Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supplementary Information

for

Biomass-Derived N-doped Porous Carbon Catalyst for Aerobic Dehydrogenation of Nitrogen Heterocycles

Jing-Jiang Liu[a, b, ¶], Fu-Hu Guo[a, ¶, *], Fu-Jun Cui[b], Ji-Hua Zhu[c], Xiao-Yu Liu[a], Arif Ullah[a], Xi-Cun Wang[a, *], and Zheng-Jun Quan[a, *]

a Gansu International Scientific and Technological Cooperation Base of Water Retention Chemical Functional Materials, College of Chemistry and Chemical Engineering, Northwest Normal University, Lanzhou, Gansu 730070, People's Republic of China. E-mail: quanzhengjun@hotmail.com.

b School of Chemistry and Chemical Engineering, Pulp & Paper Engineering State Key Laboratory of China, South China University of Technology, Guangzhou 510640, PR China.

c College of Chemistry and Chemical Engineering, Qinghai Normal University, Xining, Qinghai, 810016, China.

¶ (J.L., F.G.) These authors contributed equally to this work.

Table of Contents S1

1. General Information

Column chromatography was performed on silica gel (200-300 mesh). ^{1}H NMR spectra were recorded in $CDCl_3$ at 400 or 600 MHz, and chemical shifts (ppm) were recorded using tetramethylsilane (TMS) as an internal reference standard. ^{13}C NMR spectra were recorded in $CDCl_3$ at 100 or 150 MHz. HR-MS was obtained using a Q-TOF or Q-Orbitrap instrument equipped with an ESI source. ^{1}H NMR and ^{13}C NMR of all compounds are provided in "Support Information". The room temperature is 20–30°C. Other commercially available reagents and solvents can be used without further purification. The tetrahydroquinoline derivatives **1c&1k**,[1] **1i-1j**,[2] **1l-1t**,[3] were prepared by the same procedure in the corresponding literature. The tetrahydroquinazoline derivatives **5a-5e** are prepared from 2-aminobenzylamine and the corresponding aldehyde.[4] Tetrahydroquinoxaline and its derivatives **7a**[5] and **7b**[6] are obtained by reacting o-phenylenediamine with 1,2-dibromoethane or 1,3-butanedione.The indoline derivative **9d** was prepared according to the literature.[1] Dihydrobenzothiazole derivatives **11a-11d** are synthesized from 2-aminothiophenol and the corresponding aldehyde according to the literature.[7] 4-Substituted Hans Ester **13b-13e** is synthesized by the corresponding literature.[8]

2. Catalyst preparation results

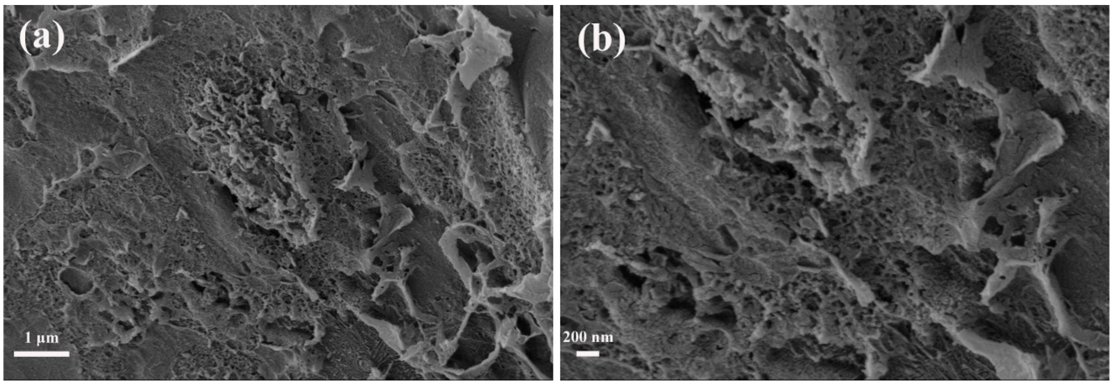
Table S1. Detailed laboratory preparation of biomass-based carbon materials.

Sample	Sugarcane bagasse (g)	Calcium salt（g）	Urea (g)	Calcium salt	Carbonization Temperature (℃)	Yield (%)
NC(100-800)	1	0	0	$CaCl_2$	800	22
NC(102-800)	1	0	2	$CaCl_2$	800	33
NC(121-800)	1	2	1	$CaCl_2$	800	34
NC(242-800)	2	4	2	$CaCl_2$	800	37
NC(363-800)	3	6	3	$CaCl_2$	800	46
NC(484-800)	4	8	4	$CaCl_2$	800	42
NC(242-800)[a]	2	4	2	$CaSO_4$•$2H_2O$	800	33
NC(242-800)[b]	2	4	2	$Ca(H_2PO_4)_2$•H_2O	800	93
NC(242-800)[c]	2	4	2	$CaHPO_4$	800	30
NC(242-800)[d]	2	4	2	$C_{12}H_{22}CaO_{14}$•H_2O	800	80
NC(242-600)	2	4	2	$CaCl_2$	600	38
NC(242-700)	2	4	2	$CaCl_2$	700	34
NC(242-900)	2	4	2	$CaCl_2$	900	48
NC(242-1000)	2	4	2	$CaCl_2$	1000	40

The superscript of NC represents the different calcium salt (“a”, “b”, “c” and “d” stands for $CaSO_4$•$2H_2O$, $Ca(H_2PO_4)_2$•H_2O, $CaHPO_4$ and $C_{12}H_{22}CaO_{14}$•H_2O, respectively).

3. Physical characterization of the NC

The morphology of NC was studied with field emission scanning electron microscopy (FE-SEM, Ultra Plus, Carl Zeiss, Germany) and transmission electron microscopy (TEM, JEM-2100, Japan). Nitrogen absorption/desorpotion measurements were performed at -196 °C using an ASAP 2020 system (Micrometitics, USA). The Raman spectra were obtained using a Bio-Rad FTS6000 Raman spectrophotometer with a 532 nm blue laser beam. X-ray diffraction (XRD) patterns of samples were examined on a diffractomter (D/Max-2400, Rigaku, Japan) advance instrument using Cu Kα radiation (□□=1.5418 Å) at 40 kV, 100 mA. The 2θ range used in the measurements was from 5 to 80^{o}. The N species were characterized by X-ray photoelectron spectroscopy (XPS, Escalab 210, Germany).

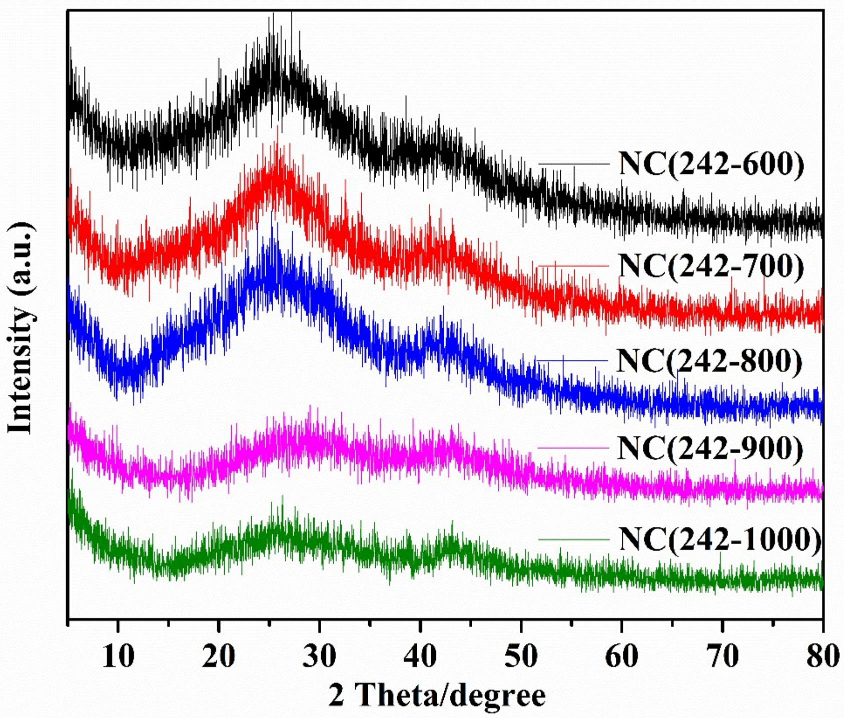


Scheme S1. SEM images of NC(242-800).

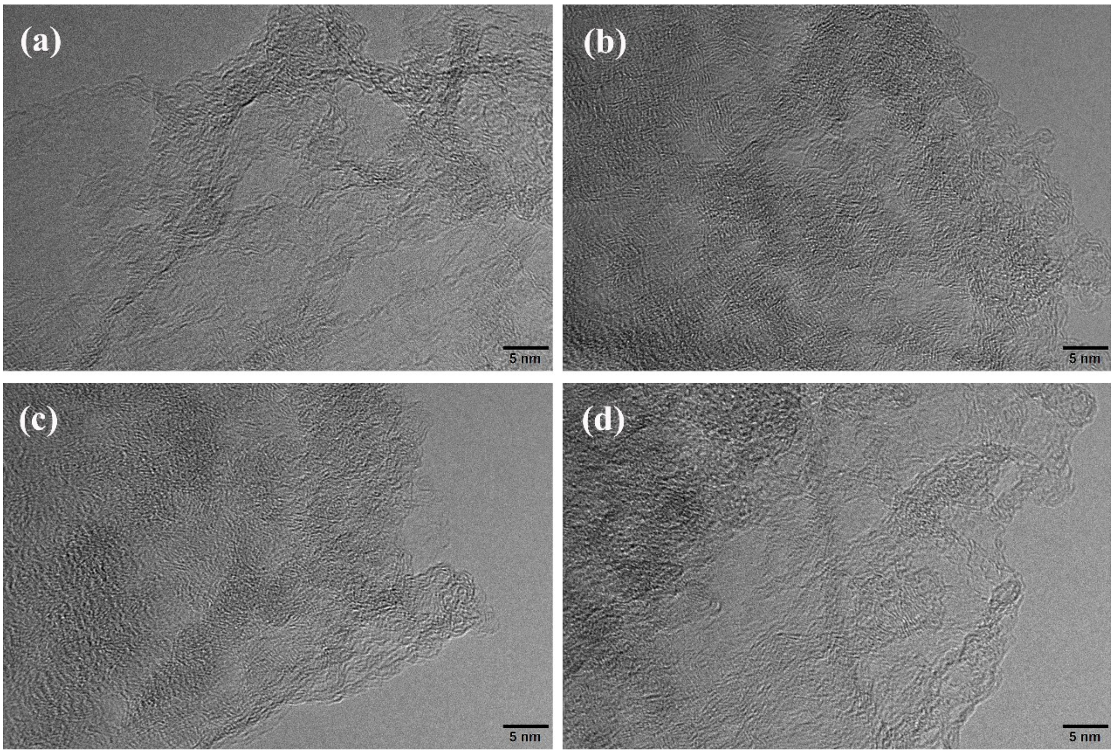
Table S2. ICP-OES analyses of NC(242-800).

Element	Solution concentration (ppm)	SD	%RSD	Int.(c/s)	Calculation concentration (ppm)
Ag 328.068	0.001054uv	0.003073	291.6	26.2910	0.000051
Al 167.019	0.093081uv	0.108998	117.1	30.8710	0.004475
As 188.980	-0.371441uv	0.387727	104.4	18.2352	-0.017858
Au 242.794	0.121253	0.108165	89.2	50.9389	0.005829
B 249.772	-0.393865uv	0.005844	1.5	72.2010	-0.018936
Ba 455.403	0.000140	0.000064	45.4	199.727	0.000007
Be 313.042	0.000410	0.000106	25.8	136.774	0.000020
Bi 223.061	-0.103836uv	0.106597	102.7	26.3413	-0.004992
Ca 422.673	99.5922x	1.26459	1.3	354121	4.78809
Cd 214.439	-0.032499uv	0.006421	19.8	17.7963	-0.001562
Ce 418.659	-0.003352uv	0.006279	187.4	60.8366	-0.000161
Co 238.892	0.006619uv	0.061985	936.4	45.0043	0.000318
Cr 267.716	-0.004903uv	0.000062	1.3	17.8287	-0.000236
Cs 672.328	89.8846x	1.52178	1.7	3678.85	4.32138
Cu 327.395	0.006348	0.004767	75.1	45.5447	0.000305
Dy 353.171	0.001507uv	0.002557	169.7	37.2377	0.000072
Er 349.910	0.000688 uv	0.001272	184.9	33.3700	0.000033
Eu 420.504	-0.002228uv	0.000048	2.1	35.7104	-0.000107
Fe 238.204	0.050345	0.020430	40.6	138.644	0.002420
Ga 294.363	0.010040	0.005808	57.8	43.4212	0.000483
Gd 342.246	-0.000504uv	0.002212	438.7	32.1195	-0.000024
Ge 209.426	-0.067982uv	0.355811	523.4	21.8757	-0.003268
Hf 264.141	-0.022904uv	0.008611	37.6	27.7476	-0.001101
Hg 184.887	-0.051016uv	0.013620	26.7	8.88573	-0.002453
Ho 345.600	-0.003883uv	0.006868	176.9	9.72666	-0.000187
In 230.606	0.132360	0.080312	60.7	27.4291	0.006363
Ir 224.268	0.064231uv	0.204623	318.6	20.4520	0.003088
K 766.491	3.23276	0.014630	0.5	2163.72	0.155421
La 333.749	0.025325	0.008267	32.6	262.148	0.001218
Li 670.783	0.036978	0.000930	2.5	2499.73	0.001778
Lu 261.541	-0.002024uv	0.000686	33.9	34.3167	-0.000097
Mg 279.553	0.005195	0.000346	6.7	241.135	0.000250
Mn 257.610	-0.001297uv	0.001042	80.3	40.8244	-0.000062
Mo 202.032	0.020865uv	0.042723	204.8	29.3588	0.001003
Na 589.592	32.7114x	0.282986	0.9	171304	1.57267
Nb 313.078	-0.001528uv	0.006544	428.4	63.3400	-0.000073
Nd 401.224	0.013658	0.016252	119.0	48.4489	0.000657
Ni 231.604	0.006818uv	0.042142	618.1	15.8719	0.000328
P 213.618	0.037003	0.179025	483.8	26.2880	0.001779

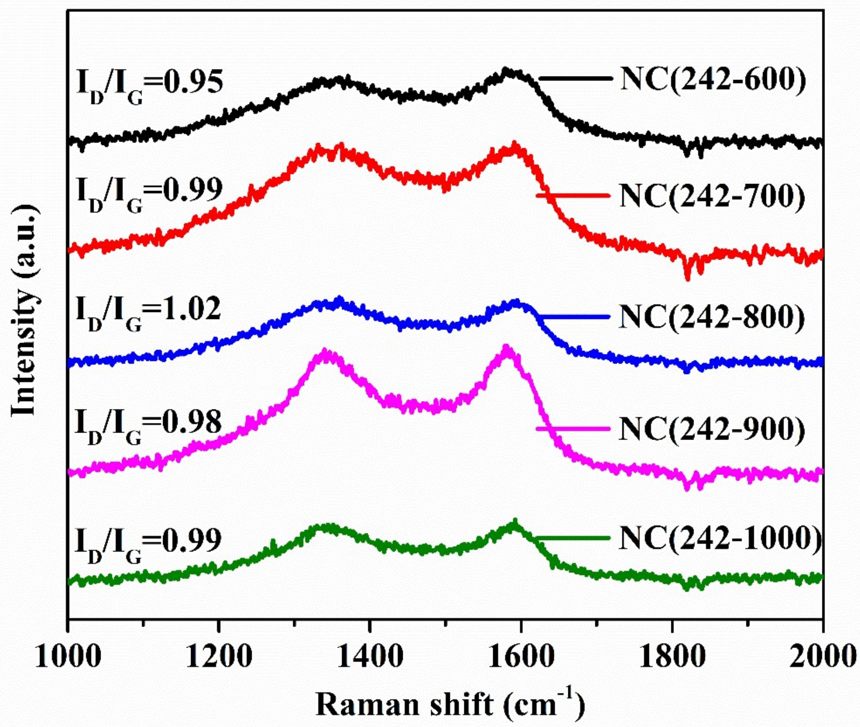
Pb 220.353	0.169701uv	0.207796	122.4	59.0922	0.008159
Pd 340.458	0.005895uv	0.010221	173.4	37.9725	0.000283
Pr 417.939	-0.020550uv	0.006317	30.7	34.2896	-0.000988
Pt 214.424	-0.156322uv	0.077958	49.9	5.68232	-0.007515
Rb 780.026	0.525713	0.007875	1.5	191.783	0.025275
Re 227.525	0.118879uv	0.148702	125.1	47.9795	0.005715
Rh 343.488	-0.012488uv	0.017520	140.3	23.5594	-0.000600
Ru 267.876	0.028341	0.006965	24.6	24.9424	0.001363
S 181.972	3.40565	0.883752	25.9	167.437	0.163733
Sb 206.834	-0.117039uv	0.072784	62.2	30.1222	-0.005627
Sc 361.383	0.000508	0.000490	96.4	34.5789	0.000024
Se 196.026	0.333303uv	0.491030	147.3	35.9509	0.016024
Si 251.611	0.078045uv	0.347800	445.6	67.4754	0.003752
Sm 359.259	-0.002885uv	0.006497	225.2	22.8420	-0.000139
Sn 189.925	-1.22860uv	0.099941	8.1	29.0405	-0.059067
Sr 407.771	0.001042	0.000036	3.4	724.206	0.000050
Ta 268.517	0.021344uv	0.048545	227.4	26.4341	0.001026
Tb 350.914	-0.005245uv	0.006467	123.3	34.1211	-0.000252
Te 214.282	-0.376858uv	0.063812	16.9	17.3739	-0.018118
Th 283.730	0.027519	0.020082	73.0	35.4283	0.001323
Ti 336.122	0.013428	0.003421	25.5	177.308	0.000646
Tl 190.794	-6.20605uv	2.25729	36.4	5.73290	-0.298368
Tm 313.125	0.000584uv	0.003084	528.4	68.0261	0.000028
U 385.957	0.142669	0.009534	6.7	82.8445	0.006859
V 292.401	0.002024	0.001673	82.6	30.0406	0.000097
W 207.912	-0.119432uv	0.072522	60.7	20.2929	-0.005742
Y 371.029	0.000965uv	0.003609	373.8	85.5237	0.000046
Yb 328.937	0.000403	0.000434	107.8	33.2388	0.000019
Zn 213.857	0.046962	0.004103	8.7	151.695	0.002258
Zr 343.823	0.003645	0.001416	38.9	77.9197	0.000175



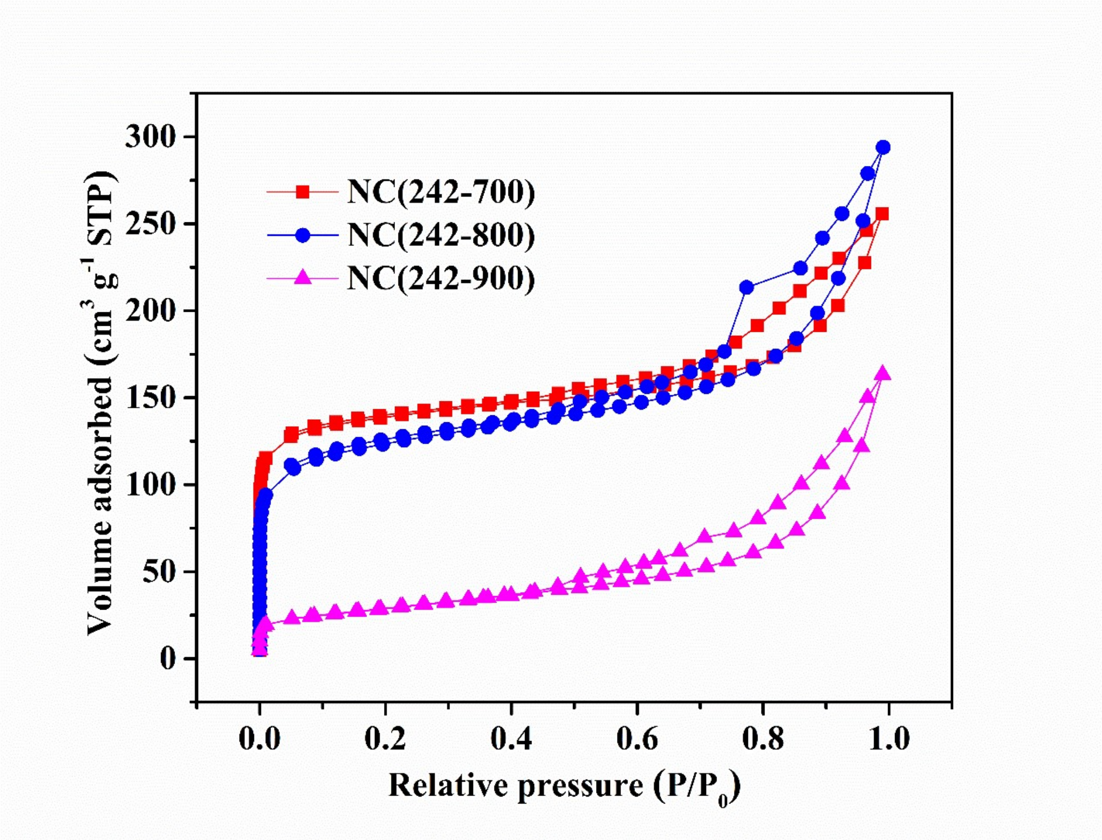
Scheme S2. XRD patterns of NC(242-600), NC(242-700), NC(242-800), NC(242-900) and NC(242-1000).



Scheme S3. HR-TEM images of NC(242-800).



Scheme S4. Raman spectra results of the NC(242-600), NC(242-700), NC(242-800), NC(242-900) and NC(242-1000).



Scheme S5. N_2 adsorption/desorption isotherms of NC(242-700), NC(242-800), and NC(242-900).

Table S3

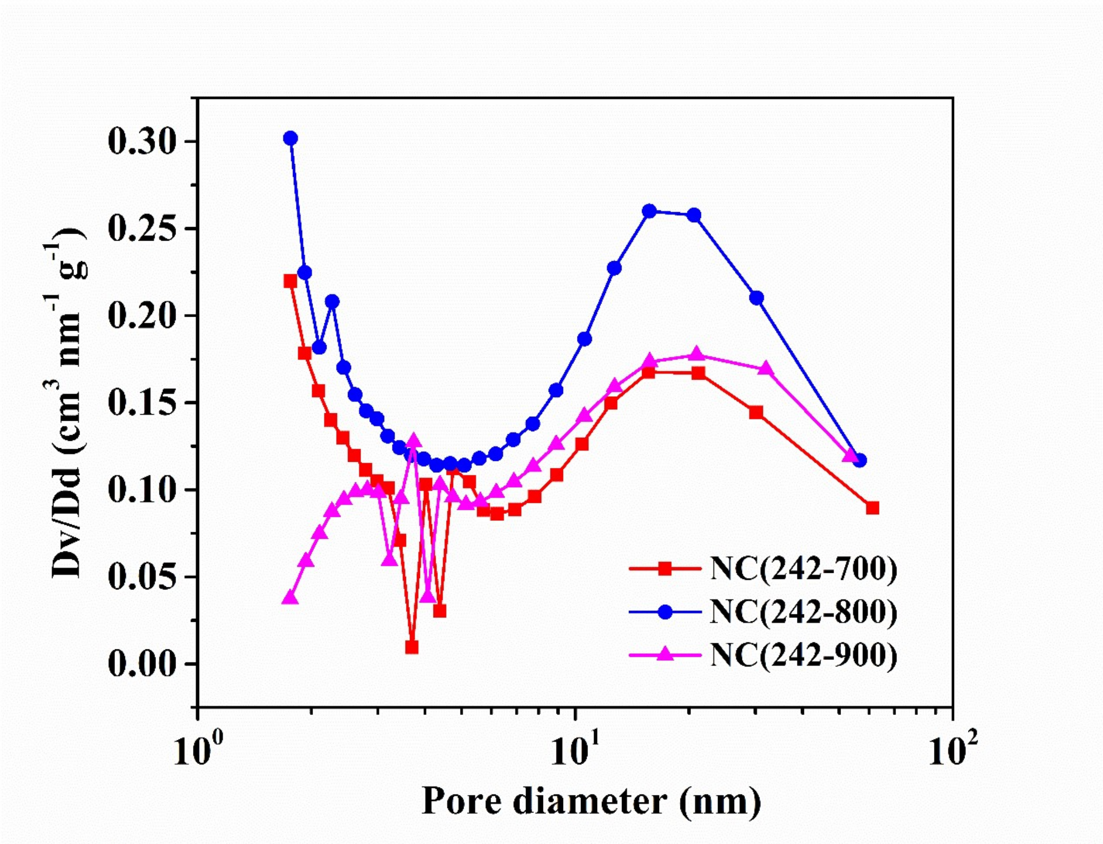
Samples	S_{BET}[a] (m^2 g^{-1})	S_{mic}[b] (m^2 g^{-1})	V_{total}[c] (cm^3 g^{-1})	D[d] (nm)
NC(242-700)	433.27	313.08	0.40	3.65
NC(242-800)	393.17	229.05	0.45	4.63
NC(242-900)	100.67	13.12	0.25	10.04

a) Specific surface area determined according to BET (Brunauer-Emmett-Teller) method.

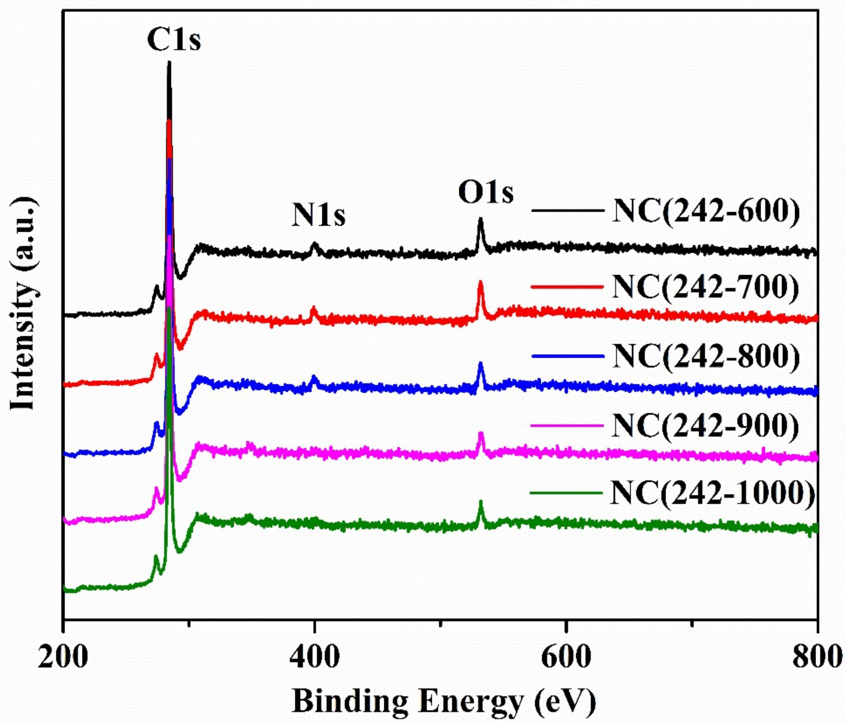
b) Micropore surface area from T-plot method.

c) Total pore volume (Single point adsorption total pore volume of pores).

d) Average pore width.



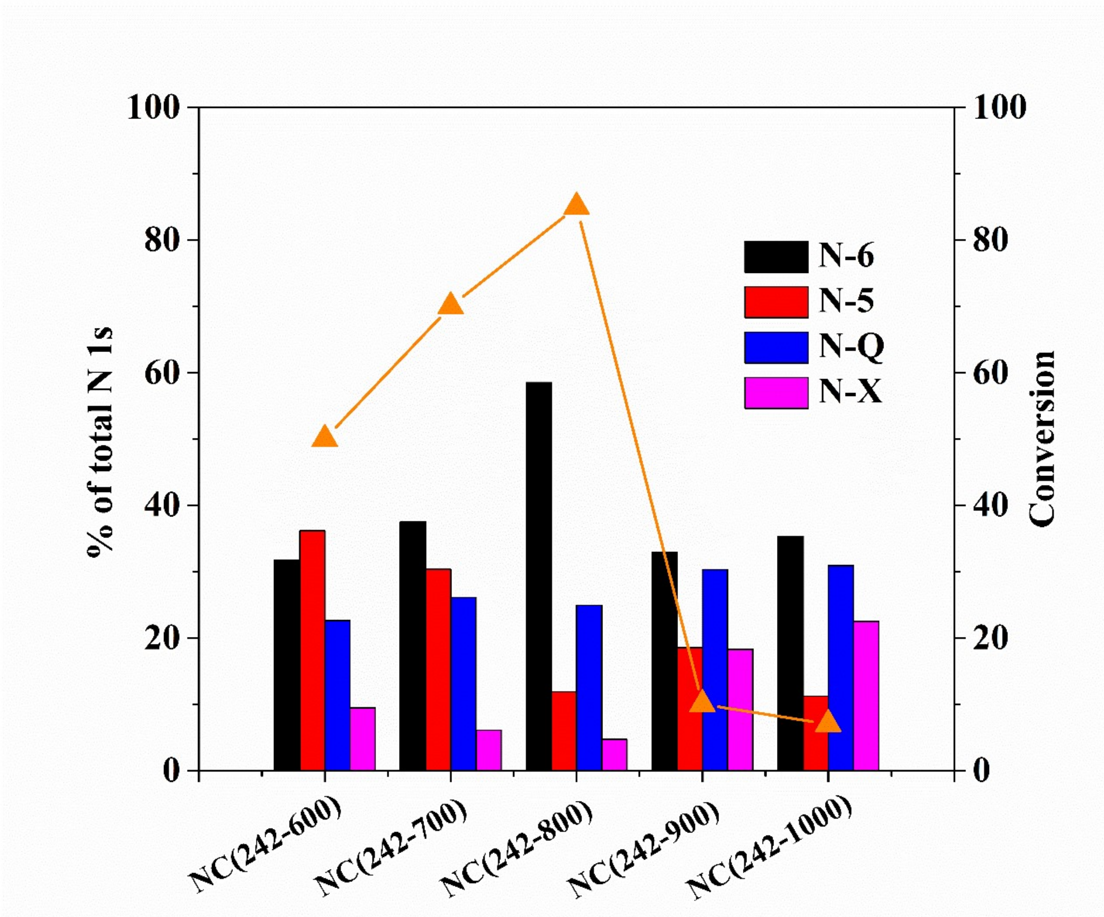
Scheme S6. Pore diameter distribution curves of NC(242-700), NC(242-800), and NC(242-900).



Scheme S7. XPS spectra of NC(242-600), NC(242-700), NC(242-800), NC(242-900) and NC(242-1000).

Table S4. Approximate distribution of N functional groups obtained by fitting the N1s core level XPS spectra of the NC samples.

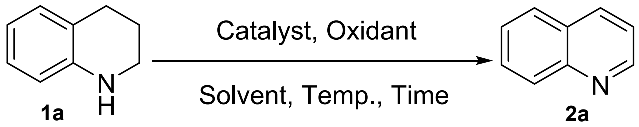
Functional groups	% of total N 1s					Total N content (at%)
	N-6 (eV)	N-5 (eV)	N-Q (eV)	N-X (eV)	N-6 + N-Q	
B. E. (eV)	398.5	400.1	401.1	403.2		
NC(242-600)	31.74	36.15	22.63	9.48	54.37	3.03
NC(242-700)	37.48	30.33	26.09	6.10	63.57	3.87
NC(242-800)	58.52	11.87	24.92	4.69	83.44	3.60
NC(242-900)	32.93	18.51	30.29	18.27	63.22	0.59
NC(242-1000)	35.30	11.25	30.93	22.52	66.23	0.34



Scheme S8. The percentage of N functional groups (bar graph), corresponding to the conversion of tetrahydroquinolone (the yellow line). The existence state of N-6 is significantly varied with the change of carbonization temperature. Especially, the ratio of N-6 is up to 58.52%, the carbon material of NC(242-800) has the highest catalytic activity, in well agreement with 85% conversion.

4. All optimization results

Table S5. More reaction conditions optimization experiments.

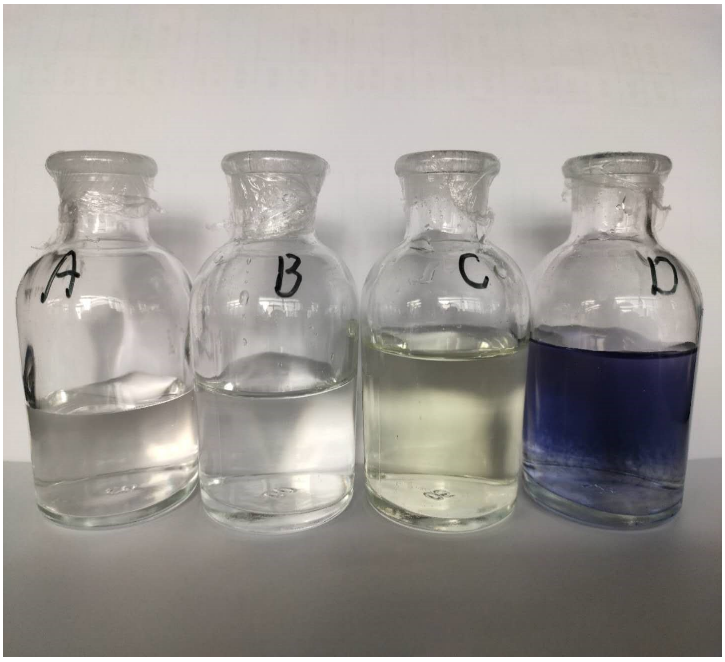


Entry	**Catalysts** (mg)	**Solvent**	**Temp.** (°C)	**Yield** (%) [b]
1	NC(100-800)/12	H_2O	60	trace
2	NC(102-800)/12	H_2O	60	trace
3	NC(121-800)/12	H_2O	60	10
4	NC(242-800)/12	H_2O	60	36
5	NC(363-800)/12	H_2O	60	16
6	NC(484-800)/12	H_2O	60	11
7[c]	NC(242-800)/12	H_2O	60	8
8	—	H_2O	60	N.R
9[d]	NC(242-800)/12	H_2O	60	N.R
10	NC(242-800)/12	EtOH	60	41
11	NC(242-800)/12	CH_3CN	60	12
12	NC(242-800)/12	EtOAc	60	14
13	NC(242-800)/12	toluene	60	trace
14	NC(242-800)/12	dioxane	60	trace
15	NC(242-800)/12	EtOH	rt	20
16	NC(242-800)/12	EtOH	40	30
17	NC(242-800)/12	EtOH	80	44
18[e]	NC(242-800)/12	EtOH	60	17
19[f]	NC(242-800)/12	EtOH	60	43
20	NC(242-800)/25	EtOH	60	60
21	NC(242-800)/40	EtOH	60	85
22[a]	NC(242-800)/40	EtOH	60	23
23[b]	NC(242-800)/40	EtOH	60	11
24[c]	NC(242-800)/40	EtOH	60	8
25[d]	NC(242-800)/40	EtOH	60	16
26	NC(242-600)/40	EtOH	60	52
27	NC(242-700)/40	EtOH	60	68
28	NC(242-900)/40	EtOH	60	13
29	NC(242-1000)/40	EtOH	60	9

[a] Unless otherwise noted, all reaction conditions: 1,2,3,4-tetrahydroquinolines (0.5 mmol), Solvent (2 mL), O_2 balloon, 60°C, 24 h. [b] Isolated yields. [c] Oxidant is air. [d] N_2 as oxidant. [e] 12 h. [f] 36 h. The superscript of NG(242-800) represents the different calcium salt ("a", "b", "c" and "d" stands for $CaSO_4 \cdot 2H_2O$, $Ca(H_2PO_4)_2 \cdot H_2O$, $CaHPO_4$ and $C_{12}H_{22}CaO_{14} \cdot H_2O$, respectively).

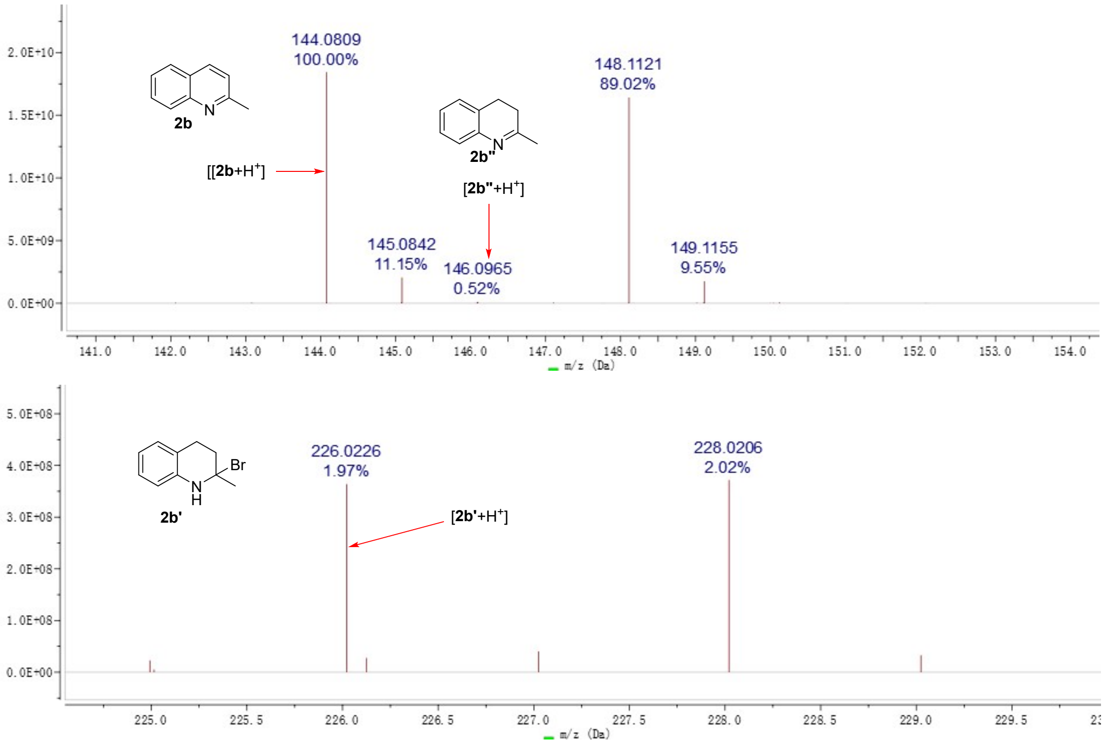
5. Mechanistic study

Detection of H_2O_2[1]

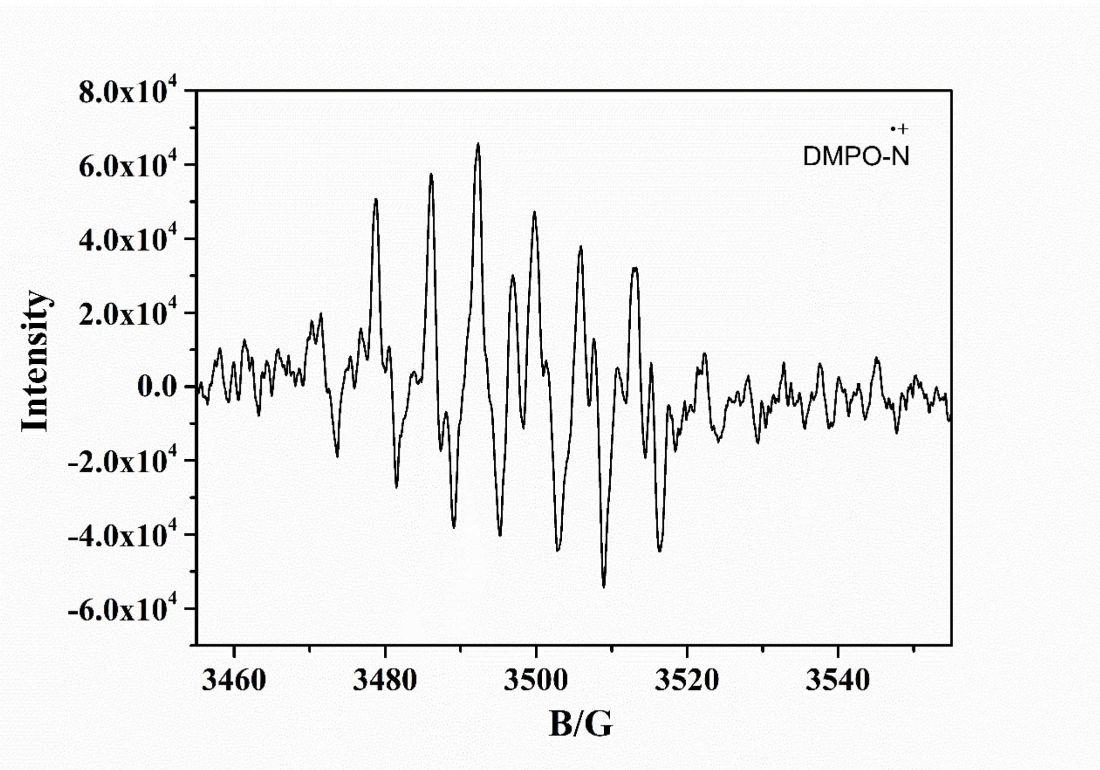


Scheme S9. Detection of H_2O_2

(A) Reaction mixture; (B) Reaction mixture + dil.H_2SO_4; (C) [Reaction mixture + dil.H_2SO_4] + KI; (D) [Reaction mixture + dil.H_2SO_4 + KI] + Starch solution.



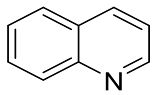
Scheme S10. HR-MS detection of 2-methyl-3,4-dihydroquinoline reaction intermediate.



Scheme S11. The DMPO spin-trapping EPR spectra for N-doped porous carbocatalyst for the amine radical cation.

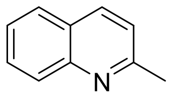
6. Spectroscopic Data

Quinoline (2a)



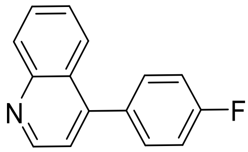
Yellow oil (55 mg, 85% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.92 (dd, *J* = 4.2, 1.2 Hz, 1H), 8.15 (d, *J* = 8.4 Hz, 1H), 8.11 (d, *J* = 8.4 Hz, 1H), 7.81 (d, *J* = 7.8 Hz, 1H), 7.74 – 7.68 (m, 1H), 7.54 (t, *J* = 7.8 Hz, 1H), 7.39 (dd, *J* = 8.4, 4.2 Hz, 1H); ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 150.4, 148.3, 136.0, 129.4, 129.4, 128.3, 127.8, 126.5, 121.0.

2-methylquinoline (2b)



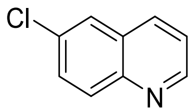
Colorless oil (49 mg, 68% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.02 (dd, *J* = 12.0, 8.4 Hz, 2H), 7.76 (d, *J* = 7.8 Hz, 1H), 7.68-7.65 (m, 1H), 7.48 – 7.45 (m, 1H), 7.27 (d, *J* = 7.8 Hz, 1H), 2.74 (s, 3H); ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 159.0, 147.9, 136.1, 129.4, 128.6, 127.4, 126.5, 125.6, 122.0, 25.4.

