132.5, 129.4, 129.0, 128.5, 126.6, 112.3, 99.0, 78.1, 61.1, 59.5, 14.3; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{23}H_{19}O_5NNa$ [$(M+Na)^+$] 412.1155; found, 412.1158.

8-(4-Methoxy-benzoyl)-7-oxo-2,3-dihydro-7*H*-oxazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



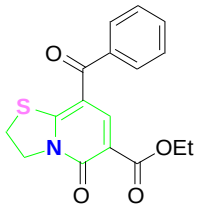
4la: White needle-like crystals; Mp 165-167 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3381, 3135, 1735, 1658, 1636, 1600, 1257, 1234, 1159, 863, 799 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.61 (s, 1H, CH), 7.71 (d, *J* = 8.08, 2H, ArH), 6.96 (d, *J* = 7.96, 2H, ArH), 4.91 (t, *J* = 8.80, 2H, OCH_2), 4.33-4.41 (m, 4H, CH_2CH_3 and $CONCH_2$), 3.87 (t, *J* = 6.44, 3H, OCH_3), 1.35 (t, *J* = 7.20, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 188.5, 164.2, 163.3, 160.9, 157.1, 149.5, 131.6, 130.3, 113.7, 113.5, 111.4, 99.6, 70.4, 61.1, 55.5, 43.8, 14.3; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{18}H_{17}O_6NNa$ [$(M+Na)^+$] 366.0948; found, 366.0950.

8-(4-Chloro-benzoyl)-7-oxo-2,3-dihydro-7*H*-oxazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



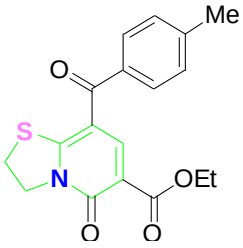
4ma: White needle-like crystals; Mp 162-164 ^{o}C; IR (KBr) (ν_{max}, cm^{-1}) 3435, 2995, 1726, 1647, 1548, 1255, 1175, 860, 678 cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.61 (s, 1H, CH), 7.63 (d, *J* = 8.08, 2H, ArH), 7.45 (d, *J* = 8.12, 2H, ArH), 4.92 (t, *J* = 8.88, 2H, OCH_2), 4.33-4.42 (m, 4H, CH_2CH_3 and $CONCH_2$), 1.36 (t, *J* = 7.20, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 188.4, 163.9, 161.2, 157.0, 149.1, 138.8, 136.4, 130.4, 128.8, 111.9, 105.6, 99.0, 70.6, 61.2, 43.7, 14.3; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{17}H_{14}O_5NClNa$ [$(M+Na)^+$] 370.0453; found, 370.0453.

8-Benzoyl-7-oxo-2,3-dihydro-7*H*-thiazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



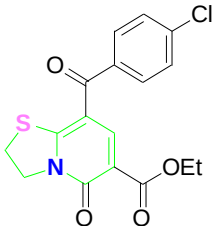
4na: Yellow-brown needle-like crystals; Mp 177-179 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3423, 3063, 2990, 1796, 1644, 1623, 1562, 1246, 1194, 1137, 864, 703 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.24 (s, 1H, CH), 7.57-7.65 (m, 5H, ArH), 4.47 (t, J = 8.32, 2H, CH_2CH_3), 4.16 (q, J = 7.04, 2H, $CONCH_2$), 3.49 (t, J = 8.32, 2H, SCH_2), 1.19 (t, J = 7.08, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 191.0, 165.3, 164.3, 157.3, 146.1, 137.7, 132.4, 129.1, 128.9, 112.6, 110.4, 60.8, 51.3, 28.2, 14.6; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{17}H_{15}O_4NNaS$ [$(M+Na)^+$] 352.0614; found, 352.0613.

8-(4-Methoxy-benzoyl)-7-oxo-2,3-dihydro-7*H*-thiazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



4oa: Light yellow granule crystals; Mp 202-204 ºC ; IR (KBr) (ν_{max}, cm^{-1}) 3438, 2992, 1699, 1677, 1572, 1266, 1193, 1149, 870, 795, 719 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.26 (s, 1H, CH), 7.52 (d, J = 7.76, 2H, ArH), 7.38 (d, J = 7.84, 2H, ArH), 4.46 (t, J = 8.28, 2H, CH_2CH_3), 4.16 (d, J = 7.04, 2H, $CONCH_2$), 3.47 (t, J = 8.28, 2H, SCH_2), 2.41 (s, 3H, $ArOCH_3$), 1.20 (t, J = 7.08, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 190.7, 165.1, 164.4, 157.3, 146.1, 142.7, 134.9, 129.6, 129.2, 112.5, 110.6, 60.8, 51.3, 28.2, 21.6, 14.6; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{18}H_{17}O_4NNaS$ [$(M+Na)^+$] 366.0770; found, 366.0770.

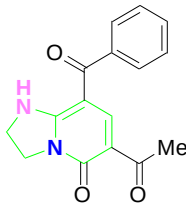
8-(4-Chloro-benzoyl)-7-oxo-2,3-dihydro-7*H*-thiazolo[3,2-*a*]pyridine-6-carboxylic acid ethyl ester



4pa: Light yellow granule crystals; Mp 217-219 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3440, 3016, 1741, 1665, 1630, 1575, 1244, 1190, 1137, 862, 770, 699 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.20 (s, 1H, CH), 7.64 (m, 4H, ArH), 4.47 (t, J = 8.32, 2H, CH_2CH_3), 4.18 (d, J = 7.08, 2H, $CONCH_2$), 3.48 (t, J = 8.36, 2H, SCH_2), 1.21 (t, J = 7.08, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 189.9, 165.4, 164.3, 157.2, 145.9, 137.2, 136.4, 130.9, 129.3, 112.8, 110.3, 60.9, 51.3, 28.3, 14.6; HRMS (ESI-TOF$^+$):

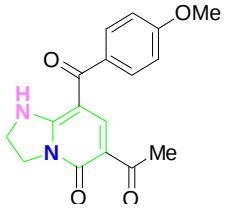
m/z Calcd for $C_{17}H_{14}O_4NClNaS$ [$(M+Na)^+$] 386.0224; found, 386.0225.

6-Acetyl-8-benzoyl-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridin-5-one



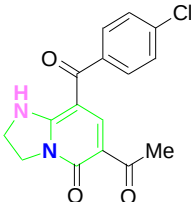
4ab: White needle-like crystals; Mp 293-294 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3412, 3292, 1665, 1598, 1159, 1073, 977, 953, 860, 548 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.24 (br, 1H, NH), 7.46-7.50 (m, 3H, ArCH), 7.39-7.43 (m, 2H, ArCH), 5.65 (d, J = 8.40, 1H, CH), 4.22-4.27 (m, 2H, $CONCH_2$), 3.93 (t, J = 9.40, 2H, $NHCH_2$), 1.70 (d, J = 6.40, 3H, $COCH_3$); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.8, 160.6, 157.3, 152.1, 143.4, 130.9, 128.4, 127.5, 109.3, 99.0, 77.3, 43.4, 42.7, 23.6; HRMS (ESI-TOF$^+$): m/z Calcd for $C_{16}H_{15}O_3N_2$ [$(M+H)^+$] 283.1077; found, 283.1075.

6-Acetyl-8-(4-methoxy-benzoyl)-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridin-5-one



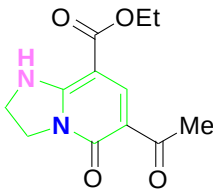
4cb: White needle-like crystals; Mp 191-193 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3409, 1658, 1607, 1443, 1165, 1075, 975, 950, 861 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.76 (br, 1H, NH), 7.49-7.52 (m, 2H, ArCH), 6.91-6.94 (m, 2H, ArCH), 5.67 (d, J = 6.40, 1H, CH), 4.24 (t, J = 8.80, 2H, $CONCH_2$), 3.90 (t, J = 9.20, 2H, $NHCH_2$), 3.86 (s, 3H, ArOCH3), 1.79 (s, 3H, $COCH_3$); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 194.0, 162.4, 160.6, 156.8, 152.1, 134.4, 130.2, 113.7, 108.9, 99.0, 55.4, 43.6, 42.6, 23.3 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{17}H_{17}O_4N_2$ [$(M+H)^+$] 313.1183; found, 313.1178.

6-Acetyl-8-(4-chloro-benzoyl)-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridin-5-one



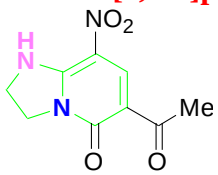
4db: White needle-like crystals; Mp 126-127 ºC; IR (KBr) (vmax, cm^{-1}) 3332, 1602, 1577, 1549, 1451, 1160, 1088, 951, 862, 848 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 9.24 (br, 1H, NH), 7.71 (d, J = 8.52, 2H, ArCH), 7.42(d, J = 8.52, 2H, ArCH), 5.24 (s, 1H, CH), 3.59 (d, J = 6.08, 2H, $CONCH_2$), 3.46 (d, J = 5.88, 2H, $NHCH_2$), 3.34 (s, 3H, $COCH_3$); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 194.2, 193.5, 160.2, 157.9, 139.9, 137.2, 134.2, 133.5, 129.6, 128.8, 98.3, 44.9, 44.8, 21.2 cm^{-1}; HRMS (ESI-TOF$^+$): m/z Calcd for $C_{16}H_{14}O_3N_2Cl$ [$(M+H)^+$] 317.0687; found, 317.0689.

6-Acetyl-5-oxo-1,2,3,5-tetrahydro-imidazo[1,2-*a*]pyridine-8-carboxylic acid ethyl ester



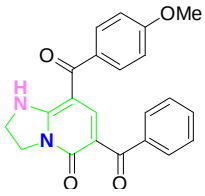
4eb: White needle-like crystals; Mp 136-138 ºC; IR (KBr) (νmax, cm^{-1}) 3388, 1676.1, 1630, 1584, 1551, 1442, 1370, 1157, 1074, 951, 861 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.73 (br, 1H, NH), 5.65 (s, 1H, CH), 4.27 (q, J = 7.12, 2H, CH_2CH_3), 4.20 (t, J = 8.84, 2H, $CONCH_2$), 3.87 (t, J = 9.28, 2H, $NHCH_2$), 2.36 (s, 3H, $COCH_3$), 1.35 (t, J = 7.12, 3H, CH_2CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 167.5, 160.6, 157.3, 153.7, 108.7, 88.7, 60.2, 43.7, 42.3, 23.8, 14.4 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{12}H_{15}O_4N_2$ $[(M+H)^+]$ 251.1026; found, 251.1026.

6-Acetyl-8-nitro-2,3-dihydro-1H-imidazo[1,2-*a*]pyridin-5-one



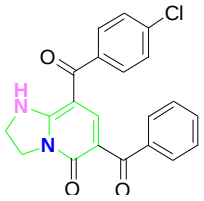
4fb: White needle-like crystals; Mp 174-176 ºC; IR (KBr) (νmax, cm^{-1}) 3428, 3296, 1678, 1649, 1612, 1560, 1209, 1161, 1075, 952, 861 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 9.82 (br, 1H, NH), 8.59 (s, 1H, CH), 4.10-4.15 (m, 2H, $CONCH_2$), 3.89-3.94 (m, 2H, $NHCH_2$), 2.49 (s, 3H, $COCH_3$); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 198.7, 164.3, 157.4, 144.3, 144.1, 119.7, 49.3, 49.0, 44.1, 35.5 cm^{-1}; HRMS (ESI-TOF$^+$): m/z Calcd for $C_9H_{19}O_4N_3$ $[(M+H)^+]$ 224.0666; found, 224.0668.

6-Benzoyl-8-(4-methoxy-benzoyl)-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridin-5-one



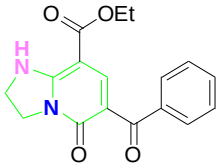
4cc: White needle-like crystals; Mp 262-264 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3430, 1584, 1440, 1159, 1074, 950, 860, 548 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 7.81 (br, 1H, NH), 7.77-7.80 (m, 1H, ArH), 7.77 (s, 1H, CH), 7.56 (t, J = 7.44, 1H, ArH), 7.48 (t, J = 7.40, 1H, ArH), 7.43 (t, J = 7.80, 2H, ArH), 7.34 (t, J = 7.80, 2H, ArH), 7.14 (d, J = 8.64, 1H, ArH), 6.78 (d, J = 8.64, 1H, ArH), 4.60 (s, 4H, $NHCH_2$ and $CONCH_2$), 3.72(s, 3H, OCH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 194.2, 193.5, 160.2, 157.9, 148.5, 139.9, 137.7, 137.2, 134.2, 133.5, 130.6, 129.6, 129.4, 129.2, 128.8, 125.0, 124.3, 114.6, 98.3, 55.6, 44.9, 44.8 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{22}H_{19}O_4N_2$ $[(M+H)^+]$ 375.1339; found, 375.1340.

6-Benzoyl-8-(4-chloro-benzoyl)-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridin-5-one



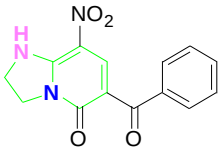
4dc: White needle-like crystals; Mp 275-277 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3413, 1685, 1654, 1585, 1448, 1161, 1074, 947, 860, 547 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 7.79 (br, 1H, NH), 7.74-7.77 (m, 1H, ArH), 7.73 (s, 1H, CH), 7.57-7.63 (m, 2H, ArH), 7.46-7.51 (m, 2H, ArH), 7.42-7.44 (m, 2H, ArH), 7.24 (d, J = 8.36, 2H, ArH), 4.44 (s, 4H, NHCH_2 and CONCH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 193.4, 157.7, 148.4, 147.5, 139.3, 137.7, 134.6, 134.3, 133.5, 131.9, 130.9, 129.6, 129.4, 128.8, 125.1, 124.7, 98.0, 45.0, 44.7 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{21}H_{16}O_3N_2Cl$ [(M+H)$^+$] 379.0844; found, 379.0848.

6-Benzoyl-5-oxo-1,2,3,5-tetrahydro-imidazo[1,2-*a*]pyridine-8-carboxylic acid ethyl ester



4ec: White needle-like crystals; Mp 187-189 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3456, 1701, 1658, 1631, 1563, 1448, 1230, 1078, 996, 547 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 8.26 (br, 1H, NH), 7.88-7.91 (m, 3H, ArH), 7.77-7.79 (m, 2H, ArH), 6.29 (s, 1H, CH), 4.86 (t, J = 8.76, 2H, CONCH_2), 4.52 (t, J = 9.20, 2H, NHCH_2), 4.44 (q, J = 7.12, 2H, CH_2CH_3), 1.32 (t, J = 7.12, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 167.0, 160.3, 157.1, 156.1, 141.3, 127.4, 127.3, 127.2, 109.2, 87.8, 59.7, 43.9, 42.7, 13.3 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{17}H_{17}O_4N_2$ [(M+H)$^+$] 313.1183; found, 313,1184.

6-Benzoyl-8-nitro-2,3-dihydro-1H-imidazo[1,2-*a*]pyridin-5-one



4fc: White needle-like crystals; Mp 234-236 ºC; IR (KBr) (ν_{max}, cm^{-1}) 3406, 1664, 1644, 1622, 1605, 1563, 1487, 1161, 1121, 1073, 984, 948, 860, 548 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 9.79 (br, 1H, NH), 8.29 (s, 1H, CH), 7.71-7.73 (m, 2H, ArH), 7.59-7.63 (m, 1H, ArH), 7.48 (t, J = 7.76, 2H, ArH), 4.12 (t, J = 9.08, 2H, CONCH_2), 3.93 (t, J = 9.20, 2H, NHCH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 192.2, 158.7, 152.6, 139.5, 138.5, 133.0, 129.5, 128.6, 116.7, 113.8, 44.5, 44.1 cm^{-1}; HRMS (ESI-TOF$^+$): *m/z* Calcd for $C_{14}H_{12}O_4N_3$ [(M+H)$^+$] 286.0822; found, 286.0822.

