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$CrCl_2$-Catalyzed α-Alkylation of Carbonyl Compounds via Borrowing Hydrogen Approach

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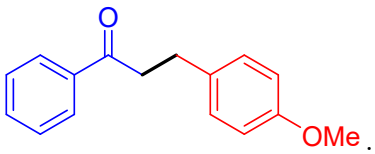
General Information. All reactions were carried out under nitrogen atmosphere in flame-dried glassware. Syringes which were used to transfer anhydrous solvents or reagents were purged with nitrogen prior to use. Toluene was continuously refluxed and freshly distilled from sodium benzophenone ketyl under nitrogen. Yields refer to isolated yields of compounds estimated to be > 95% pure as determined by ^{1}H NMR (25 °C) and capillary-GC. NMR spectra were recorded on solutions in deuterated chloroform ($CDCl_3$) with residual chloroform (δ7.25 ppm for ^{1}H NMR and δ77.0 ppm for ^{13}C NMR). Abbreviations for signal coupling are as follows: s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet, br., broad. ESI-QTOF-MS measurements were performed in the positive ion mode (m/z 50−2000 range). Column chromatographical purifications were performed using SiO_2 (200 – 300 mesh ASTM) from Branch of Qingdao Haiyang Chemical Co., Ltd if not indicated otherwise.

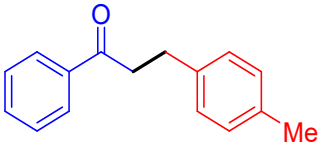
Typical procedure (TP1) for $CrCl_2$-catalyzed *α*-alkylation reaction of carbonyl compounds with alcohols. To a clean, oven-dried, screw cap reaction tube was added $CrCl_2$ (0.05 mmol, 5 mol%), PPh_3 (0.1 mmol, 10 mol%), alcohol (1.2 mmol), ketone (1 mmol), LiOH (1.2 mmol), and chlorobenznene (2 mL) under nitrogen atmosphere. The reaction mixture was stirred at 130 °C in an oil bath for 25 h. Then, the reaction mixture was diluted with water (5 mL) and extracted with EtOAc (3 × 10 mL). The resultant organic layer was dried over anhydrous $MgSO_4$ and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel column chromatography using petroleum ether/EtOAc as the eluting system.

Typical procedure (TP2) for $CrCl_2$-catalyzed dialkylation of cyclohexanone with alcohols. To a clean, oven-dried, screw cap reaction tube was added $CrCl_2$ (0.10 mmol, 10 mol%), PPh_3 (0.2 mmol, 20 mol%), alcohol (2.4 mmol), cyclohexanone (1 mmol), LiOH (2.4 mmol), and chlorobenznene (2 mL) under nitrogen atmosphere. The reaction mixture was stirred at 130 °C in an oil bath for 25 h. Then, the reaction mixture was diluted with water (5 mL) and extracted with EtOAc (3 × 10 mL). The resultant organic layer was dried over anhydrous $MgSO_4$ and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel column chromatography using petroleum ether/EtOAc as the eluting system.

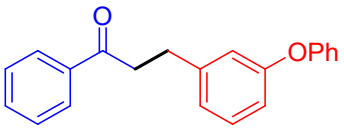
^{1}H NMR, ^{13}C NMR, HRMS and IR Data



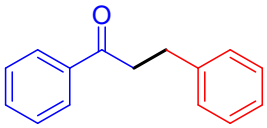
*3-(4-Methoxyphenyl)-1-phenylpropan-1-one (**3**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with (4-methoxyphenyl)methanol (166 mg, 1.2 mmol) afforded the desired product **3** as a white solid (193 mg, 80%): mp 65-66 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.96 (d, J = 7.7 Hz, 2 H), 7.55 (t, J = 7.4 Hz, 1 H), 7.45 (t, J = 7.6 Hz, 2 H), 7.18 (d, J = 8.1 Hz, 2 H), 6.86 (d, J = 8.1 Hz, 2 H), 3.78 (s, 3 H), 3.27 (t, J = 7.7 Hz, 2 H), 3.03 (t, J = 7.6 Hz, 2 H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 199.2, 157.9, 136.8, 133.2, 132.9, 129.2, 128.4, 127.9, 113.8, 55.1, 40.5, 29.1; IR (Diamond-ATR, neat) 3012, 2965, 2937, 1675, 1620, 1595, 1513, 818, 776, 523 cm^{-1}; R_f 0.33 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[1]



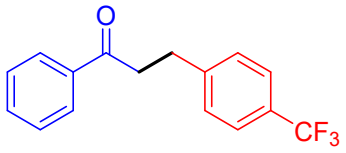
*1-Phenyl-3-(p-tolyl)propan-1-one (**4**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **4** as a pale yellow oil (184 mg, 82%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.93 (m, 2 H), 7.59–7.53 (m, 1 H), 7.52–7.40 (m, 2 H), 7.14 (q, J = 8.1 Hz, 4 H), 3.34–3.22 (m, 2H), 3.12–2.97 (m, 2H), 2.34 (s, 3 H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 199.2, 138.1, 136.8, 135.6, 132.9, 129.2, 128.5, 128.2, 127.9, 40.6, 29.7, 20.9; IR (Diamond-ATR, neat) 3021, 2922, 1680, 1514, 1449, 1362, 1292, 971, 806, 741 cm^{-1}; R_f 0.4 (petroleum ether/EtOAc, 50/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[2]



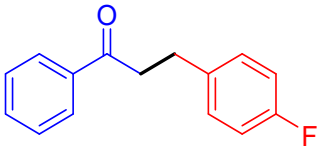
*3-(3-Phenoxyphenyl)-1-phenylpropan-1-one (**5**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 3-phenoxybenzyl alcohol (240 mg, 1.2 mmol) afforded the desired product **5** as a colorless oil (234 mg, 77%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.95–7.91 (m, 2 H), 7.60–7.50 (m, 1 H), 7.48–7.40 (m, 2 H), 7.35–7.28 (m, 2 H), 7.23 (t, J = 7.7 Hz, 1 H), 7.09 (t, J = 7.4 Hz, 1 H), 7.03–6.94 (m, 3 H), 6.91 (s, 1 H), 6.83 (dd, J = 8.1, 2.2 Hz, 1 H), 3.28 (t, J = 7.6 Hz, 2 H), 3.04 (t, J = 7.6 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.9, 157.3, 157.1, 143.3, 136.8, 133.1, 129.7, 129.6, 128.6, 128.0, 123.3, 123.2, 118.9, 118.8, 116.5, 40.1, 29.9; IR (diamond-ATR, neat) 3060, 3036, 2931, 1685, 1578, 1486, 1447, 1245, 1212, 967, 755, 689 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{21}H_{19}O_2$ [M + H]$^{+}$ 303.1385, found 303.1382; R_f 0.30 (petroleum ether/EtOAc, 30/1).



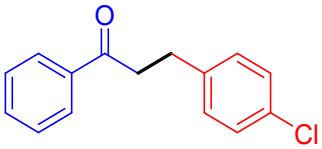
*1,3-Diphenylpropan-1-one (**6**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with benzyl alcohol (132 mg, 1.2 mmol) afforded the desired product **6** as a white solid (164 mg, 78%): mp 63-64 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.90 (m, 2H), 7.59–7.52 (m, 1H), 7.49–7.42 (m, 2H), 7.34–7.25 (m, 4H), 7.24–7.18 (m, 1H), 3.37–3.26 (m, 2H), 3.13–3.03 (m, 2H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.1, 141.2, 136.8, 133.0, 128.6, 128.5, 128.4, 127.9, 126.1, 40.4, 30.1; IR (diamond-ATR, neat) 3060, 2922, 1679, 1595, 1580, 1492, 1445, 1364, 1290, 971, 743, 689 cm^{-1}; R_f 0.41 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[3]



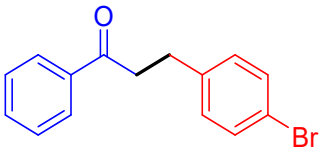
*1-Phenyl-3-(4-(trifluoromethyl)phenyl)propan-1-one (**7**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-(trifluoromethyl)benzyl alcohol (211 mg, 1.2 mmol) afforded the desired product **7** as a pale yellow oil (181 mg, 65%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.87 (m, 2H), 7.61–7.50 (m, 3H), 7.46 (dd, J = 8.4, 7.0 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 3.32 (t, J = 7.5 Hz, 2H), 3.13 (t, J = 7.5 Hz, 2H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.5, 145.4, 136.6, 133.2, 128.8, 128.6, 128.4 (q, J = 30.1 Hz), 127.9, 125.4 (q, J = 3.8 Hz), 124.2 (q, J = 270.1 Hz), 39.8, 29.7; ^{19}F NMR (376 MHz, $CDCl_3$) δ -62.4; IR (diamond-ATR, neat) 3058, 2937, 1675, 1617, 1595, 1580, 1449, 1323, 1292, 1204, 829, 743, 689 cm^{-1}; R_f 0.32 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[1]



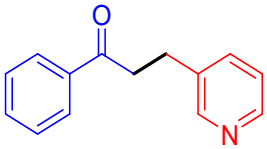
*3-(4-Fluorophenyl)-1-phenylpropan-1-one (**8**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-fluorobenzyl alcohol (151 mg, 1.2 mmol) afforded the desired product **8** as a white solid (119 mg, 52%): mp 59–60 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98–7.89 (m, 2 H), 7.62–7.50 (m, 1 H), 7.50–7.38 (m, 2 H), 7.24–7.17 (m, 2 H), 7.02-6.91 (m, 2 H), 3.28 (t, J = 7.5 Hz, 2 H), 3.04 (t, J = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.9, 161.4 (d, J = 243.9 Hz), 136.8 (d, J = 3.2 Hz), 136.8, 133.1, 129.8 (d, J = 7.8 Hz), 128.6, 127.9, 115.2 (d, J = 21.1 Hz), 40.4, 29.2; ^{19}F NMR (376 MHz, $CDCl_3$) δ -117.3; IR (diamond-ATR, neat) 3052, 2937, 1679, 1597, 1576, 1508, 1447, 1360, 1288, 1214, 973, 823, 743, 689 cm^{-1}; R_f 0.34 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[2]



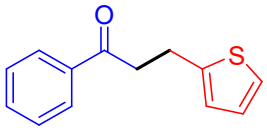
3-(4-Chlorophenyl)-1-phenylpropan-1-one ***(9)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-chlorobenzyl alcohol (171 mg, 1.2 mmol) afforded the desired product **9** as a pale yellow solid (174 mg, 71%): mp 47–49 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99–7.90 (m, 2 H), 7.60–7.52 (m, 1 H), 7.50–7.41 (m, 2 H), 7.30–7.21 (m, 2 H), 7.22–7.14 (m, 2 H), 3.27 (t, J = 7.5 Hz, 2 H), 3.04 (t, J = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.8, 139.7, 136.7, 133.1, 131.8, 129.8, 128.6, 128.5, 127.9, 40.1, 29.3; IR (diamond-ATR, neat) 3065, 2929, 1665, 1593, 1580, 1490, 1447, 1368, 1265, 1206, 981, 825, 741, 687 cm^{-1}; R_f 0.31 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[2]



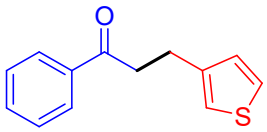
3-(4-Bromophenyl)-1-phenylpropan-1-one ***(10)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-bromobenzyl alcohol (224 mg, 1.2 mmol) afforded the desired product **10** as a white solid (211 mg, 73%): mp 59–60 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98–7.90 (m, 2 H), 7.59–7.51 (m, 1 H), 7.50–7.36 (m, 4 H), 7.15–7.06 (m, 2 H), 3.27 (t, J = 7.5 Hz, 2 H), 3.02 (t, J = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.7, 140.2, 136.7, 133.1, 131.5, 130.2, 128.6, 127.9, 119.8, 40.0, 29.4. IR (diamond-ATR, neat) 3062, 2931, 1665, 1593, 1578, 1486, 1445, 1364, 1265, 821, 776, 741, 685 cm^{-1}. R_f 0.31 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[2]



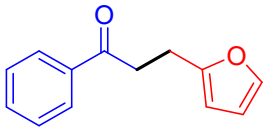
1-Phenyl-3-(pyridin-3-yl)propan-1-one ***(11)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 3-pyridinemethanol (131 mg, 1.2 mmol) afforded the desired product **11** as a white solid (108 mg, 51%): mp 86–87 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.51 (d, J = 2.2 Hz, 1 H), 8.44 (dd, J = 4.8, 1.6 Hz, 1 H), 8.01–7.85 (m, 2 H), 7.61–7.49 (m, 2 H), 7.44 (t, J = 7.6 Hz, 2 H), 7.19 (dd, J = 7.7, 4.8 Hz, 1 H), 3.30 (t, J = 7.4 Hz, 2 H), 3.06 (t, J = 7.4 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.4, 149.9, 147.6, 136.6, 136.5, 135.9, 133.2, 128.6, 127.9, 123.3, 39.7, 27.0; IR (diamond-ATR, neat) 3025, 2931, 1679, 1591, 1574, 1482, 1362, 1288, 806, 745, 689 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 1/2). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[1]



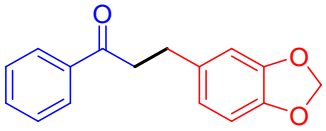
1-Phenyl-3-(thiophen-2-yl)propan-1-one ***(12)***. According to **TP1**, the reaction of acetophenone (240 mg, 2 mmol) with 2-thiophenemethanol (274 mg, 2.4 mmol) afforded the desired product **12** as a pale yellow oil (264 mg, 61%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.92 (m, 2 H), 7.61–7.52 (m, 1 H), 7.52–7.42 (m, 2 H), 7.12 (dd, *J* = 5.1, 1.2 Hz, 1 H), 6.95–6.89 (m, 1 H), 6.90–6.83 (m, 1 H), 3.36 (t, *J* = 7.5 Hz, 2 H), 3.30 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.5, 143.8, 136.7, 133.1, 128.6, 127.9, 126.8, 124.6, 123.3, 40.5, 24.2; IR (diamond-ATR, neat) 3054, 2920, 2857, 1681, 1595, 1578, 1531, 1445, 1358, 1294, 1200, 969, 829, 747, 687 cm^{-1}; R_f 0.34 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[1]



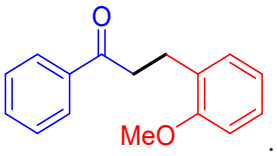
1-Phenyl-3-(thiophen-3-yl)propan-1-one ***(13)***. According to **TP1**, the reaction of acetophenone (240 mg, 2 mmol) with 3-thiophenemethanol (274 mg, 2.4 mmol) afforded the desired product **13** as a pale yellow oil (147 mg, 34%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01–7.89 (m, 2 H), 7.59–7.50 (m, 1 H), 7.49–7.36 (m, 2 H), 7.25–7.22 (m, 1 H), 7.04–6.93 (m, 2 H), 3.29 (t, *J* = 7.5 Hz, 2 H), 3.08 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.1, 141.4, 136.8, 133.1, 128.6, 128.1, 127.9, 125.6, 120.5, 39.5, 24.5. IR (diamond-ATR, neat) 3056, 2924, 2857, 1681, 1595, 1578, 1407, 1358, 835, 741, 687 cm^{-1}; R_f 0.30 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[4]



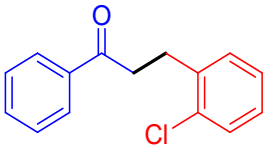
3-(Furan-2-yl)-1-phenylpropan-1-one ***(14)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 2-furanmethanol (118 mg, 1.2 mmol) afforded the desired product **14** as a pale yellow oil (60 mg, 30%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05–7.91 (m, 2 H), 7.64–7.51 (m, 1 H), 7.51–7.40 (m, 2 H), 7.30 (dd, *J* = 1.9, 0.8 Hz, 1 H), 6.28 (dd, *J* = 3.2, 1.9 Hz, 1 H), 6.05 (dd, *J* = 3.2, 1.0 Hz, 1 H), 3.33 (t, *J* = 7.5 Hz, 2 H), 3.09 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.6, 154.7, 141.1, 136.69, 133.1, 128.6, 128.0, 110.2, 105.3, 36.9, 22.5; IR (diamond-ATR, neat) 3060, 2910, 2852, 1681, 1595, 1447, 1296, 741, 689 cm^{-1}; R_f 0.20 (petroleum ether/EtOAc, 100/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[3]



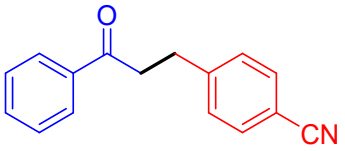
3-(Benzo[d][1,3]dioxol-5-yl)-1-phenylpropan-1-one ***(15)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 1,3-benzodioxole-5-methanol (183 mg, 1.2 mmol) afforded the desired product **11** as a white solid (211 mg, 83%): mp 56–58 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07–7.86 (m, 2 H), 7.62–7.49 (m, 1 H), 7.44 (dd, *J* = 8.3, 6.9 Hz, 2 H), 6.83–6.63 (m, 3 H), 5.90 (s, 2 H), 3.25 (t, *J* = 7.5 Hz, 2 H), 2.98 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.1, 147.5, 145.7, 136.7, 134.9, 132.9, 128.5, 127.9, 121.1, 108.8, 108.2, 100.7, 40.6, 29.8; IR (diamond-ATR, neat) 3060, 2927, 2799, 1683, 1597, 1578, 1445, 1362, 1292, 800, 747, 658 cm^{-1}; R_f 0.29 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[3]



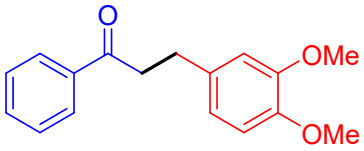
3-(2-Methoxyphenyl)-1-phenylpropan-1-one ***(16)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with (2-methoxyphenyl)methanol (166 mg, 1.2 mmol) afforded the desired product **16** as a colorless oil (144 mg, 60%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.09–7.90 (m, 2 H), 7.64–7.50 (m, 1 H), 7.51–7.40 (m, 2 H), 7.25–7.19 (m, 2 H), 6.98–6.82 (m, 2 H), 3.84 (s, 3 H), 3.28 (t, *J* = 7.5 Hz, 2 H), 3.07 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.9, 157.4, 136.9, 132.8, 130.1, 129.4, 128.4, 128.0, 127.4, 120.4, 110.1, 55.1, 38.8, 25.6; IR (diamond-ATR, neat) 3054, 2935, 2832, 1679, 1595, 1492, 1286, 736, 687 cm^{-1}; R_f 0.33 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[2]



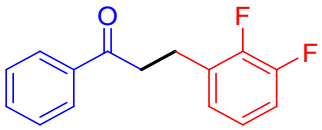
3-(2-Chlorophenyl)-1-phenylpropan-1-one ***(17)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 2-chlorobenzyl alcohol (171 mg, 1.2 mmol) afforded the desired product **17** as a pale yellow oil (162 mg, 66%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.92 (m, 2 H), 7.59–7.51 (m, 1 H), 7.49–7.41 (m, 2 H), 7.35 (dd, *J* = 7.4, 1.9 Hz, 1 H), 7.31 (dd, *J* = 7.2, 2.1 Hz, 1 H), 7.22–7.12 (m, 2 H), 3.31 (t, *J* = 7.5 Hz, 2 H), 3.17 (t, *J* = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.9, 138.8, 136.7, 133.9, 133.1, 130.8, 129.5, 128.6, 128.0, 127.7, 126.9, 38.4, 28.3; IR (diamond-ATR, neat) 3064, 2918, 1683, 1595, 1473, 1368, 1267, 747, 687 cm^{-1}; R_f 0.22 (petroleum ether/EtOAc, 100/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[5]



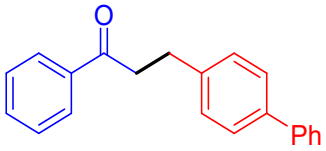
*4-(3-Oxo-3-phenylpropyl)benzonitrile (**18**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-bromobenzyl alcohol (165 mg, 1.2 mmol) afforded the desired product **18** as a white solid (83 mg, 35%): mp 86–87 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97–7.90 (m, 2 H), 7.59–7.53 (m, 3 H), 7.49–7.41 (m, 2 H), 7.39–7.33 (m, 2 H), 3.32 (t, J = 7.3 Hz, 2 H), 3.13 (t, J = 7.3 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.2, 146.9, 136.5, 133.3, 132.3, 129.3, 128.7, 127.9, 118.9, 109.9, 39.4, 29.9; IR (diamond-ATR, neat) 3060, 2920, 2852, 1677, 1597, 1578, 1447, 1336, 1212, 977, 827, 745, 687 cm^{-1}; R_f 0.30 (petroleum ether/EtOAc, 10/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[6]



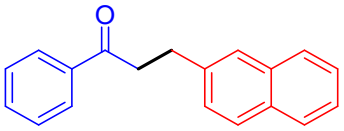
*3-(3,4-Dimethoxyphenyl)-1-phenylpropan-1-one (**19**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 3,4-dimethoxybenzyl alcohol (212 mg, 1.2 mmol) afforded the desired product **19** as a white solid (105 mg, 39%): mp 68–70 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98–7.91 (m, 2H), 7.58–7.51 (m, 1H), 7.44 (dd, J = 8.4, 7.0 Hz, 2H), 6.81–6.74 (m, 3H), 3.86 (s, 3H), 3.84 (s, 3H), 3.27 (t, J = 7.6 Hz, 2H), 3.01 (t, J = 7.6 Hz, 2H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.4, 148.9, 147.4, 136.9, 133.9, 133.0, 128.6, 128.0, 120.1, 111.8, 111.3, 55.9, 55.8, 40.7, 29.8; IR (diamond-ATR, neat) 3058, 2838, 1677, 1593, 1578, 1445, 1356, 1294, 1235, 854, 761, 745, 691 cm^{-1}; R_f 0.31 (petroleum ether/EtOAc, 5/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[7]



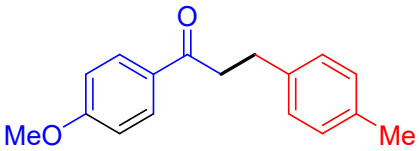
*3-(2,3-Difluorophenyl)-1-phenylpropan-1-one (**20**).* According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 2,3-difluorobenzyl alcohol (178 mg, 1.2 mmol) afforded the desired product **20** as a pale yellow oil (113 mg, 46%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00–7.89 (m, 2 H), 7.62–7.51 (m, 1 H), 7.45 (dd, J = 8.4, 7.0 Hz, 2 H), 7.08–6.91 (m, 3 H), 3.31 (t, J = 7.5 Hz, 2 H), 3.12 (t, J = 7.5 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.5, 151.1 (dd, J = 150.3, 12.8 Hz), 148.6(dd, J = 149.2, 12.8 Hz), 136.6, 133.2, 130.5(d, J = 12.4 Hz), 128.6, 127.9, 125.5 (t, J = 3.6 Hz), 123.9(dd, J = 7.0, 4.6 Hz), 115.2(d, J = 17.1 Hz), 38.5(d, J = 1.1 Hz), 23.6 (t, J = 2.6 Hz); ^{19}F NMR (376 MHz, $CDCl_3$) δ -134.70–-140.84 (m), -143.68 (dt, J = 20.9, 5.9 Hz); IR (diamond-ATR, neat) 3058, 2930, 2852, 1683, 1595, 1580, 11445, 1360, 1272, 833, 776, 743 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{15}H_{12}F_2ONa$ [M + Na]$^{+}$ 269.0754, found 269.0754; R_f 0.32 (petroleum ether/EtOAc, 20/1).



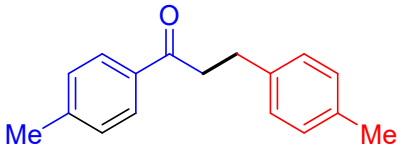
3-([1,1'-Biphenyl]-4-yl)-1-phenylpropan-1-one ***(21)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 4-phenylbenzyl alcohol (223 mg, 1.2 mmol) afforded the desired product **21** as a white solid (189 mg, 66%): mp 66–67 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.02–7.94 (m, 2 H), 7.59–7.52 (m, 5 H), 7.48–7.41 (m, 4 H), 7.35–7.31 (m, 3 H), 3.35 (t, J = 7.7 Hz, 2 H), 3.12 (t, J = 7.7 Hz, 2 H). ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.2, 140.9, 140.4, 139.1, 136.8, 133.1, 128.8, 128.7, 128.6, 128.0, 127.2, 127.1, 126.9, 40.4, 29.7; IR (diamond-ATR, neat) 3027, 2933, 1685, 1593, 1447, 1358, 827, 743, 689 cm^{-1}; R_f 0.25 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[8]



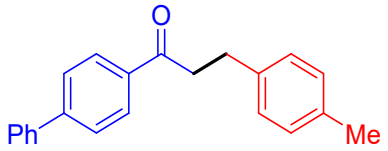
3-(Naphthalen-2-yl)-1-phenylpropan-1-one ***(22)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with naphthalen-2-ylmethanol (194 mg, 1.2 mmol) afforded the desired product **22** as a pale yellow oil (193 mg, 74%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10–8.04 (m, 1H), 8.00–7.93 (m, 2H), 7.89 (dd, J = 8.1, 1.5 Hz, 1H), 7.75 (dd, J = 5.7, 3.9 Hz, 1H), 7.58–7.47 (m, 3H), 7.47–7.37 (m, 4H), 3.56 (t, J = 7.5 Hz, 2H), 3.43 (t, J = 7.5 Hz, 2H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.2, 137.3, 136.7, 133.9, 133.1, 131.6, 128.9, 128.5, 127.9, 126.9, 126.1, 126.0, 125.6, 125.6, 123.5, 39.7, 27.1; IR (diamond-ATR, neat) 3054, 2935, 1681, 1595, 1508, 1360, 1292, 860, 776, 743, 689 cm^{-1}; R_f 0.35 (petroleum ether/EtOAc, 50/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[9]



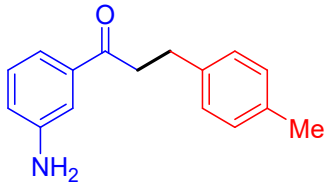
1-(4-Methoxyphenyl)-3-(p-tolyl)propan-1-one ***(23)***. According to **TP1**, the reaction of 4′-methoxyacetophenone (150 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **23** as a white solid (214 mg, 84%): mp 65–66 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98–7.91 (m, 2 H), 7.18–7.08 (m, 4 H), 6.96–6.87 (m, 2 H), 3.86 (s, 3 H), 3.23 (t, J = 7.5 Hz, 2 H), 3.02 (t, J = 7.5 Hz, 2 H), 2.32 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 197.9, 163.4, 138.3, 135.5, 130.2, 129.9, 129.1, 128.2, 113.7, 55.4, 40.2, 29.9, 20.9; IR (diamond-ATR, neat) 3013, 2964, 2914, 2842, 1667, 1601, 1574, 1469, 1414, 1251, 806, 712, 631 cm^{-1}; R_f 0.27 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[10]



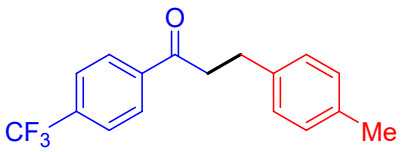
1,3-Bis(4-methylphenyl)-1-propanone ***(24)***. According to **TP1**, the reaction of 4′-methylacetophenone (134 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **24** as a white solid (203 mg, 85%): mp 133–135 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.78 (d, *J* = 8.2 Hz, 2 H), 7.19–7.12 (m, 2 H), 7.10–6.99 (m, 4 H), 3.17 (t, *J* = 7.5 Hz, 2 H), 2.94 (t, *J* = 7.5 Hz, 2 H), 2.32 (s, 3 H), 2.24 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.9, 143.7, 138.2, 135.5, 134.4, 129.2, 129.1, 128.2, 128.1, 40.5, 29.7, 21.6, 20.9; IR (diamond-ATR, neat) 3023, 2945, 1677, 1603, 1572, 1532, 1366, 1265, 800, 720 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{17}H_{19}O$ $[M + H]^{+}$ 239.1436, found 239.1429; R_f 0.51 (petroleum ether/EtOAc, 30/1).



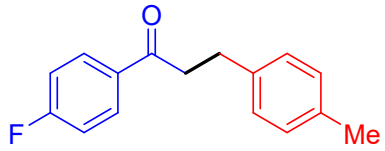
1-([1,1′-Biphenyl]-4-yl)-3-(p-tolyl)propan-1-one ***(25)***. According to **TP1**, the reaction of 4-acetylbiphenyl (200 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **25** as a white solid (243 mg, 81%): mp 120–121 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07–8.00 (m, 2 H), 7.71–7.65 (m, 2 H), 7.62 (dt, *J* = 6.1, 1.3 Hz, 2 H), 7.47 (dd, *J* = 8.3, 6.6 Hz, 2 H), 7.44–7.36 (m, 1 H), 7.19–7.10 (m, 4 H), 3.31 (t, *J* = 7.7 Hz, 2 H), 3.05 (t, *J* = 7.7 Hz, 2 H), 2.33 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.9, 145.7, 139.8, 138.2, 135.6, 135.5, 129.2, 128.9, 128.6, 128.3, 128.2, 127.2, 127.2, 40.7, 29.8, 21.0; IR (diamond-ATR, neat) 3054, 2912, 1669, 1601, 1560, 1447, 1323, 1267, 811, 767 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{22}H_{20}ONa$ $[M + Na]^{+}$ 323.1412, found 323.1415; R_f 0.50 (petroleum ether/EtOAc, 30/1).



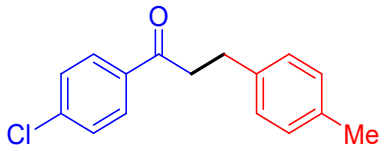
1-(3-Aminophenyl)-3-(p-tolyl)propan-1-one ***(26)***. According to **TP1**, the reaction of 3′-aminoacetophenone (138 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **26** as a yellow solid (96 mg, 40%): mp 64–65 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.42 (dt, *J* = 7.7, 1.3 Hz, 1 H), 7.38–7.34 (m, 1 H), 7.31 (t, *J* = 7.8 Hz, 1 H), 7.27–7.18 (m, 4 H), 7.00–6.88 (m, 1 H), 3.88 (s, 2 H), 3.33 (t, *J* = 7.5 Hz, 2 H), 3.10 (t, *J* = 7.5 Hz, 2 H), 2.42 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.5, 146.7, 138.2, 137.9, 135.5, 129.4, 129.1, 128.2, 119.5, 118.4, 113.9, 40.6, 29.7, 20.9; IR (diamond-ATR, neat) 3441, 3353, 3054, 2918, 2850, 1673, 1624, 1601, 1587, 1514, 1455, 1368, 1321, 1282, 992, 883, 821, 780, 683 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{16}H_{18}NO$ $[M + H]^{+}$ 240.1388, found 240.1380; R_f 0.31 (petroleum ether/EtOAc, 10/1).



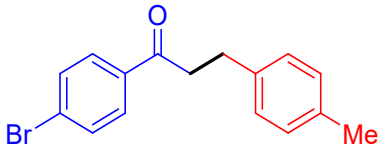
*3-(p-Tolyl)-1-(4-(trifluoromethyl)phenyl)propan-1-one (**27**).* According to **TP1**, the reaction of 4-(trifluoromethyl)acetophenone (188 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **27** as a yellow solid (208 mg, 71%): mp 69–70 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05 (d, *J* = 8.1 Hz, 2 H), 7.71 (d, *J* = 8.1 Hz, 2 H), 7.19–7.05 (m, 4 H), 3.30 (t, *J* = 7.6 Hz, 2 H), 3.05 (t, *J* = 7.6 Hz, 2 H), 2.33 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.3, 139.5, 137.7, 135.8, 134.3 (q, *J* = 32.7 Hz), 129.2, 128.3, 128.2, 125.6 (q, *J* = 3.7 Hz), 123.6 (q, *J* = 272.7 Hz), 40.9, 29.5, 20.9; ^{19}F NMR (376 MHz, $CDCl_3$) δ -63.07; IR (diamond-ATR, neat) 3023, 2920, 2869, 1680, 1578, 1407, 1321, 1267, 808, 693 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{17}H_{16}F_3O$ [M + H]$^{+}$ 293.1153, found 293.1151; R_f 0.30 (petroleum ether/EtOAc, 50/1).



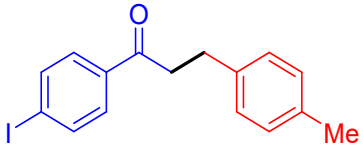
*1-(4-Fluorophenyl)-3-(p-tolyl)propan-1-one (**28**).* According to **TP1**, the reaction of 4′-fluoroacetophenone (138 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **28** as a yellow solid (196 mg, 81%): mp 48–50 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.03–7.91 (m, 2 H), 7.17–7.06 (m, 6 H), 3.24 (t, *J* = 7.7 Hz, 2 H), 3.02 (t, *J* = 7.7 Hz, 2 H), 2.32 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 197.7, 165.7 (d, *J* = 254.6 Hz), 138.0, 135.7, 133.3 (d, *J* = 2.9 Hz), 130.6 (d, *J* = 9.3 Hz), 129.2, 128.3, 115.6 (d, *J* = 21.9 Hz), 40.5, 29.7, 20.9; ^{19}F NMR (376 MHz, $CDCl_3$) δ -105.38; IR (diamond-ATR, neat) 3023, 2920, 2857, 1679, 1650, 1506, 1434, 1364, 1296, 841, 720, 693 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{16}H_{15}FONa$ [M + Na]$^{+}$ 265.1005, found 265.1001; R_f 0.33 (petroleum ether/EtOAc, 50/1).



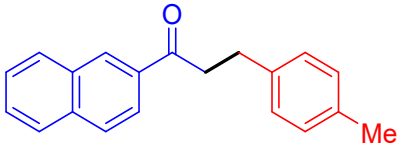
*1-(4-Chlorophenyl)-3-(p-tolyl)propan-1-one (**29**).* According to **TP1**, the reaction of 4′-chloroacetophenone (155 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **29** as a yellow solid (176 mg, 68%): mp 76–77 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99–7.79 (m, 2 H), 7.49–7.35 (m, 2 H), 7.20–7.03 (m, 4 H), 3.24 (t, *J* = 7.7 Hz, 2 H), 3.01 (t, *J* = 7.7 Hz, 2 H), 2.32 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.1, 139.4, 137.9, 135.7, 135.2, 129.4, 129.2, 128.9, 128.2, 40.6, 29.6, 20.9; IR (diamond-ATR, neat) 3023, 2916, 1677, 1587, 1512, 1426, 1397, 1319, 1200, 806, 745, 689 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{16}H_{15}ClO$ [M + Na]$^{+}$ 281.0709, found 281.0708; R_f 0.30 (petroleum ether/EtOAc, 50/1).



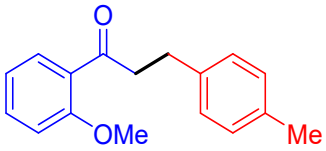
1-(4-Bromophenyl)-3-(p-tolyl)propan-1-one ***(30)***. According to **TP1**, the reaction of 4′-bromoacetophenone (199 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **30** as a yellow solid (243 mg, 80%): mp 84–85 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.89–7.74 (m, 2 H), 7.66–7.52 (m, 2 H), 7.18–7.03 (m, 4 H), 3.24 (t, J = 7.6 Hz, 2 H), 3.02 (t, J = 7.6 Hz, 2 H), 2.32 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.2, 137.9, 135.7, 135.5, 131.9, 129.5, 129.2, 128.2, 128.1, 40.5, 29.6, 20.9; IR (diamond-ATR, neat) 3021, 2916, 2857, 1673, 1601, 1582, 1512, 1482, 1395, 1267, 1200, 804, 720, 693 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[11]



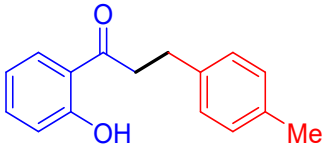
1-(4-Iodophenyl)-3-(p-tolyl)propan-1-one ***(31)***. According to **TP1**, the reaction of 4′-iodoacetophenone (246 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **31** as a white solid (267 mg, 76%): mp 108–110 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.87–7.75 (m, 2 H), 7.70–7.60 (m, 2 H), 7.16–7.06 (m, 4 H), 3.22 (t, J = 7.6 Hz, 2 H), 3.01 (t, J = 7.6 Hz, 2 H), 2.31 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.6, 137.9, 137.8, 136.1, 135.7, 129.4, 129.2, 128.2, 100.9, 40.5, 29.6, 20.9. IR (diamond-ATR, neat) 3023, 2914, 2855, 1671, 1648, 1576, 1512, 1436, 1364, 1270, 1200, 802, 693, 668 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{16}H_{16}IO$ $[M + H]^{+}$ 351.0246, found 351.0244; R_f 0.32 (petroleum ether/EtOAc, 50/1).



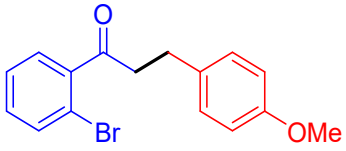
1-(Naphthalen-2-yl)-3-(p-tolyl)propan-1-one ***(32)***. According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **32** as a white solid (233 mg, 85%): mp 97–98 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.47 (s, 1 H), 8.06 (dd, J = 8.6, 1.8 Hz, 1 H), 7.96–7.85 (m, 3 H), 7.63–7.53 (m, 2 H), 7.24–7.13 (m, 4 H), 3.43 (t, J = 7.7 Hz, 2 H), 3.11 (t, J = 7.7 Hz, 2 H), 2.36 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.1, 138.2, 135.6, 135.5, 134.1, 132.4, 129.6, 129.5, 129.2, 128.4, 128.3, 128.3, 127.7, 126.7, 123.8, 40.7, 29.8, 20.9; IR (diamond-ATR, neat) 3020, 2951, 2852, 1673, 1624, 1591, 1467, 1364, 1270, 870, 738, 695 cm^{-1}; R_f 0.30 (petroleum ether/EtOAc, 50/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[12]



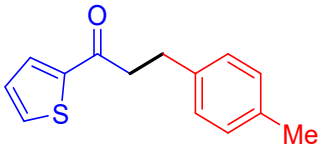
*1-(2-Methoxyphenyl)-3-(p-tolyl)propan-1-one (**33**).* According to **TP1**, the reaction of 2′-methoxyacetophenone (150 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **33** as a pale yellow solid (99 mg, 39%): mp 69–71 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.70 (dd, *J* = 7.7, 1.8 Hz, 1 H), 7.51–7.41 (m, 1 H), 7.20–7.07 (m, 4 H), 7.04–6.94 (m, 2 H), 3.88 (s, 3 H), 3.29 (t, *J* = 7.6 Hz, 2 H), 2.99 (t, *J* = 7.6 Hz, 2 H), 2.33 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 201.8, 158.5, 138.6, 135.3, 133.3, 130.3, 129.0, 128.3, 120.6, 111.4, 55.4, 45.6, 30.0, 20.9; IR (diamond-ATR, neat) 3019, 2960, 1667, 1593, 1512, 1482, 1393, 1288, 806, 757, 638 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{17}H_{19}O_2$ [M + H]$^{+}$ 255.1385, found 255.1380; R_f 0.28 (petroleum ether/EtOAc, 30/1).



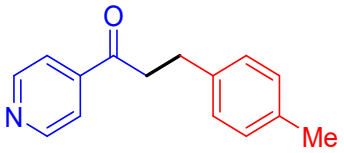
*1-(2-Hydroxyphenyl)-3-(p-tolyl)propan-1-one (**34**).* According to **TP1**, the reaction of 2′-hydroxyacetophenone (136 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **34** as a pale yellow oil (111 mg, 46%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 12.34 (s, 1 H), 7.75 (dd, *J* = 8.0, 1.6 Hz, 1 H), 7.47 (ddd, *J* = 8.6, 7.2, 1.6 Hz, 1 H), 7.20–7.10 (m, 4 H), 7.00 (dd, *J* = 8.4, 1.1 Hz, 1 H), 6.93–6.84 (m, 1 H), 3.31 (t, *J* = 7.7 Hz, 2 H), 3.04 (t, *J* = 7.7 Hz, 2 H), 2.34 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 205.4, 162.4, 137.6, 136.3, 135.8, 129.8, 129.2, 128.2, 119.2, 118.9, 118.5, 40.1, 29.6, 20.9; IR (diamond-ATR, neat) 3326, 3023, 2920, 1636, 1613, 1582, 1442, 1360, 1282, 813, 751, 654 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[13]



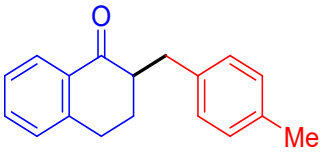
*1-(2-Bromophenyl)-3-(4-methoxyphenyl)propan-1-one (**35**).* According to **TP1**, the reaction of 2′-bromoacetophenone (199 mg, 1 mmol) with 4-methoxyphenylmethanol (167 mg, 1.2 mmol) afforded the desired product **35** as a pale yellow oil (32 mg, 10%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.58 (dd, J = 7.9, 1.2 Hz, 1 H), 7.36–7.25 (m, 3 H), 7.16–7.11 (m, 2 H), 6.85–6.79 (m, 2 H), 3.77 (s, 3 H), 3.20 (t, *J* = 7.6 Hz, 2 H), 2.99 (t, *J* = 7.6 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 203.5, 158.0, 141.7, 133.6, 132.7, 131.5, 129.3, 128.4, 127.4, 118.6, 113.9, 55.3, 44.6, 29.2; IR (diamond-ATR, neat) 3060, 2930, 2834, 1698, 1584, 1428, 1358, 1245, 825, 755, 671 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{16}H_{16}BrO_2$ [M + H]$^{+}$ 319.0334, found 319.0332; R_f 0.28 (petroleum ether/EtOAc, 20/1).



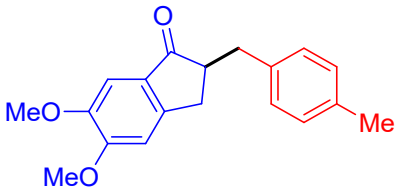
1-(Thiophen-2-yl)-3-(p-tolyl)propan-1-one ***(36)***. According to **TP1**, the reaction of 2-acetylthiophene (126 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **36** as a pale yellow oil (175 mg, 76%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (dd, *J* = 3.8, 1.1 Hz, 1 H), 7.61 (dd, *J* = 4.9, 1.1 Hz, 1 H), 7.20–7.03 (m, 5 H), 3.21 (t, *J* = 7.6 Hz, 2 H), 3.03 (t, *J* = 7.6 Hz, 2 H), 2.32 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 192.2, 144.2, 137.9, 135.7, 133.5, 131.7, 129.2, 128.3, 128.0, 41.3, 29.9, 20.9; IR (diamond-ATR, neat) 3019, 2920, 1661, 1514, 1414, 1292, 852, 724, 652 cm^{-1}; R_f 0.33 (petroleum ether/EtOAc, 50/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[14]



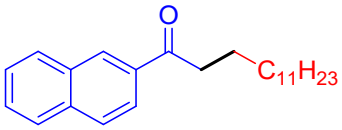
1-(Pyridin-4-yl)-3-(p-tolyl)propan-1-one ***(37)***. According to **TP1**, the reaction of 4-acetylpyridine (121 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **37** as a pale yellow solid (43 mg, 19%): mp 63–64 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.83–8.74 (m, 2 H), 7.74–7.65 (m, 2 H), 7.16–7.06 (m, 4 H), 3.27 (t, *J* = 7.6 Hz, 2 H), 3.02 (t, *J* = 7.6 Hz, 2 H), 2.31 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 198.7, 150.9, 142.6, 137.5, 135.9, 129.3, 128.2, 120.9, 40.8, 29.3, 20.9; IR (diamond-ATR, neat) 3027, 2916, 2855, 1685, 1650, 1556, 1370, 1267, 800, 648 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{15}H_{16}NO$ $[M + H]^{+}$ 226.1232, found 226.1230; R_f 0.32 (petroleum ether/EtOAc, 3/1).



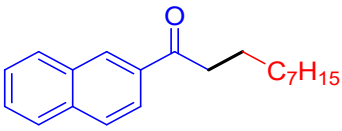
2-(4-Methylbenzyl)-3,4-dihydronaphthalen-1(2H)-one ***(38)***. According to **TP1**, the reaction of 1-tetralone (146 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **38** as a pale yellow solid (205 mg, 82%): mp 67–69 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (dd, *J* = 7.9, 1.4 Hz, 1 H), 7.46 (td, *J* = 7.4, 1.4 Hz, 1 H), 7.32 (t, *J* = 7.5 Hz, 1 H), 7.22 (d, *J* = 7.7 Hz, 1 H), 7.17–7.09 (m, 4 H), 3.46 (dd, *J* = 13.7, 4.0 Hz, 1 H), 2.93 (dt, *J* = 9.8, 4.7 Hz, 2 H), 2.77–2.69 (m, 1 H), 2.62 (dd, *J* = 13.7, 9.6 Hz, 1 H), 2.34 (s, 3 H), 2.12 (dq, *J* = 13.4, 4.5 Hz, 1 H), 1.79 (dddd, *J* = 13.4, 11.5, 10.2, 5.4 Hz, 1 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.4, 143.9, 136.8, 135.5, 133.2, 132.4, 129.1, 129.0, 128.6, 127.5, 126.5, 49.4, 35.2, 28.5, 27.6, 20.9; IR (diamond-ATR, neat) 3019, 2920, 2850, 1679, 1510, 1484, 1360, 1286, 802, 757, 720, 623 cm^{-1}; R_f 0.52 (petroleum ether/EtOAc, 30/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[12]



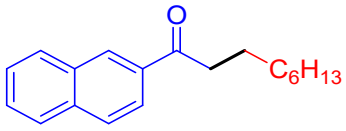
*5,6-Dimethoxy-2-(4-methylbenzyl)-2,3-dihydro-1H-inden-1-one (**39**).* According to **TP1**, the reaction of 5,6-dimethoxy-1-indanone (192 mg, 1 mmol) with *p*-tolylmethanol (147 mg, 1.2 mmol) afforded the desired product **39** as a white solid (154 mg, 52%): mp 134–135 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.18 (s, 1 H), 7.15–7.04 (m, 4 H), 6.79 (s, 1 H), 3.92 (s, 3 H), 3.89 (s, 3 H), 3.31 (dd, *J* = 13.9, 4.2 Hz, 1 H), 3.04 (dd, *J* = 16.8, 7.4 Hz, 1 H), 2.99–2.90 (m, 1 H), 2.74 (dd, *J* = 16.8, 3.2 Hz, 1 H), 2.59 (dd, *J* = 13.9, 10.3 Hz, 1 H), 2.30 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 206.5, 155.5, 149.4, 148.9, 136.6, 135.7, 129.2, 129.1, 128.7, 107.3, 104.3, 56.1, 56.0, 49.1, 36.8, 31.8, 20.9; IR (diamond-ATR, neat) 3015, 2920, 2850, 1681, 1591, 1463, 1434, 1364, 1216, 874, 837, 780, 650 cm^{-1}; R_f 0.25 (petroleum ether/EtOAc, 5/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[15]



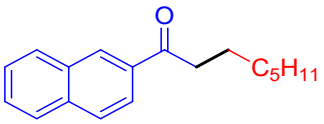
*1-(Naphthalen-2-yl)tetradecan-1-one (**40**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 1-dodecanol (224 mg, 1.2 mmol) afforded the desired product **40** as a pale yellow solid (122 mg, 36%): mp 81–82 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, *J* = 8.7, 1.7 Hz, 1 H), 7.96 (d, *J* = 8.0 Hz, 1 H), 7.91–7.84 (m, 2 H), 7.56 (dt, *J* = 18.6, 7.0 Hz, 2 H), 3.09 (t, *J* = 7.4 Hz, 2 H), 1.79 (p, *J* = 7.4 Hz, 2 H), 1.4 –1.23 (m, 20 H), 0.87 (t, *J* = 6.6 Hz, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 200.6, 135.5, 134.4, 132.5, 129.6, 129.5, 128.4, 128.3, 127.7, 126.7, 123.9, 38.7, 31.9, 29.7, 29.7, 29.6, 29.6, 29.5, 29.5, 29.4, 29.4, 24.6, 22.7, 14.1; IR (diamond-ATR, neat) 3054, 2953, 2846, 1683, 1626, 1574, 1434, 1375, 1224, 1195, 942, 854, 751 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{24}H_{34}ONa$ [M + Na]$^{+}$ 361.2507, found 361.2507; R_f 0.33 (petroleum ether/EtOAc, 100/1).



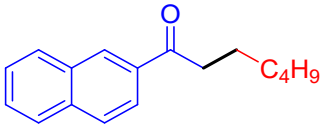
*1-(Naphthalen-2-yl)decan-1-one (**41**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 1-octanol (156 mg, 1.2 mmol) afforded the desired product **41** as a pale yellow solid (136 mg, 48%): mp 48–50 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, *J* = 8.7, 1.8 Hz, 1 H), 7.96 (d, *J* = 7.8 Hz, 1 H), 7.93–7.83 (m, 2 H), 7.65–7.47 (m, 2 H), 3.08 (t, *J* = 7.4 Hz, 2 H), 1.79 (p, *J* = 7.4 Hz, 2 H), 1.45–1.25 (m, 12 H), 0.93–0.79 (m, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 200.6, 135.5, 134.4, 132.5, 129.6 129.5, 128.4, 128.3, 127.7, 126.7, 123.9, 38.7, 31.9, 29.5, 29.5, 29.4, 29.3, 24.6, 22.7, 14.1; IR (diamond-ATR, neat) 3059, 2957, 2848, 1667, 1628, 1578, 1467, 1372, 1280, 817, 749 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{20}H_{26}ONa$ [M + Na]$^{+}$ 305.1881, found 305.1875; R_f 0.34 (petroleum ether/EtOAc, 100/1).



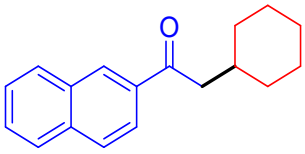
*1-(Naphthalen-2-yl)nonan-1-one (**42**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 1-heptanol (140 mg, 1.2 mmol) afforded the desired product **42** as a pale yellow solid (140 mg, 52%): mp 49–50 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, *J* = 8.7, 1.8 Hz, 1 H), 7.96 (d, *J* = 8.0 Hz, 1 H), 7.92–7.83 (m, 2 H), 7.62–7.50 (m, 2 H), 3.08 (t, *J* = 7.4 Hz, 2 H), 1.79 (p, J = 7.4 Hz, 2 H), 1.43–1.25 (m, 10 H), 0.95–0.79 (m, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 200.6, 135.5, 134.4, 132.5, 129.6, 129.5, 128.4, 128.3, 127.7, 126.7, 123.9, 38.7, 31.8, 29.5, 29.4, 29.2, 24.6, 22.7, 14.1. IR (diamond-ATR, neat) 3058, 2846, 1681, 1626, 1504, 1434, 1276, 942, 802, 753 cm^{-1}. R_f 0.34 (petroleum ether/EtOAc, 100/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[16]



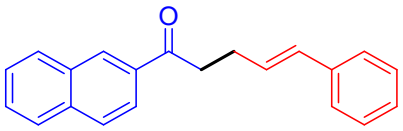
*1-(Naphthalen-2-yl)octan-1-one (**43**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 1-hexanol (123 mg, 1.2 mmol) afforded the desired product **43** as a pale yellow solid (142 mg, 56%): mp 51–53 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, *J* = 8.6, 1.7 Hz, 1 H), 7.96 (d, *J* = 8.0 Hz, 1 H), 7.90–7.85 (m, 2 H), 7.61–7.52 (m, 2 H), 3.12–3.03 (m, 2 H), 1.84–1.74 (m, 2 H), 1.42–1.28 (m, 8 H), 0.89 (t, *J* = 6.9 Hz, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 200.5, 135.5, 134.4, 132.5, 129.6, 129.5, 128.3, 128.2, 127.7, 126.7, 123.9, 38.7, 31.7, 29.4, 29.2, 24.5, 22.6, 14.1; IR (diamond-ATR, neat) 3054, 2848, 1667, 1628, 1578, 1467, 1372, 1280, 940, 821, 747 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{18}H_{22}ONa$ [M + Na]$^{+}$ 277.1568, found 277.1567; R_f 0.28 (petroleum ether/EtOAc, 100/1).



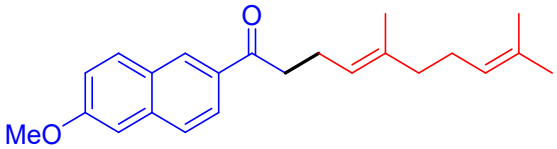
*1-(Naphthalen-2-yl)heptan-1-one (**44**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 1-pentanol (106 mg, 1.2 mmol) afforded the desired product **44** as a pale yellow solid (60 mg, 25%): mp 55–56 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, *J* = 8.6, 1.8 Hz, 1 H), 7.96 (d, *J* = 7.5 Hz, 1 H), 7.90–7.84 (m, 2 H), 7.61–7.51 (m, 2 H), 3.09 (t, *J* = 7.4 Hz, 2 H), 1.85–1.72 (m, 2 H), 1.48–1.27 (m, 6 H), 0.90 (t, *J* = 7.0 Hz, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 200.5, 135.5, 134.4, 132.5, 129.6, 129.5, 128.4, 128.3, 127.7, 126.7, 123.9, 38.7, 31.7, 29.1, 24.5, 22.5, 14.0; IR (diamond-ATR, neat) 3060, 2855, 1680, 1626, 1593, 1465, 1364, 1288, 963, 817, 794, 747 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{17}H_{20}ONa$ [M + Na]$^{+}$ 263.1412, found 263.1413; R_f 0.27 (petroleum ether/EtOAc, 100/1).



2-Cyclohexyl-1-(naphthalen-2-yl)ethan-1-one ***(45)***. According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with cyclohexanol (120 mg, 1.2 mmol) afforded the desired product **45** as a colorless oil (36 mg, 14%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.45 (s, 1 H), 8.02 (dd, *J* = 8.6, 1.8 Hz, 1 H), 7.96 (d, *J* = 8.0 Hz, 1 H), 7.92–7.83 (m, 2 H), 7.64–7.49 (m, 2 H), 2.95 (d, *J* = 6.8 Hz, 2 H), 2.13–1.95 (m, 1 H), 1.87–1.75 (m, 2 H), 1.74–1.68 (m, 2 H), 1.63–1.60 (m, 1 H), 1.32–1.25 (m, 2 H), 1.22–1.13 (m, 1 H), 1.13–0.99 (m, 2 H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 200.3, 135.5, 134.8, 132.5, 129.7, 129.5, 128.4, 128.3, 127.7, 126.7, 124.0, 46.3, 34.8, 33.5, 26.3, 26.2; IR (diamond-ATR, neat) 3056, 2850, 1677, 1626, 1434, 1354, 1278, 823, 745, 693 cm^{-1}; R_f 0.31 (petroleum ether/EtOAc, 100/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[17]

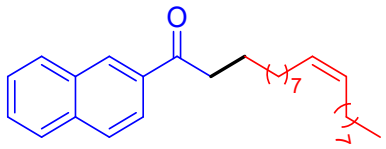


(E)-1-(Naphthalen-2-yl)-5-phenylpent-4-en-1-one ***(46)***. According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with cinnamyl alcohol (161 mg, 1.2 mmol) afforded the desired product **46** as a pale yellow solid (106 mg, 37%): mp 74–76 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.49 (s, 1 H), 8.05 (dd, *J* = 8.6, 1.8 Hz, 1 H), 7.96 (d, *J* = 8.0 Hz, 1 H), 7.93–7.84 (m, 2 H), 7.65–7.50 (m, 2 H), 7.35 (d, *J* = 7.2 Hz, 2 H), 7.32–7.25 (m, 2 H), 7.22–7.16 (m, 1 H), 6.50 (d, *J* = 15.8 Hz, 1 H), 6.34 (dt, *J* = 15.8, 6.8 Hz, 1 H), 3.29 (t, *J* = 7.4 Hz, 2 H), 2.72 (dd, *J* = 7.8, 7.3 Hz, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.3, 137.4, 135.6, 134.2, 132.5, 130.8, 129.7, 129.5, 129.1, 128.5, 128.5, 128.4, 127.8, 127.1, 126.8, 126.0, 123.8, 38.3, 27.6; IR (diamond-ATR, neat) 3062, 3021, 2846, 1679, 1624, 1553, 1492, 1368, 1274, 963, 815, 743, 693 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[18]

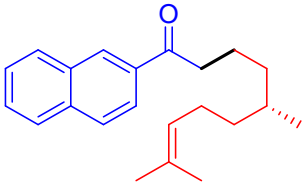


(E)-1-(6-Methoxynaphthalen-2-yl)-5,9-dimethyldeca-4,8-dien-1-one ***(47)***. According to **TP1**, the reaction of 2-acetyl-6-methoxynaphthalene (200 mg, 1 mmol) with geraniol (185 mg, 1.2 mmol) afforded the desired product **47** as a colorless oil (64 mg, 19%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.40 (d, *J* = 2.1 Hz, 1 H), 8.00 (dd, *J* = 8.6, 1.8 Hz, 1 H), 7.84 (d, *J* = 8.9 Hz, 1 H), 7.76 (d, *J* = 8.6 Hz, 1 H), 7.19 (dd, *J* = 8.9, 2.5 Hz, 1 H), 7.14 (d, *J* = 2.5 Hz, 1 H), 5.22 (ddt, *J* = 7.2, 5.9, 1.3 Hz, 1 H), 5.08 (tt, *J* = 6.9, 1.4 Hz, 1 H), 3.94 (s, 3 H), 3.10 (dd, *J* = 8.1, 7.0 Hz, 2 H), 2.47 (q, *J* = 7.4 Hz, 2 H), 2.16–1.85 (m, 4 H), 1.66 (s, 3 H), 1.64 (s, 3 H), 1.58 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 199.8, 159.6, 137.2, 136.4, 132.5, 131.4, 131.1, 129.5, 127.8, 127.1, 124.7, 124.2, 122.9, 119.7, 105.7, 55.4, 39.7, 38.7, 26.7, 25.7, 23.1, 17.7, 16.1; IR (diamond-ATR, neat) 3056, 2852, 1673,

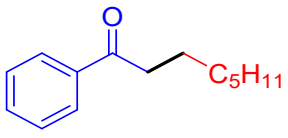
1624, 1576, 1438, 1387, 854, 745 cm^{-1}. HRMS (ESI^+) calcd for $C_{23}H_{29}O_2$ $[M + H]^+$ 337.2168, found 337.2161. R_f 0.50 (petroleum ether/EtOAc, 50/1).



