

# An electrochemical multicomponent [2+2+1] cascade cyclization of enaminones with primary amines: access to 1,2-disubstituted 4-acylimidazoles

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# **Supporting Information**

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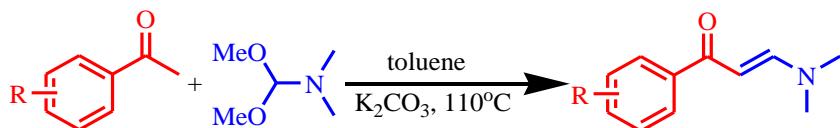
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## (A) Materials and equipment

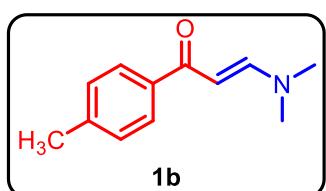
All solvents and reagents were obtained from commercial sources and were purified following the standard procedure prior to use. All glassware was oven-dried at 110 °C for hours and cooled down under a vacuum. Unless otherwise noted, materials were obtained from commercial suppliers and used without further purification. The instrument for electrolysis was a dual display potentiostat (DJS-292B) (made in China). The anode was platinum plate ( 10 mm × 10 mm × 0.2 mm ), and the cathode was nickel plate ( 10 mm × 10 mm × 0.2 mm ). Thin layer chromatography (TLC) employed glass 0.25 mm silica gel plates. Flash chromatography columns were packed with 200- 300 mesh silica gel in petroleum (bp. 60-90°C).  $^1\text{H}$ ,  $^{13}\text{C}$  and  $^{19}\text{F}$  NMR data for all compounds were recorded with Bruker Advance III (400 MHz) spectrometers with tetramethylsilane as an internal standard. All chemical shifts ( $\delta$ ) are reported in ppm and coupling constants (J) in Hz. All chemical shifts are reported relative to tetramethylsilane and D-solvent peaks (77.00 ppm, chloroform; 39.50 ppm, Dimethyl Sulfoxide), respectively. Highresolution mass spectral analysis (HRMS) data were measured on a Waters Acuity UPLC I-Class plus Xevo G2-XS (Q-TOF) mass spectrum by means of the ESI technique. **Crystallographic data** were obtained from a Bruker D8 Ouest diffractometer.

## (B) Typical experimental procedure

### 1. General procedure for the synthesis of enaminones **1**:



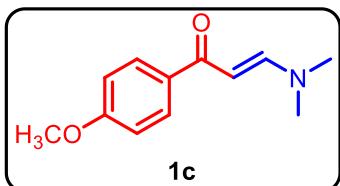
To a stirred solution of ketone (5.0 mmol, 1.0 equiv) in 5.0 mL of toluene, 1, 1-dimethoxy-N, N-dimethylmethanamine (7.0 mmol, 1.4 equiv) and  $\text{K}_2\text{CO}_3$  (5.0 mmol, 1.0 equiv) was added and stirred at 110 °C. After completion of the reaction (monitored by TLC), The resulting brown mixture was concentrated under a vacuum, the crude product was purified by flash column chromatography using petroleum ether/EtOAc (1:1) to give the enaminones **1**.<sup>[1]</sup>



**(E)-3-(dimethylamino)-1-(*p*-tolyl)prop-2-en-1-one (1b)**

According to the general procedure, **1b** was obtained using 1-(*p*-tolyl)ethan-1-one (670.9 mg, 668.2  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 75% yield (708.8 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

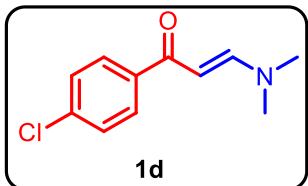
**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.815 - 7.778 (m, 3H), 7.208 (d,  $J$  = 7.6 Hz, 2H), 5.712 (d,  $J$  = 12.4 Hz, 1H), 3.117 (s, 3H), 2.937 (s, 3H), 2.384 (s, 3H).



**(E)-3-(dimethylamino)-1-(4-methoxyphenyl)prop-2-en-1-one (1c)**

According to the general procedure, **1c** was obtained using 1-(4-methoxyphenyl)ethan-1-one (751.0 mg, 694.1  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 82% yield (840.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

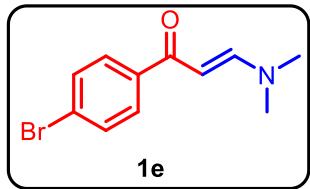
**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.896 - 7.854 (m, 2H), 7.750 (d,  $J$  = 12.4 Hz, 1H), 6.899 - 6.862 (m, 2H), 5.678 (d,  $J$  = 12.4 Hz, 1H), 3.811 (s, 3H), 3.053 (s, 3H), 2.913 (s, 3H).



**(E)-1-(4-chlorophenyl)-3-(dimethylamino)prop-2-en-1-one (1d)**

According to the general procedure, **1d** was obtained using 1-(4-chlorophenyl)ethan-1-one (773.0 mg, 664.6  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 83% yield (867.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

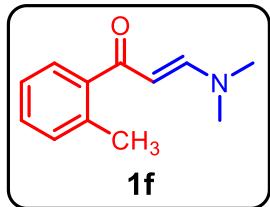
**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.840 - 7.777 (m, 3H), 7.374 - 7.347 (m, 2H), 5.647 (d,  $J$  = 12.0 Hz, 1H), 3.139 (s, 3H), 2.913 (s, 3H).



**(E)-1-(4-bromophenyl)-3-(dimethylamino)prop-2-en-1-one (1e)**

According to the general procedure, **1e** was obtained using 1-(4-bromophenyl)ethan-1-one (995.2 mg, 604.27  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 86% yield (1092.2 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

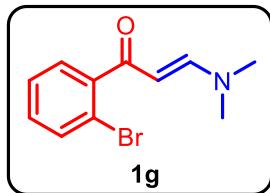
**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.808 - 7.736 (m, 3H), 7.537 - 7.503 (m, 2H), 5.640 (d,  $J$  = 12.4 Hz, 1H), 3.138 (s, 3H), 2.910 (s, 3H).



**(E)-3-(dimethylamino)-1-(o-tolyl)prop-2-en-1-one (1f)**

According to the general procedure, **1f** was obtained using 1-(o-tolyl)ethan-1-one (670.9 mg, 668.2  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 83% yield (784.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

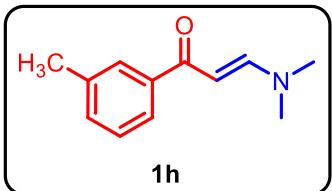
**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.361 - 7.182 (m, 5H), 5.362 (d,  $J$  = 12.8 Hz, 1H), 3.036 (s, 3H), 2.867 (s, 3H), 2.423 (s, 3H).



**(E)-1-(2-bromophenyl)-3-(dimethylamino)prop-2-en-1-one (1g)**

According to the general procedure, **1g** was obtained using 1-(2-bromophenyl)ethan-1-one (995.2 mg, 604.27  $\mu$ L, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 74% yield (939.8 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

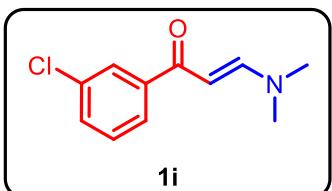
**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.827 - 7.802 (m, 1H), 7.364 - 7.318 (m, 4H), 5.640 (d, *J* = 12.4 Hz, 1H), 2.871 (s, 3H), 2.801 (s, 3H).



**(E)-3-(dimethylamino)-1-(*m*-tolyl)prop-2-en-1-one (1h)**

According to the general procedure, **1h** was obtained using 1-(*m*-tolyl)ethan-1-one (670.9 mg, 668.2 μL, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9 μL, 7 mmol) in 82% yield (774.9 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

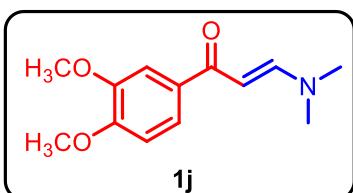
**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.664 - 7.557 (m, 3H), 7.176 - 7.107 (m, 2H), 5.600 - 5.551 (m, 1H), 2.920 (s, 3H), 2.725 (s, 3H), 2.275 - 2.255 (s, 3H).



**(E)-1-(3-chlorophenyl)-3-(dimethylamino)prop-2-en-1-one (1i)**

According to the general procedure, **1i** was obtained using 1-(3-chlorophenyl)ethan-1-one (773.0 mg, 664.6 μL, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9 μL, 7 mmol) in 78% yield (815.1 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.813 - 7.695 (m, 3H), 7.375 - 7.343 (m, 1H), 7.309-7.266 (m, 1H), 5.596 (d, *J* = 12.4 Hz, 1H), 3.108 (s, 3H), 2.883 (s, 3H).

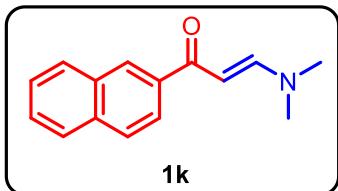


**(E)-1-(3,4-dimethoxyphenyl)-3-(dimethylamino)prop-2-en-1-one (1j)**

According to the general procedure, **1j** was obtained using 1-(3,4-dimethoxyphenyl)ethan-1-one (901.0 mg, 799.3 μL, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9 μL, 7 mmol) in 53% yield (622.8 mg) as a yellow solid (silica gel flash chromatography: petroleum

ether/EtOAc = 1:1).

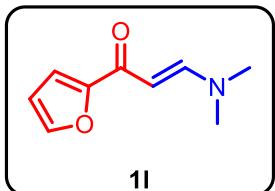
**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.750 (d, *J* = 12.4 Hz, 1H), 7.526 (d, *J* = 2.0 Hz, 1H), 7.471 (dd, *J* = 8.4, 2.0 Hz, 1H), 6.828 (d, *J* = 8.4 Hz, 1H), 5.682 (d, *J* = 12.4 Hz, 1H), 3.898 (d, *J* = 10.8 Hz, 6H), 3.053 (s, 3H), 2.918 (s, 3H).



**(E)-3-(dimethylamino)-1-(naphthalen-2-yl)prop-2-en-1-one (1k)**

According to the general procedure, **1k** was obtained using 1-(naphthalen-2-yl)ethan-1-one (850.0 mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9 μL, 7 mmol) in 77% yield (866.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

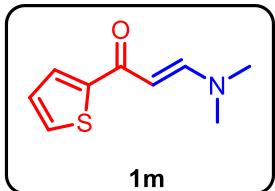
**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.390 (s, 1H), 8.008 (dd, *J* = 8.4, 1.6 Hz, 1H), 7.932 - 7.898 (m, 1H), 7.856 - 7.809 (m, 3H), 7.526 - 7.454 (m, 2H), 5.840 (d, *J* = 12.4 Hz, 1H), 3.078 (s, 3H), 2.899 (s, 3H).



**(E)-3-(dimethylamino)-1-(furan-2-yl)prop-2-en-1-one (1l)**

According to the general procedure, **1l** was obtained using 1-(furan-2-yl)ethan-1-one (550.2 mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9 μL, 7 mmol) in 81% yield (668.3 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

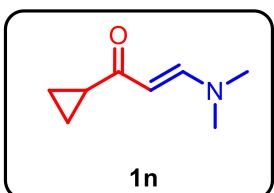
**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.676 - 7.599 (m, 1H), 7.374 - 7.341 (m, 1H), 6.945 - 6.898 (m, 1H), 6.358 - 6.308 (m, 1H), 5.563 - 5.489 (m, 1H), 2.944 (s, 3H), 2.731 (s, 3H).



**(E)-3-(dimethylamino)-1-(thiophen-2-yl)prop-2-en-1-one (1m)**

According to the general procedure, **1m** was obtained using 1-(thiophen-2-yl)ethan-1-one (630.0

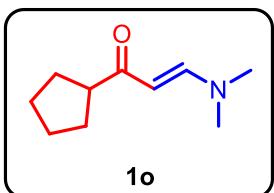
mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 79% yield (715.0 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).  
 $^1\text{H NMR}$  (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.758 - 7.704 (m, 1H), 7.601 - 7.575 (m, 1H), 7.443 - 7.414 (m, 1H), 7.057 - 7.019 (m, 1H), 5.609 - 5.563 (m, 1H), 3.069 (s, 3H), 2.871 (s, 3H).



**(E)-1-cyclopropyl-3-(dimethylamino)prop-2-en-1-one (1n)**

According to the general procedure, **1n** was obtained using 1-cyclopropylethan-1-one (420.3 mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 58% yield (403.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

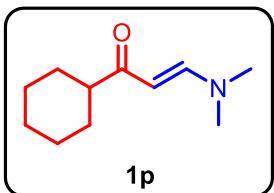
$^1\text{H NMR}$  (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.500 - 7.457 (m, 1H), 5.142 - 5.101 (m, 1H), 3.041 - 2.763 (m, 6H), 1.761 - 1.691 (m, 1H), 0.925 - 0.878 (m, 2H), 0.687 - 0.632 (m, 2H).



**(E)-1-cyclopentyl-3-(dimethylamino)prop-2-en-1-one (1o)**

According to the general procedure, **1o** was obtained using 1-cyclopentylethan-1-one (560.4 mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 69% yield (576.2 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

$^1\text{H NMR}$  (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.460 (d, *J* = 12.8 Hz, 1H), 4.978 (d, *J* = 12.4 Hz, 1H), 2.935 - 2.708 (m, 6H), 2.228 - 2.118 (s, 1H), 1.727 - 1.451 (m, 8H).



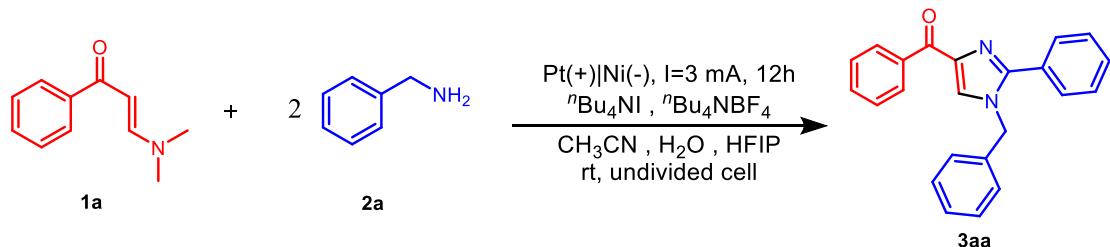
**(E)-1-cyclohexyl-3-(dimethylamino)prop-2-en-1-one (1p)**

According to the general procedure, **1p** was obtained using 1-cyclohexylethan-1-one (630.5 mg, 5 mmol) and 1,1-dimethoxy-N,N-dimethylmethanamine (834.12 mg, 929.9  $\mu$ L, 7 mmol) in 14% yield

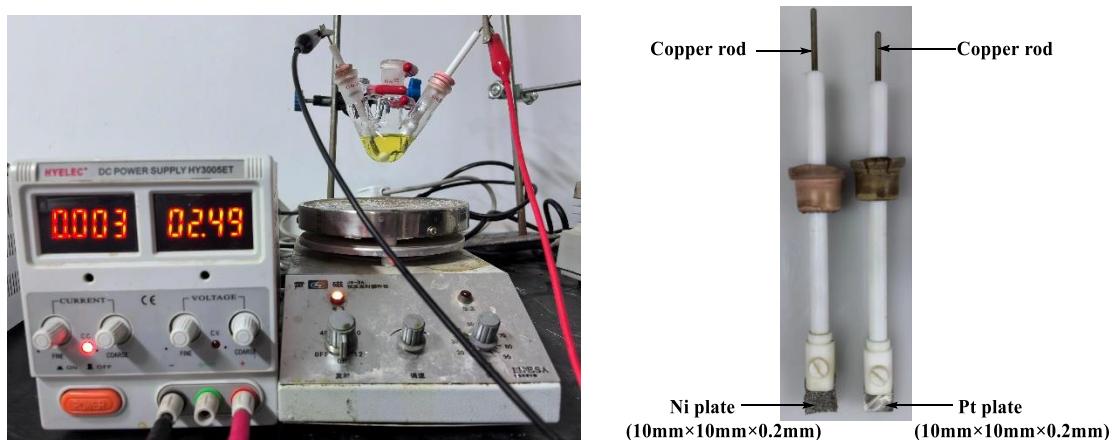
(127.0 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 1:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 7.405 - 7.305 (m, 1H), 4.903 - 4.756 (m, 1H), 2.872 - 2.640 (m, 6H), 2.138 - 1.965 (m, 1H), 1.642 - 1.410 (m, 5H), 1.281 - 0.975 (m, 5H).

## 2. General procedure for the synthesis of products **3**.



A 10-mL oven-dried undivided three-necked bottle was equipped with Pt plate anode (10 mm × 10 mm × 0.2 mm), Ni plate cathode (10 mm × 10 mm × 0.2 mm), constant current = 3 mA, **1a** (0.2 mmol), **2a** (0.4 mmol), "Bu<sub>4</sub>NI (0.6 mmol), "Bu<sub>4</sub>NBF<sub>4</sub> (0.2 mmol), H<sub>2</sub>O (0.5 mL), HFIP (0.5 mL), CH<sub>3</sub>CN (8 mL), room temperature, undivided cell. Then stirred for 12 h. The resulting brown mixture was concentrated under a vacuum, the crude product was purified by flash column chromatography using petroleum ether/EtOAc (6:1) to give the title compounds **3aa**.



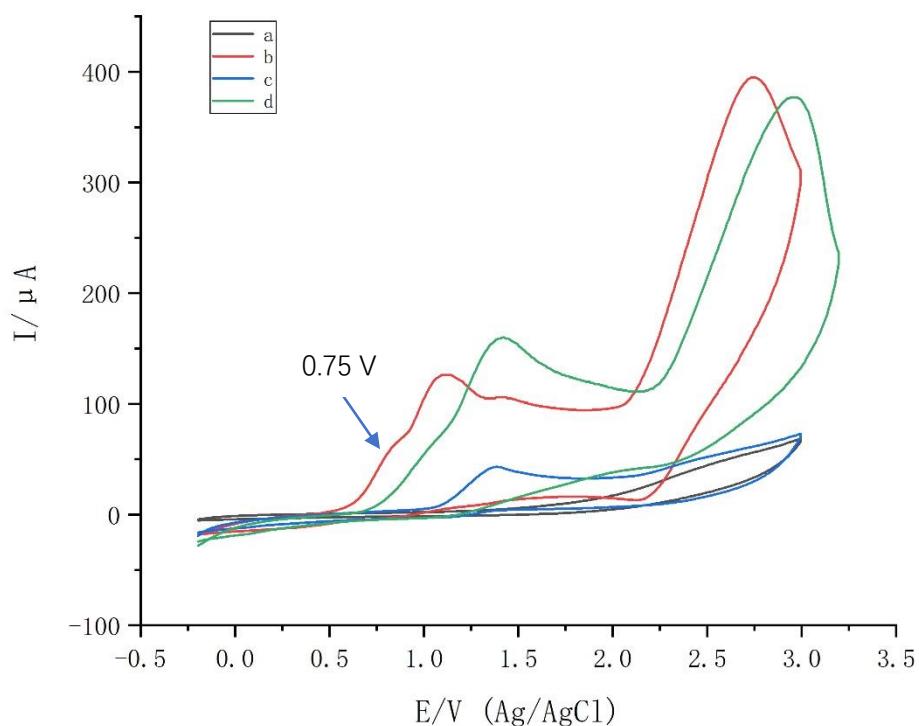
**Figure S1** Electrolysis setup



**Figure S2** Gram-scale amplification Electrolysis setup

### (C) Cyclic voltammetry experiments

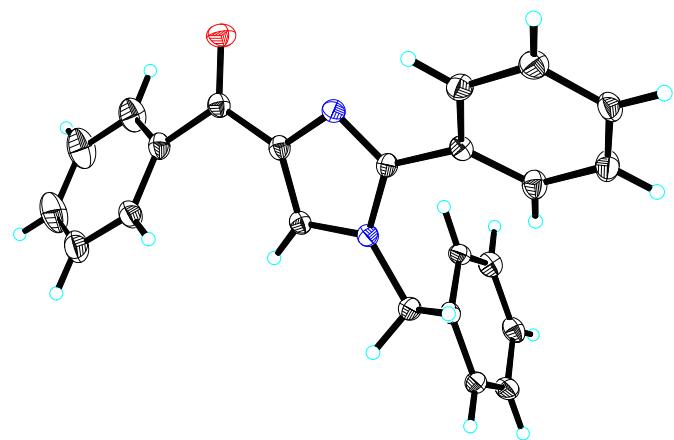
Cyclic voltammetry was performed in a three-electrode cell connected to a schlenk line under nitrogen at room temperature. The working electrode was a steady glassy carbon disk electrode, the counter electrode a platinum wire. The reference was an Ag/AgCl electrode submerged in saturated aqueous KCl solution. 8 mL of CH<sub>3</sub>CN were poured into the electrochemical cell in all experiments. The scan rate is 0.1 V/s, ranging from -0.2 V to 3.5 V.



**Figure S3** Cyclic voltammograms of in 8 mL of CH<sub>3</sub>CN solution containing different compounds:  
(a) CH<sub>3</sub>CN (8 mL), tetrabutylammonium tetrafluoroborate (0.02 mmol), benzylamine (0.04 mmol);  
(b) CH<sub>3</sub>CN (8 mL), tetrabutylammonium tetrafluoroborate (0.02 mmol), tetrabutylammonium iodide (0.06 mmol); (c) CH<sub>3</sub>CN (8 ml), tetrabutylammonium tetrafluoroborate (0.02 mmol), 3-(Dimethylamino)-1-phenyl-2-propen-1-one (0.02 mmol); (d) CH<sub>3</sub>CN (8 ml), tetrabutylammonium tetrafluoroborate (0.02 mmol), tetrabutylammonium iodide (0.06 mmol), 3-(Dimethylamino)-1-phenyl-2-propen-1-one (0.02 mmol).

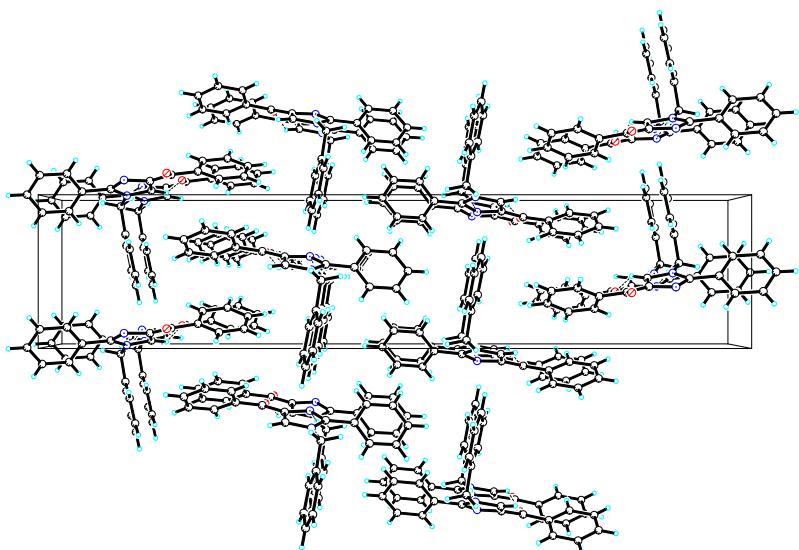
### (D) X-Ray crystallographic studies

Crystal data for md\_zxj4:  $C_{23}H_{18}N_2O$ ,  $M = 338.39$ ,  $a = 6.2987(4)$  Å,  $b = 7.8754(5)$  Å,  $c = 36.641(2)$  Å,  $\alpha = 90^\circ$ ,  $\beta = 90^\circ$ ,  $\gamma = 90^\circ$ ,  $V = 1817.6(2)$  Å $^3$ ,  $T = 150.(2)$  K, space group  $P212121$ ,  $Z = 4$ ,  $\mu(\text{Cu } \text{K}\alpha) = 0.601$  mm $^{-1}$ , 24679 reflections measured, 3343 independent reflections ( $R_{\text{int}} = 0.1563$ ). The final  $R_I$  values were 0.0706 ( $I > 2\sigma(I)$ ). The final  $wR(F^2)$  values were 0.2053 ( $I > 2\sigma(I)$ ). The final  $R_I$  values were 0.0800 (all data). The final  $wR(F^2)$  values were 0.2108 (all data). The goodness of fit on  $F^2$  was 1.096. Flack parameter = 0.1(3).



View of a molecule of md\_zxj4.

Displacement ellipsoids are drawn at the 30% probability level.



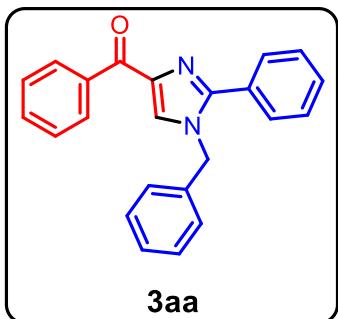
View of the pack drawing of md\_zxj4.

Hydrogen-bonds are shown as dashed lines.

Table 1. Crystal data and structure refinement for md\_zxj4\_0m.

Identification code	global	
Empirical formula	C23 H18 N2 O	
Formula weight	338.39	
Temperature	150(2) K	
Wavelength	1.54178 Å	
Crystal system	Orthorhombic	
Space group	P2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>	
Unit cell dimensions	a = 6.2987(4) Å b = 7.8754(5) Å c = 36.641(2) Å	α= 90 ° β= 90 ° γ = 90 °
Volume	1817.6(2) Å <sup>3</sup>	
Z	4	
Density (calculated)	1.237 Mg/m <sup>3</sup>	
Absorption coefficient	0.601 mm <sup>-1</sup>	
F(000)	712	
Crystal size	0.460 x 0.030 x 0.030 mm <sup>3</sup>	
Theta range for data collection	2.41 to 68.52 °	
Index ranges	-6<=h<=7, -9<=k<=9, -44<=l<=44	
Reflections collected	24679	
Independent reflections	3343 [R(int) = 0.1563]	
Completeness to theta = 68.52 °	99.7 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.98 and 0.80	
Refinement method	Full-matrix least-squares on F <sup>2</sup>	
Data / restraints / parameters	3343 / 0 / 236	
Goodness-of-fit on F <sup>2</sup>	1.096	
Final R indices [I>2sigma(I)]	R1 = 0.0706, wR2 = 0.2053	
R indices (all data)	R1 = 0.0800, wR2 = 0.2108	
Absolute structure parameter	0.1(3)	
Extinction coefficient	0.0026(9)	
Largest diff. peak and hole	0.458 and -0.272 e.Å <sup>-3</sup>	

## (E) Analytical data for products



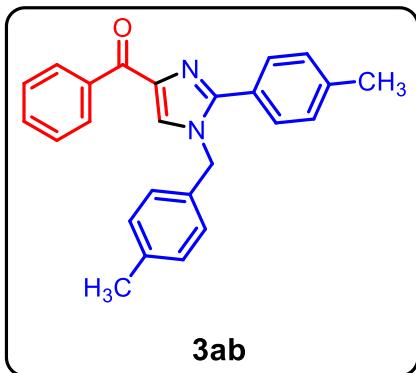
### **(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(phenyl)methanone (3aa)**

According to the general procedure, **3aa** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 84% yield (56.8 mg) as a yellow solid (silica gel flash chromatography: pet roleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  8.256 - 8.228 (m, 3H), 7.641 - 7.600 (m, 3H), 7.533 (t, *J* = 8.0 Hz, 2H), 7.484 - 7.460 (m, 3H), 7.336 - 7.266 (m, 3H), 7.058 (d, *J* = 6.8 Hz, 2H), 5.442 (s, 2H);

**<sup>13</sup>C NMR** (100 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  186.47, 147.82, 139.75, 137.79, 136.73, 132.29, 130.49, 129.86, 129.73, 129.40, 128.84, 128.74, 128.70, 128.24, 127.82, 126.82, 50.25.

**HRMS** (ESI) *m/z* calcd. for C<sub>23</sub>H<sub>19</sub>N<sub>2</sub>O (M+H)<sup>+</sup>: 339.1492, found 339.1487.



### **(1-(4-methylbenzyl)-2-(p-tolyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ab)**

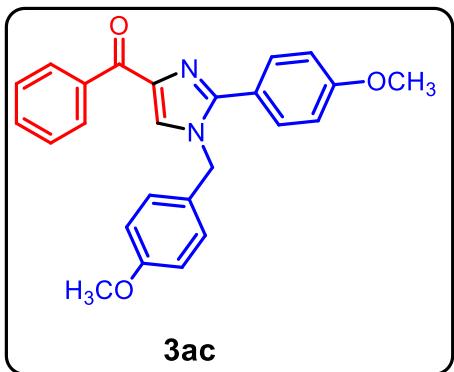
According to the general procedure, **3ab** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and *p*-tolylmethanamine **2b** (48.4 mg, 50.9  $\mu$ L, 0.4 mmol) in 61% yield (44.7 mg) as yellow oily liquid (silica gel flash chromatography: p etroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.216 - 8.192 (m, 2H), 7.622 (s, 1H), 7.482 - 7.434 (m, 3H), 7.402 -

7.362 (m, 2H), 7.176 (d,  $J$  = 7.6 Hz, 2H), 7.088 (d,  $J$  = 7.6 Hz, 2H), 6.933 (d,  $J$  = 8.0 Hz, 2H), 5.122 (s, 2H), 2.326 (s, 3H), 2.271 (s, 3H);

**$^{13}\text{C}$  NMR** (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.93, 149.22, 140.93, 139.74, 138.34, 138.21, 132.89, 132.26, 130.34, 129.94, 129.48, 129.23, 128.62, 128.23, 127.08, 126.89, 50.90, 21.50, 21.22.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{25}\text{H}_{23}\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 367.1805, found 367.1799.



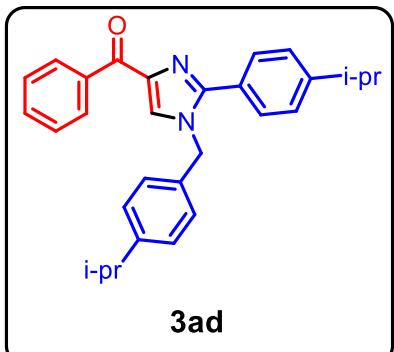
**(1-(4-methoxybenzyl)-2-(4-methoxyphenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ac)**

According to the general procedure, **3ac** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (4-methoxyphenyl)methanamine **2c** (54.8 mg, 52.2  $\mu\text{L}$ , 0.4 mmol) in 62% yield (49.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H}$  NMR** (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.272 - 8.284 (m, 2H), 7.671 (s, 1H), 7.572 - 7.502 (m, 3H), 7.447 (t,  $J$  = 7.6 Hz, 2H), 7.032 (d,  $J$  = 8.8 Hz, 2H), 6.958 (d,  $J$  = 8.8 Hz, 2H), 6.864 (d,  $J$  = 8.8 Hz, 2H), 5.151 (s, 2H), 3.835 (s, 3H), 3.786 (s, 3H);

**$^{13}\text{C}$  NMR** (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.90, 160.64, 159.66, 148.94, 140.73, 138.20, 132.23, 130.72, 130.26, 128.51, 128.47, 128.21, 127.82, 122.16, 114.61, 114.17, 55.45, 55.42, 50.61.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{25}\text{H}_{23}\text{N}_2\text{O}_3 (\text{M}+\text{H})^+$ : 399.1703, found 399.1709.



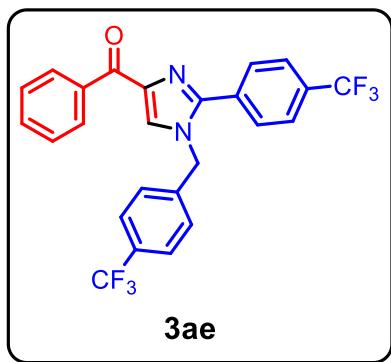
**(1-(4-isopropylbenzyl)-2-(4-isopropylphenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ad)**

According to the general procedure, **3ad** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (4-isopropylphenyl)methanamine **2d** (59.6 mg, 64.1  $\mu$ L, 0.4 mmol) in 74% yield (62.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.304 (d,  $J$  = 7.6 Hz, 2H), 7.717 (s, 1H), 7.575 - 7.517 (m, 3H), 7.458 (t,  $J$  = 7.6 Hz, 2H), 7.308 (d,  $J$  = 7.6 Hz, 2H), 7.219 (d,  $J$  = 8.0 Hz, 2H), 7.054 (d,  $J$  = 8.0 Hz, 2H), 5.221 (s, 2H), 2.973 - 2.890 (m, 2H), 1.281 - 1.236 (m, 12H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.98, 150.65, 149.35, 149.25, 141.07, 138.20, 133.29, 132.30, 130.44, 129.35, 128.62, 128.24, 127.37, 127.20, 126.94, 50.92, 34.17, 33.95, 29.84, 24.04, 23.98.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{29}\text{H}_{31}\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 423.2431, found 423.2437.



**Phenyl(1-(4-(trifluoromethyl)benzyl)-2-(4-(trifluoromethyl)phenyl)-1*H*-imidazol-4-yl)methanone (3ae)**

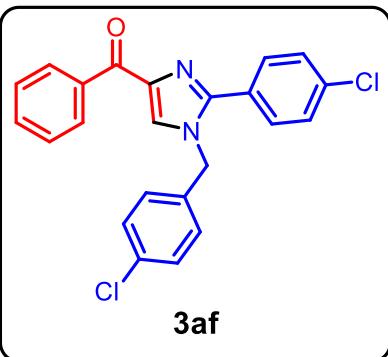
According to the general procedure, **3ae** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (4-(trifluoromethyl)phenyl)methanamine **2e** (70.0 mg, 57.3  $\mu$ L, 0.4 mmol) in 72% yield (68.3 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.312 - 8.284 (m, 2H), 7.788 (s, 1H), 7.702 (s, 4H), 7.628 (d,  $J$  = 8.0 Hz, 2H), 7.569 - 7.541 (m, 1H), 7.494 - 7.454 (m, 2H), 7.207 (d,  $J$  = 8.0 Hz, 2H), 5.347 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.56, 147.43, 141.79, 139.45, 137.70, 132.93, 132.70, 131.42 (dd,  $J$  = 33.3, 33.3 Hz), 130.34, 129.48, 129.06, 128.37, 127.38 (d,  $J$  = 26.3 Hz), 127.08, 126.46 (q,  $J$  = 7.1, 4.0 Hz), 125.93 (q,  $J$  = 8.1, 4.0 Hz), 125.18 (d,  $J$  = 4.0 Hz), 122.47 (d,  $J$  = 4.0 Hz), 50.72;

**$^{19}\text{F NMR}$**  (376 MHz,  $\text{CDCl}_3$ )  $\delta$  -62.76, -62.89.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{25}\text{H}_{17}\text{F}_6\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 475.1240, found 475.1233.



