Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information for

Strain-release enabled [3+2] annulation of 3-aminooxetanes with simple C=N bonds: facile synthesis of imidazolidines

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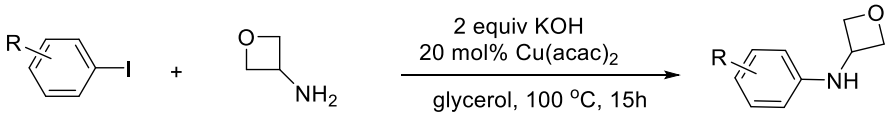
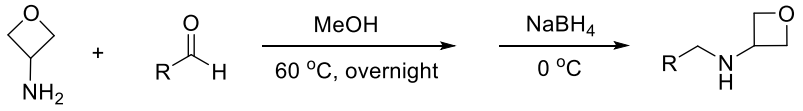
1. General Information

Unless otherwise noted, materials were purchased from commercial suppliers (Alfa, TCI and Sigma-Aldrich etc.), and used without further purification. All the solvents were treated according to general methods. All reactions were monitored by thin-layer chromatography (TLC) on silica gel plates using UV light as visualizing agent (if applicable). Flash column chromatography was performed using 200-300 mesh silica gel. ^{1}H NMR spectra were recorded on 400/600 MHz spectrophotometers. Chemical shifts are reported in delta (δ (ppm)) units in parts per million (ppm) relative to the singlet (0 ppm) for tetramethylsilane (TMS). Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, dd = doublet of doublets, m = multiplet), coupling constants (Hz) and integration. ^{13}C NMR spectra were recorded on Varian Mercury 400 (100 MHz) with complete proton decoupling spectrophotometers ($CDCl_3$: 77.0 ppm). The high resolution mass spectra (HRMS) were measured on a Shimadzu LCMS-IT-TOF mass spectrometer or DIONEX UltiMate 3000 & Bruker Compact TOF mass spectrometer by ESI. Measured values are reported to 4 decimal places of the calculated value. The calculated values are based on the most abundant isotope. An oil bath was used for the synthesis of 1,3,5-triazinanes, and a heating module was used for preparation of compounds **3aa-3af, 3ba-3va, 5aa-5ae** and **7.**

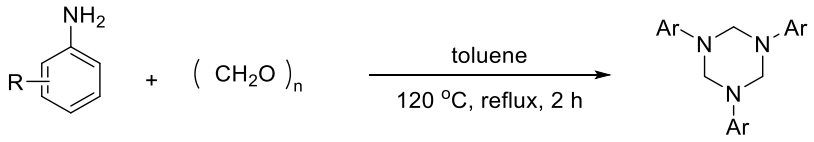
2. Preparation of 3-aminooxetanes and 1,3,5-triazinanes



To an around-bottom flask was added iodobenzene (5 mmol), 3-aminooxetane (7.5 mmol), KOH (10 mmol), $Cu(acac)_2$ (0.1 mmol) and glycerol (25 mL), stirred at 100 °C for 15 h. Then, the reaction mixture was extracted with EtOAc (3 x 10 mL). The combined organic layers dried over Na_2SO_4, and the drying material was removed via gravity filtration and the ethyl acetate filtrate evaporated to dryness under reduced pressure. The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1).



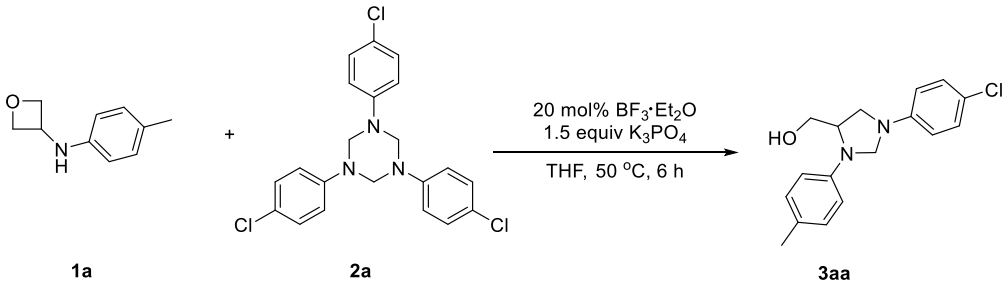
To an around-bottom flask was added the 3-aminooxetane (5 mmol) and aldehyde (6 mmol) in MeOH (20 mL). The reaction mixture was stirred at 60 °C overnight. The mixture was then cooled to 0 °C and $NaBH_4$ was added slowly. The reaction mixture was allowed to warm to room temperature, and stirred for 1h before it was quenched with NH_4Cl (20 mL), extracted with EtOAc (3×20 mL). The combined organic layers dried over Na_2SO_4, and the drying material was removed via gravity filtration and the ethyl acetate filtrate evaporated to dryness under reduced pressure. The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1).



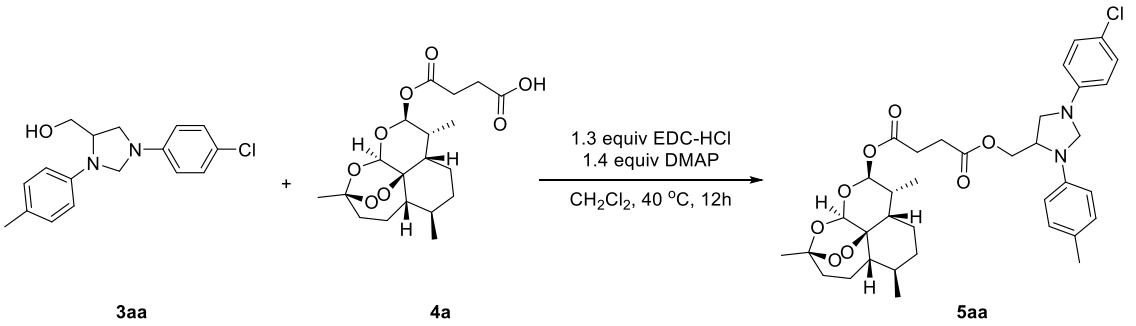
To an around-bottom flask was added the anilines (20 mmol) and paraformaldehyde (20 mmol) in toluene (24 mL), stirred at 120 °C for 2 h. Remove the solvent under reduced pressure, the solid was directly washed by cooled petroleum ether to give the pure product as white solids. All of 1,3,5-triazinanes 2 are known compounds.

3. General Procedure and Spectral Data of the Products

3.1 General procedure for the synthesis of 3aa-3ai, 3ba-3wa, 5aa-5ae, 7, 9 and gram scale reaction

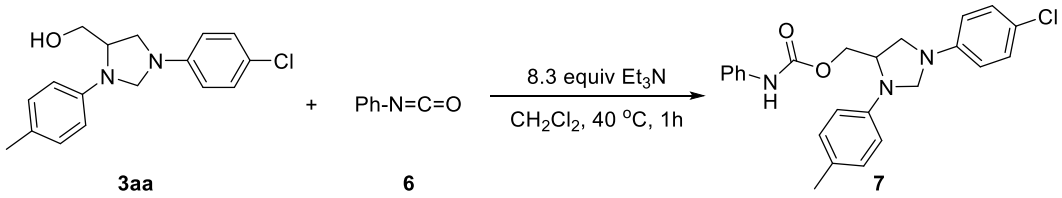


To a dried flask was added **1a** (48.9 mg, 0.3 mmol), **2a** (100.4 mg, 0.24 mmol), and K_3PO_4 (92.5 mg, 0.45 mmol) in THF (3 mL). Then, $BF_3{\cdot}Et_2O$ (20 mol%) was added to the mixture. The reaction mixture was stirred at 50 °C for 6 h. The crude product was purified by chromatography on silica gel directly to give the desired product **3aa** in 88% isolated yield as a white solid. Other products **3ab-3ai** and **3ba-3wa** were prepared according to this procedure.



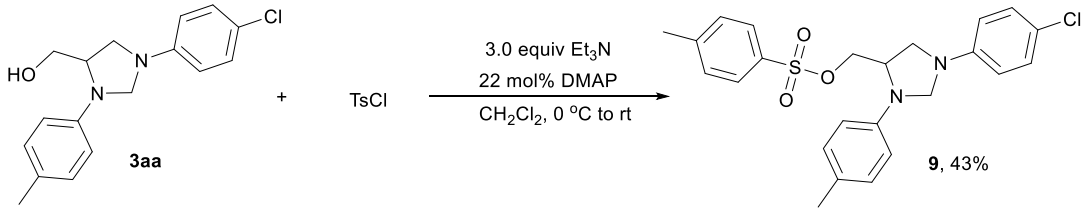
3aa **4a** **5aa**

To a dried flask was added **3aa** (60.6 mg, 0.2 mmol), **4a** (64.3 mg, 0.17 mmol), EDC-HCl (41.4 mg, 0.22 mmol), DMAP (28.6 mg, 0.23 mmol), and CH_2Cl_2 (2 mL). The reaction mixture was stirred at 40 °C for 12 h. The crude product was purified by chromatography on silica gel directly to give the desired product **5aa** in 92% isolated yield as a white solid. Other products **5ba-5ae** were prepared according to this procedure.



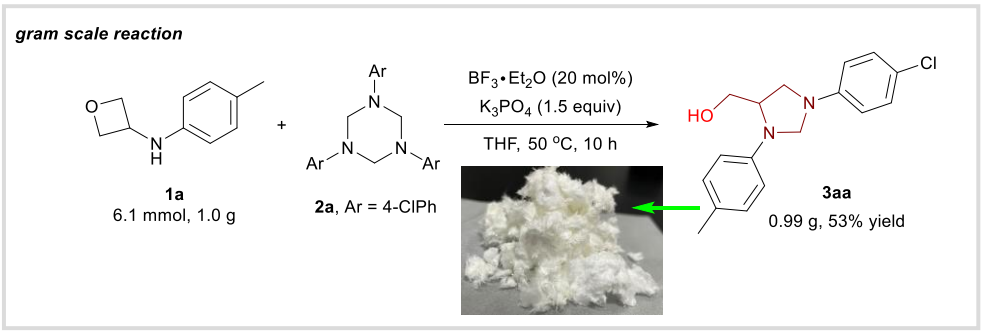
3aa **6** **7**

To a dried flask was added **3aa** (60.6 mg, 0.2 mmol), isocyanate (16.7 mg, 0.14 mmol), Et_3N (168 mg, 1.66 mmol) in CH_2Cl_2 (1 mL). The reaction mixture was stirred at 40 °C for 1 h. The crude product was purified by chromatography on silica gel directly to give the desired product **7** in 57% isolated yield as a white solid.



3aa **9**, 43%

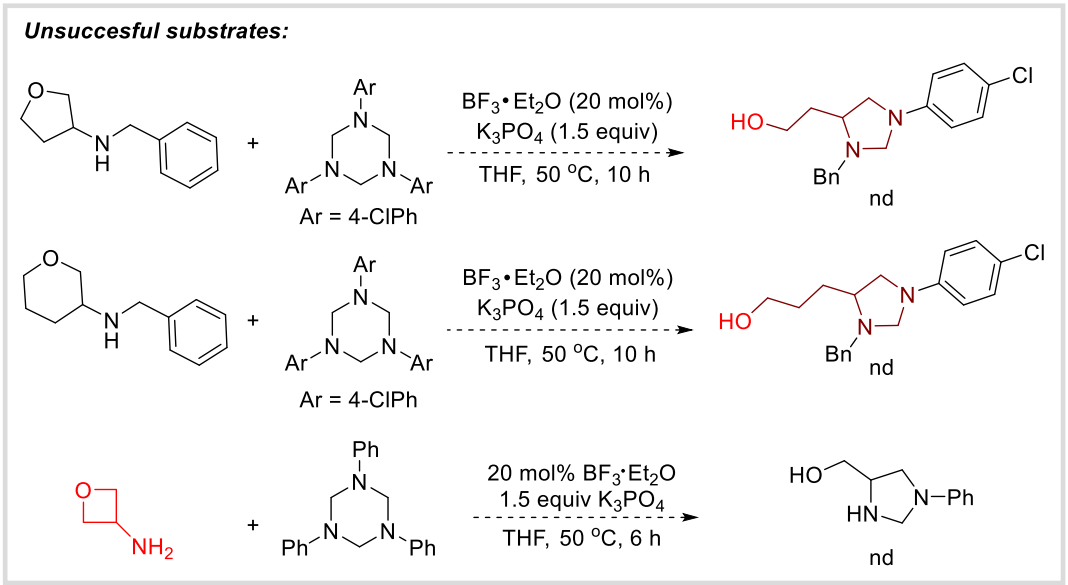
To a dried flask was added **3aa** (151.4 mg, 0.5 mmol) and anhydrous dichloromethane (5 mL) at 0 °C, 4-methylbenzenesulfonyl chloride **8** (142.9 mg, 0.75 mmol) was added followed by the addition of Et_3N (151.8 mg, 1.5 mmol) and DMAP (13.4 mg, 0.11 mmol). The reaction mixture was stirred at 0 °C for 15 minutes and then warmed to room temperature until TLC indicated the completion of the reaction. The crude product was purified by chromatography on silica gel directly to give the desired product **9** in 43% isolated yield as a yellow solid.



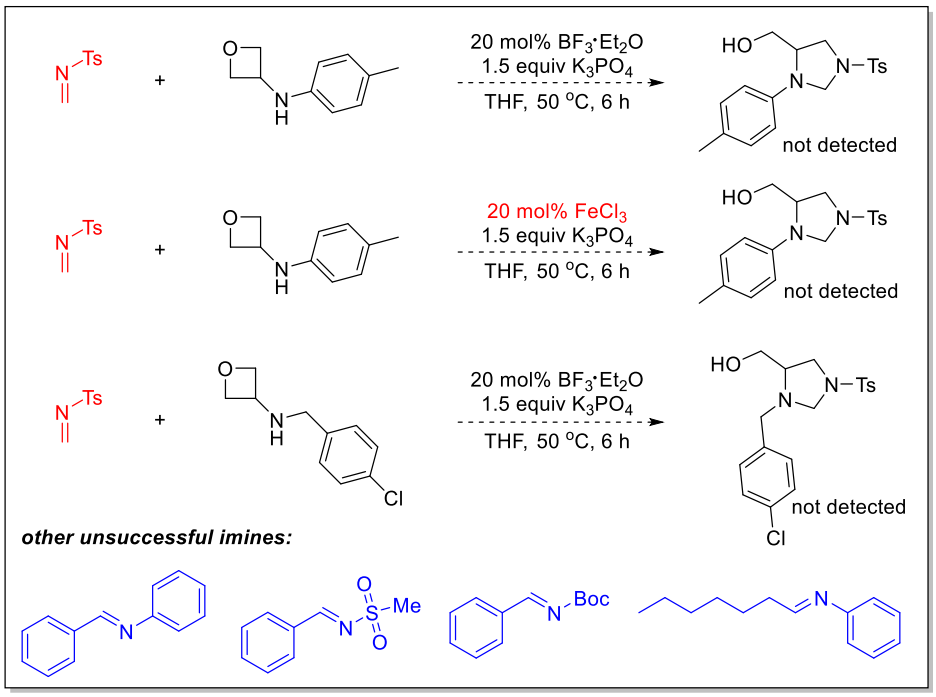
To a dried flask was added **1a** (1.0 g, 6.13 mmol), **2a** (2.05 g, 4.9 mmol), and K_3PO_4 (1.89 g, 9.2 mmol), THF (61 mL). Then, $BF_3{\cdot}Et_2O$ (20 mol%) was added to the mixture. The reaction mixture was stirred at 50 °C for 6 h. The crude product was purified by chromatography on silica gel (petroleum ether/ethyl acetate = 50:1 to 10:1) directly to give the desired product **3aa** (0.99 g) in 53% isolated yield as a white solid.

3.2 Unsuccessful substrates

Unfortunately, at current stage, five and six-membered oxygen-containing substrates such as 3-aminotetrahydrofuran and 3-aminotetrahydro-2H-pyran were found to be ineffective for this transformation due to the low strain energies.

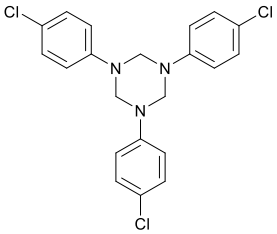


In addition, we further tested this [3+2] annulation reaction with other substituted imines. Firstly, the [3+2] reaction of N-sulfonylformaldimine with 3-aminooxetanes have been conducted under standard conditions. Other substituted imines such as N-phenyl, N-Ms and N-Boc imines have been investigated, which failed to provide the desired products.



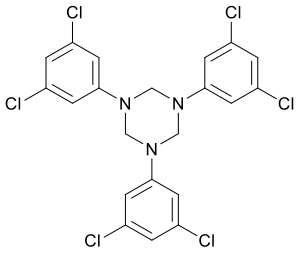
3.3 Spectral data of the products 2a-2f, 3aa-3ai, 3ba-3wa, 5aa-5ae, 7 and 9

Product 2a (known compound, CAS: 30805-14-2)



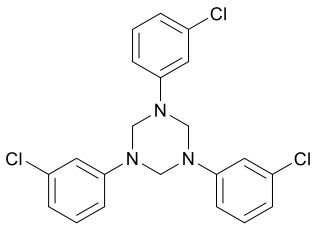
The crude product was directly washed by cooled petroleum ether, yielding **2a** in 62% yield as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.16 (d, *J* = 8.7 Hz, 6H), 6.89 (d, *J* = 8.7 Hz, 6H), 4.81 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 147.0, 129.2, 126.3, 119.0, 68.8.

Product 2b (known compound, CAS: 85680-40-6)



The crude product was directly washed by cooled petroleum ether, yielding **2b** in 70% yield as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 6.91 (s, 3H), 6.79 (s, 6H), 4.81 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 149.6, 135.7, 121.5, 115.9, 67.3.

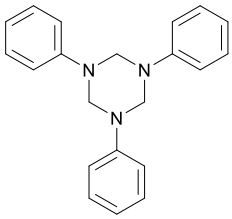
Product 2c (known compound, CAS: 109423-10-1)



The crude product was directly washed by cooled petroleum ether, yielding **2c** in 68% yield as a white solid. ^{1}H NMR (400 MHz, d^6-DMSO+CD_3COOD) δ (ppm) = 6.33 (t, *J* = 8.1 Hz, 3H), 6.27 (s, 3H), 6.19 (d, *J* = 8.2 Hz, 3H), 5.96 (d, *J* = 7.7 Hz, 3H), 4.13 (s, 6H). ^{13}C NMR (100 MHz, d^6-DMSO+CD_3COOD) δ

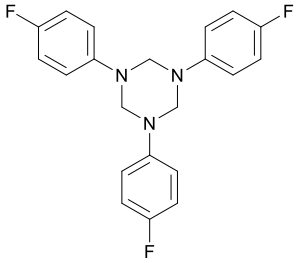
(ppm) = 150.6, 134.8, 131.5, 120.8, 117.6, 116.5, 67.3.

Product 2d (known compound, CAS: 91-78-1)



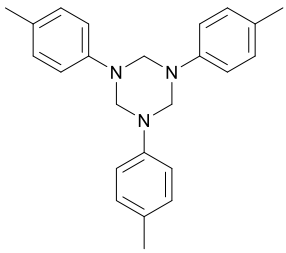
The crude product was directly washed by cooled petroleum ether, yielding **2d** in 50% yield as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.21 (t, *J* = 7.8 Hz, 6H), 7.01 (d, *J* = 8.1 Hz, 6H), 6.87 (t, *J* = 7.3 Hz, 3H), 4.89 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 148.6, 129.1, 120.9, 117.7, 68.6.

Product 2e (known compound, CAS: 109423-09-8)



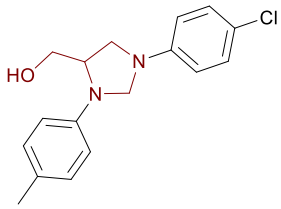
The crude product was directly washed by cooled petroleum ether, yielding **2e** in 40% yield as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.00 – 6.96 (m, 6H), 6.89 (t, *J* = 8.6 Hz, 6H), 4.76 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 157.8 (d, *J* = 239.1 Hz), 144.9 (d, *J* =2.4 Hz), 119.9 (d, *J* = 7.7 Hz), 115.7 (d, *J* = 22.1 Hz), 70.4.

Product 2f (known compound, CAS: 6639-47-0)



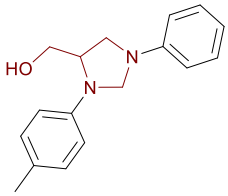
The crude product was directly washed by cooled petroleum ether, yielding **2b** in 75% yield as a white solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.03 (d, *J* = 8.2 Hz, 6H), 6.93 (d, *J* = 8.3 Hz, 6H), 4.75 (s, 6H), 2.25 (s, 9H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.3, 130.4, 129.7, 118.0, 69.5, 20.5.

Product 3aa



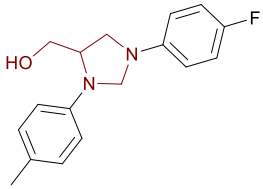
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3aa** as a white solid (79.8 mg, 88% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.24 (d, *J* = 8.7 Hz, 2H), 7.11 (d, *J* = 8.2 Hz, 2H), 6.64 (dd, *J* = 16.1, 8.6 Hz, 4H), 4.90 (d, *J* = 3.4 Hz, 1H), 4.41 (d, *J* = 3.4 Hz, 1H), 4.21 (d, *J* = 6.2 Hz, 1H), 3.78 (d, *J* = 5.6 Hz, 2H), 3.71 (dd, *J* = 8.9, 2.4 Hz, 1H), 3.49 (dd, *J* = 8.8, 7.2 Hz, 1H), 2.28 (s, 3H), 1.76 (t, *J* = 5.6 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 145.0, 143.2, 130.0, 129.1, 127.5, 123.1, 114.1, 112.8, 67.1, 62.3, 58.9, 49.7, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}ClN_2O$: 303.1259; found: 303.1244.

Product 3ab



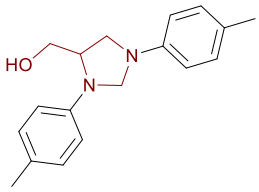
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ab** as a white solid (48.3 mg, 60% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.09 (d, *J* = 8.2 Hz, 2H), 6.83 (t, *J* = 7.3 Hz, 1H), 6.68 (dd, *J* = 21.4, 8.1 Hz, 4H), 4.92 (d, *J* = 3.4 Hz, 1H), 4.41 (d, *J* = 3.4 Hz, 1H), 4.16 (d, *J* = 6.0 Hz, 1H), 3.82 – 3.69 (m, 3H), 3.46 (t, *J* = 8.0 Hz, 1H), 2.27 (s, 3H), 1.96 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.5, 143.3, 130.0, 129.3, 127.1, 118.2, 113.1, 112.6, 67.0, 62.4, 58.8, 49.5, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{20}N_2O$: 269.1648; found: 269.1644.

Product 3ac



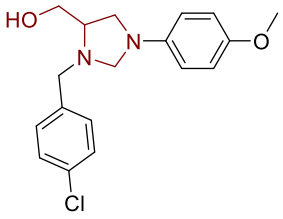
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ac** as a yellow solid (44.6 mg, 52% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.10 (d, *J* = 8.2 Hz, 2H), 7.00 (t, *J* = 8.7 Hz, 2H), 6.64 (dd, *J* = 8.8, 4.2 Hz, 4H), 4.90 (d, *J* = 3.3 Hz, 1H), 4.35 (d, *J* = 3.3 Hz, 1H), 4.16 (d, *J* = 5.8 Hz, 1H), 3.79 (s, 2H), 3.69 (dd, *J* = 8.8, 2.2 Hz, 1H), 3.46 – 3.39 (m, 1H), 2.28 (s, 3H), 1.91 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 156.3 (d, *J* = 235.2 Hz), 143.3, 144.2 (d, *J* = 1.89 Hz), 130.0, 127.2, 115.9 (d, *J* = 22.0 Hz), 114.1 (d, *J* = 7.4 Hz), 112.6, 67.7, 62.5, 58.9, 50.2, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}FN_2O$: 287.1554; found: 287.1548.

Product 3ad



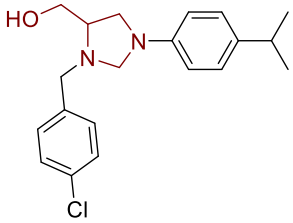
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ad** as a white solid (56.7 mg, 67% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.10 (dd, *J* = 8.2, 2.9 Hz, 4H), 6.65 (d, *J* = 7.1 Hz, 4H), 4.93 (d, *J* = 3.4 Hz, 1H), 4.35 (d, *J* = 3.4 Hz, 1H), 4.16 (d, *J* = 5.2 Hz, 1H), 3.80 (d, *J* = 5.0 Hz, 2H), 3.72 (d, *J* = 8.8 Hz, 1H), 3.42 (t, *J* = 8.0 Hz, 1H), 2.29 (s, 3H), 2.28 (s, 3H), 1.89 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 144.5, 143.5, 129.9, 129.8, 127.7, 127.0, 113.4, 112.5, 67.4, 62.7, 58.9, 50.0, 20.4, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{18}H_{22}N_2O$: 283.1805; found: 283.1797.

Product 3ae



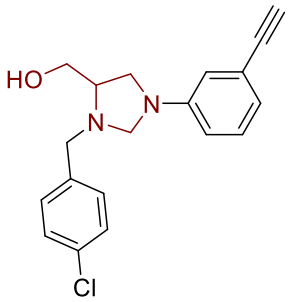
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 3:1), yielding **3ae** as a yellow solid (42.6 mg, 43% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.35 – 7.27 (m, 4H), 6.81 (d, *J* = 9.0 Hz, 2H), 6.44 (d, *J* = 9.0 Hz, 2H), 4.17 (d, *J* = 5.4 Hz, 1H), 3.98 (d, *J* = 13.3 Hz, 1H), 3.81 (d, *J* = 5.4 Hz, 1H), 3.73 (s, 3H), 3.67 (m, 1H), 3.61 (d, *J* = 13.3 Hz, 1H), 3.53 (m, 2H), 3.32 – 3.22 (m, 2H), 2.57 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 151.8, 141.1, 136.9, 133.2, 129.8, 128.7, 114.9, 113.2, 70.7, 64.1, 60.7, 57.4, 55.8, 49.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{18}H_{21}ClN_2O_2$: 333.1364; found: 333.1357.

Product 3af



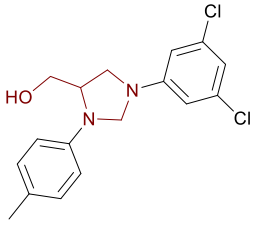
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 3:1), yielding **3af** as a yellow solid (21.8 mg, 63% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.34 – 7.26 (m, 4H), 7.08 (d, *J* = 8.5 Hz, 2H), 6.43 (d, *J* = 8.6 Hz, 2H), 4.21 (d, *J* = 5.6 Hz, 1H), 3.97 (d, *J* = 13.3 Hz, 1H), 3.85 (d, *J* = 5.6 Hz, 1H), 3.67 (m, 1H), 3.61 (d, *J* = 13.3 Hz, 1H), 3.57 – 3.51 (m, 2H), 3.33 – 3.24 (m, 2H), 2.82 (m, 1H), 2.74 (s, 1H), 1.21 (s, 3H) , 1.19 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 144.4, 137.50, 136.8, 133.2, 129.8, 128.7, 127.1, 112.1, 70.0, 64.1, 60.6, 57.4, 48.4, 33.1, 24.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{20}H_{25}ClN_2O$: 345.1728; found: 345.1726.

Product 3ag



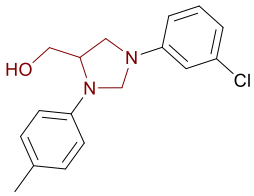
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 3:1), yielding **3ag** as a brown solid (16.2 mg, 57% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.31 (q, *J* = 8.6 Hz, 4H), 7.14 (t, *J* = 7.9 Hz, 1H), 6.85 (d, *J* = 7.6 Hz, 1H), 6.56 (s, 1H), 6.44 (dd, *J* = 8.2, 2.1 Hz, 1H), 4.22 (d, *J* = 5.6 Hz, 1H), 3.99 (d, *J* = 13.2 Hz, 1H), 3.83 (d, *J* = 5.6 Hz, 1H), 3.71 (dd, *J* = 11.4, 3.8 Hz, 1H), 3.58 (d, *J* = 2.3 Hz, 1H), 3.54 (t, *J* = 5.5 Hz, 2H), 3.33 – 3.27 (m, 2H), 3.01 (s, 1H), 2.52 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 145.8, 136.5, 133.3, 129.8, 129.2, 128.8, 122.6, 120.8, 115.2, 112.7, 84.2, 76.4, 69.7, 63.9, 60.3, 57.1, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{19}ClN_2O$: 327.1259; found: 327.1257.

Product 3ah



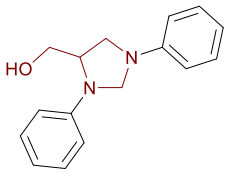
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ah** as a white solid (93.8 mg, 93% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.11 (d, *J* = 8.0 Hz, 2H), 6.77 (s, 1H), 6.65 (d, *J* = 8.0 Hz, 2H), 6.51 (s, 2H), 4.81 (d, *J* = 3.6 Hz, 1H), 4.45 (d, *J* = 3.6 Hz, 1H), 4.21 (s, 1H), 3.75 (s, 2H), 3.67 (d, *J* = 8.5 Hz, 1H), 3.53 (t, *J* = 8.0 Hz, 1H), 2.28 (s, 3H), 1.83 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 147.5, 142.8, 135.6, 130.1, 127.8, 117.5, 113.1, 110.9, 66.4, 61.8, 58.5, 58.5, 49.1, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{18}Cl_2N_2O$: 337.0869; found: 337.0862.

Product 3ai



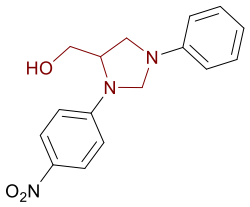
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ai** as a white solid (76.1 mg, 84% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.18 (t, *J* = 8.1 Hz, 1H), 7.10 (d, *J* = 8.2 Hz, 2H), 6.78 (d, *J* = 8.9 Hz, 1H), 6.66 (dd, *J* = 5.4, 2.9 Hz, 3H), 6.55 (dd, *J* = 8.3, 2.2 Hz, 1H), 4.87 (d, *J* = 3.5 Hz, 1H), 4.44 (d, *J* = 3.5 Hz, 1H), 4.22-4.18 (m, 1H), 3.77 (h, *J* = 5.8 Hz, 2H), 3.70 (dd, *J* = 8.9, 2.4 Hz, 1H), 3.53 – 3.48 (m, 1H), 2.28 (s, 3H), 1.81 (t, *J* = 5.5 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 147.3, 143.1, 135.1, 130.2, 130.0, 127.5, 117.8, 112.8, 111.0, 77.0, 66.7, 62.1, 58.6, 49.3, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}ClN_2O$: 303.1259; found: 303.1252.

Product 3ba



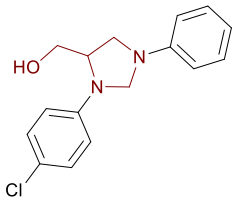
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ba** as a white solid (48.8 mg, 64% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.30 (td, *J* = 7.6, 4.6 Hz, 4H), 6.83 (dt, *J* = 14.9, 7.3 Hz, 2H), 6.73 (d, *J* = 8.1 Hz, 4H), 4.97 (d, *J* = 3.5 Hz, 1H), 4.45 (d, *J* = 3.4 Hz, 1H), 4.24 (q, *J* = 4.7 Hz, 1H), 3.87 – 3.76 (m, 3H), 3.53 – 3.45 (m, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.4, 145.3, 129.5, 129.3, 118.3, 117.7, 113.2, 112.2, 66.6, 62.4, 58.5, 49.6. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{16}H_{18}N_2O$: 255.1492; found: 255.1493.

Product 3ca



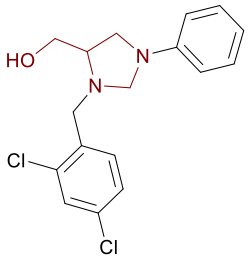
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ca** as a yellow solid (66.4 mg, 74% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.19 (d, *J* = 9.2 Hz, 2H), 7.34 (t, *J* = 7.9 Hz, 2H), 6.90 (t, *J* = 7.3 Hz, 1H), 6.75 (d, *J* = 8.0 Hz, 2H), 6.68 (d, *J* = 9.3 Hz, 2H), 5.03 (d, *J* = 4.1 Hz, 1H), 4.52 (d, *J* = 4.1 Hz, 1H), 4.38 – 4.33 (m, 1H), 3.90 (dd, *J* = 9.9, 5.0 Hz, 2H), 3.80 (dd, *J* = 10.7, 7.3 Hz, 1H), 3.51 (dd, *J* = 9.0, 6.8 Hz, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 149.4, 145.9, 138.2, 129.5, 126.4, 119.1, 113.4, 110.8, 65.8, 62.1, 58.5, 49.5. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{16}H_{17}N_3O_3$: 300.1343; found: 300.1350.

Product 3da



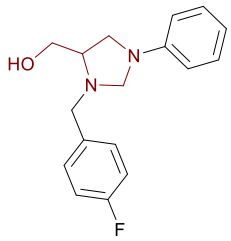
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3da** as a white solid (54.5 mg, 63% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.31 (t, *J* = 7.9 Hz, 2H), 7.23 (d, *J* = 8.9 Hz, 2H), 6.86 (t, *J* = 7.3 Hz, 1H), 6.72 (d, *J* = 8.1 Hz, 2H), 6.64 (d, *J* = 8.9 Hz, 2H), 4.92 (d, *J* = 3.4 Hz, 1H), 4.40 (d, *J* = 3.4 Hz, 1H), 4.18 (q, *J* = 4.9 Hz, 1H), 3.79 (dd, *J* = 12.0, 6.1 Hz, 3H), 3.48 (dd, *J* = 8.7, 7.2 Hz, 1H), 1.85 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.3, 143.8, 129.4, 129.3, 122.5, 118.5, 113.3, 113.2, 66.6, 62.4, 58.6, 49.6. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{16}H_{17}ClN_2O$: 289.1102; found: 289.1081.

Product 3ea



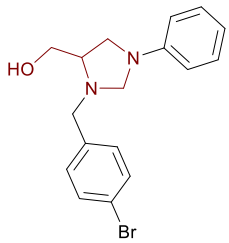
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ea** as a yellow solid (64.5 mg, 64% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.47 – 7.37 (m, 2H), 7.28 – 7.20 (m, 3H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.49 (d, *J* = 8.1 Hz, 2H), 4.24 (d, *J* = 5.5 Hz, 1H), 4.07 (d, *J* = 13.6 Hz, 1H), 3.92 (d, *J* = 5.5 Hz, 1H), 3.78 – 3.70 (m, 2H), 3.57 (q, *J* = 9.7, 8.0 Hz, 2H), 3.39 – 3.30 (m, 2H), 2.61 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.1, 134.8, 134.3, 134.0, 131.3, 129.7, 129.2, 127.3, 117.0, 112.1, 69.8, 64.3, 60.4, 54.8, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{18}Cl_2N_2O$: 337.0869; found: 337.0875.

Product 3fa



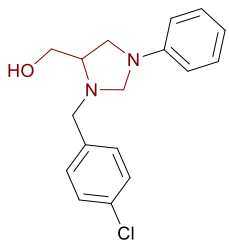
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3fa** as a yellow oil (53.2 mg, 62% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.32 (dd, *J* = 8.3, 5.6 Hz, 2H), 7.20 (t, *J* = 7.9 Hz, 2H), 7.03 (t, *J* = 8.6 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.46 (d, *J* = 8.1 Hz, 2H), 4.22 (d, *J* = 5.6 Hz, 1H), 3.97 (d, *J* = 13.1 Hz, 1H), 3.86 (d, *J* = 5.6 Hz, 1H), 3.68 (dd, *J* = 11.3, 3.7 Hz, 1H), 3.57 (dd, *J* = 12.6, 8.2 Hz, 3H), 3.30 (d, *J* = 4.9 Hz, 2H), 2.53 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 162.1 (d, *J* = 246.6 Hz), 146.1, 133.9 (d, *J* = 3.2 Hz), 130.0 (d, *J* = 8.0 Hz), 129.2, 116.9, 115.4 (d, *J* = 21.4 Hz), 111.9, 69.7, 63.9, 60.4, 57.1, 48.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}FN_2O$: 287.1554; found: 287.1548.

Product 3ga



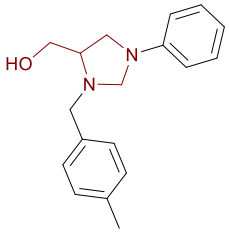
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ga** as a yellow solid (78.9 mg, 76% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.49 – 7.44 (m, 2H), 7.21 (dd, *J* = 15.5, 8.1 Hz, 4H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.45 (d, *J* = 8.0 Hz, 2H), 4.22 (d, *J* = 5.5 Hz, 1H), 3.96 (d, *J* = 13.3 Hz, 1H), 3.84 (d, *J* = 5.6 Hz, 1H), 3.68 (dd, *J* = 11.4, 3.7 Hz, 1H), 3.59 – 3.50 (m, 3H), 3.33 – 3.25 (m, 2H), 2.50 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.1, 137.3, 131.7, 130.1, 129.2, 121.3, 117.0, 112.0, 69.9, 64.0, 60.6, 57.3, 48.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}BrN_2O$: 347.0754; found: 347.0760.

Product 3ha



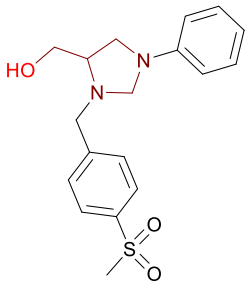
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ha** as a yellow solid (45.3 mg, 50% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.32 (d, *J* = 2.8 Hz, 4H), 7.24 – 7.19 (m, 2H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.47 (d, *J* = 8.0 Hz, 2H), 4.24 (d, *J* = 5.6 Hz, 1H), 3.99 (d, *J* = 13.3 Hz, 1H), 3.87 (d, *J* = 5.6 Hz, 1H), 3.72 (d, *J* = 3.7 Hz, 1H), 3.62 – 3.53 (m, 3H), 3.33 (t, *J* = 5.2 Hz, 2H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.1, 136.7, 133.3, 129.8, 129.2, 128.8, 117.0, 112.0, 69.8, 64.0, 60.4, 57.3, 48.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}ClN_2O$: 303.1259; found: 303.1256.