(Z)-1-(Naphthalen-2-yl)icos-11-en-1-one ***(48)***. According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with oleyl alcohol (322 mg, 1.2 mmol) afforded the desired product **48** as a yellow oil (211 mg, 50%): ^1H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, J = 8.6, 1.7 Hz, 1 H), 7.96 (d, J = 8.0 Hz, 1 H), 7.91–7.84 (m, 2 H), 7.62–7.51 (m, 2 H), 5.41–5.27 (m, 2 H), 3.09 (t, J = 7.4 Hz, 2 H), 2.06–1.93 (m, 4 H), 1.83–1.73 (m, 2 H), 1.34–1.20 (m, 24 H), 0.87 (t, J = 6.7 Hz, 3 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 200.6, 135.5, 134.4, 132.5, 129.9, 129.8, 129.6, 129.5, 128.4, 128.3, 127.7, 126.7, 123.9, 38.7, 31.9, 29.8, 29.5, 29.5, 29.4, 29.3, 29.3, 27.2, 24.6, 22.7, 14.1; IR (diamond-ATR, neat) 3058, 2852, 1681, 1622, 1463, 1368, 1276, 858, 745 cm^{-1}; HRMS (ESI^+) calcd for $C_{30}H_{45}O$ $[M + H]^+$ 421.3470, found 421.3466. R_f 0.31 (petroleum ether/EtOAc, 50/1).

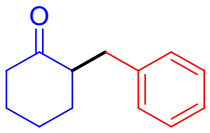


(S)-5,9-Dimethyl-1-(naphthalen-2-yl)dec-8-en-1-one ***(49)***. According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with (S)-(-)-β-citronellol (188 mg, 1.2 mmol) afforded the desired product **49** as a yellow oil (158 mg, 51%): ^1H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1 H), 8.03 (dd, J = 8.6, 1.7 Hz, 1 H), 7.96 (d, J = 8.0 Hz, 1 H), 7.91–7.84 (m, 2 H), 7.64–7.49 (m, 2 H), 5.16–5.03 (m, 1 H), 3.07 (t, J = 7.4 Hz, 2 H), 2.06–1.89 (m, 2 H), 1.89–1.70 (m, 2 H), 1.68 (s, 3 H), 1.60 (s, 3 H), 1.52–1.32 (m, 3 H), 1.28–1.16 (m, 2 H), 0.91 (d, J = 6.5 Hz, 3 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 200.5, 135.5, 134.4, 132.5, 131.1, 129.6, 129.5, 128.3, 128.2, 127.7, 126.7, 124.9, 123.9, 38.9, 36.9, 36.6, 32.3, 25.7, 25.5, 22.0, 19.5, 17.6; IR (diamond-ATR, neat) 3062, 2912, 2850, 1679, 1626, 1595, 1455, 1375, 1276, 858, 819, 745, 695 cm^{-1}; HRMS (ESI^+) calcd for $C_{22}H_{29}O$ $[M + H]^+$ 309.2218, found 309.2213; R_f 0.33 (petroleum ether/EtOAc, 50/1).

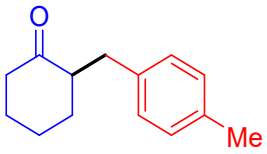


1-Phenyloctan-1-one ***(50)***. According to **TP1**, the reaction of acetophenone (120 mg, 1 mmol) with 1-hexanol (123 mg, 1.2 mmol) afforded the desired product **50** as a colorless oil (46 mg, 23%): ^1H NMR (400 MHz, $CDCl_3$) δ 7.98–7.92 (m, 2 H), 7.58–7.51 (m, 1 H), 7.48–7.42 (m, 2 H), 3.01–2.90 (m, 2 H), 1.78–1.68 (m, 2 H), 1.36–1.25 (m, 8 H), 0.89–0.84 (m, 3 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 200.6, 137.1, 132.8, 128.5, 128.0, 38.6, 31.7, 29.3, 29.1, 24.4, 22.6, 14.0; IR (diamond-ATR, neat) 3060, 3025, 2955, 2857, 1667, 1597, 1578, 1447, 1315, 1282, 973, 749 cm^{-1}; R_f 0.32

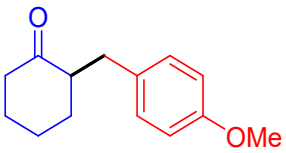
(petroleum ether/EtOAc, 100/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[19]



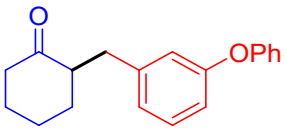
*2-Benzylcyclohexan-1-one (**51**).* According to **TP1**, the reaction of cyclohexanone (98 mg, 1 mmol) with benzyl alcohol (130 mg, 1.2 mmol) afforded the desired product **51** as a yellow oil (53 mg, 28%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.36–7.31 (m, 2 H), 7.28–7.17 (m, 3 H), 3.30 (dd, J = 13.9, 4.7 Hz, 1 H), 2.66–2.56 (m, 1 H), 2.54–2.34 (m, 3 H), 2.18–2.02 (m, 2 H), 1.95–1.84 (m, 1 H), 1.77–1.71 (m, 1 H), 1.67–1.57 (m, 1 H), 1.47–1.36 (m, 1 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 212.5, 140.3, 129.1, 128.2, 125.9, 52.4, 42.1, 35.4, 33.4, 28.0, 25.0; IR (diamond-ATR, neat) 3025, 2929, 2857, 1706, 1603, 1494, 1274, 1117, 720, 697 cm^{-1}; R_f 0.34 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[20]



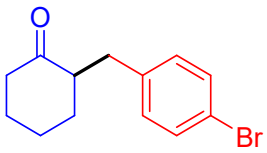
*2-(4-Methylbenzyl)cyclohexan-1-one (**52**).* According to **TP1**, the reaction of cyclohexanone (98 mg, 1 mmol) with 4-methylbenzyl alcohol (147 mg, 1.2 mmol) afforded the desired product **52** as a yellow oil (55 mg, 27%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.08 (d, J = 8.1 Hz, 2 H), 7.04 (d, J = 8.1 Hz, 2 H), 3.18 (dd, J = 13.9, 4.8 Hz, 1 H), 2.57–2.46 (m, 1 H), 2.46–2.32 (m, 3 H), 2.31 (s, 3 H), 2.10–1.97 (m, 2 H), 1.86–1.78 (m, 1 H), 1.68–1.51 (m, 2 H), 1.40–1.27 (m, 1 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 212.7, 137.2, 135.4, 128.9, 128.9, 52.5 , 42.1, 34.9, 33.3, 28.0, 25.0, 20.9; IR (diamond-ATR, neat) 3017, 2922, 2857, 1685, 1510, 1447, 1354, 1280, 825, 780, 615 cm^{-1}; R_f 0.33 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[20]



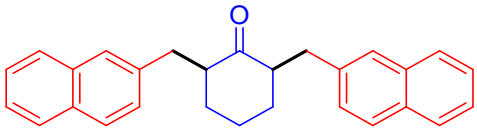
*2-(4-Methoxybenzyl)cyclohexan-1-one (**53**).* According to **TP1**, the reaction of cyclohexanone (98 mg, 1 mmol) with 4-methoxybenzyl alcohol (166 mg, 1.2 mmol) afforded the desired product **53** as a colorless oil (48 mg, 22%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.06 (d, J = 8.6 Hz, 2 H), 6.80 (d, J = 8.6 Hz, 2 H), 3.77 (s, 3 H), 3.14 (dd, J = 14.0, 4.9 Hz, 1 H), 2.55–2.25 (m, 4 H), 2.10–1.96 (m, 2 H), 1.87–1.77 (m, 1 H), 1.69–1.51 (m, 2 H), 1.40–1.30 (m, 1 H); $^{13}C\{H^1\}$ NMR (101 MHz, $CDCl_3$) δ 212.7, 157.8, 132.3, 129.9, 113.7, 55.2, 52.6, 42.1, 34.5, 33.3, 28.0, 24.9; IR (diamond-ATR, neat) 3032, 2929, 2855, 1685, 1609, 1582, 1508, 1440, 1358, 1298, 829, 734 cm^{-1}; R_f 0.30 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[20]



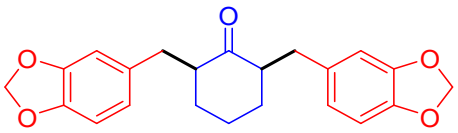
*2-(3-Phenoxybenzyl)cyclohexan-1-one (**54**).* According to **TP1**, the reaction of cyclohexanone (98 mg, 1 mmol) with 3-phenoxybenzyl alcohol (240 mg, 1.2 mmol) afforded the desired product **54** as a colorless oil (64 mg, 23%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39–7.27 (m, 2 H), 7.24–7.18 (m, 1 H), 7.09 (td, *J* = 7.4, 1.2 Hz, 1H), 7.02–6.97 (m, 2 H), 6.89 (dt, *J* = 7.5, 1.3 Hz, 1 H), 6.81 (dd, *J* = 6.5, 1.2 Hz, 2 H), 3.20 (dd, *J* = 13.9, 4.8 Hz, 1 H), 2.58–2.47 (m, 1 H), 2.46–2.27 (m, 3 H), 2.04 (dddd, *J* = 11.9, 10.7, 6.0, 2.8 Hz, 2 H), 1.87–1.78 (m, 1 H), 1.74–1.63 (m, 1 H), 1.60–1.51 (m, 1 H), 1.41–1.27 (m, 1 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 212.3, 157.2, 157.1, 142.5, 129.7, 129.5, 124.1, 123.1, 119.6, 118.8, 116.3, 52.2, 42.1, 35.3, 33.4, 27.9, 25.0; IR (diamond-ATR, neat) 3056, 2927, 2859, 1706, 1578, 1486, 1360, 1245, 883, 751, 691 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{19}H_{21}O_2$ [M + H]$^{+}$ 281.1542, found 281.1538; R_f 0.31 (petroleum ether/EtOAc, 20/1).



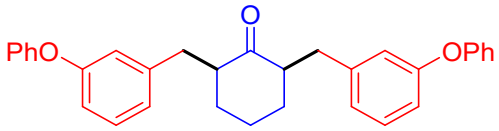
*2-(4-Bromobenzyl)cyclohexan-1-one (**55**).* According to **TP1**, the reaction of cyclohexanone (98 mg, 1 mmol) with 4-bromobenzyl alcohol (224 mg, 1.2 mmol) afforded the desired product **55** as a yellow oil (60 mg, 22%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.37 (d, *J* = 8.3 Hz, 2 H), 7.02 (d, *J* = 8.3 Hz, 2 H), 3.14 (dd, *J* = 13.9, 5.1 Hz, 1 H), 2.55–2.45 (m, 1 H), 2.45–2.25 (m, 3 H), 2.11–1.94 (m, 2 H), 1.87–1.77 (m, 1 H), 1.72–1.51 (m, 2 H), 1.40–1.27 (m, 1 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 212.2, 139.4, 131.3, 130.9, 119.7, 52.3, 42.2, 34.9, 33.5, 27.9, 25.1; IR (diamond-ATR, neat) 3021, 2922, 2852, 1706, 1486, 1311, 1010, 831, 792, 631 cm^{-1}; R_f 0.28 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[20]



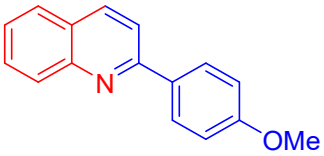
*2,6-Bis(naphthalen-2-ylmethyl)cyclohexan-1-one (**56**).* According to **TP2**, the reaction of cyclohexanone (98 mg, 1 mmol) with 2-naphthalenemethanol (380 mg, 2.4 mmol) afforded the desired product **56** as a pale yellow solid (64 mg, 17%): mp 114–116 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99–7.90 (m, 2 H), 7.90–7.83 (m, 2 H), 7.73 (d, *J* = 8.1 Hz, 2 H), 7.54–7.45 (m, 4 H), 7.43–7.37 (m, 2 H), 7.34 (dd, *J* = 7.1, 1.4 Hz, 2 H), 3.85 (dd, *J* = 14.2, 4.3 Hz, 2 H), 2.87 (dd, *J* = 14.2, 9.0 Hz, 2 H), 2.80–2.68 (m, 2 H), 2.05–1.94 (m, 2 H), 1.73–1.66 (m, 1 H), 1.53–1.35 (m, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 212.8, 136.4, 133.9, 131.9, 128.9, 127.5, 126.8, 125.8, 125.4, 125.3, 123.7, 51.7, 35.2, 32.4, 25.2; IR (diamond-ATR, neat) 3045, 2927, 2853, 1695, 1594, 11447, 1395, 1362, 1282, 858, 788 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{28}H_{26}ONa$ [M + Na]$^{+}$ 401.1881, found 401.1877; R_f 0.50 (petroleum ether/EtOAc, 20/1).



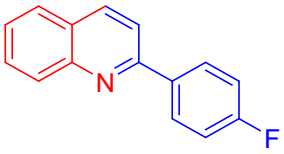
*2,6-Bis(benzo[d][1,3]dioxol-5-ylmethyl)cyclohexan-1-one (**57**).* According to **TP2**, the reaction of cyclohexanone (98 mg, 1 mmol) with piperonyl alcohol (365 mg, 2.4 mmol) afforded the desired product **57** as a pale yellow solid (73 mg, 20%): mp 147–149 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 6.70 (d, J = 7.9 Hz, 2 H), 6.64 (d, J = 1.7 Hz, 2 H), 6.59 (dd, J = 7.9, 1.7 Hz, 2 H), 5.90 (s, 4 H), 3.12 (dd, J = 14.0, 4.9 Hz, 2 H), 2.58–2.43 (m, 2 H), 2.34 (dd, J = 14.1, 8.5 Hz, 2 H), 2.12–1.99 (m, 2 H), 1.84–1.71 (m, 1 H), 1.56–1.49 (m, 1 H), 1.35–1.26 (m, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 212.8, 147.5, 145.7, 134.3, 121.9, 109.5, 108.0, 100.8, 53.0, 35.2, 34.8, 25.3; IR (diamond-ATR, neat) 2950, 2922, 2854, 1693, 1608, 1500, 1487, 1363, 1246, 1194, 927, 866, 818, 790, 637 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{22}H_{23}O_5$ [M + H]$^{+}$ 367.1545, found 367.1542; R_f 0.20 (petroleum ether/EtOAc, 20/1).



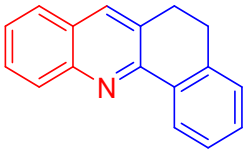
*2,6-Bis(3-phenoxybenzyl)cyclohexan-1-one (**58**).* According to **TP2**, the reaction of cyclohexanone (98 mg, 1 mmol) with 3-phenoxybenzyl alcohol (481 mg, 2.4 mmol) afforded the desired product **58** as a white solid (125 mg, 27%): mp 76–78 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.38–7.32 (m, 4 H), 7.27–7.22 (m, 2 H), 7.14–7.09 (m, 2 H), 7.04–6.99 (m, 4 H), 6.92 (dt, J = 7.7, 1.3 Hz, 2 H), 6.87–6.81 (m, 4 H), 3.22 (dd, J = 14.0, 4.8 Hz, 2 H), 2.58 (ddt, J = 13.2, 9.5, 5.1 Hz, 2 H), 2.42 (dd, J = 14.0, 8.7 Hz, 2 H), 2.12–2.03 (m, 2 H), 1.86–1.77 (m, 1 H), 1.62–1.52 (m, 1 H), 1.41–1.27 (m, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$): δ 212.3, 157.2, 157.1, 142.6, 129.7, 129.5, 124.1, 123.1, 119.5, 118.8, 116.3, 52.6, 35.3, 34.7, 25.3; IR (diamond-ATR, neat) 3040, 2926, 2855, 1708, 1582, 1443, 1252, 887, 783, 693 cm^{-1}; HRMS (ESI^{+}) calcd for $C_{32}H_{30}O_3Na$ [M + Na]$^{+}$ 485.2093, found 485.2084; R_f 0.33 (petroleum ether/EtOAc, 20/1).



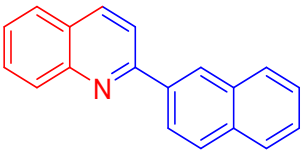
*2-(4-Methoxyphenyl)quinoline (**59**).* According to **TP1**, the reaction of 4′-methoxyacetophenone (150 mg, 1 mmol) with 2-aminobenzyl alcohol (148 mg, 1.2 mmol) afforded the desired product **59** as a white solid (115 mg, 49%): mp 122–123 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.22–8.05 (m, 4 H), 7.87–7.75 (m, 2 H), 7.76–7.62 (m, 1 H), 7.56–7.42 (m, 1 H), 7.12–6.93 (m, 2 H), 3.87 (s, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 160.7, 156.8, 148.2, 136.6, 132.2, 129.5, 129.4, 128.8, 127.4, 126.8, 125.9, 118.5, 114.2, 55.3; IR (diamond-ATR, neat) 3038, 2957, 2840, 1595, 1517, 1496, 1319, 1247, 1027, 946, 815, 747, 646 cm^{-1}; R_f 0.31 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[3]



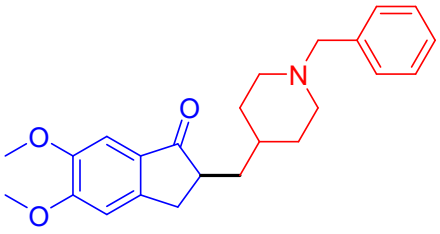
*2-(4-Fluorophenyl)quinoline (**60**).* According to **TP1**, the reaction of 4′-fluoroacetophenone (138 mg, 1 mmol) with 2-aminobenzyl alcohol (148 mg, 1.2 mmol) afforded the desired product **60** as a white solid (103 mg, 46%): mp 88–89 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.20 (d, J = 8.7 Hz, 1 H), 8.18–8.12 (m, 3 H), 7.81 (d, J = 8.6 Hz, 2 H), 7.76–7.69 (m, 1 H), 7.55–7.49 (m, 1 H), 7.24–7.16 (m, 2 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 163.8 (d, J = 249.0 Hz), 156.2, 148.2, 136.9, 135.8 (d, J = 3.1 Hz), 129.8, 129.6, 129.4 (d, J = 8.4 Hz), 127.4, 127.0, 126.3, 118.6, 115.7 (d, J = 21.6 Hz); ^{19}F NMR (376 MHz, $CDCl_3$) δ -112.52; IR (diamond-ATR, neat) 3042, 2924, 2855, 1554, 1492, 1321, 1267, 852, 755, 648 cm^{-1}; R_f 0.48 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[3]



*5,6-Dihydrobenzo[c]acridine (**61**).* According to **TP1**, the reaction of 1-tetralone (146 mg, 1 mmol) with 2-aminobenzyl alcohol (148 mg, 1.2 mmol) afforded the desired product **61** as a white solid (130 mg, 56%): mp 60–61 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.59 (dd, J = 7.6, 1.4 Hz, 1 H), 8.15 (d, J = 8.4 Hz, 1 H), 7.90 (s, 1 H), 7.73 (dd, J = 8.0, 1.4 Hz, 1 H), 7.70–7.60 (m, 1 H), 7.53–7.41 (m, 2 H), 7.41–7.34 (m, 1 H), 7.27 (d, J = 7.4 Hz, 1 H), 3.11 (t, J = 7.5 Hz, 2 H), 3.00 (t, J = 7.5 Hz, 2 H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 153.3, 147.6, 139.4, 134.7, 133.6, 130.5, 129.6, 129.3, 128.6, 127.9, 127.8, 127.3, 126.9, 125.9, 28.8, 28.3; IR (diamond-ATR, neat) 3036, 2931, 2846, 1554, 1434, 1344, 1253, 911, 860, 767, 613 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[12]



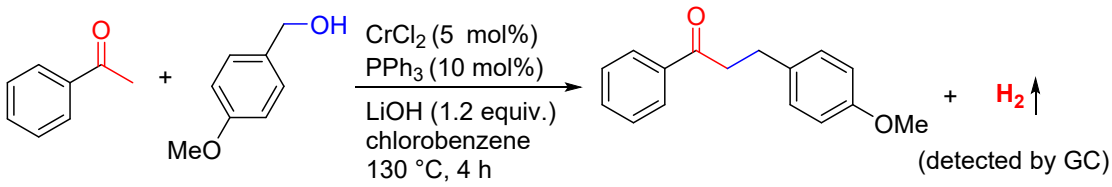
*2-(Naphthalen-2-yl)quinoline (**62**).* According to **TP1**, the reaction of 2-acetylnaphthalene (170 mg, 1 mmol) with 2-aminobenzyl alcohol (148 mg, 1.2 mmol) afforded the desired product **62** as a white solid (135 mg, 53%): mp 160–161 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.61 (s, 1 H), 8.38 (dd, J = 8.6, 1.8 Hz, 1 H), 8.24 (d, J = 8.5 Hz, 2 H), 8.07–7.95 (m, 3 H), 7.95–7.86 (m, 1 H), 7.84 (dd, J = 8.1, 1.4 Hz, 1 H), 7.78–7.72 (m, 1 H), 7.57–7.49 (m, 3 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 157.1, 148.3, 136.9, 136.8, 133.8, 133.5, 129.7, 129.6, 128.8, 128.5, 127.7, 127.5, 127.2, 127.1, 126.7, 126.3, 126.3, 125.0, 119.1; IR (diamond-ATR, neat) 3050, 2927, 2852, 1556, 1494, 1358, 1249, 817, 736, 691 cm^{-1}; R_f 0.50 (petroleum ether/EtOAc, 20/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[21]



*2-((1-Benzylpiperidin-4-yl)methyl)-5,6-dimethoxy-2,3-dihydro-1H-inden-1-one (**63**)*. According to **TP1**, the reaction of 5,6-dimethoxy-1-indanone (384 mg, 2 mmol) with (1-benzyl-4-piperidyl)methanol (493 mg, 2.4 mmol) afforded the desired product **63** as a yellow oil (205 mg, 27%): ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32–7.26 (m, 4 H), 7.26–7.19 (m, 1 H), 7.14 (s, 1 H), 6.83 (s, 1 H), 3.93 (s, 3 H), 3.88 (s, 3 H), 3.48 (s, 2 H), 3.20 (dd, J = 17.5, 8.1 Hz, 1 H), 2.95–2.82 (m, 2 H), 2.73–2.61 (m, 2 H), 2.02–1.84 (m, 3 H), 1.77–1.58 (m, 2 H), 1.52–1.25 (m, 4 H); ^{13}C{H^{1}} NMR (101 MHz, $CDCl_3$) δ 207.7, 155.4, 149.3, 148.7, 138.3, 129.2, 129.2, 128.1, 126.8, 107.3, 104.3, 63.4, 56.1, 56.0, 53.7, 53.7, 45.4, 38.6, 34.4, 33.3, 32.9, 31.7; IR (diamond-ATR, neat) 3060, 2920, 2842, 1689, 1589, 1453, 1364, 1261, 973, 862, 732, 646 cm^{-1}; R_f 0.31 (petroleum ether/EtOAc, 1/1). All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[22]

Gas Chromatography analysis

The reactions were set up following the general procedure for $CrCl_2$-catalyzed α-alkylation of ketones with alcohols. As depicted in Figure S1 and Figure S2, H_2 was detected by GC analysis (TCD detection) of the reaction headspace, which was performed on a SCION 436-GC instrument equipped with dual TCD detectors and nitrogen as the carrier gas.



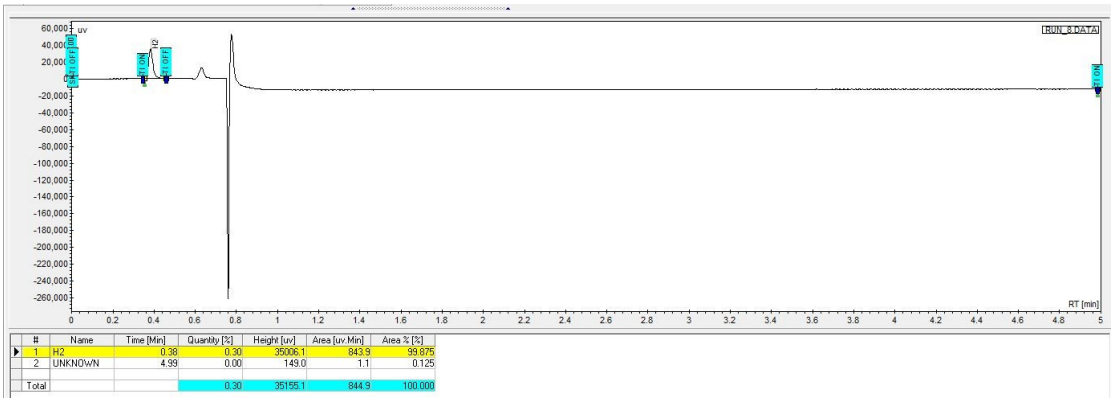
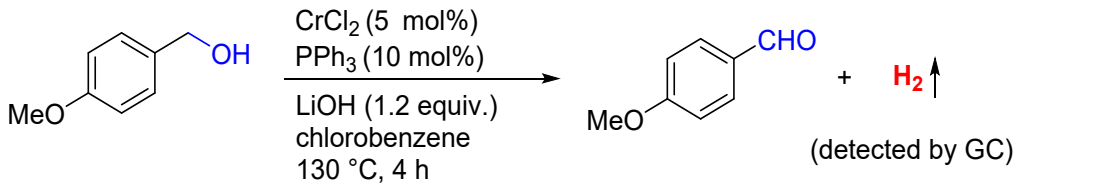


Figure S1. Evidence for H_2 evolution via GC analysis



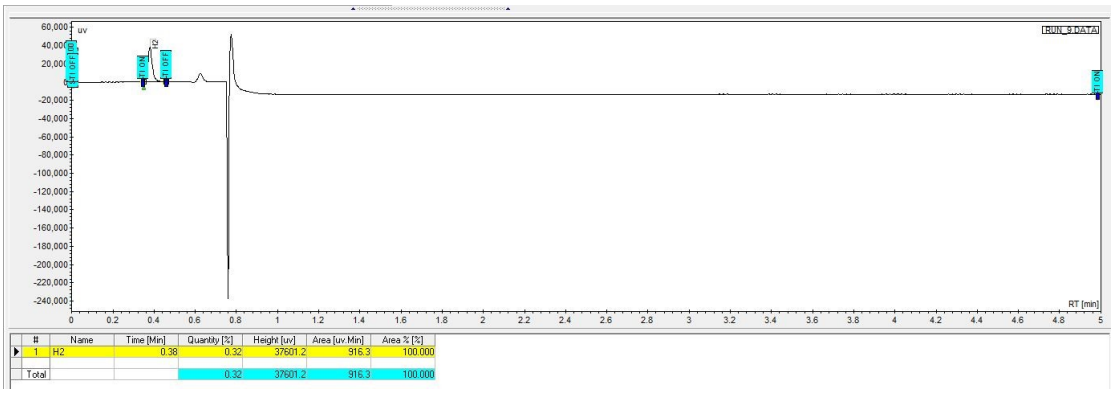
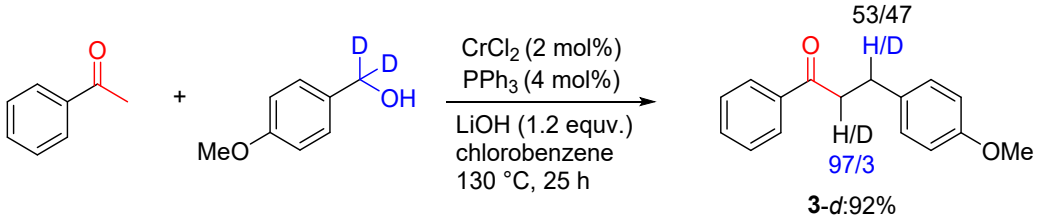


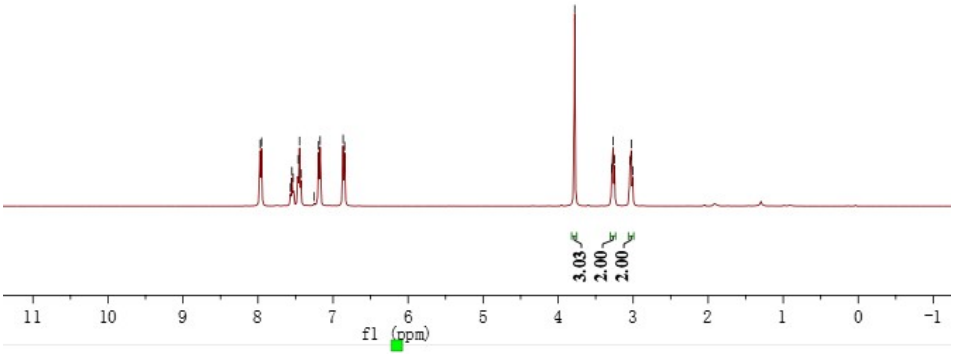
Figure S2. Evidence for H_2 evolution from the dehydrogenation of 4-methoxybenzyl alcohol via GC analysis

Deuterium labelling experiments

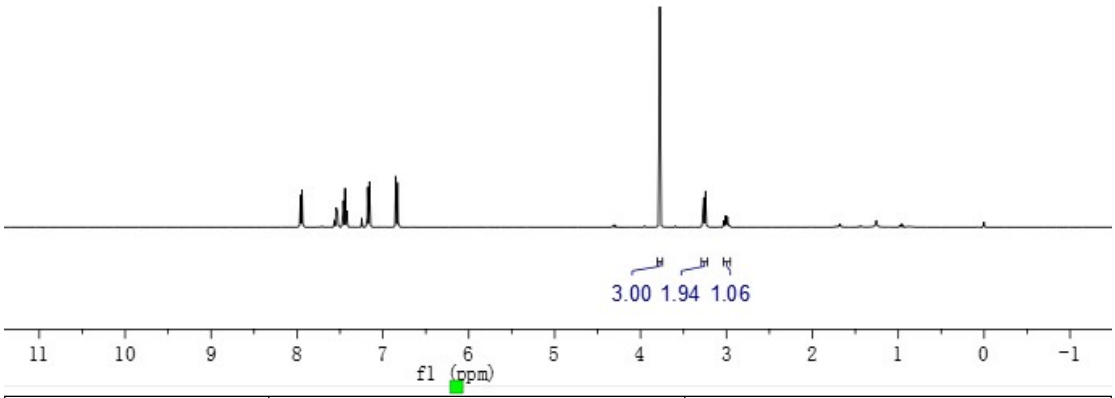


Acetophenone (51 mg, 0.42 mmol) reacted with deuterated 4-methoxybenzyl alcohol (71 mg, 0.5 mmol) in the presence of LiOH (12 mg, 0.15 mmol), $CrCl_2$ (3 mg, 0.02 mmol) and PPh_3 (11 mg, 0.042 mmol) in chlorobenzene (1 mL) at 130 °C for 25 h under nitrogen atmosphere, giving the expected product in the yield of 92% (81 mg).

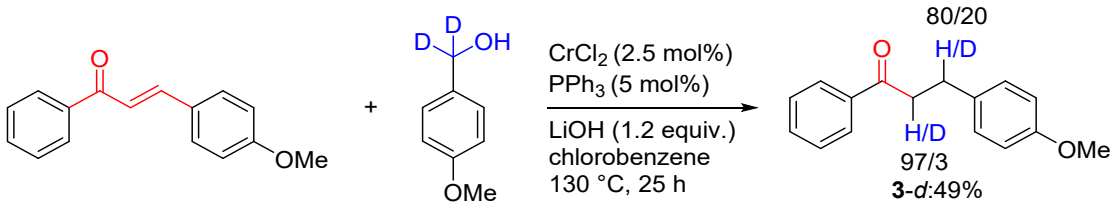
Standard ^{1}H NMR spectra of compound **3**



^{1}H NMR spectra of the product from the reaction of acetophenone with deuterated 4-methoxybenzyl alcohol



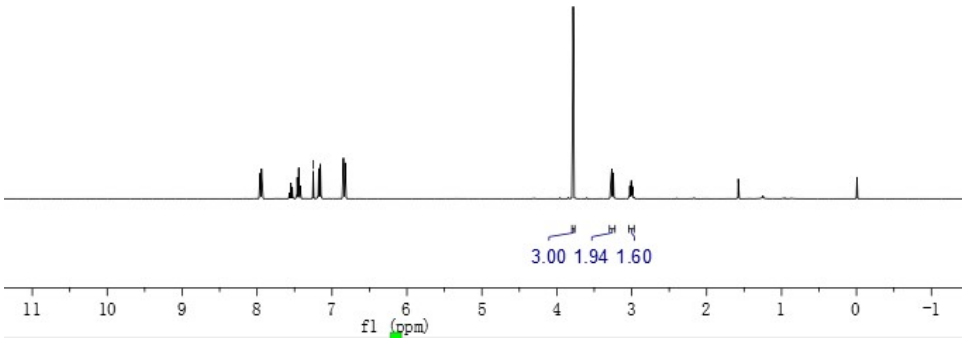
	Deuterium incorporation in α position	**Deuterium incorporation in β position**
Singnal δ	3.27 (2 H)	3.03 (2 H)
Integral value	1.94	1.06
Calculated ratio	{(2-1.94)/2}×100=3%	{(2-1.06)/2}×100=47%



4-Methoxychalcone (119 mg, 0.5 mmol) reacted with deuterated 4-methoxybenzyl alcohol (85 mg, 0.6 mmol) in the presence of LiOH (15 mg, 0.6 mmol), $CrCl_2$ (4 mg, 0.02 mmol) and PPh_3 (13 mg, 0.05 mmol) in chlorobenzene (1 mL) at 130 °C for 25 h under nitrogen atmosphere, giving the expected product in the yield of 49% (59 mg).

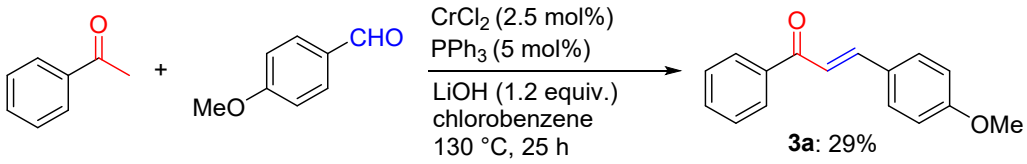
Standard ^{1}H NMR spectra of compound **3**.

^{1}H NMR spectra of the product from the reaction of 4-methoxychalcone deuterated 4-methoxybenzyl alcohol.



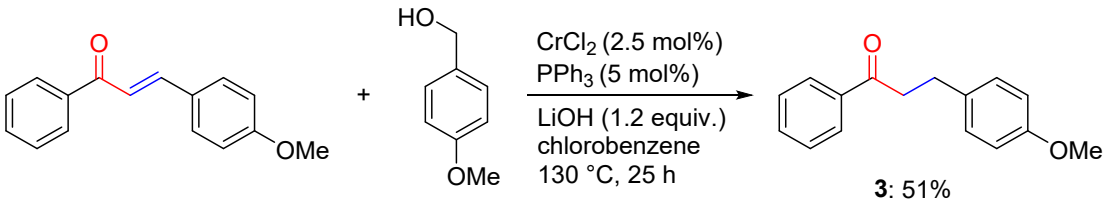
	Deuterium incorporation in α position	**Deuterium incorporation in β position**
Singnal δ	3.27 (2 H)	3.03 (2 H)
Integral value	1.94	1.60
Calculated ratio	{(2-1.94)/2}×100=3%	{(2-1.60)/2}×100=20%

Isolation of the intermediate 4-methoxychalcone 3a



Acetophenone (60 mg, 0.5 mmol) reacted with 4-methoxybenzaldehyde (82 mg, 0.6 mmol) in the presence of LiOH (15 mg, 0.6 mmol), $CrCl_2$ (4 mg, 0.02 mmol) and PPh_3 (13 mg, 0.05 mmol) in chlorobenzene (1 mL) at 130 °C for 25 h under nitrogen atmosphere, giving the expected product **3a** in the yield of 29% (35 mg). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.0 –7.98 (m, 2 H), 7.78 (d, J = 15.6 Hz, 1 H), 7.62–7.59 (m, 2 H), 7.58–7.54 (m, 1 H), 7.52–7.47 (m, 2 H), 7.41 (d, J = 15.6 Hz, 1 H), 6.96–6.91 (m, 2 H), 3.85 (s, 3 H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 190.6, 161.7, 144.7, 138.5, 132.6, 130.2, 128.6, 128.4, 127.6, 119.8, 114.4, 55.4. All the resonances in the ^{1}H and ^{13}C NMR spectra were consistent with reported values.[23]

Hydrogenation of the intermediate 4-methoxychalcone 3a



4-Methoxychalcone (119 mg, 0.5 mmol) reacted with 4-methoxybenzyl alcohol (85 mg, 0.6 mmol) in the presence of LiOH (15 mg, 0.6 mmol), $CrCl_2$ (4 mg, 0.02 mmol) and PPh_3 (13 mg, 0.05 mmol) in chlorobenzene (1 mL) at 130 °C for 25 h under nitrogen atmosphere, giving the expected product **3** in the yield of 51% (56 mg).

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^{1}H and $^{13}C\{^{1}H\}$ NMR Spectra of Compounds

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **3**

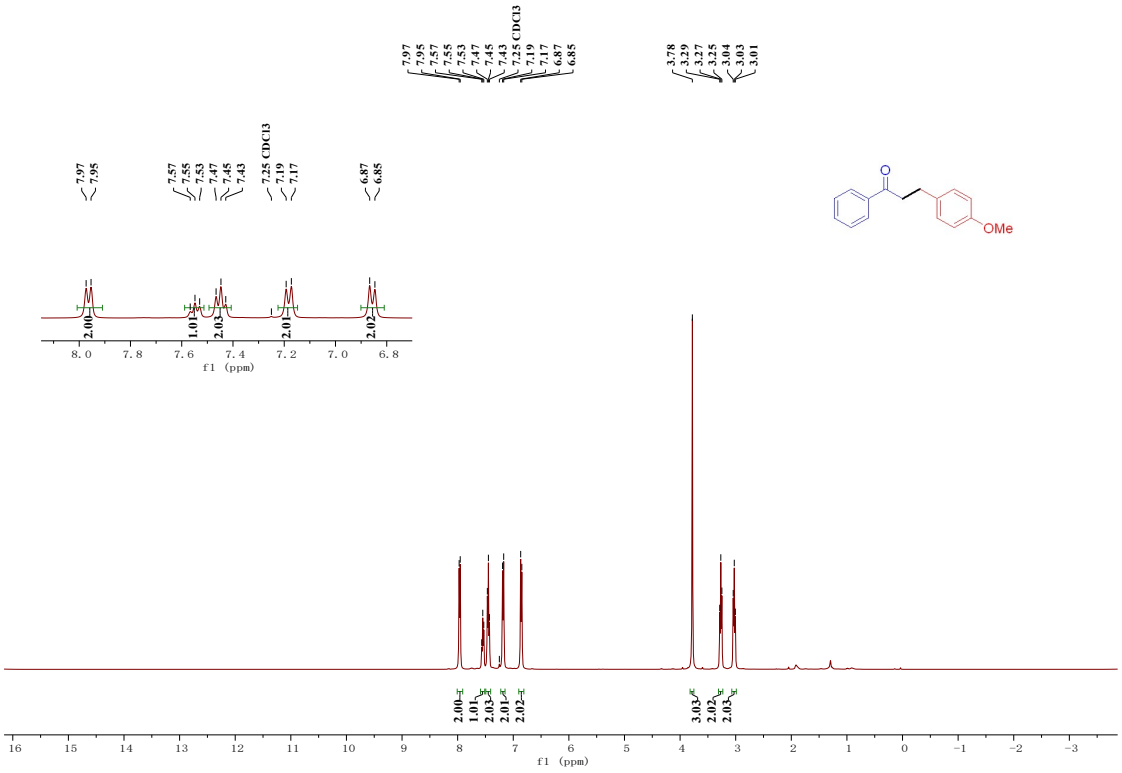


Figure S3. ^{1}H NMR (400 MHz) spectrum of compound 3 in $CDCl_3$

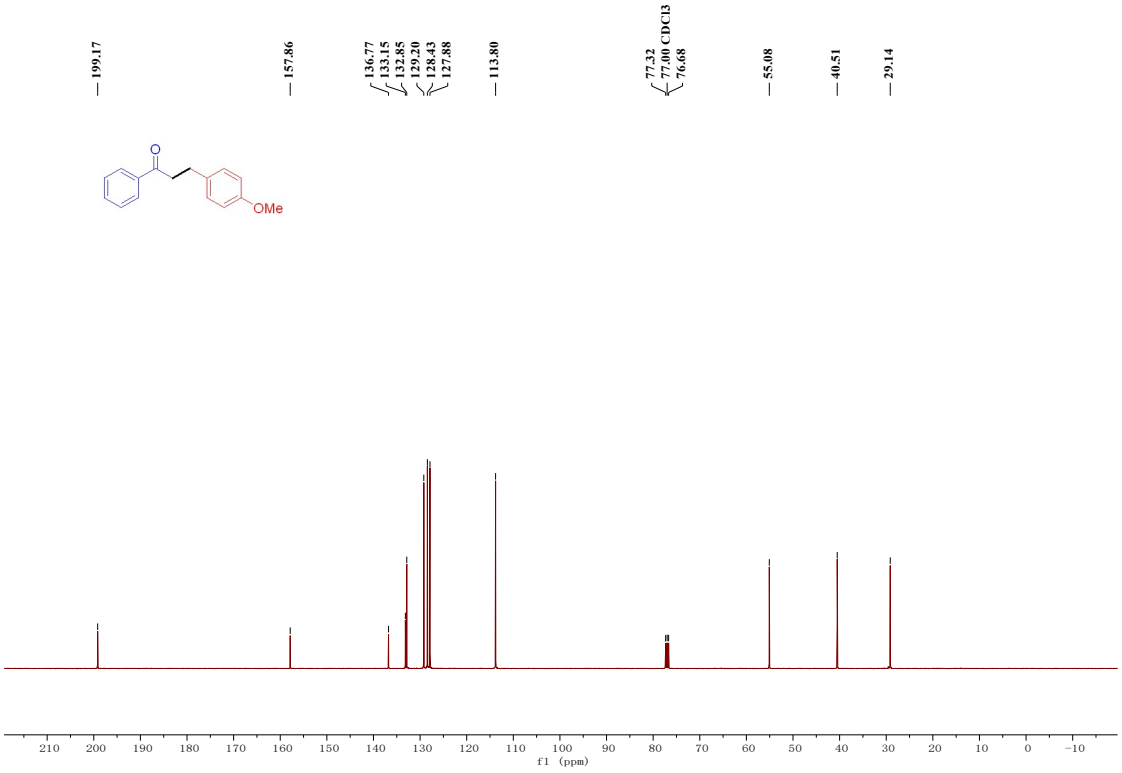


Figure S4. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 3 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **4**

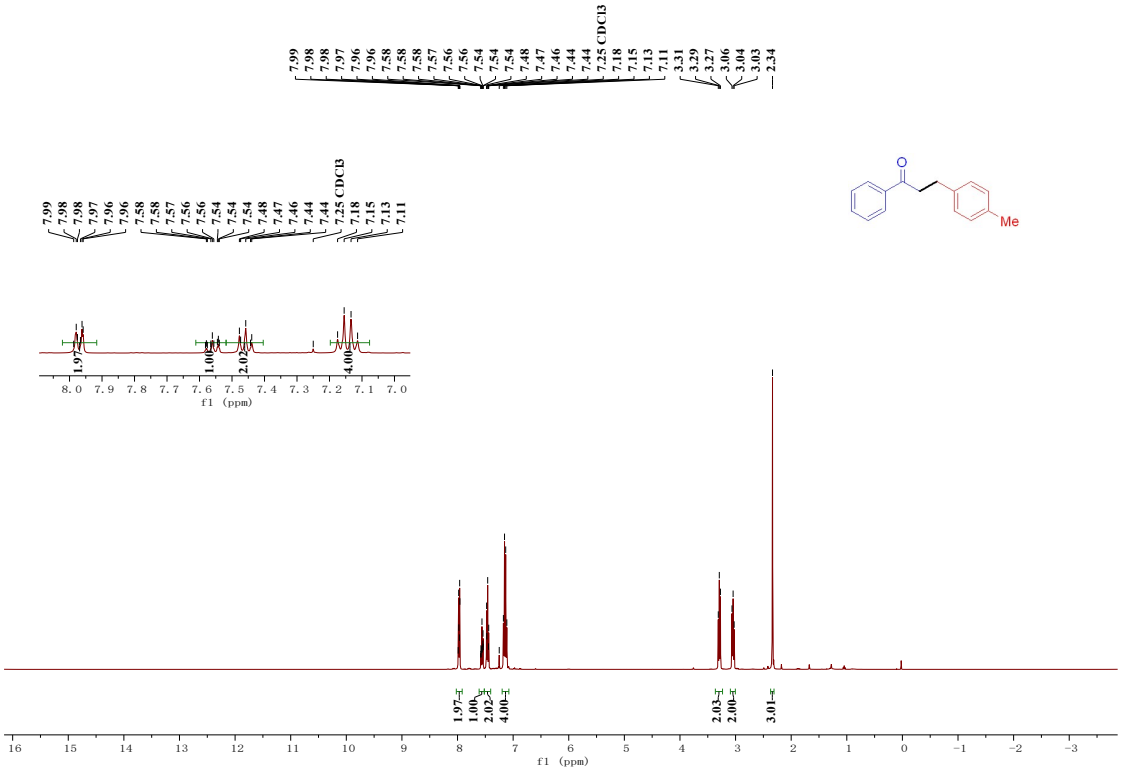


Figure S5. ^{1}H NMR (400 MHz) spectrum of compound 4 in $CDCl_3$

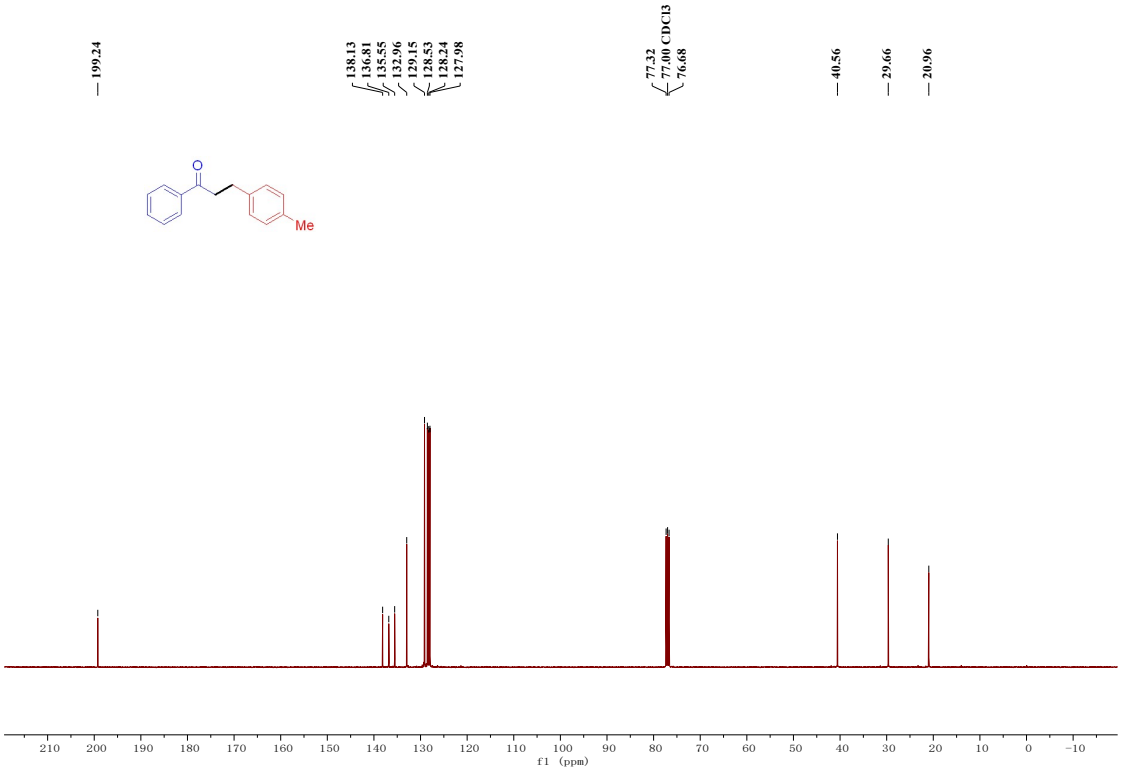


Figure S6. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 4 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **5**

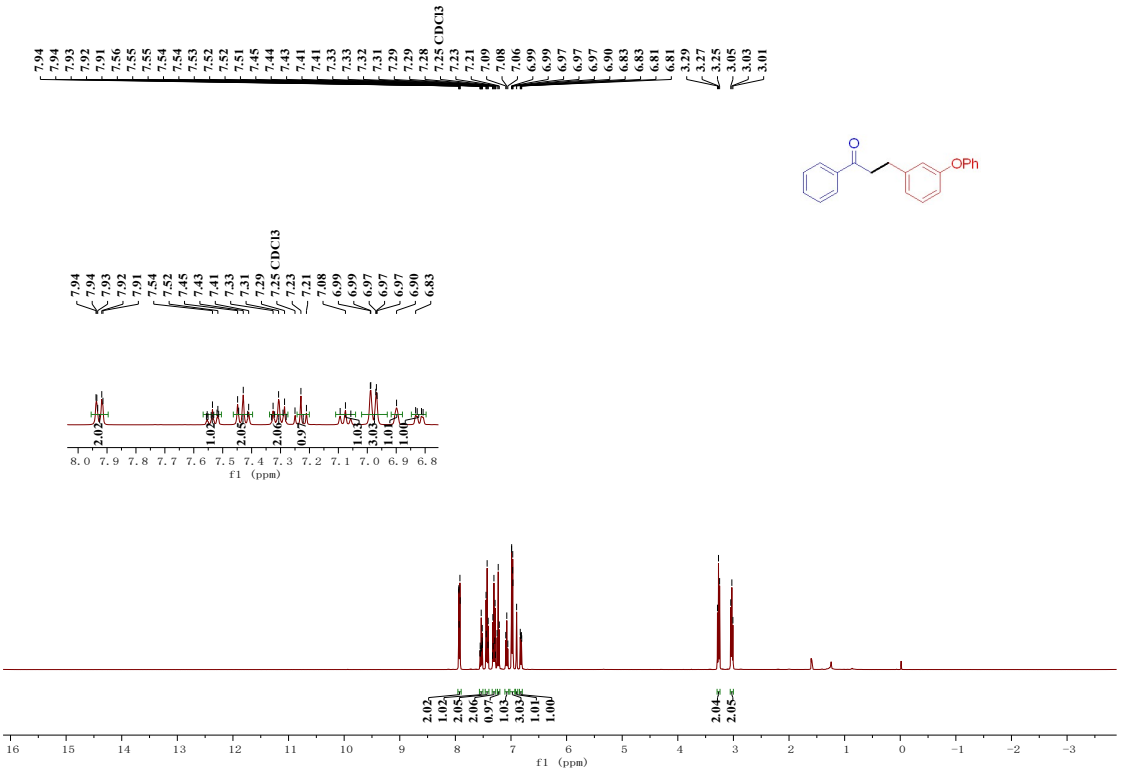


Figure S7. ^{1}H NMR (400 MHz) spectrum of compound 5 in $CDCl_3$

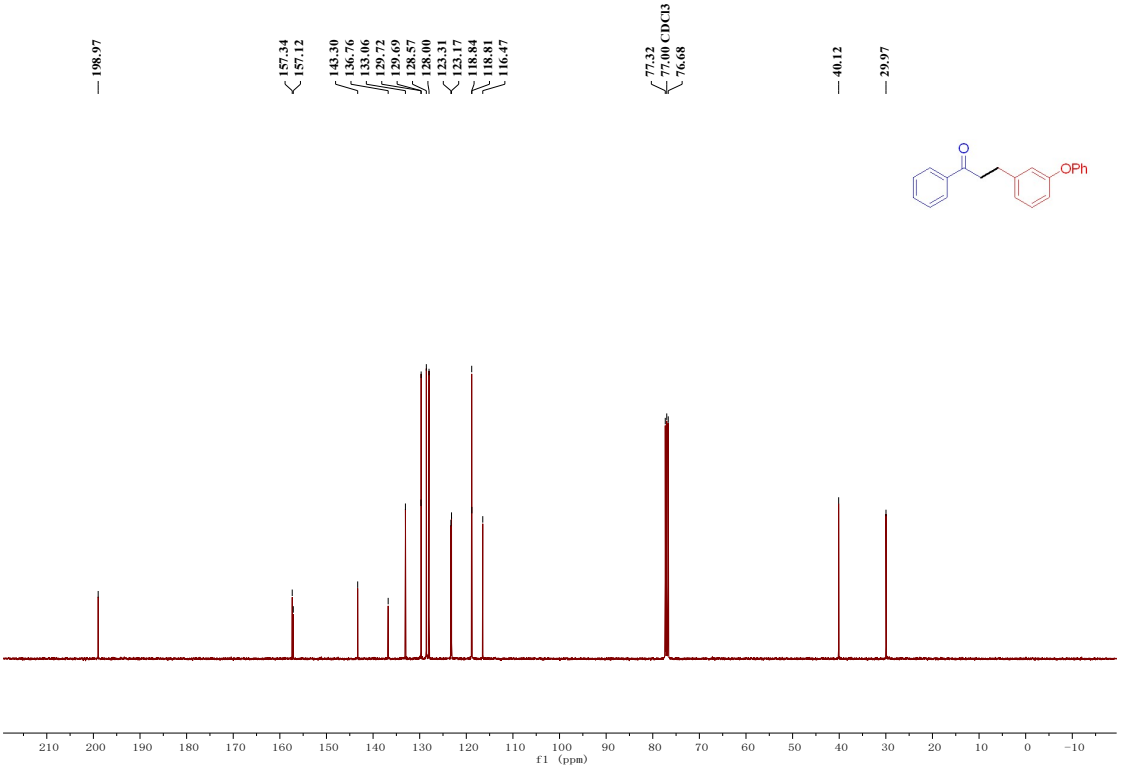


Figure S8. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 5 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **6**

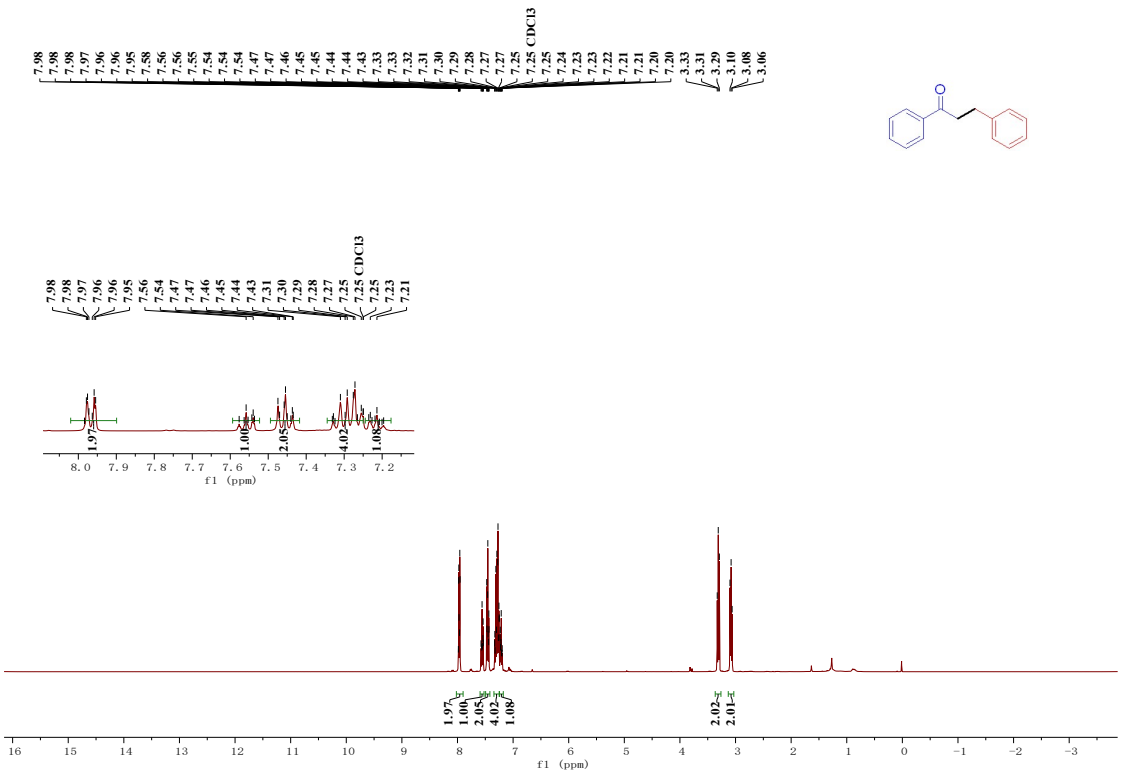


Figure S9. ^{1}H NMR (400 MHz) spectrum of compound 6 in $CDCl_3$

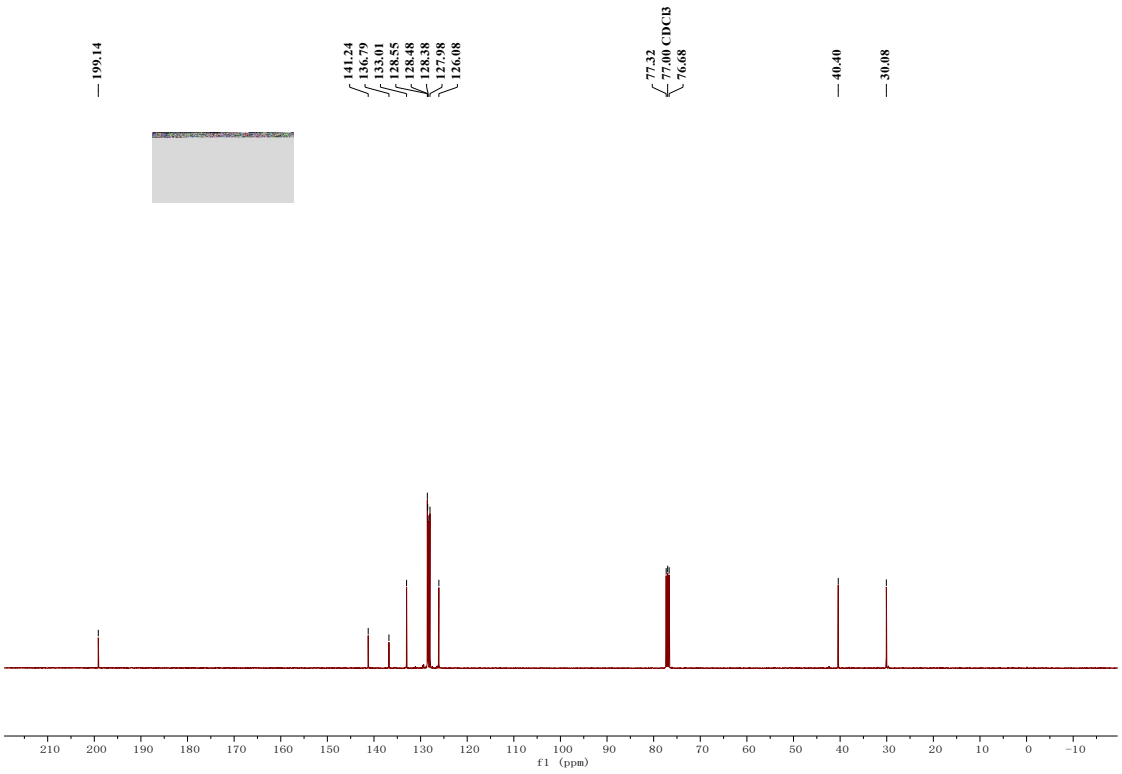


Figure S10. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 6 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **7**

