Electronic Supplementary Information

TEMPO Mediated Oxidative Annulation of Aryl Methyl Ketones with Amines/Ammonium Acetate for Imidazole Synthesis

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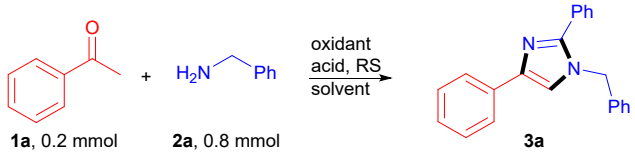
1. General Information

The reactions were carried out in Schlenk tubes of 25 mL under N_2 atmosphere. Reagents and solvents were used as received unless otherwise noted. Column chromatography was performed using Silica Gel 60 (300–400 mesh). The reactions were monitored by GC and GC-MS, GC-MS results were recorded on GC-MS QP2010, and GC analysis was performed on GC 2010 plus. The gas detection was monitored by Agilent Technologies 7820A GC system. The ^{1}H, ^{13}C NMR, and ^{19}F NMR spectra were recorded on a Brucker ADVANCE III spectrometer at 400 MHz, 101 MHz, and 372 MHz respectively, and chemical shifts were reported in parts per million (ppm). All solvents and reagents were purchased from Energy Chemical, Alfa Aesar, and Aladdin.

2. Optimization of the Reaction Conditions

2.1 The loading screen of $NaIO_4$ and TEMPO



1a, 0.2 mmol **2a**, 0.8 mmol **3a**

Table S1. Optimal amounts of $NaIO_4$ and TEMPO[a]

Entry	Amounts of $NaIO_4$	Amounts of TEMPO	Yield[b] (%)
1	2 equiv.	2 equiv.	88
2	2 equiv.	1 equiv.	56
3	2 equiv.	0.5 equiv.	31
4	2 equiv.	0.1 equiv.	12
5	1 equiv.	1 equiv.	18
6	1 equiv.	2 equiv.	37
7	0.5 equiv.	2 equiv.	10

[a]Reaction conditions: **1a** (0.2 mmol), **2a** (4.0 equiv.) in the solvent (2.0 mL) at 70 °C under N_2 for 12 h. [b]GC yield using tridecane as an internal standard.

The supplementary experiments including (1 to 2) equiv. of $NaIO_4$ and (0.1, 0.5, 1 to 2) equiv. of TEMPO have been screened, but no better results were observed when the reaction proceed under other conditions. These results indicate that sufficient oxidants and free radicals were necessary.

2.2 The investigation of oxidants under the standard conditions

Table S2. The investigation of oxidants[a]

Entry	Oxidant	Acid	Yield[b] (%)
1	$NaIO_4$	AcOH	88
2	$K_2S_2O_8$	AcOH	50
3	I_2	AcOH	35
4	$NaIO_3$	AcOH	15
5	$KBrO_3$	AcOH	10

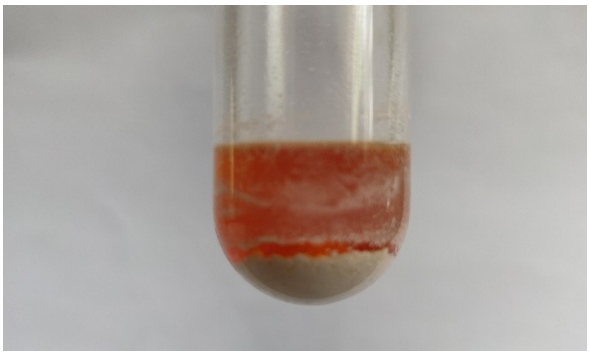
[a]Reaction conditions: **1a** (0.2 mmol), **2a** (4.0 equiv.) and TEMPO (4.0 equiv.) in the CH_3CN (2.0 mL) at 70 °C under N_2 for 12 h. [b]GC yield using tridecane as an internal standard.

These investigations suggest that $NaIO_4$ is the best oxidant, which provides the desired product in 88% yield.

3. General Experimental Procedure

3.1 General Experimental Procedure for the Synthesis of 1*H*-Imidazoles

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with aryl methyl ketones **1** (0.2 mmol), amines **2** (0.8 mmol), $NaIO_4$ (0.4 mmol), AcOH (0.4 mmol) and TEMPO (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, CH_3CN (2.0 mL) was added under N_2. The reaction mixture was stirred at 70 ºC for 12 h and monitored by GC or GC-MS. After completion, the reaction mixture was diluted with EtOAc and washed with H_2O. The organic layer was dried with anhydrous Na_2SO_4, filtered, and concentrated in vacuo. The crude product was purified by flash column chromatography on silica gel with petroleum ether / ethyl acetate to give the desired 1*H*-imidazoles **3**. The reaction mixture is shown as follow:



3.2 General Experimental Procedure for the Synthesis of 2*H*-Imidazoles

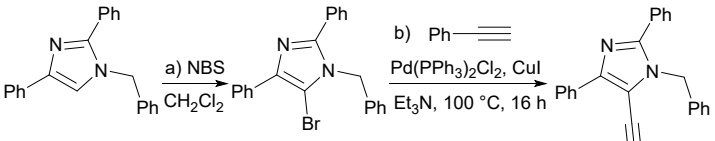
An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with aryl methyl ketones **1** (0.4 mmol), NH_4OAc (1.6 mmol), $NaIO_4$ (0.4 mmol) and TEMPO (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, CH_3CN (1.0 mL) was added under N_2. The reaction mixture was stirred at 70 ºC for 12 h and monitored by GC or GC-MS. After completion, the reaction mixture was diluted with EtOAc and washed with H_2O. The organic layer was dried with anhydrous Na_2SO_4, filtered, and concentrated in vacuo. The crude product was purified by flash column chromatography on silica gel with petroleum ether / ethyl acetate to give the desired 2*H*-imidazoles **4**.

4. Synthetic Utility

4.1 Preparation of 1-benzyl-2,4-diphenyl-1*H*-Imidazole at Gram-scales

5 mmol gram-scale: an oven-dried 250 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with acetophenone **1a** (5.0 mmol), benzylamine **2a** (20.0 mmol), $NaIO_4$ (10.0 mmol), and TEMPO (10.0 mmol) was evacuated and backfilled with N_2 three times. Then, CH_3CN (50.0 mL) and was added under N_2. The reaction mixture was stirred at 70 ºC for 12 h. After completion, the reaction mixture was diluted with EtOAc and washed with H_2O. The organic layer was dried with anhydrous Na_2SO_4, filtered, and concentrated in vacuo. The crude product was purified by column chromatography on silica gel with petroleum ether / ethyl acetate (20/1) to afford 1-benzyl-2,4-diphenyl-1*H*-imidazole (**3a**) in 85% yield (1.32g).

4.2 Synthesis of 1-benzyl-2,4-diphenyl-5-(phenylethynyl)-1*H*-imidazole (6)



Sonogashira coupling functionalization of imidazoles

a) Synthesis of 1-benzyl-5-bromo-2,4-diphenyl-1*H*-imidazole (**5**)[1]: A round-bottom flask equipped with a magnetic stirrer bar was charged with 1 mmol of imidazole (**3a**) dissolved in 5.0 mL CH_2Cl_2, and the solution of NBS (1.1 equiv. in 5.0 mL CH_2Cl_2) was dropwise added at room temperature. After completion of the reaction (as monitored by TLC), the mixture was directly subjected to flash column chromatography on silica gel to give the 1-benzyl-5-bromo-2,4-diphenyl-1*H*-imidazole (**5**) in 90% yield.

b) Synthesis of 1-benzyl-2,4-diphenyl-5-(phenylethynyl)-1*H*-imidazole (**6**)[2]: An oven-dried Schlenk tube of 25 mL equipped with a magnetic stir bar was charged with **5** (0.2 mmol), $Pd(PPh_3)_2Cl_2$ (10 mol %) and CuI (10 mol %). After charging nitrogen for three times, phenylacetylene (0.24 mmol, 1.2 equiv.), and triethylamine (1 mL) was added under nitrogen atmosphere, and the reaction mixture was stirred at 100 °C in IKA for 16 h. The precipitate was removed by filtration and washed with EtOAc, and the filtrate was washed with brine, dried over Na_2SO_4 and then concentrated under vacuum. The title compound was purified by column chromatography on silica gel and eluted with petroleum ether / ethyl acetate (10/1) to afford the 1-benzyl-2,4-diphenyl-5-(phenylethynyl)-1*H*-imidazole (**6**) in 91% yield.

5. X-ray Crystallographic Data of 3k

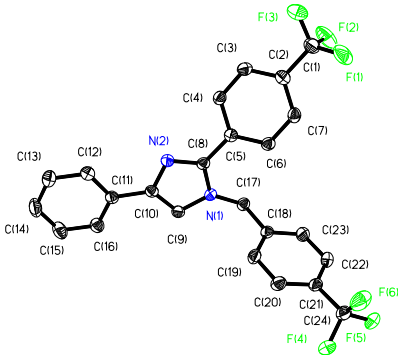


Figure S1. X-ray crystal structure of compound 3k (CCDC number: 2129228)

Table S3. Summary of X-ray crystallographic data for compound 3k.

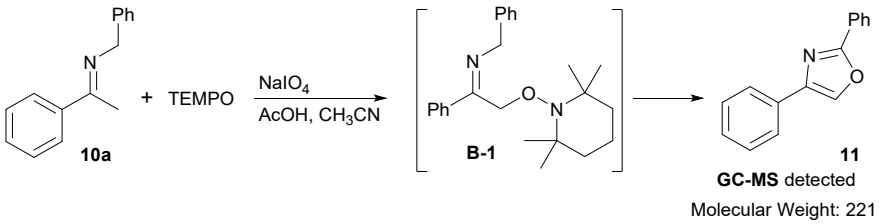
Empirical formula	$C_{24}H_{16}F_6N_2$
Formula weight	446.39
Temperature/K	293(2)
Crystal system	monoclinic
Space group	N/A
a/Å	7.6201(11)
b/Å	11.120(2)
c/Å	12.625(2)
α/°	106.776(4)

β/°	99.113(4)
γ/°	96.569(6)
Volume/Å^3	996.7(3)
Z	2
ρ_{calc}g/cm^3	1.487
μ/mm^{-1}	0.127
F(000)	456.0
Crystal size/mm^3	0.28 × 0.22 × 0.21
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	4.3 to 50.02
Index ranges	-9 ≤ h ≤ 9, -13 ≤ k ≤ 13, -15 ≤ l ≤ 15
Reflections collected	12636
Independent reflections	3488 [R_{int} = 0.0226, R_{sigma} = N/A]
Data/restraints/parameters	3488/0/289
Goodness-of-fit on F^2	1.044
Final R indexes [I>=2σ (I)]	R_1 = 0.0349, wR_2 = 0.0883
Final R indexes [all data]	R_1 = 0.0366, wR_2 = 0.0899
Largest diff. peak/hole / e Å^{-3}	0.21/-0.26

6. Investigations of the Reaction Mechanism

6.1 Verification of addition of *N*-benzyl-1-phenylethan-1-imine (7) and TEMPO

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with *N*-benzyl-1-phenylethan-1-imine (**7**, 0.2 mmol), TEMPO (0.4 mmol), and $NaIO_4$ (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, AcOH (0.4 mmol) and CH_3CN (2.0 mL) were added under N_2. The reaction mixture was stirred at 70 ºC for 1.5 h. After completion, the reaction mixture was diluted with EtOAc and monitored by GC-MS. As shown in Figure S2, a cyclization product (**11**) was observed without the use of benzylamine (**2a**), which can be generated from intermediate **B-1**.



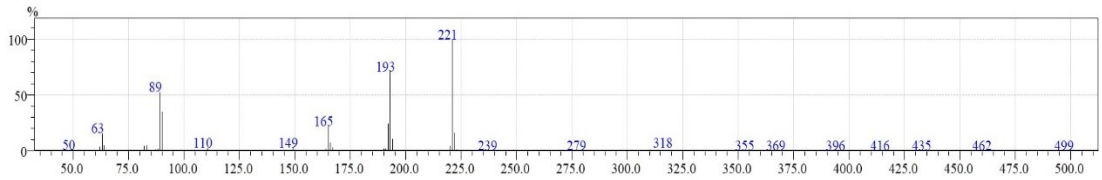


Figure S2. GC-MS analysis of 2-phenylnaphtho[2,1-*d*]oxazole under standard reaction conditions

[MS Spectrum]

# of Peaks 210

Raw Spectrum 13.370 -> 13.455 (scan : 1175 -> 1192) Base Peak m/z 221.00 (Inten : 53,996)

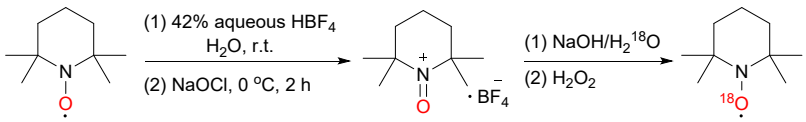
Background 13.375 (scan : 1176)

m/z	Absolute Intensity	Relative Intensity						
62.05	2057	3.81	149.05	1389	2.57	194.00	5922	10.97
63.05	8550	15.83	164.10	855	1.58	220.00	2207	4.09
64.05	2585	4.79	165.10	11776	21.81	**221.00**	**53996**	**100.00**
82.05	2311	4.28	166.05	4204	7.79	222.00	8738	16.18
83.05	2694	4.99	167.05	1764	3.27	**223.00**	**587**	**1.09**
88.05	792	1.47	190.00	859	1.59	229.10	38	0.07
89.05	28563	52.90	191.00	813	1.51			
90.05	18982	35.15	192.00	13377	24.77			
110.55	1369	2.54	193.05	38255	70.85			

6.2 TEMP^{18}O-labelling experiments

To further verify the product (**11**), a TEMP^{18}O-labelling experiment was conducted.

A) Synthesis of TEMP^{18}O.[3]



To a solution of TEMPO (4.68 g, 30 mmol) in H_2O (15 mL, 2 M) was added dropwise 42% aqueous HBF_4 (14.9 mL, 30 mmol) at room temperature. After the solution became to amber color, the aqueous NaOCl solution (16.0 mL, 30 mmol) was added dropwise at 0 ºC. When it finished, the reaction mixture stirred for additional 1 h at 0 ºC. Finally, the reaction mixture was filtered and the yellow crystalline precipitate was washed with ice-cold 5% aqueous $NaHCO_3$ (6.0 mL), water (6.0 mL), and ice-clod ether (60.0 mL). The bright yellow solid was dried at 50 ºC in vacuo to gain the TEMPO$^+$BF$_4^-$ (5.1 g, 70 %)

To the solution of TEMPO$^+$BF$_4^-$ (0.9710 g, 4 mmol) in $H_2{}^{18}O$ (1.7 mL) was added concentrated NaOH (12 N, 1.5 mL $H_2{}^{18}O$) at 0 ºC for 2 h and the color of solution was changed from orange to slightly yellow. Then, 30% H_2O_2 (0.2 mL) was added to the reaction mixture. When the color of reaction mixture became slightly red, the reaction mixture was extracted with ether. The organic layer was dried over anhydrous Na_2SO_4, filtered and concentrated under reduced pressure to gain the red crystalline solid (TEMP^{18}O), which was dried at room temperature in vacuo. The ratio of TEMP^{18}O/TEMP^{16}O was **1:0.181** determined by the GC-MS analysis, and the result is shown in Figure S3.

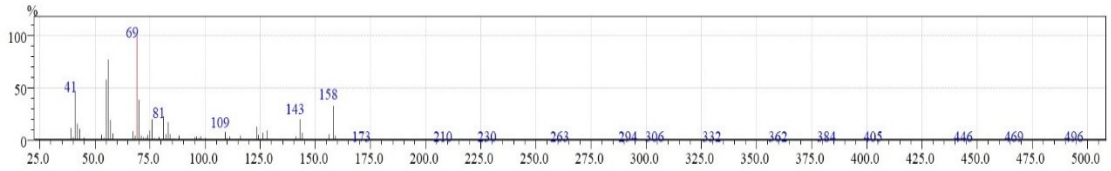


Figure S3. GC-MS analysis of TEMP^{18}O

[MS Spectrum]

# of Peaks 299

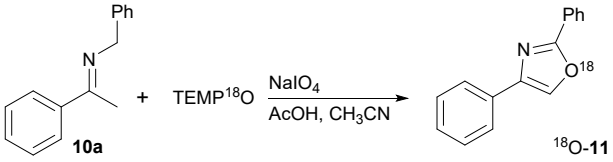
Raw Spectrum 5.875 -> 5.955 (scan : 576 -> 592) Base Peak m/z 69.05 (Inten : 2,200,485)

Background 5.945 (scan : 590)

m/z	Absolute Intensity	Relative Intensity						
39.002	57975	11.72	42.05	342828	15.58	53.05	115425	5.25
41.05	1044251	47.46	43.05	239964	10.91	55.05	1277460	58.05

56.05	1696496	77.10	76.05	446074	20.27	126.10	163007	7.41
57.05	426056	19.36	81.05	504771	22.94	128.05	207250	9.42
58.05	138285	6.28	82.05	133842	6.08	143.10	438908	19.95
67.00	193573	8.80	83.05	383612	17.43	144.10	159209	7.24
69.05	2200485	100.00	84.05	136140	6.19	**156.10**	**133235**	**6.05**
70.05	852143	38.73	109.05	171766	7.81	**158.10**	**735434**	**33.42**
74.05	115011	5.23	123.10	291143	13.23	159.10	101746	4.62
75.05	210997	9.59	124.10	115324	5.24	160.10	6602	0.30

B) Verification of addition of *N*-benzyl-1-phenylethan-1-imine (**7**) and TEMP^{18}O



GC-MS detected

Molecular Weight: 223

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with *N*-benzyl-1-phenylethan-1-imine (**10a**, 0.2 mmol), TEMP^{18}O (0.4 mmol), and $NaIO_4$ (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, AcOH (0.4 mmol) and CH_3CN (2.0 mL) were added under N_2. The reaction mixture was stirred at 70 ºC for 1.5 h. After completion, the reaction mixture was diluted with EtOAc and monitored by GC-MS. As shown in Figure S4, a ^{18}O-labelling product (^{18}O-**11**) was observed by GC-MS. The ratio of (^{18}O)-**11**/(^{16}O)-**11** was 1:0.072 determined that almost all of oxygen of oxazole (**11**) comes from TEMPO.

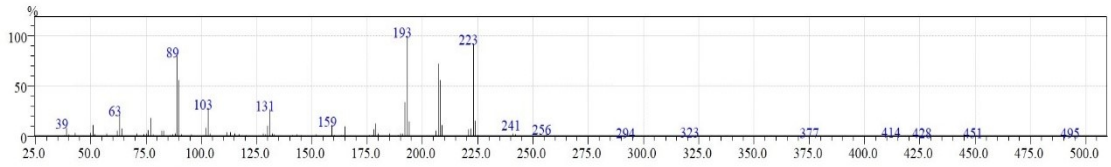


Figure S4. GC-MS analysis of ^{18}O-labelling oxazole (11)

[MS Spectrum]

# of Peaks 290

Raw Spectrum 13.355 -> 13.475 (scan : 2072 -> 2096) Base Peak m/z 193.05 (Inten : 33,147)

Background 13.400 -> 13.715 (scan : 2081 -> 2144)

m/z	Absolute Intensity	Relative Intensity						
39.05	2849	8.60	130.05	3440	10.38	207.00	23947	72.24
51.05	3763	11.35	131.05	8894	26.83	208.00	18526	55.89
63.05	7255	21.89	159.15	3696	11.15	209.00	3620	10.92
64.05	2497	7.53	165.10	3207	9.68	**221.00**	**2204**	**6.65**
77.05	6115	18.45	178.05	2294	6.92	222.05	2421	7.30
89.05	26610	80.28	179.05	4051	12.22	**223.00**	**30665**	**92.51**
90.05	18416	55.56	192.05	11088	33.45	224.00	5141	15.51
102.05	2630	7.93	193.05	33147	100.00	225.05	442	1.33
103.05	9388	28.32	194.05	4783	14.43	226.00	42	0.13

6.3 The detection of hydrogen radical

In this reaction, we speculated that there would be hydrogen radicals generated. And they may directly form H_2 or be trapped by TEMPO to give TEMPOH. To further verify the mechanism, we tried to detect the content of H_2

after the reaction completed and monitor the amount of TEMPOH changes at different time.

A) The detection of H_2

The mixture of hydrogen and nitrogen in a volume ratio of 1:100 was performed on Agilent Technologies 7820A GC system as the standard spectrum (Figure S4).

In sharp contrast, the content of H_2 was detected in the optimal conditions. The result was shown in Figure S5. The experimental procedure is shown as follow:

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with acetophenone (**1a**, 0.2 mmol), benzylamine (**2a**, 0.8 mmol), $NaIO_4$ (0.4 mmol), AcOH (0.4 mmol) and TEMPO (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, CH_3CN (2.0 mL) was added under N_2. The reaction mixture was stirred at 70 ºC for 12 h. After completion, 1 mL gas was extracted and analyzed by Agilent Technologies 7820A GC system. The GC spectrum is shown in Figure S5.

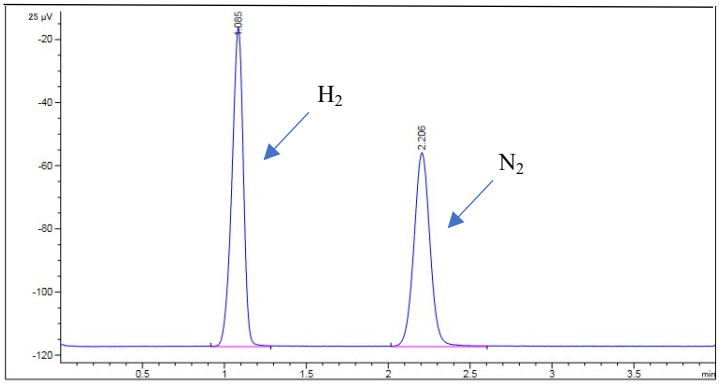


Figure S4. GC detection of H_2 and N_2 as standard spectrum

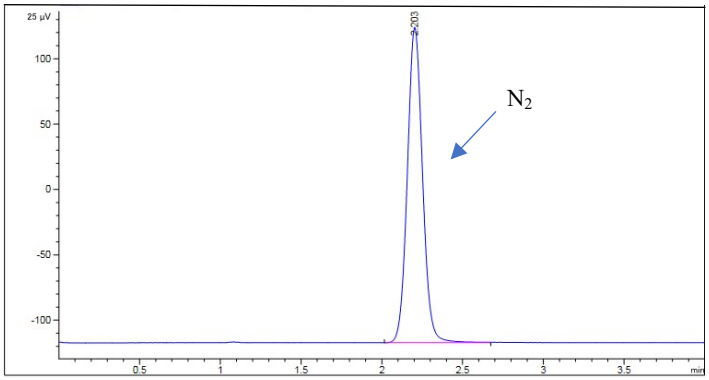


Figure S5. GC detection of the content of H_2

The spectrum suggests that there is no H_2 generated in the reaction system.

B) The tracking detection of TEMPOH

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with acetophenone (**1a**, 0.2 mmol), benzylamine (**2a**, 0.8 mmol), $NaIO_4$ (0.4 mmol), AcOH (0.4 mmol) and TEMPO (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, CH_3CN (2.0 mL) and tridecane (as an internal standard) were added under N_2. The reaction mixture was stirred at 70 ºC, and monitored by GC at 0 h, 1 h, 6 h, 12 h, 24 h, respectively.

Table S3. The detection of TEMP, TEMPOH and TEMPO[a]

Time	TEMP	TEMPOH	TEMPO

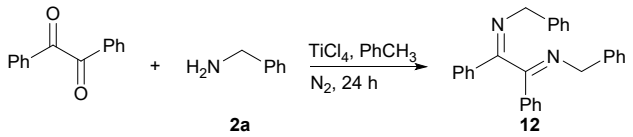
0 h	0	10%	190%
1 h	44%	8%	120%
6 h	56%	7%	112%
12 h	93%	6%	103%
24 h	94%	7%	103%

[a]Reaction conditions: **1a** (0.2 mmol), **2a** (4.0 equiv.) and TEMPO (4.0 equiv.) in the CH_3CN (2.0 mL) at 70 °C under N_2 for 12 h. [b]GC yield using tridecane as an internal standard.

The result shows that no TEMPOH was produced in the reaction system, which excluded the possibility of H radical removal.

6.4 Synthesis and cyclization of dibenzyl-1,2-diphenylethane-1,2-diimine

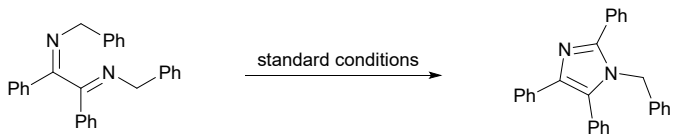
A) The synthesis of dibenzyl-1,2-diphenylethane-1,2-diimine[4]



2a **12**

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with benzil (0.5 mmol), was evacuated and backfilled with N_2 three times. Then, benzylamine (2.2 equiv.), triethylamine (10.0 equiv.) and dry $PhCH_3$ (3.5 mL) were added under N_2, and a solution of $TiCl_4$ (1 M in toluene, 1.5 equiv.) was added dropwise at 0°C. The reaction mixture was stirred at room temperature for 24 h. After completion, the reaction mixture was quenched by addition of sat. Na_2CO_3 solution and diluted with EtOAc. Then, the heterogeneous mixture was centrifuged to sediment TiO_x, the phases were carefully separated by decantation, the aq./TiO_x was again washed with EtOAc, centrifuged and the combined organic phases were evaporated. The residue was purified by column chromatography petroleum ether / ethyl acetate (10/1) or recrystallization to afford the dibenzyl-1,2-diphenylethane-1,2-diimine (**12**) as white solid.

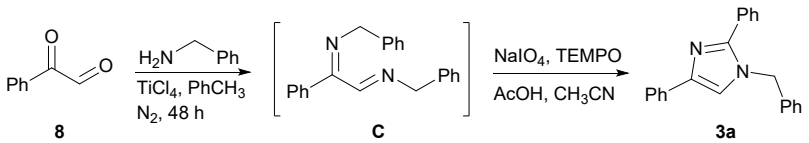
B) The cyclization of dibenzyl-1,2-diphenylethane-1,2-diimine



12 **13**

An oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with dibenzyl-1,2-diphenylethane-1,2-diimine (**12**, 0.2 mmol), $NaIO_4$ (0.4 mmol), AcOH (0.4 mmol) and TEMPO (0.4 mmol), was evacuated and backfilled with N_2 three times. Then, benzylamine (**2a**, 0.2 mmol) and CH_3CN (2.0 mL) were added under N_2. The reaction mixture was stirred at 70 ºC for 12 h. After completion, the reaction mixture was diluted with EtOAc and washed with H_2O. The organic layer was dried with anhydrous Na_2SO_4, filtered, and concentrated in vacuo. The crude product was purified by flash column chromatography on silica gel with petroleum ether / ethyl acetate (20/1) to give the desired white solid **13**.

6.5 One-pot synthesis of 3a from 2-oxo-2-phenylacetaldehyde



8 **C** **3a**

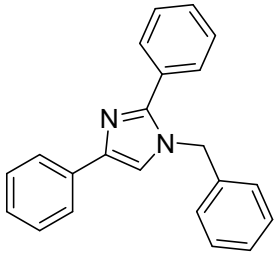
First, an oven-dried 25 mL Schlenk tube, which was equipped with a magnetic stir bar and charged with 2-oxo-

2-phenylacetaldehyde (0.2 mmol), was evacuated and backfilled with N_2 three times. Then, benzylamine (2.2 equiv.), triethylamine (10.0 equiv.) and dry $PhCH_3$ (1.5 mL) were added under N_2, and a solution of $TiCl_4$ (1 M in toluene, 1.5 equiv.) was added dropwise at 0 °C. The reaction mixture was stirred at room temperature for 48 h. After completion, the reaction mixture was quenched by addition of sat. Na_2CO_3 solution and diluted with EtOAc. Then, the heterogeneous mixture was centrifuged to sediment TiO_x, the phases were carefully separated by decantation, the aq./TiO_x was again washed with EtOAc, centrifuged and the combined organic phases were evaporated to obtain the residue.

Next, the residue was charged in an oven-dried 25 mL Schlenk tube equipped with a magnetic stir bar, which was evacuated and backfilled with N_2 three times. $NaIO_4$ (0.4 mmol), AcOH (0.4 mmol), TEMPO (0.4 mmol), and CH_3CN (2.0 mL) were then added under N_2. The reaction mixture was stirred at 70 ºC for 12 h, which provides the desired product **3a** in 40% yield.

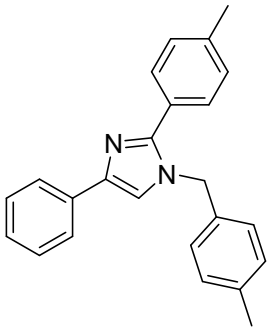
7. ^{1}H, ^{13}C and ^{19}F NMR Spectra Data of the Imidazoles

1-benzyl-2,4-diphenyl-1*H*-imidazole (3a)[5]



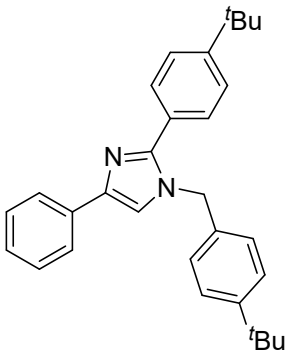
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 88% yield (54.6 mg). mp 123-124 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.88 (d, J = 7.6 Hz, 2H), 7.65 (d, J = 4.9 Hz, 2H), 7.46 (d, J = 4.6 Hz, 3H), 7.39 (m, 5H), 7.27 (d, J = 10.4 Hz, 2H), 7.17 (d, J = 7.1 Hz, 2H), 5.25 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.5, 140.4, 135.7, 133.0, 129.3, 127.9, 127.7, 127.5, 127.4, 126.9, 125.7, 125.6, 123.8, 115.7, 49.4.

1-(4-methylbenzyl)-4-phenyl-2-(p-tolyl)-1*H*-imidazole (3b)[6]



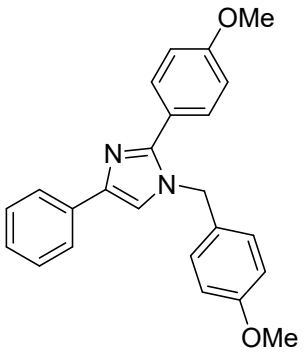
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 91% yield (61.5 mg). mp 94-95 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.92 (d, J = 7.4 Hz, 2H), 7.59 (d, J = 8.0 Hz, 2H), 7.44 (t, J = 7.7 Hz, 2H), 7.34 – 7.29 (m, 3H), 7.28 (s, 1H), 7.23 (d, J = 7.9 Hz, 2H), 7.10 (d, J = 8.0 Hz, 2H), 5.21 (s, 2H), 2.46 (s, 3H), 2.42 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 148.4, 141.1, 138.6, 137.4, 134.0, 133.7, 129.4, 129.1, 128.7, 127.4, 126.5, 126.3, 124.7, 116.4, 50.0, 21.1, 20.9.

1-(4-(tert-butyl)benzyl)-2-(4-(tert-butyl)phenyl)-4-phenyl-1*H*-imidazole (3c)[5]



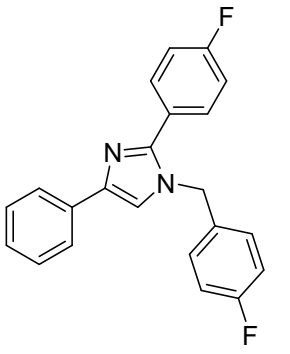
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 86% yield (72.6 mg). mp 189-190 °C.^{1}H NMR (400 MHz, $CDCl_3$): δ 7.83 (d, J = 7.2 Hz, 2H), 7.56 (d, J = 7.5 Hz, 2H), 7.43 (d, J = 7.5 Hz, 2H), 7.34 (t, J = 7.8 Hz, 4H), 7.21 (s, 2H), 7.07 (d, J = 7.4 Hz, 2H), 5.18 (s, 2H), 1.33 (s, 9H), 1.31 (s, 9H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 151.4, 150.3, 148.1, 140.8, 133.6, 133.4, 128.1, 127.9, 127.0, 126.0, 125.8, 125.3, 125.0, 124.3, 116.0, 49.5, 34.1, 33.9, 30.7, 30.6.

1-(4-methoxybenzyl)-2-(4-methoxyphenyl)-4-phenyl-1*H*-imidazole (3d)[6]



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 41% yield (30.4 mg). mp 117-119 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.84 (d, J = 7.6 Hz, 2H), 7.55 (d, J = 7.9 Hz, 2H), 7.36 (t, J = 7.3 Hz, 2H), 7.23 (d, J = 7.2 Hz, 1H), 7.19 (d, J = 6.4 Hz, 1H), 7.04 (d, J = 8.1 Hz, 2H), 6.95 (d, J = 8.0 Hz, 2H), 6.86 (d, J = 8.1 Hz, 2H), 5.08 (s, 2H), 3.81 (s, 3H), 3.77 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 160.0, 159.1, 148.2, 140.9, 134.1, 130.2, 128.7, 128.3, 127.9, 126.5, 124.7, 122.9, 116.3, 114.2, 113.9, 55.1, 54.9, 49.8.

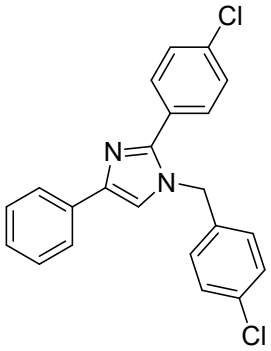
1-(4-fluorobenzyl)-2-(4-fluorophenyl)-4-phenyl-1*H*-imidazole (3e)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 82% yield (56.8 mg). mp

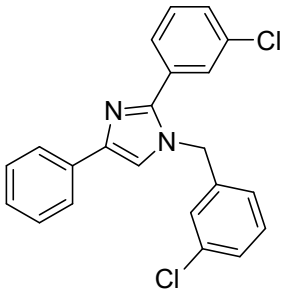
82-83 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.83 (d, J = 7.5 Hz, 2H), 7.66 – 7.51 (m, 2H), 7.38 (t, J = 7.4 Hz, 2H), 7.29 – 7.21 (m, 2H), 7.09 (m, 6H), 5.16 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.2 (J = 247.8 Hz), 162.4 (J = 245.9 Hz), 147.5, 141.6, 133.8, 132.3 (d, J = 3.3 Hz), 130.9 (d, J = 8.4 Hz), 128.6, 128.4, 128.3, 126.9, 116.7, 116.1, 115.9 (d, J = 6.0 Hz), 115.6. ^{19}F NMR (376 MHz, $CDCl_3$) δ -111.6, -113.8.