Product 3ia



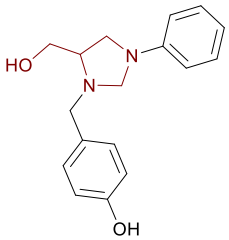
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ia** as a yellow oil (49.9 mg, 59% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.27 – 7.12 (m, 6H), 6.70 (t, *J* = 7.3 Hz, 1H), 6.45 (d, *J* = 8.0 Hz, 2H), 4.24 (d, *J* = 5.5 Hz, 1H), 3.96 (d, *J* = 13.0 Hz, 1H), 3.88 (d, *J* = 5.5 Hz, 1H), 3.69 (dd, *J* = 11.3, 2.9 Hz, 1H), 3.61 – 3.49 (m, 3H), 3.30 (q, *J* = 8.8, 7.5 Hz, 2H), 2.35 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2, 137.2, 135.0, 129.2, 129.1, 128.5, 116.7, 111.9, 69.8, 63.8, 60.2, 57.5, 48.2, 21.1. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{18}H_{22}N_2O$: 283.1805; found: 283.1804.

Product 3ja



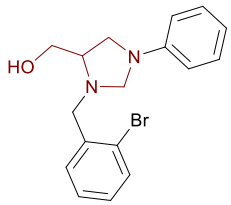
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **3ja** as a yellow solid (59.2 mg, 57% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.91 (d, *J* = 8.1 Hz, 2H), 7.59 (d, *J* = 8.1 Hz, 2H), 7.21 (t, *J* = 7.8 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.47 (d, *J* = 8.2 Hz, 2H), 4.26 (d, *J* = 5.5 Hz, 1H), 4.12 (d, *J* = 14.0 Hz, 1H), 3.85 (d, *J* = 5.5 Hz, 1H), 3.76 – 3.68 (m, 2H), 3.63 – 3.56 (m, 2H), 3.37 – 3.29 (m, 2H), 3.06 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) =146.0, 144.9, 139.5, 129.2 (overlap), 127.6, 117.0, 112.0, 70.0, 64.2, 60.9, 57.5, 48.2, 44.4. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{18}H_{23}N_2O_3S$: 347.1424; found: 347.1418.

Product 3ka



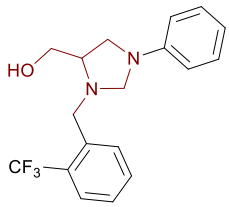
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **3ka** as a white solid (44.3 mg, 52% yield). ^{1}H NMR (400 MHz, DMSO) δ (ppm) = 8.16 (s, 1H), 5.99 (dd, *J* = 18.1, 8.1 Hz, 4H), 5.56 (d, *J* = 8.4 Hz, 2H), 5.45 (t, *J* = 7.3 Hz, 1H), 5.26 (d, *J* = 8.0 Hz, 2H), 3.56 (t, *J* = 5.3 Hz, 1H), 2.90 (d, *J* = 5.3 Hz, 1H), 2.77 (d, *J* = 13.0 Hz, 1H), 2.58 (d, *J* = 5.3 Hz, 1H), 2.43 (dd, *J* = 10.5, 5.2 Hz, 1H), 2.33 – 2.27 (m, 2H), 2.02 – 1.89 (m, 2H), 1.35 (s, 1H). ^{13}C NMR (100 MHz, DMSO) δ (ppm) = 156.4, 146.4, 129.7, 129.0, 129.0, 115.9, 115.0, 111.6, 70.2, 63.7, 62.3, 56.8, 49.5. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{17}H_{20}N_2O_2$: 285.1598; found: 285.1593.

Product 3la



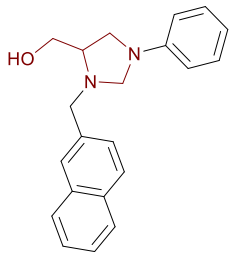
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3la** as a yellow oil (92.4 mg, 89% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.57 (d, *J* = 7.9 Hz, 1H), 7.42 (d, *J* = 7.3 Hz, 1H), 7.30 (t, *J* = 7.4 Hz, 1H), 7.18 (dt, *J* = 17.1, 7.6 Hz, 3H), 6.71 (t, *J* = 7.3 Hz, 1H), 6.47 (d, *J* = 8.2 Hz, 2H), 4.22 (d, *J* = 5.6 Hz, 1H), 4.08 (d, *J* = 13.4 Hz, 1H), 3.92 (d, *J* = 5.6 Hz, 1H), 3.76 – 3.70 (m, 2H), 3.59 – 3.50 (m, 2H), 3.33 (t, *J* = 5.1 Hz, 2H), 2.74 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.1, 137.2, 133.1, 130.7, 129.1, 127.5, 124.5, 116.8, 112.0, 69.7, 64.2, 60.3, 57.7, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}BrN_2O$: 347.0754; found: 347.0750.

Product 3ma



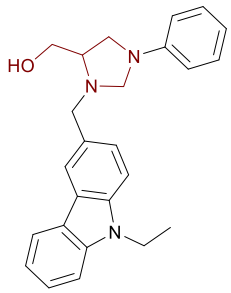
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ma** as a yellow oil (62.5 mg, 62% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.76 (d, *J* = 7.7 Hz, 1H), 7.67 (d, *J* = 7.8 Hz, 1H), 7.55 (t, *J* = 7.6 Hz, 1H), 7.39 (t, *J* = 7.6 Hz, 1H), 7.21 (t, *J* = 7.8 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.48 (d, *J* = 8.1 Hz, 2H), 4.31 (d, *J* = 5.5 Hz, 1H), 4.18 (d, *J* = 14.5 Hz, 1H), 3.93 – 3.84 (m, 2H), 3.69 (dd, *J* = 11.5, 3.6 Hz, 1H), 3.57 (dd, *J* = 11.0, 4.7 Hz, 2H), 3.35 (q, *J* = 5.5, 4.7 Hz, 2H), 2.46 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2, 137.1, 132.1, 130.2, 129.2, 128.3 (d, *J* = 30.0 Hz), 127.4, 126.1 (d, *J* = 5.8 Hz), 124.4 (d, *J* = 275.0 Hz), 117.0, 112.1, 70.1, 64.4, 60.6, 54.1, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{18}H_{19}F_3N_2O$: 337.1522; found: 337.1519.

Product 3na



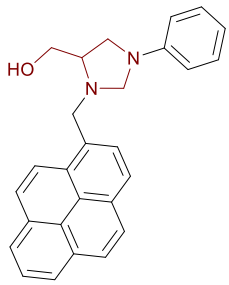
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3na** as a yellow solid (62.0 mg, 65% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.23 (d, *J* = 8.3 Hz, 1H), 7.88 (d, *J* = 7.9 Hz, 1H), 7.83 (d, *J* = 7.9 Hz, 1H), 7.58-7.42 (m, 4H), 7.26 – 7.18 (m, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.47 (d, *J* = 8.1 Hz, 2H), 4.44 (d, *J* = 12.9 Hz, 1H), 4.20 (d, *J* = 6.0 Hz, 1H), 4.14 – 4.02 (m, 2H), 3.72 – 3.60 (m, 2H), 3.53 (dd, *J* = 11.1, 5.3 Hz, 1H), 3.45 (dt, *J* = 12.1, 5.3 Hz, 1H), 3.28 (dd, *J* = 8.7, 5.6 Hz, 1H), 2.51 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.3, 133.9, 133.8, 131.9, 129.2, 128.8, 128.6, 127.1, 126.4, 125.9, 125.2, 123.7, 116.9, 112.1, 69.7, 64.5, 60.8, 56.6, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{21}H_{22}N_2O$: 319.1805; found: 319.1800.

Product 3oa



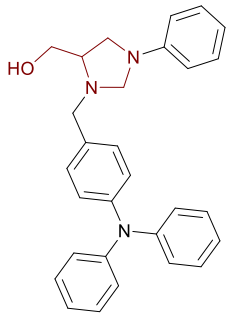
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **3oa** as a yellow oil (53.2 mg, 46% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.17 – 8.00 (m, 2H), 7.47 (t, *J* = 6.9 Hz, 2H), 7.44 – 7.36 (m, 2H), 7.24 (t, *J* = 3.6 Hz, 1H), 7.22 – 7.16 (m, 2H), 6.70 (t, *J* = 7.3 Hz, 1H), 6.46 (d, *J* = 8.3 Hz, 2H), 4.37 (q, *J* = 7.2 Hz, 2H), 4.28 (d, *J* = 5.7 Hz, 1H), 4.21 (d, *J* = 12.7 Hz, 1H), 3.99 (d, *J* = 5.7 Hz, 1H), 3.83 – 3.72 (m, 2H), 3.58 (q, *J* = 5.7, 4.4 Hz, 2H), 3.43 – 3.28 (m, 2H), 1.44 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.3, 140.2, 139.4, 129.2, 128.3, 126.4, 125.8, 122.9, 122.6, 120.5, 120.4, 118.8, 116.7, 112.0, 108.6, 108.5, 69.7, 63.9, 60.2, 58.3, 48.3, 37.6, 13.8. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{25}H_{27}N_3O$: 386.2227; found: 386.2221.

Product 3pa



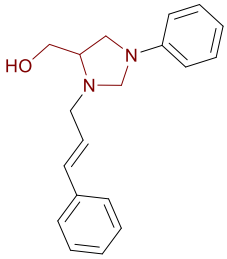
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 30:1 to 10:1), yielding **3pa** as a yellow solid (67.1 mg, 57% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.44 (d, *J* = 9.2 Hz, 1H), 8.21 – 8.10 (m, 4H), 8.08 – 7.93 (m, 4H), 7.18 (t, *J* = 7.8 Hz, 2H), 6.70 (t, *J* = 7.3 Hz, 1H), 6.44 (d, *J* = 8.0 Hz, 2H), 4.63 (d, *J* = 12.8 Hz, 1H), 4.33 (d, *J* = 12.9 Hz, 1H), 4.17 (d, *J* = 5.9 Hz, 1H), 4.08 (d, *J* = 6.0 Hz, 1H), 3.63 (t, *J* = 7.9 Hz, 2H), 3.57 – 3.42 (m, 2H), 3.28 (dd, *J* = 8.7, 5.5 Hz, 1H) , 2.58 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2, 131.3, 131.2, 131.2, 130.7, 129.5, 129.2, 128.0, 127.7, 127.5, 127.3, 126.0, 125.3, 125.3, 125.1, 124.7, 124.6, 123.0, 116.9, 112.1, 69.6, 64.5, 60.9, 56.5, 48.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{27}H_{24}N_2O$: 393.1961; found: 393.1955.

Product 3qa



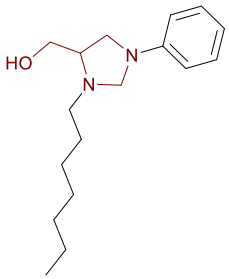
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **3qa** as a yellow solid (79.6 mg, 61% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.26 – 7.19 (m, 8H), 7.11 – 6.99 (m, 9H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.50 (d, *J* = 7.9 Hz, 2H), 4.31 (d, *J* = 5.6 Hz, 1H), 3.99 – 3.91 (m, 2H), 3.72 (dd, *J* = 11.3, 3.5 Hz, 1H), 3.61 – 3.53 (m, 3H), 3.32 (q, *J* = 6.3 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ (ppm) = 147.7, 147.2, 146.2, 132.0, 129.4, 129.2, 129.2, 124.3, 123.7, 122.8, 116.8, 112.0, 69.8, 63.8, 60.2, 57.4, 48.2. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{29}N_3O$: 436.2383; found: 436.2578.

Product 3ra



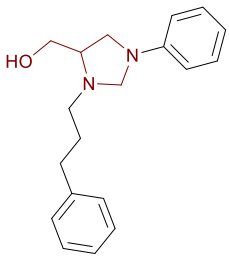
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ra** as a white solid (37.1 mg, 42% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.39 (d, *J* = 7.5 Hz, 2H), 7.33 (t, *J* = 7.5 Hz, 2H), 7.27 – 7.19 (m, 3H), 6.74 (t, *J* = 7.3 Hz, 1H), 6.59 (d, *J* = 15.9 Hz, 1H), 6.52 (d, *J* = 8.1 Hz, 2H), 6.35-6.27 (m, 1H), 4.39 (d, *J* = 5.8 Hz, 1H), 4.01 (d, *J* = 5.8 Hz, 1H), 3.72 (dd, *J* = 11.3, 4.1 Hz, 1H), 3.64 (dd, *J* = 13.6, 5.4 Hz, 1H), 3.55 (dd, *J* = 9.8, 6.1 Hz, 2H), 3.37 – 3.23 (m, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2, 136.5, 133.0, 129.2, 128.6, 127.8, 126.4, 126.1, 116.9, 112.1, 69.5, 63.8, 60.4, 56.3, 48.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{22}N_2O$: 295.1805; found: 295.1803.

Product 3sa



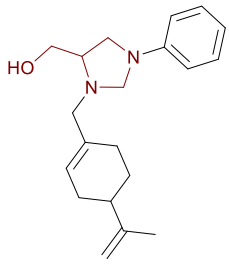
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3sa** as a yellow oil (43.9 mg, 53% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.27 – 7.21 (m, 2H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.52 (d, *J* = 7.9 Hz, 2H), 4.45 (d, *J* = 5.0 Hz, 1H), 3.85 (d, *J* = 5.0 Hz, 1H), 3.70 (dd, *J* = 11.2, 3.9 Hz, 1H), 3.53 – 3.44 (m, 2H), 3.35 – 3.29 (m, 1H), 3.14 (tt, *J* = 7.4, 3.9 Hz, 1H), 2.77 (dt, *J* = 11.9, 8.0 Hz, 1H), 2.50 – 2.43 (m, 1H), 1.56 – 1.51 (m, 2H), 1.35 – 1,29 (m, 8H), 0.89 (t, *J* = 6.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2, 129.2, 116.7, 111.9, 70.2, 64.1, 59.7, 53.6, 48.1, 31.8, 29.1, 29.0, 27.2, 22.6, 14.1. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{28}N_2O$: 277.2274; found: 277.2271.

Product 3ta



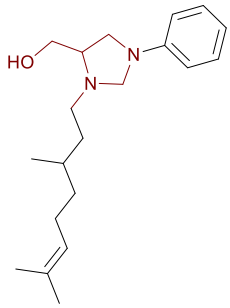
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ta** as a yellow solid (48.9 mg, 55% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.32 – 7.27 (m, 2H), 7.25 – 7.17 (m, 5H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.51 (d, *J* = 8.1 Hz, 2H), 4.45 (d, *J* = 5.1 Hz, 1H), 3.85 (d, *J* = 5.0 Hz, 1H), 3.66 (dd, *J* = 11.2, 4.0 Hz, 1H), 3.52 – 3.43 (m, 2H), 3.34 – 3.27 (m, 1H), 3.14 (tt, *J* = 7.4, 4.0 Hz, 1H), 2.85 – 2.59 (m, 4H), 2.54 – 2.48 (m, 1H), 1.92 – 1.84 (m, *J* = 2H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) =146.1, 141.5, 129.2, 128.4, 128.3, 125.9, 116.8, 111.9, 70.2, 64.2, 59.9, 53.1, 48.1, 33.3, 30.5. HRMS (EI): m/z $[M + H]^+$ calcd for $C_{19}H_{24}N_2O$: 297.1961; found: 297.1958.

Product 3ua



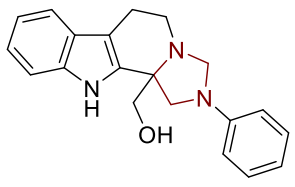
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3ua** as a colorless oil (63.7 mg, dr = 1:1, 68% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.23 (t, *J* = 7.9 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.51 (dd, *J* = 8.1, 3.7 Hz, 2H), 5.68 (s, 1H), 4.73 (s, 2H), 4.28 (dd, *J* = 15.6, 5.6 Hz, 1H), 3.83 (dd, *J* = 13.8, 5.6 Hz, 1H), 3.68 (ddd, *J* = 11.0, 6.8, 4.1 Hz, 1H), 3.53 – 3.46 (m, 2H), 3.28 (t, *J* = 10.5 Hz, 2H), 3.22 – 3.16 (m, 1H), 2.97 (t, *J* = 12.0 Hz, 1H), 2.65 (s, 1H), 2.23 – 2.15 (m, 3H), 2.03 – 2.01 (m, 2H), 1.85 (d, *J* = 12.2 Hz, 1H), 1.75 (s, 3H), 1.55 – 1.45 (m, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 149.6, 149.5, 146.2, 146.2, 134.8 (overlap), 129.1 (overlap), 124.6, 124.5, 116.7, 116.7, 111.9, 111.9, 108.7 (overlap), 69.6 (overlap), 63.7, 63.6, 60.7, 60.2, 60.0, 60.0, 48.1, 48.0, 41.0, 40.9, 30.5, 30.5, 27.5, 27.4 (overlap), 27.0, 20.8, 20.7. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{20}H_{28}N_2O$: 313.2274; found: 313.2269.

Product 3va



The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3va** as a yellow solid (47.4 mg, dr = 1:1, 50% yield). 1H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.24 (t, *J* = 7.8 Hz, 2H), 6.73 (t, *J* = 7.3 Hz, 1H), 6.52 (d, *J* = 8.0 Hz, 2H), 5.10 (t, *J* = 7.3 Hz, 1H), 4.44 (d, *J* = 5.1 Hz, 1H), 3.85 (dd, *J* = 7.3, 5.1 Hz, 1H), 3.70 (dt, *J* = 11.0, 4.5 Hz, 1H), 3.54 – 3.43 (m, 2H), 3.34 – 3.27 (m, 1H), 3.18 – 3.14 (m, 1H), 2.87 – 2.78 (m, 1H), 2.51 – 2.41 (m, 1H), 2.06 – 1.92 (m, 2H), 1.69 (s, 3H), 1.61 (s, 3H), 1.42 – 1.14 (m, 5H), 0.93 – 0.91 (m, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.2 (overlap), 131.3, 131.3, 129.2 (overlap), 124.6, 124.6, 116.7 (overlap), 111.9, 111.8, 70.3, 70.2, 64.1 (overlap), 59.9, 59.7, 51.8, 51.5, 48.2, 48.1, 37.4, 36.7, 36.1, 35.9, 30.6, 30.4, 25.7 (overlap), 25.5, 25.3, 19.8, 19.4, 17.6 (overlap). HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{20}H_{32}N_2O$: 317.2587; found: 317.2583.

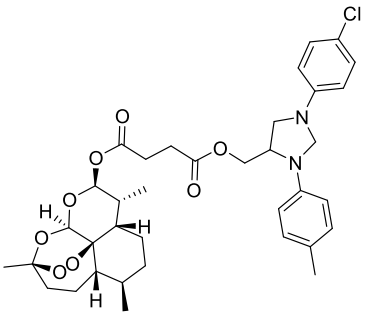
Product 3wa



The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 50:1 to 10:1), yielding **3wa** as a yellow solid (51.8 mg, 20% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.13 (s, 1H), 7.49 (d, *J* = 7.7 Hz, 1H), 7.30 (d, *J* = 8.0 Hz, 1H), 7.24 – 7.08 (m, 4H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.48 (d, *J* = 7.9 Hz, 2H), 4.47

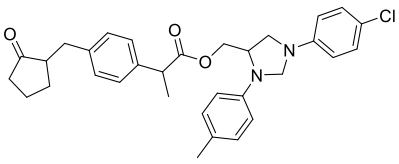
(d, *J* = 5.0 Hz, 1H), 4.28 (d, *J* = 5.0 Hz, 1H), 3.84 – 3.66 (m, 3H), 3.55 (d, *J* = 8.8 Hz, 1H), 3.30 – 3.16 (m, 3H), 3.03-2.95 (m, 1H), 2.73 (m, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 146.1, 136.1, 133.5, 129.2, 126.6, 122.2, 119.6, 118.4, 116.9, 111.9, 111.0, 108.9, 67.8, 64.5, 64.2, 52.7, 43.5, 17.8. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{20}H_{21}N_3O$: 320.1757; found: 320.1759.

Product 5aa



The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **5aa** as a white solid (104.5 mg, dr = 1:1, 92% yield). solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.30 – 7.19 (m, 2H), 7.10 (d, *J* = 8.3 Hz, 2H), 6.72 – 6.54 (m, 4H), 5.78 (dd, *J* = 9.9, 1.8 Hz, 1H), 5.40 (d, *J* = 16.4 Hz, 1H), 4.84 (t, *J* = 3.0 Hz, 1H), 4.53 – 4.45 (m, 1H), 4.37 (t, *J* = 2.6 Hz, 1H), 4.30 (s, 1H), 4.04 – 3.83 (m, 1H), 3.67 (dd, *J* = 9.0, 3.5 Hz, 1H), 3.45 – 3.40 (m, 1H), 2.72 (s, 2H), 2.70 – 2.51 (m, 3H), 2.40 – 2.33 (m, 1H), 2.27 (s, 3H), 2.02 (d, *J* = 14.7 Hz, 1H), 1.94 – 1.83 (m, 1H), 1.79 – 1.68 (m, 2H), 1.61 (d, *J* = 13.7 Hz, 1H), 1.39 – 1.19 (m, 4H), 0.98 – 0.92 (m, 3H), 0.87 – 0.83 (m, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 171.9, 171.9, 171.0, 171.0, 145.0, 145.0, 142.2, 142.2, 130.0 (overlap), 129.1, 129.0, 126.9 (overlap), 122.9 (overlap), 114.1, 114.0, 112.1 (overlap), 104.4, 104.4, 92.2, 91.4, 91.4, 80.0, 80.0, 65.8, 65.8, 63.1 (overlap), 55.4, 55.4, 51.4, 49.8, 49.7, 45.1 (overlap), 41.3, 37.2, 37.1, 36.1, 36.0, 34.0, 31.7, 29.1, 29.1, 29.0, 28.8, 28.8, 28.8, 27.6, 25.9, 24.5, 22.6, 21.9, 20.4, 20.3, 20.1, 19.4, 18.7, 14.3, 12.0, 11.4. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{37}H_{47}ClN_2O_8$: 691.2754; found: 691.2742.

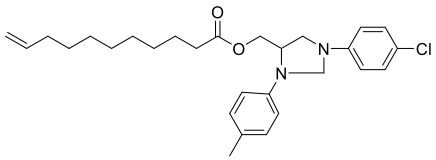
Product 5ab



The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **5ab** as a white solid (88.3 mg, dr = 2:1, 98% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.26 – 7.14 (m, 4H), 7.14 – 7.01 (m, 4H), 6.61 (t, *J* = 7.5 Hz, 2H), 6.55 – 6.45 (m, 2H), 4.74 (dd, *J* = 13.2, 2.9 Hz, 1H), 4.54 – 4.39 (m, 1H), 4.33 (d, *J* = 2.8 Hz, 1H), 4.29 – 4.15 (m, 1H), 3.97 – 3.80 (m, 1H), 3.68 – 3.66 (m, 1H), 3.50 – 3.34 (m, 1H), 3.34 – 3.16 (m, 1H), 3.14 – 3.10 (m, 1H), 2.51 – 2.48 (m, 1H), 2.36 – 2.30 (m, 2H), 2.26 (s, 3H), 2.14 – 2.03 (m, 2H), 1.97 – 1.93 (m, 1H), 1.77 – 1.68 (m, 1H), 1.58 – 1.44 (m, 4H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 220.0, 220.0, 174.5, 174.4, 144.8 (overlap), 142.2, 142.2, 139.0, 139.0, 138.0, 138.0, 130.0, 130.0, 129.1, 129.1,

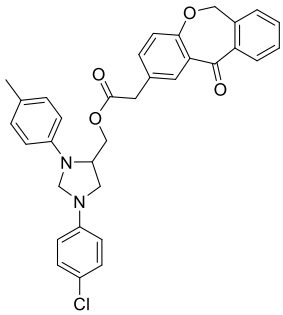
129.0 (overlap), 127.5, 127.4, 127.0, 126.9, 122.9 (overlap), 113.8 (overlap), 112.1, 112.1, 77.3, 76.7, 65.9, 65.8, 63.3, 63.1, 55.5, 55.2, 50.9, 50.8, 49.5, 49.4, 45.0, 44.9, 38.1 (overlap), 35.1 (overlap), 29.2 (overlap), 20.5, 20.3, 18.2, 18.2, 18.0. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{32}H_{35}ClN_2O_3$: 553.2228; found: 533.2215.

Product 5ac



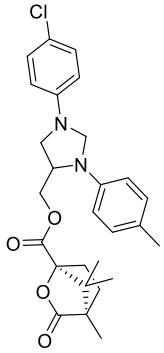
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **5ac** as a white solid (70.8 mg, 89% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.27 – 7.20 (m, 2H), 7.11 (d, *J* = 8.2 Hz, 2H), 6.63 (dd, *J* = 22.9, 8.7 Hz, 4H), 5.86 – 5.76 (mf, 1H), 5.04 – 4.89 (m, 2H), 4.84 (d, *J* = 3.1 Hz, 1H), 4.50 (dd, *J* = 11.0, 3.6 Hz, 1H), 4.40 (d, *J* = 3.1 Hz, 1H), 4.32 – 4.27 (m, 1H), 3.90 (dd, *J* = 11.0, 8.6 Hz, 1H), 3.64 (d, *J* = 8.2 Hz, 1H), 3.43 (dd, *J* = 8.7, 6.9 Hz, 1H), 2.29 (d, *J* = 9.4 Hz, 5H), 2.04 (q, *J* = 6.9 Hz, 2H), 1.67 – 1.58 (m, 3H), 1.28 – 1.25 (m, 9H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 173.7, 145.0, 142.3, 139.1, 130.1, 129.1, 127.0, 123.0, 114.2, 113.9, 112.2, 65.9, 62.9, 55.6, 49.9, 34.2, 33.8, 29.3, 29.2, 29.1, 29.0, 28.9, 24.8, 20.3. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{28}H_{37}ClN_2O_2$: 469.2616; found: 469.2604.

Product 5ad



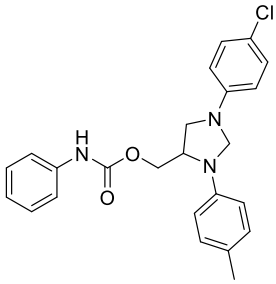
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **5ad** as a white solid (89.2 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 8.12 (d, *J* = 2.3 Hz, 1H), 7.89 (d, *J* = 7.7 Hz, 1H), 7.56 (t, *J* = 7.4 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.37 – 7.33 (m, 2H), 7.19 (d, *J* = 8.8 Hz, 2H), 7.09 (d, *J* = 8.3 Hz, 2H), 7.00 (d, *J* = 8.4 Hz, 1H), 6.63 (d, *J* = 8.5 Hz, 2H), 6.58 (d, *J* = 8.9 Hz, 2H), 5.16 (s, 2H), 4.81 (d, *J* = 3.2 Hz, 1H), 4.53 (dd, *J* = 11.0, 3.8 Hz, 1H), 4.35 (d, *J* = 3.2 Hz, 1H), 4.31 – 4.27 (m, 1H), 3.93 (dd, *J* = 11.0, 8.5 Hz, 1H), 3.61 (d, *J* = 15.8 Hz, 3H), 3.42 – 3.34 (m, 1H), 2.26 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 190.7, 171.3, 160.5, 144.9, 142.2, 140.2, 136.3, 135.5, 132.8, 132.4, 130.0, 129.4, 129.2, 129.1, 127.8, 127.3, 127.0, 125.0, 122.9, 121.1, 114.0, 112.1, 73.5, 65.9, 63.4, 55.4, 49.7, 40.0, 20.3. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{33}H_{29}ClN_2O_4$: 575.1708; found: 575.1702.

Product 5ae



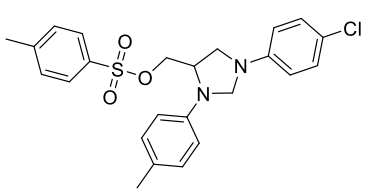
The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 5:1), yielding **5ae** as a white solid (78.7 mg, 96% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.28 – 7.21 (m, 3H), 7.12 (d, *J* = 8.0 Hz, 2H), 6.66 (dd, *J* = 8.4, 3.9 Hz, 2H), 6.61 (dd, *J* = 8.8, 2.0 Hz, 2H), 4.89 (d, *J* = 3.0 Hz, 1H), 4.63 (dd, *J* = 10.9, 2.9 Hz, 1H), 4.41 – 4.31 (m, 2H), 4.11 – 4.05 (m, 1H), 3.67 (d, *J* = 9.0 Hz, 1H), 3.49 – 3.42 (m, 1H), 2.44 – 2.35 (m, 1H), 2.28 (s, 3H), 2.05 – 1.88 (m, 2H), 1.73 – 1.61 (m, 1H), 1.12 (s, 3H), 1.04 (d, *J* = 14.5 Hz, 3H), 0.94 (d, *J* = 10.0 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 178.0, 167.6, 144.9, 142.1, 130.2, 129.2, 127.3, 123.3, 114.1, 112.2, 90.9, 66.0, 64.1, 55.5, 54.8, 54.3, 49.9, 30.7, 28.9, 20.3, 16.7, 16.7, 9.7. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{27}H_{31}ClN_2O_4$: 483.2045; found: 483.2027.

Product 7



The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 20:1 to 3:1), yielding **7** as a white solid (33.6 mg, 57% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.32 (q, *J* = 8.5, 8.1 Hz, 4H), 7.23 (d, *J* = 8.8 Hz, 2H), 7.10 (t, *J* = 8.1 Hz, 3H), 6.70 (d, *J* = 8.4 Hz, 2H), 6.66 – 6.58 (m, 3H), 4.87 (d, *J* = 3.0 Hz, 1H), 4.55 (dd, *J* = 11.0, 3.7 Hz, 1H), 4.39 (d, *J* = 3.1 Hz, 2H), 3.99 (t, *J* = 9.7 Hz, 1H), 3.72 (d, *J* = 9.0 Hz, 1H), 3.47 – 3.40 (m, 1H), 2.27 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 145.0, 142.4, 137.4, 130.1, 129.1, 129.1, 127.1, 123.7, 123.1, 118.8, 114.1, 112.2, 66.0, 63.8, 55.9, 49.8, 20.3. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{24}H_{24}ClN_3O_2$: 444.1449; found: 444.1446.

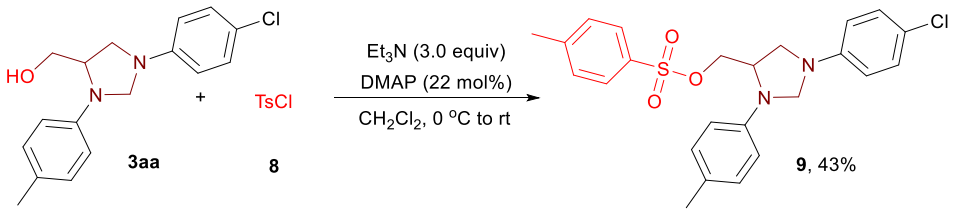
Product 9

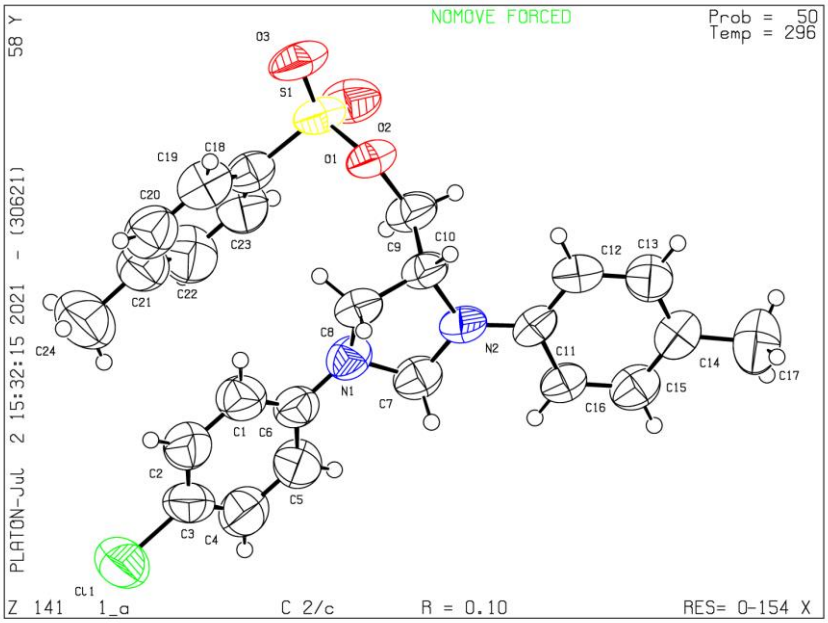


The crude product was purified by column chromatography (SiO_2, petroleum ether/ethyl acetate = 30:1 to 15:1), yielding **9** as a yellow solid (98.1 mg, 43% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm) = 7.77 (d, *J* = 8.2 Hz, 2H), 7.31 (d, *J* = 8.1 Hz, 2H), 7.22 (d, *J* = 8.8 Hz, 2H), 7.07 (d, *J* = 8.3 Hz, 2H), 6.55 (d, *J* = 8.8 Hz, 2H), 6.48 (d, *J* = 8.4 Hz, 2H), 4.76 (d, *J* = 3.0 Hz, 1H), 4.29 (d, *J* = 3.1 Hz, 2H), 4.21 (dd, *J* = 10.1, 3.1 Hz, 1H), 3.88 (t, *J* = 9.8 Hz, 1H), 3.66 (d, *J* = 9.3 Hz, 1H), 3.42 – 3.34 (m, 1H), 2.43 (s, 3H), 2.27 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm) = 145.1, 144.8, 141.8, 132.7, 130.2,

130.0, 129.1, 127.9, 127.4, 123.5, 114.3, 111.9, 67.6, 65.9, 55.9, 49.7, 21.6, 20.3. HRMS (ESI): m/z [M + Na]$^+$ calcd for $C_{24}H_{25}ClN_2O_3S$: 479.1167; found: 479.1151.

4. X-Ray structure of 9





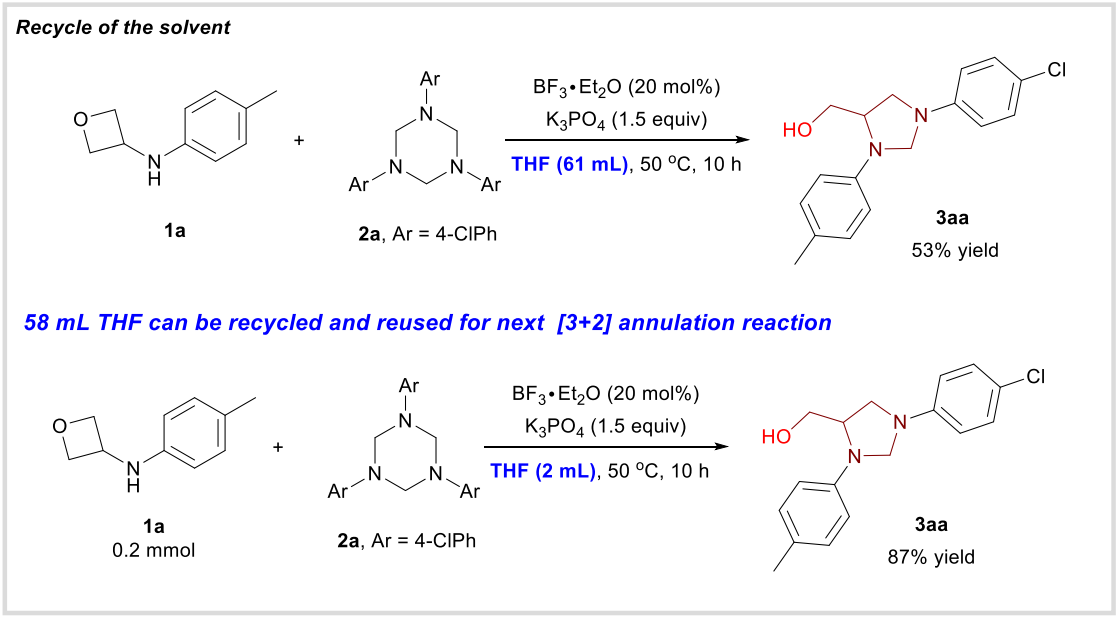
CCDC number: 2096196

5. Recycling the solvent of a gram-scale reaction

To a dried flask was added **1a** (1.0 g, 6.13 mmol), **2a** (2.05 g, 4.9 mmol), and K_3PO_4 (1.89 g, 9.2 mmol), THF (61 mL). Then, $BF_3{\cdot}Et_2O$ (20 mol%) was added to the mixture. The reaction mixture was stirred at 50 °C for 6 h. Then, the solvent THF was recycled through vacuum distillation and the crude product was purified by chromatography on silica gel directly to give the desired product **3aa** in 53% isolated yield as a white solid.

As a result, 95% of THF (58 mL) can be recycled. The resulting THF can be reused for the next [3+2] annulation reaction to give the expected product without the loss of reaction efficiency. These results

strongly show that this transformation is a sustainable method for the synthesis of imidazolidines.