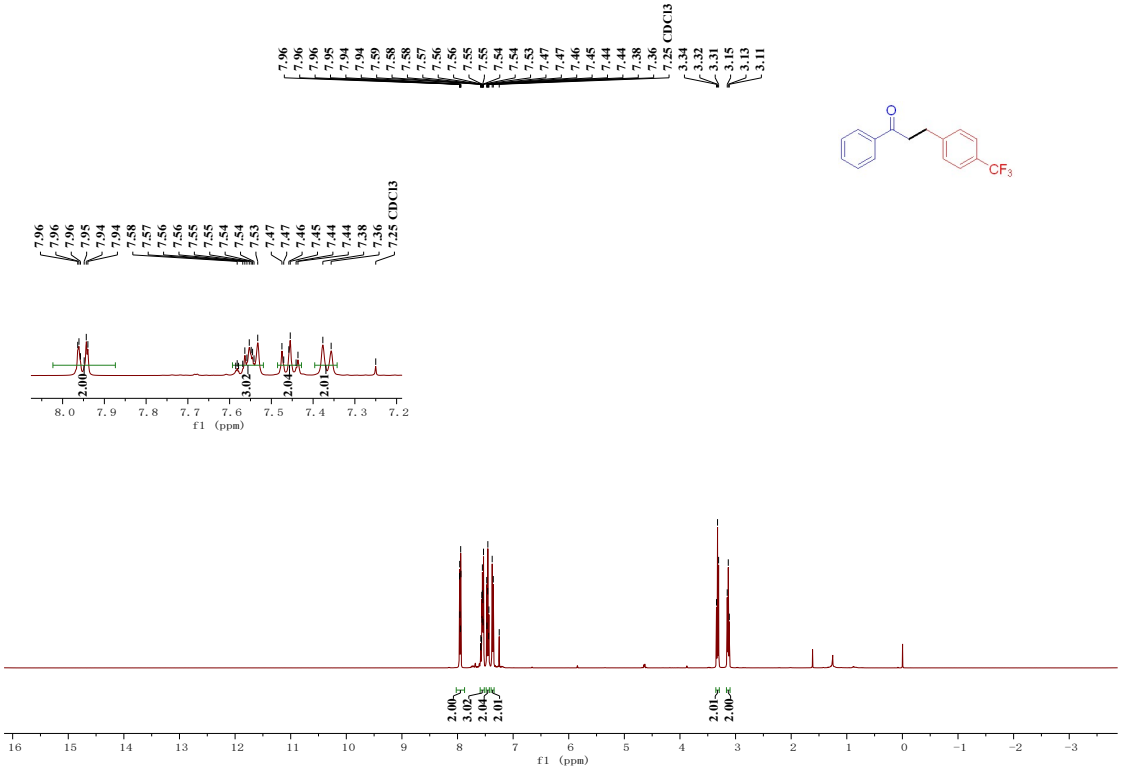


Figure S11. ^{1}H NMR (400 MHz) spectrum of compound 7 in $CDCl_3$

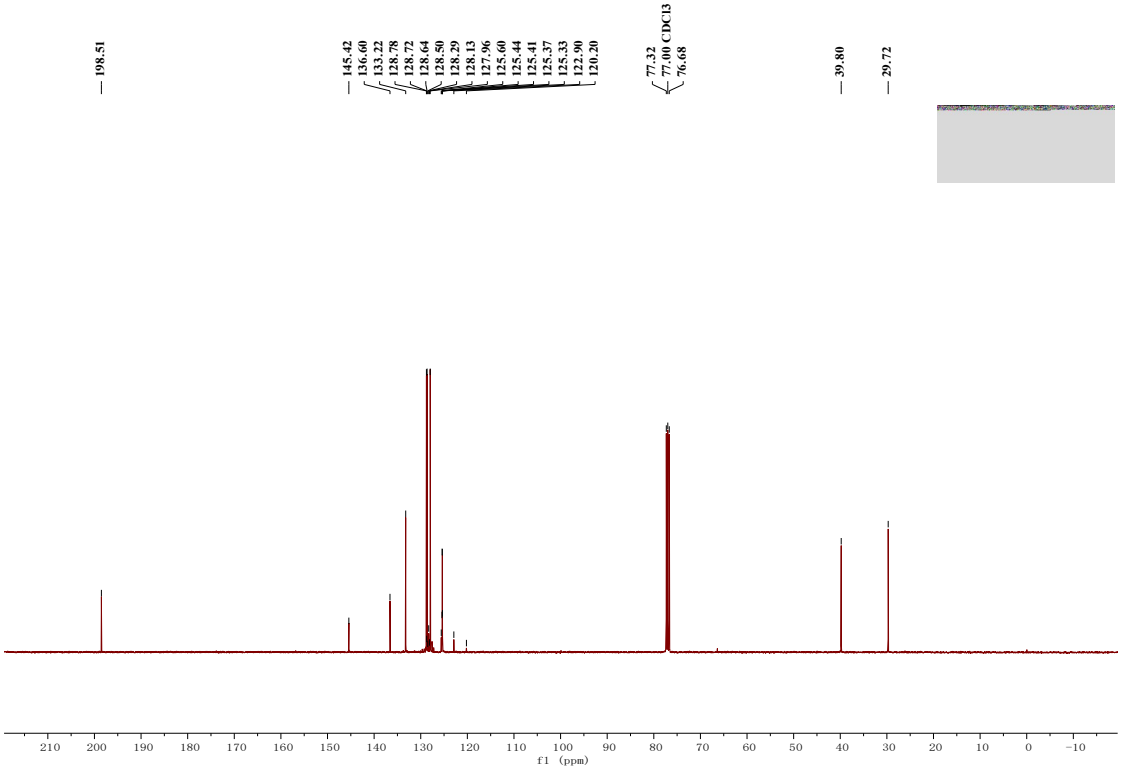


Figure S12. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 7 in $CDCl_3$

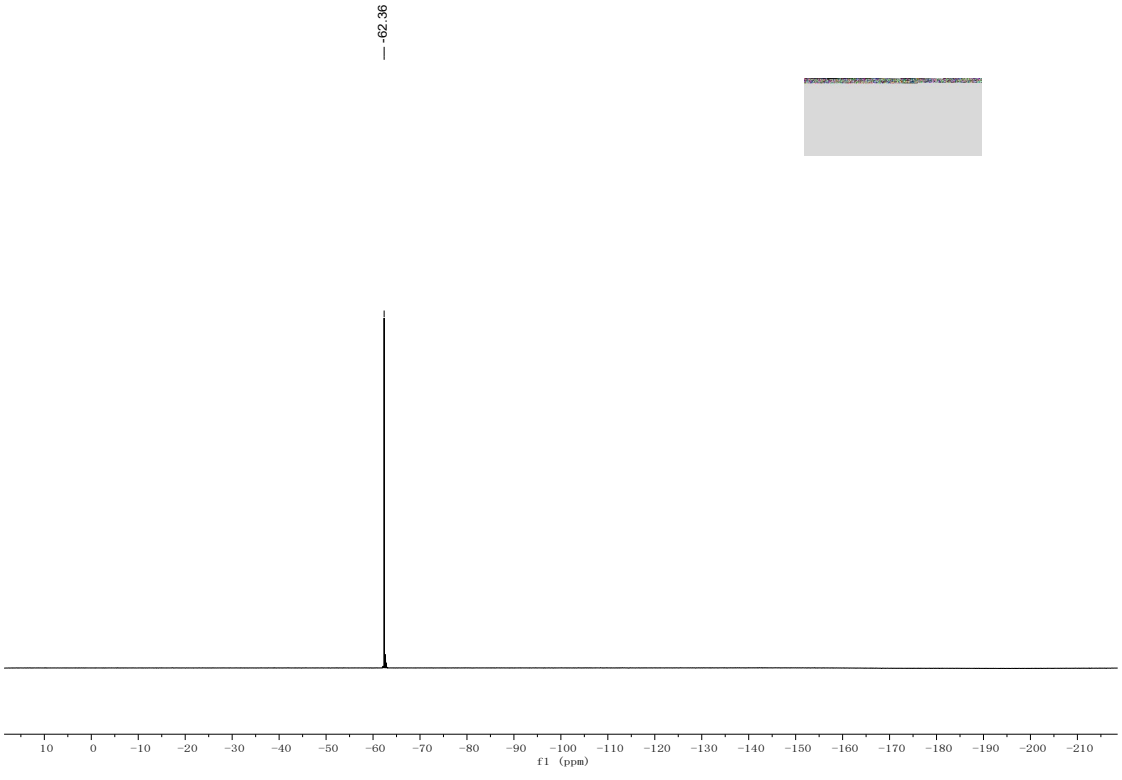


Figure S13. ^{19}F NMR (376 MHz) spectrum of compound 7 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **8**

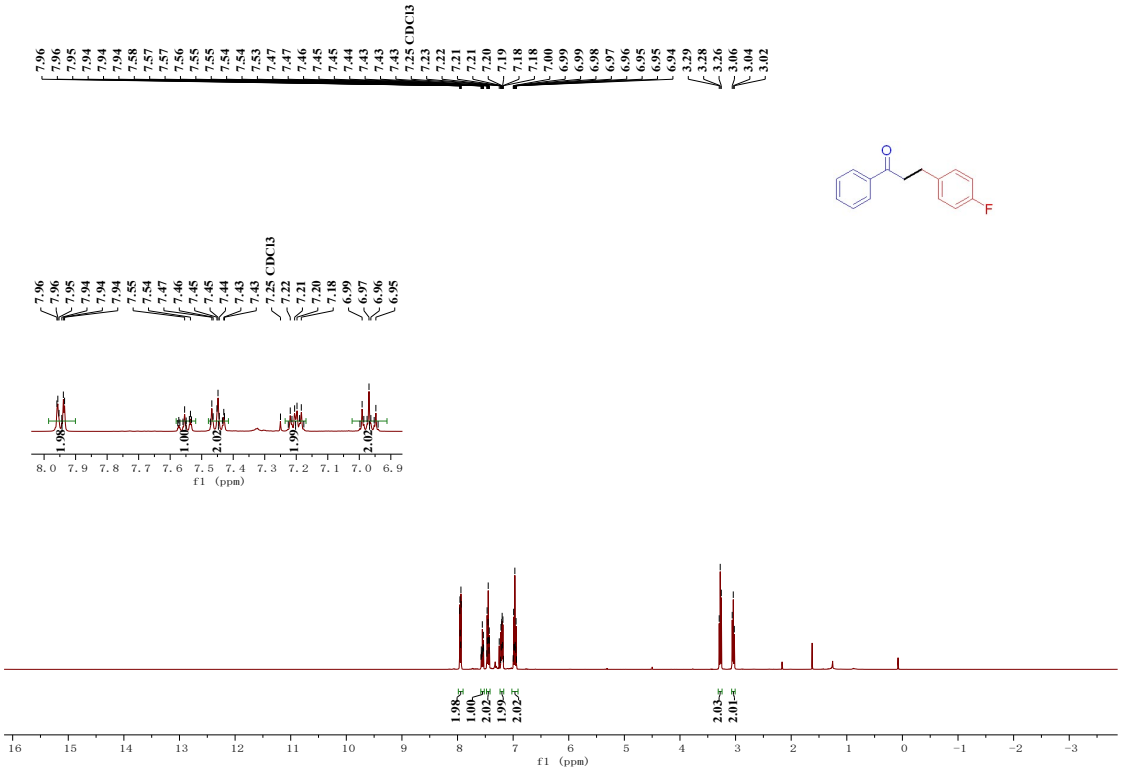


Figure S14. ^{1}H NMR (400 MHz) spectrum of compound 8 in $CDCl_3$

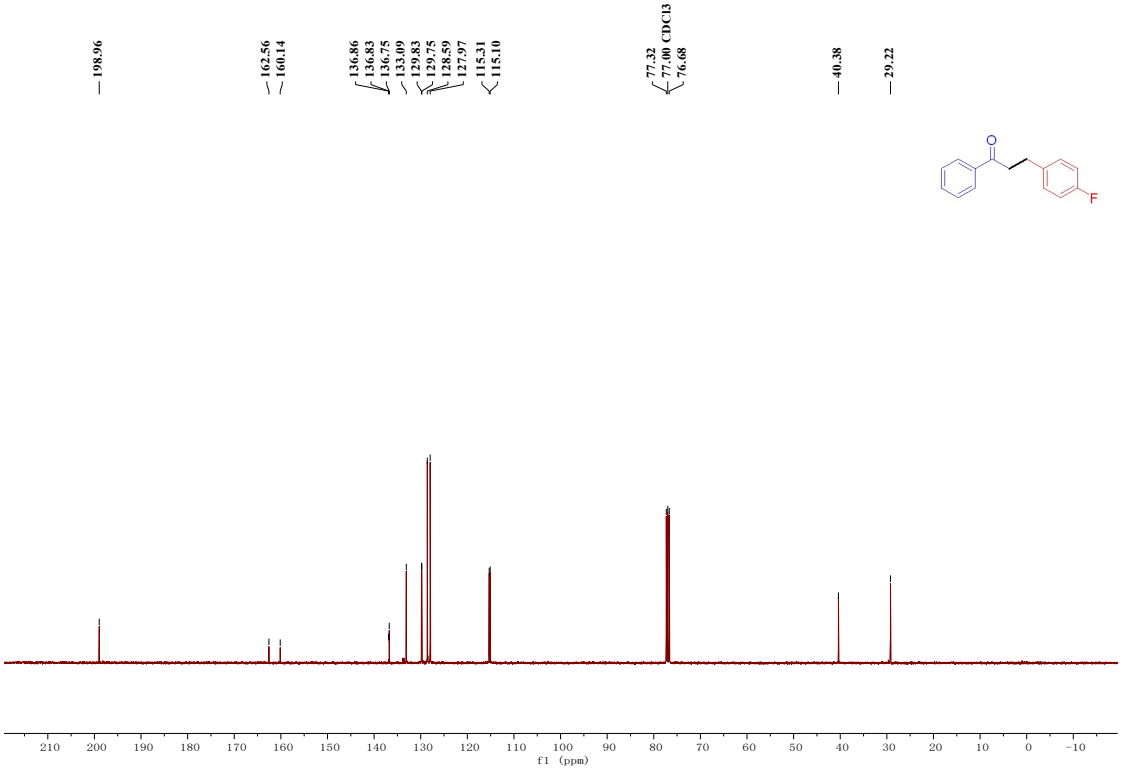


Figure S15. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 8 in $CDCl_3$

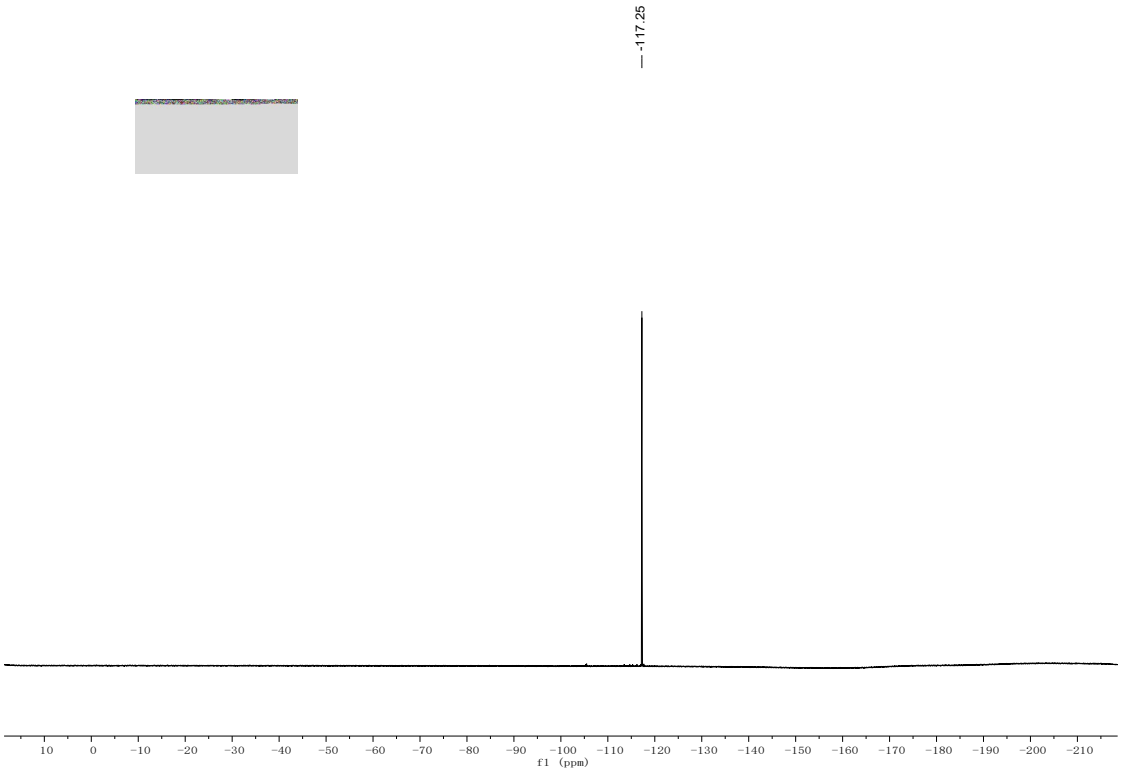


Figure S16. ^{19}F NMR (376 MHz) spectrum of compound 8 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **9**

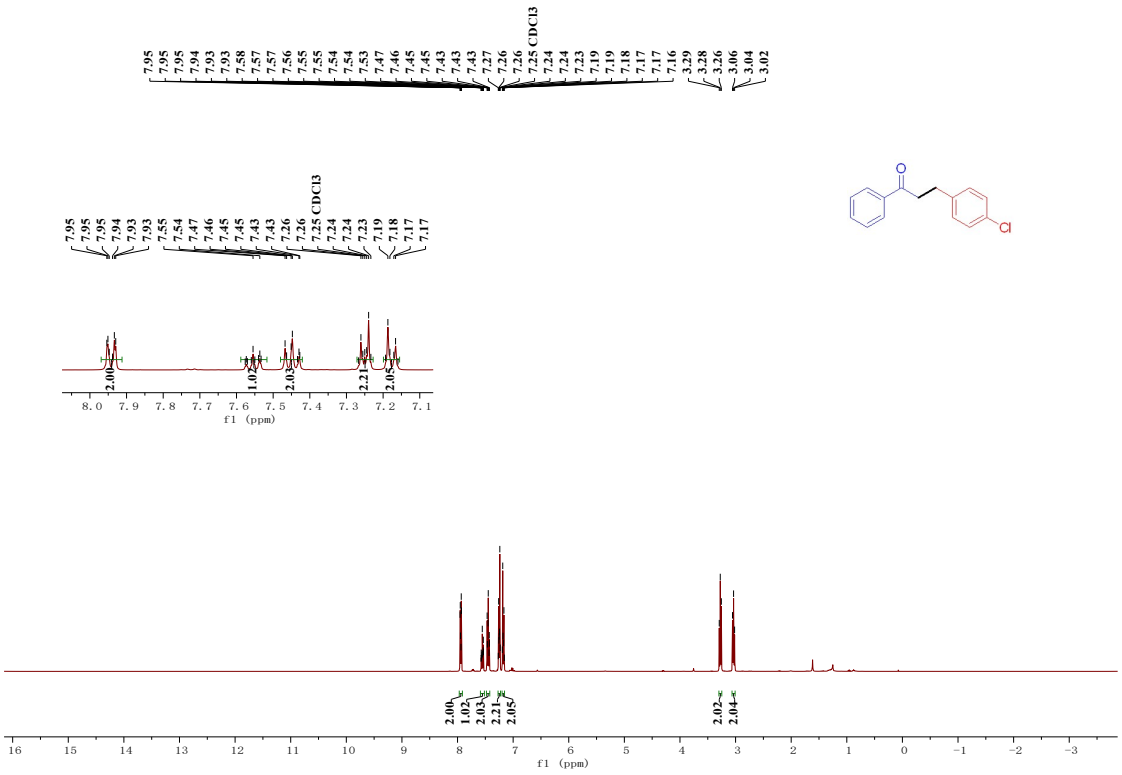


Figure S17. ^{1}H NMR (400 MHz) spectrum of compound 9 in $CDCl_3$

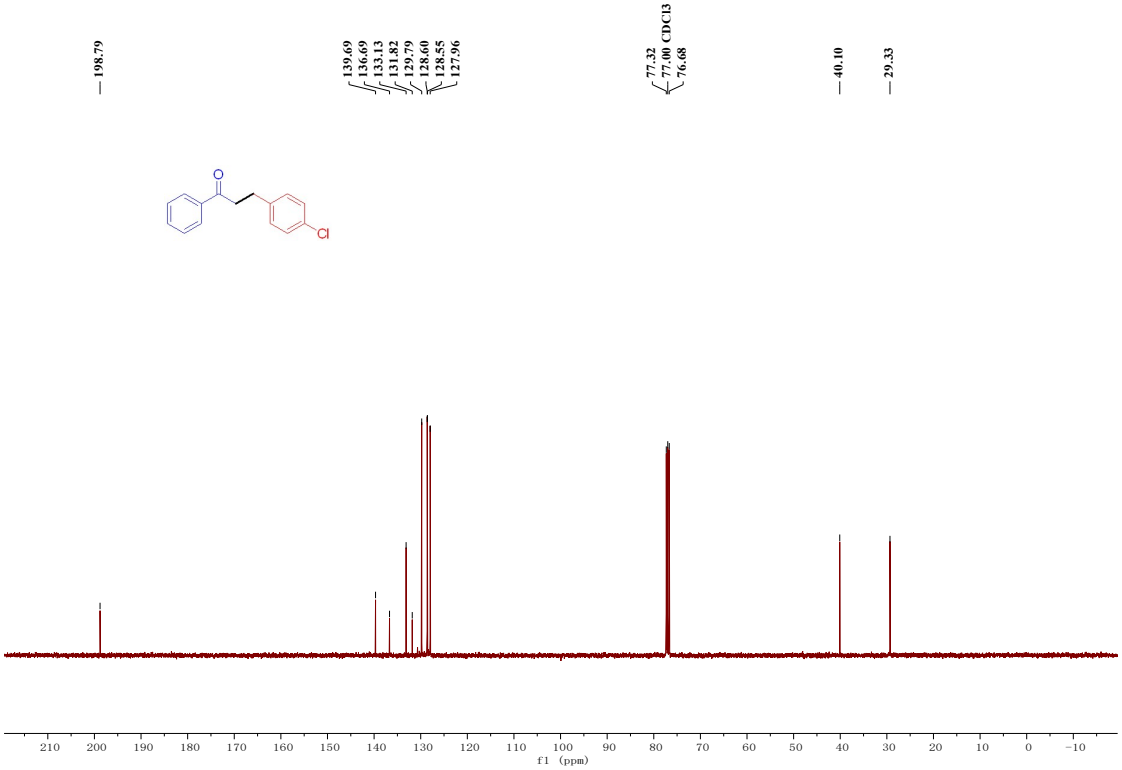


Figure S18. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 9 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **10**

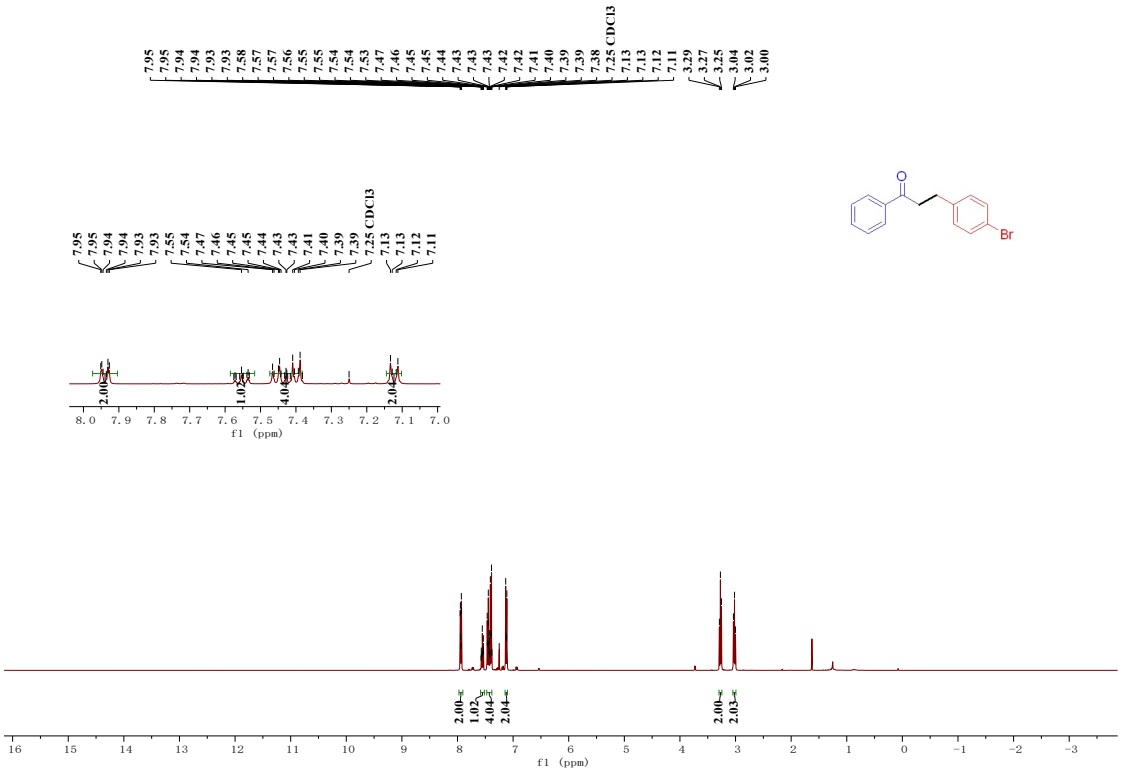


Figure S19. ^{1}H NMR (400 MHz) spectrum of compound 10 in $CDCl_3$

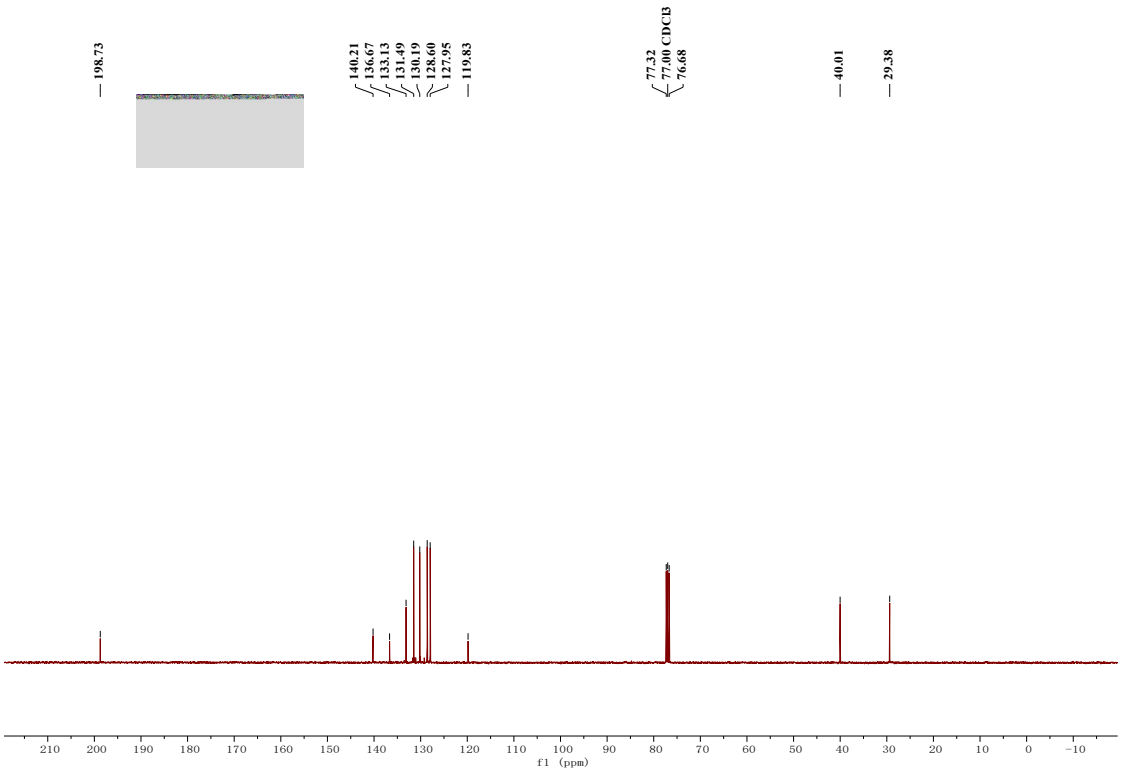


Figure S20. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 10 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **11**

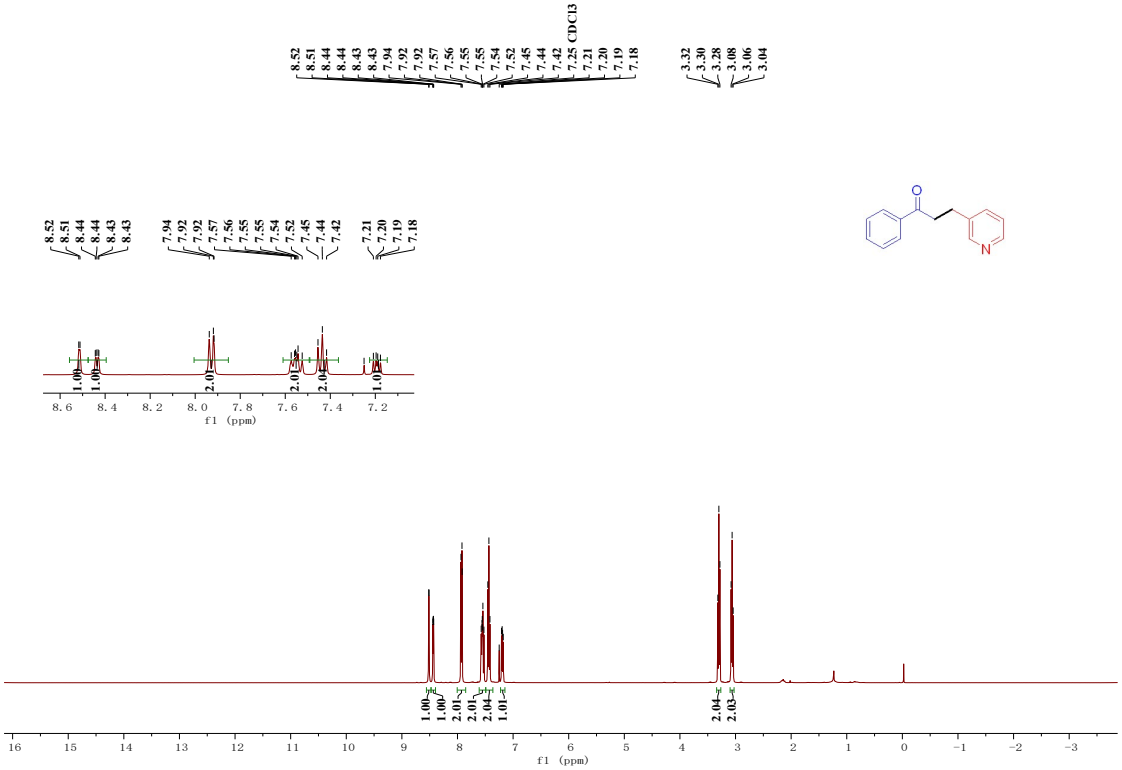


Figure S21. ^{1}H NMR (400 MHz) spectrum of compound 11 in $CDCl_3$

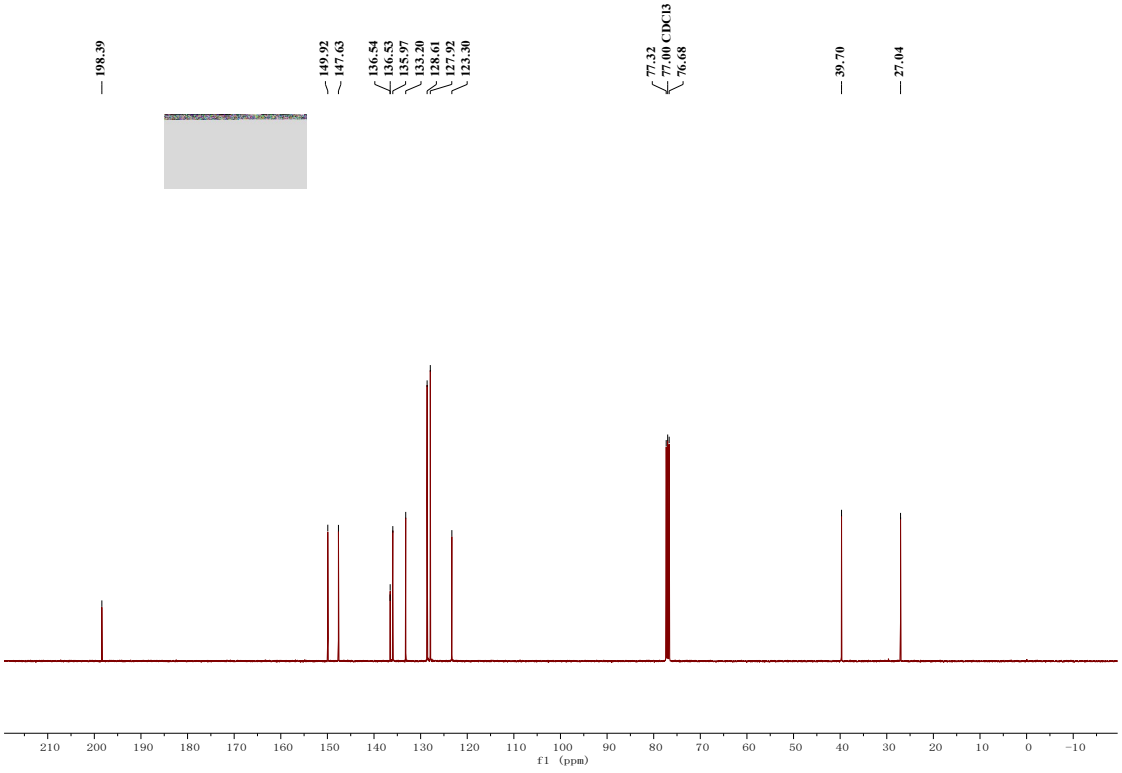


Figure S22. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 11 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **12**

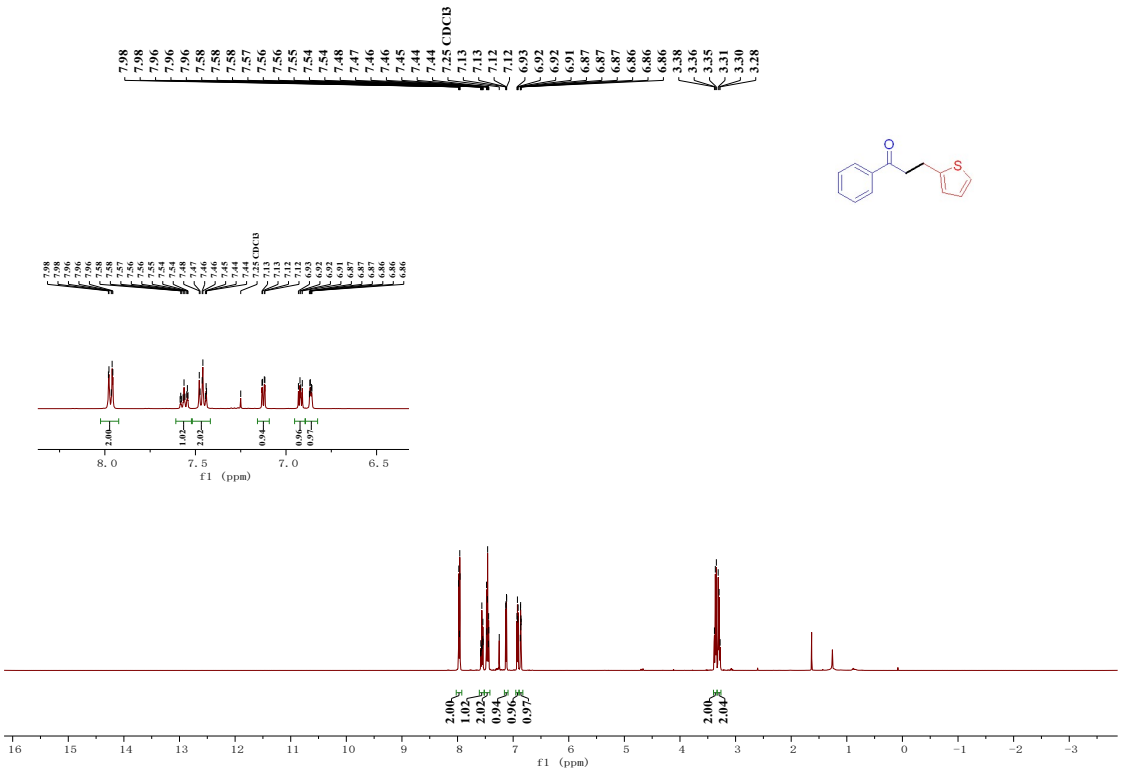


Figure S23. ^{1}H NMR (400 MHz) spectrum of compound 12 in $CDCl_3$

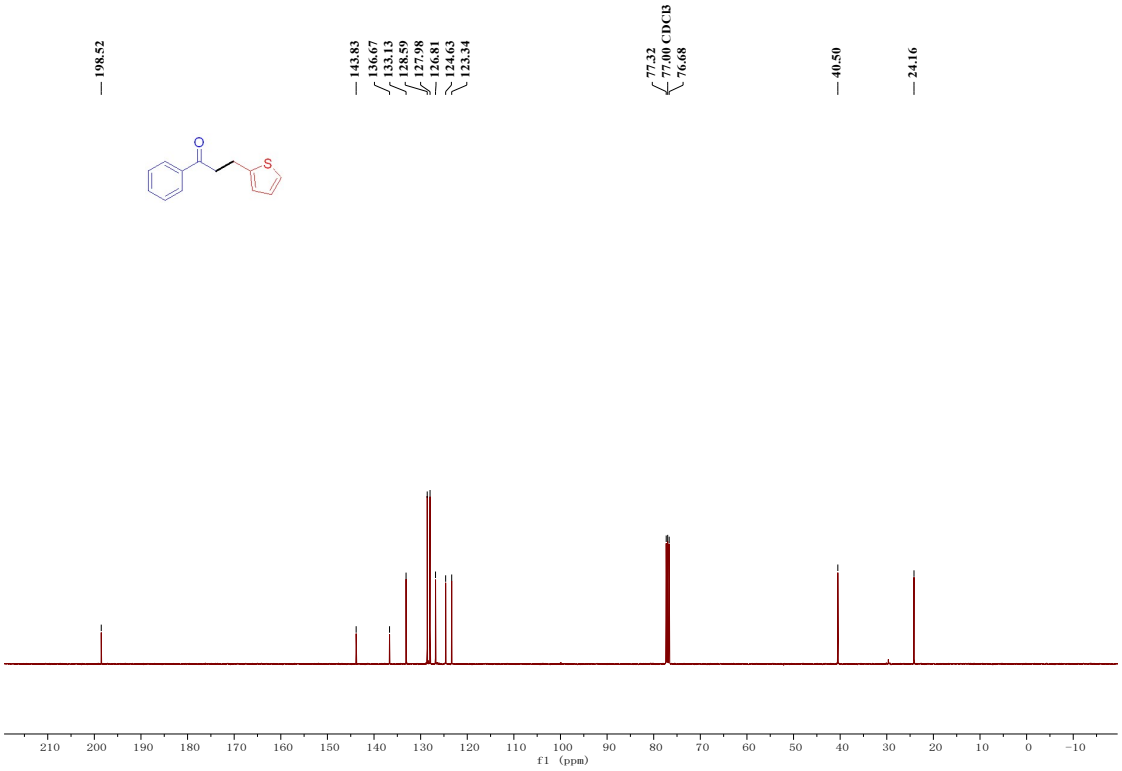


Figure S24. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 12 in $CDCl_3$

^{1}H and ^{13}C{^{1}H} NMR spectra of compound **13**

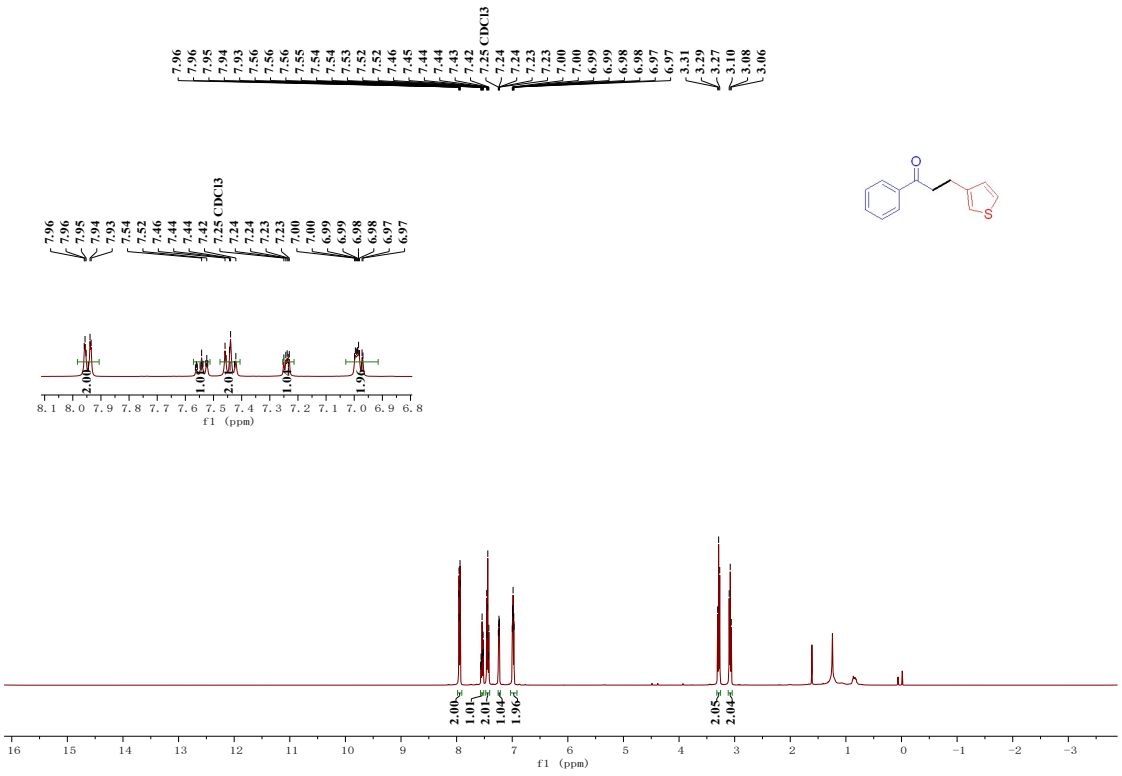


Figure S25. ^{1}H NMR (400 MHz) spectrum of compound 13 in $CDCl_3$

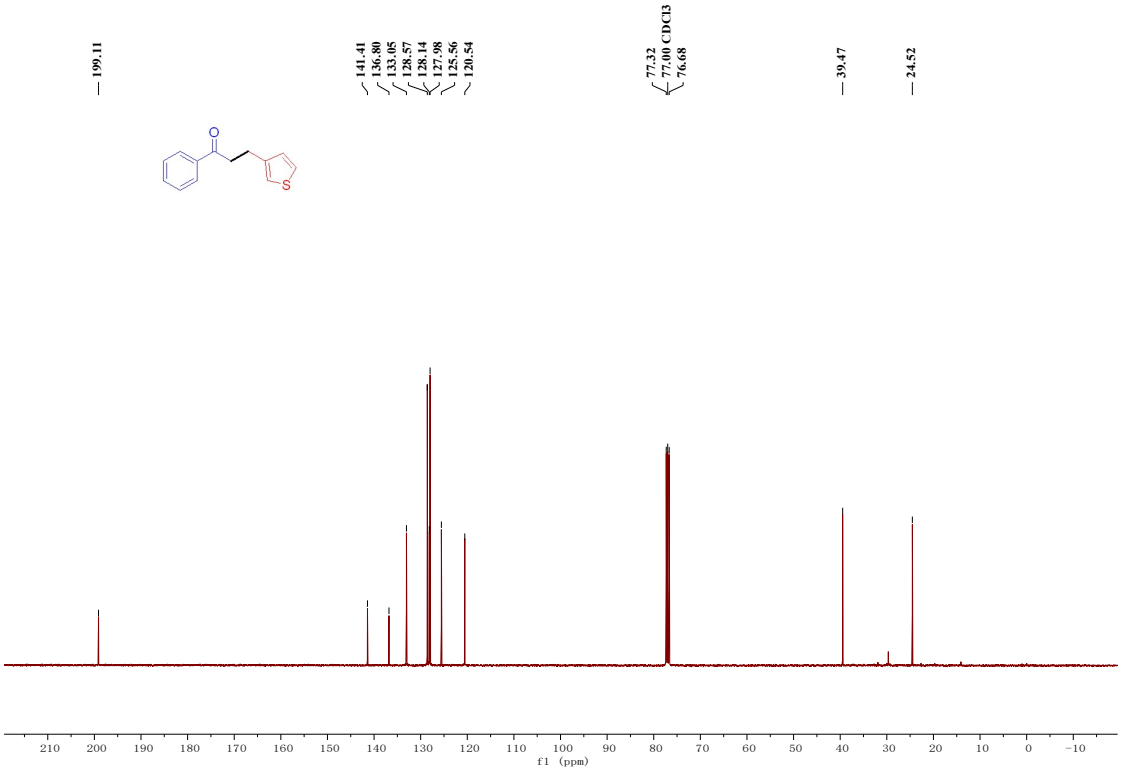


Figure S26. ^{13}C{^{1}H} NMR (101 MHz) spectrum of compound 13 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **14**

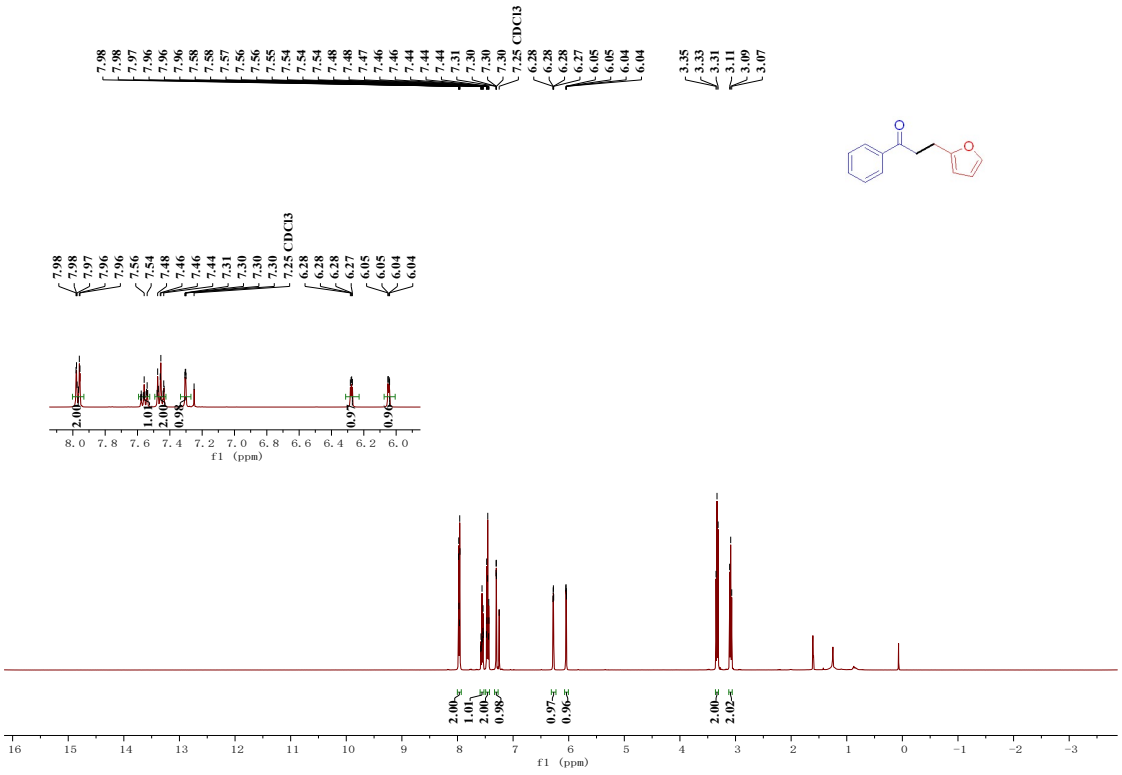


Figure S27. ^{1}H NMR (400 MHz) spectrum of compound 14 in $CDCl_3$

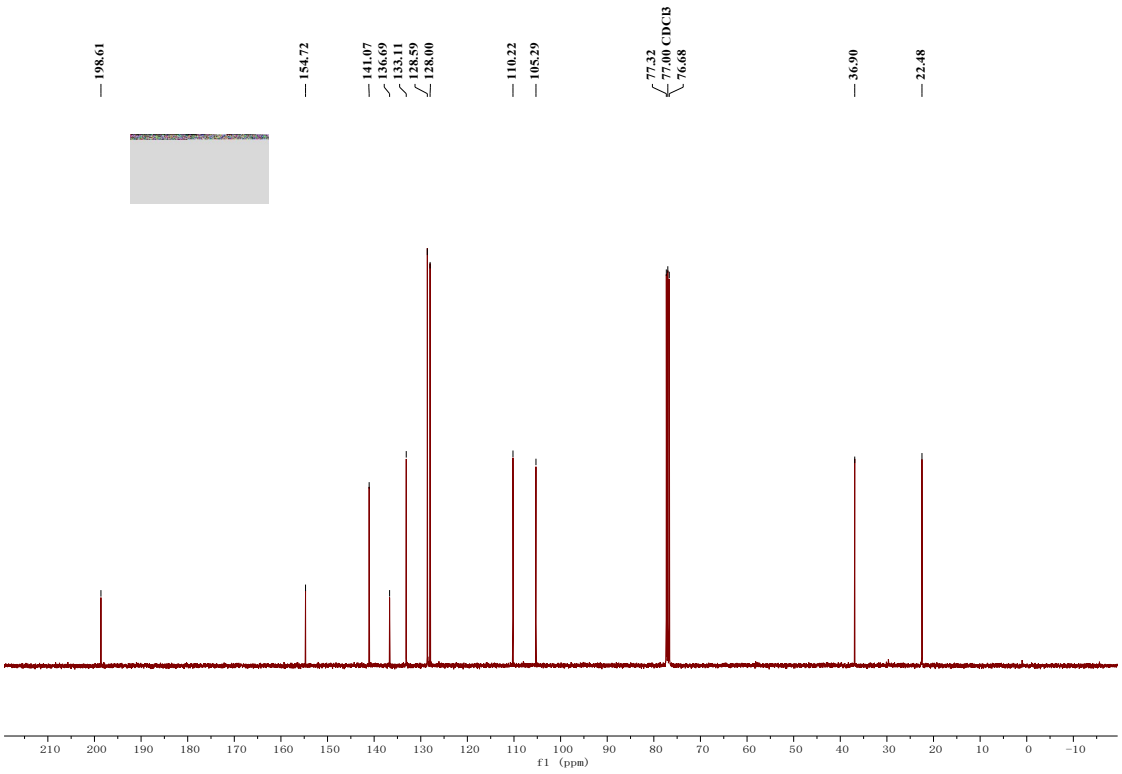


Figure S28. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 14 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **15**

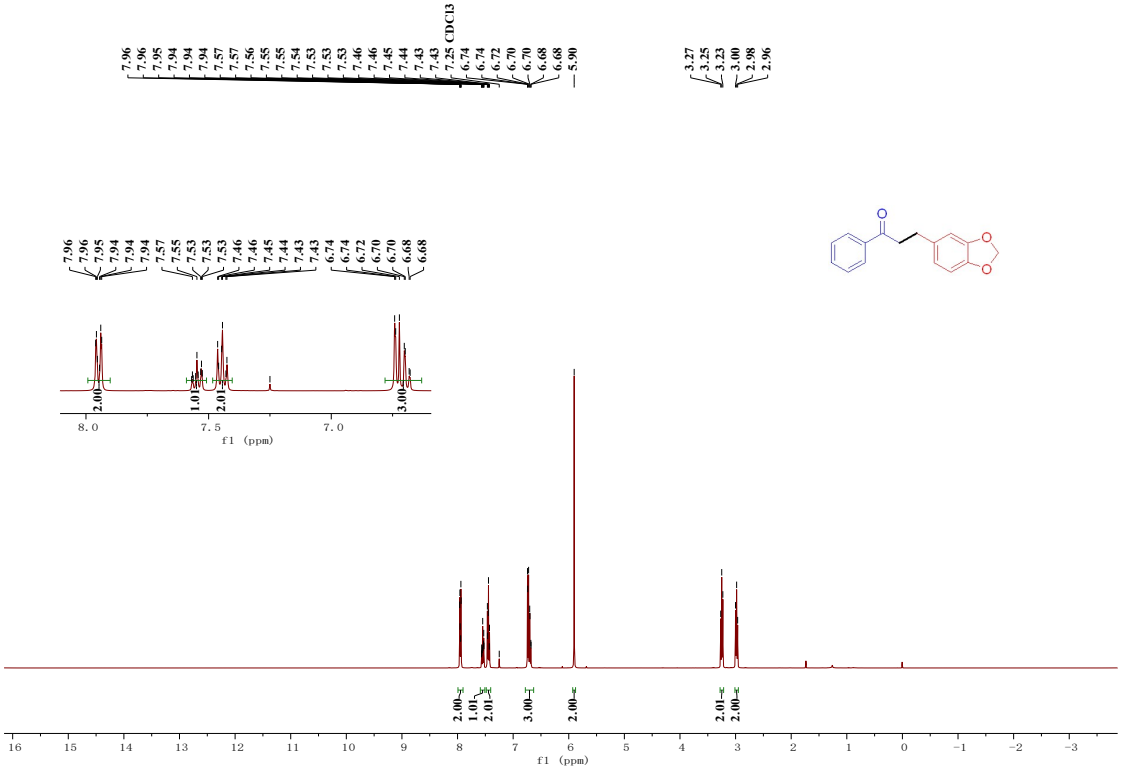


Figure S29. ^{1}H NMR (400 MHz) spectrum of compound 15 in $CDCl_3$

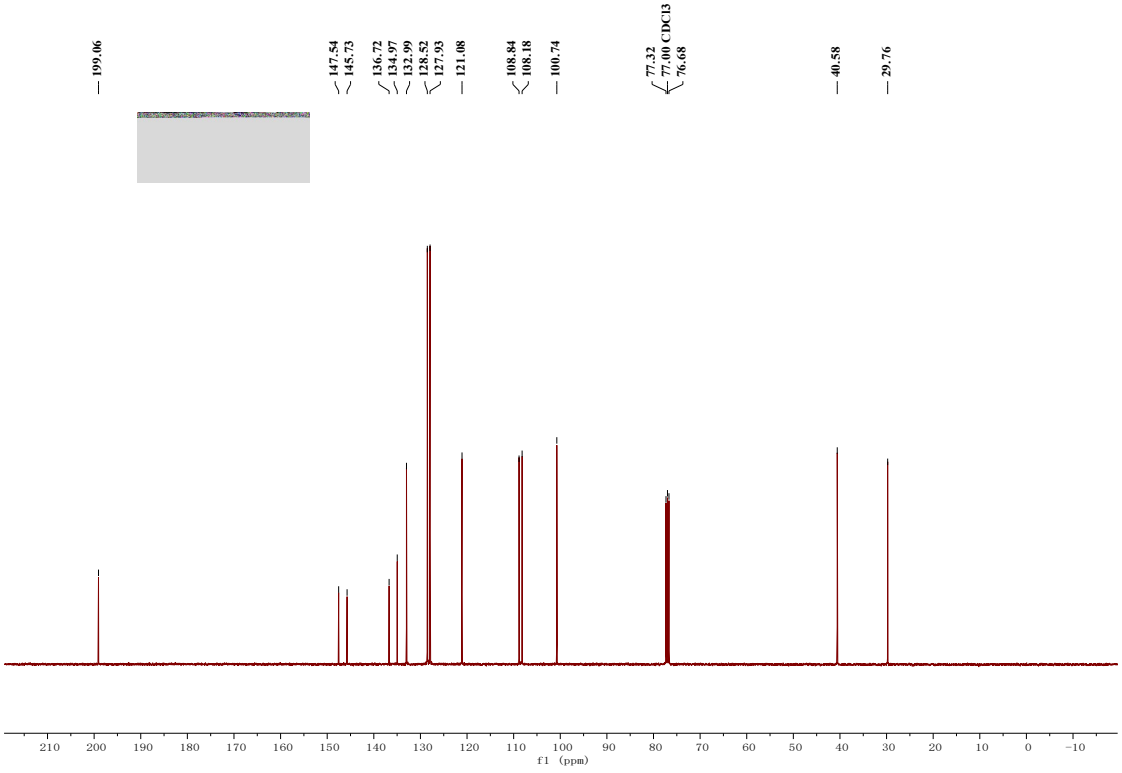


Figure S30. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 15 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **16**

