Electronic Supplementary Information

Substituent-oriented C-N bond formation *via* N-H insertion or Wolff rearrangement of 5-aryl-1*H*-pyrazoles and diazo compounds

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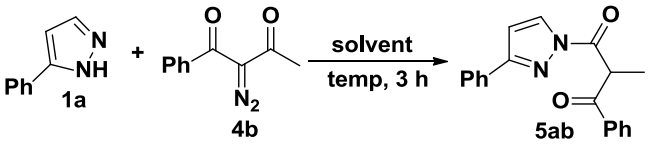
Table S1. Optimization of the Wolff-rearrangement Reaction Conditions.[a]



entry	solvent	temp (^{o}C)	yield[b] (%)
1	toluene	reflux	46
2	MeOH	reflux	trace
3	DMF	90	trace
4	DMSO	90	trace
5	DCM	reflux	42
6	DCE	reflux	81
7	MeCN	reflux	65
8	DCE	rt	12
9	DCE	50	46
10	DCE	70	64
11[c]	DCE	reflux	38
12[d]	DCE	reflux	81

[a] Reaction conditions: 5-phenyl-1*H*-pyrazole **1a** (0.5 mmol), 2-diazo-1-phenylbutane-1,3-dione **4b** (0.5 mmol), solvent (3 mL) for 3 h. [b] Isolated yields. [c] Reaction time for 1 h, [d] Reaction time for 6 h.

Experimental

General experimental method

Unless otherwise specified, all reagents and starting materials were purchased from commercial sources and used as received, and the solvents were purified and dried using standard procedures. The chromatography solvents were technical grade and distilled prior to use. The substrates of cyclic 2-diazo-1,3-diketones **1** and pyrazoles **2** are known, and synthesized by reported methods.[20,21] Flash chromatography was performed using 200-300 mesh silica gel with the indicated solvent system according to standard techniques. The ^{1}H and ^{13}C NMR data were recorded on 300, 500 MHz NMR spectrometers, unless otherwise specified. Chemical shifts (δ) in parts per million are reported relative to the residual signals of chloroform (7.26 ppm for ^{1}H and 77.16 ppm for ^{13}C), and all ^{13}C NMR were recorded with proton broadband decoupling and indicated as $^{13}C\{^{1}H\}$NMR. Multiplicities are described as s (singlet), d (doublet), t (triplet), q (quartet), or m (multiplet), and the coupling constants (*J*) are reported in Hertz. HRMS analysis with a quadrupole time-of-flight mass spectrometer yielded ion mass/charge (m/z) ratios in atomic mass units. IR spectra were measured as dry films (KBr), and the peaks are reported in terms of wave number (cm^{-1}). The melting points were measured using SGWX-4 melting point apparatus.

General procedure for the synthesis of the 3 or 7. A mixture of 5-aryl-1*H*-pyrazoles **1** (0.5 mmol) or 4,5,6,7-tetrahydro-1*H*-indazole **6a** (0.5 mmol), cyclic 2-diazo-1,3-diketones **2** (0.5 mmol), and CuTc (0.05 mmol) in DCE (3 mL) was heated to reflux in an oil bath for 6 h. After the reaction was complete (as determined using TLC), the reaction mixture was cooled to room temperature, extracted with CH_2Cl_2 (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na_2SO_4, filtered, and then evaporated under vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:8, v/v) as the elution solvent to give desired product **3** or **7**.

General procedure for the synthesis 5 or 8 or 11ab. A mixture of 5-aryl-1*H*-pyrazoles **1** (0.5 mmol) or 4,5,6,7-tetrahydro-1*H*-indazole **6a** (0.5 mmol) or 1*H*-pyrazole **6b** (0.5 mmol) or 4-methyl-1*H*-indazole **7** (0.5 mmol), and noncyclic diazo compounds **4** (0.5 mmol) in DCE (3 mL) was heated to reflux in an oil bath for 3 h. After the reaction was complete (as determined using TLC), the reaction mixture was cooled to room temperature, extracted with CH_2Cl_2 (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na_2SO_4, filtered, and then evaporated under vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:8, v/v) as the elution solvent to give desired product **5**, **8** or **11ab**.

General procedure for the synthesis 10. A mixture of 4-methyl-1*H*-indazole **9** (0.5 mmol), cyclic 2-diazo-1,3-diketones **2** (0.5 mmol), and silver carbonate (0.05 mmol) in DCE (3 mL) was heated to reflux in an oil bath for 6 h. After the reaction was complete (as determined using TLC), the reaction mixture was cooled to room temperature, extracted with CH_2Cl_2 (3 × 10 mL), and washed with brine. The organic layers were combined, dried over Na_2SO_4, filtered, and then evaporated under vacuum. The residue was purified using flash column chromatography with a silica gel (200-300 mesh), using ethyl acetate and petroleum ether (1:8, v/v) as the elution solvent to give desired products **10aa-10ac**.

Characterization Data of Compounds 3 and 7

3-Hydroxy-5,5-dimethyl-2-(3-phenyl-1*H*-pyrazol-1-yl)cyclohex-2-en-1-one (**3aa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 83% yield (117 mg, 0.42 mmol); mp 126-127 ℃; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.76 (s, 1H), 7.78 (d, J = 7.2 Hz, 2H), 7.46-7.36 (m, 3H), 6.69 (d, J = 2.4 Hz, 1H), 2.54 (s, 4H), 1.15 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.0, 150.6, 135.0, 133.9, 129.4, 128.3, 126.0, 117.4, 103.4, 47.0, 32.3, 28.6; IR (KBr) v 3157, 2976, 2357, 1667, 1610, 1498, 1387, 1264, 1218, 1102, 1083, 996, 835, 775 cm^{-1}; HRMS (APCI-TOF) m/z: [M + H]$^+$ calcd for [$C_{18}H_{20}N_2O_2$ + H]$^+$ 283.1441, found 283.1445.

3-Hydroxy-5,5-dimethyl-2-[3-(*p*-tolyl)-1*H*-pyrazol-1-yl]cyclohex-2-en-1-one (**3ba**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 83% yield (123 mg, 0.42 mmol); mp 100-101 ºC; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.77 (s, 1H), 7.67 (d, J = 8.1 Hz, 2H), 7.24 (d, J = 8.1 Hz, 2H), 6.64 (s, 1H), 2.54 (s, 4H), 2.39 (s, 3H), 1.56(s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 181.8, 150.6, 137.6, 134.8, 131.1, 130.0, 125.9, 117.2, 103.1, 47.1, 32.2, 28.6, 21.6; IR (KBr) v 3149, 2972, 2402, 1689, 1670, 1587, 1459, 1348, 1247, 1198, 1121, 1079, 1005, 917, 875, 786 cm^{-1}; HRMS (APCI-TOF) m/z: [M + H]$^+$ calcd for [$C_{18}H_{20}N_2O_2$ + H]$^+$ 297.1598, found 297.1597.

3-Hydroxy-2-[3-(4-methoxyphenyl)-1*H*-pyrazol-1-yl]-5,5 dimethylcyclohex-2-en-1-one (**3ca**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 82% yield (128 mg, 0.41 mmol); mp 112-113 ºC; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.85 (d, J = 2.4 Hz, 1H), 7.87 (d, J = 7.8 Hz, 2H), 7.68 (d, J = 7.8 Hz, 2H), 6.73 (d, J = 2.4 Hz, 1H), 3.85 (s, 3H), 2.55 (s, 4H), 1.17 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 181.8, 159.7, 150.3, 134.8, 128.8, 127.3, 126.5, 117.2, 114.8, 114.5, 102.8, 55.9, 47.1, 32.3, 28.6; IR (KBr) v 3165, 2966, 2362, 2335, 1651, 1614, 1557, 1536, 1449, 1385, 1322, 1218, 1070, 838, 778, 684 cm^{-1}; HRMS (APCI-TOF) m/z: [M + H]$^+$ calcd for [$C_{18}H_{20}N_2O_3$ + H]$^+$ 313.1547, found 313.1550.

2-[3-(4-Chlorophenyl)-1*H*-pyrazol-1-yl]-3-hydroxy-5,5-dimethylcyclohex-2-en-1-one (**3da**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (128 mg, 0.41 mmol); mp 123-124 ºC; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.84 (d, J = 2.1 Hz, 1H), 7.69 (d, J = 8.4 Hz, 2H), 7.40 (d, J = 8.4 Hz, 2H), 6.64 (d, J = 2.1 Hz, 1H), 2.54 (s, 4H), 1.16 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.3, 149.7, 135.4, 133.0, 132.7, 129.5, 127.7, 117.5, 103.6, 47.0, 32.3, 28.6; IR (KBr) v 3112, 2894, 1810, 1674, 1648, 1573, 1543, 1473, 1362, 1231, 1134, 1087, 1057, 967, 835, 755 cm^{-1}; HRMS (APCI-TOF) m/z: [M + H]$^+$ calcd for [$C_{17}H_{17}ClN_2O_2$ + H]$^+$ 317.1051, found 317.1052.

3-Hydroxy-5,5-dimethyl-2-{3-[4-(trifluoromethyl)phenyl]-1Hpyrazol-1-yl}cyclohex-2-en-1-one (**3ea**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 58 % yield (101 mg, 0.29 mmol); mp 96-97 ºC; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.85 (d, J = 2.1 Hz, 1H), 7.87 (d, J = 7.8 Hz, 2H), 7.68 (d, J = 7.8 Hz, 2H), 6.73 (d, ^4J = 2.1 Hz, 1H), 2.55 (s, 4H), 1.17 (s, 6H); δ 187.1, 154.2, 142.8, 140.5, 140.3, 133.2 (q, $^2J_{C\text{-}F}$ = 31.5 Hz), 131.2, 131.1, 130.0 (q, $^1J_{C\text{-}F}$ = 270.1 Hz), 122.3, 108.9, 51.7, 37.1, 33.4; ^{19}F NMR (470 MHz, $CDCl_3$) δ -62.6; IR (KBr) v 3167, 3048, 2534, 2415, 1858, 1689, 1634, 1599, 1547, 1531, 1474, 1428, 1316, 1208, 1120, 925, 810, 719 cm^{-1}; HRMS (APCI-TOF) m/z: [M + H]$^+$ calcd for [$C_{18}H_{17}F_3N_2O_2$ + H]$^+$ 351.1315, found 351.1320.

2-(3-([1,1'-Biphenyl]-4-yl)-1*H*-pyrazol-1-yl)-3-hydroxy-5,5-dimethylcyclohex-2-en-1-one

(**3fa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 92 % yield (165 mg, 0.46 mmol); mp 149-150 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.85 (s, 1H), 7.85 (d, J = 7.8 Hz, 2H), 7.65 (t, J = 9.0 Hz, 4H), 7.46 (t, J = 7.5 Hz, 2H), 7.38 (d, J = 7.2 Hz, 1H), 6.71 (d, J = 1.8 Hz, 1H), 2.55 (s, 4H), 1.17 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.0, 150.3, 140.5, 139.8, 135.1, 133.1, 129.8, 128.3, 127.7, 127.3, 126.5, 117.4, 103.5, 47.0, 32.3, 28.6; IR (KBr) v 3148, 3032, 2506, 2412, 1810, 1654, 1620, 1548, 1532, 1506, 1449, 1436, 1318, 1209, 1172, 914, 836, 724 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{23}H_{22}N_2O_2 + H]^+$ 359.1754, found 359.1759.

3-Hydroxy-5,5-dimethyl-2-(3-(*m*-tolyl)-1*H*-pyrazol-1-yl)cyclohex-2-en-1-one (**3ga**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 81 % yield (119 mg, 0.41 mmol); mp 89-90 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.79 (s, 1H), 7.57-7.61 (m, 2H), 7.32 (t, J = 7.5 Hz, 1H), 7.18 (d, J = 7.5 Hz, 1H), 6.66 (s, 1H), 2.54 (s, 4H), 2.41(s, 3H), 1.16 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.0, 150.6, 138.5, 134.9, 133.8, 129.3, 129.0, 126.5, 123.2, 117.3, 103.3, 47.0, 32.3, 28.6, 21.9; IR (KBr) v 3147, 3028, 2516, 2434, 1811, 1630, 1624, 1540, 1520, 1508, 1428, 1416, 1306, 1218, 1129, 938, 824, 797 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{18}H_{20}N_2O_2 + H]^+$ 297.1598, found 297.1563.

2-(3-(3-Bromophenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5,5-dimethylcyclohex-2-en-1-one (**3ha**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 82 % yield (147 mg, 0.41 mmol); mp 85-86 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.89 (s, 1H), 7.68 (d, J = 7.8 Hz, 1H), 7.48 (d, J = 7.8 Hz, 1H), 7.31 (d, J = 7.8 Hz, 1H), 6.65 (s, 1H), 2.54 (s, 4H), 1.16 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.5, 149.4, 136.5, 135.5, 131.7, 130.9, 128.3, 124.9, 122.9, 117.5, 103.8, 47.0, 32.3, 28.6; IR (KBr) v 3159, 3049, 2537, 2436, 1825, 1641, 1625, 1531, 1516, 1538, 1448, 1433, 1315, 1215, 1137, 930, 859, 748, 672 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{17}H_{17}BrN_2O_2 + H]^+$ 361.0546, found 361.0543.

2-(3-(2-Chlorophenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5,5-dimethylcyclohex-2-en-1-one (**3ia**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (134 mg, 0.43 mmol); mp 96-97 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.88 (d, J = 2.4 Hz, 1H), 7.68-7.65 (m, 1H), 7.50-7.47 (m, 1H), 7.35-7.29 (m, 2H), 6.91 (d, J = 2.4 Hz, 1H), 2.54 (s, 4H), 1.16 (s, 6H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 181.6, 147.6, 133.8, 132.0, 131.2, 130.9, 130.8, 129.6, 127.7, 116.7, 106.5, 46.6, 31.9, 28.2; IR (KBr) v 3167, 3042, 2515, 2440, 1812, 1614, 1630, 1518, 1503, 1498, 1458, 1413, 1375, 1265, 1187, 991, 876, 748, 513 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{17}H_{17}ClN_2O_2 + H]^+$ 317.1051, found 317.1055.

3-Hydroxy-5-methyl-2-(3-phenyl-1*H*-pyrazol-1-yl)cyclohex-2-en-1-one (**3ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (114 mg, 0.43 mmol); mp 84-85 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.83 (s, 1H), 7.73 (d, J = 6.9 Hz, 2H), 7.43-7.35 (m, 3H), 6.66 (s, 1H), 2.71-2.66 (m, 2H), 2.40-2.36 (m, 3H), 1.14 (s, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.8, 150.6, 135.0, 133.9, 129.5, 128.3, 125.9, 117.9, 103.3, 41.4, 28.1, 21.3; IR (KBr) v 3174, 3029, 2563, 2427, 1828, 1664, 1632, 1536, 1516, 1499, 1448, 1409, 1336, 1209, 1118, 937, 873, 749, 519 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{16}H_{16}N_2O_2 + H]^+$ 269.1285, found 269.1286.

3-Hydroxy-2-(3-(4-methoxyphenyl)-1*H*-pyrazol-1-yl)-5-methylcyclohex-2-en-1-one (**3cb**).

This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (127 mg, 0.43 mmol); mp 82-83 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.77 (s, 1H), 7.70 (d, J = 8.4 Hz, 2H), 6.96 (d, J = 8.1 Hz, 2H), 6.59 (s, 1H), 3.85 (s, 3H), 2.71-2.68 (m, 2H), 2.43-2.36 (m, 3H), 1.13 (d, J = 5.1 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.6, 159.6, 150.3, 134.7, 127.3, 126.5, 117.8, 114.8, 102.8, 55.9, 41.5, 28.0, 21.3; IR (KBr) ν 3137, 3021, 2542, 2424, 1836, 1681, 1672, 1563, 1554, 1454, 1449, 1381, 1236, 1218, 927, 871, 772, 649 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{17}H_{18}N_2O_3$ + H]$^+$ 299.1441, found 299.1443.

2-(3-(4-Fluorophenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5-methylcyclohex-2-en-1-one (**3kb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 88 % yield (126 mg, 0.44 mmol); mp 100-101 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.81 (s, 1H), 7.75-7.70 (m, 2H), 7.14-7.09 (m, 2H), 6.61 (d, J = 2.4 Hz, 1H), 2.71-2.67 (m, 2H), 2.44-2.36 (m, 3H), 1.14 (d, J = 5.4 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.1, 162.5 (q, d, $^1J_{C\text{-}F}$ = 242.3 Hz), 149.9, 135.2, 130.7 (d, $^4J_{C\text{-}F}$ = 2.5 Hz),, 127.9 (d, $^3J_{C\text{-}F}$ = 8.0 Hz), 117.4, 116.3 (d, $^2J_{C\text{-}F}$ = 21.4 Hz), 103.3, 47.0, 32.3, 28.6; ^{19}F NMR (470 MHz, $CDCl_3$) δ -112.9; IR (KBr) ν 3188, 2956, 2876, 2365, 1647, 1614, 1493, 1416, 1382, 1258, 1208, 1073, 1046, 1016, 889, 758, 590 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{16}H_{15}FN_2O_2$ + H]$^+$ 287.1190, found 287.1189.

2-(3-(4-Chlorophenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5-methylcyclohex-2-en-1-one (**3db**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 89 % yield (134 mg, 0.43 mmol); mp 111-112 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.74 (s, 1H), 7.70 (d, J = 8.4 Hz, 2H), 7.40 (d, J = 8.4 Hz, 2H), 6.65 (d, J = 2.4 Hz, 1H), 2.73-2.68 (m, 2H), 2.44-2.37 (m, 3H), 1.13 (d, J = 6.0 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.9, 149.6, 135.3, 133.0, 132.7, 129.5, 127.6, 118.1, 103.6, 41.4, 28.1, 21.3; IR (KBr) ν 3164, 2930, 2856, 2330, 1630, 1624, 1427, 1406, 1316, 1240, 1248, 1021, 1003, 872, 740, 674 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{16}H_{15}ClN_2O_2$ + H]$^+$ 303.0895, found 303.0893.

2-(3-(3-Bromophenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5-methylcyclohex-2-en-1-one (**3hb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86 % yield (149 mg, 0.43 mmol); mp 83-84 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.81 (d, J = 2.4 Hz, 1H), 7.88 (s, 1H), 7.67 (d, J = 7.8 Hz, 1H), 7.47 (d, J = 7.8 Hz, 1H), 7.30 (d, J = 8.1 Hz, 1H), 6.65 (d, J = 2.4 Hz, 1H), 2.72-2.67 (m, 2H), 2.44-2.36 (m, 3H), 1.14 (d, J = 5.7 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.9, 149.3, 136.5, 135.4, 131.7, 130.9, 128.3, 124.8, 122.9, 118.1, 103.8, 41.3, 28.1, 21.3; IR (KBr) ν 3187, 2956, 2830, 2379, 1663, 1624, 1518, 1414, 1407, 1327, 1243, 1201, 1029, 1003, 948, 832, 748, 578 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{16}H_{15}BrN_2O_2$ + H]$^+$ 347.0390, found 347.0391.

2-(3-(2,4-Dimethylphenyl)-1*H*-pyrazol-1-yl)-3-hydroxy-5-methylcyclohex-2-en-1-one (**3mb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (129 mg, 0.44 mmol); mp 77-78 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.84 (d, J = 2.4 Hz, 1H), 7.44 (d, J = 7.8 Hz, 1H), 7.10-7.06 (m, 2H), 6.51 (d, J = 2.4 Hz, 1H), 2.70-2.66 (m, 2H), 2.46 (s, 3H), 2.39-2.36 (m, 6H), 1.13 (d, J = 5.4 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.1, 150.4, 137.5, 135.8, 133.5, 132.4, 130.3, 129.5, 127.3, 117.3, 105.8, 41.5, 28.0, 22.0, 21.5, 21.2; IR (KBr) ν 3188, 2964, 2849, 2336, 1645, 1663, 1579, 1468, 1448, 1382, 1229, 1203, 1121, 1097, 948, 876, 768, 676 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{18}H_{20}N_2O_2$ + H]$^+$ 297.1598, found 297.1597.

5-Hydroxy-4-(3-phenyl-1*H*-pyrazol-1-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3ac**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 83 % yield (137 mg, 0.42 mmol); mp 90-91 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.83 (s, 1H), 7.78 (d, J = 7.2 Hz, 2H), 7.47-7.28 (m, 7H), 6.70 (s, J = 2.4 Hz, 1H), 3.57-3.44 (m, 1H), 2.94-2.91 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.5, 150.7, 143.8, 135.0, 134.0, 129.5, 125.4, 128.3, 127.8, 127.7, 125.9, 118.1, 103.4, 40.7, 38.4; IR (KBr) v 3174, 2897, 2469, 2403, 1874, 1651, 1634, 1542, 1524, 1442, 1379, 1374, 1215, 1070, 827, 718, 675 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{21}H_{18}N_2O_2 + H]^+$ 331.1441, found 331.1440.

5-Hydroxy-4-(3-(*p*-tolyl)-1*H*-pyrazol-1-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3bc**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (146 mg, 0.43 mmol); mp 151-152 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.81 (s, 1H), 7.67 (d, J = 8.1 Hz, 2H), 7.39-7.35 (m, 2H), 7.30-7.23 (m, 5H), 6.66 (d, J = 2.4 Hz, 1H), 3.54-3.44 (m, 1H), 3.93-2.91 (m, 4H), 2.39 (s, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.3, 150.7, 143.8, 137.6, 134.8, 131.2, 130.1, 129.4, 127.8, 125.9, 118.0, 103.2, 40.7, 38.2, 21.6; IR (KBr) v 3168, 2896, 2502, 2494, 1836, 1681, 1674, 1667, 1549, 1526, 1534, 1442, 1381, 1374, 1215, 1193, 927, 737, 594 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{22}H_{20}N_2O_2 + H]^+$ 345.1598, found 345.1602.

5-Hydroxy-4-(3-(4-methoxyphenyl)-1*H*-pyrazol-1-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3cc**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86% yield (154 mg, 0.43 mmol); mp 150-151 °C; ^{1}H NMR (300MHz, $CDCl_3$) δ 8.82 (s, 1H), 7.70 (d, J = 8.7 Hz, 2H), 7.40-7.26 (m, 5H), 6.97 (d, J = 8.7 Hz, 2H), 6.61 (d, J = 2.7 Hz, 1H), 3.85 (s, 3H), 3.54-3.43 (m, 1H), 2.93-2.90 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.2, 159.7, 150.5, 143.8, 134.7, 129.4, 127.8, 127.6 127.3, 126.6, 117.9, 114.9, 102.9, 55.9, 40.8, 38.4; IR (KBr) v 3154, 2836, 2520, 2434, 1812, 1675, 1677, 1636, 1579, 1549, 1542, 1435, 1336, 1309, 1294, 1173, 997, 749, 694 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{22}H_{20}N_2O_3 + H]^+$ 361.1547, found 361.1544.

4-(3-(4-Chlorophenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3dc**): This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86% yield (157 mg, 0.43 mmol); mp 156-157 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.86 (d, J = 2.1 Hz, 1H), 7.70 (d, J = 8.4 Hz, 2H), 7.42-7.36 (m, 3H), 7.31-7.26 (m, 4H), 6.67 (d, J = 2.4 Hz, 1H), 3.54-5.44 (m, 1H), 2.94-2.91 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.6, 149.8, 143.8, 135.3, 133.0, 132.7, 129.5, 129.4, 127.8, 127.6, 118.2, 103.7, 40.7, 38.4; IR (KBr) v 3179, 2963, 2881, 2372, 1664, 1670, 1549, 1498, 1428, 1372, 1259, 1236, 1179, 1075, 937, 876, 768 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{21}H_{17}ClN_2O_2 + H]^+$ 365.1051, found 365.1053.

4-(3-(4-Bromophenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3nc**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (177 mg, 0.44 mmol); mp 175-176 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.82 (s, 1H), 7.64 (d, J = 8.4 Hz, 2H), 7.56 (d, J = 8.4 Hz, 2H), 7.40-7.35 (m, 2H), 7.30-7.26 (m, 3H), 6.68 (d, J = 2.7 Hz, 1H), 3.55-3.44 (m, 1H), 2.94-2.91 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.6, 149.8, 143.8, 135.3, 133.4, 132.4, 129.4, 129.0, 127.8, 127.7, 121.2, 118.2, 103.7, 40.6, 38.4; IR (KBr) v 3184, 2976, 2849, 2314, 1687, 1679, 1548, 1494, 1400, 1382, 1378, 1279, 1179, 937, 876, 597 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{21}H_{17}BrN_2O_2 + H]^+$ 409.0546, found 409.0546.

4-(3-([1,1'-Biphenyl]-4-yl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3fc**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 93% yield (189 mg, 0.47 mmol); mp 174-175 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.84 (s, 1H), 7.85 (d, J = 8.1 Hz, 2H), 7.69-7.62 (m, 4H), 7.47 (t, J = 7.5 Hz, 2H), 7.40-7.35 (m, 3H), 7.31-7.29 (m, 3H), 6.74 (d, J = 2.4 Hz, 1H), 3.53-3.48 (m, 1H), 2.95-2.93 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.5, 150.4, 143.8, 140.5, 139.8, 135.1, 133.2, 129.8, 129.4, 128.3, 127.8, 127.7, 127.3, 126.5, 118.1, 103.6, 40.7, 38.4; IR (KBr) v 3182, 2850, 2583, 2498, 1851, 1689, 1649, 1638, 1587, 1565, 1518, 1436, 1370, 1348, 1276, 1150, 983, 747, 675 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{27}H_{22}N_2O_2$ + H]$^+$ 407.1754, found 407.1758.

4-(3-(3-Bromophenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3hc**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86% yield (175 mg, 0.43 mmol); mp 142-143 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.85 (s, 1H), 7.90 (s, 1H), 7.69 (d, J = 6.9 Hz, 1H), 7.49 (d, J = 6.6 Hz, 1H), 7.38-7.26 (m, 6H), 6.68 (s, 1H), 3.52-3.47 (m, 1H), 2.94-2.92 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.3, 149.1, 143.2, 136.2, 135.0, 131.5, 130.5, 129.0, 128.0, 127.4, 127.3, 124.5, 122.6, 117.9, 103.5, 40.4, 38.0; IR (KBr) v 3183, 2848, 2580, 2489, 1815, 1698, 1649, 1631, 1548, 1525, 1511, 1463, 1384, 1307, 1276, 1142, 938, 774, 689, 574 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{21}H_{17}BrN_2O_2$ + H]$^+$ 409.0546, found 409.0548.

4-(3-(2-Chlorophenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3ic**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85% yield (155 mg, 0.43 mmol); mp 125-126 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.89 (d, J = 2.4 Hz, 1H), 7.68-7.65 (m, 1H), 7.51-7.48 (m, 1H), 7.36-7.31 (m, 4H), 7.26-7.21 (m, 3H), 6.83 (d, J = 2.7 Hz, 1H), 3.53-3.42 (m, 1H), 2.93-2.87 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.2, 148.1, 142.7, 134.1, 132.4, 132.2, 131.6, 131.2, 129.9, 129.7, 129.3, 128.1, 117.8, 107.0, 40.6, 37.7; IR (KBr) v 3197, 2875, 2497, 2472, 1872, 1674, 1660, 1536, 1472, 1416, 1379, 1248, 1232, 1197, 1037, 927, 842, 748 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{21}H_{17}ClN_2O_2$ + H]$^+$ 365.1051, found 365.1047.

4'-Chloro-5-hydroxy-4-(3-(*p*-tolyl)-1*H*-pyrazol-1-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3bd**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86% yield (162 mg, 0.43 mmol); mp 165-166 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.82 (s, 1H), 7.66 (d, J = 8.1 Hz, 2H), 7.34 (d, J = 8.1 Hz, 2H), 7.26-7.20 (m, 4H), 6.66 (d, J = 2.4 Hz, 1H), 3.50-3.41 (m, 1H), 2.90-2.87 (m, 4H), 2.39 (s, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.2, 150.7, 142.8, 137.6, 134.8, 132.2, 130.0, 129.8, 129.3, 125.9, 118.0, 103.2, 40.7, 37.7, 21.6; IR (KBr) v 3163, 2856, 2463, 2428, 1832, 1644, 1596, 1572, 1463, 1397, 1284, 1223, 1179, 1073, 972, 824, 784 cm^{-1}; HRMS (APCI-TOF) *m/z*: [M + H]$^+$ calcd for [$C_{22}H_{19}ClN_2O_2$ + H]$^+$ 379.1208, found 379.1211.

4'-Chloro-4-(3-(4-fluorophenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3kd**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 89 % yield (170 mg, 0.45 mmol); mp 148-149 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.84 (s, 1H), 7.73 (m, 2H), 7.36-7.31 (m, 2H), 7.24-7.19 (m, 2H), 7.16-7.11 (m, 2H), 6.62 (s, 1H), 3.60-3.34 (m, 1H), 2.90-2.88 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.3, 162.6 (d, $^{1}J_{C\text{-}F}$ = 242.5 Hz), 150.0, 142.7, 135.1, 132.2, 130.7(d, $^{4}J_{C\text{-}F}$ = 2.5 Hz), 129.1, 129.3, 127.9 (d, $^{3}J_{C\text{-}F}$ = 7.9 Hz), 118.1, 116.4 (d, $^{2}J_{C\text{-}F}$ = 21.3 Hz), 103.4, 40.7, 37.8; ^{19}F NMR (470 MHz, $CDCl_3$) δ -112.7; IR (KBr) v 3174, 2869, 2454, 2448, 1872, 1656, 1572, 1569, 1493, 1378,

1279, 1273, 1197, 1167, 927, 842, 737, 634 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{22}H_{19}FClN_2O_2 + H]^+$ 383.0957，found 383.0962.

4-(3-(4-Bromophenyl)-1*H*-pyrazol-1-yl)-4'-chloro-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3nd**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 89 % yield (197 mg, 0.45 mmol); mp 181-182 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.84 (s, 1H), 7.63 (m, 2H), 7.56 (d, *J* = 8.4 Hz, 2H), 7.35 (d, *J* = 8.4 Hz, 2H), 7.23 (d, *J* = 8.4 Hz, 2H), 6.66 (m, 1H), 3.55-3.38 (m, 1H), 2.90-2.87 (m, 4H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.1, 149.5, 142.4, 134.9, 133.0, 132.0, 131.8, 129.4, 128.9, 127.6, 120.9, 117.8, 103.3, 40.4, 37.4; IR (KBr) *v* 3172, 2896, 2488, 2445, 1862, 1672, 1593, 1569, 1556, 1487, 1374, 1237, 1179, 1170, 972, 824, 773, 534 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{22}H_{19}BrClN_2O_2 + H]^+$ 443.0156, found 443.0159.

4'-Chloro-5-hydroxy-4-(3-(*m*-tolyl)-1*H*-pyrazol-1-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3gd**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (161 mg, 0.43 mmol); mp 125-126 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.82 (s, 1H), 7.57 (d, *J* = 6.6 Hz, 2H), 7.35-7.30 (m, 3H), 7.23-7.18 (m, 3H), 6.68 (s, 1H), 3.52-3.42 (m, 1H), 2.90-2.87 (m, 4H), 2.41 (s, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.3, 150.8, 142.8, 138.5, 134.9, 133.9, 132.5, 129.8, 129.4, 129.3, 129.0, 126.5, 123.1, 118.1, 103.4, 40.7, 37.8, 21.9; IR (KBr) *v* 3179, 2785, 2479, 2427, 1827, 1647, 1606, 1596, 1442, 1436, 1319, 1298, 1202, 1147, 1057, 946, 748, 549 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{22}H_{19}ClN_2O_2 + H]^+$ 379.1208, found 379.1210.

4'-Chloro-4-(3-(2,4-dimethylphenyl)-1*H*-pyrazol-1-yl)-5-hydroxy-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one (**3md**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87% yield (170 mg, 0.44 mmol); mp 169-170 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.87 (s, 1H), 7.44 (d, *J* = 7.8 Hz, 1H), 7.34 (d, *J* = 8.1 Hz, 2H), 7.22 (d, *J* = 8.1 Hz, 2H), 7.11-7.07 (m, 2H), 6.55 (s, 1H), 3.47-3.43 (m, 1H), 2.89-2.86 (m, 4H), 2.47 (s, 3H), 2.37 (s, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 181.4, 150.3, 142.4, 137.1, 135.4, 133.2, 132.0, 131.8, 130.0, 129.4, 129.1, 128.9, 126.9, 117.2, 105.6, 40.4, 37.3, 21.7, 21.1; IR (KBr) *v* 3169, 2799, 2579, 2545, 1772, 1762, 1649, 1596, 1556, 1549, 1407, 1347, 1273, 1207, 1174, 1037, 924, 774, 681 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{23}H_{21}ClN_2O_2 + H]^+$ 393.1364, found 393.1363.

3-Hydroxy-2-(3-(*p*-tolyl)-1*H*-pyrazol-1-yl)cyclohex-2-en-1-one (**3be**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:4) to afford a white solid in 77% yield (103 mg, 0.39 mmol); mp 125-126 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.82 (s, 1H), 7.66 (d, *J* = 7.8 Hz, 2H), 7.24 (d, *J* = 7.8 Hz, 2H), 6.64 (d, *J* =2.4 Hz, 1H), 2.67 (t, *J* = 6.3 Hz, 4H), 2.39 (s, 3H), 2.05 (t, *J* = 6.3 Hz, 2H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 180.4, 147.7, 143.7, 138.4, 132.7, 129.5, 128.6, 125.7, 115.0, 101.9, 33.9, 21.3, 20.0; IR (KBr) *v* 3197, 2875, 2469, 2464, 1856, 1614, 1636, 1518, 1440, 1421, 1373, 1294, 1242, 1167, 1021, 927, 848, 764, 673 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{16}H_{16}N_2O_2 + H]^+$ 269.1285, found 269.1288.

3-Hydroxy-2-(3-(4-methoxyphenyl)-1*H*-pyrazol-1-yl)cyclohex-2-en-1-one (**3ce**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:4) to afford a white solid in 78 % yield (111 mg, 0.39 mmol); mp 68-69 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.76 (s, 1H), 7.69 (d, *J* = 8.1 Hz, 2H), 6.96 (d, *J* = 8.1 Hz, 2H), 6.58 (s, 1H), 3.84 (s, 3H), 2.66 (m, 4H), 2.05 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 183.2, 159.6, 150.4, 134.7, 127.2, 126.6, 118.2, 114.8, 102.7, 55.9, 33.6, 20.6; IR (KBr) *v* 3176, 2846, 2672, 2554, 1842, 1800, 1649, 1569,

1565, 1549, 1470, 1408, 1327, 1249, 1174, 1048, 974, 687 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{16}H_{16}N_2O_3 + H]^+$ 285.1234, found 285.1232.

2-(3-(4-Fluorophenyl)-1*H*-pyrazol-1-yl)-3-hydroxycyclohex-2-en-1-one (**3ke**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:4) to afford a white solid in 79 % yield (107 mg, 0.40 mmol); mp 83-84 °C; ^1H NMR (300 MHz, $CDCl_3$) δ 8.78 (s, 1H), 7.75-7.70 (m, 2H), 7.15-7.09 (m, 2H), 6.61 (s, 1H), 2.67 (t, J = 6.3 Hz, 4H), 2.10-2.01 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 183.5, 162.5 (d, $^1J_{C\text{-}F}$ = 242.2 Hz), 149.8, 135.2, 130.7 (d, $^4J_{C\text{-}F}$ = 2.5 Hz), 127.9 (d, $^3J_{C\text{-}F}$ = 8.0 Hz), 118.4, 116.3 (d, $^2J_{C\text{-}F}$ = 21.3 Hz), 103.3, 33.5, 20.6; ^{19}F NMR (470 MHz, $CDCl_3$) δ -112.9; IR (KBr) v 3142, 2832, 2642, 2520, 1863, 1836, 1654, 1587, 1536, 1530, 1428, 1408, 1349, 1232, 1148, 1074, 987, 674 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{15}H_{13}FN_2O_2 + H]^+$ 273.1034, found 273.1033.

2-(3-(4-Bromophenyl)-1*H*-pyrazol-1-yl)-3-hydroxycyclohex-2-en-1-one (**3ne**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:4) to afford a white solid in 80 % yield (133 mg, 0.40 mmol); mp 123-124 °C; ^1H NMR (300 MHz, $CDCl_3$) δ 8.78 (s, 1H), 7.60-7.56 (m, 4H), 6.64 (s, 1H), 2.83 (m, 4H), 2.19-1.97 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 183.5, 149.7, 135.3, 133.4, 132.4, 127.9, 121.2, 118.4, 103.5, 33.5, 20.6; IR (KBr) v 3199, 2767, 2554, 2572, 1726, 1769, 1672, 1596, 1565, 1507, 1474, 1328, 1221, 1204, 1171, 1021, 824, 628, 611 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{15}H_{13}BrN_2O_2 + H]^+$ 333.0233, found 333.0230.

2-(3-(2,4-Dimethylphenyl)-1*H*-pyrazol-1-yl)-3-hydroxycyclohex-2-en-1-one (**3me**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:4) to afford a white solid in 78 % yield (110 mg, 0.39 mmol); mp 75-76 °C; ^1H NMR (300 MHz, $CDCl_3$) δ 8.81 (d, J = 2.4 Hz, 1H), 7.43 (d, J = 7.8 Hz, 1H), 7.10-7.06 (m, 2H), 6.52 (d, J = 2.4 Hz, 1H), 2.66 (t, J = 6.3 Hz, 4H), 2.46 (s, 3H), 2.36 (s, 3H), 2.05 (t, J = 6.3 Hz, 2H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 182.7, 150.4, 137.5, 135.8, 133.6, 132.4, 130.3, 129.5, 127.3, 117.6, 105.8, 33.7, 22.0, 21.5, 20.5; IR (KBr) v 3167, 2754, 2572, 2526, 1772, 1769, 1696, 1565, 1506, 1481, 1428, 1321, 1204, 1113, 1011, 879, 682 cm^{-1}; HRMS (APCI-TOF) *m/z*: $[M + H]^+$ calcd for $[C_{17}H_{18}N_2O_2 + H]^+$ 283.1441, found 283.1445.

3-Hydroxy-5,5-dimethyl-2-(4,5,6,7-tetrahydro-2*H*-indazol-2-yl)cyclohex-2-en-1-one (**7aa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 76 % yield (99 mg, 0.38 mmol); mp 112-113 °C; ^1H NMR (300 MHz, $CDCl_3$) δ 8.37 (s, 1H), 2.68 (t, J = 6.3 Hz, 2H), 2.58 (t, J = 6.3 Hz, 2H), 2.51-2.47 (m, 4H), 1.84-1.76 (m, 4H), 1.12 (s, 6H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 179.3, 145.6, 128.7, 115.5, 48.1, 31.9, 28.5, 23.6, 23.4, 23.1, 20.7; IR (KBr) v 3187, 2887, 1898, 1657, 1684, 1579, 1543, 1510, 1443, 1265, 1121, 954, 873 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{15}H_{20}N_2O_2 + H]^+$ 261.1598, found 261.1595.

