Supporting Information for:
Cooperative effect in organocatalytic intramolecular hydroamination of unfunctionalized olefins

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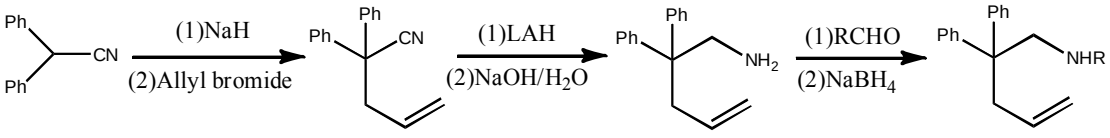
1. General information

Unless otherwise noted, all reactions were performed with commercially available reagents in oven-dried apparatus. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker DRX 400 spectrometer at 298 K using deuterated chloroform as solvent and tetramethylsilane as internal standard. Column chromatography was performed employing 200-300 mesh silica gel. Starting materials and reagents used in the reactions were purchased from J&K Chemicals (Beijing) or Aladdin Reagents (Shanghai) and were used as received without further purification.

2. Synthesis and spectral data of substrates[1]



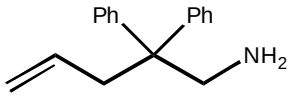
S1 S2

1a R=Bn
1b R=4-MeBn
1c R=4-MeOBn
1d R=4-O2NBn
1e R=4-FBn
1f R=4-ClBn
1g R=i-Bu
1h R=n-Bu

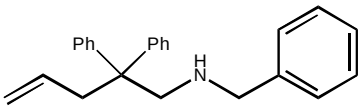
A solution of diphenylacetonitrile (4.83 g, 25 mmol) in DMF (10 mL) was added slowly to a suspension of NaH (0.66 g, 27.5 mmol) in DMF (25 mL) and the resulting mixture was stirred at room temperature for 1 h. The resulting bright yellow suspension was cooled to 0 °C, treated with allyl bromide (3.33 g, 27.5 mmol), warmed to room temperature and stirred at room temperature for 12 hours. The resulting solution was poured into ice/water (100 mL) and was extracted with CH_2Cl_2 (3 × 100 mL). The combined organic layer was washed with water (2 × 50 mL), dried with $MgSO_4$, and concentrated to give 2,2-diphenyl-4-pentenenitrile (S1) (5.41g, 93%), which was used in the subsequent step without further purification. To a suspension of $LiAlH_4$ (1.52 g, 40 mmol) in ether (130 mL) was added S1 (2.33 g, 10 mmol) at 0 °C. The mixture was slowly warmed to room temperature and stirred overnight. The resulting suspension was cooled to 0 °C and quenched by slow addition of 6 M NaOH (50 mL). The resulting mixture was extracted with ether (4 × 100 mL) and the combined ether extracts were dried ($MgSO_4$) and concentrated to give

2,2-diphenyl-4-penten-1-amine (S2) (2.01g, 85%) as a pale yellow, viscous oil. A solution of S2 (1.19 g, 5 mmol) and benzaldehyde (0.54 g, 5.1 mmol) in MeOH (20 mL) was stirred at room temperature for 5 h, then treated with $NaBH_4$ (0.29g, 7.5 mmol) and the mixture was stirred overnight. The resulting mixture was treated with water (50 mL), 1 M NaOH (20 mL) and was extracted with CH_2Cl_2 (3 × 100 mL). The combined organic layer was dried ($MgSO_4$) and concentrated. The resulting oily residue was chromatographed (petroleum ether : dichloromethane = 10:1) to give **1a** (1.32 g, 86%) as a viscous oil[1].

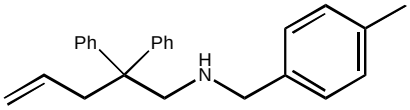
The substituted N-arylmethyl-(2,2-diphenyl-4-pentenyl)amines **1b**-**1h** were synthesized via reductive amination of S2 with the corresponding aldehydes via procedures similar to that used to synthesize 1a.



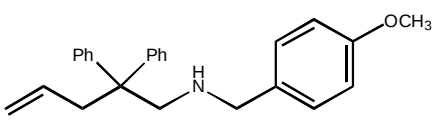
2,2-Diphenylpent-4-en-1-amine (**S2**) 1H NMR (400 MHz, $CDCl_3$) δ= 7.40-7.09 (m, 10H), 5.54-5.31 (m, 1H), 5.09-4.90 (m, 2H), 3.32 (s, 2H), 2.92 (d, J = 7.1 Hz, 2H), 0.90 (m, 2H). 13C NMR (100 MHz, CDCl3) δ= 146.26, 134.64, 128.21, 128.07, 126.07, 117.66, 51.39, 48.61, 41.16. Spectral data were consistent with known aminoalkene[2].



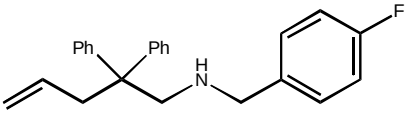
N-Benzyl-2,2-diphenylpent-4-en-1-amine (**1a**) 1H NMR (400 MHz, $CDCl_3$) δ =7.40-7.11 (m, 15H), 5.47-5.28 (m, 1H), 5.08-4.87 (m, 2H), 3.76 (s, 2H), 3.24 (s, 2H), 3.08 (d, J = 7.1 Hz, 2H). 13C NMR (100 MHz, CDCl3) δ=145.81, 139.72, 133.87, 127.16, 127.04, 126.91, 126.88, 125.66, 124.93, 116.55, 54.29, 53.15, 49.15, 40.60. Spectral data were consistent with known aminoalkene[1].



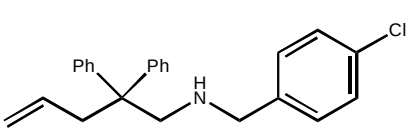
N-(4-Methylbenzyl)-2,2-diphenylpent-4-en-1-amine (**1b**) 1H NMR (400 MHz, $CDCl_3$) δ= 7.49-7.13 (m, 14H), 5.58–5.48 (m, 1H), 5.13 (dd, J = 40.5, 13.5 Hz, 2H), 3.85 (s, 2H), 3.37 (s, 2H), 3.22 (d, J = 7.0 Hz, 2H), 2.48 (s, 3H), 0.98 (s, 1H). 13C NMR (101 MHz, CDCl3) δ =145.79, 136.59, 135.08, 133.84, 127.80, 127.00, 126.86, 126.77, 124.87, 116.54, 54.18, 52.83, 49.06, 40.55, 20.03. Spectral data were consistent with known aminoalkene[3].



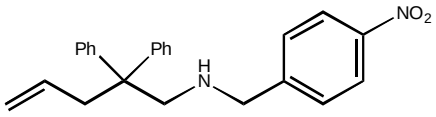
N-(4-Methoxybenzyl)-2,2-diphenylpent-4-en-1-amine (**1c**) ^{1}H NMR (400 MHz, $CDCl_3$) δ=7.30-7.00 (m, 12H), 6.80 (d, *J* = 8.6 Hz, 2H), 5.39–5.30 (m, 1H), 5.01-4.86 (m, 2H), 3.76 (s, 3H), 3.63 (s, 2H), 3.17 (s, 2H), 3.02 (d, *J* = 7.1 Hz, 2H), 1.20 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ=158.46, 146.83, 134.88, 132.80, 129.04, 128.06, 127.98, 125.93, 117.56, 113.95, 55.23, 55.18, 53.55, 50.14, 41.64. Spectral data were consistent with known aminoalkene[4].



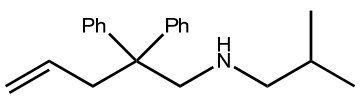
N-(4-Fluorobenzyl)-2,2-diphenylpent-4-en-1-amine (**1d**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.31-7.05 (m, 12H), 6.89 (t, *J* = 8.7 Hz, 2H), 5.37–5.29 (m, 1H), 4.91 (dd, *J* = 37.3, 13.5 Hz, 2H), 3.61 (s, 2H), 3.17 (s, 2H), 3.02 (d, *J* = 7.0 Hz, 2H), 0.83 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 162.96, 160.5, 146.75, 134.77, 129.39, 129.31, 127.99, 127.93, 125.96, 117.58, 114.97, 114.76, 55.15, 53.34, 50.09, 41.57. Spectral data were consistent with known aminoalkene[5].



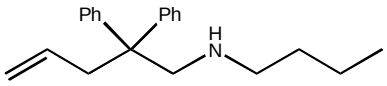
N-(4-chlorbenzyl)-2,2-diphenylpent-4-en-1-amine (**1e**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.43-7.19 (m, 14H), 5.50-5.36 (m, 1H), 5.15-4.93 (m, 2H), 3.76 (d, *J* = 3.0 Hz, 2H), 3.33-3.23 (m, 2H), 3.14 (d, *J* = 6.4 Hz, 2H), 0.99 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =146.83, 139.29, 134.90, 132.42, 129.37, 128.40, 128.13, 126.14, 117.78, 55.27, 53.50, 50.21, 41.68. Spectral data were consistent with known aminoalkene[4].



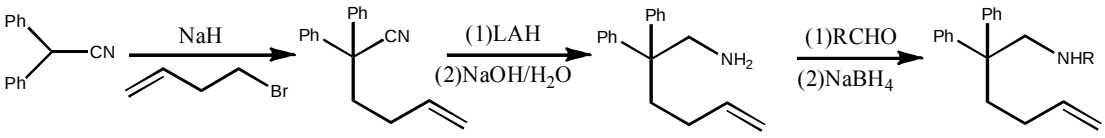
N-(4-Nitrobenzyl)-2,2-diphenylpent-4-en-1-amine (**1f**) ^{1}H NMR (400 MHz, $CDCl_3$) δ=8.11 (d, *J* = 8.7 Hz, 2H), 7.41-7.00 (m, 12H), 5.38-5.26 (m, 1H), 5.11-4.81 (m, 2H), 3.79 (s, 2H), 3.18 (s, 2H), 3.04 (d, *J* = 7.0 Hz, 2H), 1.16 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 148.55, 146.95, 146.52, 134.70, 128.45, 128.05, 127.98, 126.15, 123.44, 117.70, 55.40, 53.40, 50.14, 41.55. Spectral data were consistent with known aminoalkene[1].



N-isobutyl-2,2-diphenylpent-4-en-1-amine (**1g**) 1H NMR (400 MHz, CDCl3) δ= 7.45-7.13 (m, 10H), 5.49–5.40 (m, 1H), 5.15-4.94 (m, 2H), 3.24 (s, 2H), 3.09 (d, J= 7.0 Hz, 2H), 2.39 (d, J= 6.8 Hz, 2H), 1.69 (td, J= 13.3, 6.6 Hz, 1H), 0.83 (d, J= 6.6 Hz, 6H), 0.47 (s, 1H).13C NMR (100 MHz, $CDCl_3$) δ= 147.06 , 135.14 , 128.13, 127.97, 125.96, 117.58, 58.52 , 55.96 , 50.21, 41.68 , 27.85 , 20.56[5].



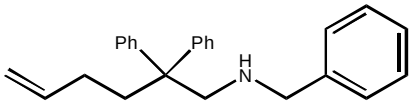
N-butyl-2,2-diphenylpent-4-en-1-amine (**1h**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.36 (m, 10H), 5.59–5.50 (m, 1H), 5.14 (dd, *J* = 41.1, 13.5 Hz, 1H), 3.36 (s, 1H), 3.18 (d, *J* = 6.4 Hz, 2H), 2.68 (t, *J* = 6.6 Hz, 2H), 1.54-1.44 (m, 2H), 1.43-1.30 (m, 2H), 1.00 (t, *J* = 7.1 Hz, 3H), 0.57 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 145.89, 133.91, 126.98, 126.85, 124.82, 116.48, 54.90, 49.00, 40.62, 30.95, 19.30, 12.94. Spectral data were consistent with known aminoalkene.



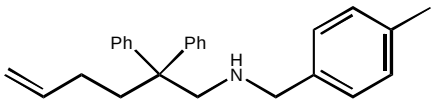
S3 S4

1i R=Bn
1j R=4-MeBn
1k R=4-MeOBn
1l R=4-FBn
1m R=4-ClBn

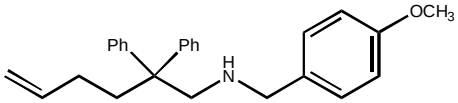
Compound **1i-1m** was synthesized from 4-Bromo-1-butene and diphenylacetonitrile employing a procedure similar to that used to synthesize **1a**.



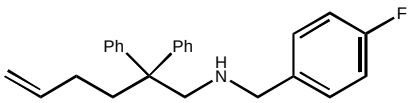
N-benzyl-2,2-diphenylhex-5-en-1-amine (**1i**) ^{1}H NMR (400 MHz, $CDCl_3$) δ =7.37-7.16 (m, 15H), 5.86-5.76 (m, 1H), 4.97 (dd, *J* = 23.7, 6.3 Hz, 2H), 3.78 (s, 2H), 3.25 (s, 2H), 2.43-2.28 (m, 2H), 1.82-1.57 (m, 2H), 1.11 (s, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ =147.27, 139.32, 129.35, 128.40, 128.21, 128.16, 128.04, 127.88, 126.12, 114.31, 55.28, 50.40, 42.70, 36.42, 28.83. Spectral data were consistent with known aminoalkene[6].



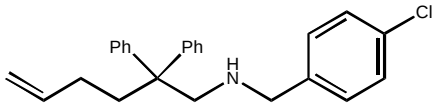
N-(4-Methylbenzyl)-2,2-diphenylhex-5-en-1-amine (**1j**) ^{1}H NMR (400 MHz, $CDCl_3$) δ =7.29-7.01 (m,14H), 5.79–5.69 (m, 1H), 4.91 (dd, *J* = 23.9, 6.3 Hz, 2H), 3.68 (s, 2H), 3.19 (s, 2H), 2.39-2.27 (m, 5H), 1.66 (dd, *J* = 15.8, 7.1 Hz, 2H), 0.90 (s, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ= 146.05, 138.12, 136.55, 135.13, 127.84, 126.98, 126.89, 126.74, 124.85, 113.00, 54.08, 52.68, 49.18, 35.23, 27.59, 20.03.



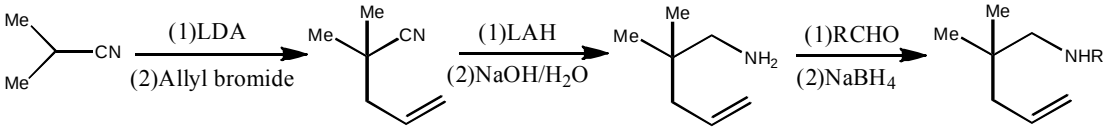
N-(4-Methoxybenzyl)-2,2-diphenylhex-5-en-1-amine (**1k**) ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28-6.92 (m, 12H), 6.70 (d, *J* = 8.5 Hz, 2H), 5.80-5.44 (m, 1H), 4.90-4.67 (m, 2H), 3.63 (s, 3H), 3.54 (s, 2H), 3.07 (s, 2H), 2.28-2.13 (m, 2H), 1.55 (d, *J* = 8.7 Hz, 2H), 0.89 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 157.41, 146.04, 138.10, 131.65, 127.94, 126.97, 126.90, 124.85, 113.02, 112.54, 54.13, 53.97, 52.35, 49.16, 35.22, 27.57.



N-(4-Fluorobenzyl)-2,2-diphenylhex-5-en-1-amine (**1l**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.30-7.07 (m, 12H), 6.95 (t, *J* = 8.7 Hz, 2H), 5.80–5.73 (m, 1H), 4.91 (dd, *J* = 21.1, 5.5 Hz, 2H), 3.67 (s, 2H), 3.17 (s, 2H), 2.39-2.22 (m, 2H), 1.64 (dd, J = 15.8, 7.2 Hz, 2H). 13C NMR (100 MHz, CDCl3) δ =161.95, 159.53, 145.95, 138.03, 135.28, 128.30, 128.25, 126.86, 124.92, 113.06, 54.01, 52.18, 49.17, 35.16, 27.56.

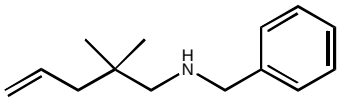


N-(4-chlorbenzyl)-2,2-diphenylhex-5-en-1-amine (**1m**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.32-7.02 (m, 14H), 5.80–5.74 (m, 1H), 4.92 (dd, *J* = 20.8, 5.4 Hz, 2H), 3.68 (s, 2H), 3.16 (s, 2H), 2.42-2.25 (m, 2H), 1.70-1.54 (m, 2H), 0.92 (s, 1H).^{13}C NMR (100 MHz, $CDCl_3$) δ =145.90, 138.14, 138.00, 131.30, 128.15, 127.27, 126.95, 124.94, 113.09, 54.05, 52.20, 49.18, 35.14, 27.57.

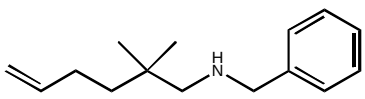


S5 S6 R=Bn

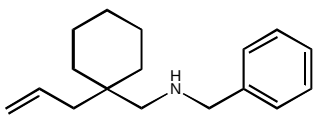
Isobutyronitrile (4.25 g, 11.5 mmol) was added to a solution of LDA at –78 °C and stirred for 45 min. To the resulting solution was added allyl bromide (10.7 mL, 124 mmol). The solution was warmed to room temperature and was stirred overnight. CH_2Cl_2 (75 mL) was added and the resulting biphasic mixture washed with water (3 × 150 mL), dried ($MgSO_4$), and concentrated. The residue was distilled under reduced pressure to give 2,2-dimethyl-4-pentenenitrile(S5) (4.21 g, 63%). Conversion of S5 to **1n** was accomplished in a manner similar to that employed for the conversion of S1 to **1a**.



N-Benzyl-2,2-dimethylpent-4-en-1-amine (**1n**)
^{1}H NMR (400 MHz, $CDCl_3$) δ =7.30-7.12 (m, 5H), 5.82-5.59 (m, 1H), 4.93 (d, *J* = 12.2 Hz, 2H), 3.71 (s, 2H), 2.29 (s, 2H), 1.95 (d, *J* = 7.5 Hz, 2H), 1.22 (s, 1H), 0.82 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ = 140.01, 134.60, 127.25, 126.94, 125.71, 115.70, 58.69, 53.69, 52.39, 43.64, 33.34, 24.50. Spectral data were consistent with known aminoalkene[1].

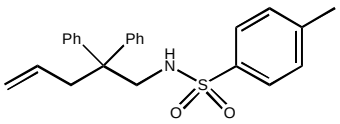


N-Benzyl-2,2-dimethylhex-5-en-1-amine (**1o**)
Compound **1o** was synthesized employing a procedure similar to that used to synthesize **1n** starting from 4-Bromo-1-butene. ^{1}H NMR (400 MHz, $CDCl_3$) δ =7.34-7.19 (m, 5H), 5.86–5,78 (m, 1H), 5.04-4.83 (m,2H), 3.79 (s,2H), 2.36 (s, 2H), 2.02-1.89 (m, 2H), 1.39-1.25 (m, 2H), 1.14 (s, 1H), 0.89 (s, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =141.06, 139.74, 128.27, 127.96, 126.74, 113.78, 99.13, 59.72, 54.72, 39.34, 33.93, 28.49, 25.62. Spectral data were consistent with known aminoalkene[7].

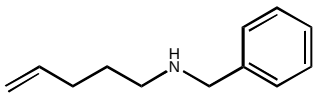


1-(1-Allylcyclohexyl)-N-b enzylmethanamine (**1p**)
Compound **1p** was synthesized employing a procedure similar to that used to synthesize **1n** starting from cyclohexanenitrile. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27-7.12 (m, 5H), 5.75–5.67 (m, 1H), 4.95-4.86 (m, 2H), 3.68 (s, 2H), 2.33 (s, 2H),

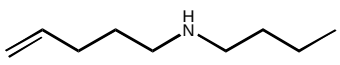
2.04 (d, *J* = 7.5 Hz, 2H), 1.36-1.17 (m, 10H), 1.12 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =140.09, 134.37, 127.21, 126.96, 125.68, 115.56, 54.86, 53.75, 39.66, 35.64, 33.00, 25.40, 20.57. Spectral data were consistent with known aminoalkene[1].



N-(2,2-Diphenylpent-4-enyl)-4-methylbenzenesulfonamide (**1q**) To a solution of 2,2-diphenylpent-4-en-1-amine (1.0 mmol) in 10 mL of dry toluene was added *p*-toluenesulfonyl chloride (1.0 mmol) and pyridine (0.5 mL, 2 mmol). The mixture was stirred at 25 °C for 24 h, diluted with 5 mL of 1 N HCl (aq) and was extracted with Et_2O (3 × 30 mL). The combined organic layer was dried over $MgSO_4$, filtered and concentrated in vacuo. The resulting oily residue was chromatographed (petroleum ether : dichloromethane= 5:1) to give 1q (0.27g, 70%) as a viscous oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.52 (d, *J* = 8.2 Hz, 2H), 7.24-6.86 (m, 12H), 5.21-5.09 (m, 1H), 4.92-4.77 (m, 2H), 3.45 (d, *J* = 6.4 Hz, 2H), 2.82 (d, *J* = 7.1 Hz, 2H), 2.35 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =143.53, 142.41, 135.23, 132.10, 128.67, 127.40, 126.75, 126.13, 125.73, 118.04, 48.41, 48.27, 40.24, 20.50. Spectral data were consistent with known aminoalkene[2].



N-Benzylpent-4-en-1-amine (**1r**) To a solution of benzylamine (5 mmol) and 5-bromo-1-pentene (1 mmol) in ethanol (30 mL) was added NaI (0.1 mmol,). The mixture was stirred overnight at 75 °C and the solvent was removed in vacuo. The resulting oily residue was chromatographed (petroleum ether : ethylacetate= 5:1) to give **1r** (0.09 g, 53%) as a clear oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25-7.07 (m, 5H), 5.81-5.53 (m, 1H), 4.94-4.82 (m, 2H), 3.67 (s, 2H), 2.54 (t, *J* = 7.2 Hz, 2H), 2.00 (dd, *J* = 14.5, 7.1 Hz, 2H), 1.56-1.45 (m, 2H), 1.22 (s, 1H).^{13}C NMR (100 MHz, $CDCl_3$) δ =139.55, 137.44, 127.30, 127.03, 125.80, 113.57, 52.97, 47.71, 30.49, 28.26. Spectral data were consistent with known aminoalkene[7].



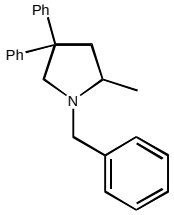
N-butylpent-4-en-1-amine (**1s**) Compound **1s** was synthesized via procedure similar to that used to synthesize **1r**. ^{1}H NMR (400 MHz, $CDCl_3$) δ 5.86-5.76 (m, 1H), 4.98 (ddd, *J* = 13.6, 11.0, 1.3 Hz, 2H), 2.60 (dd, *J* = 14.2, 6.8 Hz, 4H), 2.09 (dd, *J* =

14.6, 7.0 Hz, 2H), 1.58 (dd, *J* = 14.7, 7.3 Hz, 2H), 1.51-1.42 (m, 2H), 1.37–1.33 (m, 2H), 0.92 (t, *J* = 7.3 Hz, 4H). ^{13}C NMR (101 MHz, $CDCl_3$) δ =137.52, 113.51, 48.81, 48.60, 31.53, 30.73, 28.57, 19.63, 13.06. Spectral data were consistent with known aminoalkene[2].

3. Typical procedure for hydroamination and spectral data of products

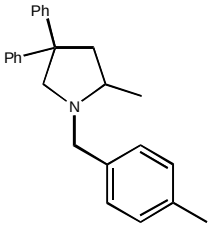
Typical procedure for intramolecular hydroamination reaction: all commercially available compounds were purchased from Aladdin Reagents (Shanghai) or J&K Chemicals (Beijing) and were used as received.

To a 25 mL Vacuum Tube was added 0.5 mmol alkenylamine, 20 mol% 3-hydroxy-2-naphthoic acid, and 3 mL Xylene. The reaction mixture was stirred for an indicated period at 130 °C and was then concentrated to give an oil. Column chromatography gave the corresponding products.



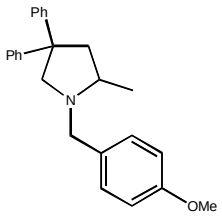
1-Benzyl-2-methyl-4,4-diphenyl-pyrrolidine (**2a**)

1H NMR (400 MHz, CDCl3) δ= 7.33-7.05 (m, 15H), 4.00 (d, J = 13.2 Hz, 1H), 3.56 (d, J = 9.9 Hz, 1H), 3.17 (d, J = 13.3 Hz, 1H), 2.87-2.79 (m, 1H), 2.79-2.65 (m, 2H), 2.12 (dd, J = 12.6, 7.7 Hz, 1H), 1.08 (d, J = 5.9 Hz, 3H). 13C NMR (101 MHz, CDCl3) δ =149.57, 147.67, 139.04, 128.16, 127.56, 127.17, 127.09, 126.78, 126.39, 126.21, 125.75, 124.76, 65.39, 58.61, 56.97, 51.52, 46.96, 18.47. Spectral data were consistent with known aminoalkene[1].

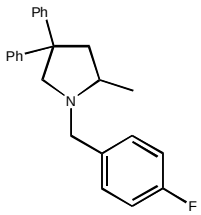


1-(4-Methylbenzyl)-2-(methyl)-4,4-diphenylpyrrolidin (**2b**)

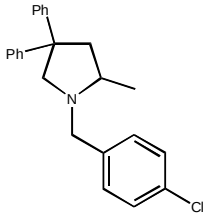
1H NMR (400 MHz, CDCl3) δ 7.22-6.90 (m, 14H), 3.94 (d, J = 13.1 Hz, 1H), 3.54 (d, J = 9.9 Hz, 1H), 3.10 (d, J = 13.1 Hz, 1H), 2.87-2.73 (m, 1H), 2.76-2.59 (m, 2H), 2.23 (s, 3H), 2.09 (dd, J = 12.7, 7.7 Hz, 1H), 1.05 (d, J = 6.0 Hz, 3H). 13C NMR (100 MHz, CDCl3) δ= 149.62, 147.70, 135.90, 135.18, 127.83, 127.48, 127.05, 126.74, 126.38, 126.21, 124.71, 124.31, 65.33, 58.53, 56.63, 51.44, 46.97, 20.07, 18.47.Spectral data were consistent with known aminoalkene[3].



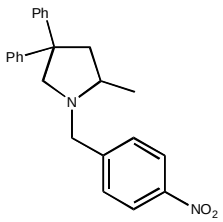
1-(4-Methoxybenzyl)-2-(methyl)-4,4-diphenylpyrrolidin (**2c**) ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27-7.05 (m, 12H), 6.85 (d, *J* = 8.6 Hz, 2H), 4.01 (d, *J* = 13.0 Hz, 1H), 3.79 (s, 3H), 3.61 (d, *J* = 9.8 Hz, 1H), 3.18 (d, *J* = 13.0 Hz, 1H), 2.90 (dd, *J* = 12.8, 7.8 Hz, 1H), 2.84-2.76 (m, 1H), 2.74 (d, *J* = 9.9 Hz, 1H), 2.18 (dd, *J* = 12.7, 7.8 Hz, 1H), 1.14 (d, *J* = 6.0 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =158.52, 150.66, 148.78, 132.11, 129.66, 128.10, 127.79, 127.42, 127.25, 125.75, 125.35, 113.58, 66.32, 59.51, 57.27, 55.23, 52.45, 48.04, 19.52. Spectral data were consistent with known aminoalkene[4].



1-(4- Fluorobenzyl)-2-(methyl)-4,4-diphenylpyrrolidin (**2d**) ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33-7.12 (m, 12H), 6.98 (t, *J* = 8.7 Hz, 2H), 4.00 (d, *J* = 13.2 Hz, 1H), 3.59 (d, *J* = 9.8 Hz, 1H), 3.21 (d, *J* = 13.2 Hz, 1H), 2.90 (dd, *J* = 12.7, 7.8 Hz, 1H), 2.81 (dd, *J* = 13.6, 7.5 Hz, 1H), 2.75 (d, *J* = 9.8 Hz, 1H), 2.20 (dd, *J* = 12.7, 7.7 Hz, 1H), 1.14 (d, *J* = 6.0 Hz, 3H).^{13}C NMR (100 MHz, $CDCl_3$) δ =150.47, 148.62, 135.77, 135.75, 129.97, 128.13, 127.83, 127.37, 127.17, 125.81, 125.44, 115.04, 66.34, 59.56, 57.16, 52.50, 47.93, 19.50. Spectral data were consistent with known aminoalkene[5].

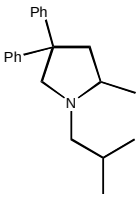


1-(4-chlorbenzyl)-2-(methyl)-4,4-diphenylpyrrolidin (**2e**) ^{1}H NMR (400 MHz, $CDCl_3$) δ =7.18-6.98 (m, 14H), 3.90 (d, J = 13.4 Hz, 1H), 3.50 (d, J = 9.8 Hz, 1H), 3.11 (d, J = 13.4 Hz, 1H), 2.80 (dd, J = 12.6, 7.8 Hz, 1H), 2.72 (ddd, J = 13.7, 9.7, 3.6 Hz, 1H), 2.66 (d, J = 9.8 Hz, 1H), 2.11 (dd, J = 12.6, 7.6 Hz, 1H), 1.04 (d, J = 5.9 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =150.51, 148.68, 138.79, 132.50, 129.95, 128.44, 128.27, 127.96, 127.47, 127.26, 125.95, 125.59, 66.46, 59.71, 57.35, 52.66, 47.98, 19.62. Spectral data were consistent with known aminoalkene[4].



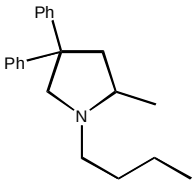
1-(4- Nitrobenzyl)-2-(methyl)-4,4-diphenylpyrrolidin (**2f**) ^{1}H NMR (400 MHz, $CDCl_3$) δ =8.15 (d, *J* = 8.6 Hz, 2H), 7.50 (d, *J* = 8.4 Hz, 2H), 7.28-7.10 (m, 10H), 4.10 (d, *J* = 14.3 Hz, 1H), 3.59 (d, *J* = 9.7 Hz, 1H), 3.40 (d, *J* = 14.3 Hz, 1H), 2.95-2.86 (m, 2H), 2.84 (d, *J* = 9.7 Hz, 1H), 2.27

(q, *J* = 11.0 Hz, 1H), 1.17 (d, *J* = 5.5 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ =150.03, 148.31, 148.23, 147.03, 128.98, 128.22, 127.94, 127.28, 127.03, 125.96, 125.65, 123.51, 66.44, 59.70, 57.30, 52.74, 47.61, 19.49. Spectral data were consistent with known aminoalkene[1].



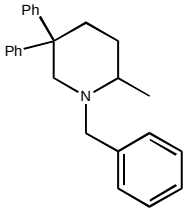
1-Isobutyl-2-methyl-4,4-diphenyl-pyrrolidine (**2g**)

^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.61-6.92 (m, 10H), 3.90 (d, *J* = 9.6 Hz, 1H), 2.92 (dd, *J* = 12.8, 7.6 Hz, 1H), 2.84 (d, *J* = 9.6 Hz, 1H), 2.77-2.65 (m, 1H), 2.48 (t, *J* = 11.0 Hz, 1H), 2.26-2.10 (m, 2H), 1.94-1.77 (m, 1H), 1.16 (d, *J* = 6.0 Hz, 3H), 1.05 (t, *J* = 10.5 Hz, 3H), 0.99 (d, *J* = 6.6 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ= 151.38, 148.94, 128.19, 127.86, 127.66, 127.36, 125.86, 125.45, 67.49, 62.69, 60.33, 52.77, 48.12, 27.82, 21.46, 20.97, 19.65. Spectral data were consistent with known aminoalkene[5].



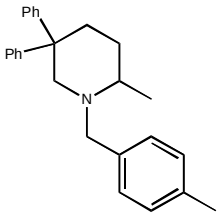
1-Butyl-2-methyl-4,4-diphenyl-pyrrolidine (**2h**)

^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.53-6.86 (m, 10H), 3.91 (d, *J* = 9.9 Hz, 1H), 2.90-2.70 (m, 3H), 2.64 (dd, *J* = 13.9, 7.6 Hz, 1H), 2.21-2.03 (m, 2H), 1.56-1.50 (m, 2H), 1.38-1.34 (m, 2H), 1.09 (d, *J* = 6.1 Hz, 3H), 0.93 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =150.75, 148.87, 128.16, 127.93, 127.45, 127.20, 125.80, 125.44, 66.84, 60.40, 53.87, 52.57, 47.96, 31.06, 20.86, 19.37, 14.05.