^{1}H NMR and ^{13}C NMR Spectra for Bicyclic Pyridones

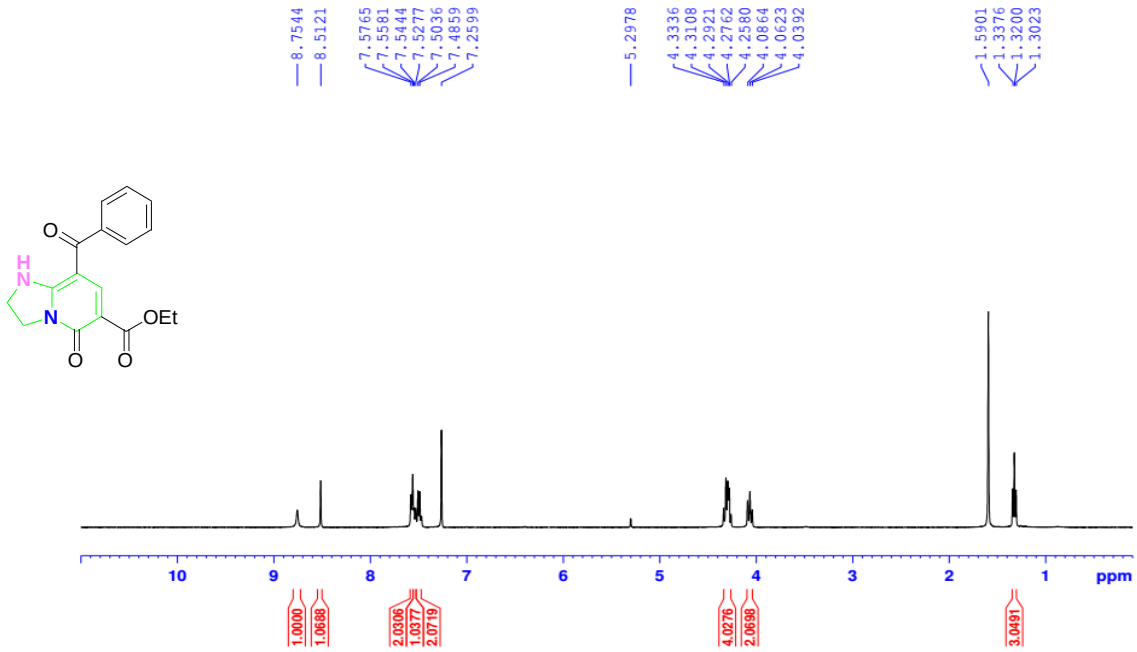


Figure 1. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4aa**

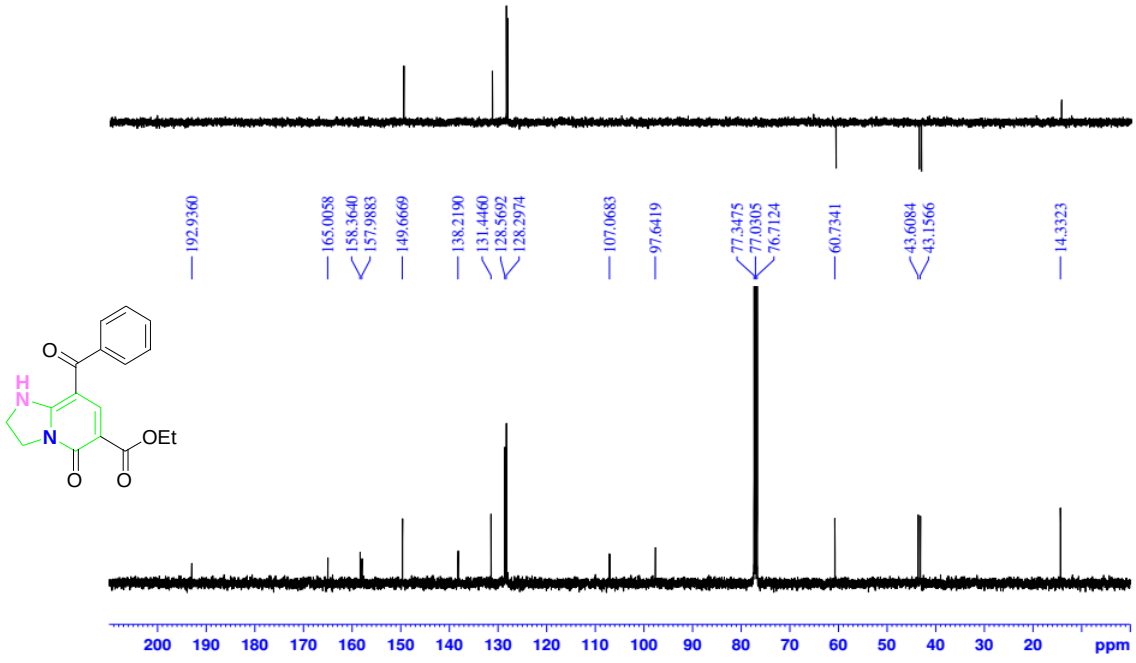


Figure 2. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4aa**

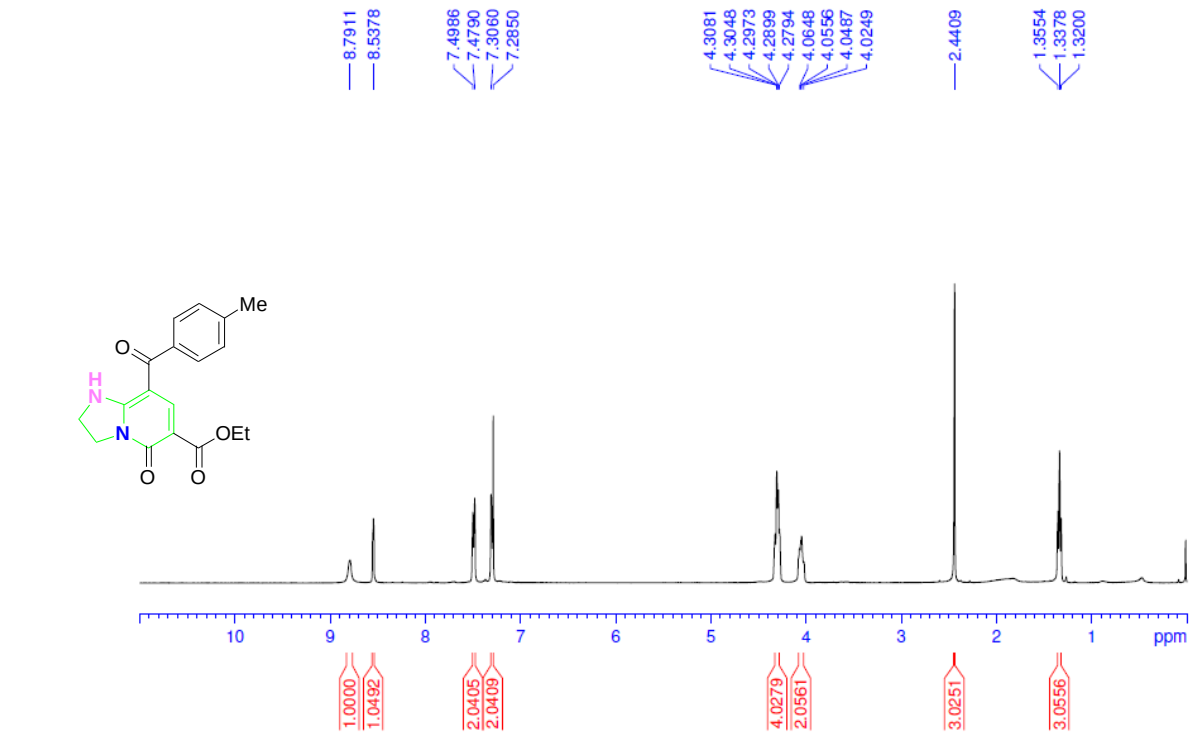


Figure 3. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ba**

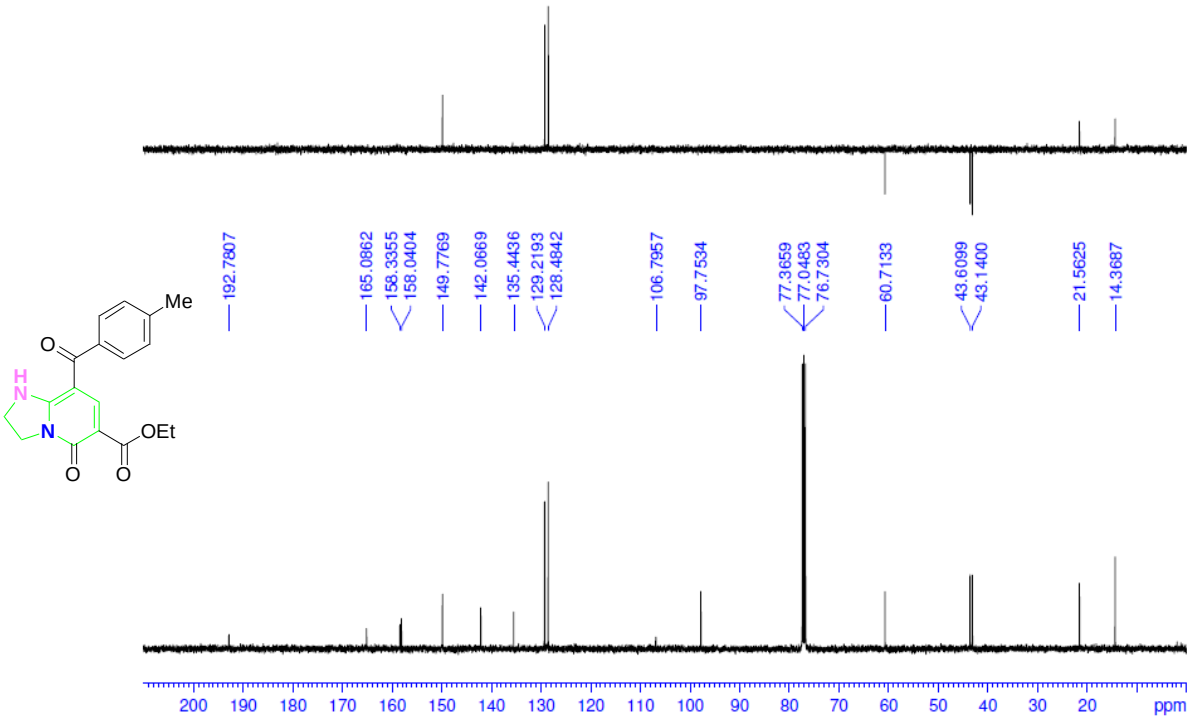


Figure 4. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ba**

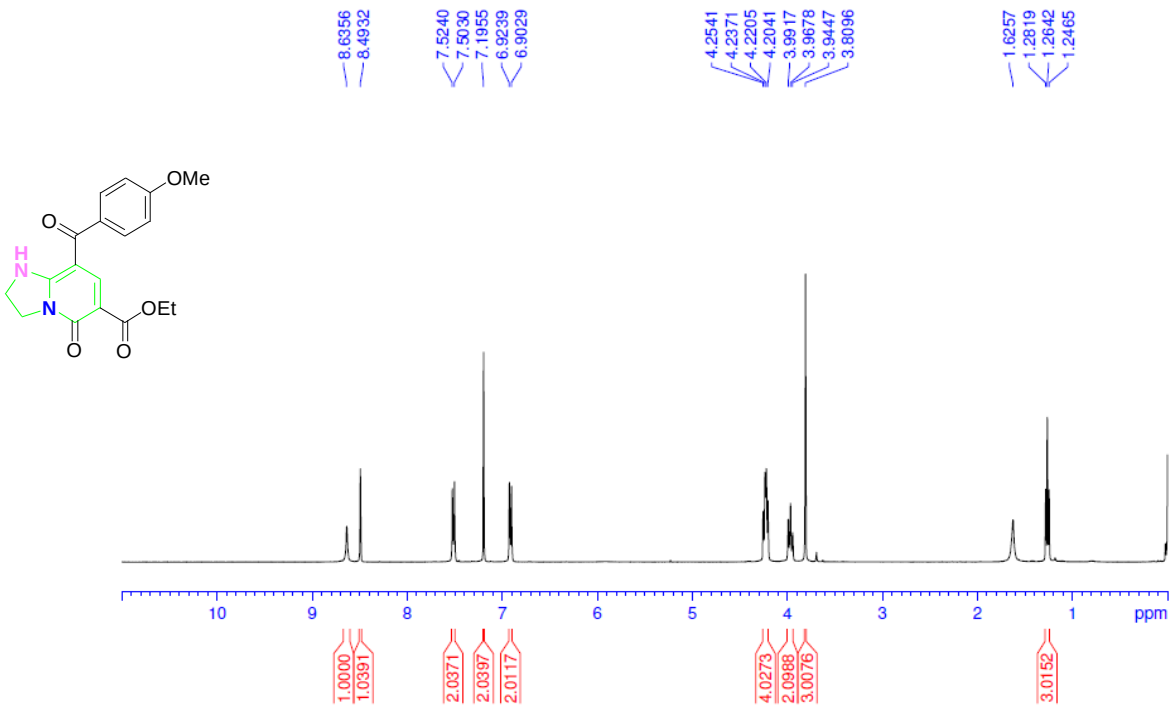


Figure 5. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ca**

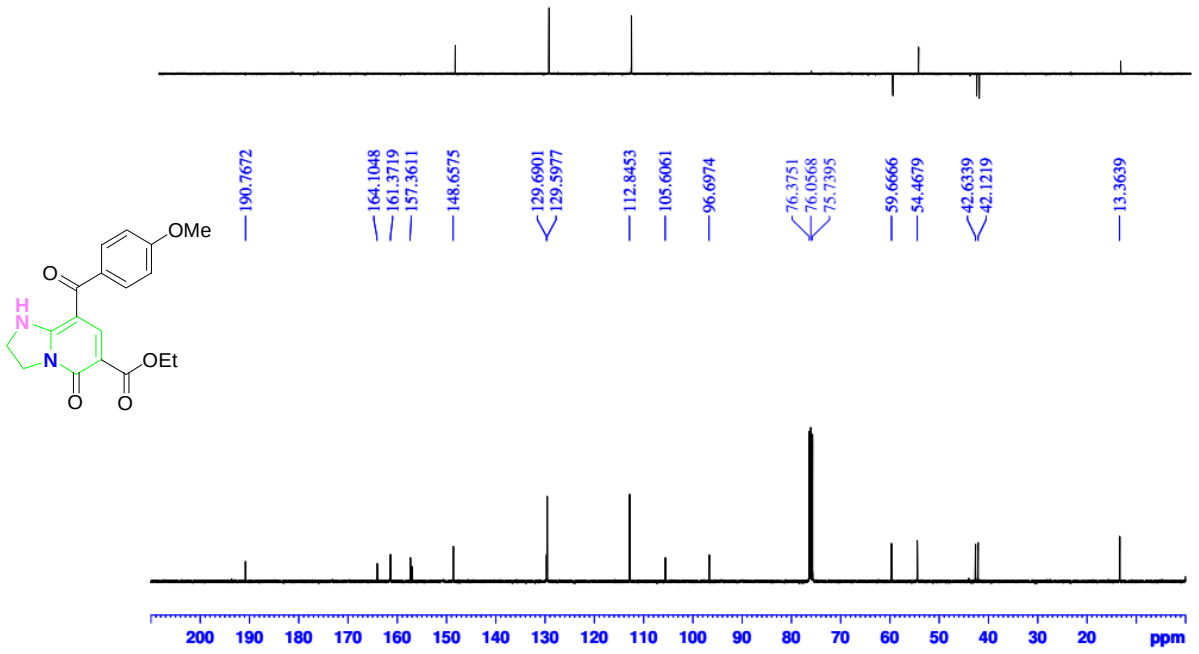


Figure 6. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ca**

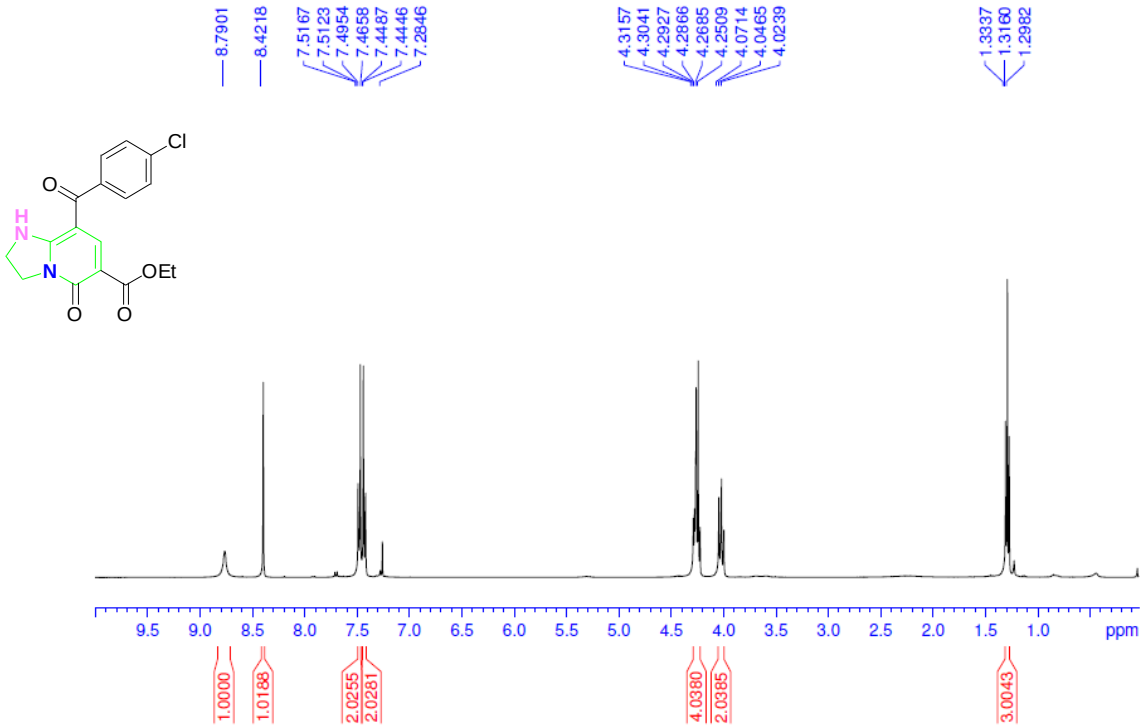


Figure 7. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4da**