1-(4-chlorobenzyl)-2-(4-chlorophenyl)-4-phenyl-1*H*-imidazole (3f)[5]



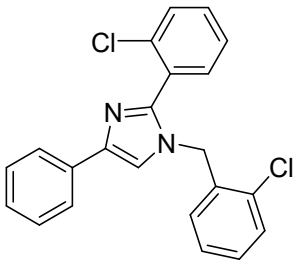
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 81% yield (61.2 mg). mp 100-101 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.80 (d, J = 7.5 Hz, 2H), 7.48 (d, J = 7.8 Hz, 2H), 7.35 (t, J = 7.7 Hz, 4H), 7.28 (d, J = 7.7 Hz, 2H), 7.23 (d, J = 8.4 Hz, 1H), 7.18 (s, 1H), 6.98 (d, J = 7.8 Hz, 2H), 5.09 (s, 2H).
^{13}C NMR (101 MHz, $CDCl_3$): δ 147.0, 141.6, 134.9, 134.8, 133.8, 133.5, 129.9, 129.0, 128.7, 128.5, 128.3, 127.7, 126.8, 124.7, 116.8, 49.7.

1-(3-chlorobenzyl)-2-(3-chlorophenyl)-4-phenyl-1*H*-imidazole (3g)[6]



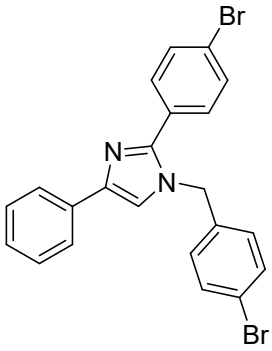
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 87% yield (65.8 mg). mp 125-127 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.81 (d, J = 7.5 Hz, 2H), 7.61 (s, 1H), 7.36 (q, J = 6.4, 5.5 Hz, 4H) 7.31 (d, J = 7.7 Hz, 1H), 7.24 (d, J = 9.8 Hz, 3H), 7.20 (s, 1H), 7.07 (s, 1H), 6.93 (d, J = 6.6 Hz, 1H), 5.11 (s, 2H).
^{13}C NMR (101 MHz, $CDCl_3$): δ 146.8, 141.8, 138.4, 134.9, 134.6, 133.6, 131.8, 130.3, 129.8, 129.1, 129.0, 128.5, 128.3, 127.0, 126.7, 126.6, 124.8, 124.6, 117.1, 49.9.

1-(2-chlorobenzyl)-2-(2-chlorophenyl)-4-phenyl-1*H*-imidazole (3h)[6]



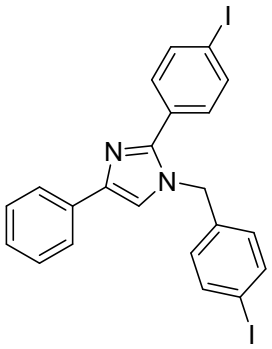
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 80% yield (60.5 mg). mp 135-136 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.81 (d, J = 7.5 Hz, 2H), 7.47 (d, J = 6.2 Hz, 2H), 7.35 (m, 5H), 7.20 (m, 4H), 6.93 (d, J = 7.3 Hz, 1H), 5.10 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 145.7, 141.4, 134.5, 133.7, 133.5, 132.9, 132.6, 130.8, 129.8, 129.5, 129.3, 129.0, 128.3, 127.0, 126.8, 126.6, 124.7, 115.6, 47.9.

1-(4-bromobenzyl)-2-(4-bromophenyl)-4-phenyl-1*H*-imidazole (3i)[7]



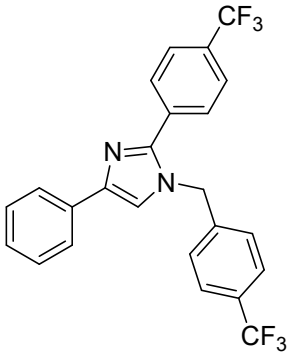
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 76% yield (70.8 mg). mp 102-103 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.80 (d, J = 7.5 Hz, 2H), 7.53 (d, J = 7.9 Hz, 2H), 7.43 (t, J = 9.9 Hz, 4H), 7.35 (t, J = 7.4 Hz, 2H), 7.24 (d, J = 6.9 Hz, 1H), 7.19 (s, 1H), 6.94 (d, J = 7.8 Hz, 2H), 5.09 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.2, 141.8, 135.4, 133.6, 132.1, 131.8, 130.2, 129.0, 128.5, 128.1, 126.9, 124.8, 123.3, 122.0, 116.9, 49.8.

1-(4-iodobenzyl)-2-(4-iodophenyl)-4-phenyl-1*H*-imidazole (3j)



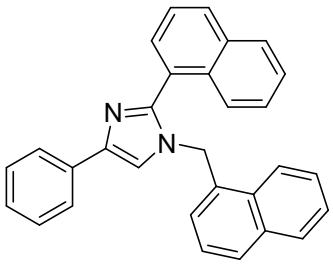
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 62% yield (69.7 mg). mp 130-131 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.79 (d, J = 7.6 Hz, 2H), 7.73 (d, J = 7.6 Hz, 2H), 7.64 (d, J = 7.6 Hz, 2H), 7.35 (t, J = 7.3 Hz, 2H), 7.28 (d, J = 7.6 Hz, 2H), 7.24 (d, J = 5.6 Hz, 1H), 7.19 (s, 1H), 6.81 (d, J = 7.7 Hz, 2H), 5.08 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.3, 141.8, 138.0, 137.7, 136.1, 133.6, 129.6, 128.5, 128.3, 127.0, 124.8, 117.0, 95.2, 93.5, 49.9. HRMS (EI) m/z: $[M]^+$ calcd for $C_{22}H_{16}I_2N_2$ 561.9403; found 561.9400.

4-phenyl-1-(4-(trifluoromethyl)benzyl)-2-(4-(trifluoromethyl)phenyl)-1*H*-imidazole (3k)[7]



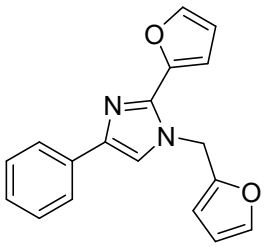
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a yellow solid in 78% yield (69.6 mg). mp 107-108 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.85 (d, J = 7.6 Hz, 2H), 7.77 – 7.66 (m, 4H), 7.63 (d, J = 7.8 Hz, 2H), 7.39 (t, J = 7.3 Hz, 2H), 7.28 (s, 2H), 7.26 – 7.12 (m, 2H), 5.29 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.0, 142.5, 140.4, 133.6, 133.5, 131.0 (q, J =99.0 Hz),130.6 (q, J=99.8Hz), 129.1, 128.7, 127.3, 126.7, 126.2 (q, J = 3.6 Hz), 125.7 (q, J = 3.6 Hz), 125.0, 117.5, 50.2.^{19}F NMR (376 MHz, $CDCl_3$): δ -62.7(s, 1F), -62.8(s, 1F).

2-(naphthalen-1-yl)-1-(naphthalen-1-ylmethyl)-4-phenyl-1*H*-imidazole (3l)[7]



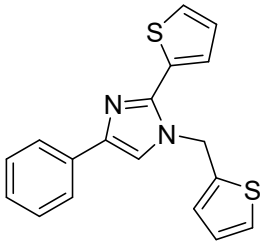
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow oil in 79% yield (64.8 mg).
^{1}H NMR (400 MHz, $CDCl_3$): δ 7.94 (q, J = 12.7, 10.2 Hz, 3H), 7.84 (d, J = 7.9 Hz, 3H), 7.80 (d, J = 8.5 Hz, 1H), 7.65 (d, J = 7.0 Hz, 1H), 7.61 (d, J = 8.4 Hz, 1H), 7.57 – 7.52 (m, 2H), 7.51 (d, J = 8.2 Hz, 1H), 7.46 (d, J = 7.9 Hz, 1H), 7.43 (d, J = 5.6 Hz, 1H), 7.39 (d, J = 9.4 Hz, 1H), 7.34 (t, J = 6.7 Hz, 3H), 7.27 (s, 1H), 7.20 (d, J = 7.0 Hz, 1H), 5.40 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.0, 141.5, 134.1, 133.8, 133.7, 132.9, 131.9, 130.6, 130.0, 128.9, 128.8, 128.7, 128.5, 128.3, 128.0, 127.0, 126.7, 126.6, 126.3, 126.1, 125.9, 125.8, 125.4, 125.0, 124.9, 122.5, 115.8, 48.6.

2-(furan-2-yl)-1-(furan-2-ylmethyl)-4-phenyl-1*H*-imidazole (3m)[6]



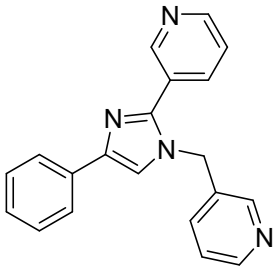
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 36% yield (20.9 mg). mp 103-104 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.8 (d, J = 7.5 Hz, 2H), 7.5 (s, 1H), 7.4 (m, 3H), 7.2 (d, J = 9.2 Hz, 2H), 6.9 (s, 1H), 6.5 (s, 1H), 6.3 (d, J = 17.3 Hz, 2H), 5.4 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 149.3, 145.4, 143.0, 142.7, 141.7, 139.1, 133.7, 128.4, 126.9, 125.0, 116.6, 111.5, 110.6, 110.2, 108.9, 43.9.

4-phenyl-2-(thiophen-2-yl)-1-(thiophen-2-ylmethyl)-1*H*-imidazole (3n)[7]



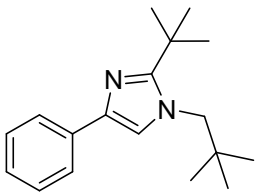
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 76% yield (49.0 mg). mp 92-93 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.81 (d, J = 7.5 Hz, 2H), 7.42 (d, J = 4.7 Hz, 1H), 7.36 (t, J = 7.4 Hz, 2H), 7.31 (d, J = 8.0 Hz, 3H), 7.23 (d, J = 7.3 Hz, 1H), 7.10 (s, 1H), 7.02 – 6.97 (m, 1H), 6.95 (s, 1H), 5.45 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 142.0, 141.8, 138.7, 133.7, 131.9, 127.5, 127.3, 127.2, 127.1, 127.0, 126.4, 126.0, 125.0, 116.7, 45.9.

3-(4-phenyl-1-(pyridin-3-ylmethyl)-1*H*-imidazol-2-yl)pyridine (3o)



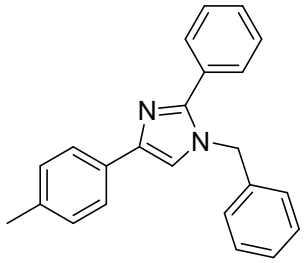
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (1/1) to afford a yellow oil in 35% yield (21.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.86 (s, 1H), 8.69 (s, 1H), 8.60 (s, 1H), 8.48 (s, 1H), 7.95 (d, J = 7.9 Hz, 1H), 7.82 (d, J = 7.4 Hz, 2H), 7.39 (t, J = 7.5 Hz, 4H), 7.30 (d, J = 6.7 Hz, 2H), 7.27 (s, 1H), 5.28 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 150.1, 149.7, 149.2, 148.2, 145.4, 142.6, 136.4, 134.2, 133.3, 131.8, 128.6, 127.2, 126.5, 124.9, 123.9, 123.5, 117.2, 48.3. HRMS (EI) m/z: $[M]^+$ calcd for $C_{20}H_{16}N_4$ 312.1375; found 312.1372.

2-(tert-butyl)-1-neopentyl-4-phenyl-1*H*-imidazole (3p)



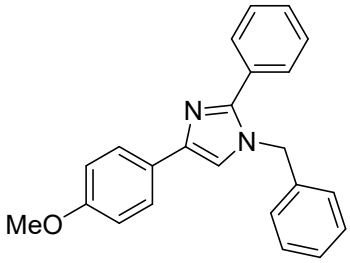
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow oil in 63% yield (34.0 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.82 (d, J = 7.4 Hz, 2H), 7.38 (t, J = 7.3 Hz, 2H), 7.21 (d, J = 7.1 Hz, 2H), 3.96 (s, 2H), 1.53 (s, 9H), 1.12 (s, 9H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 154.4, 137.7, 134.1, 127.7, 125.4, 123.9, 115.8, 57.0, 33.1, 32.2, 30.1, 28.0. HRMS (EI) m/z: $[M]^+$ calcd for $C_{18}H_{26}N_2$ 270.2096; found 270.2094.

1-benzyl-2-phenyl-4-(p-tolyl)-1*H*-imidazole (3q)[5]



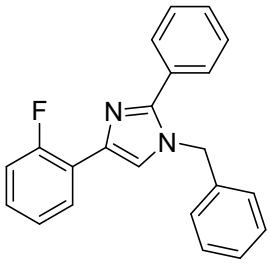
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 91% yield (59.0 mg). mp 138-140 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.73 (d, J = 7.6 Hz, 2H), 7.68 – 7.58 (m, 2H), 7.42 (d, J = 5.1 Hz, 3H), 7.35 (m, 3H), 7.20 (d, J = 9.3 Hz, 2H), 7.18 – 7.08 (m, 3H), 5.22 (s, 2H), 2.36 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 148.4, 141.5, 136.8, 136.3, 131.2, 130.4, 129.1, 128.9, 128.8, 128.6, 128.5, 127.8, 126.6, 124.7, 116.3, 50.4, 21.1.

1-benzyl-4-(4-methoxyphenyl)-2-phenyl-1*H*-imidazole (3r)[5]



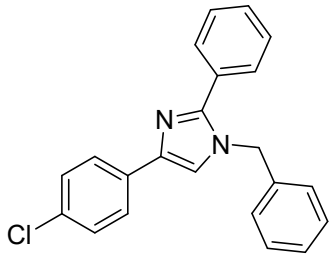
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 66% yield (44.9 mg). mp 128-130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.74 (d, J = 7.9 Hz, 2H), 7.58 (s, 2H), 7.37 (s, 3H), 7.30 (t, J = 8.2 Hz, 3H), 7.10 (d, J = 7.6 Hz, 3H), 6.89 (d, J = 7.9 Hz, 2H), 5.16 (s, 2H), 3.78 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 157.6, 147.2, 140.3, 135.8, 129.4, 127.9, 127.8, 127.5, 126.8, 125.9, 125.6, 125.1, 114.7, 112.8, 54.2, 49.4.

1-benzyl-4-(2-fluorophenyl)-2-phenyl-1*H*-imidazole (3s)[6]



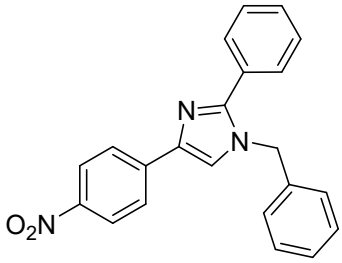
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 90% yield (59.1 mg). mp 68–69 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.29 (t, J = 8.8 Hz, 1H), 7.64 – 7.58 (m, 2H), 7.48 (d, J = 3.9 Hz, 1H), 7.45 – 7.39 (m, 3H), 7.33 (m, 3H), 7.23 – 7.18 (m, 2H), 7.13 (d, J = 7.3 Hz, 3H), 5.25 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 160.9, 158.4, 148.0, 136.8, 135.0, 130.3, 129.1, 129.0 (d, J = 2.9 Hz), 128.6, 127.9, 127.7 (d, J = 4.2 Hz), 127.5 (d, J = 8.4 Hz), 126.5, 124.3 (d, J = 3.2 Hz), 121.8 (d, J = 12.7 Hz), 121.1 (d, J = 14.5 Hz), 115.3 (d, J = 21.9 Hz), 50.5. ^{19}F NMR (376 MHz, $CDCl_3$): δ -114.6.

1-benzyl-4-(4-chlorophenyl)-2-phenyl-1*H*-imidazole (3t)[5]



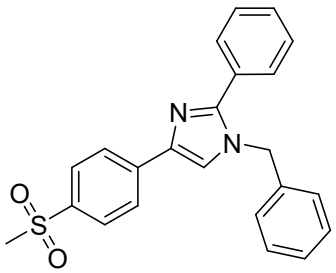
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 85% yield (58.5 mg). mp 126-128 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.76 (d, J = 8.3 Hz, 2H), 7.65 – 7.54 (m, 2H), 7.44 – 7.40 (m, 3H), 7.34 (m, 5H), 7.24 (d, J = 11.3 Hz, 1H), 7.13 (d, J = 7.2 Hz, 2H), 5.21 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 148.8, 140.4, 136.6, 132.6, 132.3, 130.2, 129.1, 129.0, 128.8, 128.6, 128.3, 128.0, 126.7, 126.1, 116.9, 50.5.

1-benzyl-4-(4-nitrophenyl)-2-phenyl-1*H*-imidazole (3u)[5]



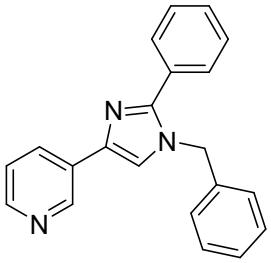
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a yellow solid in 91% yield (64.6 mg). mp 118-119 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.22 (d, J = 7.7 Hz, 2H), 7.96 (d, J = 7.8 Hz, 2H), 7.66 – 7.59 (m, 2H), 7.46 (s, 3H), 7.41 (s, 1H), 7.37 (d, J = 7.8 Hz, 2H), 7.26 (s, 1H), 7.15 (d, J = 7.0 Hz, 2H), 5.25 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 149.6, 146.3, 140.5, 139.4, 136.2, 129.9, 129.5, 129.2, 129.0, 128.8, 128.3, 126.8, 125.1, 124.1, 119.1, 50.8.

1-benzyl-4-(4-(methylsulfonyl)phenyl)-2-phenyl-1*H*-imidazole (3v)



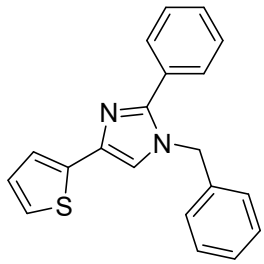
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a yellow solid in 88% yield (68.3 mg). mp 110-111 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.01 (d, J = 7.7 Hz, 2H), 7.91 (d, J = 7.9 Hz, 2H), 7.64 – 7.57 (m, 2H), 7.45 (s, 3H), 7.37 (d, J = 12.9 Hz, 3H), 7.26 (s, 1H), 7.14 (d, J = 7.0 Hz, 1H), 5.24 (s, 2H), 3.05 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 149.4, 139.6, 139.5, 138.0, 136.3, 130.0, 129.4, 129.1, 129.0, 128.7, 128.2, 127.7, 126.8, 125.3, 118.7, 50.7, 44.6. HRMS (EI) m/z: $[M]^+$ calcd for $C_{23}H_{20}N_2O_2S$ 388.1245; found 388.1241.

(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)pyridine (3w)



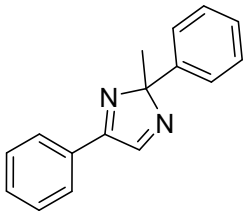
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (5/1) to afford a yellow oil in 86% yield (53.5 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.99 (s, 1H), 8.44 (d, J = 3.5 Hz, 1H), 8.15 (d, J = 7.8 Hz, 1H), 7.63 – 7.51 (m, 2H), 7.44 – 7.39 (m, 3H), 7.31 (m, 5H), 7.12 (d, J = 7.1 Hz, 2H), 5.22 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 149.2, 147.7, 146.4, 138.4, 136.5, 132.2, 130.0, 129.2, 129.1, 129.0, 128.7, 128.1, 127.8, 127.5, 126.7, 117.3, 50.6. HRMS (EI) m/z: $[M]^+$ calcd for $C_{21}H_{17}N_3$ 311.1422; found 311.1421.

1-benzyl-2-phenyl-4-(thiophen-2-yl)-1*H*-imidazole (3x)[6]



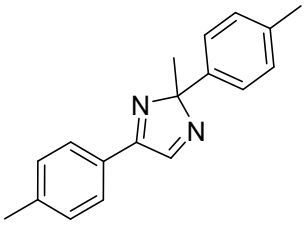
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a yellow solid in 74% yield (46.8 mg). mp 98-100 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.62 – 7.57 (m, 2H), 7.43 – 7.39 (m, 3H), 7.39 – 7.31 (m, 4H), 7.18 (d, J = 4.6 Hz, 1H), 7.13 (d, J = 9.1 Hz, 3H), 7.03 (s, 1H), 5.18 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 148.4, 137.8, 136.7, 136.6, 130.1, 129.0, 128.9, 128.5, 127.9, 127.4, 126.6, 124.9, 123.2, 122.0, 116.2, 50.4.

2-methyl-2,4-diphenyl-2*H*-imidazole (4a)[8]



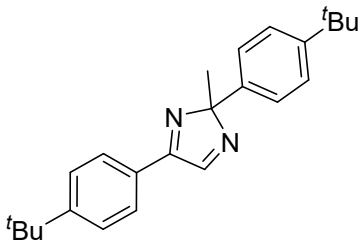
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a blue solid in 37% yield (17.3 mg). mp 84-86 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.46 (s, 1H), 8.04 (d, J = 6.9 Hz, 2H), 7.71 (d, J = 7.6 Hz, 2H), 7.49 (d, J = 5.0 Hz, 3H), 7.33 (t, J = 7.2 Hz, 2H), 7.30 – 7.21 (m, 1H), 1.84 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.7, 154.2, 140.1, 131.3, 131.0, 129.0, 128.2, 127.6, 126.9, 109.1, 27.0.

2-methyl-2,4-di-p-tolyl-2*H*-imidazole (4b)[8]



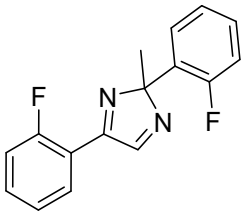
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a blue solid in 38% yield (19.9 mg). mp 78-79 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.4 (s, 1H), 7.9 (d, J = 8.0 Hz, 2H), 7.6 (d, J = 8.0 Hz, 2H), 7.3 (d, J = 7.9 Hz, 2H), 7.2 (d, J = 7.9 Hz, 2H), 2.4 (s, 3H), 2.3 (s, 3H), 1.8 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.5, 154.3, 141.8, 137.3, 137.2, 129.7, 128.9, 128.4, 128.2, 126.7, 108.8, 26.9, 21.5, 21.0.

2,4-bis(4-(tert-butyl)phenyl)-2-methyl-2*H*-imidazole (4c)



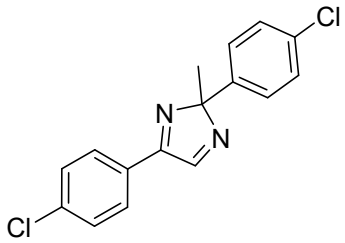
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a blue oil in 42% yield (29.1 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.50 (s, 1H), 8.04 (d, J = 8.1 Hz, 2H), 7.68 (d, J = 8.2 Hz, 2H), 7.58 (d, J = 8.0 Hz, 2H), 7.41 (d, J = 8.2 Hz, 2H), 1.90 (s, 3H), 1.42 (s, 9H), 1.36 (s, 9H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 162.6, 154.0, 153.5, 149.6, 136.2, 127.5, 127.3, 125.7, 125.1, 124.3, 108.0, 34.1, 33.6, 30.4, 30.3, 26.0. HRMS (EI) m/z: $[M]^+$ calcd for $C_{24}H_{30}N_2$ 346.2409; found 346.2406.

2,4-bis(2-fluorophenyl)-2-methyl-2*H*-imidazole (4d)



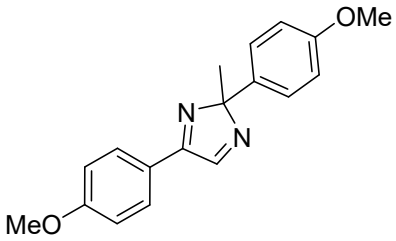
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a green oil in 44% yield (23.8 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.49 (d, J = 4.7 Hz, 1H), 8.20 (t, J = 7.5 Hz, 1H), 7.51 (dt, J = 13.5, 6.8 Hz, 2H), 7.28 (t, J = 6.1 Hz, 2H), 7.19 (m, J = 11.1, 8.3 Hz, 1H), 7.15 – 6.99 (m, 2H), 1.91 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 162.3 (d, J = 72.0 Hz), 161.1, 159.8 (d, J = 70.5 Hz), 156.3 (d, J = 11.7 Hz), 133.1 (d, J = 8.7 Hz), 130.8, 129.7, 128.5 (d, J = 3.7 Hz), 126.1, 124.7 (d, J = 3.4 Hz), 123.8 (d, J = 3.5 Hz), 118.9, 116.3 (d, J = 11.7 Hz), 116.1 (d, J = 10.9 Hz), 105.4 (d, J = 4.6 Hz), 23.7. ^{19}F NMR (372 MHz, $CDCl_3$) δ -110.6, -112.8. HRMS (EI) m/z: $[M]^+$ calcd for $C_{16}H_{12}F_2N_2$ 270.0969; found 270.0966.

2,4-bis(4-chlorophenyl)-2-methyl-2*H*-imidazole (4e)



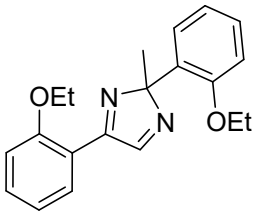
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (10/1) to afford a yellow oil in 45% yield (27.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.42 (s, 1H), 7.98 (d, J = 8.6 Hz, 2H), 7.64 (d, J = 8.5 Hz, 2H), 7.48 (d, J = 8.6 Hz, 2H), 7.30 (d, J = 8.6 Hz, 2H), 1.79 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 162.9, 154.1, 138.5, 137.8, 133.6, 129.6, 129.4, 129.3, 128.4, 128.3, 108.8, 27.1. HRMS (EI) m/z: $[M]^+$ calcd for $C_{16}H_{12}Cl_2N_2$ 302.0378; found 302.0376.

2,4-bis(4-methoxyphenyl)-2-methyl-2*H*-imidazole (4f)



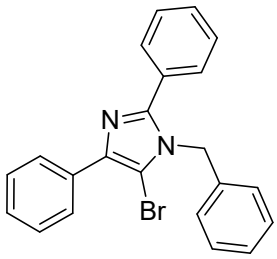
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (20/1) to afford a light green oil in 36% yield (21.2 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.43 (s, 1H), 8.00 (d, J = 8.5 Hz, 2H), 7.60 (d, J = 8.9 Hz, 2H), 7.00 (d, J = 6.6 Hz, 2H), 6.86 (d, J = 8.4 Hz, 2H), 3.86 (s, 3H), 3.77 (s, 3H), 1.81 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.0, 162.1, 159.0, 154.2, 132.5, 129.9, 127.9, 123.8, 114.4, 113.6, 108.5, 55.4, 55.2, 26.9. HRMS (EI) m/z: $[M]^+$ calcd for $C_{18}H_{18}N_2O_2$ 294.1368; found 294.1366.

2,4-bis(2-ethoxyphenyl)-2-methyl-2*H*-imidazole (4g)



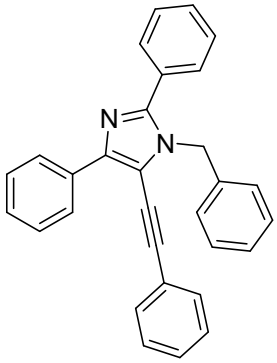
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ ethyl acetate (5/1) to afford a green oil in 55% yield (35.4 mg). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.63 (s, 1H), 8.14 (m, J = 7.7, 1.8 Hz, 1H), 7.55 (m, J = 8.1 Hz, 1H), 7.49 – 7.43 (m, 1H), 7.30 – 7.24 (m, 1H), 7.07 (m, J = 7.5 Hz, 1H), 7.00 (m, J = 8.5 Hz, 1H), 6.91 (t, J = 7.4 Hz, 2H), 4.18 (q, J = 7.0 Hz, 2H), 4.11 (q, J = 9.8 Hz, 2H), 1.95 (s, 3H), 1.50 (t, J = 7.0 Hz, 3H), 1.41 (t, J = 7.0 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 163.5, 157.7, 157.4, 157.2, 132.2, 131.0, 129.0, 128.2, 126.6, 120.9, 120.6, 120.1, 112.5, 111.9, 105.4, 63.9, 63.8, 23.3, 14.6, 14.5. HRMS (EI) m/z: $[M]^+$ calcd for $C_{20}H_{22}N_2O_2$ 322.1681; found 322.1680.

1-Benzyl-5-bromo-2,4-diphenyl-1*H*-imidazole (5)[1]



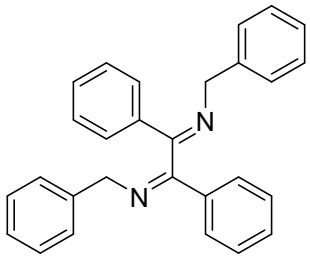
White solid, mp 106–108 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.18 (d, *J* = 7.3 Hz, 2H), 7.61 (dd, *J* = 7.3, 2.3 Hz, 2H), 7.48 (t, *J* = 7.6 Hz, 2H), 7.42 (m, *J* = 5.3 Hz, 3H), 7.39 – 7.30 (m, 4H), 7.09 (d, *J* = 6.9 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 149.0, 138.5, 136.2, 133.1, 130.3, 129.2, 128.8, 128.6, 128.5, 128.1, 127.5, 127.2, 126.7, 125.8, 101.5, 49.2.

1-Benzyl-5-bromo-2,4-diphenyl-1*H*-imidazole (6)



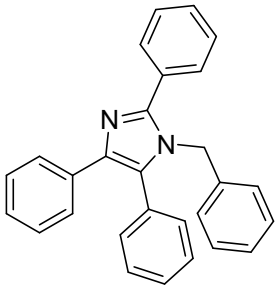
Light yellow solid, mp 128–130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.32 (d, *J* = 7.6 Hz, 2H), 7.66 – 7.59 (m, 2H), 7.50 – 7.40 (m, 5H), 7.32 (m, 9H), 7.19 (d, *J* = 7.1 Hz, 2H), 5.41 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 148.8, 144.3, 136.8, 133.7, 131.0, 130.0, 129.3, 128.9, 128.8, 128.6, 128.4, 128.3, 128.2, 127.6, 127.5, 126.3, 126.2, 122.6, 113.2, 99.2, 79.8, 49.0.

dibenzyl-1,2-diphenylethane-1,2-diimine (12)[9]



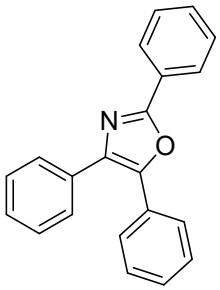
White solid, mp 97–98 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.90 (d, *J* = 6.8 Hz, 4H), 7.50 – 7.39 (m, 6H), 7.35 (m, 8H), 7.28 (t, *J* = 3.6 Hz, 2H), 4.69 – 4.55 (m, 4H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 165.89, 139.53, 135.74, 131.02, 128.82, 128.35, 127.89, 127.44, 126.80, 57.77.

1-benzyl-2,4,5-triphenyl-1*H*-imidazole (13)[10]



White solid, mp 128–130 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.69 – 7.61 (m, 2H), 7.58 (d, *J* = 7.5 Hz, 2H), 7.42 – 7.26 (m, 7H), 7.23 (d, *J* = 6.0 Hz, 2H), 7.21 – 7.19 (m, 2H), 7.18 (d, *J* = 2.0 Hz, 2H), 7.14 (d, *J* = 7.1 Hz, 1H), 6.79 (d, *J* = 5.2 Hz, 2H), 5.10 (s, 2H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 147.89, 137.90, 137.36, 134.29, 130.90, 130.86, 130.78, 129.87, 128.89, 128.65 (d, *J* = 11.2 Hz), 128.43, 128.40, 128.38, 127.88, 127.16, 126.61, 126.17, 125.83, 48.10.

2,4,5-triphenyloxazole (15)[11]



White solid, mp 111–113 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.21 – 8.16 (m, 2H), 7.79 – 7.73 (m, 2H), 7.73 – 7.67 (m, 2H), 7.54 – 7.47 (m, 33H), 7.47 – 7.34 (m, 6H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 160.10, 145.51, 136.75, 132.56, 130.30, 129.85, 128.95, 128.71, 128.64, 128.57, 128.51, 128.19, 128.11, 127.36, 126.52, 126.42.