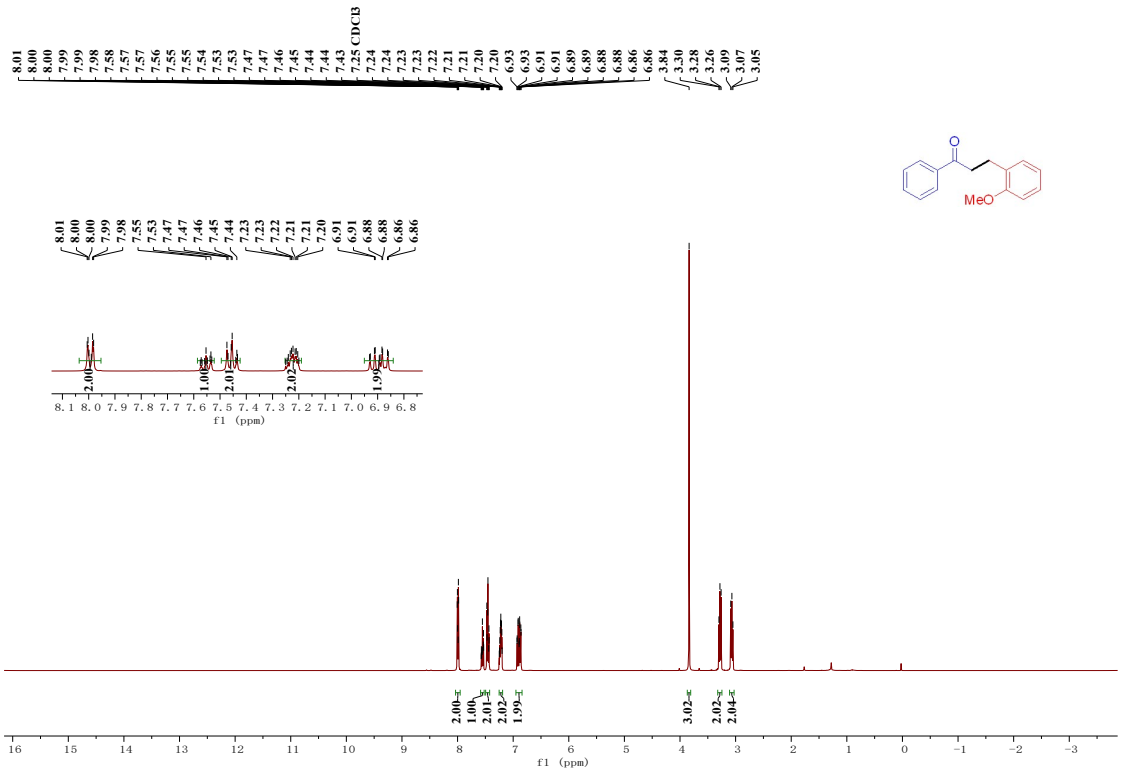


Figure S31. ^{1}H NMR (400 MHz) spectrum of compound 16 in $CDCl_3$

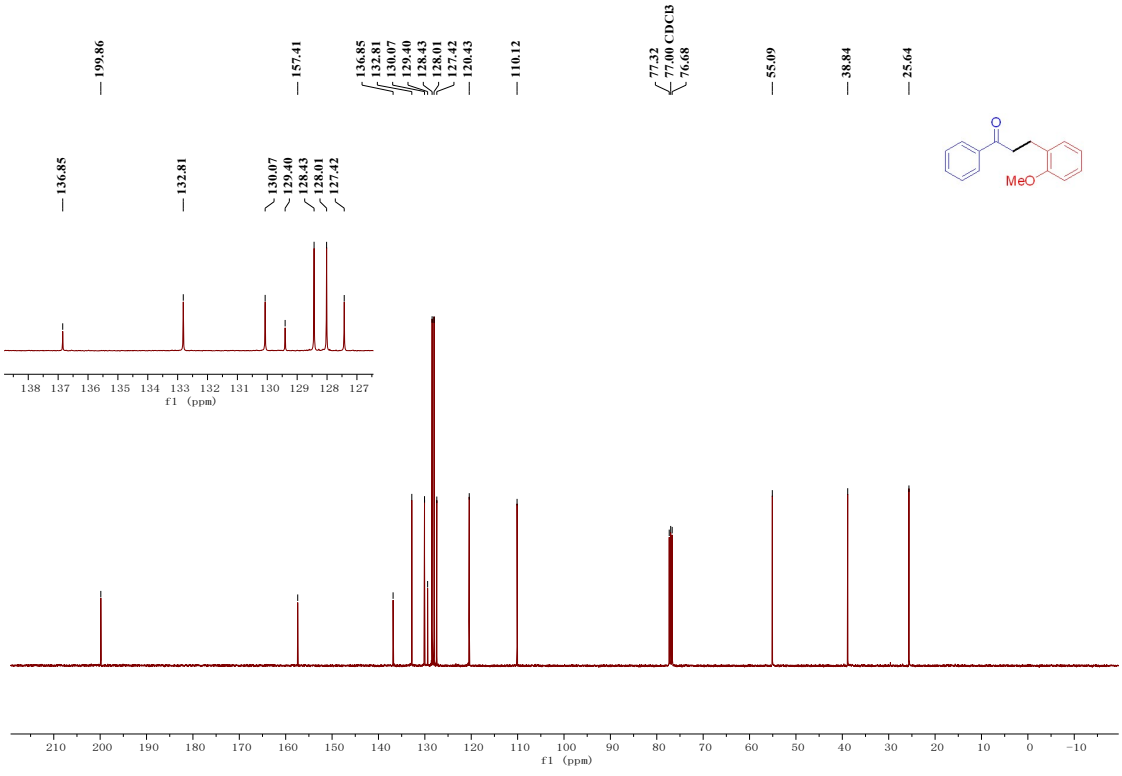


Figure S32. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 16 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **17**

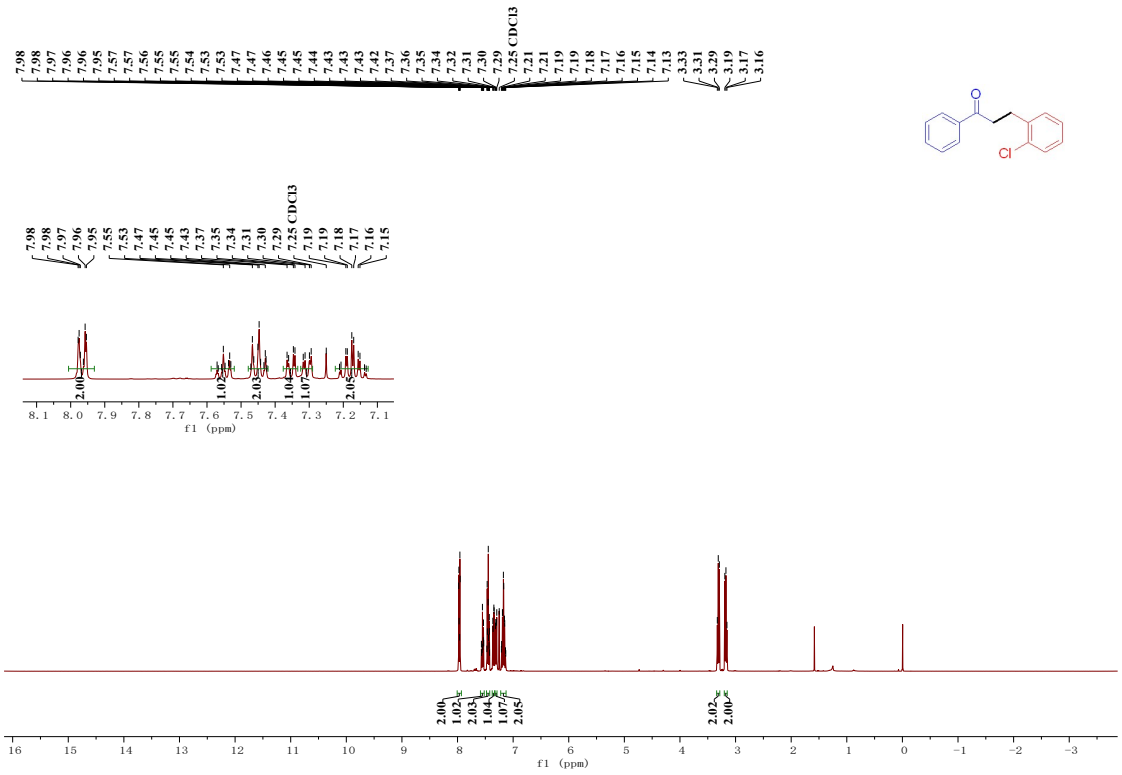


Figure S33. ^{1}H NMR (400 MHz) spectrum of compound 17 in $CDCl_3$

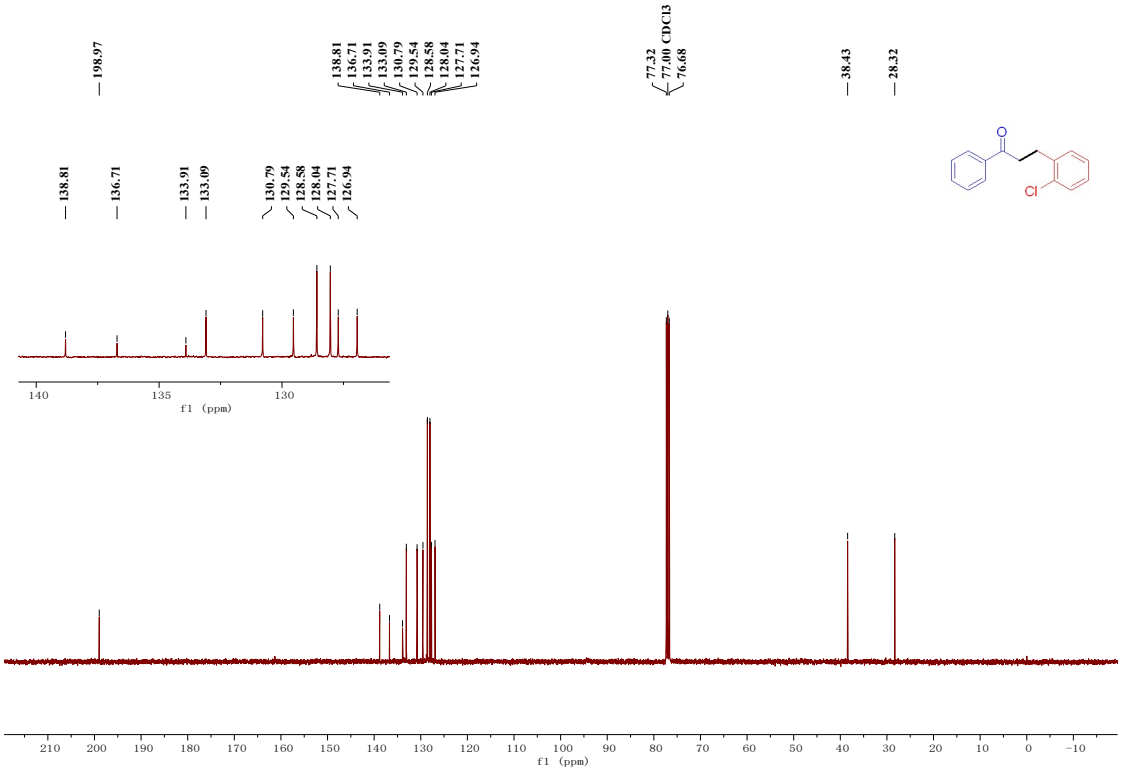


Figure S34. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 17 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **18**

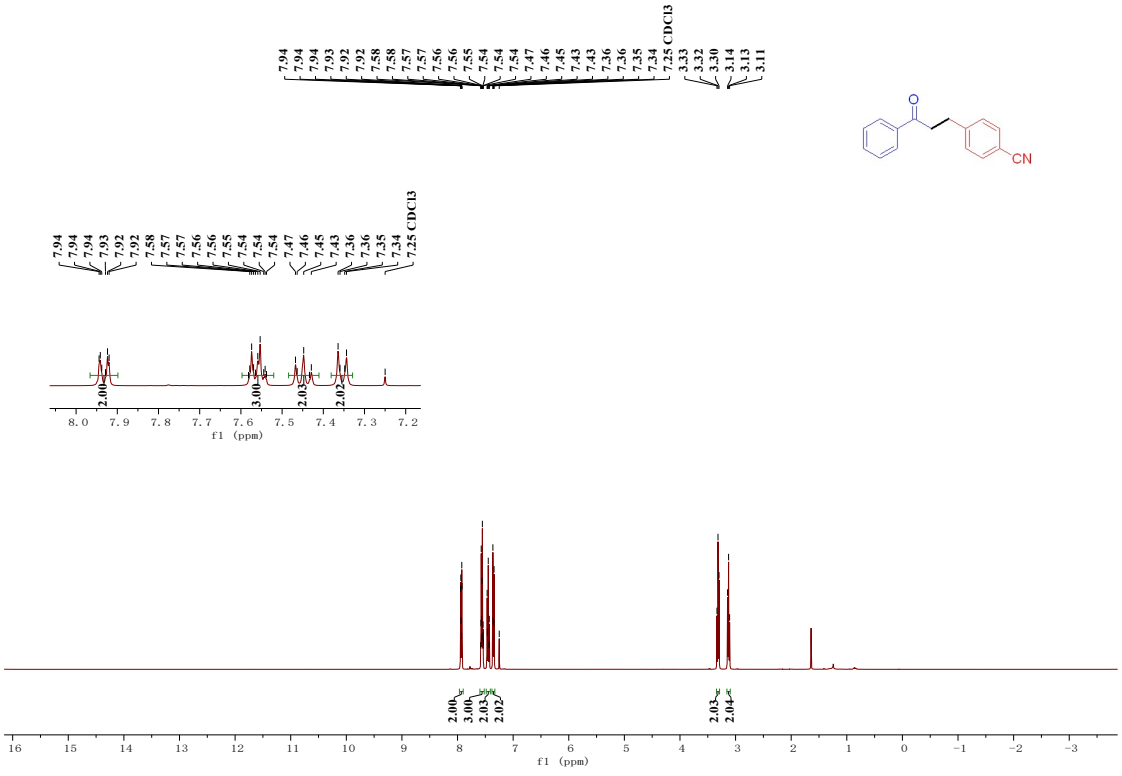


Figure S35. ^{1}H NMR (400 MHz) spectrum of compound 18 in $CDCl_3$

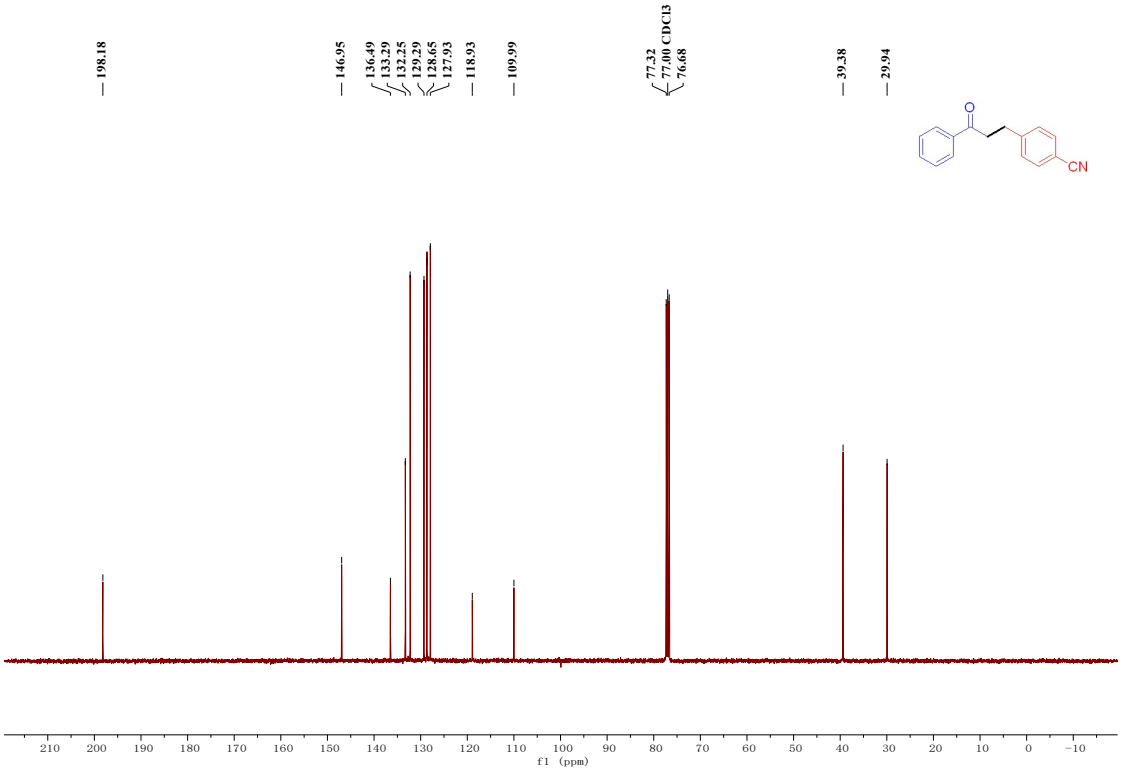


Figure S36. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 18 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **19**

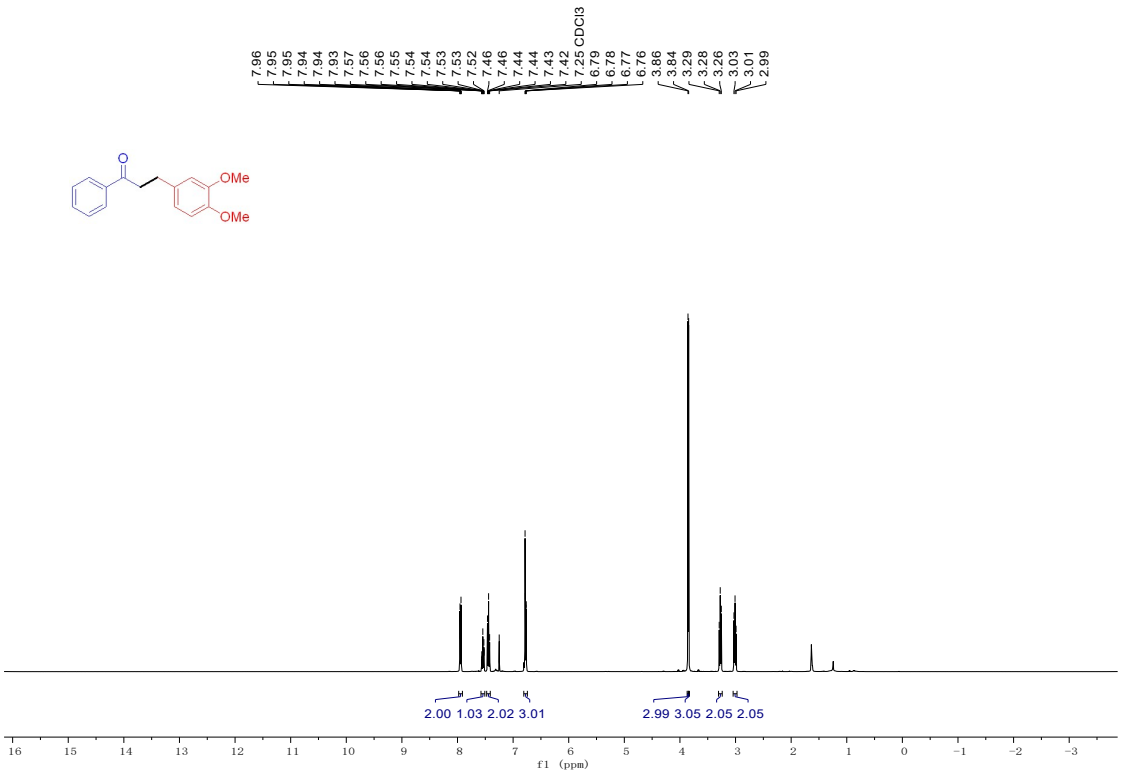


Figure S37. ^{1}H NMR (400 MHz) spectrum of compound 19 in $CDCl_3$

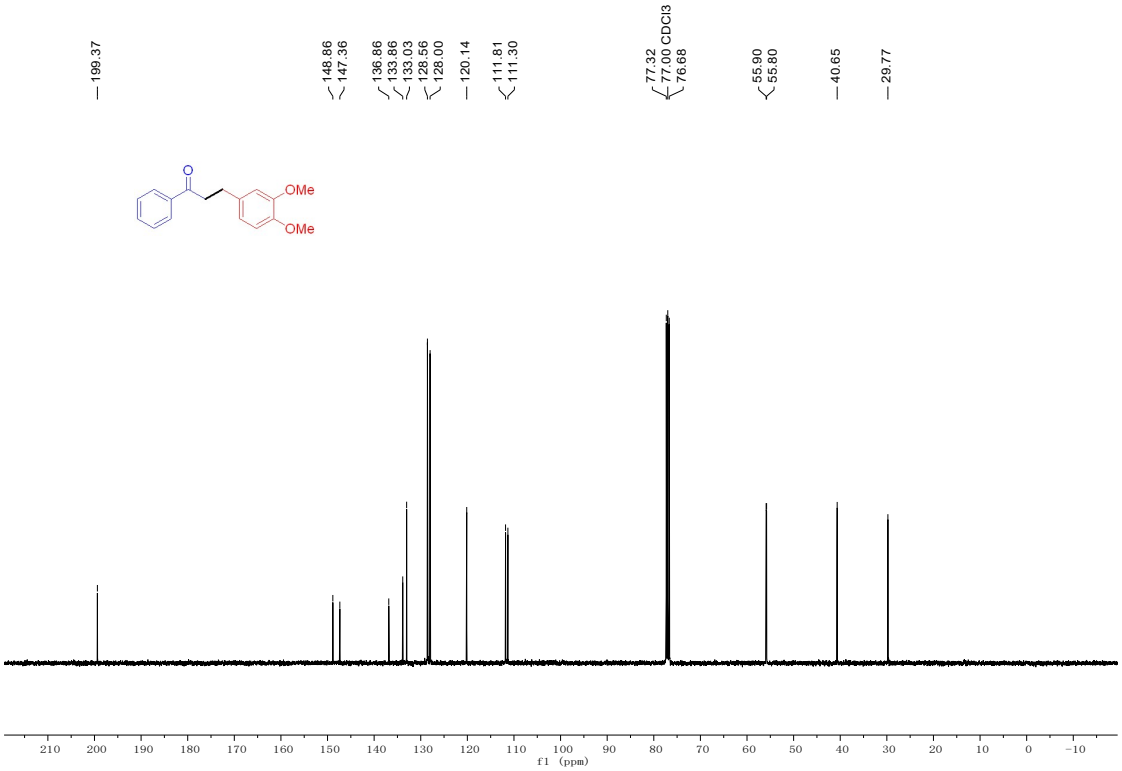


Figure S38. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 19 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **20**

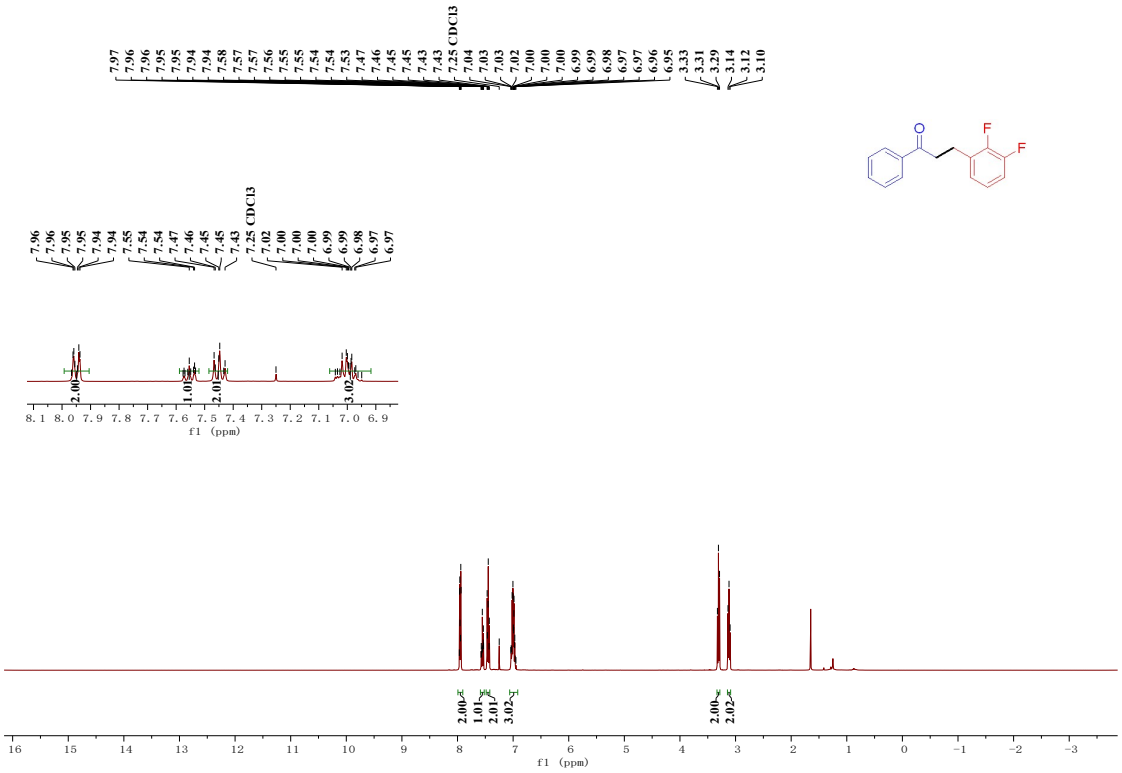


Figure S39. ^{1}H NMR (400 MHz) spectrum of compound 20 in $CDCl_3$

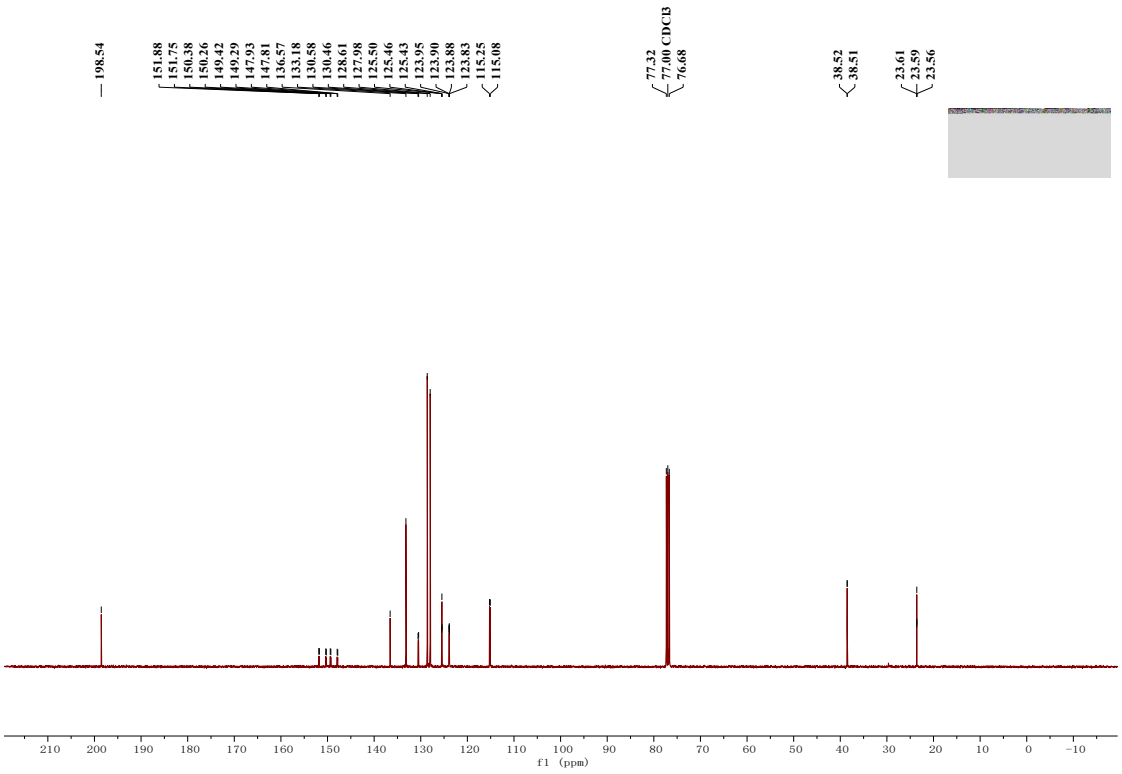


Figure S40. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 20 in $CDCl_3$

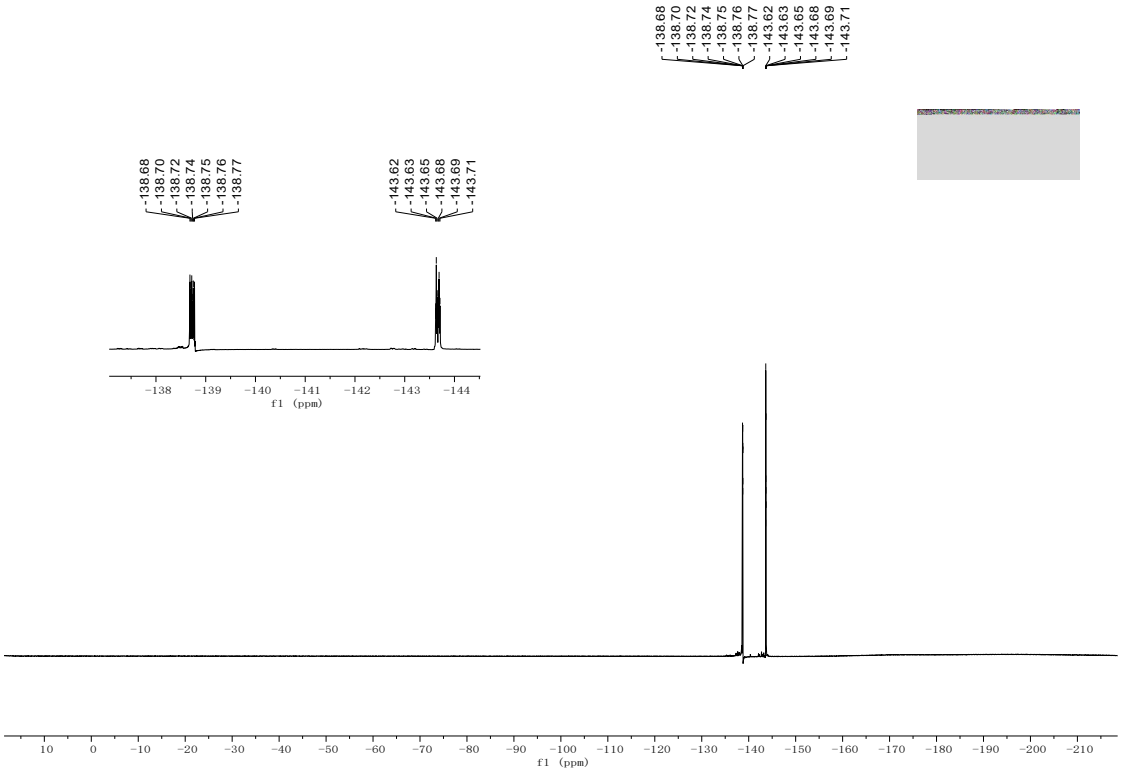


Figure S41. ^{19}F NMR (376 MHz) spectrum of compound 20 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **21**

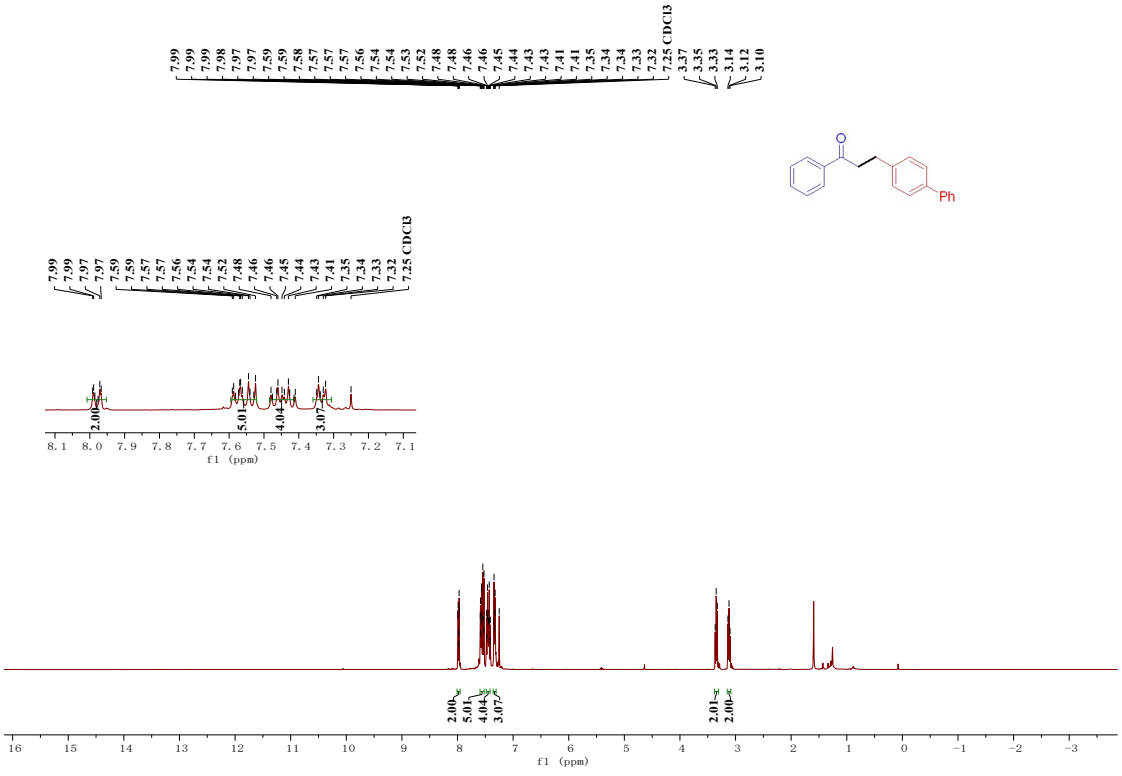


Figure S42. ^{1}H NMR (400 MHz) spectrum of compound 21 in $CDCl_3$

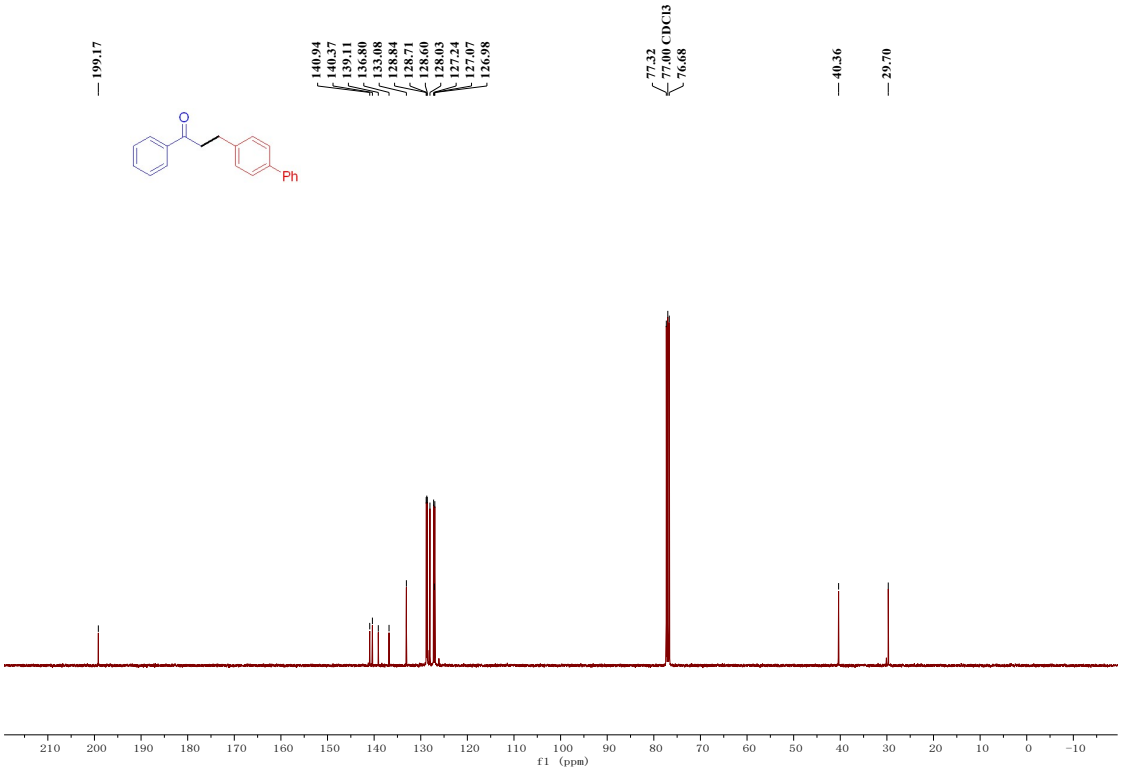


Figure S43. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 21 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **22**

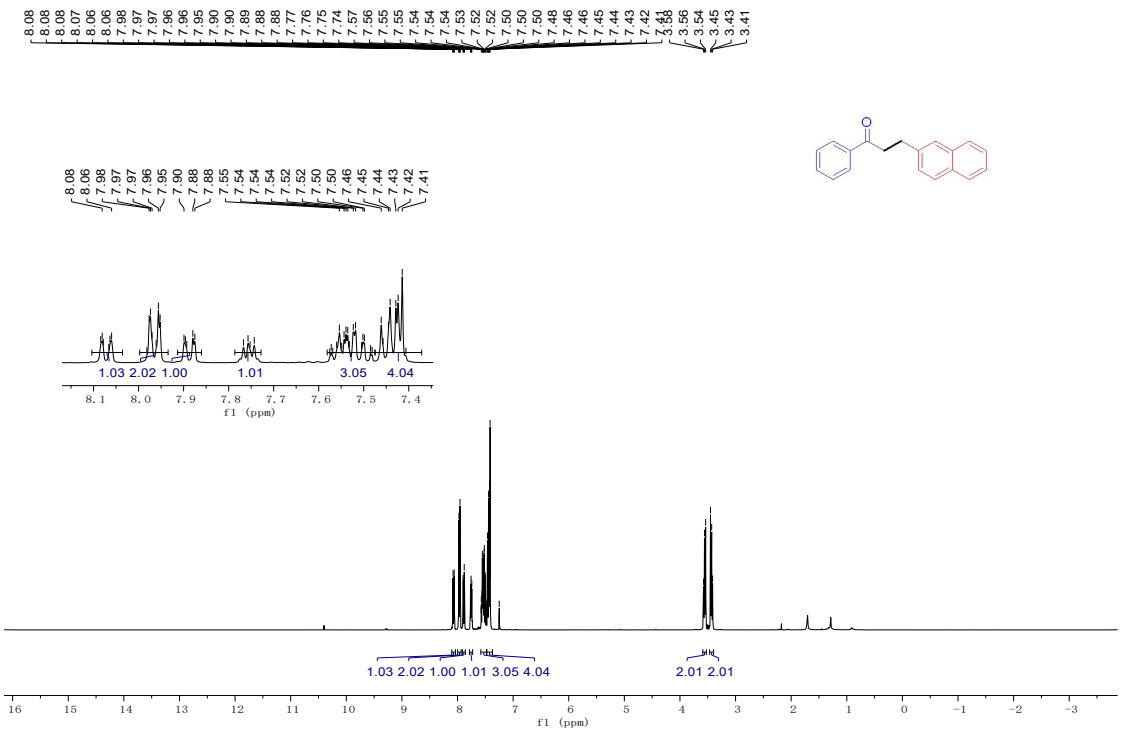


Figure S44. ^{1}H NMR (400 MHz) spectrum of compound 22 in $CDCl_3$

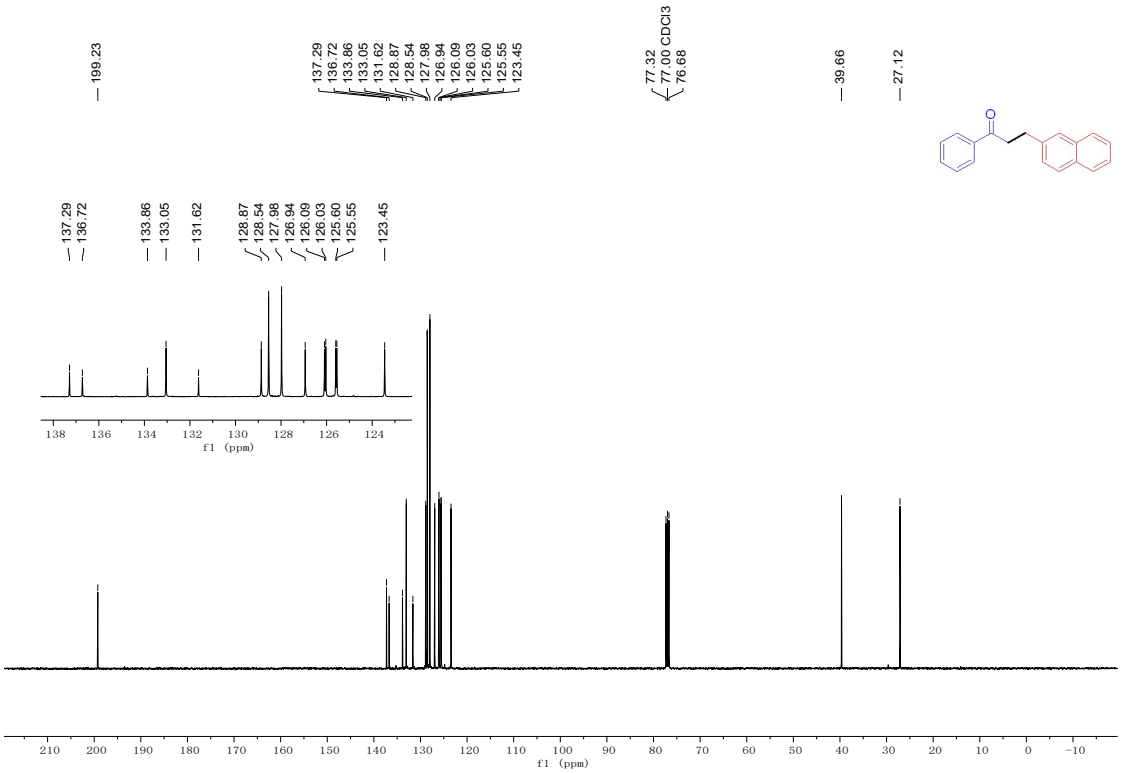


Figure S45. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 22 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **23**

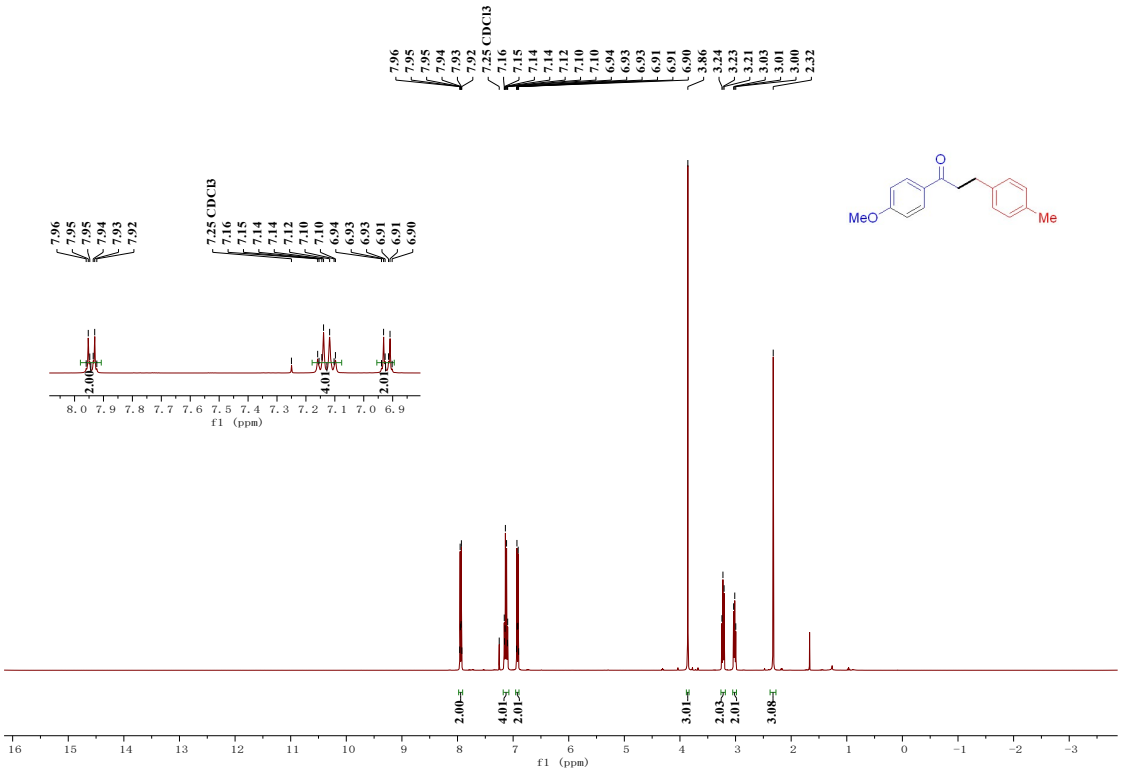


Figure S46. ^{1}H NMR (400 MHz) spectrum of compound 23 in $CDCl_3$

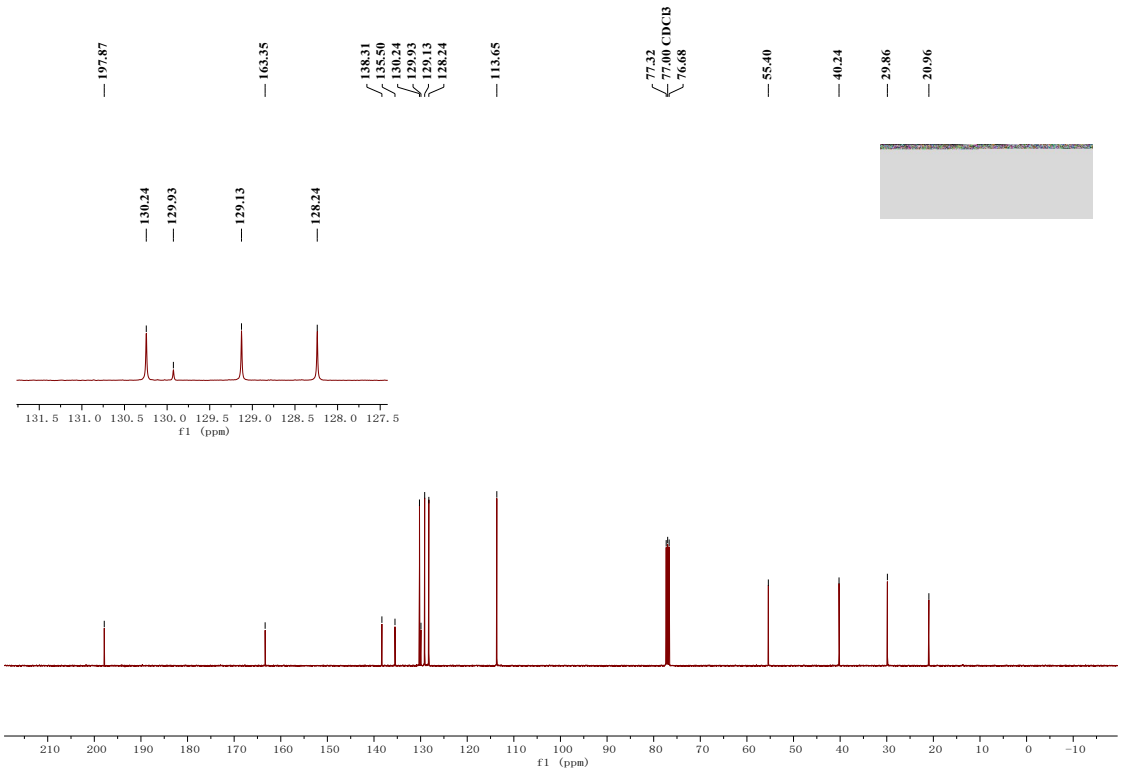


Figure S47. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 23 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **24**

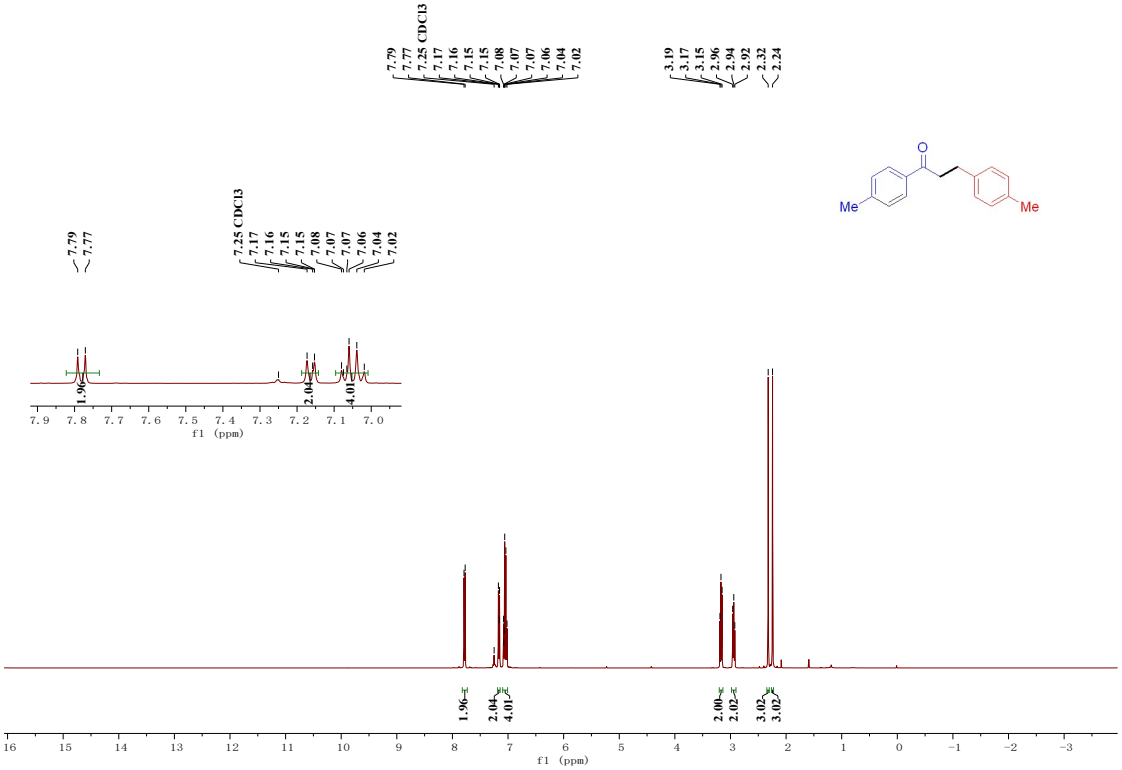


Figure S48. ^{1}H NMR (400 MHz) spectrum of compound 24 in $CDCl_3$

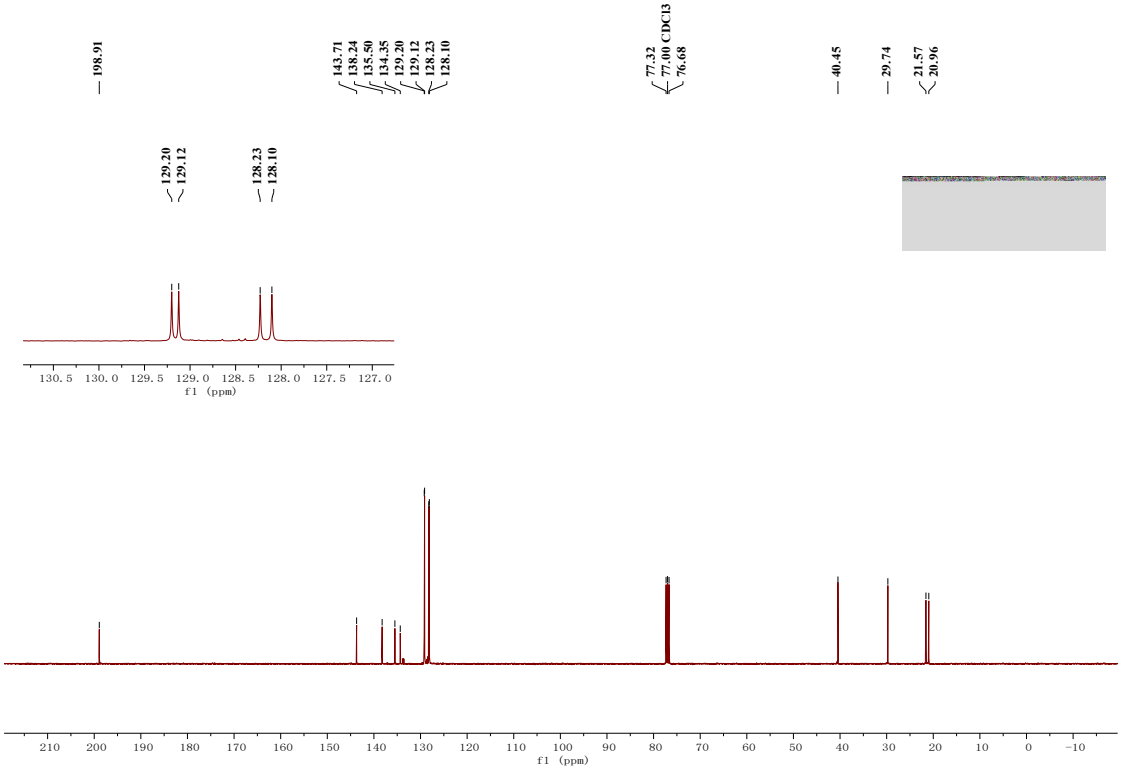


Figure S49. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 24 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **25**