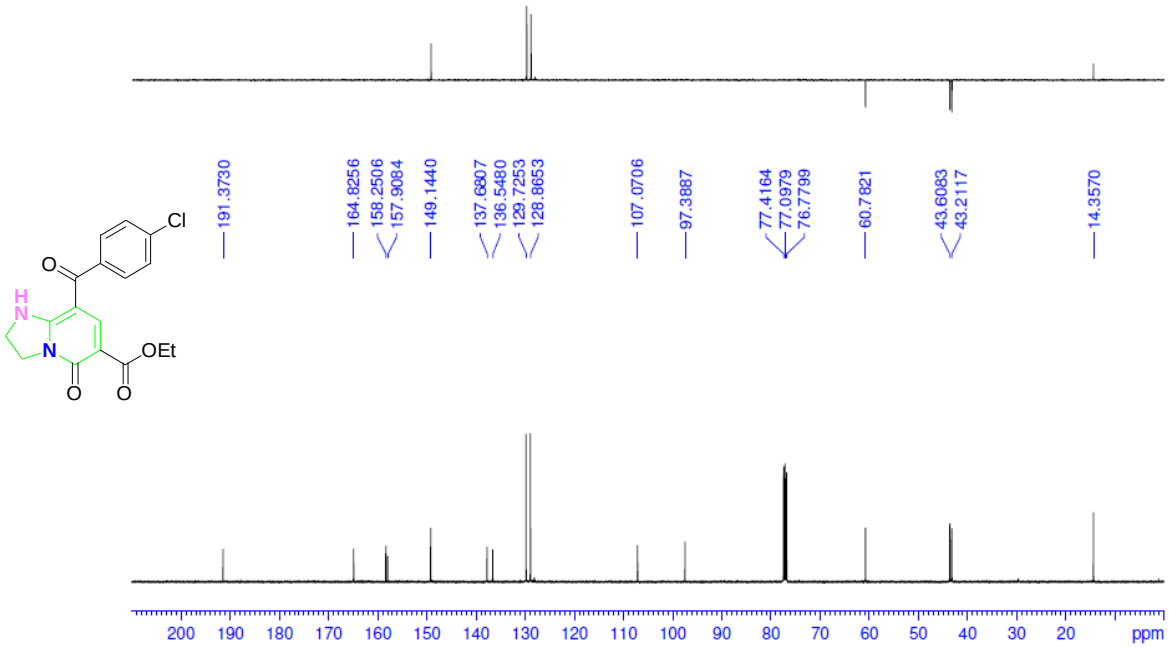


Figure 8. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4da**

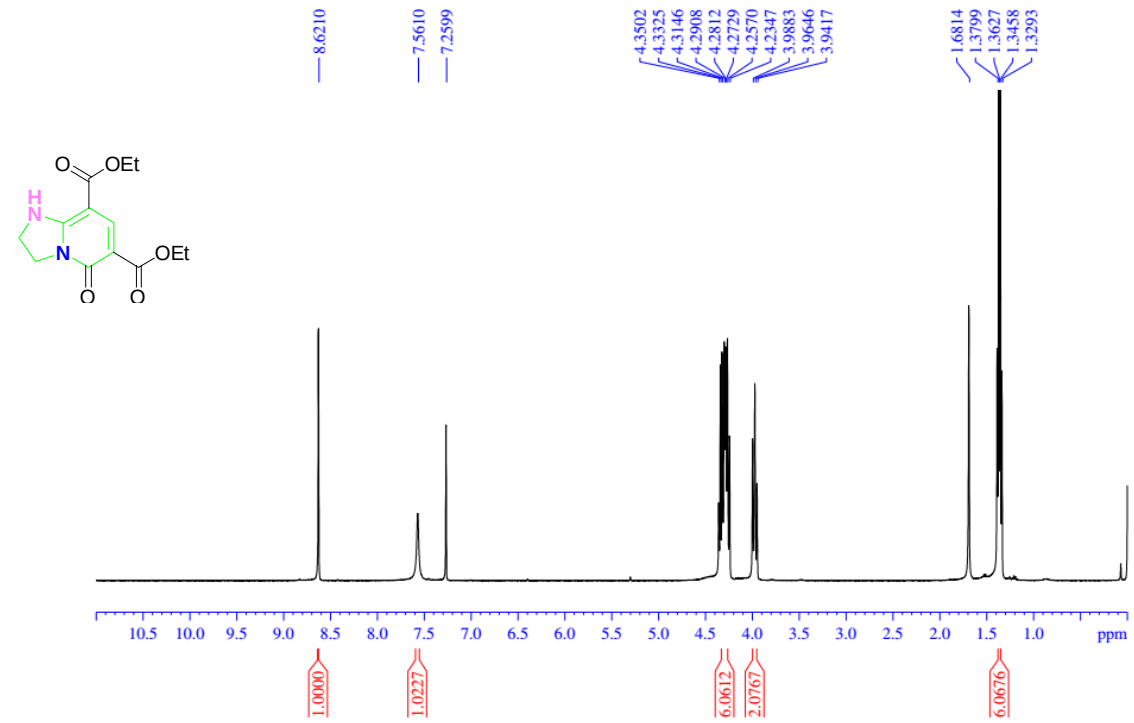


Figure 9. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ea**

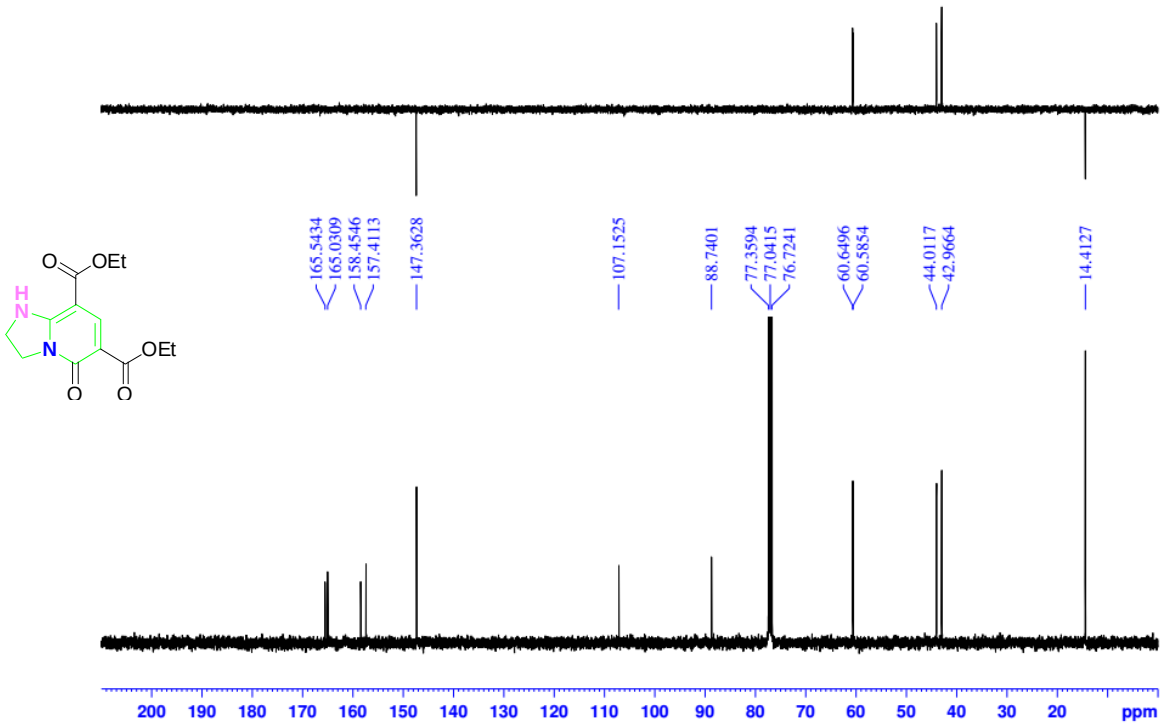


Figure 10. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ea**

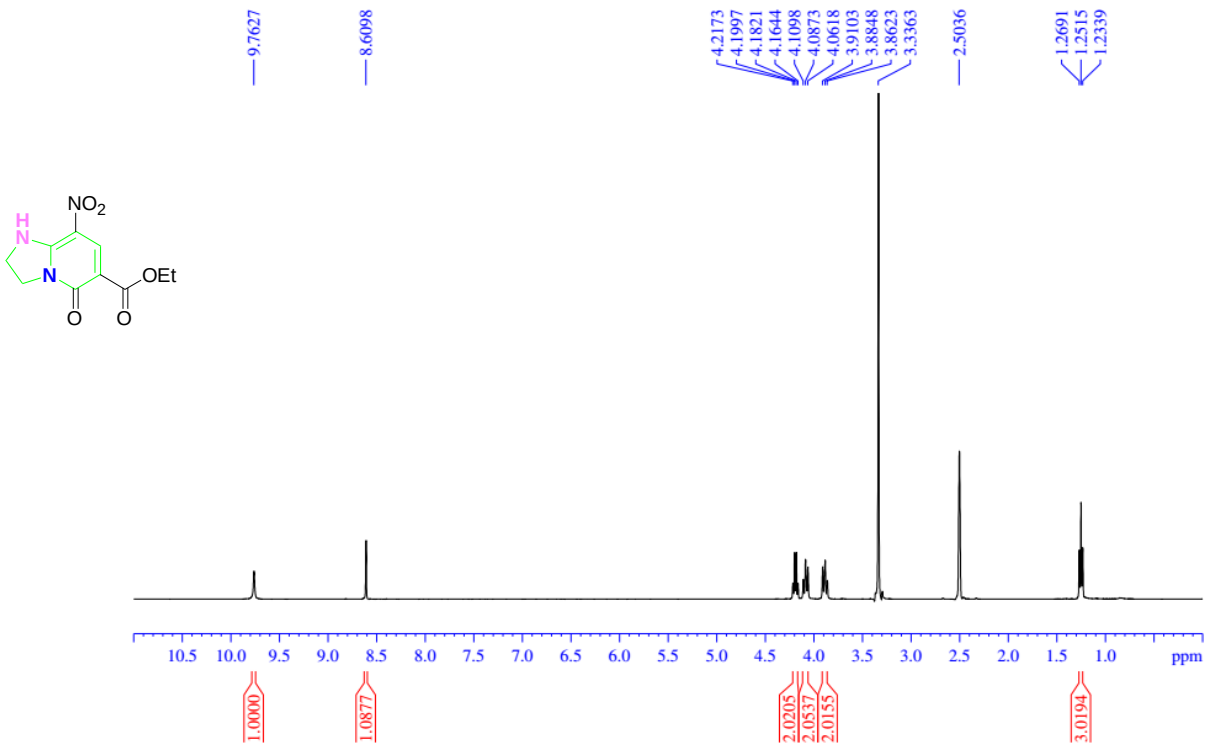


Figure 11. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4fa**

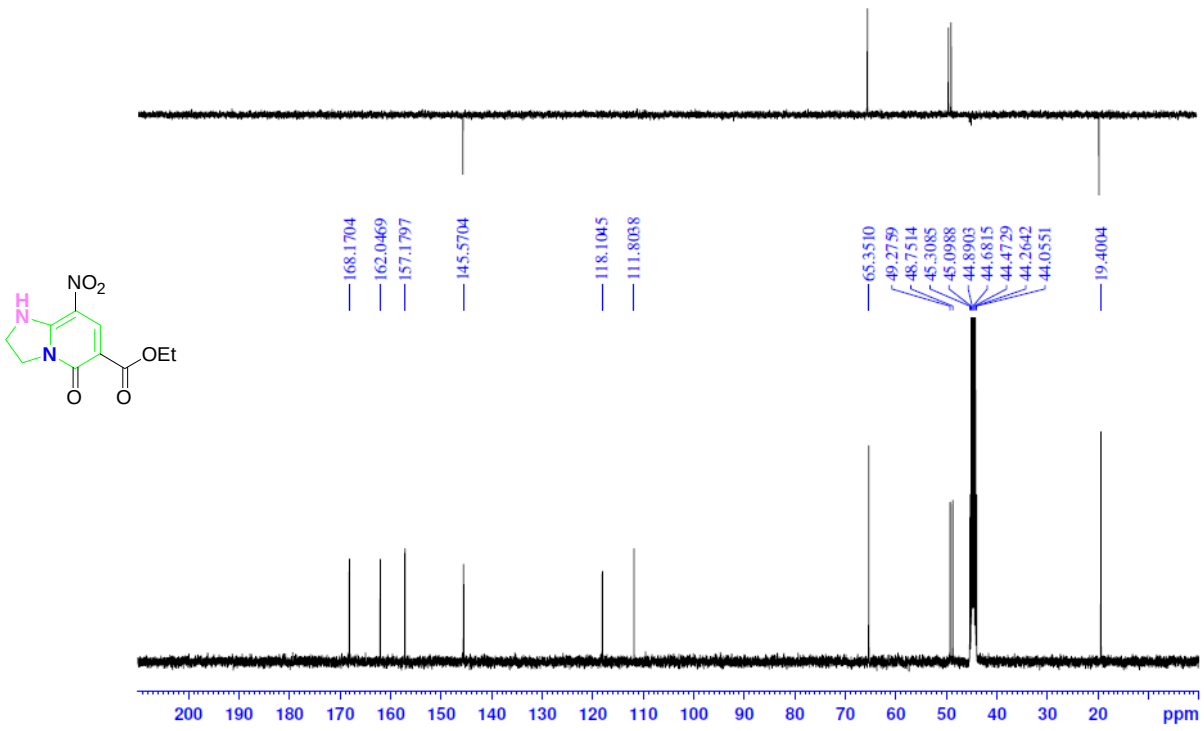


Figure 12. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4fa**

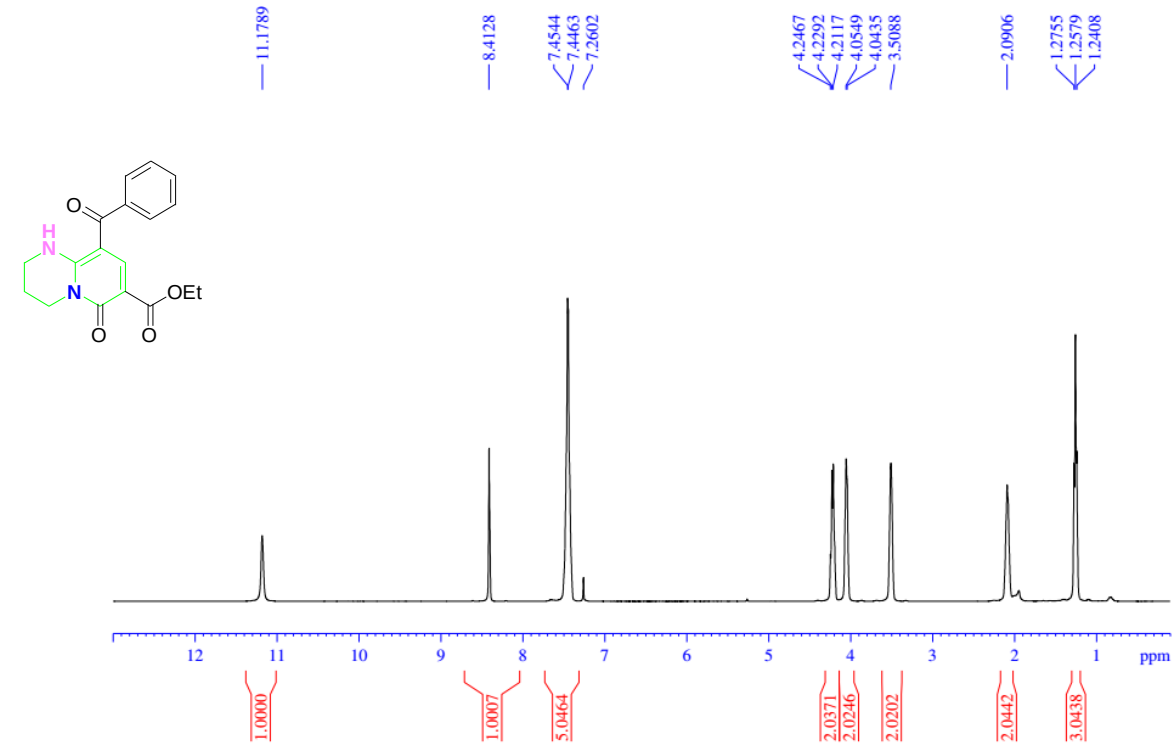


Figure 13. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ga**

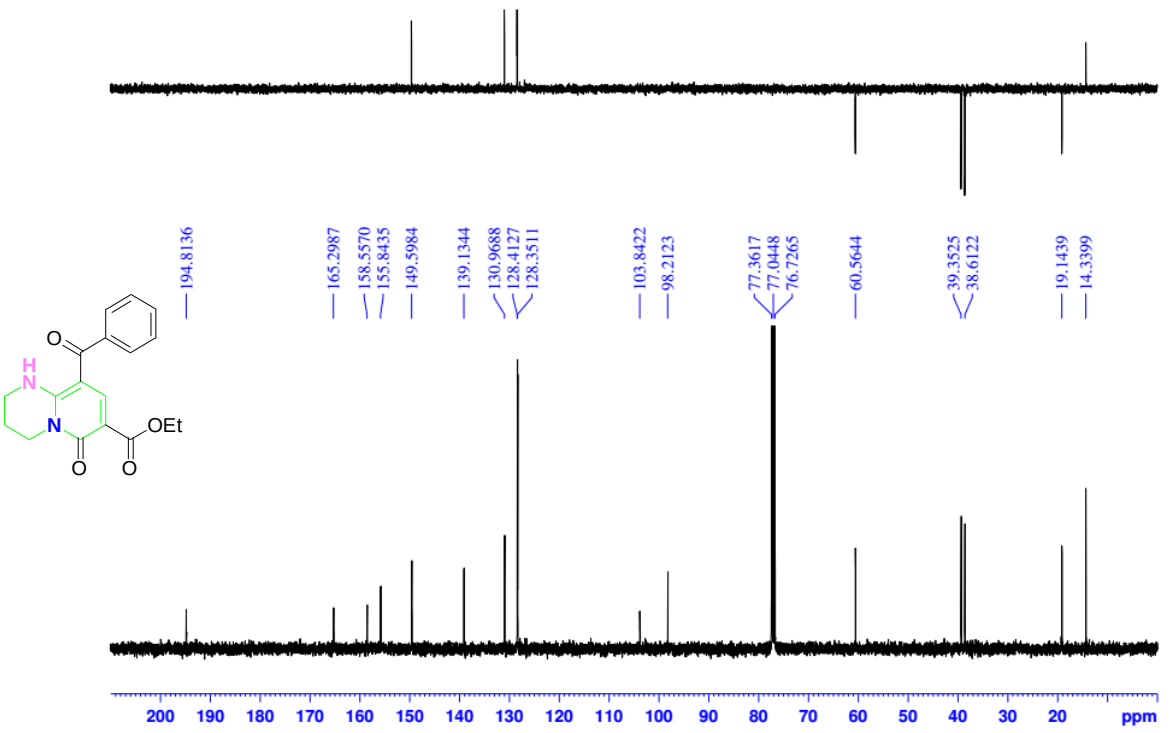


Figure 14. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ga**

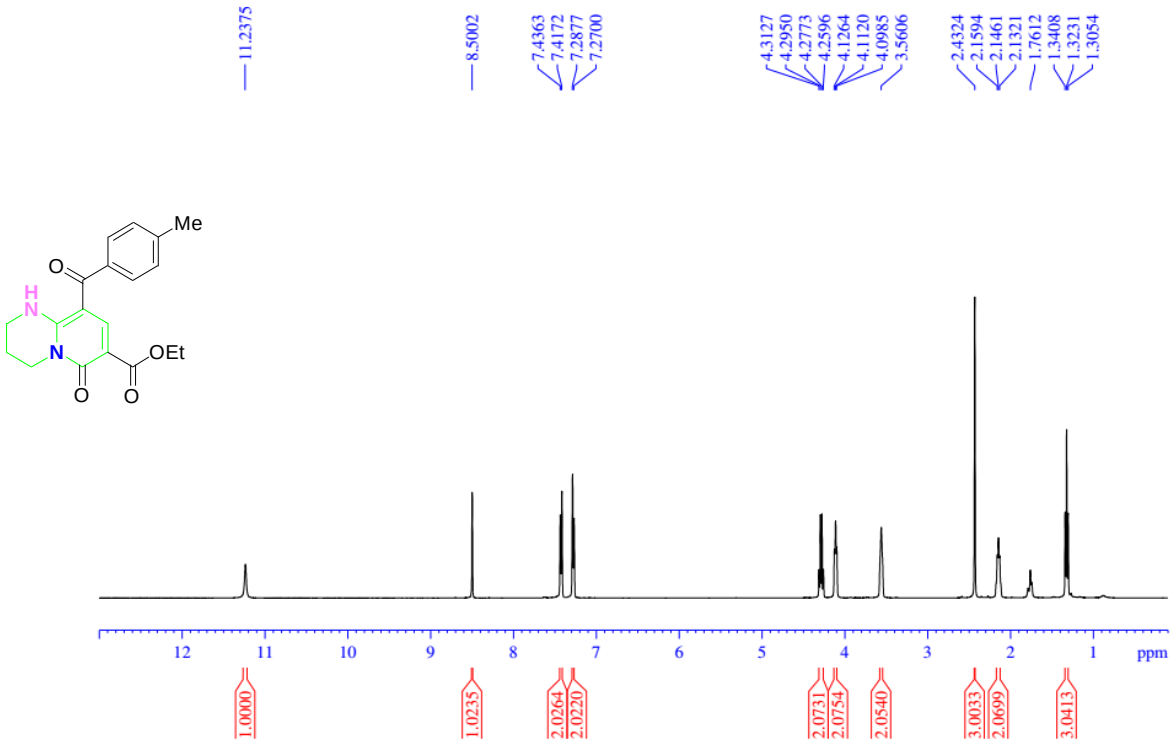


