Supporting Information

Synthesis of a Fe-Pd bimetallic catalyst for N-alkylation of amines with alcohols *via* a hydrogen auto-transfer methodology

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1. General Information

All chemical reagents are obtained from commercial suppliers and used without further purification. GC yields were determined with hexadecane as an internal standard, and the product was then isolated by column chromatography (100–200 meshed silica gel, ethyl acetate/n-hexane = 1/20-1/100) in isolated yield. ^{1}H NMR and ^{13}C NMR spectra are recorded on an AVANCE III Bruker spectrometer operating at 500 MHz and 125 MHz in $CDCl_3$, respectively, and chemical shifts were reported in ppm relative to the center of the singlet at 7.26 ppm for $CDCl_3$. GC-MS analyses are performed on an ISQ Trace 1300 in the electron ionization (EI) mode. GC analyses are performed on an Agilent 7890A instrument (Column: Agilent 19091J413: 30 m × 320 μm × 0.25 μm, carrier gas: H_2, FID detection.

2. Table of catalytic performance of latest heterogeneous catalysts for N-alkylation of amines with alcohols *via* HAT methodology

Table S1 catalytic performance of latest heterogeneous catalysts for N-alkylation of amines with alcohols *via* HAT methodology

Catalyst	Catalyst amount	Temperature/Time	Base/Solvent	Yield	Ref.
Ir(III)@carbon black	0.01 mol% Ir	100 °C/24 h	KO^{t}Bu/toluene-d_8	42 %	1
Ru/N-C	0.2 mol% Ru	110 °C/24 h	KOH/toluene	95 %	2
Co_2Rh_2/C	5 mol% Rh	100 °C/24 h	-/-	99 %	3
$CuNiAlO_x$	15 mol% Cu	180 °C/24 h	NaOH/mesitylene	96 %	4
$(Ni_{0.5}Cu_{0.5})Fe_2O_4^{MAIN}$	10 mol% Cu	240 °C (μw)/1 h	-/TAA (iPrOH)	71 %	5
Nano-Fe_2O_3	30 mol % Fe	135 °C/24 h	KOH/toluene	89 %	6
Self-supported Cu	5 mol % Cu	160 °C/24 h	KOH/toluene	99 %	7
Ti-Pd alloy	0.2 mol % Pd	135 °C/48 h	KOH/toluene	97 %	8
Pd@SiO_2	1 mol % Pd	150 °C/30 h	-/o-xylene	97 %	9
$Fe_{10}Pd_1$/NC500	0.2 mol % Pd	120 °C/16 h	-/-	98 %	This work

3. The magnetic property of $Fe_{10}Pd_1$/NC500

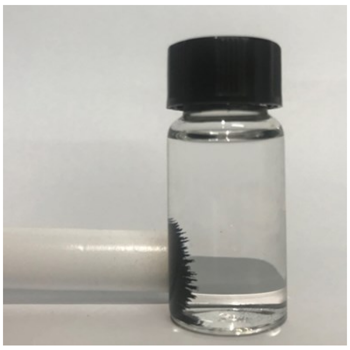


Fig. S1 The magnetic property of $Fe_{10}Pd_1$/NC500.

4. ICP-MS results of catalysts

Table S2 ICP-MS analysis of catalysts

Catalyst	Metal Content (wt %)		
	Fe	Pd	Al
$Fe_{10}Pd_1$/NC500	15.96	3.18	0
$Fe_{10}Pd_1$/NC500[a]	15.90	3.14	0
Fe/NC500	16.50	0	0
Pd/NC500	0	3.65	0.34
$Fe_{15}Pd_1$/NC500	14.98	19.47	0
Fe_5Pd_1/NC500	15.59	5.78	0
$Fe_{10}Pd_1$/AC	14.75	3.86	0
$Fe_{10}Pd_1$/NC(cellulose)	14.07	3.39	0
$Fe_{10}Pd_1$/NC(cellulose)[b]	12.50	2.48	0
$Fe_{10}Pd_1$/C500	13.11	2.89	0

[a] $Fe_{10}Pd_1$/NC500 after eight cycles.

[b] $Fe_{10}Pd_1$/NC(cellulose) after the first cycle.

5. Raman spectra and XPS survey spectra of $Fe_{10}Pd_1$/NC500

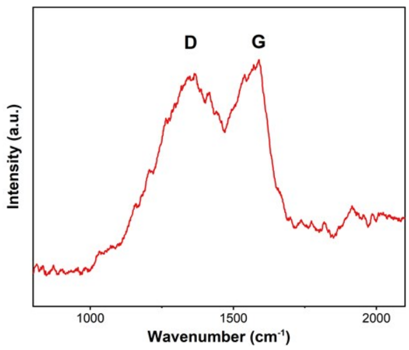


Fig. S2 Raman spectra of $Fe_{10}Pd_1$/NC500.

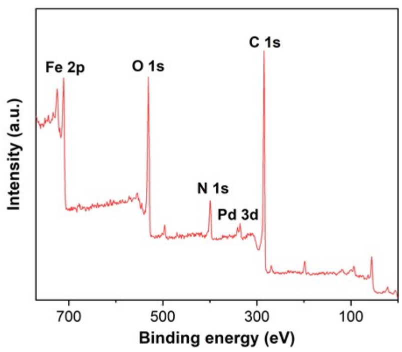


Fig. S3 XPS survey spectra of $Fe_{10}Pd_1$/NC500

6. XPS spectra of Pd/NC500 and NH_2-MIL-101($Fe_{10}Pd_1$)

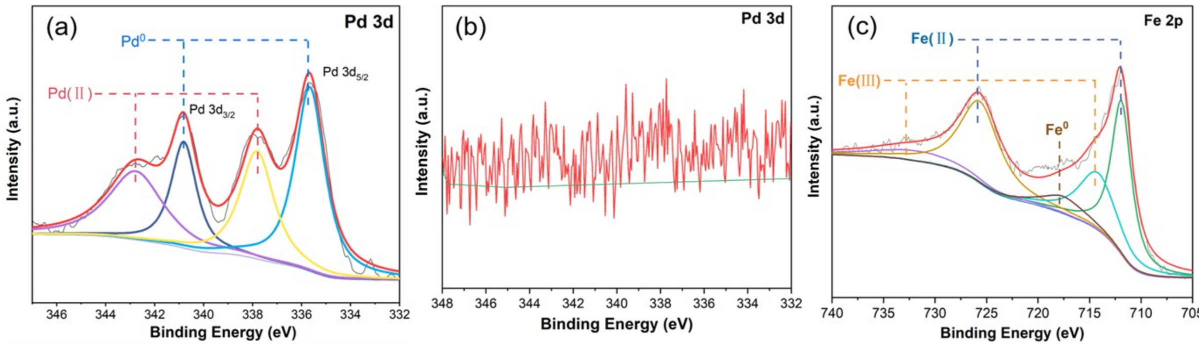


Fig. S4 Pd 3d region XPS spectra of (a) Pd/NC500, (b) NH_2-MIL-101($Fe_{10}Pd_1$), (c) Fe 2p region XPS spectra of NH_2-MIL-101($Fe_{10}Pd_1$).

7. The atomic contents of N, Fe and Pd on the surfaces of catalysts

Table S3 The atomic contents of N, Fe and Pd on the surfaces of catalysts

Catalyst	Atomic Content (%)		
	N	Fe	Pd
$Fe_{10}Pd_1$/NC400	5.42	1.87	0.23
$Fe_{10}Pd_1$/NC500	9.29	5.53	0.43
$Fe_{10}Pd_1$/NC600	5.05	5.29	0.40

8. TEM image of $Fe_{10}Pd_1$/NC500 after eight cycles

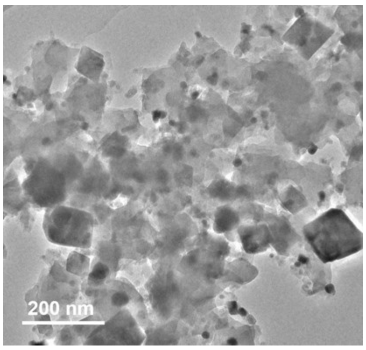
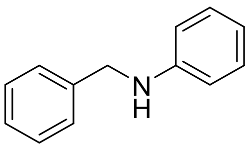


Fig. S5 TEM image of $Fe_{10}Pd_1$/NC500 after eight cycles

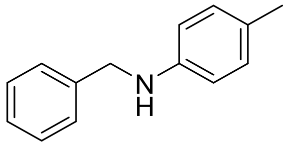
9. Characterization data

N-benzyl aniline (3a)[10]



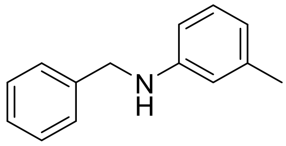
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.58 (td, J = 6.1, 4.7 Hz, 4H), 7.52 (s, 1H), 7.47 – 7.36 (m, 2H), 6.98 (d, J = 7.2 Hz, 1H), 6.84 (d, J = 7.7 Hz, 2H), 4.50 (d, J = 6.6 Hz, 2H), 4.10 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 148.46 (s), 139.81 (s), 129.59 (s), 128.94 (s), 127.80 (s), 127.51 (s), 117.84 (s), 113.19 (s), 48.52 (s).

N-benzyl-4-methylaniline (3b)[10]



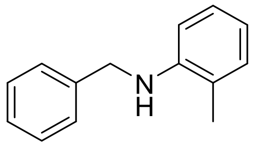
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.42 – 7.28 (m, 4H), 7.25 (s, 1H), 6.98 (d, *J* = 7.6 Hz, 2H), 6.53 (d, *J* = 7.0 Hz, 2H), 4.25 (s, 2H), 3.75 (s, 1H), 2.24 (s, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.24 (s), 140.03 (s), 130.07 (s), 128.89 (s), 127.79 (s), 127.43 (s), 126.91 (s), 113.34 (s), 48.85 (s), 20.76 (s).

N-benzyl-3-methylaniline (3c)[11]



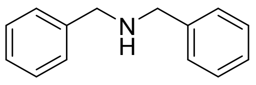
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.51 – 7.38 (m, 4H), 7.34 (d, *J* = 6.7 Hz, 1H), 7.13 (t, *J* = 7.7 Hz, 1H), 6.62 (d, *J* = 7.2 Hz, 1H), 6.58 – 6.47 (m, 2H), 4.37 (s, 2H), 3.78 (s, 1H), 2.34 (s, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 148.19 (s), 139.57 (s), 139.13 (s), 129.25 (s), 128.71 (s), 127.66 (s), 127.31 (s), 118.74 (s), 113.84 (s), 110.18 (s), 48.51 (s), 21.73 (s).

N-benzyl-2-methylaniline (3d)[8]



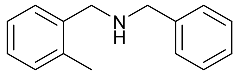
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.42 (dt, *J* = 15.0, 7.4 Hz, 4H), 7.33 (t, *J* = 7.1 Hz, 1H), 7.15 (dd, *J* = 14.8, 7.3 Hz, 2H), 6.74 (t, *J* = 7.3 Hz, 1H), 6.67 (d, *J* = 8.0 Hz, 1H), 4.42 (s, 2H), 4.00 (s, 1H), 2.22 (s, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.05 (s), 139.51 (s), 130.18 (s), 128.75 (s), 127.66 (s), 127.31 (d, *J* = 12.9 Hz), 122.09 (s), 117.37 (s), 110.18 (s), 48.44 (s), 17.66 (s).

Dibenzylamine (3e)[9]



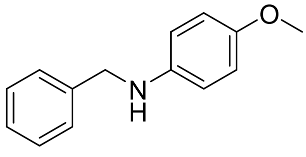
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.58 – 7.46 (m, 8H), 7.45 – 7.39 (m, 2H), 3.95 (s, 4H), 1.88 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 140.64 (s), 128.65 (s), 128.42 (s), 127.19 (s), 53.39 (s).

2-Methyldibenzylamine (3f)[12]



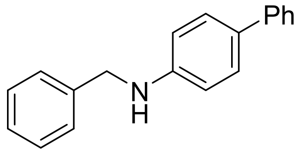
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.52 – 7.43 (m, 5H), 7.38 (dd, *J* = 10.3, 3.8 Hz, 1H), 7.32 – 7.27 (m, 3H), 3.98 (s, 2H), 3.91 (s, 2H), 2.46 (s, 3H), 1.72 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 140.56 (s), 138.36 (s), 136.56 (s), 130.40 (s), 128.53 (d, *J* = 6.6 Hz), 128.28 (s), 127.09 (d, *J* = 4.1 Hz), 126.01 (s), 53.75 (s), 51.10 (s), 19.08 (s).

N-benzyl-4-methoxyaniline (3g)[1]



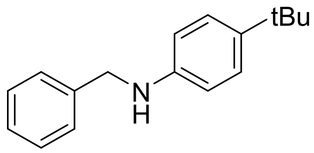
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.39 (dt, *J* = 15.0, 7.4 Hz, 4H), 7.31 (t, *J* = 7.0 Hz, 1H), 6.82 (d, *J* = 8.9 Hz, 2H), 6.64 (d, *J* = 8.9 Hz, 2H), 4.31 (s, 2H), 3.77 (s, 3H), 3.62 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 152.32 (s), 142.43 (s), 139.72 (s), 128.68 (s), 127.65 (s), 127.26 (s), 114.99 (s), 114.27 (s), 55.87 (s), 49.35 (s).

N-Benzyl-4-biphenylamine (3h)[13]



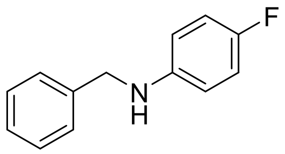
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.61 (d, J = 7.4 Hz, 2H), 7.51 (d, J = 8.5 Hz, 2H), 7.44 (dq, J = 15.5, 7.7 Hz, 6H), 7.38 – 7.29 (m, 2H), 6.76 (d, J = 8.5 Hz, 2H), 4.42 (s, 2H), 4.20 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 147.62 (s), 141.33 (s), 139.39 (s), 130.59 (s), 128.79 (d, J = 2.4 Hz), 128.06 (s), 127.61 (s), 127.41 (s), 126.41 (s), 126.20 (s), 113.27 (s), 48.43 (s).

N-Benzyl-4-*tert*-butylbenzenamine (3i)[14]



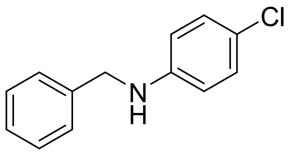
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.60 (s, 2H), 7.59 – 7.54 (m, 2H), 7.54 – 7.49 (m, 1H), 7.49 – 7.43 (m, 2H), 6.83 (d, J = 7.4 Hz, 2H), 4.51 (s, 2H), 4.01 (s, 1H), 1.56 (s, 9H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.18 (s), 140.52 (s), 140.05 (s), 128.91 (s), 127.85 (s), 127.47 (s), 126.33 (s), 112.92 (s), 48.88 (s), 34.17 (s), 31.93 (s).

N-benzyl-4-fluoroaniline (3j)[1]



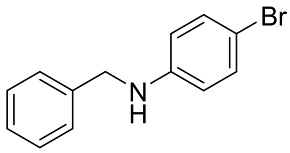
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.55 – 7.38 (m, 5H), 7.04 (t, J = 8.4 Hz, 2H), 6.67 (dd, J = 7.5, 4.6 Hz, 2H), 4.39 (s, 2H), 3.96 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 157.01 (s), 155.14 (s), 144.78 (s), 139.56 (s), 128.91 (s), 127.72 (s), 115.98 (s), 113.92 (s), 49.02 (s).

N-benzyl-4-chloroaniline (3k)[1]



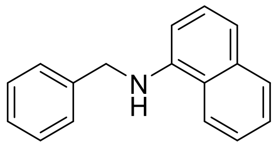
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.37 (d, J = 4.4 Hz, 4H), 7.31 (dd, J = 8.9, 4.5 Hz, 1H), 7.13 (d, J = 8.8 Hz, 2H), 6.57 (d, J = 8.8 Hz, 2H), 4.31 (s, 2H), 4.18 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.61 (s), 138.93 (s), 129.14 (s), 128.77 (s), 127.48 (d, J = 6.5 Hz), 122.25 (s), 114.07 (s), 48.45 (s).

N-benzyl-4-bromoaniline (3l)[1]

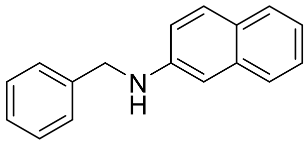


^{1}H NMR (500 MHz, $CDCl_3$) δ 7.37 (d, J = 3.0 Hz, 4H), 7.34 – 7.29 (m, 1H), 7.29 – 7.25 (m, 2H), 6.54 (d, J = 8.8 Hz, 2H), 4.32 (s, 2H), 4.19 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.99 (s), 138.84 (s), 132.00 (s), 128.77 (s), 127.47 (d, J = 3.6 Hz), 114.59 (s), 109.31 (s), 48.36 (s).

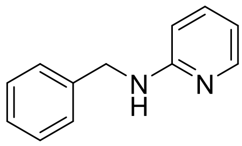
N-benzylnaphthalen-1-amine (3m)[15]


^{1}H NMR (500 MHz, $CDCl_3$) δ 8.02 (d, *J* = 8.1 Hz, 1H), 7.92 (d, *J* = 8.5 Hz, 1H), 7.66 (s, 1H), 7.61 (t, *J* = 7.2 Hz, 2H), 7.59 – 7.54 (m, 4H), 7.53 – 7.47 (m, 2H), 6.81 (d, *J* = 7.4 Hz, 1H), 4.86 (s, 1H), 4.61 (s, 2H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 143.53 (s), 139.44 (s), 134.64 (s), 129.01 (s), 128.58 (s), 128.00 (s), 127.66 (s), 126.99 (s), 126.07 (s), 125.03 (s), 123.72 (s), 120.31 (s), 117.93 (s), 105.10 (s), 48.76 (s).

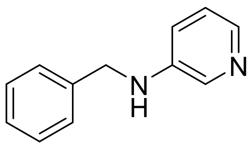
N-benzylnaphthalen-2-amine (3n)[9]


^{1}H NMR (500 MHz, $CDCl_3$) δ 7.94 (d, *J* = 8.1 Hz, 1H), 7.85 (dd, *J* = 8.3, 3.5 Hz, 2H), 7.65 – 7.57 (m, 5H), 7.54 (d, *J* = 5.7 Hz, 1H), 7.47 (t, *J* = 7.4 Hz, 1H), 7.04 (d, *J* = 8.6 Hz, 2H), 4.54 (s, 2H), 4.16 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.11 (s), 139.54 (s), 135.57 (s), 129.29 (s), 129.01 (s), 128.16 – 127.85 (m), 127.63 (s), 126.72 (s), 126.41 (s), 122.42 (s), 118.26 (s), 105.01 (s), 48.53 (s).

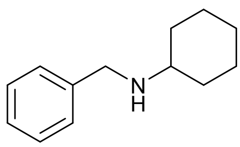
N-benzylpyridin-2-amine (3o)[15]


^{1}H NMR (500 MHz, $CDCl_3$) δ 8.09 (d, *J* = 4.9 Hz, 1H), 7.40 (s, 1H), 7.37 – 7.31 (m, 4H), 7.28 (d, *J* = 6.9 Hz, 1H), 6.64 – 6.54 (m, 1H), 6.38 (d, *J* = 8.4 Hz, 1H), 5.05 (s, 1H), 4.51 (d, *J* = 5.7 Hz, 2H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 158.60 (s), 147.99 (s), 139.15 (s), 137.67 (s), 128.69 (s), 127.37 (d, *J* = 16.3 Hz), 113.17 (s), 106.91 (s), 46.37 (s).

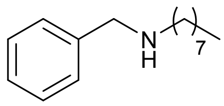
N-benzylpyridin-3-amine (3p)[16]


^{1}H NMR (500 MHz, $CDCl_3$) δ 8.05 (d, *J* = 2.9 Hz, 1H), 7.94 (d, *J* = 4.7 Hz, 1H), 7.37 – 7.26 (m, 5H), 7.05 (dd, *J* = 8.3, 4.7 Hz, 1H), 6.86 (d, *J* = 8.3 Hz, 1H), 4.32 (s, 2H), 2.67 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 144.15 (s), 138.80 (s), 138.60 (s), 136.12 (s), 128.81 (s), 127.49 (d, *J* = 10.1 Hz), 123.79 (s), 118.64 (s), 47.86 (s).

N-Cyclohexylbenzylamine (3q)[17]

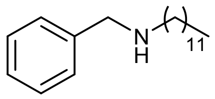

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.35 (d, *J* = 13.7 Hz, 4H), 7.28 (ddd, *J* = 11.2, 6.9, 4.4 Hz, 1H), 3.86 (s, 2H), 2.59 – 2.49 (m, 1H), 2.01 – 1.92 (m, 2H), 1.79 (dd, *J* = 9.3, 3.7 Hz, 2H), 1.67 (d, *J* = 11.4 Hz, 1H), 1.50 (s, 1H), 1.35 – 1.13 (m, 5H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 141.04 (s), 128.44 (s), 128.16 (s), 126.83 (s), 56.26 (s), 51.12 (s), 33.62 (s), 26.30 (s), 25.09 (s).

N-benzyloctylamine (3r)[18]



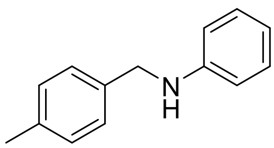
[1]H NMR (500 MHz, $CDCl_3$) δ 7.35 (d, J = 4.5 Hz, 4H), 7.30 – 7.23 (m, 1H), 3.81 (s, 2H), 2.70 – 2.63 (m, 2H), 1.83 (s, 1H), 1.59 – 1.50 (m, 2H), 1.33 (s, 10H), 0.94 (dd, J = 7.5, 6.6 Hz, 3H). **[13]C NMR** (126 MHz, $CDCl_3$) δ 140.53 (s), 128.41 (s), 128.19 (s), 126.91 (s), 54.11 (s), 49.54 (s), 31.94 (s), 30.15 (s), 29.63 (s), 29.38 (s), 27.46 (s), 22.76 (s), 14.19 (s).

N-benzyldodecylamine (3s)



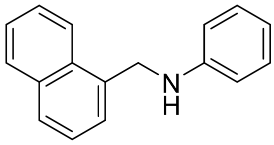
[1]H NMR (500 MHz, $CDCl_3$) δ 7.45 – 7.33 (m, 4H), 7.27 (t, J = 6.4 Hz, 1H), 3.83 (s, 2H), 2.68 (t, J = 7.2 Hz, 2H), 1.59 – 1.56 (m, 1H), 1.36 (s, 20H), 0.98 (t, J = 6.9 Hz, 3H). **[13]C NMR** (126 MHz, $CDCl_3$) δ 140.68 (s), 128.32 (s), 128.08 (s), 126.81 (s), 54.21 (s), 49.63 (s), 32.06 (s), 30.27 (s), 29.97 – 29.60 (m), 29.51 (s), 27.50 (s), 22.82 (s), 14.20 (s).

N-(4-methylbenzyl)aniline (3t)[10]



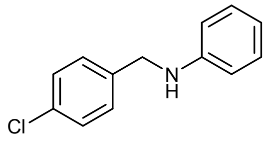
[1]H NMR (500 MHz, $CDCl_3$) δ 7.40 (d, J = 7.7 Hz, 2H), 7.31 (t, J = 7.4 Hz, 4H), 6.87 (td, J = 7.3, 1.0 Hz, 1H), 6.79 – 6.74 (m, 2H), 4.40 (s, 2H), 4.03 (s, 1H), 2.50 (s, 3H). **[13]C NMR** (126 MHz, $CDCl_3$) δ 148.44 (s), 137.02 (s), 136.62 (s), 129.50 (d, J = 7.3 Hz), 127.73 (s), 117.69 (s), 113.08 (s), 48.25 (s), 21.33 (s).

N-(naphthalen-1-ylmethyl)aniline (3u)[16]



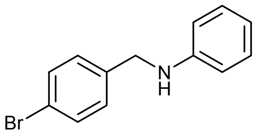
[1]H NMR (500 MHz, $CDCl_3$) δ 8.17 (d, J = 4.0 Hz, 1H), 8.01 (dd, J = 3.9, 2.4 Hz, 1H), 7.92 (d, J = 8.0 Hz, 1H), 7.71 – 7.59 (m, 3H), 7.57 – 7.49 (m, 1H), 7.34 (dd, J = 1.9, 0.9 Hz, 2H), 6.89 (dd, J = 2.6, 1.3 Hz, 1H), 6.77 (d, J = 8.3 Hz, 2H), 4.79 (s, 2H), 4.04 (s, 1H). **[13]C NMR** (126 MHz, $CDCl_3$) δ 148.25 (s), 146.83 (s), 144.93 (s), 131.32 (s), 129.35 (s), 120.59 (s), 117.78 (s), 114.52 (s), 113.10 (s), 110.39 (s), 58.52 (s), 55.99 (s), 48.50 (s), 18.44 (s).