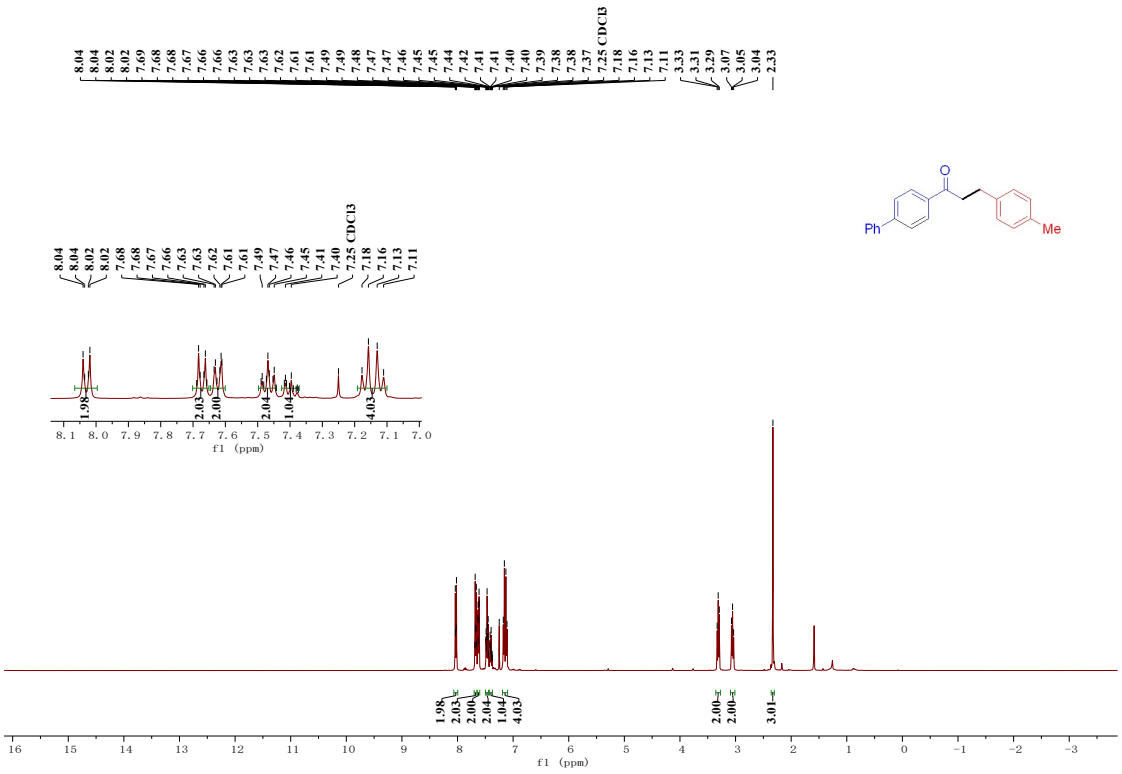


Figure S50. ^{1}H NMR (400 MHz) spectrum of compound 25 in $CDCl_3$

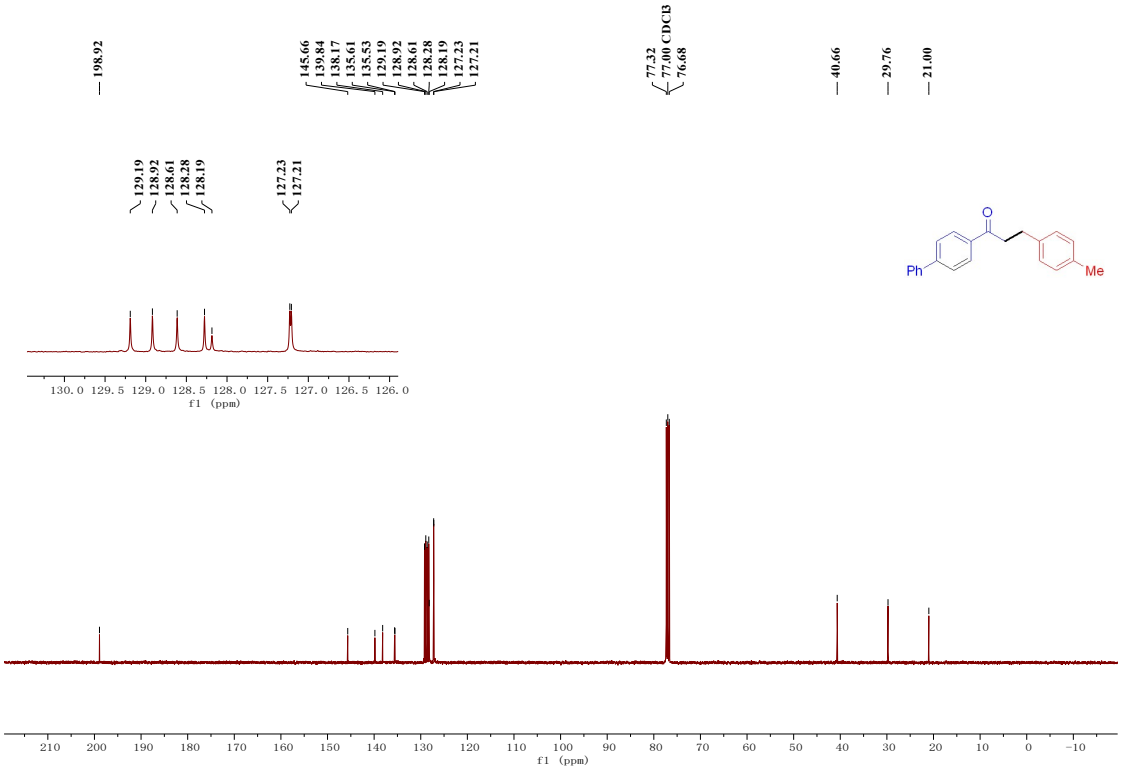


Figure S51. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 25 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **26**

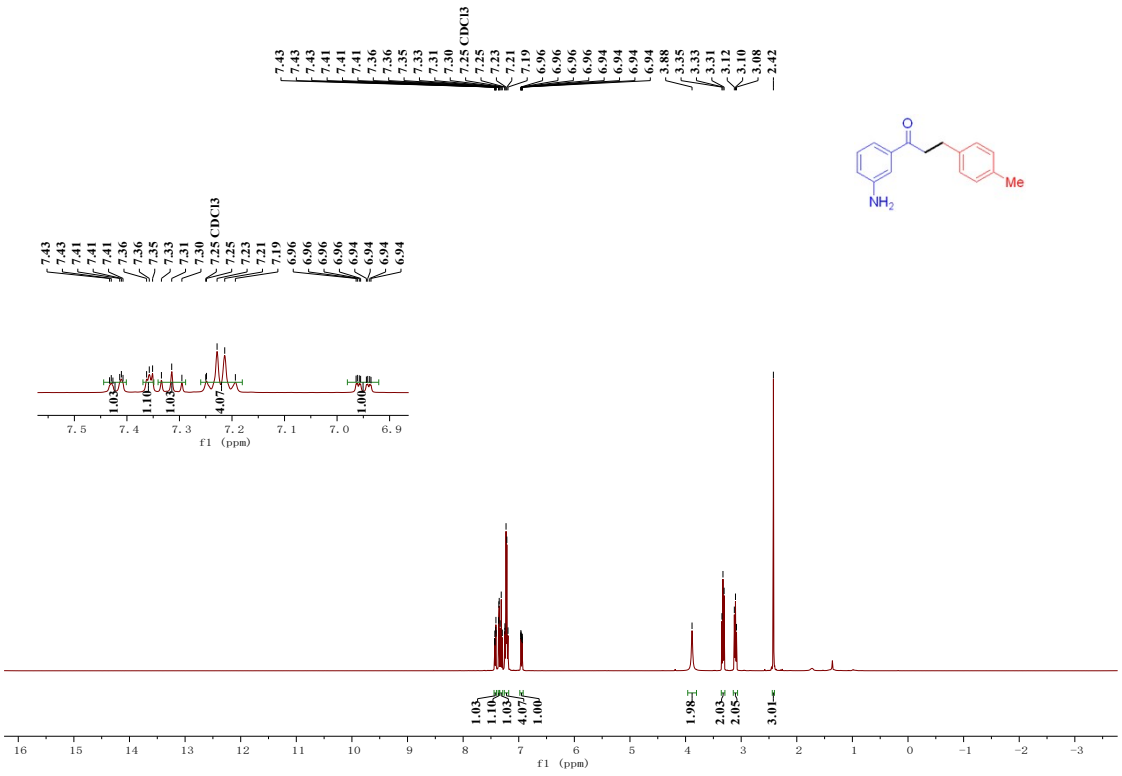


Figure S52. ^{1}H NMR (400 MHz) spectrum of compound 26 in $CDCl_3$

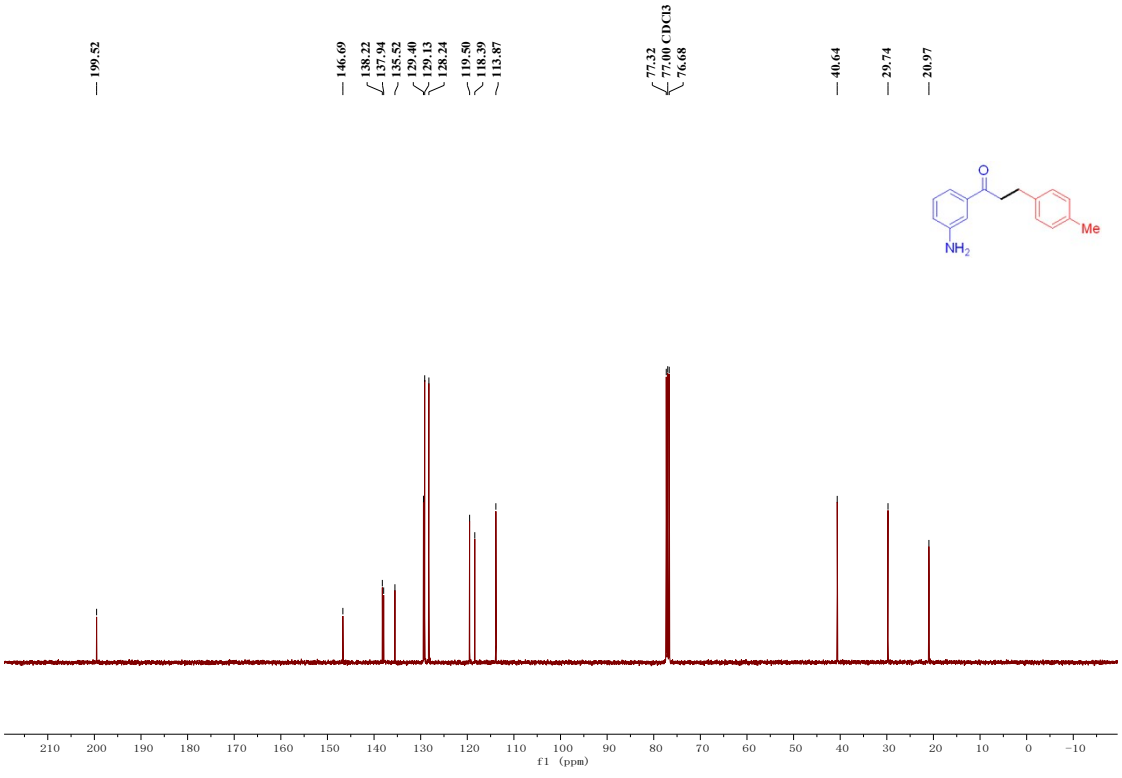


Figure S53. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 26 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **27**

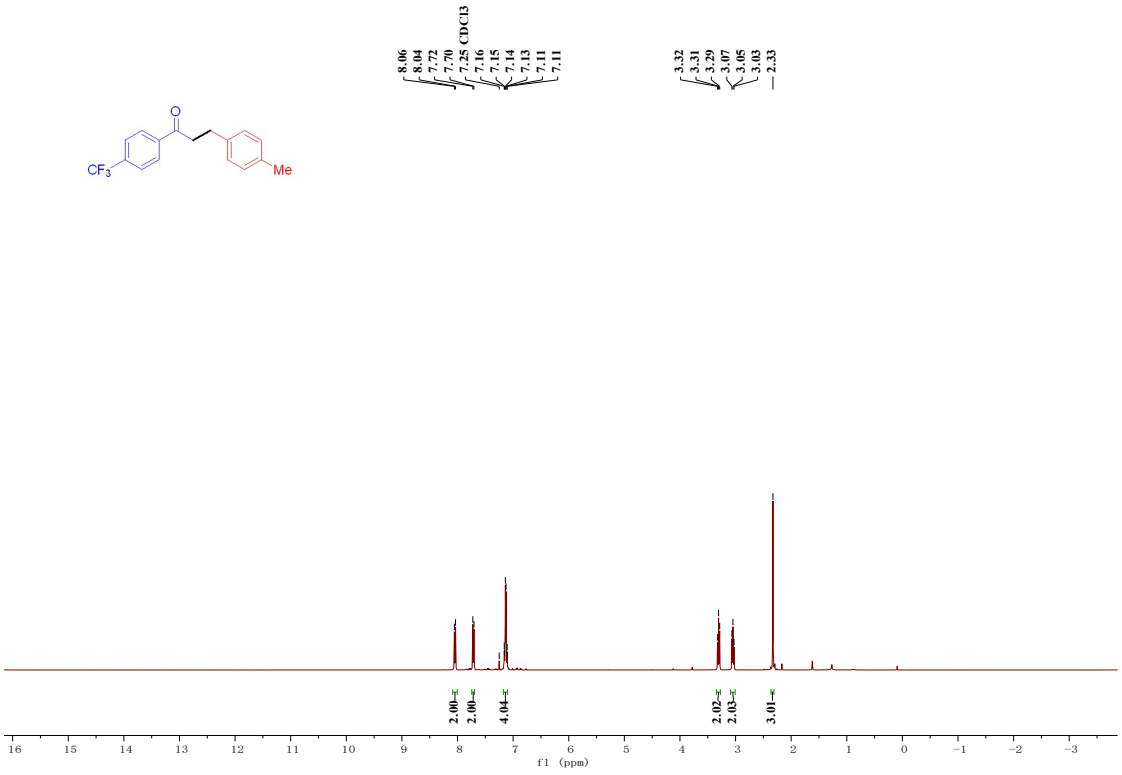


Figure S54. ^{1}H NMR (400 MHz) spectrum of compound 27 in $CDCl_3$

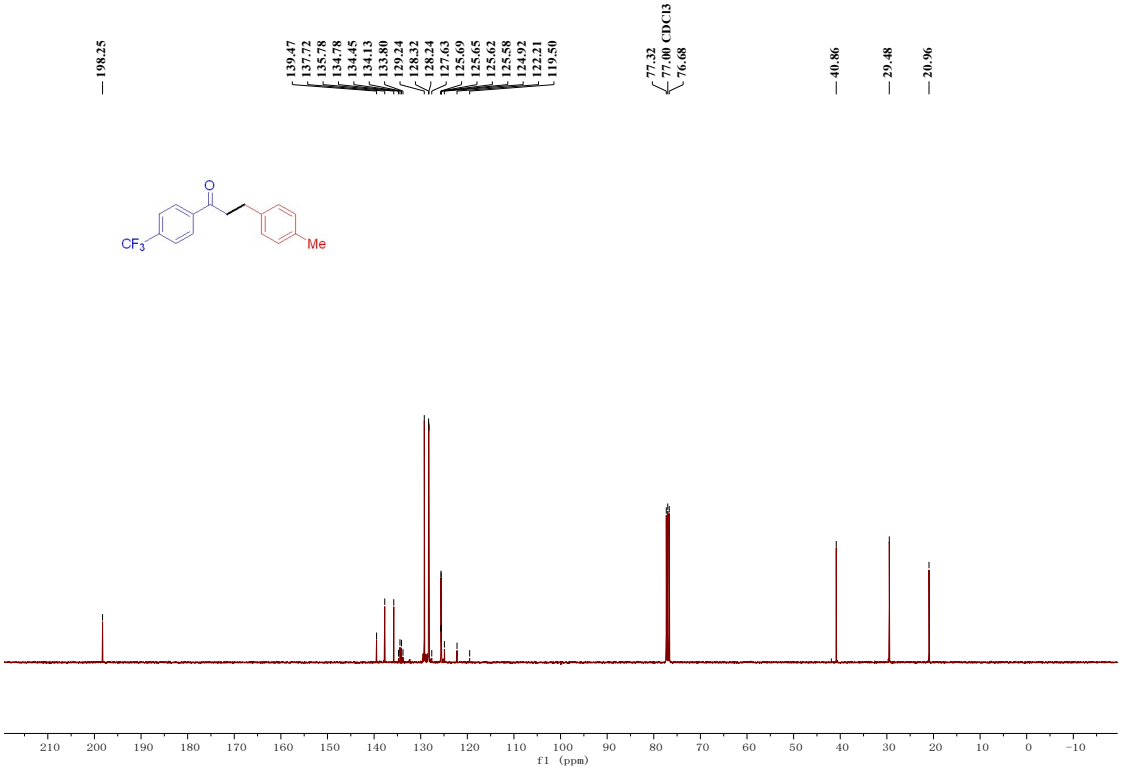


Figure S55. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 27 in $CDCl_3$

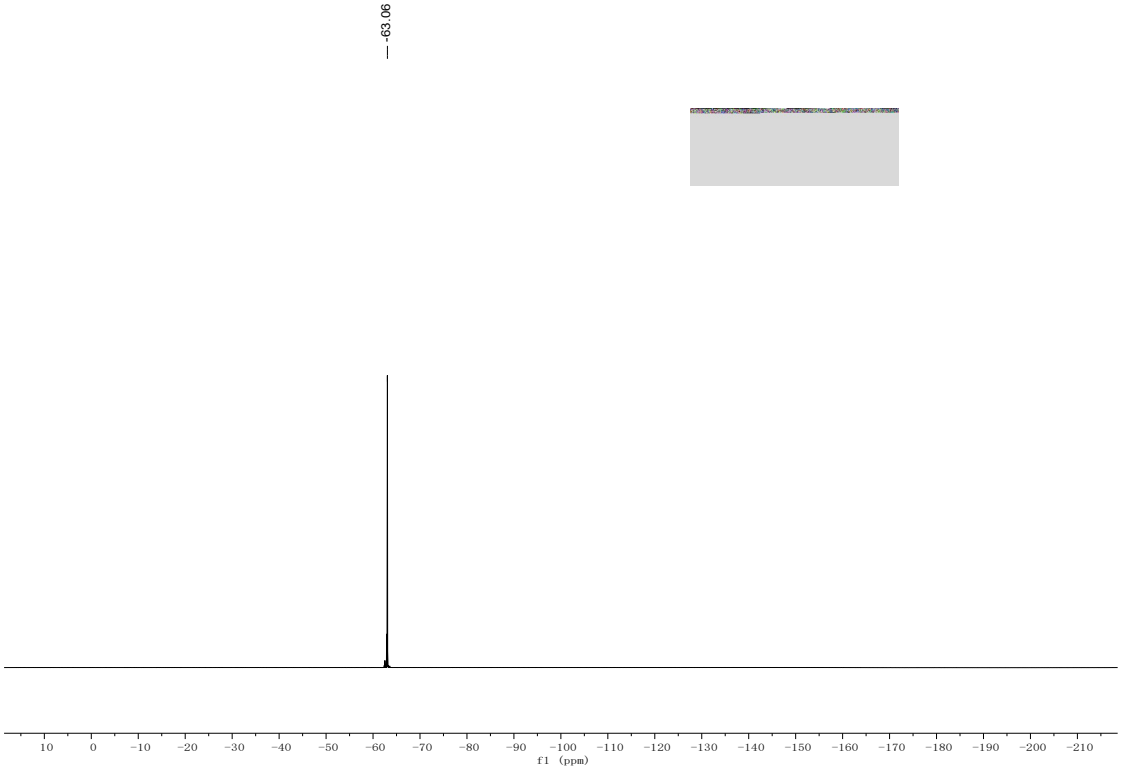


Figure S56. ^{19}F NMR (376 MHz) spectrum of compound 27 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **28**

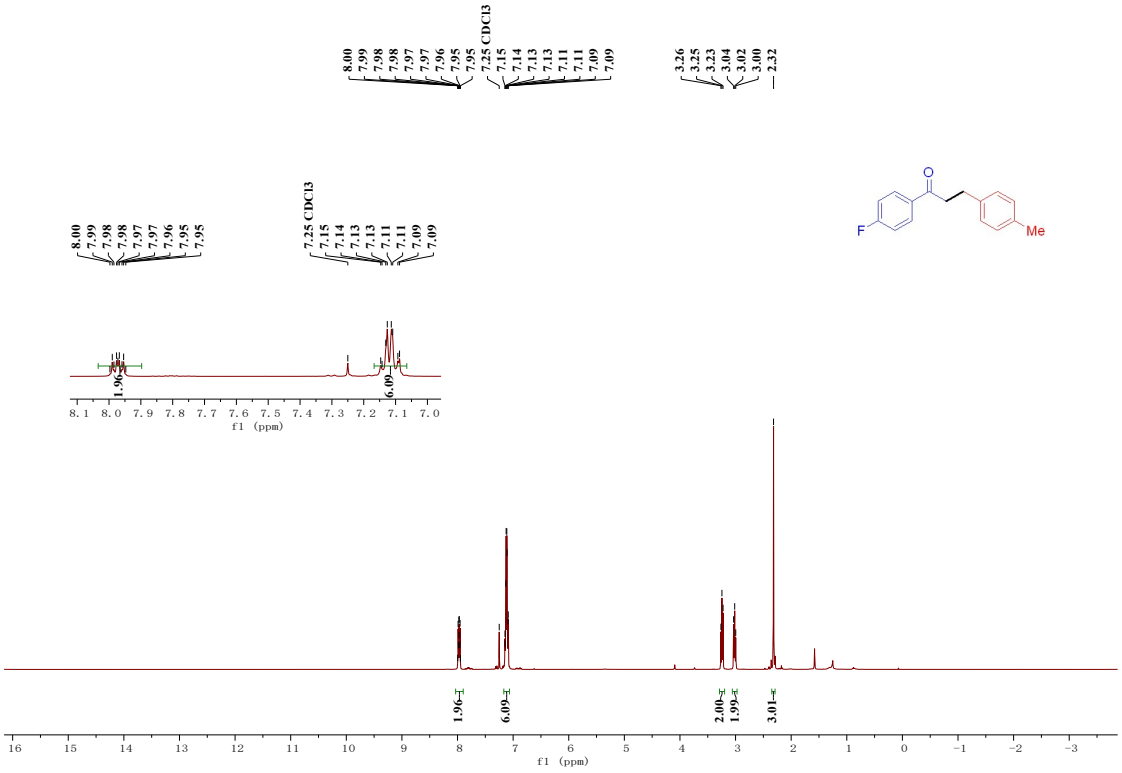


Figure S57. ^{1}H NMR (400 MHz) spectrum of compound 28 in $CDCl_3$

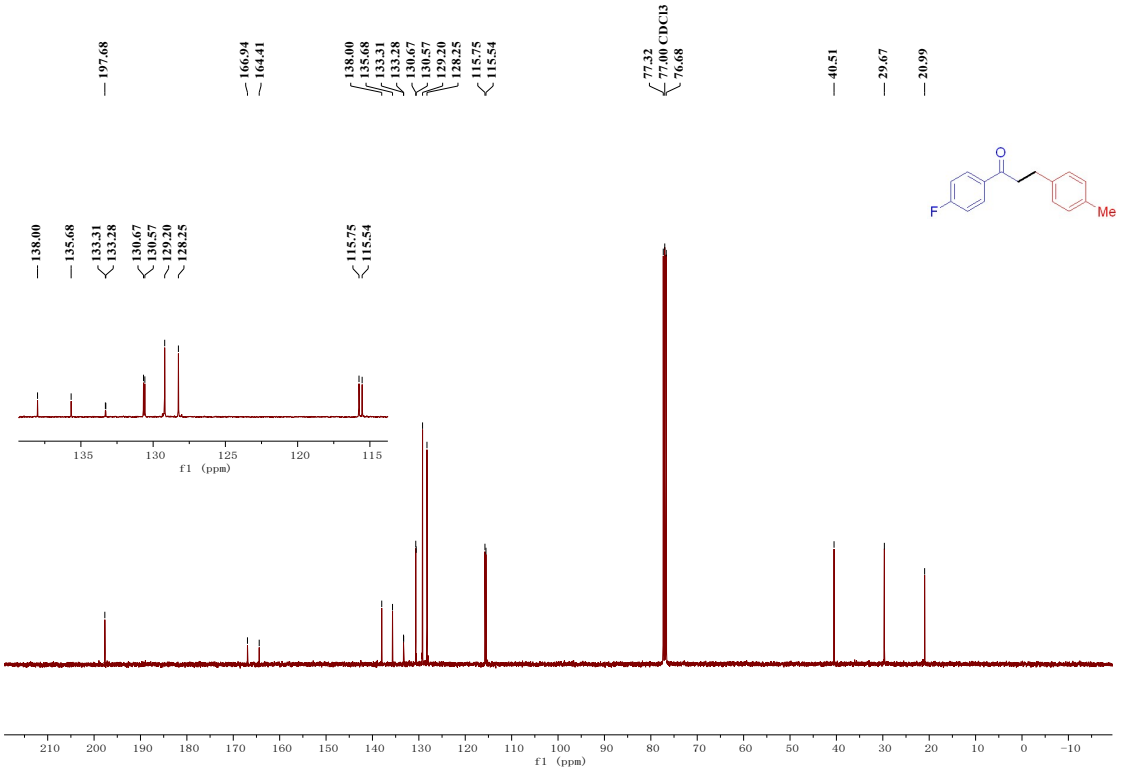


Figure S58. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 28 in $CDCl_3$

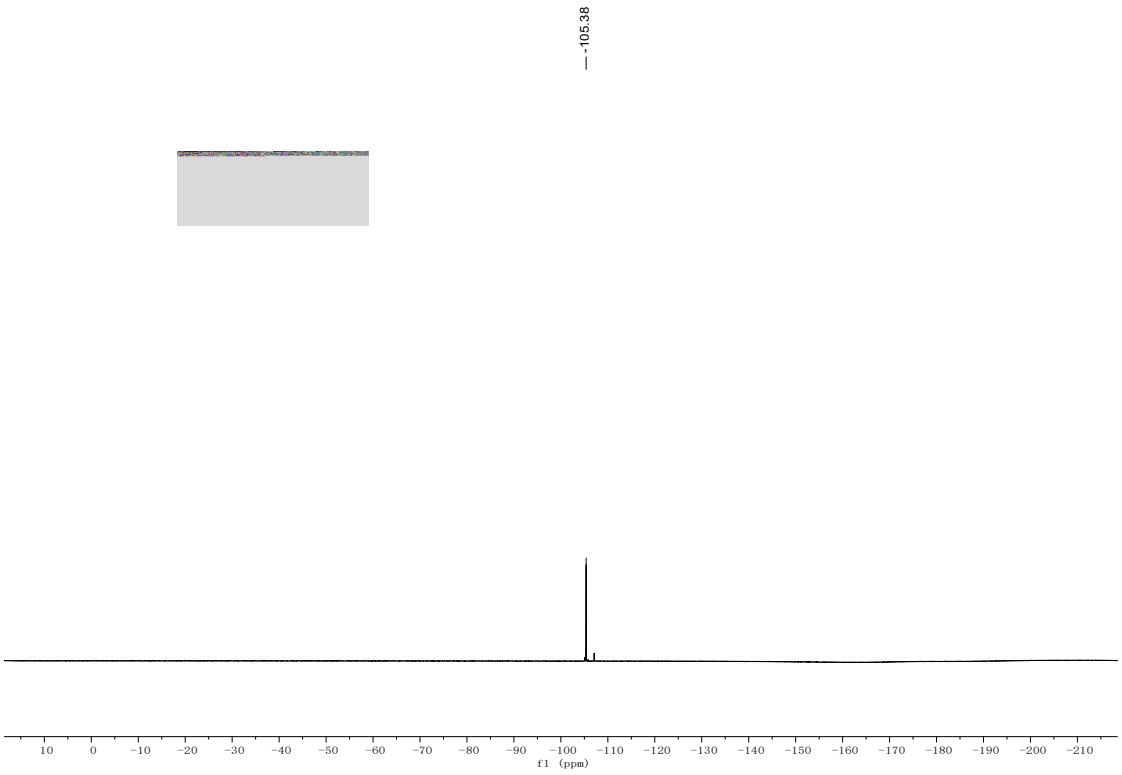


Figure S59. ^{19}F NMR (376 MHz) spectrum of compound 28 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **29**

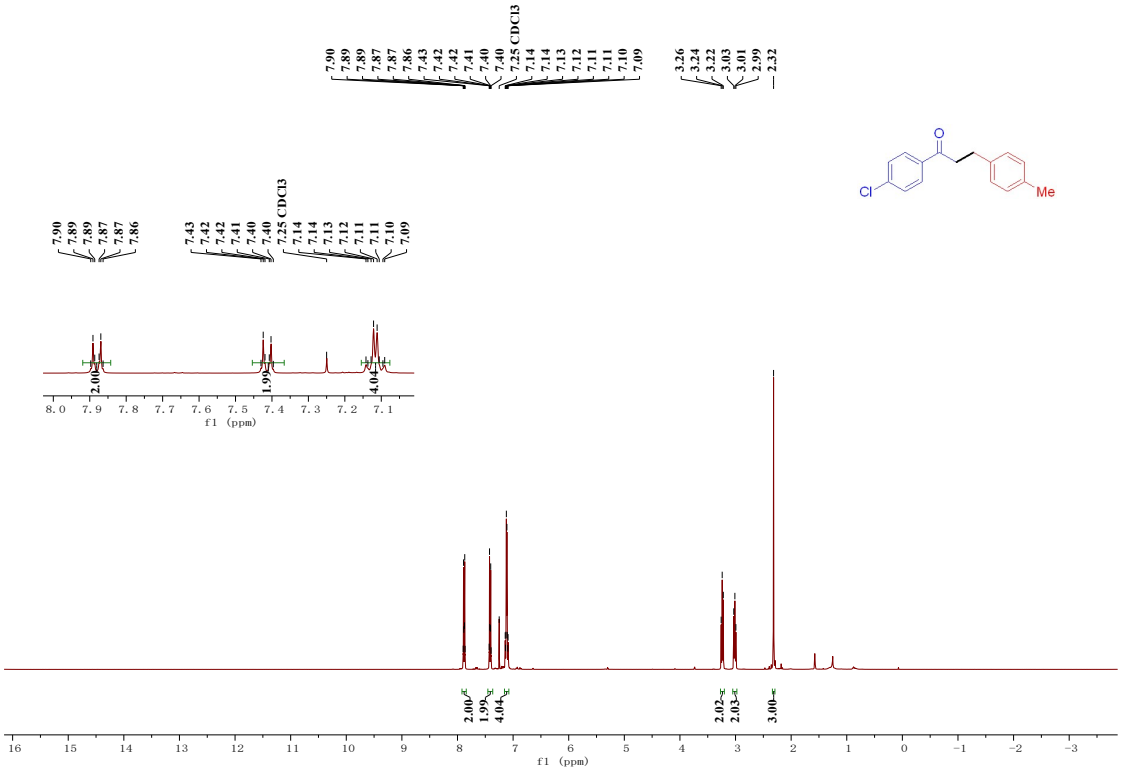


Figure S60. ^{1}H NMR (400 MHz) spectrum of compound 29 in $CDCl_3$

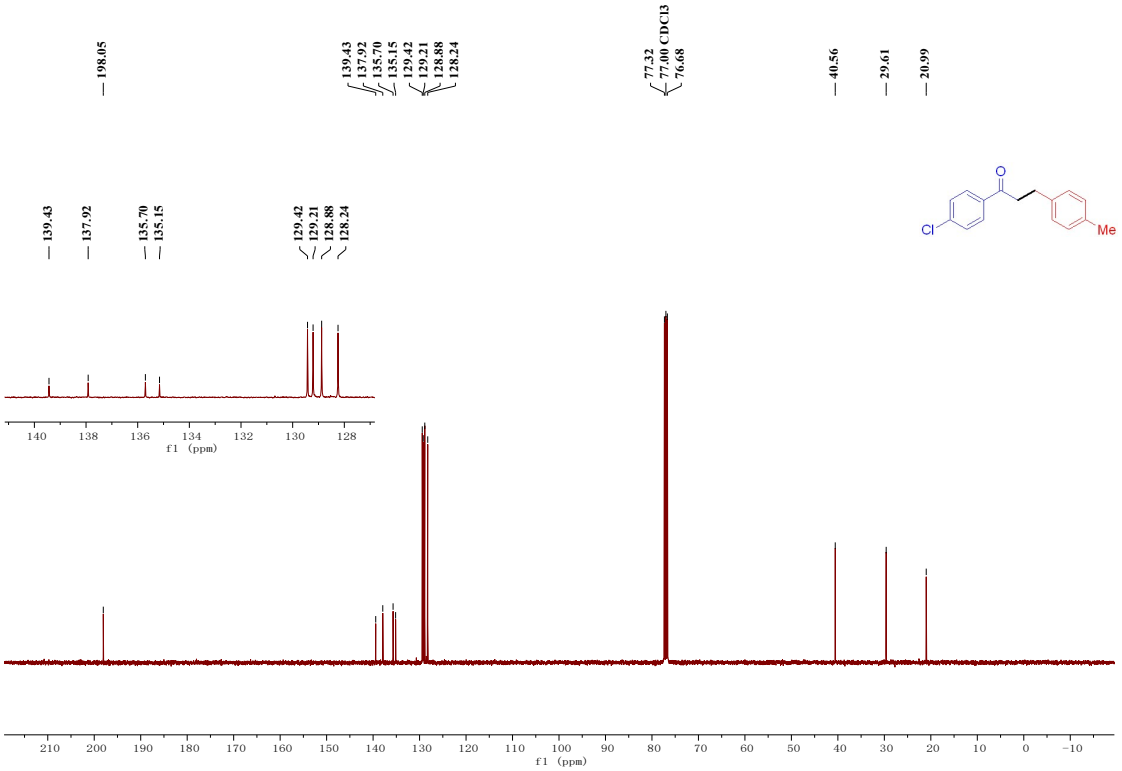


Figure S61. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 29 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **30**

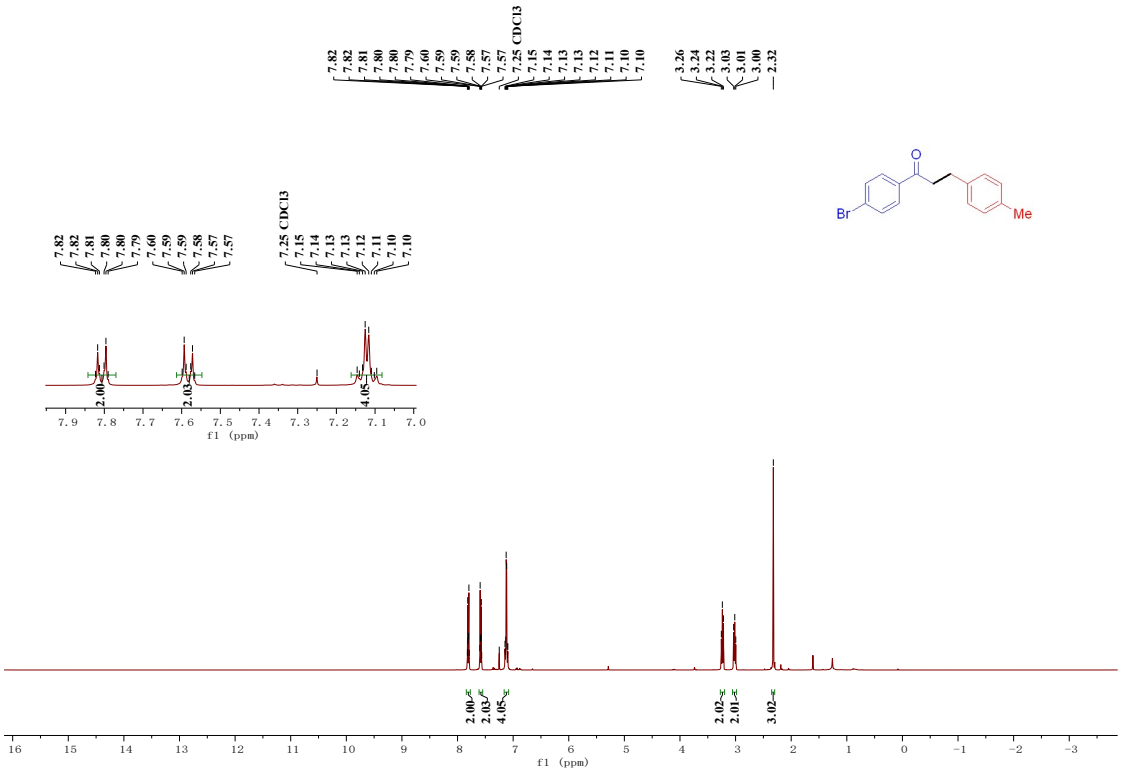


Figure S62. ^{1}H NMR (400 MHz) spectrum of compound 30 in $CDCl_3$

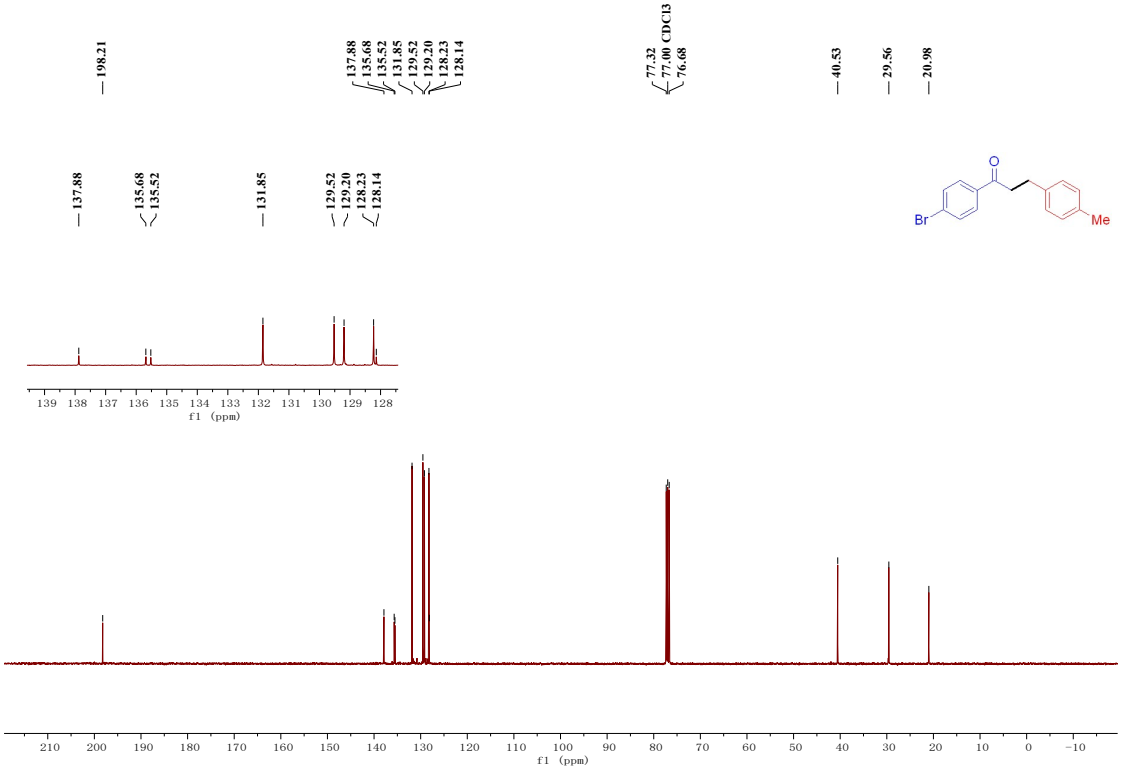


Figure S63. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 30 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **31**

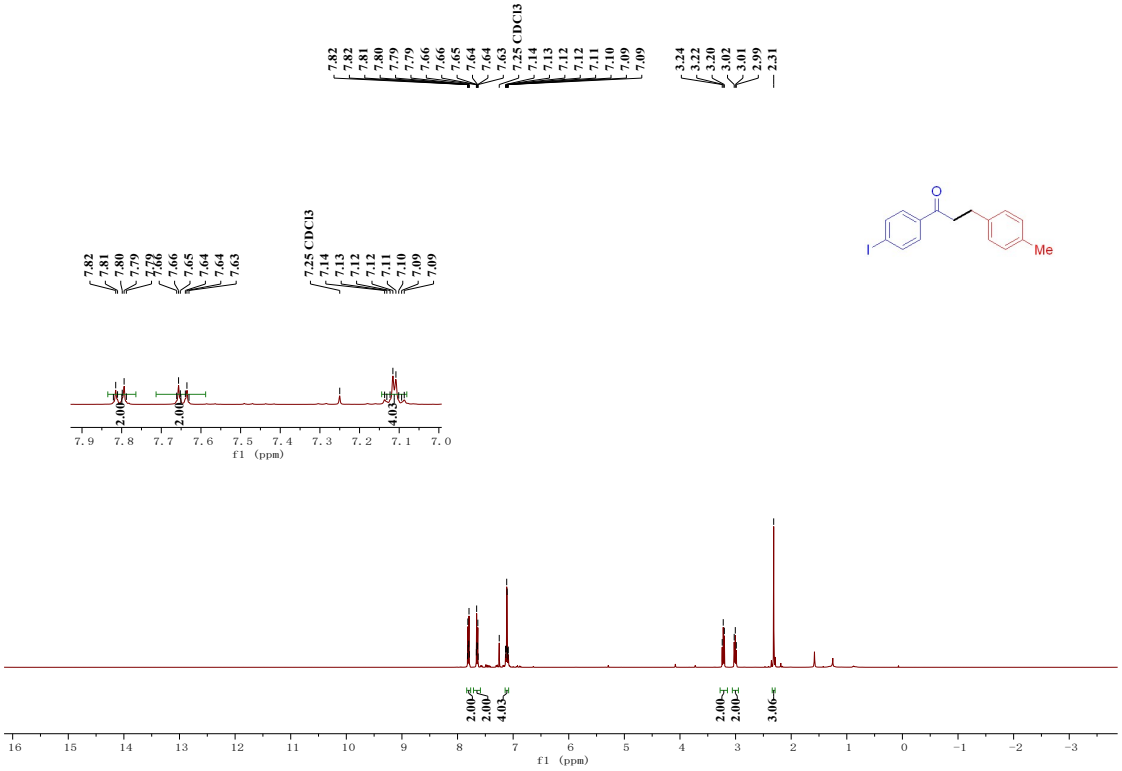


Figure S64. ^{1}H NMR (400 MHz) spectrum of compound 31 in $CDCl_3$

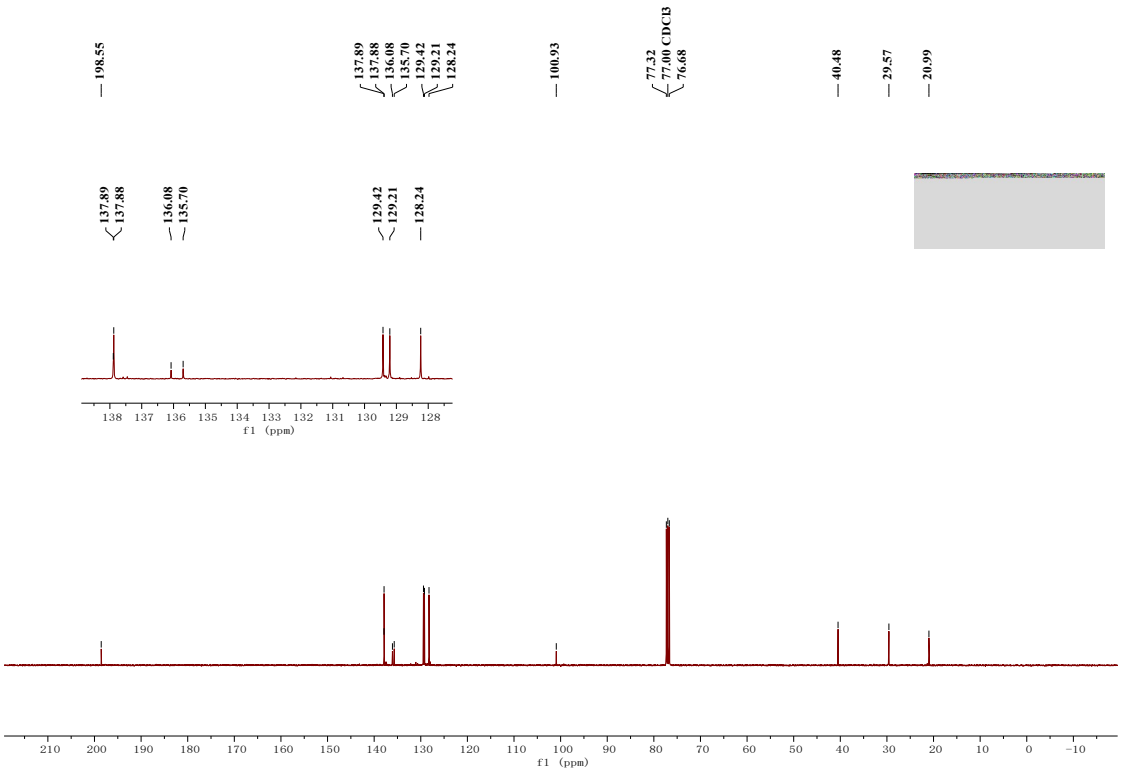


Figure S65. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 31 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **32**

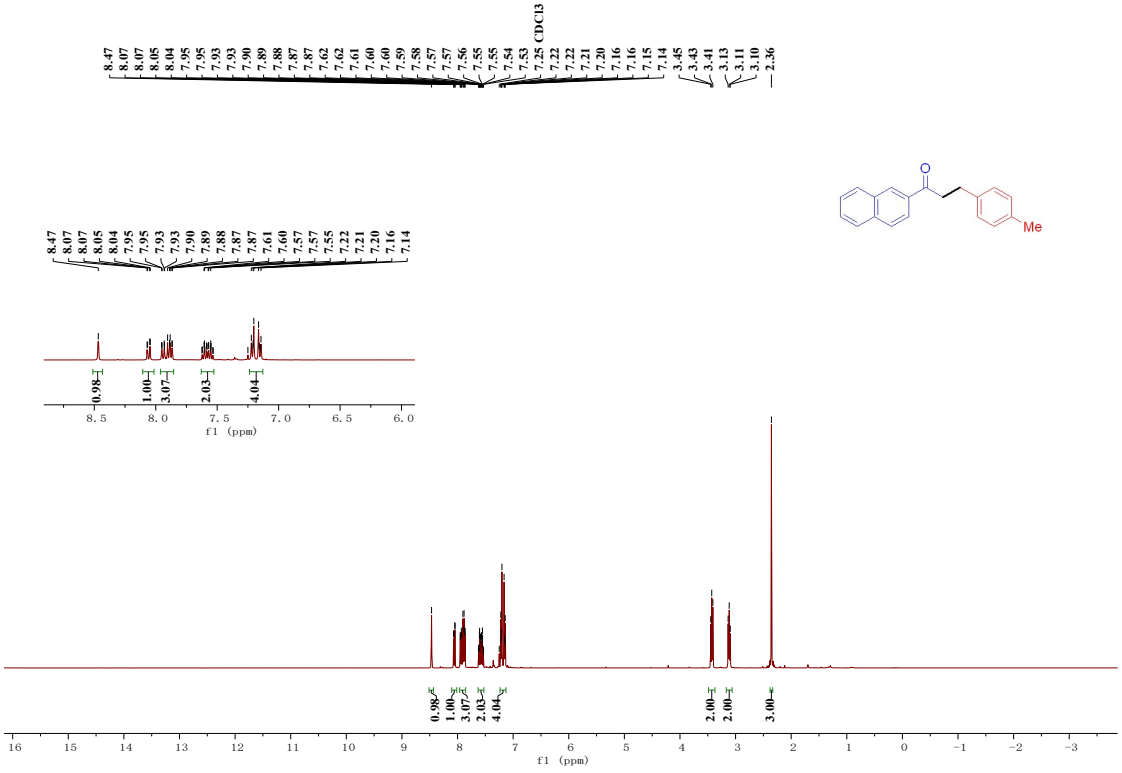


Figure S66. ^{1}H NMR (400 MHz) spectrum of compound 32 in $CDCl_3$

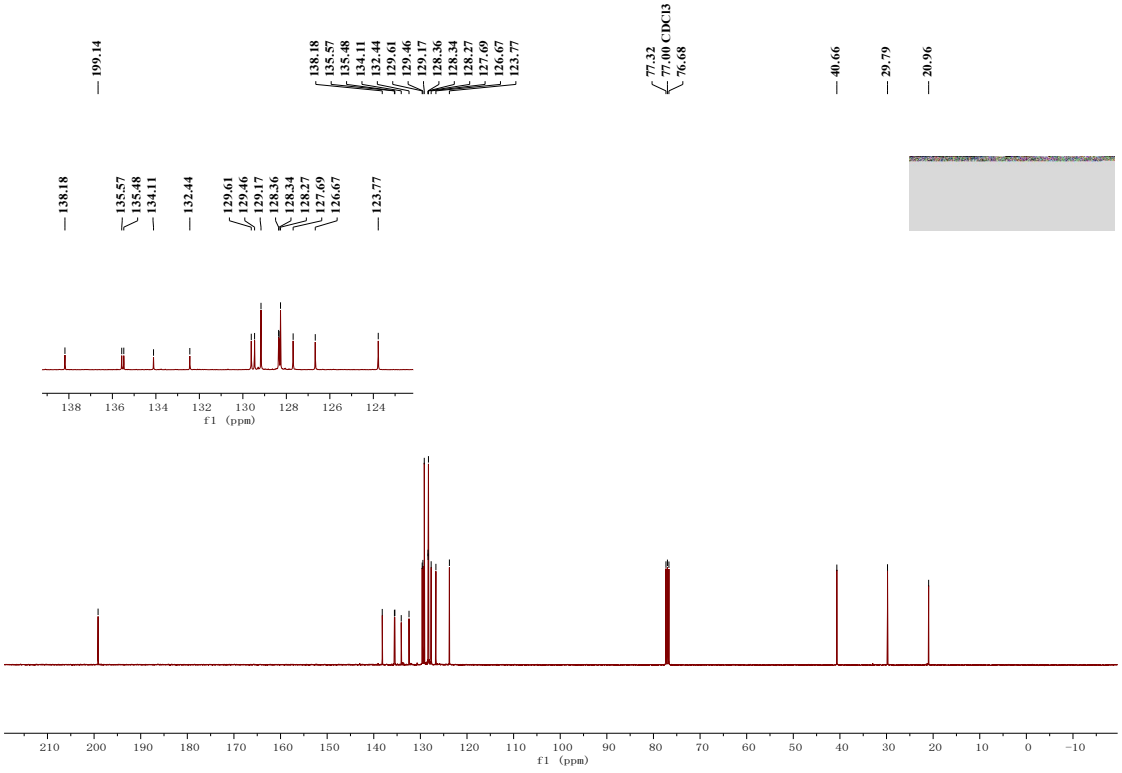


Figure S67. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 32 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **33**

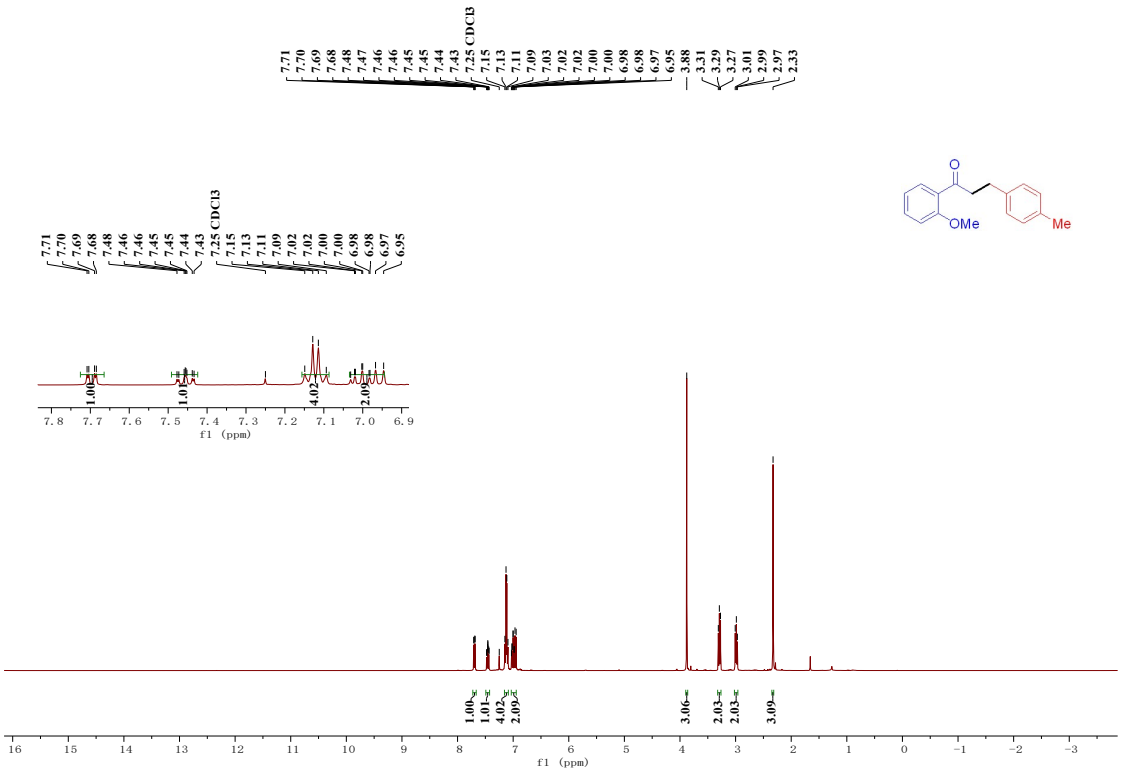


Figure S68. ^{1}H NMR (400 MHz) spectrum of compound 33 in $CDCl_3$

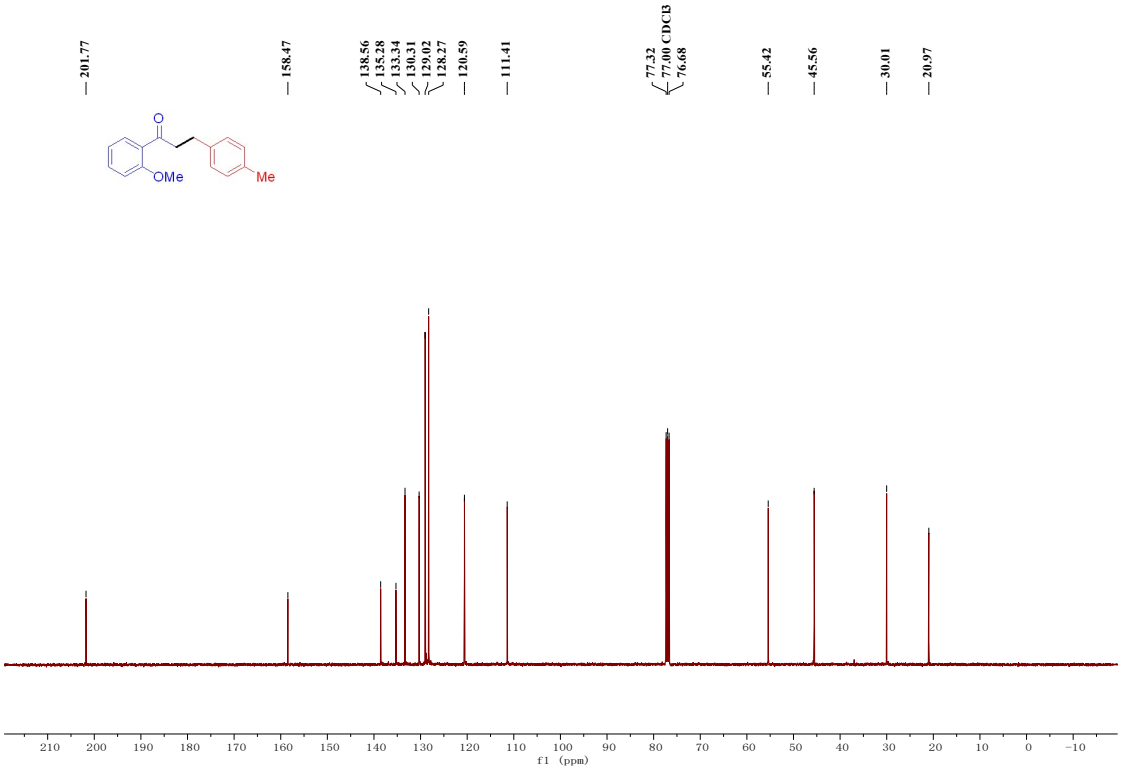


Figure S69. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 33 in $CDCl_3$

^{1}H and ^{13}C{^{1}H} NMR spectra of compound **34**

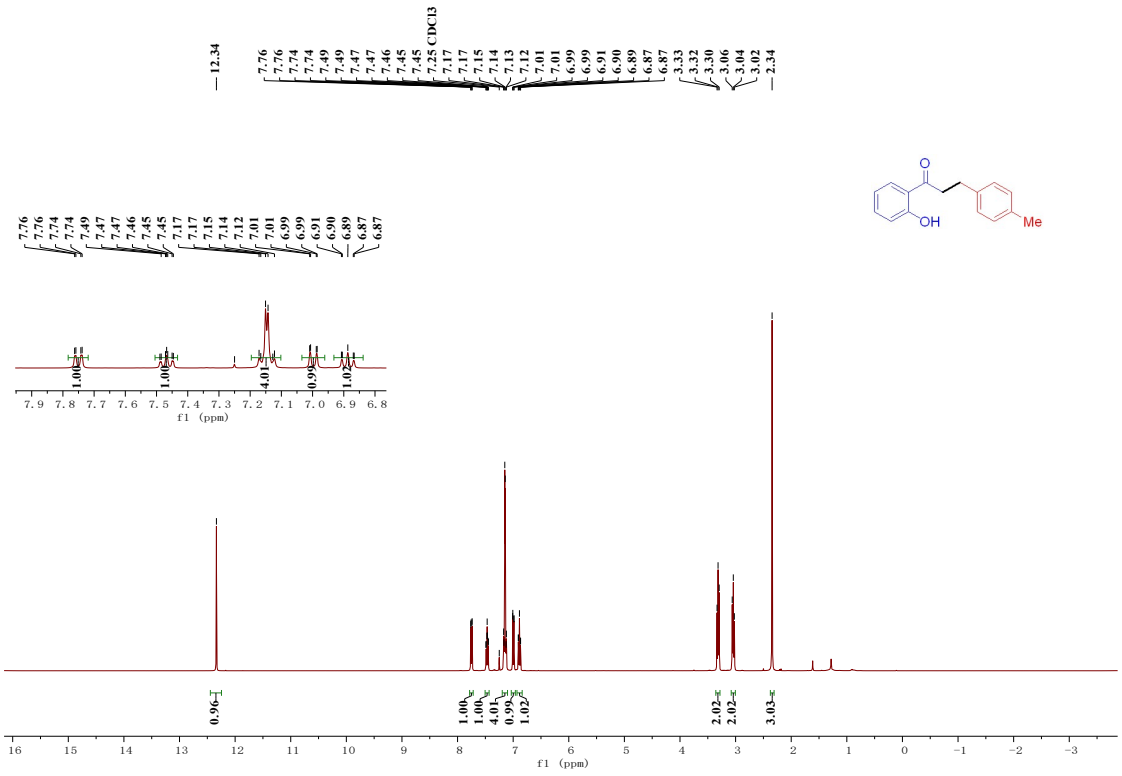


Figure S70. ^{1}H NMR (400 MHz) spectrum of compound 34 in $CDCl_3$

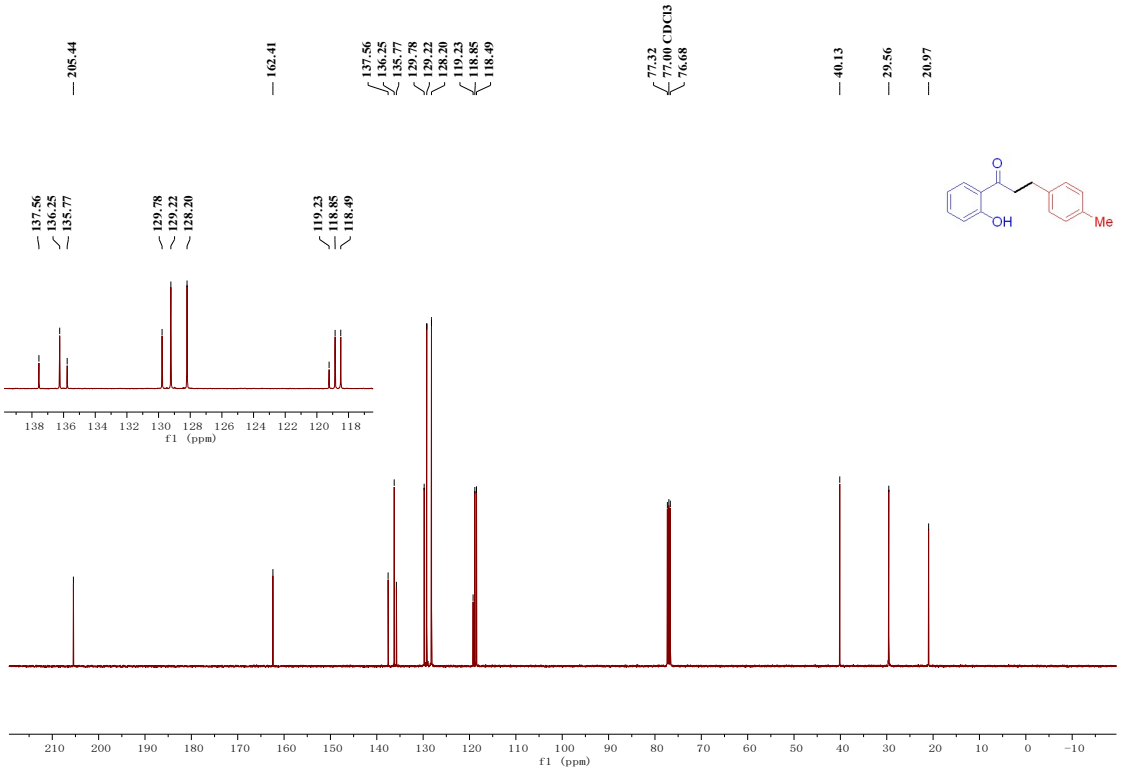


Figure S71. ^{13}C{^{1}H} NMR (101 MHz) spectrum of compound 34 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **35**