Figure 15. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ha**

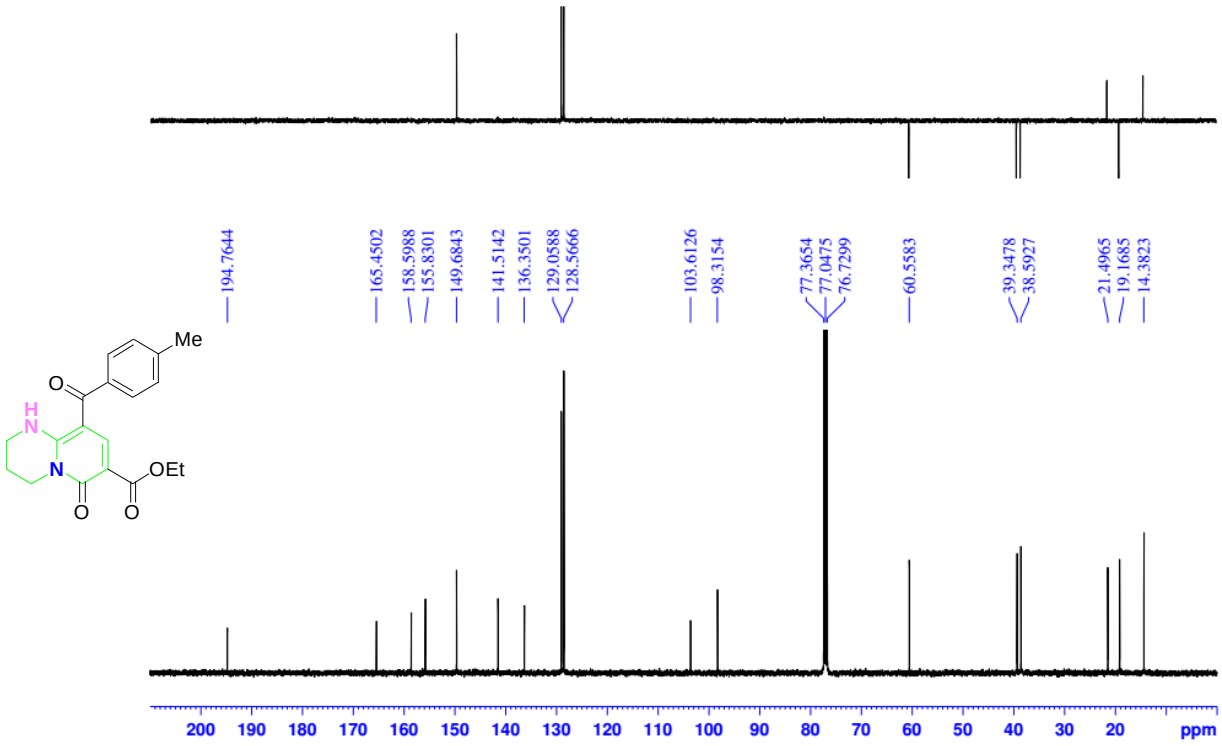


Figure 16. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ha**

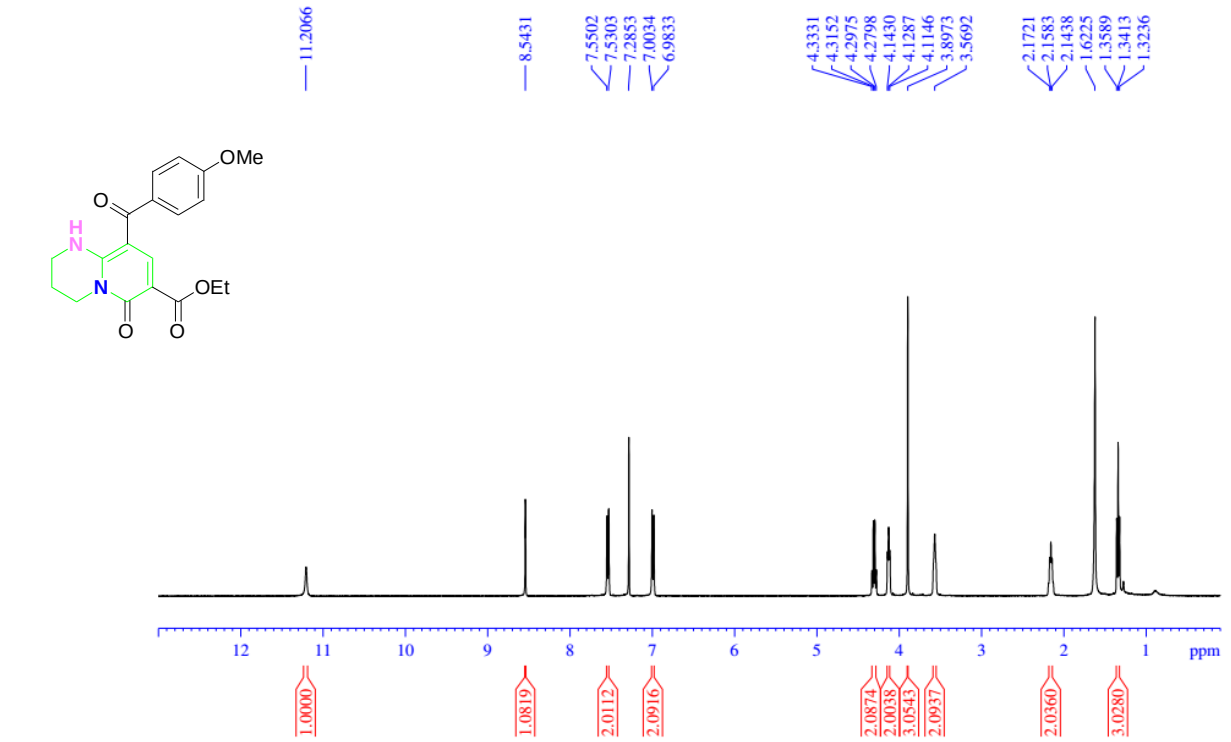


Figure 17. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ia**

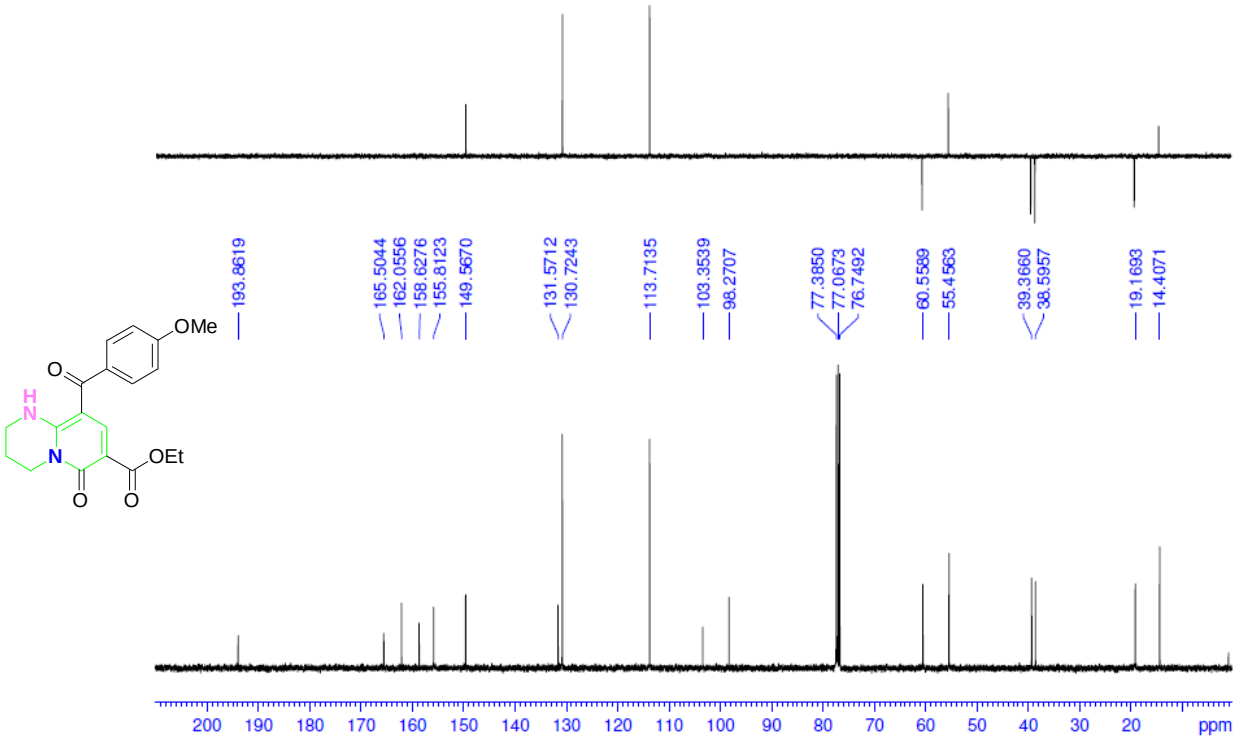


Figure 18. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ia**

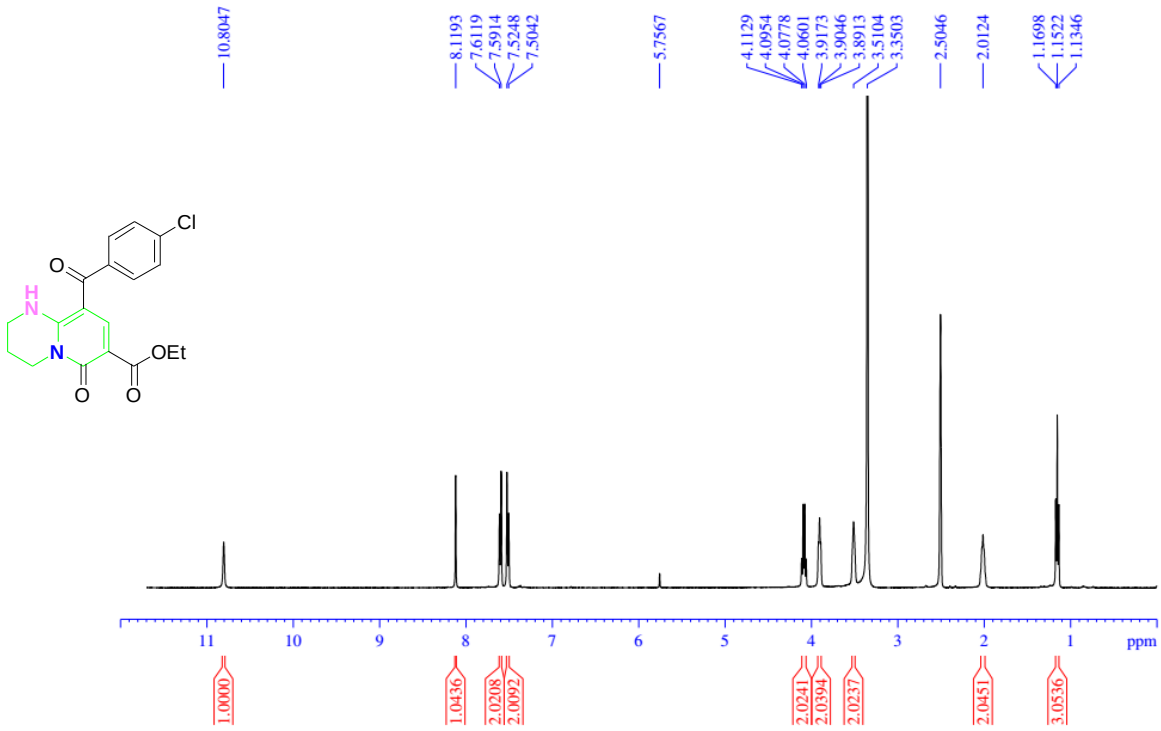


Figure 19. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4ja**

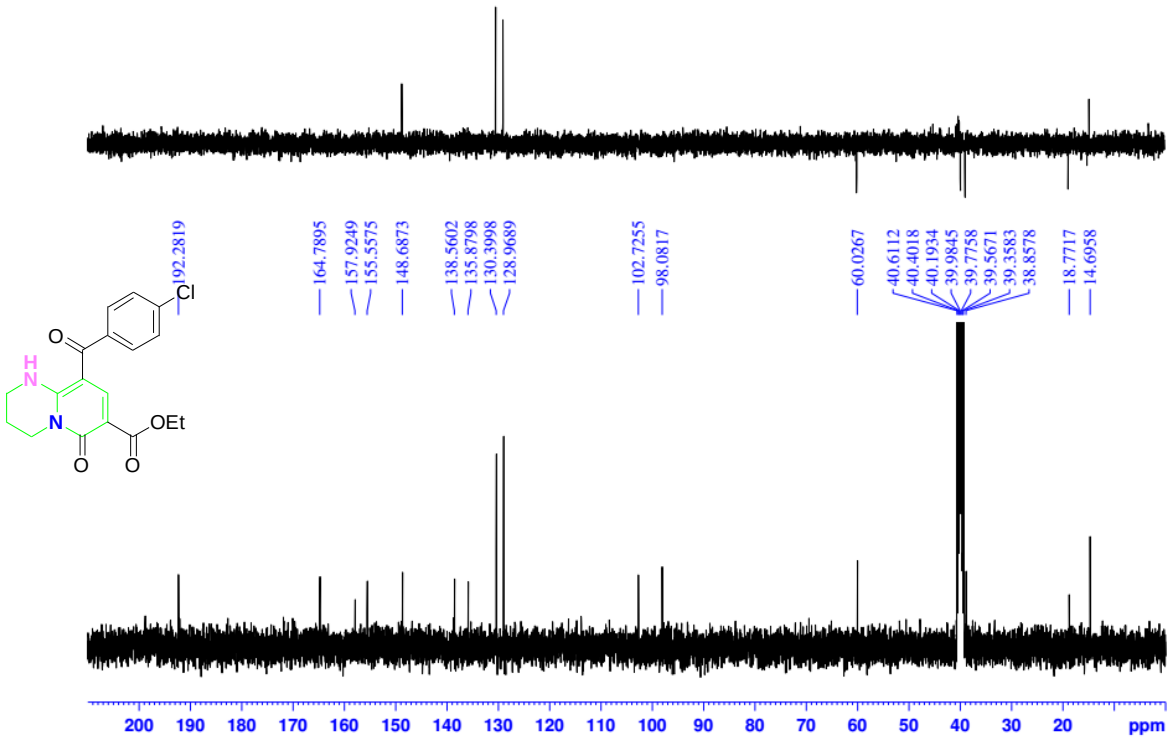


Figure 20. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4ja**

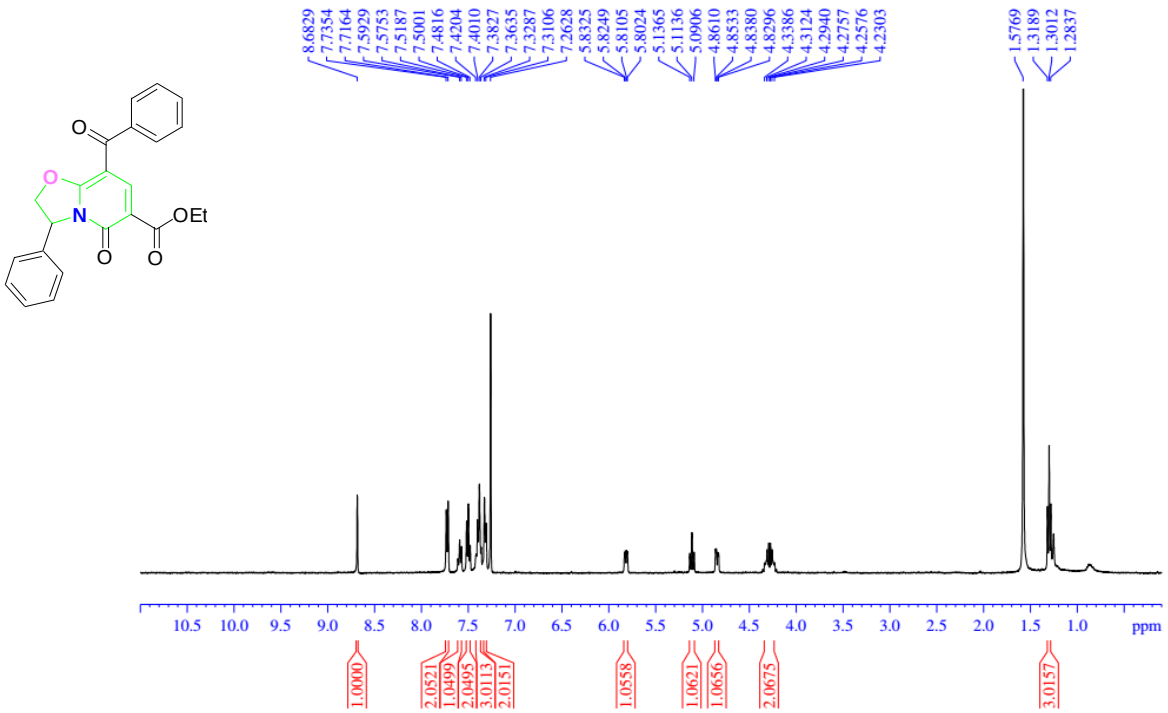


Figure 21. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ka**

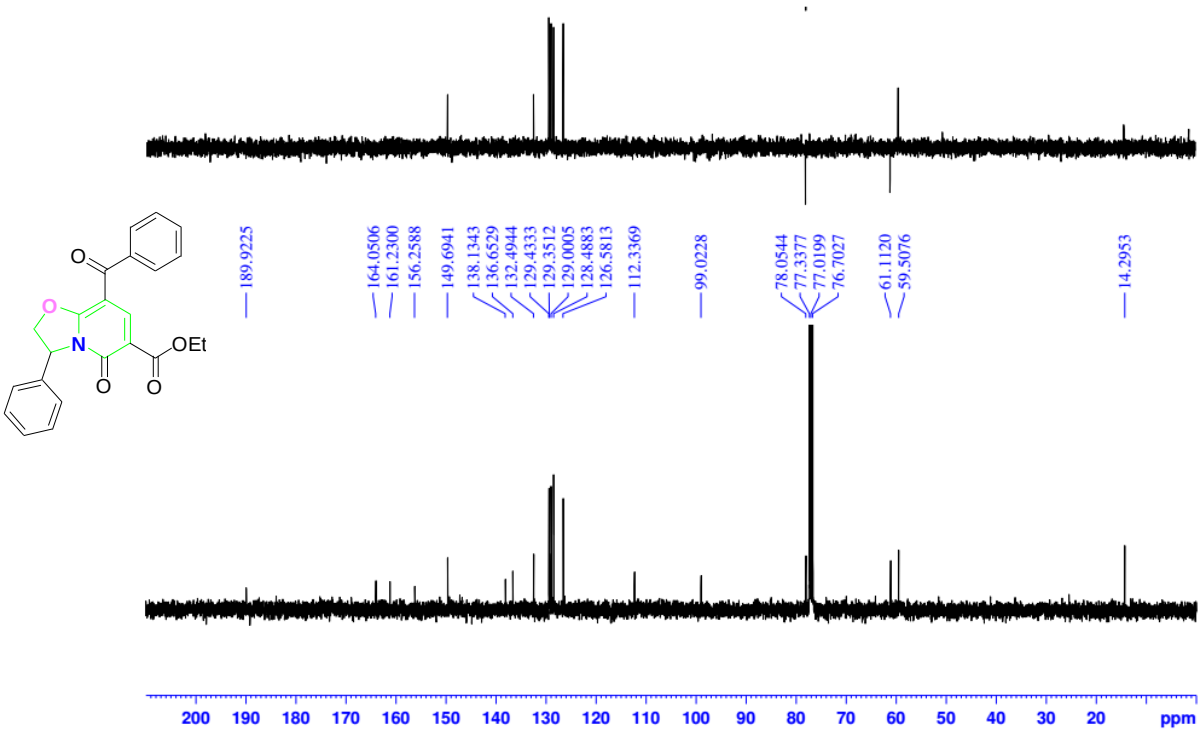


