Supporting Information

Palladium-catalyzed three-component reaction of *N*-tosyl hydrazones, isonitriles and amines leading to amidines

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Content:

1. General considerations.

All reactions were performed in a dried sealed tube with magnetic stirring. All solvents were distilled prior to use. The boiling point of petroleum ether is between 60 and 90 °C. For chromatography, 300-400 mesh silica gel (Qingdao, China) was employed. ^{1}H and ^{13}C NMR spectra were recorded at 400 MHz and 100 MHz with Bruker ARX 400. Chemical shifts are reported in ppm using tetramethylsilane as internal standard. IR spectra were recorded with a Nicolet IS5 infrared spectrometer. HMRS were obtained on a 6540 UHD Accurate-Mass Q-TOF LC/MS.

2. The procedures for the synthesis of substrates.

The preparation of aldehyde hydrazones was followed by literature procedure.[1]

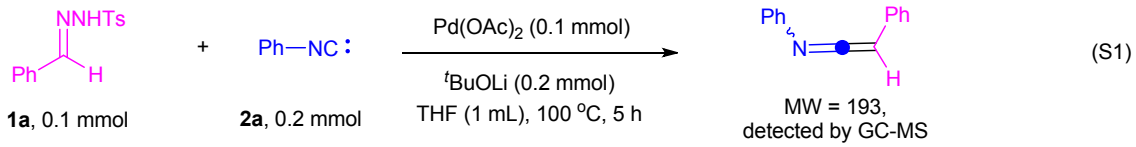
The preparation of aryl isonitriles was followed by literature procedure.[2]

3. Typical procedure for palladium-catalyzed three-component reactions.

$Pd(OAc)_2$ (2.2 mg, 0.01 mmol), PPh_3 (7.9 mg, 0.03 mmol), LiO^tBu (32.0 mg, 0.4 mmol), aldehyde hydrazone **1** (0.2 mmol), aryl isonitrile **2** (0.4 mmol), and amine **3** (0.2 mmol) was added to a sealed tube, THF (2 mL) was added *via* syringe. Then the tube was sealed and the mixture was stirred at 100 °C for 5 hours. After the reaction ended, solution was extracted by ethyl acetate and the combined extracts were removed in *vacuo* to leave a residue which was purified by flash silica gel chromatography to afford pure product **5-37**(5 ‰ Triethylamine was needed to add into the eluent).

4. Mechanism study.

To further explore the reaction possibilities of our system, we tested the reaction of *N*-tosylhydrazone (**1a**) with aryl isonitrile (**2a**) under the simplified reaction conditions (S1) to get ketenimine intermediate. Because ketenimine is unstable, we tried to use GC-MS to detect it. To our delight, a species with molecular weight 193, probably assigned to ketenimine, was detected by GC-MS (Figure 1).



(S1)

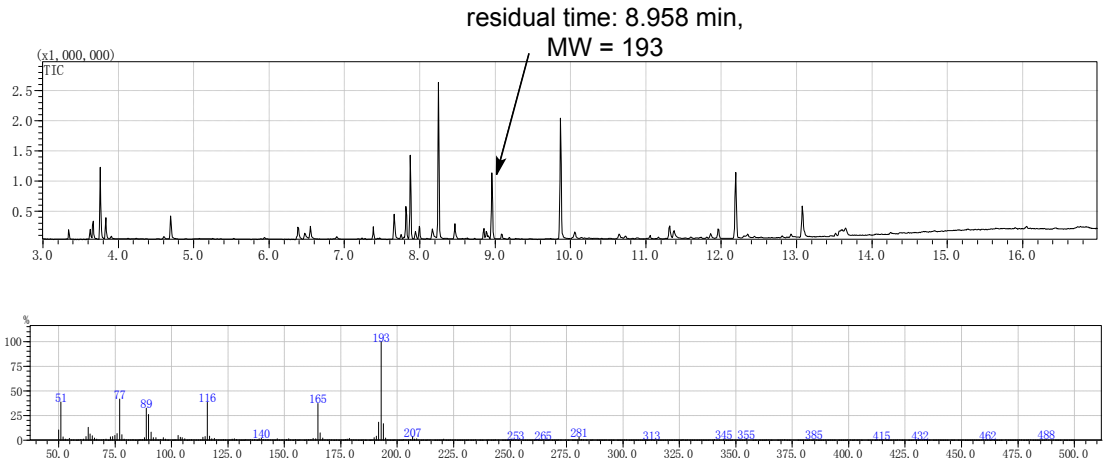
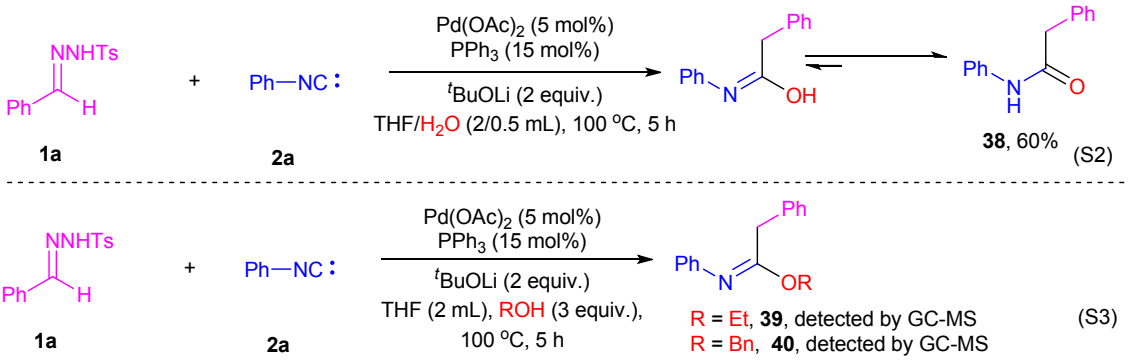


Figure 1. Detected ketenimine intermediate by GC-MS.

Furthermore, we also tried to use nucleophilic reagents such as water and alcohols to trap the ketenimine intermediate (S 2, 3). To our delight, the desired amides (**38**) was obtained in 60% yield, while the yields of corresponding products **39** and **40** were very low. We can only detect them by GC-MS (Figure 2, 3).



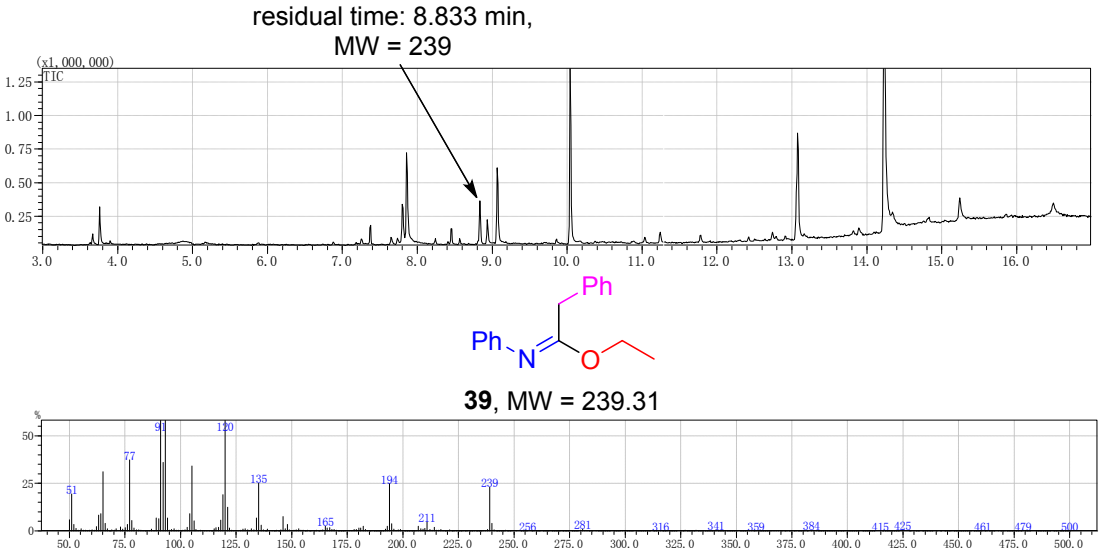


Figure 2. Captured the ketenimine intermediate by EtOH.

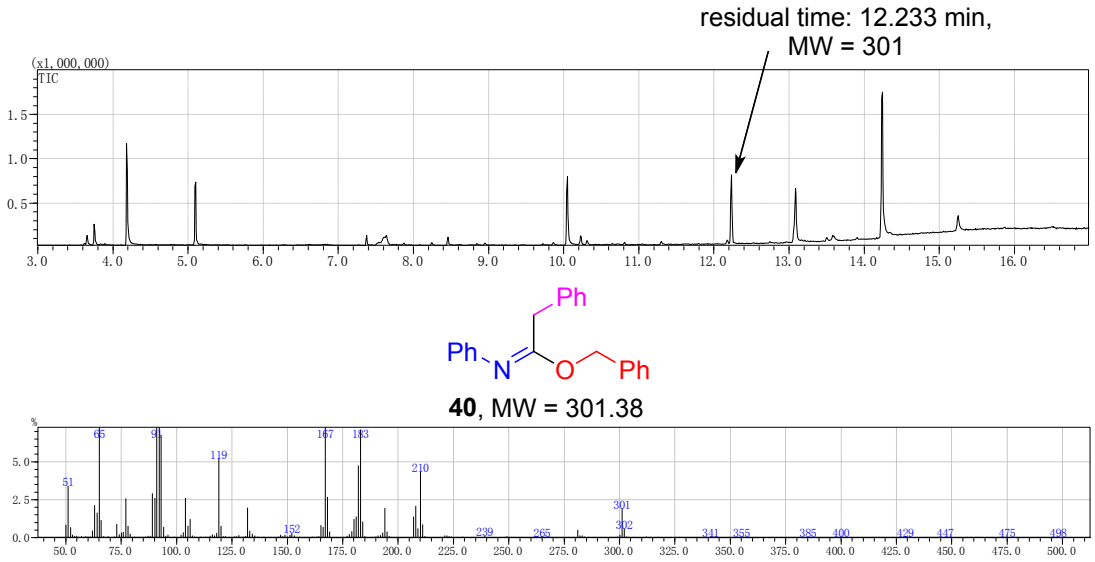


Figure 3. Captured the ketenimine intermediate by BnOH.

The results supported the possibility of our hypothesis that ketenimine intermediate did exist in our system. Thus, we consider the mechanism described in Scheme 3 is most plausible with the information currently available, although vigorous experiments are needed to unambiguously establish this mechanism.

5. X-ray structure of product 5

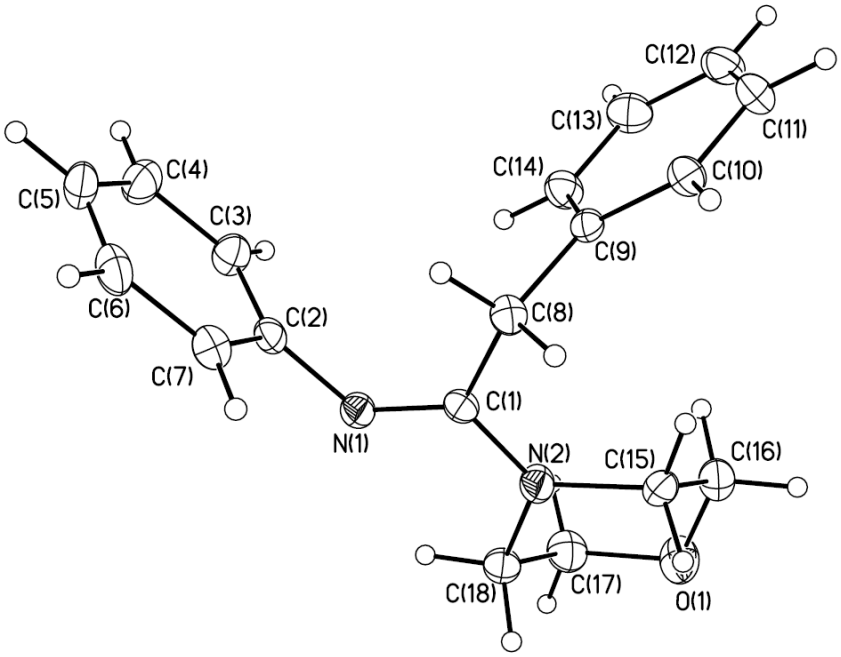
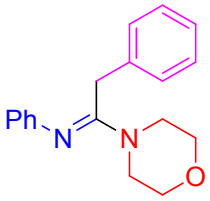


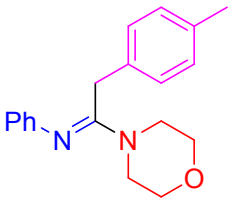
Figure 4. The configuration of C=N in **5** was confirmed to be *E* by single crystal test.

6. Spectra Data.



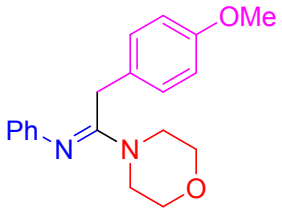
***(E)-N*-(1-morpholino-2-phenylethylidene)aniline (5)**

Yellow solid. m.p.: 99-100 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.6 Hz, 2 H), 7.24-7.16 (m, 5 H), 6.93 (t, *J* = 7.2 Hz, 1 H), 6.76 (d, *J* = 8.4 Hz, 2 H), 3.71 (s, 2 H), 3.59 (t, *J* = 4.4 Hz, 4 H), 3.46 (t, *J* = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.9, 151.0, 136.2, 128.8, 128.7, 127.7, 126.4, 121.9, 121.7, 66.6, 45.2, 33.6. IR 3056, 2964, 2896, 1611, 1591, 1428, 1295, 1108, 965 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{21}N_2O$ $[M+H]^+$ 281.1648, found 281.1650.



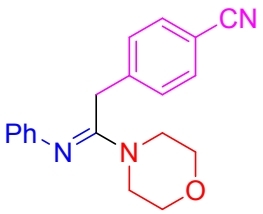
***(E)-N*-(1-morpholino-2-(4-tolyl)ethylidene)aniline (6)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.19 (t, *J* = 7.6 Hz, 2 H), 7.12 (d, *J* = 8.0 Hz, 2 H), 7.05 (d, *J* = 8.0 Hz, 2 H), 6.93 (t, *J* = 7.2 Hz, 1 H), 6.76 (d, *J* = 7.2 Hz, 2 H), 3.66 (s, 2 H), 3.60 (t, *J* = 4.8 Hz, 4 H), 3.46 (t, *J* = 4.8 Hz, 4 H), 2.33 (s, 3 H) . ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.1, 151.0, 136.0, 133.1, 129.4, 128.8, 127.6, 121.9, 121.7, 66.6, 45.2, 33.2, 20.9. IR 3051, 2960, 2852, 1615, 1592, 1445, 1236, 1118, 970 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O$ $[M+H]^+$ 295.1805, found 295.1804.



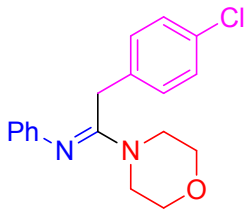
***(E)-N*-(2-(4-methoxyphenyl)-1-morpholinoethylidene)aniline (7)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.19 (t, *J* = 7.8 Hz, 2 H), 7.07 (d, *J* = 8.8 Hz, 2 H), 6.93 (t, *J* = 7.2 Hz, 1 H), 6.84 (d, *J* = 6.4 Hz, 2 H), 6.75 (d, *J* = 8.2 Hz, 2 H), 3.79 (s, 3 H), 3.63 (s, 2 H), 3.59 (t, *J* = 4.4 Hz, 4 H), 3.46 (t, *J* = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.1, 157.3, 151.0, 128.8, 128.7, 128.1, 121.9, 121.7, 114.1, 66.6, 55.2, 45.2, 32.7. IR 3056, 2959, 2852, 1621, 1510, 1454, 1244, 1118, 970 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O_2$ $[M+H]^+$ 311.1754, found 311.1753.



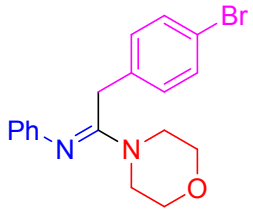
***(E)*-4-(2-morpholino-2-(phenylimino)ethyl)benzonitrile (8)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59 (d, J = 8.4 Hz, 2 H), 7.26 (d, J = 8.4 Hz, 2 H), 7.19 (t, J = 7.6 Hz, 2 H), 6.94 (t, J = 7.6 Hz, 1 H), 6.69 (d, J = 7.6 Hz, 2 H), 3.76 (s, 2 H), 3.60 (t, J = 4.4 Hz, 4 H), 3.41 (t, J = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.7, 150.5, 141.8, 132.5, 129.0, 128.5, 122.3, 121.4, 118.5, 110.6, 66.5, 45.3, 33.5. IR 3054, 2962, 2853, 2227, 1622, 1504, 1417, 1288, 1116, 998 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{20}N_3O$ $[M+H]^+$ 306.1601, found 306.1602.



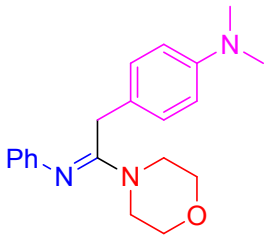
***(E)*-*N*-(2-(4-chlorophenyl)-1-morpholinoethylidene)aniline (9)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 (d, J = 8.4 Hz, 2 H), 7.20 (t, J = 7.8 Hz, 2 H), 7.09 (d, J = 8.8 Hz, 2 H), 6.94 (t, J = 7.2 Hz, 1 H), 6.72 (d, J = 7.2 Hz, 2 H), 3.66 (s, 2 H), 3.60 (t, J = 5.2 Hz, 4 H), 3.43 (t, J = 4.8 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.5, 150.8, 134.7, 132.3, 129.0, 128.9, 128.8, 122.1, 121.6, 66.6, 45.2, 32.8. IR 3055, 2962, 2853, 1667, 1614, 1490, 1443, 1236, 1117, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{20}ClN_2O$ $[M+H]^+$ 315.1259, found 315.1257.



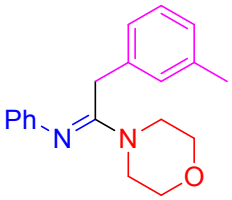
***(E)*-*N*-(2-(4-bromophenyl)-1-morpholinoethylidene)aniline (10)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.43 (d, J = 8.4 Hz, 2 H), 7.20 (t, J = 8.0 Hz, 2 H), 7.03 (d, J = 8.0 Hz, 2 H), 6.94 (t, J = 7.6 Hz, 1 H), 6.72 (d, J = 7.6 Hz, 2 H), 3.64 (s, 2 H), 3.60 (t, J = 4.8 Hz, 4 H), 3.43 (t, J = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.4, 150.8, 135.2, 131.8, 129.4, 128.9, 122.1, 121.5, 120.4, 66.6, 45.2, 32.9. IR 3056, 2961, 2852, 1610, 1487, 1430, 1236, 1117, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{20}BrN_2O$ $[M+H]^+$ 359.0754, found 359.0752.



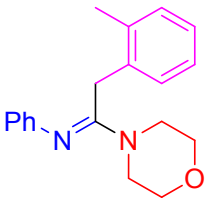
***(E)-N,N*-dimethyl-4-(2-morpholino-2-(phenylimino)ethyl)aniline (11)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.19 (t, *J* = 7.6 Hz, 2 H), 7.02 (d, *J* = 8.8 Hz, 2 H), 6.92 (t, *J* = 7.2 Hz, 1 H), 6.77 (d, *J* = 7.2 Hz, 2 H), 6.68 (d, *J* = 8.8 Hz, 2 H), 3.61-3.59 (m, 6 H), 3.47 (t, *J* = 4.4 Hz, 4 H), 2.93 (s, 6 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.6, 151.1, 149.2, 128.7, 128.4, 123.8, 121.8, 121.8, 112.8, 66.7, 45.2, 40.6, 32.6. IR 3055, 2958, 2852, 1614, 1519, 1446, 1236, 1118, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{20}H_{26}N_3O$ $[M+H]^+$ 324.2070, found 324.2068.



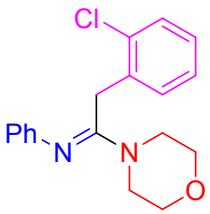
***(E)-N*-(1-morpholino-2-(3-tolyl)ethylidene)aniline (12)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.20 (t, *J* = 8.0 Hz, 3 H), 7.03 (d, *J* = 7.6 Hz, 1 H), 6.98-6.91 (m, 3 H), 6.76 (d, *J* = 7.2 Hz, 2 H), 3.68 (s, 2 H), 3.60 (t, *J* = 4.4 Hz, 4 H), 3.46 (t, *J* = 4.4 Hz, 4 H), 2.33 (s, 3 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.0, 151.0, 138.3, 136.1, 128.8, 128.5, 128.5, 127.2, 124.7, 121.9, 121.7, 66.7, 45.2, 33.5, 21.4. IR 3015, 2960, 2852, 1614, 1488, 1436, 1240, 1118, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O$ $[M+H]^+$ 295.1805, found 295.1806.



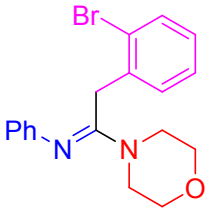
***(E)-N*-(1-morpholino-2-(2-tolyl)ethylidene)aniline (13)**

Yellow solid. m.p.: 136-138 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 (d, *J* = 7.2 Hz, 1 H), 7.22 (t, *J* = 7.6 Hz, 1 H), 7.19-7.11 (m, 4 H), 6.92 (t, *J* = 7.6 Hz, 1 H), 6.73 (d, *J* = 8.4 Hz, 2 H), 3.63 (t, *J* = 4.8 Hz, 4 H), 3.55 (s, 2 H), 3.44 (t, *J* = 4.8 Hz, 4 H), 2.13 (s, 3 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.3, 150.9, 135.3, 134.4, 130.1, 128.8, 127.2, 126.6, 126.2, 121.9, 121.6, 66.7, 45.0, 31.1, 19.6. IR 3056, 2970, 2854, 1612, 1589, 1428, 1287, 1107, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O$ $[M+H]^+$ 295.1805, found 295.1806.



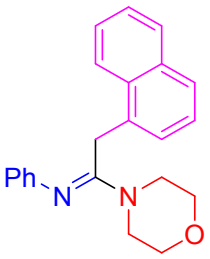
***(E)-N*-(2-(2-chlorophenyl)-1-morpholinoethylidene)aniline (14)**

Yellow solid. m.p.: 117-119 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 (d, J = 8.0 Hz, 2 H), 7.28 (t, J = 7.6 Hz, 1 H), 7.22-7.18 (m, 3 H), 6.94 (t, J = 7.6 Hz, 1 H), 6.75 (d, J = 8.4 Hz, 2 H), 3.75 (s, 2 H), 3.60 (t, J = 4.8 Hz, 4 H), 3.41 (t, J = 4.8 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.5, 150.7, 133.9, 133.3, 129.5, 128.9, 128.9, 128.0, 127.0, 122.1, 121.5, 66.6, 45.1, 31.1. IR 3052, 2971, 2854, 1615, 1588, 1426, 1258, 1115, 968 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{20}ClN_2O$ $[M+H]^+$ 315.1259, found 315.1260.



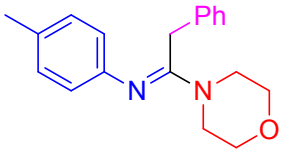
***(E)-N*-(2-(2-bromophenyl)-1-morpholinoethylidene)aniline (15)**

Yellow solid. m.p.: 97-98 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.52 (d, J = 8.0 Hz, 1 H), 7.33-7.30 (m, 2 H), 7.20 (t, J = 7.6 Hz, 2 H), 7.13-7.08 (m, 1 H), 6.94 (t, J = 7.2 Hz, 1 H), 6.75 (d, J = 8.0 Hz, 2 H), 3.74 (s, 2 H), 3.60 (t, J = 4.4 Hz, 4 H), 3.40 (t, J = 3.6 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.6, 150.7, 135.6, 132.8, 129.0, 128.9, 128.3, 127.7, 124.0, 122.1, 121.5, 66.6, 45.1, 33.9. IR 3069, 2917, 2846, 1617, 1590, 1420, 1255, 1119, 968 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{20}BrN_2O$ $[M+H]^+$ 359.0754, found 359.0753.



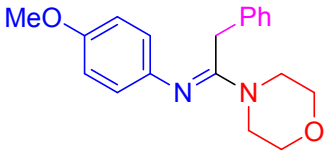
***(E)-N*-(1-morpholino-2-(naphthalen-1-yl)ethylidene)aniline (16)**

Yellow solid. m.p.: 127-129 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.89-7.87 (m, 1 H), 7.82-7.78 (m, 2 H), 7.52-7.48 (m, 4 H), 7.15 (t, J = 8.0 Hz, 2 H), 6.90 (t, J = 7.2 Hz, 1 H), 6.81 (d, J = 8.4 Hz, 2 H), 4.08 (s, 2 H), 3.63 (t, J = 4.4 Hz, 4 H), 3.48 (t, J = 4.0 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.0, 150.8, 133.7, 131.7, 131.0, 128.9, 128.8, 127.4, 126.3, 125.8, 125.5, 124.9, 122.6, 122.0, 121.4, 66.7, 45.1, 30.6. IR 3049, 2960, 2850, 1612, 1590, 1417, 1234, 1116, 968 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{22}H_{23}N_2O$ $[M+H]^+$ 331.1805, found 331.1806.