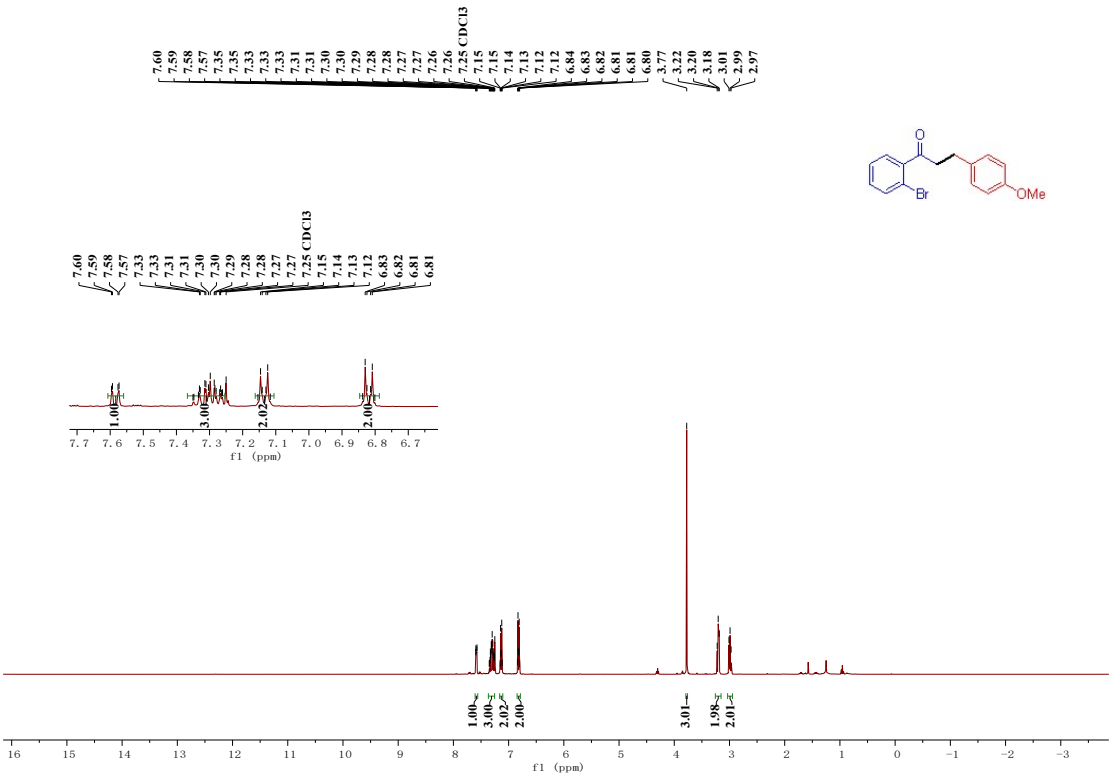


Figure S72. ^{1}H NMR (400 MHz) spectrum of compound 35 in $CDCl_3$

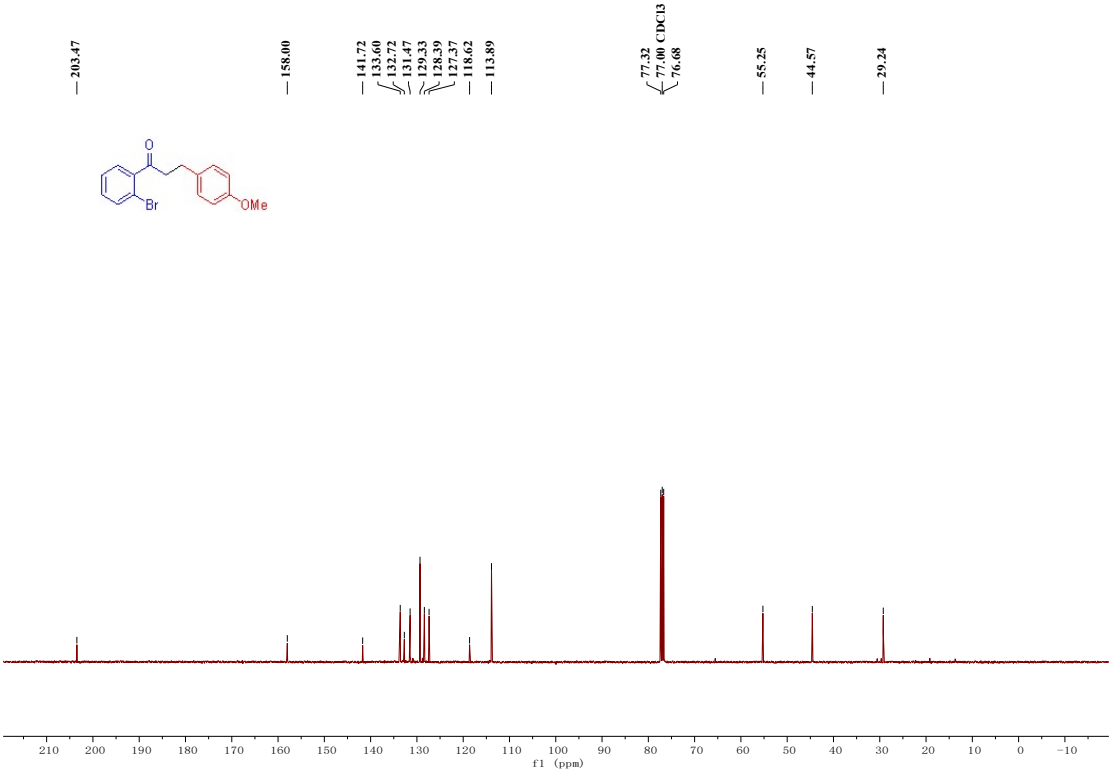


Figure S73. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 35 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **36**

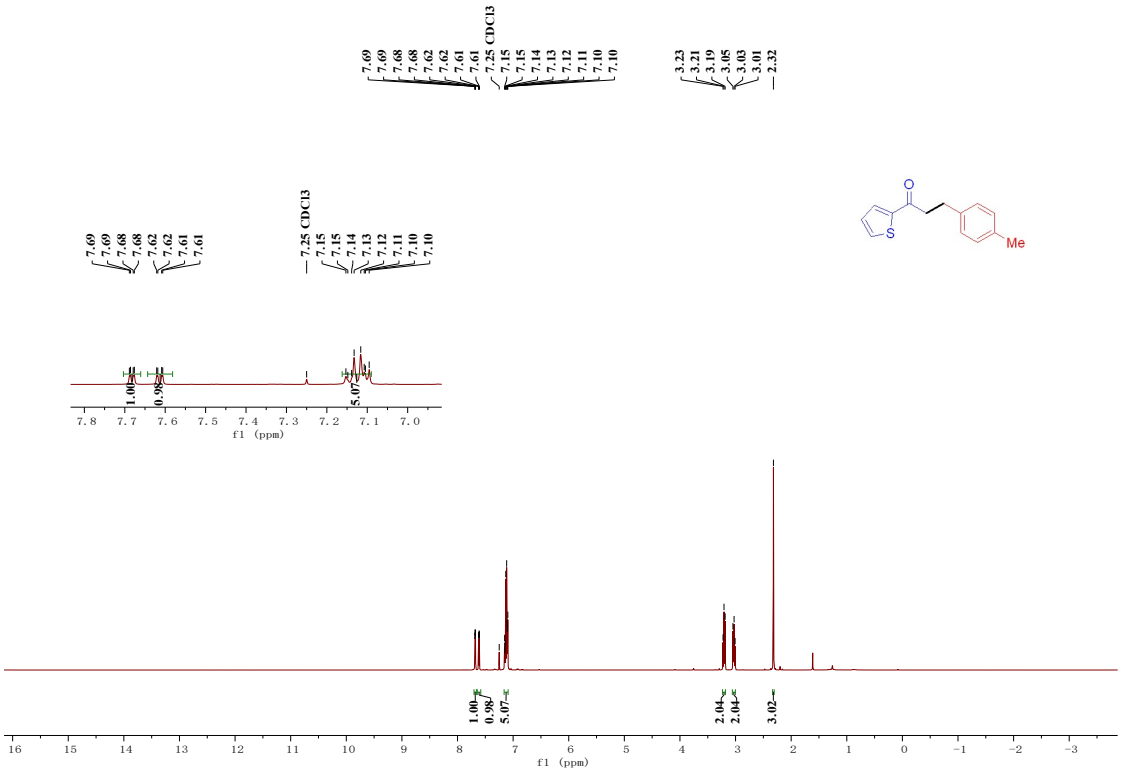


Figure S74. ^{1}H NMR (400 MHz) spectrum of compound 36 in $CDCl_3$

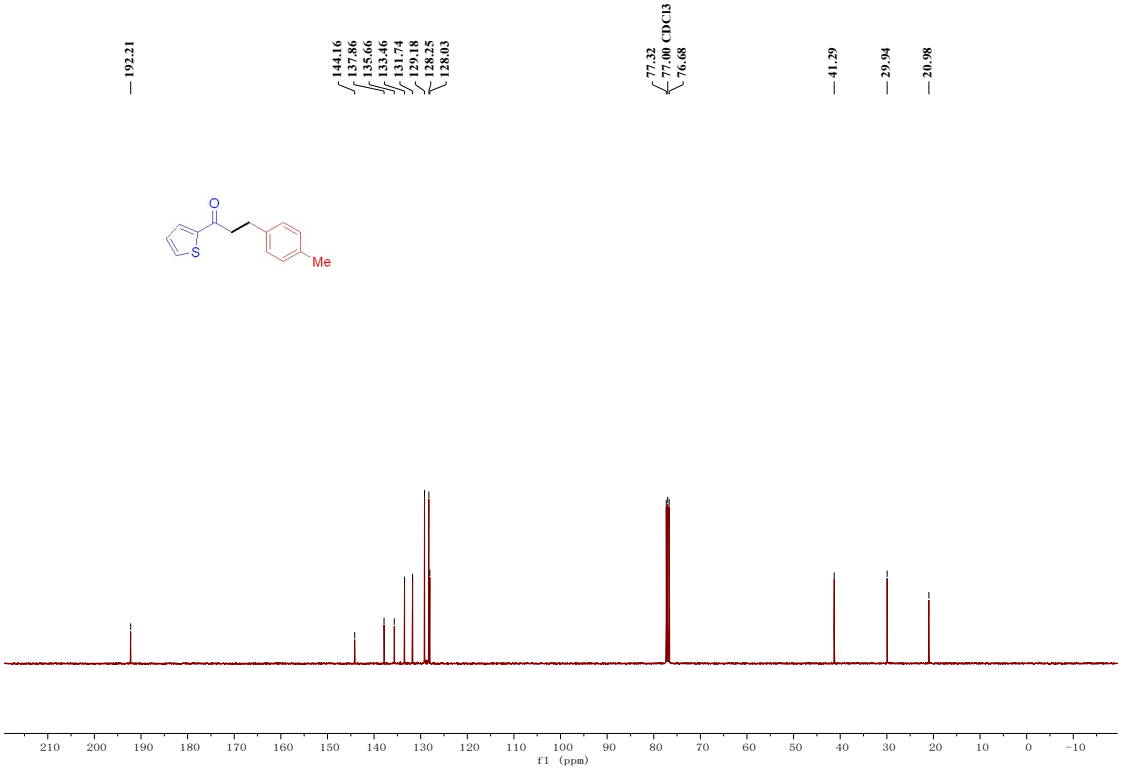


Figure S75. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 36 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **37**

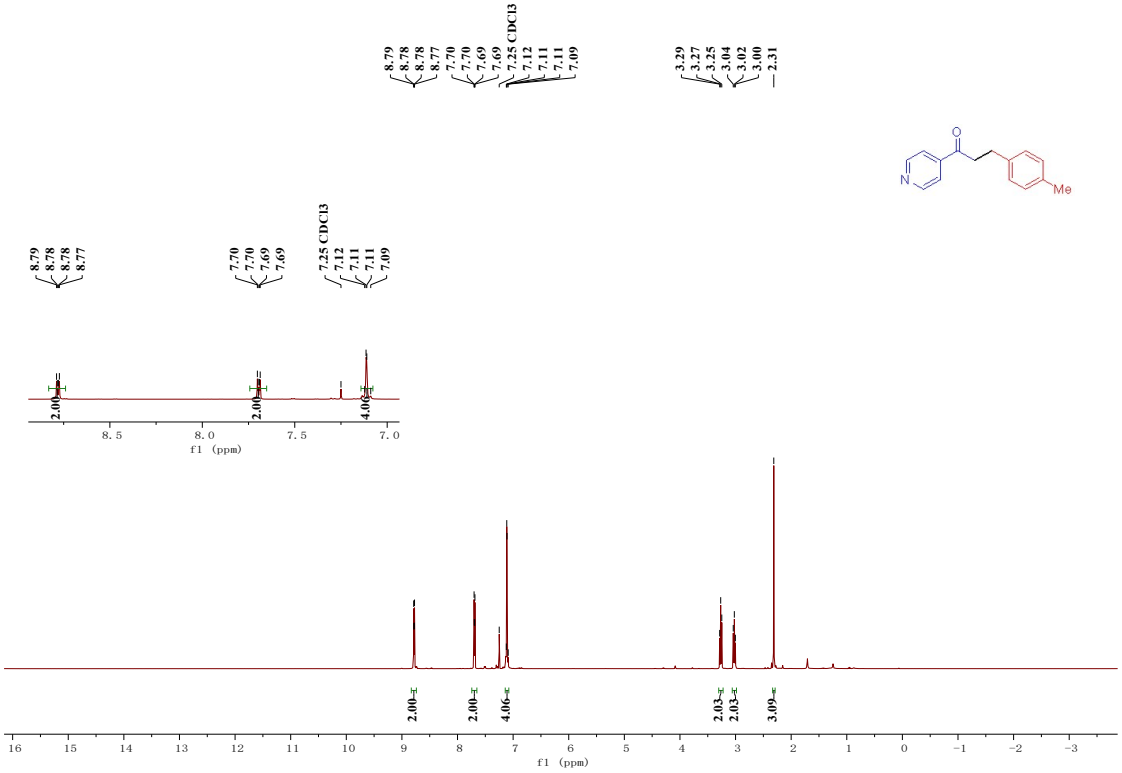


Figure S76. ^{1}H NMR (400 MHz) spectrum of compound 37 in $CDCl_3$

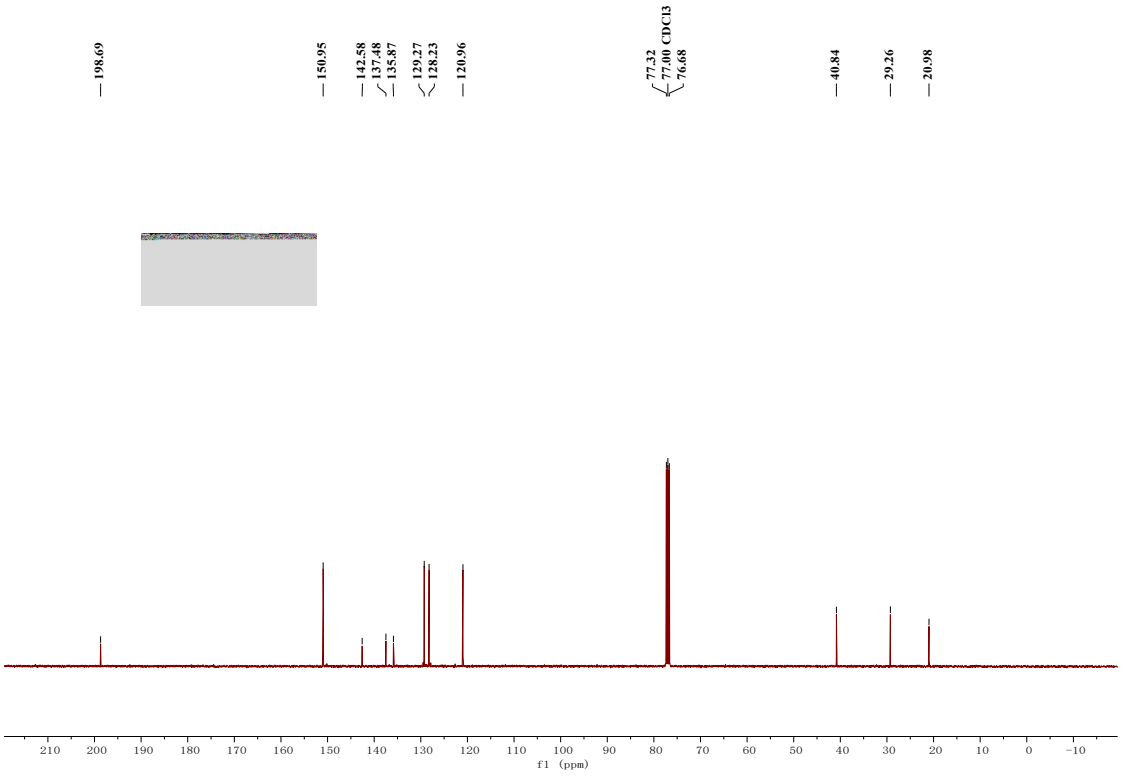


Figure S77. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 37 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **38**

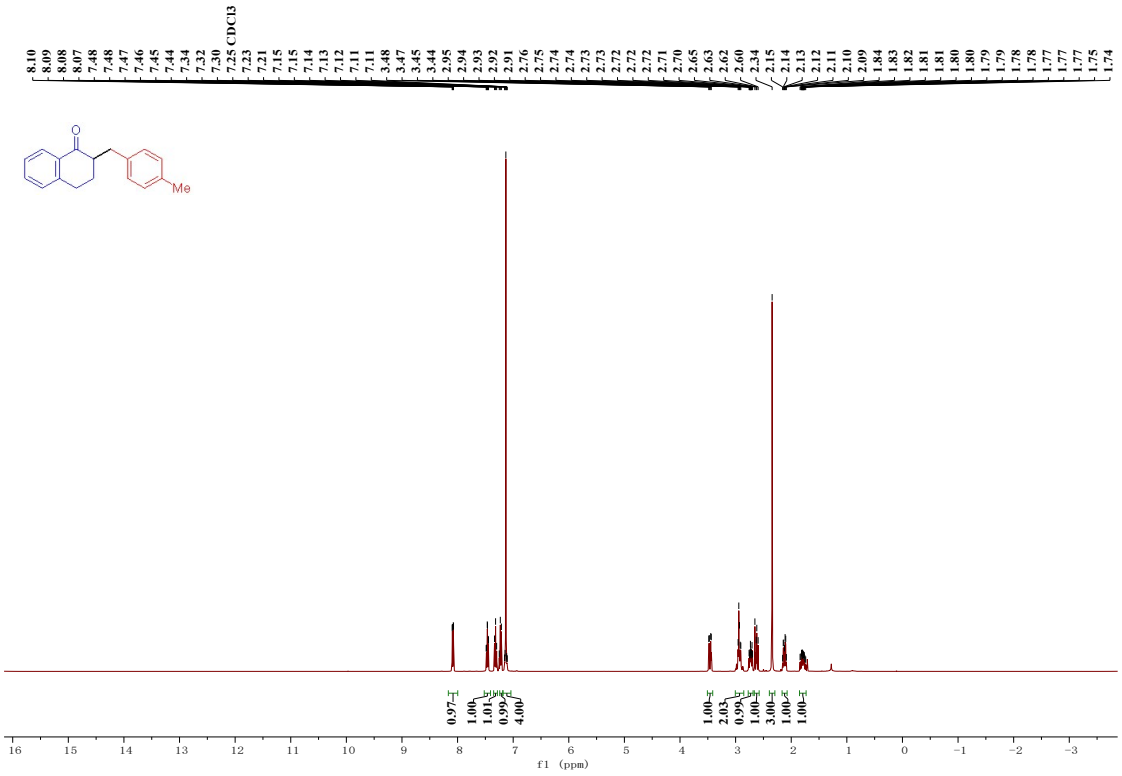


Figure S78. ^{1}H NMR (400 MHz) spectrum of compound 38 in $CDCl_3$

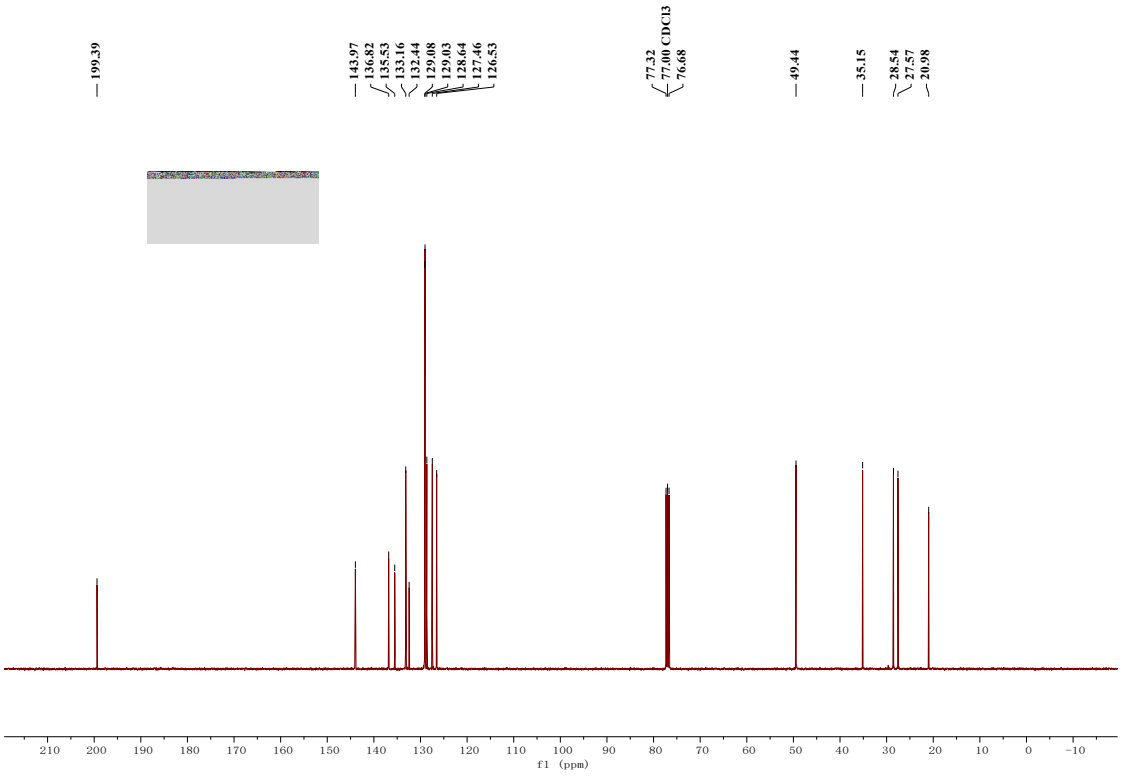


Figure S79. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 38 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **39**

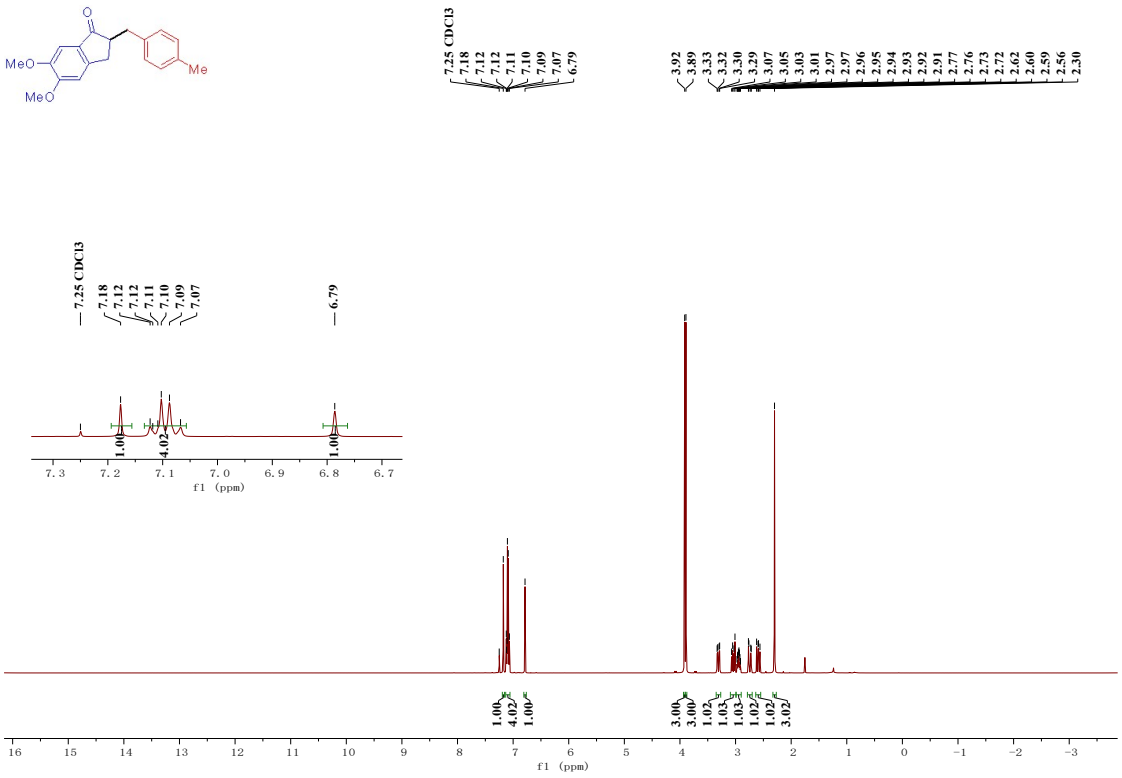


Figure S80. ^{1}H NMR (400 MHz) spectrum of compound 39 in $CDCl_3$

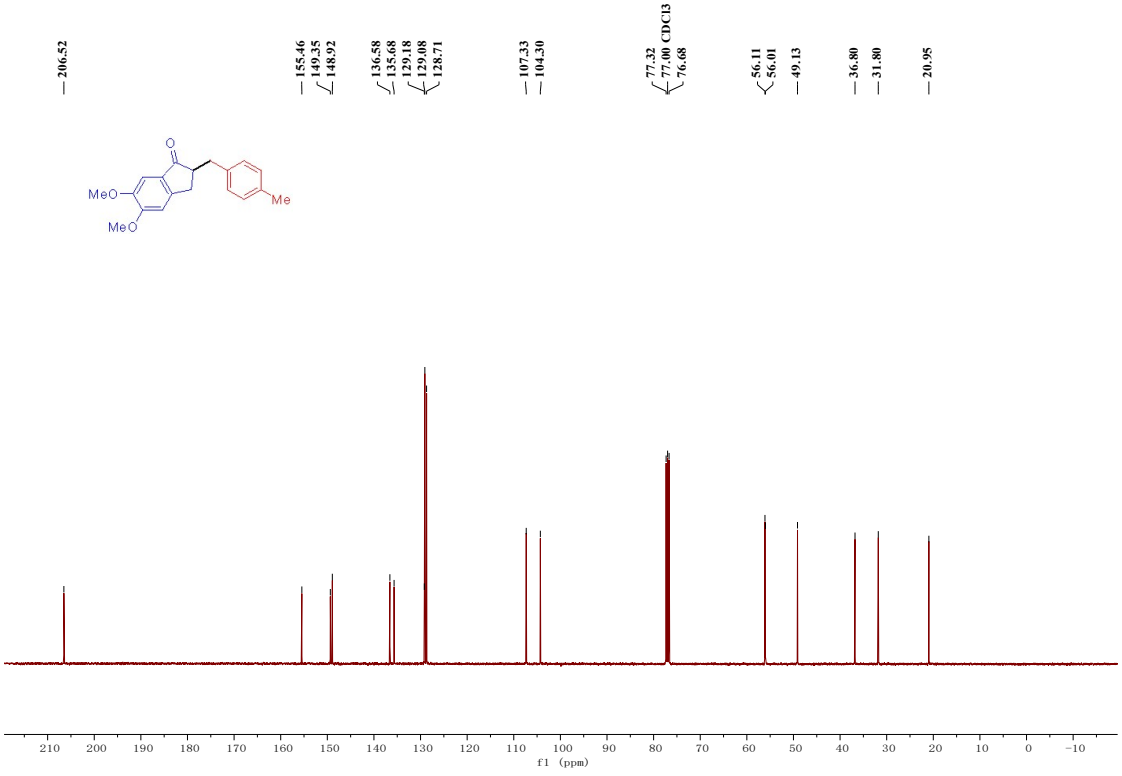


Figure S81. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 39 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **40**

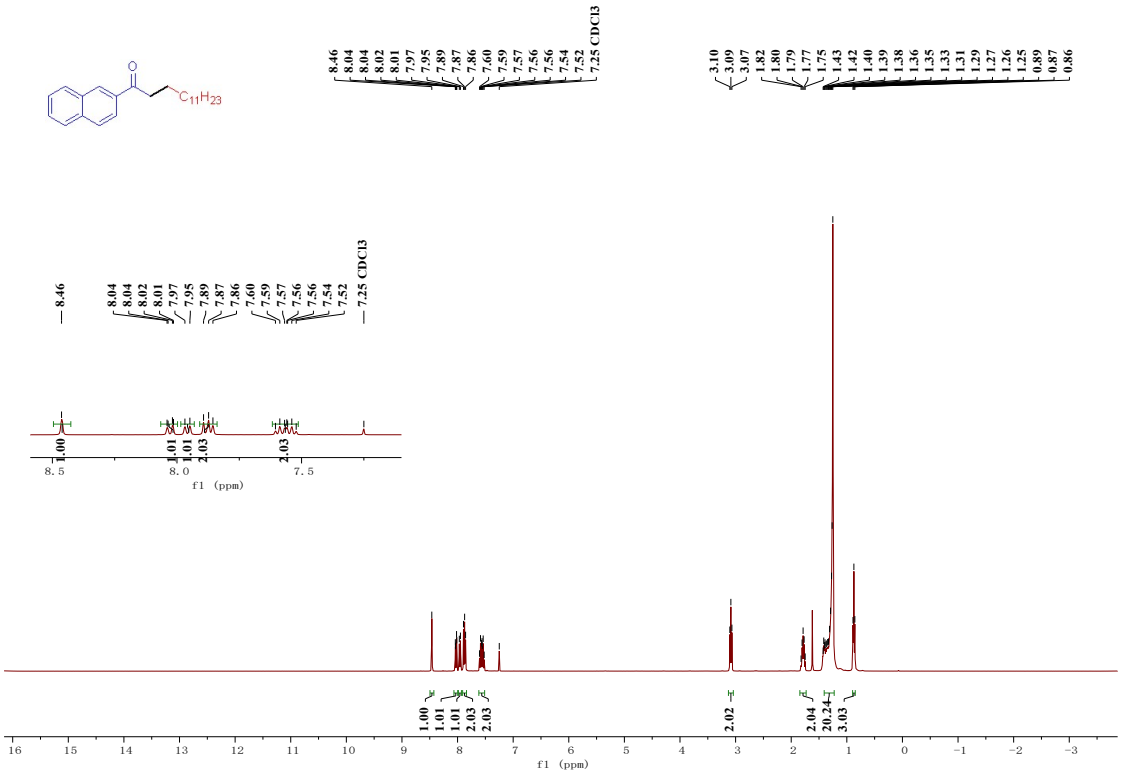


Figure S82. ^{1}H NMR (400 MHz) spectrum of compound 40 in $CDCl_3$

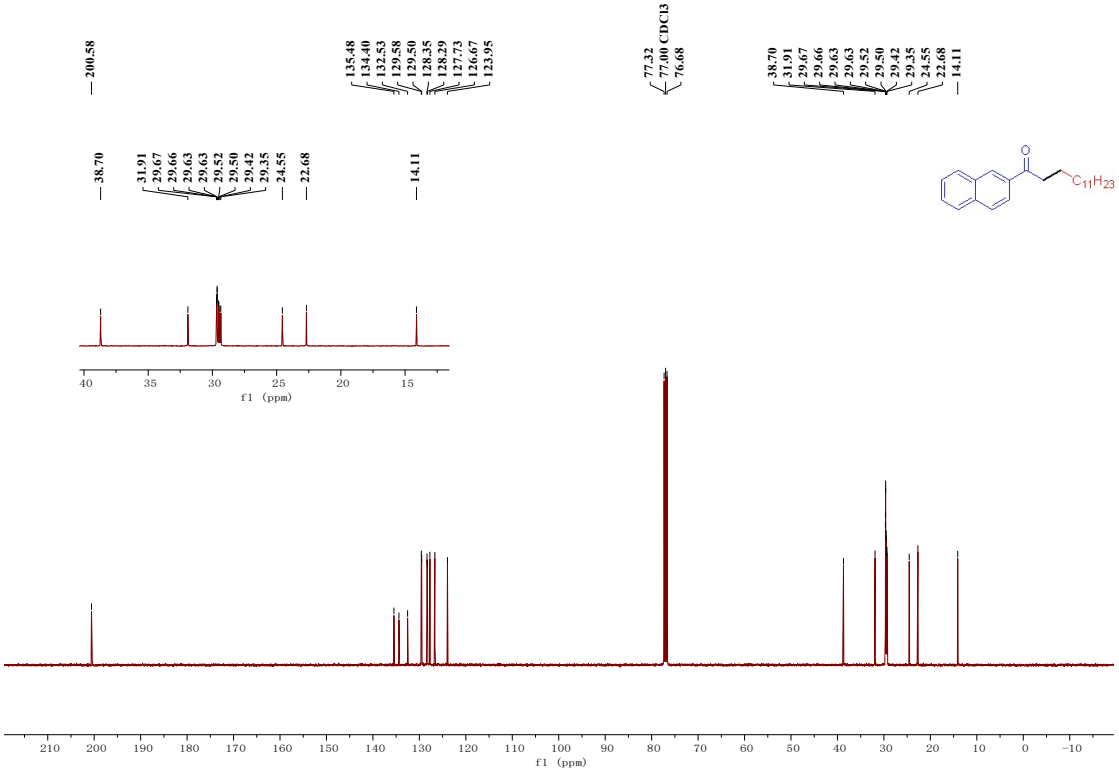


Figure S83. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 40 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **41**

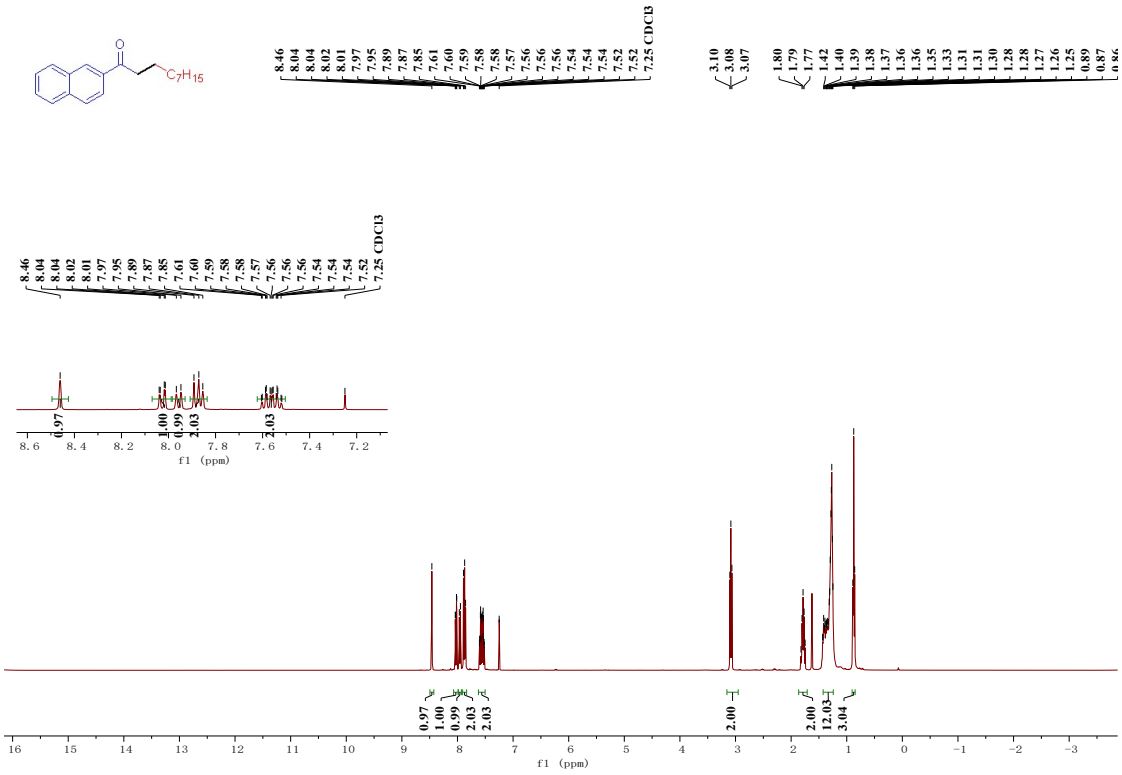


Figure S84. ^{1}H NMR (400 MHz) spectrum of compound 41 in $CDCl_3$

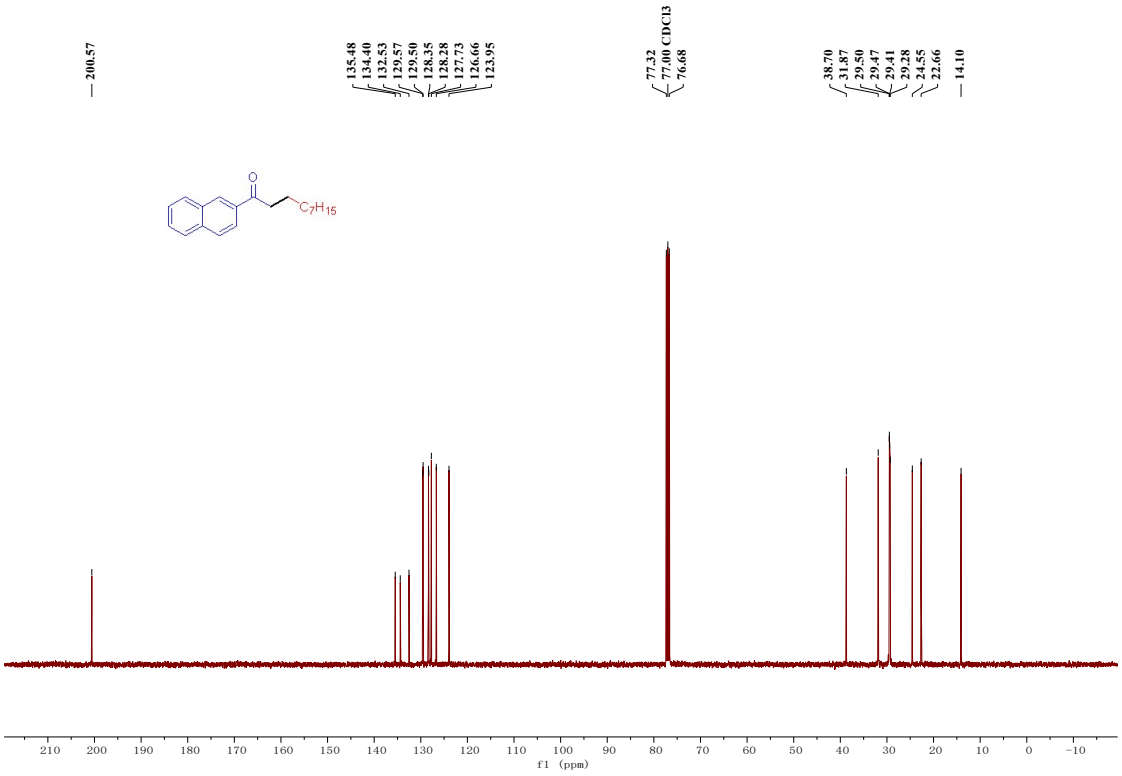


Figure S85. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 41 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **42**

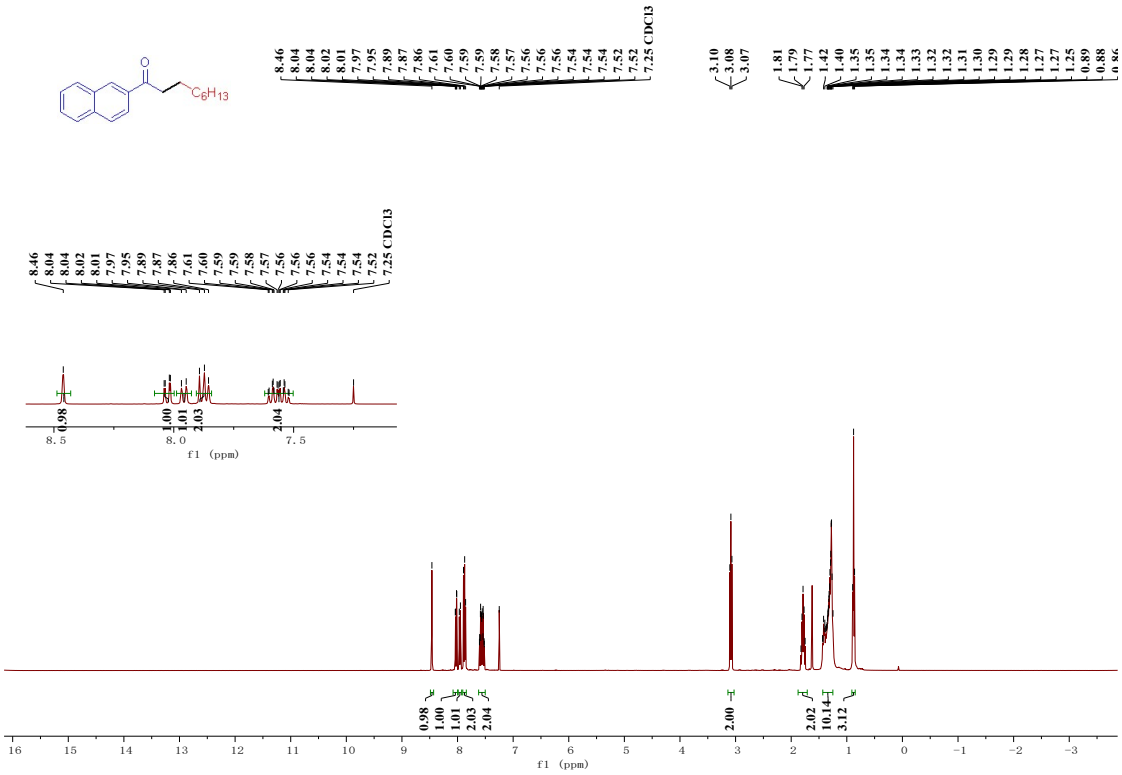


Figure S86. ^{1}H NMR (400 MHz) spectrum of compound 42 in $CDCl_3$

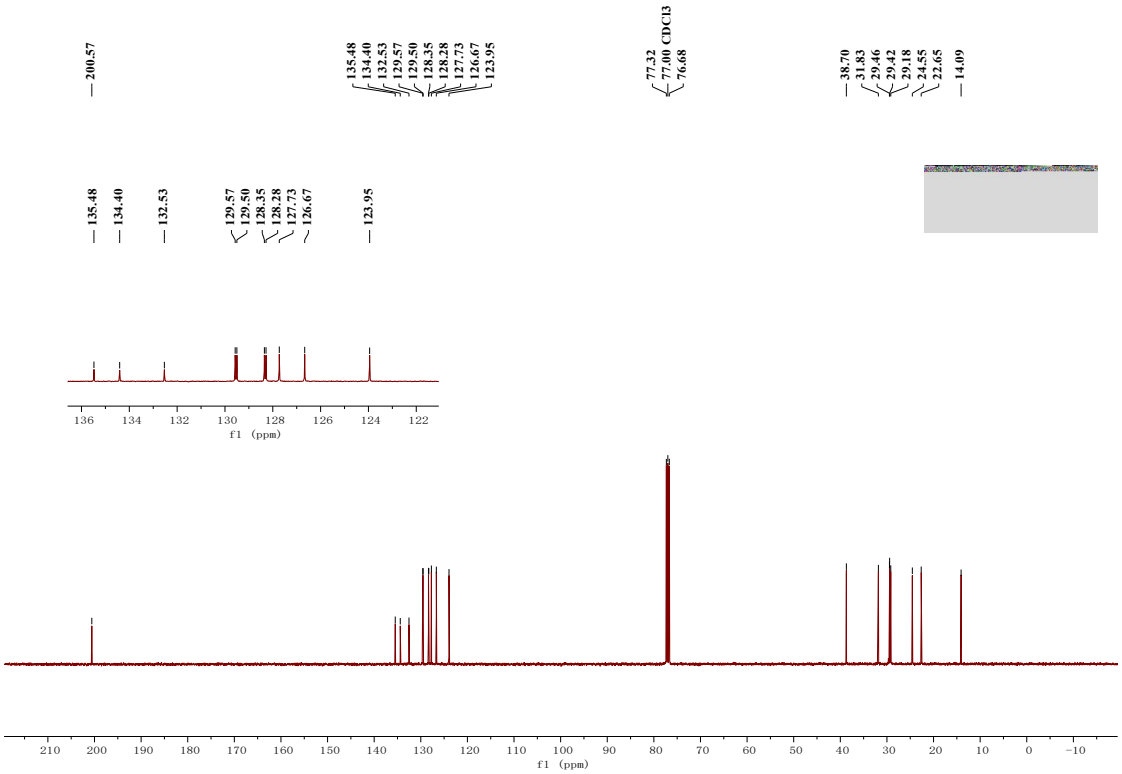


Figure S87. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 42 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **43**

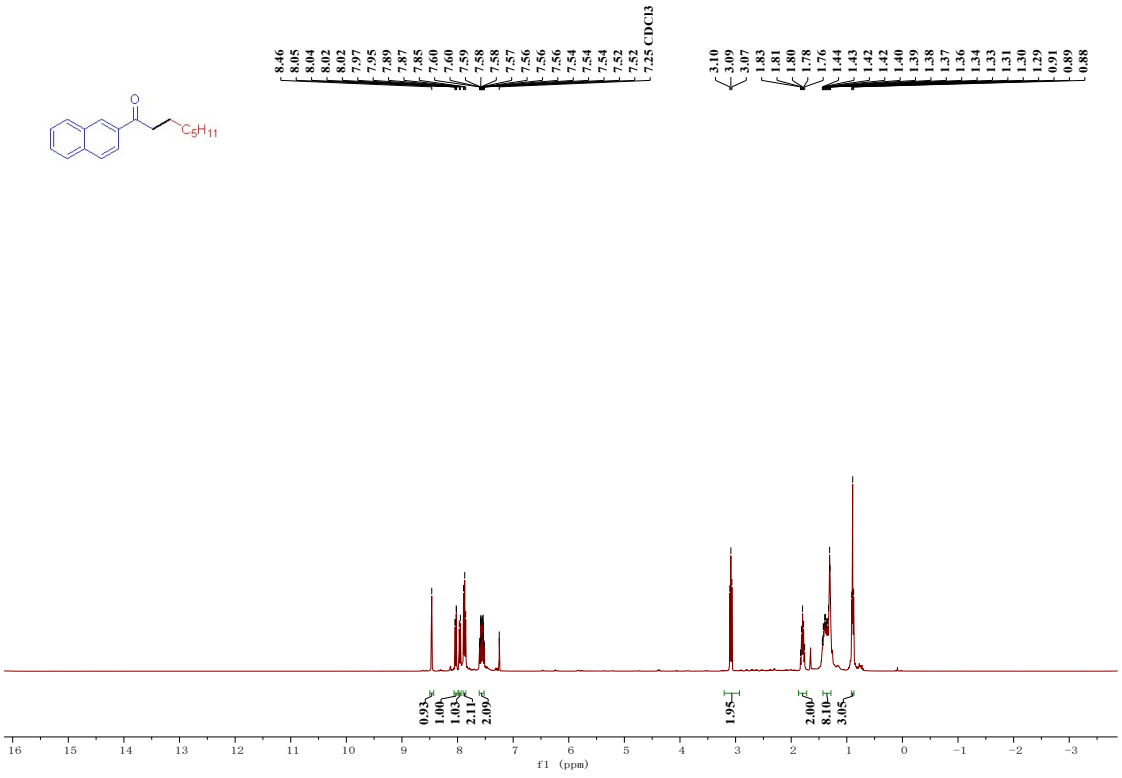


Figure S88. ^{1}H NMR (400 MHz) spectrum of compound 43 in $CDCl_3$

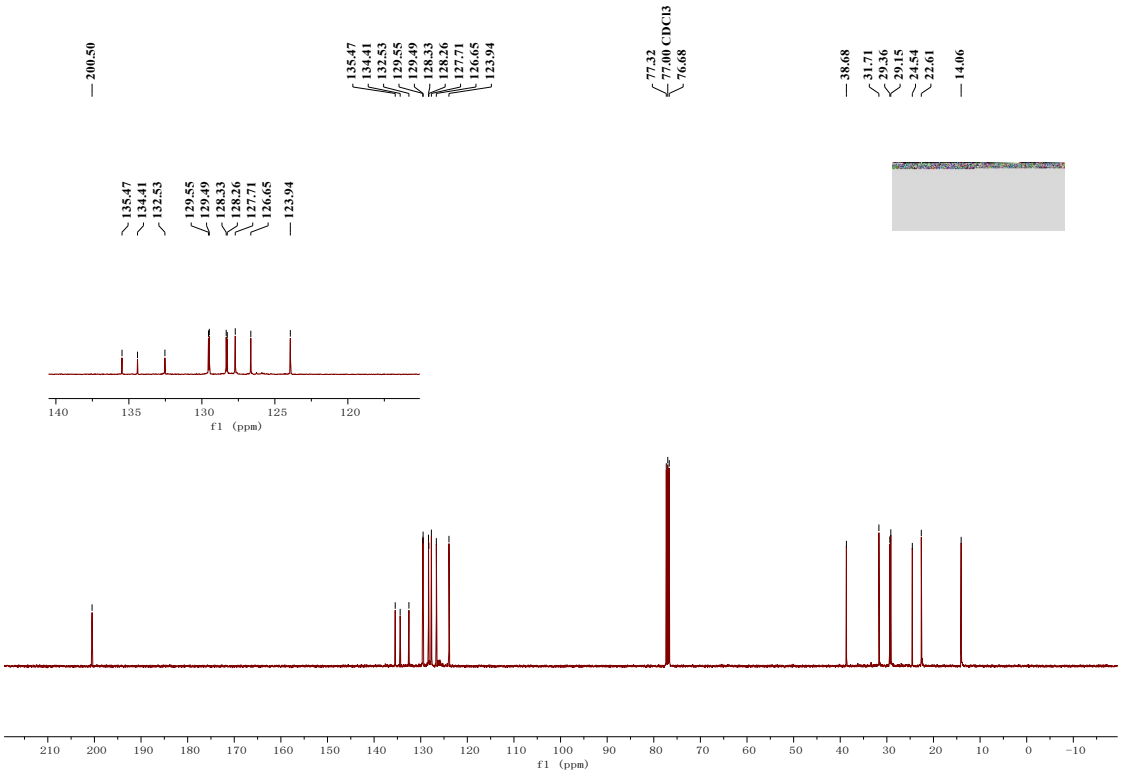


Figure S89. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 43 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **44**

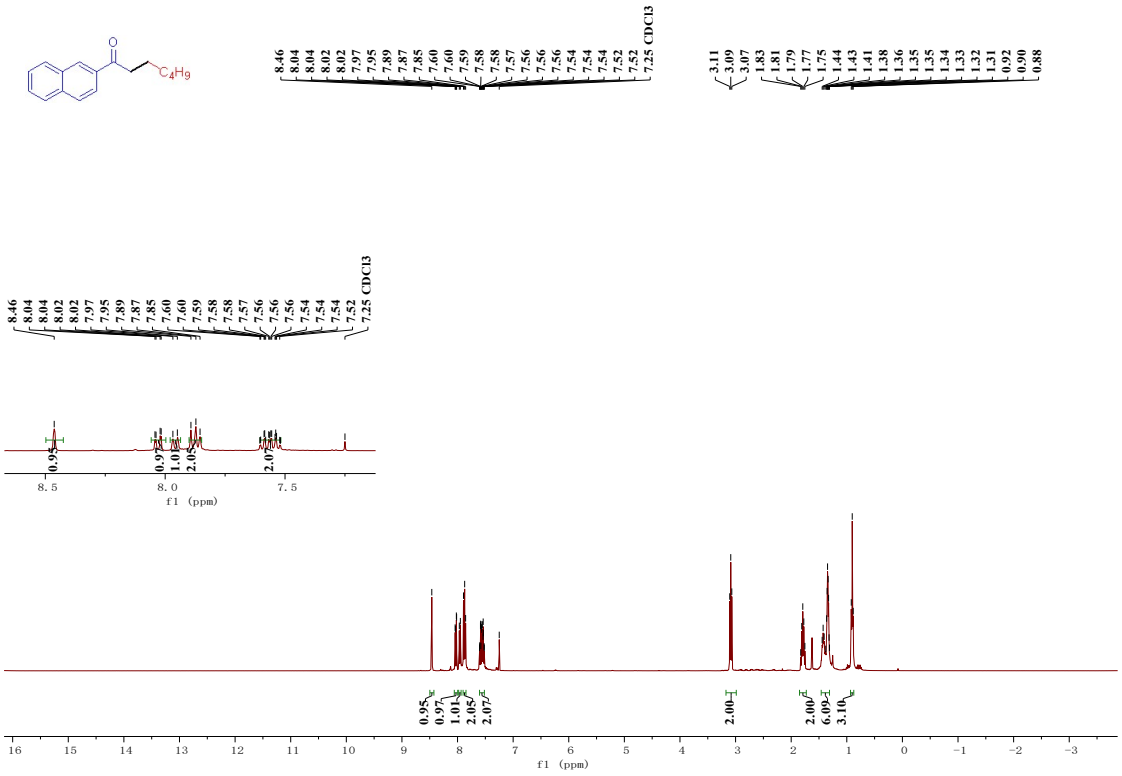


Figure S90. ^{1}H NMR (400 MHz) spectrum of compound 44 in $CDCl_3$

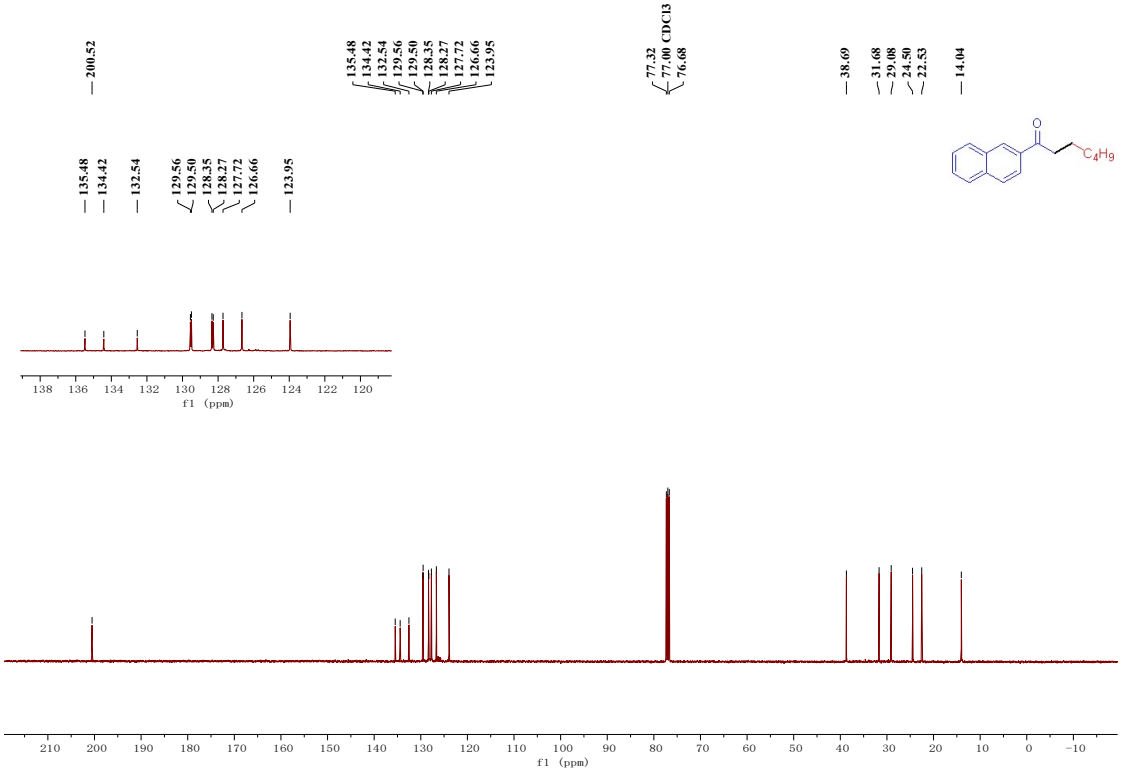


Figure S91. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 44 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **45**