6. Calculation of the atom economy and E-factor for this reaction.

Steps	Reagent 1	Reagent 2	Solvent	Product
1	3-aminooxetane (**1a**), 6.13 mmol, MW:163.10, 1000 mg	1,3,5-triazinane (**2a**), 4.9 mmol, MW: 418.75, 2051.6 mg	THF, 61 mL, MW: 72.1, 54523.2 mg	**3aa**, 3.3 mmol, MW: 302.8, 987.9 mg

$$Atom\ Economy = \frac{302.8}{163.1+418.8} \times 100\% = 52.0\%$$

With solvent recycling

$$E\text{-}factor = \frac{\sum 1000+2051.6-987.9}{987.9} = 2.1$$

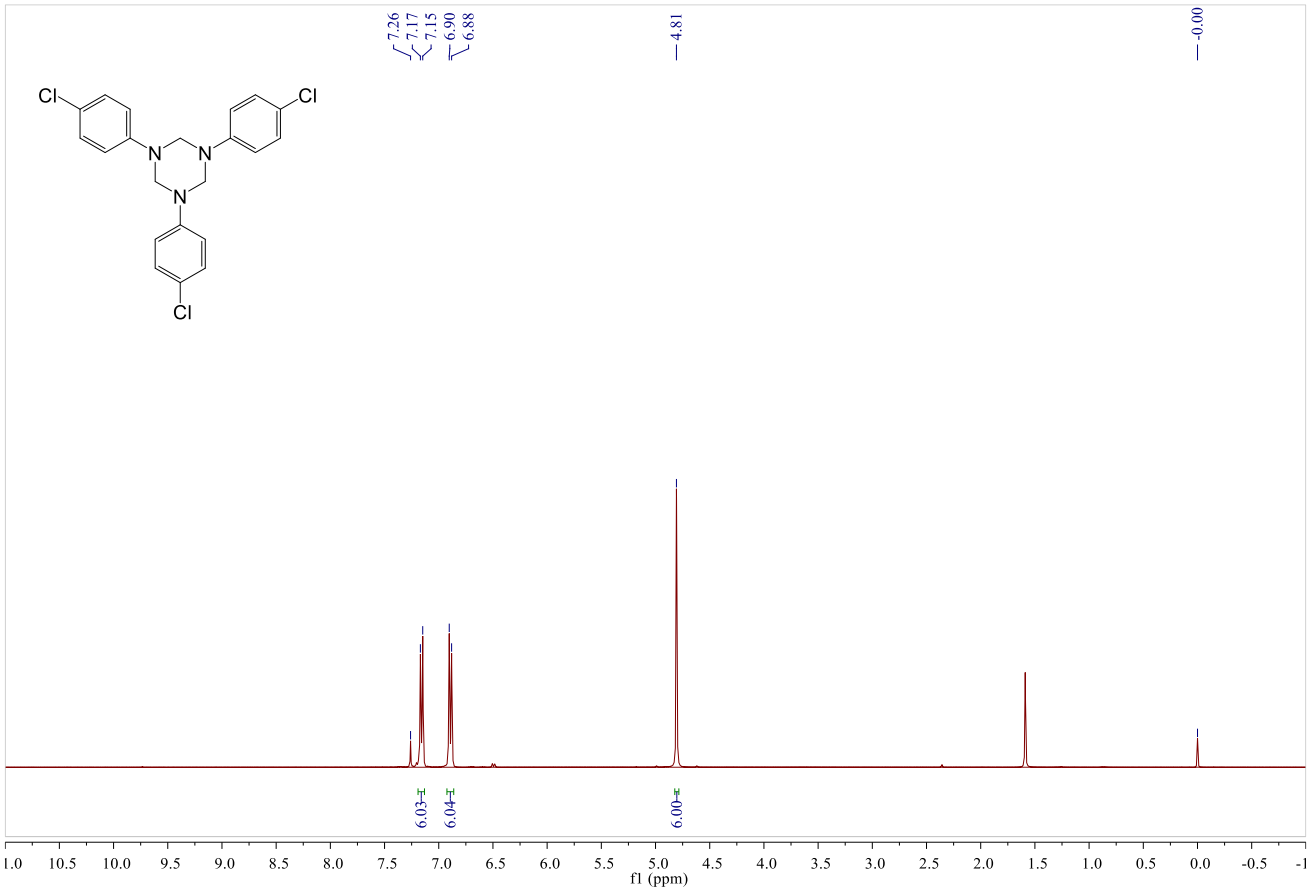
Without solvent recycling

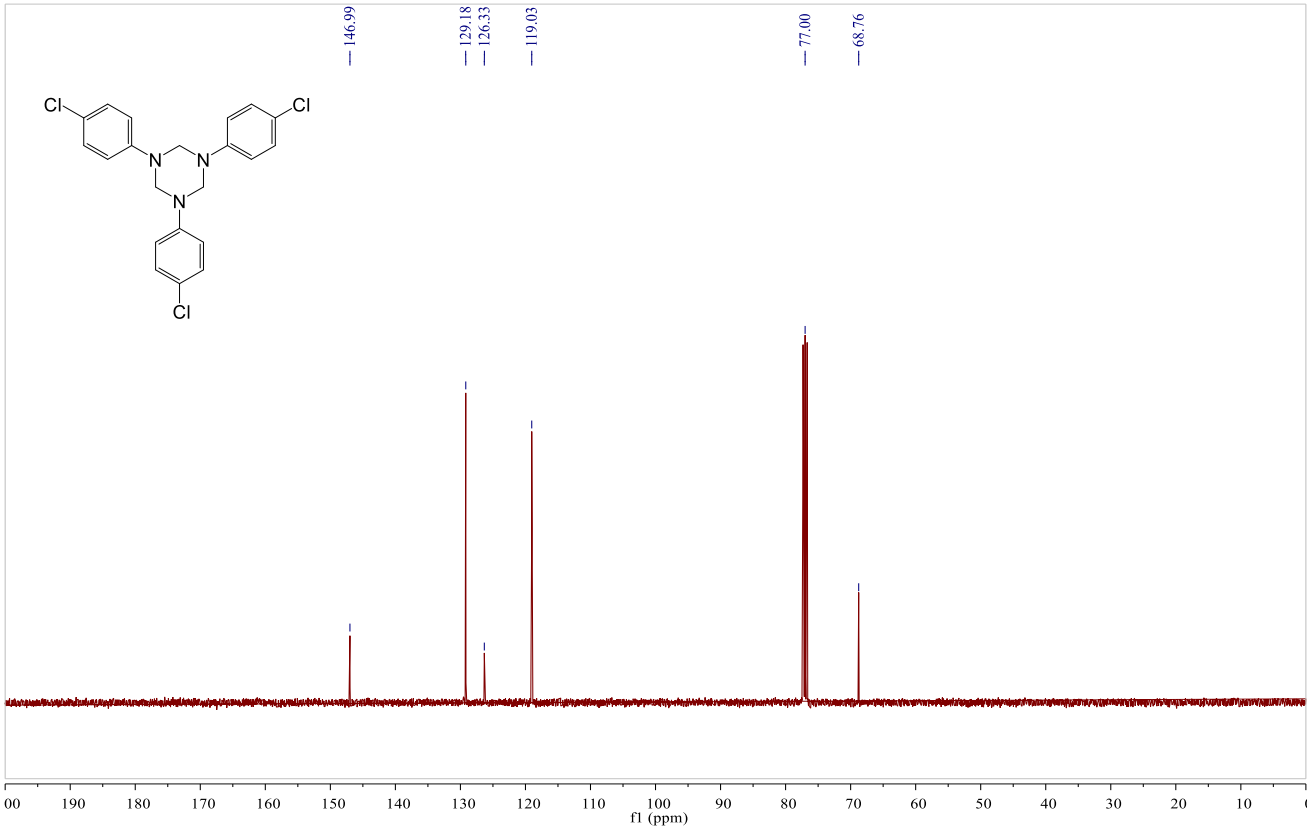
$$E\text{-}factor = \frac{\sum 1000+2051.6+54523.2-987.9}{987.9} = 57.3$$

The green chemistry metrics of this reaction such as atom economy and E-factor were calculated. The present reaction scored moderate atom economy (52%) and E-factor (2.1).

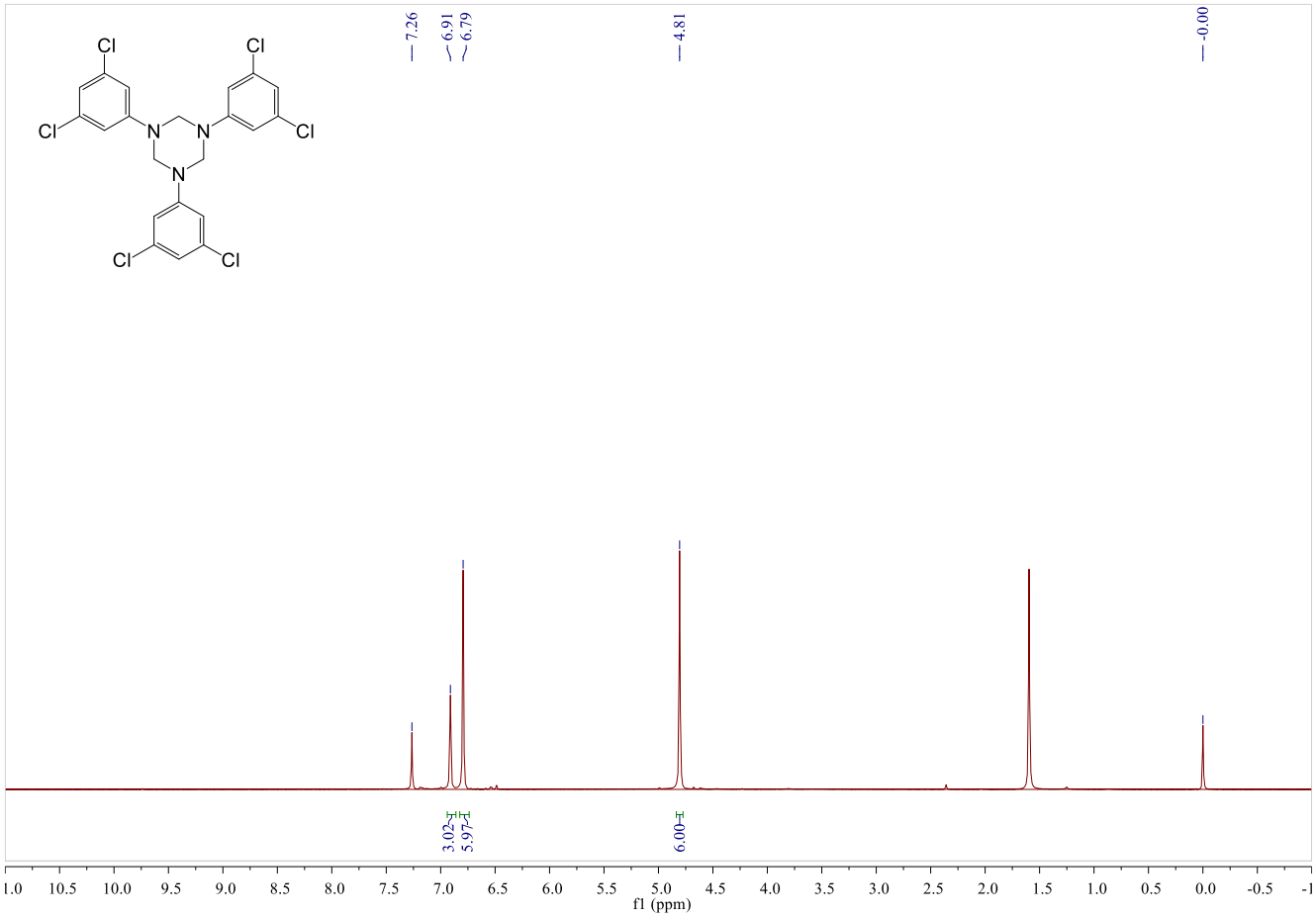
7. NMR Spectra of products 2a-2f, 3aa–3ai, 3ba–3wa, 5aa-5ae, 7 and 9

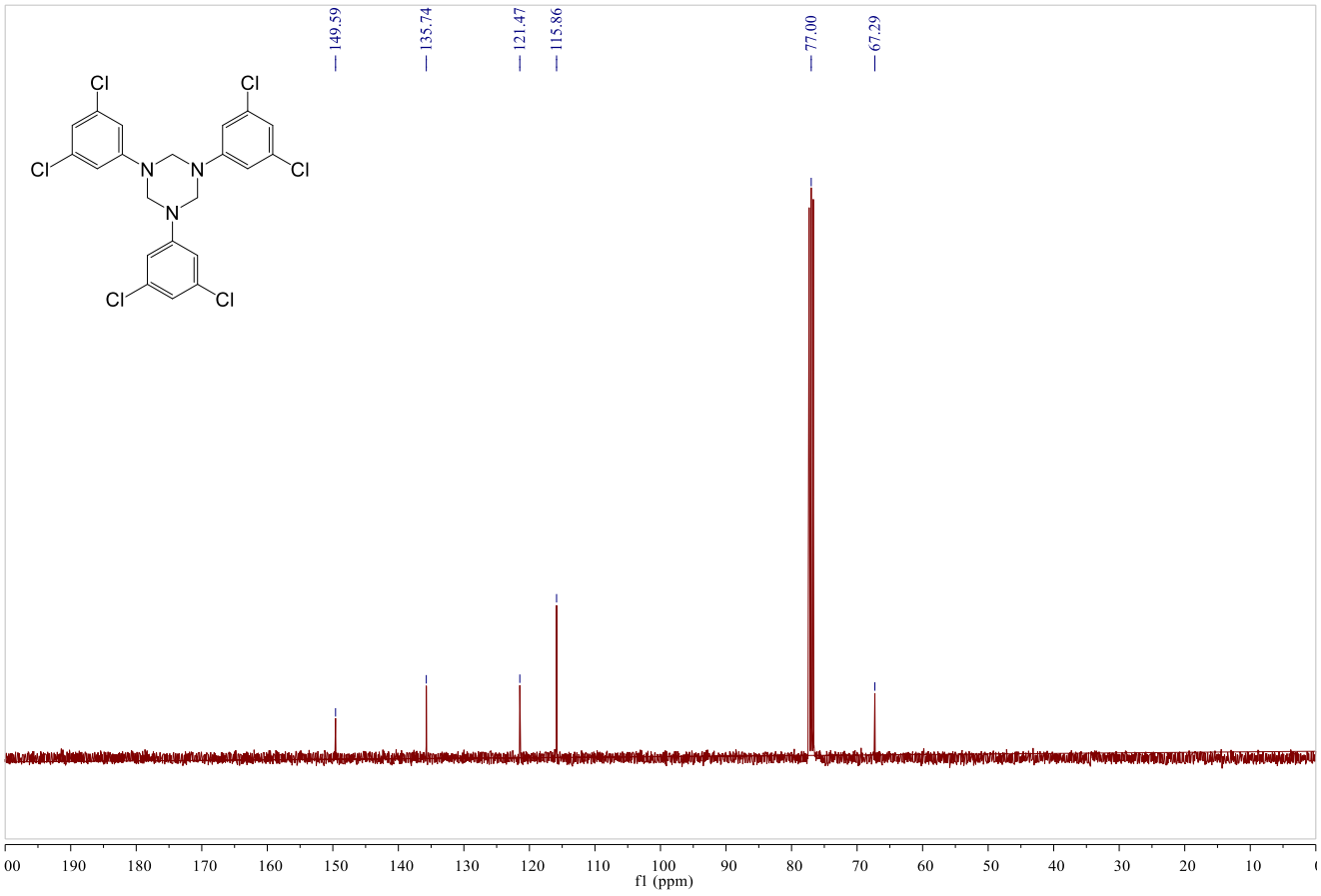
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 2a



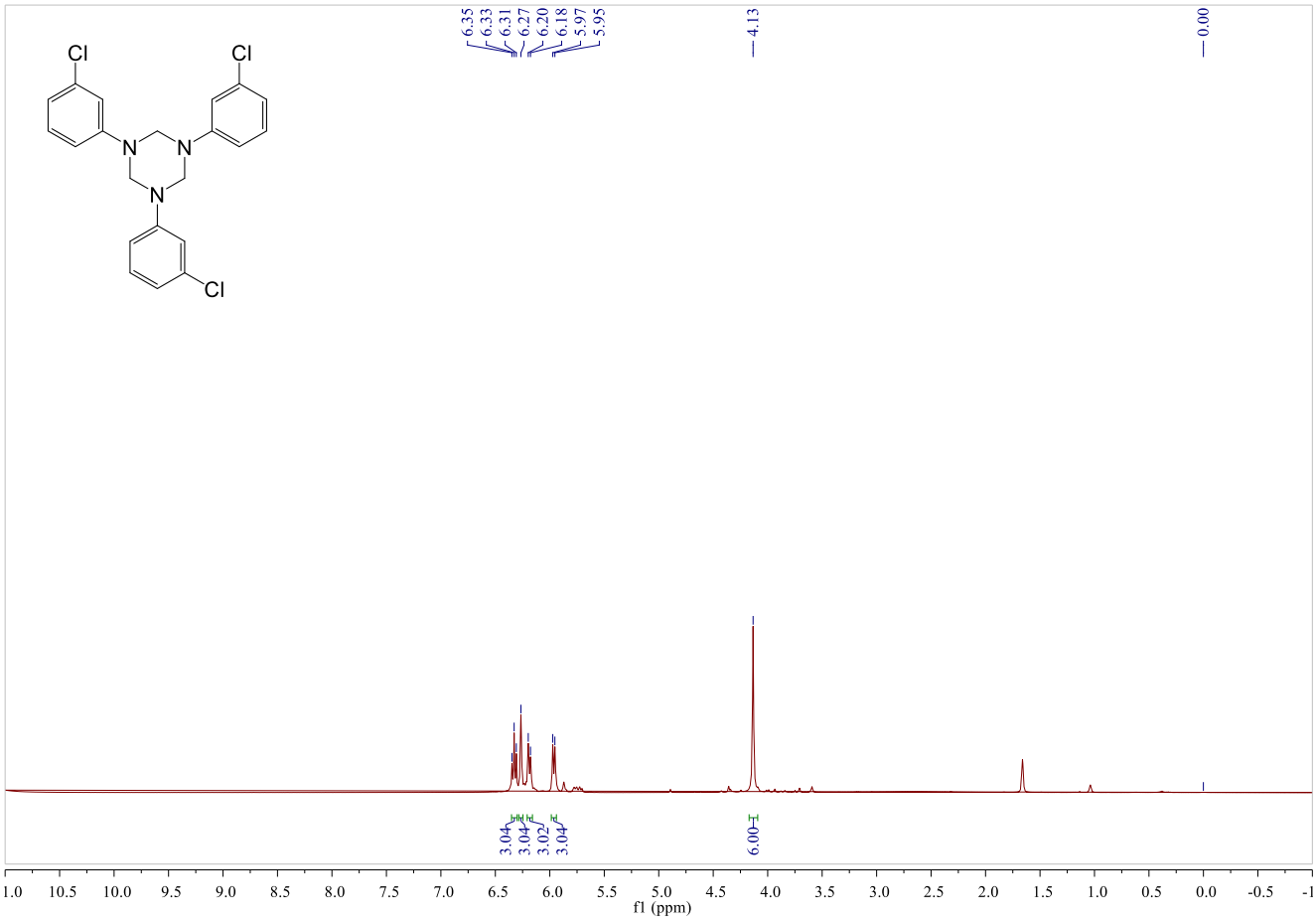


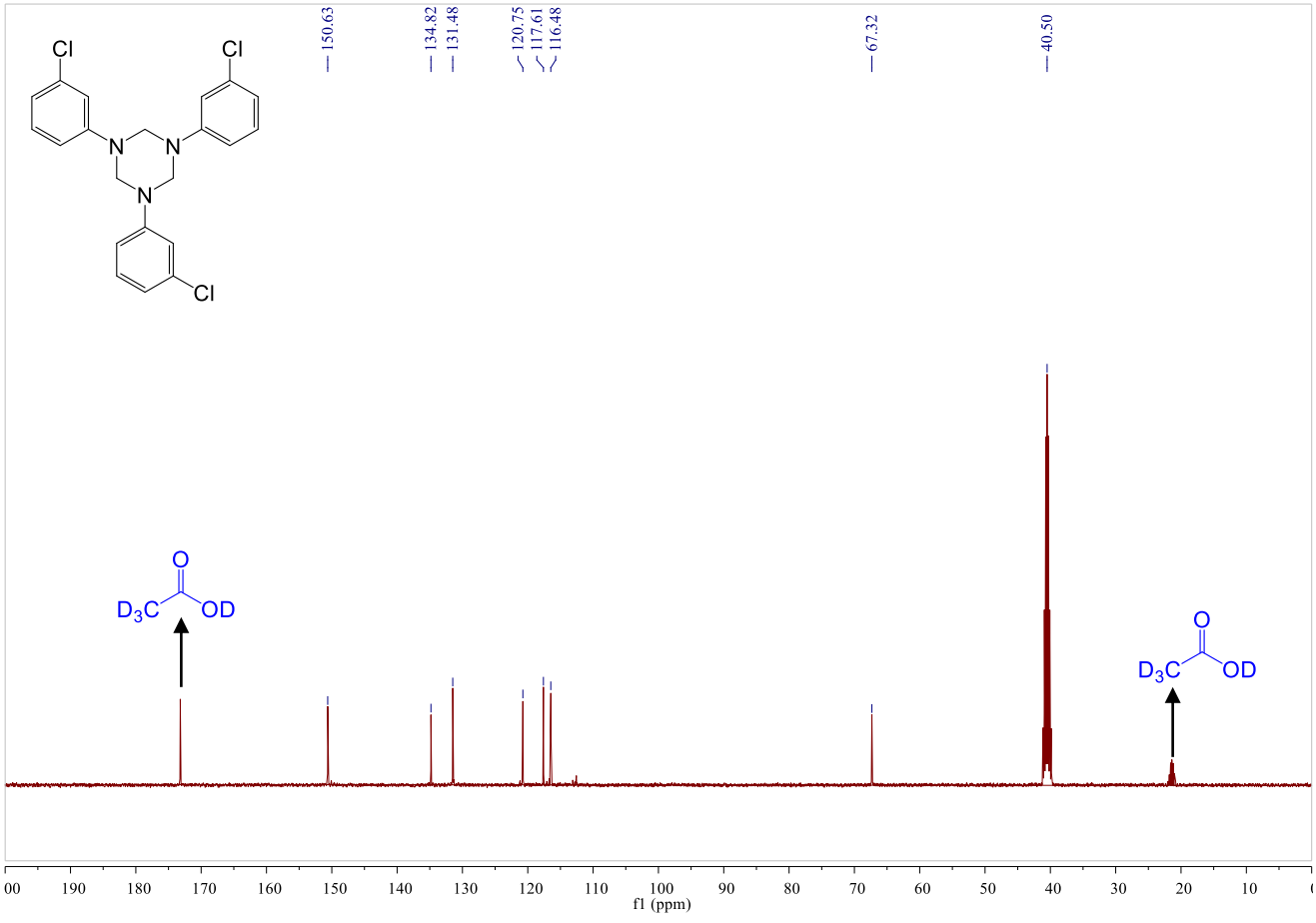
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 2b



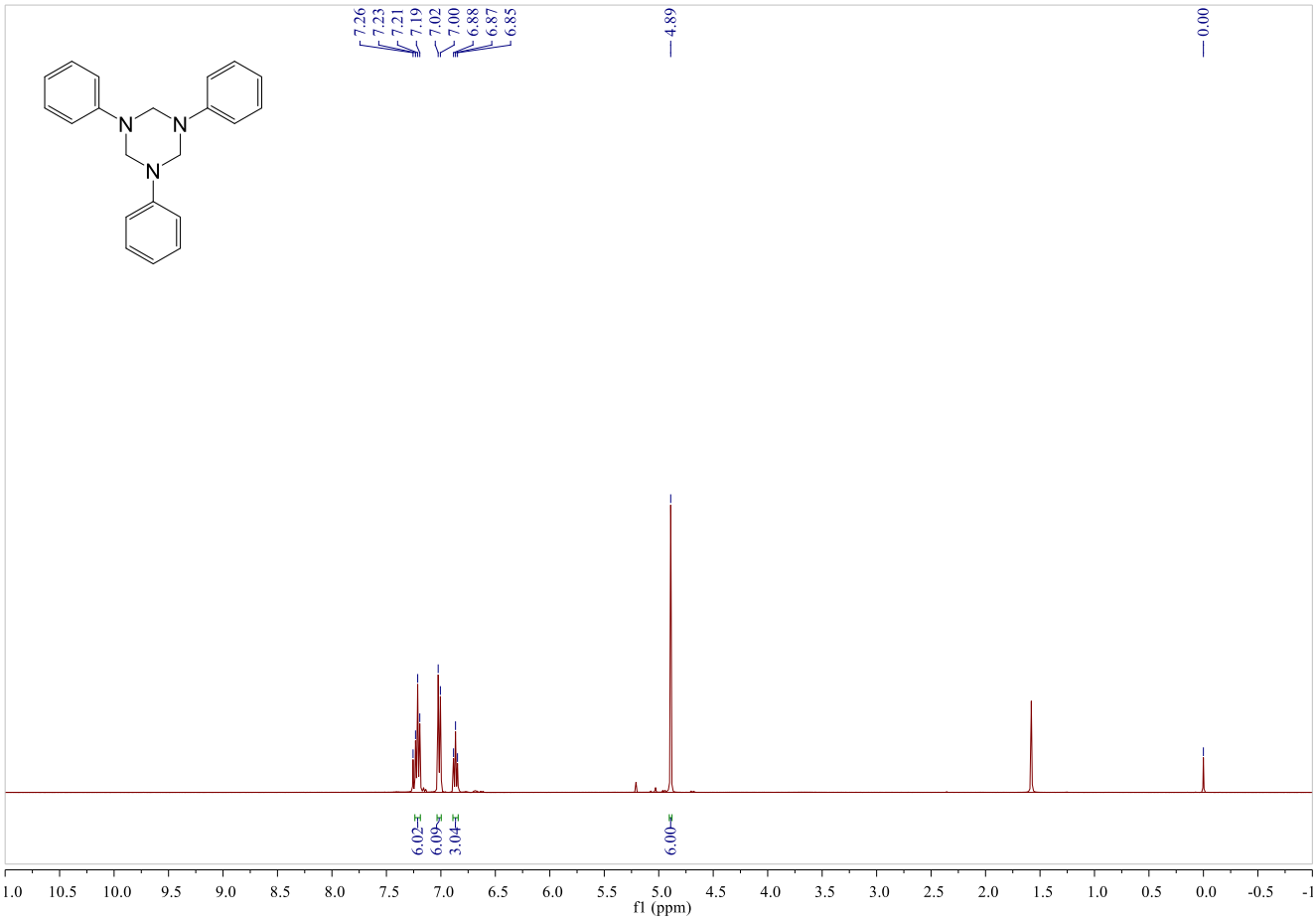


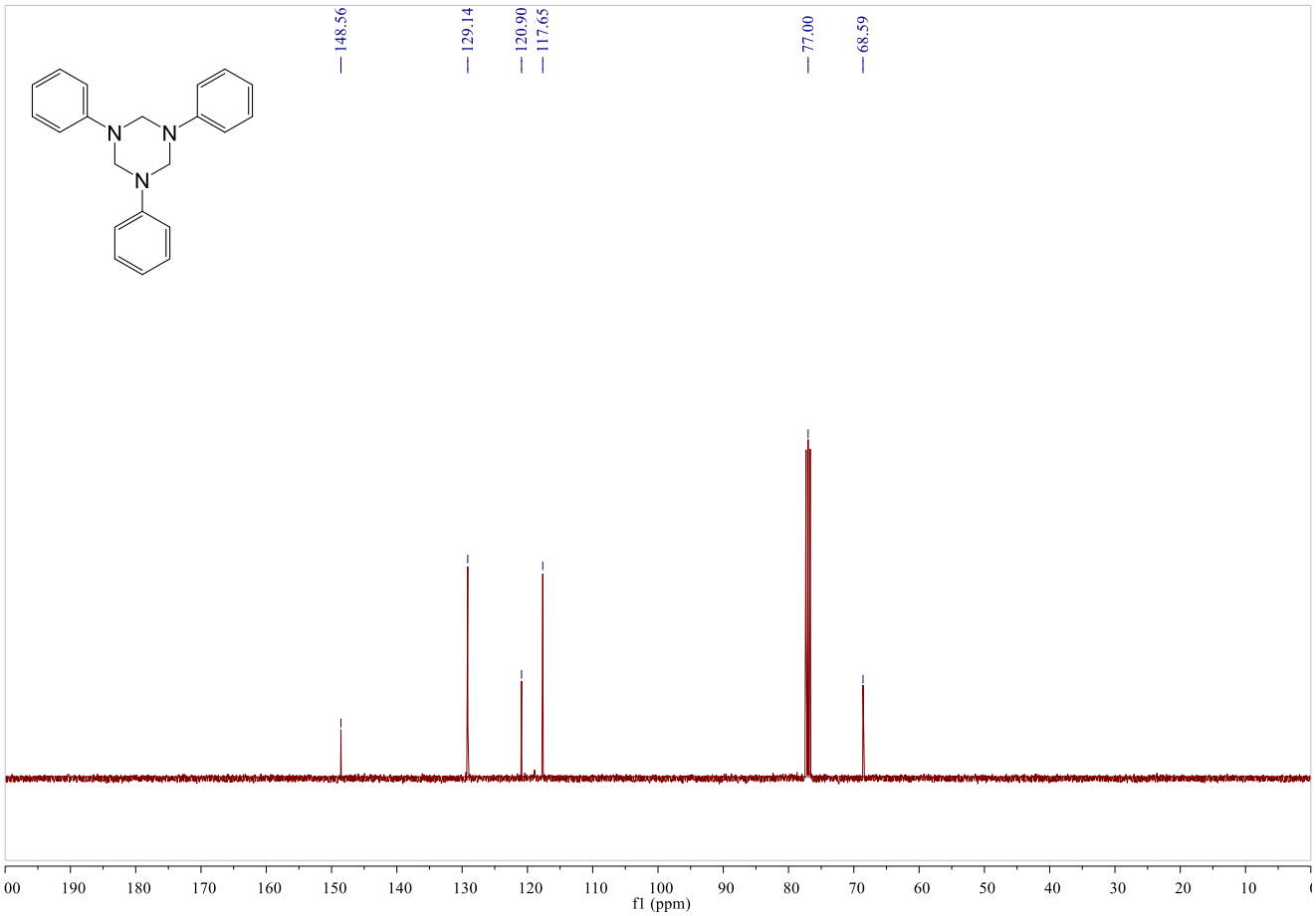
^{1}H NMR (400 MHz, d^{6}-DMSO+CD$_3$COOD) and ^{13}C NMR (d^{6}-DMSO+CD$_3$COOD) spectra of product 2c



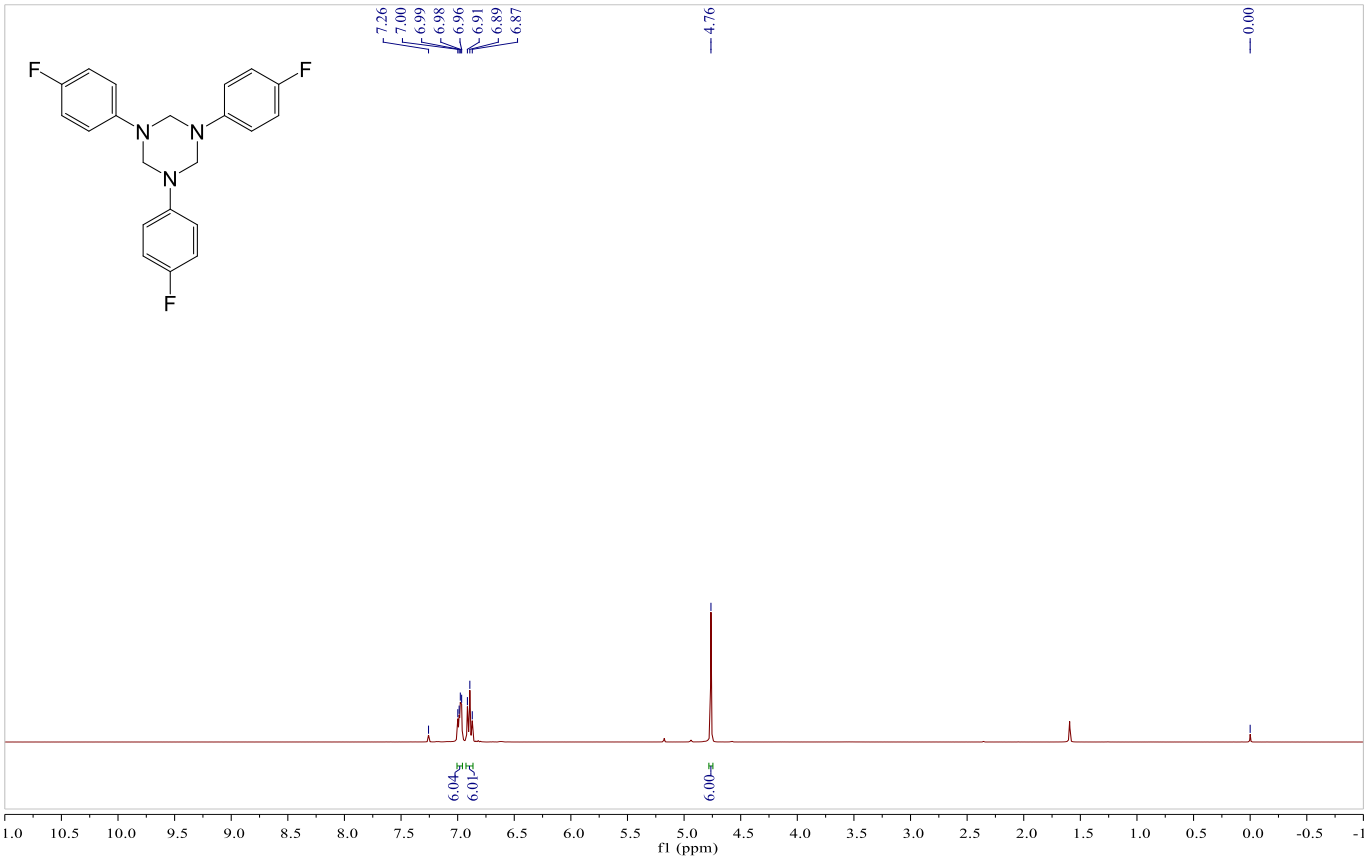


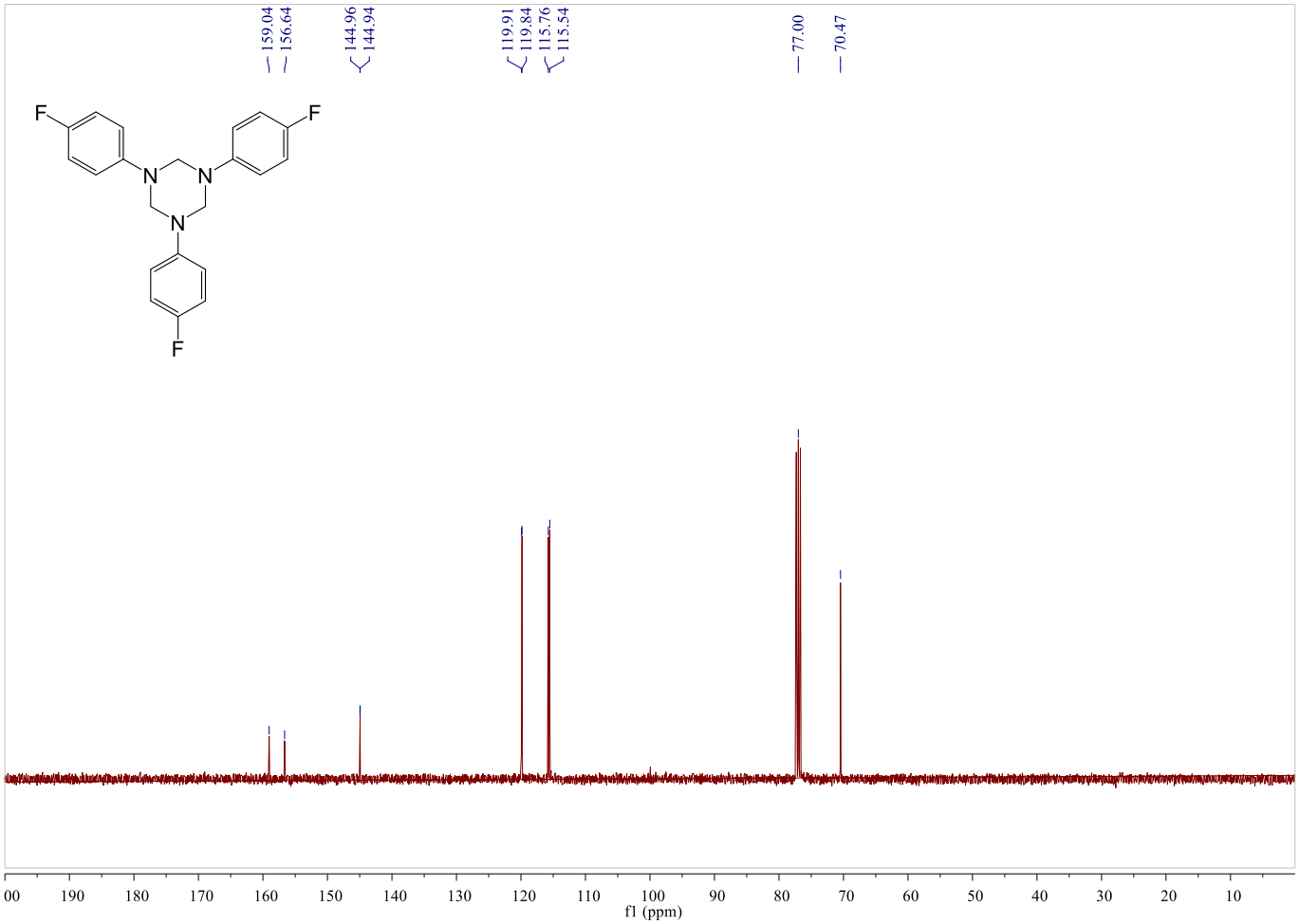
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 2d