3-Hydroxy-5-methyl-2-(4,5,6,7-tetrahydro-2*H*-indazol-2-yl)cyclohex-2-en-1-one (**7ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 72 % yield (88 mg, 0.36 mmol); mp 105-106 °C; ^1H NMR (300 MHz, $CDCl_3$) δ 8.39 (s, 1H), 2.69-2.55 (m, 6H), 2.83-2.31 (m, 3H), 1.82-1.75 (m, 4H), 1.10 (d, J = 5.4 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 180.2, 145.6, 128.9, 115.6, 42.4, 28.0, 23.5, 23.4, 23.1, 21.1, 20.6; IR (KBr) v 3198, 2851, 2805, 1859, 1797, 1685, 1628, 1587, 1563, 1486, 1357, 1213, 1194, 1023, 989, 847 cm^{-1};HRMS (APCI-TOF) calcd for $[C_{14}H_{18}N_2O_2 + H]^+$ 247.1441, found 247.1448.

5-Hydroxy-4-(4,5,6,7-tetrahydro-2*H*-indazol-2-yl)-1,6-dihydro-[1,1'-biphenyl]-3(2*H*)-one

(**7ac**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 63 % yield (97 mg, 0.31 mmol); mp 141-142 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.45 (s, 1H), 7.38-7.26 (m, 5H), 3.49-3.41 (m, 1H), 2.91-2.85 (m, 4H), 2.67 (t, *J* = 5. 7 Hz, 2H), 2.59 (t, *J* = 5.7 Hz, 2H), 1.84-1.77 (m, 4H); ^{13}C NMR (75 MHz, $CDCl_3$) δ179.0, 145.2, 142.1, 128.8, 128.3, 127.1, 126.6, 115.2, 114.7, 41.0, 38.0, 23.2, 23.0, 22.7, 20.3; IR (KBr) *v* 3198, 2883, 1836, 1789, 1684, 1654, 1611, 1593, 1564, 1486, 1358, 1309, 1248, 865, 786 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{20}N_2O_2 + H]^+$ 309.1598, found 309.1594.

Characterization Data of Compounds 5, 8 and 11ab

2-Methyl-1-(3-phenyl-1*H*-pyrazol-1-yl)butane-1,3-dione (**5aa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 76 % yield (92 mg, 0.38 mmol); mp 65-66 °C; ^{1}H NMR (300MHz, $CDCl_3$) δ 8.28 (d, *J* = 2.7 Hz, 1H), 7.83 (d, *J* = 6.3 Hz, 2H), 7.44 (m, 3H), 6.79 (d, *J* = 2.7 Hz, 1H), 4.80 (q, *J* = 7.2 Hz, 1H), 2.40 (s, 3H), 1.52 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75MHz, $CDCl_3$) δ 204.0, 169.0, 155.6, 153.4, 131.5, 129.7, 129.3, 128.8, 126.2, 107.9, 52.2, 29.0, 12.7; IR (KBr) *v* 3150, 2379, 2354, 1649, 1627, 1539, 1520, 1459, 1431, 1380, 1356, 1282, 1174, 768, 697 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{14}H_{14}N_2O_2 + H]^+$ 243.1128, found 243.1133.

2-Methyl-1-(3-(*p*-tolyl)-1*H*-pyrazol-1-yl)butane-1,3-dione (**5ba**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 80 % yield (102 mg, 0.40 mmol); mp 56-57 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.27 (d, *J* = 1.8 Hz, 1H), 7.63 (d, *J* = 7.2 Hz, 2H), 7.33 (t, *J* = 7.2 Hz, 1H), 7.22 (d, *J* = 7.5 Hz, 1H), 6.78 (d, *J* = 1.8 Hz, 1H), 4.82 (q, *J* = 7.2 Hz, 1H), 2.42 (s, 3H), 2.40 (s, 3H), 1.52 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 204.1, 169.1, 155.7, 139.5, 129.6, 129.5, 128.7, 126.1, 125.6, 107.8, 52.2, 29.0, 21.4, 12.7; IR (KBr) *v* 3163, 2354, 2336, 1628, 1645, 1559, 1559, 1469, 1408, 1380, 1316, 1174, 778, 547 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{15}H_{16}N_2O_2 + H]^+$ 257.1285, found 257.1286.

1-(3-([1,1'-Biphenyl]-4-yl)-1*H*-pyrazol-1-yl)-2-methylbutane-1,3-dione (**5fa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (135 mg, 0.43 mmol); mp 115-116 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.31 (s, 1H), 7.90 (d, *J* = 8.1 Hz, 2H), 7.69-7.62 (m, 4H), 7.47 (t, *J* = 7.2 Hz, 2H), 7.39 (d, *J* = 7.2 Hz, 1H), 6.83 (s, 1H), 4.82 (q, *J* = 7.2 Hz, 1H), 2.42 (s, 3H), 1.53 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 204.0, 169.0, 155.3, 142.1, 140.3, 130.4, 129.8, 128.9, 127.7, 127.5, 127.0, 126.7, 107.9, 52.2, 29.0, 12.7; IR (KBr) *v* 3159, 2779, 1849, 1697, 1634, 1625, 1593, 1548, 1495, 1413, 1356, 1308, 1228, 1147, 786, 679 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{20}H_{18}N_2O_2 + H]^+$ 319.1441, found 319.1442.

1-(3-(4-Bromophenyl)-1*H*-pyrazol-1-yl)-2-methylbutane-1,3-dione (**5na**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (139 mg, 0.44 mmol); mp 86-87°C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.29 (d, *J* = 2.7 Hz, 1H), 7.69 (d, *J* = 8.7 Hz, 2H), 7.57 (d, *J* = 8.7 Hz, 2H), 6.76 (d, *J* = 2.7 Hz, 1H), 4.78 (q, *J* = 7.2 Hz, 1H), 2.39 (s, 3H), 1.52 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 203.9, 168.9, 154.6, 132.0, 130.5, 129.9, 127.7, 123.5, 107.7, 52.2, 28.9, 12.7; IR (KBr) *v* 3148, 2797, 1856, 1687, 1643, 1635, 1575, 1568, 1459, 1431, 1365, 1282, 1174, 768, 668 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{14}H_{13}BrN_2O_2 + H]^+$ 321.0233, found 321.0230.

2-Methyl-1-(3-(4-(trifluoromethyl)phenyl)-1*H*-pyrazol-1-yl)butane-1,3-dione (**5ea**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford

a white solid in 86 % yield (139 mg, 0.44 mmol); mp 86-87°C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.32 (d, J = 2.4 Hz, 1H), 7.93 (d, J = 8.1 Hz, 2H), 7.70 (d, J = 8.1 Hz, 2H), 6.83 (d, J = 2.4 Hz, 1H), 4.80 (q, J = 7.2Hz, 1H), 2.40 (s, 3H), 1.54 (d, J = 7.2 Hz, 3H); ^{13}C NMR (125 MHz, DMSO-d_6) δ 206.0, 169.8, 154.0, 135.9, 131.5,130.3 (q, $^2J_{C\text{-}F}$ = 31.9 Hz), 127.5, 126.5, 126.5, 125.0 (q, $^1J_{C\text{-}F}$ = 270.4 Hz), 109.2, 52.7, 29.7, 13.1; ^{19}F NMR (470 MHz, $CDCl_3$) δ -62.7; IR (KBr) ν 3148, 2899, 1823, 1696, 1670, 1616, 1558, 1473, 1240, 1197, 1046; 948, 857 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{15}H_{13}F_3N_2O_2 + H]^+$ 311.1002, found 311.1004.

2-Methyl-1-(3-(4-nitrophenyl)-1*H*-pyrazol-1-yl)butane-1,3-dione (**5la**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (122 mg, 0.43 mmol); mp 99-100 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.35-8.29 (m, 3H), 7.99 (d, J = 8.7 Hz, 2H), 6.87 (d, J = 2.7 Hz, 1H), 4.80 (q, J = 7.2 Hz, 1H), 2.41 (s, 3H), 1.55 (d, J = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 203.7, 168.9, 153.3, 148.1, 137.6, 130.3, 126.9, 124.2, 108.1, 52.3, 28.8, 12.7; IR (KBr) ν 3101, 2875, 1743, 1651, 1640, 1594, 1548, 1512, 1365, 1287, 1017, 893, 753, 648 cm^{-1} HRMS (APCI-TOF) calcd for $[C_{14}H_{13}N_3O_4 + H]^+$ 288.0979, found 288.0974.

2-Methyl-1-(3-(*m*-tolyl)-1*H*-pyrazol-1-yl)butane-1,3-dione (**5ga**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 78 % yield (100 mg, 0.39 mmol); mp 56-57 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.28-8.27 (m, 1H), 7.63 (m, 2H), 7.33 (t, J = 7.2 Hz, 1H), 7.22 (m, 1H), 6.78 (m, 1H), 4.82 (q, J = 7.2 Hz, 1H), 2.42 (s, 3H), 2.40 (s, 3H), 1.52 (d, J = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 204.0, 169.1, 155.8, 138.5, 131.4, 130.1, 129.6, 128.7, 126.9, 123.4, 108.0, 52.2, 29.0, 21.4, 12.7; IR (KBr) ν 3198, 2959, 1864, 1672, 1652, 1589, 1552, 1348, 1209, 1109, 806, 766 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{15}H_{16}N_2O_2 + H]^+$ 257.1285, found 257.1286.

2-Methyl-1-phenyl-3-(3-phenyl-1*H*-pyrazol-1-yl)propane-1,3-dione (**5ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 81 % yield (123 mg, 0.40 mmol); mp 94-95 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.26 (d, J = 3.0 Hz, 1H), 8.10 (d, J = 7.5 Hz, 2H), 7.64 (t, J = 7.5 Hz, 1H), 7.56-7.48 (m, 4H), 7.29-7.26 (m, 4H), 6.75 (d, J = 3.0 Hz, 1H), 5.62 (q, J = 6.9 Hz, 1H), 1.63 (d, J = 6.9 Hz, 1H); ^{13}C NMR (75 MHz $CDCl_3$) δ 196.9, 169.4, 155.2, 135.6, 133.3, 131.4, 129.6, 129.1, 128.9, 128.8, 128.7, 128.6, 126.3, 126.1, 107.6, 47.3, 13.6; IR (KBr) ν 3198, 2844, 1867, 1778, 1762, 1654, 1625, 1568, 1451, 1411, 1245, 1067, 937, 826, 759 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{16}N_2O_2 + H]^+$ 305.1285, found 305.1282.

2-Methyl-1-phenyl-3-(3-(*p*-tolyl)-1*H*-pyrazol-1-yl)propane-1,3-dione (**5bb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 84 % yield (133 mg, 0.42 mmol); mp 129-130 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.26 (d, J = 3.0 Hz, 1H), 8.10 (d, J = 7.5 Hz, 2H), 7.64 (t, J = 7.5 Hz, 1H), 7.53 (t, J = 7.5 Hz, 2H), 7.38 (d, J = 7.8 Hz, 2H), 7.08 (d, J = 7.8 Hz, 2H), 6.71 (d, J = 3.0 Hz, 1H), 5.61 (q, J = 7.2 Hz, 1H), 2.33 (s, 3H), 1.62 (d, J = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.9, 169.4, 155.3, 139.2, 135.7, 133.3, 129.5, 129.3, 128.7, 128.6, 126.0, 109.9, 107.5, 47.3, 21.3, 13.6; IR (KBr) ν 3198, 2889, 1853, 1663, 1647, 1632, 1582, 1569, 1456, 1354, 1258, 1175, 947, 832 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{20}H_{18}N_2O_2 + H]^+$ 319.1441, found 319.1445.

1-(3-(4-Methoxyphenyl)-1*H*-pyrazol-1-yl)-2-methyl-3-phenylpropane-1,3-dione (**5cb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (142 mg, 0.42 mmol); mp 117-118 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ

8.24 (d, *J* = 2.7 Hz, 1H), 8.09 (d, *J* = 6.9 Hz, 2H), 7.66-7.62 (m, 1H), 7.53 (t, *J* = 7.5 Hz, 2H), 7.43-7.41 (m, 2H), 6.79 (d, *J* = 8.7 Hz, 2H), 6.68 (d, *J* = 2.7 Hz, 1H), 5.60 (q, *J* = 7.2 Hz, 1H), 3.80 (s, 3H), 1.62 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 197.0, 169.3, 160.4, 155.0, 135.7, 133.3, 129.5, 128.8, 128.7, 127.4, 124.1, 113.9, 107.3, 55.2, 47.3, 13.6; IR (KBr) *v* 3121, 2812, 1818, 1651, 1629, 1569, 1524, 1479, 1345, 1289, 1161, 1023, 852, 739 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{20}H_{18}N_2O_3 + H]^+$ 335.1390, found 335.1393.

1-(3-(4-Fluorophenyl)-1*H*-pyrazol-1-yl)-2-methyl-3-phenylpropane-1,3-dione (**5kb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 86 % yield (138 mg, 0.43 mmol); mp 118-119 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.27 (d, *J* = 2.7 Hz, 1H), 8.09 (d, *J* = 7.2 Hz, 2H), 7.65 (t, *J* = 7.2 Hz, 1H), 7.54 (t, *J* = 7.8 Hz, 2H), 7.46-7.41 (m, 2H), 6.94 (t, *J* = 8.7 Hz, 2H), 6.69 (d, *J* = 2.7 Hz, 1H), 5.58 (q, *J* = 6.9 Hz, 1H), 1.63 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.9, 169.3, 163.2 (d, $^1J_{C\text{-}F}$ = 247.6 Hz), 154.3, 135.6, 133.4, 129.7, 128.8 (d, $^3J_{C\text{-}F}$ = 9.1 Hz), 128.6, 127.9 (d, $^4J_{C\text{-}F}$ = 8.2 Hz), 127.6, 127.6, 115.6 (d, $^2J_{C\text{-}F}$ = 21.6 Hz), 107.3, 47.3, 13.6; ^{19}F NMR (470 MHz, $CDCl_3$) δ -111.9; IR (KBr) *v* 3118, 2891, 1854, 1703, 1682, 1642, 1582, 1526, 1452, 1352, 1265, 1174, 1083, 974, 674 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{15}FN_2O_2 + H]^+$ 323.1190, found 323.1194.

1-(3-(4-Chlorophenyl)-1*H*-pyrazol-1-yl)-2-methyl-3-phenylpropane-1,3-dione (**5db**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (147 mg, 0.43 mmol); mp 148-149 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.28 (d, *J* = 2.7 Hz, 1H), 8.09 (d, *J* = 7.2 Hz, 2H), 7.66 (t, *J* = 7.2 Hz, 1H), 7.54 (d, *J* = 7.2 Hz, 2H), 7.39-7.36 (m, 2H), 7.24-7.21 (m, 2H), 6.70 (d, *J* = 2.4 Hz, 1H), 5.57 (q, *J* = 7.2 Hz, 1H), 1.63 (d, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.9, 169.3, 154.1, 135.5, 135.0, 133.4, 129.9, 129.8, 128.8, 128.8, 128.7, 127.3, 107.3, 47.3, 13.5; IR (KBr) *v* 3127, 2874, 1832, 1683, 1661, 1630, 1576, 1529, 1459, 1431, 1380, 1254, 1127, 957, 769 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{15}ClN_2O_2 + H]^+$ 339.0895, found 339.0898.

2-Methyl-1-phenyl-3-(3-(4-(trifluoromethyl)phenyl)-1*H*-pyrazol-1-yl)propane-1,3-dione (**5eb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (147 mg, 0.43 mmol); mp 134-135 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.32 (d, *J* = 2.7 Hz, 1H), 8.10 (d, *J* = 7.2 Hz, 2H), 7.68 (t, *J* = 7.2 Hz, 1H), 7.58-7.48 (m, 6H), 6.78 (d, *J* = 2.7 Hz, 1H), 5.59 (q, *J* = 6.9 Hz, 1H), 1.64 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 197.3, 169.7, 154.1, 135.8, 135.2, 133.9, 131.2 (q, $^2J_{C\text{-}F}$ = 32.3 Hz), 130.3, 129.3, 129.1, 126.6, 129.5, 125.9, 124.3 (q, $^1J_{C\text{-}F}$ = 270.0 Hz), 107.9, 47.7, 14.0; ^{19}F NMR (470 MHz, $CDCl_3$) δ -62.7; IR (KBr) *v* 3197, 2866, 1896, 1759, 1650, 1645, 1601, 1530, 1526, 1456, 1357, 1203, 1187, 1086, 1034, 978, 854 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{15}F_3N_2O_2 + H]^+$ 373.1158, found 373.1154.

2-Methyl-1-(3-(4-nitrophenyl)-1*H*-pyrazol-1-yl)-3-phenylpropane-1,3-dione (**5lb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 88 % yield (153 mg, 0.44 mmol); mp 152-153 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.34 (d, *J* = 2.4 Hz, 1H), 8.10 (d, *J* = 7.8 Hz, 4H), 7.73-7.69 (m, 1H), 7.60-7.55 (m, 4H), 6.81 (d, *J* = 2.4 Hz, 1H), 5.57 (q, *J* = 6.9 Hz, 1H), 1.65 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.8, 169.2, 152.8, 147.9, 137.5, 135.3, 133.7, 130.2, 129.0, 128.7, 126.6, 123.9, 107.7, 47.3, 13.5; IR (KBr) *v* 3131, 2894, 1857, 1710, 1658, 1641, 1526, 1453, 1394, 1203, 1187, 1031, 945, 794 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{15}N_3O_4 + H]^+$ 350.1135, found 350.1133.

1-(3-(2-Chlorophenyl)-1*H*-pyrazol-1-yl)-2-methyl-3-phenylpropane-1,3-dione (**5ib**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 84 % yield (142 mg, 0.42 mmol); mp 78-79 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.30 (d, *J* = 2.7 Hz, 1H), 8.08 (d, *J* = 7.5 Hz, 2H), 7.62 (t, *J* = 7.5 Hz, 1H), 7.50 (t, *J* = 7.5 Hz, 2H), 7.39 (d, *J* = 7.8 Hz, 1H), 7.31-7.21 (m, 2H), 7.07 (t, *J* = 7.5 Hz, 1H), 6.99 (d, *J* = 2.7 Hz, 1H), 5.63 (q. *J* = 6.9 Hz, 1H), 1.63 (d, *J* = 6.9 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.8, 169.4, 153.4, 135.4, 133.4, 132.6, 130.5, 130.4, 129.9, 128.8, 128.7, 126.7, 111.4, 47.3, 13.6; IR (KBr) *v* 3187, 2895, 1867, 1698, 1607, 1590, 1548, 1460, 1440, 1349, 1187, 1015, 986, 795 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{19}H_{15}ClN_2O_2 + H]^+$ 339.0895, found 339.0896.

1-(3-(2,4-Dimethylphenyl)-1*H*-pyrazol-1-yl)-2-methyl-3-phenylpropane-1,3-dione (**5mb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 85 % yield (142 mg, 0.42 mmol); mp 89-90 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.30 (d, *J* = 2.7 Hz, 1H), 8.04 (d, *J* = 7.5 Hz, 2H), 7.60 (t, *J* = 7.5 Hz, 1H), 7.48 (t, *J* = 7.5 Hz, 2H), 6.97-6.94 (m, 2H), 6.63 (d, *J* = 2.7 Hz, 1H), 5.63 (q, *J* = 7.2 Hz, 1H), 2.30 (s, 3H), 2.18 (s, 3H), 1.63 (d, *J* = 7.2 Hz, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.7, 169.6, 156.2, 138.6, 136.4, 135.2, 133.5, 131.9, 130.1, 129.3, 129.2, 129.0, 128.8, 126.5, 110.5, 47.3, 21.1, 21.1, 13.7; IR (KBr) *v* 3176, 2876, 1868, 1766, 1684, 1630, 1583, 1564, 1461, 1243, 1086, 961, 746 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{21}H_{20}N_2O_2 + H]^+$ 333.1598, found 333.1600.

2-Methyl-1-phenyl-3-(4,5,6,7-tetrahydro-2*H*-indazol-2-yl)propane-1,3-dione (**8ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 81 % yield (115 mg, 0.41 mmol); mp 117-118 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.01 (d, *J* = 7.5 Hz, 2H), 7.91 (s, 1H), 7.57 (t, *J* = 7.2 Hz, 1H), 7.47 (t, *J* = 7.5 Hz, 2H), 5.57 (q, *J* = 7.2 Hz, 1H), 2.57 (m, 4H), 1.84-1.71 (m, 4H), 1.58-1.55 (m, 4H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.9, 169.3, 155.4, 135.6, 133.2, 128.7, 124.7, 121.3, 47.0, 23.7, 22.8, 20.6, 13.8; IR (KBr) *v* 3115, 2810, 1798, 1657, 1649. 1597, 1568, 1564, 1404, 1311, 1389, 1134, 943, 883 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{17}H_{18}N_2O_2 + H]^+$ 283.1441, found 283.1445.

2-Methyl-1-phenyl-3-(1*H*-pyrazol-1-yl)propane-1,3-dione (**8bb**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 83 % yield (95mg, 0.41 mmol); mp 117-118 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.27 (d, *J* = 8.4 Hz, 1H), 8.06-8.04 (m, 3H), 7.61-7.57 (m, 1H), 7.51-7.42 (m, 3H), 5.65 (q, *J* = 7.2 Hz, 1H), 2.57 (s, 3H), 1.65 (q, *J* = 7.2 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.7, 169.6, 144.1, 135.2, 133.5, 128.8, 128.7, 128.5, 109.9, 47.4, 13.7; IR (KBr) *v* 3176, 1766, 1671, 1664, 1508, 1464, 1243, 1177, 966, 746 cm^{-1};HRMS (APCI-TOF) calcd for $[C_{13}H_{12}N_2O_2 + H]^+$ 229.0972, found 229.0975.

2-Methyl-1-(4-methyl-1*H*-indazol-1-yl)-3-phenylpropane-1,3-dione (**11ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 87 % yield (127 mg, 0.43 mmol); mp 112-113°C; ^{1}H NMR (300MHz, $CDCl_3$) δ 8.27 (d, *J* = 8.4 Hz, 1H), 8.05 (t, *J* = 7.5 Hz, 3H), 7.59 (t, *J* = 7.5 Hz, 1H), 7.48-7.42 (m, 3H), 7.13 (d, *J* = 7.4 Hz, 1H), 5.65 (q, *J* = 7.2 Hz, 1H), 2.57 (s, 3H), 1.65 (d, *J* = 7.5 Hz, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 197.1, 170.8, 139.0, 135.5, 133.3, 131.5, 128.8, 128.7, 126.5, 125.0, 112.8, 48.2, 18.3, 13.8; IR (KBr) *v* 3109, 2863, 1865, 1767, 1684, 1621, 1570, 1542, 1531, 1489, 1360, 1254, 1195, 1089, 876, 790 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{13}H_{14}N_2O_2 + H]^+$ 293.1285, found 293.1290.

Characterization Data of Compounds 10aa-10ac

2,2-Dimethyl-5-(4-methyl-1*H*-indazole-1-carbonyl)cyclopentanone (**10aa**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 78 % yield (105 mg, 0.39 mmol); mp 115-116 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.22 (m, 2H), 7.45 (t, J = 7.8 Hz, 1H), 7.15 (d, J = 7.2 Hz, 1H), 4.91-4.85 (m, 1H), 2.61 (s, 3H), 2.51-2.40 (m, 2H), 2.32-2.21 (m, 2H), 1.30 (s, 3H), 1.18 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 212.2, 169.9, 139.2, 139.0, 131.5, 129.7, 126.6, 125.2, 113.0, 54.3, 53.3, 41.3, 34.8, 29.2, 27.9, 18.3; IR (KBr) v 3176, 2859, 1860, 1785, 1600, 1564, 1516, 1471, 1354, 1258, 1134, 1041, 923, 827 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{18}H_{18}N_2O_2 + H]^+$ 271.1441, found 271.1444.

2-Methyl-5-(4-methyl-1*H*-indazole-1-carbonyl)cyclopentanone (**10ab**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 77 % yield (98 mg, 0.38 mmol); mp 115-116 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.26-8.18 (m, 2H), 7.45 (t, J = 7.8 Hz, 1H), 7.15 (d, J = 7.2 Hz, 1H), 4.85-4.70 (m, 1H), 2.71-2.50 (m, 5H), 2.37-2.07 (m, 3H), 1.26-1.18 (m, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 211.9, 169.6, 139.1, 139.0, 131.5, 129.7, 126.6, 125.1, 113.0, 56.5, 52.9, 46.8, 46.7, 35.9, 35.2, 30.0, 29.0, 20.7, 19.9, 18.3; IR (KBr) v 3110, 2812, 1842, 1678, 1666, 1580, 1539, 1501, 1458, 1346, 1258, 1134, 1082, 847, 796 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{15}H_{16}N_2O_2 + H]^+$ 257.1285, found 257.1281.

2-(4-methyl-1*H*-indazole-1-carbonyl)-5-phenylcyclopentanone (**10ac**). This compound was purified by column chromatography (ethyl acetate/petroleum ether = 1:8) to afford a white solid in 84 % yield (133 mg, 0.42 mmol); mp 115-116 ^{o}C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 8.27-8.22 (m, 2H), 7.47 (t, J = 7.8 Hz, 1H), 7.37-7.29 (m, 5H), 7.16 (d, J = 6.9 Hz, 1H), 4.96-4.78 (m, 1H), 3.59-3.46 (m, 1H), 3.00-2.69 (m, 3H), 2.62 (s, 3H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 210.5, 169.3, 152.4, 143.0, 141.9, 139.4, 139.4, 139.0, 131.6, 129.8, 128.8, 127.0, 126.9, 126.7, 126.6, 125.3, 113.0, 112.9, 56.6, 53.2, 46.0, 45.6, 40.4, 39.6, 35.6, 35.4, 18.3; IR (KBr) v 3129, 2872, 1865, 1762, 1640, 1594, 1588, 1456, 1371, 1360, 1265, 1074, 985, 693 cm^{-1}; HRMS (APCI-TOF) calcd for $[C_{20}H_{18}N_2O_2 + H]^+$ 319.1441,found 319.1444.

X-Ray Crystallography structures of Compound **3nc, 5cb** and **10ac**

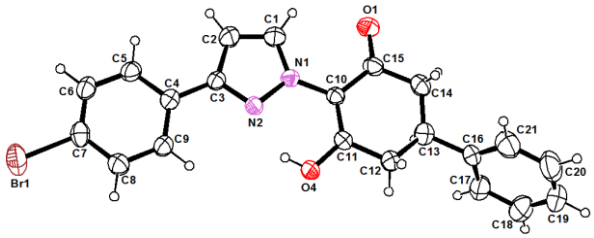


Figure S1. X-ray crystal structure of **3nc**

Crystal data for **3nc**: $C_{21}H_{16}BrN_2O_2$, Mr = 408.26, Orthorhombic, *a* = 11.3784(10) Å, *b* = 8.5648(7) Å, *c* = 37.271(3) Å, *α* = 90 °, *β* = 90 °, *γ* = 90 °, *V* = 3632.2(5) Å^3, *T* = 293 (2) K, space group Pbca, Z = 8, 34343 reflections collected, 3336 unique (Rint = 0.0666) which were used in all calculation. The ellipsoid contour probability level in the caption of 30 %. Crystallographic data for compound **3nc** reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. **CCDC-1919855**.

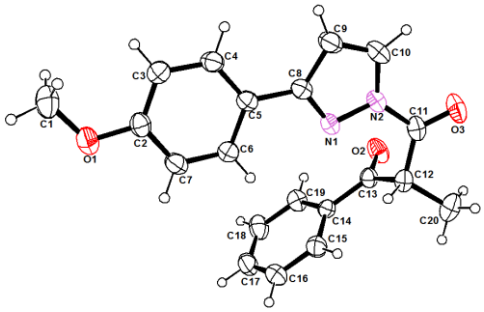


Figure S2. X-ray crystal structure of **5cb**

Crystal data for **5cb**: $C_{20}H_{18}N_2O_3$, Mr = 334.36, Orthorhombic, *a* = 10.0547(13) Å, *b* = 19.198(3) Å, *c* = 8.7595(13) Å, *α* = 90 °, *β* = 90 °, *γ* = 90 °, *V* = 1690.8(4) Å^3, *T* = 293 (2) K, space group $Pna2_1$, Z = 4, 10996 reflections collected, 3330 unique (R(int) = 0.0727) which were used in all calculation. The ellipsoid contour probability level in the caption of 30 %. Crystallographic data for compound **5cb** reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. **CCDC-1919853**.

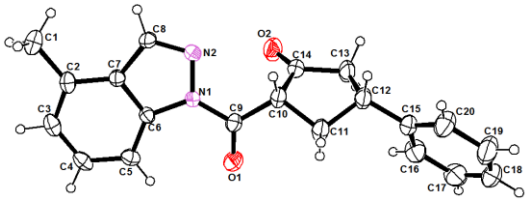
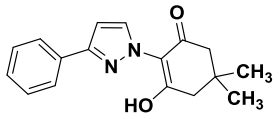
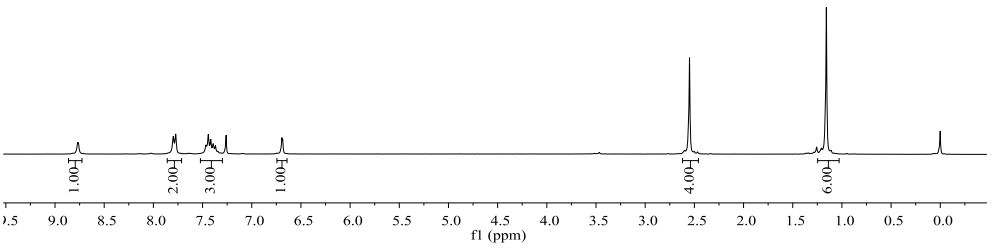


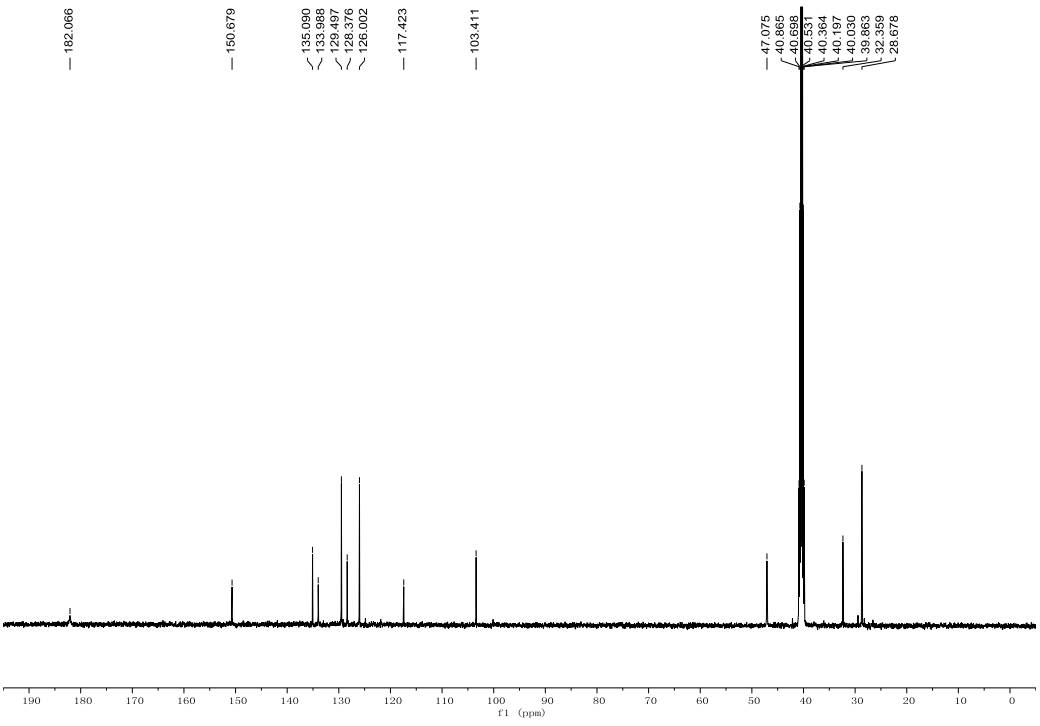
Figure S3. X-ray crystal structure of **10ac**

Crystal data for **10ac**: $C_{20}H_{18}N_2O_2$, Mr = 318.36, Monoclinic, *a* = 6.8253(4) Å, *b* = 7.8483(4) Å, *c* = 30.9897(18) Å, *α* = 90 °, *β* = 97.986(2) °, *γ* = 90 °, *V* = 1690.8(4) Å^3, *T* = 293 (2) K, space group $P2_1/c$, Z = 4, 32327 reflections collected, 2988 unique (R(int) = 0.0260) which were used in all calculation. The ellipsoid contour probability level in the caption of 30 %. Crystallographic data for compound **5cb** reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. **CCDC-1919853**.