1-Benzyl-2- methyl -5,5-diphenyl -piperidine (**2i**)

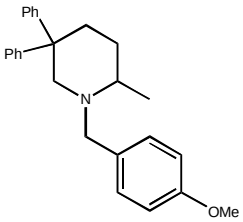
^{1}H NMR (400 MHz, $CDCl_3$) δ =7.40-6.93 (m, 15H), 4.04 (d, *J* = 13.3 Hz, 1H), 3.35 (d, *J* = 12.2 Hz, 1H), 3.13 (d, *J* = 13.3 Hz, 1H), 2.51-2.37 (m, 3H), 2.23-2.09 (m, 1H), 1.64-1.56 (m, 1H), 1.36 (ddd, *J* = 24.7, 9.4, 3.2 Hz, 1H), 1.13 (d, *J* = 6.1 Hz, 3H). 13C NMR (101 MHz, CDCl3) δ =147.52, 145.68, 138.39, 129.02, 128.45, 127.41, 126.89, 126.62, 125.99, 125.84, 124.60, 124.25, 59.91, 57.85, 55.07, 45.48, 33.19, 29.94, 17.59.Spectral data were consistent with known aminoalkene[6].



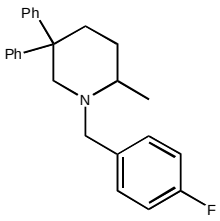
1-(4-Methylbenzyl)-2-methyl-5,5-diphenyl-piperidine (**2j**)

^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.47-7.15 (m, 14H), 4.15 (d, *J* = 13.2 Hz, 1H), 3.50 (d, *J* = 12.3 Hz, 1H), 3.24 (d, *J* = 13.2 Hz, 1H), 2.56-2.55 (m, 3H), 2.50 (s, 3H), 2.30 (t, *J* = 11.8 Hz, 1H), 1.78-1.71 (m, 1H), 1.55-1.43 (m, 1H), 1.27 (dd, *J* =

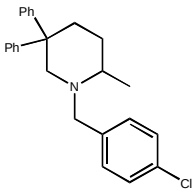
6.1, 1.8 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =147.56, 145.73, 135.20, 128.38, 127.65, 127.45, 127.20, 126.86, 126.55, 126.00, 124.57, 124.22, 59.81, 57.49, 54.98, 45.47, 33.23, 29.93, 20.12.



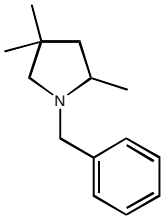
1-(4-Methoxybenzyl)-2-methyl-5,5-diphenyl-piperidine (**2k**) ^{1}H NMR (400 MHz, $CDCl_3$) δ =7.32-7.03 (m, 12H), 6.87 (d, *J* = 8.5 Hz, 2H), 3.97 (d, *J* = 13.1 Hz, 1H), 3.79 (s, 3H), 3.33 (d, *J* = 12.2 Hz, 1H), 3.05 (d, *J* = 13.1 Hz, 1H), 2.46-2.39 (m, 2H), 2.37 (d, *J* = 12.4 Hz, 1H), 2.15 (td, *J* = 12.9, 3.5 Hz, 1H), 1.64-1.53 (m, 1H), 1.34 (d, *J* = 12.0 Hz, 1H), 1.11 (d, *J* = 6.1 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 157.56, 147.57, 145.75, 130.29, 129.53, 127.43, 126.88, 126.57, 126.01, 124.58, 124.23, 112.35,59.65, 57.05, 54.95, 54.17, 45.48, 33.22, 29.97.



1-(4-Fluorobenzyl)-2-methyl-5,5-diphenyl-piperidine (**2l**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.33-6.82 (m, 14H), 3.90 (d, *J* = 13.2 Hz, 1H), 3.23 (d, *J* = 12.2 Hz, 1H), 2.99 (d, *J* = 12.8 Hz, 1H), 2.42-2.22 (m, 3H), 2.07 (t, *J* = 12.3 Hz, 1H), 1.59-1.47 (m, 1H), 1.34-1.20 (m, 1H), 1.07-0.96 (m, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ = 148.54, 146.70, 135.17, 130.83, 128.43, 128.05, 127.73, 127.06, 125.77, 125.43, 114.96, 114.75. 60.93, 58.12, 56.18, 46.60, 34.25, 31.06, 18.33.

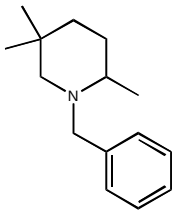


1-(4-chlorbenzyl)-2-methyl-5,5-diphenyl-piperidine (**2m**) ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.21-6.93 (m, 14H), 3.90 (d, *J* = 13.5 Hz, 1H), 3.23 (d, *J* = 12.3 Hz, 1H), 3.02 (d, *J* = 13.5 Hz, 1H), 2.42-2.30 (m, 3H), 2.12-2.00 (m, 1H), 1.61-1.49 (m, 1H), 1.29 (d, *J* = 12.0 Hz, 1H), 1.29 (d, *J* = 12.0 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ= 147.39, 145.53, 137.03, 131.51, 129.68, 127.32, 127.13, 126.96, 126.66, 125.95, 124.70, 124.36, 60.03, 57.19, 55.16, 45.51, 33.14, 29.94, 17.59.



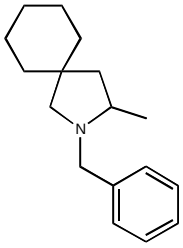
1-Benzyl-2-methyl-4,4-dimethyl-pyrrolidine (**2n**) ^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.35-7.15 (m, 5H), 4.01 (d, *J* = 13.2 Hz, 1H), 2.64 (d, *J* = 9.1 Hz, 1H), 2.61-2.50 (m, 1H), 1.94 (d, *J* = 9.1 Hz, 1H), 1.72 (dd, *J* = 12.4, 7.3 Hz, 1H), 1.31 (dd, *J* = 12.3, 9.0 Hz, 1H), 1.14 (d, *J* = 6.0 Hz, 3H), 1.07 (s, 3H), 0.97 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =140.10, 128.72, 128.08,

126.59, 68.41, 59.80, 58.06, 49.14, 35.39, 30.65, 29.28, 19.49. Spectral data were consistent with known aminoalkene[1].



1-Benzyl-2-methyl-5,5-dimethyl-piperidine (**2o**)

^{1}H NMR (400 MHz, $CDCl_3$) δ =7.47-7.18 (m, 5H), 4.05 (d, *J* = 13.8 Hz, 1H), 3.13 (d, *J* = 13.8 Hz, 1H), 2.36 (dd, *J* = 11.3, 1.8 Hz, 1H), 2.29 (s, 1H), 1.74 (d, *J* = 11.3 Hz, 1H), 1.66-1.52 (m, 2H), 1.41 (dtd, *J* = 12.9, 4.3, 2.0 Hz, 1H), 1.27-1.20 (m, 1H), 1.17 (d, *J* = 6.2 Hz, 3H), 1.00 (s, 3H), 0.85 (s, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ =140.62, 128.46, 127.98, 126.36, 63.85, 58.26, 56.56, 37.09, 31.60, 30.85, 28.94, 25.19, 18.86. Spectral data were consistent with known aminoalkene[7].



1-Benzyl-2-methyl-5-allylcyclohexyl-piperidine (**2p**)

^{1}H NMR (400 MHz, $CDCl_3$) δ= 7.26 (m, 5H), 4.01 (d, *J* = 13.3 Hz, 1H), 3.07 (d, *J* = 13.3 Hz, 1H), 2.77 (d, *J* = 9.3 Hz, 1H), 2.54-2.39 (m, 1H), 1.86 (d, *J* = 9.4 Hz, 1H), 1.74 (dd, *J* = 12.4, 6.9 Hz, 1H), 1.45-1.14 (m , 11H), 1.13 (d, *J* = 6.0 Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$) δ=140.01, 128.61, 128.04, 126.51, 66.67, 58.97, 57.96, 46.95, 39.27, 38.50, 26.06, 23.62, 23.51, 19.29. Spectral data were consistent with known aminoalkene[1].

4. References

[1] C. F. Bender, R. A. Widenhoefer, *Journal of the American Chemical Society* **2005**, *127*, 1070-1071.

[2] G.-Q. Liu, W. Li, Y.-M. Li, *Advanced Synthesis & Catalysis* **2013**, *355*, 395-402.

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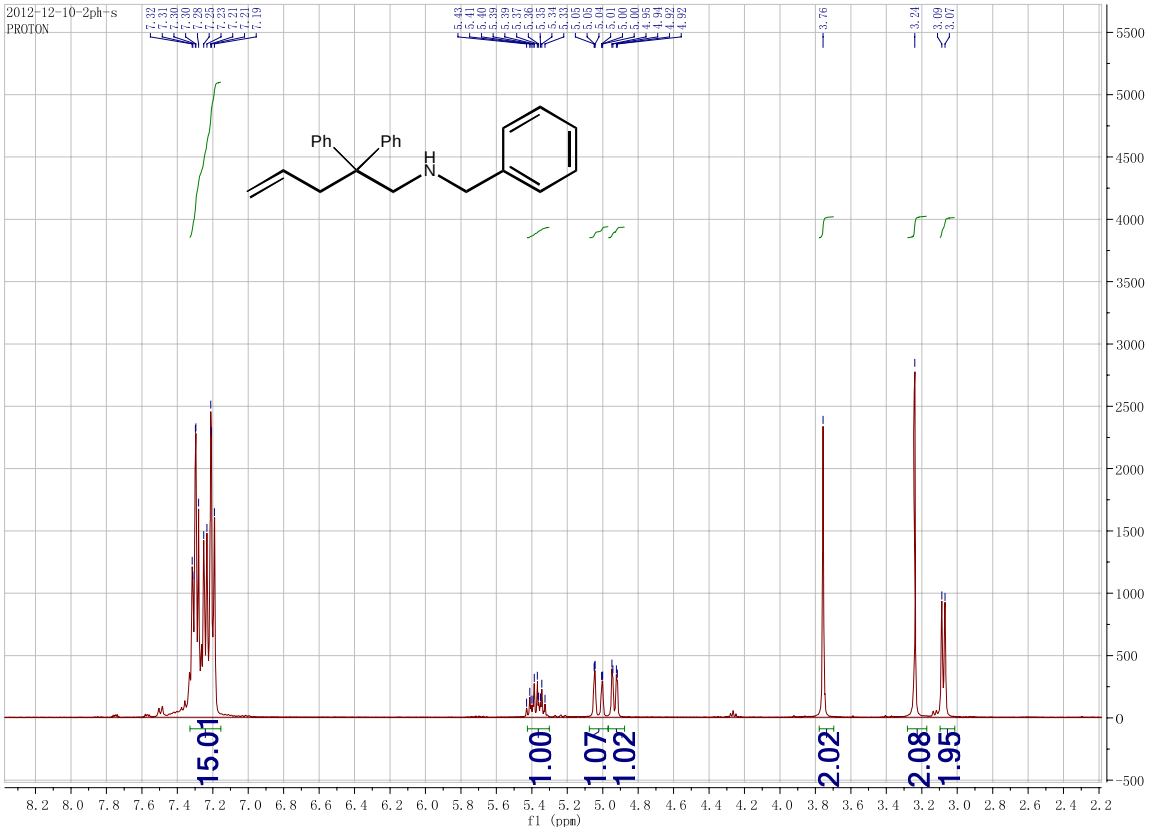
[4] L. Ackermann, L. T. Kaspar, A. Althammer, *Organic & Biomolecular Chemistry* **2007**, *5*, 1975-1978.

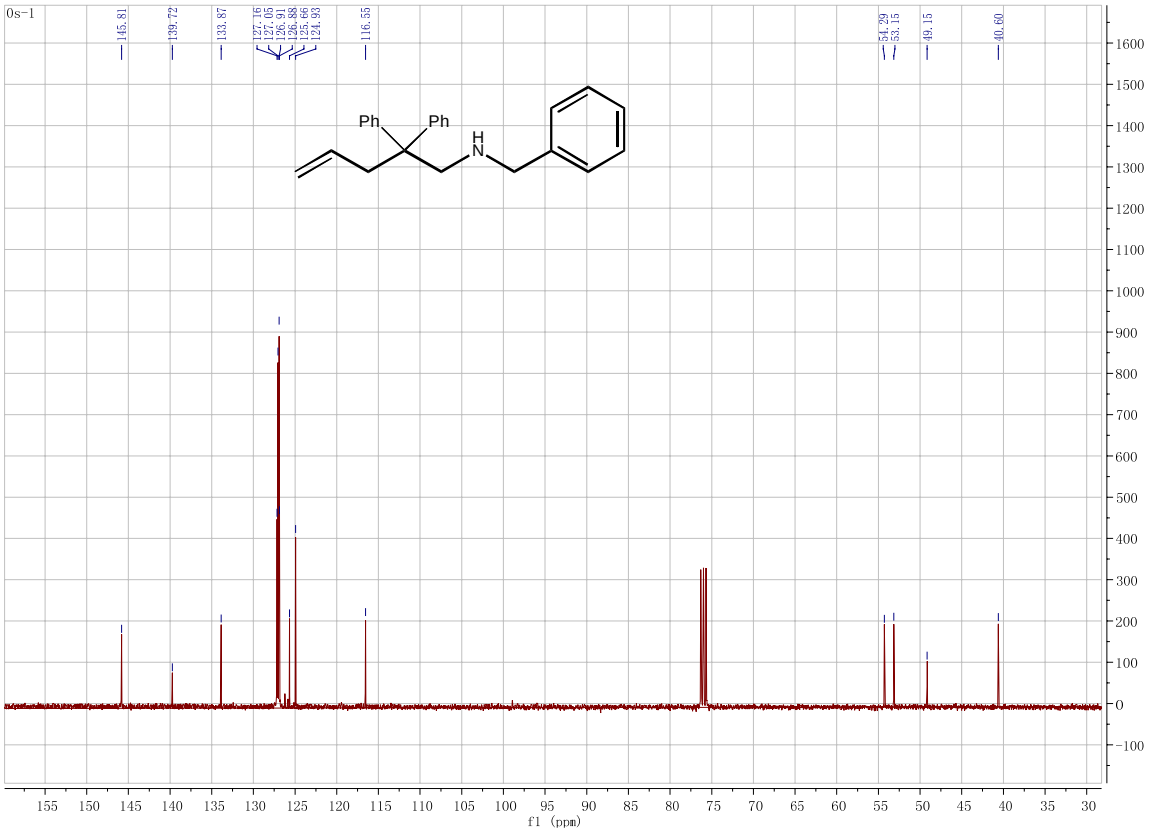
[5] H. Ohmiya, T. Moriya, M. Sawamura, *Organic Letters* **2009**, *11*, 2145-2147.

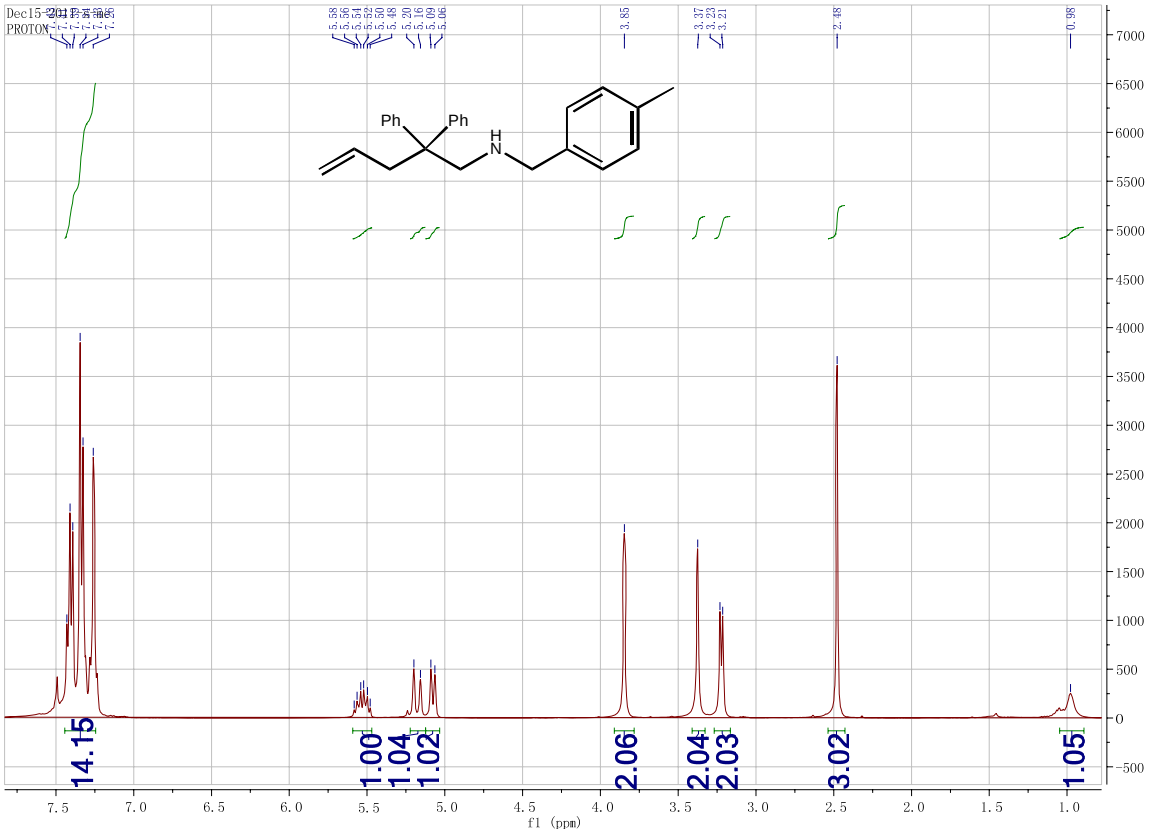
[6] E. B. Bauer, G. T. S. Andavan, T. K. Hollis, R. J. Rubio, J. Cho, G. R. Kuchenbeiser, T. R. Helgert, C. S. Letko, F. S. Tham, *Organic Letters* **2008**, *10*, 1175-1178.

[7] C. Quinet, P. Jourdain, C. Hermans, A. Atest, I. Lucas, I. E. Marko, *Tetrahedron* **2008**, *64*, 1077-1087.