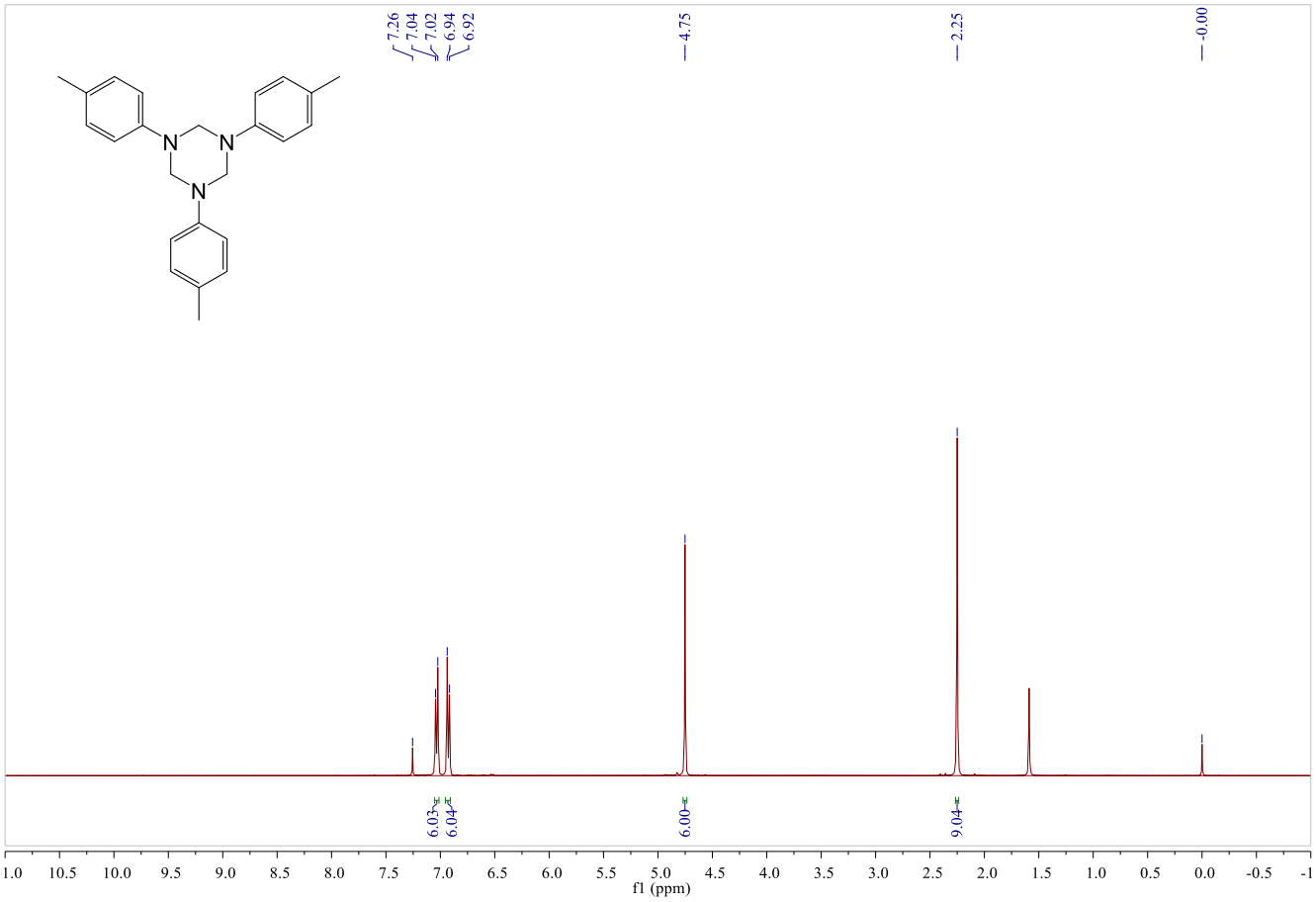


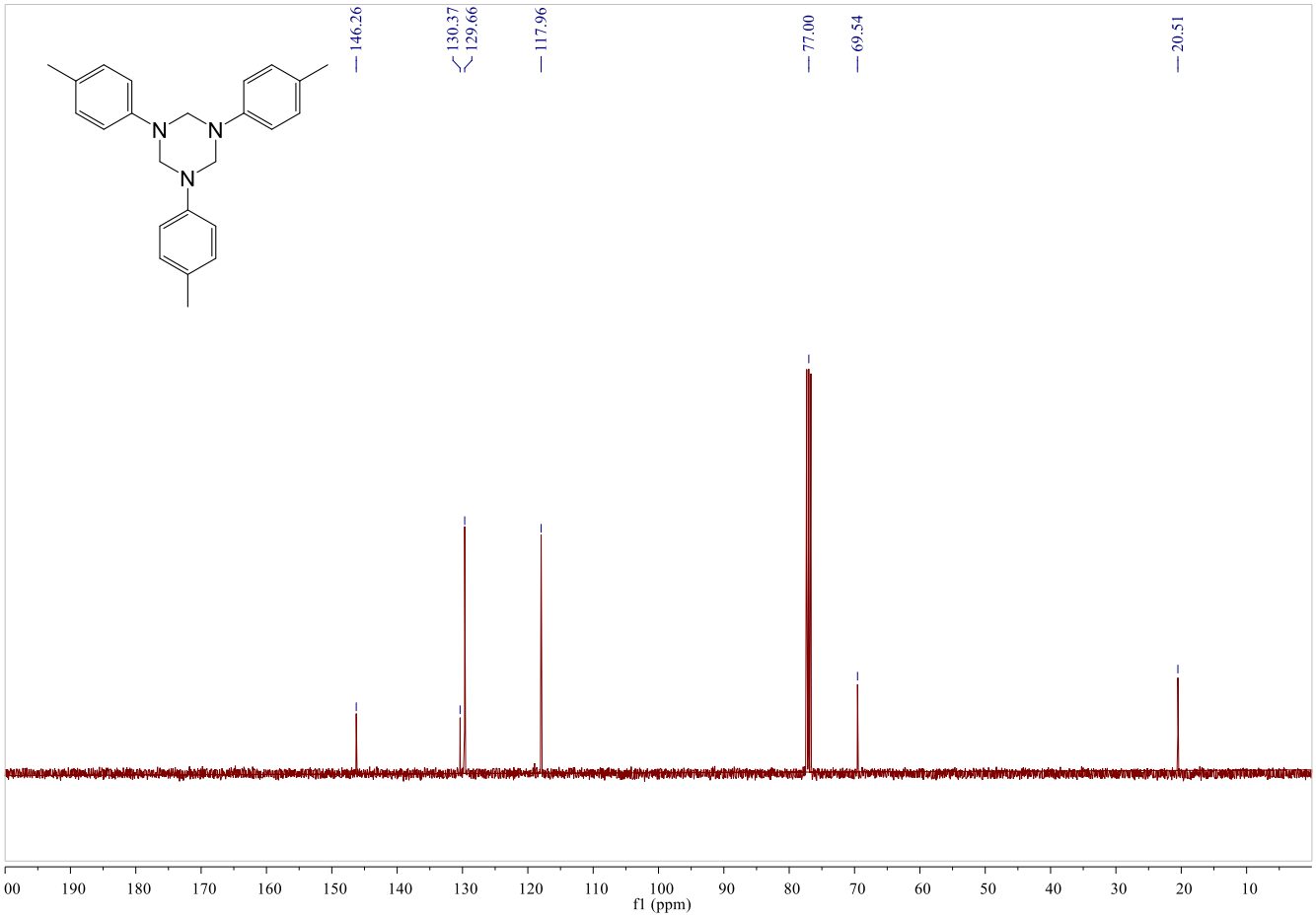
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 2e



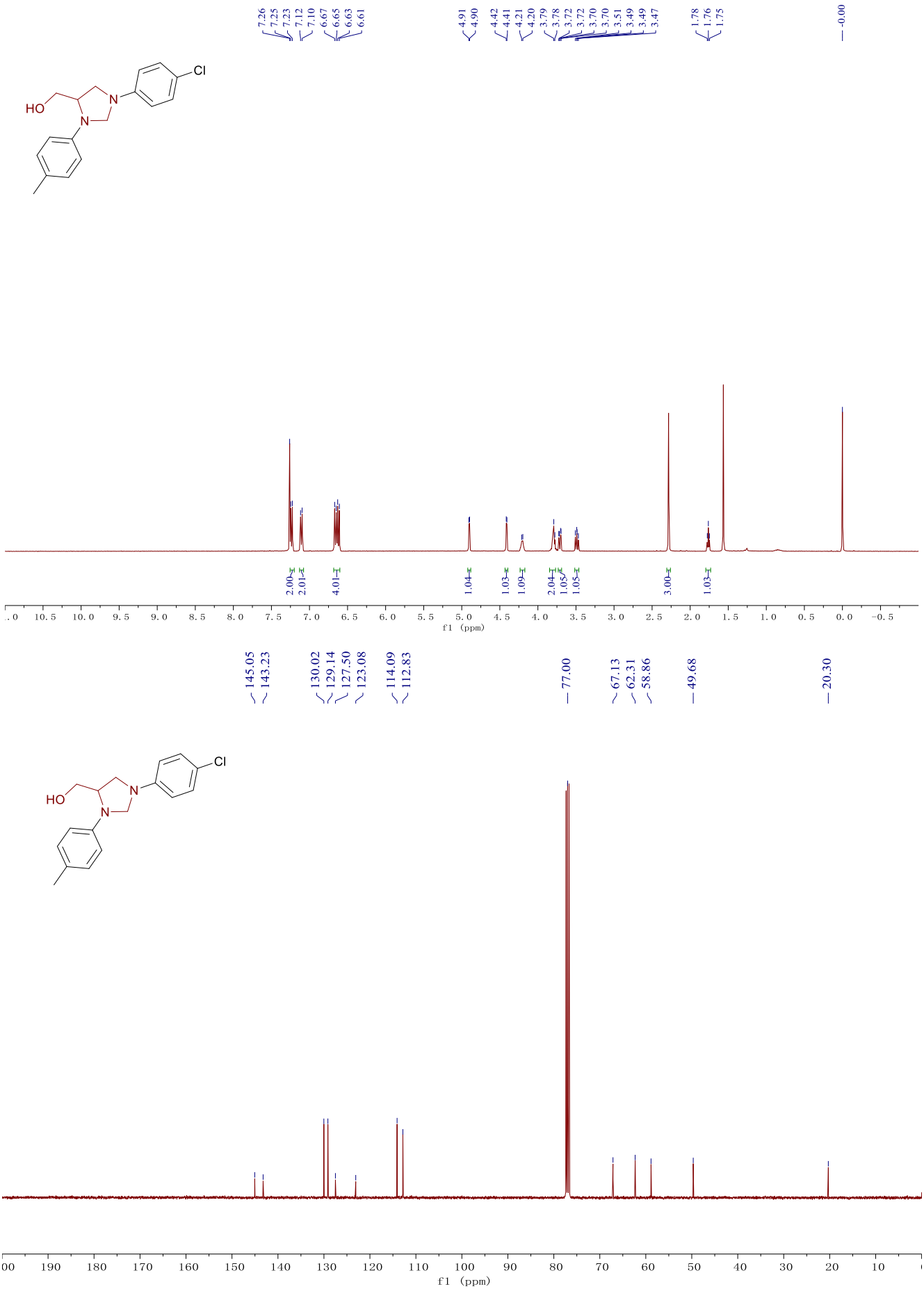


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 2f

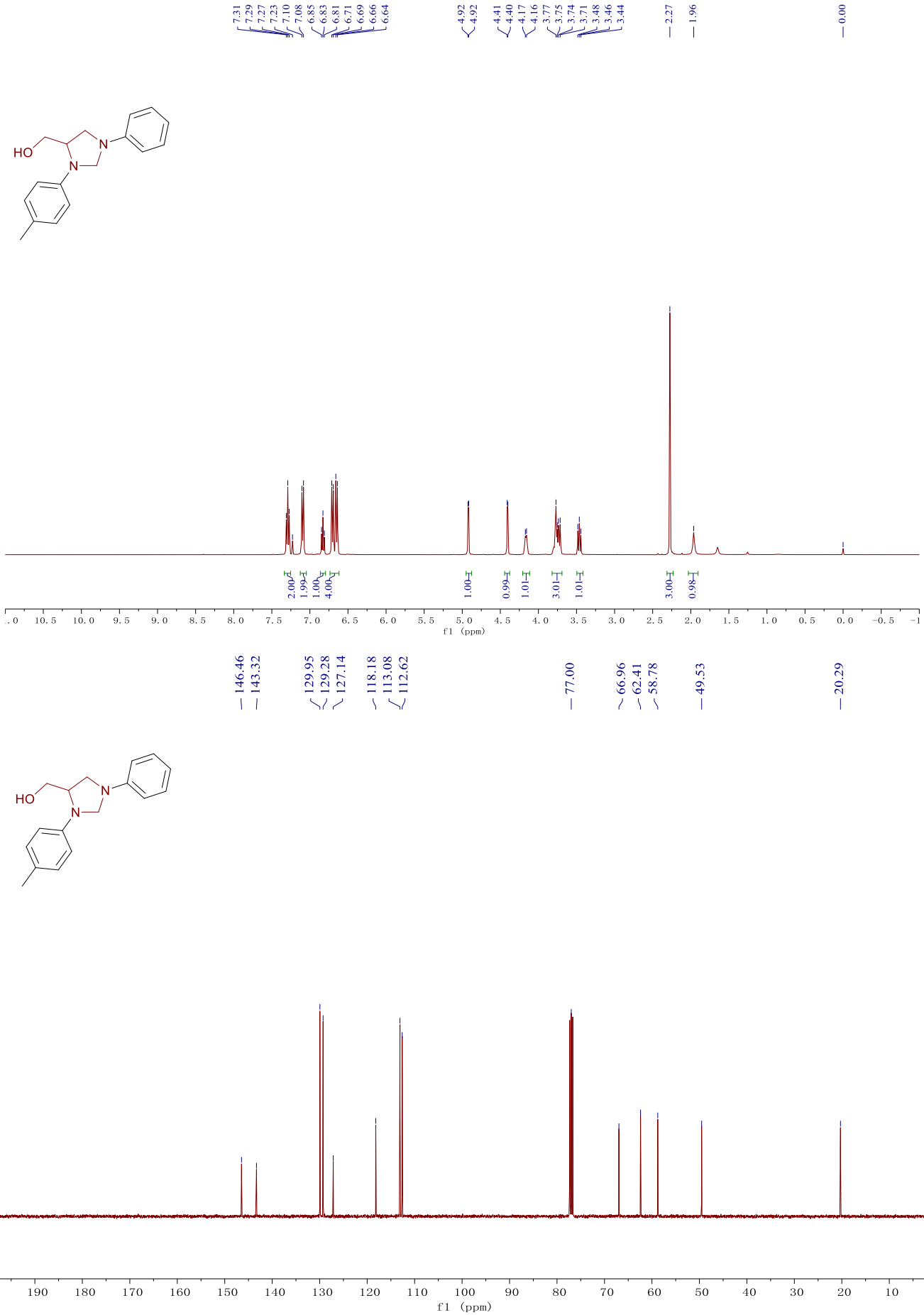




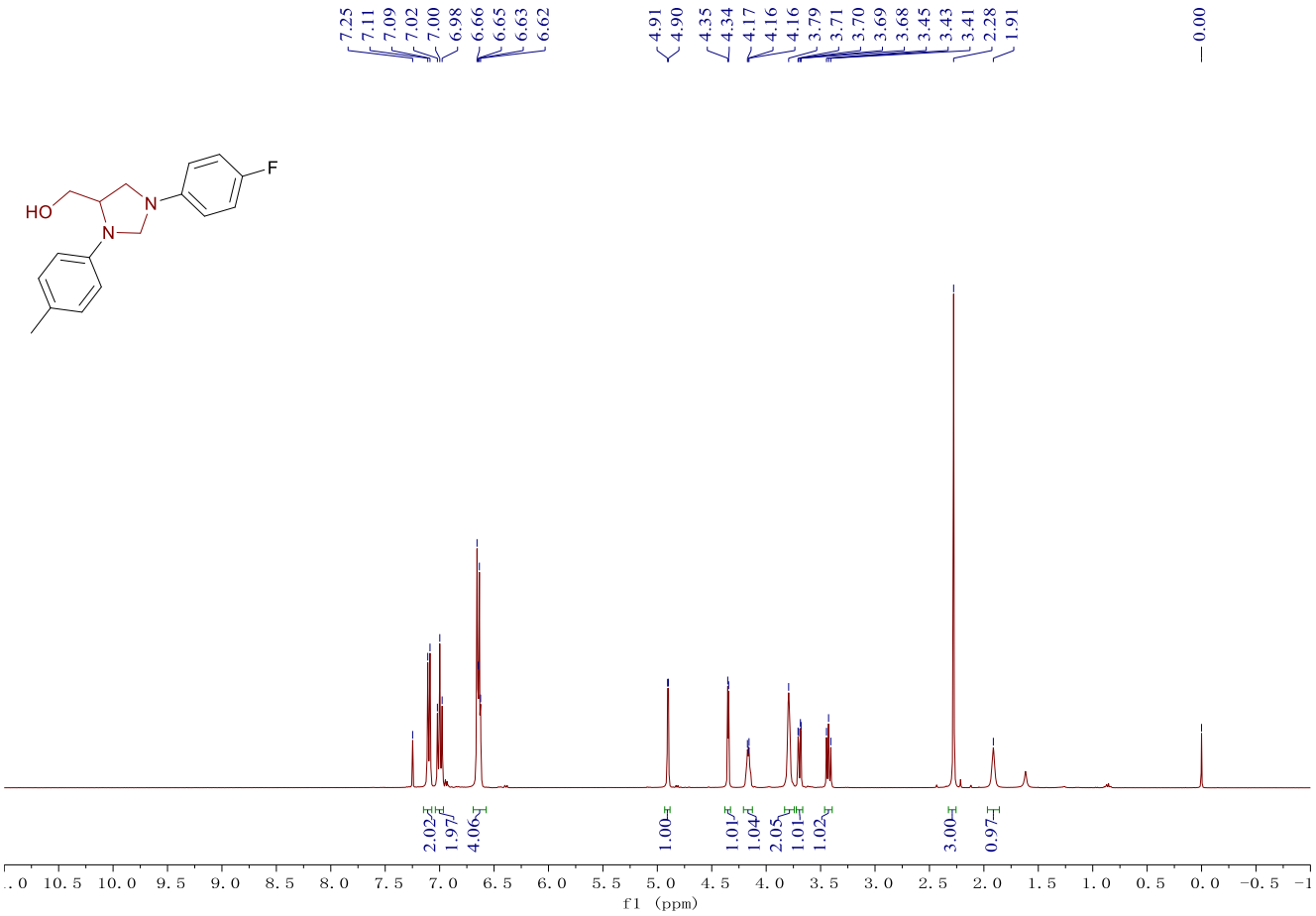
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3aa

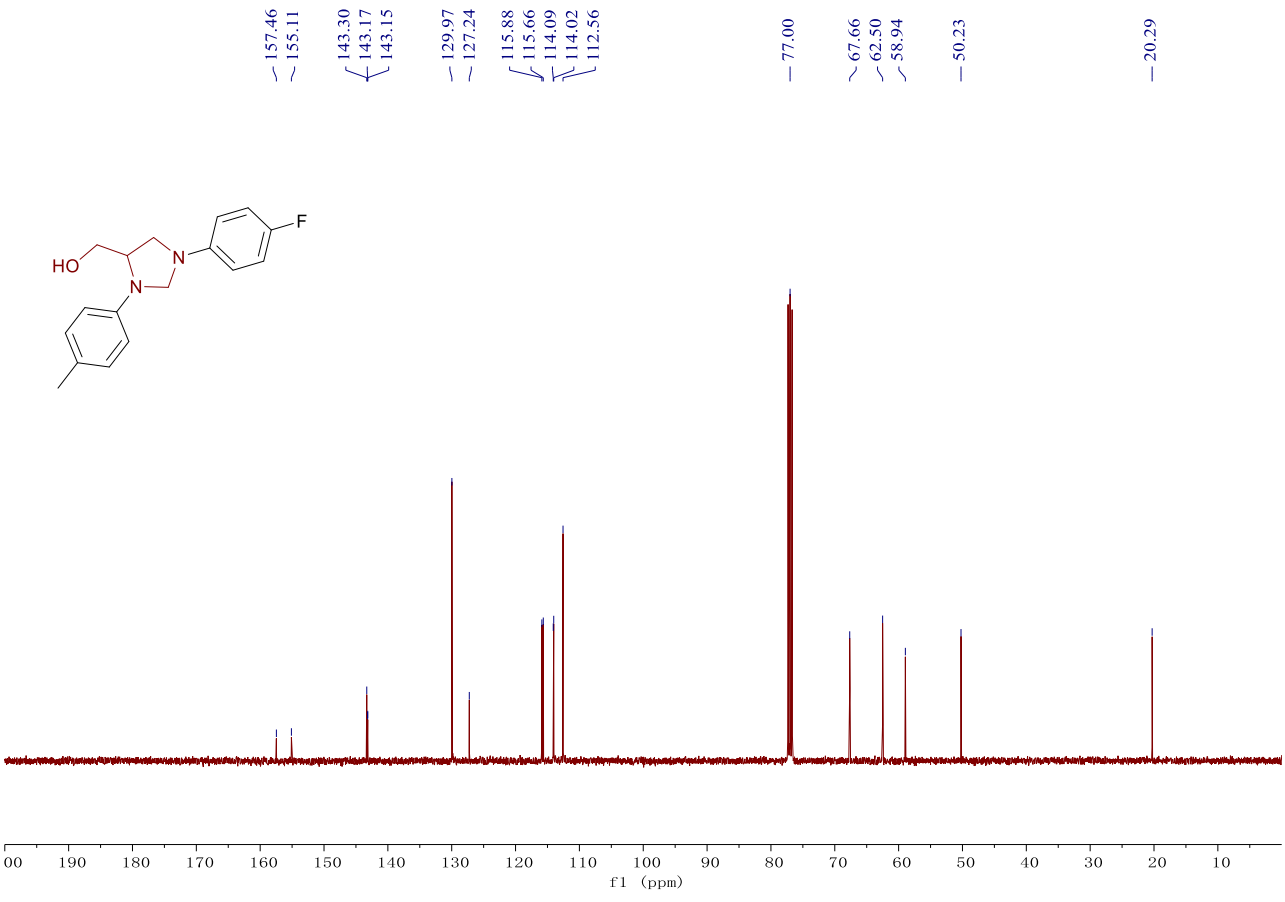


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ab

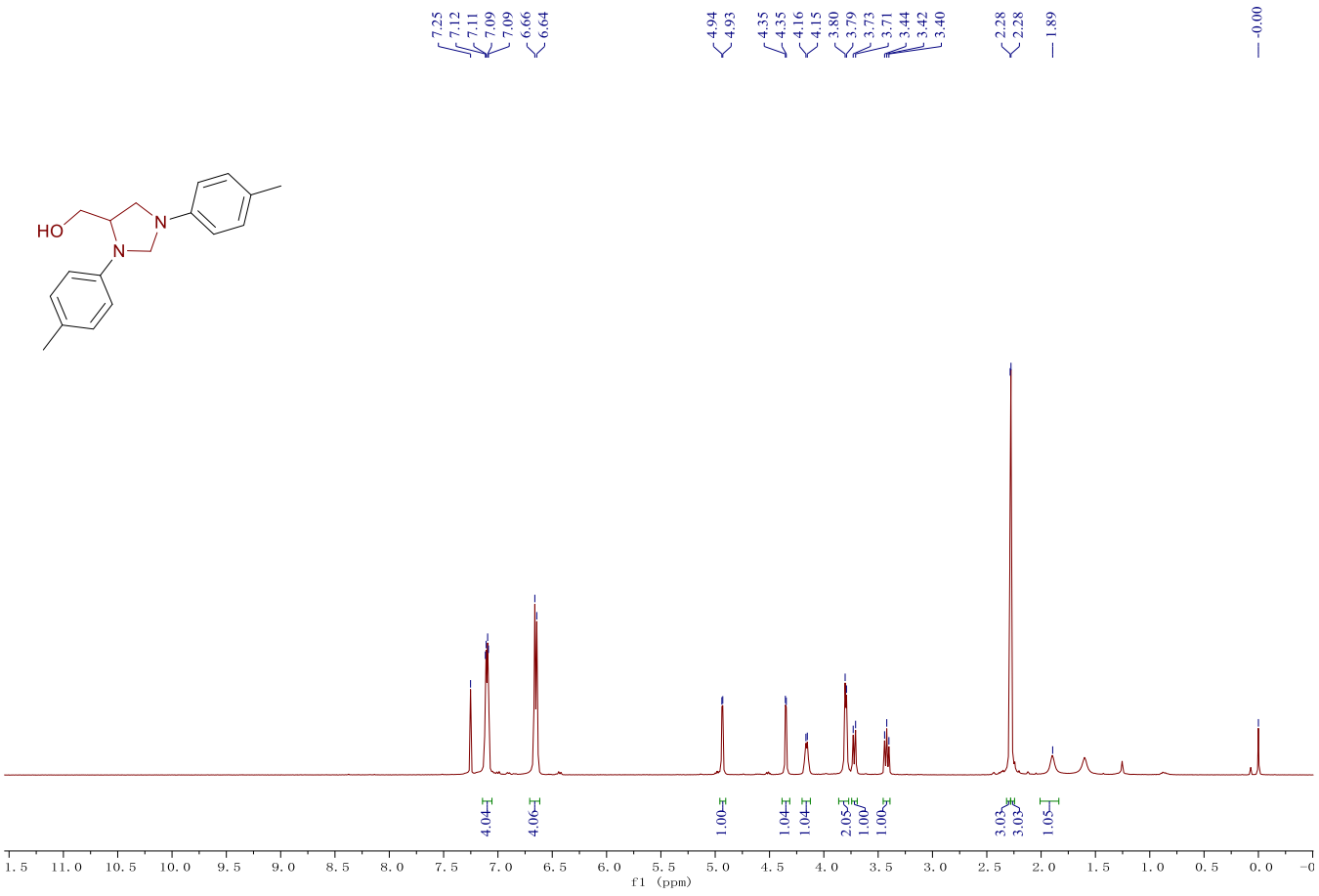


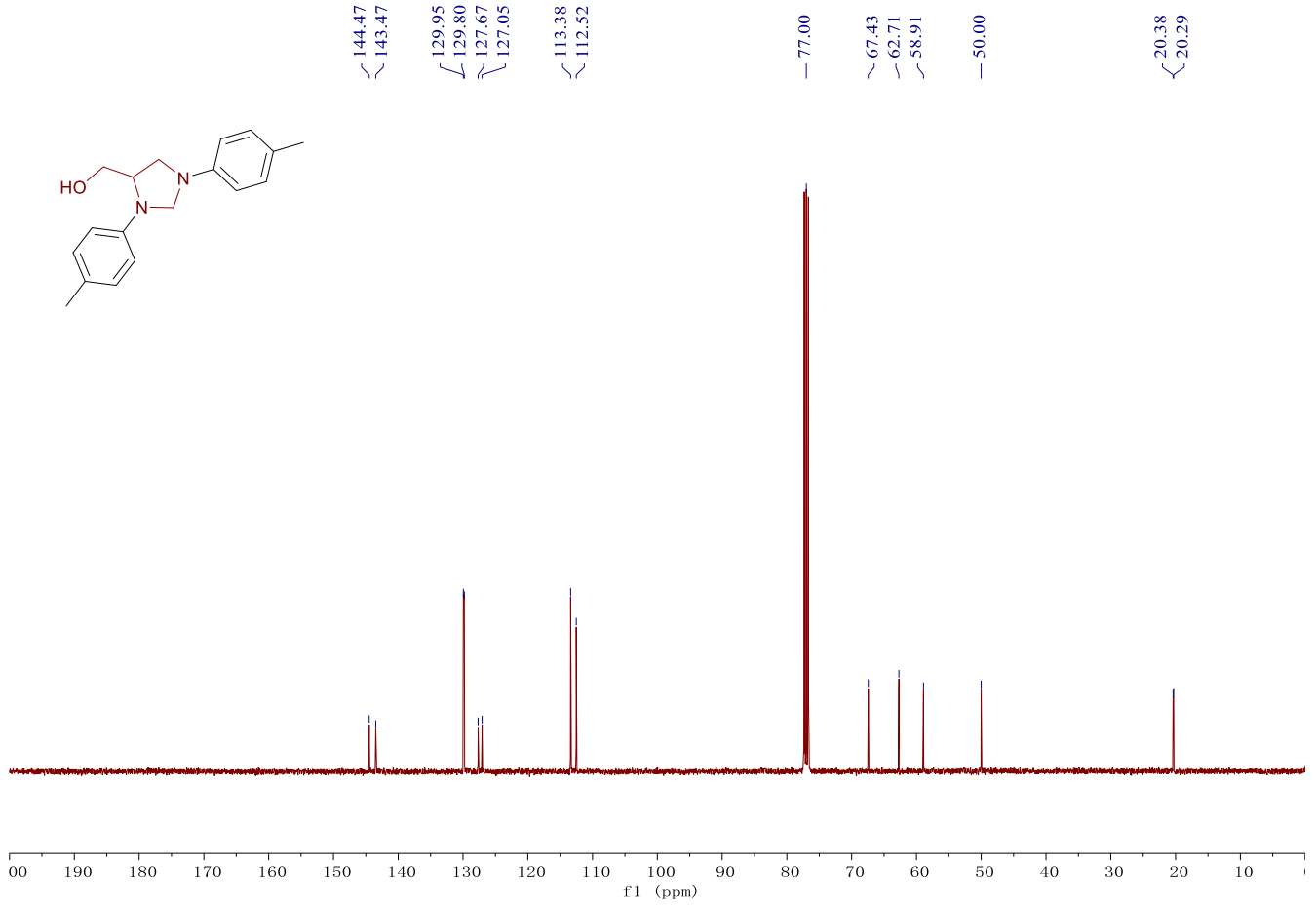
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ac



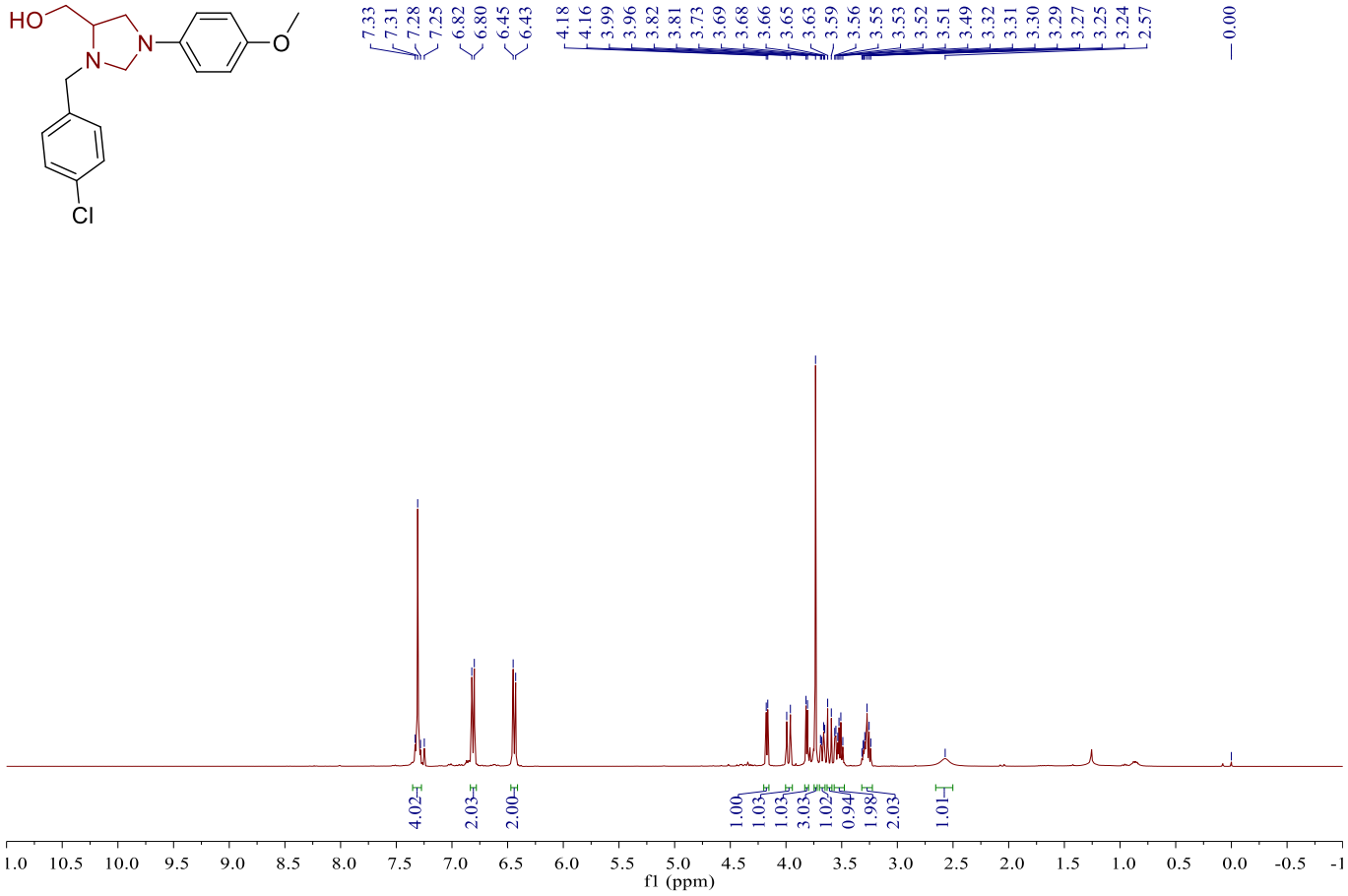


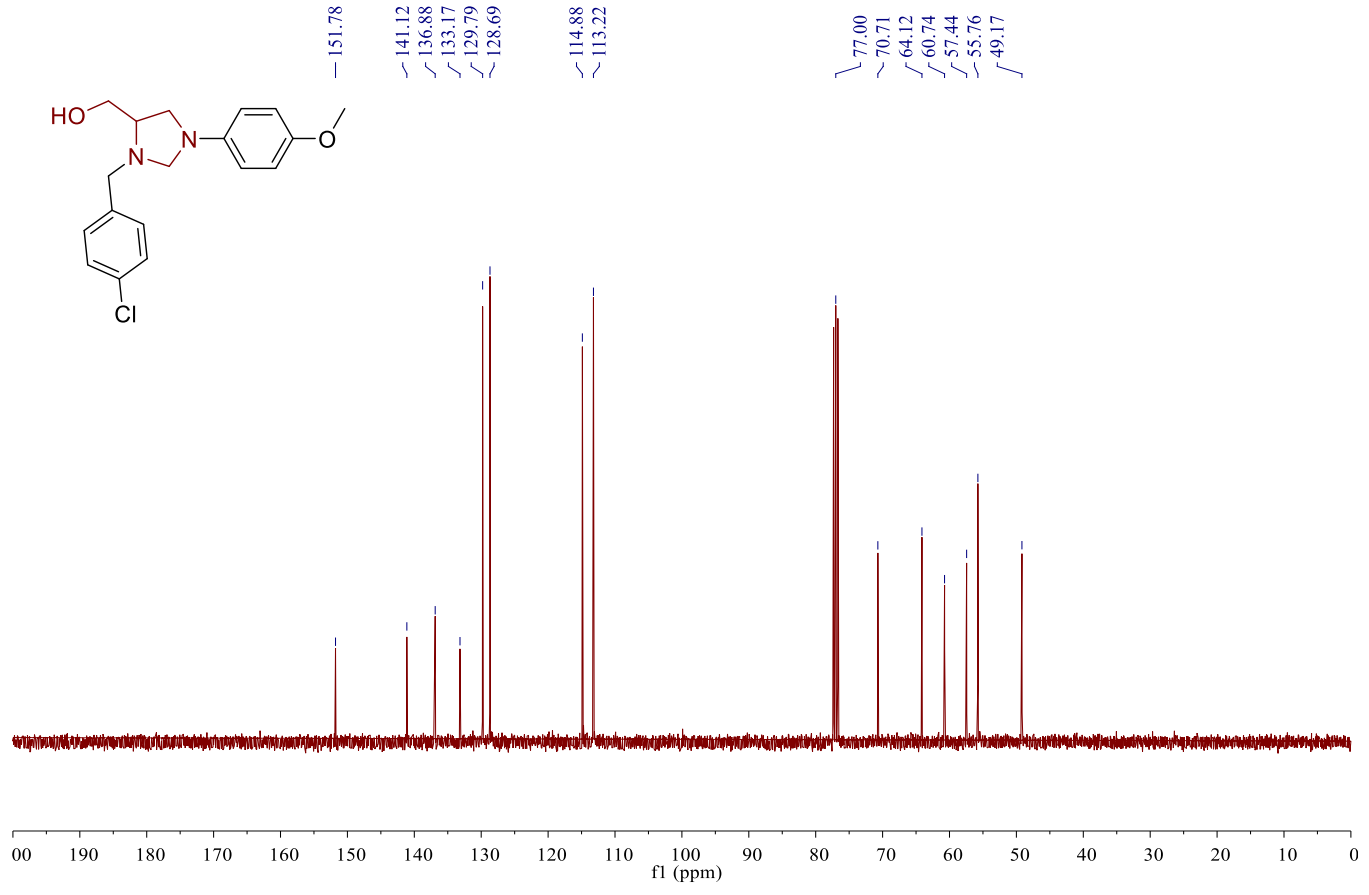
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ad



