

## Electronic Supplementary Information

### Switchable Electrooxidative *N*-Methyl Amines: Access to C3-Aminomethylated and C3-Arylmethylated Imidazo[1,2-*a*]pyridines

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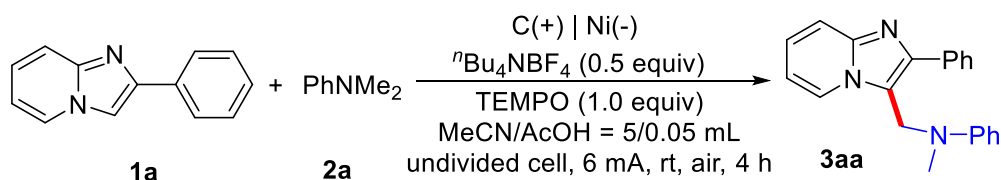
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## 1. General information

All the  $^1\text{H}$  NMR,  $^{13}\text{C}$  NMR and  $^{19}\text{F}$  NMR spectra of isolated compounds were recorded on JEOL 400 MHz spectrometer in  $\text{CDCl}_3$ . Chemical shifts ( $\delta$ ) were reported in ppm with tetramethylsilane as internal standard, and  $J$  values were given in Hz,  $\delta$  values relative to internal  $\text{CHCl}_3$  ( $\delta$  7.26 for  $^1\text{H}$  NMR and 77.16 for  $^{13}\text{C}$  NMR in  $\text{CDCl}_3$ ).  $^{19}\text{F}$  NMR chemical shifts were determined as  $\delta$  values relative to external standard  $\text{PhCF}_3$  at  $-63.0$ . High-resolution mass spectra (HRMS) were obtained on a 4G mass spectrometer by using electrospray ionization (ESI) analyzed by a quadrupole time-of-flight (QToF) instrument. Cyclic voltammetry was performed on CHI660E using the cyclic voltammetry mode. All melting points were measured with the samples after column chromatography and uncorrected. All reactions were monitored by thin layer chromatography (TLC), and column chromatography was carried out on 200-300 mesh of silica gel purchased from Qing Dao Hai Yang Chemical Industry Co. All the electrodes, cables and power supplies were purchased from online shopping platforms. Unless otherwise stated, all commercially available reagents and solvents were used directly without further purification. imidazo[1,2-*a*]pyridines **1**<sup>1-3</sup> were all known and prepared according to the previous reported protocols

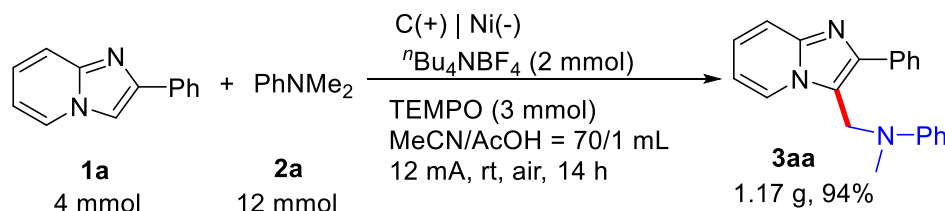
## 2. Typical procedure for electrochemical oxidative C3 aminomethylation



Imidazo[1,2-*a*]pyridine **1a** (0.2 mmol, 38.8 mg), *N,N*-dimethylaniline **2a** (1.0 mmol, 121 mg, 5.0 equiv.),  $t\text{Bu}_4\text{BF}_4$  (0.1 mmol, 33.0 mg, 0.5 equiv.), TEMPO (0.2 mmol, 31.2 mg, 1.0 equiv.), acetic acid (50  $\mu\text{L}$ ) and MeCN (5.0 mL) were added into a 10 mL tube equipped with a stir bar. The tube was equipped with a graphite plate (20.0 mm  $\times$  15.0 mm  $\times$  2.0 mm) anode and a nickel plate (20.0 mm  $\times$  10.0 mm  $\times$  0.2 mm) cathode, and the two electrodes were then submerged into the solution for 10 mm. The reaction mixture was then stirred and electrolyzed at a constant current of 6 mA at room temperature 4 h. After the completion of reaction as monitored by TLC, the resulting mixture was allowed to poured to saturated  $\text{NaHCO}_3$  (5 mL) before extracted by ethyl acetate (3  $\times$  10 mL). The organic phase was washed by saturated brine and then dried over  $\text{Na}_2\text{SO}_4$  before concentrated

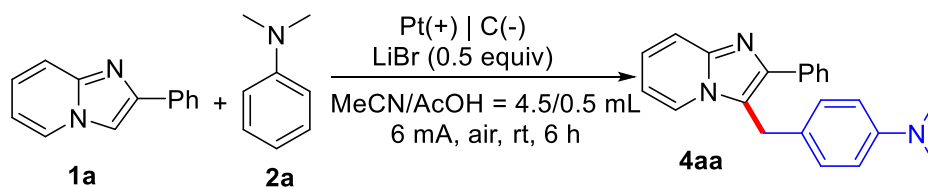
and dried under vacuum. The residue was then purified by silica gel column by using petroleum ether and ethyl acetate (petroleum ether/ethyl acetate = 5:1 to 3:1) as a mixed eluent to provide the desired products **3aa** (61 mg, 98%).

### 2.1 Gram-scale C3-aminomethylation reaction of 2-phenylimidazo[1,2-*a*]pyridine



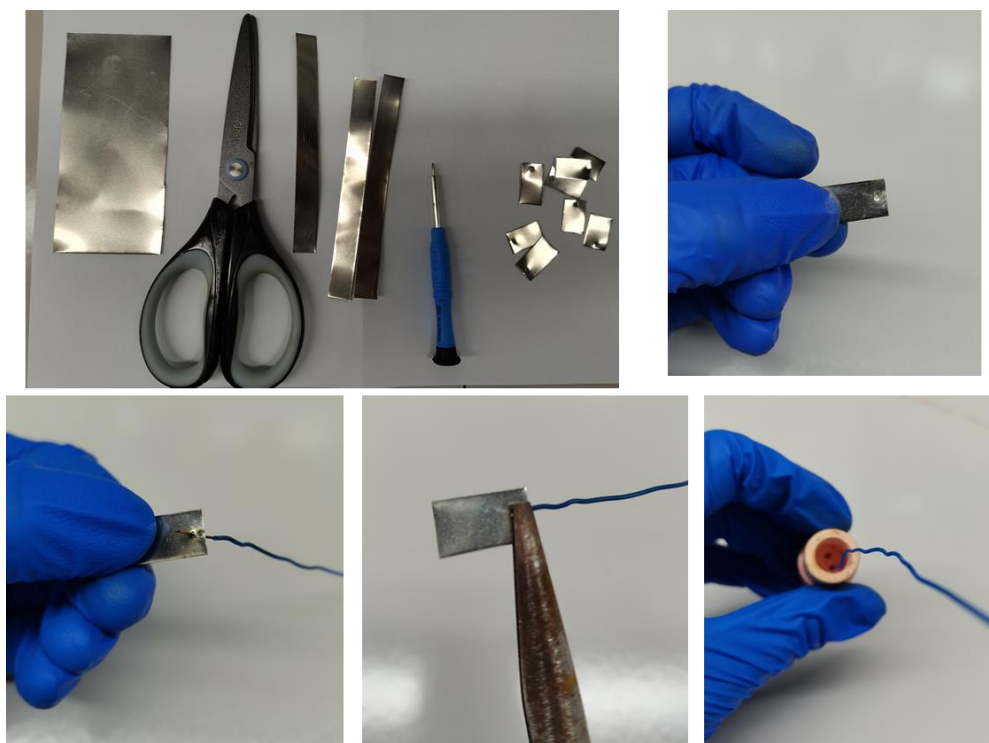
Imidazo[1,2-*a*]pyridine **1a** (4 mmol, 776 mg), *N,N*-dimethylaniline **2a** (12 mmol, 1.45 g, 3.0 equiv.),  $t\text{Bu}_4\text{BF}_4$  (3 mmol, 16.5 mg, 0.25 equiv.), TEMPO (3 mmol, 468 mg, 0.75 equiv.), acetic acid (1.0 mL) and MeCN (70 mL) were added into a 150 mL conical flask and a stir bar was added. The tube was equipped with a graphite plate (20.0 mm × 15.0 mm × 2.0 mm) anode and a nickel plate (20.0 mm × 10.0 mm × 0.2 mm) cathode, and the two electrodes were then submerged into the solution for 10 mm. The reaction mixture was then stirred and electrolyzed at a constant current of 12 mA at room temperature 14 h. After the completion of reaction as monitored by TLC, the resulting mixture was allowed to poured to saturated  $\text{NaHCO}_3$  (30 mL) before extracted by ethyl acetate (3 × 50 mL). The organic phase was washed by saturated brine and then dried over  $\text{Na}_2\text{SO}_4$  before concentrated and dried under vacuum. The residue was then purified by silica gel column by using petroleum ether and ethyl acetate (petroleum ether/ethyl acetate = 5:1 to 3:1) as a mixed eluent to provide the desired products **3aa** (1.17 g, 94%).

### 3. Typical procedure for electrochemical C3 *para*-selective arylmethylation

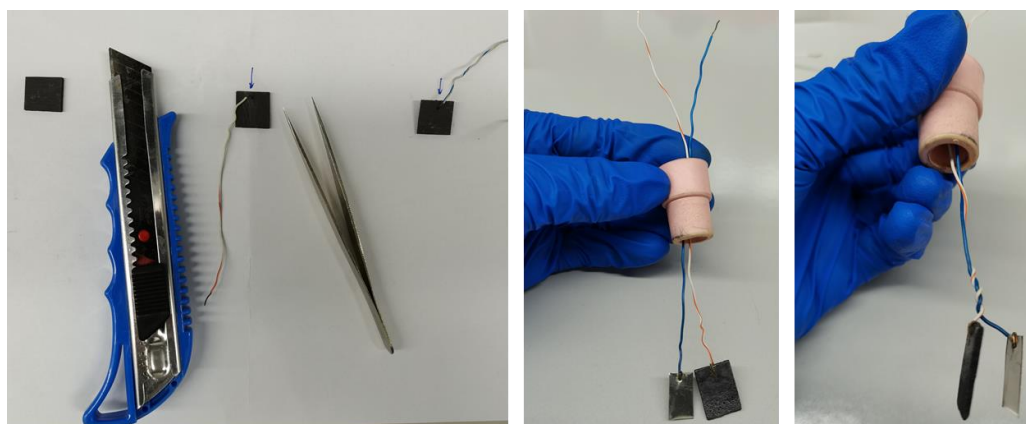


Imidazo[1,2-*a*]pyridine **1a** (0.2 mmol, 38.8 mg), *N,N*-dimethylaniline **2a** (1.0 mmol, 121 mg, 5.0 equiv.), LiBr (0.1 mmol, 8.7 mg, 0.5 equiv.), acetic acid (0.5 mL) and MeCN (4.5 mL) were added into a 10 mL tube equipped with a stir bar. The tube was equipped with a Pt plate (10.0 mm × 10.0 mm × 0.1 mm) anode and a graphite plate (20.0 mm × 15.0 mm × 2.0 mm) cathode, and the two

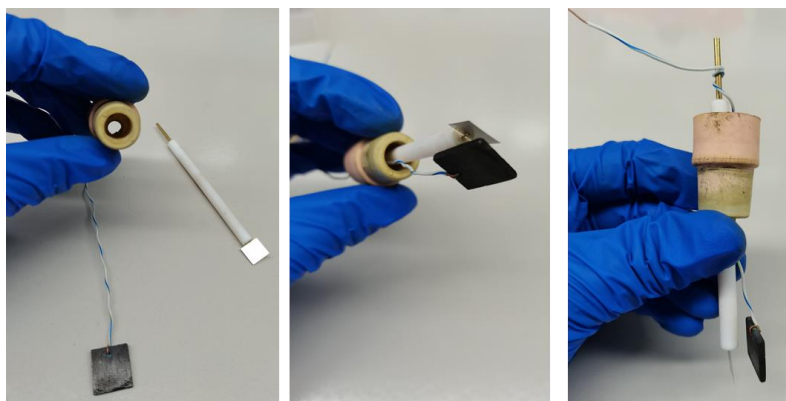
electrodes were then submerged into the solution for 10 mm. The reaction mixture was then stirred and electrolyzed at a constant current of 6 mA at room temperature 6 h. After the completion of reaction as monitored by TLC, the resulting mixture was allowed to poured to saturated  $\text{NaHCO}_3$  (5 mL) before extracted by ethyl acetate ( $3 \times 10$  mL). The organic phase was washed by saturated brine and then dried over  $\text{Na}_2\text{SO}_4$  before concentrated and dried under vacuum. The residue was then purified by silica gel column by using petroleum ether and ethyl acetate (petroleum ether/ethyl acetate = 10:1 to 5:1) as a mixed eluent to provide the desired products **4aa** (43 mg, 66%).



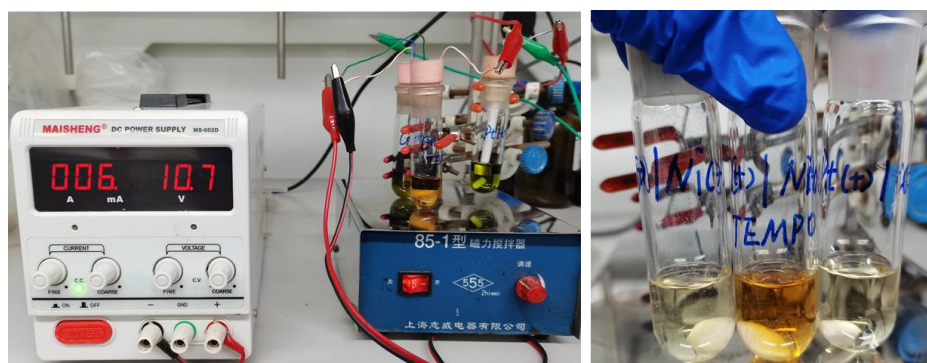
**The preparation of nickel cathode**



**The preparation of C(+) | Ni(-) electrodes**



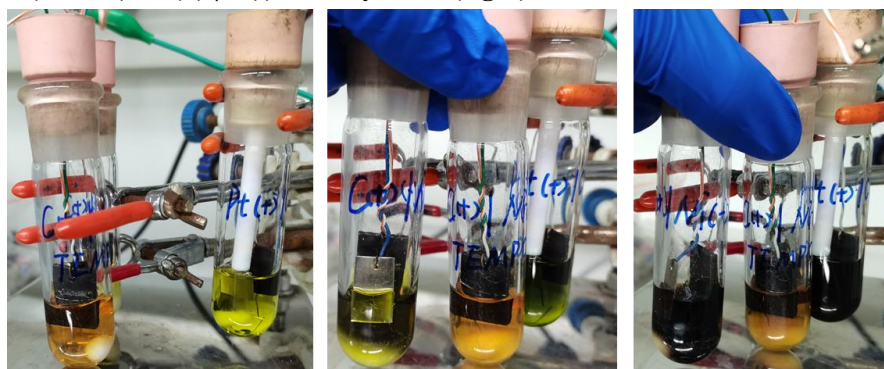
The preparation of Pt(+) | C(-) electrodes



Reaction setup (5 min)

0 h

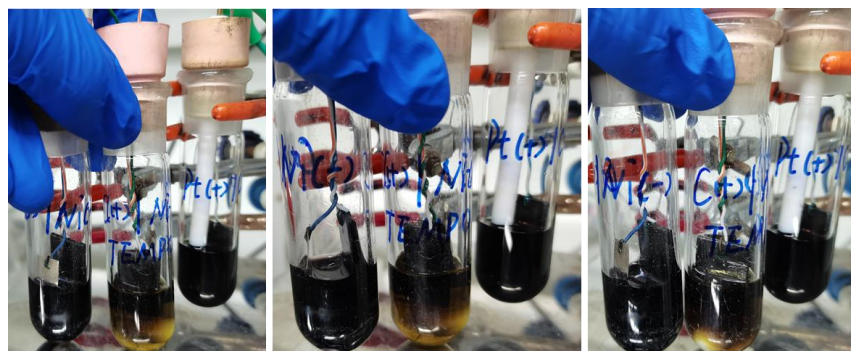
C(+) | Ni(-) for methylation without TEMPO (left); C(+) | Ni(-) for methylation in standard conditions (middle); Pt(+) | C(-) for alkylation (right).



1 min

5 min

30 min



2 h

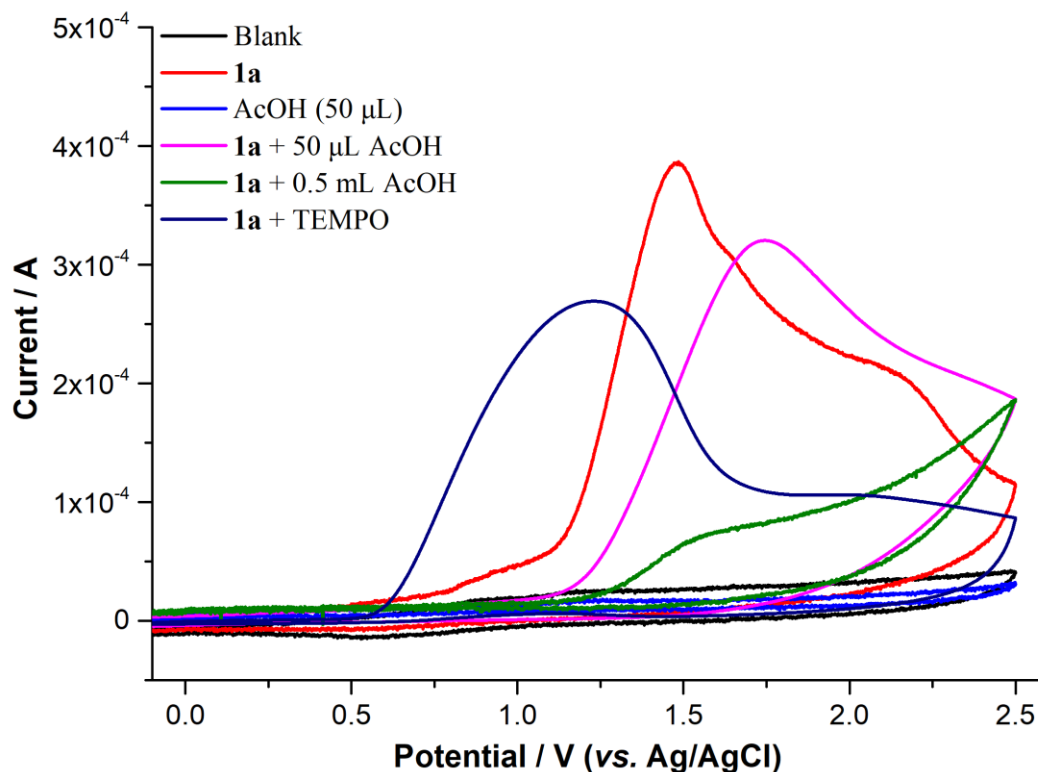
4 h

6 h

The reaction processes

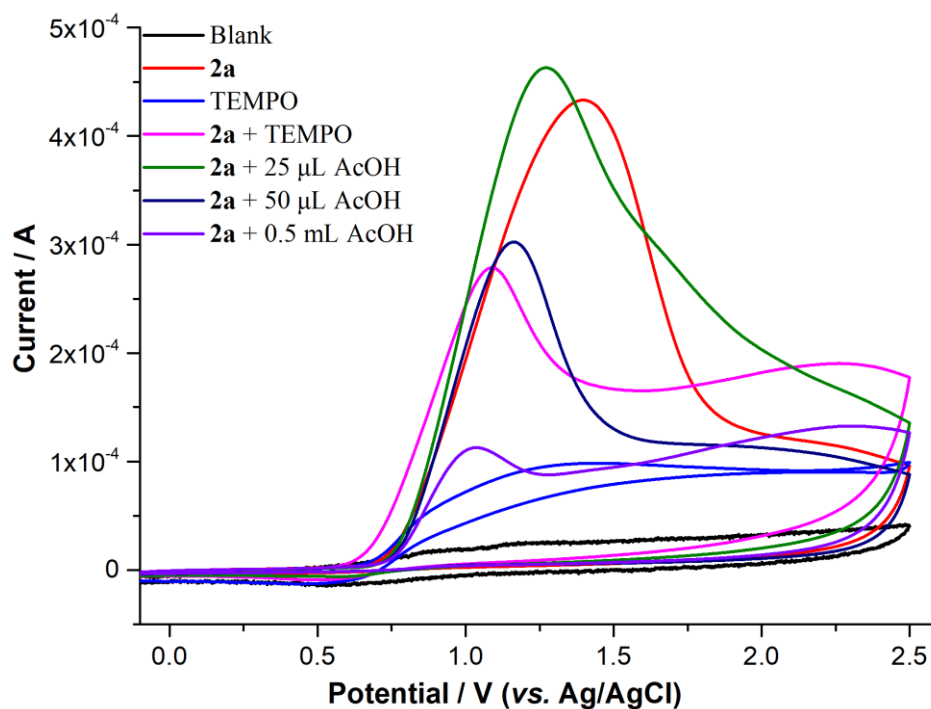
#### 4. Procedure for cyclic voltammetry (CV)

Cyclic voltammetry was performed on CHI660E using the cyclic voltammetry mode. A glassy carbon disc (diameter 3 mm) working electrode, a platinum plate (10 x 10 x 1 mm) counter electrode and an Ag wire (in saturated aqueous KCl solution) reference electrode were used at a scan rate of 50 mV/s. The experiments were conducted in a 10 mL tube without stirring in CH<sub>3</sub>CN (5 mL) or mixed solvent (CH<sub>3</sub>CN/AcOH = 4.5/0.5 mL) using <sup>t</sup>Bu<sub>4</sub>NBF<sub>4</sub> (0.1 mmol, 0.02 M) as electrolyte.



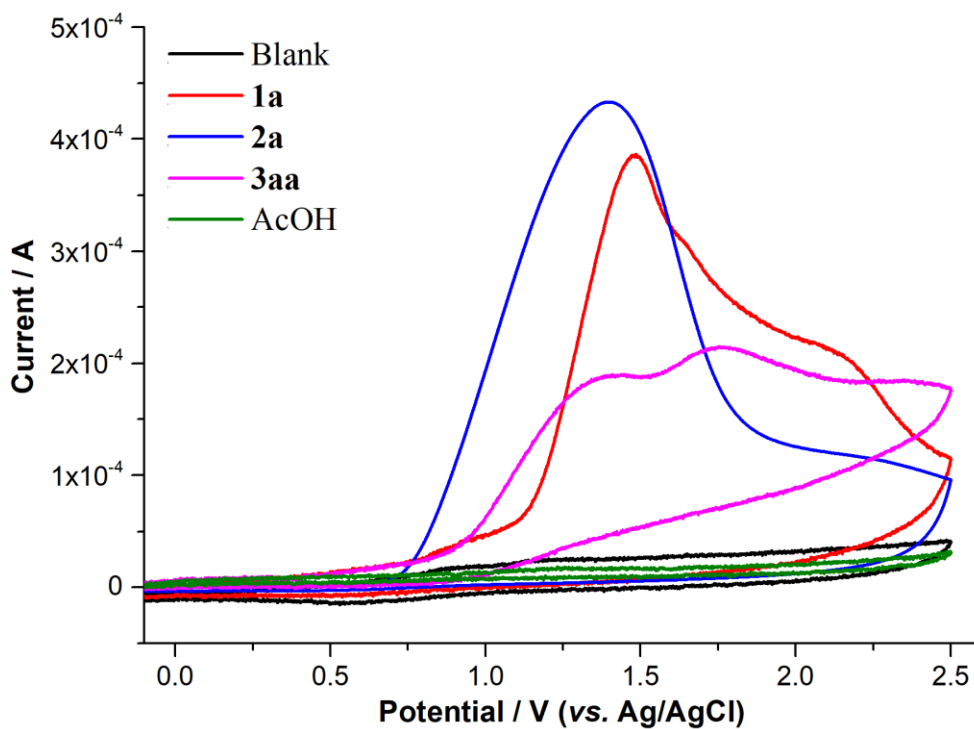
Cyclic voltammetry curves: (**1a**) 0.04 M **1a** in CH<sub>3</sub>CN; (AcOH) 50 μL acetic acid in CH<sub>3</sub>CN; (**1a** + 50 μL AcOH) 0.04 M **1a** and 50 μL AcOH in MeCN; (**1a** + 0.5 mL AcOH) 0.04 M **1a** in mixed solvent; (**1a** + TEMPO) 0.04 M **1a** and 0.02 M TEMPO in CH<sub>3</sub>CN.

Figure S1 Cyclic voltammetry experiments of **1a**



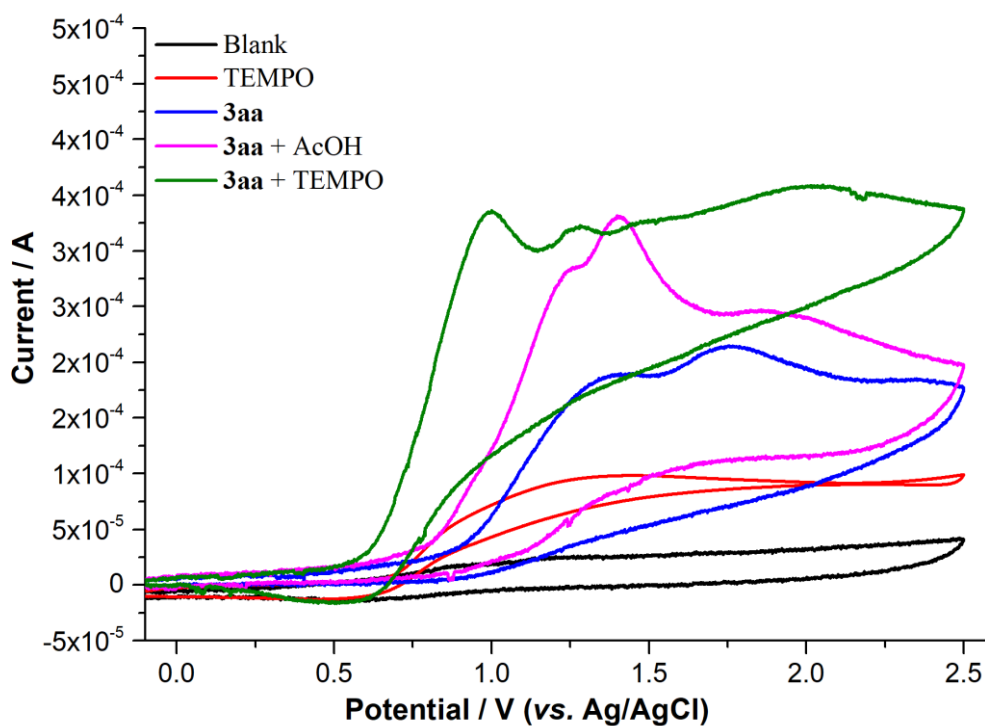
Cyclic voltammetry curves: (**2a**) 0.2 M **2a** in CH<sub>3</sub>CN; (TEMPO) 0.04 M TEMPO in CH<sub>3</sub>CN; (**2a** + TEMPO) 0.2 M **2a** and 0.04 M TEMPO in CH<sub>3</sub>CN; (**2a** + 50 μL AcOH) 0.2 M **2a** and 25 μL AcOH in MeCN; (**2a** + 25 μL AcOH) 0.2 M **2a** and 50 μL AcOH in MeCN; (**2a** + 0.5 mL AcOH) 0.2 M **2a** in mixed solvent.

Figure S2 Cyclic voltammetry experiments of **2a**



Cyclic voltammetry curves: (**1a**) 0.04 M **1a** in CH<sub>3</sub>CN; (**2a**) 0.2 M **2a** in MeCN; (**3aa**) 0.04 M **3aa** in CH<sub>3</sub>CN; (**3aa** + AcOH) 0.04 M **3aa** in mixed solvent; (AcOH) 0.5 mL AcOH in 4.5 mL CH<sub>3</sub>CN.

Figure S3 Cyclic voltammetry experiments of coupling partners

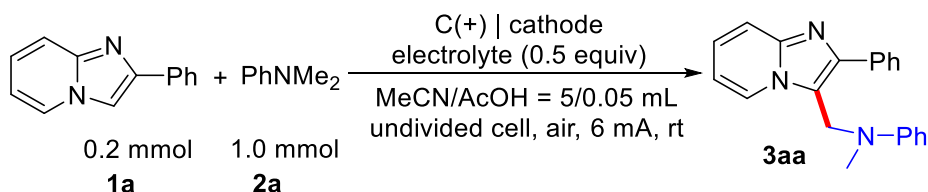


Cyclic voltammetry curves: (TEMPO) 0.04 M TEMPO in CH<sub>3</sub>CN; (**3aa**) 0.04 M **3aa** in CH<sub>3</sub>CN; (**3aa** + AcOH) 0.04 M **3aa** in mixed solvent; (**3aa** + TEMPO) 0.04 M **3aa** and 0.04 M TEMPO in CH<sub>3</sub>CN.

Figure S4 Cyclic voltammetry experiments of **3aa**

## 5. The optimization of reaction conditions

Table S1 Optimization of reaction conditions for the aminomethylation



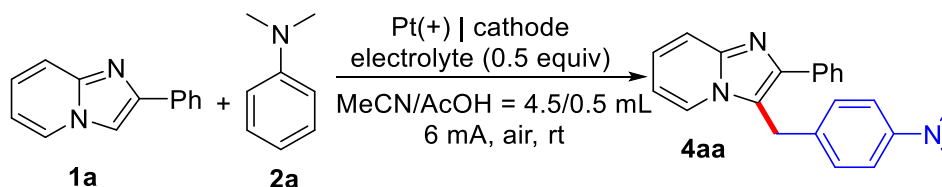
entry	electrolyte	cathode	time (h)	yield (%) <sup>[b]</sup>
1	none	Ni	12	41
2	<sup>t</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	8	63
3 <sup>[c]</sup>	<sup>t</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	6	56
4	<sup>t</sup> Bu <sub>4</sub> NPF <sub>6</sub>	Ni	12	46
5	<sup>t</sup> Bu <sub>4</sub> NClO <sub>4</sub>	Ni	12	34
6	<sup>t</sup> Bu <sub>4</sub> NI	Ni	12	37
7	<sup>t</sup> Bu <sub>4</sub> NBr	Ni	12	45
8	<sup>t</sup> Bu <sub>4</sub> NHSO <sub>4</sub>	Ni	12	N.R.
9	Et <sub>4</sub> NBF <sub>4</sub>	Ni	12	46



<b>10</b> <sup>[d]</sup>	CAN	Ni	12	39
<b>11</b> <sup>[d]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Fe	12	42
<b>12</b> <sup>[d]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Al	12	27
<b>13</b>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Graphite	12	44
<b>14</b>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Pt	12	10
<b>15</b> <sup>[e]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	8	62
<b>16</b> <sup>[f]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	24	trace
<b>17</b> <sup>[g]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	4	98
<b>18</b> <sup>[h]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	Ni	4	87

[a] Reaction conditions: graphite anode, cathode, 2-phenyl imidazo[1,2-*a*]pyridine **1a** (0.2 mmol), *N,N*-dimethylaniline **2a** (1.0 mmol), electrolyte (0.1 mmol) and AcOH (50  $\mu$ L) were dissolved in 5 mL solvent, the mixture was stirred under air at room temperature for specific time, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-Methylanisole as the standard. [c] 0.2 mmol <sup>n</sup>Bu<sub>4</sub>BF<sub>4</sub> was used. [d] An unexpected product **4aa** was isolated. [e] The reaction was conducted under N<sub>2</sub>. [f] 0.2 mmol 2,6-Di-*tert*-butyl-4-methylphenol (BHT) was added. [g] 0.2 mmol TEMPO was added. [h] 0.05 mmol TEMPO was added. CAN = Diammonium cerium(IV) nitrate. TEMPO = 2,2,6,6-tetramethylpiperidinyl-1-oxide.

Table S2 Optimization of reaction conditions for arylmethylation

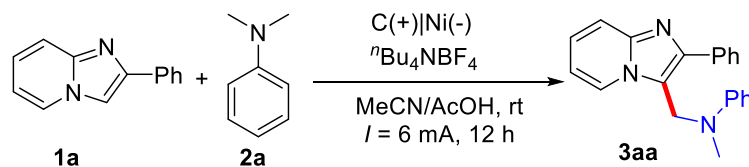


entry	electrolyte	cathode	time (h)	yield (%) <sup>[b]</sup>
<b>1</b>	none	graphite	8	23
<b>2</b>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	graphite	8	40
<b>3</b> <sup>[c]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	graphite	8	37
<b>4</b> <sup>[d]</sup>	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	graphite	8	21
<b>5</b> <sup>[e]</sup>	<sup>n</sup> Bu <sub>4</sub> NPF <sub>6</sub>	graphite	8	43
<b>6</b>	<sup>n</sup> Bu <sub>4</sub> NClO <sub>4</sub>	graphite	8	30
<b>7</b>	<sup>n</sup> Bu <sub>4</sub> NI	graphite	8	39
<b>8</b>	<sup>n</sup> Bu <sub>4</sub> NBr	graphite	8	trace
<b>9</b> <sup>[e]</sup>	<sup>n</sup> Bu <sub>4</sub> NHSO <sub>4</sub>	graphite	8	43
<b>10</b>	Et <sub>4</sub> NBF <sub>4</sub>	graphite	8	42

<b>11</b>	LiBr	graphite	6	66
<b>12<sup>[f]</sup></b>	LiBr	graphite	6	67
<b>13</b>	LiBr	Fe	8	36
<b>14<sup>[e]</sup></b>	LiBr	Ni	8	40
<b>15</b>	LiBr	Pt	8	47
<b>16<sup>[g]</sup></b>	LiBr	graphite	12	21
<b>17<sup>[h]</sup></b>	LiBr	graphite	12	69
<b>18<sup>[i]</sup></b>	LiBr	graphite	12	67

[a] Reaction conditions: Pt anode, cathode, 2-phenyl imidazo[1,2-*a*]pyridine **1a** (0.2 mmol), *N,N*-dimethylaniline **2a** (1.0 mmol), electrolyte (0.1 mmol) and AcOH (0.5 mL) were dissolved in 4.5 mL solvent, the mixture was stirred under air at room temperature for specific time, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-Methylanisole as the standard. [c] 200 μL AcOH was used. [d] 1.0 mL AcOH was used. [e] Product **3aa** was detected in <sup>1</sup>H NMR. [f] The reaction was conducted under N<sub>2</sub>. [g] 0.2 mmol 2,6-Di-*tert*-butyl-4-methylphenol (BHT) was added. [h] 0.2 mmol TEMPO was added. [i] 0.05 mmol TEMPO was added.

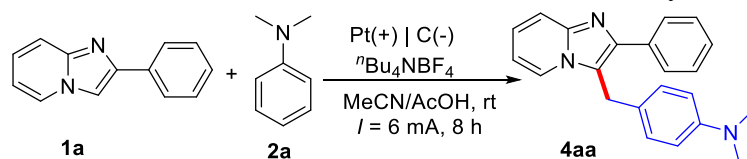
Table S3 The effect of acetic acid and <sup>n</sup>Bu<sub>4</sub>NBF<sub>4</sub> for the aminomethylation



entry	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub> loading	amounts of AcOH	Yield (%) <sup>[b]</sup>
<b>1</b>	1 equiv	none	trace
<b>2<sup>[c]</sup></b>	1 equiv	25 μL	21
<b>3</b>	1 equiv	50 μL	56
<b>4<sup>[c]</sup></b>	1 equiv	100 μL	32
<b>5</b>	none	50 μL	41
<b>6</b>	0.25 equiv	50 μL	47
<b>7</b>	0.5 equiv	50 μL	63
<b>8</b>	1.5 equiv	50 μL	53

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, specific amounts of <sup>n</sup>Bu<sub>4</sub>NBF<sub>4</sub> and AcOH were dissolved in 5 mL acetonitrile, the mixture was stirred under air at room temperature for 12 h, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-Methylanisole as the standard. [c] The reaction was not complete.

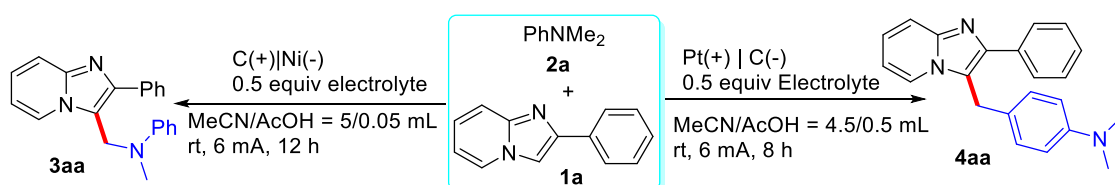
Table S4 The effect of acetic acid and <sup>n</sup>Bu<sub>4</sub>NBF<sub>4</sub> for the arylmethylation



entry	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub> loading	amounts of AcOH	Yield (%) <sup>[b]</sup>
1 <sup>[c]</sup>	1 equiv	50 μL	N.P.
2 <sup>[d]</sup>	1 equiv	100 μL	trace
3 <sup>[e]</sup>	1 equiv	200 μL	21
4	1 equiv	0.5 mL	39
5	1 equiv	1.0 mL	37
6	1 equiv	1.5 mL	32
7	none	0.5 mL	23
8	0.25 equiv	0.5 mL	34
9	0.5 equiv	0.5 mL	40
10	2.0 equiv	0.5 mL	39

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, specific amounts of <sup>n</sup>Bu<sub>4</sub>NBF<sub>4</sub> and AcOH were dissolved in 5 mL acetonitrile, the mixture was stirred under air at room temperature for 12 h, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-Methylanisole as the standard. [c] 11% **3aa** was detected. [d] 18% **3aa** was detected. [e] 29% **3aa** was detected. N.P. = No product.

Table S5 The effects of electrolytes

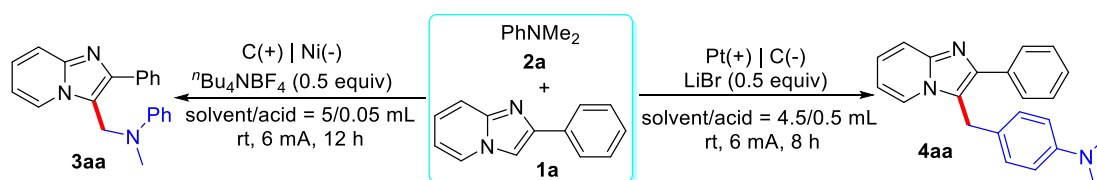


entry	electrolyte	Yield <b>3aa</b> (%) <sup>[b]</sup>	Yield <b>4aa</b> (%) <sup>[b]</sup>
1	<sup>n</sup> Bu <sub>4</sub> NBF <sub>4</sub>	63 (61)	40
2	<sup>n</sup> Bu <sub>4</sub> NPF <sub>6</sub>	46	43; 27 <sup>[c]</sup>
3	<sup>n</sup> Bu <sub>4</sub> NClO <sub>4</sub>	34	30
4	<sup>n</sup> Bu <sub>4</sub> NI	37	39
5 <sup>[c]</sup>	<sup>n</sup> Bu <sub>4</sub> NBr	45	trace
6	<sup>n</sup> Bu <sub>4</sub> NF·3H <sub>2</sub> O	trace	57; 21 <sup>[c]</sup>
7	<sup>n</sup> Bu <sub>4</sub> NHSO <sub>4</sub>	n.r.	43; 12 <sup>[c]</sup>
8	<sup>n</sup> Bu <sub>4</sub> NOAc	34	trace
9	<sup>n</sup> Bu <sub>4</sub> NBr <sub>3</sub>	trace	15
10	<sup>n</sup> Bu <sub>4</sub> PBr	trace	45

11	Et <sub>4</sub> NBF <sub>4</sub>	46	42
12	NH <sub>4</sub> PF <sub>6</sub>	trace	51; 4 <sup>[c]</sup>
13	Et <sub>4</sub> NOTs	21	46; 5 <sup>[c]</sup>
14	Et <sub>4</sub> NCl	trace	27; 12 <sup>[c]</sup>
15	Et <sub>4</sub> NClO <sub>4</sub>	38	41; 7 <sup>[c]</sup>
16	NaOPiv·H <sub>2</sub> O	36	6
17 <sup>[e]</sup>	NH <sub>4</sub> I	15	16
18	NH <sub>4</sub> OAc	n.r.	30; 18 <sup>[c]</sup>
19	KPF <sub>6</sub>	41	31
20	LiBr	trace	66 (64)
21	Li <sub>2</sub> CO <sub>3</sub>	trace	43
22	KI	42	trace
23	CAN	39 <sup>[c]</sup>	20
24	[BMIM]PF <sub>6</sub>	43	trace
25	AcONa	trace	46; 20 <sup>[c]</sup>

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, 0.1 mmol electrolyte and 50 μL /0.5 mL AcOH were dissolved in 5/4.5 mL acetonitrile, the mixture was stirred under air at room temperature for 12 h, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-methylanisole as the standard. Isolated yield was presented in parenthesis. [c] **3aa** was detected in <sup>1</sup>H NMR. [d] The reaction was not complete. [e] **4aa** was detected in <sup>1</sup>H NMR.

Table S6 The effect of aliphatic acid and solvent

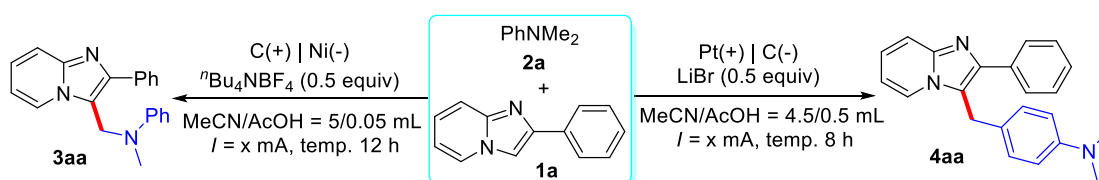


entry	Solvent	acid	Yield <b>3aa</b> (%) <sup>[b]</sup>	Yield <b>4aa</b> (%) <sup>[b]</sup>
1	MeCN	AcOH	63 (62)	66 (64)
2	MeCN	propionic acid	43	37, 31 <sup>[c]</sup>
3	MeCN	butyric acid	trace <sup>[d]</sup>	5, 30% <sup>[c]</sup>
4	MeCN	isobutyric acid	n.r.	n.p., 48 <sup>[c]</sup>
5	MeCN	pivalic acid	n.r.	n.p., 52 <sup>[c]</sup>
6	MeCN	CH <sub>3</sub> SO <sub>3</sub> H	n.p. 8 <sup>[c]</sup>	n.p., 8% dimer
7	DME	AcOH	trace	n.p. 23 <sup>[c]</sup>
8	DCE	AcOH	trace	36, 11 <sup>[c]</sup>
9	DCM	AcOH	trace	trace

10	DMSO	AcOH	trace	trace
11	DMF	AcOH	n.r.	n.p. 15 <sup>[c]</sup>
12	THF	AcOH	trace	n.p. 15 <sup>[c]</sup>
13	MeOH	AcOH	Trace	trace, 59 <sup>[c]</sup> (57)
14	EtOH	AcOH	trace	trace

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, 0.1 mmol electrolyte and 50  $\mu$ L /0.5 mL AcOH were dissolved in 5/4.5 mL acetonitrile, the mixture was stirred under air at room temperature for 12 h, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-methylanisole as the standard. Isolated yield was presented in parenthesis. [c] **3aa** was detected in <sup>1</sup>H NMR. [d] The reaction was not complete. [e] **4aa** was detected in <sup>1</sup>H NMR. n.r.= no reaction. n.p. = no product.

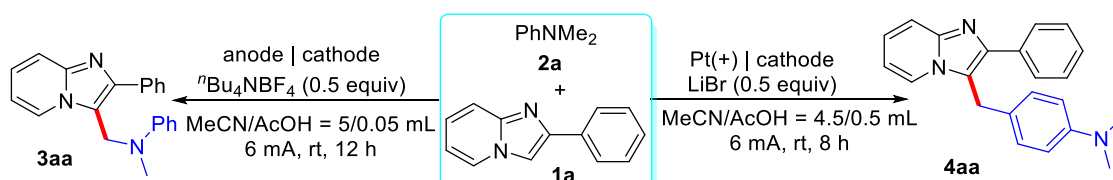
Table S7 The effect of current and temperature



entry	x	Temp.	Yield 3aa (%) <sup>[b]</sup>	Yield 4aa (%) <sup>[b]</sup>
1	4	r.t.	62	42
2	6	r.t.	63	66
3	8	r.t.	60	49
4	10	r.t.	60	42
5	6	50	54	63
6	6	70	47	41

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, 0.1 mmol electrolyte and 50  $\mu$ L /0.5 mL AcOH were dissolved in 5/4.5 mL acetonitrile, the mixture was stirred under air at room temperature, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-methylanisole as the standard.

Table S8 The effect of electrode

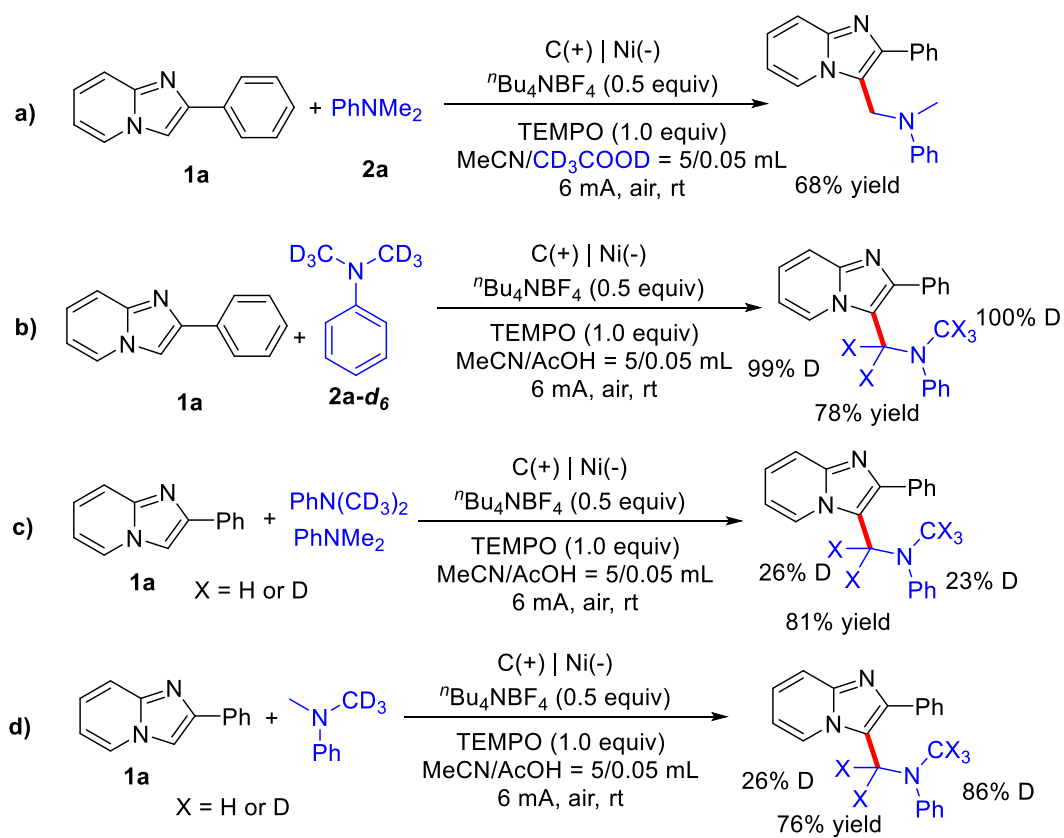


entry	Anode	Cathode	Yield 3aa (%) <sup>[b]</sup>	Cathode	Yield 4aa (%) <sup>[b]</sup>
1	Graphite	Ni	63	Graphite	66
2	Graphite	Ni foam	8	RVC	45

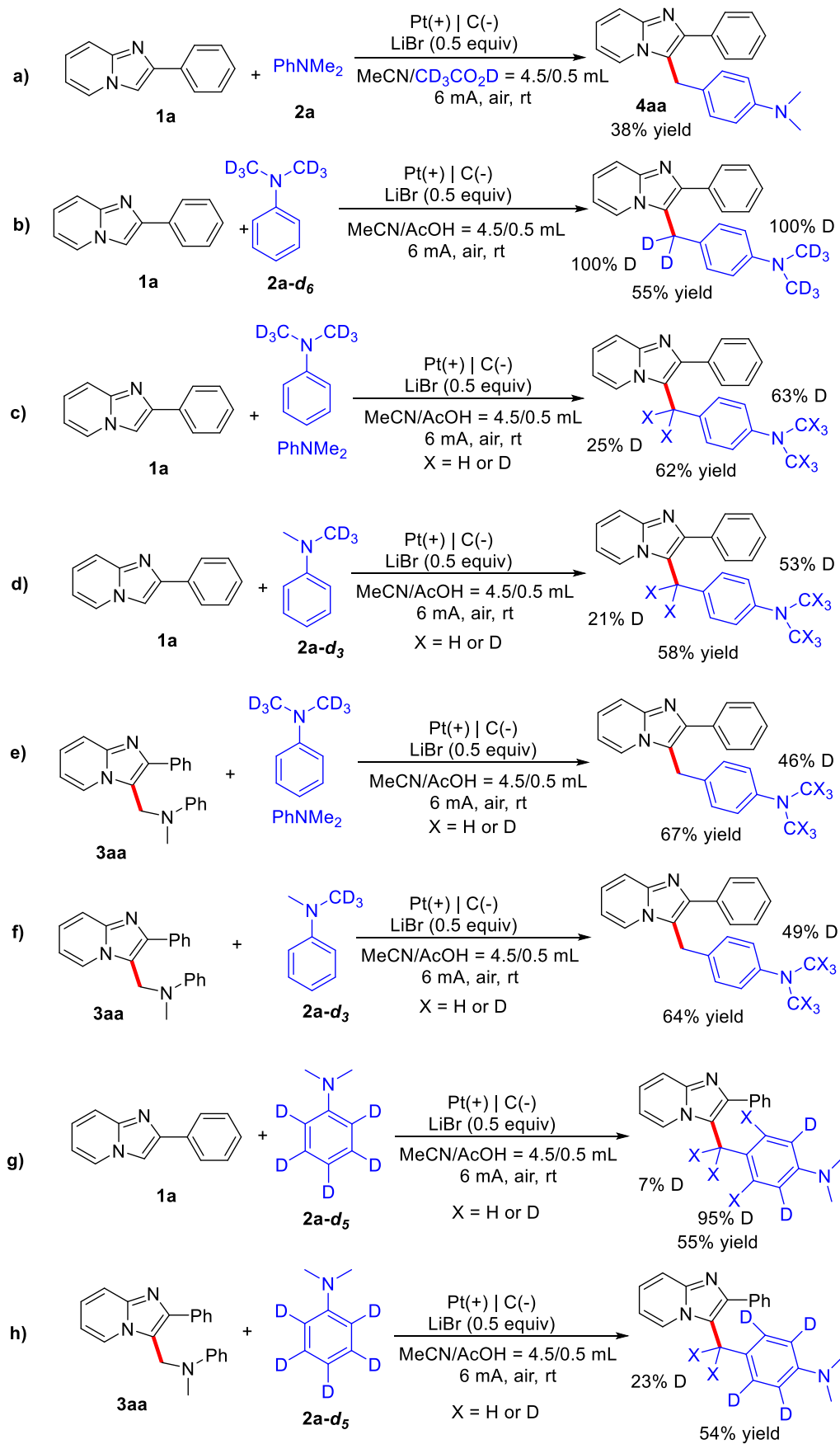
3	Graphite	Fe	42; 12 <sup>[c]</sup>	C paper	49
4	Graphite	SS	45	C foam	34
5	Graphite	Pt	10	C felt	trace
6	Graphite	Cu	trace <sup>[d]</sup>	Fe	36
7	Graphite	Al	27; 8 <sup>[c]</sup>	SS	40
8	Graphite	Zn	10	Ni	40; 20 <sup>[e]</sup>
9	Graphite	Sn	Trace <sup>[d]</sup>	Ni foam	Trace <sup>[d]</sup>
10	Graphite	Nb	5	Cu	30; 20 <sup>[e]</sup>
11	Graphite	W	46	Cu foam	trace
12	Graphite	Ti	trace	Pt	47
13	Graphite	Pb	n.p.	Al	37
14	Graphite	Mo	trace	Zn	35
15	Graphite	Graphite	44	Mg	53
16	Graphite	RVC	41	Sn	45
17	Graphite	C paper	21	Nb	38
18	Graphite	C foam	13	W	trace
19	Graphite	C felt	23	Ti	51
20	RVC	Ni	26	Pb	63
21	C paper	Ni	47	Mo	54
22	C foam	Ni	34	Graphite	67 <sup>[f]</sup>
23	C felt	Ni	51		
24	Graphite	Ni	62 <sup>[f]</sup>		

[a] Reaction condition: graphite anode, nickel cathode, 0.2 mmol **1a**, 1.0 mmol **2a**, 0.1 mmol electrolyte and 50  $\mu$ L /0.5 mL AcOH were dissolved in 5/4.5 mL acetonitrile, the mixture was stirred under air at room temperature for 12 h, constant current = 6 mA. [b] Yield was determined by <sup>1</sup>H NMR with 3-methylanisole as the standard. [c] **4aa** was detected in <sup>1</sup>H NMR. [d] The reaction was not complete. [e] **3aa** was detected in <sup>1</sup>H NMR. [f] The reaction was conducted under N<sub>2</sub>. n.r.= no reaction. n.p. = no product. RVC = reticulated glass carbon. SS = stainless steel.

## 6. Deuterium-labeling experiments



Scheme S1 The deuterium-labelling experiment for **3aa**



Scheme S2 The deuterium-labelling experiment for **4aa**



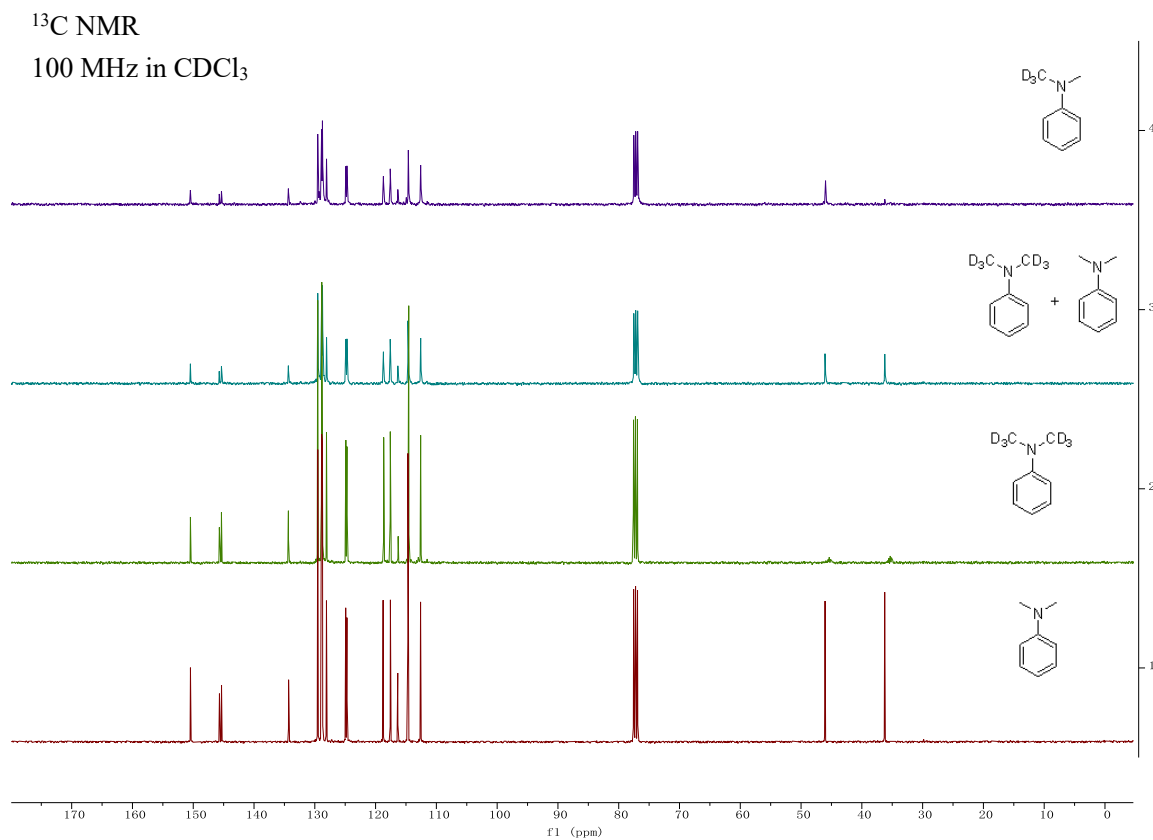
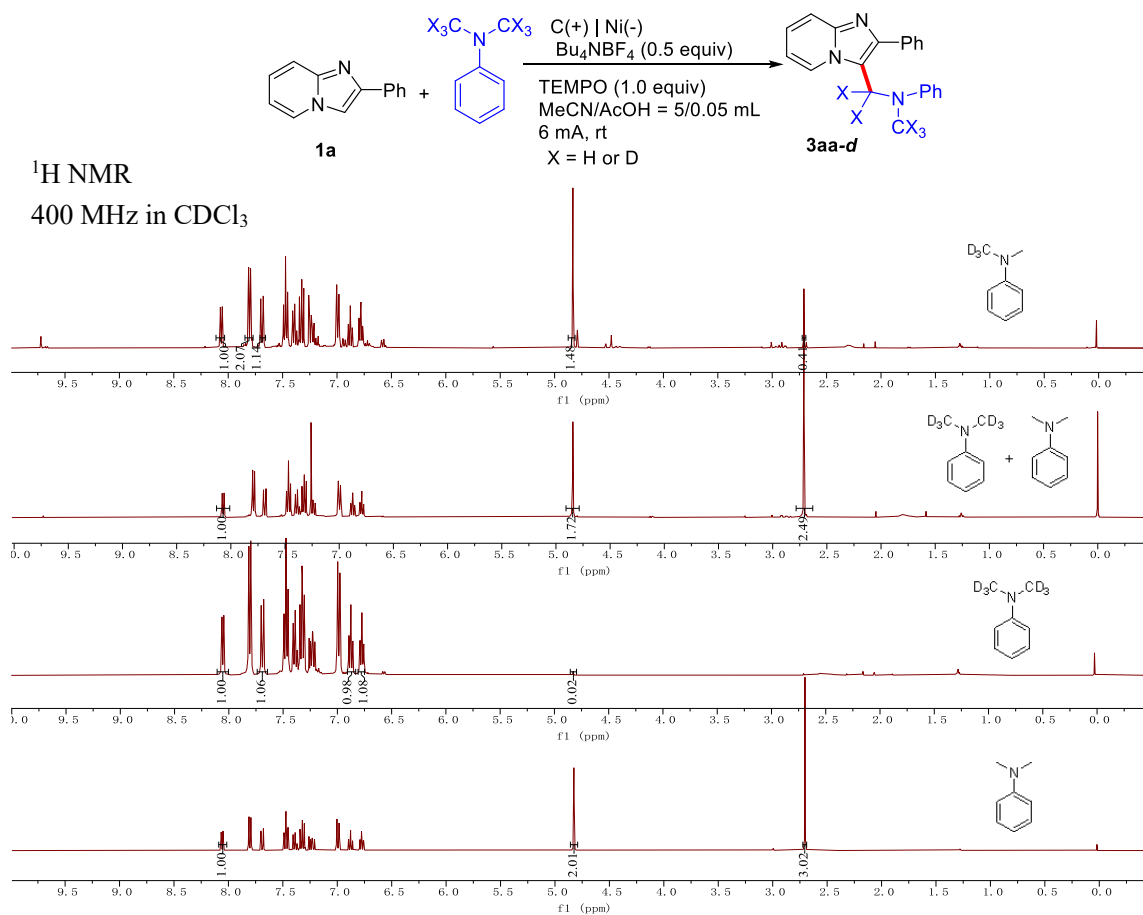


