Electronic Supplementary Material (ESI) for Green Chemistry.

Electrochemical-Induced Solvent-tuned Selective Transfer Hydrogenation of Imidazopyridines with Carbazates as Hydrogen Donor

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Contents

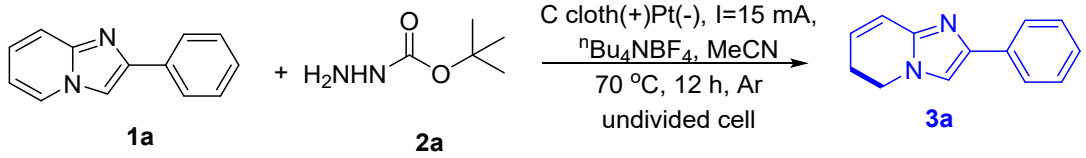
1. General Information

All reactions were carried out under Ar unless otherwise noted. Commercial reagents were used as received without additional purification unless otherwise noted. Substituted imidazopyridines were prepared according to the literature procedure.[1] Reactions were monitored by thin layer chromatography (TLC) using Silicycle glass-backed TLC plates with 250 μm silica and F254 indicator. Visualization was accomplished by UV light.

^{1}H NMR, ^{13}C NMR, and ^{19}F NMR spectra were recorded on a AM-500 Fourier transform NMR spectrometer at 400/600 MHz, 125/151 MHz, 376 MHz respectively. Chemical shifts are reported relative to the solvent resonance peak δ 2.50 (DMSO-d_6) for ^{1}H；δ 39.52 (DMSO-d_6) or 77.16 ($CDCl_3$) for ^{13}C. Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, b = broad singlet, m = multiplet), coupling constants, and number of protons. High resolution mass spectra were obtained using a VG autospec with an ionization mode of EI-TOF. Infrared spectra are reported in cm^{-1}. Column chromatography was performed with silica gel (50-63 μm mesh particle size).

2. Optimization of The Reaction Conditions

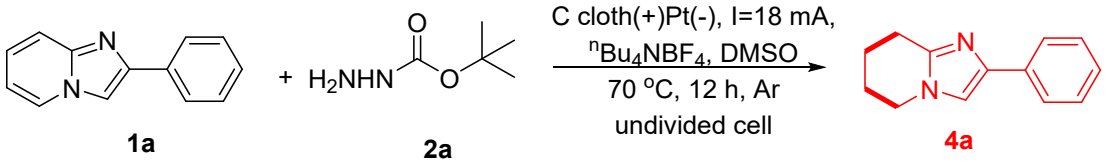
Table S1. Optimization of Transfer Hydrogenation for **3a**[a]



Entry	Electrode	Electrolyte	Solvent	Temp./°C	Yield[b] of **3a**[%]	Yield[b] of **4a**[%]
1	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	**82**	trace
2	C cloth(+)|Pt(-)	$^{n}Bu_4NClO_4$	MeCN	70	N. D.	N. D.
3	C cloth(+)|Pt(-)	$^{n}Bu_4NBr$	MeCN	70	N. D.	N. D.
4	C cloth(+)|Pt(-)	$^{n}Bu_4NPF_6$	MeCN	70	72	trace
5	Pt(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	68	trace
6	C cloth(+)|Ni(-)	$^{n}Bu_4NBF_4$	MeCN	70	75	10
7	GC(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	trace	trace
8	C cloth(+)|C cloth(-)	$^{n}Bu_4NBF_4$	MeCN	70	60	14
9	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	DMSO	70	8	72
10	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	THF	70	N. D.	N. D.
11	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	DCE	70	N. D.	N. D.
12	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	DMF	70	N. D.	N. D.
13	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	H_2O	70	N. D.	N. D.
14[c]	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	65	18
15[d]	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	70	13
16	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	r. t.	33	trace
17	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	90	67	22
18[e]	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	trace	trace
19[f]	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	N. D.	N. D.
20[g]	C cloth(+)|Pt(-)	$^{n}Bu_4NBF_4$	MeCN	70	N. D.	N. D.

[a] Standard conditions: undivided cell, carbon cloth anode (10 × 10 mm), Pt cathode (10 × 10 × 0.1 mm), **1a** (0.5 mmol), **2a** (2.5 mmol), $^{n}Bu_4NBF_4$ (0.1 M), MeCN (10 mL), CCE = 15 mA, 12 h, 70 °C, under Ar. [b] Isolated yields. [c] CCE = 10 mA, 18 h. [d] CCE = 20 mA, 8 h. [e] Under air. [f] No electric current. [g] Without carbazate **2a**.

Table S2. Optimization of Transfer Hydrogenation for **4a**[a]

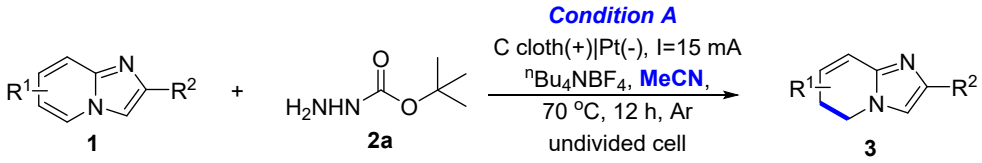


1a + H_2NHN (**2a**) → **4a**

Entry	Electrode	Electrolyte	I/mA	Time/h	Yield[b] of **3a**[%]	Yield[b] of **4a**[%]
1	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	18	12	trace	80
2	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	20	12	trace	76
3	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	18	18	trace	**85**
4	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	18	24	trace	81
5	Pt(+)|Pt(-)	$^nBu_4NBF_4$	18	18	11	69
6	Pt(+)|Ni(-)	$^nBu_4NBF_4$	18	18	trace	80
7	GC(+)|Pt(-)	$^nBu_4NBF_4$	18	18	trace	trace
8	C cloth(+)|C cloth(-)	$^nBu_4NBF_4$	18	18	trace	66
9	C cloth(+)|Pt(-)	$^nBu_4NClO_4$	18	18	trace	trace
10	C cloth(+)|Pt(-)	$^nBu_4NBr$	18	18	trace	trace
11	C cloth(+)|Pt(-)	$^nBu_4NPF_6$	18	18	trace	78
12[c]	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	18	18	trace	trace
13	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	0	18	N. D.	N. D.
14[d]	C cloth(+)|Pt(-)	$^nBu_4NBF_4$	18	18	N. D.	N. D.

[a] Reaction conditions: undivided cell, carbon cloth anode (10 × 10 mm), Pt cathode (10 × 10 × 0.1 mm), Ni foam cathode (10 × 10 mm), Glassy carbon anode (10 × 10 × 1 mm), **1a** (0.5 mmol), **2a** (2.5 mmol), $^nBu_4NBF_4$ (0.1 M), DMSO (10 mL), CCE = 15 mA, 12 h, 70 °C, under Ar. [b] Isolated yields. [c] Under air. [d] Without carbazate **2a**.

3. General Experimental Procedures



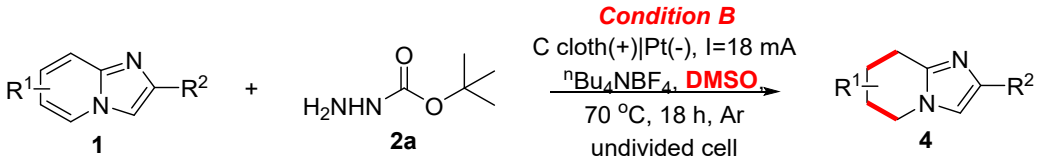
1 + H_2NHN (**2a**) → **3**

General Procedure A – for the preparation of products **3**:

Imidazo[1,2-*a*]pyridine (**1**, 0.5 mmol), *tert*-butyl carbazate (**2a**, 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) were added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. After completion, the reaction mixture was cooled to room temperature and concentrated under reduced pressure. The reaction mixture was

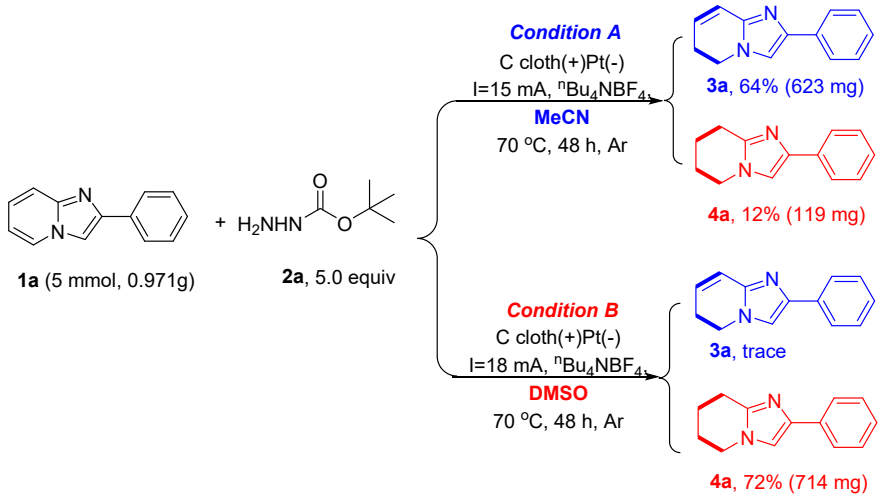
purified by column chromatography over silica gel eluting with petroleum ether/dichloromethane to give the desired products **3**.

General Procedure B - for the preparation of products **4**:



Imidazo[1,2-*a*]pyridine (**1**, 0.5 mmol), *tert*-butyl carbazate (**2a**, 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) were added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. After completion, the reaction mixture was washed with water and extracted with CH_2Cl_2 (10 mL x 3). The organic layers were combined, dried over Na_2SO_4, and concentrated. The reaction mixture was purified by column chromatography over silica gel eluting with petroleum ether/dichloromethane to give the desired products **4**.

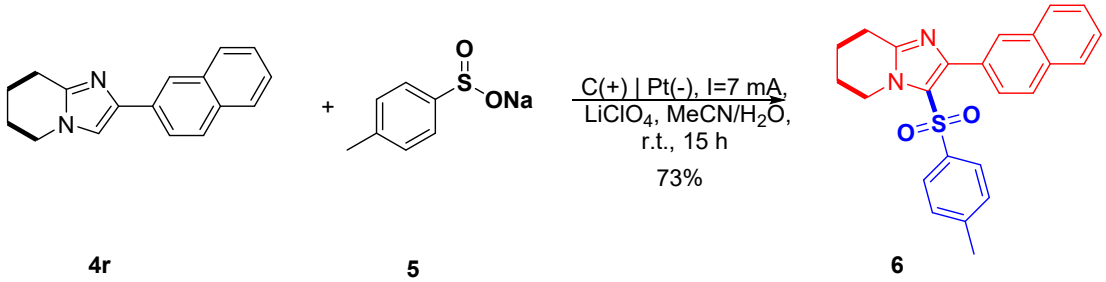
General Procedure C - for gram scale preparation of **3a** and **4a**:



Imidazo[1,2-*a*]pyridine **1a** (5 mmol, 0.97 g), *tert*-butyl carbazate **2a** (25 mmol, 3.30 g), $^{n}Bu_4NBF_4$ (2.5 mmol, 823.2 mg) were added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The reaction was carried out with 15 mA current using MeCN as solvent or 18 mA current using DMSO as solvent for 48 h at 70 °C under argon. After completion, the reaction mixture was washed with water and extracted with CH_2Cl_2 (10 mL x 3). The organic layers were combined, dried over Na_2SO_4, and concentrated. The reaction mixture was purified by column chromatography over silica gel eluting with petroleum ether/dichloromethane to give the desired products **3a** (64%, 623 mg)

under condition A and **4a** (72%, 714 mg) under condition B.

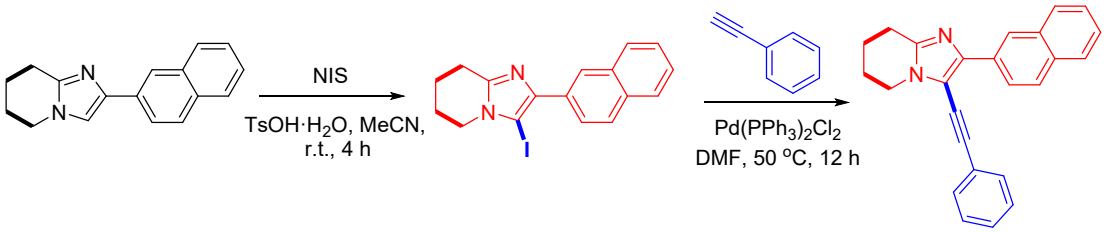
4. General procedure for the synthesis of compound 6[1]



4r **5** **6**

The hydrogenation product **4r** was obtained from **1r** in 78% yield by general procedure **C**. A solution of **4r** (1.0 mmol), sodium sulfinate (1.5 mmol), and $LiClO_4$ (320 mg, 0.42 M) in a mixture of CH_3CN and H_2O (14 mL, v/v = 2.5:1) was added to the undivided cell equipped with a carbon anode and a platinum cathode. The reaction mixture was stirred and electrolyzed at a constant current of 7 mA under room temperature for 15 h. After the reaction was complete, the residue was diluted with EtOAc (10 mL), washed with water, dried over Na_2SO_4, and concentrated under reduced pressure. The residue was purified by flash column chromatography to afford the desired product **6**.

5. General procedure for the synthesis of compound 8[2-3]



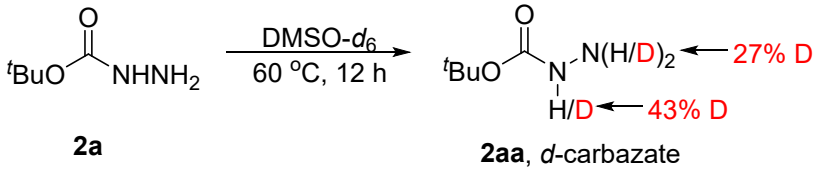
4r **7**, 81% **8**, 78% overall yields

To an oven dried 12 mL scintillation vial equipped with a magnetic stir bar, the compound **4r** (0.6 mmol, 1.0 equiv), *N*-iodosuccinimide (0.6 mmol, 1.0 equiv), *p*-toluenesulfonic acid (0.6 mmol, 1.0 equiv) were dissolved in the CH_3CN (6 mL, 0.1 M). The reaction mixture was stirred at ambient temperature for 4 h under argon atmosphere. After the reaction was complete by TLC, it was quenched with sat. $Na_2S_2O_3$ (10 mL), neutralized with sat. $NaHCO_3$ and extracted with DCM (20 mL × 3). The combined organic layers were washed with sat. $NaHCO_3$ (20 mL), followed by brine (20 mL), dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. The crude product was purified by silica gel column chromatography using EA/DCM as an eluent to provide the desired product **7** (181.8 mg, 81%).

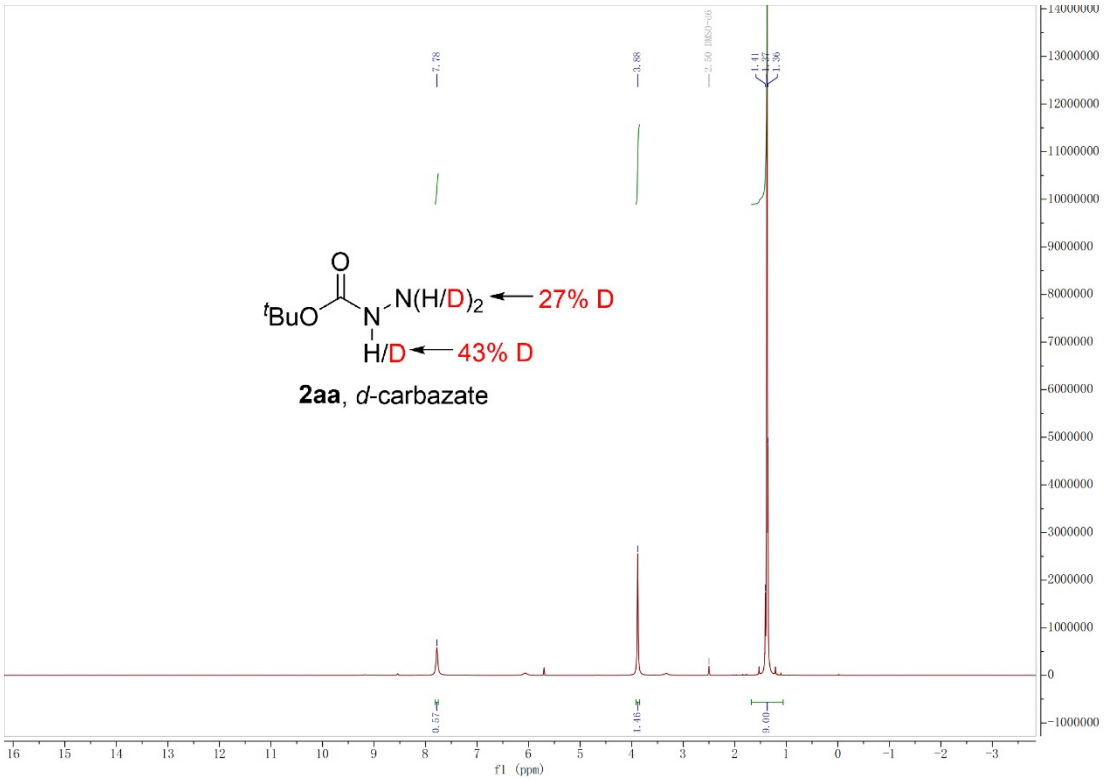
Compound **7** (150.0 mg, 0.4 mmol), Pd $(PPh_3)_2Cl_2$ (28 mg, 0.04 mmol, 10%) and CuI (8 mg, 0.05 mmol, 10%) were introduced into a screwcap test tube. The tube was sealed with a rubber septum and then evacuated and refilled with argon thrice. DMF (2 mL), Et_3N (225 µL, 1.6 mmol, 4 eq.) and phenylacetylene (44.9 mg, 0.44 mmol, 1.1 eq.)

were then added. The reaction mixture was stirred for 1.5 h at 80 °C. After cooling down to room temperature, the reaction mixture was partitioned between CH_2Cl_2 (10 mL) and brine (10 mL). The aqueous phase was extracted twice with CH_2Cl_2 (10 mL). Organic phases were reunited, dried over $MgSO_4$ and evaporated to dryness. The crude mixture was purified by silica gel column chromatography (EtOAc/petroleum ether = 1:5) to afford compound **8** as a white solid (133.8 mg, 78% overall yield).

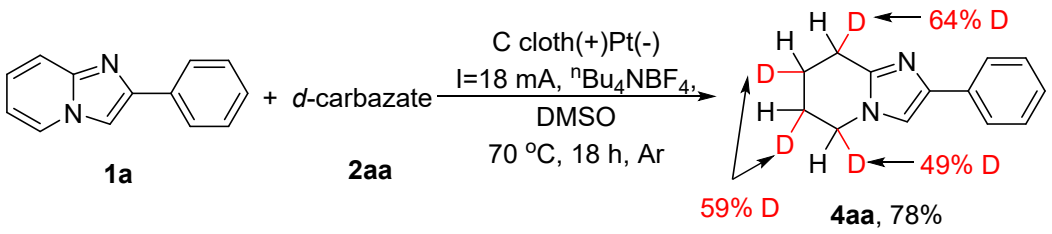
6. Deuterium-labelling experiments

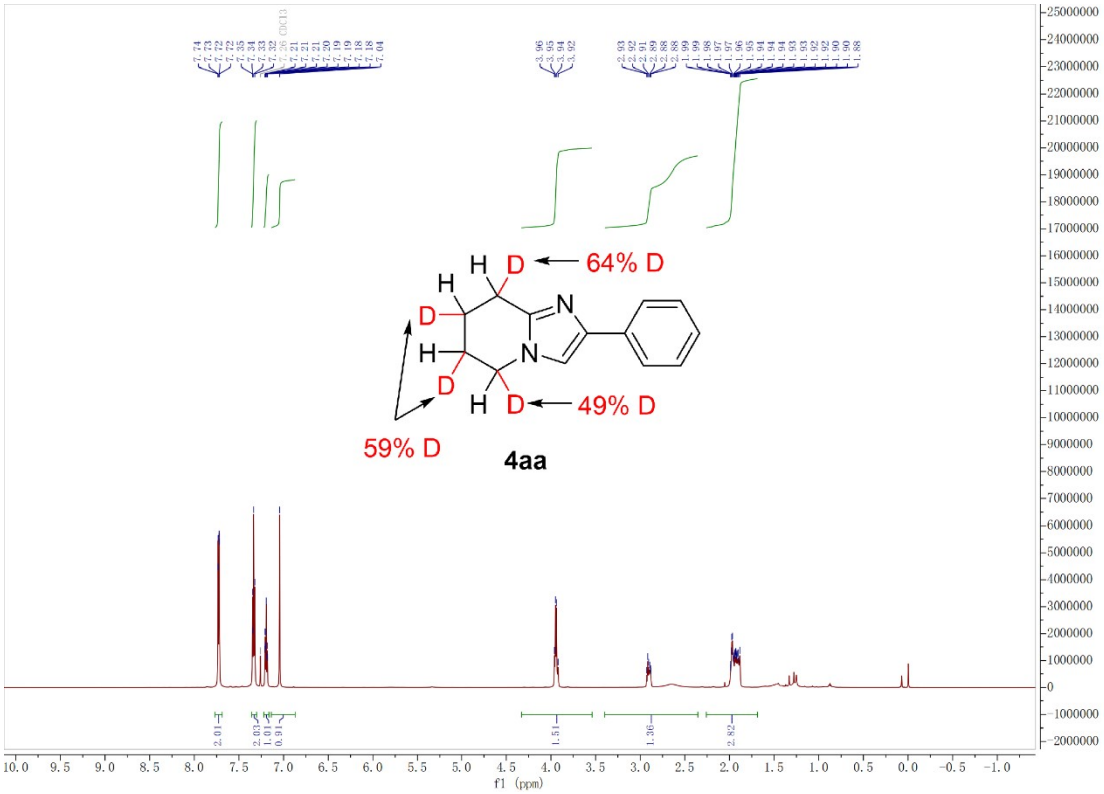


2a **2aa**, *d*-carbazate



To an oven dried 25 mL scintillation vial equipped with a magnetic stir bar, the compound **2a** (10 mmol, 1.32 g) were dissolved in DMSO-d_6 (10 mL). The reaction mixture was stirred at 60 °C for 12 h under argon atmosphere for hydrogen-deuterium exchange. After the reaction was complete, it was washed by brine (20 mL) and extracted with EA (20 mL × 3), followed by dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure to provide the deuterated carbazate **2aa** (1.03 g, 78%).

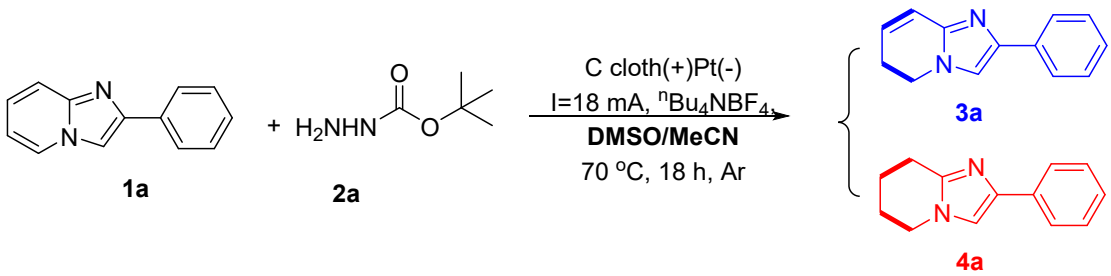




Imidazo[1,2-*a*]pyridine (**1a**, 0.5 mmol), deuterated *tert*-butyl carbazate (**2aa**, 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) were added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. **4aa** was isolated in 78% yield by column chromatography with 49-64% D-incorporation as revealed by ^{1}H NMR.