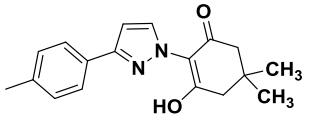
^{1}H NMR and ^{13}C NMR Spectra of Compound 3aa

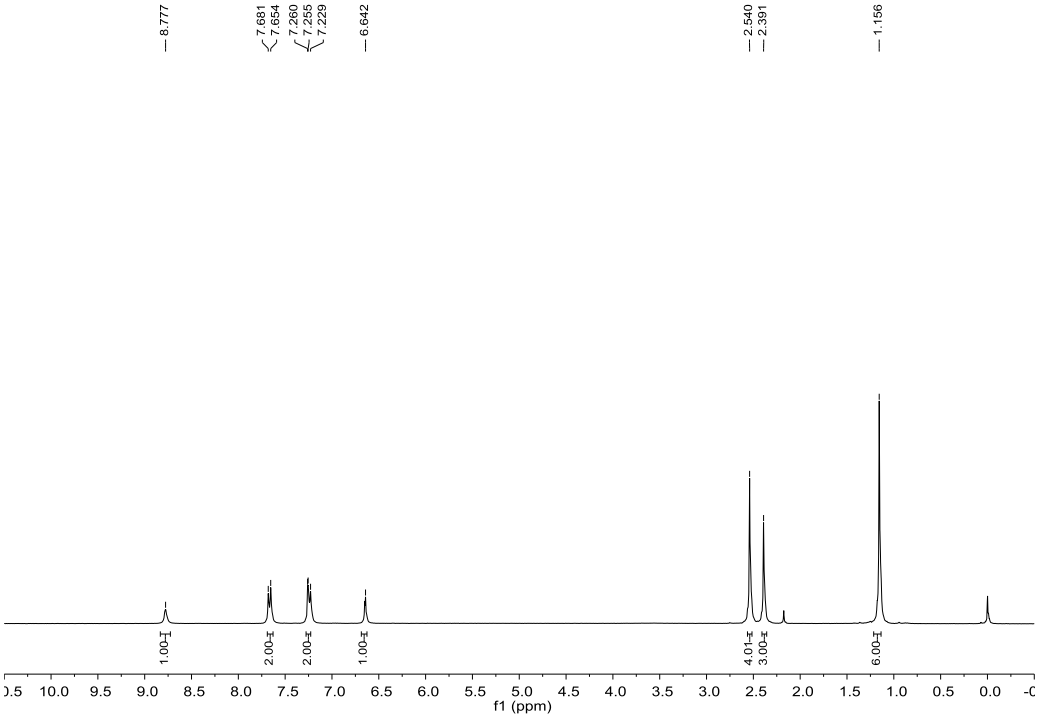


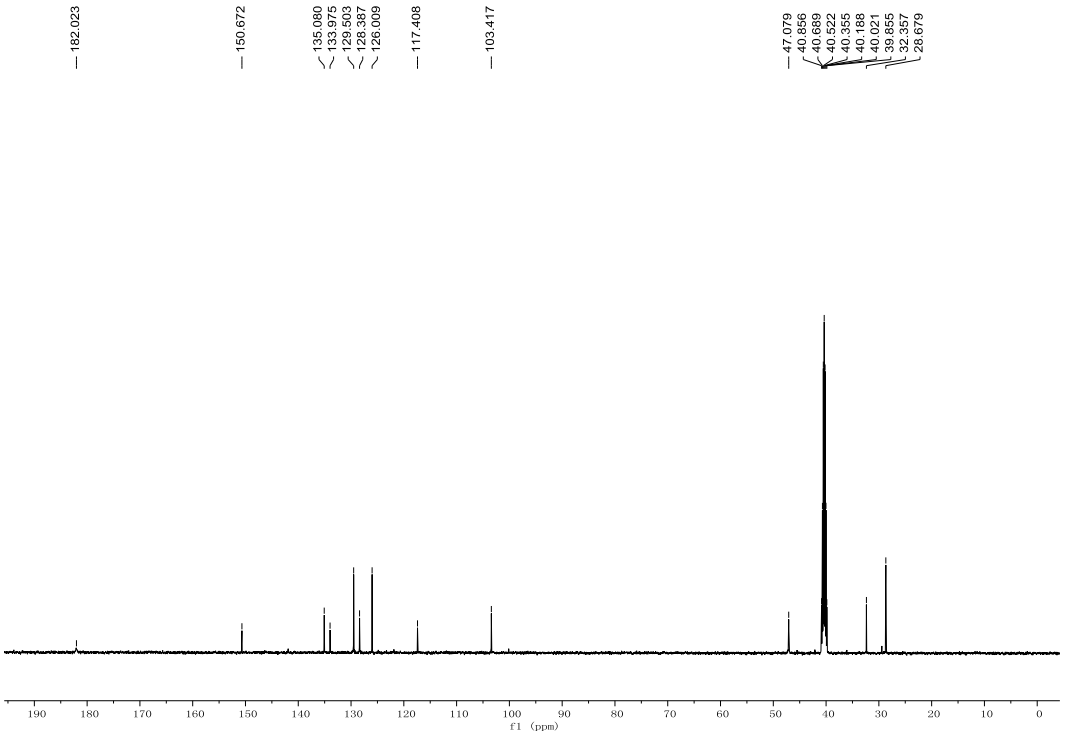




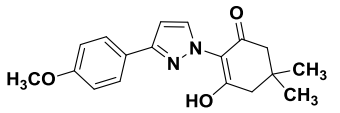
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ba







^{1}H NMR and ^{13}C NMR Spectra of Compound 3ca



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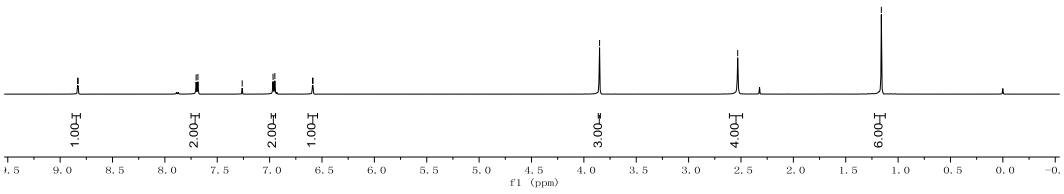
1.160



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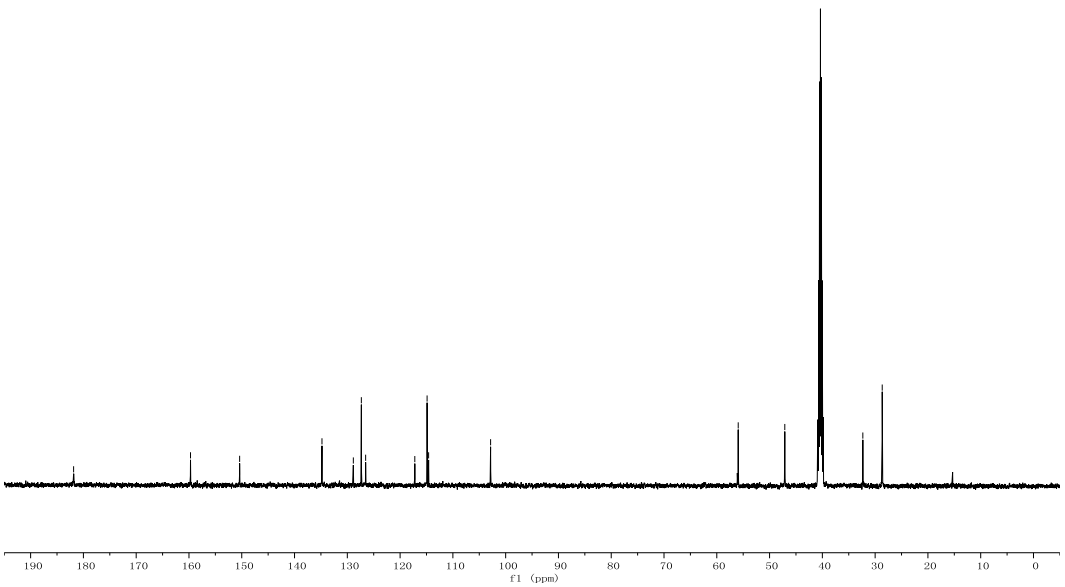
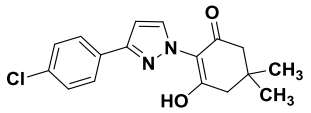
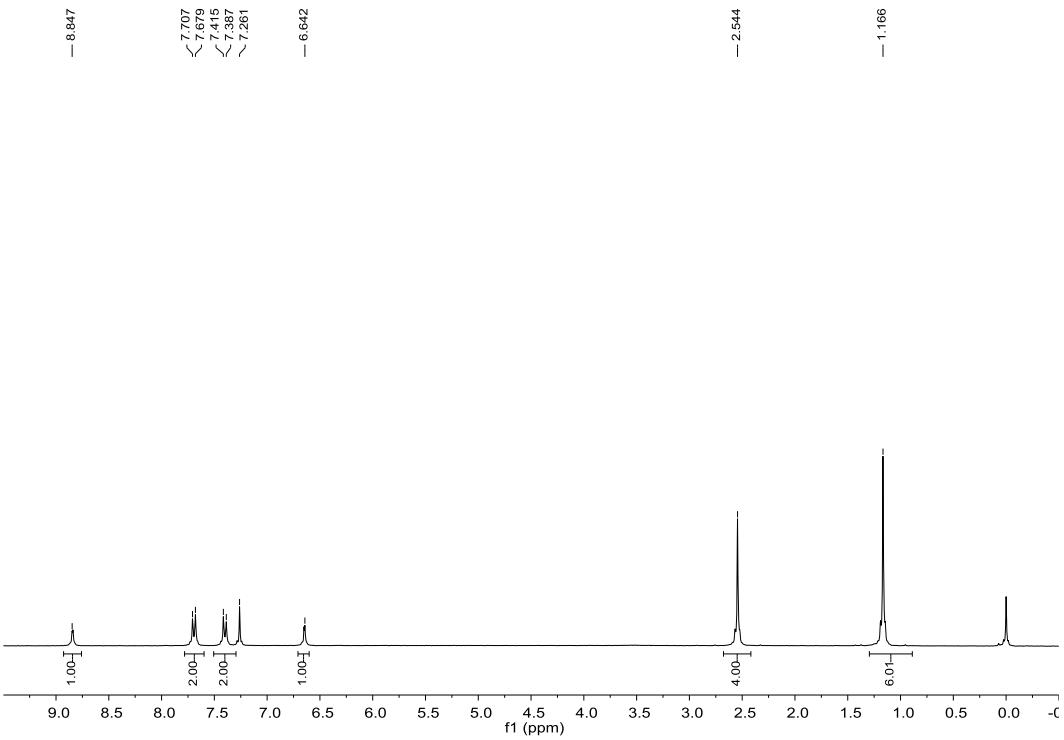
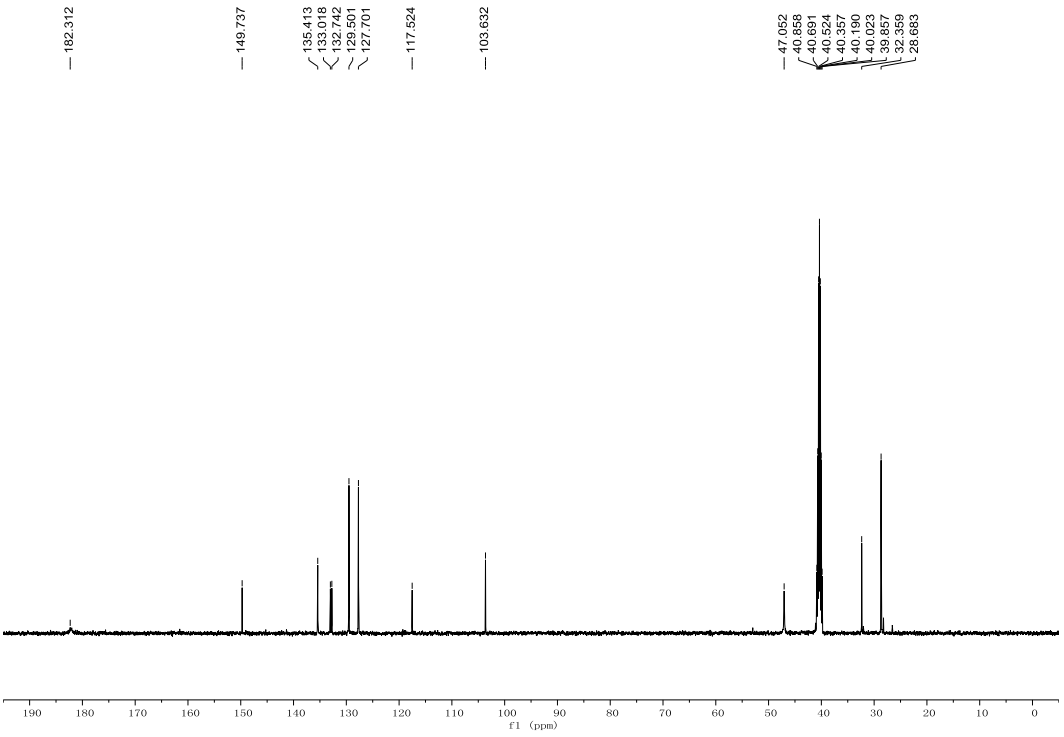


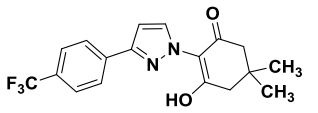
^{1}H NMR and ^{13}C NMR Spectra of Compound 3da

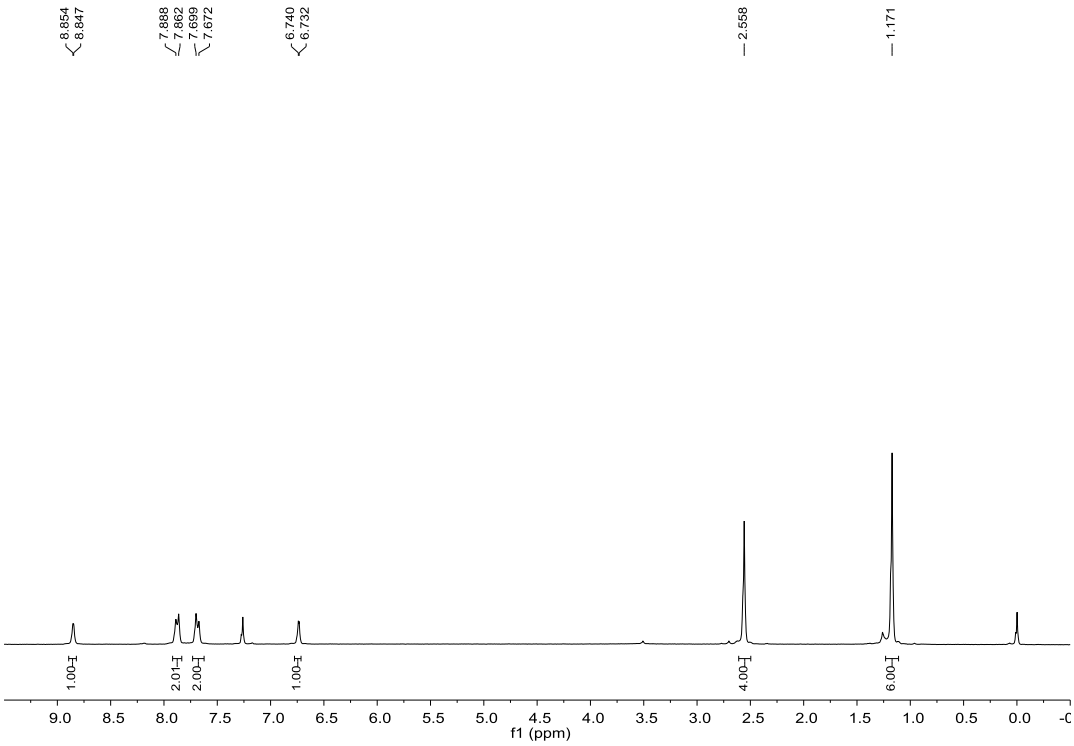


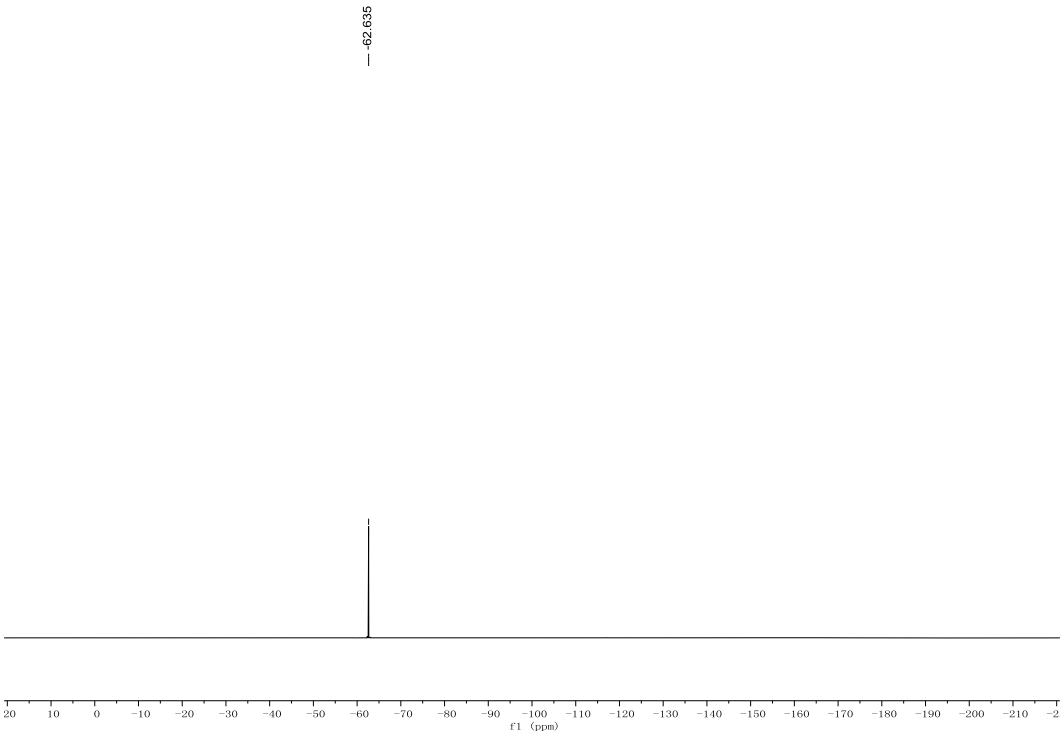


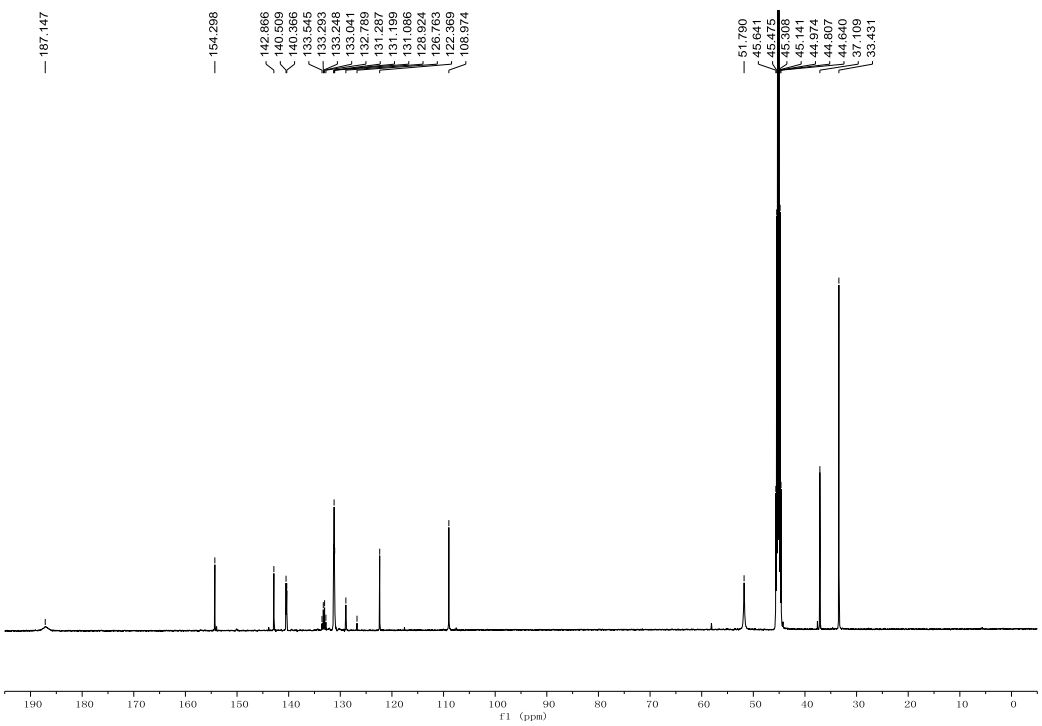


^{1}H NMR, ^{19}F NMR and ^{13}C NMR Spectra of Compound 3ea





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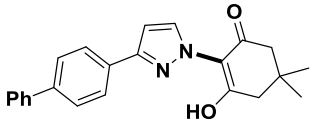
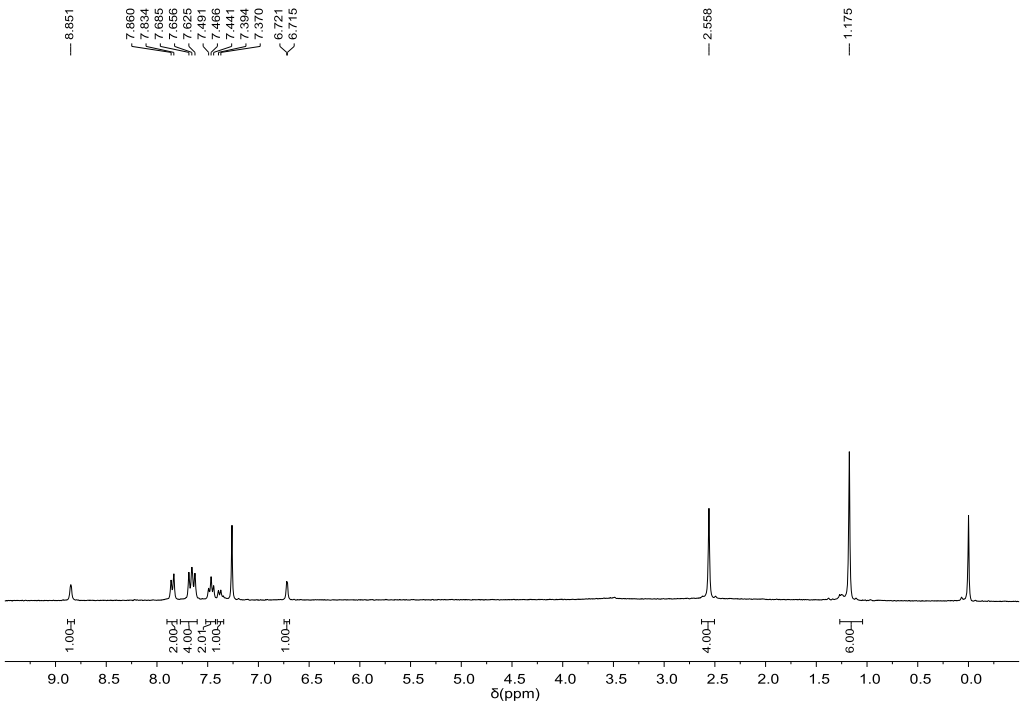
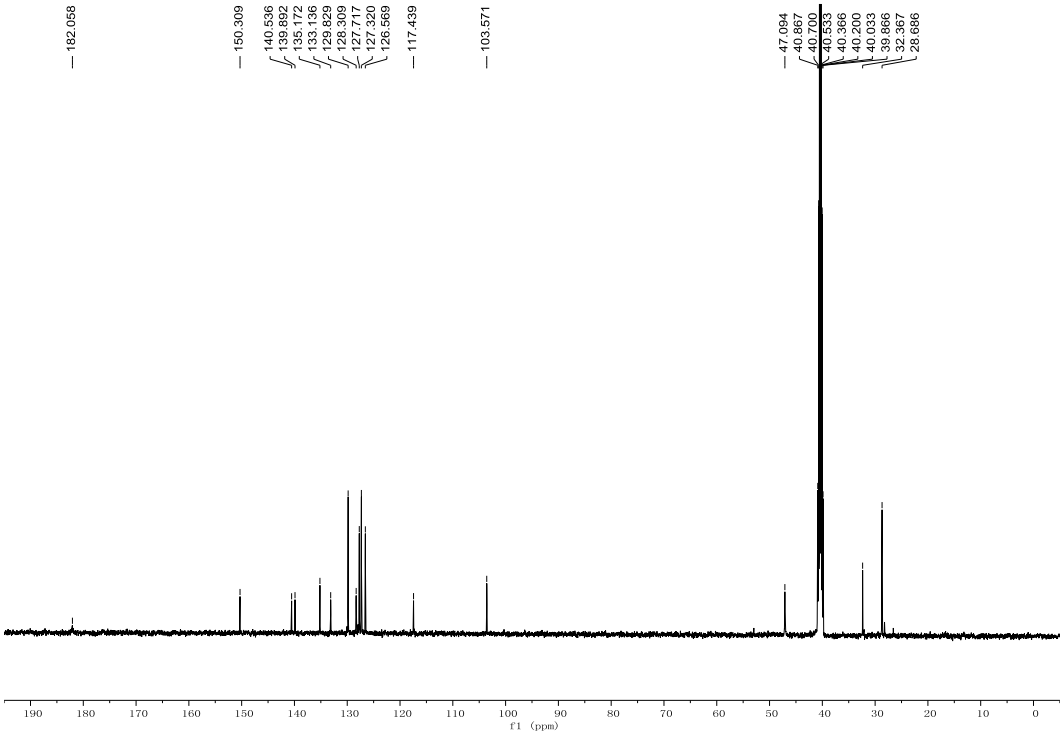
f1 (ppm)

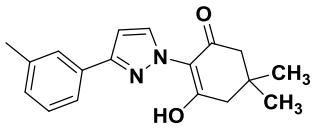
^{1}H NMR and ^{13}C NMR Spectra of Compound 3fa

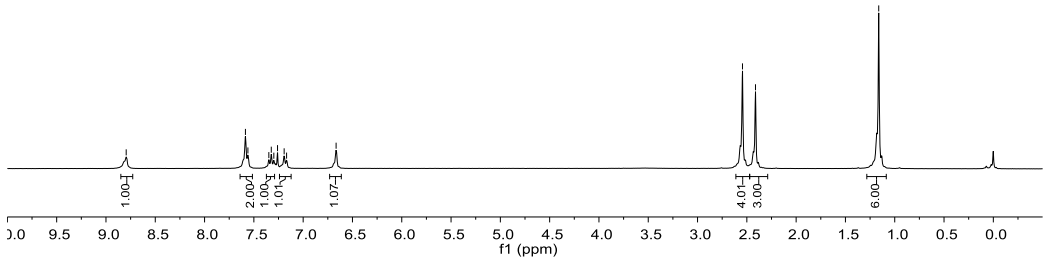


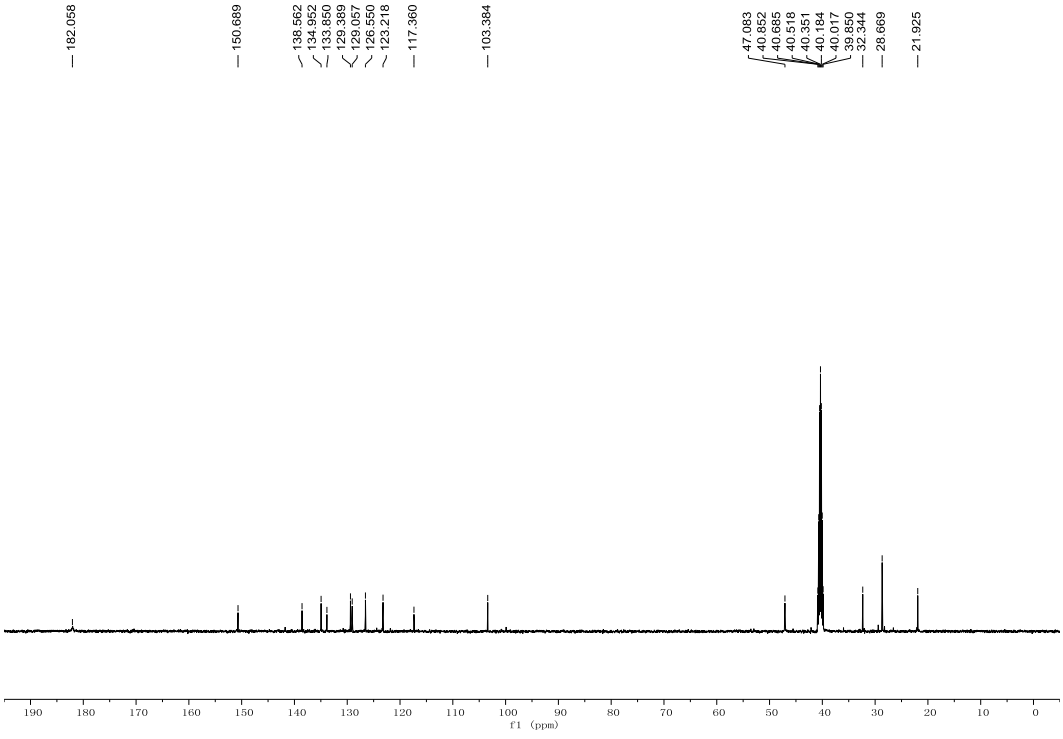




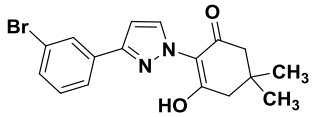
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ga

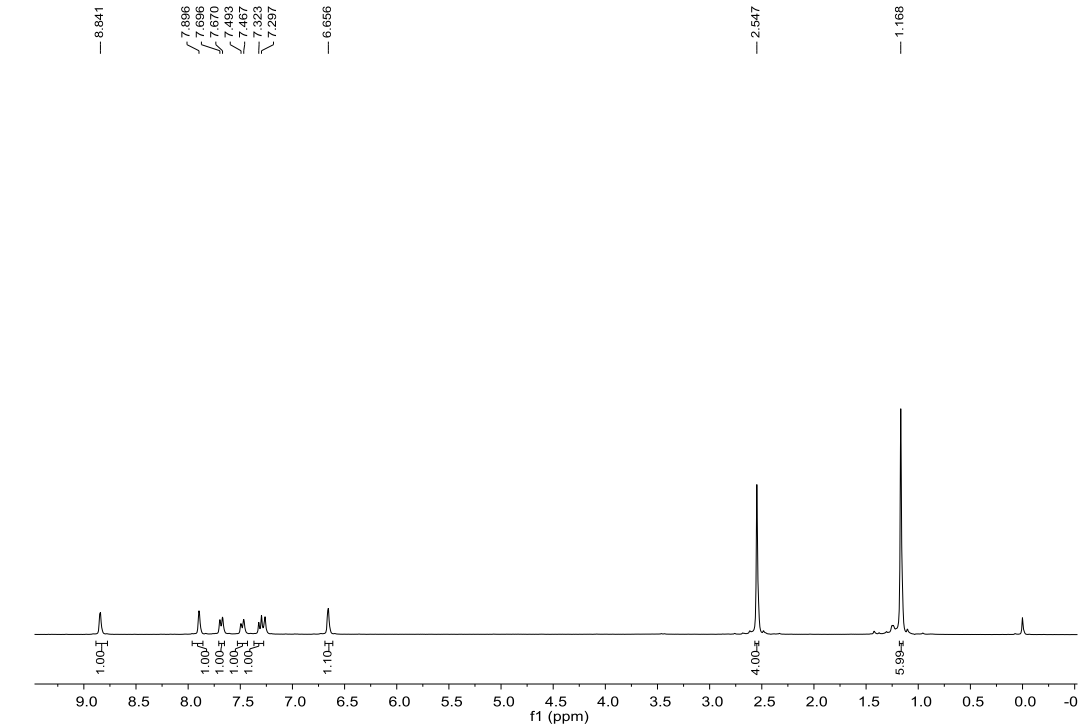


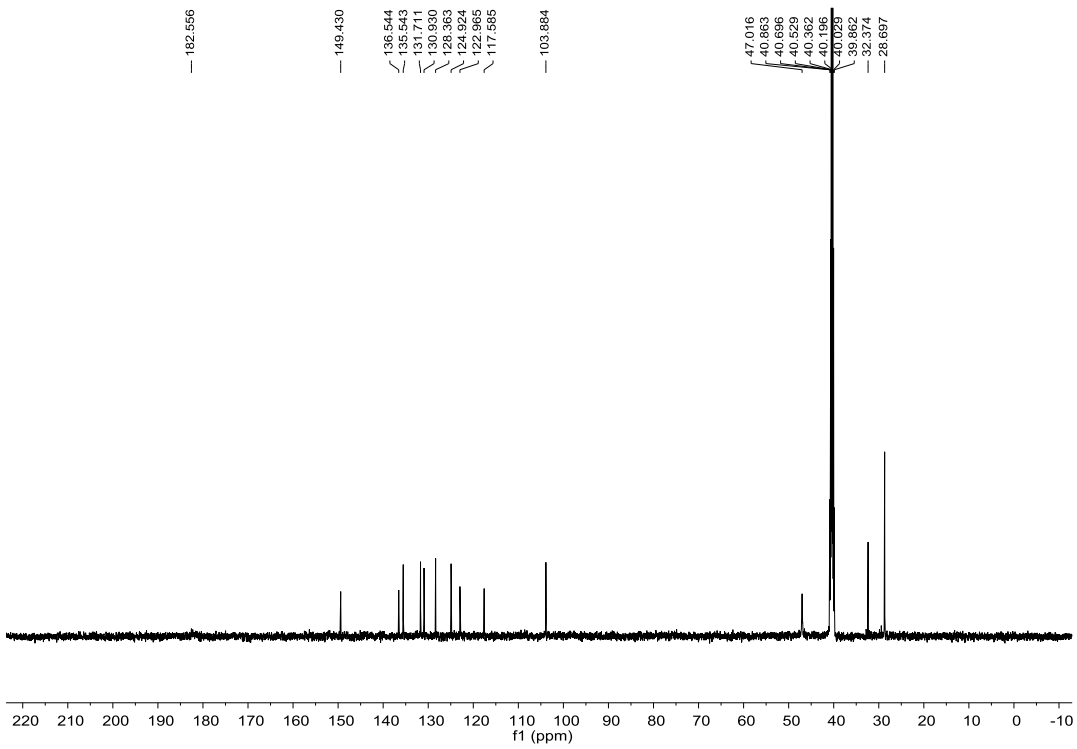




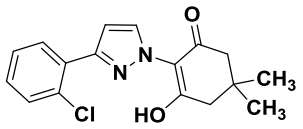
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ha

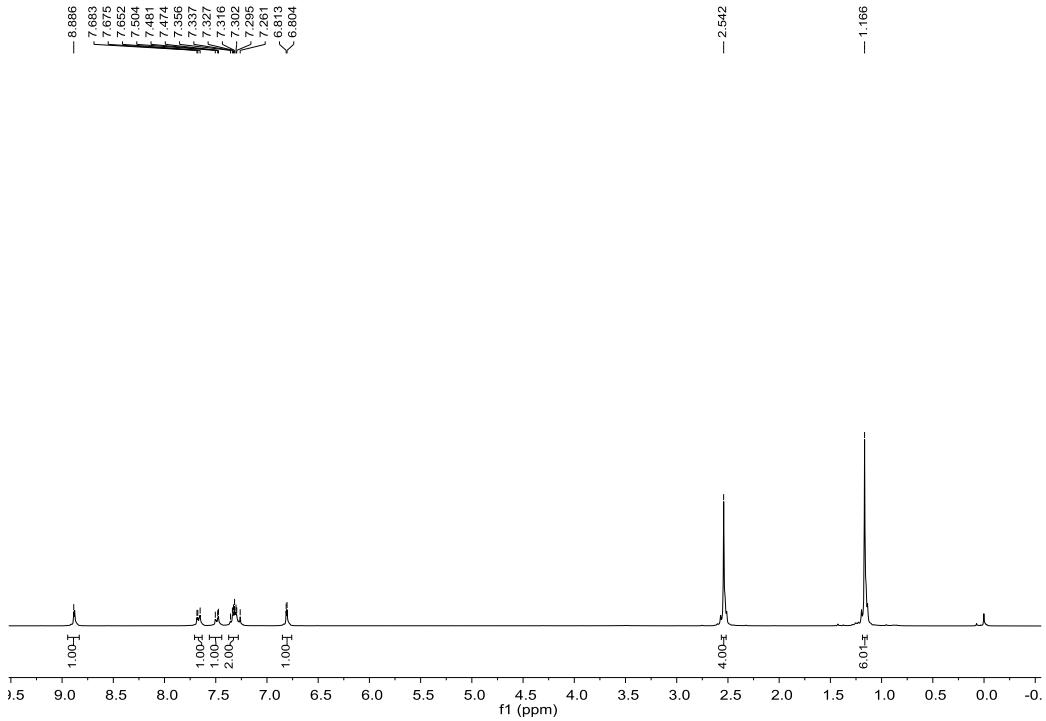


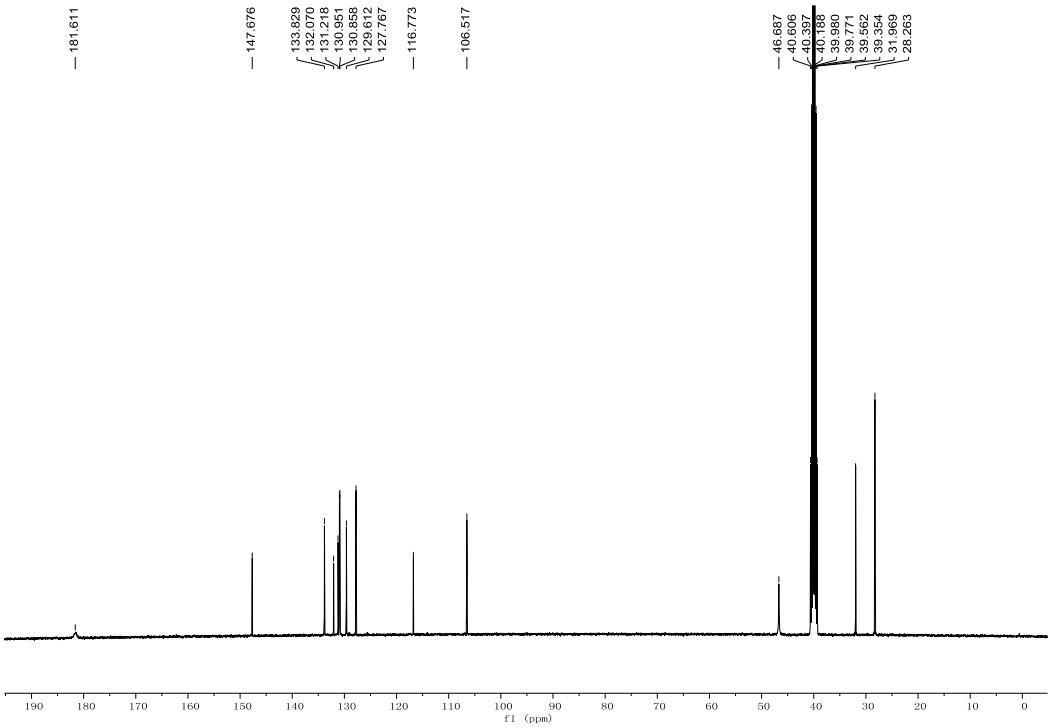




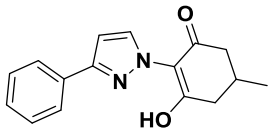
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ia

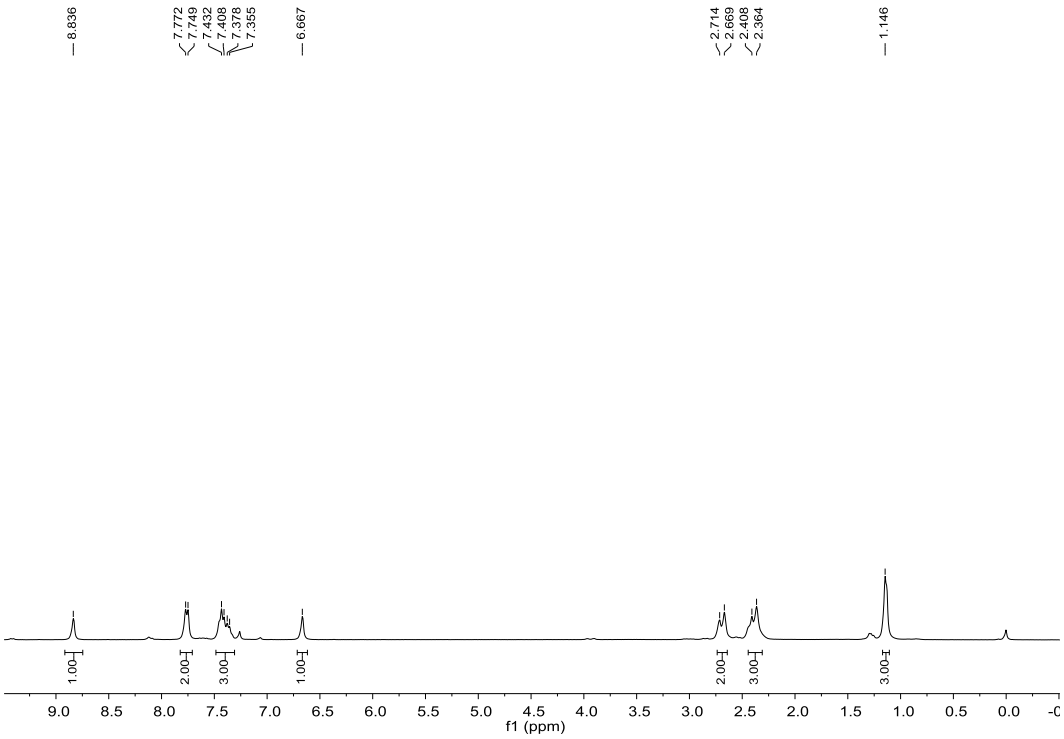


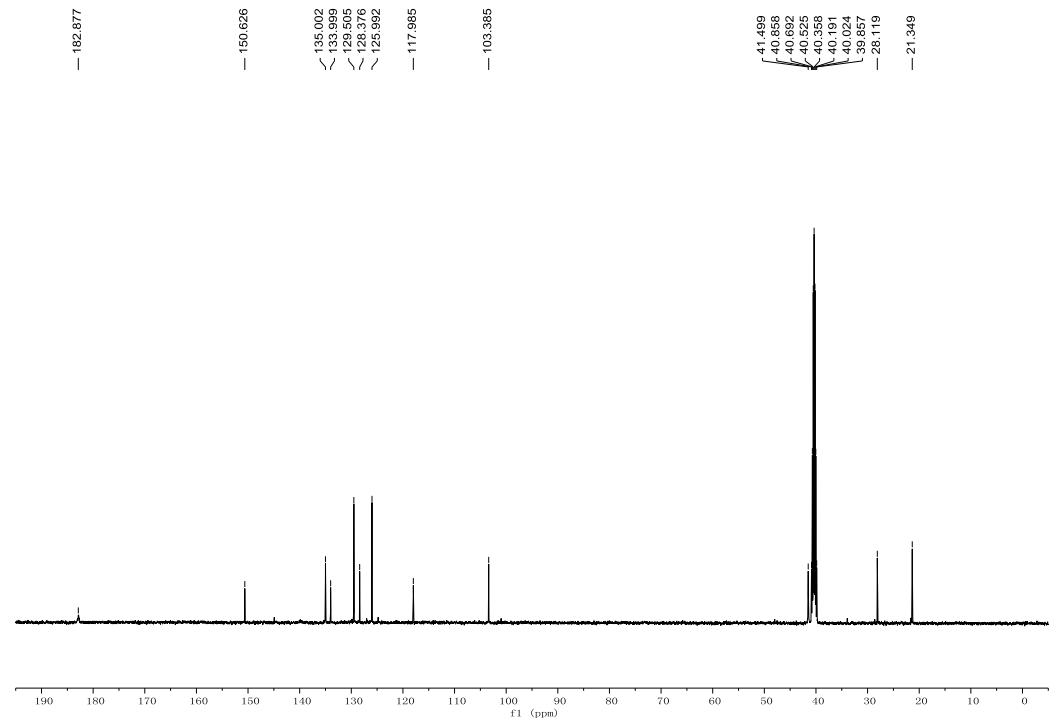




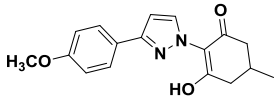
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ab