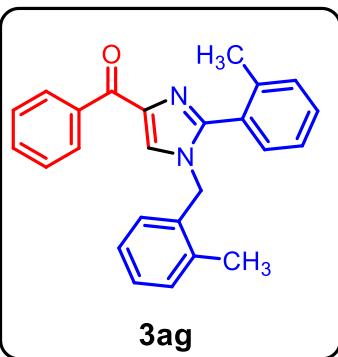
**(1-(4-chlorobenzyl)-2-(4-chlorophenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3af)**

According to the general procedure, **3af** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (4-chlorophenyl)methanamine **2f** (56.4 mg, 48.4  $\mu$ L, 0.4 mmol) in 66% yield (53.7 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.290 - 8.268 (m, 2H), 7.732 (s, 1H), 7.572 - 7.511 (m, 3H), 7.470 (t,  $J$  = 7.6 Hz, 2H), 7.416 (d,  $J$  = 8.4 Hz, 2H), 7.334 (d,  $J$  = 8.4 Hz, 2H), 7.018 (d,  $J$  = 8.4 Hz, 2H), 5.212 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.69, 147.83, 141.41, 137.88, 136.09, 134.68, 134.08, 132.55, 130.50, 130.34, 129.63, 129.20, 128.74, 128.34, 128.25, 128.03, 50.56.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{17}\text{Cl}_2\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 407.0712, found 407.0706.



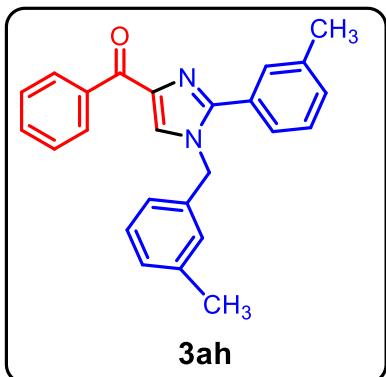
**(1-(2-methylbenzyl)-2-(o-tolyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ag)**

According to the general procedure, **3ag** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and o-tolylmethanamine **2g** (48.4 mg, 49.6  $\mu$ L, 0.4 mmol) in 57% yield (41.7 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.214 (d,  $J$  = 7.6 Hz, 2H), 7.582 (s, 1H), 7.431 (t,  $J$  = 7.6 Hz, 1H),

7.357 (t,  $J = 7.6$  Hz, 2H), 7.285 (t,  $J = 7.6$  Hz, 1H), 7.224 (d,  $J = 7.6$  Hz, 2H), 7.174 - 7.126 (m, 2H), 7.098 - 7.049 (m, 2H), 6.872 (d,  $J = 7.6$  Hz, 1H), 4.867 (s, 2H), 2.186 (s, 3H), 1.987 (s, 3H);  
 $^{13}\text{C}$  NMR (100 MHz, CDCl<sub>3</sub>) δ 188.04, 148.27, 140.93, 138.77, 138.05, 136.08, 133.40, 132.25, 130.93, 130.75, 130.43, 130.39, 130.00, 129.42, 128.77, 128.55, 128.19, 127.17, 126.73, 125.88, 48.98, 19.98, 18.98.

**HRMS** (ESI)  $m/z$  calcd. for C<sub>25</sub>H<sub>23</sub>N<sub>2</sub>O (M+H)<sup>+</sup>: 367.1805, found 367.1801.



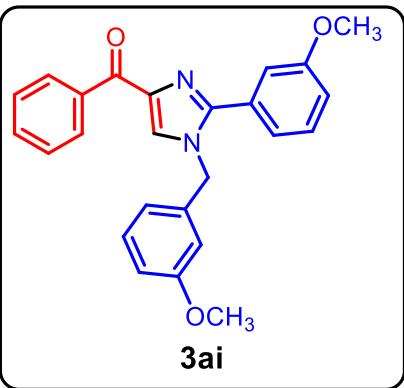
**(1-(3-methylbenzyl)-2-(m-tolyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ah)**

According to the general procedure, **3ah** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and m-tolylmethanamine **2h** (48.4 mg, 49.6 μL, 0.4 mmol) in 64% yield (46.8 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

$^1\text{H}$  NMR (400 MHz, CDCl<sub>3</sub>) δ 8.217 (d,  $J = 7.2$  Hz, 2H), 7.643 (s, 1H), 7.452 (t,  $J = 7.2$  Hz, 1H), 7.378 (t,  $J = 7.2$  Hz, 3H), 7.292 (d,  $J = 7.6$  Hz, 1H), 7.234 (t,  $J = 7.2$  Hz, 1H), 7.186 - 7.139 (m, 2H), 7.051 (d,  $J = 8.0$  Hz, 1H), 6.837 (s, 2H), 5.114 (s, 2H), 2.297 (s, 3H), 2.241 (s, 3H);

$^{13}\text{C}$  NMR (100 MHz, CDCl<sub>3</sub>) δ 187.91, 149.25, 140.99, 139.11, 138.64, 138.17, 135.84, 132.29, 130.44, 130.37, 130.26, 129.64, 129.24, 129.15, 128.70, 128.58, 128.25, 127.83, 126.19, 124.24, 51.16, 21.49, 21.47.

**HRMS** (ESI)  $m/z$  calcd. for C<sub>25</sub>H<sub>22</sub>N<sub>2</sub>O<sup>+</sup> (M+Na)<sup>+</sup>: 389.1624, found 389.1616.



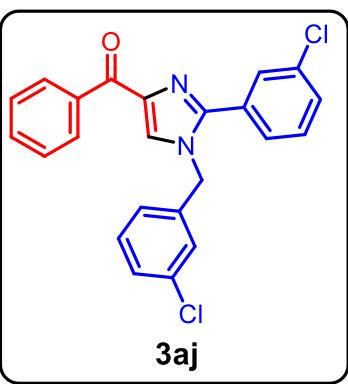
**(1-(3-methoxybenzyl)-2-(3-methoxyphenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ai)**

According to the general procedure, **3ai** was obtained using (*E*)-3-(dimethylamino)-1-phenyl prop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (3-methoxyphenyl)methanamine **2i** (54.8 mg, 51.1  $\mu$ L, 0.4 mmol) in 62% yield (49.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.269 - 8.245 (m, 2H), 7.711 (s, 1H), 7.509 (t, *J* = 7.2 Hz, 1H), 7.431 (t, *J* = 8.0 Hz, 2H), 7.311 (t, *J* = 8.0 Hz, 1H), 7.240 - 7.220 (m, 1H), 7.153 - 7.124 (m, 2H), 6.971 - 6.944 (m, 1H), 6.833 - 6.806 (m, 1H), 6.664 (d, *J* = 7.6 Hz, 1H), 6.595 (s, 1H), 5.195 (s, 2H), 3.734 (s, 3H), 3.724 (s, 3H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>)  $\delta$  187.85, 160.33, 159.85, 148.92, 141.02, 138.11, 137.46, 132.35, 130.89, 130.44, 130.34, 129.86, 128.87, 128.27, 121.50, 119.15, 115.85, 114.66, 113.67, 112.86, 55.45, 55.39, 51.06.

**HRMS (ESI)** *m/z* calcd. for C<sub>25</sub>H<sub>23</sub>N<sub>2</sub>O<sub>3</sub> (M+H)<sup>+</sup>: 399.1703, found 399.1712.



**(1-(3-chlorobenzyl)-2-(3-chlorophenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3aj)**

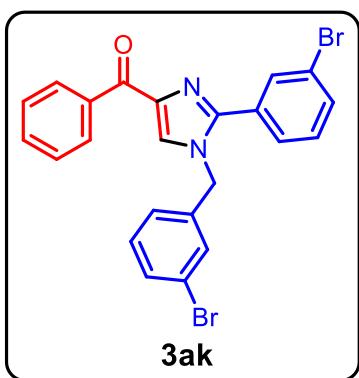
According to the general procedure, **3aj** was obtained using (*E*)-3-(dimethylamino)-1-phenyl prop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (3-chlorophenyl)methanamine **2j** (56.4 mg, 4

8.4  $\mu$ L, 0.4 mmol) in 48% yield (39.1 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.302 (d,  $J$  = 7.6 Hz, 2H), 7.76 (s, 1H), 7.620 (s, 1H), 7.561 (t,  $J$  = 7.2 Hz, 1H), 7.492 (d,  $J$  = 7.6 Hz, 2H), 7.463 - 7.432 (m, 2H), 7.384 (d,  $J$  = 6.8 Hz, 1H), 7.310 (d,  $J$  = 6.8 Hz, 2H), 7.097 (s, 1H), 6.972 (d,  $J$  = 6.8 Hz, 1H), 5.230 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.68, 147.46, 141.50, 137.81, 137.47, 135.45, 135.00, 132.60, 131.22, 130.74, 130.40, 130.17, 130.00, 129.52, 128.99, 128.78, 128.36, 127.23, 127.19, 125.11, 50.69.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{17}\text{Cl}_2\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 407.0712, found 407.0708.



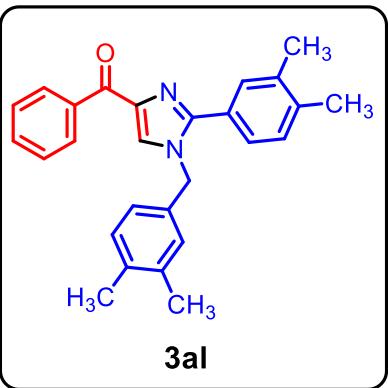
**(1-(3-bromobenzyl)-2-(3-bromophenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ak)**

According to the general procedure, **3ak** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (3-bromophenyl)methanamine **2k** (74.0 mg, 50.0  $\mu$ L, 0.4 mmol) in 62% yield (61.5 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.305 - 8.282 (m, 2H), 7.760 (s, 2H), 7.605 - 7.580 (m, 1H), 7.552 (d,  $J$  = 6.8 Hz, 1H), 7.500 - 7.464 (m, 4H), 7.311 (t,  $J$  = 7.6 Hz, 1H), 7.254 - 7.220 (m, 2H), 7.014 (d,  $J$  = 8.8 Hz, 1H), 5.214 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.68, 147.32, 141.51, 137.81, 137.69, 132.90, 132.59, 132.38, 131.93, 131.46, 130.99, 130.39, 130.36, 130.17, 128.79, 128.36, 127.63, 125.60, 123.50, 123.00, 50.62.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{17}\text{Br}_2\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 494.9702, found 494.9700.

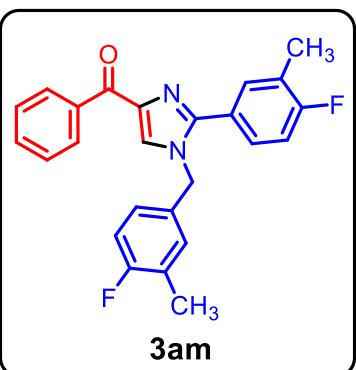


**(1-(3,4-dimethylbenzyl)-2-(3,4-dimethylphenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3al)**

According to the general procedure, **3al** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (3,4-dimethylphenyl)methanamine **2l** (54.0 mg, 56.9  $\mu$ L, 0.4 mmol) in 72% yield (56.7 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>)  $\delta$  8.285 (d, *J* = 7.2 Hz, 2H), 7.691 (s, 1H), 7.529 (t, *J* = 6.0 Hz, 1H), 7.456 (t, *J* = 6.8 Hz, 3H), 7.326 (d, *J* = 7.6 Hz, 1H), 7.193 (d, *J* = 8.0 Hz, 1H), 7.112 (d, *J* = 7.6 Hz, 1H), 6.872 (t, *J* = 8.4 Hz, 2H), 5.161 (s, 2H), 2.302 (d, *J* = 6.4 Hz, 6H), 2.242 (d, *J* = 8.0 Hz, 6H); **<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>)  $\delta$  187.96, 149.35, 140.79, 138.37, 138.25, 137.58, 137.17, 136.92, 133.31, 132.20, 130.69, 130.39, 130.33, 129.86, 128.62, 128.48, 128.20, 127.25, 126.50, 124.69, 50.93, 19.87, 19.81, 19.77, 19.53.

**HRMS** (ESI) *m/z* calcd. for C<sub>27</sub>H<sub>27</sub>N<sub>2</sub>O (M+H)<sup>+</sup>: 395.2118, found 395.2126.



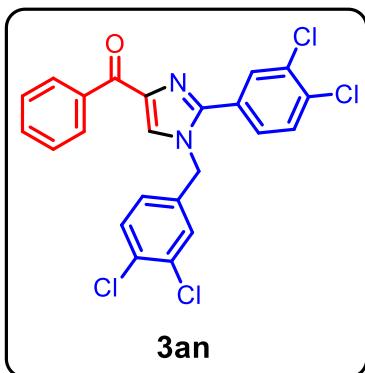
**(1-(4-fluoro-3-methylbenzyl)-2-(4-fluoro-3-methylphenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3am)**

According to the general procedure, **3am** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (4-fluoro-3-methylphenyl)methanamine **2m**

(55.6 mg, 52.5  $\mu$ L, 0.4 mmol) in 81% yield (65.1 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.273 - 8.249 (m, 2H), 7.700 (s, 1H), 7.562 - 7.518 (m, 1H), 7.484 - 7.439 (m, 3H), 7.355 - 7.316 (m, 1H), 7.056 (t,  $J$  = 9.2 Hz, 1H), 6.975 (t,  $J$  = 8.4 Hz, 1H), 6.917 - 6.853 (m, 2H), 5.142 (s, 2H), 2.292 (d,  $J$  = 2.0 Hz, 3H), 2.236 (d,  $J$  = 2.0 Hz, 3H);  
 **$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.84, 163.46, 162.48, 160.99, 160.03, 148.28, 140.96, 138.02, 132.93 (d,  $J$  = 6.1 Hz), 132.42, 131.16 (d,  $J$  = 4.0 Hz), 130.29 (t,  $J$  = 3.0 Hz), 128.52, 128.30, 128.28 (d,  $J$  = 9.1 Hz), 126.15 (d,  $J$  = 8.1 Hz), 125.95 (d,  $J$  = 12.1 Hz), 125.71, 125.47 (d,  $J$  = 4.0 Hz), 115.86 (d,  $J$  = 23.2 Hz), 115.44 (d,  $J$  = 23.2 Hz), 50.56, 14.64 (dd,  $J$  = 5.0, 4.0 Hz);  
 **$^{19}\text{F NMR}$**  (376 MHz,  $\text{CDCl}_3$ )  $\delta$  -115.00, -117.36.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{25}\text{H}_{21}\text{F}_2\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 403.1616, found 403.1620.



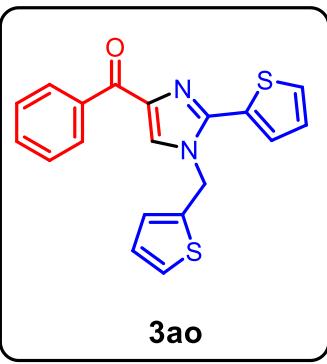
**(1-(3,4-dichlorobenzyl)-2-(3,4-dichlorophenyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3an)**

According to the general procedure, **3an** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and (3,4-dichlorophenyl)methanamine **2n** (70.8 mg, 53.6  $\mu$ L, 0.4 mmol) in 53% yield (50.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.286 (d,  $J$  = 7.6 Hz, 2H), 7.757 (s, 1H), 7.711 (s, 1H), 7.571 (t,  $J$  = 7.2 Hz, 1H), 7.528 - 7.440 (m, 4H), 7.384 - 7.359 (m, 1H), 7.195 (s, 1H), 6.929 - 6.903 (m, 1H), 5.208 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.53, 146.47, 141.74, 137.66, 135.41, 134.51, 133.90, 133.48, 133.29, 132.74, 131.54, 131.16, 130.98, 130.38, 129.26, 128.95, 128.82, 128.42, 128.05, 126.12, 50.21.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{14}\text{Cl}_4\text{N}_2\text{ONa} (\text{M}+\text{Na})^+$ : 496.9752, found 496.9760.



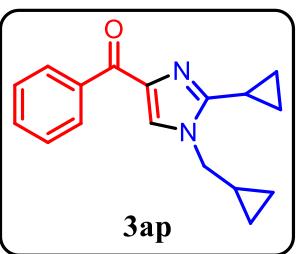
**Phenyl(2-(thiophen-2-yl)-1-(thiophen-2-ylmethyl)-1*H*-imidazol-4-yl)methanone (3ao)**

According to the general procedure, **3ao** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and thiophen-2-ylmethanamine **2o** (45.2 mg, 41.0 µL, 0.4 mmol) in 59% yield (41.3 mg) as a yellow solid (silica gel flash chromatograph y: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.296 (d, *J* = 7.2 Hz, 2H), 7.768 (s, 1H), 7.567 - 7.523 (m, 1H), 7.484 - 7.447 (m, 3H), 7.388 - 7.376 (m, 1H), 7.317 - 7.301 (m, 1H), 7.129 - 7.107 (m, 1H), 6.997 - 6.976 (m, 1H), 6.957 (s, 1H), 5.490 (s, 2H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 187.49, 142.61, 141.18, 137.87, 137.36, 132.46, 131.08, 130.41, 128.42, 128.26, 128.16, 128.06, 127.76, 127.51, 127.24, 126.73, 46.32.

**HRMS (ESI) *m/z*** calcd. for C<sub>19</sub>H<sub>15</sub>N<sub>2</sub>OS<sub>2</sub>(M+H)<sup>+</sup>: 351.0620, found 351.0624.



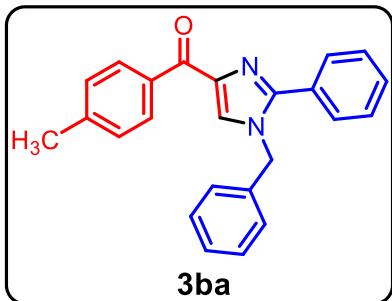
**(2-cyclopropyl-1-(cyclopropylmethyl)-1*H*-imidazol-4-yl)(phenyl)methanone (3ap)**

According to the general procedure, **3ap** was obtained using (*E*)-3-(dimethylamino)-1-phenylprop-2-en-1-one **1a** (35.0 mg, 0.2 mmol) and cyclopropylmethanamine **2p** (28.4 mg, 34.2 µL, 0.4 mmol) in 28% yield (14.9 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 4:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.162 - 8.138 (m, 2H), 7.673 (s, 1H), 7.550 - 7.513 (m, 1H), 7.469 - 7.429 (m, 2H), 3.900 (d, *J* = 7.2 Hz, 2H), 1.853 - 1.787 (m, 1H), 1.257 - 1.233 (m, 1H), 1.175 - 1.136 (m, 2H), 1.025 - 0.978 (m, 2H), 0.728 - 0.680 (m, 2H), 0.414 - 0.375 (m, 2H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 187.89, 150.71, 139.20, 138.62, 132.04, 130.11, 128.19, 127.64, 50.85, 11.35, 7.42, 4.34, 1.16.

**HRMS (ESI) *m/z*** calcd. for C<sub>18</sub>H<sub>18</sub>N<sub>2</sub>O(M+H)<sup>+</sup>: 267.1492, found 267.1493.



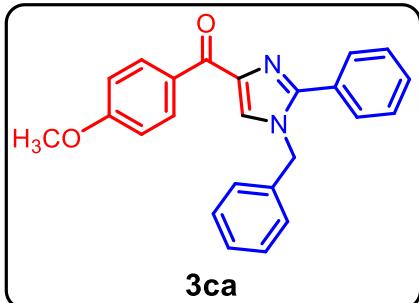
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(p-tolyl)methanone (3ba)**

According to the general procedure, **3ba** was obtained using (*E*)-3-(dimethylamino)-1-(p-tolyl)prop-2-en-1-one **1b** (37.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6 μL, 0.4 mmol) in 66% yield (46.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.151 (d, *J* = 8.4 Hz, 2H), 7.649 (s, 1H), 7.552 - 7.528 (m, 2H), 7.371 - 7.354 (m, 3H), 7.302 - 7.244 (m, 3H), 7.190 (d, *J* = 6.8 Hz, 2H), 7.041 - 7.017 (m, 2H), 5.173 (s, 2H), 2.328 (s, 3H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 187.58, 148.93, 143.01, 141.32, 135.90, 135.48, 130.52, 129.80, 129.65, 129.32, 129.30, 128.98, 128.80, 128.52, 128.49, 127.03, 51.10, 21.78.

**HRMS (ESI) *m/z*** calcd. for C<sub>24</sub>H<sub>21</sub>N<sub>2</sub>O (M+H)<sup>+</sup>: 353.1648, found 353.1644.



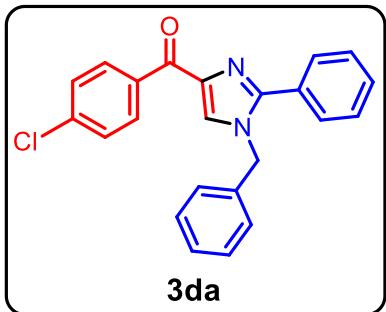
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(4-methoxyphenyl)methanone (3ca)**

According to the general procedure, **3ca** was obtained using (*E*)-3-(dimethylamino)-1-(4-methoxyphenyl)prop-2-en-1-one **1c** (41.0 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6 μL, 0.4 mmol) in 63% yield (46.4 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.457 - 8.427 (m, 2H), 7.757 (s, 1H), 7.632 - 7.608 (m, 2H), 7.462 - 7.428 (m, 3H), 7.381 - 7.321 (m, 3H), 7.125 - 7.102 (m, 2H), 6.980 - 6.943 (m, 2H), 5.251 (s, 2H), 3.863 (s, 3H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 186.27, 163.14, 148.63, 141.64, 135.93, 132.90, 130.82, 129.89, 129.61, 129.30, 129.28, 128.80, 128.46, 128.26, 127.05, 113.52, 55.51, 51.08.

**HRMS** (ESI) *m/z* calcd. for C<sub>24</sub>H<sub>21</sub>N<sub>2</sub>O<sub>2</sub> (M+H)<sup>+</sup>: 369.1598, found 369.1596.



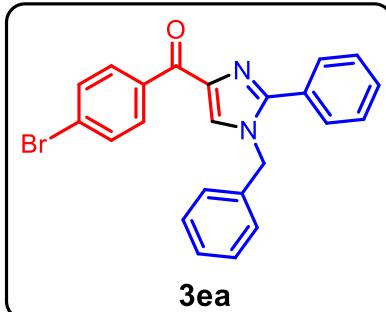
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(4-chlorophenyl)methanone (3da)**

According to the general procedure, **3da** was obtained using (*E*)-1-(4-chlorophenyl)-3-(dimethylamino)prop-2-en-1-one **1d** (41.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6 μL, 0.4 mmol) in 66% yield (49.2 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.386 - 8.345 (m, 2H), 7.788 (s, 1H), 7.622 - 7.598 (m, 2H), 7.475 (m, 4H), 7.437 - 7.421 (m, 1H), 7.381 - 7.330 (m, 3H), 7.124 - 7.100 (m, 2H), 5.254 (s, 2H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 186.38, 148.93, 141.13, 138.78, 136.22, 135.72, 132.06, 129.78, 129.68, 129.34, 129.29, 128.87, 128.71, 128.58, 128.52, 127.10, 51.19.

**HRMS** (ESI) *m/z* calcd. for C<sub>23</sub>H<sub>18</sub>ClN<sub>2</sub>O (M+H)<sup>+</sup>: 373.1102, found 373.1111.



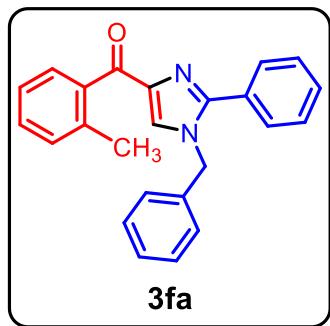
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(4-bromophenyl)methanone (3ea)**

According to the general procedure, **3ea** was obtained using (*E*)-1-(4-bromophenyl)-3-(dimethylamino)prop-2-en-1-one **1e** (41.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6 μL, 0.4 mmol) in 66% yield (49.2 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

thylamino)prop-2-en-1-one **1e** (50.6 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 73% yield (60.9 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.274 (d,  $J$  = 8.4 Hz, 2H), 7.784 (s, 1H), 7.604 (d,  $J$  = 8.0 Hz, 4H), 7.463 - 7.446 (m, 3H), 7.388 - 7.313 (m, 3H), 7.112 (d,  $J$  = 8.4 Hz, 2H), 5.252 (s, 2H);  
 **$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  186.57, 148.97, 141.09, 136.66, 135.72, 132.19, 131.52, 129.80, 129.68, 129.35, 129.30, 128.89, 128.74, 128.60, 127.54, 127.12, 51.21.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{17}\text{BrN}_2\text{ONa} (\text{M}+\text{Na})^+$ : 439.0416, found 439.0419.

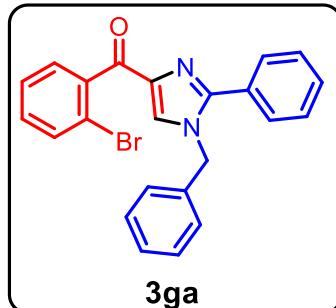


#### (1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(o-tolyl)methanone (**3fa**)

According to the general procedure, **3fa** was obtained using (*E*)-3-(dimethylamino)-1-(o-tolyl)prop-2-en-1-one **1f** (37.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 77% yield (54.2 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.675 - 7.638 (m, 3H), 7.544 (s, 1H), 7.491 - 7.464 (m, 3H), 7.391 (t,  $J$  = 7.6 Hz, 4H), 7.320 - 7.278 (m, 2H), 7.140 - 7.117 (m, 2H), 5.281 (s, 2H), 2.500 (s, 3H);  
 **$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  190.98, 149.85, 141.34, 138.84, 137.27, 135.80, 131.13, 130.31, 129.76, 129.44, 129.31, 129.28, 129.19, 129.04, 128.73, 128.48, 126.87, 125.13, 51.08, 20.16.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{24}\text{H}_{21}\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 353.1648, found 353.1653.



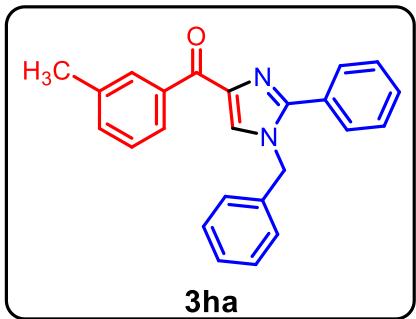
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(2-bromophenyl)methanone (3ga)**

According to the general procedure, **3ga** was obtained using (*E*)-1-(2-bromophenyl)-3-(dimethylamino)prop-2-en-1-one **1g** (50.6 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 66% yield (57.5 mg) as a yellow solid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.303 (d,  $J$  = 7.2 Hz, 2H), 7.750 (s, 1H), 7.636 - 7.612 (m, 2H), 7.487 (s, 1H), 7.468 - 7.441 (m, 4H), 7.348 (t,  $J$  = 7.6 Hz, 3H), 7.126 - 7.103 (m, 2H), 5.265 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.89, 149.07, 141.20, 138.12, 135.87, 132.37, 130.41, 129.78, 129.72, 129.36, 129.34, 128.84, 128.73, 128.54, 128.28, 127.07, 51.17.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{18}\text{BrN}_2\text{O} (\text{M}+\text{H})^+$ : 417.0597, found 417.0588.



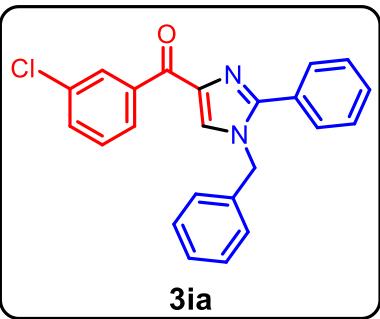
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(m-tolyl)methanone (3ha)**

According to the general procedure, **3ha** was obtained using (*E*)-3-(dimethylamino)-1-(m-tolyl)prop-2-en-1-one **1h** (37.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 75% yield (52.8 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.105 - 8.078 (m, 1H), 8.016 (s, 1H), 7.708 (s, 1H), 7.634 - 7.609 (m, 2H), 7.455 - 7.437 (m, 3H), 7.364 - 7.346 (m, 5H), 7.114 - 7.100 (m, 2H), 5.261 (s, 2H), 2.416 (s, 3H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  188.22, 149.15, 141.12, 138.22, 138.03, 135.90, 133.17, 130.62, 129.76, 129.72, 129.37, 129.34, 128.83, 128.71, 128.54, 128.15, 127.65, 127.07, 51.16, 21.58.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{24}\text{H}_{21}\text{N}_2\text{O} (\text{M}+\text{H})^+$ : 353.1648, found 353.1654.



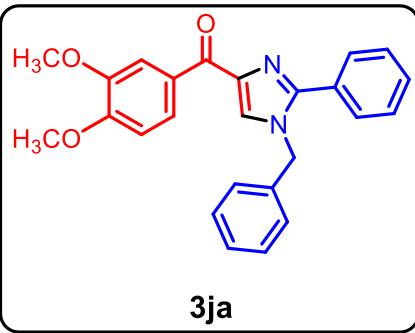
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(3-chlorophenyl)methanone (3ia)**

According to the general procedure, **3ia** was obtained using (*E*)-1-(3-chlorophenyl)-3-(dimethylamino)prop-2-en-1-one **1i** (41.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 69% yield (51.5 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.311 (s, 1H), 8.267 (d,  $J = 8.0$  Hz, 1H), 7.777 (s, 1H), 7.631 - 7.607 (m, 2H), 7.526 - 7.498 (m, 1H), 7.478 - 7.452 (m, 3H), 7.427 - 7.407 (m, 1H), 7.388 - 7.354 (m, 3H), 7.127 - 7.104 (m, 2H), 5.264 (s, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  186.33, 149.15, 140.85, 139.53, 135.70, 134.40, 132.35, 130.46, 129.84, 129.61, 129.39, 129.33, 128.99, 128.91, 128.73, 128.64, 127.13, 51.25.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{23}\text{H}_{18}\text{ClN}_2\text{O} (\text{M}+\text{H})^+$ : 373.1102, found 373.1111.



**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(3,4-dimethoxyphenyl)methanone (3ja)**

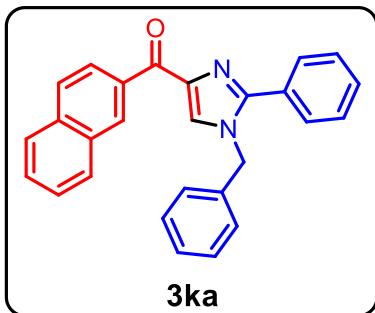
According to the general procedure, **3ja** was obtained using (*E*)-1-(3,4-dimethoxyphenyl)-3-(dimethylamino)prop-2-en-1-one **1j** (47.0 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 78% yield (62.1 mg) as brown oil filled liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.271 - 8.240 (m, 1H), 7.947 (s, 1H), 7.754 (s, 1H), 7.628 - 7.603 (m, 2H), 7.461 - 7.425 (m, 3H), 7.360 - 7.321 (m, 3H), 7.130 - 7.110 (m, 2H), 6.931 (d,  $J =$

8.4 Hz, 1H), 5.258 (s, 2H), 3.943 (d,  $J$  = 2.8 Hz, 6H);

**$^{13}\text{C}$  NMR** (100 MHz,  $\text{CDCl}_3$ )  $\delta$  186.08, 152.92, 148.73, 148.67, 141.56, 135.91, 130.85, 129.86, 129.64, 129.31, 129.25, 128.83, 128.51, 128.38, 127.08, 125.66, 112.80, 110.14, 56.12, 56.08, 51.12.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{25}\text{H}_{23}\text{N}_2\text{O}_3$  ( $\text{M}+\text{H}$ ) $^+$ : 399.1703, found 399.1704.



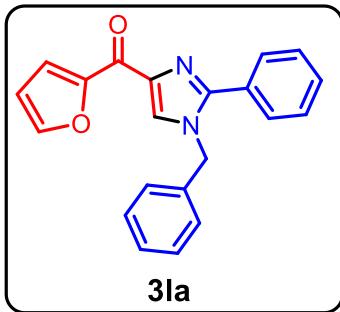
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(naphthalen-2-yl)methanone (3ka)**

According to the general procedure, **3ka** was obtained using (*E*)-3-(dimethylamino)-1-(naphthalen-2-yl)prop-2-en-1-one **1k** (45.0 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 76% yield (59.0 mg) as brown oil filled liquid (silica gel flash ch chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H}$  NMR** (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.978 (s, 1H), 8.293 - 8.268 (m, 1H), 7.987 (d,  $J$  = 8.0 Hz, 1H), 7.920 - 7.861 (m, 2H), 7.794 (s, 1H), 7.665 - 7.641 (m, 2H), 7.597 - 7.501 (m, 3H), 7.472 - 7.456 (m, 2H), 7.370 - 7.331 (m, 3H), 7.152 - 7.118 (m, 2H), 5.286 (s, 2H).

**$^{13}\text{C}$  NMR** (100 MHz,  $\text{CDCl}_3$ )  $\delta$  187.68, 149.13, 141.13, 135.81, 135.45, 135.36, 132.66, 132.30, 129.91, 129.82, 129.61, 129.38, 129.35, 129.21, 128.79, 128.58, 128.17, 128.05, 127.82, 127.12, 126.49, 126.10, 51.24.

**HRMS** (ESI)  $m/z$  calcd. for  $\text{C}_{27}\text{H}_{20}\text{N}_2\text{ONa}$  ( $\text{M}+\text{Na}$ ) $^+$ : 411.1468, found 411.1467.



**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(furan-2-yl)methanone (3la)**

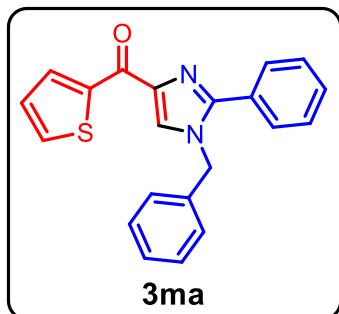
According to the general procedure, **3la** was obtained using (*E*)-3-(dimethylamino)-1-(furan-2-yl)prop-2-en-1-one **1l** (33.0 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$

L, 0.4 mmol) in 73% yield (47.9 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.094 (d, *J* = 3.6 Hz, 1H), 7.908 (s, 1H), 7.661 - 7.657 (m, 1H), 7.626 - 7.602 (m, 2H), 7.469 - 7.432 (m, 3H), 7.378 - 7.319 (m, 3H), 7.118 - 7.094 (m, 2H), 6.575 - 6.561 (m, 1H), 5.254 (s, 2H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 174.41, 152.05, 148.95, 146.72, 140.02, 135.77, 129.75, 129.69, 129.32, 129.26, 128.85, 128.54, 128.11, 127.08, 121.70, 112.34, 51.21.

**HRMS** (ESI) *m/z* calcd. for C<sub>21</sub>H<sub>16</sub>N<sub>2</sub>O<sub>2</sub>Na (M+Na)<sup>+</sup>: 351.1104, found 351.1113.