Figure S5  $^1\text{H-NMR}$ ,  $^{13}\text{C-NMR}$  of deuterated **3aa**

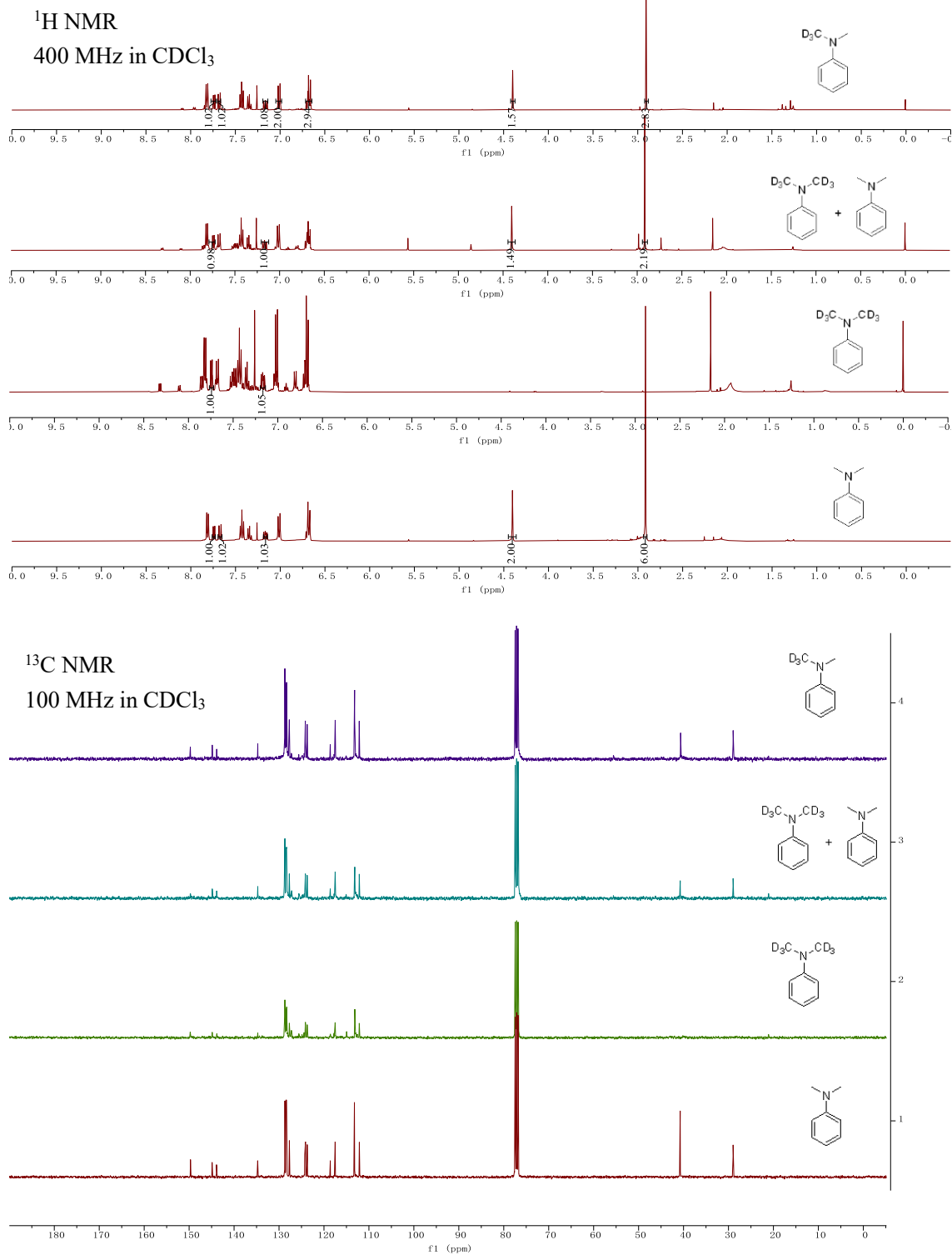
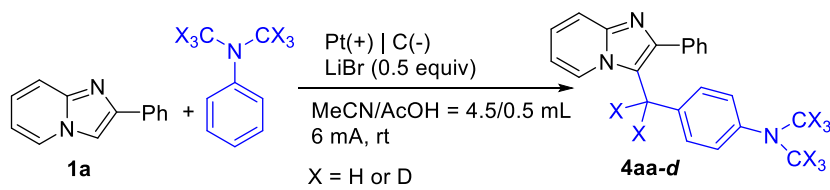
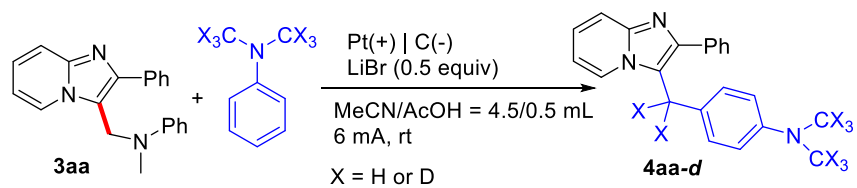
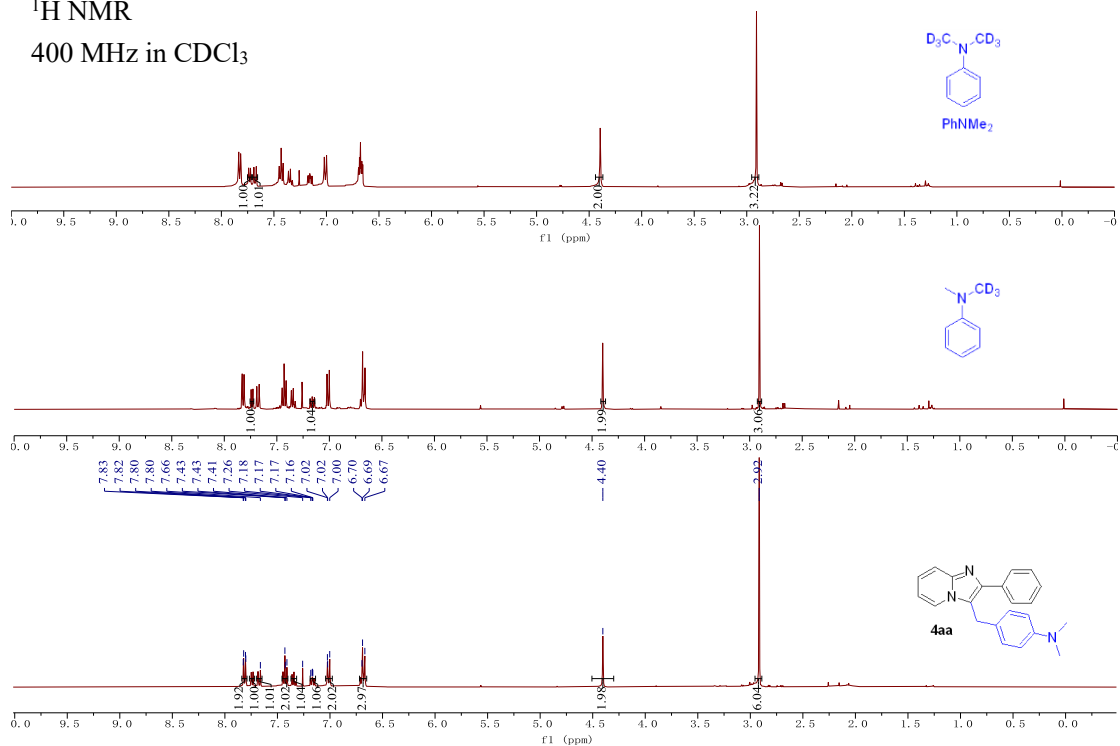


Figure S6 <sup>1</sup>H-NMR, <sup>13</sup>C-NMR of deuterated **4aa**



$^1\text{H}$  NMR

400 MHz in  $\text{CDCl}_3$



$^{13}\text{C}$  NMR

100 MHz in  $\text{CDCl}_3$

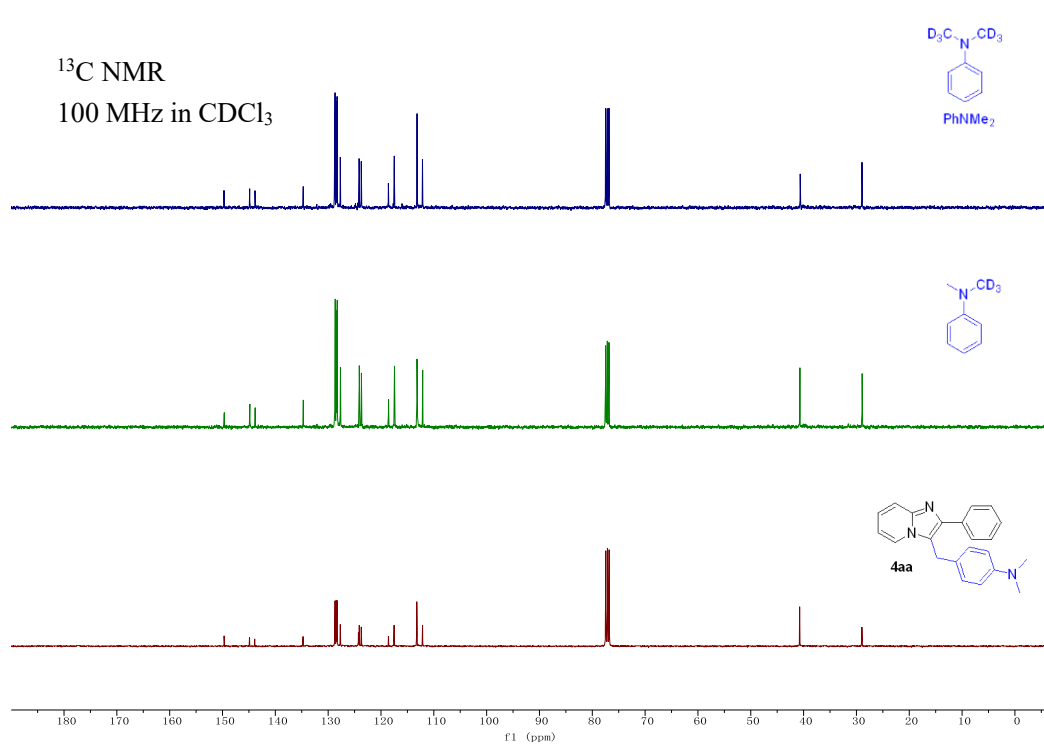


Figure S7  $^1\text{H}$ -NMR,  $^{13}\text{C}$ -NMR for the reaction of **3aa** with deuterated anilines

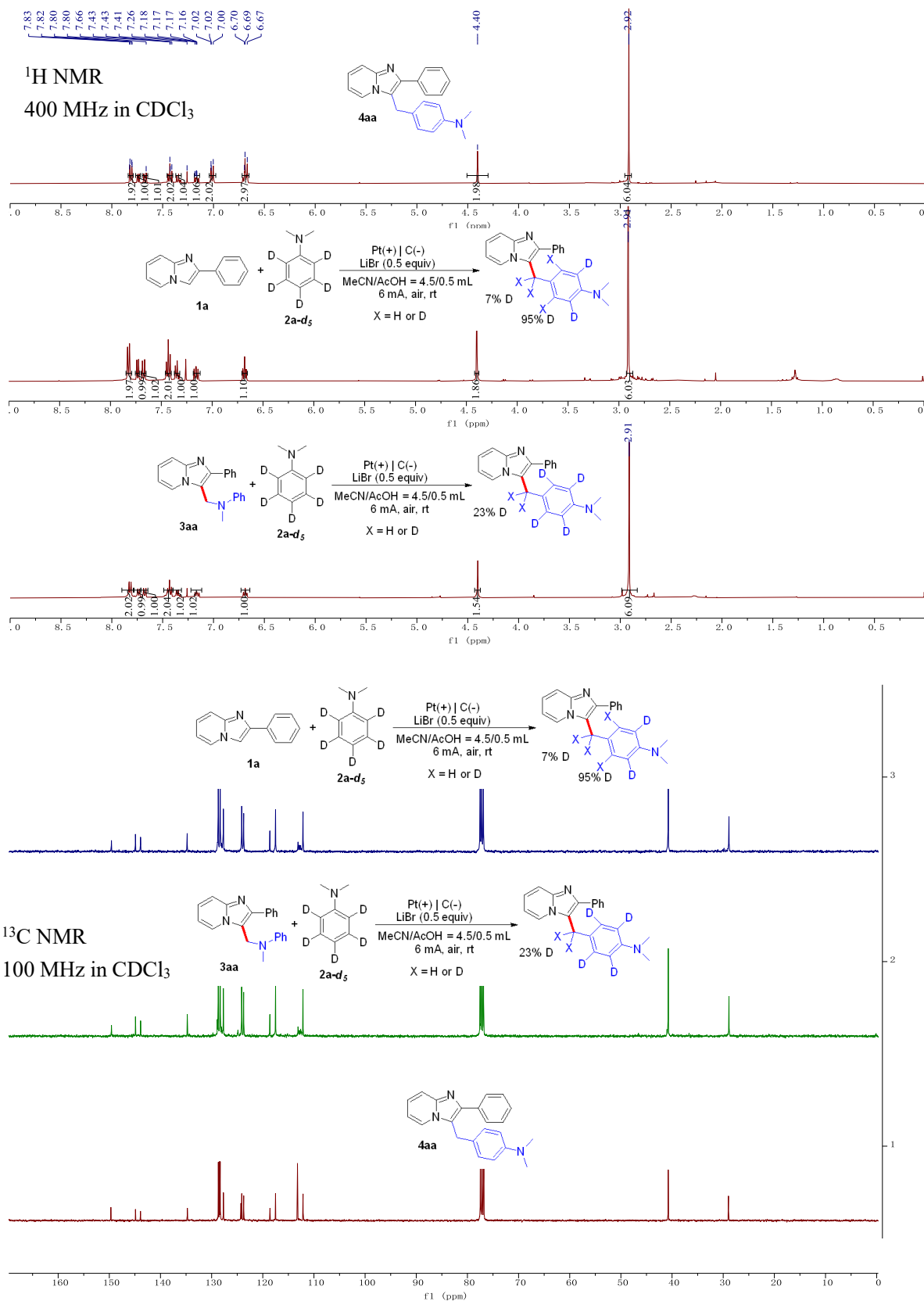
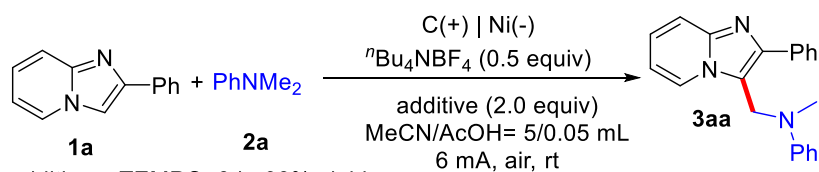
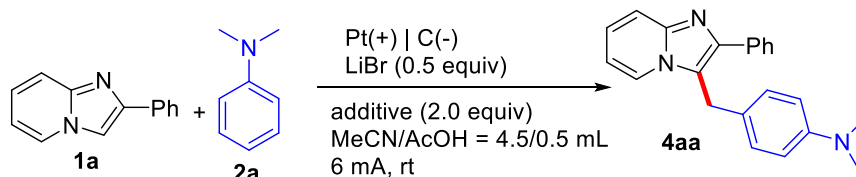


Figure S8 <sup>1</sup>H-NMR, <sup>13</sup>C-NMR of deuterated **4aa-D<sub>5</sub>**

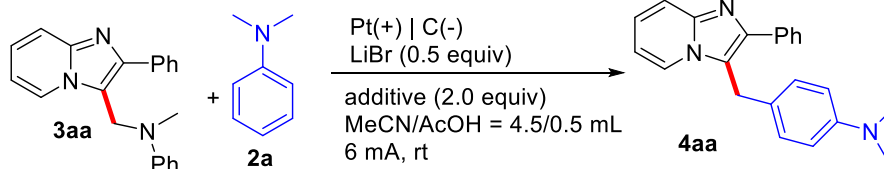
## 7. Free radical trap experiments and proposed mechanisms for 3aa



additive = TEMPO, 3 h, 98% yield  
 additive = BHT, 12 h, trace  
 additive = 1,1-diphenylethylene, 12 h, 21% yield  
 additive = 2-benzylidenemalononitrile, 12 h, 13% yield

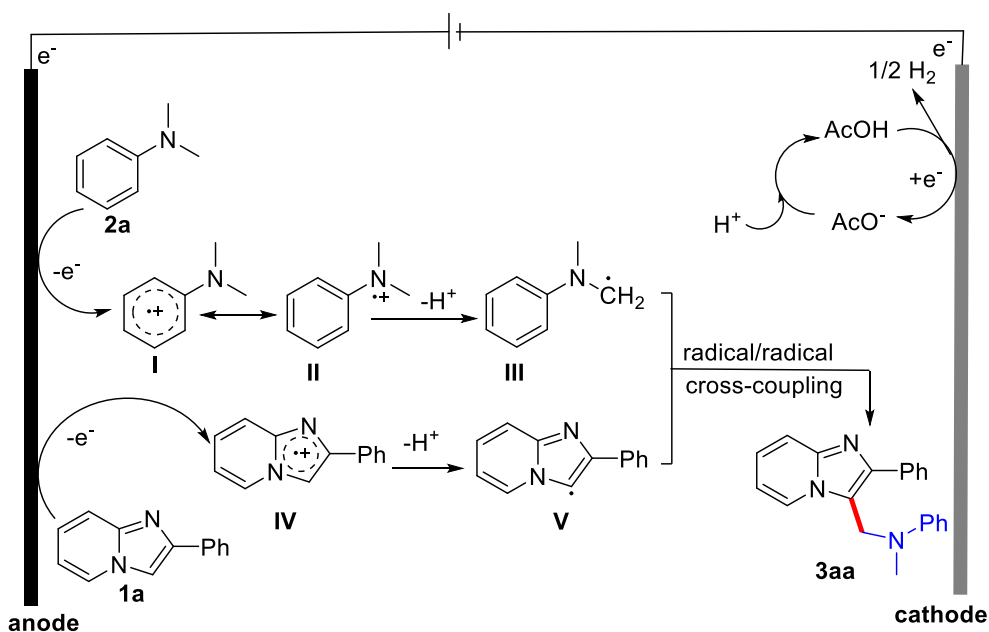


additive = TEMPO, 12 h, 71% yield  
 additive = BHT, 12 h, 30% yield  
 additive = 1,1-diphenylethylene, 12 h, 14% yield  
 additive = 2-benzylidenemalononitrile, 12 h, 8% yield



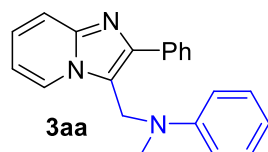
additive = TEMPO, 12 h, 75% yield  
 additive = BHT, 12 h, 65% yield  
 additive = 1,1-diphenylethylene, 12 h, 28% yield  
 additive = 2-benzylidenemalononitrile, 12 h, 42% yield

Scheme S3 The Free radical trap experiments

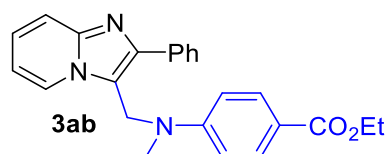


Scheme S4 Proposed mechanisms for 3aa

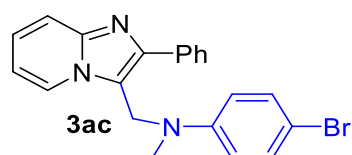
## 8. Characterization data (<sup>1</sup>H-NMR, <sup>13</sup>C-NMR and <sup>19</sup>F-NMR) of products



*N*-Methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3aa** (61 mg, Yield = 98%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 197–199 °C. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.06 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.82–7.78 (m, 2H), 7.69 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.51–7.44 (m, 2H), 7.41–7.36 (m, 1H), 7.35–7.29 (m, 2H), 7.23 (ddd,  $J = 9.1, 6.7, 1.3$  Hz, 1H), 7.02–6.96 (m, 2H), 6.90–6.85 (m, 1H), 6.78 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.83 (s, 2H), 2.70 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  150.39, 145.62, 145.28, 134.25, 129.45, 128.82, 128.68, 128.02, 124.85, 124.65, 118.70, 117.50, 116.29, 114.61, 112.54, 45.98, 36.18; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>21</sub>H<sub>20</sub>N<sub>3</sub> 314.1652; Found 314.1650. <sup>1</sup>H NMR and <sup>13</sup>C NMR data are consistent with the reported values.<sup>4-6</sup>

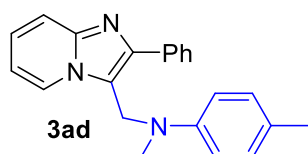


Ethyl 4-(methyl((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)amino)benzoate. Compound **3ab** (47 mg, Yield = 61%,  $R_f = 0.3$  (PE/EA = 2:1)) was isolated as a light brown paste. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.02–7.96 (m, 2H), 7.85 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.77–7.73 (m, 2H), 7.69 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.50–7.44 (m, 2H), 7.42–7.36 (m, 1H), 7.27–7.22 (m, 1H), 6.94–6.87 (m, 2H), 6.79 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.95 (s, 2H), 4.34 (q,  $J = 7.1$  Hz, 2H), 2.76 (s, 3H), 1.38 (t,  $J = 7.1$  Hz, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  166.86, 153.07, 146.13, 145.48, 134.01, 131.59, 128.85, 128.82, 128.30, 125.14, 124.16, 119.35, 117.80, 115.47, 113.03, 112.10, 60.50, 44.70, 35.12, 14.58; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>24</sub>H<sub>24</sub>N<sub>3</sub>O<sub>2</sub> 386.1863; Found 386.1863.

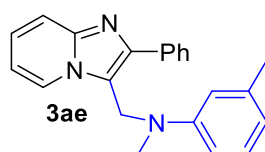


4-Bromo-*N*-methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ac** (68

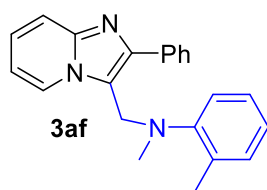
mg, Yield = 87%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid.  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.96 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.78–7.72 (m, 2H), 7.68 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.48–7.42 (m, 2H), 7.41–7.33 (m, 3H), 7.22 (ddd,  $J = 9.1, 6.7, 1.3$  Hz, 1H), 6.83–6.77 (m, 3H), 4.78 (s, 2H), 2.67 (s, 3H);  $^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.21, 145.83, 145.35, 134.16, 132.12, 128.80, 128.74, 128.14, 124.94, 124.38, 117.65, 115.97, 115.85, 112.71, 110.63, 45.77, 36.18; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{Br}$  392.0757; Found 392.0759.



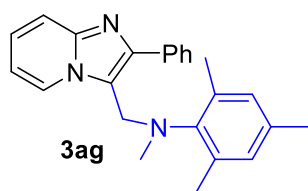
*N*,4-Dimethyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ad** (62 mg, Yield = 89%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 121–122 °C.  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.11 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.83–7.78 (m, 2H), 7.68 (dt,  $J = 9.0, 1.2$  Hz, 1H), 7.49–7.43 (m, 2H), 7.42–7.36 (m, 1H), 7.25–7.19 (m, 1H), 7.15–7.09 (m, 2H), 6.96–6.89 (m, 2H), 6.77 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.77 (s, 2H), 2.68 (s, 3H), 2.31 (s, 3H);  $^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  148.46, 145.52, 145.26, 134.34, 129.94, 128.97, 128.87, 128.66, 128.37, 127.98, 124.80, 117.47, 116.52, 115.29, 112.43, 46.66, 36.91, 20.42; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1809.  $^1\text{H NMR}$  and  $^{13}\text{C NMR}$  data are consistent with the literature.<sup>4, 5</sup>



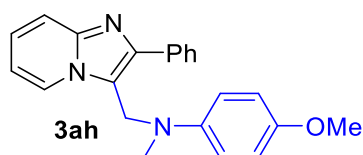
*N*,3-Dimethyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ae** (58 mg, Yield = 89%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.08 (dt,  $J = 6.9, 1.1$  Hz, 1H), 7.84–7.78 (m, 2H), 7.69 (dt,  $J = 9.1, 1.2$  Hz, 1H), 7.51–7.44 (m, 2H), 7.42–7.37 (m, 1H), 7.27–7.18 (m, 2H), 6.85–6.75 (m, 3H), 6.71 (d,  $J = 7.5$  Hz, 1H), 4.83 (s, 2H), 2.72 (s, 3H), 2.35 (s, 3H);  $^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.51, 145.64, 145.32, 139.22, 134.35, 129.31, 128.88, 128.70, 128.03, 124.83, 124.75, 119.64, 117.55, 116.42, 115.42, 112.52, 111.84, 46.10, 36.42, 21.96; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1807.



*N*,2-Dimethyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3af** (57 mg, Yield = 87%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.24 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.83–7.76 (m, 2H), 7.64 (dt,  $J$  = 9.1, 1.2 Hz, 1H), 7.52–7.45 (m, 2H), 7.43–7.37 (m, 1H), 7.23–7.16 (m, 2H), 7.15–7.08 (m, 2H), 7.02 (td,  $J$  = 7.2, 1.7 Hz, 1H), 6.77 (td,  $J$  = 6.8, 1.2 Hz, 1H), 4.60 (s, 2H), 2.52 (s, 3H), 2.26 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  151.24, 145.33, 145.14, 134.72, 133.73, 131.31, 129.04, 128.56, 127.84, 126.81, 125.24, 124.58, 124.18, 120.76, 117.39, 117.05, 111.78, 49.23, 42.50, 17.96; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1808.



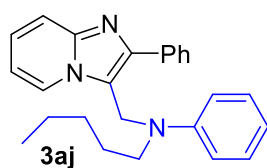
*N*,2,4,6-Tetramethyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ag** (60 mg, Yield = 85%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.93 (dt,  $J$  = 6.8, 1.2 Hz, 1H), 7.74–7.68 (m, 2H), 7.65 (dt,  $J$  = 9.0, 1.2 Hz, 1H), 7.47–7.41 (m, 2H), 7.39–7.34 (m, 1H), 7.20 (ddd,  $J$  = 9.0, 6.7, 1.3 Hz, 1H), 6.83 (s, 2H), 6.76 (td,  $J$  = 6.8, 1.2 Hz, 1H), 4.62 (s, 2H), 2.71 (s, 3H), 2.26 (s, 3H), 2.17 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.61, 144.97, 144.92, 136.73, 135.10, 134.73, 130.11, 128.93, 128.46, 127.74, 124.79, 124.53, 118.64, 117.35, 111.83, 48.56, 40.48, 20.75, 19.25; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{24}\text{H}_{26}\text{N}_3$  356.2121; Found 356.2122.



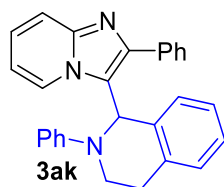
4-Methoxy-*N*-methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ah** (62



mg, Yield = 90%,  $R_f = 0.2$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.17 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.81–7.76 (m, 2H), 7.66 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.48–7.42 (m, 2H), 7.40–7.33 (m, 1H), 7.25–7.17 (m, 1H), 6.97–6.92 (m, 2H), 6.88–6.83 (m, 2H), 6.79–6.74 (m, 1H), 4.67 (s, 2H), 3.77 (s, 3H), 2.65 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  153.60, 145.43, 145.18, 145.08, 134.40, 128.86, 128.59, 127.90, 124.89, 124.72, 117.83, 117.41, 116.62, 114.70, 112.27, 55.68, 48.03, 38.42; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3\text{O}$  344.1757; Found 344.1757.

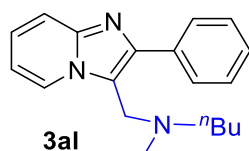


*N*-Pentyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3aj** (34 mg, Yield = 46%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.03 (dt,  $J = 7.0, 1.2$  Hz, 1H), 7.77–7.73 (m, 2H), 7.69 (dt,  $J = 9.1, 1.2$  Hz, 1H), 7.50–7.45 (m, 2H), 7.42–7.37 (m, 1H), 7.29–7.20 (m, 3H), 6.96–6.91 (m, 2H), 6.83 (tt,  $J = 7.4, 1.0$  Hz, 1H), 6.78 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.85 (s, 2H), 3.04–2.95 (m, 2H), 1.24–1.16 (m, 2H), 1.06–0.98 (m, 2H), 0.96–0.89 (m, 2H), 0.68 (t,  $J = 7.2$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.31, 145.69, 145.25, 134.45, 129.47, 128.95, 128.69, 128.06, 124.96, 124.83, 118.59, 117.53, 116.65, 115.50, 112.41, 49.16, 43.75, 29.16, 25.56, 22.34, 13.98; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{25}\text{H}_{28}\text{N}_3$  370.2278; Found 370.2280.

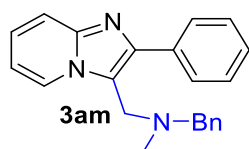


2-Phenyl-1-(2-phenylimidazo[1,2-*a*]pyridin-3-yl)-1,2,3,4-tetrahydroisoquinoline. Compound **3ak** (51 mg, Yield = 64%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light yellow solid; mp 117–118 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.19 (d,  $J = 6.7$  Hz, 1H), 7.53 (d,  $J = 9.1$  Hz, 1H), 7.46–7.41 (m, 2H), 7.40–7.33 (m, 3H), 7.28–7.24 (m, 2H), 7.20 (t,  $J = 7.5$  Hz, 1H), 7.11–7.06 (m, 1H), 7.03–6.95 (m, 3H), 6.94–6.88 (m, 2H), 6.72–6.66 (m, 2H), 6.62–6.56 (m, 1H), 5.98 (s, 1H), 3.57–3.51 (m, 1H),

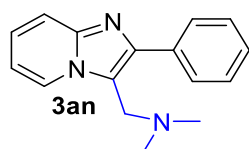
3.49–3.39 (m, 2H), 3.05–2.93 (m, 1H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  151.32, 146.81, 144.99, 135.43, 134.86, 134.55, 128.96, 128.77, 128.60, 128.59, 128.29, 127.84, 127.06, 126.70, 126.50, 126.14, 124.43, 123.50, 119.62, 117.17, 111.52, 58.90, 52.63, 30.47; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{28}\text{H}_{24}\text{N}_3$  402.1965; Found 402.1966.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>4</sup>



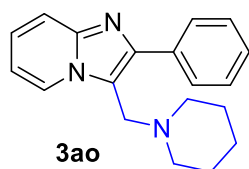
*N*-Methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)butan-1-amine. Compound **3al** (50 mg, Yield = 85%,  $R_f$  = 0.2 (PE/EA = 5:1)) was isolated as a light yellow paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.42 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.83–7.78 (m, 2H), 7.62 (dt,  $J$  = 9.1, 1.1 Hz, 1H), 7.48–7.41 (m, 2H), 7.38–7.30 (m, 1H), 7.18 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 6.78 (td,  $J$  = 6.8, 1.2 Hz, 1H), 3.93 (s, 2H), 2.43–2.35 (m, 2H), 2.16 (s, 3H), 1.51–1.39 (m, 2H), 1.31–1.22 (m, 2H), 0.83 (t,  $J$  = 7.3 Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.00, 144.80, 134.74, 128.97, 128.45, 127.66, 125.61, 124.48, 117.53, 117.17, 111.72, 56.95, 51.70, 41.60, 29.45, 20.51, 14.01; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{19}\text{H}_{24}\text{N}_3$  294.1965; Found 294.1964.



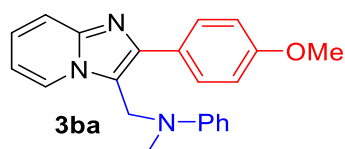
*N*-Benzyl-*N*-methyl-1-(2-phenylimidazo[1,2-*a*]pyridin-3-yl)methanamine. Compound **3am** (50 mg, Yield = 76%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.41–8.36 (m, 1H), 7.89–7.80 (m, 2H), 7.68–7.61 (m, 1H), 7.52–7.45 (m, 2H), 7.42–7.36 (m, 1H), 7.35–7.18 (m, 6H), 6.87–6.79 (m, 1H), 4.01 (s, 2H), 3.52 (s, 2H), 2.19 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.08, 145.04, 138.77, 134.68, 129.09, 129.01, 128.51, 128.35, 127.76, 127.25, 125.54, 124.59, 117.25, 111.81, 61.74, 50.98, 42.13, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1808.



*N,N*-Dimethyl-1-(2-phenylimidazo[1,2-*a*]pyridin-3-yl)methanamine. Compound **3an** (26 mg, Yield = 51%,  $R_f = 0.15$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.40 (d,  $J = 6.9$  Hz, 1H), 7.84–7.77 (m, 2H), 7.64 (d,  $J = 9.1$  Hz, 1H), 7.50–7.43 (m, 2H), 7.41–7.34 (m, 1H), 7.25–7.17 (m, 1H), 6.87–6.77 (m, 1H), 3.89 (s, 2H), 2.26 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.13, 144.95, 134.77, 129.06, 128.55, 127.78, 125.54, 124.64, 117.48, 117.34, 111.98, 53.02, 45.15; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{16}\text{H}_{18}\text{N}_3$  252.1495; Found 252.1497.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>5-7</sup>

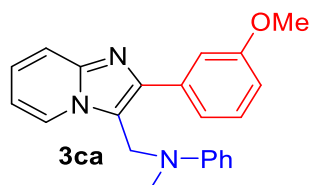


2-Phenyl-3-(piperidin-1-ylmethyl)imidazo[1,2-*a*]pyridine. Compound **3ao** (44 mg, Yield = 76%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 78–90 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.50–8.44 (m, 1H), 7.83–7.79 (m, 2H), 7.64–7.59 (m, 1H), 7.47–7.41 (m, 2H), 7.37–7.31 (m, 1H), 7.20–7.14 (m, 1H), 6.77 (tt,  $J = 6.8, 1.6$  Hz, 1H), 3.89 (d,  $J = 1.4$  Hz, 2H), 2.40 (br s, 4H), 1.56–1.48 (m, 4H), 1.47–1.35 (m, 2H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.02, 144.80, 134.77, 129.90, 128.98, 128.44, 127.63, 125.77, 124.42, 117.14, 111.68, 54.29, 52.64, 26.13, 24.46; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{19}\text{H}_{22}\text{N}_3$  292.1808; Found 292.1808.

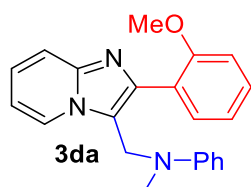


*N*-((2-(4-Methoxyphenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3ba** (64 mg, Yield = 93%,  $R_f = 0.2$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.04 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.75–7.71 (m, 2H), 7.66 (dt,  $J = 9.0, 1.1$  Hz, 1H), 7.35–7.28 (m, 2H), 7.22 (ddd,  $J = 9.0, 6.7, 1.3$  Hz, 1H), 7.01–6.97 (m, 4H), 6.87 (tt,  $J = 7.3, 1.1$  Hz, 1H), 6.77 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.81 (s, 2H), 3.85 (s, 3H), 2.70 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,

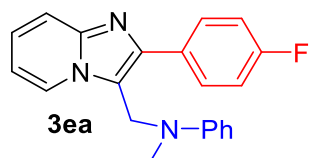
CDCl<sub>3</sub>)  $\delta$  159.62, 150.49, 145.60, 145.28, 130.06, 129.51, 126.85, 124.75, 124.57, 118.71, 117.41, 115.70, 114.65, 114.20, 112.47, 55.44, 46.10, 36.18; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>22</sub>H<sub>22</sub>N<sub>3</sub>O 344.1757; Found 344.1757.



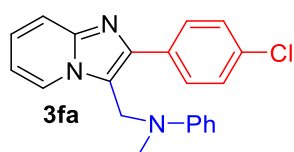
*N*-((2-(3-Methoxyphenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3ca** (62 mg, Yield = 90%, R<sub>f</sub> = 0.2 (PE/EA = 5:1)) was isolated as a light brown paste. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.07 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.69 (dt,  $J$  = 9.0, 1.1 Hz, 1H), 7.38–7.35 (m, 3H), 7.33–7.29 (m, 2H), 7.25–7.21 (m, 1H), 7.00–6.97 (m, 2H), 6.95–6.91 (m, 1H), 6.87 (tt,  $J$  = 7.2, 6.2, 1.0 Hz, 1H), 6.79 (td,  $J$  = 6.8, 1.2 Hz, 1H), 4.83 (s, 2H), 3.84 (s, 3H), 2.71 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  159.93, 150.46, 145.56, 145.27, 135.64, 129.70, 129.51, 124.96, 124.70, 121.29, 118.80, 117.59, 116.49, 114.71, 114.31, 113.90, 112.66, 55.44, 46.05, 36.33; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>22</sub>H<sub>22</sub>N<sub>3</sub>O 344.1757; Found 344.1757.



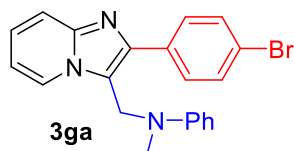
*N*-((2-(2-Methoxyphenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3da** (57 mg, Yield = 83%, R<sub>f</sub> = 0.2 (PE/EA = 5:1)) was isolated as a light brown solid; mp 84–86 °C. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.05 (d,  $J$  = 6.9 Hz, 1H), 7.66 (d,  $J$  = 8.9 Hz, 1H), 7.56 (dd,  $J$  = 7.5, 1.8 Hz, 1H), 7.41–7.36 (m, 1H), 7.30–7.24 (m, 2H), 7.21–7.15 (m, 1H), 7.07 (t,  $J$  = 7.4 Hz, 1H), 6.98 (d,  $J$  = 8.3 Hz, 1H), 6.93 (d,  $J$  = 7.9 Hz, 2H), 6.81 (t,  $J$  = 7.3 Hz, 1H), 6.74 (td,  $J$  = 6.8, 1.2 Hz, 1H), 4.65 (s, 2H), 3.79 (s, 3H), 2.65 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  156.95, 150.58, 145.38, 142.35, 132.32, 129.84, 129.41, 124.81, 124.28, 123.38, 120.93, 118.23, 118.05, 117.58, 114.30, 112.38, 111.07, 55.54, 46.42, 36.24; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>22</sub>H<sub>22</sub>N<sub>3</sub>O 344.1757; Found 344.1755.



*N*-((2-(4-Fluorophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3ea** (62 mg, Yield = 94%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 101–102 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.08–8.02 (m, 1H), 7.78–7.73 (m, 2H), 7.69–7.65 (m, 1H), 7.36–7.29 (m, 2H), 7.26–7.20 (m, 1H), 7.18–7.12 (m, 2H), 7.03–6.96 (m, 2H), 6.91–6.86 (m, 1H), 6.82–6.76 (m, 1H), 4.78 (s, 2H), 2.69 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  162.68 (d,  $J = 246.2$  Hz), 150.30, 145.21, 144.67, 130.45 (d,  $J = 8.1$  Hz), 129.44, 124.95, 124.57, 118.85, 117.43, 116.13, 115.63 (d,  $J = 21.2$  Hz), 114.70, 112.58, 46.00, 36.25, (1C is merged with other peaks);  $^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ )  $\delta$  –113.75 (s, 1F). HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{F}$  332.1558; Found 332.1558.

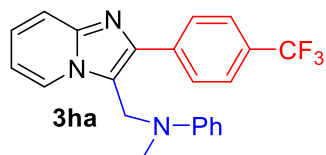


*N*-((2-(4-Chlorophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3fa** (64 mg, Yield = 92%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 134–136 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.06 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.80–7.71 (m, 2H), 7.67 (d,  $J = 9.1$  Hz, 1H), 7.44–7.40 (m, 2H), 7.36–7.29 (m, 2H), 7.27–7.22 (m, 1H), 6.99 (d,  $J = 7.8$  Hz, 2H), 6.89 (t,  $J = 7.3$  Hz, 1H), 6.82–6.76 (m, 1H), 4.78 (s, 2H), 2.69 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.31, 145.31, 144.44, 133.99, 132.82, 129.99, 129.48, 128.88, 125.07, 124.61, 118.95, 117.53, 116.46, 114.77, 112.69, 46.06, 36.32; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{Cl}$  348.1262; Found 348.1263.



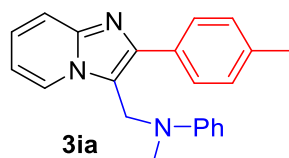
*N*-((2-(4-Bromophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3ga** (67 mg, Yield = 86%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 108–109 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.11–8.04 (m, 1H), 7.71–7.64 (m, 3H), 7.62–7.55 (m, 2H), 7.38–7.29

(m, 2H), 7.27–7.21 (m, 1H), 7.04–6.96 (m, 2H), 6.93–6.86 (m, 1H), 6.83–6.76 (m, 1H), 4.78 (s, 2H), 2.69 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.34, 145.36, 144.45, 133.27, 131.85, 130.32, 129.50, 125.13, 124.65, 122.30, 119.00, 117.56, 116.53, 114.83, 112.74, 46.11, 36.38; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{Br}$  392.0757; Found 392.0756.

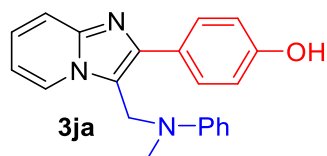


*N*-Methyl-*N*-((2-(4-(trifluoromethyl)phenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline.

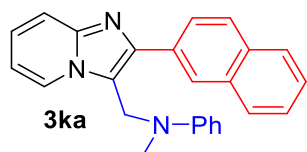
Compound **3ha** (61 mg, Yield = 80%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.10 (dt,  $J$  = 6.9, 1.1 Hz, 1H), 7.95–7.91 (m, 2H), 7.74–7.67 (m, 3H), 7.36–7.31 (m, 2H), 7.29–7.24 (m, 1H), 7.02–6.97 (m, 2H), 6.90 (tt,  $J$  = 7.3, 1.1 Hz, 1H), 6.82 (td,  $J$  = 6.8, 1.2 Hz, 1H), 4.82 (s, 2H), 2.71 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.37, 145.52, 144.13, 137.96, 129.94 (q,  $J$  = 32.2 Hz), 129.58, 129.04, 125.63 (q,  $J$  = 3.8 Hz), 125.41, 124.80, 124.34 (q,  $J$  = 270.8 Hz), 119.22, 117.79, 117.22, 114.99, 112.98, 46.23, 36.53;  $^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ )  $\delta$  –62.30 (s, 3F). HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{F}_3$  382.1526; Found 382.1524.



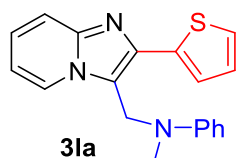
*N*-Methyl-*N*-((2-(*p*-tolyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ia** (56 mg, Yield = 86%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown solid; mp 96–97 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.05 (dt,  $J$  = 6.9, 1.4 Hz, 1H), 7.72–7.65 (m, 3H), 7.35–7.30 (m, 2H), 7.28 (d,  $J$  = 7.7 Hz, 2H), 7.25–7.20 (m, 1H), 7.03–6.97 (m, 2H), 6.87 (tt,  $J$  = 7.4, 1.1 Hz, 1H), 6.77 (tt,  $J$  = 6.9, 1.1 Hz, 1H), 4.82 (s, 2H), 2.70 (s, 3H), 2.41 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.45, 145.73, 145.27, 137.85, 131.37, 129.47, 129.43, 128.70, 124.74, 124.61, 118.64, 117.45, 116.02, 114.58, 112.46, 46.01, 36.12, 21.37; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1807.



4-(3-((Methyl(phenyl)amino)methyl)imidazo[1,2-*a*]pyridin-2-yl)phenol. Compound **3ja** (57 mg, Yield = 87%,  $R_f = 0.2$  (PE/EA = 2:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.07 (d,  $J = 7.0$  Hz, 1H), 7.70 (d,  $J = 9.1$  Hz, 1H), 7.51 (d,  $J = 8.7$  Hz, 2H), 7.35–7.28 (m, 2H), 7.25–7.20 (m, 1H), 6.98 (d,  $J = 7.7$  Hz, 2H), 6.89–6.83 (m, 3H), 6.82–6.77 (m, 1H), 4.80 (s, 2H), 2.70 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  158.02, 150.50, 145.40, 144.94, 130.30, 129.53, 125.41, 124.78, 124.22, 118.80, 116.77, 116.19, 115.77, 114.77, 112.91, 46.07, 36.47; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{20}\text{N}_3\text{O}$  330.1601; Found 330.1603.

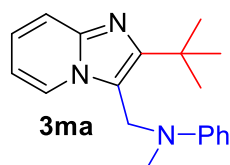


*N*-Methyl-*N*-((2-(naphthalen-2-yl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ka** (54 mg, Yield = 74%,  $R_f = 0.2$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.26 (s, 1H), 8.11–8.05 (m, 1H), 8.00–7.91 (m, 2H), 7.90–7.83 (m, 2H), 7.75–7.70 (m, 1H), 7.54–7.46 (m, 2H), 7.37–7.29 (m, 2H), 7.28–7.21 (m, 1H), 7.05–6.96 (m, 2H), 6.93–6.87 (m, 1H), 6.81–6.76 (m, 1H), 4.86 (s, 2H), 2.71 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.35, 145.47, 145.34, 133.45, 132.94, 131.67, 129.45, 128.37, 128.31, 127.85, 127.73, 126.60, 126.31, 126.26, 124.92, 124.59, 118.77, 117.47, 116.69, 114.70, 112.57, 46.10, 36.30; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{25}\text{H}_{22}\text{N}_3$  364.1808; Found 364.1807.

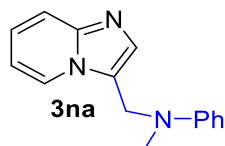


*N*-Methyl-*N*-((2-(thiophen-2-yl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3la** (57 mg, Yield = 89%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste; mp 120–122 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.00 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.65 (dt,  $J = 9.1, 1.2$  Hz, 1H), 7.49 (dd,  $J = 3.7, 1.2$  Hz, 1H), 7.40–7.30 (m, 3H), 7.21 (ddd,  $J = 9.1, 6.8, 1.3$  Hz, 1H), 7.12 (dd,  $J = 5.1, 3.6$  Hz, 1H), 7.06–6.99 (m, 2H), 6.92–6.86 (m, 1H), 6.76 (td,  $J = 6.8, 1.3$  Hz, 1H), 4.88 (s, 2H), 2.71 (s, 3H);  $^{13}\text{C}$

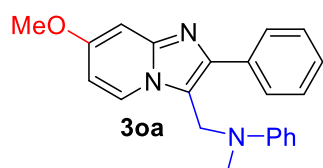
NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  150.51, 145.29, 139.85, 137.14, 129.53, 127.81, 126.03, 125.57, 125.20, 124.52, 118.86, 117.34, 115.73, 114.74, 112.69, 45.97, 36.45; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>19</sub>H<sub>18</sub>N<sub>3</sub>S 320.1216; Found 320.1218.



*N*-((2-(Tert-butyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3ma** (51 mg, Yield = 87%, R<sub>f</sub> = 0.3 (PE/EA = 10:1)) was isolated as a light brown solid; mp 109–110 °C. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.94 (dt, *J* = 6.9, 1.3 Hz, 1H), 7.63 (dt, *J* = 9.1, 1.2 Hz, 1H), 7.40–7.33 (m, 2H), 7.19–7.13 (m, 1H), 7.05–6.99 (m, 2H), 6.91–6.85 (m, 1H), 6.71 (td, *J* = 6.8, 1.2 Hz, 1H), 4.82 (s, 2H), 2.63 (s, 3H), 1.51 (s, 9H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  154.28, 150.49, 143.90, 129.55, 124.32, 124.00, 118.46, 117.12, 114.39, 114.30, 112.17, 45.92, 35.12, 33.63, 31.44; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>19</sub>H<sub>24</sub>N<sub>3</sub> 294.1965; Found 294.1964.

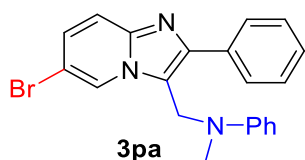


*N*-(Imidazo[1,2-*a*]pyridin-3-ylmethyl)-*N*-methylaniline. Compound **3na** (40 mg, Yield = 85%, R<sub>f</sub> = 0.2 (PE/EA = 2:1)) was isolated as a light brown solid; mp 77–78 °C. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.01–7.96 (m, 1H), 7.65–7.61 (m, 1H), 7.56 (s, 1H), 7.33–7.28 (m, 2H), 7.20–7.15 (m, 1H), 7.00–6.94 (m, 2H), 6.87–6.82 (m, 1H), 6.76 (tt, *J* = 6.8, 1.3 Hz, 1H), 4.67 (s, 2H), 2.77 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  150.32, 146.35, 133.57, 129.43, 124.35, 124.24, 120.62, 118.58, 117.90, 114.51, 112.43, 46.94, 37.27; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>15</sub>H<sub>16</sub>N<sub>3</sub> 238.1339; Found 238.1339.

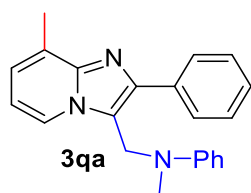




*N*-((7-Methoxy-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3oa** (40 mg, Yield = 58%,  $R_f = 0.2$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 115–116 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.87 (d,  $J = 7.6$  Hz, 1H), 7.79–7.74 (m, 2H), 7.47–7.42 (m, 2H), 7.39–7.36 (m, 1H), 7.34–7.29 (m, 2H), 7.00–6.94 (m, 3H), 6.86 (tt,  $J = 7.3, 1.1$  Hz, 1H), 6.50 (dd,  $J = 7.5, 2.5$  Hz, 1H), 4.78 (s, 2H), 3.87 (s, 3H), 2.70 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  158.18, 150.49, 146.82, 145.10, 134.44, 129.49, 128.67, 128.64, 127.86, 125.18, 118.72, 115.15, 114.67, 107.52, 94.75, 55.60, 46.04, 36.13; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3\text{O}$  344.1757; Found 344.1755.

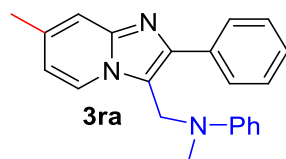


*N*-((6-Bromo-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-*N*-methylaniline. Compound **3pa** (65 mg, Yield = 83%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 157–158 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.24–8.20 (m, 1H), 7.79–7.73 (m, 2H), 7.58–7.53 (m, 1H), 7.50–7.44 (m, 2H), 7.42–7.37 (m, 1H), 7.35–7.24 (m, 3H), 7.03–6.96 (m, 2H), 6.94–6.87 (m, 1H), 4.77 (s, 2H), 2.70 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.47, 146.39, 143.68, 133.80, 129.46, 128.80, 128.75, 128.28, 128.22, 124.83, 119.33, 118.13, 116.91, 115.30, 107.19, 46.54, 36.95; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{21}\text{H}_{19}\text{N}_3\text{Br}$  392.0757; Found 392.0758.

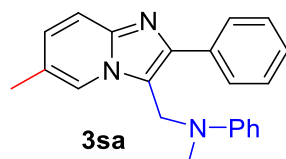


*N*-Methyl-*N*-((8-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3qa** (54 mg, Yield = 83%,  $R_f = 0.3$  (PE/EA = 10:1)) was isolated as a light brown solid; mp 100–102 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.94 (d,  $J = 6.9$  Hz, 1H), 7.84–7.79 (m, 2H), 7.51–7.45 (m, 2H), 7.42–7.37 (m, 1H), 7.36–7.30 (m, 2H), 7.04 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.02–6.98 (m, 2H), 6.89 (tt,  $J = 7.3, 1.1$  Hz, 1H), 6.71 (t,  $J = 6.9$  Hz, 1H), 4.82 (s, 2H), 2.72 (s, 3H), 2.72 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.35, 145.64, 145.18, 134.47, 129.35, 128.93, 128.58, 127.81, 127.37, 123.54, 122.33, 118.43, 116.51, 114.39, 112.48, 45.87, 35.95, 17.19; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$

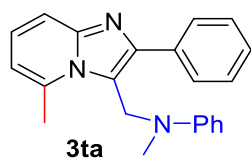
328.1808; Found 328.1808.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>4</sup>



*N*-Methyl-*N*-((7-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ra** (49 mg, Yield = 75%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.94 (d,  $J$  = 7.0 Hz, 1H), 7.79–7.75 (m, 2H), 7.49–7.42 (m, 3H), 7.40–7.36 (m, 1H), 7.34–7.29 (m, 2H), 7.02–6.96 (m, 2H), 6.87 (tt,  $J$  = 7.3, 1.1 Hz, 1H), 6.63 (dd,  $J$  = 7.1, 1.7 Hz, 1H), 4.81 (s, 2H), 2.70 (s, 3H), 2.42 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.47, 145.78, 145.39, 135.84, 134.46, 129.47, 128.78, 128.67, 127.91, 123.86, 118.60, 115.95, 115.67, 115.18, 114.55, 45.96, 36.03, 21.46; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1808.

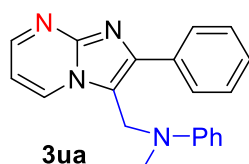


*N*-Methyl-*N*-((6-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3sa** (54 mg, Yield = 82%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light brown solid; mp 81–83 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.84–7.81 (m, 1H), 7.79–7.76 (m, 2H), 7.63 (d,  $J$  = 9.1 Hz, 1H), 7.48–7.43 (m, 2H), 7.40–7.30 (m, 3H), 7.12 (dd,  $J$  = 9.1, 1.7 Hz, 1H), 7.03–6.98 (m, 2H), 6.88 (tt,  $J$  = 7.3, 1.0 Hz, 1H), 4.80 (s, 2H), 2.71 (s, 3H), 2.29 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.52, 145.51, 144.37, 134.41, 129.41, 128.70, 128.64, 128.01, 127.88, 122.18, 118.61, 116.83, 115.93, 114.62, 46.02, 35.98, 18.49; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1807.

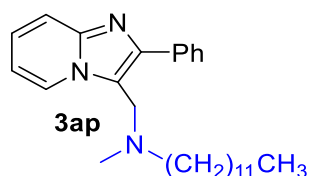


*N*-Methyl-*N*-((5-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3ta** (37 mg, Yield = 57%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown solid; mp 197–199 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.70–7.63 (m, 2H), 7.57 (d,  $J$  = 9.0 Hz, 1H), 7.46–7.40 (m, 2H), 7.39–7.35 (m, 1H), 7.34–7.29 (m, 2H), 7.14 (dd,  $J$  = 9.0, 6.8 Hz, 1H), 6.91–6.81 (m, 3H), 6.56 (dt,  $J$  =

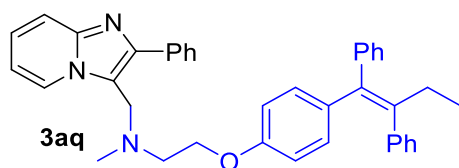
6.9, 1.1 Hz, 1H), 4.82 (s, 2H), 2.78 (s, 3H), 2.64 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.29, 148.14, 147.37, 137.09, 134.52, 129.45, 129.26, 128.65, 128.07, 125.47, 118.06, 117.13, 115.89, 113.94, 113.65, 45.43, 34.78, 19.28; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1811.



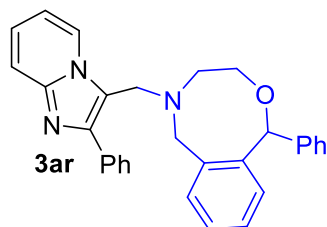
*N*-Methyl-*N*-((2-phenylimidazo[1,2-*a*]pyrimidin-3-yl)methyl)aniline. Compound **3ua** (58 mg, Yield = 93%,  $R_f$  = 0.3 (PE/EA = 2:1)) was isolated as a light brown solid; mp 156–158 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.54 (dd,  $J$  = 4.1, 2.0 Hz, 1H), 8.39 (dd,  $J$  = 6.9, 2.0 Hz, 1H), 7.86–7.81 (m, 2H), 7.49–7.44 (m, 2H), 7.42–7.38 (m, 1H), 7.33–7.27 (m, 2H), 7.00–6.94 (m, 2H), 6.88 (tt,  $J$  = 7.3, 1.1 Hz, 1H), 6.81 (dd,  $J$  = 6.9, 4.1 Hz, 1H), 4.84 (s, 2H), 2.69 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  150.27, 149.93, 148.30, 146.82, 133.59, 132.56, 129.53, 129.00, 128.71, 128.46, 119.32, 115.11, 115.08, 108.69, 46.43, 37.00; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{20}\text{H}_{19}\text{N}_4$  315.1604; Found 315.1601.



*N*-Methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)dodecan-1-amine. Compound **3ap** (73 mg, Yield = 90%,  $R_f$  = 0.3 (PE/EA = 5:1)) was isolated as a light yellow oil.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.44 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.84–7.77 (m, 2H), 7.62 (dt,  $J$  = 9.1, 1.2 Hz, 1H), 7.47–7.42 (m, 2H), 7.39–7.32 (m, 1H), 7.19 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 6.79 (td,  $J$  = 6.8, 1.2 Hz, 1H), 3.94 (s, 2H), 2.38 (t,  $J$  = 7.3 Hz, 2H), 2.17 (s, 3H), 1.50–1.42 (m, 2H), 1.30–1.18 (m, 18H), 0.87 (t,  $J$  = 7.1 Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.05, 144.85, 134.79, 129.02, 128.48, 127.69, 125.69, 124.49, 117.59, 117.23, 111.73, 57.20, 51.77, 41.68, 32.03, 29.78, 29.75, 29.72, 29.57, 29.46, 27.41, 27.33, 22.80, 14.24, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{27}\text{H}_{40}\text{N}_3$  406.3217; Found 406.3220.

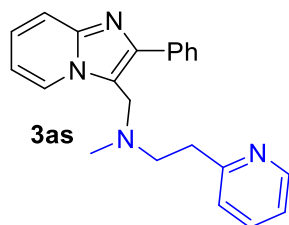


(*Z*)-2-(4-(1,2-Diphenylbut-1-en-1-yl)phenoxy)-*N*-methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)ethan-1-amine. Compound **3aq** (92 mg, Yield = 82%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light yellow paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.48 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.78–7.75 (m, 2H), 7.64–7.59 (m, 1H), 7.46–7.40 (m, 2H), 7.37–7.33 (m, 3H), 7.30–7.27 (m, 1H), 7.25–7.23 (m, 2H), 7.21–7.14 (m, 3H), 7.14–7.10 (m, 3H), 6.78–6.74 (m, 2H), 6.68 (td,  $J = 6.8, 1.2$  Hz, 1H), 6.52–6.47 (m, 2H), 4.06 (s, 2H), 3.93 (t,  $J = 5.4$  Hz, 2H), 2.79 (t,  $J = 5.4$  Hz, 2H), 2.46 (q,  $J = 7.4$  Hz, 2H), 2.26 (s, 3H), 0.93 (t,  $J = 7.4$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  156.67, 145.16, 144.94, 143.90, 142.54, 141.52, 138.29, 135.78, 134.71, 132.05, 129.82, 129.58, 129.01, 128.55, 128.23, 128.00, 127.77, 126.66, 126.12, 126.00, 124.67, 117.18, 113.34, 111.87, 65.79, 55.80, 51.56, 42.44, 29.16, 13.73, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{39}\text{H}_{38}\text{N}_3\text{O}$  564.3009; Found 564.3012.



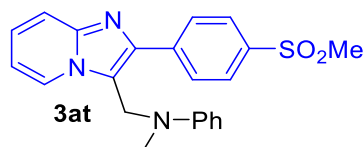
1-Phenyl-5-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-3,4,5,6-tetrahydro-1H-benzo[*f*][1,4]oxazocine. Compound **3ar** (65 mg, Yield = 73%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.42 (d,  $J = 7.2$  Hz, 1H), 7.82–7.76 (m, 2H), 7.68 (d,  $J = 9.4$  Hz, 1H), 7.49–7.44 (m, 2H), 7.41–7.36 (m, 1H), 7.30–7.22 (m, 6H), 7.14–7.08 (m, 1H), 7.01–6.92 (m, 2H), 6.83–6.76 (m, 1H), 6.57 (d,  $J = 6.2$  Hz, 1H), 5.78 (s, 1H), 4.74 (d,  $J = 12.9$  Hz, 1H), 4.20 (d,  $J = 2.3$  Hz, 2H), 4.11 (ddd,  $J = 12.4, 8.0, 2.6$  Hz, 1H), 3.82 (ddd,  $J = 12.4, 6.1, 2.2$  Hz, 1H), 3.63 (d,  $J = 12.8$  Hz, 1H), 2.85 (ddd,  $J = 13.8, 8.0, 2.3$  Hz, 1H), 2.67 (ddd,  $J = 13.8, 6.1, 2.6$  Hz, 1H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  145.26, 142.78, 140.83, 134.62, 132.82, 129.09, 128.65, 128.56, 127.89, 127.68, 127.62, 127.44, 127.35, 125.91, 124.78, 117.28, 117.22, 111.94, 84.37,

68.89, 55.45, 52.35, 49.85, (3C are merged with other peaks); HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{30}H_{28}N_3O$  446.2227; Found 446.2228.

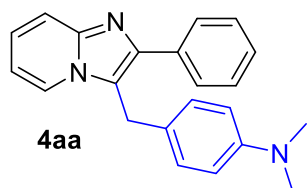


*N*-Methyl-*N*-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-2-(pyridin-2-yl)ethan-1-amine.

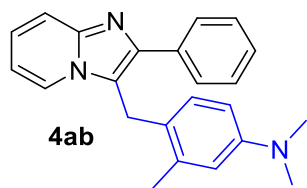
Compound **3as** (62 mg, Yield = 91%,  $R_f$  = 0.2 (PE/EA = 5:1)) was isolated as a light yellow paste.  $^1H$  NMR (400 MHz,  $CDCl_3$ )  $\delta$  8.46 (ddd,  $J$  = 4.9, 1.9, 0.9 Hz, 1H), 8.00 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.78–7.73 (m, 2H), 7.56 (dt,  $J$  = 9.1, 1.1 Hz, 1H), 7.47 (td,  $J$  = 7.6, 1.9 Hz, 1H), 7.44–7.39 (m, 2H), 7.36–7.30 (m, 1H), 7.11 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 7.06 (ddd,  $J$  = 7.5, 4.9, 1.2 Hz, 1H), 6.97 (dt,  $J$  = 7.8, 1.1 Hz, 1H), 6.56 (td,  $J$  = 6.8, 1.2 Hz, 1H), 3.97 (s, 2H), 2.97–2.91 (m, 2H), 2.89–2.84 (m, 2H), 2.22 (s, 3H);  $^{13}C$  NMR (100 MHz,  $CDCl_3$ )  $\delta$  160.39, 149.19, 144.97, 144.71, 136.26, 134.63, 128.94, 128.47, 127.68, 125.68, 124.46, 123.23, 121.21, 117.16, 116.98, 111.54, 56.91, 51.58, 41.23, 35.99; HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{22}H_{23}N_4$  343.1917; Found 343.1918.



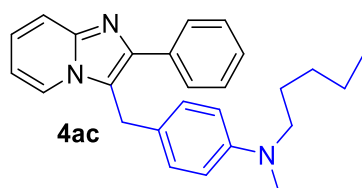
*N*-Methyl-*N*-((2-(4-(methylsulfonyl)phenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **3at** (64 mg, Yield = 82%,  $R_f$  = 0.3 (PE/EA = 1:1)) was isolated as a light brown paste.  $^1H$  NMR (400 MHz,  $CDCl_3$ )  $\delta$  8.12 (d,  $J$  = 7.0 Hz, 1H), 8.02 (s, 4H), 7.70 (d,  $J$  = 8.8 Hz, 1H), 7.37–7.27 (m, 3H), 7.03–6.96 (m, 2H), 6.94–6.89 (m, 1H), 6.87–6.82 (m, 1H), 4.83 (s, 2H), 3.09 (s, 3H), 2.71 (s, 3H);  $^{13}C$  NMR (100 MHz,  $CDCl_3$ )  $\delta$  150.18, 145.49, 143.31, 139.91, 139.45, 129.48, 129.41, 127.69, 125.55, 124.72, 119.29, 117.71, 115.02, 113.06, 46.20, 44.57, 36.61, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{22}H_{22}N_3OS$  392.1427; Found 392.1429.



*N,N*-Dimethyl-4-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4aa** (43 mg, Yield = 66%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.84–7.79 (m, 2H), 7.74 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.67 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.46–7.40 (m, 2H), 7.37–7.32 (m, 1H), 7.16 (ddd,  $J = 9.0, 6.7, 1.3$  Hz, 1H), 7.01 (d,  $J = 8.7$  Hz, 2H), 6.72–6.65 (m, 3H), 4.40 (s, 2H), 2.92 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.73, 144.92, 143.94, 134.78, 128.72, 128.50, 128.35, 127.73, 124.30, 124.14, 123.76, 118.62, 117.54, 113.25, 112.16, 40.78, 29.00; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{22}\text{N}_3$  328.1808; Found 328.1808.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>8, 9</sup>

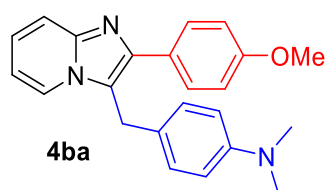


*N,N,3*-Trimethyl-4-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ab** (42 mg, Yield = 59%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.78–7.74 (m, 2H), 7.70 (dt,  $J = 9.0, 1.1$  Hz, 1H), 7.66 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.44–7.39 (m, 2H), 7.35–7.31 (m, 1H), 7.18 (ddd,  $J = 9.1, 6.7, 1.3$  Hz, 1H), 6.72–6.68 (m, 2H), 6.54 (d,  $J = 8.5$  Hz, 1H), 6.41 (dd,  $J = 8.5, 2.8$  Hz, 1H), 4.28 (s, 2H), 2.91 (s, 6H), 2.41 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.73, 144.94, 144.16, 137.03, 134.73, 128.68, 128.24, 127.65, 127.57, 124.08, 123.67, 122.40, 118.30, 117.52, 115.06, 112.17, 110.66, 40.73, 26.83, 20.40; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{24}\text{N}_3$  342.1965; Found 342.1963.