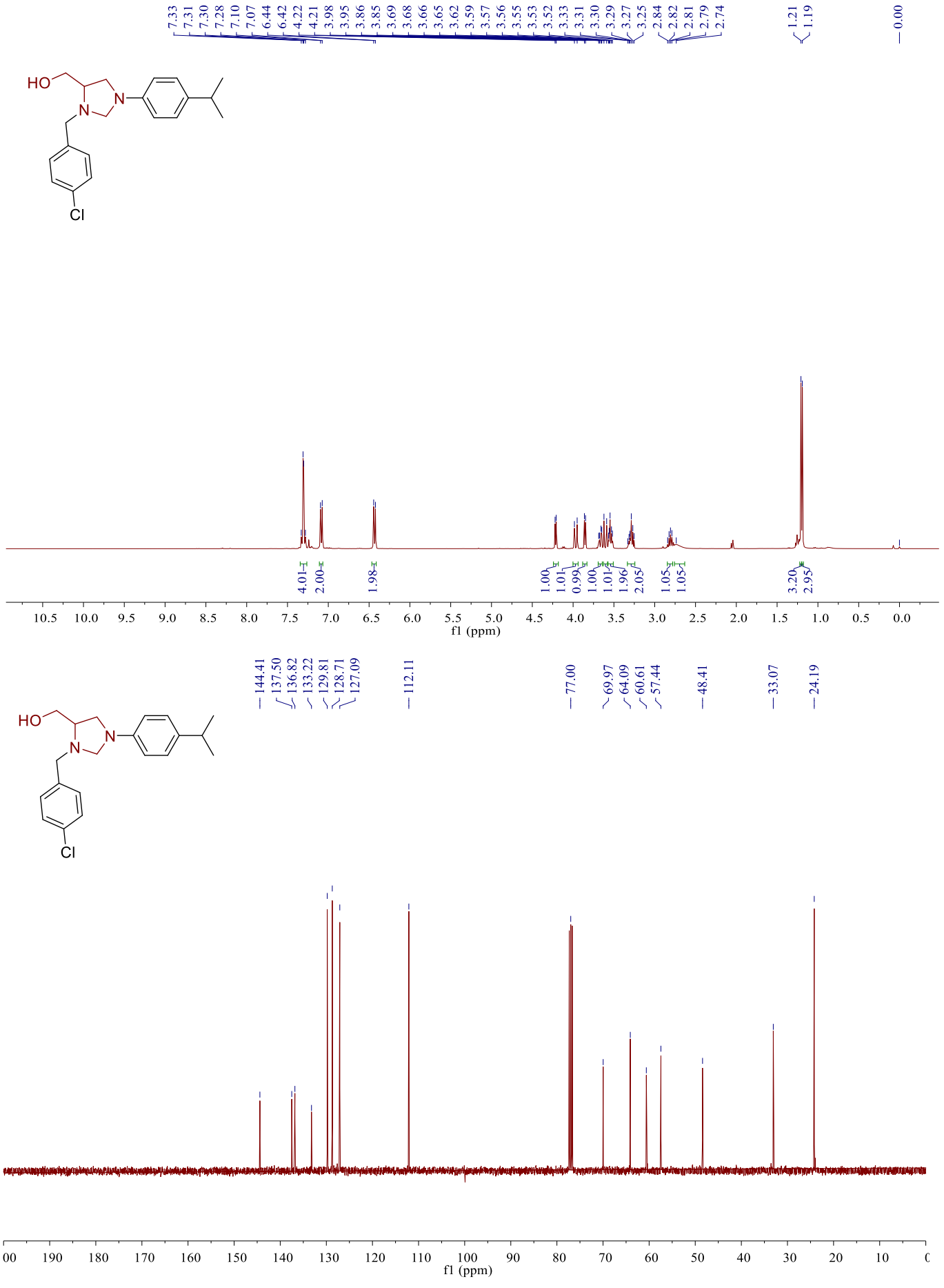


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ae

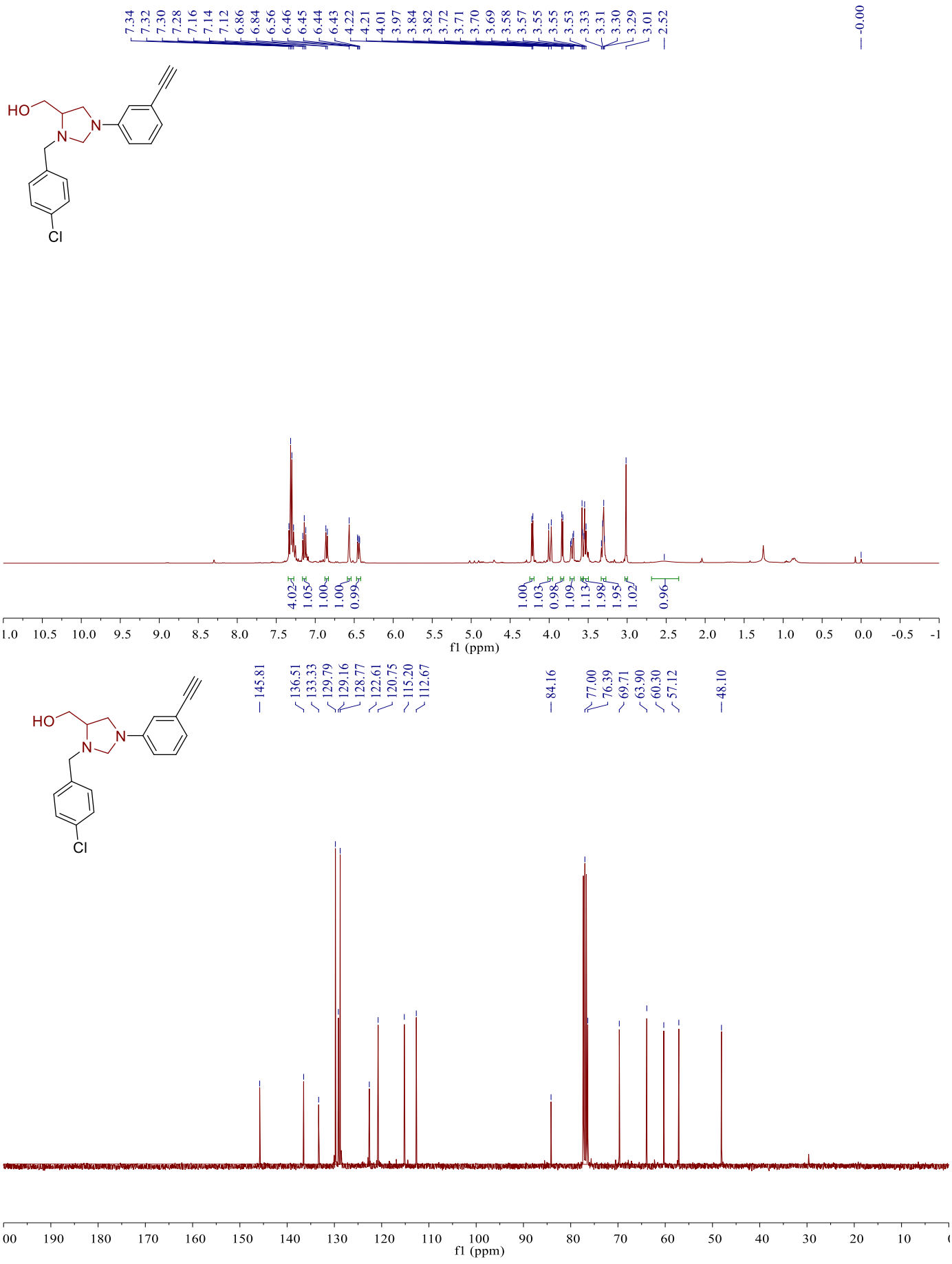




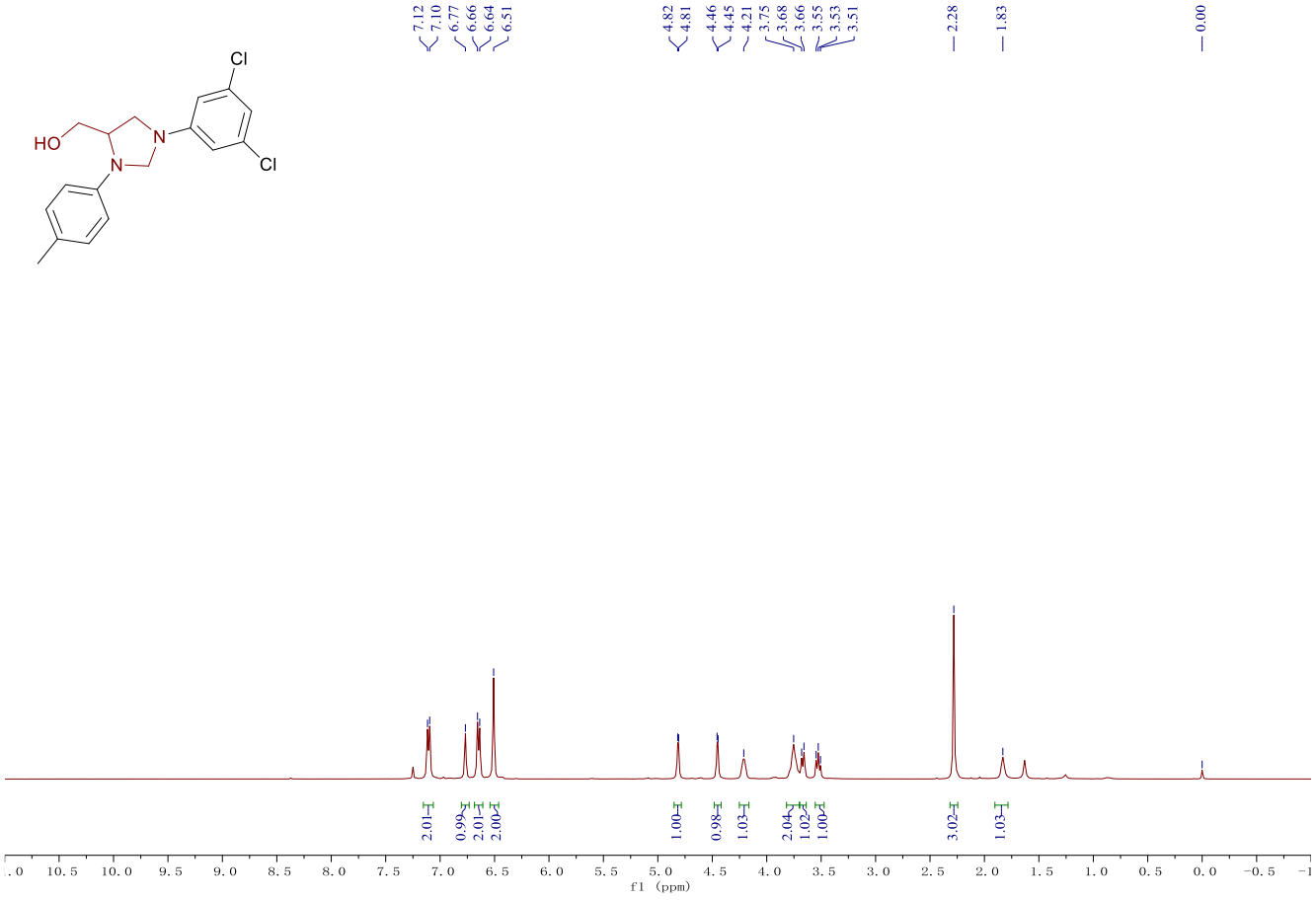
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3af

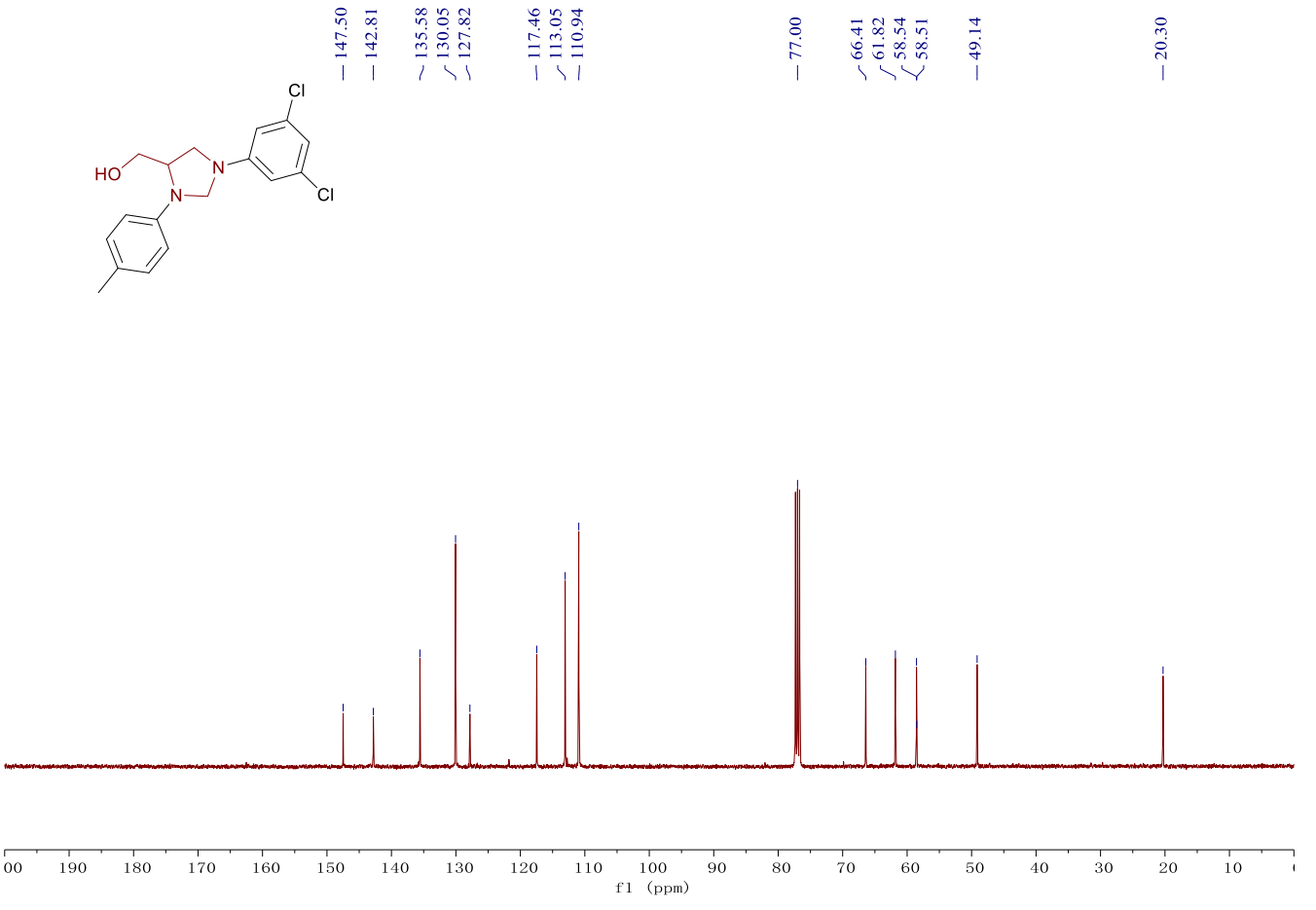


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ag

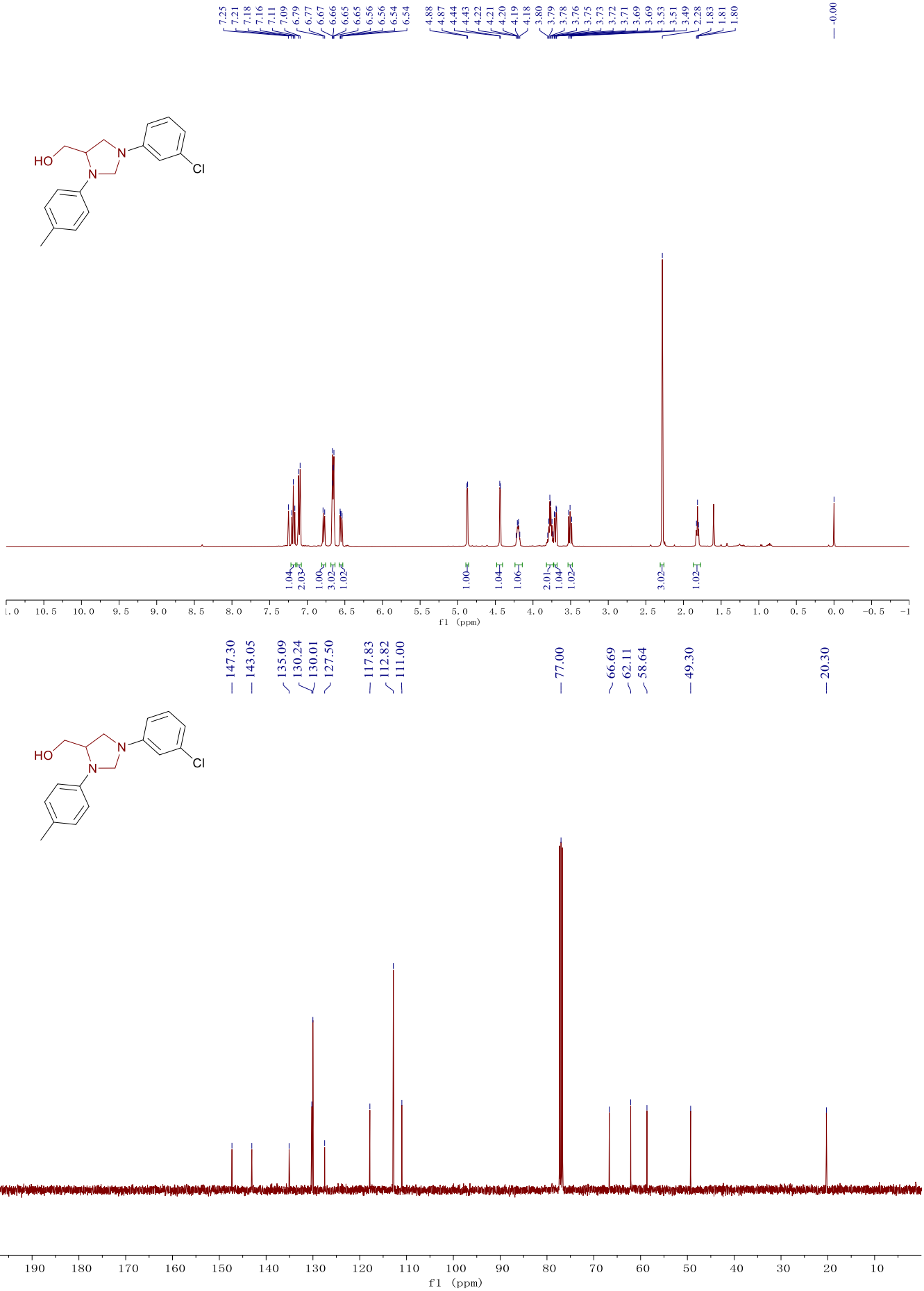


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ah

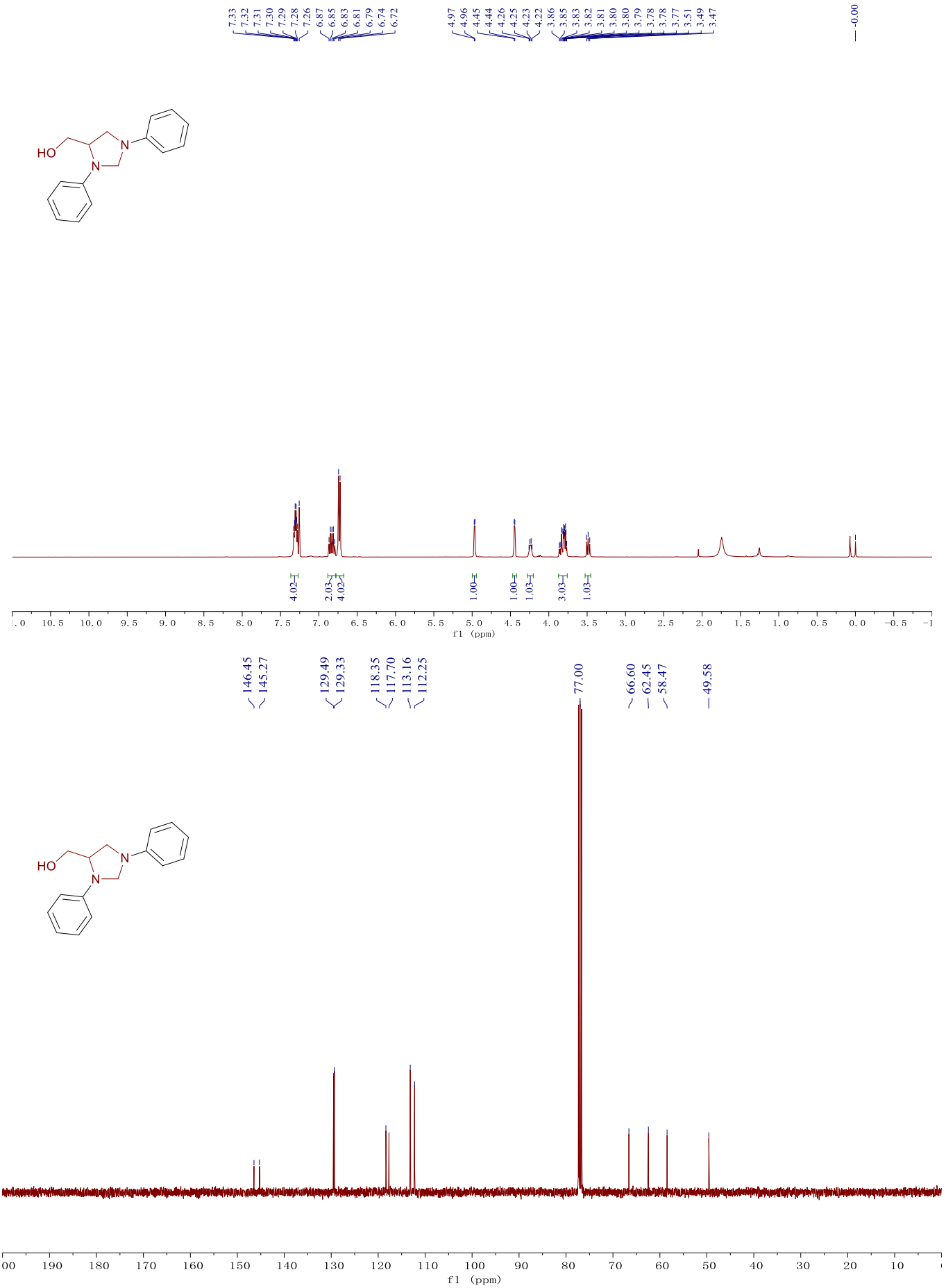




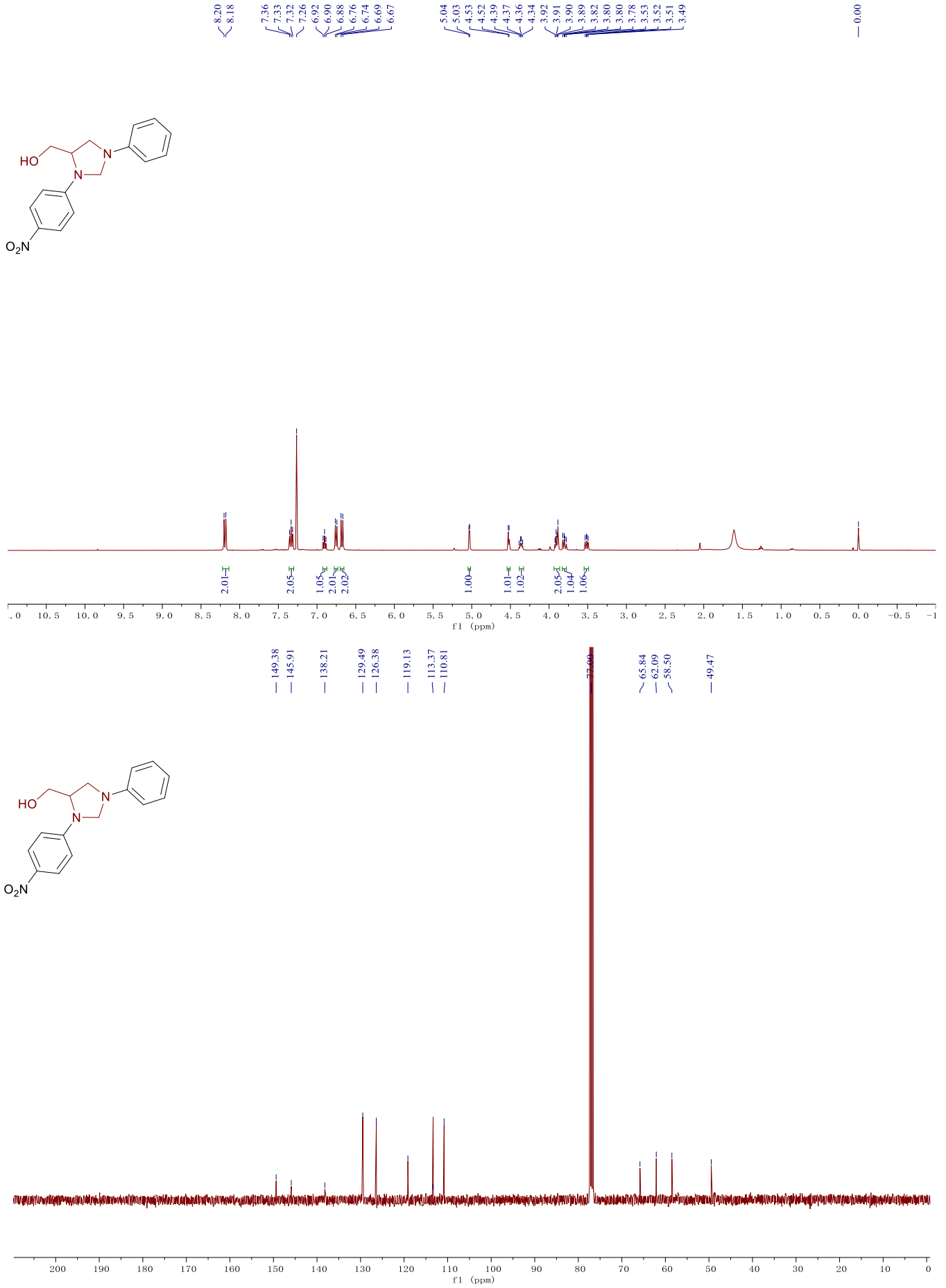
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ai



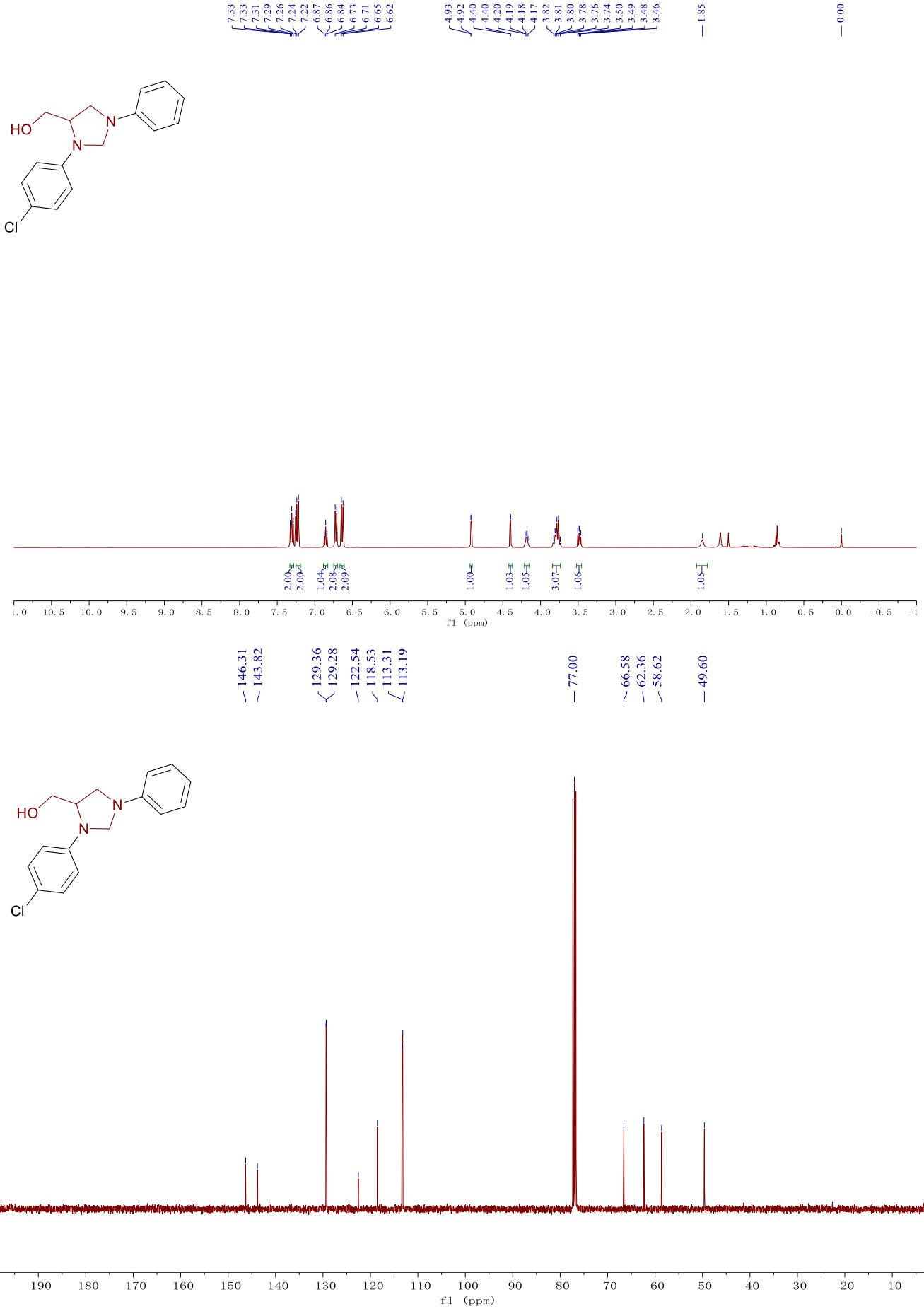
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ba



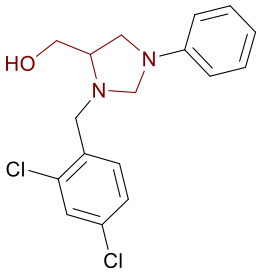
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ca

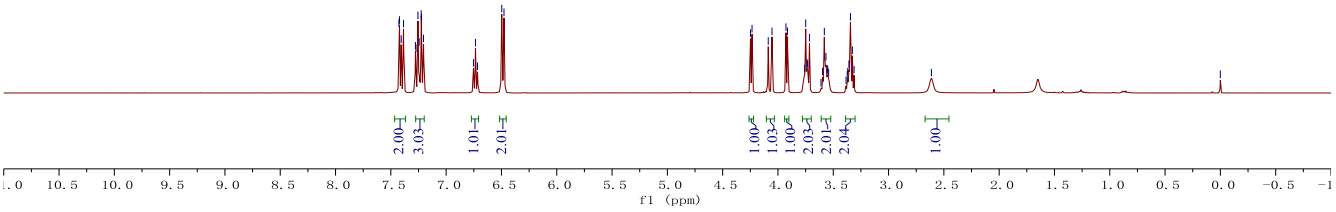


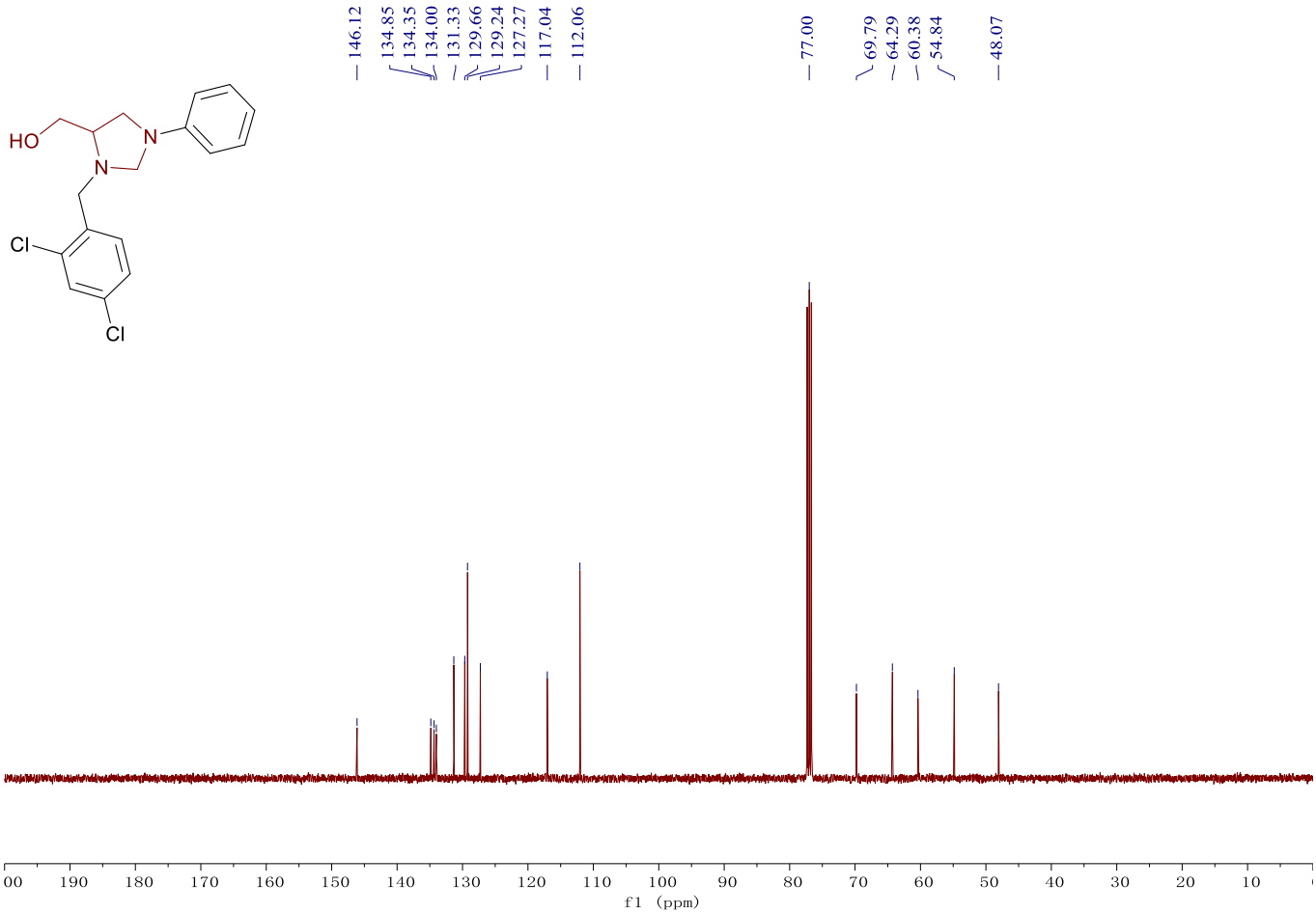
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3da



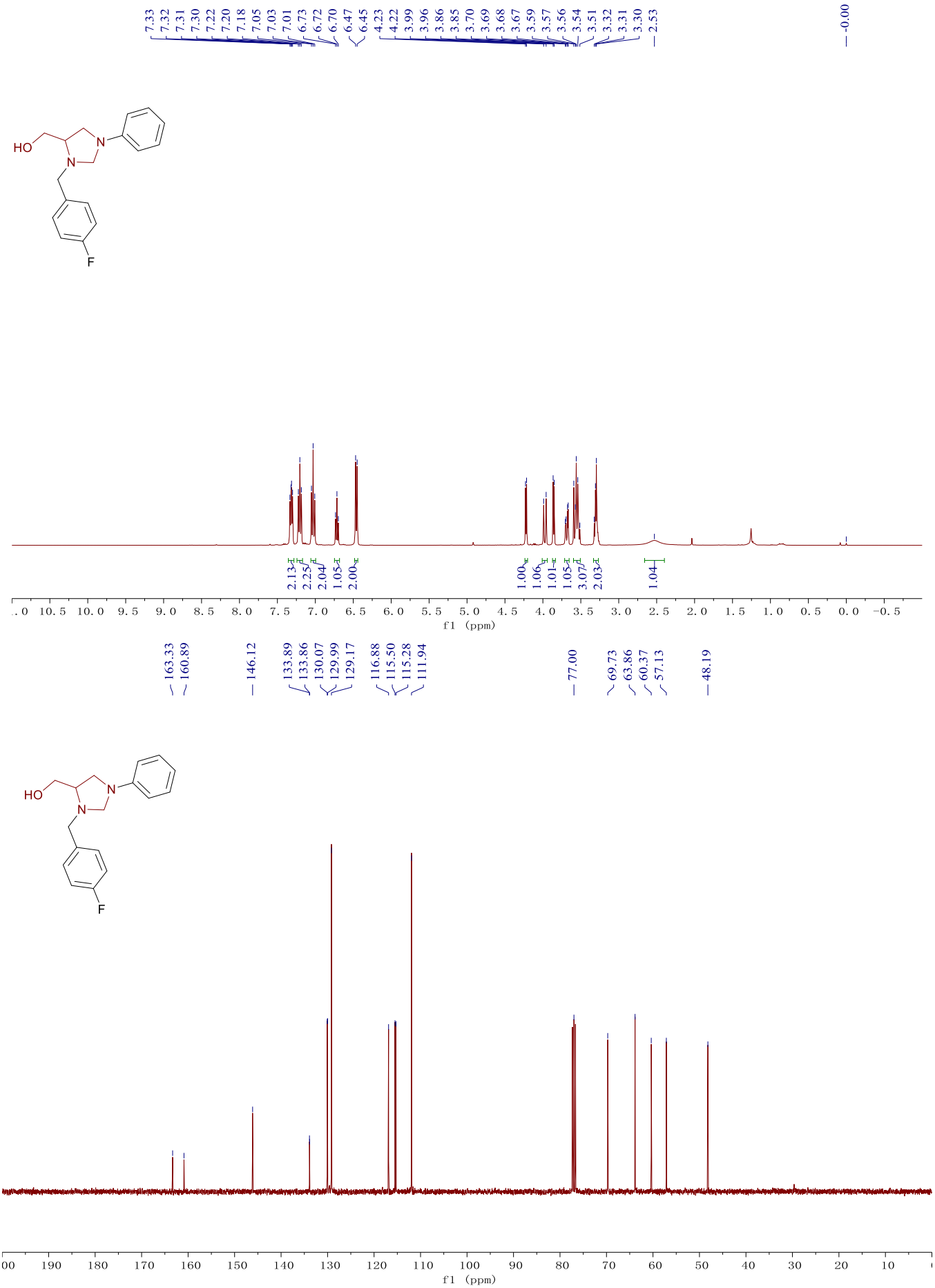
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ea