N-(4-chlorobenzyl)benzenamine (3v)[19]



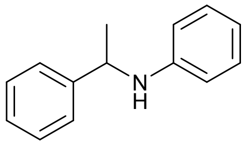
[1]H NMR (500 MHz, $CDCl_3$) δ 7.46 – 7.36 (m, 4H), 7.35 – 7.26 (m, 2H), 6.89 (t, J = 7.3 Hz, 1H), 6.73 (d, J = 8.4 Hz, 2H), 4.38 (s, 2H), 4.12 (s, 1H). **[13]C NMR** (126 MHz, $CDCl_3$) δ 148.08 (s), 138.29 (s), 132.98 (s), 129.55 (s), 128.93 (d, J = 2.9 Hz), 117.98 (s), 113.12 (s), 47.70 (s).

N-(4-bromobenzyl)benzenamine (3w)[19]



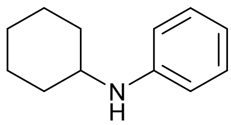
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.43 (d, *J* = 8.4 Hz, 2H), 7.21 (d, *J* = 8.3 Hz, 2H), 7.16 (dd, *J* = 8.4, 7.5 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.58 (d, *J* = 7.7 Hz, 2H), 4.25 (s, 2H), 4.04 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 147.87 (s), 138.64 (s), 131.79 (s), 129.41 (s), 129.16 (s), 121.01 (s), 117.93 (s), 113.01 (s), 47.73 (s).

N-(1-phenylethyl)aniline (3x)[3]



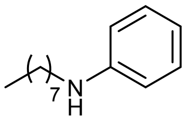
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 (d, *J* = 7.8 Hz, 2H), 7.25 – 7.21 (m, 1H), 7.20 – 7.11 (m, 2H), 7.08 – 6.97 (m, 2H), 6.59 (t, *J* = 7.3 Hz, 1H), 6.46 (d, *J* = 8.0 Hz, 2H), 4.42 (d, *J* = 6.7 Hz, 1H), 4.11 (s, 1H), 1.46 (d, *J* = 6.7 Hz, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 146.95 (s), 145.01 (s), 129.15 (s), 128.69 (s), 126.98 (s), 125.96 (s), 117.59 (s), 113.64 (s), 53.80 (s), 24.91 (s).

N-cyclohexylaniline (3y)[20]



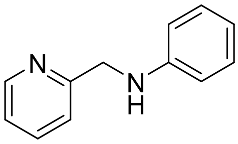
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.22 (t, *J* = 7.8 Hz, 2H), 6.72 (t, *J* = 7.3 Hz, 1H), 6.65 (d, *J* = 8.2 Hz, 2H), 3.61 (s, 1H), 3.31 (t, *J* = 3.7 Hz, 1H), 2.12 (dd, *J* = 13.3, 3.1 Hz, 2H), 1.82 (dd, *J* = 10.5, 6.7 Hz, 2H), 1.75 – 1.67 (m, 1H), 1.43 (d, *J* = 12.9 Hz, 2H), 1.30 (s, 1H), 1.25 – 1.13 (m, 2H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 147.42 (s), 129.36 (s), 117.00 (s), 113.32 (s), 51.84 (s), 33.56 (s), 26.06 (s), 25.15 (s).

N-octylaniline (3z)[8]



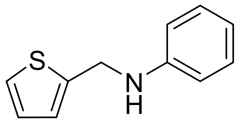
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 – 7.24 (m, 2H), 6.81 (t, *J* = 7.3 Hz, 1H), 6.70 (d, *J* = 8.1 Hz, 2H), 3.67 (s, 1H), 3.19 (t, *J* = 7.2 Hz, 2H), 1.75 – 1.64 (m, 2H), 1.45 (dd, *J* = 23.5, 20.7 Hz, 11H), 1.03 (t, *J* = 6.7 Hz, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 148.71 (s), 129.36 (s), 117.17 (s), 112.82 (s), 44.14 (s), 32.06 (s), 29.86 – 29.41 (m), 27.40 (s), 22.89 (s), 14.32 (s).

N-((pyridin-2-yl)methyl)aniline (3aa)[19]



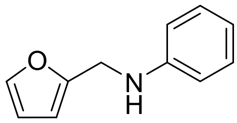
^{1}H NMR (500 MHz, $CDCl_3$) δ 8.59 (d, *J* = 4.8 Hz, 1H), 7.65 (td, *J* = 7.7, 1.7 Hz, 1H), 7.35 (d, *J* = 7.8 Hz, 1H), 7.19 (dd, *J* = 8.4, 7.4 Hz, 3H), 6.76 – 6.64 (m, 3H), 4.47 (s, 2H), 4.09 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 158.56 (s), 149.15 (s), 147.91 (s), 136.81 (s), 129.31 (s), 122.19 (s), 121.70 (s), 117.67 (s), 113.11 (s), 49.29 (s).

N-((thiophen-2-yl)methyl)aniline (3bb)[21]



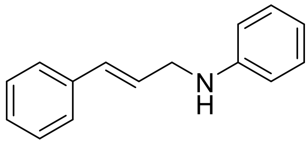
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.24 – 7.19 (m, 3H), 7.07 – 7.02 (m, 1H), 6.99 (dd, *J* = 5.0, 3.6 Hz, 1H), 6.78 (t, *J* = 7.3 Hz, 1H), 6.73 – 6.69 (m, 2H), 4.53 (s, 2H), 4.13 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 147.54 (s), 142.88 (s), 129.36 (s), 126.93 (s), 125.17 (s), 124.69 (s), 118.27 (s), 113.34 (s), 43.61 (s).

N-((furan-2-yl)methyl)aniline (3cc)[21]



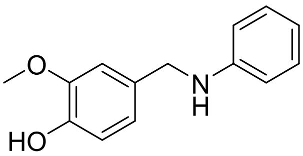
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.54 – 7.49 (m, 1H), 7.37 (dd, *J* = 11.9, 4.0 Hz, 2H), 6.93 (td, *J* = 7.3, 0.8 Hz, 1H), 6.84 – 6.78 (m, 2H), 6.48 (s, 1H), 6.38 (d, *J* = 3.2 Hz, 1H), 4.43 (s, 2H), 4.08 (s, 1H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 153.11 (s), 147.92 (s), 142.10 (s), 129.48 (s), 118.19 (s), 113.39 (s), 110.63 (s), 107.22 (s), 41.58 (s).

N-(cinnamyl)aniline (3dd)[21]



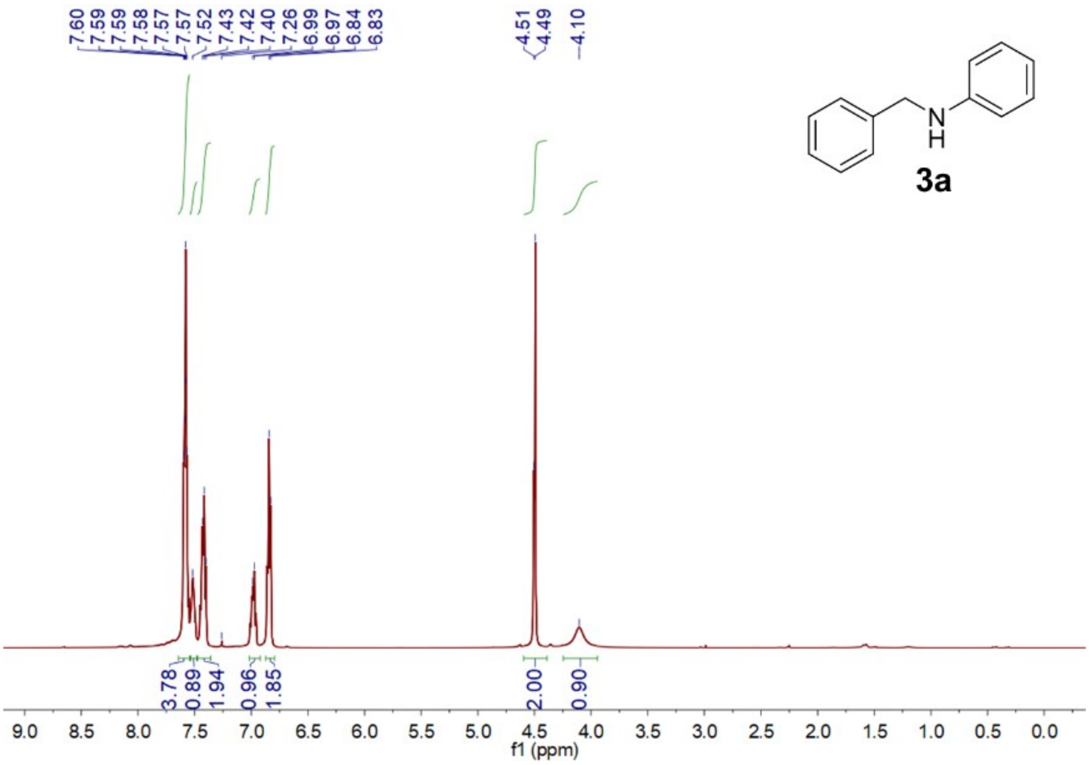
^{1}H NMR (500 MHz, $CDCl_3$) δ 7.37 (d, *J* = 7.5 Hz, 3H), 7.31 (t, *J* = 7.6 Hz, 2H), 7.25 – 7.19 (m, 3H), 6.74 (t, *J* = 7.2 Hz, 3H), 6.63 (d, *J* = 15.9 Hz, 1H), 6.34 (dt, *J* = 15.9, 5.9 Hz, 1H), 3.95 (d, *J* = 5.9 Hz, 2H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 147.94 (s), 136.89 (s), 131.66 (s), 129.33 (s), 128.62 (s), 127.59 (s), 126.99 (s), 126.39 (s), 117.82 (s), 113.23 (s), 46.37 (s).

4-Anilinomethyl-2-methoxyphenol (3ee)[22]

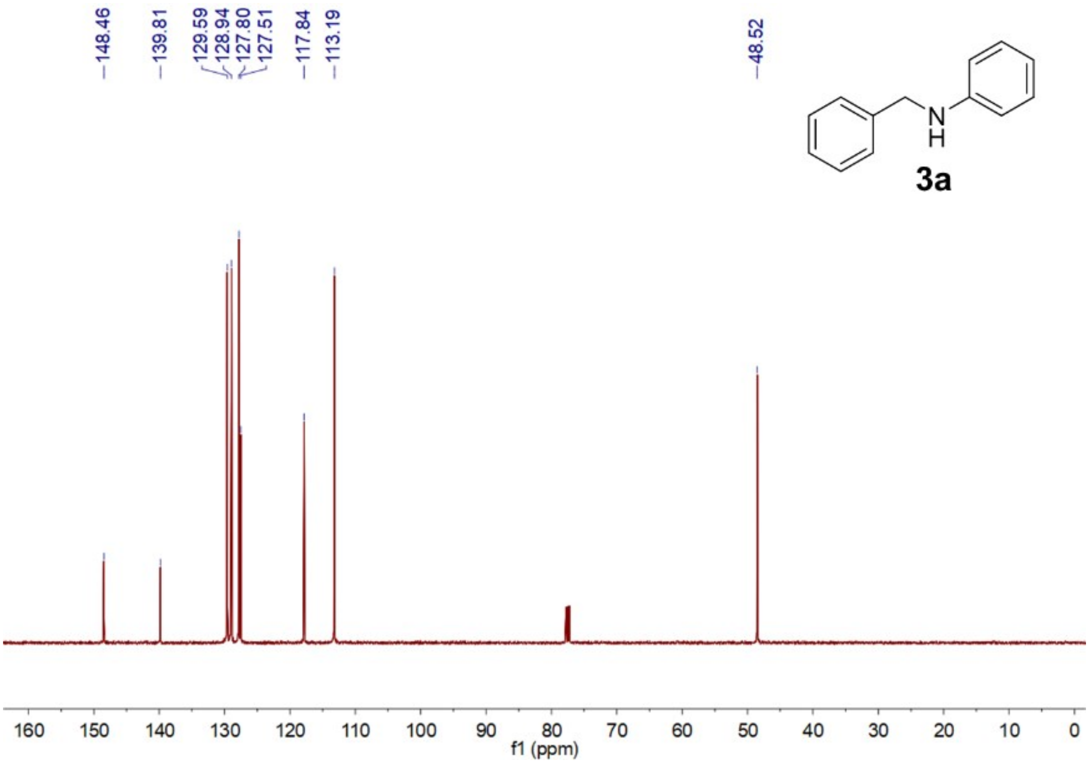


^{1}H NMR (500 MHz, $CDCl_3$) δ 7.22 (t, *J* = 7.9 Hz, 2H), 6.90 (dd, *J* = 14.0, 8.0 Hz, 3H), 6.77 (t, *J* = 7.3 Hz, 1H), 6.68 (d, *J* = 7.9 Hz, 2H), 4.55 (s, 1H), 4.26 (s, 2H), 3.87 (s, 3H). **^{13}C NMR** (126 MHz, $CDCl_3$) δ 148.25 (s), 146.83 (s), 144.93 (s), 131.32 (s), 129.35 (s), 120.59 (s), 117.78 (s), 114.52 (s), 113.10 (s), 110.39 (s), 55.99 (s), 48.50 (s).