5. Copies of NMR Spectra



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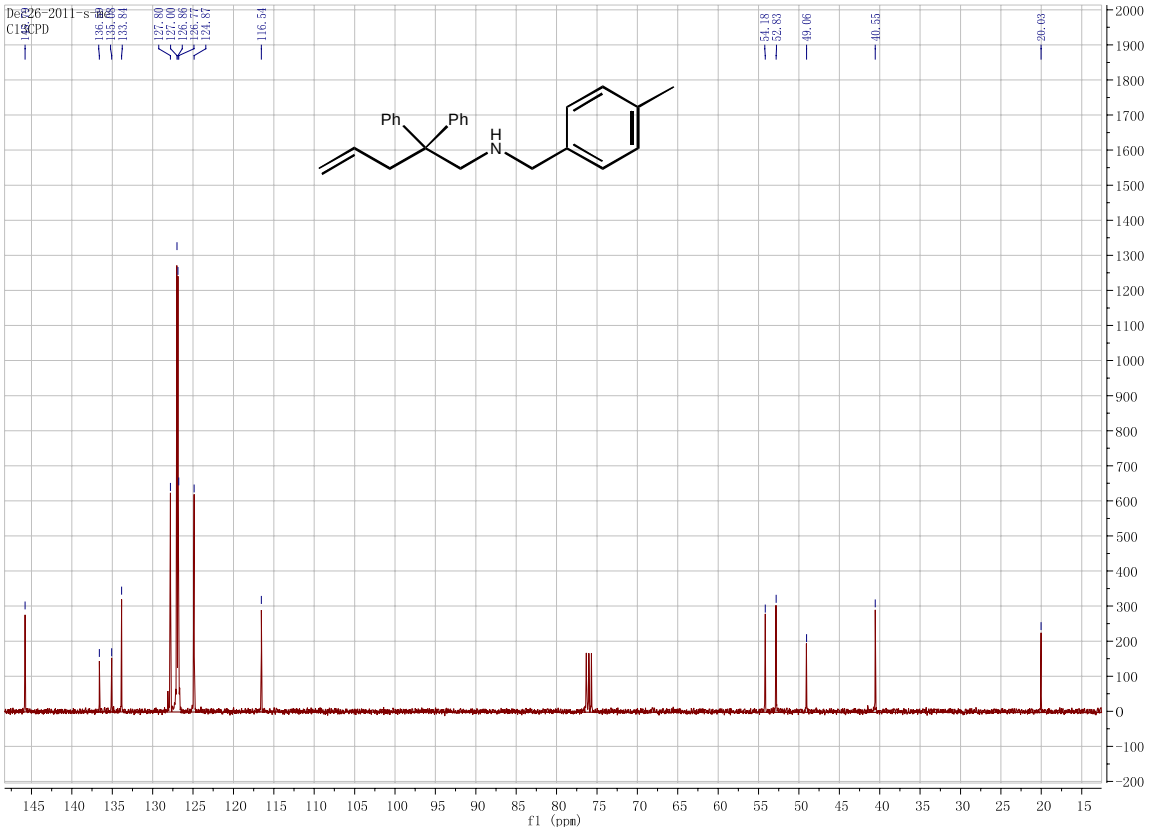
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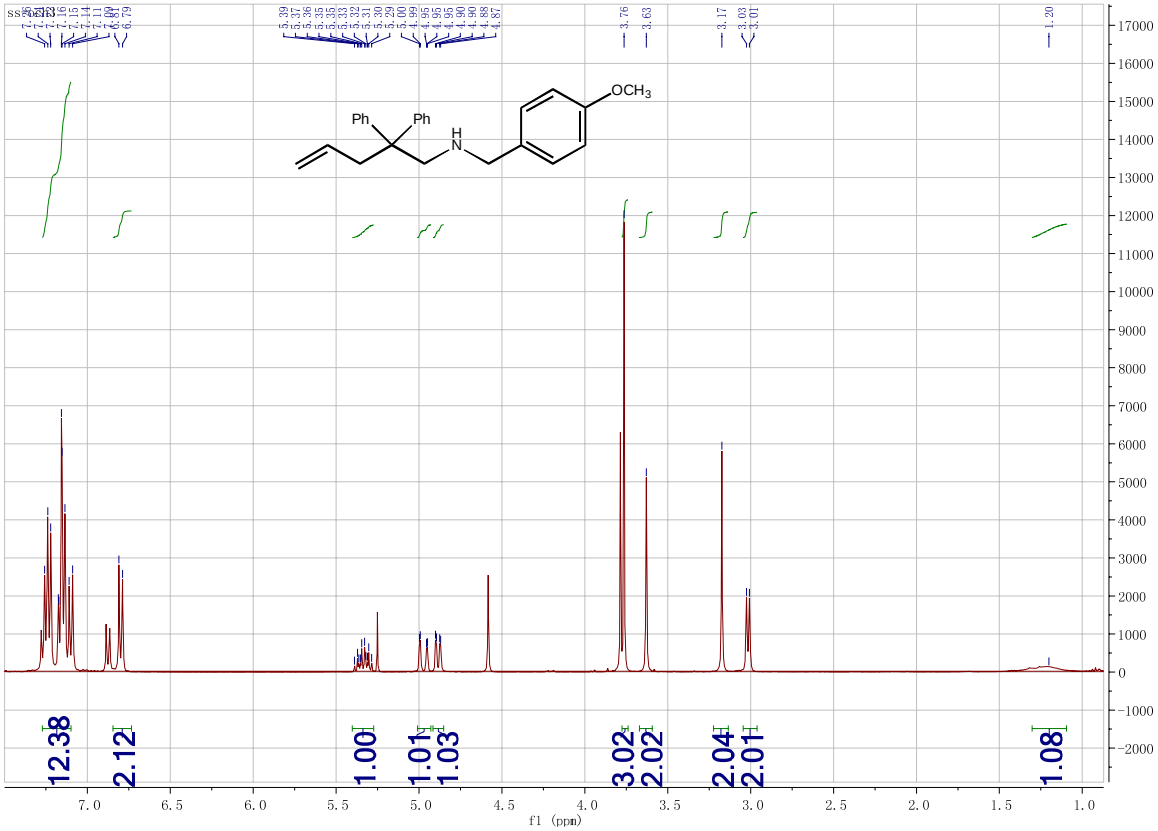
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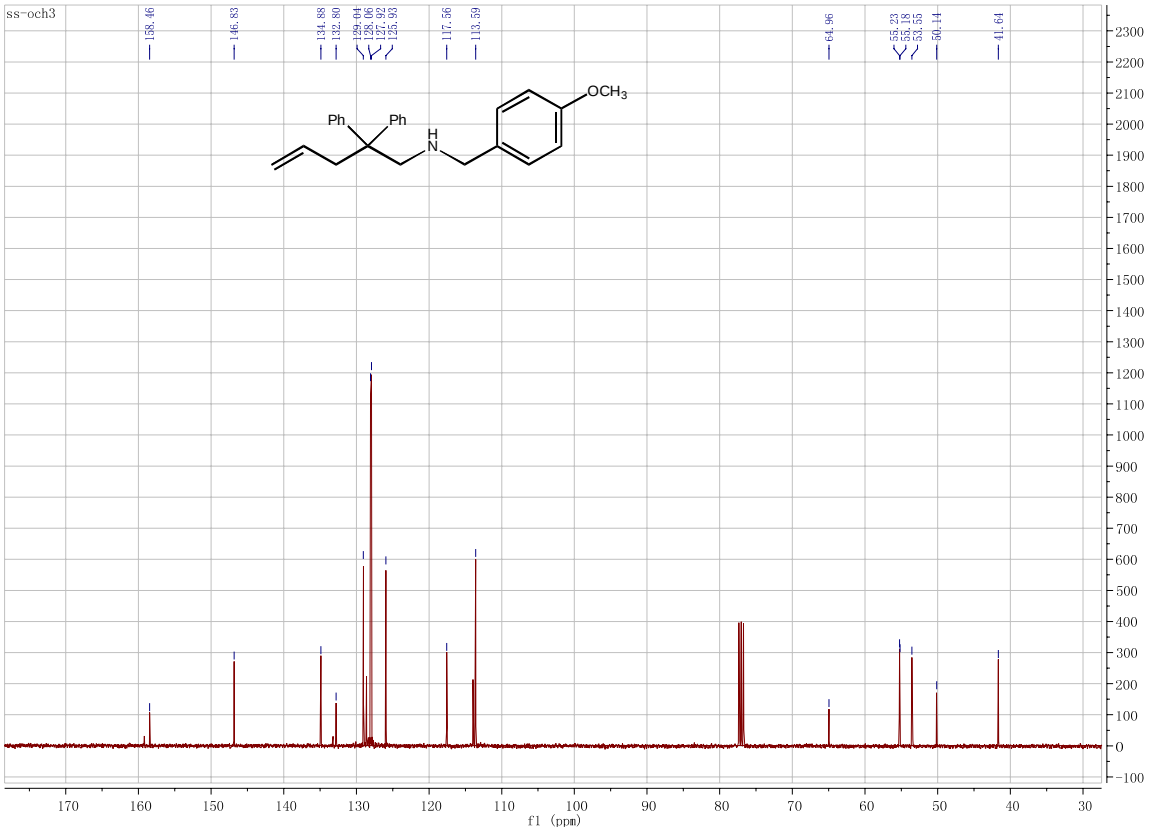
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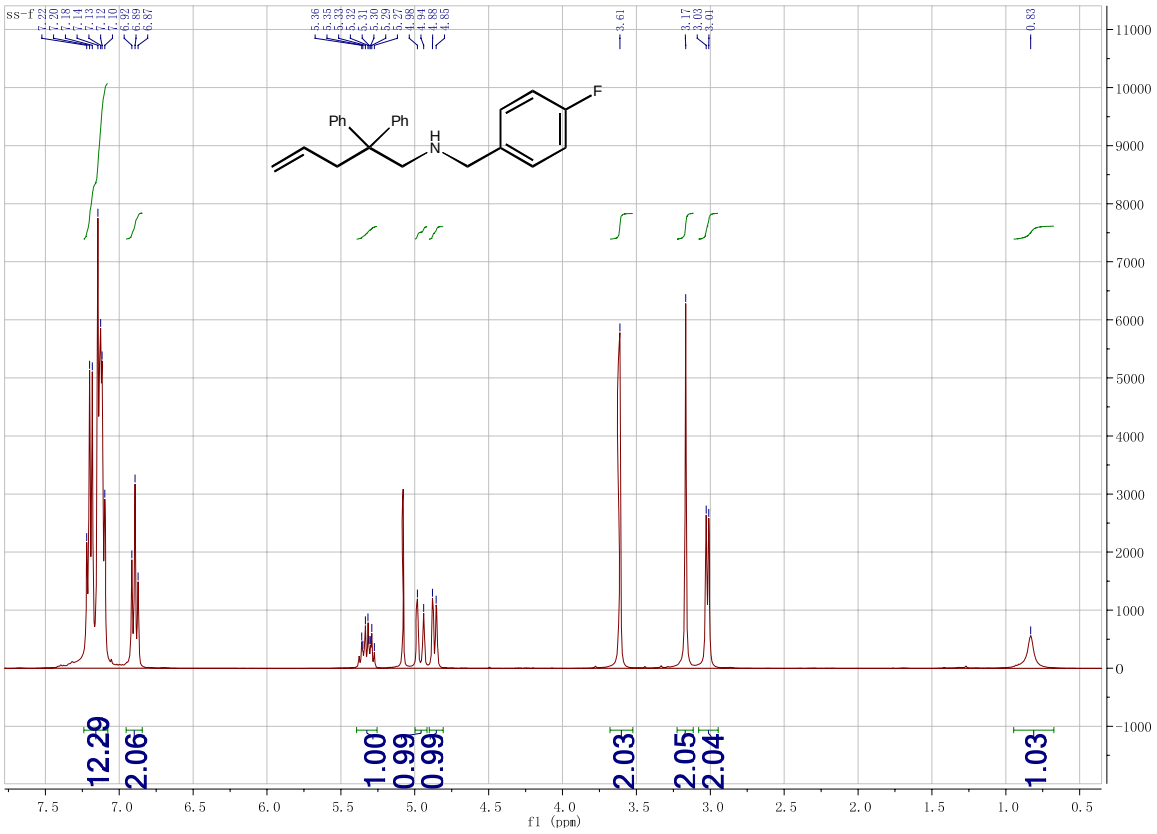
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5.33
5.32
5.31
5.30
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4.84
4.85
3.61
3.17
3.03
3.01
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Ph
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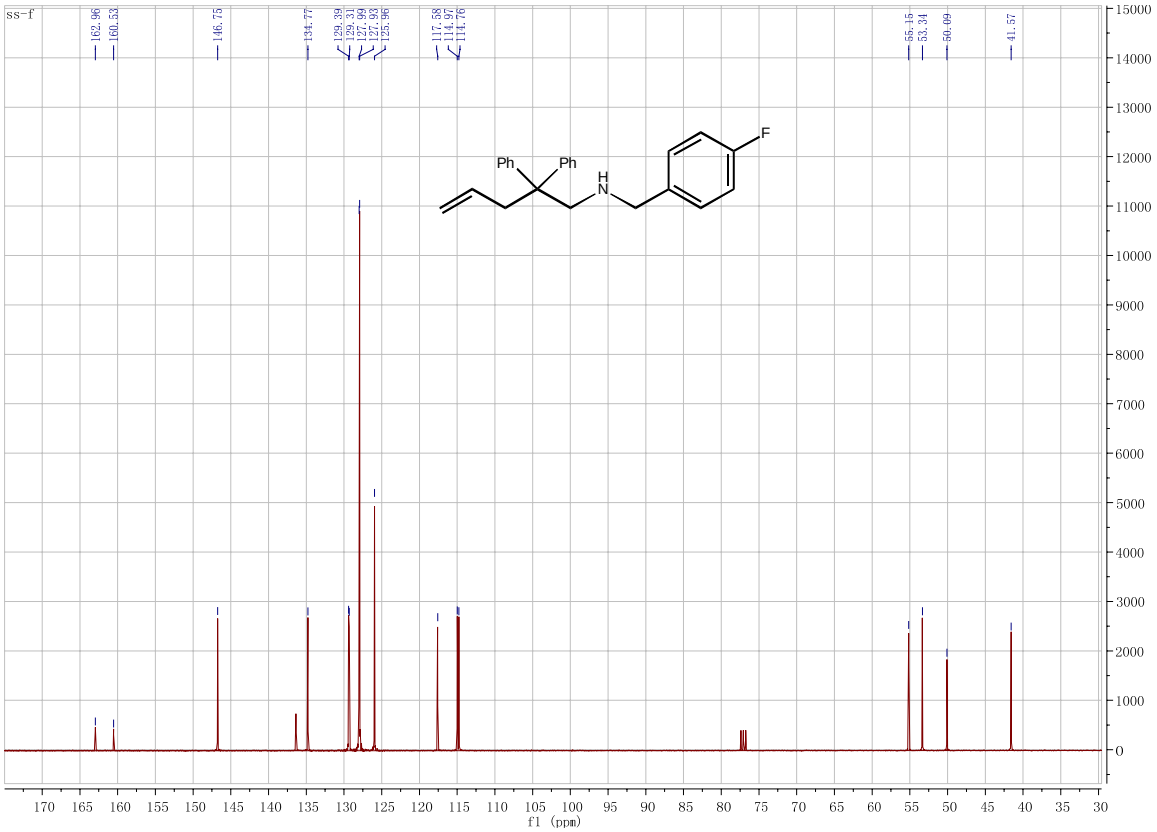
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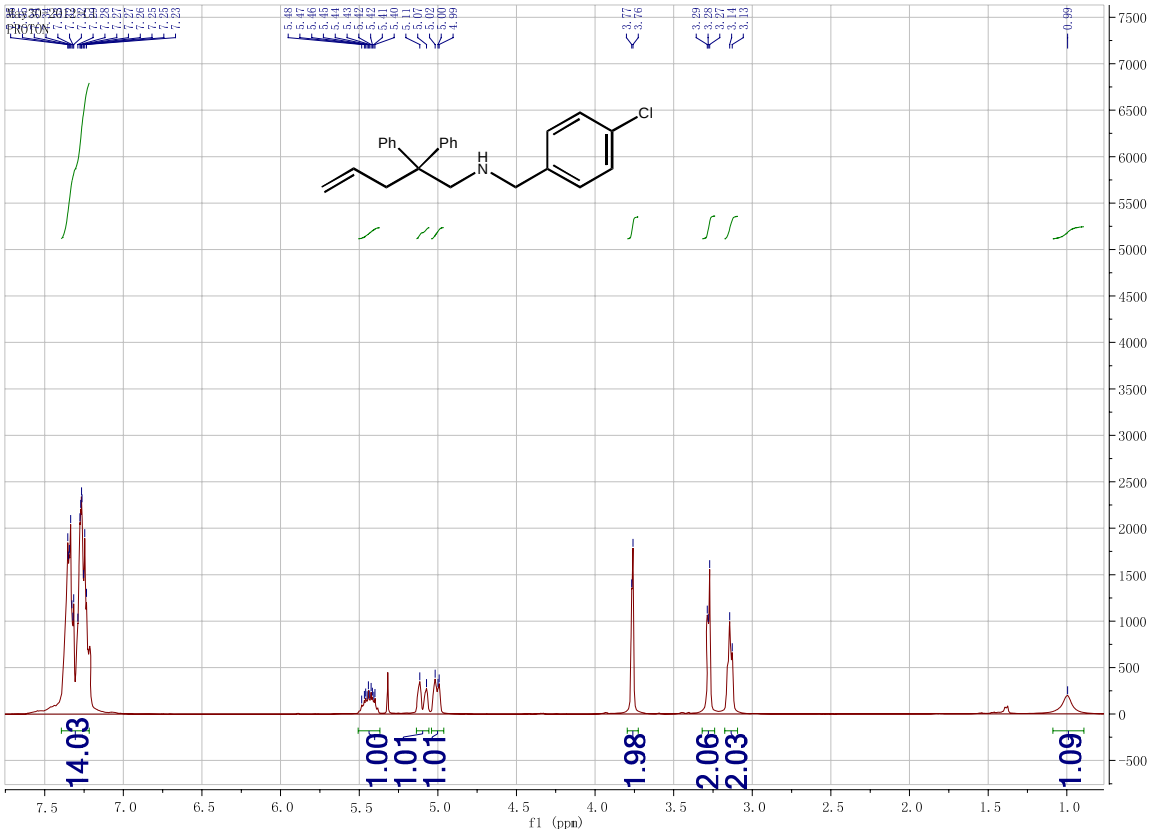
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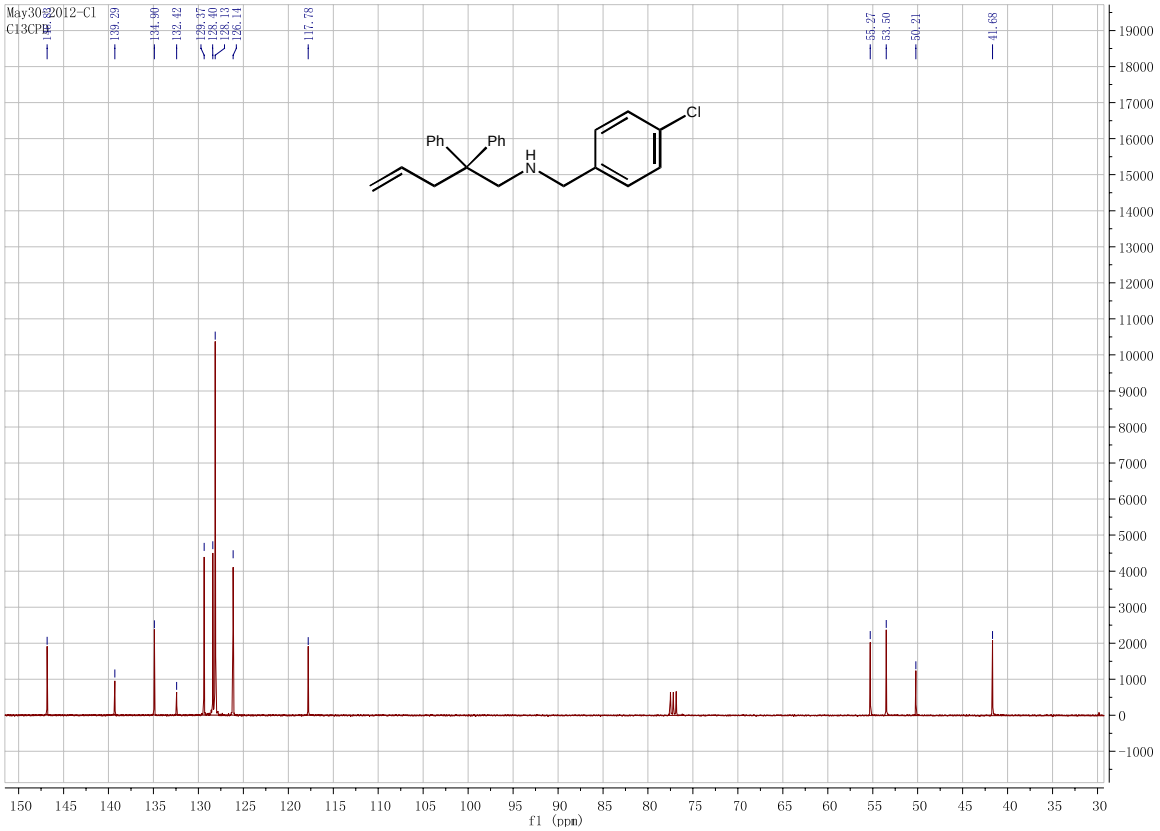
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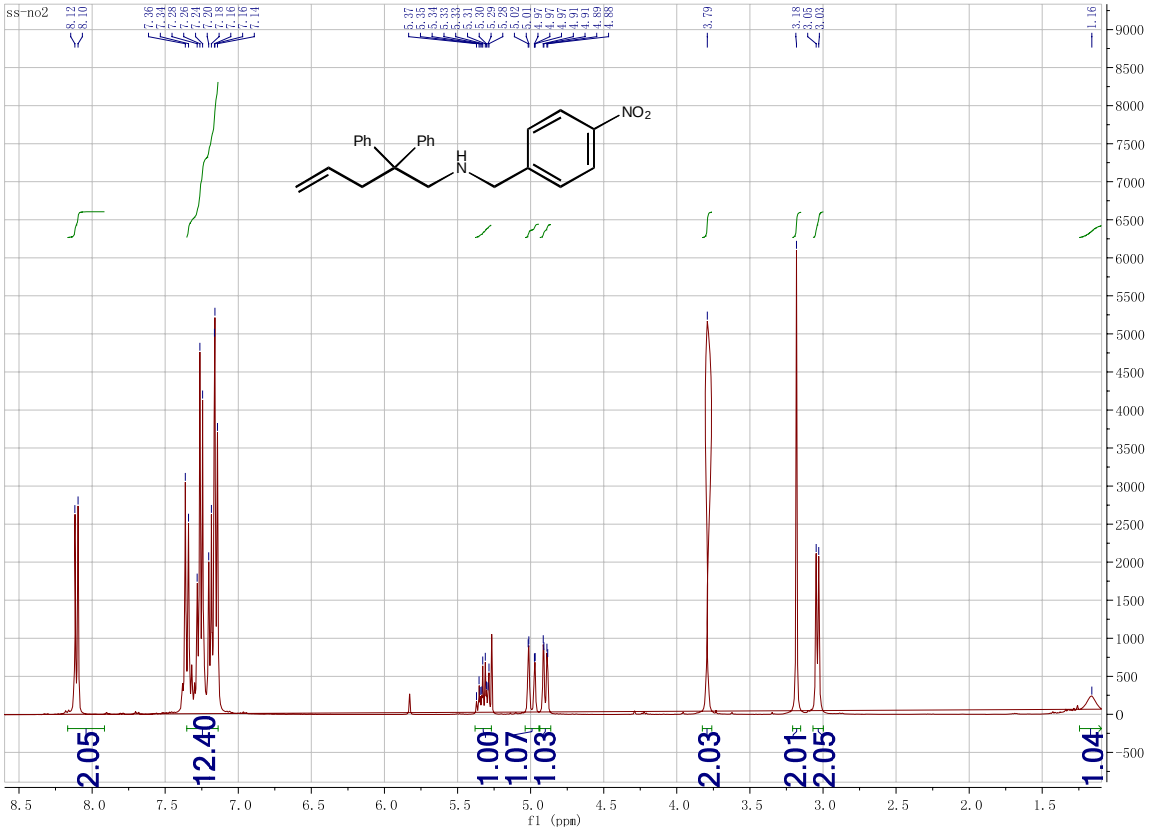
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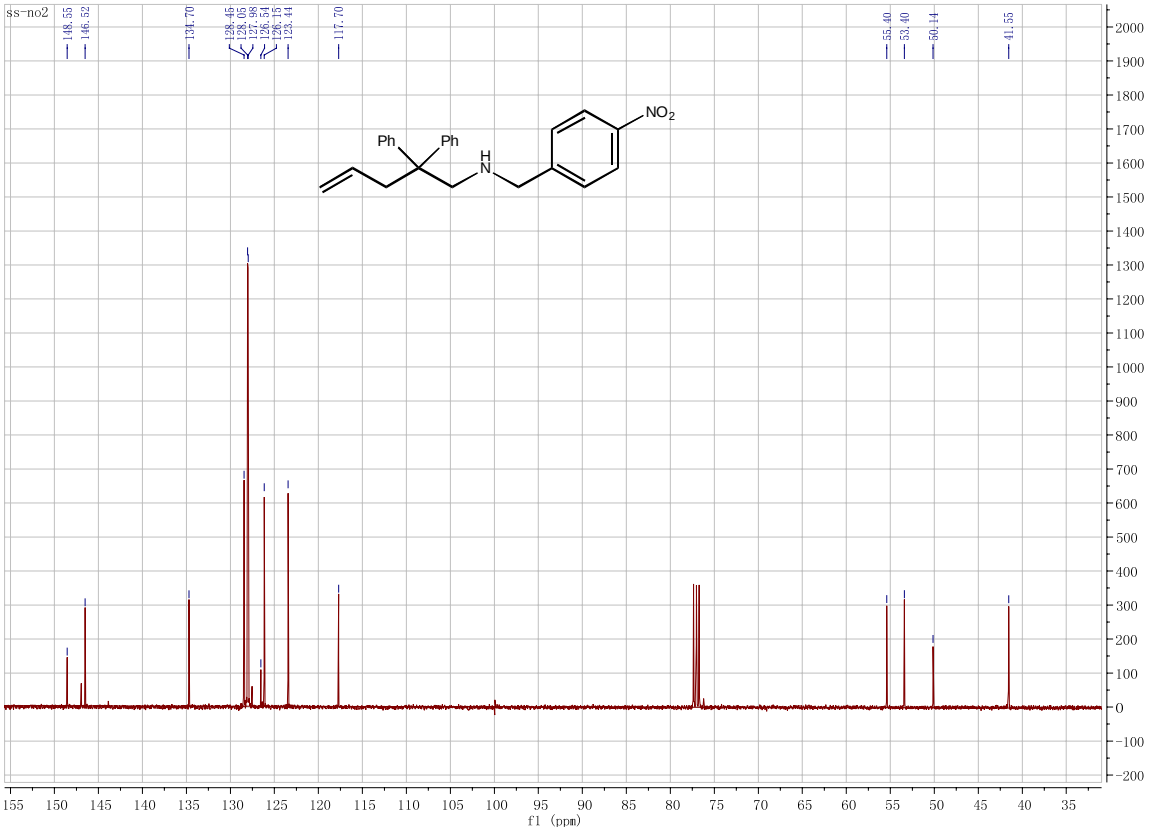
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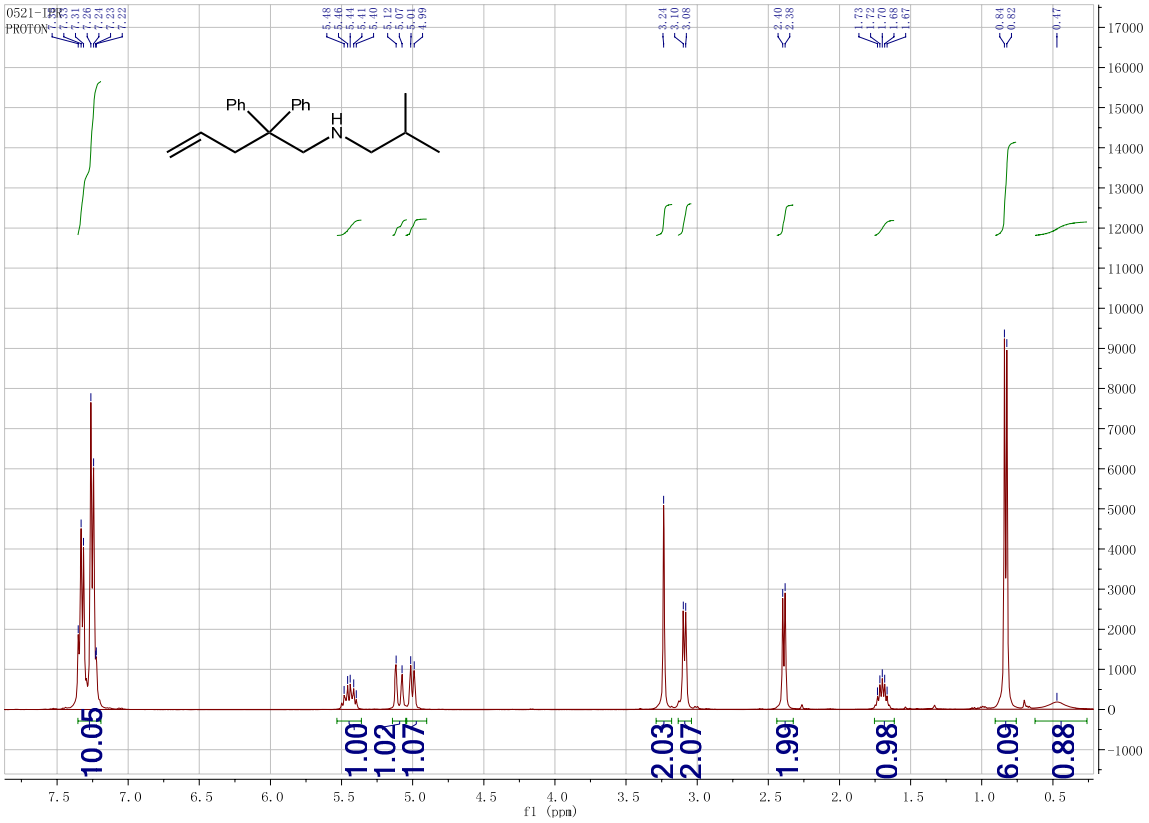
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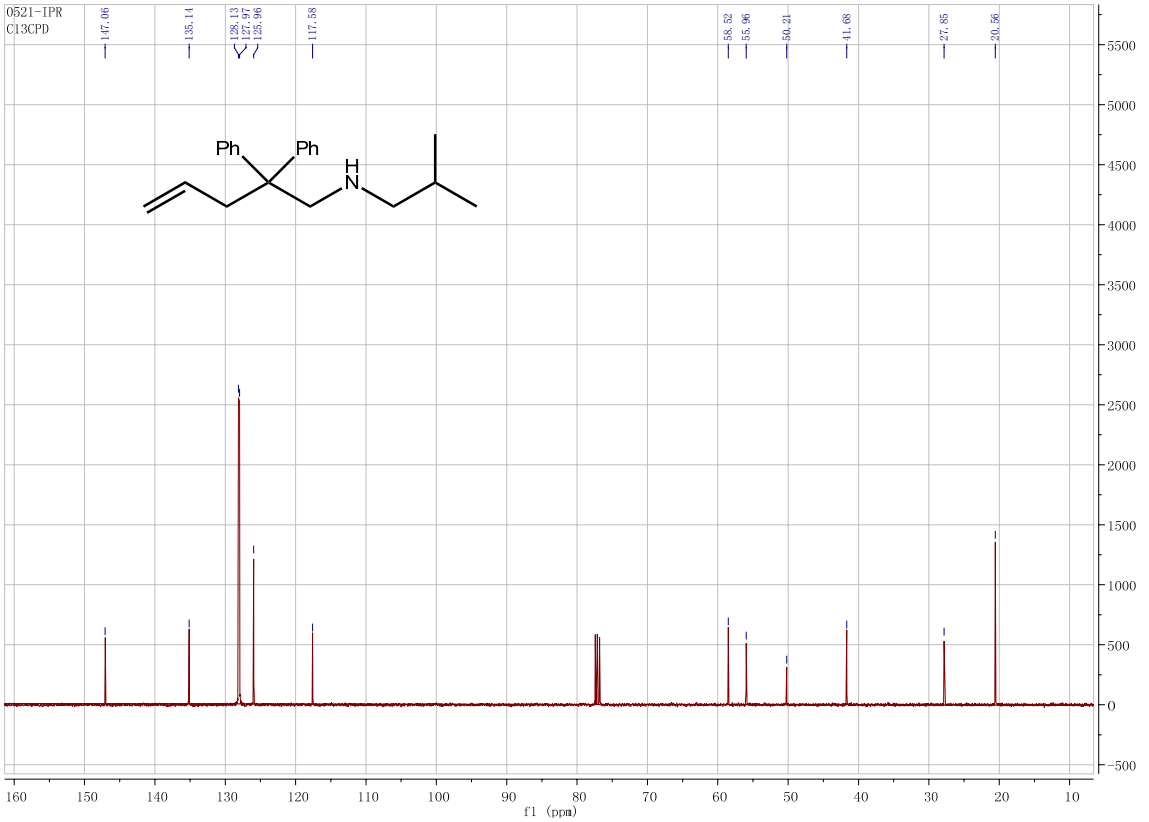
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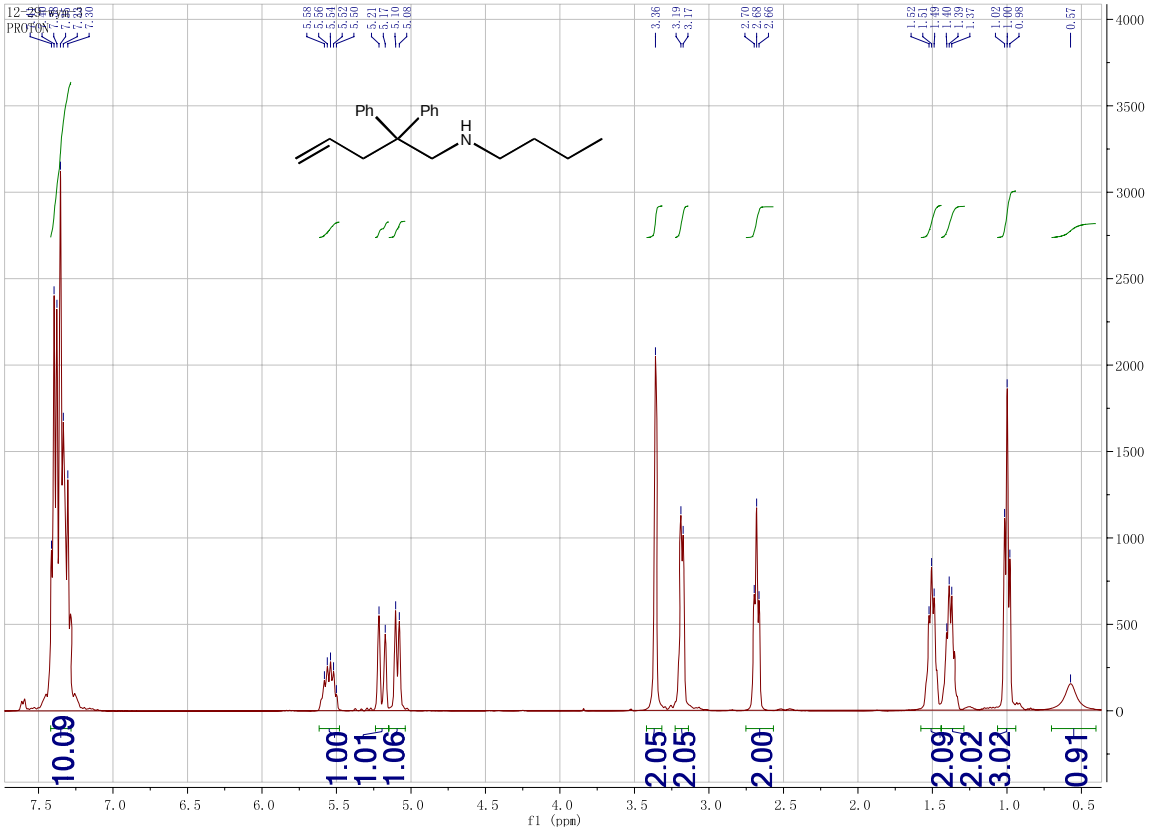
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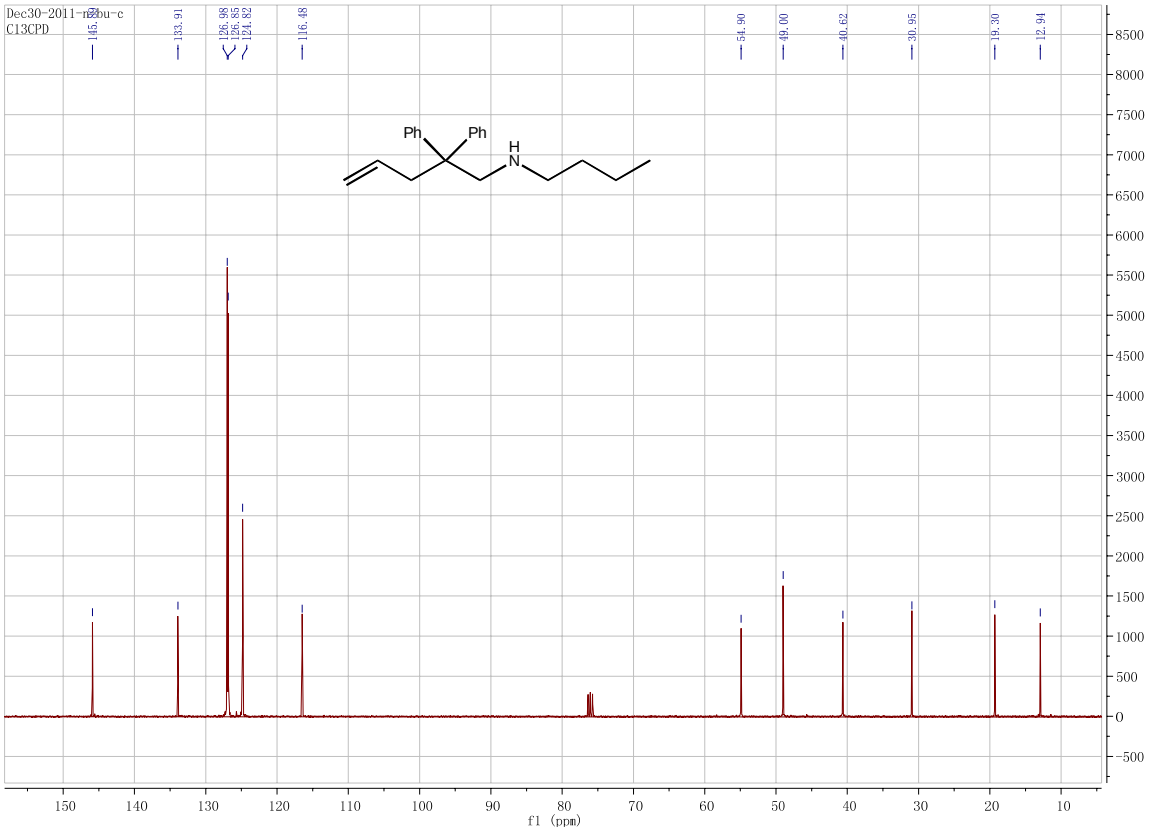
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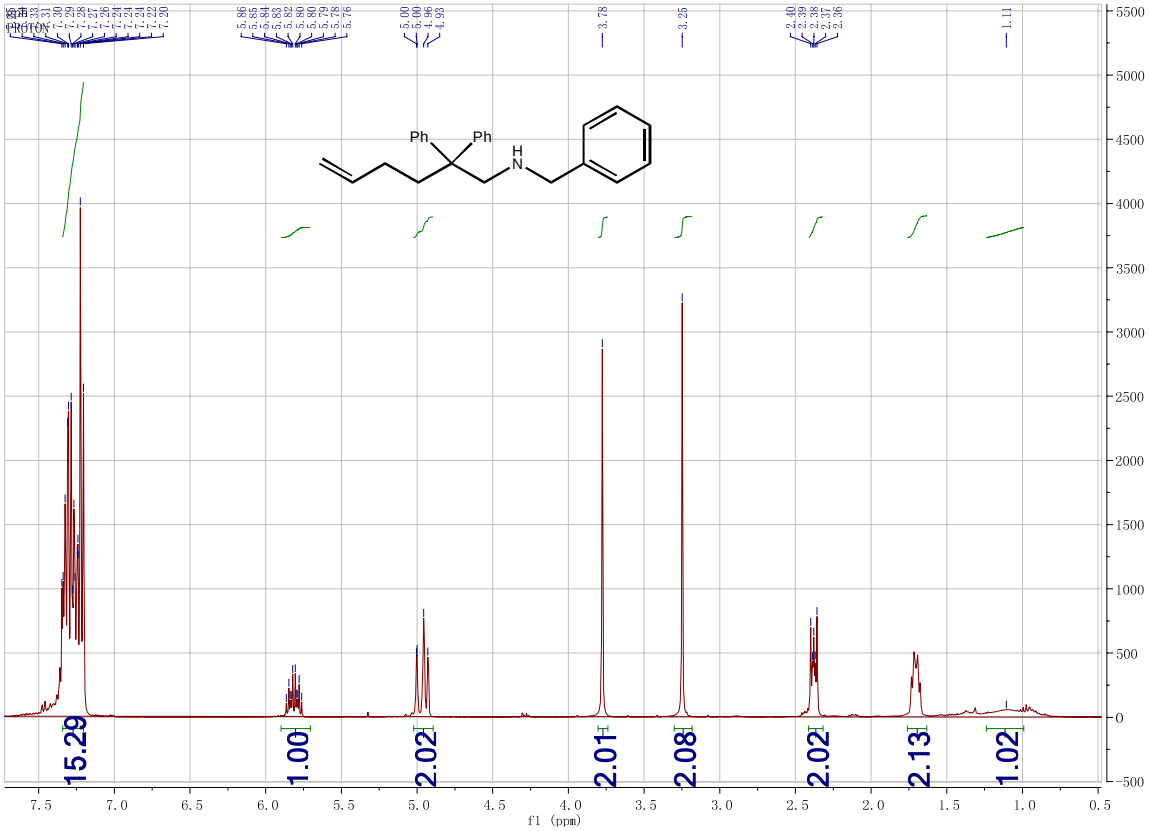
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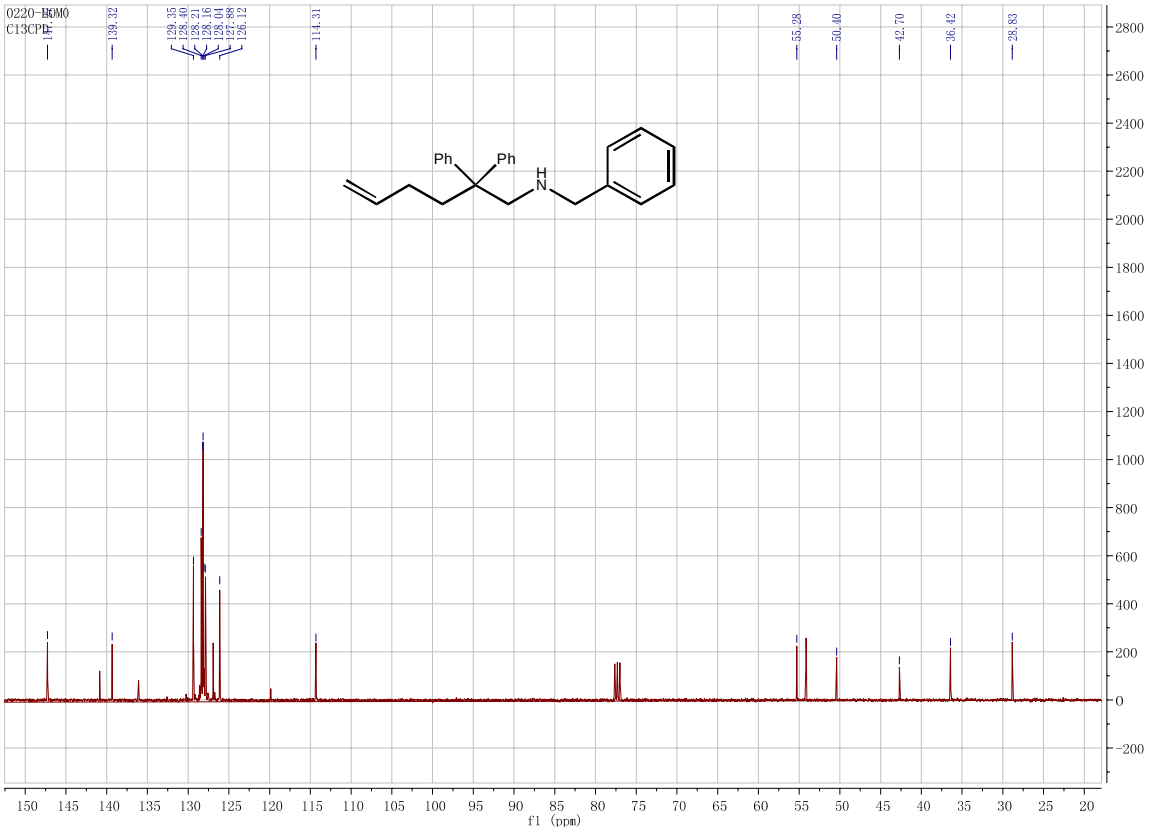
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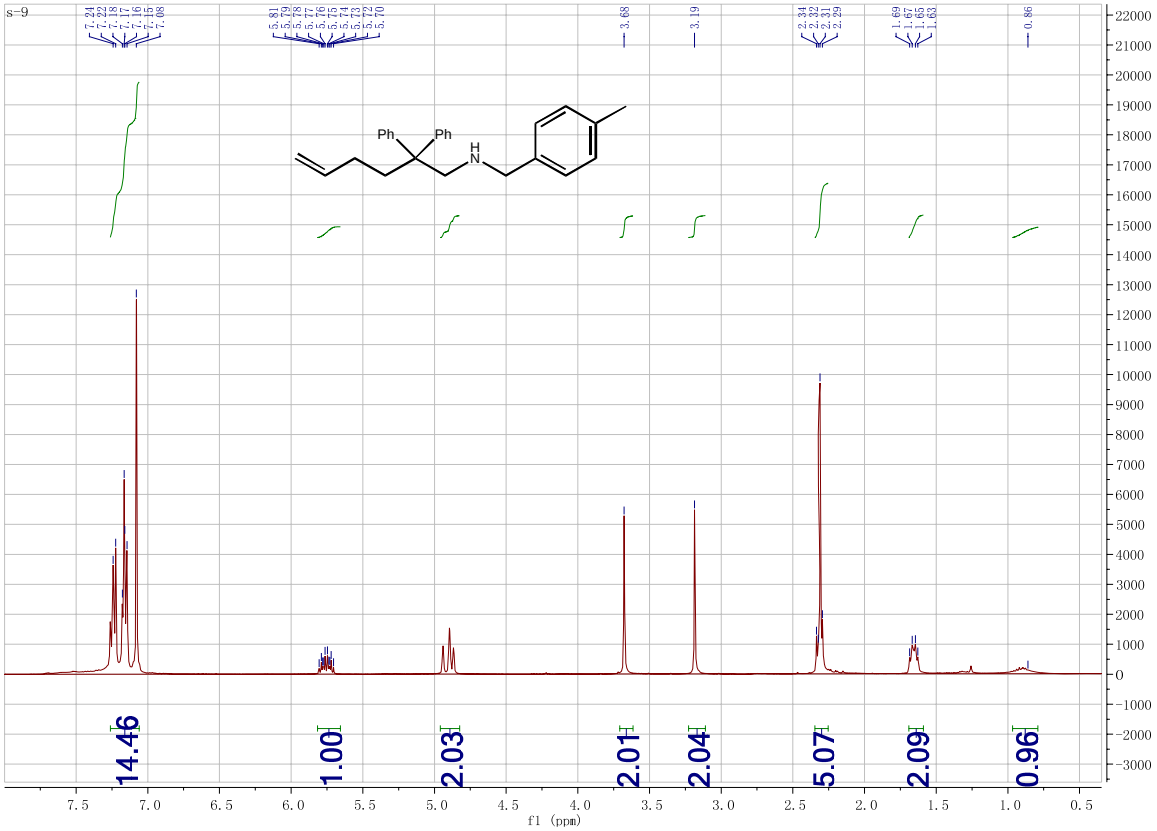
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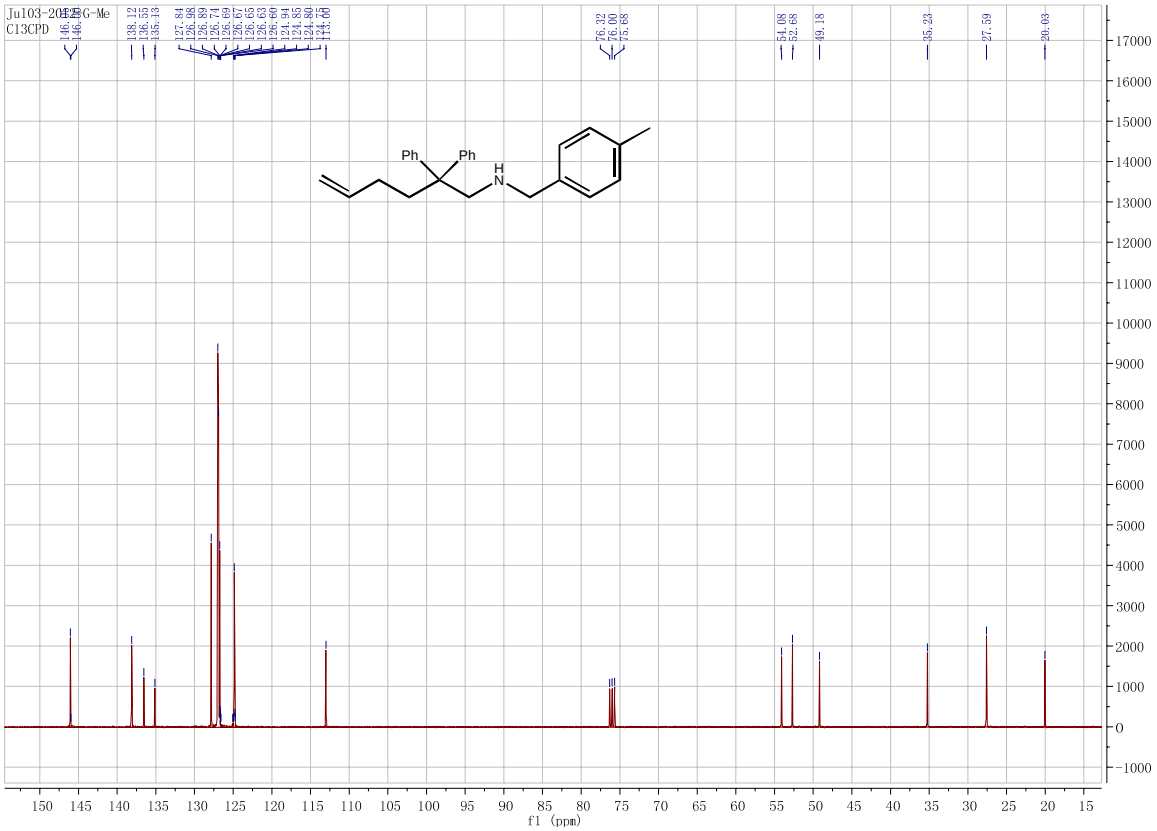