7. Exploration of solvent effect

Table S3. Exploration of solvent effect[a]



Entry	Solvent ratio	Yield%	
		3a	**4a**
1	**MeCN (10 mL)**	80	trace
2	**MeCN:DMSO=9:1**	74	16
3	**MeCN:DMSO=7:3**	56	35
4	**MeCN:DMSO=5:5**	41	46
5	**MeCN:DMSO=3:7**	15	72
6	**MeCN:DMSO=1:9**	9	80
7	**DMSO (10 mL)**	trace	84

[a] Standard conditions: **1a** (0.5 mmol), **2a** (2.5 mmol), undivided cell, carbon cloth anode (10 × 10 mm), Pt cathode (10 × 10 × 0.1 mm), CCE = 18 mA, $^{n}Bu_4NBF_4$ (0.1 M), solvent (10 mL), 18 h, 70 °C, under Ar. [b] Isolated yields

8. Cyclic voltammetry studies

The cyclic voltammograms were recorded in 0.1 M $^{n}Bu_4NBF_4$ solution in MeCN/DMSO with glassy carbon as the working electrode, Pt wire as the counter electrode and an Ag/AgCl (KCl sat'd) reference electrode as a reference electrode at room temperature. The scan rate was 100 mV/s. The reduction peak potential of 2-phenylimidazo[1,2-*a*]pyridine **1a** was observed at E_p = -1.33 ~ -1.05 V *vs.* Ag/AgCl.

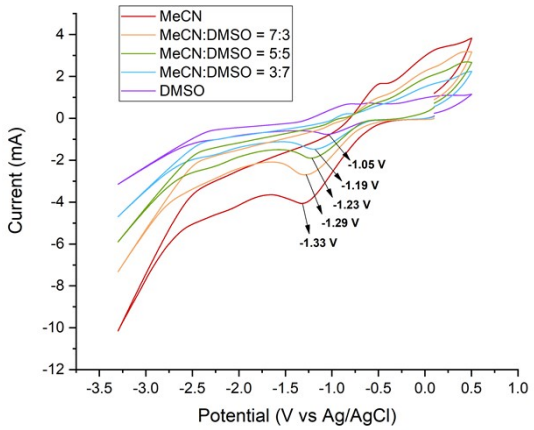
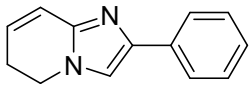


Figure S1. The cyclic voltammetry test of **1a** with different ratio of MeCN/DMSO.

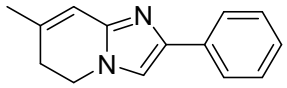
9. Characterization Data of Products

2-phenyl-5,6-dihydroimidazo[1,2-a]pyridine (3a)



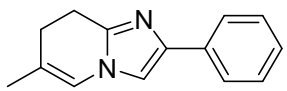
General procedure A was followed using **1a** (97.0 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3a** in 82% yield (80.4 mg) as a yellow solid: ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.80 – 7.73 (m, 2H), 7.41 – 7.33 (m, 2H), 7.25 – 7.19 (m, 1H), 7.14 (s, 1H), 6.09 – 6.02 (m, 1H), 5.94 – 5.86 (m, 1H), 4.84 – 4.49 (m, 2H), 3.85 – 3.40 (m, 2H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{13}H_{13}N_2$ 197.1079, found 197.0986. Spectral data match those previously reported.[4]

7-methyl-2-phenyl-5,6-dihydroimidazo[1,2-a]pyridine (3b)



General procedure A was followed using **1b** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3b** in 85% yield (89.3 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.84 – 7.64 (m, 2H), 7.36 (t, *J* = 7.7 Hz, 2H), 7.26 – 7.20 (m, 1H), 7.15 (s, 1H), 5.81 – 5.46 (m, 1H), 4.71 – 4.46 (m, 2H), 3.44 (t, *J* = 5.0 Hz, 2H), 1.90 (d, *J* = 2.1 Hz, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{15}N_2$ 211.1235, found 211.1226. Spectral data match those previously reported.[4]

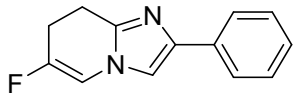
6-methyl-2-phenyl-7,8-dihydroimidazo[1,2-a]pyridine (3c)



General procedure A was followed using **1c** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3c** in 75% yield (78.8 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.91 – 7.63 (m, 2H), 7.36 (dd, *J* = 8.4, 7.1 Hz, 2H), 7.25 – 7.19

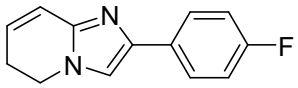
(m, 1H), 7.12 (s, 1H), 5.78 – 5.60 (m, 1H), 4.50 – 4.30 (m, 2H), 3.61 – 3.43 (m, 2H), 1.93 – 1.74 (m, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{15}N_2$ 211.1235, found 211.1222. Spectral data match those previously reported.[4]

6-fluoro-2-phenyl-7,8-dihydroimidazo[1,2-a]pyridine (3e)



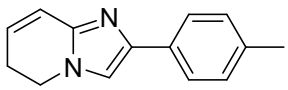
General procedure A was followed using **1e** (106.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3e** in 58% yield (62.1 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.02 – 7.63 (m, 2H), 7.37 (dd, *J* = 8.4, 6.9 Hz, 2H), 7.26 – 7.21 (m, 1H), 7.14 (s, 1H), 5.94 – 5.49 (m, 1H), 4.98 – 4.50 (m, 2H), 3.81 – 3.48 (m, 2H).HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{13}H_{12}N_2F$ 215.0985, found 215.0975. Spectral data match those previously reported.[4]

2-(4-fluorophenyl)-5,6-dihydroimidazo[1,2-a]pyridine (3f)



General procedure A was followed using **1f** (106.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3f** in 64% yield (68.5 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.84 – 7.54 (m, 2H), 7.07 (s, 1H), 7.07 – 6.89 (m, 2H), 6.18 – 5.97 (m, 1H), 5.97 – 5.84 (m, 1H), 4.72 – 4.43 (m, 2H), 3.67 – 3.42 (m, 2H).HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{13}H_{12}N_2F$ 215.0985, found 215.0980. Spectral data match those previously reported.[4]

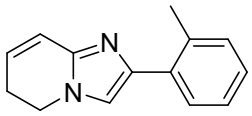
2-(p-tolyl)-5,6-dihydroimidazo[1,2-a]pyridine (3j)



General procedure A was followed using **1j** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3j** in 79% yield (83.0 mg) as a yellow solid: ^{1}H NMR (400

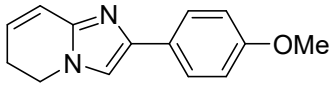
MHz, Chloroform-*d*) δ 7.96 – 7.54 (m, 2H), 7.17 (d, *J* = 7.9 Hz, 2H), 7.09 (s, 1H), 6.15 – 5.97 (m, 1H), 5.93 – 5.70 (m, 1H), 4.70 – 4.44 (m, 2H), 3.87 – 3.44 (m, 2H), 2.35 (s, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{15}N_2$ 211.1235, found 211.1233. Spectral data match those previously reported.[4]

2-(o-tolyl)-5,6-dihydroimidazo[1,2-a]pyridine (3k)



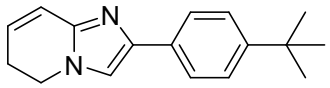
General procedure A was followed using **1k** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3k** in 82% yield (86.1 mg) as a yellow solid (mp 141-143 ℃): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.74 (dd, *J* = 7.5, 1.5 Hz, 1H), 7.17 – 7.04 (m, 3H), 6.88 (s, 1H), 6.22 – 5.88 (m, 1H), 5.86 – 5.77 (m, 1H), 4.72 – 4.23 (m, 2H), 3.77 – 3.35 (m, 2H), 2.38 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 141.58, 140.24, 134.74, 133.65, 130.68, 128.45, 126.66, 125.93, 123.18, 119.76, 115.83, 44.98, 25.05, 21.84. IR (film) 3441, 3317, 2975, 1702, 1642, 1480, 1388, 1365, 1248, 1152, 1048, 1017, 933, 848, 786, 732, 701, 601, 509 cm^{-1}; HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{15}N_2$ 211.1235, found 211.1227.

2-(4-methoxyphenyl)-5,6-dihydroimidazo[1,2-a]pyridine (3l)



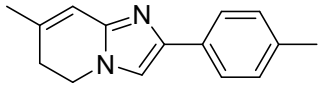
General procedure A was followed using **1l** (112.0 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3l** in 80% yield (90.4 mg) as a yellow solid (mp 102-104 ℃): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.88 – 7.50 (m, 2H), 7.04 (s, 1H), 6.97 – 6.82 (m, 2H), 6.14 – 5.98 (m, 1H), 5.95 – 5.81 (m, 1H), 4.74 – 4.53 (m, 2H), 3.82 (s, 3H), 3.63 – 3.49 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 158.66, 142.29, 141.10, 127.29, 123.13, 119.83, 114.04, 112.15, 55.35, 45.01, 25.18. IR (film) 3389, 3133, 3039, 2961, 2908, 2837, 1905, 1707, 1660, 1558, 1519, 1479, 1443, 1296, 1247, 1178, 1057, 1033, 979, 948, 840, 755, 669, 637, 539 cm^{-1}; HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{15}N_2O$ 227.1184, found 227.1175.

2-(4-(tert-butyl)phenyl)-5,6-dihydroimidazo[1,2-a]pyridine (3m)



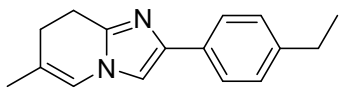
General procedure A was followed using **1m** (125.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3m** in 89% yield (112.2 mg) as a yellow solid (mp 121-123 °C): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.79 – 7.61 (m, 2H), 7.50 – 7.33 (m, 2H), 7.11 (s, 1H), 6.34 – 5.94 (m, 1H), 5.94 – 5.56 (m, 1H), 4.76 – 4.40 (m, 2H), 3.78 – 3.44 (m, 2H), 1.33 (s, 9H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 149.73, 142.42, 141.32, 131.55, 125.57, 124.66, 123.27, 119.82, 112.85, 45.10, 34.65, 31.49, 25.24. IR (film) 3135, 3039, 2961, 2904, 2867, 1905, 1710, 1611, 1519, 1478, 1426, 1396, 1265, 1195, 1163, 956, 841, 756, 725, 663, 555 cm^{-1}; HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{17}H_{21}N_2$ 253.1705, found 253.1697.

7-methyl-2-(p-tolyl)-5,6-dihydroimidazo[1,2-a]pyridine (3n)



General procedure A was followed using **1n** (111.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3n** in 79% yield (88.6 mg) as a yellow solid: ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.77 – 7.51 (m, 2H), 7.17 (d, *J* = 7.9 Hz, 2H), 7.09 (s, 1H), 5.62 – 5.56 (m, 1H), 4.60 – 4.47 (m, 2H), 3.42 (t, *J* = 5.3 Hz, 2H), 2.35 (s, 3H), 1.90 (d, *J* = 2.3 Hz, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{15}H_{17}N_2$ 255.1392, found 255.1381. Spectral data match those previously reported.[4]

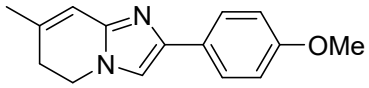
2-(4-ethylphenyl)-6-methyl-7,8-dihydroimidazo[1,2-a]pyridine (3o)



General procedure A was followed using **1o** (118.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3o** in 67% yield (79.8 mg) as a yellow solid (mp 109-111 °C): ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.68 (d, *J* = 8.1 Hz, 2H), 7.19 (d, *J* = 7.9 Hz,

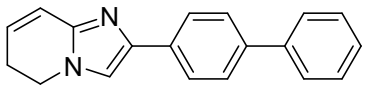
2H), 7.08 (s, 1H), 5.89 – 5.53 (m, 1H), 4.44 (t, *J* = 5.1 Hz, 2H), 3.60 – 3.37 (m, 2H), 2.65 (q, *J* = 7.6 Hz, 2H), 1.85 (d, *J* = 2.5 Hz, 3H), 1.25 (t, *J* = 7.6 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 142.84, 142.67, 141.51, 131.83, 128.16, 127.15, 124.92, 117.86, 112.45, 48.57, 28.72, 25.20, 20.44, 15.68. IR (film) 3419, 2963, 2877, 1619, 1558, 1526, 1488, 1433, 1381, 1342, 1058, 883, 837, 795, 764, 740, 530 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{16}H_{19}N_2$ 239.1548, found 239.1540.

2-(4-methoxyphenyl)-7-methyl-5,6-dihydroimidazo[1,2-a]pyridine (3p)



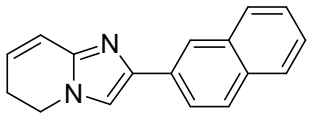
General procedure A was followed using **1p** (119.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), ^{n}Bu$_4$NBF$_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3p** in 88% yield (105.7 mg) as a yellow solid (mp 130-132 ℃): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.06 – 7.53 (m, 2H), 7.04 (s, 1H), 6.94 – 6.85 (m, 2H), 5.64 – 5.49 (m, 1H), 4.81 – 4.45 (m, 2H), 3.82 (s, 3H), 3.46 – 3.34 (m, 2H), 2.09 – 1.67 (m, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.71, 143.04, 141.19, 131.16, 127.21, 126.15, 114.22, 114.09, 112.10, 55.40, 45.04, 29.70, 23.02. IR (film) 2959, 2875, 1708, 1609, 1511, 1481, 1372, 1244, 1172, 1052, 1027, 956, 834, 732, 701, 601, 523 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{15}H_{17}N_2O$ 241.1341, found 241.1339.

2-([1,1'-biphenyl]-4-yl)-5,6-dihydroimidazo[1,2-a]pyridine (3q)



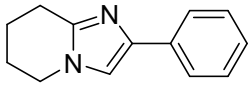
General procedure A was followed using **1q** (135.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), ^{n}Bu$_4$NBF$_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3q** in 85% yield (115.7 mg) as a yellow solid (mp 154-156 ℃): ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.91 – 7.74 (m, 2H), 7.67 – 7.60 (m, 4H), 7.43 (t, *J* = 7.7 Hz, 2H), 7.37 – 7.27 (m, 1H), 7.19 (s, 1H), 6.14 – 6.01 (m, 1H), 5.96 – 5.90 (m, 1H), 4.68 – 4.57 (m, 2H), 3.64 – 3.54 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 142.73, 141.10, 140.91, 139.50, 133.35, 128.87, 127.40, 127.22, 127.03, 125.30, 119.80, 113.39, 45.18, 25.25. IR (film) 3386, 3038, 2962, 2900, 1913, 1667, 1596, 1612, 1558, 1488, 1450, 1427, 1396, 1373, 1326, 1191, 1157, 1080, 1002, 948, 894, 847, 756, 725, 694, 663, 486 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{19}H_{17}N_2$ 273.1392, found 273.1384.

2-(naphthalen-2-yl)-5,6-dihydroimidazo[1,2-a]pyridine (3r)



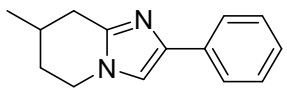
General procedure A was followed using **1r** (122.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and MeCN (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (15 mA) electrolysis was carried out at 70 °C under argon for 12 h. Chromatography (DCM/EA = 50/1) afforded **3r** in 83% yield (102.2 mg) as a yellow solid (mp 144-146 °C): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.30 (s, 1H), 7.87 (dd, *J* = 7.6, 1.6 Hz, 1H), 7.84 – 7.78 (m, 4H), 7.50 – 7.36 (m, 2H), 6.16 – 5.98 (m, 1H), 5.99 – 5.85 (m, 1H), 4.83 – 4.51 (m, 2H), 3.70 – 3.48 (m, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 142.79, 141.08, 133.89, 132.60, 131.51, 128.12, 127.65, 126.11, 125.34, 123.64, 123.09, 122.79, 119.69, 113.70, 45.07, 25.15. IR (film) 3131, 3046, 2897, 1928, 1837, 1710, 1666, 1599, 1518, 1419, 1396, 1372, 1318, 1186, 1163, 995, 923, 863, 770, 728, 669, 640, 594, 476 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^{+}$ Calcd for $C_{17}H_{15}N_2$ 247.1235, found 247.1225.

2-phenyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4a)



General procedure B was followed using **1a** (97.0 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4a** in 85% yield (84.2 mg) as a yellow solid: ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.83 – 7.56 (m, 2H), 7.45 (s, 1H), 7.31 (t, *J* = 7.7 Hz, 2H), 7.24 – 7.05 (m, 1H), 3.95 (t, *J* = 5.9 Hz, 2H), 2.75 (t, *J* = 6.3 Hz, 2H), 2.04 – 1.88 (m, 2H), 1.88 – 1.82 (m, 2H). HRMS (ESI-TOF) m/z [M + H]$^{+}$ Calcd for $C_{13}H_{15}N_2$ 199.1235, found 199.1238. Spectral data match those previously reported.[5]

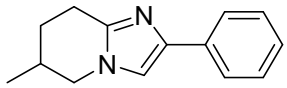
7-methyl-2-phenyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4b)



General procedure B was followed using **1b** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography

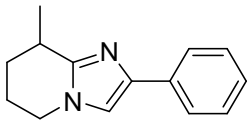
(DCM/EA = 8/1) afforded **4b** in 78% yield (82.8 mg) as a yellow oil: ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.97 – 7.61 (m, 2H), 7.45 (s, 1H), 7.31 (t, J = 7.8 Hz, 2H), 7.22 – 7.10 (m, 1H), 4.17 – 3.79 (m, 2H), 3.00 – 2.80 (m, 1H), 2.39 – 2.25 (m, 1H), 2.13 – 1.89 (m, 2H), 1.74 – 1.51 (m, 1H), 1.07 (d, J = 6.5 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 145.43, 140.52, 134.34, 128.66, 126.69, 124.87, 114.01, 45.03, 24.65, 23.16, 21.23. IR (film) 3116, 3067, 2975, 2940, 1719, 1643, 1504, 1473, 1365, 1303, 1245, 1080, 1065, 919, 864, 786, 724, 694 cm^{-1}; HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{17}N_2$ 213.1392, found 213.1379.

6-methyl-2-phenyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4c)



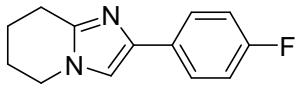
General procedure B was followed using **1c** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4c** in 70% yield (74.3 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.81 – 7.66 (m, 2H), 7.34 (dd, J = 8.4, 7.0 Hz, 2H), 7.24 – 7.15 (m, 1H), 7.04 (s, 1H), 4.14 – 3.96 (m, 1H), 3.52 (dd, J = 12.1, 10.2 Hz, 1H), 3.07 (d, J = 17.1, 3.2 Hz, 1H), 2.93 – 2.77 (m, 1H), 2.22 – 1.97 (m, 2H), 1.69 – 1.50 (m, 1H), 1.12 (d, J = 6.7 Hz, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{17}N_2$ 213.1392, found 213.1382. Spectral data match those previously reported.[2]

8-methyl-2-phenyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4d)



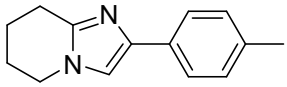
General procedure B was followed using **1d** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4d** in 59% yield (62.6 mg) as a yellow oil: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.74 (dd, J = 8.1, 1.5 Hz, 2H), 7.34 (t, J = 7.7 Hz, 2H), 7.21 – 7.15 (m, 1H), 7.01 (s, 1H), 4.16 – 3.75 (m, 2H), 3.13 – 2.77 (m, 1H), 2.13 – 2.00 (m, 2H), 1.96 – 1.84 (m, 1H), 1.60 – 1.51 (m, 1H), 1.45 (d, J = 6.9 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 149.86, 140.61, 134.69, 128.50, 126.40, 124.87, 113.87, 45.13, 30.26, 29.82, 21.65, 19.97; IR (film) 2938, 2864, 1718, 1640, 1511, 1473, 1452, 1380, 1318, 1257, 1118, 1072, 1026, 948, 909, 856, 727, 694, 641, 506 cm^{-1}; HRMS (ESI-TOF) calcd for ($[C_{14}H_{17}N_2]^+$) $[M+H]^+$ m/z = 213.1392; found 213.1379.

2-(4-fluorophenyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4f)



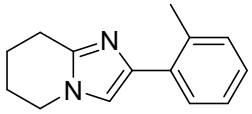
General procedure B was followed using **1f** (106.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4f** in 73% yield (78.9 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.92 – 7.53 (m, 2H), 7.34 (t, *J* = 7.7 Hz, 1H), 7.24 – 7.16 (m, 1H), 7.04 (d, *J* = 12.8 Hz, 1H), 3.96 (dd, *J* = 7.3, 4.3 Hz, 2H), 2.96 – 2.87 (m, 2H), 2.06 – 1.85 (m, 4H).HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{13}H_{14}N_2F$ 217.1141, found 217.1148. Spectral data match those previously reported.[5]

2-(p-tolyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4j)



General procedure B was followed using **1j** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4j** in 88% yield (93.4 mg) as a yellow solid: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.80 – 7.48 (m, 2H), 7.15 (d, *J* = 7.9 Hz, 2H), 7.02 (d, *J* = 1.1 Hz, 1H), 3.96 (t, *J* = 5.8 Hz, 2H), 2.92 (t, *J* = 6.1 Hz, 2H), 2.34 (s, 3H), 2.11 – 1.84 (m, 4H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 145.26, 140.52, 136.32, 131.46, 129.35, 124.80, 113.54, 44.97, 24.58, 23.14, 21.31, 21.19.; IR (film) 2945, 2896, 1714, 1640, 1558, 1509, 1481, 1430, 1383, 1309, 1258, 1186, 1109, 1074, 910, 834, 727, 646, 550 cm^{-1}; HRMS (ESI-TOF) calcd for ($[C_{14}H_{17}N_2]^+$) $[M+H]^+$ m/z = 213.1392; found 213.1385.

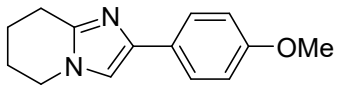
2-(o-tolyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4k)



General procedure B was followed using **1k** (104.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4k** in 79% yield (83.8 mg) as a yellow solid:^{1}H NMR (400

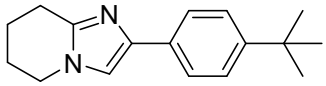
MHz, Chloroform-*d*) δ 7.83 (dd, *J* = 7.7, 1.4 Hz, 1H), 7.30 – 7.04 (m, 3H), 6.90 (s, 1H), 3.99 (t, *J* = 5.8 Hz, 2H), 2.93 (t, *J* = 6.3 Hz, 2H), 2.46 (s, 3H), 2.19 – 1.88 (m, 4H). HRMS (ESI-TOF) calcd for ($[C_{14}H_{17}N_2]^+$) $[M+H]^+$ m/z = 213.1392; found 213.1386. Spectral data match those previously reported.[5]

2-(4-methoxyphenyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4l)



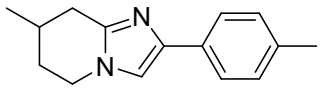
General procedure B was followed using **1l** (97.0 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4l** in 68% yield (77.6 mg) as a yellow solid:^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.22 – 7.41 (m, 2H), 6.96 (s, 1H), 6.89 (d, *J* = 8.8 Hz, 2H), 3.95 (t, *J* = 5.8 Hz, 2H), 3.81 (s, 3H), 2.91 (t, *J* = 6.2 Hz, 2H), 2.11 – 1.87 (m, 4H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{14}H_{17}N_2O$ 229.1341, found 229.1338. Spectral data match those previously reported.[5]

2-(4-(tert-butyl)phenyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4m)



General procedure B was followed using **1m** (125.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^nBu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4m** in 81% yield (103.0 mg) as a yellow oil: ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.89 – 7.49 (m, 2H), 7.44 – 7.31 (m, 2H), 7.02 (s, 1H), 3.97 (t, *J* = 5.9 Hz, 2H), 2.93 (t, *J* = 6.4 Hz, 2H), 2.28 – 1.75 (m, 4H), 1.32 (s, 9H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 149.50, 145.21, 140.66, 131.68, 125.52, 124.58, 113.59, 44.96, 34.62, 31.50, 24.69, 23.19, 21.28.; IR (film) 3409, 2959, 2876, 1911, 1706, 1617, 1557, 1512, 1481, 1426, 1379, 1295, 1267, 1193, 1073, 1037, 948, 864, 825, 761, 670, 509 cm^{-1}; HRMS (ESI-TOF) calcd for ($[C_{17}H_{23}N_2]^+$) $[M+H]^+$ m/z = 255.1856; found 255.1851.