4-(4-fluorophenyl)quinoline (2c)



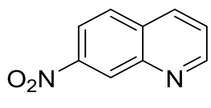
White solid (84 mg, 75% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.93 (d, *J* = 4.2 Hz, 1H), 8.17 (d, *J* = 8.4 Hz, 1H), 7.86 (d, *J* = 8.4 Hz, 1H), 7.74-7.71 (m, 1H), 7.52 – 7.45 (m, 3H), 7.30 (d, *J* = 4.2 Hz, 1H), 7.23-7.19 (m, 2H); ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 163.7, 162.1, 149.9, 148.7, 147.4, 133.9, 133.9, 131.2, 131.2, 129.9, 129.4, 126.7, 126.7, 125.5, 121.3, 115.7, 115.6.

6-chloroquinoline (2d)



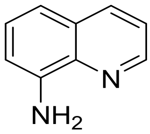
Colorless oil (29 mg, 35% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.91 (dd, *J* = 4.2, 1.8 Hz, 1H), 8.15 – 8.09 (m, 2H), 7.75 (d, *J* = 9.0 Hz, 1H), 7.49 (dd, *J* = 8.4, 2.4 Hz, 1H), 7.39 (dd, *J* = 8.4, 3.6 Hz, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 151.3, 148.6, 135.8, 135.3, 129.0, 128.5, 127.6, 126.6, 121.2.

7-nitroquinoline (2e)



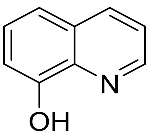
Pale yellow solid (39 mg, 45% yield) Mp. 111-113°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.07 (dd, *J* = 4.2, 1.8 Hz, 1H), 8.98 (d, *J* = 2.4 Hz, 1H), 8.30 (dd, *J* = 8.4, 2.4 Hz, 1H), 8.26 (d, *J* = 8.4 Hz, 1H), 7.96 (d, *J* = 9.0 Hz, 1H), 7.59-7.57 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 152.7, 148.0, 147.1, 135.9, 131.3, 129.4, 125.8, 123.9, 120.0.

quinolin-8-amine (2f)



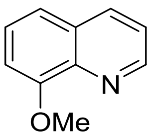
Brown solid (36 mg, 50% yield) Mp. 62-64℃; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.76 (dd, *J* = 4.2, 1.8 Hz, 1H), 8.05 (dd, *J* = 8.4, 1.8 Hz, 1H), 7.37 – 7.29 (m, 2H), 7.15 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.92 (dd, *J* = 7.8, 1.2 Hz, 1H), 5.00 (br, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 147.4, 144.0, 138.4, 136.0, 128.8, 127.4, 121.3, 116.0, 110.0.

quinolin-8-ol (2g)



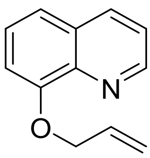
White solid (37 mg, 60% yield) Mp. 71-73℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.80 (dd, *J* = 6.6, 2.4 Hz, 1H), 8.15 (dd, *J* = 12.0, 2.4 Hz, 1H), 7.50 – 7.39 (m, 2H), 7.33 (d, *J* = 12.6 Hz, 1H), 7.21 (d, *J* = 11.4 Hz, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 152.3, 147.9, 138.3, 136.1, 128.5, 127.7, 121.8, 117.9, 110.1.

8-methoxyquinoline （2h）



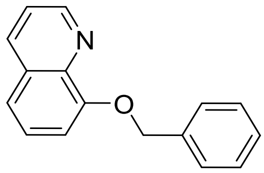
Brown oil (50 mg, 63% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.86 (s, 1H), 8.04 (d, *J* = 8.4 Hz, 1H), 7.39 (t, *J* = 8.4 Hz, 1H), 7.36 – 7.29 (m, 2H), 7.01 – 6.93 (m, 1H), 4.02 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 155.3, 149.2, 140.2, 135.8, 129.3, 126.6, 121.6, 119.5, 107.5, 55.9.

8-(allyloxy)quinoline hydrate (2i)



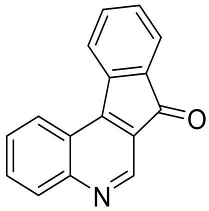
Colorless oil (46 mg, 47% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.94 (dd, *J* = 4.2, 1.8 Hz, 1H), 8.11 (dd, *J* = 8.4, 1.8 Hz, 1H), 7.44 – 7.36 (m, 3H), 7.06 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.24-6.17 (m, 1H), 5.46 (dd, *J* = 17.4, 1.8 Hz, 1H), 5.32 (dd, *J* = 10.2, 1.2 Hz, 1H), 4.86 (d, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 154.3, 149.3, 140.4, 135.8, 133.1, 129.5, 126.5, 121.6, 119.7, 118.3, 109.2, 69.8.

8-(benzyloxy)quinoline (2j)



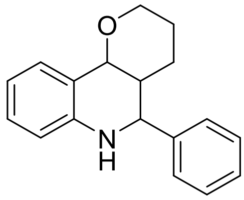
Colorless oil (59 mg, 50%); [1]H NMR (600 MHz, Chloroform-*d*) δ 8.96 (dd, *J* = 4.2, 1.8 Hz, 1H), 8.10 (dd, *J* = 8.4, 1.8 Hz, 1H), 7.51 (d, *J* = 7.8 Hz, 2H), 7.41 (dd, *J* = 7.8, 4.2 Hz, 1H), 7.36-7.33 (m, 4H), 7.28 (t, *J* = 7.8 Hz, 1H), 7.02-7.00 (m, 1H), 5.44 (s, 2H). [13]C NMR (151 MHz, Chloroform-*d*) *δ* 154.3, 149.4, 140.5, 136.9, 135.9, 129.5, 128.6, 127.8, 127.1, 126.5, 121.6, 119.8, 109.9, 70.7.

7H-indeno[2,1-c]quinolin-7-one (2k)



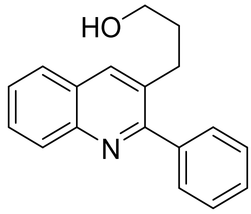
Pale yellow solid (81 mg, 70% yield) Mp 222-224; [1]H NMR (600 MHz, Chloroform-*d*) δ 9.03 (s, 1H), 8.31 (d, *J* = 8.4 Hz, 1H), 8.07 (d, *J* = 8.4 Hz, 1H), 7.96 (d, *J* = 7.2 Hz, 1H), 7.78 – 7.74 (m, 1H), 7.66 (d, *J* = 7.2 Hz, 1H), 7.62 – 7.57 (m, 1H), 7.53 (t, *J* = 7.8 Hz, 1H), 7.41 (t, *J* = 7.5 Hz, 1H). [13]C NMR (151 MHz, Chloroform-*d*) *δ* 192.8, 152.4, 150.9, 144.6, 142.3, 134.6, 133.8, 132.0, 131.0, 131.0, 128.1, 124.8, 124.7, 124.7, 124.3, 123.4.

5-phenyl-3,4,4a,5,6,10 b-hexahydro-2H-pyrano[3,2-c] quinoline (1l)



[1]H NMR (600 MHz, Chloroform-*d*) δ 7.46 (d, *J* = 7.1 Hz, 2H), 7.42 (t, *J* = 7.4 Hz, 2H), 7.37 (t, *J* = 7.2 Hz, 1H), 7.28 (d, *J* = 7.6 Hz, 1H), 7.15 – 7.10 (m, 1H), 6.75 (t, *J* = 7.4 Hz, 1H), 6.54 (d, *J* = 8.0 Hz, 1H), 4.74 (d, *J* = 10.8 Hz, 1H), 4.43 (d, *J* = 2.7 Hz, 1H), 4.12 (s, 2H), 3.76 (td, *J* = 11.6, 2.5 Hz, 1H), 2.12 (t, *J* = 10.5 Hz, 1H), 1.94 – 1.84 (m, 1H), 1.73 – 1.65 (m, 1H), 1.52 (d, *J* = 13.2 Hz, 1H), 1.38 (d, *J* = 13.5 Hz, 1H). **HRMS**(ESI) calcd for $C_{18}H_{20}NO^+$:266.1540. Found:266.1539(MH^+).

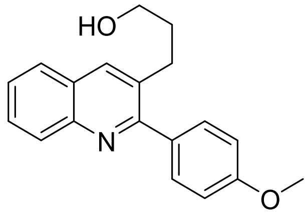
3-(2-phenylquinolin-3-yl)propan-1-ol (2l)



White solid (82 mg, 62% yield) 102-104℃; [1]H NMR (600 MHz, Chloroform-*d*) δ 8.12 (dd, *J* = 8.4, 1.2 Hz, 1H), 8.05 (s, 1H), 7.79 (d, *J* = 1.8 Hz, 1H), 7.67 (m, 1H), 7.52 (t, *J* = 7.2 Hz, 3H), 7.46 (t, *J* = 7.2

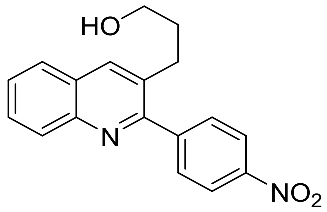
Hz, 2H), 7.42 (t, *J* = 7.2 Hz, 1H), 3.51 (t, *J* = 6.6 Hz, 2H), 2.89 – 2.84 (m, 2H), 1.78 – 1.72 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 160.6, 146.4, 140.7, 135.9, 133.1, 129.2, 129.0, 128.7, 128.3, 128.2, 127.6, 126.9, 126.5, 61.9, 33.3, 29.1. **HRMS**(ESI) calcd for $C_{18}H_{18}NO^+$: 264.1383. Found: 264.1383(MH$^+$).

3-(2-(4-methoxyphenyl)quinolin-3-yl)propan-1-ol (2m)



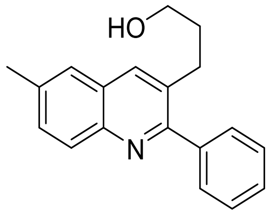
Yellow solid (59 mg, 40% yield) Mp. 118-120°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.10 (d, *J* = 8.4 Hz, 1H), 8.02 (s, 1H), 7.77 (d, *J* = 8.4 Hz, 1H), 7.65 (t, *J* = 7.2 Hz, 1H), 7.51-7.48 (m, 3H), 6.98 (d, J = 8.4 Hz, 2H), 3.84 (s, 3H), 3.52 (t, *J* = 6.6 Hz, 2H), 2.88 (m, 2H), 1.75 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 160.2, 159.6, 146.4, 135.9, 133.3, 133.2, 130.1, 129.1, 128.9, 127.4, 126.9, 126.3, 113.8, 61.9, 55.3, 33.3, 29.2.

3-(2-(4-nitrophenyl)quinolin-3-yl)propan-1-ol (2n)



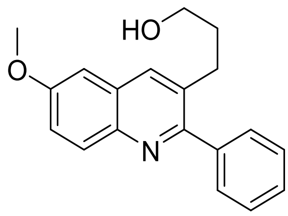
Pale yellow solid (77 mg, 50% yield) Mp. 100-102°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.34 (d, *J* = 9.0 Hz, 2H), 8.13 – 8.07 (m, 2H), 7.83 (d, *J* = 8.4 Hz, 1H), 7.75-7.68 (m, 3H), 7.59 – 7.55 (m, 1H), 3.57 (t, *J* = 6.6 Hz, 2H), 2.90 – 2.84 (m, 2H), 1.81 – 1.74 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 158.0, 147.7, 147.2, 146.4, 136.5, 132.6, 130.0, 129.5, 129.2, 127.8, 127.2, 127.1, 123.6, 61.7, 33.2, 29.0. **HRMS**(ESI) calcd for $C_{18}H_{17}N_2O_3^+$: 309.1234. Found: 309.1234(MH$^+$).

3-(6-methyl-2-phenylquinolin-3-yl)propan-1-ol (2o)



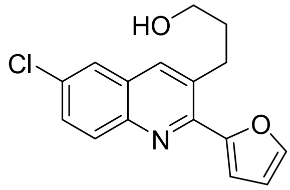
White solid (133 mg, 96% yield) Mp. 101-103; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.00 (d, *J* = 8.4 Hz, 1H), 7.90 (s, 1H), 7.51 – 7.44 (m, 4H), 7.40-7.34 (m, 3H), 3.38 (t, *J* = 6.6 Hz, 2H), 2.79 – 2.74 (m, 2H), 2.59 (s, 1H), 2.52 (s, 3H), 1.69 – 1.62 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 159.6, 144.9, 140.7, 136.3, 135.3, 133.2, 131.3, 128.7, 128.7, 128.2, 128.0, 127.6, 125.7, 61.5, 33.3, 29.1, 21.6. **HRMS**(ESI) calcd for $C_{19}H_{20}NO^+$: 278.1539. Found: 278.1536(MH$^+$).

3-(6-methoxy-2-phenylquinolin-3-yl)propan-1-ol (2p)



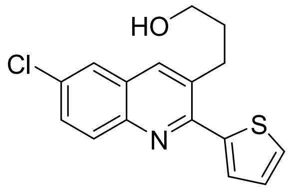
(63 mg, 43% yield); [1]H NMR (600 MHz, Chloroform-*d*) δ 8.01 (d, *J* = 9.0 Hz, 1H), 7.95 (s, 1H), 7.54 – 7.50 (m, 2H), 7.47-7.38 (m, 3H), 7.32 (dd, *J* = 9.0, 2.8 Hz, 1H), 7.06 (d, *J* = 2.4 Hz, 1H), 3.94 (s, 3H), 3.53 (t, *J* = 6.6 Hz, 2H), 2.88 – 2.82 (m, 2H), 1.79 – 1.71 (m, 2H). [13]C NMR (151 MHz, Chloroform-*d*) *δ* 158.1, 157.8, 142.5, 140.8, 134.8, 133.3, 130.7, 128.8, 128.5, 128.3, 128.0, 121.7, 104.4, 61.9, 55.5, 33.4, 29.0.

3-(6-chloro-2-(furan-2-yl)quinolin-3-yl)propan-1-ol (2q)



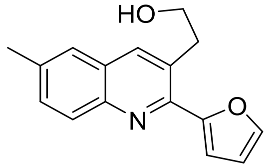
Brown solid (99 mg, 69% yield) Mp. 115-117℃; δ_H (600 MHz, Chloroform-*d*) 8.00 (1 H, d, *J* 9.0), 7.85 (1 H, s), 7.70 – 7.51 (3 H, m), 7.15 (1 H, d, *J* 3.5), 6.65 – 6.45 (1 H, m), 3.71 (2 H, t, *J* 6.3), 3.14 (2 H, t, *J* 7.8), 2.02 – 1.84 (3 H, m); [13]C NMR (151 MHz, Chloroform-*d*) *δ* 153.5, 148.9, 144.8, 143.7, 136.0, 133.3, 132.1, 130.6, 130.0, 127.8, 125.4, 112.5, 111.9, 62.0, 33.1, 29.7.

3-(6-chloro-2-(thiophen-2-yl)quinolin-3-yl)propan-1-ol (2r)



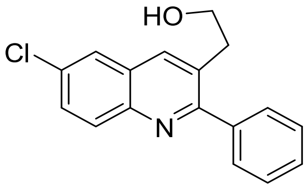
Pale yellow solid (108 mg, 71% yield) Mp. 102-104℃; [1]H NMR (600 MHz, Chloroform-*d*) δ 7.98 (d, *J* = 9.0 Hz, 1H), 7.85 (d, *J* = 3.0 Hz, 1H), 7.66 (s, 1H), 7.58 – 7.50 (m, 2H), 7.44 (d, *J* = 5.4 Hz, 1H), 7.13 – 7.08 (m, 1H), 3.66 (t, *J* = 6.0 Hz, 2H), 3.11 – 3.04 (m, 2H), 1.93 – 1.85 (m, 2H). [13]C NMR (151 MHz, Chloroform-*d*) *δ* 152.9, 144.7, 143.7, 135.6, 133.5, 132.1, 130.5, 130.0, 128.2, 127.8, 127.7, 127.7, 125.4, 61.8, 32.7, 29.8.

2-(2-(furan-2-yl)-6-methylquinolin-3-yl)ethan-1-ol (2s)



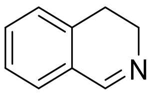
Pale yellow solid (96 mg, 76% yield) Mp. 126-128℃; [1]H NMR (600 MHz, Chloroform-*d*) δ 7.91 (dd, *J* = 8.4, 1.8 Hz, 1H), 7.82 (d, *J* = 3.0 Hz, 1H), 7.54 (d, *J* = 1.8 Hz, 1H), 7.43 (d, *J* = 8.4 Hz, 1H), 7.34 (d, *J* = 4.2 Hz, 1H), 7.05 (t, *J* = 3.0 Hz, 1H), 6.52-6.49 (m, 1H), 3.90 (t, *J* = 6.6 Hz, 2H), 3.23 (t, *J* = 6.6 Hz, 2H), 2.47 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 153.8, 147.8, 145.1, 143.3, 137.6, 136.4, 131.6, 129.0, 128.5, 127.0, 125.7, 111.9, 111.7 , 62.5, 36.7, 21.6.

2-(6-chloro-2-phenylquinolin-3-yl)ethan-1-ol (2t)



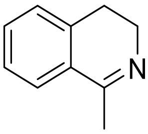
White solid (85 mg, 60% yield) Mp. 89-91℃; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.97 (t, *J* = 7.8 Hz, 2H), 7.69 (s, 1H), 7.55 (dd, *J* = 8.4, 2.4 Hz, 1H), 7.46 – 7.37 (m, 5H), 3.61 (t, *J* = 6.5 Hz, 2H), 2.93 (t, *J* = 6.4 Hz, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 160.8, 144.7, 140.0, 136.0, 132.2, 131.2, 130.6, 130.1, 128.7, 128.4, 128.4, 128.0, 125.6, 62.0, 35.7.

3,4-dihydroisoquinoline (4a)



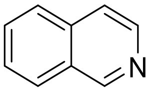
Brown oil (56 mg, 85% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8,31 (s, 1H), 7.34-7.31 (m, 1H), 7.29 – 7.22 (m, 2H), 7.13 (d, *J* = 7.2 Hz, 1H), 3.75 (m, 2 H), 2.72 (t, J =7.8Hz, 2 H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 160.3, 136.3, 131.0, 128.5, 127.4, 127.2, 127.0, 47.3, 25.0.

1-methyl-3,4-dihydroisoquinoline (4b)



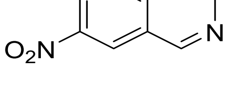
Yellow oil (48 mg, 67% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.48 (d, *J* = 7.2 Hz, 1H), 7.36-7.34 (m, 1H), 7.31 – 7.27 (m, 1H), 7.18 (d, *J* = 7.2 Hz, 1H), 3.67-3.64 (m, 2H), 2.70 (t, J = 7.2 Hz, 2H), 2.38 (t, *J* = 1.8 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 164.3, 137.4, 130.6, 129.5, 127.4, 126.9, 125.3, 46.9, 26.0, 23.2.

Isoquinoline (4c)



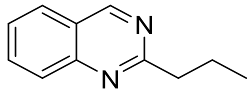
Brown oil (20 mg, 31% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.26 (s, 1H), 8.52 (d, *J* = 5.4 Hz, 1H), 7.97 (d, *J* = 8.4 Hz, 1H), 7.82 (d, *J* = 8.4 Hz, 1H), 7.70 (m, 1H), 7.65 (d, *J* = 6.0 Hz, 1H), 7.61 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 152.5, 142.9, 135.8, 130.3, 128.7, 127.6, 127.2, 126.4, 120.5.

7-nitro-3,4-dihydroisoquinoline (4d)



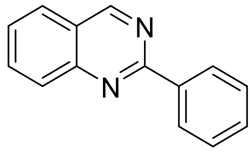
Yellow solid (36 mg, 26%) Mp. 165-167°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.42 (t, *J* = 2.4 Hz, 1H), 8.22 (dd, *J* = 7.8, 2.4 Hz, 1H), 8.13 (d, *J* = 2.4 Hz, 1H), 7.35 (d, J=8.4 Hz, 1H), 3.87 – 3.83 (m, 2H), 2.86 (t, *J* = 7.8 Hz, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 158.2, 147.2, 143.5, 128.8, 128.5, 125.6, 121.8, 46.8, 25.1.

2-propylquinazoline (6a)



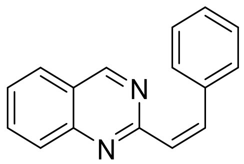
Pale brown solid (95 mg, 70% yield) Mp. 121-123°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.29 (s, 1H), 7.93 (d, *J* = 8.4 Hz, 1H), 7.82 (t, *J* = 8.4 Hz, 2H), 7.53 (t, *J* = 7.8 Hz, 1H), 3.05 (t, *J* = 7.8 Hz, 2H), 1.91 (m, 2H), 1.00 (t, *J* = 7.8 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 172.1, 153.3, 135.1, 125.8, 124.6, 122.5, 121.4, 36.2, 23.1, 13.7.

2-phenylquinazoline (6b)



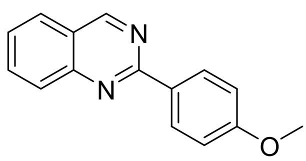
Yellow solid (67 mg, 65% yield) 99-101°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.43 (s, 1H), 8.64 – 8.59 (m, 2H), 8.07 (d, *J* = 8.4 Hz, 1H), 7.90 – 7.84 (m, 2H), 7.57 (t, *J* = 7.8 Hz, 1H), 7.52 (m, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 161.0, 160.5, 160.4, 150.8, 138.0, 134.1, 130.6, 128.6, 128.6, 127.2, 127.1, 123.6.

(E)-2-styrylquinazoline (6c)



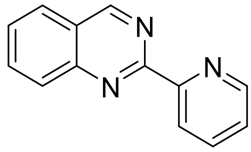
White solid (78 mg, 65% yield) Mp. 116-118°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.36 (s, 1H), 8.15 (d, *J* = 16.3 Hz, 1H), 7.98 (d, *J* = 9.0 Hz, 1H), 7.87 (t, *J* = 7.8 Hz, 2H), 7.67 (d, *J* = 7.2 Hz, 2H), 7.57 (t, *J* = 6.6 Hz, 1H), 7.43 – 7.38 (m, 3H), 7.34 (t, *J* = 7.2 Hz, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 161.3, 160.2, 150.6, 138.5, 136.2, 134.2, 129.0, 128.8, 128.1, 127.9, 127.7, 127.2, 127.1, 123.3.

2-(4-methoxyphenyl)quinazoline (6d)



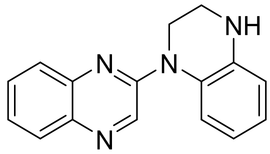
Yellow solid (87 mg, 74% yield) 87-89°C; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.42 (s, 1H), 8.58 (d, *J* = 13.2 Hz, 2H), 8.04 (d, *J* = 12.0 Hz, 1H), 7.92 – 7.84 (m, 2H), 7.57 (t, *J* = 11.4 Hz, 1H), 7.05 (d, *J* = 13.8 Hz, 2H), 3.90 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 161.8, 160.8, 160.4, 150.8, 134.0, 130.7, 130.2, 128.4, 127.1, 126.8, 123.3, 114.0, 55.4.

2-(pyridin-2-yl)quinazoline (6e)



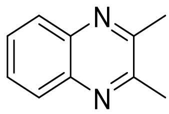
Brown solid (83 mg, 80% yield) Mp. 91-93°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.50 (t, *J* = 1.2 Hz, 1H), 8.85 (d, *J* = 4.8 Hz, 1H), 8.63 (dd, *J* = 7.8, 1.2 Hz, 1H), 8.18 (d, *J* = 8.4 Hz, 1H), 7.92 – 7.81 (m, 3H), 7.60 (t, *J* = 6.6 Hz, 1H), 7.38 – 7.33 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 160.9, 159.9, 155.1, 150.7, 150.2, 136.9, 134.3, 129.1, 128.0, 127.1, 124.7, 124.1, 124.0.

3,4-dihydro-2H-1,2'-biquinoxaline (8a)



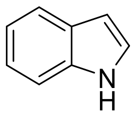
Brown oil (52 mg, 80% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.68 (d, *J* = 1.8 Hz, 1H), 8.61 (d, *J* = 1.8 Hz, 1H), 7.91 (d, *J* = 9.0 Hz, 1H), 7.78 (dd, *J* = 9.0, 2.4 Hz, 1H), 7.55 (d, *J* = 2.4 Hz, 1H), 7.08 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.85-6.82 (m, 1H), 6.66 – 6.58 (m, 2H), 4.05 (s, 1H), 3.84 (t,*J* = 7.2 Hz, 2H), 3.48 (t, *J* = 5.4 Hz, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 149.0, 145.0, 144.7, 142.2, 139.4, 136.5, 129.5, 128.9, 126.1, 123.1, 119.3, 117.6, 115.4, 115.3, 47.1, 41.3. **HRMS**(ESI) calcd for $C_{16}H_{15}N_4^+$: 263.1291. Found: 263.1289(MH$^+$).

2,3-dimethylquinoxaline (8b)



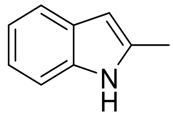
Pink solid (52 mg, 66% yield) Mp. 107-109°C; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.93 (dd, *J* = 9.0, 5.4 Hz, 2H), 7.62 (dd, *J* = 9.6, 5.4 Hz, 2H), 2.68 (s, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 153.4, 141.0, 128.7, 128.3 , 23.1.

1H-indole (10a)



White solid (57 mg, 96% yield) MP. 50-52°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.00 (br, 1H), 7.74 (d, *J* = 7.8 Hz, 1H), 7.43 – 7.36 (m, 1H), 7.28 (t, *J* = 7.2 Hz, 1H), 7.23 – 7.17 (m, 2H), 6.62 (s, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 135.8, 127.9, 124.2, 122.0, 120.8, 119.9, 111.1, 102.6.

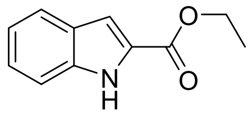
2-methyl-1H-indole (10b)



White solid (57 mg, 87% yield) Mp. 53-55°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.74 (br, 1H), 7.55 (d, *J* = 7.8 Hz, 1H), 7.28 (d, *J* = 8.4 Hz, 1H), 7.16-7.08 (m, 2H), 6.24 (s, 1H), 2.44 (s, 3H). ^{13}C NMR

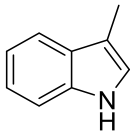
(151 MHz, Chloroform-*d*) δ 136.0, 135.1, 129.1, 120.9, 119.6, 110.2, 110.2, 100.4, 13.7.

ethyl 1H-indole-2-carboxylate (10d)



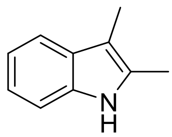
White solid (85mg, 90% yield) Mp. 120-122°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 9.30 (s, 1H), 7.70 (d, *J* = 7.8 Hz, 1H), 7.44 (d, *J* = 8.4 Hz, 1H), 7.33 (t, *J* = 7.2 Hz, 1H), 7.25 (s, 1H), 7.16 (t, *J* = 7.2 Hz, 1H), 4.47-4.42 (m, 2H), 1.44 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 162.2, 137.0, 127.5, 125.3, 122.6, 120.7, 111.9, 108.6, 61.1, 14.4.

3-methyl-1H-indole (10e)



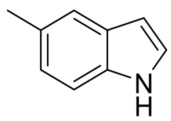
White solid (55 mg, 84% yield) Mp. 97-99°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.81 (s, 1H), 7.62 (d, *J* = 7.8 Hz, 1H), 7.35 (d, *J* = 8.4 Hz, 1H), 7.23 (t, *J* = 7.2 Hz, 1H), 7.16 (t, *J* = 7.8 Hz, 1H), 6.97 (s, 1H), 2.38 (d, *J* = 1.2 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 136.3, 128.3, 121.9, 121.6, 119.1, 118.8, 111.7, 110.9, 9.67.

2,3-dimethyl-1H-indole (10f)



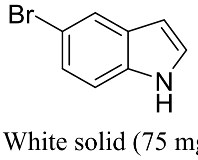
White solid (59 mg, 81% yield) Mp. 108-110°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.58 (br, 1H), 7.51 (d, *J* = 7.2 Hz, 1H), 7.27 – 7.23 (m, 1H), 7.17-7.11 (m, 2H), 2.36 (s, 3H), 2.27 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 135.2, 130.7, 129.4, 120.9, 119.0, 117.9, 110.0, 107.1, 11.5, 8.5.

5-methyl-1H-indole (10g)



Brown solid (59 mg, 90% yield) Mp. 57-59°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.92 (br, 1H), 7.50 (s, 1H), 7.29 (d, *J* = 8.4 Hz, 1H), 7.15 (t, *J* = 3.0 Hz, 1H), 7.09 (dd, *J* = 8.4, 1.2 Hz, 1H), 6.54-6.52 (m, 1.0 Hz, 1H), 2.52 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 134.1, 129.0, 128.2, 124.3, 123.6, 120.4, 110.7, 102.1, 21.5.

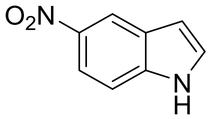
5-bromo-1H-indole(10h)



White solid (75 mg, 77% yield) Mp. 91-93°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.14 (br, 1H), 7.79

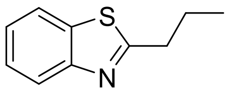
(s, 1H), 7.29 (dd, *J* = 8.4, 1.8 Hz, 1H), 7.24 (d, *J* = 9.0 Hz, 1H), 7.19 (s, 1H), 6.50 (s, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 134.4, 129.6, 125.4, 124.8, 123.2, 113.0, 112.5, 102.3.

5-nitro-1H-indole (10i)



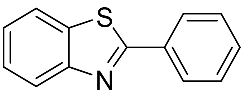
Yellow solid (39 mg, 48% yield) Mp. 139-141°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.73 (br, 1H), 8.60 (d, *J* = 2.4 Hz, 1H), 8.10 (dd, *J* = 9.0, 2.4 Hz, 1H), 7.44 (d, *J* = 9.0 Hz, 1H), 7.40 – 7.36 (m, 1H), 6.75 – 6.71 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 141.9, 138.8, 127.4, 127.2, 118.0, 117.6, 111.0, 105.0.

2-propylbenzo[d]thiazole (12a)



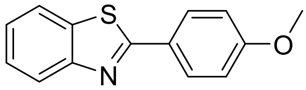
Colorless oil (125 mg, 90% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.96 (d, *J* = 7.8 Hz, 1H), 7.81 (d, *J* = 7.8 Hz, 1H), 7.44 – 7.39 (m, 1H), 7.31 (t, *J* = 7.8 Hz, 1H), 3.10 – 3.04 (m, 2H), 1.93-1.86 (m, *J* = 7.4 Hz, 2H), 1.04 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 172.1, 153.3, 135.1, 125.8, 124.6, 122.5, 121.4, 36.2, 23.1, 13.7.

2-phenylbenzo[d]thiazole (12b)



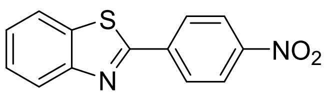
White solid (98 mg, 93% yield) 112-114°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.12 – 8.06 (m, 3H), 7.90 (d, *J* = 9.0 Hz, 1H), 7.52 – 7.47 (m, 4H), 7.40 – 7.36 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 168.0, 154.1, 135.1, 133.6, 130.9, 129.0, 127.5, 126.3, 125.2, 123.2, 121.6.

2-(4-methoxyphenyl)benzo[d]thiazole (12c)



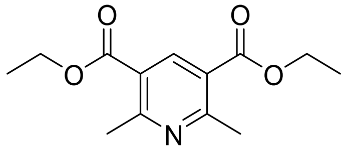
White solid (100 mg, 82% yield) Mp. 120-122°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.03 (dd, *J* = 8.4, 3.0 Hz, 3H), 7.87 (d, *J* = 8.4 Hz, 1H), 7.49 – 7.44 (m, 1H), 7.35 (t, *J* = 7.8 Hz, 1H), 7.00 (d, *J* = 9.0 Hz, 2H), 3.88 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 167.8, 161.9, 154.2, 134.8, 129.1, 126.4, 126.2, 124.8, 122.8, 121.5, 114.3, 55.4.

2-(4-nitrophenyl)benzo[d]thiazole (12d)



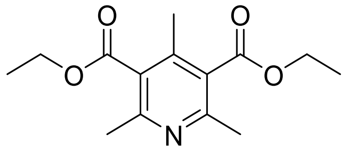
Yellow solid (81 mg,63% yield) Mp. 234-236°C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 8.34 (d, *J* = 9.0 Hz, 2H), 8.26 (d, *J* = 9.0 Hz, 2H), 8.12 (d, *J* = 8.4 Hz, 1H), 7.95 (d, *J* = 7.8 Hz, 1H), 7.57 – 7.52 (m, 1H), 7.48 – 7.44 (m, 1H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 164.8, 154.1, 149.0, 139.1, 135.5, 128.2, 126.9, 126.2, 124.3, 123.9, 121.8.

diethyl 2,6-dimethylpyridine-3,5-dicarboxylate (14a)



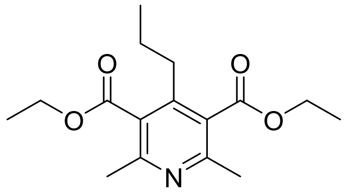
White solid (120 mg, 95%) Mp. 71-73℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.62 (d, *J* = 3.0 Hz, 1H), 4.38-4.32 (m, 2H), 2.81 – 2.77 (m, 6H), 1.37 (t, *J* = 10.2 Hz, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 165.9, 162.2, 140.8, 123.0, 61.3, 24.9, 14.2.

diethyl 2,4,6-trimethylpyridine-3,5-dicarboxylate (14b)



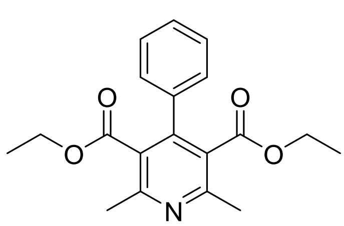
Colorless oil (120 mg, 90%yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 4.41-4.37 (m, 4H), 2.50 (s, 6H), 2.25 (s, 3H), 1.37 (t, *J* = 7.2 Hz, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 168.3, 154.9, 142.0, 127.5, 61.5, 22.9, 16.9, 14.1.

diethyl 2,6-dimethyl-4-propylpyridine-3,5-dicarboxylate (14c)



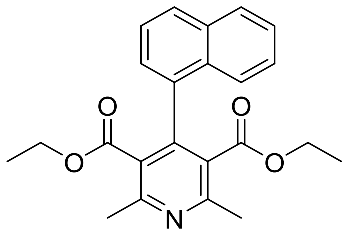
Colorless oil (125 mg, 85% yield); ^{1}H NMR (600 MHz, Chloroform-*d*) δ 4.41-4.36 (m, 4H), 2.55 – 2.51 (m, 2H), 2.48 (s, 6H), 1.54 (d, *J* = 7.8 Hz, 2H), 1.37 (t, *J* = 7.2 Hz, 6H), 0.91 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 168.5, 155.0, 146.3, 127.2, 61.5, 33.4, 24.2, 22.9, 14.4, 14.1.

diethyl 2,6-dimethyl-4-phenylpyridine-3,5-dicarboxylate (14d)



Yellow solid (102 mg, 62% yield) 61-63℃; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.36 – 7.33 (m, 3H), 7.25 – 7.22 (m, 2H), 4.00-3.96 (m, 4H), 2.59 (s, 6H), 0.88 (t, *J* = 7.2 Hz, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 167.8, 155.4, 146.1, 136.5, 128.4, 128.1, 128.0, 126.9, 61.3, 22.9, 13.5.

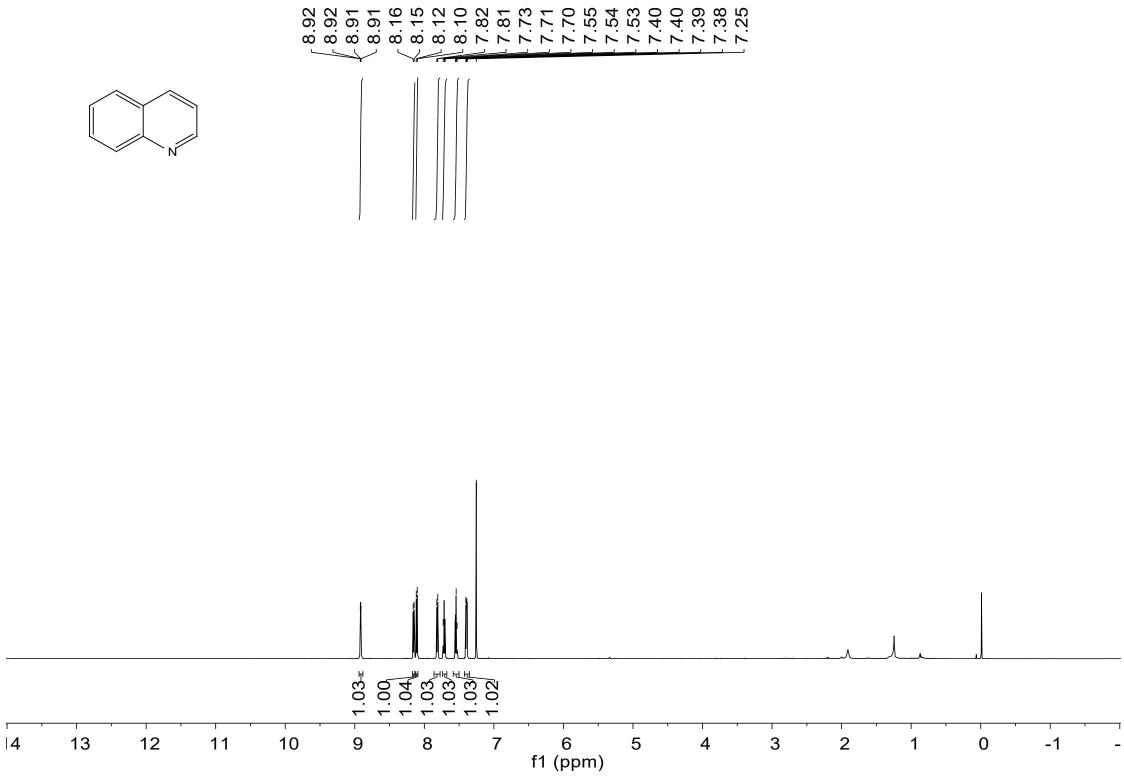
diethyl 2,6-dimethyl-4-(naphthalen-1-yl)pyridine-3,5-dicarboxylate (14e)



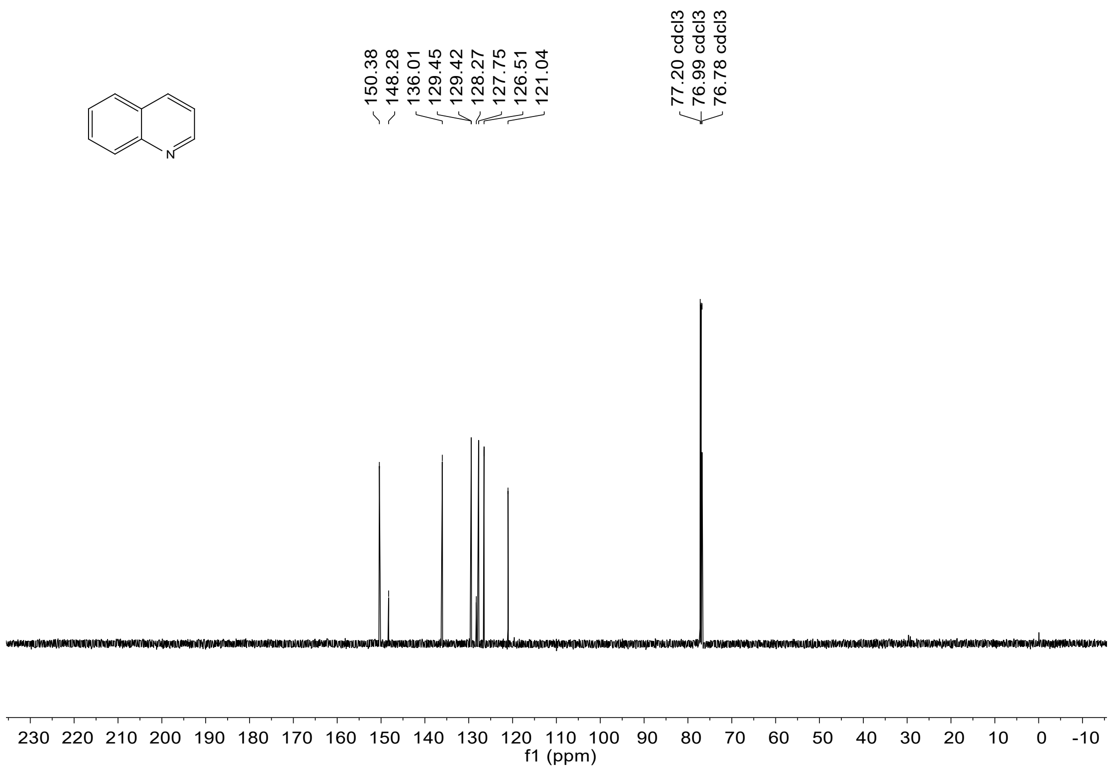
Pink solid (98 mg, 52% yield) Mp. 57-59℃; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.84 – 7.79 (m, 2H), 7.47 – 7.37 (m, 4H), 7.26 (d, *J* = 7.8 Hz, 1H), 3.68 (dd, *J* = 8.4, 7.2 Hz, 4H), 2.65 (s, 6H), 0.43 (s, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) *δ* 167.4, 155.8, 145.3, 134.0, 133.0, 131.4, 128.6, 127.7, 127.7, 126.5, 126.3, 126.1, 126.0, 124.6, 23.1, 13.0.