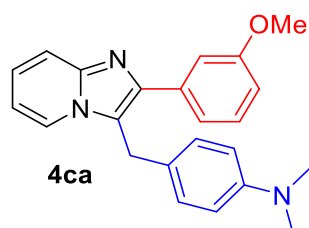


*N*-Methyl-*N*-pentyl-4-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ac** (45 mg, Yield = 59%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,

CDCl<sub>3</sub>)  $\delta$  7.84–7.80 (m, 2H), 7.76 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.68 (dt,  $J$  = 9.0, 1.1 Hz, 1H), 7.46–7.40 (m, 2H), 7.37–7.32 (m, 1H), 7.17 (ddd,  $J$  = 9.1, 6.7, 1.2 Hz, 1H), 6.99 (d,  $J$  = 8.7 Hz, 2H), 6.70 (td,  $J$  = 6.8, 1.2 Hz, 1H), 6.65–6.59 (m, 2H), 4.39 (s, 2H), 3.26 (t,  $J$  = 7.5 Hz, 2H), 2.89 (s, 3H), 1.61–1.51 (m, 2H), 1.37–1.24 (m, 4H), 0.89 (t,  $J$  = 7.0 Hz, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  148.46, 144.86, 143.83, 134.75, 128.71, 128.57, 128.35, 127.72, 124.16, 123.82, 123.36, 118.72, 117.50, 112.62, 112.16, 52.97, 38.41, 29.45, 28.96, 26.47, 22.72, 14.22; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>26</sub>H<sub>30</sub>N<sub>3</sub> 384.2434; Found 384.2437.

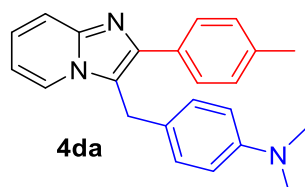


4-((2-(4-Methoxyphenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4ba** (49 mg, Yield = 69%,  $R_f$  = 0.2 (PE/EA = 5:1)) was isolated as a light brown paste. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.80–7.68 (m, 3H), 7.65 (dt,  $J$  = 9.1, 1.2 Hz, 1H), 7.17–7.09 (m, 1H), 7.03–6.92 (m, 4H), 6.70–6.62 (m, 3H), 4.36 (s, 2H), 3.82 (s, 3H), 2.90 (s, 6H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  159.34, 149.66, 144.75, 143.72, 129.45, 128.43, 127.35, 124.35, 123.91, 123.57, 117.87, 117.25, 114.13, 113.20, 111.97, 55.35, 40.72, 28.93; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>23</sub>H<sub>24</sub>N<sub>3</sub>O 358.1914; Found 358.1916. <sup>1</sup>H NMR and <sup>13</sup>C NMR data are consistent with the reported values.<sup>8</sup>

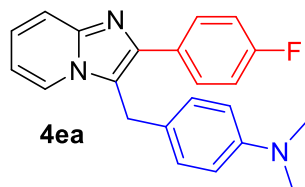


4-((2-(3-Methoxyphenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4ca** (42 mg, Yield = 59%,  $R_f$  = 0.2 (PE/EA = 5:1)) was isolated as a light brown paste. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.75 (dt,  $J$  = 6.9, 1.2 Hz, 1H), 7.67 (dt,  $J$  = 9.1, 1.2 Hz, 1H), 7.40 (dd,  $J$  = 2.7, 1.5 Hz, 1H), 7.37–7.32 (m, 2H), 7.17 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 7.03–6.98 (m, 2H), 6.90 (ddd,  $J$  = 7.9, 2.6, 1.4 Hz, 1H), 6.72–6.65 (m, 3H), 4.41 (s, 2H), 3.82 (s, 3H), 2.91 (s, 6H); <sup>13</sup>C NMR (100

MHz, CDCl<sub>3</sub>)  $\delta$  159.98, 149.74, 144.86, 143.83, 136.19, 129.68, 128.51, 124.36, 124.16, 123.75, 120.73, 118.84, 117.59, 114.16, 113.28, 112.20, 55.44, 40.80, 29.04, (1C is merged with other peaks); HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>23</sub>H<sub>24</sub>N<sub>3</sub>O 358.1914; Found 358.1915.

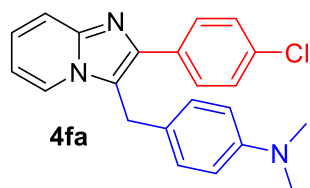


*N,N*-Dimethyl-4-((2-(*p*-tolyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4da** (42 mg, Yield = 61%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown paste. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.75–7.63 (m, 4H), 7.27–7.22 (m, 2H), 7.18–7.11 (m, 1H), 7.01 (d,  $J$  = 12.2 Hz, 2H), 6.72–6.62 (m, 3H), 4.38 (s, 2H), 2.91 (s, 6H), 2.39 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  149.68, 144.81, 143.93, 137.45, 131.85, 129.41, 128.48, 128.18, 124.38, 123.98, 123.65, 118.31, 117.39, 113.21, 112.03, 40.74, 28.99, 21.37; HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>23</sub>H<sub>24</sub>N<sub>3</sub> 342.1965; Found 342.1962. <sup>1</sup>H NMR and <sup>13</sup>C NMR data are consistent with the reported values.<sup>8</sup>

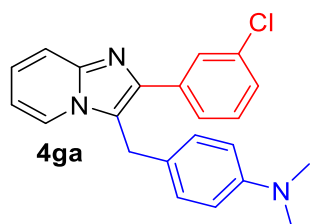


4-((2-(4-Fluorophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4ea** (46 mg, Yield = 67%,  $R_f$  = 0.3 (PE/EA = 2:1)) was isolated as a light brown solid; mp 94–95 °C. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.78–7.72 (m, 3H), 7.66 (dt,  $J$  = 9.1, 1.2 Hz, 1H), 7.17 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 7.14–7.08 (m, 2H), 7.02–6.96 (m, 2H), 6.73–6.65 (m, 3H), 4.36 (s, 2H), 2.91 (s, 6H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$  162.64 (d,  $J$  = 245.4 Hz), 149.76, 144.87, 143.03, 130.89 (d,  $J$  = 3.3 Hz), 129.97 (d,  $J$  = 8.1 Hz), 128.42, 124.30, 124.04, 123.75, 118.41, 117.47, 115.66 (d,  $J$  = 21.4 Hz), 113.24, 112.27, 40.75, 28.90; <sup>19</sup>F NMR (376 MHz, CDCl<sub>3</sub>)  $\delta$  –114.50 (s, 1F). HRMS (ESI)  $m/z$ : [M + H]<sup>+</sup> Calcd for C<sub>22</sub>H<sub>21</sub>N<sub>3</sub>F 346.1714; Found 346.1712.

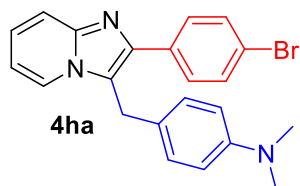




4-((2-(4-Chlorophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4fa** (37 mg, Yield = 51%,  $R_f$  = 0.3 (PE/EA = 2:1)) was isolated as a light brown solid; mp 124–126 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.76–7.71 (m, 3H), 7.66 (dt,  $J$  = 9.1, 1.1 Hz, 1H), 7.41–7.36 (m, 2H), 7.18 (ddd,  $J$  = 9.1, 6.7, 1.3 Hz, 1H), 7.00–6.96 (m, 2H), 6.71 (td,  $J$  = 6.7, 1.1 Hz, 1H), 6.69–6.65 (m, 2H), 4.37 (s, 2H), 2.91 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.79, 144.97, 142.77, 133.66, 133.33, 129.53, 128.91, 128.43, 124.40, 123.94, 123.76, 118.80, 117.56, 113.26, 112.34, 40.74, 28.94; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{21}\text{N}_3\text{Cl}$  362.1419; Found 362.1415.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>9</sup>

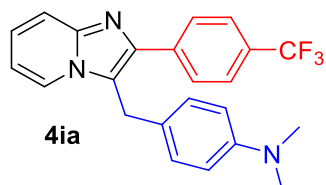


4-((2-(3-Chlorophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4ga** (37 mg, Yield = 51%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.89–7.85 (m, 1H), 7.75 (dt,  $J$  = 6.8, 1.2 Hz, 1H), 7.68–7.62 (m, 2H), 7.36–7.29 (m, 2H), 7.18 (ddd,  $J$  = 9.0, 6.7, 1.3 Hz, 1H), 7.02–6.96 (m, 2H), 6.75–6.65 (m, 3H), 4.39 (s, 2H), 2.91 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.78, 144.96, 142.49, 136.71, 134.73, 129.91, 128.47, 128.40, 127.76, 126.30, 124.46, 123.93, 123.83, 119.18, 117.66, 113.27, 112.38, 40.75, 28.96; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{21}\text{N}_3\text{Cl}$  362.1419; Found 362.1417.



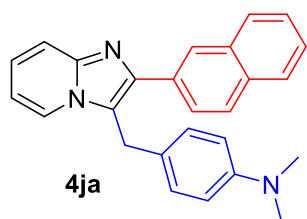
4-((2-(4-Bromophenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4ha** (55 mg, Yield = 68%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown solid; mp 108–110 °C.

$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.73 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.70–7.63 (m, 3H), 7.56–7.50 (m, 2H), 7.17 (ddd,  $J = 9.1, 6.7, 1.3$  Hz, 1H), 7.01–6.94 (m, 2H), 6.73–6.62 (m, 3H), 4.35 (s, 2H), 2.91 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.74, 144.92, 142.68, 133.73, 131.81, 129.79, 128.38, 124.40, 123.84, 123.71, 121.85, 118.81, 117.51, 113.21, 112.32, 40.69, 28.89; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{21}\text{N}_3\text{Br}$  406.0913; Found 406.0914.



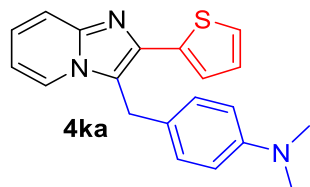
*N,N*-Dimethyl-4-((2-(4-(trifluoromethyl)phenyl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline.

Compound **4ia** (40 mg, Yield = 50%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.92 (d,  $J = 7.9$  Hz, 2H), 7.78 (dt,  $J = 6.8, 1.2$  Hz, 1H), 7.71–7.65 (m, 3H), 7.21 (ddd,  $J = 9.1, 6.7, 1.2$  Hz, 1H), 7.03–6.96 (m, 2H), 6.74 (td,  $J = 6.8, 1.2$  Hz, 1H), 6.70–6.65 (m, 2H), 4.41 (s, 2H), 2.92 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.84, 145.11, 142.41, 138.40, 129.22 (q,  $J = 32.2$  Hz), 128.42, 125.66 (q,  $J = 3.9$  Hz), 124.68, 124.45 (q,  $J = 271.0$  Hz), 123.85, 123.72, 119.56, 117.77, 113.28, 112.56, 40.75, 28.96, (1C is merged with other peaks);  $^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ )  $\delta$  -62.30 (s, 3F). HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{21}\text{N}_3\text{F}_3$  396.1682; Found 396.1683.

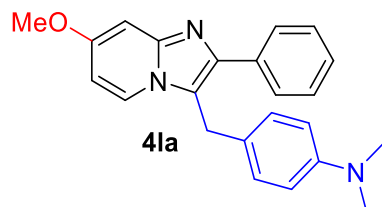


*N,N*-Dimethyl-4-((2-(naphthalen-2-yl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ja** (46 mg, Yield = 61%,  $R_f = 0.2$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.30 (s, 1H), 7.97 (dd,  $J = 8.5, 1.8$  Hz, 1H), 7.93–7.83 (m, 3H), 7.79 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.73 (dt,  $J = 9.0, 1.1$  Hz, 1H), 7.51–7.45 (m, 2H), 7.20 (ddd,  $J = 9.0, 6.7, 1.3$  Hz, 1H), 7.01 (d,  $J = 8.6$  Hz, 2H), 6.76–6.67 (m, 3H), 4.48 (s, 2H), 2.93 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.71, 144.96, 143.70, 133.65, 132.92, 132.20, 128.52, 128.45, 128.29, 127.74, 127.15, 126.41,

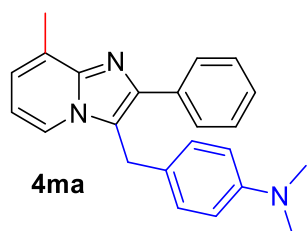
126.17, 126.04, 124.27, 123.73, 119.09, 117.48, 113.25, 112.23, 40.73, 29.09, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{26}H_{24}N_3$  378.1965; Found 378.1962.



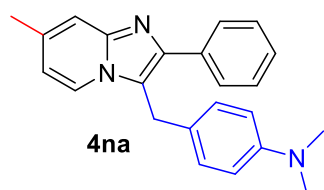
*N,N*-Dimethyl-4-((2-(thiophen-2-yl)imidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ka** (47 mg, Yield = 70%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1H$  NMR (400 MHz,  $CDCl_3$ )  $\delta$  7.75 (dt,  $J$  = 7.0, 1.2 Hz, 1H), 7.63 (dt,  $J$  = 9.1, 1.1 Hz, 1H), 7.42 (dd,  $J$  = 3.6, 1.1 Hz, 1H), 7.34 (dd,  $J$  = 5.1, 1.1 Hz, 1H), 7.14 (ddd,  $J$  = 9.2, 6.7, 1.3 Hz, 1H), 7.08 (dd,  $J$  = 5.1, 3.6 Hz, 1H), 7.05–6.99 (m, 2H), 6.70–6.62 (m, 3H), 4.44 (s, 2H), 2.89 (s, 6H);  $^{13}C$  NMR (100 MHz,  $CDCl_3$ )  $\delta$  149.71, 144.77, 138.31, 137.85, 128.53, 127.79, 125.47, 124.55, 124.37, 123.83, 123.45, 118.32, 117.31, 113.16, 112.29, 40.70, 28.95; HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{20}H_{20}N_3S$  334.1372; Found 334.1375.  $^1H$  NMR and  $^{13}C$  NMR data are consistent with the reported values.<sup>8</sup>



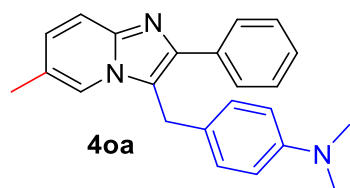
4-((7-Methoxy-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4la** (57 mg, Yield = 80%,  $R_f$  = 0.25 (PE/EA = 5:1)) was isolated as a light brown paste.  $^1H$  NMR (400 MHz,  $CDCl_3$ )  $\delta$  7.81–7.77 (m, 2H), 7.54 (dd,  $J$  = 7.5, 0.7 Hz, 1H), 7.44–7.38 (m, 2H), 7.35–7.29 (m, 1H), 7.04–6.98 (m, 2H), 6.95 (d,  $J$  = 2.6 Hz, 1H), 6.69–6.63 (m, 2H), 6.40 (dd,  $J$  = 7.4, 2.5 Hz, 1H), 4.34 (s, 2H), 3.85 (s, 3H), 2.91 (s, 6H);  $^{13}C$  NMR (100 MHz,  $CDCl_3$ )  $\delta$  157.73, 149.66, 146.19, 143.10, 134.89, 128.64, 128.45, 128.02, 127.46, 124.55, 124.22, 117.35, 113.20, 107.08, 94.72, 55.54, 40.75, 28.89; HRMS (ESI)  $m/z$ :  $[M + H]^+$  Calcd for  $C_{23}H_{24}N_3O$  358.1914; Found 358.1912.



*N,N*-Dimethyl-4-((8-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ma** (40 mg, Yield = 59%,  $R_f = 0.4$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 91–92 °C.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.82–7.78 (m, 2H), 7.62 (d,  $J = 6.9$  Hz, 1H), 7.45–7.40 (m, 2H), 7.35–7.31 (m, 1H), 7.03–6.98 (m, 2H), 6.96 (d,  $J = 6.9$  Hz, 1H), 6.70–6.65 (m, 2H), 6.62 (t,  $J = 6.8$  Hz, 1H), 4.37 (s, 2H), 2.91 (s, 6H), 2.69 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.65, 145.34, 143.54, 135.09, 128.68, 128.53, 127.55, 127.45, 124.66, 122.91, 121.63, 118.94, 113.22, 112.13, 40.79, 29.08, 17.32, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{24}\text{N}_3$  342.1965; Found 342.1966.  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR data are consistent with the reported values.<sup>8</sup>

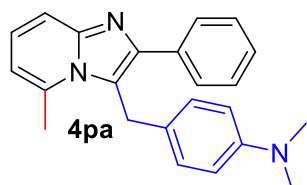


*N,N*-Dimethyl-4-((7-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4na** (40 mg, Yield = 59%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.83–7.78 (m, 2H), 7.60 (d,  $J = 7.0$  Hz, 1H), 7.44–7.39 (m, 3H), 7.35–7.30 (m, 1H), 7.02–6.98 (m, 2H), 6.69–6.65 (m, 2H), 6.51 (dd,  $J = 6.9, 1.7$  Hz, 1H), 4.36 (s, 2H), 2.91 (s, 6H), 2.37 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.65, 145.28, 143.44, 134.99, 134.90, 128.62, 128.44, 128.22, 127.52, 124.52, 122.94, 117.94, 115.86, 114.72, 113.19, 40.72, 28.90, 21.38; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{24}\text{N}_3$  342.1965; Found 342.1962.

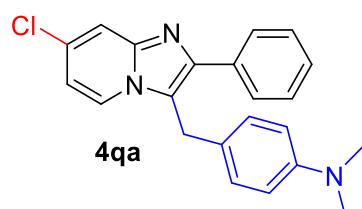


*N,N*-Dimethyl-4-((6-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4oa** (31 mg, Yield = 46%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown solid; mp 148–150 °C.

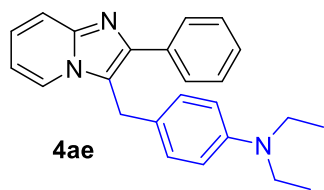
$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.82–7.77 (m, 2H), 7.58 (dd,  $J = 9.2, 1.0$  Hz, 1H), 7.54–7.51 (m, 1H), 7.44–7.39 (m, 2H), 7.35–7.29 (m, 1H), 7.04–6.99 (m, 3H), 6.72–6.65 (m, 2H), 4.37 (s, 2H), 2.92 (s, 6H), 2.24 (d,  $J = 1.1$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.63, 143.96, 143.65, 134.89, 128.65, 128.45, 128.16, 127.53, 127.30, 124.51, 121.72, 121.23, 118.28, 116.84, 113.21, 40.75, 28.90, 18.52; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{24}\text{N}_3$  342.1965; Found 342.1966.



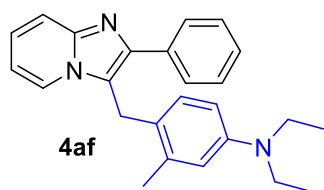
*N,N*-Dimethyl-4-((5-methyl-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4pa** (23 mg, Yield = 34%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.70–7.66 (m, 2H), 7.54 (dt,  $J = 8.9, 1.0$  Hz, 1H), 7.40–7.35 (m, 2H), 7.34–7.30 (m, 1H), 7.03 (dd,  $J = 9.0, 6.8$  Hz, 1H), 6.91–6.87 (m, 2H), 6.72–6.65 (m, 2H), 6.40 (dt,  $J = 6.8, 1.1$  Hz, 1H), 4.56 (s, 2H), 2.92 (s, 6H), 2.64 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.29, 146.86, 145.60, 136.59, 135.04, 128.91, 128.75, 128.50, 128.31, 127.63, 124.45, 119.89, 115.88, 113.50, 113.29, 40.76, 30.76, 20.18; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{23}\text{H}_{24}\text{N}_3$  342.1965; Found 342.1966.



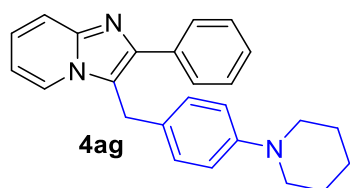
4-((7-Chloro-2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)-*N,N*-dimethylaniline. Compound **4qa** (39 mg, Yield = 54%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.81–7.75 (m, 2H), 7.68–7.61 (m, 2H), 7.46–7.41 (m, 2H), 7.38–7.34 (m, 1H), 7.01–6.96 (m, 2H), 6.71–6.64 (m, 3H), 4.38 (s, 2H), 2.92 (s, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  149.81, 144.81, 144.61, 134.35, 130.67, 128.79, 128.45, 128.32, 127.99, 124.09, 123.73, 118.97, 116.36, 113.74, 113.26, 40.73, 28.94; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{22}\text{H}_{21}\text{N}_3\text{Cl}$  362.1419; Found 362.1416.



*N,N*-Diethyl-4-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4ae** (60 mg, Yield = 85%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.85–7.81 (m, 2H), 7.77 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.67 (dt,  $J = 9.1, 1.1$  Hz, 1H), 7.46–7.39 (m, 2H), 7.37–7.32 (m, 1H), 7.16 (ddd,  $J = 9.0, 6.7, 1.3$  Hz, 1H), 7.01–6.95 (m, 2H), 6.70 (td,  $J = 6.8, 1.2$  Hz, 1H), 6.64–6.58 (m, 2H), 4.38 (s, 2H), 3.32 (q,  $J = 7.1$  Hz, 4H), 1.14 (t,  $J = 7.0$  Hz, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  146.87, 144.87, 143.85, 134.80, 128.69, 128.34, 127.68, 124.09, 123.84, 122.91, 118.74, 117.48, 112.34, 112.10, 44.42, 28.92, 12.67, (1C is merged with other peaks); HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{24}\text{H}_{26}\text{N}_3$  356.2121; Found 356.2123.



*N,N*-Diethyl-3-methyl-4-((2-phenylimidazo[1,2-*a*]pyridin-3-yl)methyl)aniline. Compound **4af** (66 mg, Yield = 89%,  $R_f = 0.3$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.78–7.74 (m, 2H), 7.71–7.67 (m, 2H), 7.44–7.39 (m, 2H), 7.36–7.31 (m, 1H), 7.18 (ddd,  $J = 9.0, 6.7, 1.2$  Hz, 1H), 6.71 (td,  $J = 6.8, 1.1$  Hz, 1H), 6.62 (d,  $J = 2.8$  Hz, 1H), 6.50 (d,  $J = 8.5$  Hz, 1H), 6.35 (dd,  $J = 8.6, 2.8$  Hz, 1H), 4.27 (s, 2H), 3.35–3.28 (m, 4H), 2.39 (s, 3H), 1.14 (t,  $J = 7.1$  Hz, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  146.89, 144.93, 144.12, 137.23, 134.76, 128.70, 128.29, 127.79, 127.66, 124.09, 123.81, 121.05, 118.46, 117.50, 114.07, 112.17, 109.77, 44.34, 26.79, 20.54, 12.75; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{25}\text{H}_{28}\text{N}_3$  370.2278; Found 370.2276.



2-Phenyl-3-(4-(piperidin-1-yl)benzyl)imidazo[1,2-*a*]pyridine. Compound **4ag** (38 mg, Yield = 52%,  $R_f = 0.25$  (PE/EA = 5:1)) was isolated as a light brown paste.  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.82–

7.78 (m, 2H), 7.72 (dt,  $J = 6.9, 1.2$  Hz, 1H), 7.68 (dt,  $J = 9.1, 1.2$  Hz, 1H), 7.45–7.40 (m, 2H), 7.37–7.32 (m, 1H), 7.17 (ddd,  $J = 9.1, 6.7, 1.3$  Hz, 1H), 7.04–6.99 (m, 2H), 6.90–6.85 (m, 2H), 6.70 (td,  $J = 6.8, 1.2$  Hz, 1H), 4.41 (s, 2H), 3.15–3.08 (m, 4H), 1.73–1.65 (m, 4H), 1.60–1.53 (m, 2H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  151.25, 144.92, 144.00, 134.73, 128.74, 128.45, 128.35, 127.76, 126.99, 124.20, 123.76, 118.42, 117.57, 117.04, 112.18, 50.72, 29.13, 25.98, 24.35; HRMS (ESI)  $m/z$ :  $[\text{M} + \text{H}]^+$  Calcd for  $\text{C}_{25}\text{H}_{26}\text{N}_3$  368.2121; Found 368.2119.

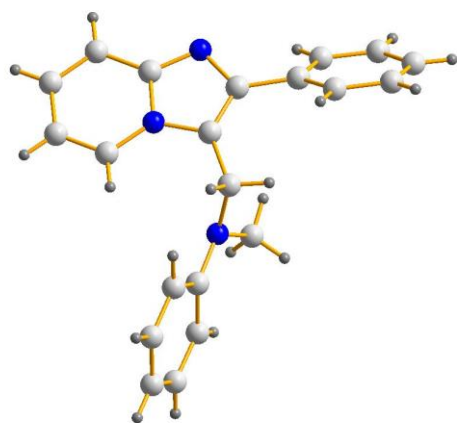
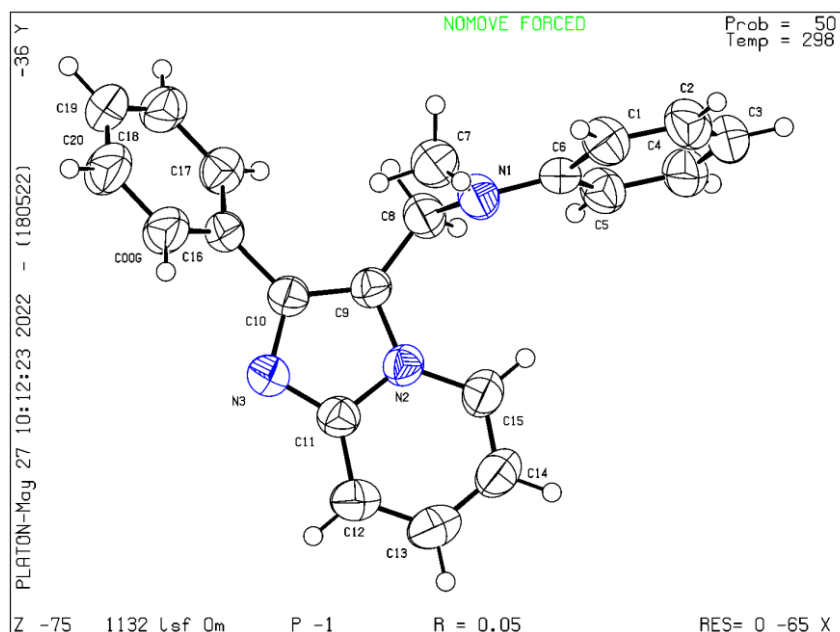
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9. R. Kumar; D. Rawat; S. Adimurthy, Polyethylene Glycol (PEG-400) as Methylene Spacer and Green Solvent for the Synthesis of Heterodiarylmethanes under Metal-Free Conditions. *Eur. J. Org. Chem.* 2020, **2020**, 3499-3507.

## 10. Crystal data and structure refinement of 3aa, 3la and 4ha

The crystal was prepared in ethyl acetate underneath methanol through evaporation, the structure was measured by 'Bruker APEX-II CCD' diffractometer.

### 10.1 X-ray structure of 3aa



CCDC 2175986

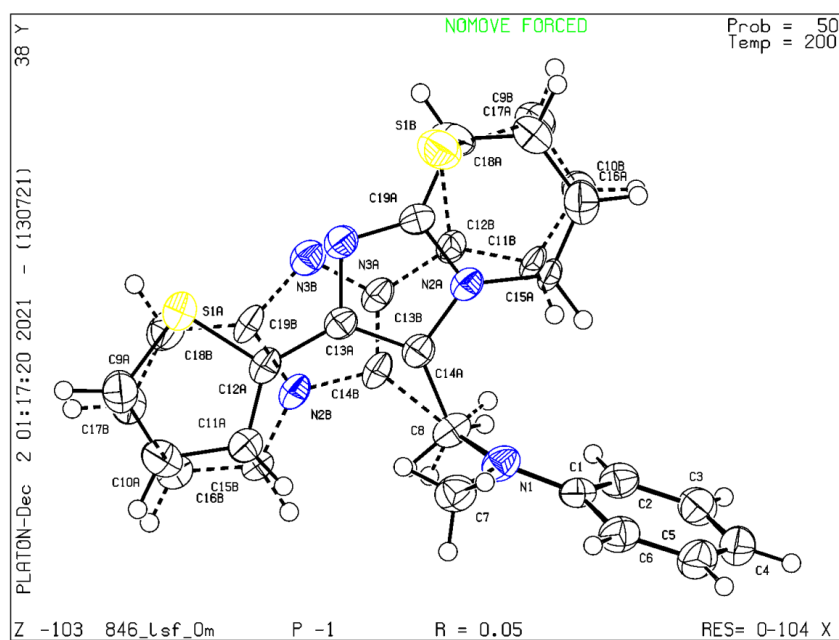
**Table S9 Crystal data and structure refinement for 3aa**

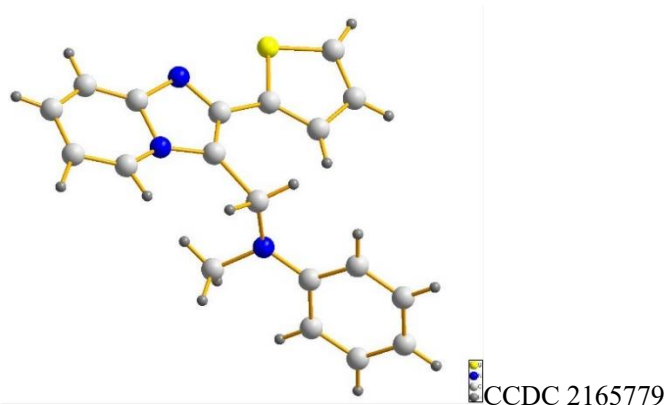
Identification code	1132_LSF_0m
Empirical formula	C <sub>21</sub> H <sub>19</sub> N <sub>3</sub>
Formula weight	313.39
Temperature/K	298.0
Crystal system	triclinic
Space group	P-1



a/Å	8.365(4)
b/Å	10.655(5)
c/Å	10.809(5)
$\alpha$ /°	115.055(14)
$\beta$ /°	99.780(14)
$\gamma$ /°	98.016(15)
Volume/Å <sup>3</sup>	835.6(7)
Z	2
$\rho_{\text{calc}}/\text{cm}^3$	1.246
$\mu/\text{mm}^{-1}$	0.075
F(000)	332.0
Crystal size/mm <sup>3</sup>	0.6 × 0.6 × 0.6
Radiation	MoK $\alpha$ ( $\lambda$ = 0.71073)
2 $\theta$ range for data collection/°	4.302 to 54.966
Index ranges	-9 ≤ h ≤ 10, -13 ≤ k ≤ 13, -14 ≤ l ≤ 14
Reflections collected	13642
Independent reflections	3805 [R <sub>int</sub> = 0.0552, R <sub>sigma</sub> = 0.0503]
Data/restraints/parameters	3805/0/218
Goodness-of-fit on F <sup>2</sup>	1.033
Final R indexes [I ≥ 2 $\sigma$ (I)]	R <sub>1</sub> = 0.0470, wR <sub>2</sub> = 0.1198
Final R indexes [all data]	R <sub>1</sub> = 0.0676, wR <sub>2</sub> = 0.1352
Largest diff. peak/hole / e Å <sup>-3</sup>	0.15/-0.14

## 10.2 X-ray structure of 3la

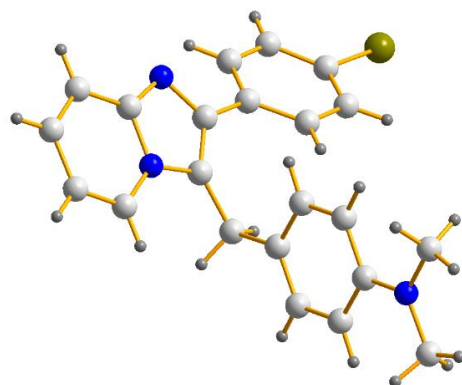
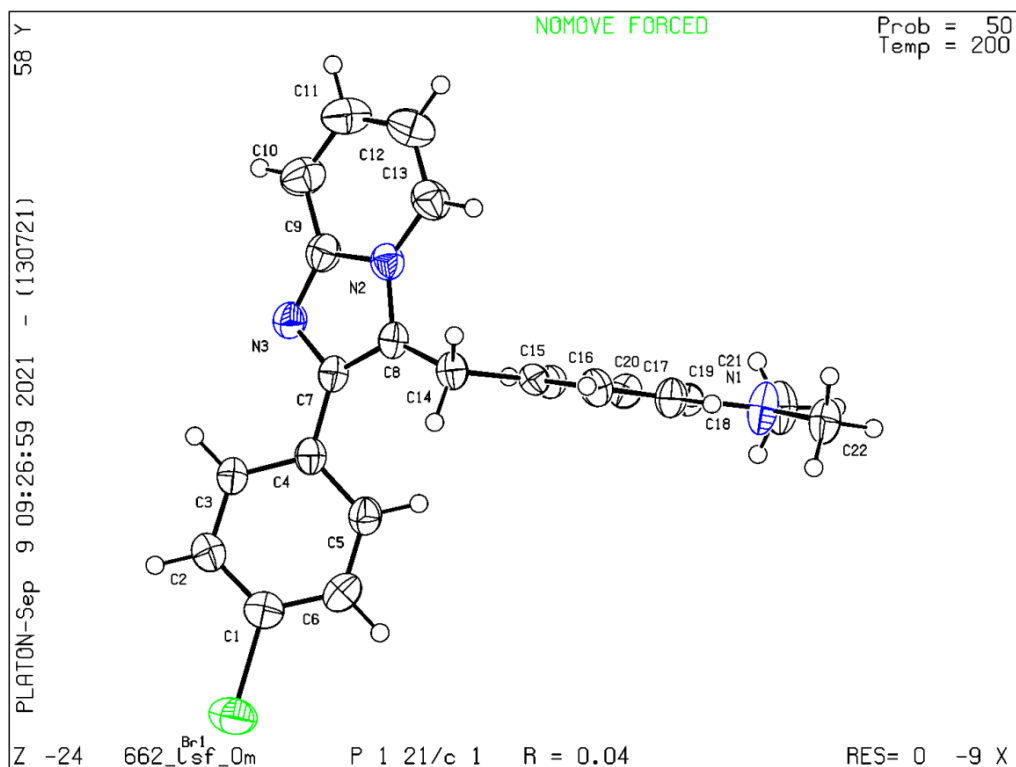




**Table S10 Crystal data and structure refinement for 3la**

Identification code	846_LSF_0m
Empirical formula	C <sub>19</sub> H <sub>17</sub> N <sub>3</sub> S
Formula weight	319.41
Temperature/K	200.0
Crystal system	triclinic
Space group	P-1
a/Å	8.3612(2)
b/Å	9.7442(2)
c/Å	10.6475(2)
α/°	65.8180(10)
β/°	86.3210(10)
γ/°	79.4170(10)
Volume/Å <sup>3</sup>	777.84(3)
Z	2
ρ <sub>calc</sub> /cm <sup>3</sup>	1.364
μ/mm <sup>-1</sup>	1.853
F(000)	336.0
Crystal size/mm <sup>3</sup>	0.2 × 0.1 × 0.08
Radiation	CuKα (λ = 1.54178)
2θ range for data collection/°	9.104 to 136.372
Index ranges	-10 ≤ h ≤ 9, -11 ≤ k ≤ 11, -12 ≤ l ≤ 12
Reflections collected	10004
Independent reflections	2825 [R <sub>int</sub> = 0.0474, R <sub>sigma</sub> = 0.0410]
Data/restraints/parameters	2825/872/324
Goodness-of-fit on F <sup>2</sup>	1.099
Final R indexes [I ≥ 2σ (I)]	R <sub>1</sub> = 0.0466, wR <sub>2</sub> = 0.1197
Final R indexes [all data]	R <sub>1</sub> = 0.0503, wR <sub>2</sub> = 0.1229
Largest diff. peak/hole / e Å <sup>-3</sup>	0.29/-0.28

### 10.3 X-ray structure of 4ha



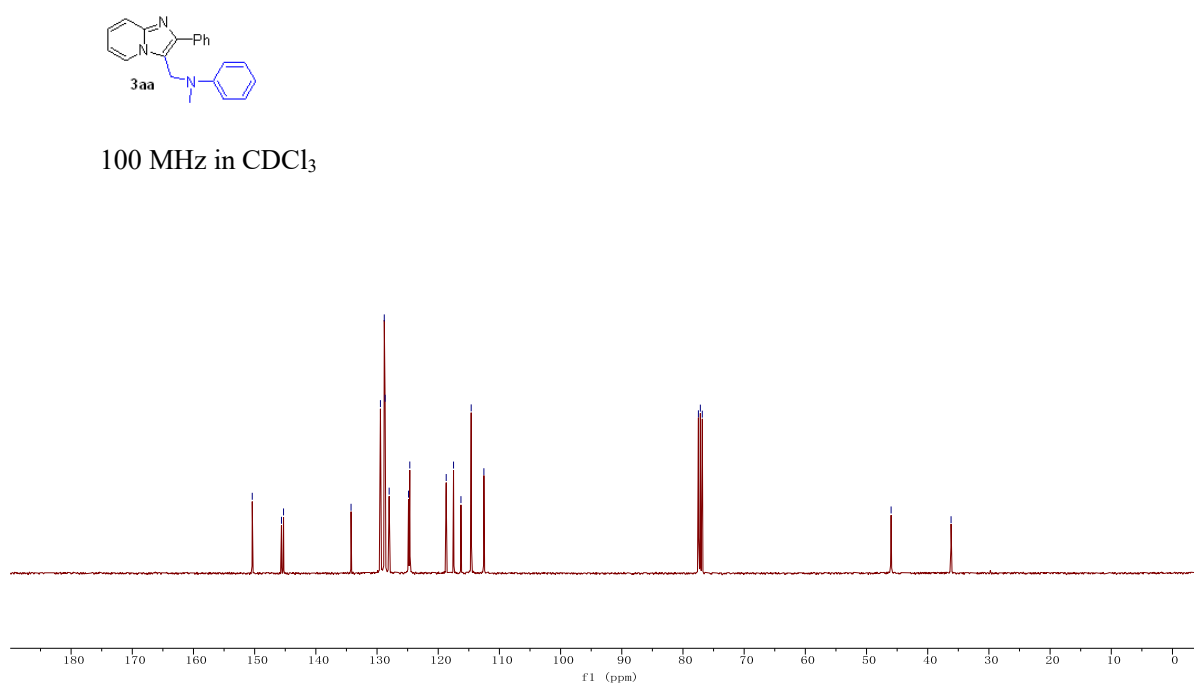
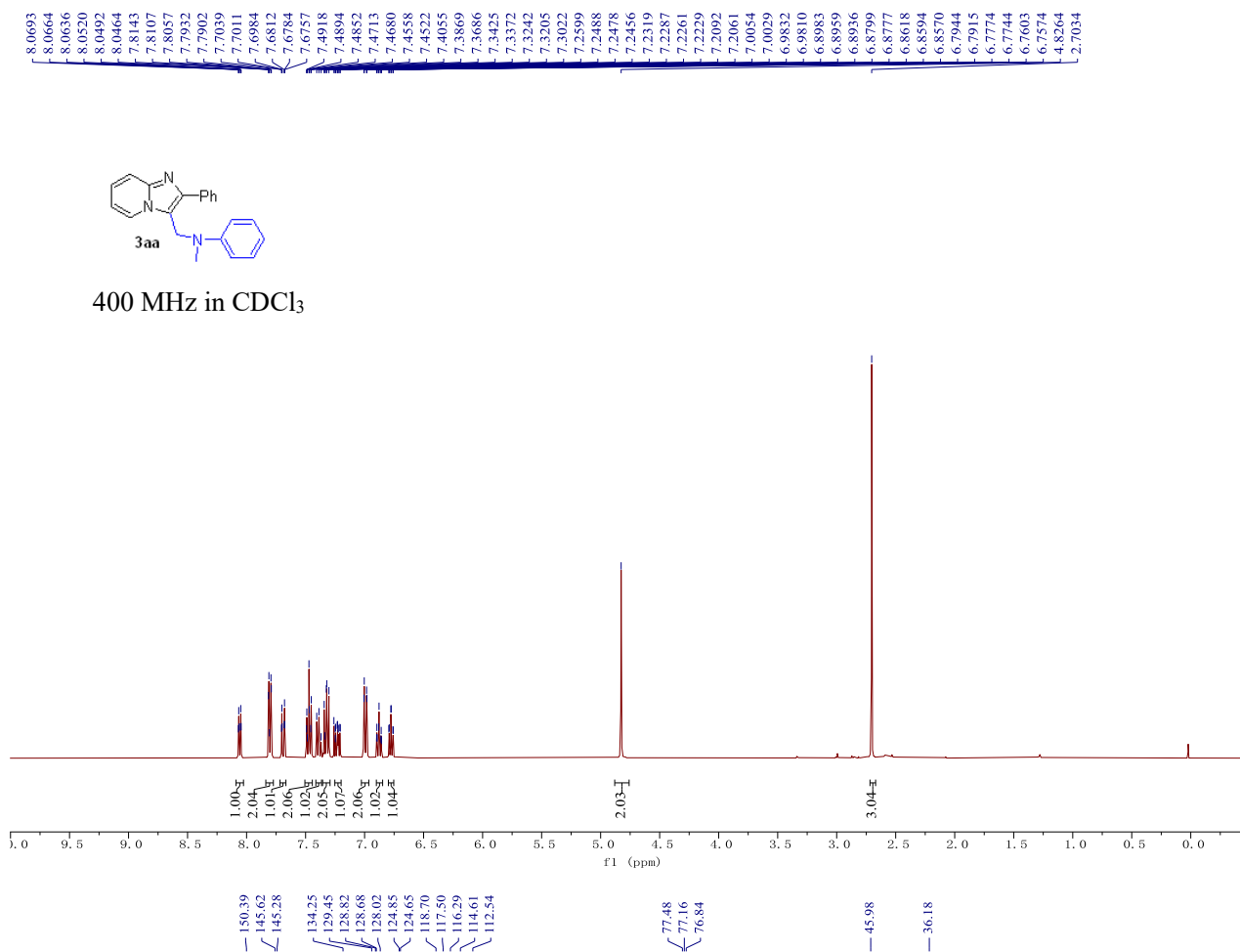
CCDC 2165780

**Table S11 Crystal data and structure refinement for 4ha**

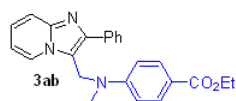
Identification code	662_LSF_0m
Empirical formula	C <sub>22</sub> H <sub>20</sub> BrN <sub>3</sub>
Formula weight	406.32
Temperature/K	200.0
Crystal system	monoclinic
Space group	P2 <sub>1</sub> /c
a/Å	6.3317(2)
b/Å	10.0072(3)
c/Å	29.5714(8)
α/°	90

$\beta/^\circ$	92.360(2)
$\gamma/^\circ$	90
Volume/ $\text{\AA}^3$	1872.13(10)
Z	4
$\rho_{\text{calc}}/\text{g/cm}^3$	1.442
$\mu/\text{mm}^{-1}$	3.057
F(000)	832.0
Crystal size/ $\text{mm}^3$	$0.4 \times 0.03 \times 0.01$
Radiation	CuK $\alpha$ ( $\lambda = 1.54178$ )
$2\theta$ range for data collection/ $^\circ$	5.982 to 136.386
Index ranges	$-7 \leq h \leq 6, -12 \leq k \leq 12, -35 \leq l \leq 34$
Reflections collected	14919
Independent reflections	3421 [ $R_{\text{int}} = 0.0655, R_{\text{sigma}} = 0.0561$ ]
Data/restraints/parameters	3421/0/237
Goodness-of-fit on $F^2$	1.044
Final R indexes [ $I \geq 2\sigma(I)$ ]	$R_1 = 0.0417, wR_2 = 0.1057$
Final R indexes [all data]	$R_1 = 0.0543, wR_2 = 0.1127$
Largest diff. peak/hole / $e \text{\AA}^{-3}$	0.49/-0.53

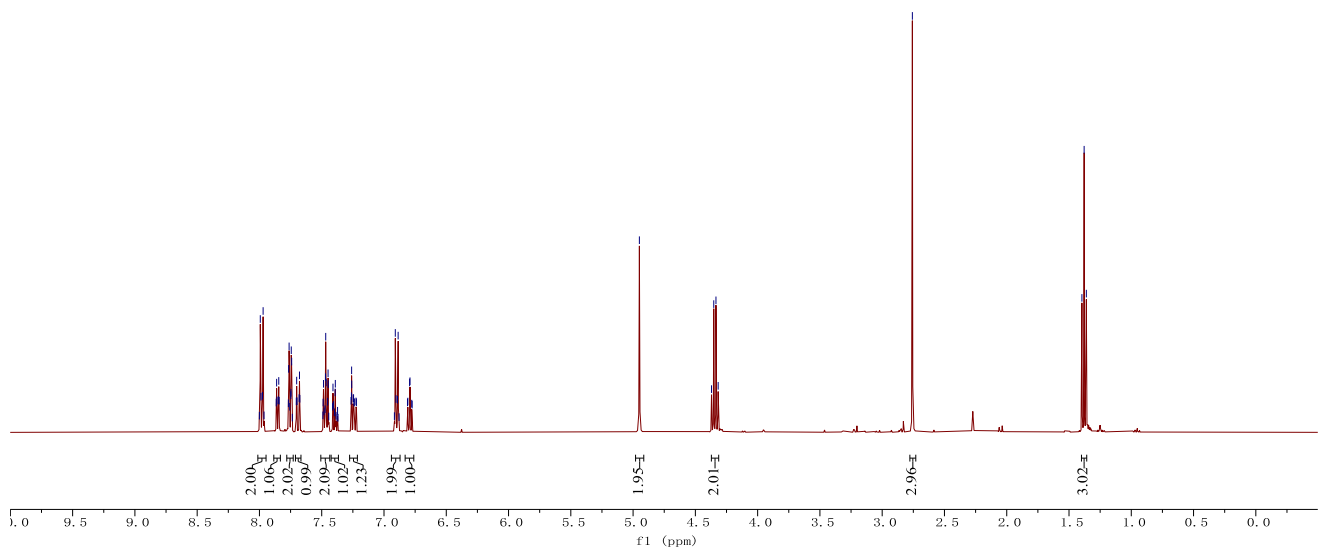
## 11. NMR spectra of C3 methylated imidazo[1,2-a]pyridines



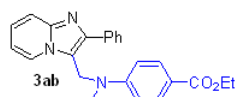
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7.8450  
7.8420  
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7.7656  
7.7619  
7.7569  
7.7486  
7.7470  
7.7445  
7.7415  
7.7040  
7.7012  
7.6983  
7.6813  
7.6785  
7.6757  
7.4906  
7.4886  
7.4858  
7.4815  
7.4729  
7.4696  
7.4679  
7.4642  
7.4526  
7.4491  
7.4474  
7.4131  
7.4096  
7.4062  
7.3968  
7.3922  
7.3908  
7.3728  
7.2643  
7.2615  
7.2600  
7.2595  
7.2475  
7.2443  
7.2416  
7.2384  
7.2247  
7.2215  
6.9092  
6.9040  
6.8917  
6.8865  
6.8113  
6.8084  
6.7943  
6.7913  
6.7773  
6.7742  
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2.7576  
1.3964  
1.3785  
1.3607



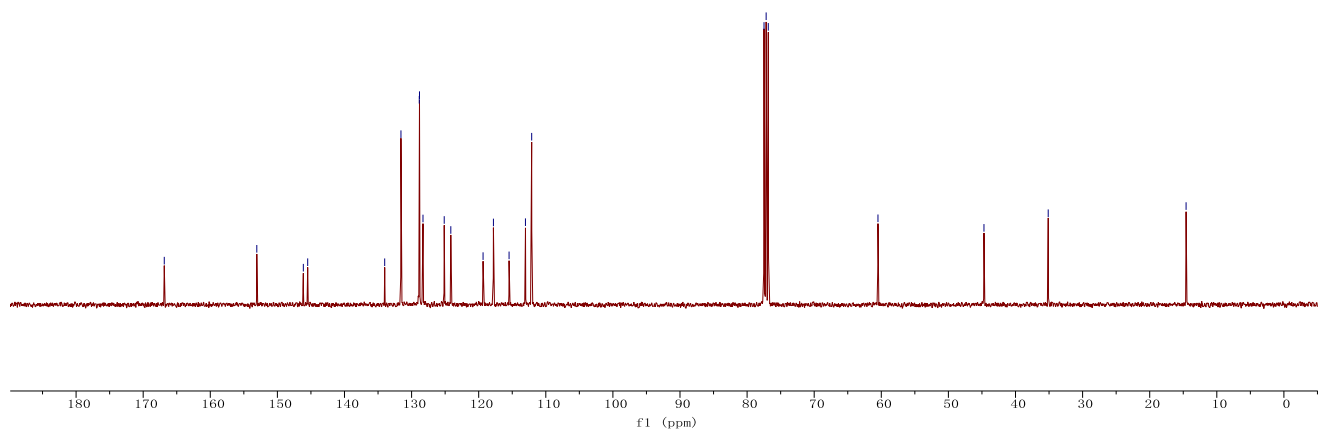
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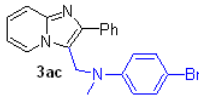
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145.48  
134.01  
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128.30  
125.14  
124.16  
119.35  
117.80  
115.47  
113.03  
112.10  
77.48  
77.16  
76.84  
60.50  
44.70  
35.12  
14.58



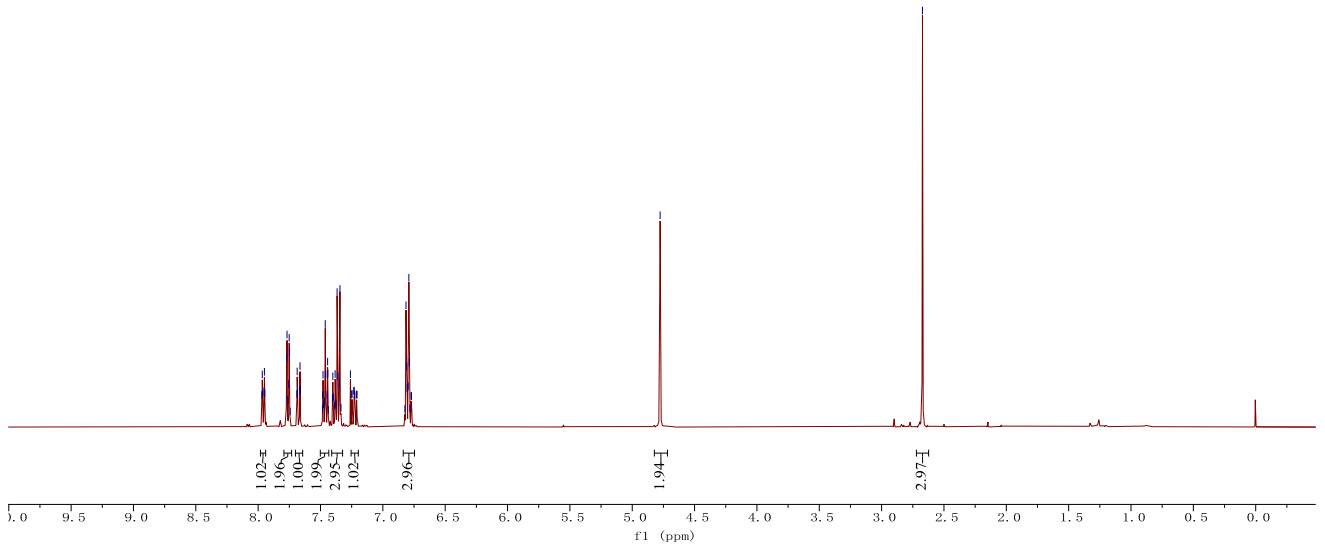
100 MHz in CDCl<sub>3</sub>



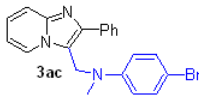
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7.7511  
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7.7428  
7.6899  
7.6870  
7.6843  
7.6672  
7.6643  
7.6616  
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7.4825  
7.4798  
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7.4666  
7.4620  
7.4582  
7.4464  
7.4430  
7.4412  
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7.4010  
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7.3826  
7.3762  
7.3668  
7.3612  
7.3498  
7.3441  
7.3356  
7.2600  
7.2501  
7.2469  
7.2332  
7.2299  
7.2273  
7.2242  
7.2105  
7.2072  
6.8229  
6.8144  
6.8084  
6.8049  
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6.7708  
4.7763  
2.6723



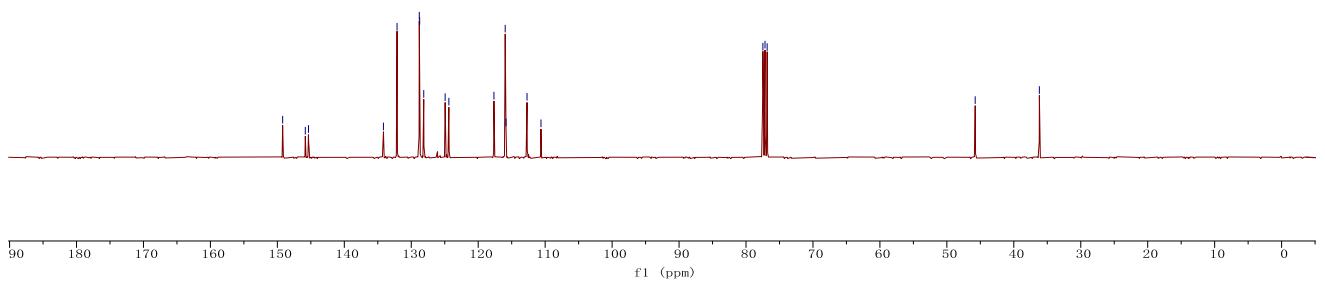
400 MHz in CDCl<sub>3</sub>



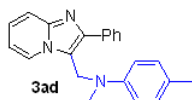
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132.12  
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124.94  
124.38  
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115.85  
112.71  
110.63  
77.48  
77.16  
76.84  
45.77  
36.18



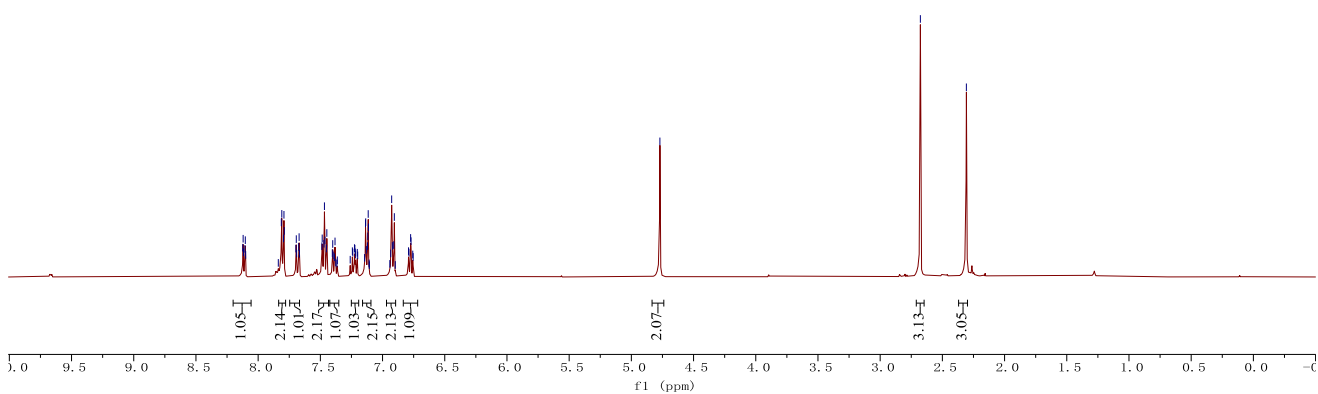
100 MHz in CDCl<sub>3</sub>



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8.1004  
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7.7962  
7.7926  
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7.6963  
7.6934  
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7.6736  
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7.6679  
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7.4671  
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7.1132  
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6.8976  
6.7912  
6.7882  
6.7744  
6.7712  
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4.7706  
2.6769  
2.3070



**3ad**  
400 MHz in CDCl<sub>3</sub>



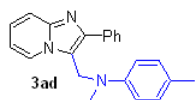
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124.80  
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16.52  
15.29  
112.43

77.48  
77.16  
76.84

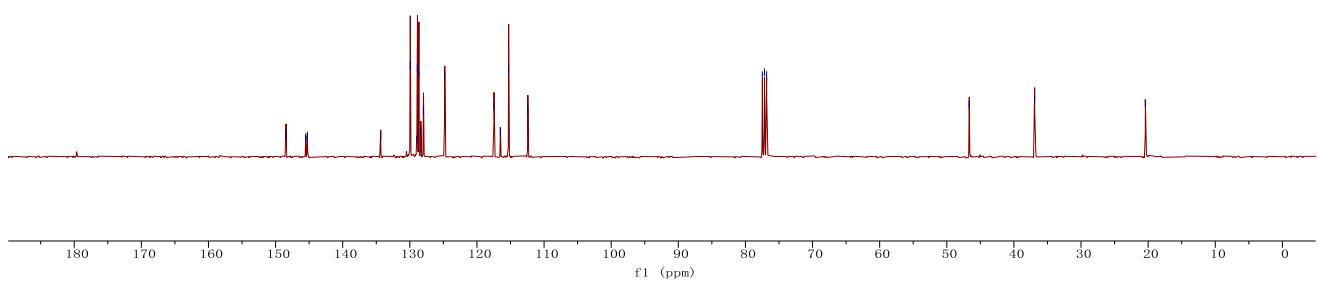
46.66

36.91

20.42

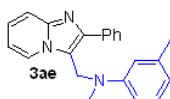


**3ad**  
100 MHz in CDCl<sub>3</sub>

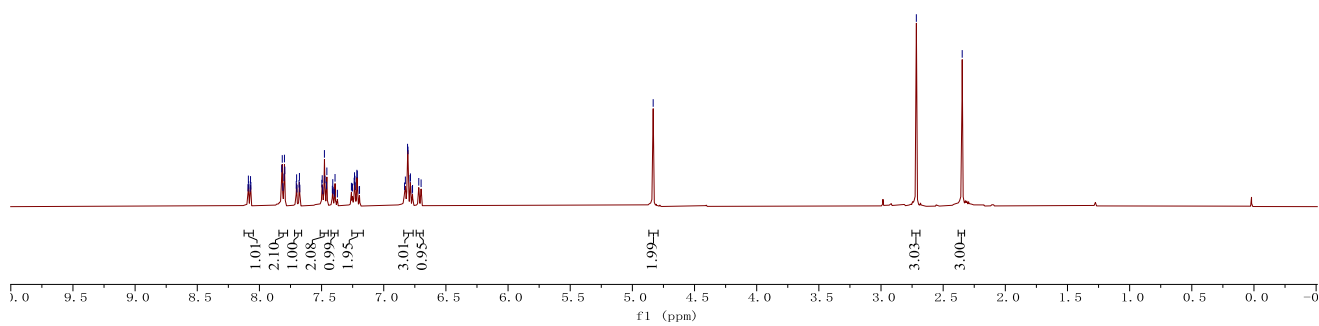




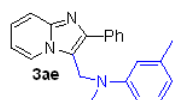
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7.8026  
7.7987  
7.7959  
7.7038  
7.7009  
7.6979  
7.6811  
7.6781  
7.6752  
7.4975  
7.4948  
7.4904  
7.4771  
7.4729  
7.4613  
7.4576  
7.4136  
7.4100  
7.4065  
7.3918  
7.2600  
7.2547  
7.2517  
7.2380  
7.2351  
7.2321  
7.2291  
7.2161  
7.2128  
7.1978  
7.1948  
6.8326  
6.8260  
6.8086  
6.8051  
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4.8326  
2.3457



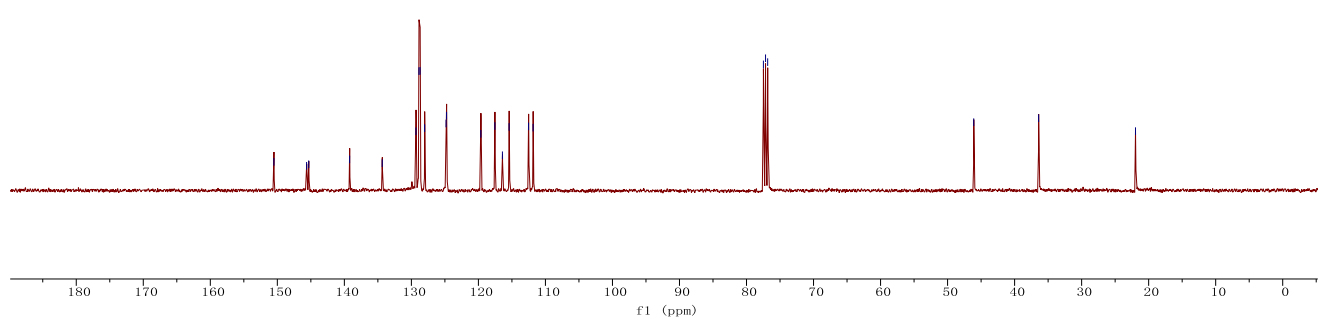
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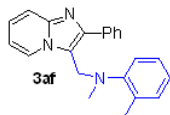
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119.64  
117.55  
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21.96



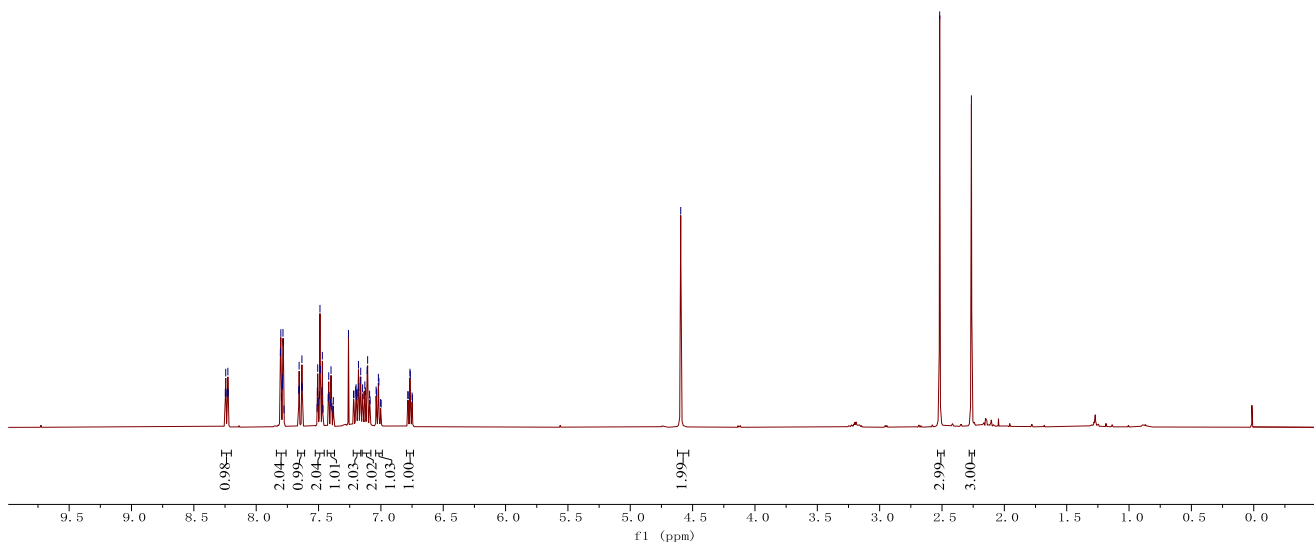
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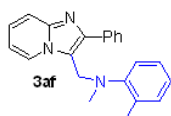
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7.0905  
7.0864  
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7.0201  
7.0160  
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2.2644