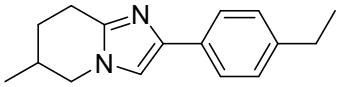
7-methyl-2-(p-tolyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4n)



General procedure B was followed using **1n** (111.1 mg, 0.5 mmol), *tert*-butyl

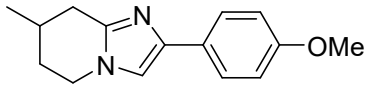
carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4n** in 72% yield (81.4 mg) as a yellow solid: (mp 136-138 °C): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.72 – 7.53 (m, 2H), 7.15 (d, *J* = 7.9 Hz, 2H), 7.02 (s, 1H), 4.23 – 3.75 (m, 2H), 3.28 – 2.90 (m, 1H), 2.45 (dd, *J* = 16.6, 10.4 Hz, 1H), 2.33 (s, 3H), 2.18 – 1.92 (m, 2H), 1.72 – 1.61 (m, 1H), 1.14 (d, *J* = 6.6 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 145.27, 140.87, 136.05, 131.65, 129.21, 124.62, 113.27, 43.99, 32.76, 30.93, 28.01, 21.21. IR (film) 2954, 2923, 1707, 1558, 1511, 1480, 1449, 1427, 1373, 1329, 1303, 1269, 1186, 1107, 1072, 909, 824, 763, 727, 646, 547, 509 cm^{-1}; HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{15}H_{19}N_2$ 227.1548, found 227.1537.

2-(4-ethylphenyl)-6-methyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4o)



General procedure B was followed using **1o** (118.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4o** in 79% yield (94.9 mg) as a yellow solid:^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.83 – 7.53 (m, 2H), 7.22 – 7.05 (m, 2H), 6.99 (s, 1H), 4.10 – 3.94 (m, 1H), 3.49 (dd, *J* = 12.0, 10.2 Hz, 1H), 3.13 – 2.96 (m, 1H), 2.94 – 2.76 (m, 1H), 2.64 (q, *J* = 7.6 Hz, 2H), 2.17 – 1.95 (m, 2H), 1.65 – 1.51 (m, 1H), 1.24 (t, *J* = 7.6 Hz, 3H), 1.11 (d, *J* = 6.6 Hz, 3H). HRMS (ESI-TOF) m/z $[M + H]^+$ Calcd for $C_{16}H_{21}N_2$ 241.1700, found 241.1693. Spectral data match those previously reported.[4]

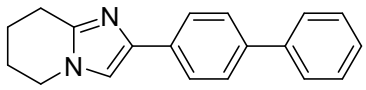
2-(4-methoxyphenyl)-7-methyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4p)



General procedure B was followed using **1p** (119.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), $^{n}Bu_4NBF_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4p** in 70% yield (84.8 mg) as a yellow solid (mp 143-145 °C): ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.68 – 7.62 (m, 2H), 6.97 (s, 1H), 6.90 – 6.86 (m, 2H), 4.06 – 3.98 (m, 1H), 3.92 – 3.86 (m, 1H), 3.81 (s, 3H), 3.11 – 3.03 (m, 1H), 2.45 (dd, *J* = 16.5, 10.4 Hz, 1H), 2.08 – 1.98 (m, 2H), 1.73 – 1.62 (m, 1H), 1.14

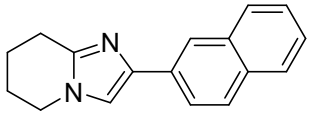
(d, J = 6.6 Hz, 3H). ^{13}C NMR (101 MHz, DMSO-d_6) δ 158.61, 145.33, 140.76, 127.40, 126.07, 114.06, 112.76, 55.39, 44.09, 32.81, 31.04, 28.11, 21.29. IR (film) 2963, 1710, 1611, 1556, 1460, 1385, 1365, 1295, 1241, 1171, 1032, 948, 832, 787, 761, 641, 524 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{15}H_{19}N_2O$ 243.1497, found 243.1489.

2-([1,1'-biphenyl]-4-yl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4q)



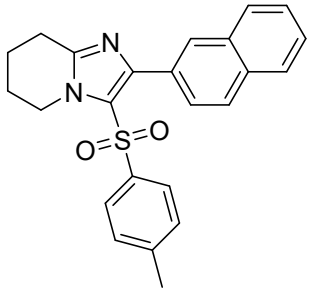
General procedure B was followed using **1q** (135.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), ^{n}Bu$_4$NBF$_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4q** in 75% yield (102.8 mg) as a yellow solid:^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.94 – 7.71 (m, 2H), 7.70 – 7.56 (m, 4H), 7.43 (dd, J = 8.3, 7.0 Hz, 2H), 7.37 – 7.29 (m, 1H), 7.11 (s, 1H), 3.99 (t, J = 5.8 Hz, 2H), 2.95 (t, J = 6.2 Hz, 2H), 2.15 – 1.87 (m, 4H). HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{19}H_{19}N_2$ 275.1548, found 275.1536. Spectral data match those previously reported.[6]

2-(naphthalen-2-yl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (4r)



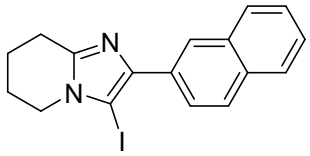
General procedure B was followed using **1r** (122.1 mg, 0.5 mmol), *tert*-butyl carbazate **2a** (330.4 mg 2.5 mmol), ^{n}Bu$_4$NBF$_4$ (0.5 mmol, 164.6 mg), and DMSO (10 mL) added to a three-necked, round-bottomed flask equipped with carbon cloth anode (10 × 10 mm) and platinum plate cathode (10 × 10 × 0.1 mm). The constant current (18 mA) electrolysis was carried out at 70 °C under argon for 18 h. Chromatography (DCM/EA = 8/1) afforded **4r** in 86% yield (106.7 mg) as a yellow oil: ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.28 (s, 1H), 7.94 – 7.69 (m, 4H), 7.51 – 7.33 (m, 2H), 7.16 (s, 1H), 3.97 (t, J = 5.7 Hz, 2H), 2.96 (t, J = 6.1 Hz, 2H), 2.05 – 1.89 (m, 4H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 145.58, 140.37, 133.91, 132.51, 131.67, 128.09, 128.05, 127.63, 126.06, 125.24, 123.65, 122.60, 114.49, 44.93, 24.63, 23.03, 21.12. IR (film) 3052, 2947, 2186, 1712, 1628, 1519, 1481, 1424, 1374, 1341, 1318, 1196, 1071, 932, 905, 857, 818, 755, 725, 641, 585, 475 cm^{-1}. HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{17}H_{17}N_2$ 249.1386, found 249.1380.

2-(naphthalen-2-yl)-3-tosyl-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (6)



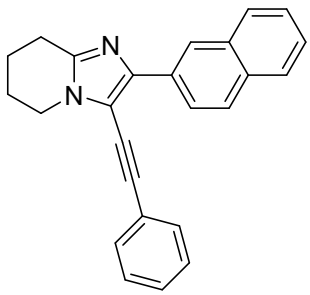
It was obtained in 73% yield (293.8 mg) as a yellow solid (mp 157-159 °C): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.11 (d, *J* = 1.7 Hz, 1H), 7.95 – 7.80 (m, 3H), 7.75 (dd, *J* = 8.5, 1.7 Hz, 1H), 7.57 – 7.52 (m, 2H), 7.52 – 7.46 (m, 2H), 7.14 (d, *J* = 8.1 Hz, 2H), 4.23 (t, *J* = 6.0 Hz, 2H), 2.96 (t, *J* = 6.4 Hz, 2H), 2.32 (s, 3H), 2.04 – 1.97 (m, 2H), 1.94 – 1.87 (m, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 148.97, 148.13, 144.25, 139.47, 133.41, 132.89, 130.68, 129.79, 128.67, 127.92, 127.75, 127.18, 127.01, 126.44, 126.08, 123.98, 45.74, 25.28, 22.77, 21.61, 19.79. IR (film) 3055, 2946, 1596, 1506, 1419, 1442, 1316, 1141, 1118, 1080, 964, 894, 825, 810, 762, 694, 655, 586, 539, 475 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^{+}$ Calcd for $C_{24}H_{23}N_2O_2S$ 403.1480, found 403.1476.

3-iodo-2-(naphthalen-2-yl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (7)



It was obtained in 81% yield (181.9 mg) as a yellow solid (mp 152-154 °C): ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.45 – 8.33 (m, 1H), 8.09 (dd, *J* = 8.5, 1.7 Hz, 1H), 7.95 – 7.78 (m, 3H), 7.54 – 7.41 (m, 2H), 3.88 (t, *J* = 6.0 Hz, 2H), 2.99 (t, *J* = 6.4 Hz, 2H), 2.11 – 1.94 (m, 4H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 149.06, 142.78, 133.51, 132.75, 131.65, 128.37, 127.81, 127.75, 126.09, 125.96, 125.83, 125.67, 66.84, 46.85, 25.58, 23.44, 21.30. IR (film) 3047, 2925, 1712, 1627, 1496, 1481, 1419, 1358, 1334, 1103, 994, 934, 894, 853, 817, 743, 702, 678, 593, 473 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^{+}$ Calcd for $C_{17}H_{16}N_2I$ 375.0358, found 375.0345.

2-(naphthalen-2-yl)-3-(phenylethynyl)-5,6,7,8-tetrahydroimidazo[1,2-a]pyridine (8)



It was obtained in 78% overall yield (133.8 mg) as a yellow solid (mp 183-185 °C): ^{1}H

NMR (400 MHz, Chloroform-*d*) δ 8.66 (d, *J* = 1.7 Hz, 1H), 8.37 (dd, *J* = 8.6, 1.8 Hz, 1H), 7.89 (dd, *J* = 8.8, 3.6 Hz, 2H), 7.85 – 7.79 (m, 1H), 7.61 – 7.53 (m, 2H), 7.51 – 7.33 (m, 5H), 4.07 (t, *J* = 5.9 Hz, 2H), 2.99 (t, *J* = 6.3 Hz, 2H), 2.20 – 1.85 (m, 4H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 146.31, 143.55, 133.77, 132.89, 131.63, 131.18, 128.68, 128.56, 128.43, 127.97, 127.76, 126.09, 125.73, 124.64, 124.49, 123.20, 110.82, 98.79, 79.88, 43.67, 25.07, 22.79, 20.85. IR (film) 3444, 3343, 3054, 2947, 2198, 1956, 1669, 1519, 1442, 1389, 1234, 1191, 1092, 964, 910, 817, 749, 689, 647, 586, 564, 484 cm^{-1}; HRMS (ESI-TOF) m/z [M + H]$^+$ Calcd for $C_{25}H_{21}N_2$ 349.1699, found 349.1696.

10.References

[1] W. Kim, H. Y. Kim, K. Oh, *J Org Chem* **2021**, *86*, 15973-15991.

[2] D. V. Patil, Y. Lee, H. Y. Kim, K. Oh, *Org Lett* **2022**, *24*, 5840-5844.

[3] P.-O. Delaye, M. Pénichon, H. Allouchi, C. Enguehard-Gueiffier, A. Gueiffier, *Org Biomol Chem* **2017**, *15*, 4199-4204.

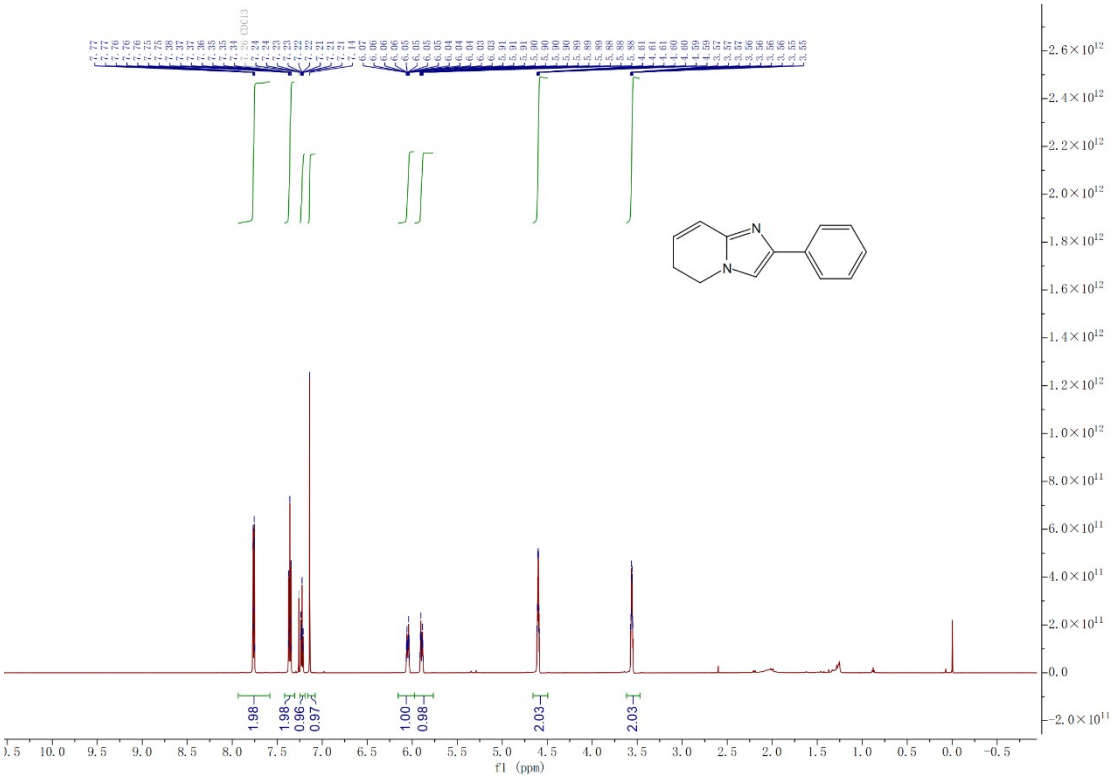
[4] J. Wen, H. Qin, K. Yan, X. Yang, X. Sun, W. Wei, J. Yang, H. Wang, *Org Lett* **2020**, *22*, 8824-8828.

[5] Q. Xuan, Q. Song, *Org Lett* **2016**, *18*, 4250-4253.

[6] J. Li, P. Zhang, M. Jiang, H. Yang, Y. Zhao, H. Fu, *Org Lett* **2017**, *19*, 1994-1997.

11. Copies of 1H NMR, 13C NMR and 19F NMR

3a



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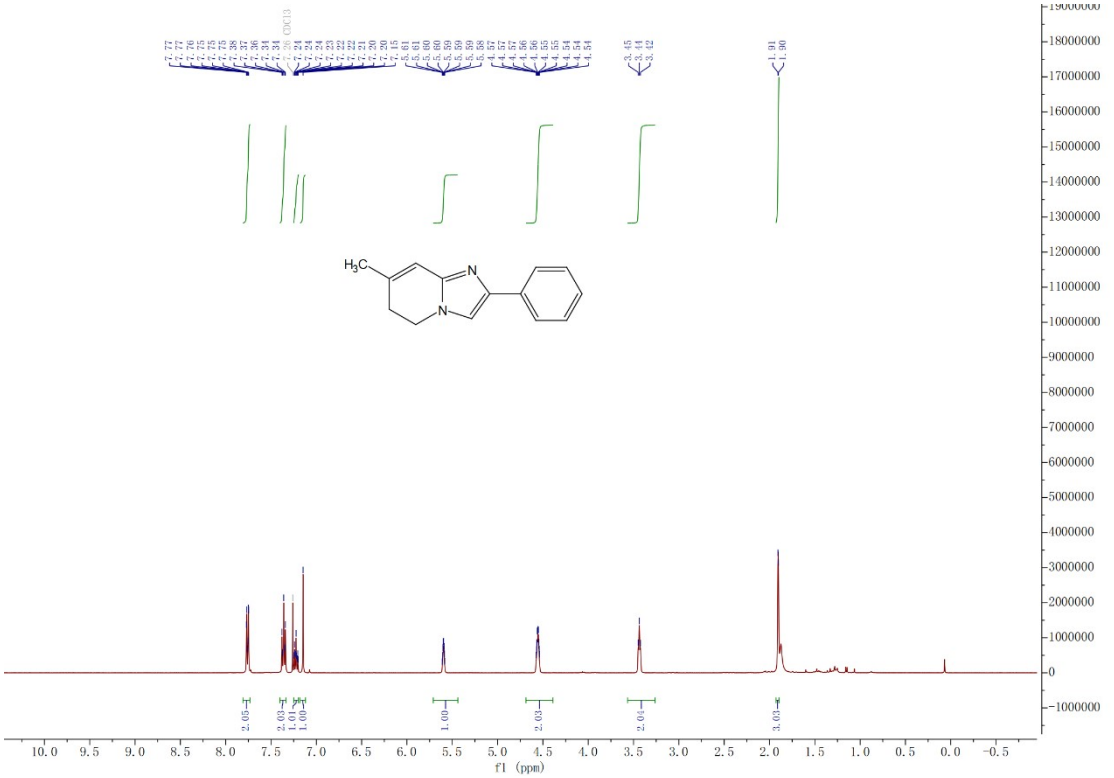

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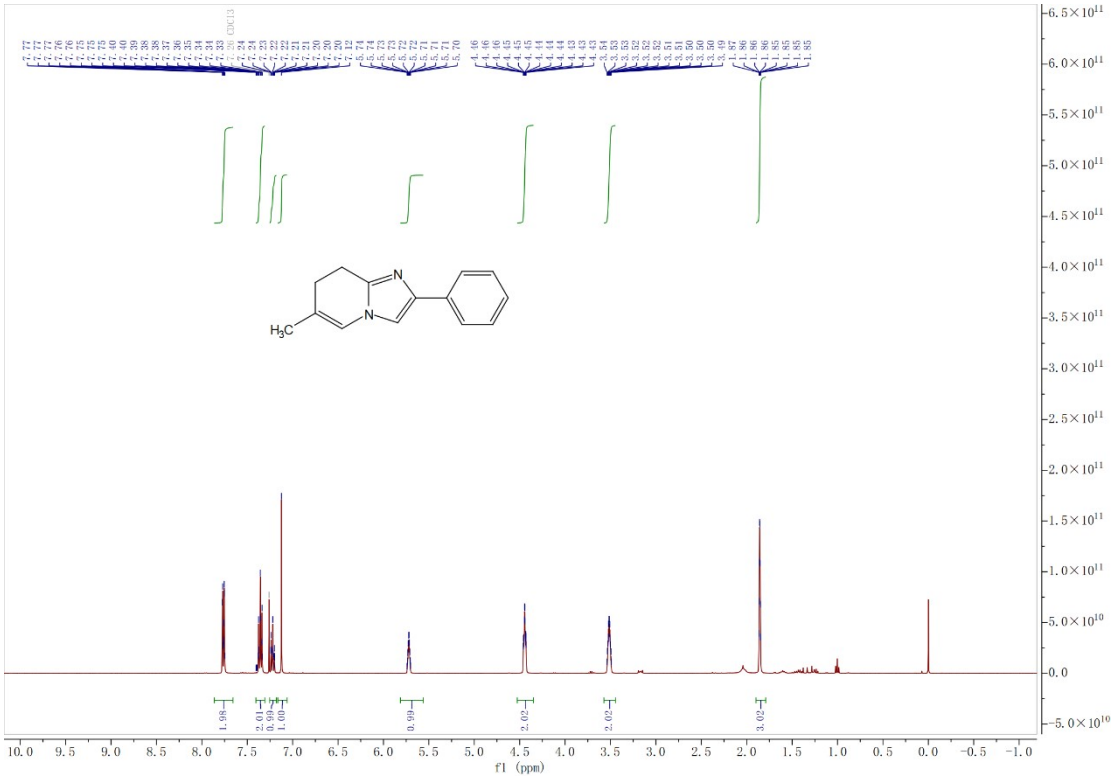

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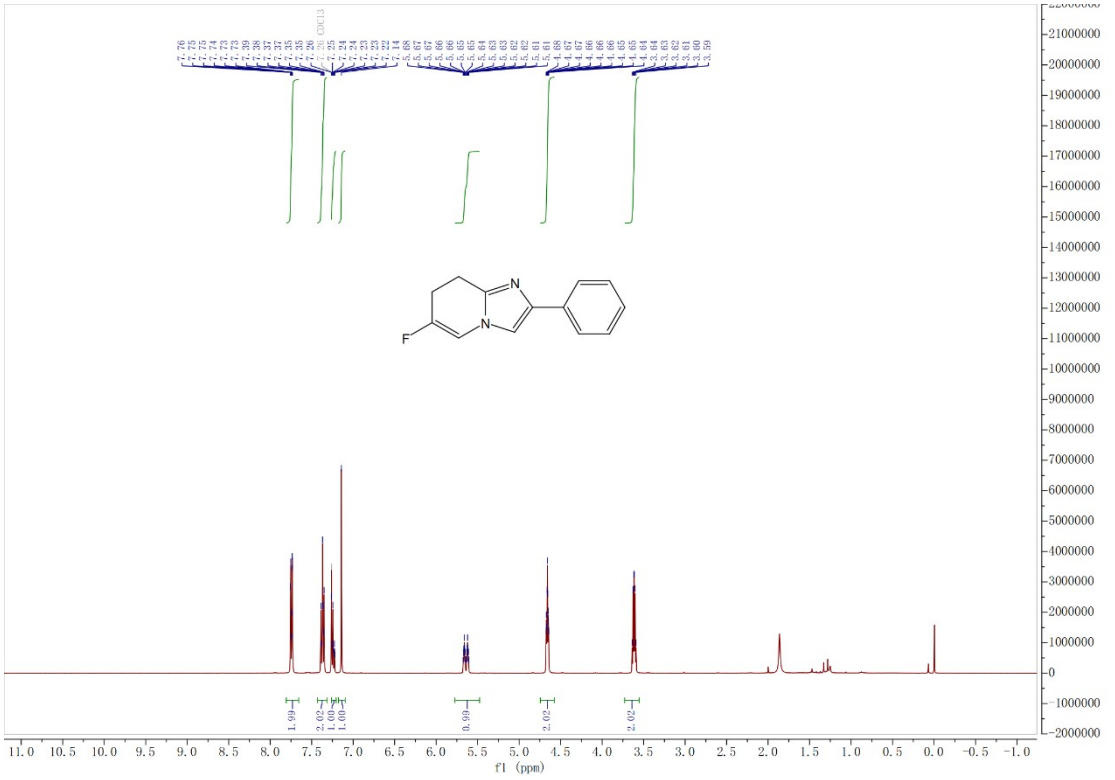