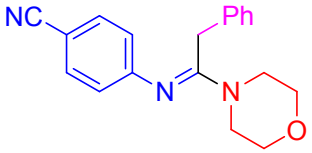
***(E)*-4-methyl-*N*-(1-morpholino-2-phenylethylidene)aniline (17)**[3]

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.6 Hz, 2 H), 7.22 (t, *J* = 7.6 Hz, 1 H), 7.17 (d, *J* = 7.2 Hz, 2 H), 7.01 (d, *J* = 8.0 Hz, 2 H), 6.67 (d, *J* = 8.0 Hz, 2 H), 3.72 (s, 2 H), 3.59 (t, *J* = 4.8 Hz, 4 H), 3.45 (t, *J* = 5.2 Hz, 4 H), 2.26 (s, 3 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.1, 148.3, 136.3, 131.1, 129.4, 128.6, 127.7, 126.4, 121.4, 66.6, 45.2, 33.5, 20.6.



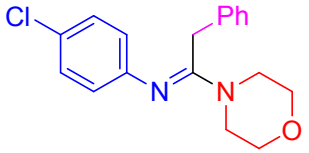
***(E)*-4-methoxy-*N*-(1-morpholino-2-phenylethylidene)aniline (18)**

Yellow solid. m.p.: 121-123 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.40 (t, *J* = 7.6 Hz, 2 H), 7.31 (t, *J* = 7.6 Hz, 1 H), 7.25 (d, *J* = 7.6 Hz, 2 H), 6.85 (d, *J* = 8.8 Hz, 2 H), 6.78 (d, *J* = 8.8 Hz, 2 H), 3.82 (s, 3 H), 3.81 (s, 2 H), 3.68 (t, *J* = 4.8 Hz, 4 H), 3.54 (t, *J* = 4.8 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.5, 154.9, 144.3, 136.3, 128.7, 127.7, 126.4, 122.4, 114.2, 66.6, 55.4, 45.2, 33.4. IR 3041, 2964, 2836, 1629, 1599, 1505, 1416, 1232, 1117, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O_2$ $[M+H]^+$ 311.1754, found 311.1756.



***(E)*-4-((1-morpholino-2-phenylethylidene)amino)benzonitrile (19)**

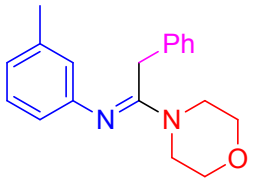
White solid. m.p.: 111-112 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44 (d, *J* = 8.4 Hz, 2 H), 7.31 (t, *J* = 7.6 Hz, 2 H), 7.23 (t, *J* = 7.2 Hz, 1 H), 7.12 (d, *J* = 7.6 Hz, 2 H), 6.77 (d, *J* = 8.4 Hz, 2 H), 3.67 (s, 2 H), 3.59 (t, *J* = 4.4 Hz, 4 H), 3.47 (t, *J* = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.5, 155.4, 135.3, 133.0, 128.9, 127.4, 126.8, 122.4, 119.6, 104.5, 66.4, 45.1, 33.7. IR 3065, 2966, 2856, 2219, 1611, 1587, 1453, 1241, 1120, 972 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{20}N_3O$ $[M+H]^+$ 306.1601, found 306.1600.



***(E)*-4-chloro-*N*-(1-morpholino-2-phenylethylidene)aniline (20)**

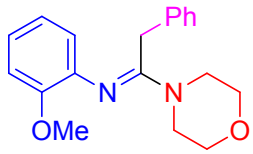
Yellow solid. m.p.: 124-125 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.6 Hz, 2 H), 7.22 (t, *J* = 7.2 Hz, 1 H), 7.14 (d, *J* = 6.8 Hz, 4 H), 6.68 (d, *J* = 8.4 Hz, 2 H), 3.69 (s, 2 H), 3.59 (t, *J* = 4.8 Hz, 4 H), 3.45 (t, *J* = 4.4 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.2, 149.6, 135.8, 128.8, 128.8, 127.6, 127.0, 126.6, 123.0, 66.6, 45.1,

33.5. IR 3030, 2959, 2850, 1618, 1588, 1448, 1234, 1119, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{20}ClN_2O$ $[M+H]^+$ 315.1259, found 315.1261.



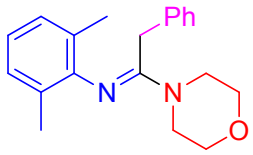
***(E)*-3-methyl-*N*-(1-morpholino-2-phenylethylidene)aniline (21)**

Yellow solid. m.p.: 95-96 °C. ^1H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.2 Hz, 2 H), 7.22 (t, *J* = 7.2 Hz, 1 H), 7.17 (d, *J* = 7.6 Hz, 2 H), 7.09 (t, *J* = 7.6 Hz, 1 H), 6.77 (d, *J* = 7.2 Hz, 1 H), 6.61 (s, 1 H), 6.57 (d, *J* = 8.0 Hz, 1 H), 3.73 (s, 2 H), 3.59 (t, *J* = 4.8 Hz, 4 H), 3.46 (t, *J* = 4.8 Hz, 4 H), 2.26 (s, 3 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.8, 150.9, 138.4, 136.3, 128.6, 128.6, 127.7, 126.4, 122.7, 122.5, 118.5, 66.6, 45.2, 33.6, 21.3. IR 3023, 2959, 2852, 1611, 1594, 1428, 1249, 1117, 908 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O$ $[M+H]^+$ 295.1805, found 295.1804.



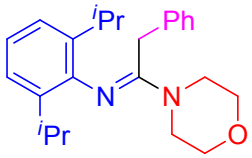
***(E)*-2-methoxy-*N*-(1-morpholino-2-phenylethylidene)aniline (22)**

Yellow solid. m.p.: 80-82 °C. ^1H NMR (400 MHz, $CDCl_3$) δ 7.28 (t, *J* = 6.8 Hz, 2 H), 7.19 (t, *J* = 7.6 Hz, 3 H), 6.93 (t, *J* = 7.6 Hz, 1 H), 6.85-6.81 (m, 2 H), 6.77 (d, *J* = 7.6 Hz, 1 H), 3.74 (s, 3 H), 3.62 (s, 2 H), 3.58 (t, *J* = 4.8 Hz, 4 H), 3.50 (t, *J* = 4.8 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.3, 150.8, 140.1, 136.3, 128.5, 127.9, 126.2, 122.9, 122.7, 120.8, 111.2, 66.6, 55.2, 45.3, 34.1. IR 3054, 2963, 2830, 1610, 1587, 1414, 1240, 1116, 969 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2O_2$ $[M+H]^+$ 311.1754, found 311.1752.



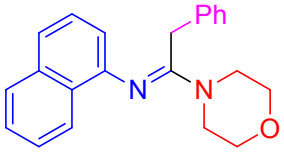
***(E)*-2,6-dimethyl-*N*-(1-morpholino-2-phenylethylidene)aniline (23)**

Yellow solid. m.p.: 63-65 °C. ^1H NMR (400 MHz, $CDCl_3$) δ 7.26 (t, *J* = 7.6 Hz, 2 H), 7.19 (t, *J* = 7.6 Hz, 1 H), 7.08 (d, *J* = 7.6 Hz, 2 H), 6.98 (d, *J* = 7.2 Hz, 2 H), 6.81 (t, *J* = 7.2 Hz, 1 H), 3.67 (t, *J* = 4.8 Hz, 4 H), 3.54 (t, *J* = 4.8 Hz, 4 H), 3.52 (s, 2 H), 2.06 (s, 6 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.0, 147.9, 135.9, 128.5, 128.3, 127.9, 127.8, 126.4, 121.7, 66.7, 45.7, 33.6, 18.5. IR 3064, 2963, 2857, 1605, 1588, 1399, 1247, 1113, 965 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{20}H_{25}N_2O$ $[M+H]^+$ 309.1961, found 309.1962.



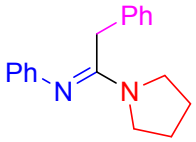
***(E)*-2,6-diisopropyl-*N*-(1-morpholino-2-phenylethylidene)aniline (24)**

Yellow solid. m.p.: 108-109 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.26 (t, *J* = 7.6 Hz, 2 H), 7.18 (t, *J* = 7.2 Hz, 1 H), 7.12 (d, *J* = 7.2 Hz, 2 H), 7.07 (d, *J* = 7.6 Hz, 2 H), 6.98 (t, *J* = 7.4 Hz, 1 H), 3.69 (t, *J* = 4.4 Hz, 4 H), 3.55-3.54 (m, 6 H), 2.93 (hept, *J* = 6.8 Hz, 2 H), 1.15 (d, *J* = 6.8 Hz, 6 H), 1.08 (d, *J* = 7.2 Hz, 6 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.0, 145.4, 138.4, 135.8, 128.4, 128.0, 126.3, 122.7, 122.1, 66.8, 45.8, 33.8, 28.0, 23.7, 22.4. IR 3055, 2962, 2838, 1621, 1588, 1407, 1239, 1119, 970 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{24}H_{33}N_2O$ $[M+H]^+$ 365.2587, found 365.2589.



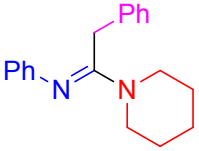
***(E)*-*N*-(1-morpholino-2-phenylethylidene)naphthalen-1-amine (25)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08-8.05 (m, 1 H), 7.84-7.82 (m, 1 H), 7.51-7.48 (m, 3 H), 7.35-7.28 (m, 3 H), 7.22 (t, *J* = 7.2 Hz, 1 H), 7.16 (d, *J* = 7.6 Hz, 2 H), 6.82 (d, *J* = 7.2 Hz, 1 H), 3.68-3.67 (m, 6 H), 3.62 (t, *J* = 4.8 Hz, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.2, 147.1, 136.3, 134.3, 128.6, 128.5, 127.8, 127.7, 126.4, 126.0, 125.7, 124.9, 123.9, 121.9, 115.4, 66.7, 45.4, 33.9. IR 3056, 2961, 2852, 1736, 1614, 1495, 1385, 1254, 1115, 963 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{22}H_{23}N_2O$ $[M+H]^+$ 331.1805, found 331.1803.



***(E)*-*N*-(2-phenyl-1-(pyrrolidin-1-yl)ethylidene)aniline (26)**

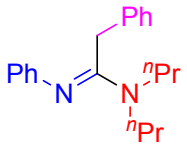
Yellow solid. m.p.: 87-88 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 (t, *J* = 7.6 Hz, 2 H), 7.21-7.13 (m, 5 H), 6.90 (t, *J* = 7.6 Hz, 1 H), 6.79 (d, *J* = 7.6 Hz, 2 H), 3.71 (s, 2 H), 3.55-3.36 (m, 4 H), 1.84 (s, 4 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.7, 151.5, 136.4, 128.6, 128.5, 127.8, 126.2, 122.6, 121.5, 46.9, 35.2. IR 3056, 3021, 2972, 2868, 1604, 1587, 1440, 1426, 1340, 1232, 1156 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{21}N_2$ $[M+H]^+$ 265.1699, found 265.1703.



***(E)*-*N*-(2-phenyl-1-(piperidin-1-yl)ethylidene)aniline (27)**

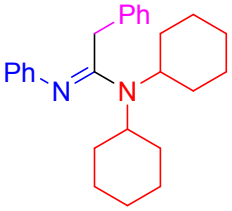
Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30 (t, *J* = 7.6 Hz, 2 H), 7.23-7.17 (m, 5 H), 6.91 (t, *J* = 7.2 Hz, 1 H), 6.77 (d, *J* = 7.2 Hz, 2 H), 3.73 (s, 2 H), 3.45 (t, *J* = 5.2 Hz, 4 H), 1.58 (pent, *J* = 5.2 Hz, 2 H), 1.45 (pent, *J* = 5.6 Hz, 4 H). ^{13}C NMR (100

MHz, $CDCl_3$) δ 156.7, 151.6, 136.8, 128.7, 128.5, 127.8, 126.2, 122.0, 121.4, 45.8, 33.8, 25.7, 24.6. IR 3059, 2934, 2852, 1610, 1589, 1442, 1267, 1204, 1017, 957 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{19}H_{23}N_2$ $[M+H]^+$ 279.1856, found 279.1854.



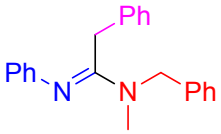
***(E)-N'*,2-diphenyl-*N*,*N*-dipropylacetimidamide (28)**

Yellow oil. ^1H NMR (400 MHz, $CDCl_3$) δ 7.30 (t, *J* = 7.6 Hz, 2 H), 7.23-7.16 (m, 5 H), 6.90 (t, *J* = 6.8 Hz, 1 H), 6.76 (d, *J* = 8.4 Hz, 2 H), 3.72 (s, 2 H), 3.25 (s, 4 H), 1.60 (d, *J* = 6.0 Hz, 4 H), 0.87 (t, *J* = 7.6 Hz, 6 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.0, 152.0, 137.0, 128.6, 128.5, 127.7, 126.2, 122.1, 121.2, 49.4, 33.8, 21.2, 11.4. IR 3060, 2962, 2872, 1611, 1590, 1452, 1235, 1142, 1030, 761 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{20}H_{27}N_2$ $[M+H]^+$ 295.2169, found 295.2170.



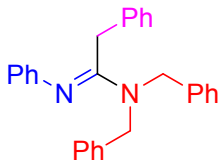
***(E)-N*,*N*-dicyclohexyl-*N'*,2-diphenylacetimidamide (29)**

Yellow solid. m.p.: 108-109 °C. ^1H NMR (400 MHz, $CDCl_3$) δ 7.28 (t, *J* = 7.2 Hz, 2 H), 7.21-7.16 (m, 5 H), 6.86 (t, *J* = 7.2 Hz, 1 H), 6.76 (d, *J* = 7.6 Hz, 2 H), 3.68 (s, 2 H), 3.16 (s, 2 H), 1.70-1.09 (m, 20 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 154.2, 152.1, 137.7, 128.7, 128.4, 127.9, 126.1, 121.6, 120.5, 57.4, 35.3, 30.7, 26.5, 25.5. IR 3055, 2926, 2844, 1609, 1592, 1446, 1365, 1231, 1145, 894, 729 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{26}H_{35}N_2$ $[M+H]^+$ 375.2795, found 375.2800.



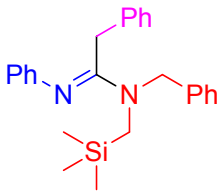
***(E)-N*-benzyl-*N*-methyl-*N'*,2-diphenylacetimidamide (30)**

Yellow oil. ^1H NMR (400 MHz, $CDCl_3$) δ 7.41-7.32 (m, 5 H), 7.28-7.21 (m, 7 H), 6.98 (t, *J* = 7.2 Hz, 1 H), 6.87 (d, *J* = 8.0 Hz, 2 H), 4.64 (s, 2 H), 3.81 (s, 2 H), 2.99 (s, 3 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.3, 151.5, 138.5, 136.5, 128.7, 128.6, 128.5, 127.8, 127.1, 127.0, 126.3, 122.1, 121.6, 53.0, 35.9, 33.9. IR 3060, 3026, 2923, 1610, 1495, 1397, 1295, 1212, 1111, 954 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{22}H_{23}N_2$ $[M+H]^+$ 315.1856, found 315.1853.



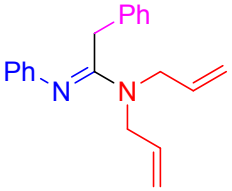
***(E)*-*N*,*N*-dibenzyl-*N*',2-diphenylacetimidamide (31)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39-7.35 (m, 4 H), 7.33-7.30 (m, 4 H), 7.27-7.23 (m, 7 H), 7.18 (d, *J* = 7.6 Hz, 2 H), 6.97 (t, *J* = 7.2 Hz, 1 H), 6.87 (d, *J* = 7.6 Hz, 2 H), 4.61 (s, 4 H), 3.79 (s, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.3, 151.3, 138.5, 136.5, 128.8, 128.7, 128.5, 127.8, 127.3, 127.0, 126.5, 122.0, 121.7, 49.9, 33.8. IR 3061, 3027, 2919, 2851, 1613, 1494, 1361, 1224, 1075, 955 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{28}H_{27}N_2$ $[M+H]^+$ 391.2169, found 391.2171.



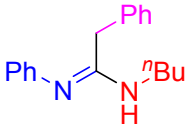
***(E)*-*N*-benzyl-*N*',2-diphenyl-*N*-((trimethylsilyl)methyl)acetimidamide (32)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.39-7.32 (m, 5 H), 7.27-7.16 (m, 7 H), 6.97 (t, *J* = 7.2 Hz, 1 H), 6.85 (d, *J* = 7.2 Hz, 2 H), 4.46 (s, 2 H), 3.77 (s, 2 H), 3.06 (s, 2 H), 0.14 (d, *J* = 4.0 Hz, 9 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.5, 151.8, 138.7, 137.0, 128.8, 128.7, 128.0, 128.0, 127.1, 126.7, 126.5, 122.3, 121.4, 53.4, 40.0, 33.8, -0.6. IR 3061, 3027, 2950, 2896, 1590, 1450, 1219, 1145, 853, 697 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{25}H_{31}N_2Si$ $[M+H]^+$ 387.2251, found 387.2256.



***(E)*-*N*,*N*-diallyl-*N*',2-diphenylacetimidamide (33)**

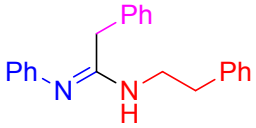
Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.6 Hz, 2 H), 7.24-7.16 (m, 5 H), 6.92 (t, *J* = 7.6 Hz, 1 H), 6.78 (d, *J* = 7.6 Hz, 2 H), 5.84-5.78 (m, 2 H), 5.16 (dd, *J* = 11.6 Hz, *J* = 1.2 Hz, 4 H), 3.96 (s, 4 H), 3.73 (s, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.3, 151.4, 136.6, 134.2, 128.7, 128.6, 127.7, 126.3, 122.0, 121.6, 116.2, 49.2, 33.7. IR 3061, 3026, 2922, 2853, 1613, 1445, 1304, 1230, 1167, 922 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{20}H_{23}N_2$ $[M+H]^+$ 291.1856, found 291.1859.



***(E)*-*N*-butyl-*N*',2-diphenylacetimidamide (34)**

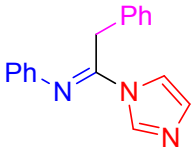
Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33-7.24 (m, 5 H), 7.14 (d, *J* = 6.8 Hz, 2 H), 6.98 (t, *J* = 7.2 Hz, 1 H), 6.84 (d, *J* = 7.6 Hz, 2 H), 4.10 (s, 1 H), 3.51 (s, 2 H), 3.31 (s, 2 H), 1.48 (t, *J* = 6.8 Hz, 2 H), 1.31 (t, *J* = 7.2 Hz, 2 H), 0.90 (t, *J* = 7.2 Hz, 3

H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.0, 151.4, 136.3, 129.0, 128.7, 126.8, 122.3, 121.8, 41.0, 37.0, 31.1, 20.1, 13.8. IR 3436, 3061, 3027, 2957, 2870, 1633, 1591, 1486, 1211, 762, 699 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{18}H_{23}N_2$ $[M+H]^+$ 267.1856, found 267.1858.



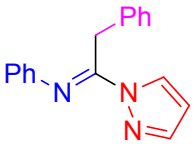
***(E)*-*N*-phenethyl-*N*',2-diphenylacetimidamide (35)**

Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30-7.20 (m, 8 H), 7.07-6.98 (m, 5 H), 6.85 (d, *J* = 7.6 Hz, 2 H), 4.17 (s, 1 H), 3.59 (s, 2 H), 3.46 (s, 2 H), 2.83 (t, *J* = 6.4 Hz, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.9, 151.3, 139.4, 136.0, 129.2, 128.9, 128.9, 128.8, 128.5, 127.0, 126.2, 122.4, 122.0, 42.2, 37.1, 35.1. IR 3431, 3060, 3026, 2924, 2858, 1634, 1591, 1486, 1234, 1071, 1030, 698 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{22}H_{23}N_2$ $[M+H]^+$ 315.1856, found 315.1858.



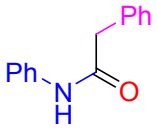
***(E)*-*N*-(1-(1*H*-imidazol-1-yl)-2-phenylethylidene)aniline (36)**

Yellow solid. m.p.: 57-58 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.09 (s, 1 H), 7.59 (s, 1 H), 7.36-7.22 (m, 5 H), 7.13 (t, *J* = 6.8 Hz, 3 H), 7.04 (s, 1 H), 6.88 (d, *J* = 7.6 Hz, 2 H), 4.08 (s, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.4, 147.1, 136.3, 134.4, 130.1, 129.4, 129.3, 127.7, 127.4, 124.5, 119.9, 116.7, 35.1. IR 3160, 3063, 2926, 1670, 1594, 1475, 1378, 1296, 1212, 1045, 984 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{17}H_{15}N_3Na$ $[M+Na]^+$ 284.1158, found 284.1157.



***(E)*-*N*-(2-phenyl-1-(1*H*-pyrazol-1-yl)ethylidene)aniline (37)**

Yellow solid. m.p.: 91-92 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.50 (d, *J* = 2.8 Hz, 1 H), 7.71 (s, 1 H), 7.34 (t, *J* = 7.6 Hz, 2 H), 7.23-7.17 (m, 3 H), 7.13 (t, *J* = 7.2 Hz, 1 H), 7.08 (d, *J* = 7.2 Hz, 2 H), 6.84 (d, *J* = 7.6 Hz, 2 H), 6.45 (s, 1 H), 4.37 (s, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 154.2, 147.5, 142.1, 135.9, 129.1, 128.5, 128.4, 127.6, 126.5, 124.0, 120.3, 108.5, 33.7. IR 3155, 3058, 3026, 1668, 1594, 1422, 1392, 1226, 1039, 943 cm^{-1}. HRMS (ESI) *m/z* calcd for $C_{17}H_{15}N_3Na$ $[M+Na]^+$ 284.1158, found 284.1159.



***N*,2-diphenylacetamide (38)**[4]

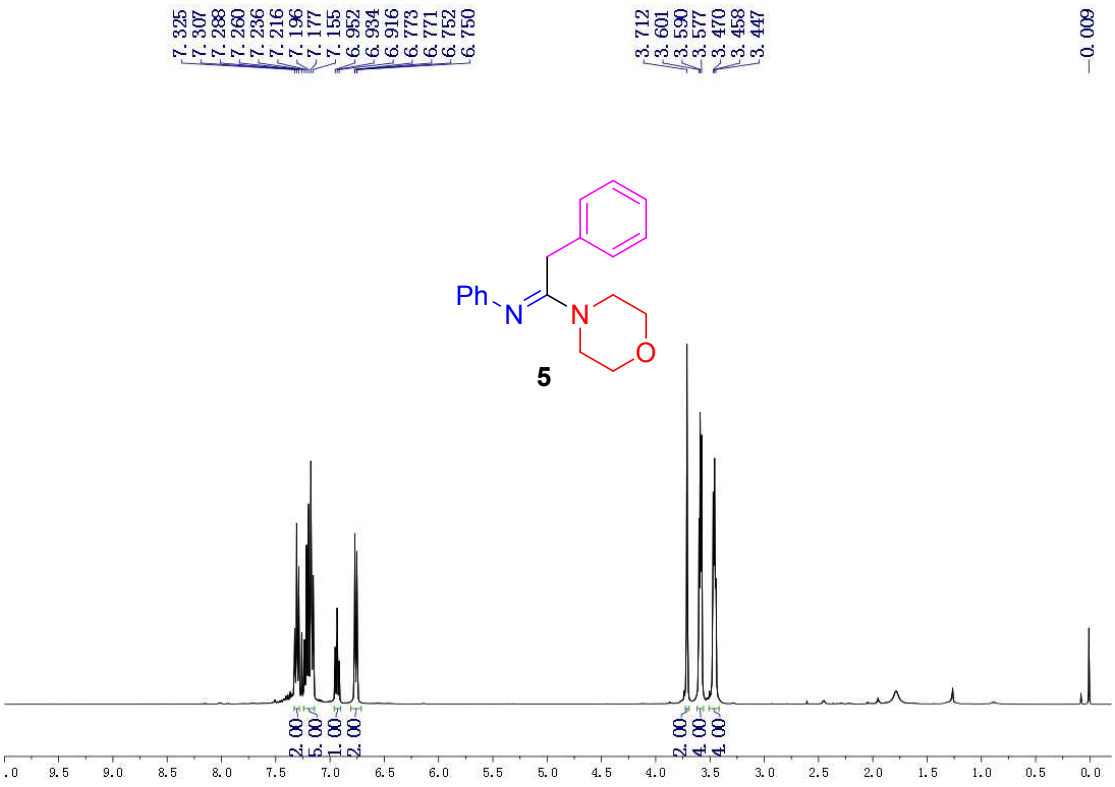
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44-7.26 (m, 9 H), 7.09 (t, *J* = 7.6 Hz, 1 H), 3.72 (s, 2 H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 169.2, 137.6, 134.4, 129.4, 129.1, 128.9, 127.6, 124.4, 119.8, 44.7.