7. ^{1} H and ^{13}C NMR Spectra

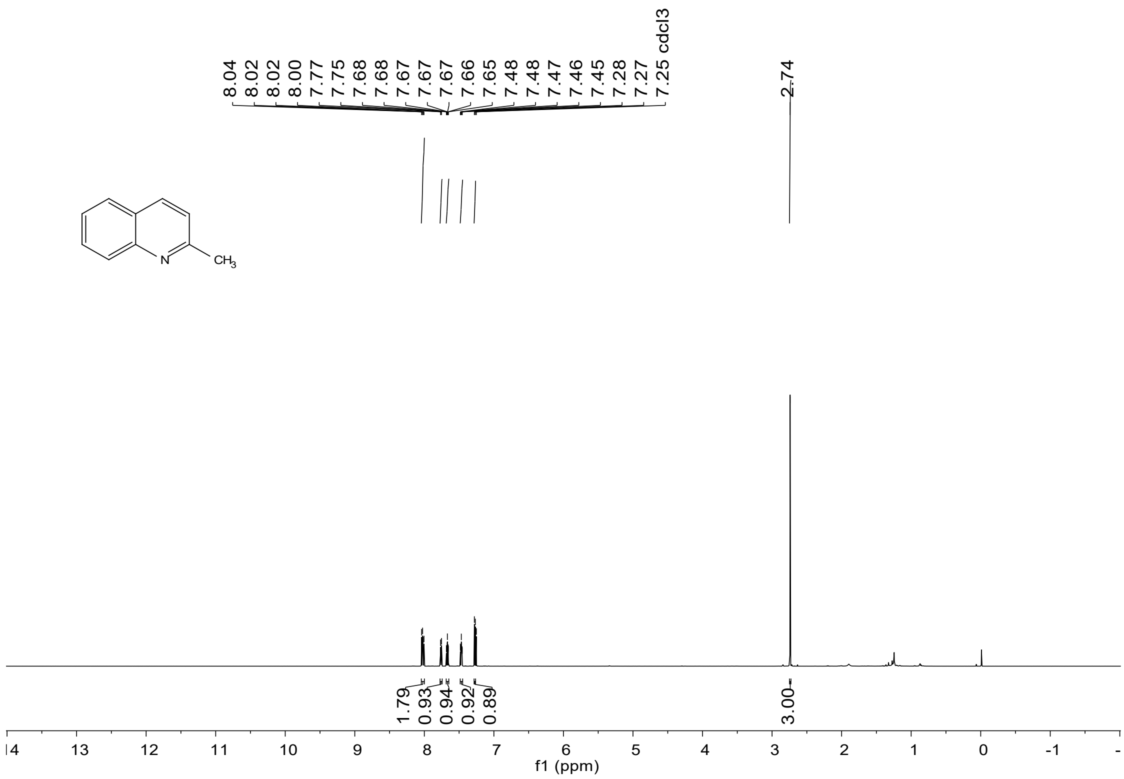
^{1}H NMR spectrum of 2a



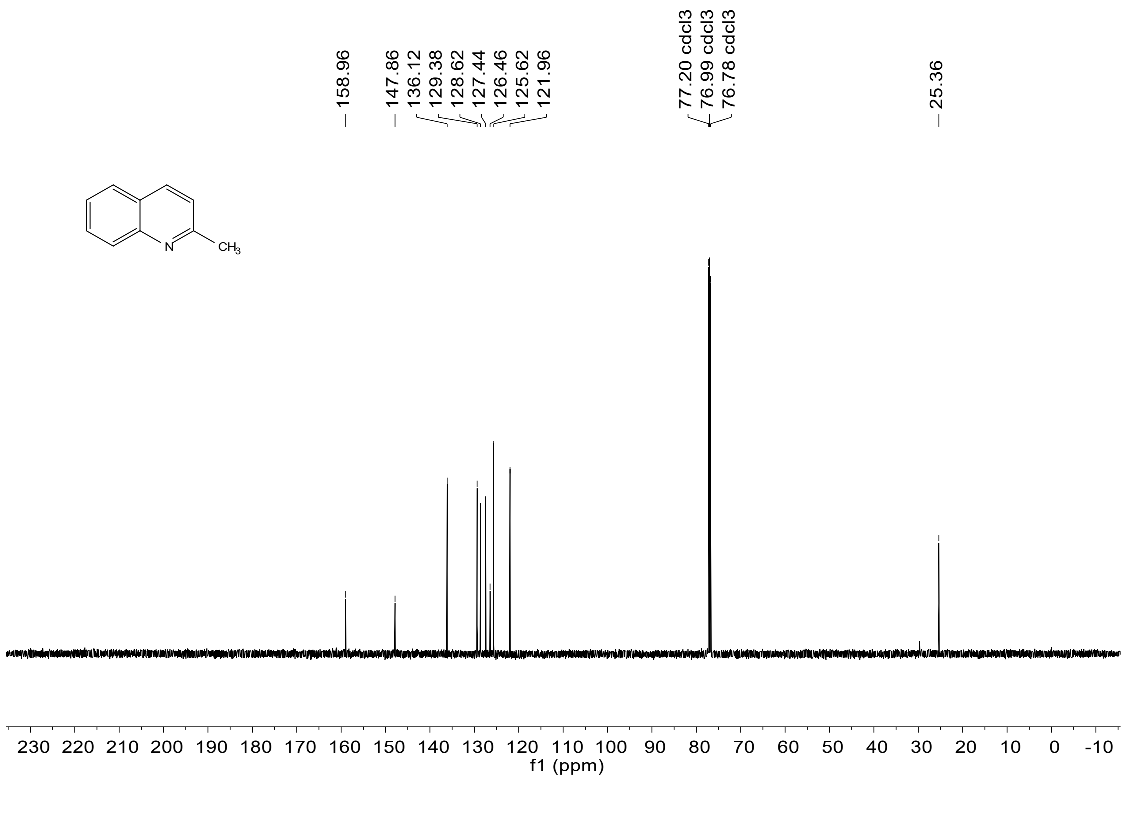
^{13}C NMR spectrum of 2a



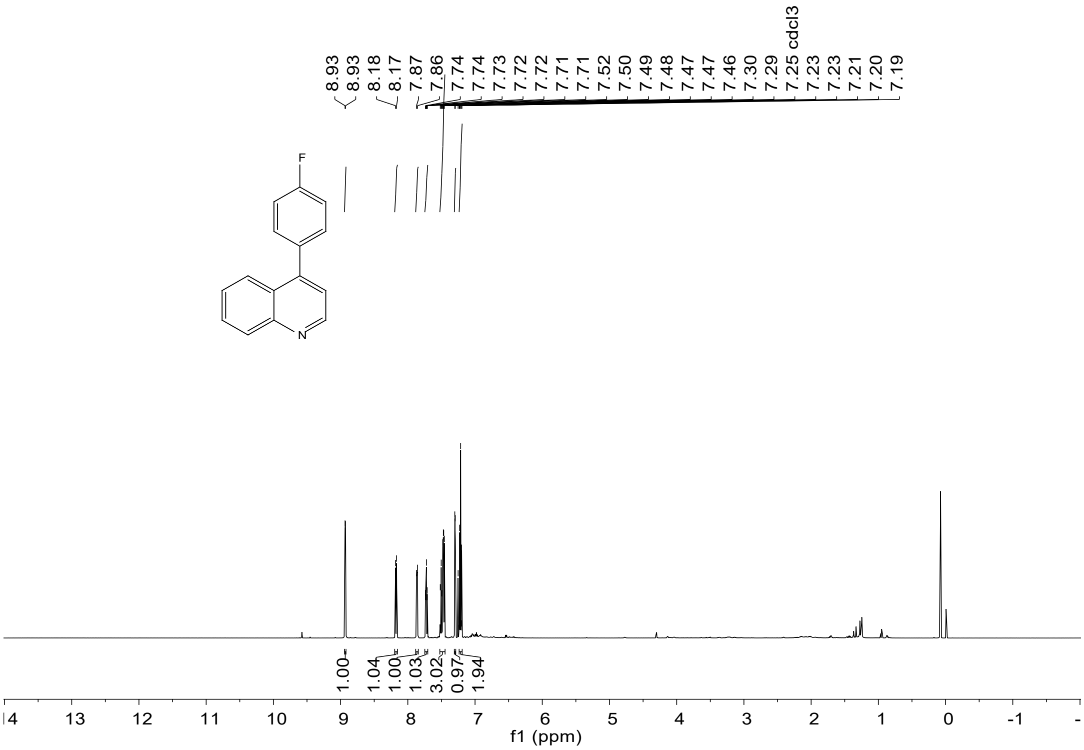
^{1}H NMR spectrum of 2b



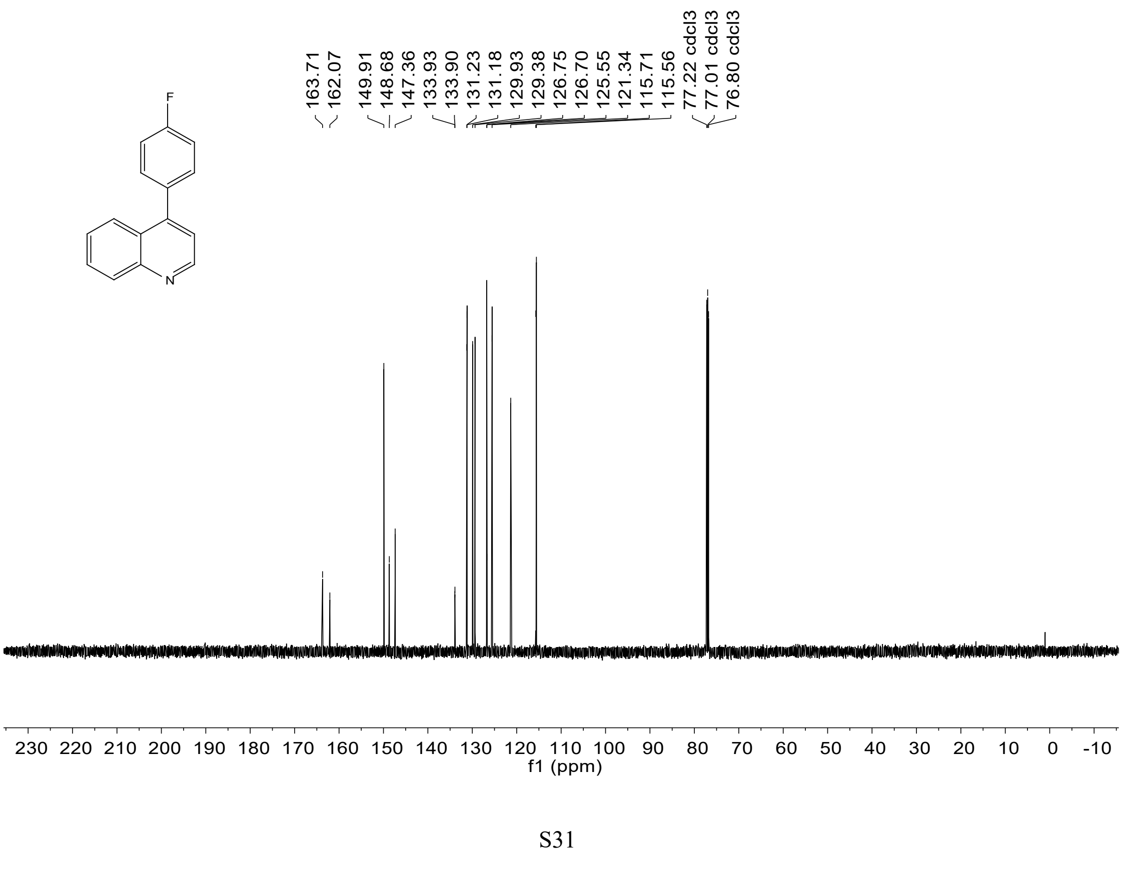
^{13}C NMR spectrum of 2b



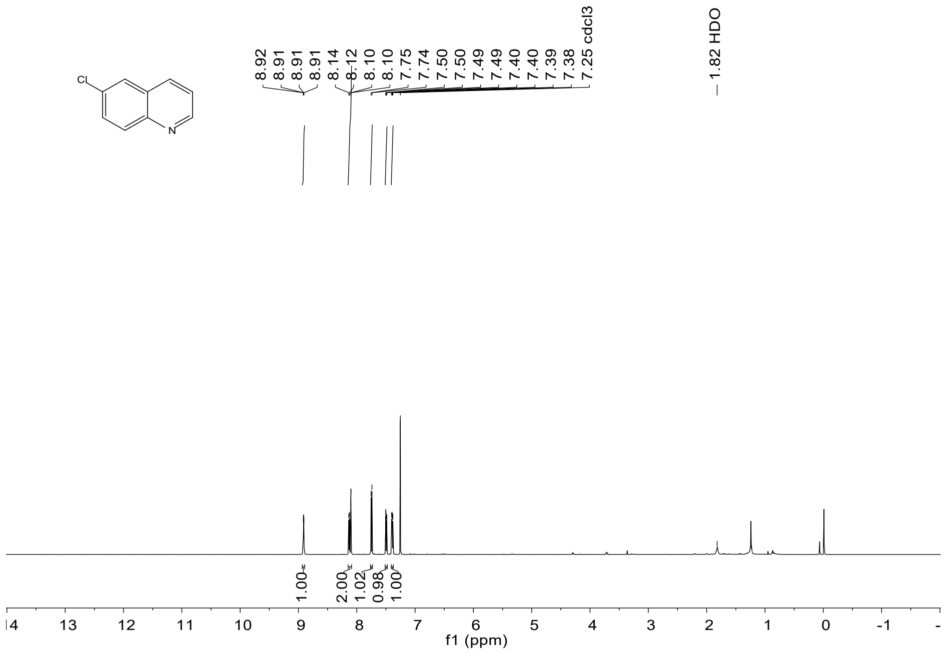
[1]H NMR spectrum of 2c



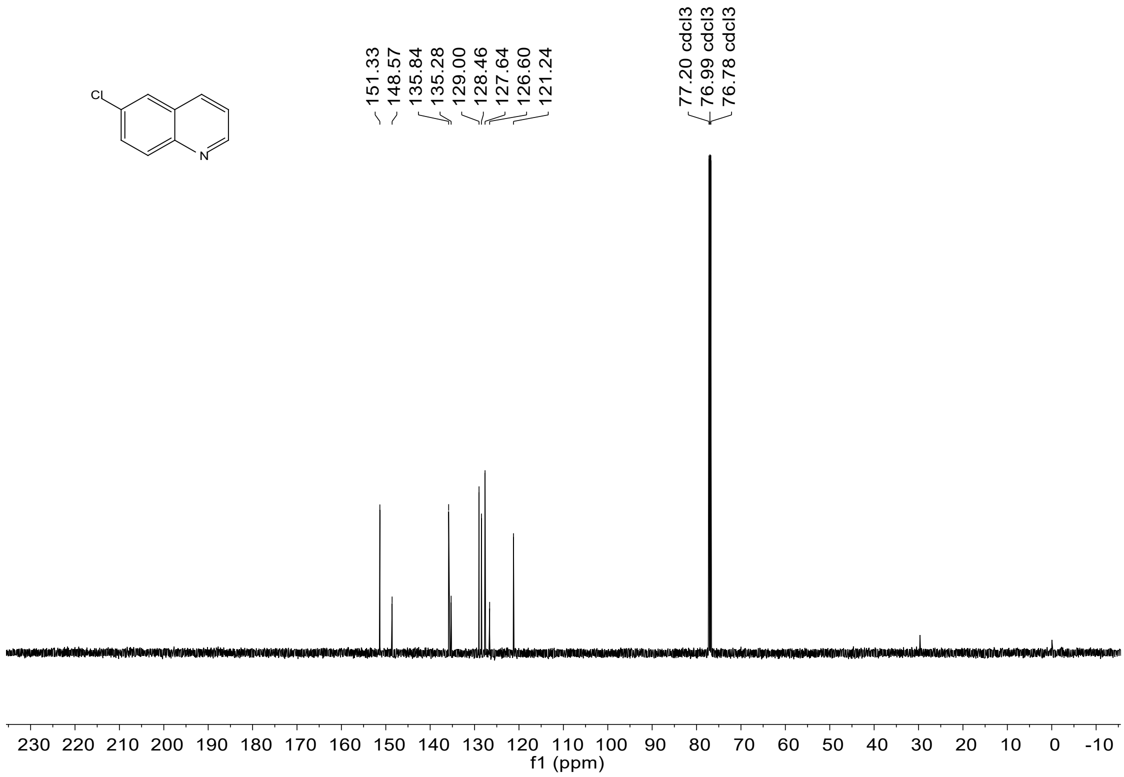
[13]C NMR spectrum of 2c



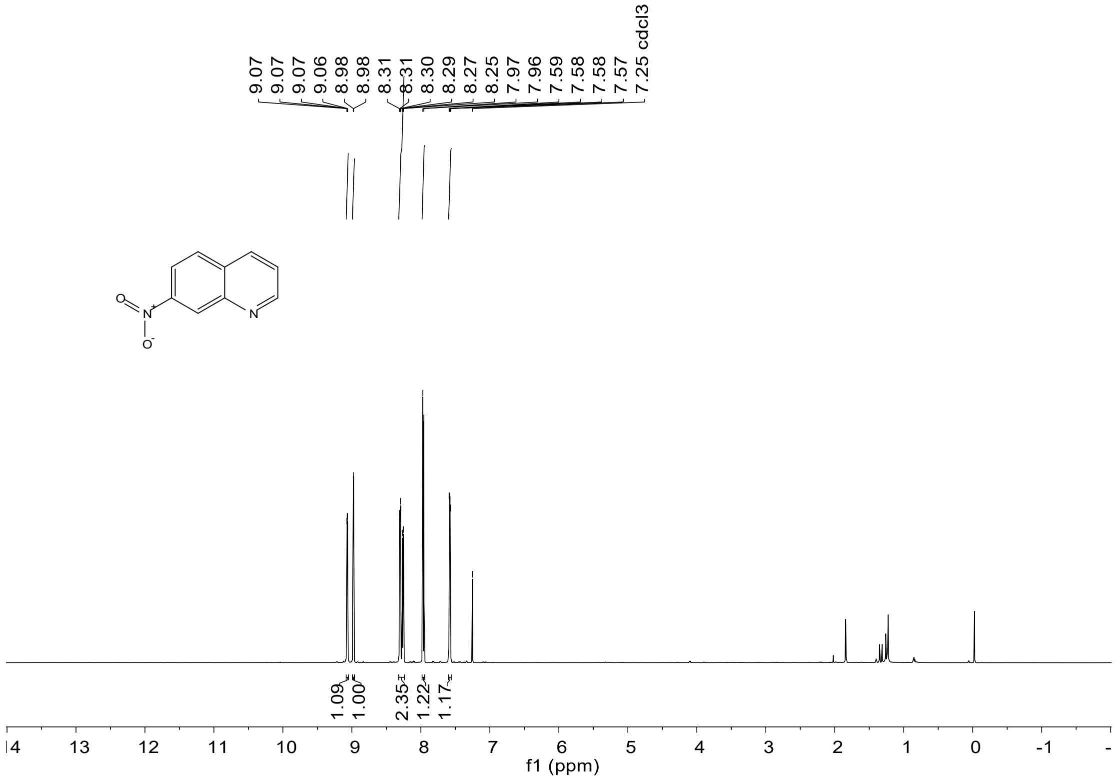
[1]H NMR spectrum of 2d



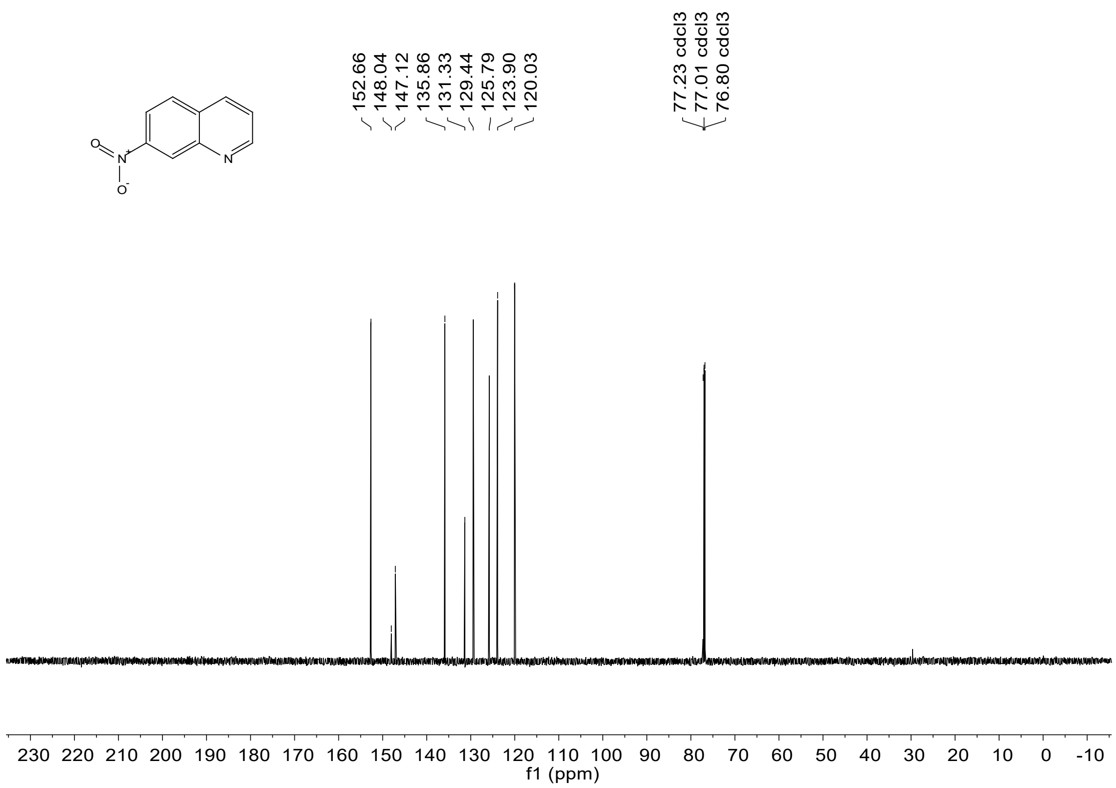
[13]C NMR spectrum of 2d



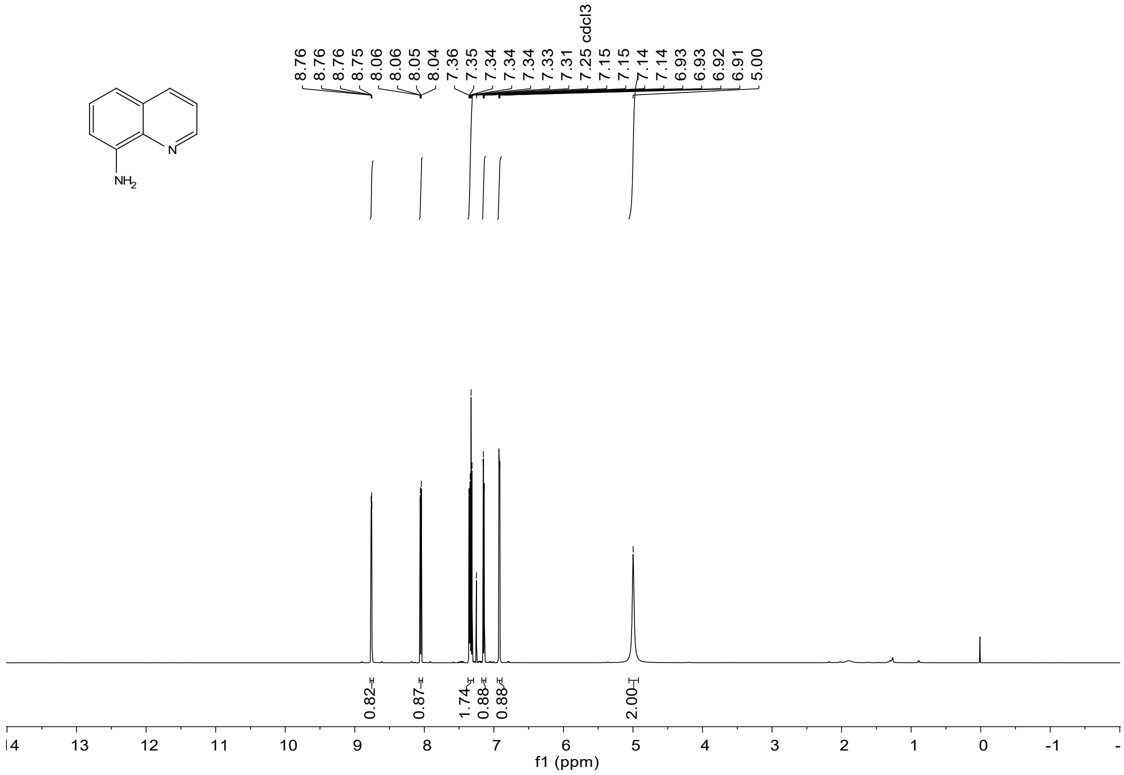
^{1}H NMR spectrum of 2e



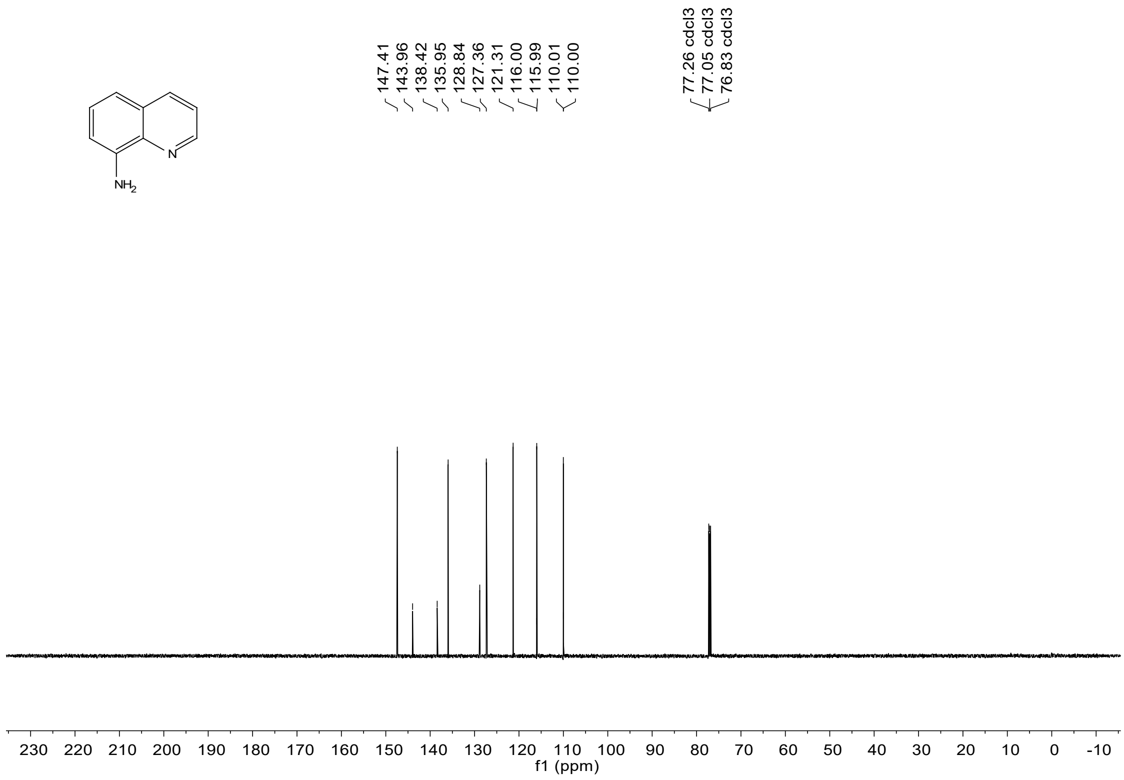
^{13}C NMR spectrum of 2e



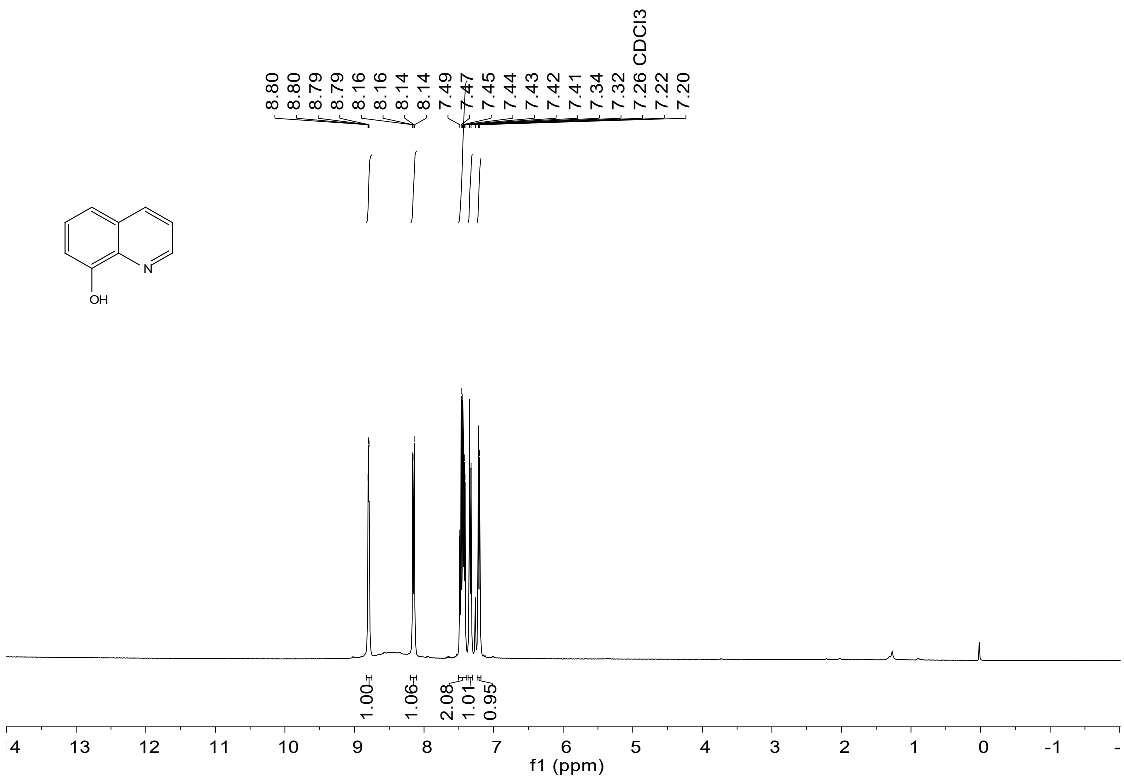
[1]H NMR spectrum of 2f



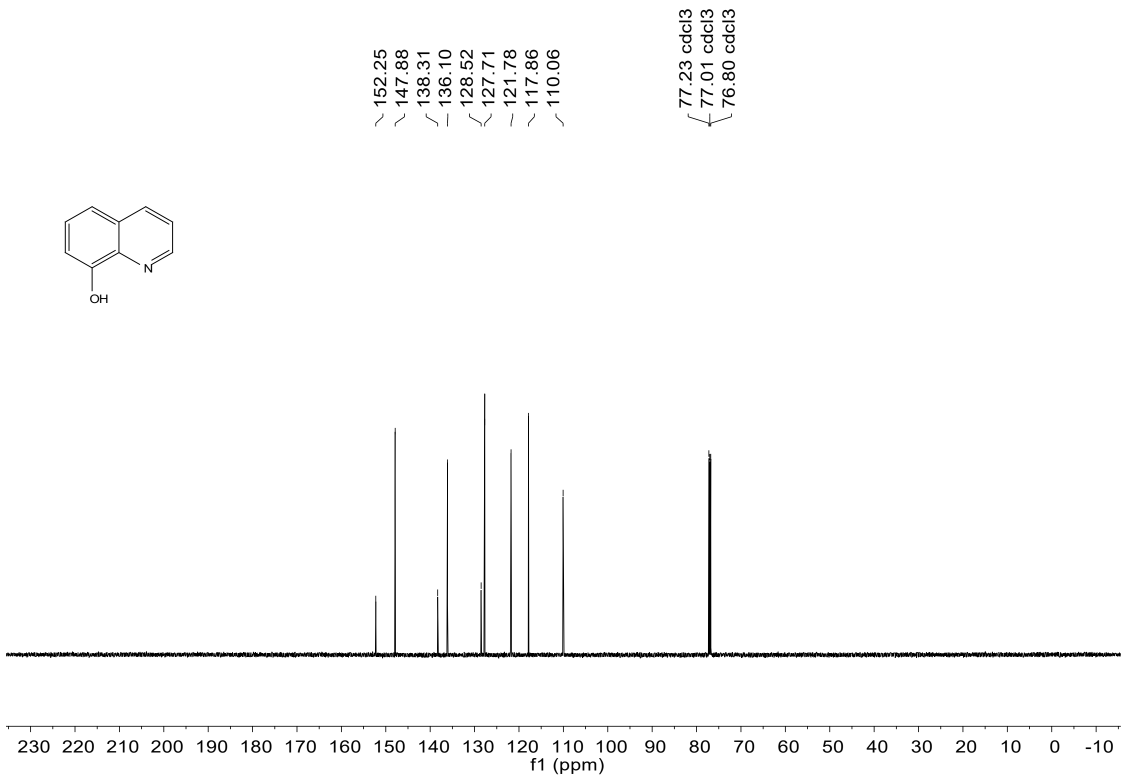
[13]C NMR spectrum of 2f



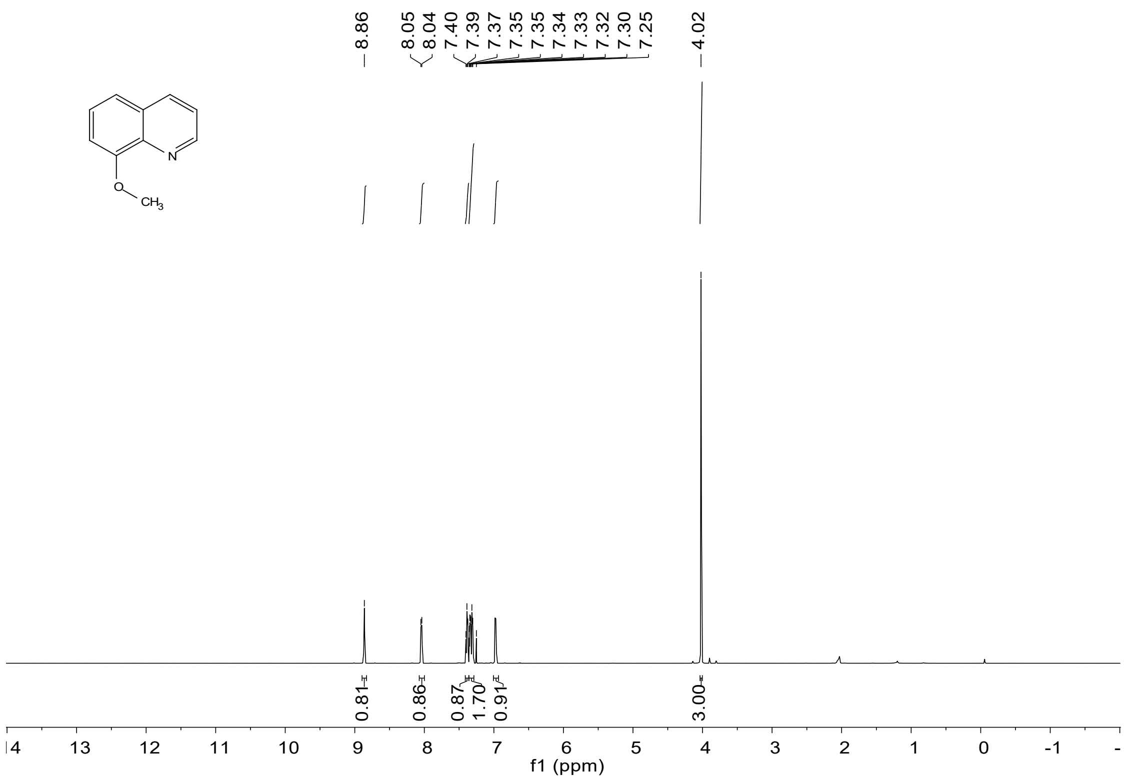
^{1}H NMR spectrum of 2g



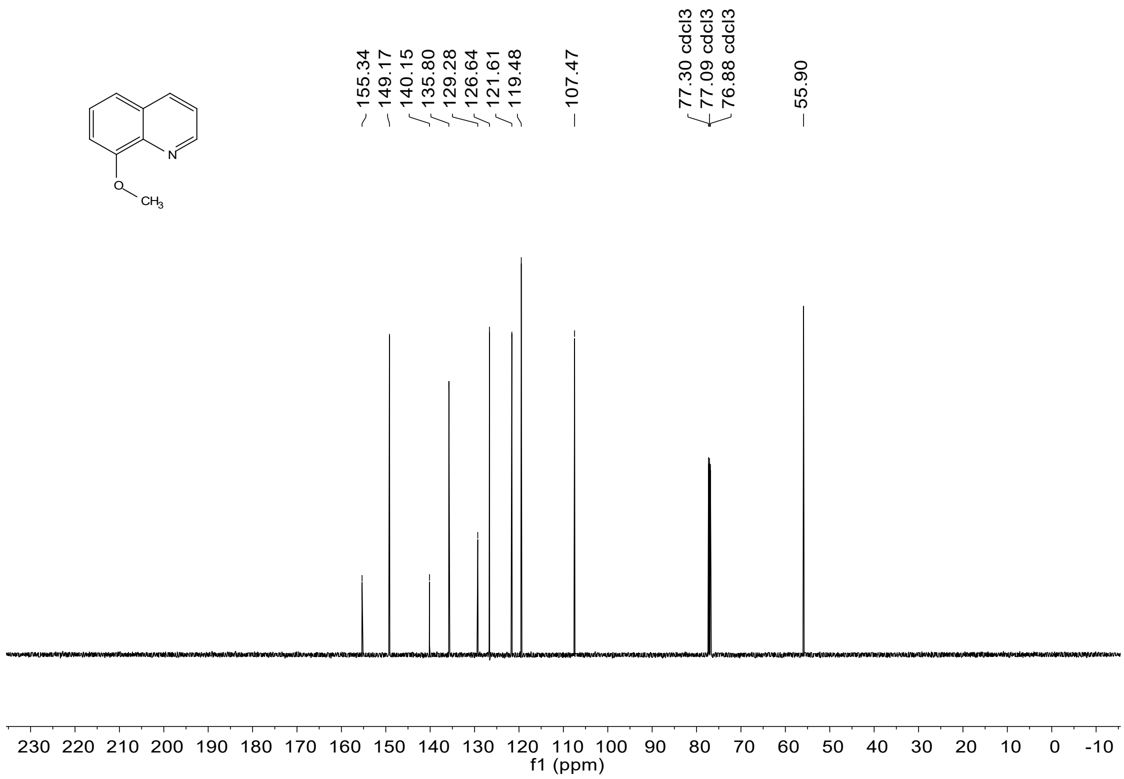
^{13}C NMR spectrum of 2g



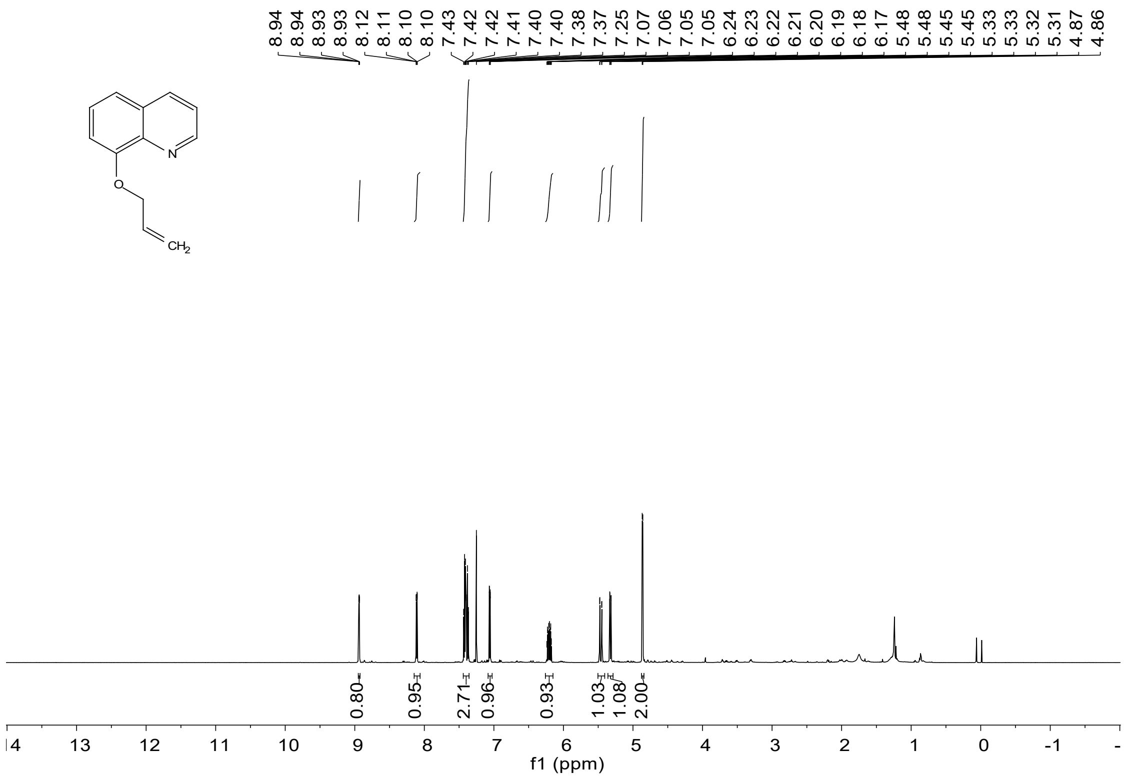
^{1}H NMR spectrum of 2h



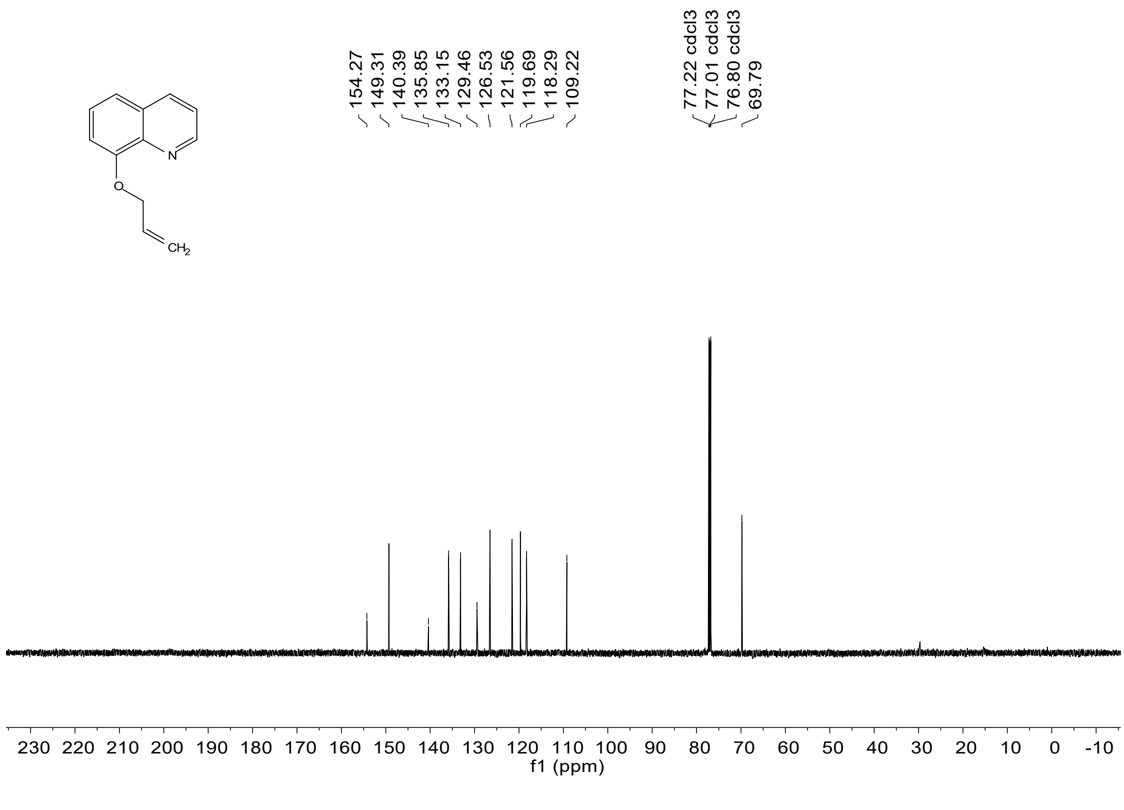
^{13}C NMR spectrum of 2h



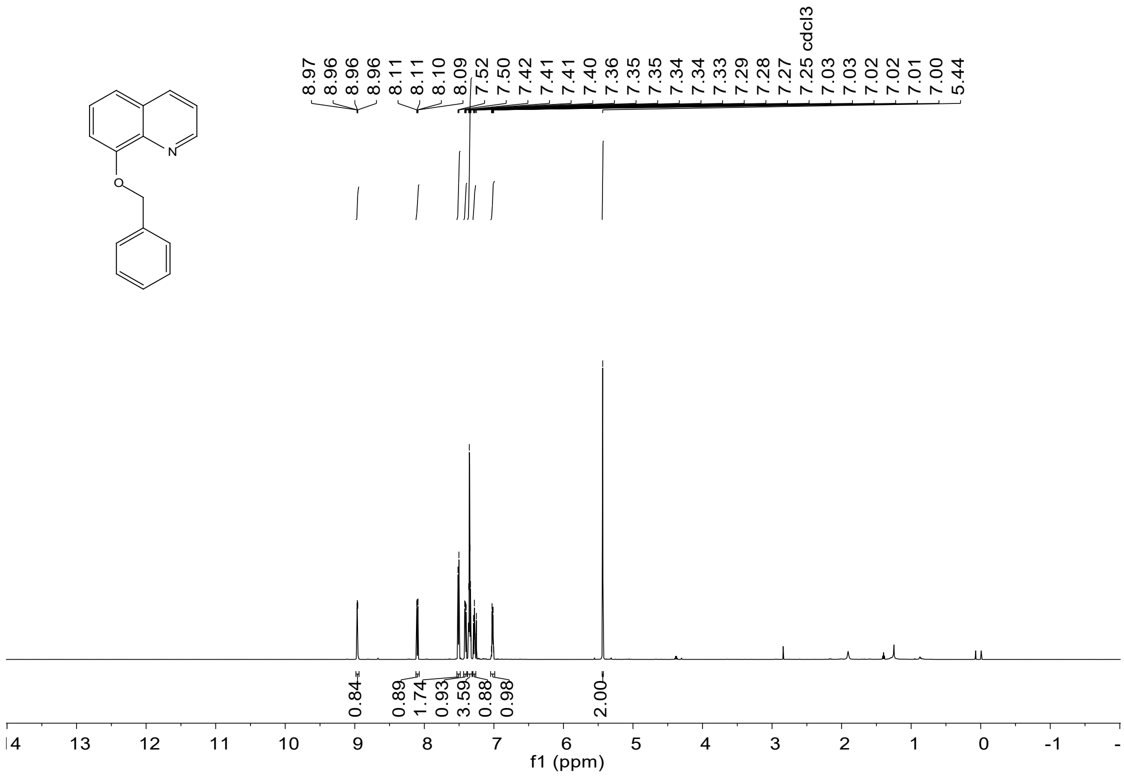
^{1}H NMR spectrum of 2i



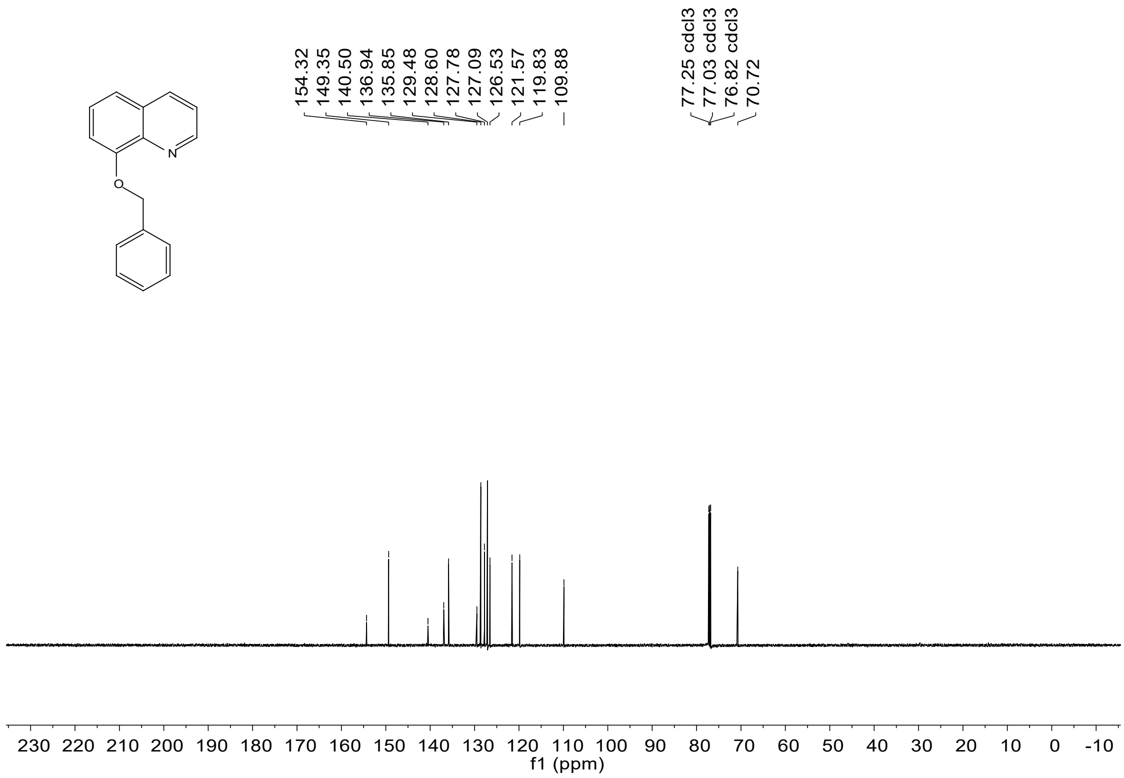