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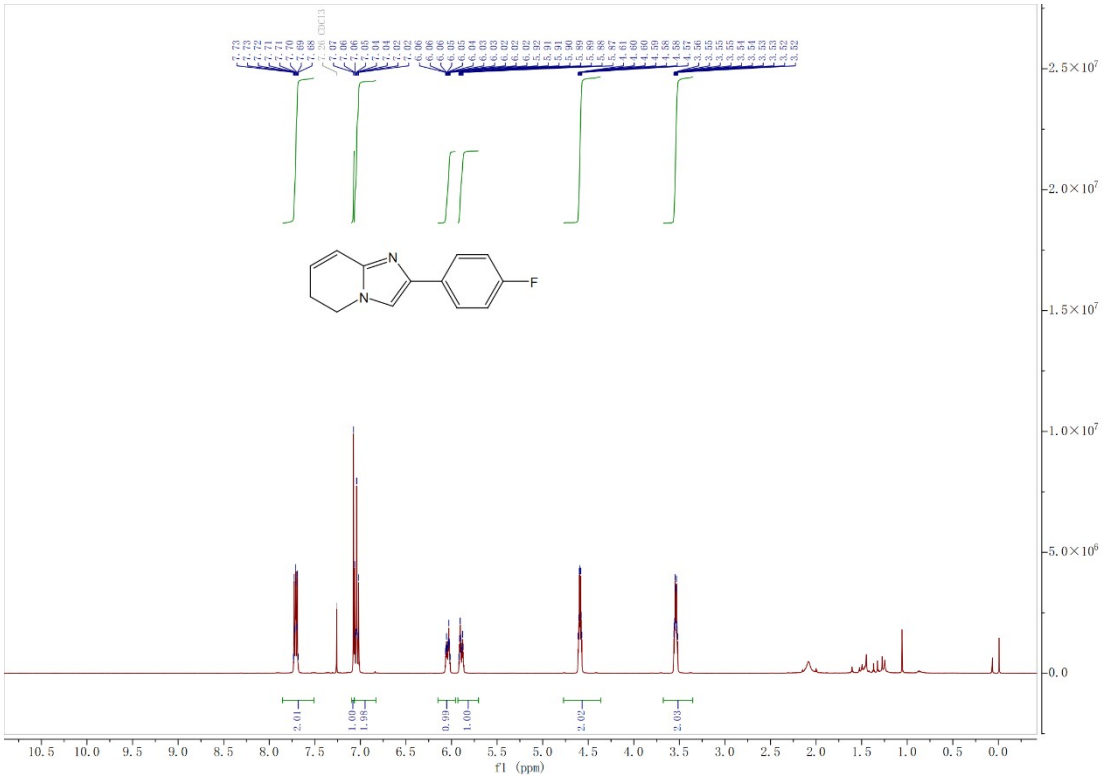
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N
F
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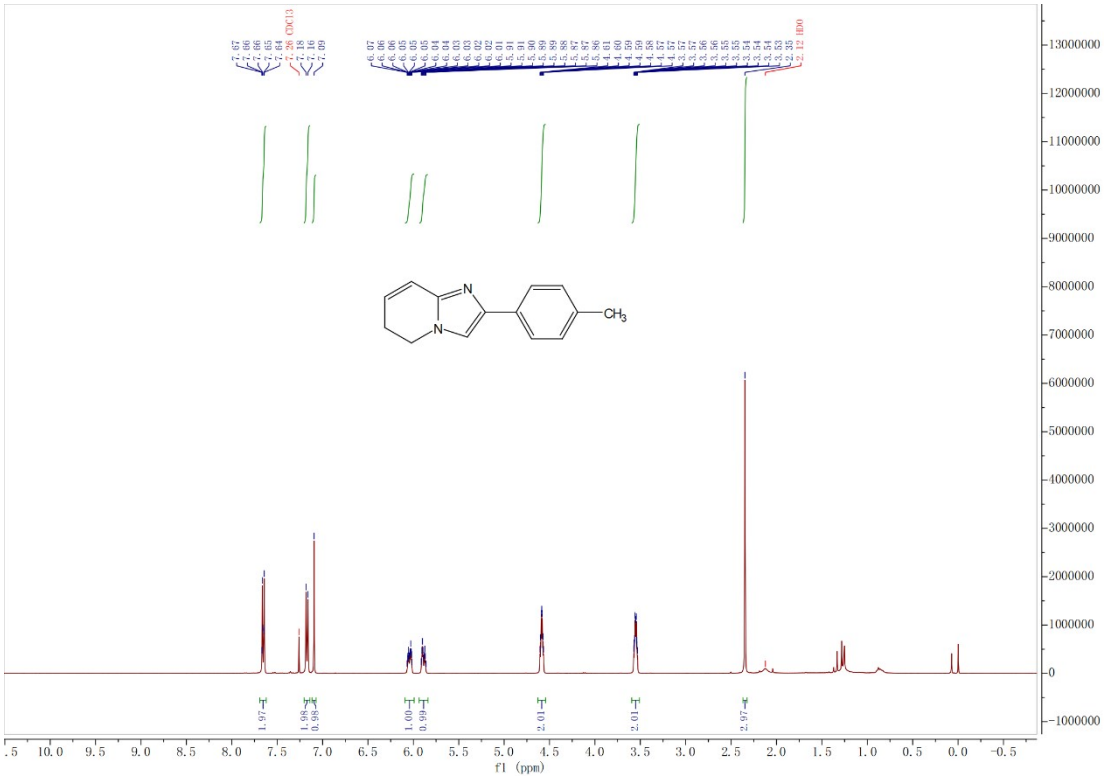
1.98
0.99
1.00
2.02
2.03
f1 (ppm)

3j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HDO
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N
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1.98
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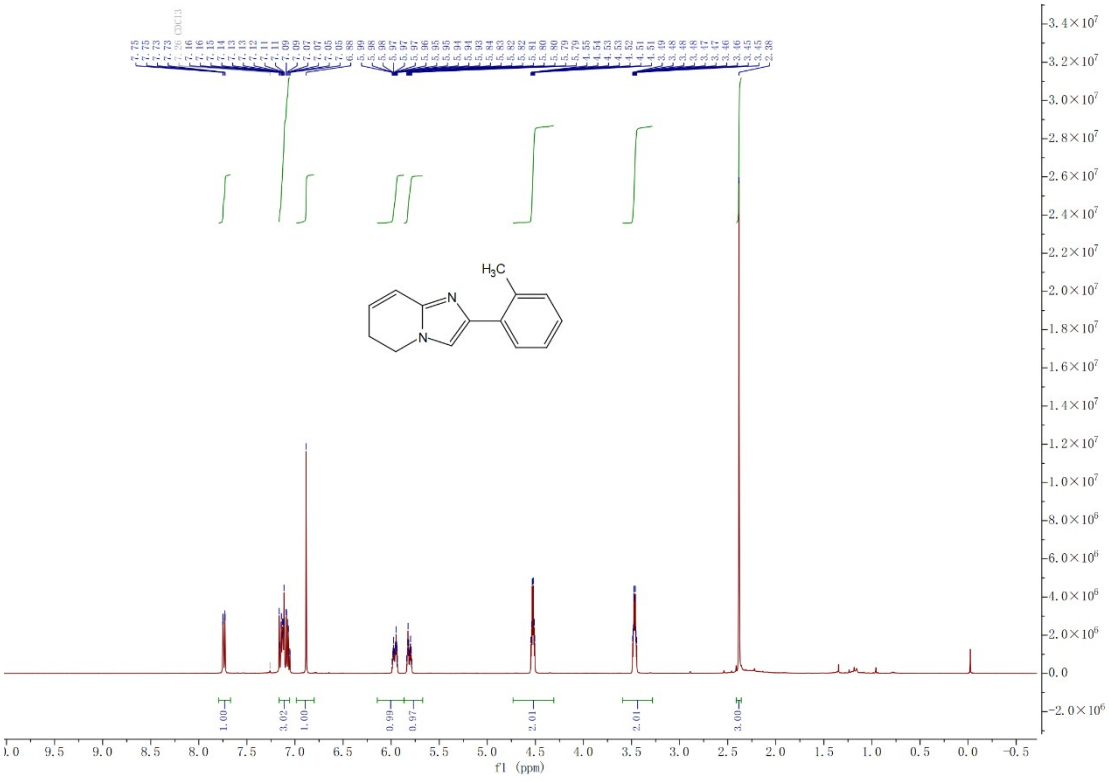
1.69
0.99
2.01
2.01
2.97
f1 (ppm)

3k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N
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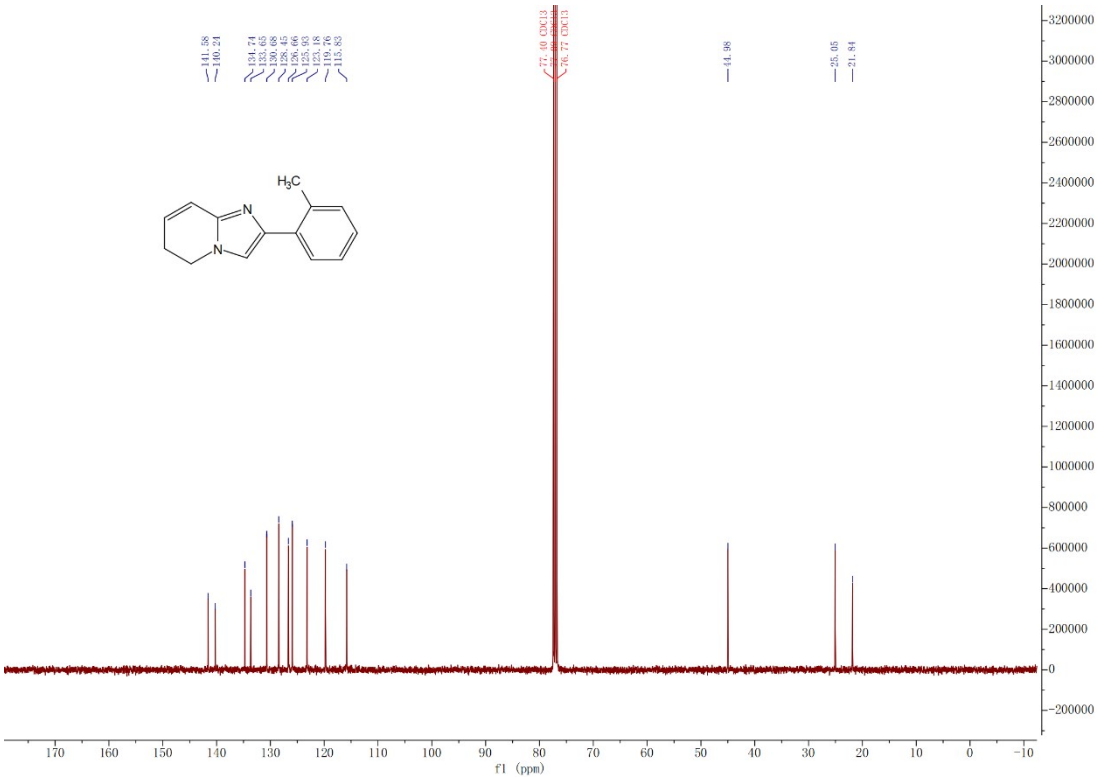
3.02
1.00
0.99
0.97
2.01
2.01
3.09
f1 (ppm)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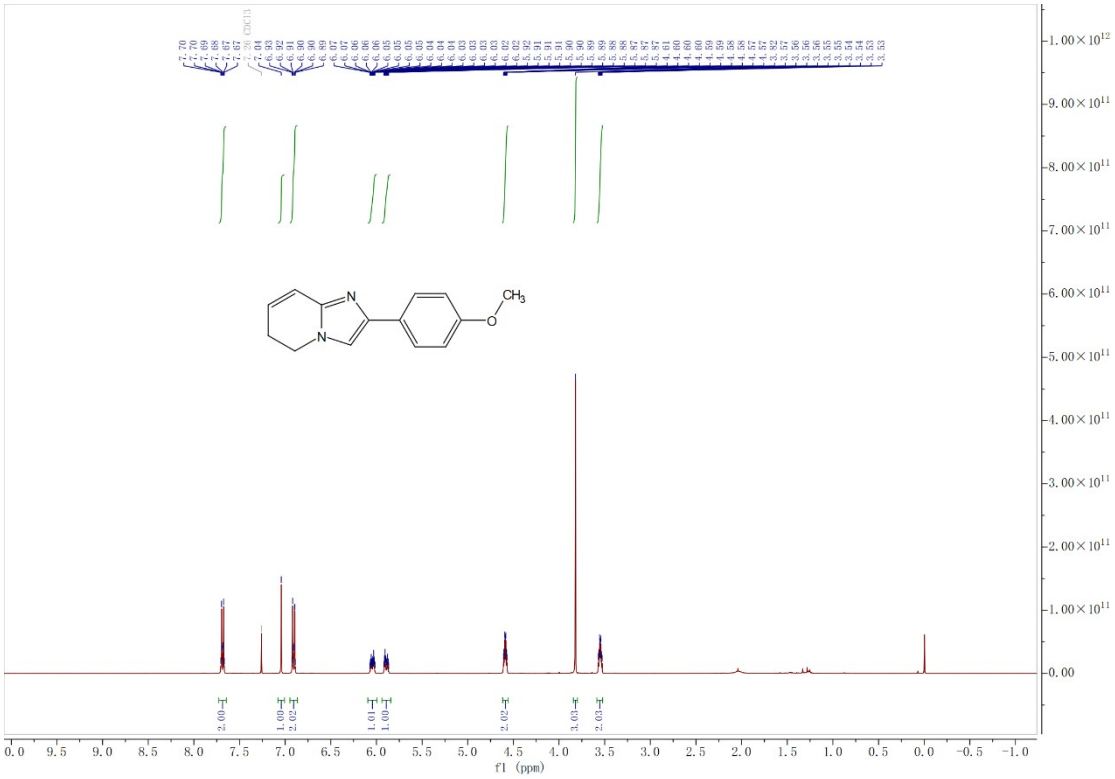
140.24
134.74
133.65
130.68
128.45
126.66
125.93
123.18
119.76
115.83
77.40 CDCl3
76.77 CDCl3
44.98
25.05
21.84
H3C
N
N
f1 (ppm)

3l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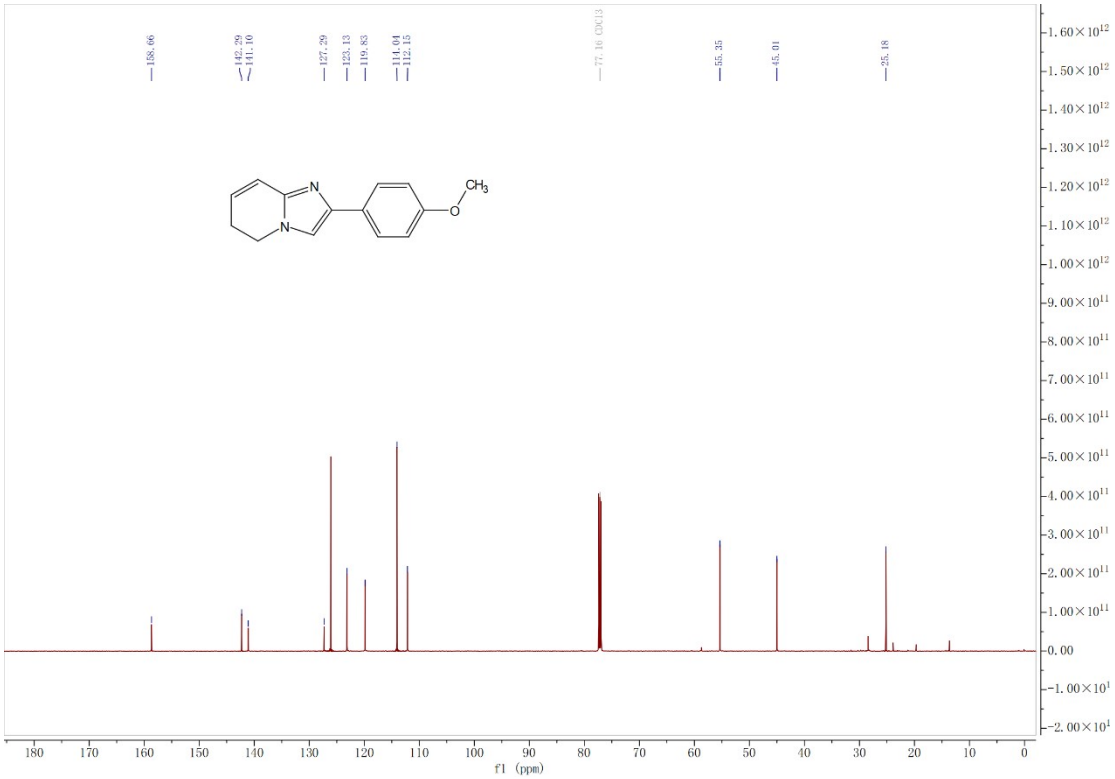
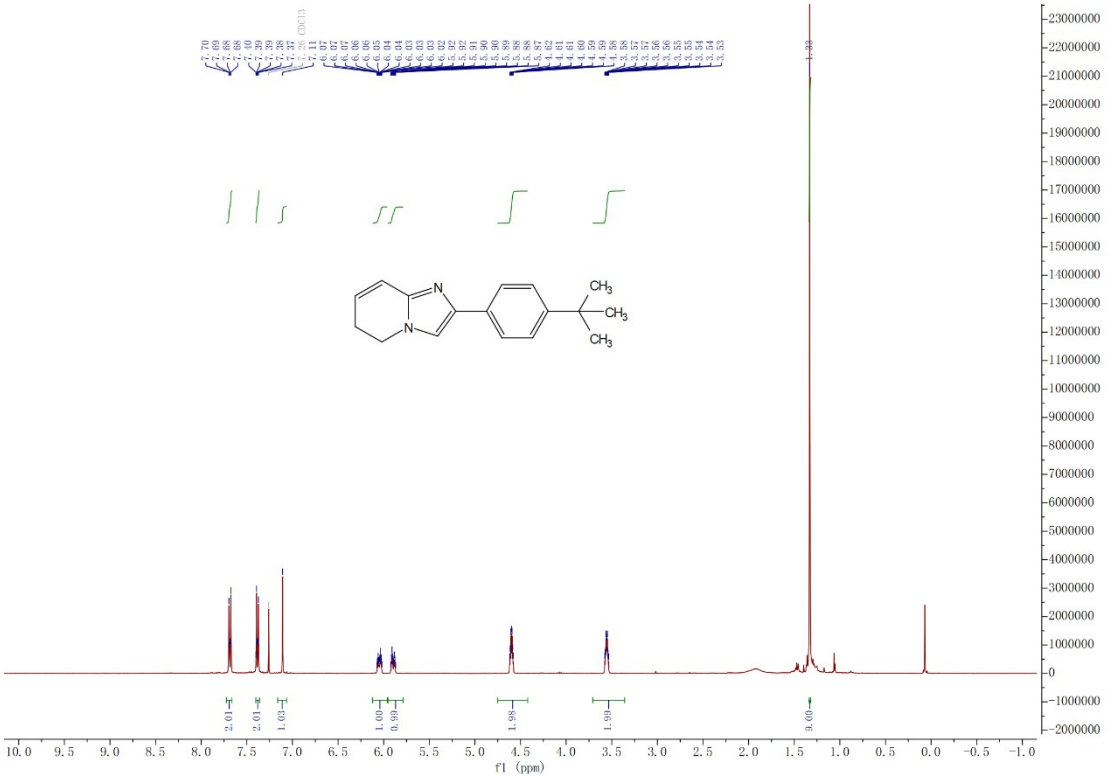
7.70
7.69
7.68
7.67
7.67
7.26 CDCl3
6.93
6.92
6.91
6.90
6.90
6.89
6.07
6.06
6.06
6.05
6.05
6.04
6.04
6.03
6.03
6.02
6.02
5.92
5.91
5.91
5.90
5.90
5.89
5.88
5.88
5.87
5.87
4.61
4.60
4.60
4.59
4.59
4.58
4.58
4.57
4.57
3.92
3.56
3.56
3.55
3.55
3.54
3.54
3.53
3.53
N
N
CH3
O
2.00
1.00
2.02
1.01
1.09
2.02
3.03
2.03
f1 (ppm)


158.66
142.29
141.10
127.29
123.13
119.83
114.04
112.15
77.16 CDCl3
55.35
45.01
25.18
N
N
CH3
O
f1 (ppm)

3m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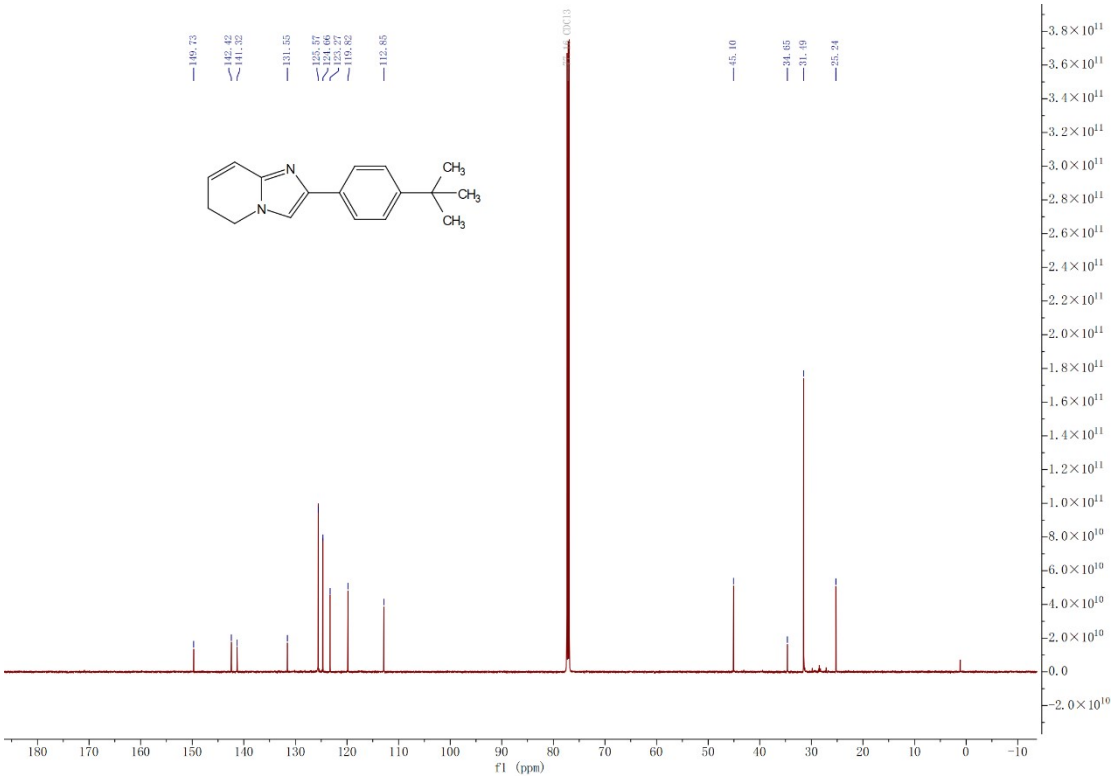
2.01
1.03
1.00
0.99
1.98
1.99
9.00
f1 (ppm)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123.37
119.82
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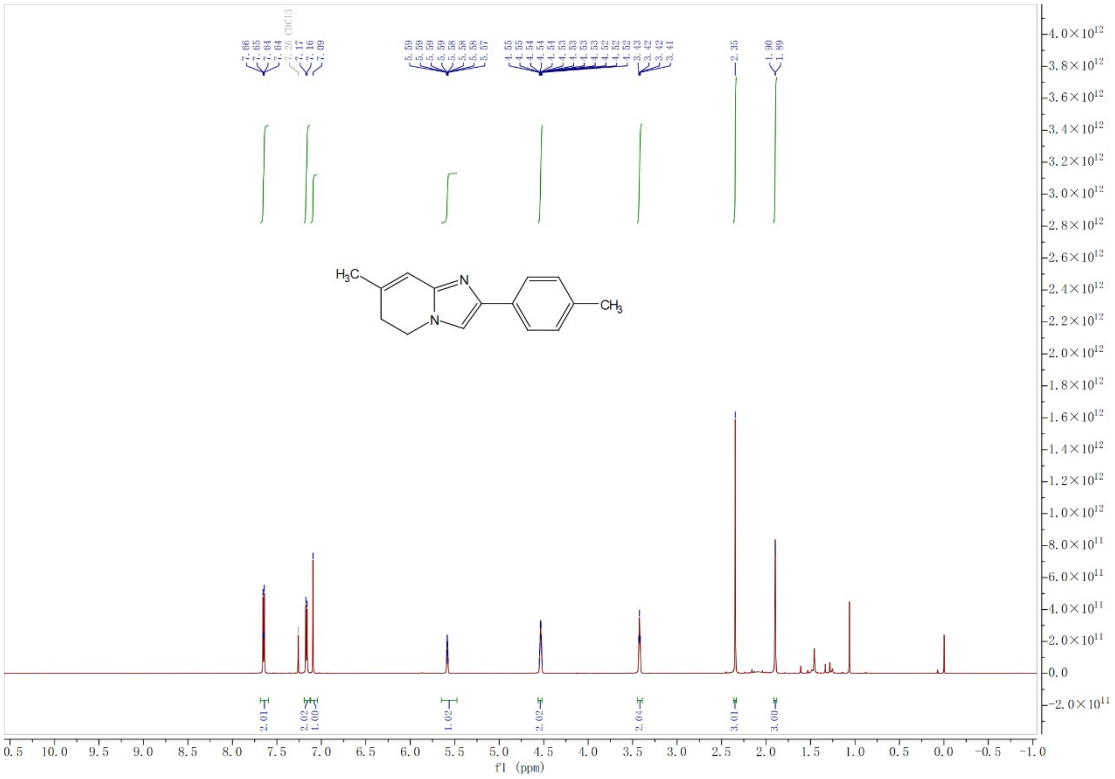
34.65
31.49
25.24
f1 (ppm)