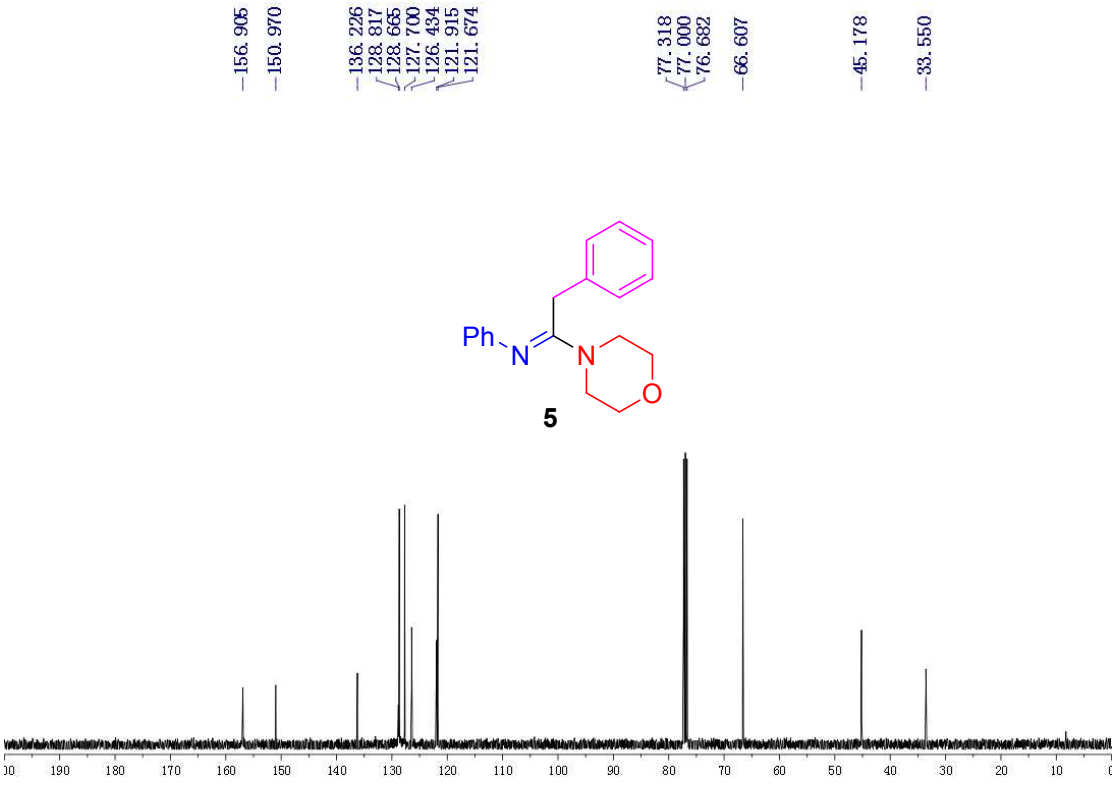
7. Reference.

1. F. Ye, X. Ma, Q. Xiao, H. Li, Y. Zhang and J. Wang, *J. Am. Chem. Soc.,* 2012, **134**, 5742.
2. S. Kamijo, T. Jin and Y. Yamamoto, *Angew. Chem., Int. Ed.,* 2002, **41**, 1780.
3. A. R. Katritzky, C. Cai and S. K. Singh, *J. Org. Chem.,* 2006, **71**, 3375.
4. L. Gu, J. Lim, J. L. Cheong and S. S. Lee, *Chem. Commun.,* 2014, **50**, 7017.

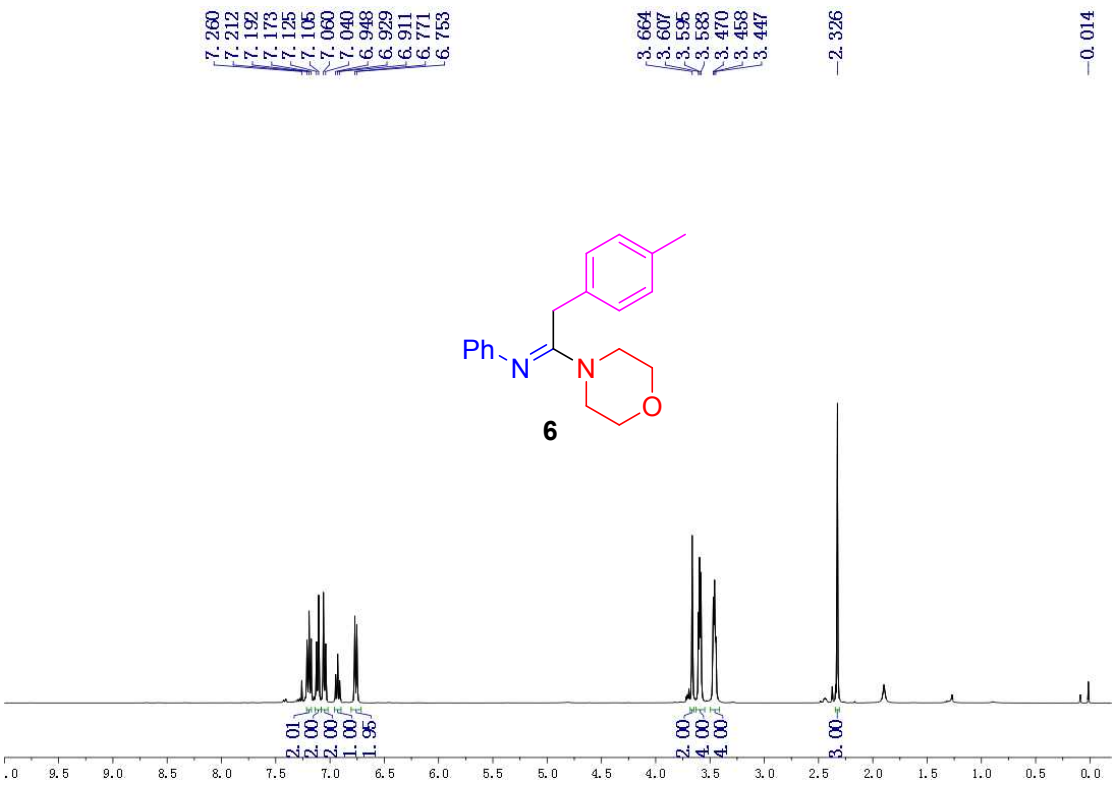
8. ^{1}H and ^{13}C Spectra.

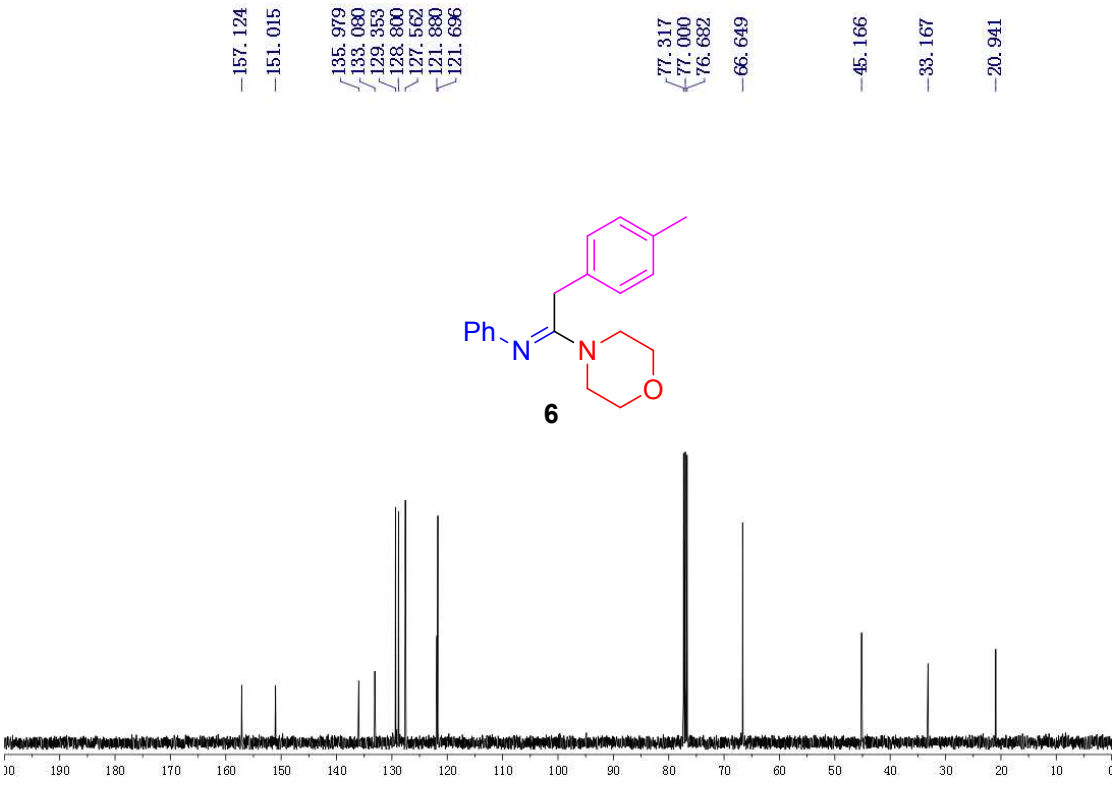
(E)-N-(1-morpholino-2-phenylethylidene)aniline (5)



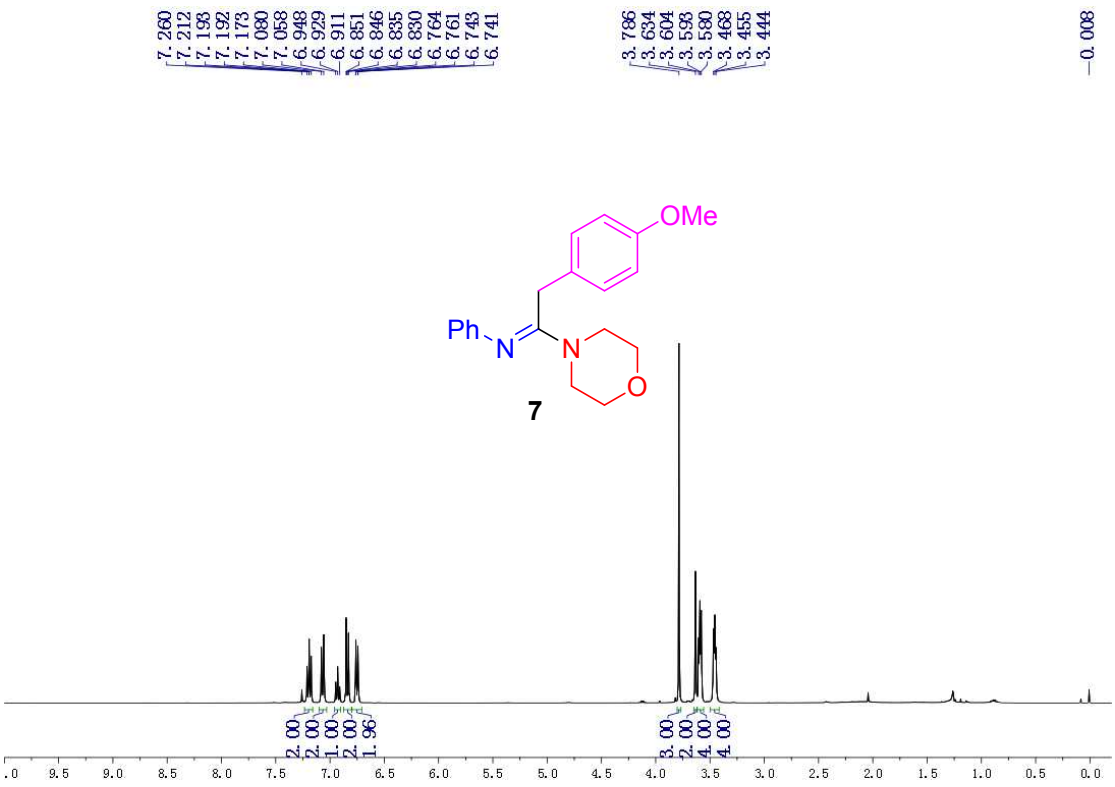


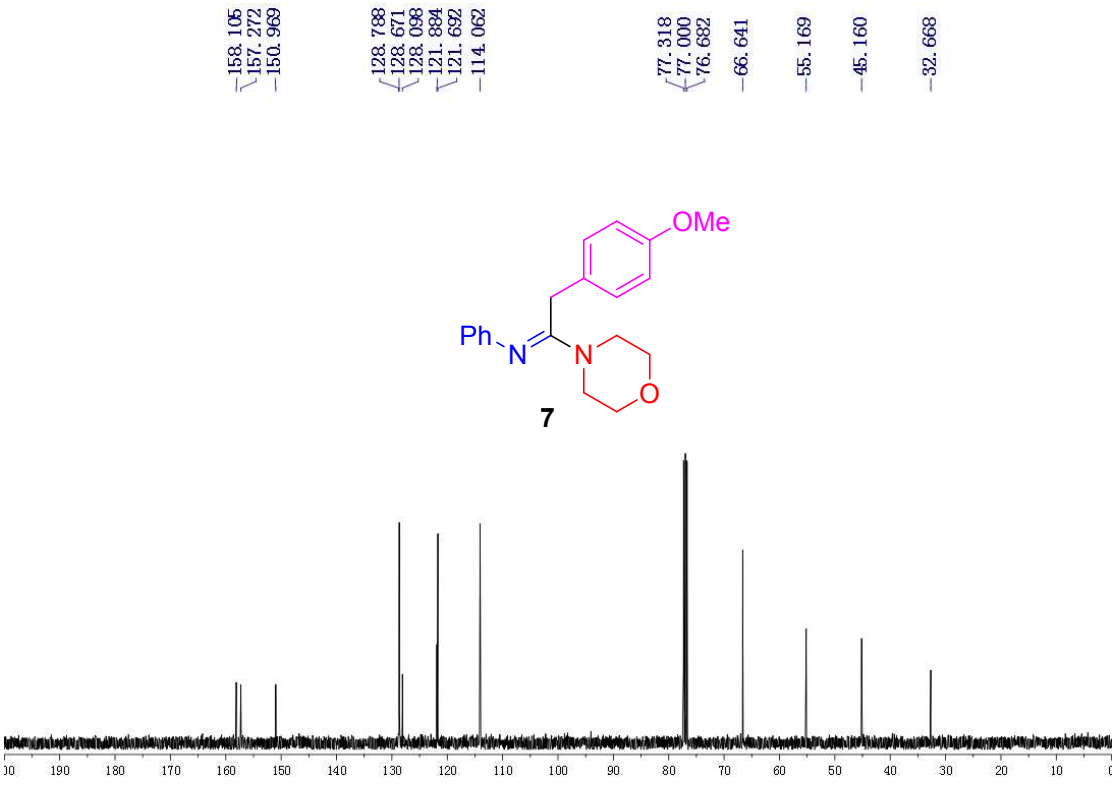
***(E)-N*-(1-morpholino-2-(4-tolyl)ethylidene)aniline (6)**



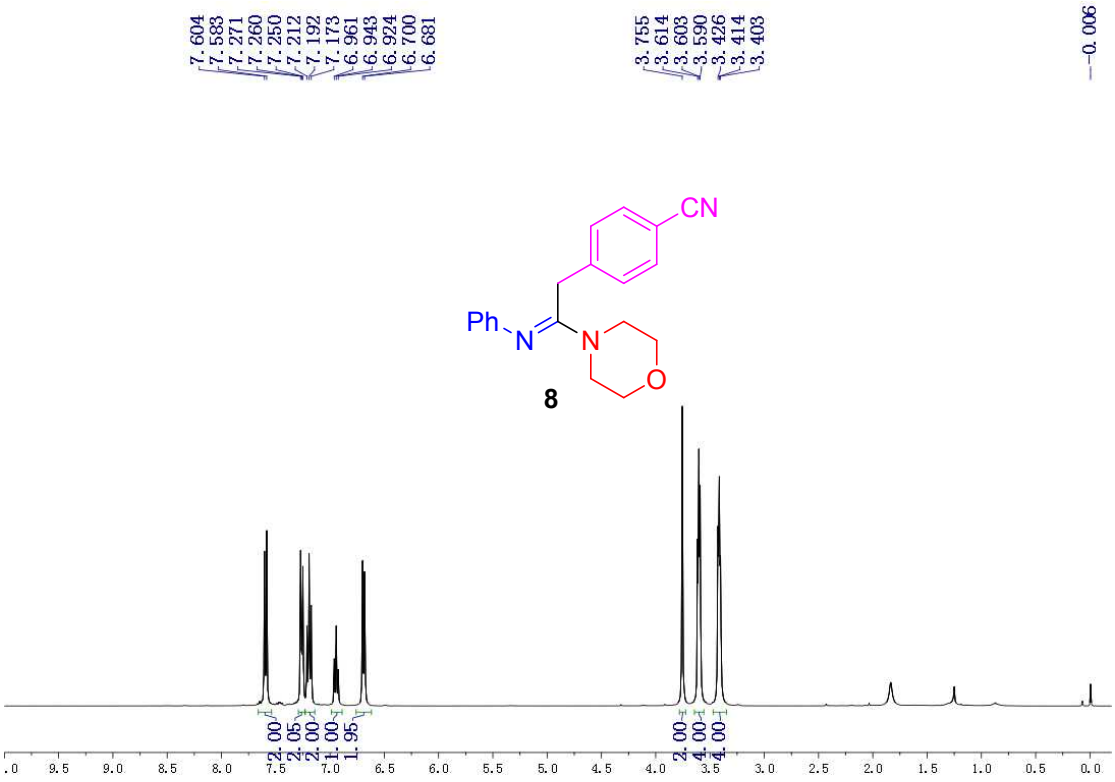


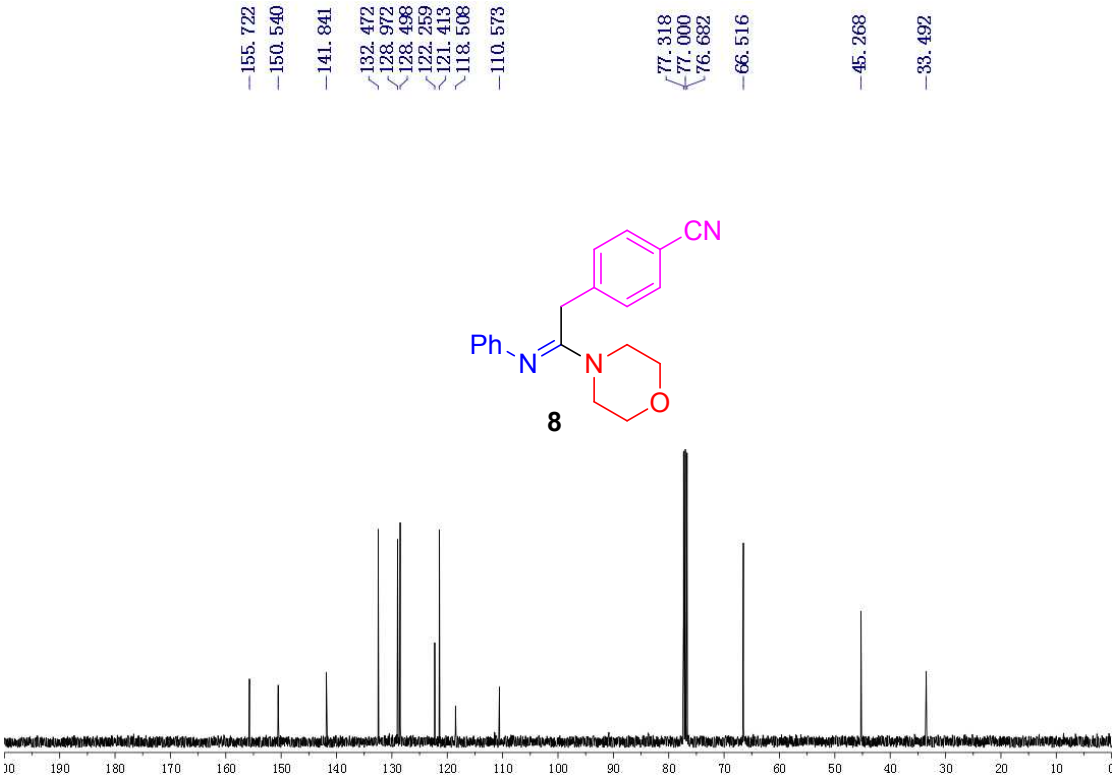
***(E)-N*-(2-(4-methoxyphenyl)-1-morpholinoethylidene)aniline (7)**



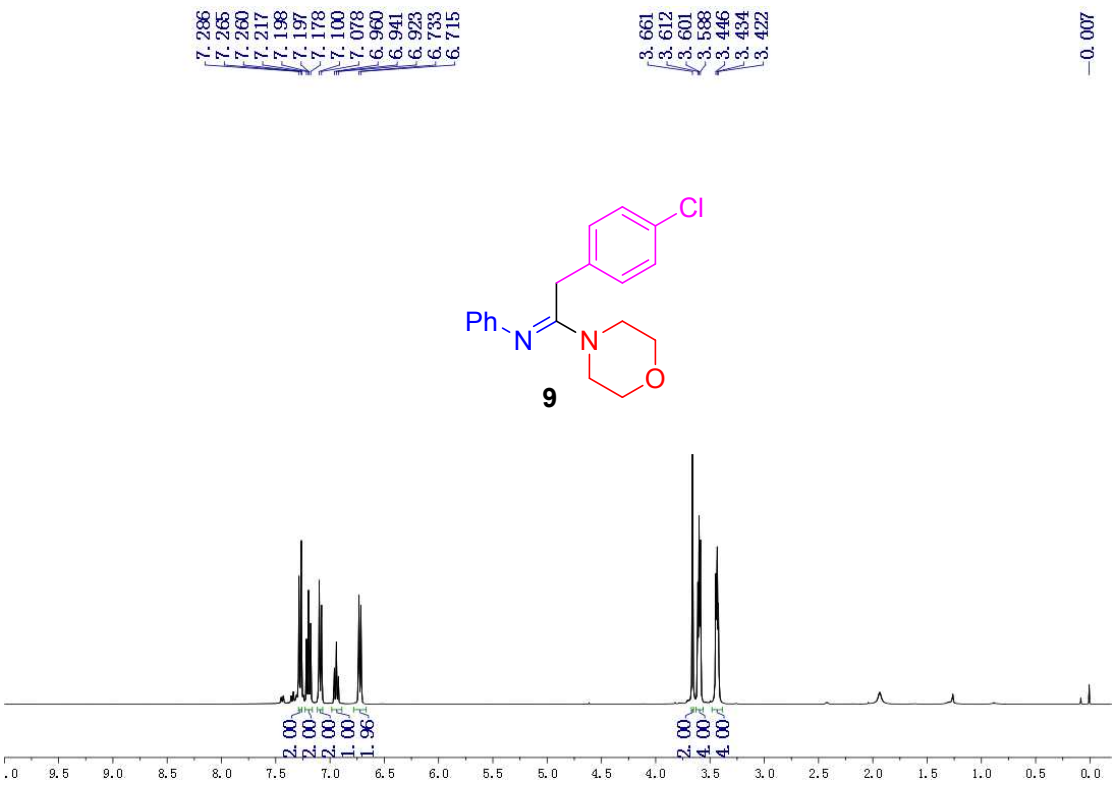


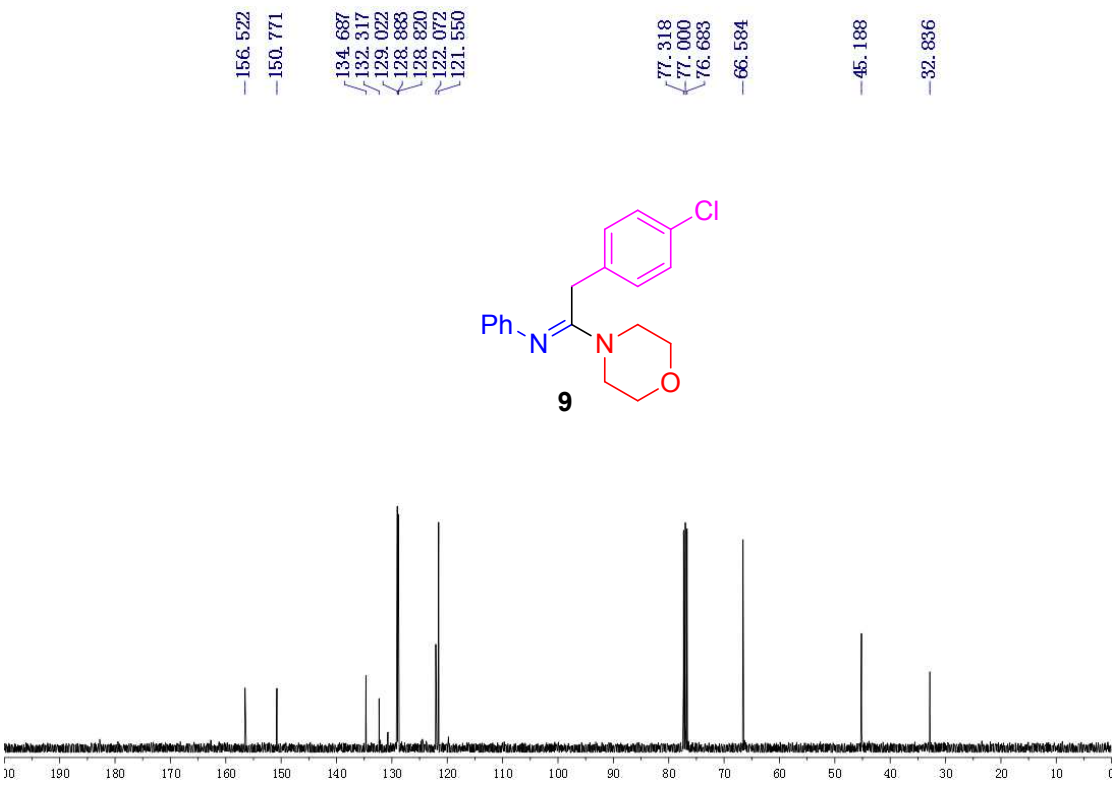
***(E)*-4-(2-morpholino-2-(phenylimino)ethyl)benzonitrile (8)**