8. References

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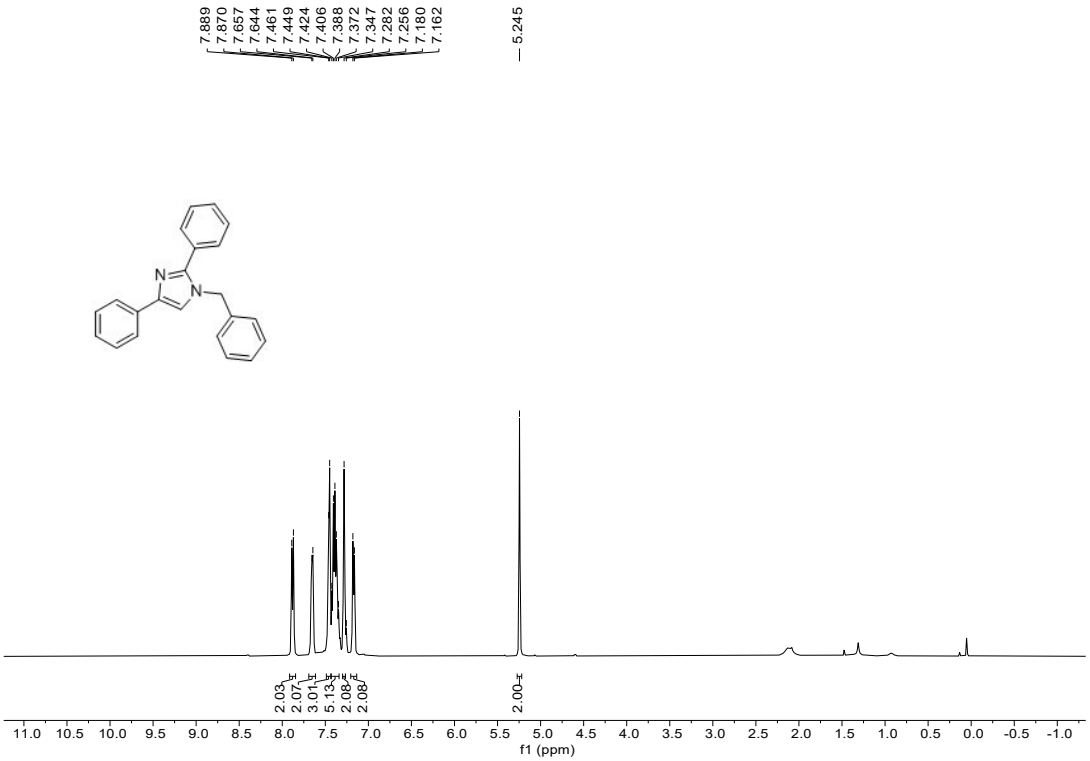
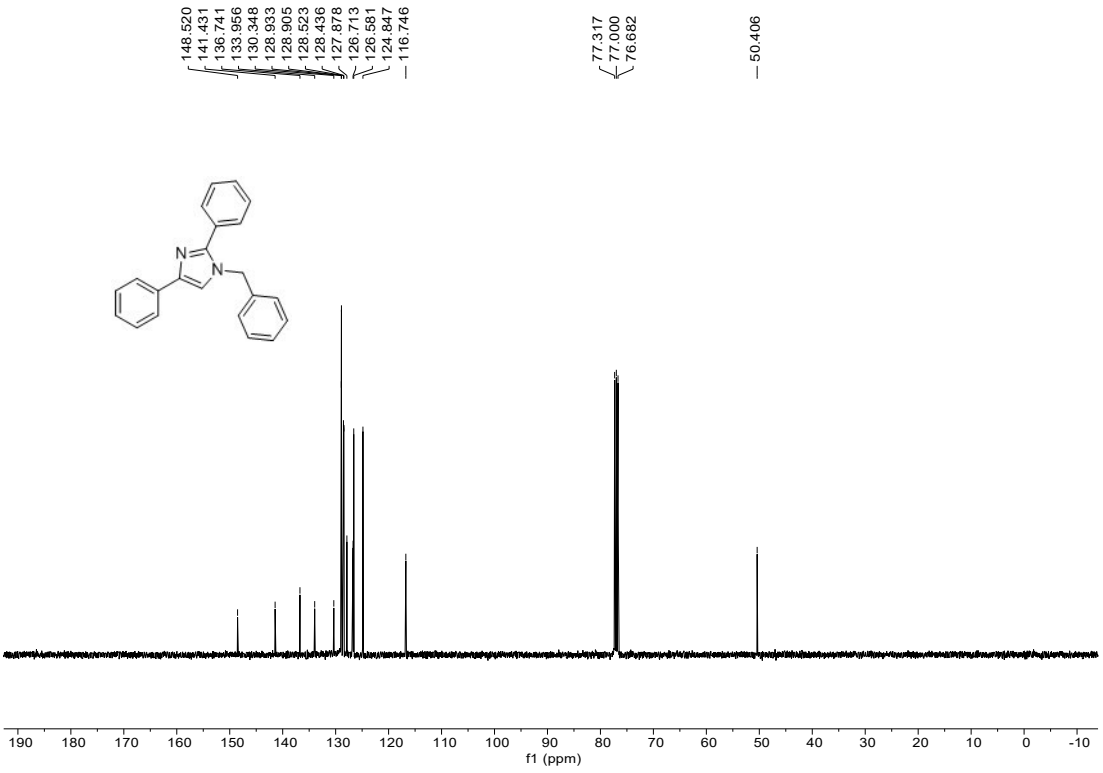
9. Copies of ^{1}H, ^{13}C and ^{19}F NMR Charts of the Imidazoles