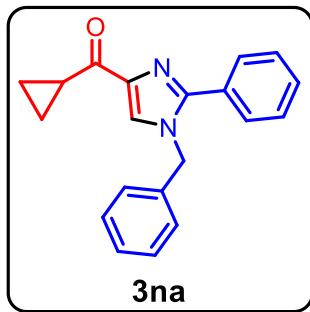
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(thiophen-2-yl)methanone (3ma)**

According to the general procedure, **3ma** was obtained using (*E*)-3-(dimethylamino)-1-(thiophen-2-yl)prop-2-en-1-one **1m** (36.2 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6 μL, 0.4 mmol) in 77% yield (53.0 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>) δ 8.680 - 8.667 (m, 1H), 7.834 (s, 1H), 7.668 - 7.628 (m, 3H), 7.468 - 7.445 (m, 3H), 7.369 - 7.322 (m, 3H), 7.177 - 7.155 (m, 1H), 7.125 - 7.101 (m, 2H), 5.266 (s, 2H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>) δ 179.21, 148.59, 143.05, 140.93, 135.77, 135.62, 134.16, 129.78, 129.68, 129.30, 129.20, 128.87, 128.51, 128.03, 127.78, 127.07, 51.21.

**HRMS** (ESI) *m/z* calcd. for C<sub>21</sub>H<sub>16</sub>N<sub>2</sub>OSK (M+K)<sup>+</sup>: 383.0615, found 383.0606.



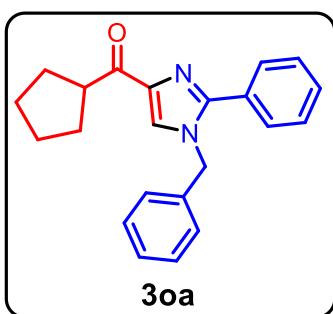
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(cyclopropyl)methanone (3na)**

According to the general procedure, **3na** was obtained using (*E*)-1-cyclopropyl-3-(dimethylamino)prop-2-en-1-one **1n** (27.8 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 52% yield (31.4 mg) as brown oil filled liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.651 (s, 1H), 7.590 - 7.566 (m, 2H), 7.455 - 7.427 (m, 3H), 7.363 7.331 (m, 3H), 7.094 - 7.070 (m, 2H), 5.209 (s, 2H), 3.125 - 3.062 (m, 1H), 1.231 - 1.192 (m, 2H), 1.029 - 0.983 (m, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  196.61, 148.90, 141.92, 135.81, 129.77, 129.34, 129.31, 128.92, 128.54, 127.13, 124.83, 51.17, 17.23, 11.80.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{20}\text{H}_{18}\text{N}_2\text{O}\text{Na} (\text{M}+\text{Na})^+$ : 325.1311, found 325.1316.



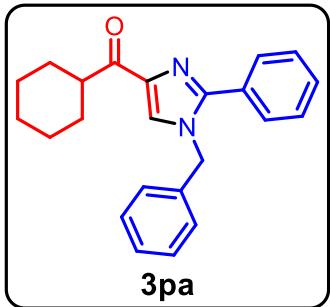
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(cyclopentyl)methanone (3oa)**

According to the general procedure, **3oa** was obtained using (*E*)-1-cyclopentyl-3-(dimethylamino)prop-2-en-1-one **1o** (33.4 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 80% yield (52.8 mg) as yellow oily liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**$^1\text{H NMR}$**  (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.637 (s, 1H), 7.569 - 7.544 (m, 2H), 7.451 - 7.415 (m, 3H), 7.366 - 7.315 (m, 3H), 7.093 - 7.070 (m, 2H), 5.186 (s, 2H), 3.944 - 3.863 (m, 1H), 1.990 - 1.927 (m, 2H), 1.904 - 1.848 (m, 2H), 1.756 - 1.714 (m, 2H), 1.659 - 1.599 (m, 2H);

**$^{13}\text{C NMR}$**  (100 MHz,  $\text{CDCl}_3$ )  $\delta$  199.73, 148.72, 141.54, 135.90, 129.89, 129.65, 129.34, 129.28, 128.87, 128.49, 127.11, 125.52, 51.08, 46.85, 30.09, 26.46.

**HRMS (ESI)**  $m/z$  calcd. for  $\text{C}_{22}\text{H}_{22}\text{N}_2\text{O}\text{Na} (\text{M}+\text{Na})^+$ : 353.1624, found 353.1624.



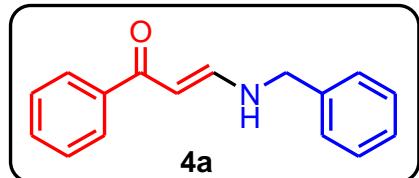
**(1-benzyl-2-phenyl-1*H*-imidazol-4-yl)(cyclohexyl)methanone (3pa)**

According to the general procedure, **3pa** was obtained using (*E*)-1-cyclohexyl-3-(dimethylamino)prop-2-en-1-one **1p** (36.2 mg, 0.2 mmol) and phenylmethanamine **2a** (42.8 mg, 43.6  $\mu$ L, 0.4 mmol) in 83% yield (57.1 mg) as brown oil filled liquid (silica gel flash chromatography: petroleum ether/EtOAc = 6:1).

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>)  $\delta$  7.637 (s, 1H), 7.570 - 7.546 (m, 2H), 7.462 - 7.416 (m, 3H), 7.364 - 7.316 (m, 3H), 7.090 - 7.066 (m, 2H), 5.185 (s, 2H), 3.522 - 3.448 (m, 1H), 1.967 - 1.929 (m, 2H), 1.821 - 1.773 (m, 2H), 1.725 - 1.674 (m, 1H), 1.537 - 1.348 (m, 5H);

**<sup>13</sup>C NMR** (100 MHz, CDCl<sub>3</sub>)  $\delta$  200.32, 148.68, 140.88, 135.86, 129.77, 129.70, 129.37, 129.29, 128.88, 128.49, 127.10, 125.72, 51.10, 45.75, 29.07, 26.17, 25.84.

**HRMS (ESI)** *m/z* calcd. for C<sub>23</sub>H<sub>25</sub>N<sub>2</sub>O (M+H)<sup>+</sup>: 345.1961, found 345.1968.



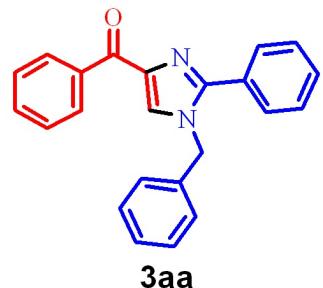
**(E)-3-(benzylamino)-1-phenylprop-2-en-1-one (4a)<sup>2</sup>**

**<sup>1</sup>H NMR** (400 MHz, CDCl<sub>3</sub>)  $\delta$  10.615 (brs, 1H), 7.897 - 7.876 (m, 2H), 7.432 - 7.407 (m, 3H), 7.364 - 7.342 (m, 2H), 7.317 - 7.285 (m, 2H), 7.042 - 6.986 (m, 1H), 5.789 - 5.766 (m, 1H), 4.469 - 4.449 (m, 2H).

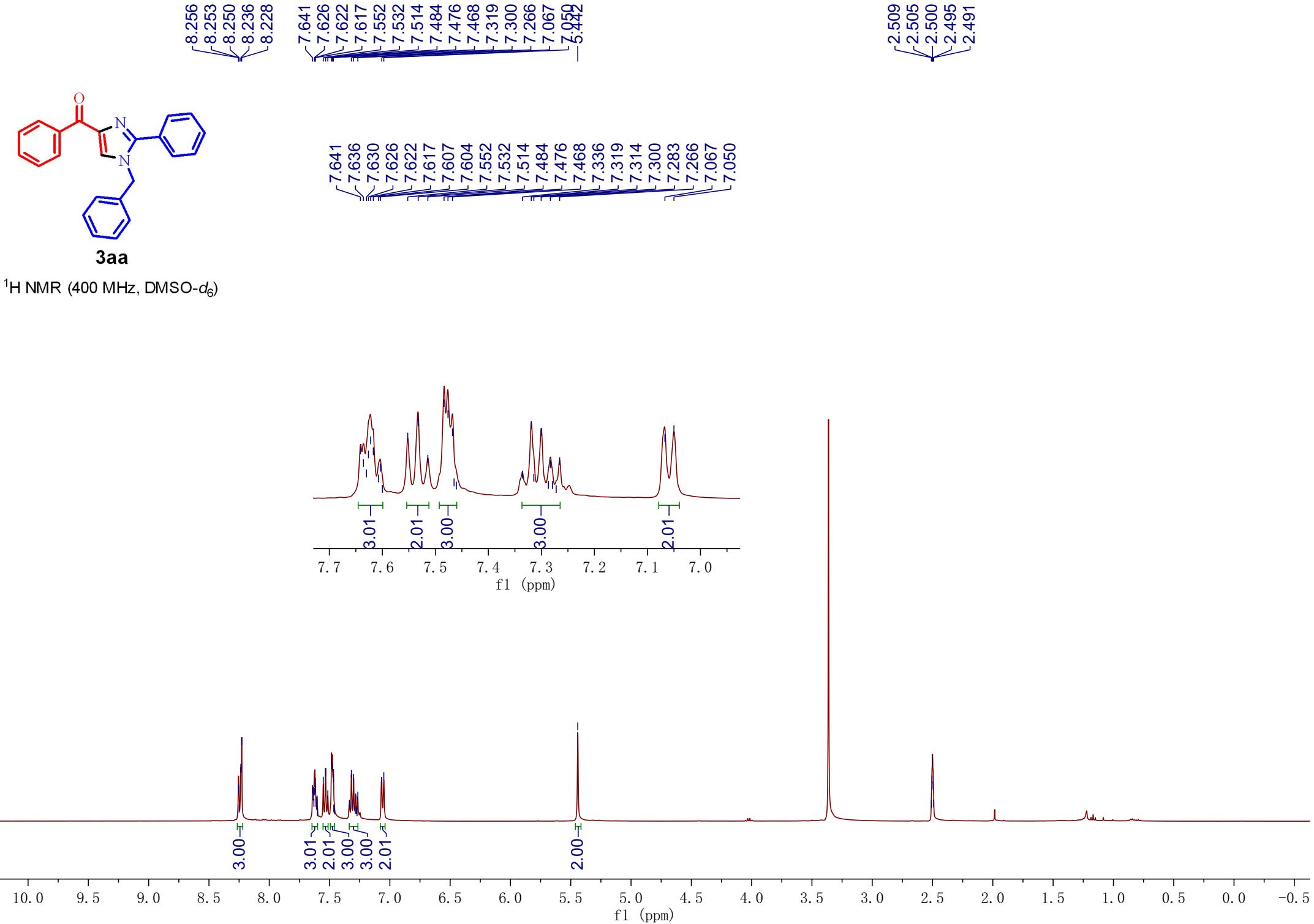
[1]. Wang, F.; Sun, W.; Wang, Y.; Jiang, Y.; Loh, T.-P. J. O. l., Highly site-selective metal-free C-H acyloxylation of stable enamines. Org. Lett., **2018**, *20* (4), 1256-1260.

[2]. Zhou, S.; Liu, D.-Y.; Wang, S.; Tian, J.-S.; Loh, T.-P., An efficient method for the synthesis of 2-pyridones via C-H bond functionalization. Chem. Commun. 2020, *56*(95), 15020-15023.

## (F) Spectra



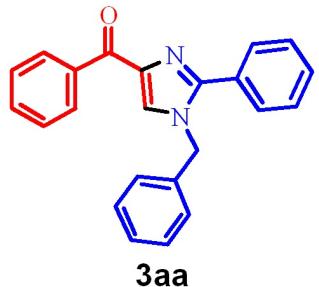
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )



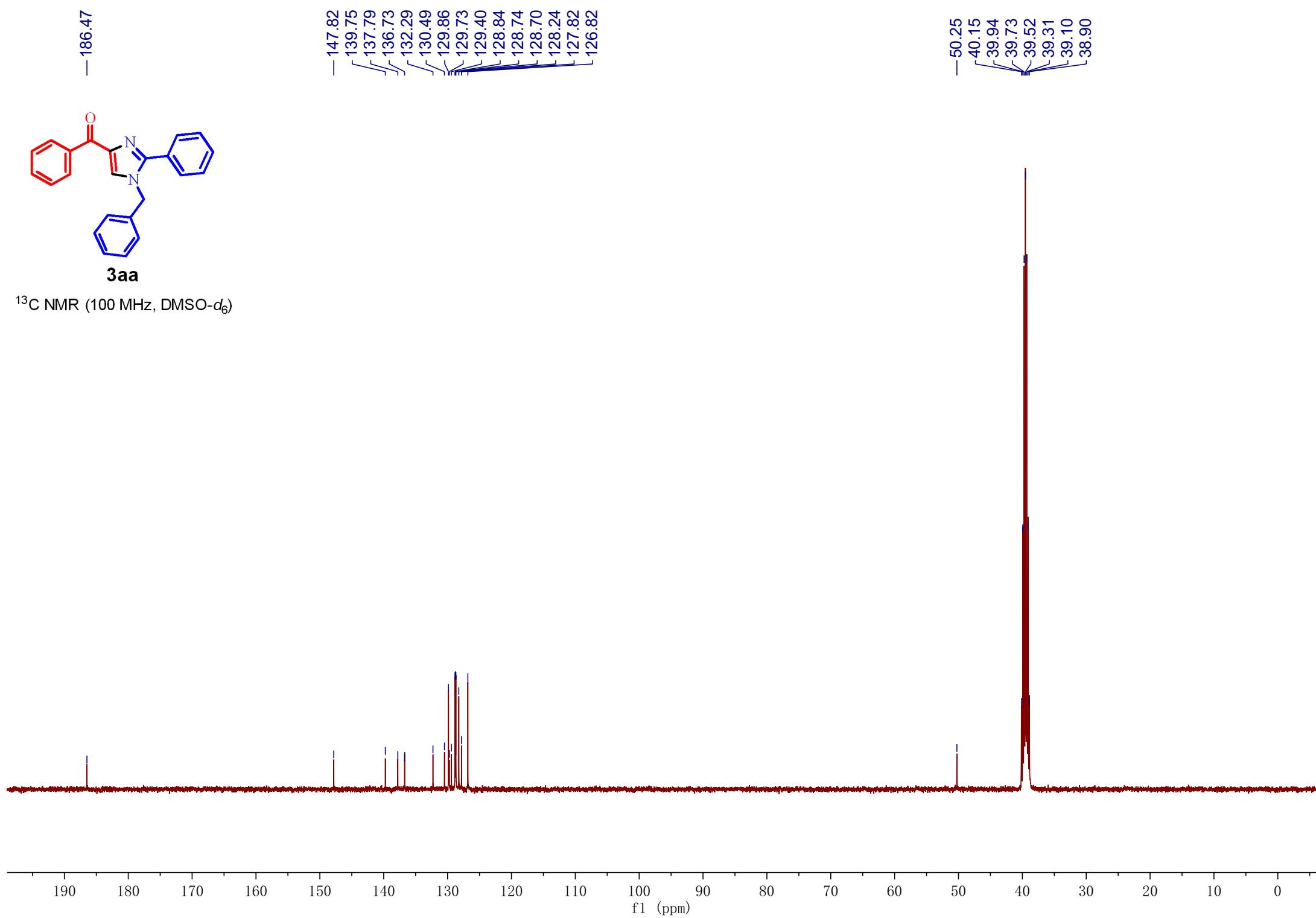
-186.47

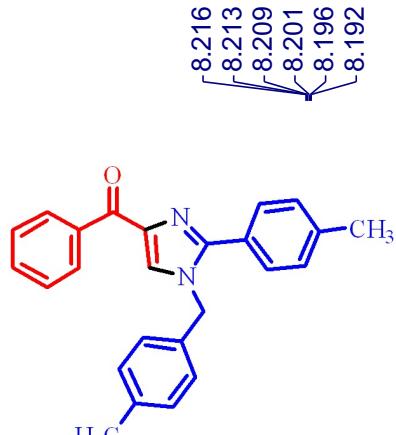
-147.82  
-139.75  
-137.79  
-136.73  
-132.29  
-130.49  
-129.86  
-129.73  
-129.40  
-128.84  
-128.74  
-128.70  
-128.24  
-127.82  
-126.82

-50.25  
-40.15  
-39.94  
-39.73  
-39.52  
-39.31  
-39.10  
-38.90



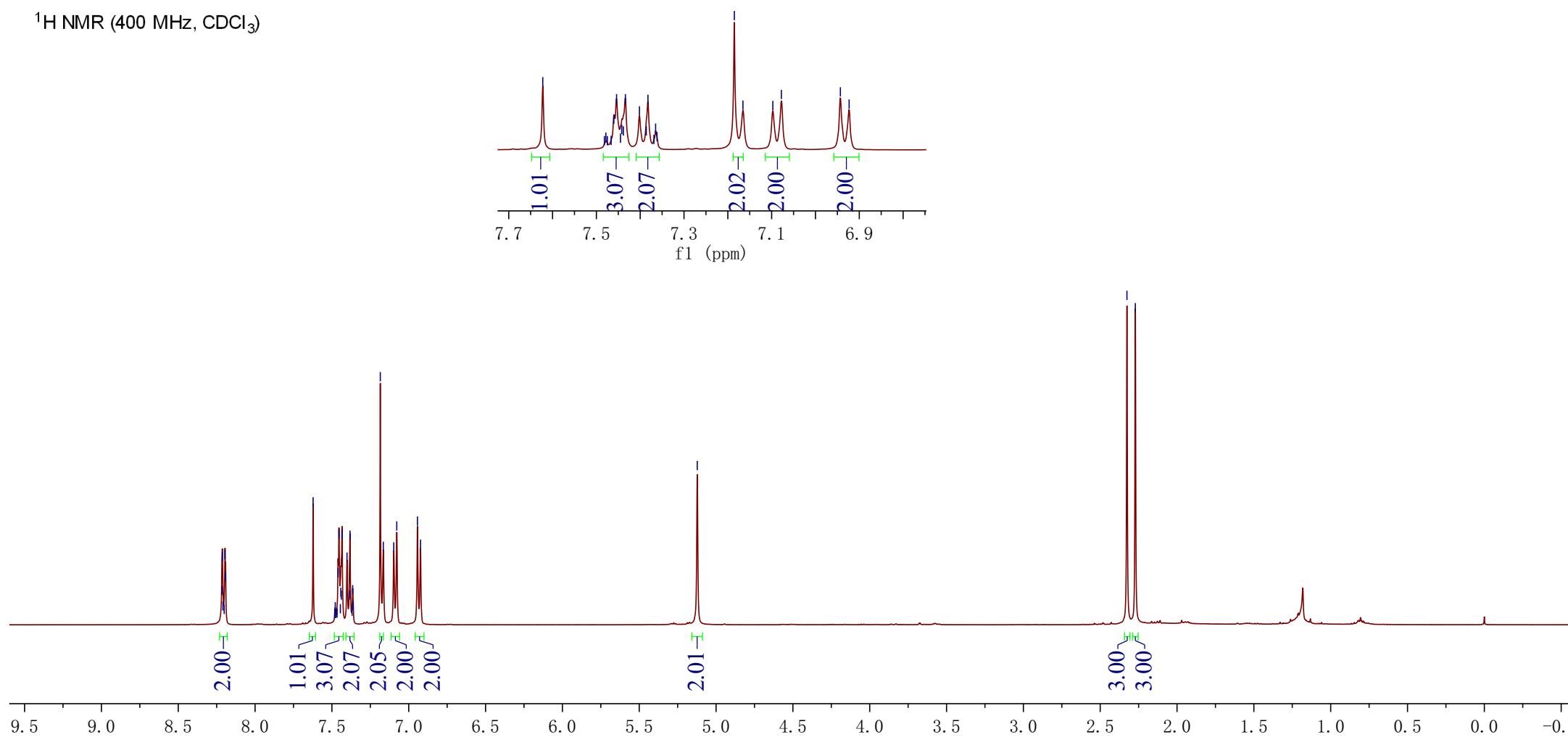
$^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO}-d_6$ )

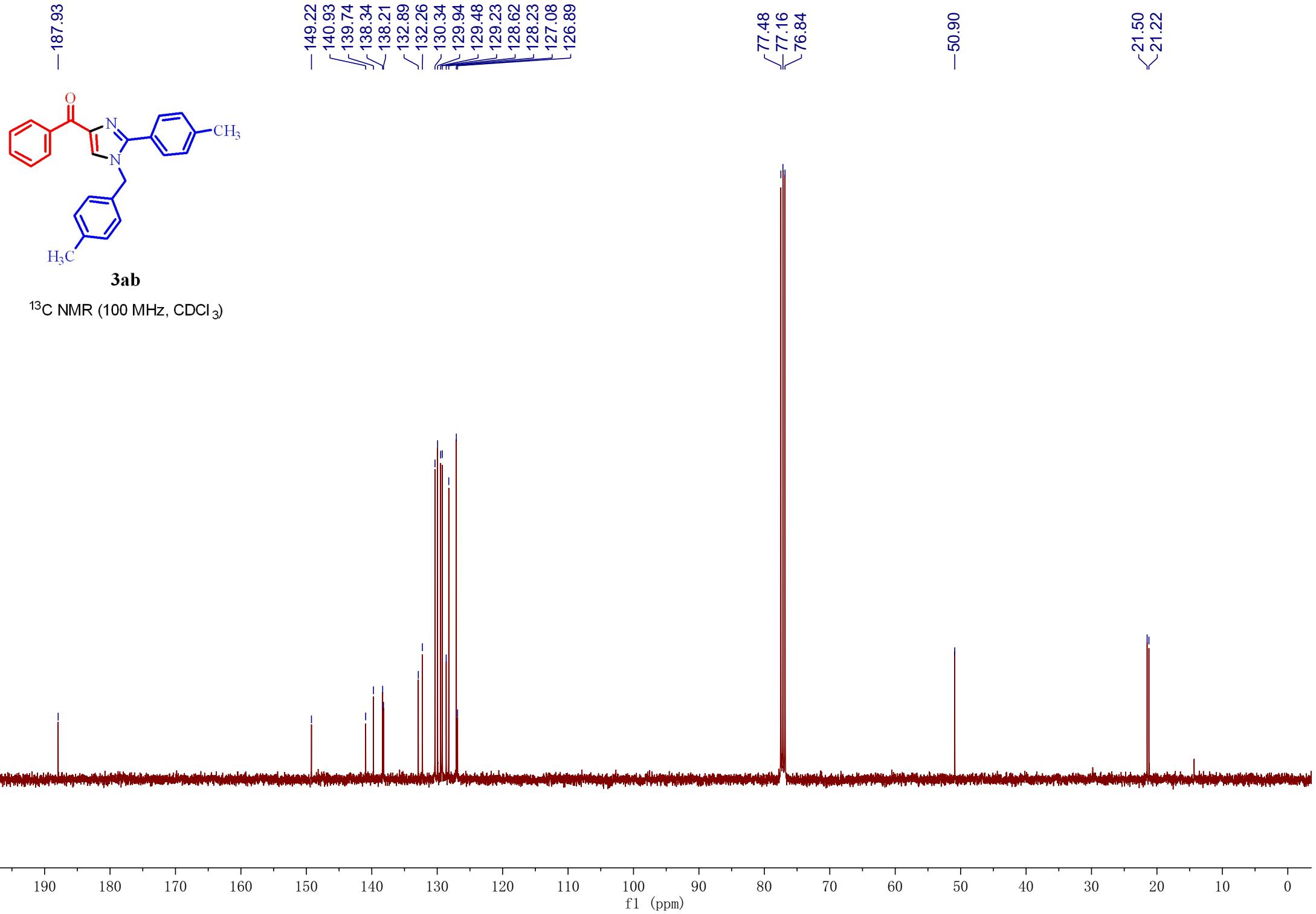


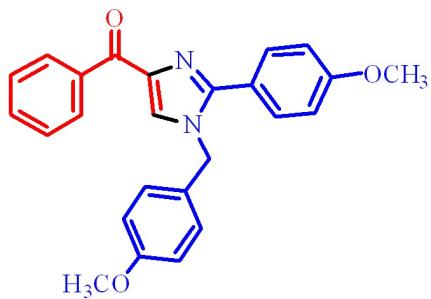


3ab

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

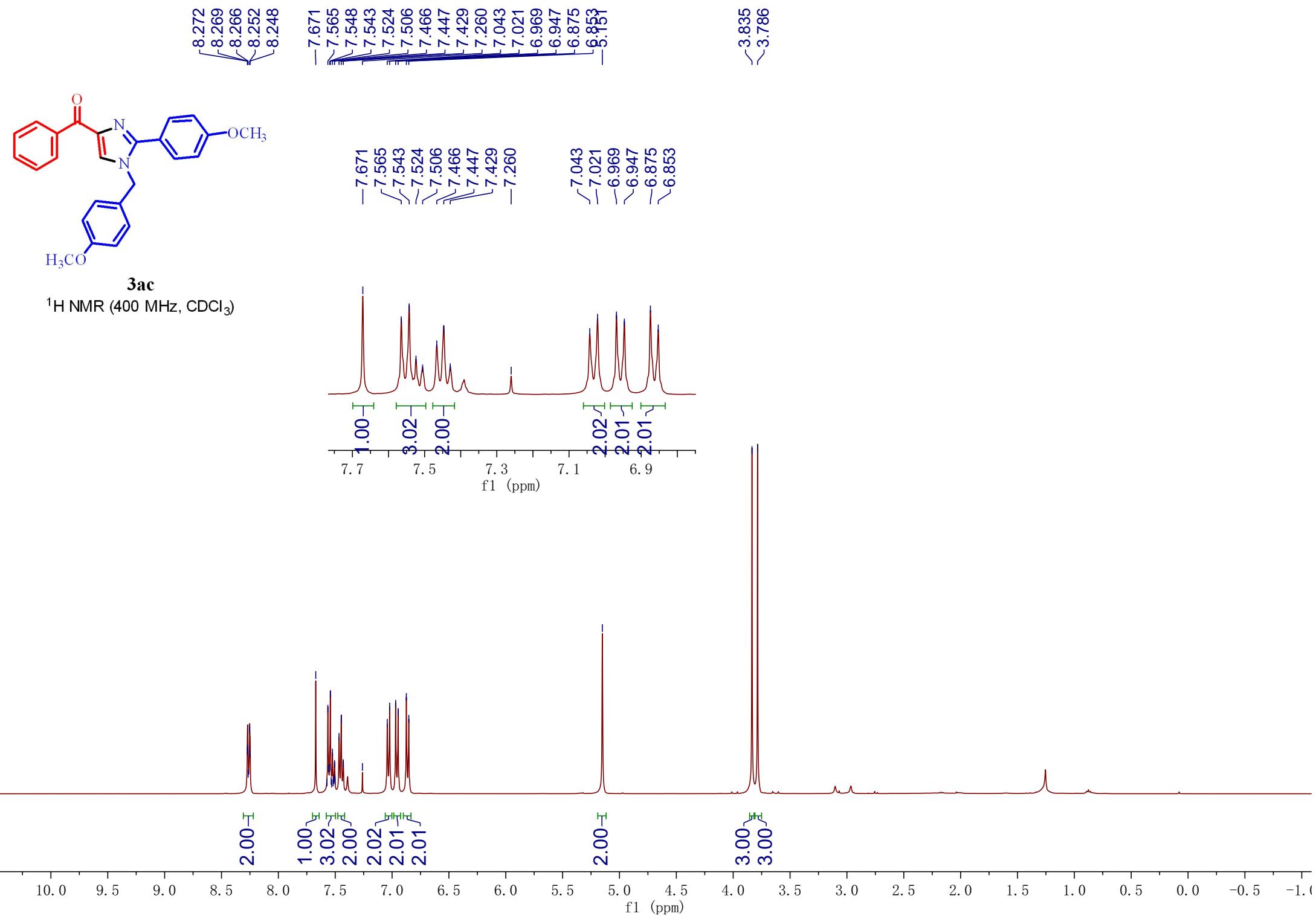


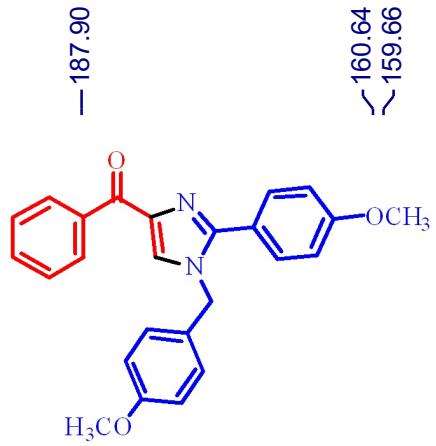




3ac

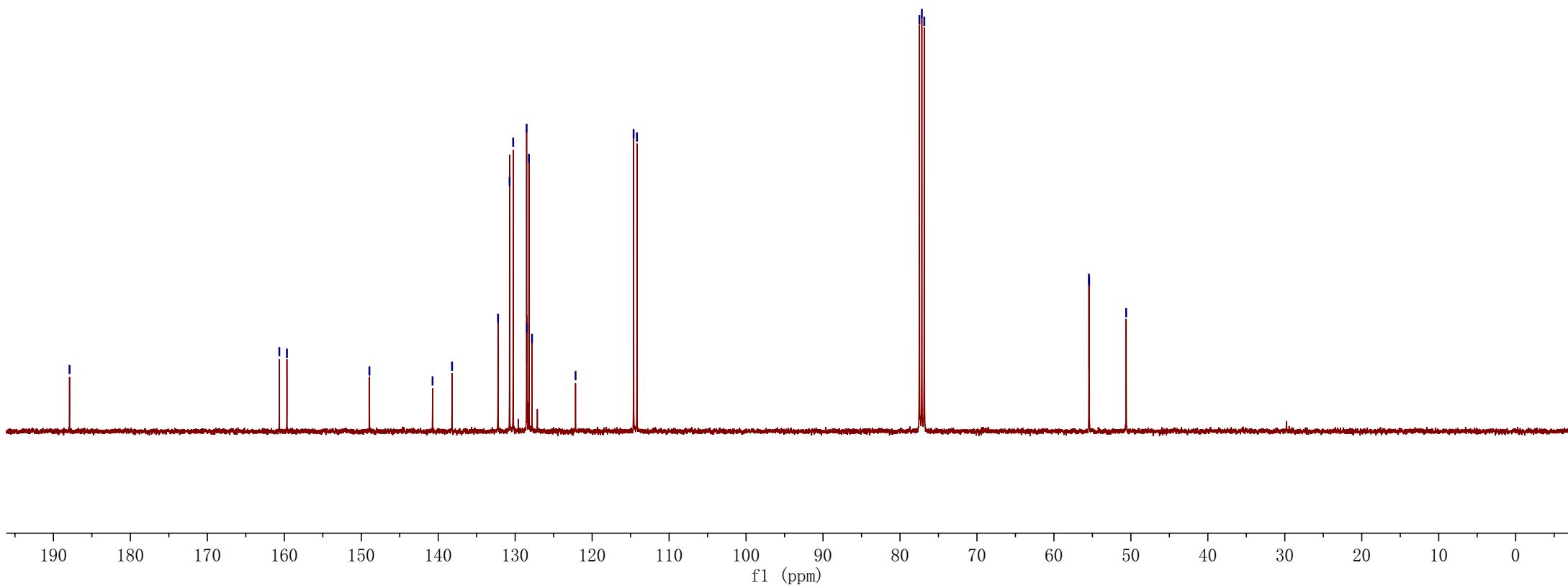
<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

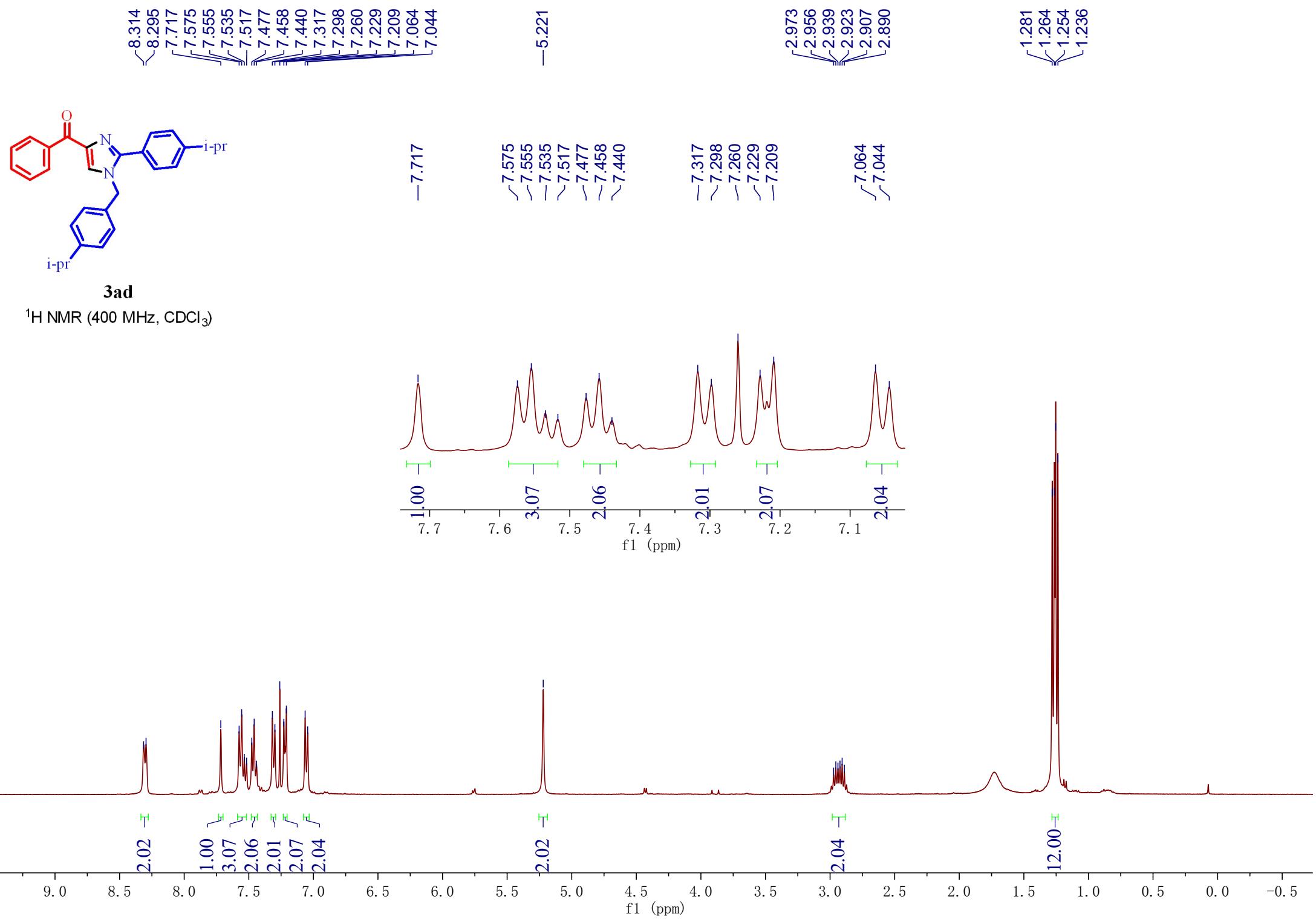




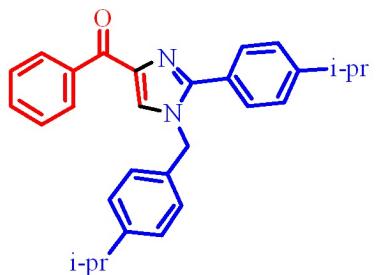
**3ac**

$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )