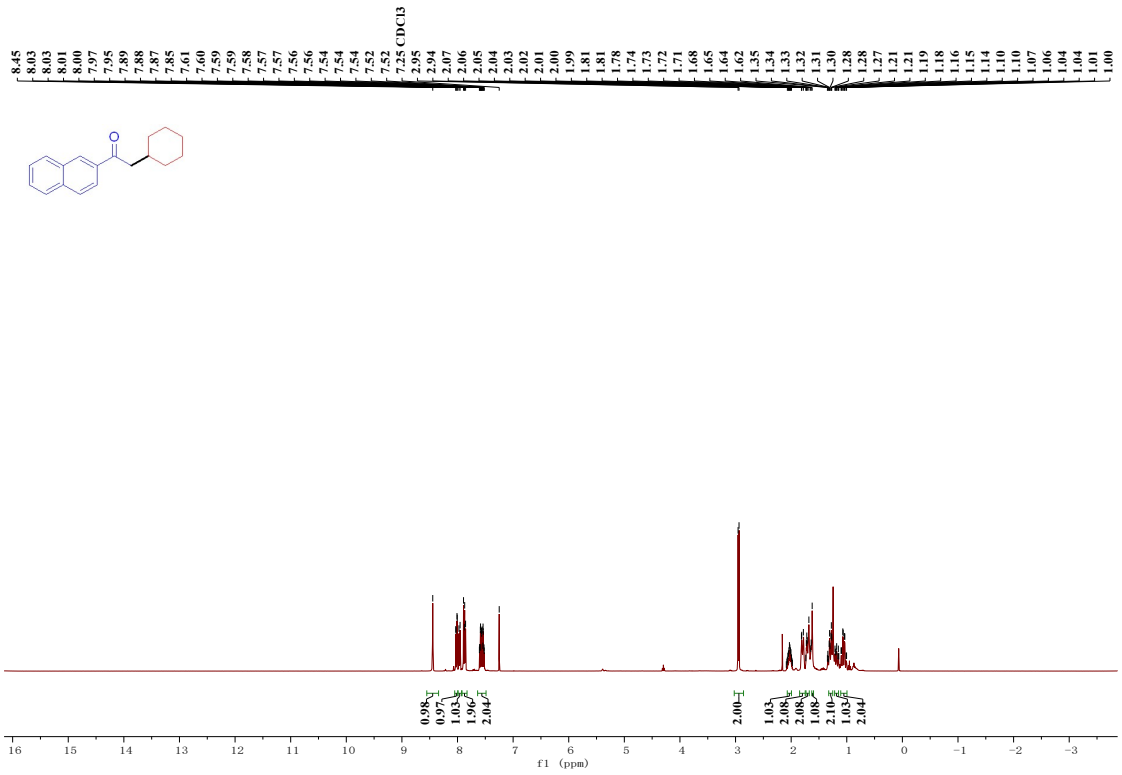


Figure S92. ^{1}H NMR (400 MHz) spectrum of compound 45 in $CDCl_3$

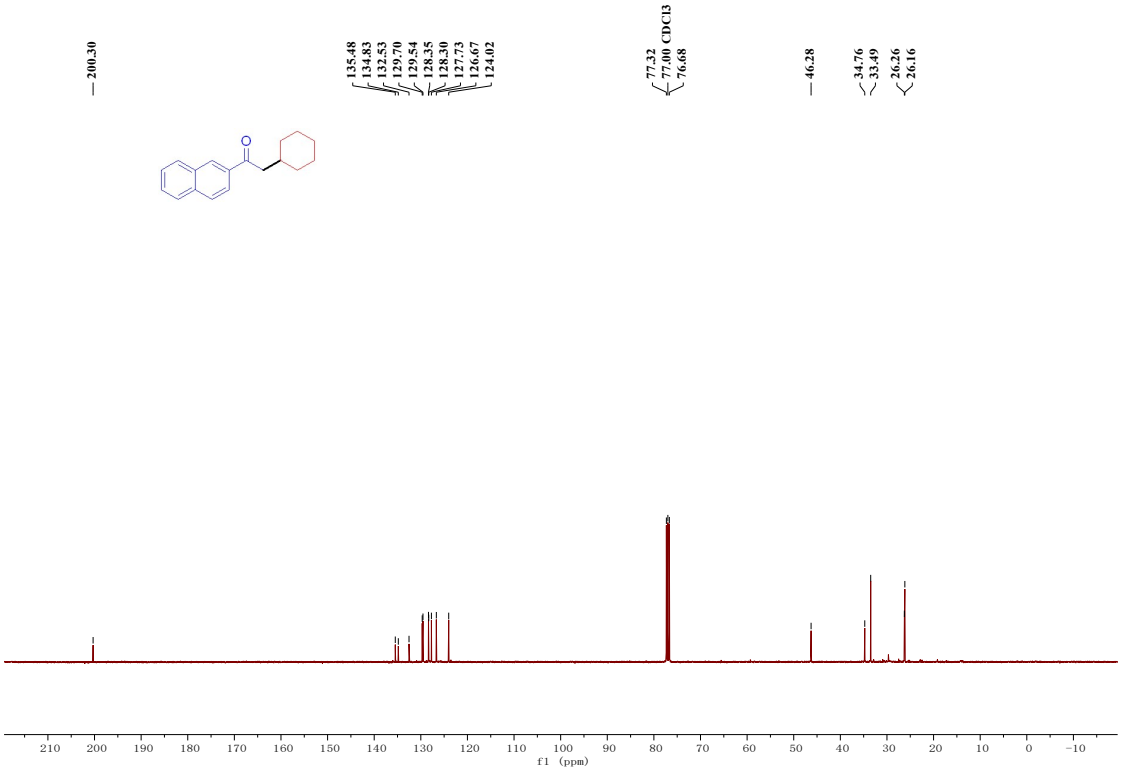


Figure S93. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 45 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **46**

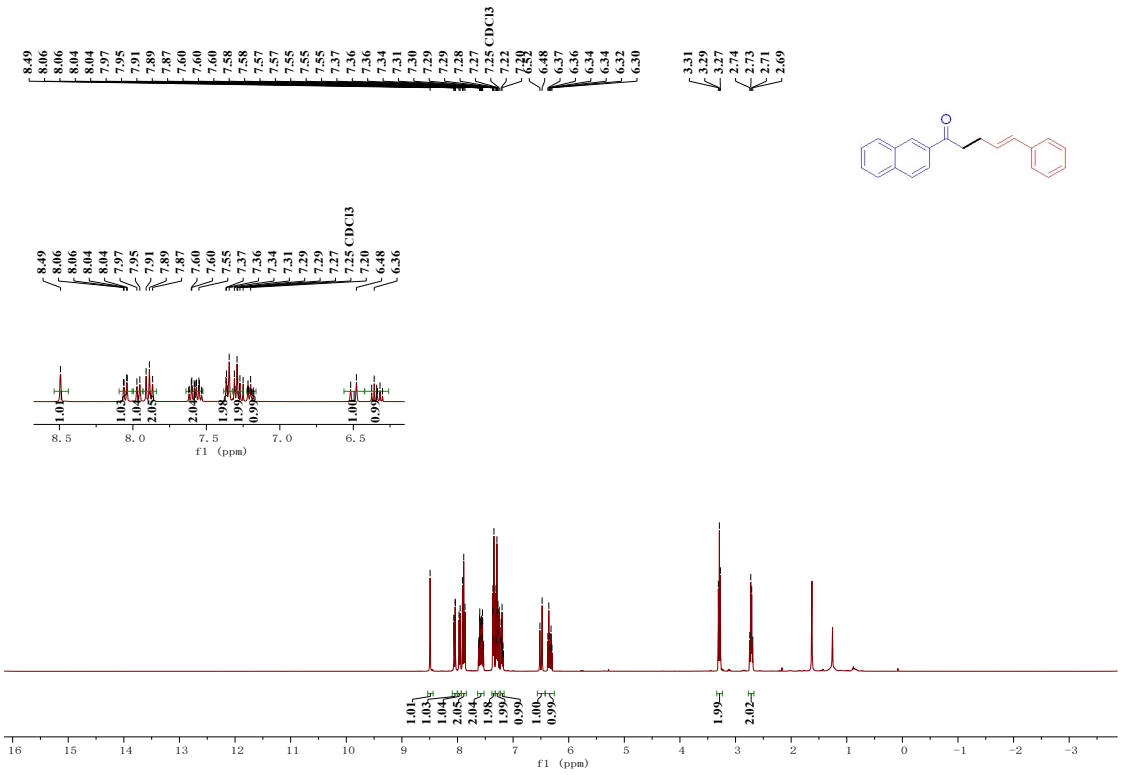


Figure S94. ^{1}H NMR (400 MHz) spectrum of compound 46 in $CDCl_3$

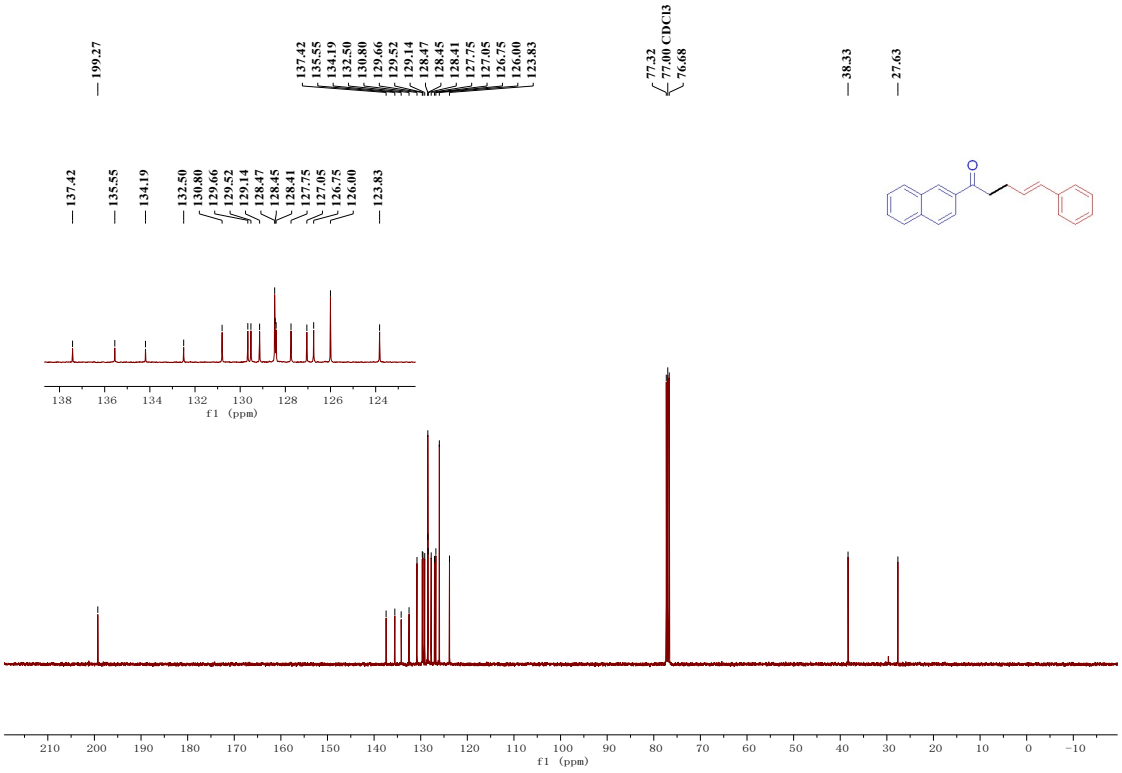


Figure S95. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 46 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **47**

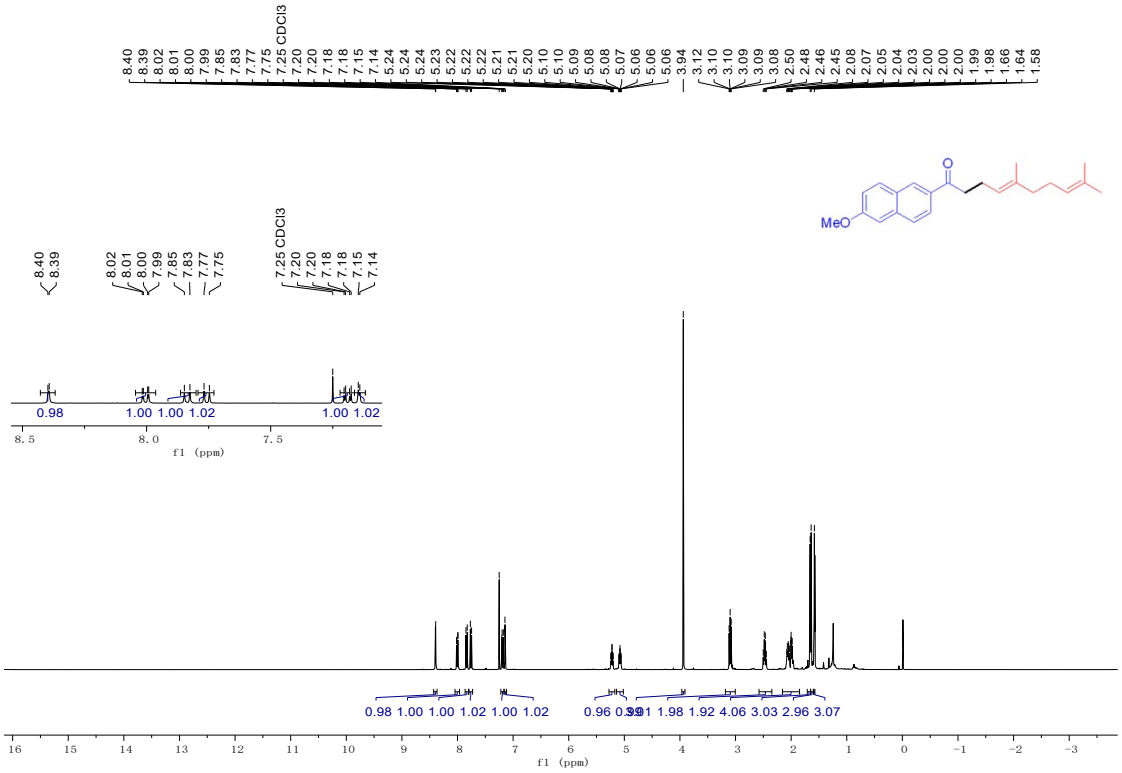


Figure S96. ^{1}H NMR (400 MHz) spectrum of compound 47 in $CDCl_3$

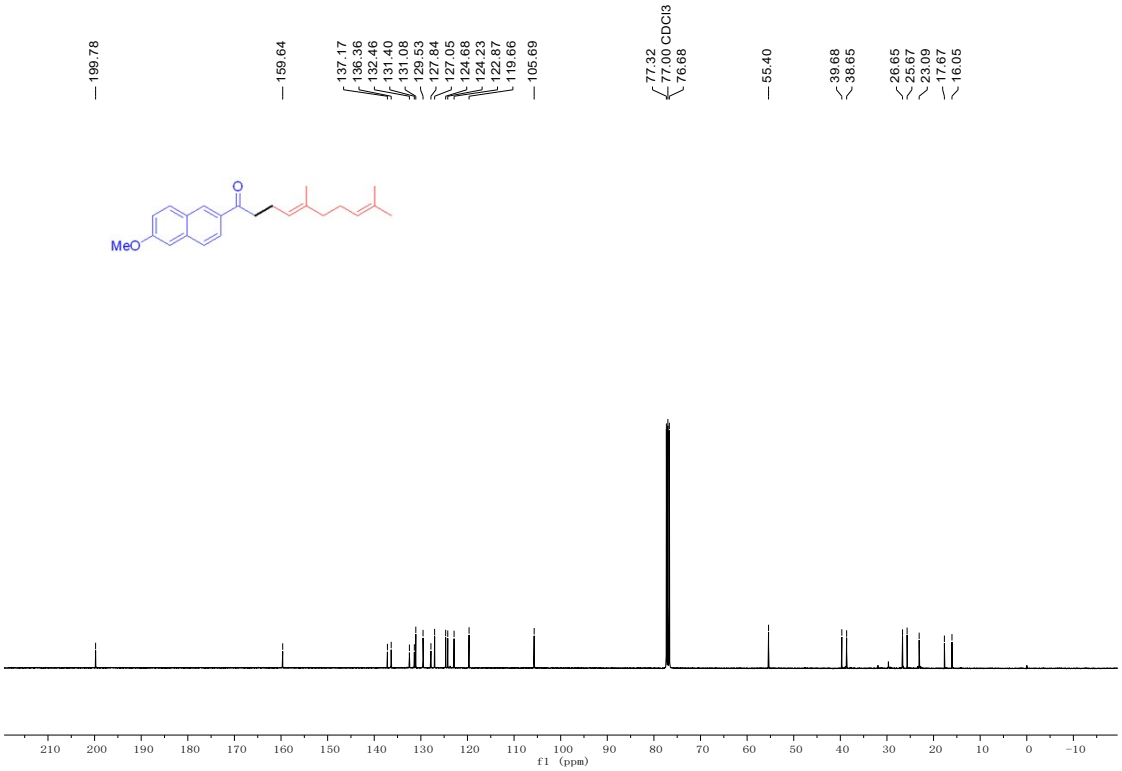


Figure S97. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 47 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **48**

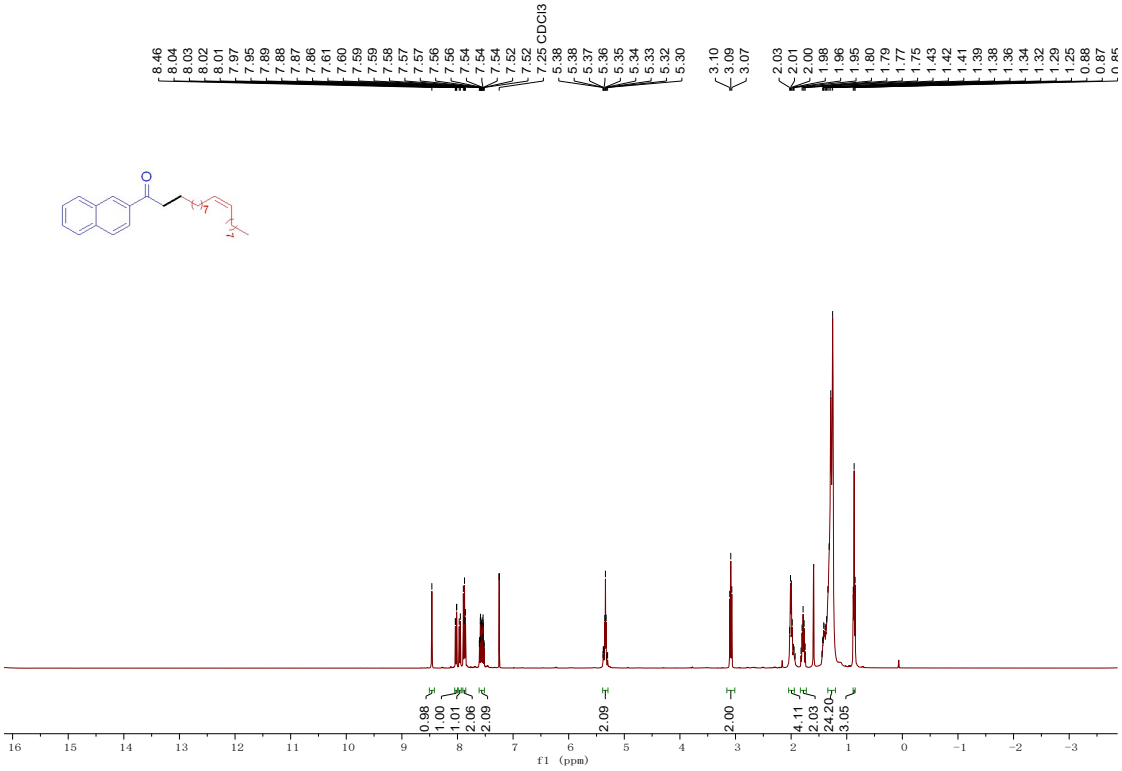


Figure S98. ^{1}H NMR (400 MHz) spectrum of compound 48 in $CDCl_3$

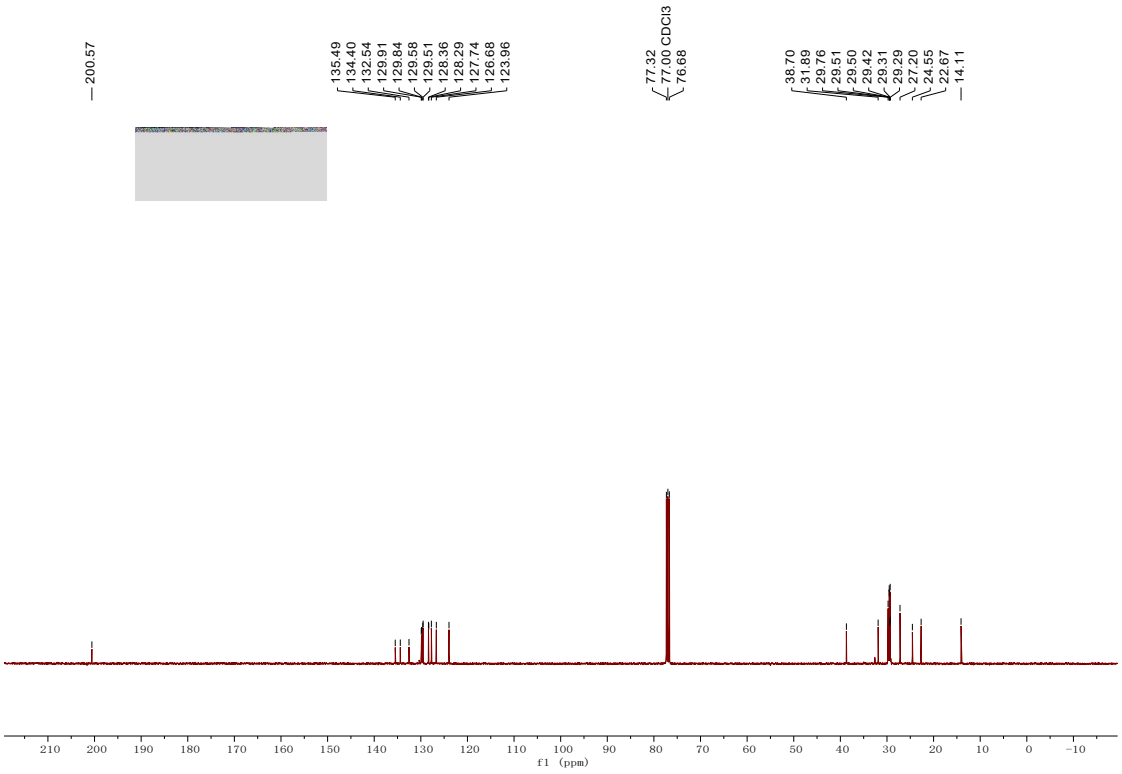


Figure S99. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 48 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **49**

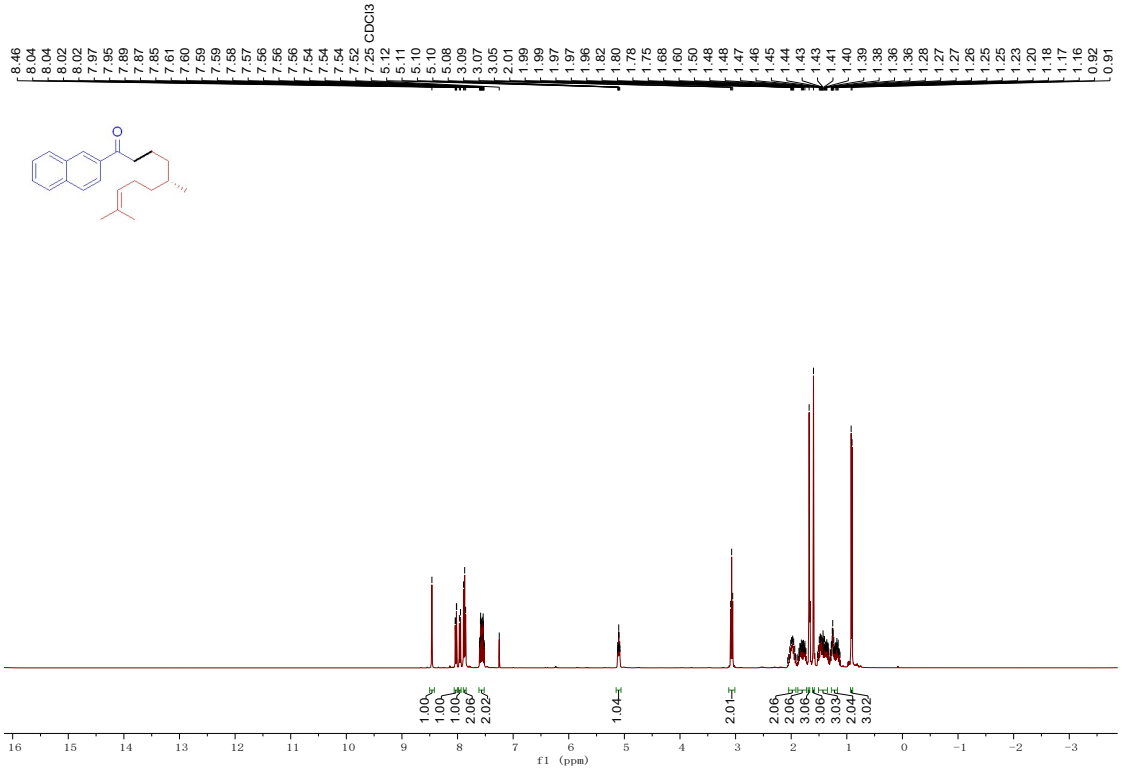


Figure S100. ^{1}H NMR (400 MHz) spectrum of compound 49 in $CDCl_3$

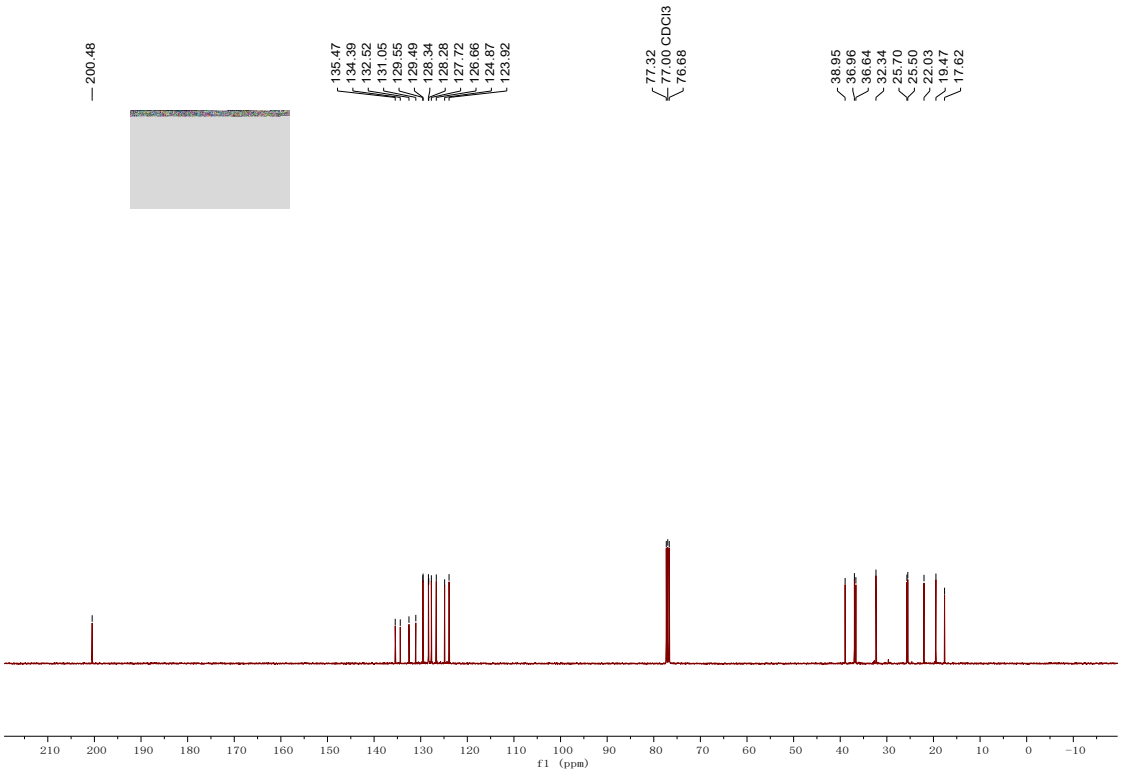


Figure S101. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 49 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **50**

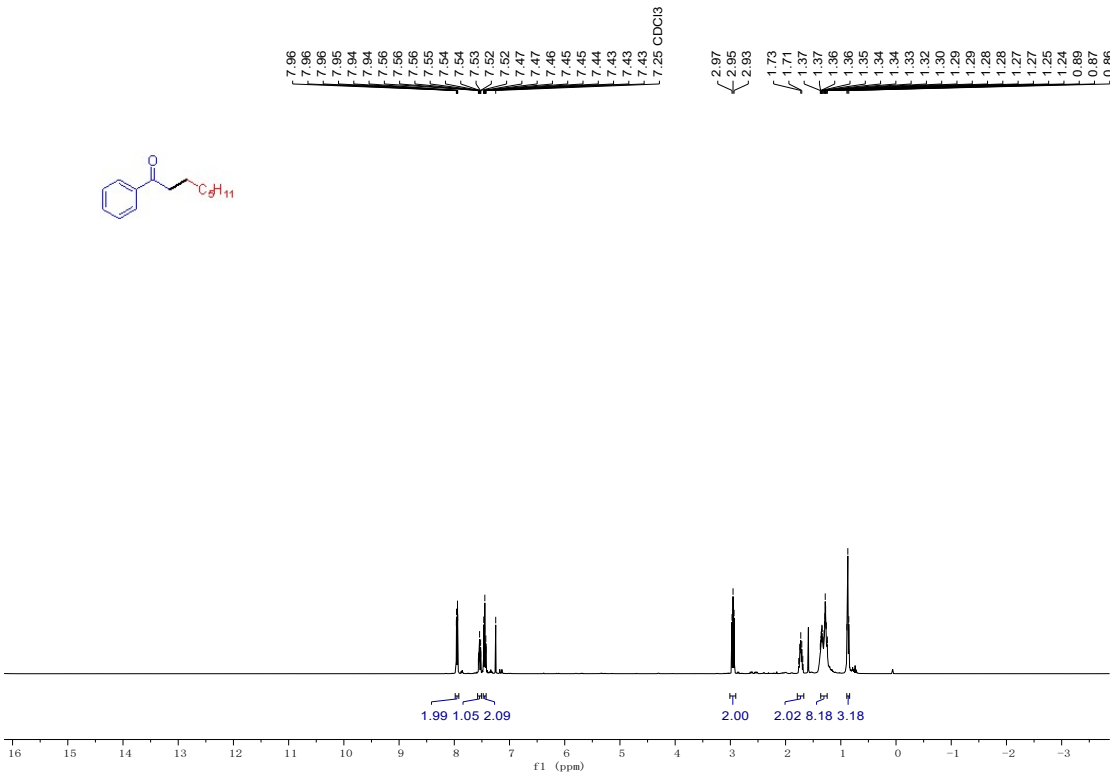


Figure S102. ^{1}H NMR (400 MHz) spectrum of compound 50 in $CDCl_3$

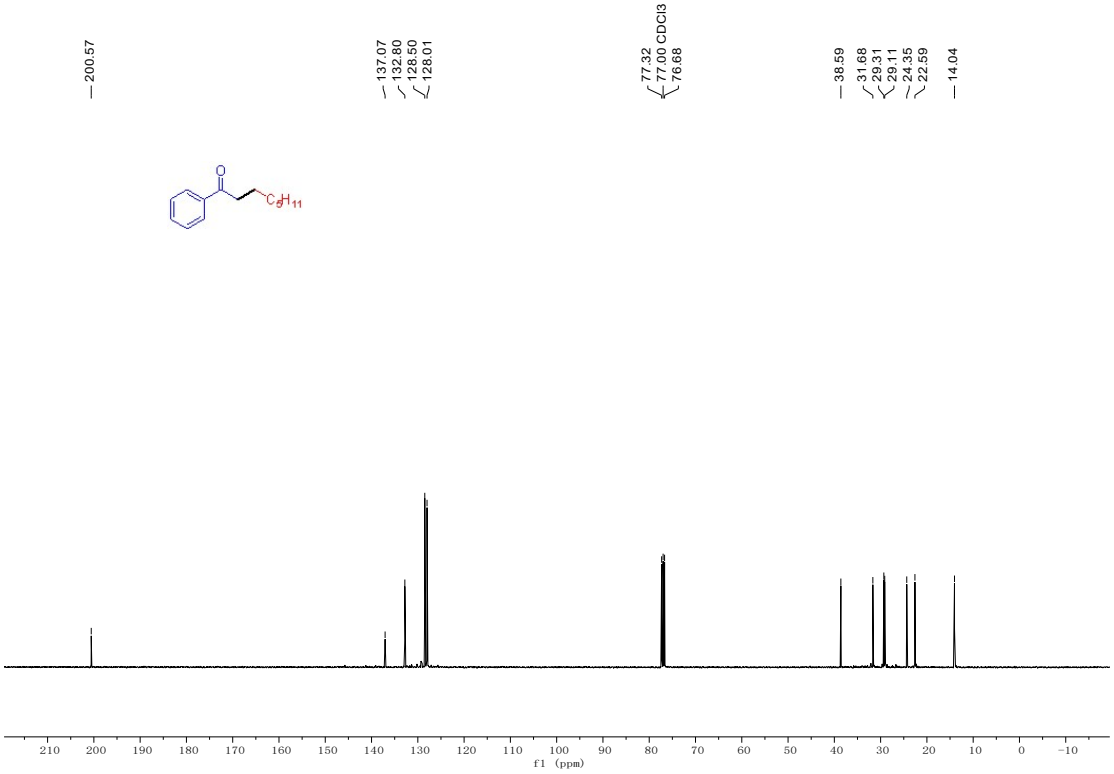


Figure S103. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 50 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **51**

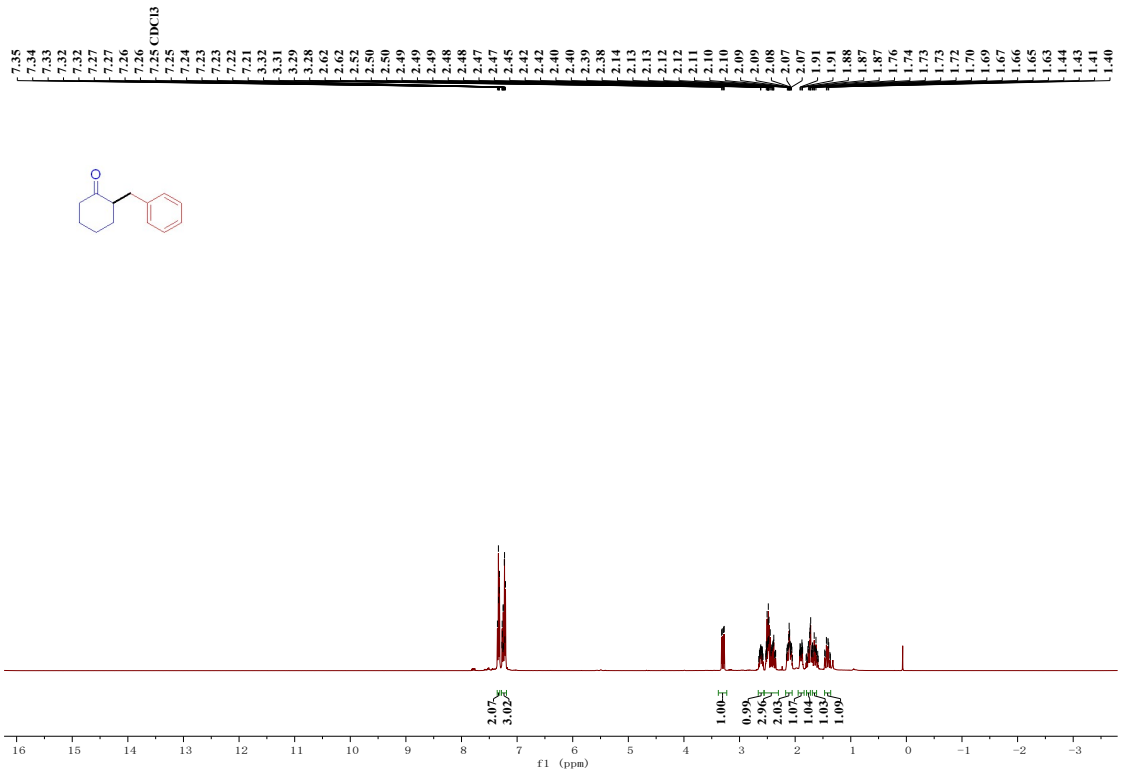


Figure S104. ^{1}H NMR (400 MHz) spectrum of compound 51 in $CDCl_3$

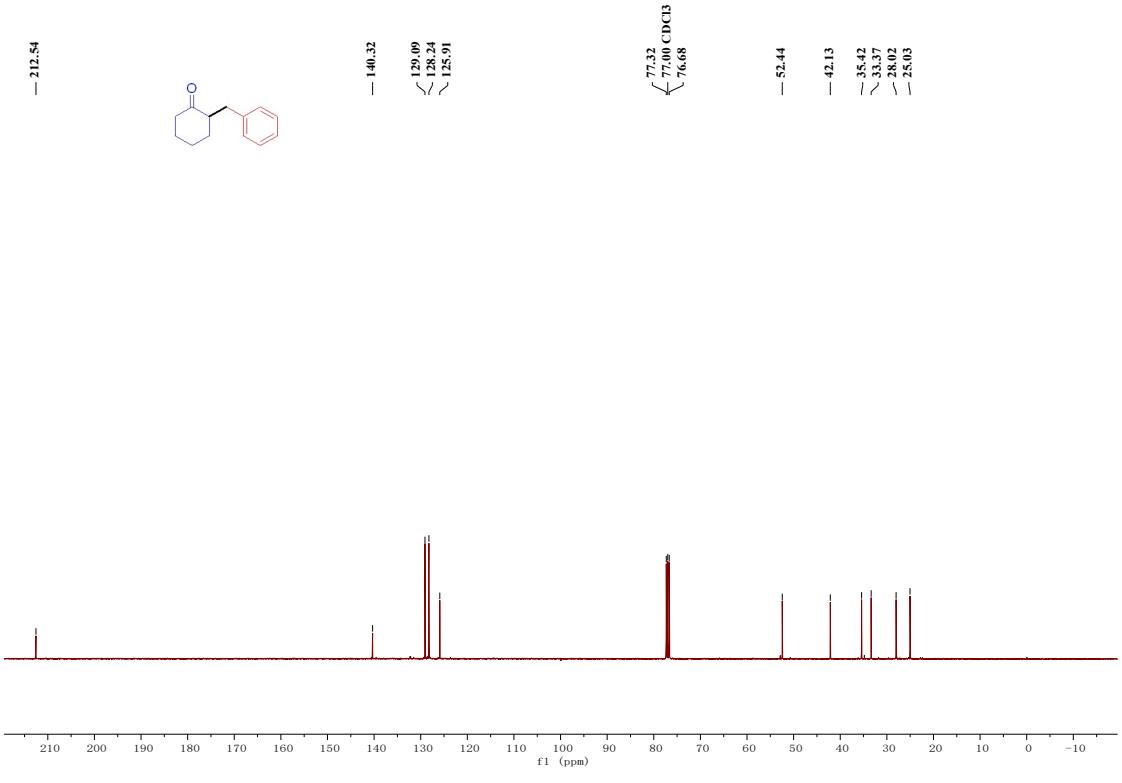


Figure S105. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 51 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **52**

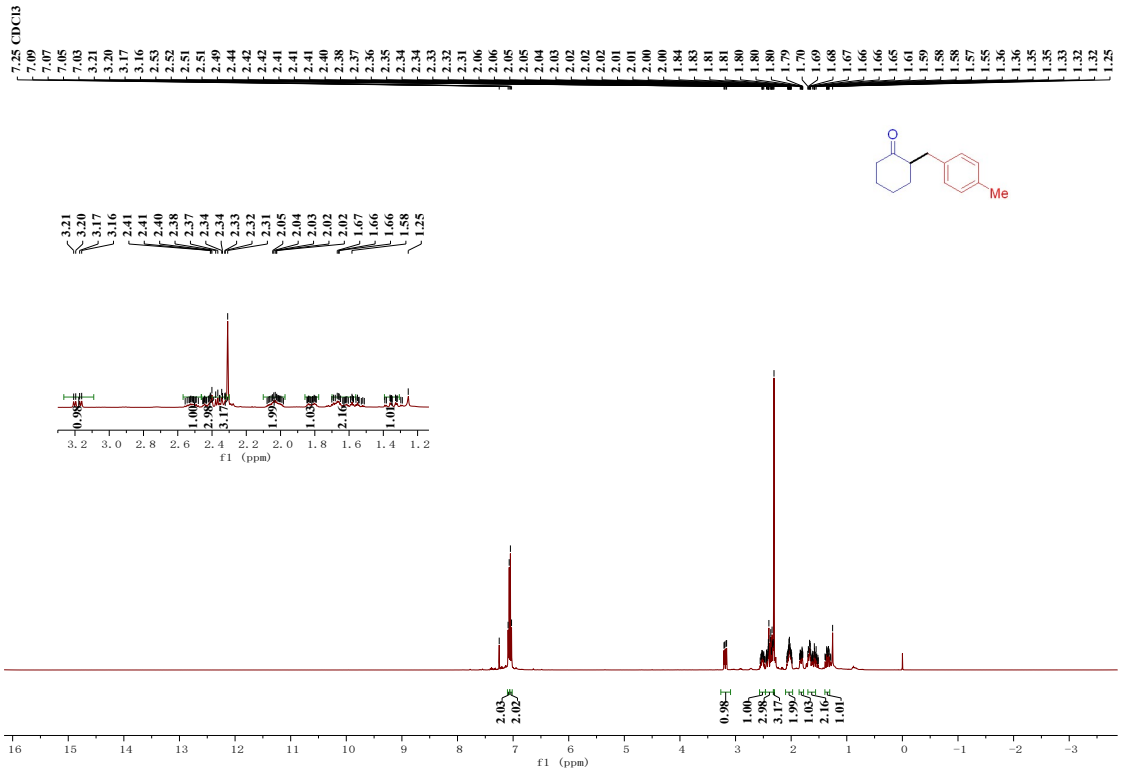


Figure S106. ^{1}H NMR (400 MHz) spectrum of compound 52 in $CDCl_3$

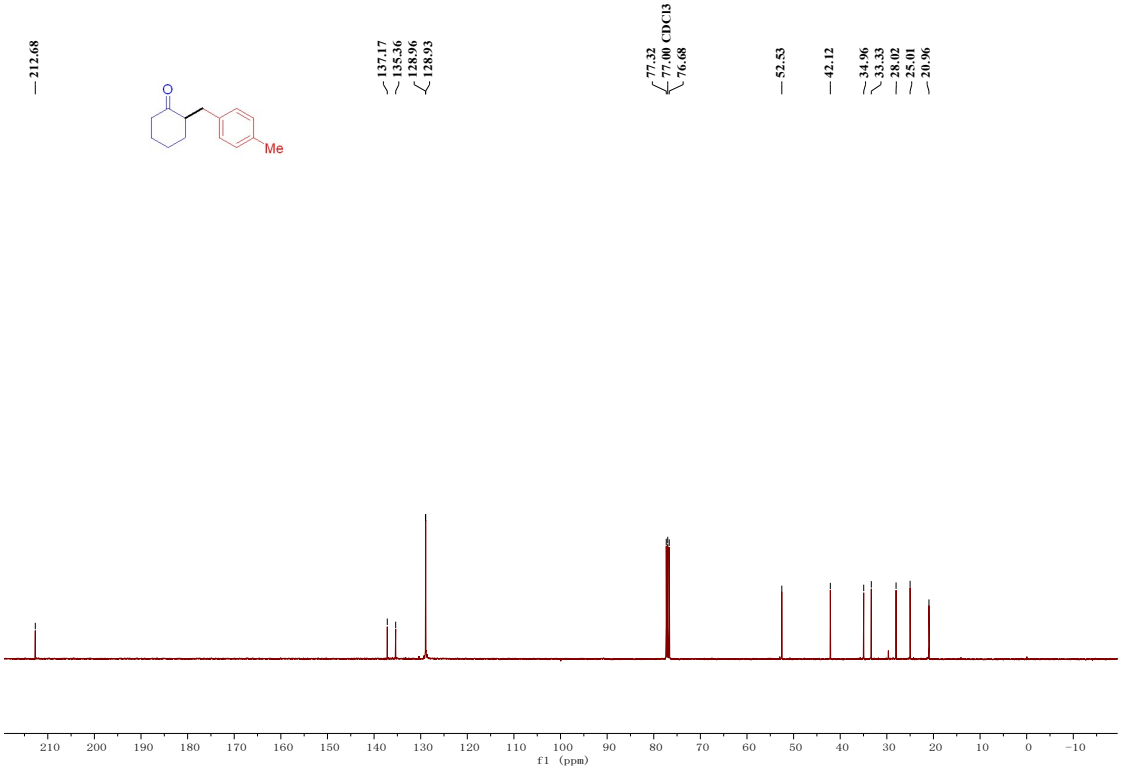


Figure S107. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 52 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **53**

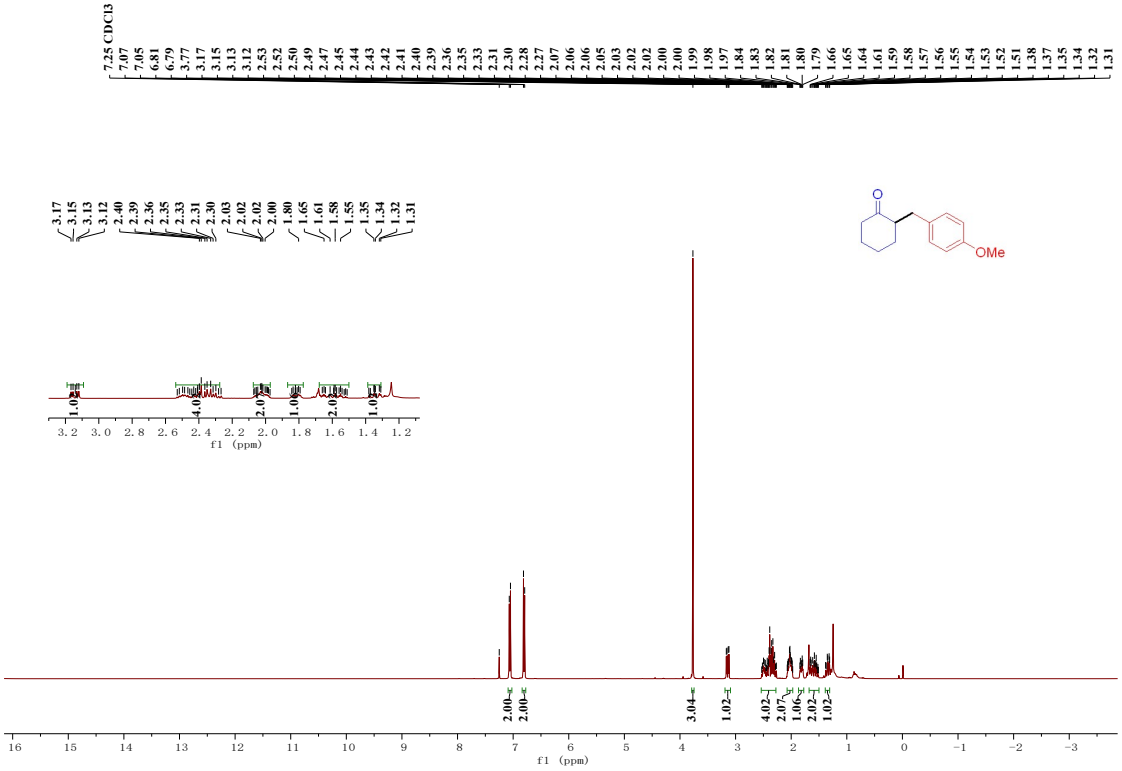


Figure S108. ^{1}H NMR (400 MHz) spectrum of compound 53 in $CDCl_3$

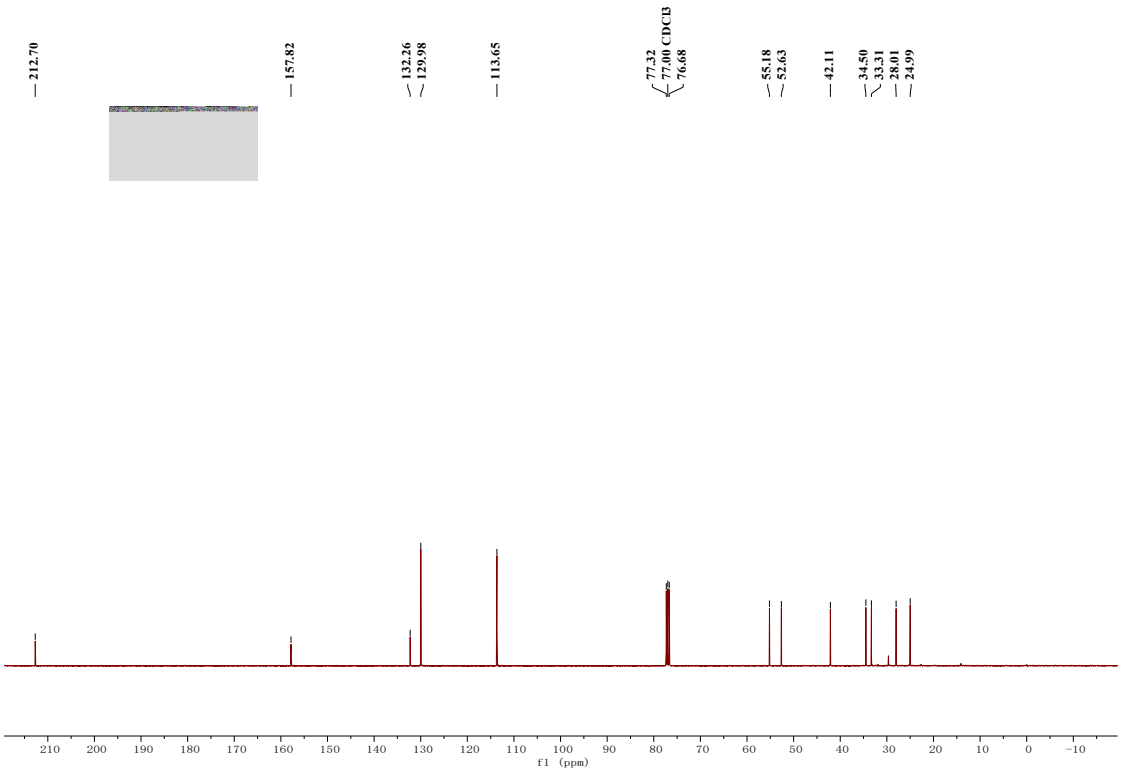


Figure S109. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 53 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **54**

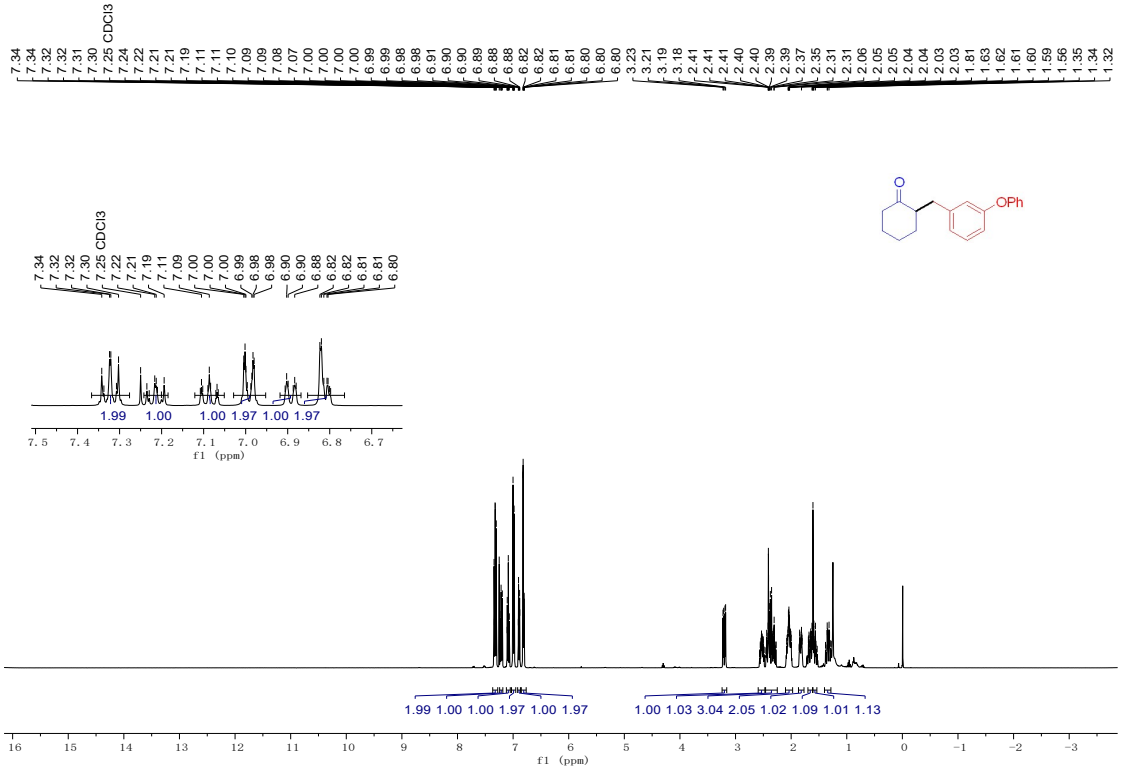


Figure S110. ^{1}H NMR (400 MHz) spectrum of compound 54 in $CDCl_3$

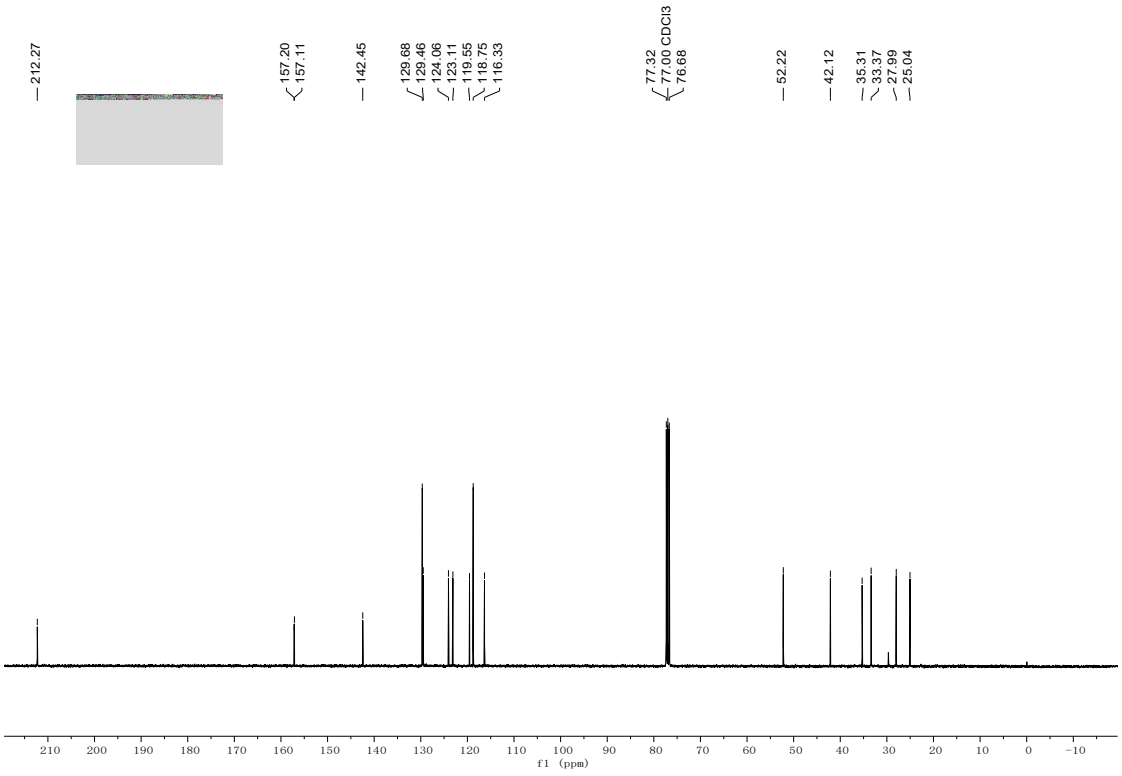


Figure S111. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 54 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **55**