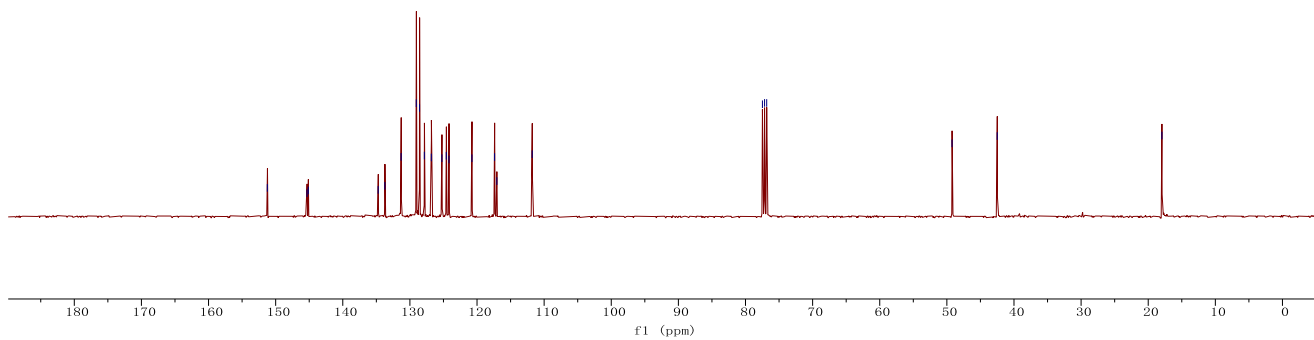
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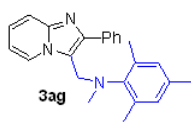
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125.24  
124.58  
124.18  
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76.84  
49.23  
42.50  
17.96



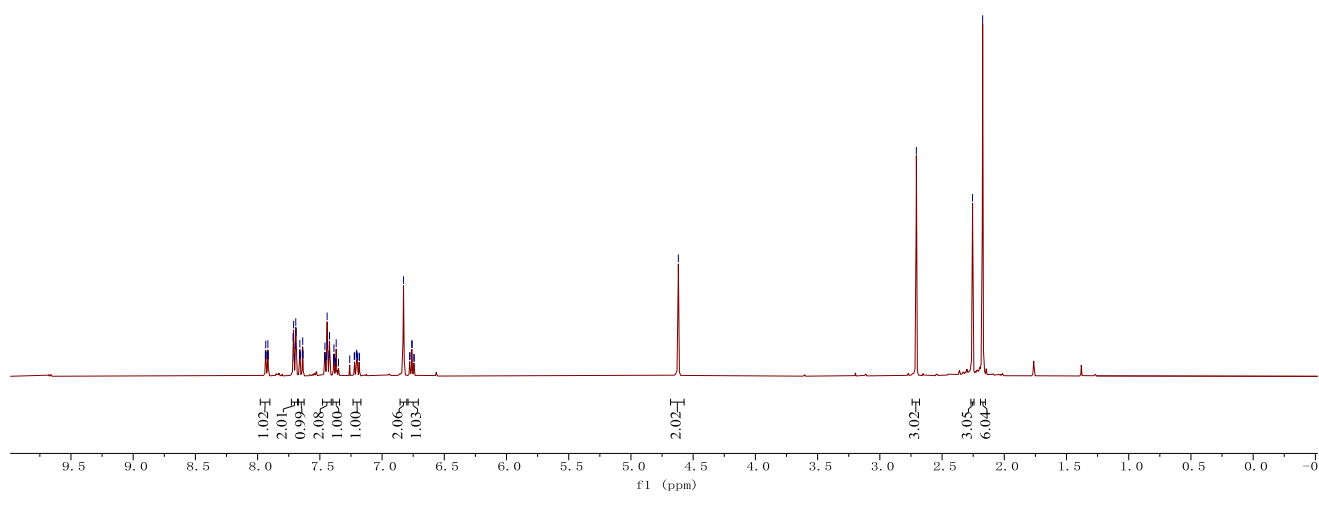
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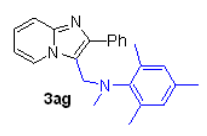
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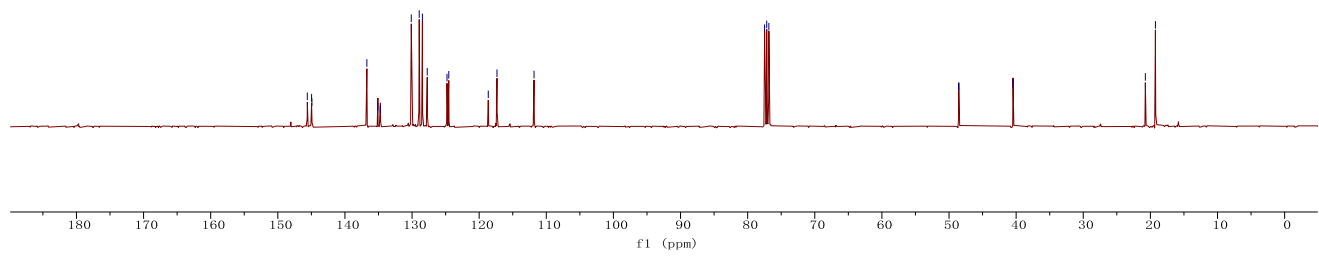
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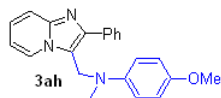
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124.53  
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117.35  
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77.16  
76.84  
48.56  
40.48  
20.75  
19.25



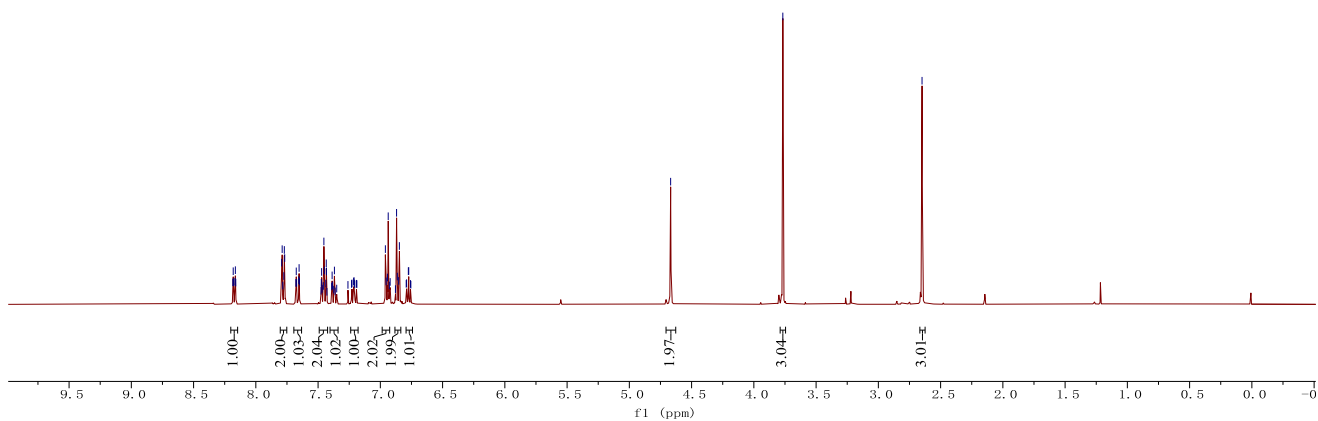
100 MHz in CDCl<sub>3</sub>



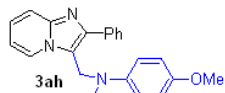
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7.7748  
7.7710  
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7.4740  
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7.4344  
7.4326  
7.3907  
7.3872  
7.3838  
7.3741  
7.3688  
7.3632  
7.3504  
7.2601  
7.2313  
7.2281  
7.2144  
7.2112  
7.2086  
7.2054  
7.1918  
7.1886  
6.9595  
6.9536  
6.9429  
6.9367  
6.9277  
6.9211  
6.8786  
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6.8470  
6.7921  
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6.7720  
6.7579  
6.7549  
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3.7673  
3.6401



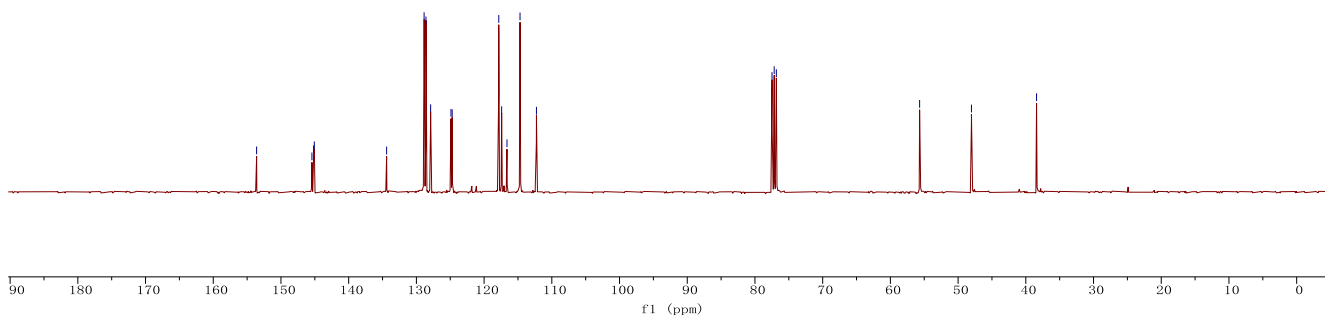
400 MHz in CDCl<sub>3</sub>



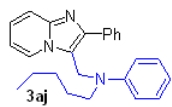
153.60  
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145.18  
145.08  
134.40  
128.86  
128.59  
127.90  
124.89  
124.72  
117.83  
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114.70  
112.27  
77.48  
77.16  
76.84  
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48.03  
38.42



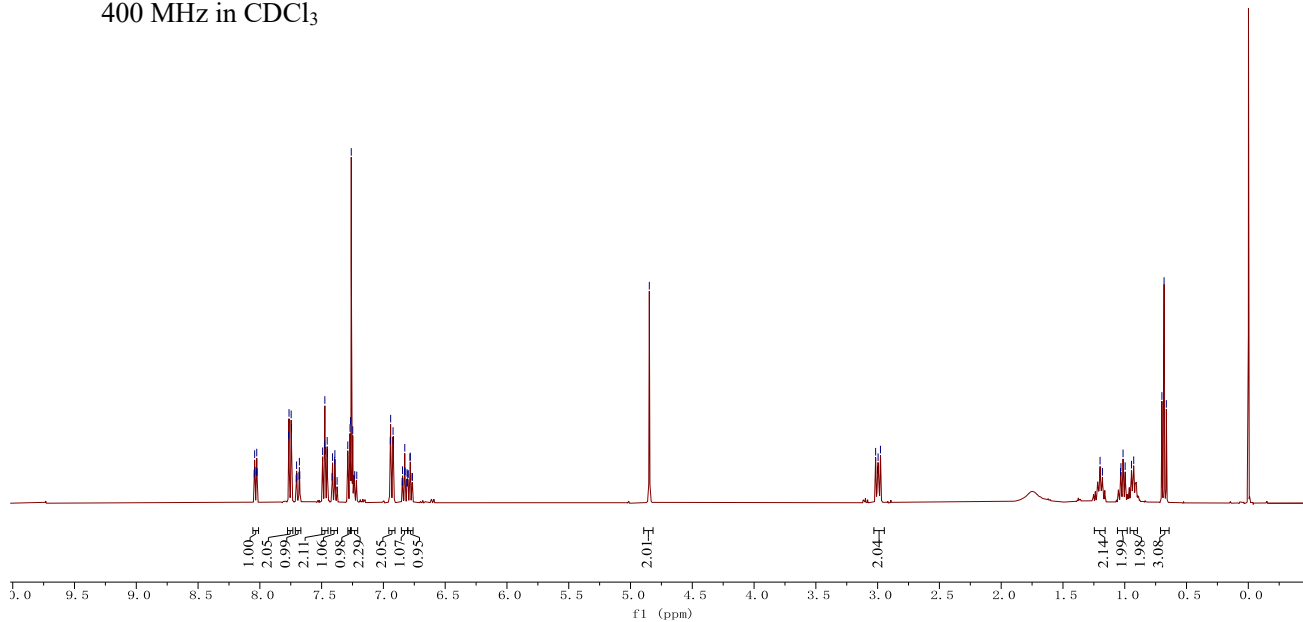
100 MHz in CDCl<sub>3</sub>



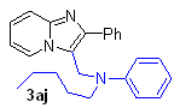
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7.7056  
7.7026  
7.6998  
7.6829  
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7.4118  
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7.2601  
7.2481  
7.2324  
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6.9442  
6.9416  
6.9219  
6.9194  
6.8478  
6.8453  
6.8427  
6.8295  
6.8271  
6.8245  
6.8088  
6.8061  
6.8023  
6.7993  
6.7852  
6.7822  
6.7683  
6.7652  
4.8479  
4.8479  
3.0162  
3.0013  
2.9962  
2.9773  
1.1998  
1.1805  
1.0341  
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0.6634



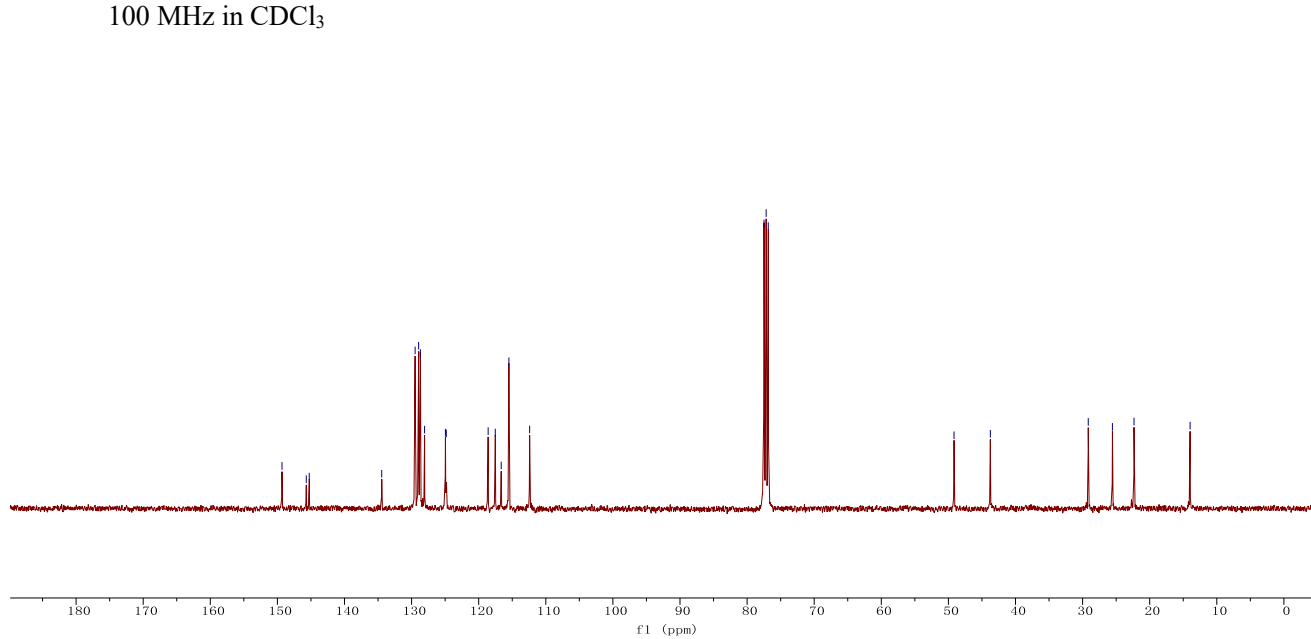
400 MHz in CDCl<sub>3</sub>

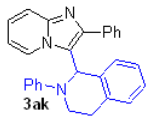


149.31  
145.69  
145.25  
134.45  
129.47  
128.95  
128.69  
128.06  
124.96  
124.83  
118.59  
117.53  
116.65  
115.50  
112.41  
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77.16  
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49.16  
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29.16  
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22.34  
13.98

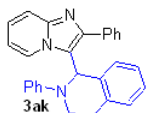
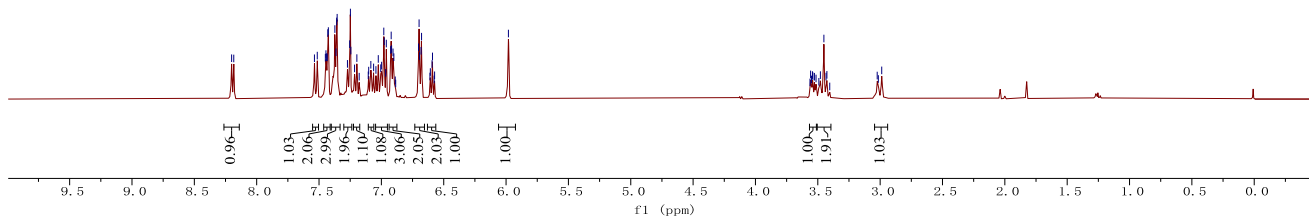


100 MHz in CDCl<sub>3</sub>

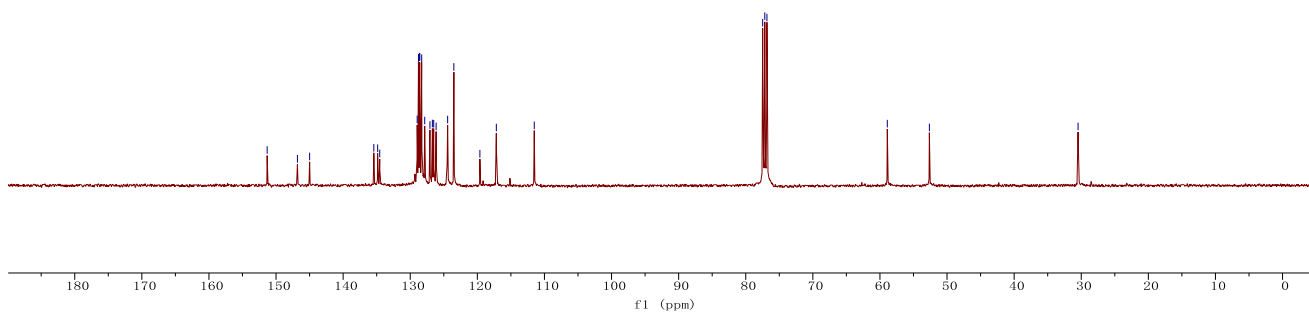




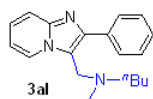
400 MHz in CDCl<sub>3</sub>



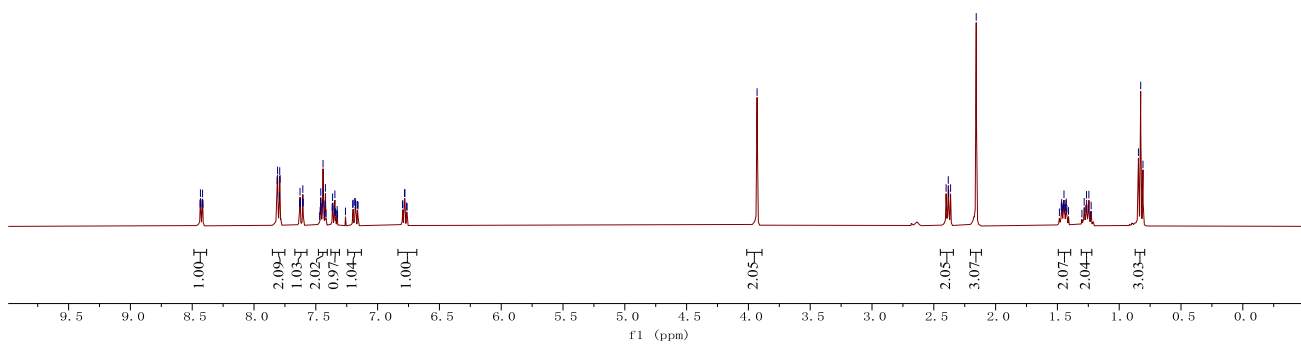
100 MHz in CDCl<sub>3</sub>



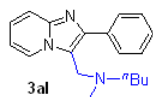
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7.8094  
7.7948  
7.7920  
7.7889  
7.6305  
7.6277  
7.6248  
7.6078  
7.6050  
7.6021  
7.4644  
7.4621  
7.4597  
7.4555  
7.4458  
7.4416  
7.4381  
7.4262  
7.4223  
7.3677  
7.3644  
7.3610  
7.3510  
7.3458  
7.3274  
7.2015  
7.1981  
7.1847  
7.1814  
7.1787  
7.1755  
7.1620  
7.1587  
6.7991  
6.7960  
6.7821  
6.7791  
6.7650  
6.7619  
3.9299  
2.4007  
2.3829  
2.3797  
2.3644  
2.1569  
2.1569  
1.4689  
1.4661  
1.4634  
1.4600  
1.4519  
1.4469  
1.4403  
1.4330  
1.4311  
1.4282  
1.4234  
1.2834  
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0.8262  
0.8078



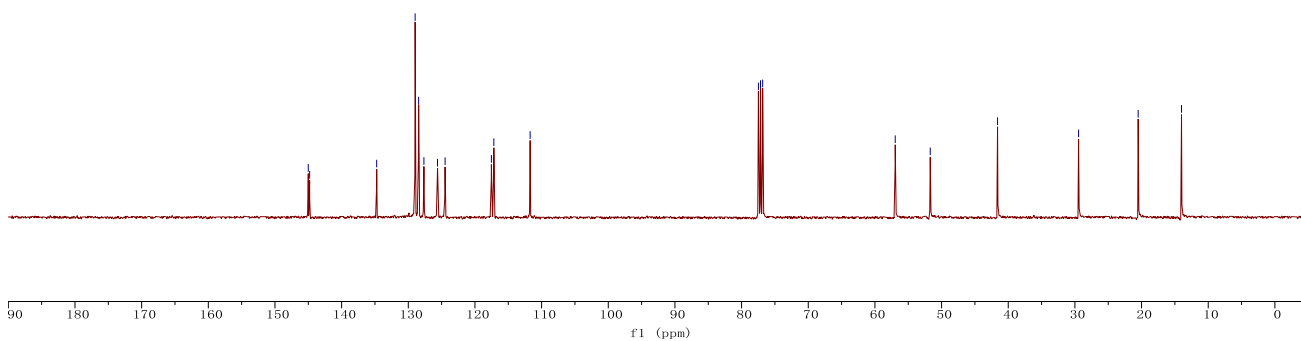
400 MHz in CDCl<sub>3</sub>

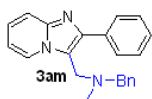


145.00  
144.80  
134.74  
128.97  
128.45  
127.66  
125.61  
124.48  
117.53  
117.17  
111.72  
77.48  
77.16  
76.84  
56.95  
51.70  
41.60  
29.45  
20.51  
14.01

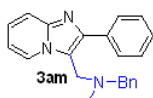
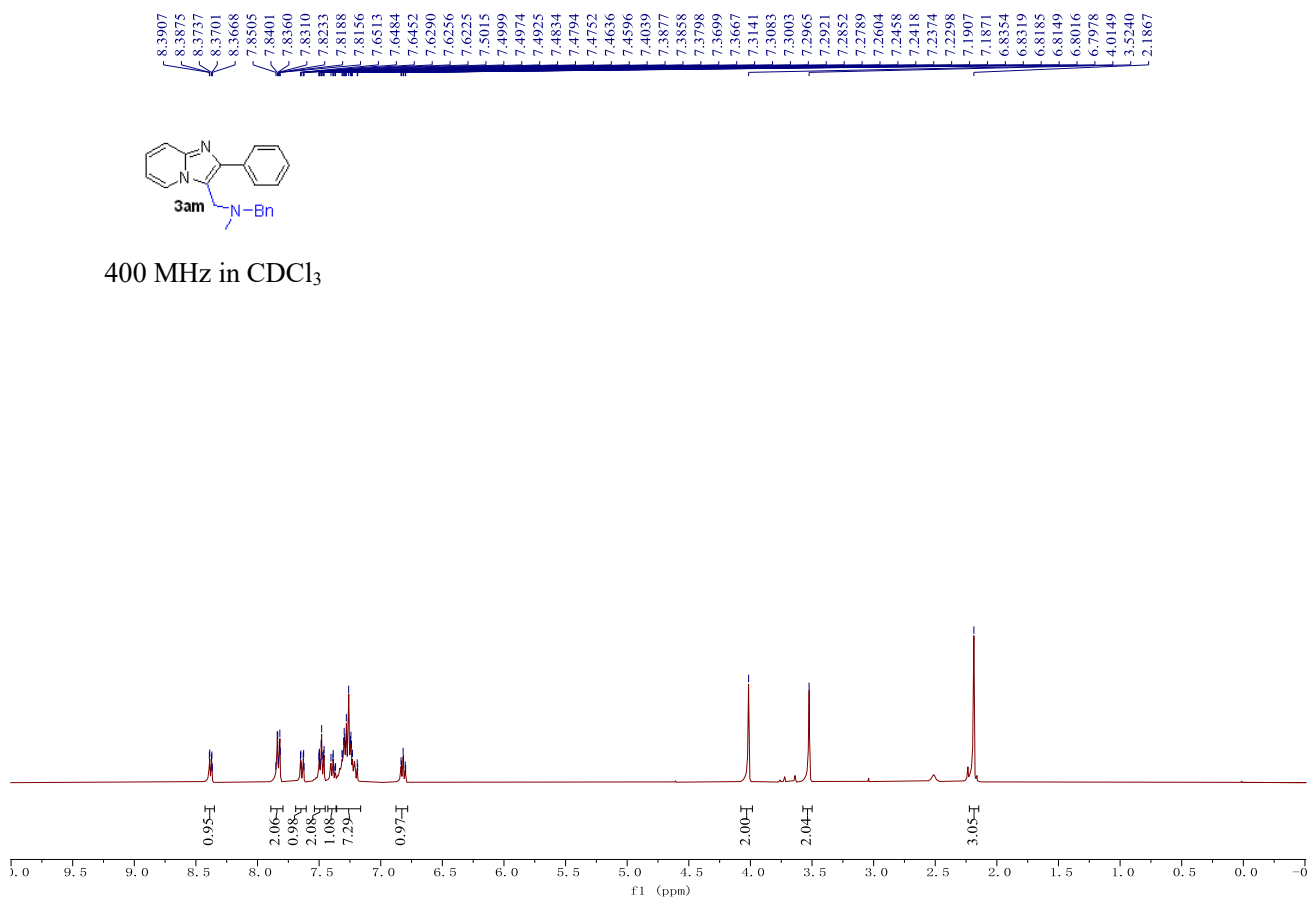


100 MHz in CDCl<sub>3</sub>

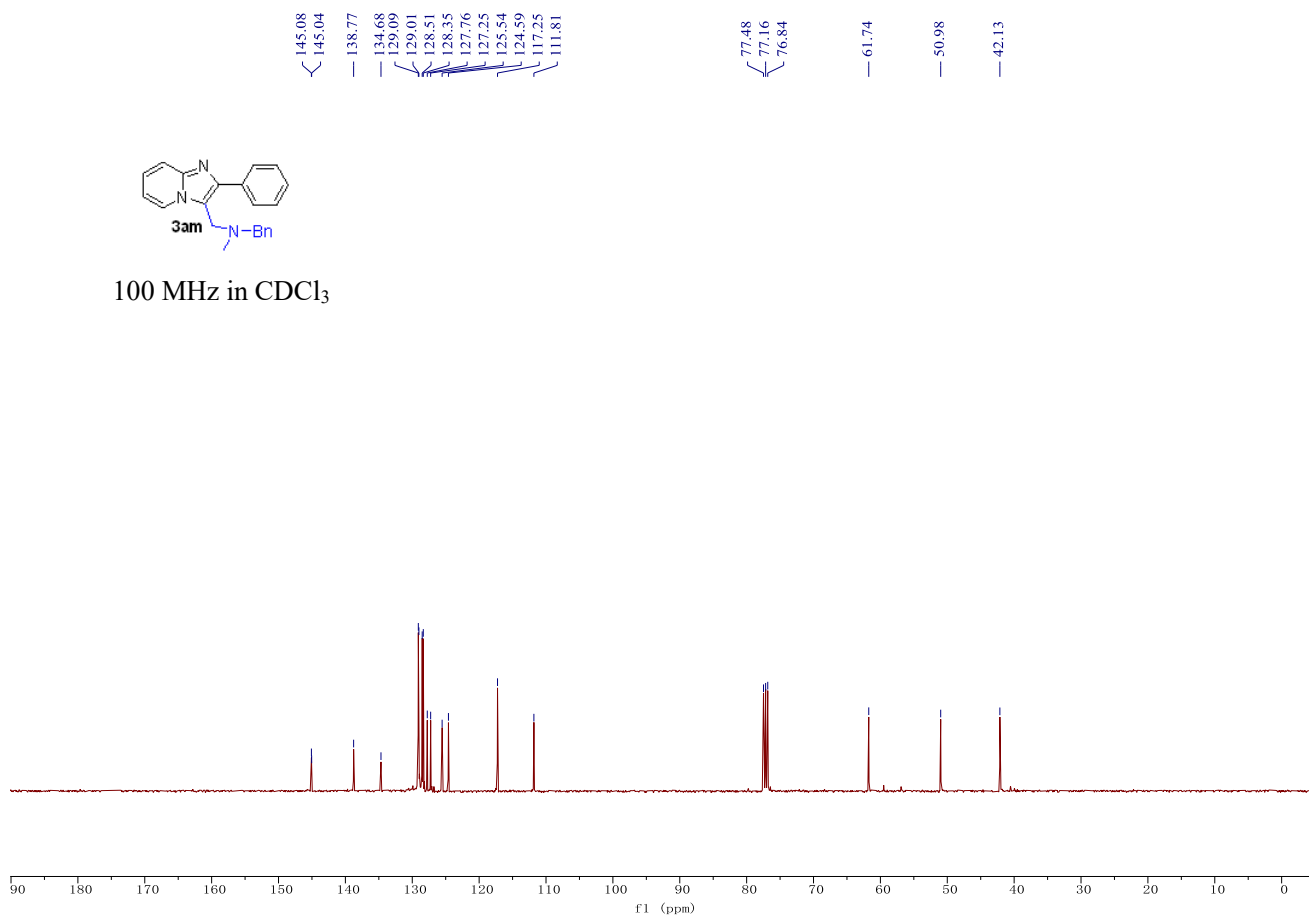




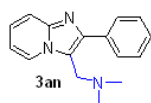
400 MHz in CDCl<sub>3</sub>



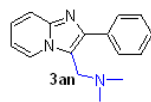
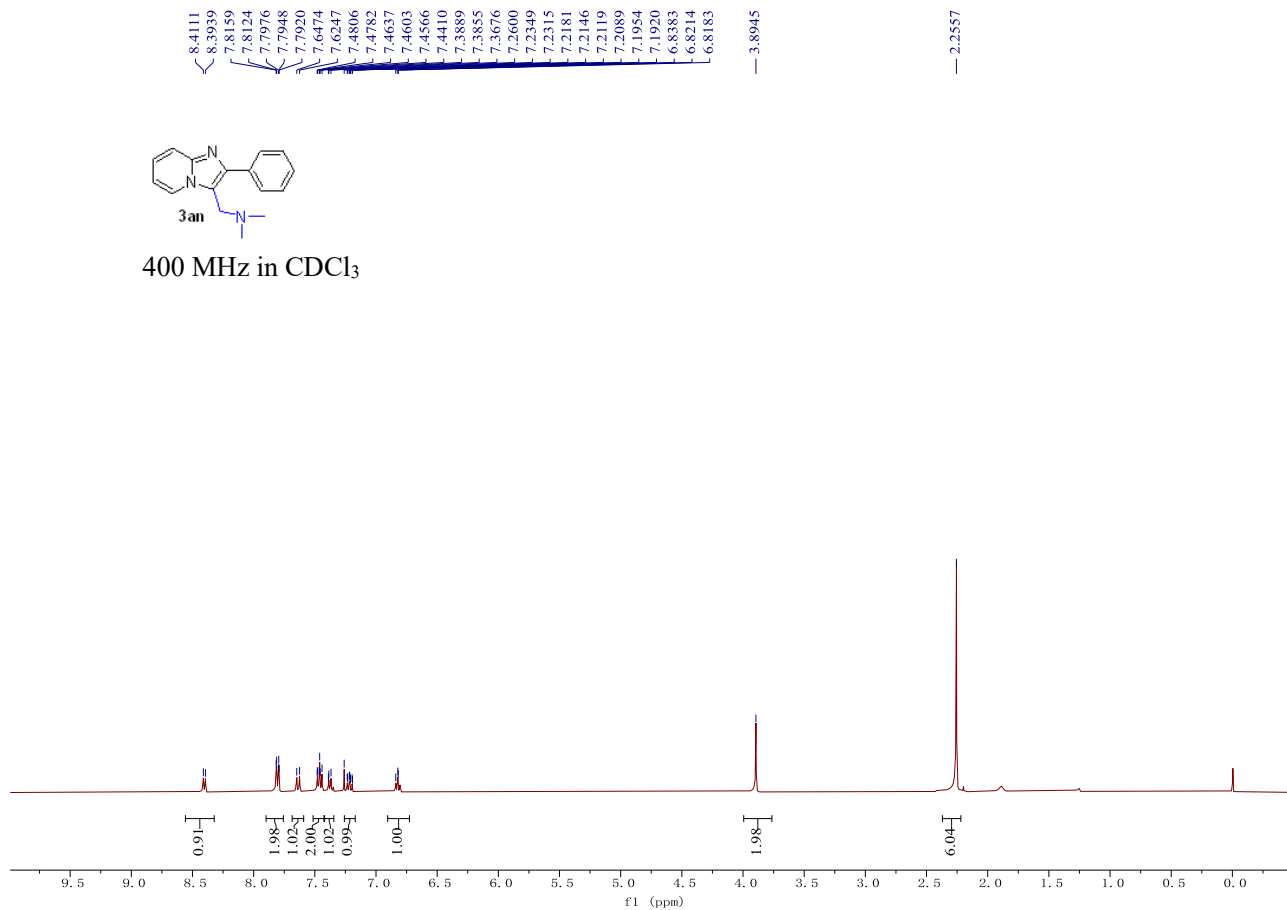
100 MHz in CDCl<sub>3</sub>



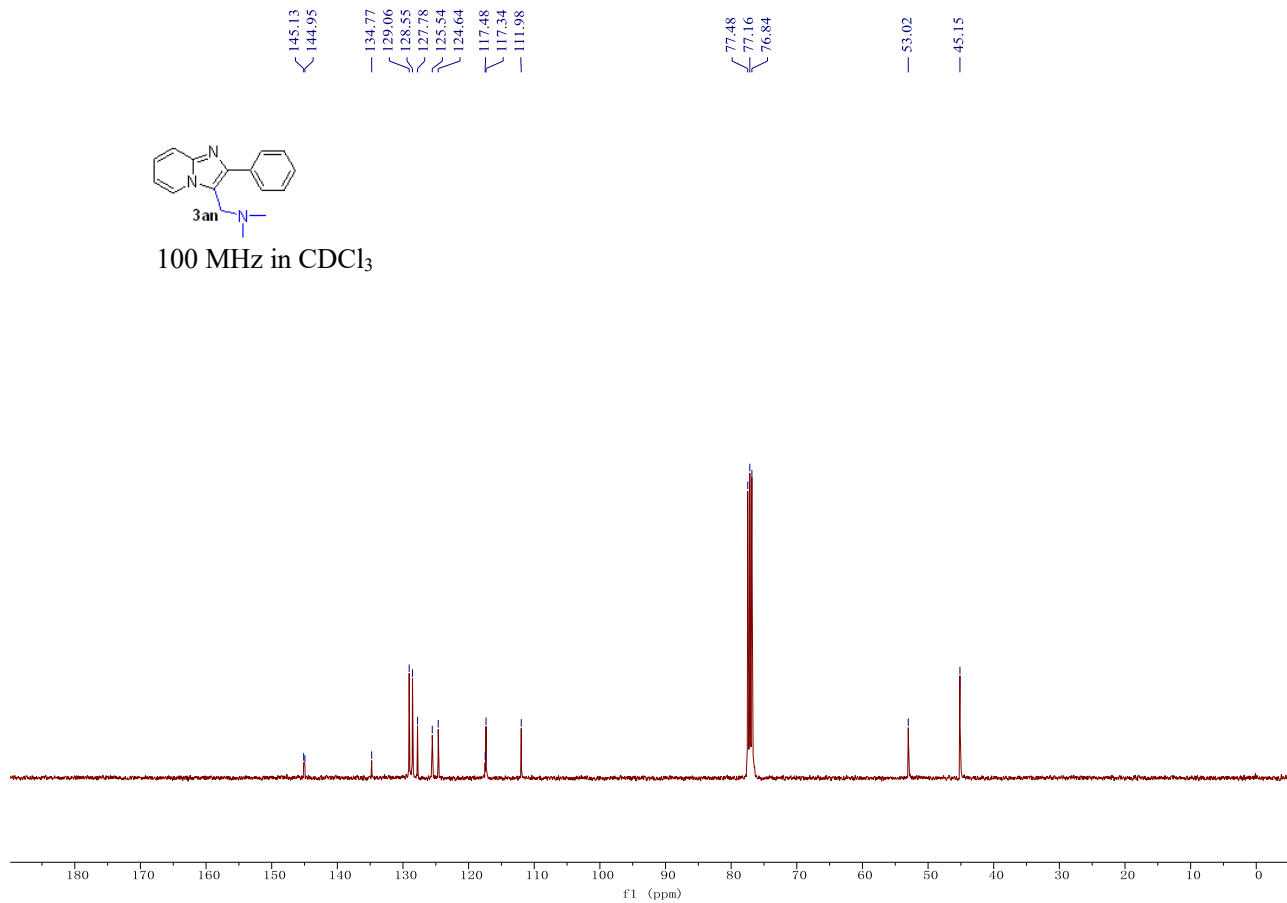


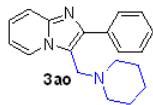


400 MHz in CDCl<sub>3</sub>

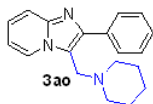
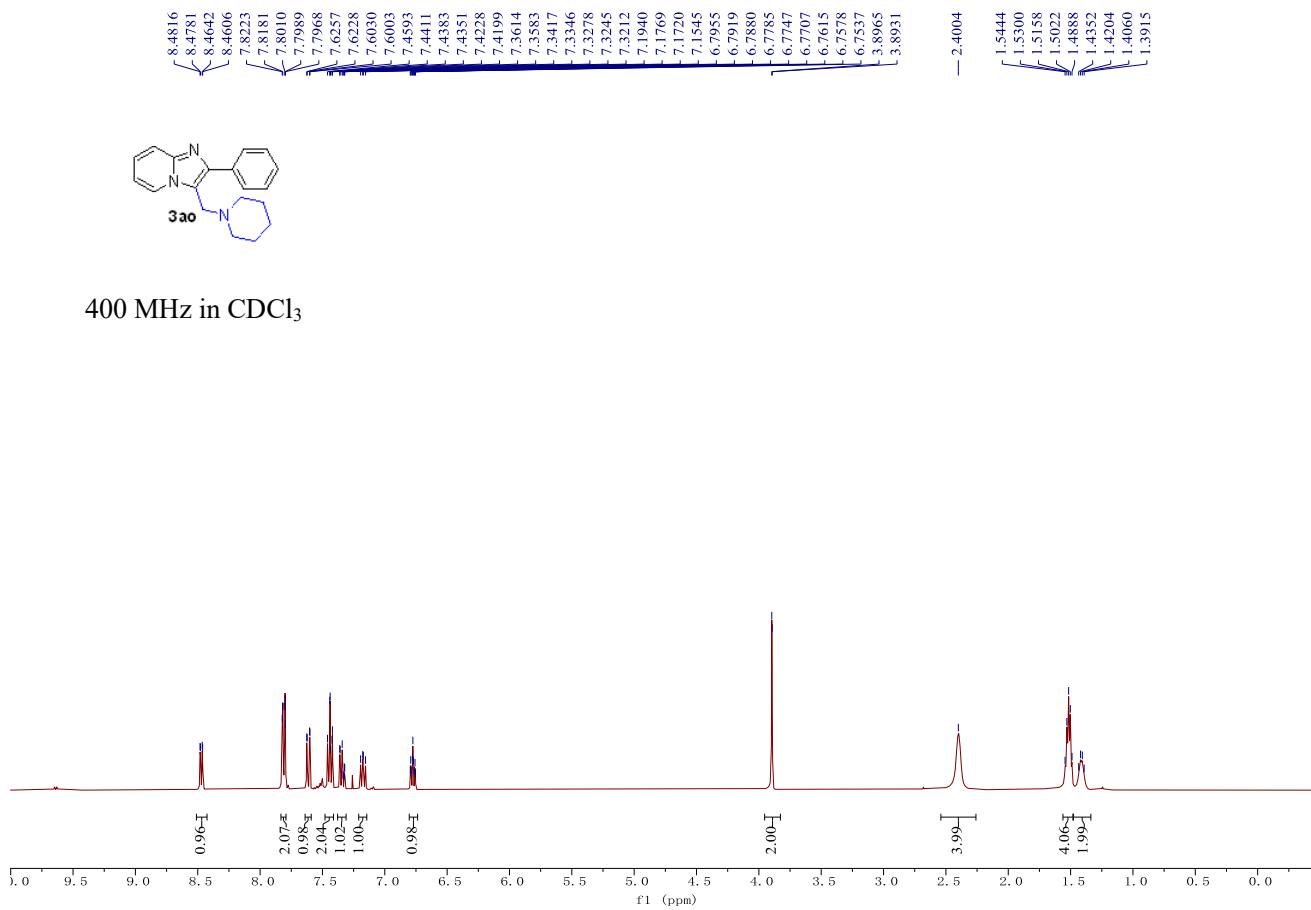


100 MHz in CDCl<sub>3</sub>

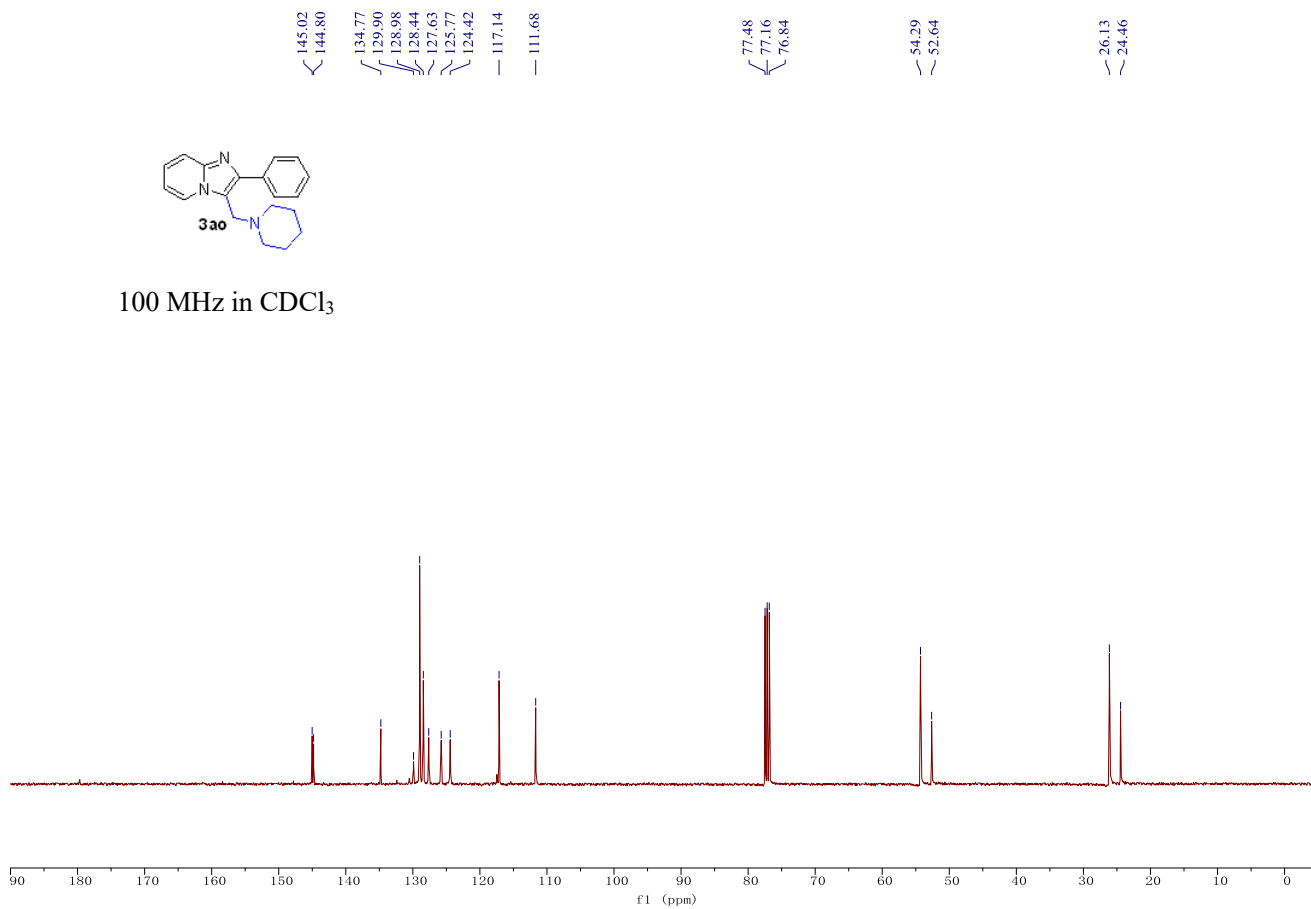




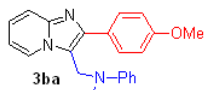
400 MHz in CDCl<sub>3</sub>



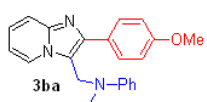
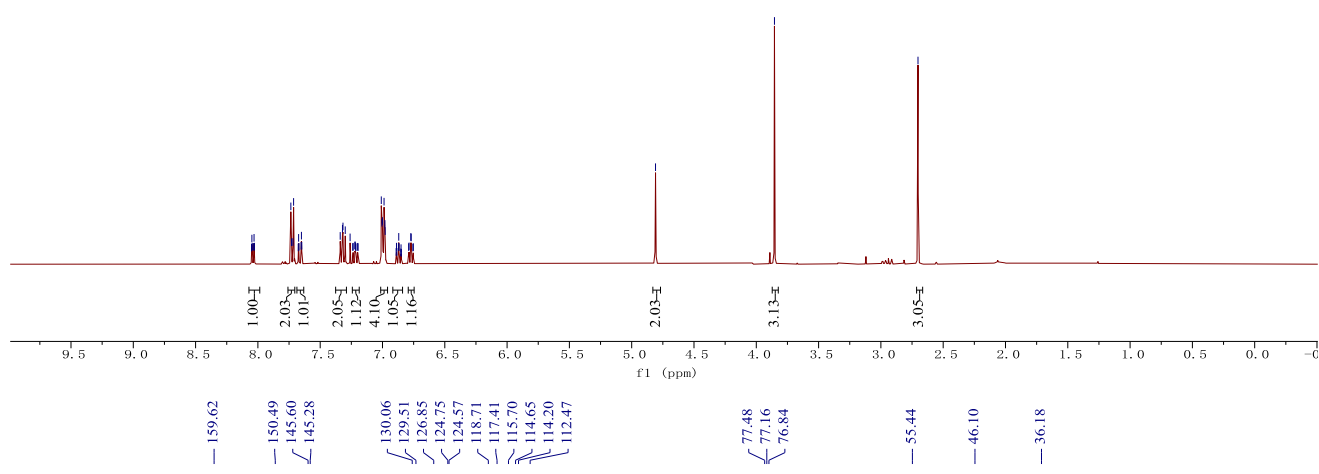
100 MHz in CDCl<sub>3</sub>



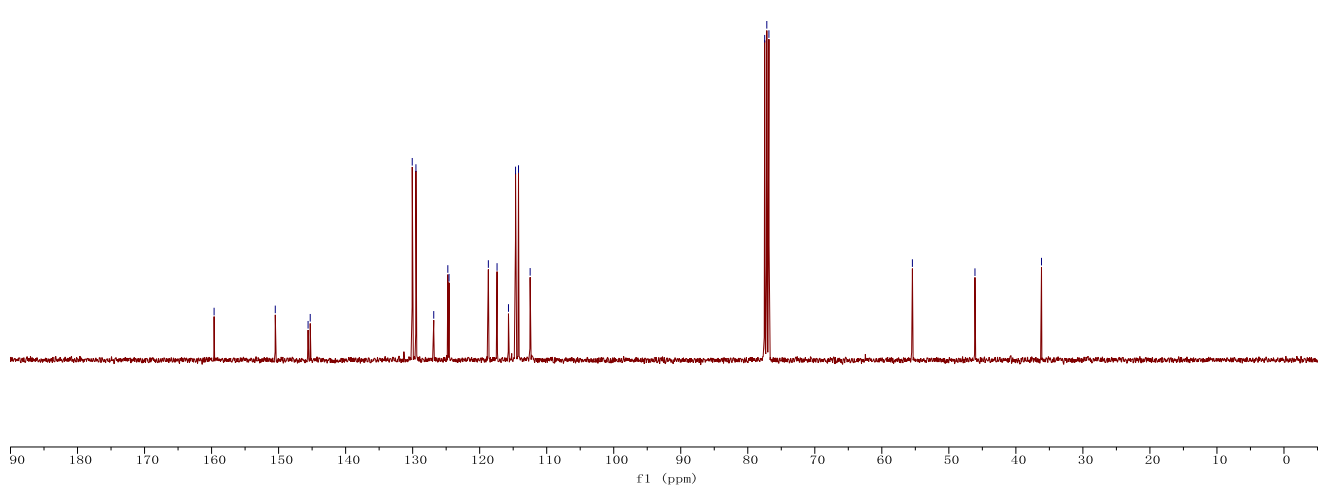
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7.7134  
7.6761  
7.6733  
7.6704  
7.6533  
7.6506  
7.6478  
7.3394  
7.3212  
7.3172  
7.2990  
7.2960  
7.2388  
7.2357  
7.2220  
7.2188  
7.2162  
7.2130  
7.1994  
7.1962  
7.0097  
7.0037  
7.0005  
6.9875  
6.9808  
6.9784  
6.8903  
6.8877  
6.8850  
6.8721  
6.8695  
6.8669  
6.8538  
6.8512  
6.8485  
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6.7869  
6.7729  
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3.8542  
2.7022



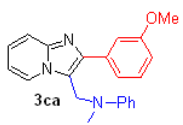
400 MHz in CDCl<sub>3</sub>



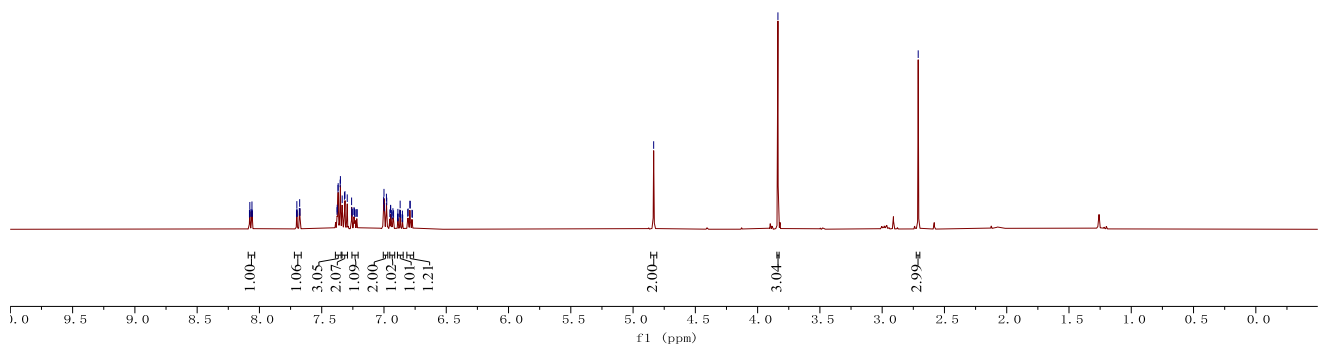
100 MHz in CDCl<sub>3</sub>



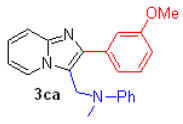
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7.6769  
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7.3754  
7.3692  
7.3678  
7.3518  
7.3498  
7.3355  
7.3172  
7.3133  
7.2950  
7.2600  
7.2580  
7.2550  
7.2412  
7.2380  
7.2354  
7.2322  
7.2185  
7.2154  
7.0026  
7.0000  
6.9975  
6.9803  
6.9778  
6.9528  
6.9468  
6.9467  
6.9410  
6.9353  
6.9282  
6.9230  
6.8912  
6.8884  
6.8859  
6.8729  
6.8701  
6.8675  
6.8546  
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6.8497  
6.8092  
6.8062  
6.7921  
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3.8370  
2.7106



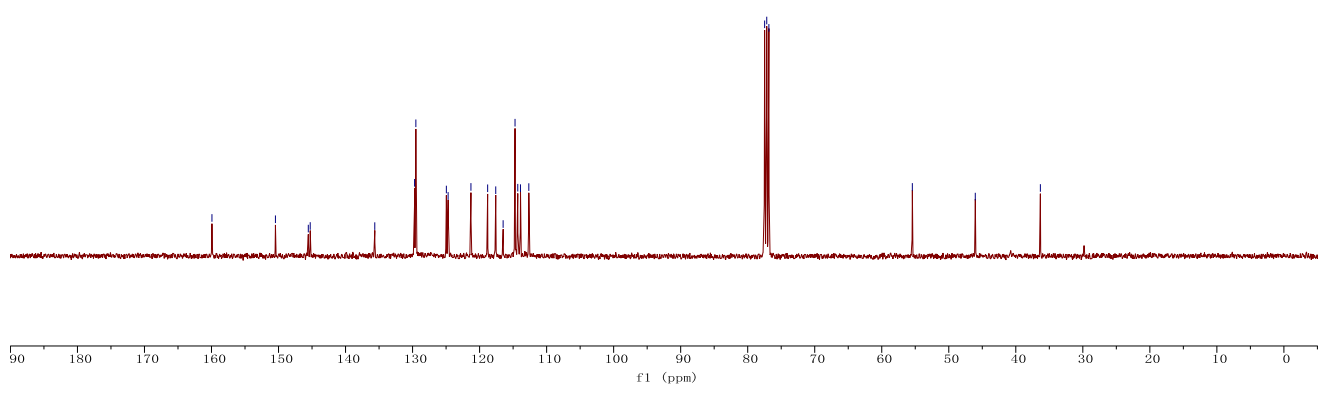
400 MHz in CDCl<sub>3</sub>

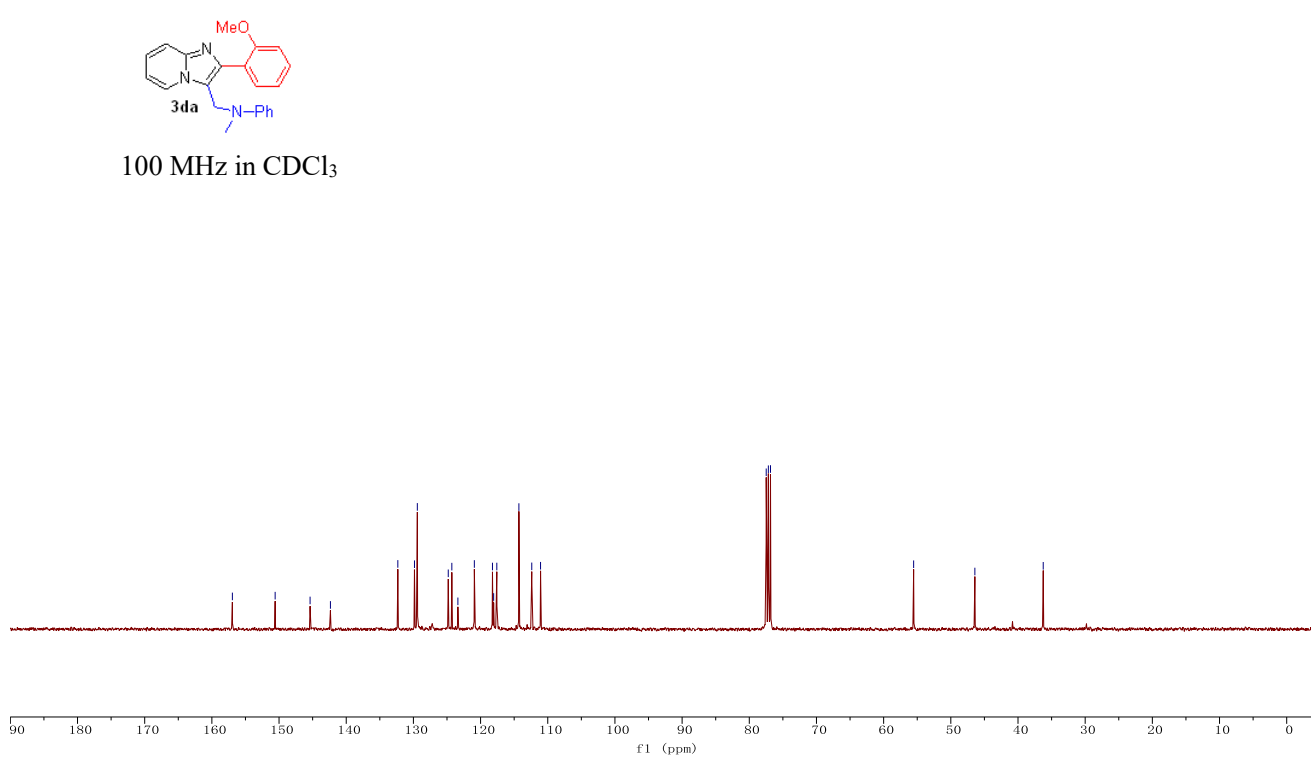
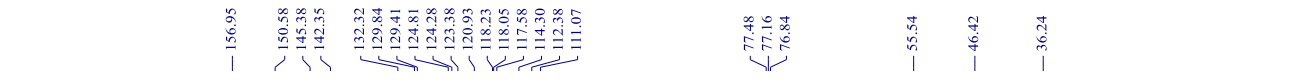
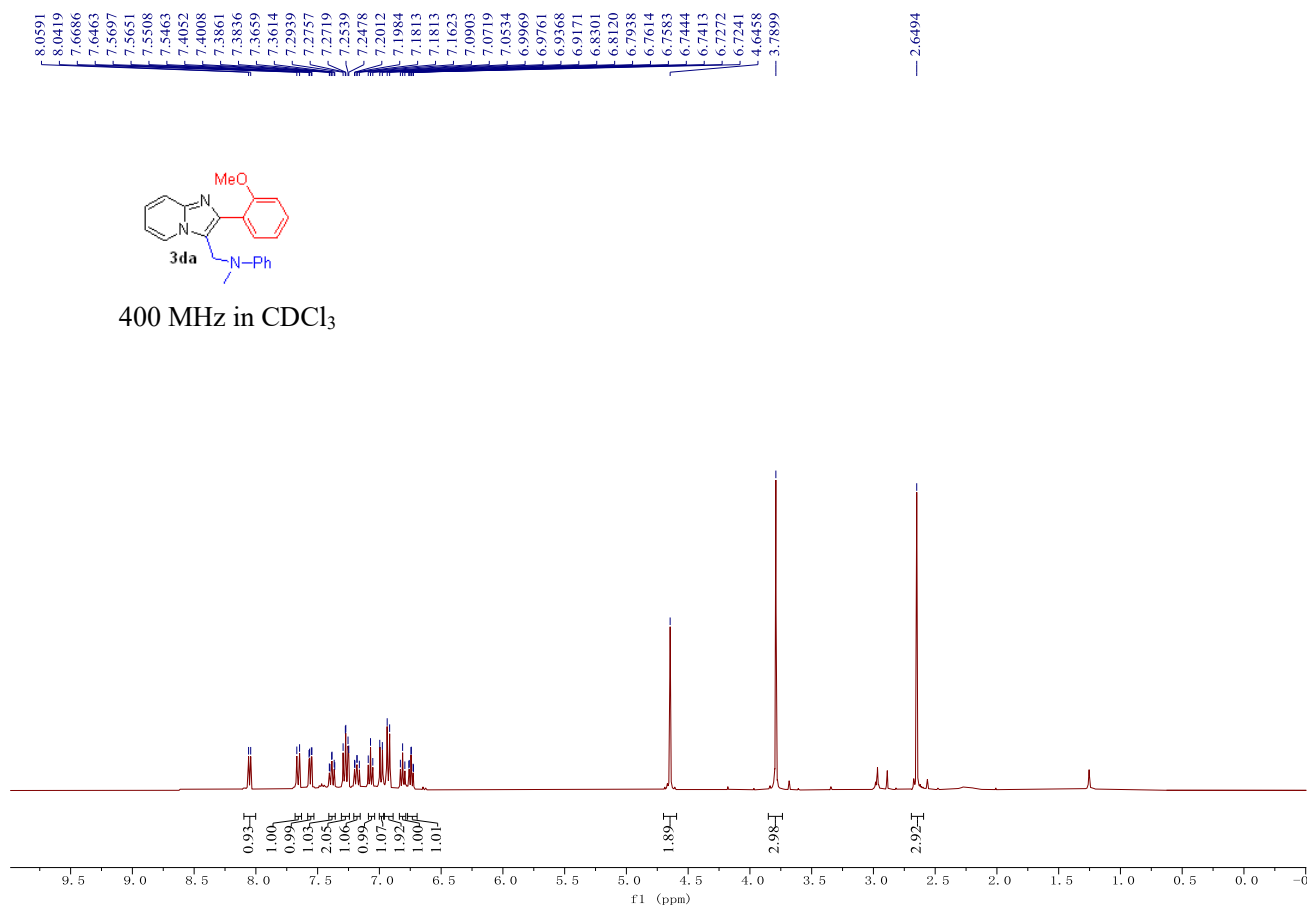