Figure 22. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ka**

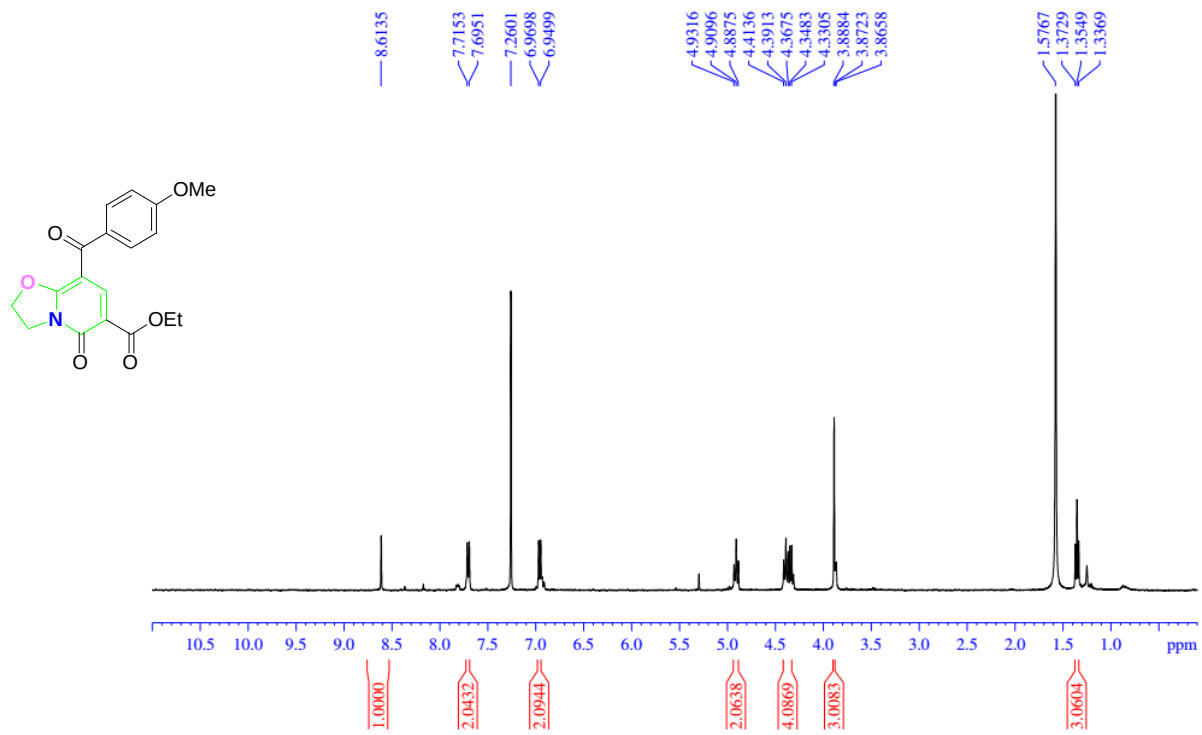


Figure 23. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4la**

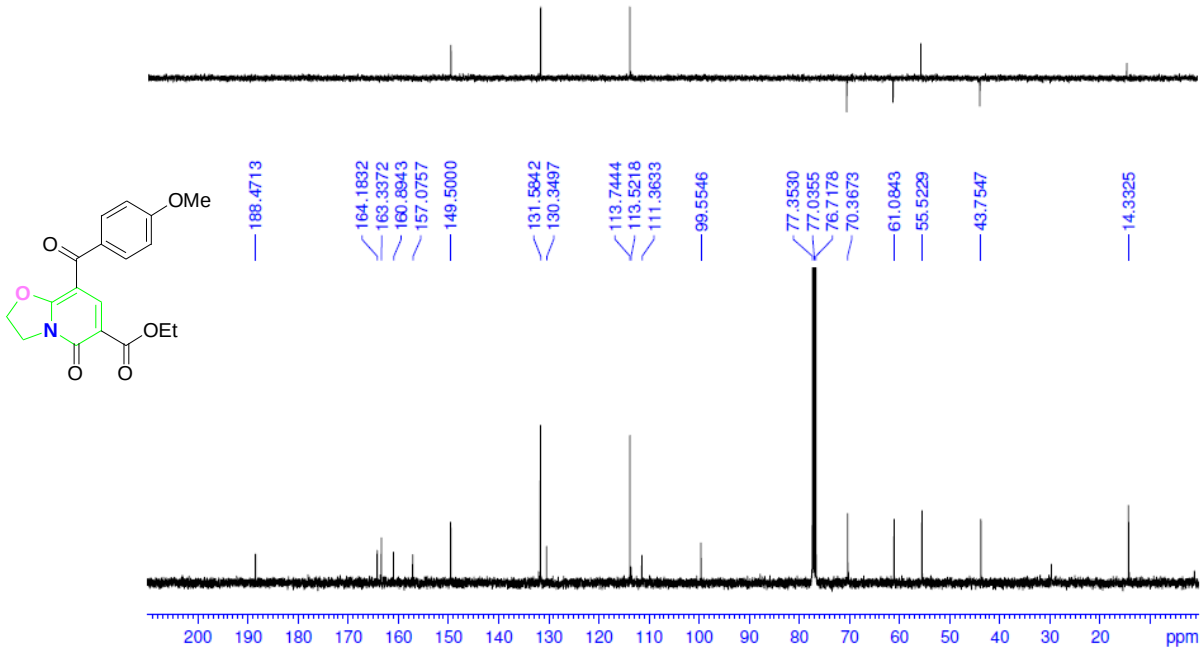


Figure 24. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4la**

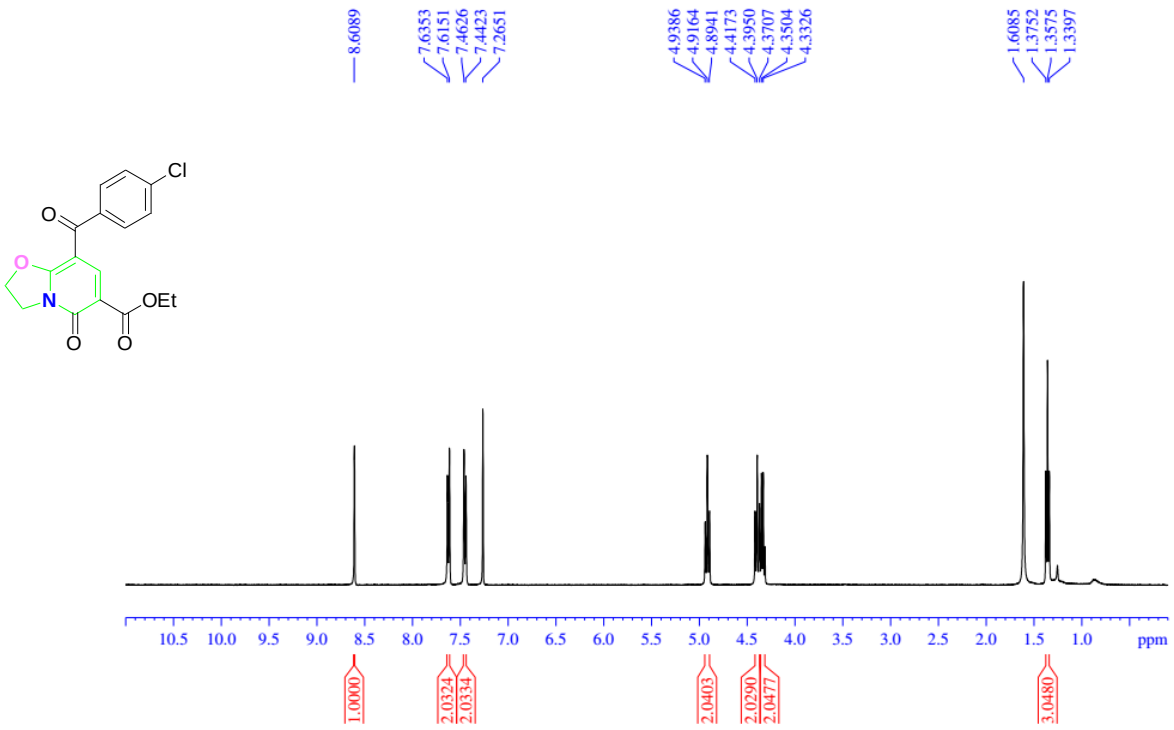


Figure 25. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ma**

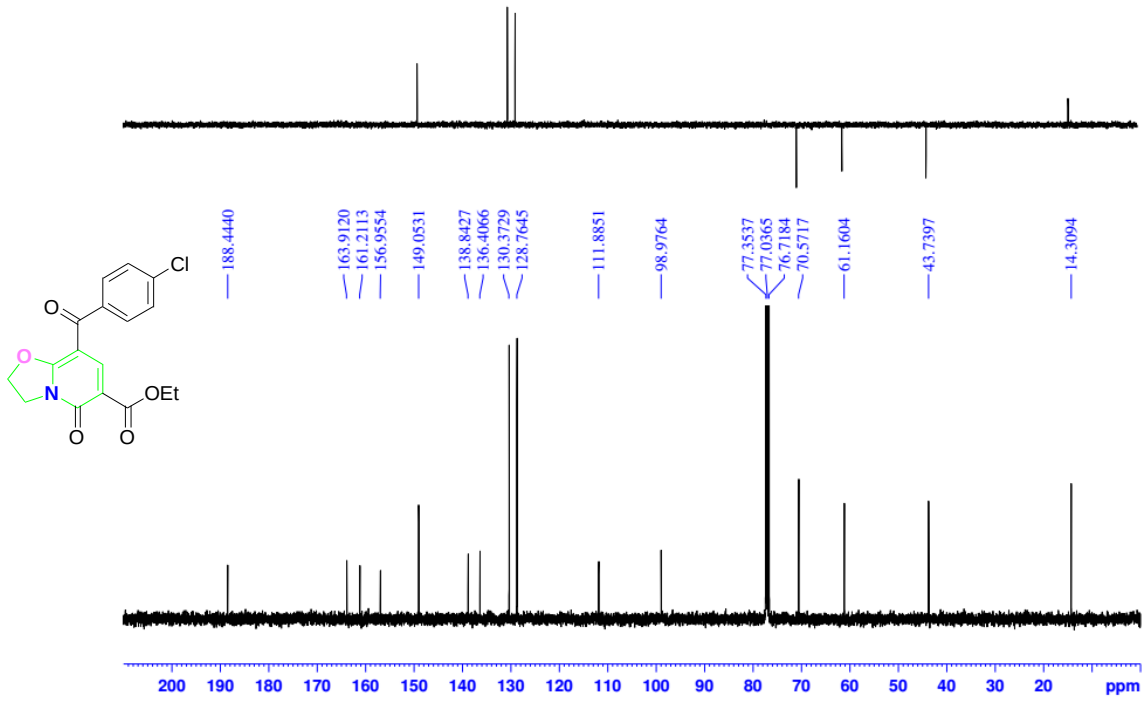


Figure 26. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ma**

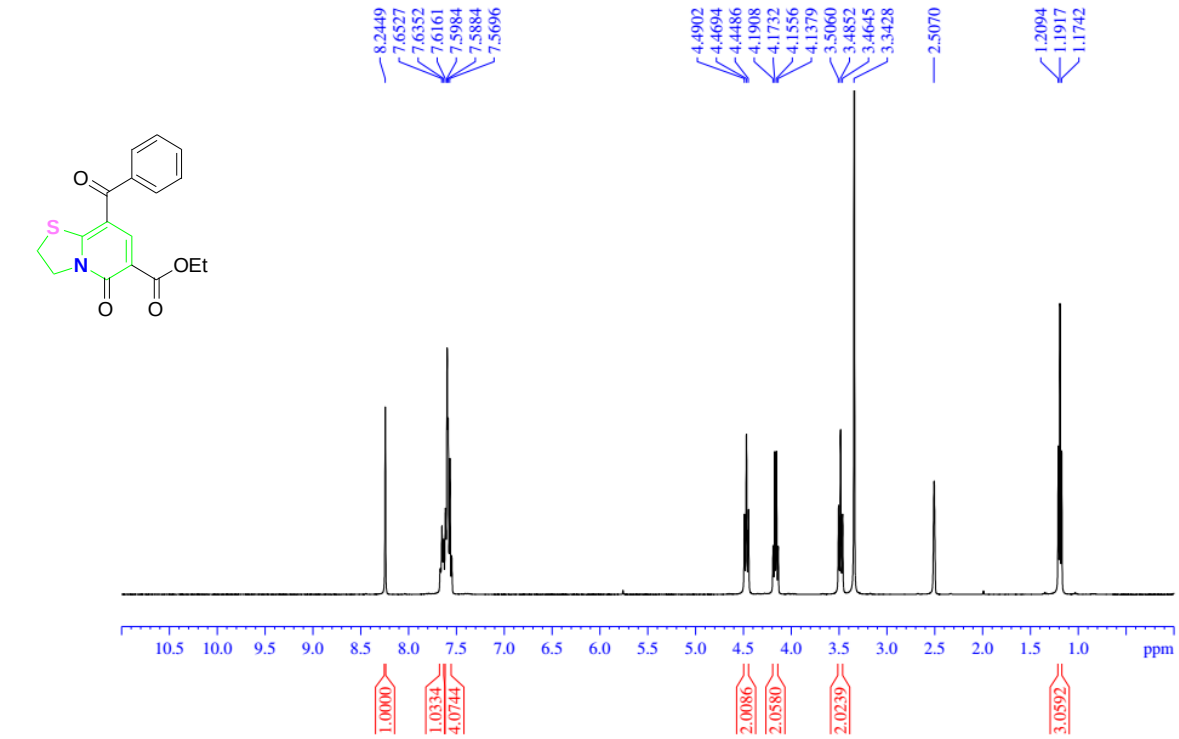


Figure 27. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4na**

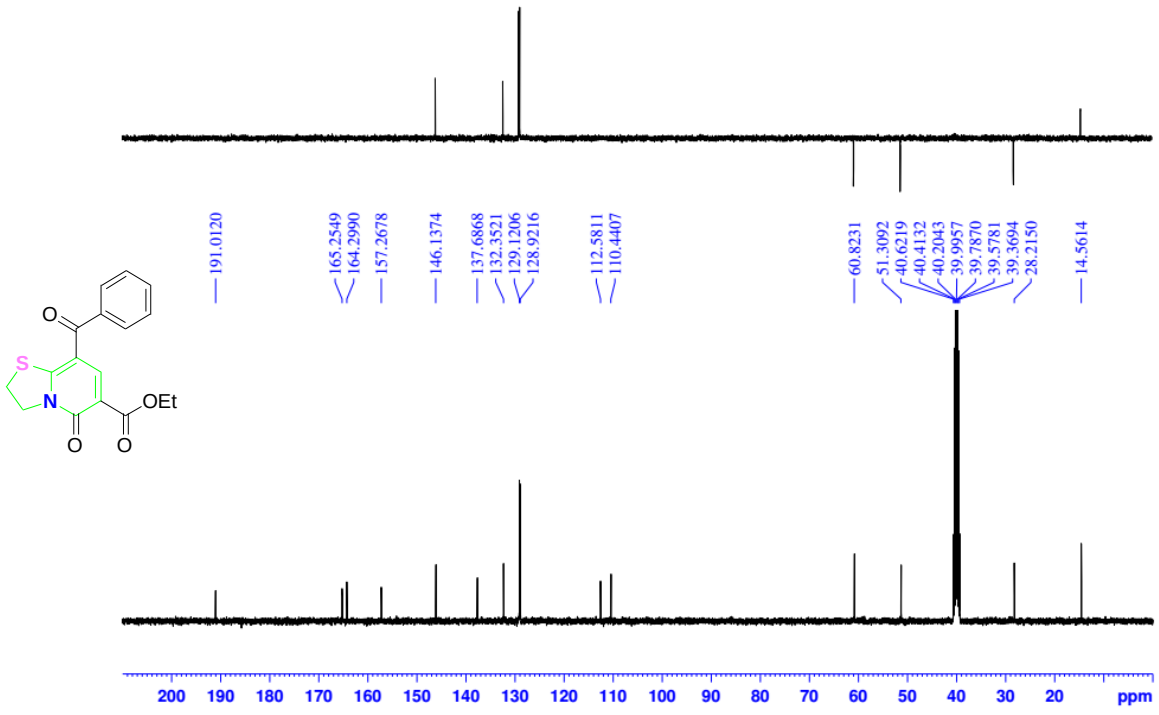


Figure 28. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4na**

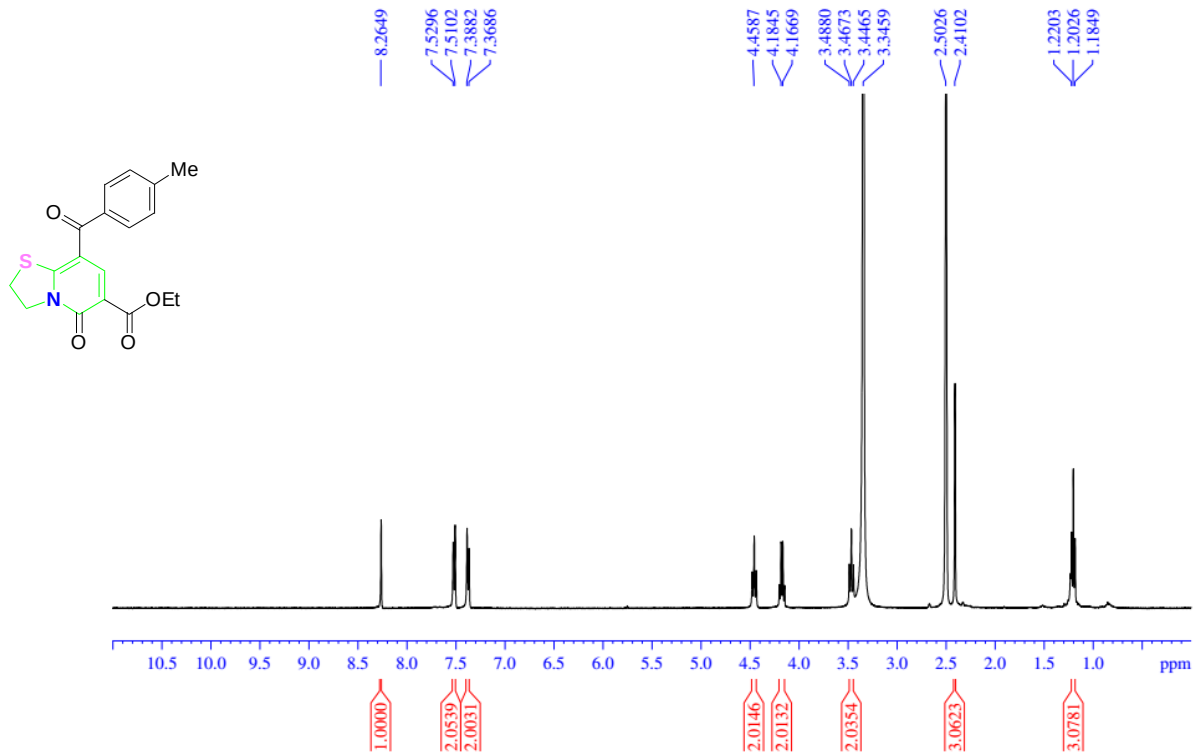


Figure 29. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4oa**