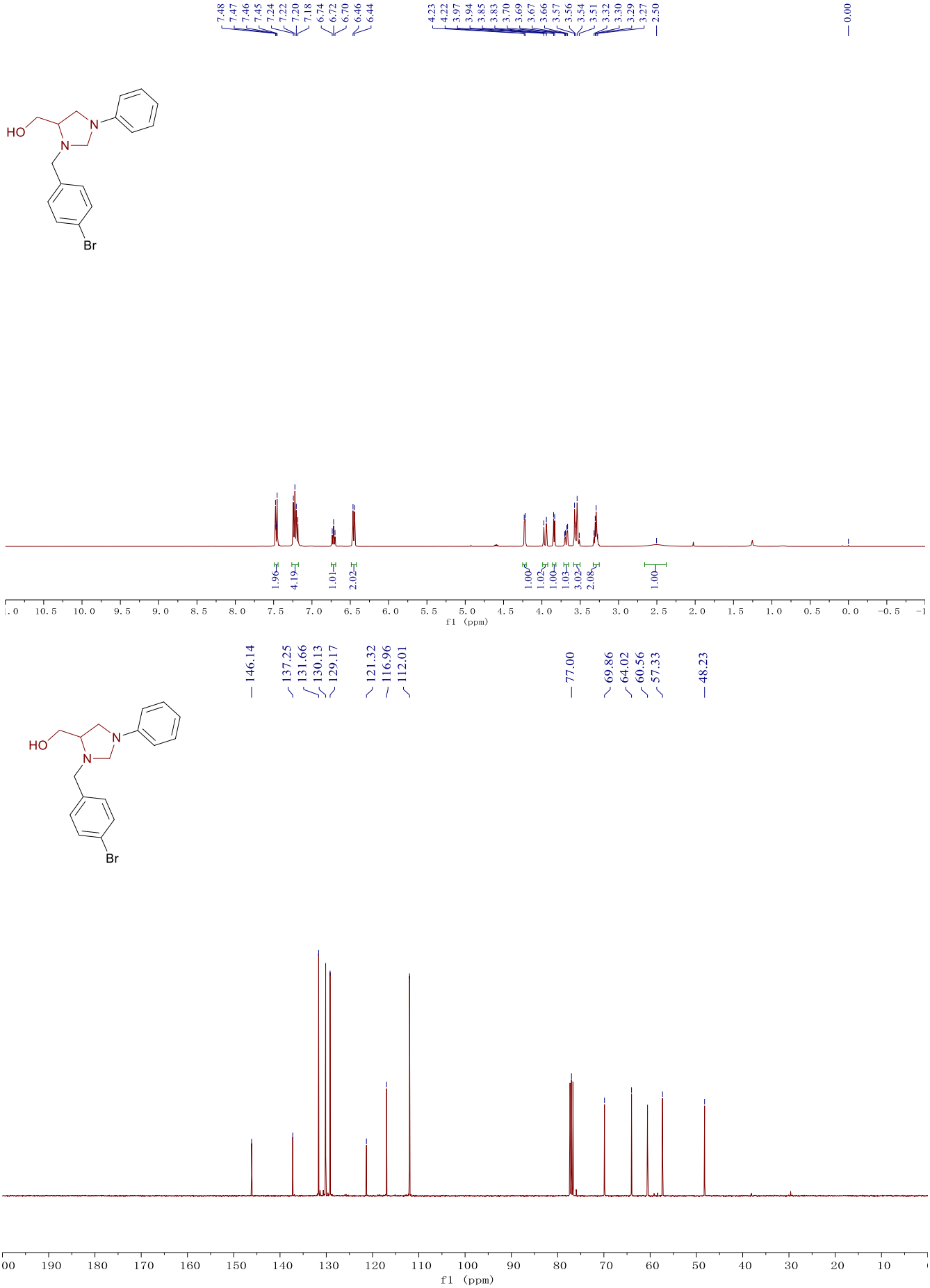




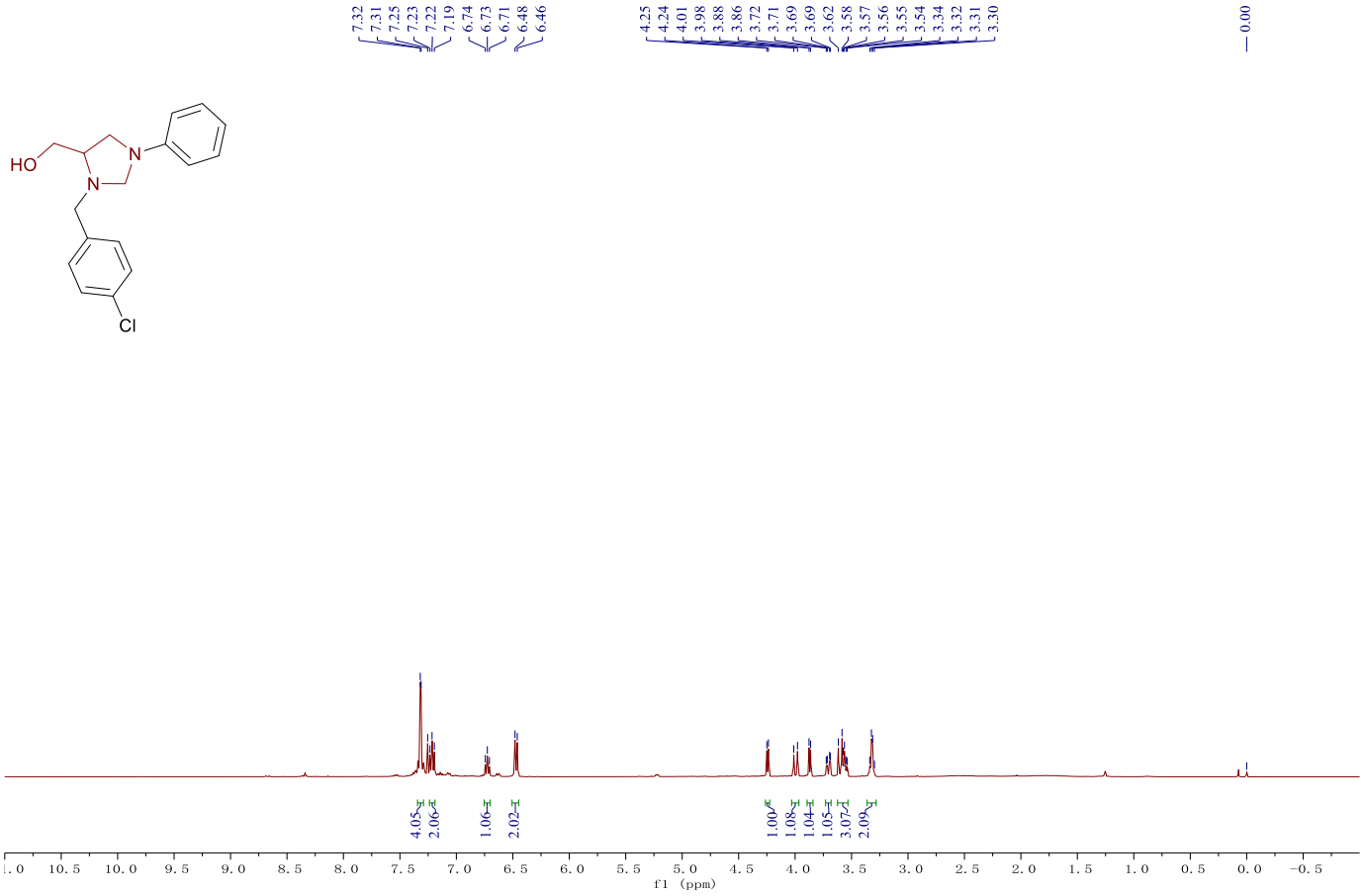
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3fa

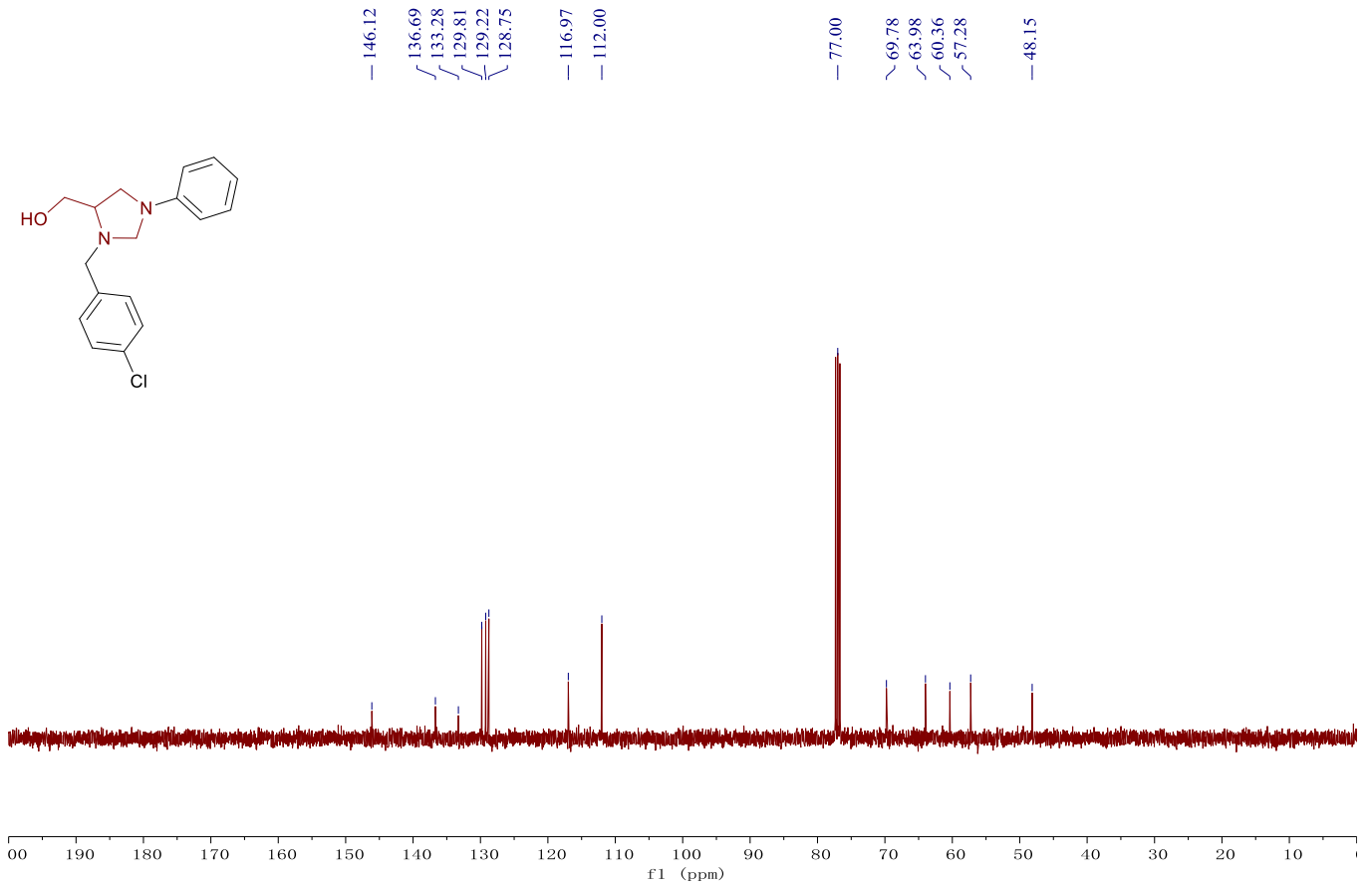


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ga

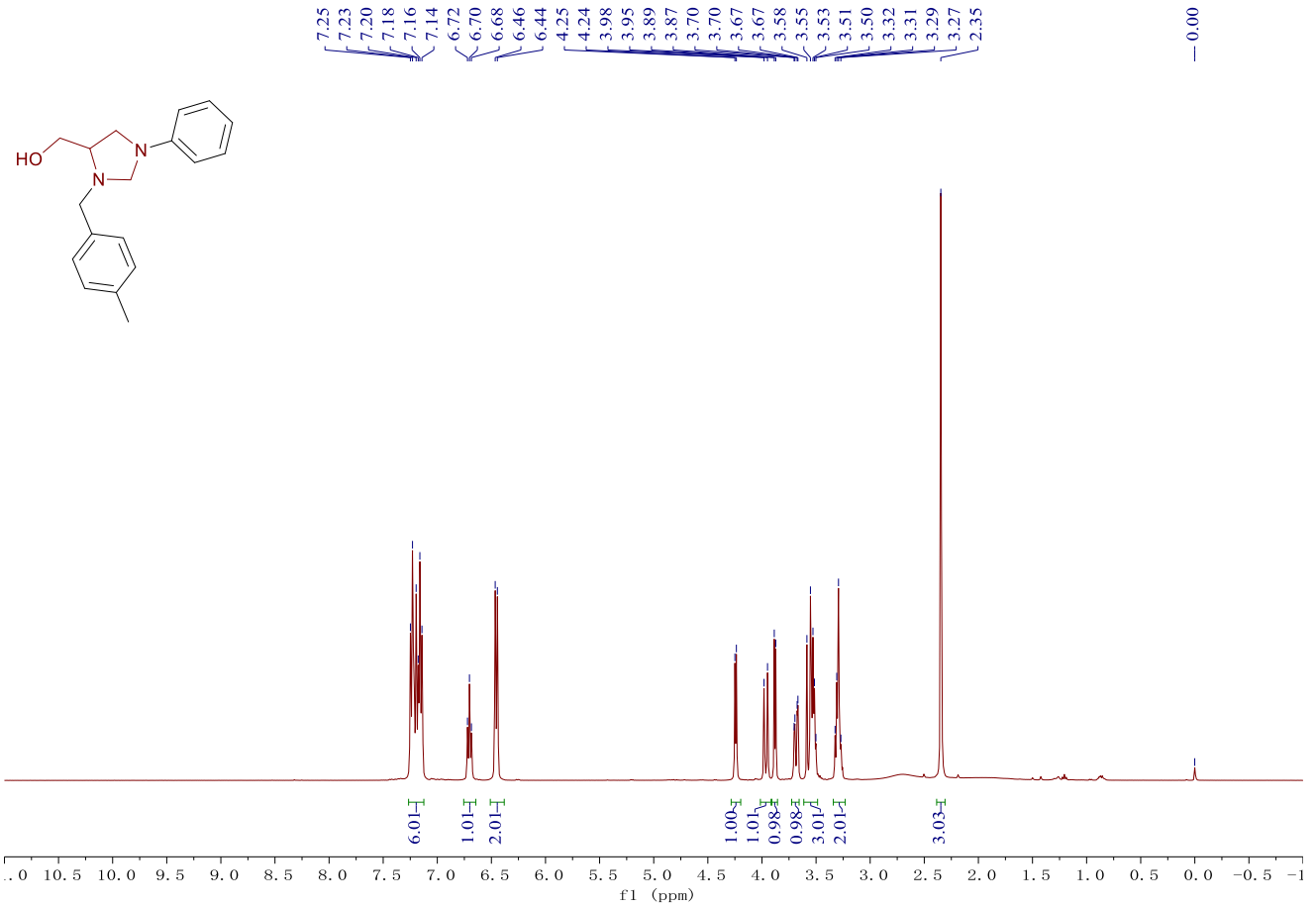


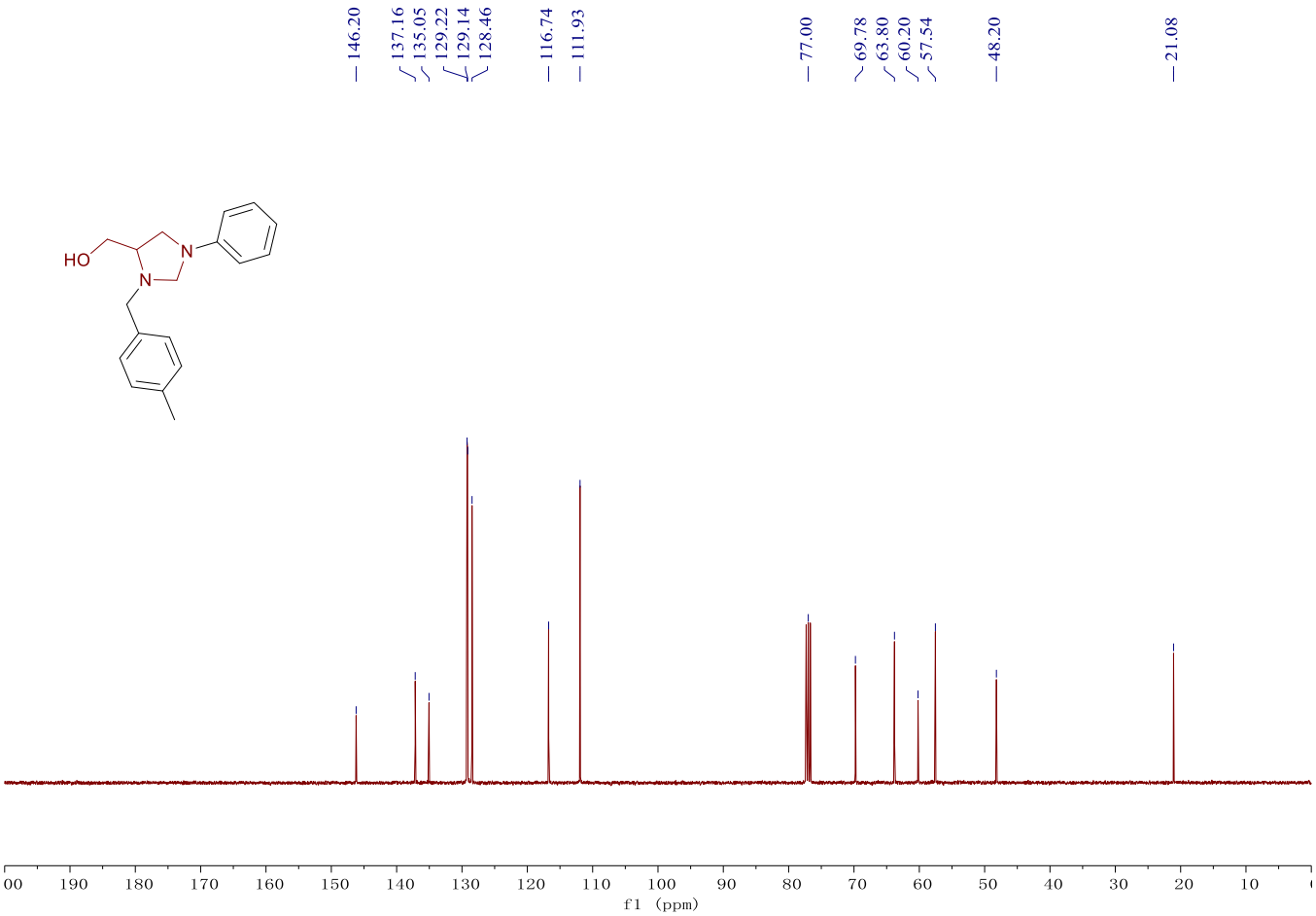
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ha



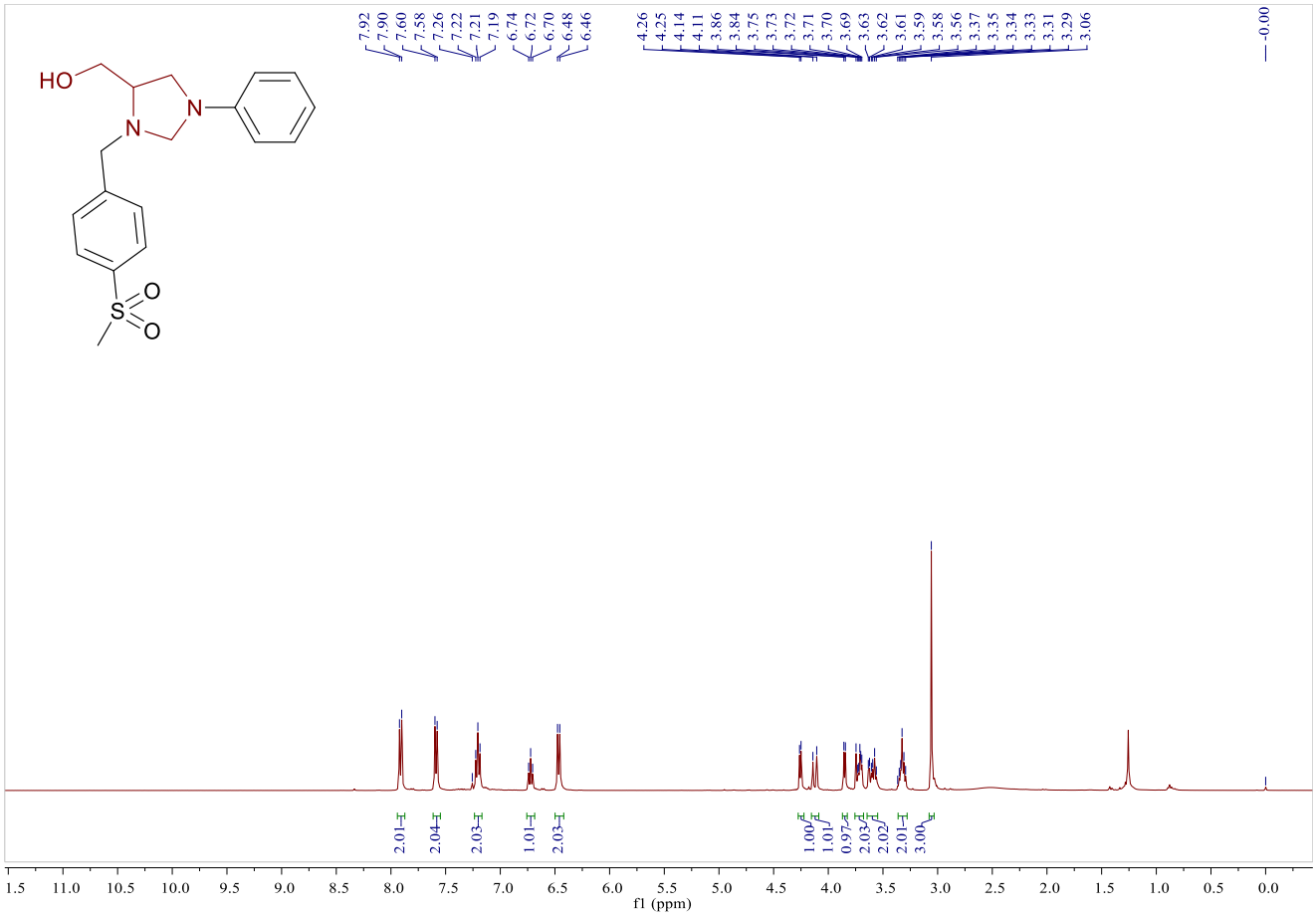


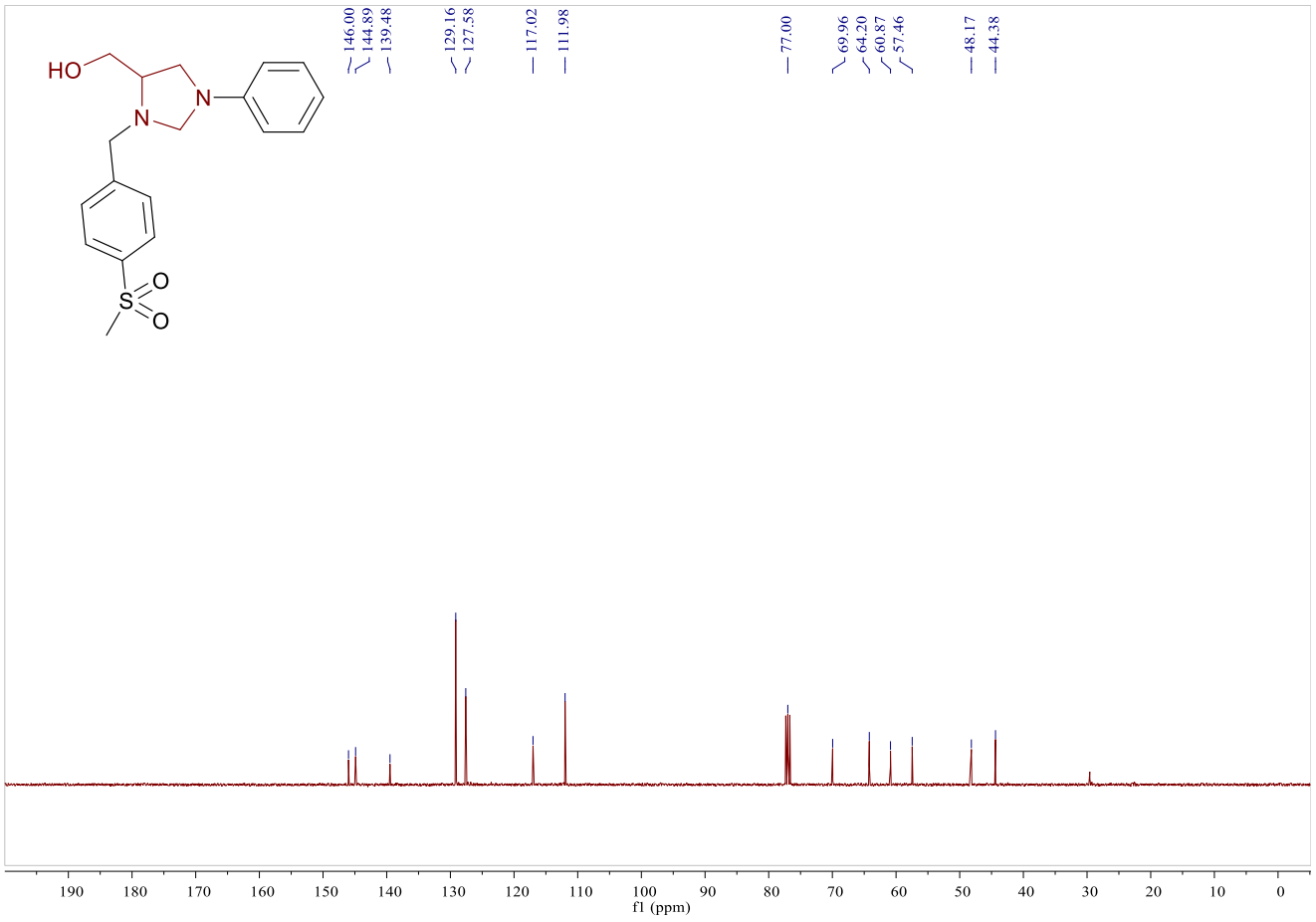
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ia



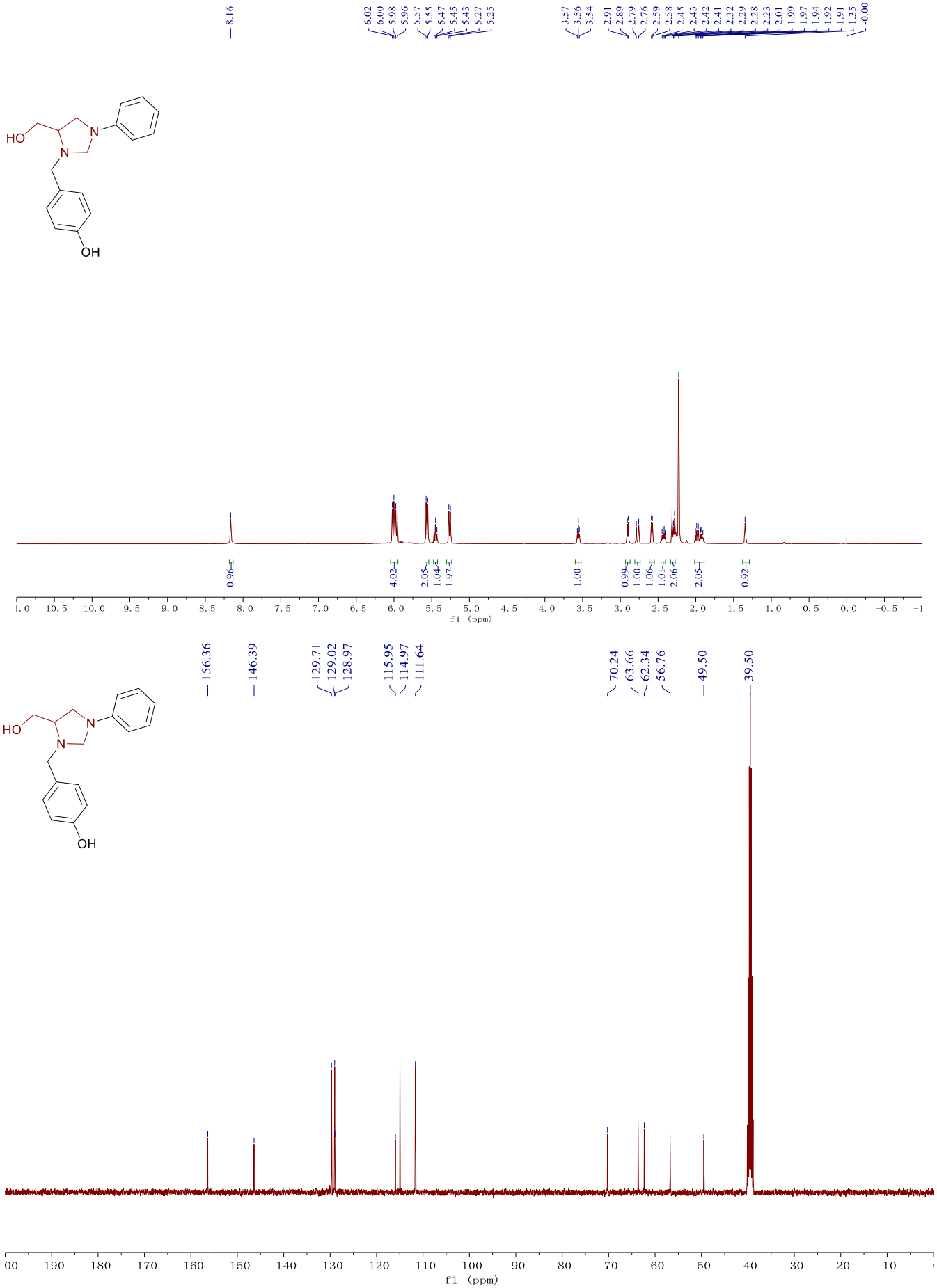


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ja

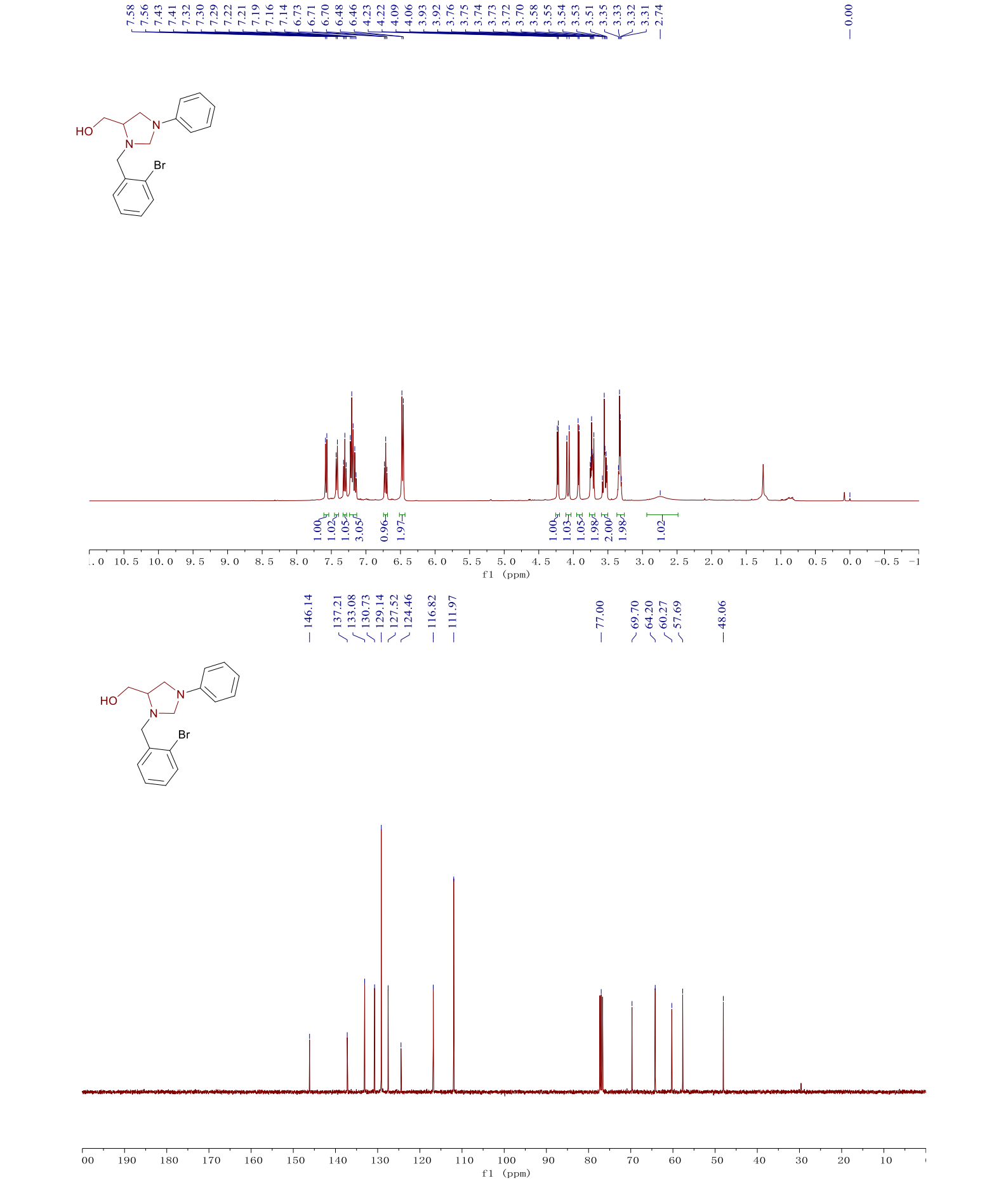




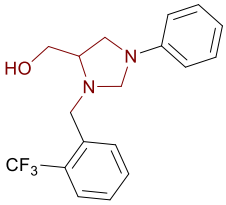
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ka

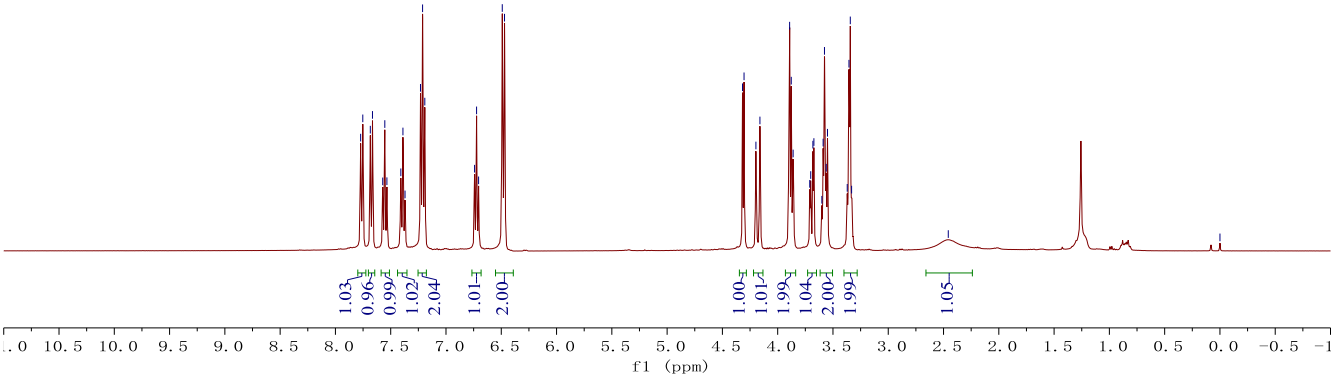


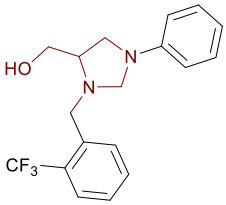
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3la