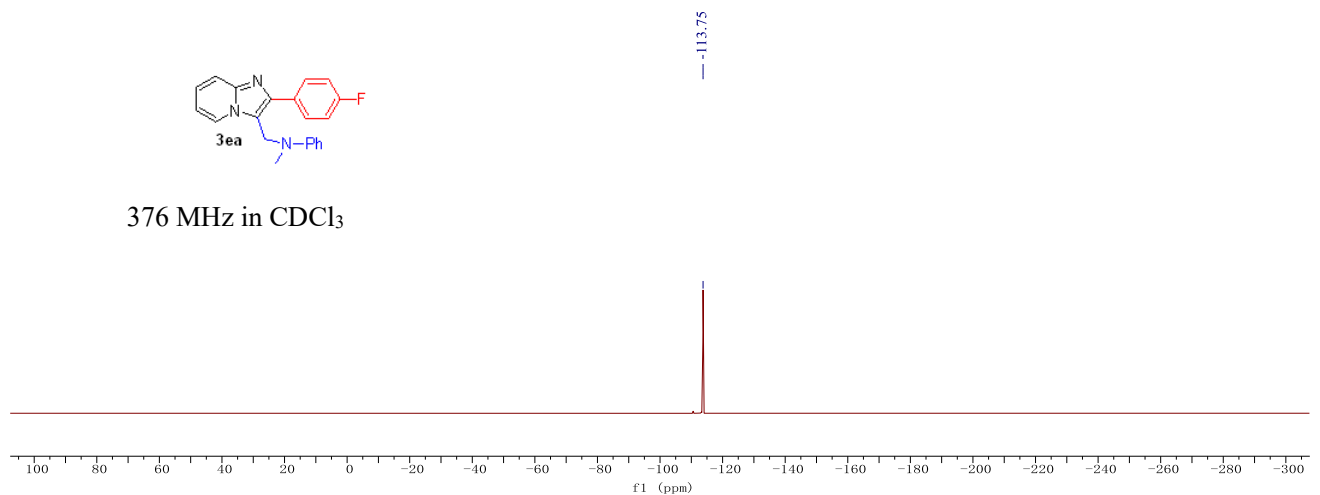
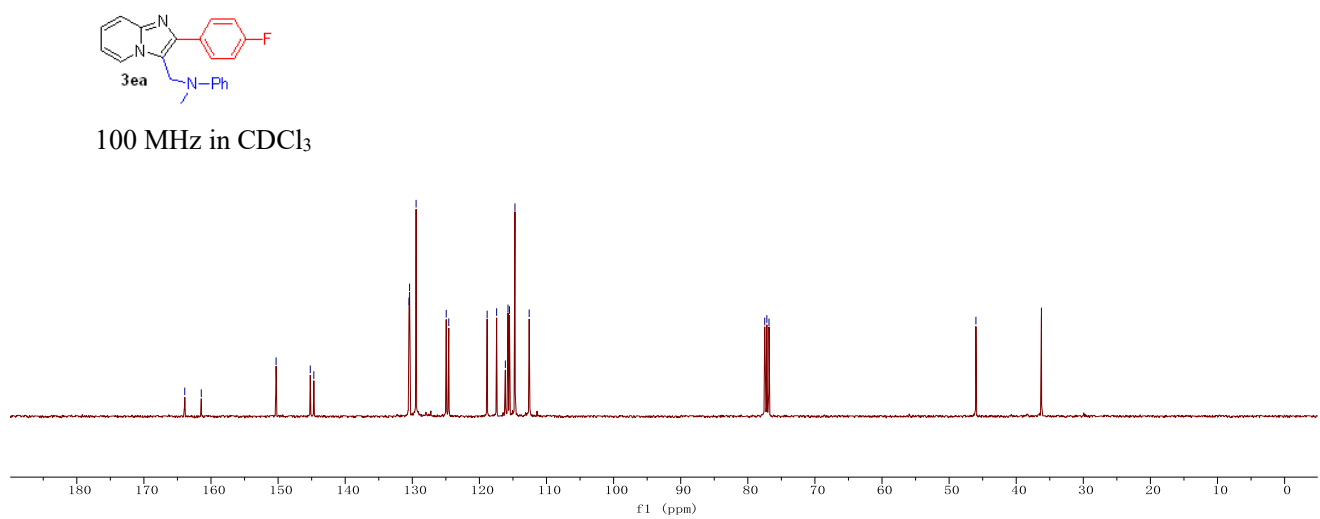
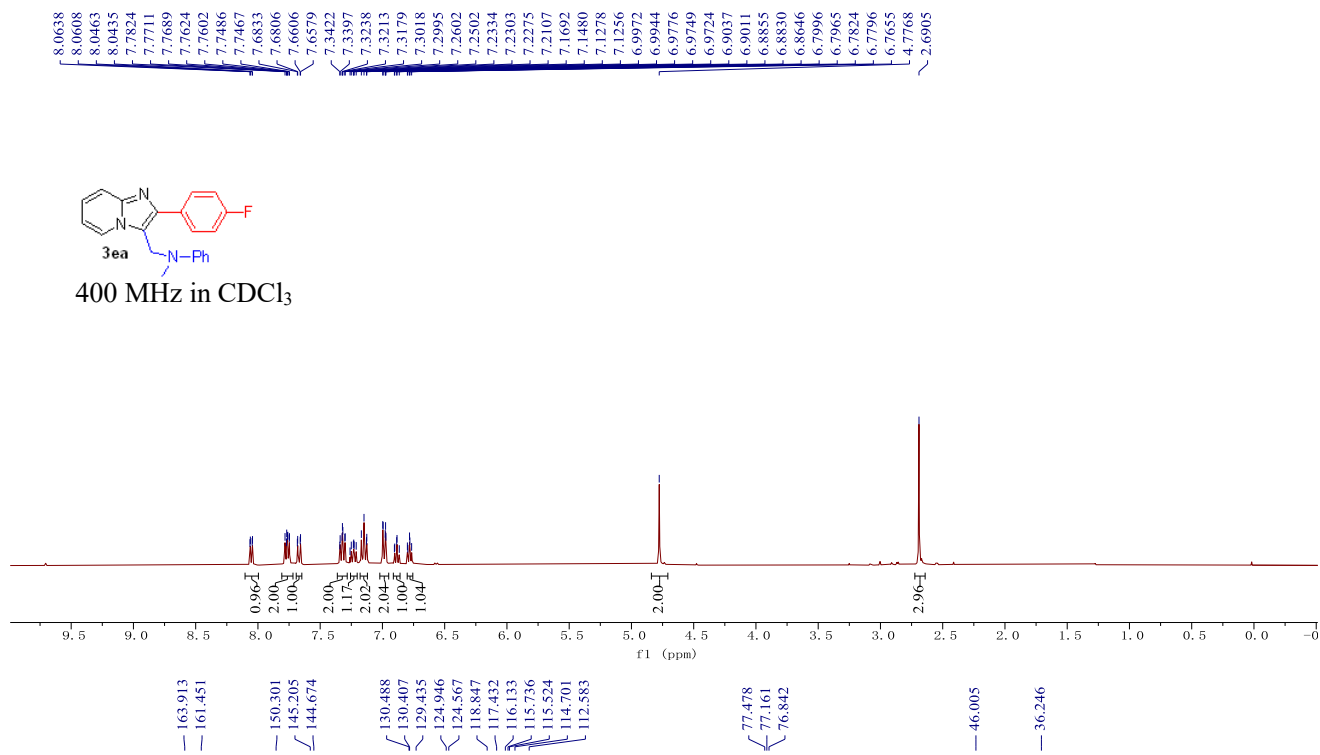
159.93  
150.46  
145.56  
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135.64  
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129.51  
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124.70  
121.29  
118.80  
117.59  
116.49  
114.71  
114.31  
113.90  
112.66  
77.48  
77.16  
76.84  
55.44  
46.05  
36.33

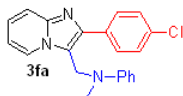


100 MHz in CDCl<sub>3</sub>

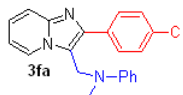
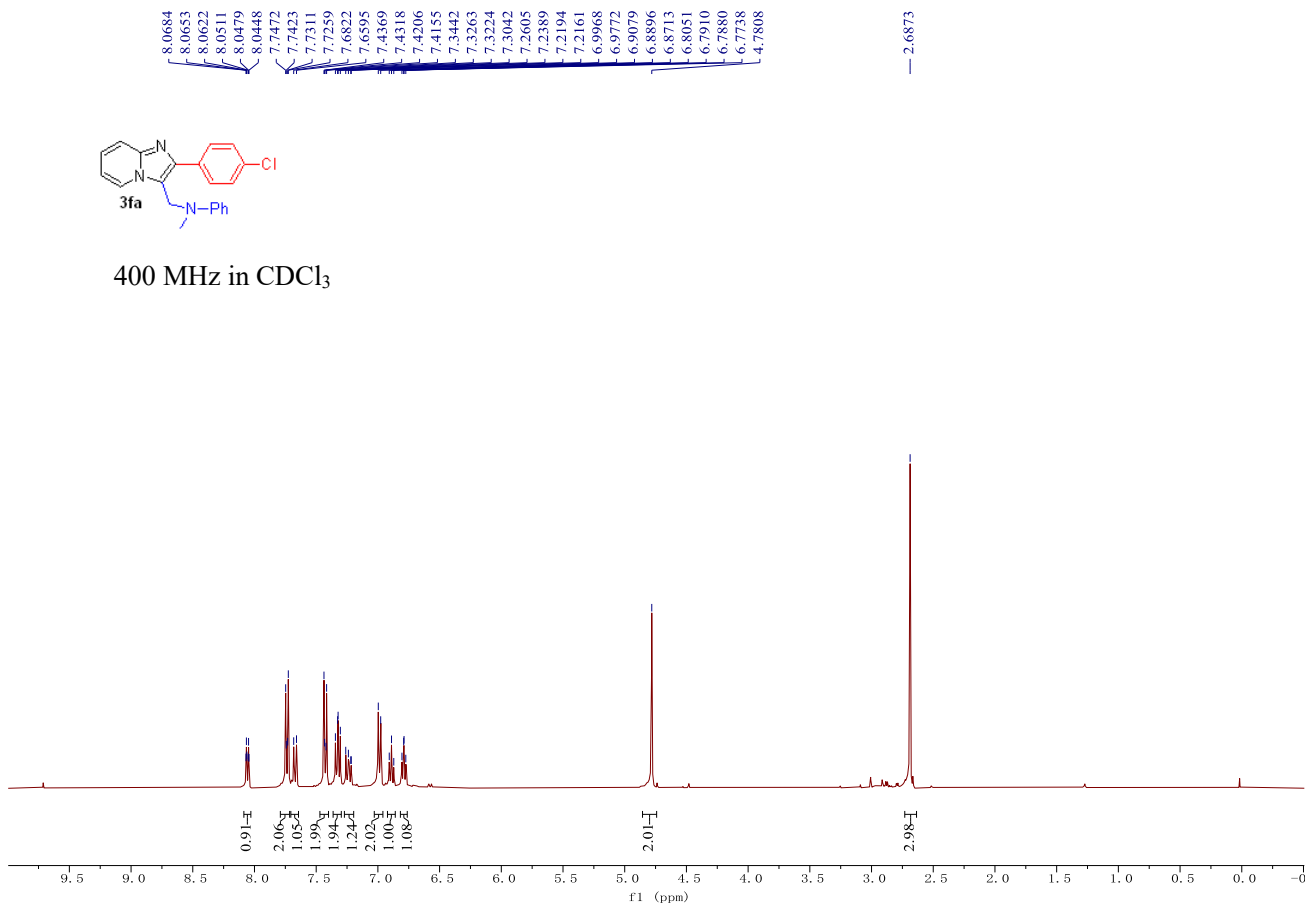




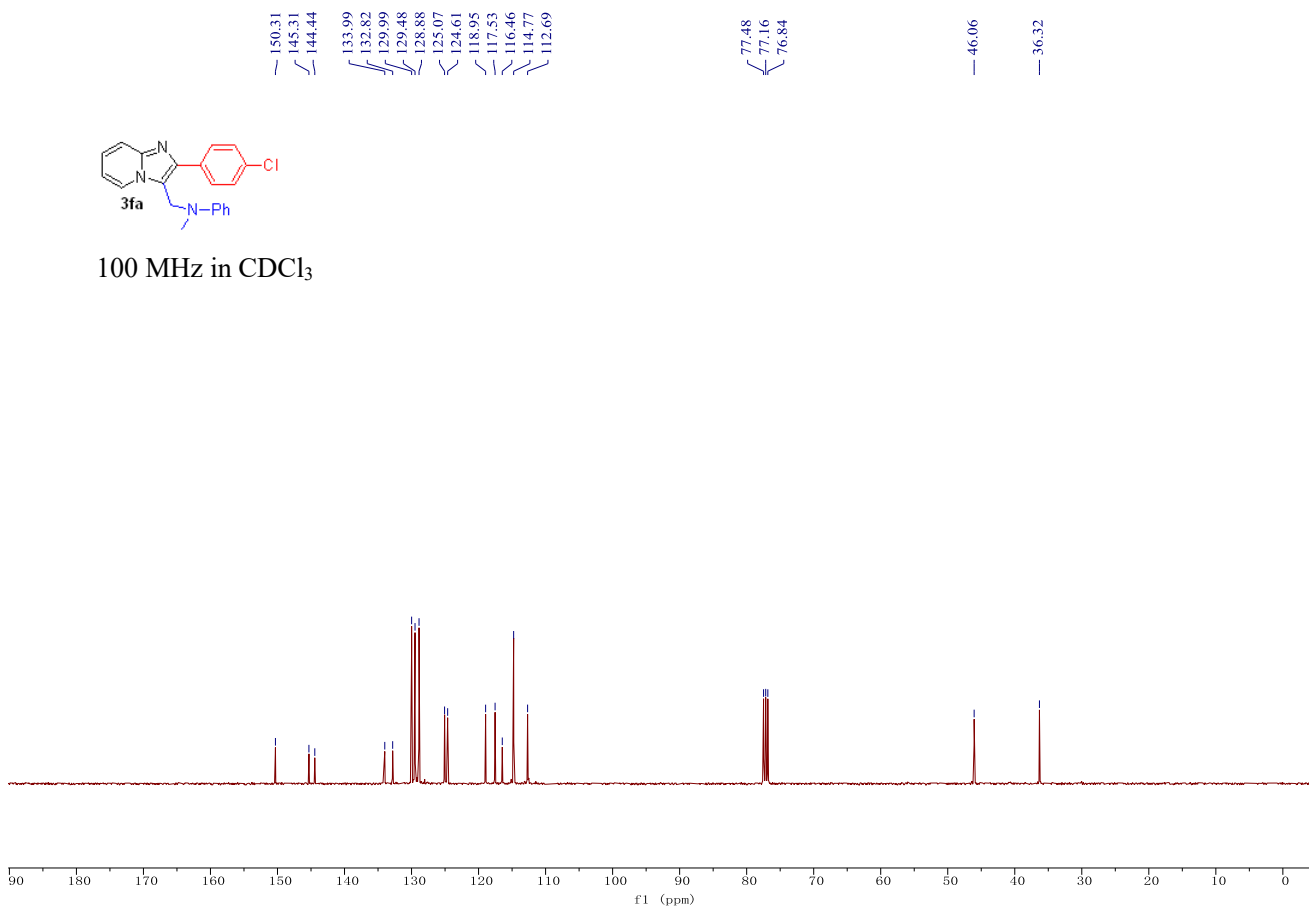


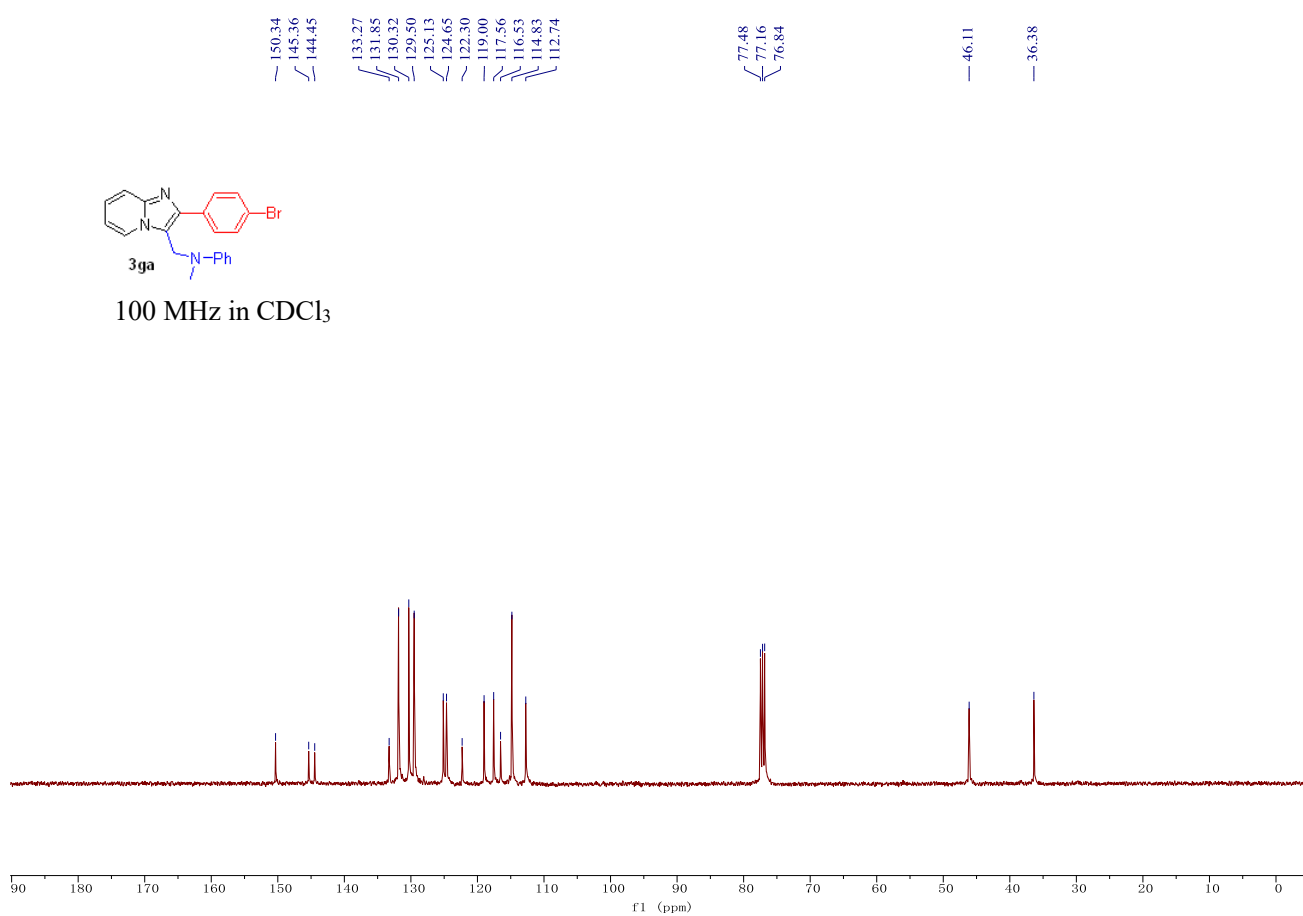
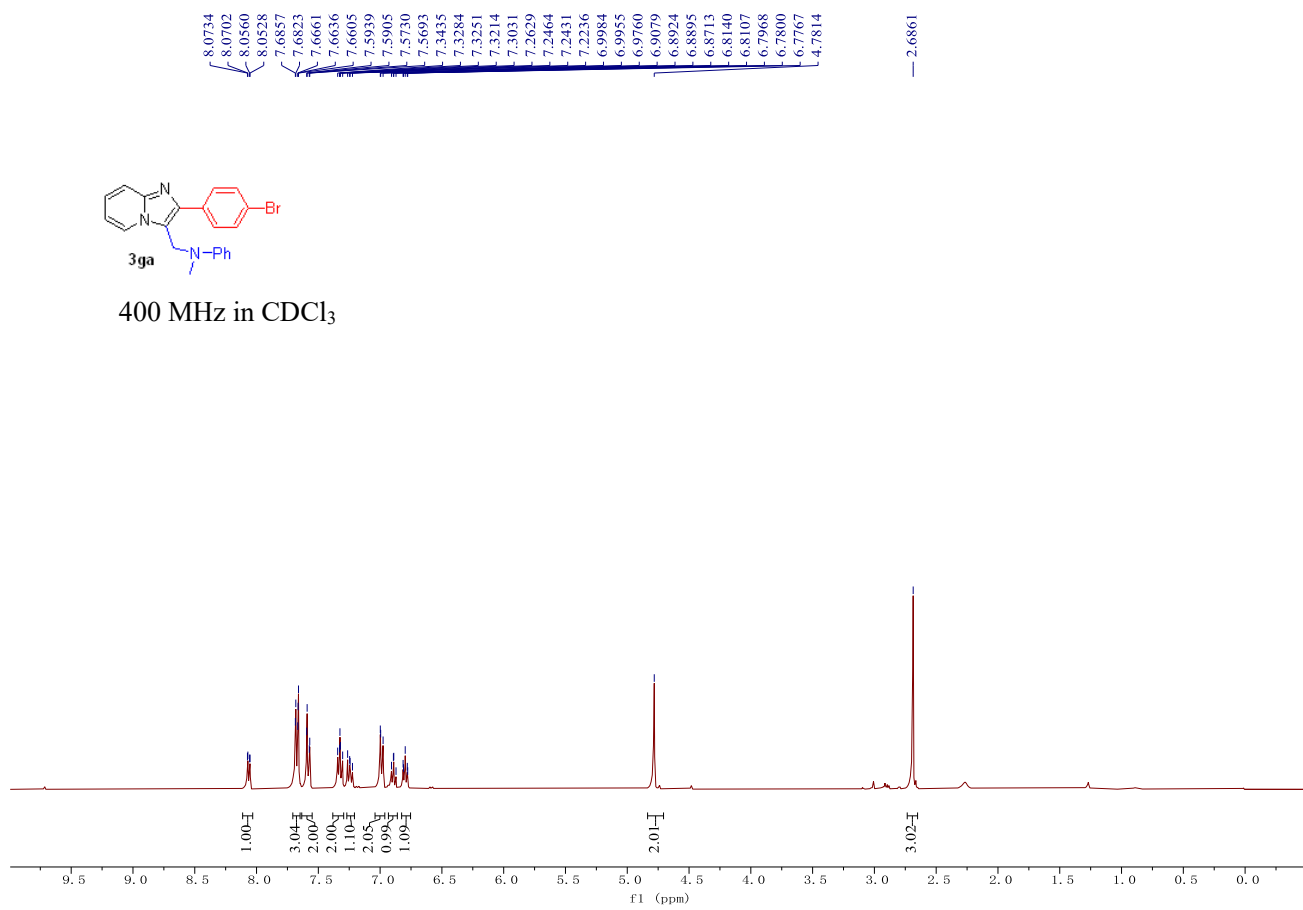


400 MHz in CDCl<sub>3</sub>

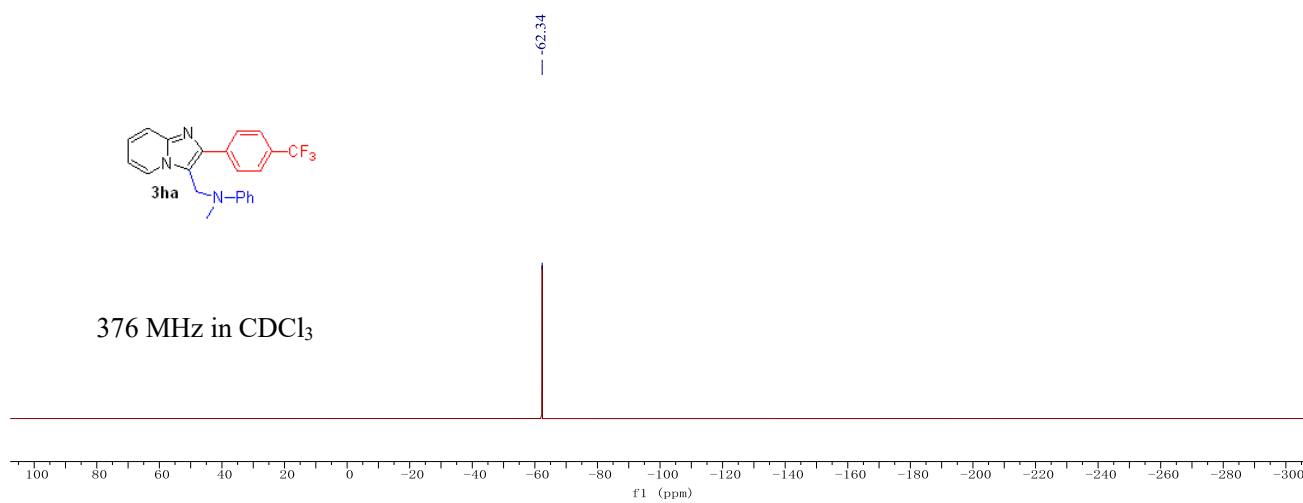
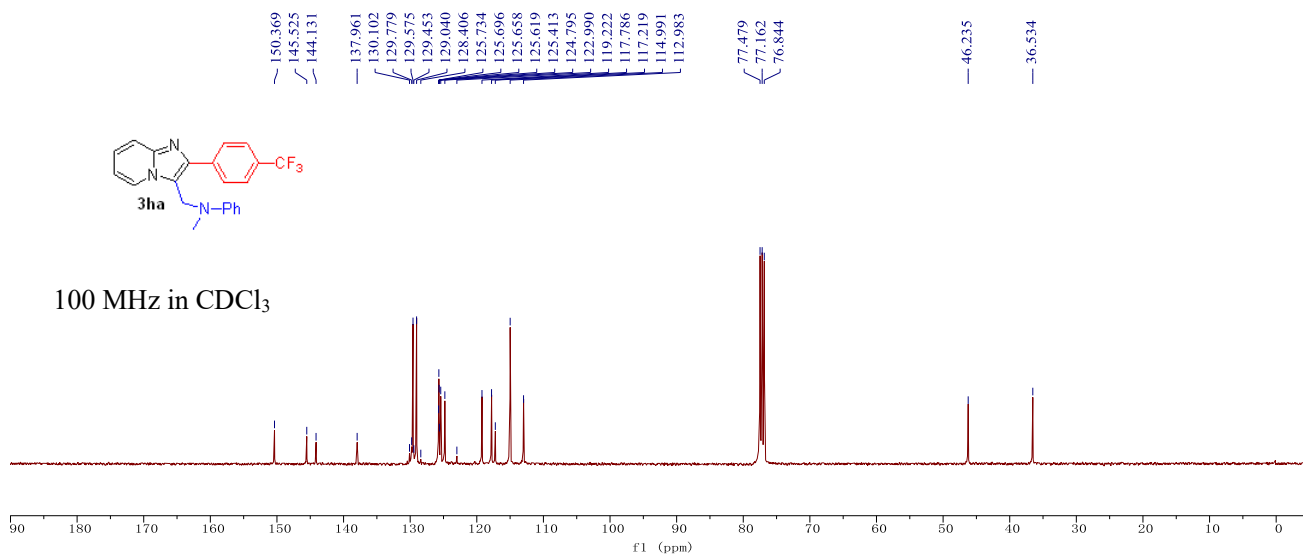
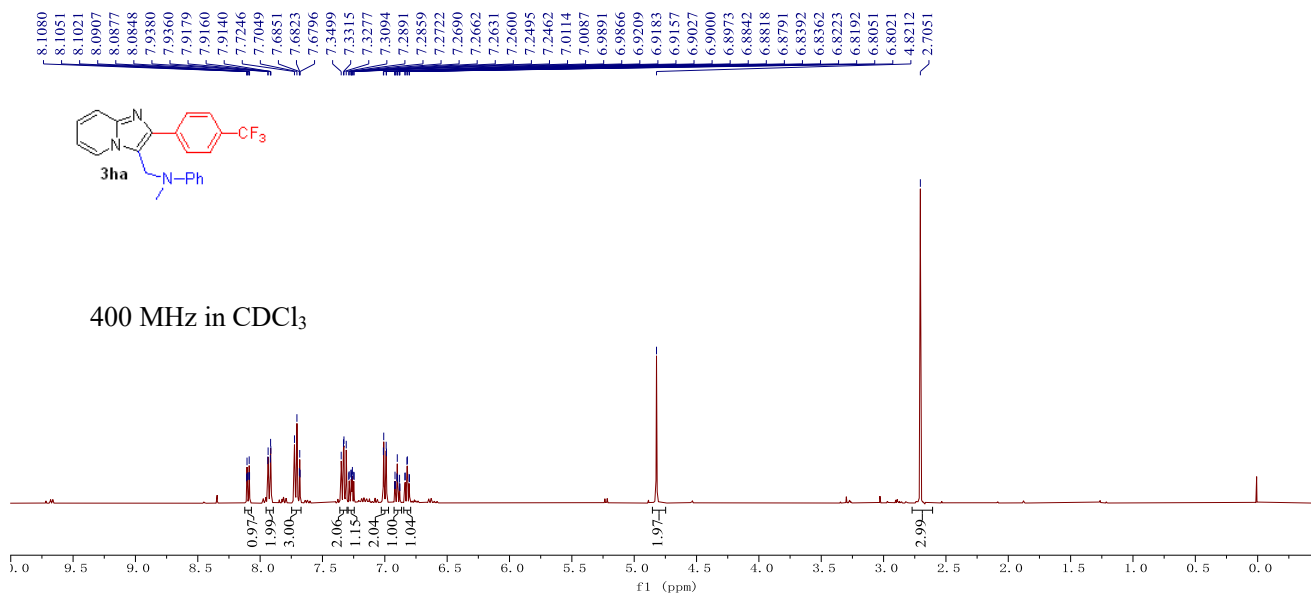


100 MHz in CDCl<sub>3</sub>

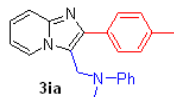




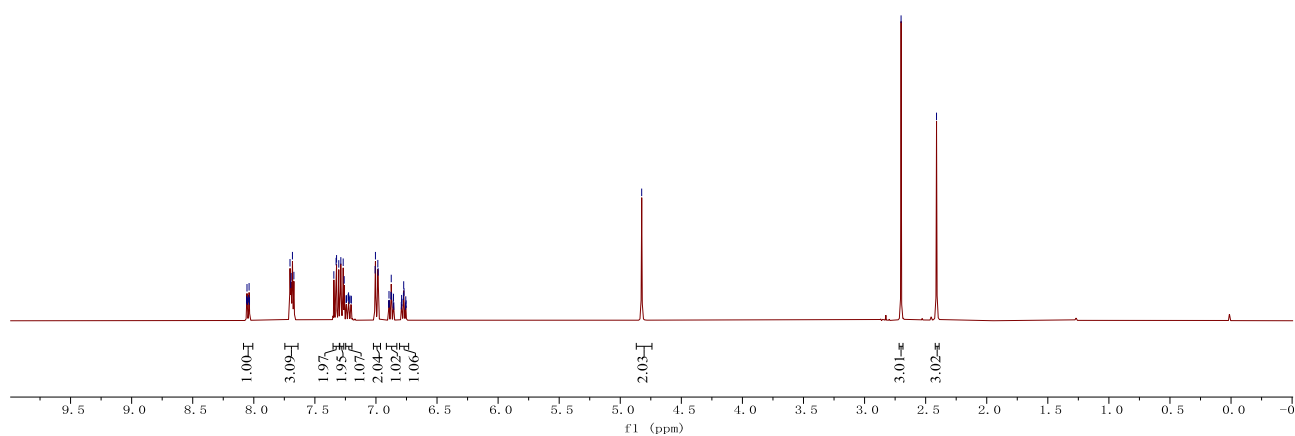




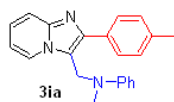
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8.0352  
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7.6976  
7.6944  
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7.6717  
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7.2444  
7.2412  
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7.2215  
7.2184  
7.2049  
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7.0042  
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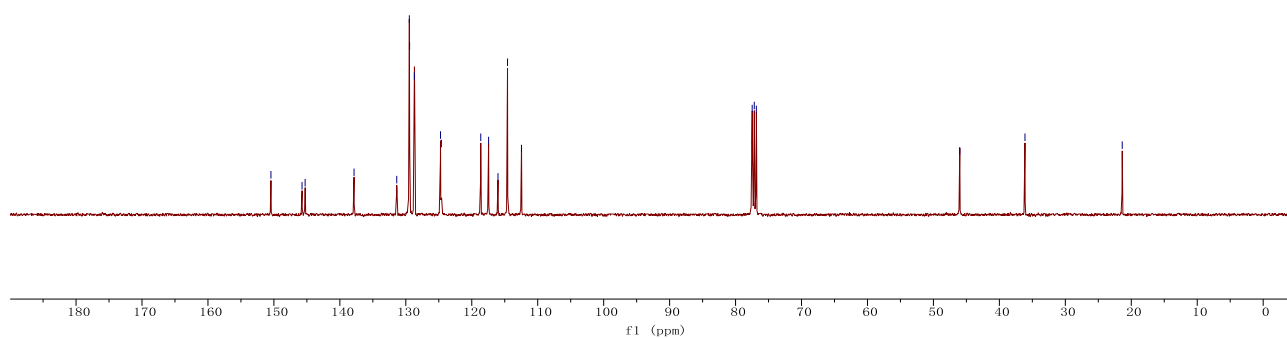
400 MHz in CDCl<sub>3</sub>

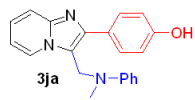


150.45  
145.73  
145.27  
137.85  
131.37  
129.47  
129.43  
128.70  
124.74  
124.61  
118.64  
117.45  
116.02  
114.58  
112.46  
77.48  
77.16  
76.85  
46.01  
36.12  
21.37

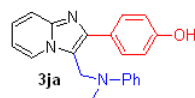
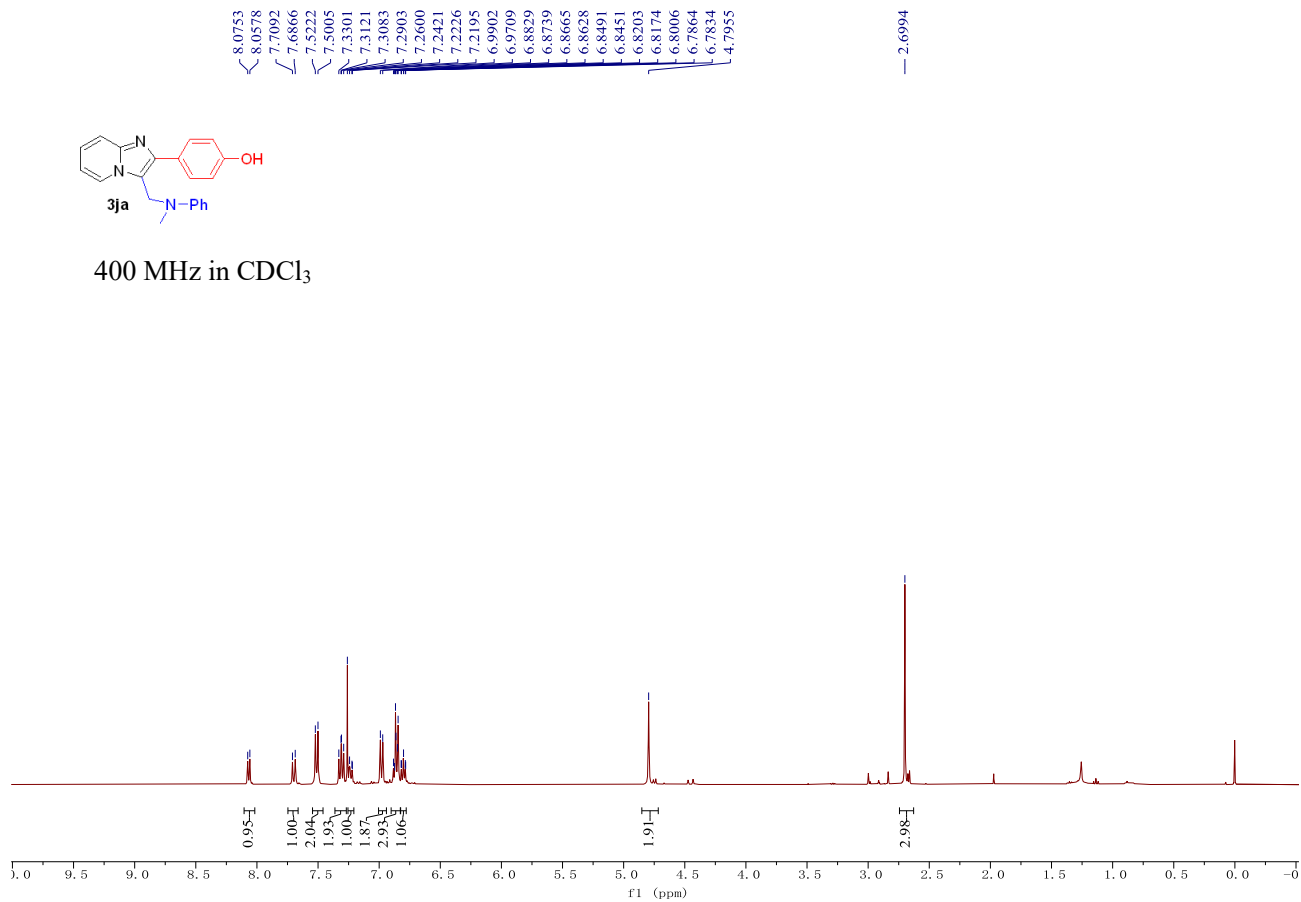


100 MHz in CDCl<sub>3</sub>

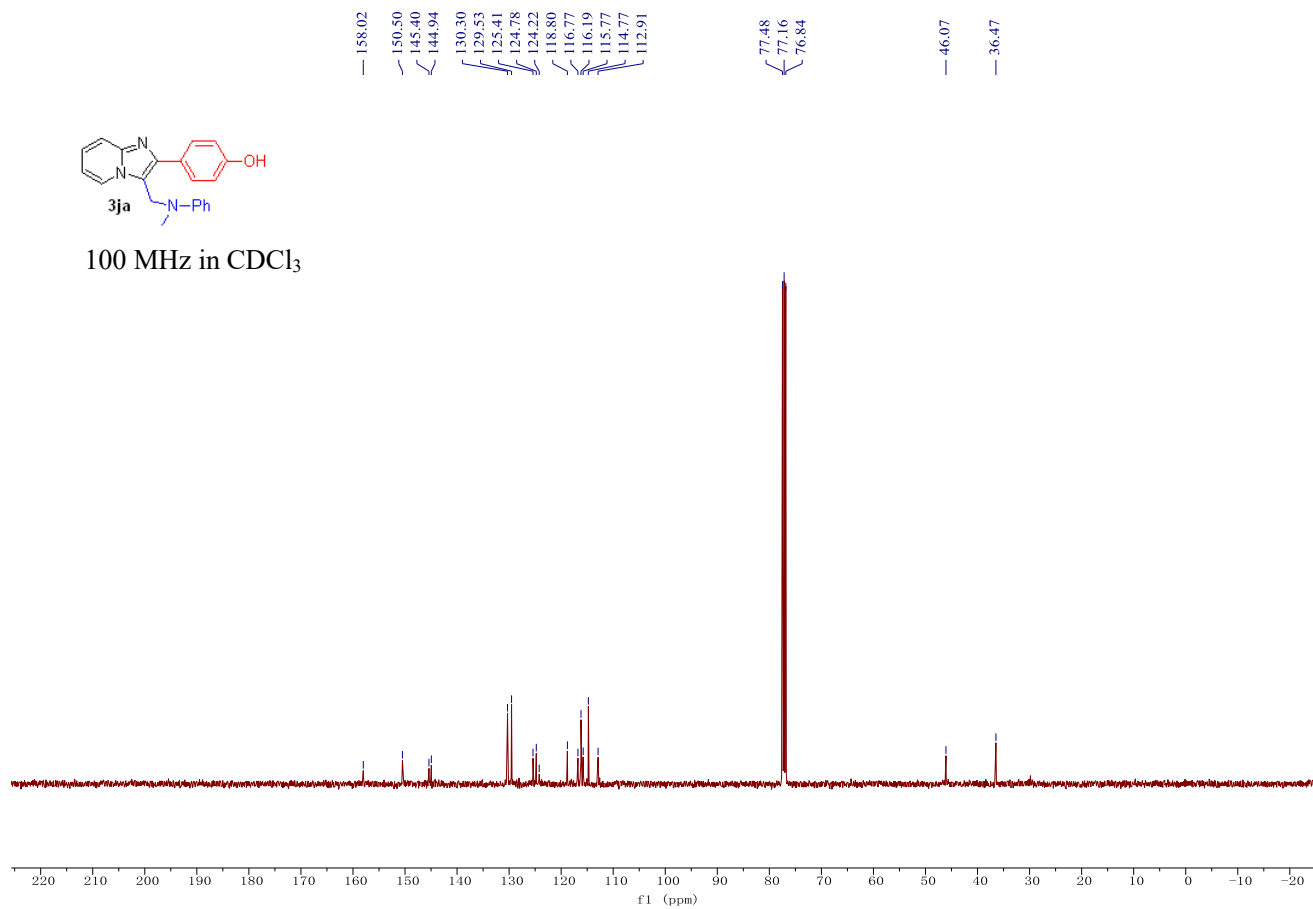




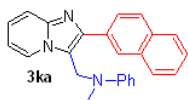
400 MHz in CDCl<sub>3</sub>



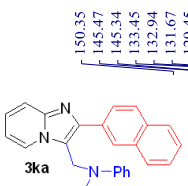
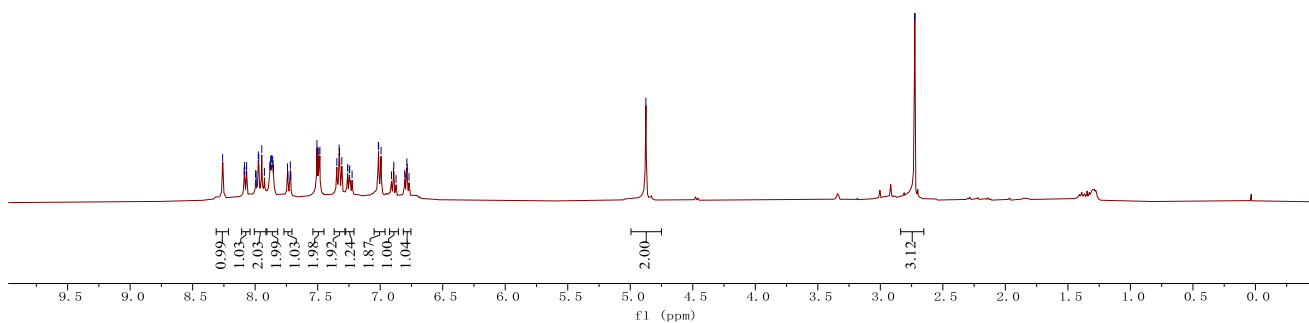
100 MHz in CDCl<sub>3</sub>



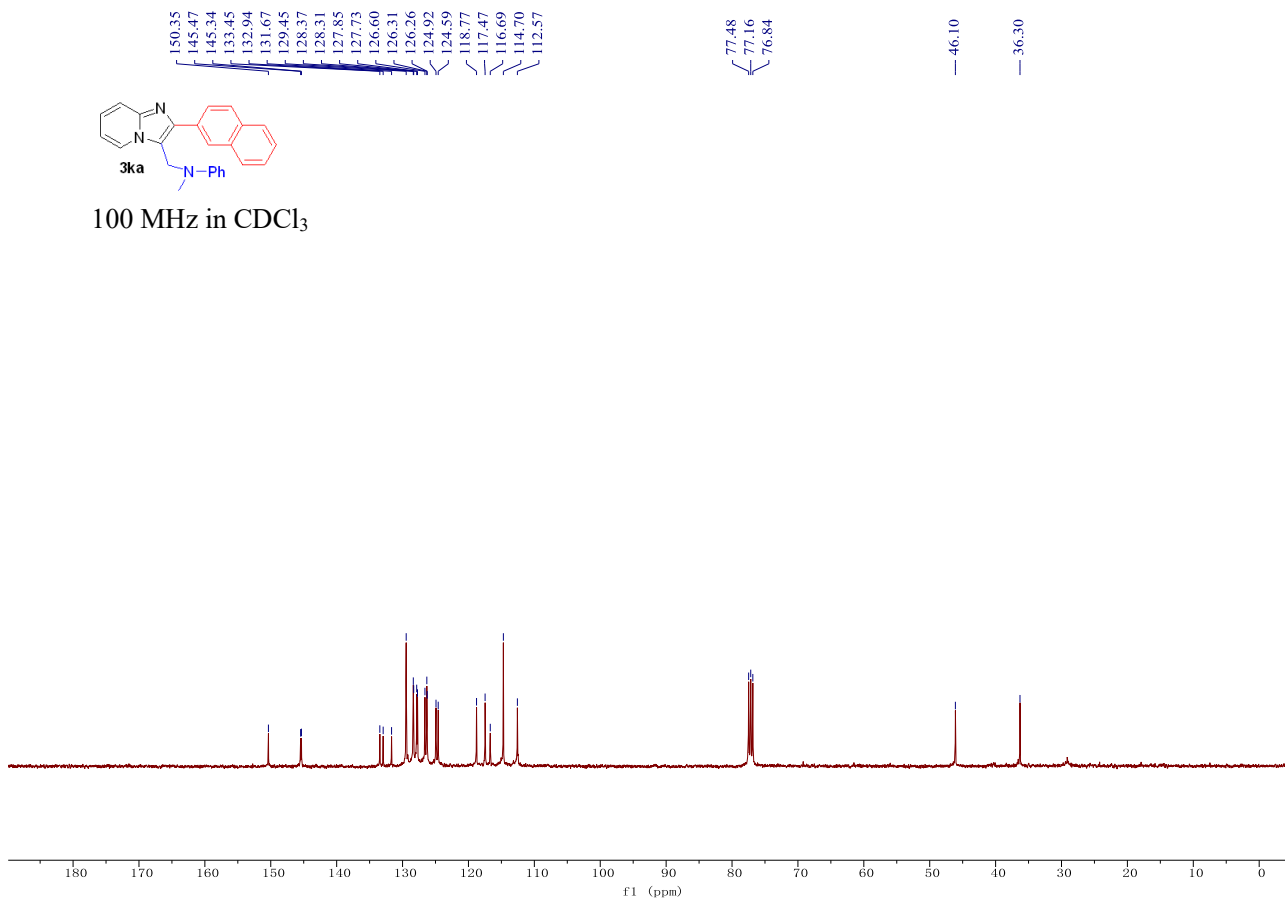
8.2601  
8.0883  
8.0850  
8.0710  
8.0679  
7.9964  
7.9867  
7.9752  
7.9709  
7.9472  
7.9258  
7.8832  
7.8788  
7.8761  
7.8682  
7.8602  
7.8577  
7.8530  
7.7414  
7.7379  
7.7185  
7.7152  
7.5060  
7.4978  
7.4903  
7.4821  
7.3469  
7.3287  
7.3248  
7.3067  
7.2604  
7.2473  
7.2438  
7.2247  
7.0149  
7.0127  
6.9930  
6.9095  
6.8939  
6.8912  
6.8729  
6.8035  
6.8004  
6.7866  
6.7835  
6.7694  
4.8752  
2.7248

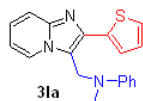


400 MHz in CDCl<sub>3</sub>

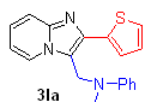
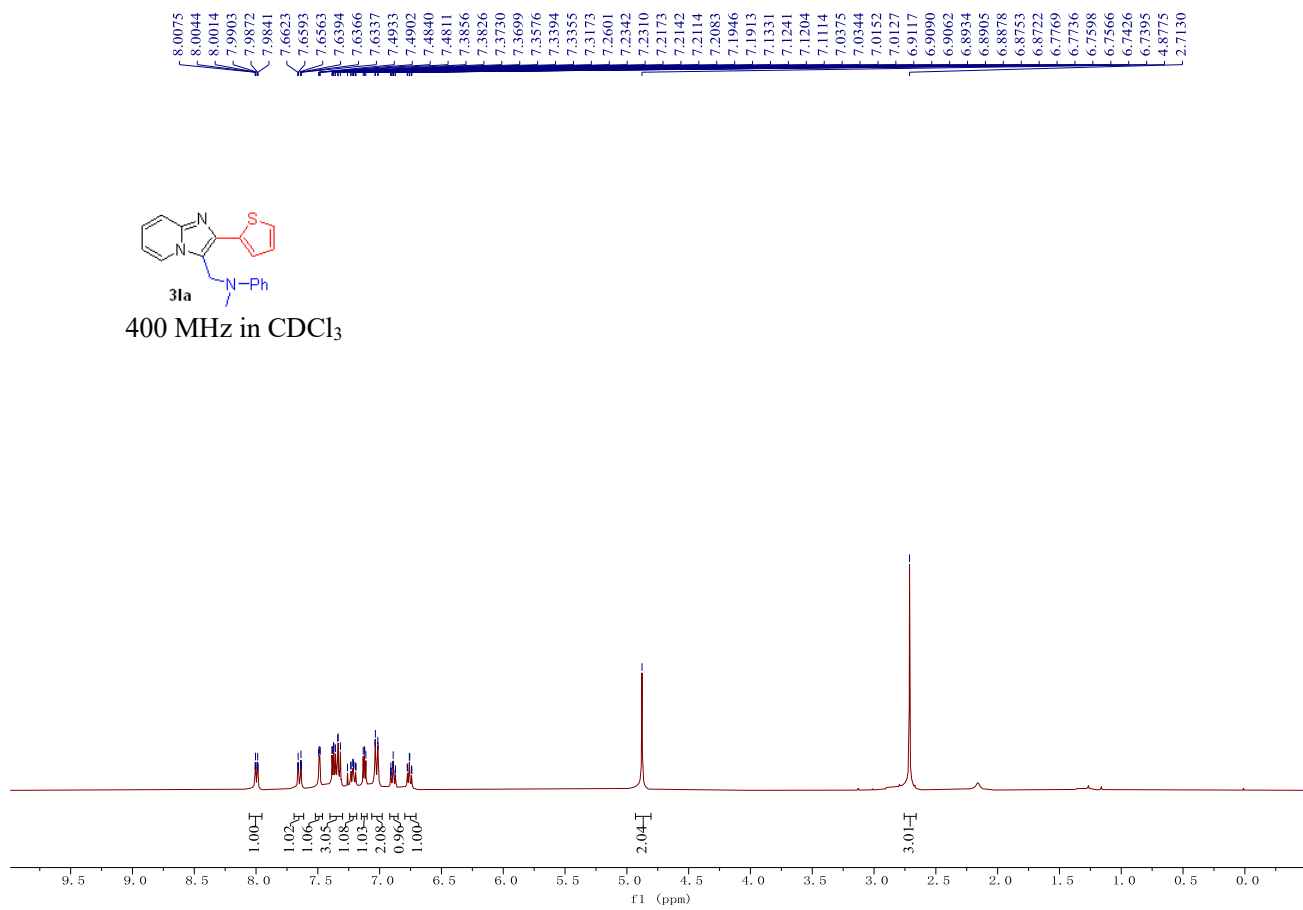


100 MHz in CDCl<sub>3</sub>

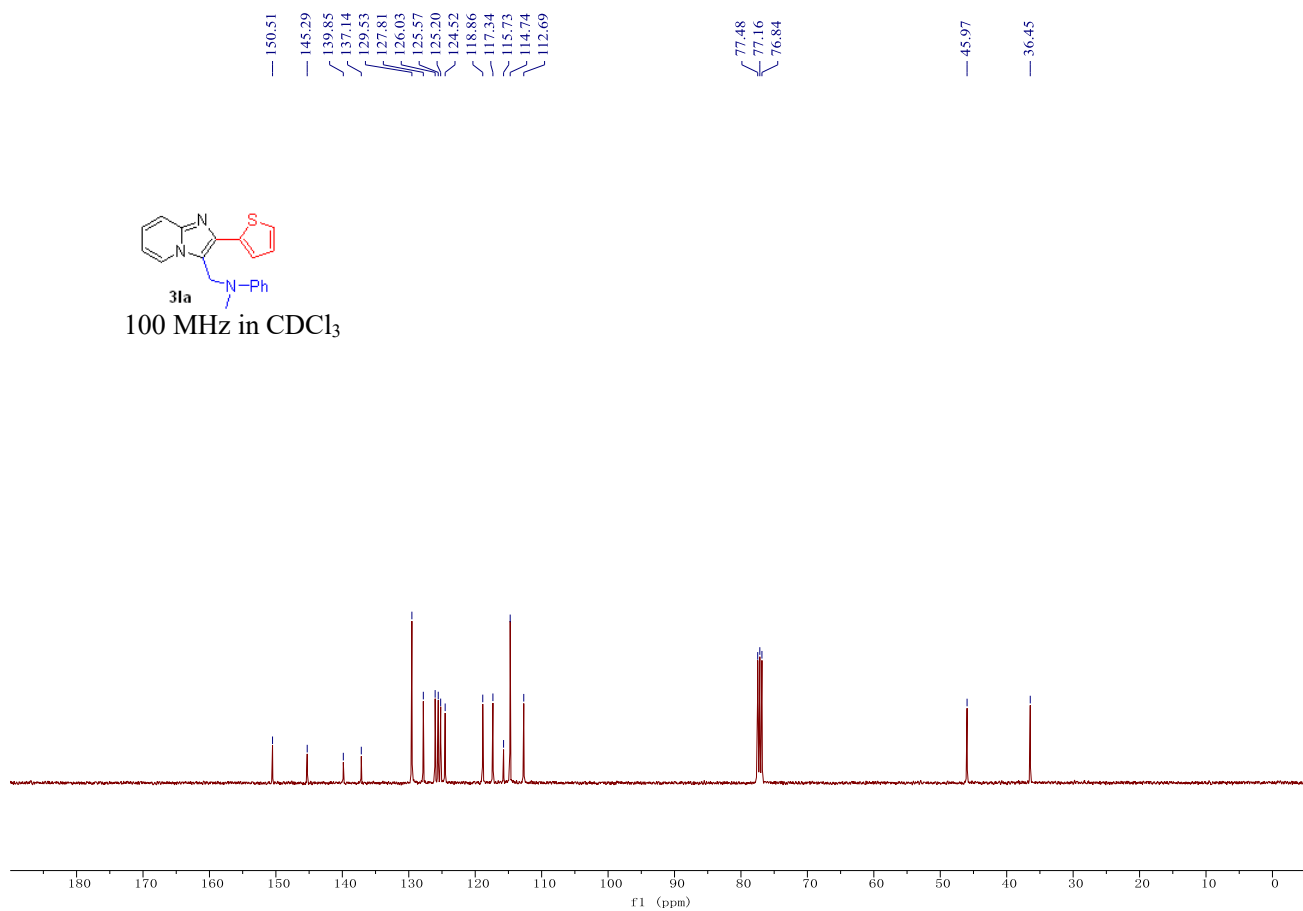




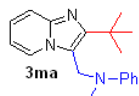
400 MHz in CDCl<sub>3</sub>



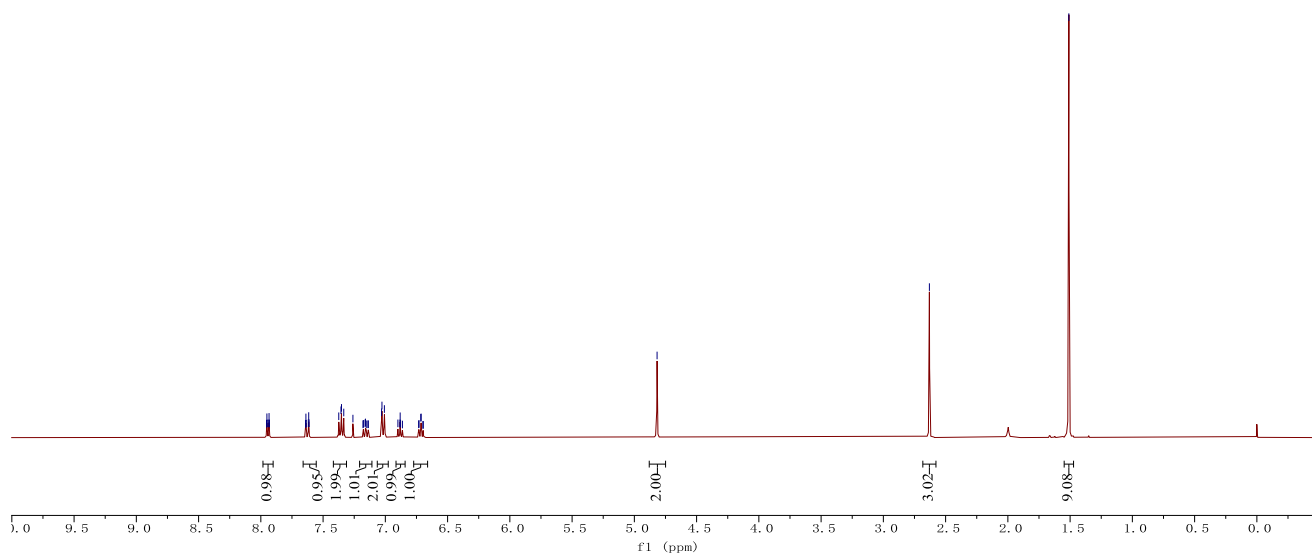
100 MHz in CDCl<sub>3</sub>



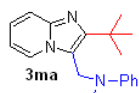
7.9544  
7.9512  
7.9481  
7.9373  
7.9341  
7.9309  
7.6411  
7.6381  
7.6351  
7.6186  
7.6155  
7.6124  
7.3741  
7.3562  
7.3522  
7.3340  
7.2605  
7.1794  
7.1762  
7.1627  
7.1595  
7.1535  
7.1399  
7.1367  
7.0302  
7.0272  
7.0076  
6.8991  
6.8838  
6.8808  
6.8779  
6.8626  
6.7325  
6.7294  
6.7154  
6.7123  
6.6985  
6.6954  
4.8175



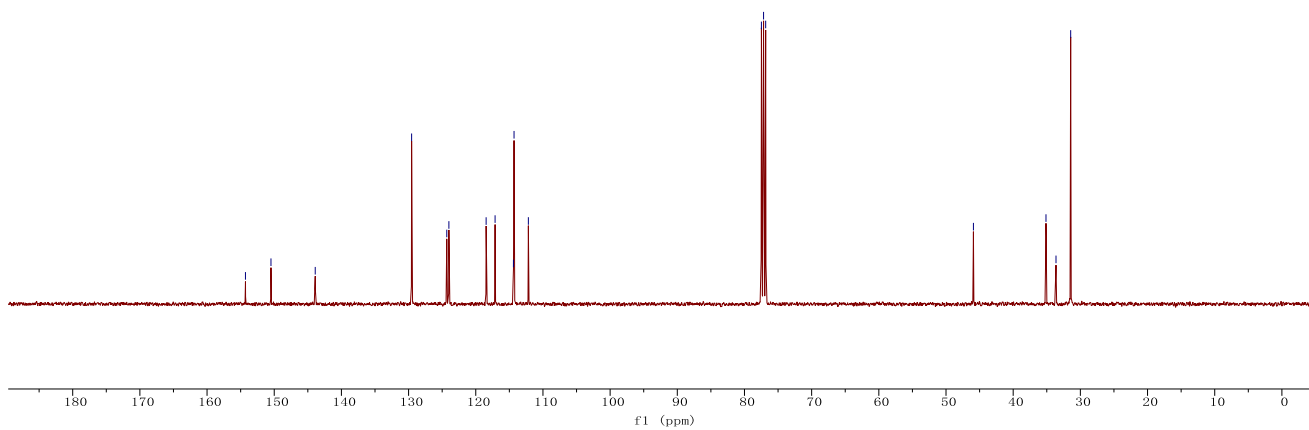
400 MHz in CDCl<sub>3</sub>

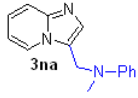


154.28  
150.49  
143.90  
129.55  
124.32  
124.00  
118.46  
117.12  
114.39  
114.30  
112.17  
77.48  
77.16  
76.84  
45.92  
35.12  
33.63  
31.44

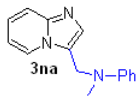
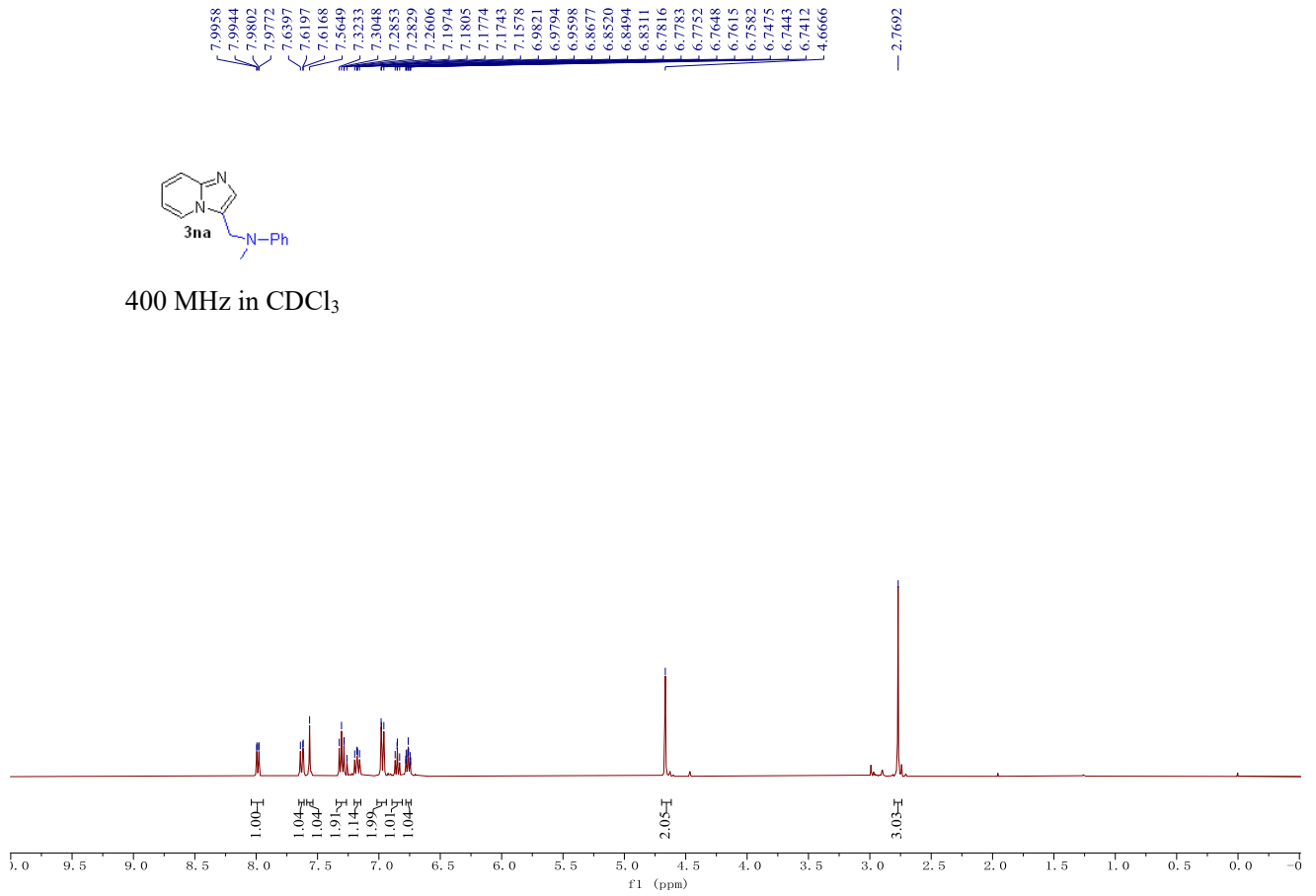