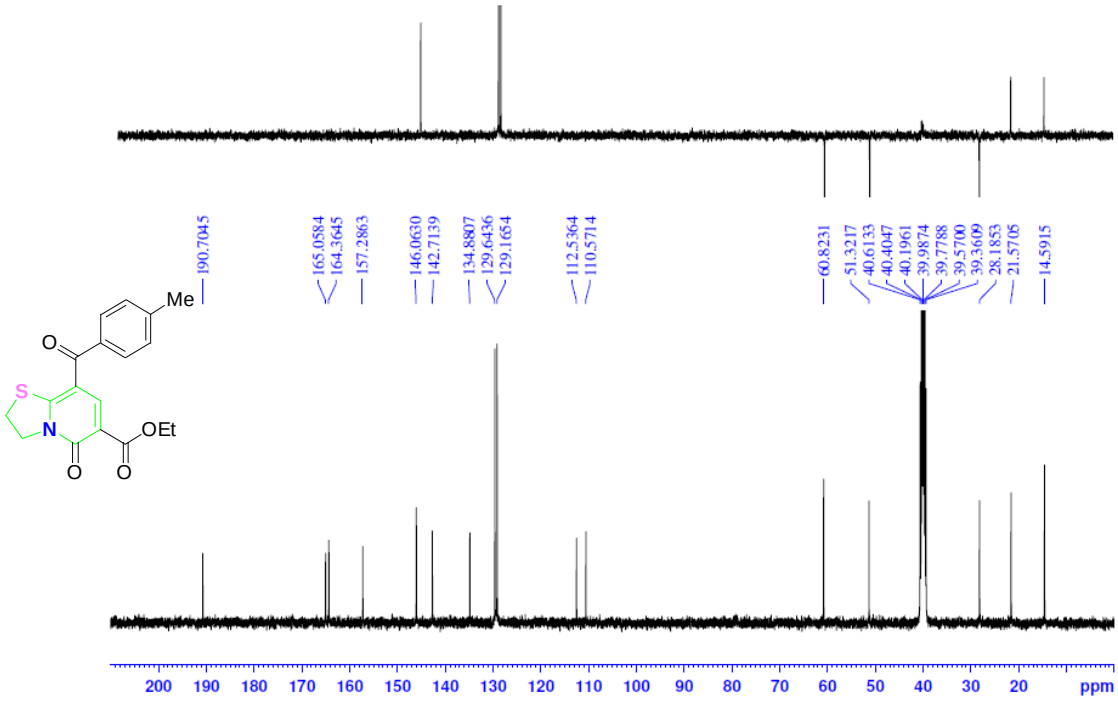


Figure 30. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4oa**

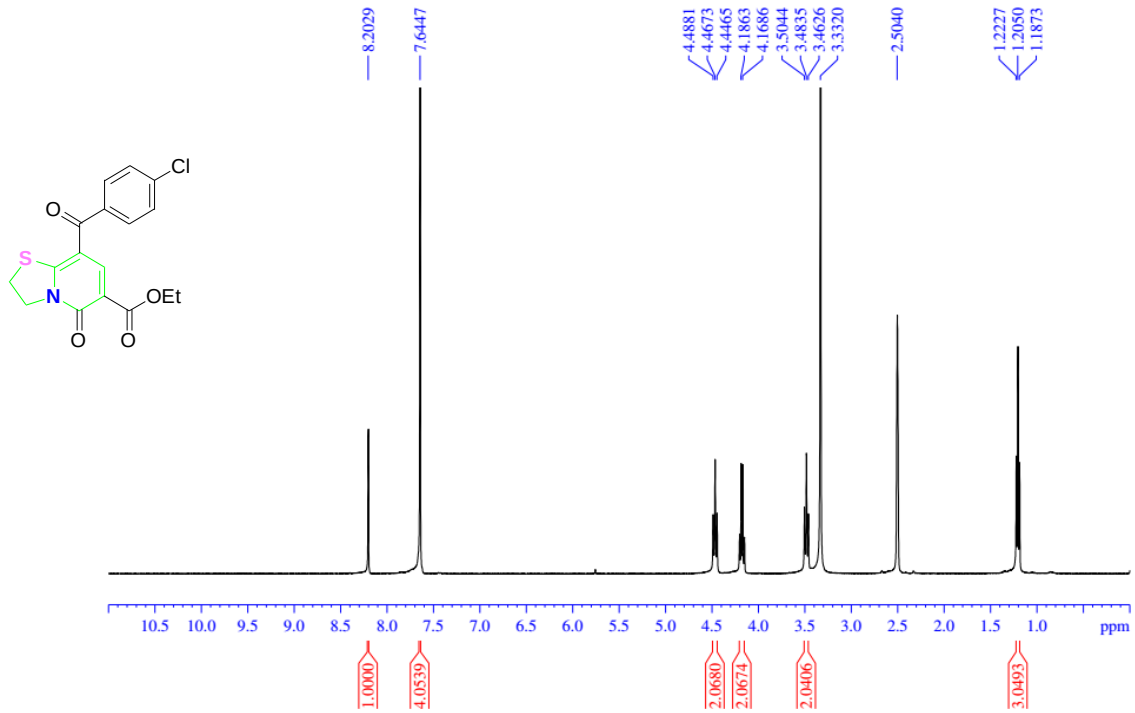


Figure 31. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4pa**

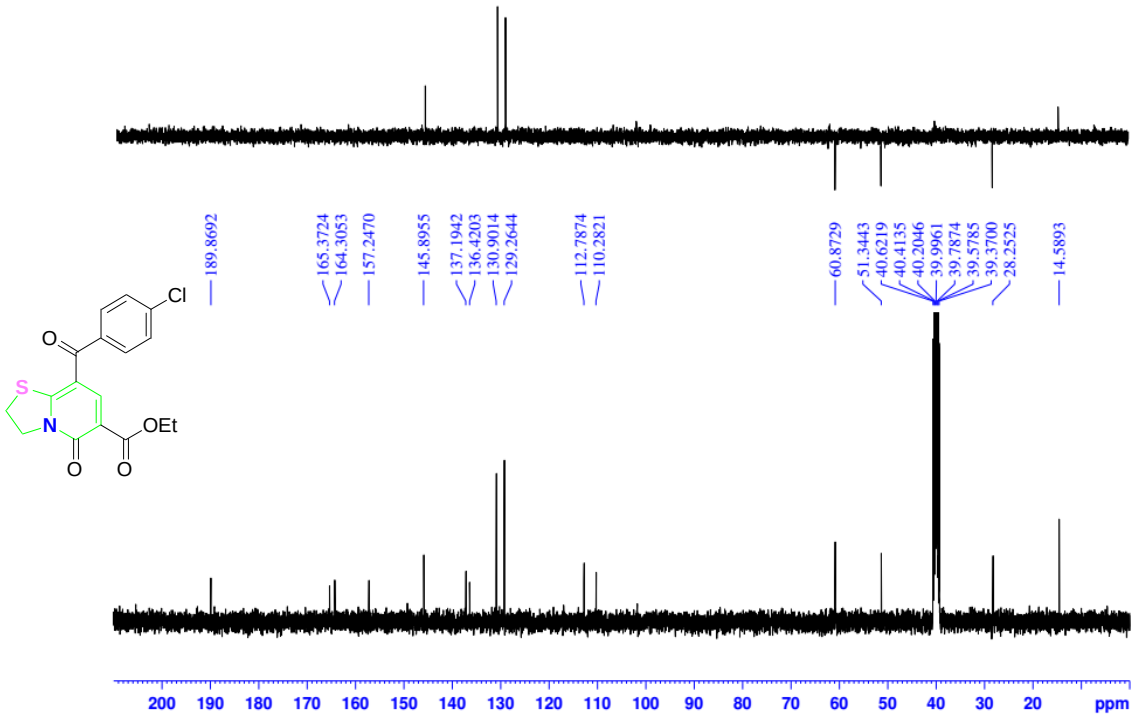


Figure 32. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4pa**

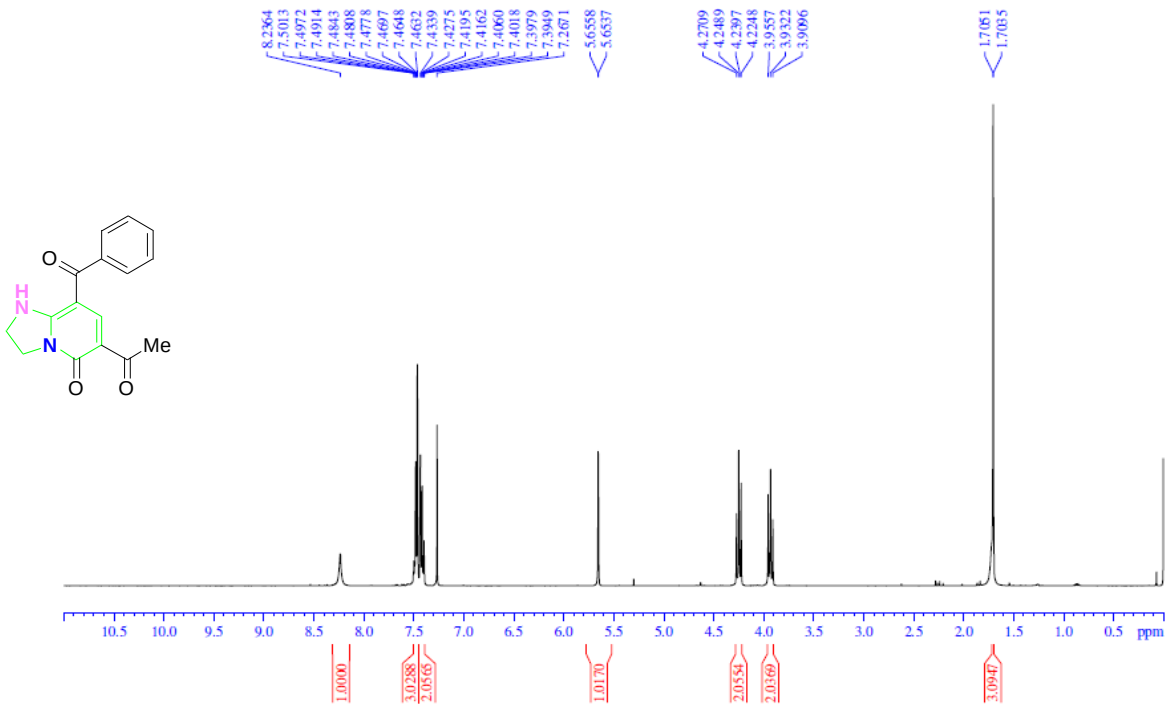


Figure 33. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ab**

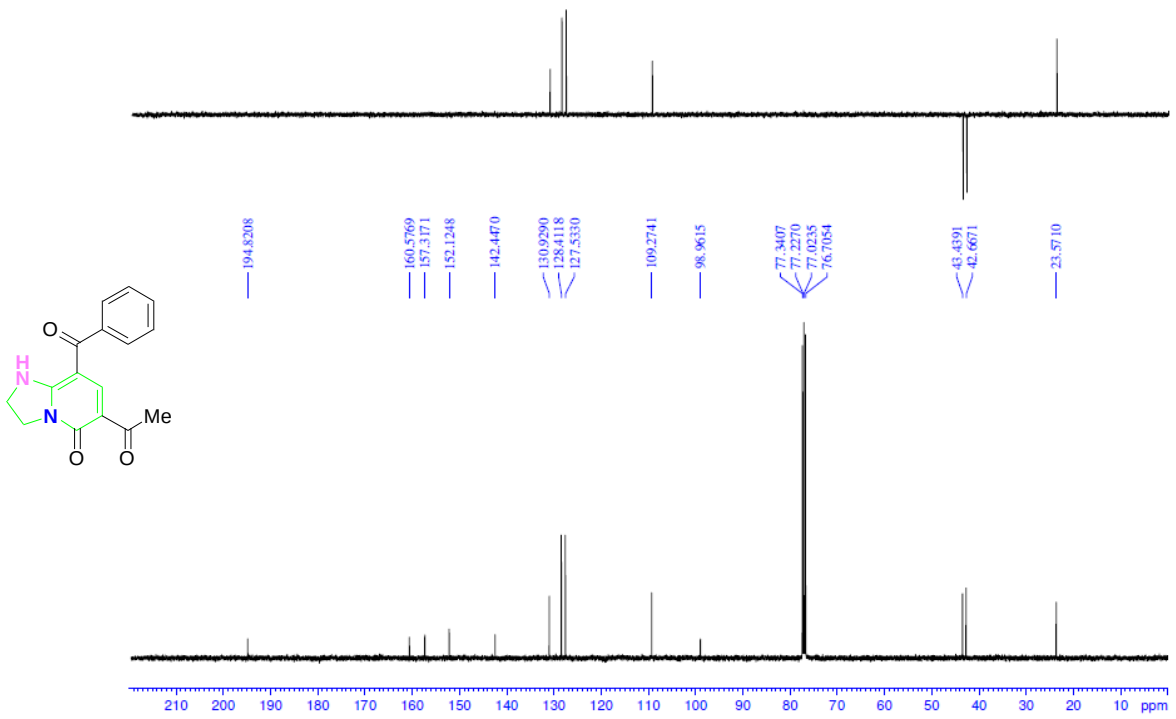


Figure 34. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ab**

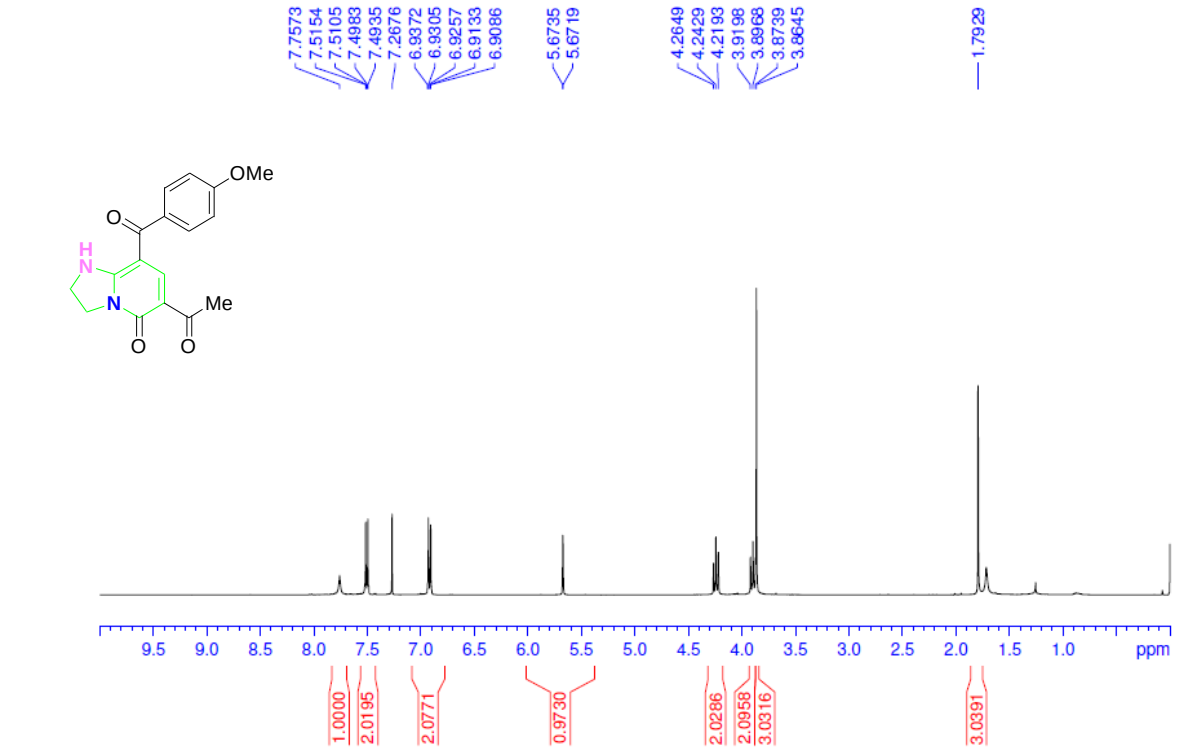


Figure 35. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4cb**

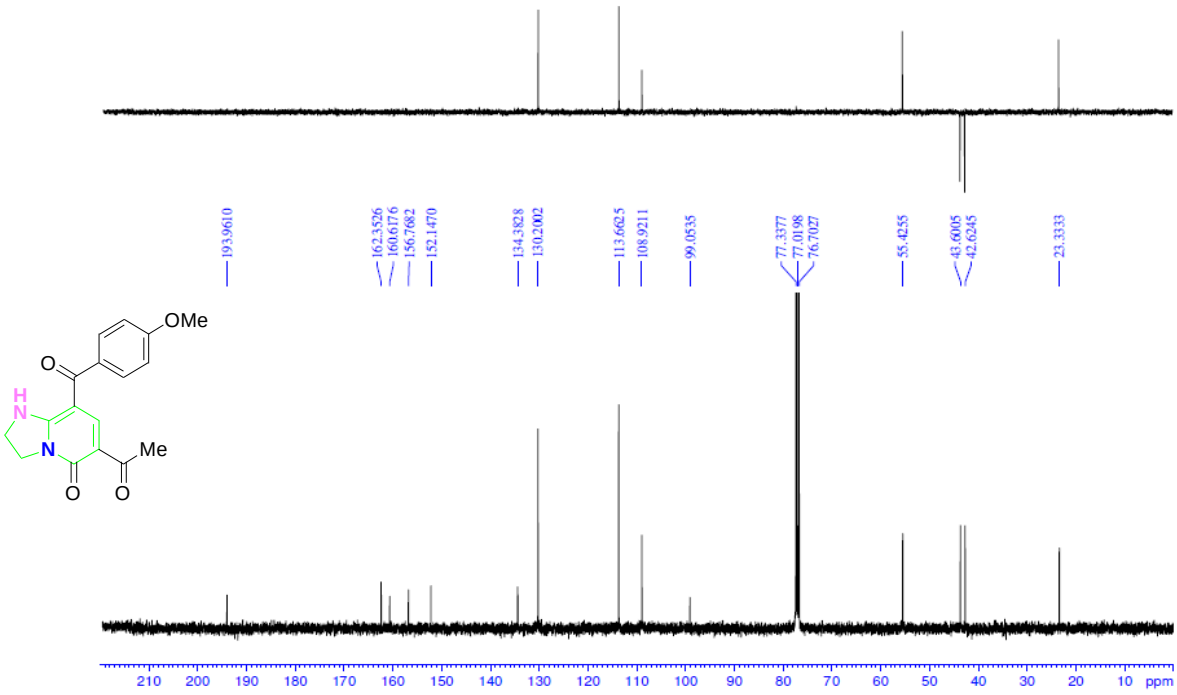


Figure 36. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4cb**

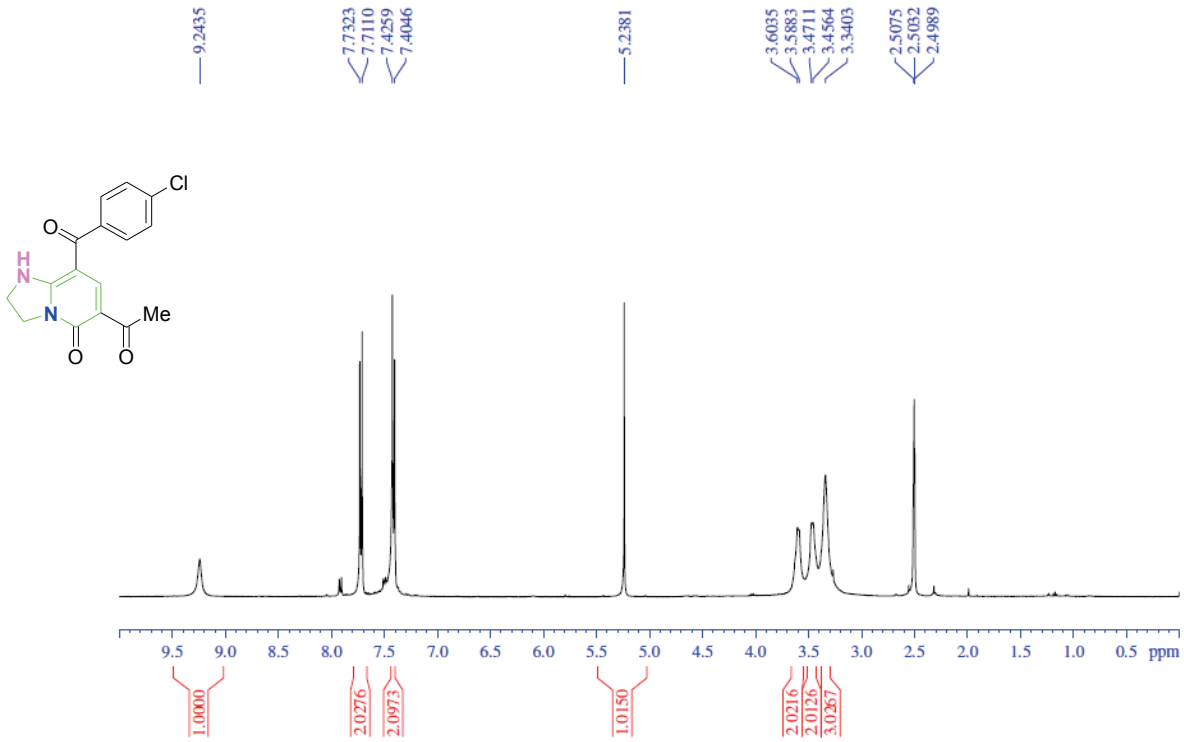