^{13}C NMR spectrum of 2i



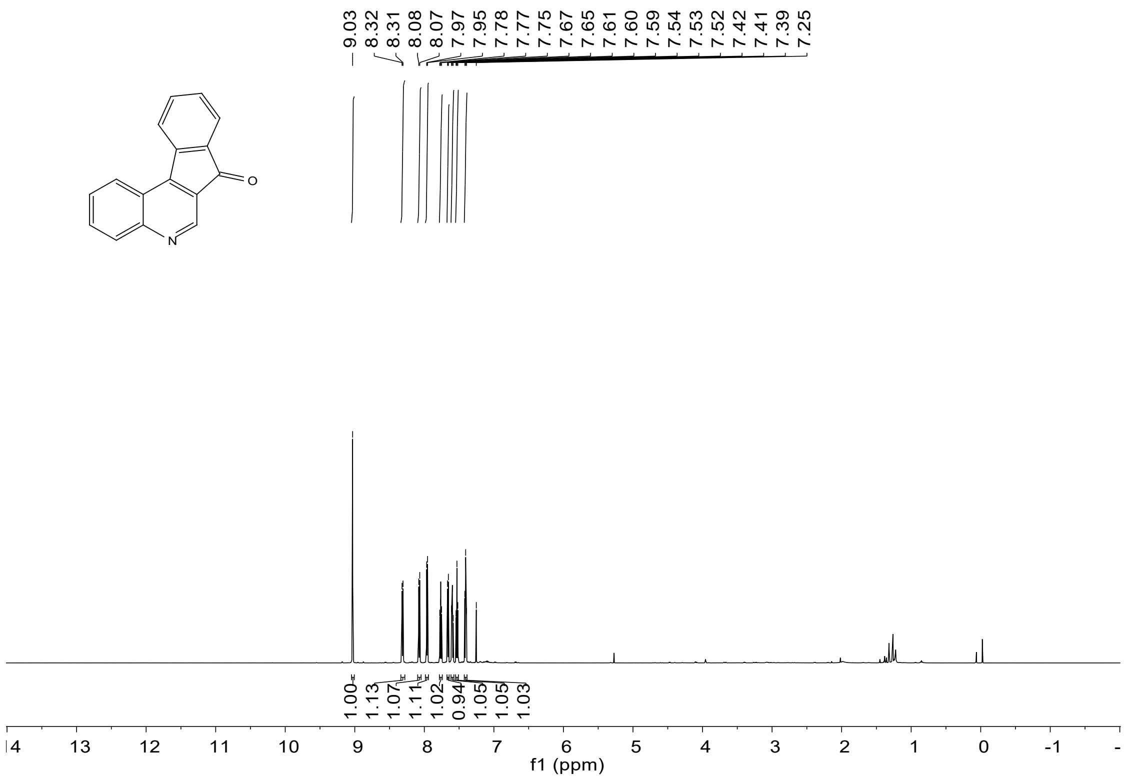
[1]H NMR spectrum of 2j



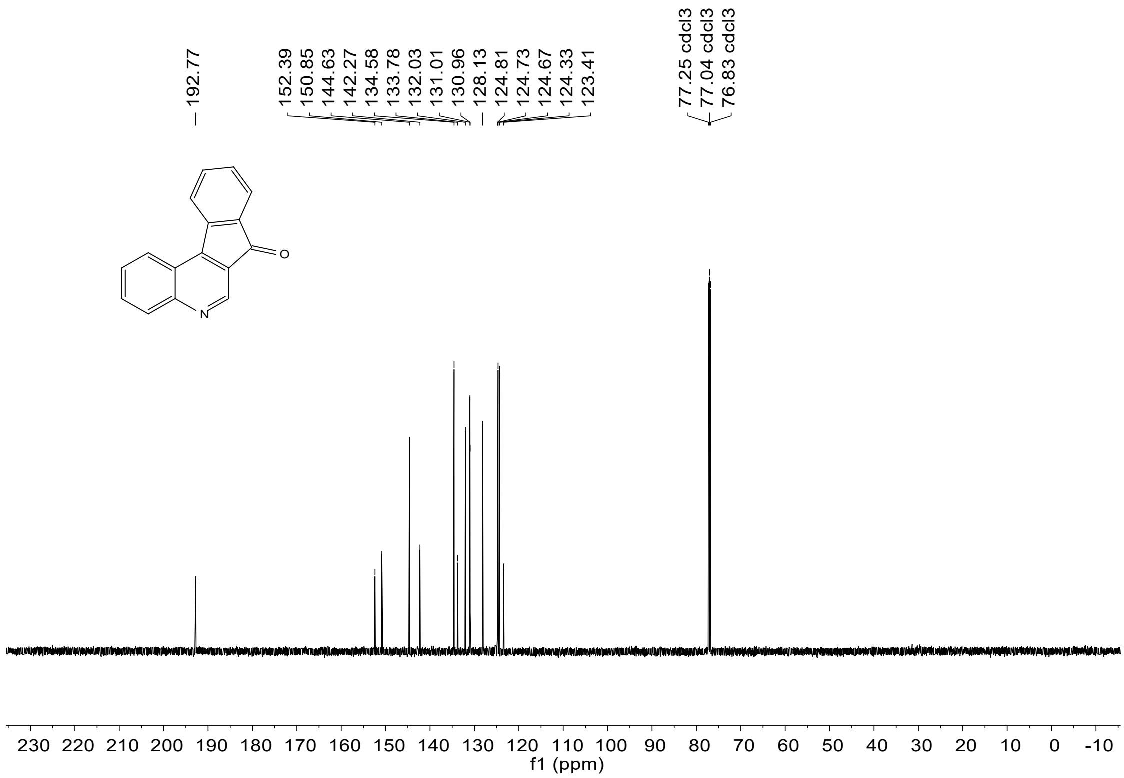
[13]C NMR spectrum of 2j



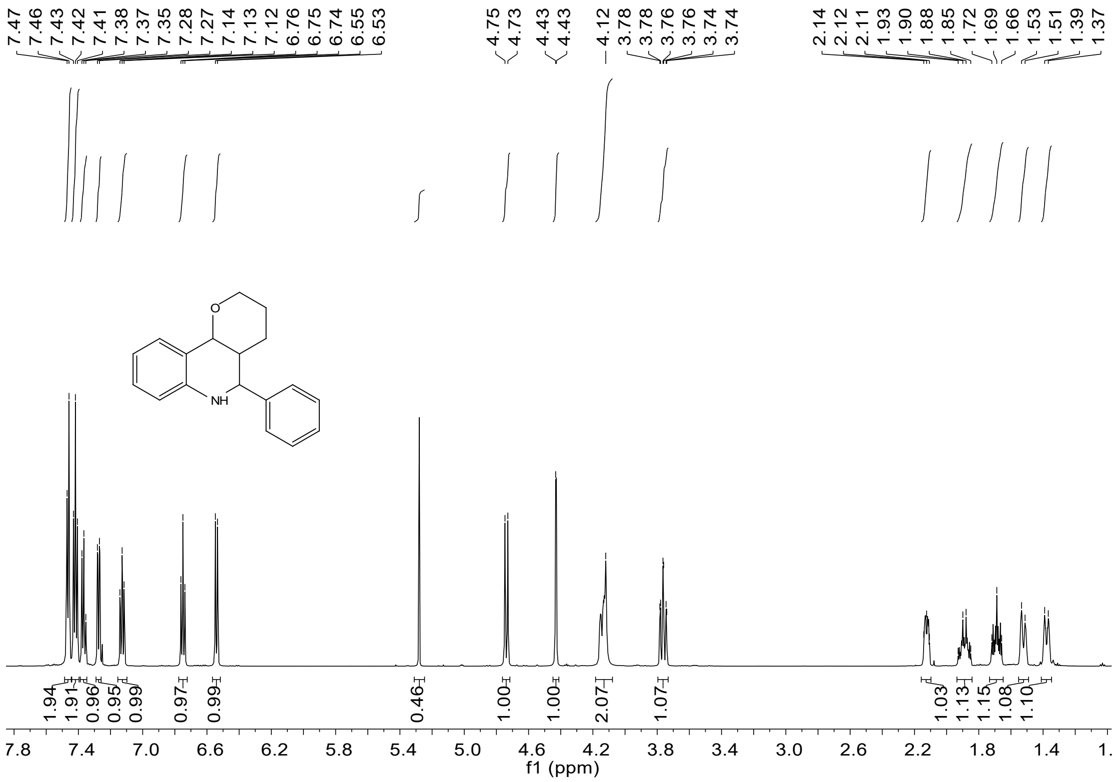
[1]H NMR spectrum of 2k



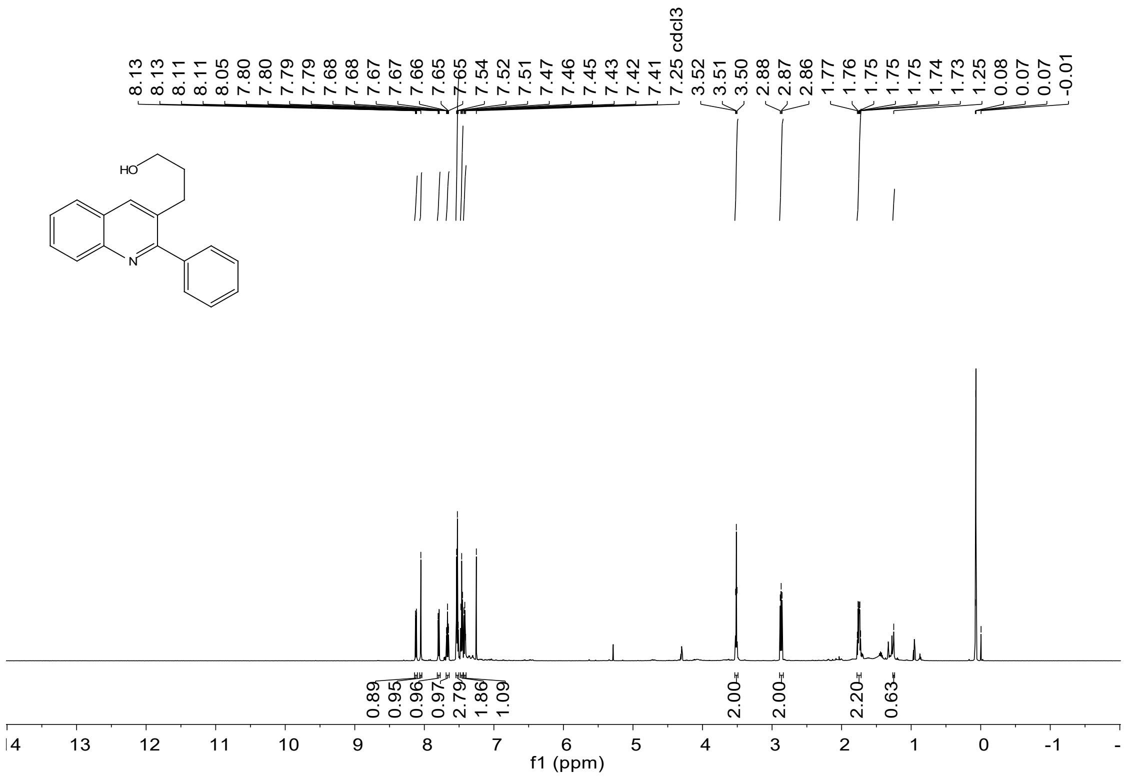
[13]C NMR spectrum of 2k



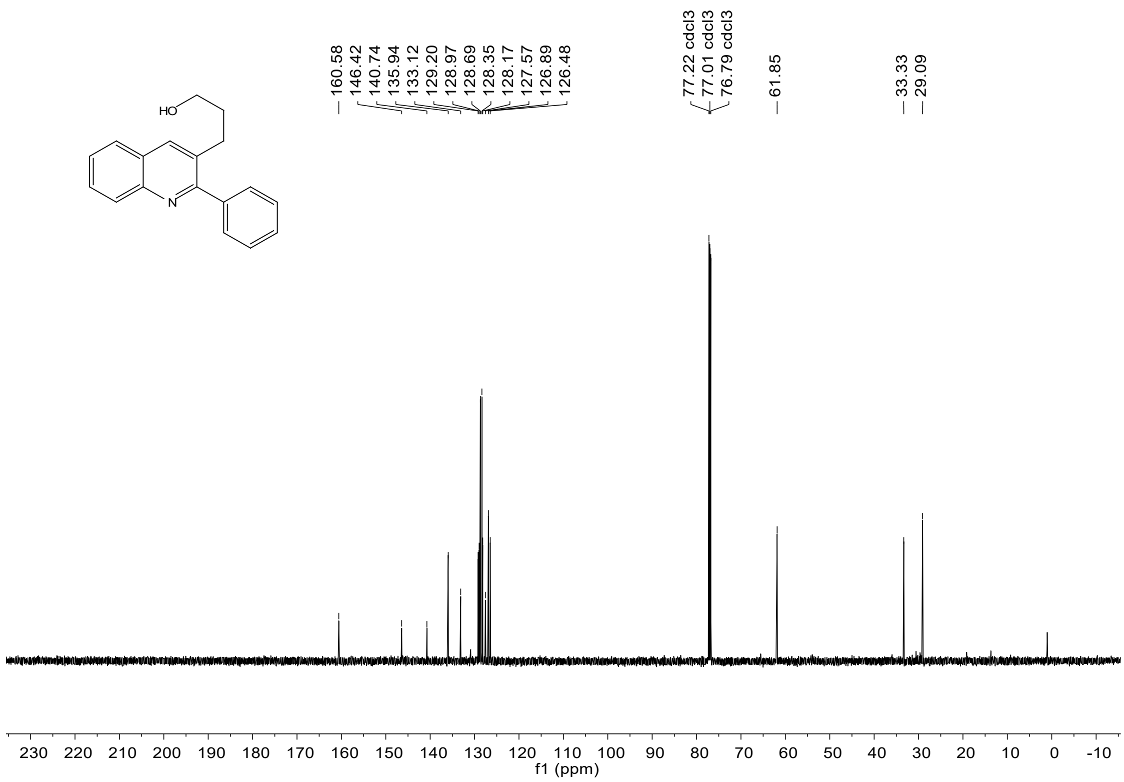
^{1}H NMR spectrum of 1l



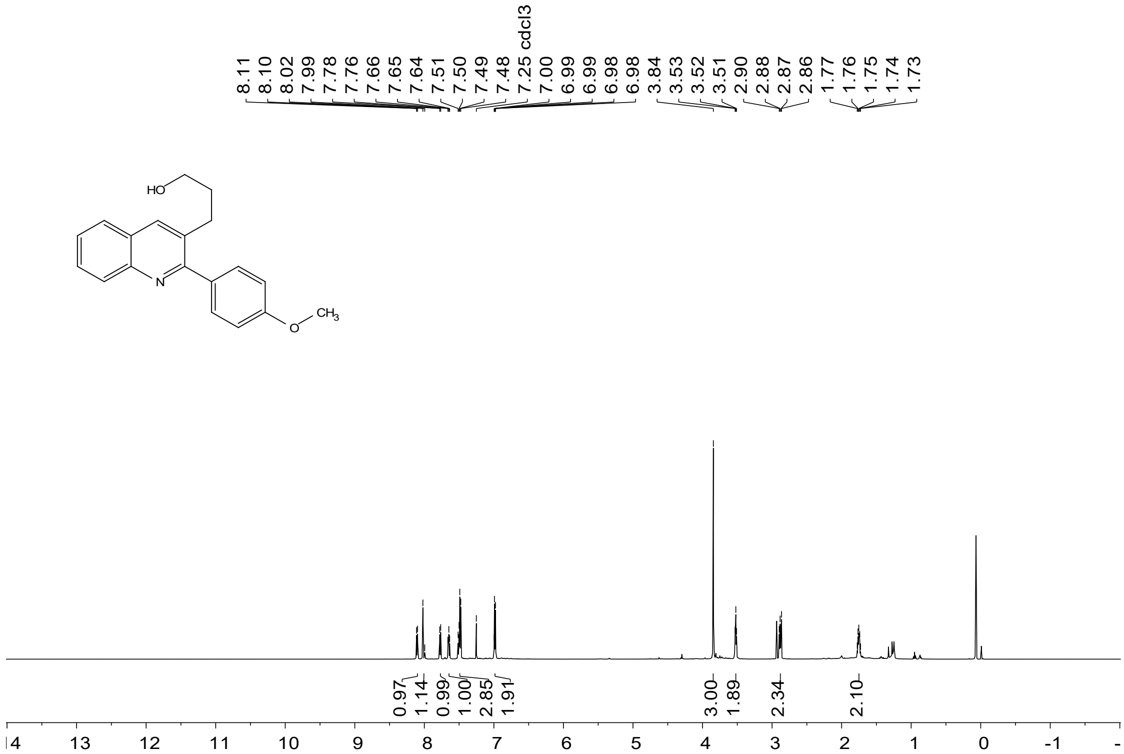
^{1}H NMR spectrum of 2l



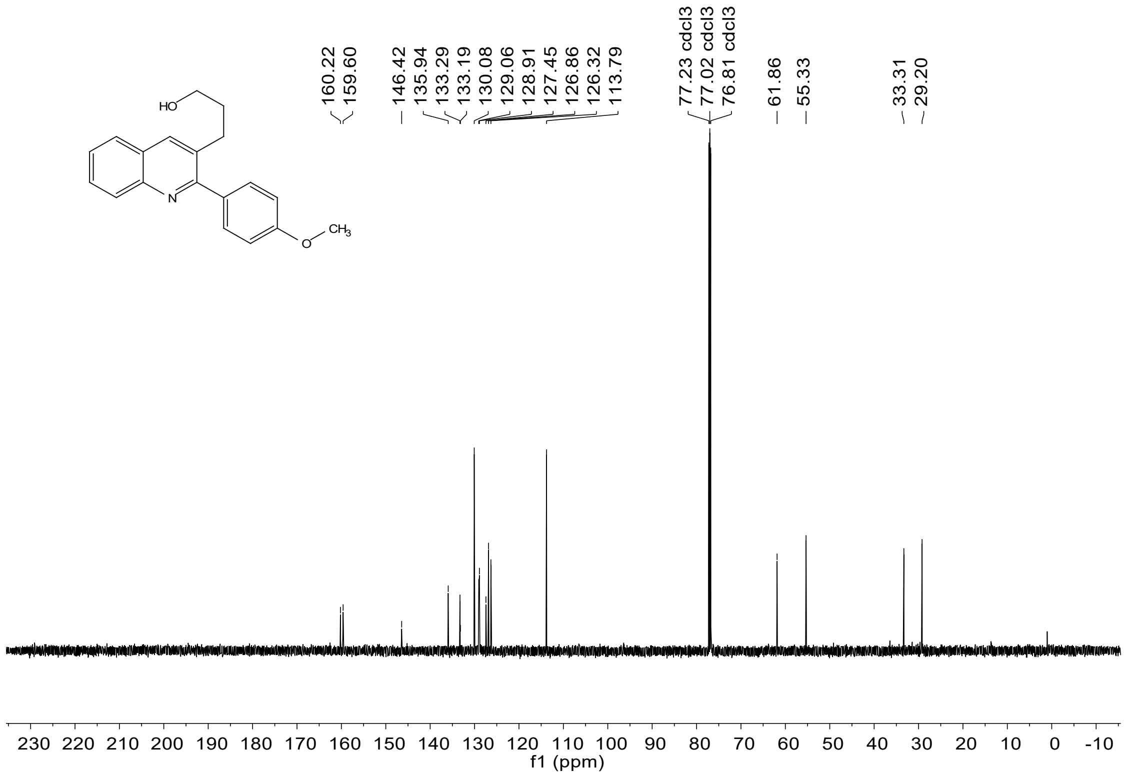
^{13}C NMR spectrum of 2l



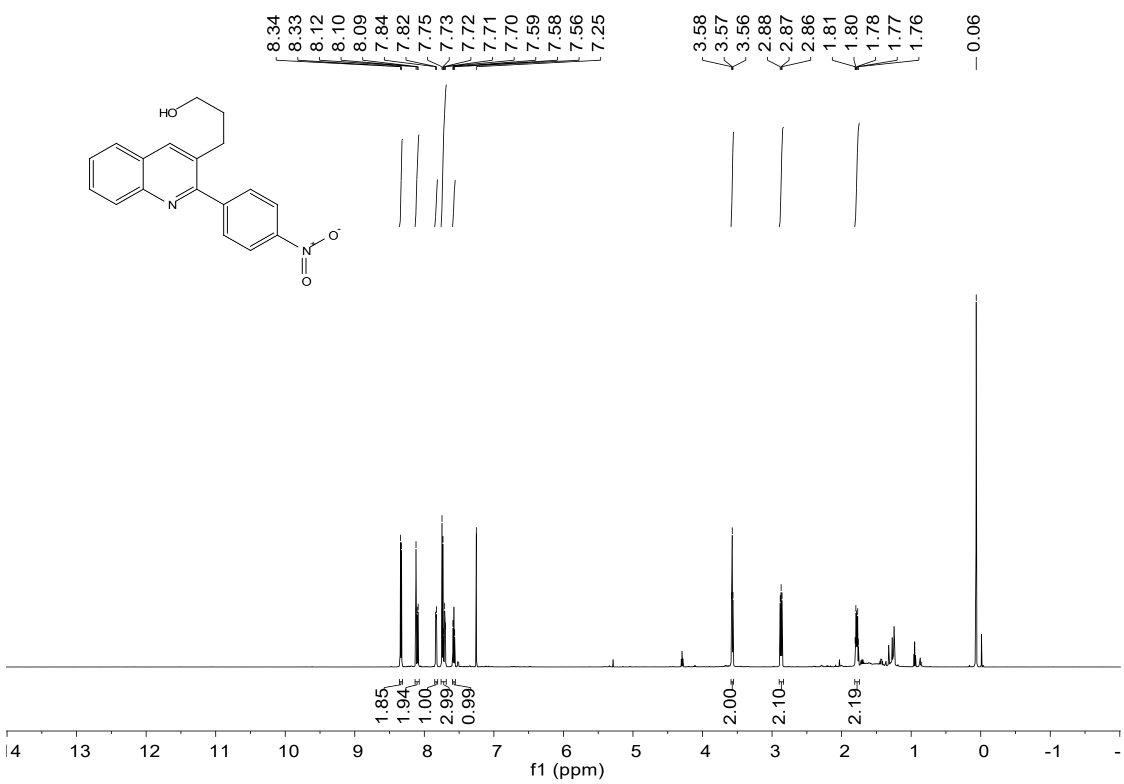
[1]H NMR spectrum of 2m



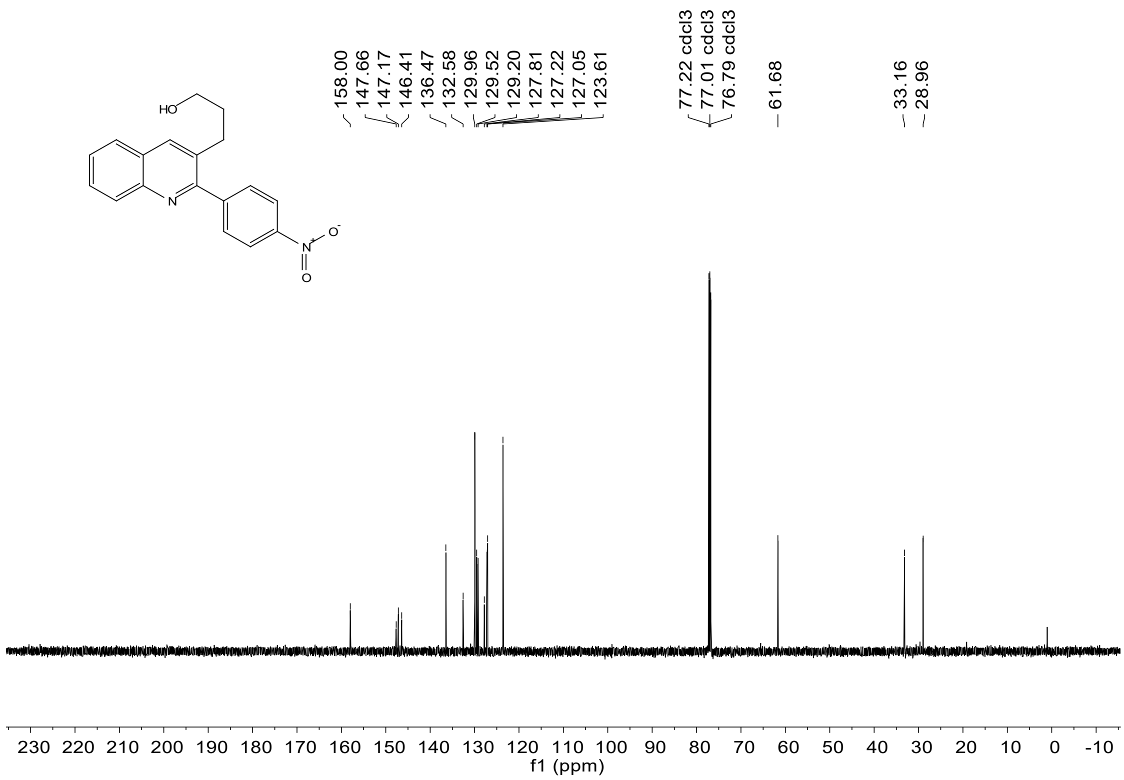
^{13}C NMR spectrum of 2m



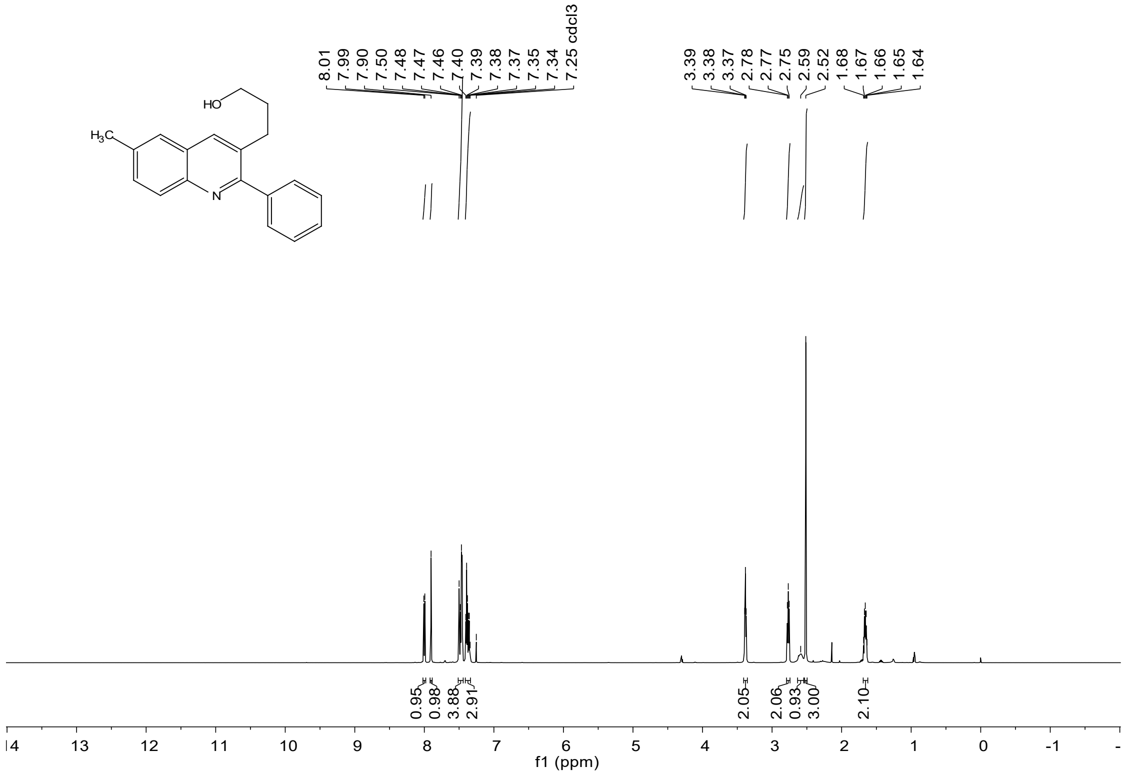
^{1}H NMR spectrum of 2n



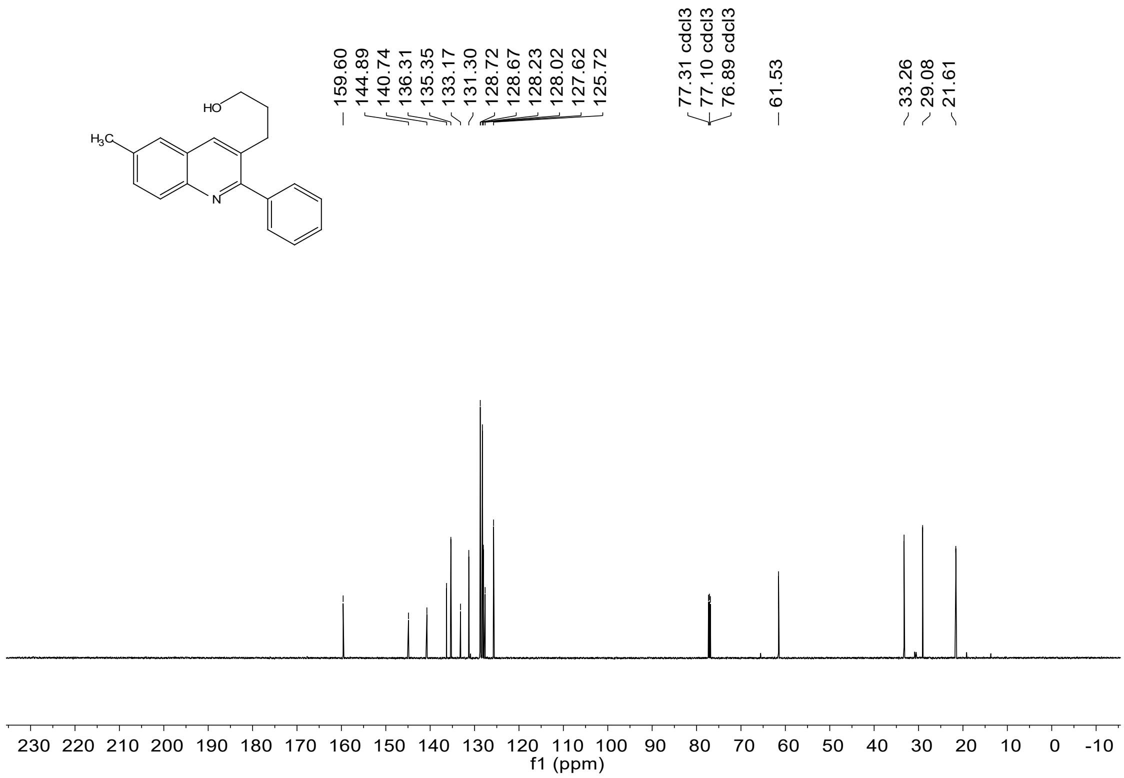
^{13}C NMR spectrum of 2n



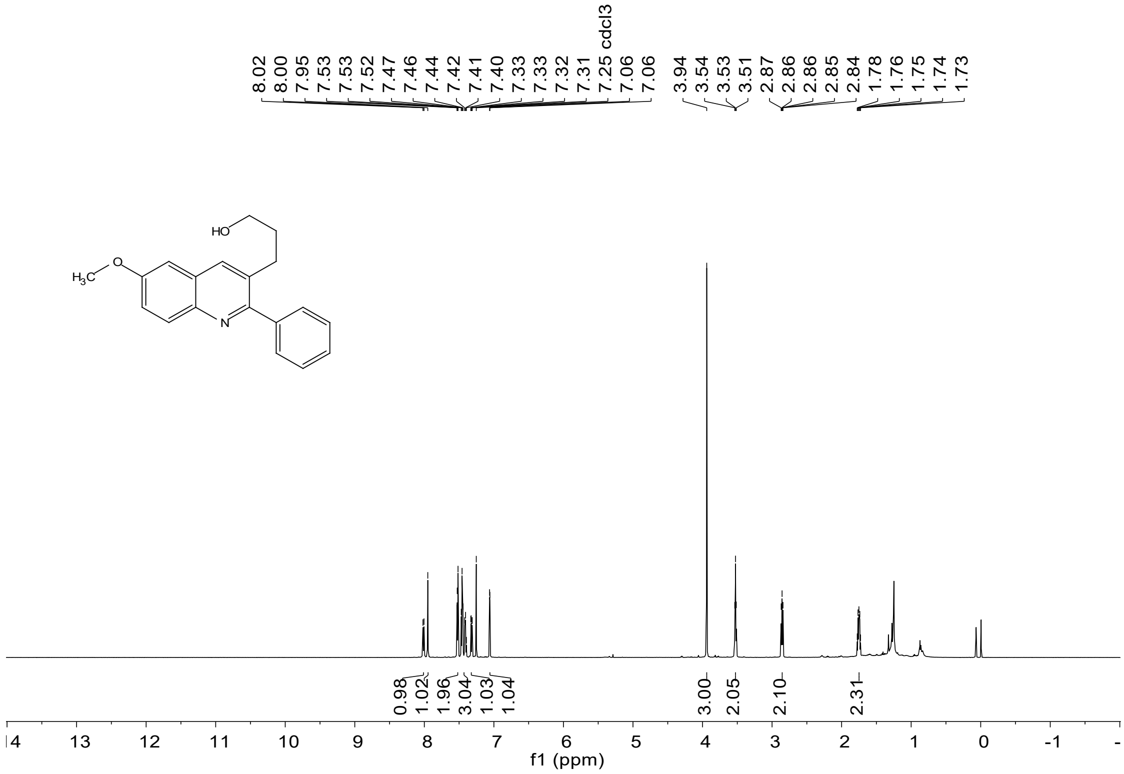
^{1}H NMR spectrum of 2o



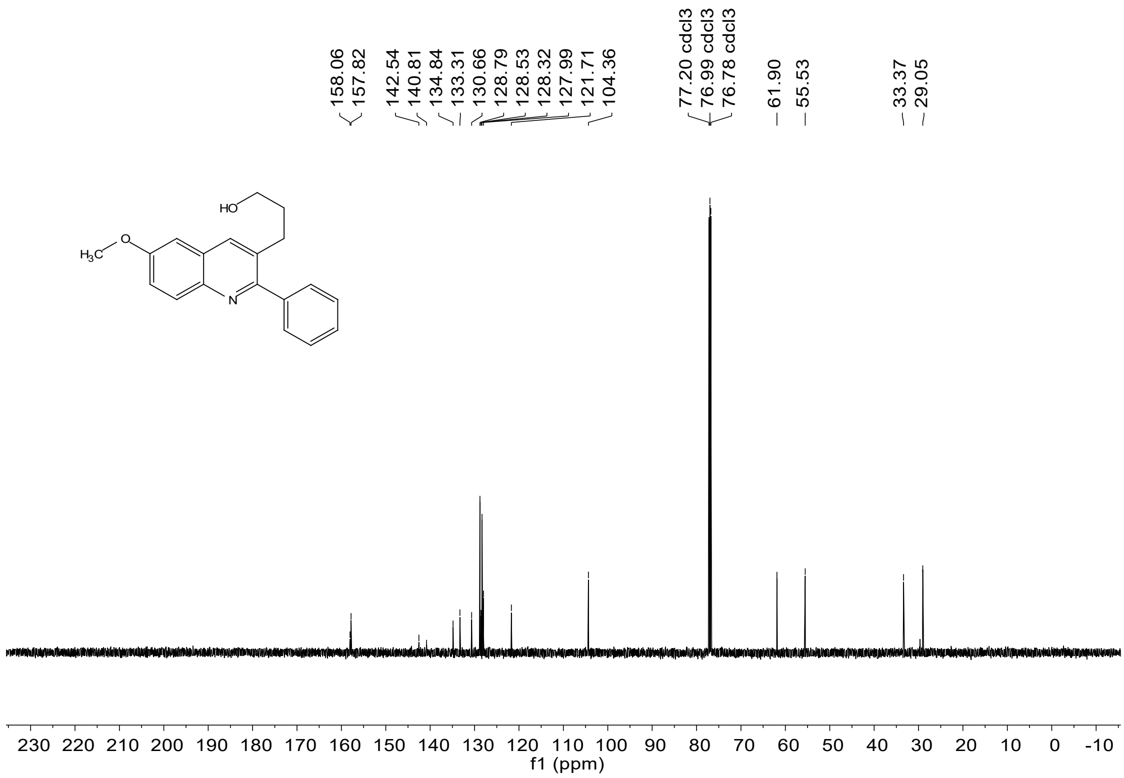
^{13}C NMR spectrum of 2o



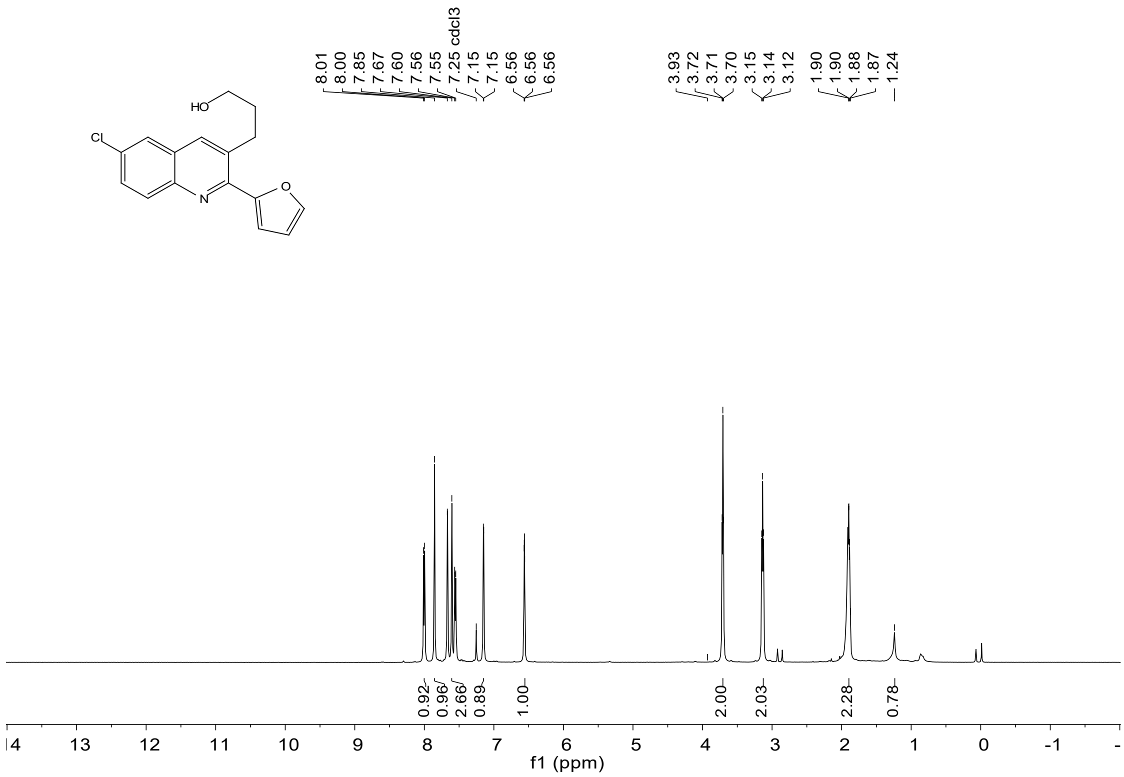
[1]H NMR spectrum of 2p



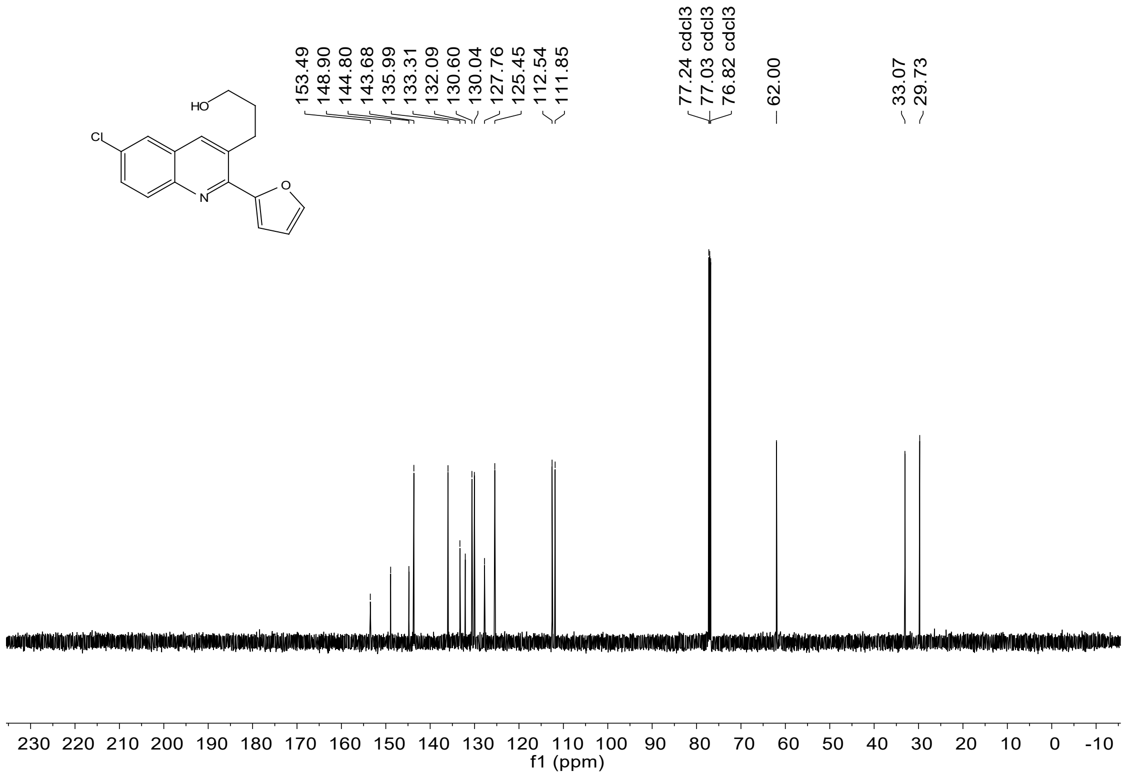
[13]C NMR spectrum of 2p



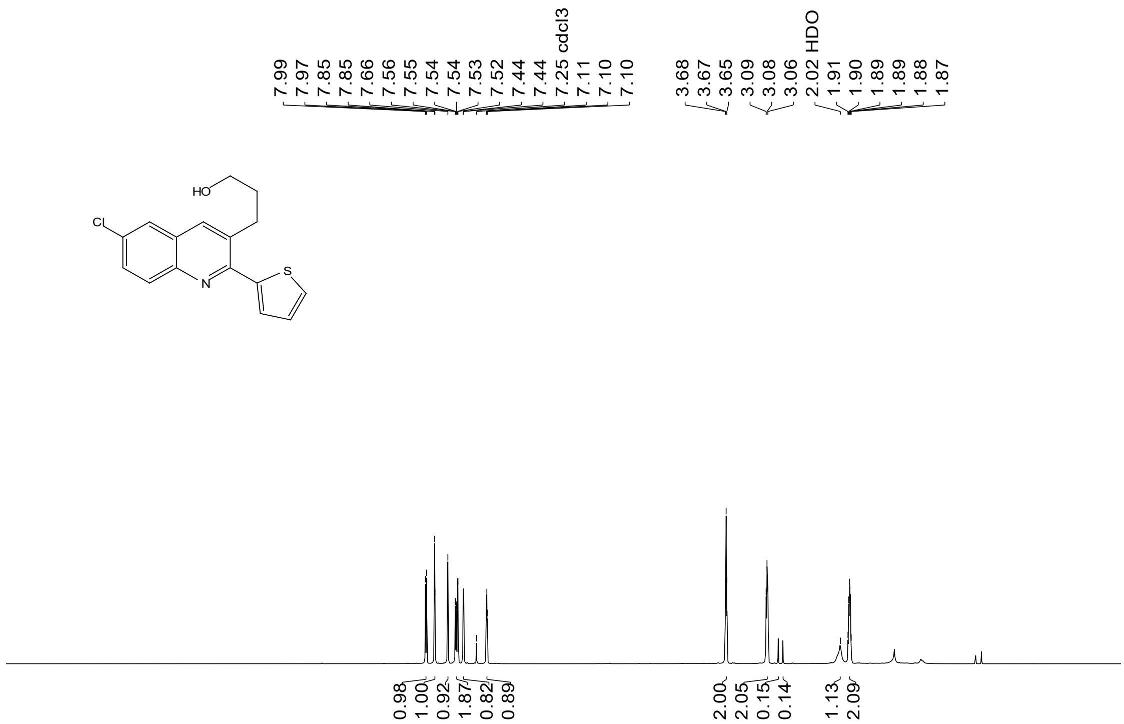
^{1}H NMR spectrum of 2q



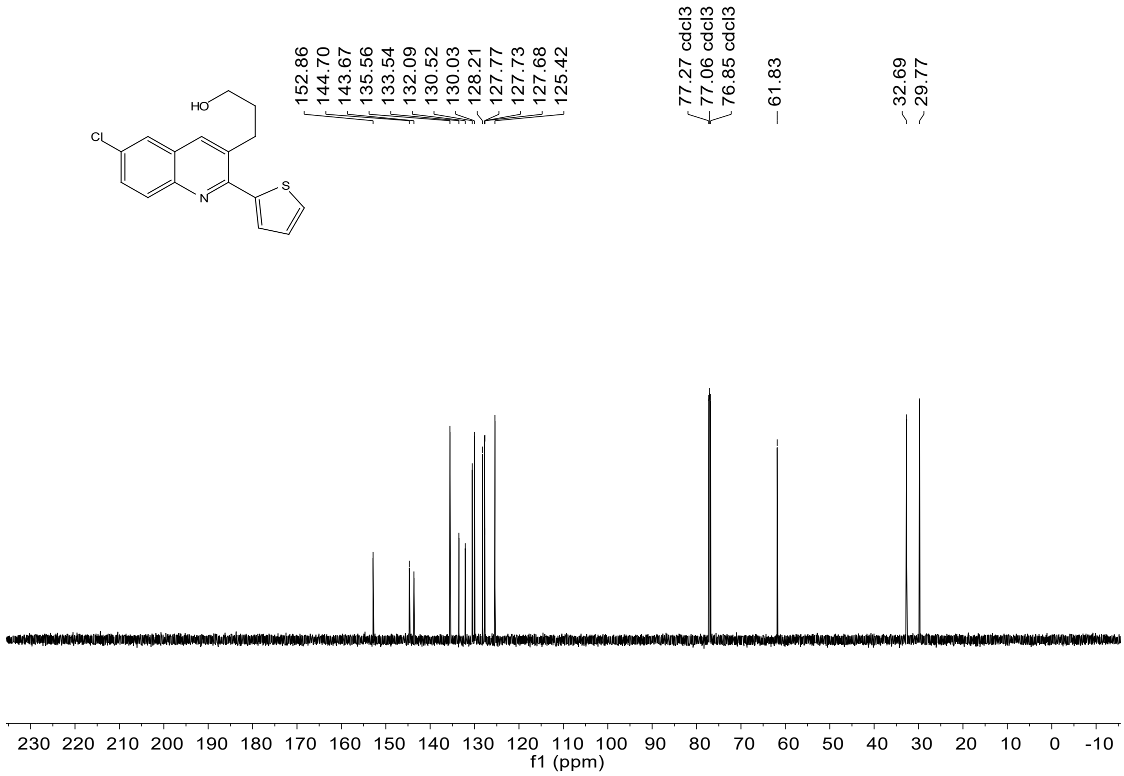
^{13}C NMR spectrum of 2q



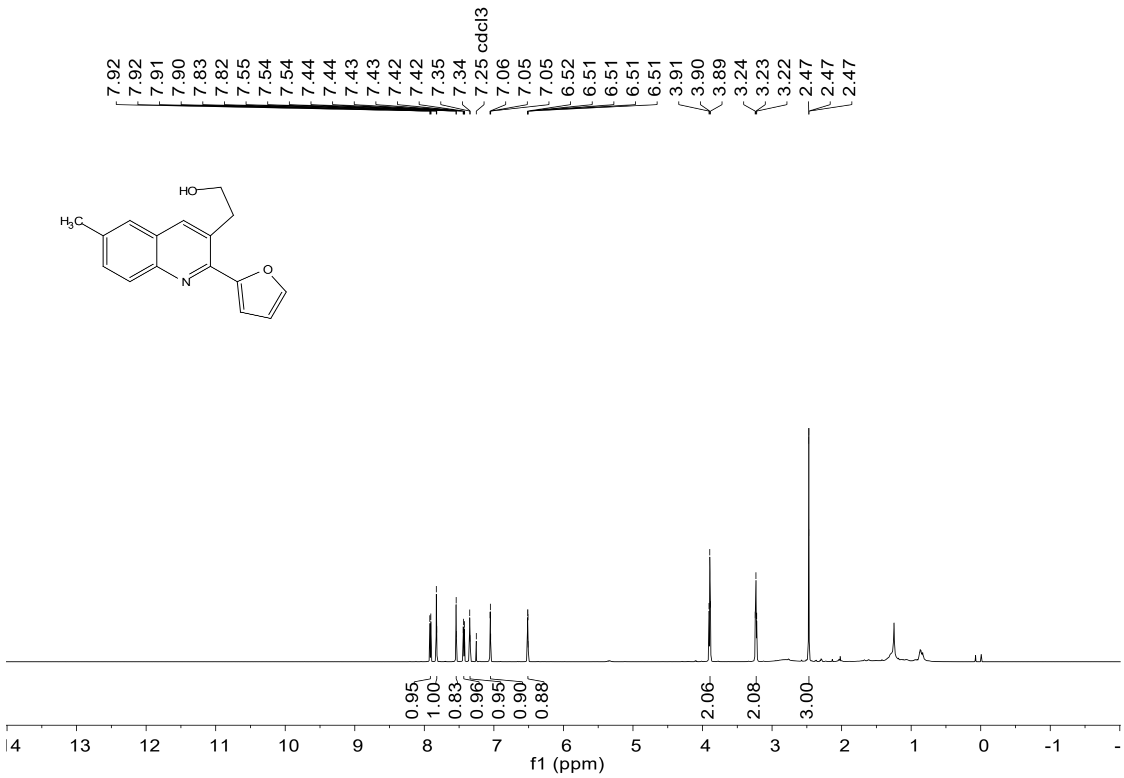
[1]H NMR spectrum of 2r