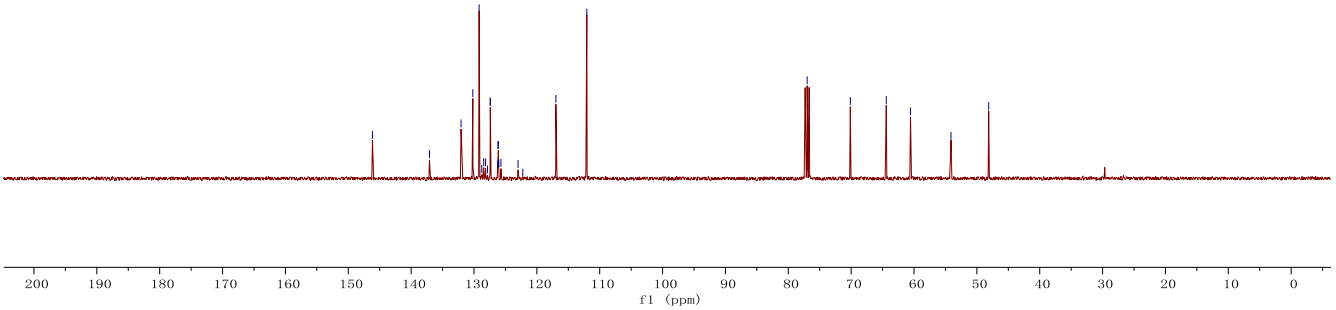


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ma

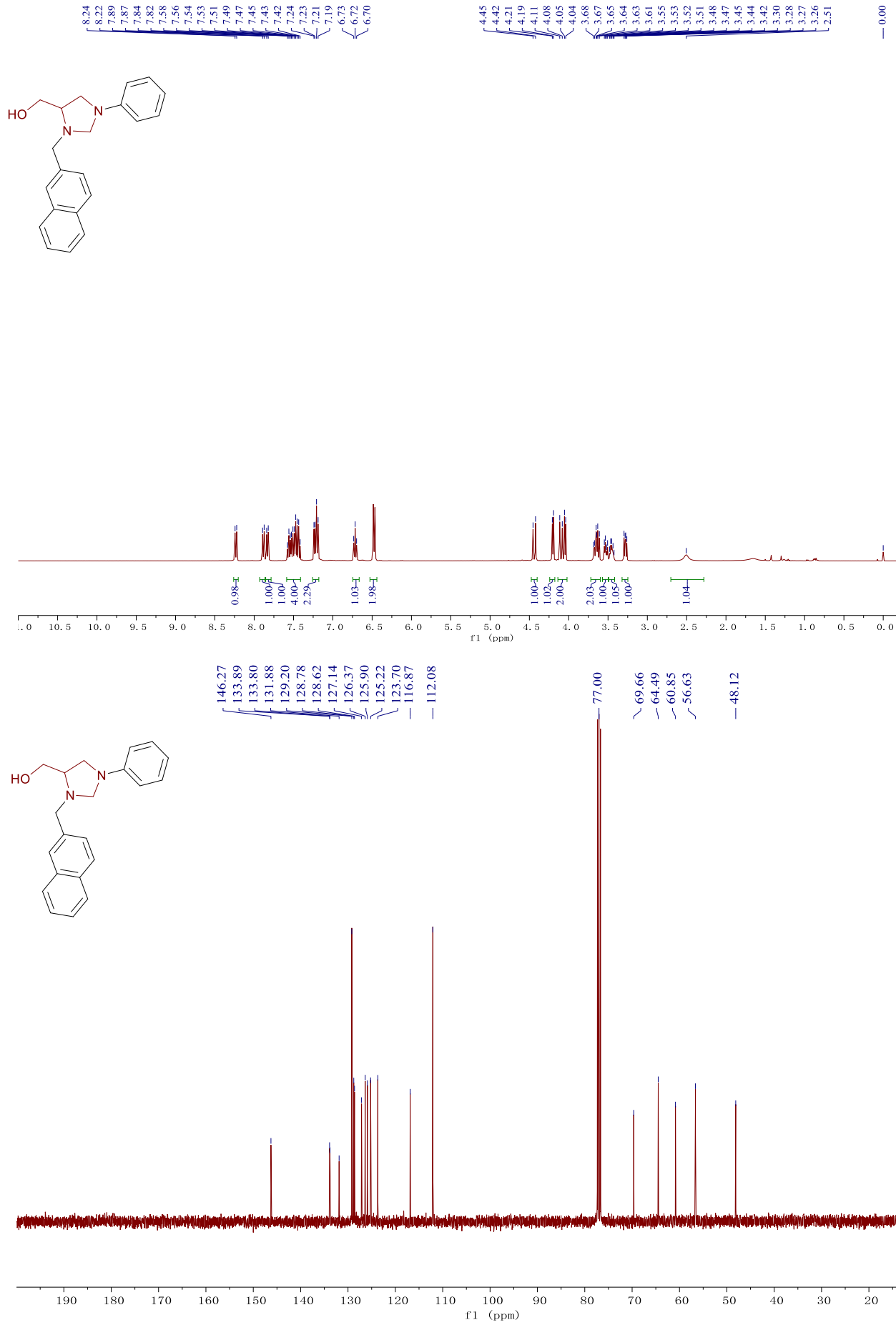




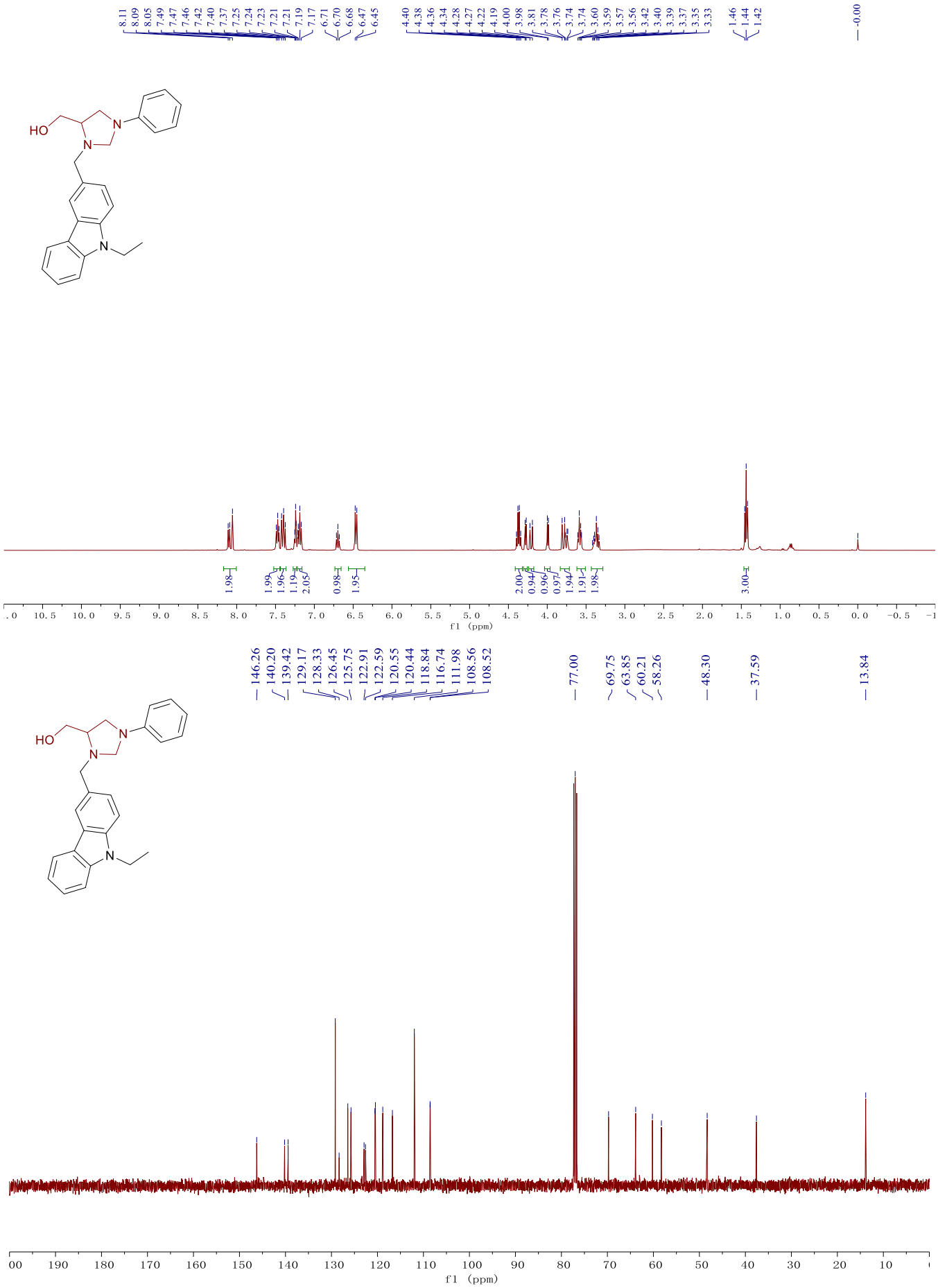




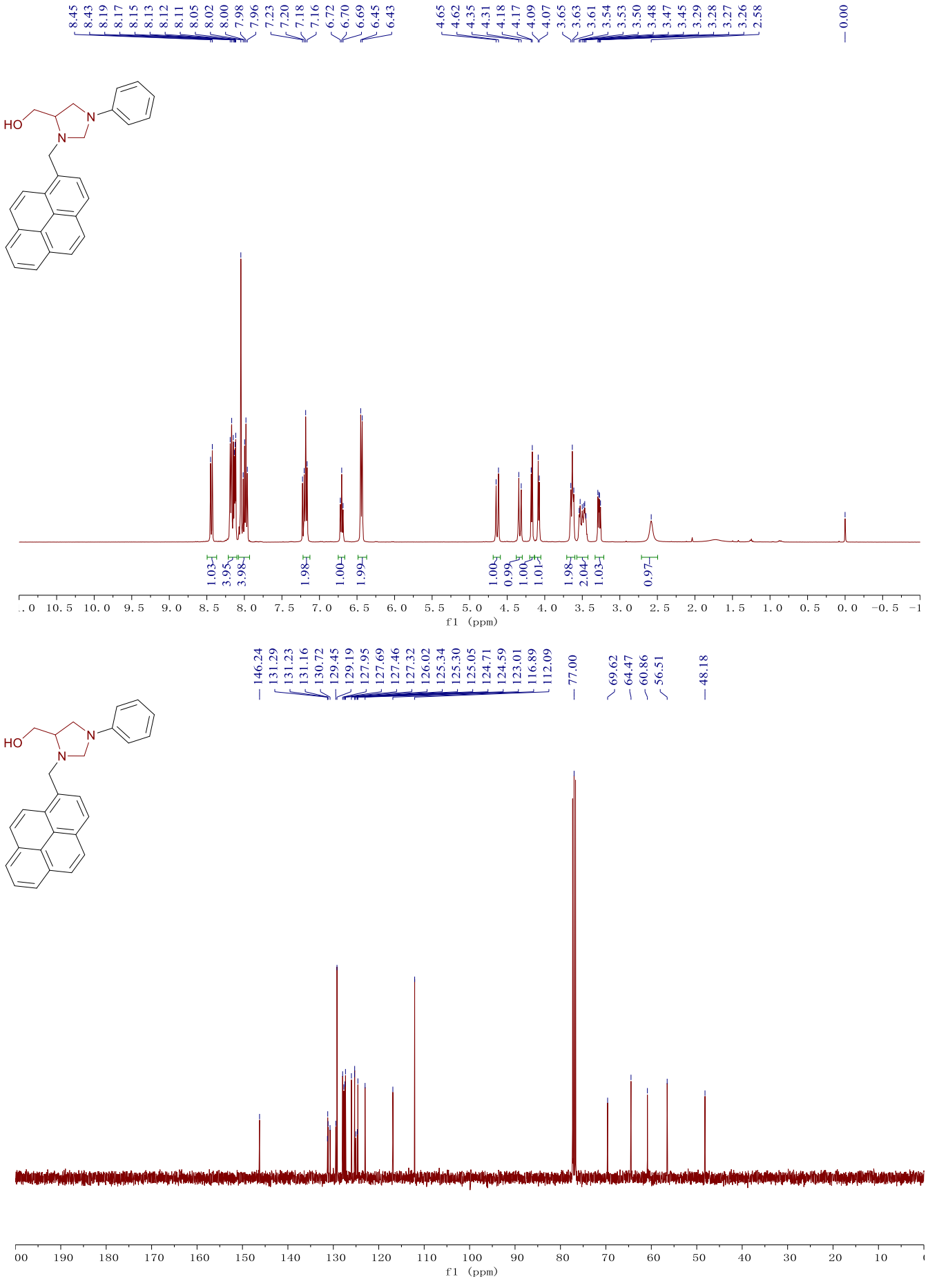
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3na



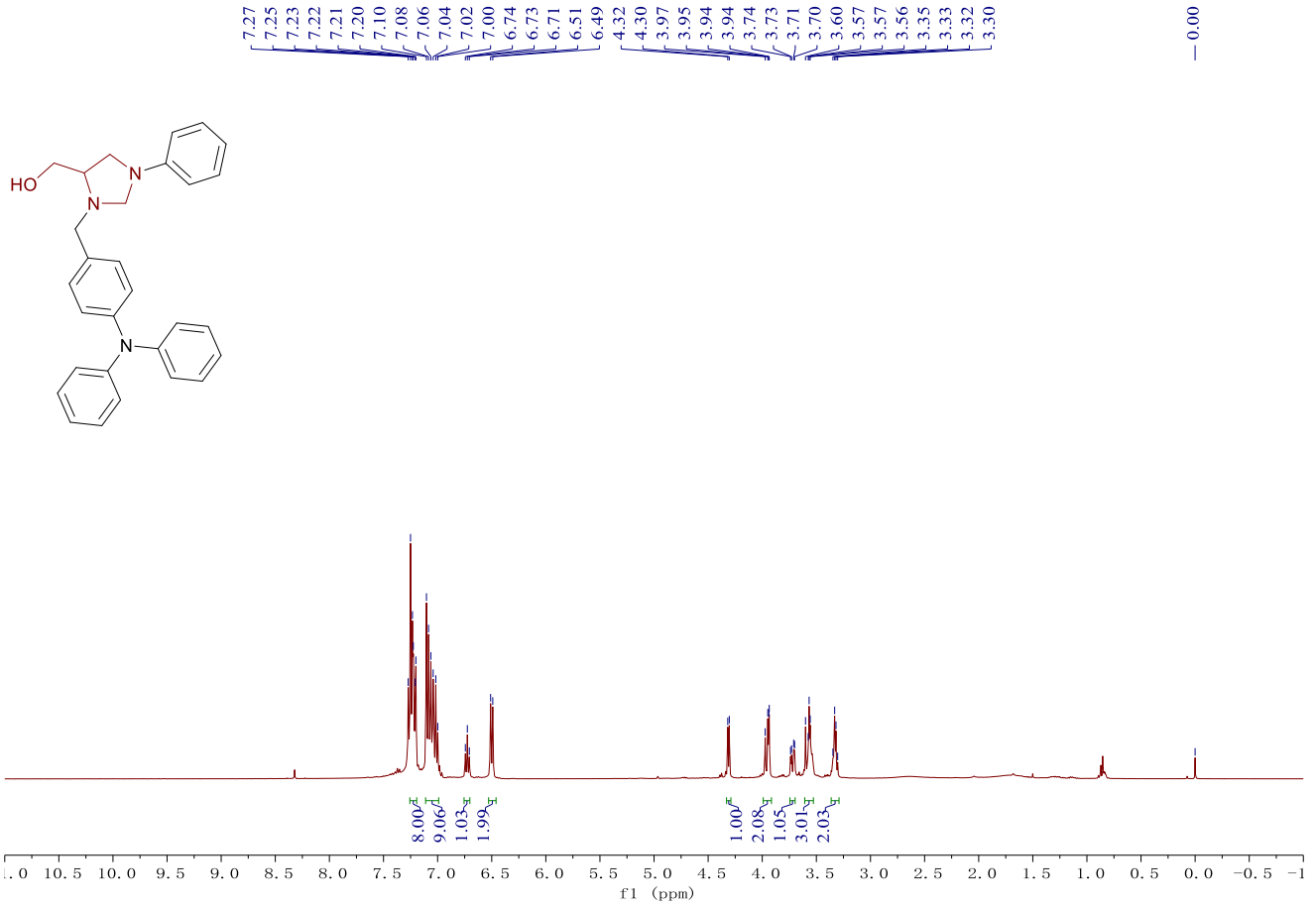
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3oa

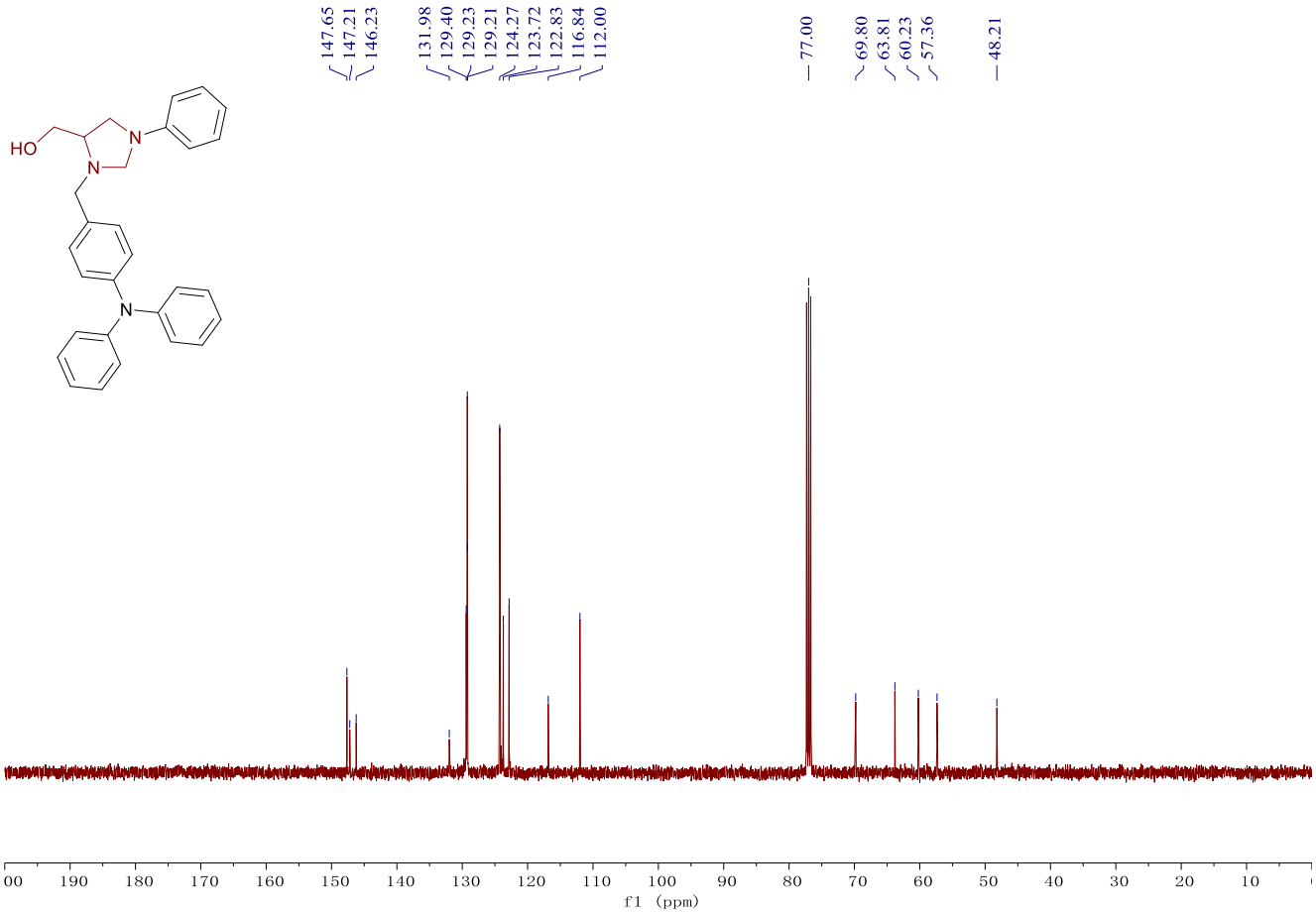


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3pa

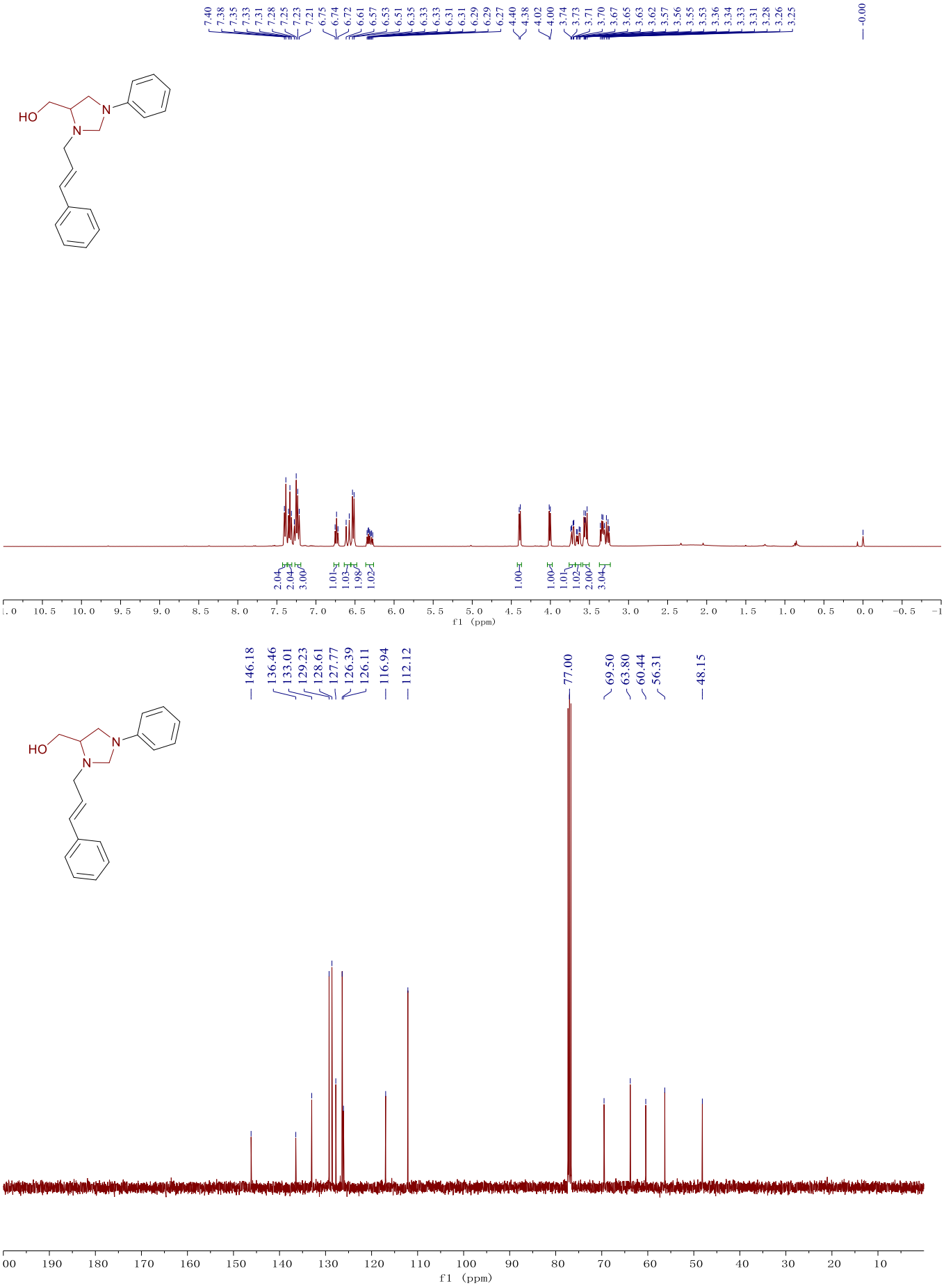


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3qa

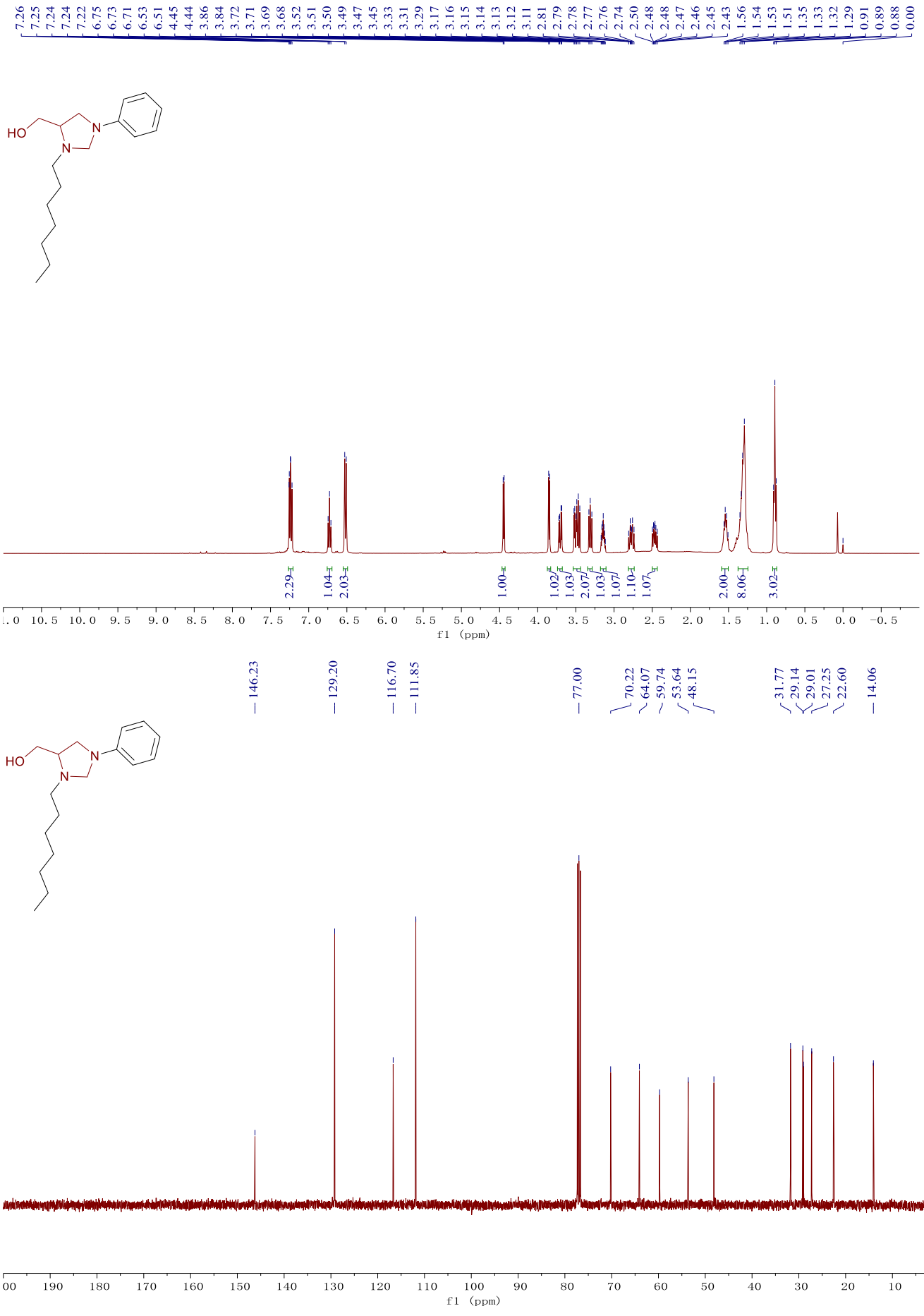




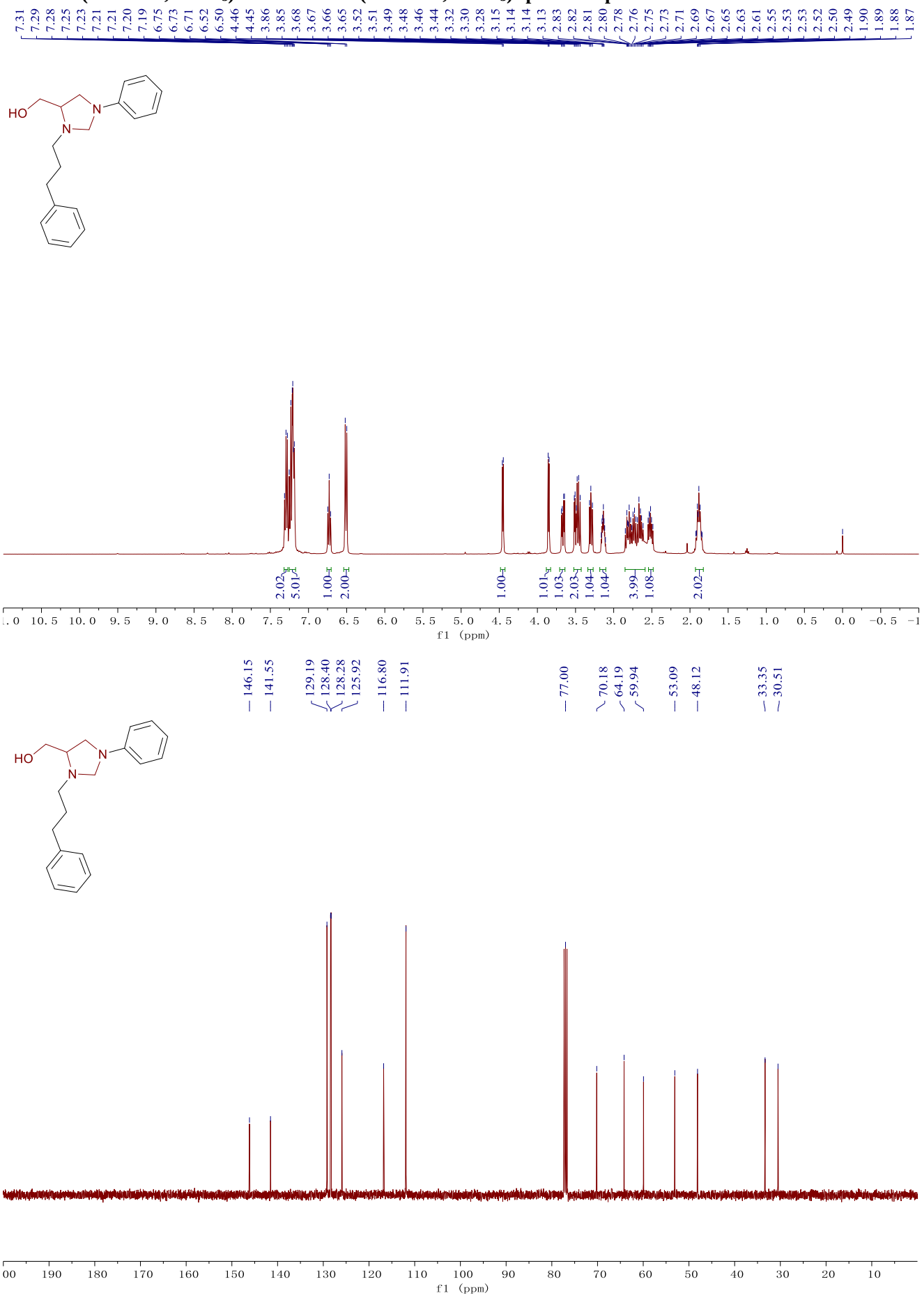
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ra



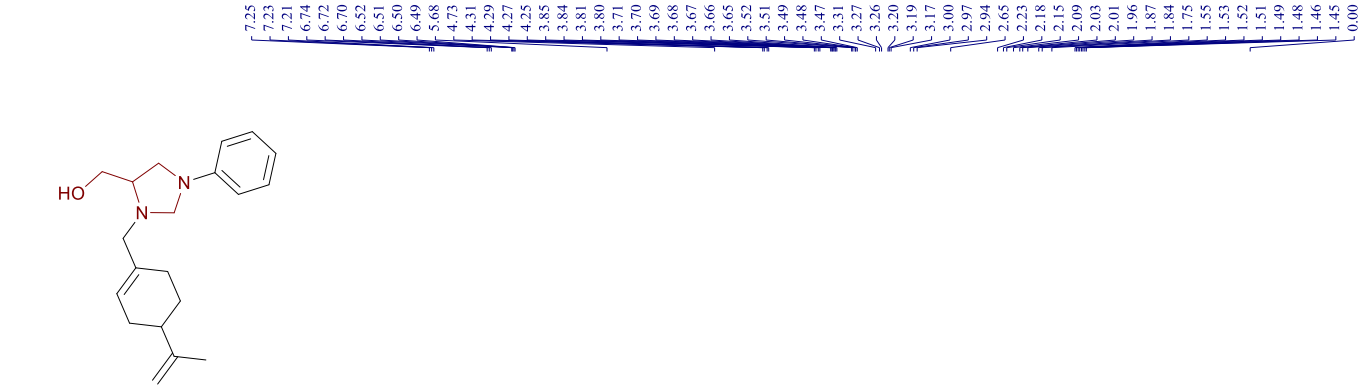
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3sa

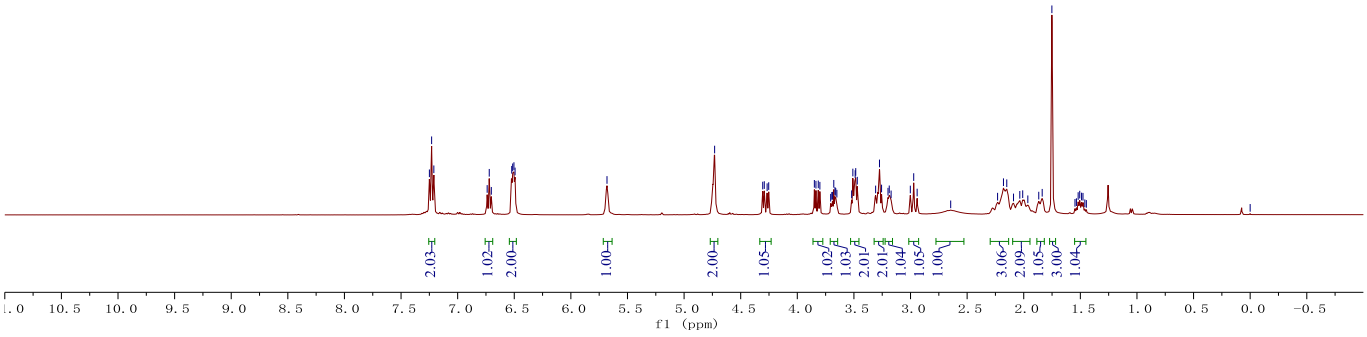


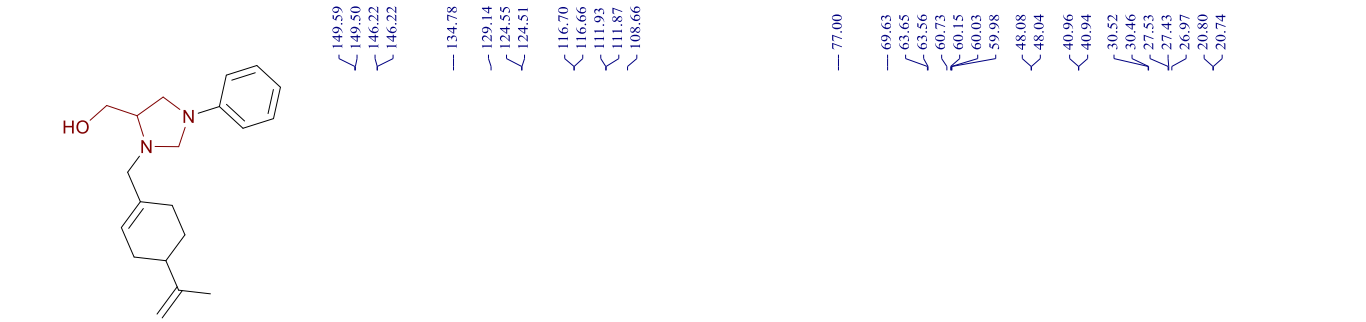
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ta

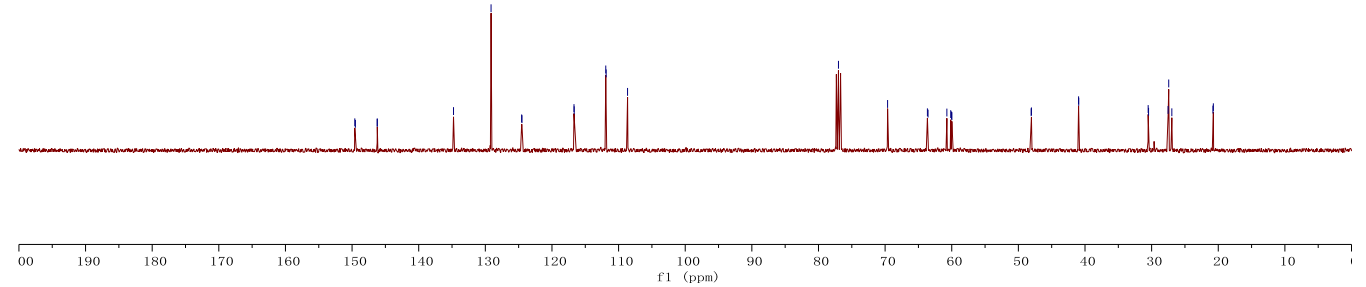


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3ua

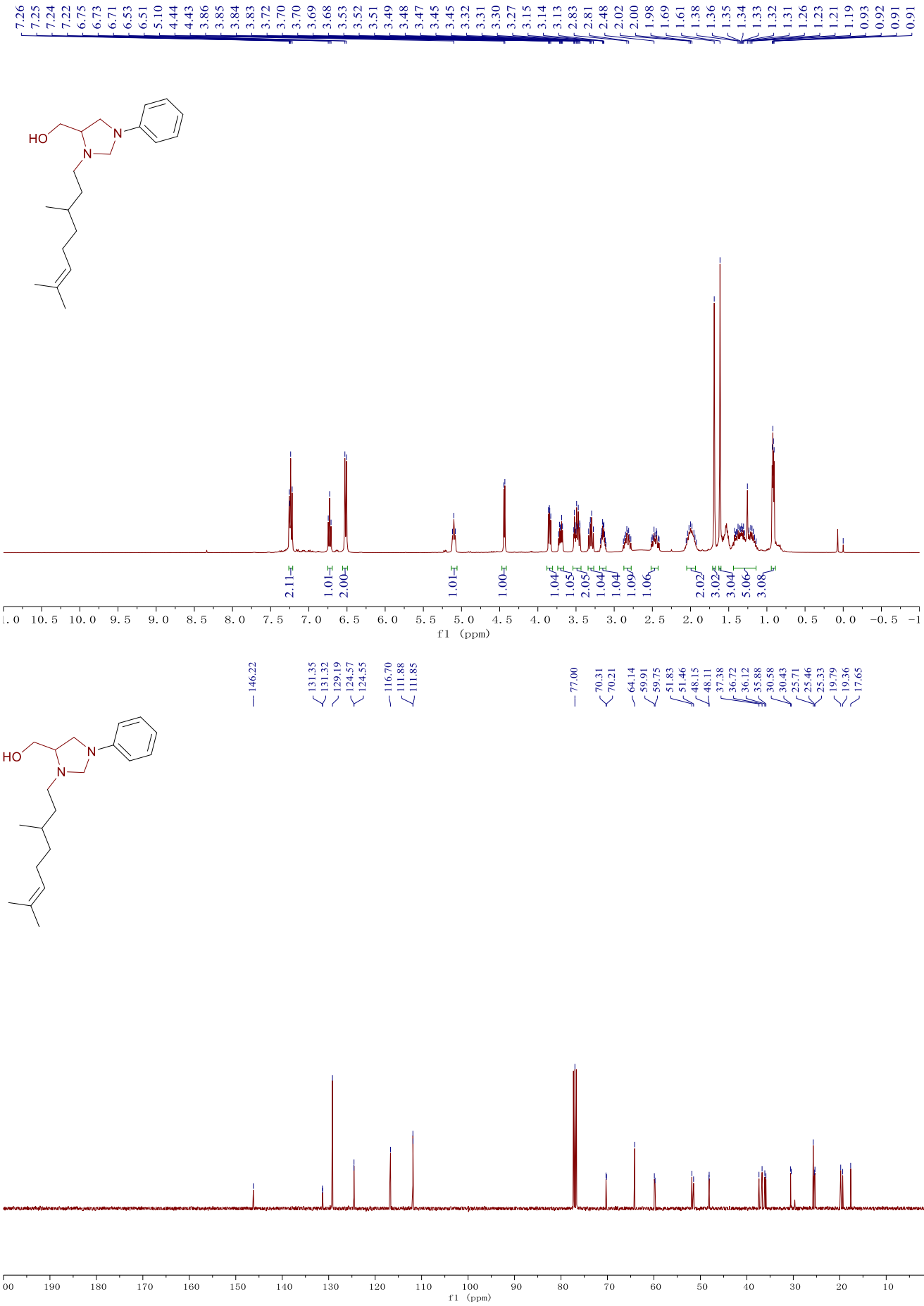




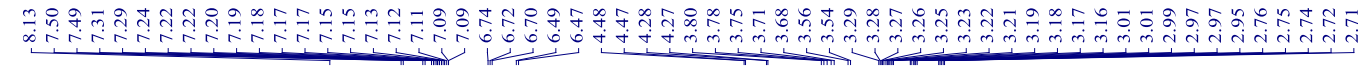


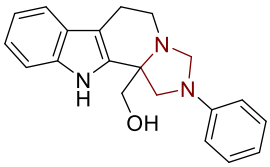


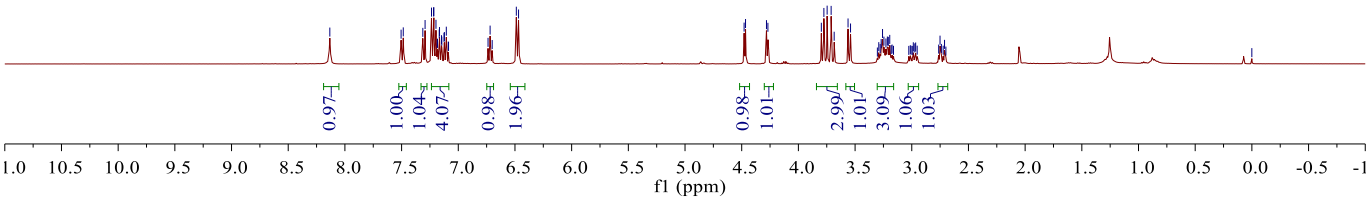
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3va