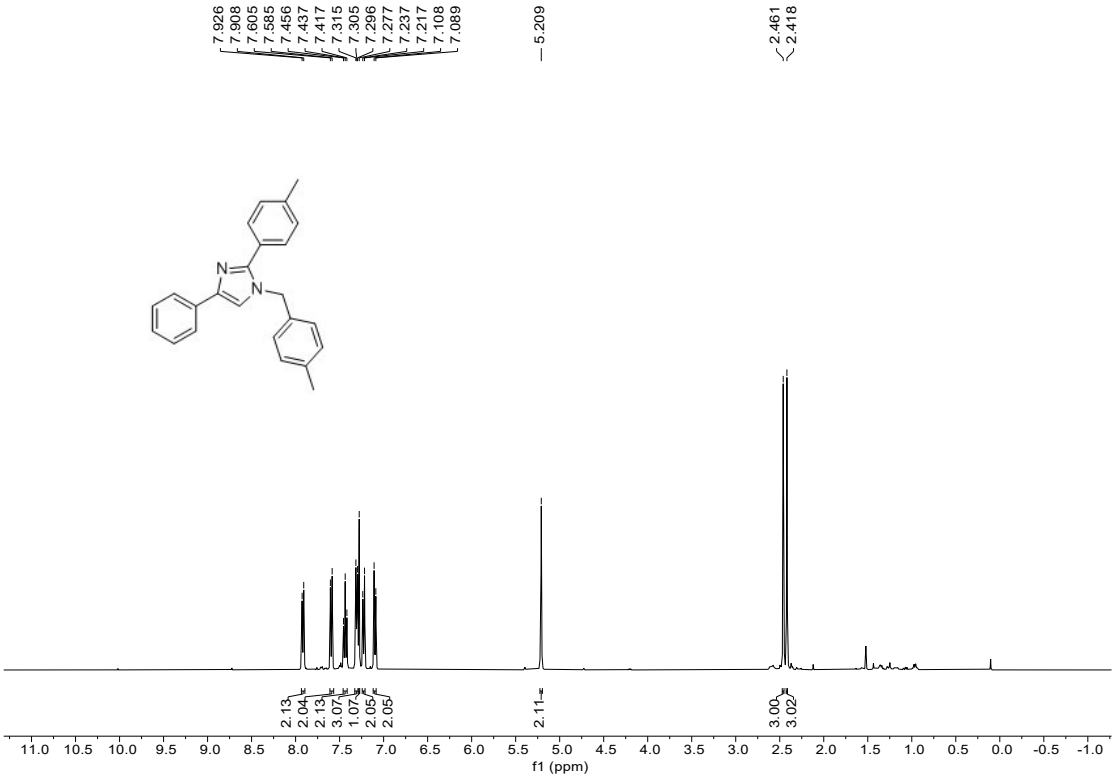
^{1}H NMR Spectrum of **3a**



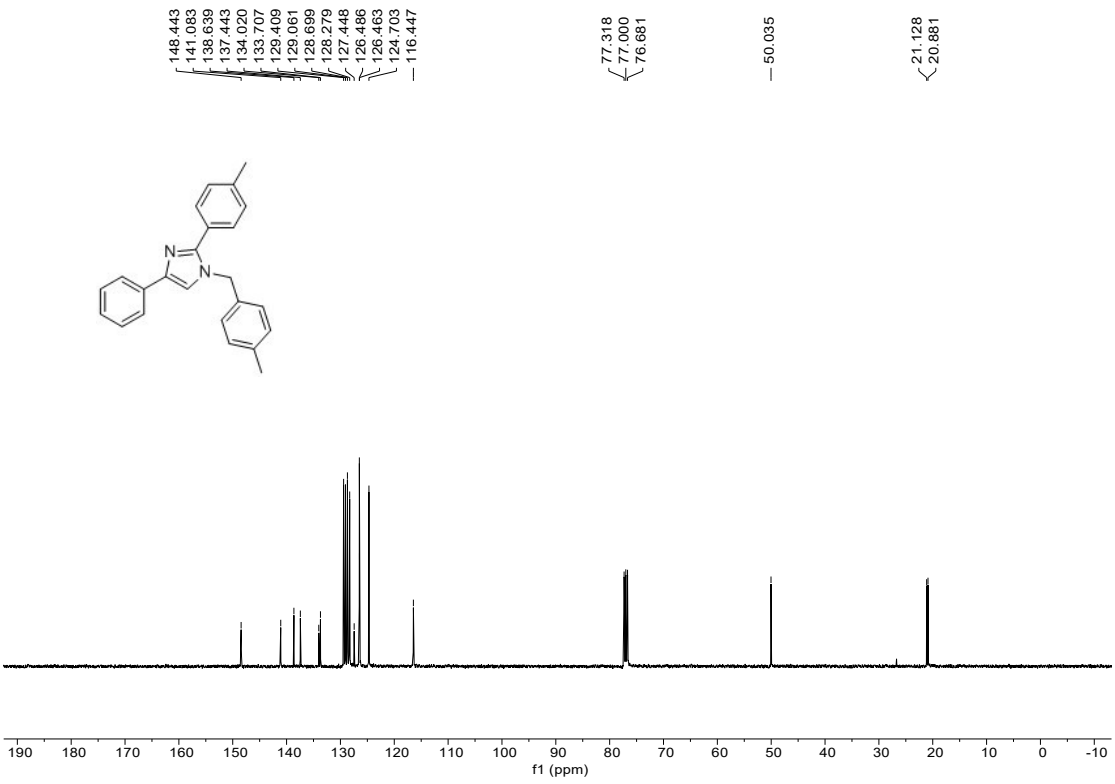
^{13}C NMR Spectrum of **3a**



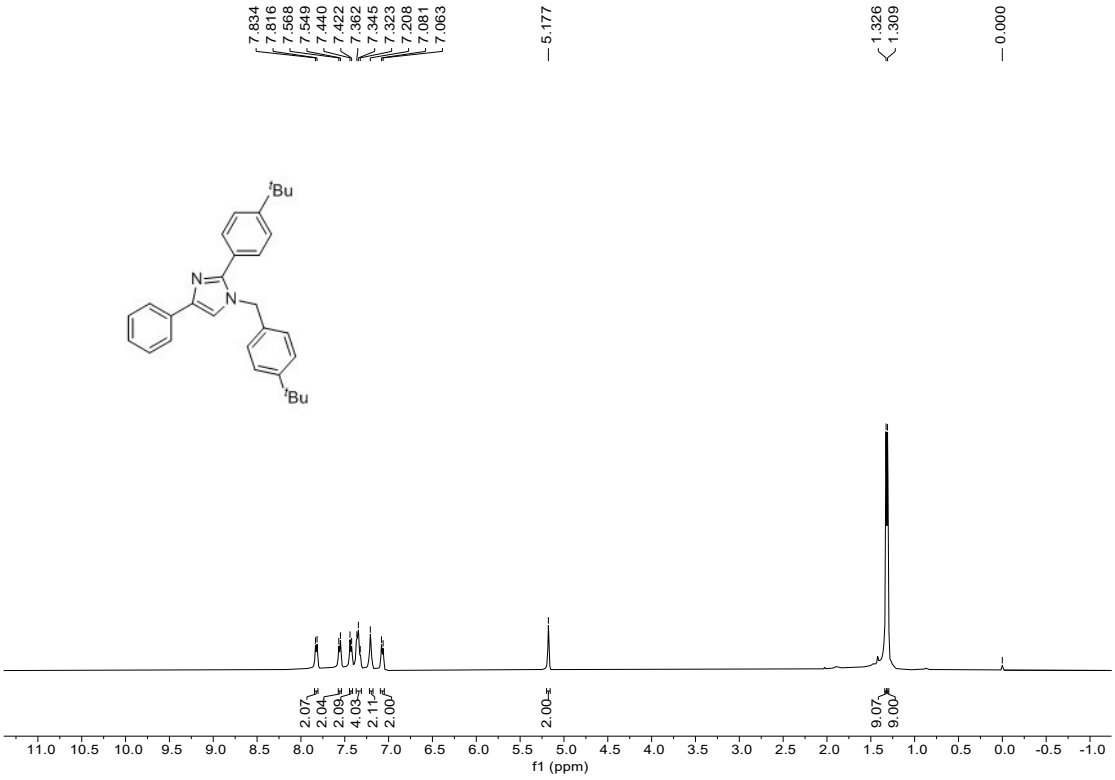
^{1}H NMR Spectrum of **3b**



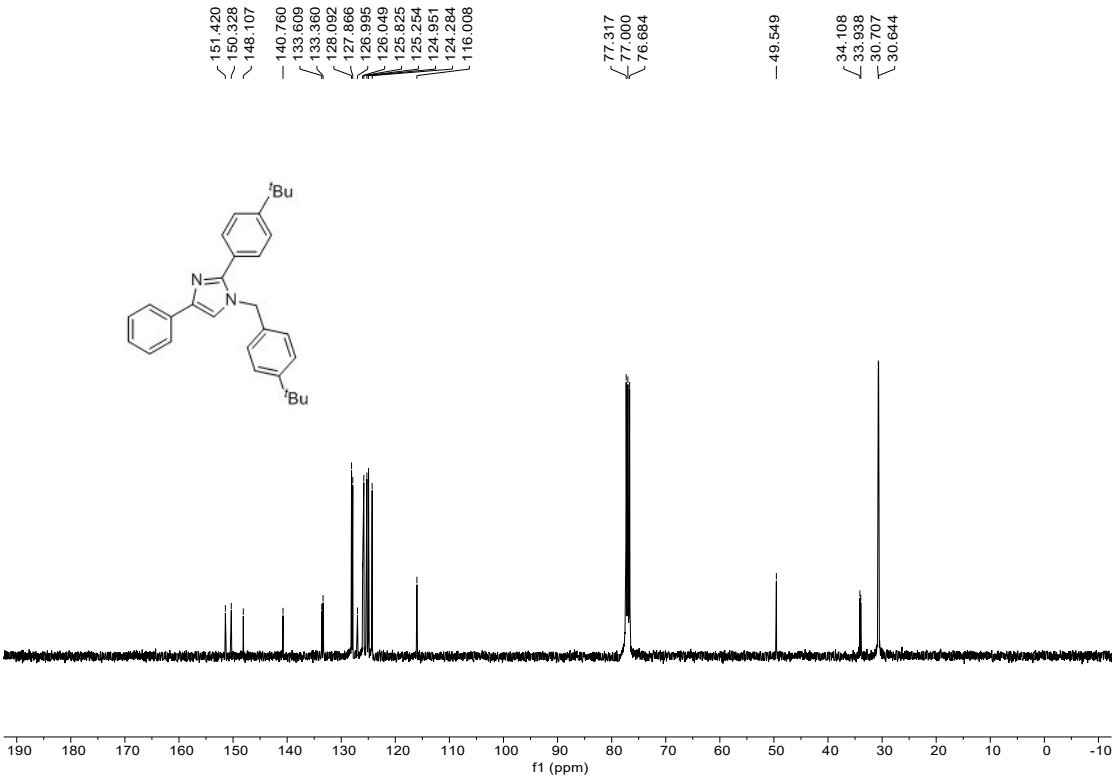
^{13}C NMR Spectrum of **3b**



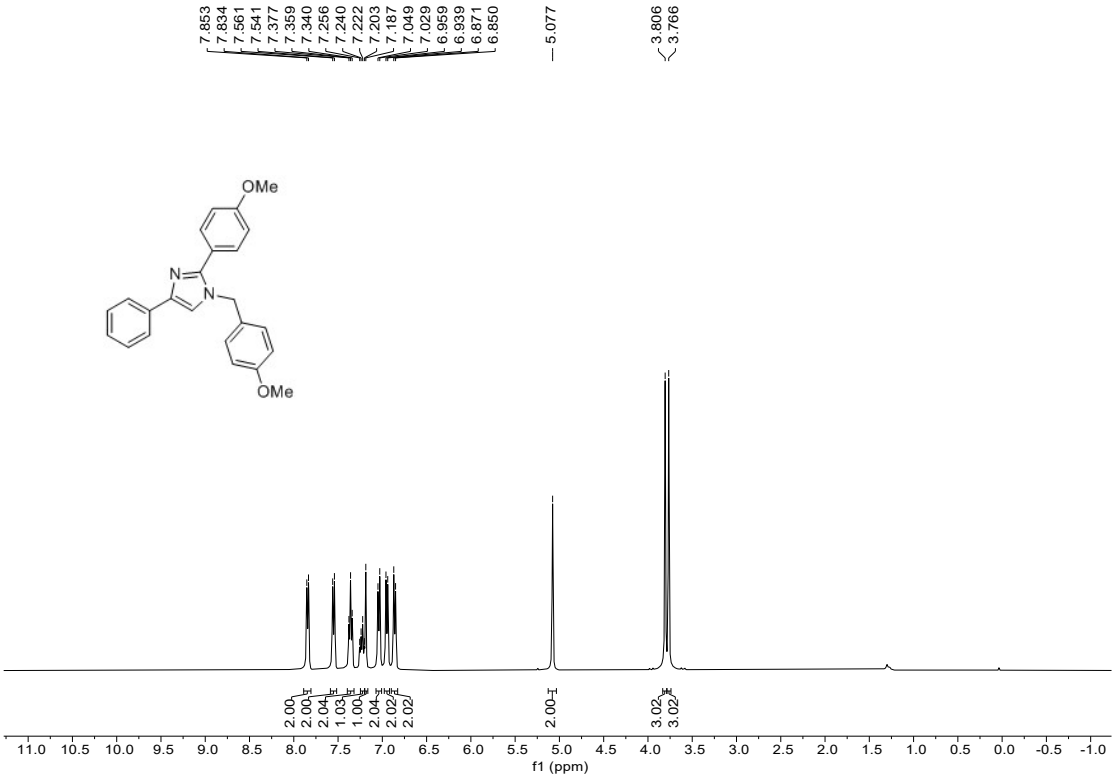
^{1}H NMR Spectrum of **3c**



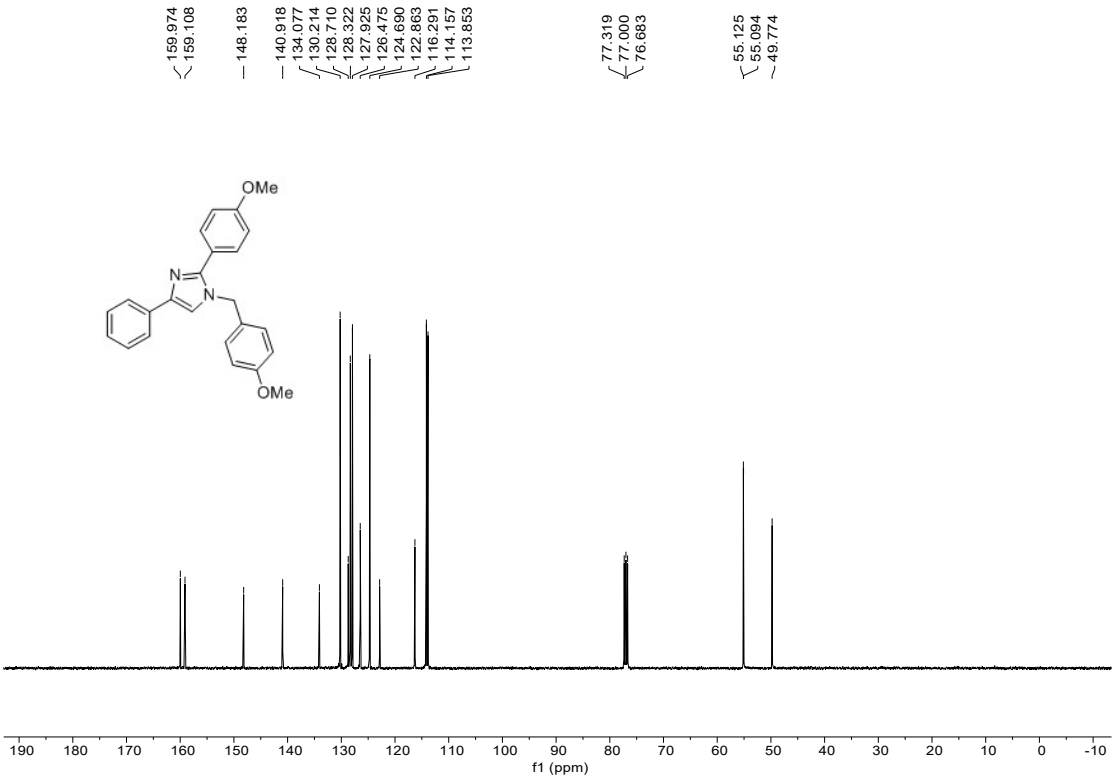
^{13}C NMR Spectrum of **3c**



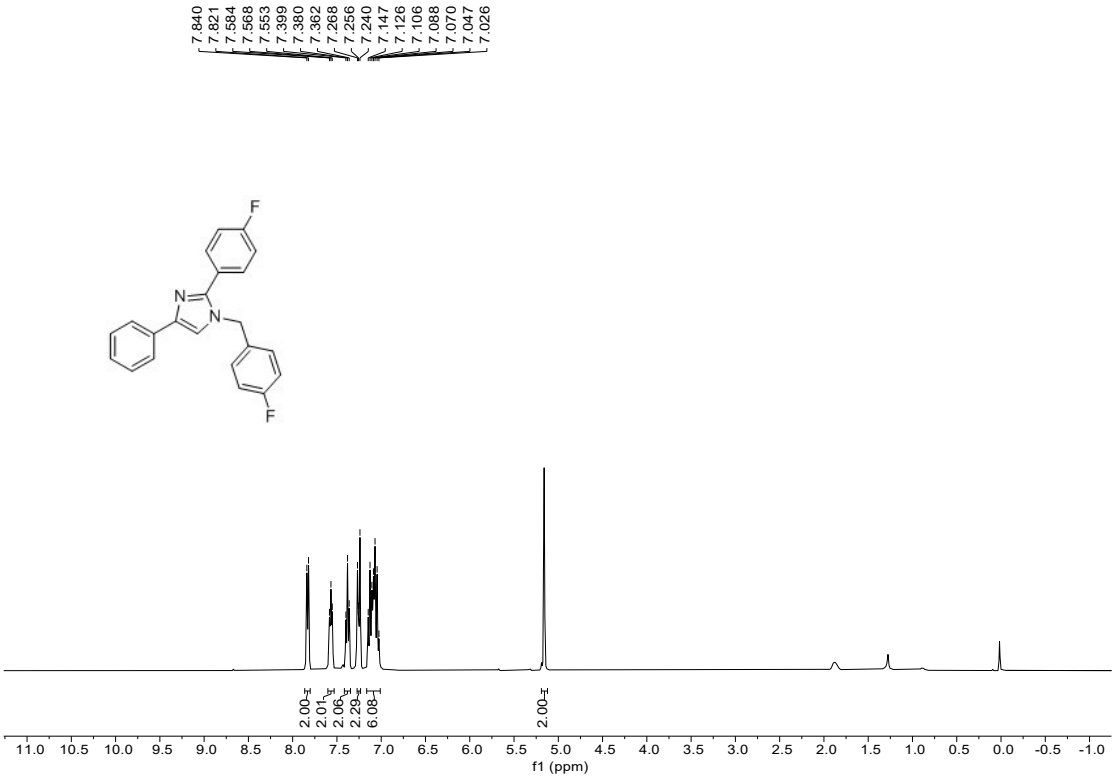
^{1}H NMR Spectrum of **3d**



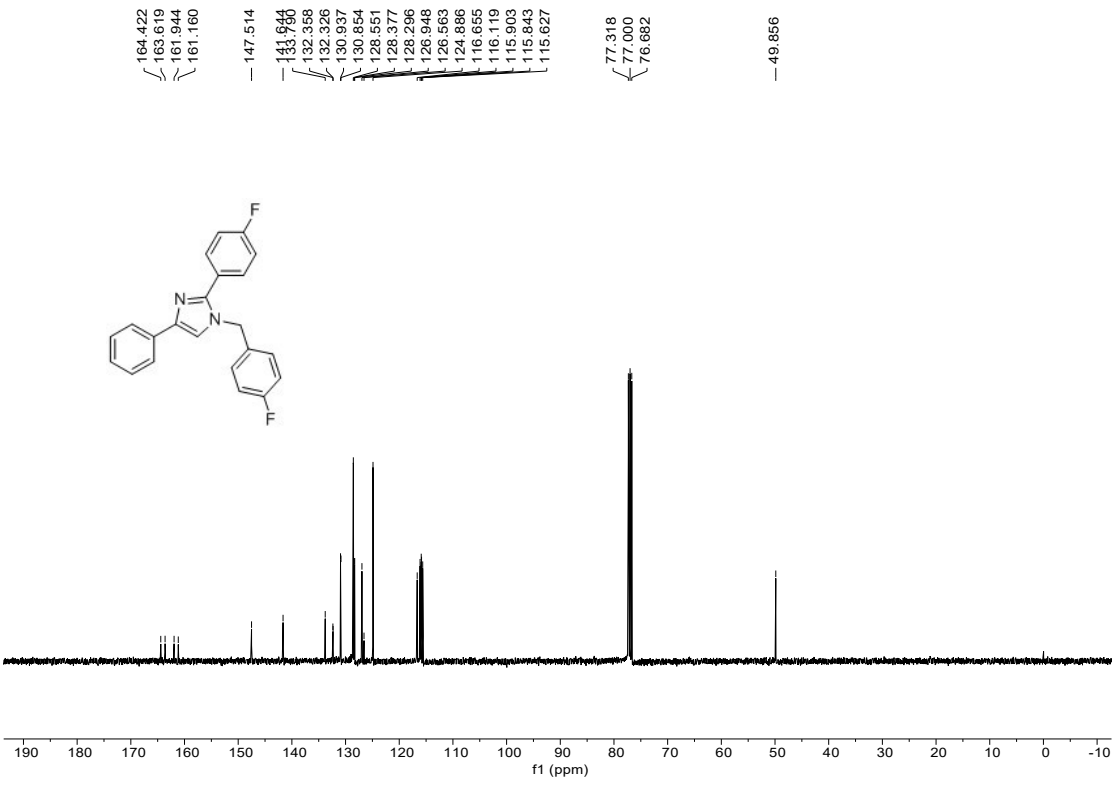
^{13}C NMR Spectrum of **3d**



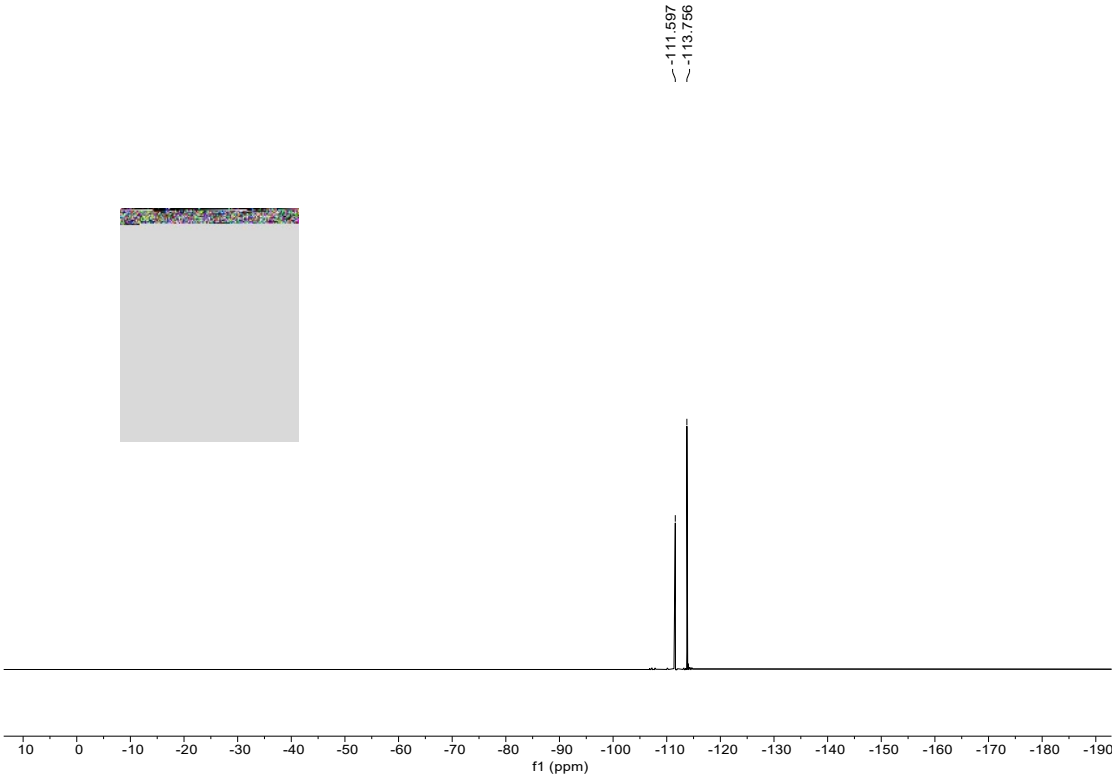
^{1}H NMR Spectrum of **3e**



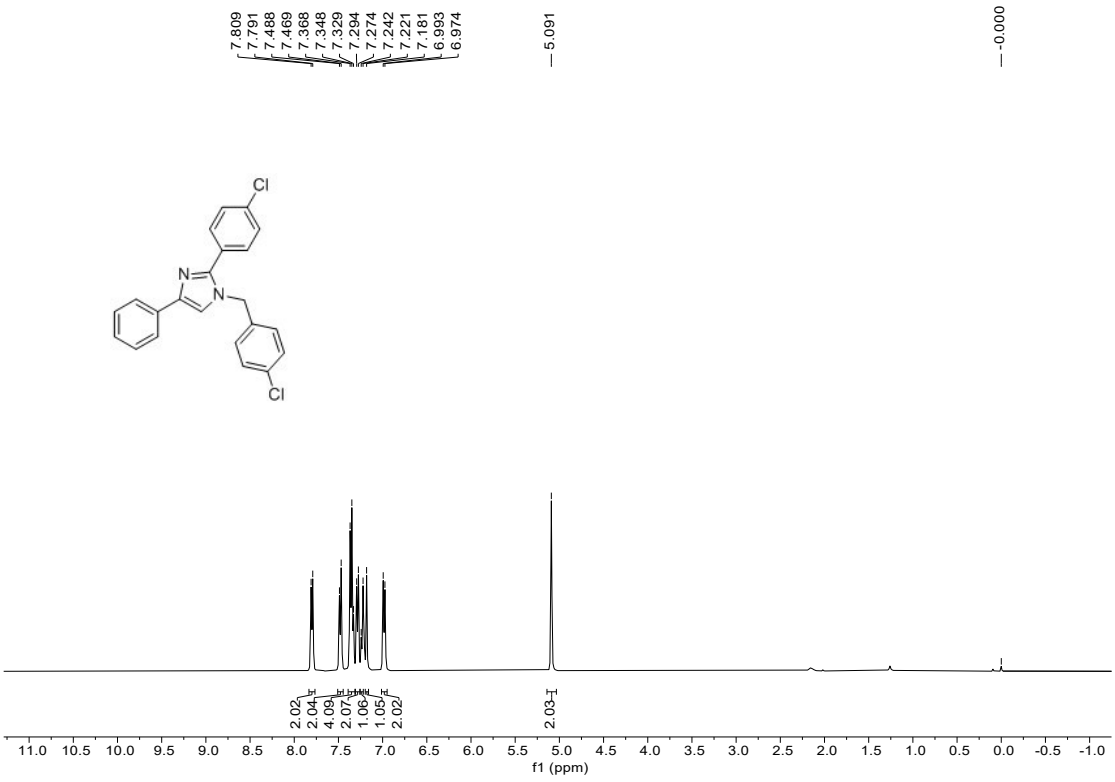
^{13}C NMR Spectrum of **3e**



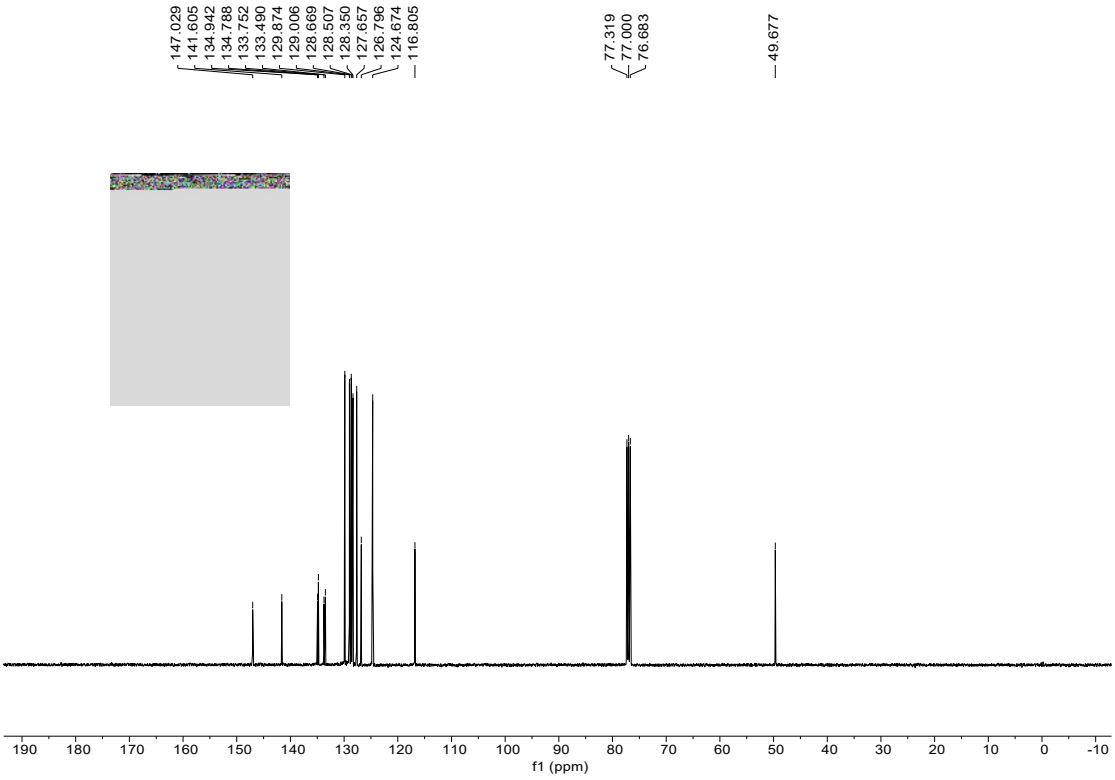
^{19}F NMR Spectrum of **3e**



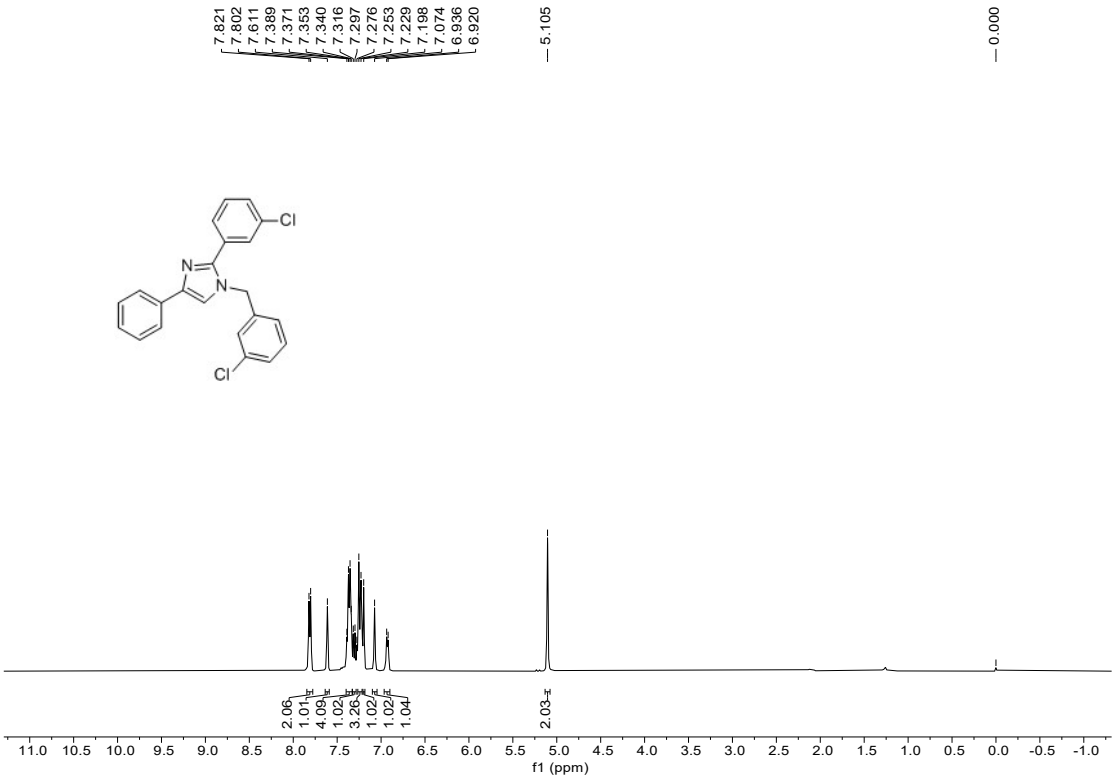
^{1}H NMR Spectrum of **3f**



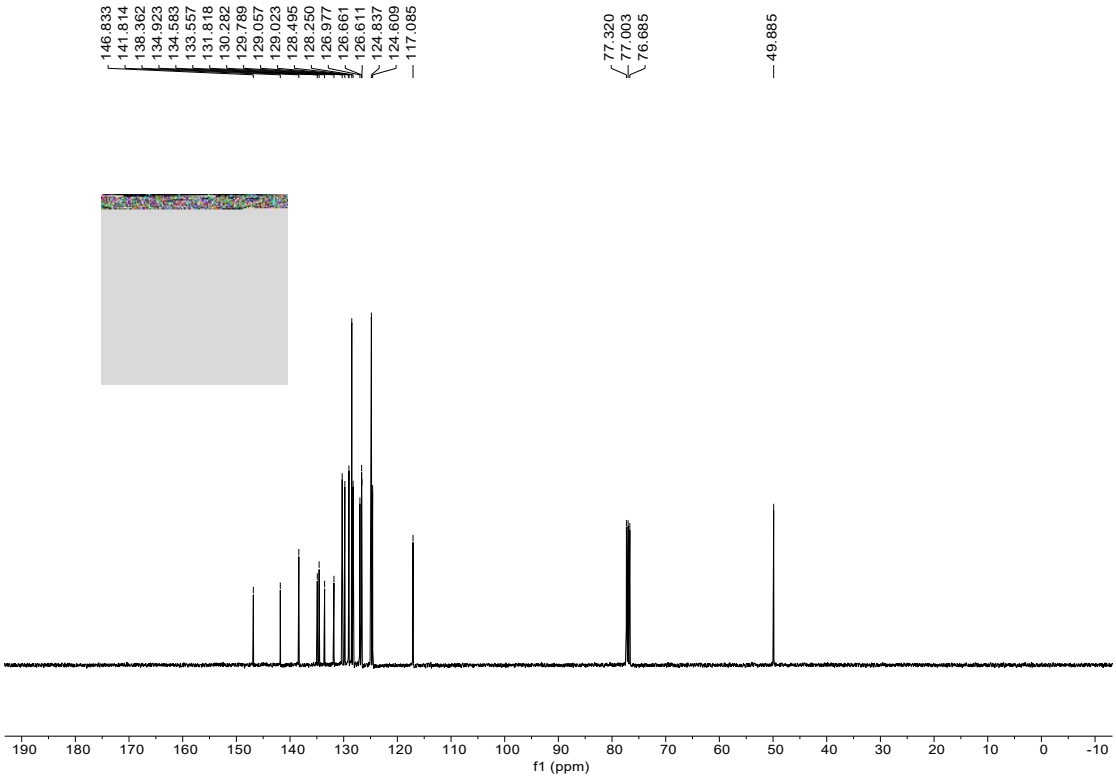
^{13}C NMR Spectrum of **3f**



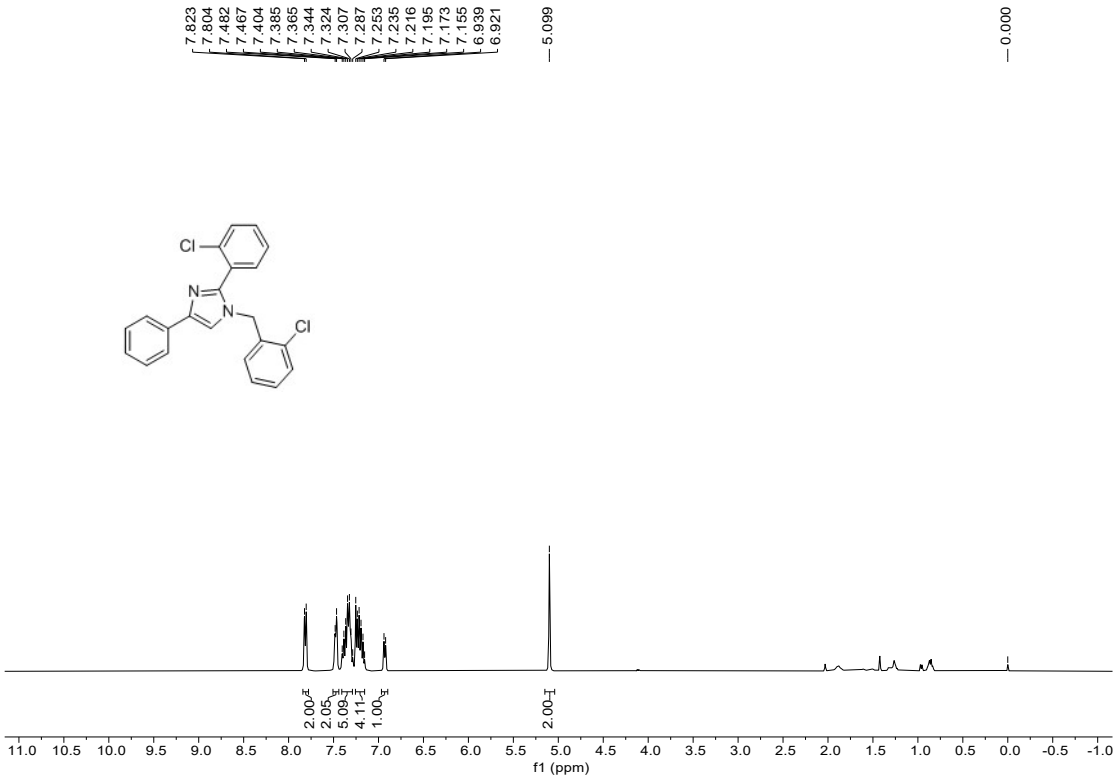
^{1}H NMR Spectrum of **3g**



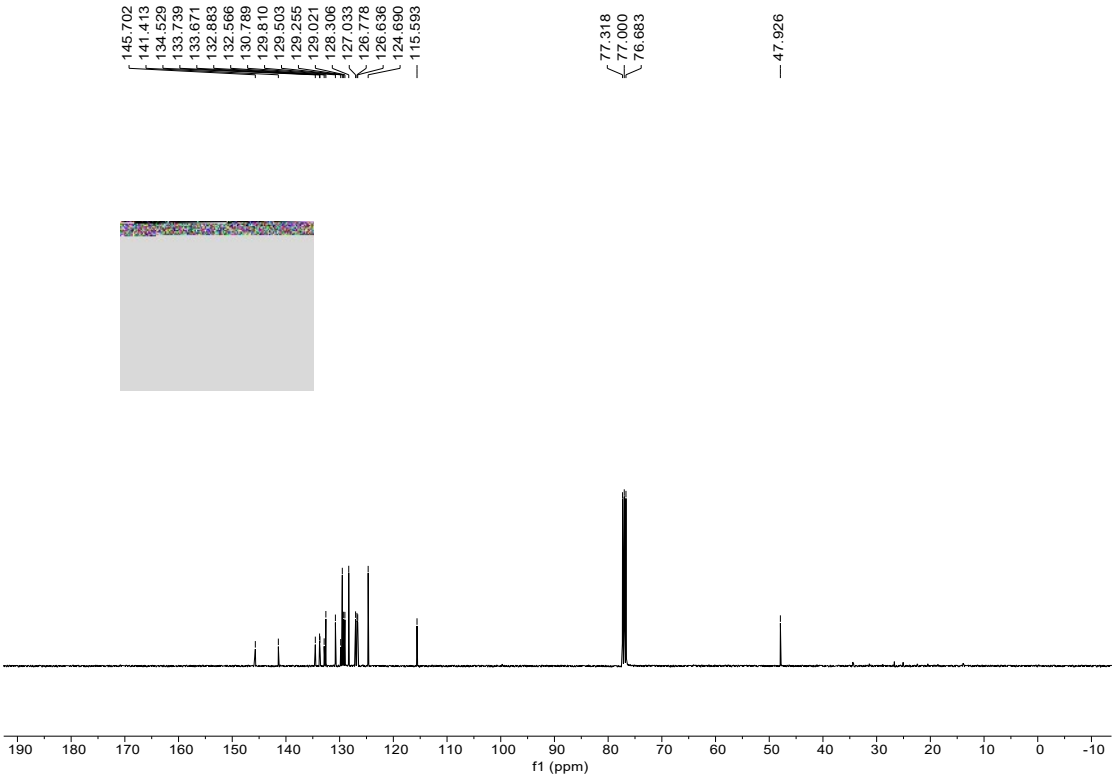
^{13}C NMR Spectrum of **3g**