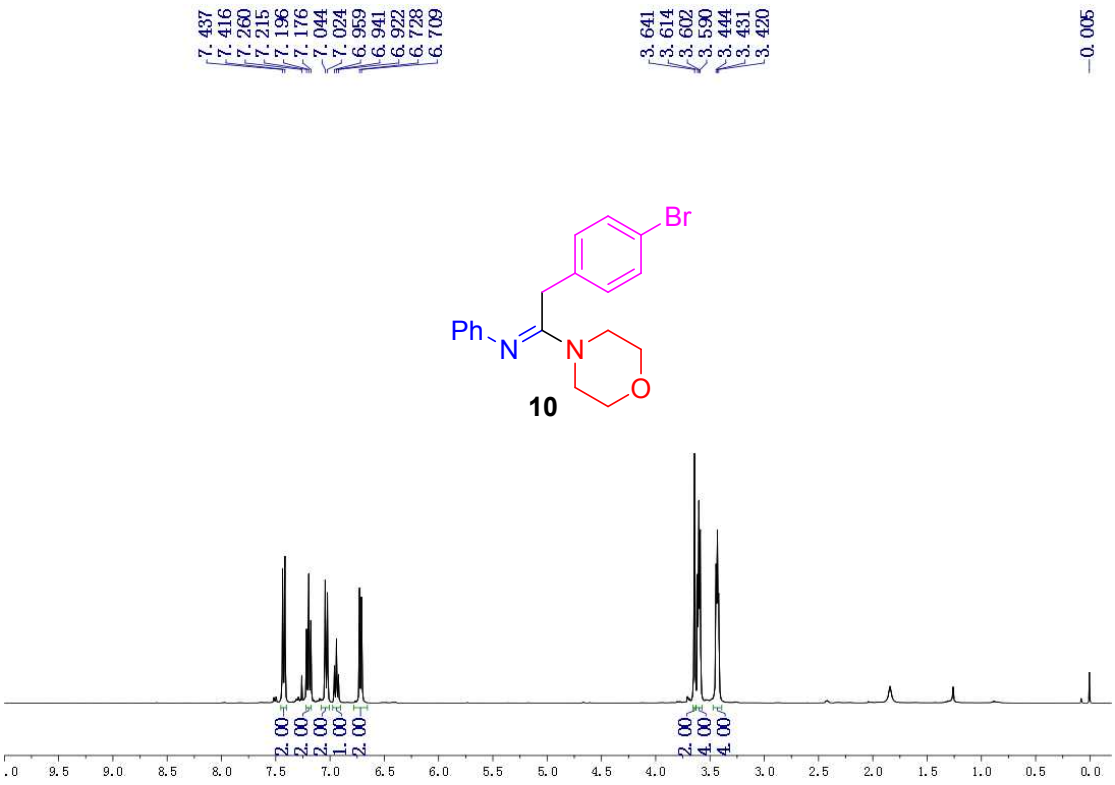


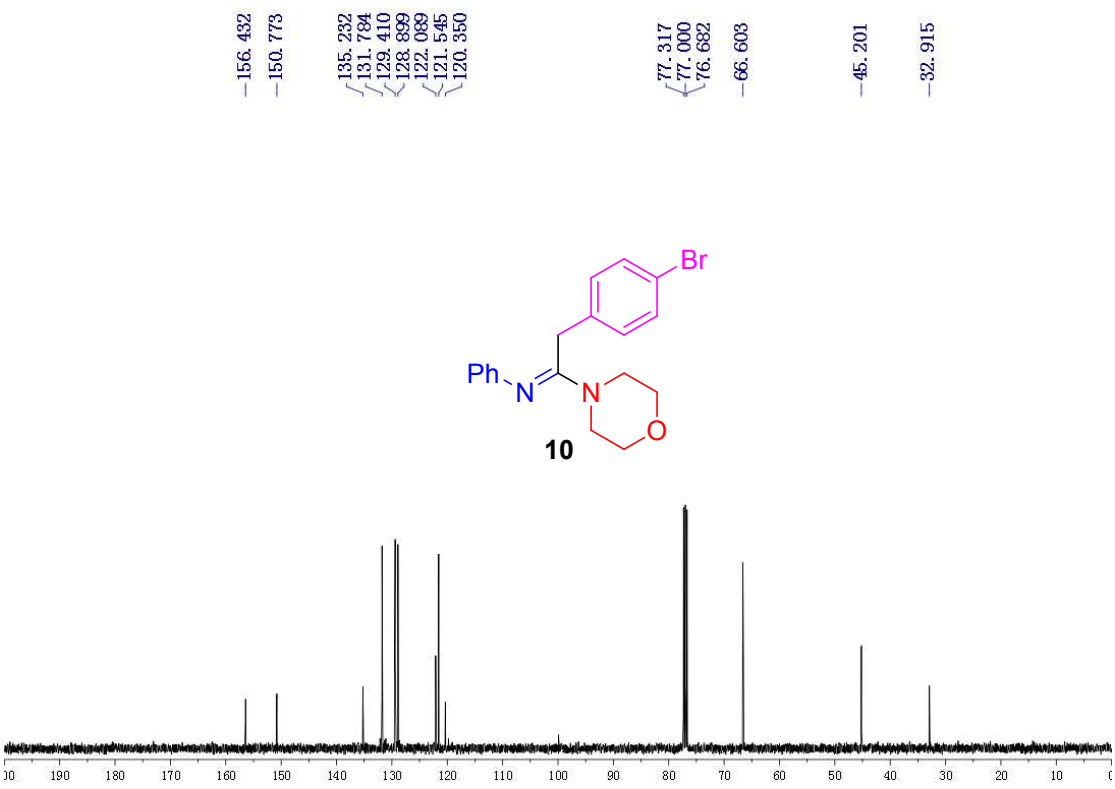
***(E)-N*-(2-(4-chlorophenyl)-1-morpholinoethylidene)aniline (9)**



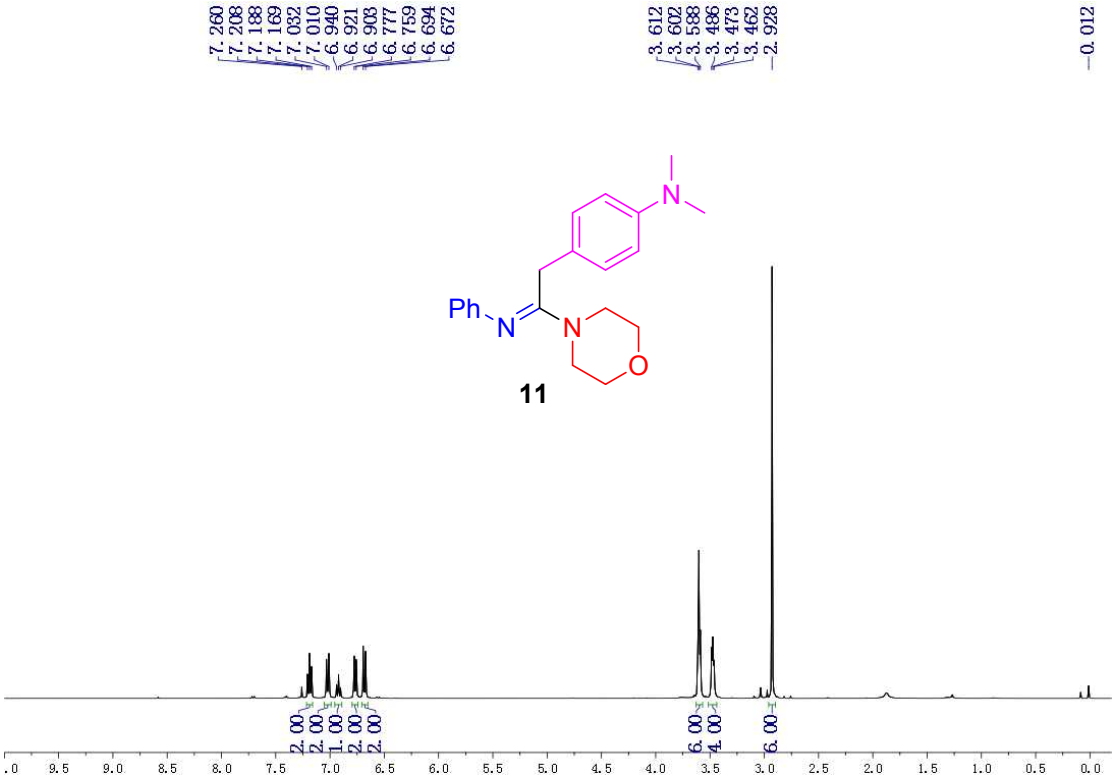


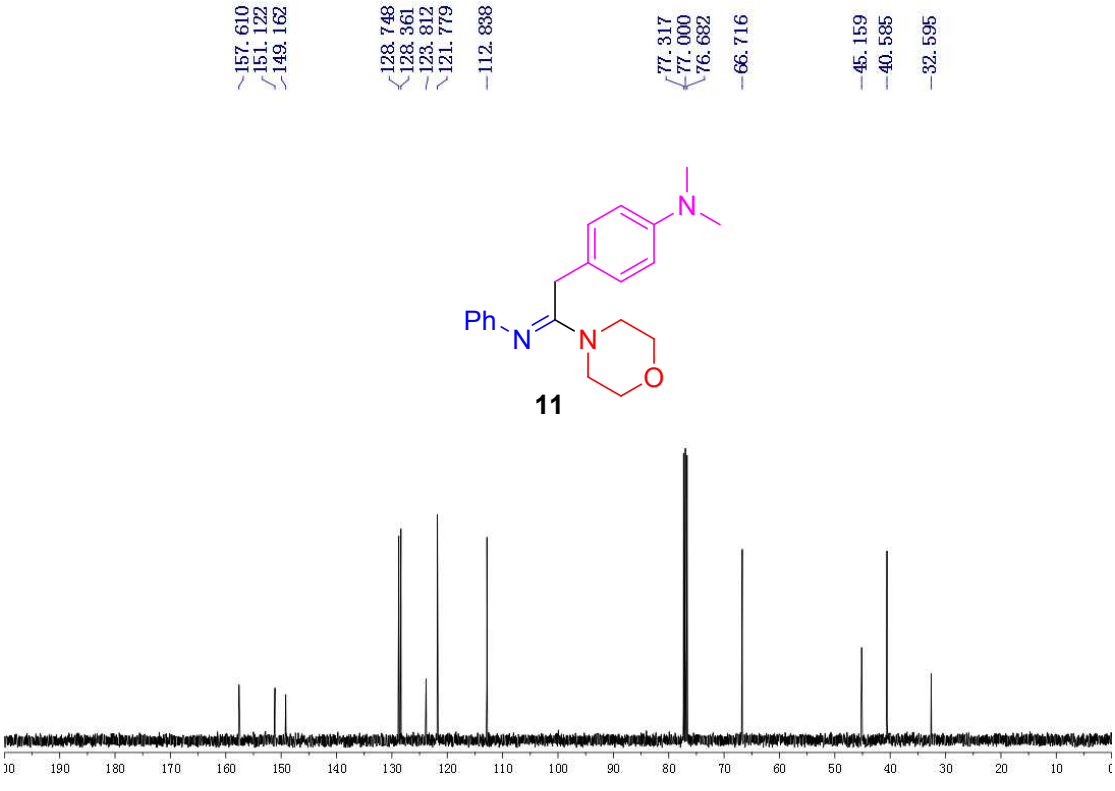
***(E)-N*-(2-(4-bromophenyl)-1-morpholinoethylidene)aniline (10)**



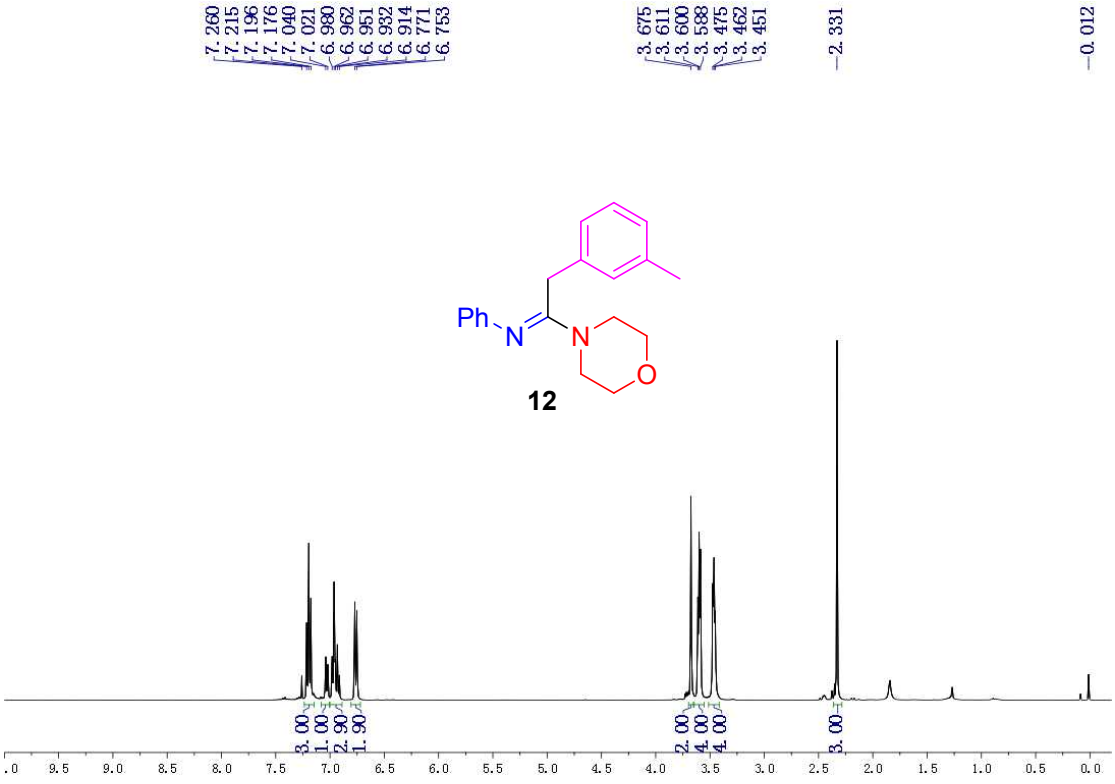


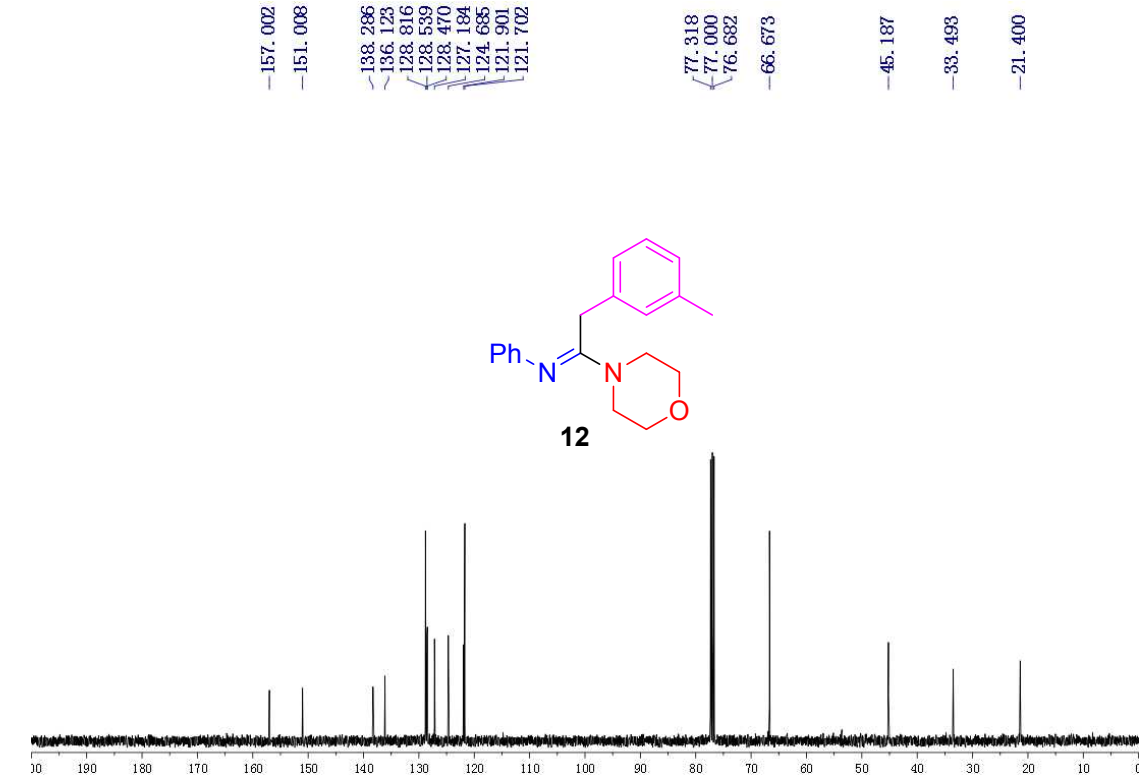
***(E)*-*N*,*N*-dimethyl-4-(2-morpholino-2-(phenylimino)ethyl)aniline (11)**



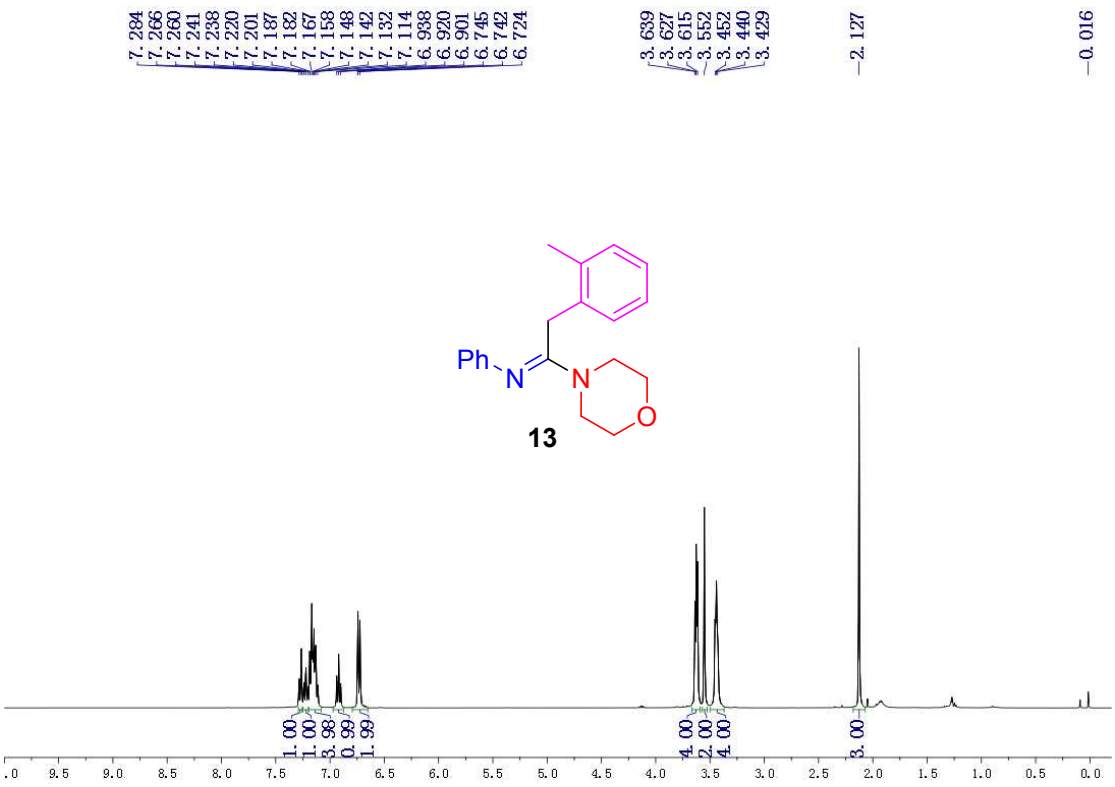


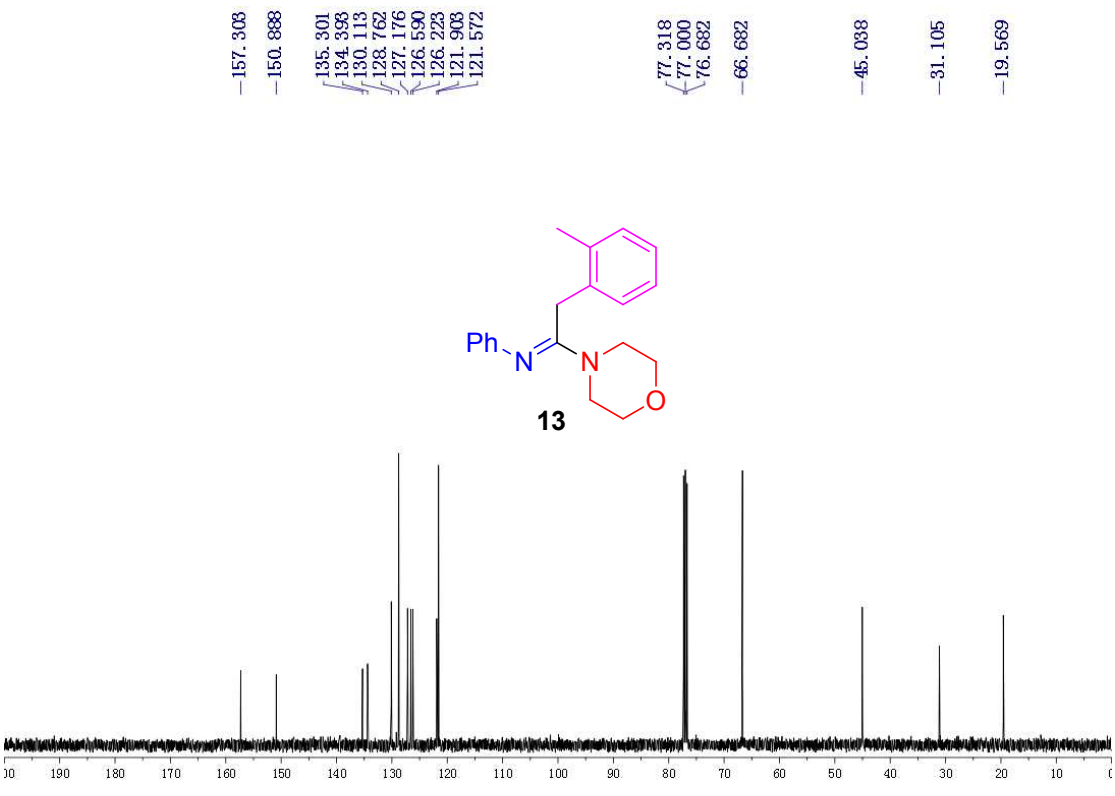
***(E)-N*-(1-morpholino-2-(3-tolyl)ethylidene)aniline (12)**



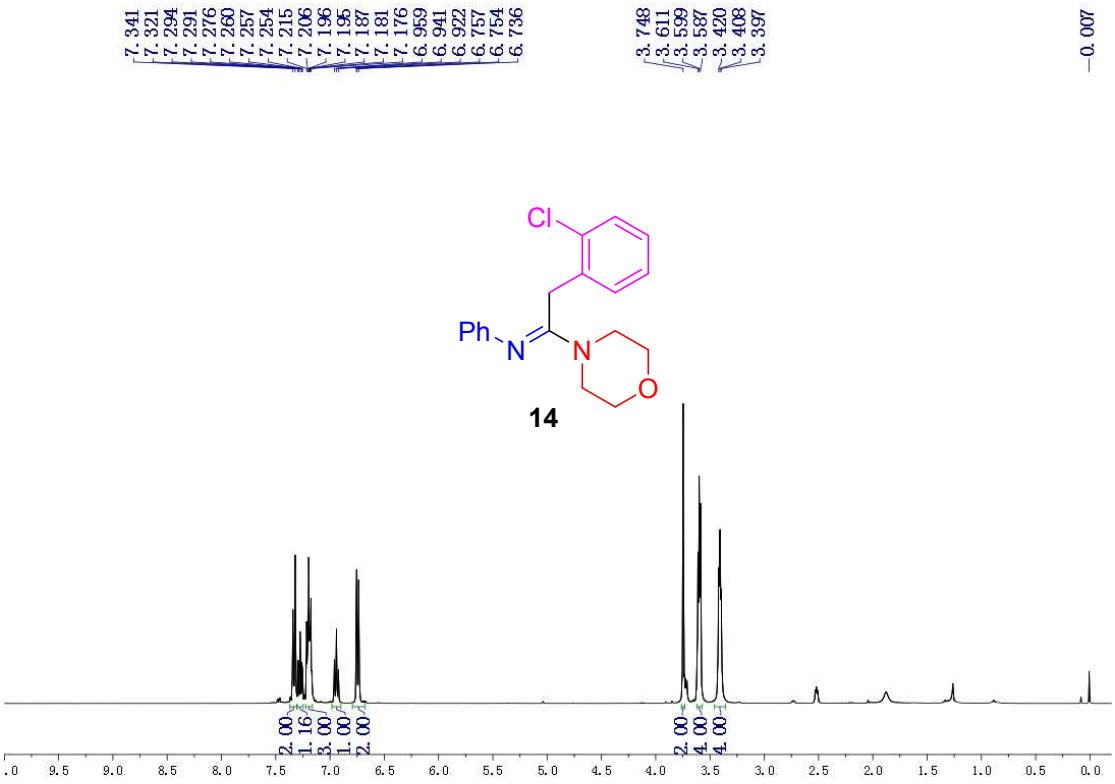


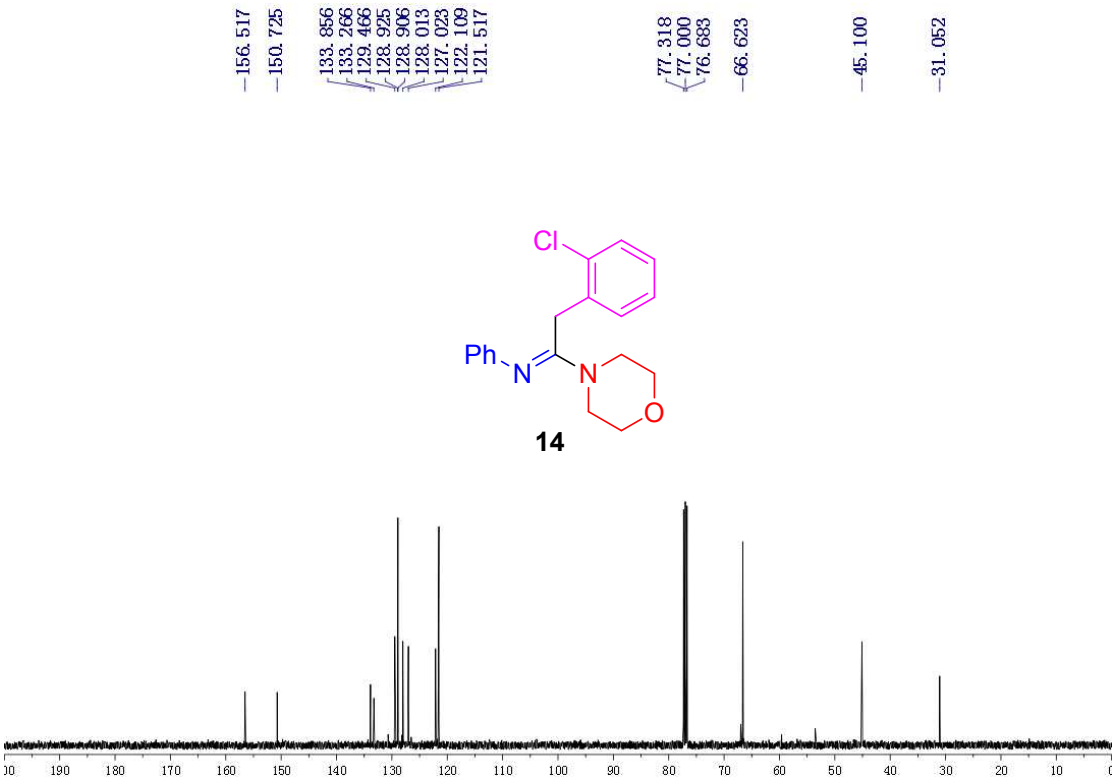
***(E)-N*-(1-morpholino-2-(2-tolyl)ethylidene)aniline (13)**