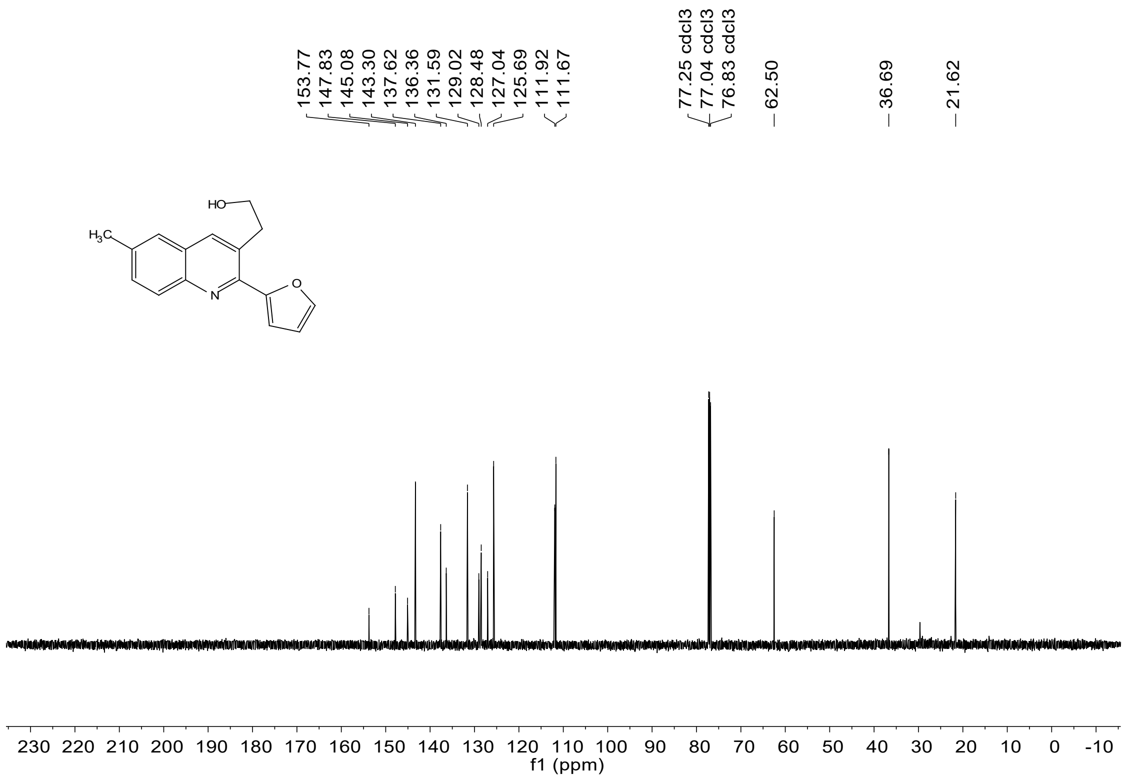
^{13}C NMR spectrum of 2r



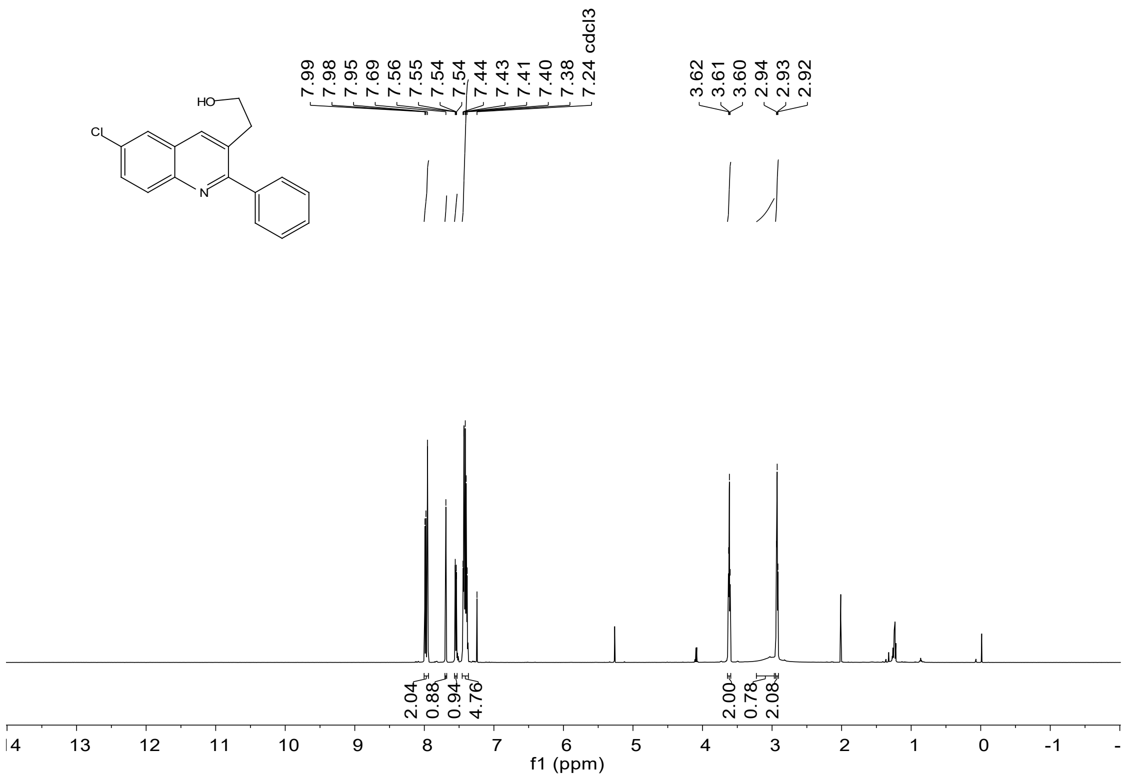
^{1}H NMR spectrum of 2s



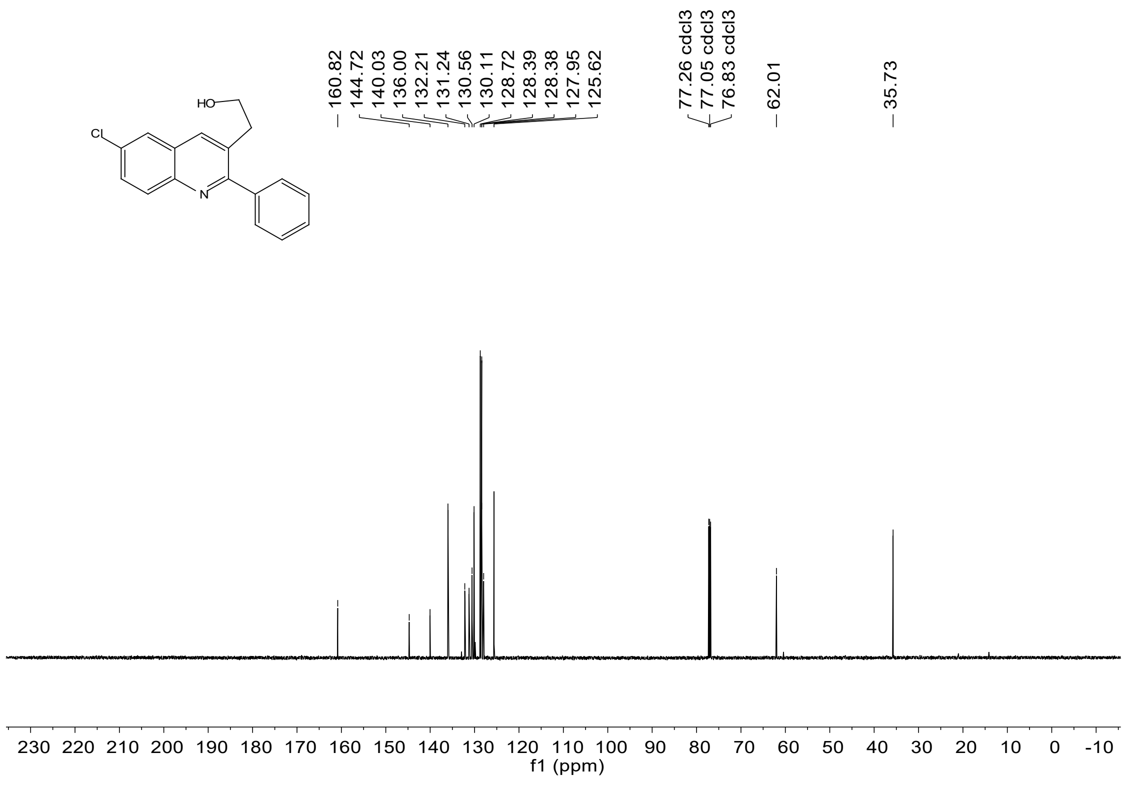
^{13}C NMR spectrum of 2s



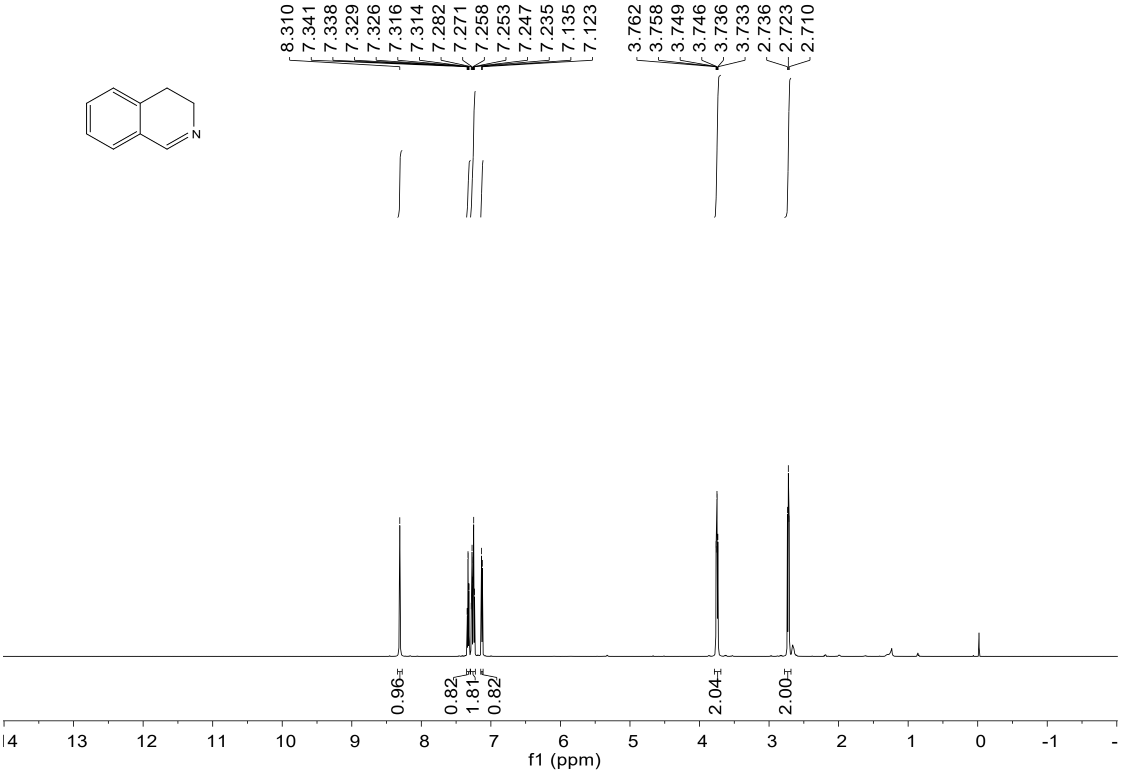
^{1}H NMR spectrum of 2t



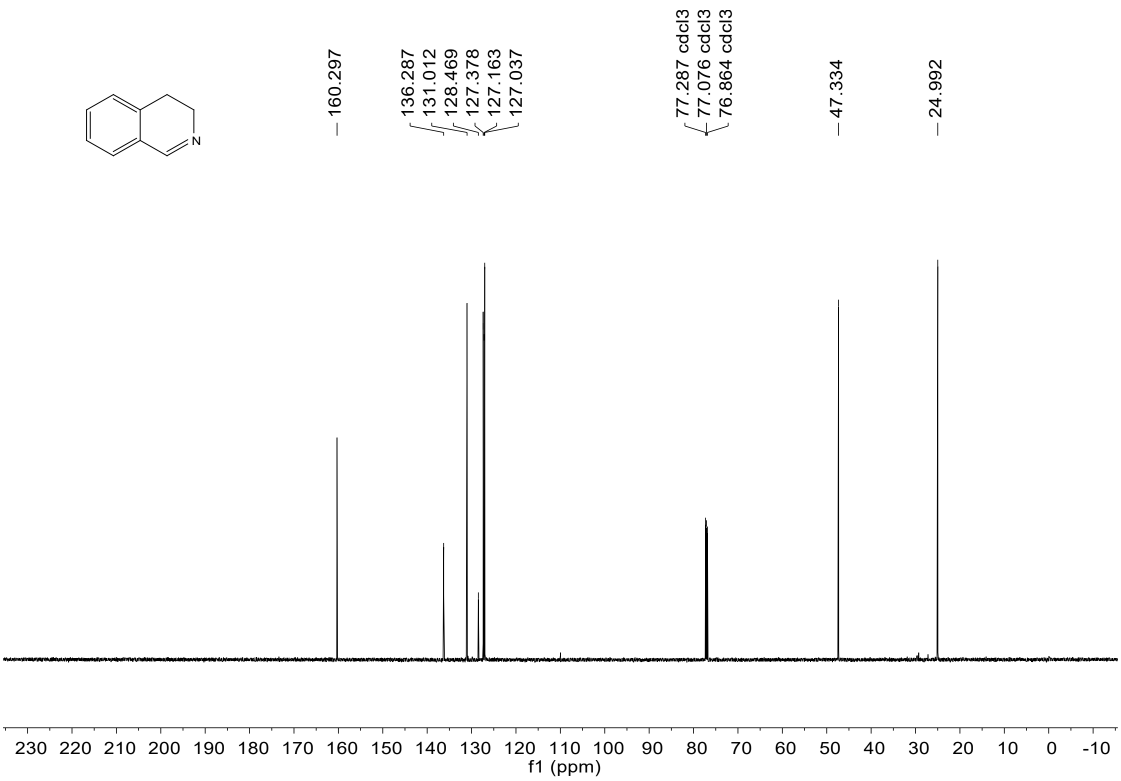
^{13}C NMR spectrum of 2t



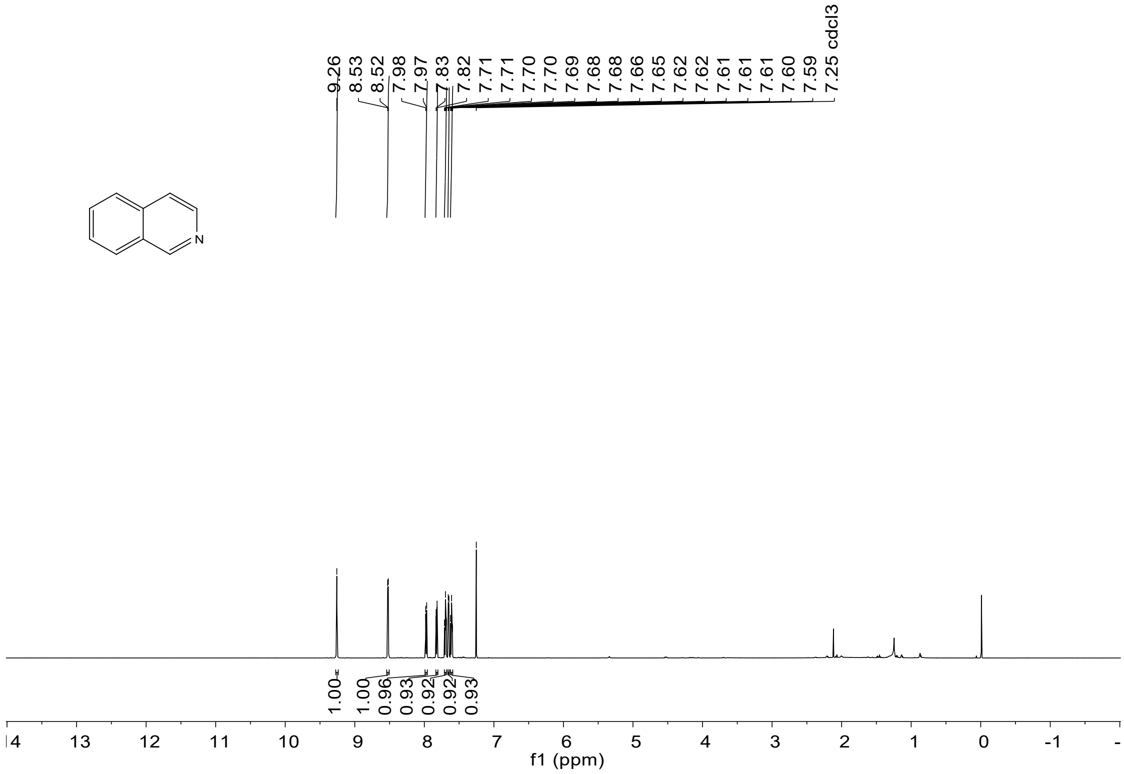
[1]H NMR spectrum of 4a



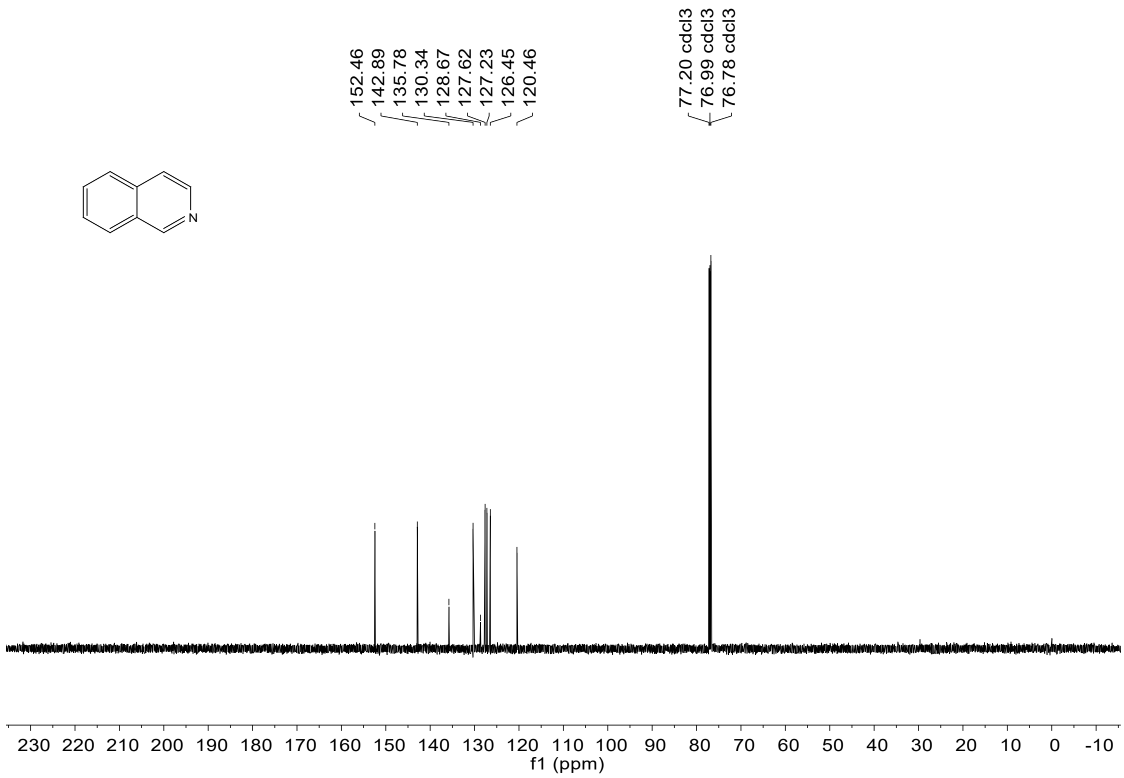
[13]C NMR spectrum of 4a



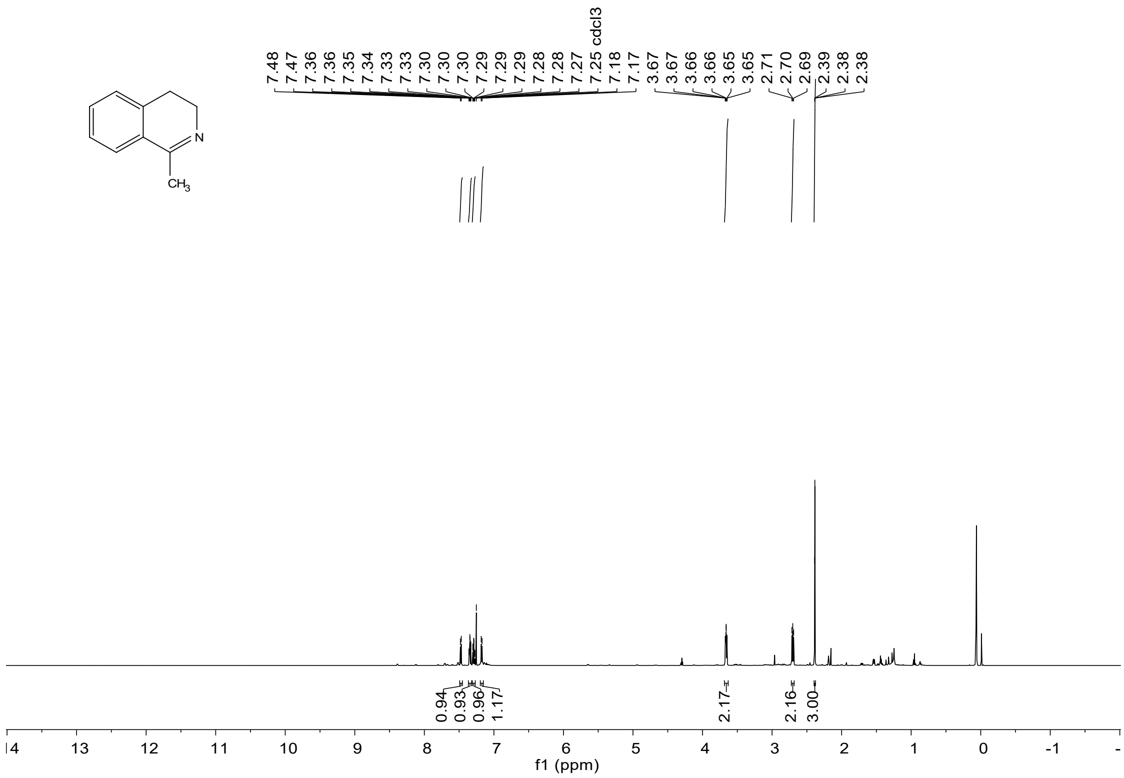
[1]H NMR spectrum of 4b



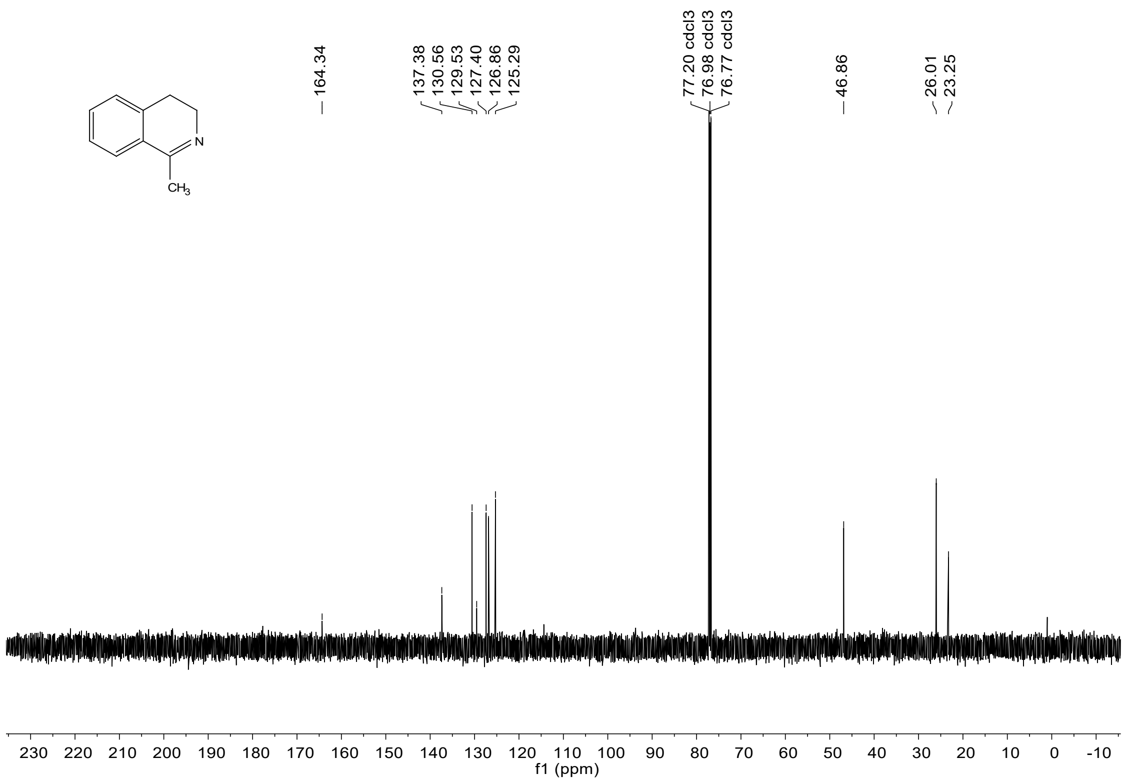
[13]C NMR spectrum of 4b



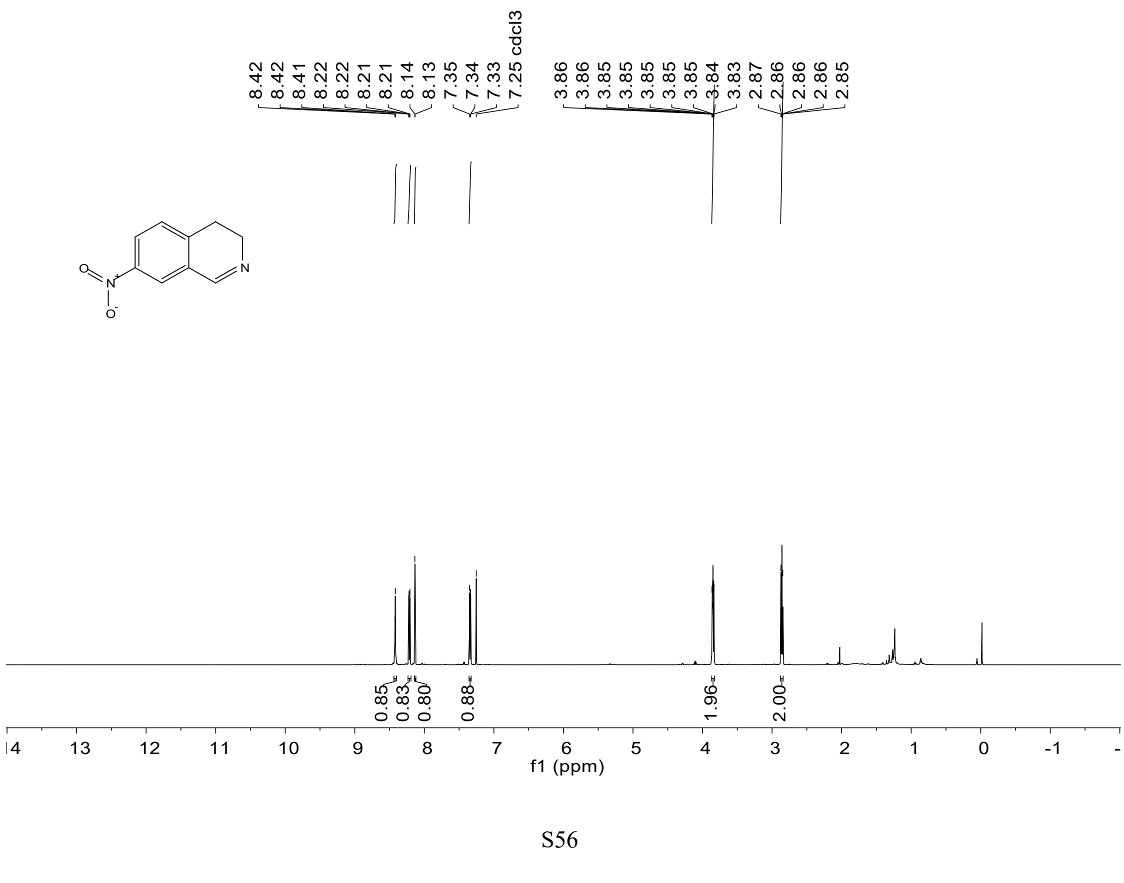
^{1}H NMR spectrum of 4c



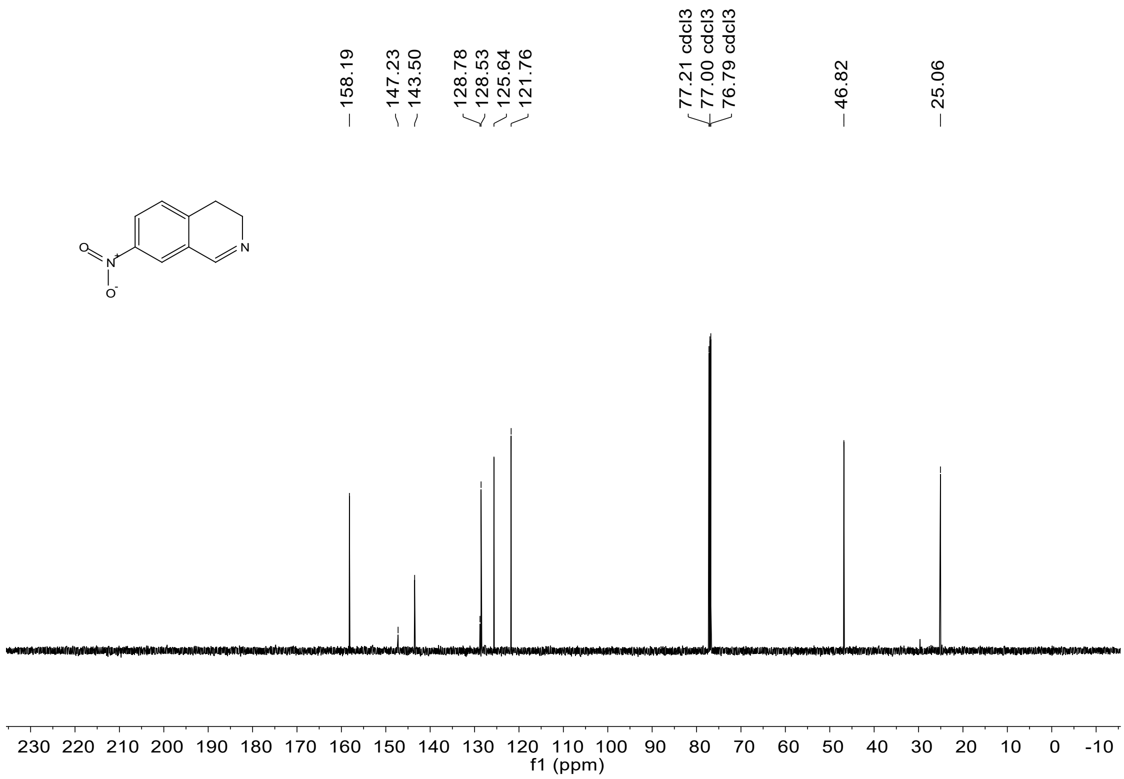
^{13}C NMR spectrum of 4c



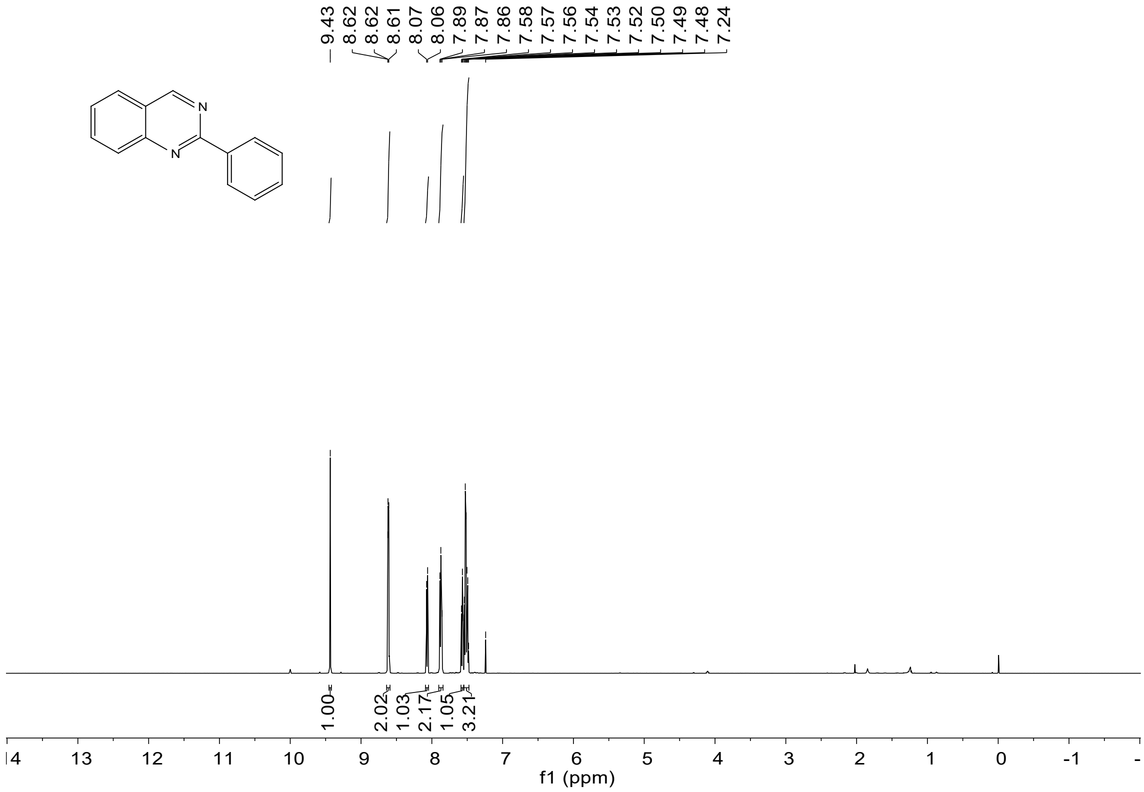
[1]H NMR spectrum of 4d



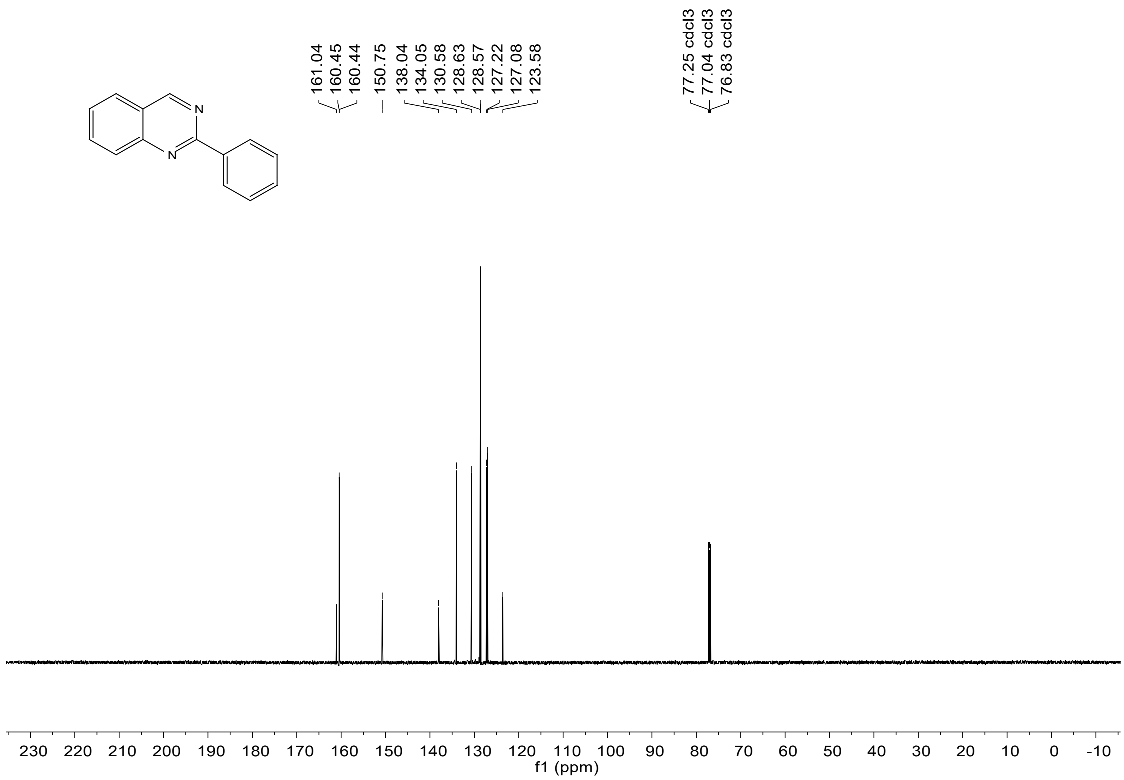
^{13}C NMR spectrum of 4d



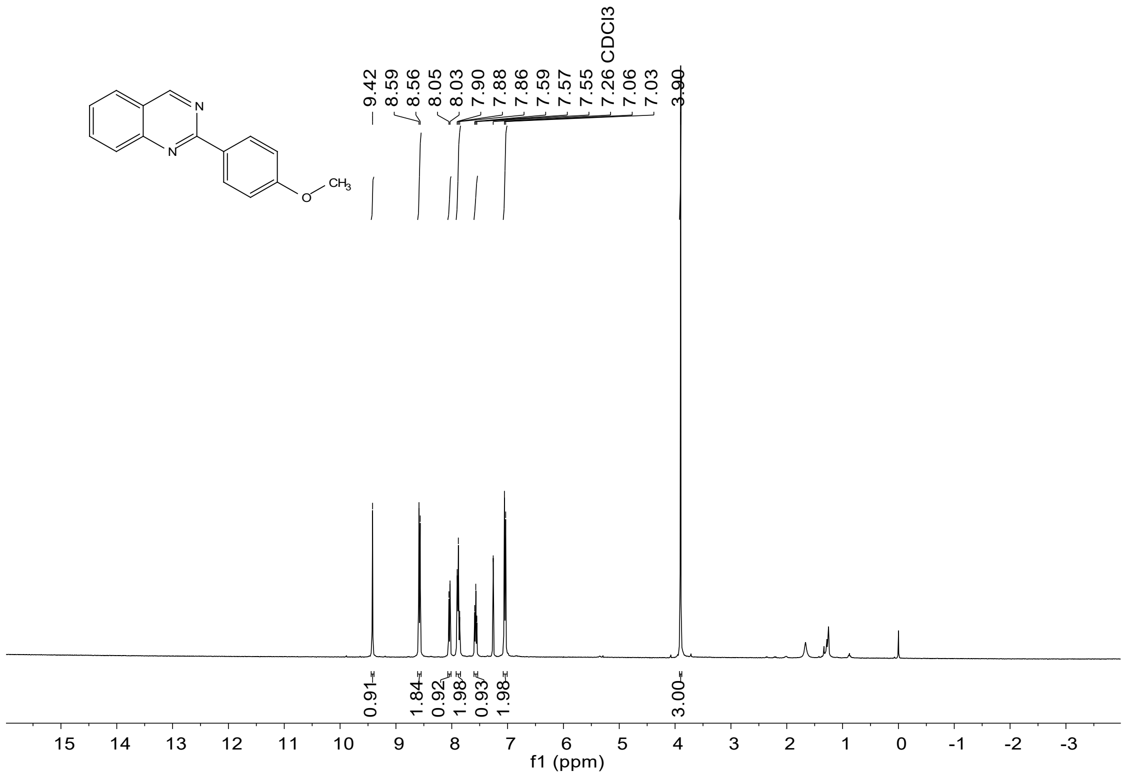
^{1}H NMR spectrum of 6a



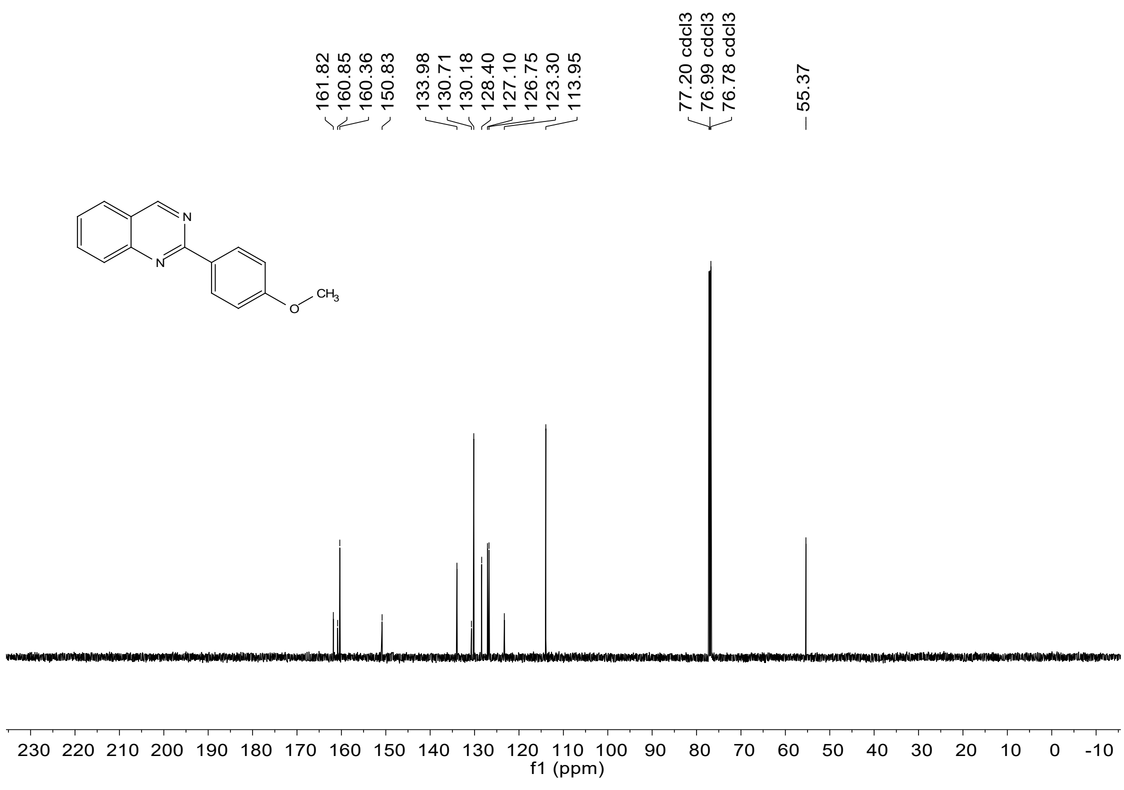
^{13}C NMR spectrum of 6a



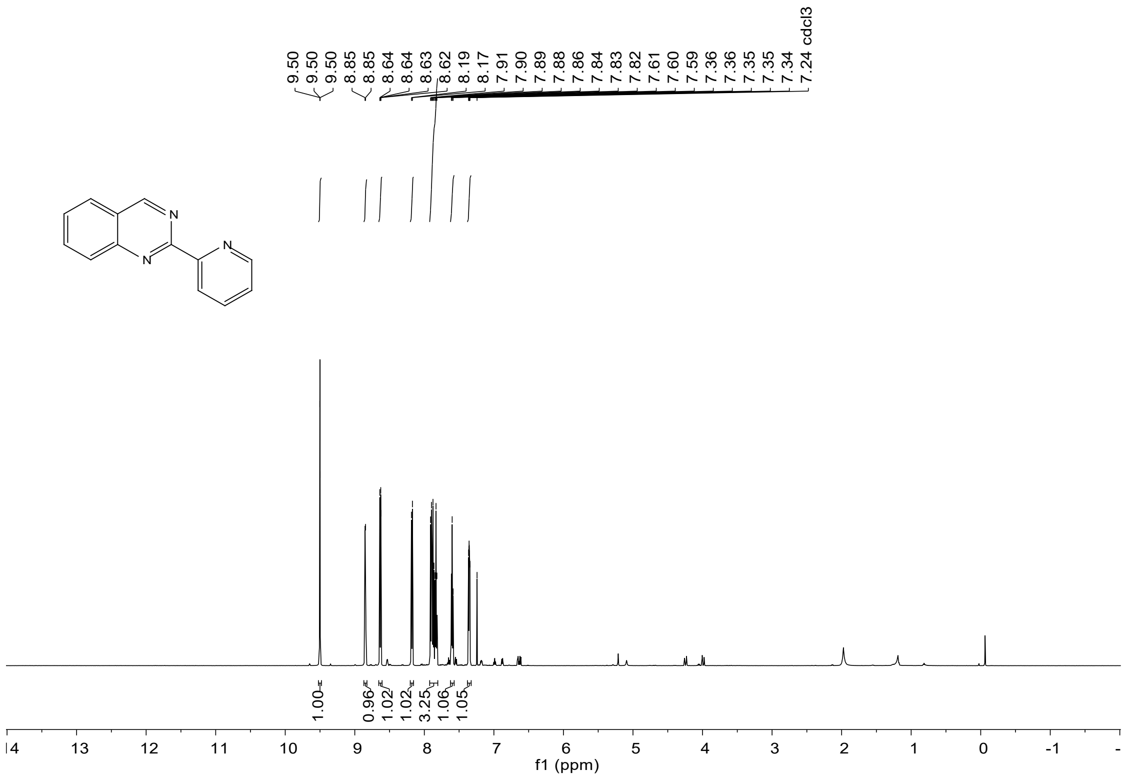
^{1}H NMR spectrum of 6b



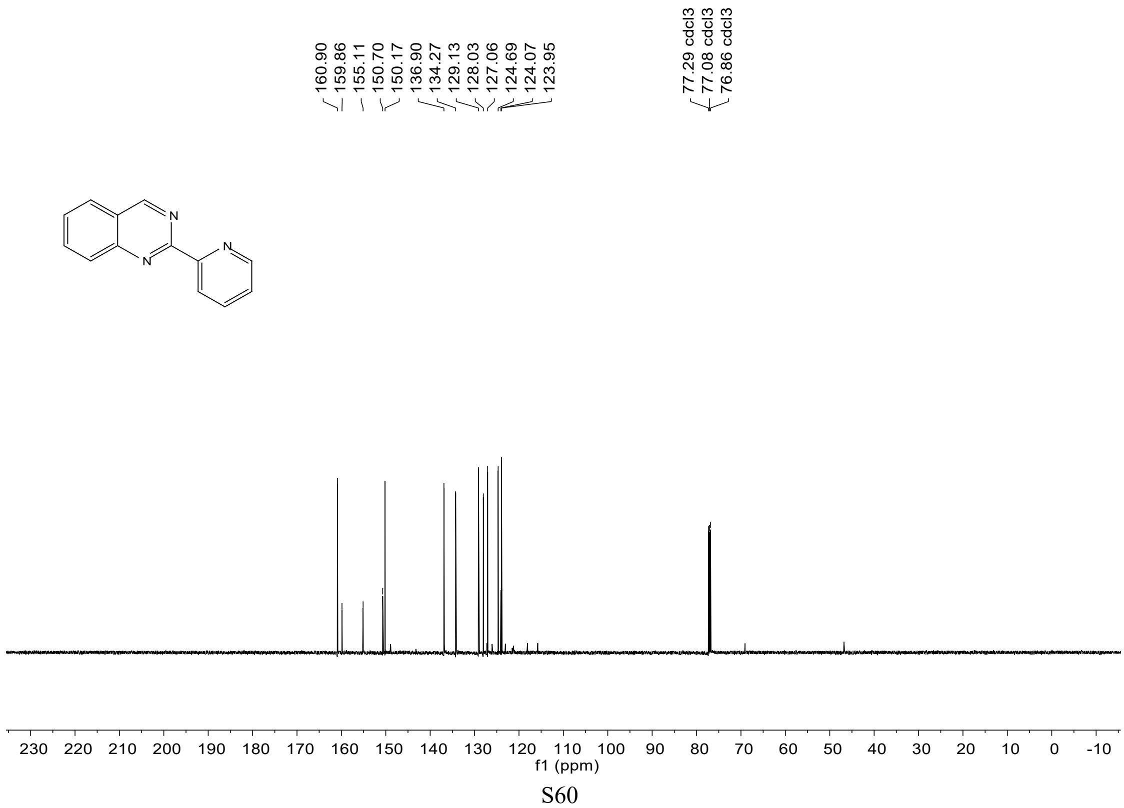
^{13}C NMR spectrum of 6b



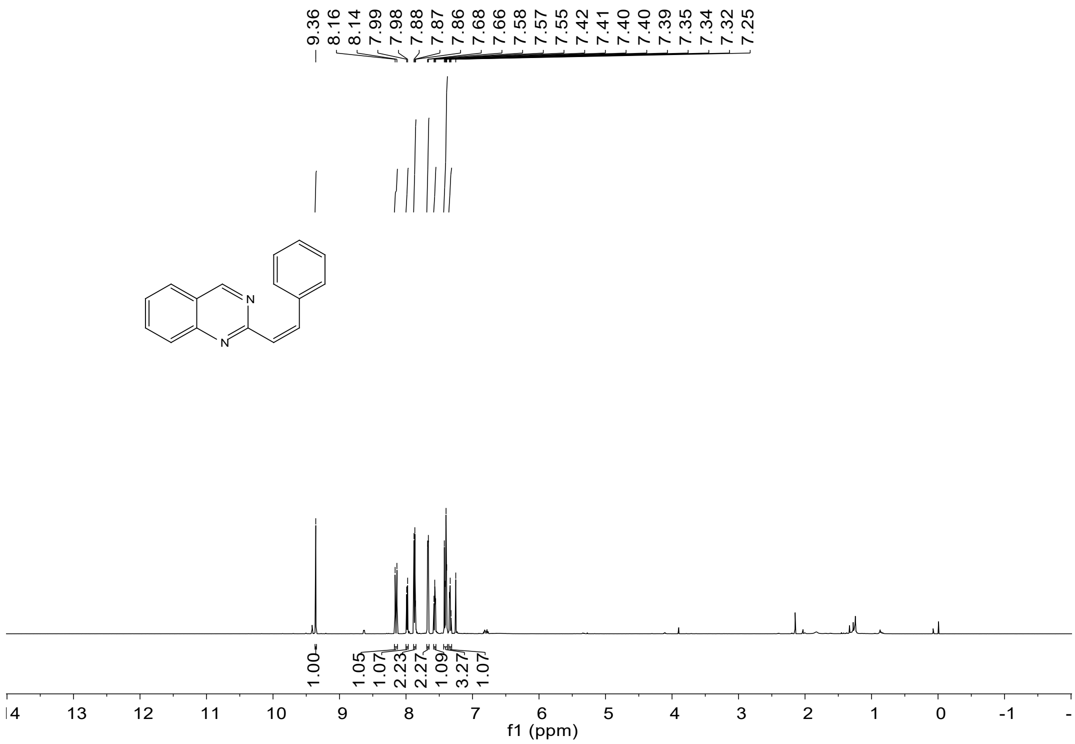
1H NMR spectrum of 6c



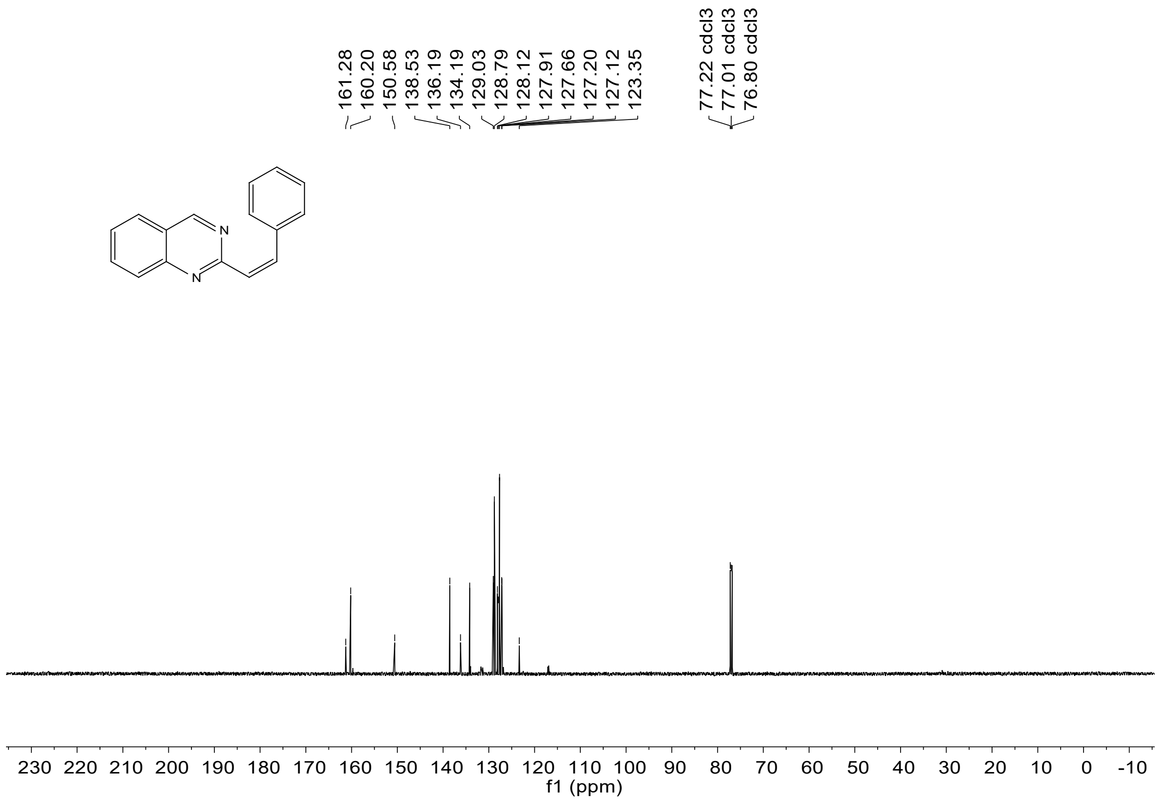