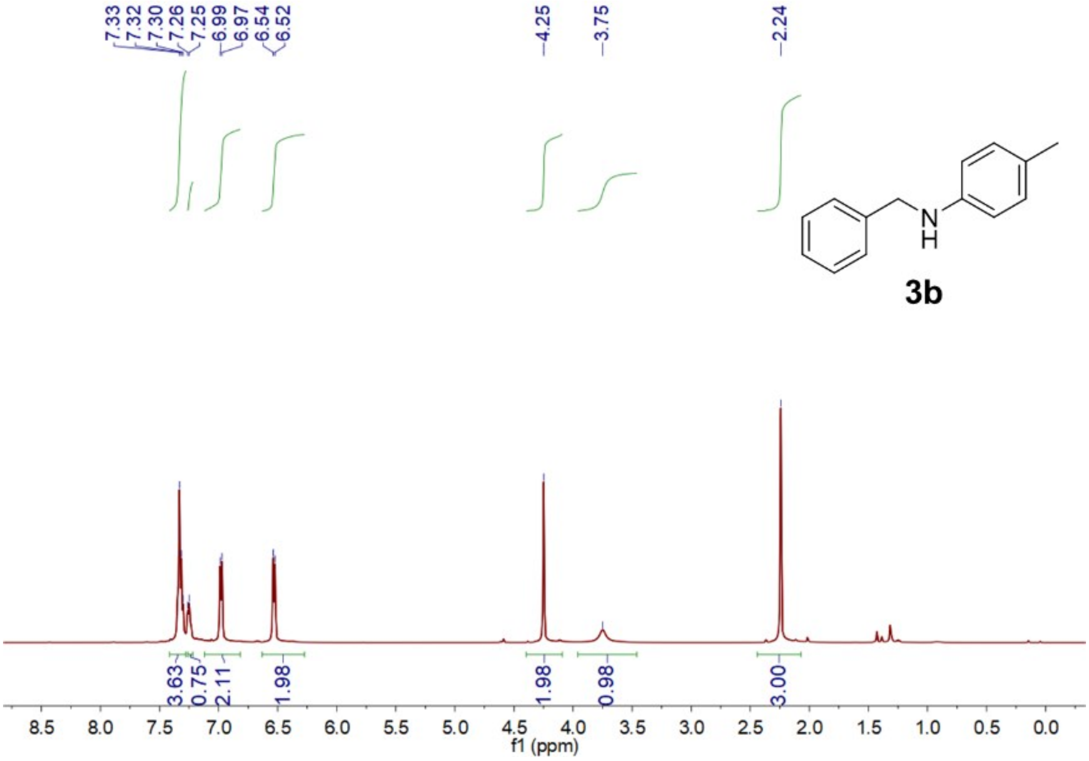
10. NMR spectra



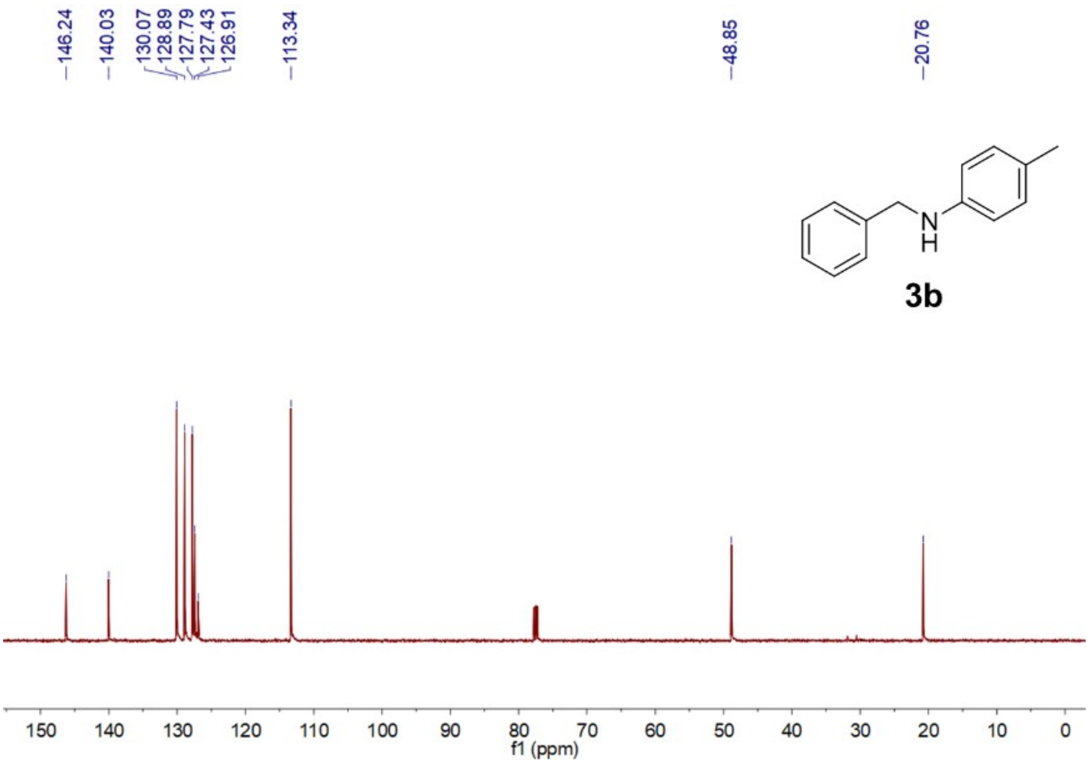
^{1}H NMR **3a**



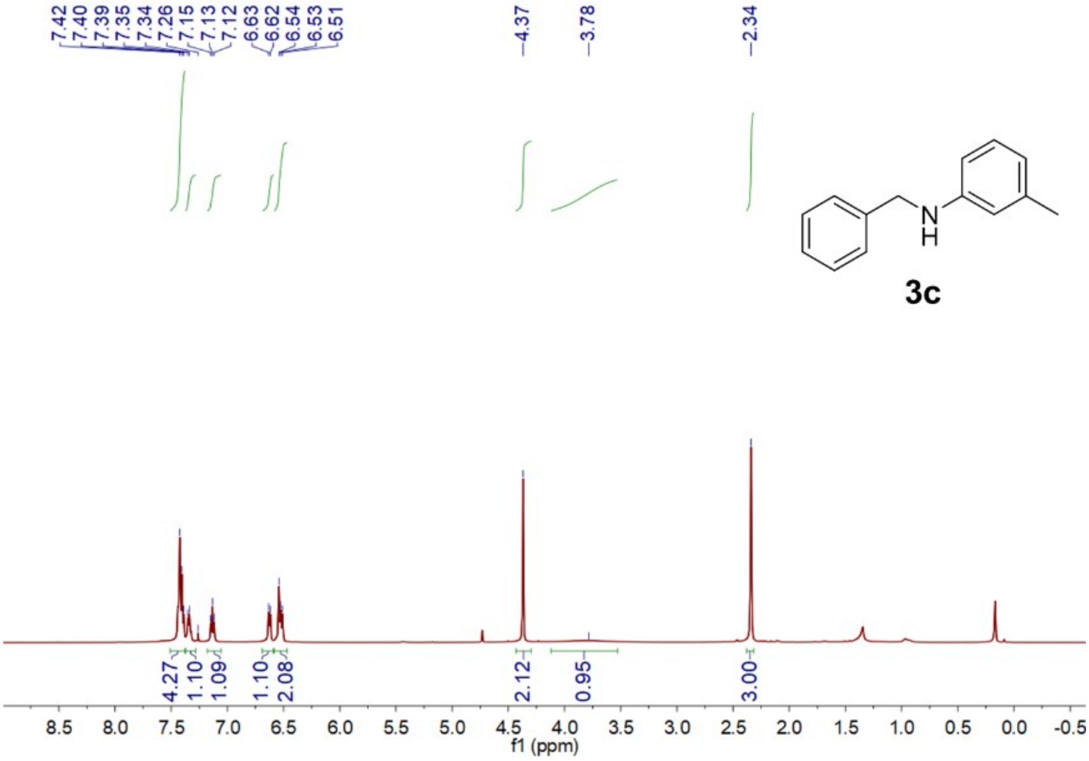
^{13}C NMR **3a**



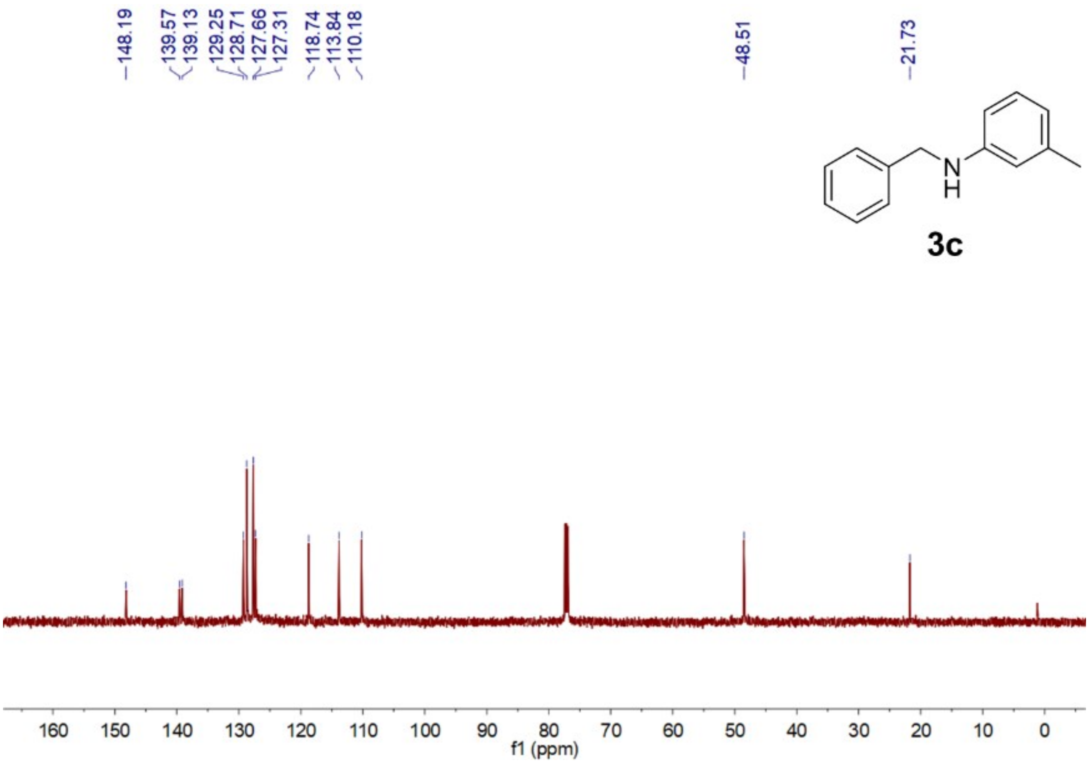
^{1}H NMR **3b**



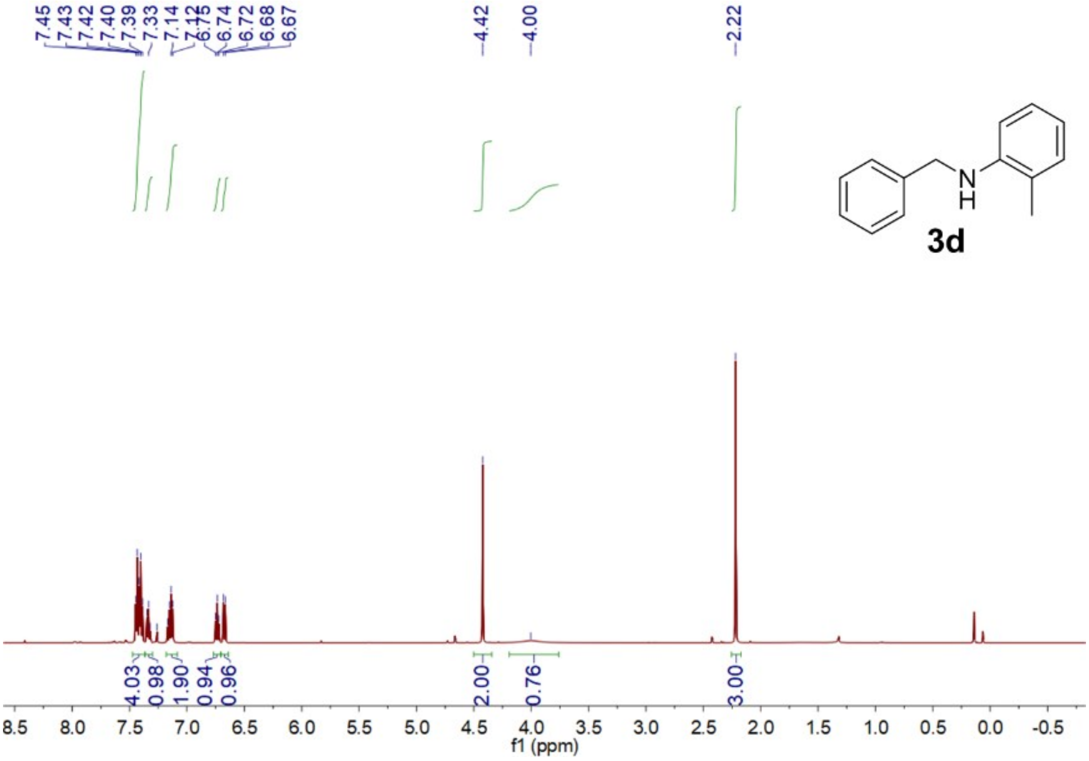
^{13}C NMR **3b**



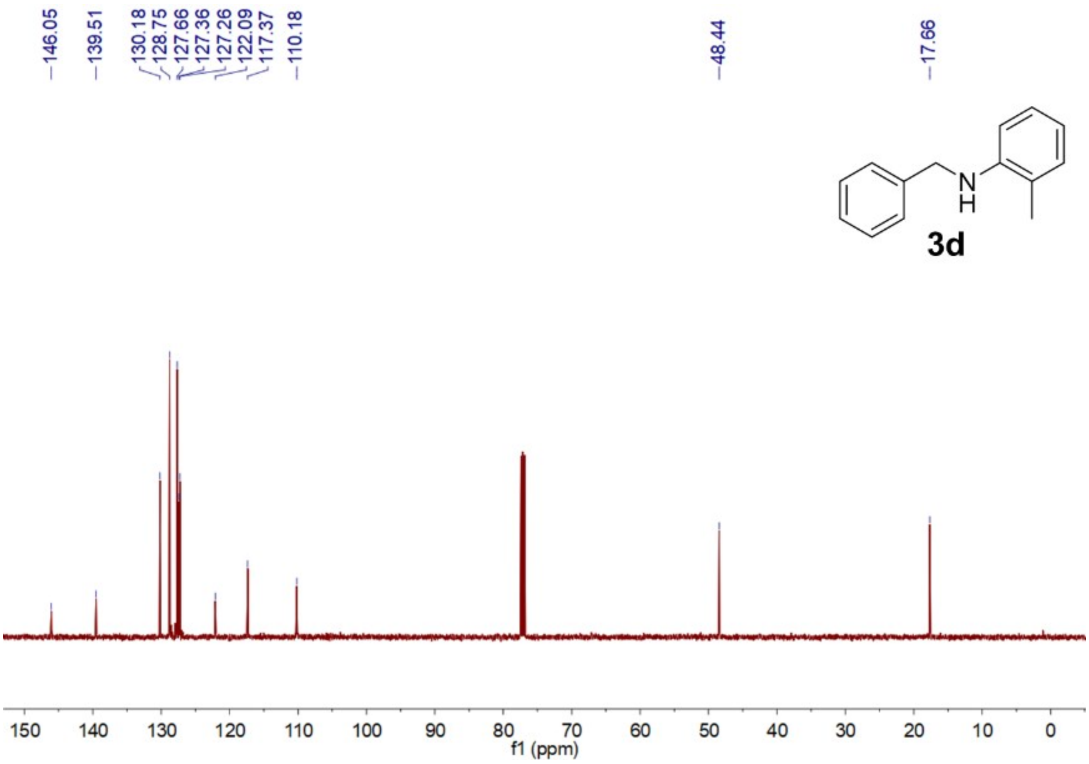
^{1}H NMR **3c**



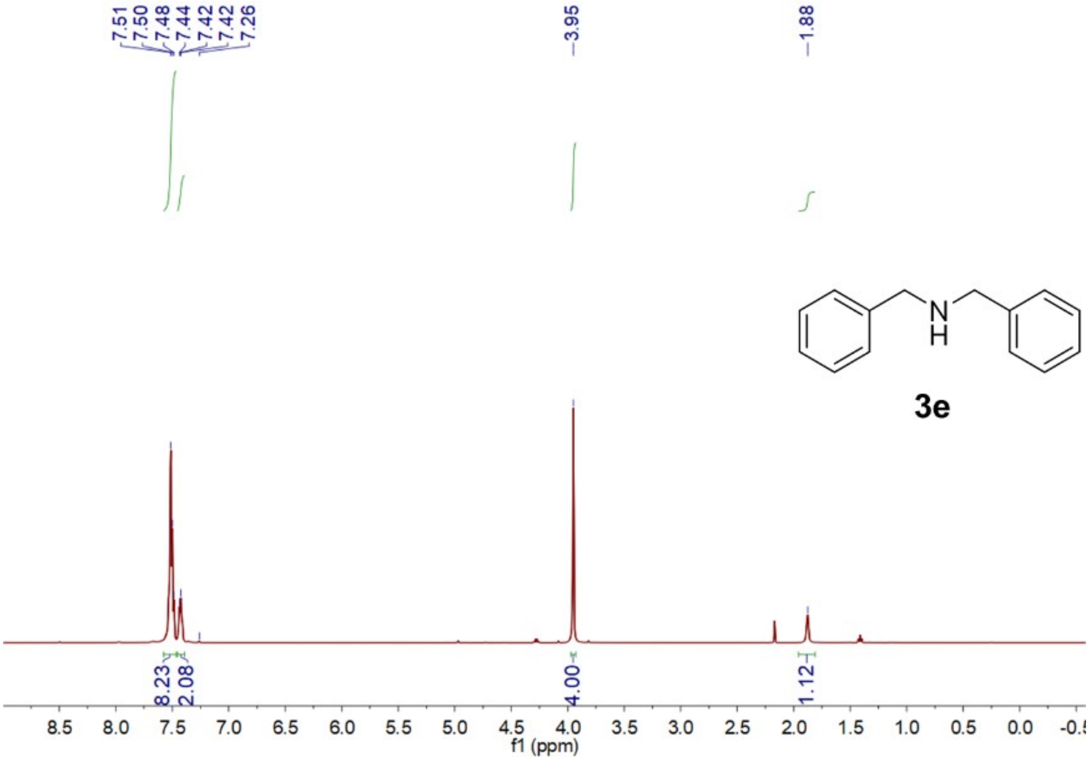
^{13}C NMR **3c**



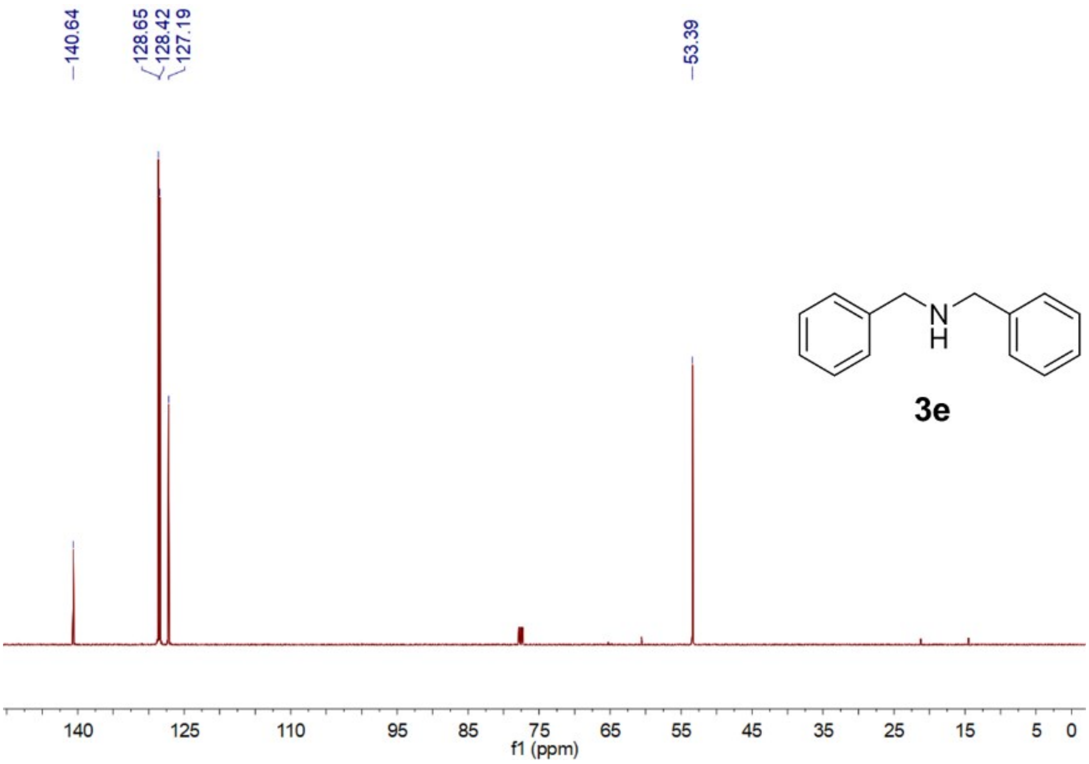
[1]H NMR **3d**



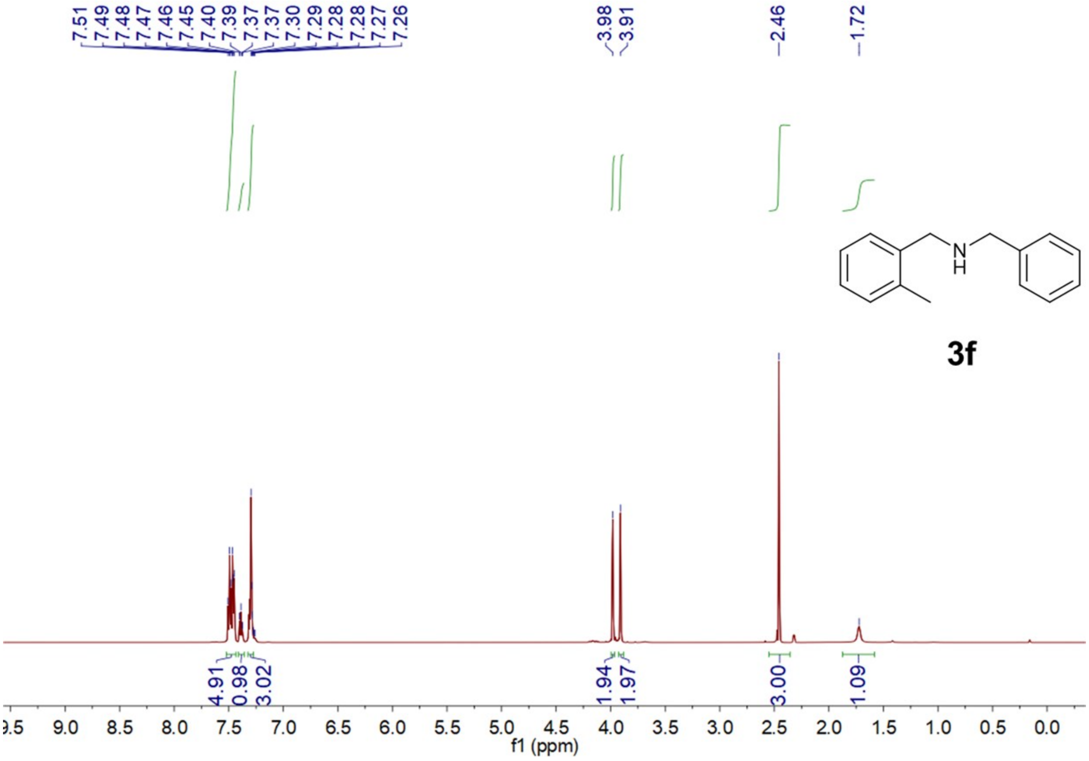
[13]C NMR **3d**



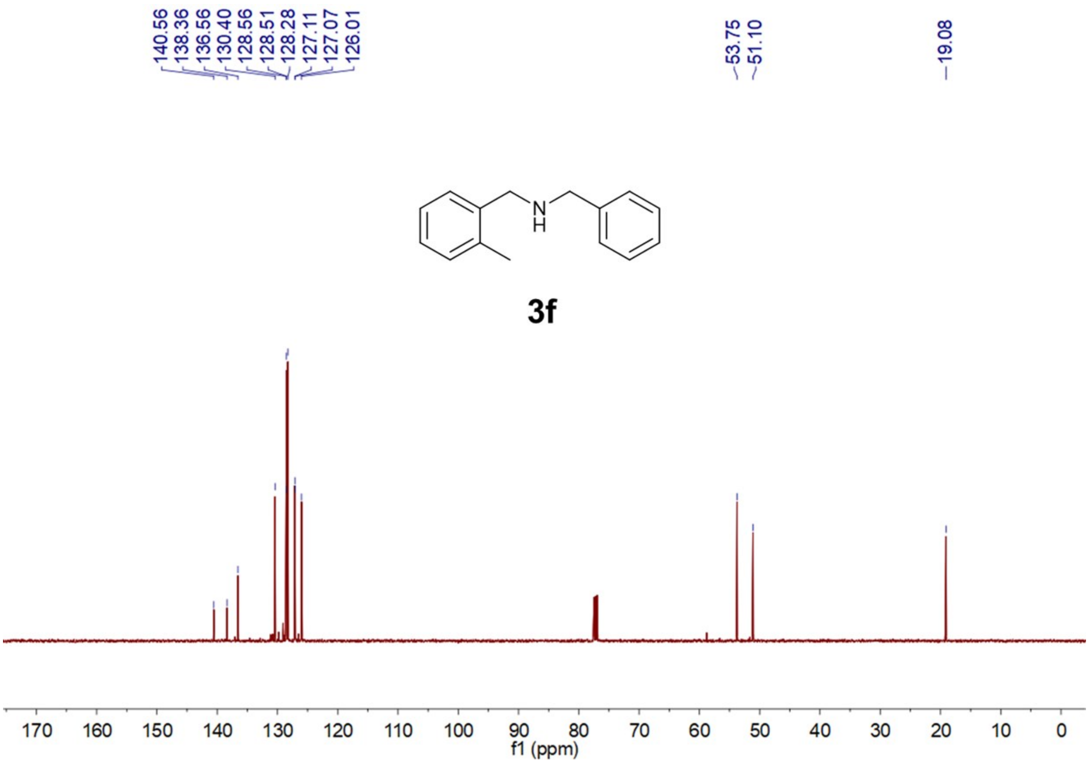
^{1}H NMR **3e**



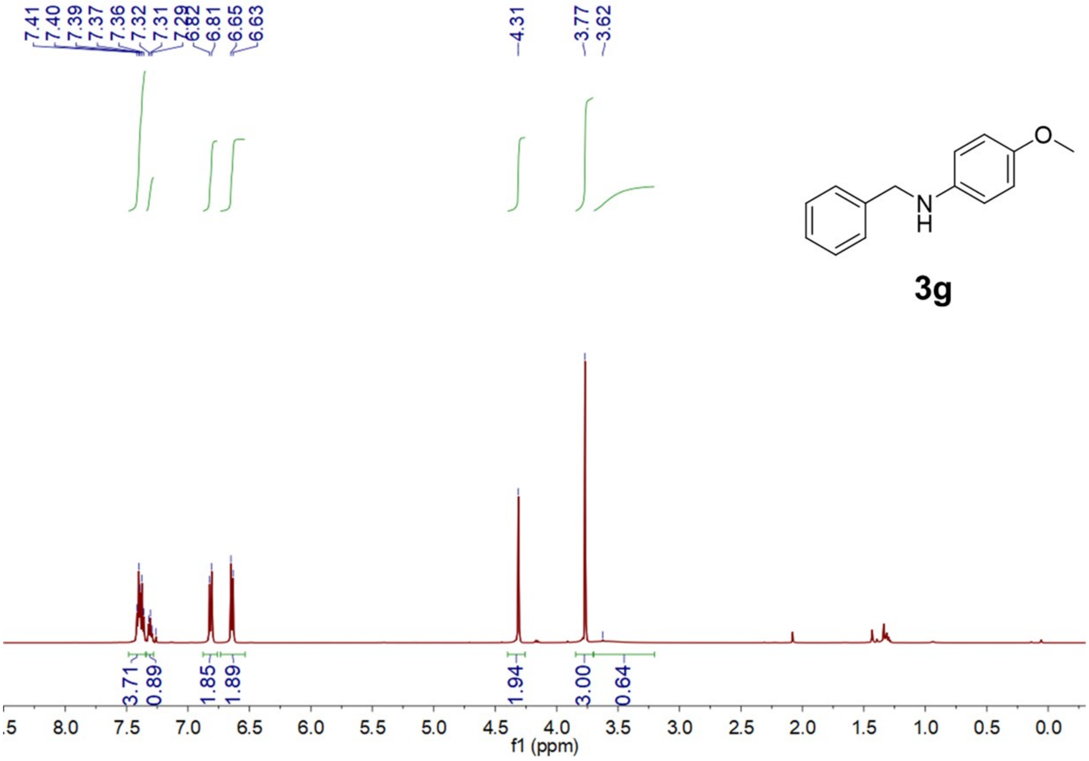