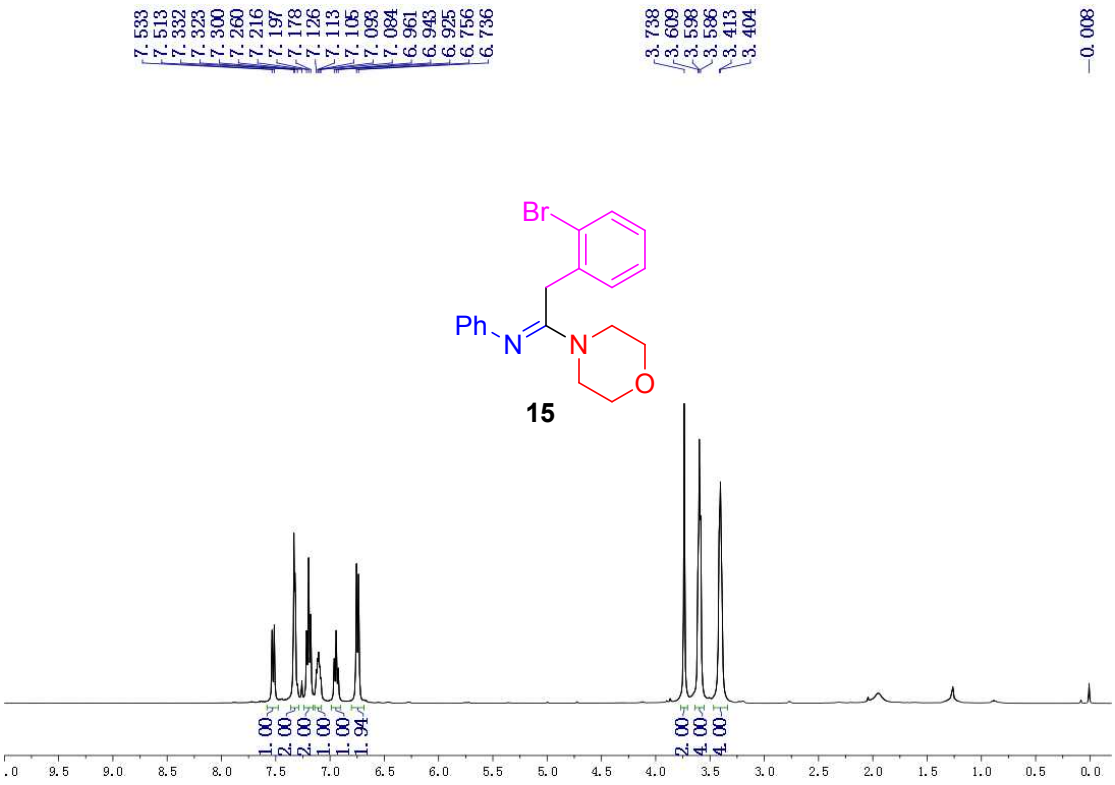


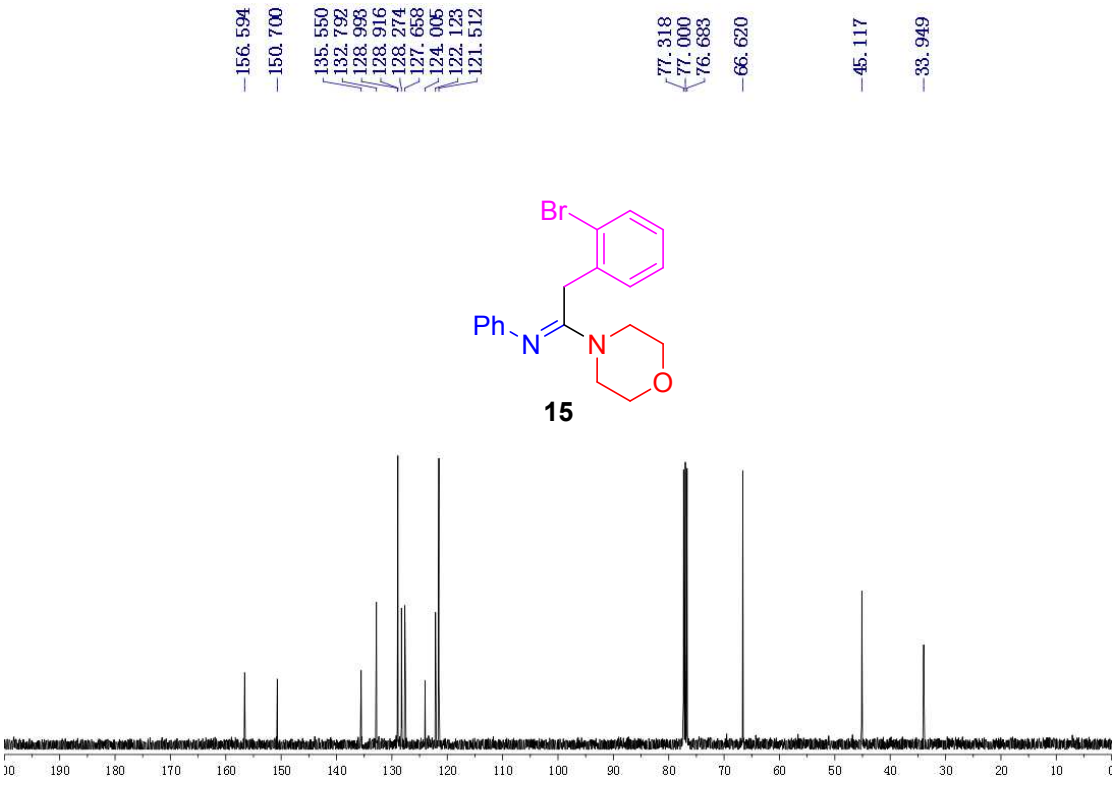
***(E)-N*-(2-(2-chlorophenyl)-1-morpholinoethylidene)aniline (14)**



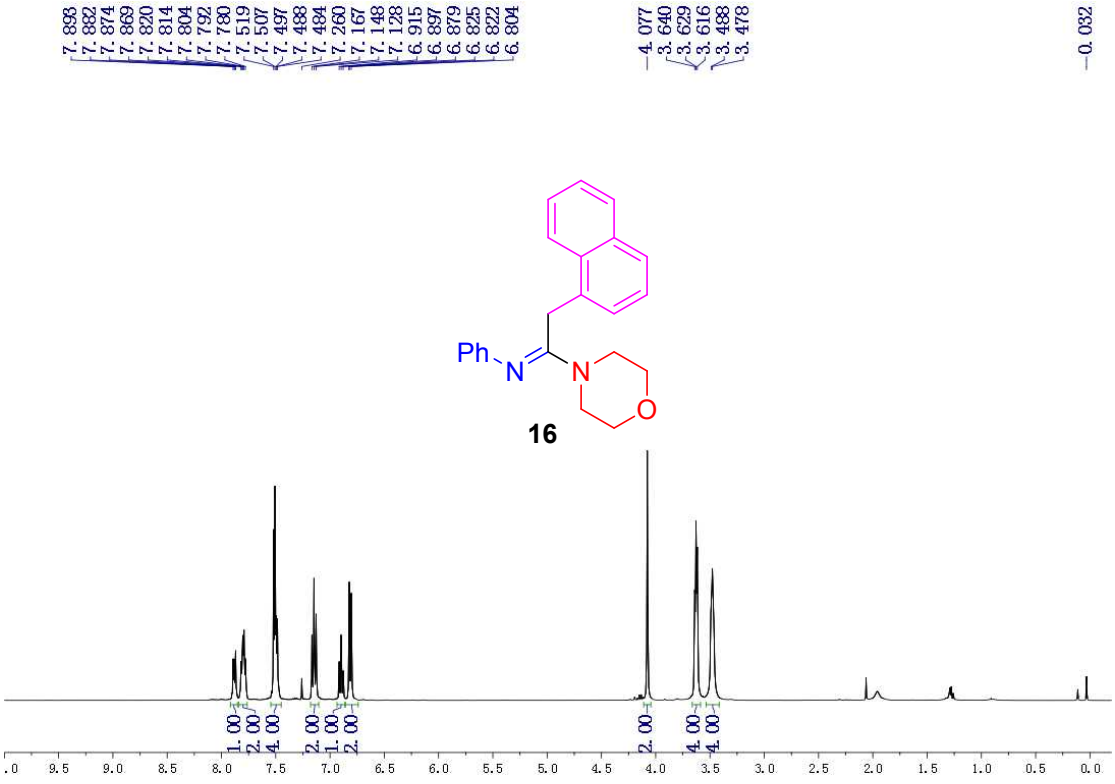


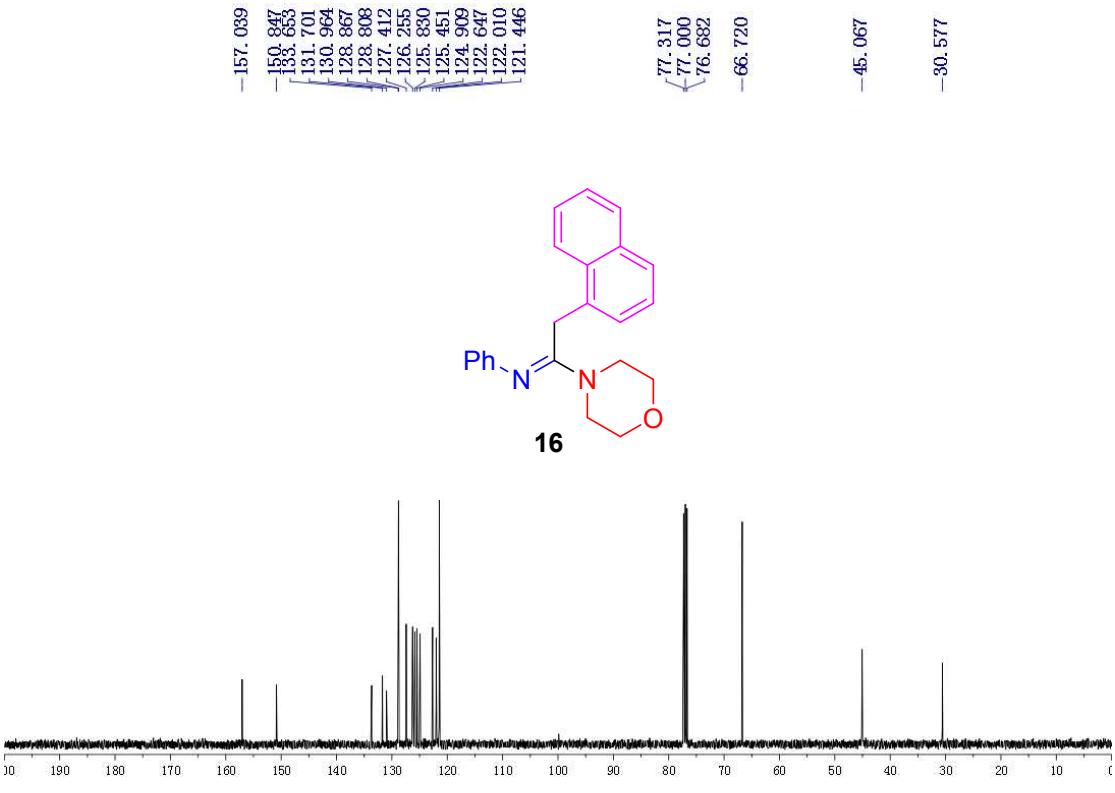
***(E)-N*-(2-(2-bromophenyl)-1-morpholinoethylidene)aniline (15)**



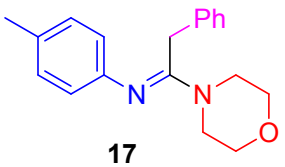


(E)-N-(1-morpholino-2-(naphthalen-1-yl)ethylidene)aniline (16)





***(E)*-4-methyl-*N*-(1-morpholino-2-phenylethylidene)aniline(17)**



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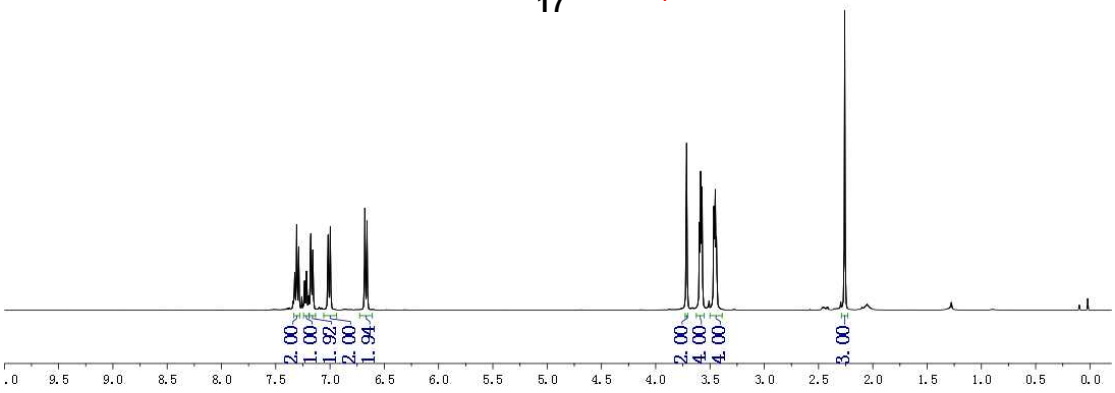

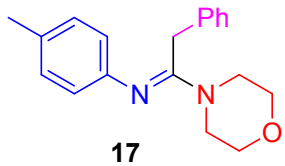

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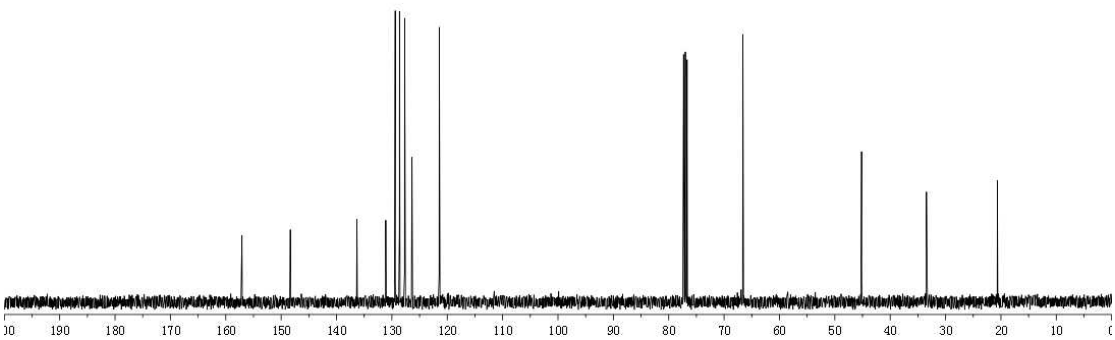
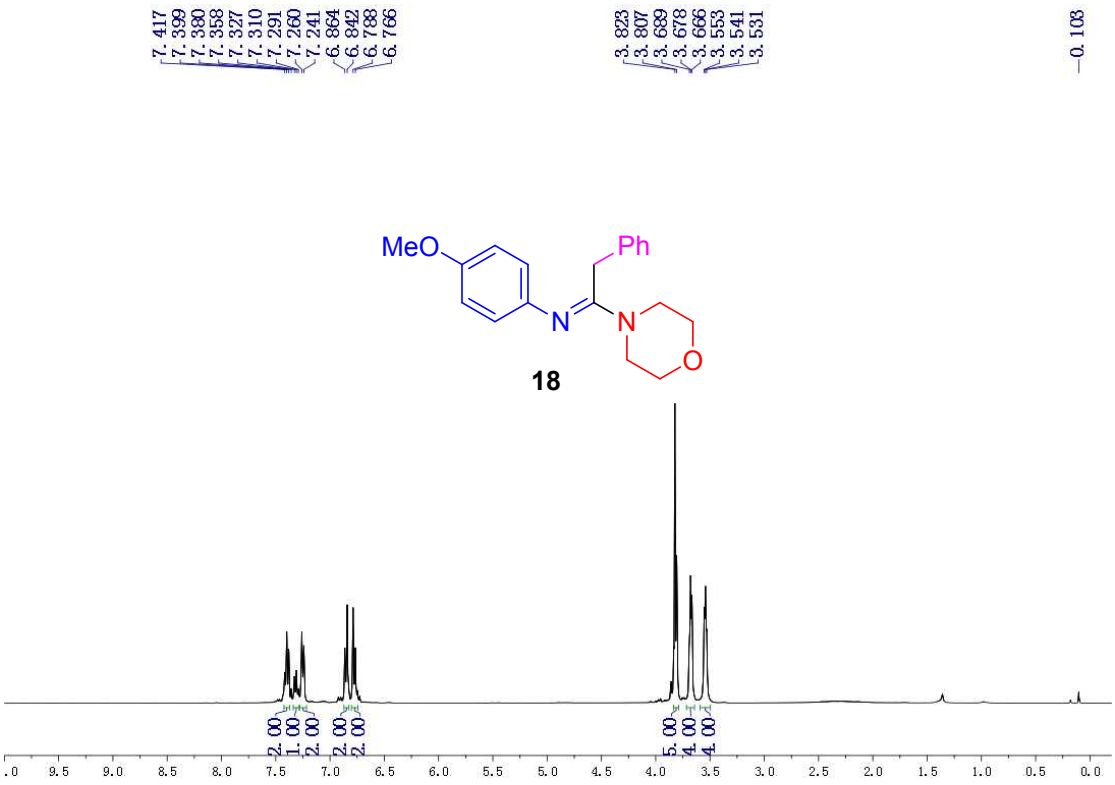
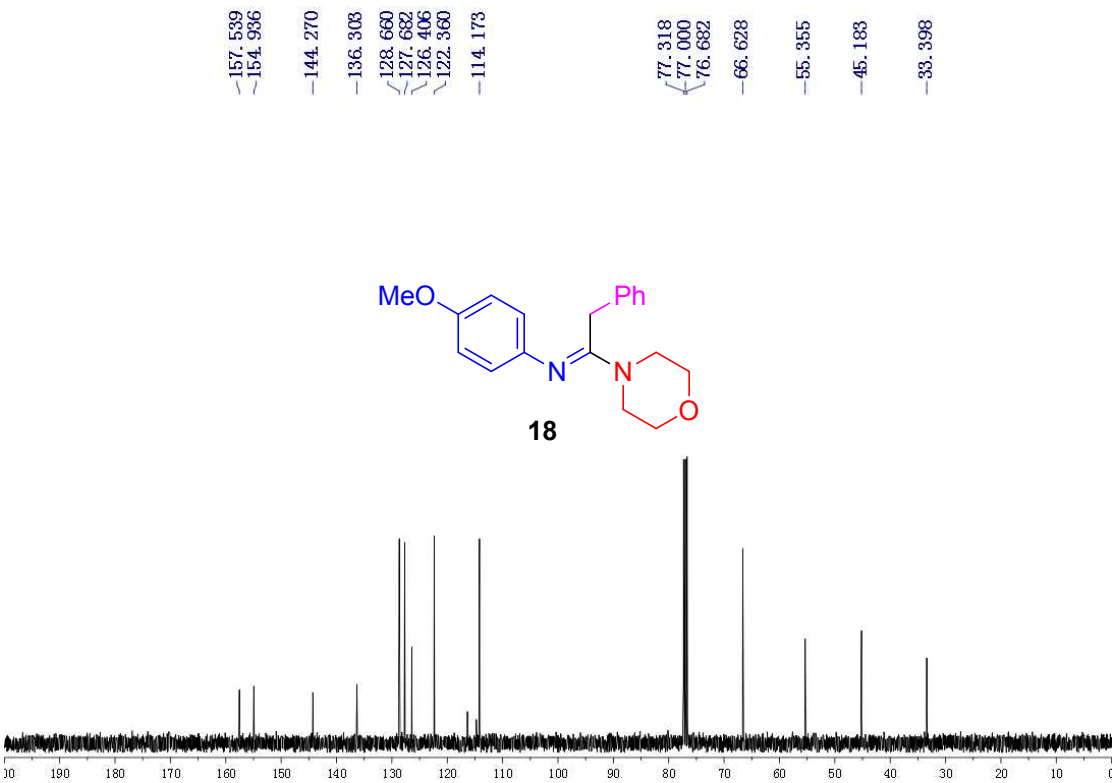
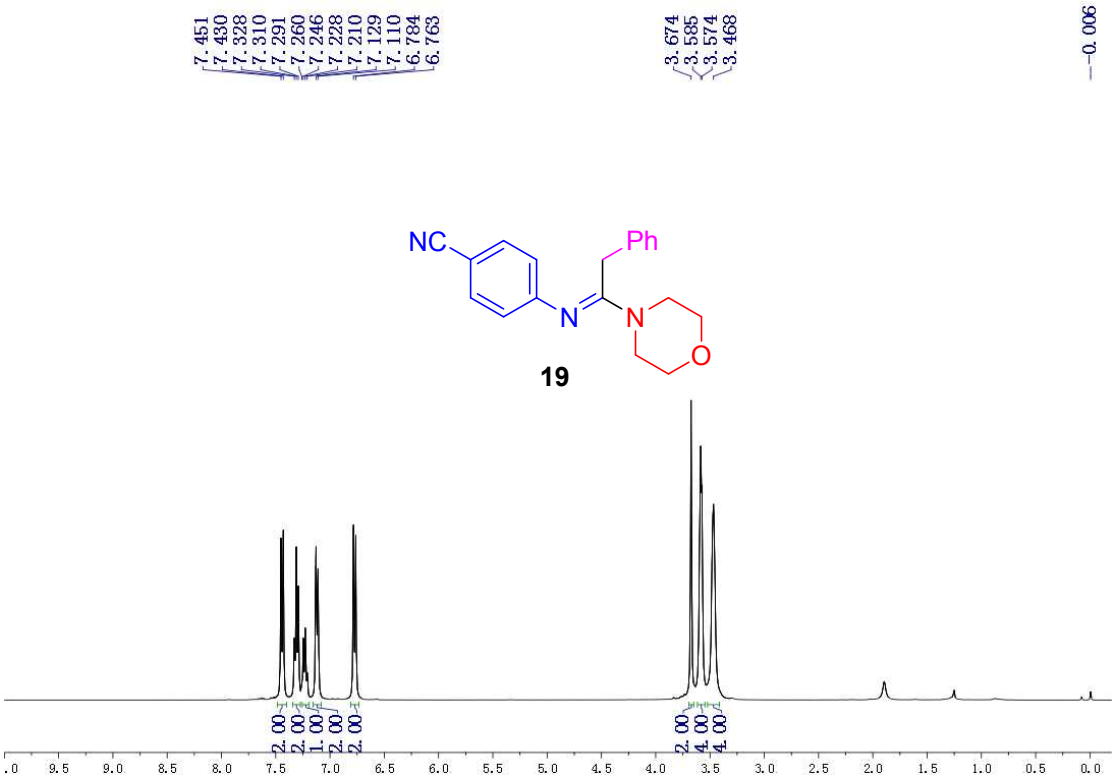