3n



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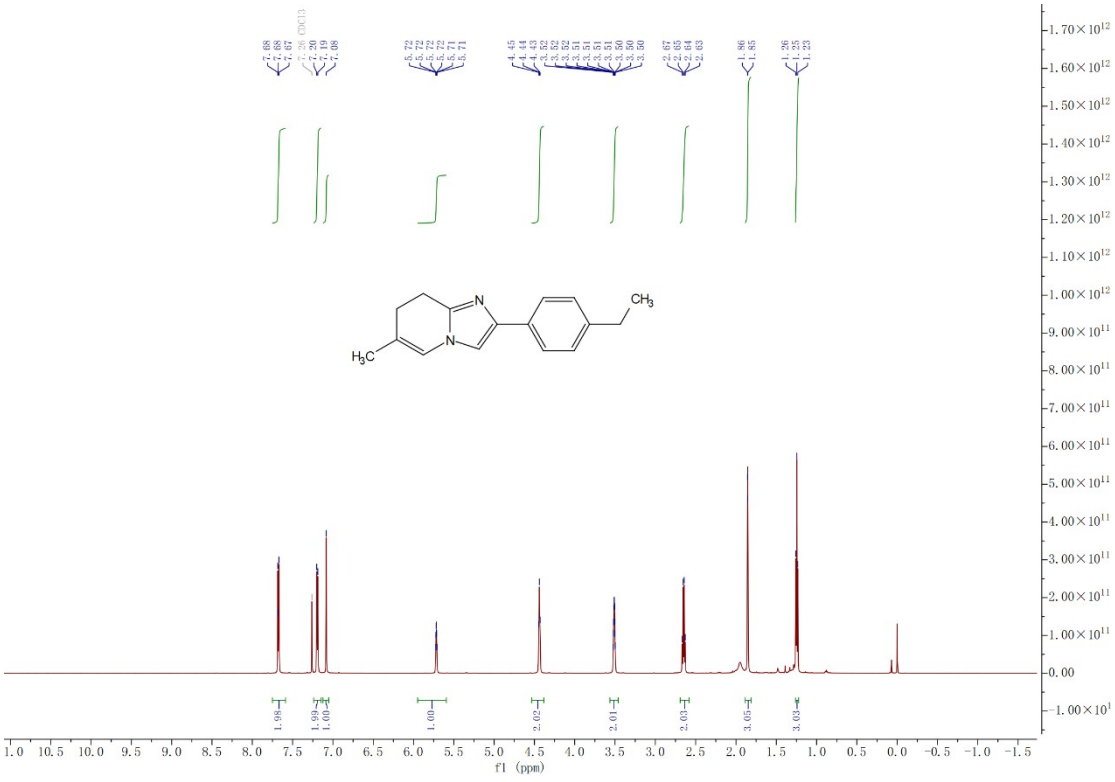

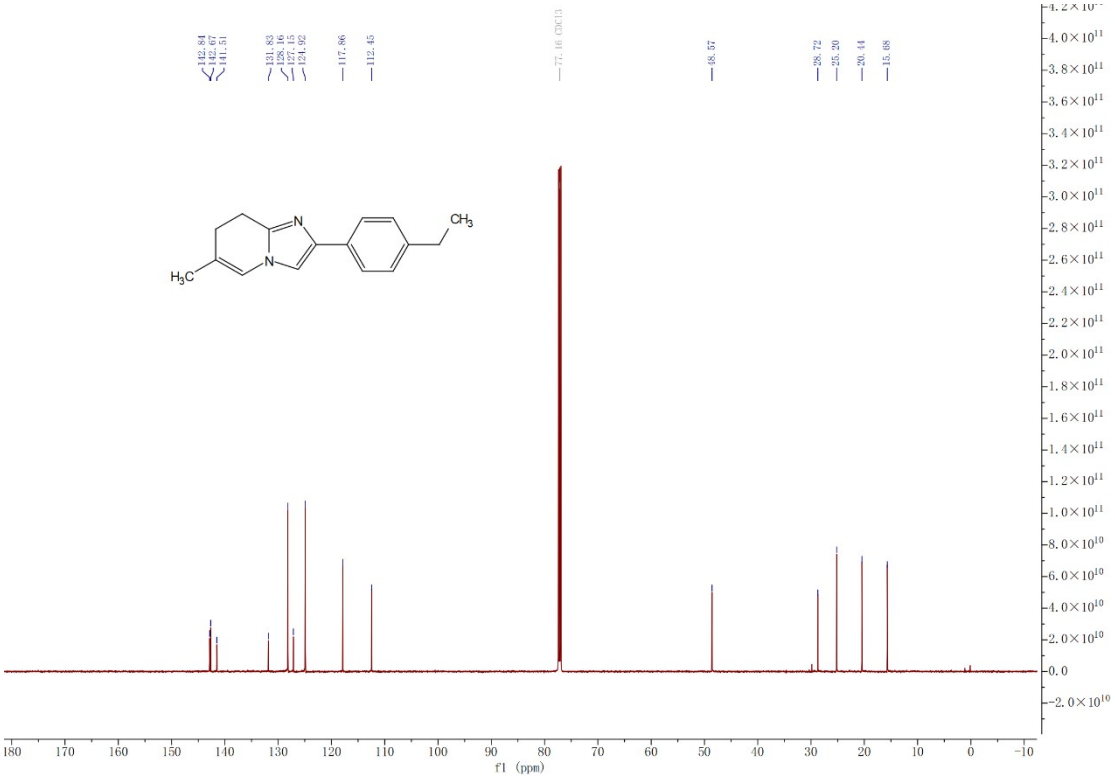
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124.92
117.86
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28.72
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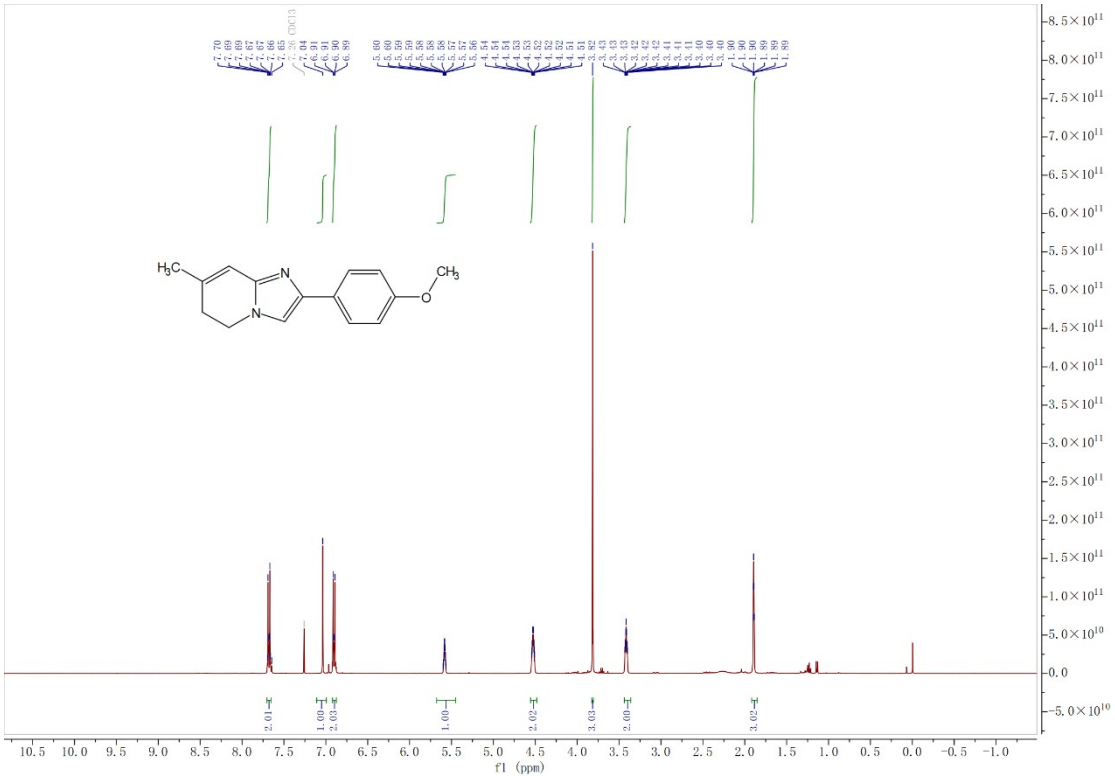
20.44
15.08
N
N
CH3
H3C
f1 (ppm)

3p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7.66
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6.91
6.91
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3.41
3.41
3.41
3.40
3.40
1.90
1.90
1.89
1.89
1.89
H3C
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O
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1.00
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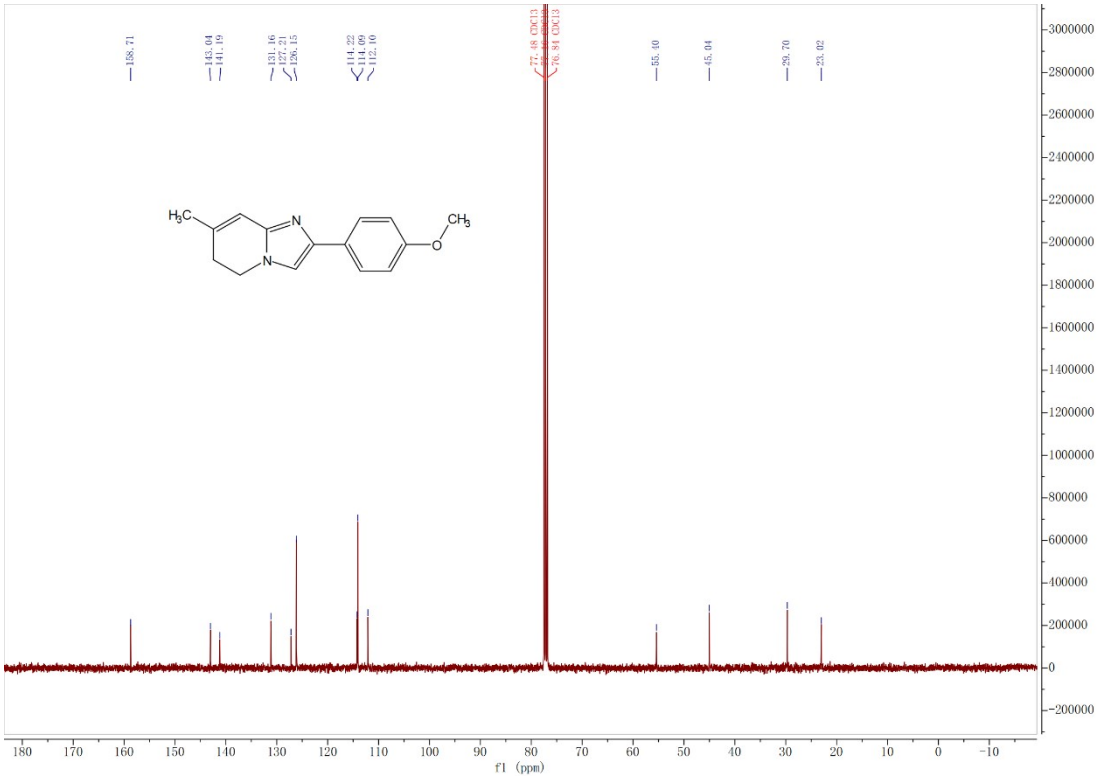
3.03
2.00
3.02
f1 (ppm)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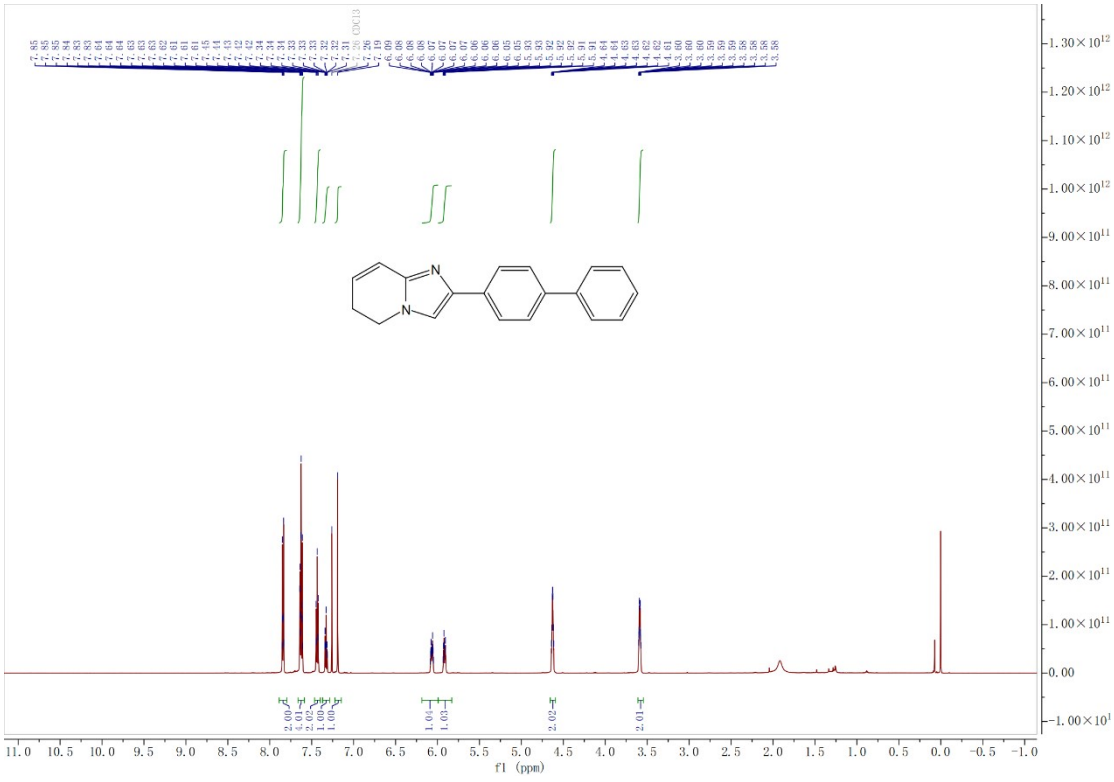
111.09
112.10
77.48 CDCl3
77.16 CDCl3
76.84 CDCl3
55.49
45.04
29.70
23.02
H3C
N
N
O
CH3
f1 (ppm)

3q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7.63
7.62
7.61
7.61
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7.45
7.44
7.43
7.42
7.34
7.34
7.34
7.33
7.33
7.33
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7.32
7.31
7.26 CDCl3
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6.06
6.06
6.05
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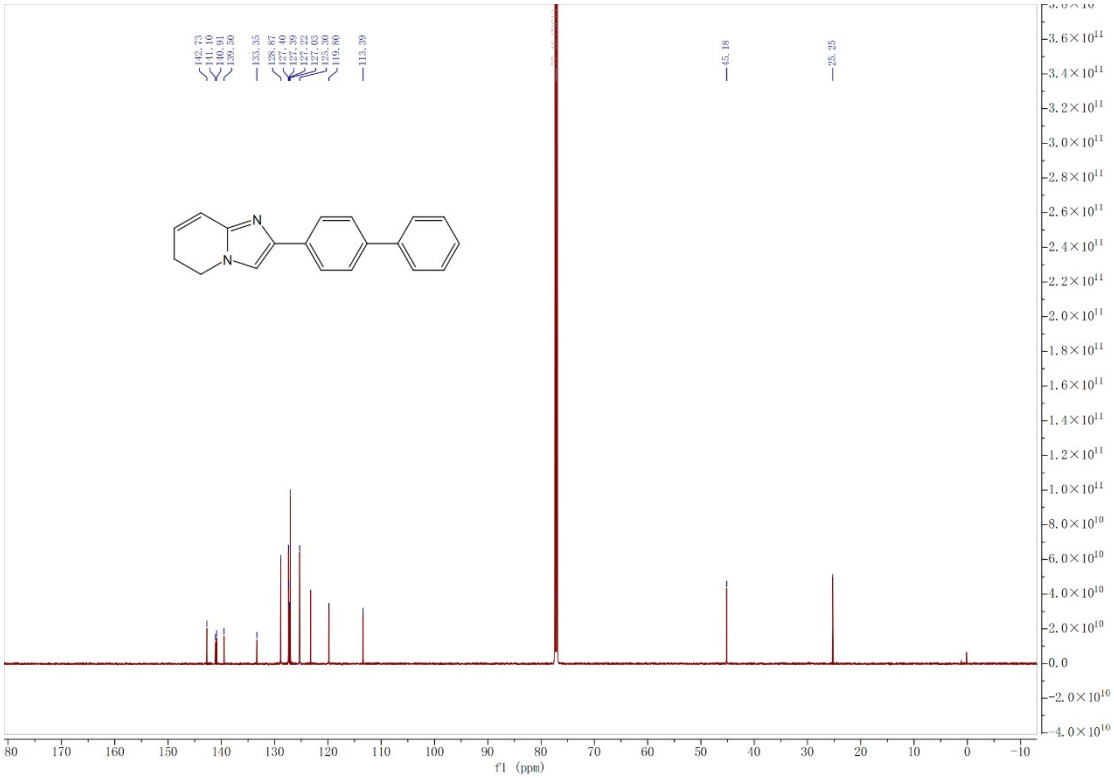
2.01
f1 (ppm)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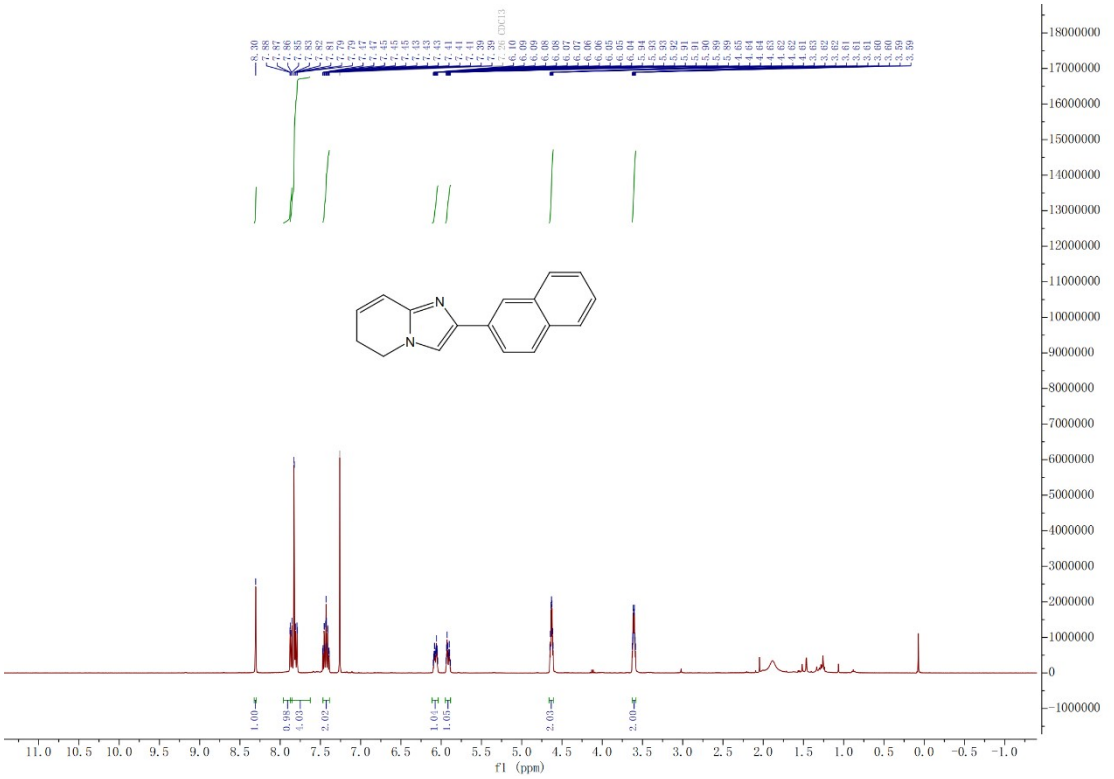
f1 (ppm)

3r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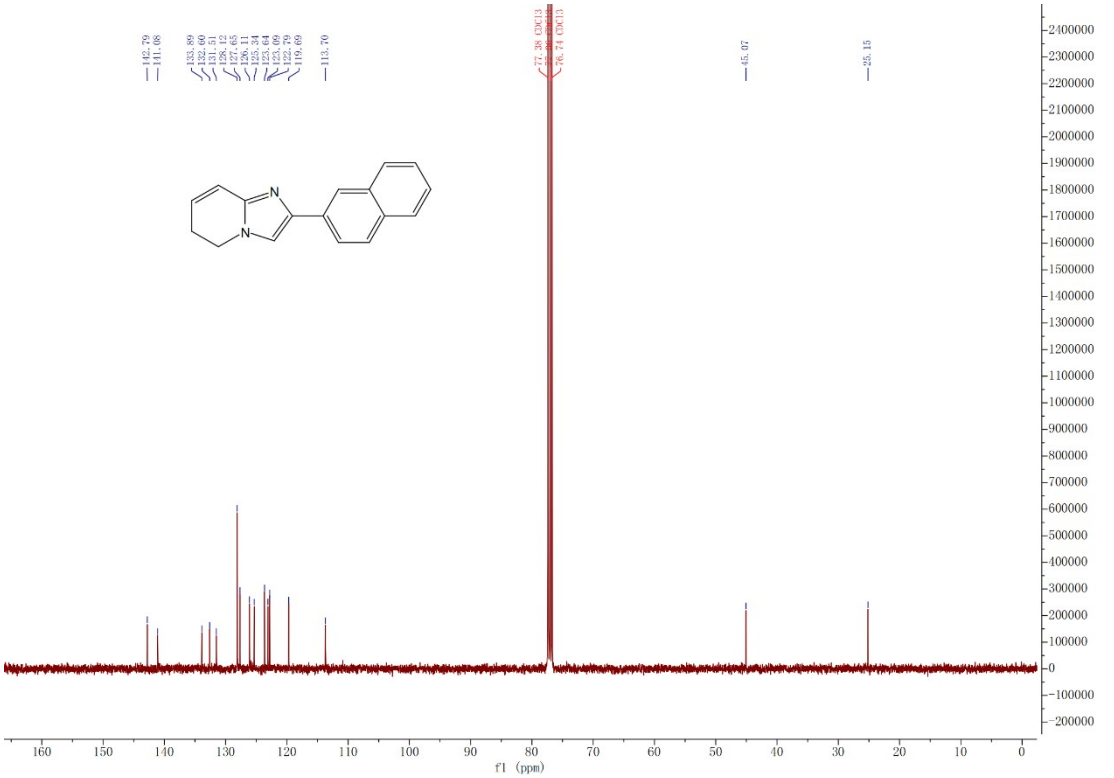