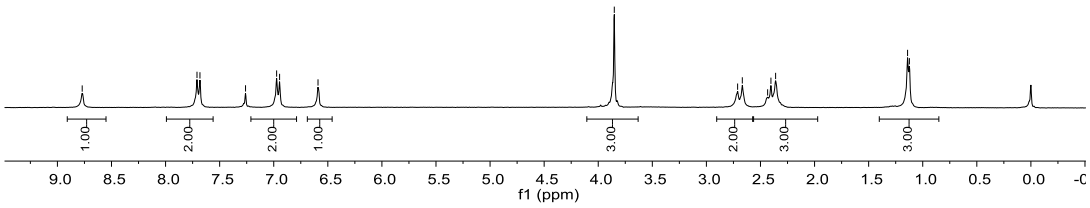




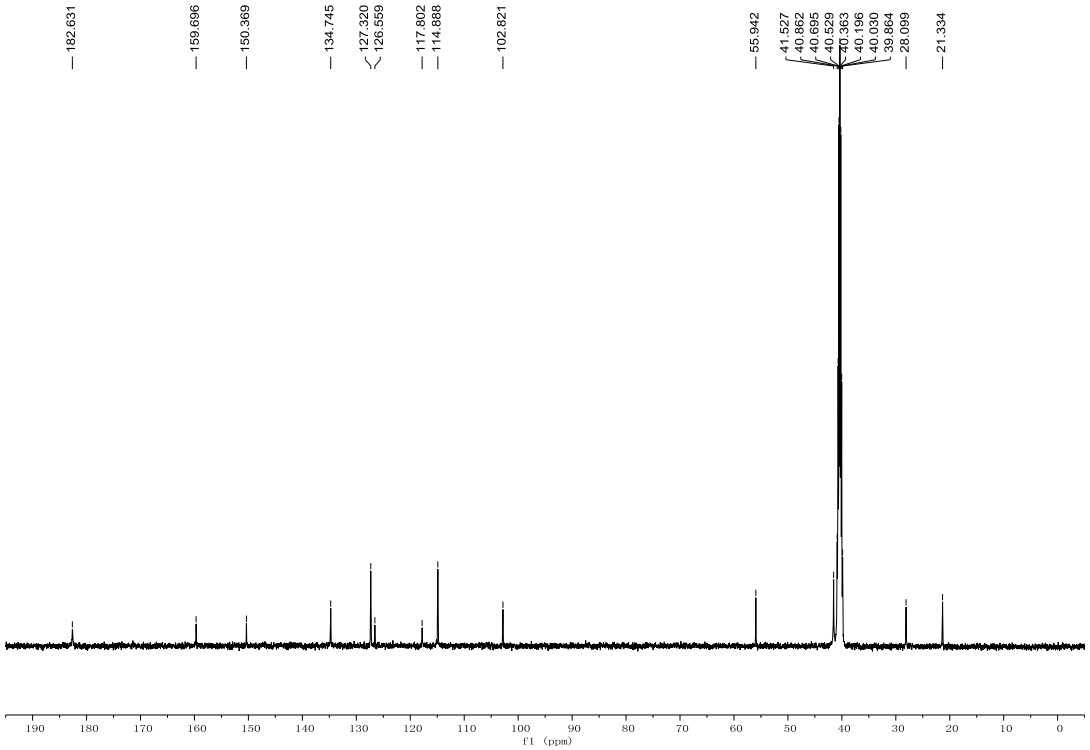
^{1}H NMR and ^{13}C NMR Spectra of Compound 3cb



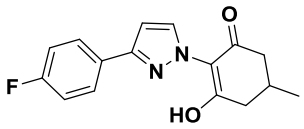
8.771
7.710
7.682
7.261
6.973
6.946
6.591
3.852
2.711
2.668
2.434
2.403
2.360
1.140
1.123

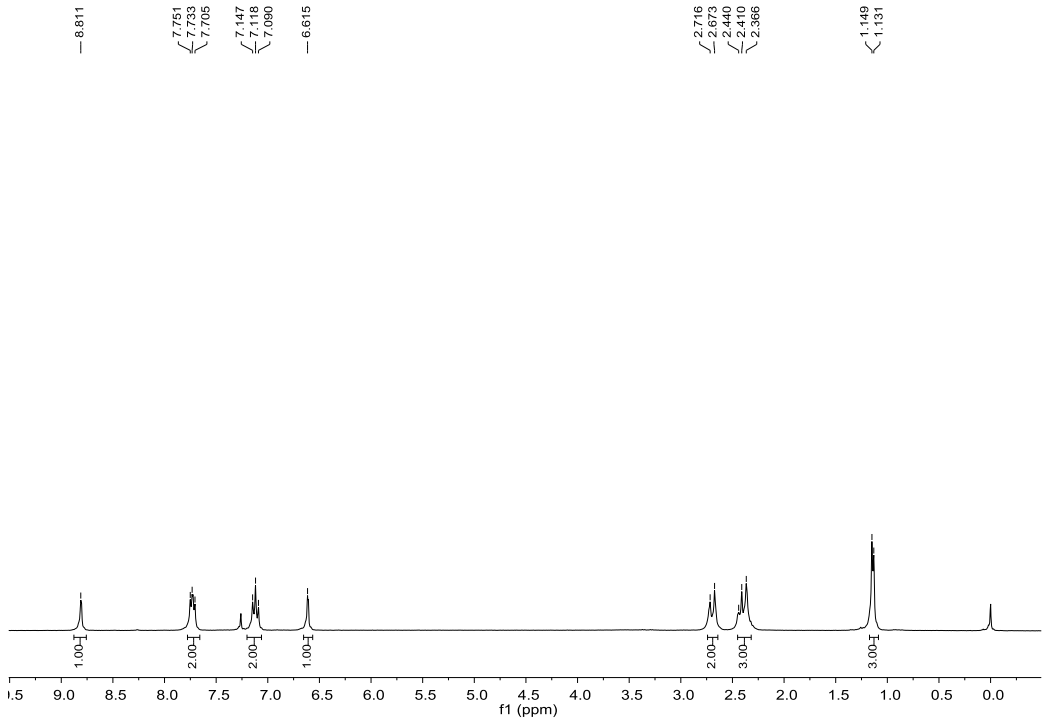


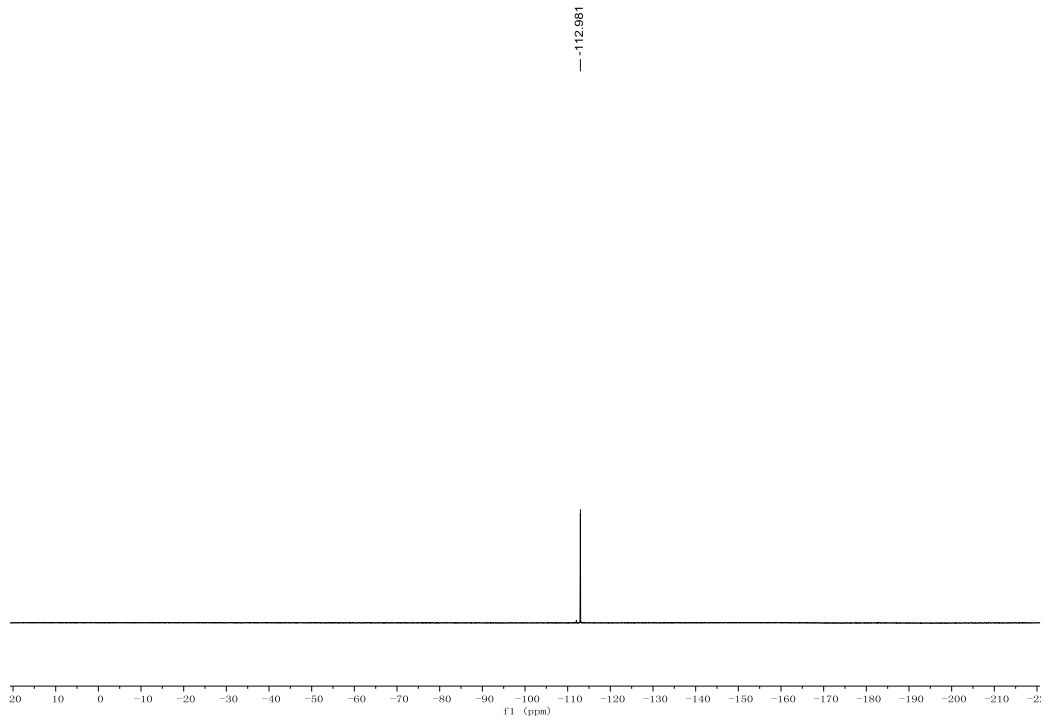
182.631
159.696
150.369
134.745
127.320
126.559
117.802
114.888
102.821
55.942
41.527
40.862
40.695
40.529
40.363
40.196
40.030
39.864
28.099
21.334

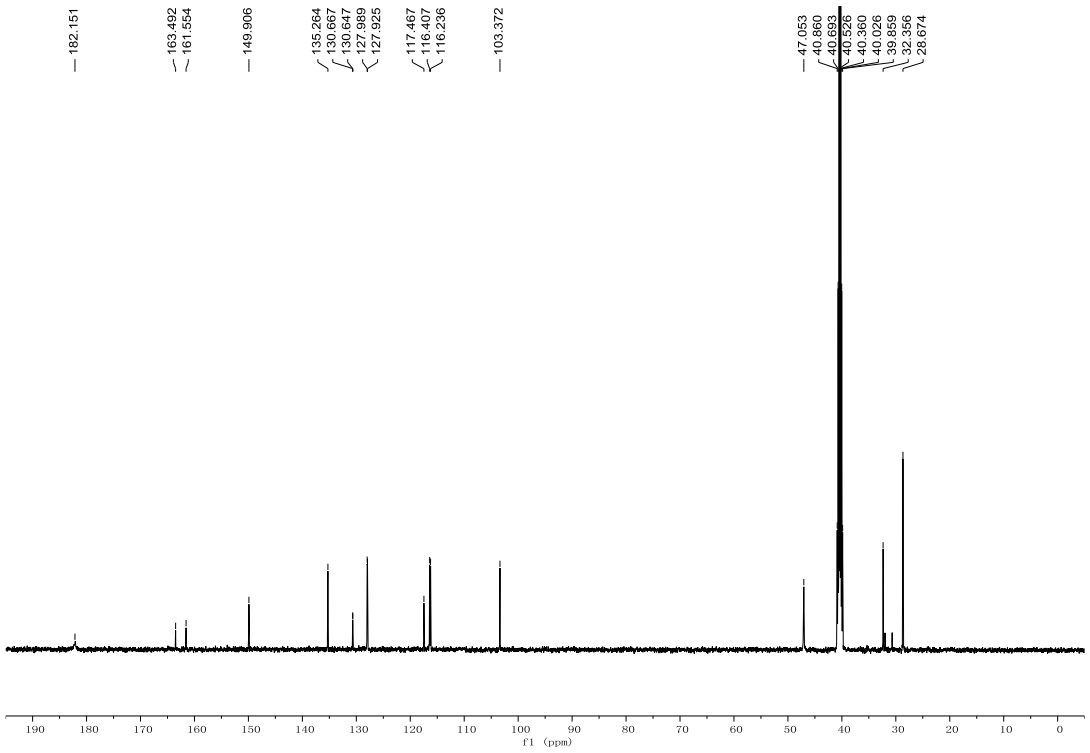


^{1}H NMR, ^{19}F NMR and ^{13}C NMR Spectra of Compound 3kb

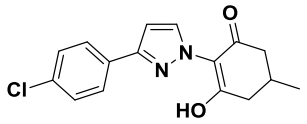


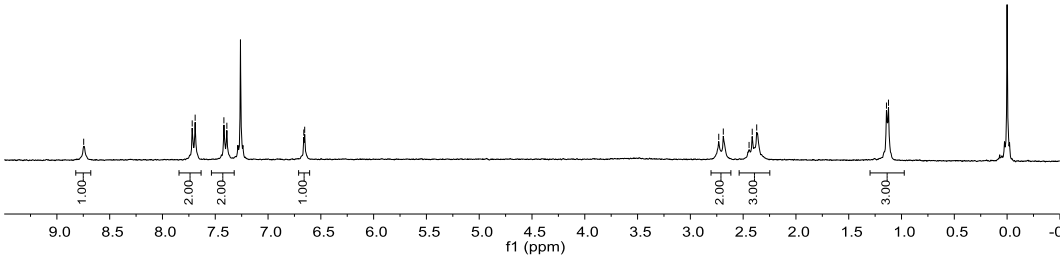


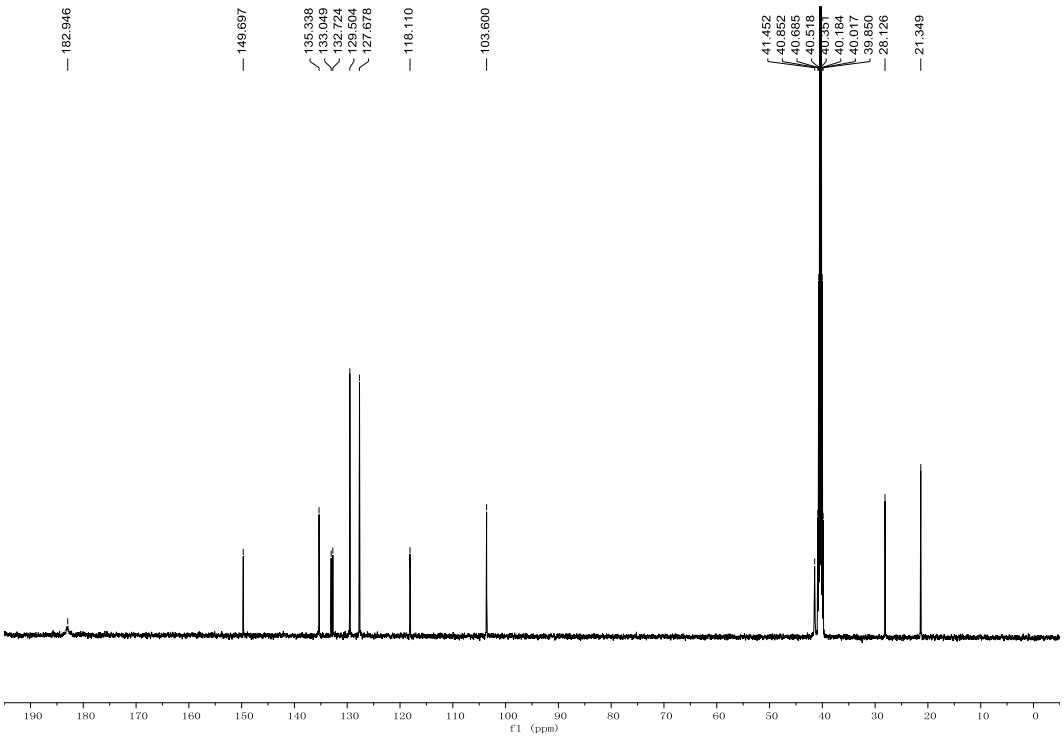



182.151
163.492
161.554
149.906
135.264
130.667
130.647
127.989
127.925
117.467
116.407
116.236
103.372
47.053
40.860
40.693
40.526
40.360
40.026
39.859
32.356
28.674
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

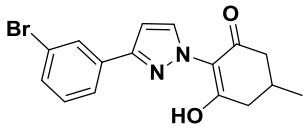
^{1}H NMR and ^{13}C NMR Spectra of Compound 3db

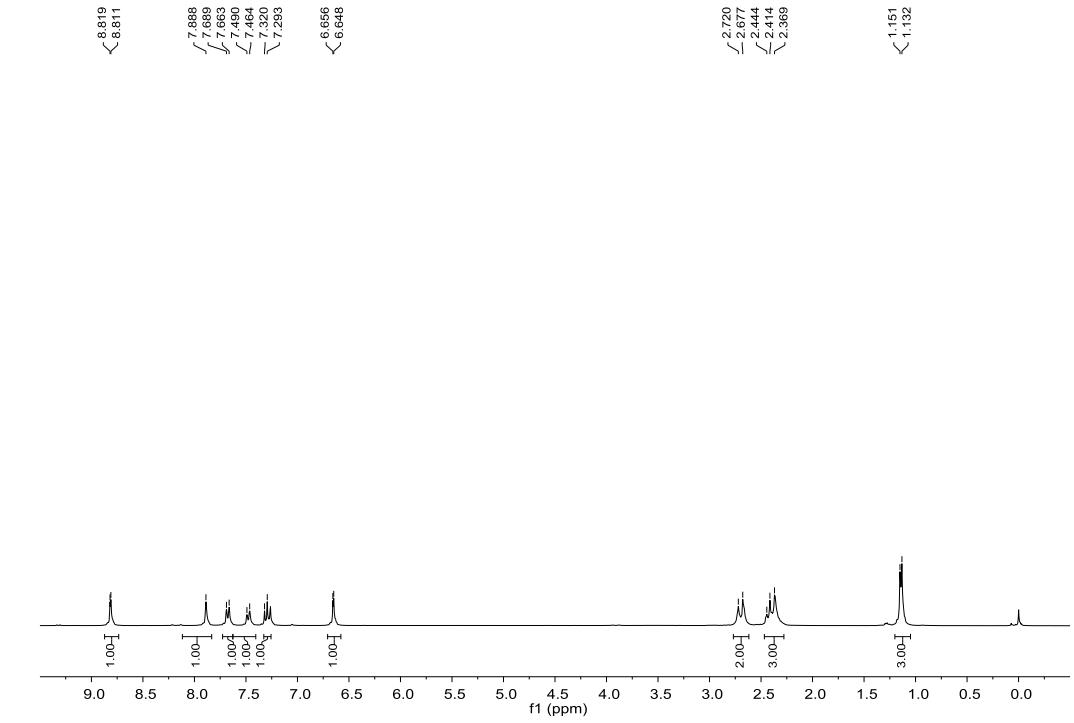


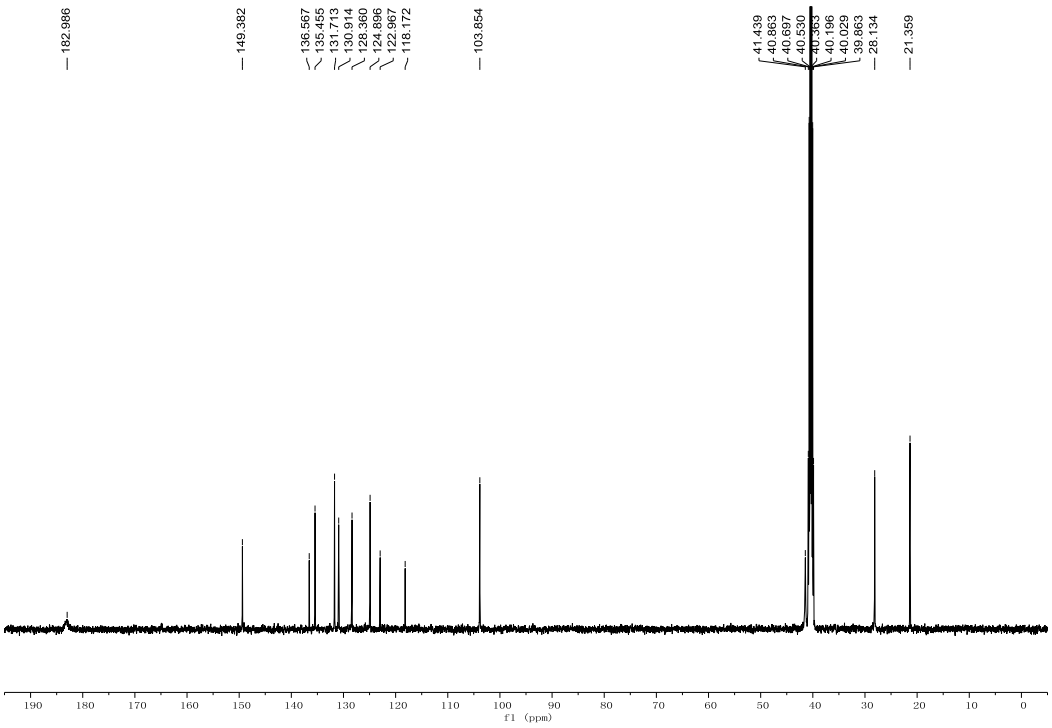




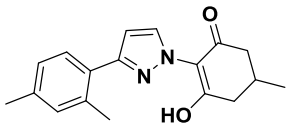
^{1}H NMR and ^{13}C NMR Spectra of Compound 3hb

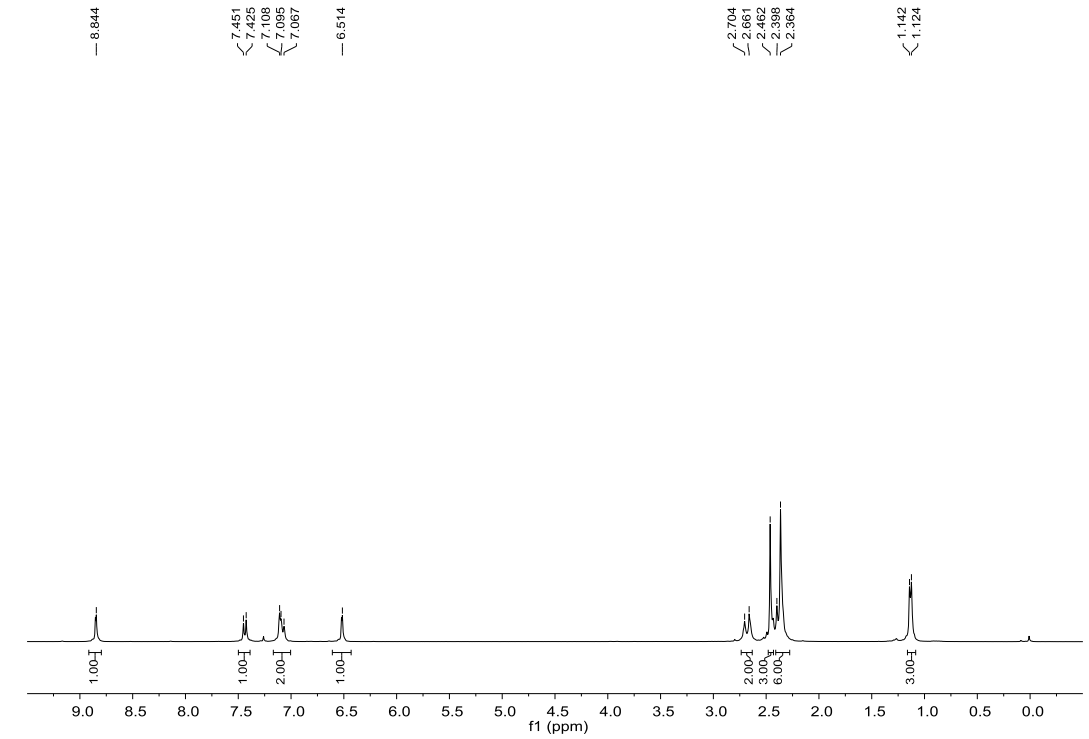


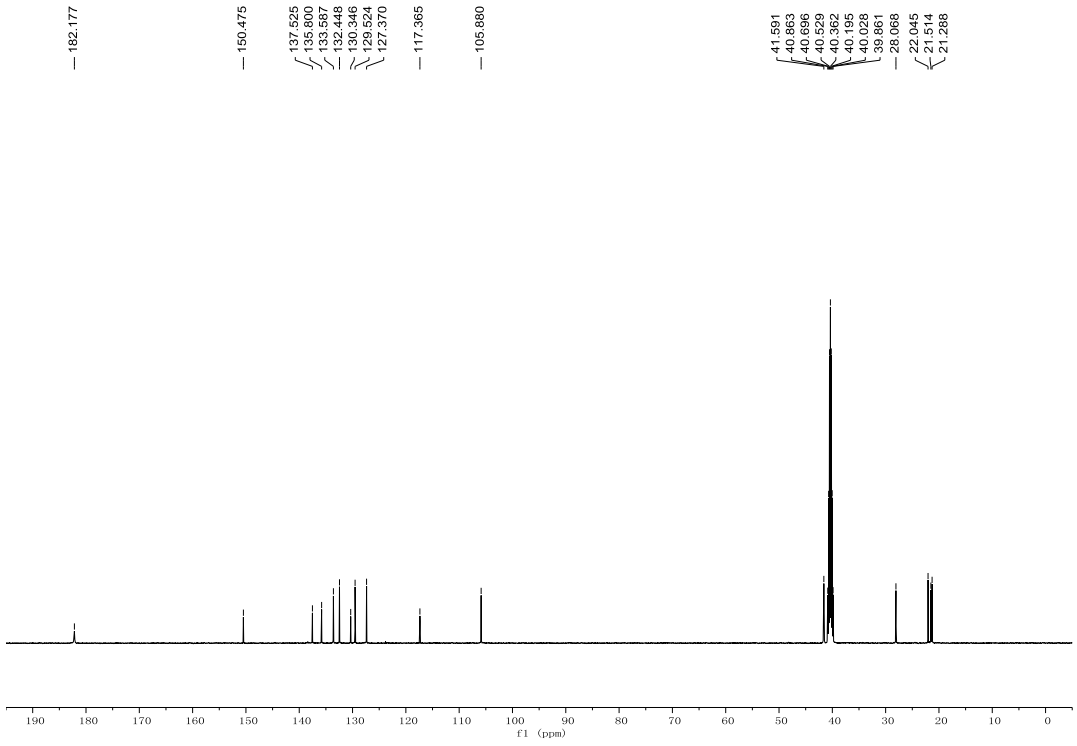




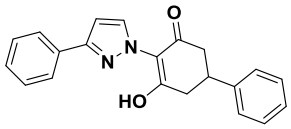
^{1}H NMR and ^{13}C NMR Spectra of Compound 3mb

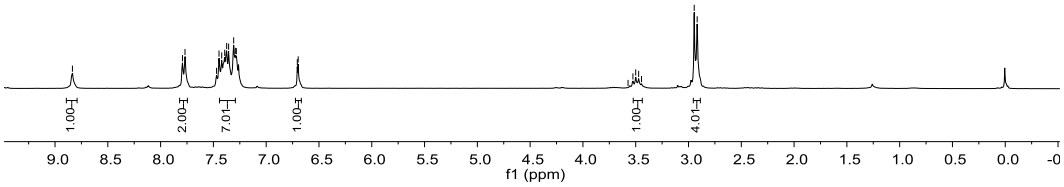


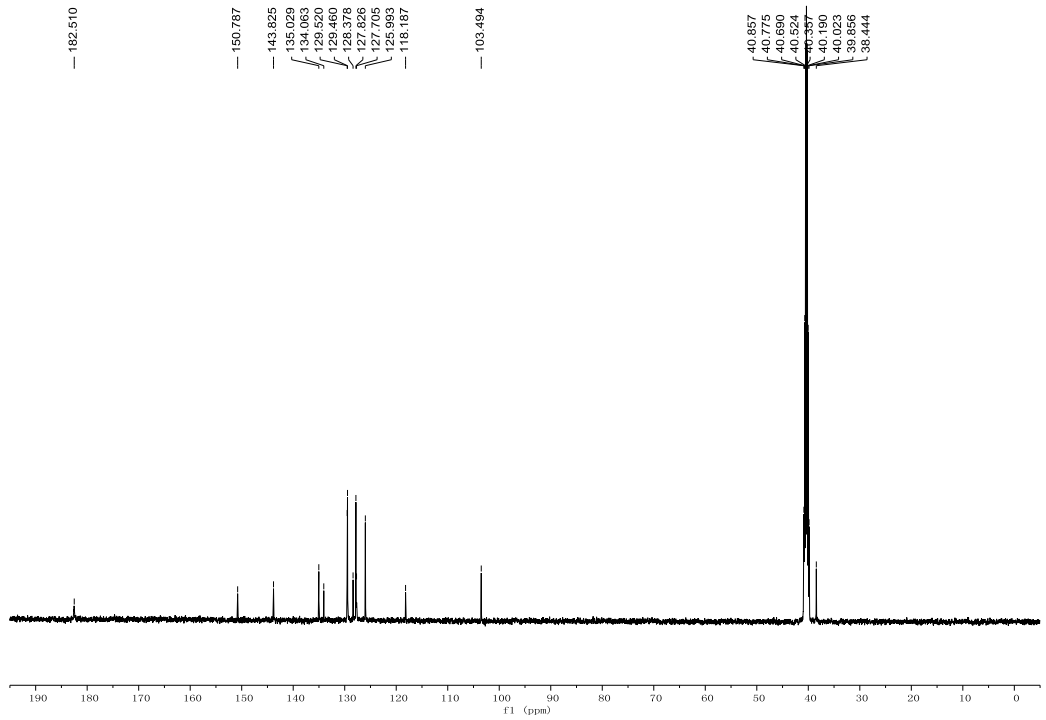




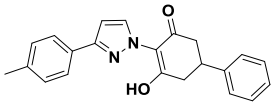
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ac

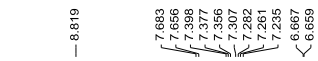


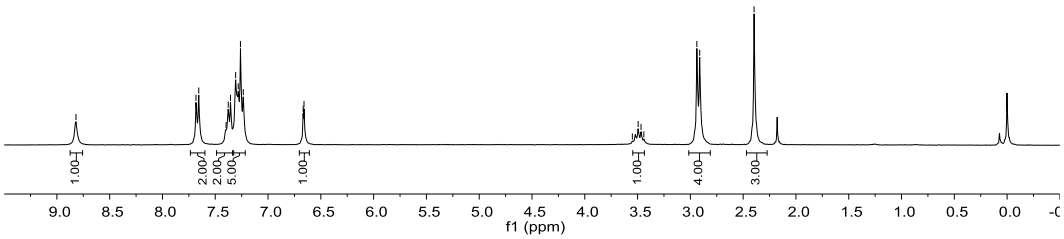


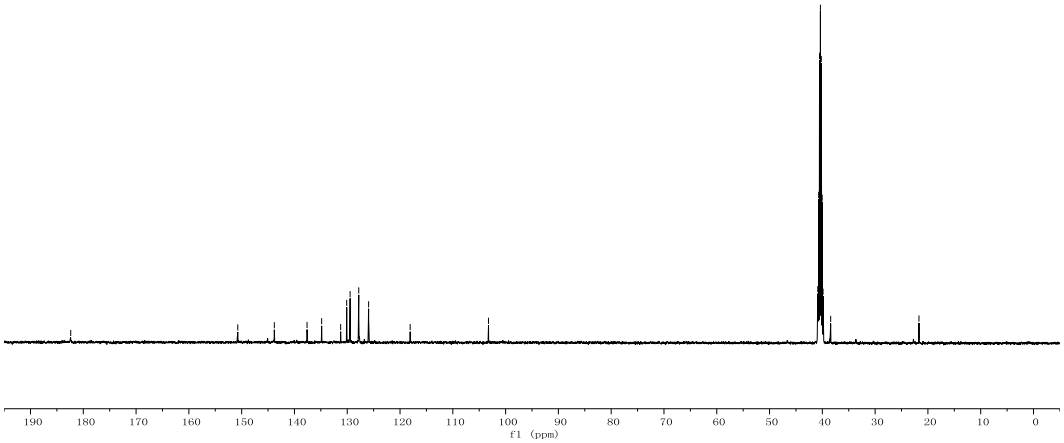


^{1}H NMR and ^{13}C NMR Spectra of Compound 3bc

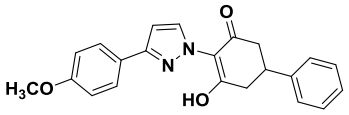


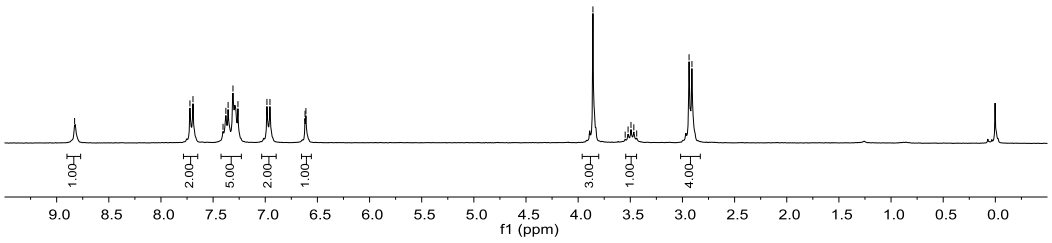


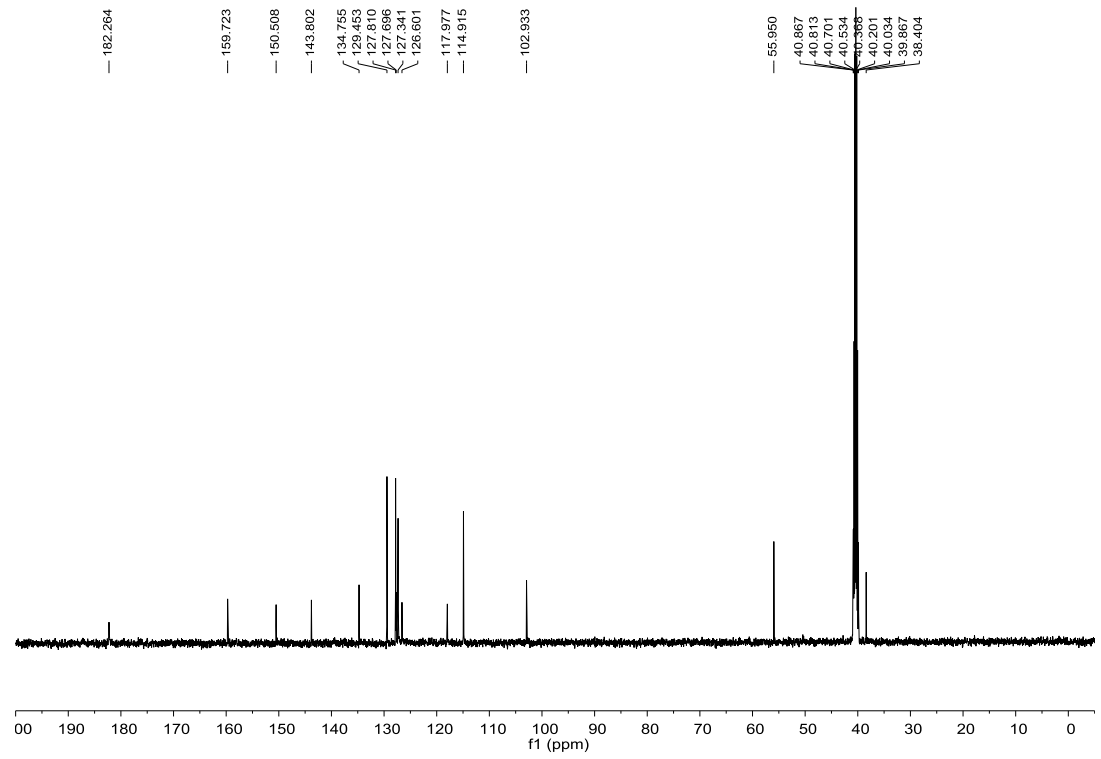




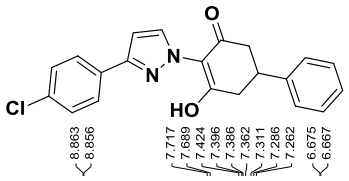
^{1}H NMR and ^{13}C NMR Spectra of Compound 3cc

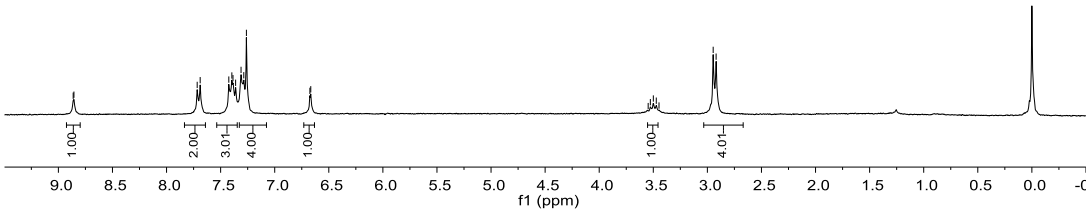


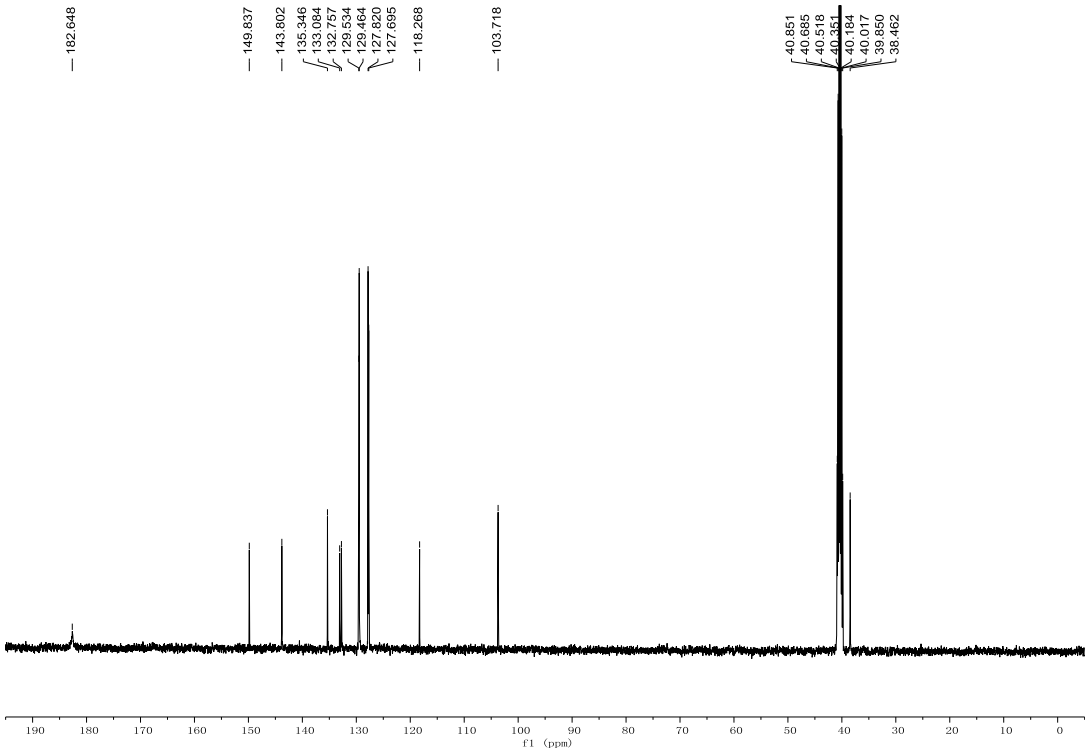




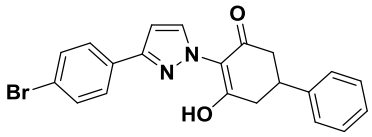
^{1}H NMR and ^{13}C NMR Spectra of Compound 3dc