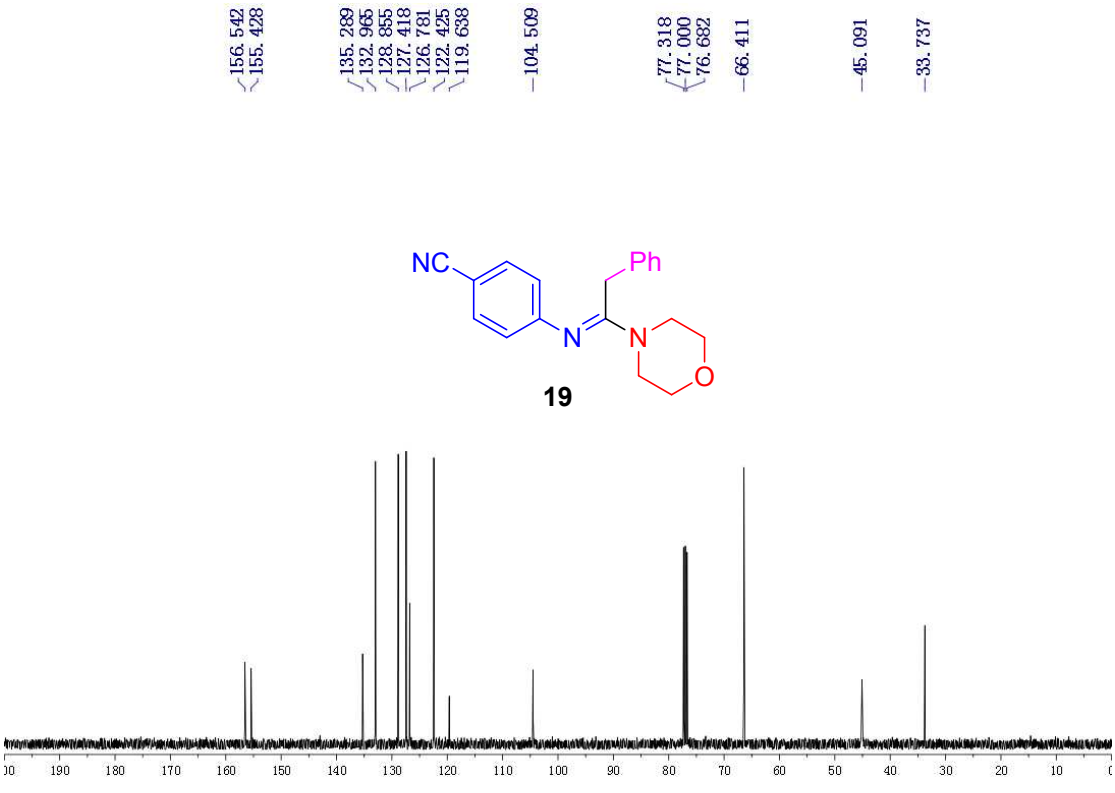
***(E)*-4-methoxy-*N*-(1-morpholino-2-phenylethylidene)aniline (18)**



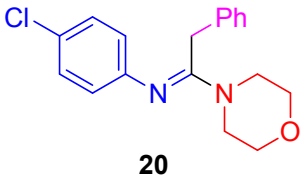


(E)-4-((1-morpholino-2-phenylethylidene)amino)benzonitrile (19)





***(E)*-4-chloro-*N*-(1-morpholino-2-phenylethylidene)aniline (20)**



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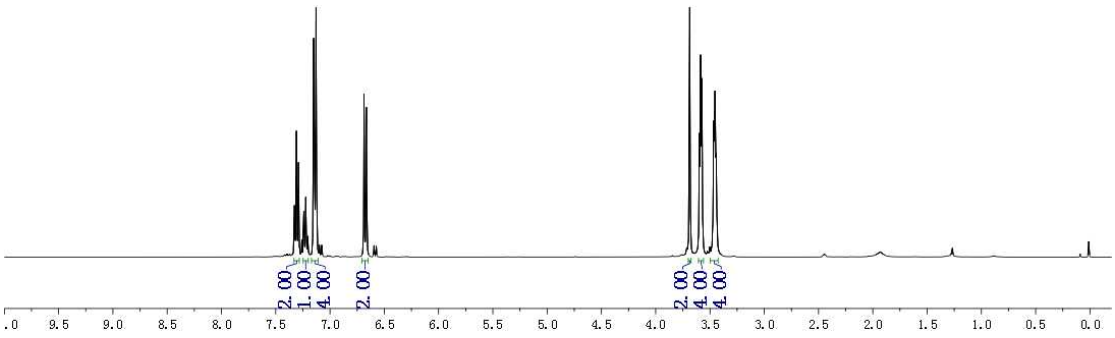

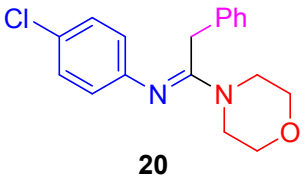

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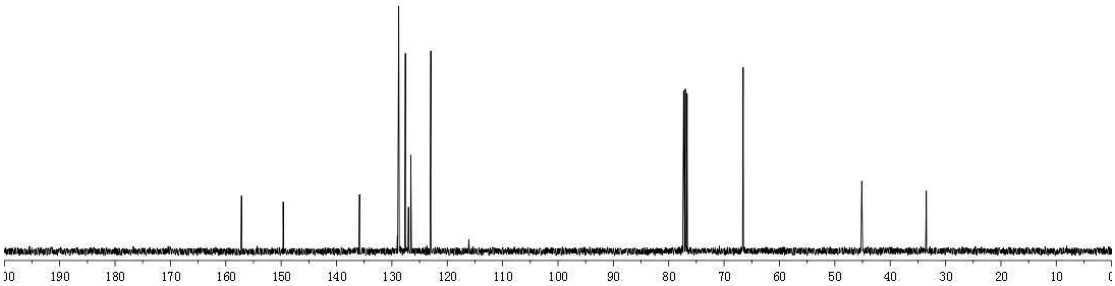
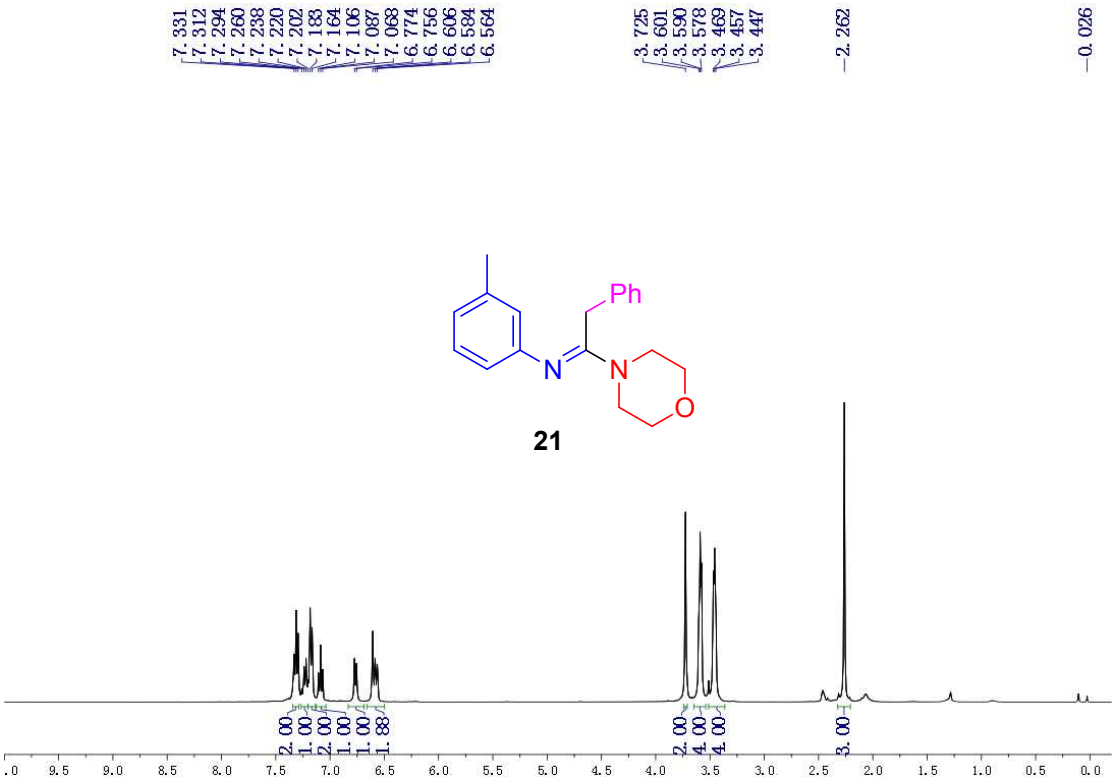
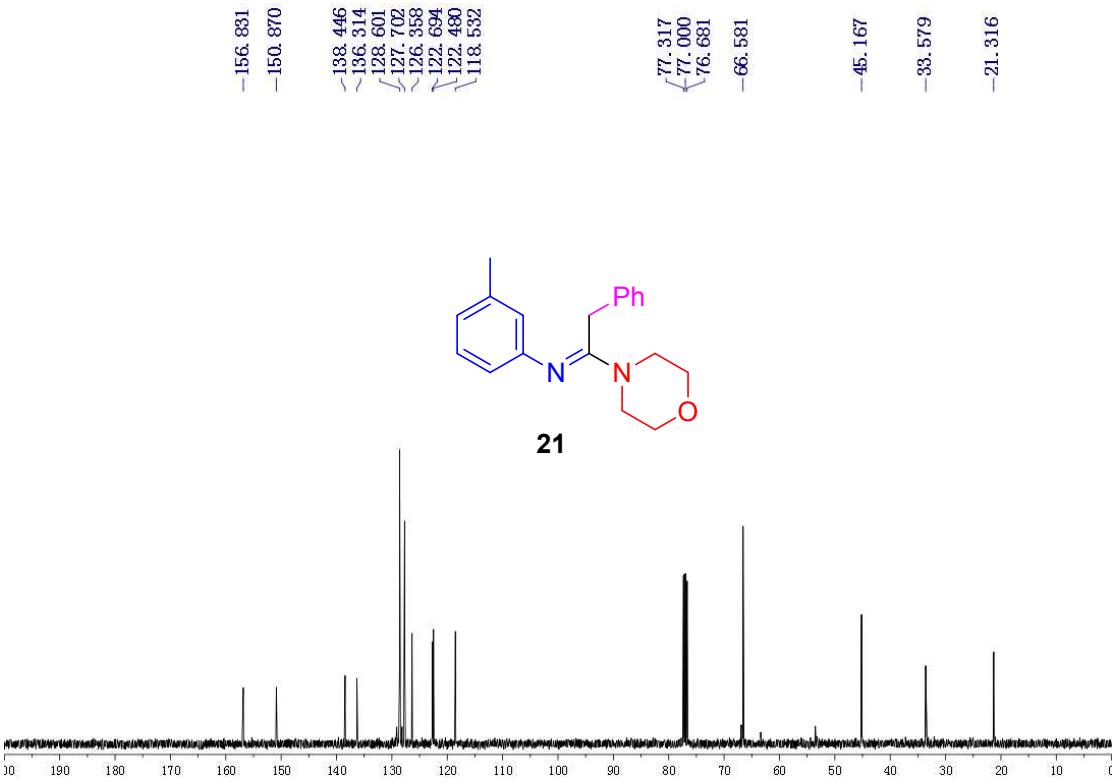
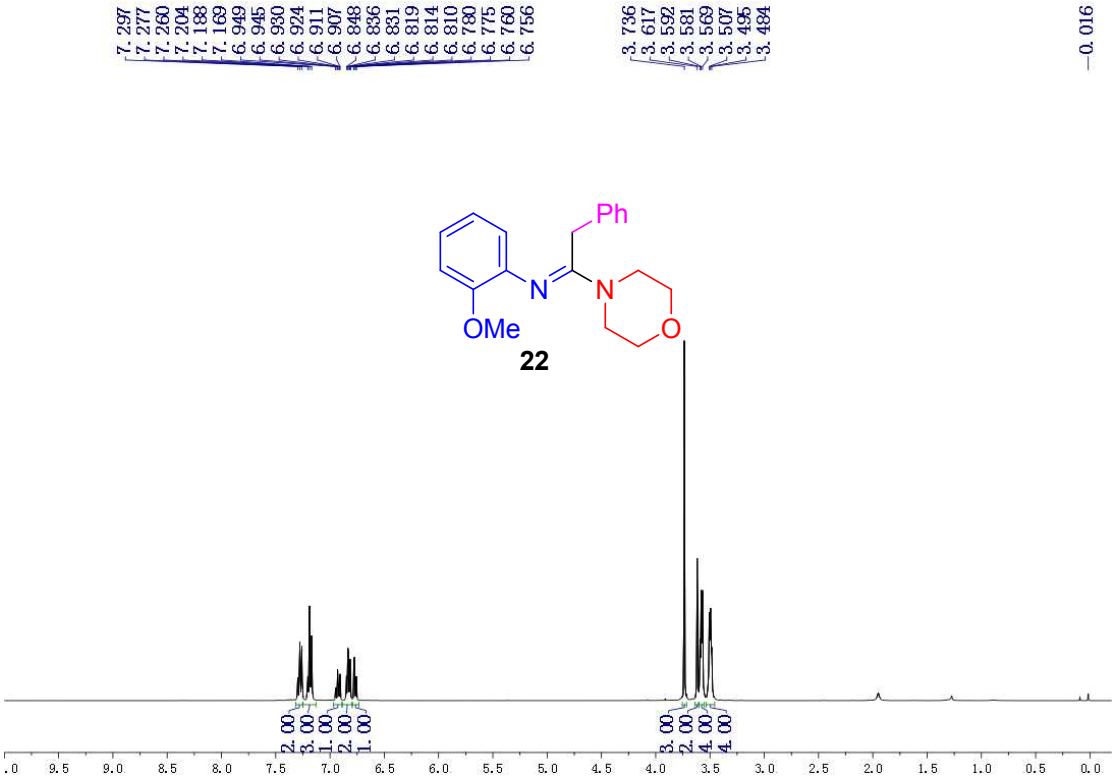


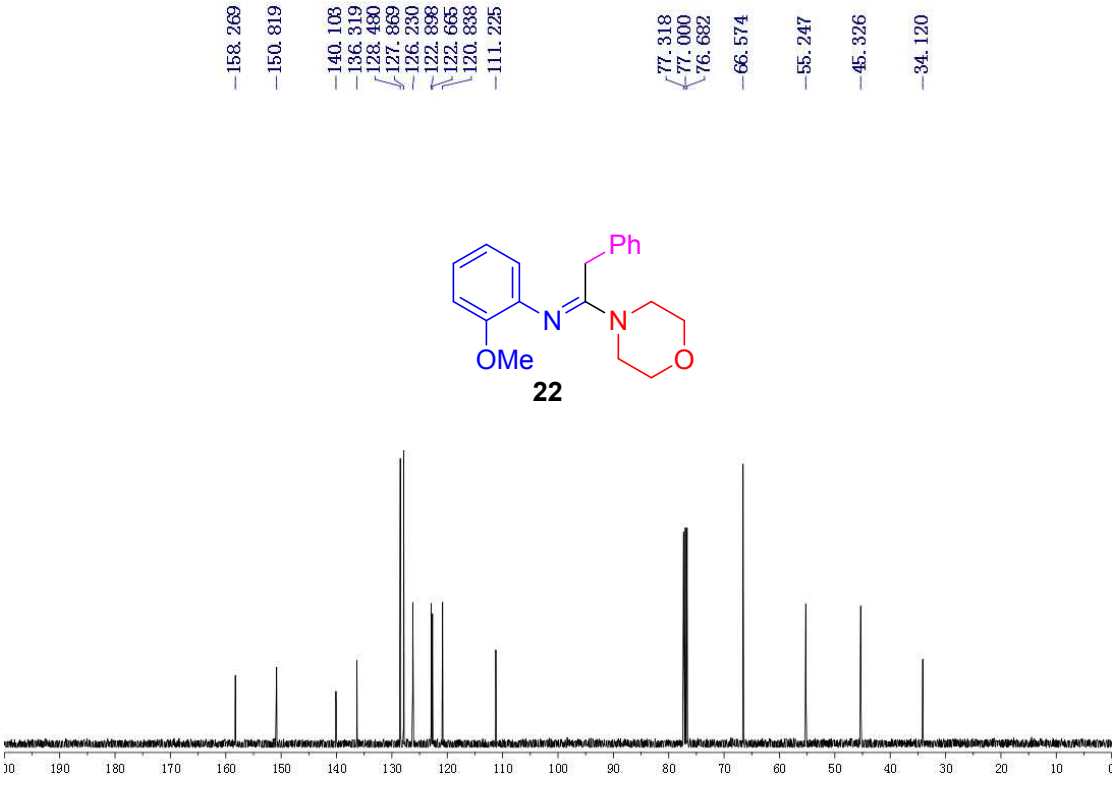
***(E)*-3-methyl-*N*-(1-morpholino-2-phenylethylidene)aniline (21)**



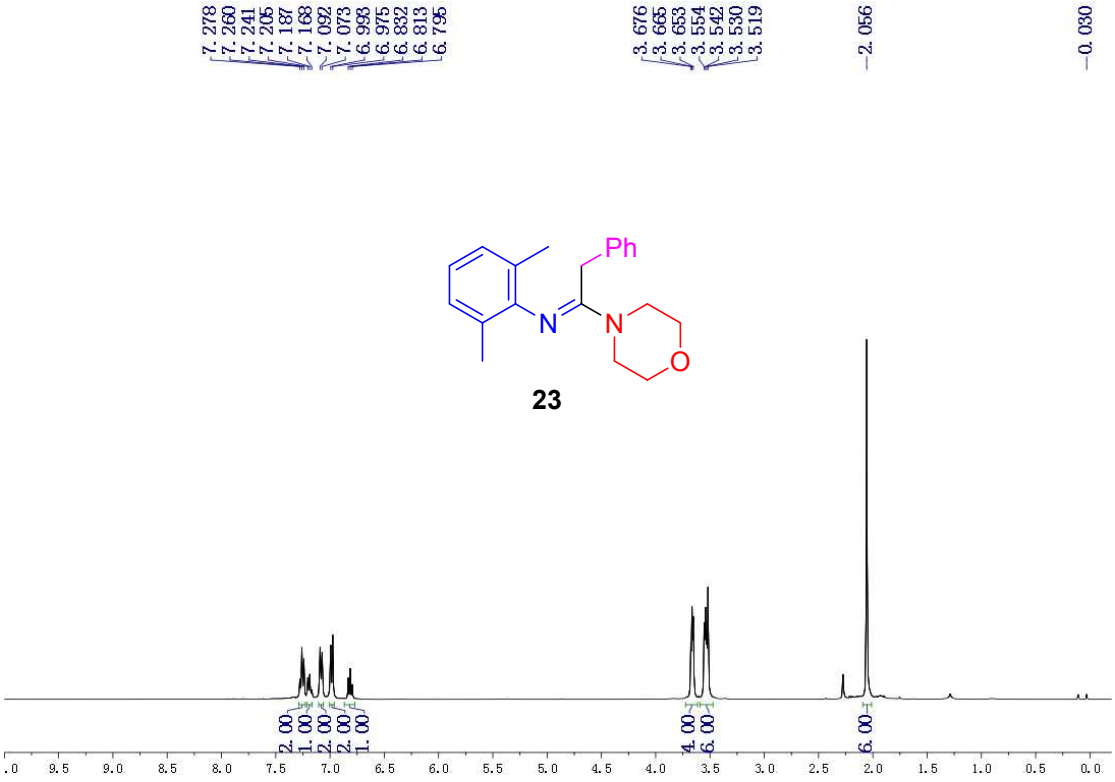


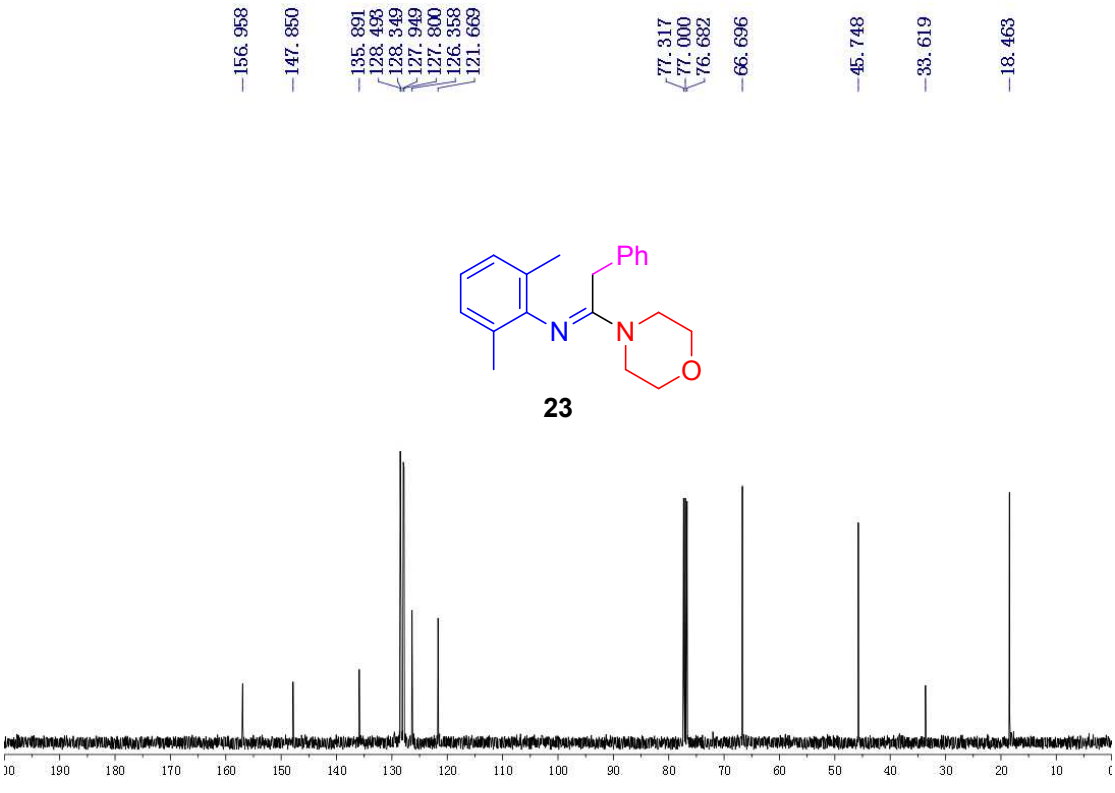
***(E)*-2-methoxy-*N*-(1-morpholino-2-phenylethylidene)aniline (22)**



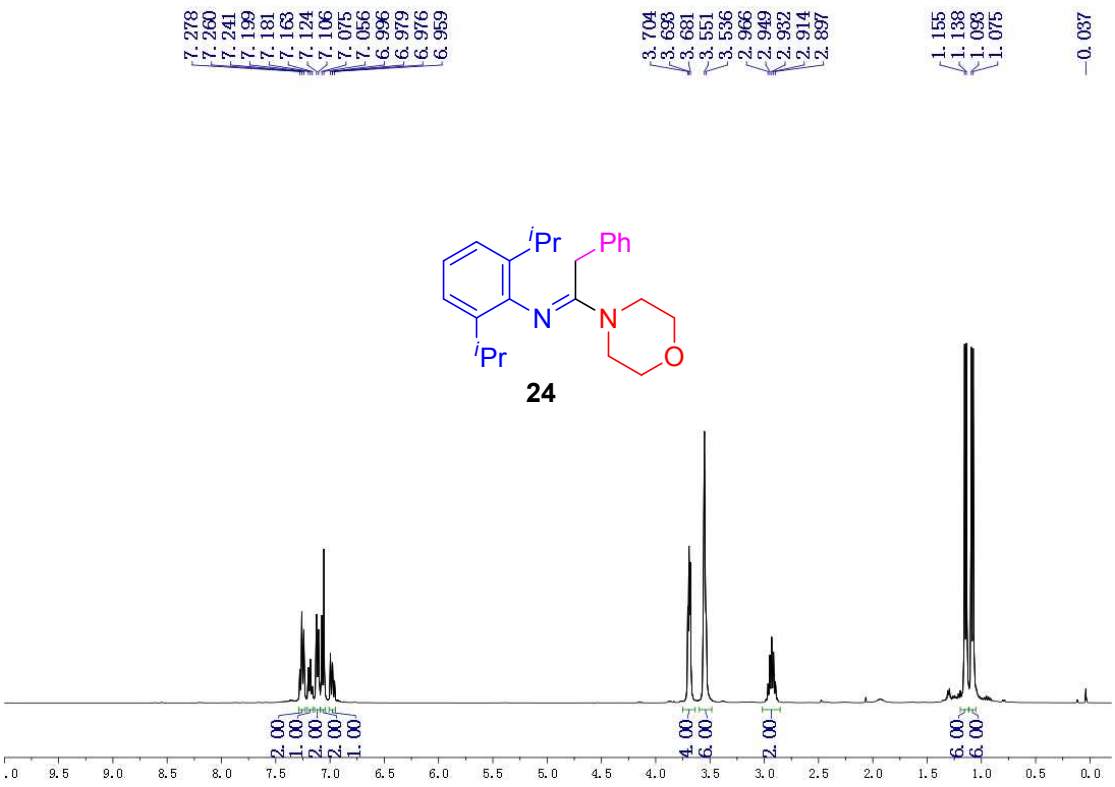


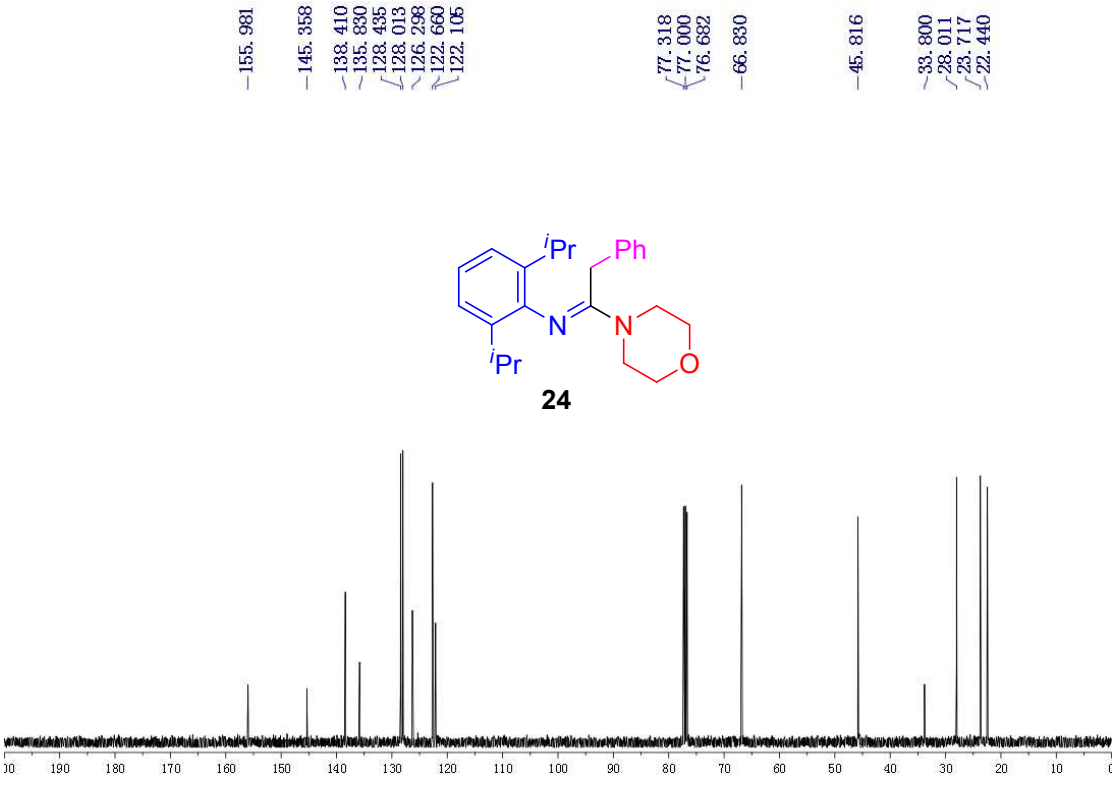
(E)-2,6-dimethyl-*N*-(1-morpholino-2-phenylethylidene)aniline (23)