13C NMR spectrum of 6c



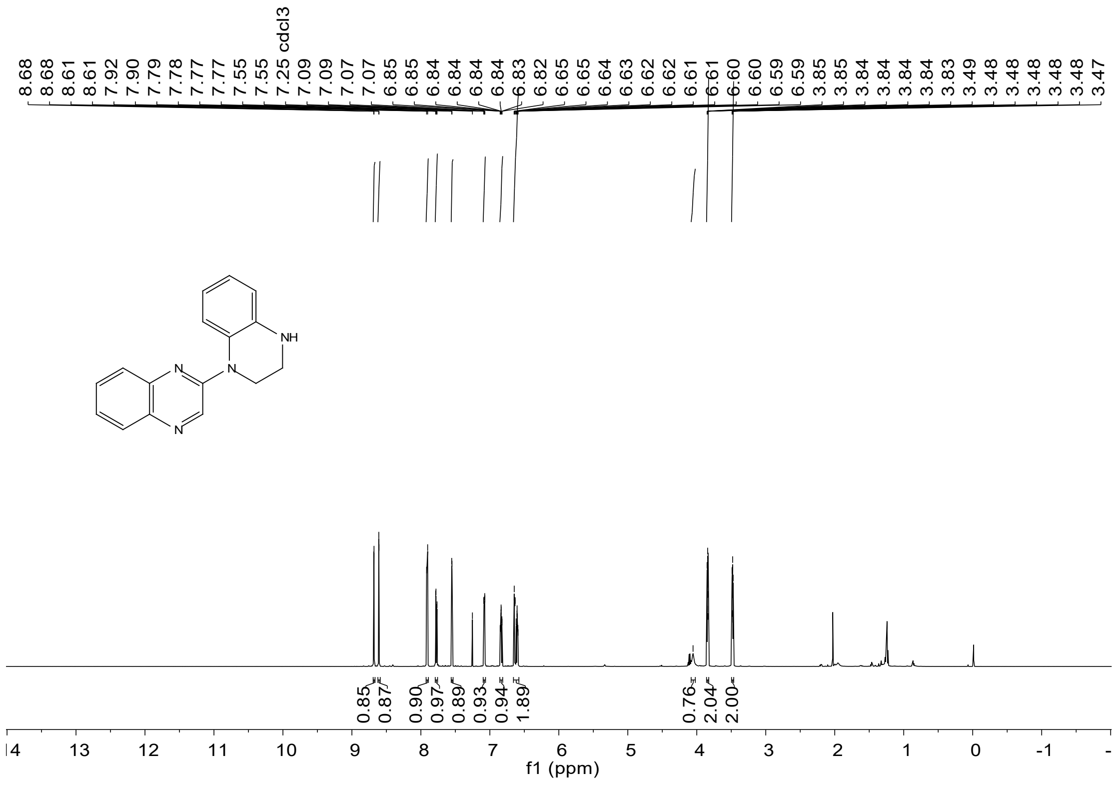
^{1}H NMR spectrum of 6d



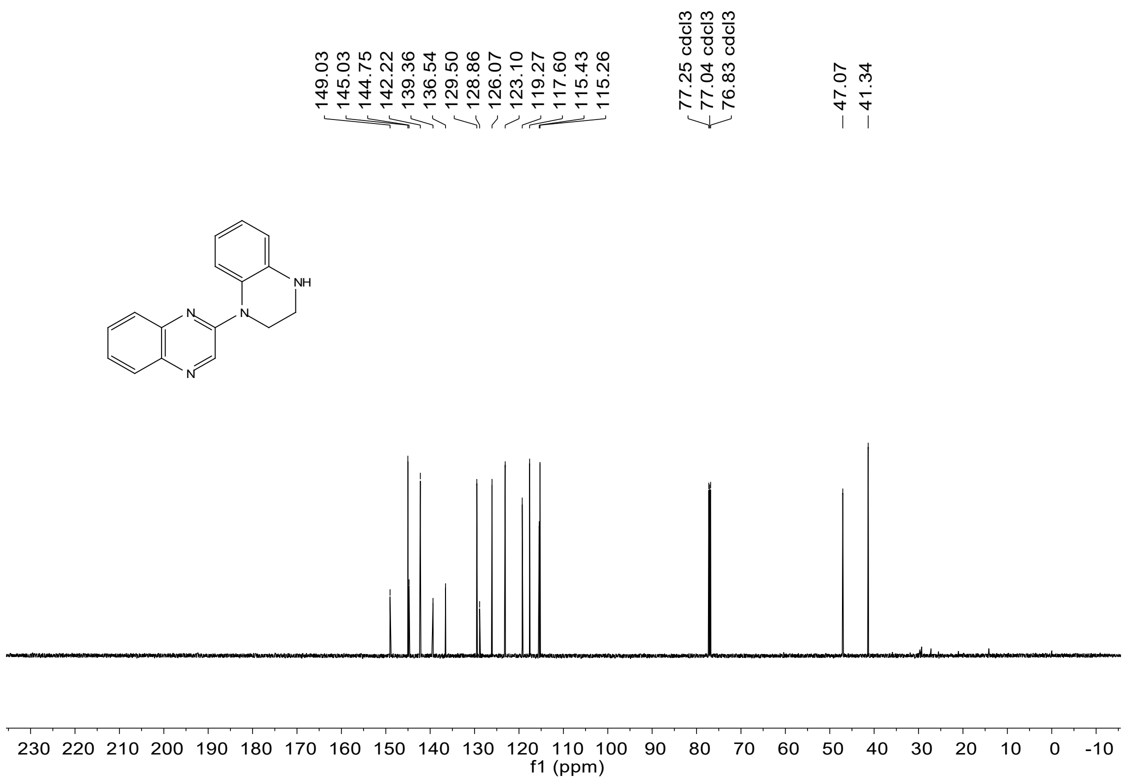
^{13}C NMR spectrum of 6d



^{1}H NMR spectrum of 8a

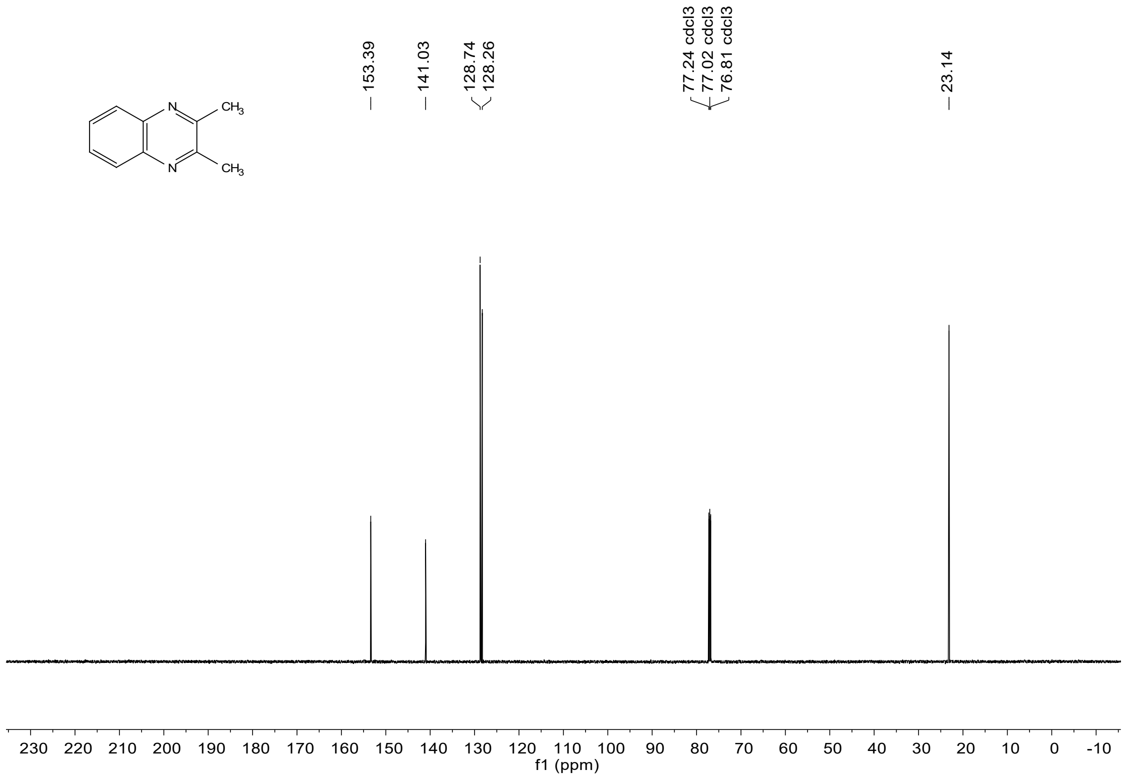


^{13}C NMR spectrum of 8a

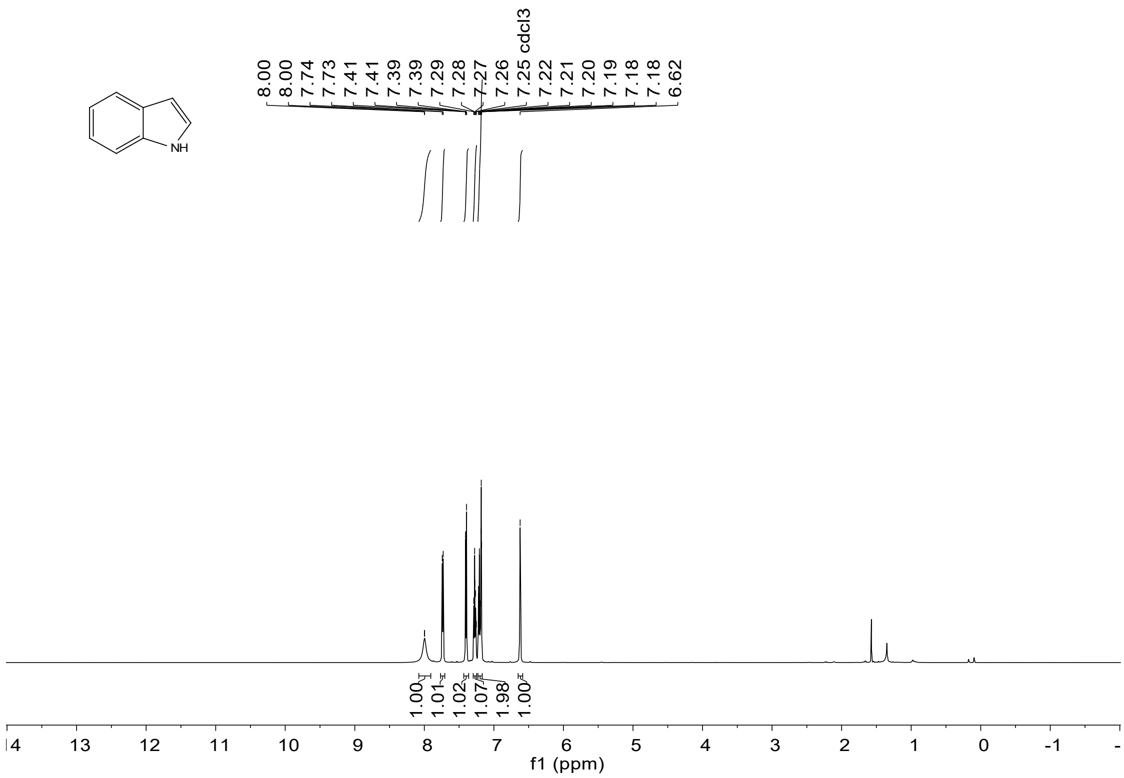


^{1}H NMR spectrum of 8b

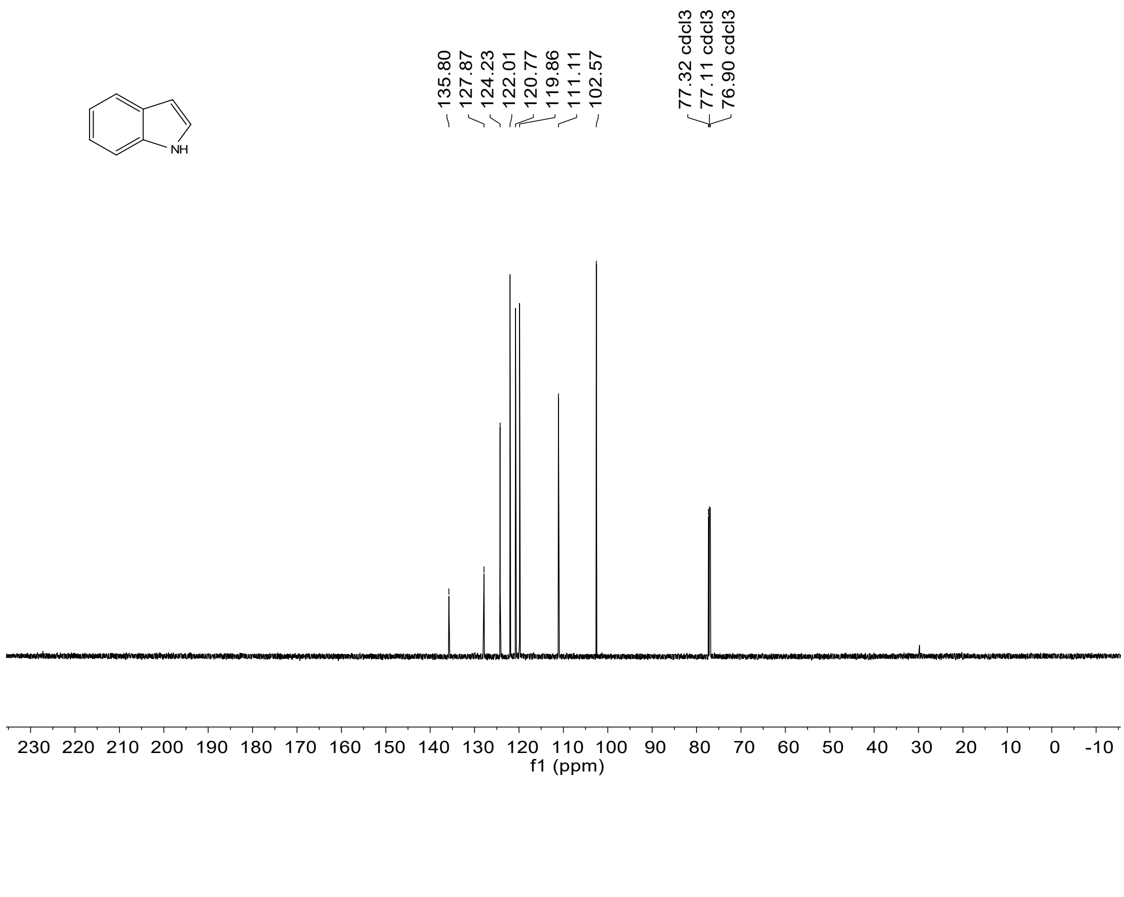
[13]C NMR spectrum of 8b



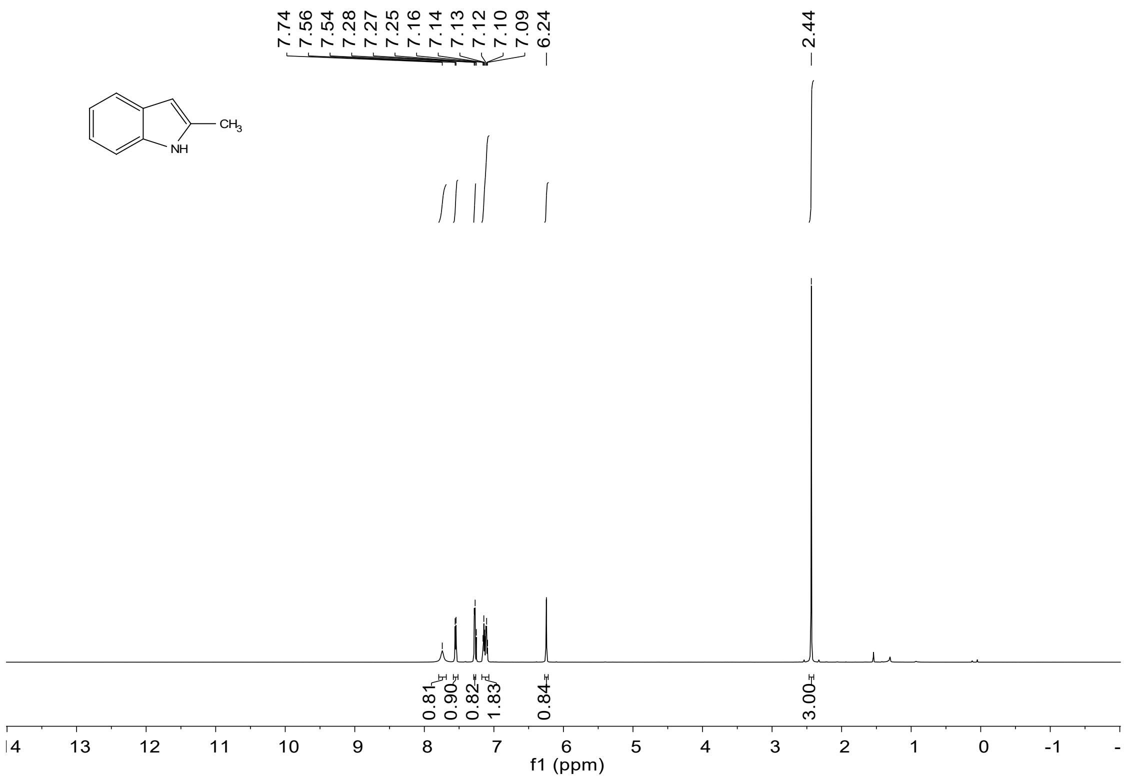
[1]H NMR spectrum of 10a



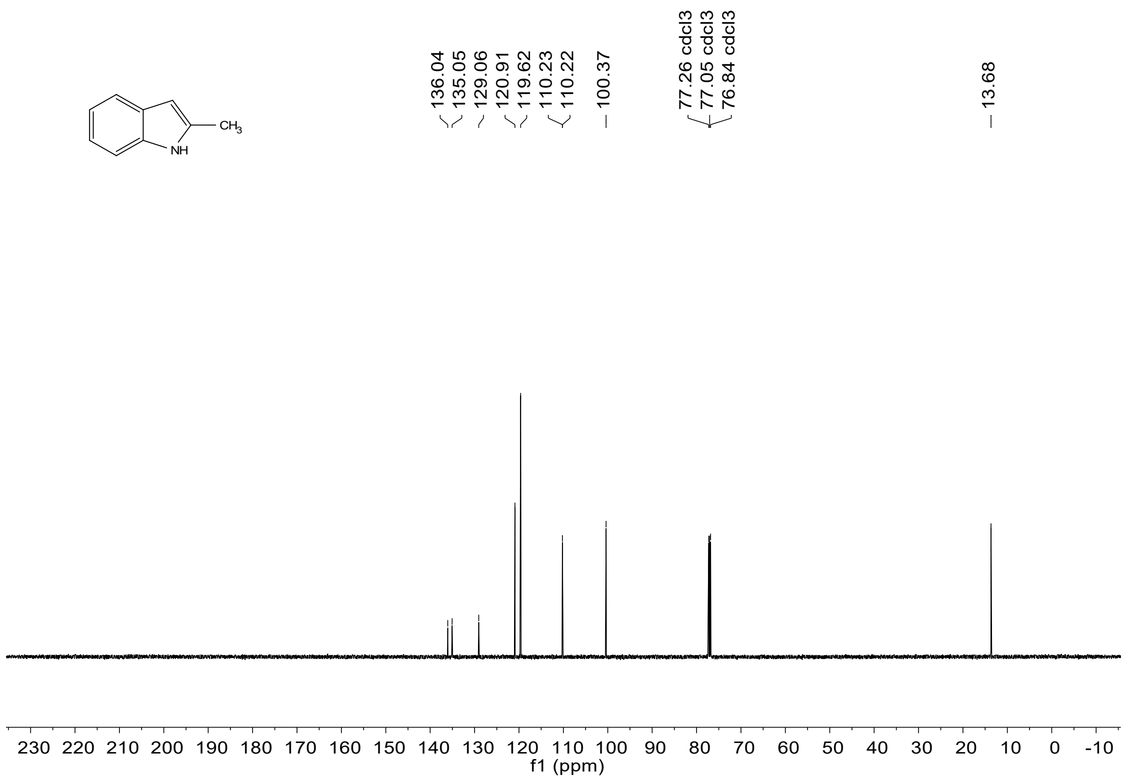
^{13}C NMR spectrum of 10a



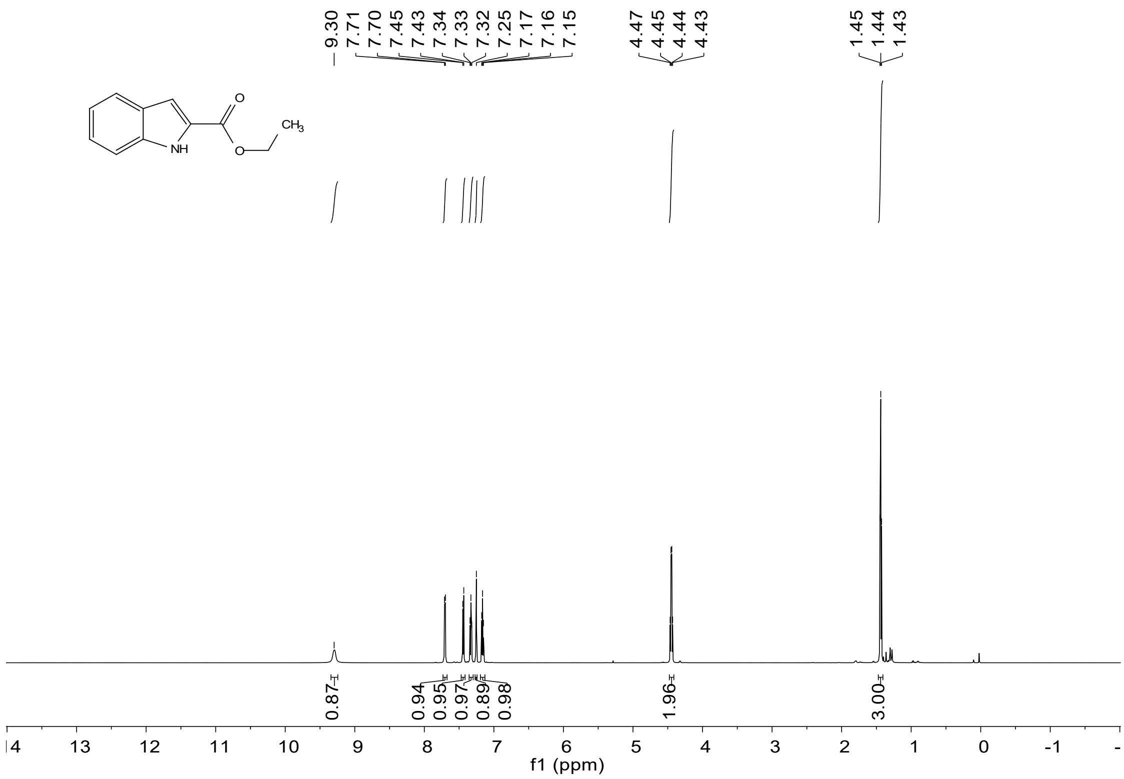
^{1}H NMR spectrum of 10b



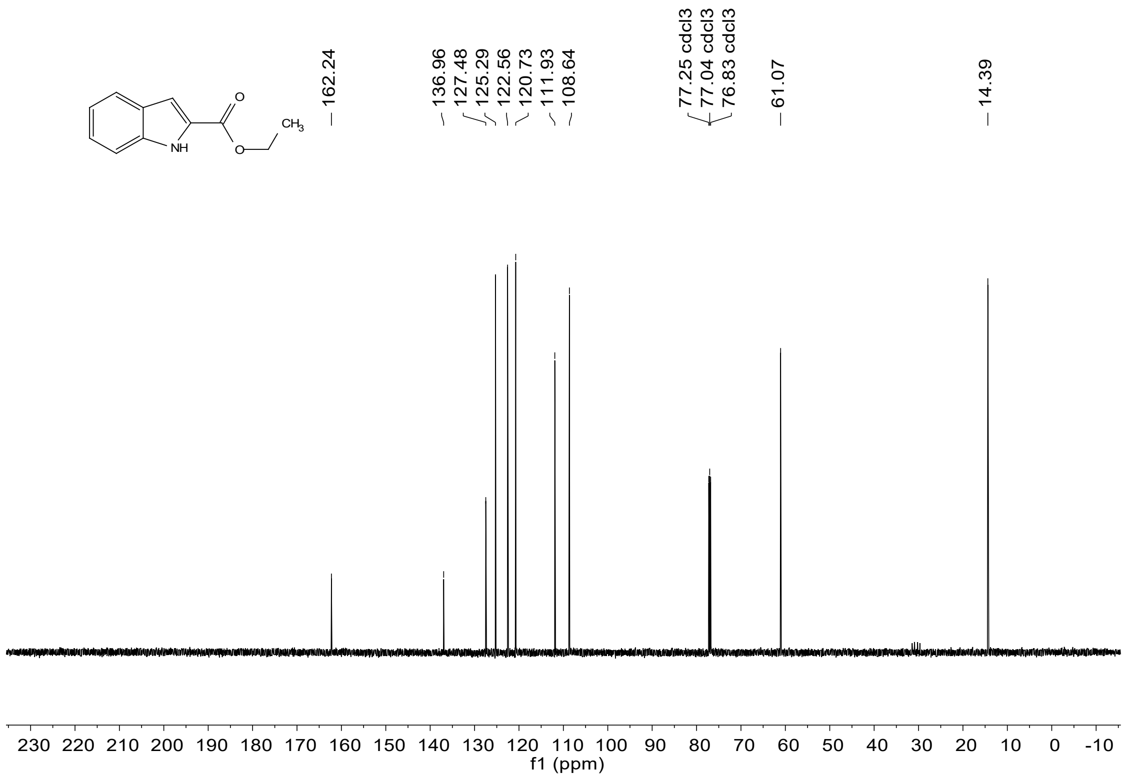
^{13}C NMR spectrum of 10b



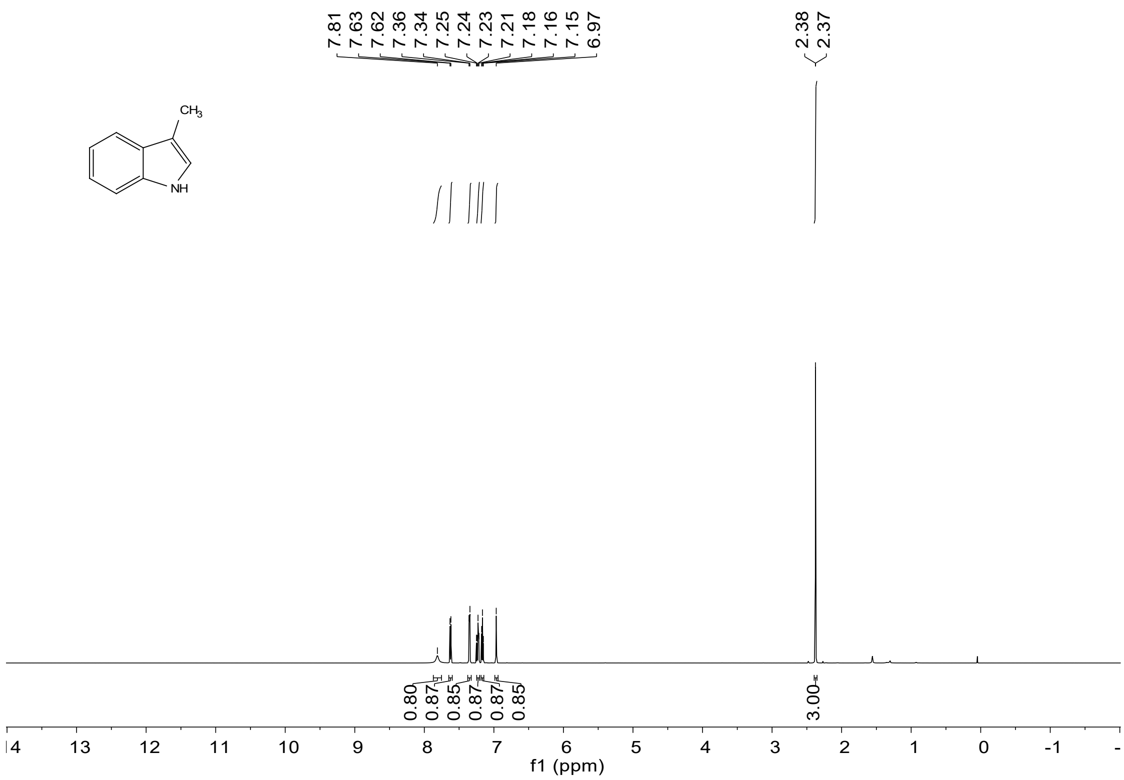
^{1}H NMR spectrum of 10d



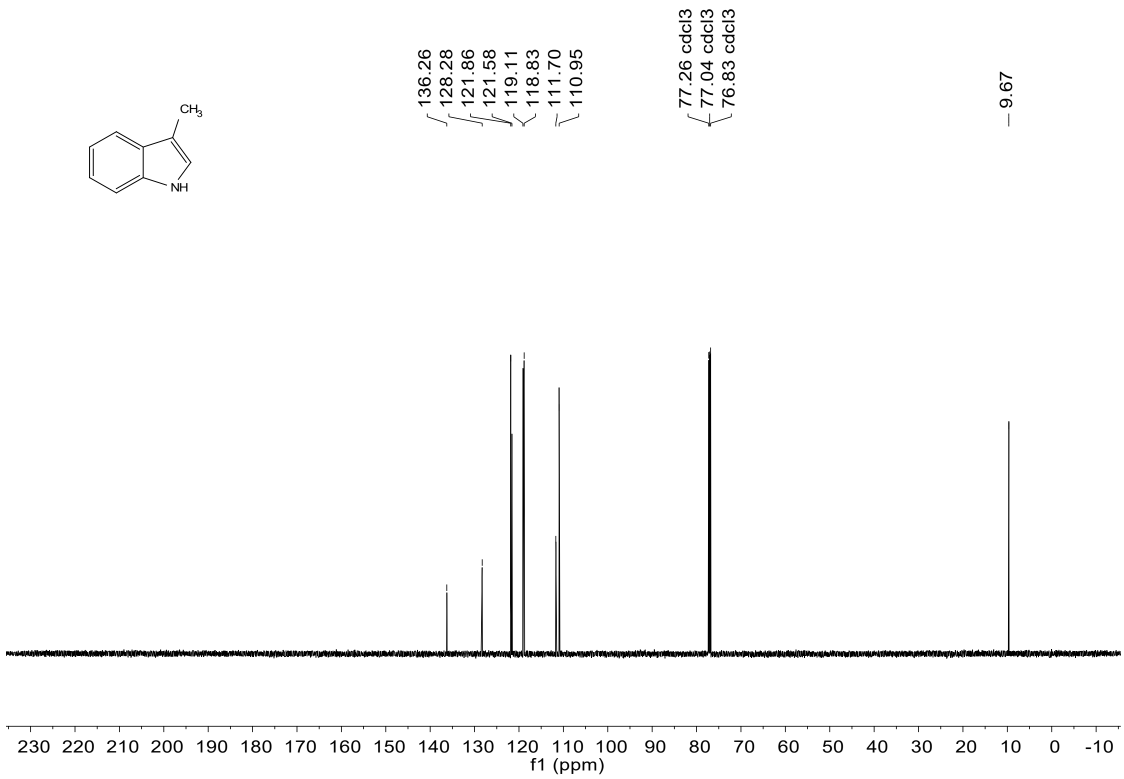
^{13}C NMR spectrum of 10d



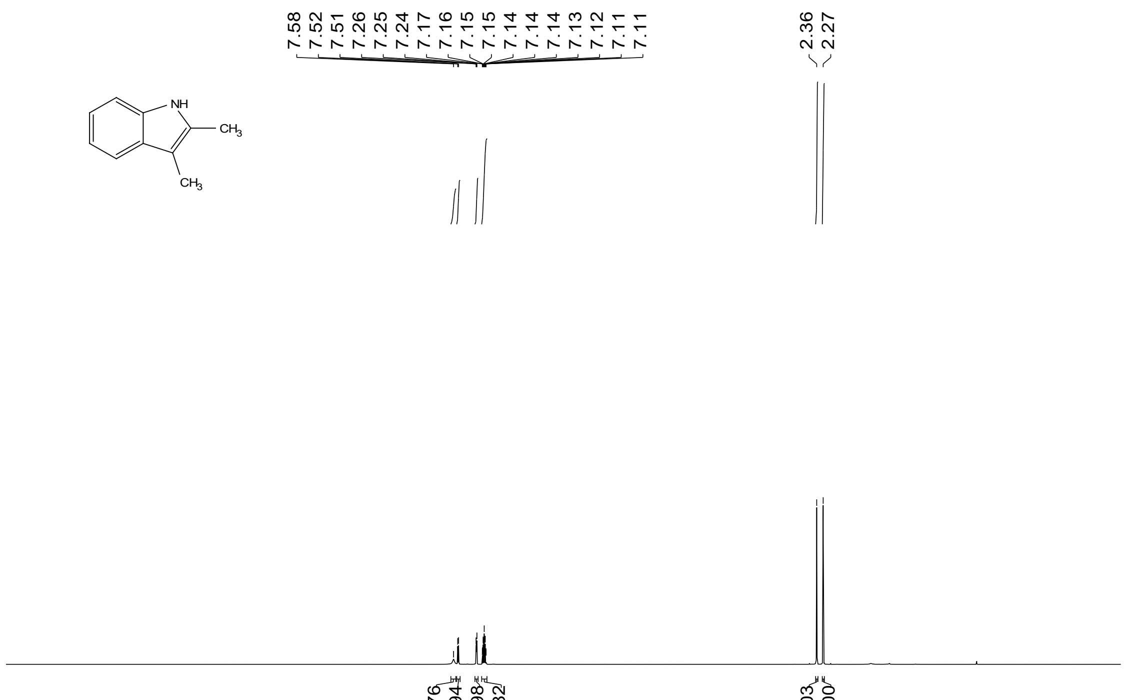
[1]H NMR spectrum of 10e



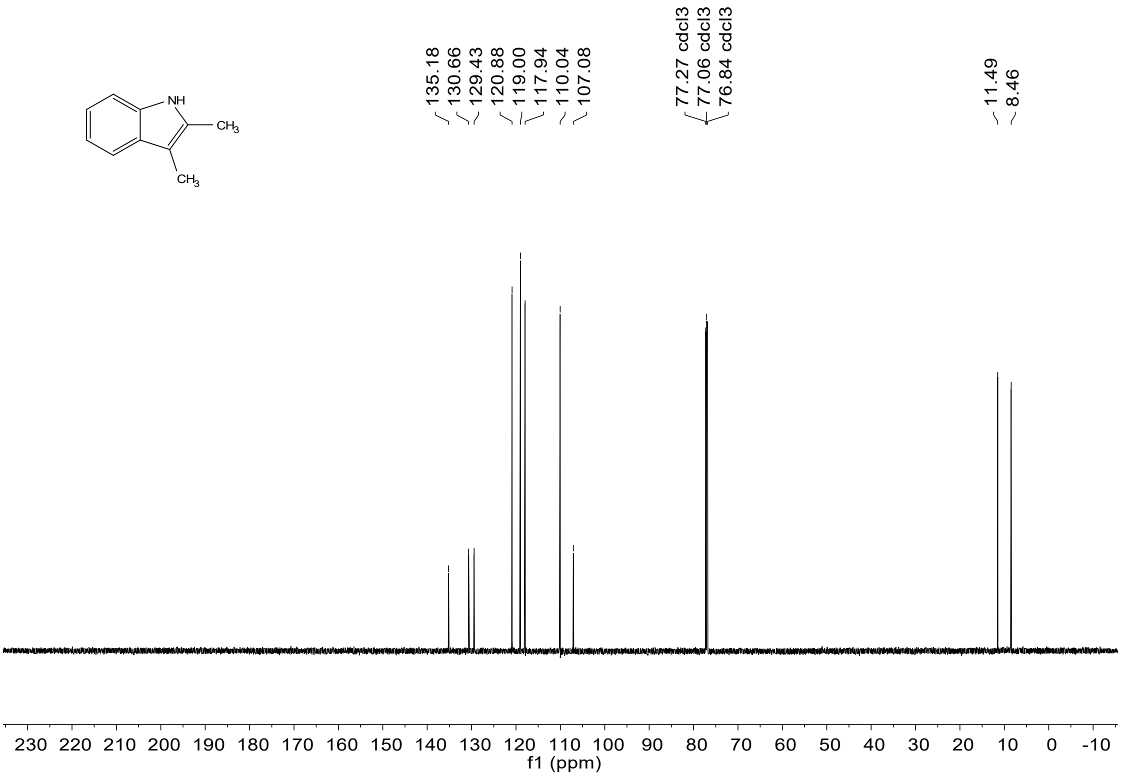
^{13}C NMR spectrum of 10e



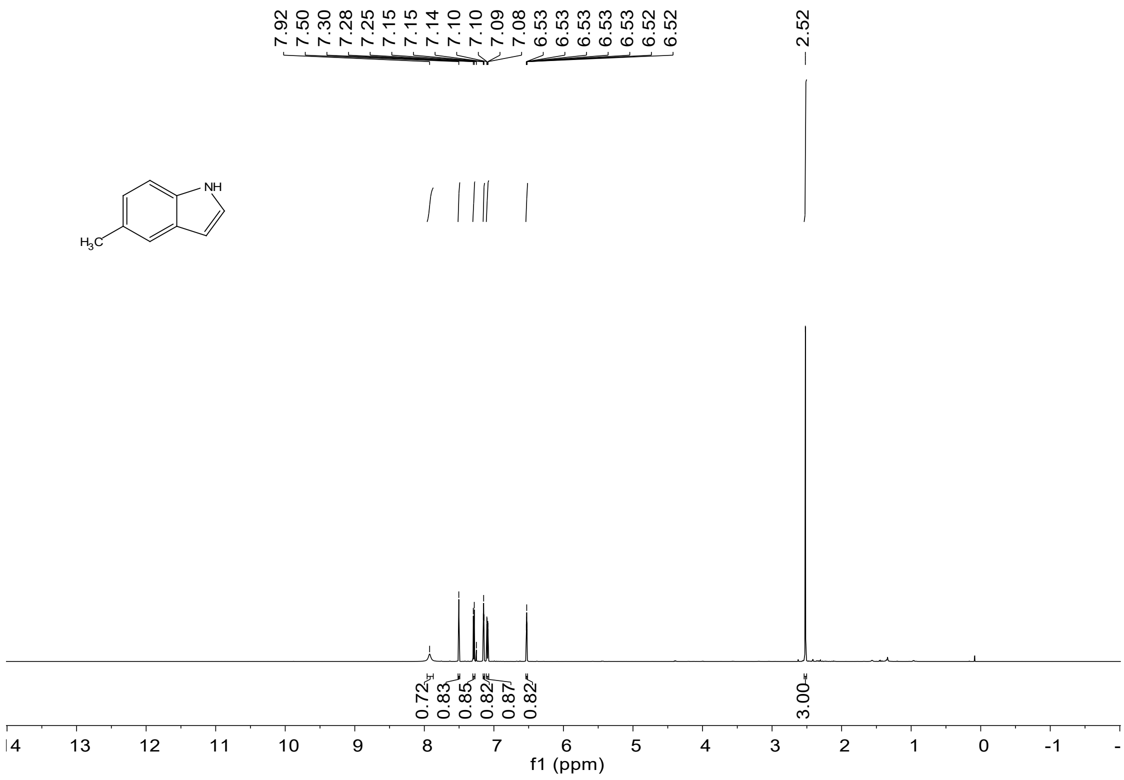
^{1}H NMR spectrum of 10f



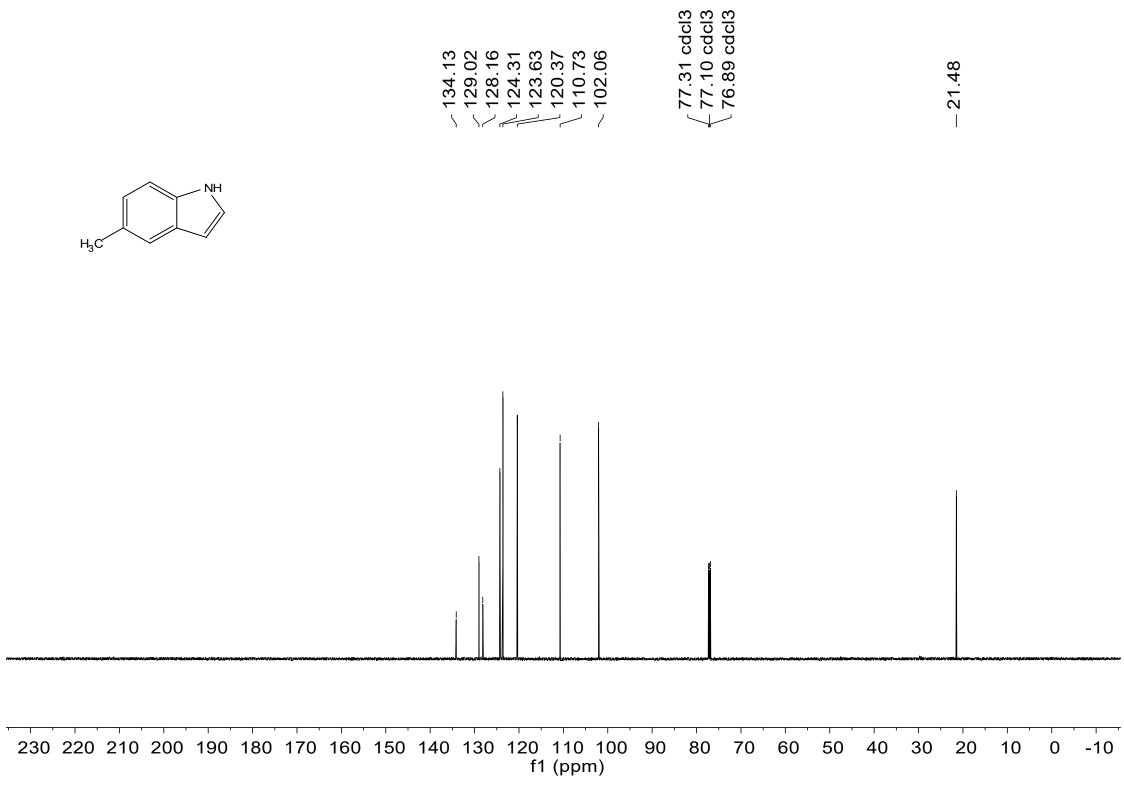
^{13}C NMR spectrum of 10f



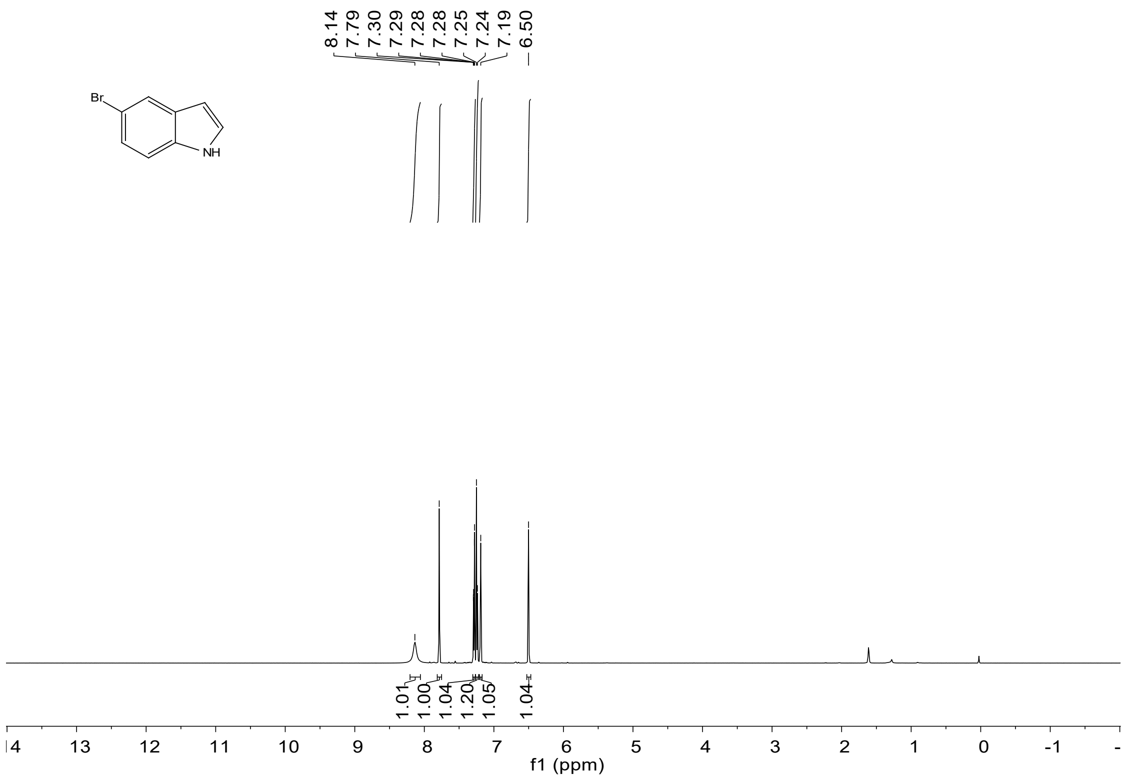
^{1}H NMR spectrum of 10g



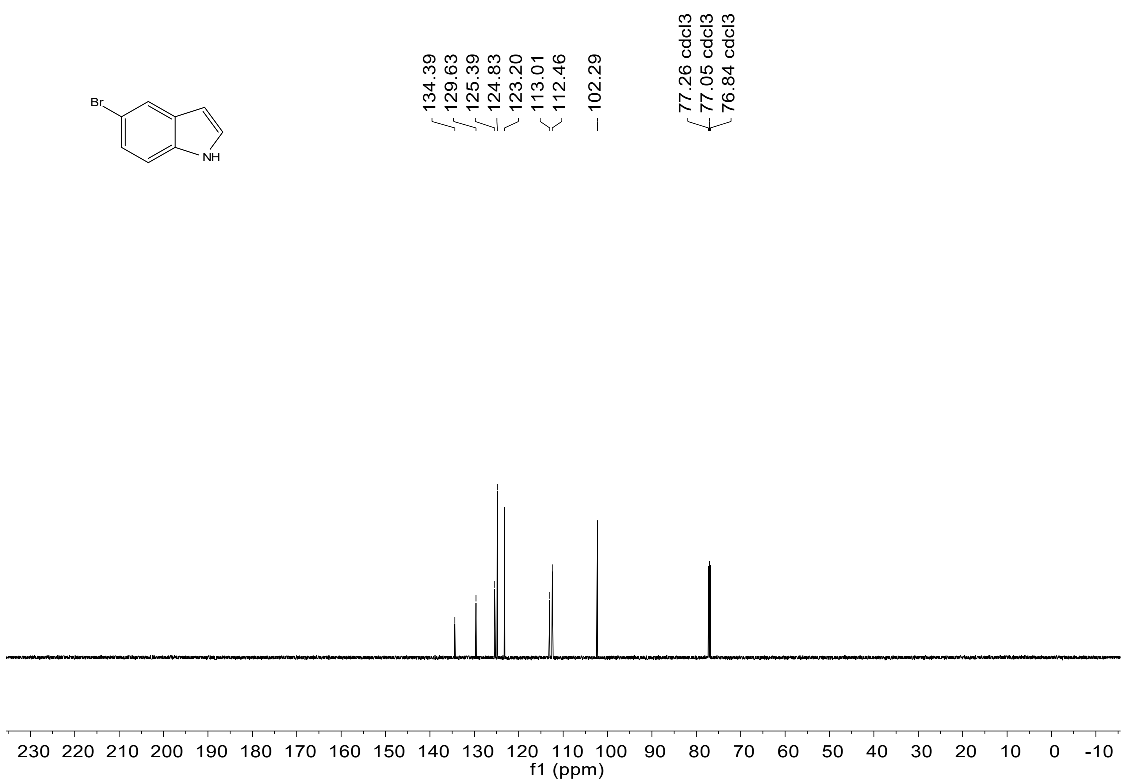
^{13}C NMR spectrum of 10g