^{13}C NMR **3e**



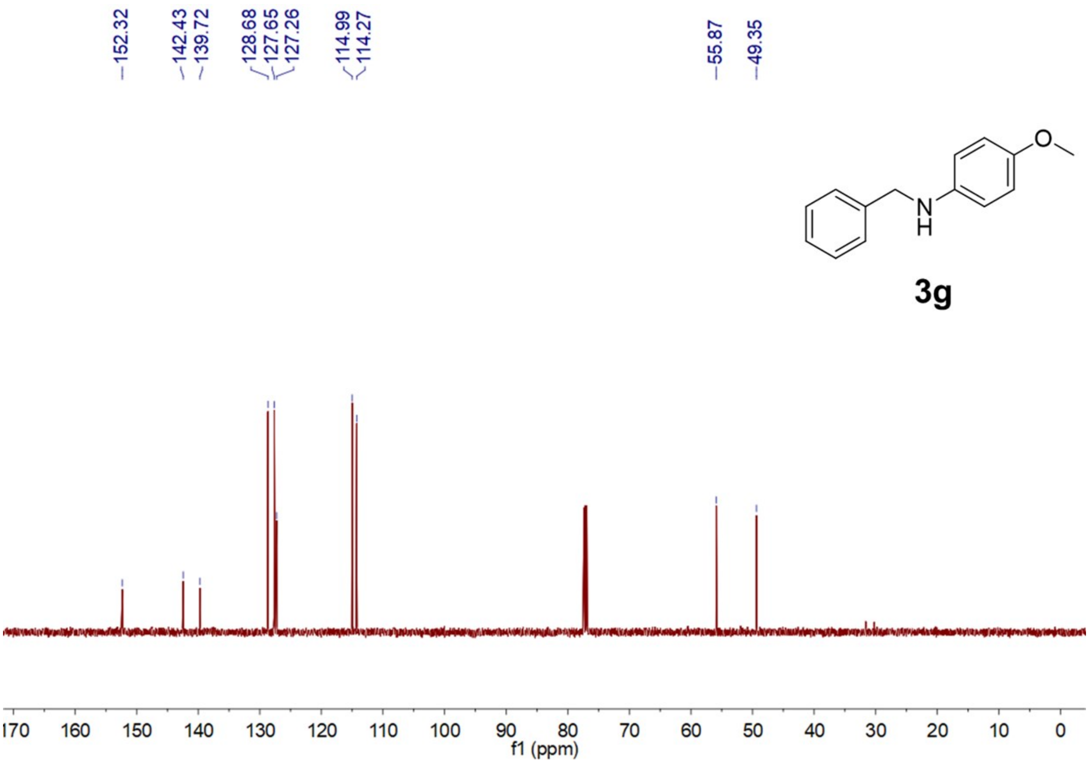
[1]H NMR **3f**



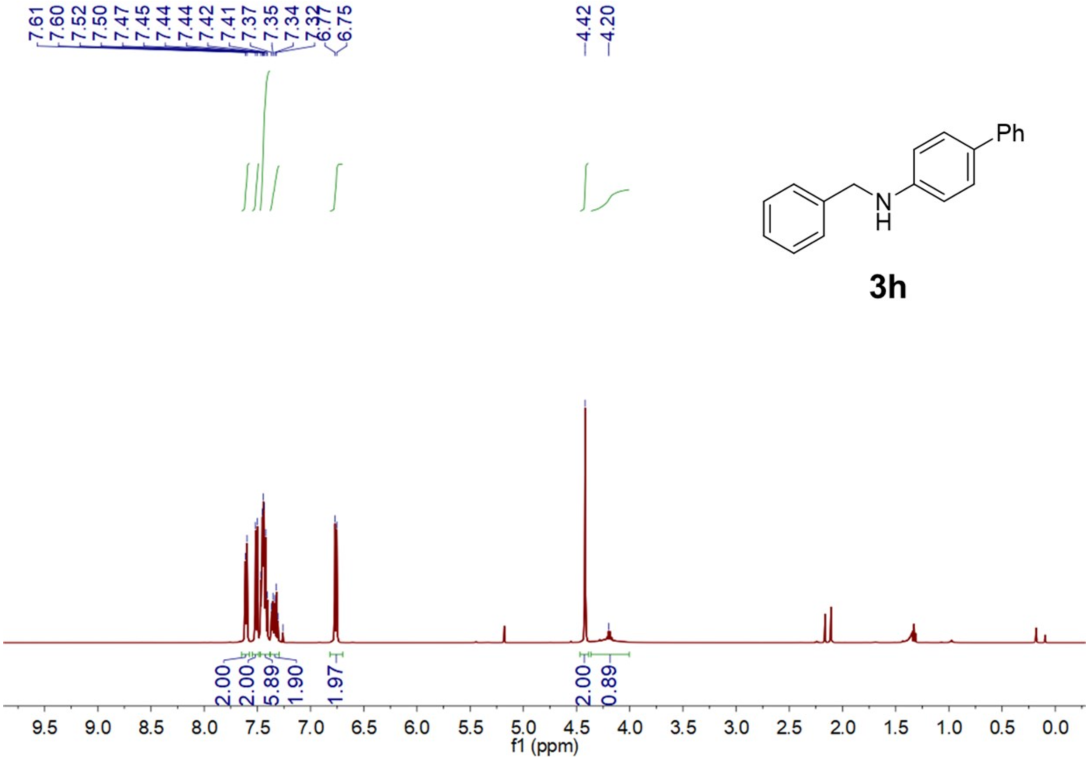
[13]C NMR **3f**



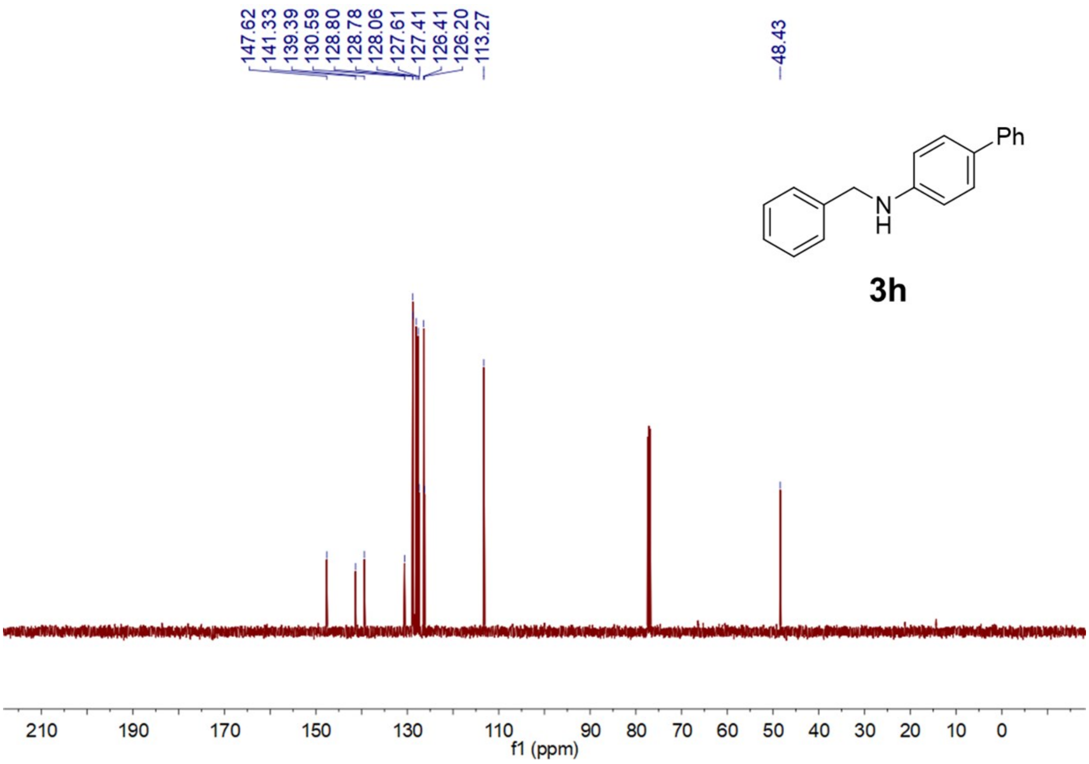
^{1}H NMR **3g**



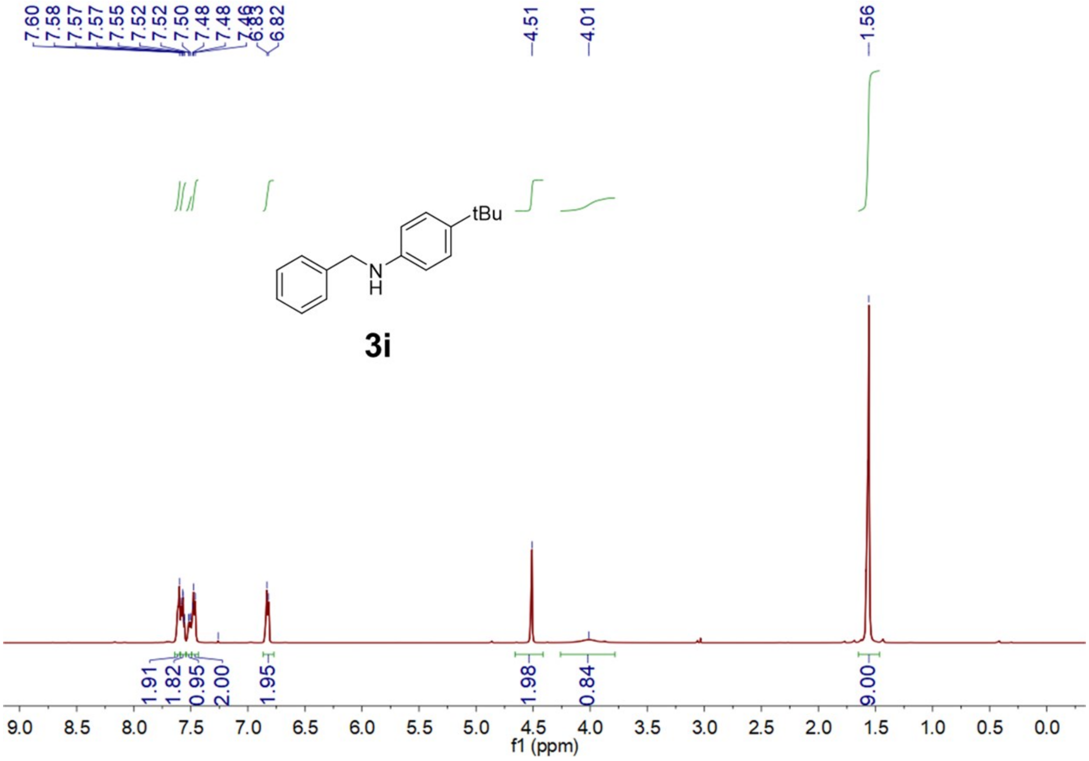
^{13}C NMR **3g**



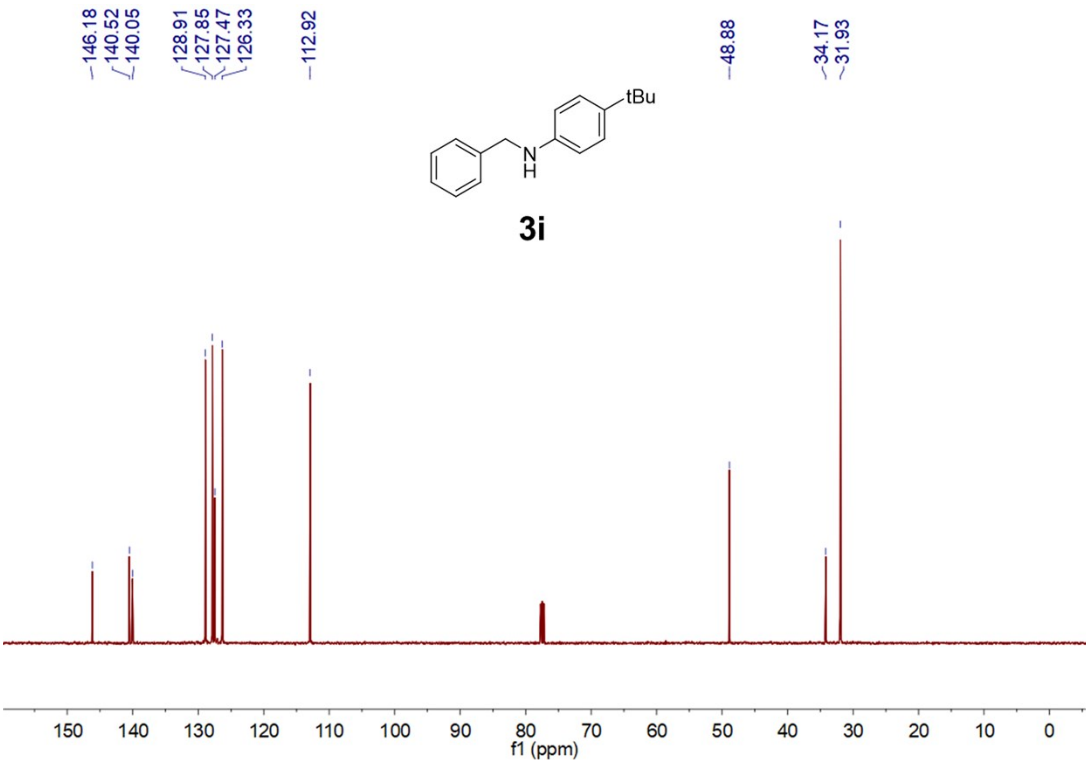
^{1}H NMR **3h**



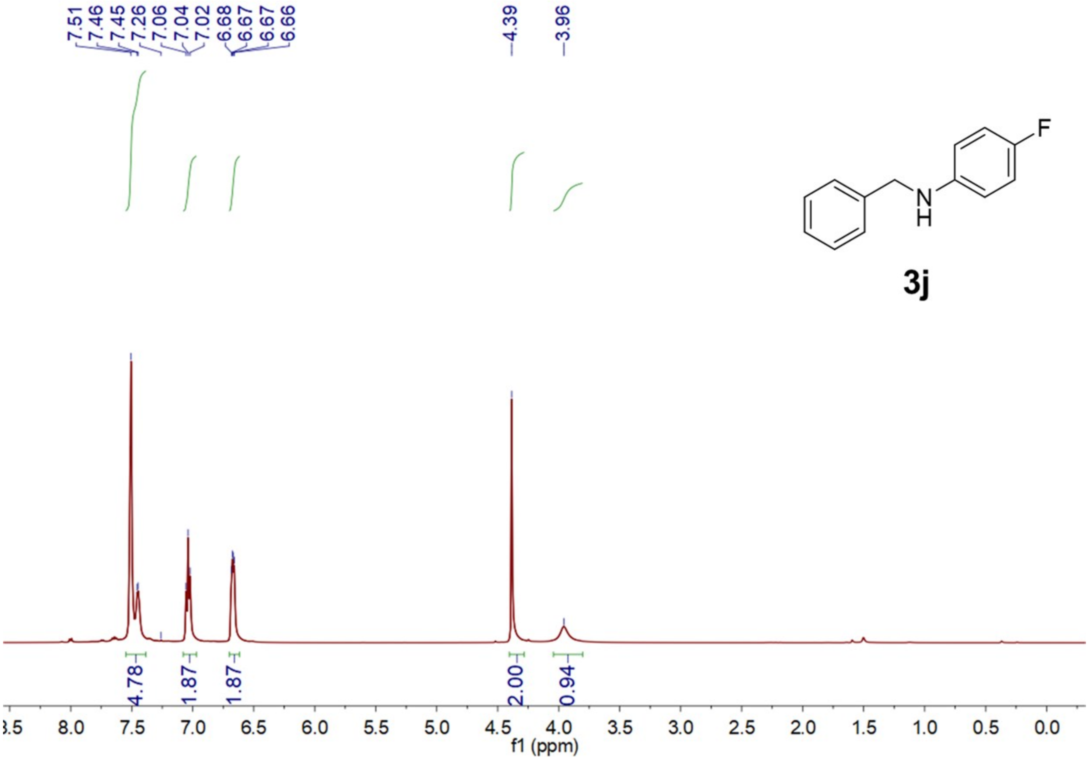
^{13}C NMR **3h**



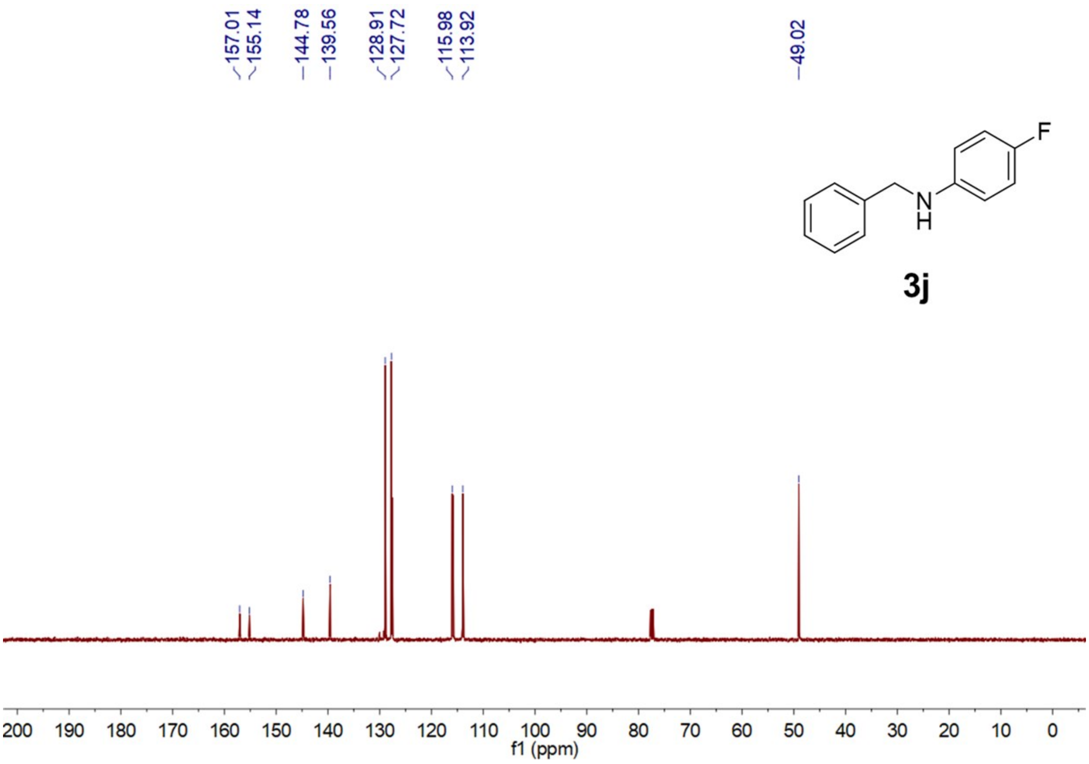
^{1}H NMR **3i**



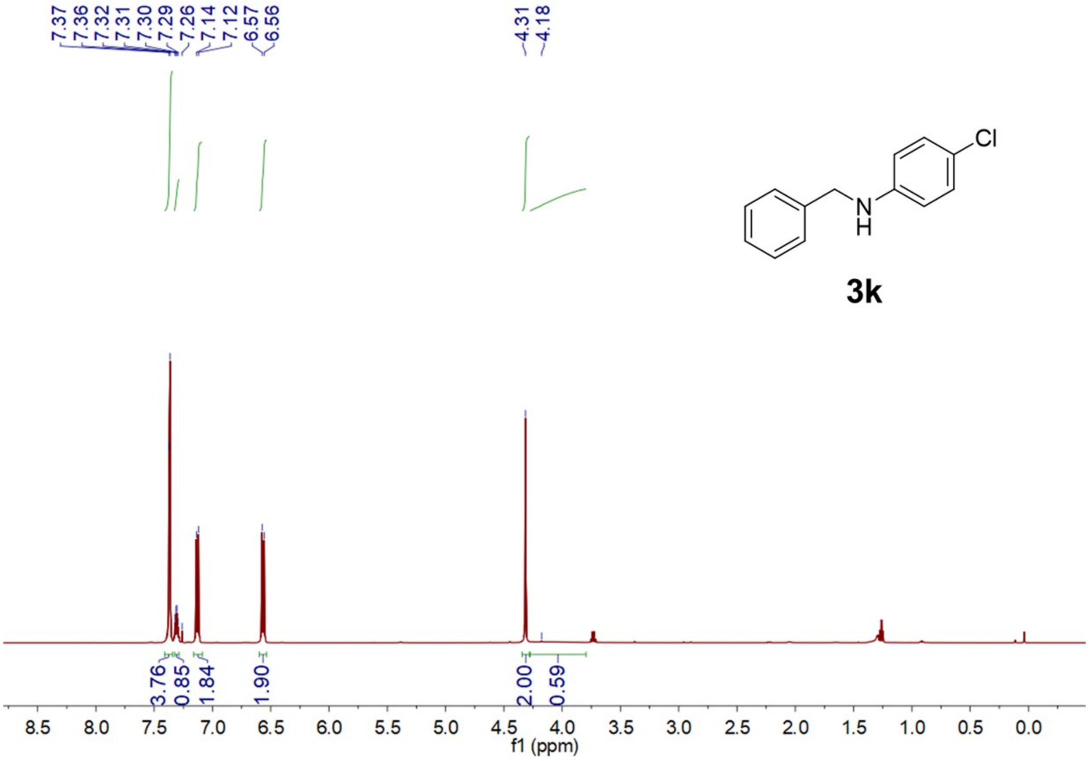
^{13}C NMR **3i**



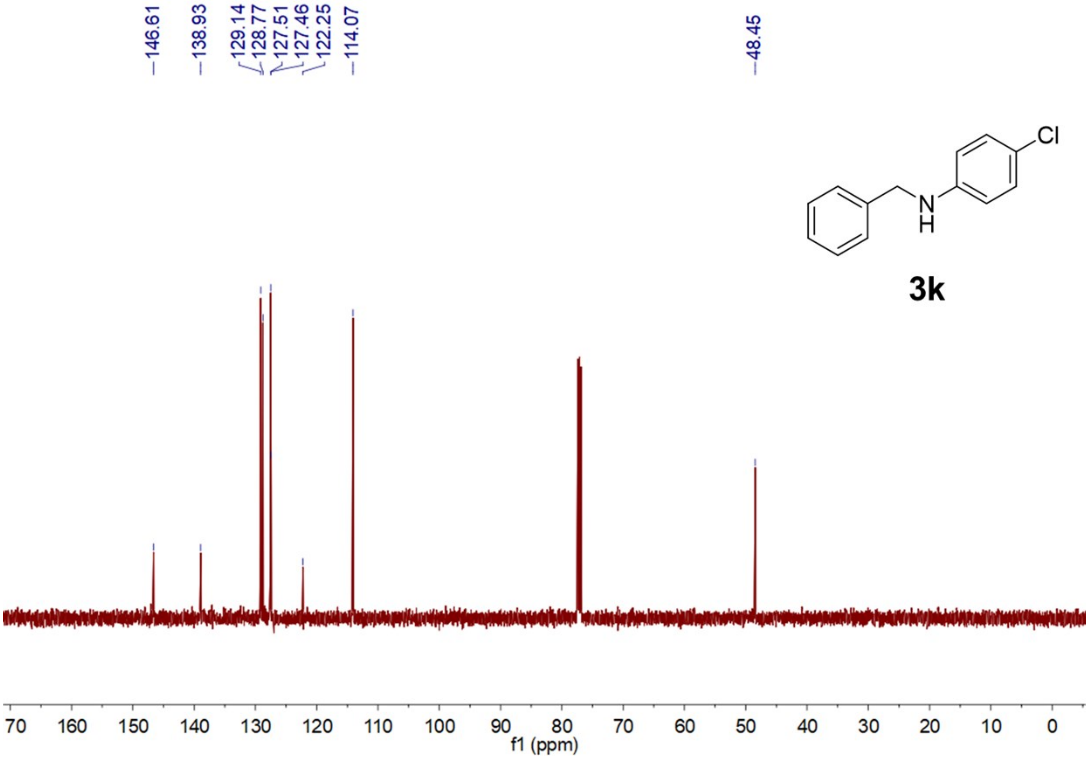
^{1}H NMR **3j**



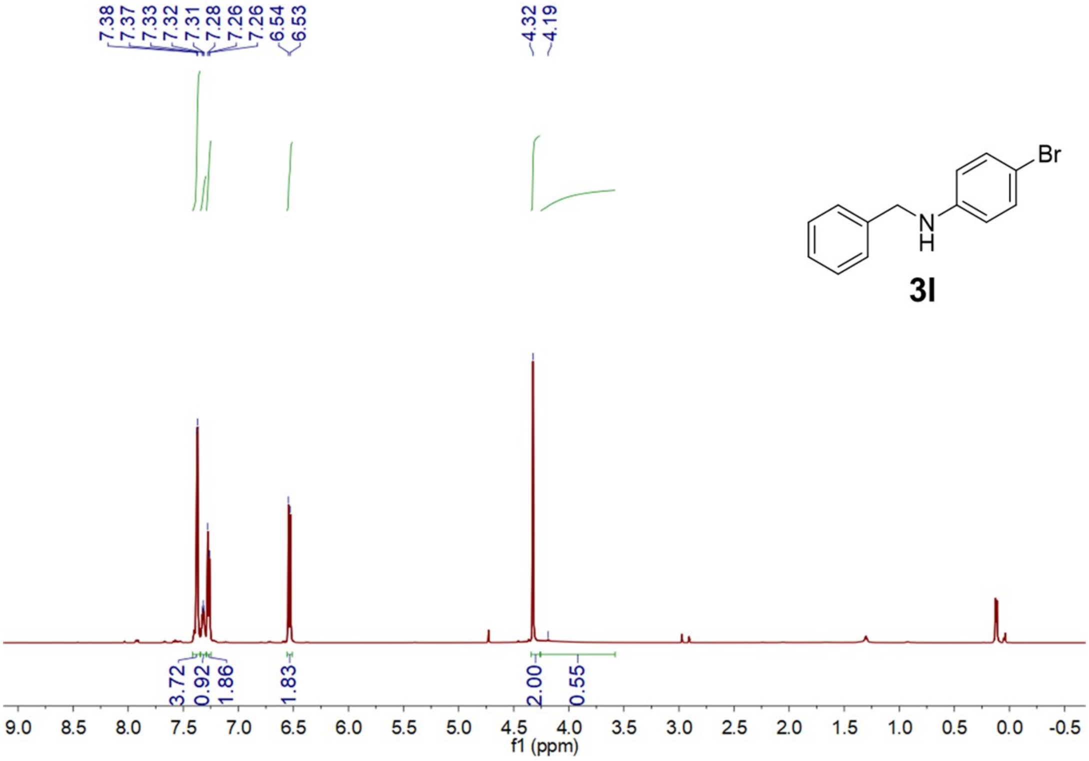