100 MHz in CDCl<sub>3</sub>

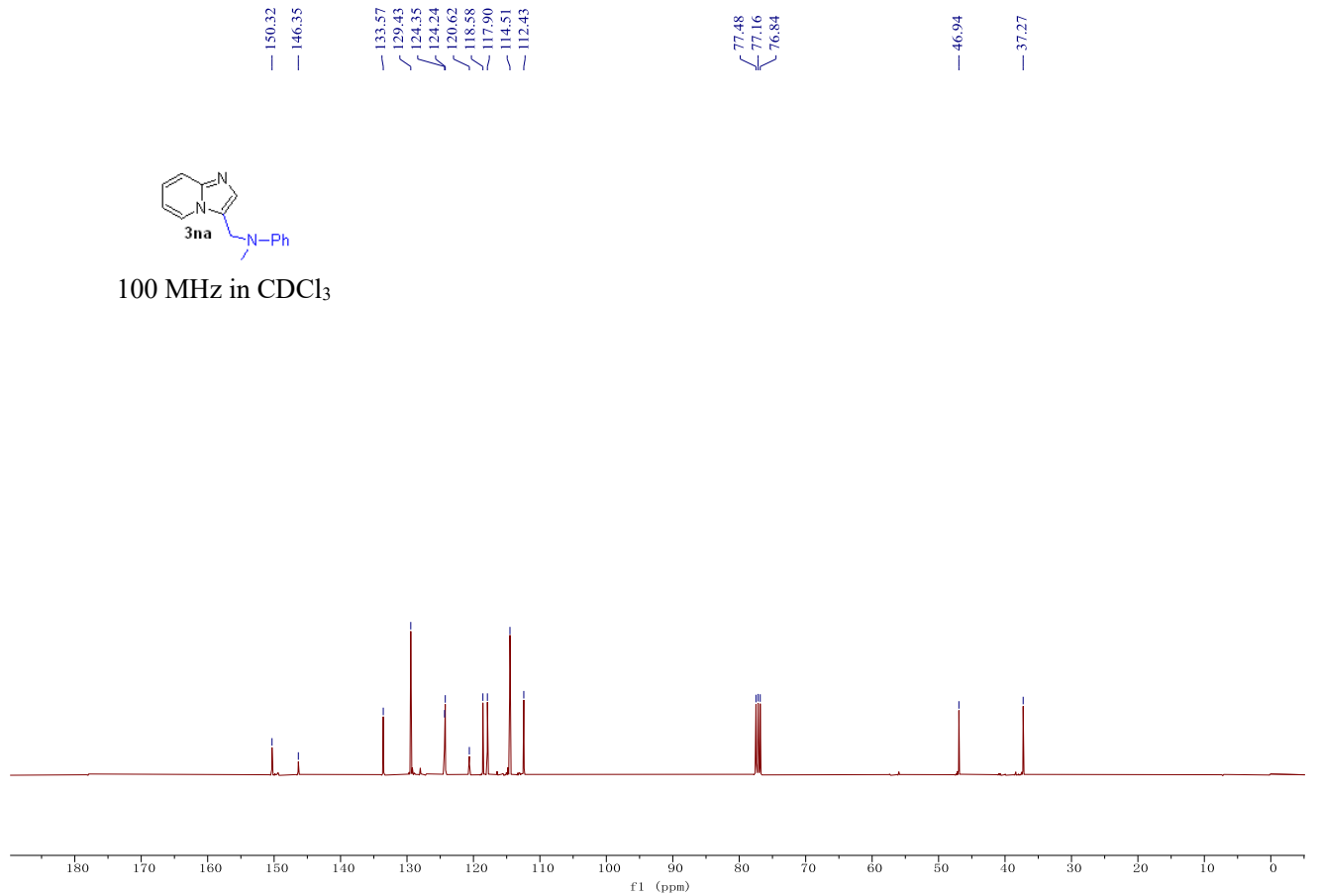


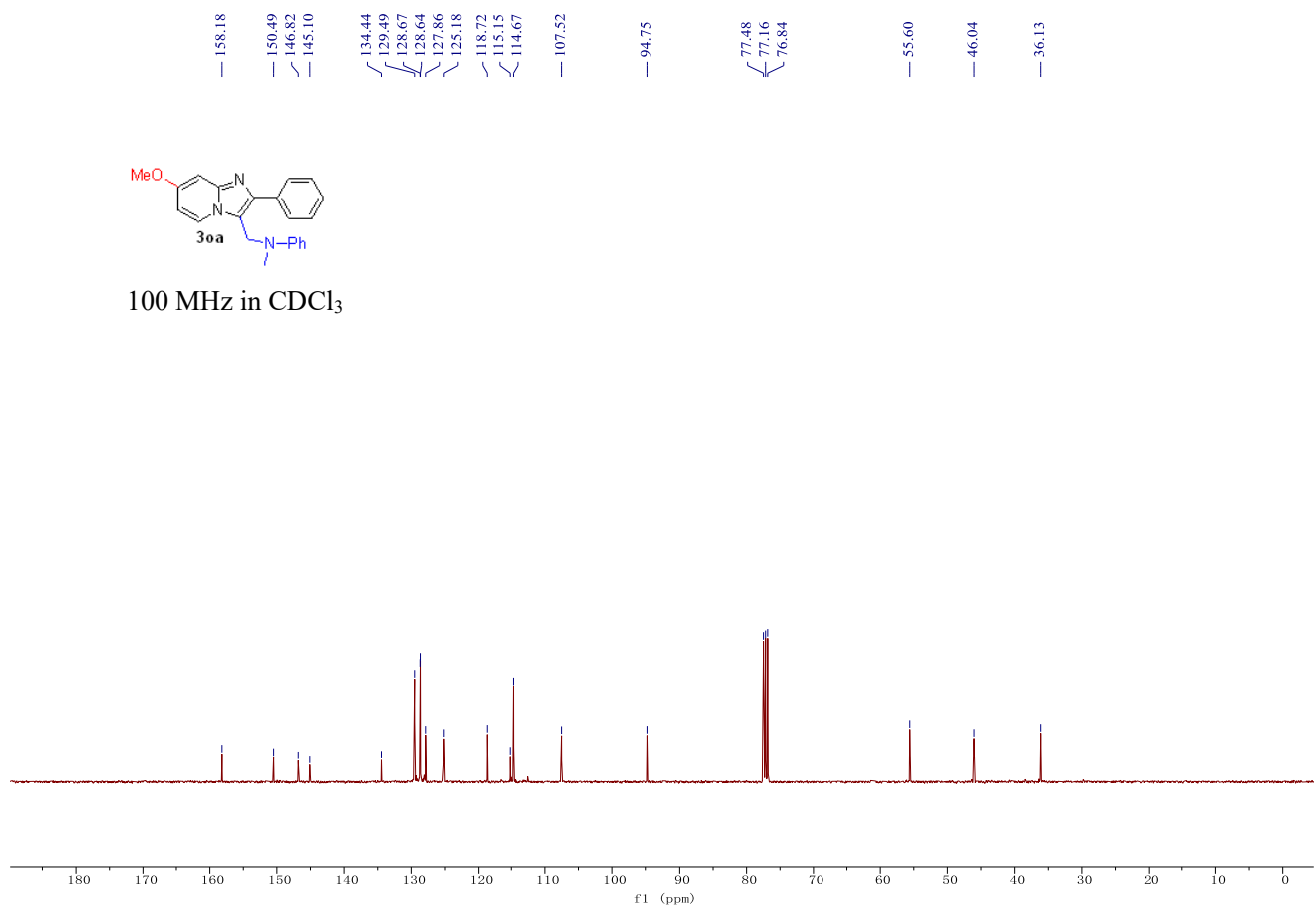
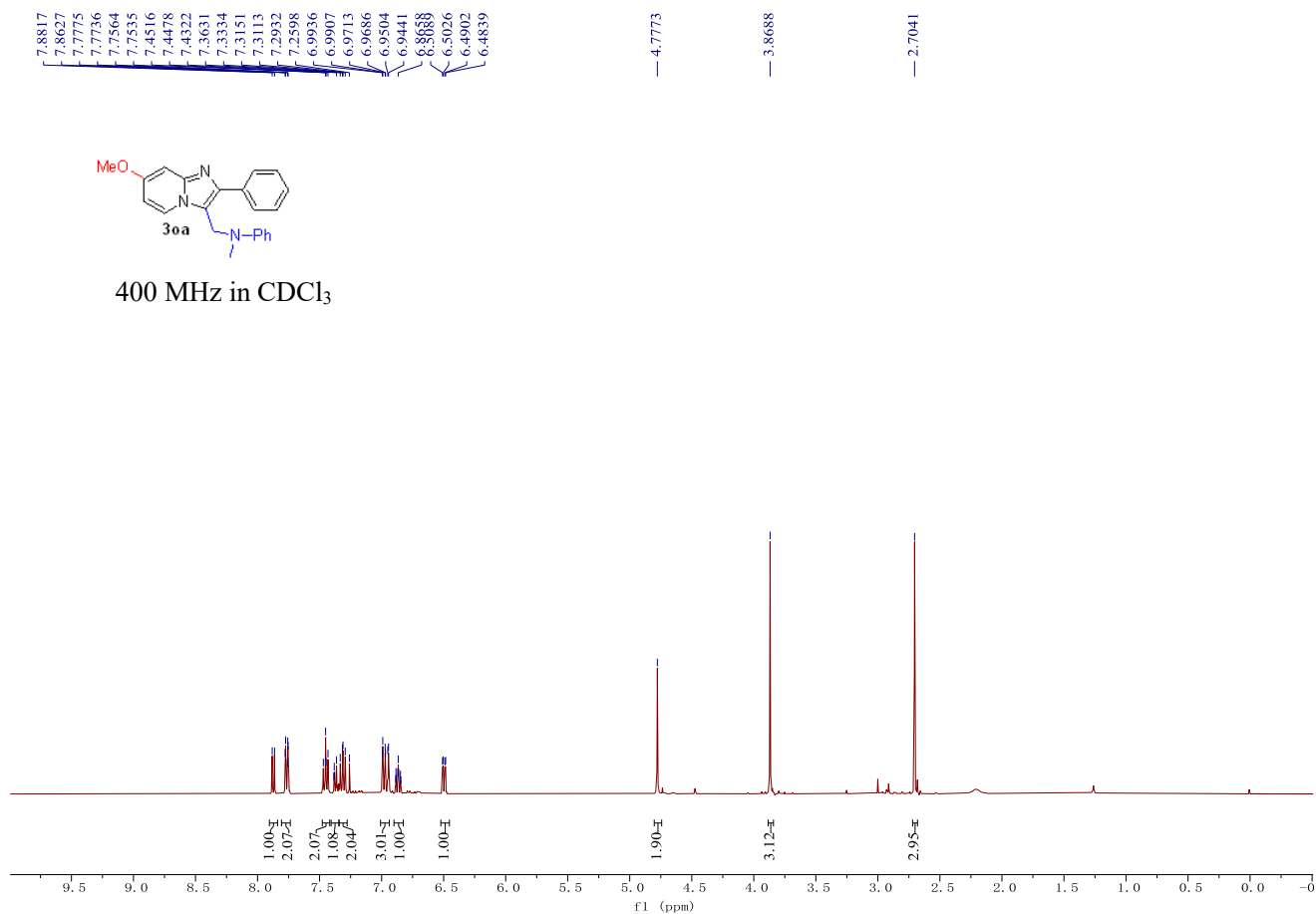


400 MHz in CDCl<sub>3</sub>

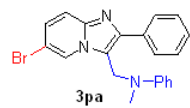


100 MHz in CDCl<sub>3</sub>

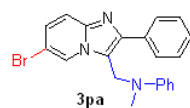
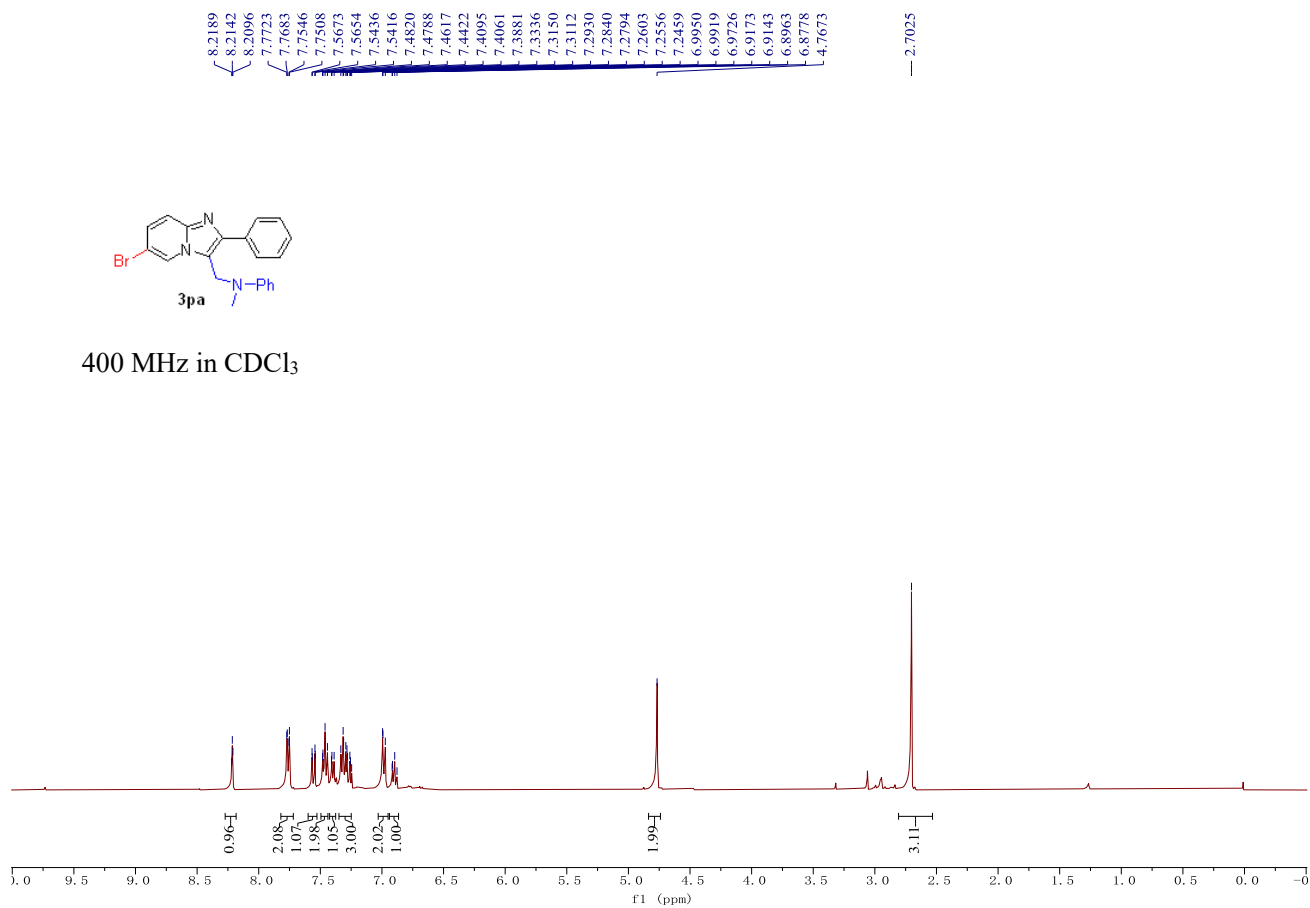




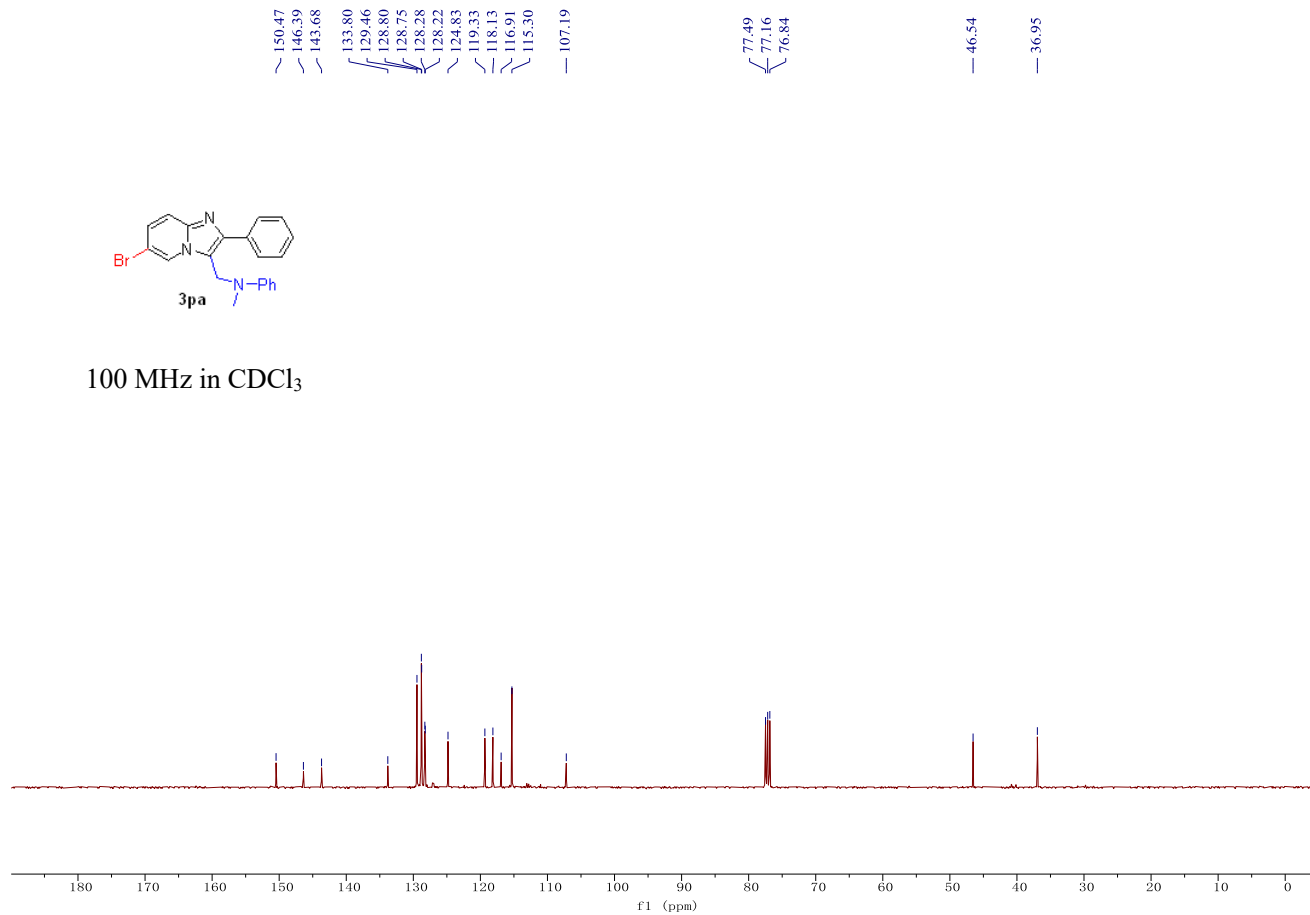


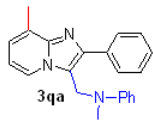


400 MHz in CDCl<sub>3</sub>

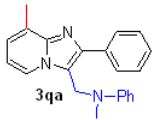
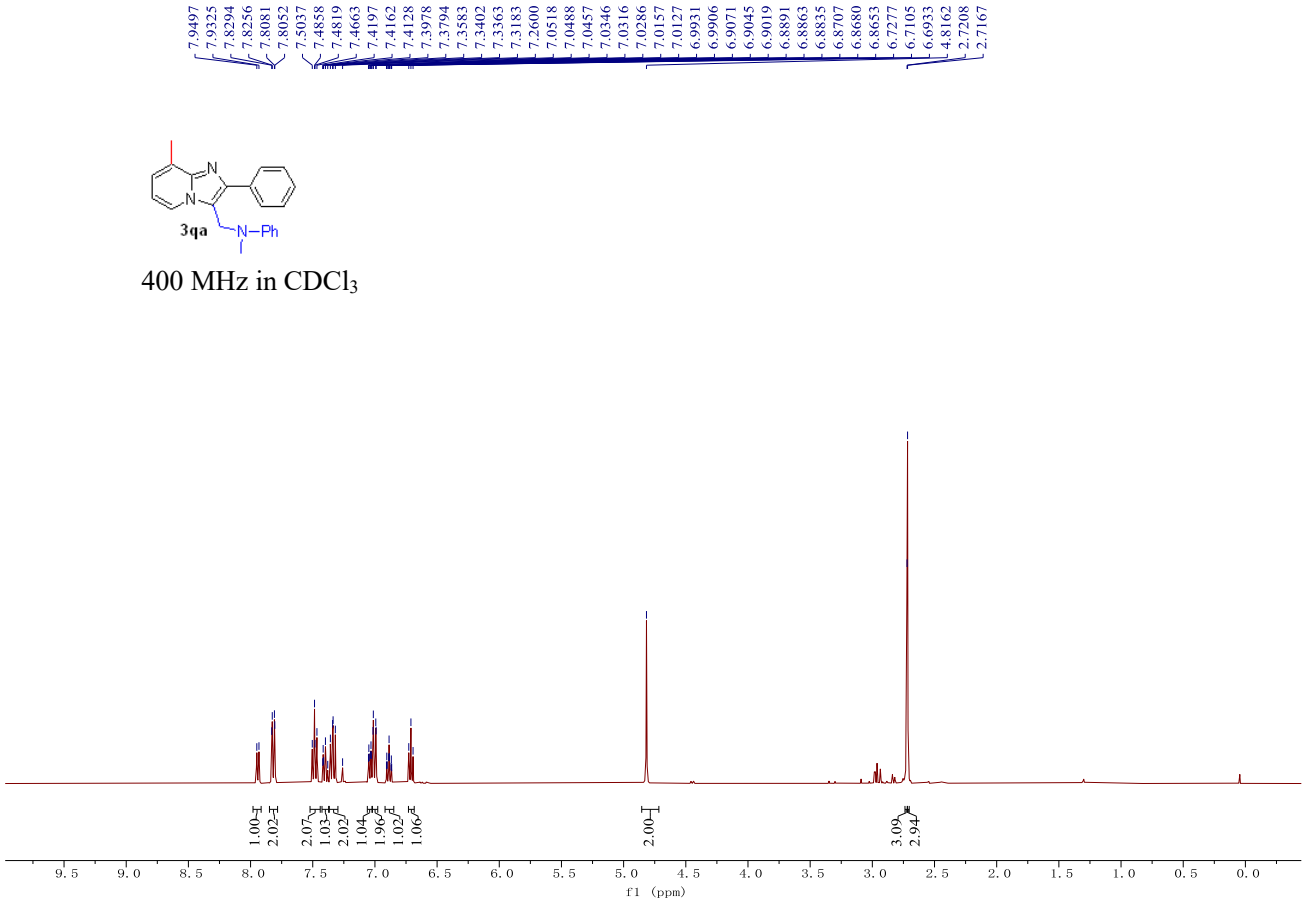


100 MHz in CDCl<sub>3</sub>

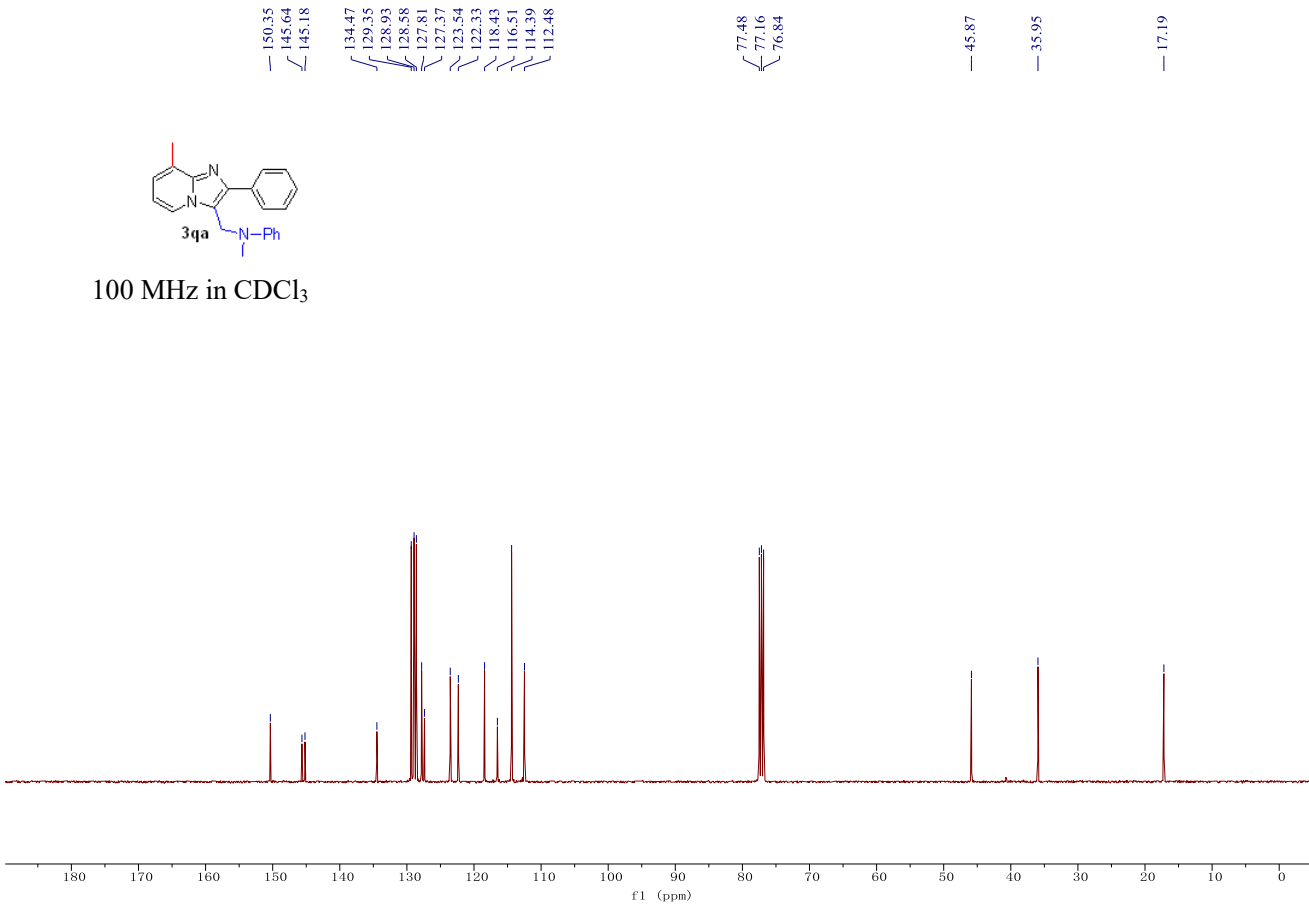


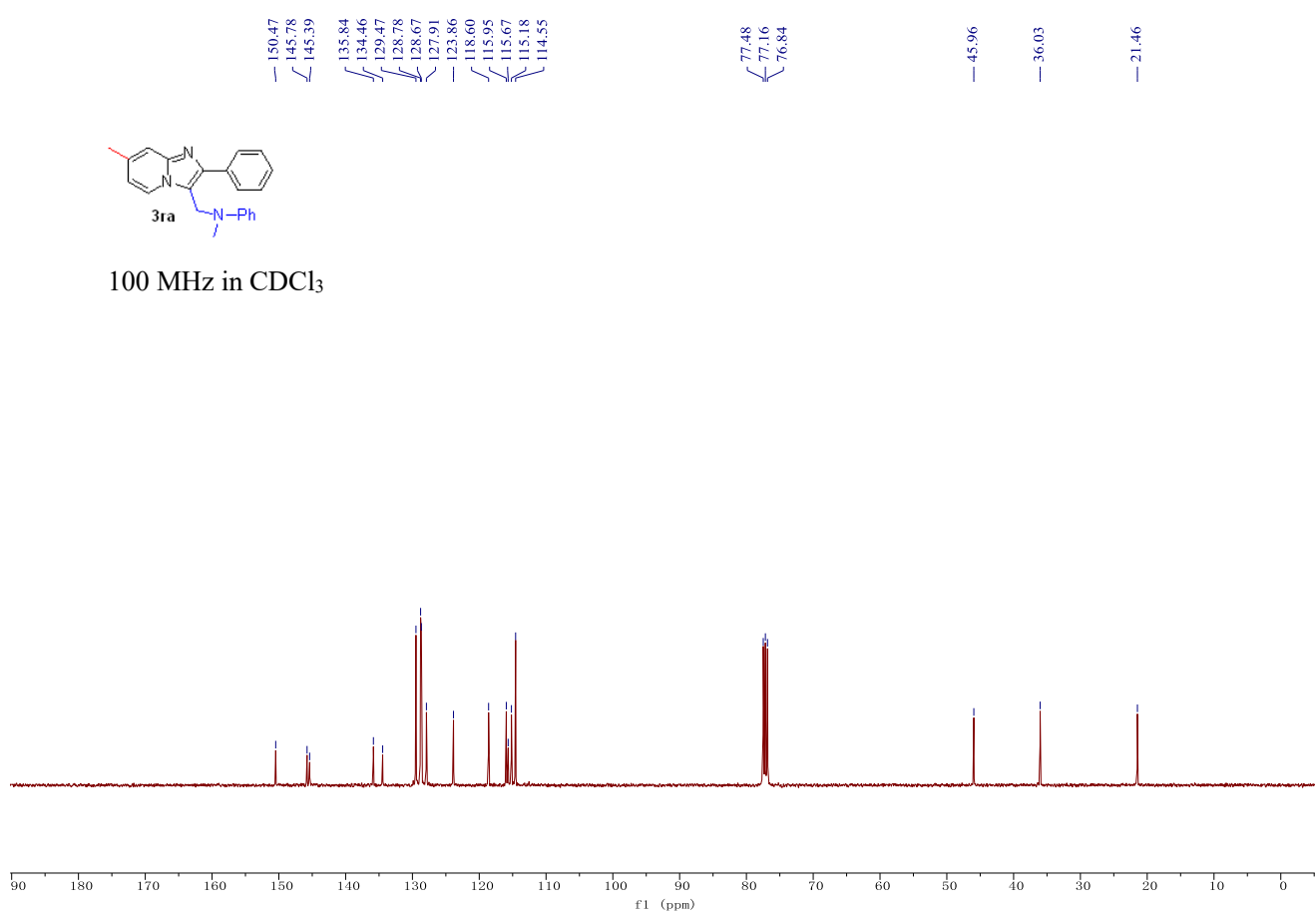
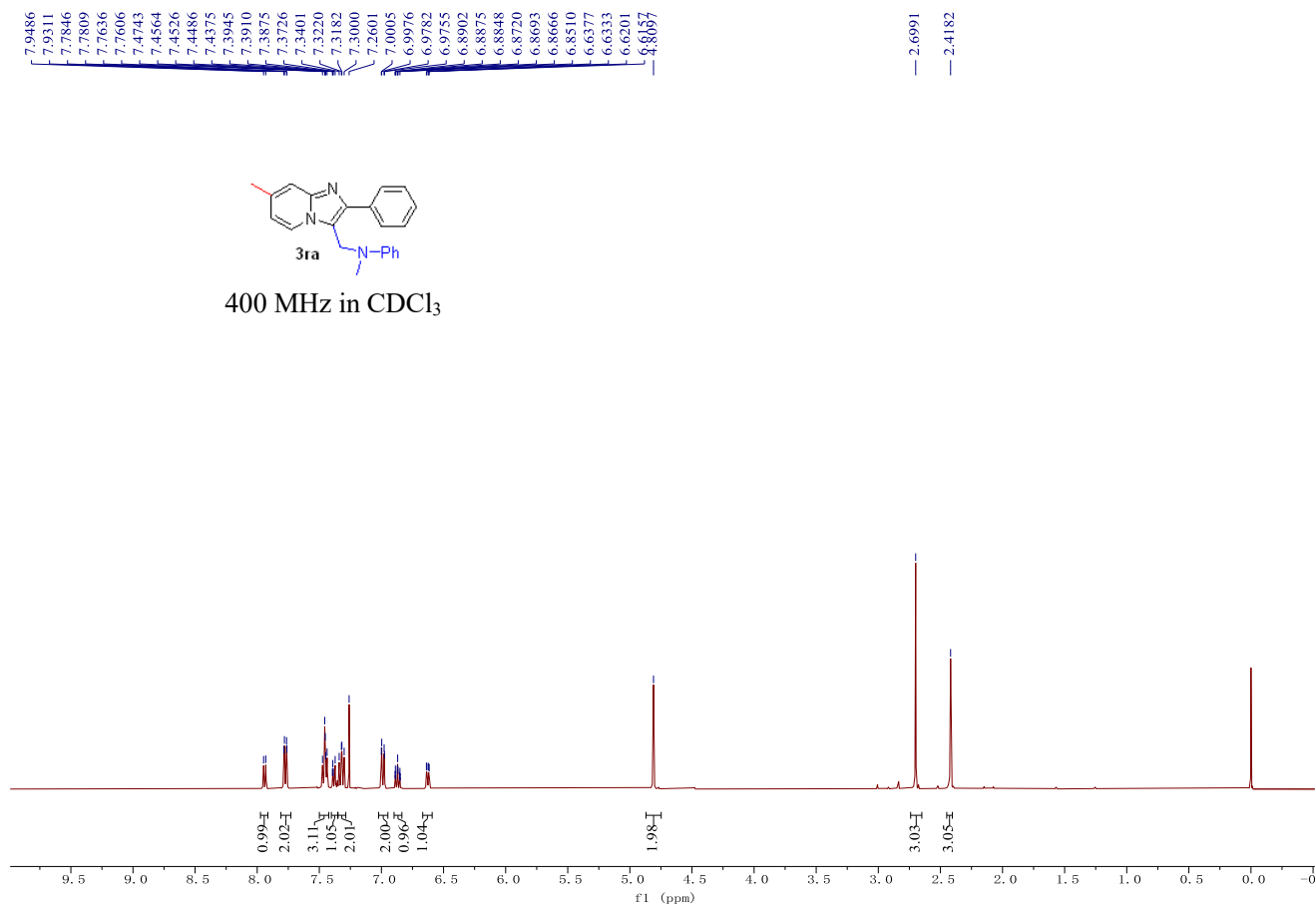


400 MHz in CDCl<sub>3</sub>

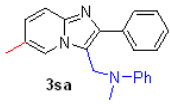


100 MHz in CDCl<sub>3</sub>

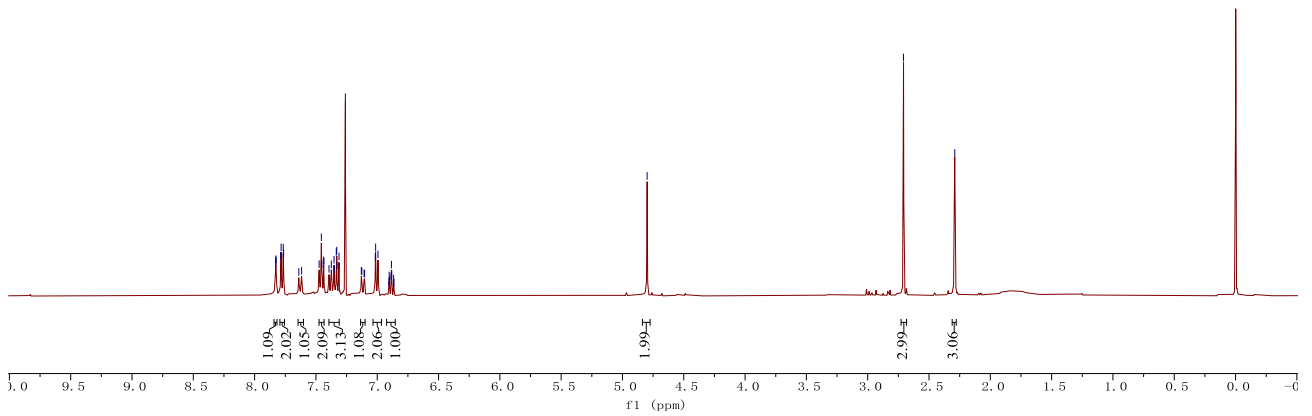




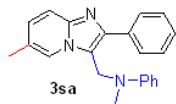
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7.7877  
7.7838  
7.7664  
7.7635  
7.6397  
7.6168  
7.4737  
7.4558  
7.4520  
7.4369  
7.4352  
7.3933  
7.3745  
7.3523  
7.3506  
7.3340  
7.3303  
7.3284  
7.3118  
7.2599  
7.1300  
7.1258  
7.1072  
7.1029  
7.0161  
7.0131  
6.9939  
6.9051  
6.9024  
6.9000  
6.8869  
6.8842  
6.8818  
6.8684  
6.8660  
6.8636  
4.7988



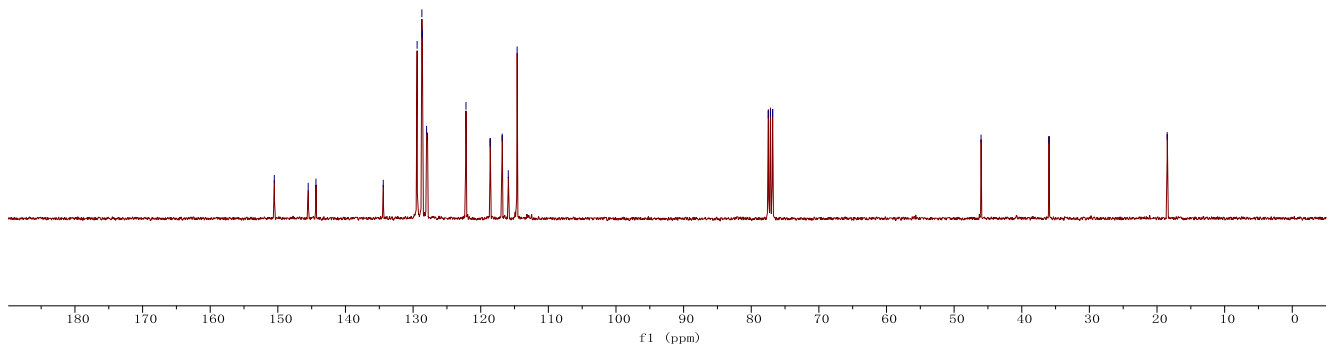
400 MHz in CDCl<sub>3</sub>



150.52  
145.51  
144.37  
134.41  
129.41  
128.70  
128.64  
128.01  
127.88  
122.18  
118.61  
116.83  
115.93  
114.62  
77.48  
77.16  
76.85  
46.02  
35.98  
18.49

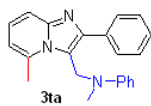


100 MHz in CDCl<sub>3</sub>

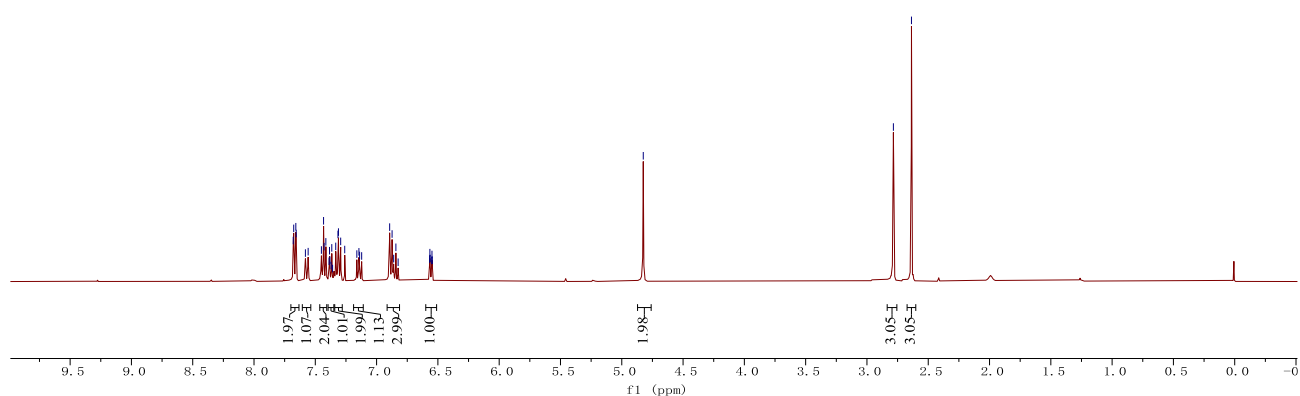


7.6801  
7.6761  
7.6588  
7.6559  
7.5807  
7.5583  
7.4502  
7.4326  
7.4286  
7.4140  
7.3875  
7.3840  
7.3805  
7.3716  
7.3655  
7.3334  
7.3151  
7.3112  
7.2929  
7.2600  
7.1612  
7.1442  
7.1386  
7.1218  
6.8935  
6.8737  
6.8607  
6.8427  
6.8244  
6.5681  
6.5653  
6.5624  
6.5511  
6.5482  
6.5374  
6.5274

— 2.7843  
— 2.6368



400 MHz in CDCl<sub>3</sub>



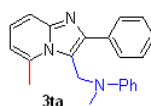
149.29  
148.14  
147.37  
137.09  
134.52  
129.45  
129.26  
128.65  
128.07  
125.47  
118.06  
117.13  
115.89  
113.94  
113.65

77.47  
77.16  
76.84

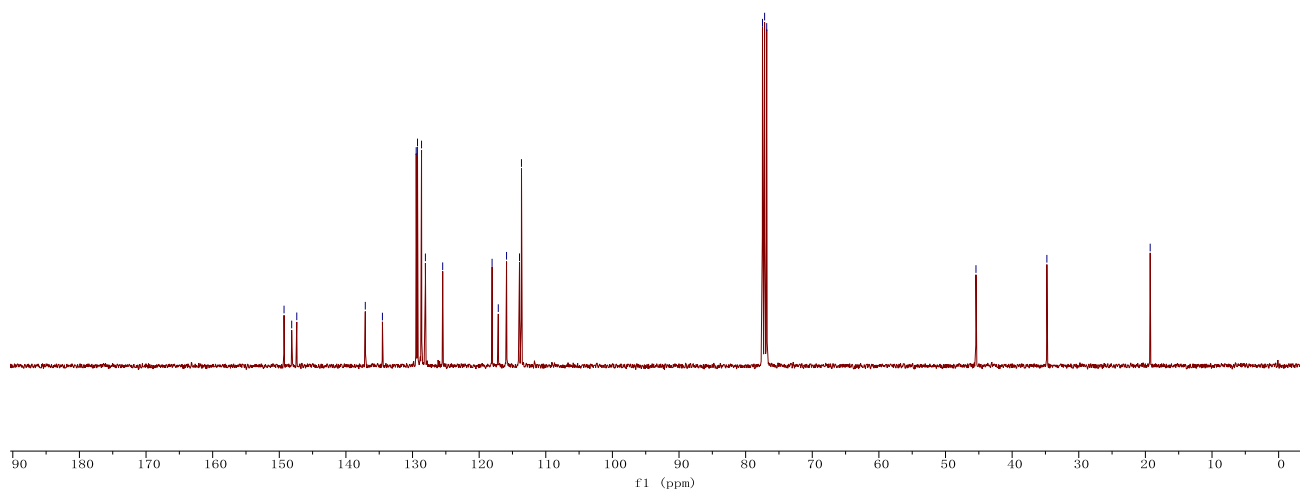
— 45.43

— 34.78

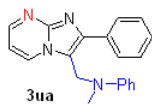
— 19.28



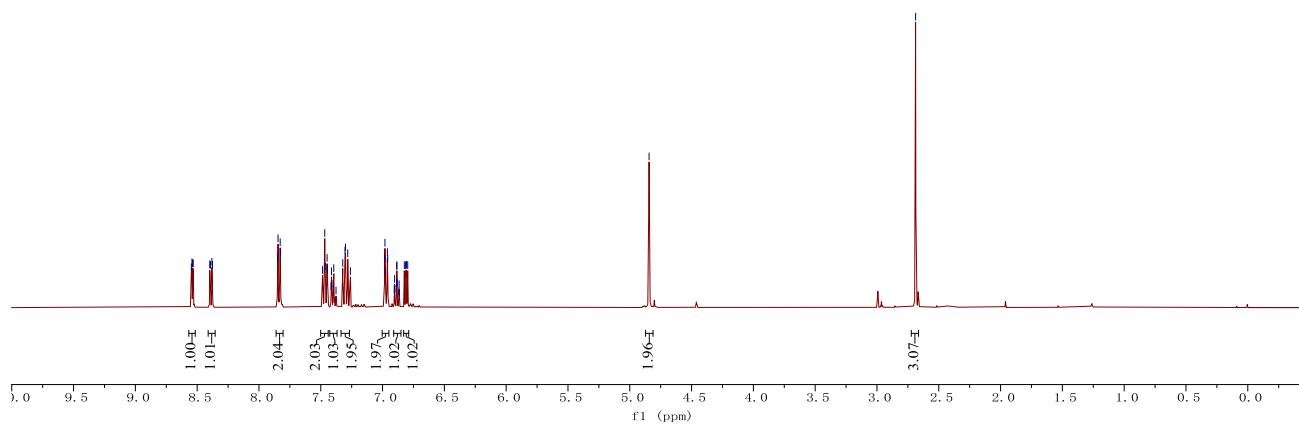
100 MHz in CDCl<sub>3</sub>



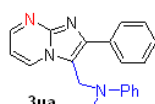
8.5474  
8.5423  
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8.5320  
8.3983  
8.3931  
8.3811  
8.3760  
7.8496  
7.8457  
7.8283  
7.8253  
7.4856  
7.4678  
7.4659  
7.4489  
7.4470  
7.4169  
7.4133  
7.4098  
7.3950  
7.3766  
7.3221  
7.3039  
7.3001  
7.2818  
7.2600  
6.9834  
6.9807  
6.9781  
6.9611  
6.9585  
6.9055  
6.9028  
6.9003  
6.8873  
6.8848  
6.8845  
6.8819  
6.8689  
6.8663  
6.8636  
6.8240  
6.8136  
6.8068  
6.7965  
4.8438  
2.6876



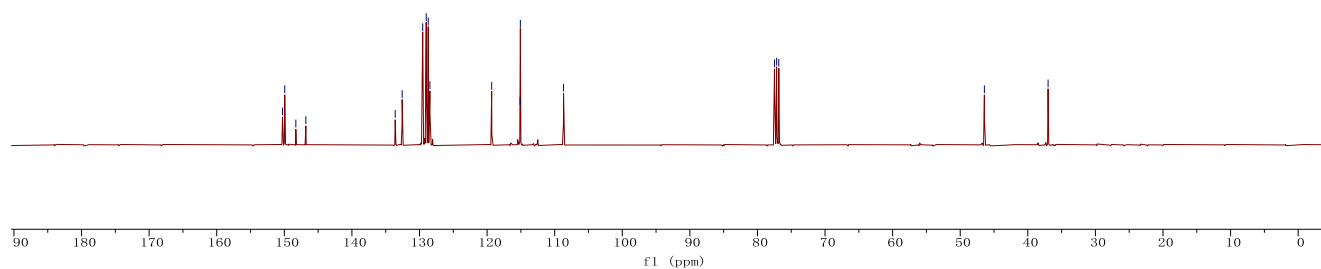
400 MHz in CDCl<sub>3</sub>



150.27  
149.93  
148.30  
146.82  
133.59  
132.56  
129.53  
129.00  
128.71  
128.46  
119.32  
115.11  
115.08  
108.69  
77.48  
77.16  
76.84  
46.43  
37.00

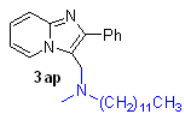


100 MHz in CDCl<sub>3</sub>

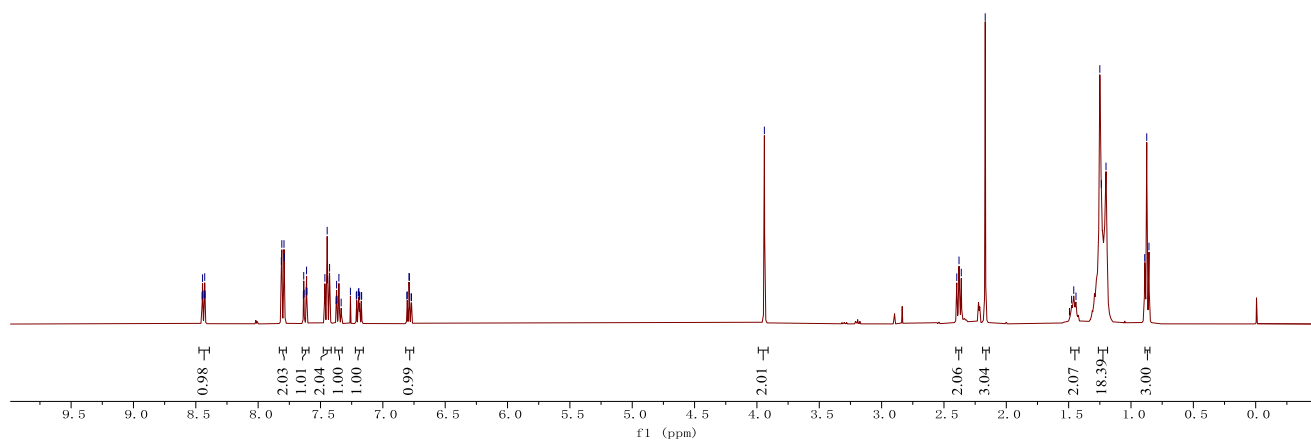


8.4489  
8.4459  
8.4428  
8.4317  
8.4386  
8.4255  
7.8140  
7.8103  
7.7928  
7.7899  
7.6374  
7.6345  
7.6315  
7.6147  
7.6118  
7.6088  
7.4648  
7.4466  
7.4430  
7.4271  
7.3744  
7.3710  
7.3677  
7.3525  
7.3342  
7.2600  
7.2126  
7.2095  
7.1959  
7.1927  
7.1900  
7.1867  
7.1734  
7.1700  
6.8073  
6.8043  
6.7902  
6.7871  
6.7732  
6.7702  
3.9402

2.3973  
2.3793  
2.3611  
2.1691  
1.4931  
1.4764  
1.4592  
1.4413  
1.2379  
1.2379  
1.2006  
0.8901  
0.8738  
0.8561



400 MHz in CDCl<sub>3</sub>

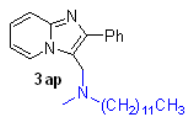


145.05  
144.85  
134.79  
129.02  
128.48  
127.69  
125.69  
124.49  
117.59  
117.23  
111.73

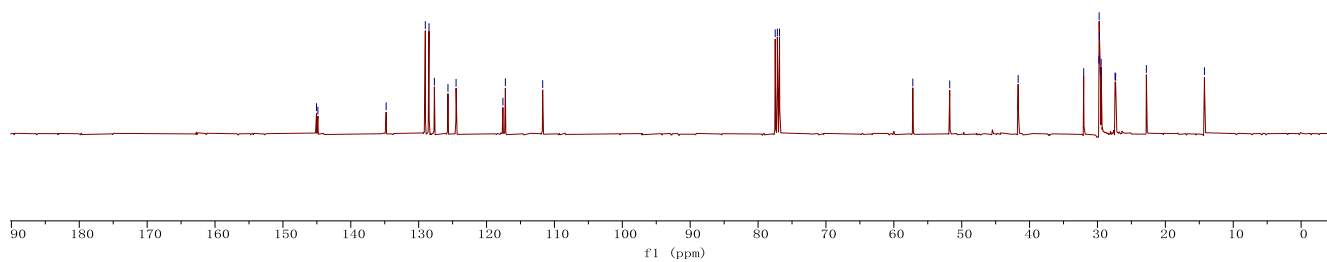
77.48  
77.16  
76.84

57.20  
51.77

41.68  
32.03  
29.78  
29.75  
29.72  
29.57  
29.46  
27.41  
27.33  
22.80  
14.24



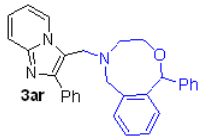
100 MHz in CDCl<sub>3</sub>



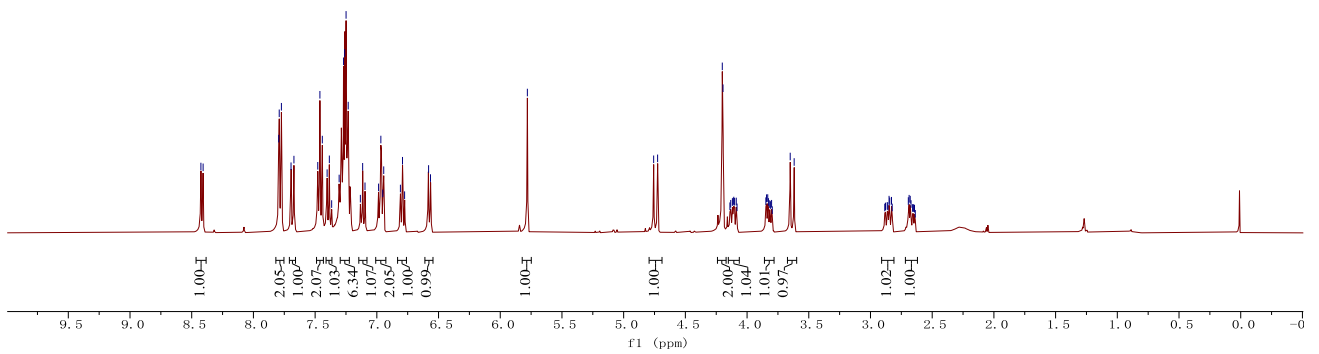




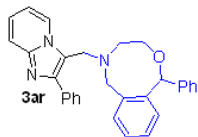
8.4256  
8.4076  
7.7951  
7.7909  
7.7732  
7.6952  
7.6717  
7.4786  
7.4613  
7.4418  
7.4029  
7.3848  
7.3661  
7.3054  
7.2696  
7.2607  
7.2493  
7.2319  
7.1334  
7.1149  
7.0959  
6.9858  
6.9674  
6.9521  
6.9441  
6.8090  
6.7920  
6.7750  
6.5807  
6.5653  
5.7803  
4.7570  
4.7248  
4.2008  
4.1950  
4.1390  
4.1325  
4.1190  
4.1124  
4.1076  
4.1012  
4.0877  
4.0813  
3.8478  
3.8422  
3.8326  
3.8269  
3.8165  
3.8110  
3.8013  
3.7958  
3.6504  
3.6184  
2.8842  
2.8786  
2.8643  
2.8585  
2.8495  
2.8440  
2.8297  
2.8240  
2.6935  
2.6869  
2.6780  
2.6588  
2.6523  
1.6434



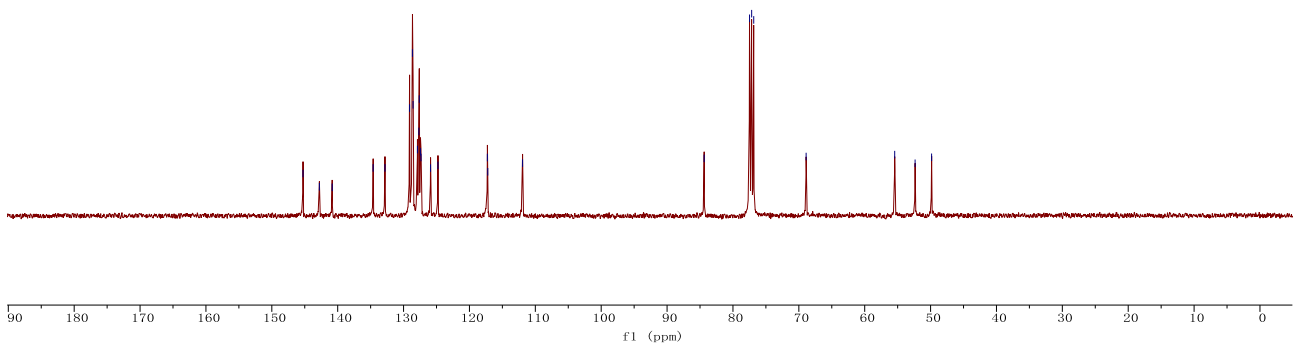
400 MHz in CDCl<sub>3</sub>



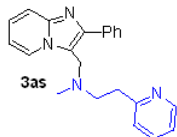
145.26  
142.78  
140.83  
134.62  
132.82  
129.09  
128.65  
128.56  
127.89  
127.68  
127.62  
127.44  
127.35  
125.91  
124.78  
117.28  
117.22  
111.94  
84.37  
77.48  
77.16  
76.85  
68.89  
55.45  
52.35  
49.85



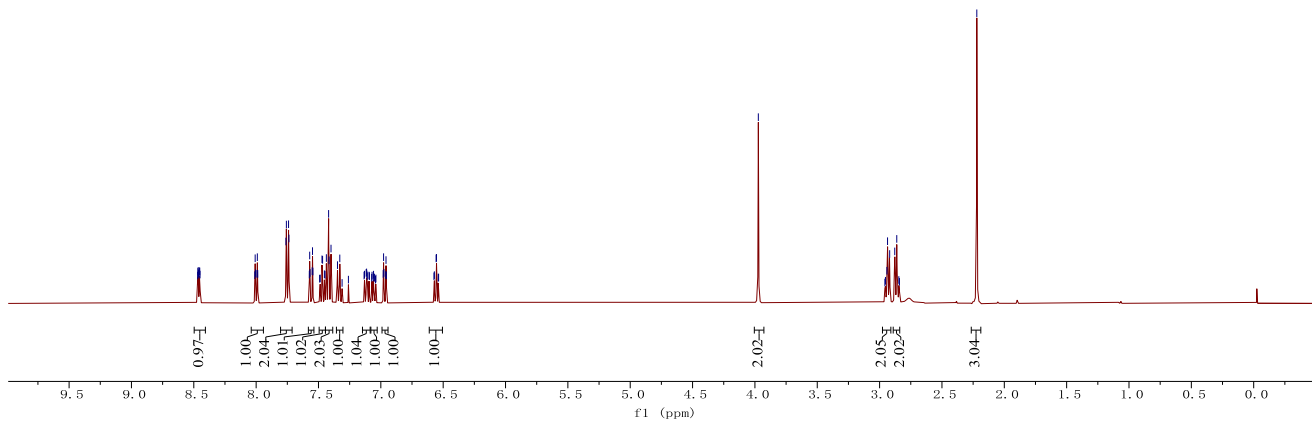
100 MHz in CDCl<sub>3</sub>



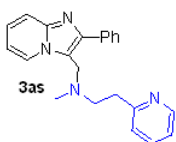
8.4693  
8.4671  
8.4647  
8.4623  
8.4570  
8.4548  
8.4523  
8.4500  
8.0107  
8.0078  
8.0048  
7.9934  
7.9904  
7.9874  
7.7612  
7.7575  
7.7400  
7.7371  
7.7334  
7.5706  
7.5678  
7.5507  
7.5479  
7.5450  
7.4909  
7.4863  
7.4719  
7.4672  
7.4527  
7.4480  
7.4368  
7.4188  
7.4151  
7.3995  
7.3476  
7.3291  
7.2600  
7.1344  
7.1311  
7.1176  
7.1143  
7.1117  
7.1084  
7.0949  
7.0916  
7.0736  
7.0706  
7.0612  
7.0584  
7.0547  
7.0517  
7.0426  
7.0396  
6.9807  
6.9780  
6.9753  
6.9612  
6.9585  
6.9557  
6.5737  
6.5707  
6.5567  
6.5536  
6.5397  
6.5367  
3.9726  
2.9414  
2.9194  
2.8785  
2.8624  
2.2205



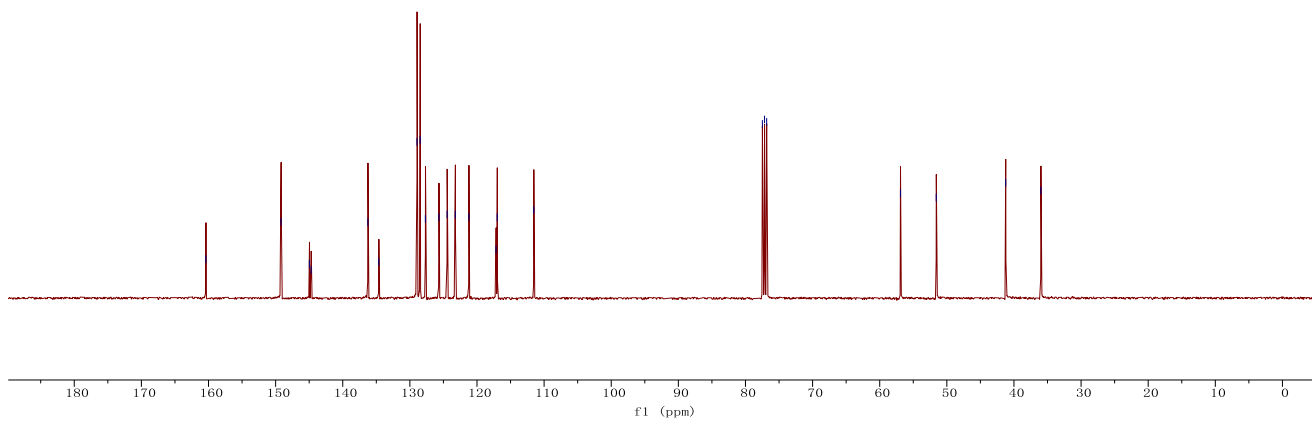
400 MHz in CDCl<sub>3</sub>



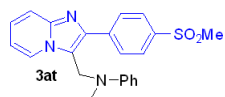
160.39  
149.19  
144.97  
144.71  
136.26  
134.63  
128.94  
128.47  
127.68  
125.68  
124.46  
123.23  
121.21  
117.16  
116.98  
111.54  
77.48  
77.16  
76.84  
56.91  
51.58  
41.23  
35.99



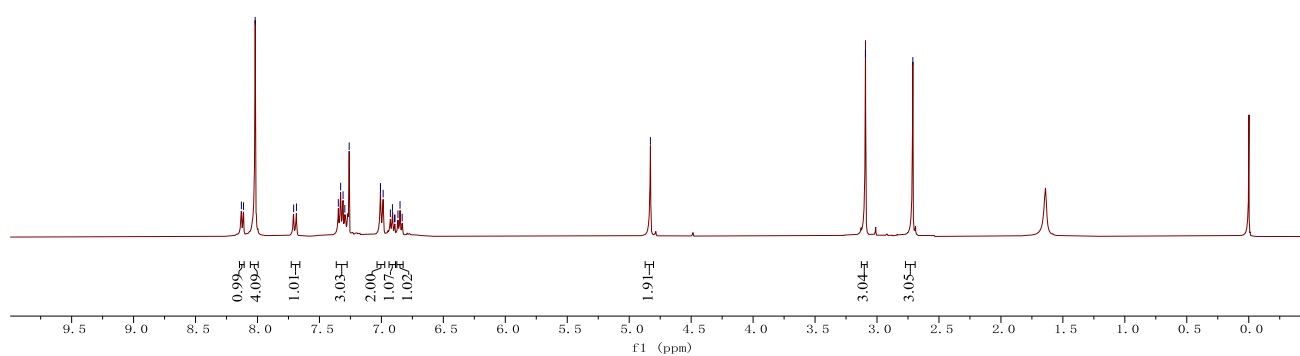
100 MHz in CDCl<sub>3</sub>



8.1300  
8.1127  
8.0188  
7.7084  
7.6856  
7.3477  
7.3291  
7.3094  
7.2953  
7.2600  
7.0078  
7.0072  
6.9860  
6.9275  
6.9109  
6.8935  
6.8909  
6.8667  
6.8496  
6.8324  
— 4.8290  
— 3.0934  
— 2.7109