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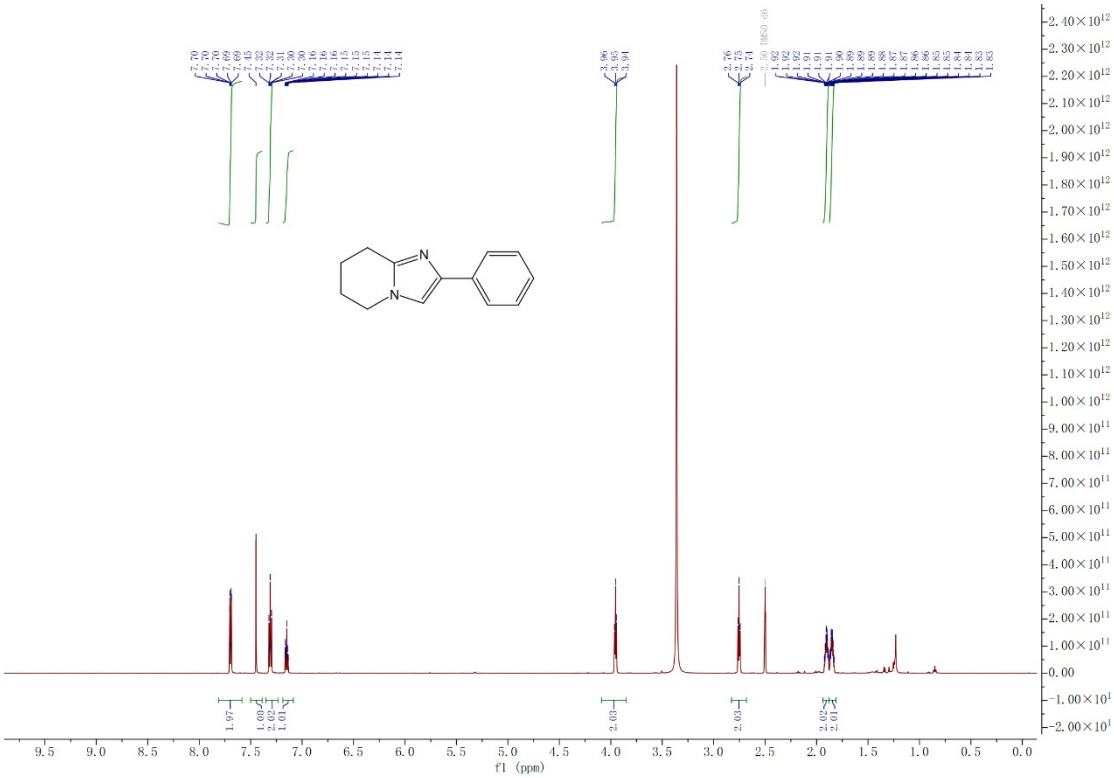

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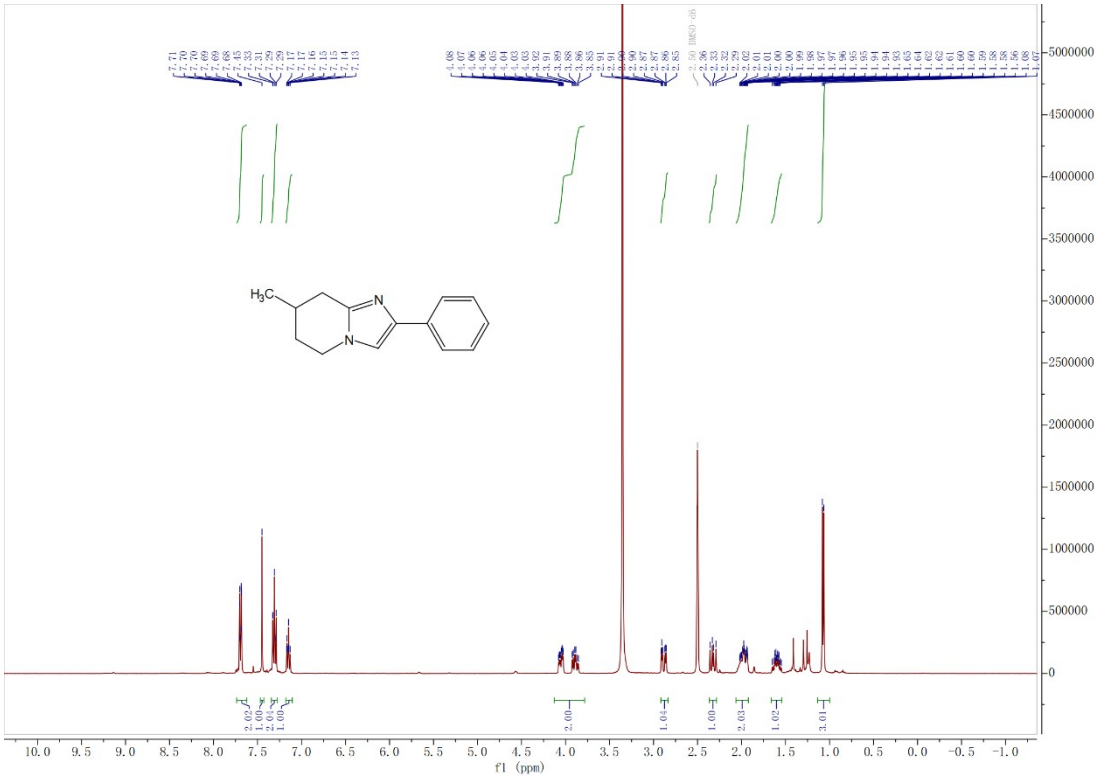
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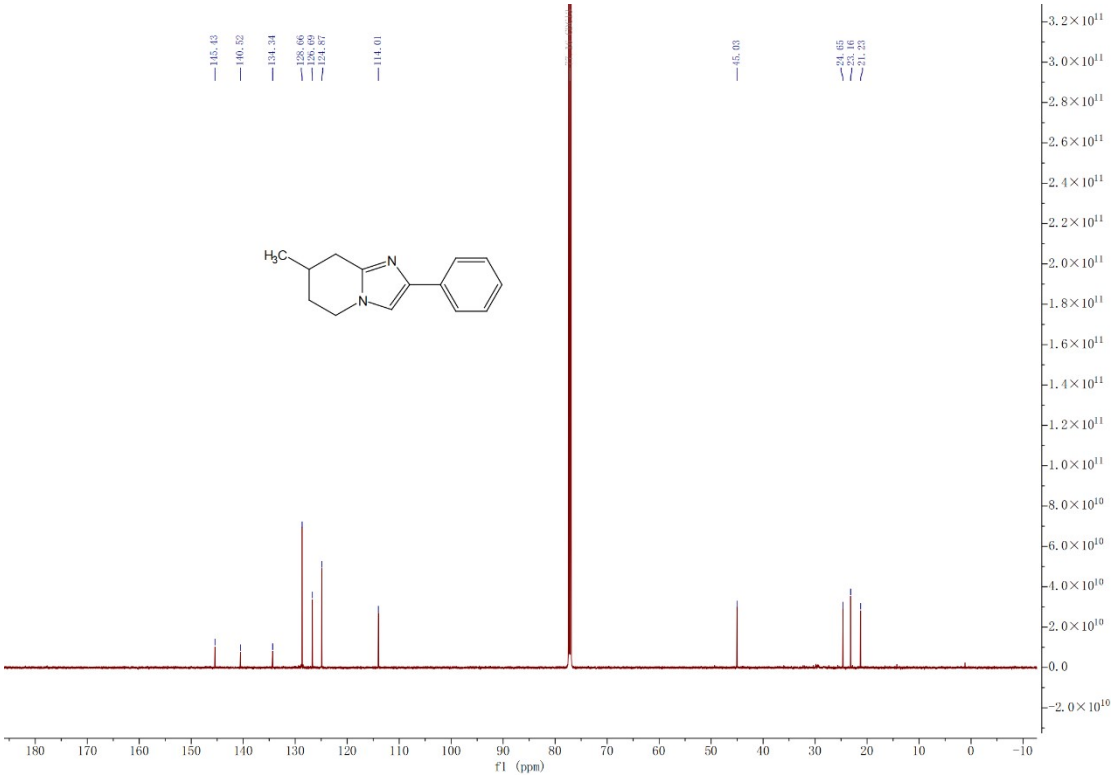
N
N
f1 (ppm)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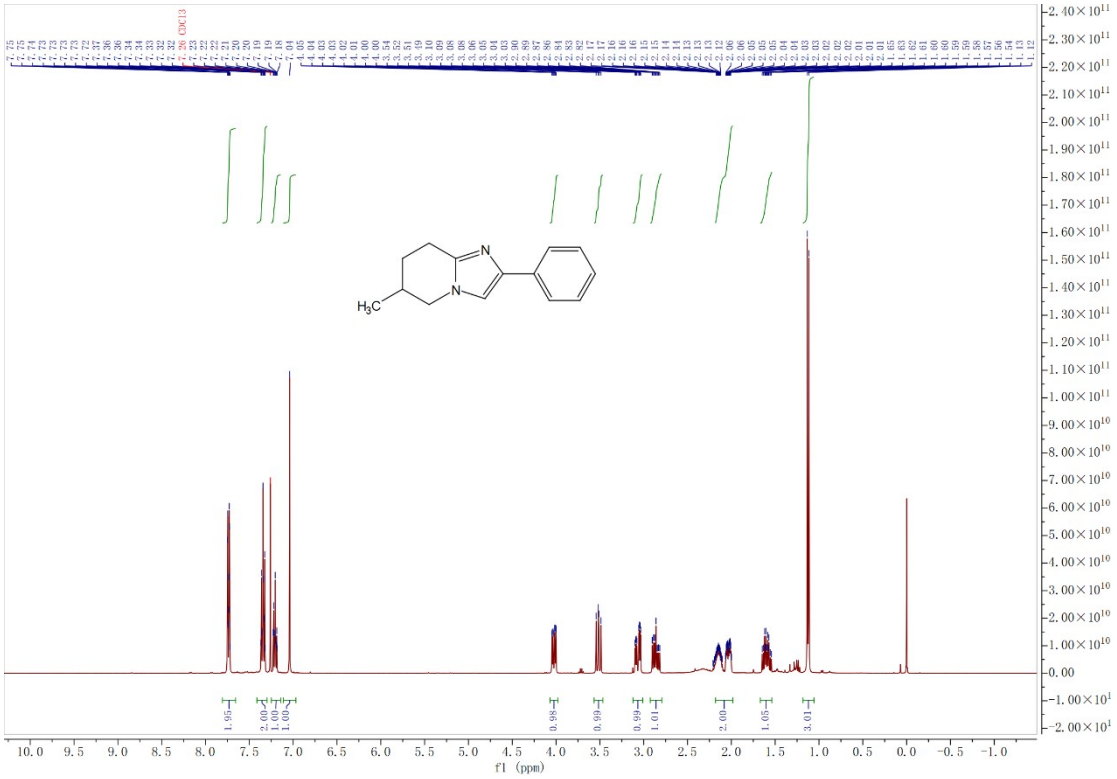
N
N
f1 (ppm)

4c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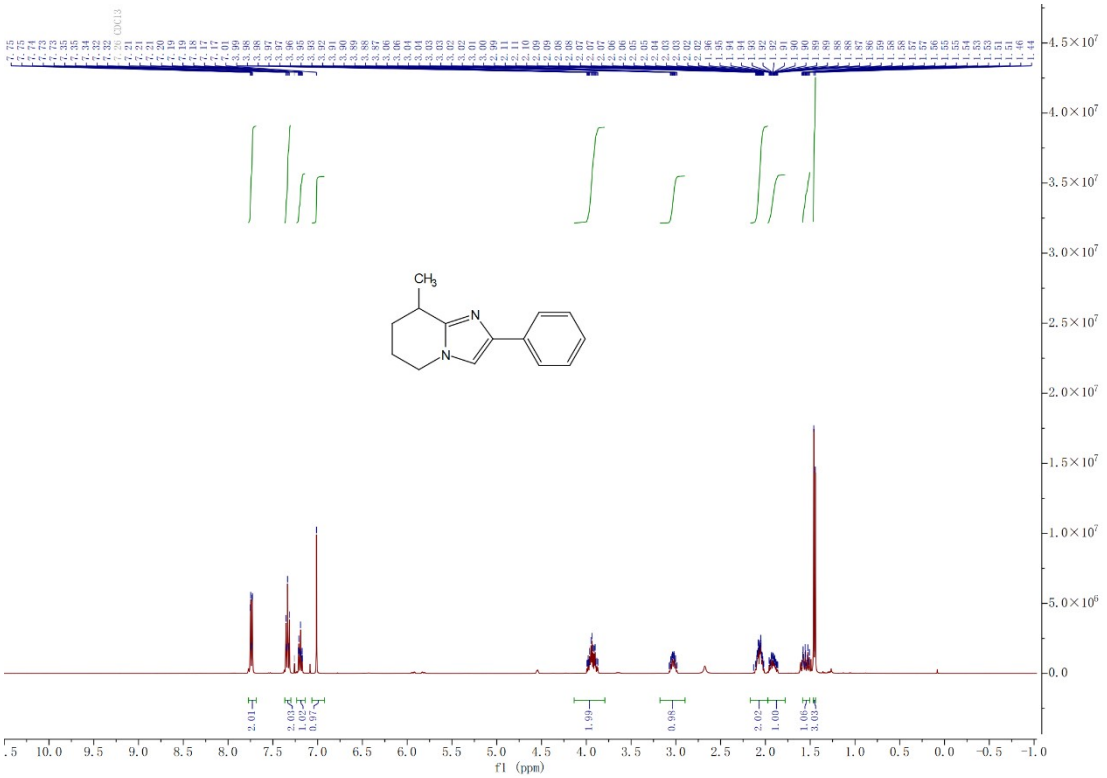
f1 (ppm)

4d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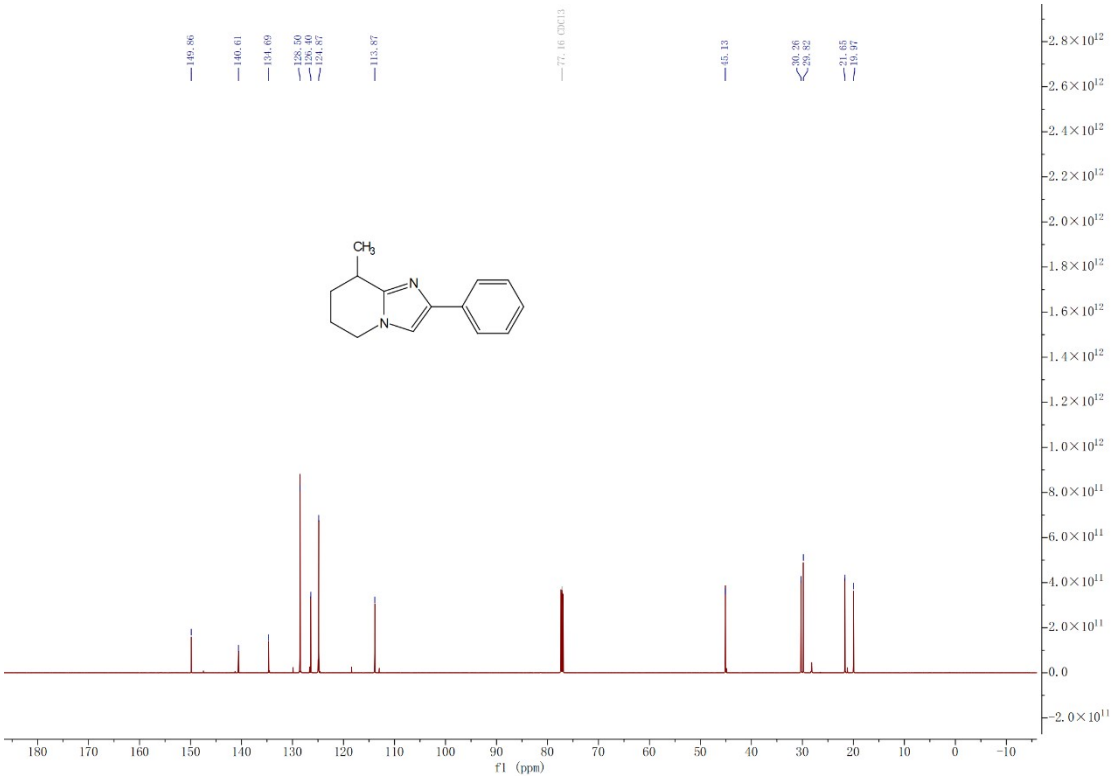
1.00
3.03
f1 (ppm)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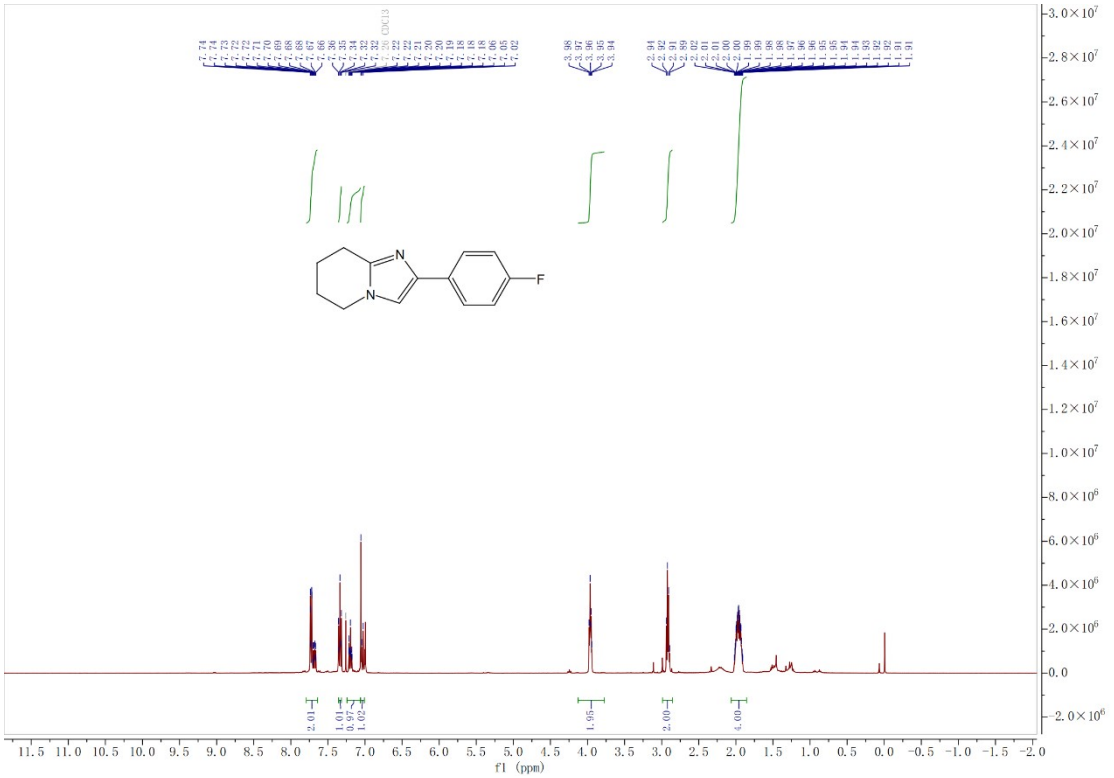
N
f1 (ppm)

4f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7.32
7.30 CDCl3
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7.21
7.20
7.19
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7.05
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1.95
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1.93
1.92
1.92
1.91
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N
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F
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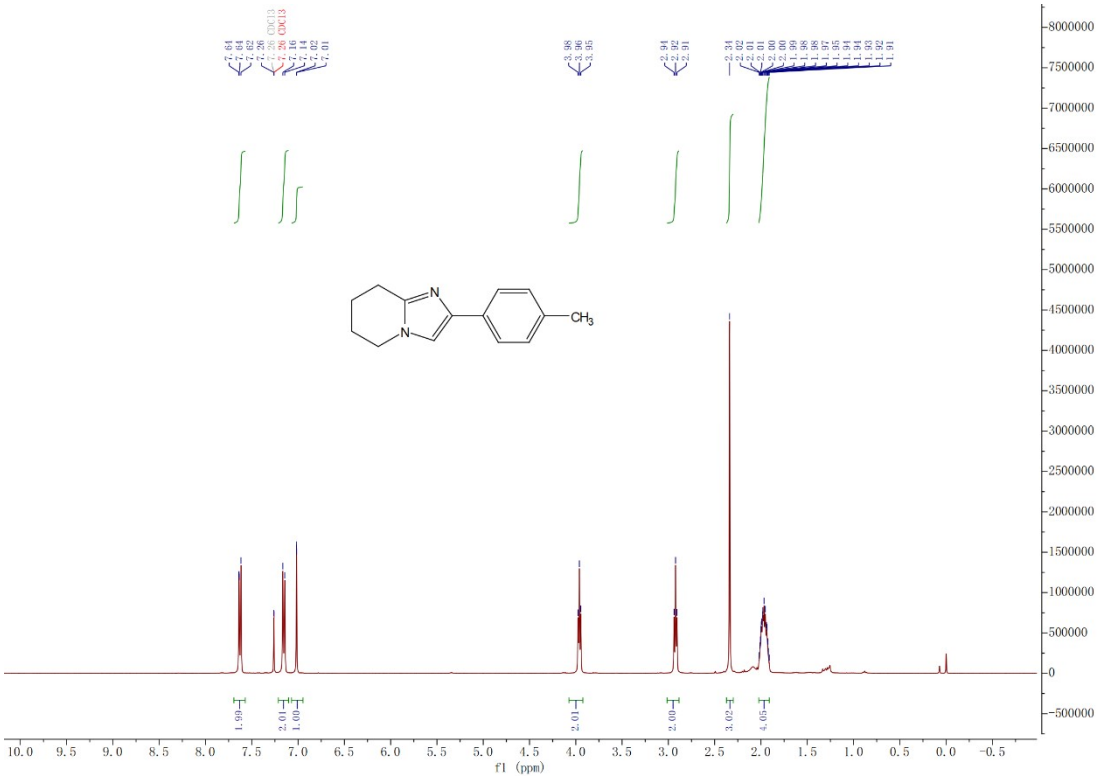
4.00
f1 (ppm)