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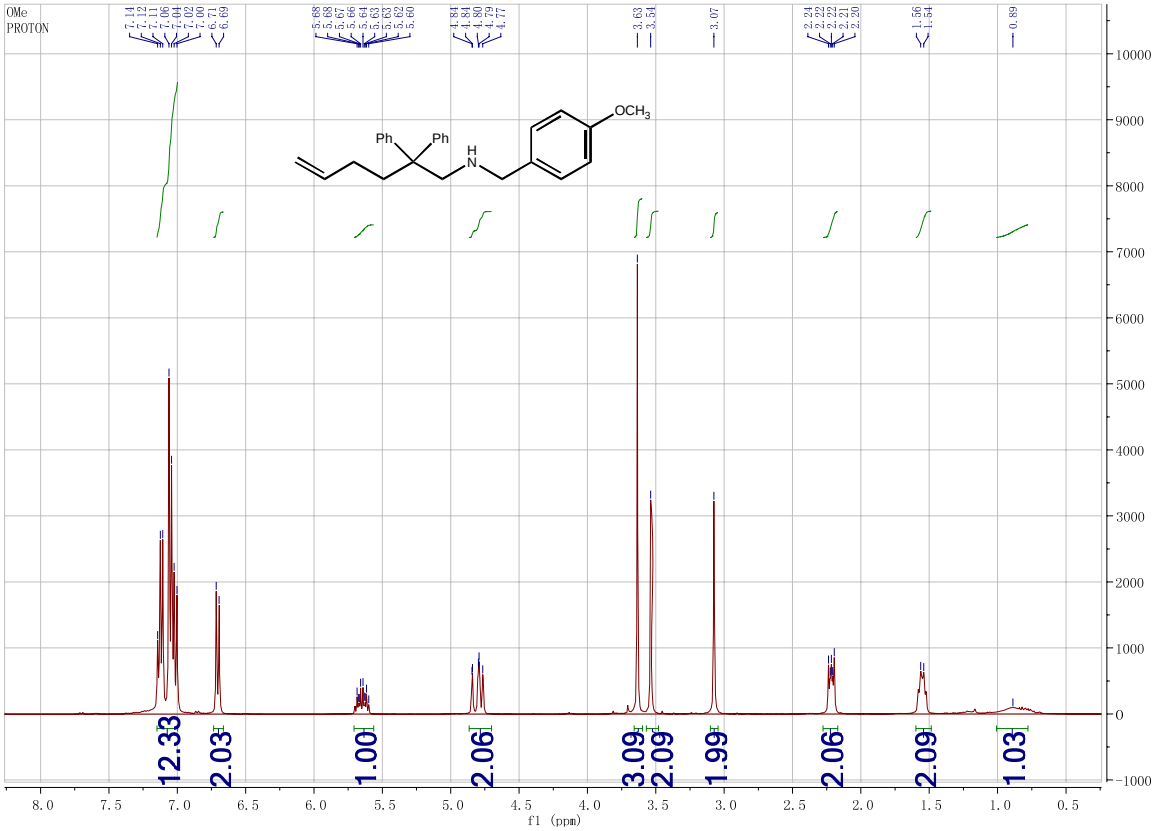
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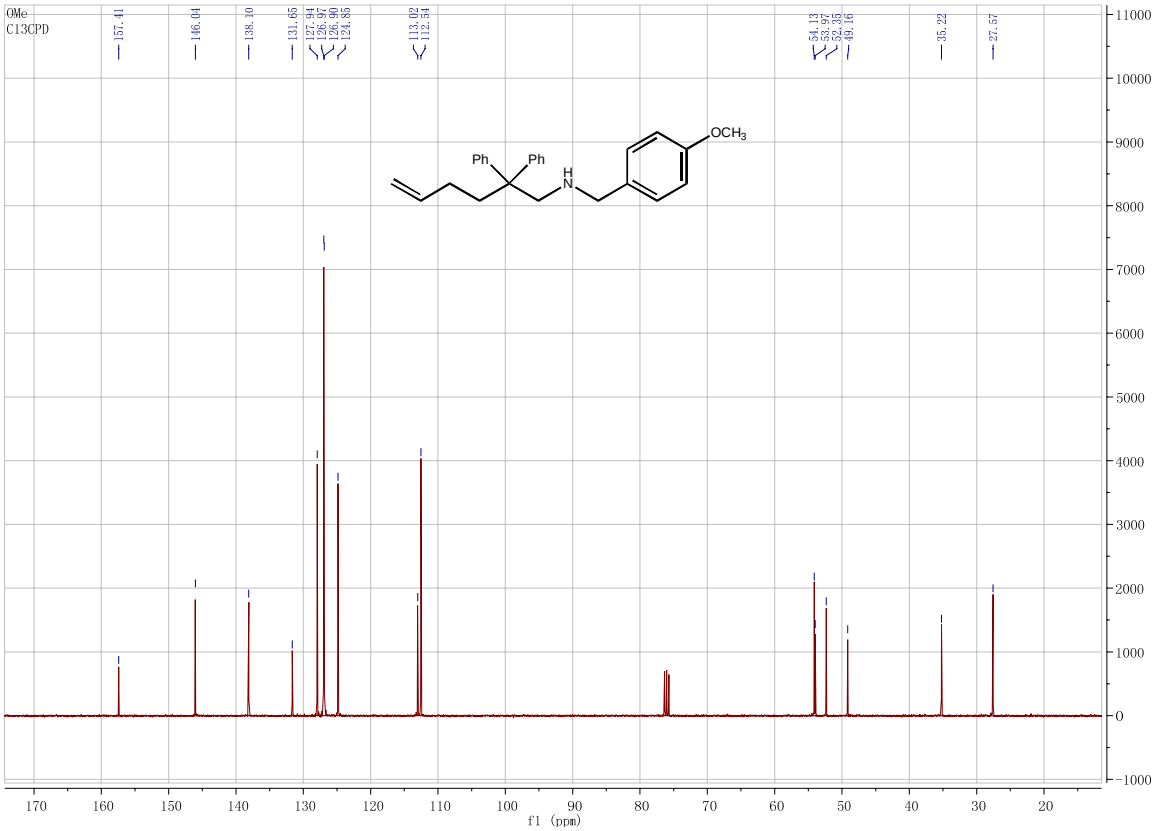
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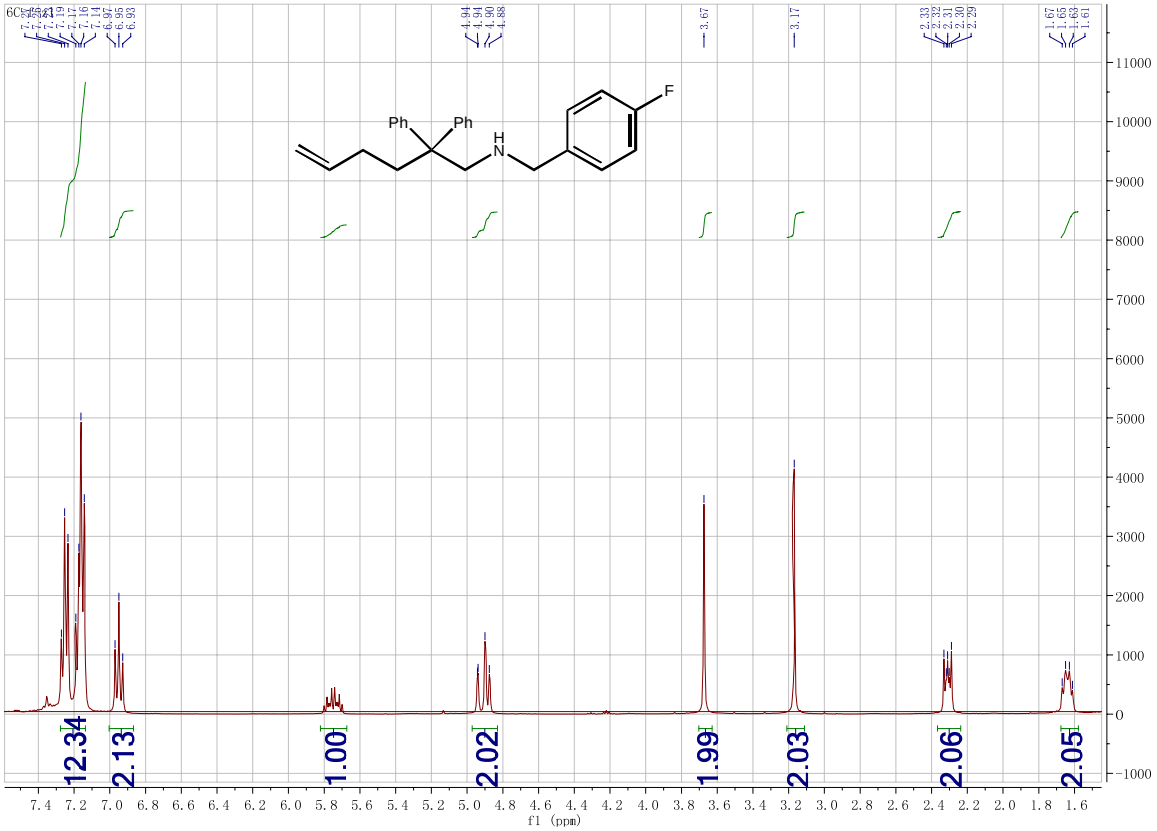
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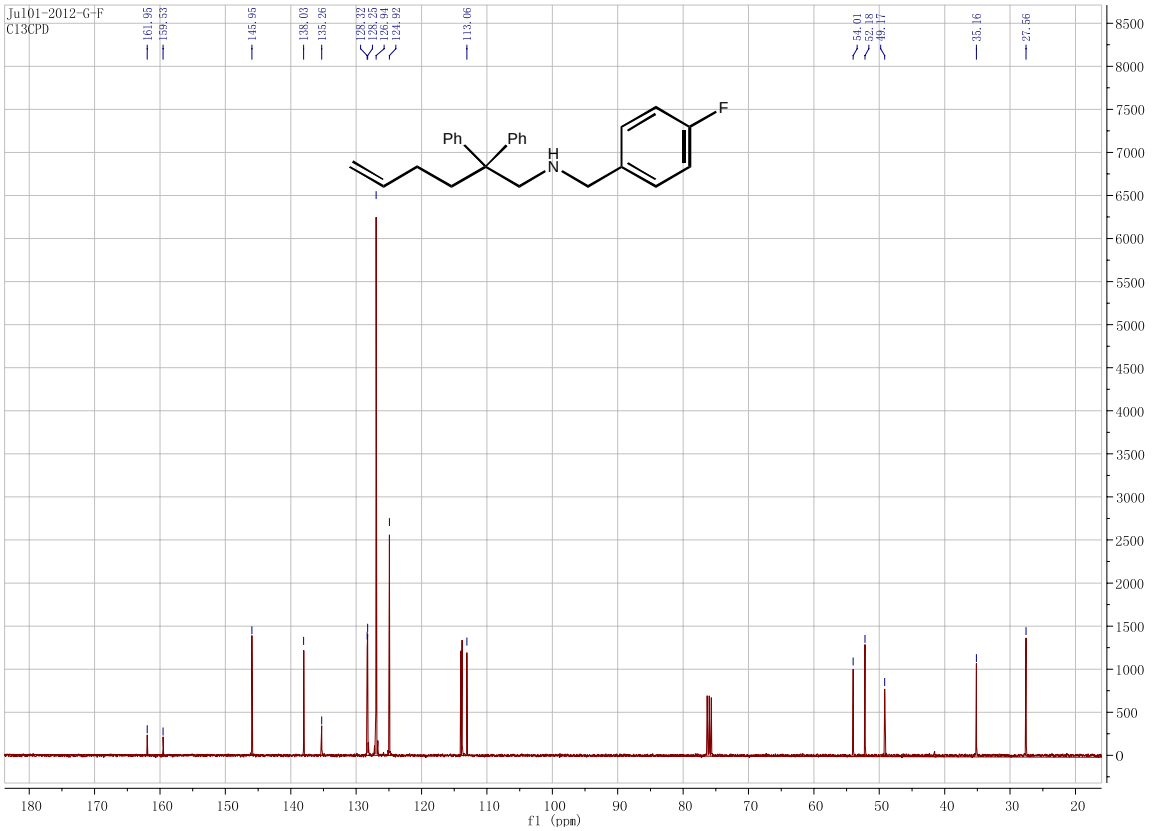
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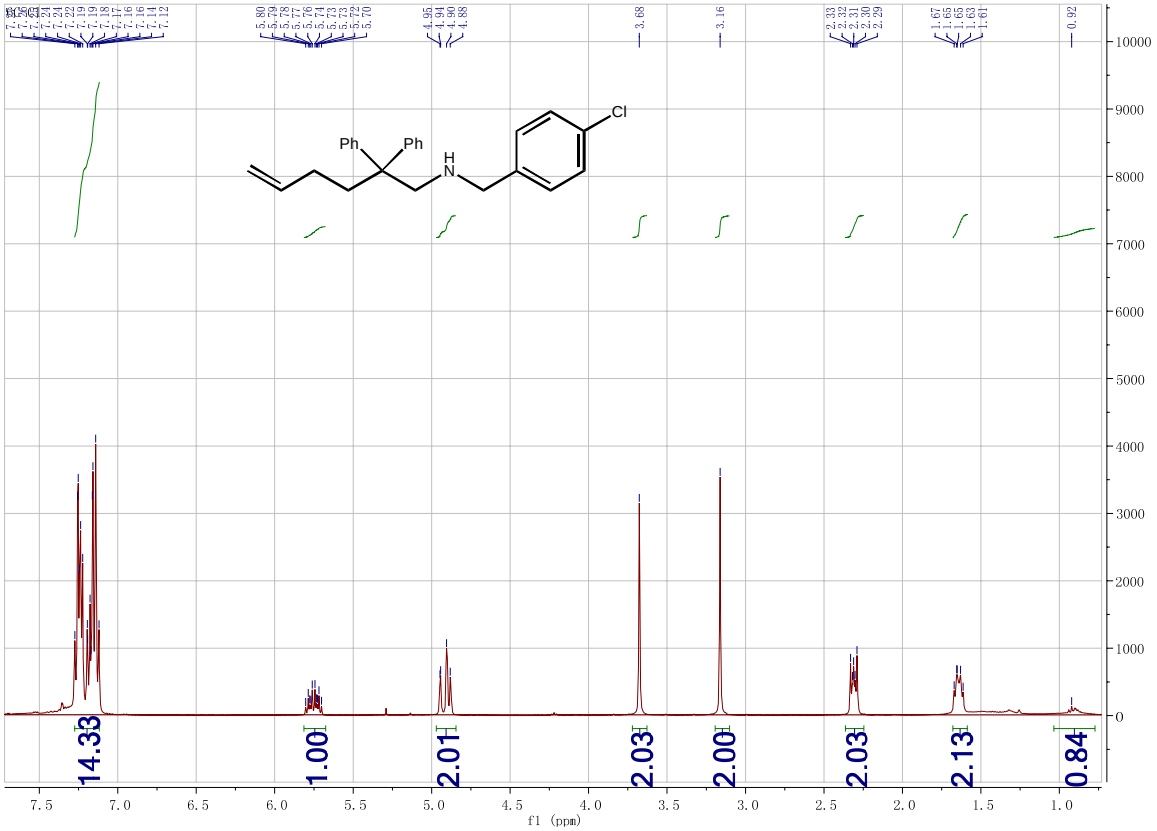
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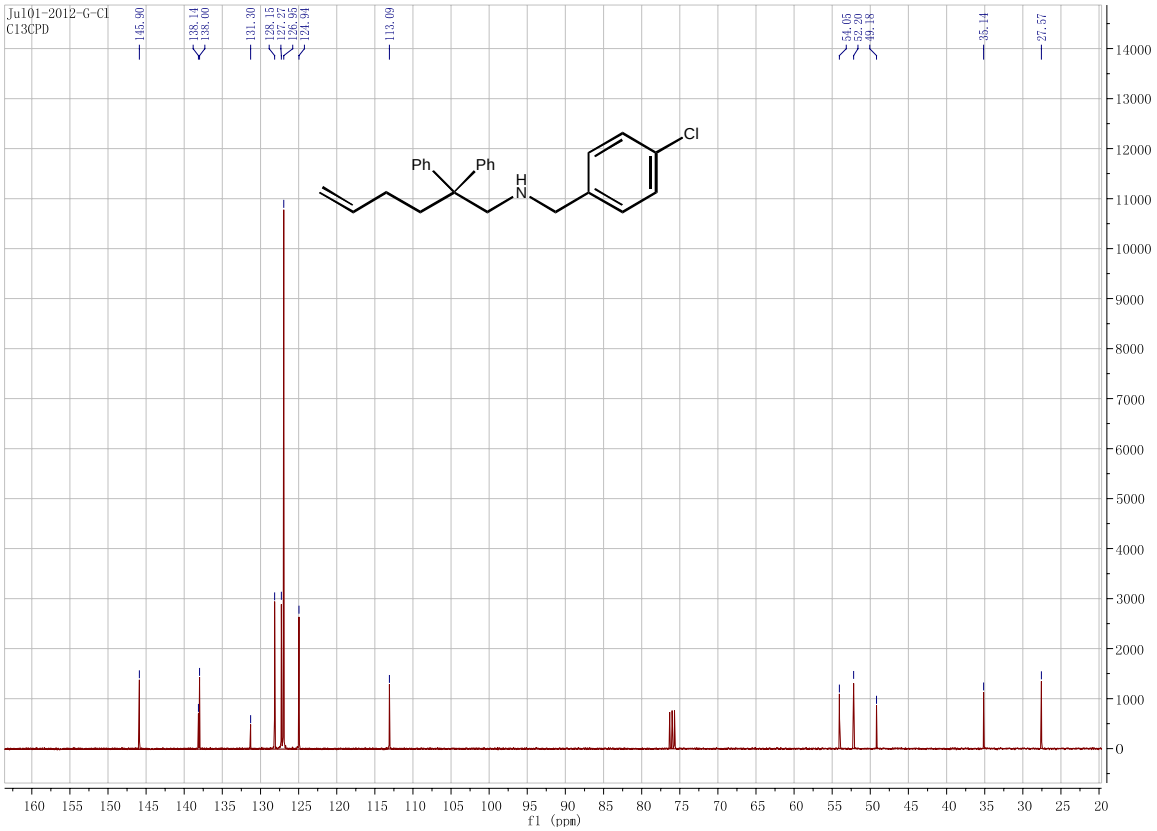
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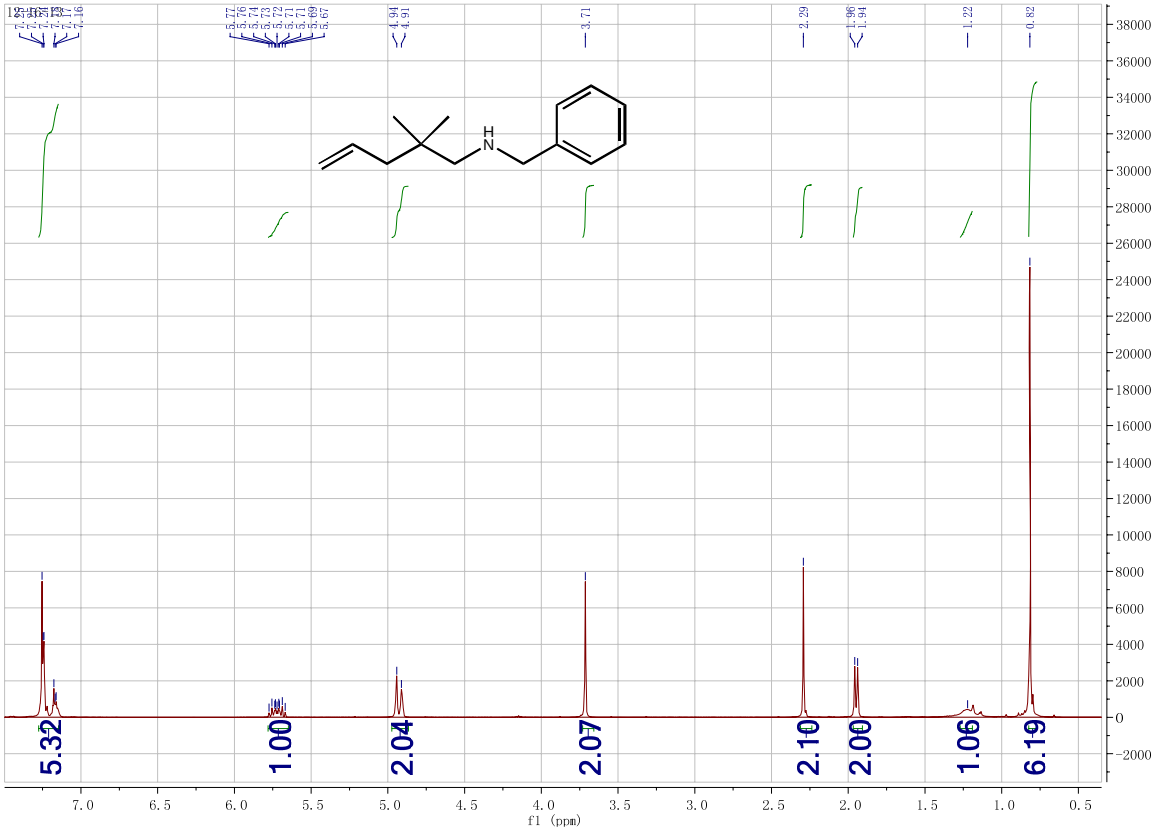
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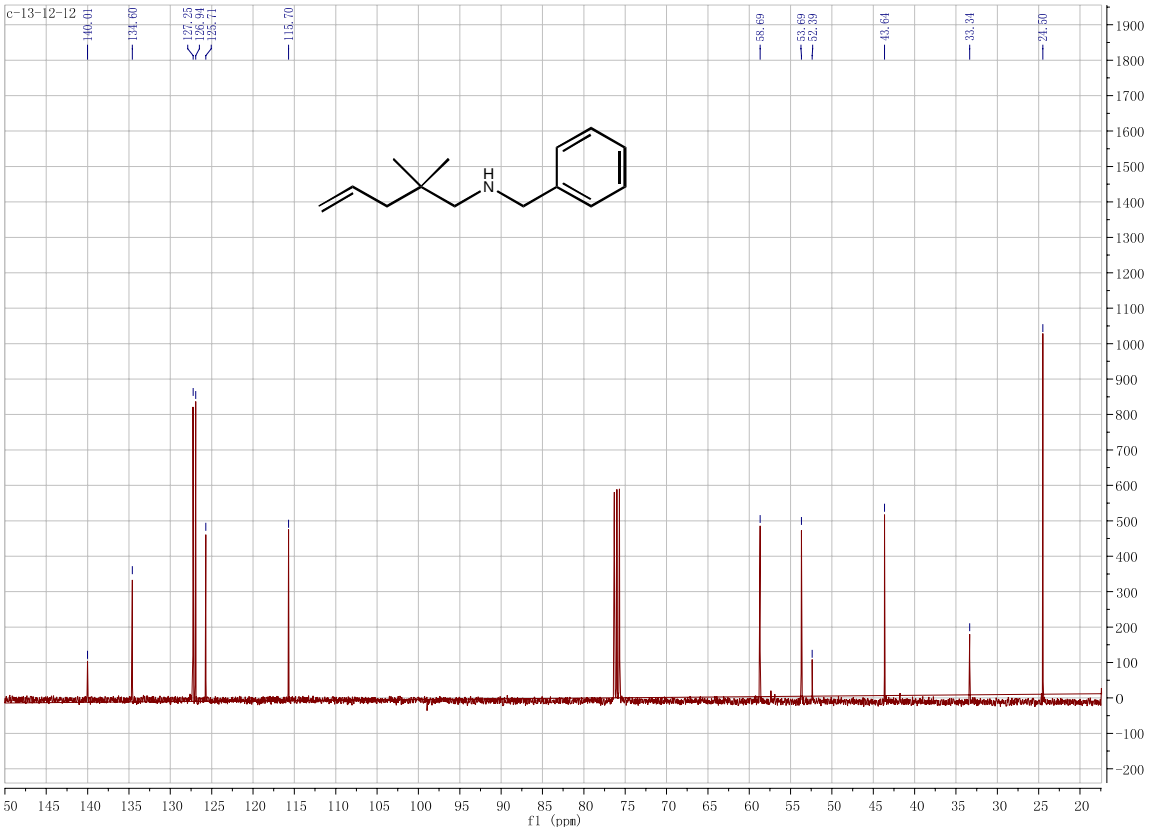
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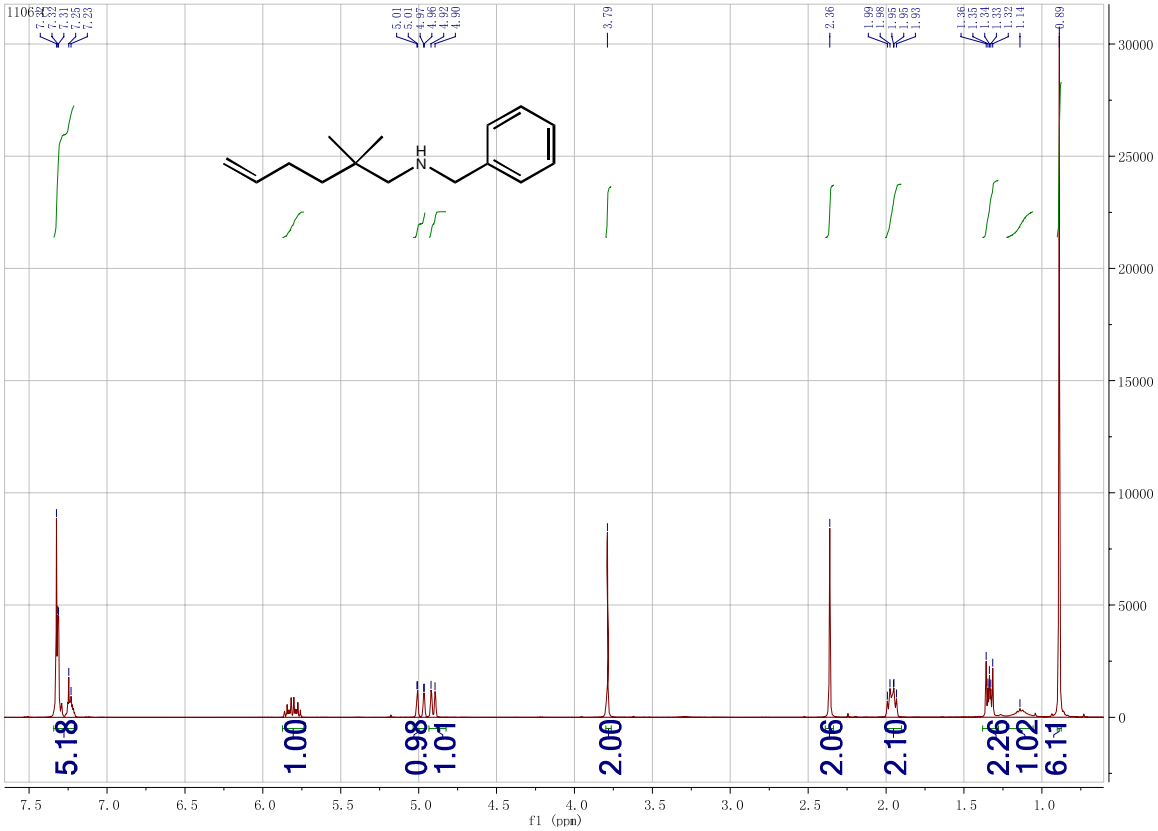
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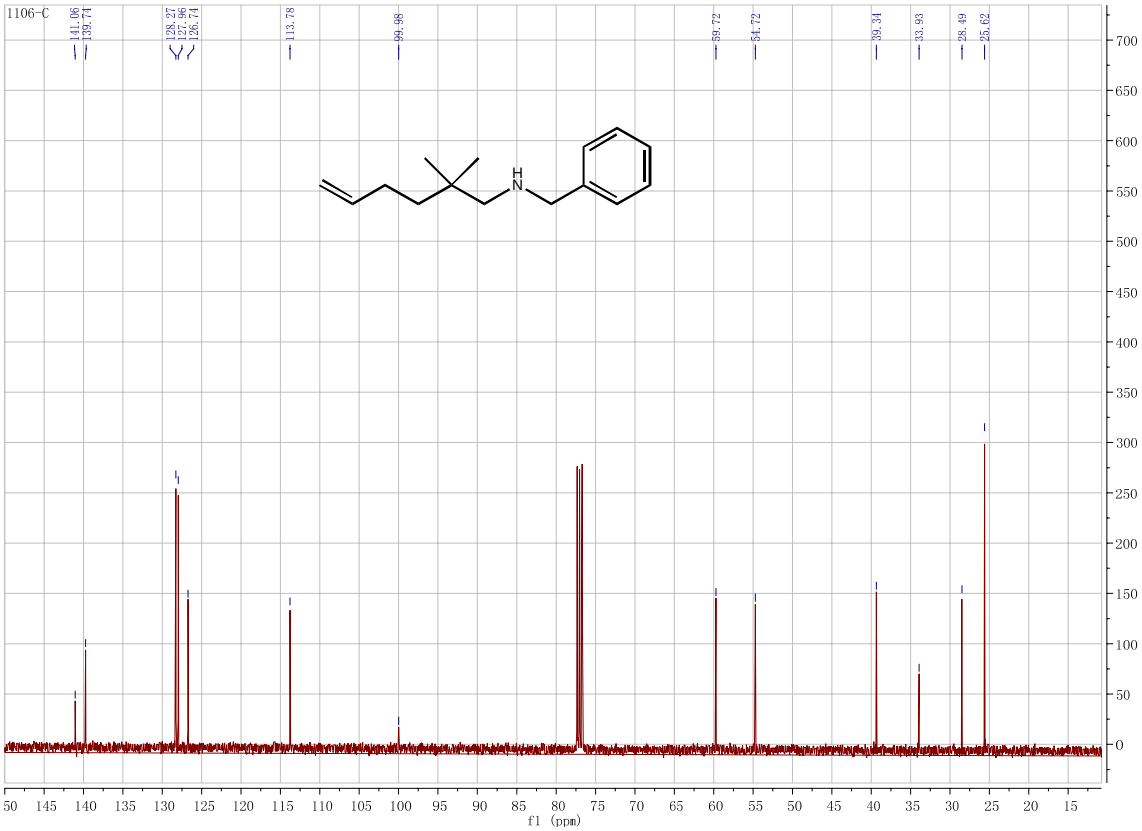
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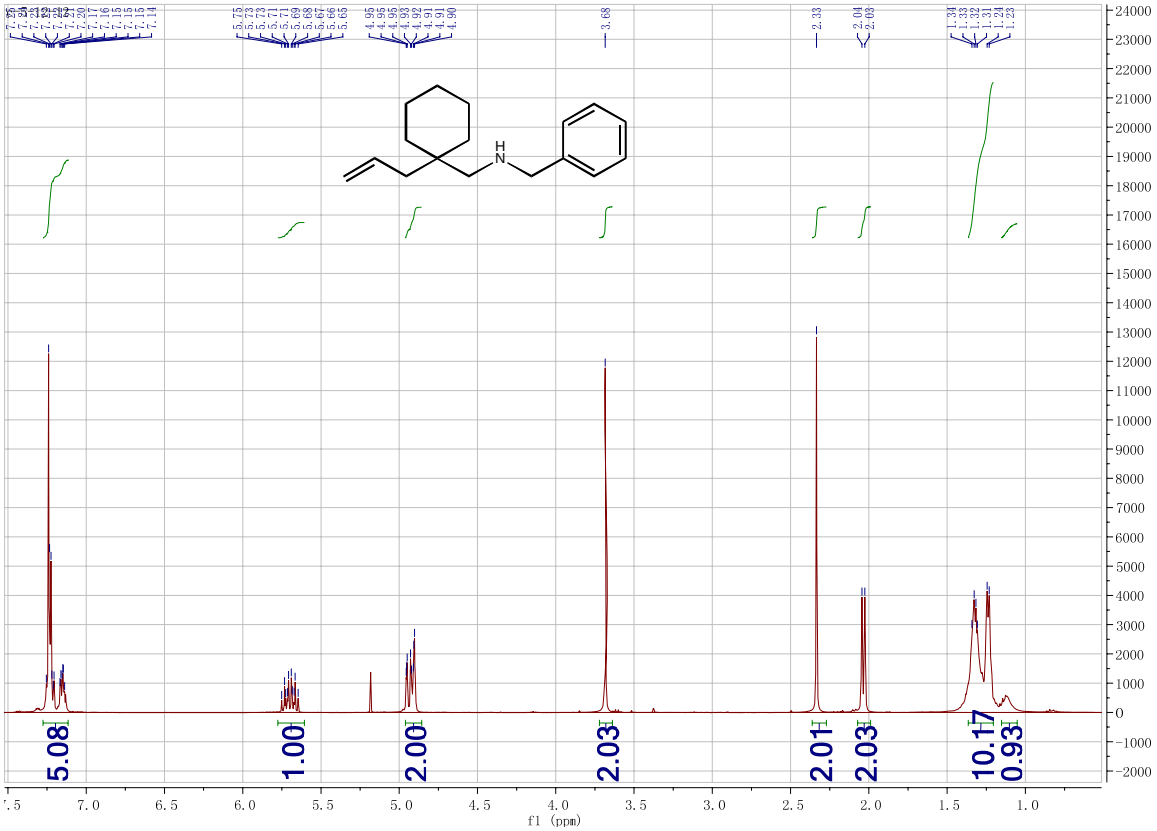
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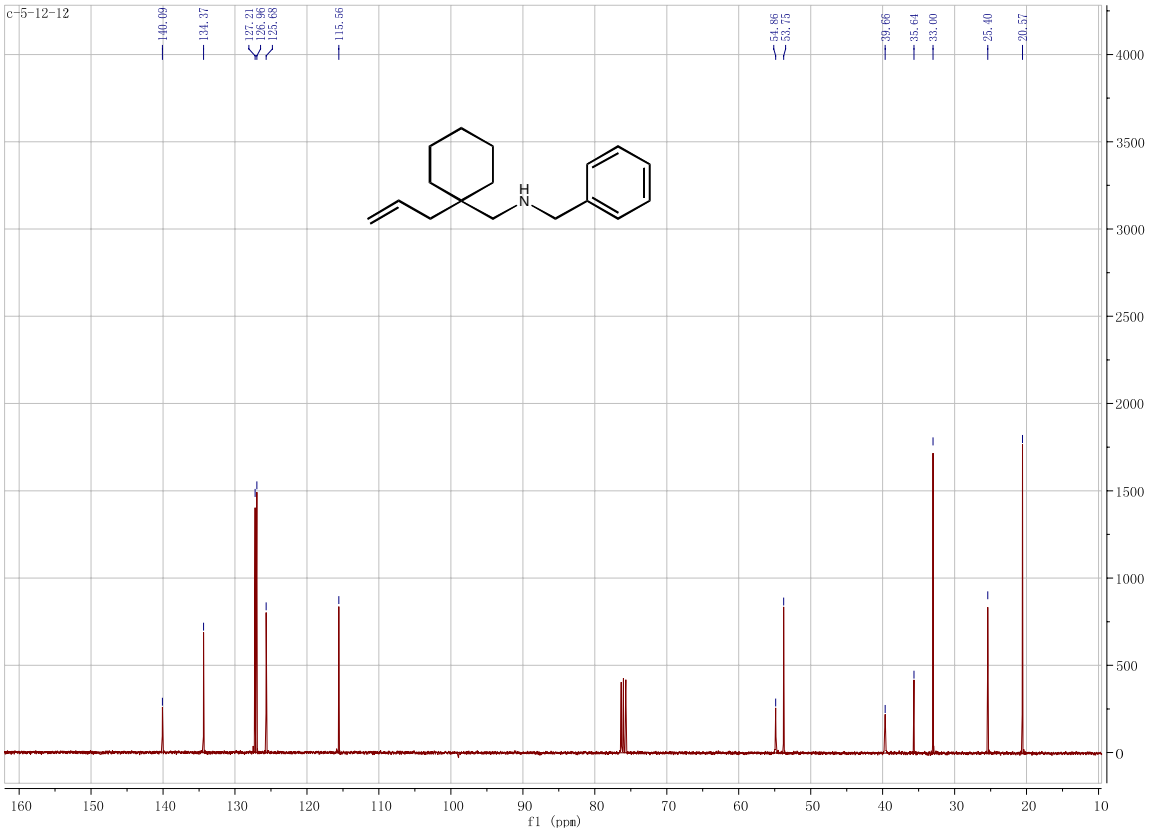
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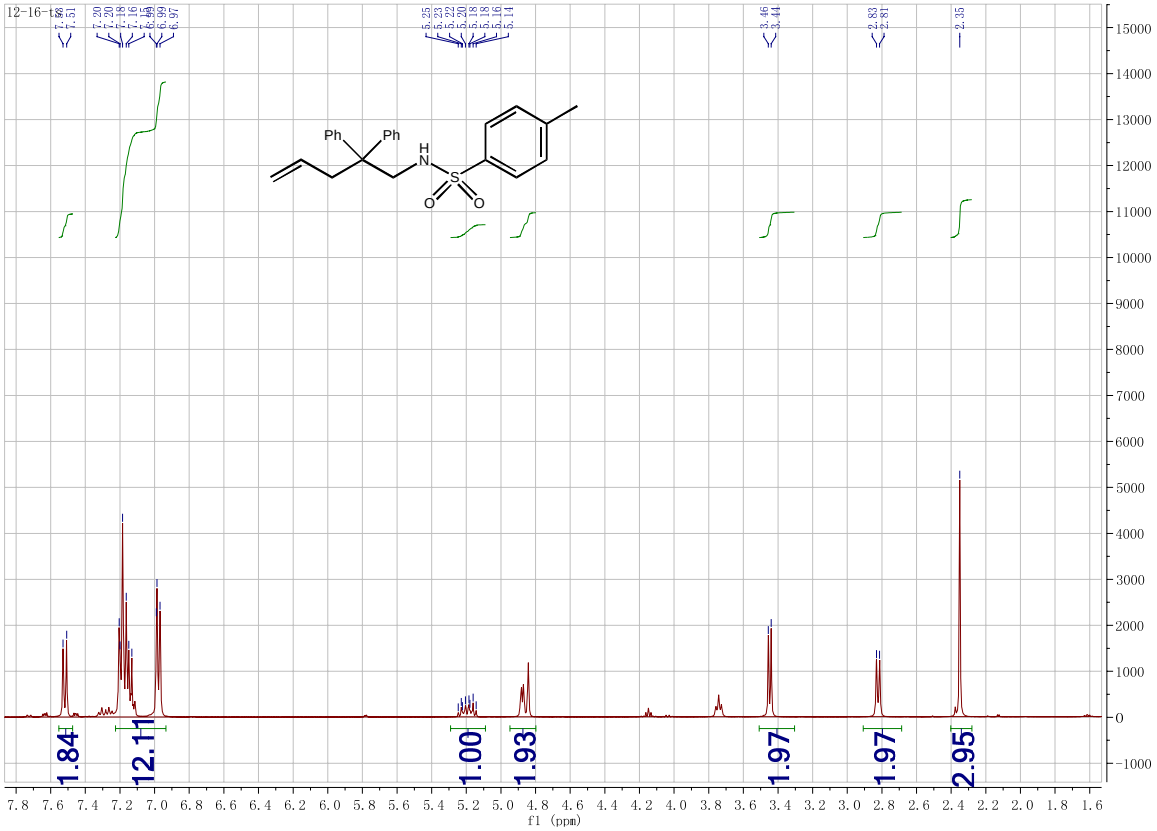
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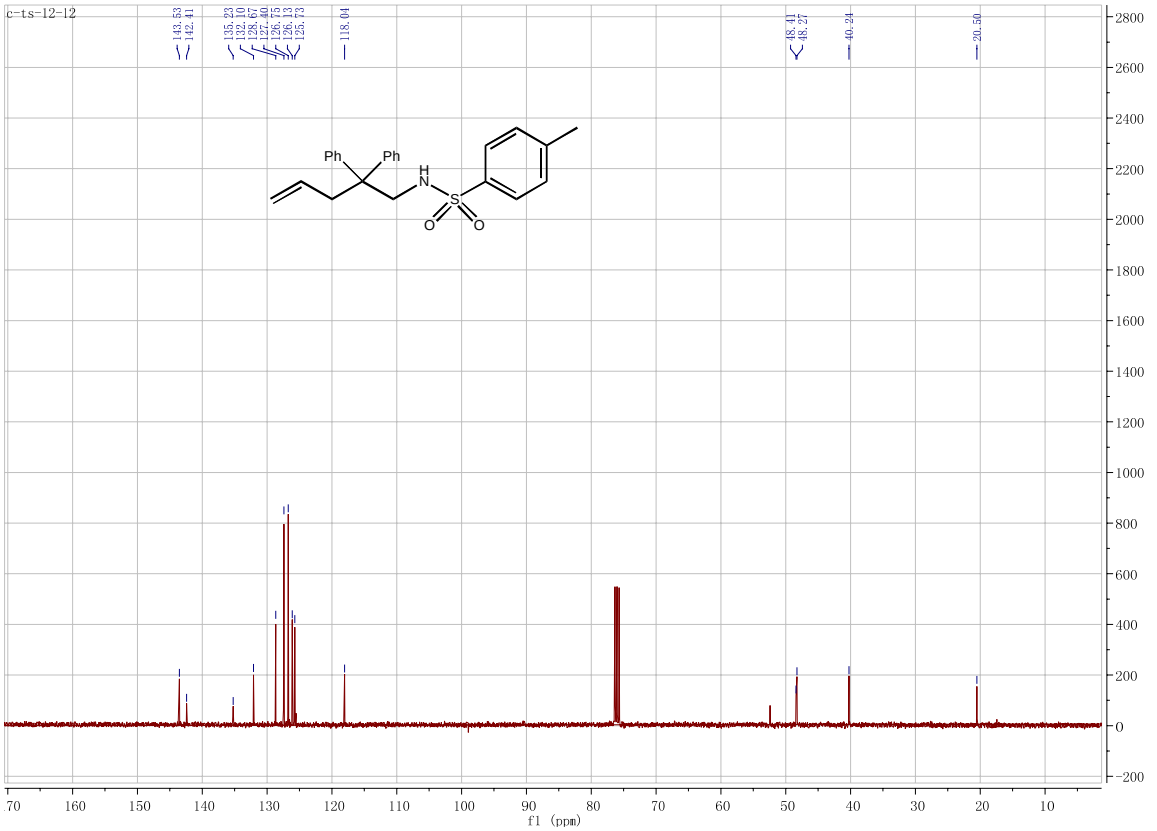
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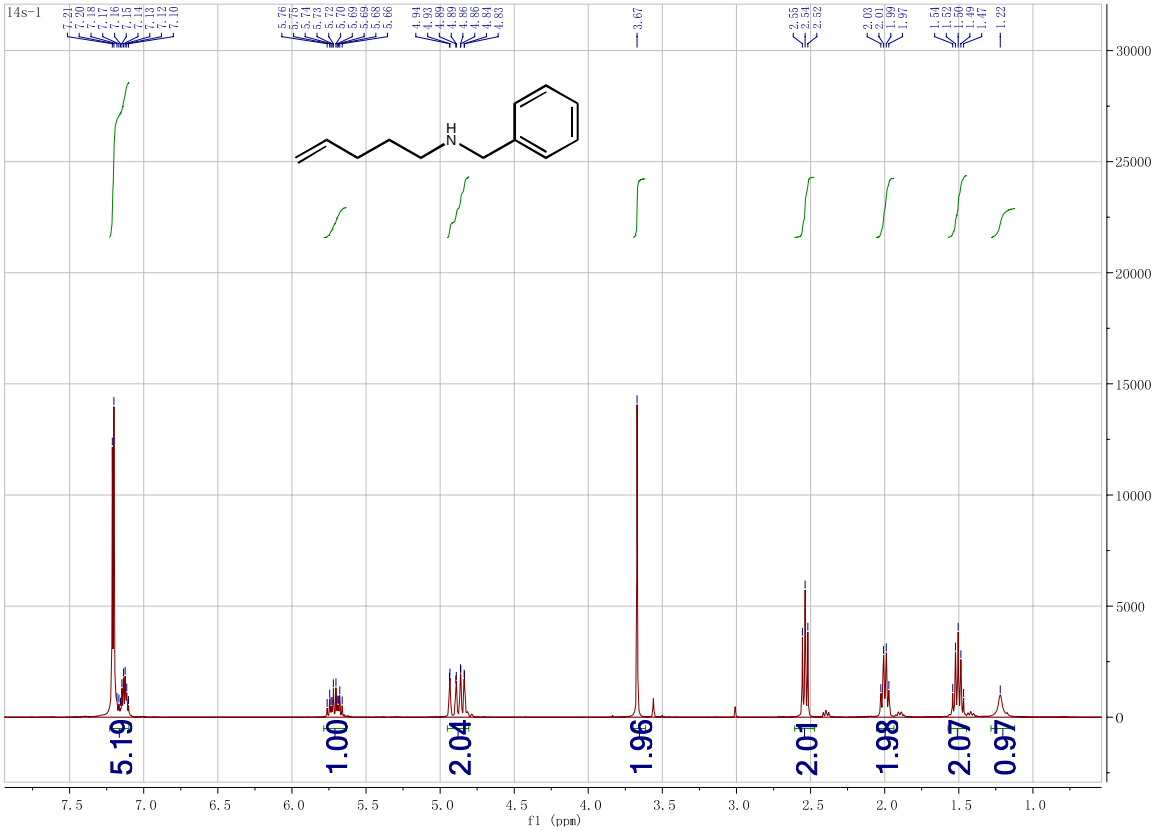
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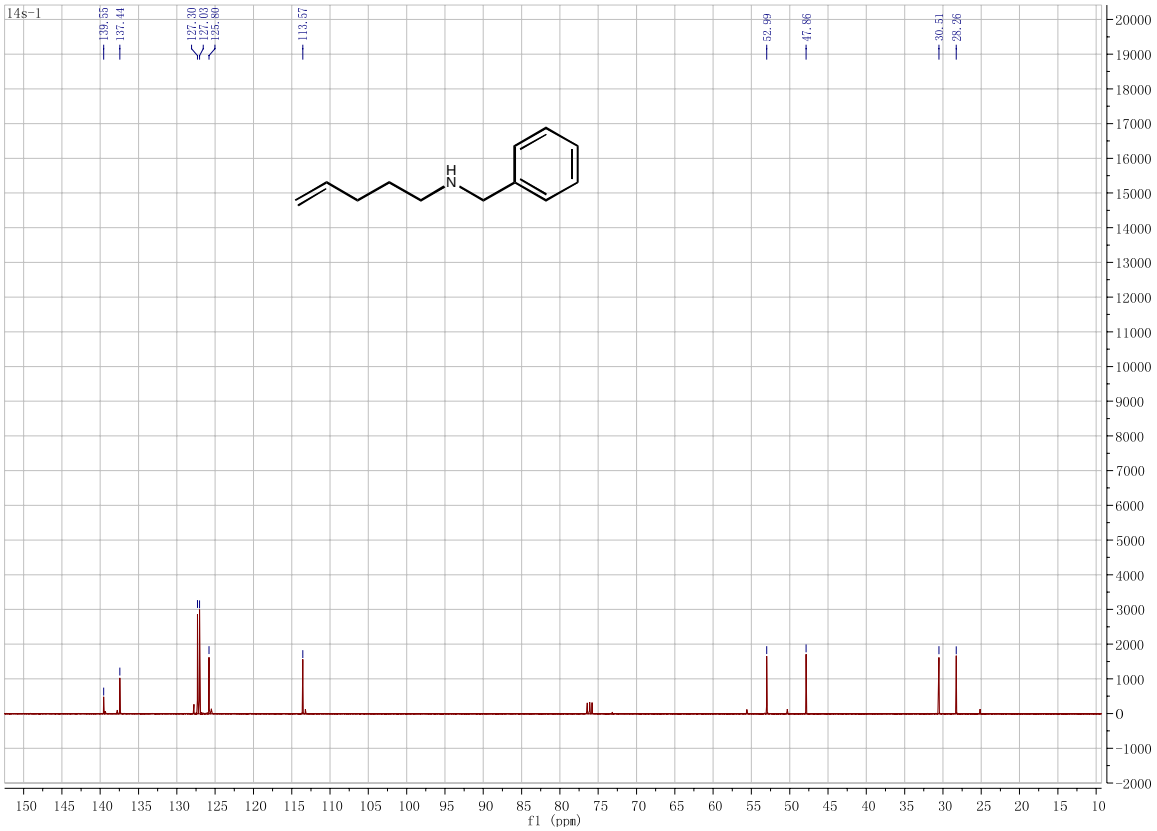
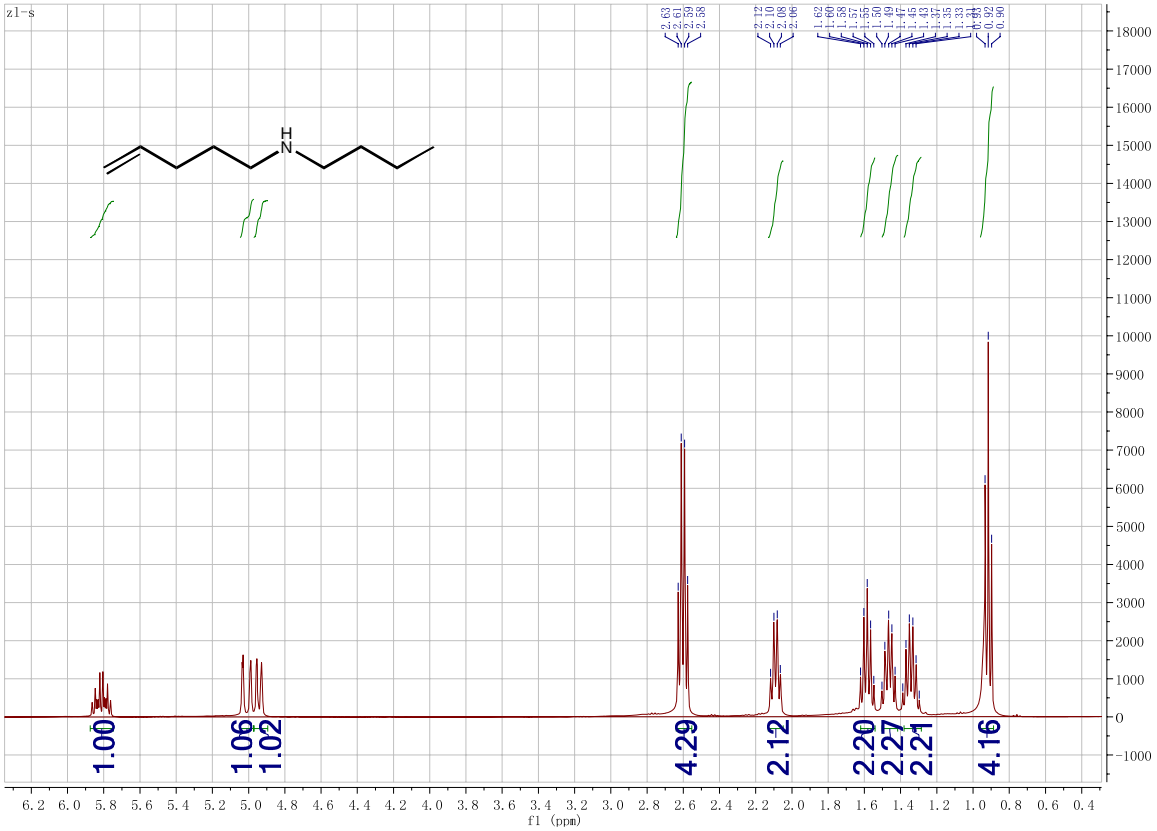
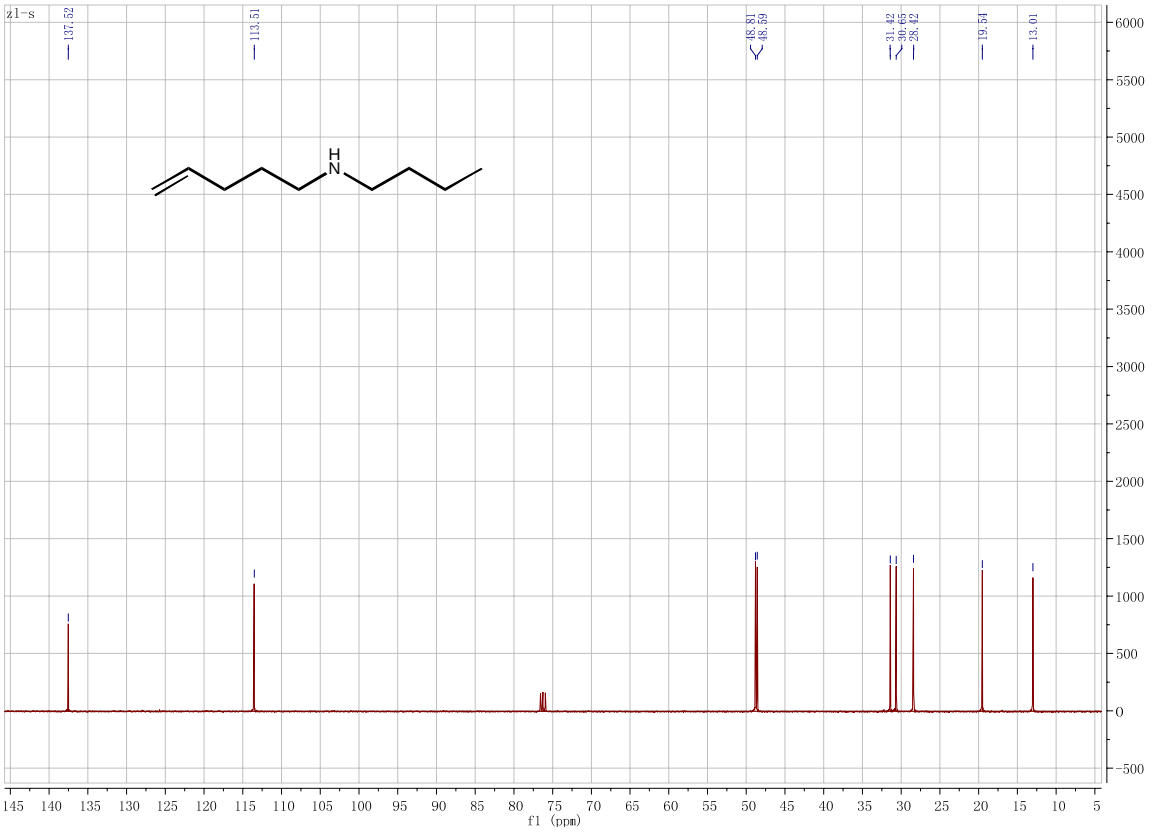
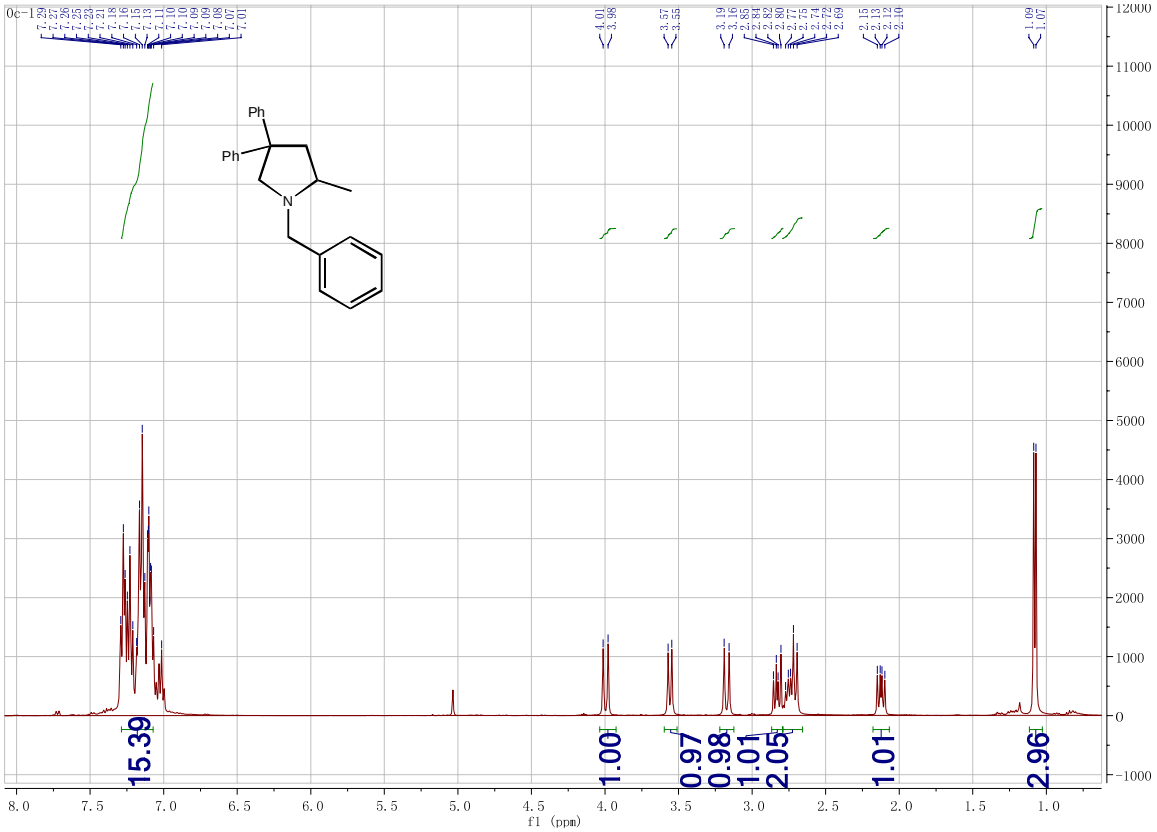
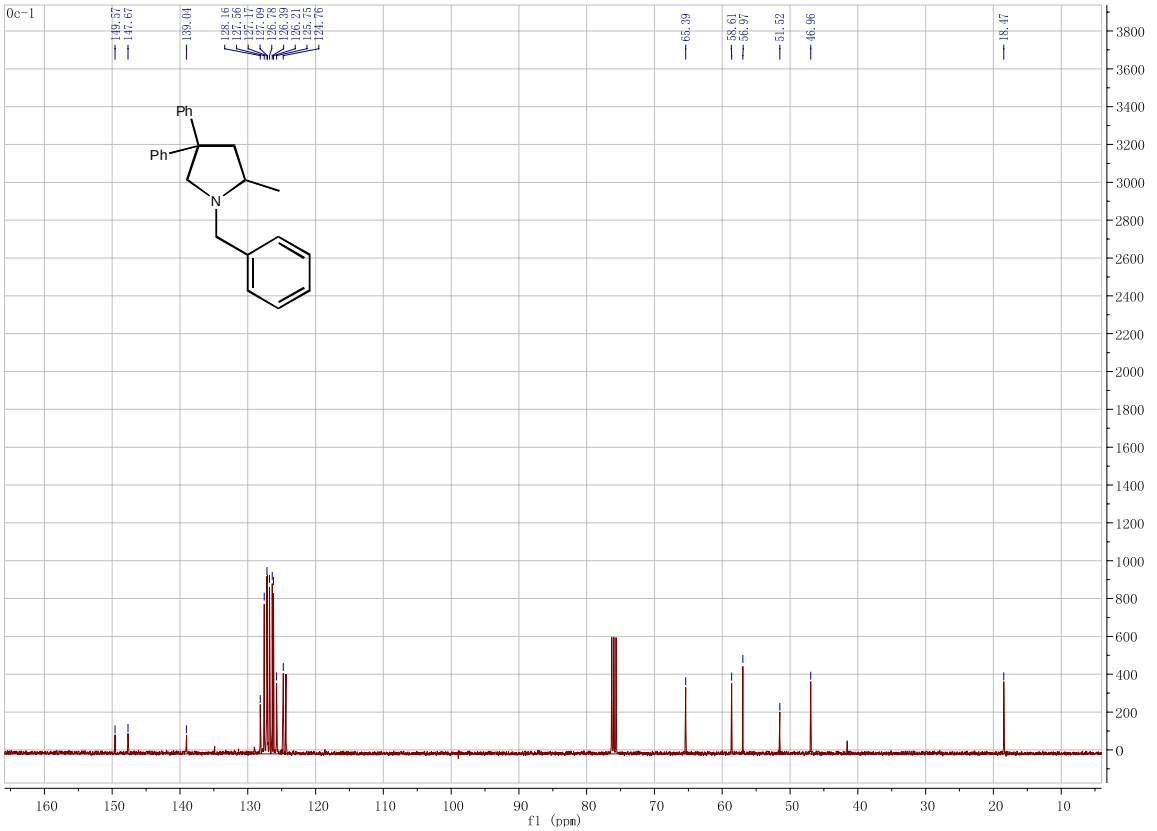
5.70
5.69
5.68
5.68
5.66
4.94
4.93
4.89
4.89
4.86
4.85
4.84
4.83
3.67
2.55
2.54
2.52
2.03
2.01
1.99
1.97
1.54
1.52
1.50
1.49
1.47
1.22
5.19
1.00
2.04
1.96
2.01
1.98
2.07
0.97
f1 (ppm)