Figure 37. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4db**

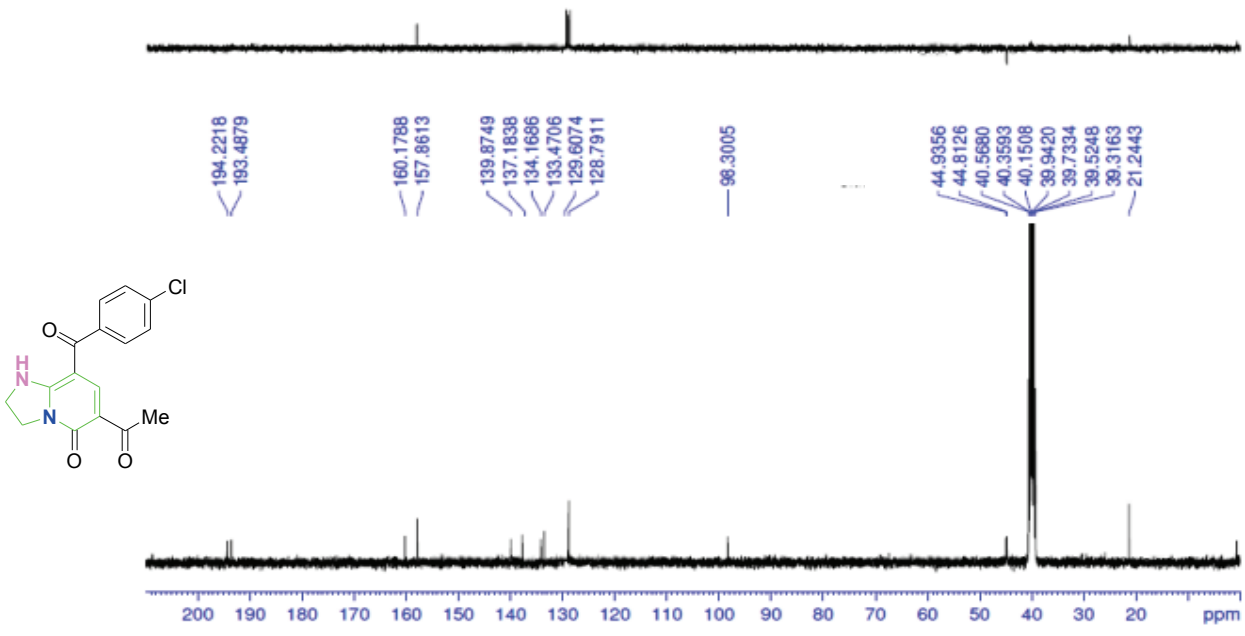


Figure 38. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4db**

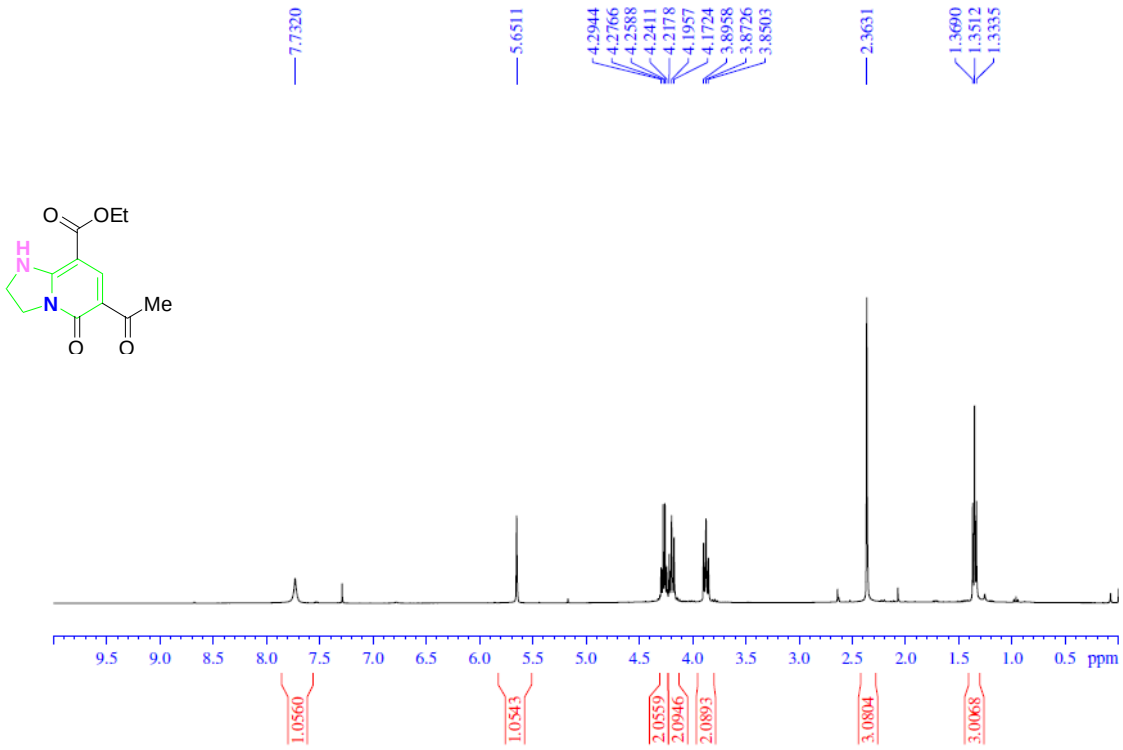


Figure 39. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4eb**

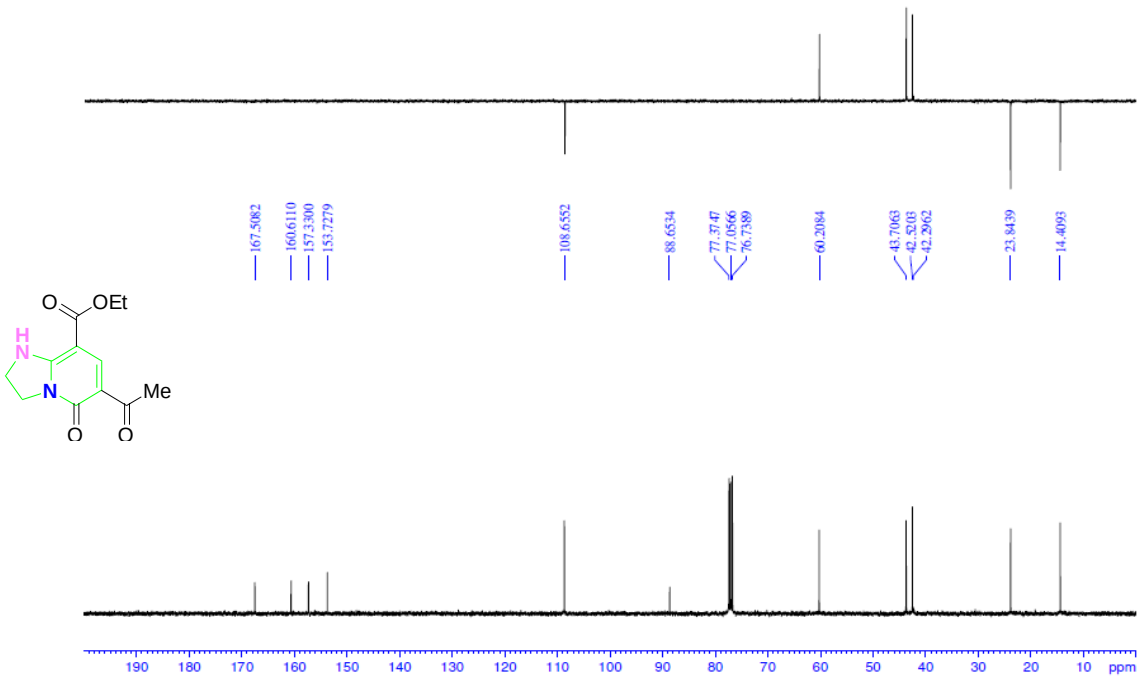


Figure 40. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4eb**

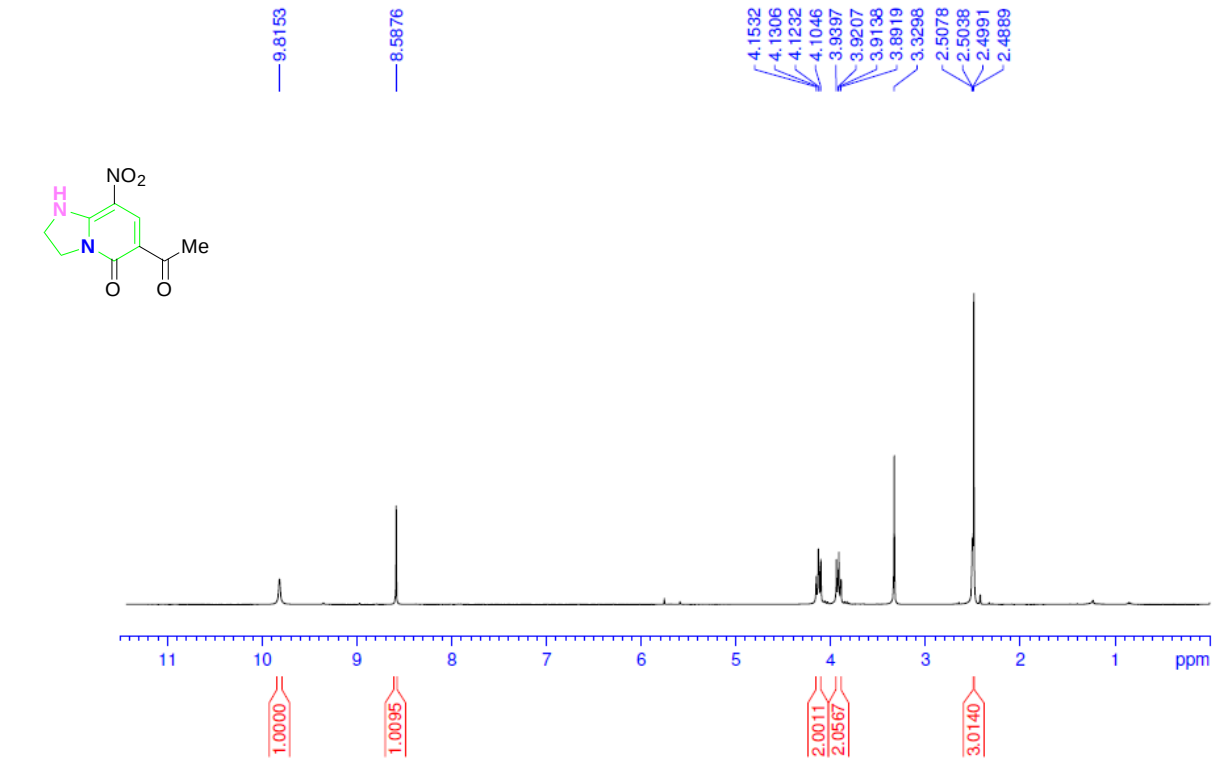


Figure 41. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4fb**

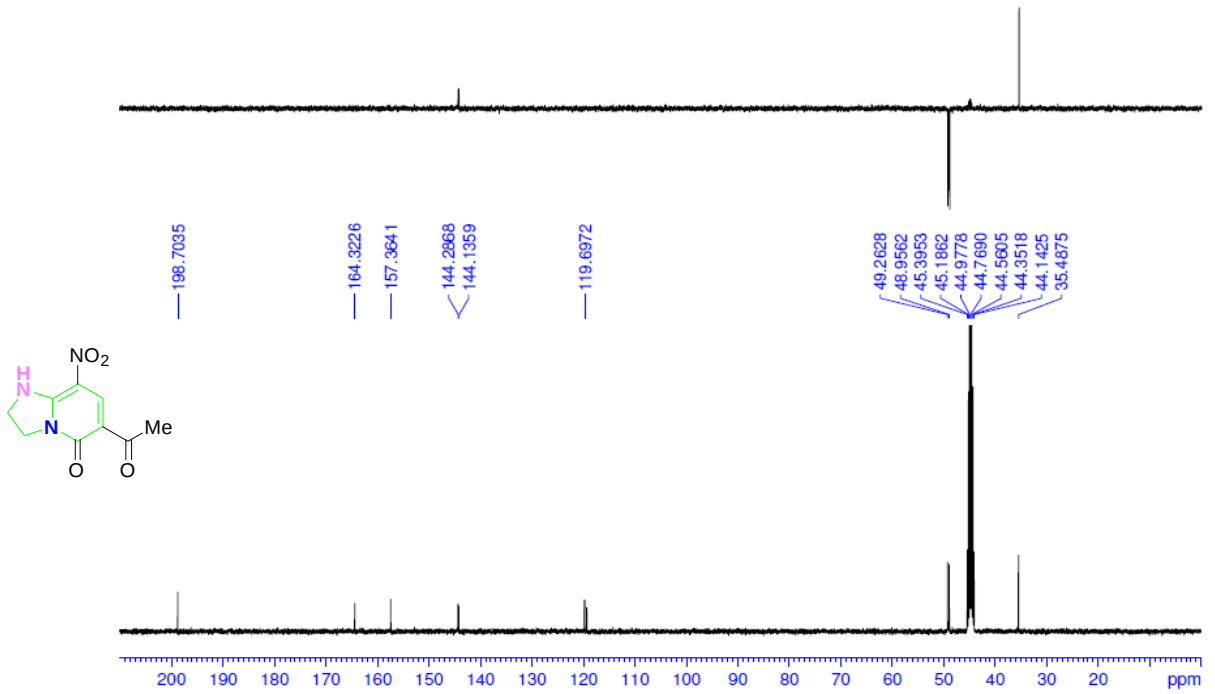


Figure 42. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4fb**

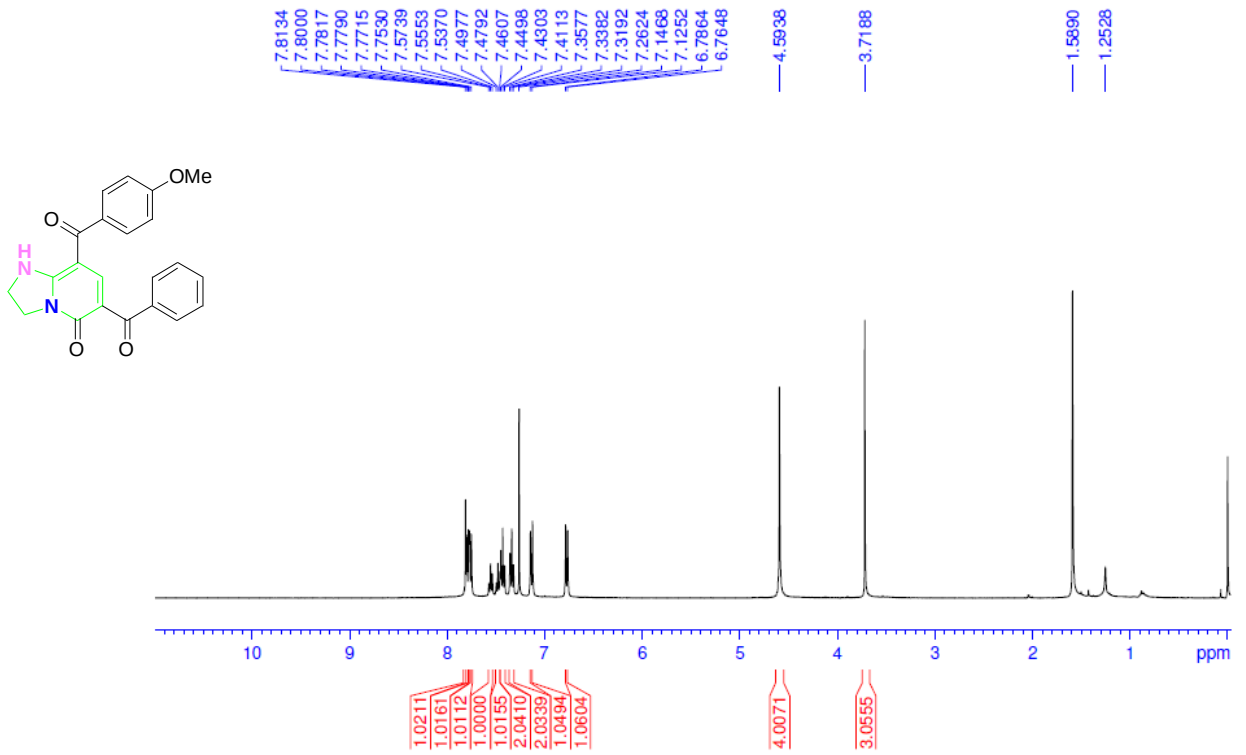


Figure 43. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4cc**

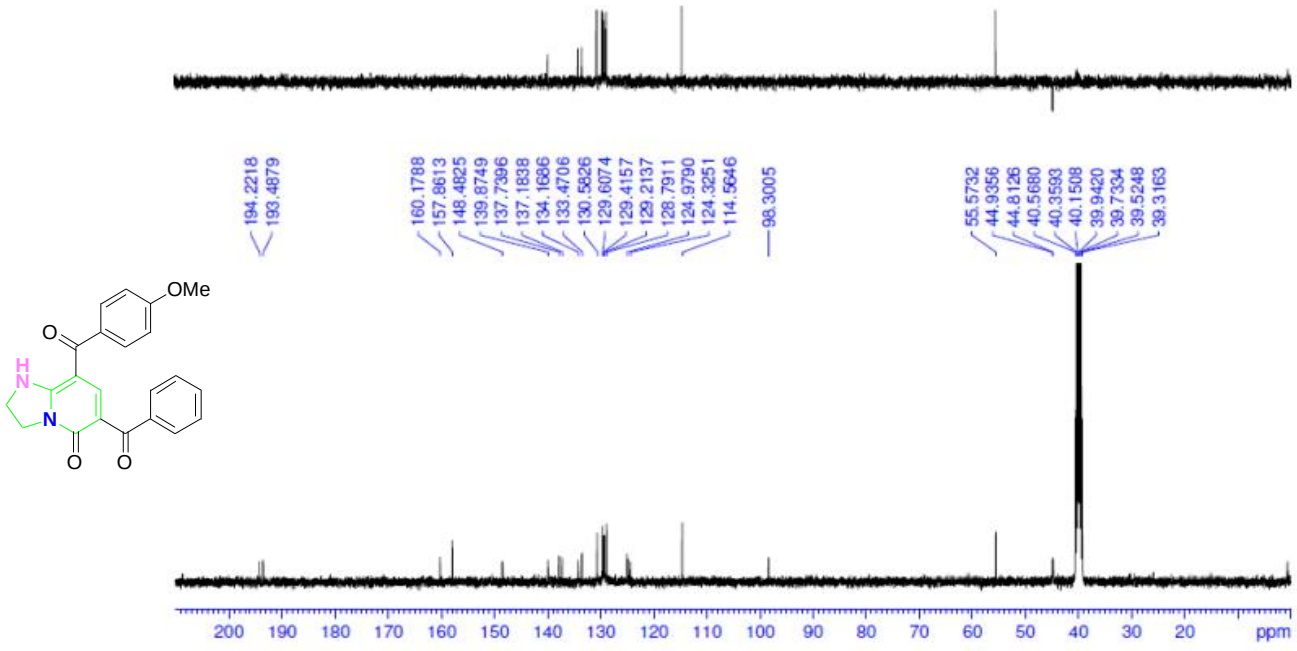