4j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CDCl3
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7.01
3.98
3.96
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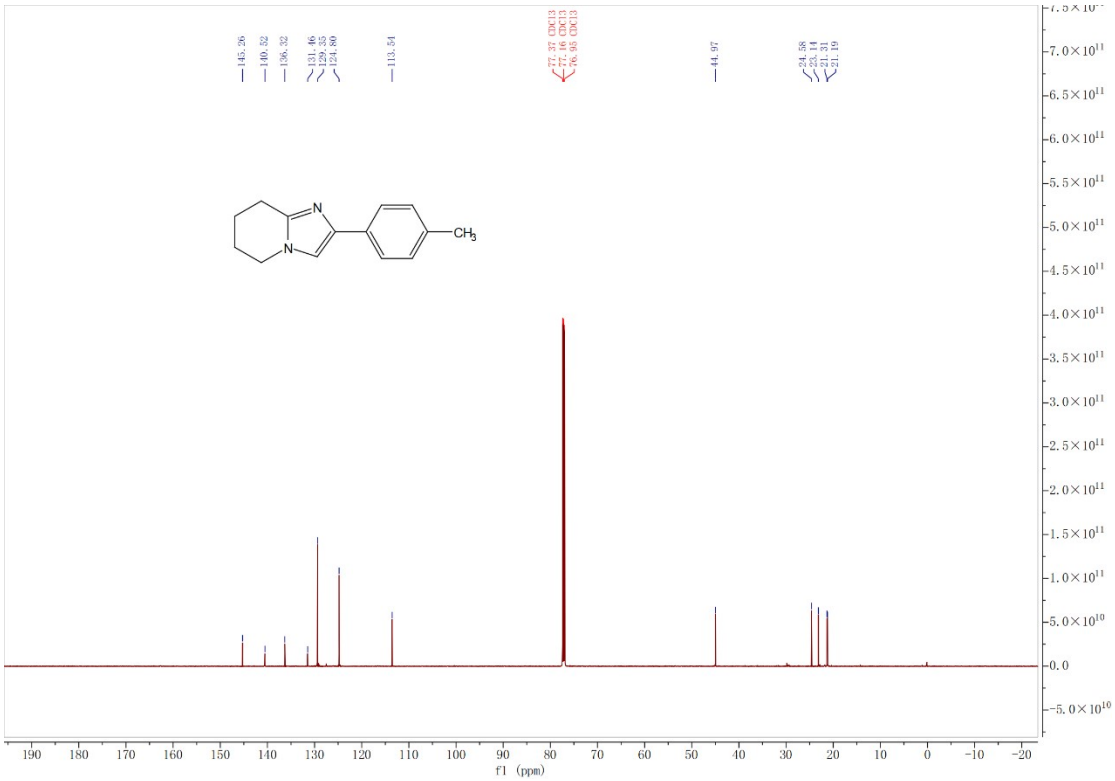
3.02
4.05
f1 (ppm)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124.80
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77.37
CDCl3
77.06
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CDCl3
44.97
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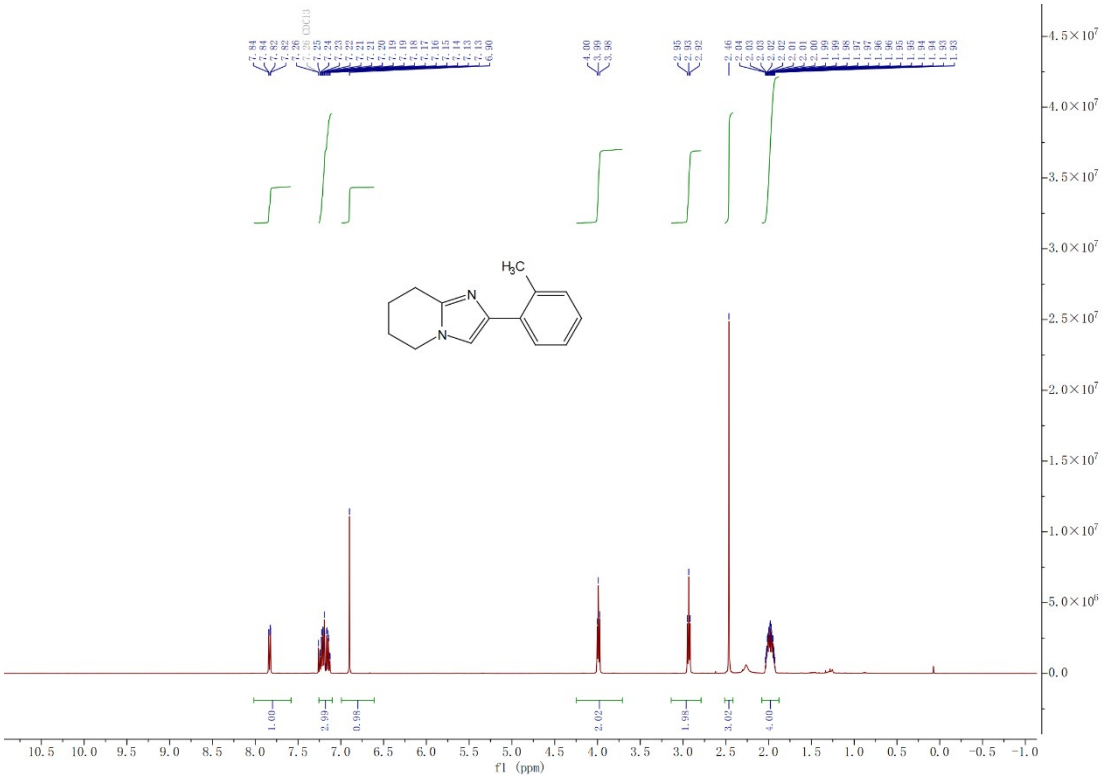
21.19
N
N
CH3
f1 (ppm)

4k



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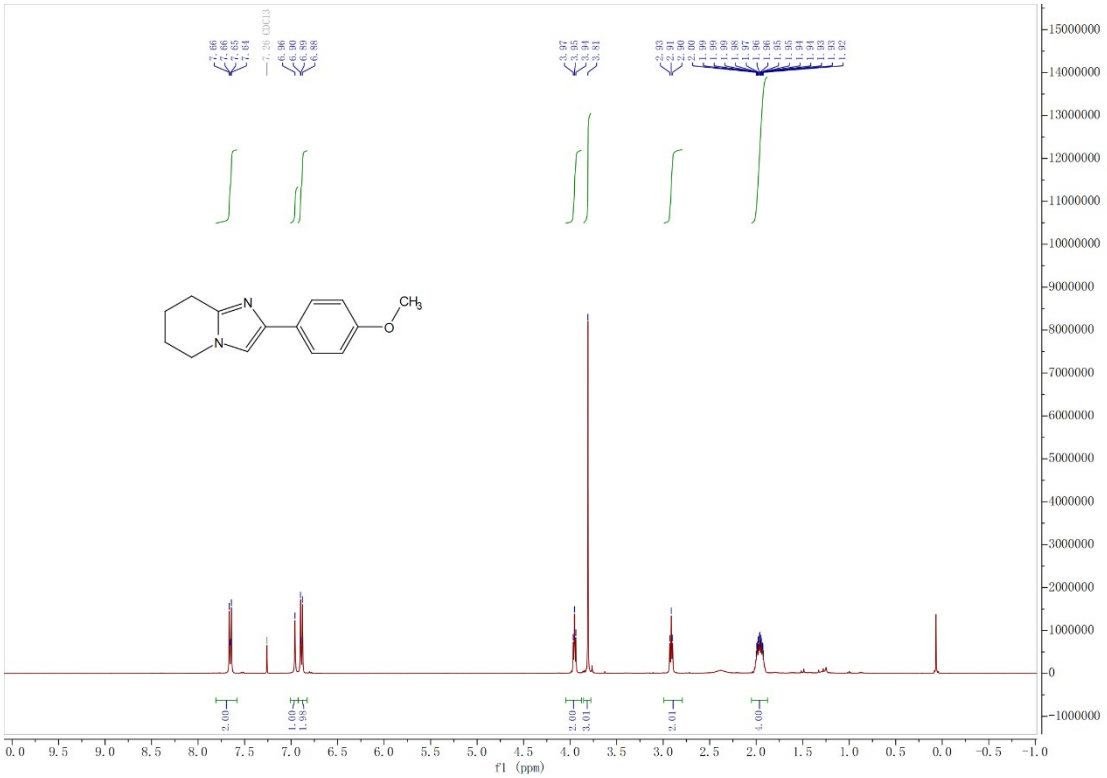

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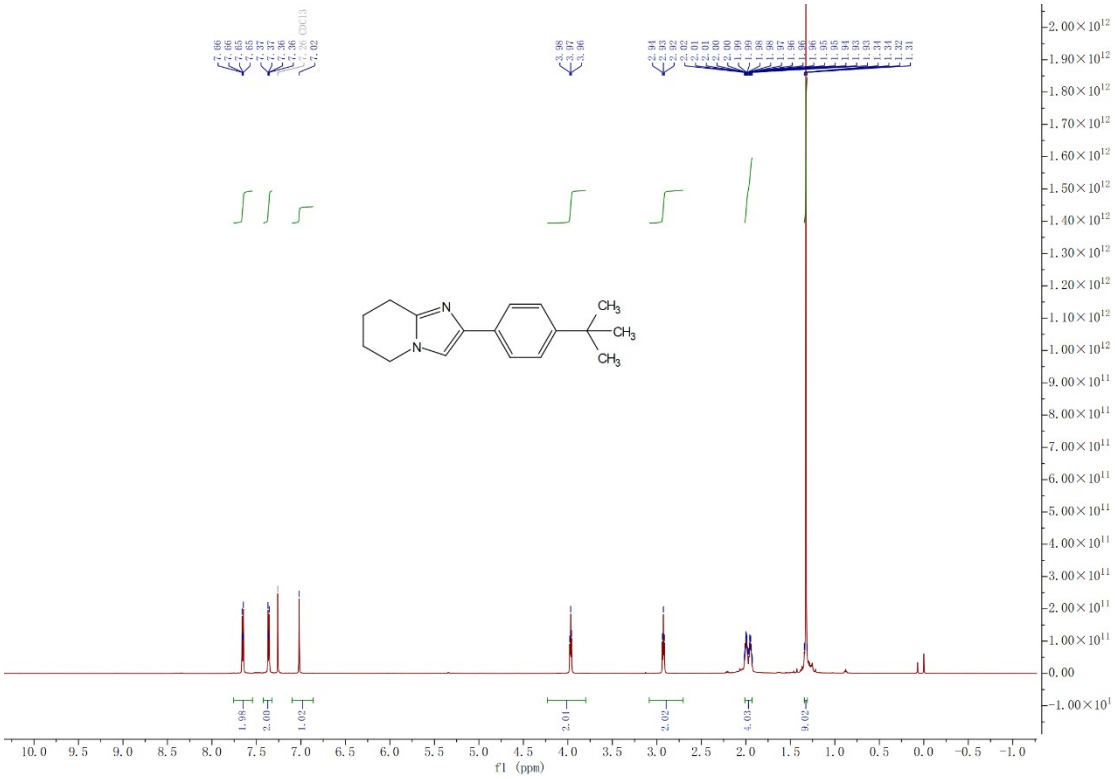
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7.85
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2.94
2.93
2.92
2.92
2.91
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2.90
2.90
2.89
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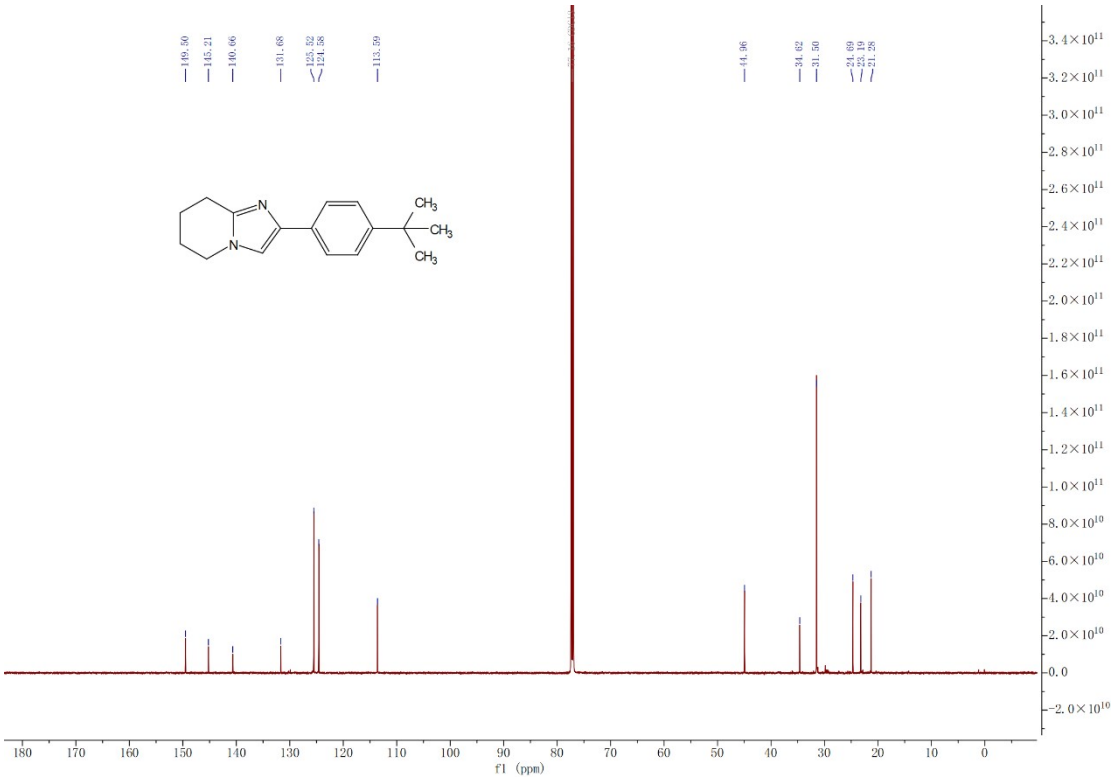
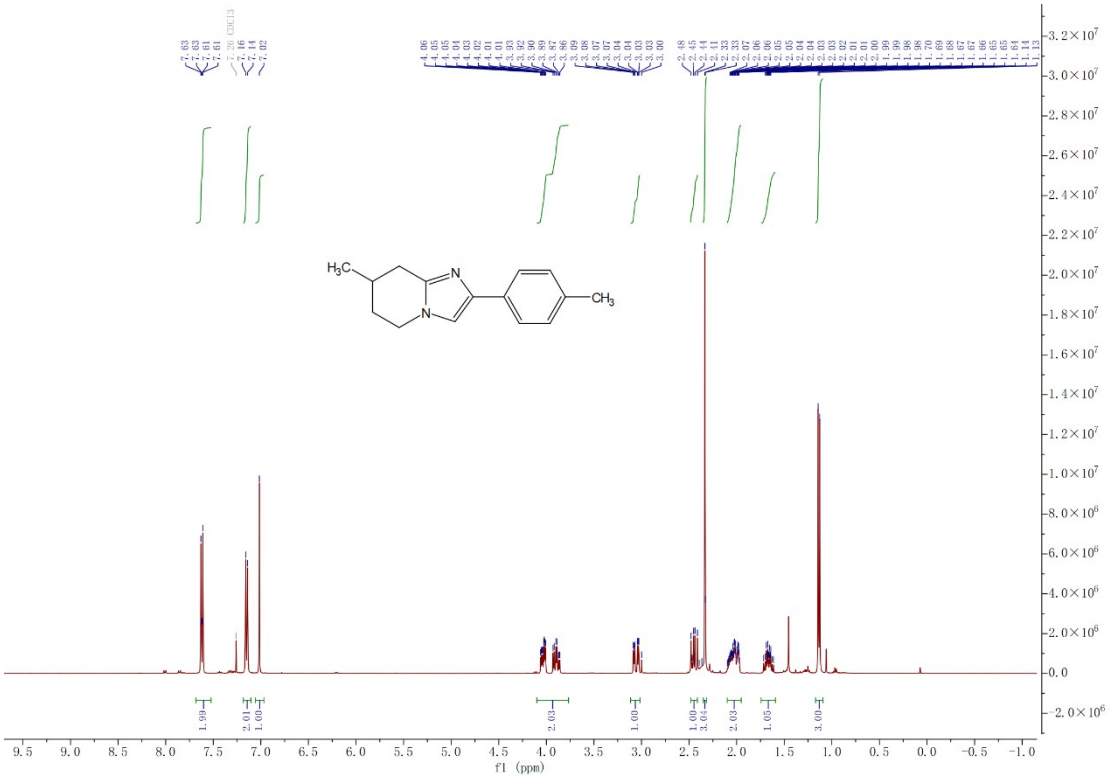
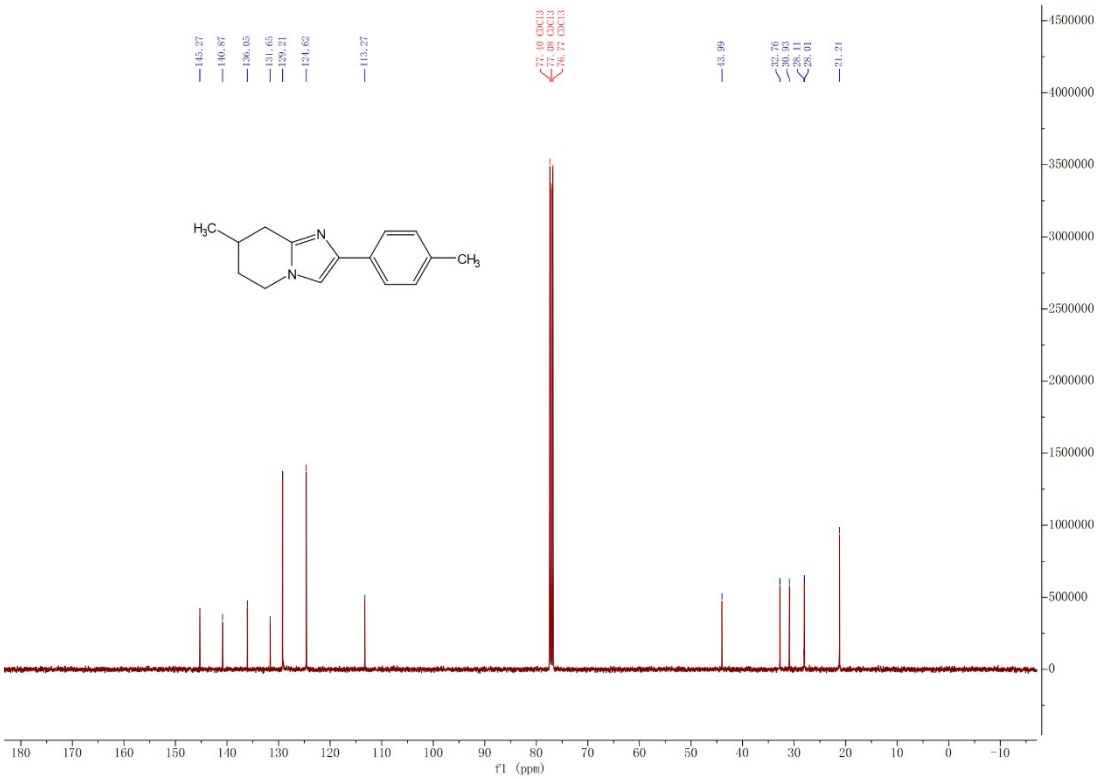
1.99
1.98
1.98
1.97
1.97
1.96
1.96
1.95
1.95
1.94
1.93
1.93
1.34
1.34
1.32
1.31
N
N
CH3
CH3
CH3
1.98
2.00
1.02
2.01
2.02
4.03
9.02
f1 (ppm)

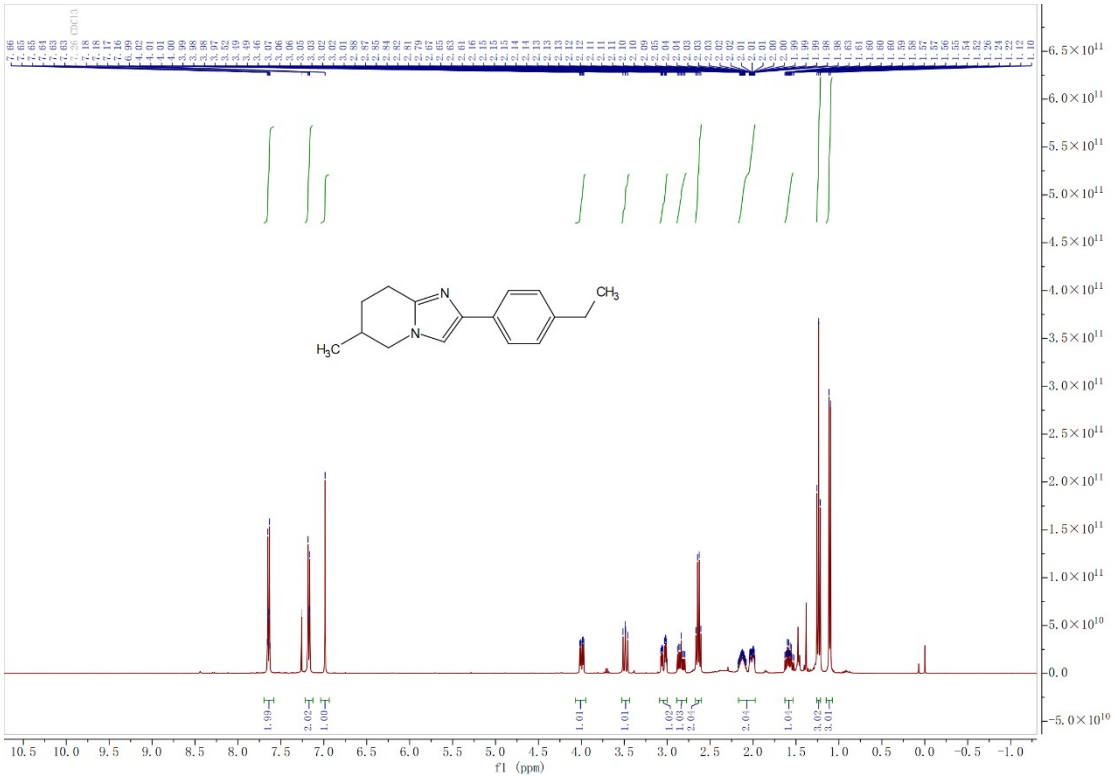

149.50
145.21
140.66
131.68
125.52
124.58
113.59
77.36
77.04
76.73
44.96
34.62
31.56
24.69
23.19
21.28
N
N
CH3
CH3
CH3
f1 (ppm)

4n

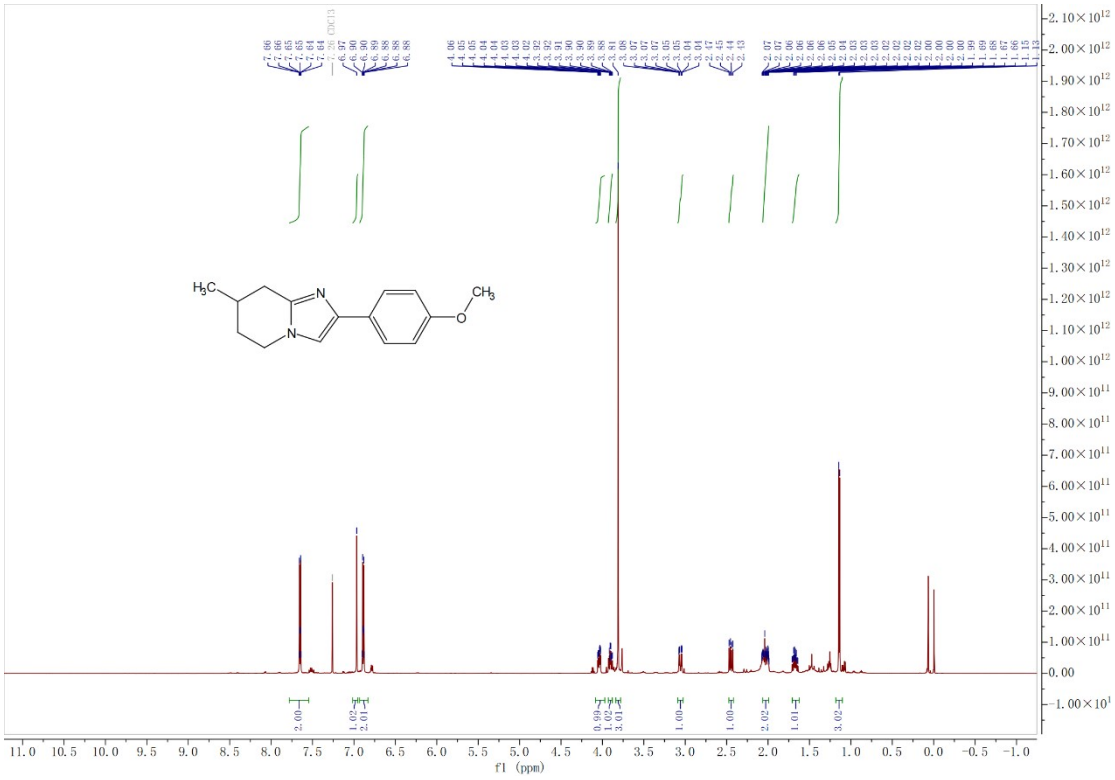

H3C
N
N
CH3
f1 (ppm)