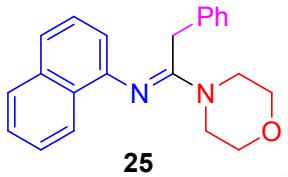


***(E)*-2,6-diisopropyl-*N*-(1-morpholino-2-phenylethylidene)aniline (24)**





(E)-N-(1-morpholino-2-phenylethylidene)naphthalen-1-amine (25)



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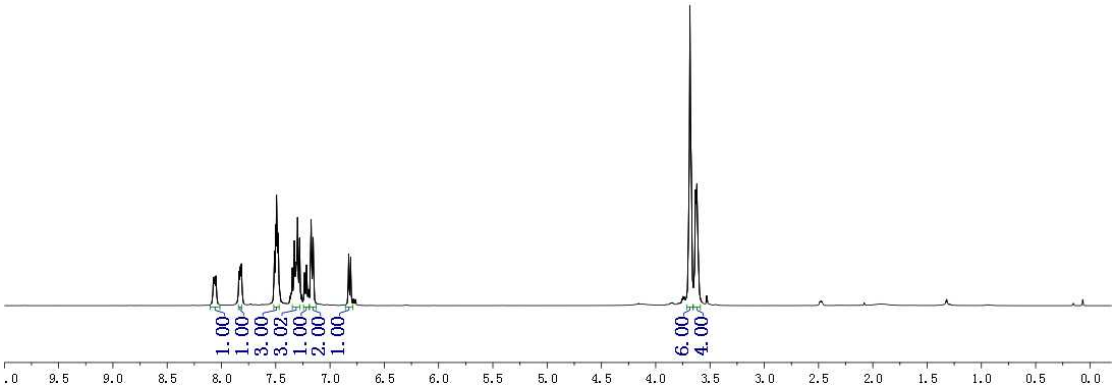

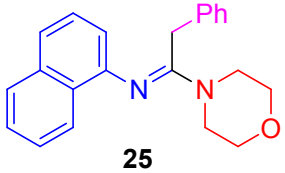

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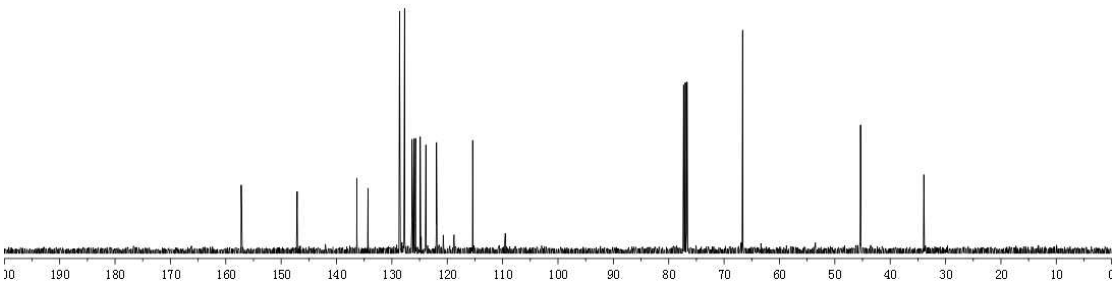
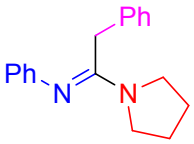


***(E)-N*-(2-phenyl-1-(pyrrolidin-1-yl)ethylidene)aniline (26)**



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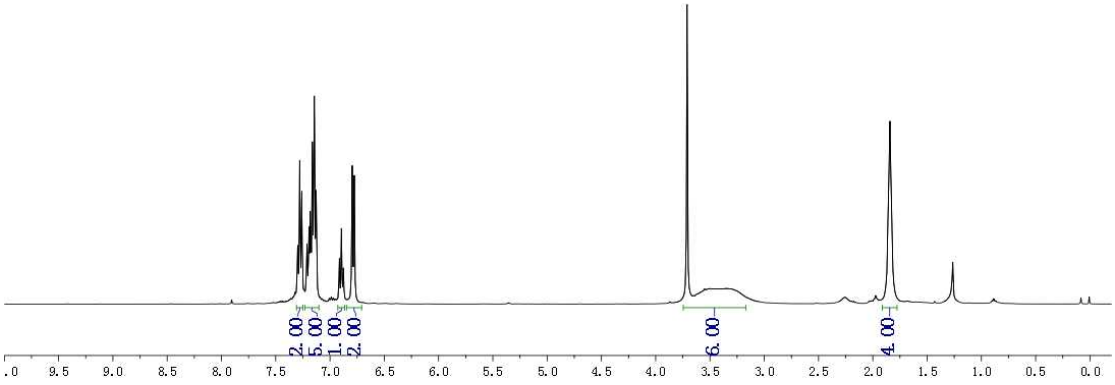

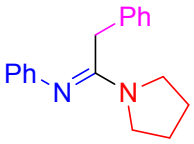

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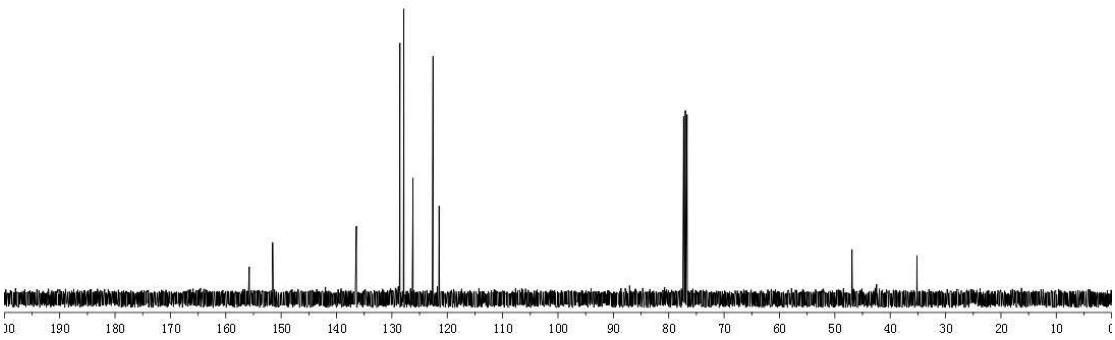
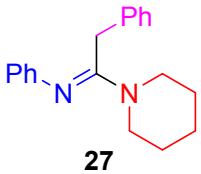


***(E)-N*-(2-phenyl-1-(piperidin-1-yl)ethylidene)aniline (27)**



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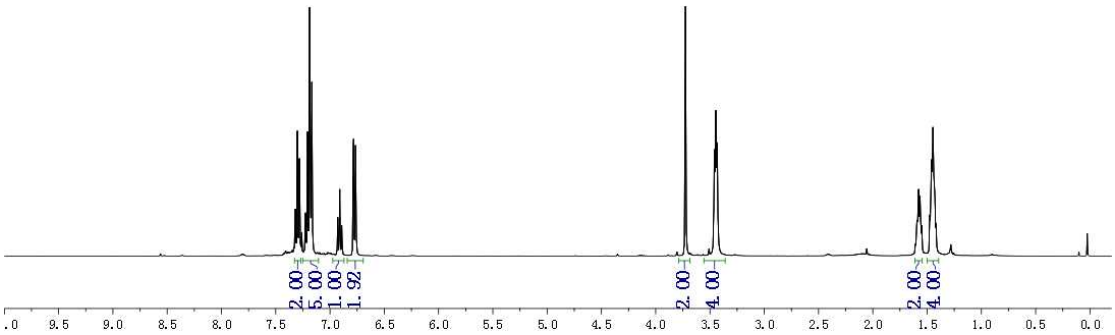

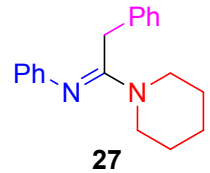

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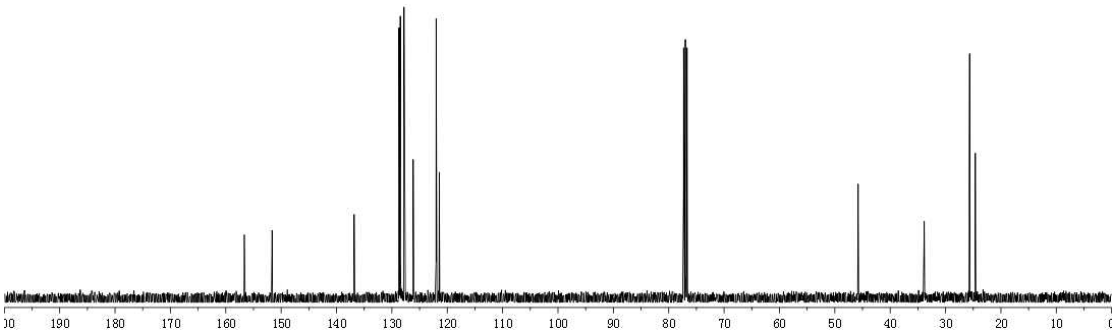
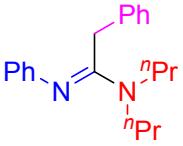


(E)-*N*',2-diphenyl-*N*,*N*-dipropylacetimidamide (28)



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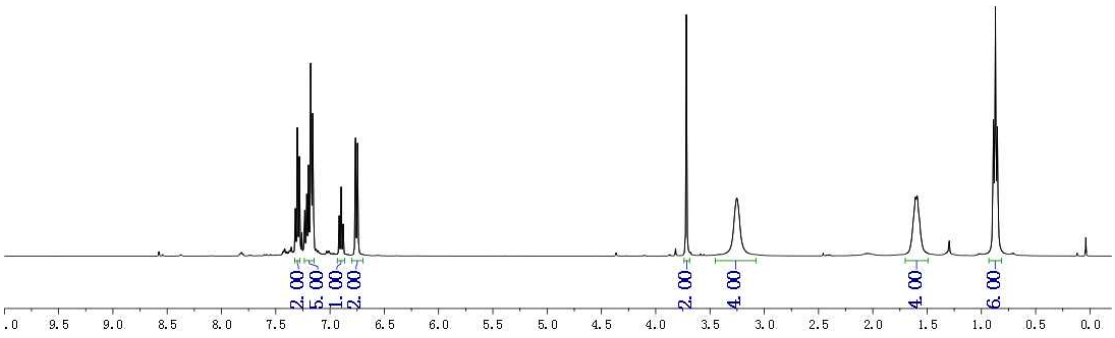

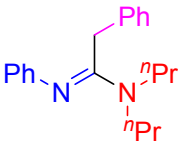

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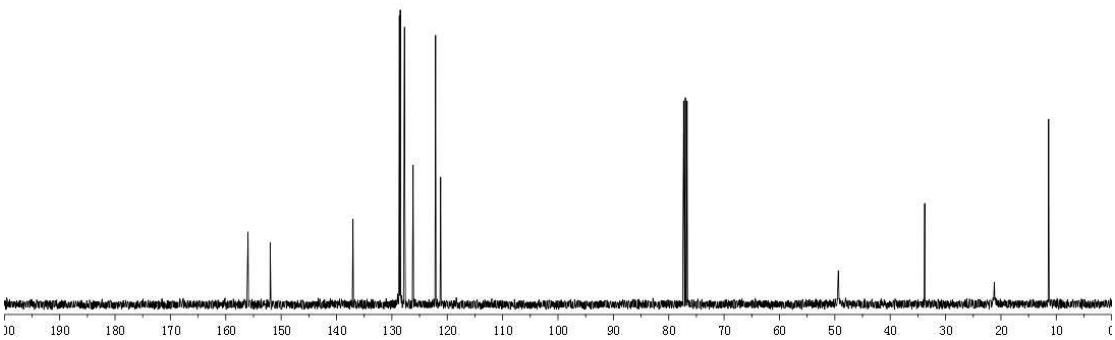
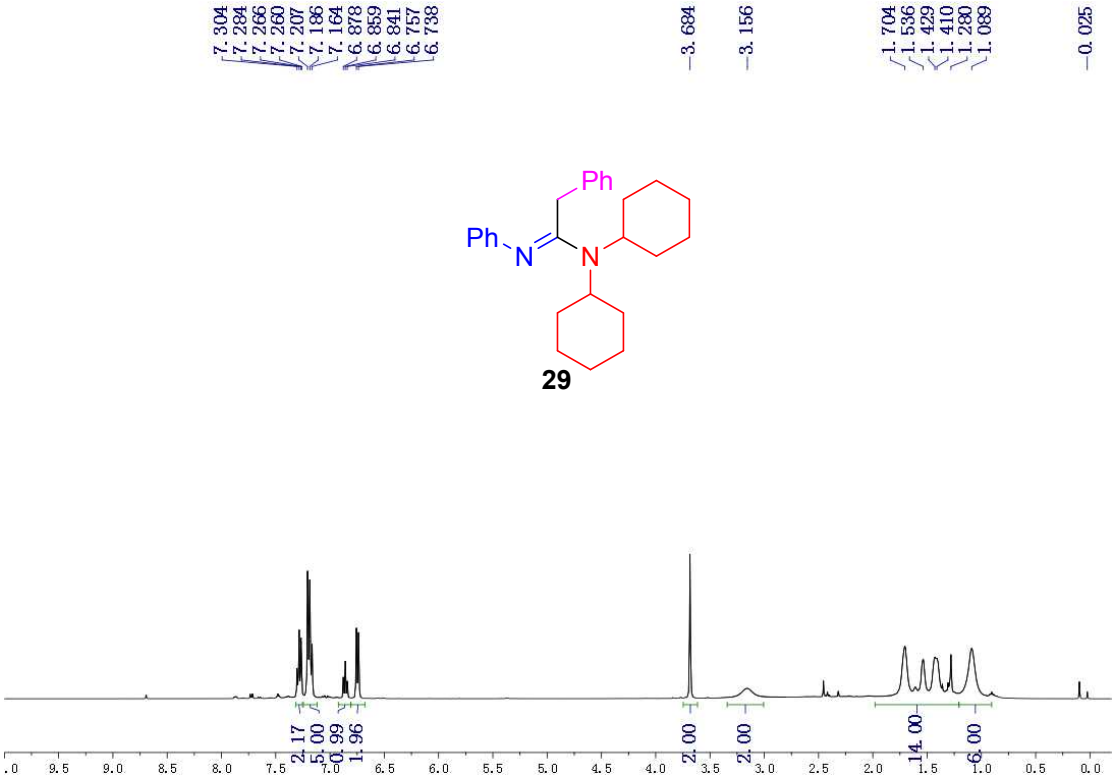
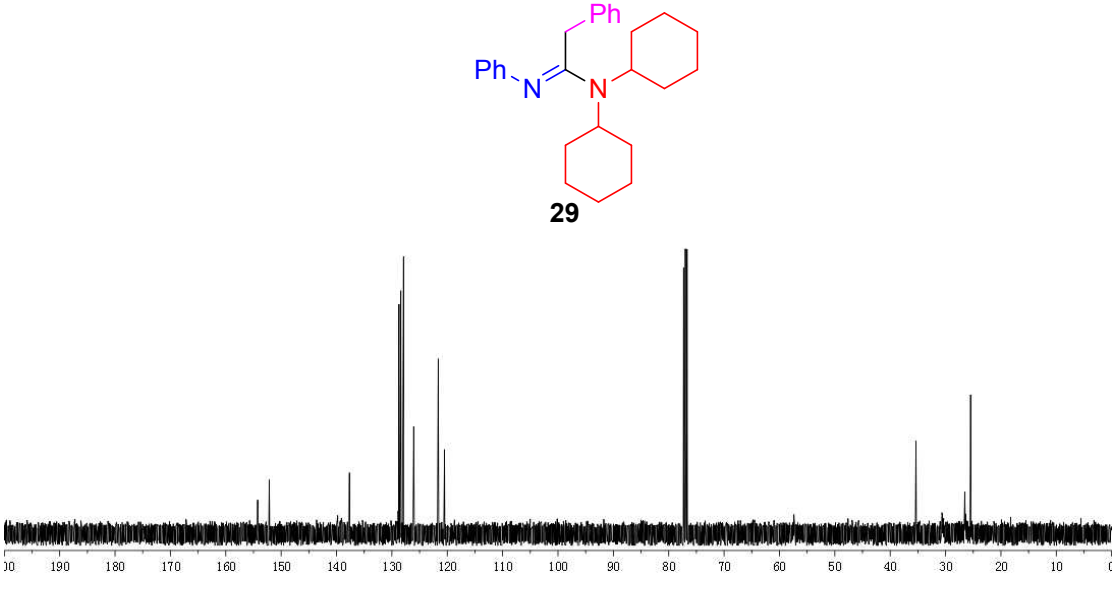
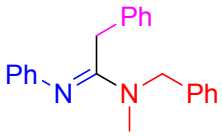


***(E)*-*N*,*N*-dicyclohexyl-*N*',2-diphenylacetimidamide (29)**

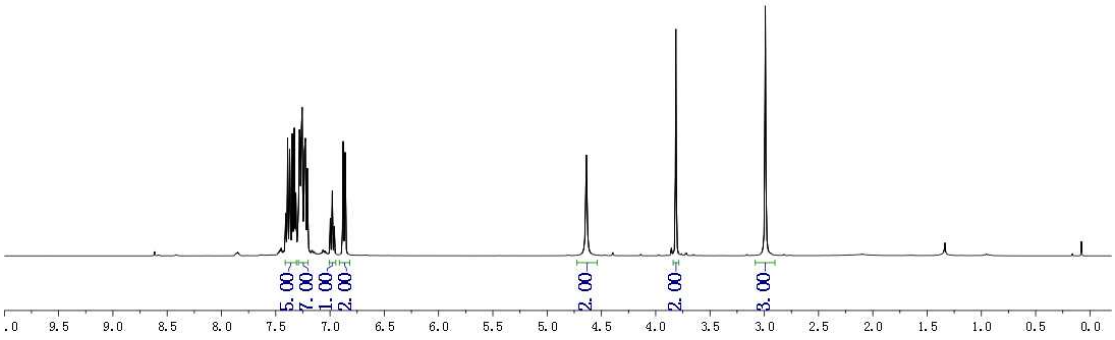


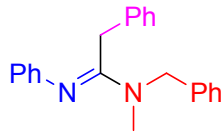


(E)-*N*-benzyl-*N*-methyl-*N*',2-diphenylacetimidamide (30)



30





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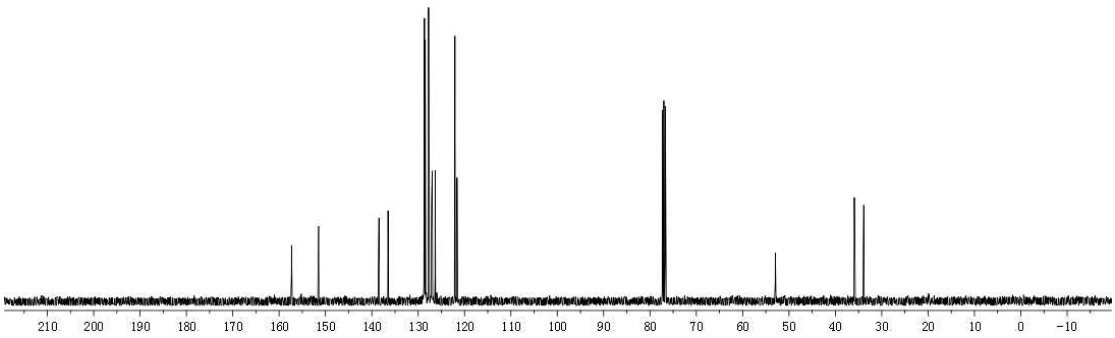
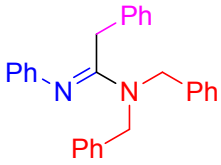


(E)-*N*,*N*-dibenzyl-*N*',2-diphenylacetimidamide (31)



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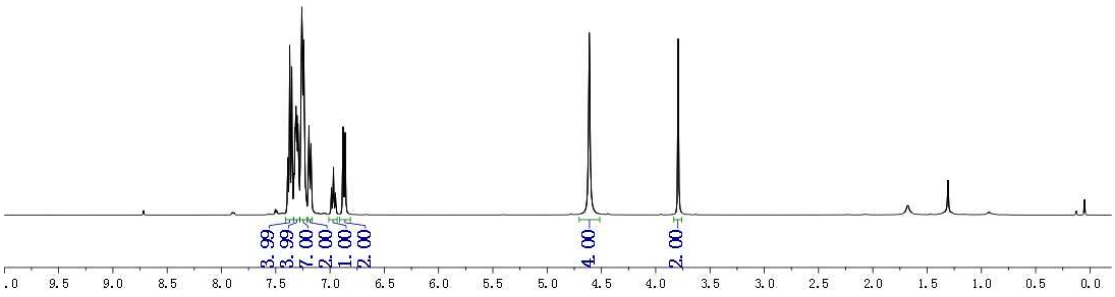

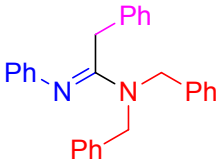

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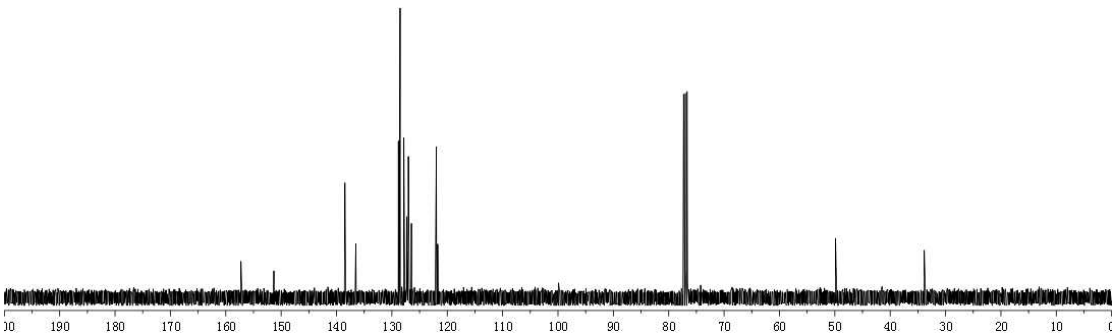
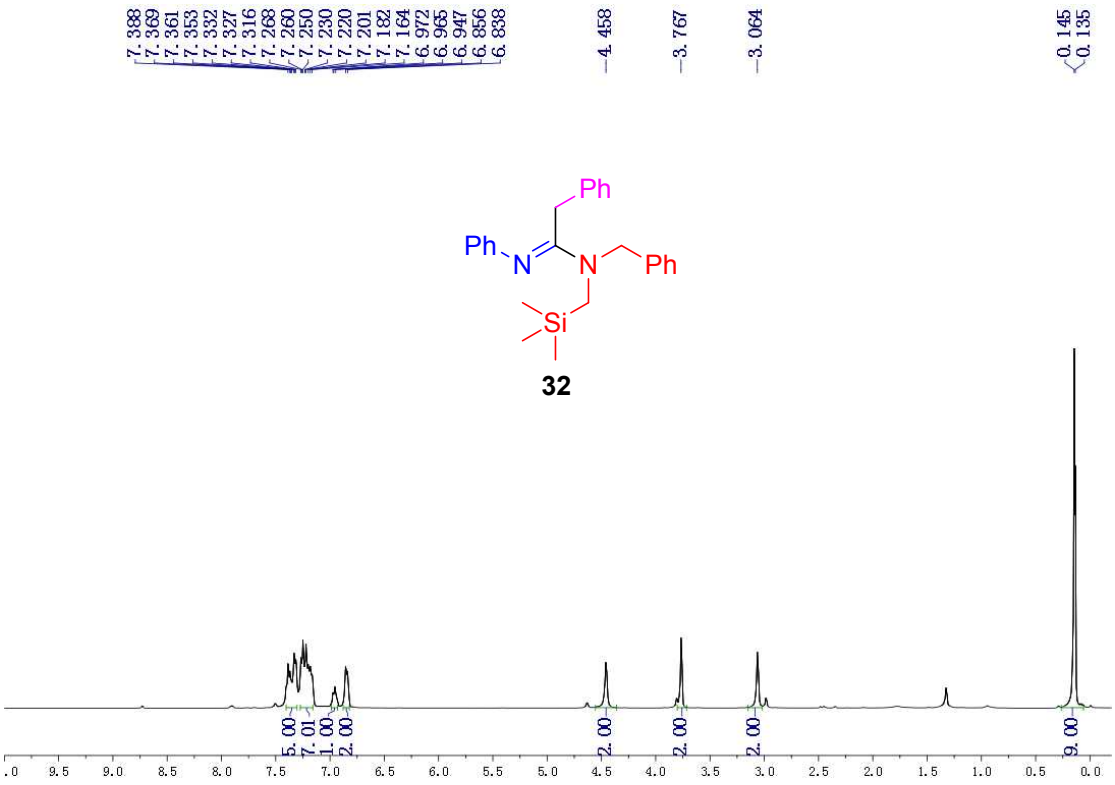