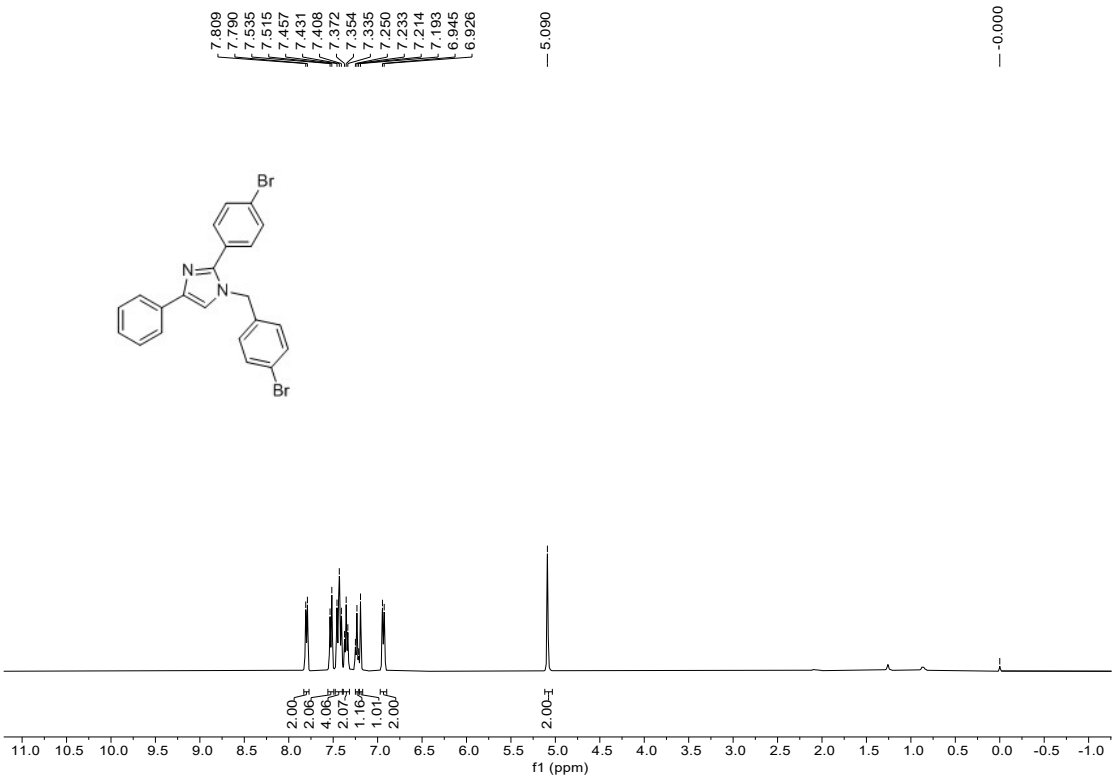
^{1}H NMR Spectrum of **3h**



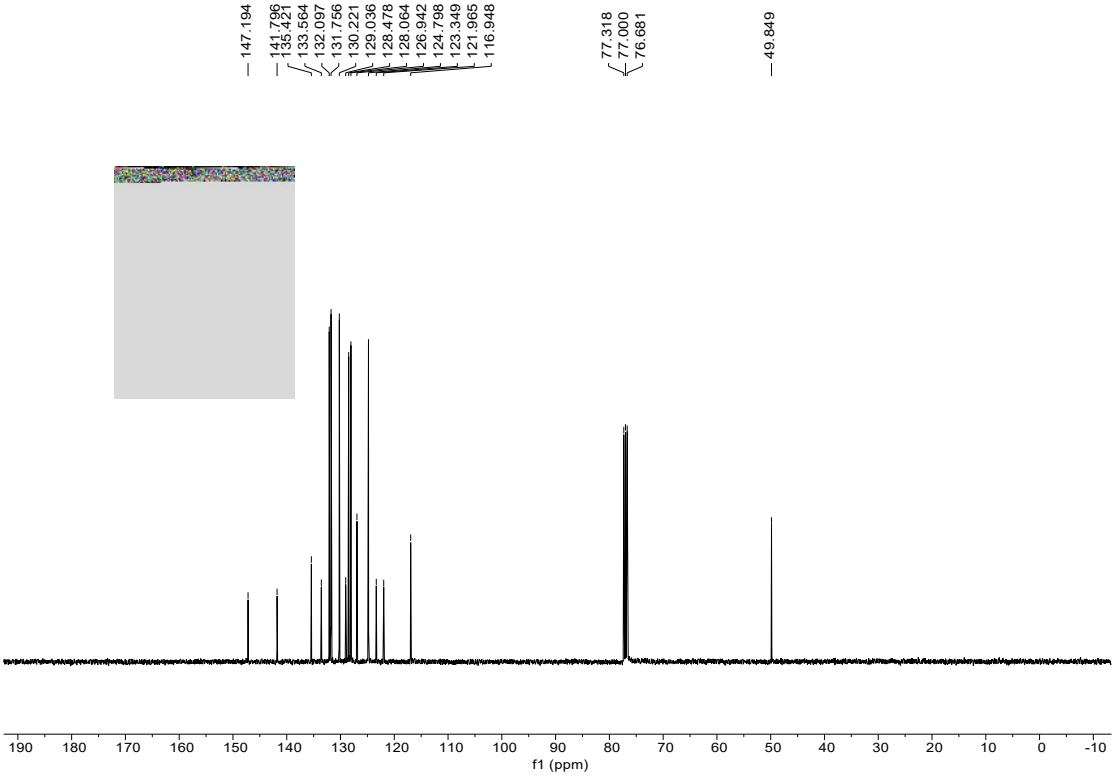
^{13}C NMR Spectrum of **3h**



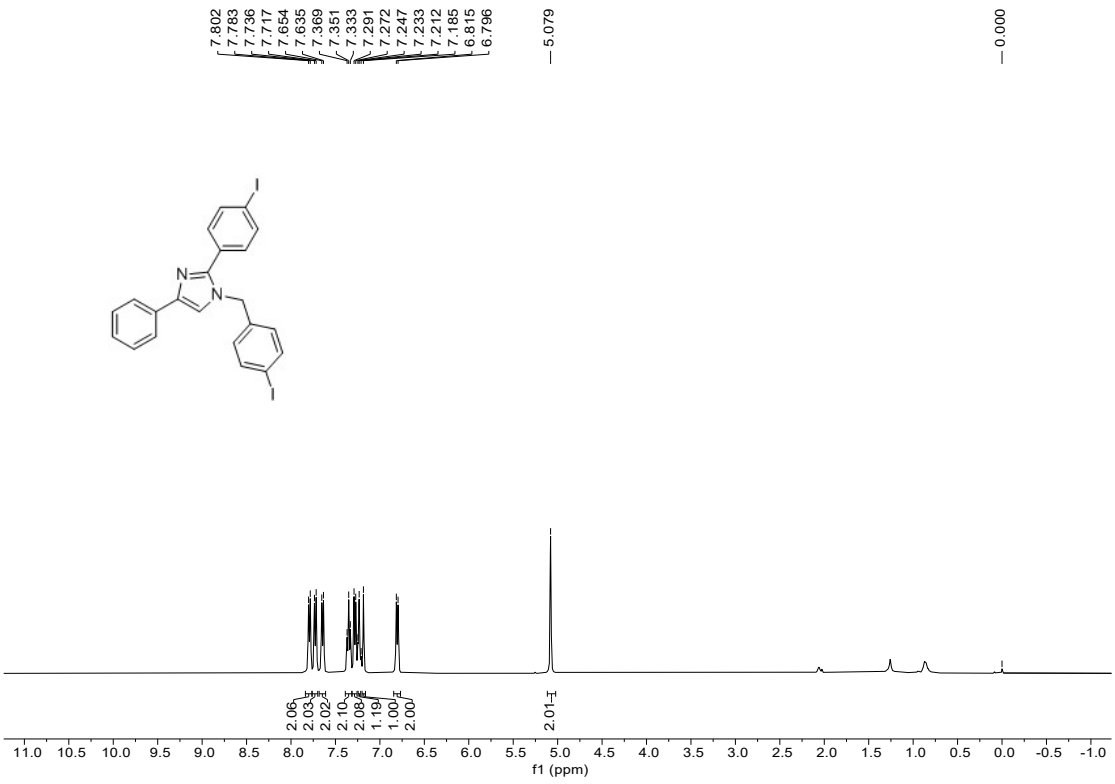
^{1}H NMR Spectrum of **3i**



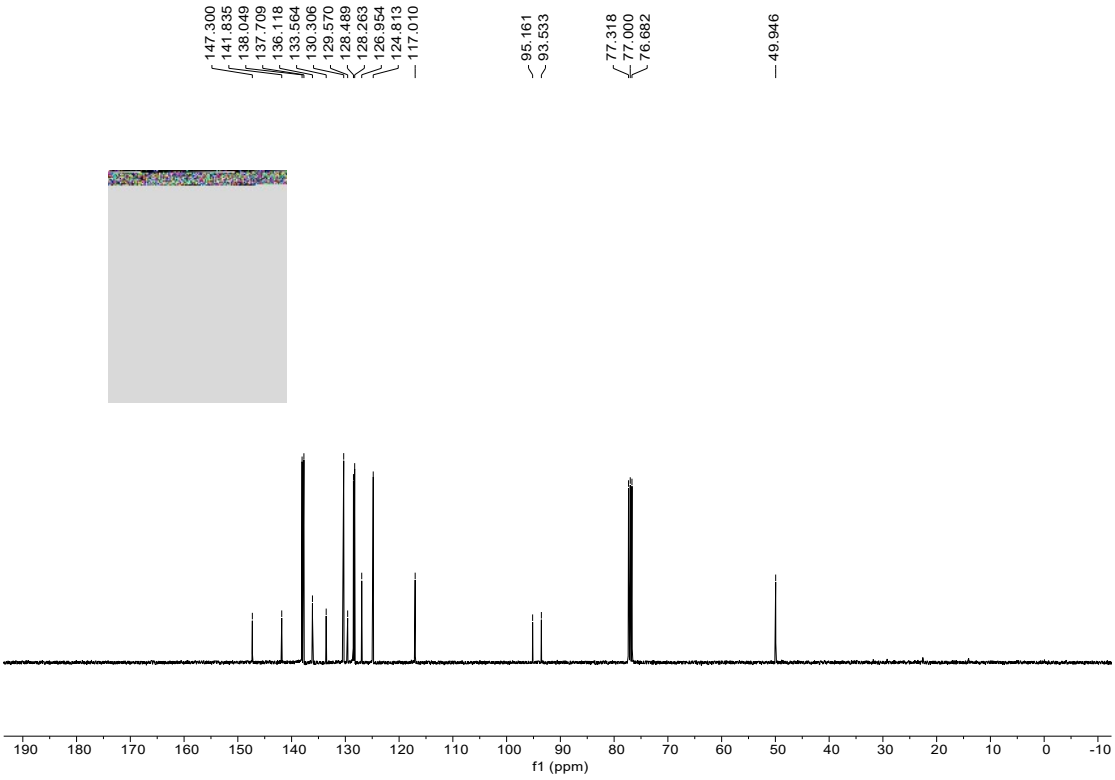
^{13}C NMR Spectrum of **3i**



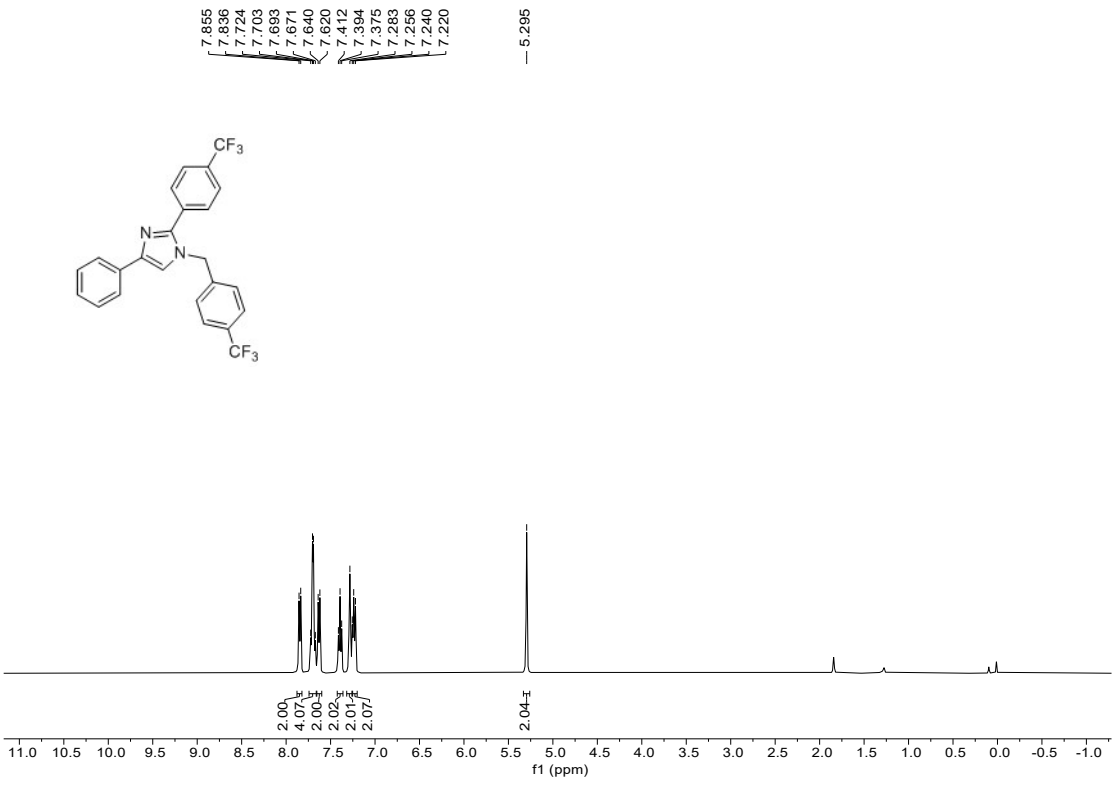
^{1}H NMR Spectrum of **3j**



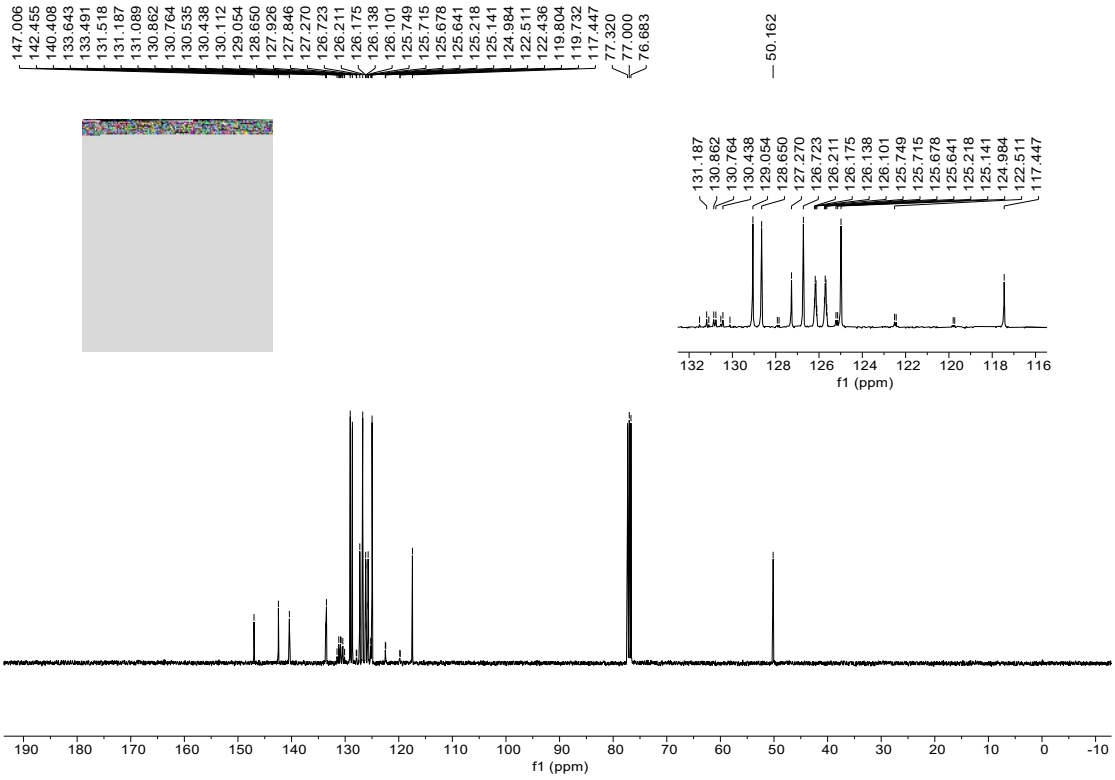
^{13}C NMR Spectrum of **3j**



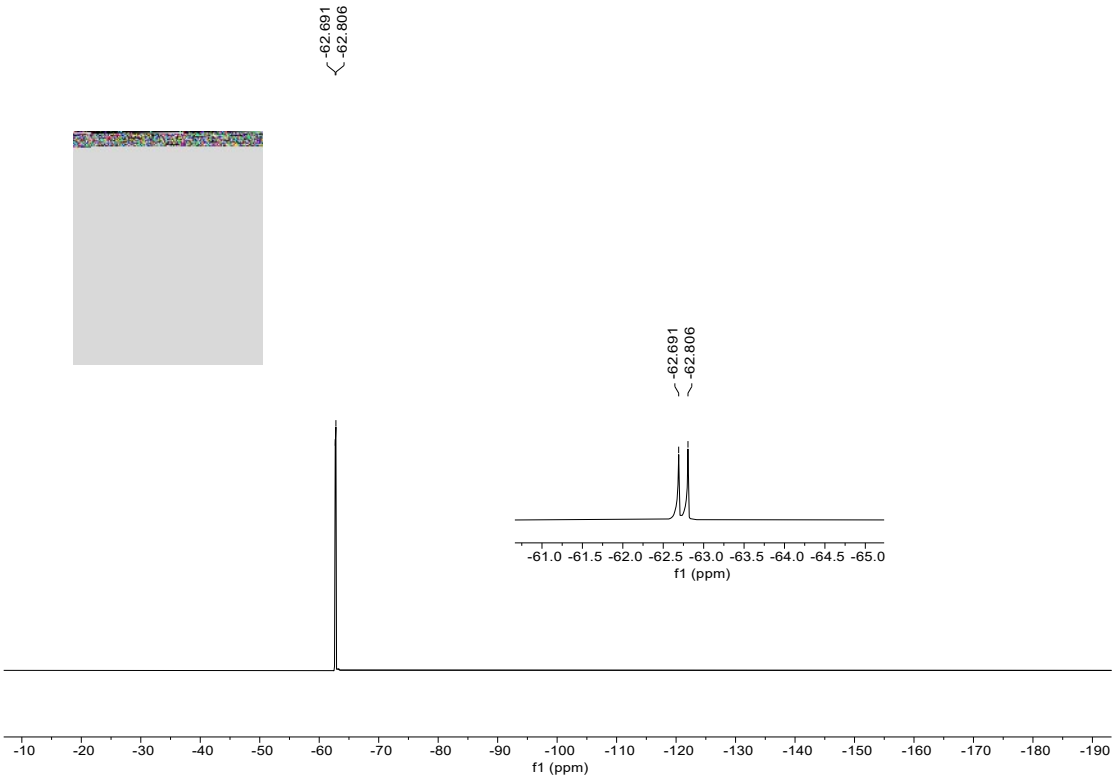
^{1}H NMR Spectrum of **3k**



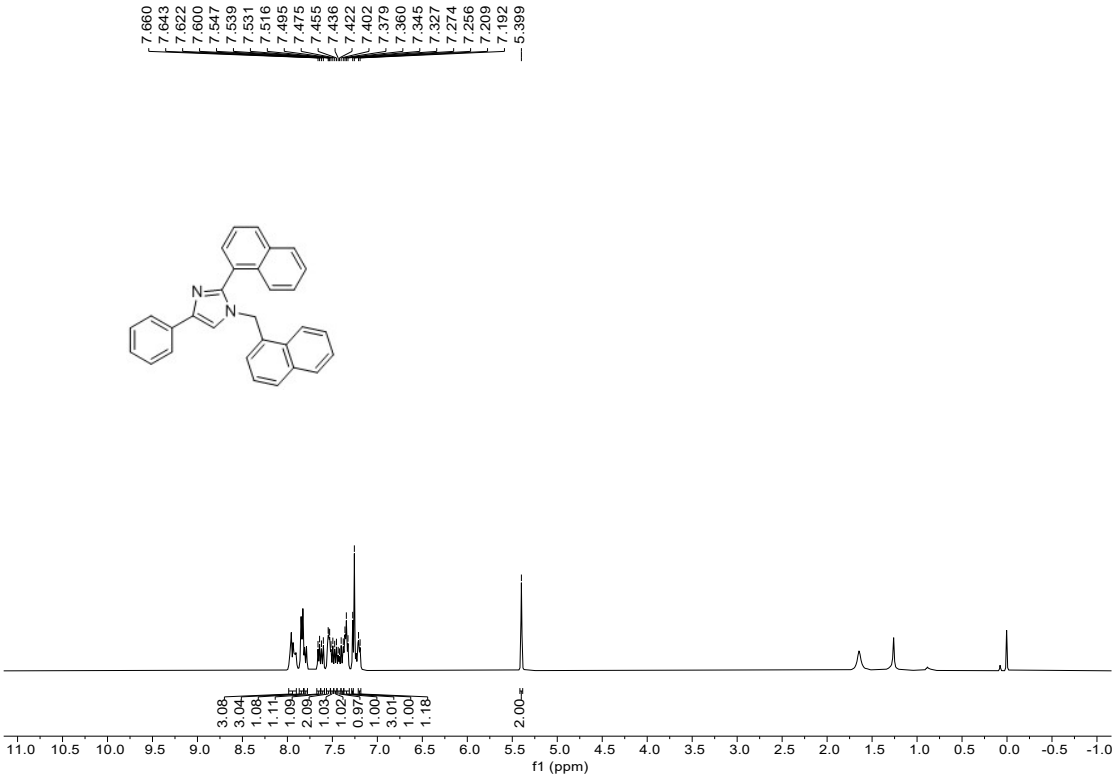
^{13}C NMR Spectrum of **3k**



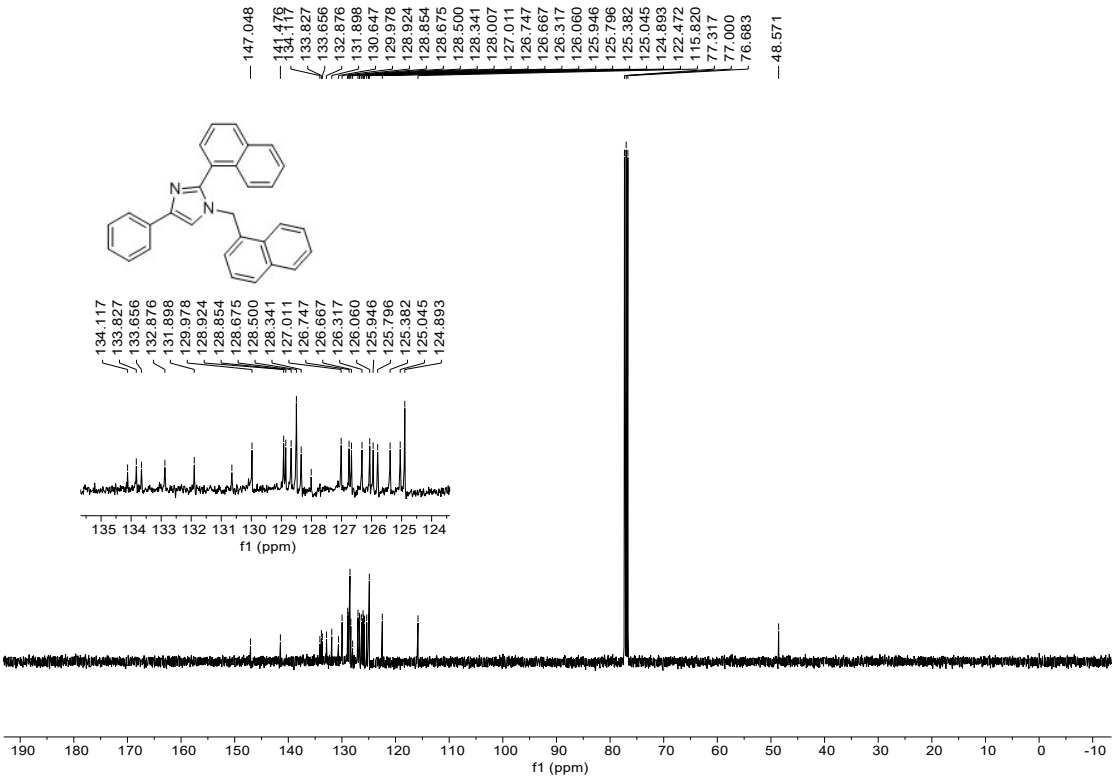
^{19}F NMR Spectrum of **3k**



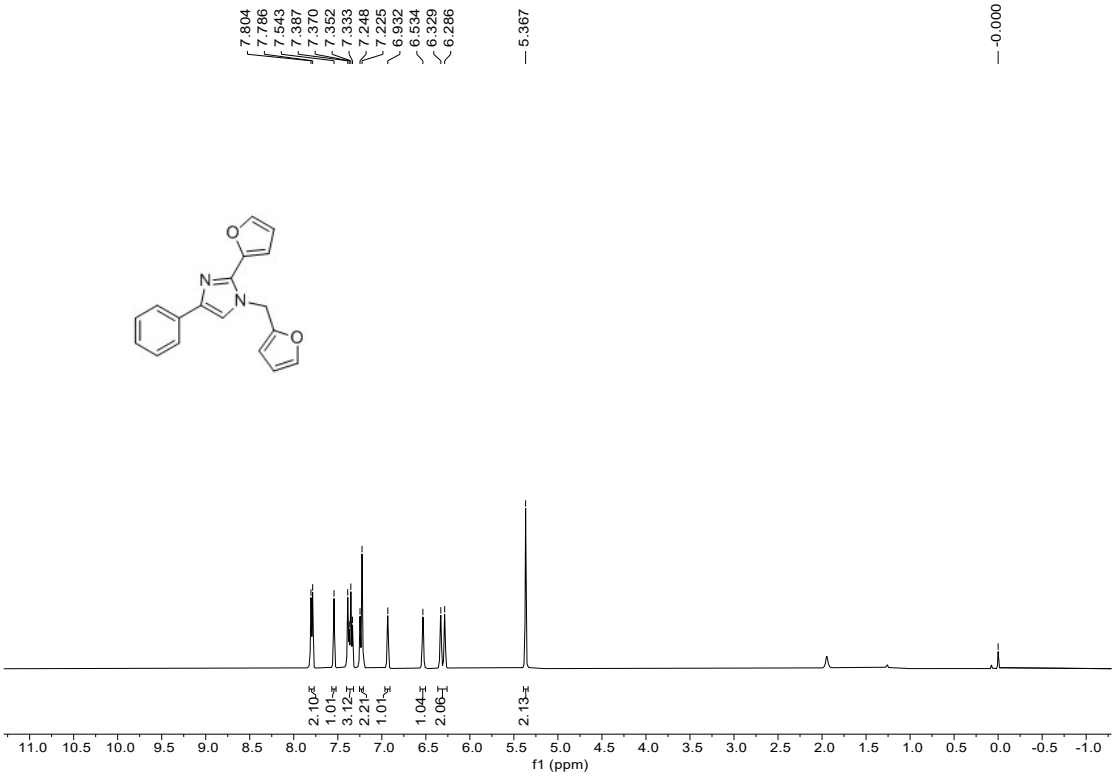
^{1}H NMR Spectrum of **3l**



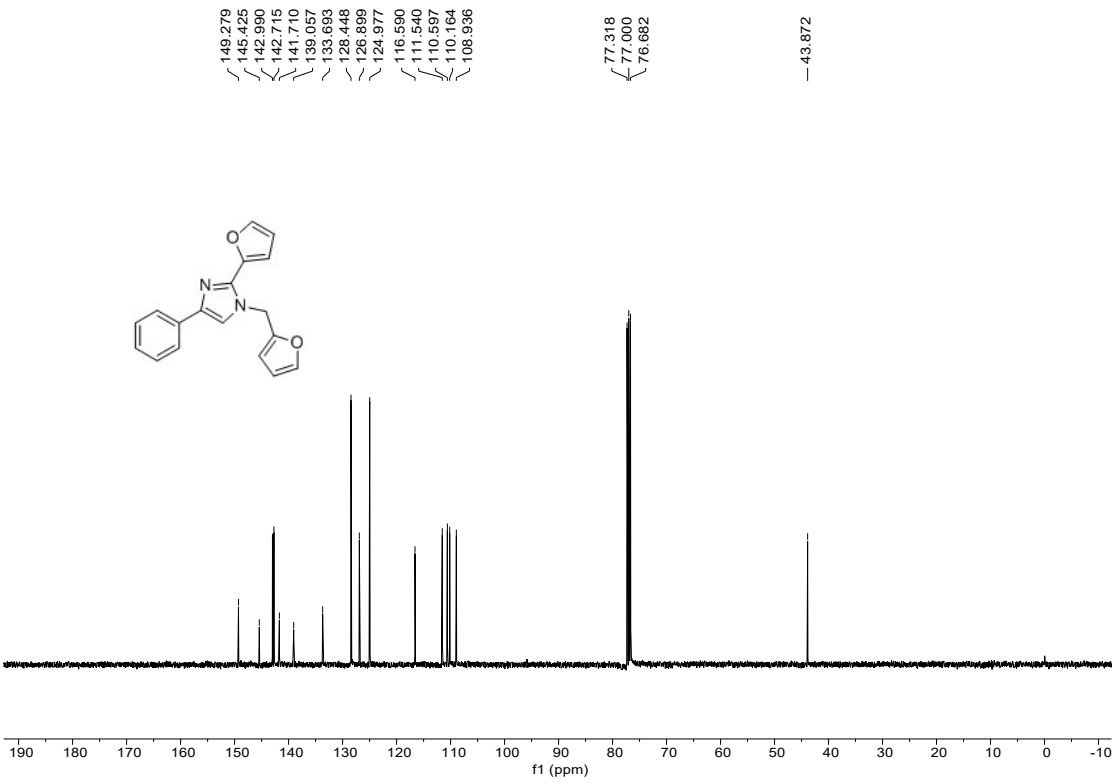
^{13}C NMR Spectrum of **3l**



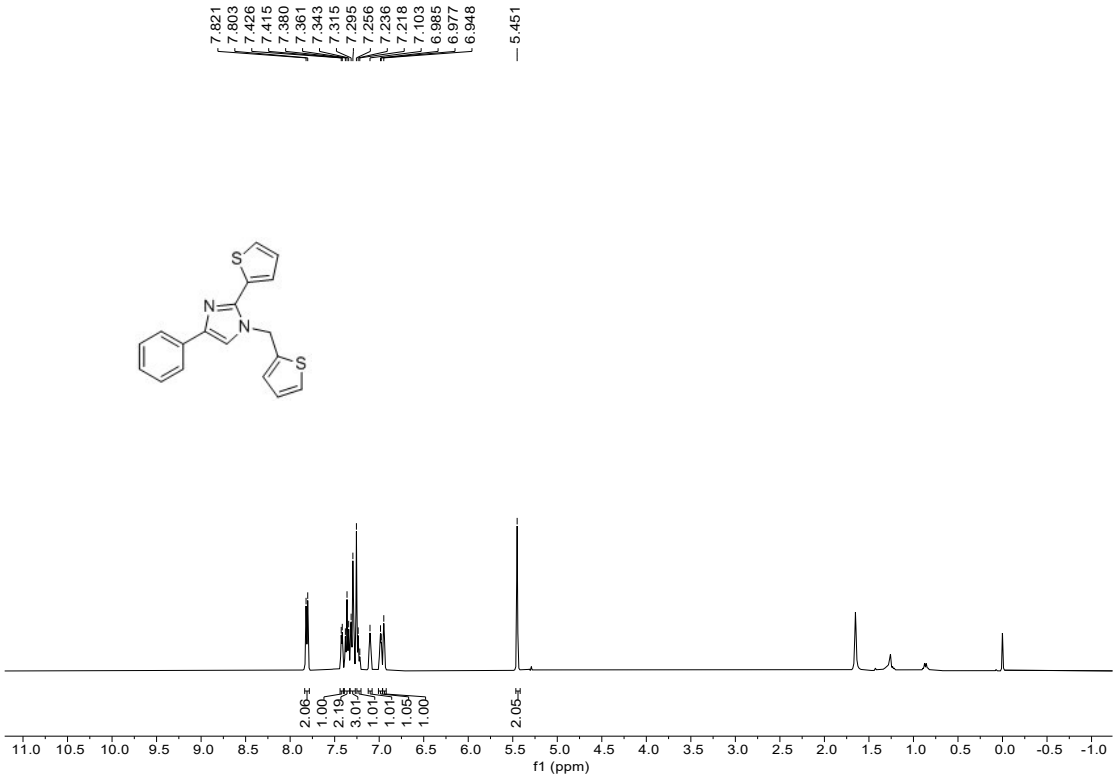
^{1}H NMR Spectrum of **3m**



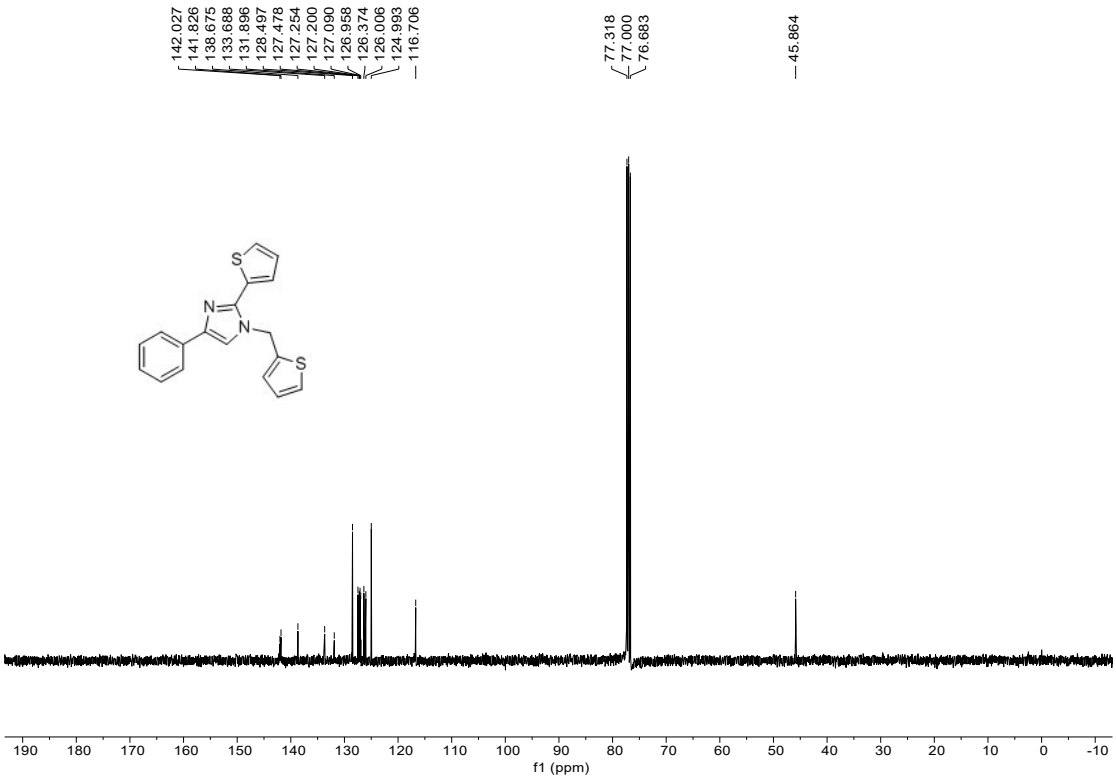
^{13}C NMR Spectrum of **3m**



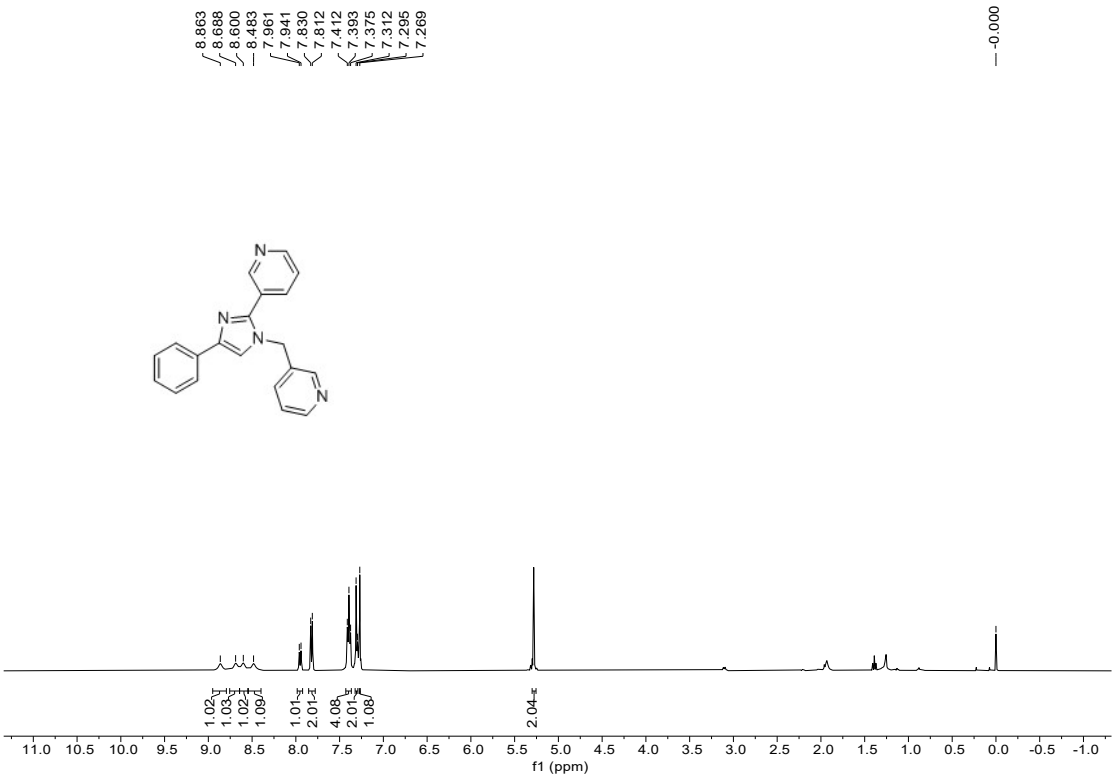
^{1}H NMR Spectrum of **3n**



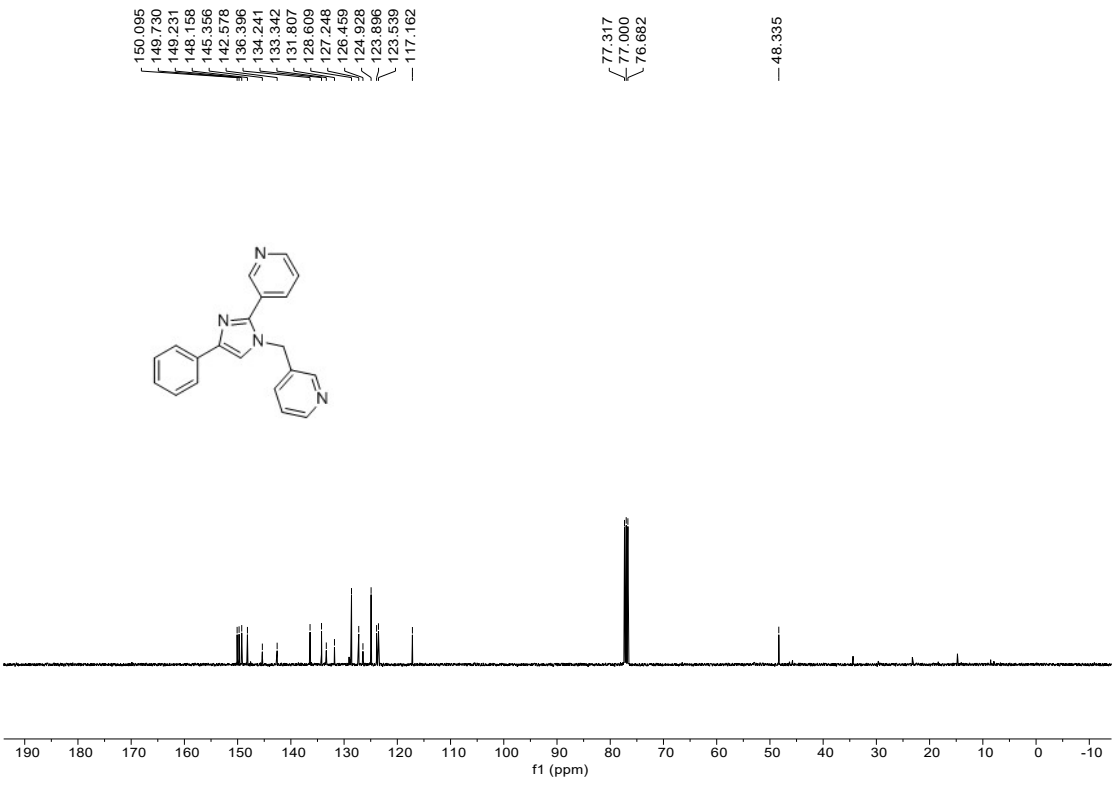