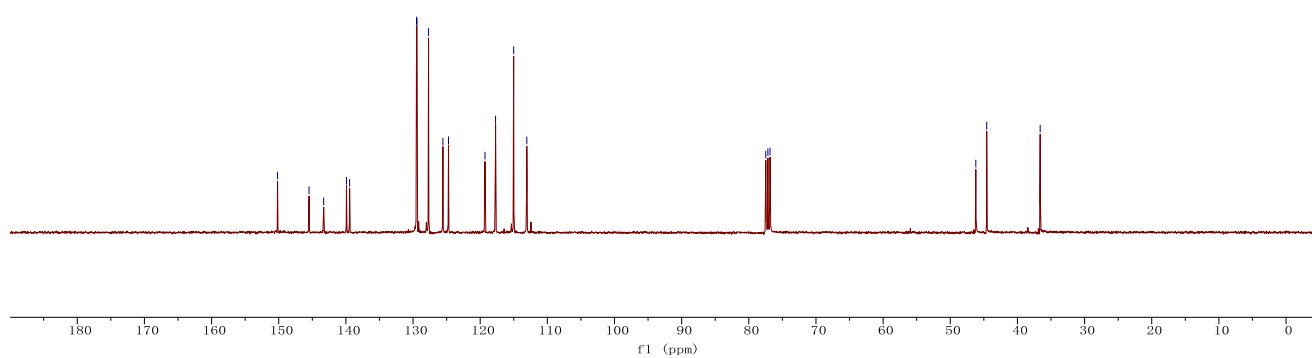
400 MHz in CDCl<sub>3</sub>

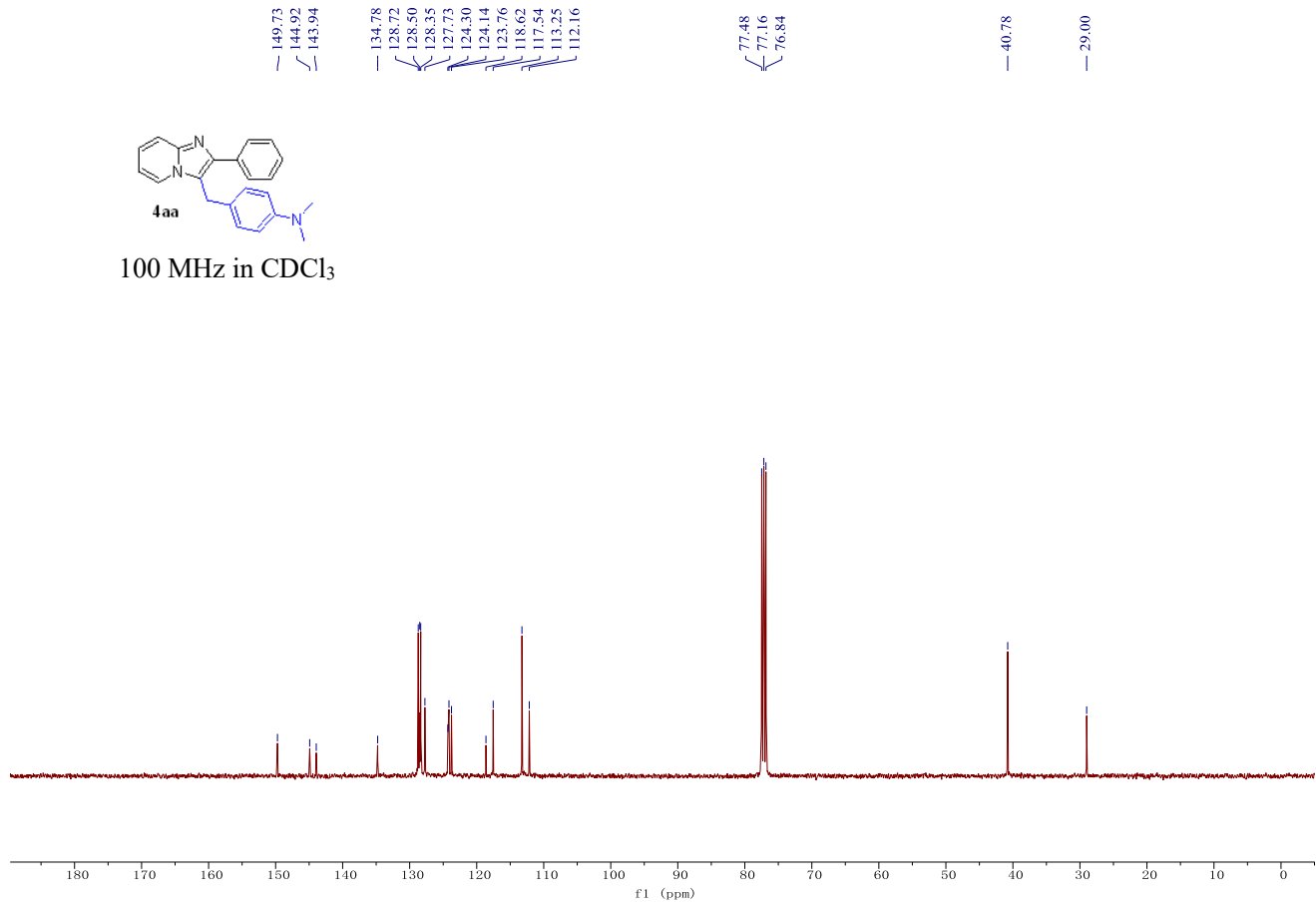
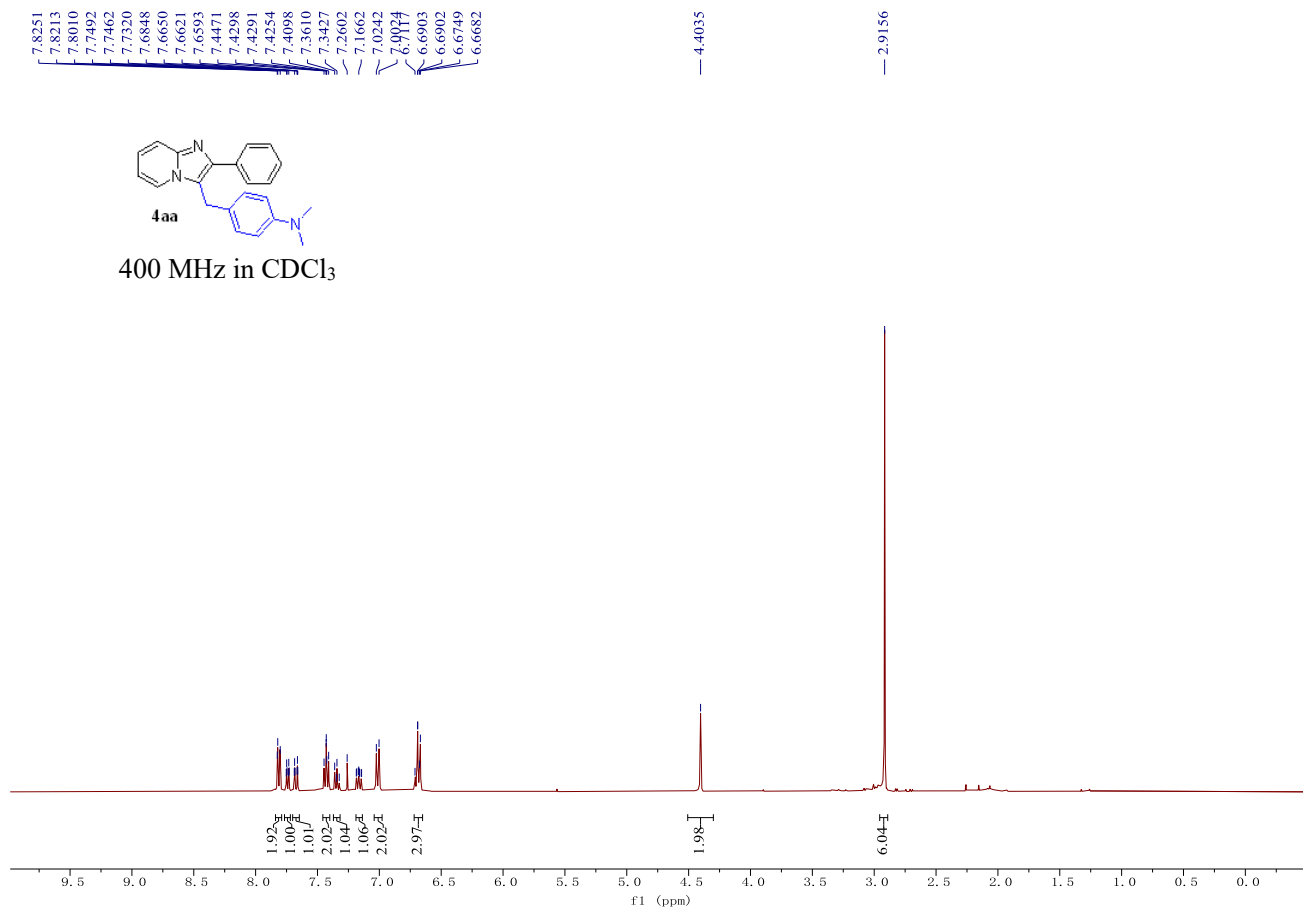


150.18  
145.49  
145.31  
139.91  
139.45  
129.48  
129.41  
127.69  
125.55  
124.72  
119.29  
117.71  
115.02  
113.06  
77.48  
77.16  
76.84  
46.20  
44.57  
36.61



100 MHz in CDCl<sub>3</sub>



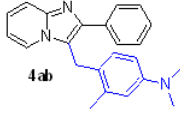


7.7681  
7.7644  
7.7470  
7.7440  
7.7129  
7.7100  
7.6903  
7.6874  
7.6679  
7.6649  
7.6508  
7.6478  
7.4322  
7.4143  
7.3954  
7.3937  
7.3505  
7.3321  
7.2601  
7.1843  
7.1813  
6.7183  
6.7012  
6.6913  
6.6875  
6.5525  
6.5313  
6.4281  
6.4212  
6.4069  
6.4000

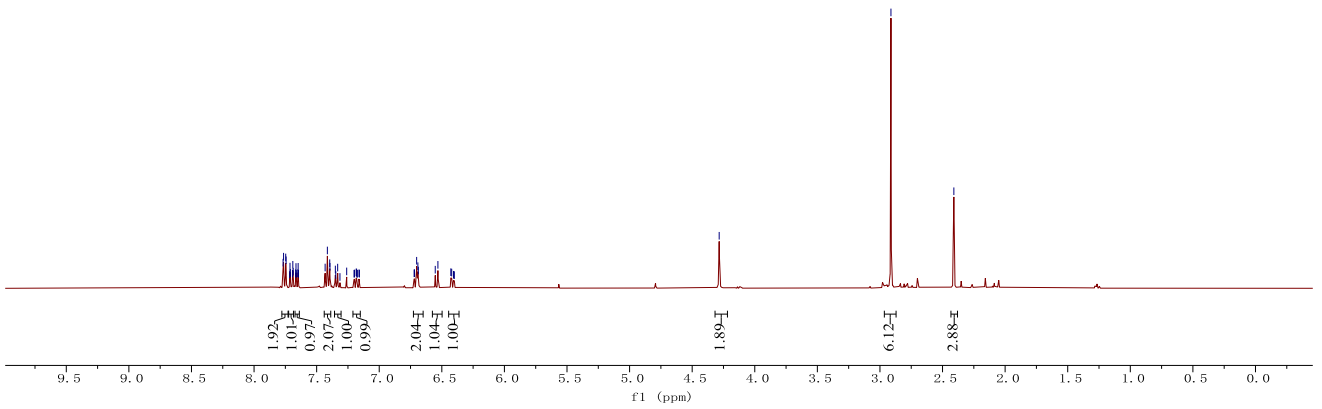
4.2843

2.9121

2.4096



400 MHz in CDCl<sub>3</sub>



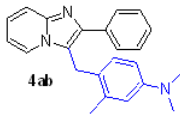
149.73  
144.94  
144.16  
137.03  
134.73  
128.68  
128.24  
127.65  
127.57  
124.08  
123.67  
122.40  
118.30  
117.52  
115.06  
112.17  
110.66

77.48  
77.16  
76.84

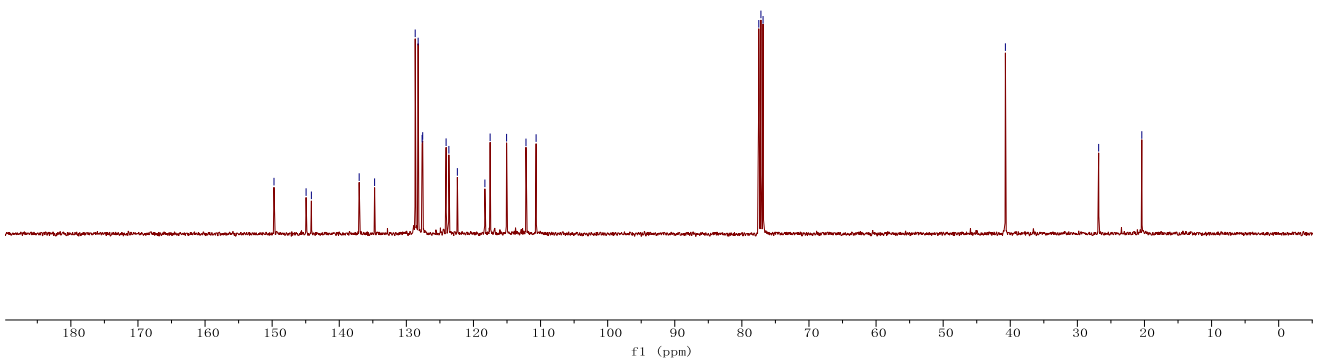
40.73

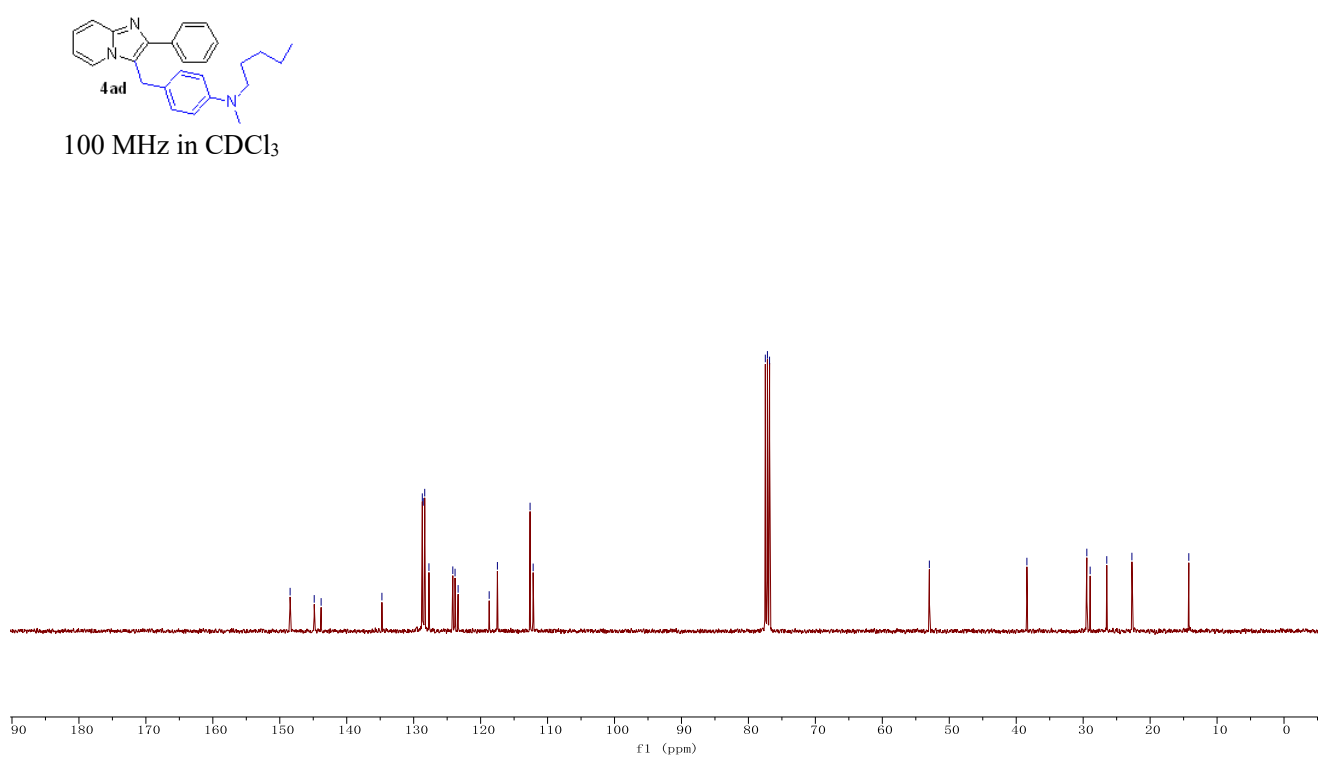
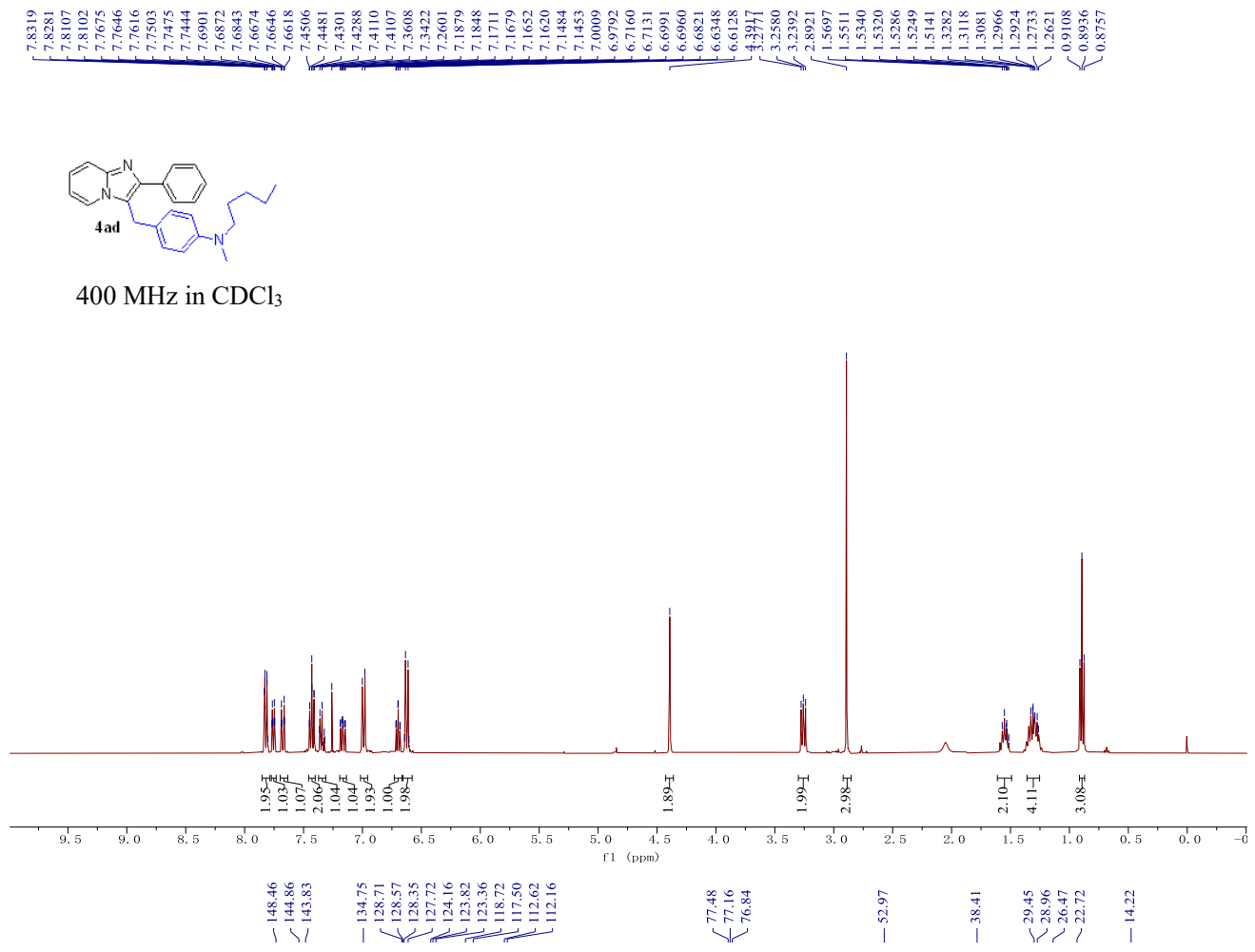
26.83

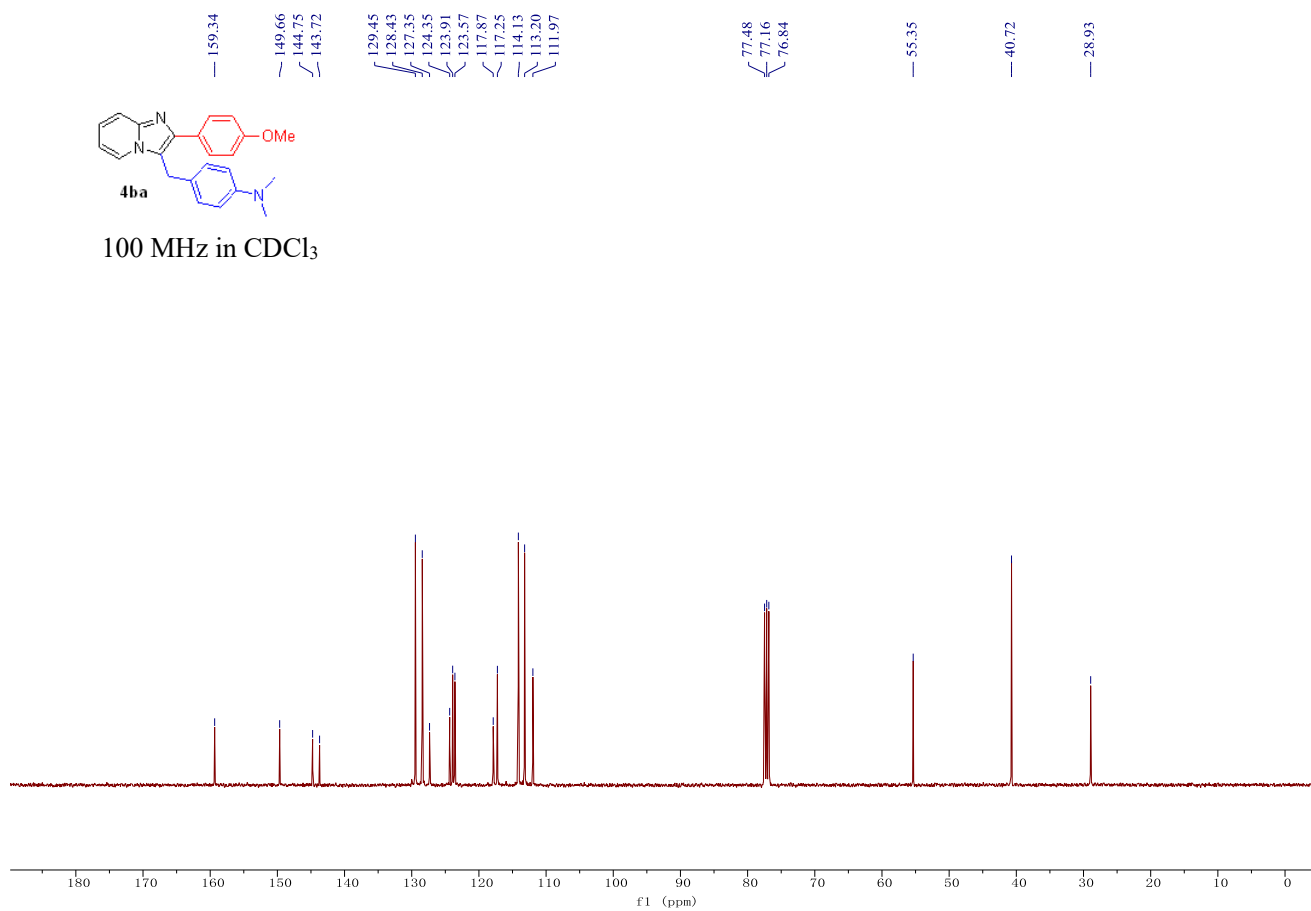
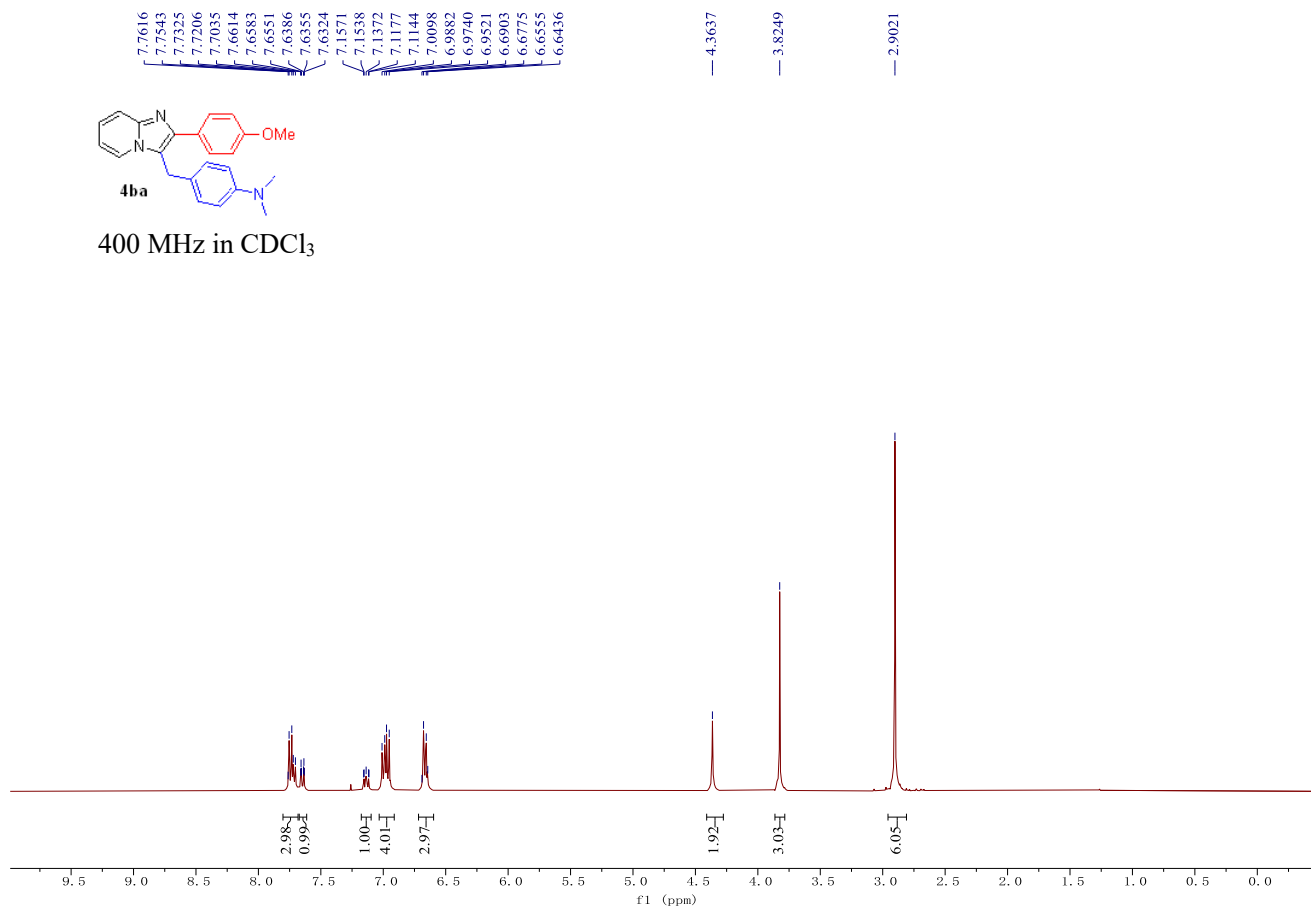
20.40

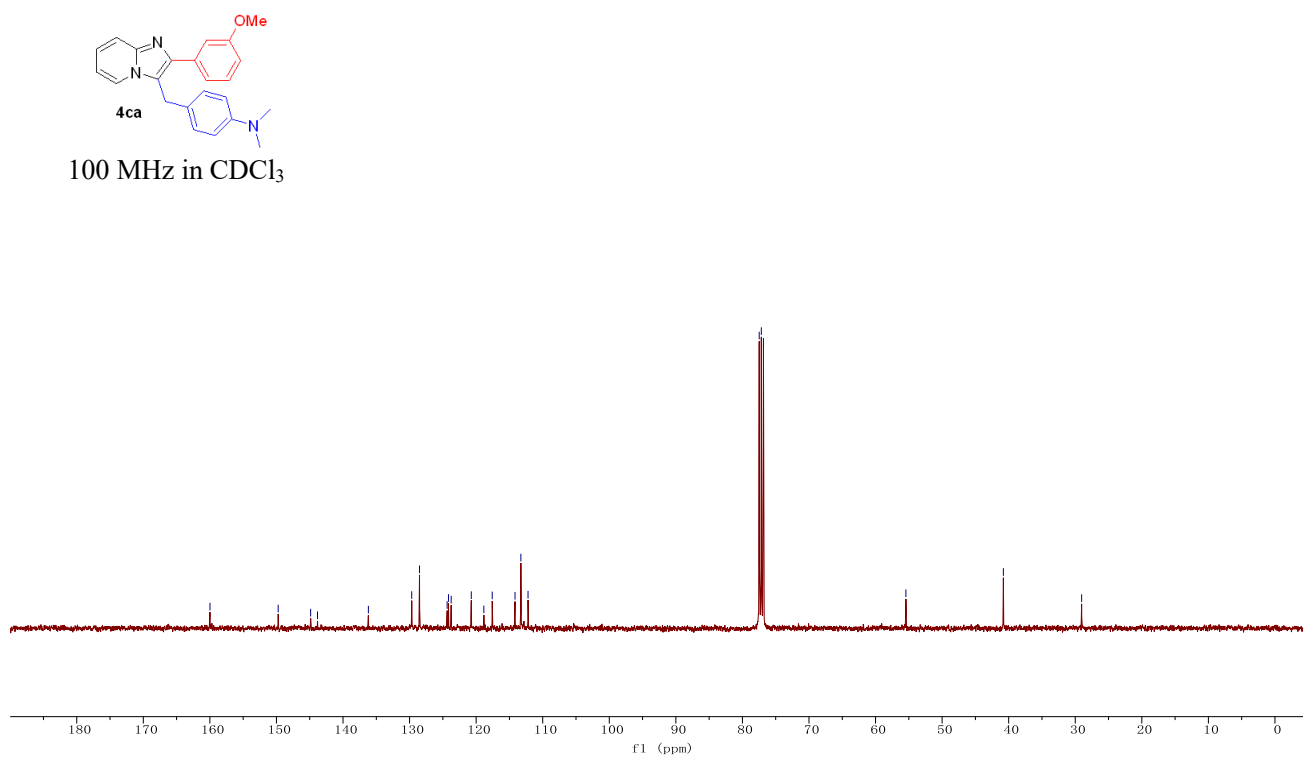
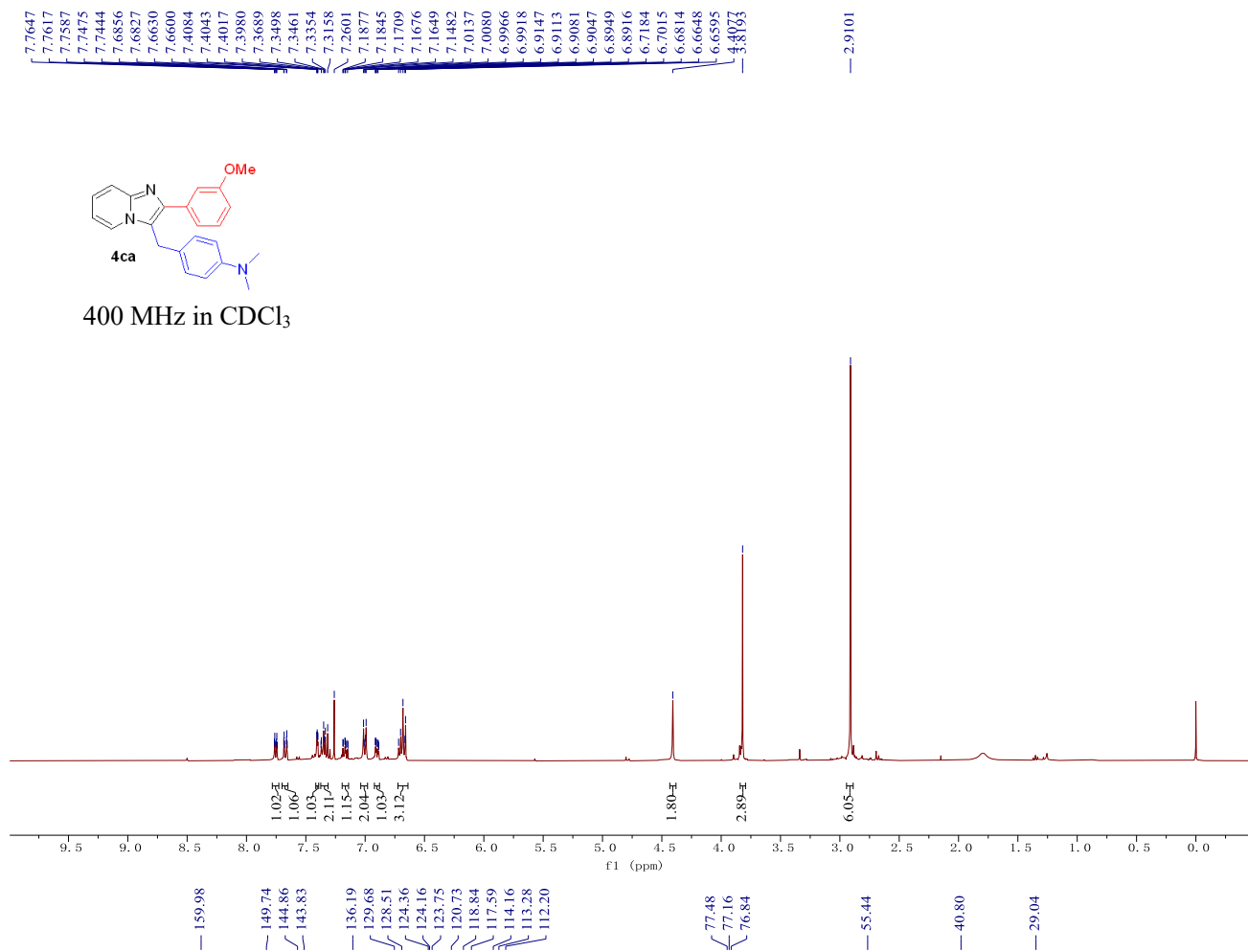


100 MHz in CDCl<sub>3</sub>



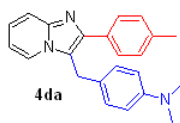




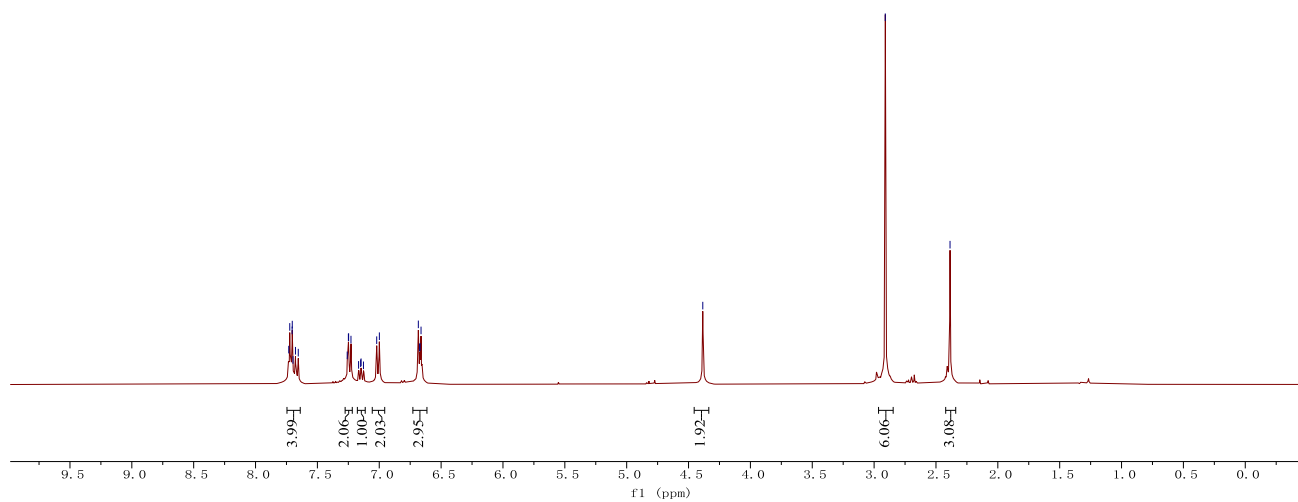




7.7315  
7.7235  
7.7042  
7.7031  
7.6975  
7.6772  
7.6546  
7.2590  
7.2495  
7.2476  
7.2286  
7.1670  
7.1504  
7.1469  
7.1277  
7.0206  
6.9990  
6.6841  
6.6738  
6.6705  
6.6623

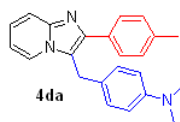


400 MHz in CDCl<sub>3</sub>

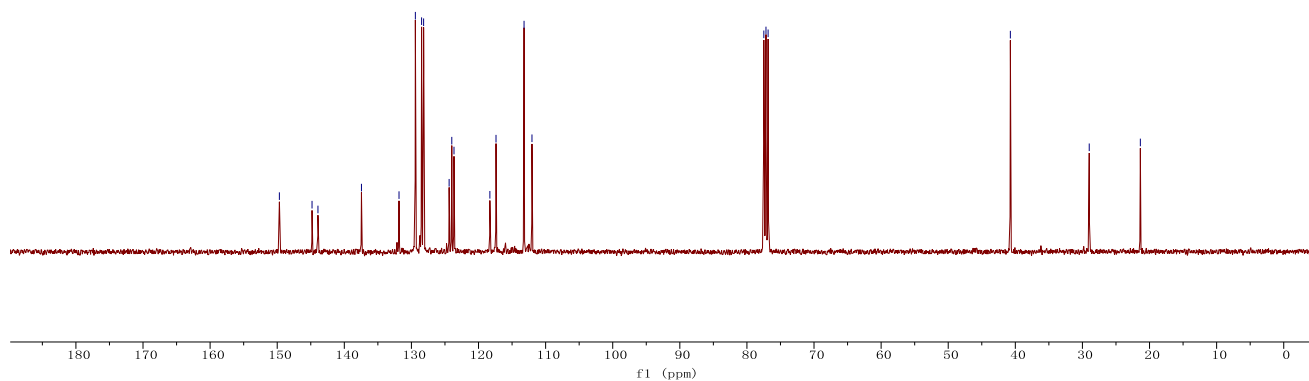


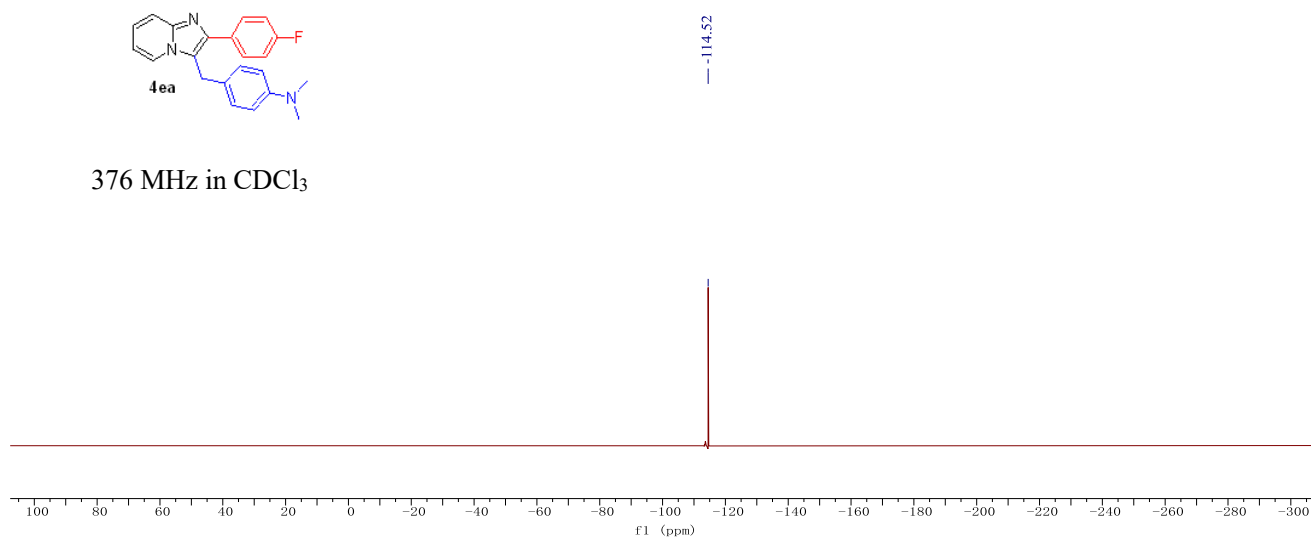
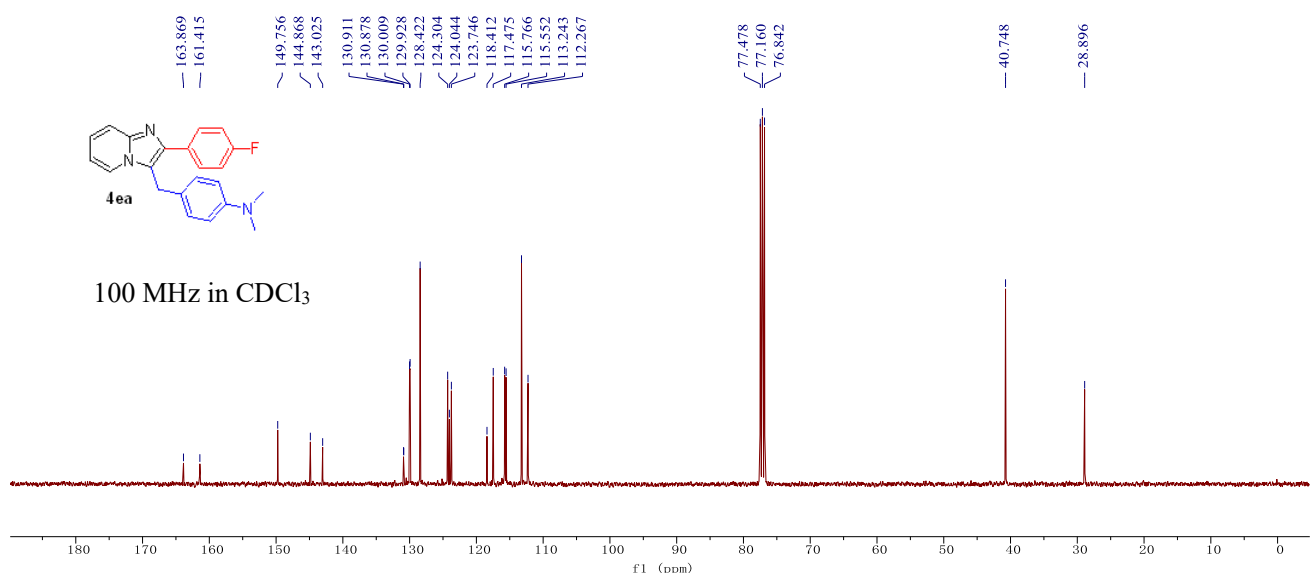
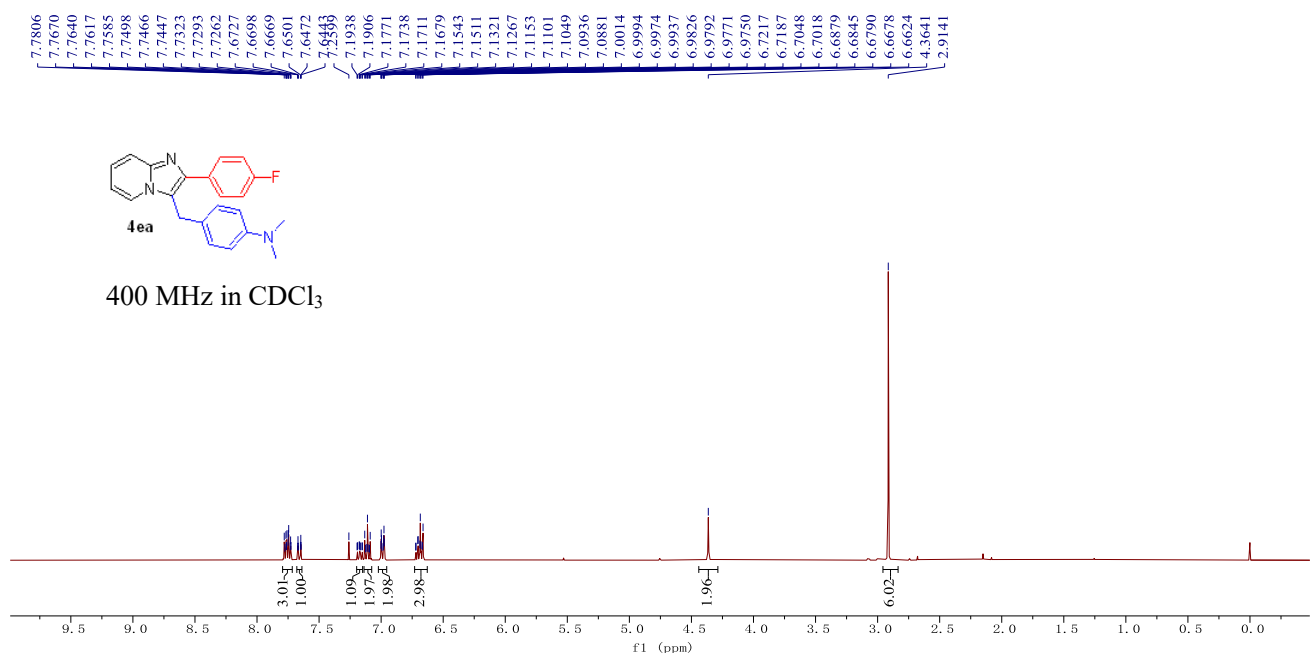
— 4.3849  
— 2.9098  
— 2.3863

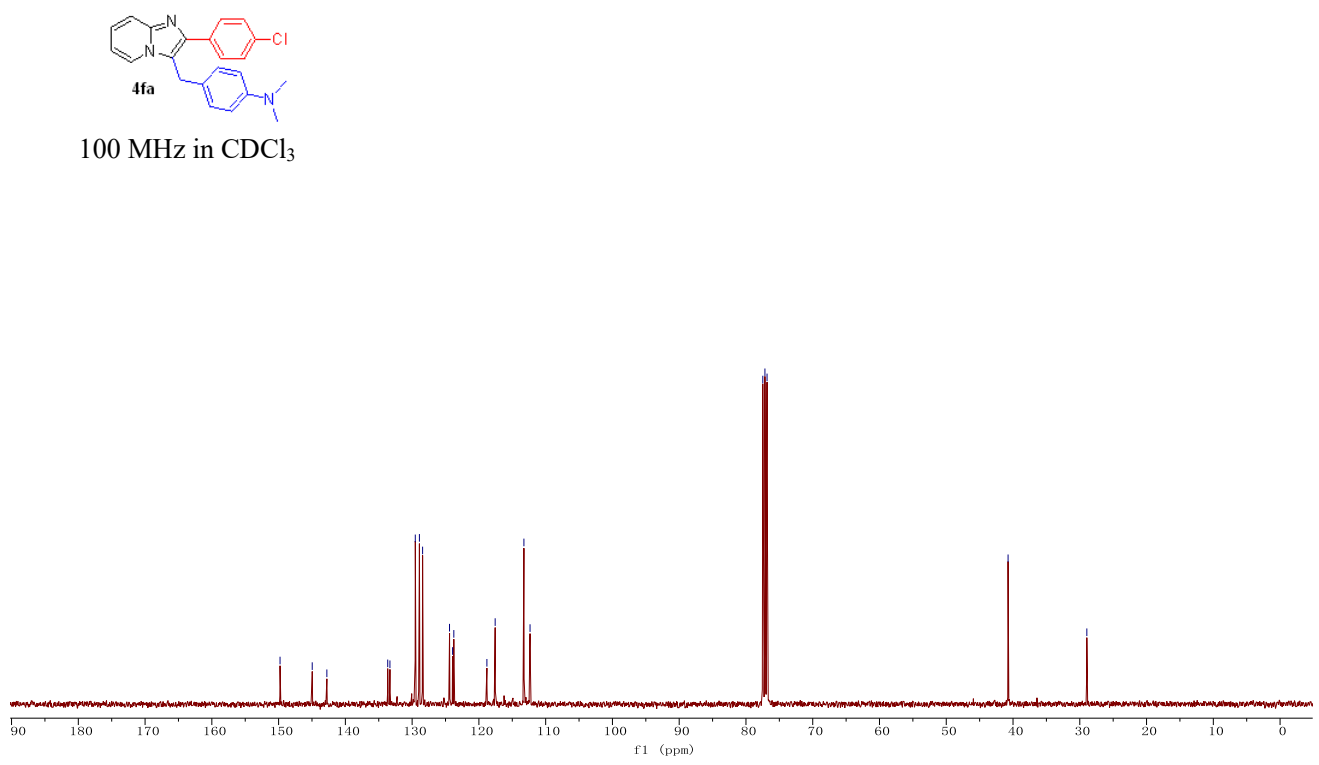
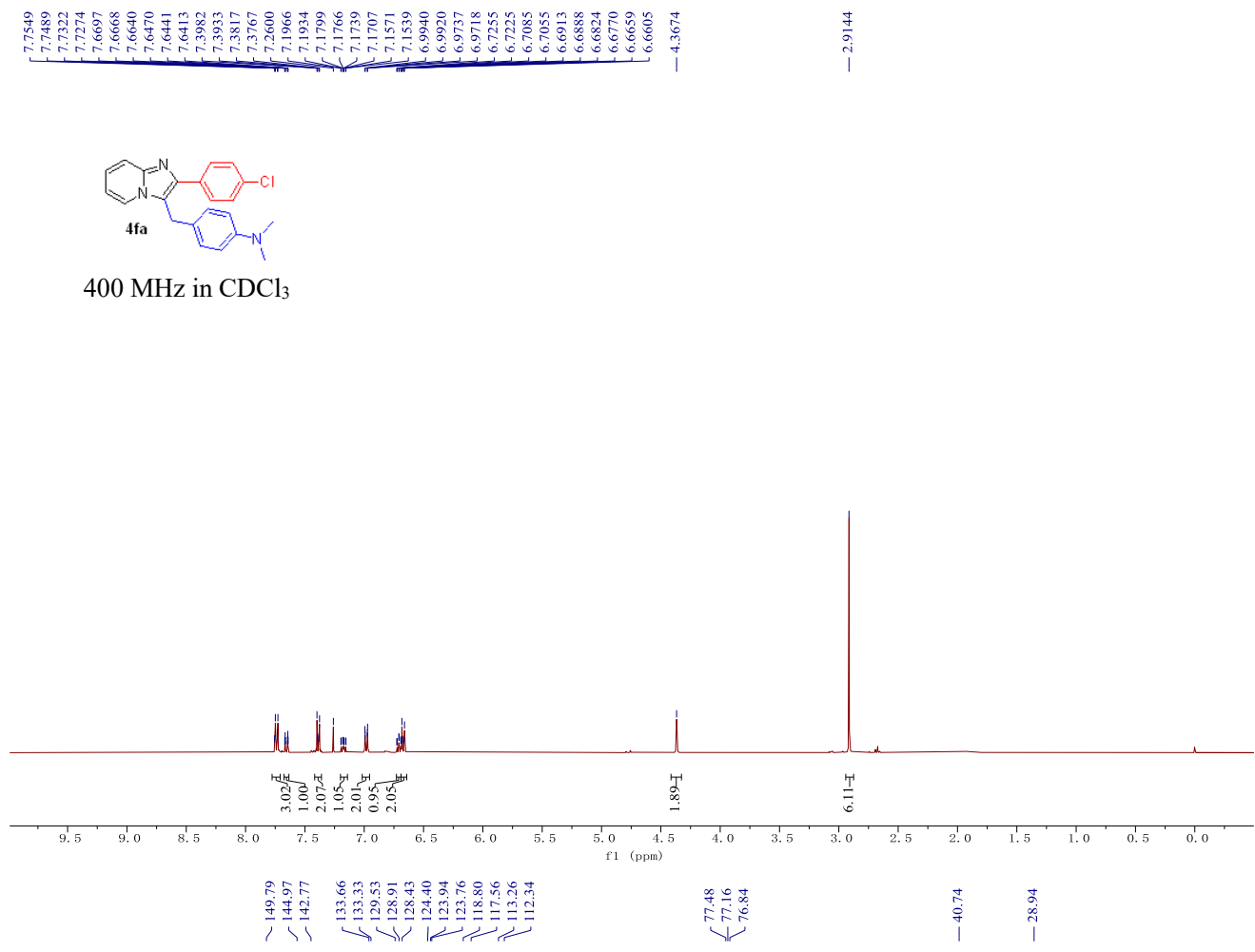
149.68  
144.81  
143.93  
137.45  
131.85  
129.41  
128.48  
128.18  
124.38  
123.98  
123.65  
118.31  
117.39  
113.21  
112.03



100 MHz in CDCl<sub>3</sub>

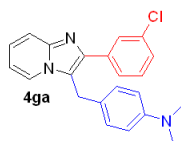




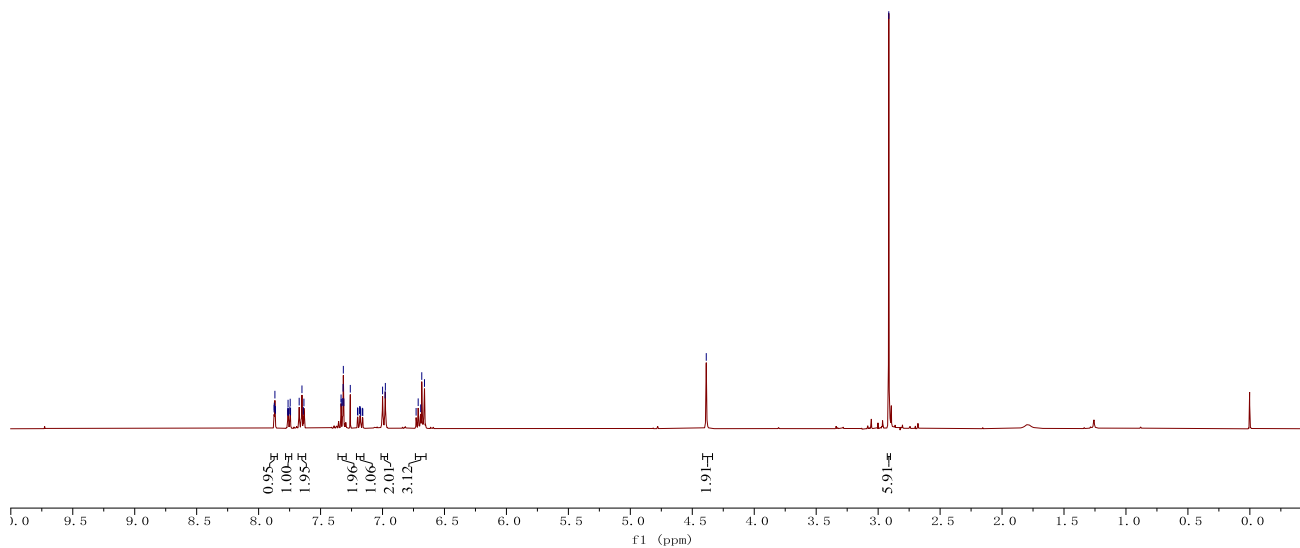


7.8751  
7.8736  
7.8686  
7.8644  
7.7654  
7.7624  
7.7593  
7.7479  
7.7451  
7.7423  
7.6727  
7.6503  
7.6396  
7.6345  
7.3347  
7.3331  
7.3217  
7.3182  
7.3163  
7.3119  
7.2601  
7.2020  
7.1988  
7.1852  
7.1818  
7.1791  
7.1760  
7.1625  
7.1593  
6.9996  
6.9793  
6.9773  
6.7291  
6.7122  
6.6920  
6.6833  
6.6613  
4.3872

2.9142



400 MHz in CDCl<sub>3</sub>

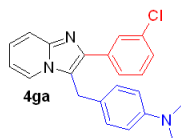


149.78  
144.96  
142.49  
136.71  
134.73  
129.91  
128.47  
128.40  
127.76  
126.30  
124.46  
123.93  
123.83  
119.18  
117.66  
113.27  
112.38

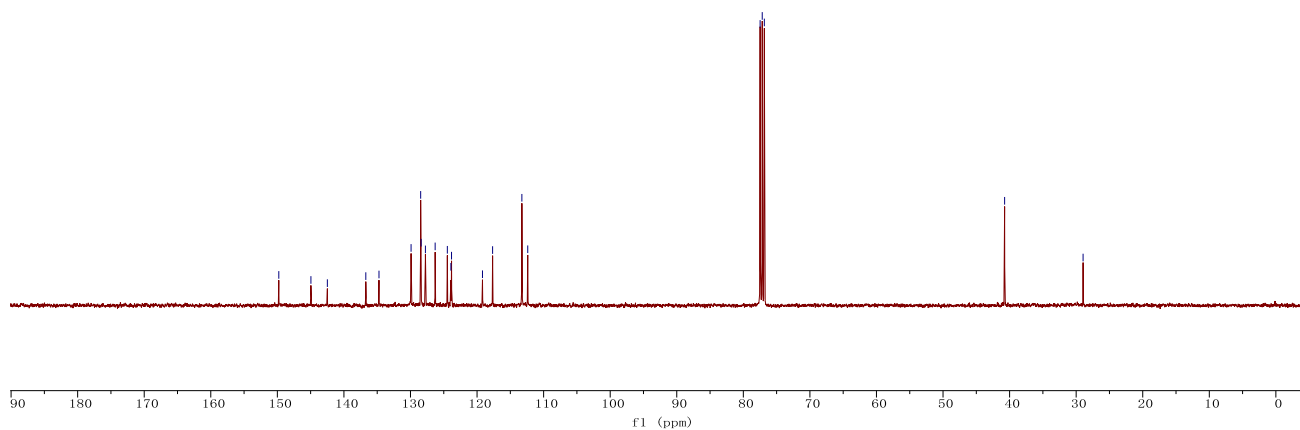
77.48  
77.16  
76.84

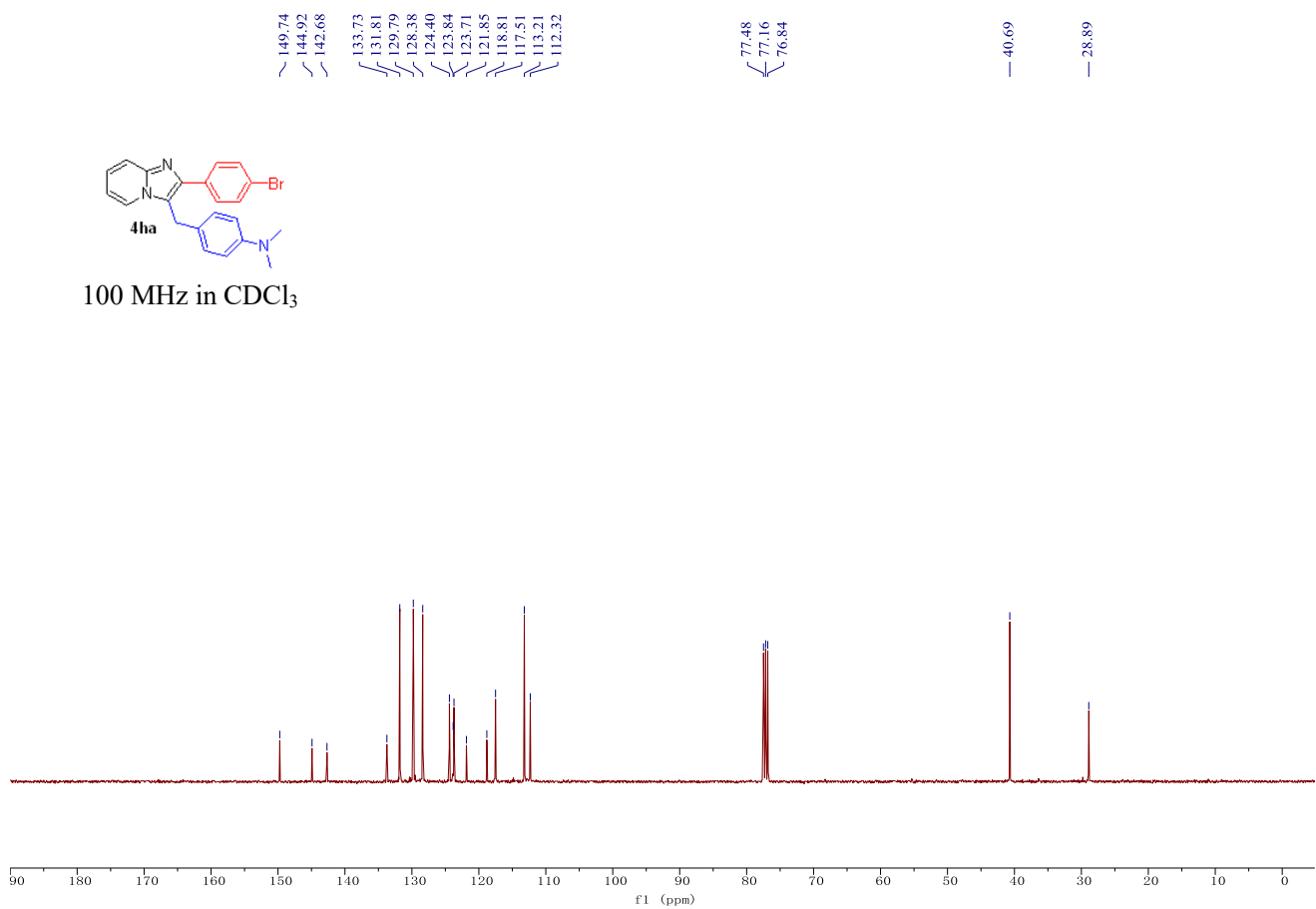
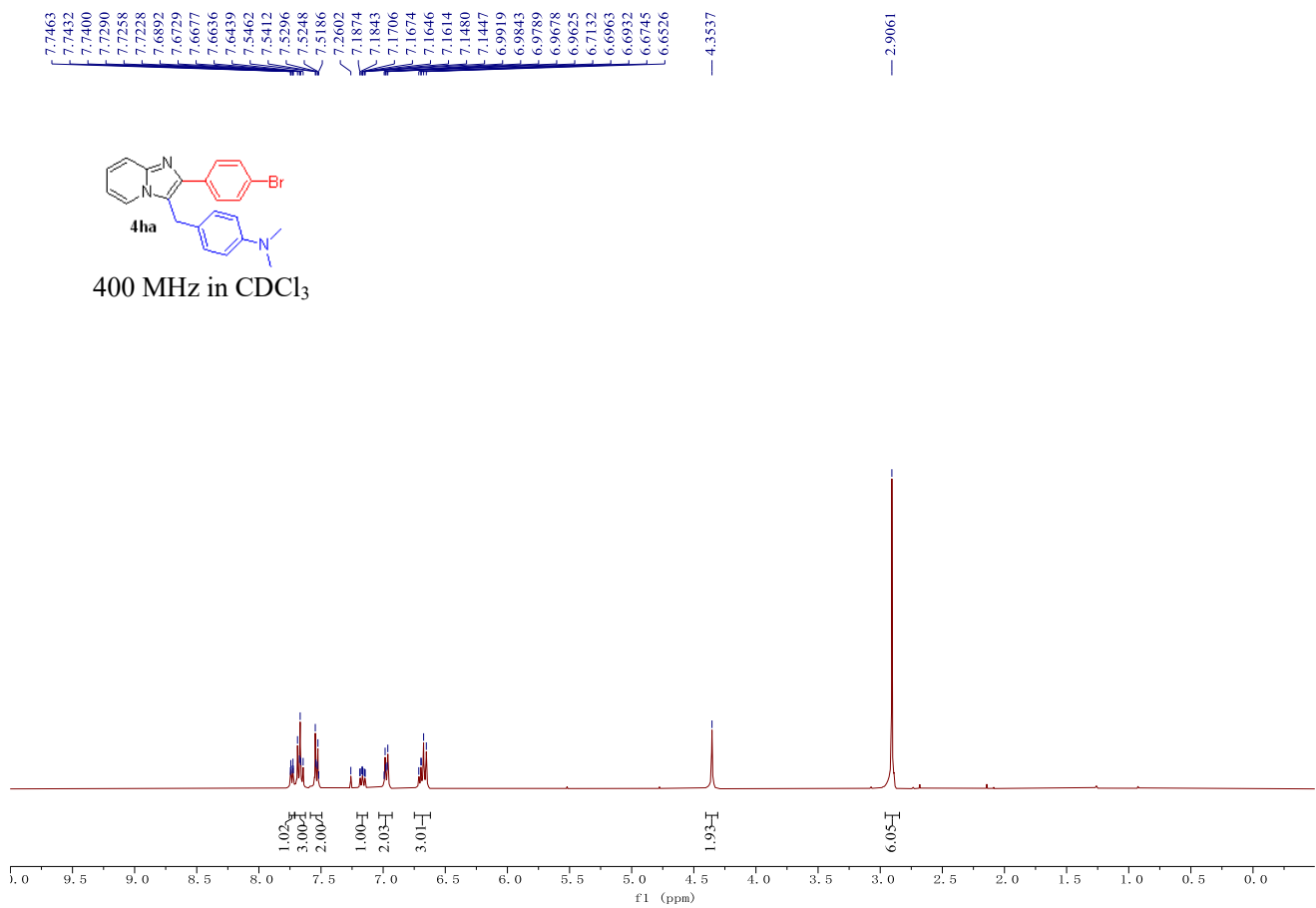
40.75