^{13}C NMR **3j**



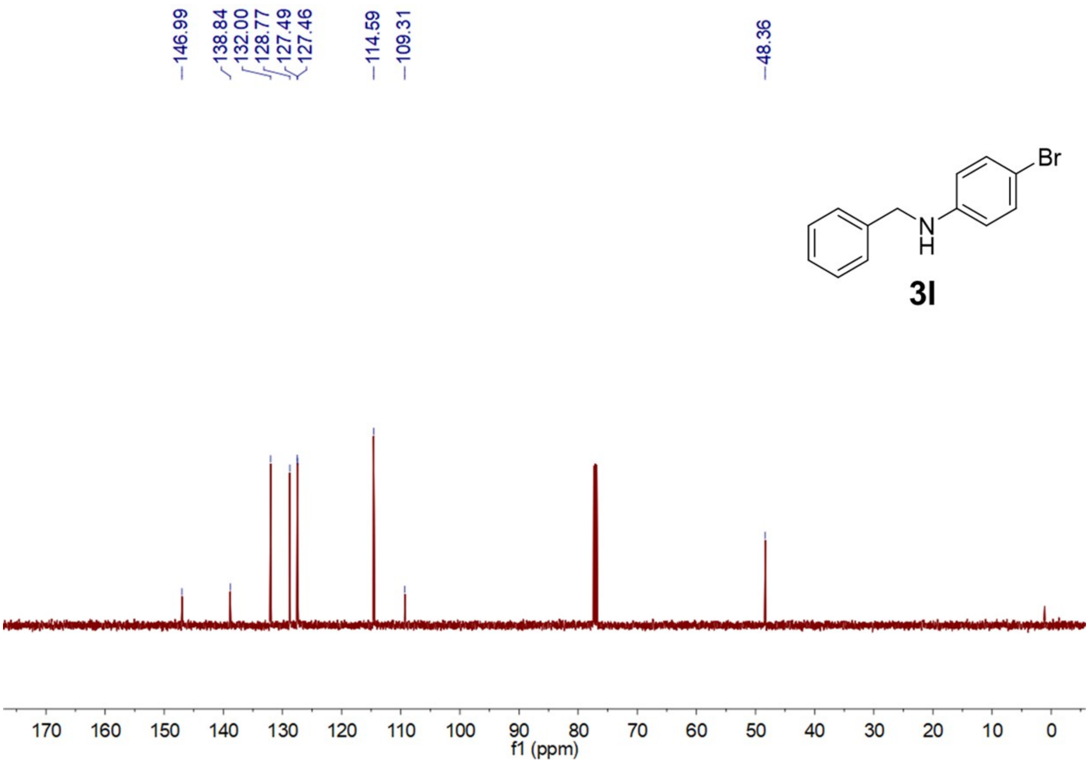
[1]H NMR **3k**



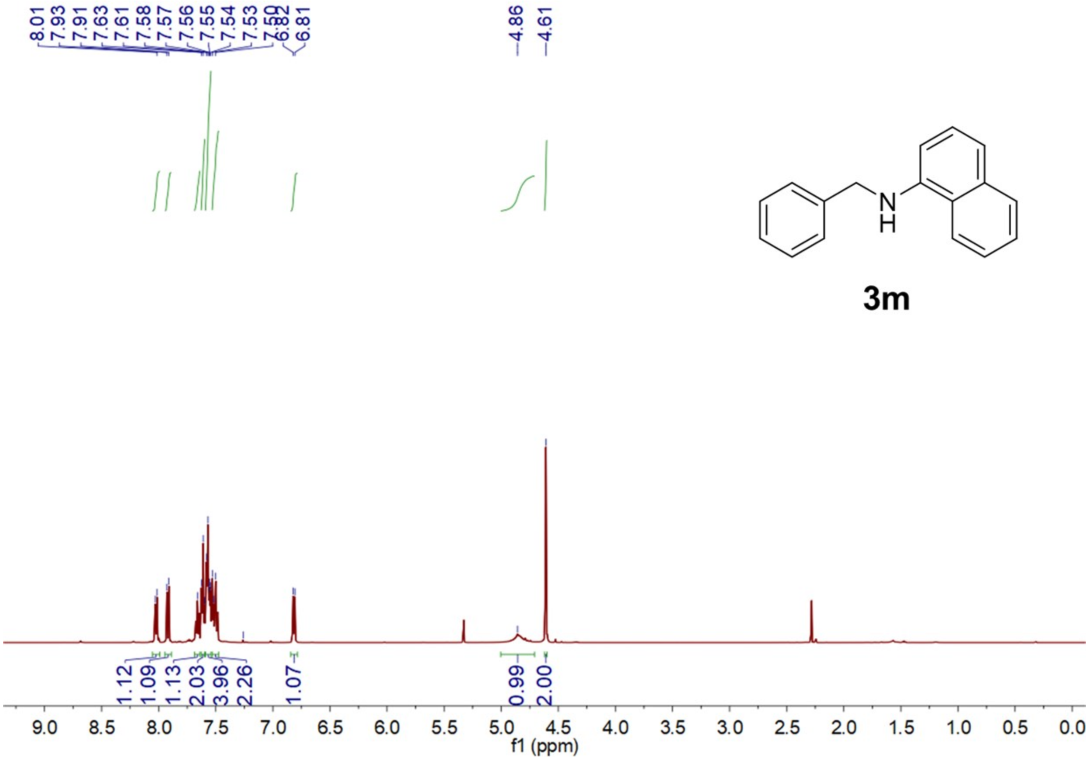
[13]C NMR **3k**



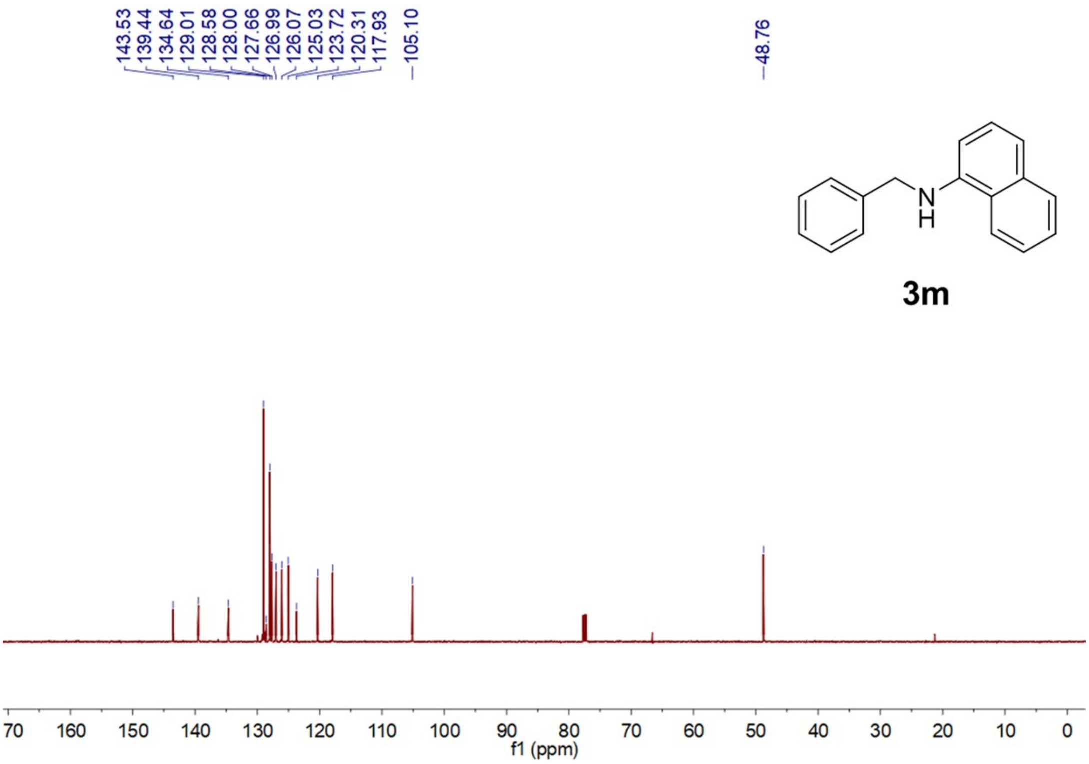
^{1}H NMR **3l**



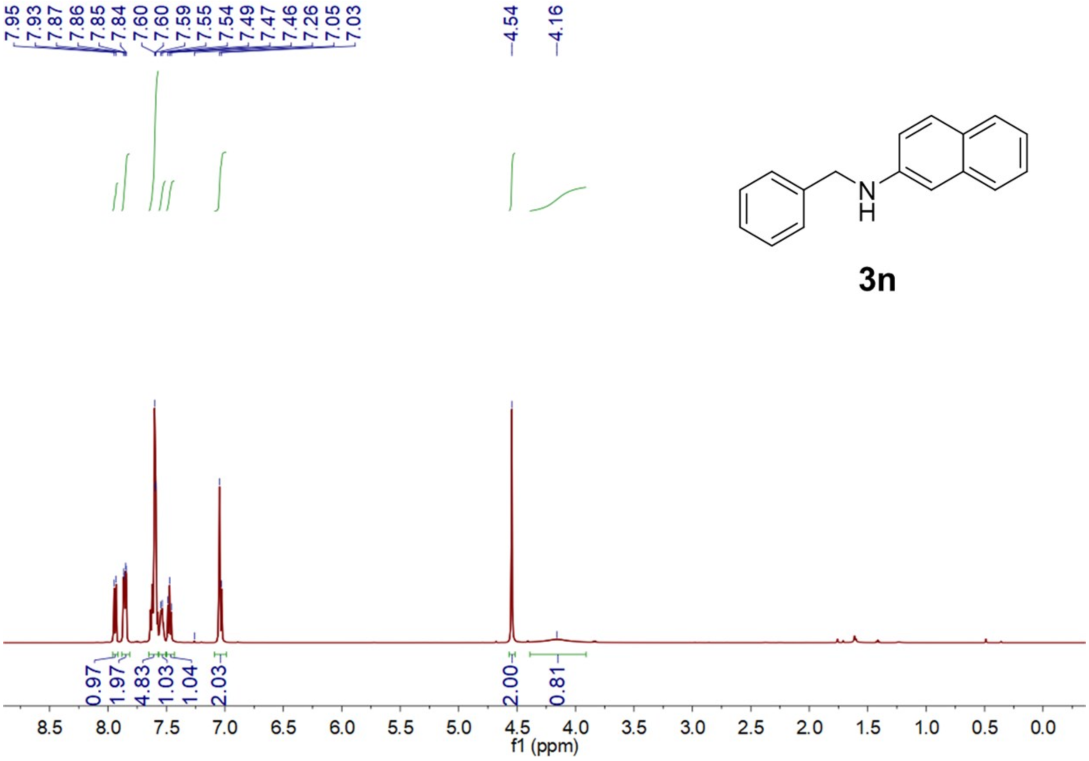
^{13}C NMR **3l**



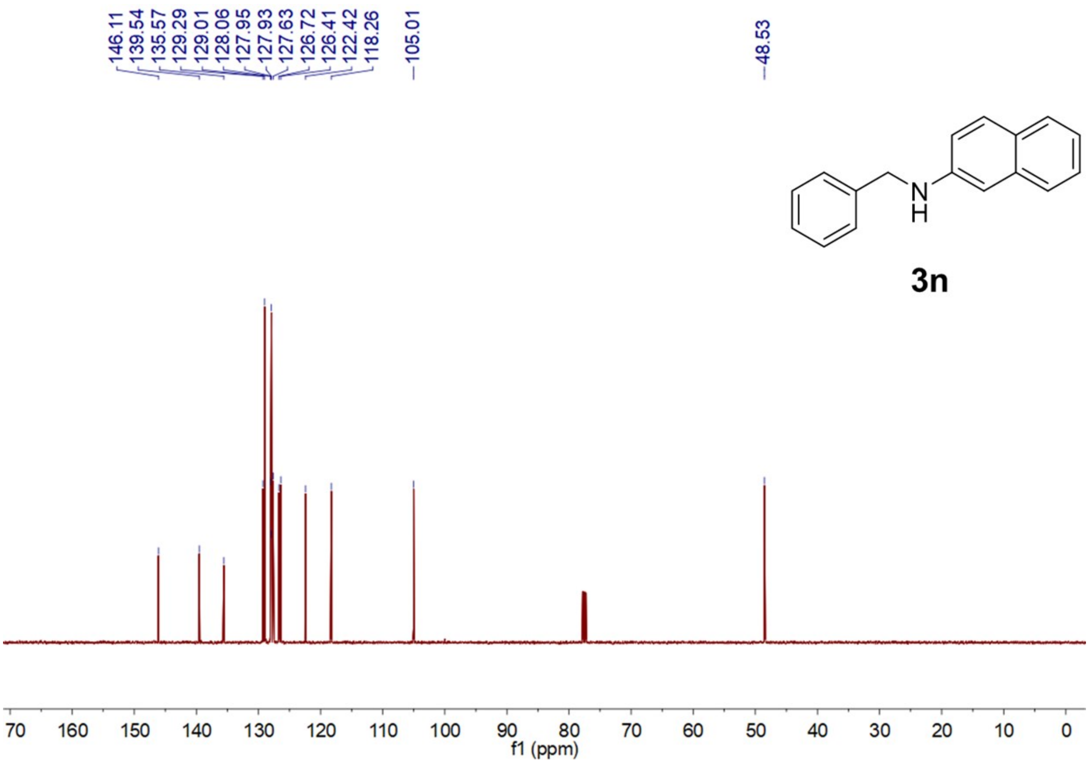
^{1}H NMR **3m**



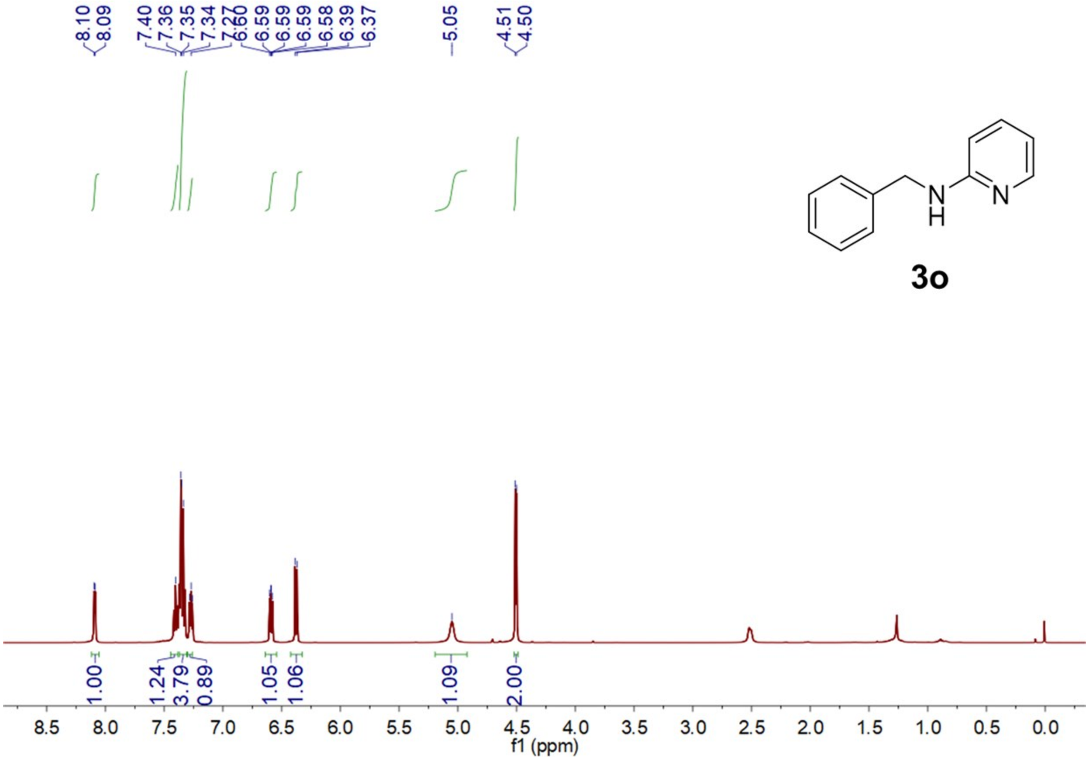
^{13}C NMR **3m**



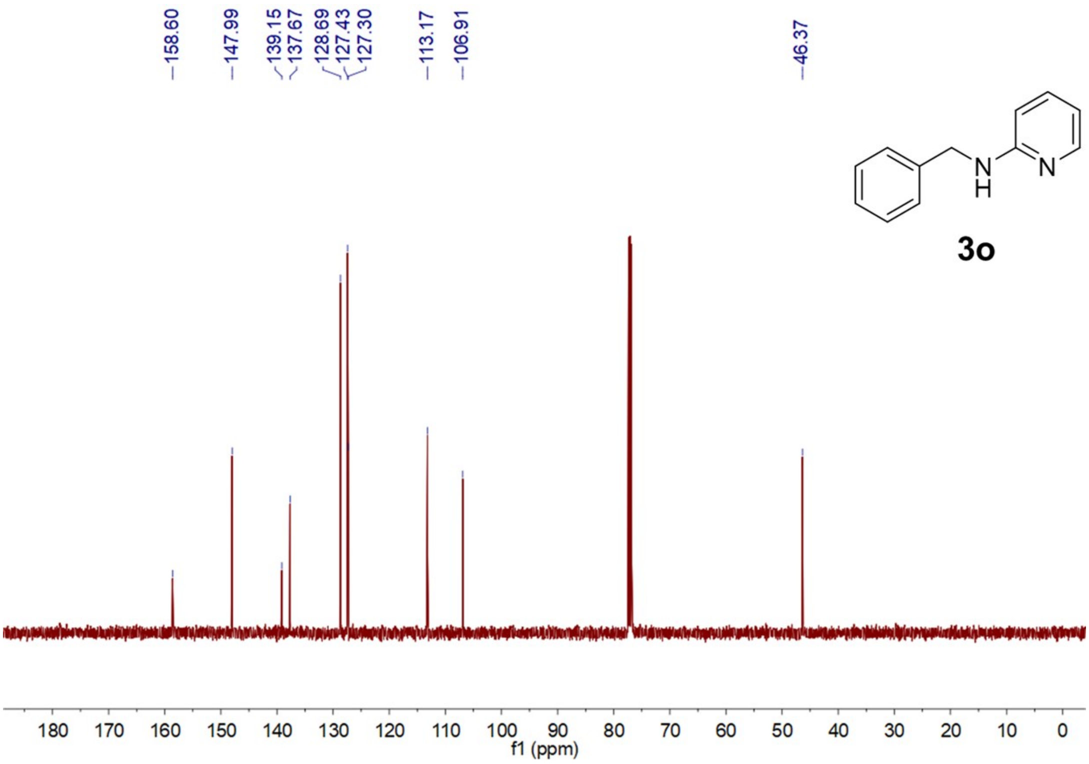
[1]H NMR **3n**



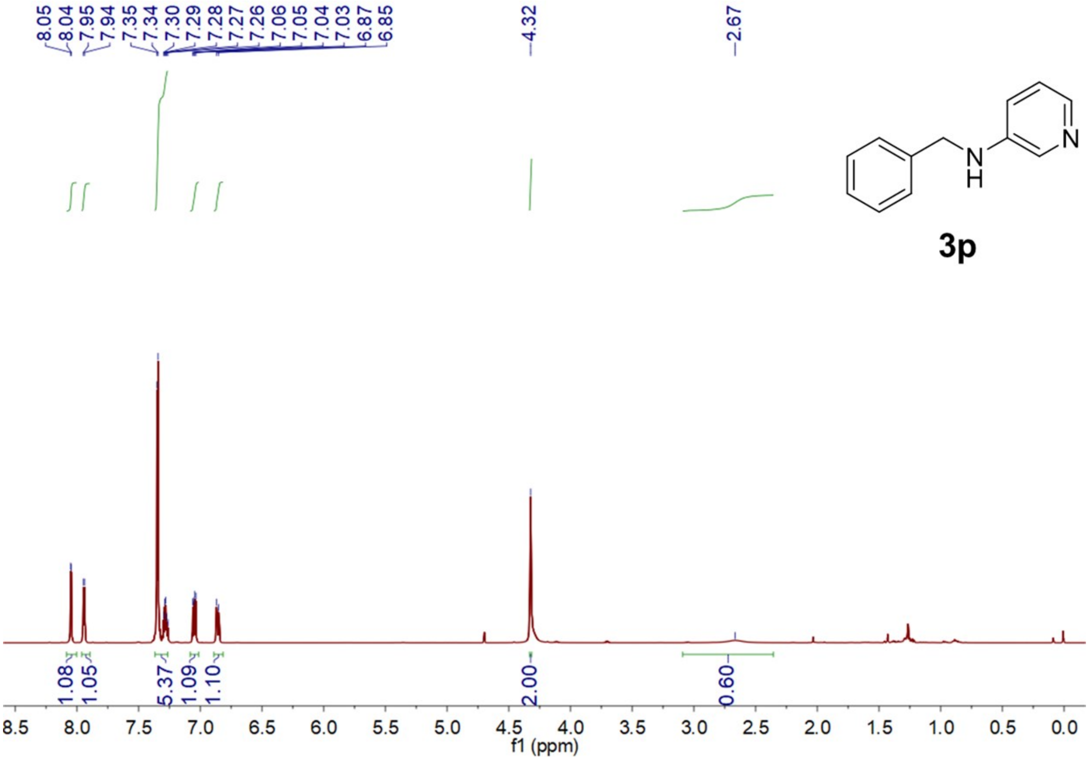
[13]C NMR **3n**



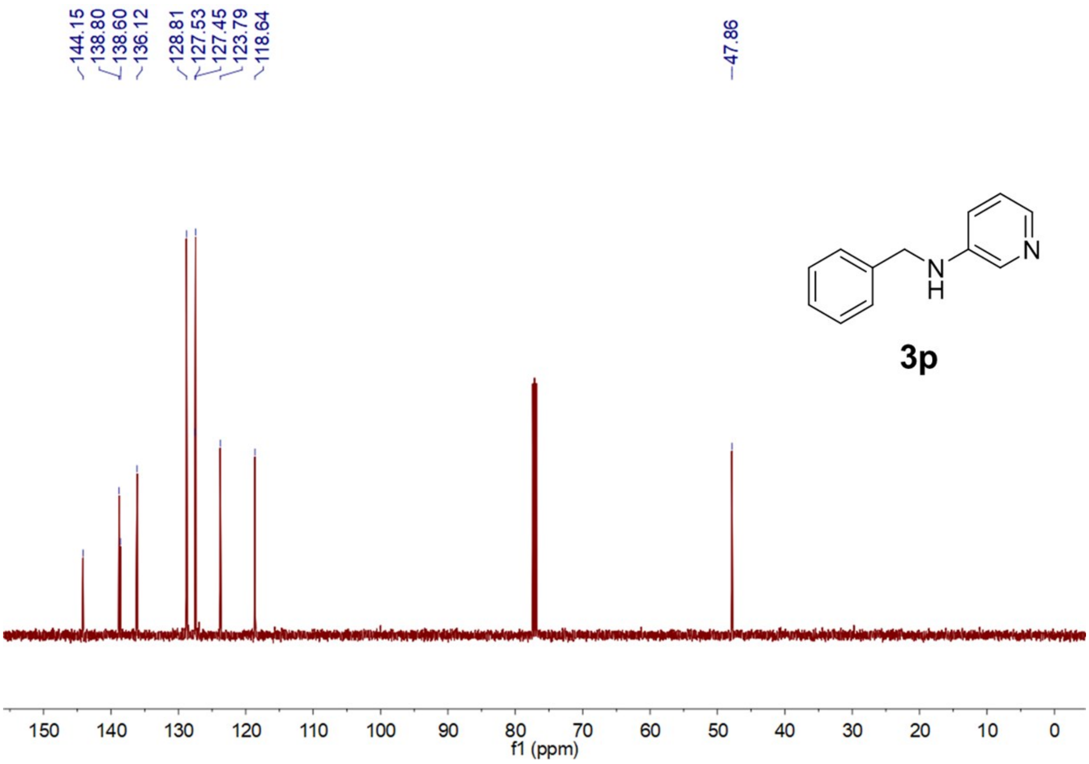
^{1}H NMR **3o**



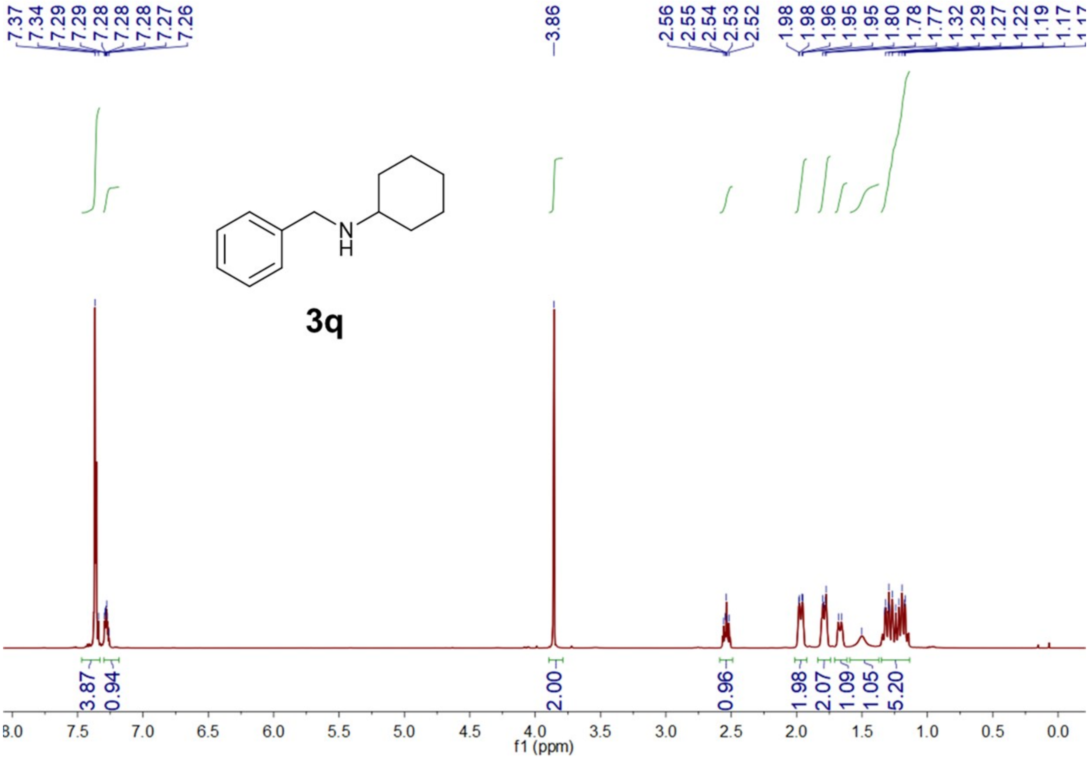
^{13}C NMR **3o**