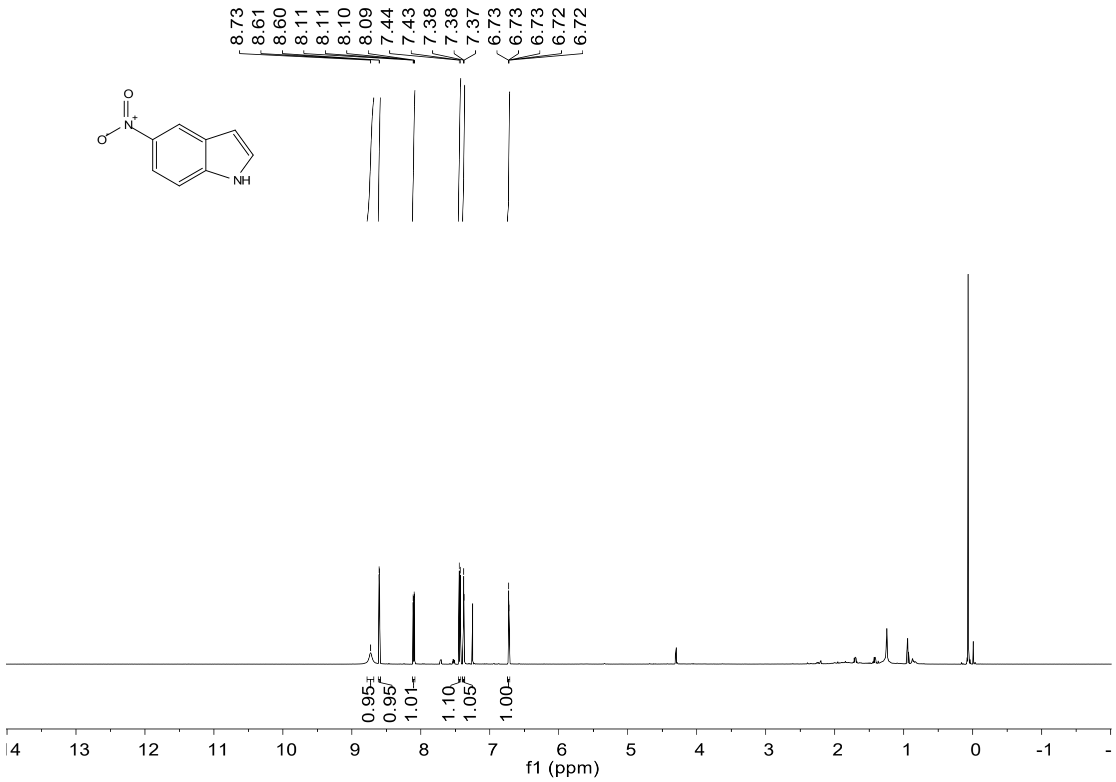
^{1}H NMR spectrum of 10h



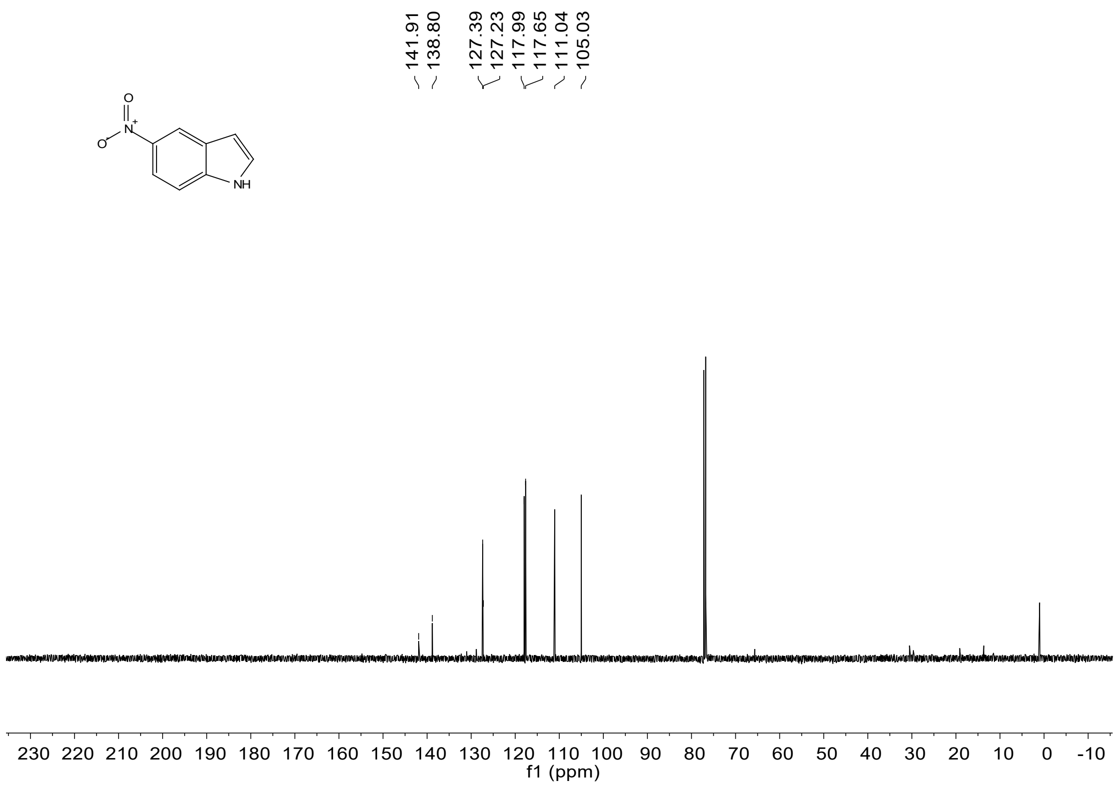
^{13}C NMR spectrum of 10h



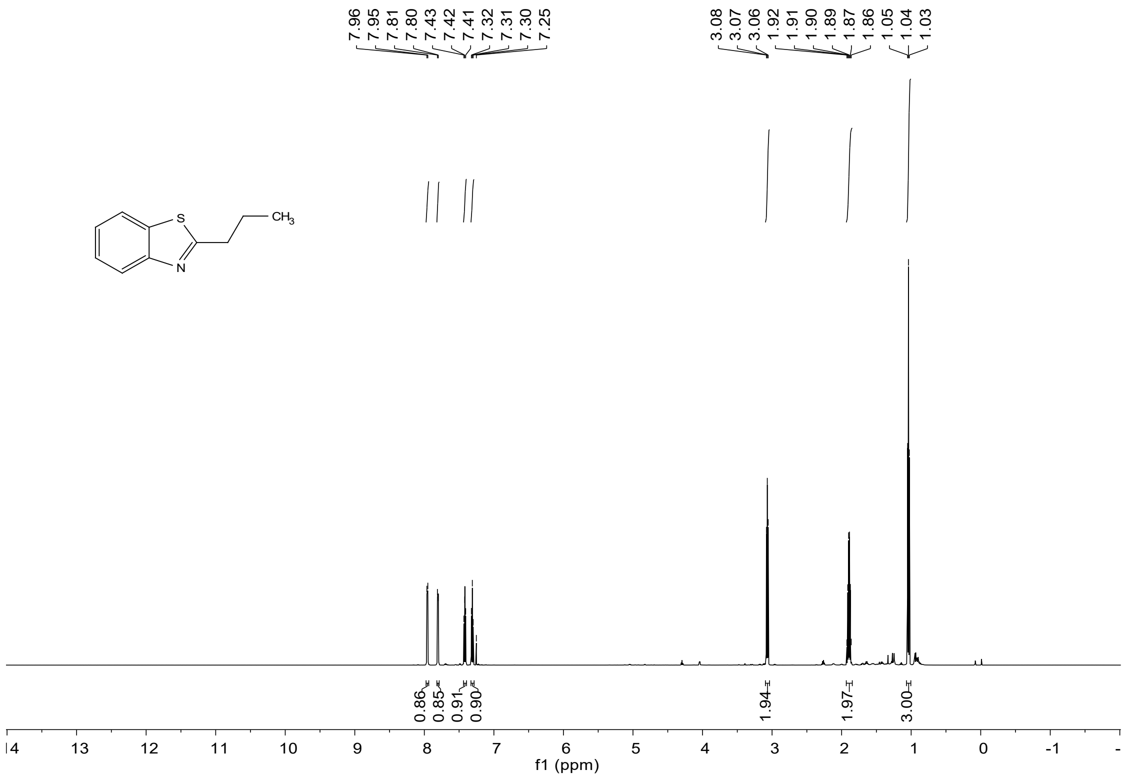
¹H NMR spectrum of 10i



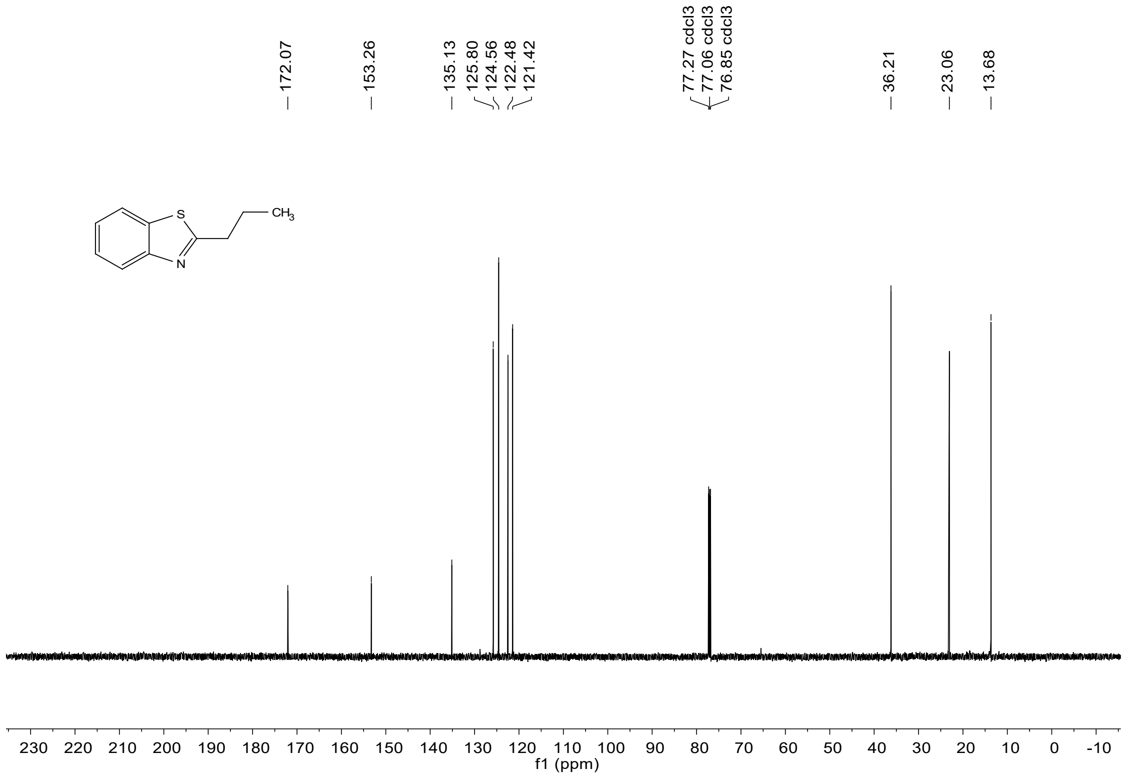
¹³C NMR spectrum of 10i



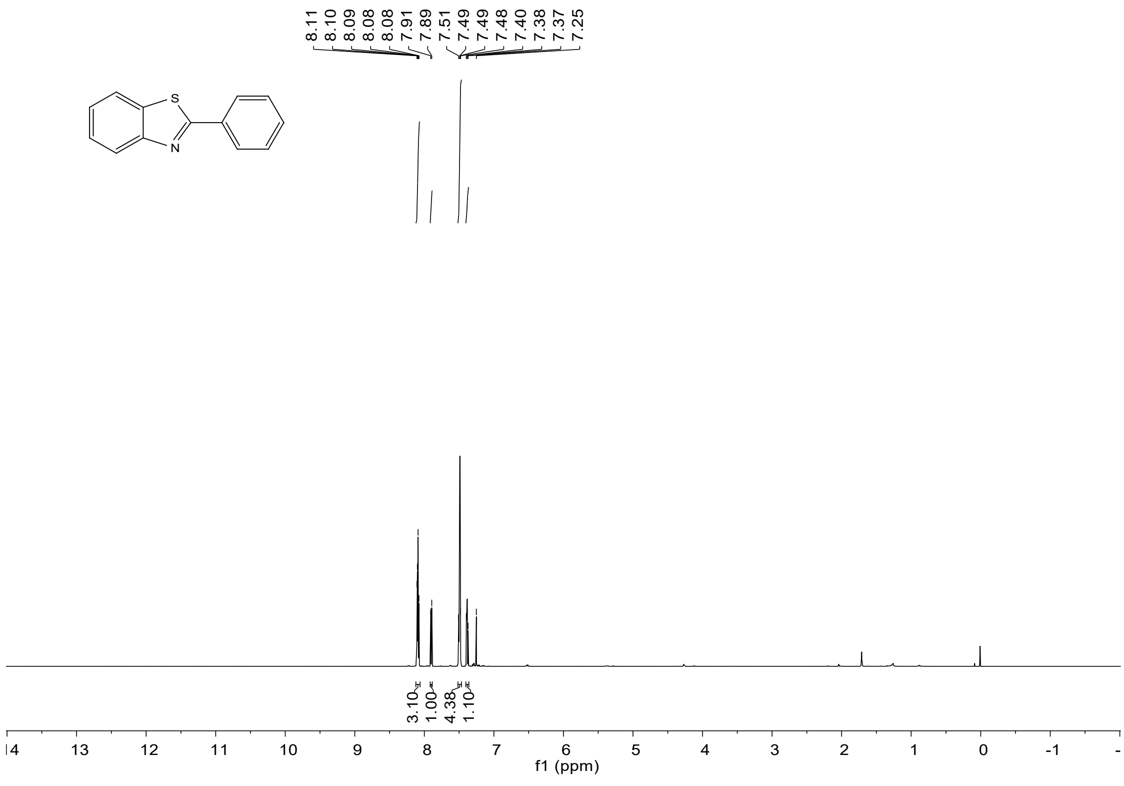
^{1}H NMR spectrum of 12a



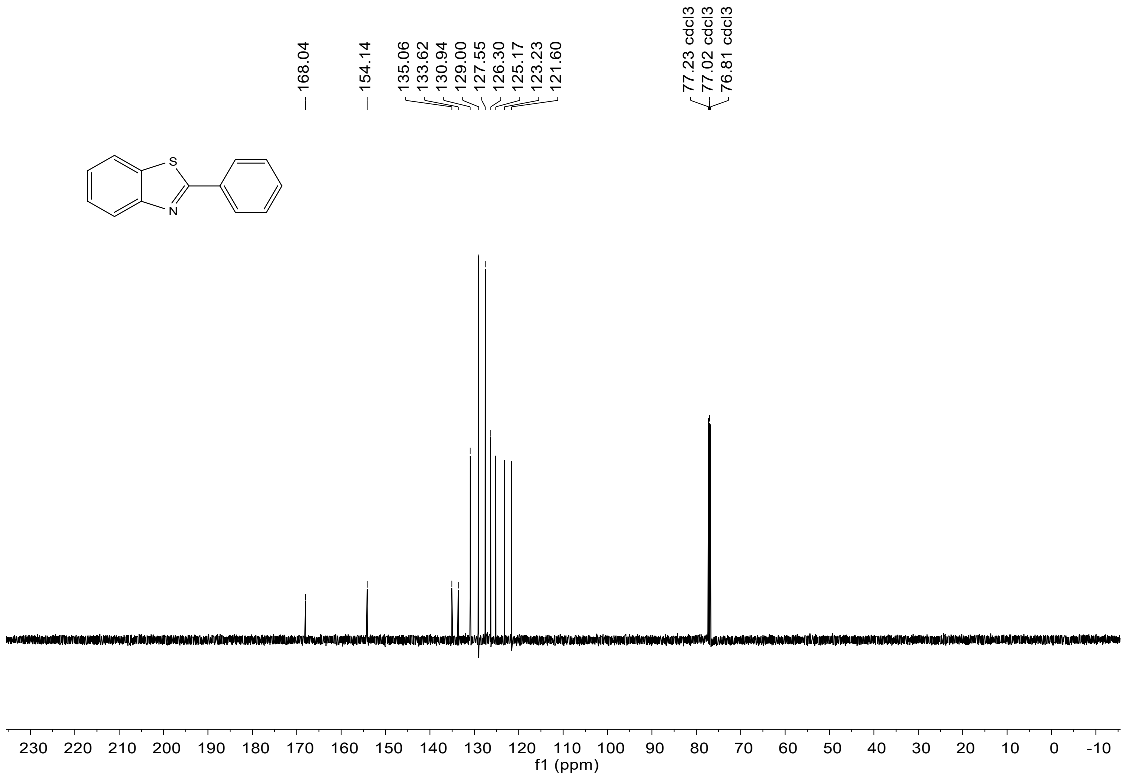
^{13}C NMR spectrum of 12a



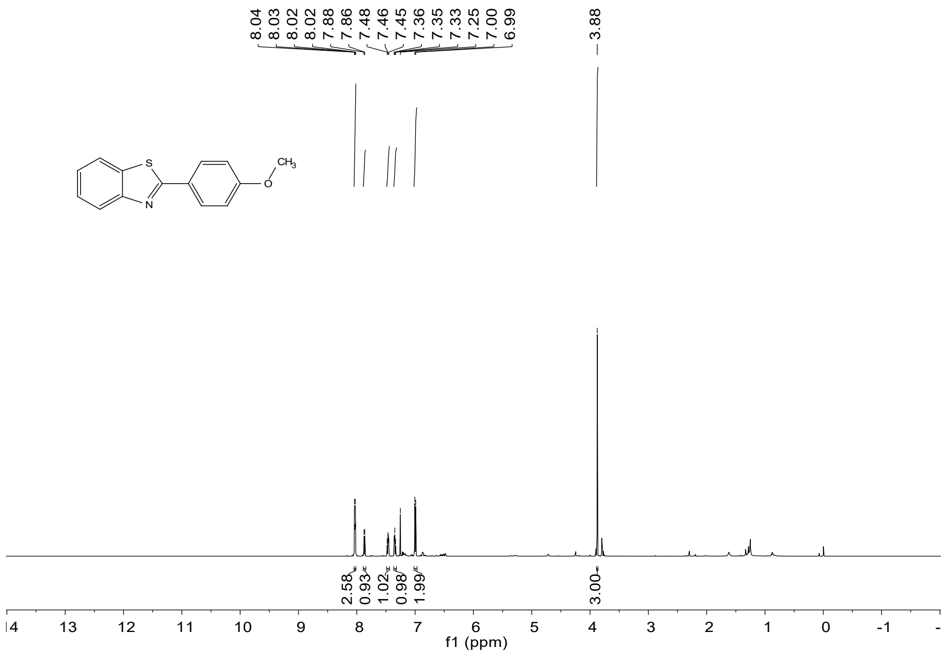
[1]H NMR spectrum of 12b



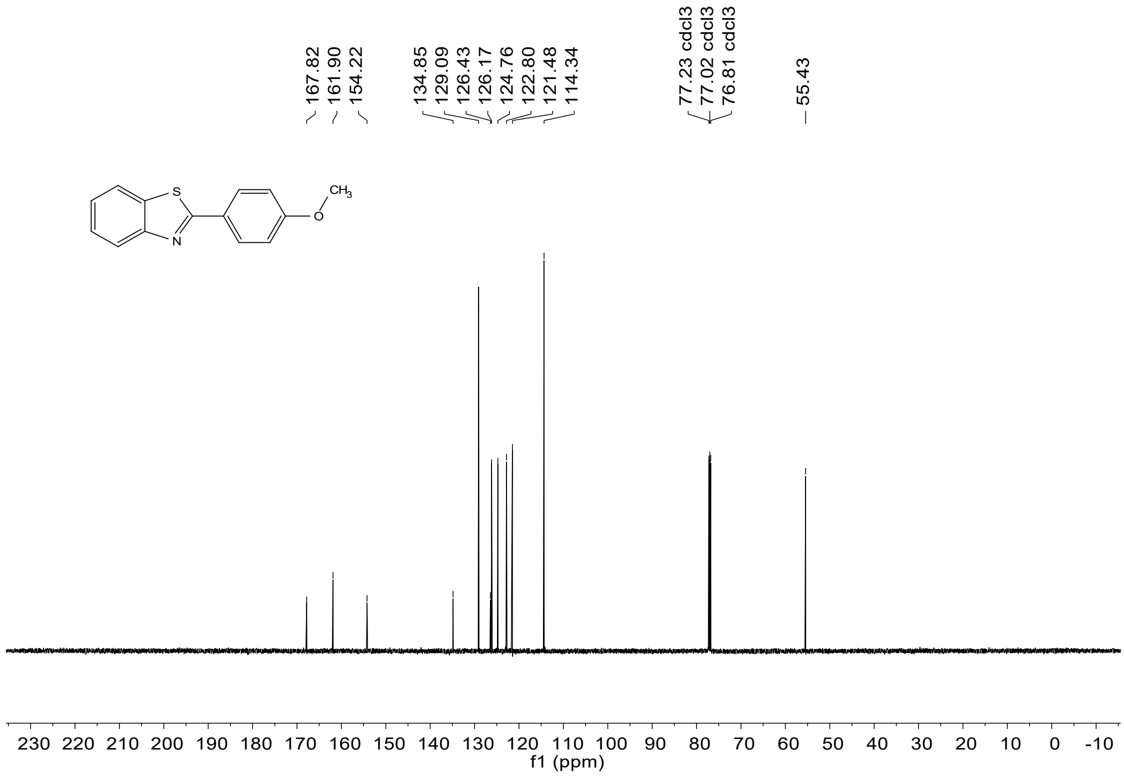
^{13}C NMR spectrum of 12b



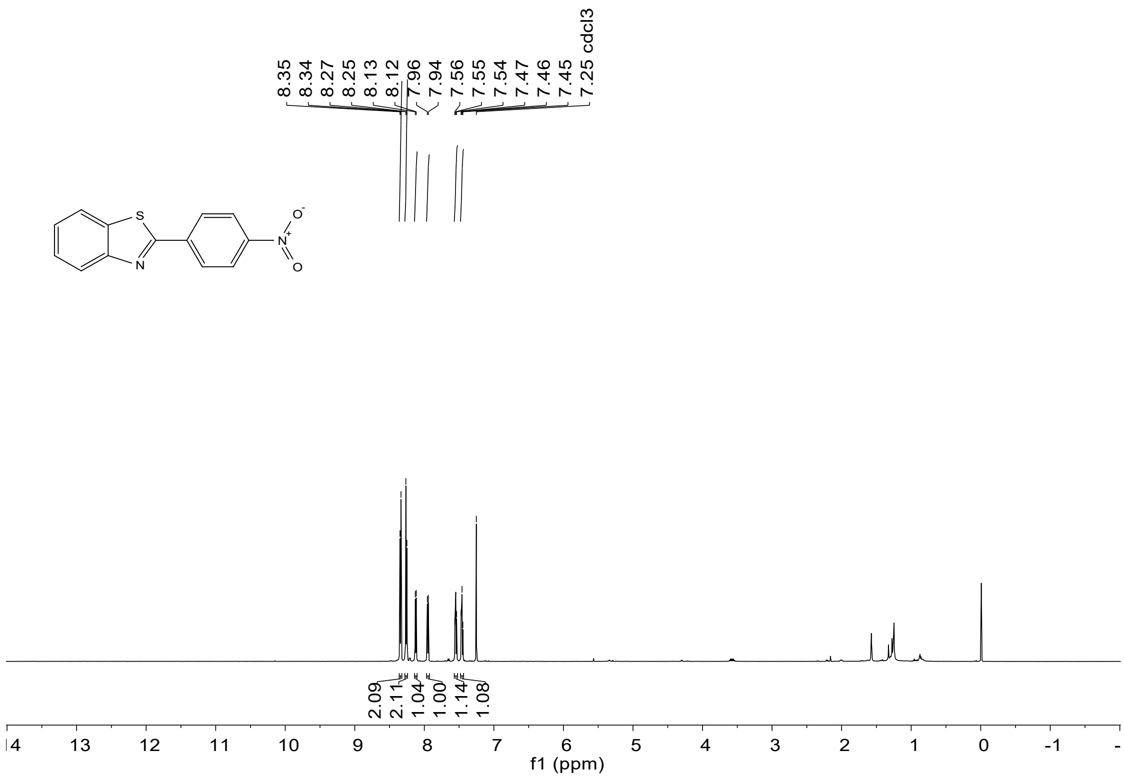
^{1}H NMR spectrum of 12c



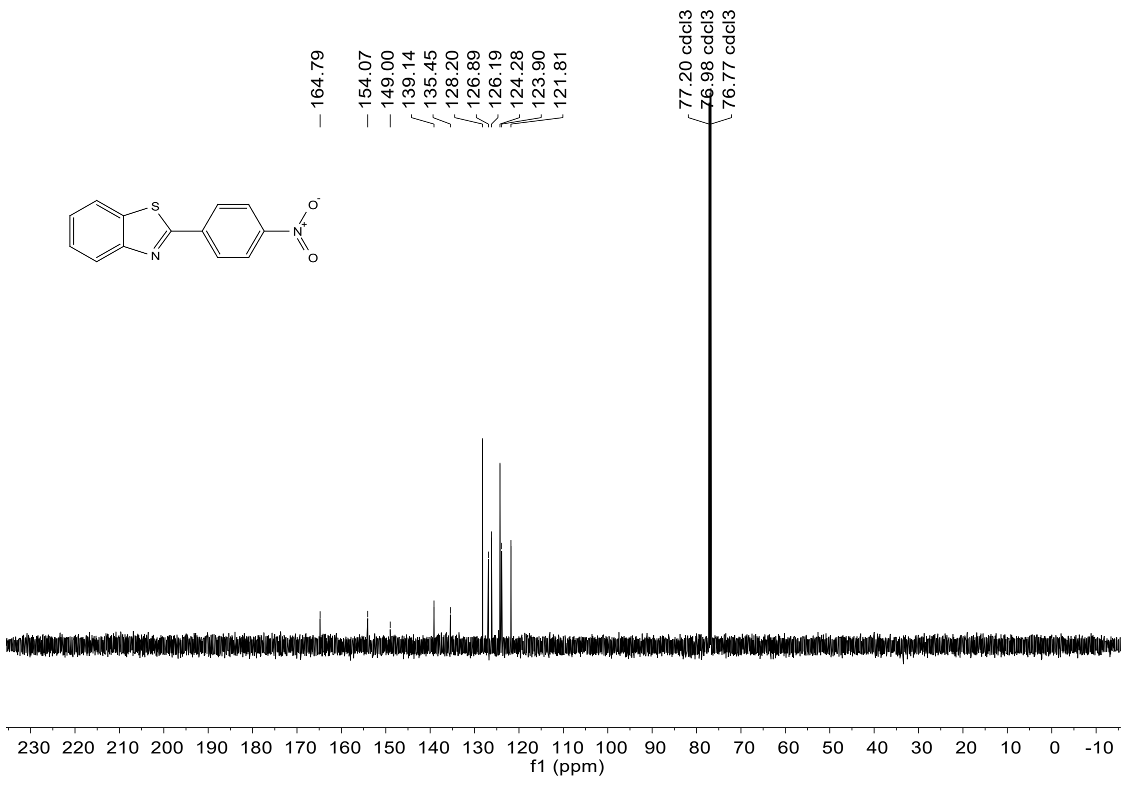
^{13}C NMR spectrum of 12c



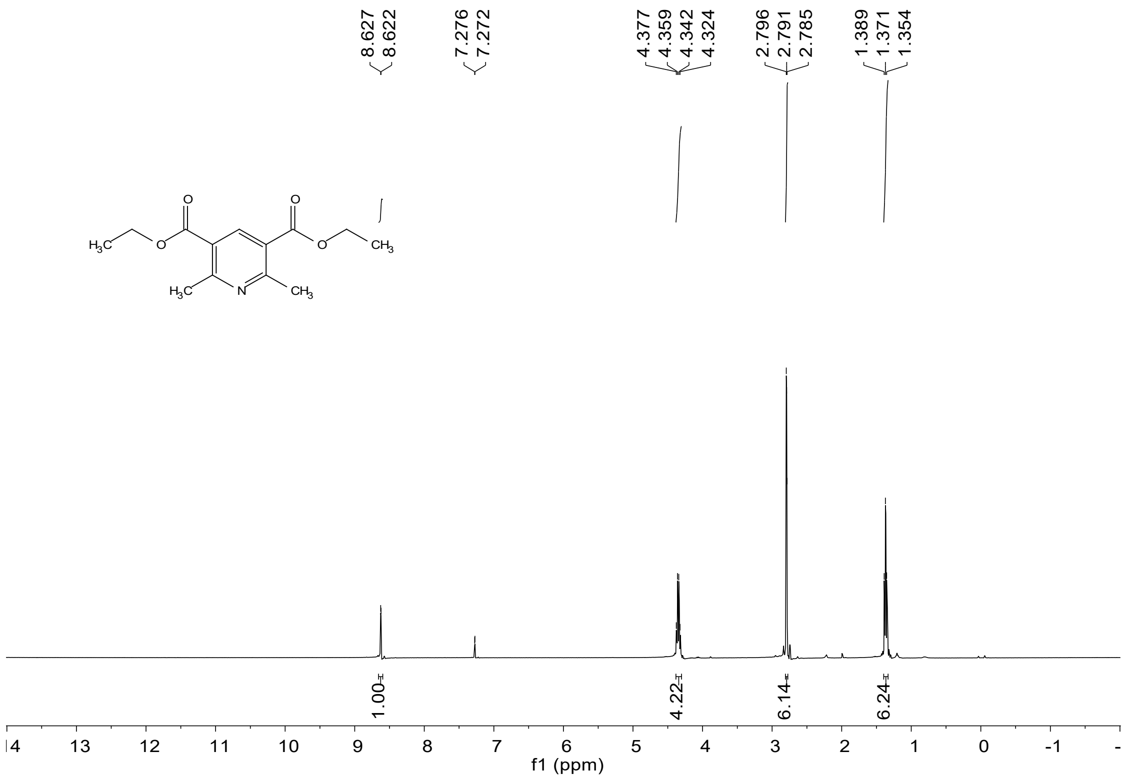
[1]H NMR spectrum of 12d



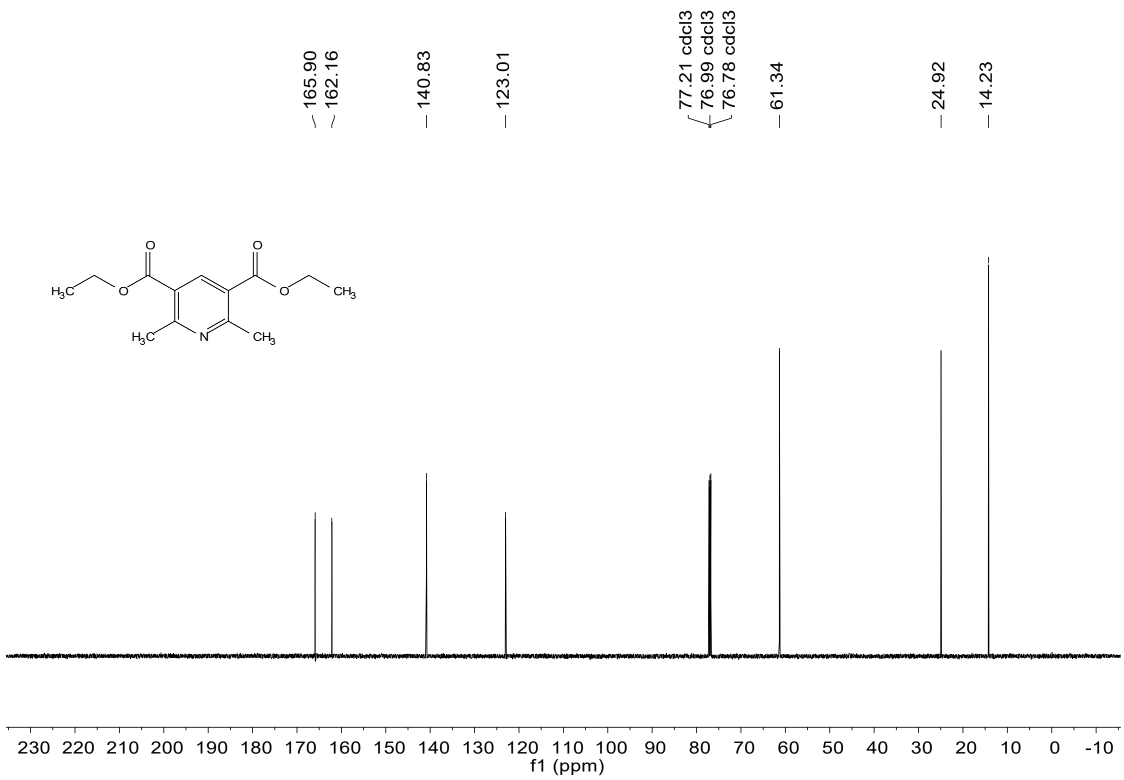
^{13}C NMR spectrum of 12d



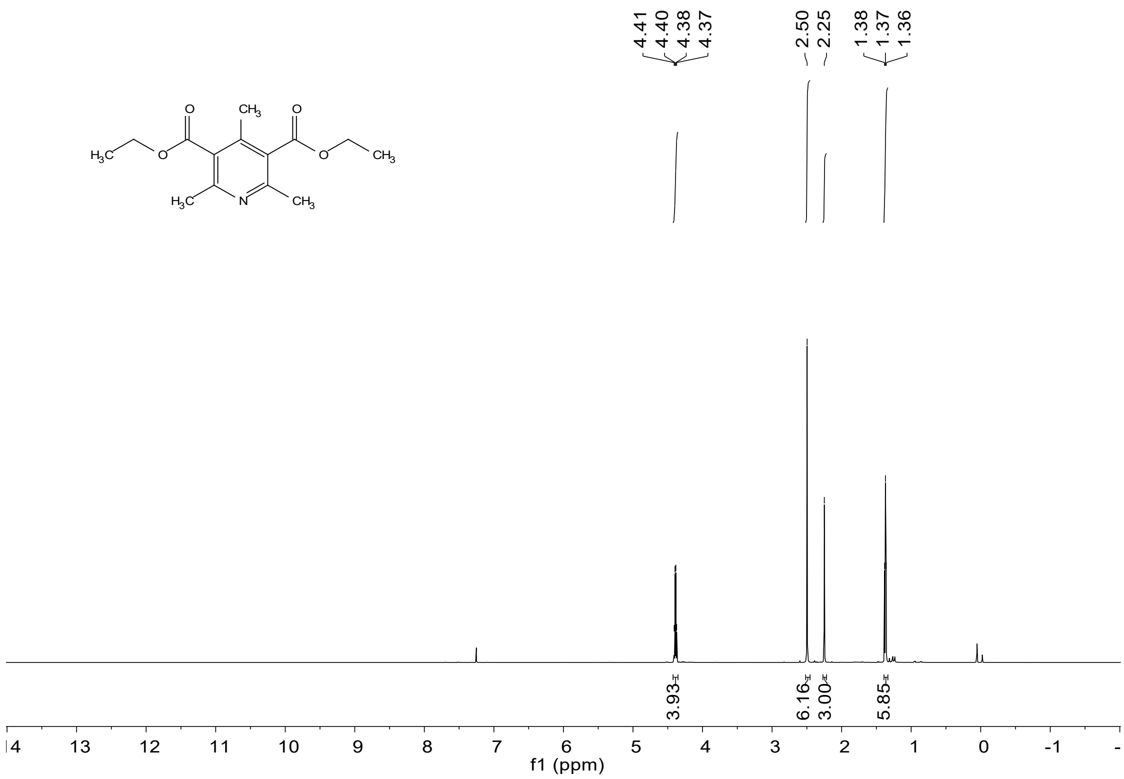
[1]H NMR spectrum of 14a



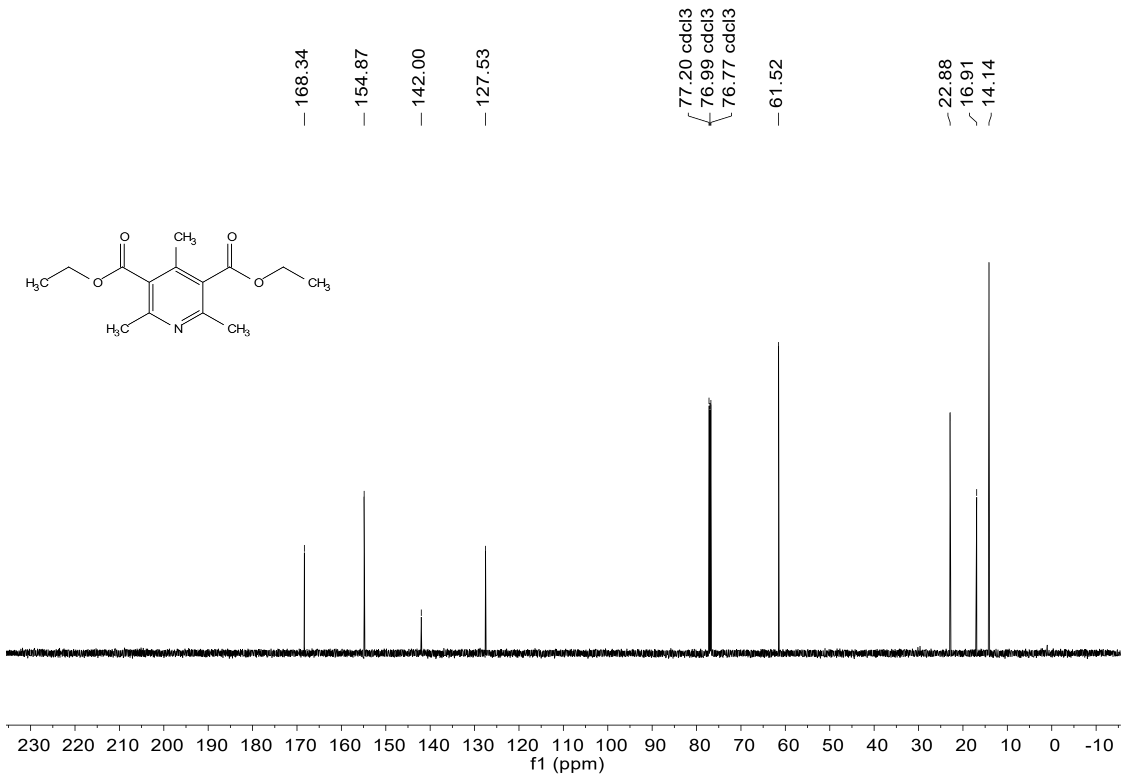
[13]C NMR spectrum of 14a



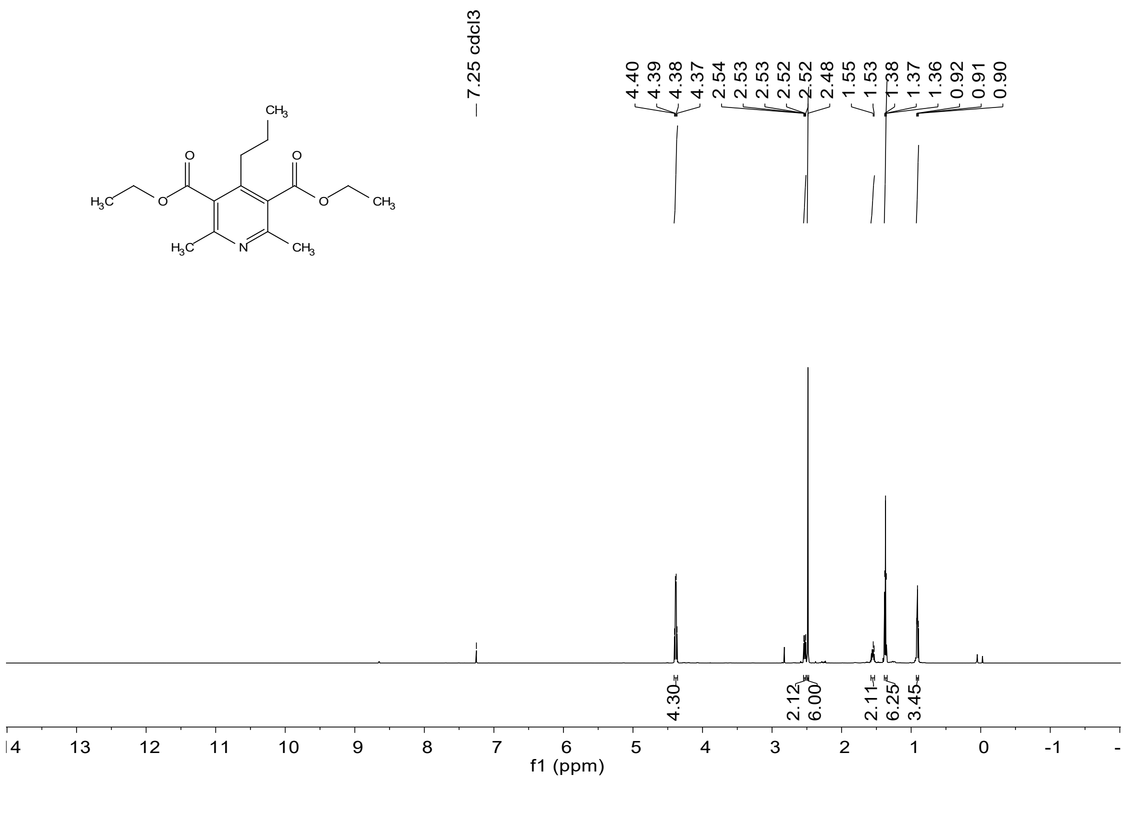
^{1}H NMR spectrum of 14b



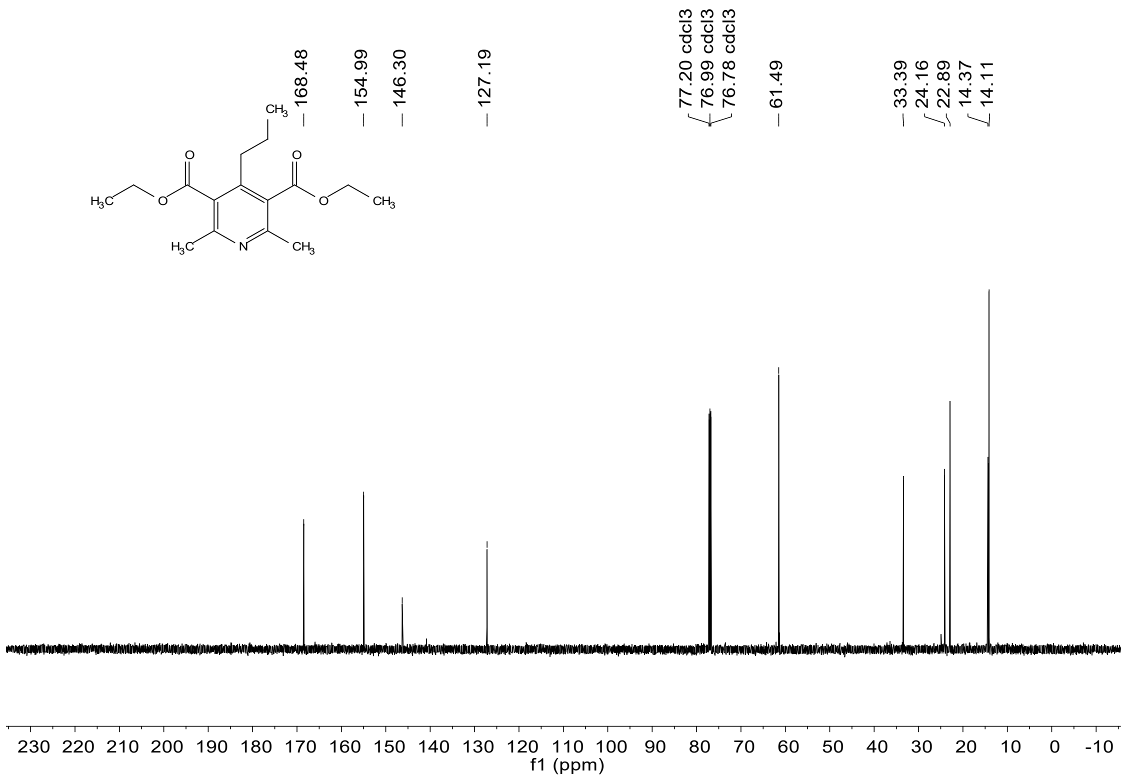
^{13}C NMR spectrum of 14b



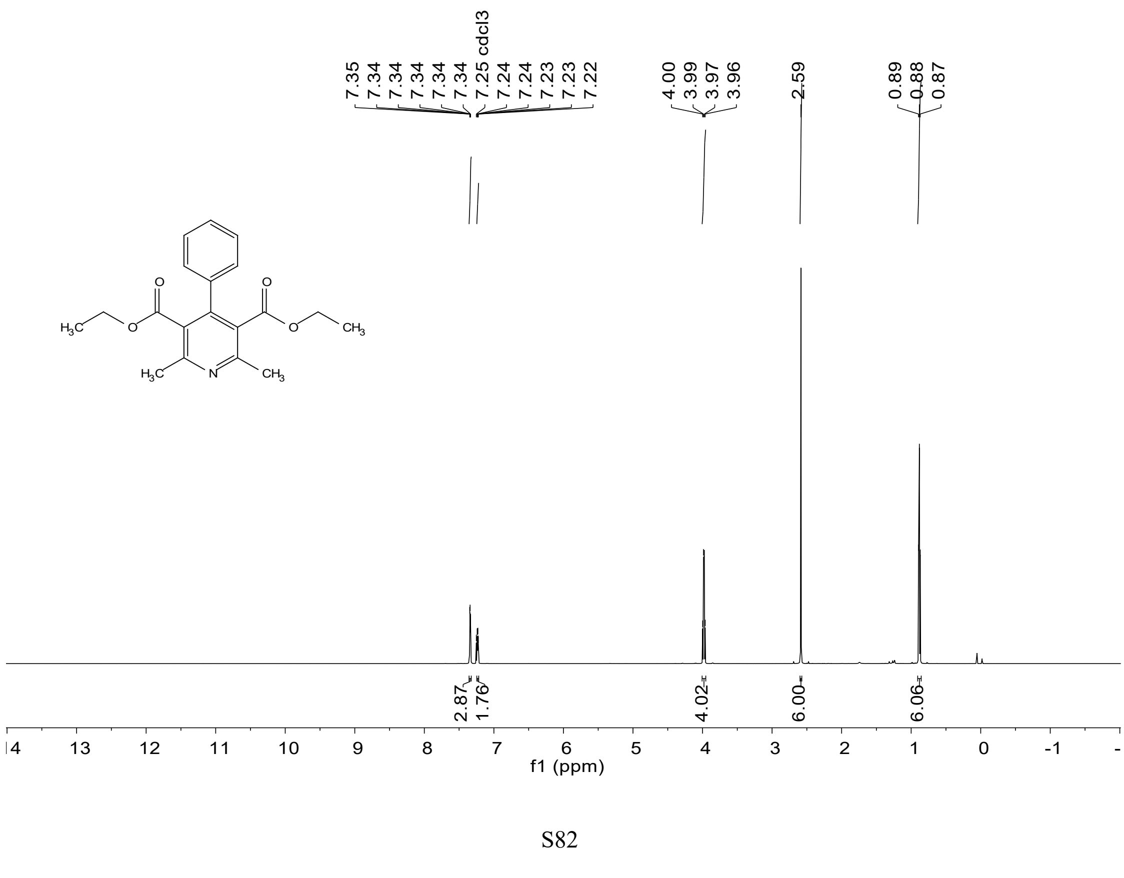
^{1}H NMR spectrum of 14c



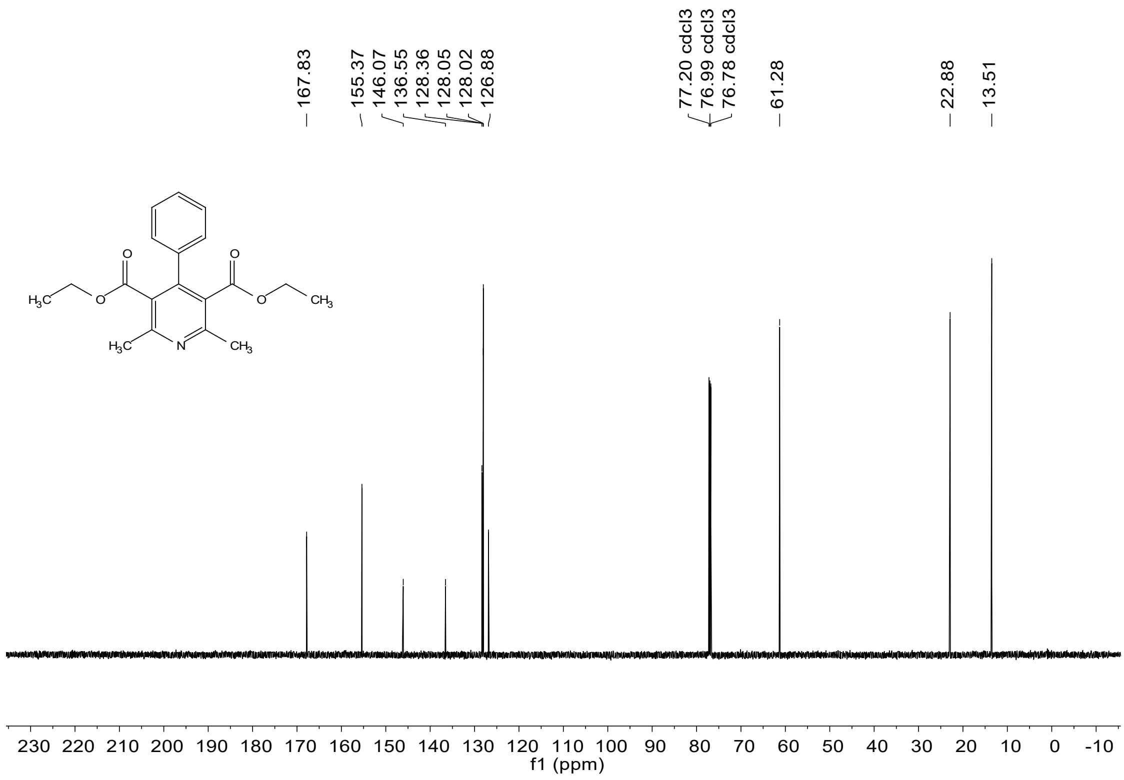
^{13}C NMR spectrum of 14c