-187.98



**3ad**

<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)

150.65  
149.35  
149.25  
141.07  
138.20  
133.29  
132.30  
130.44  
129.35  
128.62  
128.24  
127.37  
127.20  
126.94

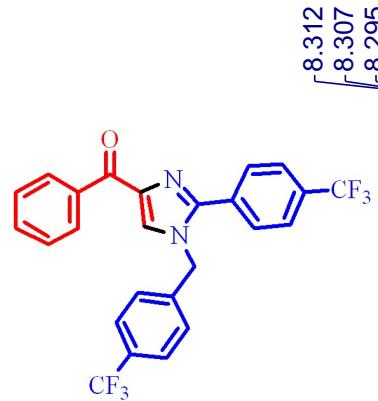
77.48  
77.16  
76.84

-50.92

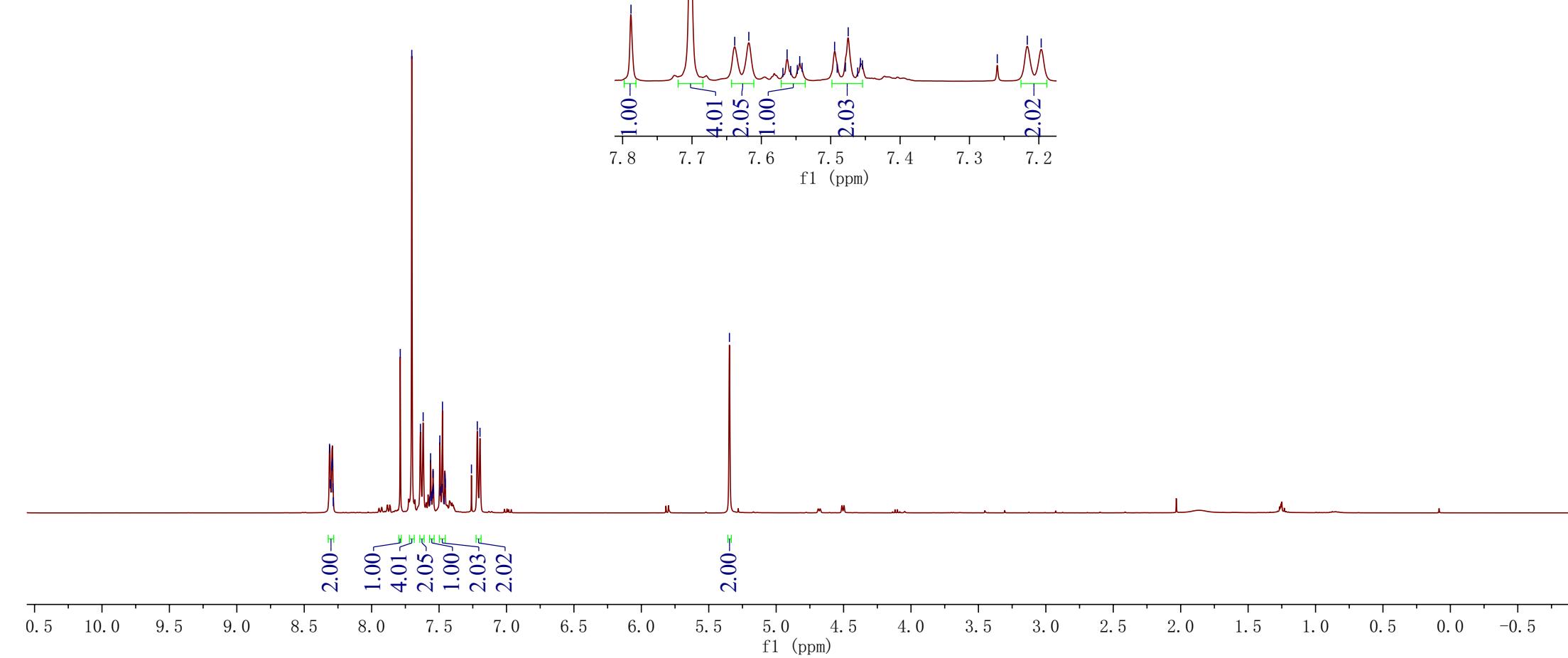
34.17  
33.95  
~29.84  
24.04  
23.98

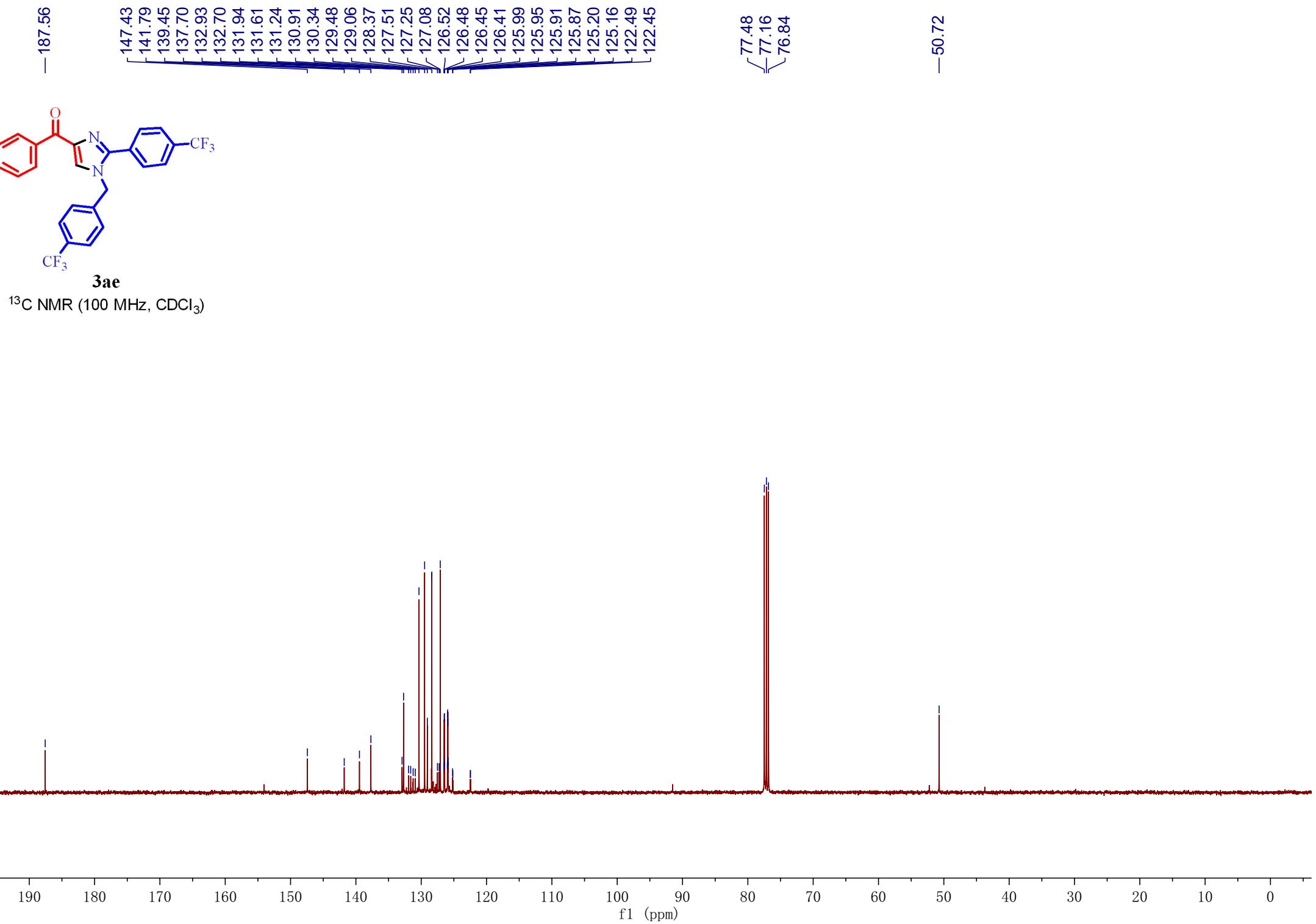
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0

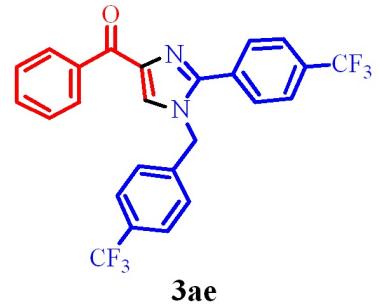
f1 (ppm)



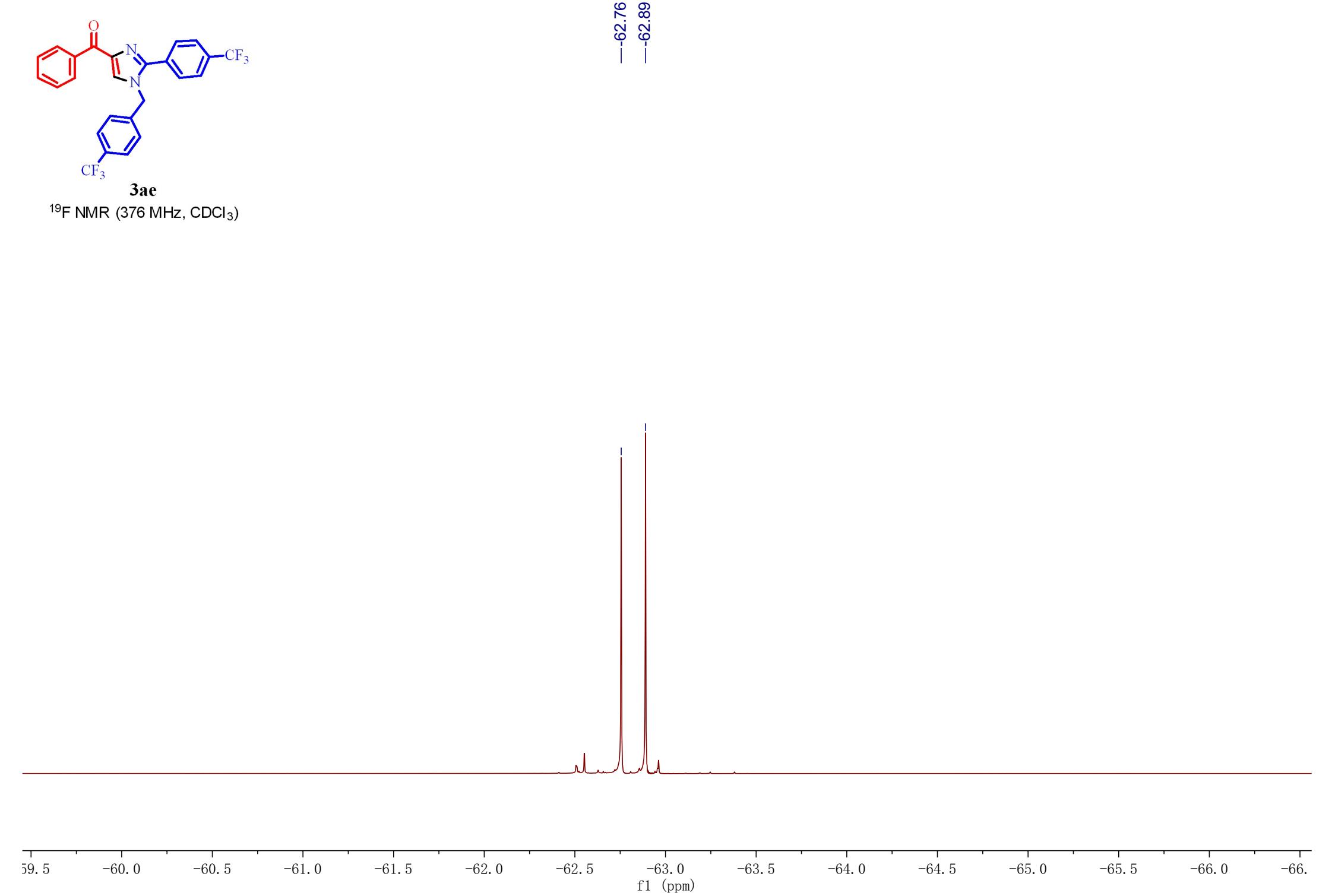
<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

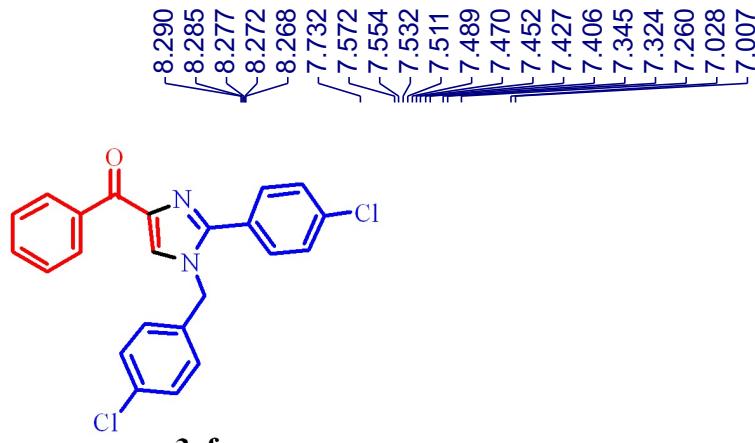




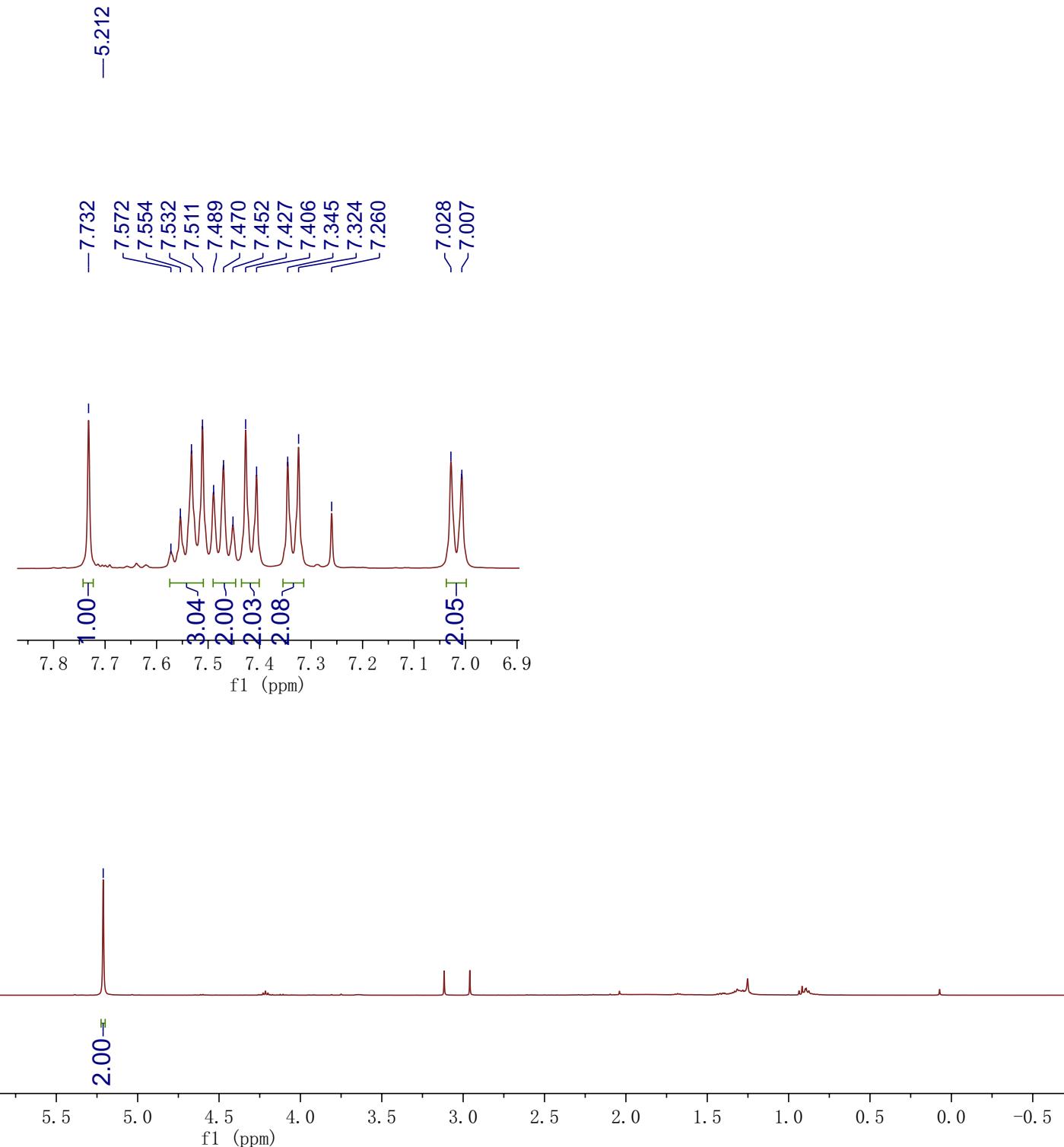


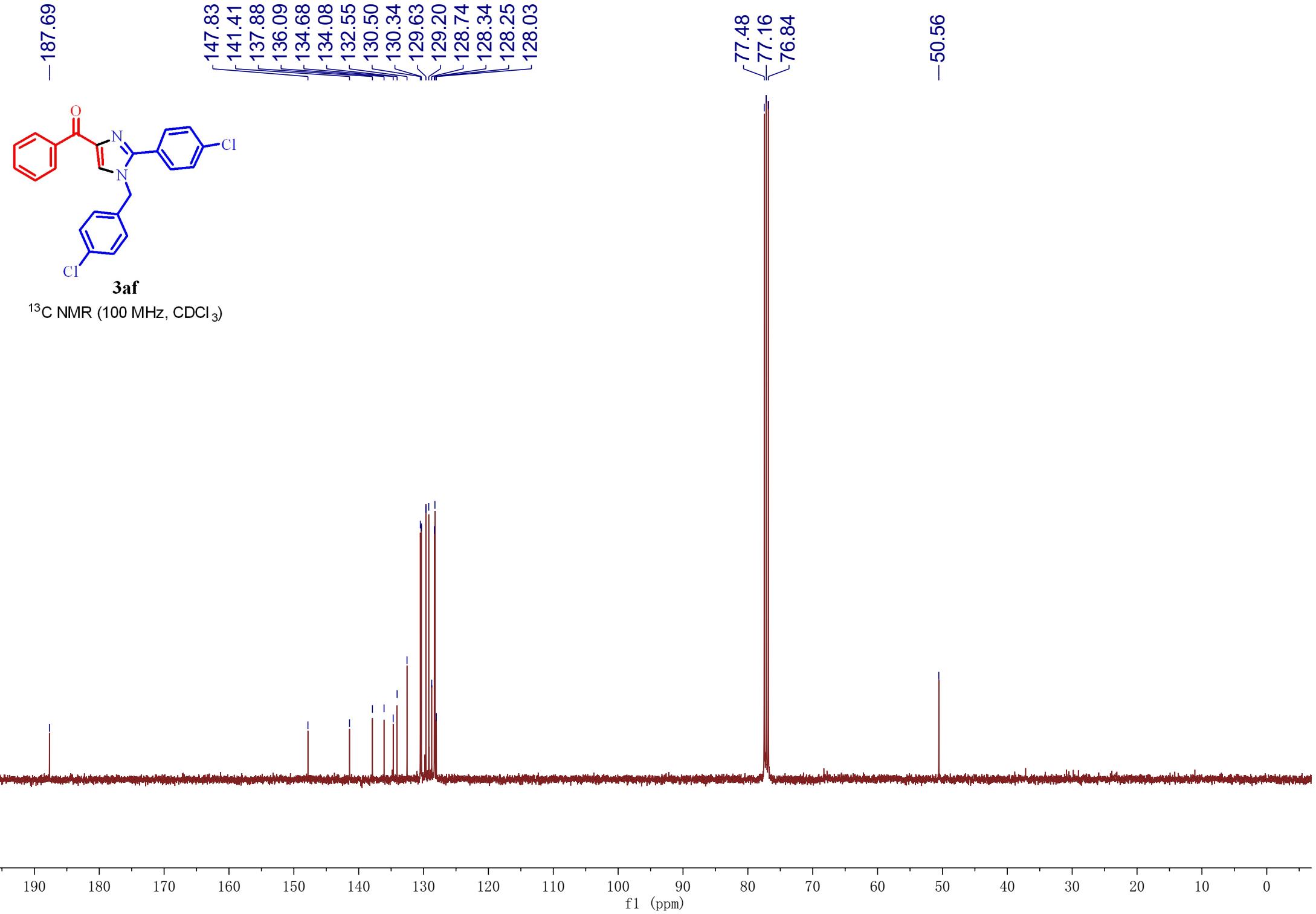
$^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ )

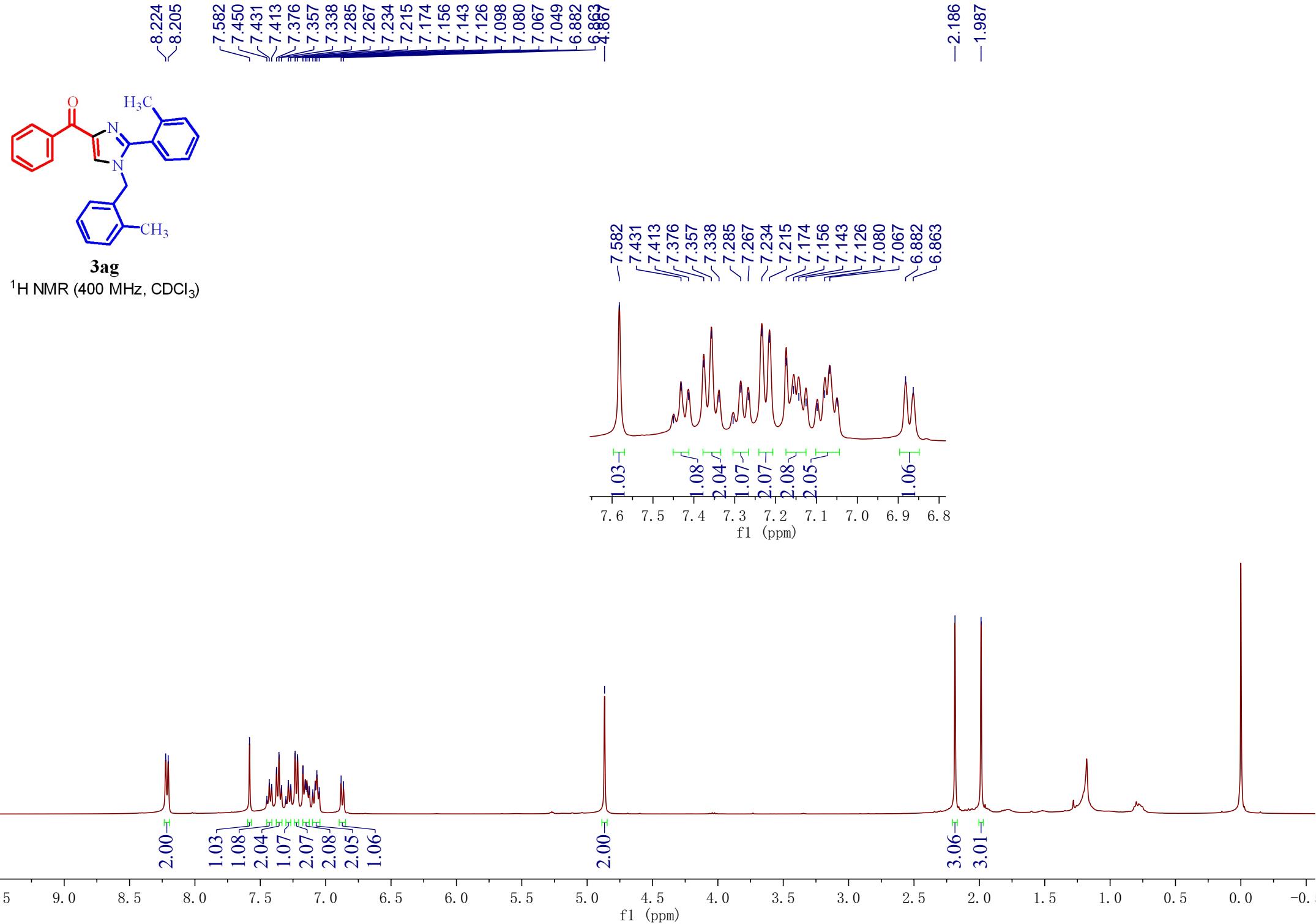


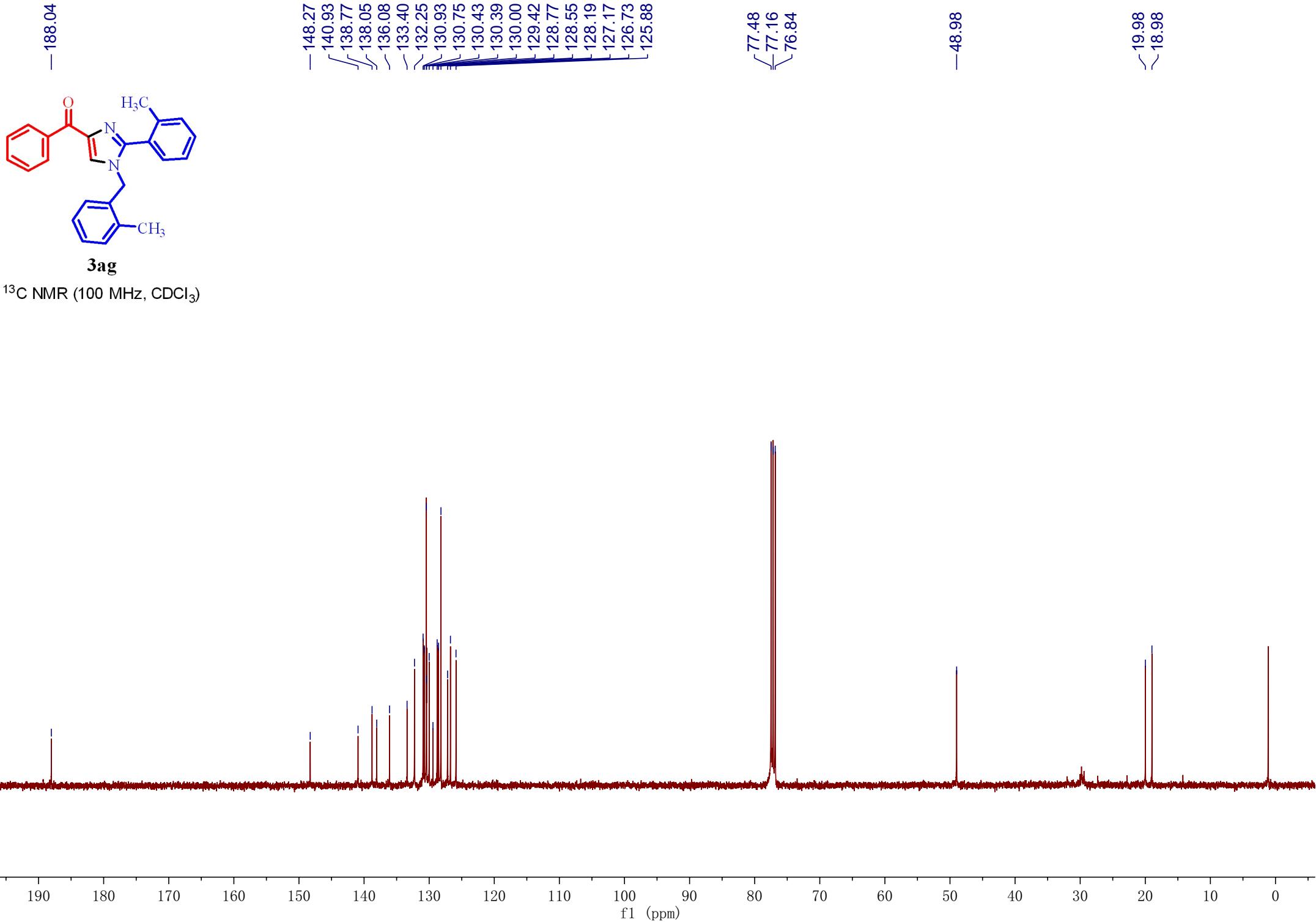


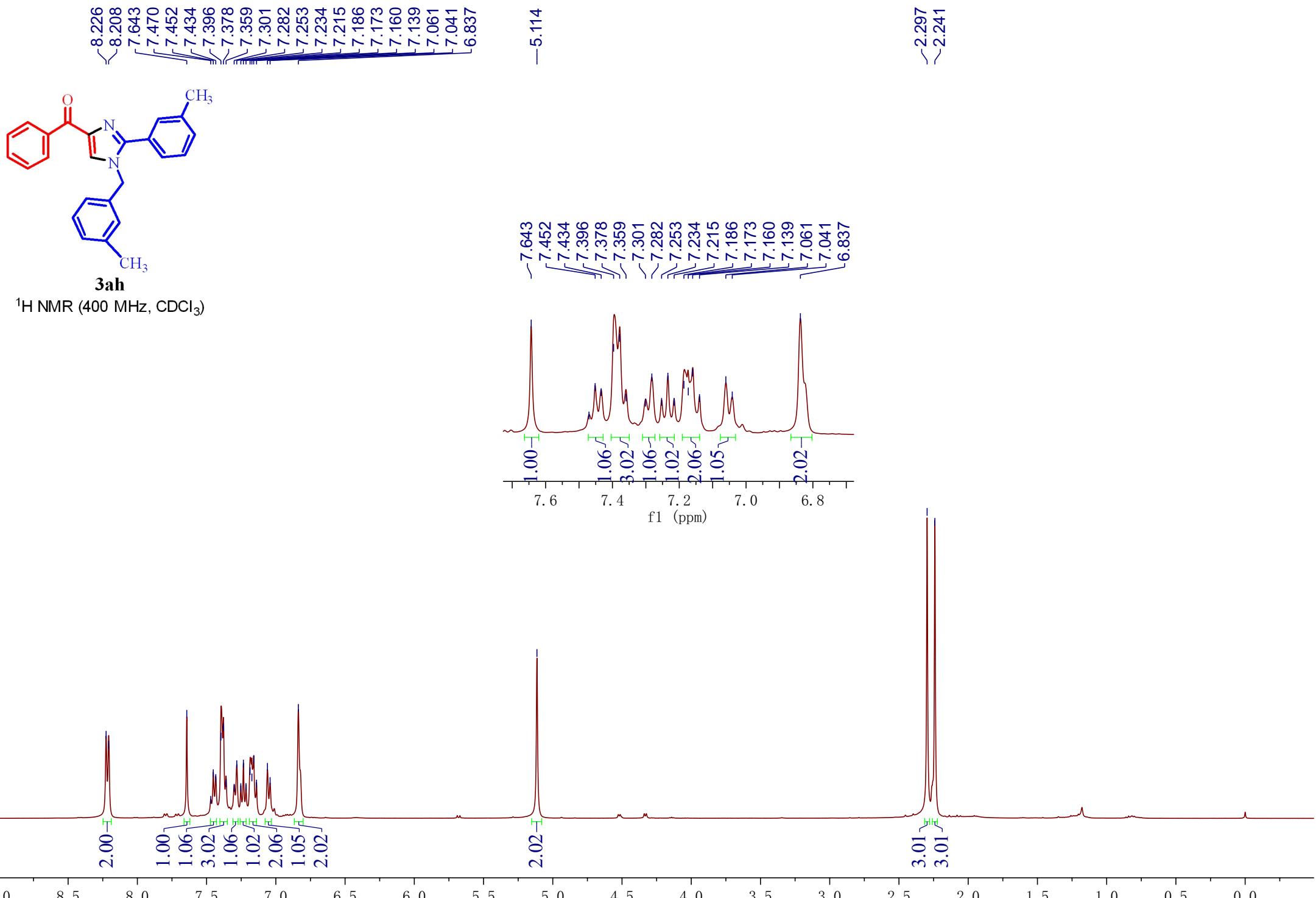
<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