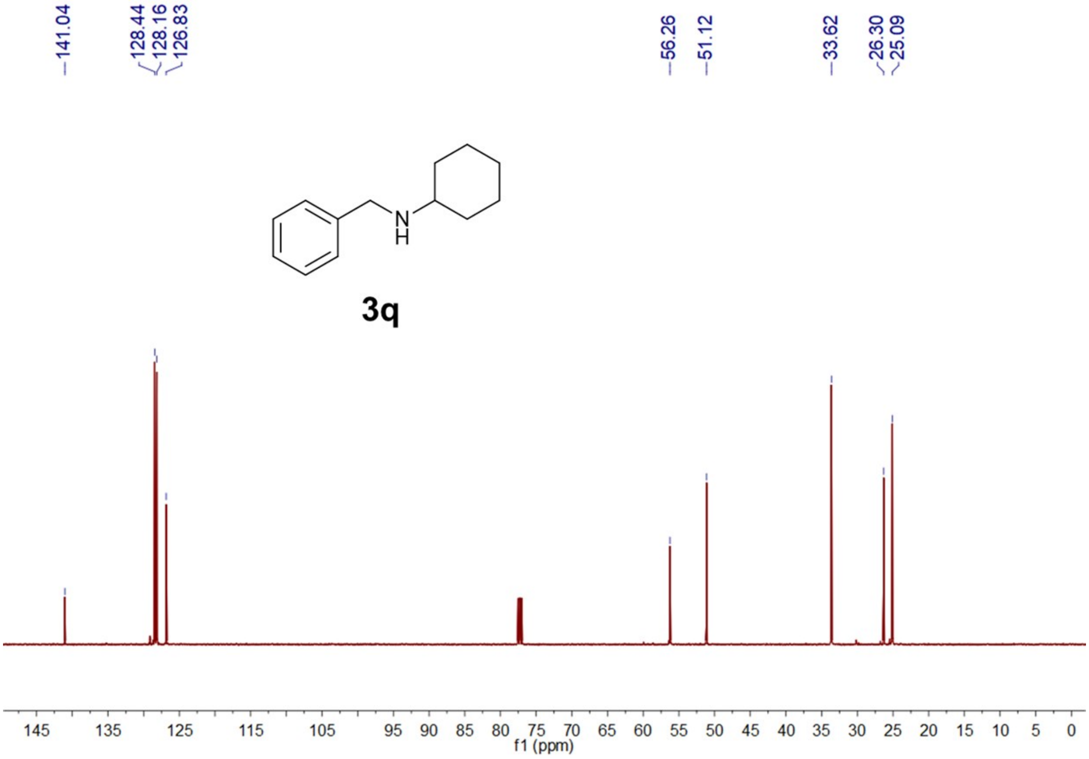
^{1}H NMR **3p**



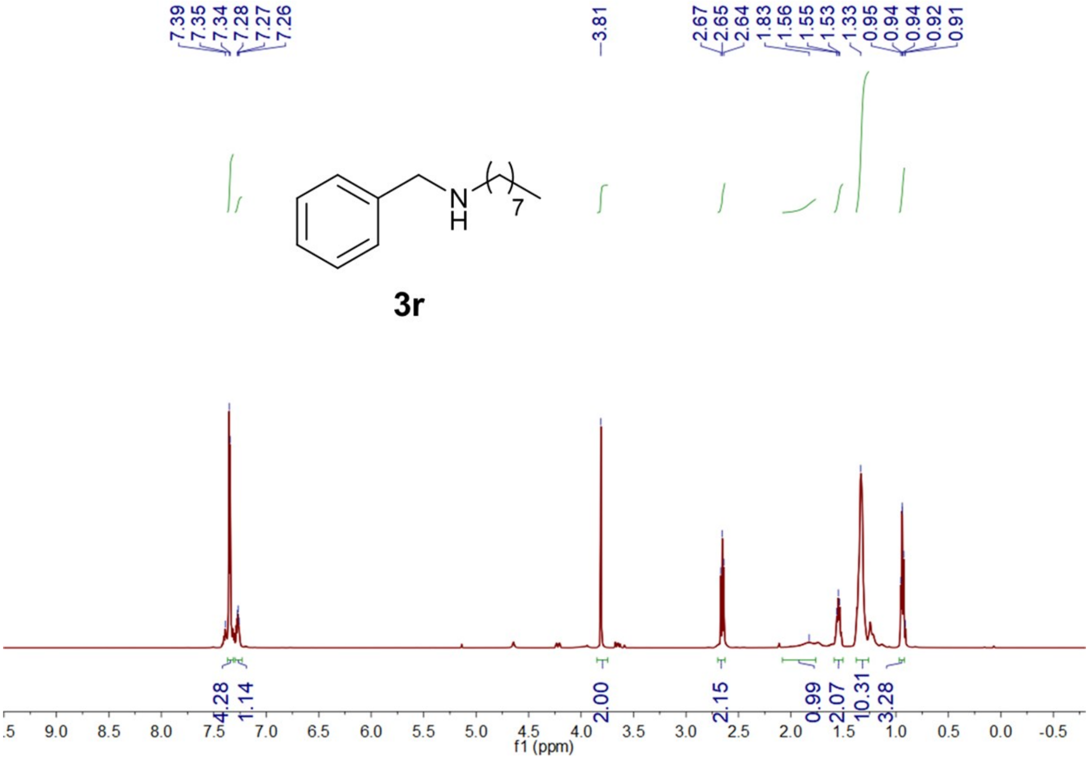
^{13}C NMR **3p**



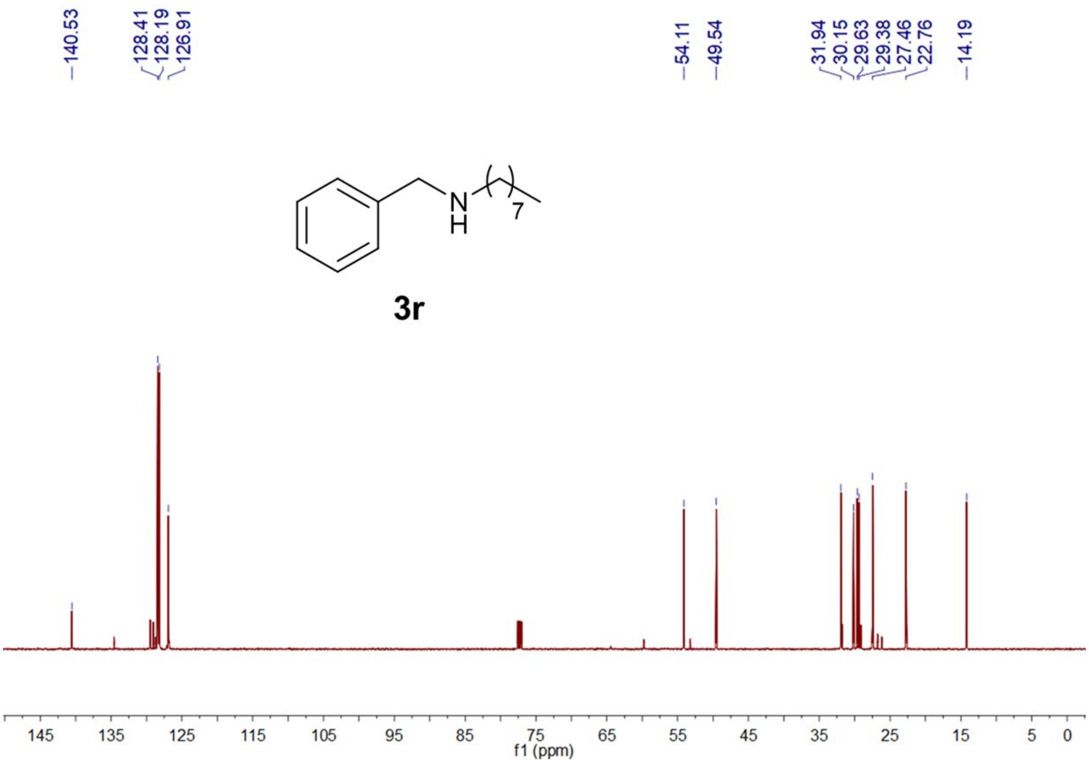
^{1}H NMR **3q**



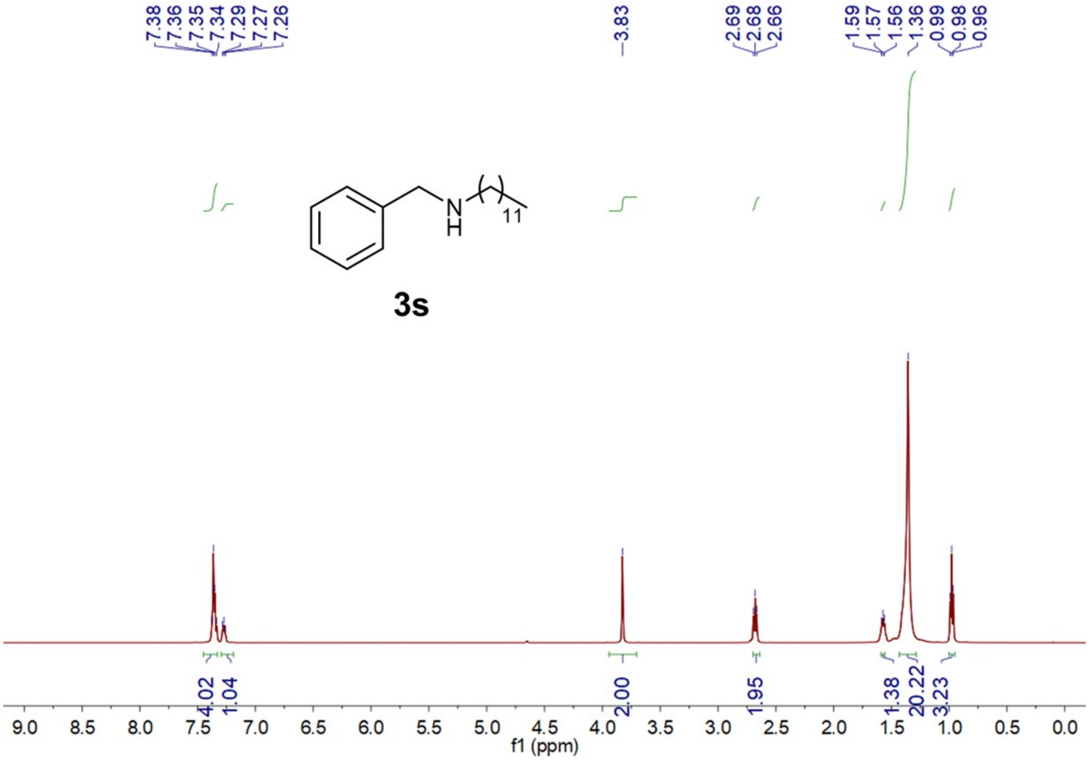
^{13}C NMR **3q**



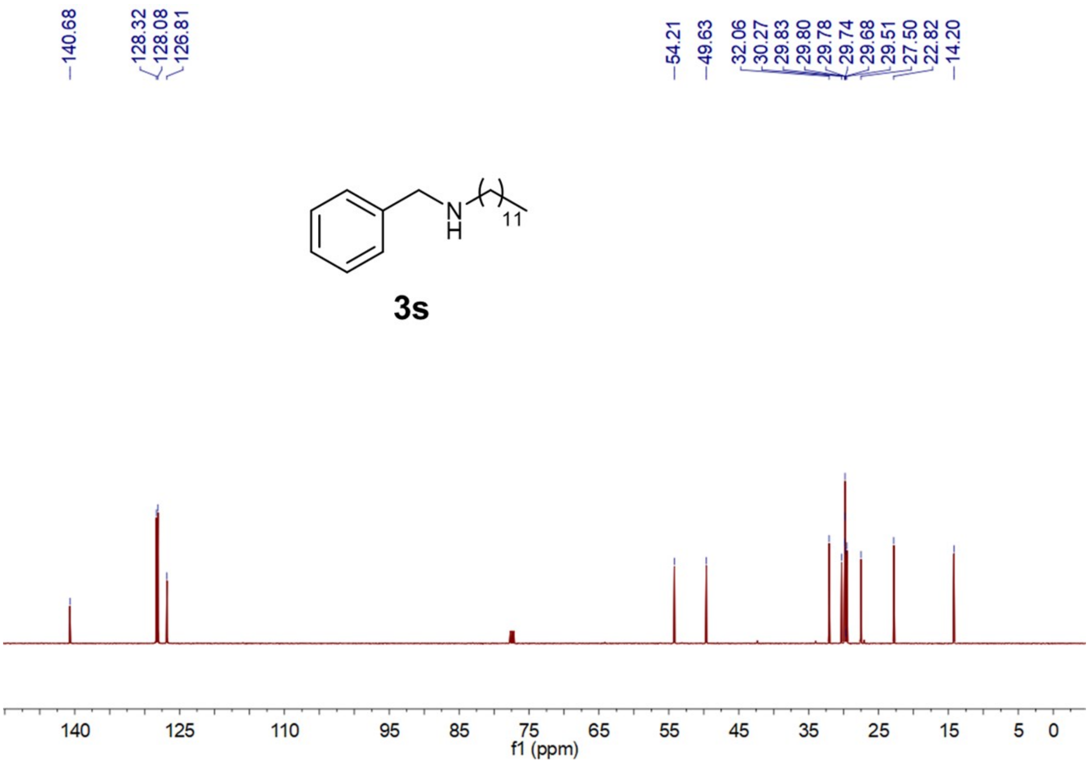
^{1}H NMR **3r**



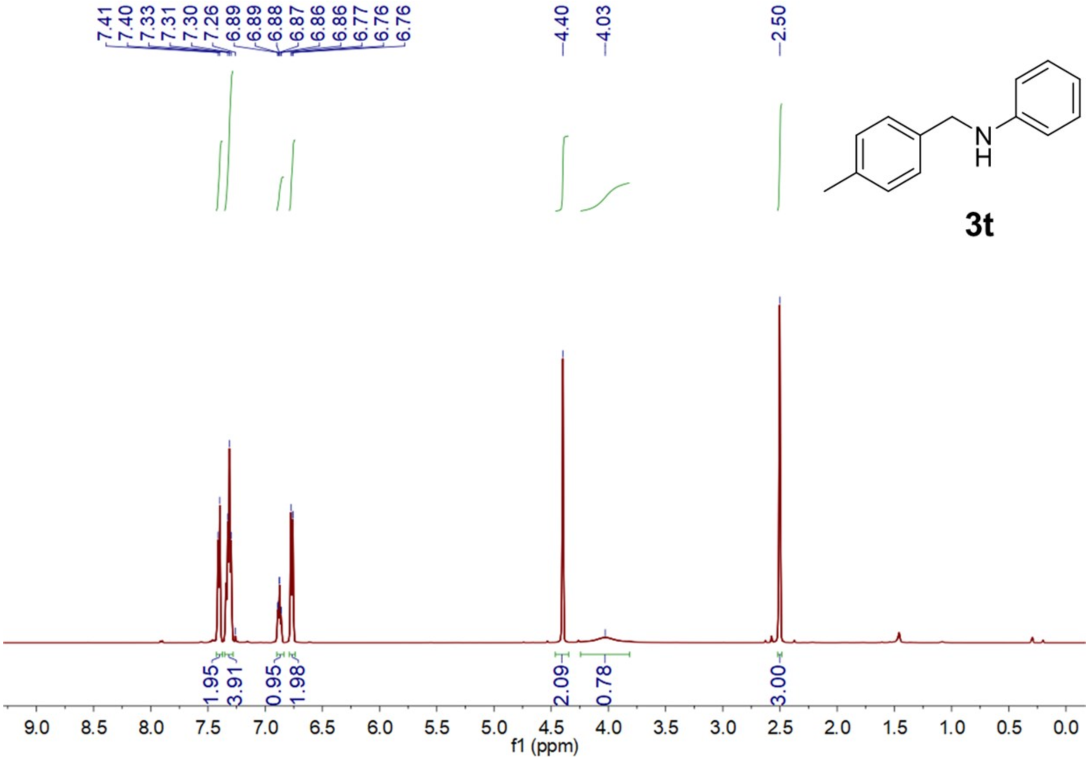
^{13}C NMR **3r**



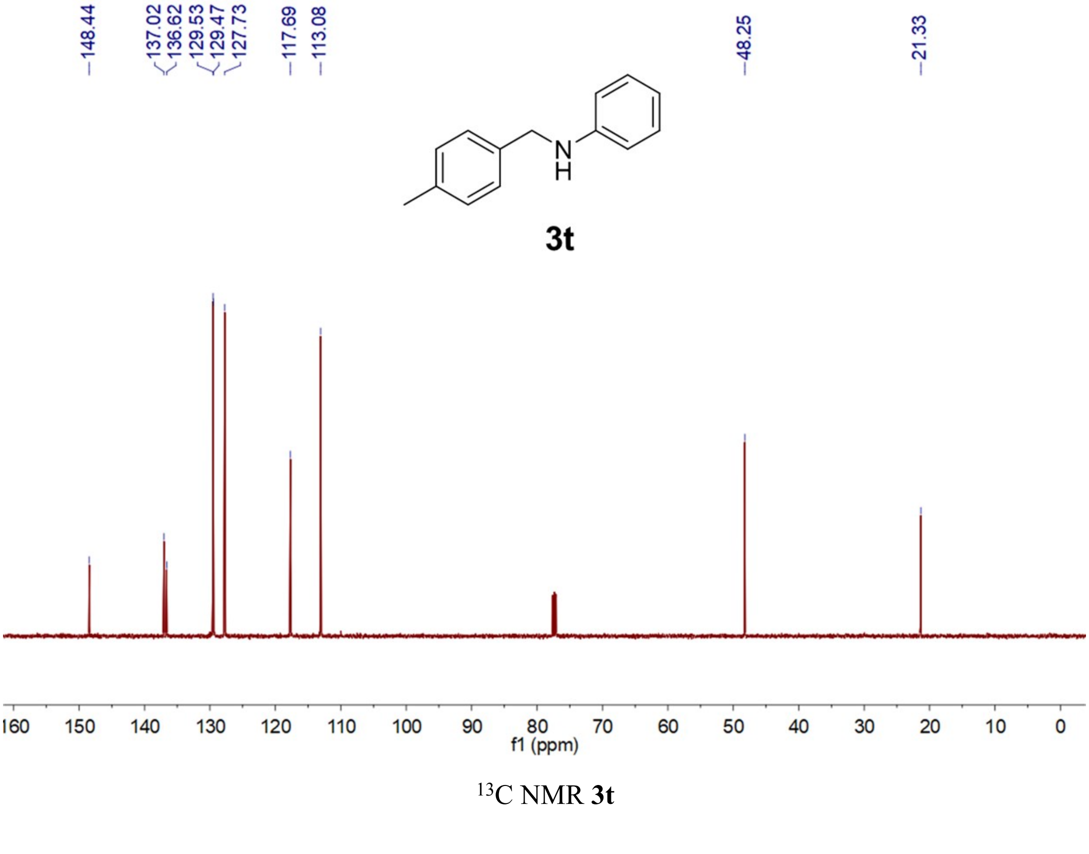
^{1}H NMR **3s**



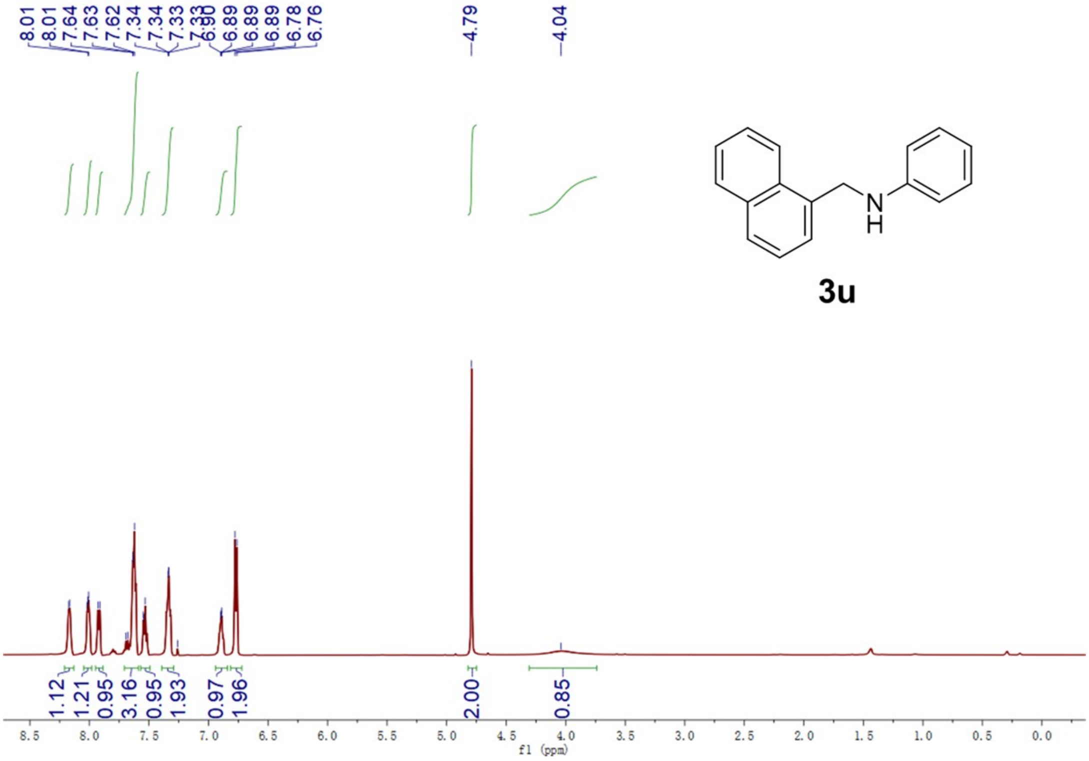
^{13}C NMR **3s**



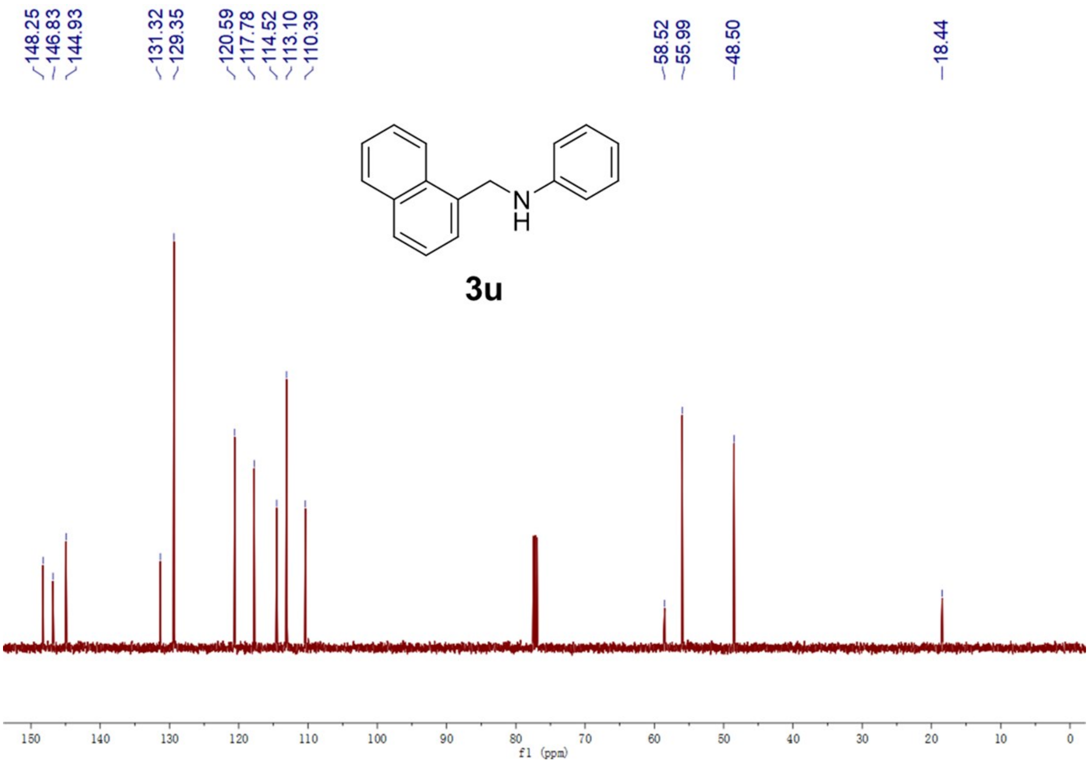
^{1}H NMR **3t**



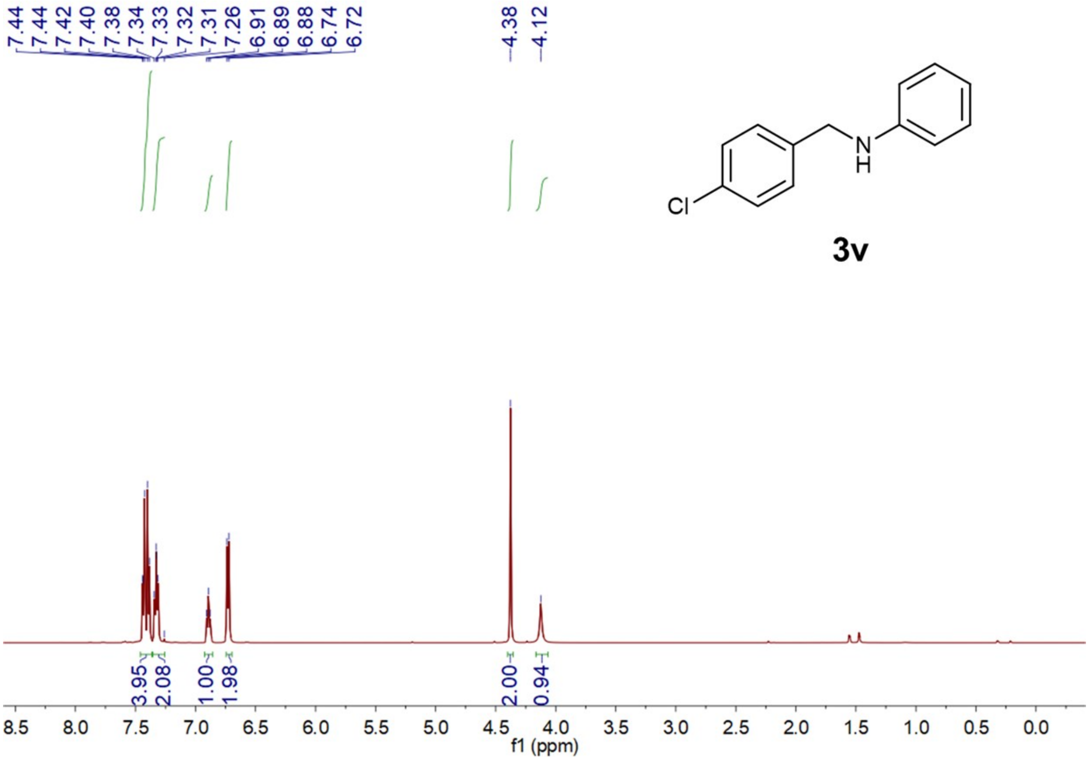
^{13}C NMR **3t**



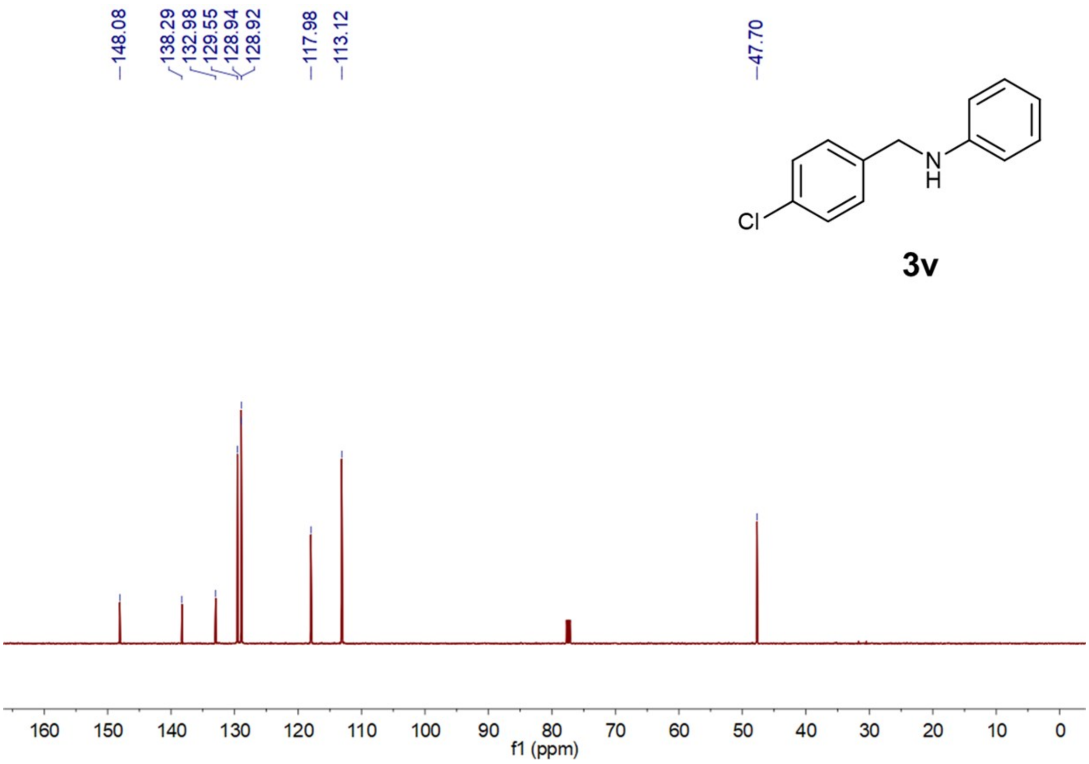