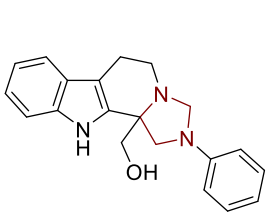


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 3wa

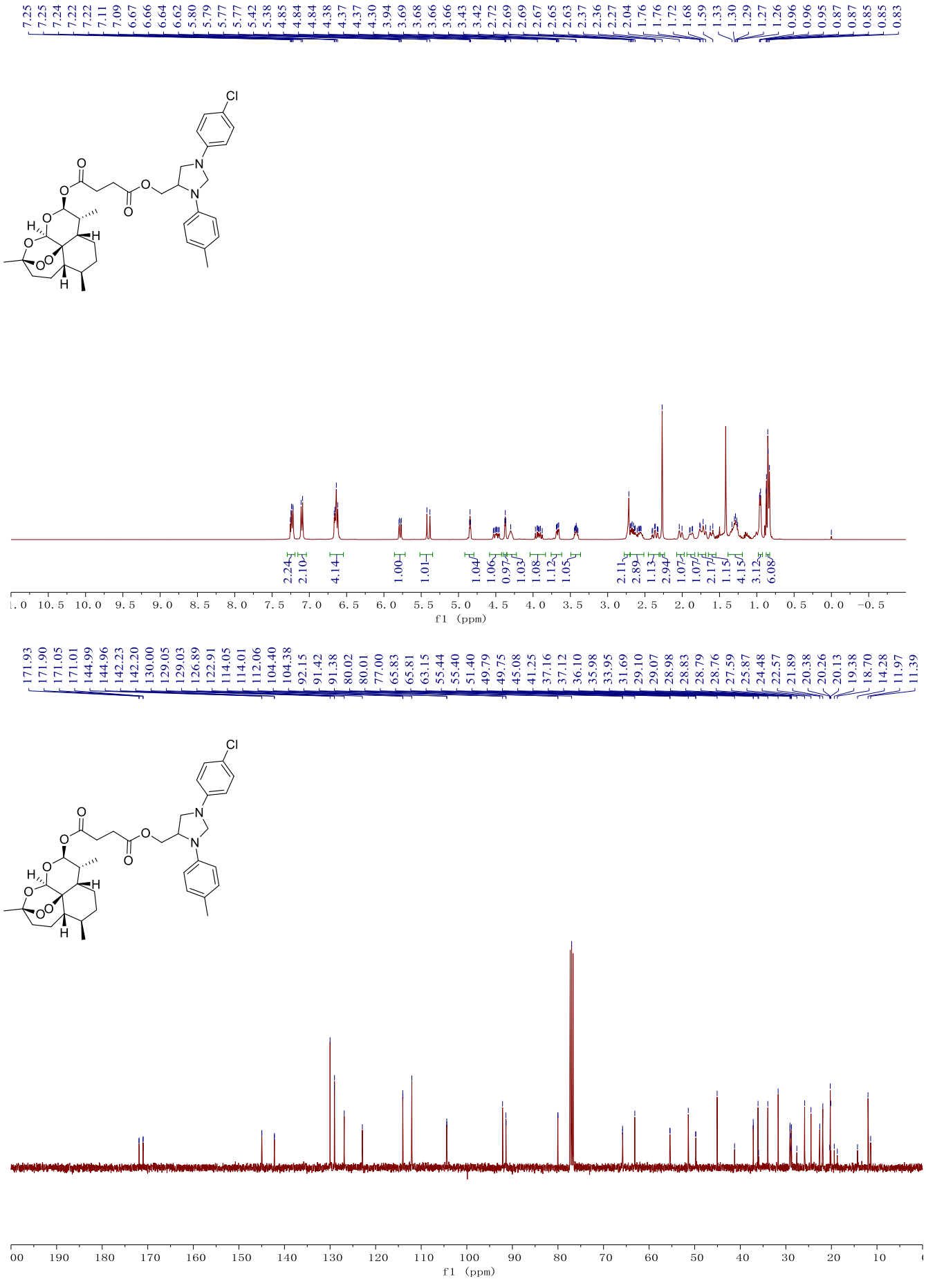




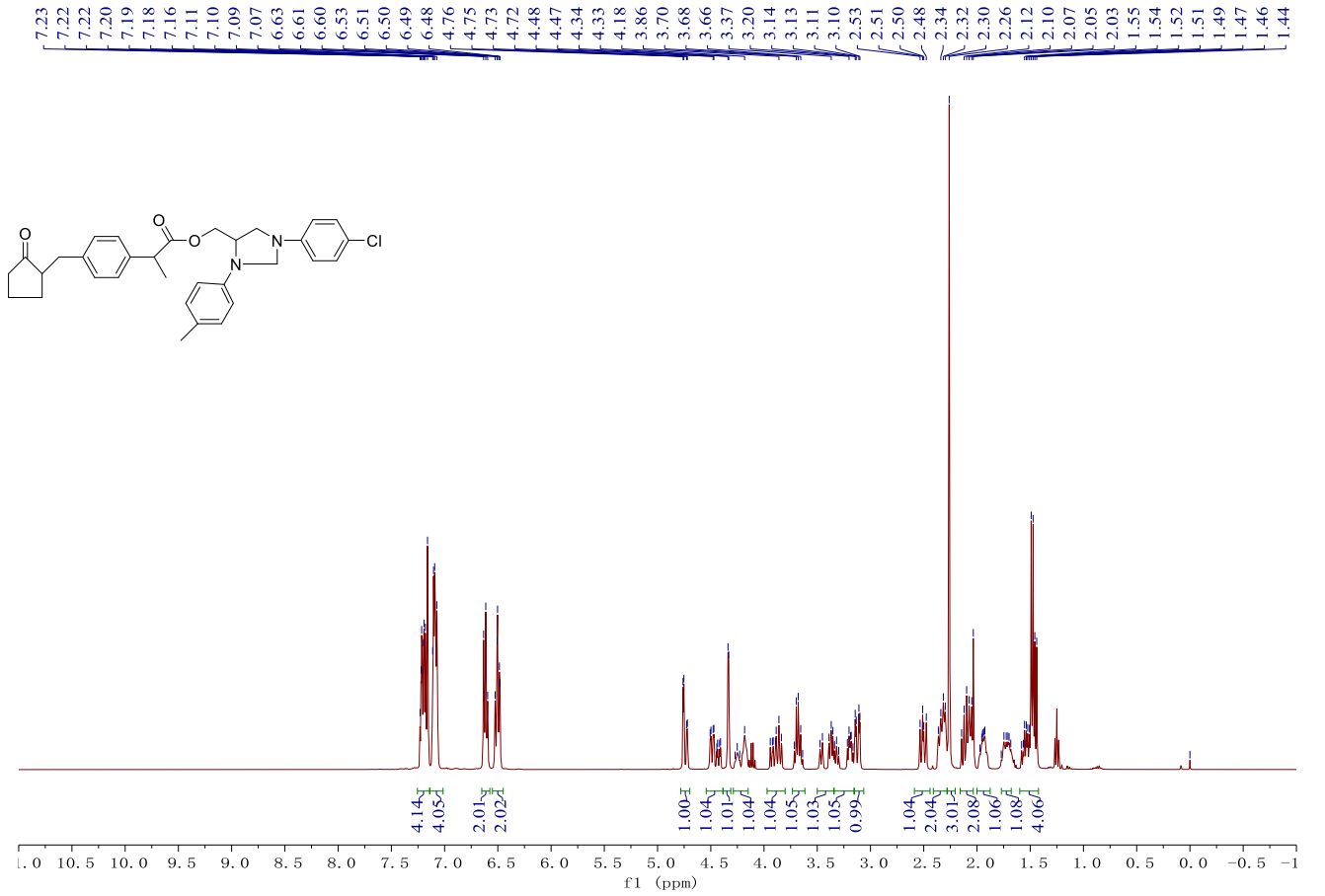


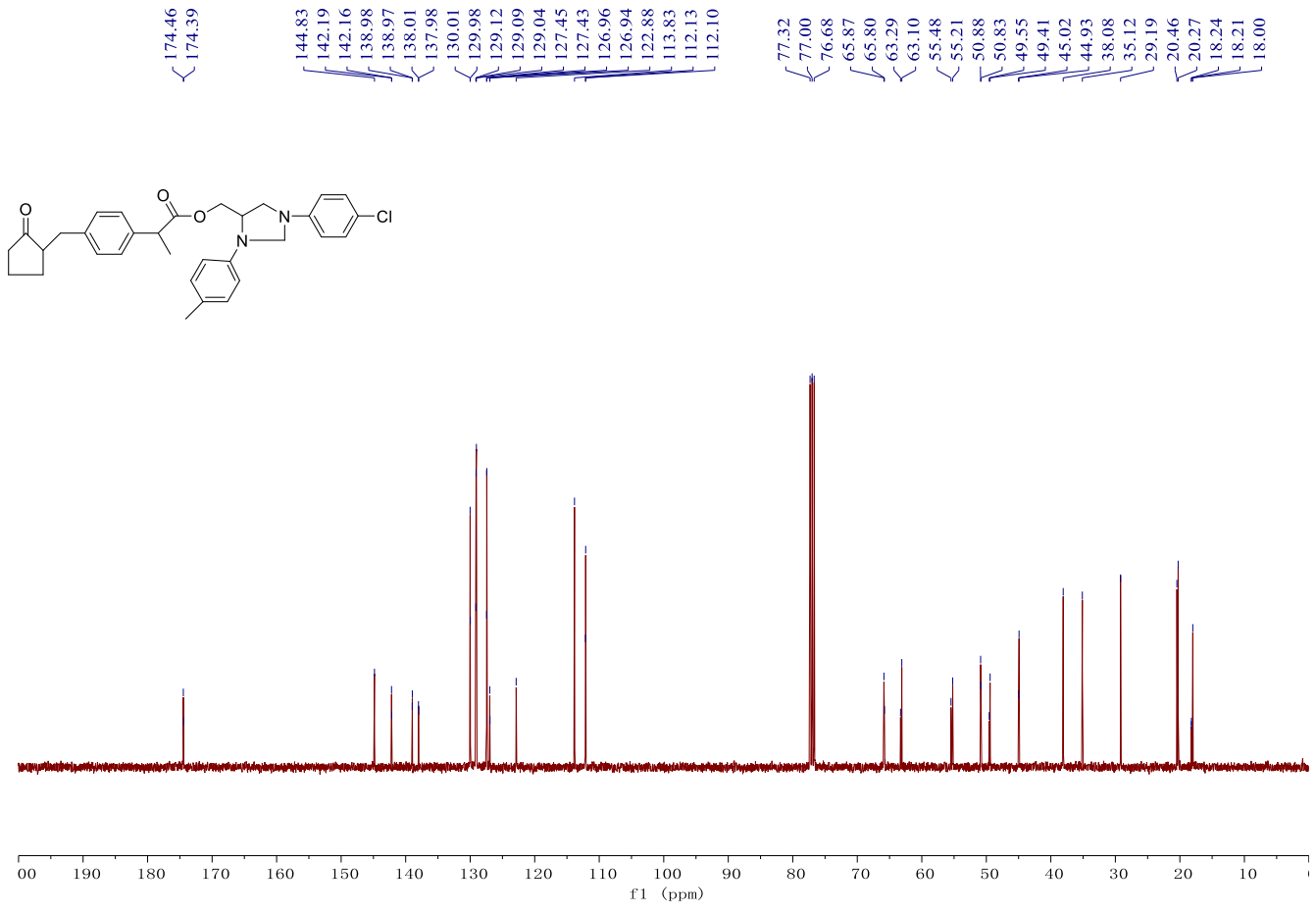


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 5aa

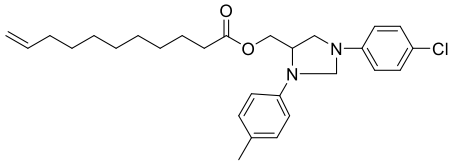


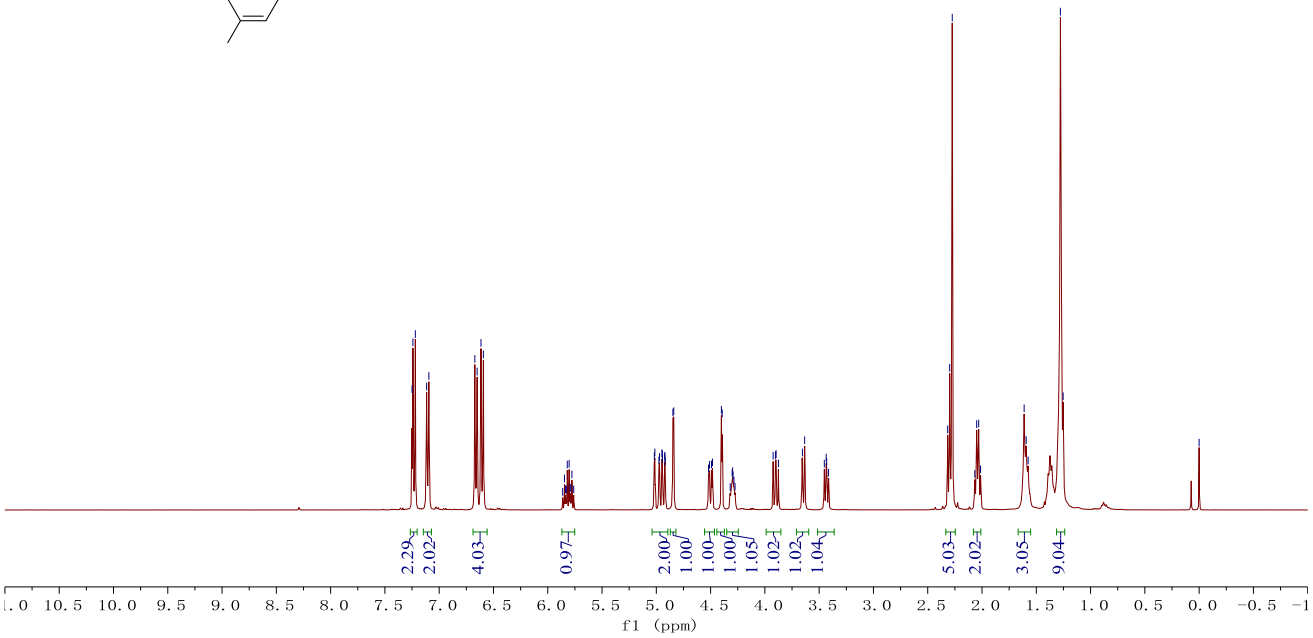
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 5ab

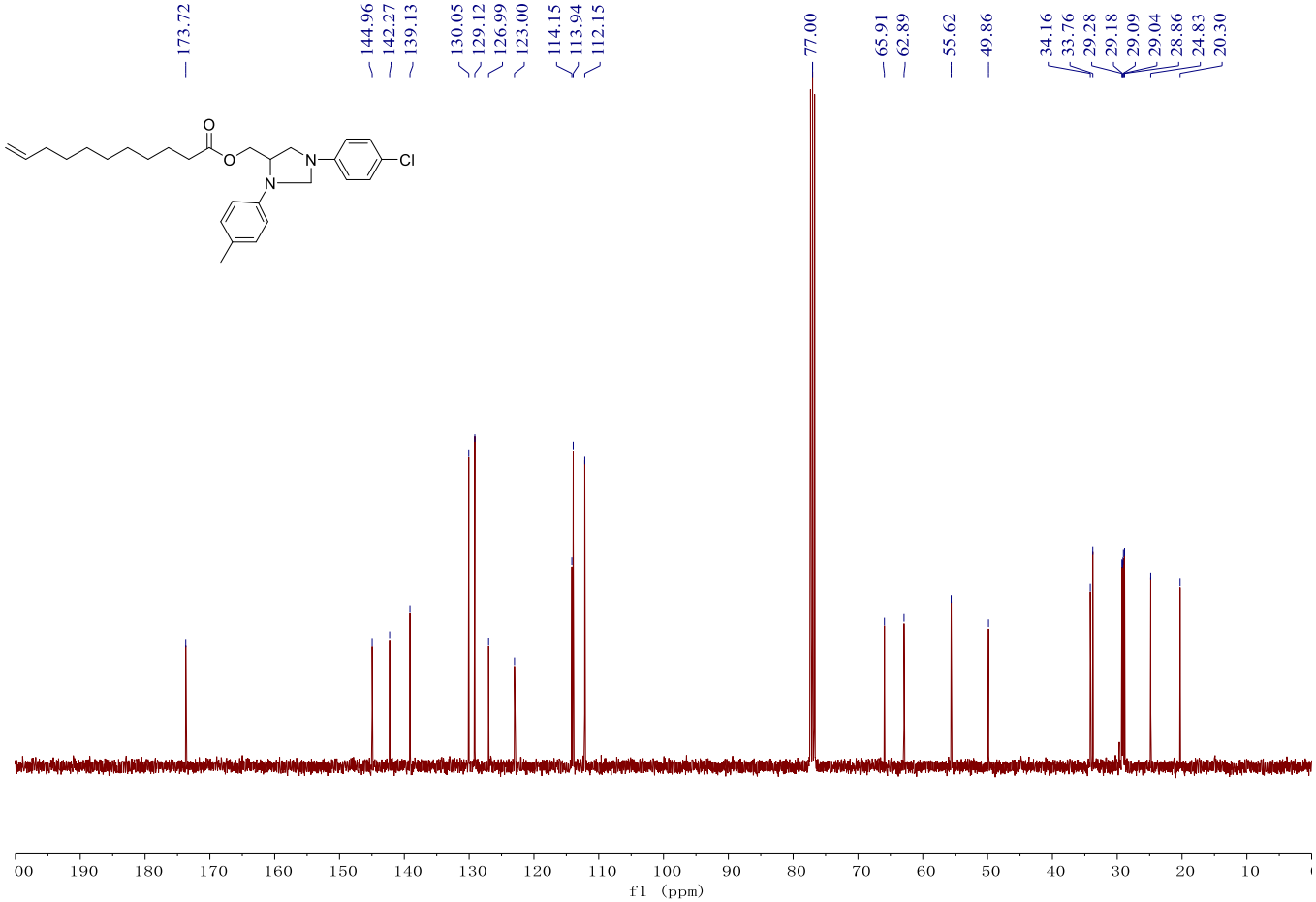




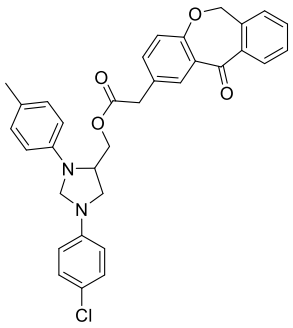
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 5ac

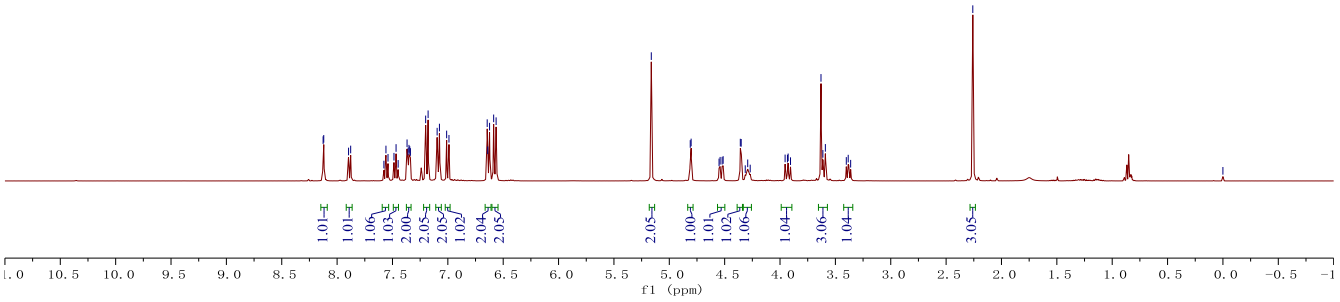


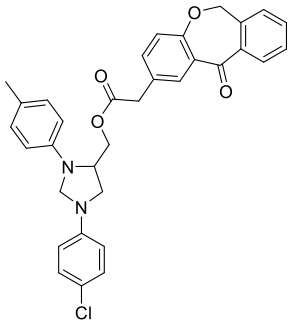


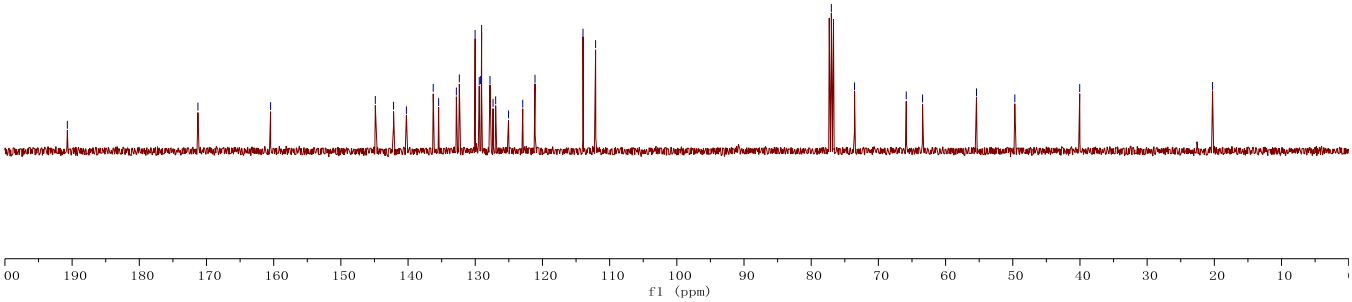


^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 5ad

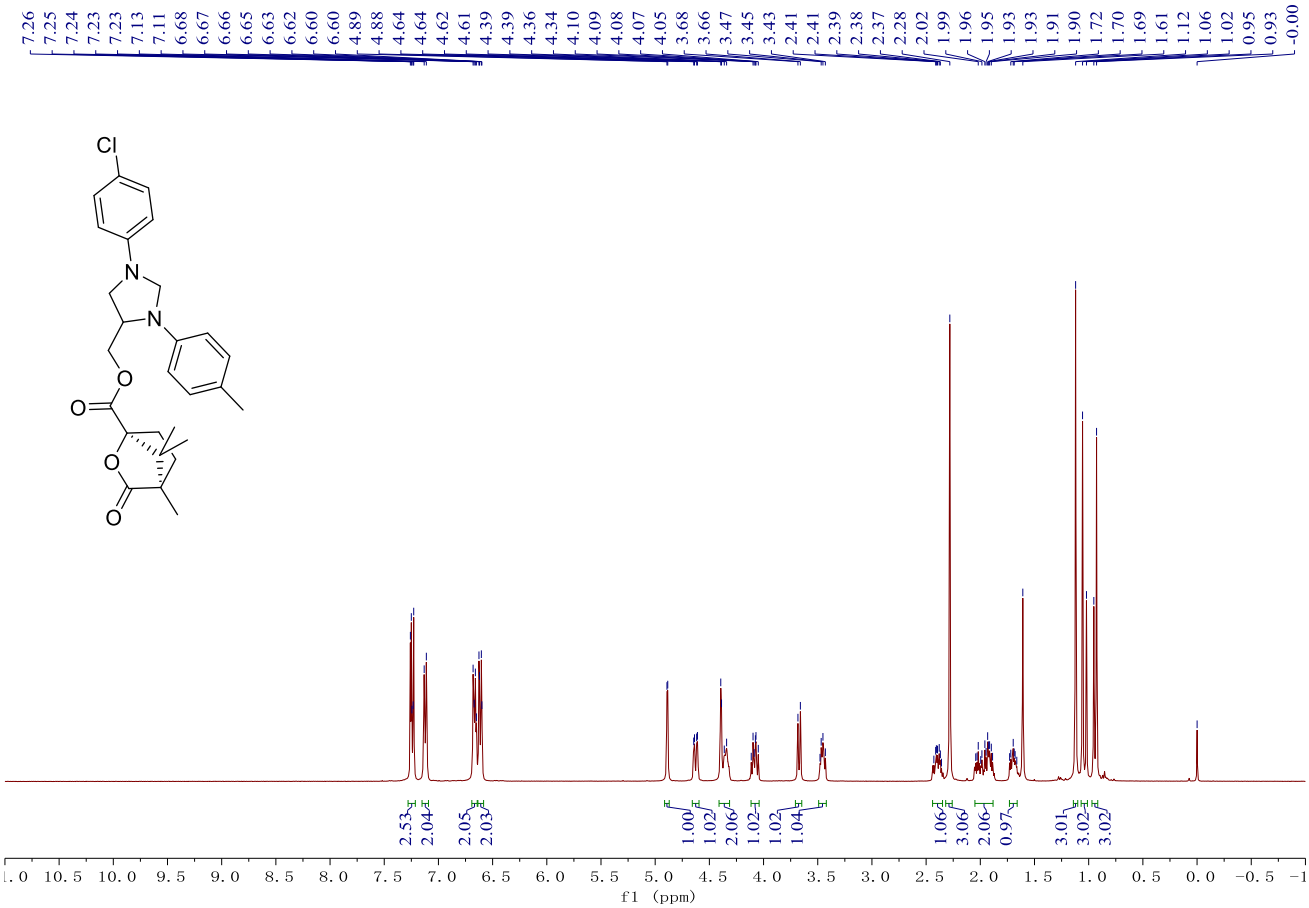


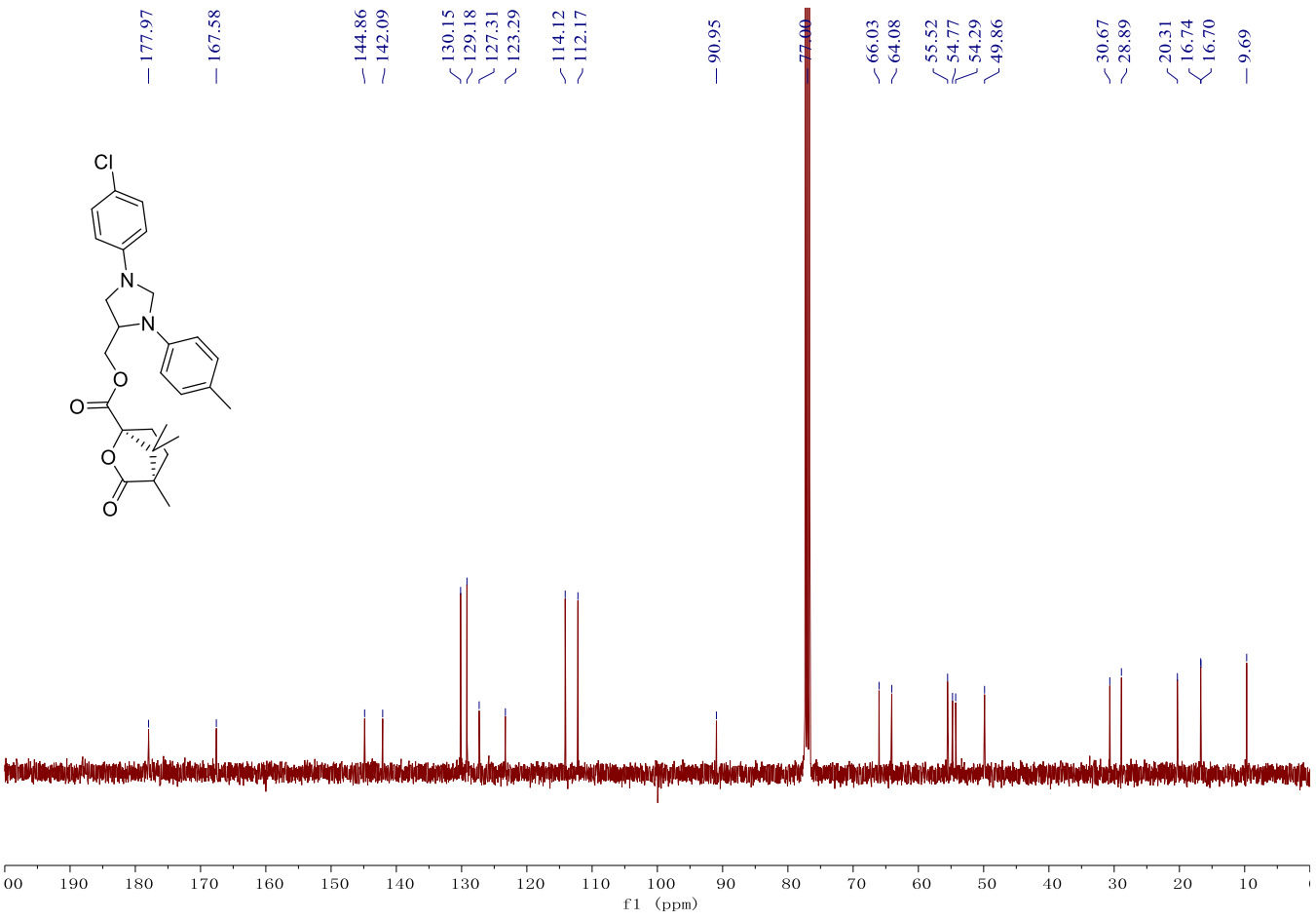




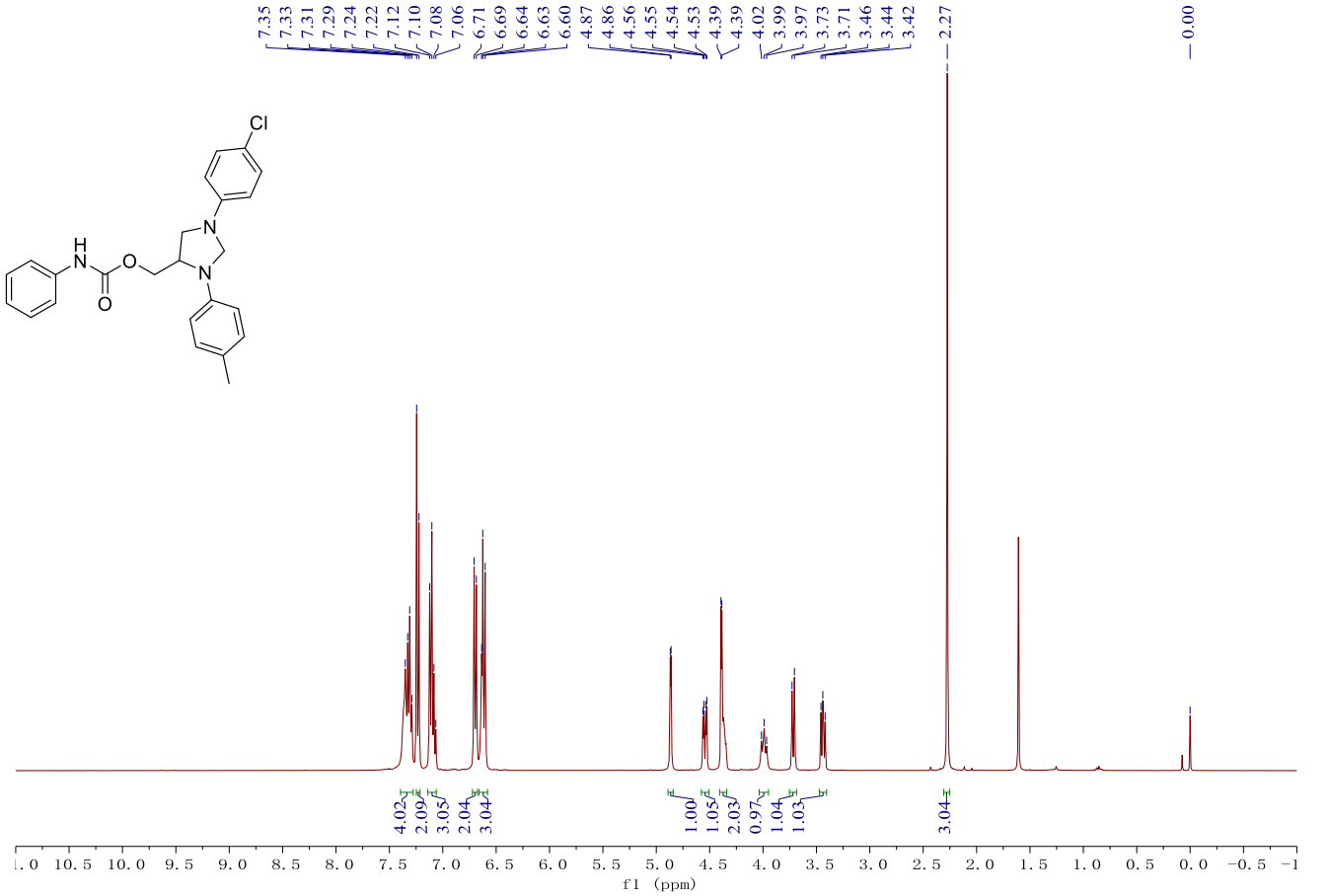


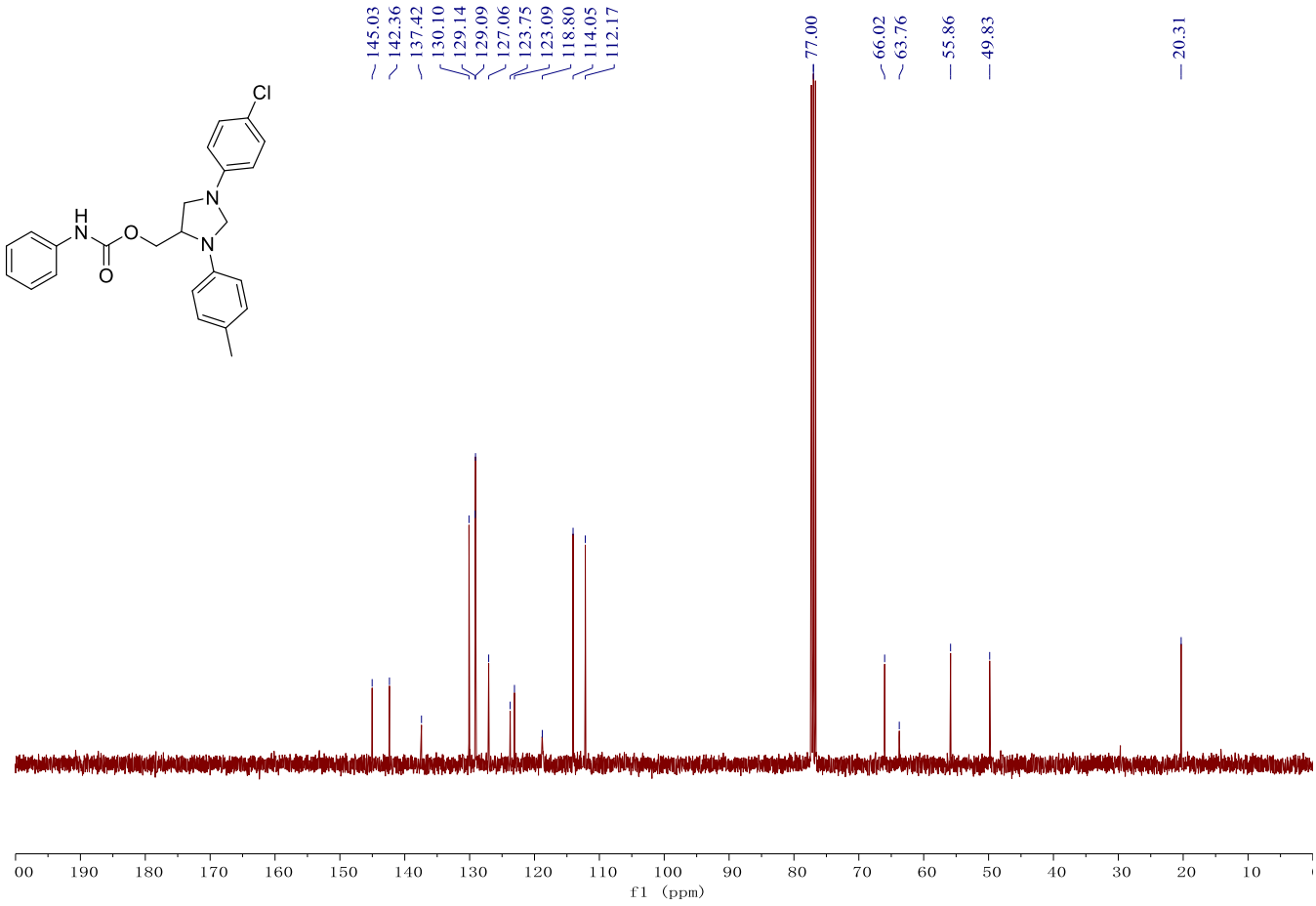
^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 5ae





^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 7





^{1}H NMR (400 MHz, $CDCl_3$) and ^{13}C NMR (100 MHz, $CDCl_3$) spectra of product 9