***(E)*-*N*-benzyl-*N*',2-diphenyl-*N*-((trimethylsilyl)methyl)acetimidamide (32)**



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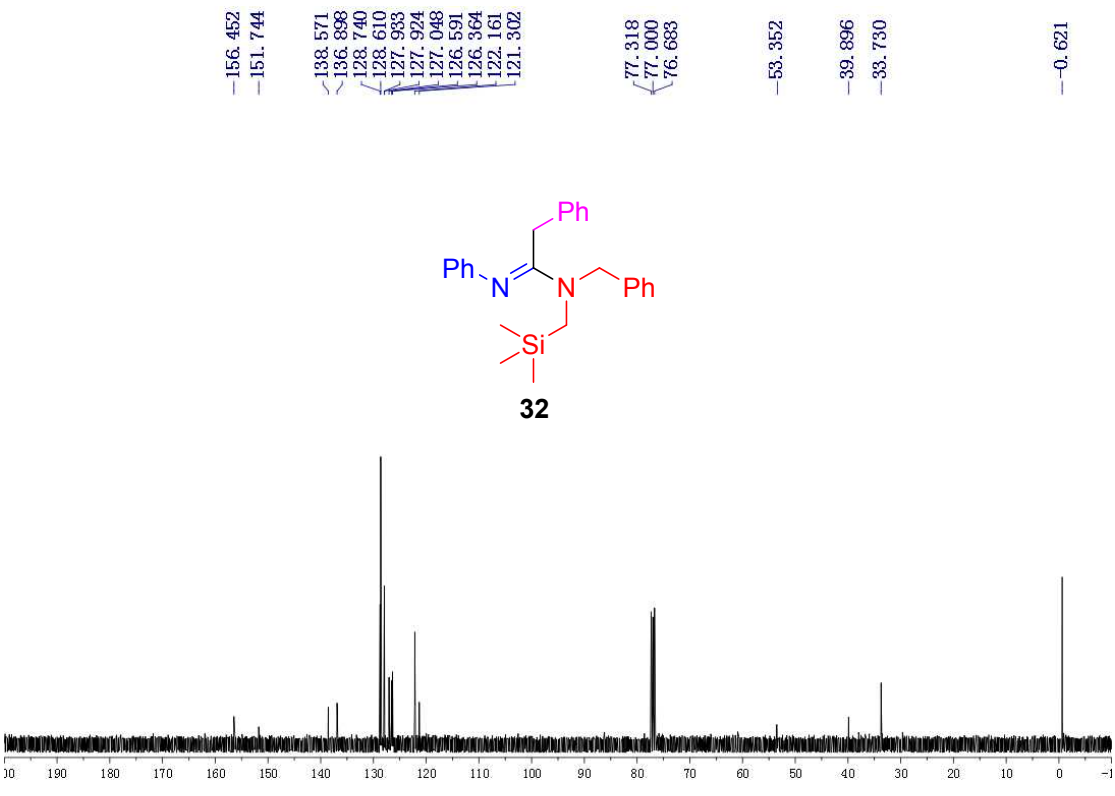
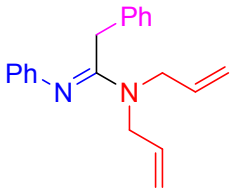


32

(E)-*N*,*N*-diallyl-*N*',2-diphenylacetimidamide (33)



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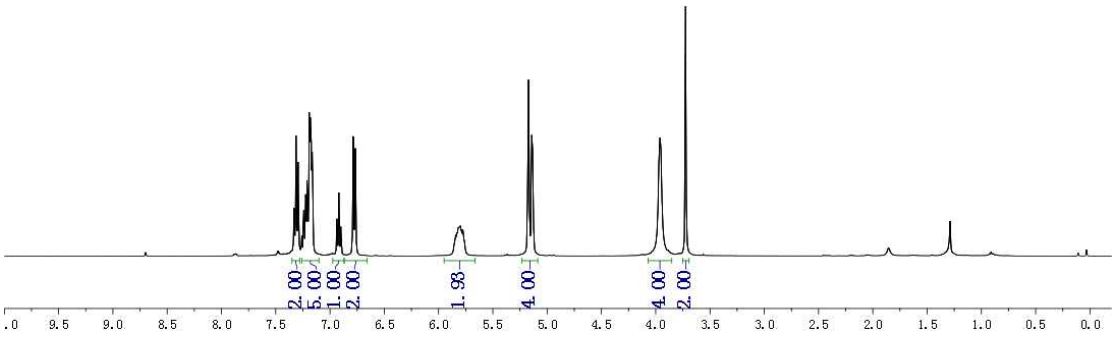

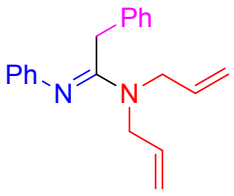

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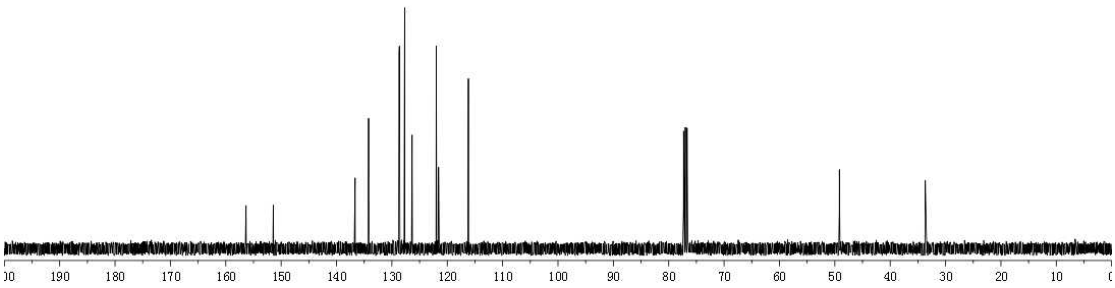
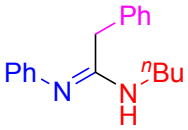


(E)-*N*-butyl-*N*',2-diphenylacetimidamide (34)



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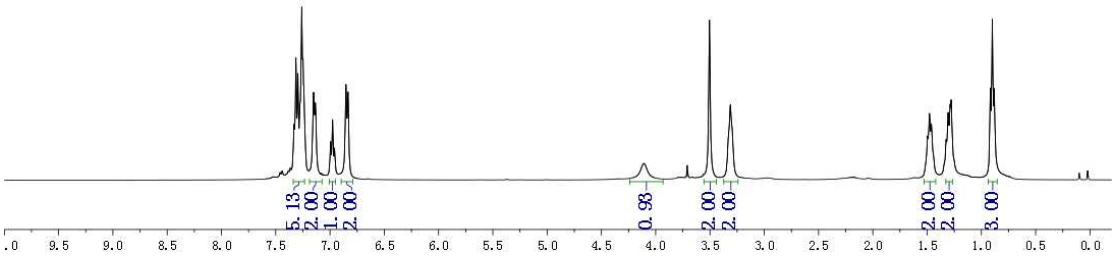

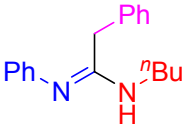

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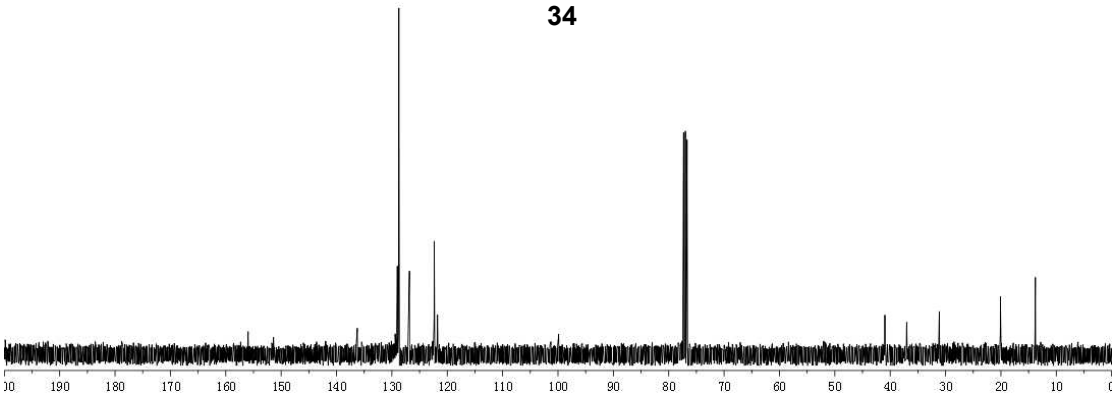
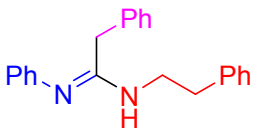


(E)-*N*-phenethyl-*N*',2-diphenylacetimidamide (35)



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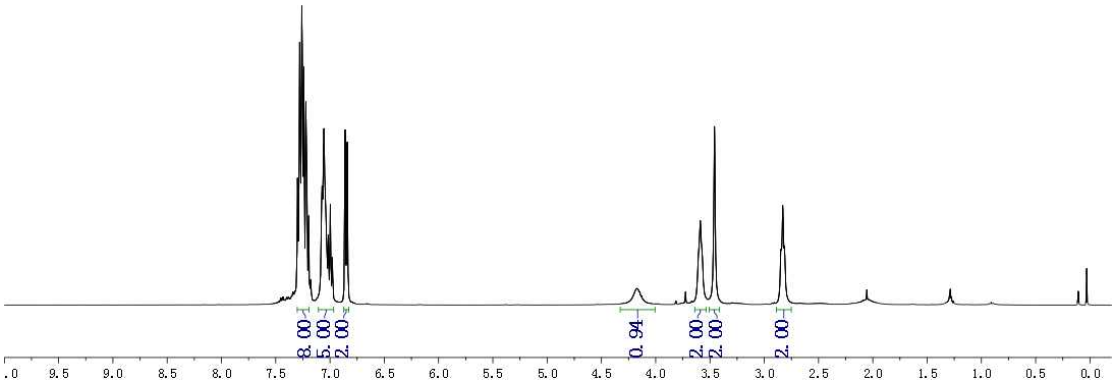

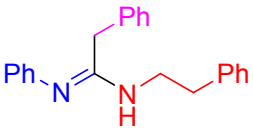

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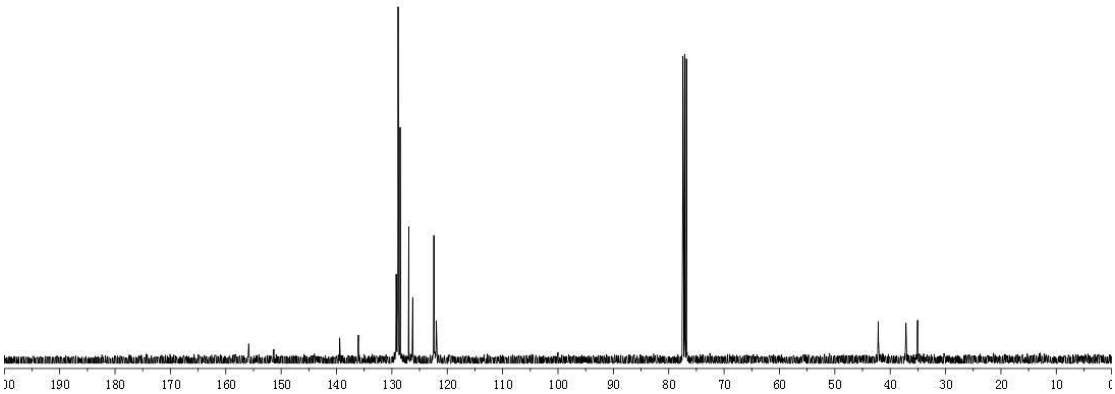
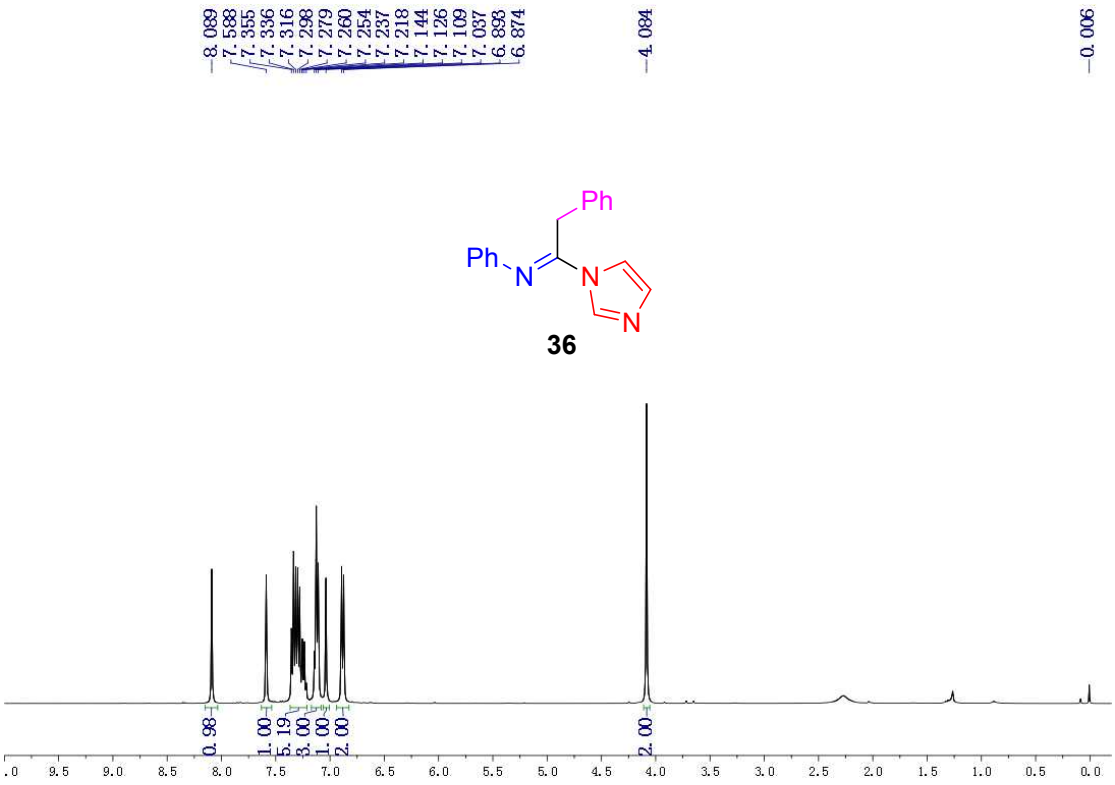
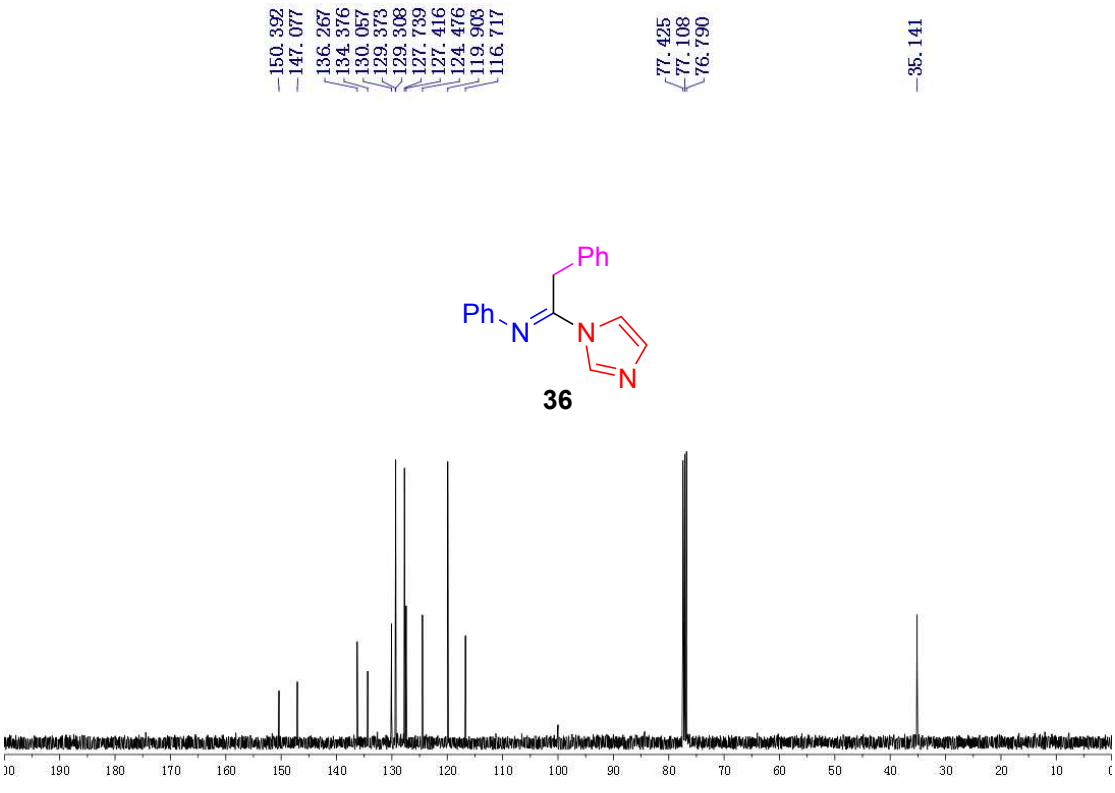
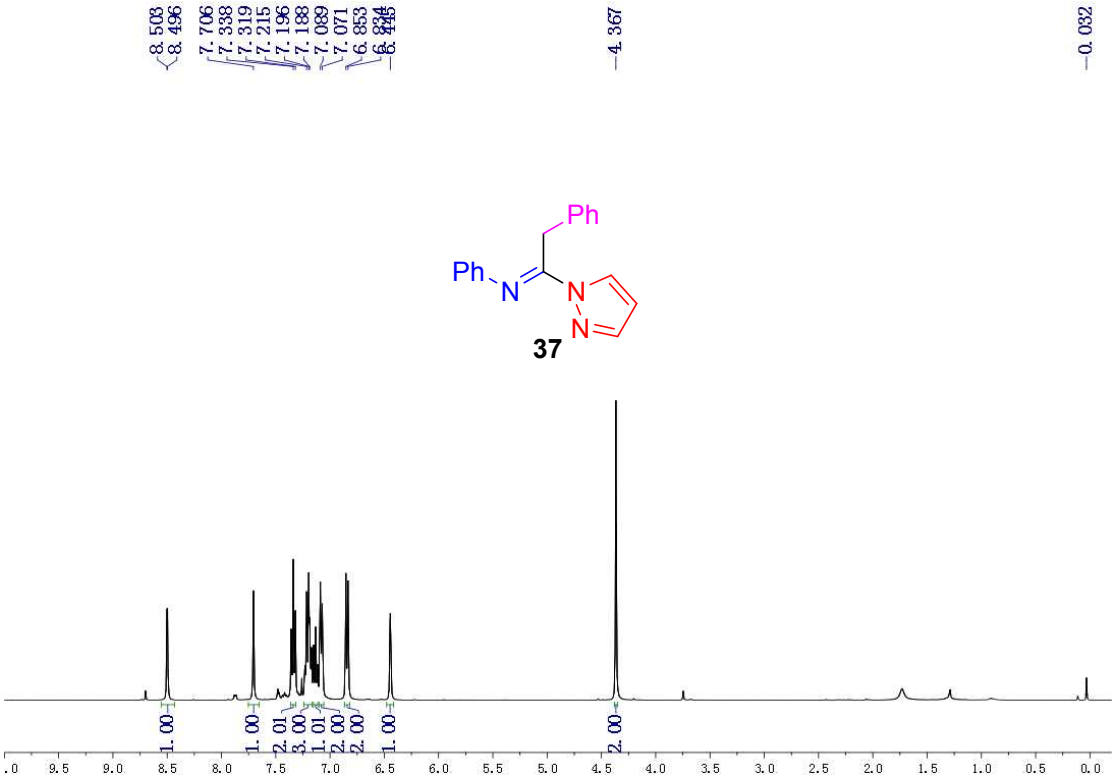


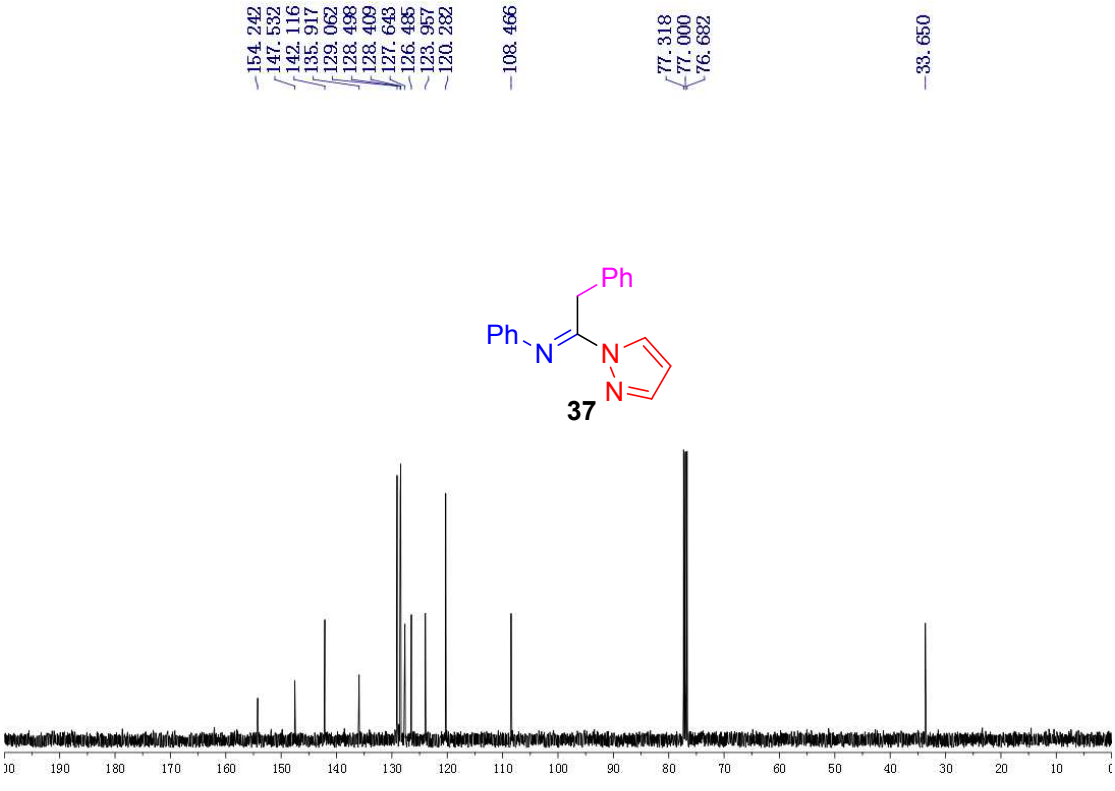
(E)-N-(1-(1*H*-imidazol-1-yl)-2-phenylethylidene)aniline (36)



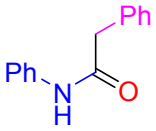


***(E)*-*N*-(2-phenyl-1-(1*H*-pyrazol-1-yl)ethylidene)aniline (37)**





N,2-diphenylacetamide (38)



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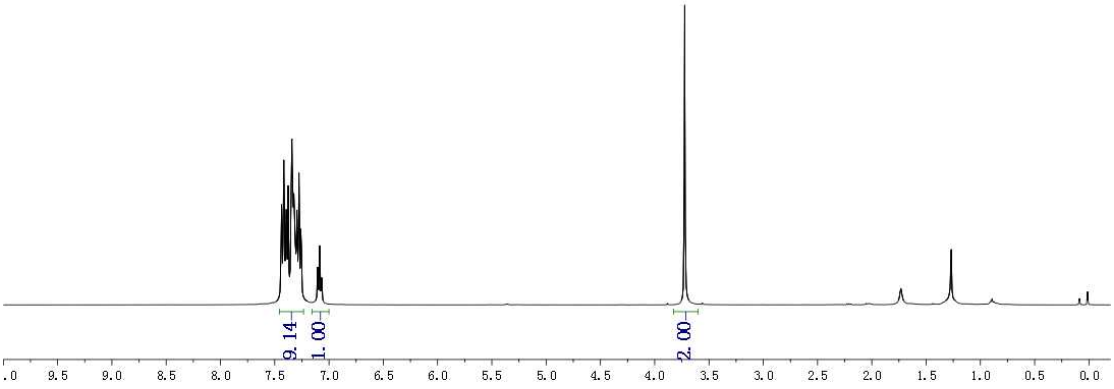

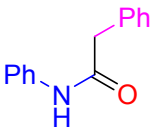

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