Figure 44. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4cc**

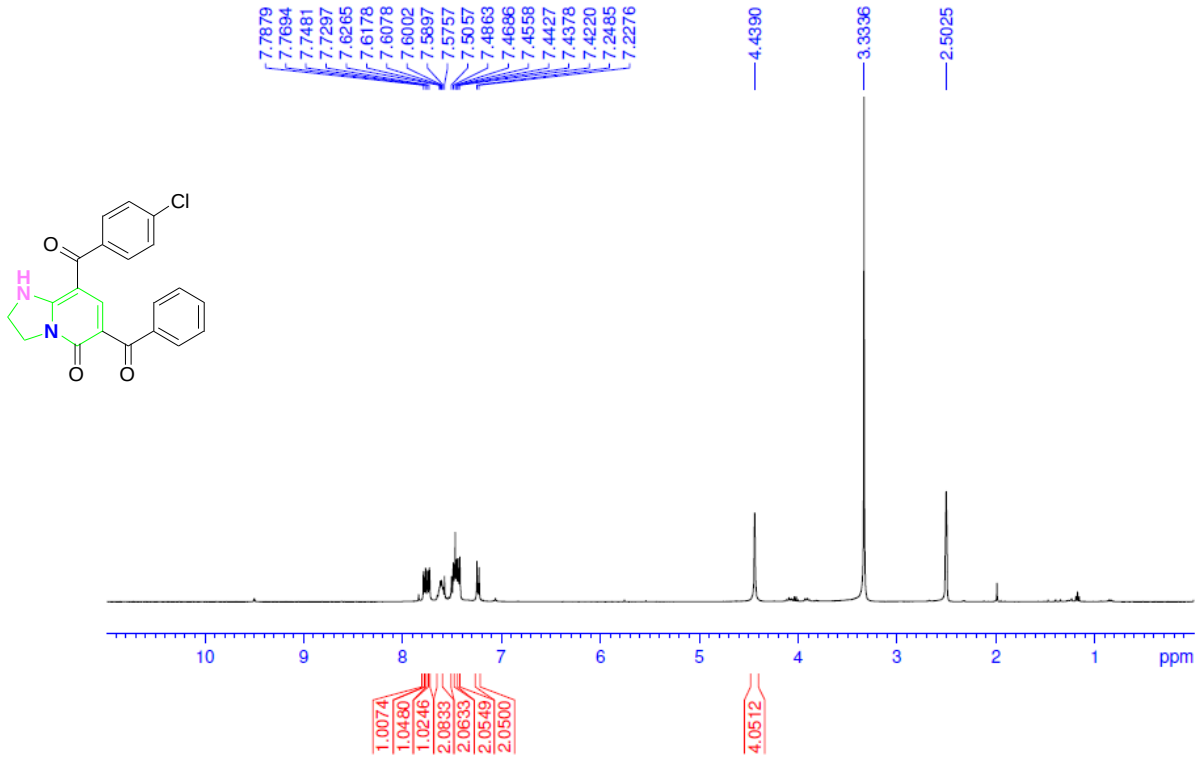


Figure 45. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4dc**

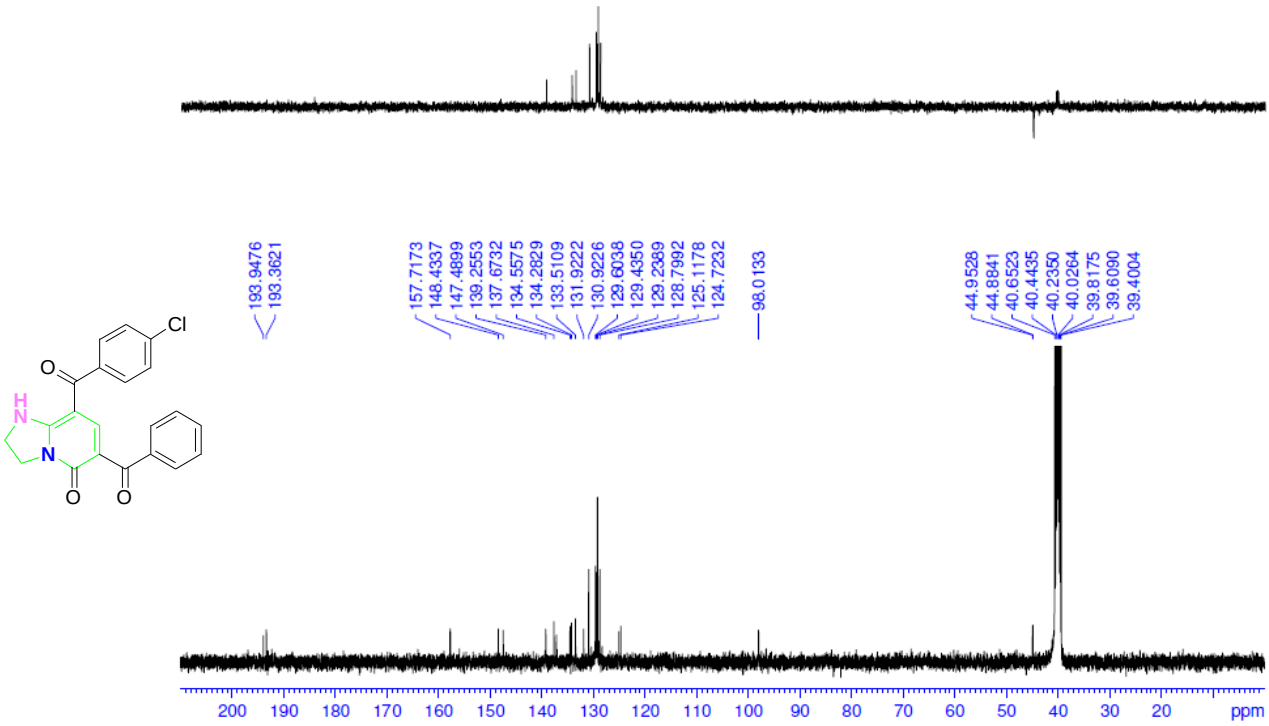


Figure 46. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4dc**

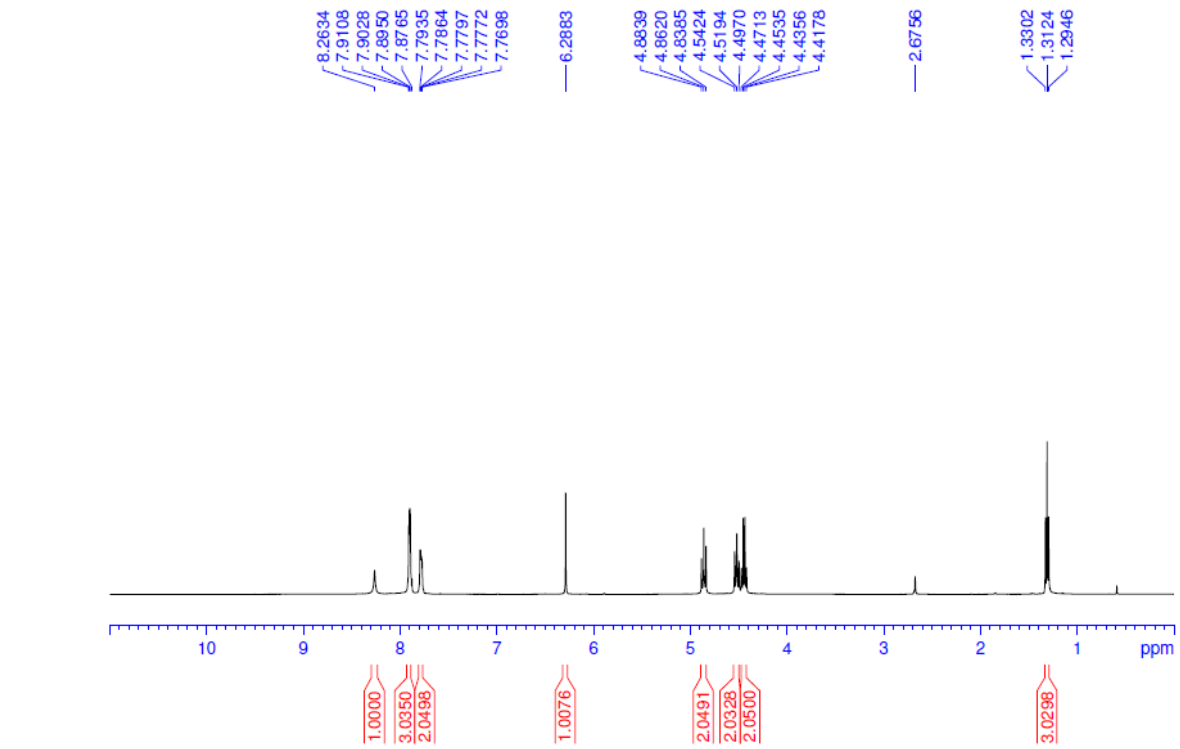


Figure 47. ^{1}H NMR (400 MHz, $CDCl_3$) spectra of compound **4ec**

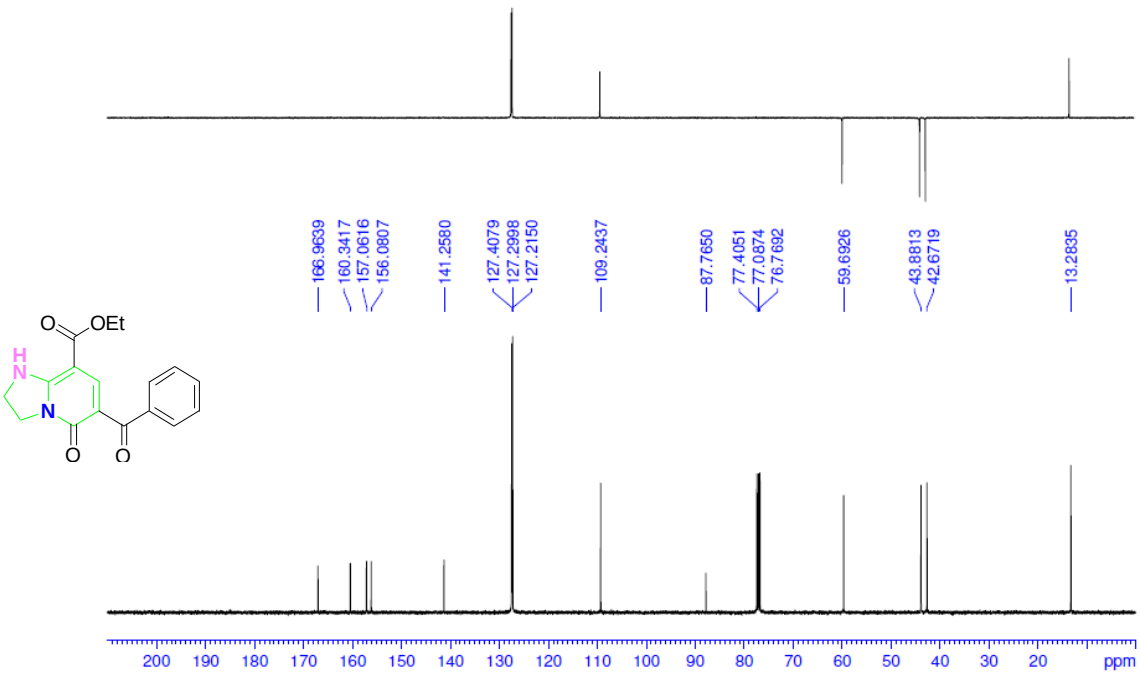


Figure 48. ^{13}C NMR (100 MHz, $CDCl_3$) spectra of compound **4ec**

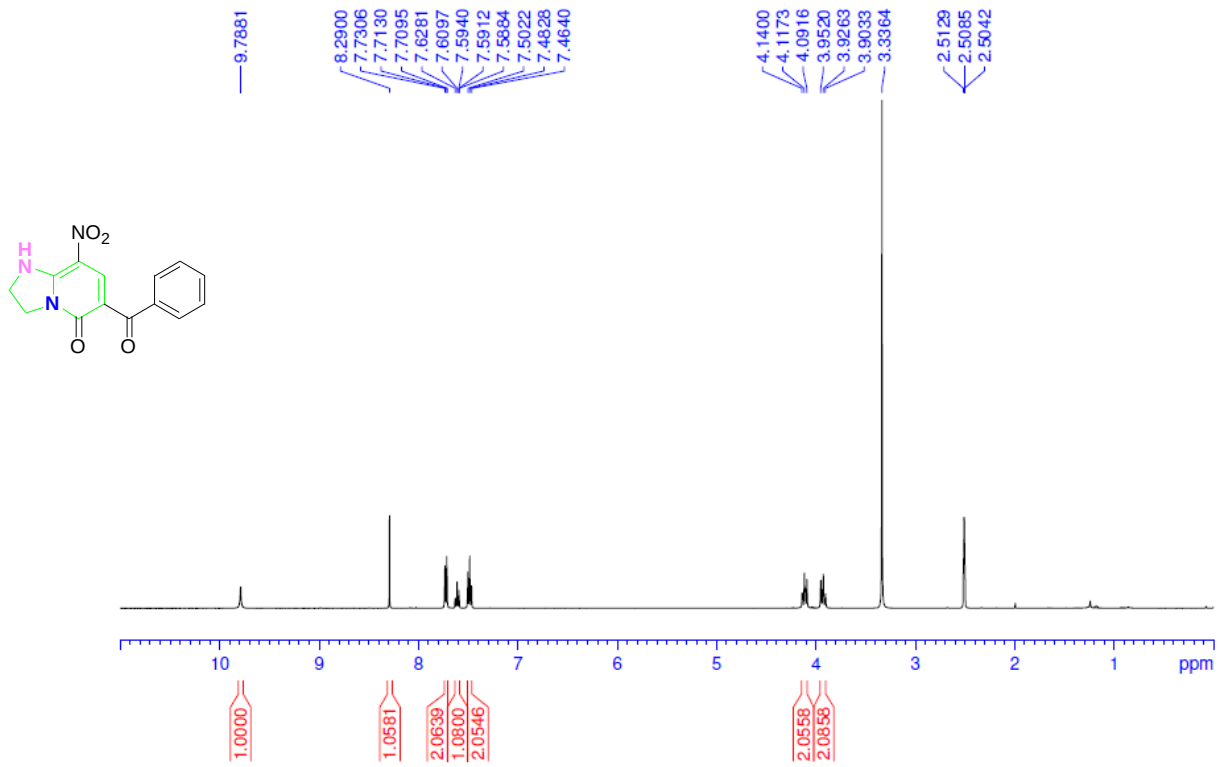


Figure 49. ^{1}H NMR (400 MHz, DMSO-d_6) spectra of compound **4fc**

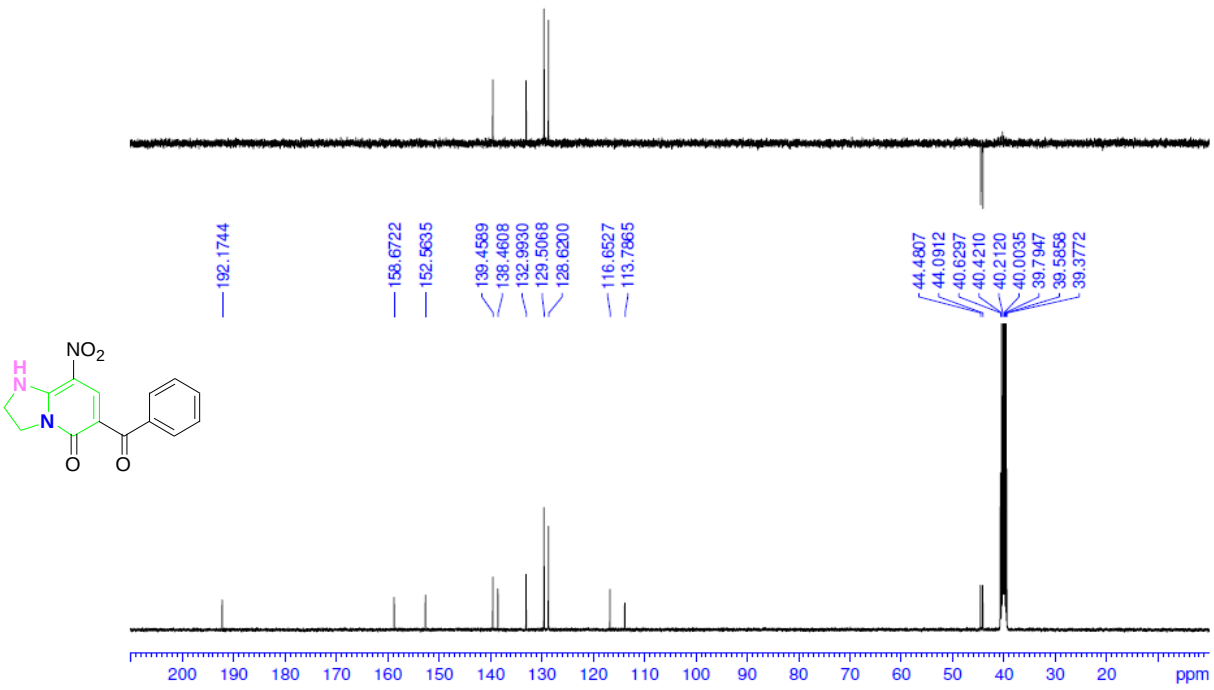


Figure 50. ^{13}C NMR (100 MHz, DMSO-d_6) spectra of compound **4fc**

Reference

1. S. Yan, Y. Chen, L. Liu, N. He and J. Lin, *Green Chem.*, 2010, **12**, 2043.