^{1}H NMR **3u**



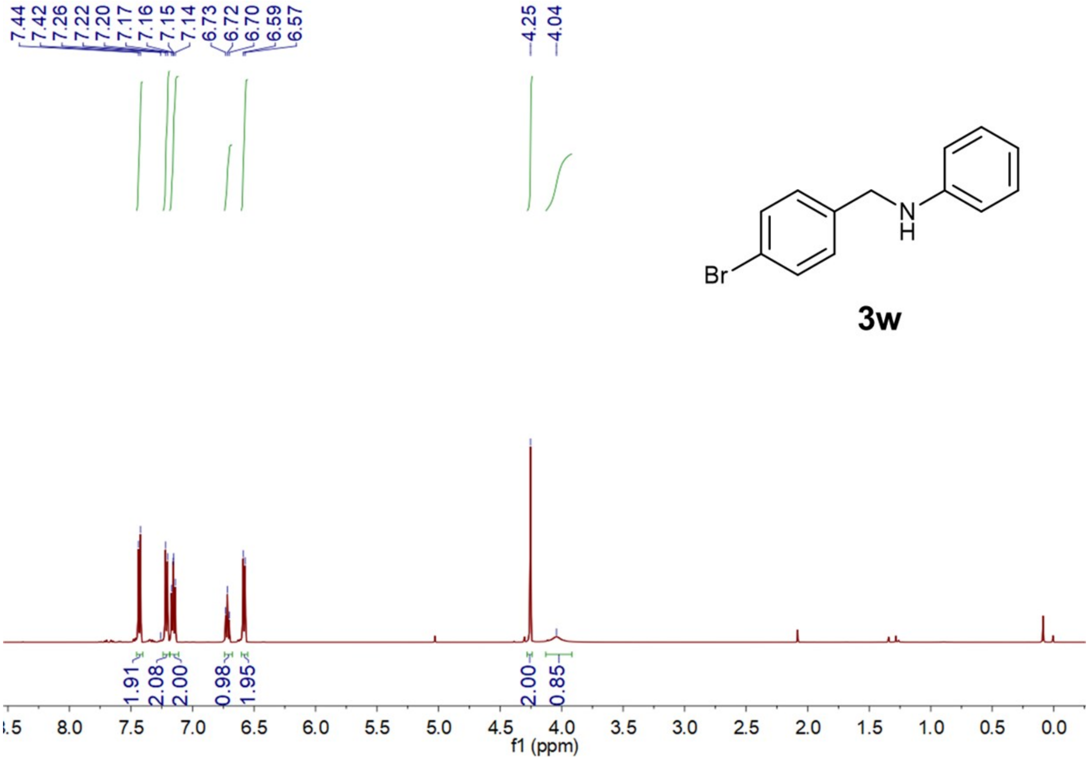
^{13}C NMR **3u**



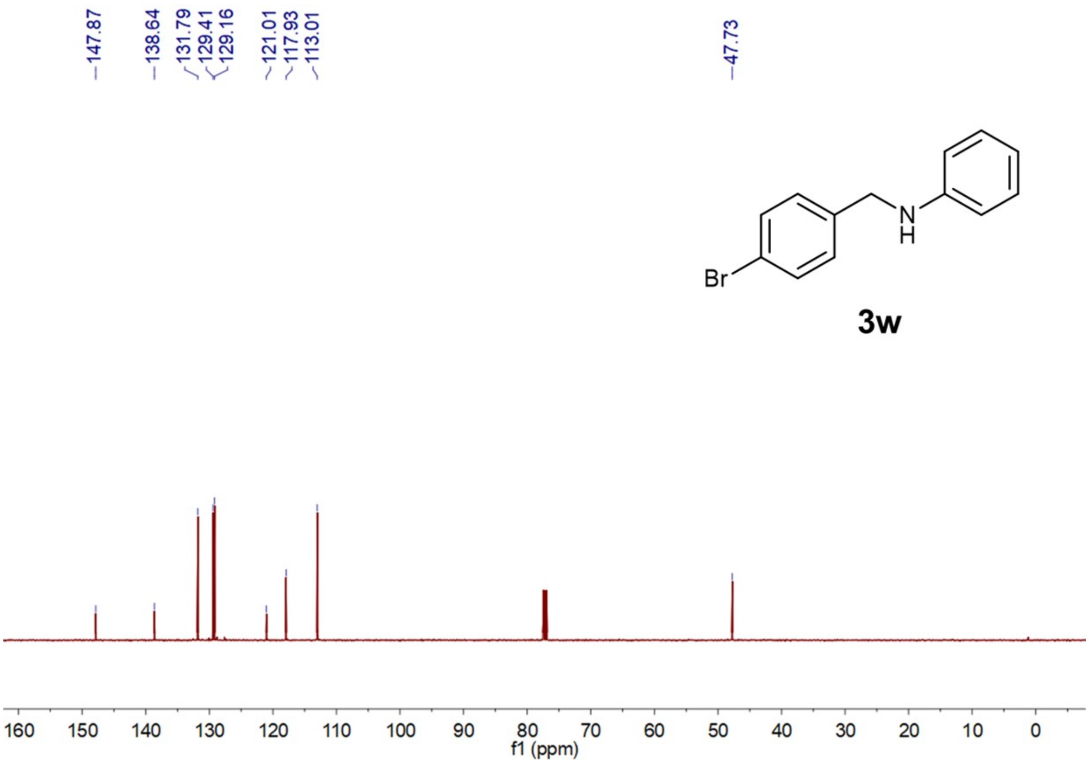
^{1}H NMR **3v**



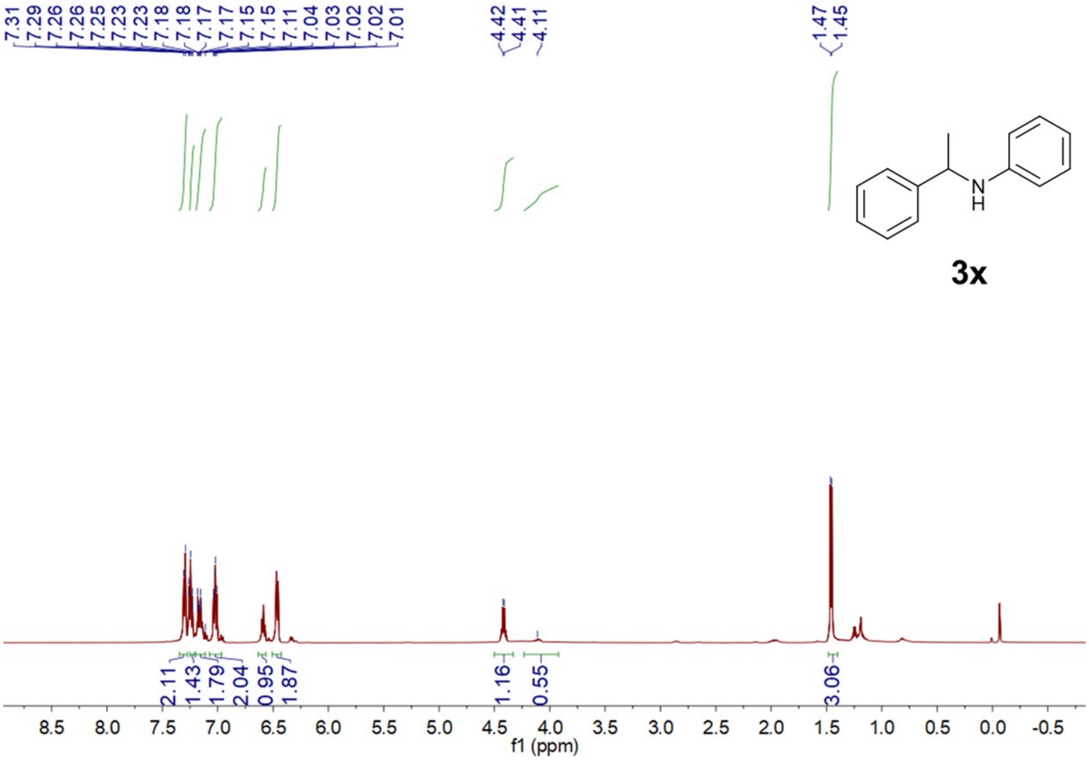
^{13}C NMR **3v**



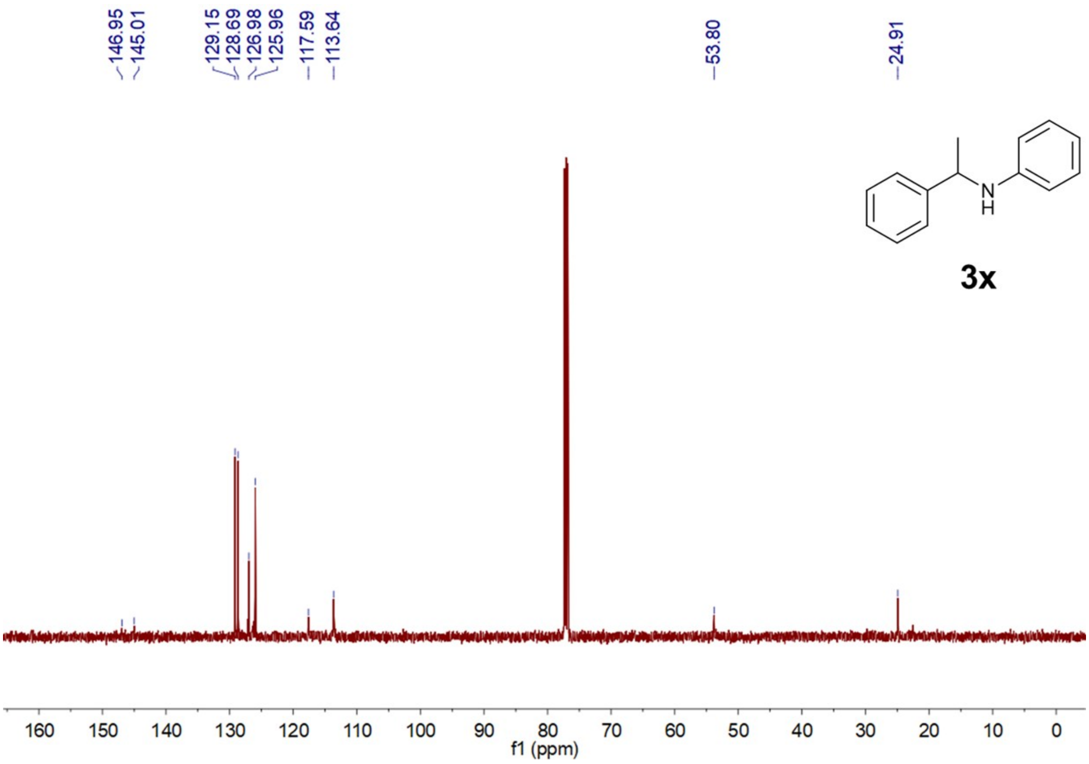
^{1}H NMR **3w**



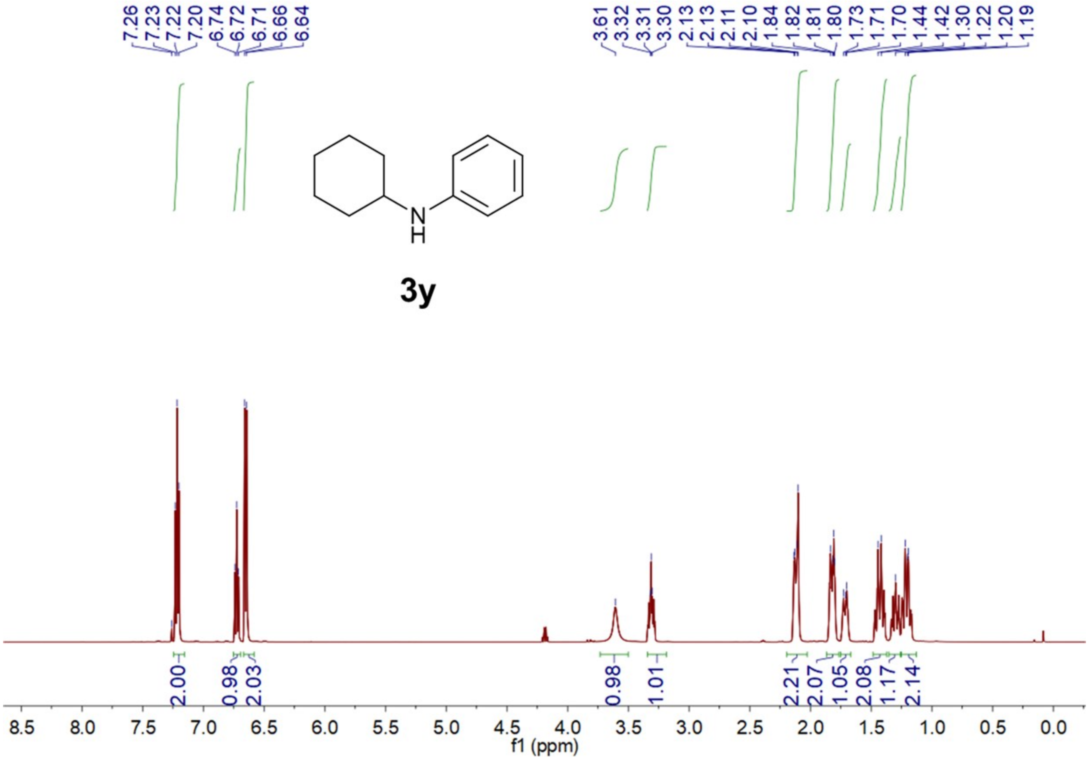
^{13}C NMR **3w**



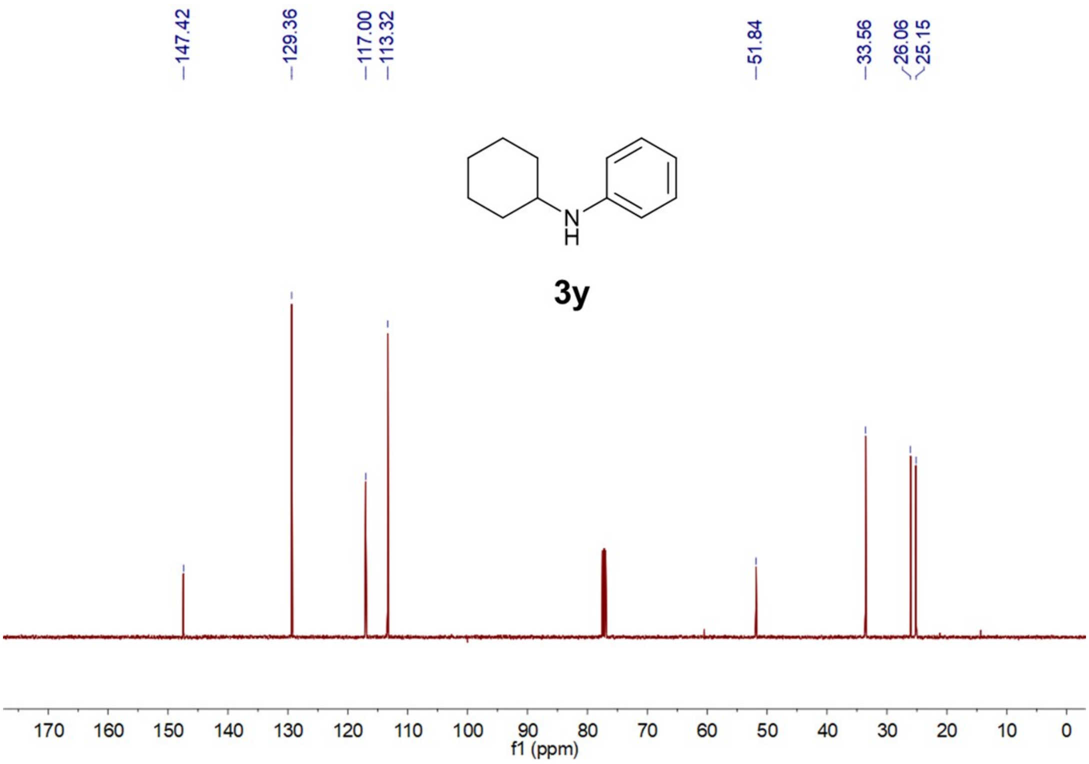
^{1}H NMR **3x**



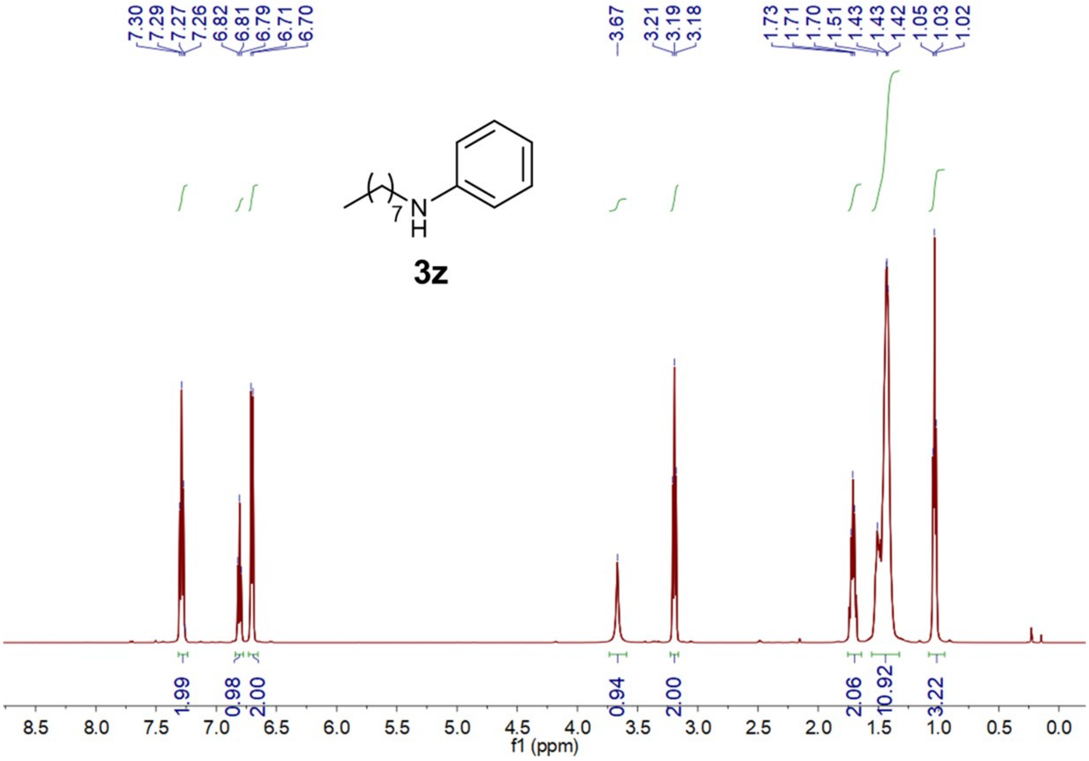
^{13}C NMR **3x**



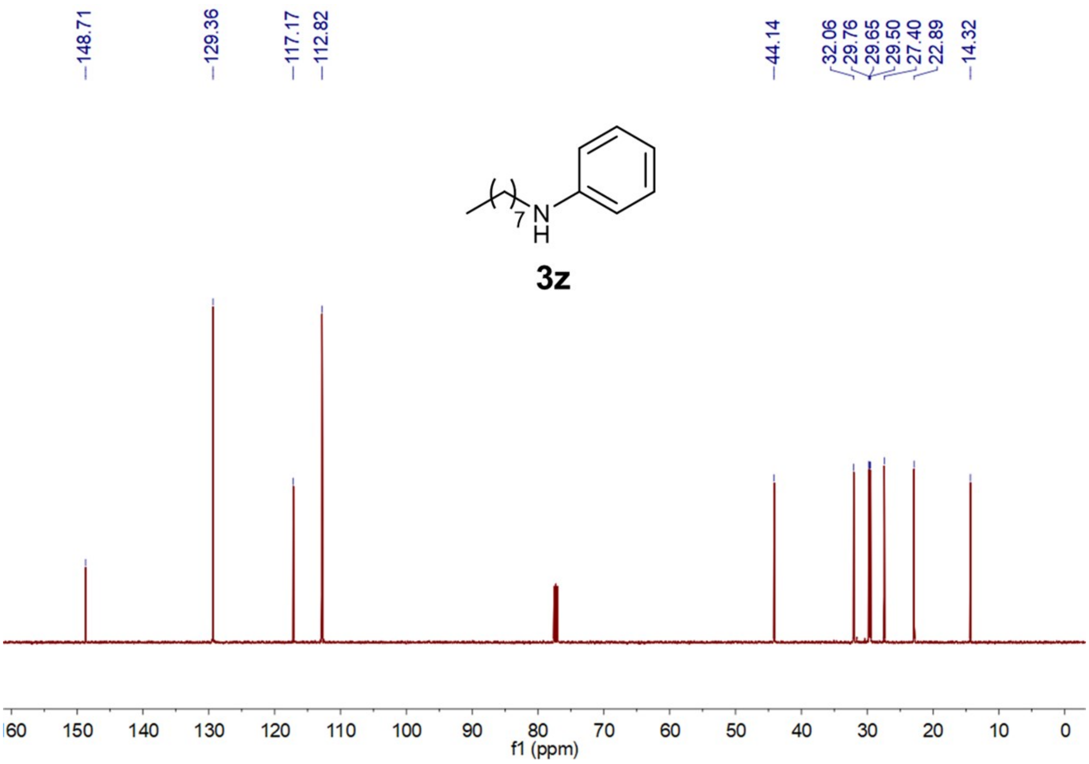
^{1}H NMR **3y**



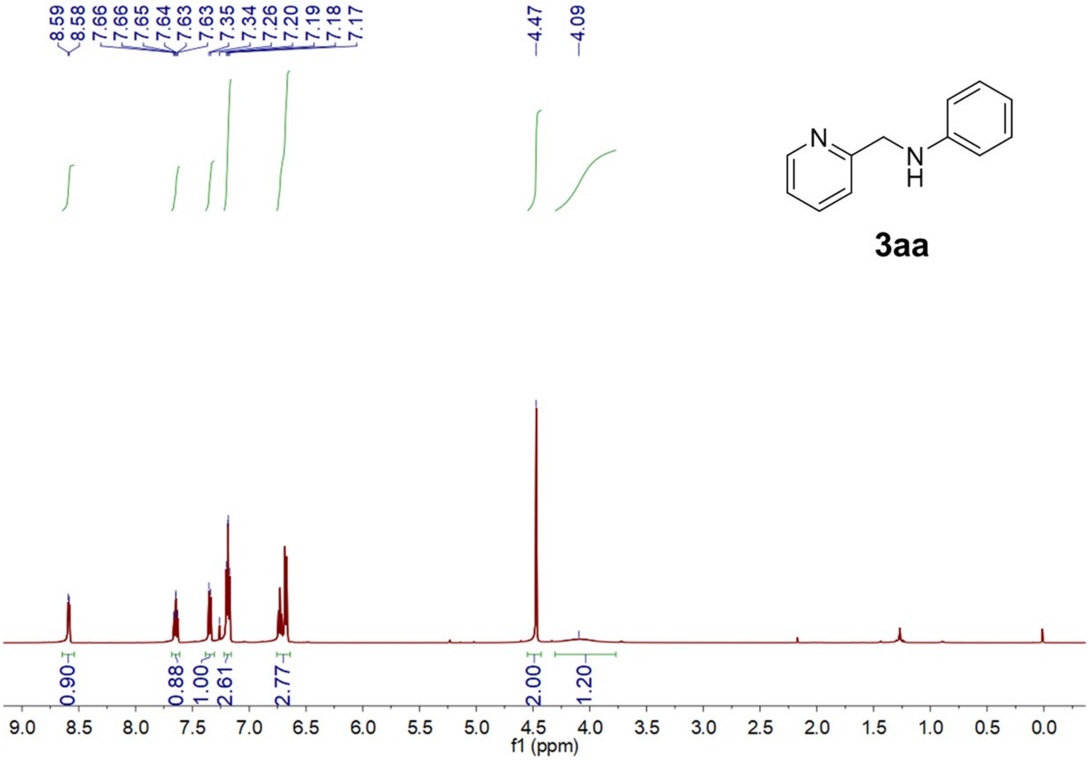
^{13}C NMR **3y**



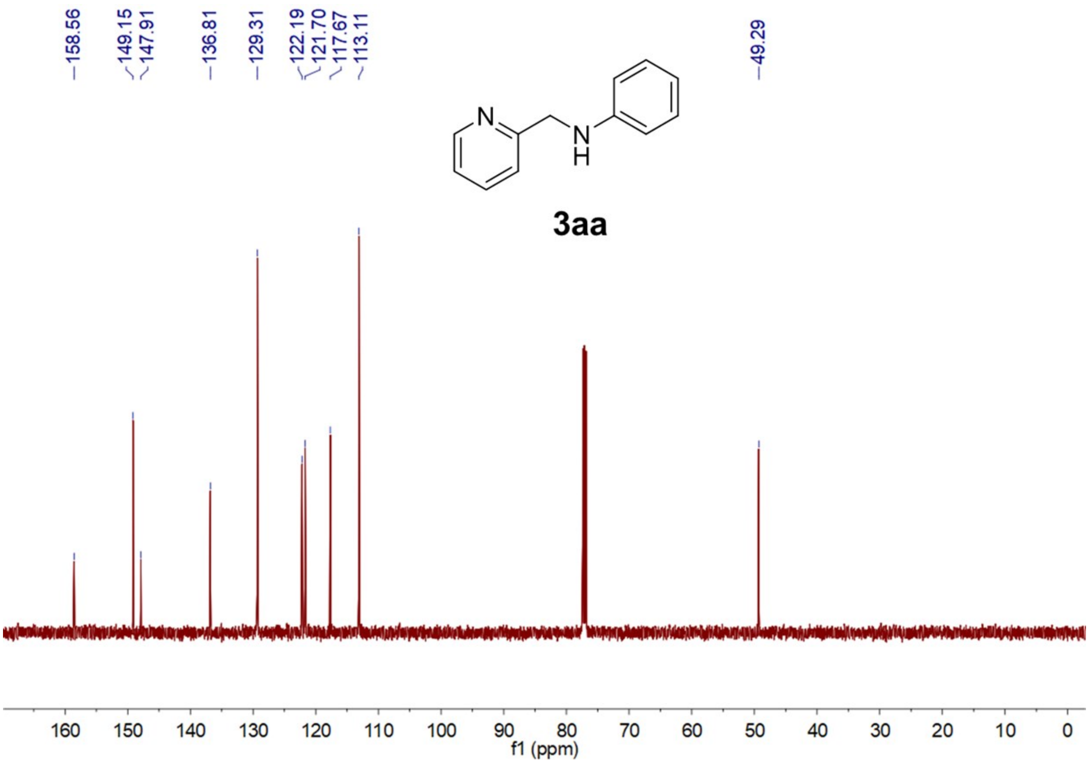