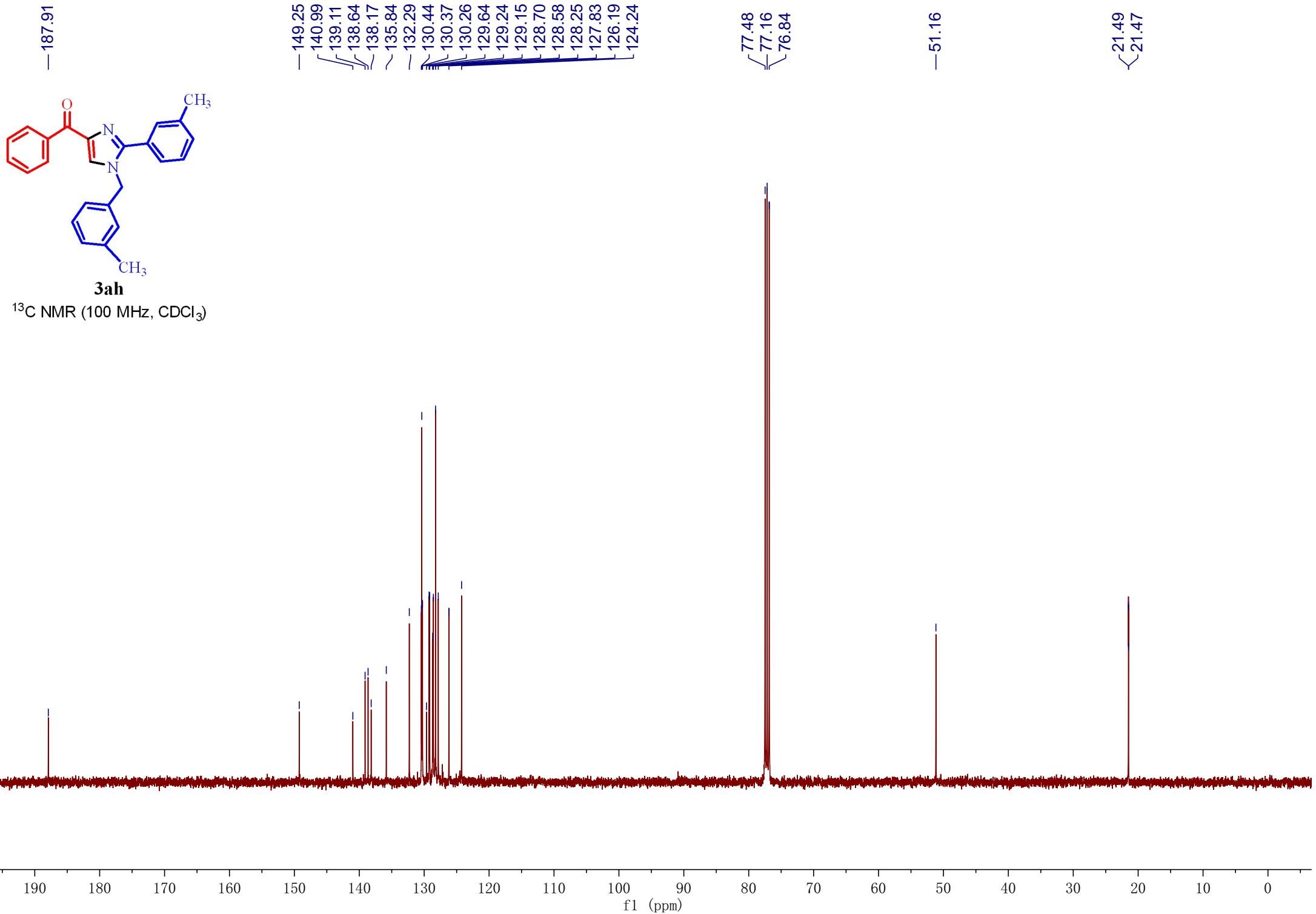


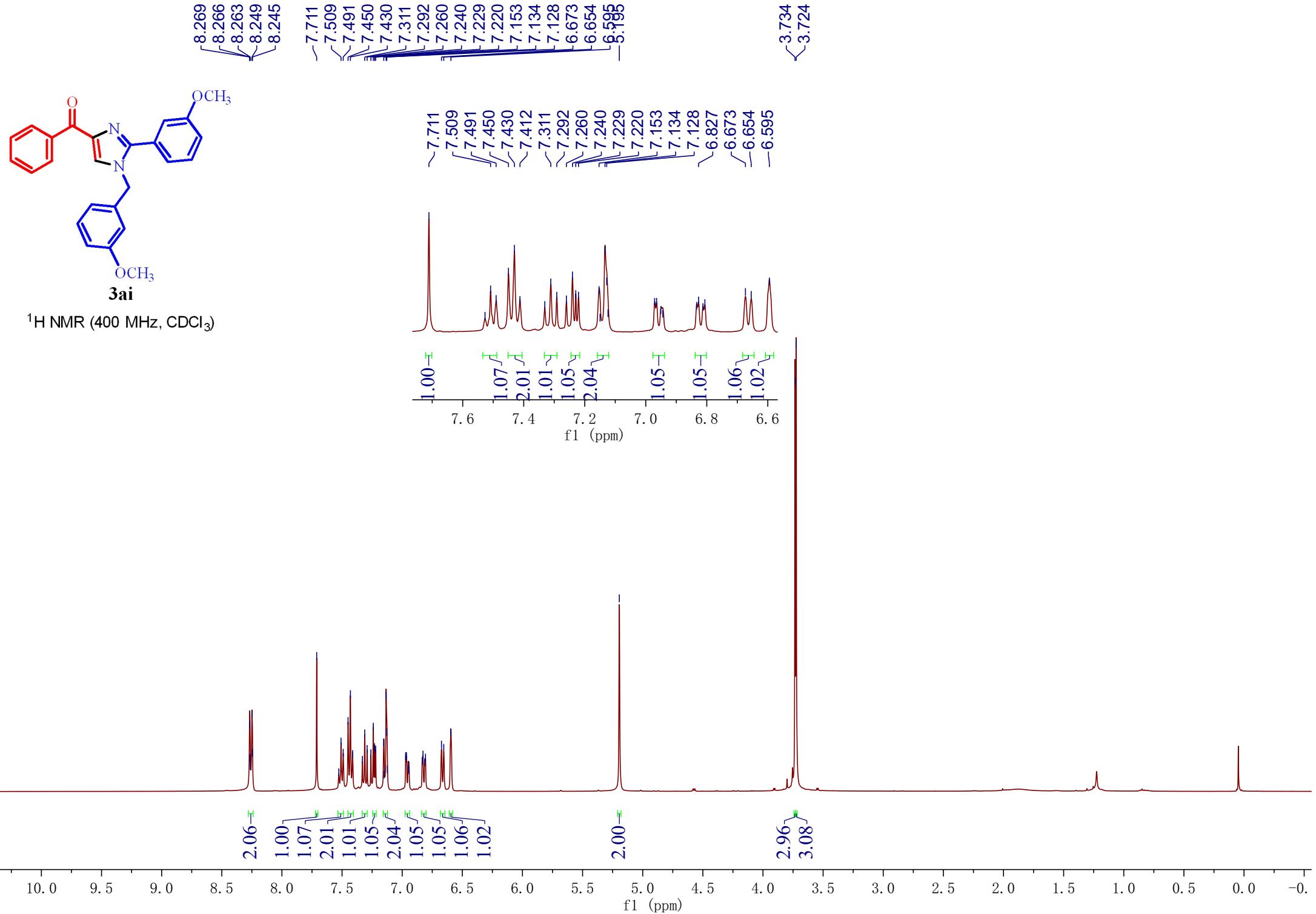


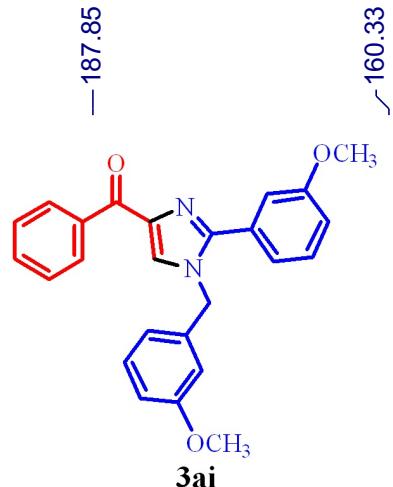




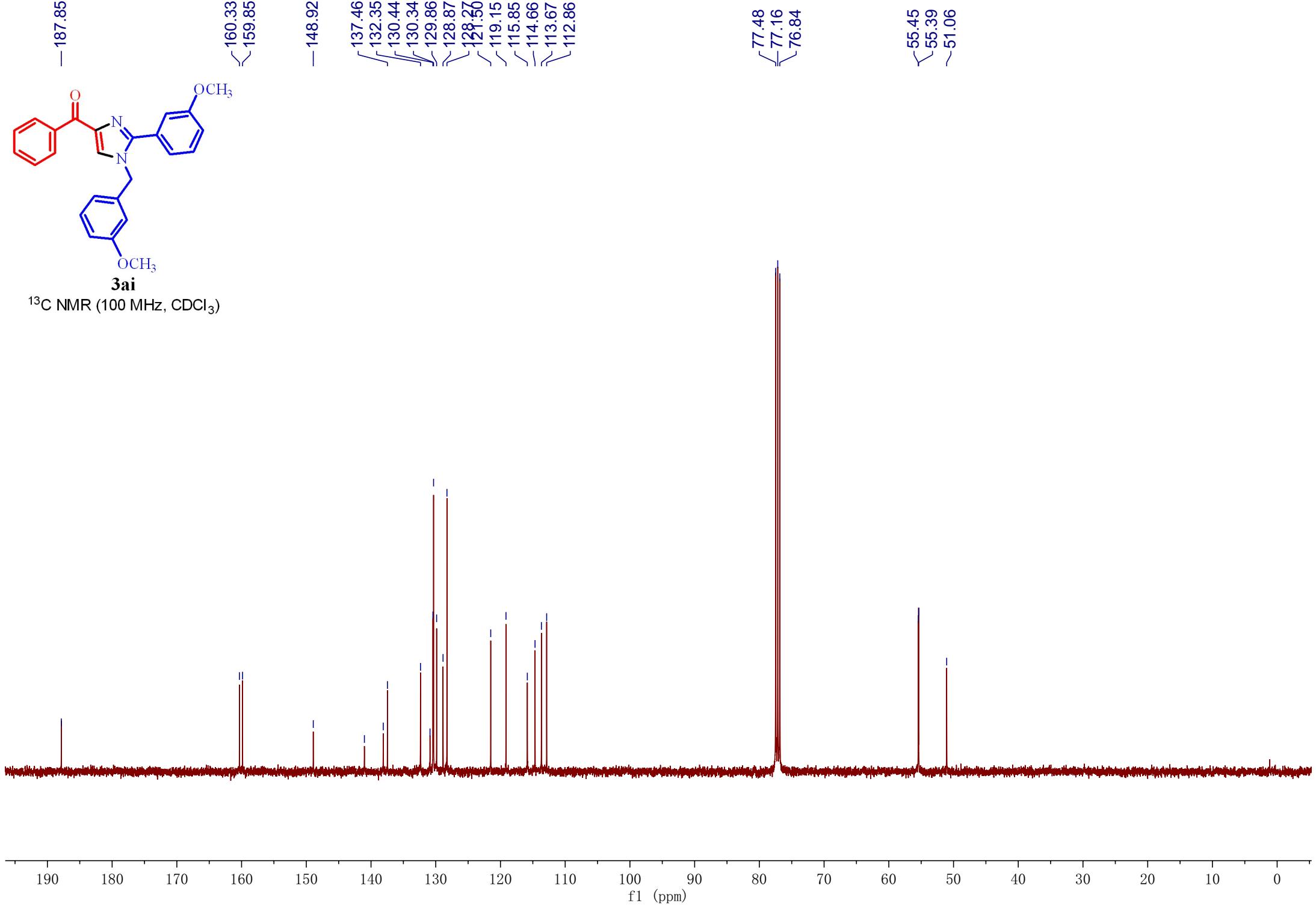


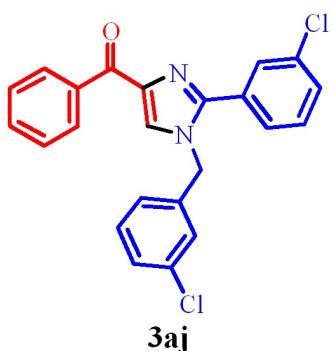




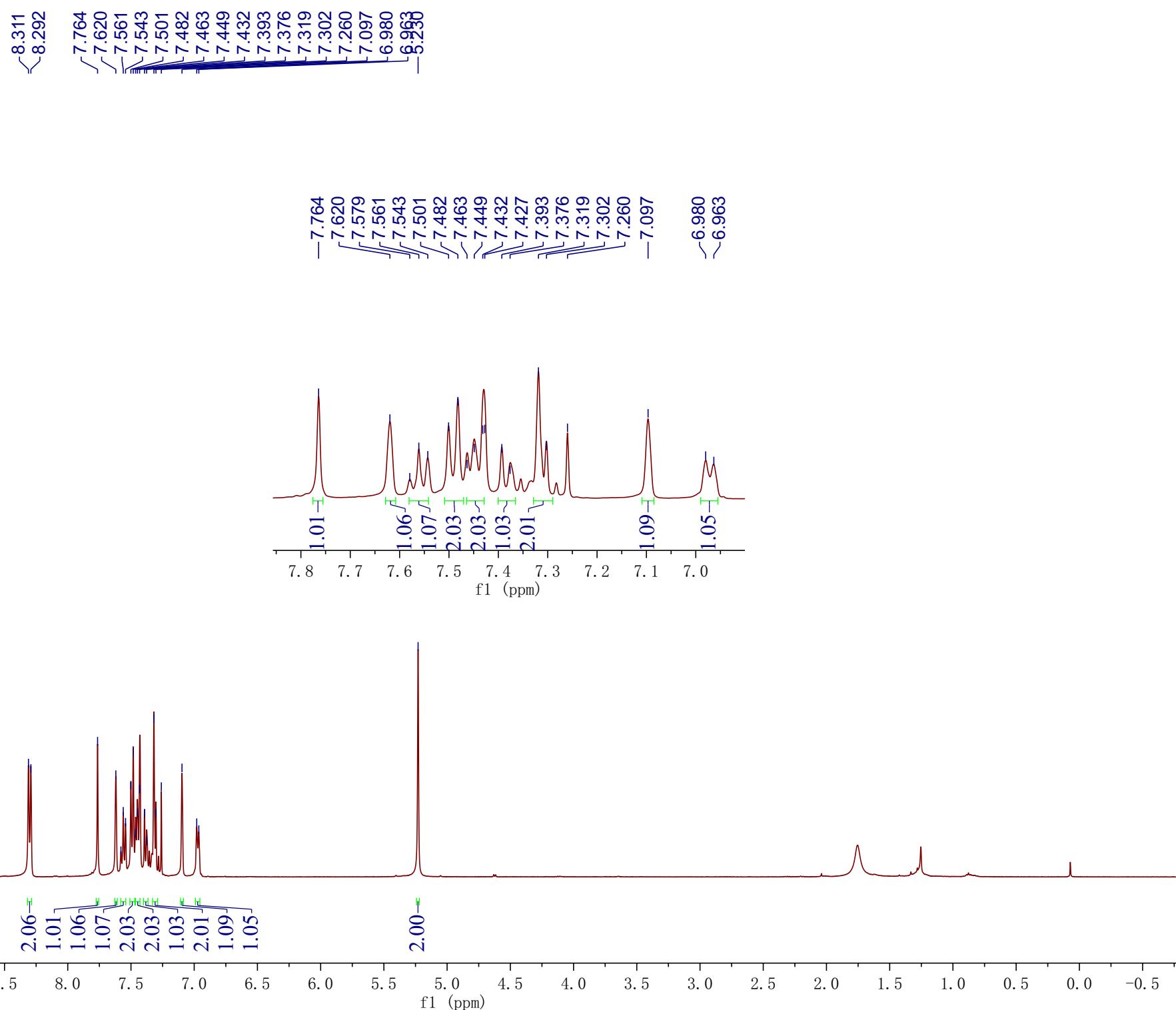


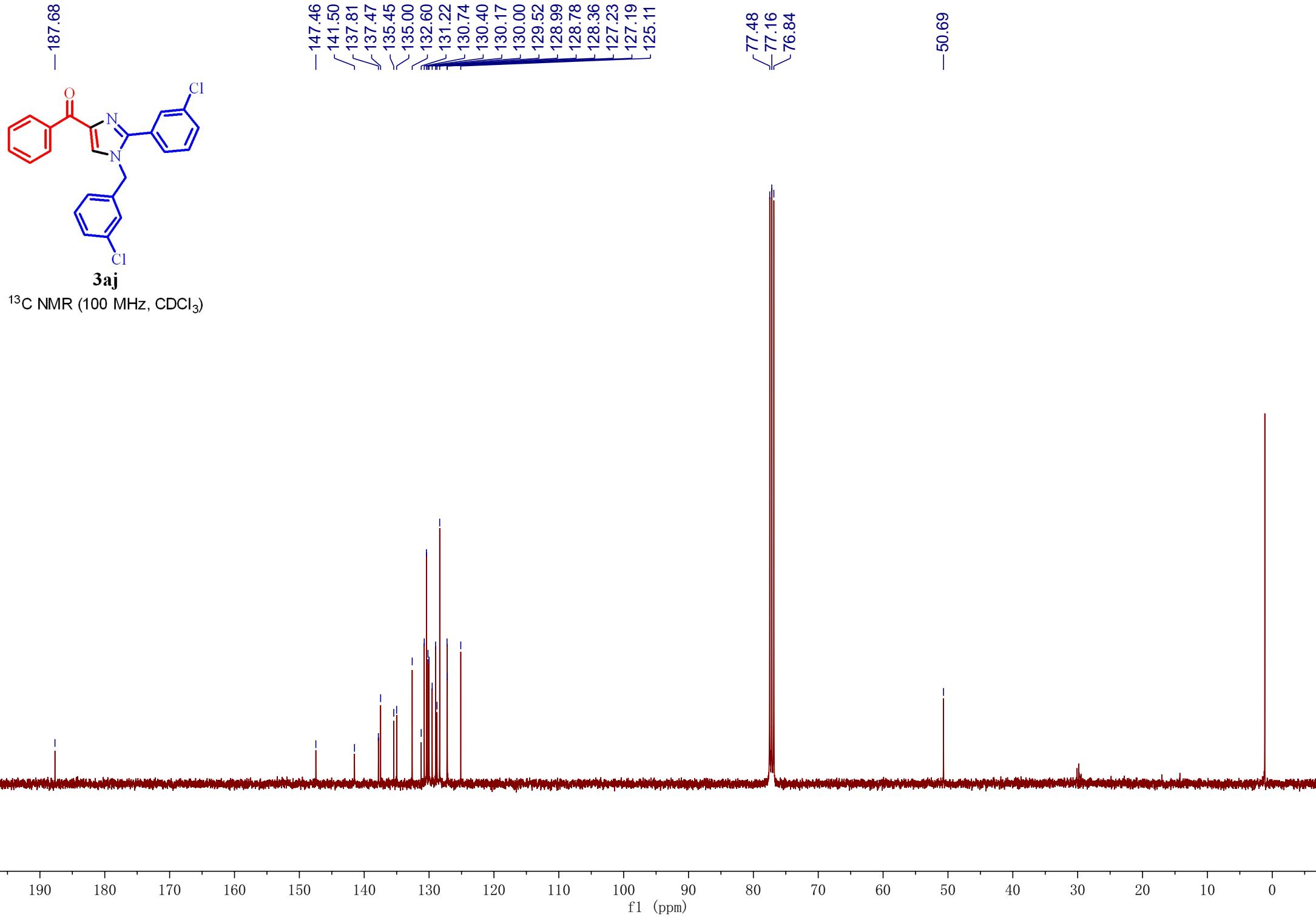
$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )

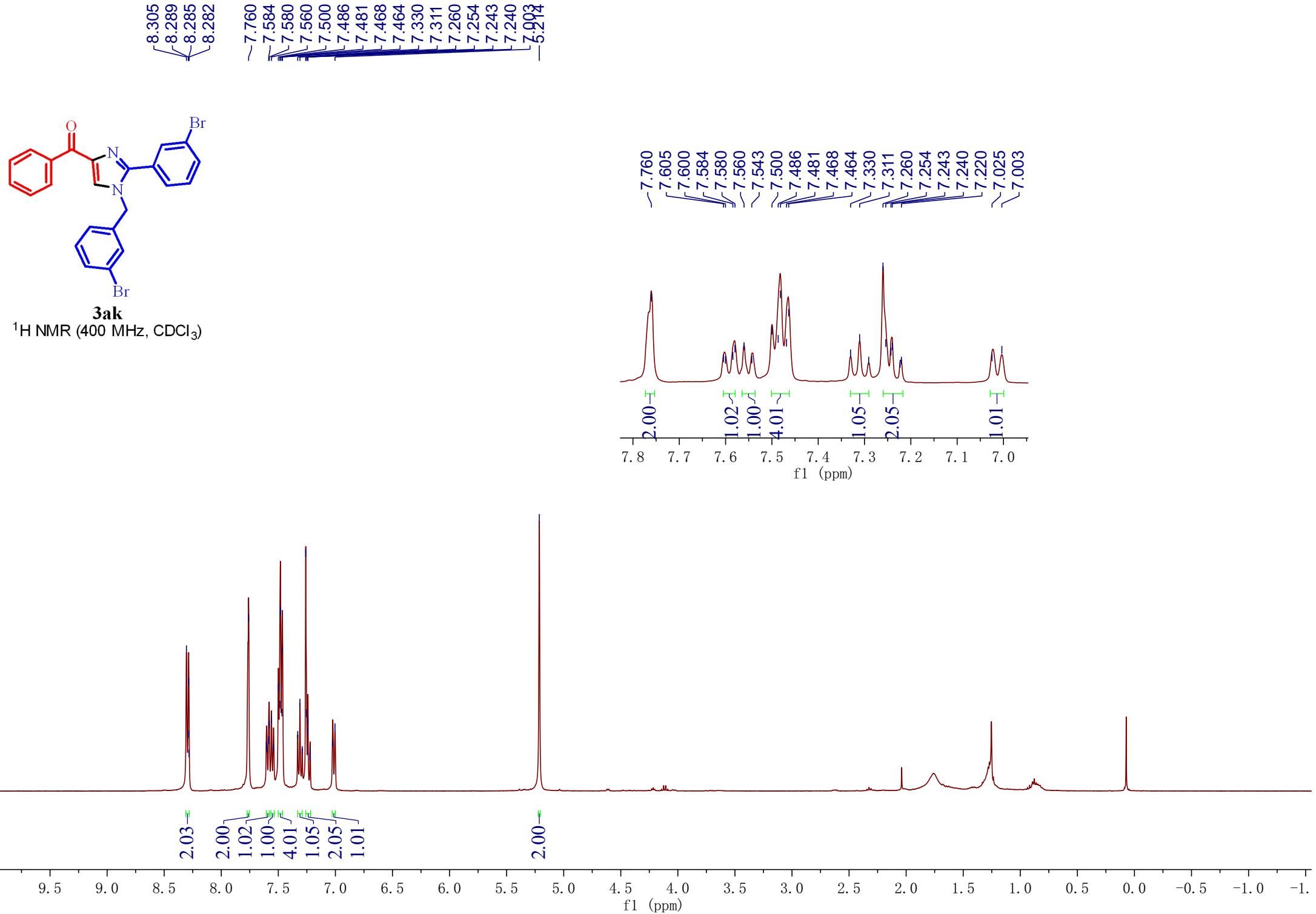




$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )





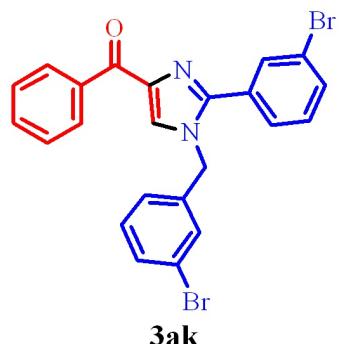


-187.68

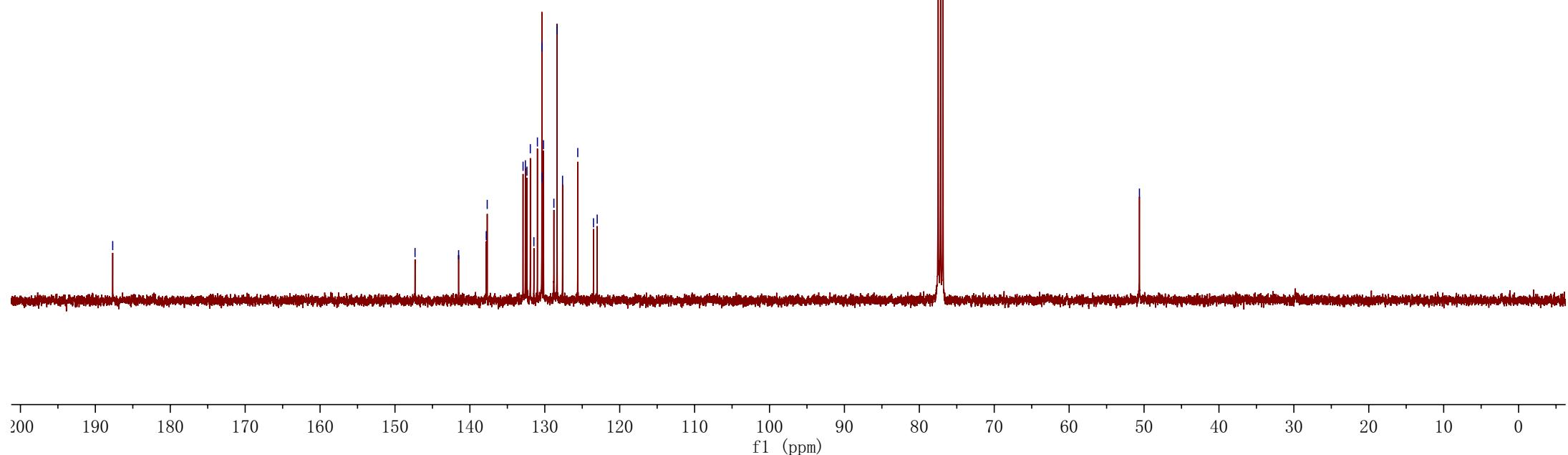
147.32  
141.51  
137.81  
137.69  
132.90  
132.59  
132.38  
131.93  
131.46  
130.99  
130.39  
130.36  
130.17  
128.79  
128.36  
127.63  
125.60  
123.50  
123.00

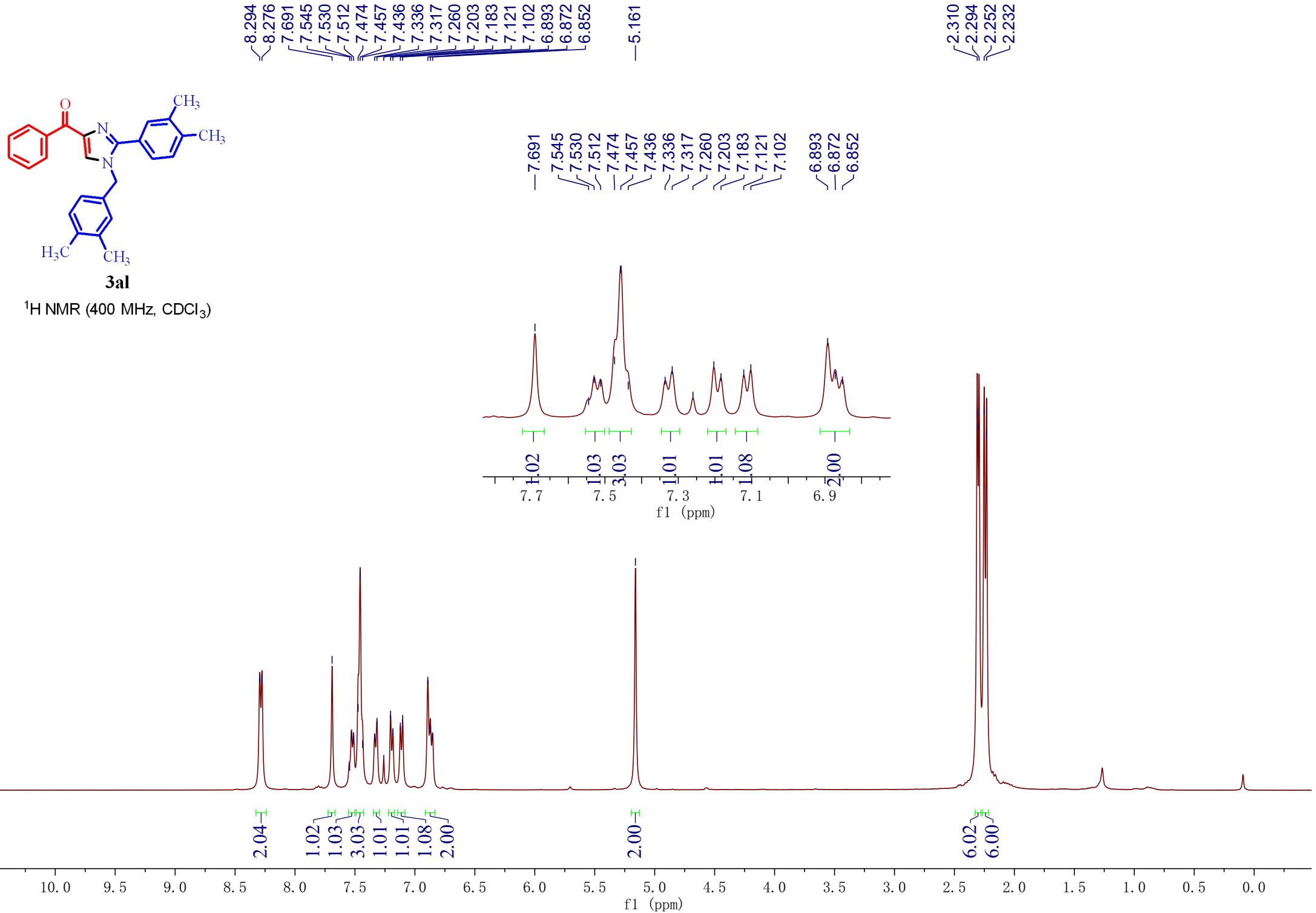
77.48  
77.16  
76.84

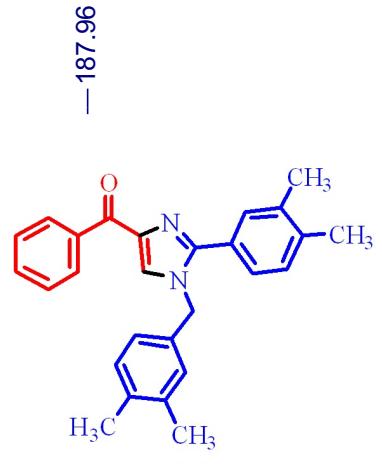
-50.62



<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)

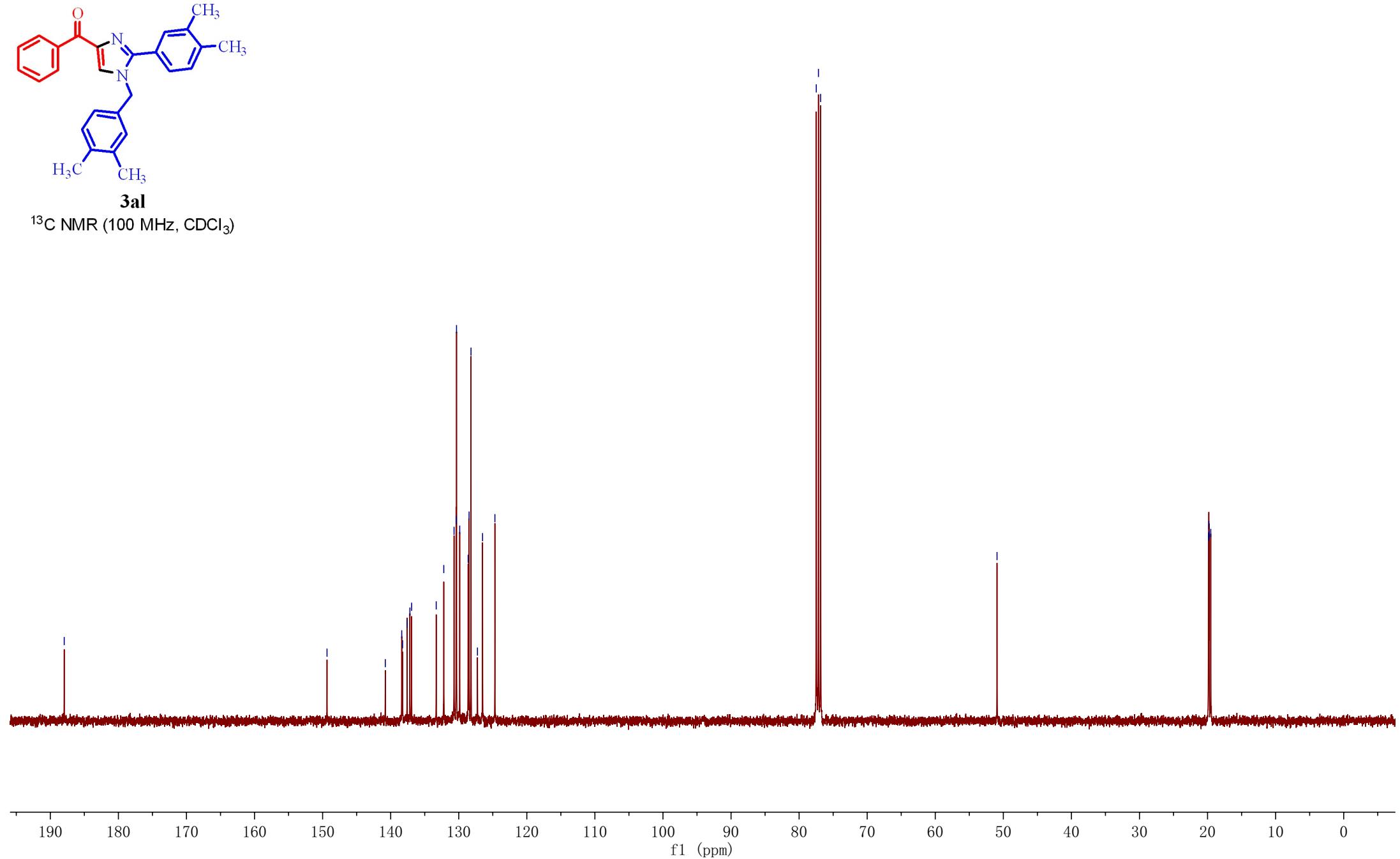






**3al**

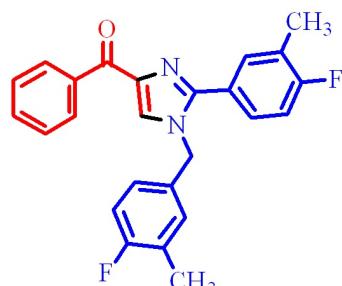
$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )



8.273  
8.270  
8.266  
8.258  
8.253  
8.249

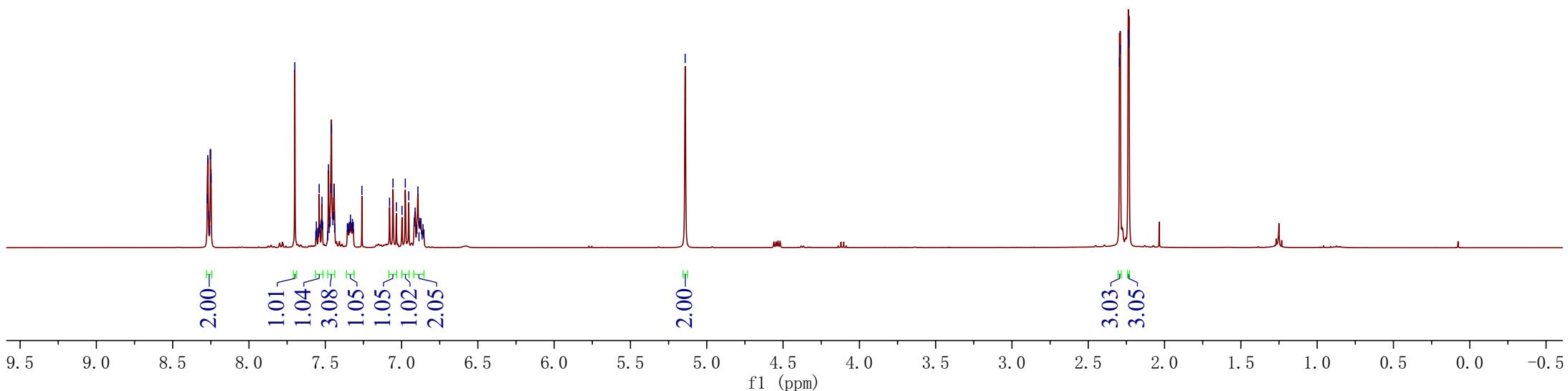
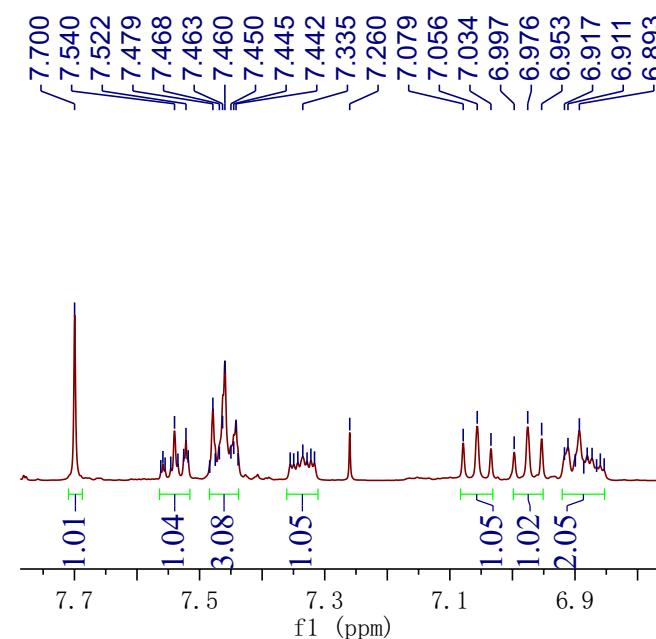
7.700  
7.540  
7.522  
7.479  
7.463  
7.460  
7.450  
7.445  
7.442  
7.335  
7.260  
7.079  
7.056  
7.034  
6.997  
6.976  
6.953  
6.911  
6.893