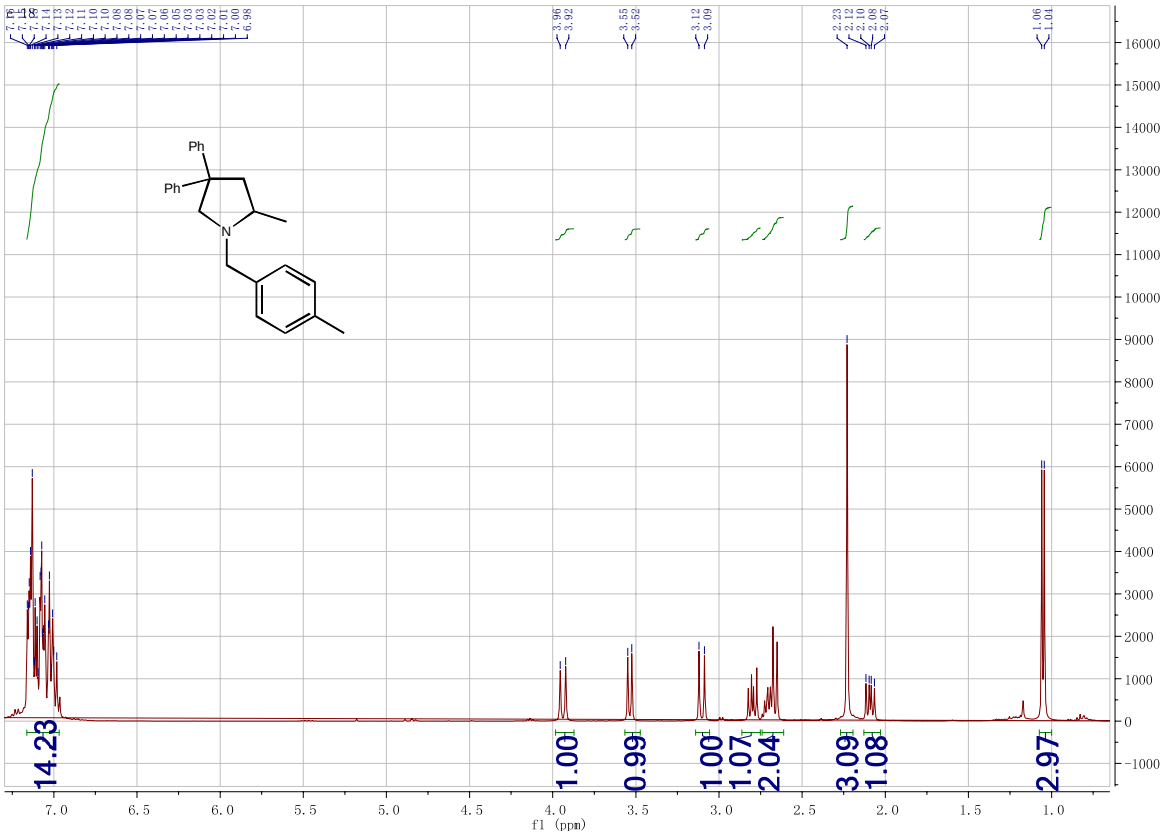

14s-1
139.55
137.44
127.30
127.03
126.80
113.57
52.99
47.86
30.51
28.26
f1 (ppm)