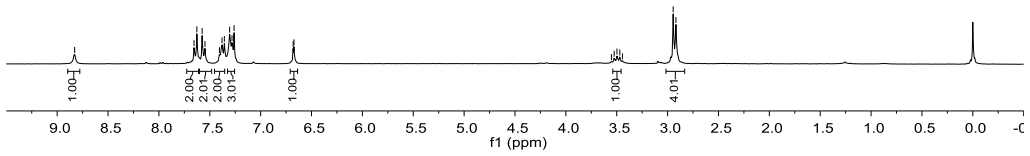


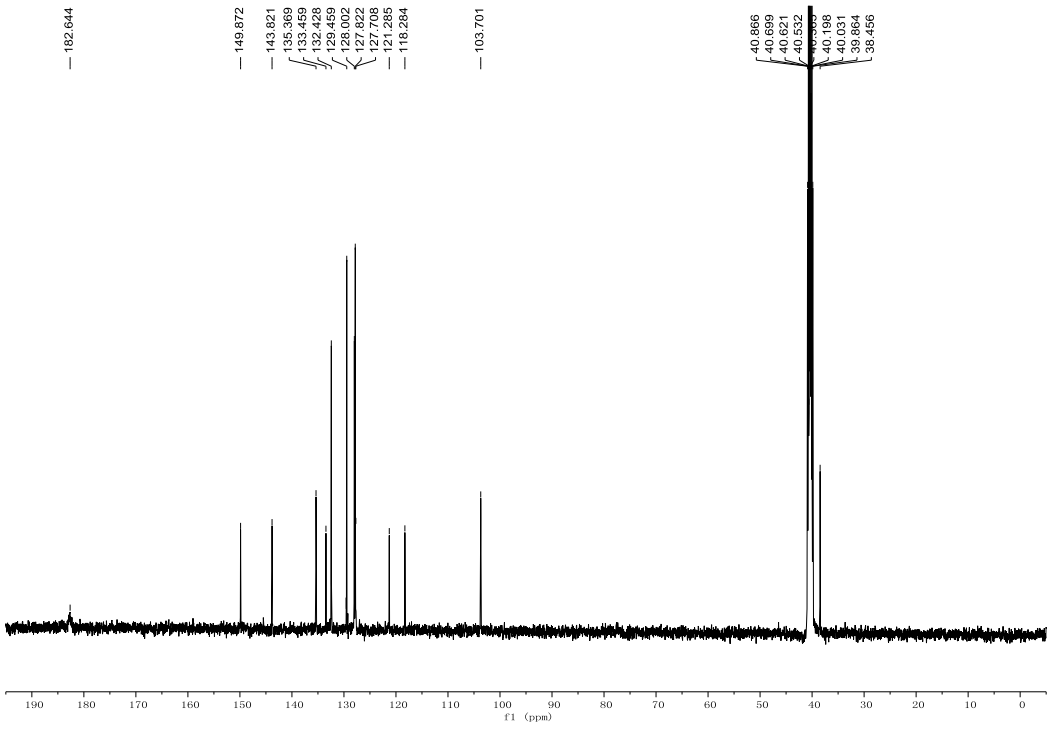




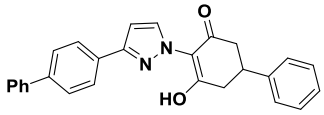
^{1}H NMR and ^{13}C NMR Spectra of Compound 3nc

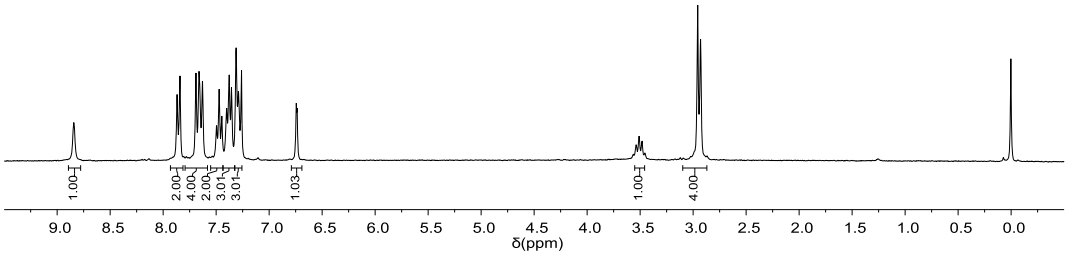


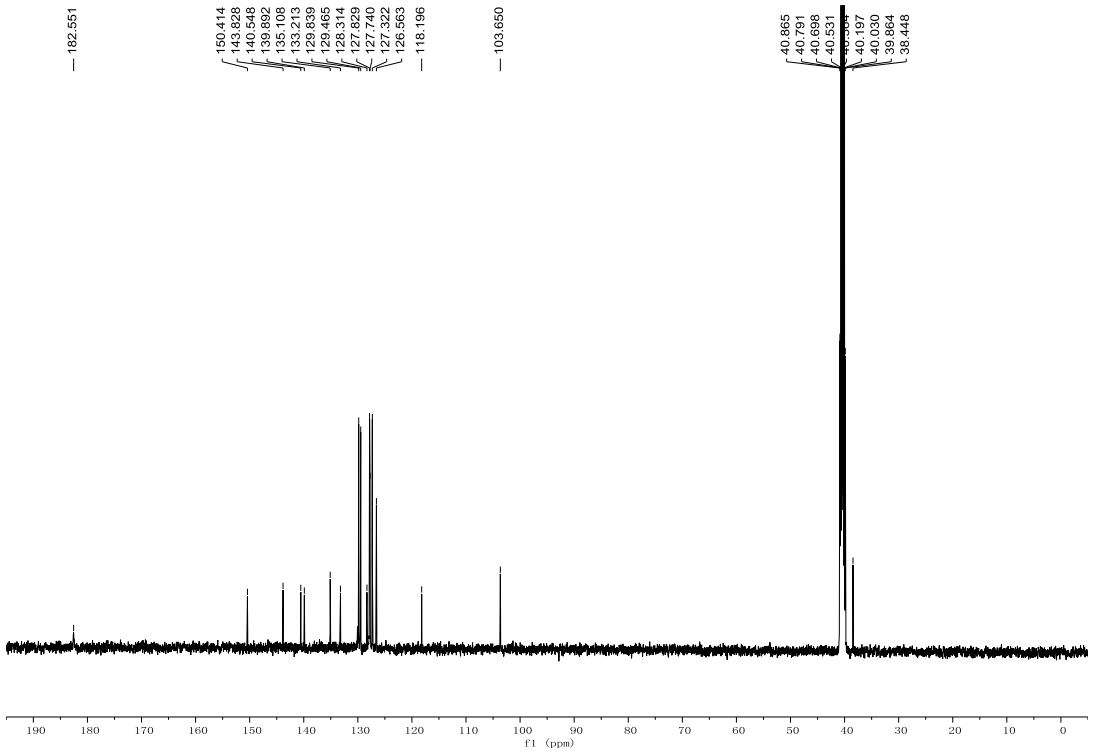




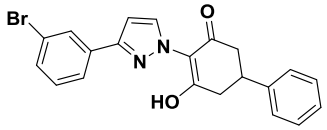
^{1}H NMR and ^{13}C NMR Spectra of Compound 3fc

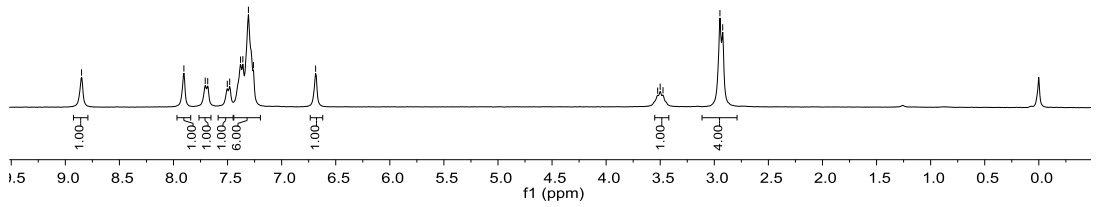


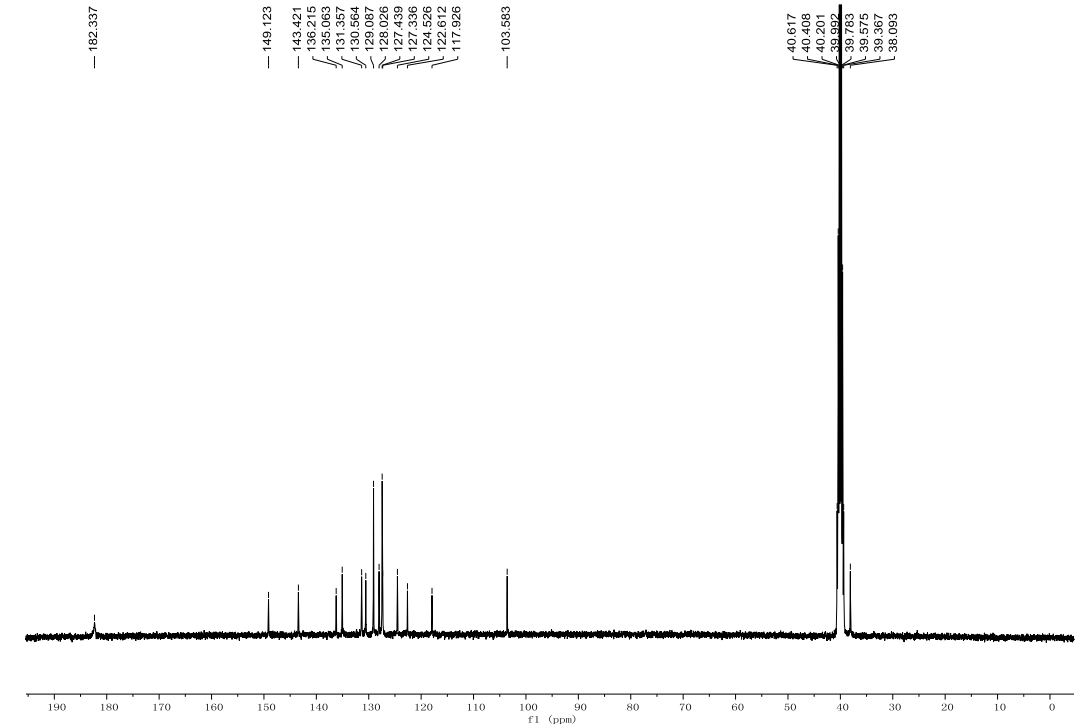




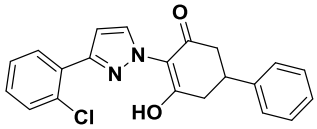
^{1}H NMR and ^{13}C NMR Spectra of Compound 3hc

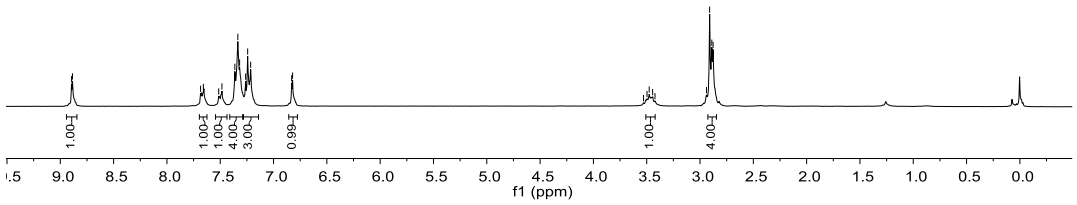


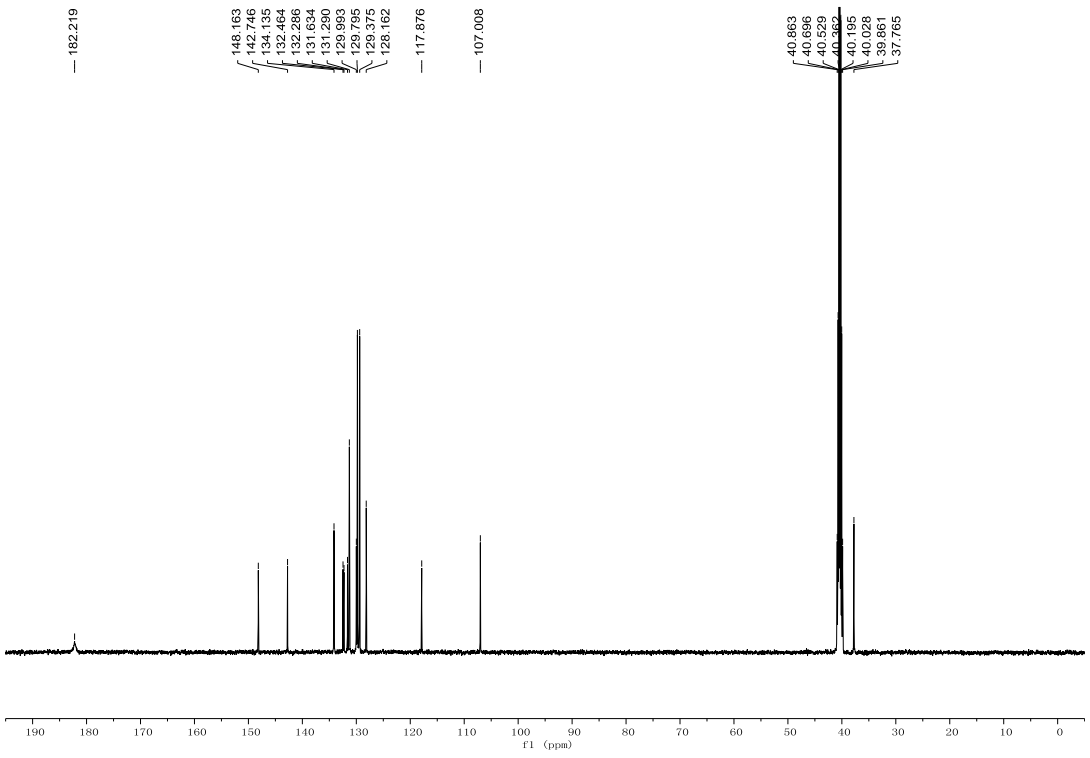




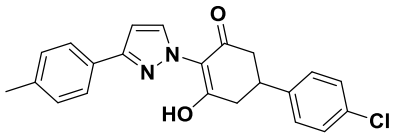
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ic

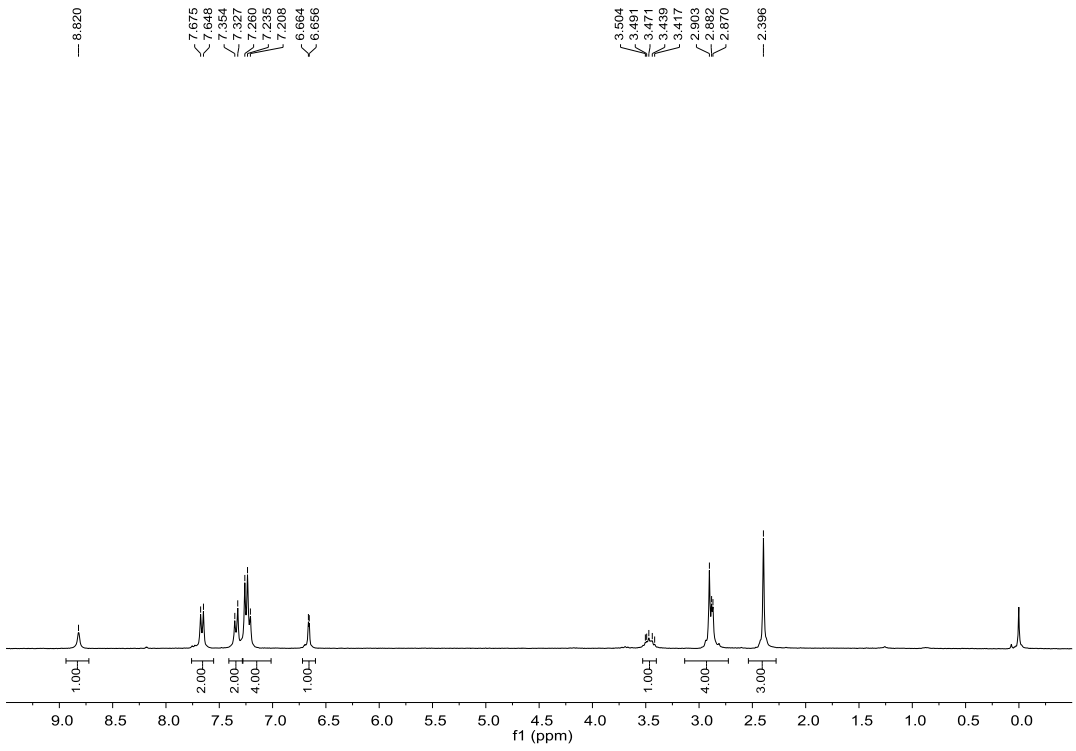


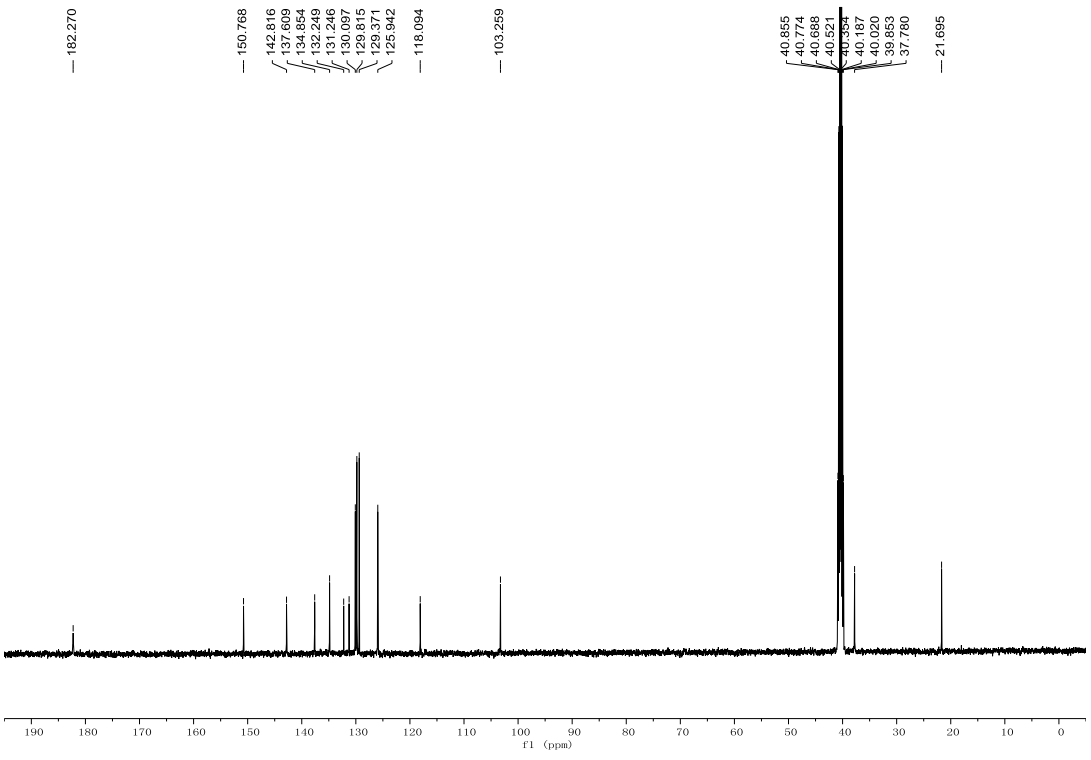




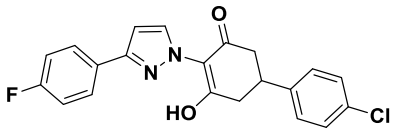
^{1}H NMR and ^{13}C NMR Spectra of Compound 3bd

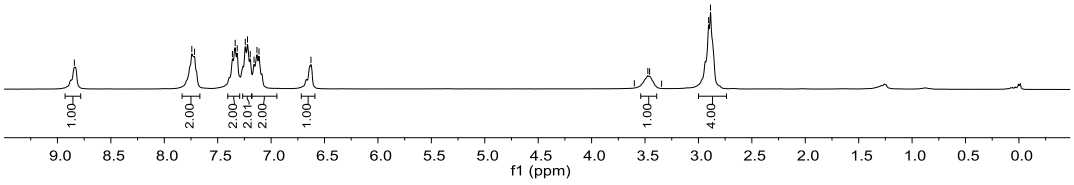


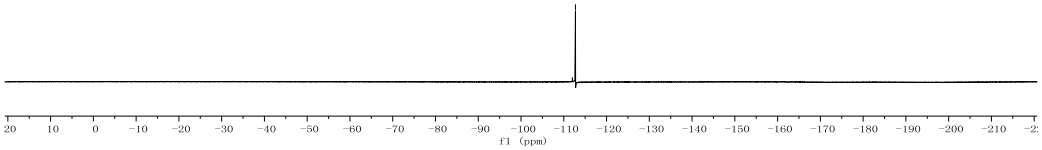


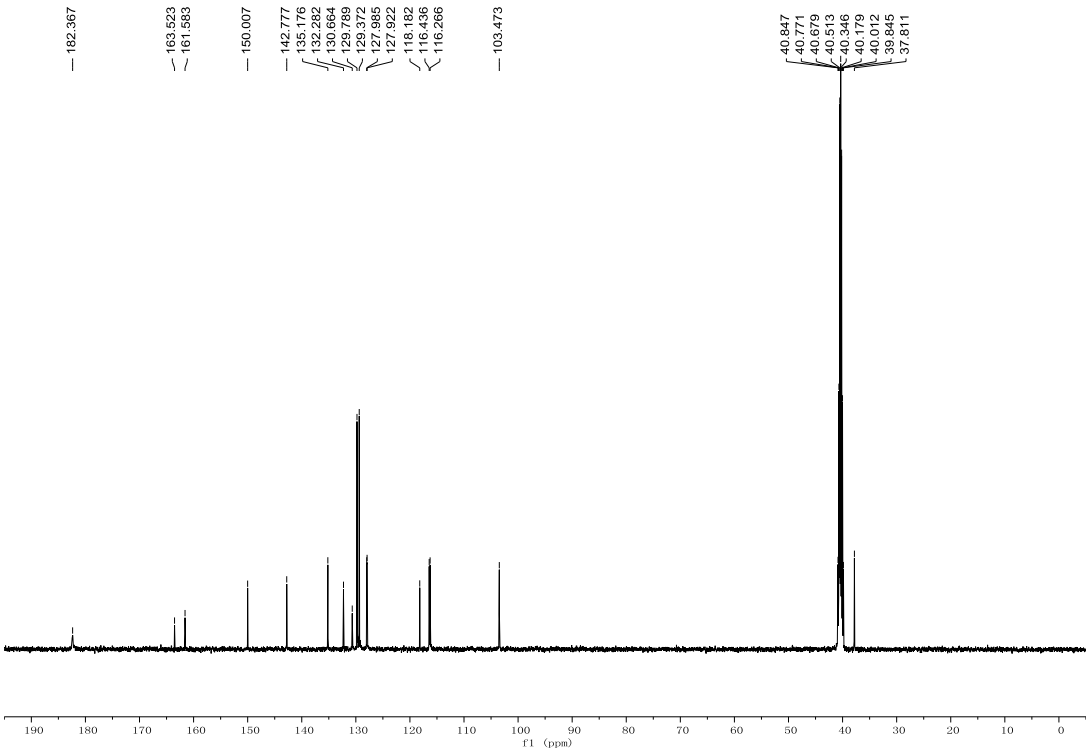


^{1}H NMR, ^{19}F NMR ^{13}C NMR Spectra of Compound 3kd





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129.372
127.985
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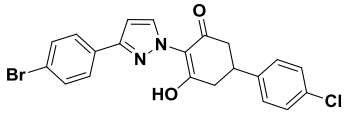
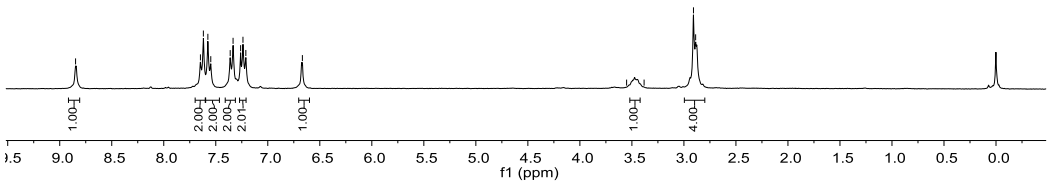
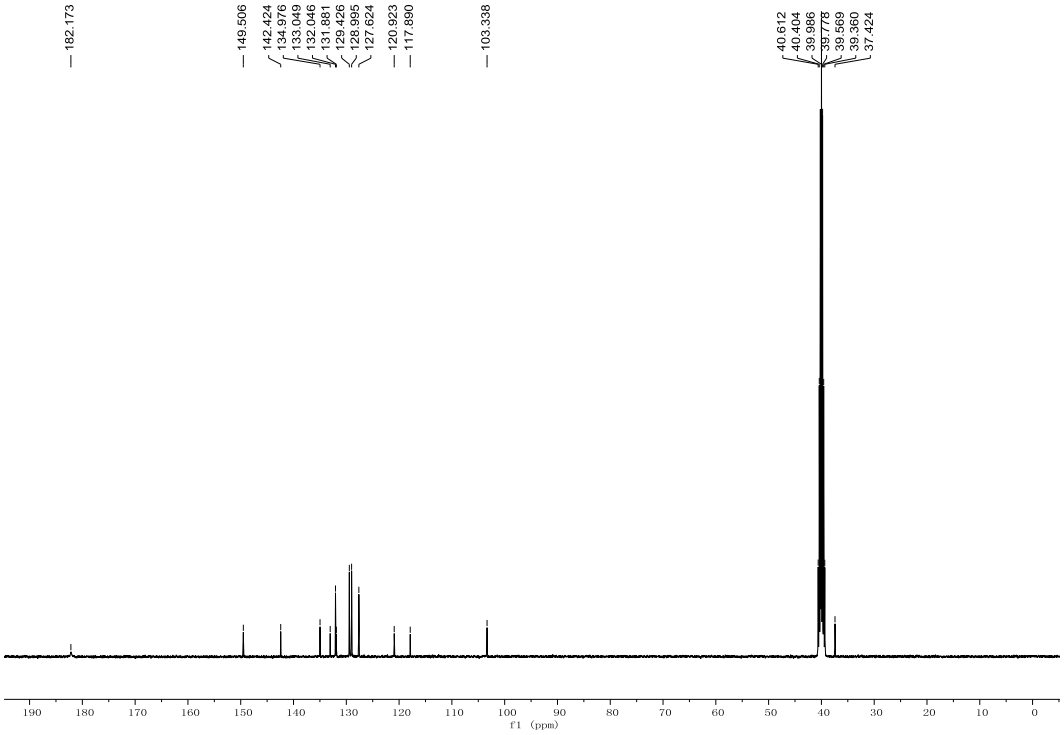
118.182
116.436
116.266
103.473
40.847
40.771
40.679
40.513
40.346
40.179
40.012
39.845
37.811
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

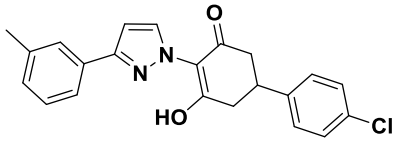
^{1}H NMR and ^{13}C NMR Spectra of Compound 3nd

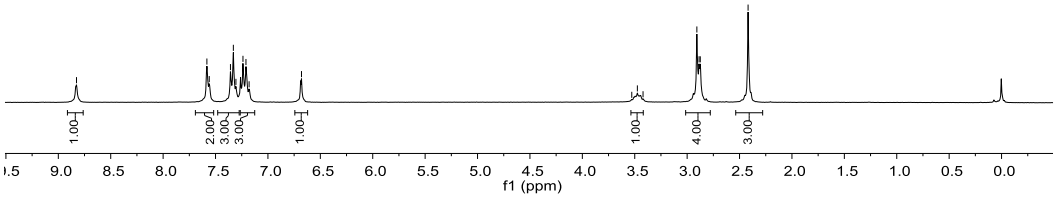


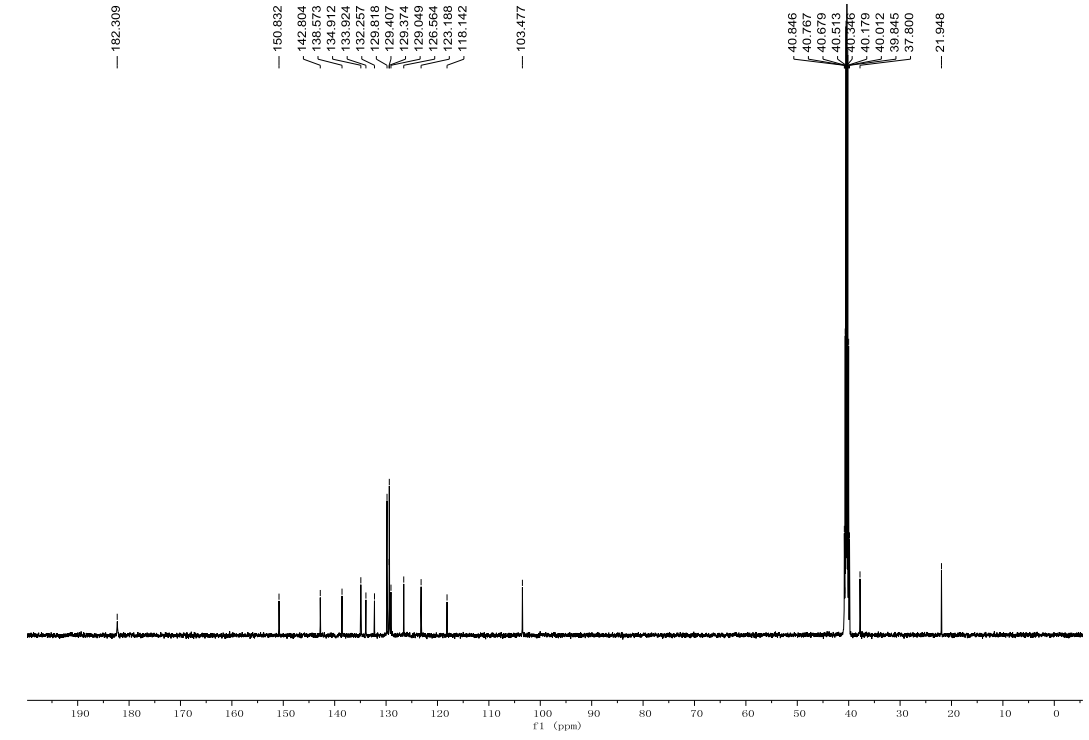




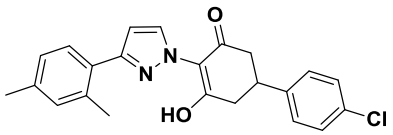
^{1}H NMR and ^{13}C NMR Spectra of Compound 3gd

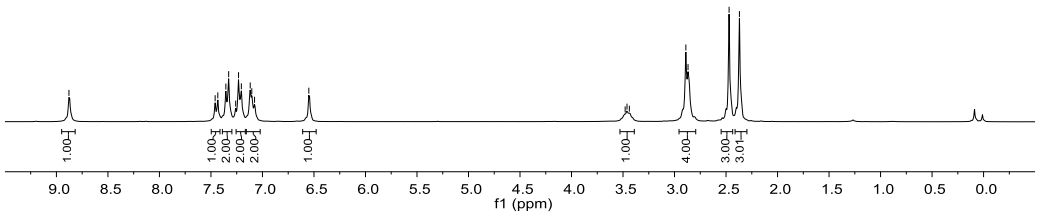


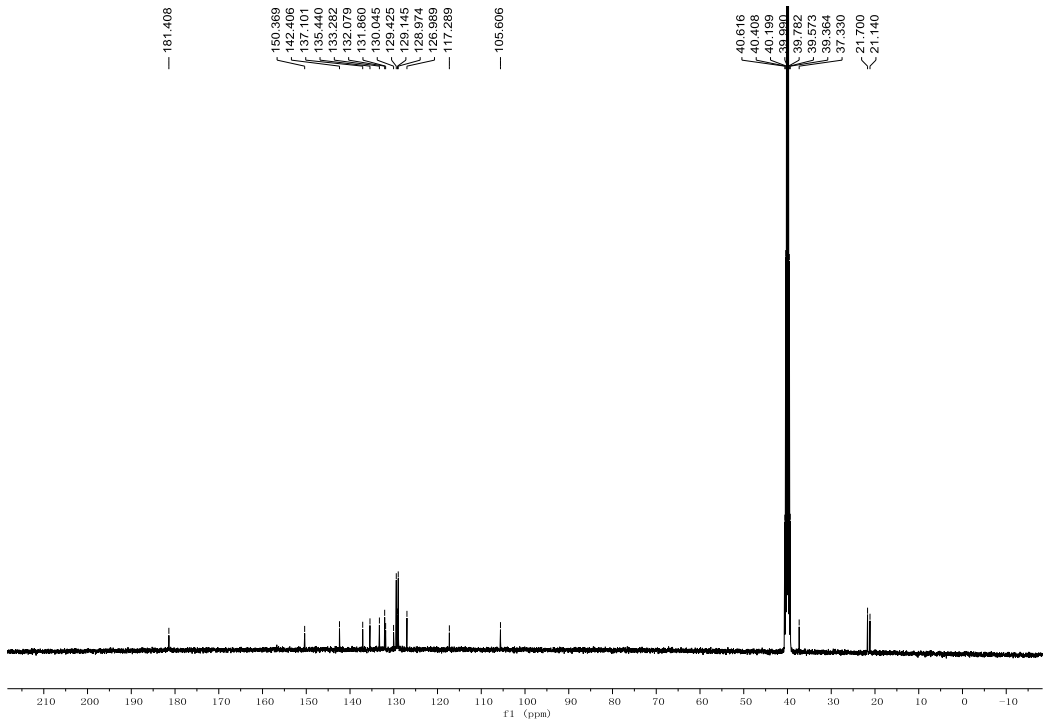




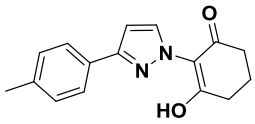
^{1}H NMR and ^{13}C NMR Spectra of Compound 3md

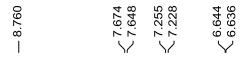


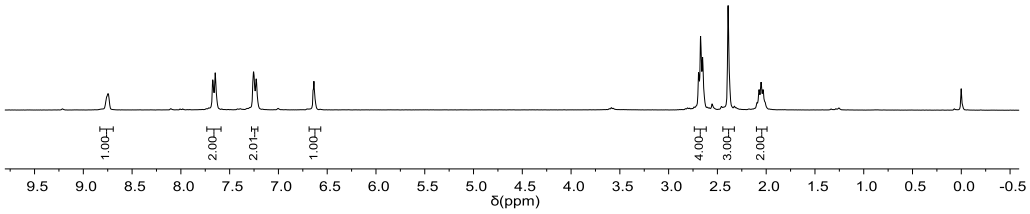


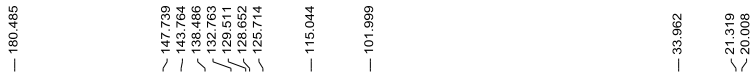


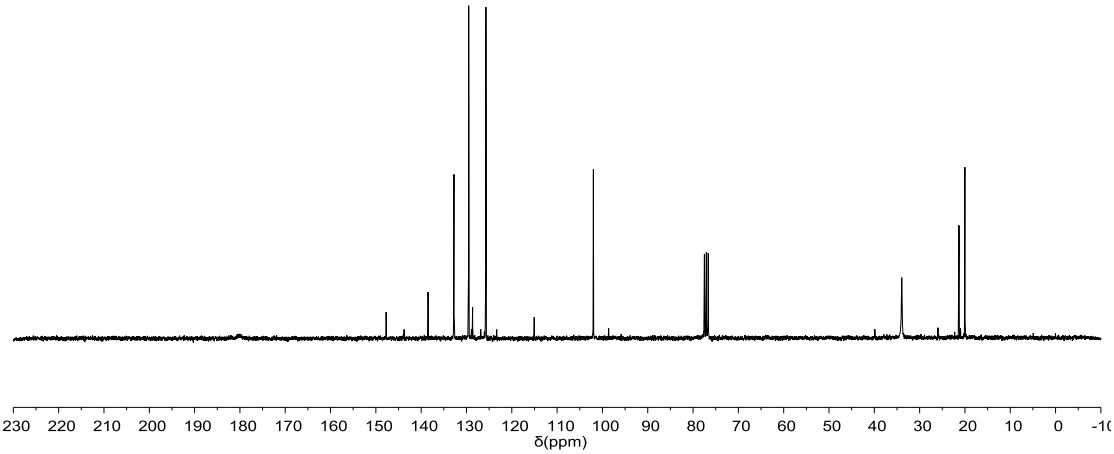
^{1}H NMR and ^{13}C NMR Spectra of Compound 3be