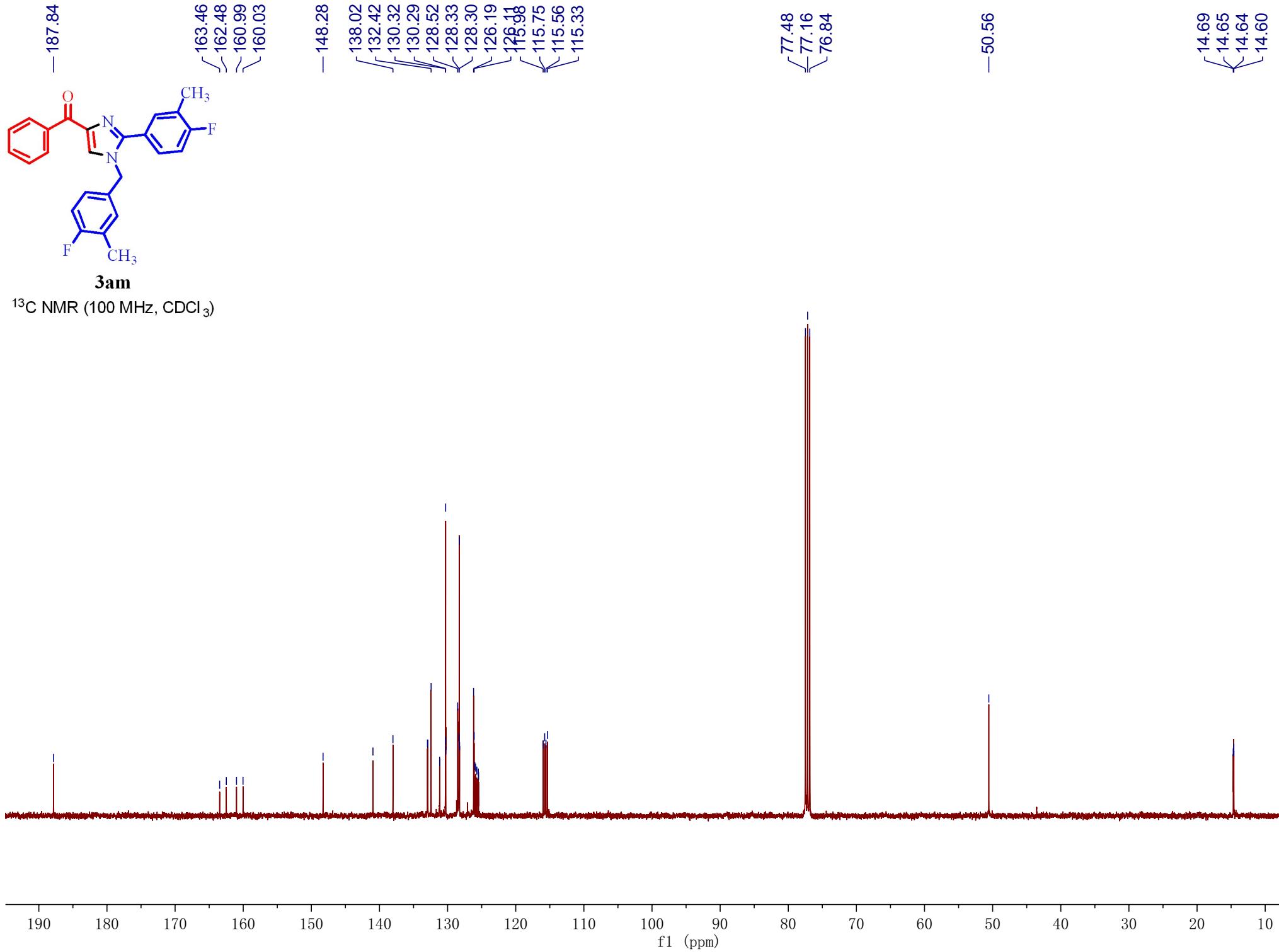
2.295  
2.290  
2.238  
2.233

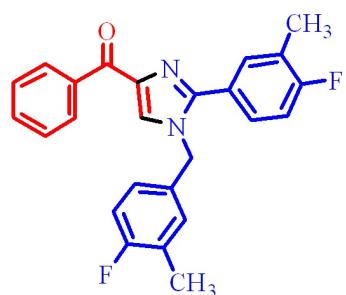


**3am**

$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )







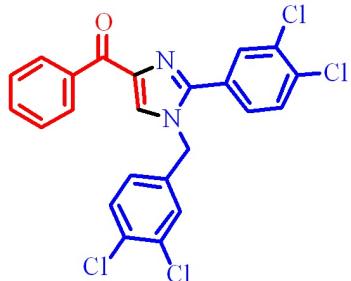
**3am**

$^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ )

~ -115.00  
~ -117.36

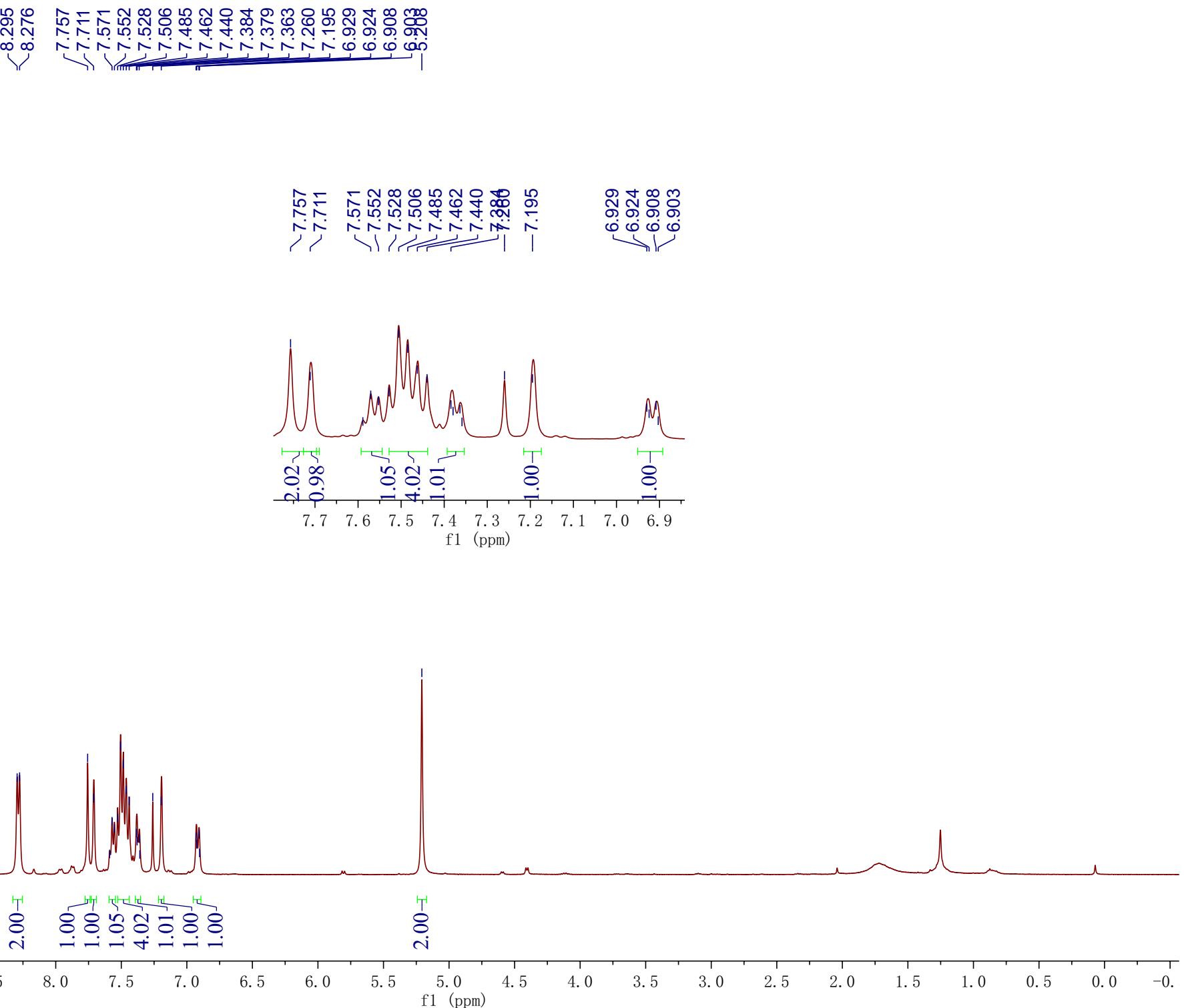
10 0 -10 -20 -30 -40 -50 -60 -70 -80 -90 -100 -110 -120 -130 -140 -150 -160 -170 -180 -190 -200 -210

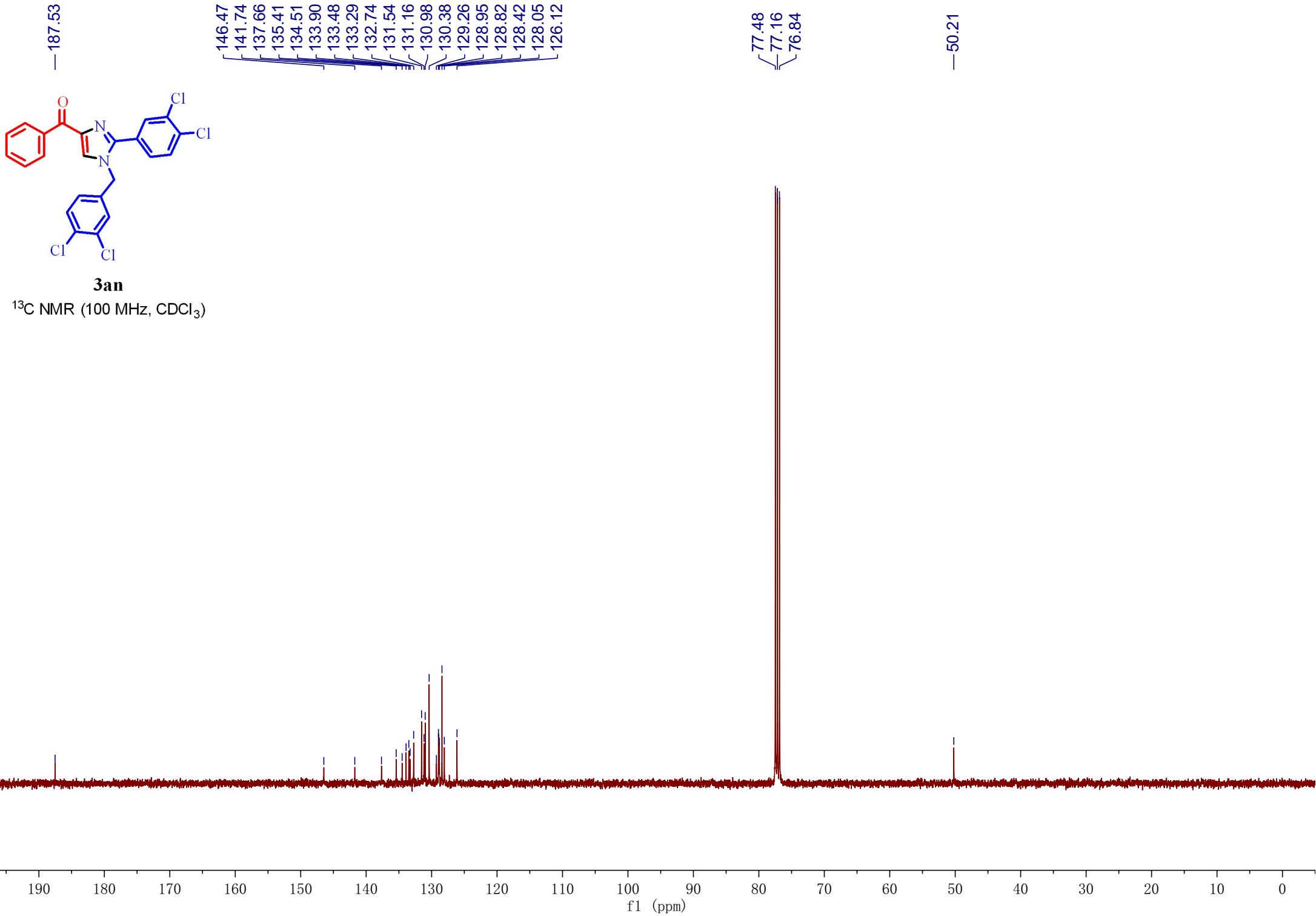
f1 (ppm)

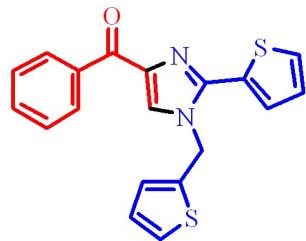


3an

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

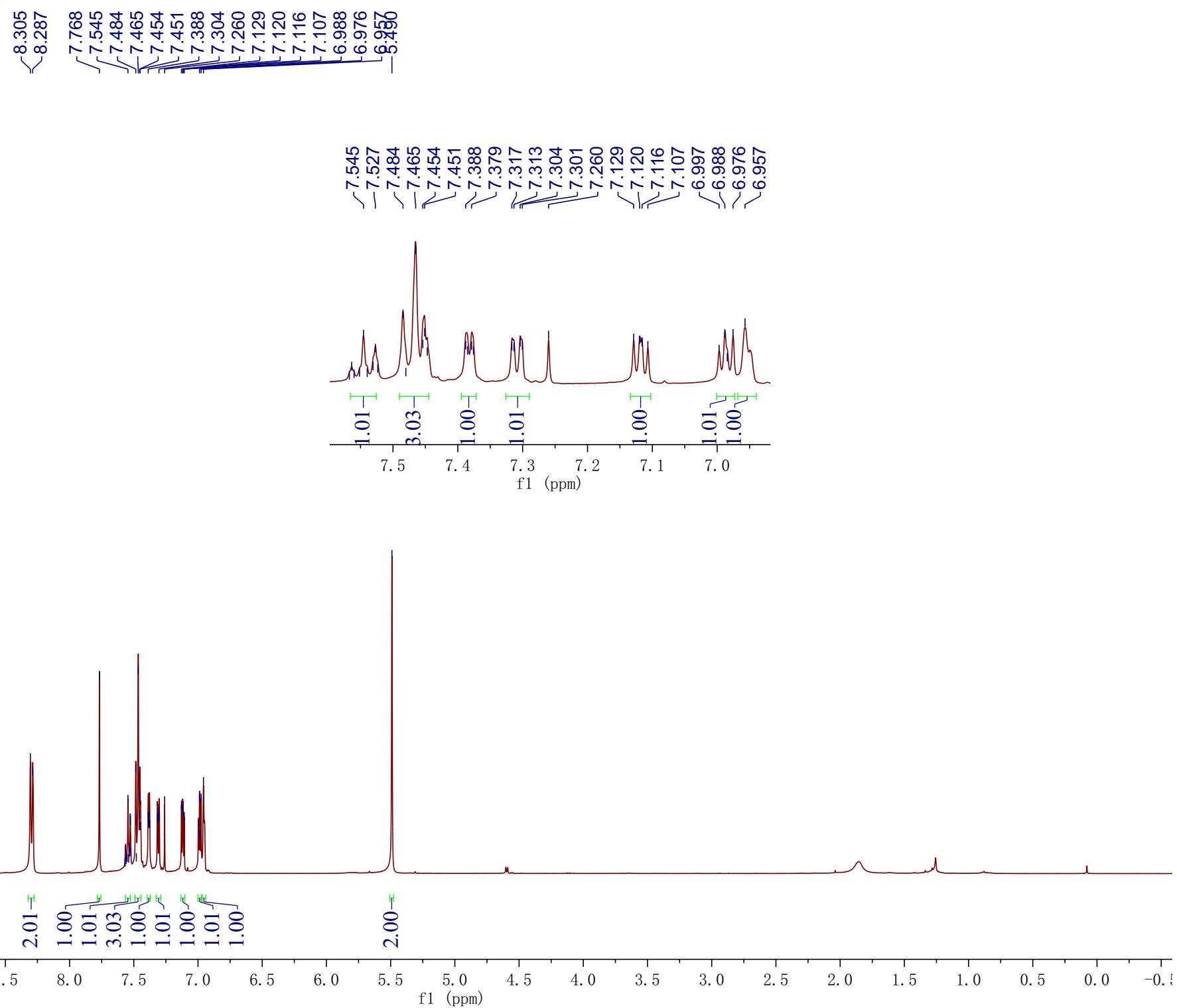




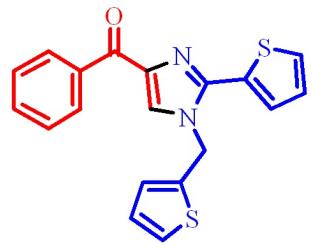


3ao

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)



-187.49



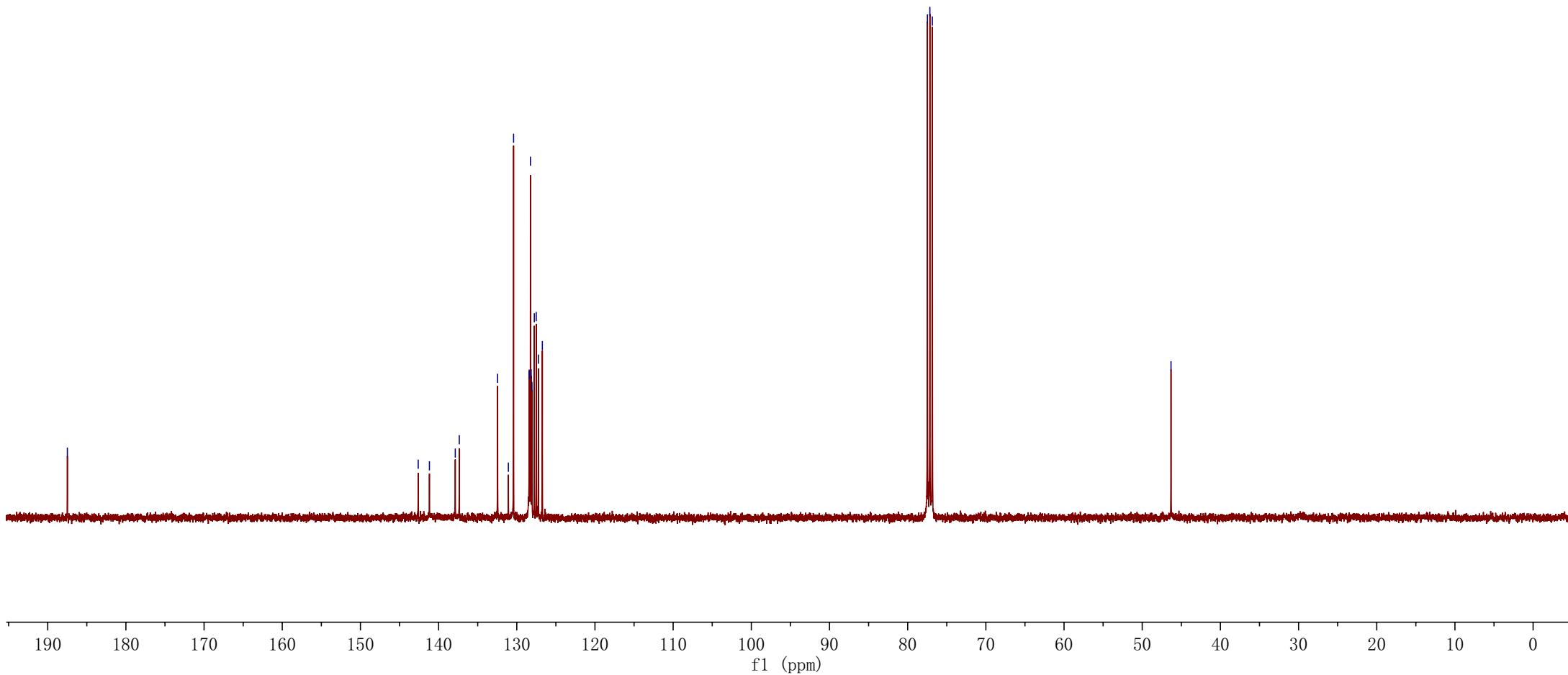
**3ao**

$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )

142.61  
141.18  
137.87  
137.36  
132.46  
132.46  
131.08  
130.41  
128.42  
128.26  
128.16  
128.06  
127.76  
127.51  
127.24  
126.73

77.48  
77.16  
76.84

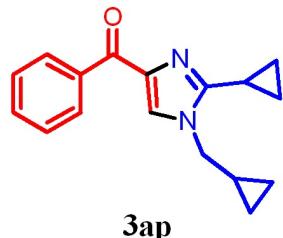
-46.32



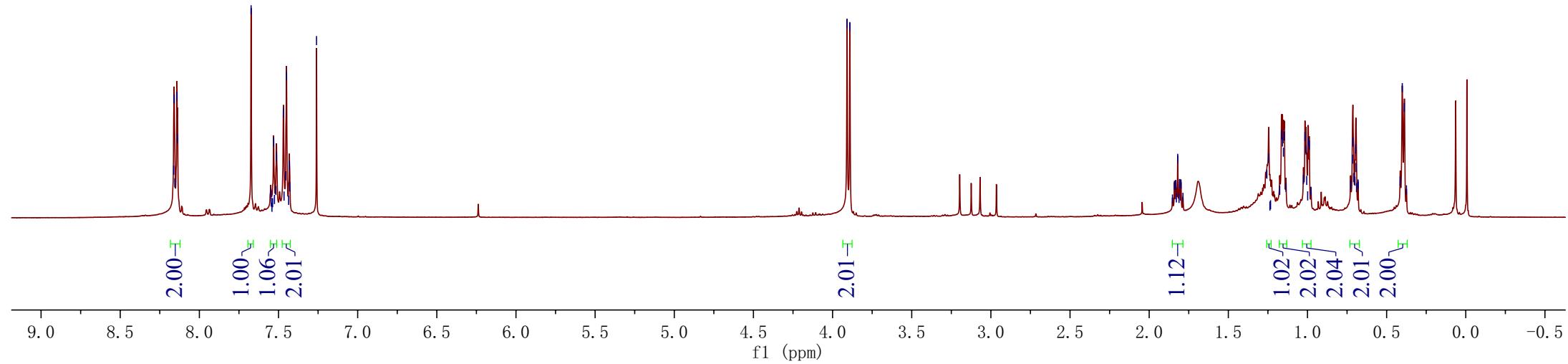
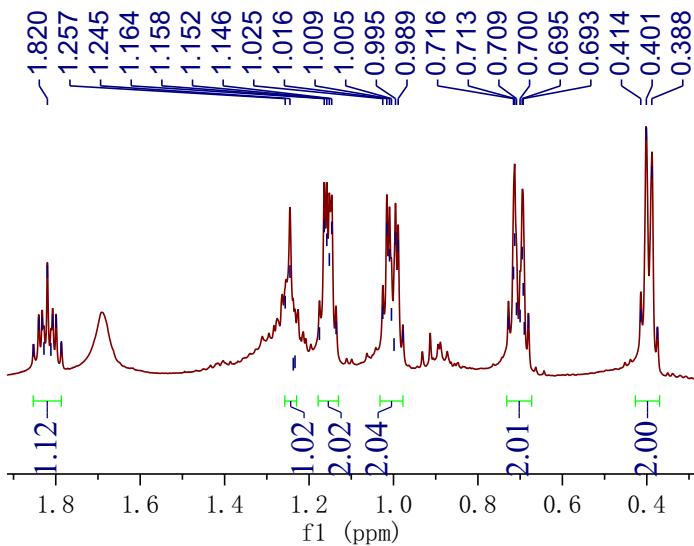
8.162  
8.159  
8.155  
8.142  
8.138  
7.673  
7.550  
7.546  
7.541  
7.537  
7.526  
7.516  
7.513  
7.469  
7.531  
7.526  
7.465  
7.436  
7.432  
7.429  
7.260

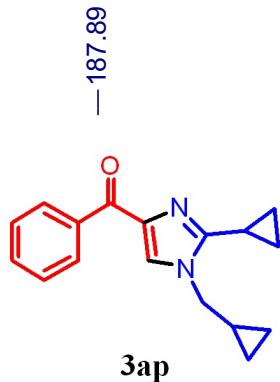
3.909  
3.891

1.853  
1.840  
1.832  
1.828  
1.820  
1.812  
1.807  
1.799  
1.787  
1.245  
1.164  
1.158  
1.152  
1.146  
1.025  
1.016  
1.009  
1.005  
0.995  
0.989  
0.716  
0.713  
0.709  
0.700  
0.695  
0.693  
0.414  
0.401  
0.388

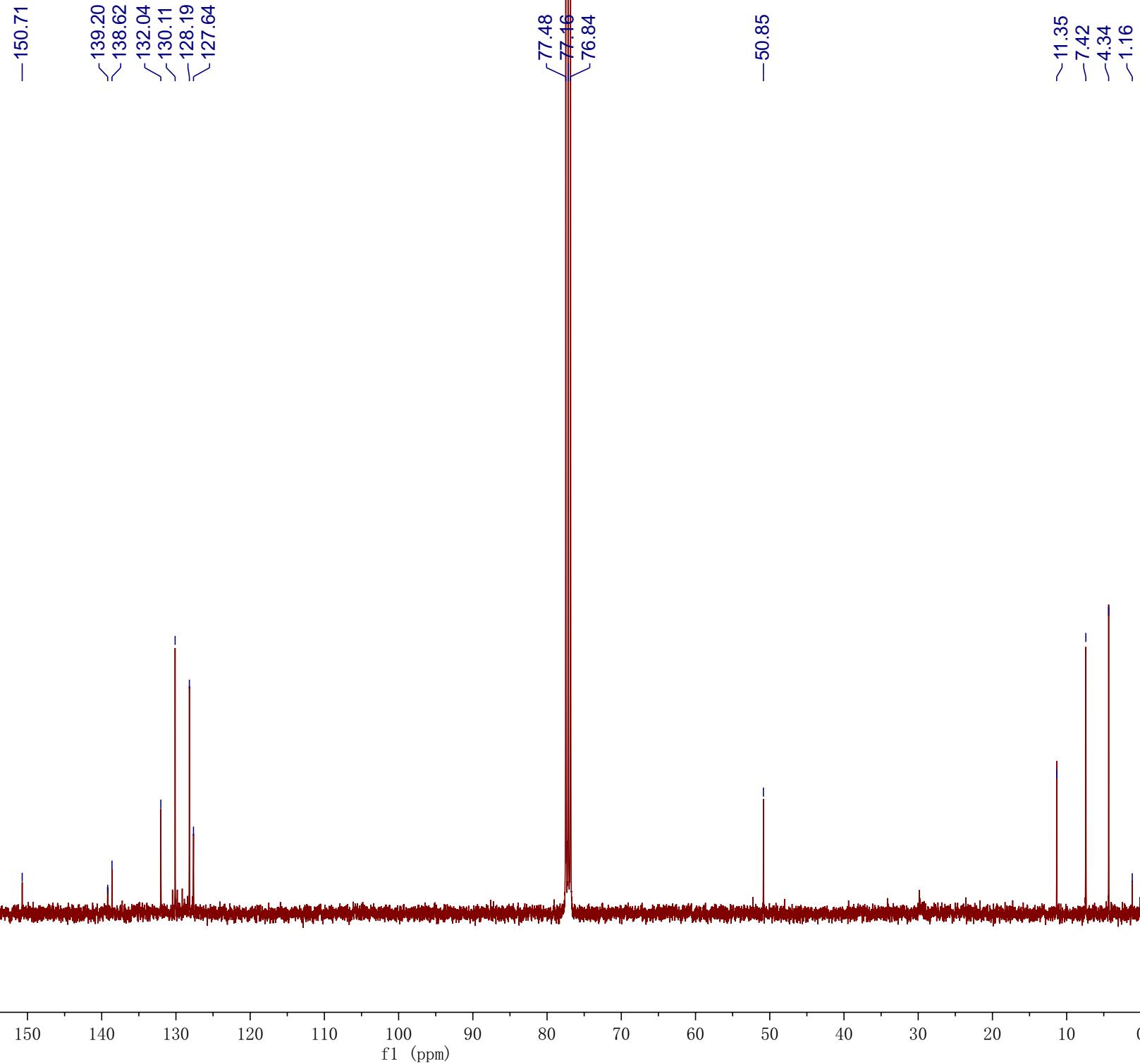


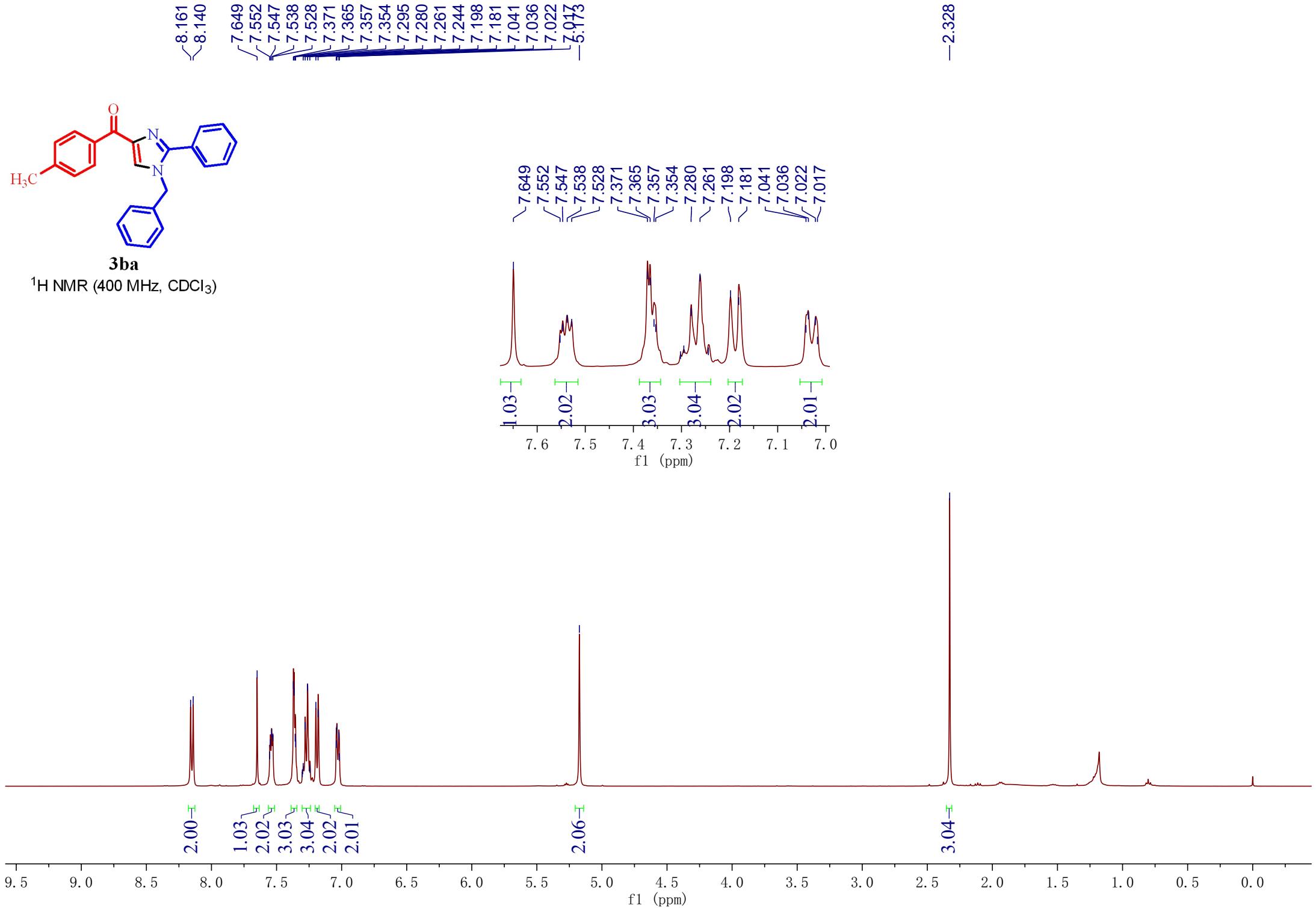
$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )

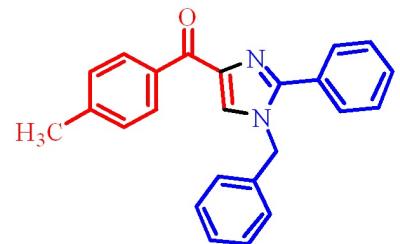




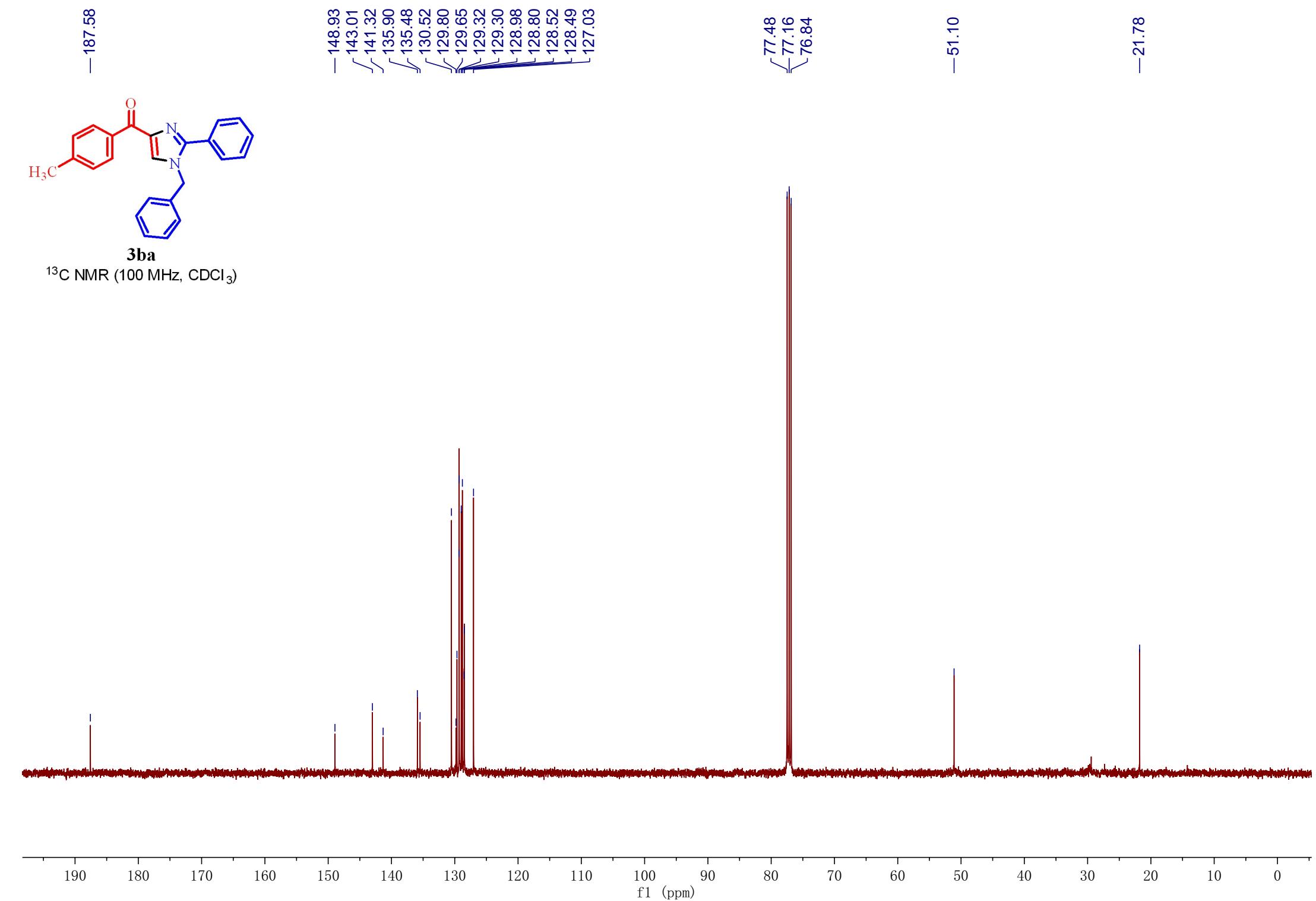
$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )