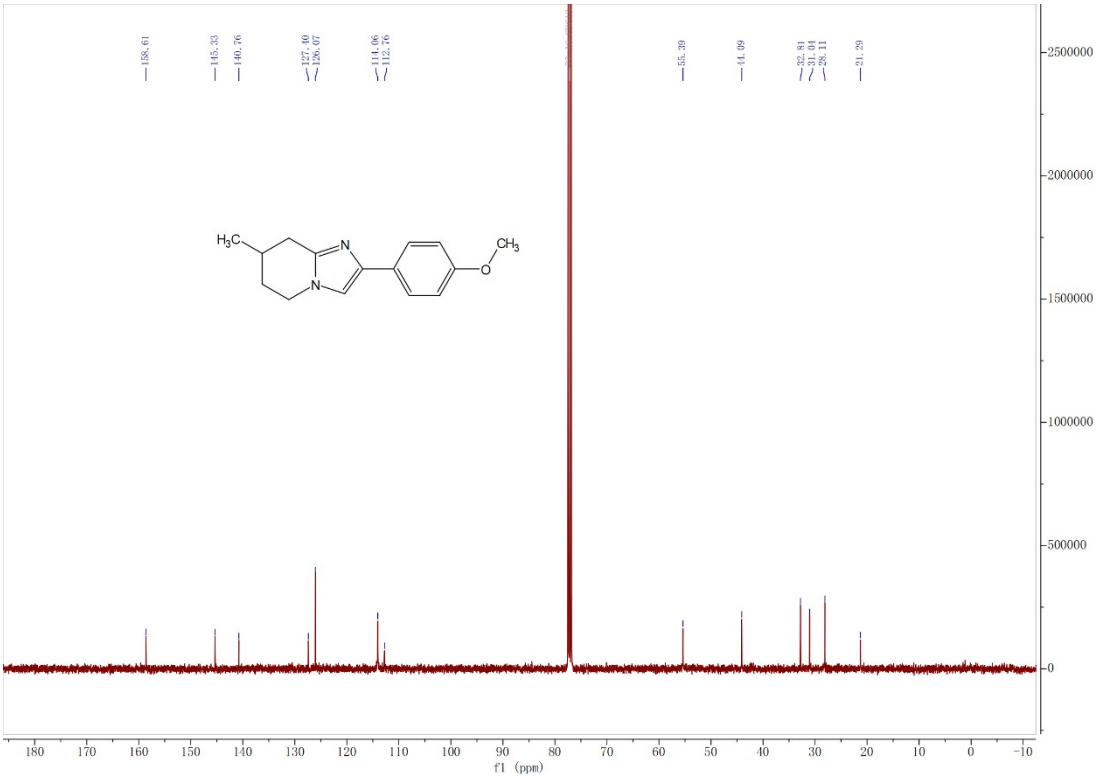

145.27
140.87
136.05
131.65
129.21
124.62
113.27
77.10 CDCl3
77.48 CDCl3
76.77 CDCl3
43.99
32.76
30.93
28.11
28.01
21.21
H3C
N
N
CH3
f1 (ppm)

4o

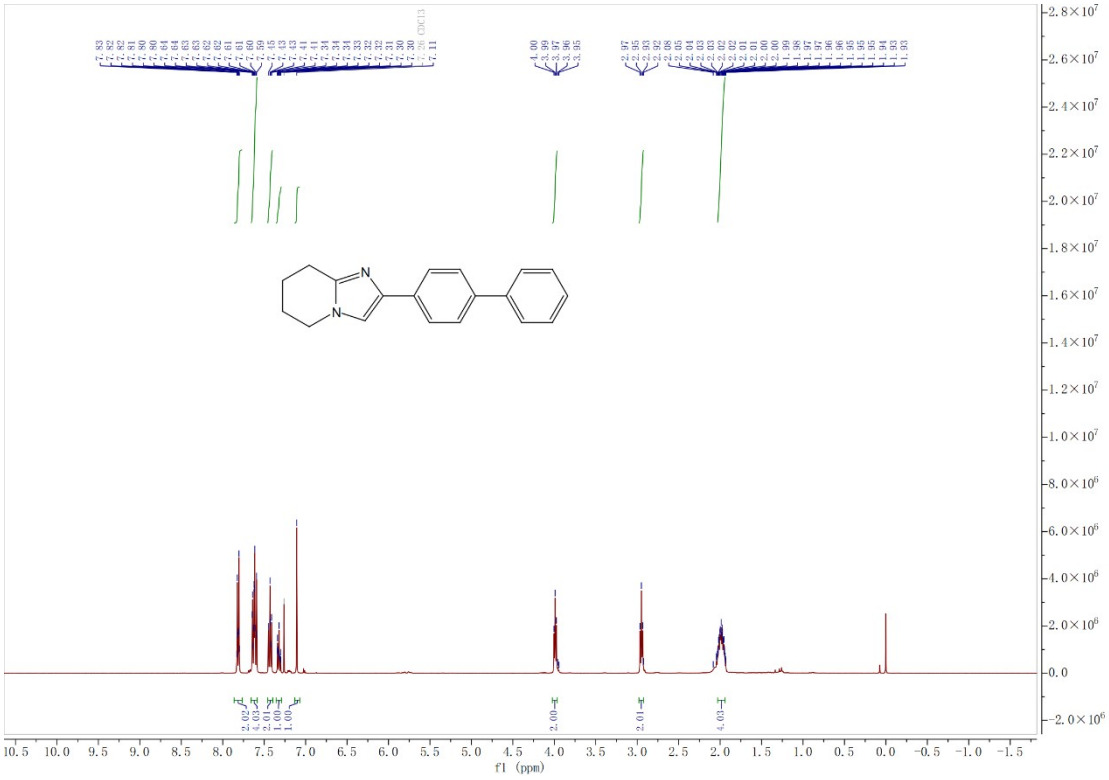


4p

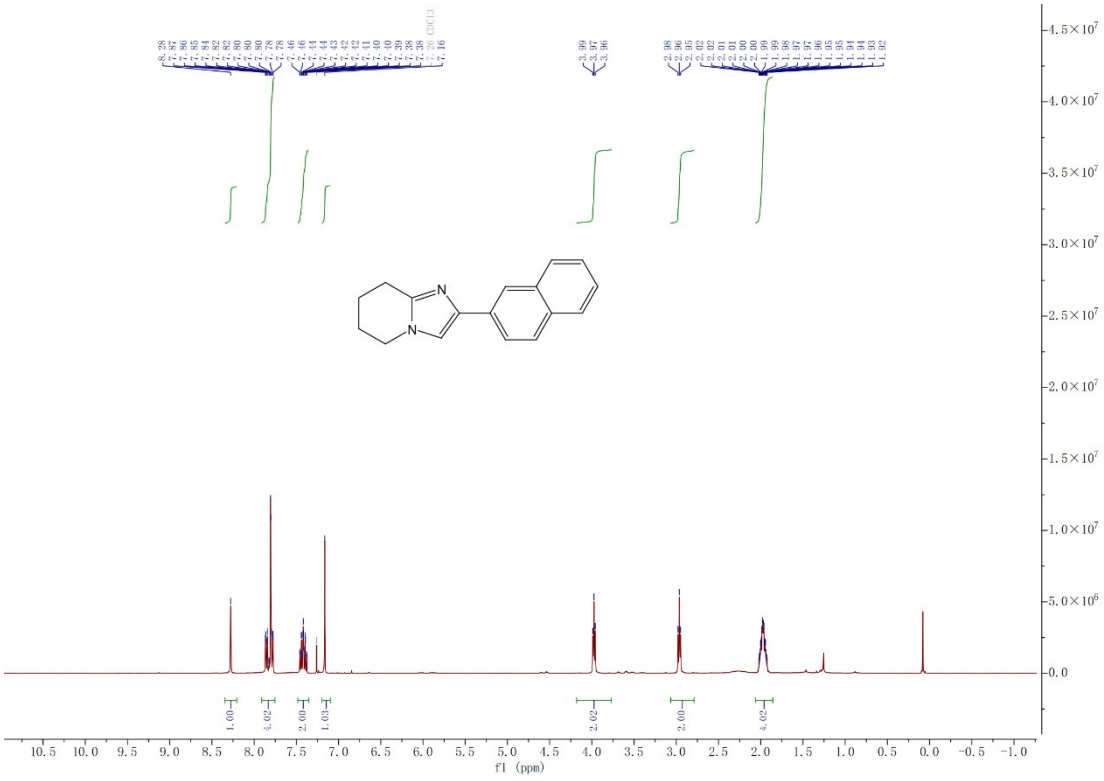


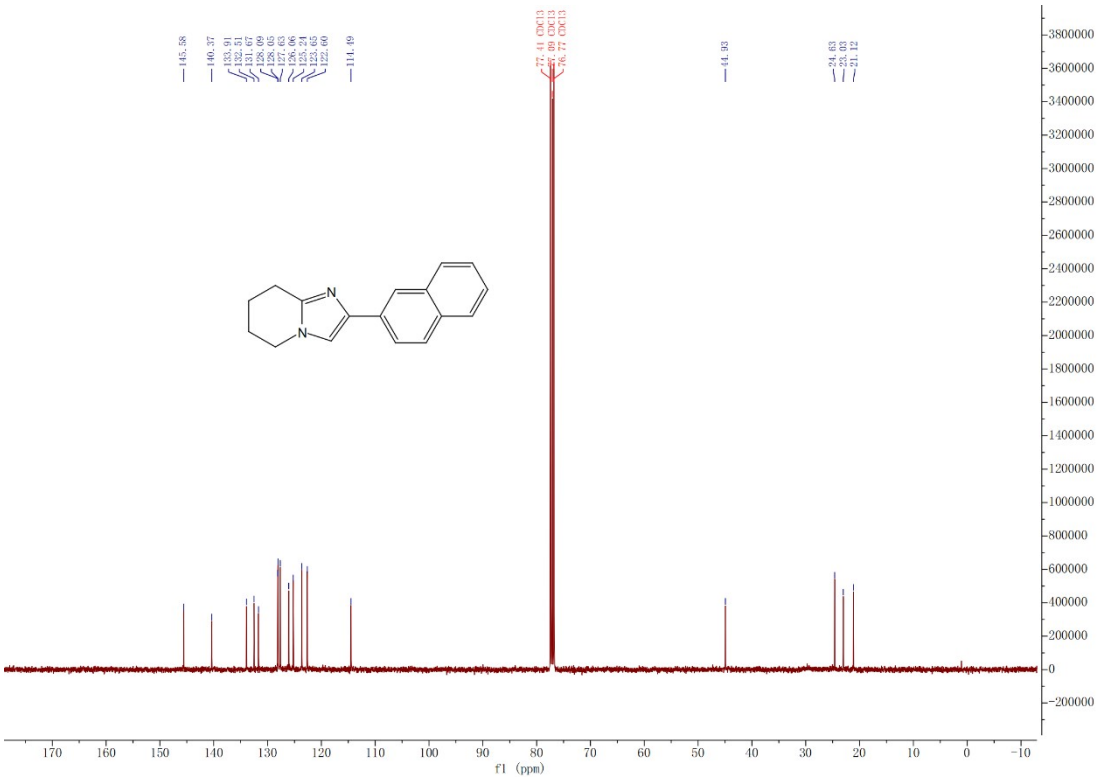

158.61
145.33
140.76
127.40
126.67
114.06
112.76
55.39
44.09
22.81
31.04
28.11
21.29
H3C
N
N
O
CH3
f1 (ppm)

4q

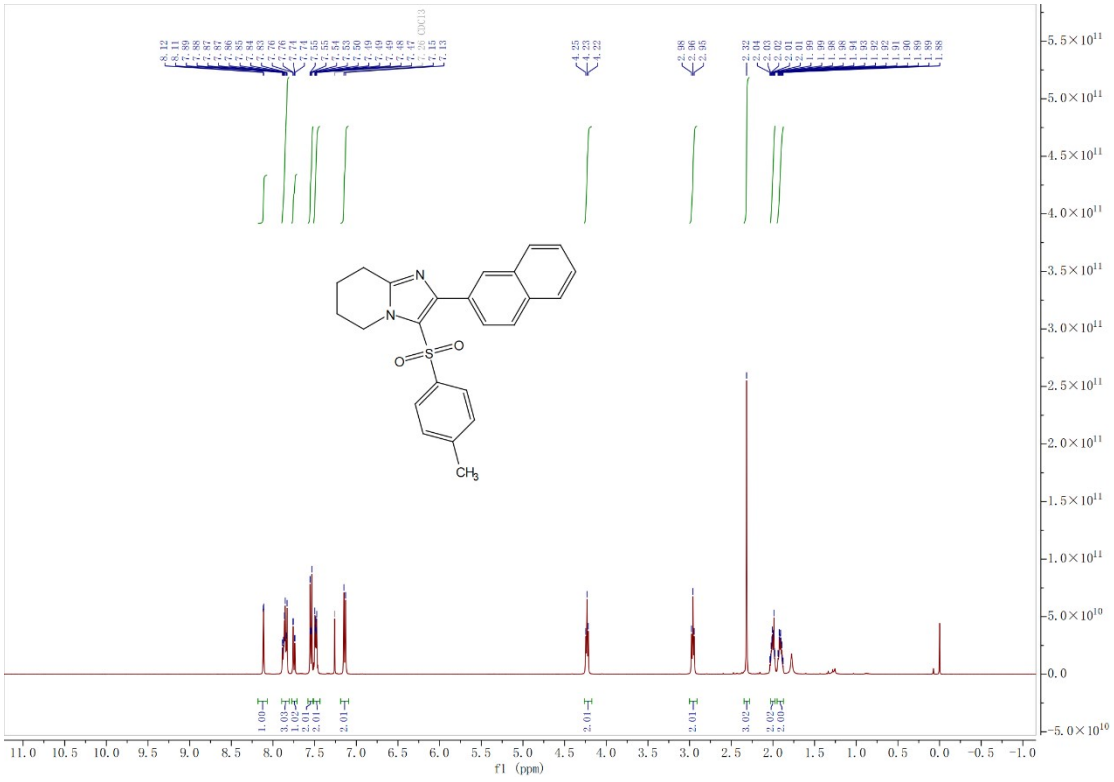

CDCl3
N
N
f1 (ppm)

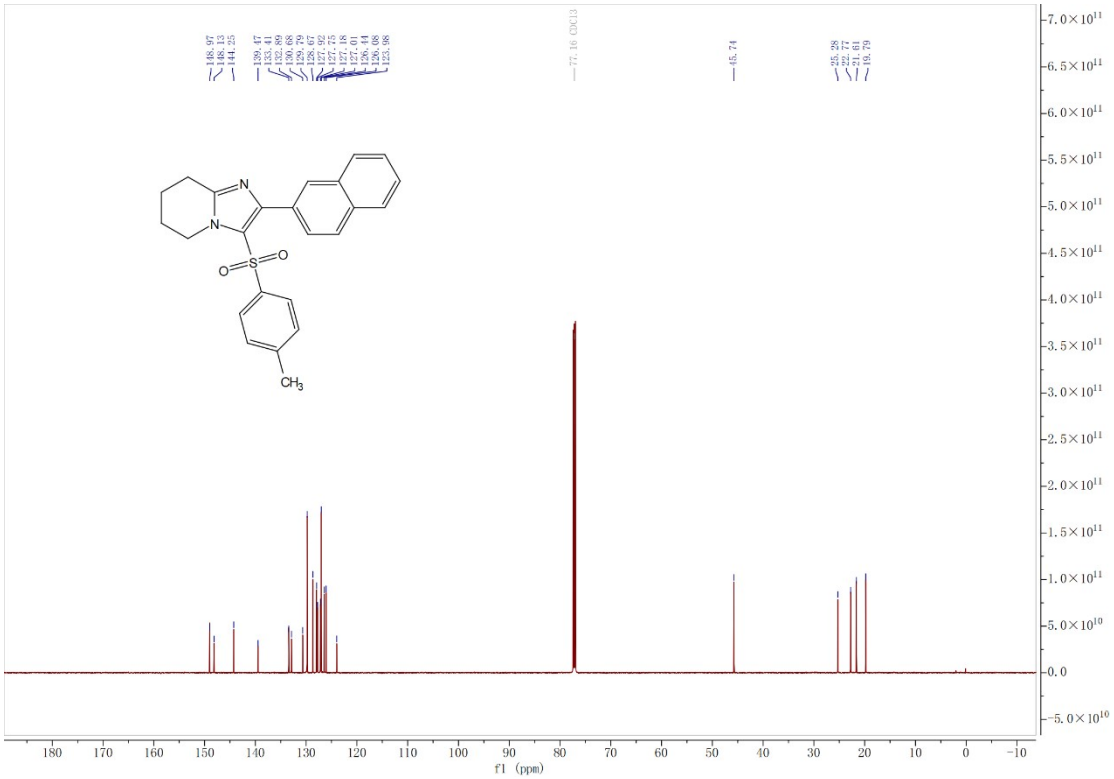
4r



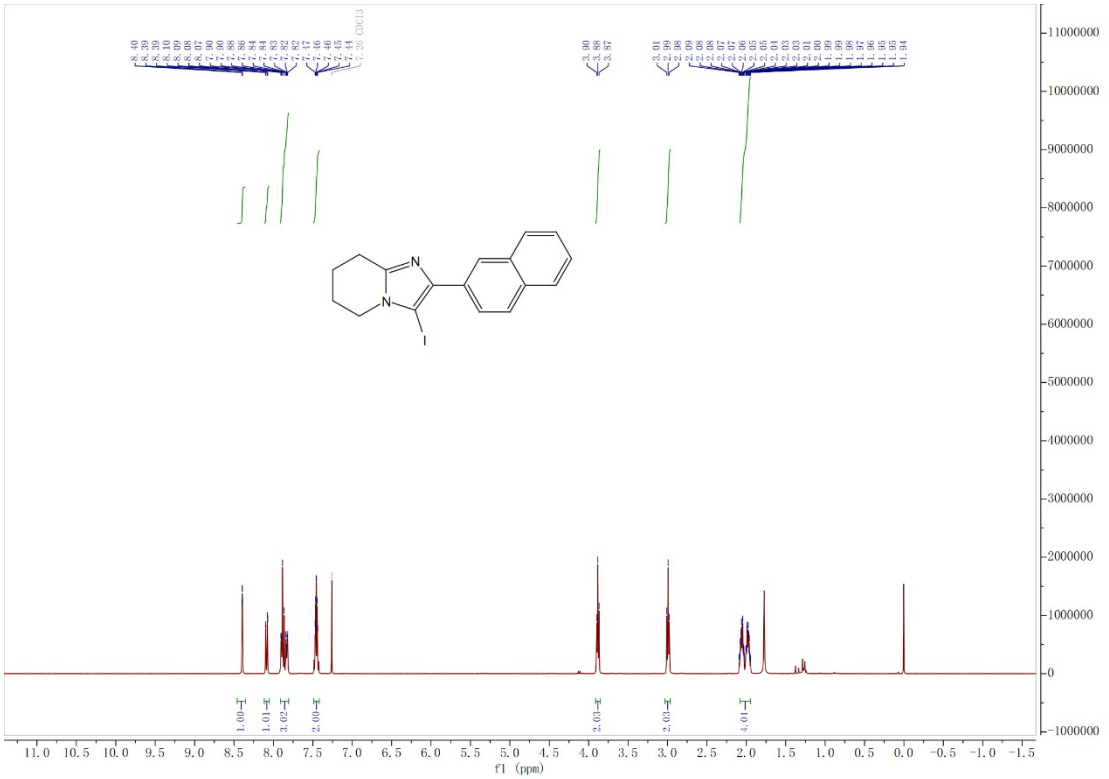


6


8.12
8.11
7.89
7.88
7.87
7.87
7.86
7.85
7.84
7.83
7.76
7.76
7.74
7.74
7.55
7.55
7.53
7.53
7.50
7.49
7.49
7.48
7.48
7.47
7.26
CDCl3
7.15
7.13
4.25
4.23
4.22
2.98
2.96
2.95
2.32
2.04
2.03
2.02
2.01
2.01
1.99
1.99
1.98
1.98
1.94
1.93
1.92
1.92
1.91
1.90
1.89
1.89
1.88
N
N
O
S
O
CH3
1.00
3.03
2.01
2.01
2.01
2.01
2.01
3.02
2.02
2.08
f1 (ppm)


148.97
148.13
144.25
139.47
135.41
132.89
130.68
129.79
128.97
127.75
127.18
127.01
126.44
126.08
123.98
77.16
CDCl3
65.74
25.28
22.77
21.84
19.79
N
N
O
S
O
CH3
f1 (ppm)

7


8.40
8.39
8.39
8.10
8.09
8.08
8.07
7.90
7.90
7.88
7.86
7.84
7.83
7.82
7.82
7.47
7.46
7.45
7.44
7.26 CDCl3
3.90
3.88
3.87
3.01
2.99
2.98
2.98
2.08
2.07
2.07
2.06
2.05
2.05
2.04
2.03
2.03
2.01
2.00
1.99
1.99
1.98
1.97
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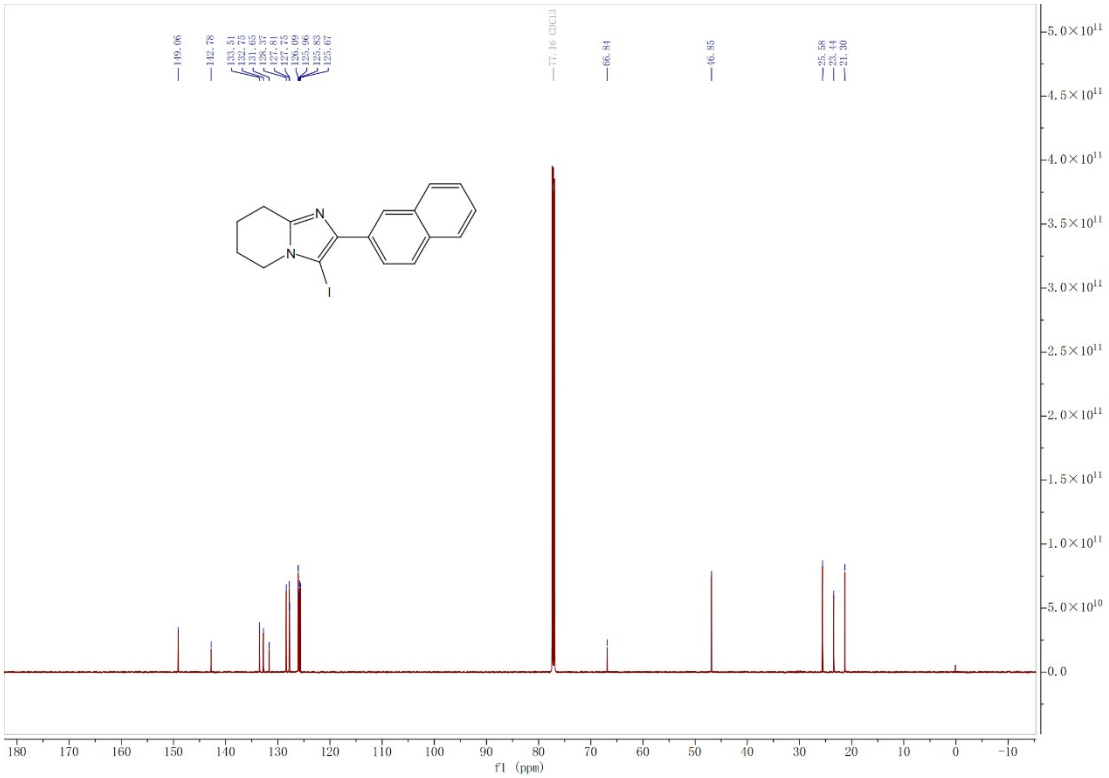
1.01
3.02
2.09
2.03
2.03
4.01
f1 (ppm)


149.06
142.78
133.51
132.75
131.65
128.37
127.81
127.73
126.66
125.96
125.83
125.67
77.16 CDCl3
66.84
46.85
25.58
23.44
21.30
f1 (ppm)

8