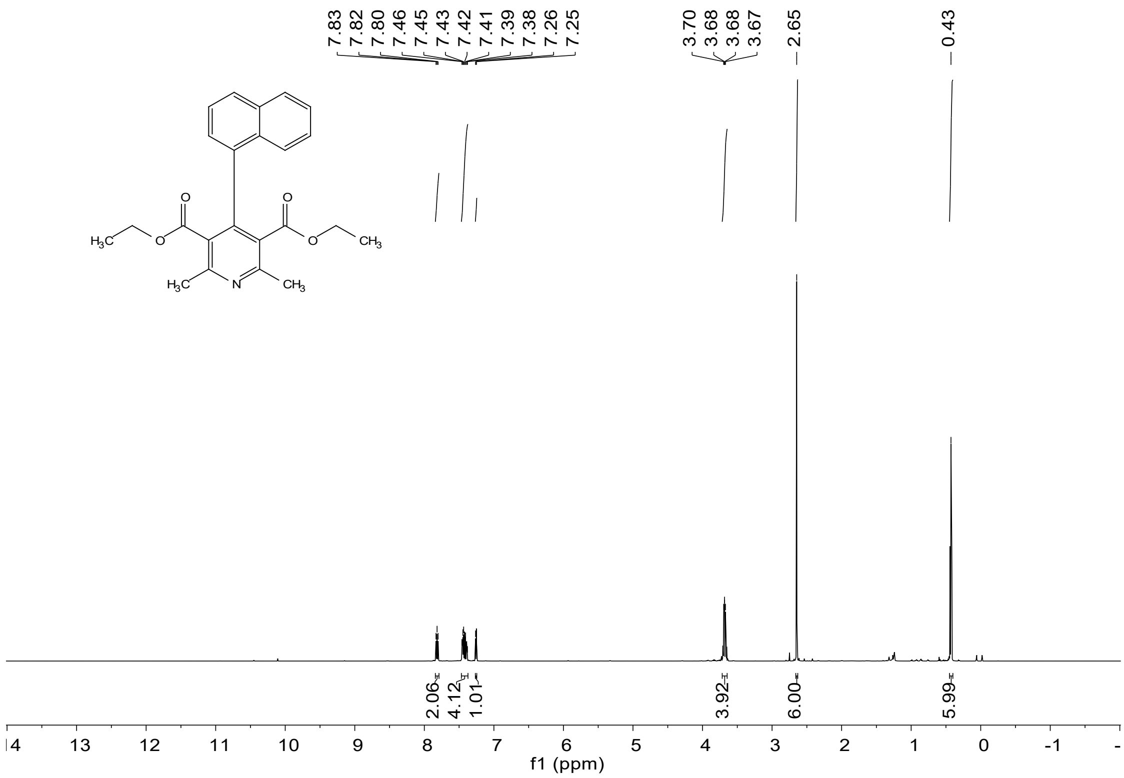
^{1}H NMR spectrum of 14d



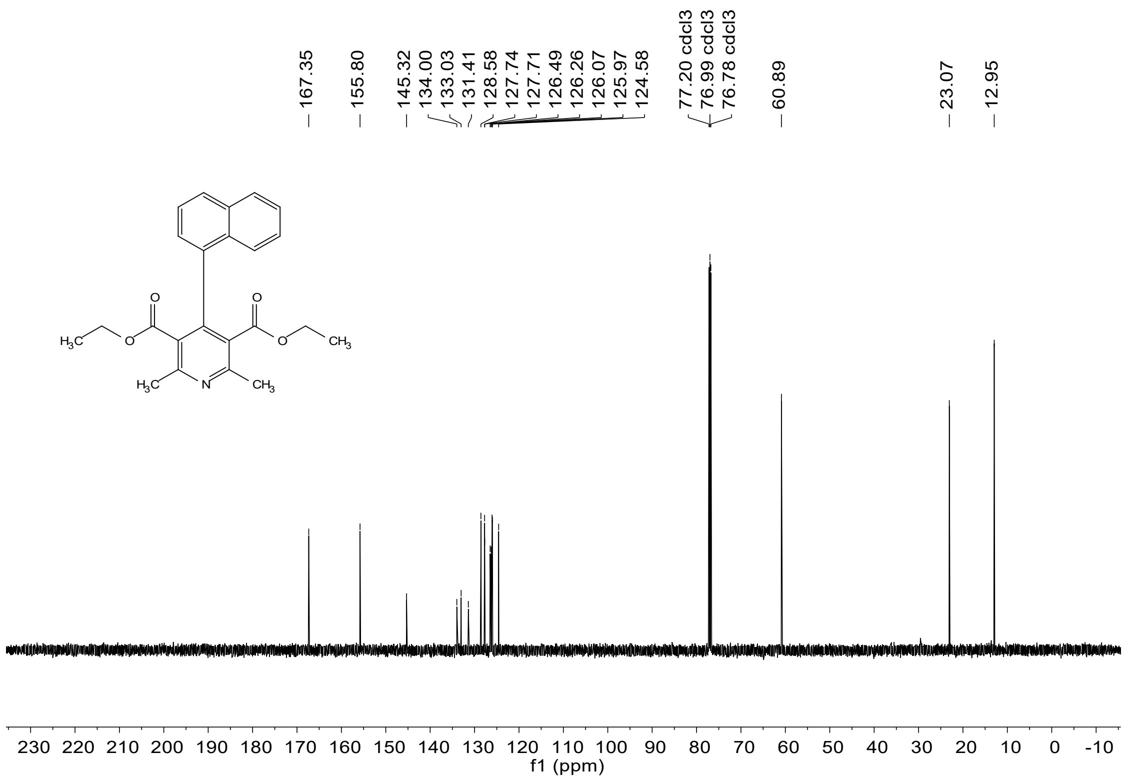
^{13}C NMR spectrum of 14d



^{1}H NMR spectrum of 14e



^{13}C NMR spectrum of 14e



8. References

[1] M. K. Sahoo, G. Jaiswal, J. Rana, E. Balaraman, *Chemistry* **2017**, *23*, 14167-14172.

[2] D. Bhattacharyya, S. Nandi, P. Adhikari, B. K. Sarmah, M. Konwar, A. Das, *Org. Biomol. Chem.* **2020**, *18*, 1214-1220.

[3] aN. Ravindranath, C. Ramesh, M. R. Reddy, B. Das, *Chem. Lett.* **2003**, *32*, 222-223; b) C.-F. Yao, S. More, M. Sastry, *Synlett* **2006**, *2006*, 1399-1403.

[4] J. Zhang, S. Chen, F. Chen, W. Xu, G.-J. Deng, H. Gong, *Adv. Synth. Catal.* **2017**, *359*, 2358-2363.

[5] F. Ye, X. Y. Liu, L. H. Qu, Y. Fu, L. X. Zhao, H. H. Qu, *Indian J. Heterocy. Ch.* **2014**, *24*, 167-174.

[6] K. Aghapoor, H. R. Darabi, F. Mohsenzadeh, Y. Balavar, H. Daneshyar, *Transition Metal Chemistry* **2009**, *35*, 49-53.

[7] aW. Waengdongbung, V. Hahnvajanawong, P. Theramongkol, *Orient. J. Chem.* **2016**, *32*, 941-945; bZ. L. Xie, D. L. Pennington, D. G. Boucher, J. Lo, M. J. Rose, *Inorg. Chem.* **2018**, *57*, 10028-10039.

[8] V. Siddaiah, G. M. Basha, G. P. Rao, U. V. Prasad, R. S. Rao, *Synth. Commun.* **2011**, *42*, 627-634.