^{1}H NMR **3z**



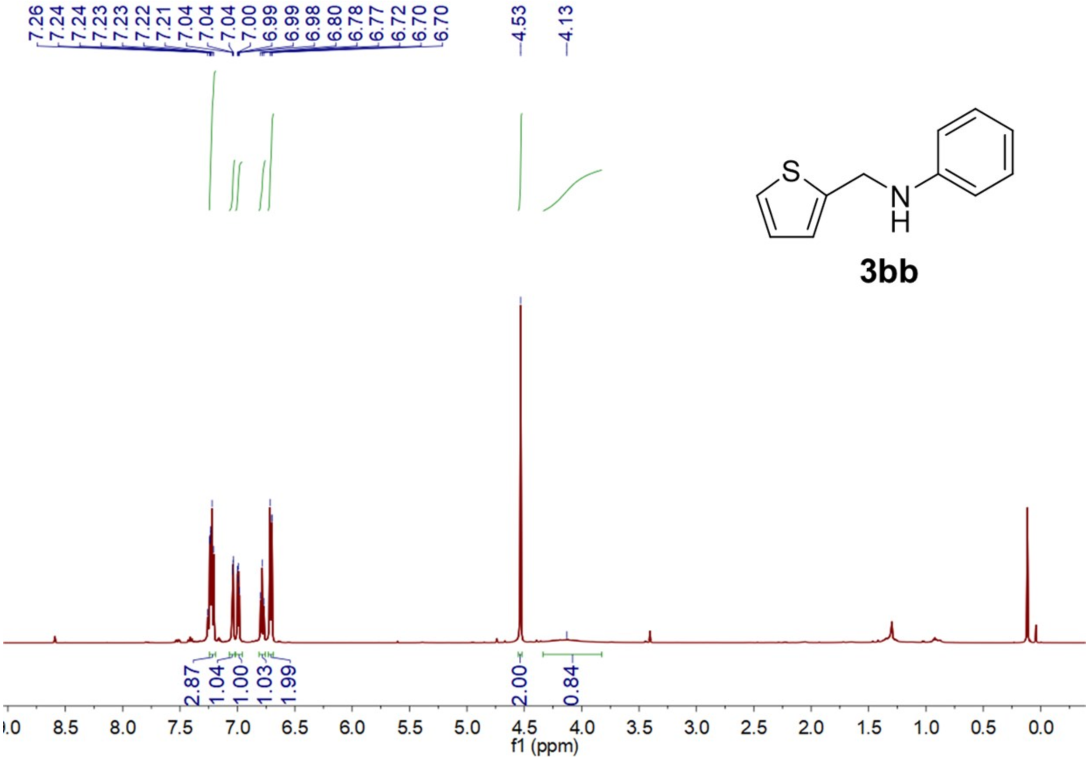
^{13}C NMR **3z**



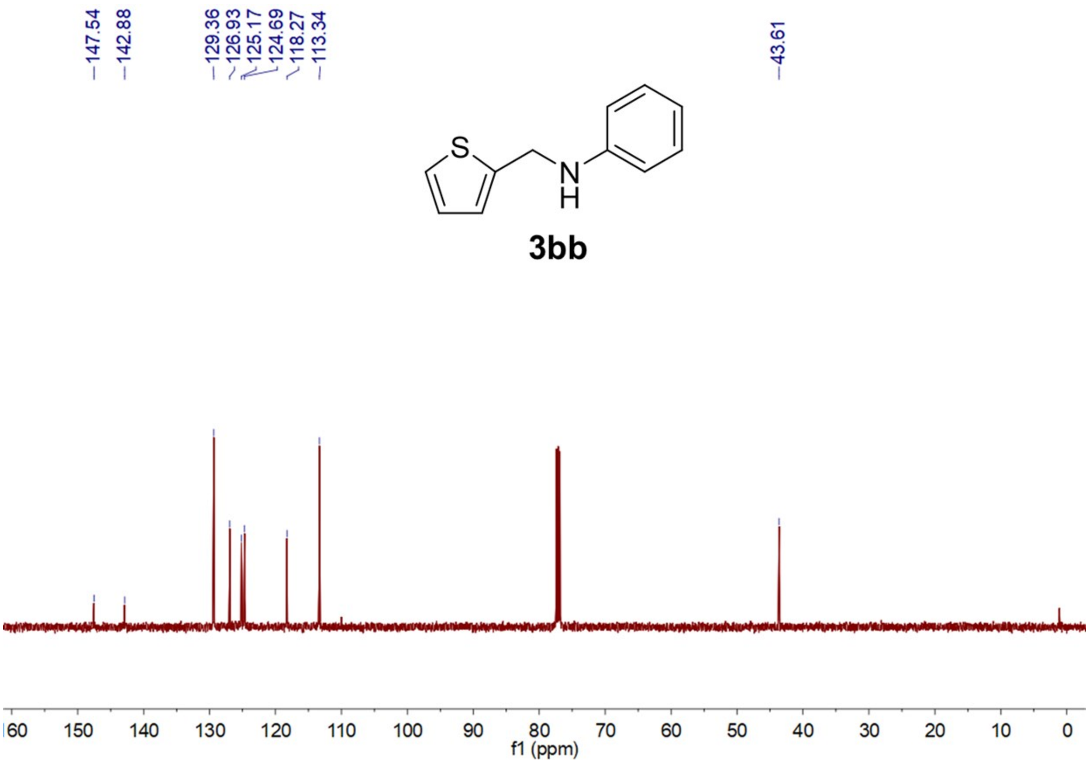
^{1}H NMR **3aa**



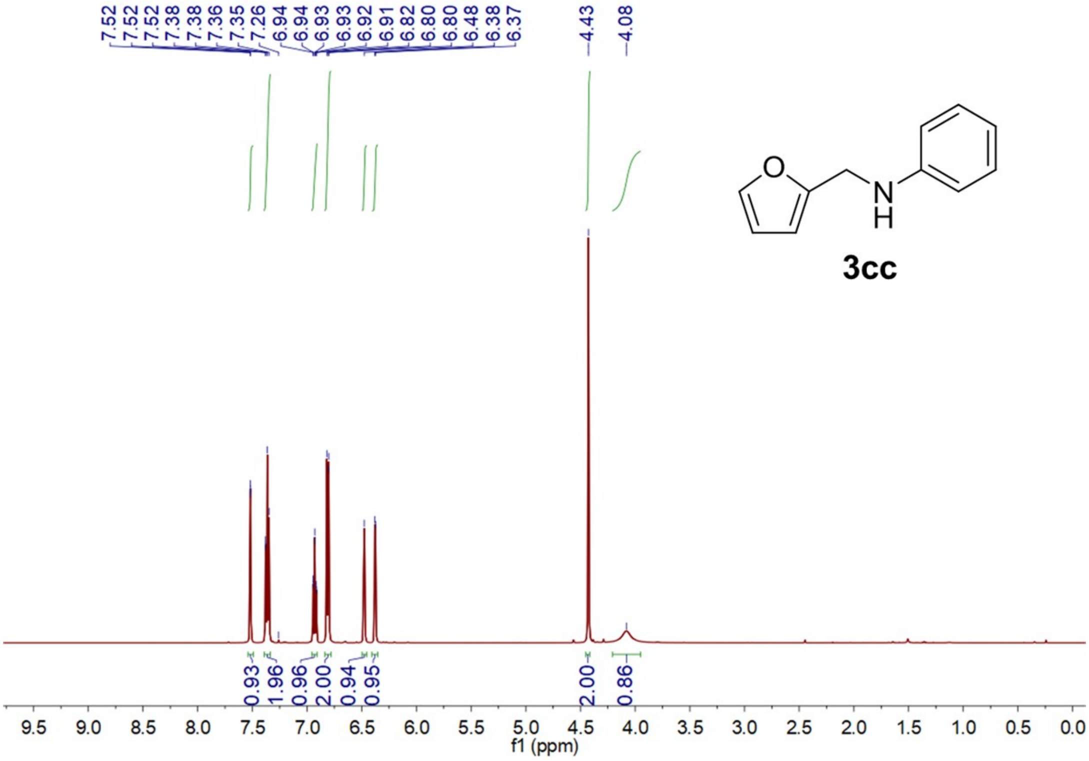
^{13}C NMR **3aa**



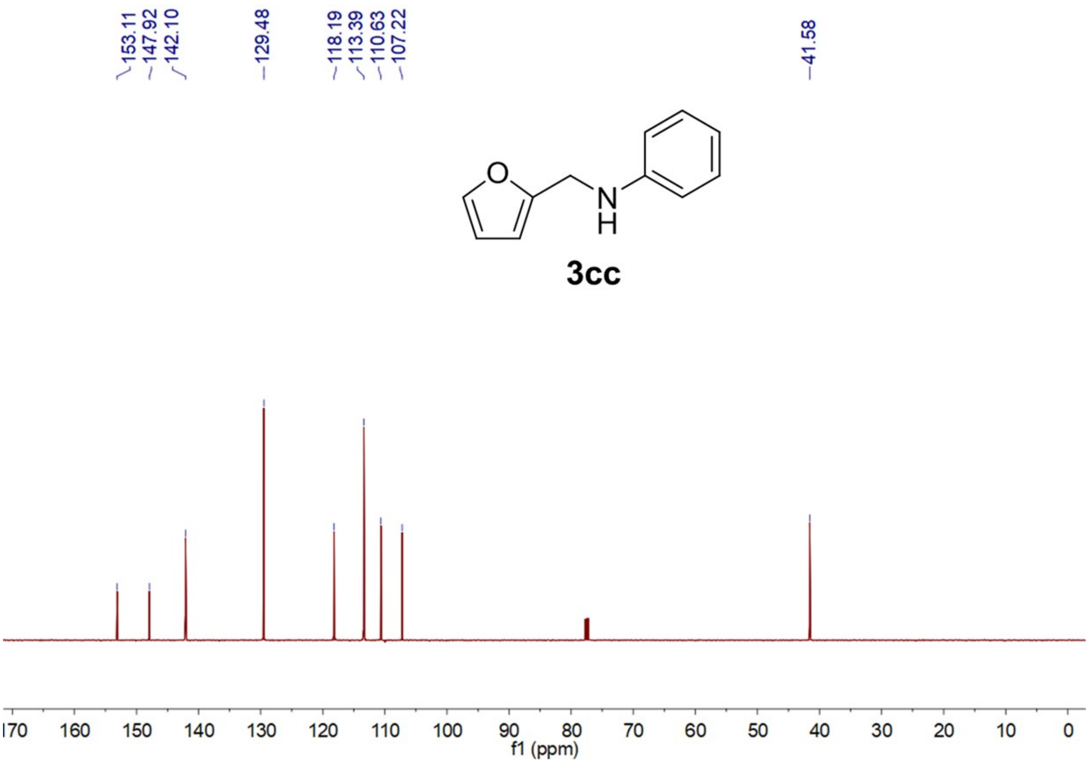
[1]H NMR **3bb**



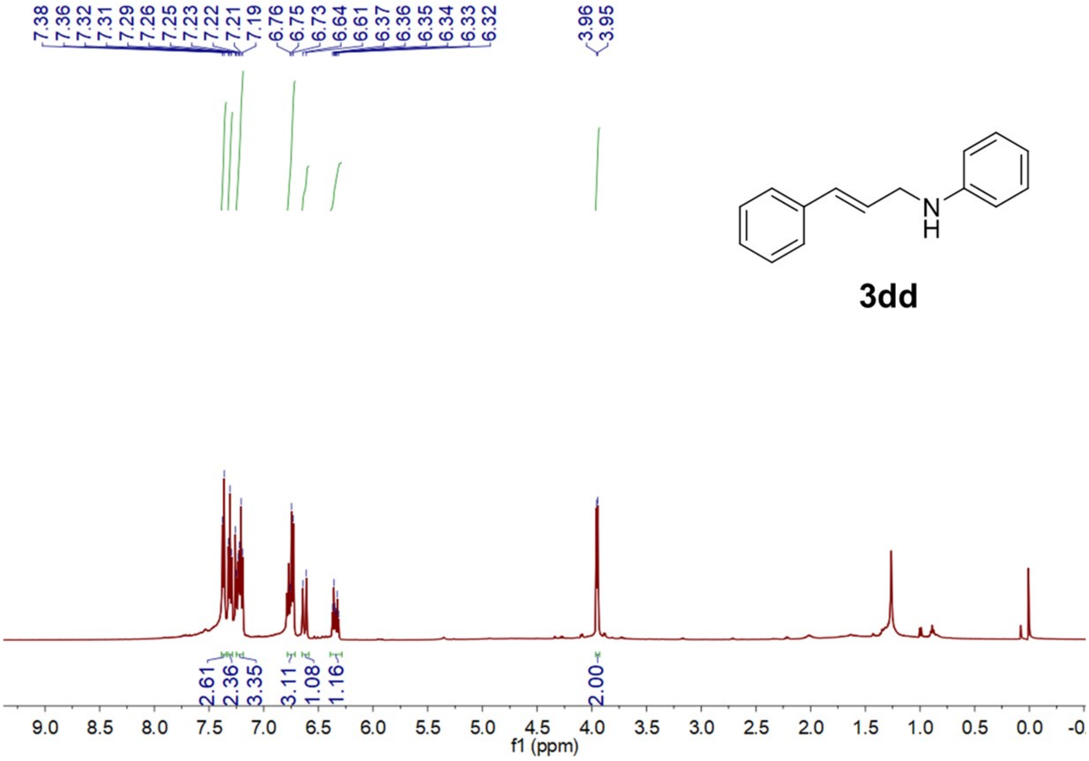
[13]C NMR **3bb**



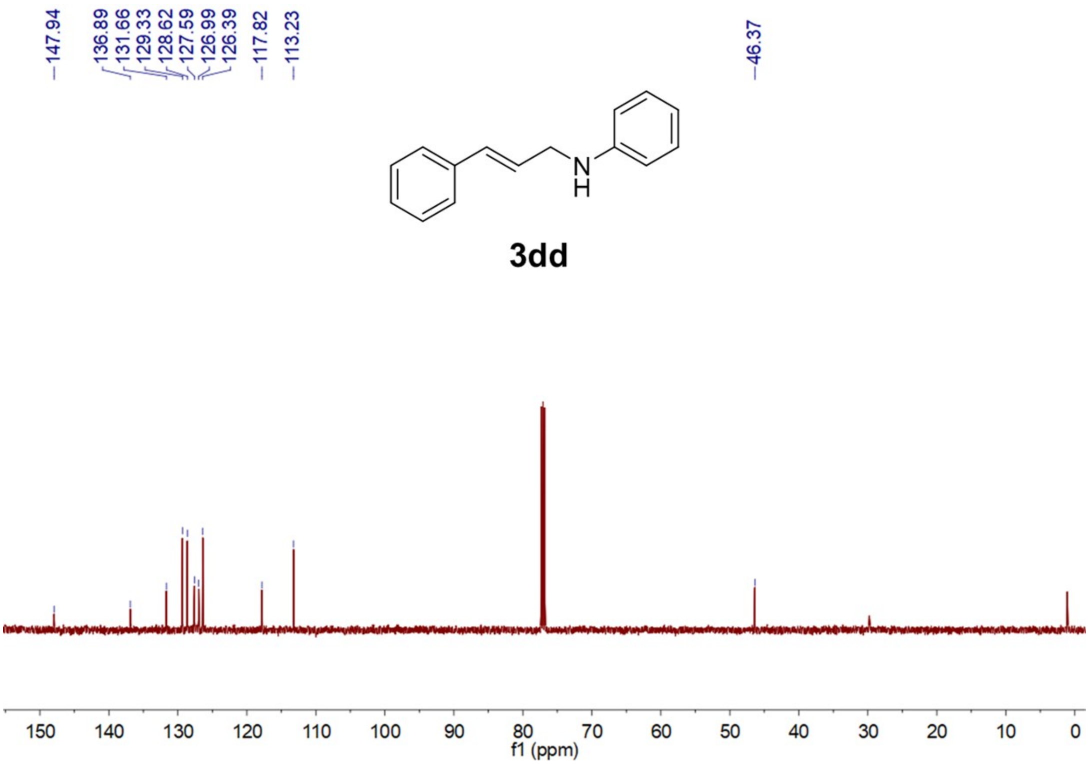
[1]H NMR **3cc**



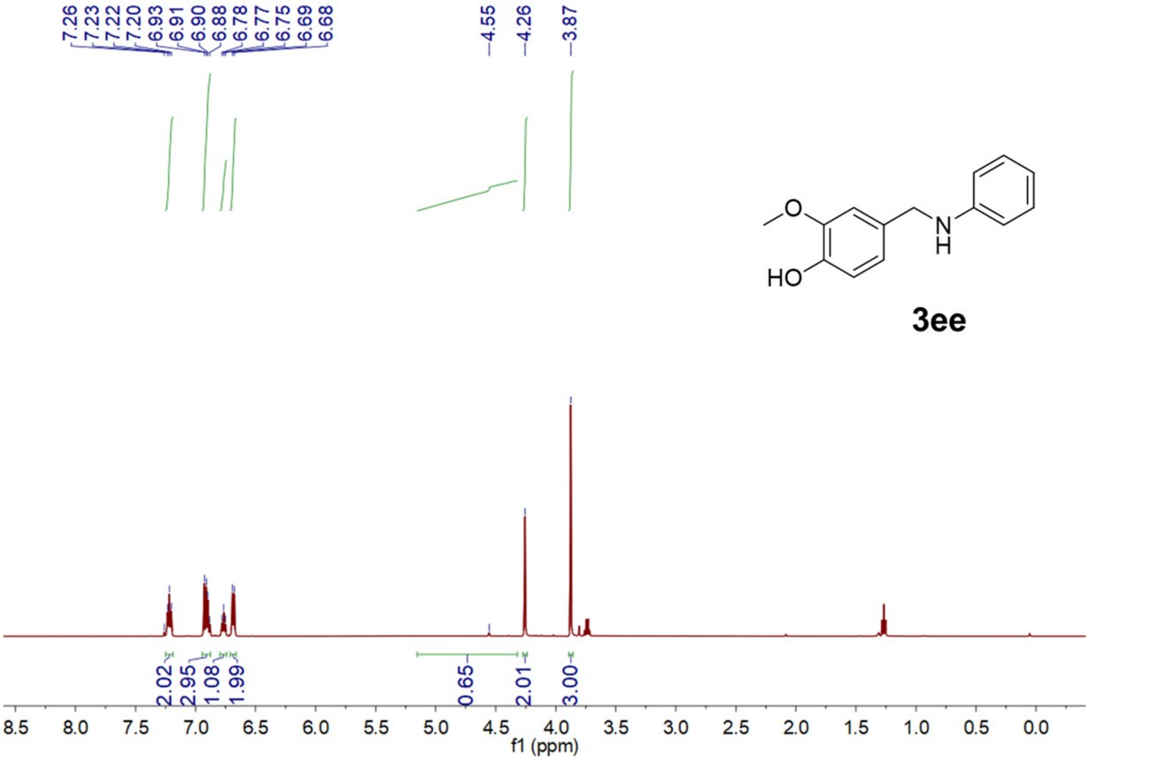
[13]C NMR **3cc**



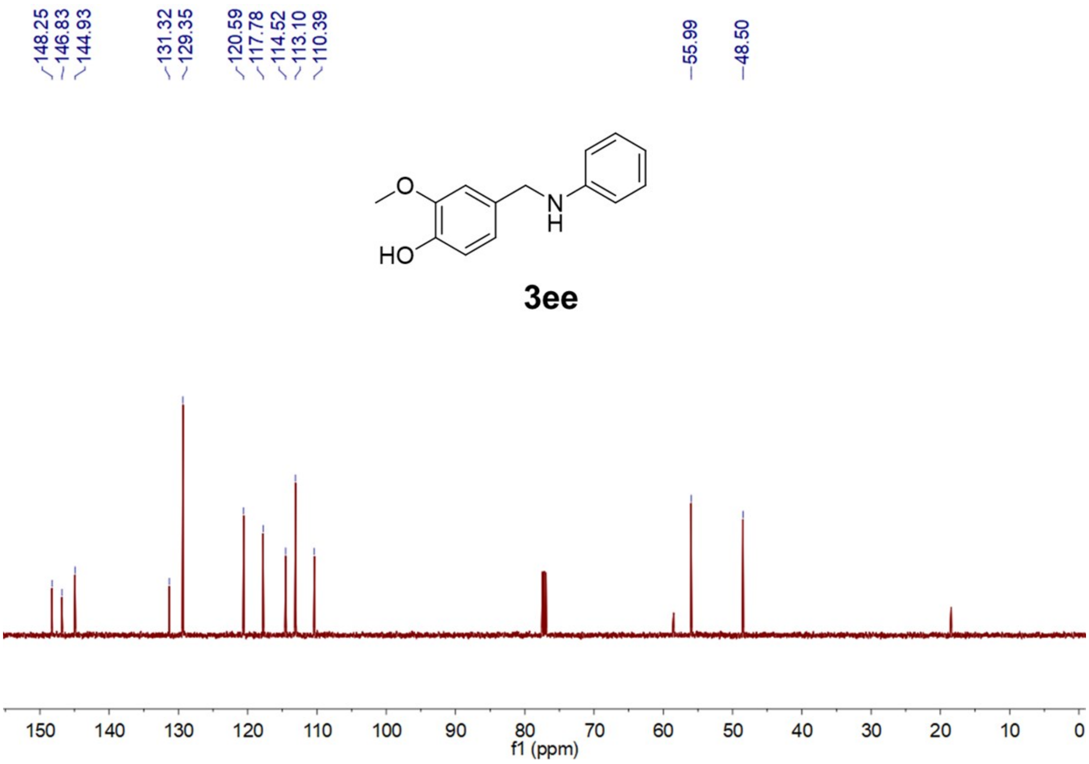
^{1}H NMR **3dd**



^{13}C NMR **3dd**



^{1}H NMR **3ee**



^{13}C NMR **3ee**

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