^{13}C NMR Spectrum of **3n**



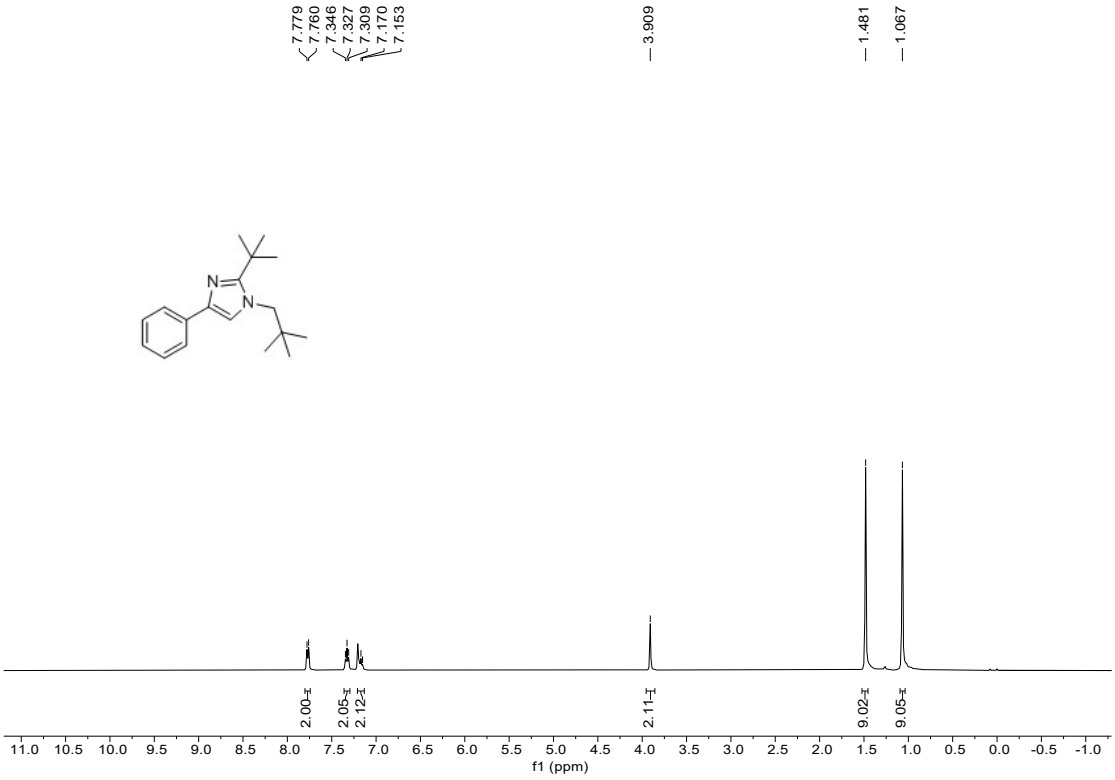
^{1}H NMR Spectrum of **3o**



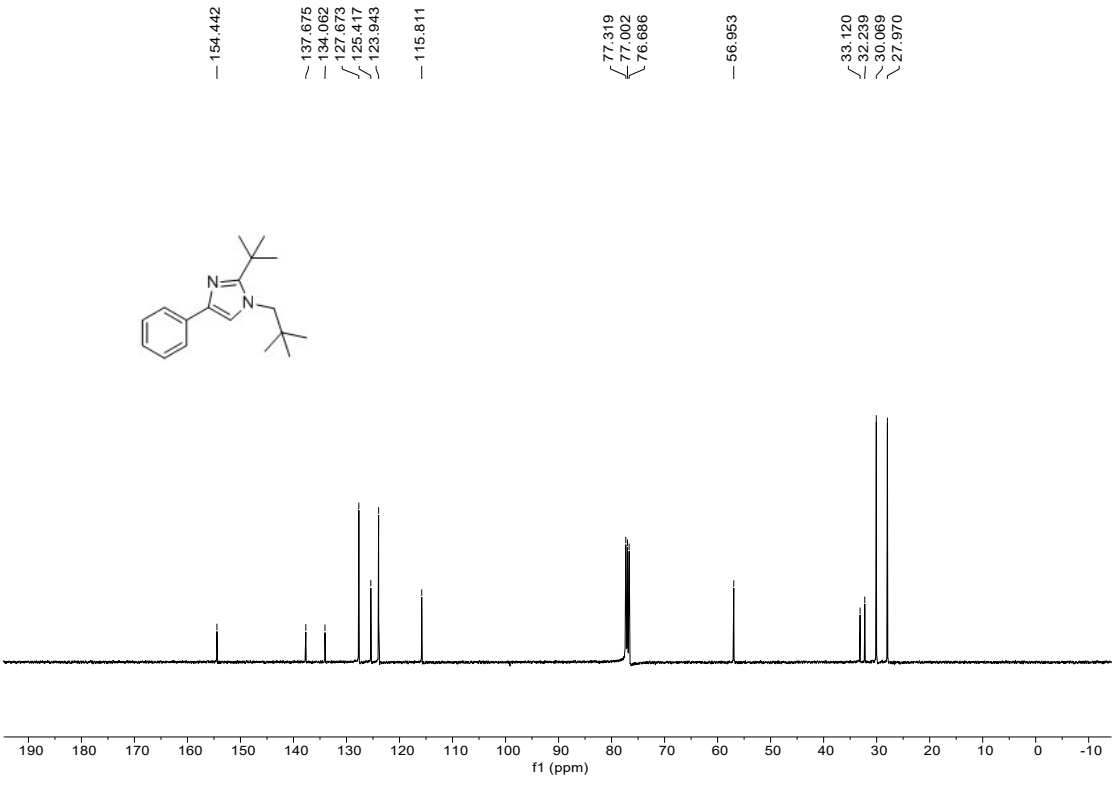
^{13}C NMR Spectrum of **3o**



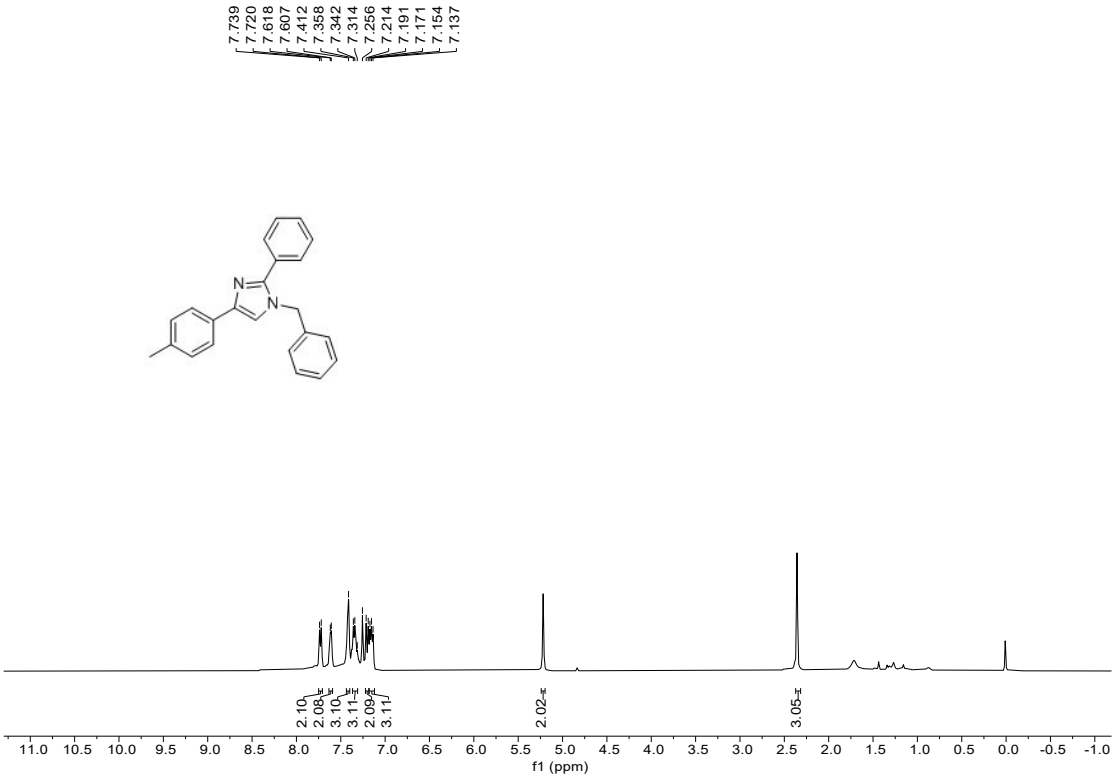
^{1}H NMR Spectrum of **3p**



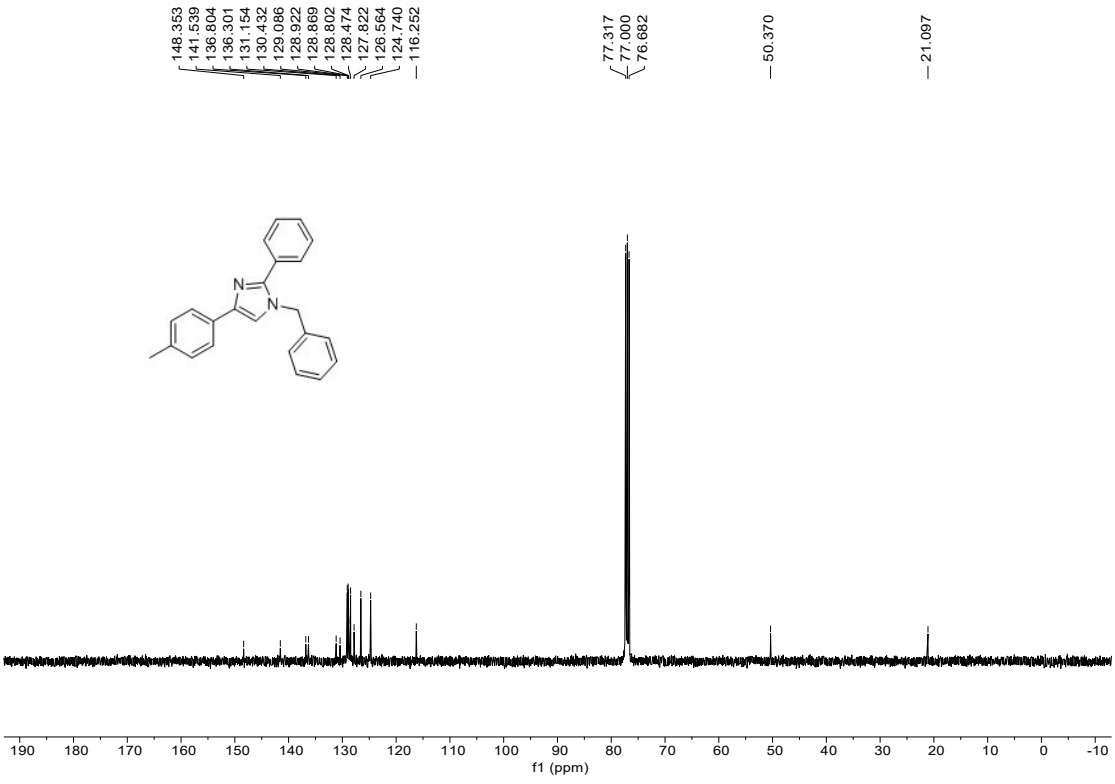
^{13}C NMR Spectrum of **3p**



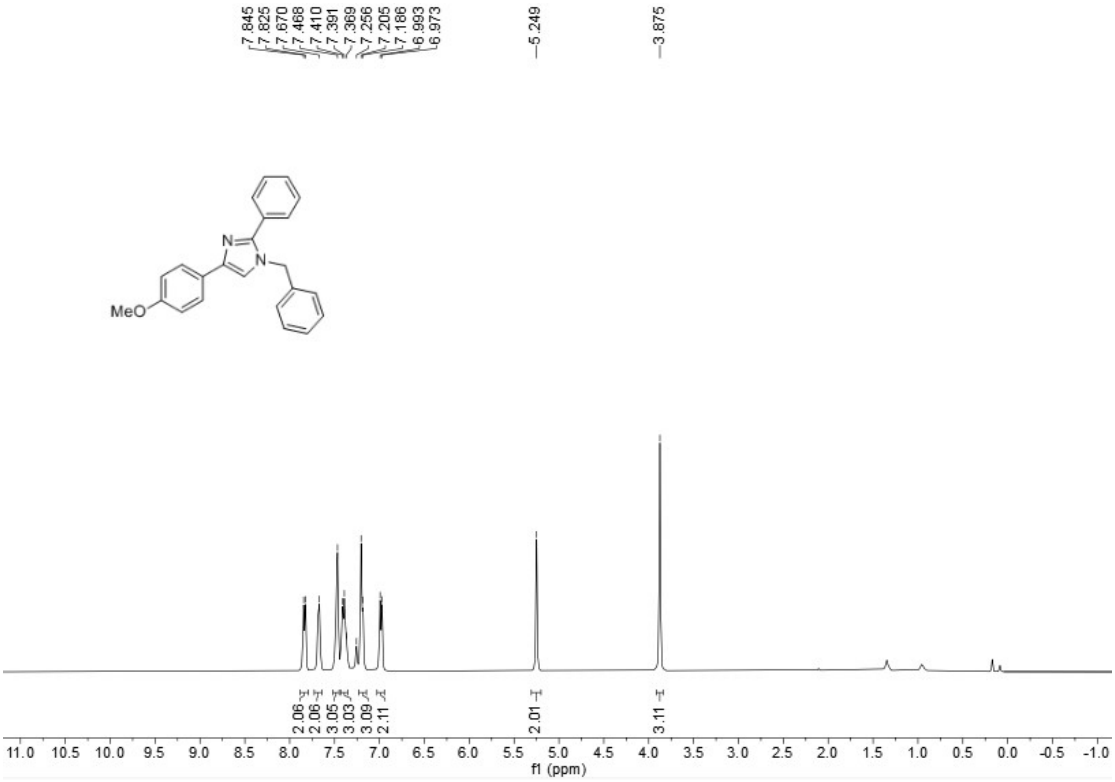
^{1}H NMR Spectrum of **3q**



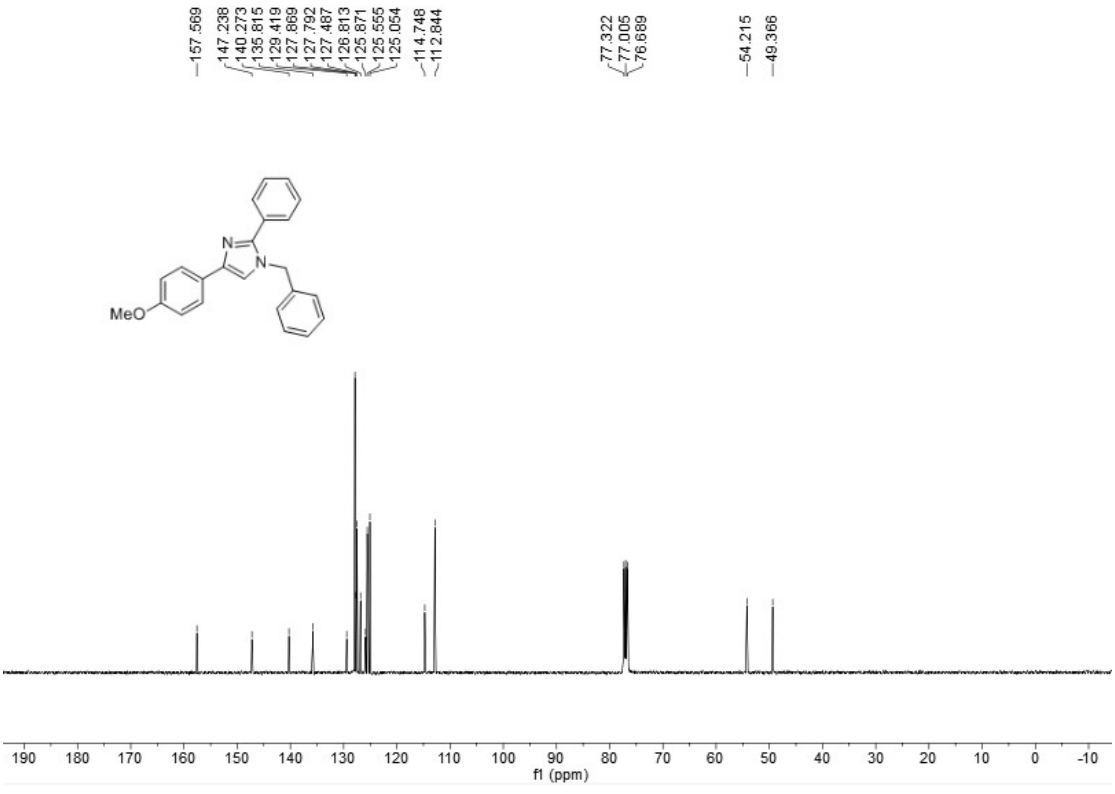
^{13}C NMR Spectrum of **3q**



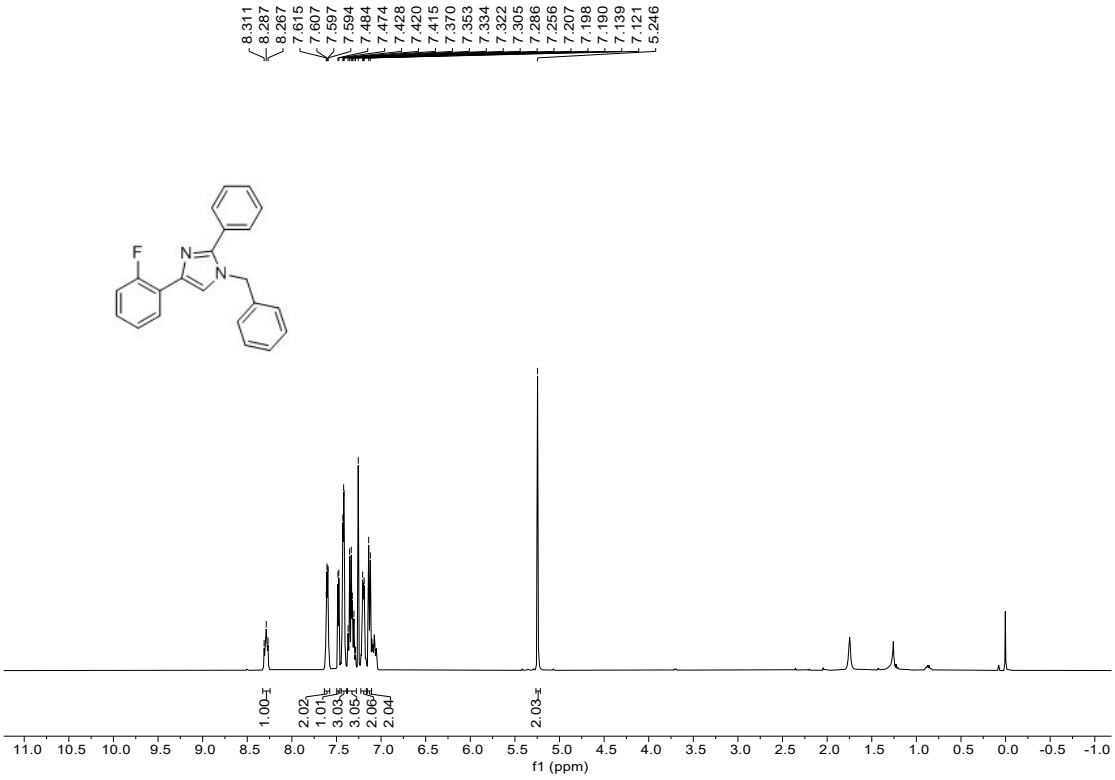
^{1}H NMR Spectrum of **3r**



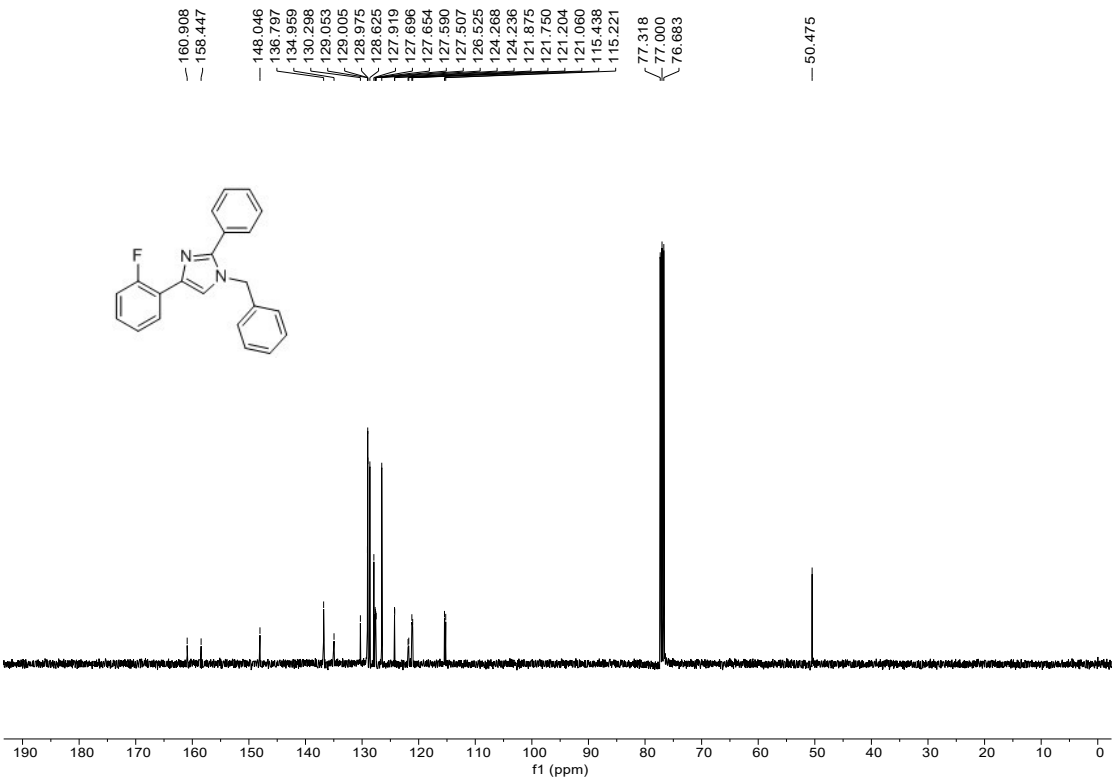
^{13}C NMR Spectrum of **3r**



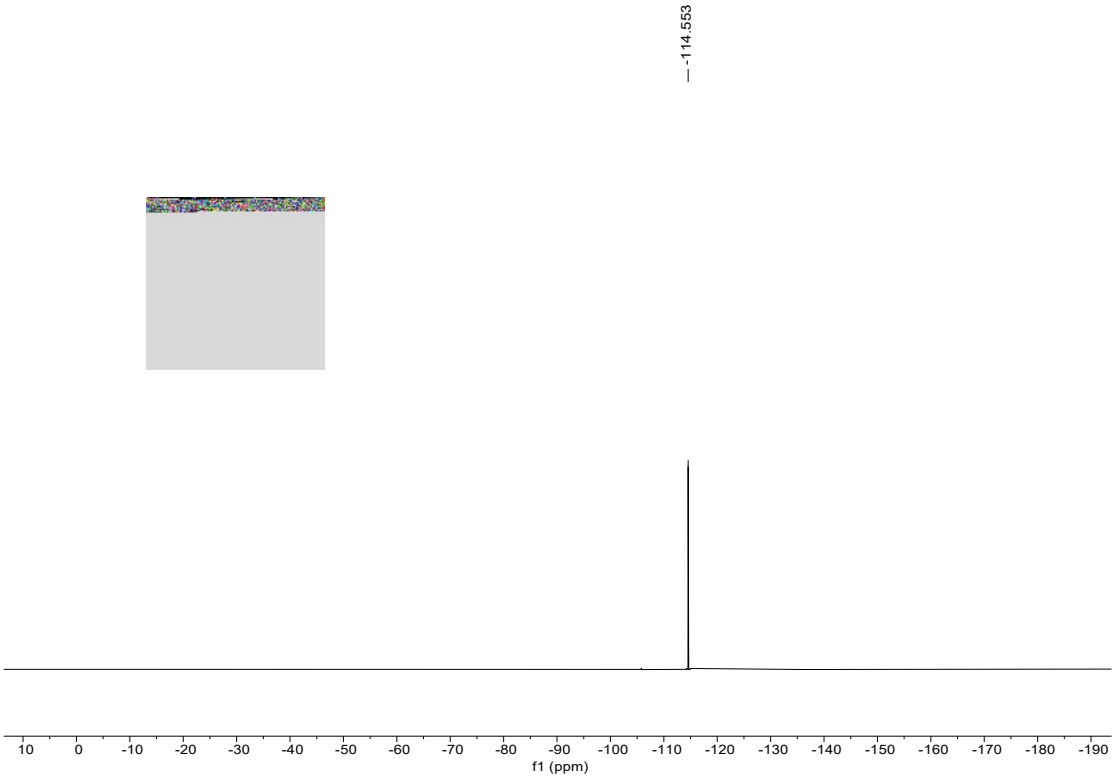
^{1}H NMR Spectrum of **3s**



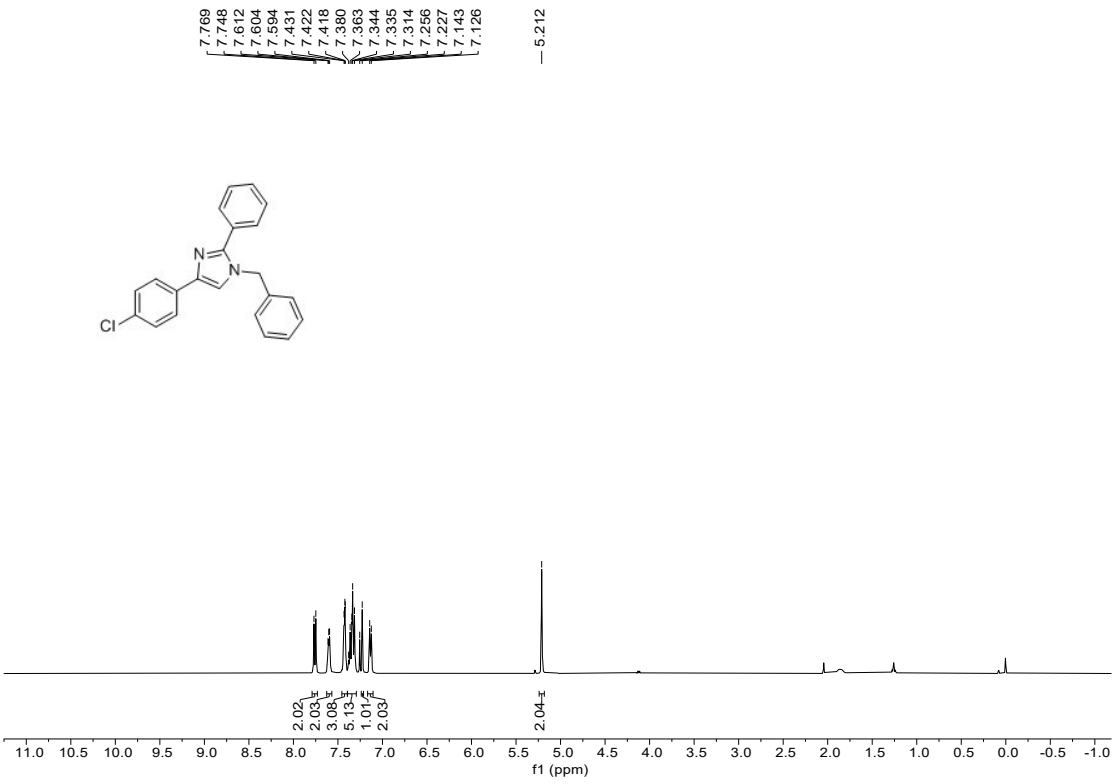
^{13}C NMR Spectrum of **3s**



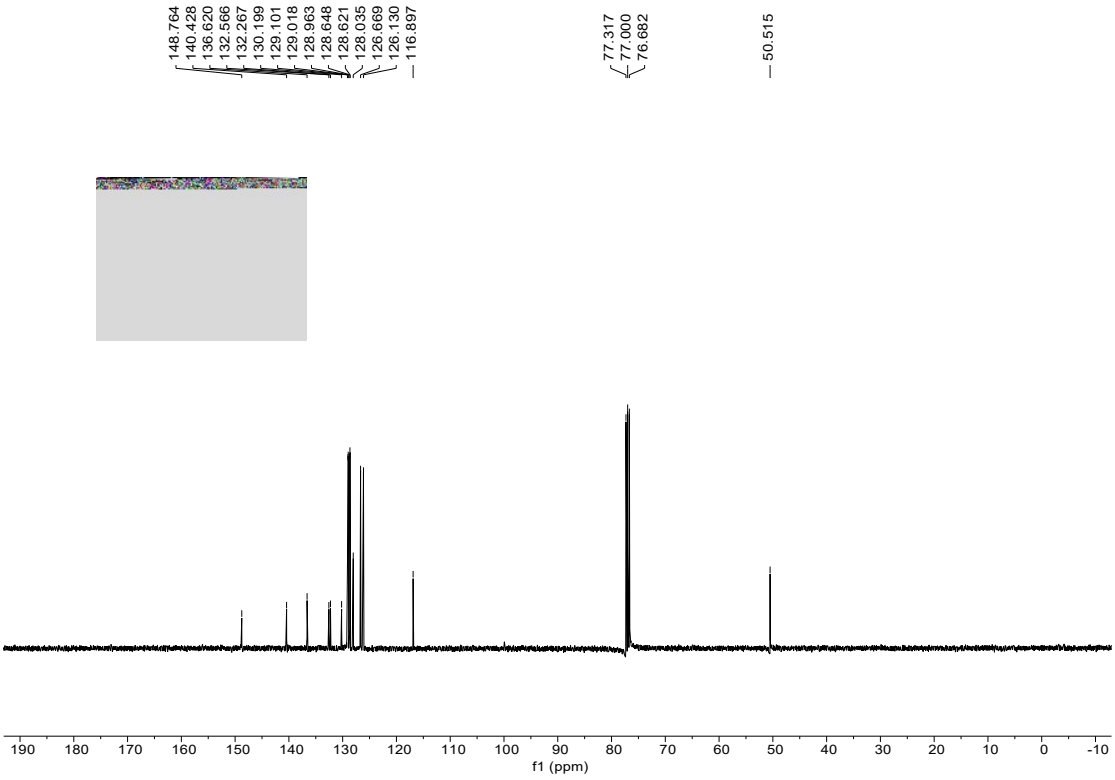
^{19}F NMR Spectrum of **3s**



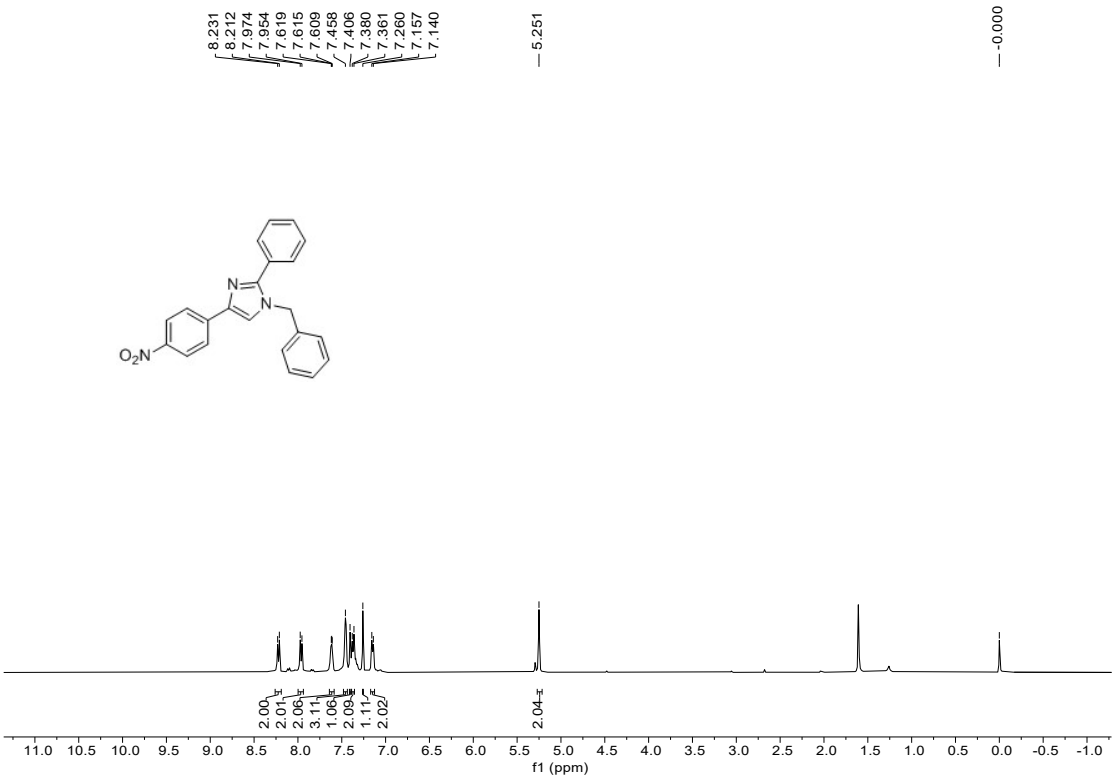
^{1}H NMR Spectrum of **3t**



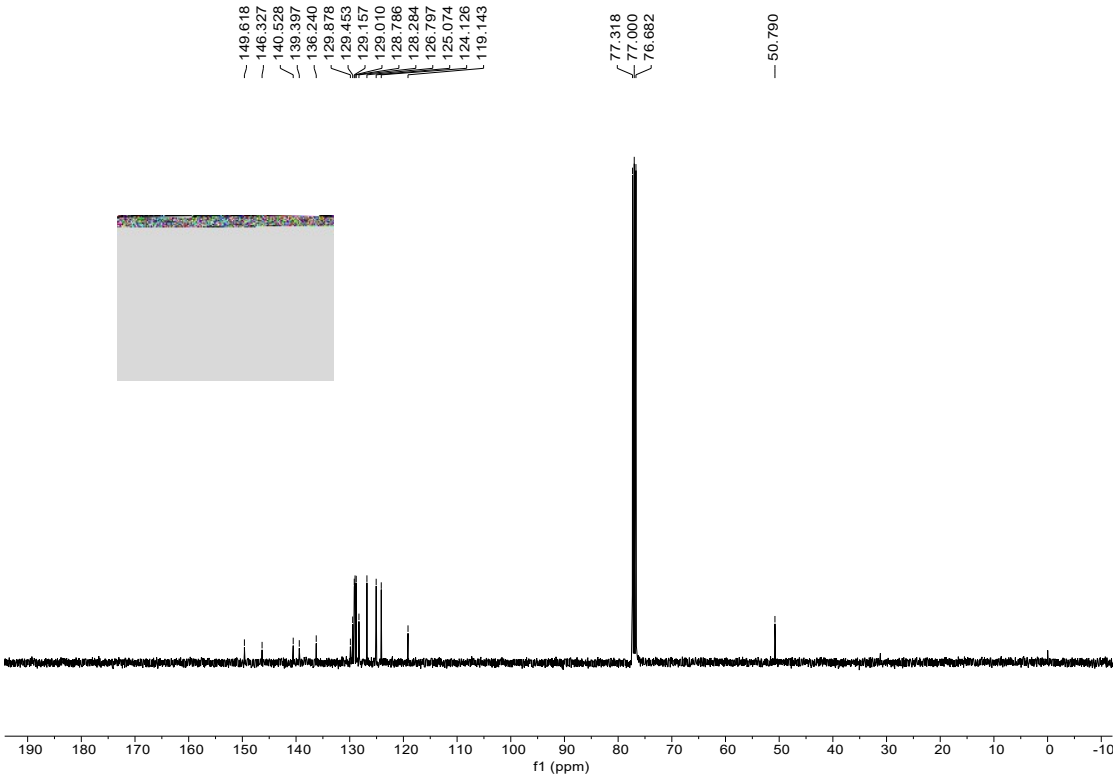
^{13}C NMR Spectrum of **3t**



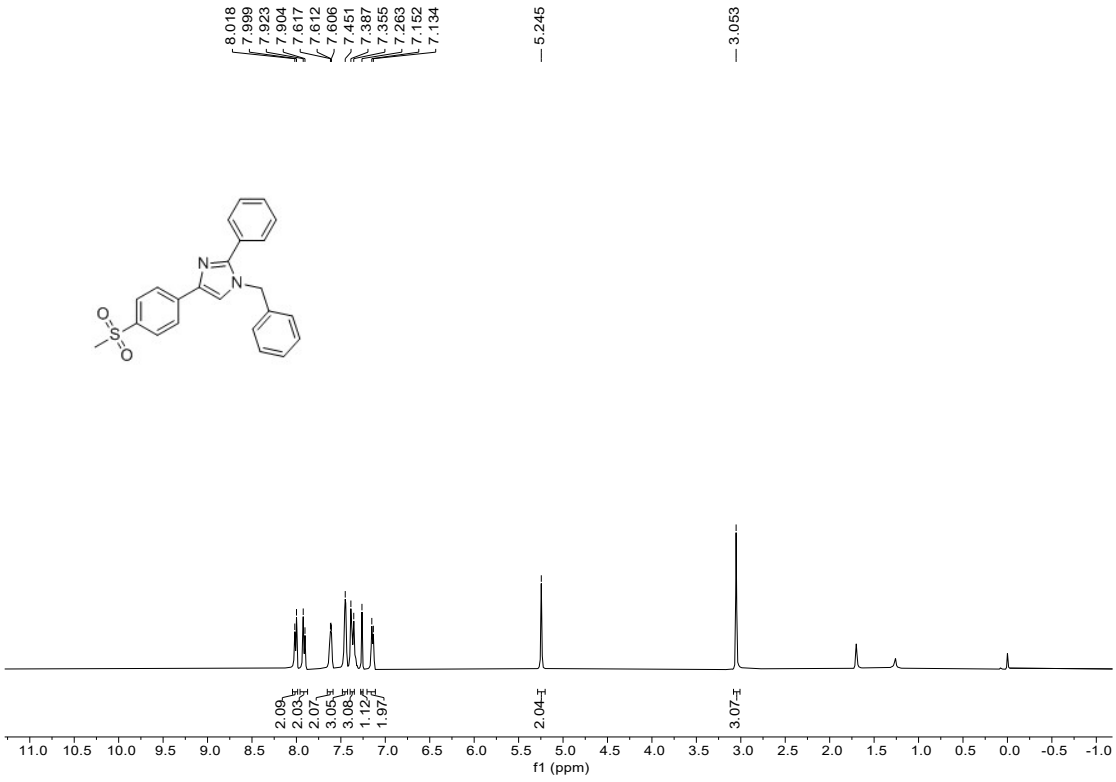
^{1}H NMR Spectrum of **3u**



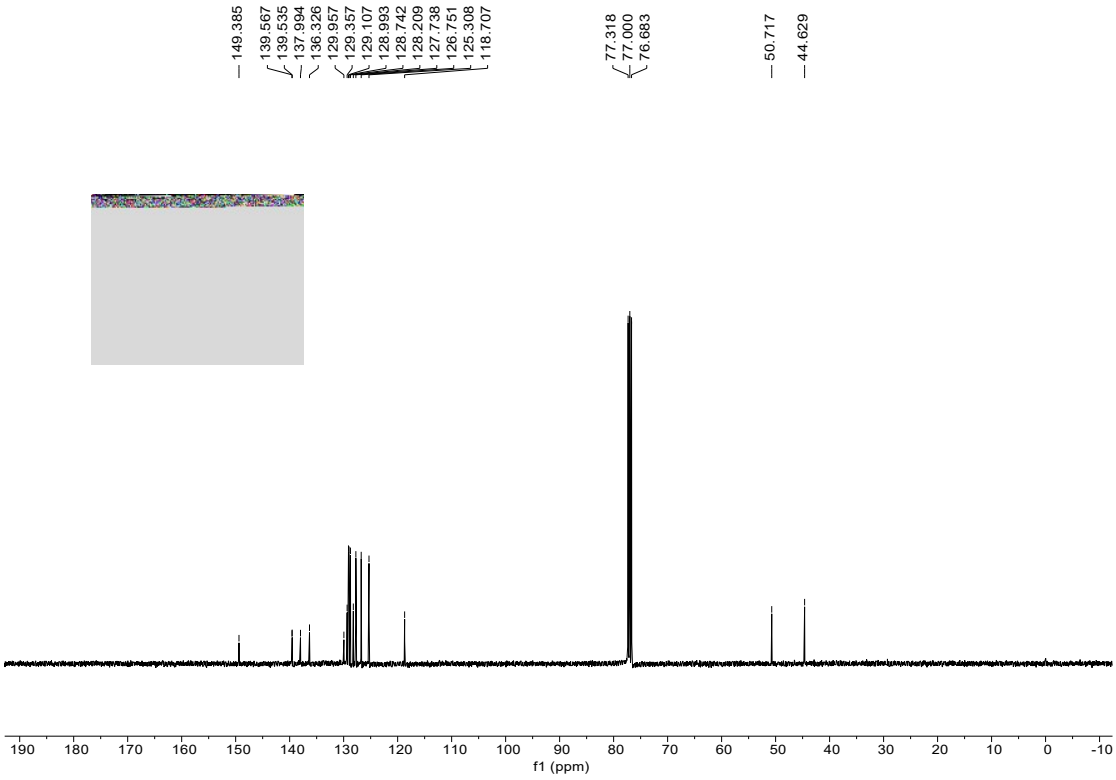