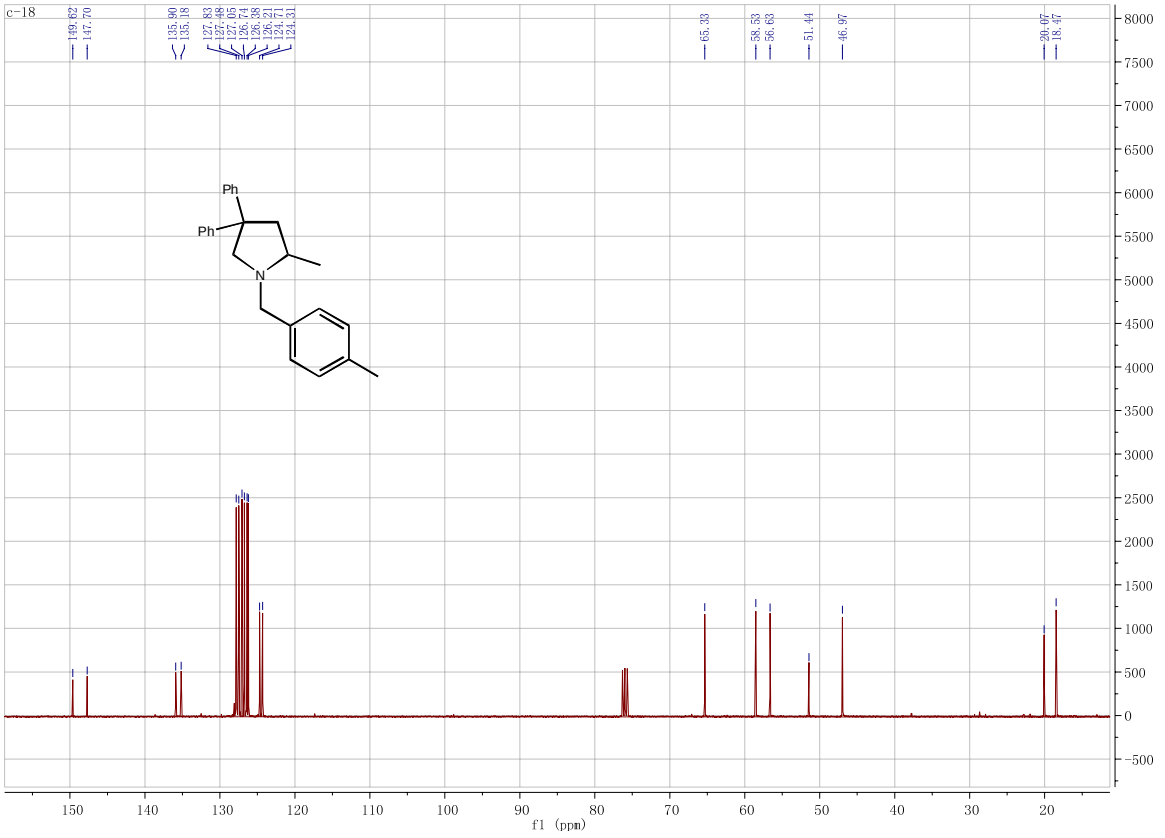

zl-s
1.00
1.06
1.02
4.29
2.12
2.20
2.27
2.21
4.16
f1 (ppm)

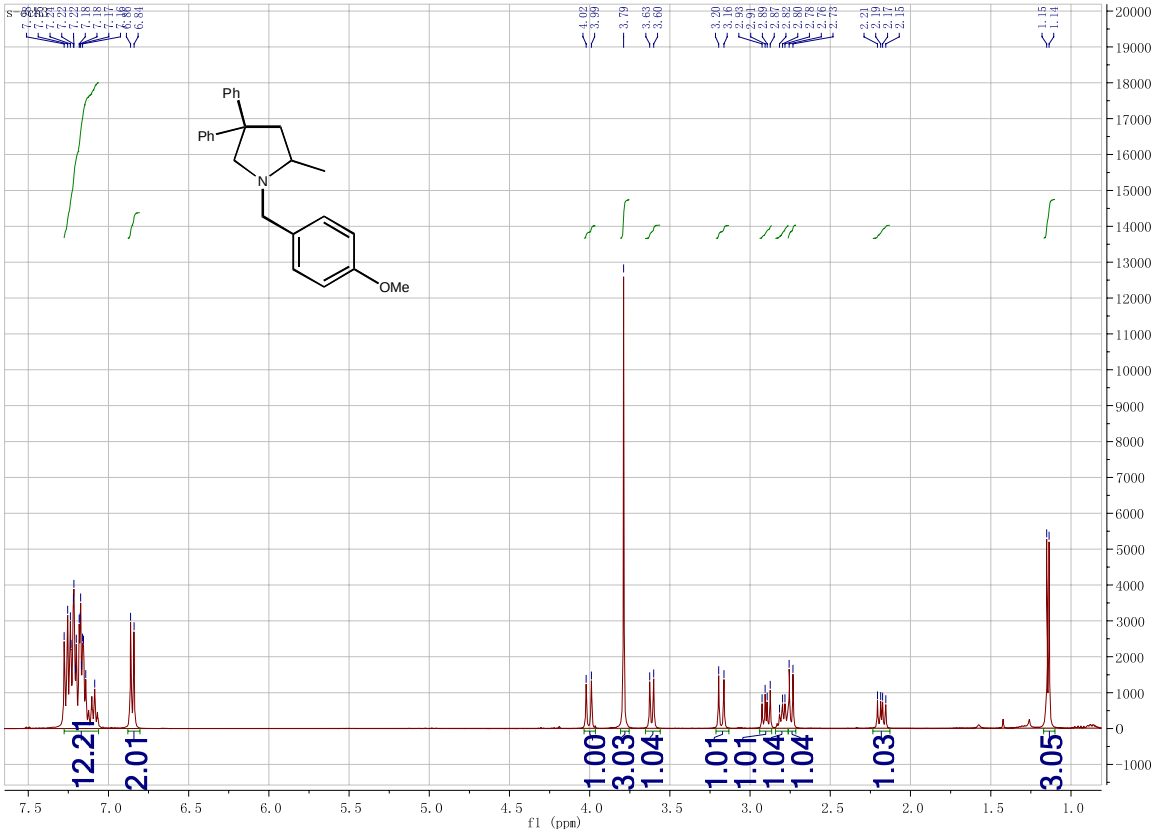

zl-s
137.52
113.51
48.81
48.59
31.42
30.65
28.42
19.54
13.01
f1 (ppm)

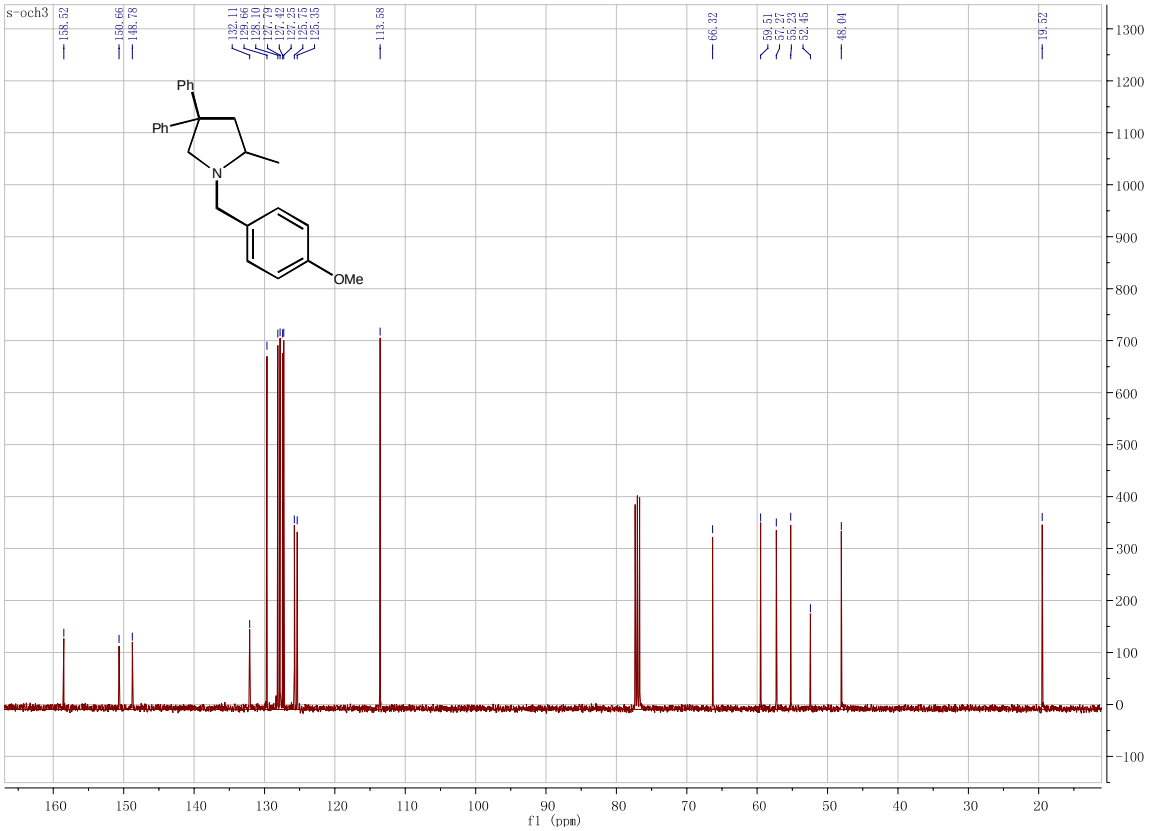

0c-1
Ph
Ph
N
15.39
1.00
0.97
0.98
1.01
2.05
1.01
2.96
f1 (ppm)

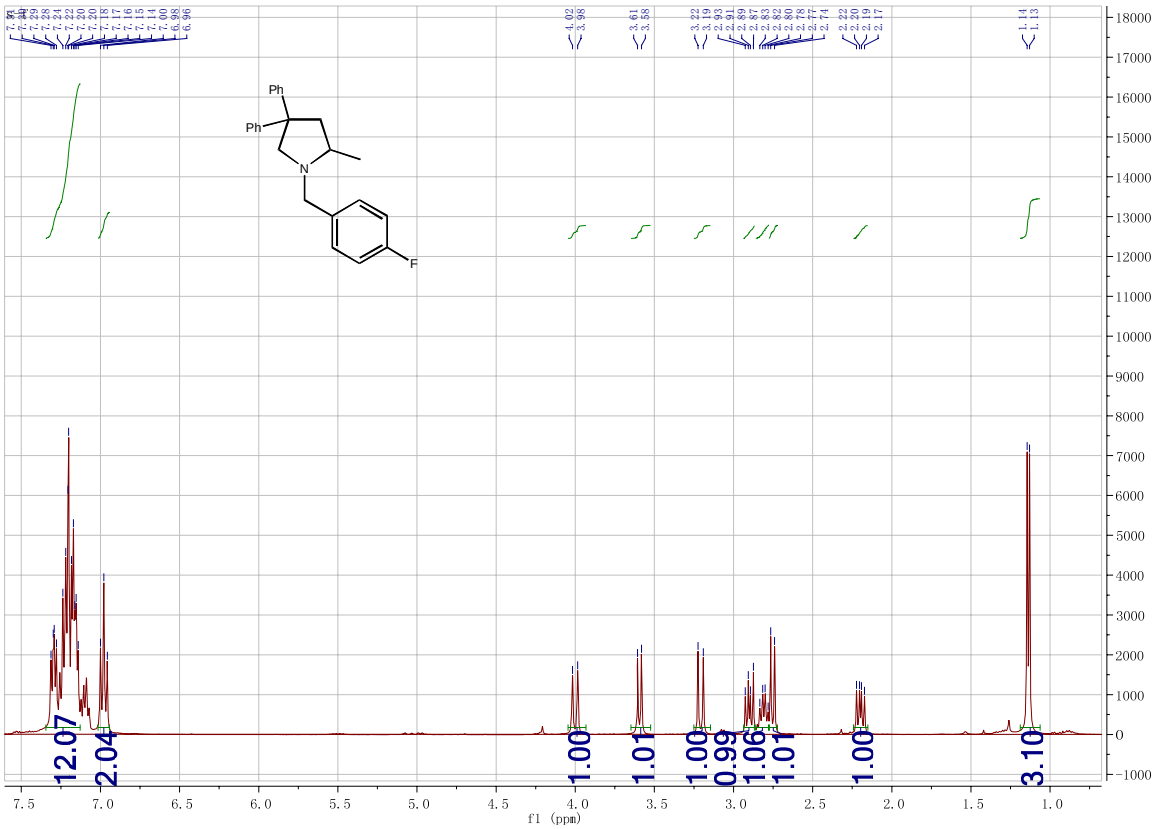

0c-1
Ph
Ph
N
f1 (ppm)

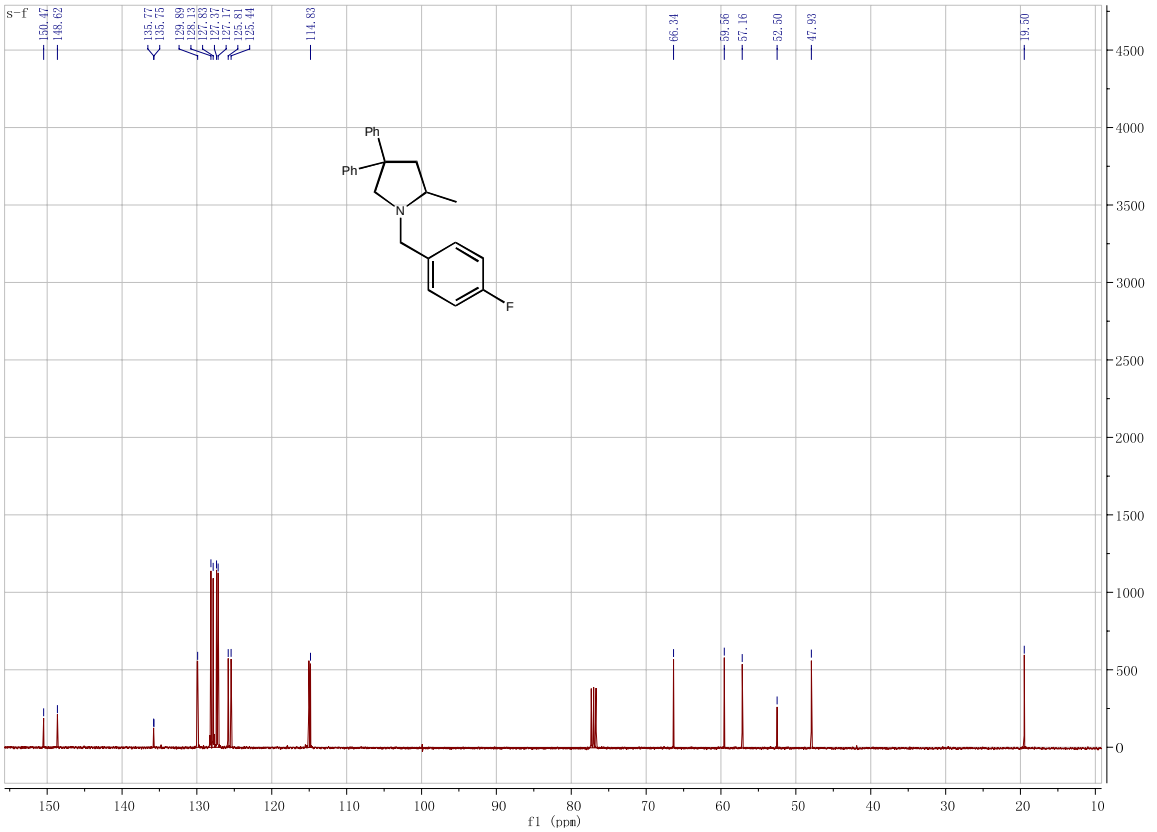

Ph
Ph
N
14.23
1.00
0.99
1.00
1.07
2.04
3.09
1.08
2.97
f1 (ppm)

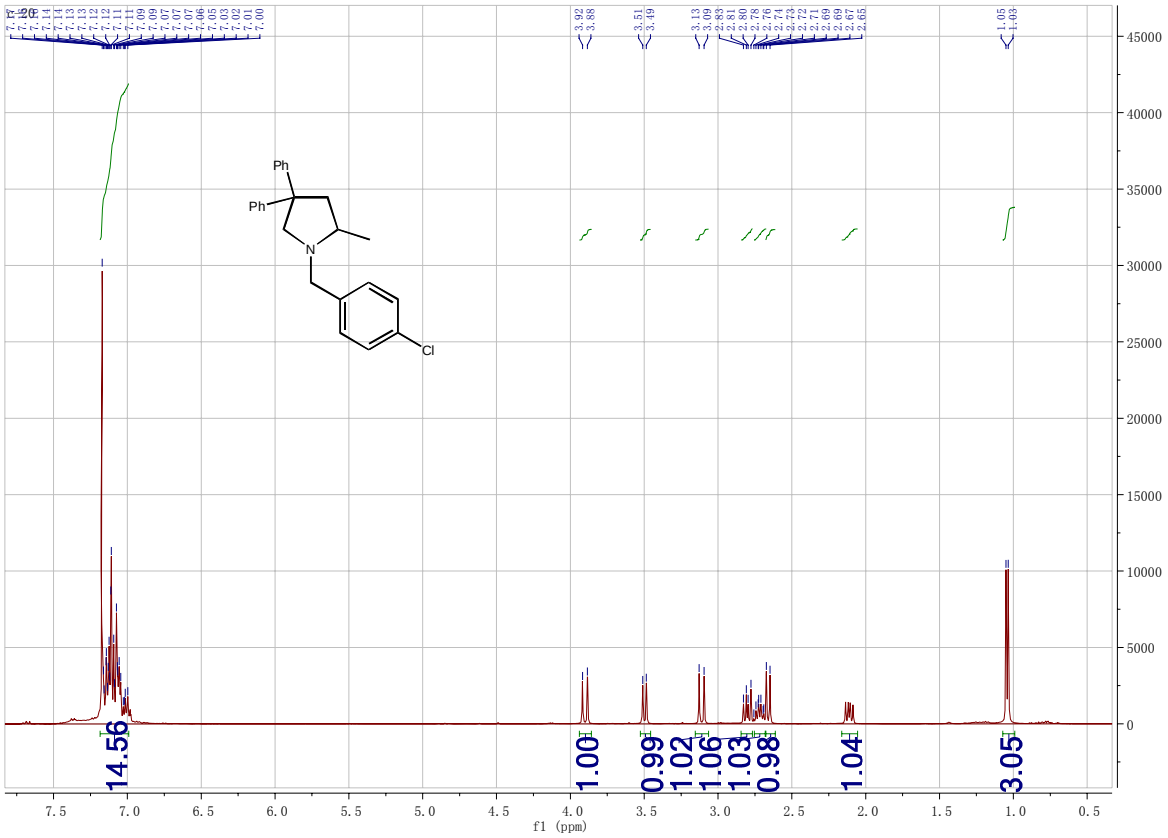

c-18
Ph
Ph
N
f1 (ppm)

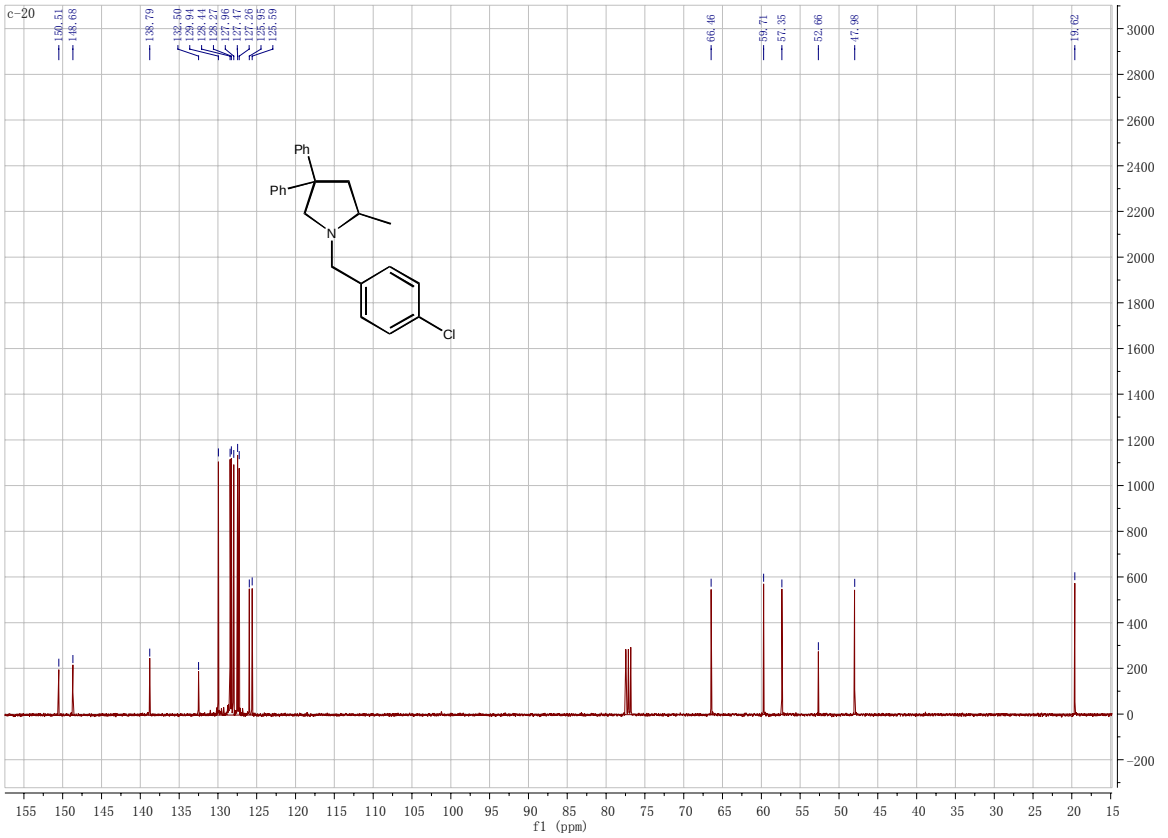

Ph
Ph
N
OMe
12.21
2.01
1.00
3.03
1.04
1.01
1.01
1.04
1.04
1.03
3.05
f1 (ppm)

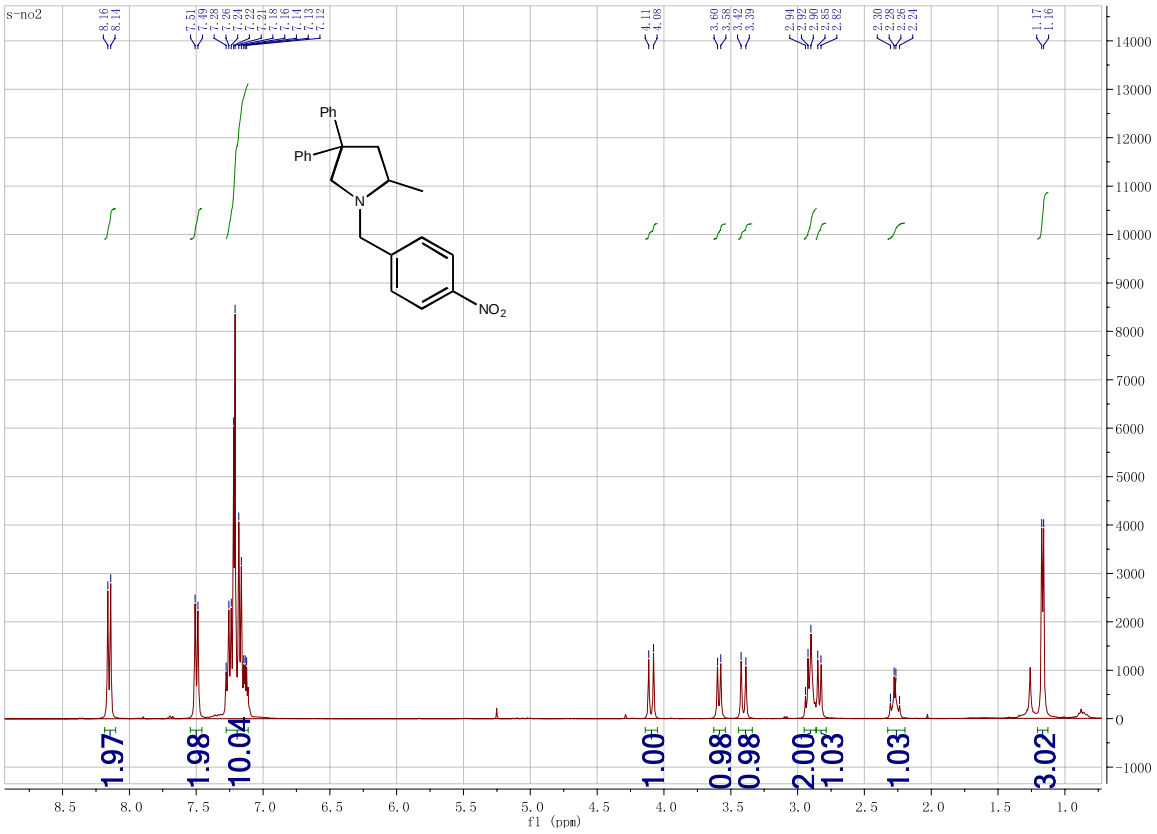

s-och3
Ph
Ph
N
OMe
f1 (ppm)

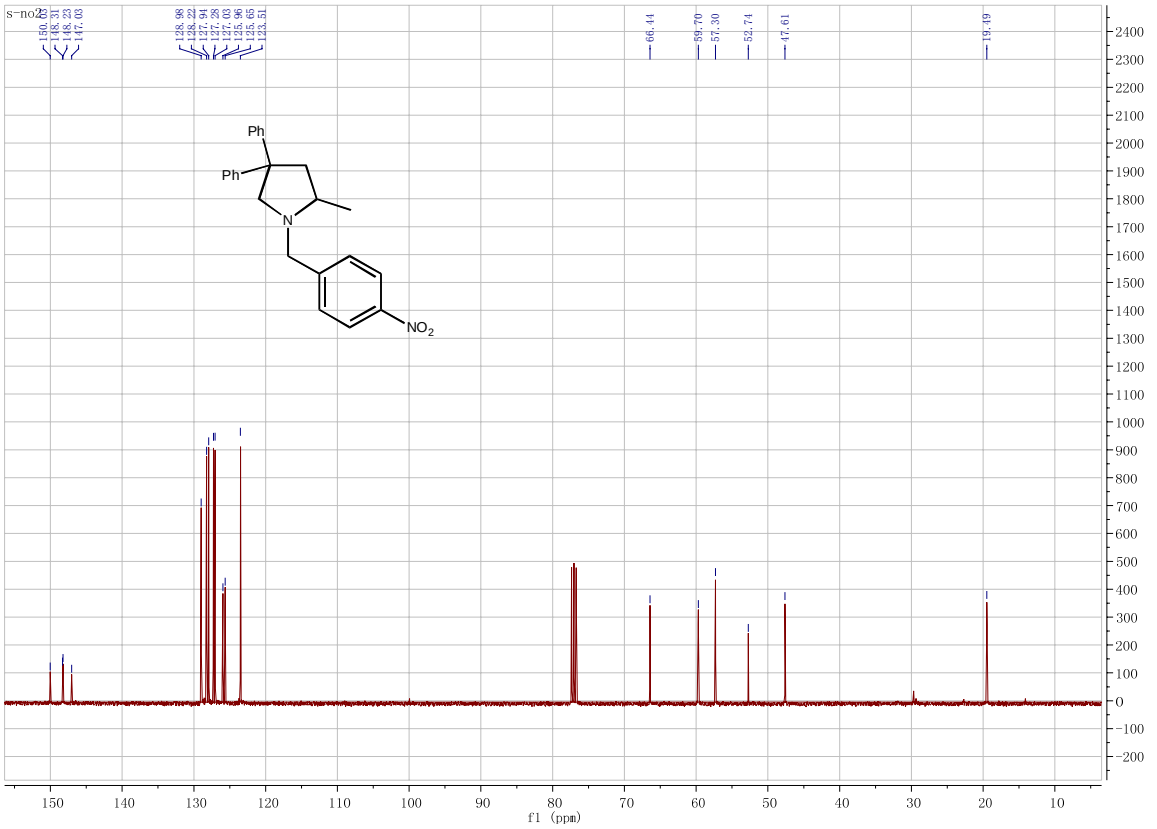

Ph
Ph
N
F
12.07
2.04
1.00
1.01
1.00
0.99
1.06
1.01
1.00
3.10
f1 (ppm)