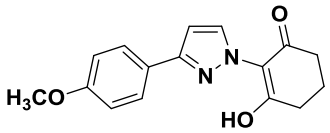


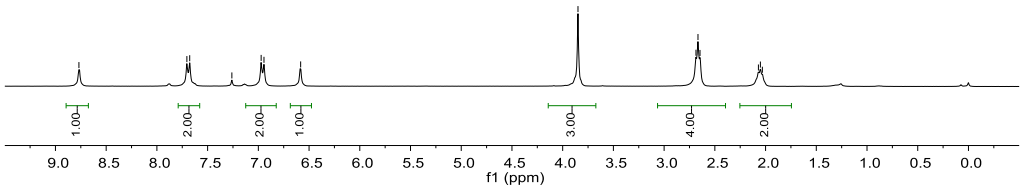


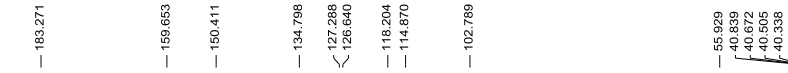


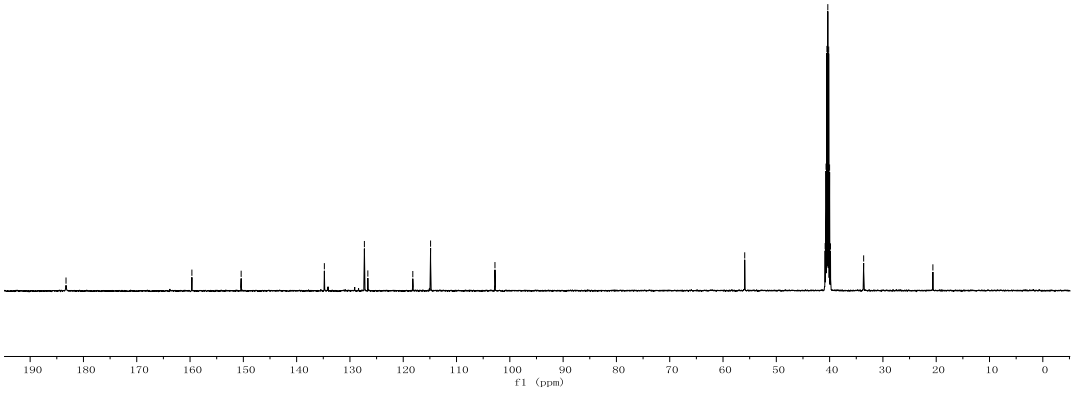


^{1}H NMR and ^{13}C NMR Spectra of Compound 3ce

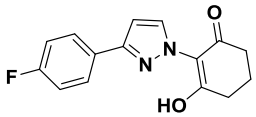


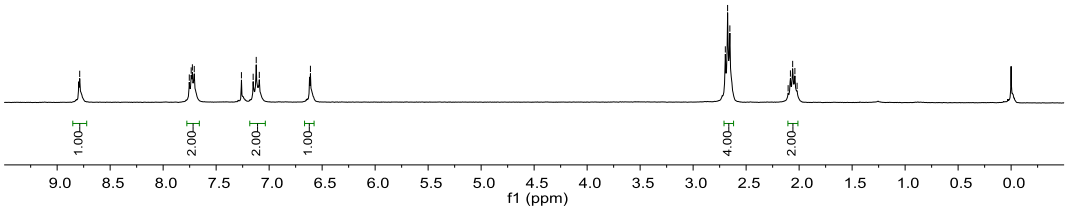


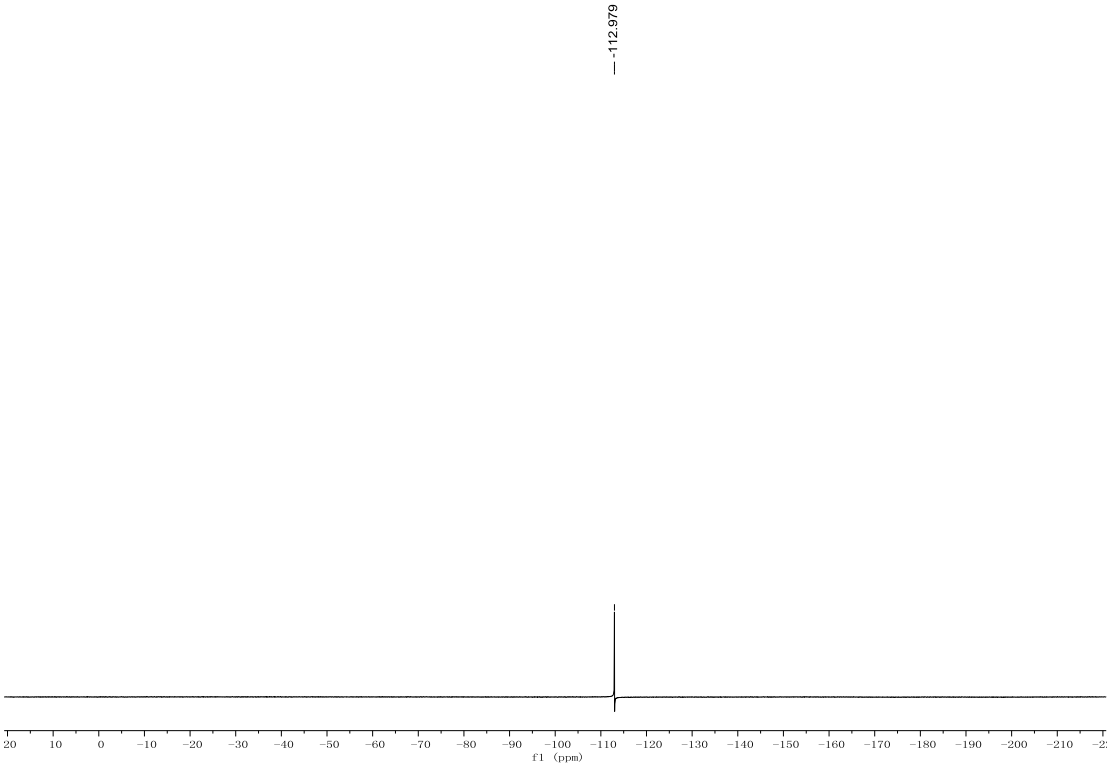


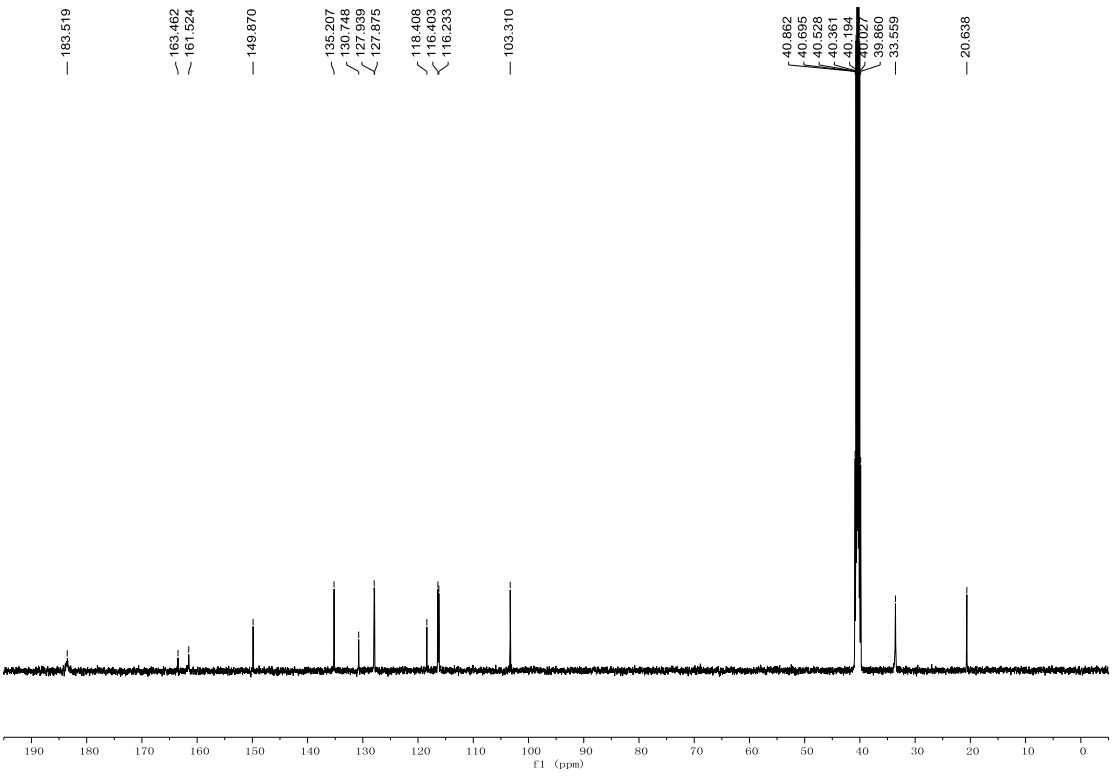


^{1}H NMR, ^{19}F NMR and ^{13}C NMR Spectra of Compound 3ke





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127.875
118.408
116.403
116.233
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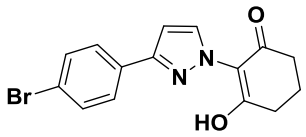
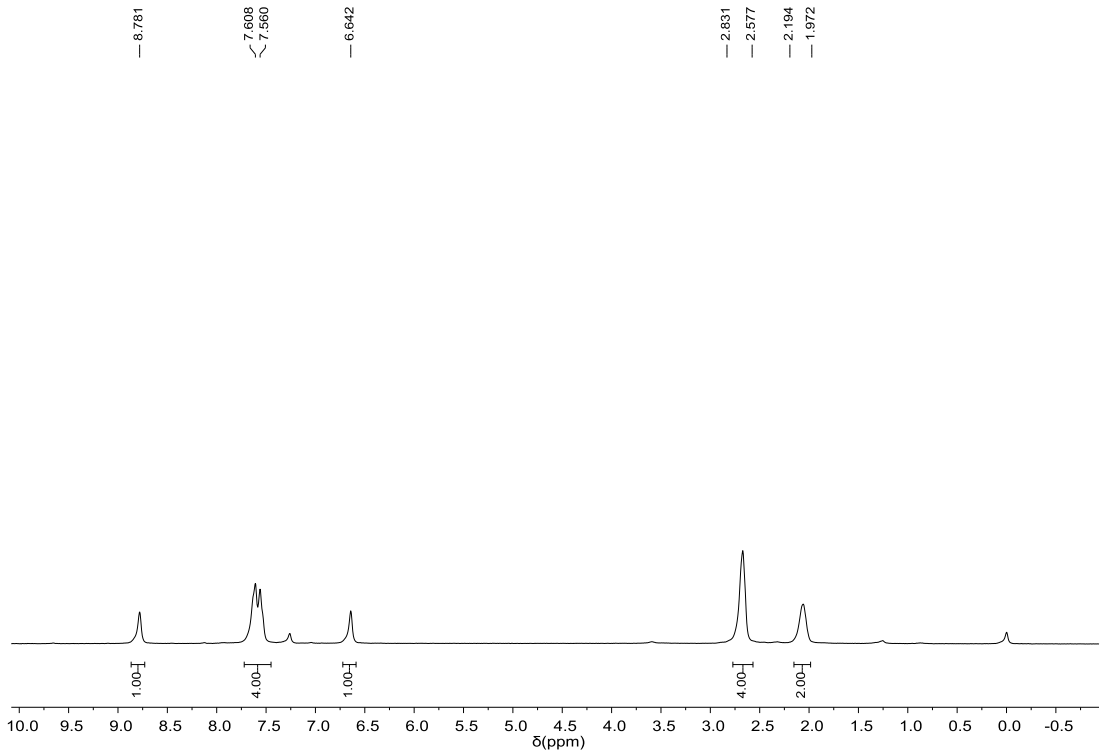
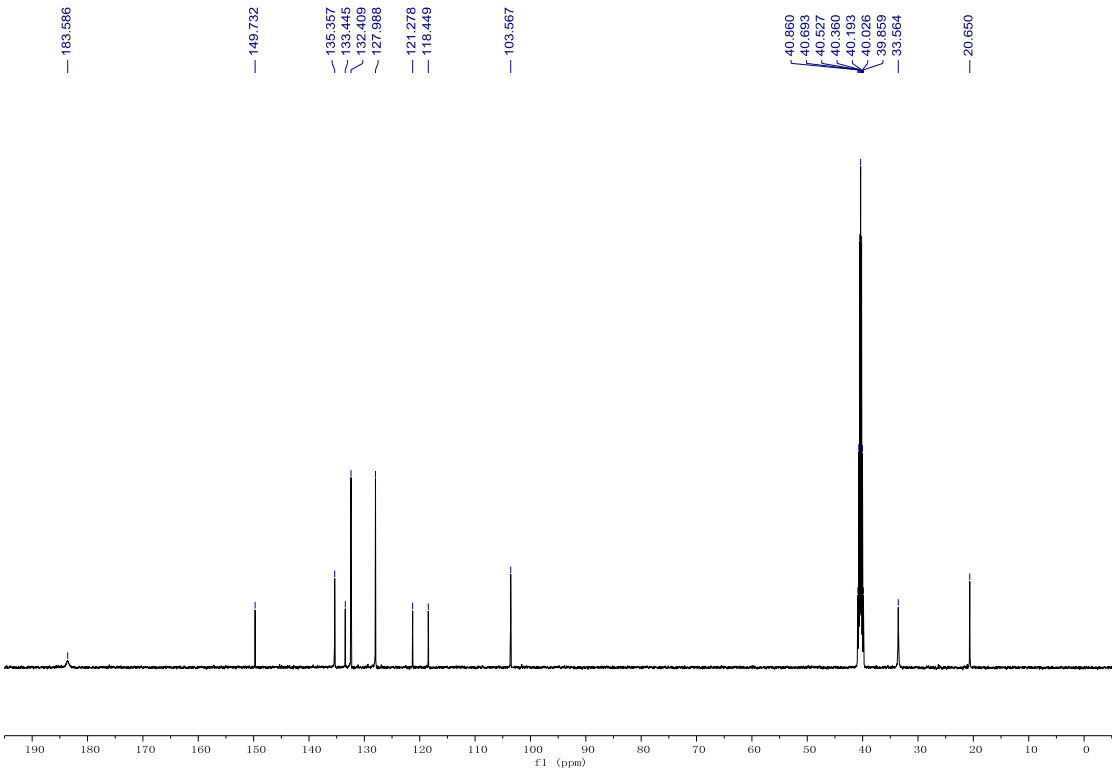
40.862
40.695
40.528
40.361
40.194
40.027
39.860
33.559
20.638
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

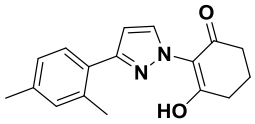
^{1}H NMR and ^{13}C NMR Spectra of Compound 3ne

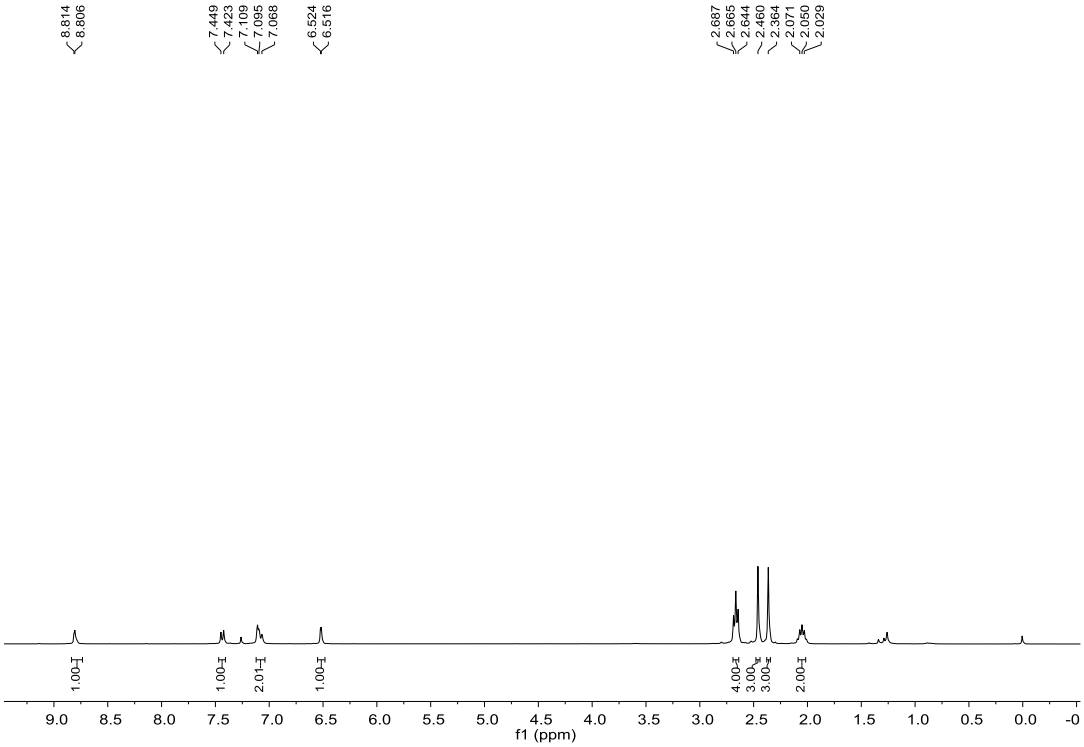


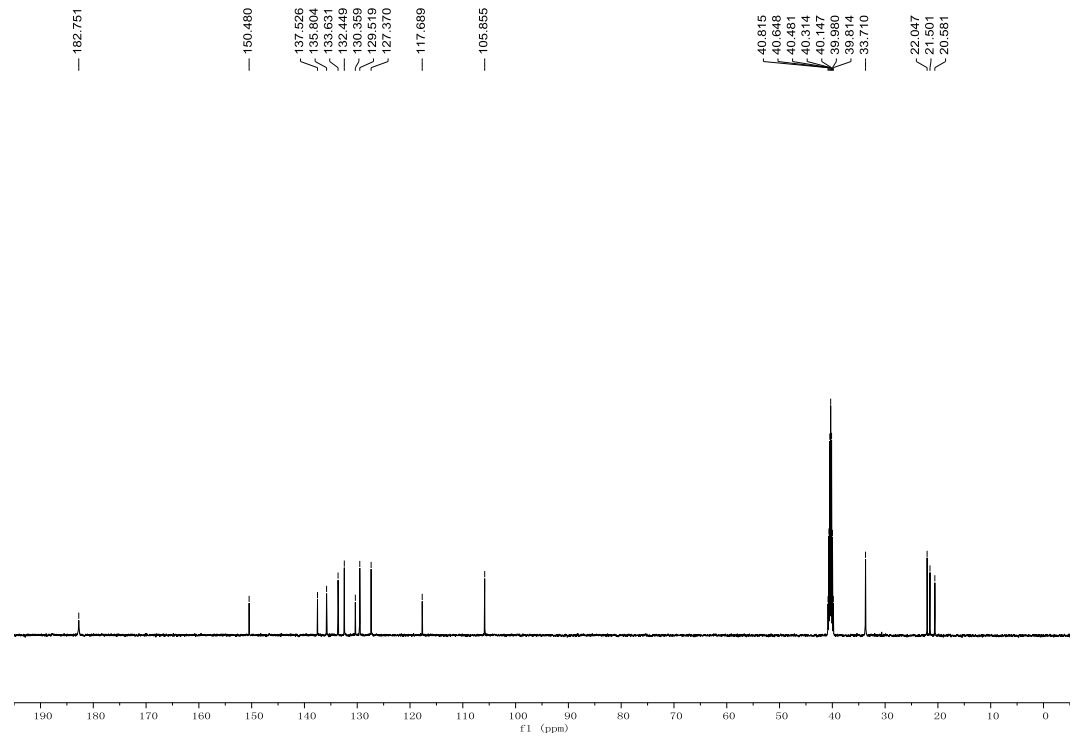




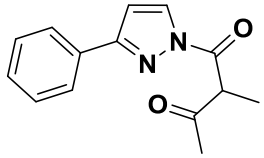
^{1}H NMR and ^{13}C NMR Spectra of Compound 3me

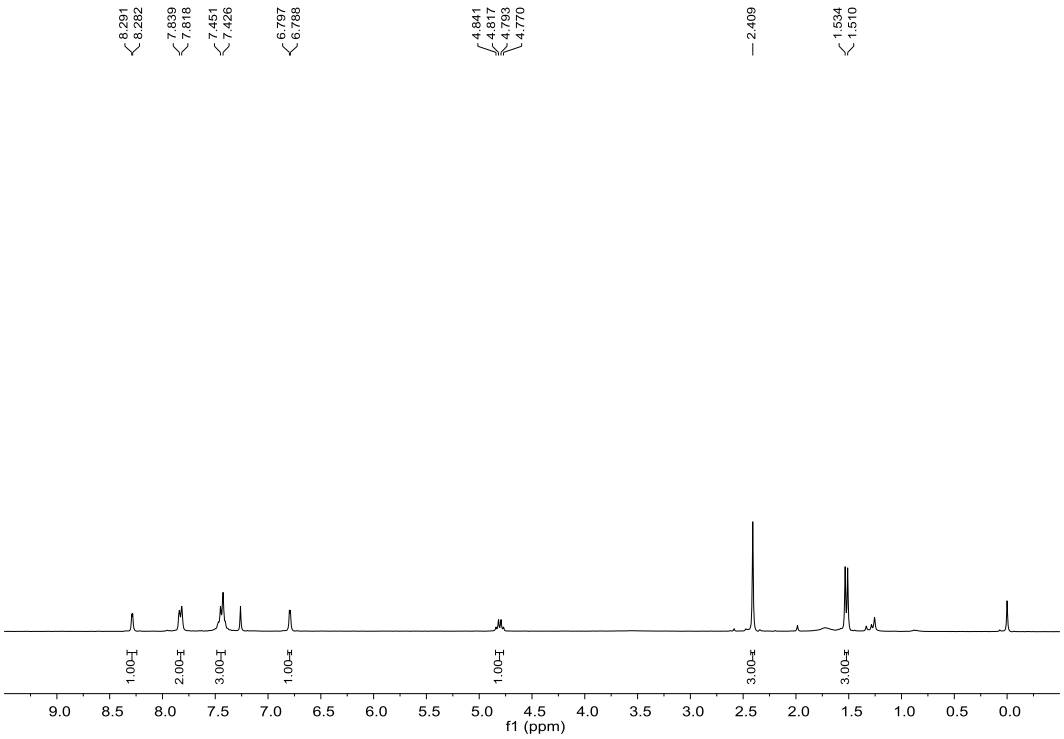


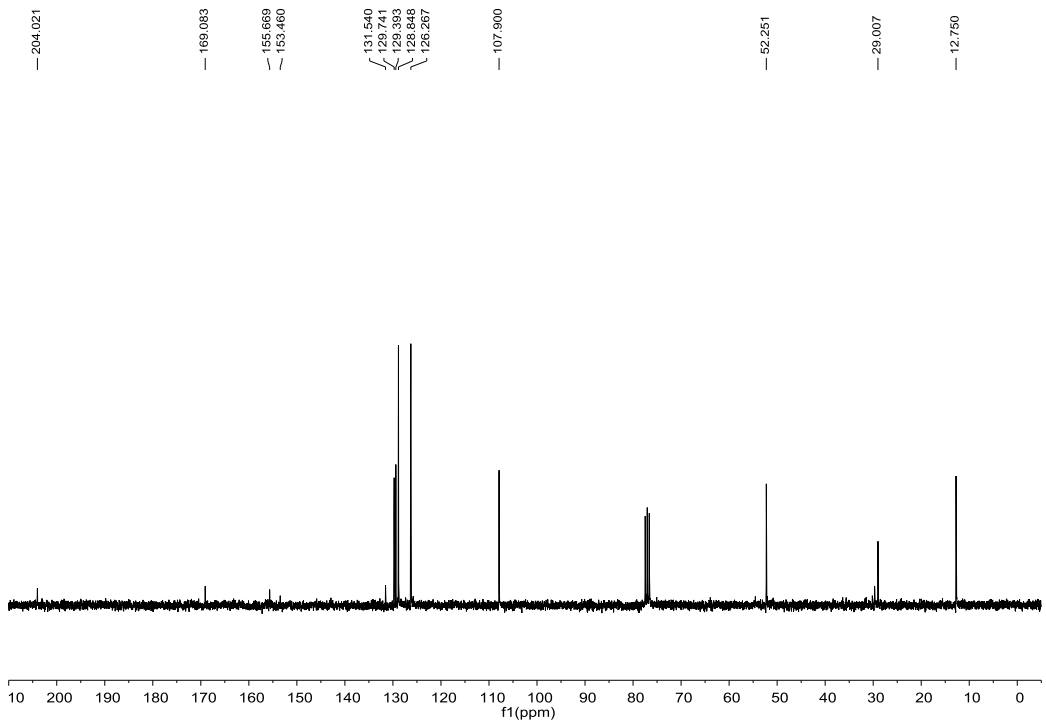




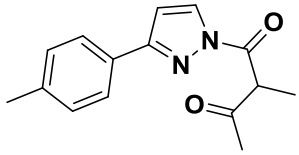
^{1}H NMR and ^{13}C NMR Spectra of Compound 5aa

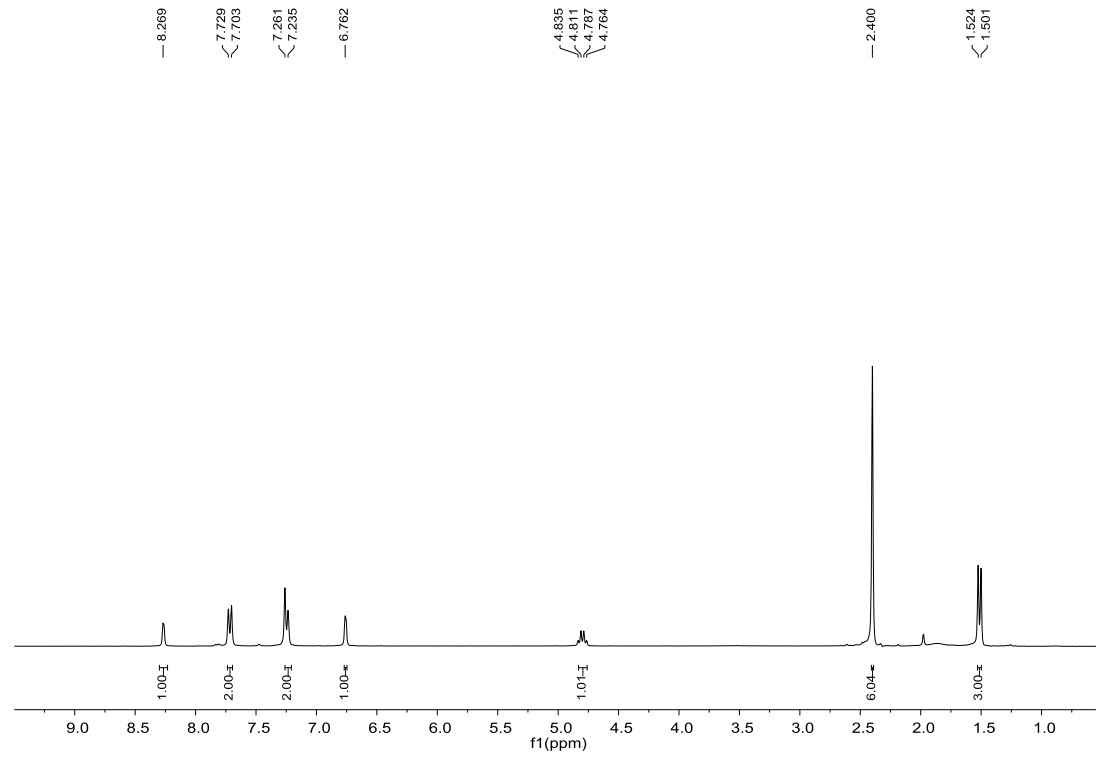


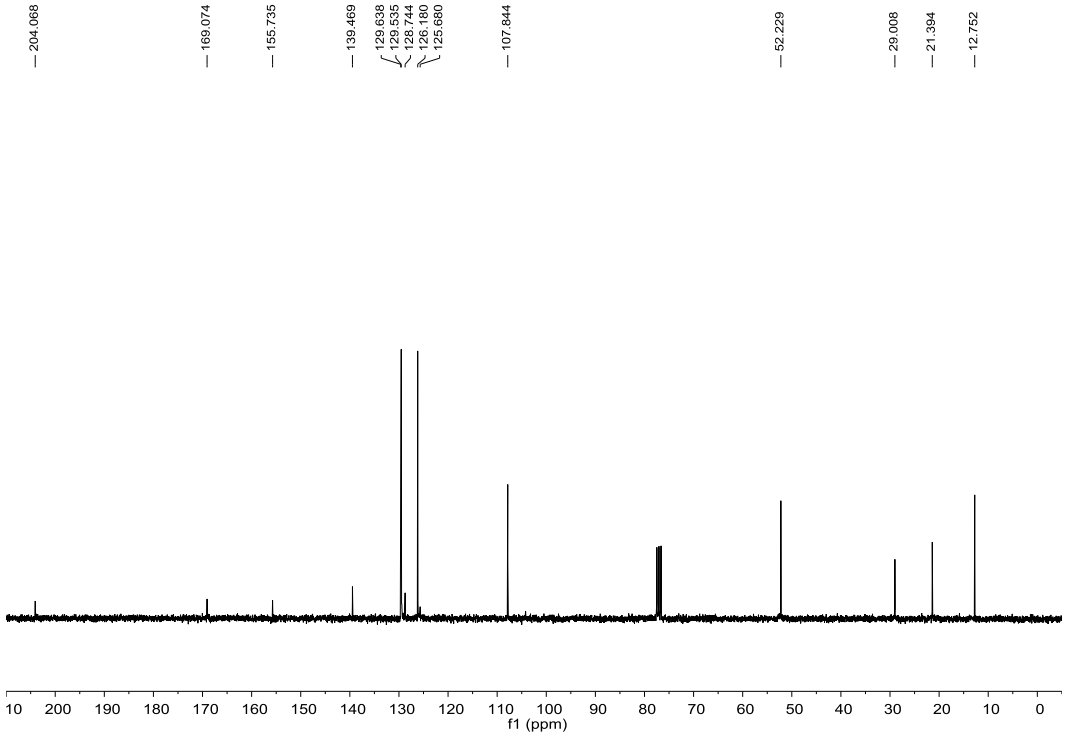




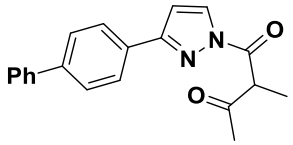
^{1}H NMR and ^{13}C NMR Spectra of Compound 5ba

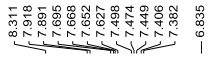


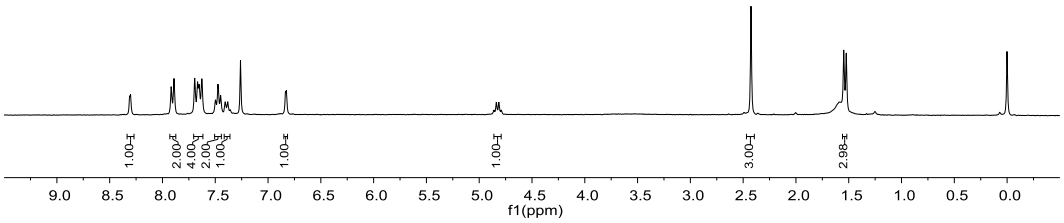


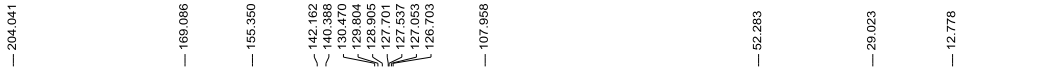


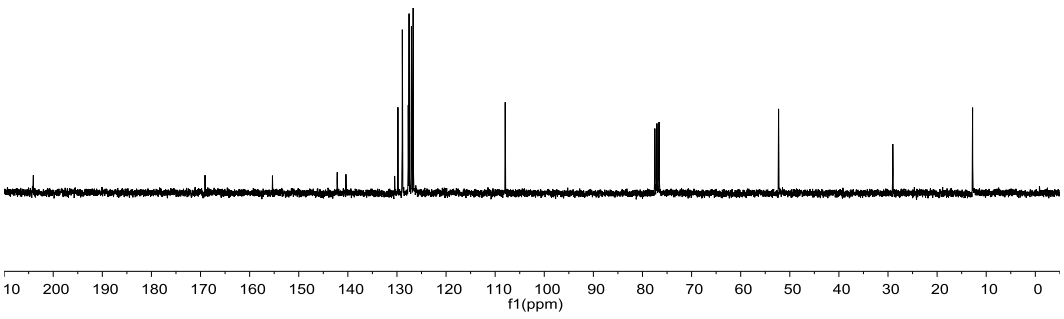
^{1}H NMR and ^{13}C NMR Spectra of Compound 5fa



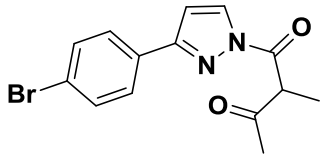


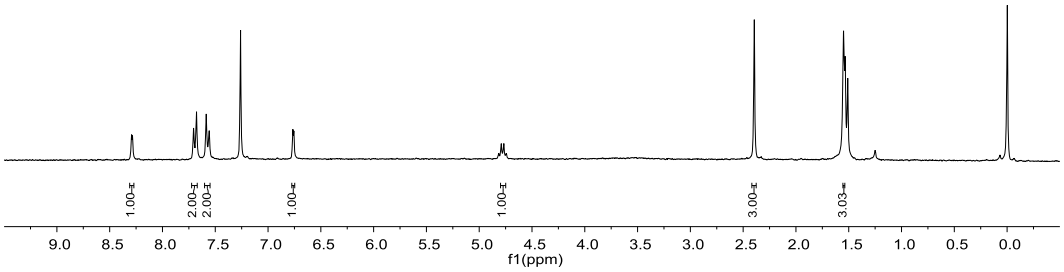


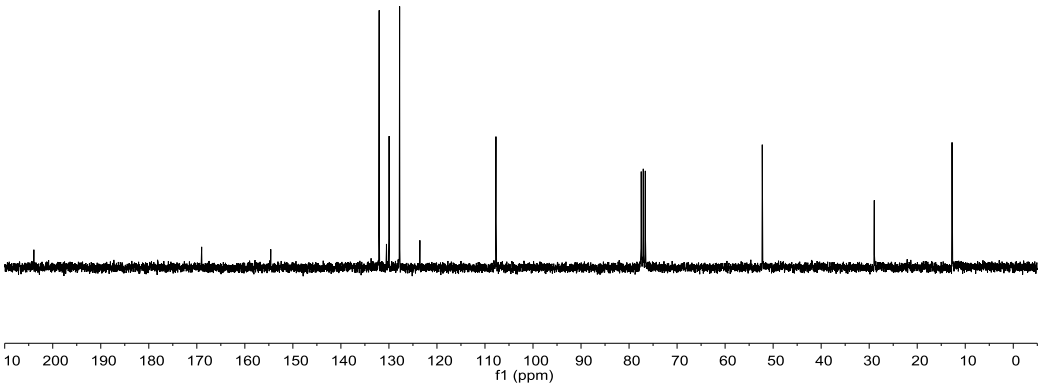




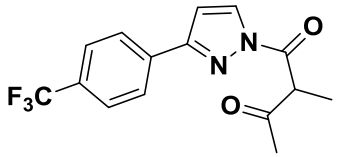
^{1}H NMR and ^{13}C NMR Spectra of Compound 5na

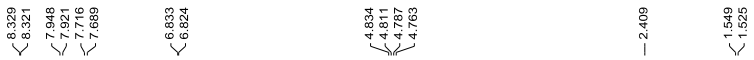


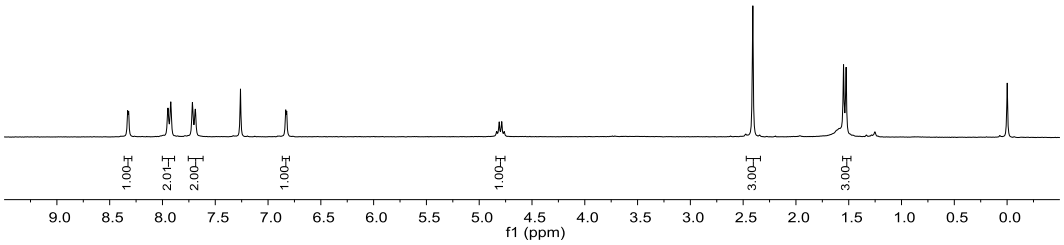


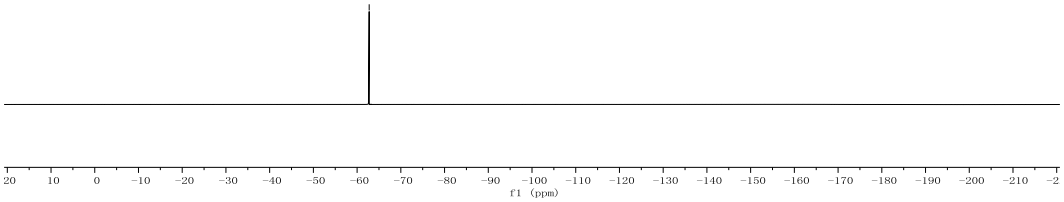


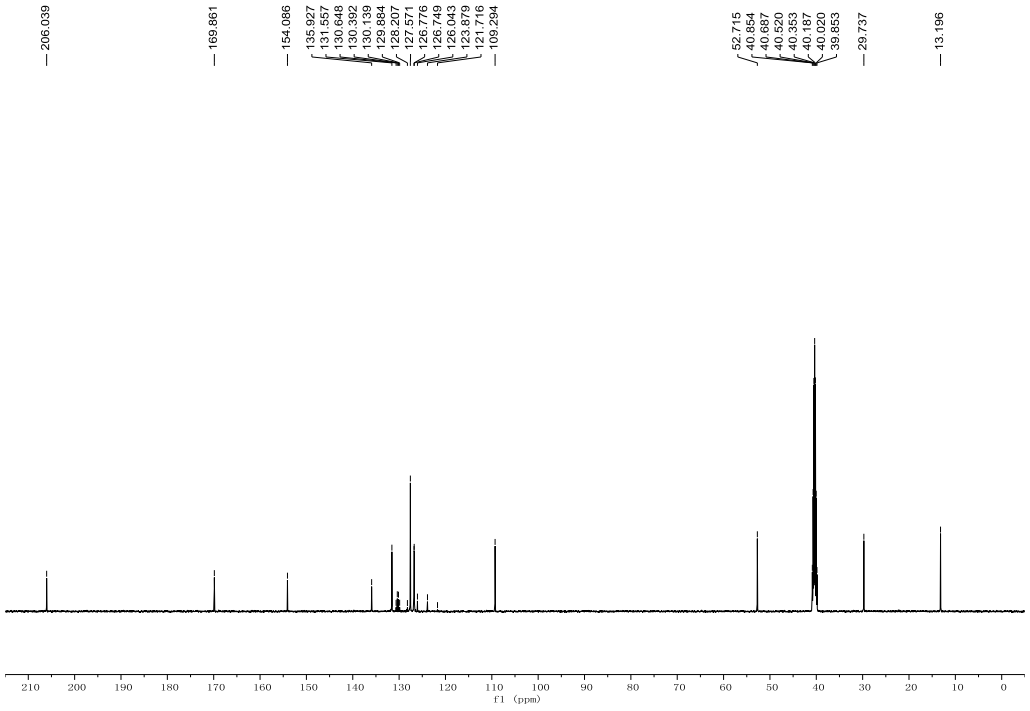
^{1}H NMR, ^{19}F NMR and ^{13}C NMR Spectra of Compound 5ea