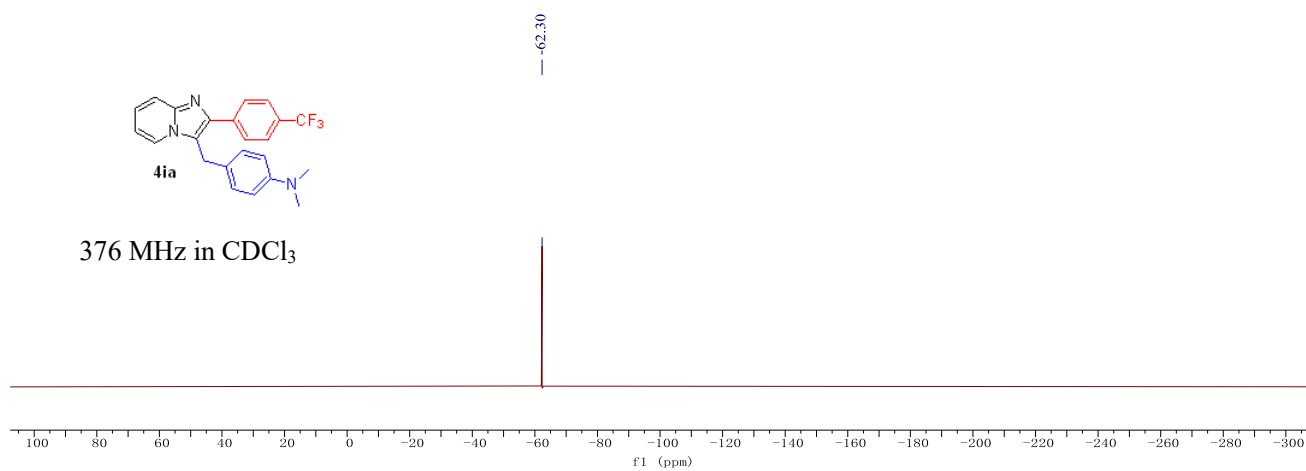
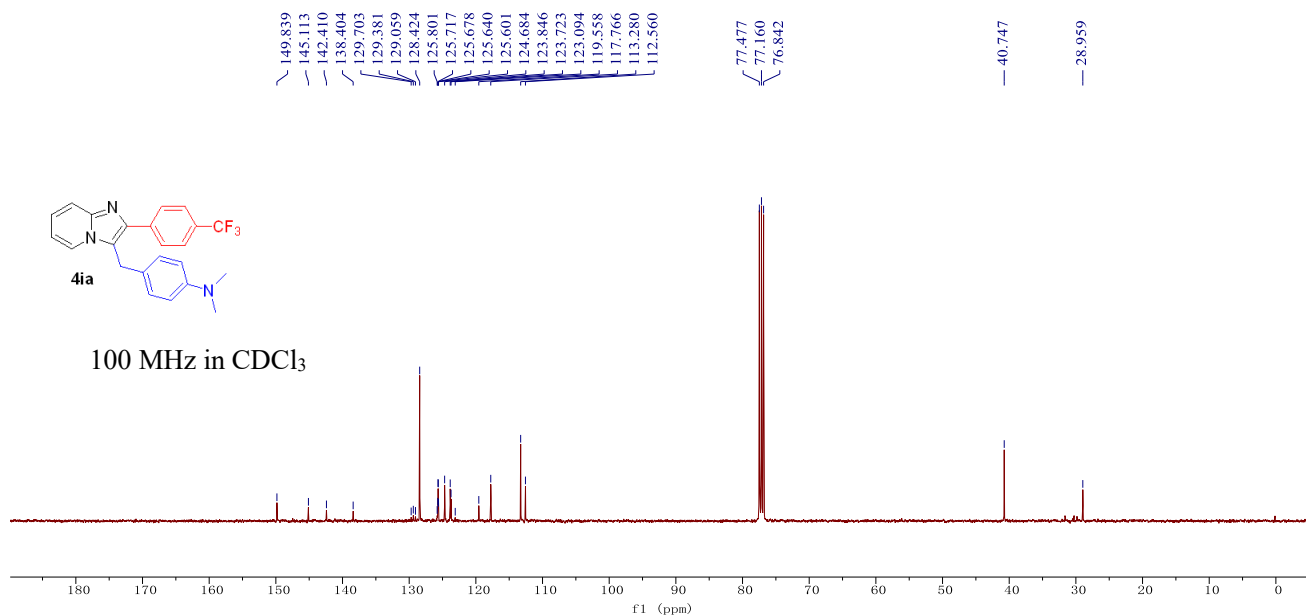
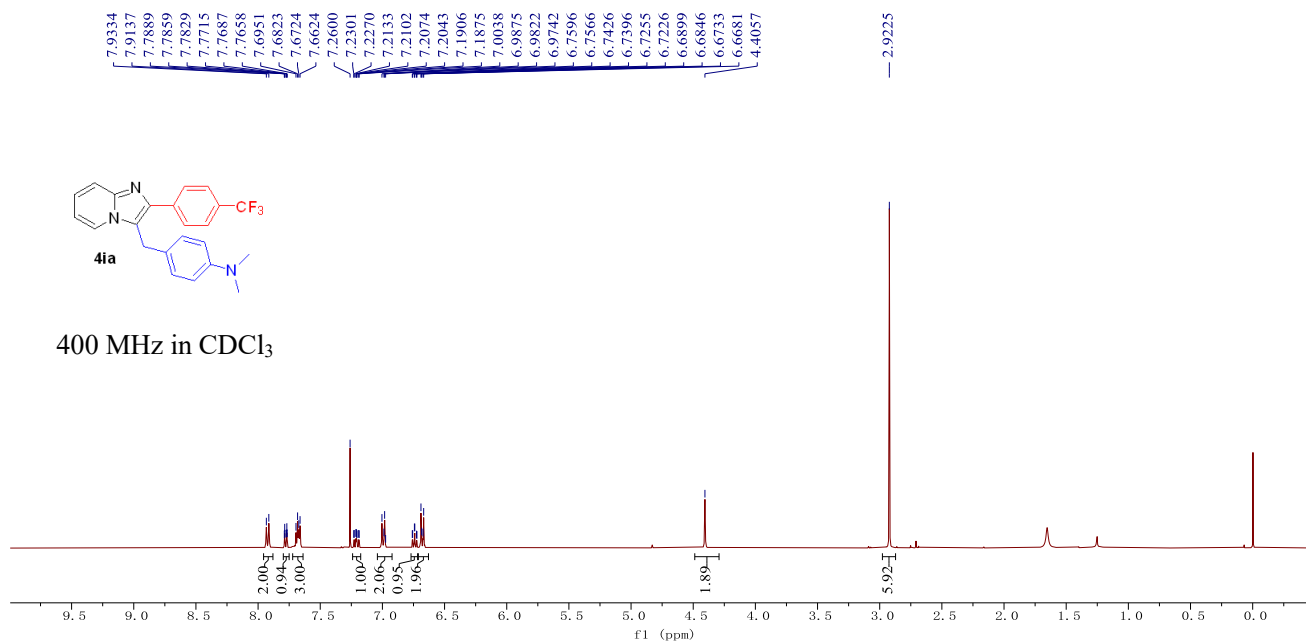
28.96

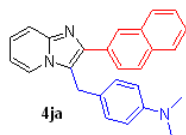


100 MHz in CDCl<sub>3</sub>

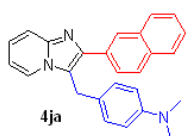
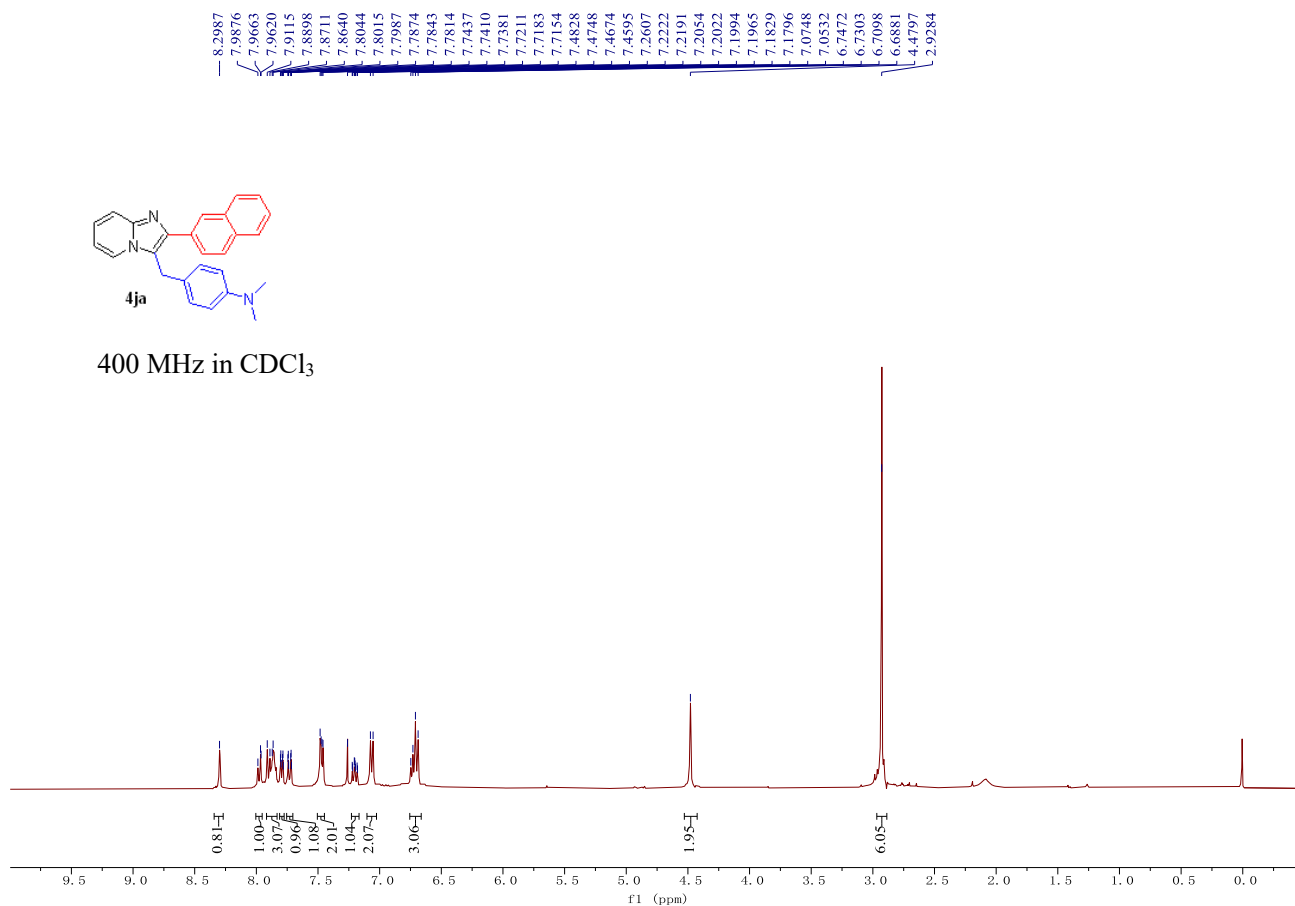




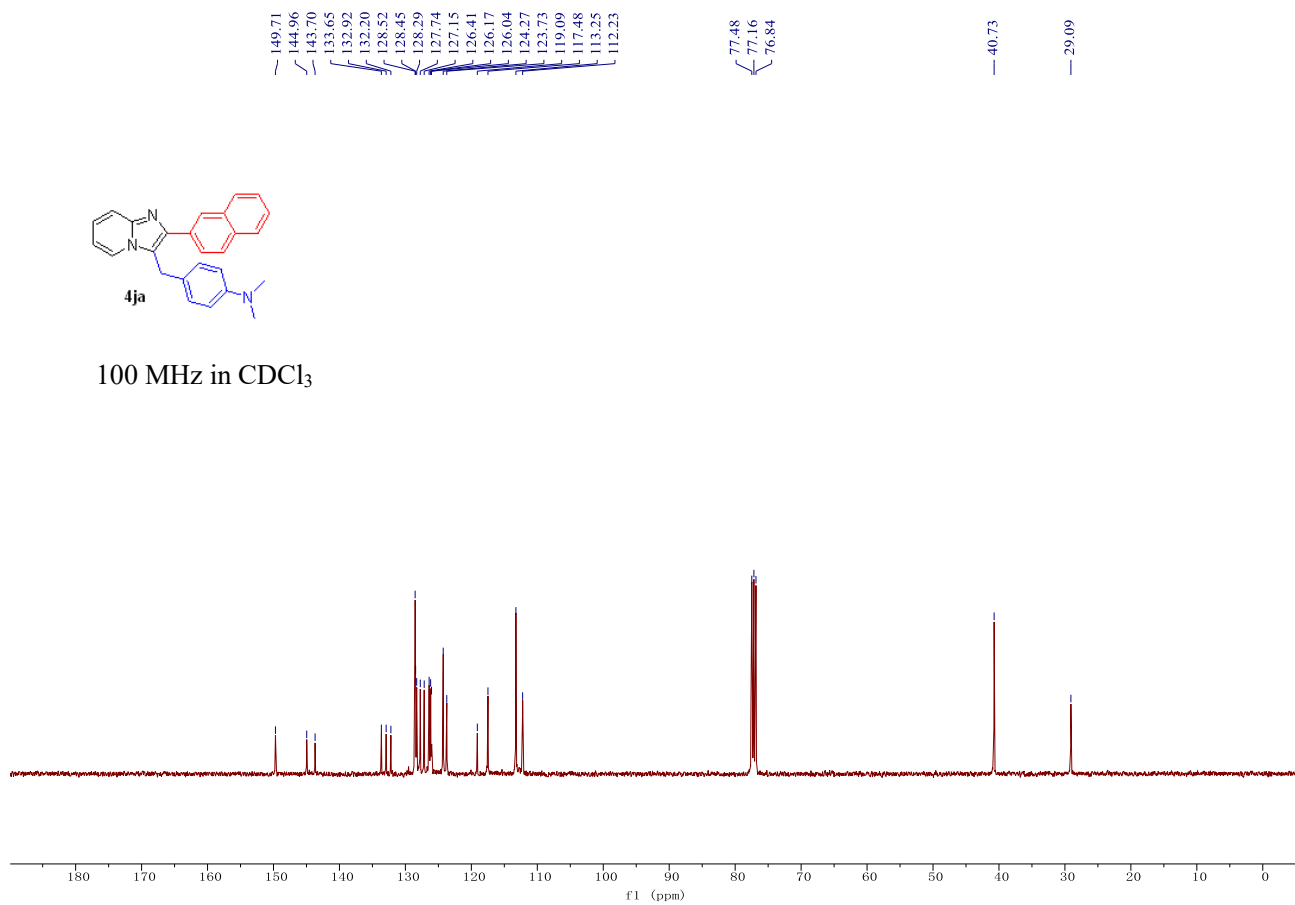




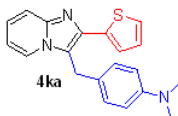
400 MHz in CDCl<sub>3</sub>



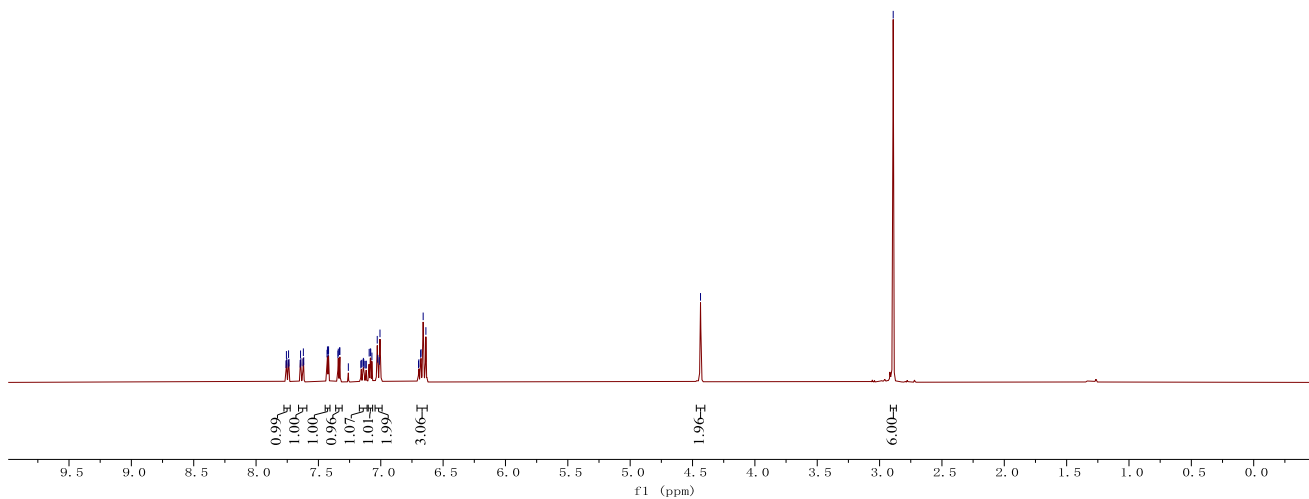
100 MHz in CDCl<sub>3</sub>



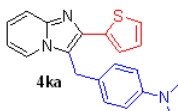
7.7596  
7.7566  
7.7536  
7.7424  
7.7393  
7.7362  
7.6461  
7.6433  
7.6406  
7.6235  
7.6206  
7.6178  
7.4304  
7.4276  
7.4215  
7.4186  
7.3434  
7.3406  
7.3306  
7.3278  
7.2600  
7.1593  
7.1561  
7.1426  
7.1395  
7.1367  
7.1333  
7.1198  
7.1165  
7.0930  
7.0841  
7.0804  
7.0711  
7.0282  
7.0240  
7.0062  
6.6973  
6.6943  
6.6802  
6.6771  
6.6600  
6.6381  
4.4360  
2.8908



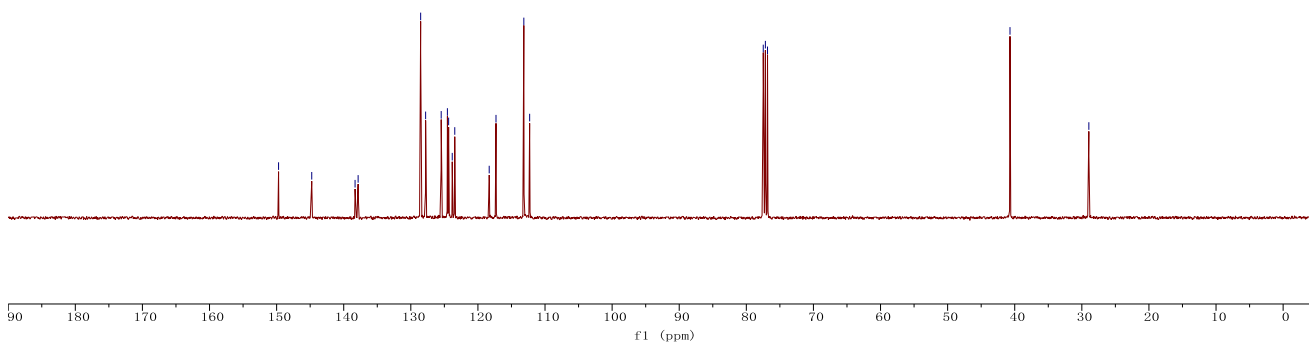
400 MHz in CDCl<sub>3</sub>



149.71  
144.77  
138.31  
137.85  
128.53  
127.79  
125.47  
124.55  
124.37  
123.83  
123.45  
118.32  
117.31  
113.16  
112.29  
77.48  
77.16  
76.84  
40.70  
28.95

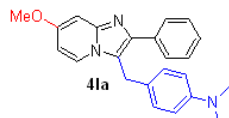


100 MHz in CDCl<sub>3</sub>

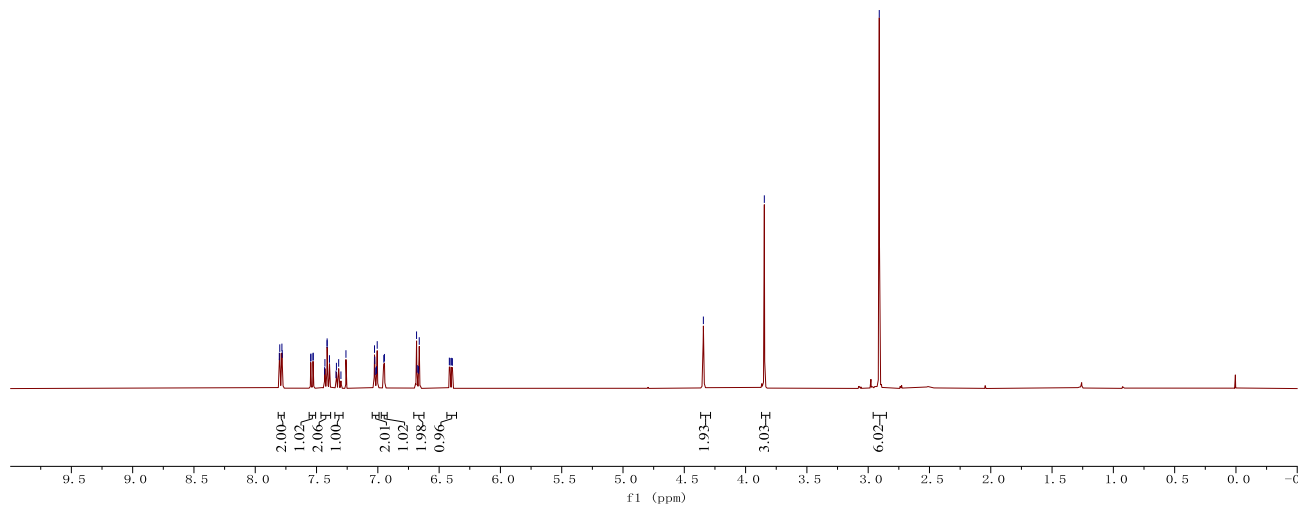




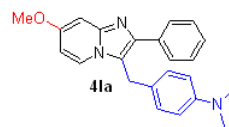
7.8037  
7.8000  
7.7825  
7.7798  
7.5475  
7.5458  
7.5288  
7.5271  
7.4340  
7.4317  
7.4138  
7.4136  
7.3946  
7.3412  
7.3378  
7.3194  
7.3010  
7.2600  
7.2071  
7.0251  
7.0216  
7.0105  
7.0051  
6.9512  
6.9463  
6.6884  
6.6786  
6.6675  
6.6622  
6.4167  
6.4105  
6.3981  
6.3919  
— 4.3443  
— 3.8472  
— 2.9096



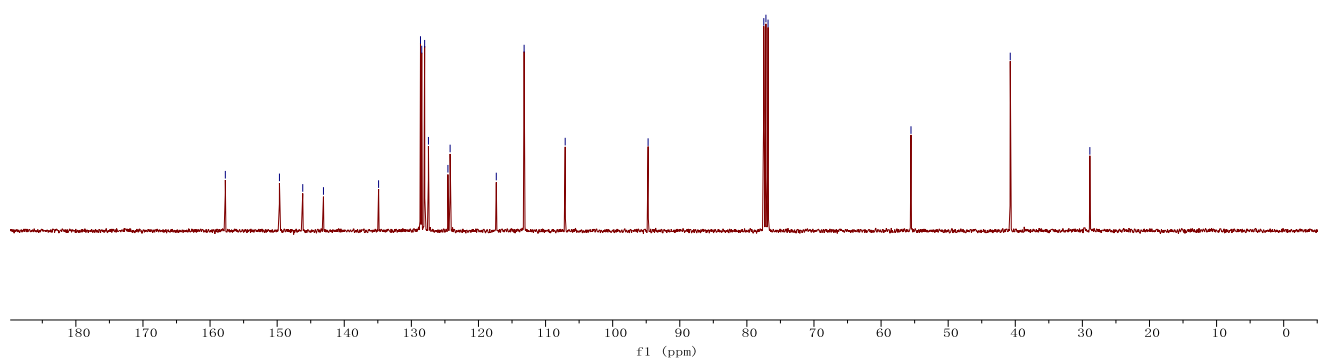
400 MHz in CDCl<sub>3</sub>



157.73  
149.66  
146.19  
143.10  
134.89  
128.64  
128.45  
128.02  
127.46  
124.55  
124.22  
117.35  
113.20  
107.08  
— 94.72  
77.48  
77.16  
76.84  
55.54  
40.75  
28.89



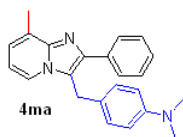
100 MHz in CDCl<sub>3</sub>



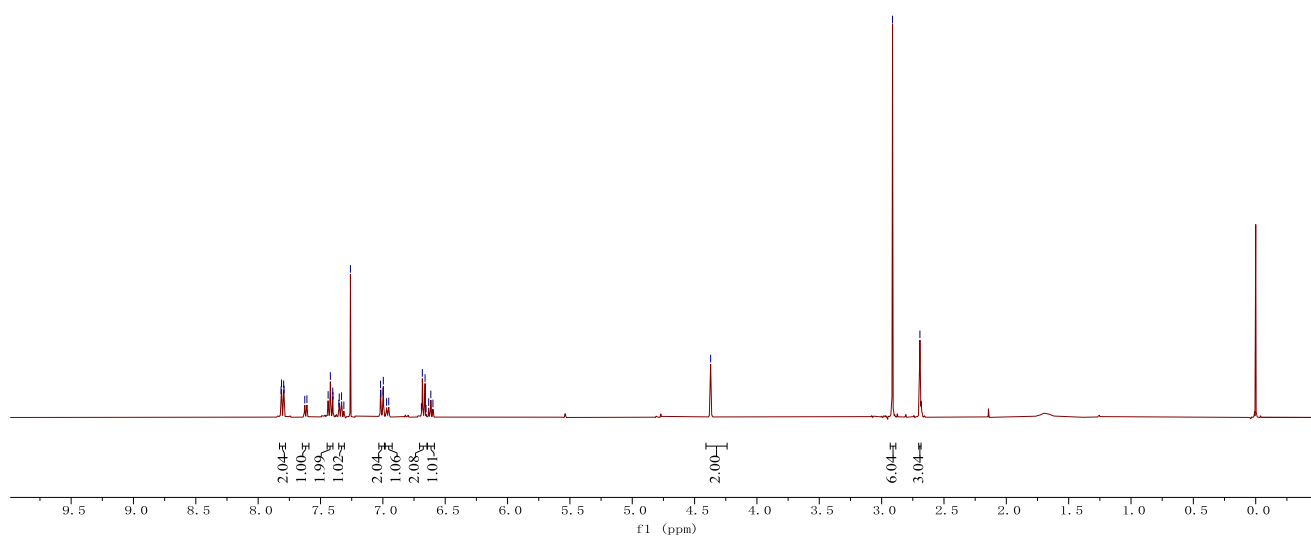
7.8166  
7.8128  
7.7956  
7.7924  
7.6263  
7.6091  
7.4390  
7.4209  
7.4021  
7.4003  
7.3529  
7.3498  
7.3314  
7.3128  
7.2600  
7.0185  
7.0165  
6.9983  
6.9901  
6.9701  
6.9530  
6.6909  
6.6833  
6.6614  
6.6538  
6.6324  
6.6154  
6.5984

— 4.3715

— 2.9127  
— 2.6930



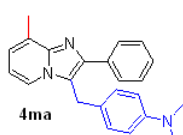
400 MHz in CDCl<sub>3</sub>



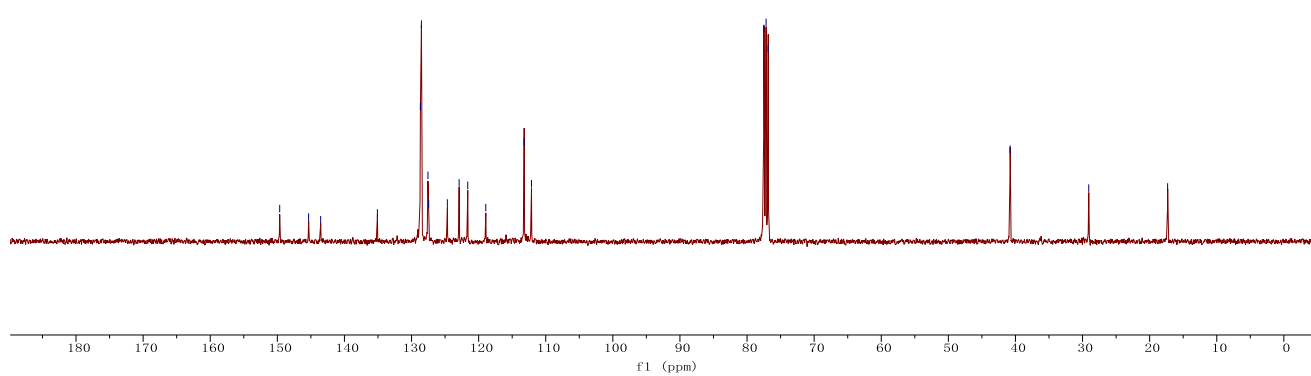
149.65  
145.34  
143.54  
135.09  
128.68  
128.53  
127.55  
127.45  
124.66  
122.91  
121.63  
118.94  
113.22  
112.13

77.48  
77.16  
76.85

— 40.79  
— 29.08  
— 17.32



100 MHz in CDCl<sub>3</sub>

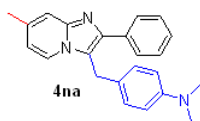


7.8161  
7.8124  
7.7951  
7.7922  
7.6118  
7.5944  
7.4361  
7.4279  
7.4182  
7.3986  
7.3486  
7.3456  
7.3270  
7.3085  
7.2605  
7.0144  
7.0093  
6.9982  
6.6818  
6.6764  
6.6655  
6.6601  
6.5190  
6.5148  
6.5016  
6.4974

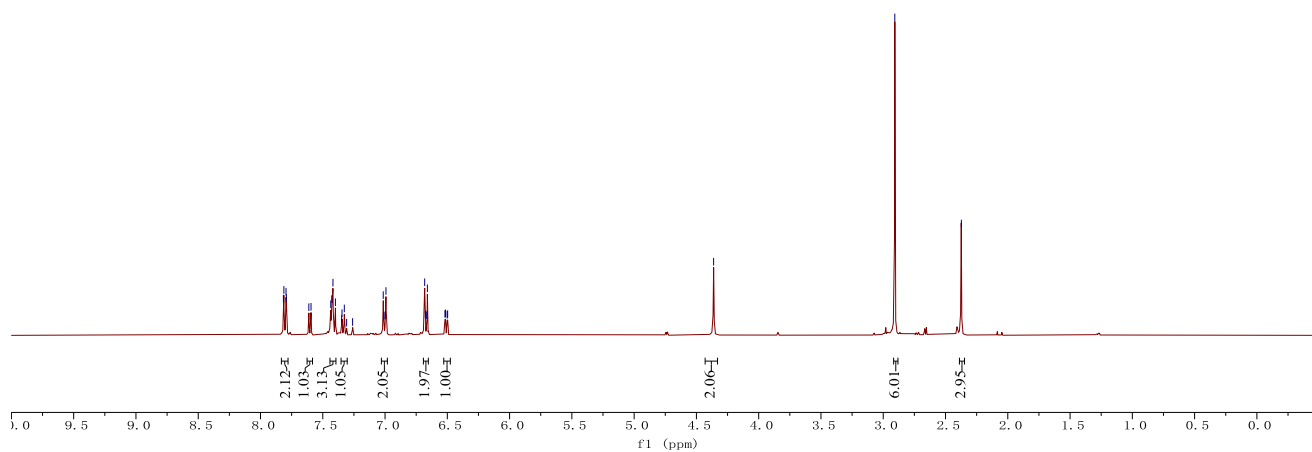
— 4.3619

— 2.9070

— 2.3727



400 MHz in CDCl<sub>3</sub>



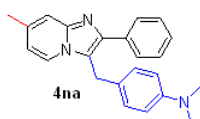
149.65  
145.28  
143.44  
134.99  
134.90  
128.62  
128.44  
128.22  
127.52  
124.52  
122.94  
117.94  
115.86  
114.72  
113.19

77.48  
77.16  
76.84

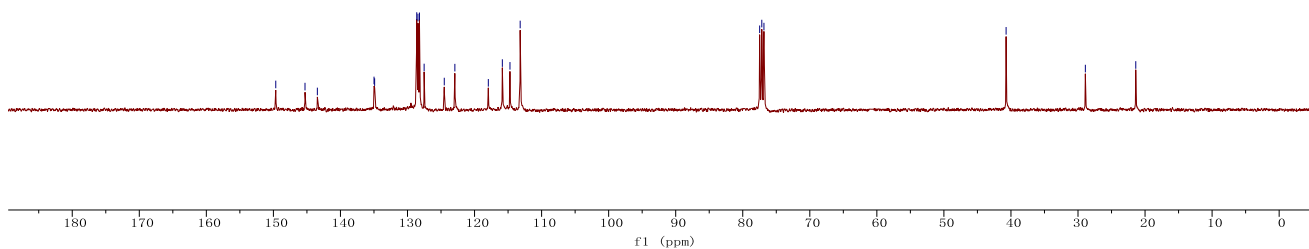
— 40.72

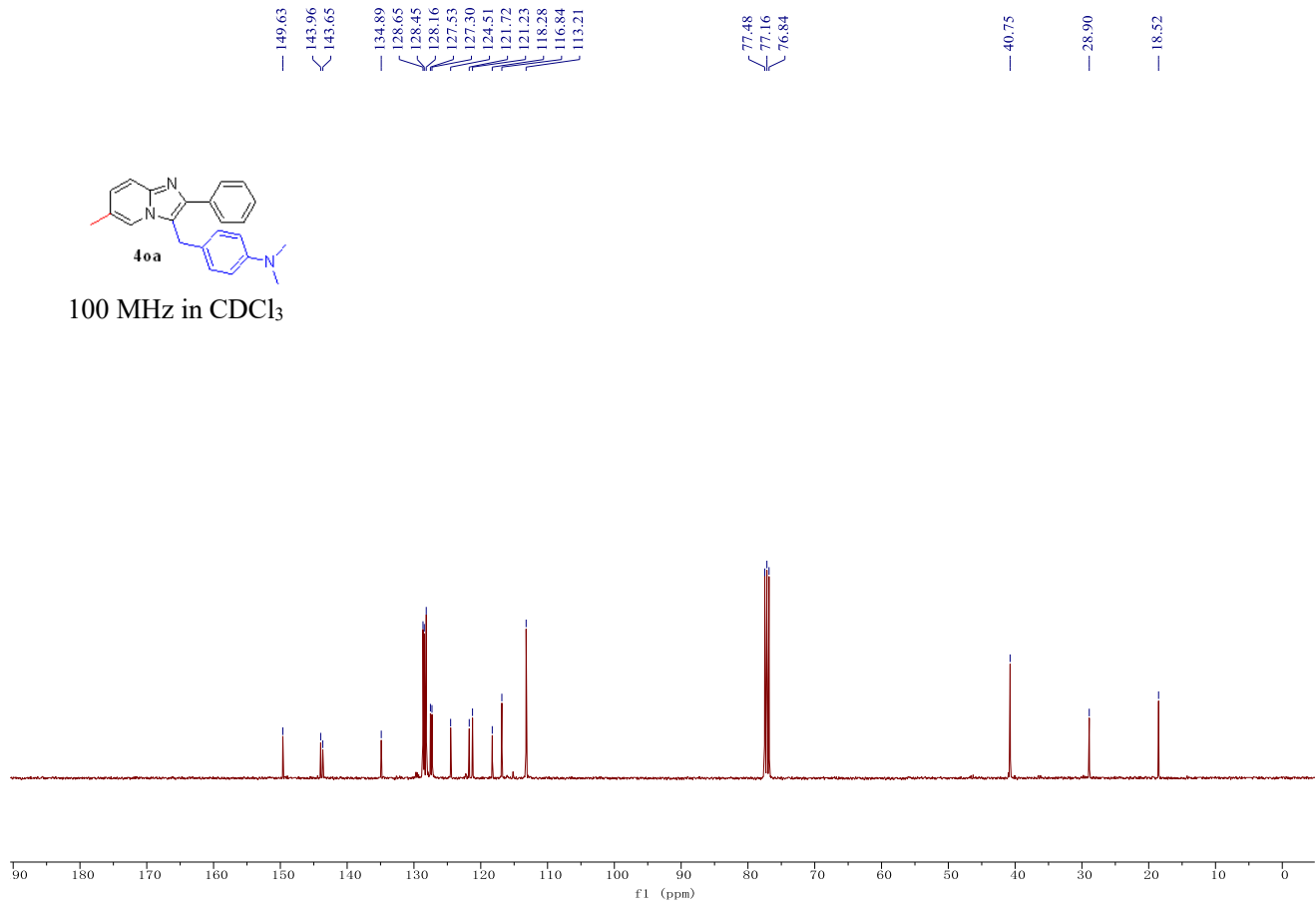
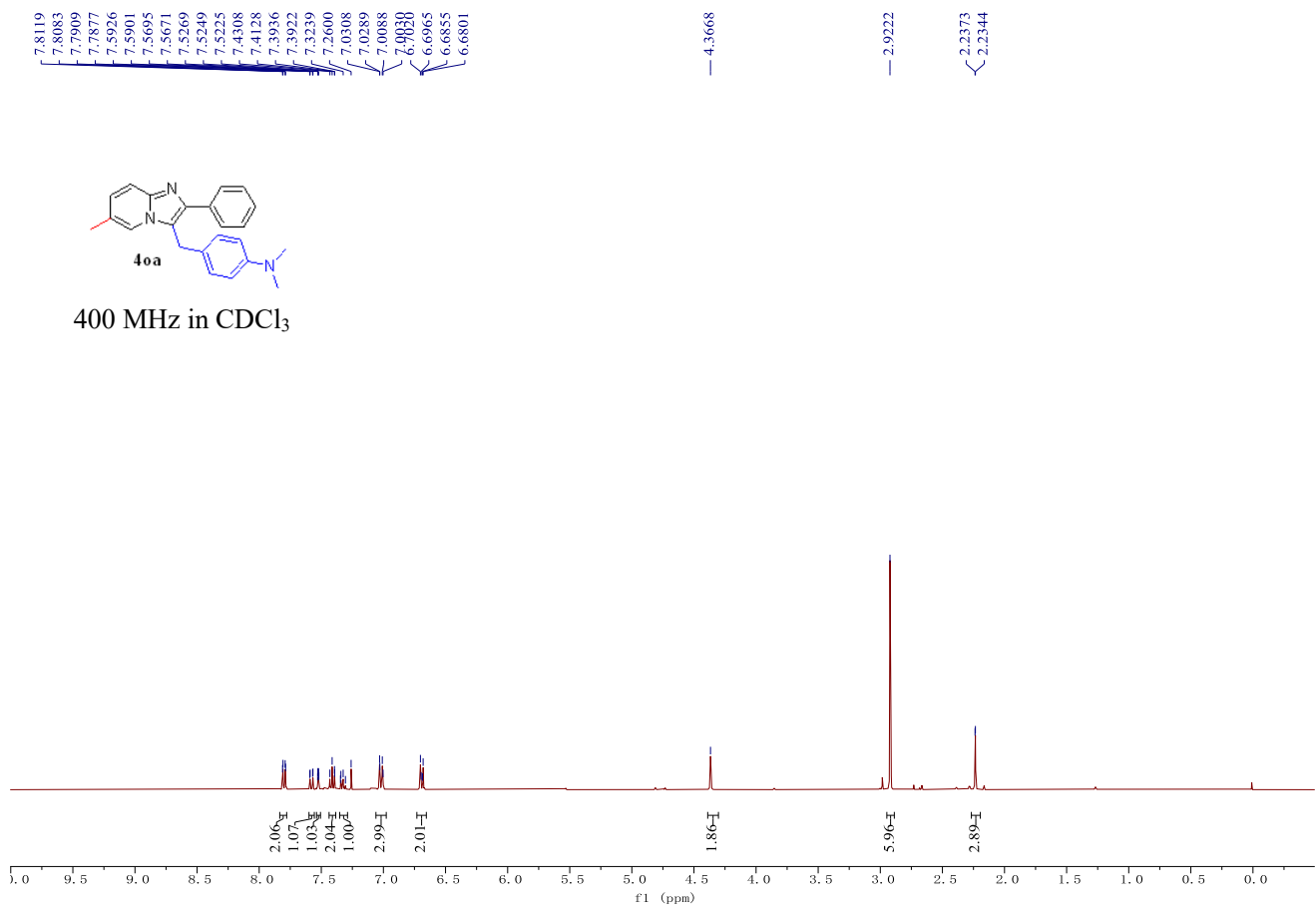
— 28.90

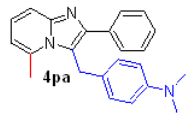
— 21.38



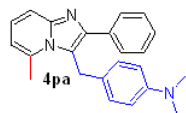
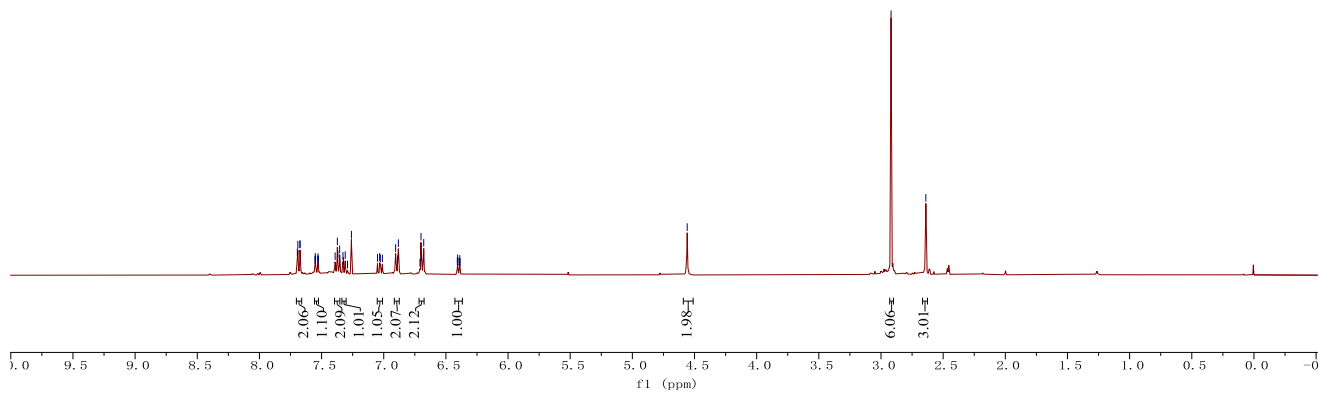
100 MHz in CDCl<sub>3</sub>



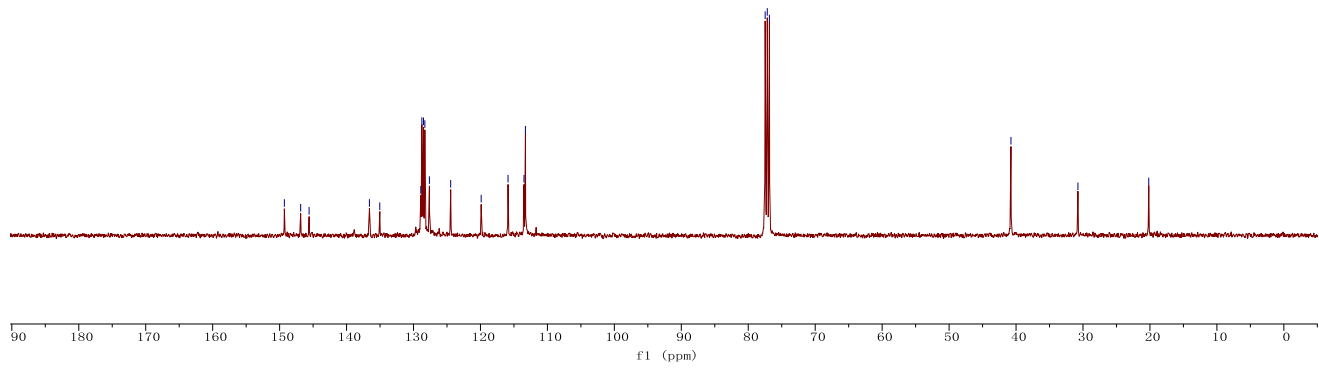


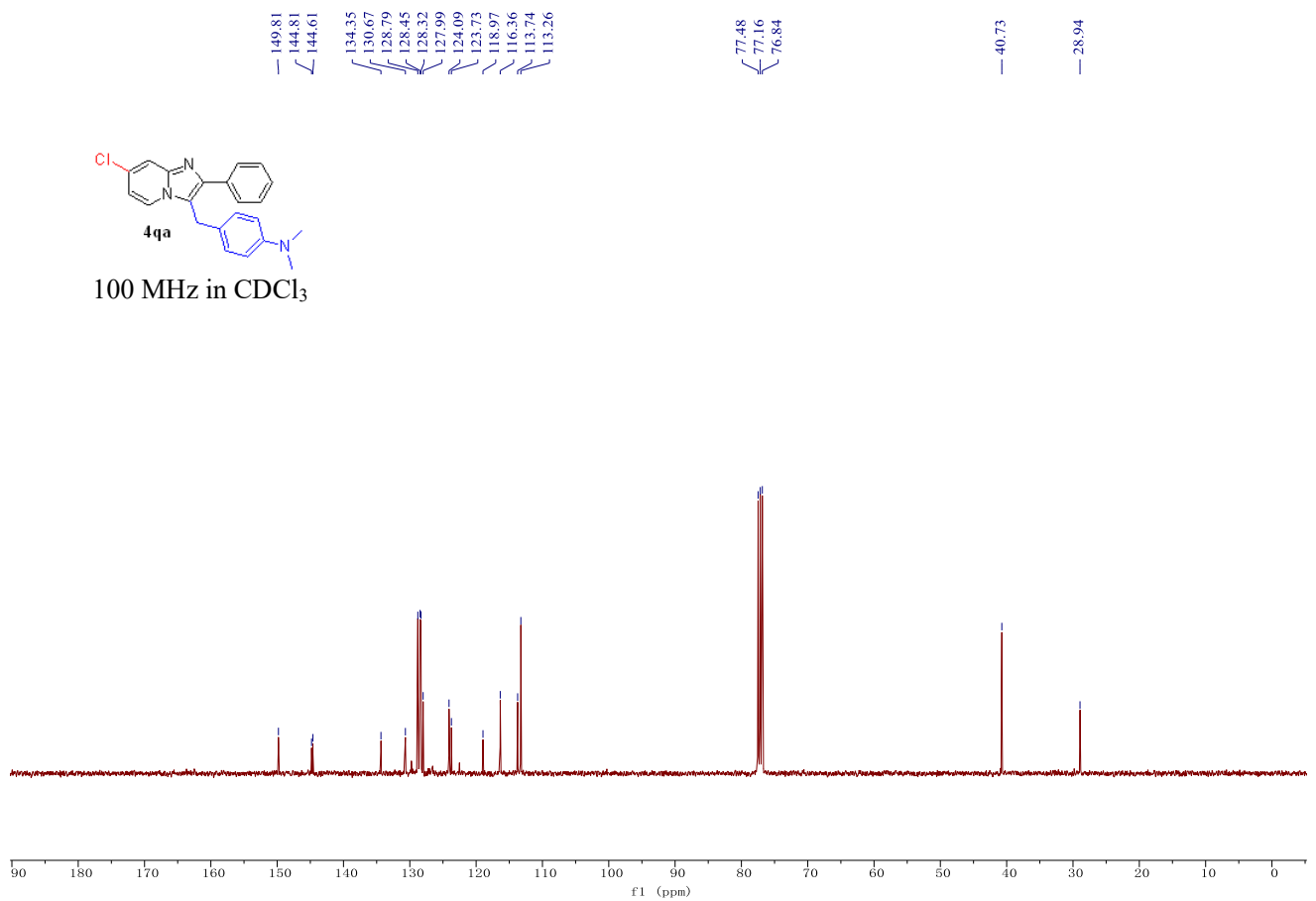
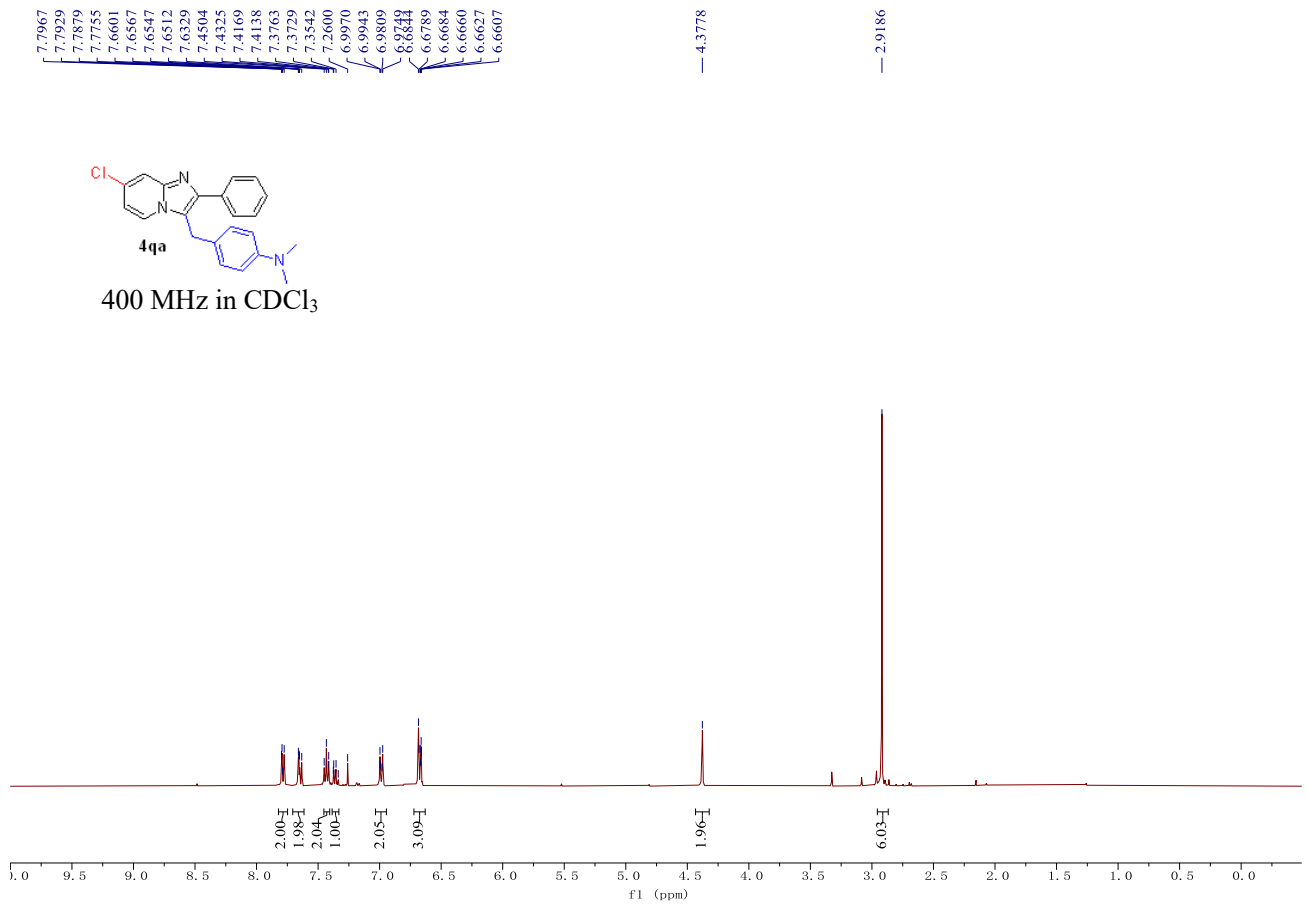


400 MHz in CDCl<sub>3</sub>



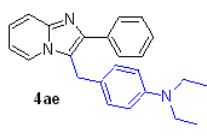
100 MHz in CDCl<sub>3</sub>



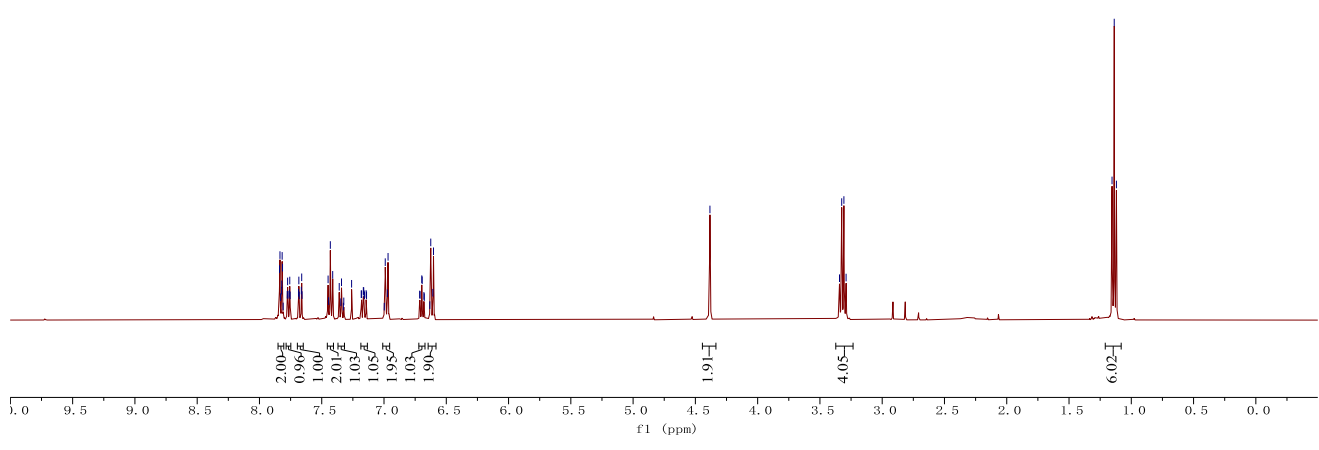


7.8391  
7.8353  
7.8221  
7.8206  
7.8178  
7.8148  
7.7769  
7.7739  
7.7710  
7.7596  
7.7567  
7.7538  
7.6866  
7.6838  
7.6809  
7.6638  
7.6609  
7.6581  
7.4490  
7.4310  
7.4121  
7.3596  
7.3413  
7.2602  
7.1829  
7.1797  
7.1661  
7.1629  
7.1601  
7.1570  
7.1435  
7.1403  
6.9900  
6.9732  
6.9676  
6.7136  
6.7105  
6.6967  
6.6936  
6.6796  
6.6246  
6.6079  
6.6027  
3.3338  
3.3328  
3.3251  
3.3074  
3.2899

1.1546  
1.1371  
1.1194



400 MHz in CDCl<sub>3</sub>



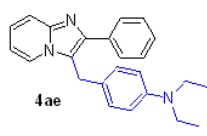
146.87  
144.87  
143.85  
134.80  
128.69  
128.34  
127.68  
124.09  
123.84  
122.91  
118.74  
117.48  
112.34  
112.10

77.48  
77.16  
76.84

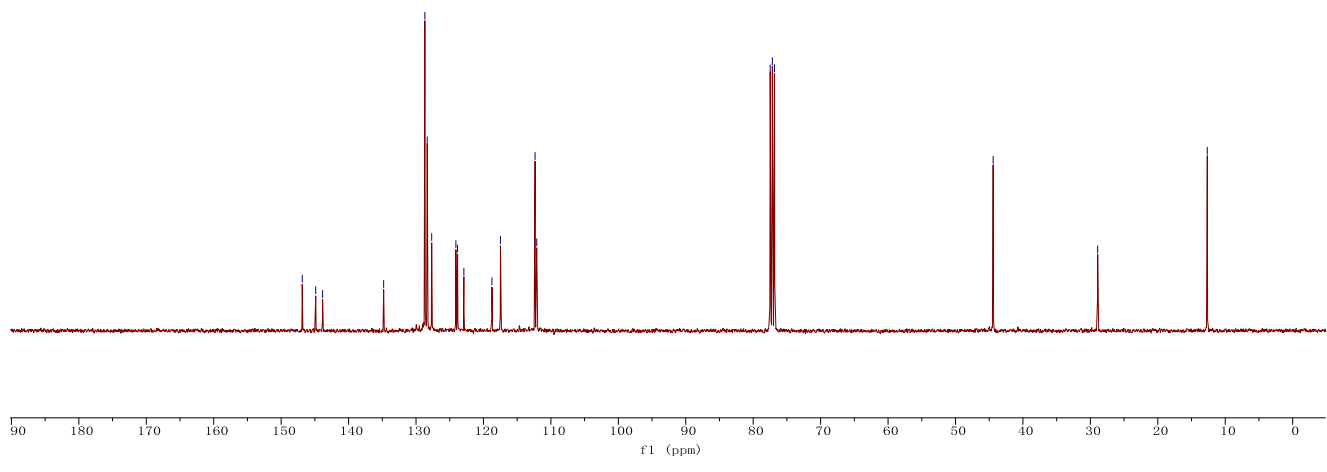
44.42

28.92

12.67



100 MHz in CDCl<sub>3</sub>



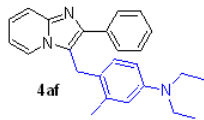
7.7676  
7.7626  
7.7502  
7.7069  
7.6887  
7.6863  
7.6738  
7.4322  
7.4142  
7.4106  
7.3949  
7.3478  
7.3295  
7.2033  
7.2006  
7.1865  
7.1831  
7.1800  
7.1773  
7.1639  
7.1596  
6.7269  
6.7129  
6.7101  
6.6953  
6.6929  
6.6269  
6.6198  
6.5085  
6.4872  
6.3592  
6.3523  
6.3379  
6.3308

4.2711

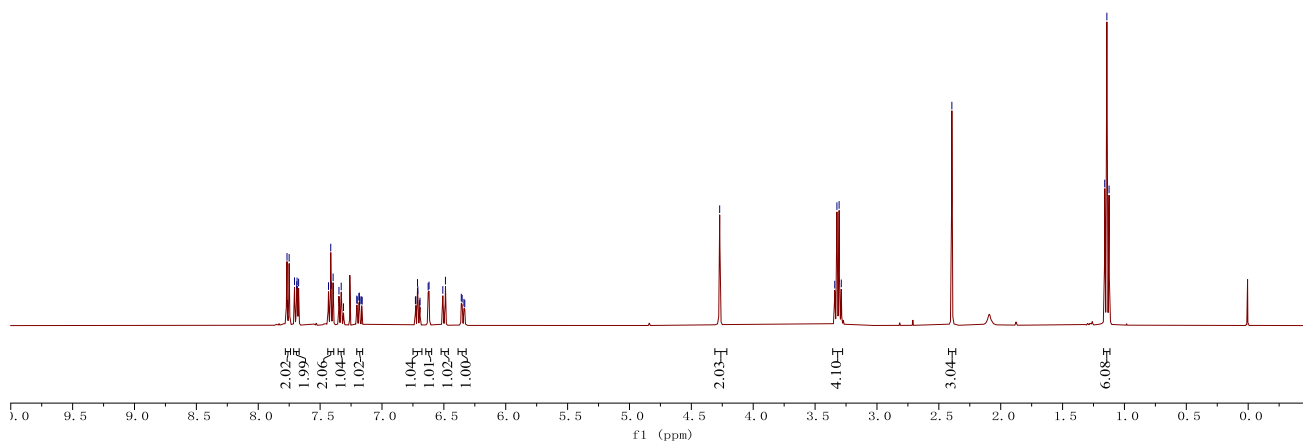
3.3412  
3.3237  
3.3059  
3.2883

2.3947

1.1595  
1.1420  
1.1242



400 MHz in CDCl<sub>3</sub>



146.89  
144.93  
144.12  
137.23  
134.76  
128.70  
128.29  
127.79  
127.66  
124.09  
123.81  
121.05  
118.46  
117.50  
114.07  
112.17  
109.77

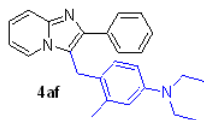
77.48  
77.16  
76.84

44.34

26.79

20.54

12.75



100 MHz in CDCl<sub>3</sub>