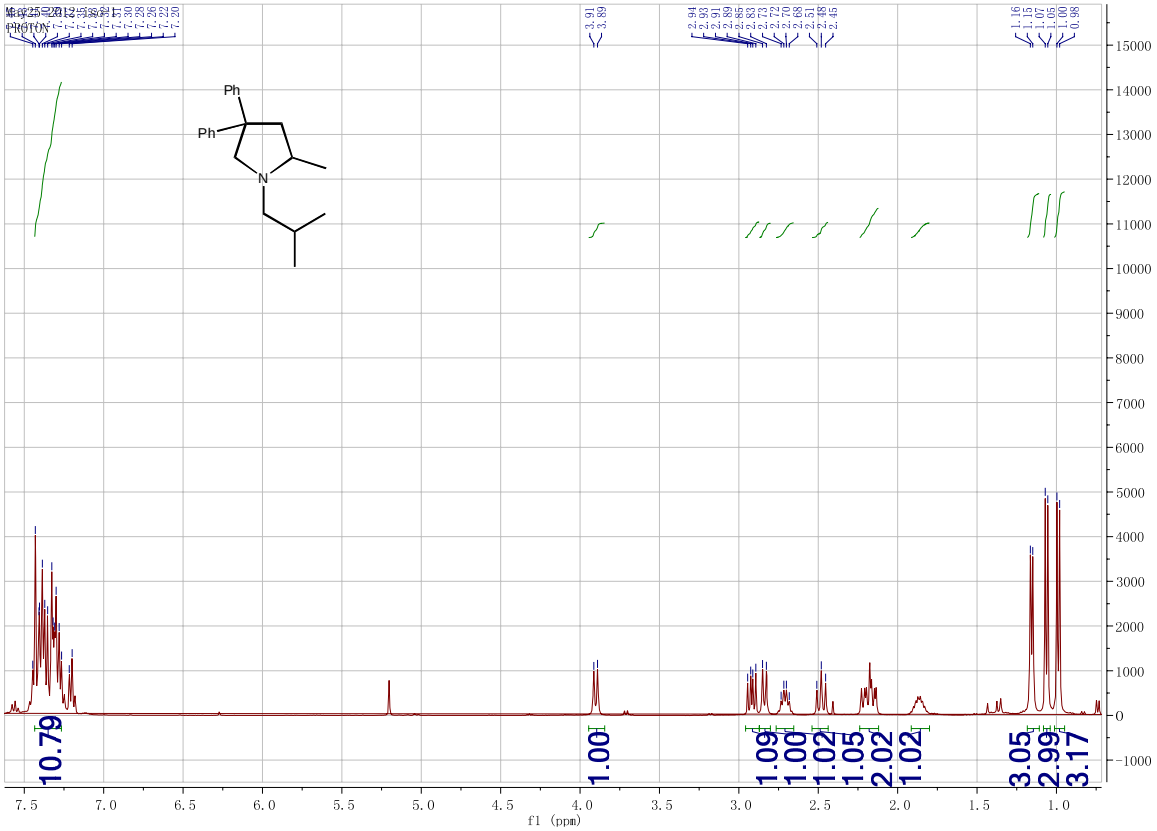

s-f
150.47
148.62
135.77
135.75
129.89
128.13
127.83
127.37
127.17
125.81
125.44
114.83
66.34
59.56
57.16
52.50
47.93
19.50
Ph
Ph
N
F
f1 (ppm)

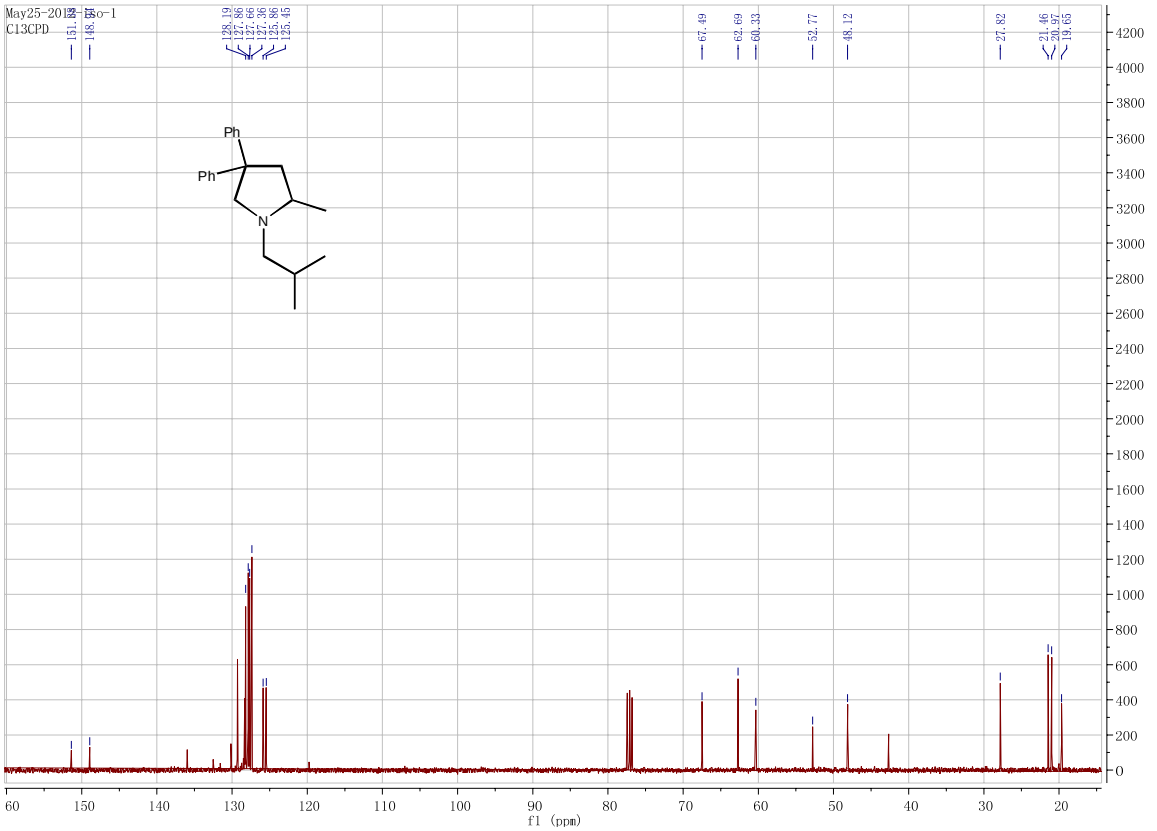

c-20
Ph
Ph
N
Cl
14.56
1.00
0.99
1.02
1.06
1.03
0.98
1.04
3.05
f1 (ppm)

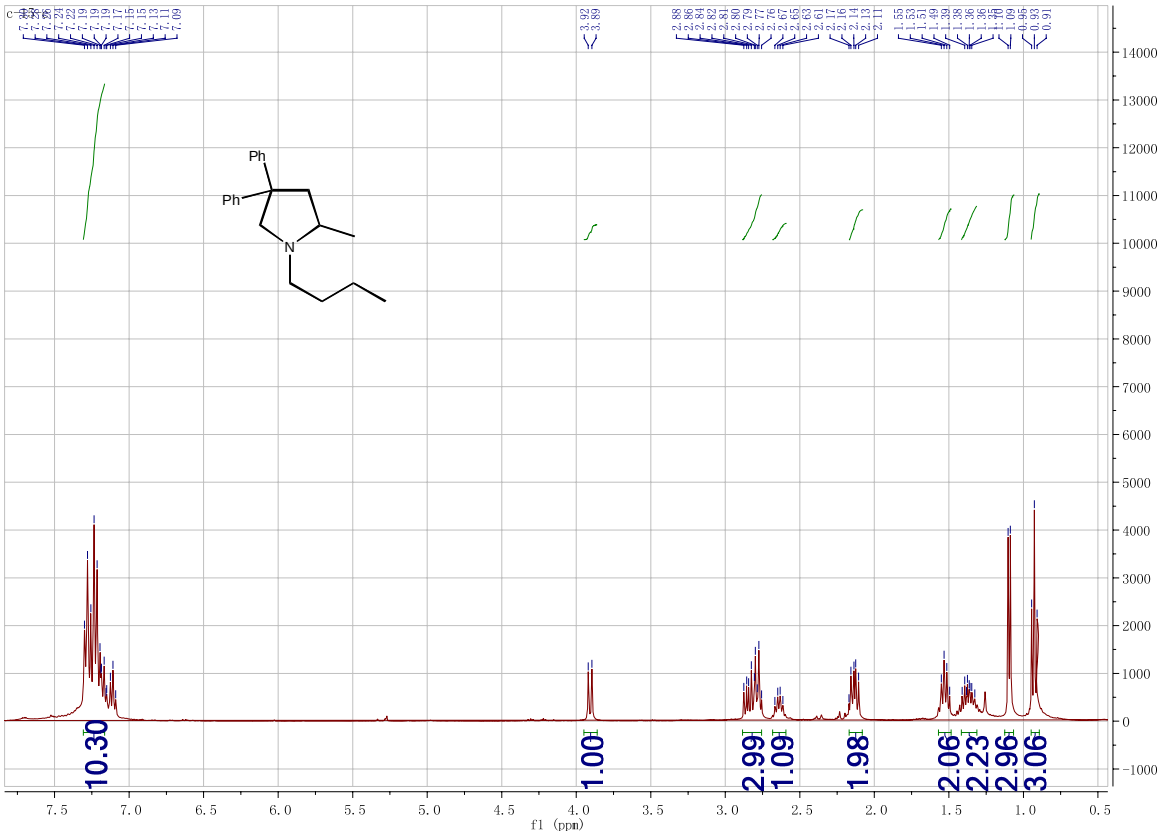

c-20
Ph
Ph
N
Cl
f1 (ppm)

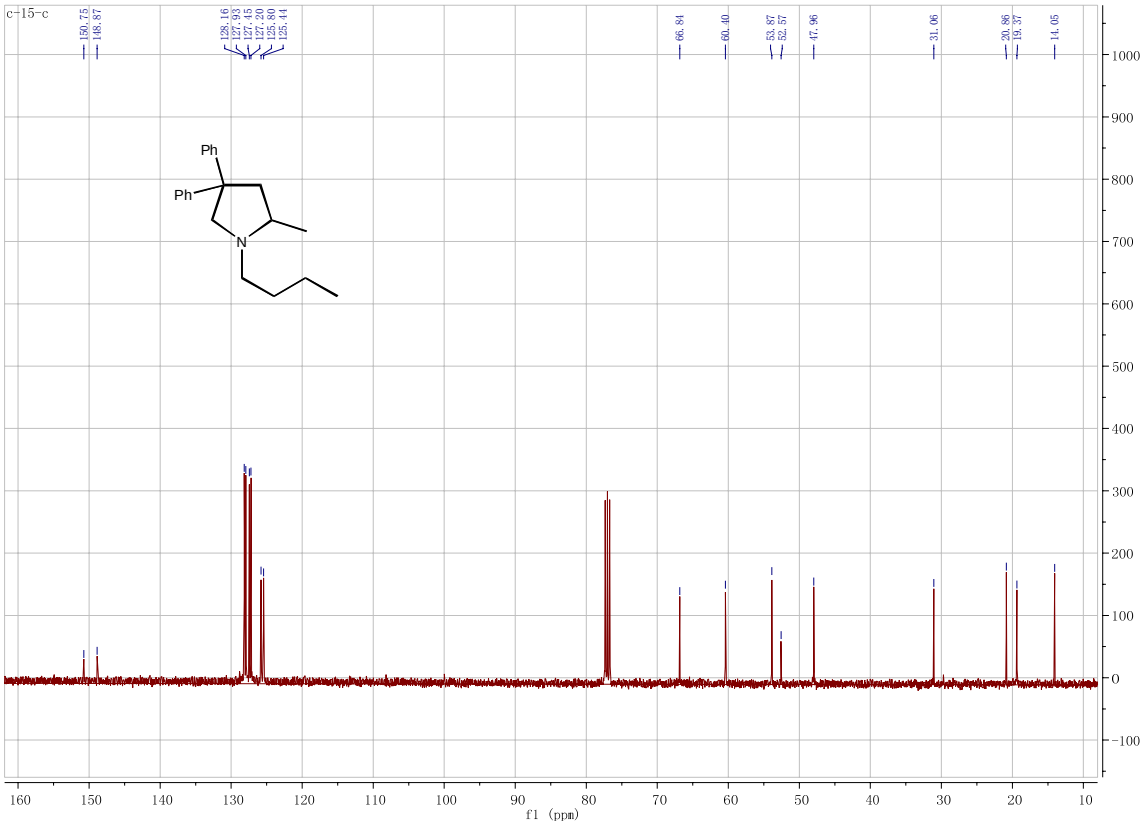

s-no2
8.16
8.14
7.51
7.49
7.28
7.25
7.23
7.22
7.21
7.18
7.16
7.14
7.13
7.12
4.11
4.08
3.60
3.58
3.42
3.39
2.94
2.92
2.90
2.85
2.82
2.30
2.28
2.26
2.24
1.17
1.16
Ph
Ph
N
NO2
1.97
1.98
10.04
1.00
0.98
0.98
2.00
1.03
1.03
3.02
f1 (ppm)

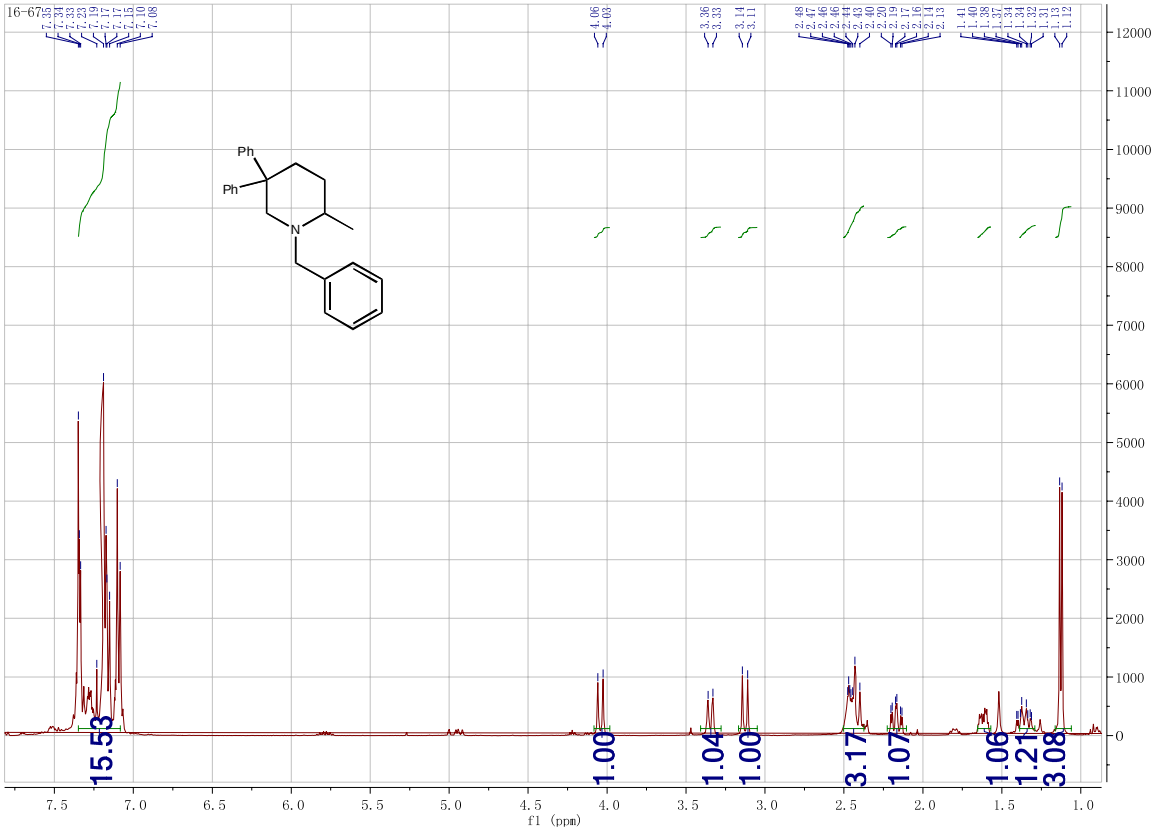

s-no2
150.07
148.31
148.23
147.03
128.98
128.22
127.94
127.28
127.03
125.96
125.65
123.51
66.44
59.70
57.30
52.74
47.61
19.49
Ph
Ph
N
NO2
f1 (ppm)

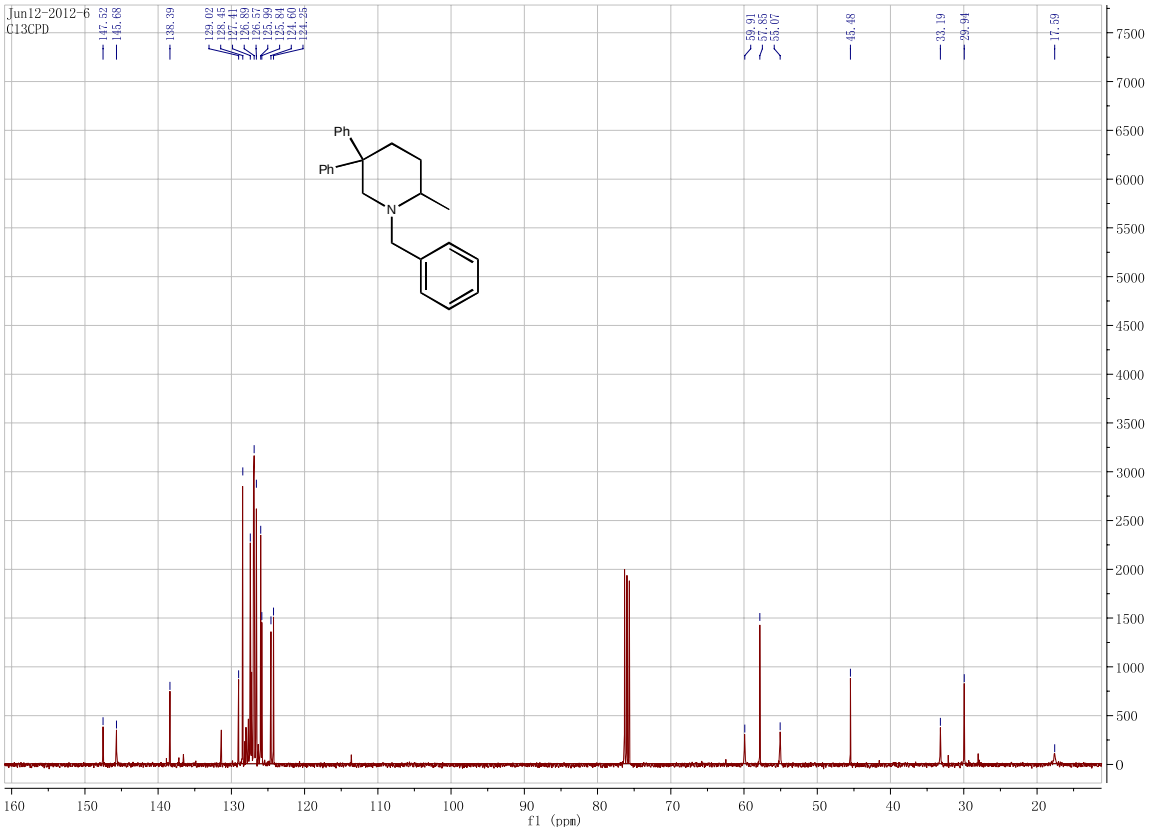

Ph
Ph
N
10.79
1.00
1.09
1.00
1.02
1.05
2.02
1.02
3.05
2.99
3.17
f1 (ppm)

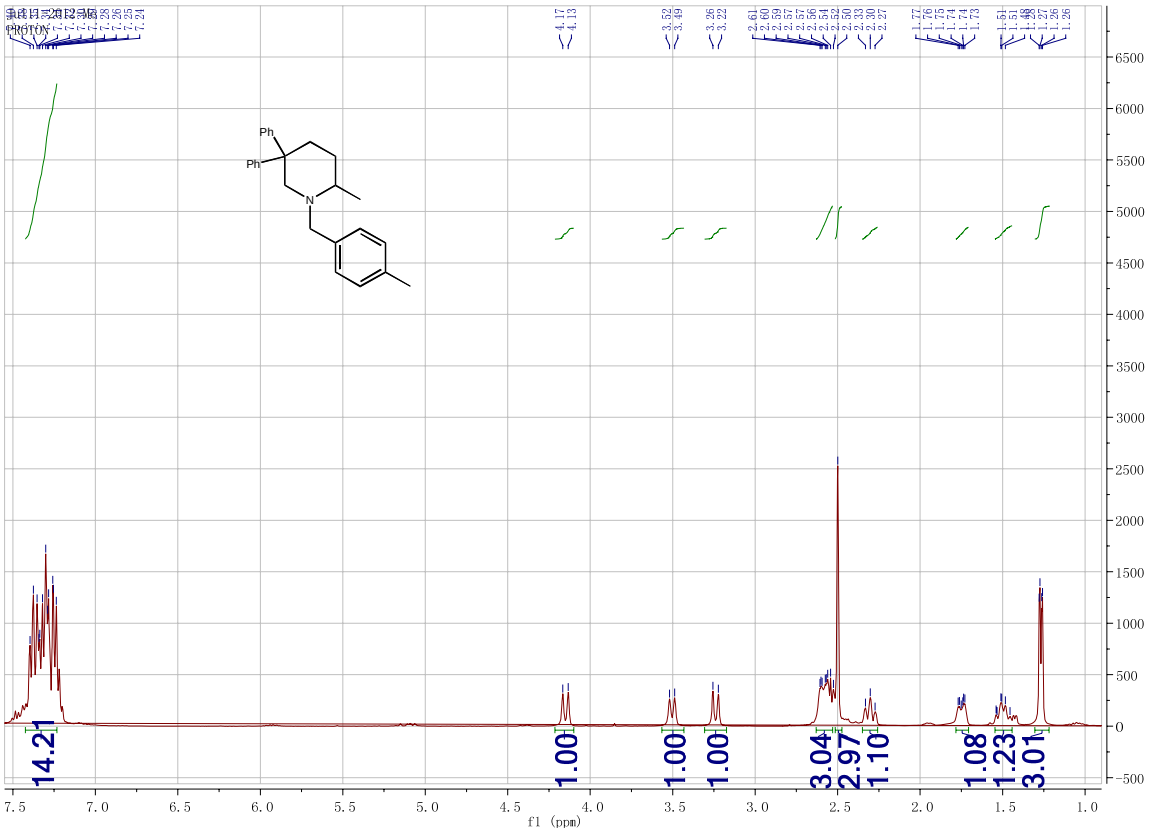

Ph
Ph
N
f1 (ppm)

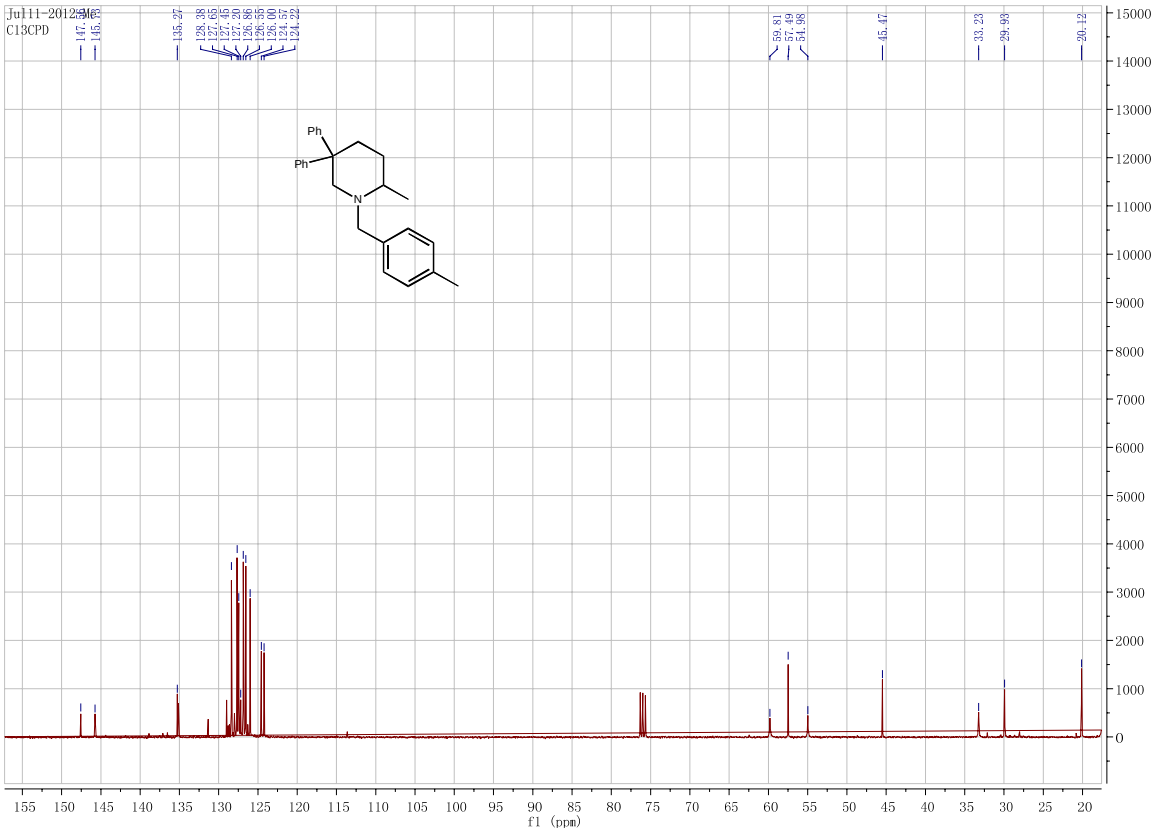

Ph
Ph
N
10.30
1.00
2.99
1.09
1.98
2.06
2.23
2.96
3.06
f1 (ppm)

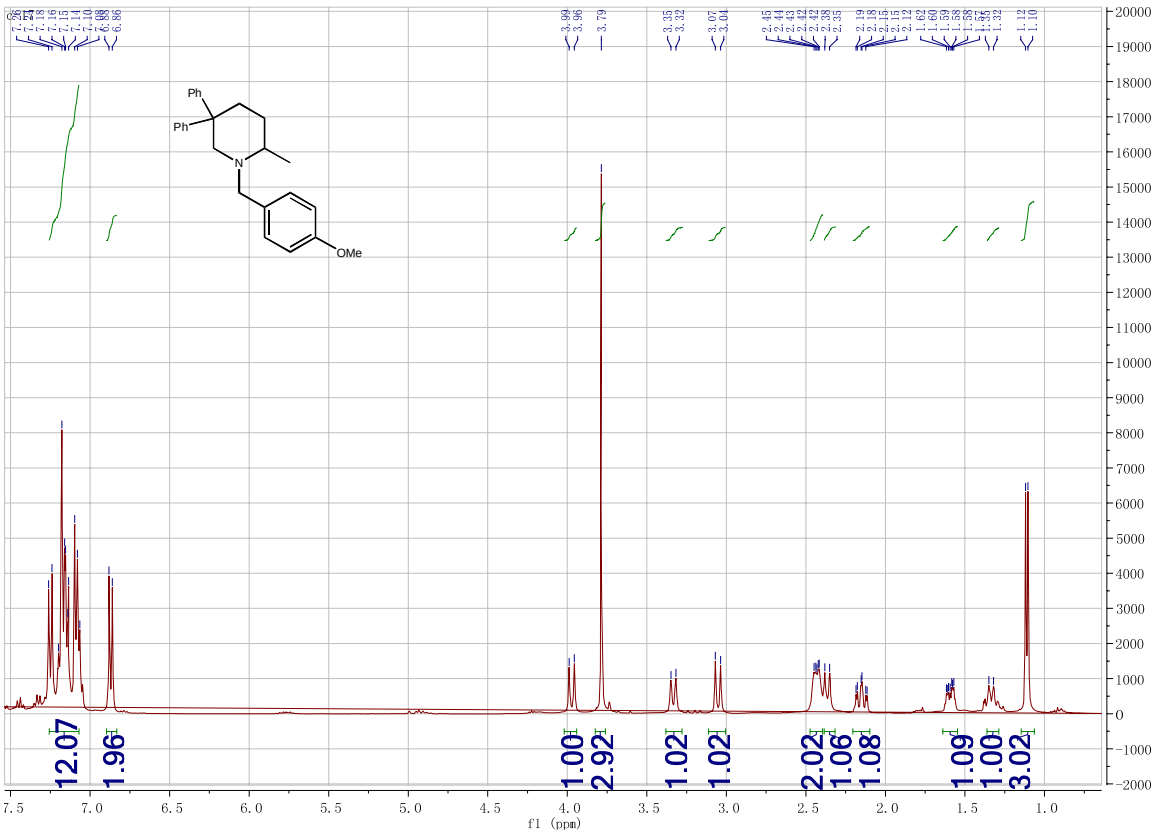

c-15-c
Ph
Ph
N
f1 (ppm)

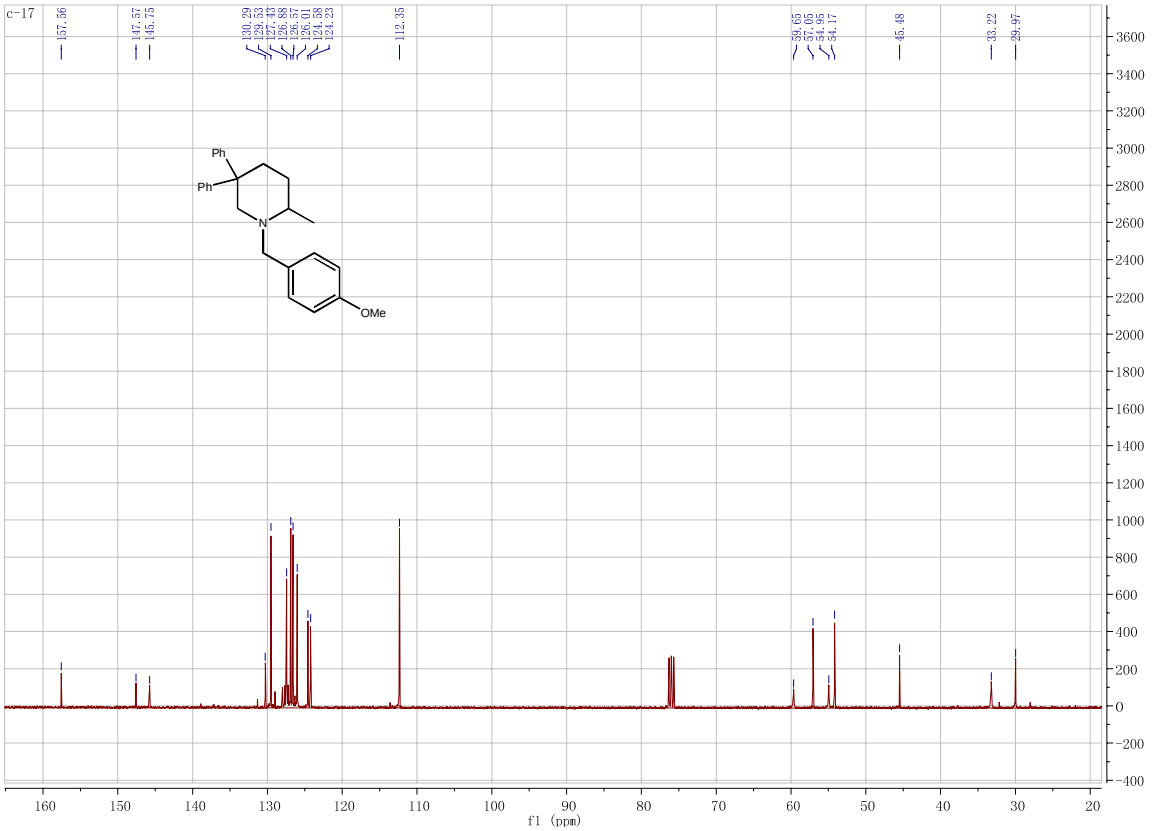

16-67
Ph
Ph
N
15.53
1.00
1.04
1.00
3.17
1.07
1.06
1.21
3.08
f1 (ppm)