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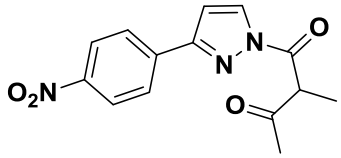
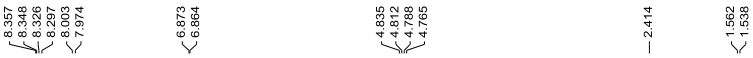
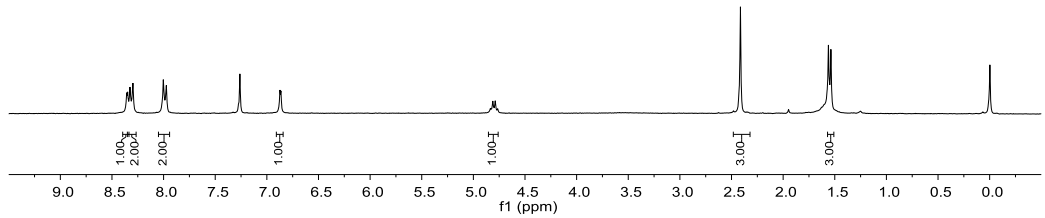
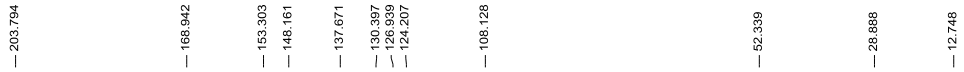
121.716
109.294
52.715
40.854
40.687
40.520
40.353
40.187
40.020
39.853
29.737
13.196
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

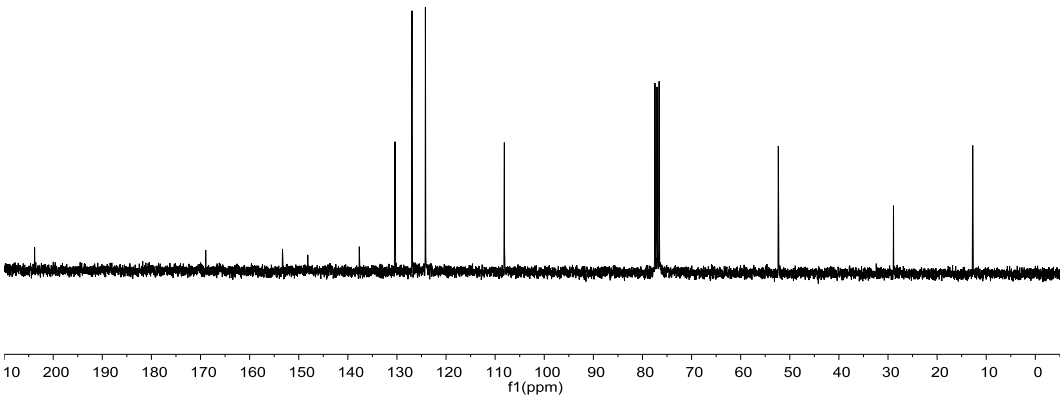
^{1}H NMR and ^{13}C NMR Spectra of Compound 5la



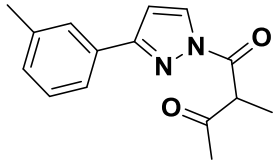


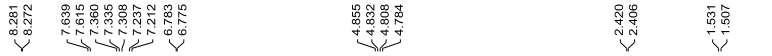


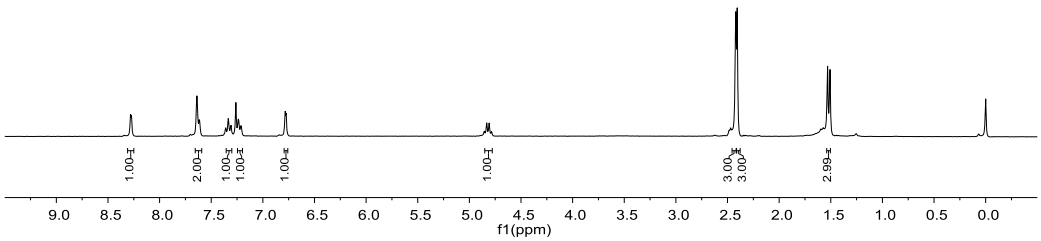


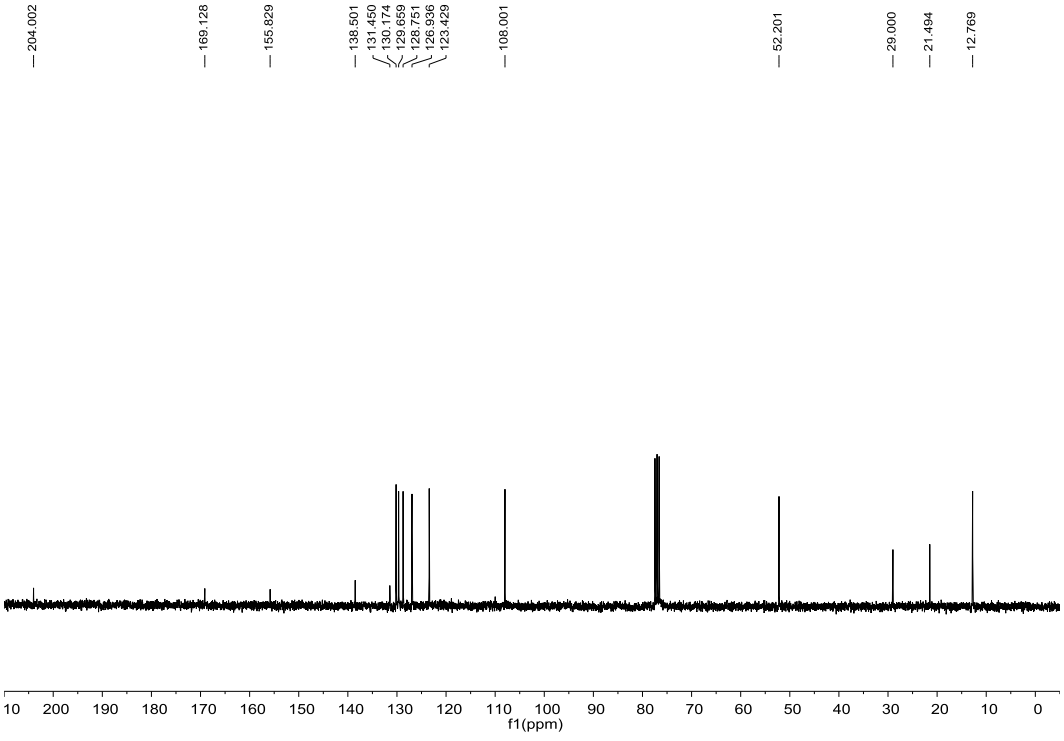


^{1}H NMR and ^{13}C NMR Spectra of Compound 5ga

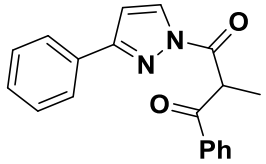


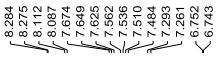


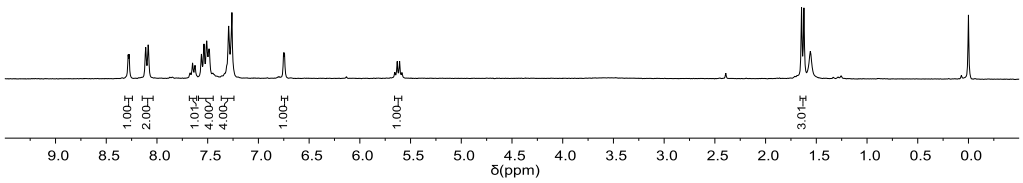


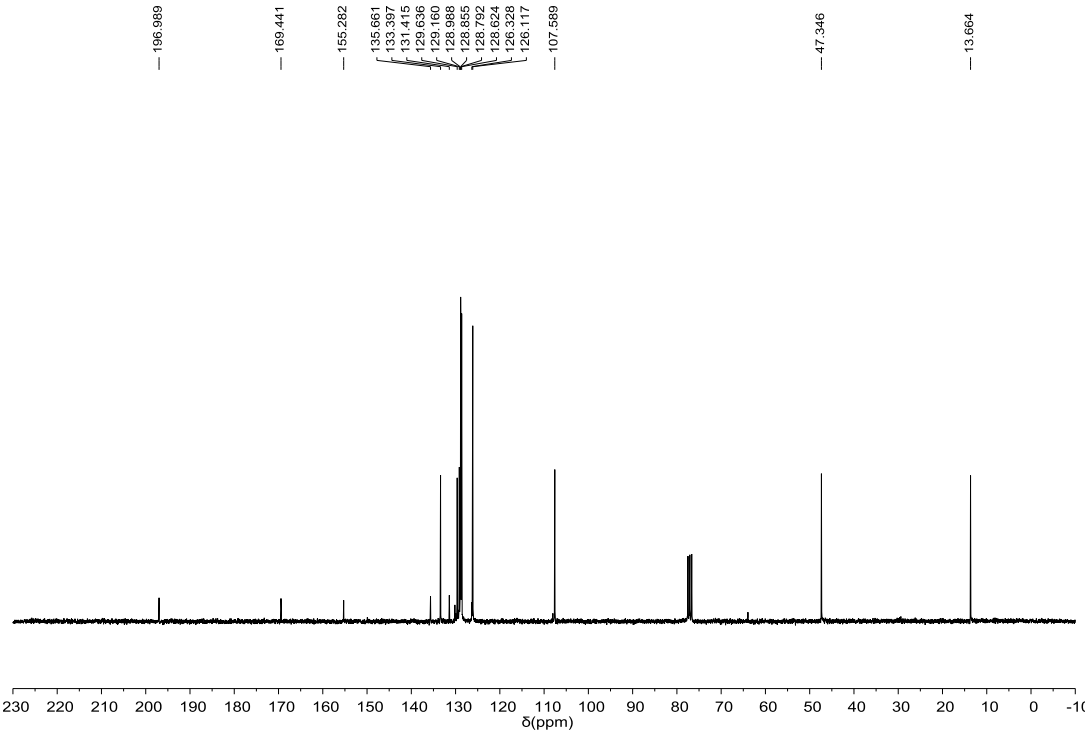


^{1}H NMR and ^{13}C NMR Spectra of Compound 5ab

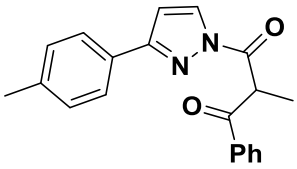


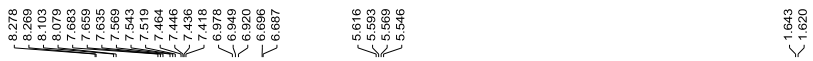


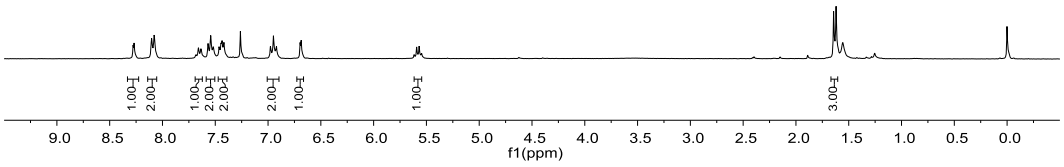


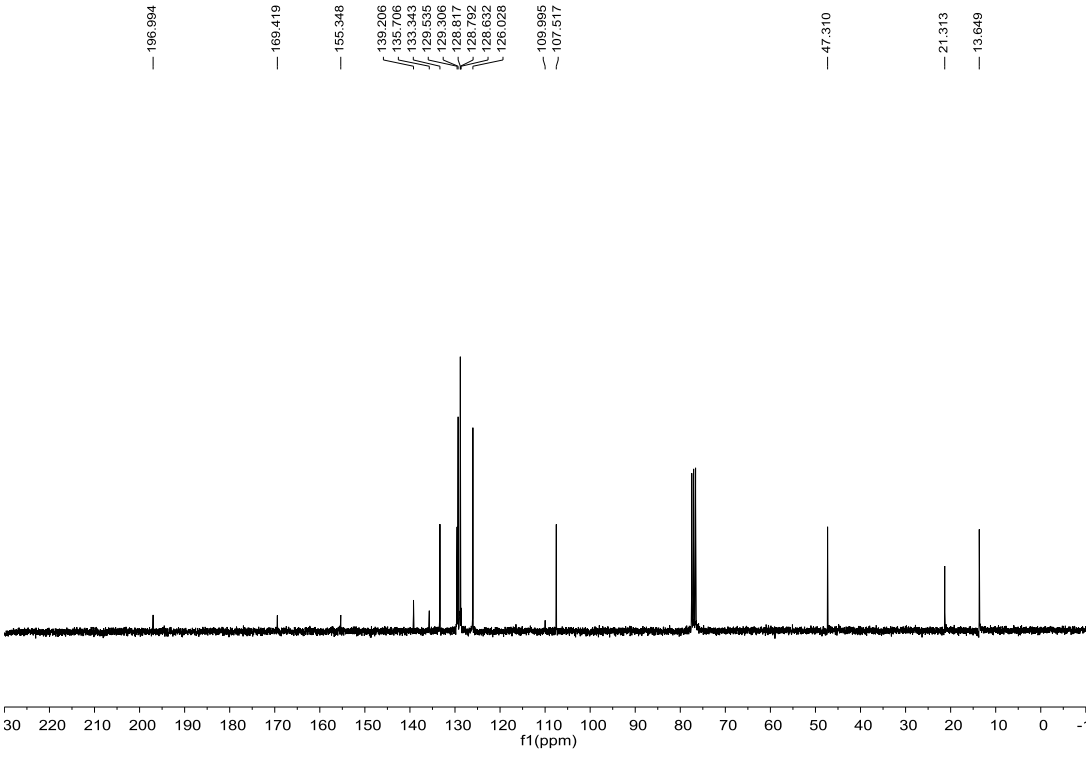


^{1}H NMR and ^{13}C NMR Spectra of Compound 5bb

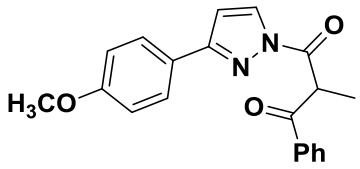


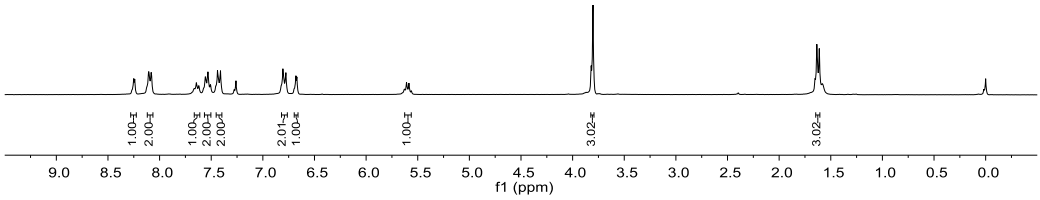


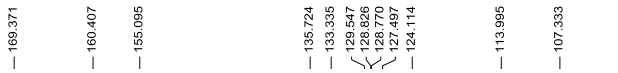


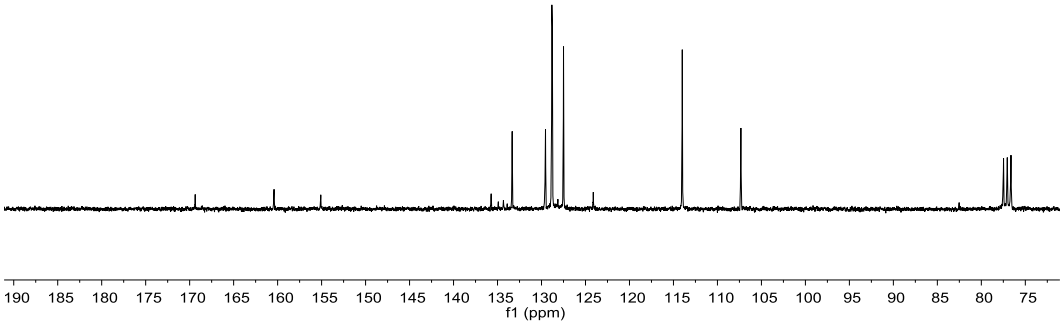


^{1}H NMR and ^{13}C NMR Spectra of Compound 5cb

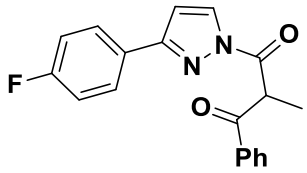


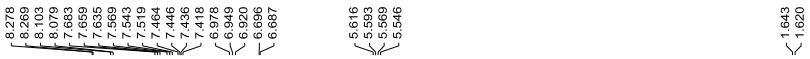


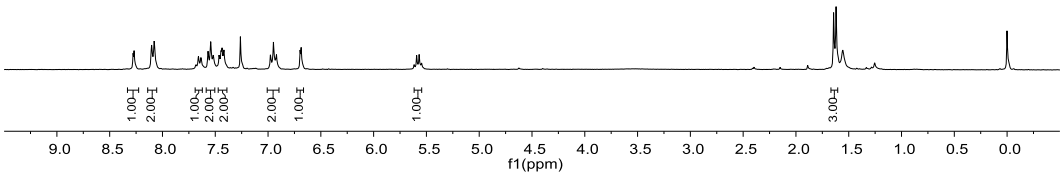


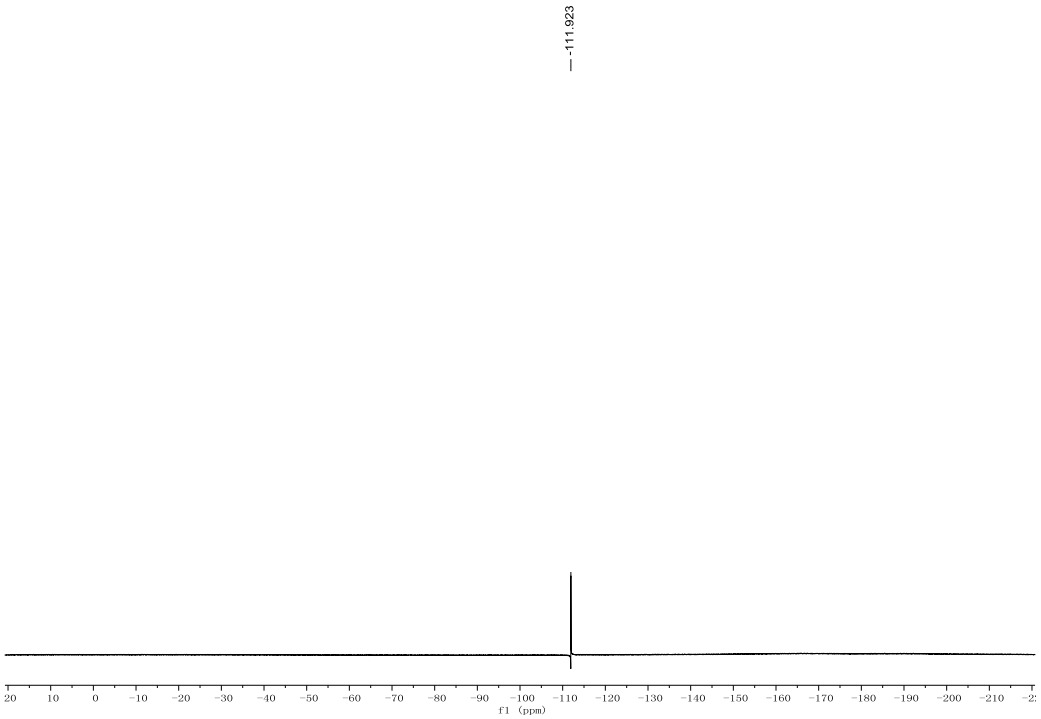


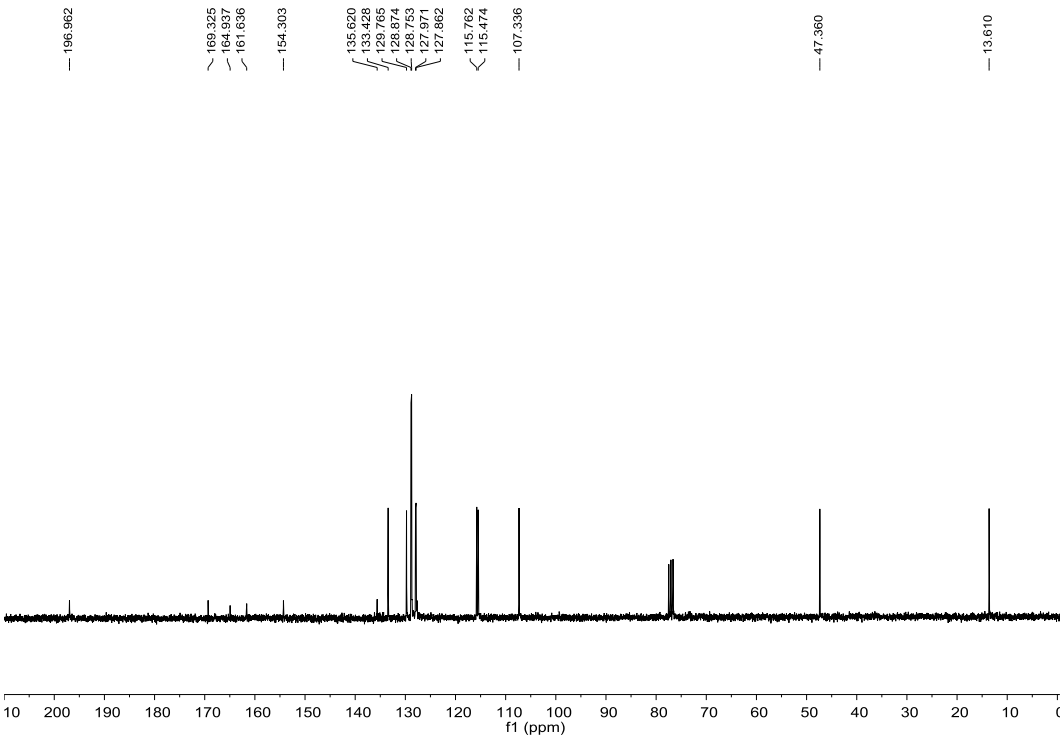
^{1}H NMR and ^{13}C NMR Spectra of Compound 5kb



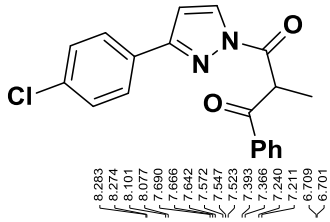


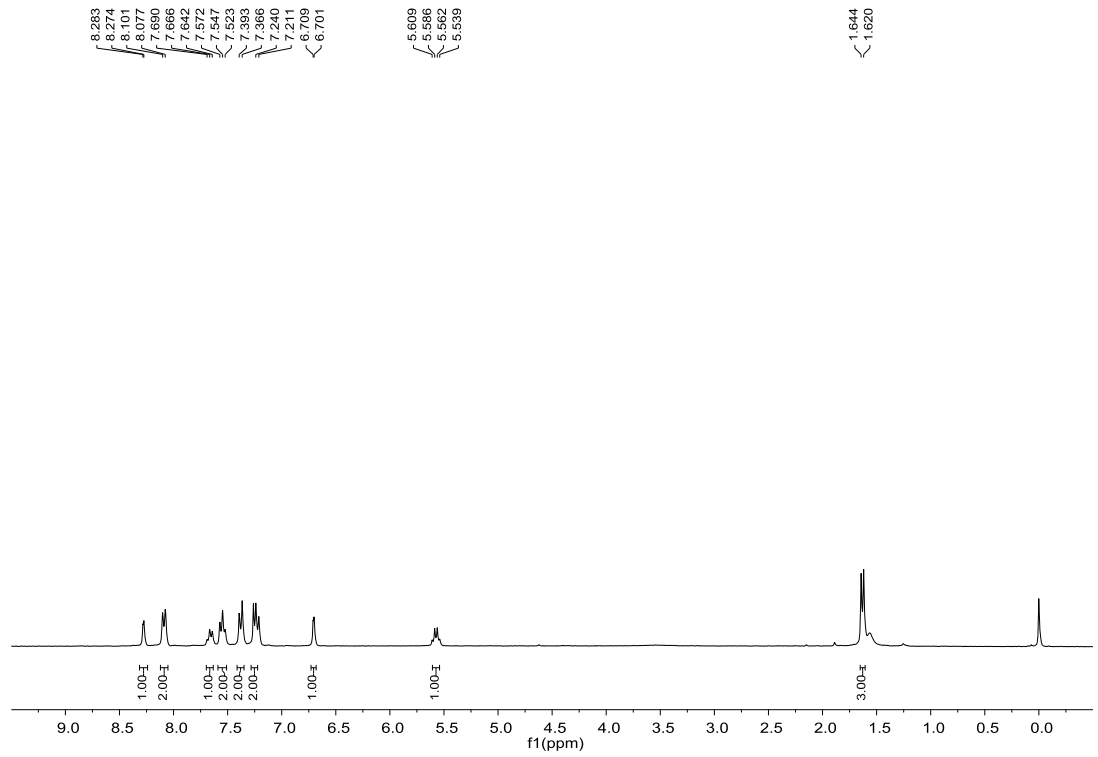


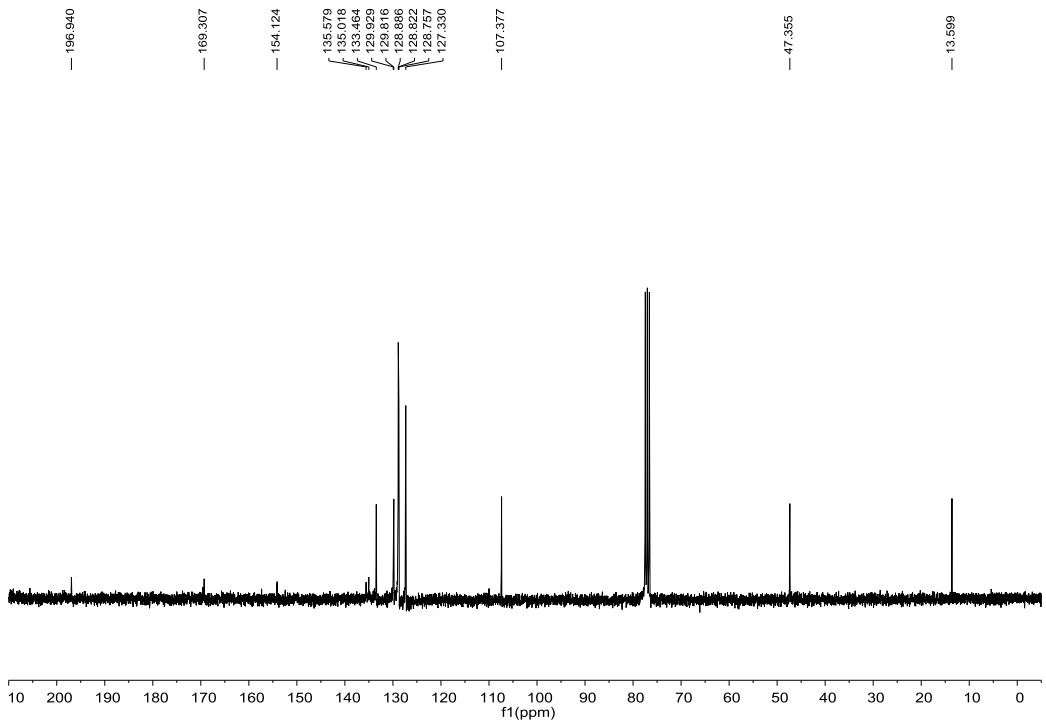



196.962
169.325
164.937
161.636
154.303
135.620
133.428
129.765
128.874
128.753
127.971
127.862
115.762
115.474
107.336
47.360
13.610
f1 (ppm)

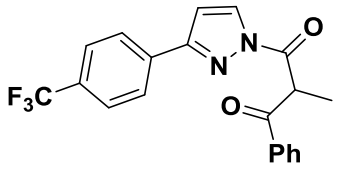
^{1}H NMR and ^{13}C NMR Spectra of Compound 5db

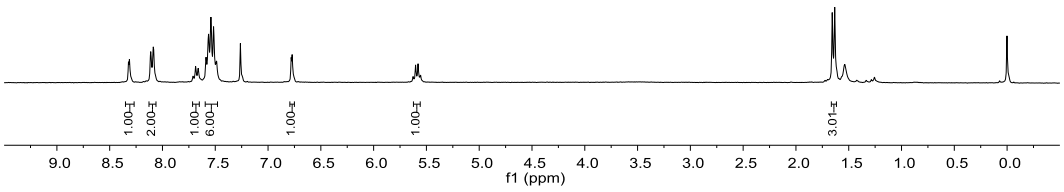


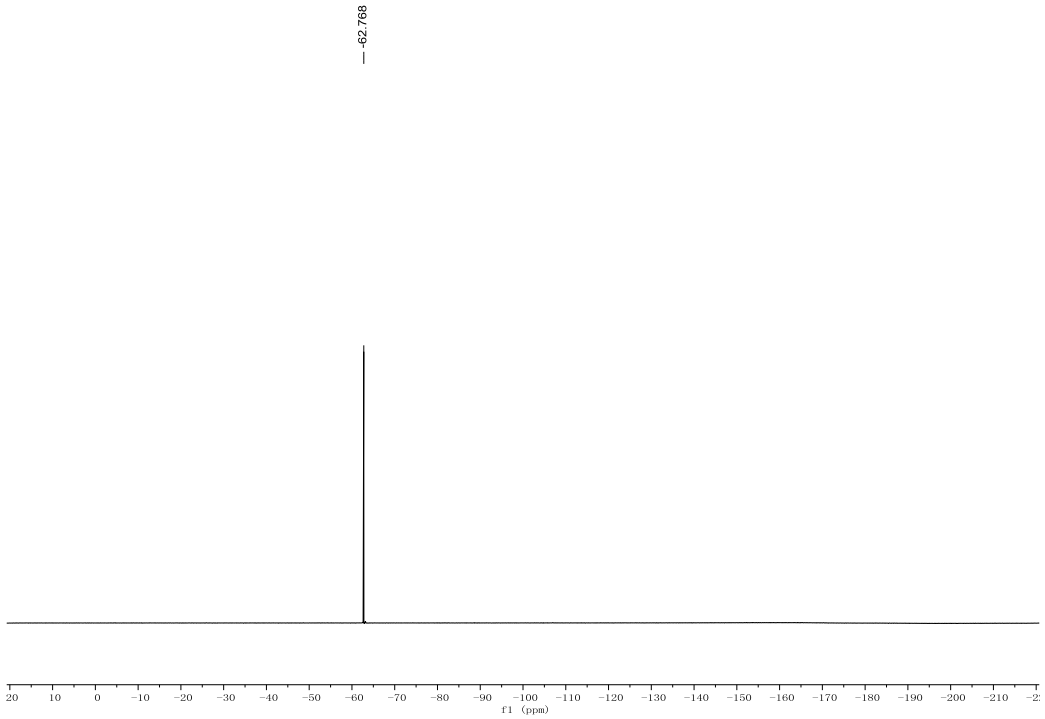


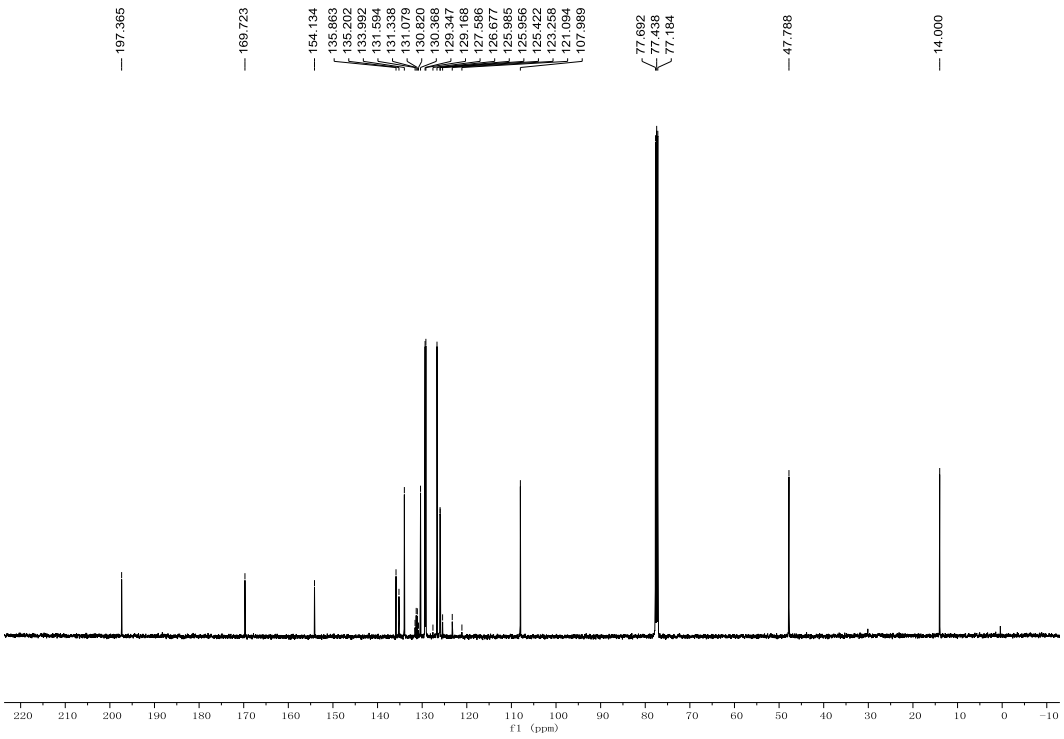


^{1}H NMR, ^{19}F NMR and ^{13}C NMR Spectra of Compound 5eb





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169.723
154.134
135.863
135.202
133.992
131.594
131.338
131.079
130.820
130.368
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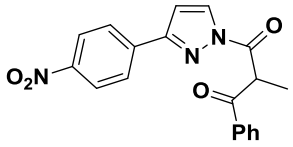
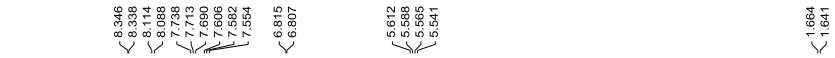
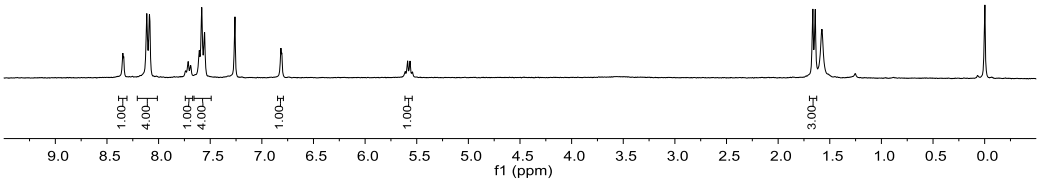
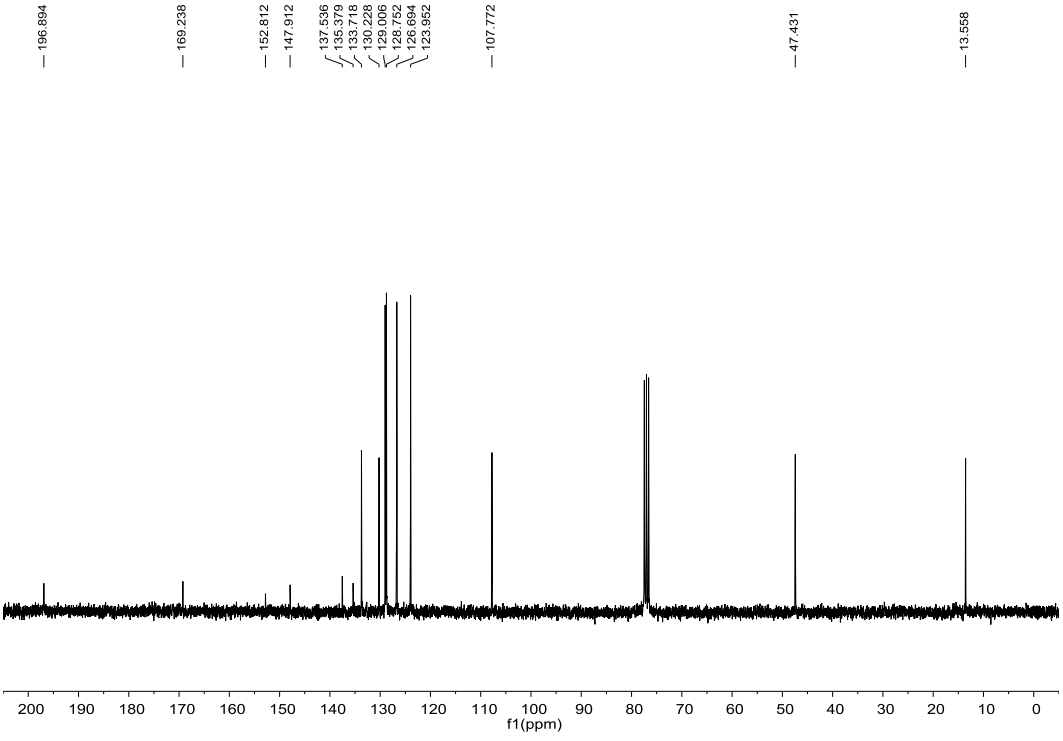
129.168
127.586
126.677
125.985
125.956
125.422
123.258
121.094
107.989
77.692
77.438
77.184
47.788
14.000
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 −10
f1 (ppm)