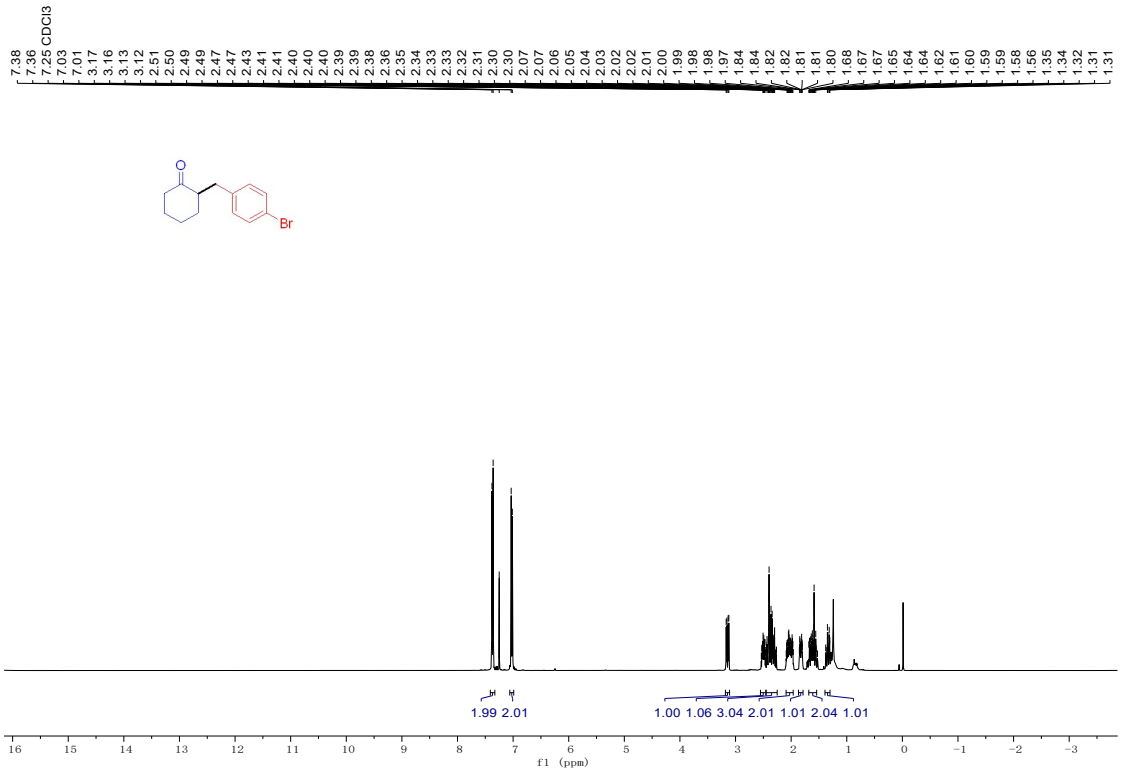


Figure S112. ^{1}H NMR (400 MHz) spectrum of compound 55 in $CDCl_3$

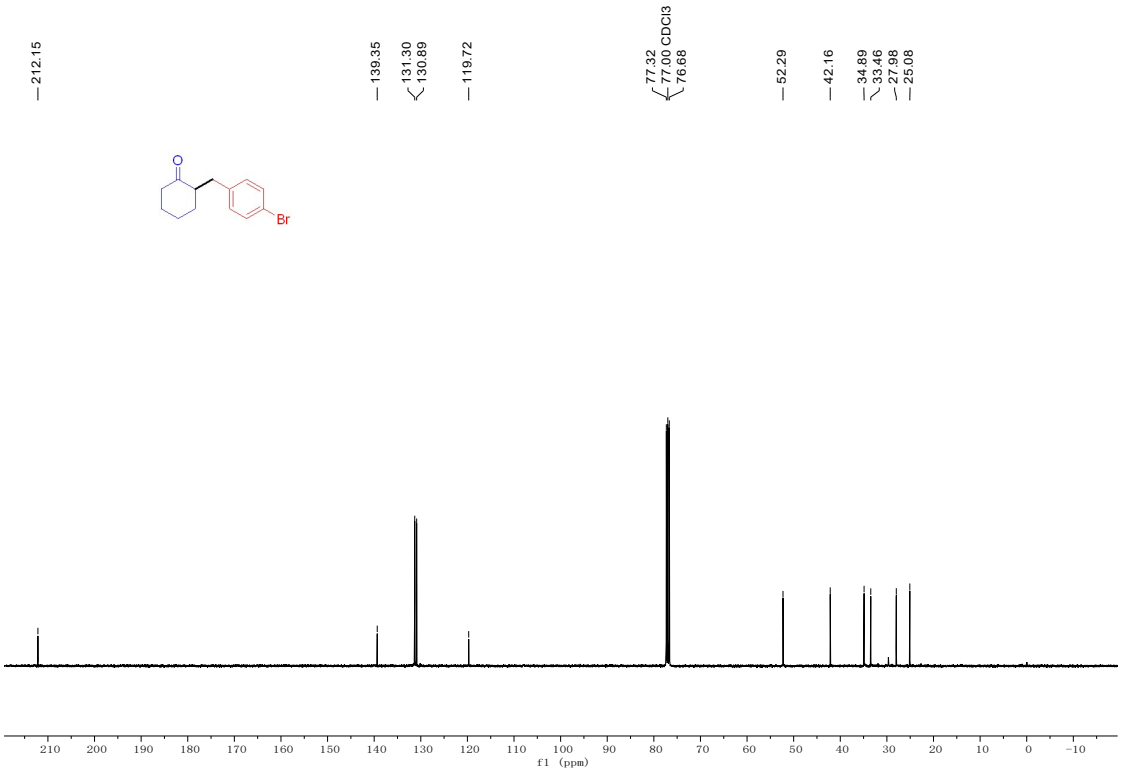


Figure S113. ^{13}C{^{1}H} NMR (101 MHz) spectrum of compound 55 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **56**

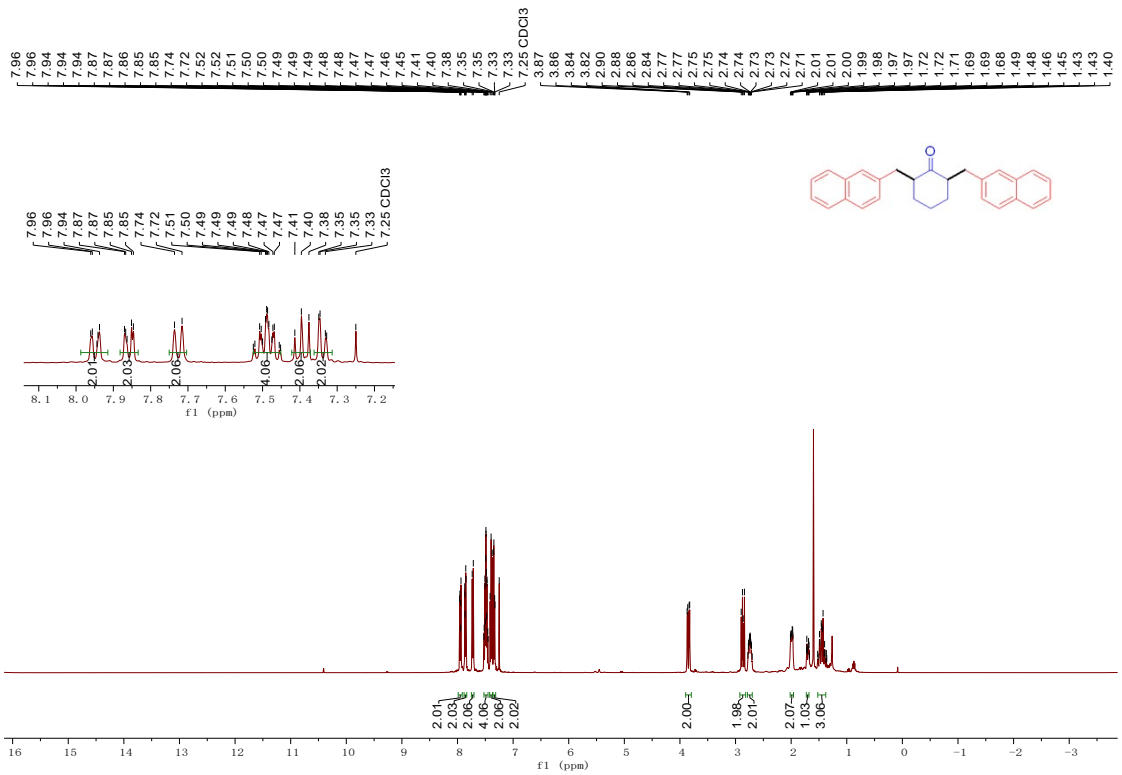


Figure S114. ^{1}H NMR (400 MHz) spectrum of compound 56 in $CDCl_3$

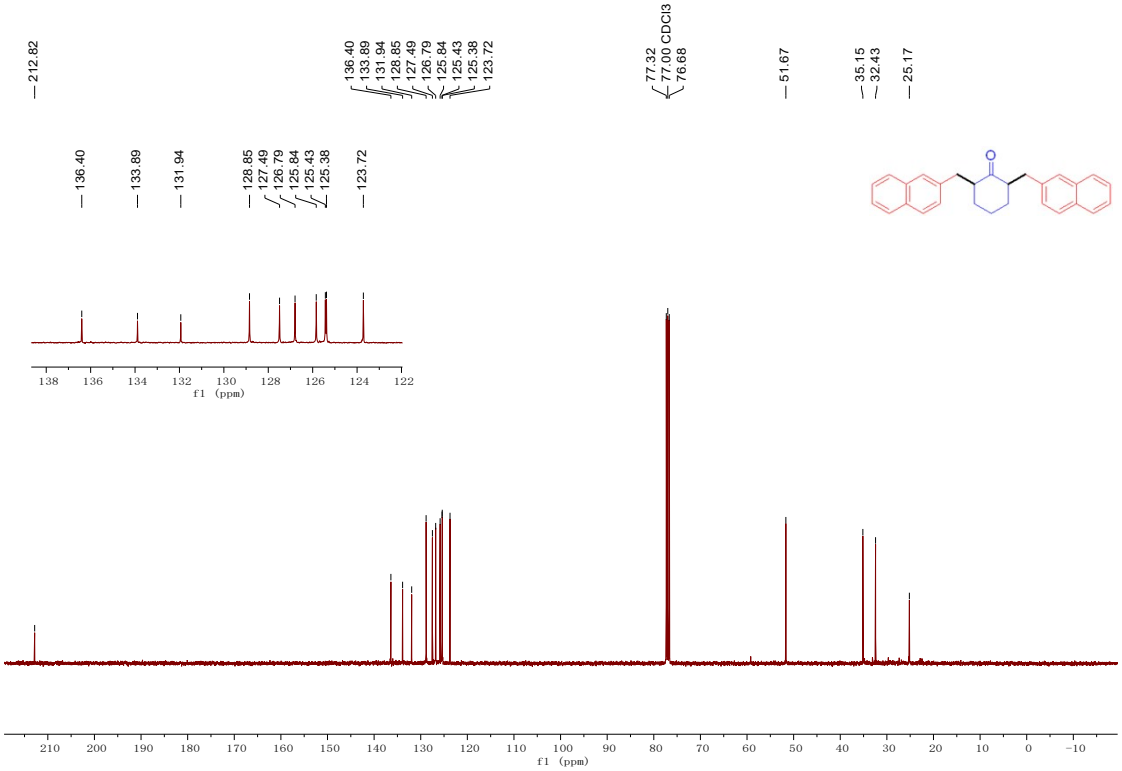


Figure S115. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 56 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **57**

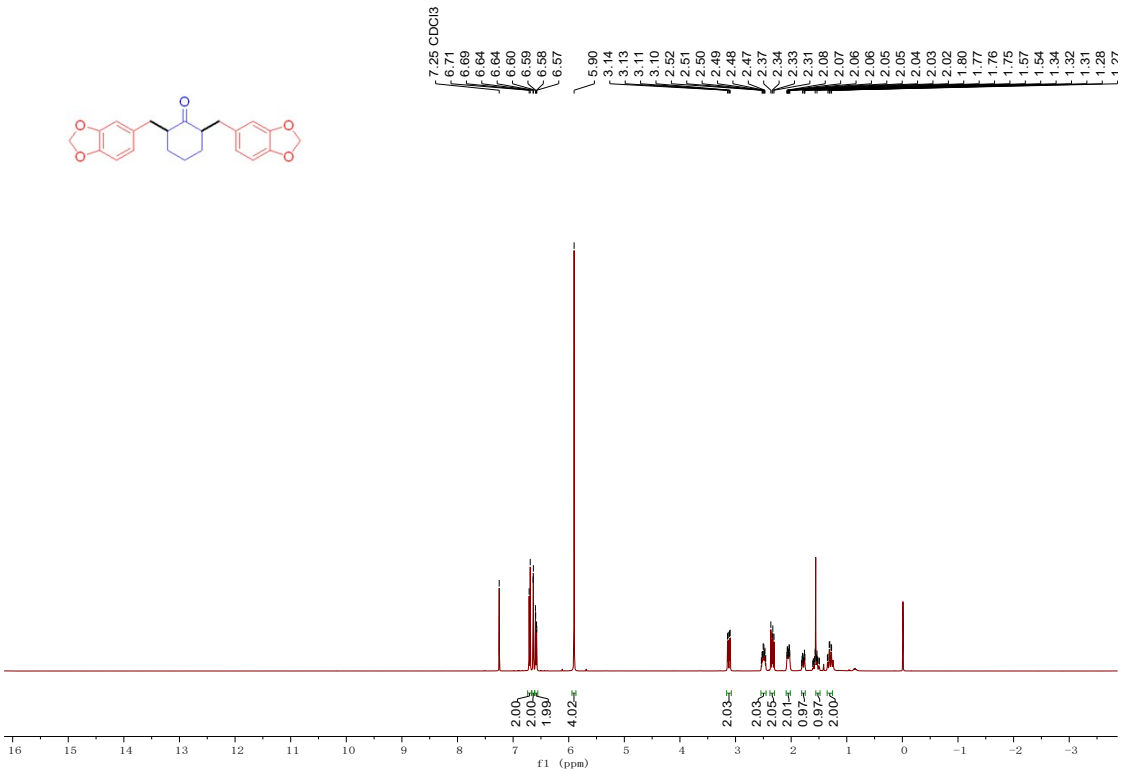


Figure S116. ^{1}H NMR (400 MHz) spectrum of compound 57 in $CDCl_3$

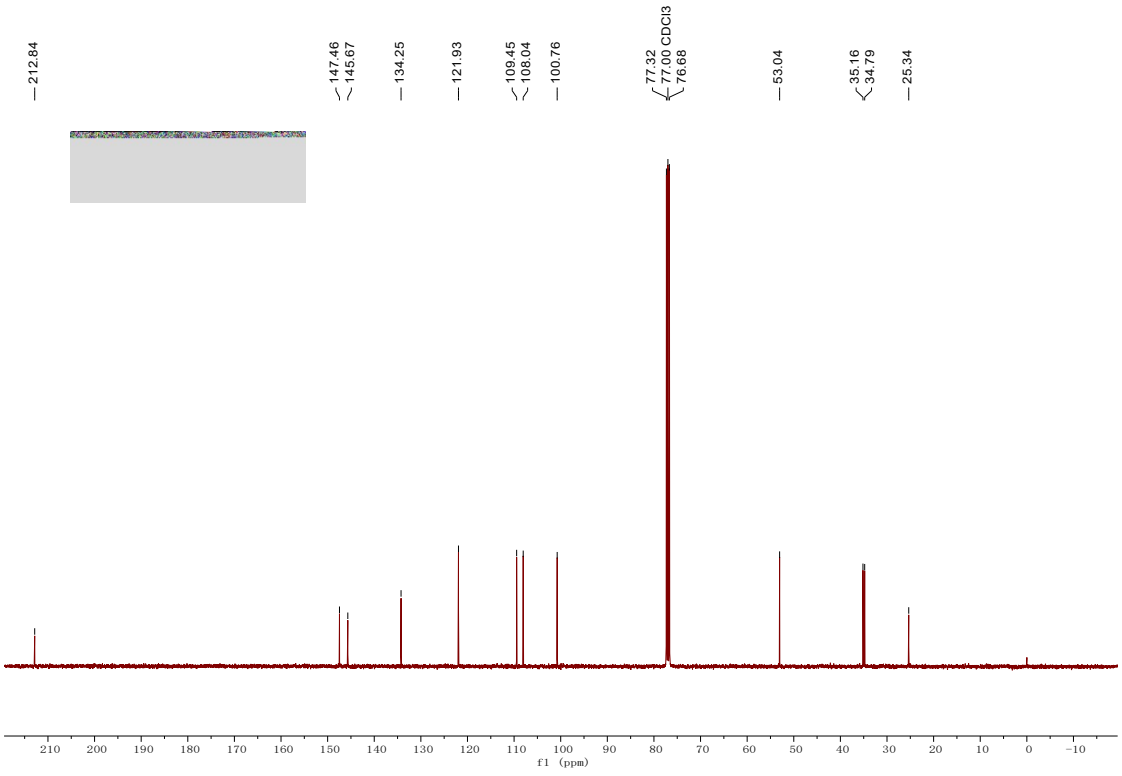


Figure S117. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 57 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **58**

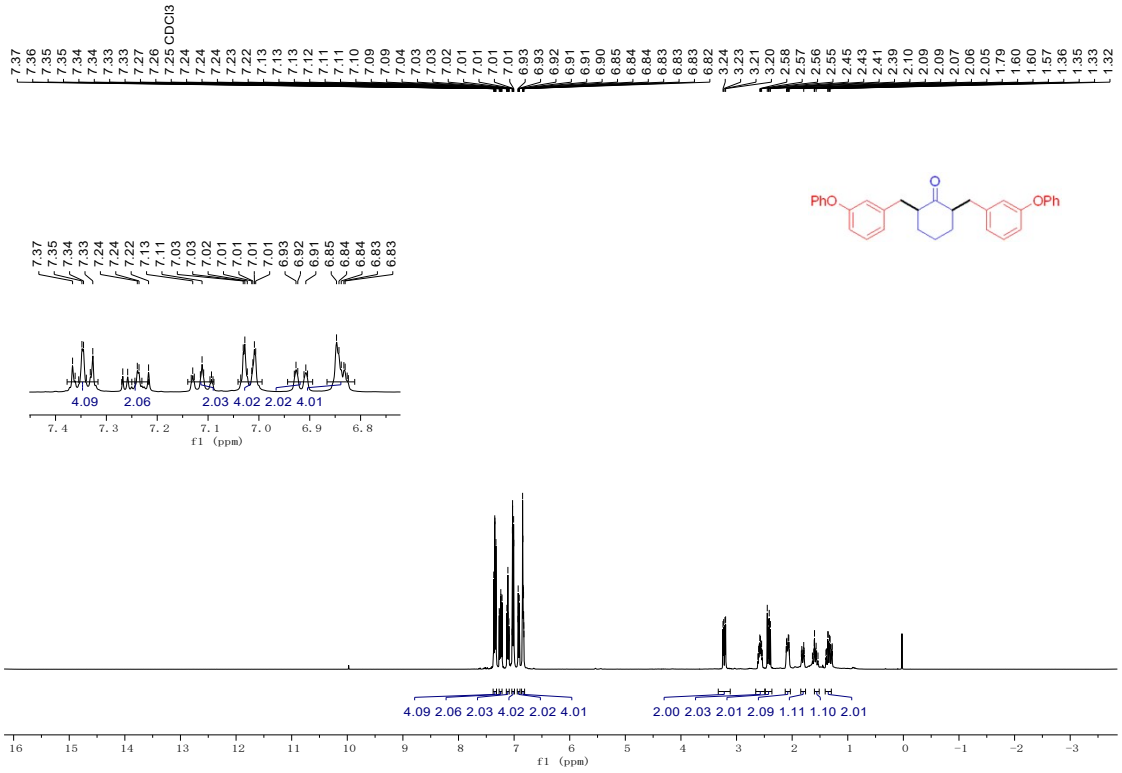


Figure S118. ^{1}H NMR (400 MHz) spectrum of compound 58 in $CDCl_3$

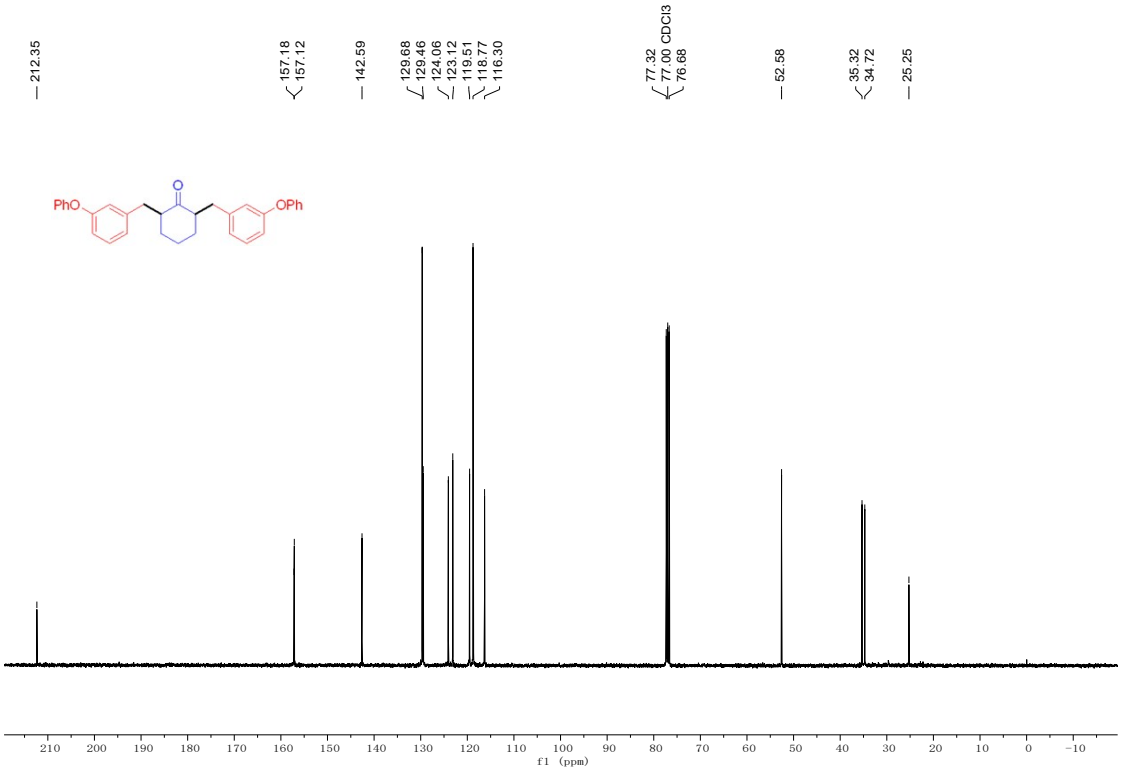


Figure S119. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 58 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **59**

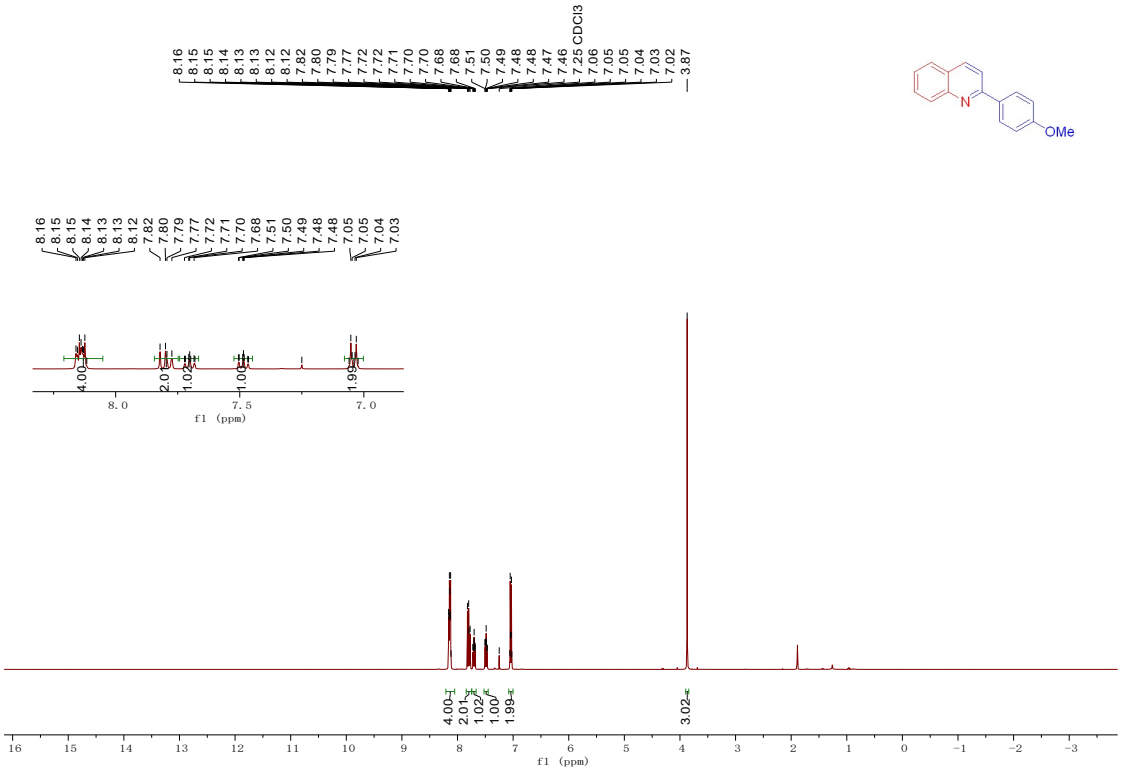


Figure S120. ^{1}H NMR (400 MHz) spectrum of compound 59 in $CDCl_3$

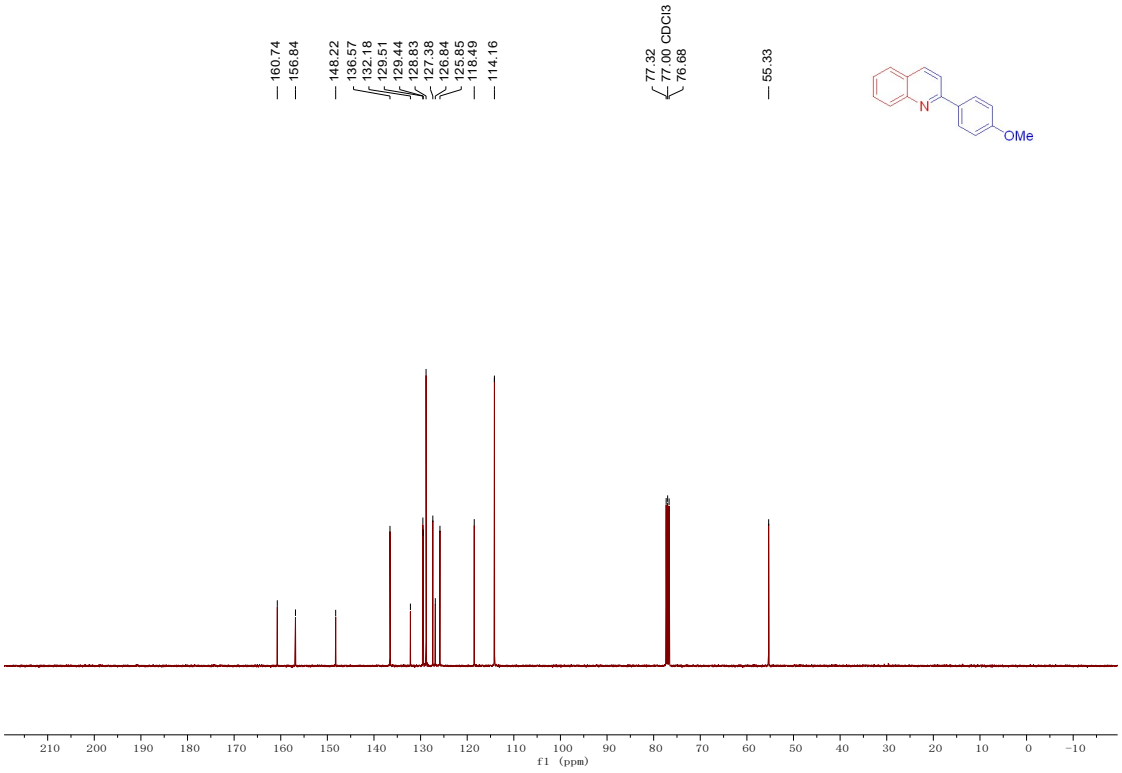


Figure S121. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 59 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **60**

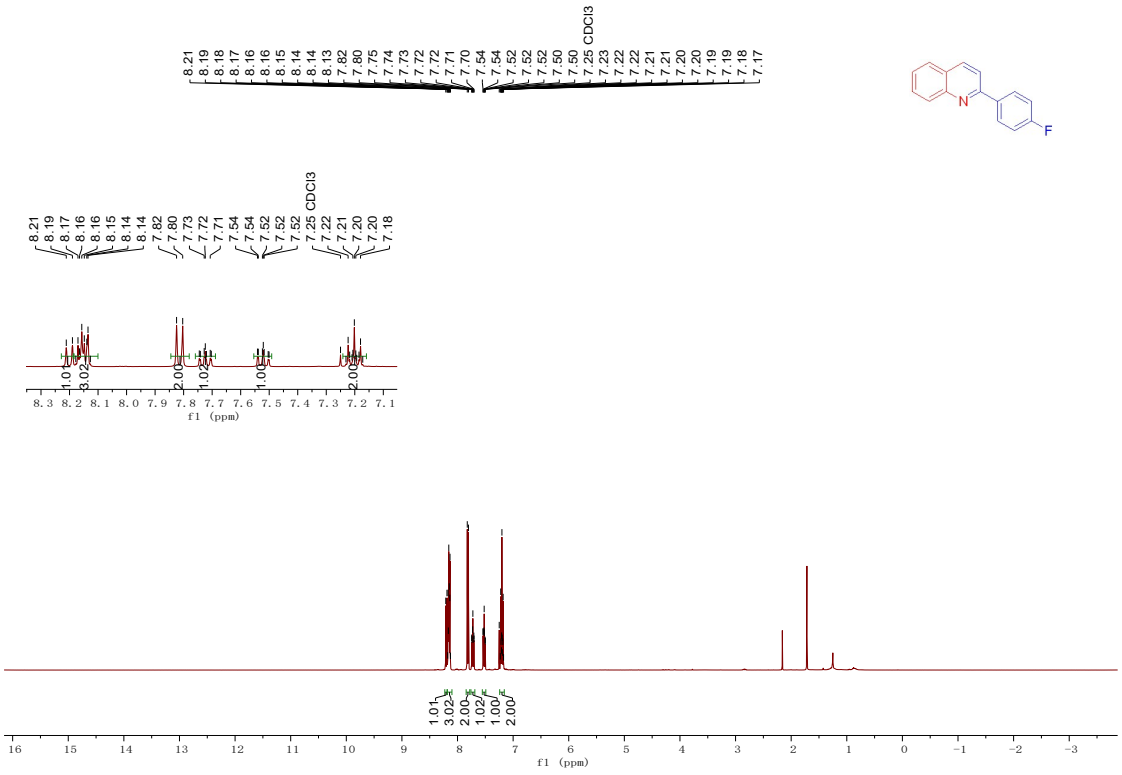


Figure S122. ^{1}H NMR (400 MHz) spectrum of compound 60 in $CDCl_3$

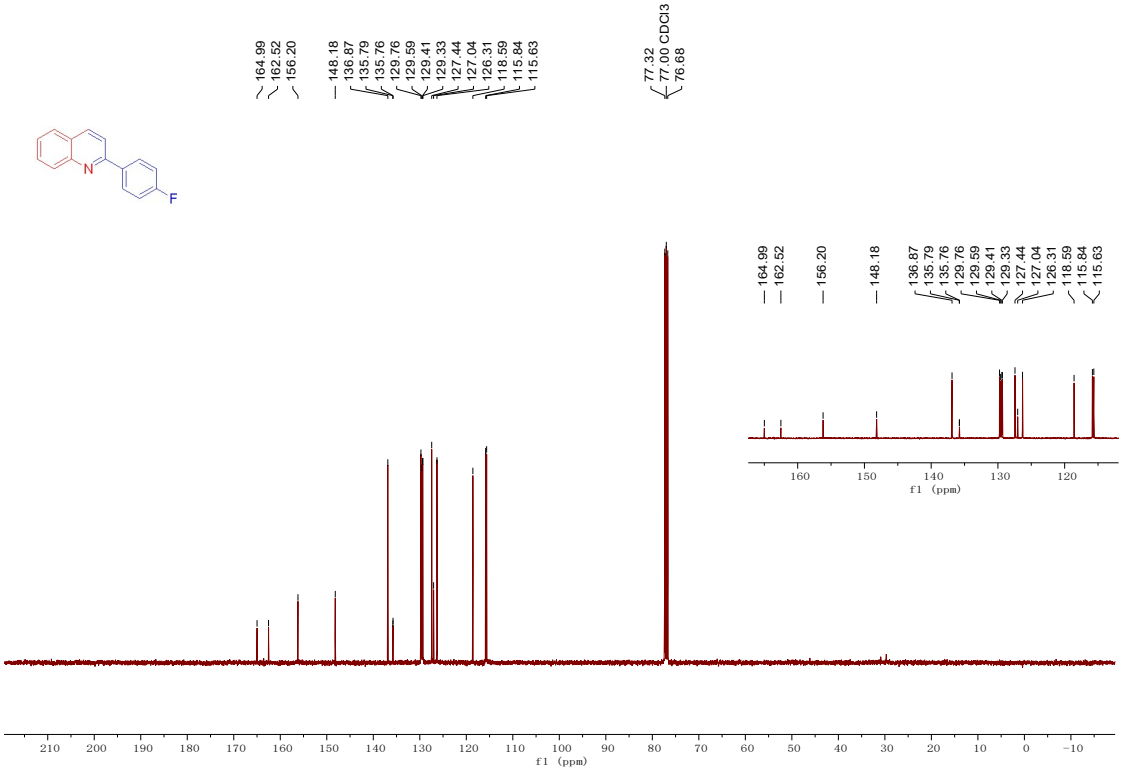


Figure S123. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 60 in $CDCl_3$

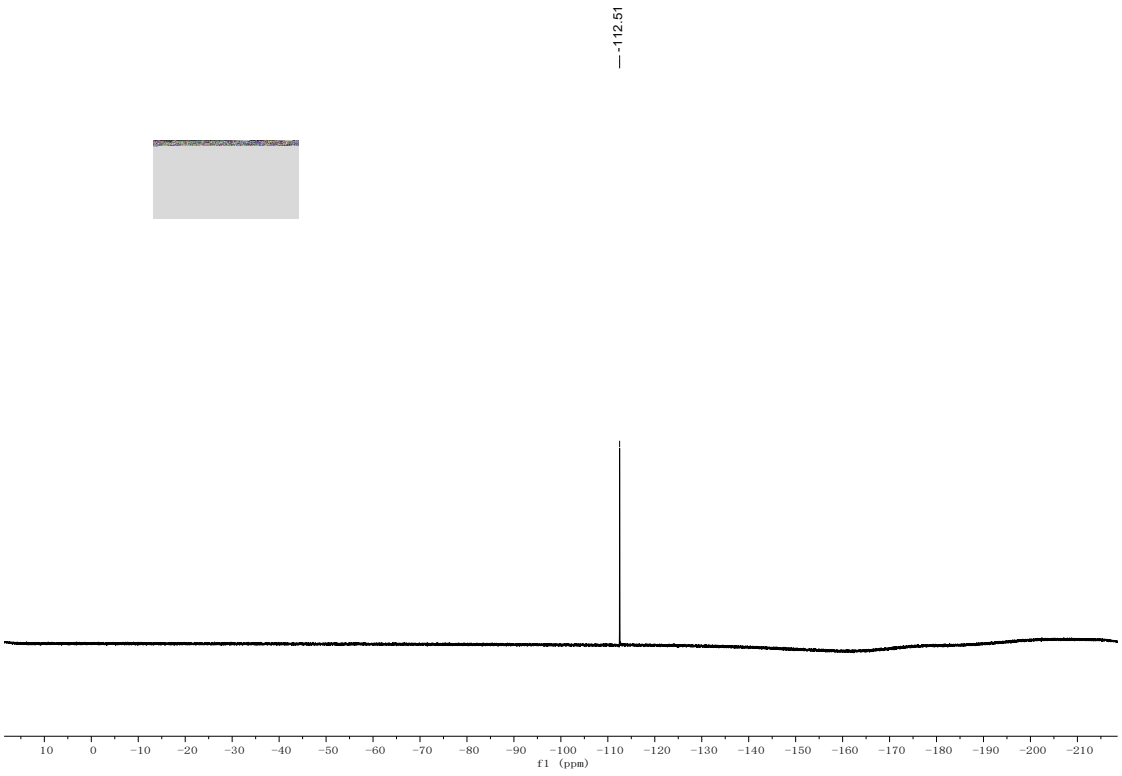


Figure S124. ^{19}F NMR (376 MHz) spectrum of compound 60 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **61**

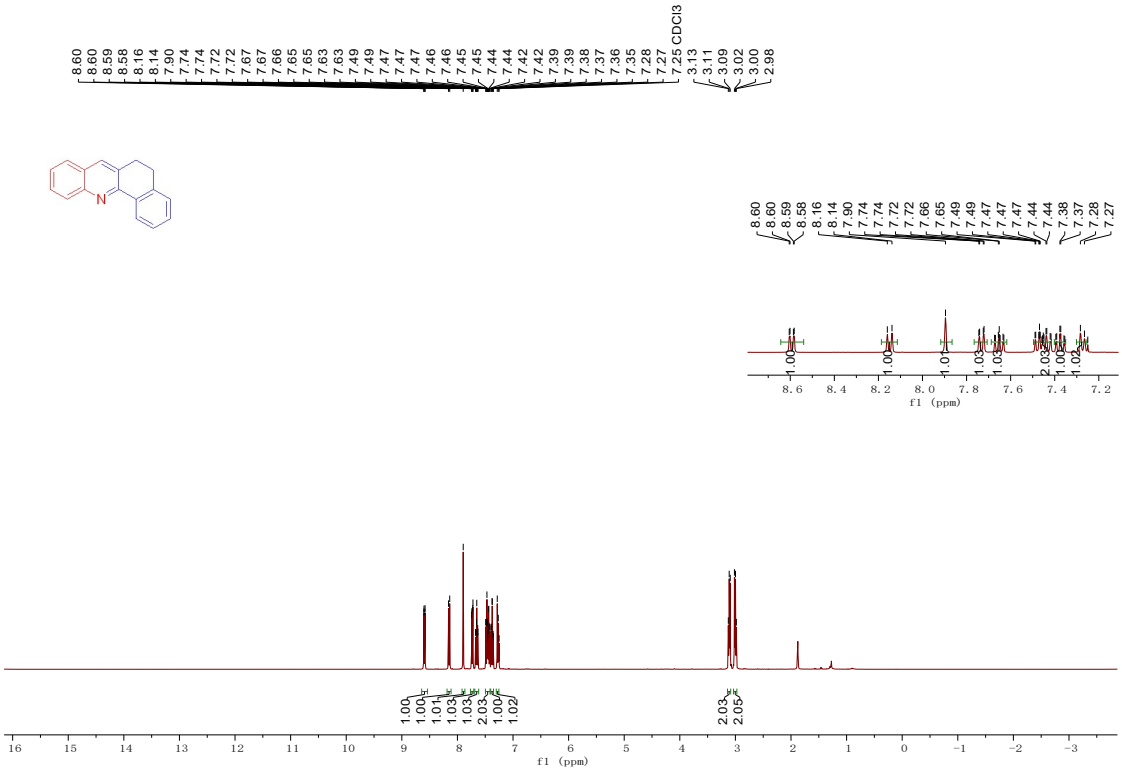


Figure S125. ^{1}H NMR (400 MHz) spectrum of compound 61 in $CDCl_3$

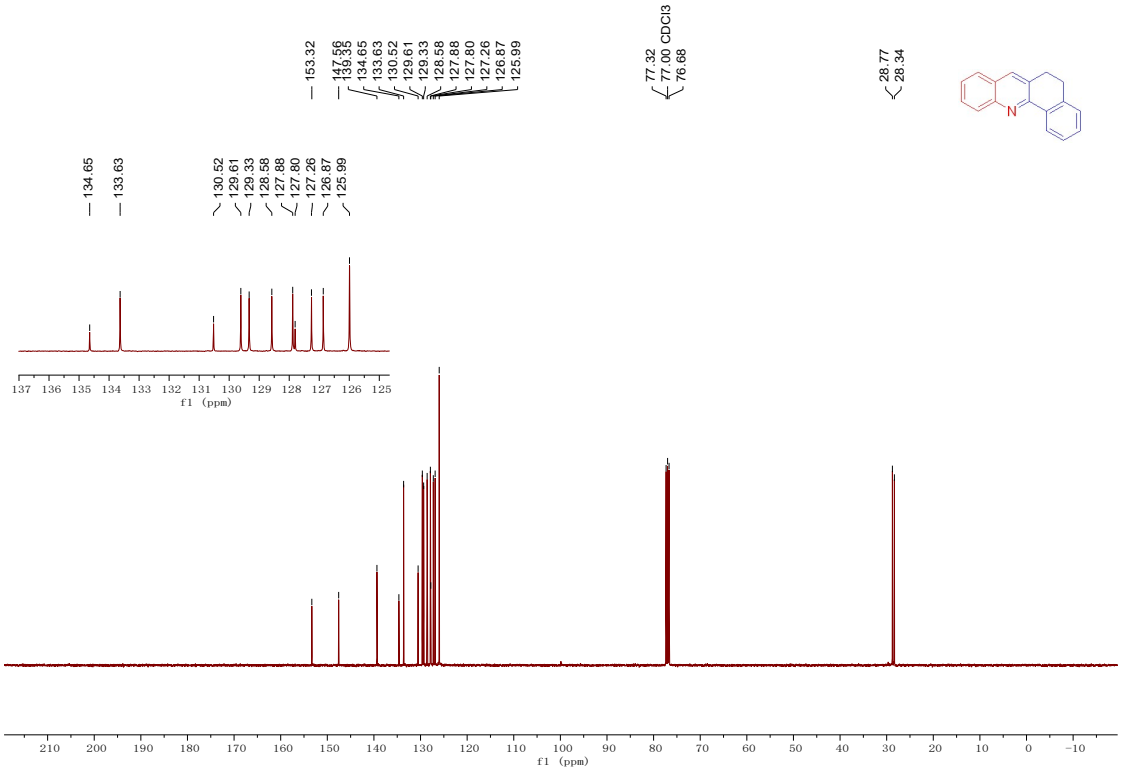


Figure S126. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 61 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **62**

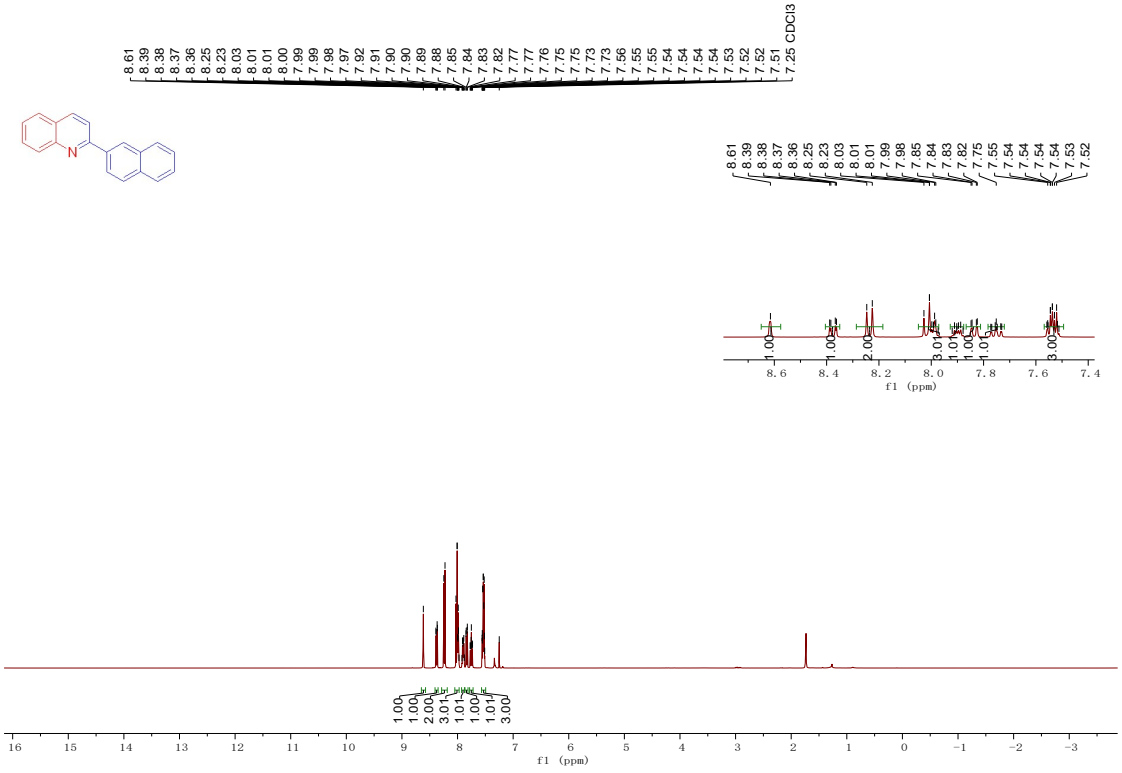


Figure S127. ^{1}H NMR (400 MHz) spectrum of compound 62 in $CDCl_3$

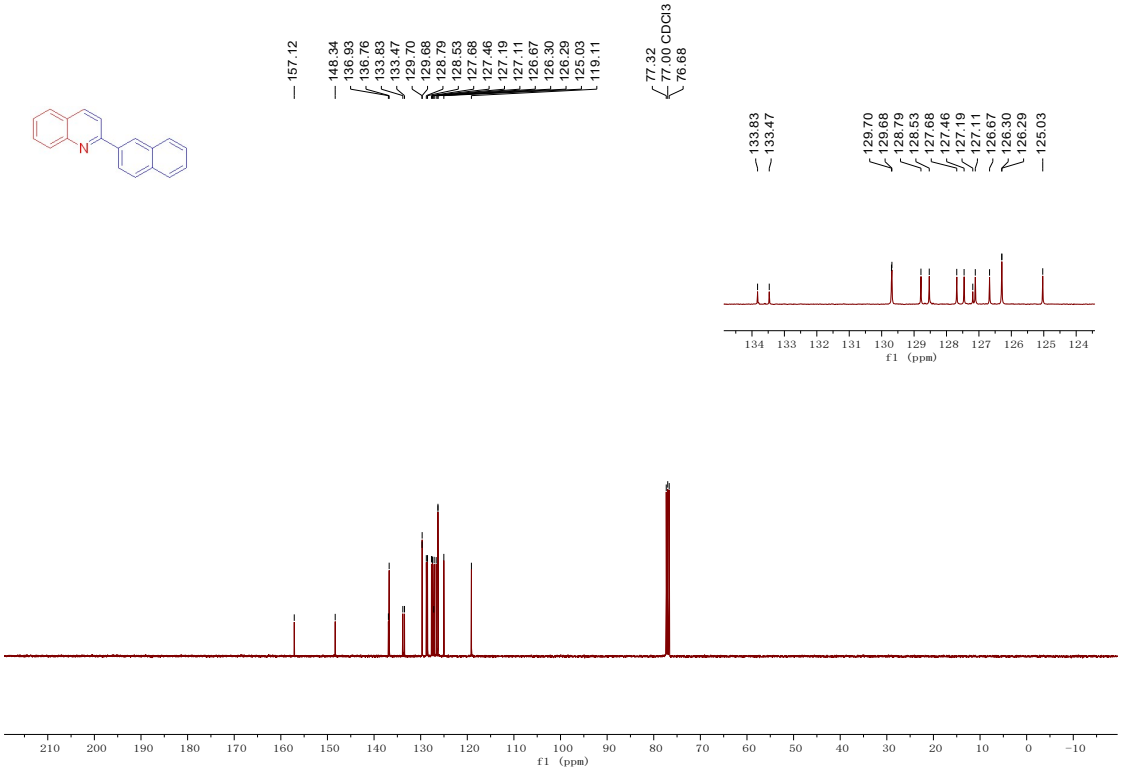


Figure S128. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 62 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **63**

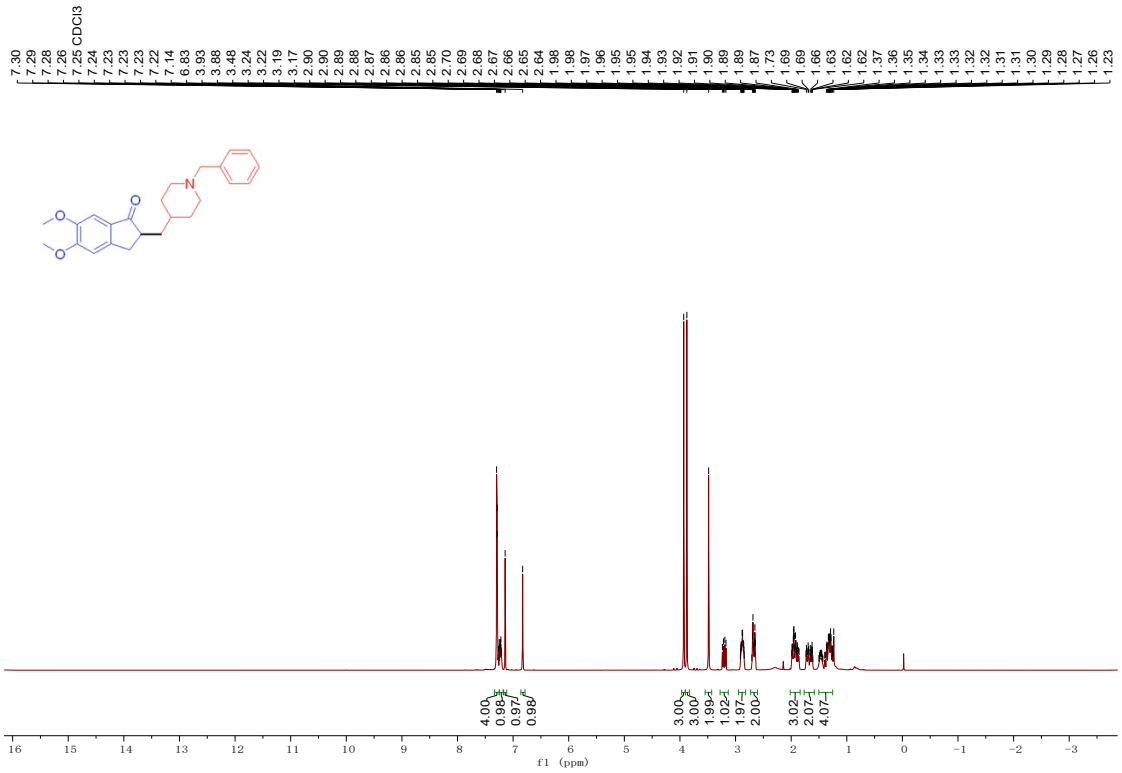


Figure S129. ^{1}H NMR (400 MHz) spectrum of compound 63 in $CDCl_3$

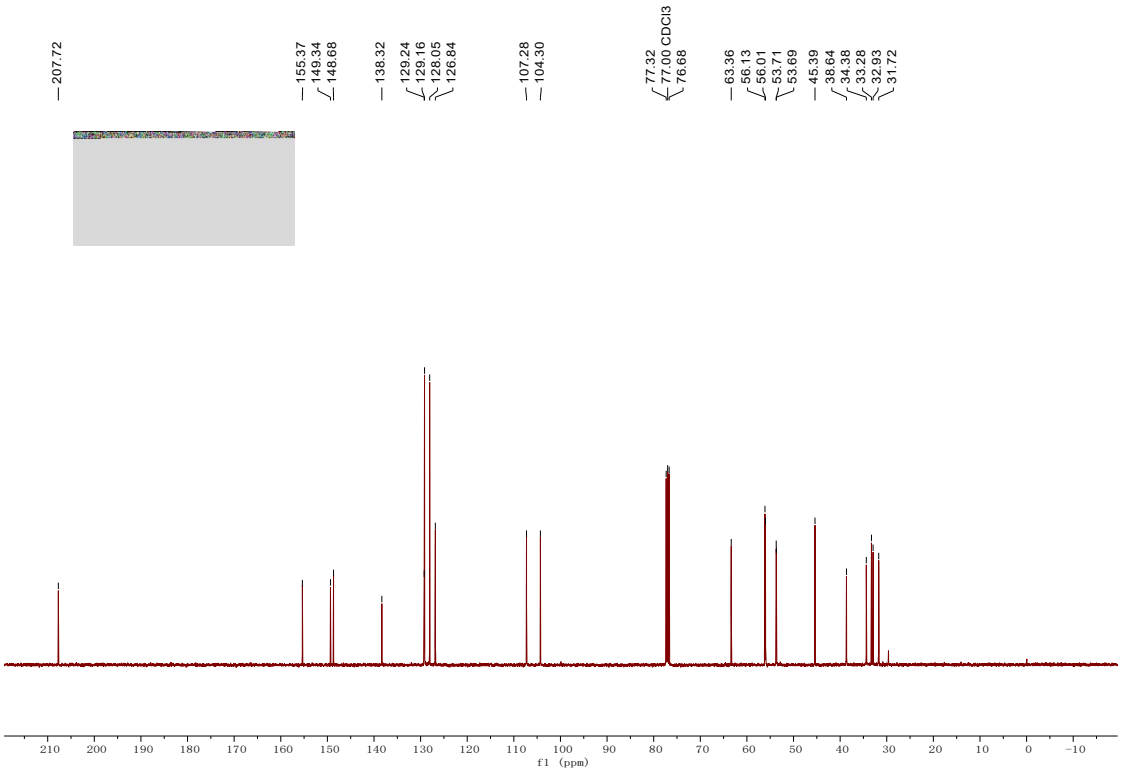
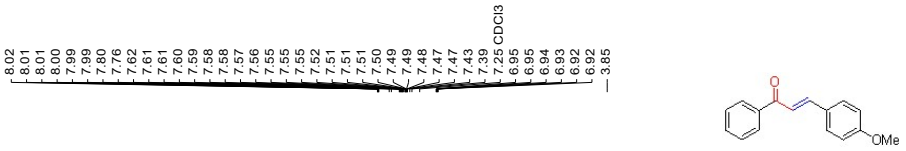


Figure S130. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 63 in $CDCl_3$

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra of compound **3a**



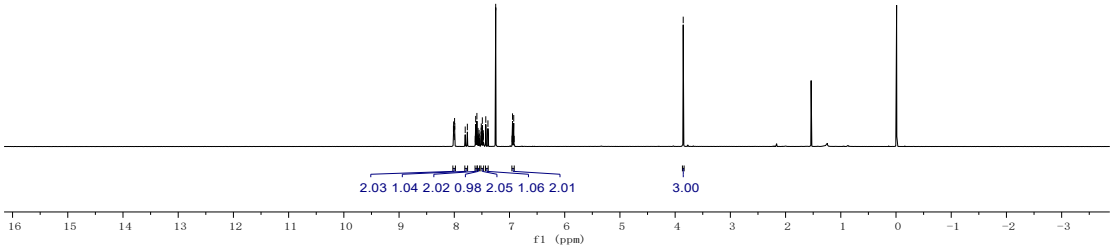


Figure S129. ^{1}H NMR (400 MHz) spectrum of compound 3a in $CDCl_3$

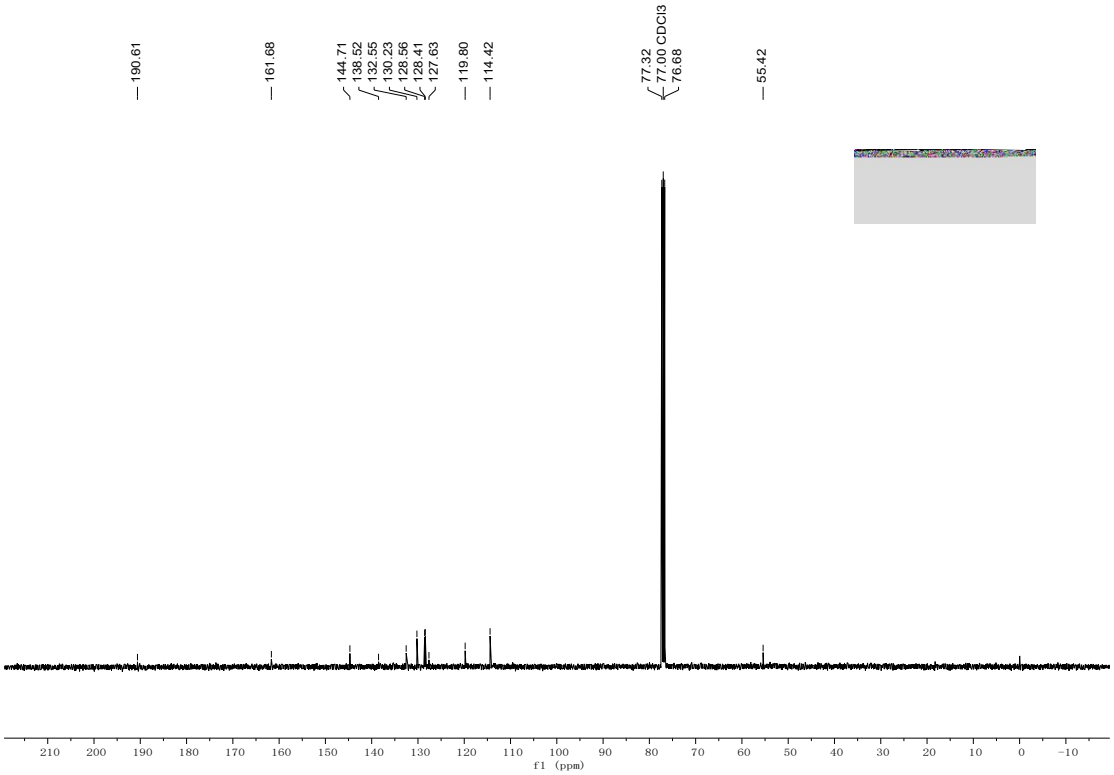


Figure S130. $^{13}C\{^{1}H\}$ NMR (101 MHz) spectrum of compound 3a in $CDCl_3$