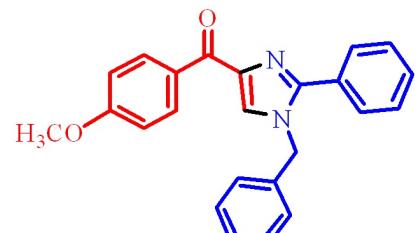
<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)



8.457  
8.451  
8.440  
8.434  
8.427

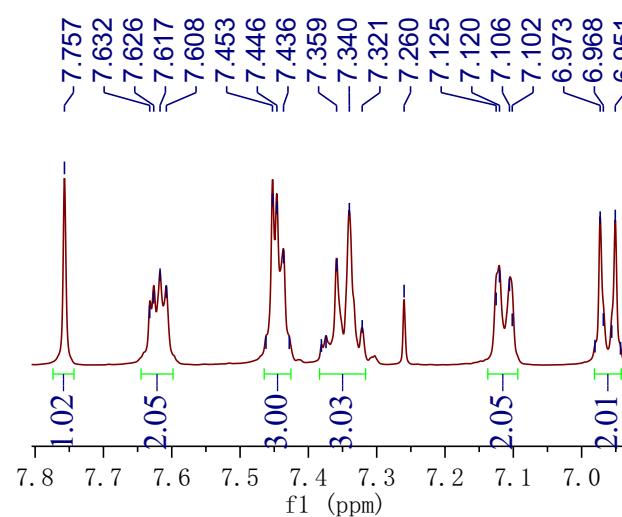
7.757  
7.632  
7.626  
7.617  
7.608  
7.453  
7.446  
7.436  
7.359  
7.340

7.321  
7.260  
7.125  
7.120  
7.106  
7.102  
6.973  
6.968  
6.951



**3ca**

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)



f1 (ppm)

2.00

1.02

2.05

3.00

3.03

2.05

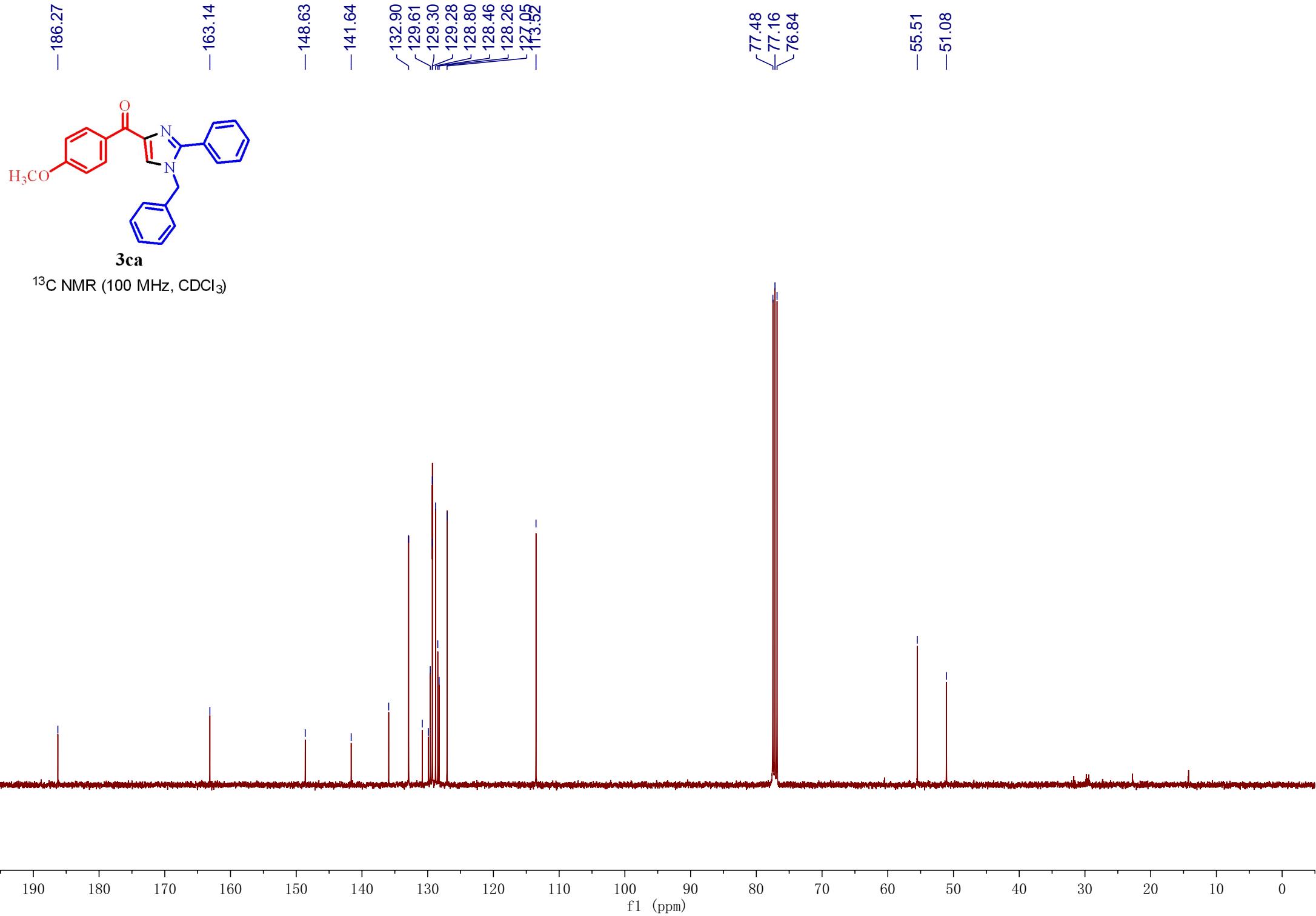
2.01

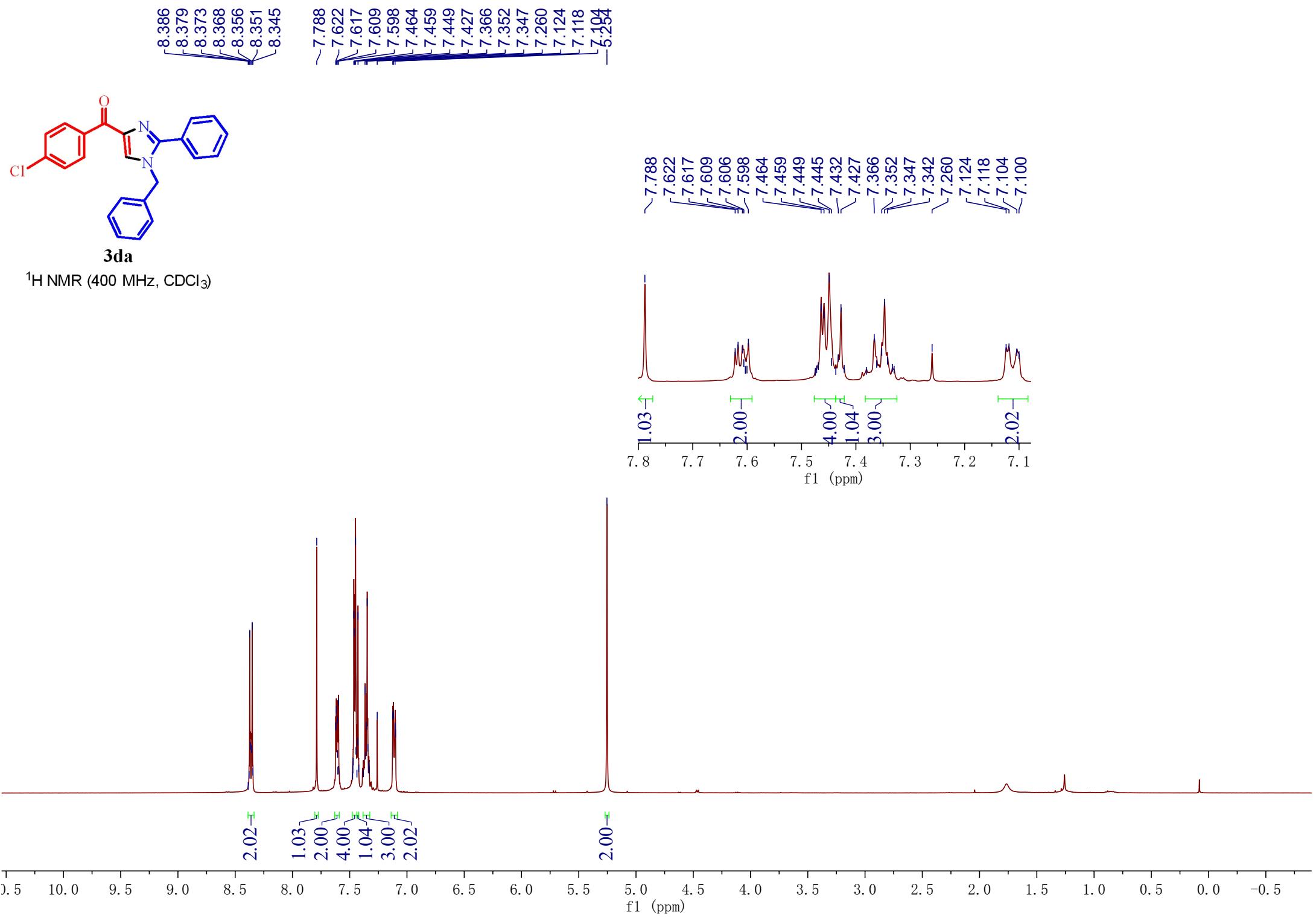
2.02

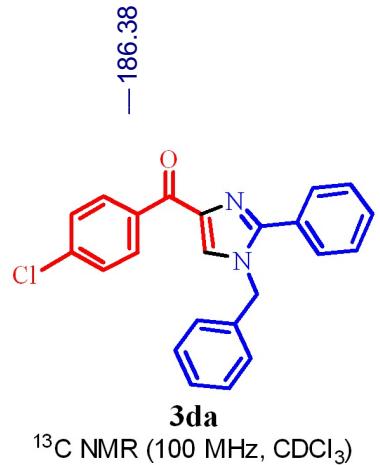
3.02

f1 (ppm)

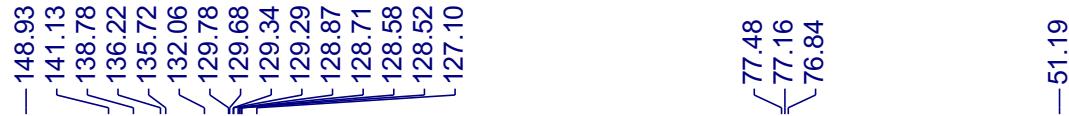
9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0 -0.1

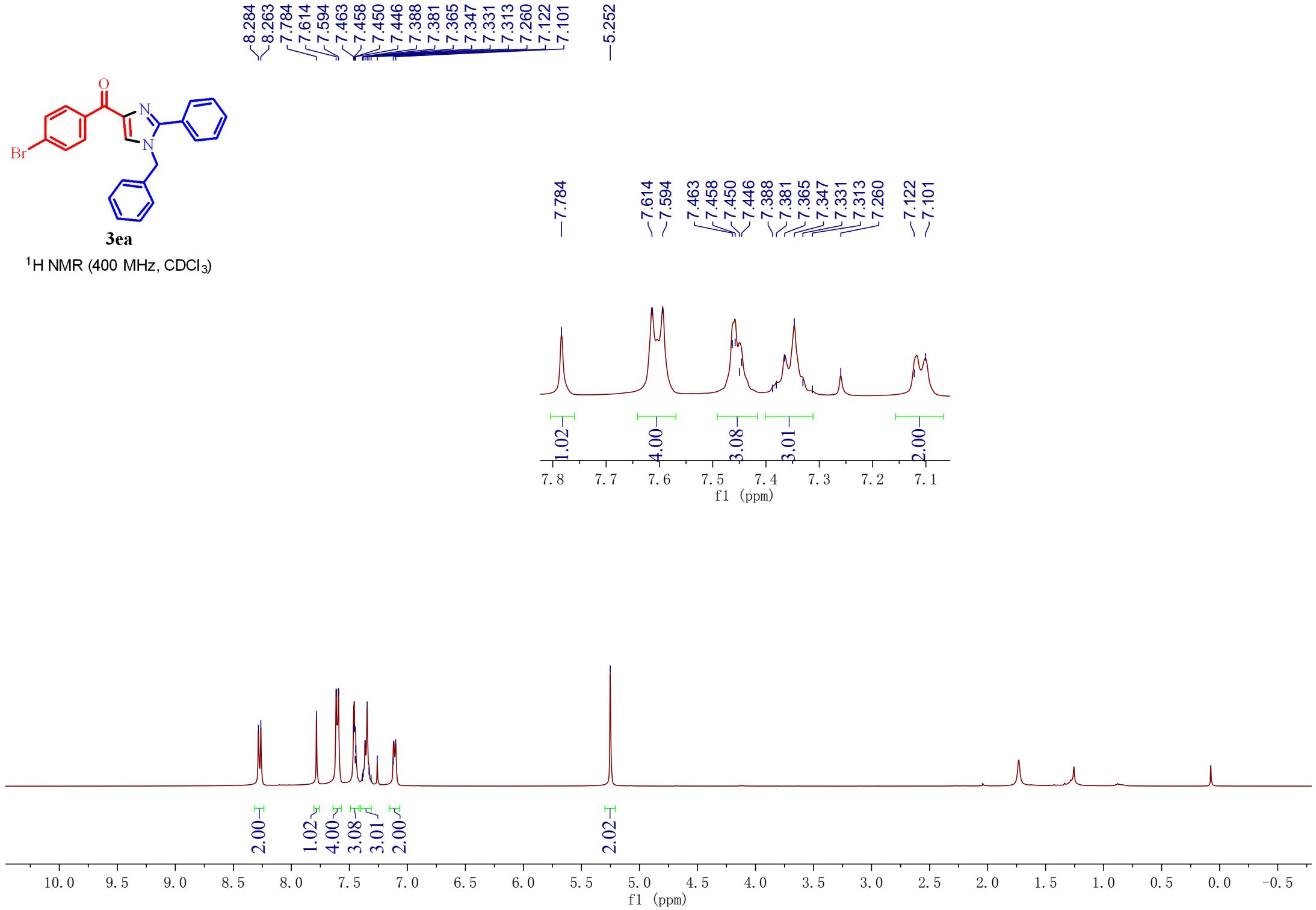






<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)



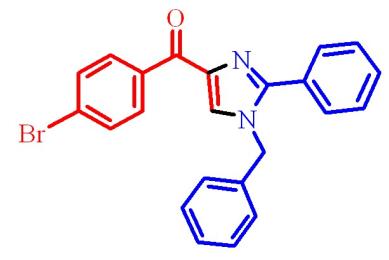


-186.57

-148.97  
-141.09  
-136.66  
-135.72  
-132.19  
-131.52  
-129.80  
-129.68  
-129.35  
-129.30  
-128.89  
-128.74  
-128.60  
-127.54  
-127.12

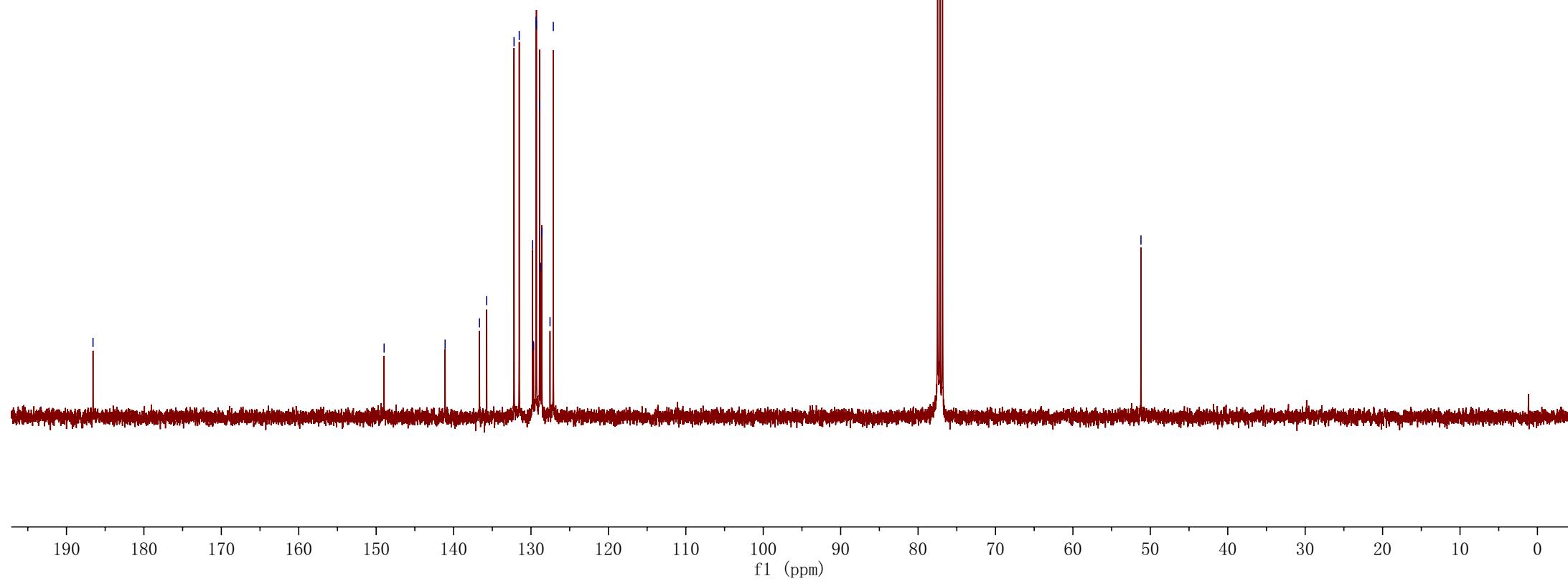
77.48  
77.16  
76.84

-51.21



**3ea**

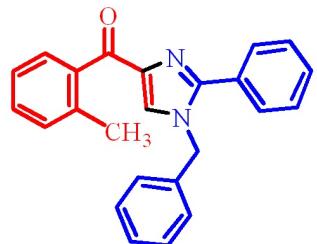
$^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )



7.675  
7.671  
7.662  
7.657  
7.652  
7.644  
7.638  
7.544  
7.491  
7.482  
7.478  
7.464  
7.409  
7.390  
7.375  
7.320  
7.310  
7.298  
7.292  
7.278  
7.260  
7.140  
7.135  
7.121  
7.117

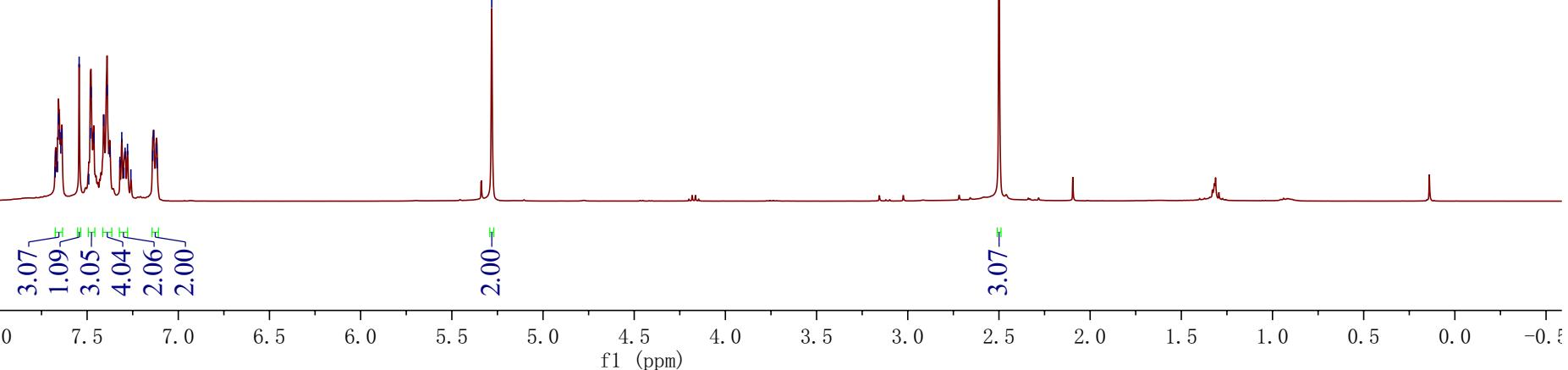
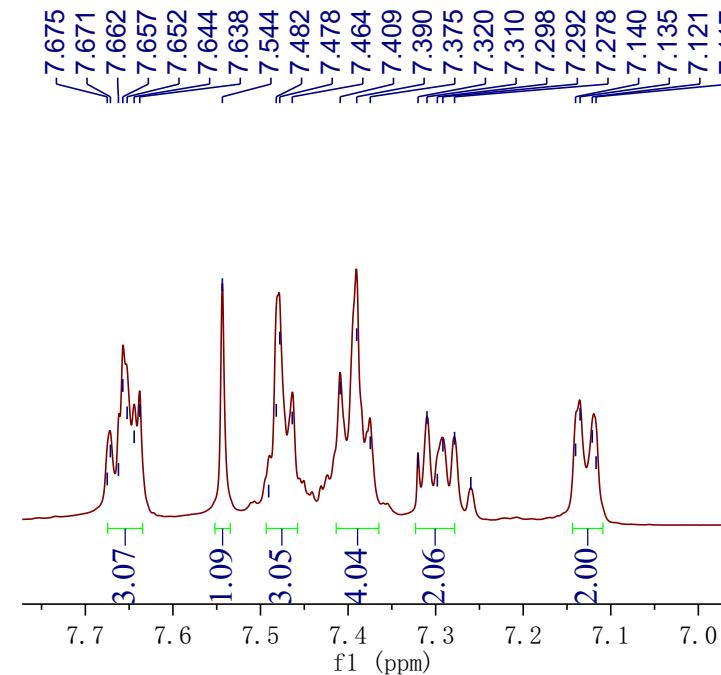
— 5.281

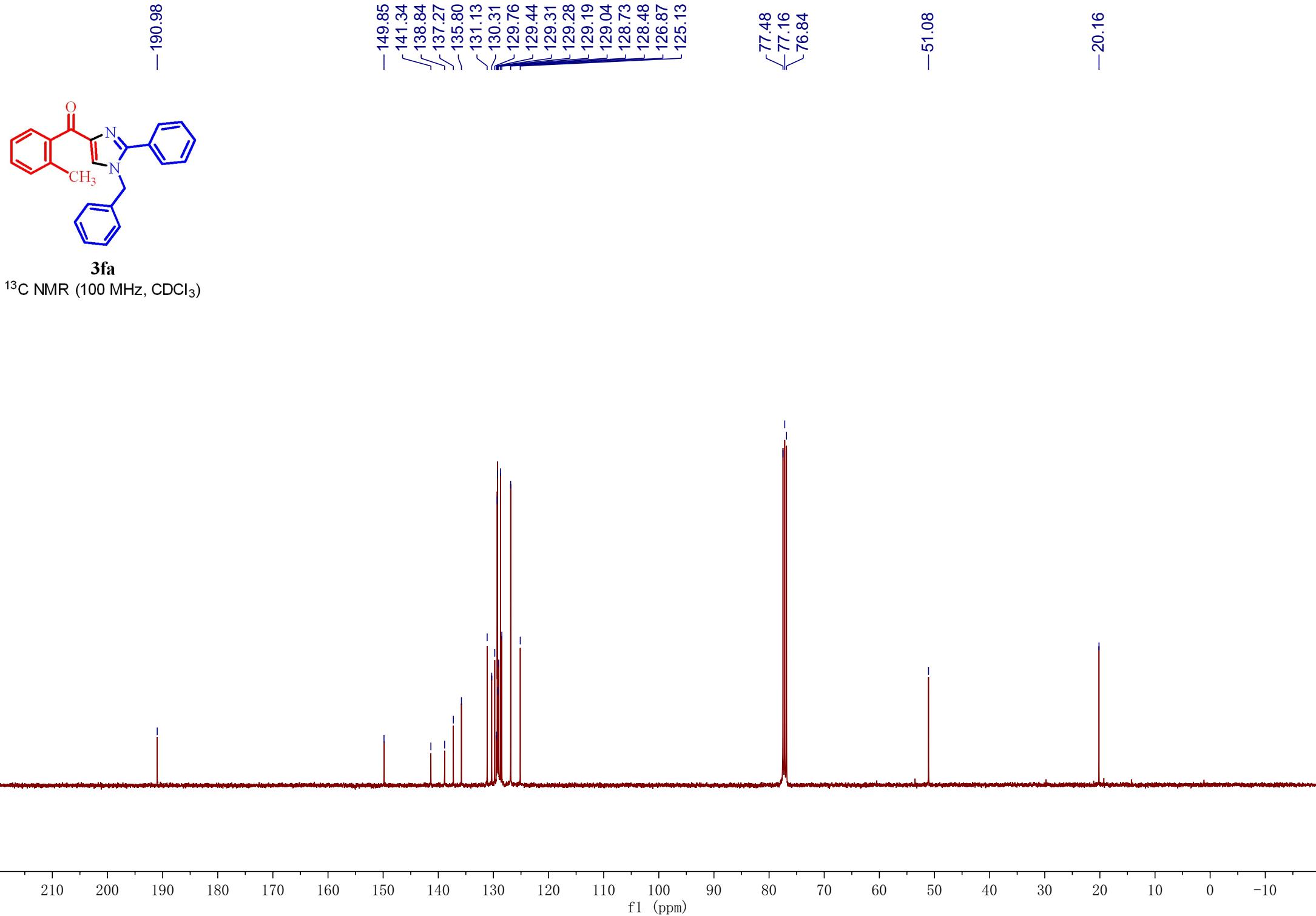
— 2.500

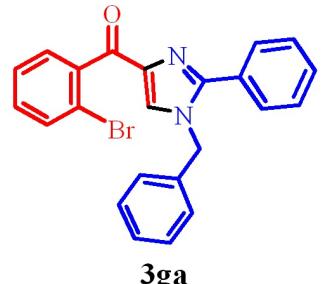


**3fa**

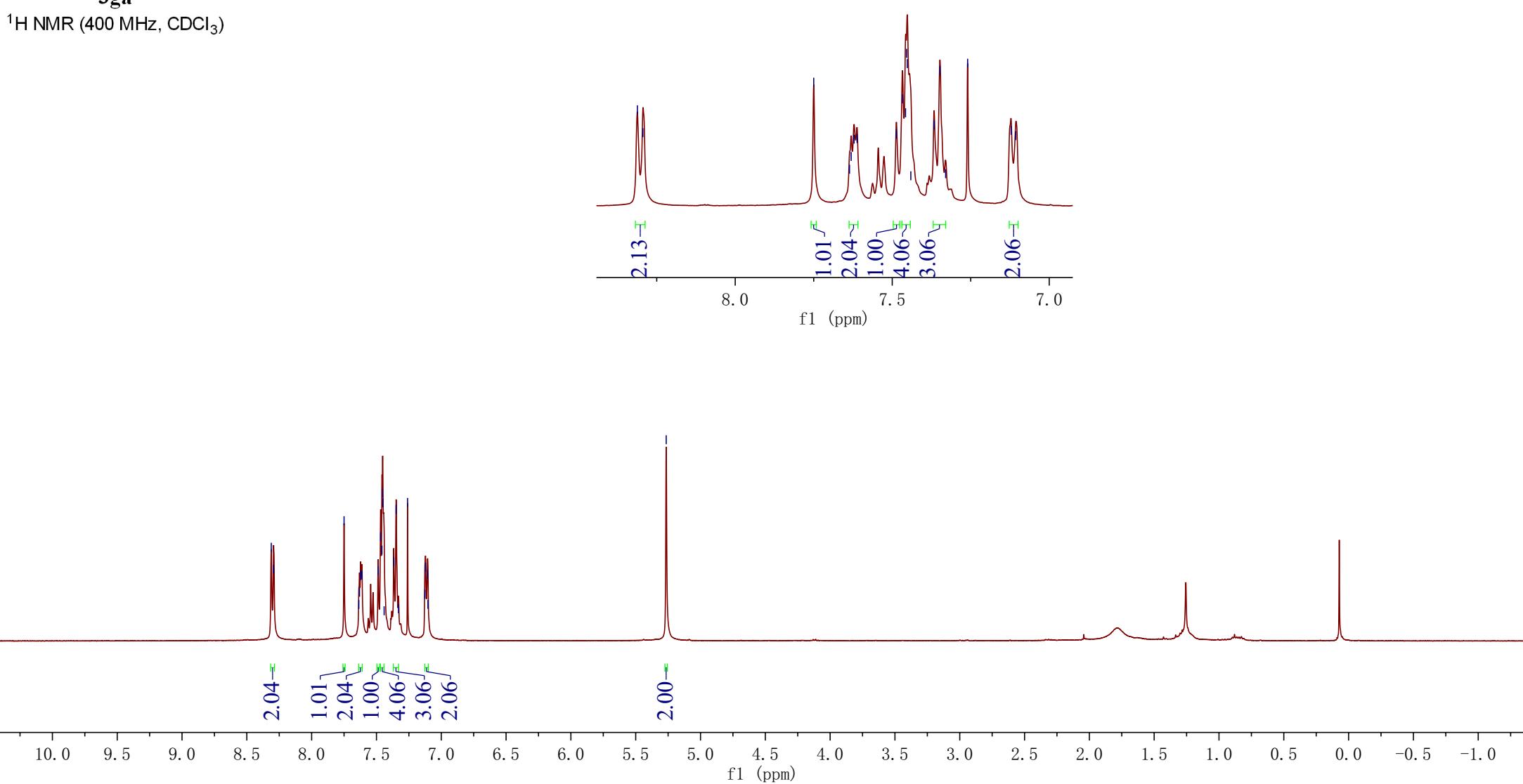
$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )

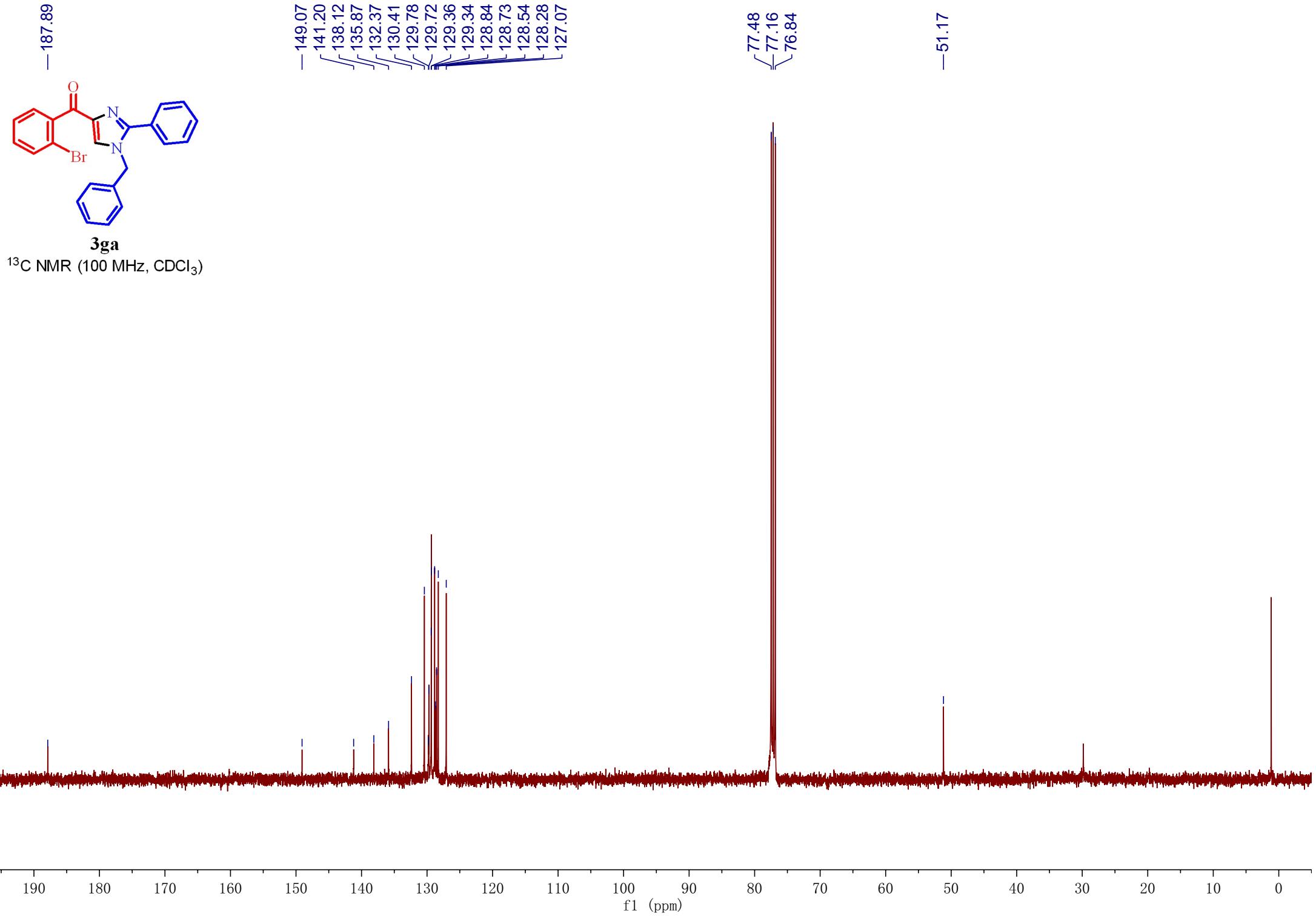


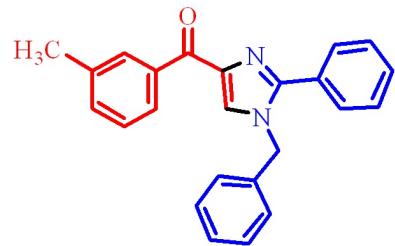
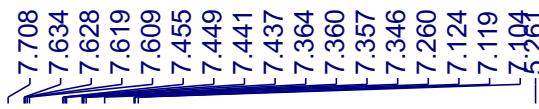
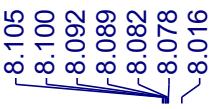