^{1}H NMR and ^{13}C NMR Spectra of Compound 5lb

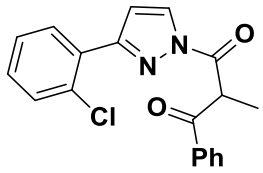


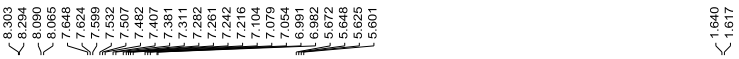


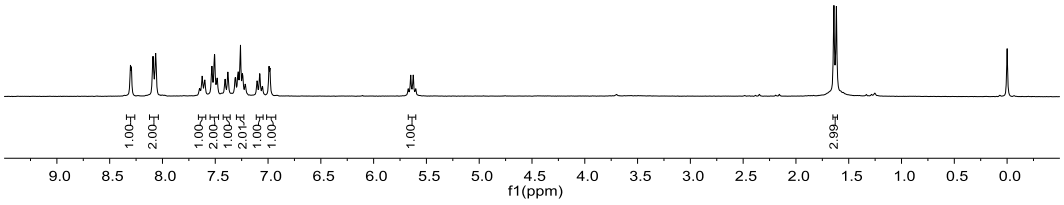


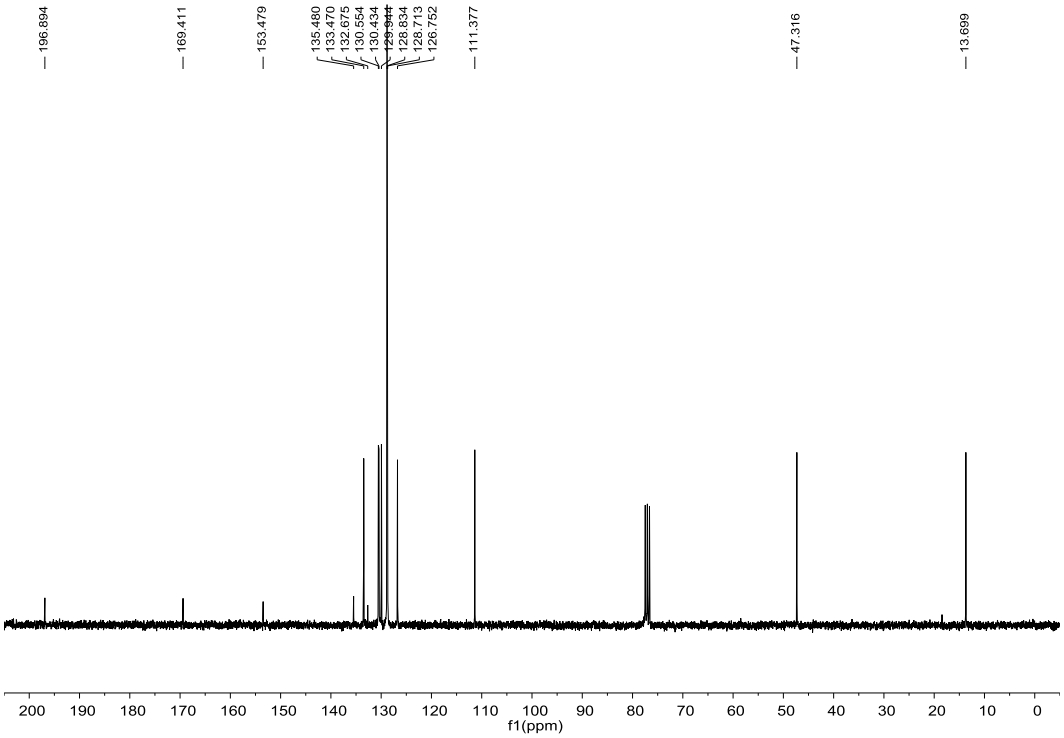


^{1}H NMR and ^{13}C NMR Spectra of Compound 5ib

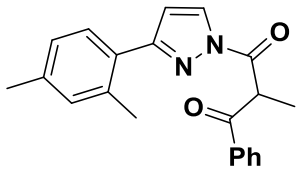


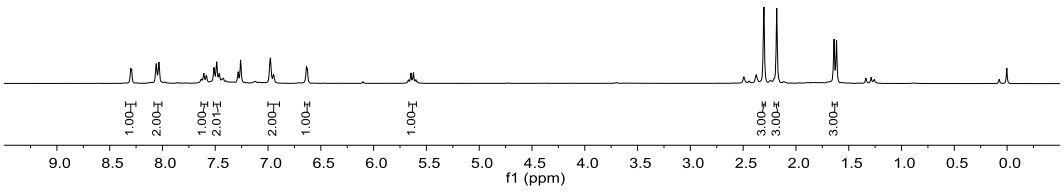


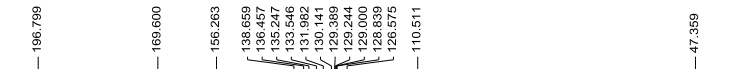


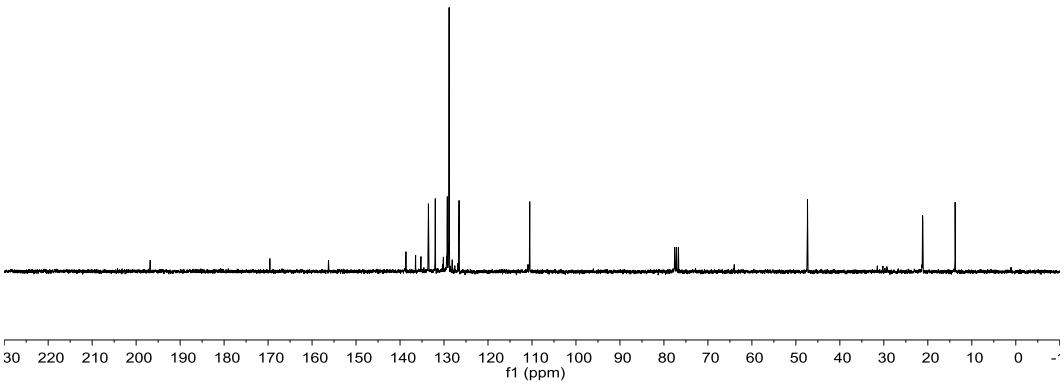


^{1}H NMR and ^{13}C NMR Spectra of Compound 5mb

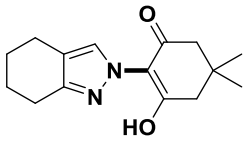


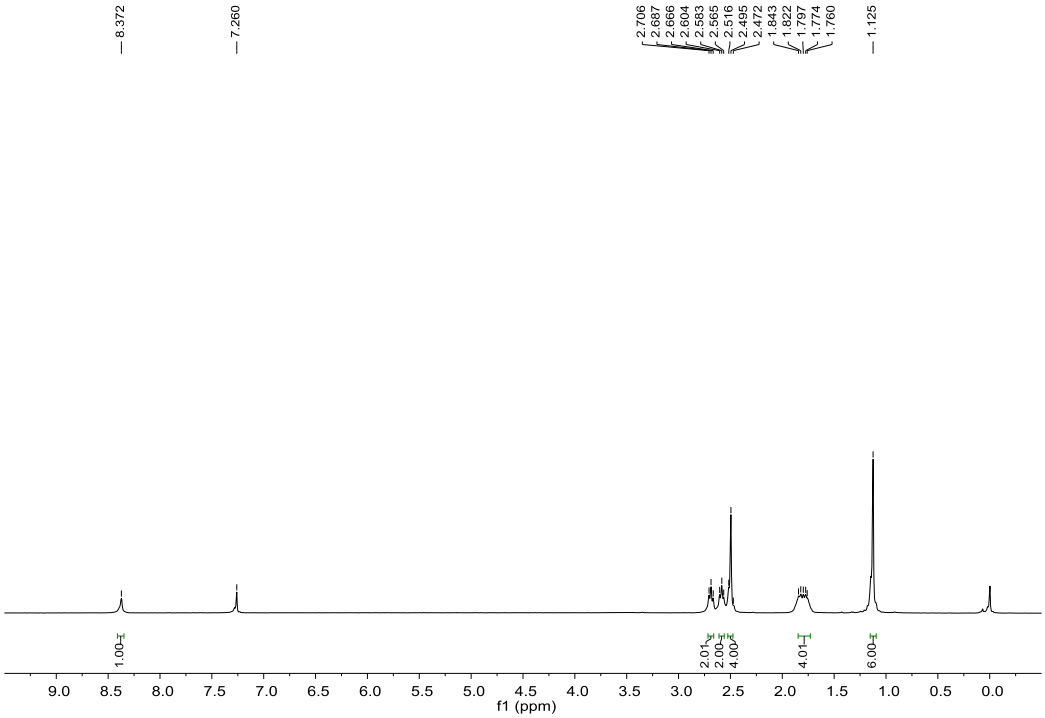


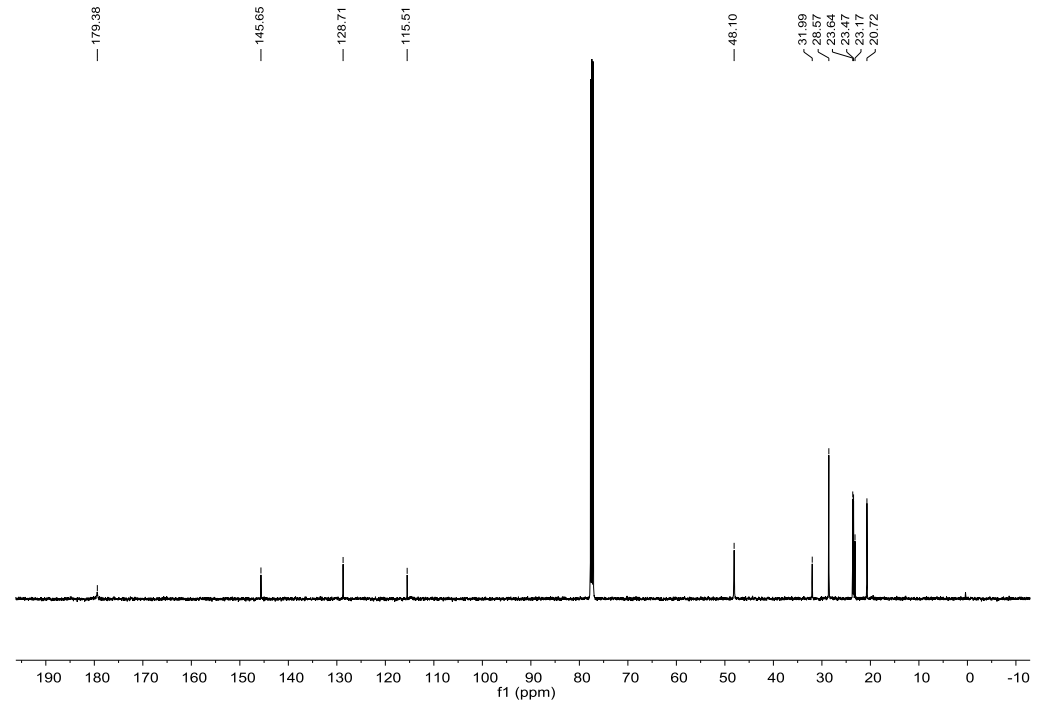




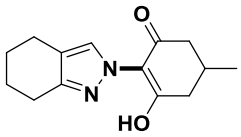
^{1}H NMR and ^{13}C NMR Spectra of Compound 7aa

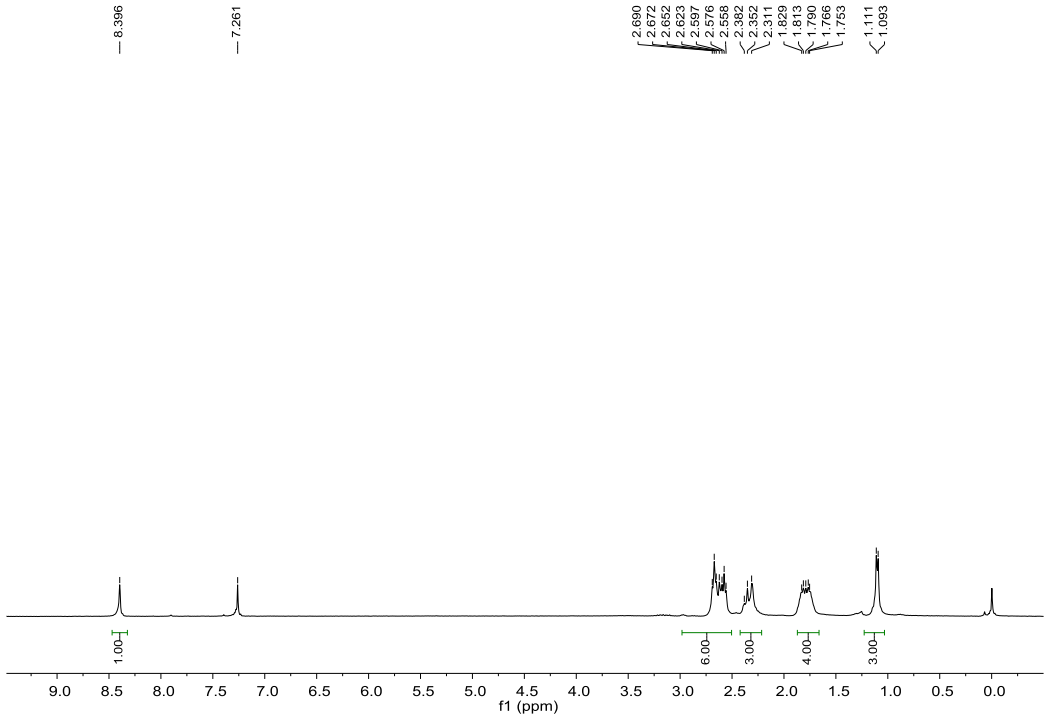


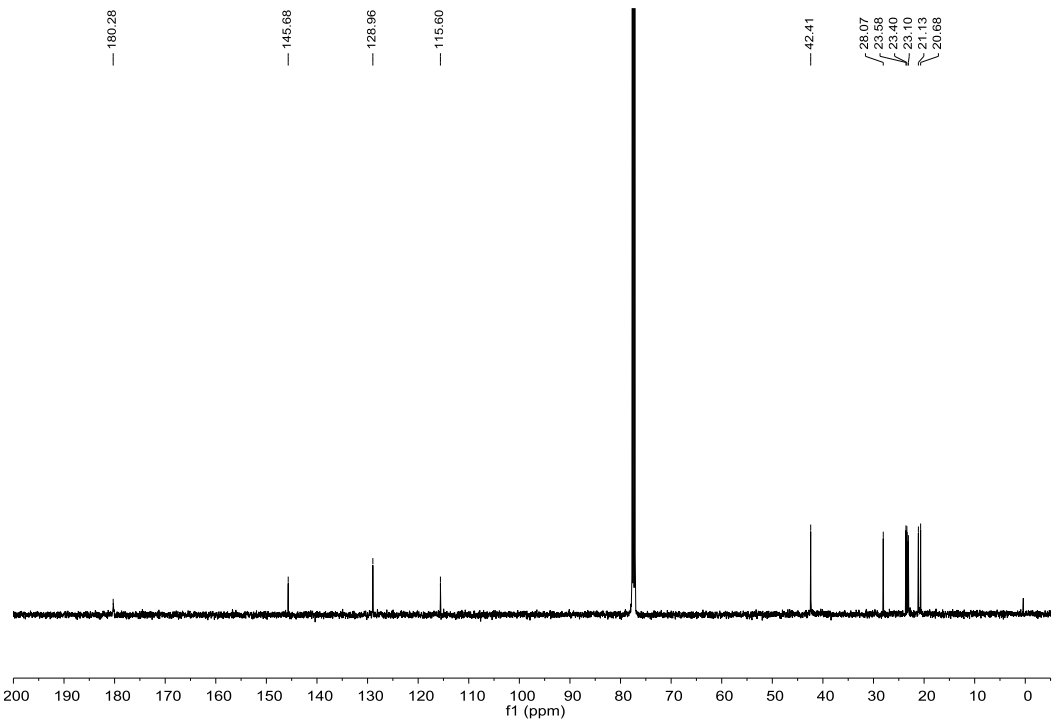




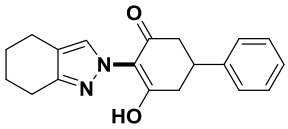
^{1}H NMR and ^{13}C NMR Spectra of Compound 7ab

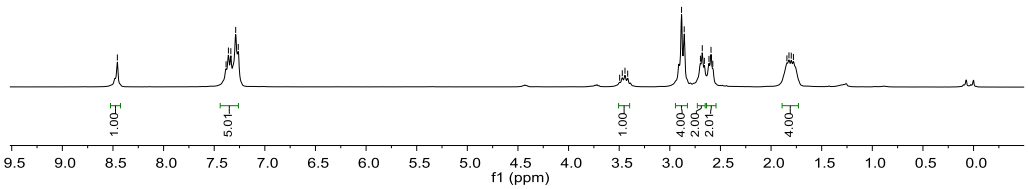


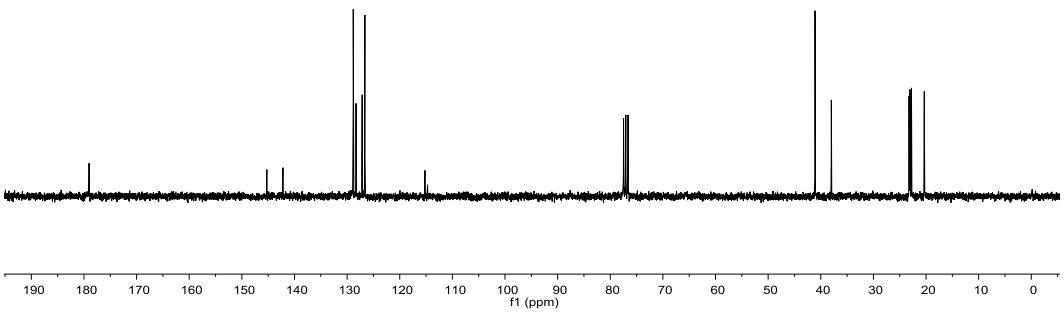




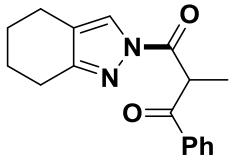
^{1}H NMR and ^{13}C NMR Spectra of Compound 7ac

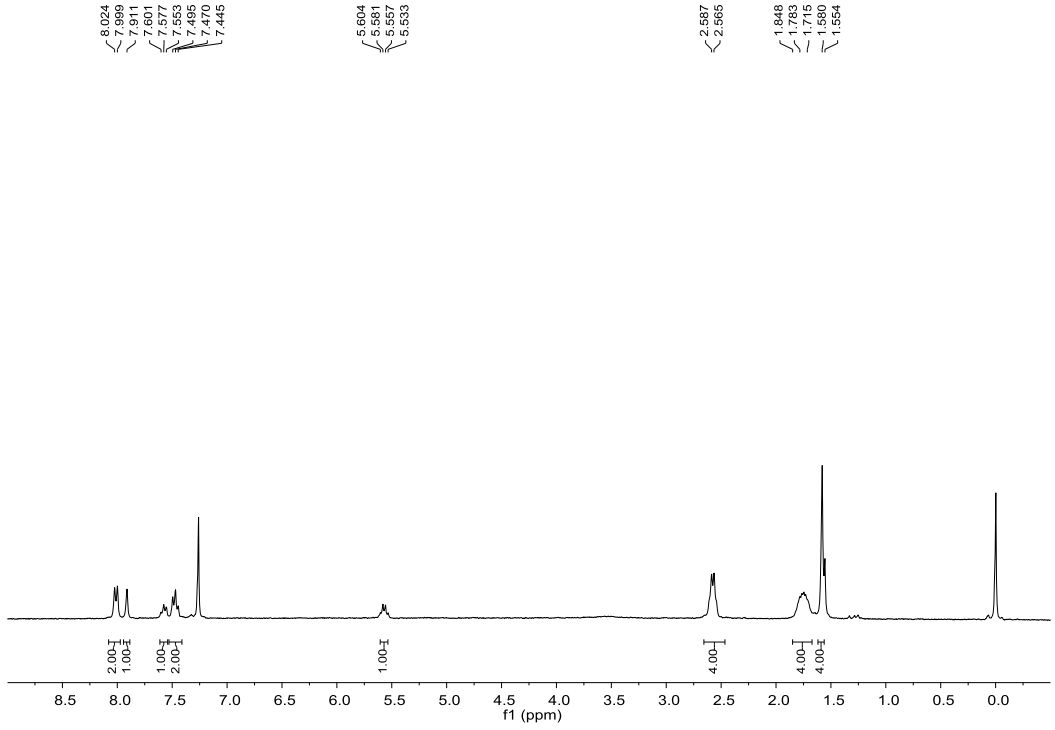


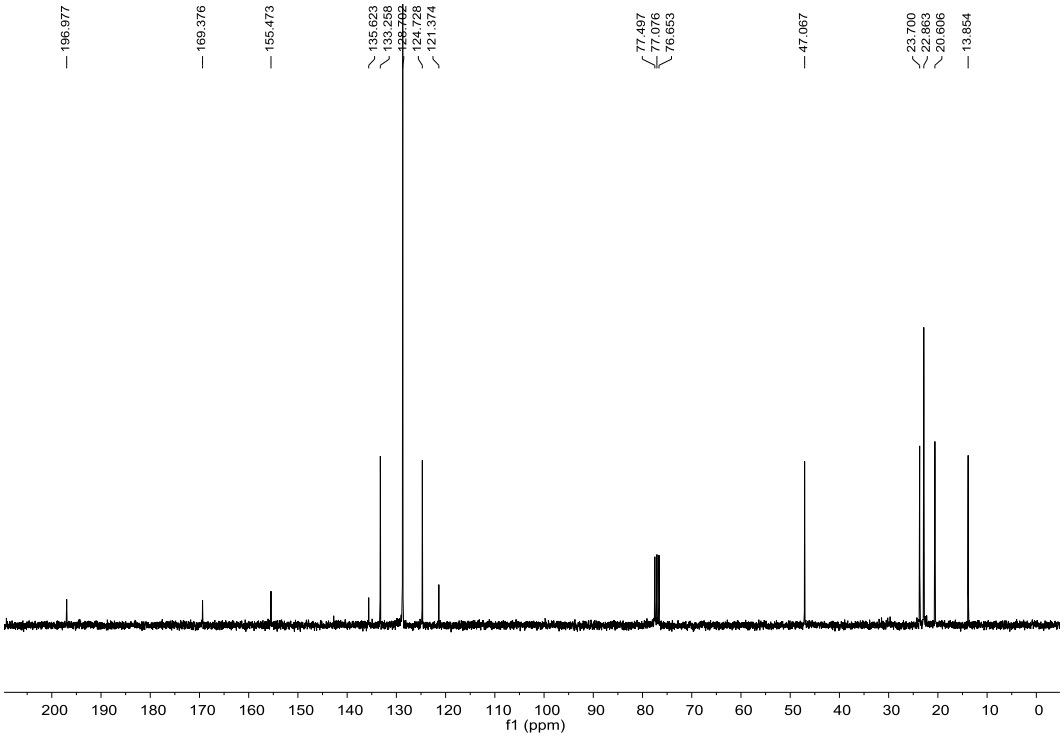




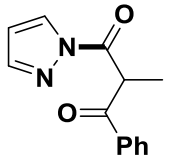
^{1}H NMR and ^{13}C NMR Spectra of Compound 8ab

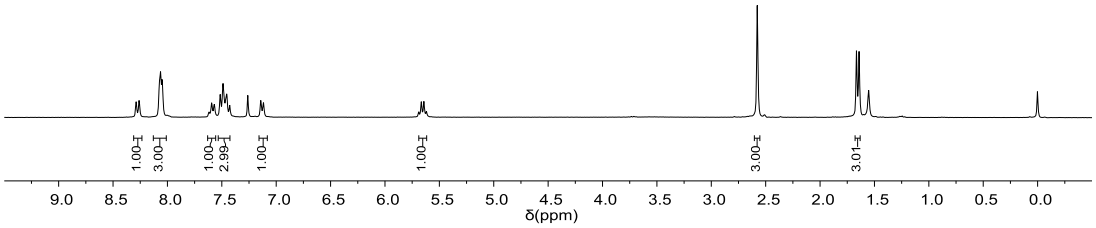


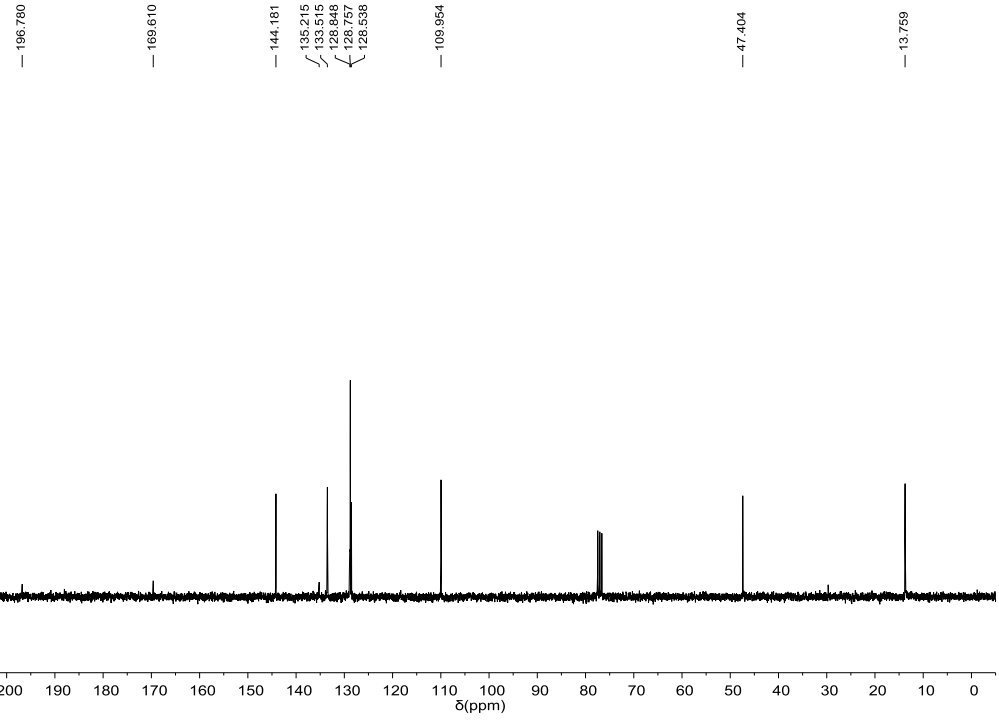




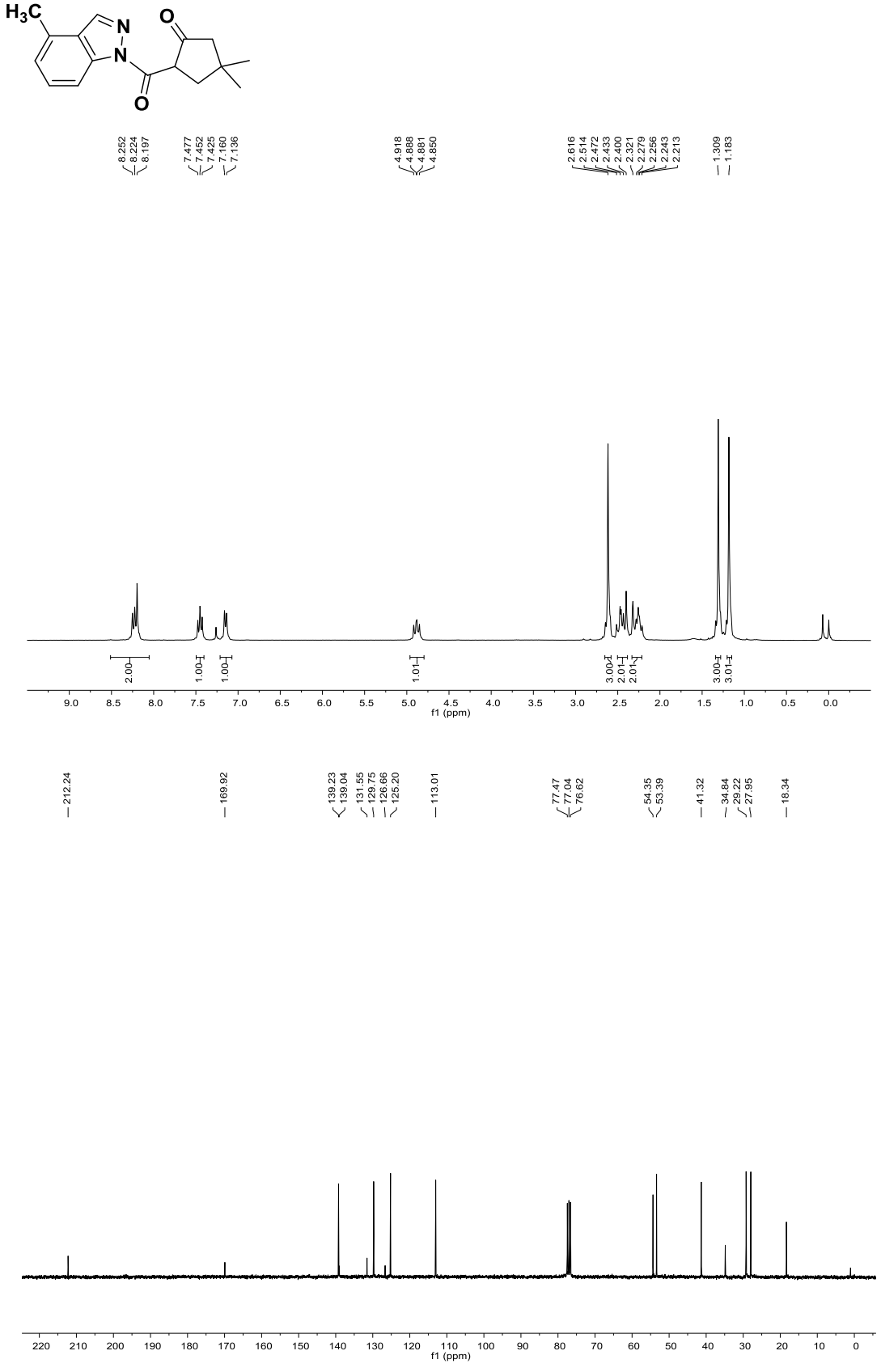
^{1}H NMR and ^{13}C NMR Spectra of Compound 8bb



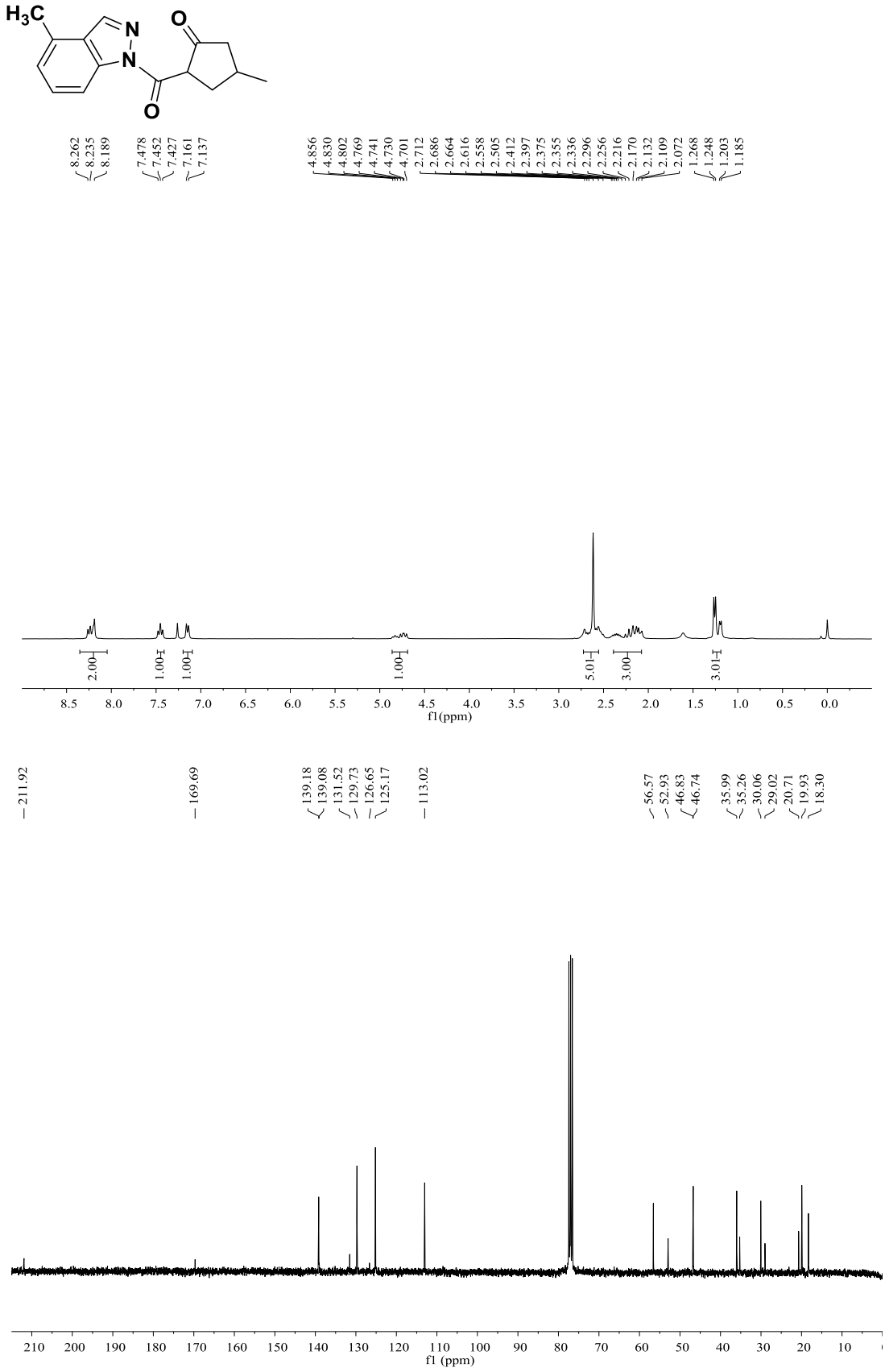




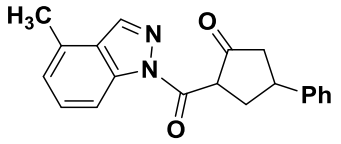
^{1}H NMR and ^{13}C NMR Spectra of Compound 10aa

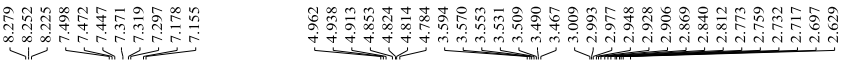


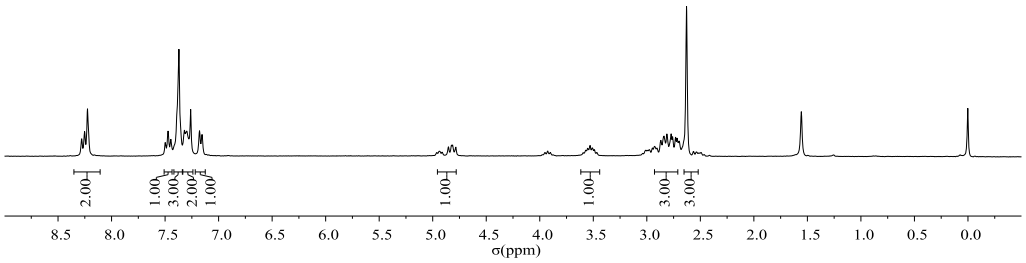
^{1}H NMR and ^{13}C NMR Spectra of Compound 10ab

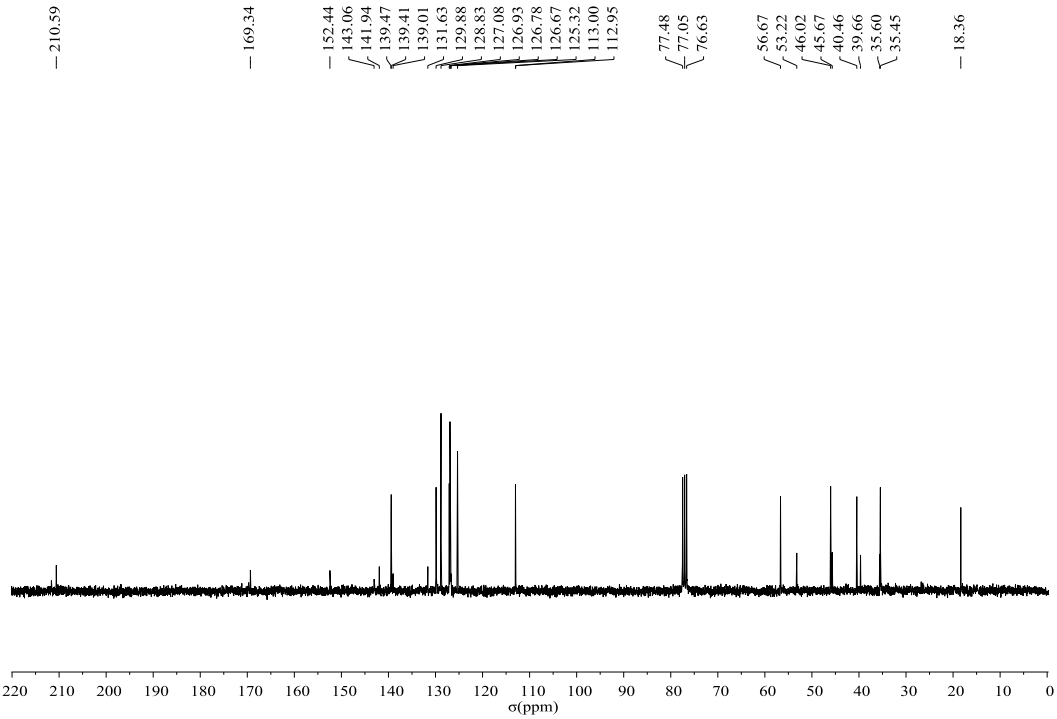


^{1}H NMR and ^{13}C NMR Spectra of Compound 10ac

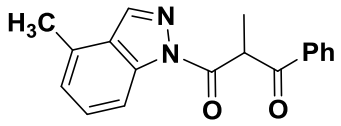








^{1}H NMR and ^{13}C NMR Spectra of Compound 11ab

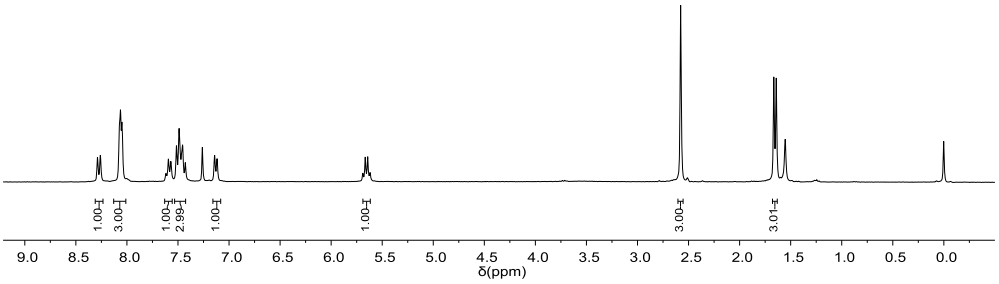


8.288
8.260
8.064
8.049
7.618
7.593
7.570
7.515
7.489
7.455
7.428
7.142
7.118

5.690
5.666
5.642
5.619

2.577

1.665
1.641



197.168
170.885
139.036
135.573
133.364
131.555
129.781
128.814
128.719
126.501
125.042
112.871
77.493
77.070
76.646
48.276
18.306
13.853