^{13}C NMR Spectrum of **3u**



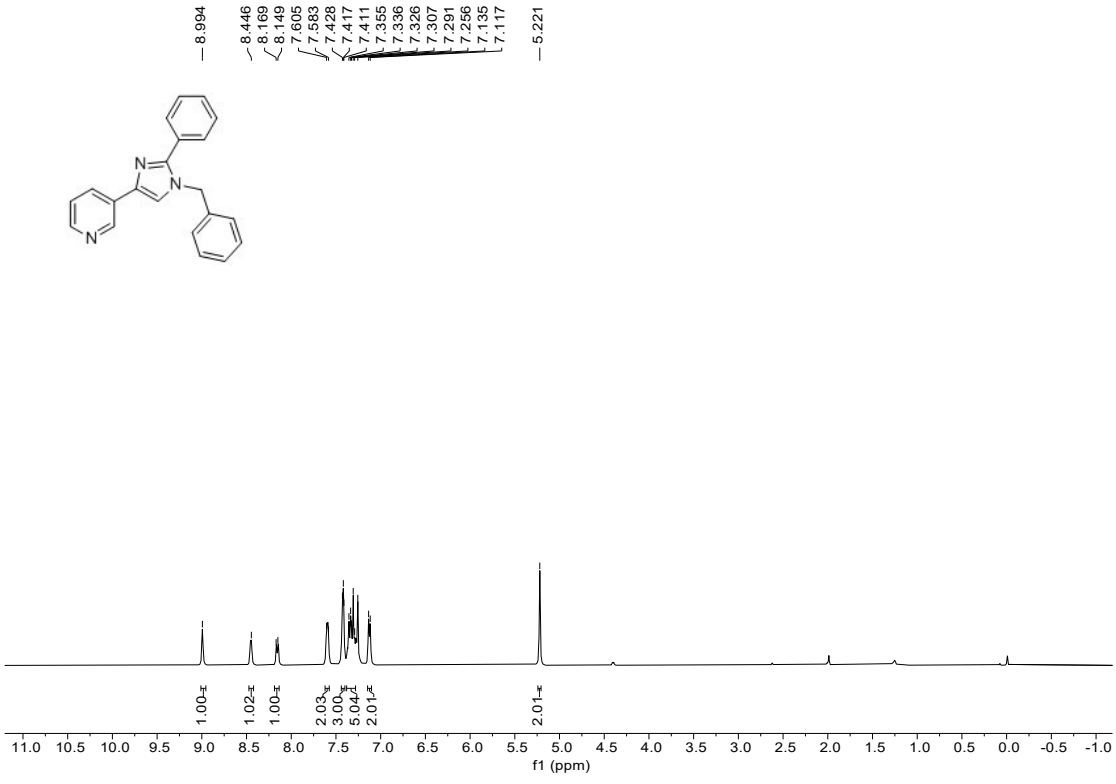
^{1}H NMR Spectrum of **3v**



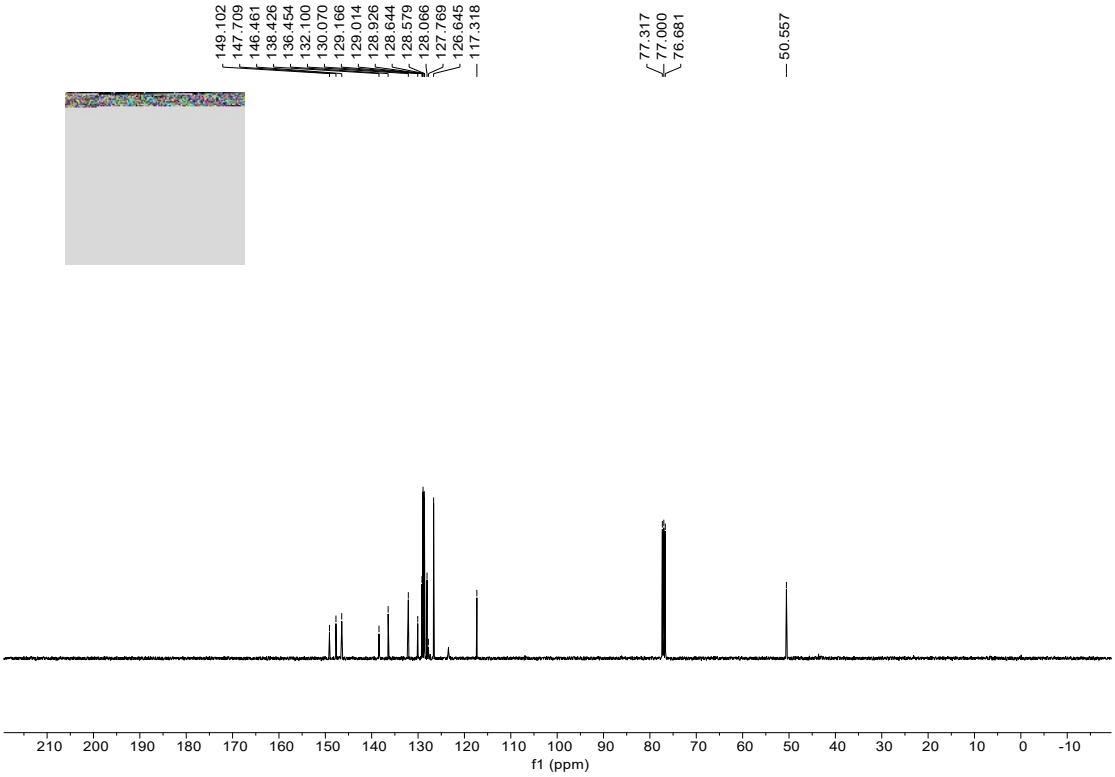
^{13}C NMR Spectrum of **3v**



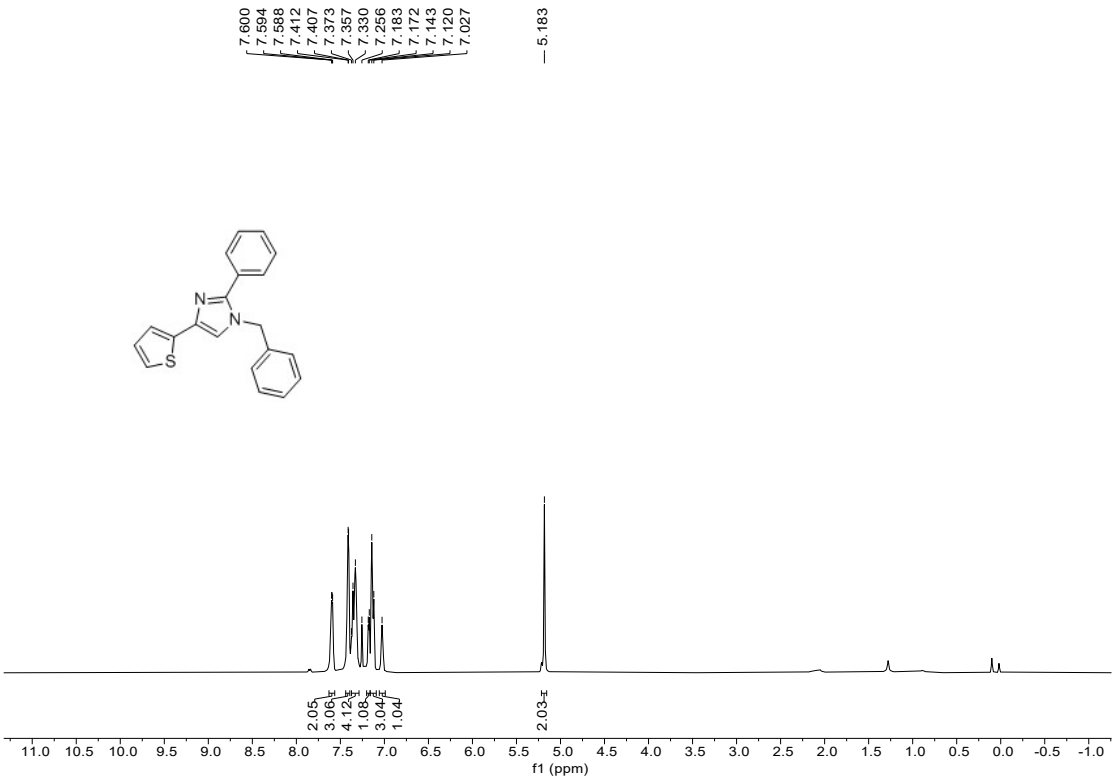
^{1}H NMR Spectrum of **3w**



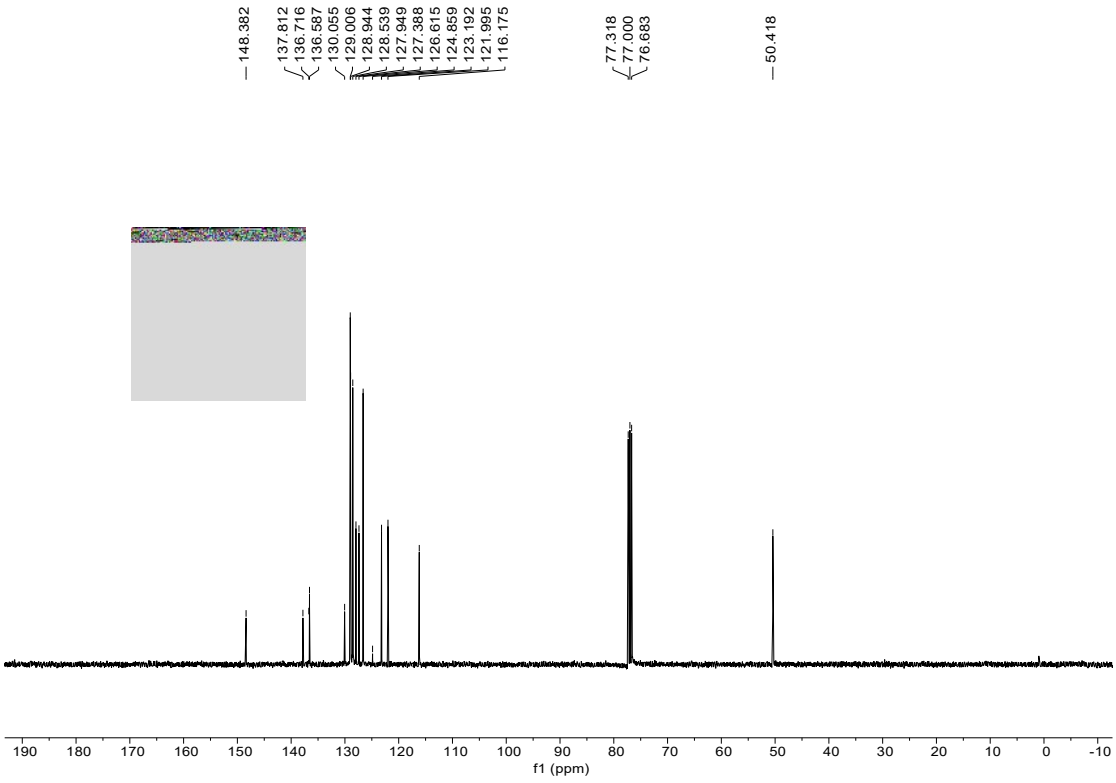
^{13}C NMR Spectrum of **3w**



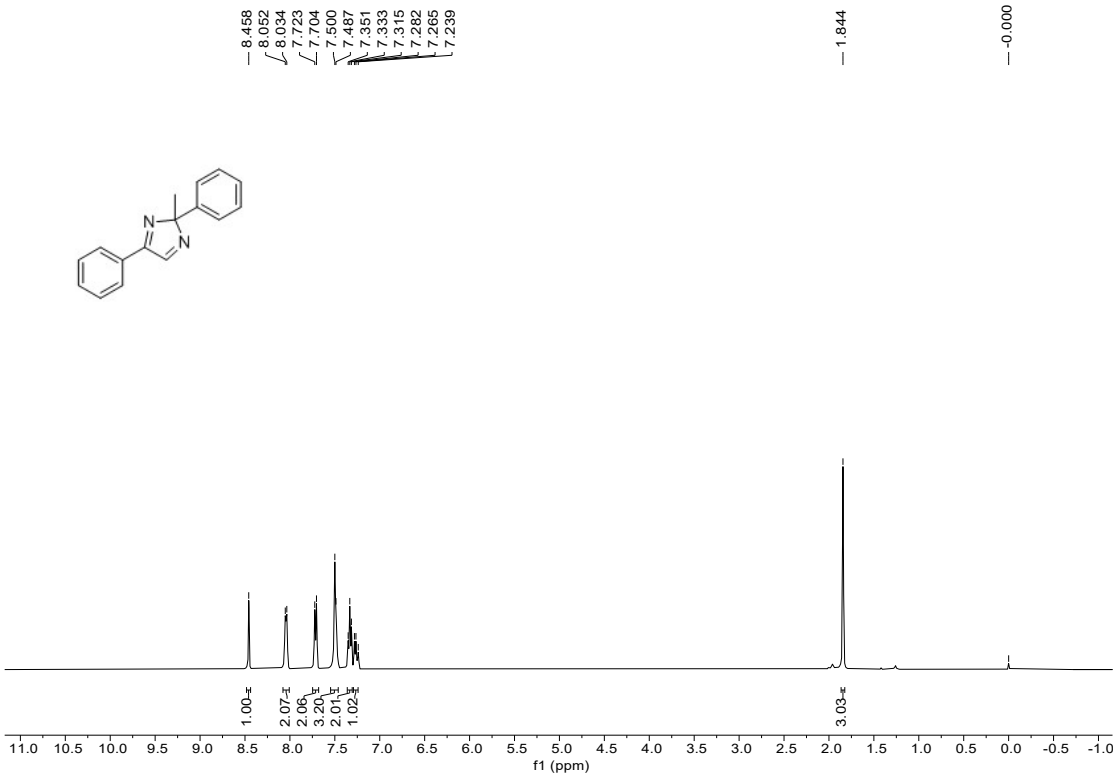
^{1}H NMR Spectrum of **3x**



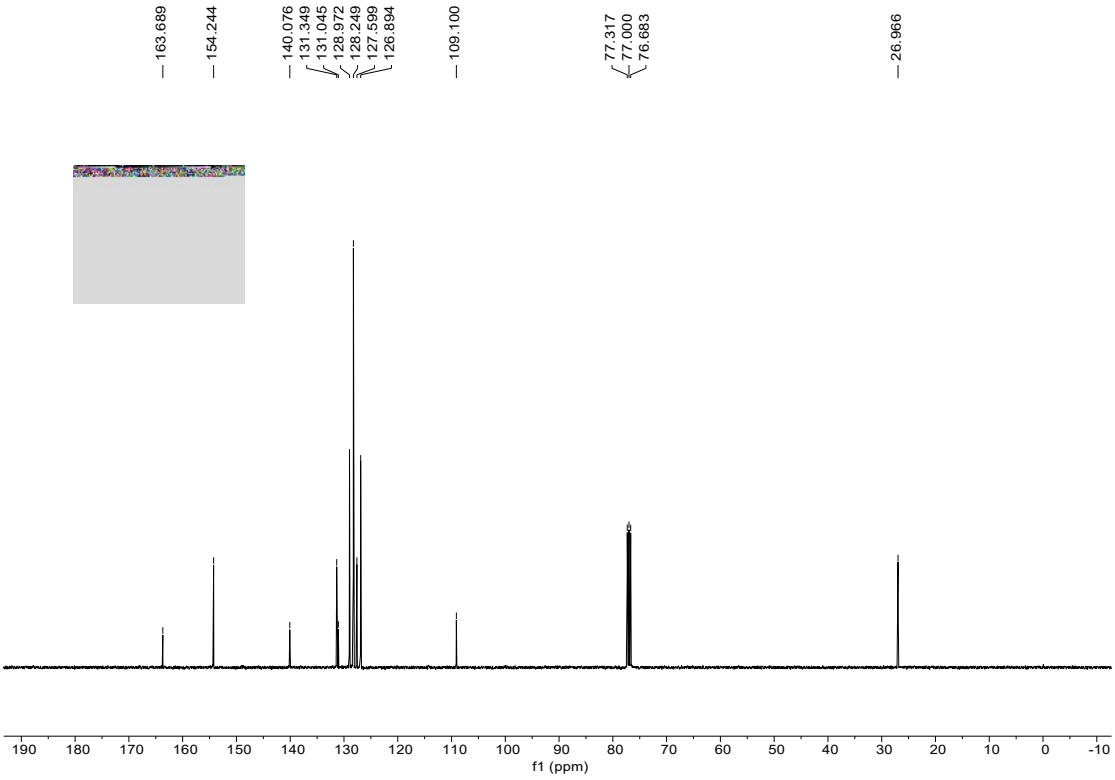
^{13}C NMR Spectrum of **3x**



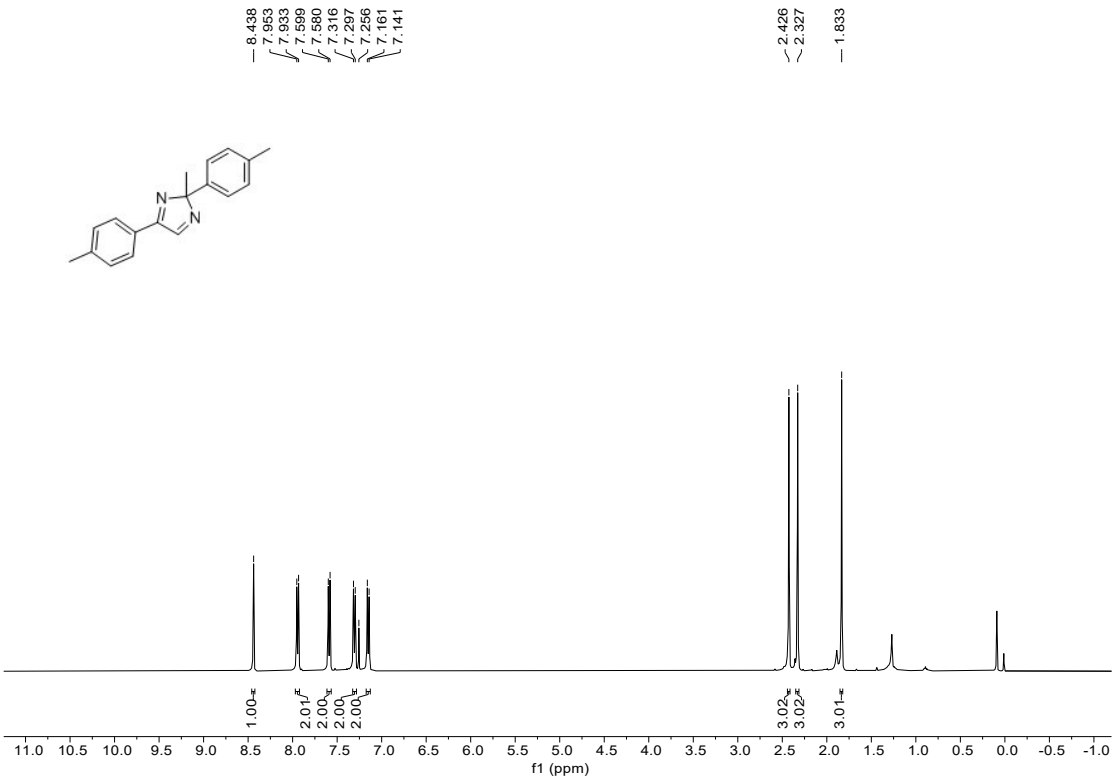
^{1}H NMR Spectrum of **4a**



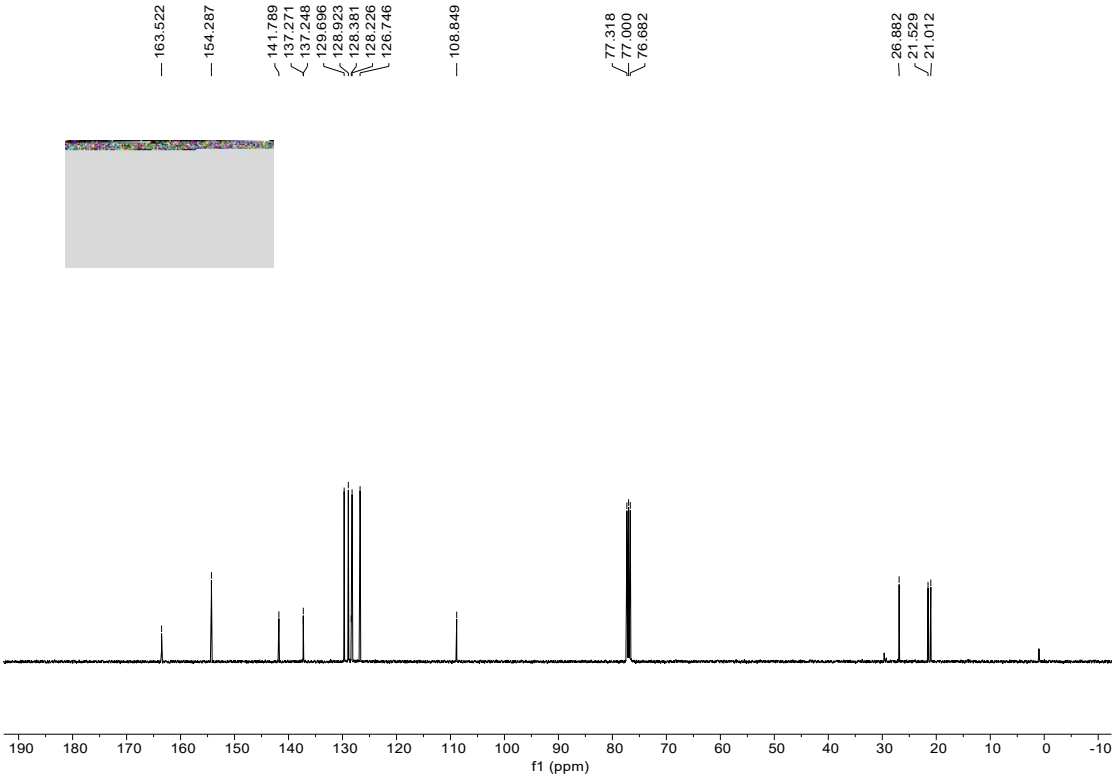
^{13}C NMR Spectrum of **4a**



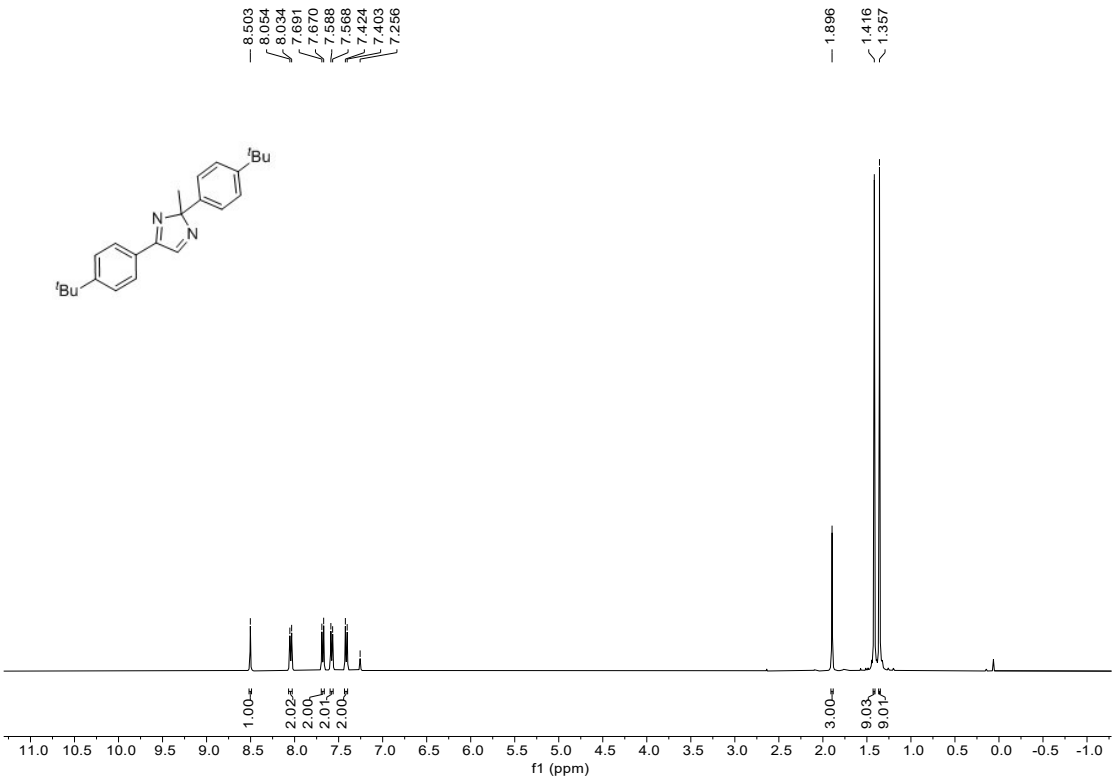
^{1}H NMR Spectrum of **4b**



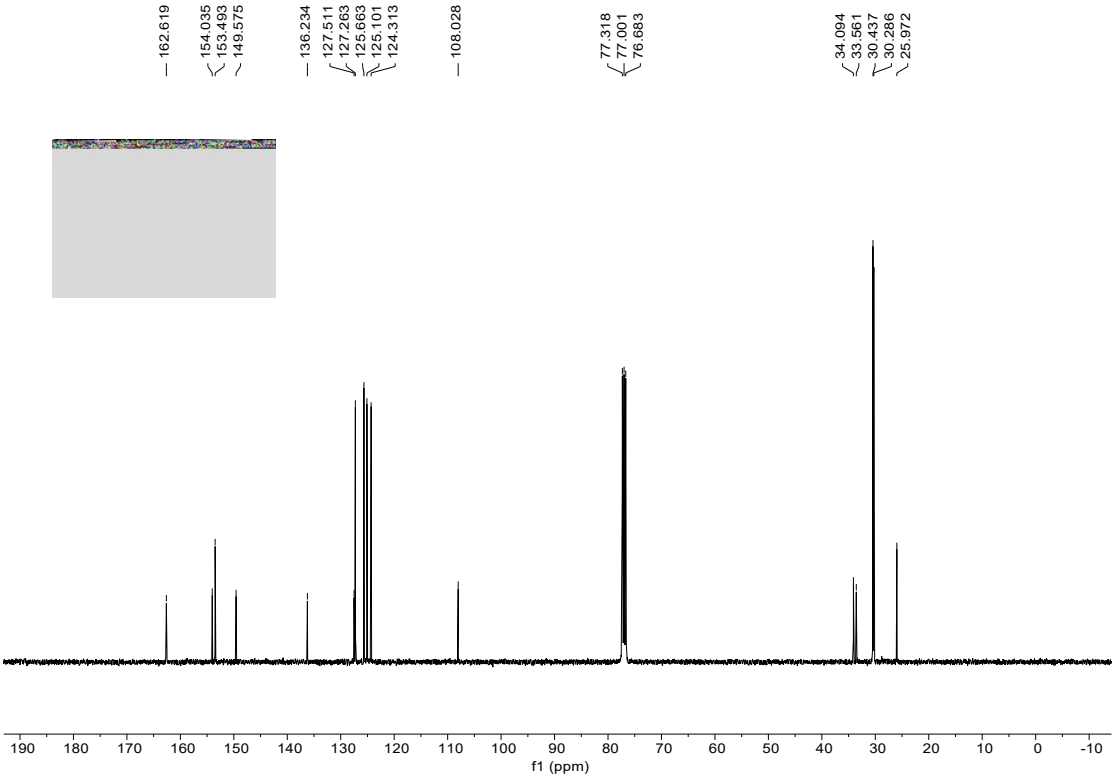
^{13}C NMR Spectrum of **4b**



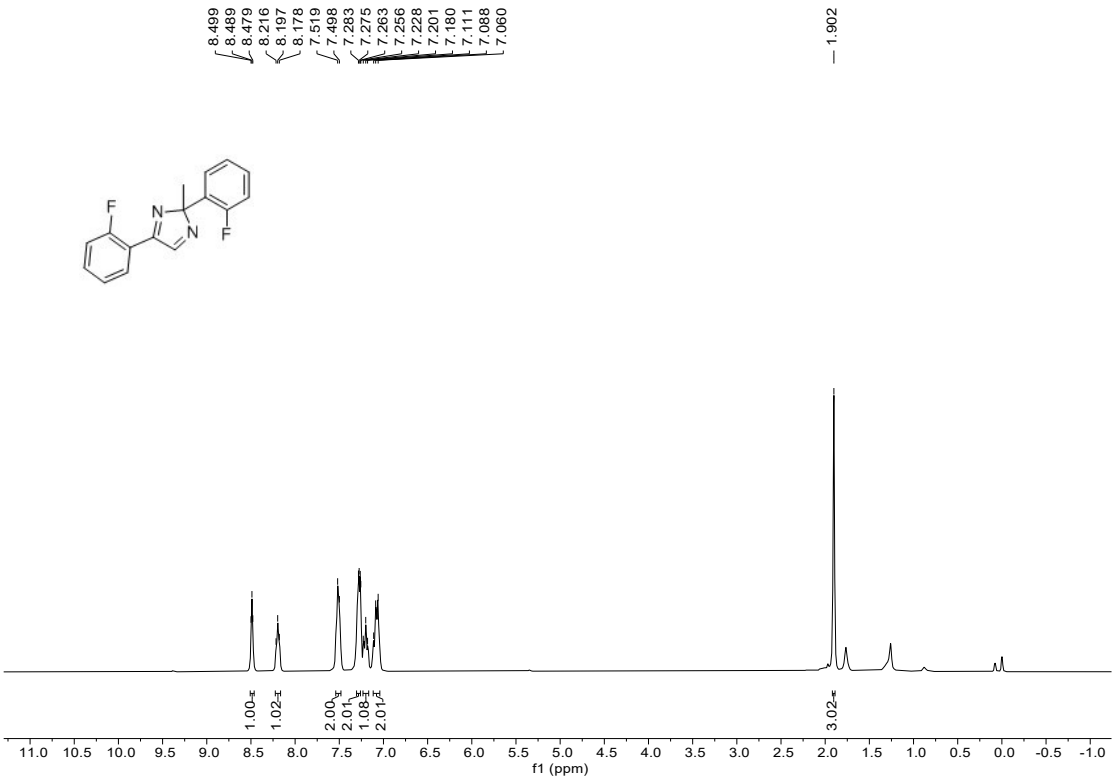
^{1}H NMR Spectrum of **4c**



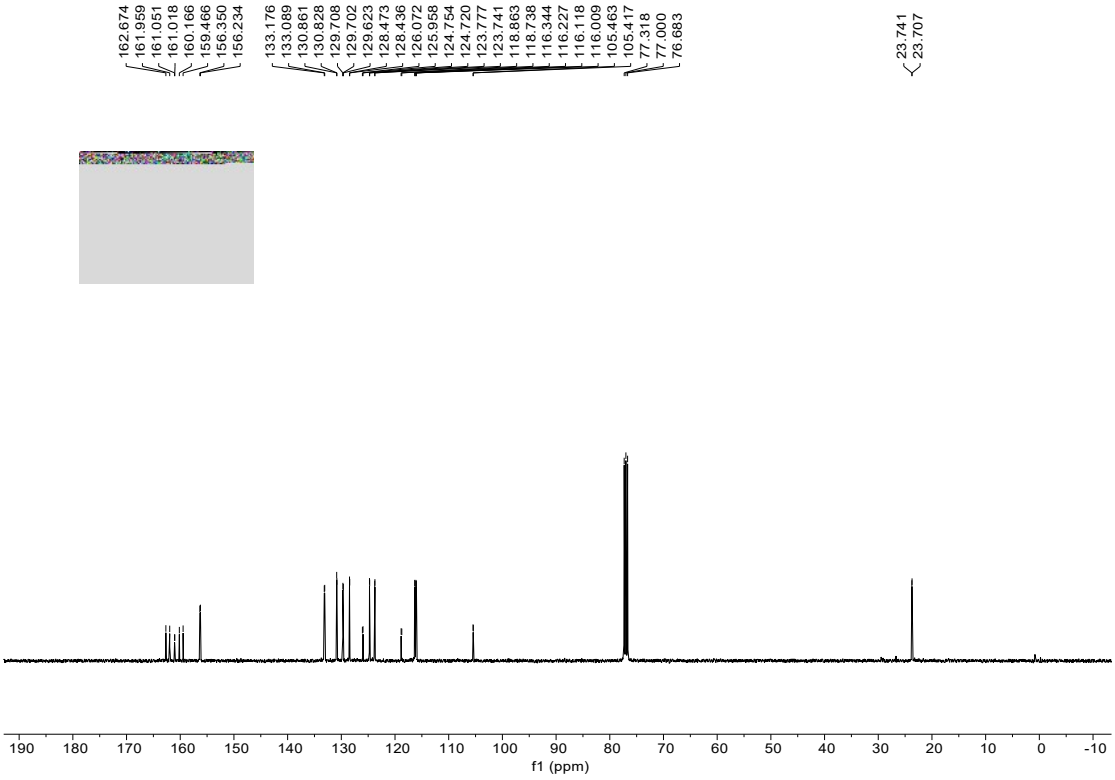
^{13}C NMR Spectrum of **4c**



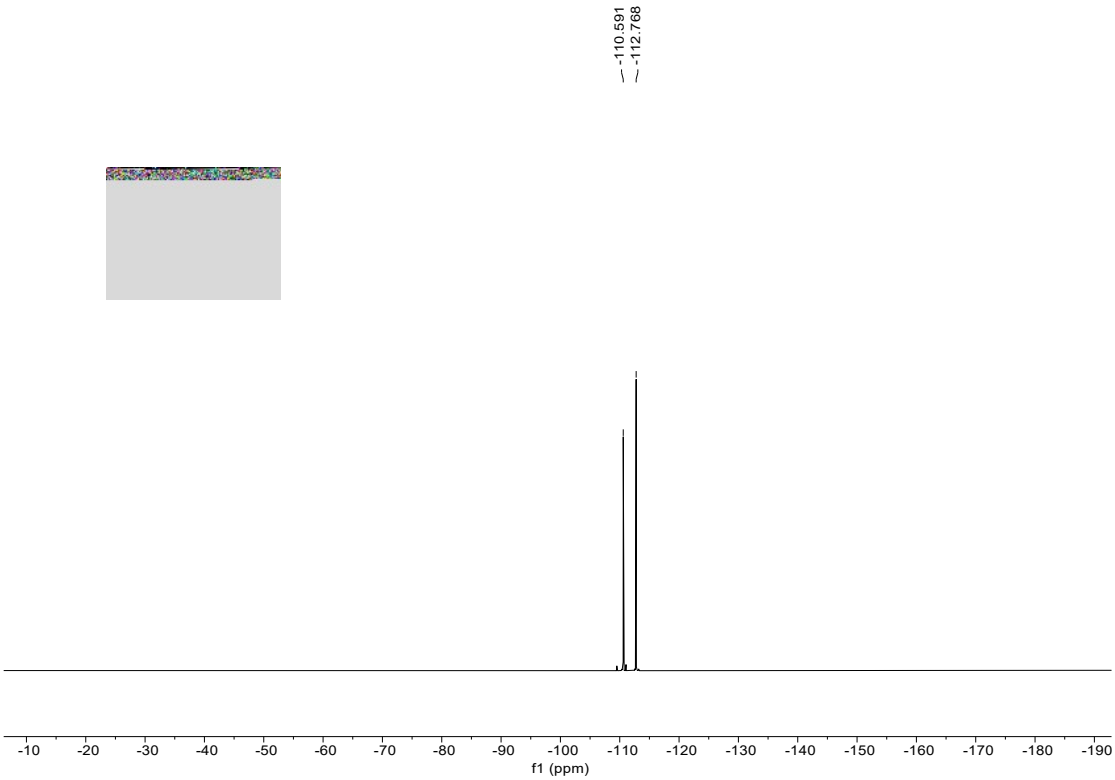
^{1}H NMR Spectrum of **4d**



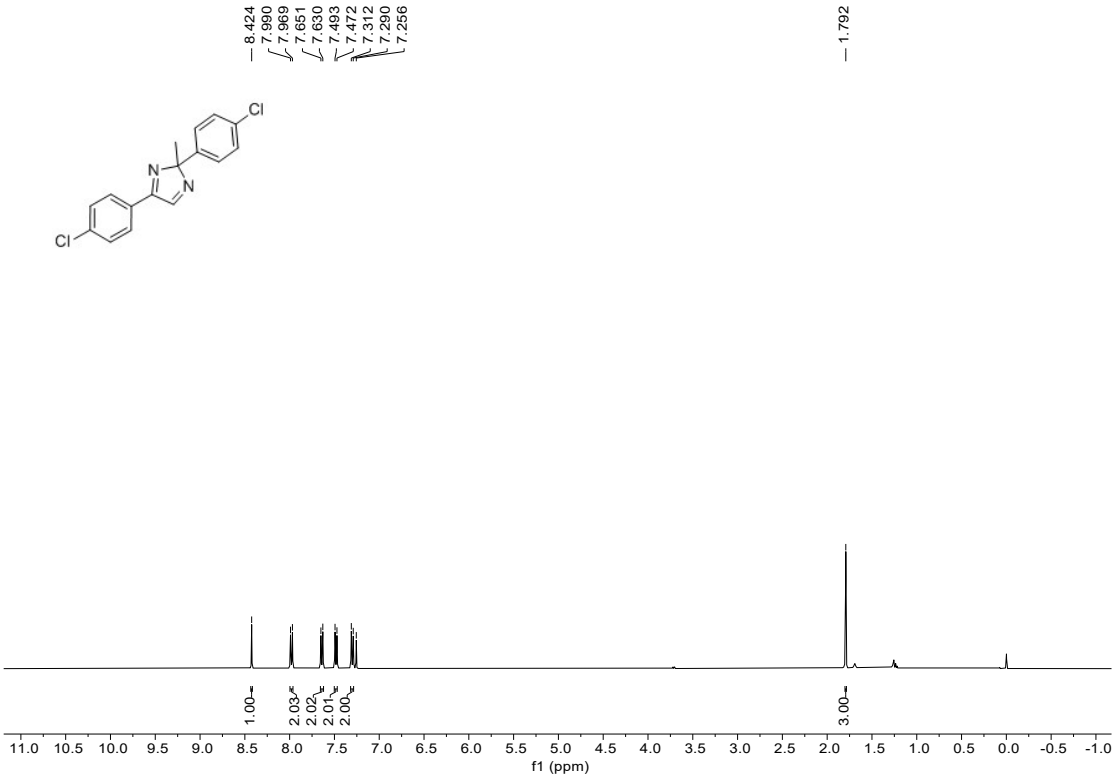
^{13}C NMR Spectrum of **4d**



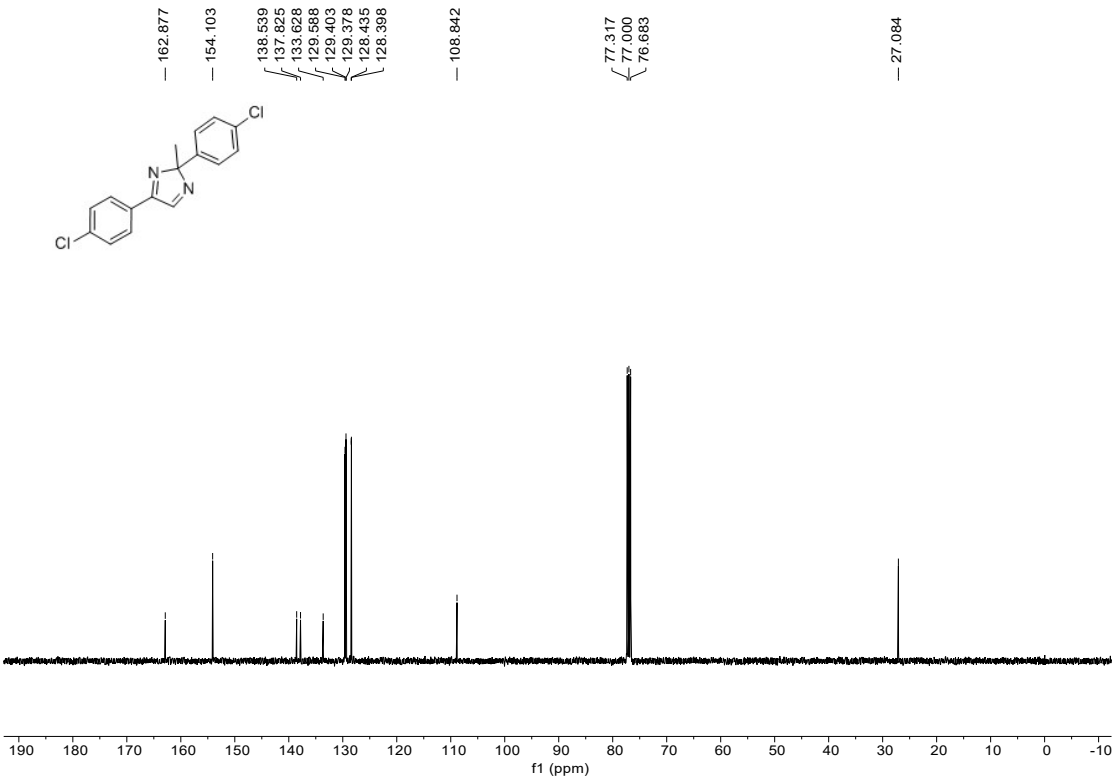