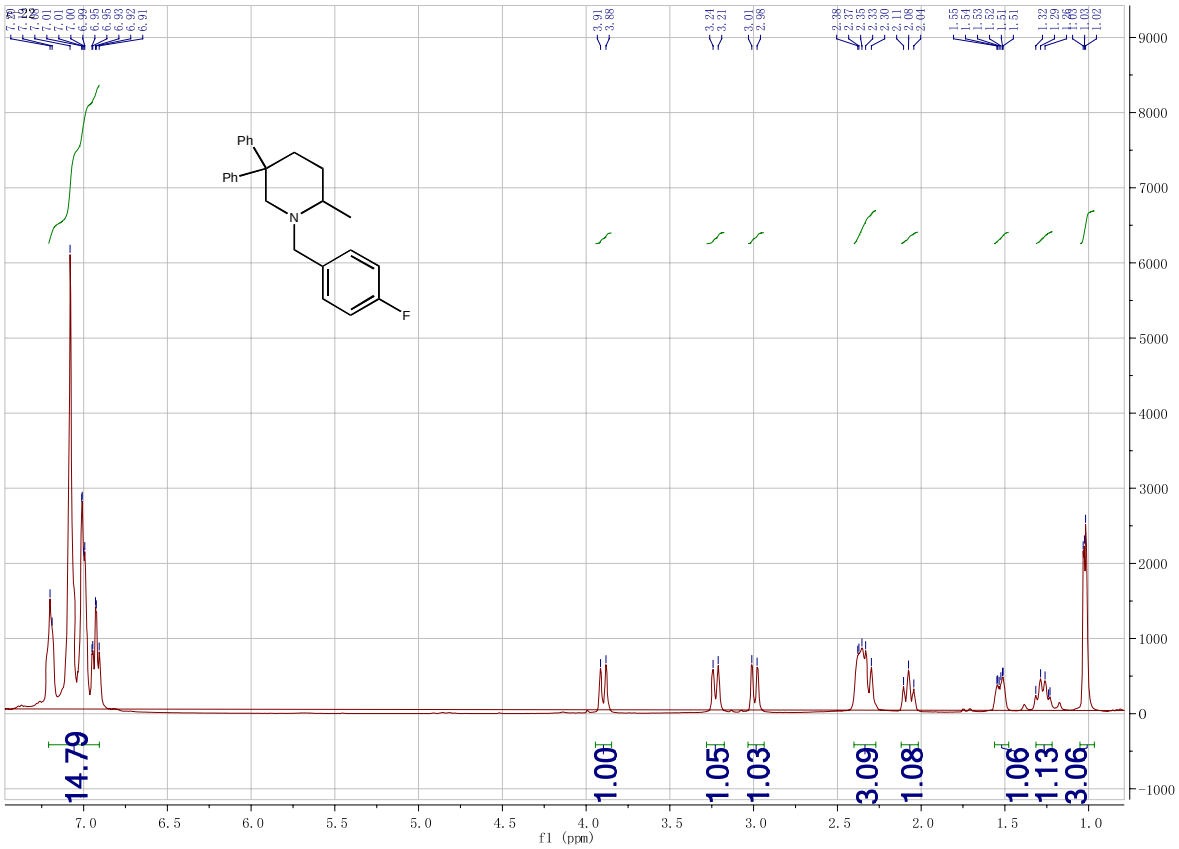

Jun12-2012-6
C13CPD
Ph
Ph
N
f1 (ppm)

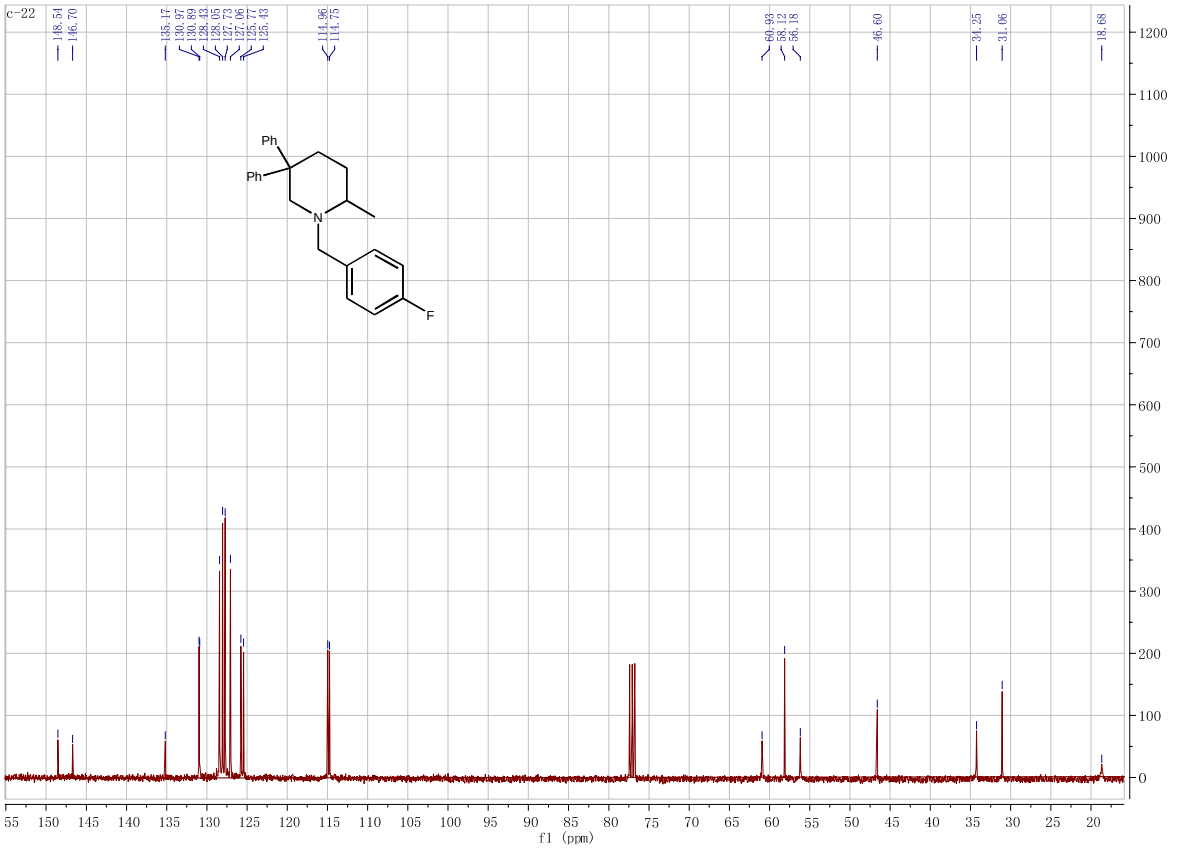

Ph
Ph
N
14.21
1.00
1.00
1.00
3.04
2.97
1.10
1.08
1.23
3.01
f1 (ppm)

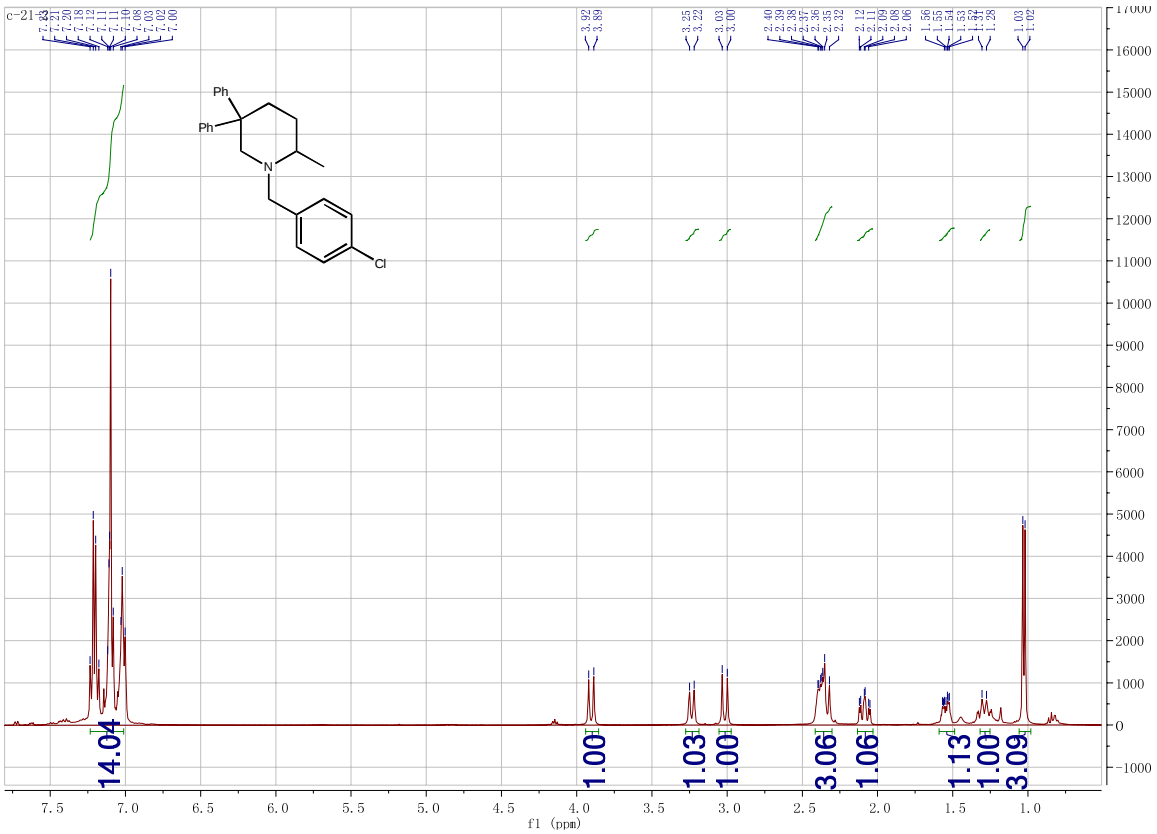

Ph
Ph
N
f1 (ppm)

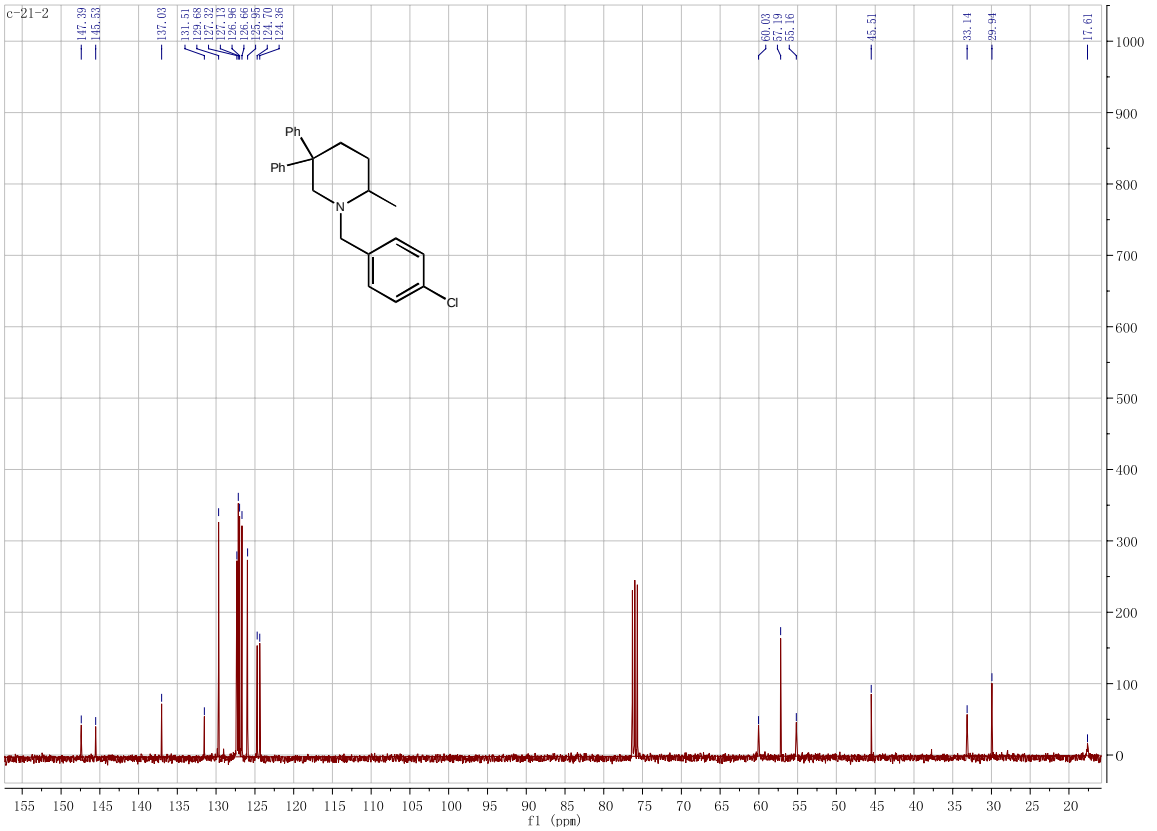

Ph
Ph
N
OMe
12.07
1.96
1.00
2.92
1.02
1.02
2.02
1.06
1.08
1.09
1.00
3.02
f1 (ppm)

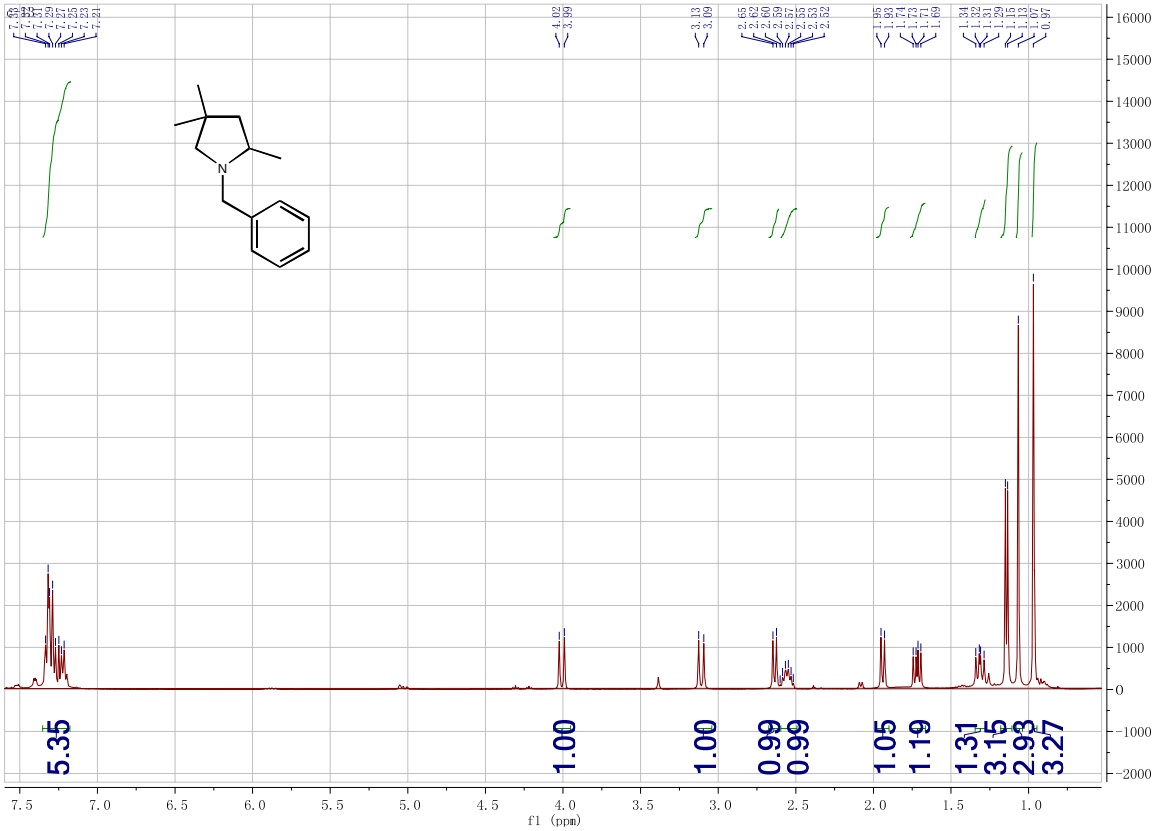

c-17
157.56
147.57
146.75
130.29
129.53
127.43
126.88
126.57
126.01
124.58
124.23
112.35
59.65
57.05
54.96
54.17
45.48
33.22
29.97
Ph
Ph
N
OMe
f1 (ppm)

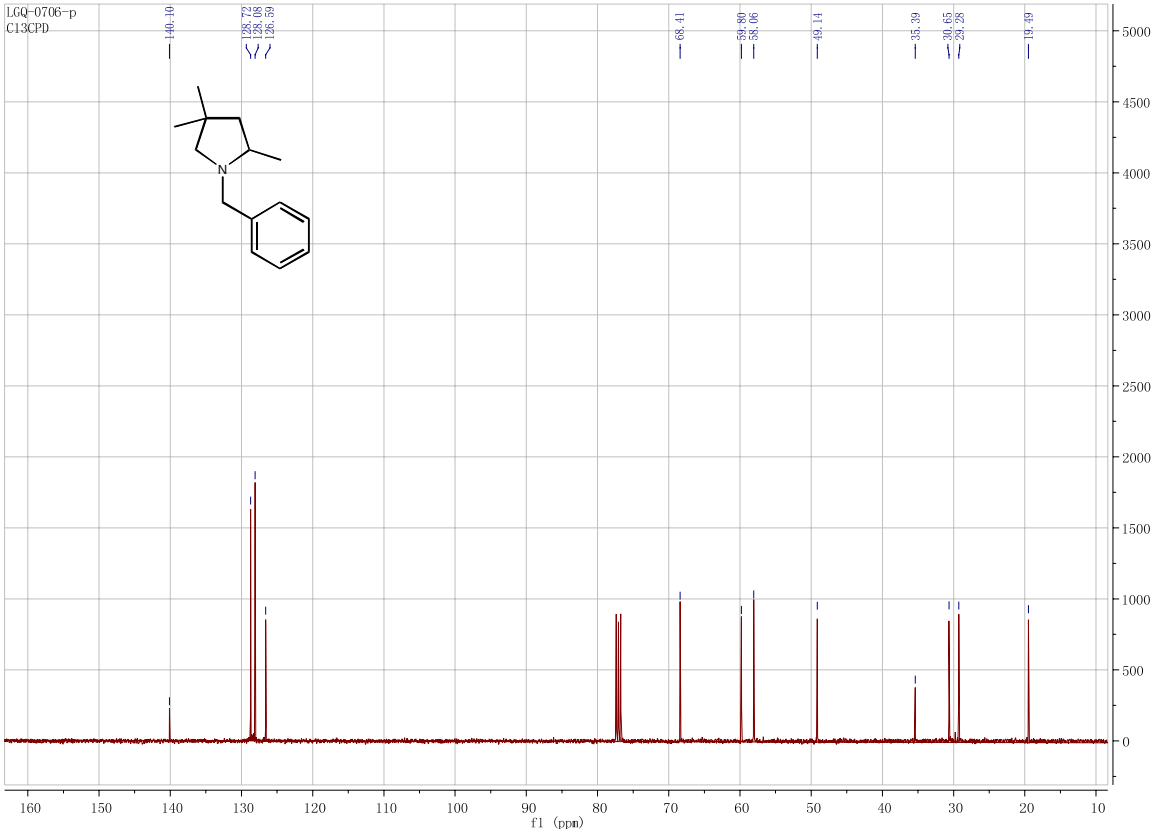

Ph
Ph
N
F
14.79
1.00
1.05
1.03
3.09
1.08
1.06
1.13
3.06
f1 (ppm)

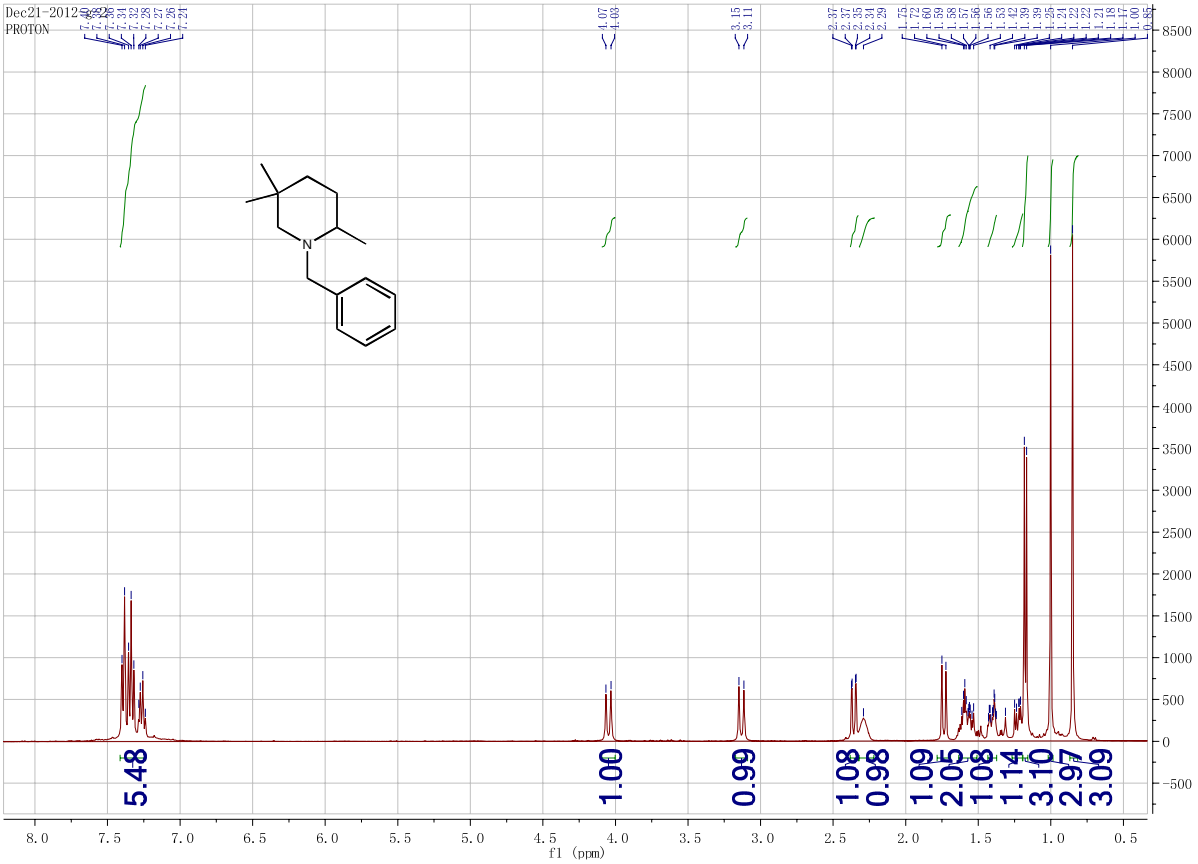

c-22
Ph
Ph
N
F
f1 (ppm)

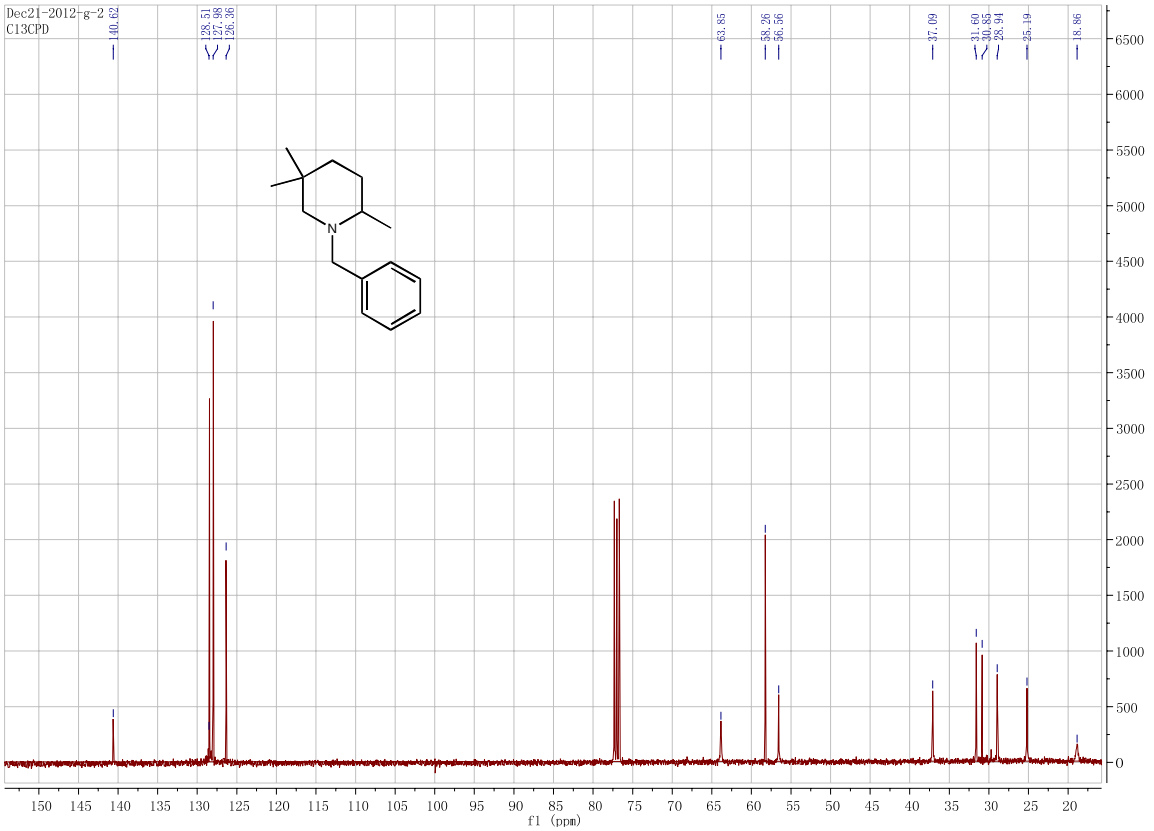

c-21-2
Ph
Ph
N
Cl
14.04
1.00
1.03
1.00
3.06
1.06
1.13
1.00
3.09
f1 (ppm)

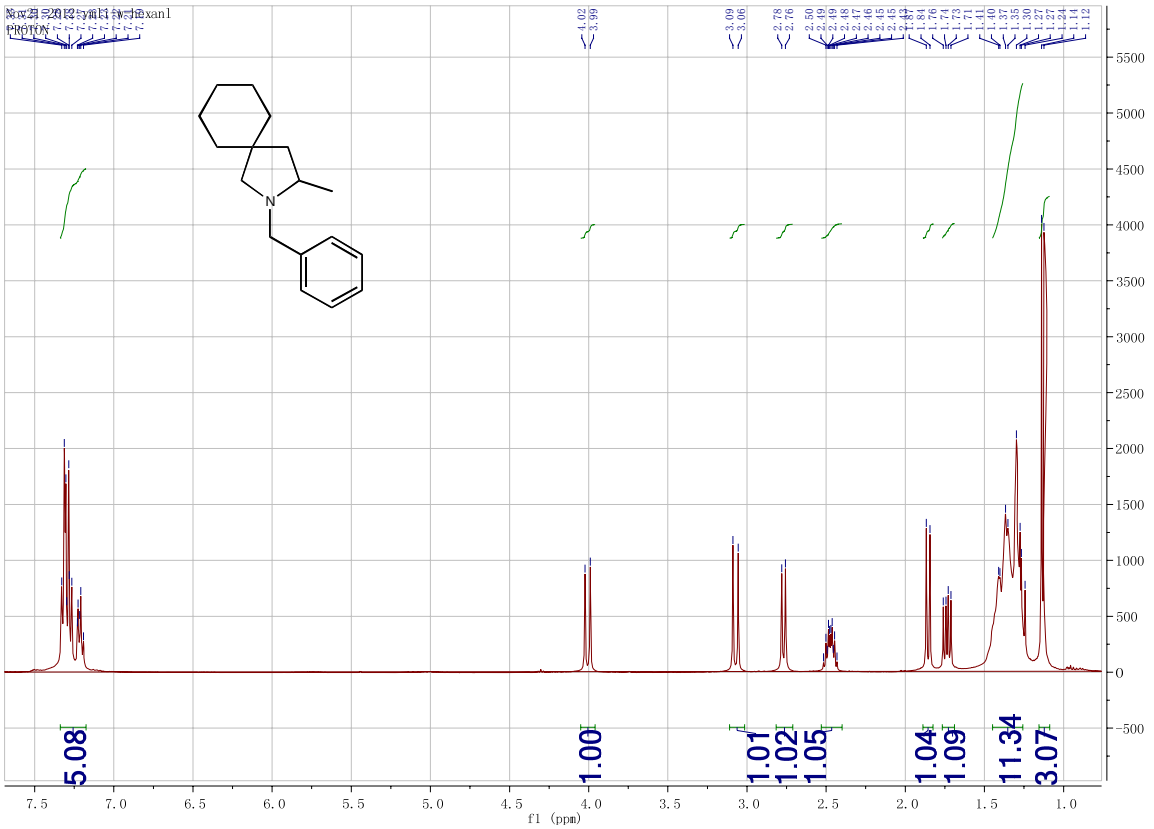

c-21-2
Ph
Ph
N
Cl
f1 (ppm)

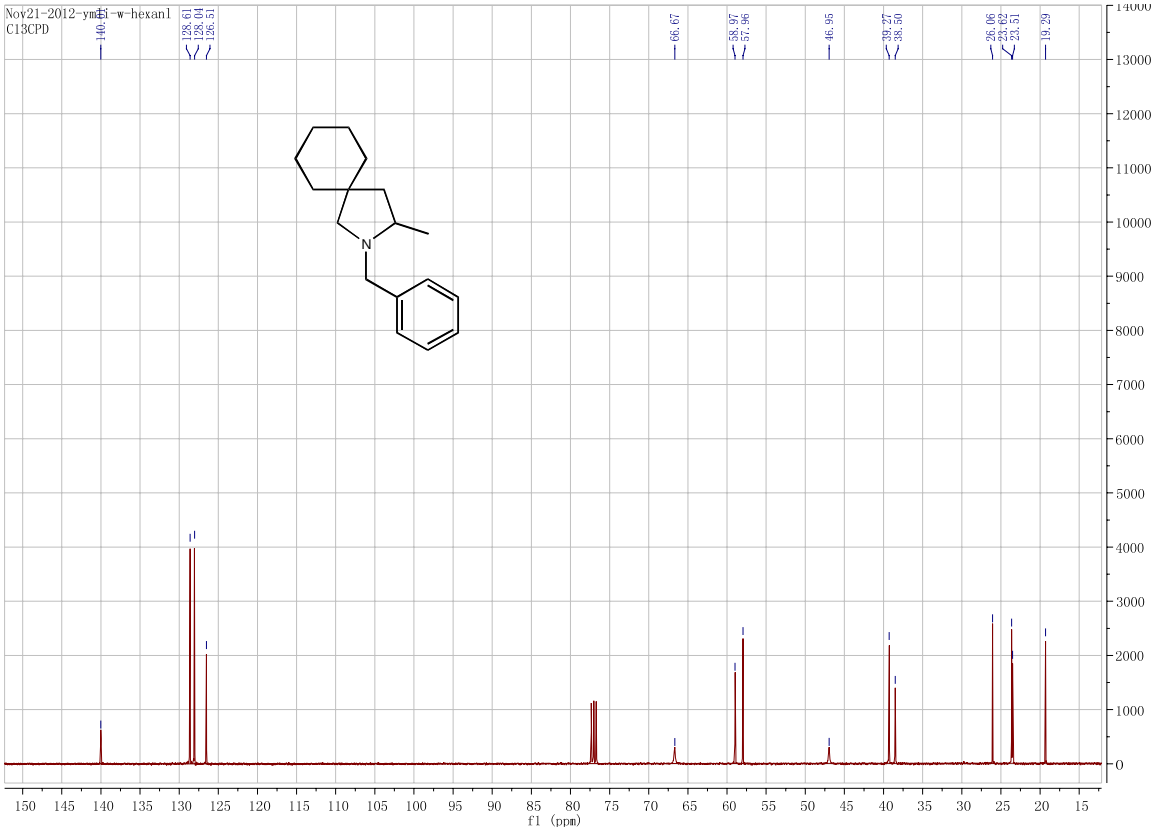

5.35
1.00
1.00
0.99
0.99
1.05
1.19
1.31
3.15
2.93
3.27
f1 (ppm)


LGQ-0706-p
C13CPD
f1 (ppm)


Dec21-2012-g-1
PROTON
5.48
1.00
0.99
1.08
0.98
1.09
2.05
1.08
1.14
3.10
2.97
3.09
f1 (ppm)


Dec21-2012-g-2
C13CPD
f1 (ppm)


PROTON
4.02
3.99
3.09
3.06
2.78
2.76
2.50
2.49
2.48
2.47
2.46
2.45
2.43
1.84
1.76
1.74
1.73
1.71
1.41
1.40
1.37
1.35
1.30
1.27
1.24
1.14
1.12
5.08
1.00
1.01
1.02
1.05
1.04
1.09
11.34
3.07
f1 (ppm)


C13CPD
140.11
128.61
128.04
126.51
66.67
58.97
57.96
46.95
39.27
38.50
26.06
23.62
23.51
19.29
f1 (ppm)