^{19}F NMR Spectrum of **4d**



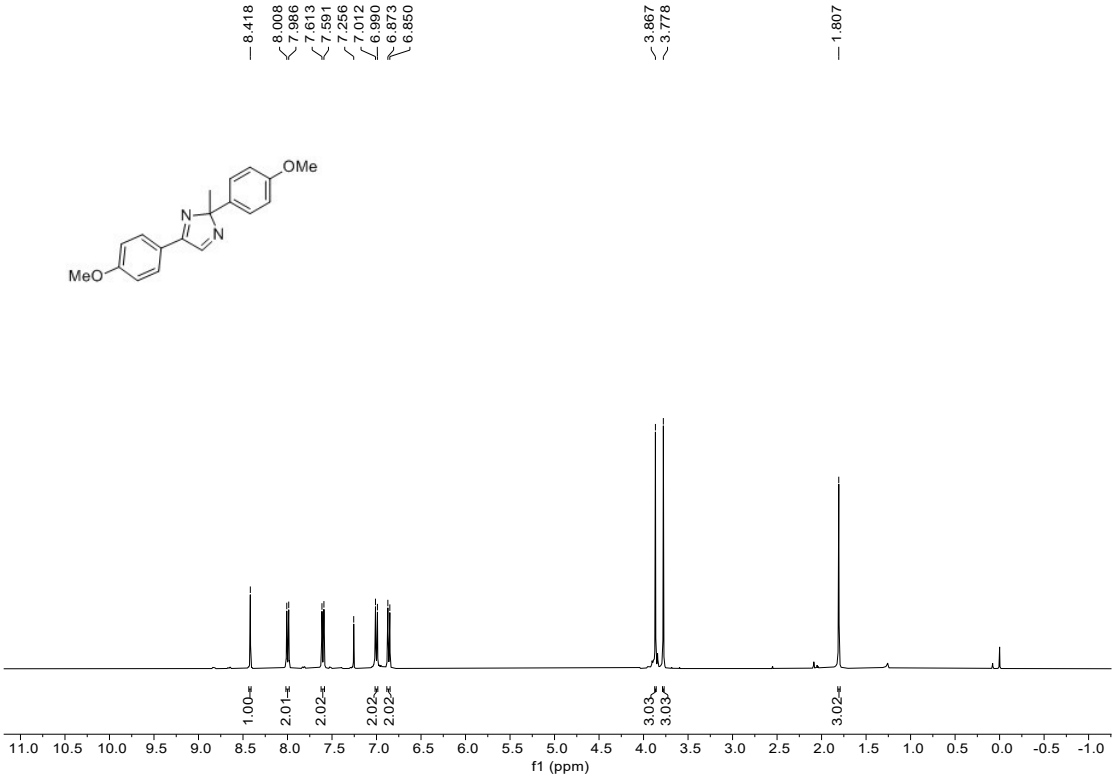
^{1}H NMR Spectrum of **4e**



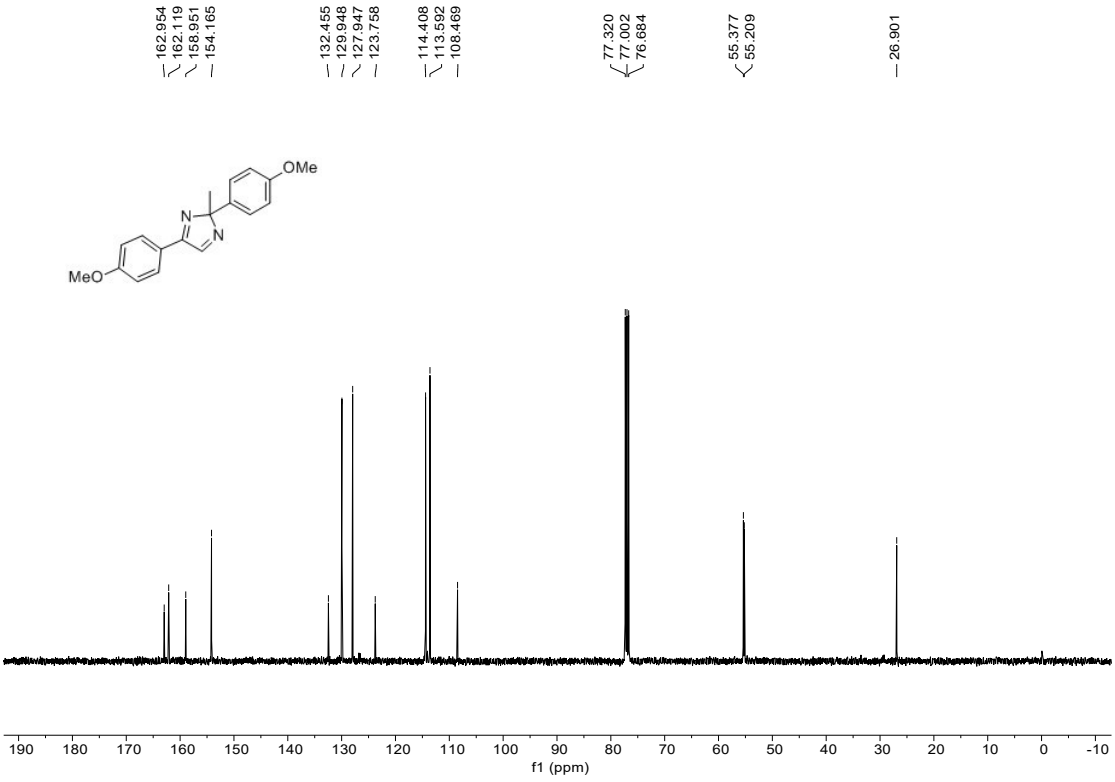
^{13}C NMR Spectrum of **4e**



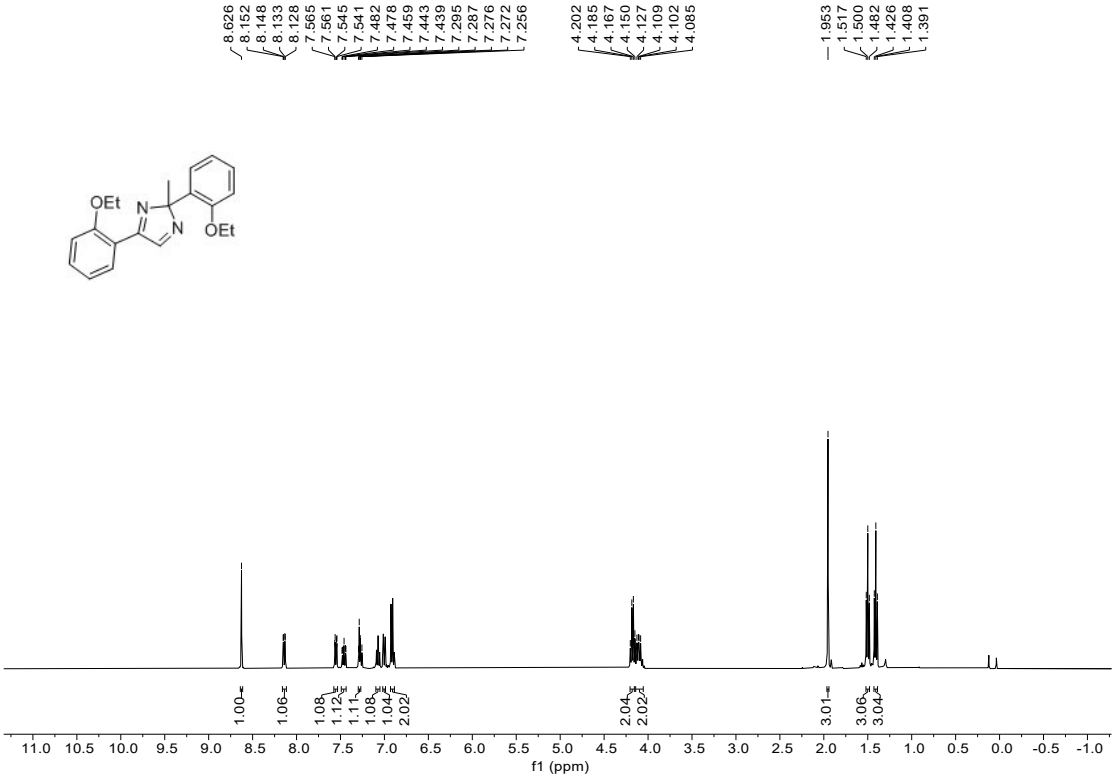
^{1}H NMR Spectrum of **4f**



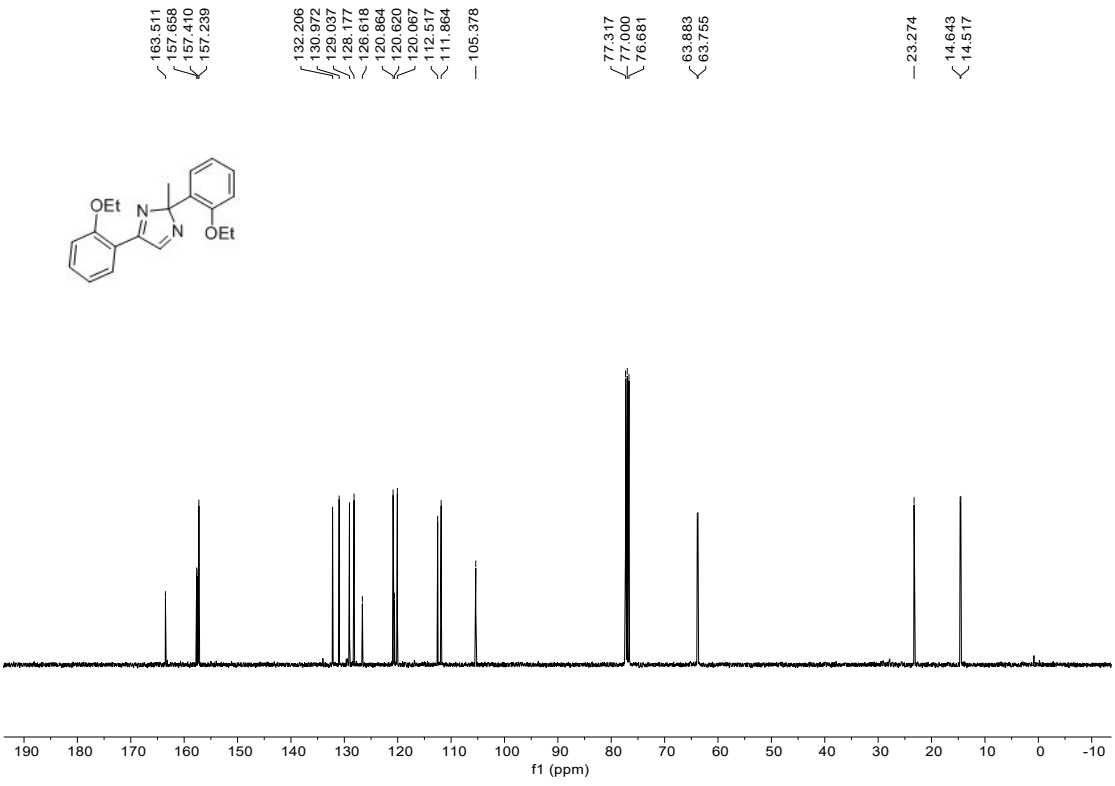
^{13}C NMR Spectrum of **4f**



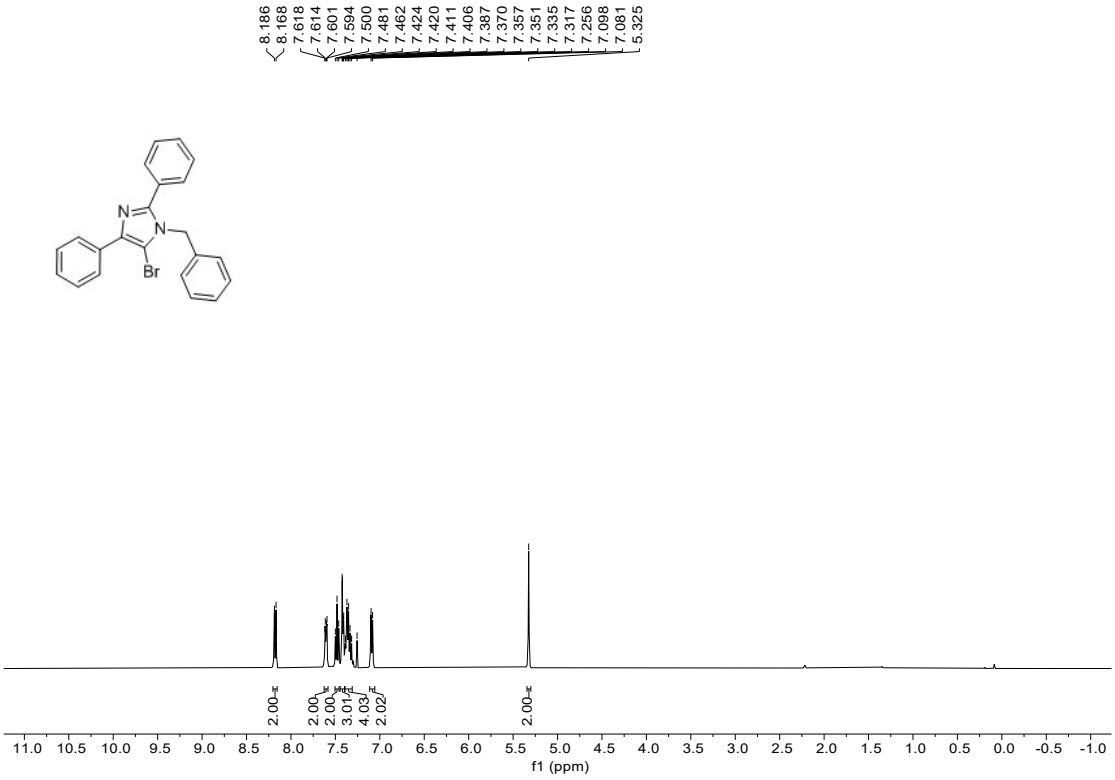
^{1}H NMR Spectrum of **4g**



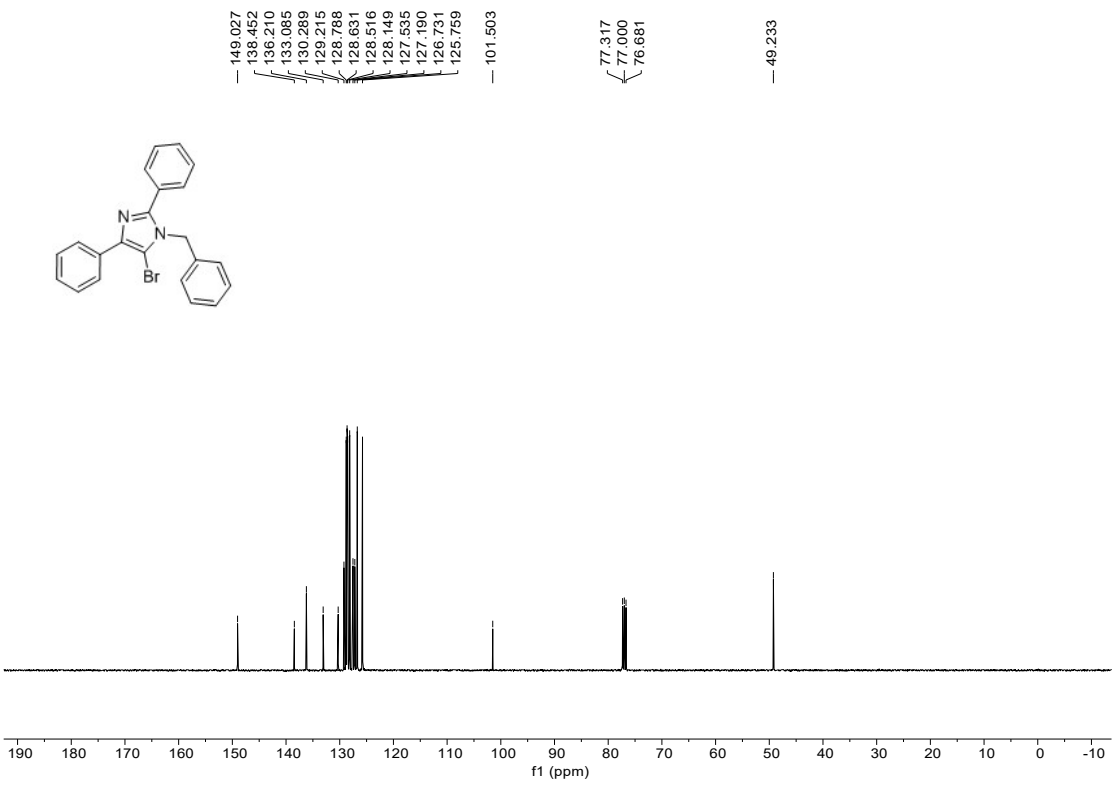
^{13}C NMR Spectrum of **4g**



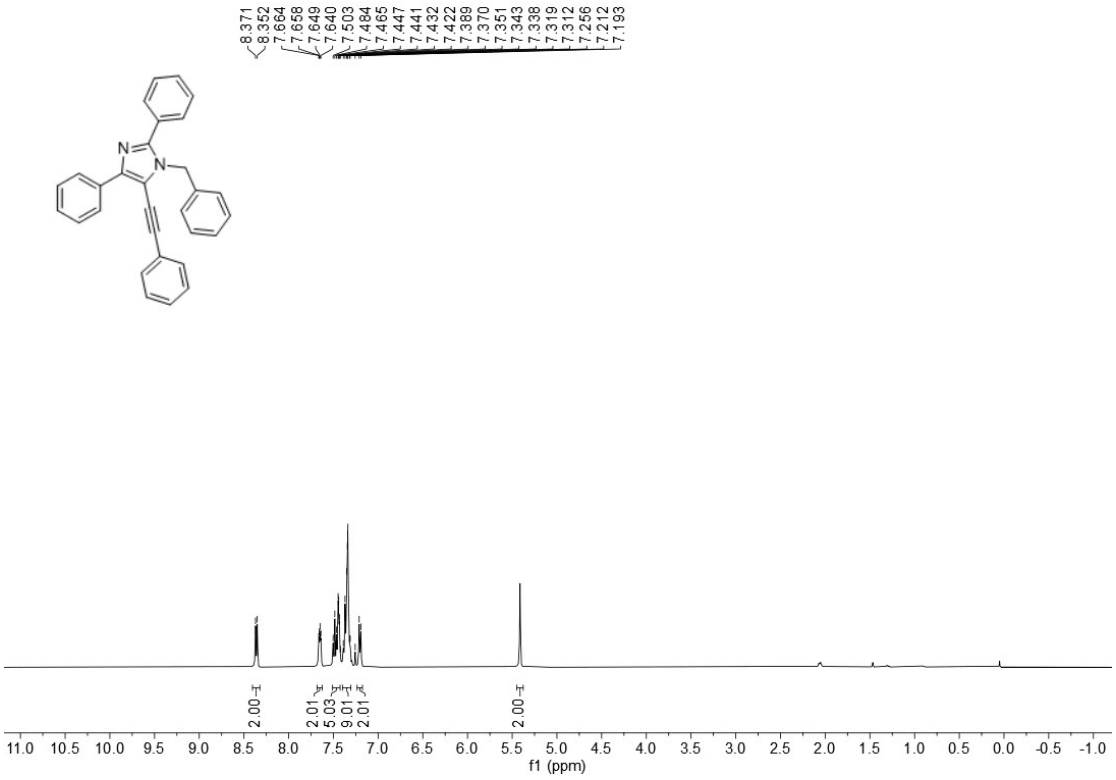
^{1}H NMR Spectrum of **5**



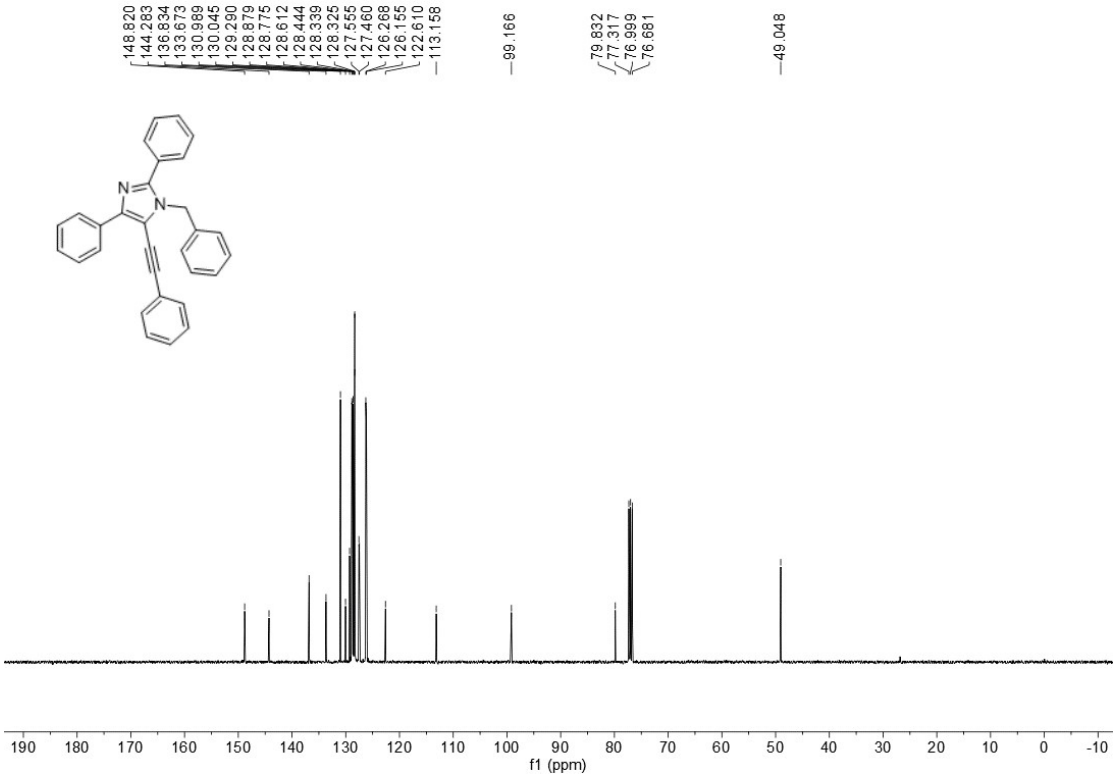
^{13}C NMR Spectrum of **5**



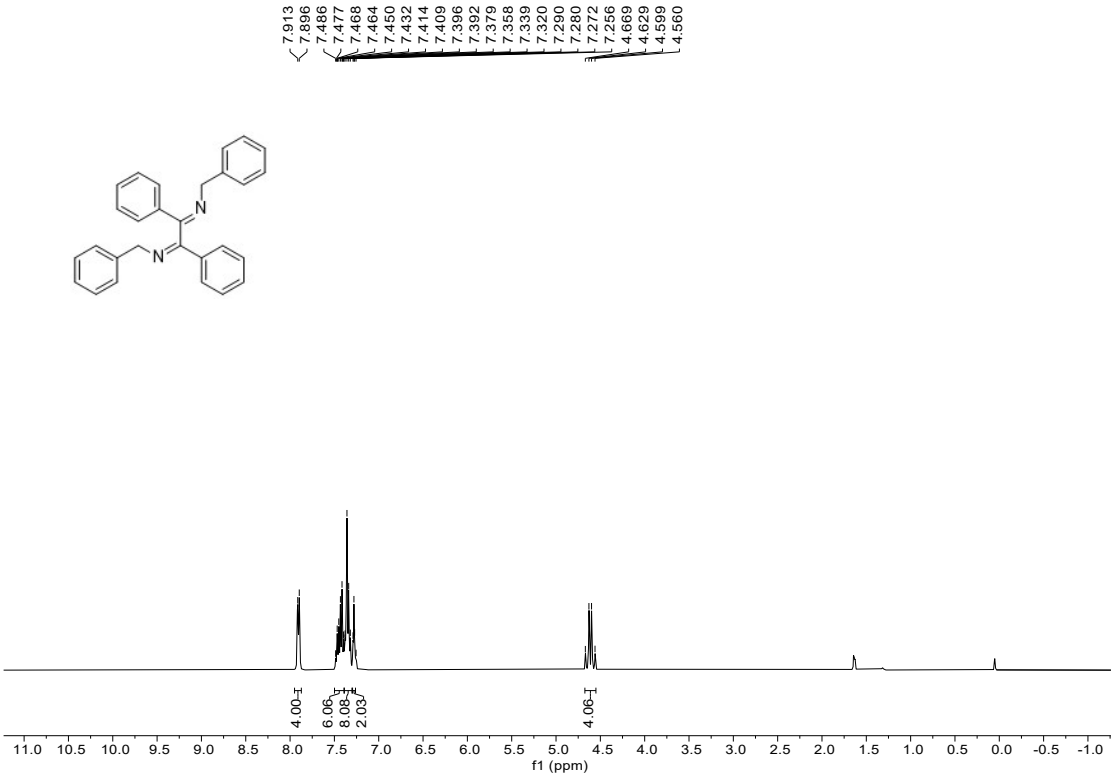
^{1}H NMR Spectrum of **6**



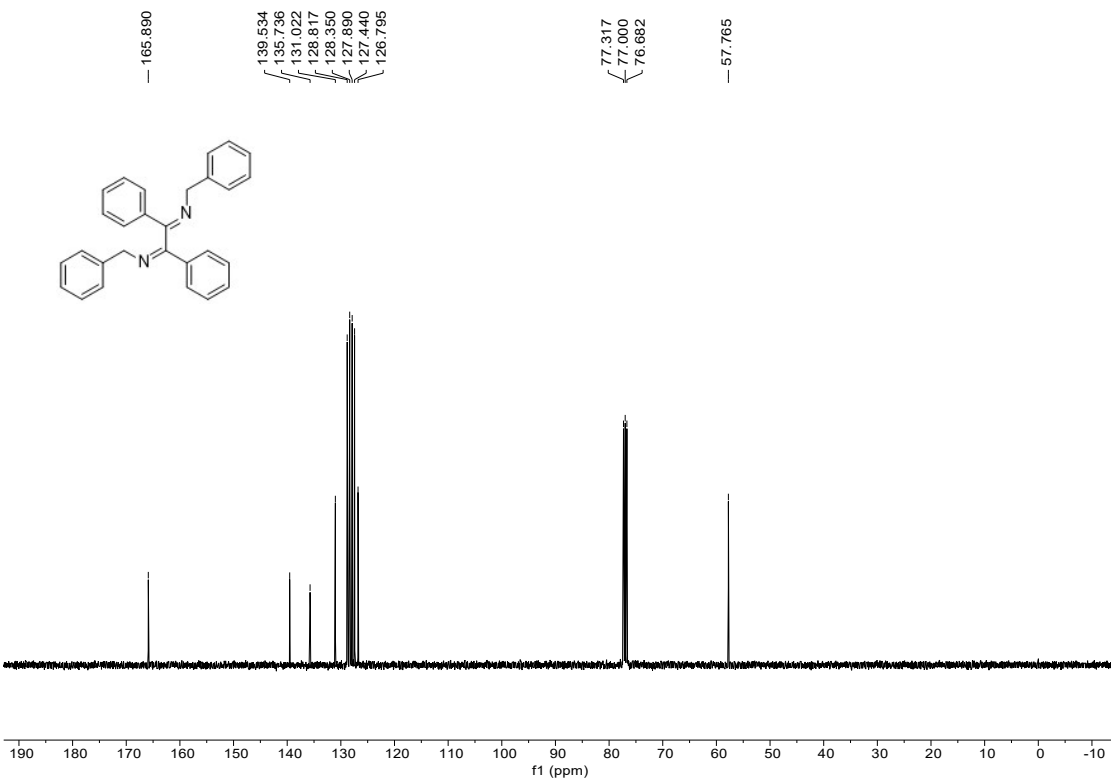
^{13}C NMR Spectrum of **6**



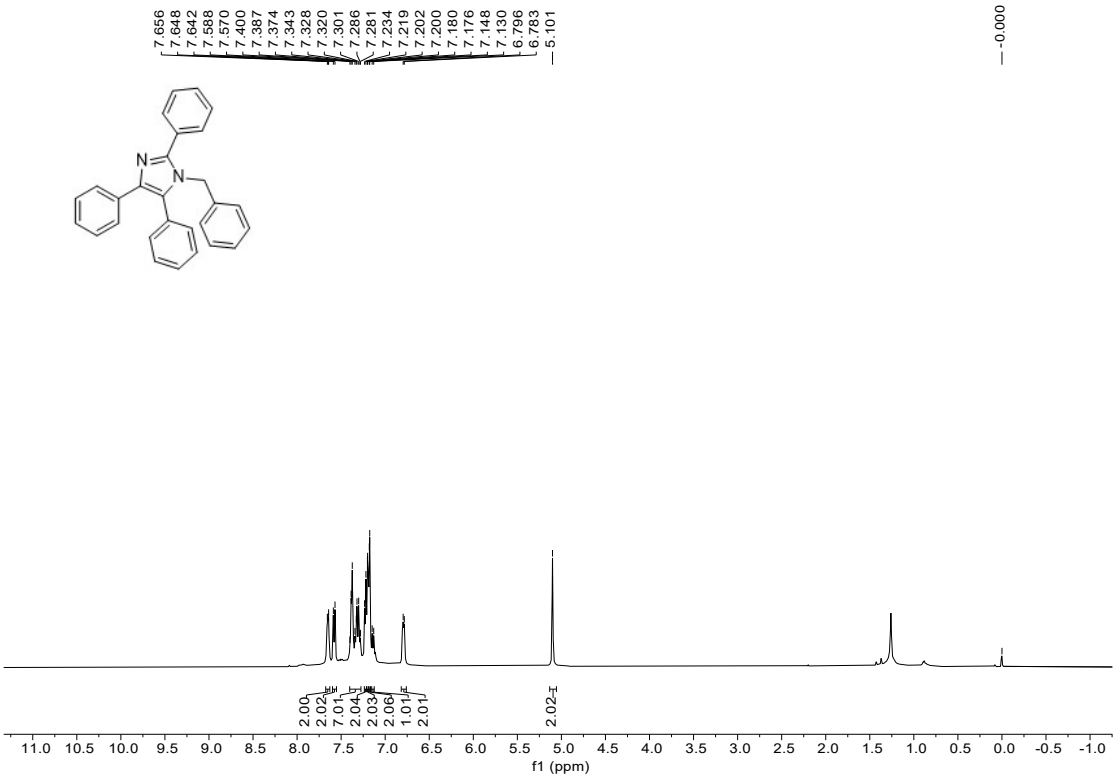
^{1}H NMR Spectrum of **12**



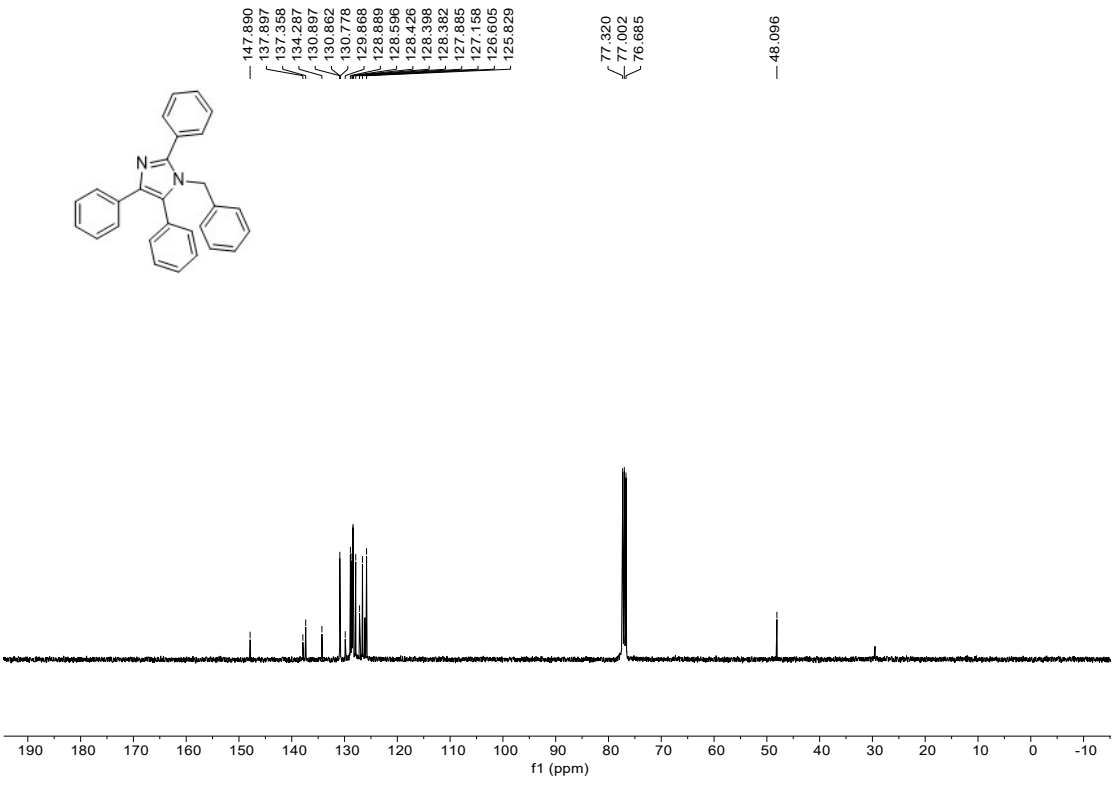
^{13}C NMR Spectrum of **12**



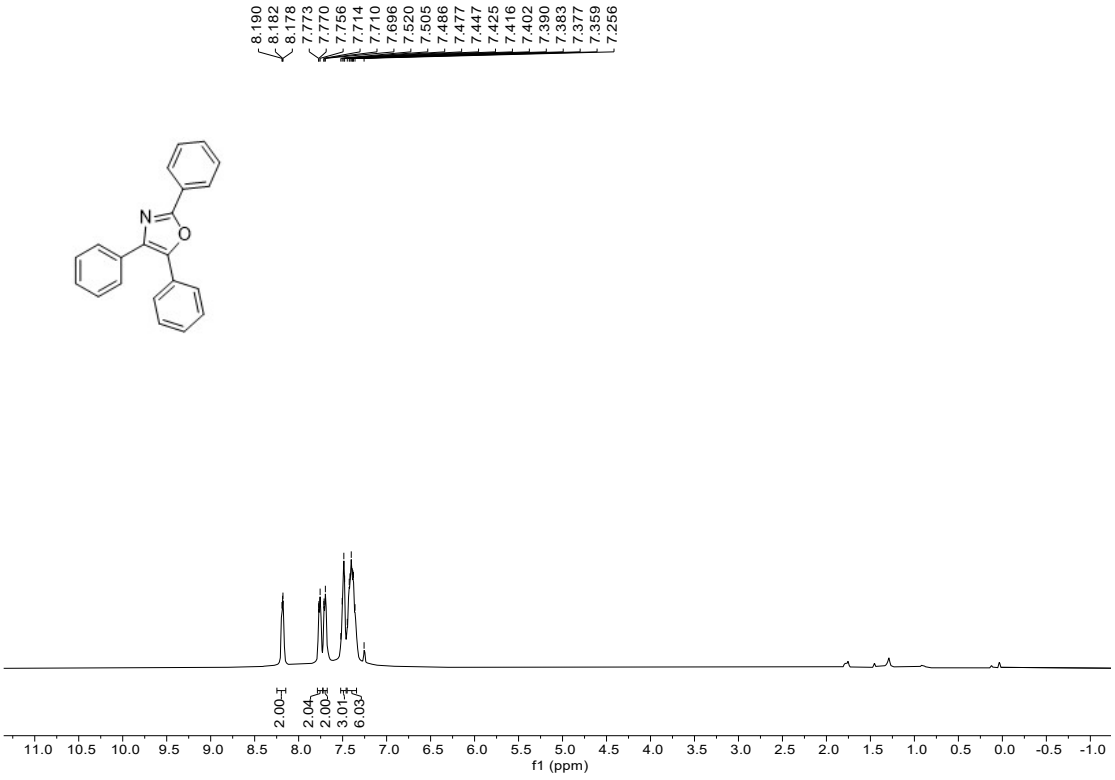
^{1}H NMR Spectrum of **13**



^{13}C NMR Spectrum of **13**



^{1}H NMR Spectrum of **15**



^{13}C NMR Spectrum of **15**