<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

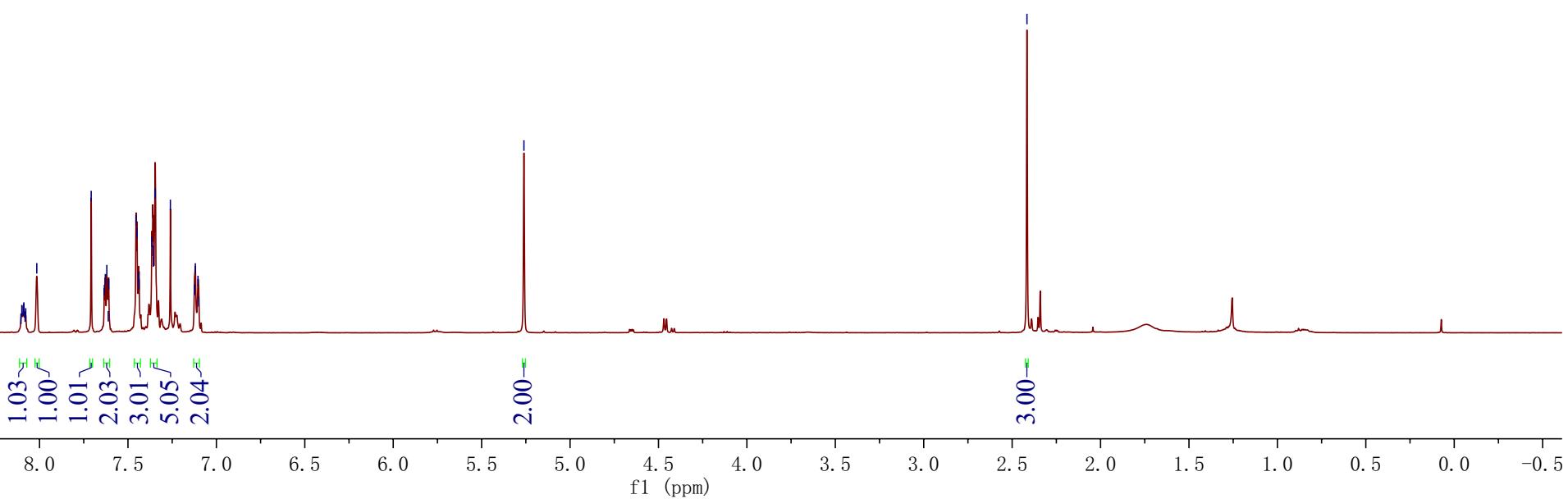
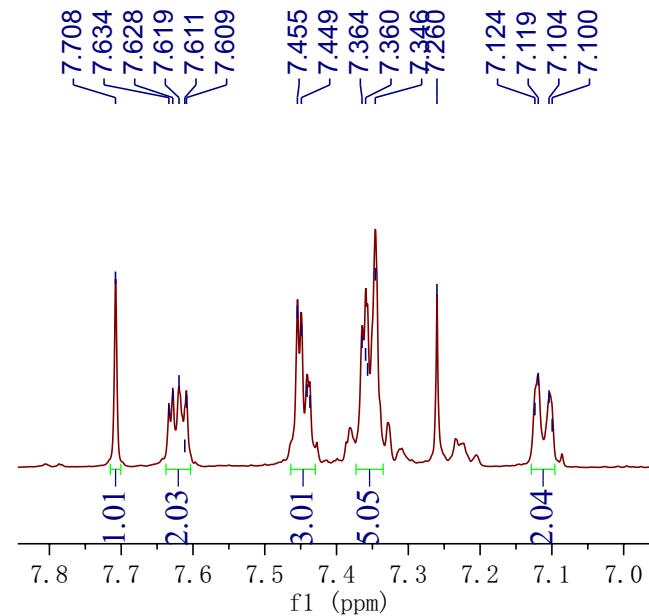


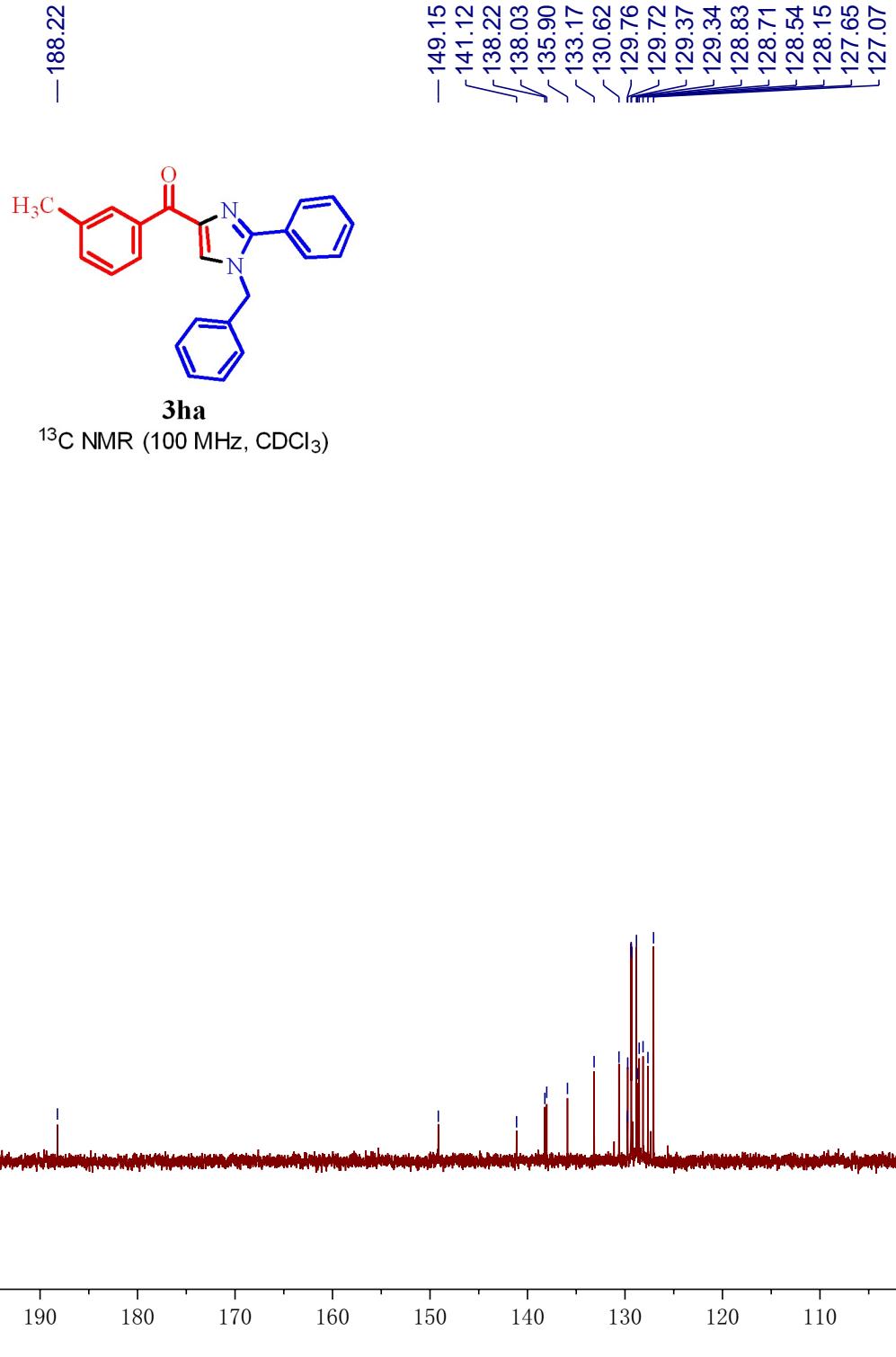




3ha

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)





8.311  
8.277  
8.257

7.777

7.631

7.626

7.618

7.607

7.469

7.464

7.407

7.456

7.452

7.427

7.354

7.388

7.373

7.359

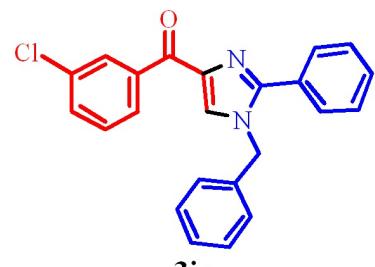
7.260

7.127

7.122

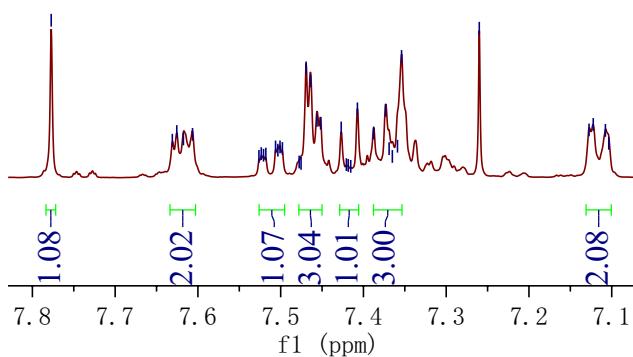
7.108

7.104



<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)

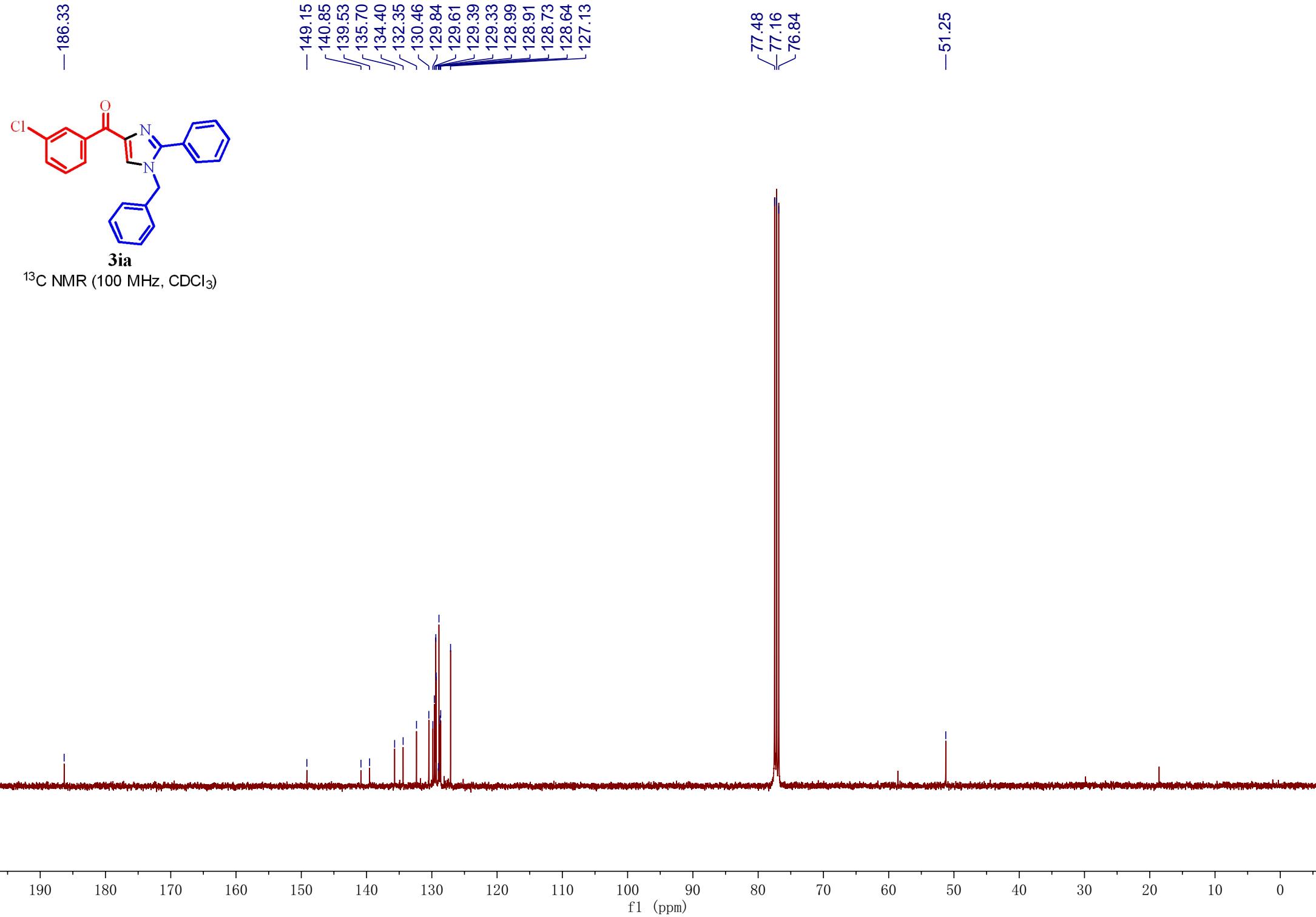
7.777  
7.631  
7.626  
7.618  
7.607  
7.469  
7.464  
7.456  
7.427  
7.388  
7.373  
7.359  
7.260  
7.127  
7.122  
7.108  
7.104

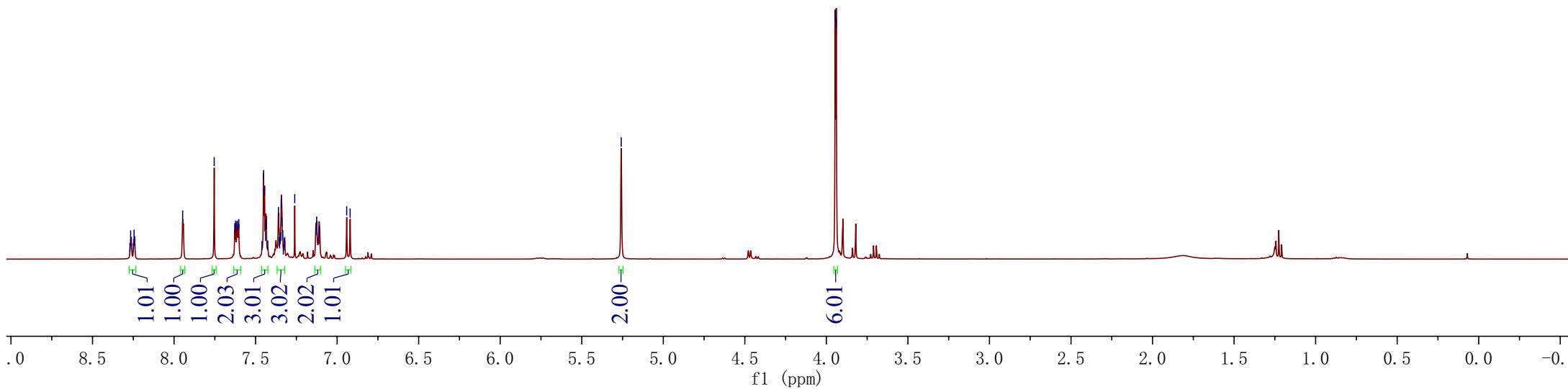
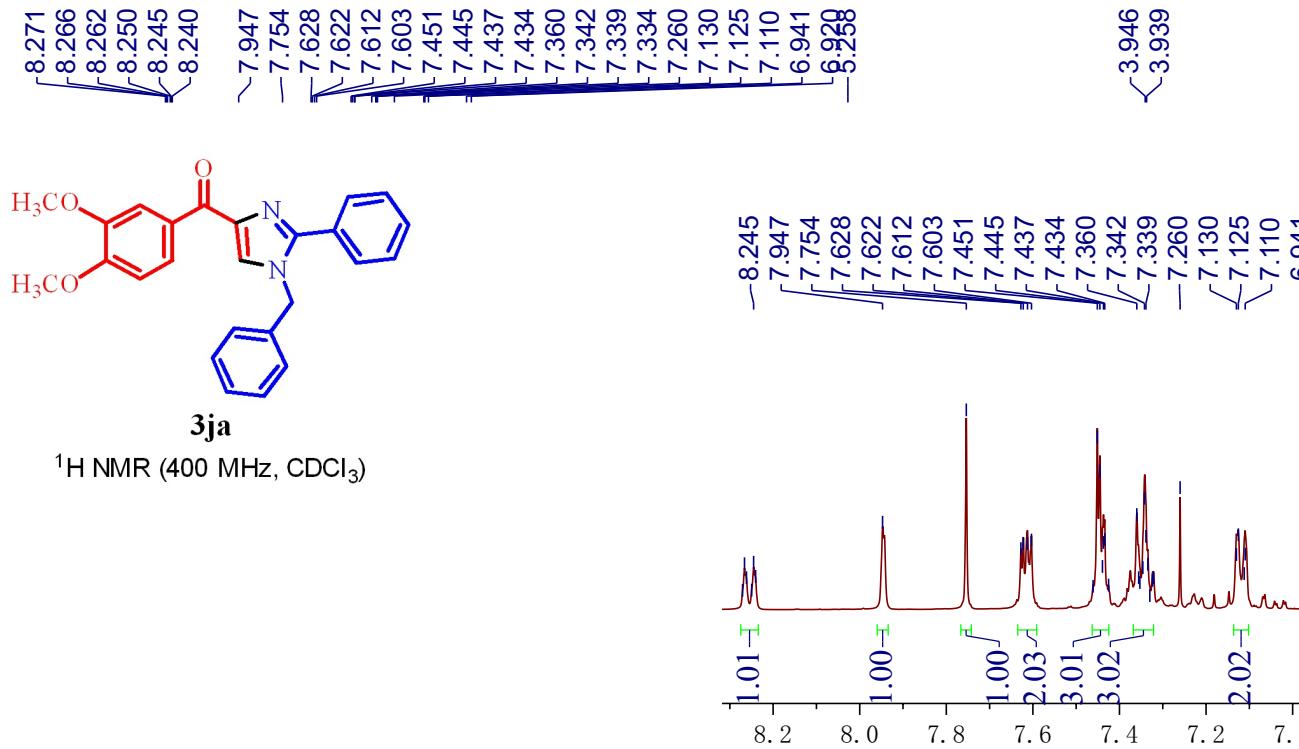


1.01  
1.03  
1.08  
2.02  
1.07  
3.04  
1.01  
3.00  
2.08

2.00

f1 (ppm)



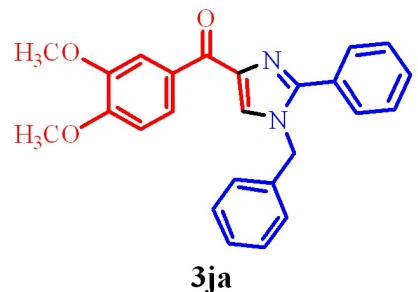


-186.08

~152.92  
148.73  
148.67  
135.91  
130.85  
129.86  
129.64  
129.31  
129.25  
128.83  
128.51  
128.38  
127.08  
125.88  
110.14

77.48  
77.16  
76.84

56.12  
56.08  
51.12

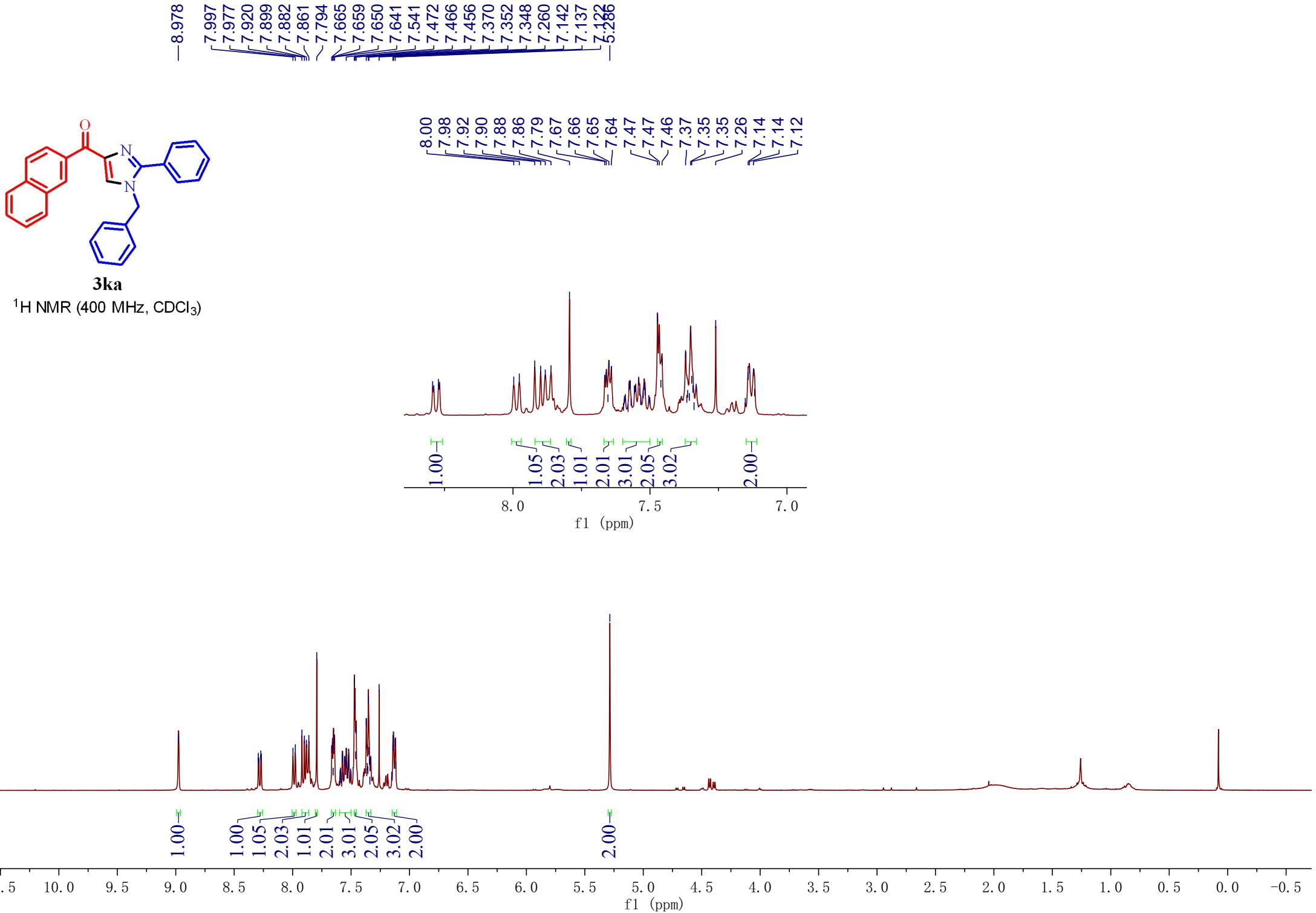


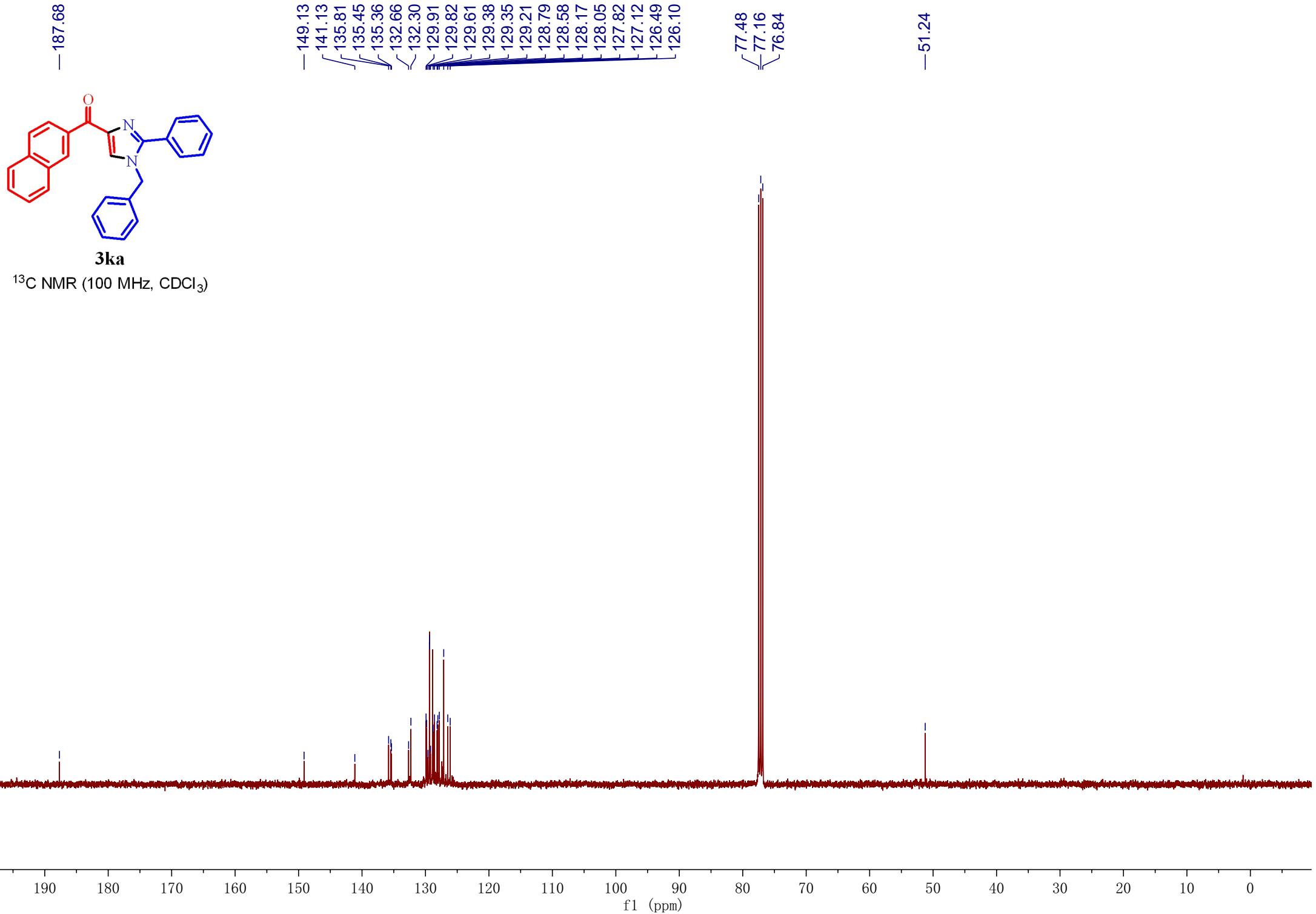
**3ja**

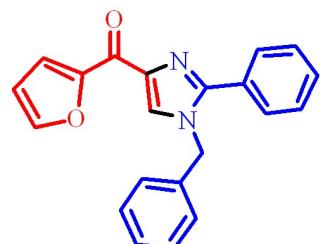
<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)

190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0

f1 (ppm)

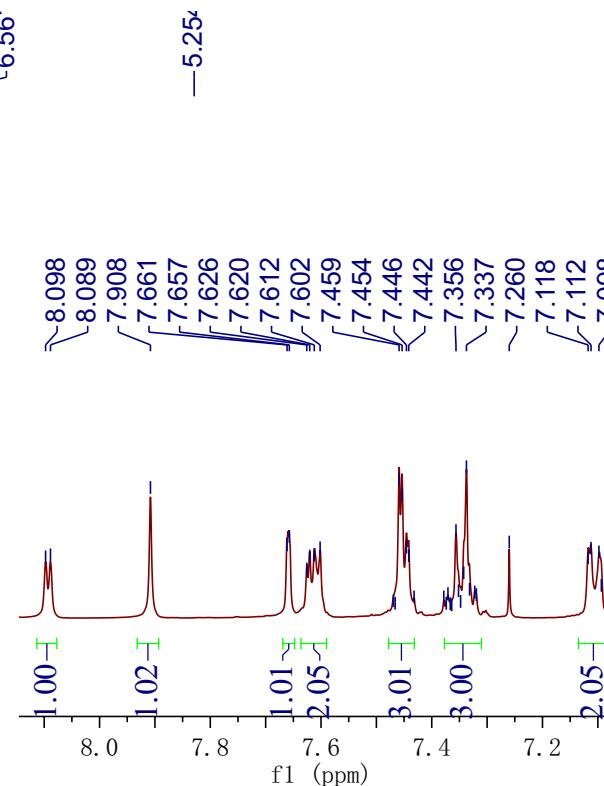
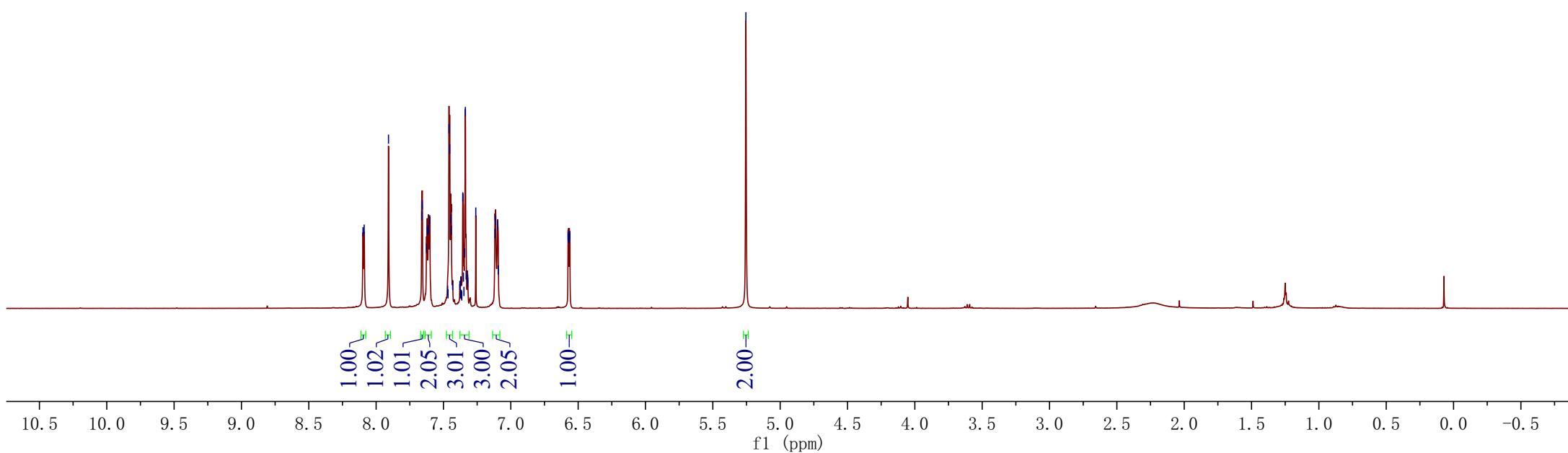


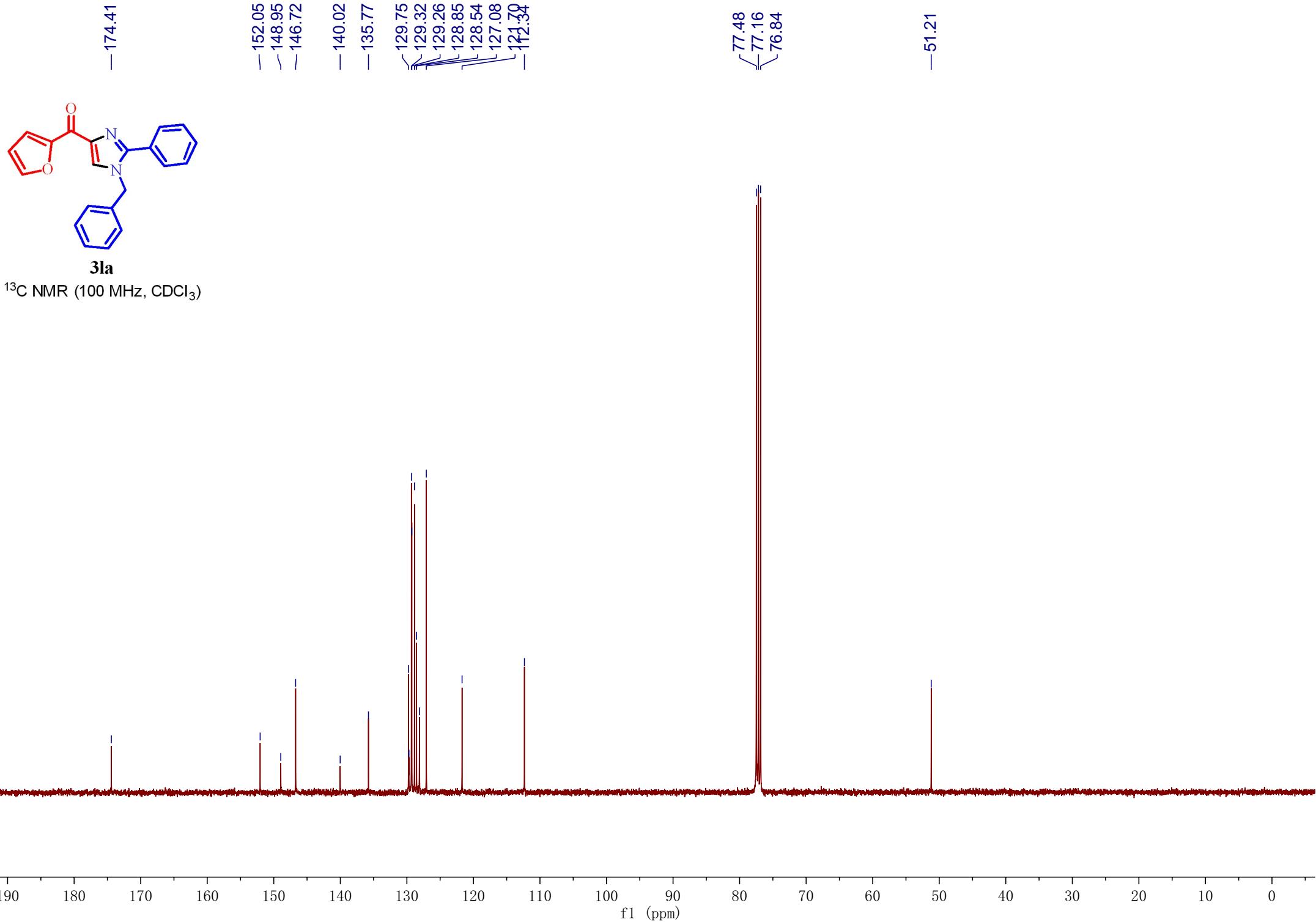


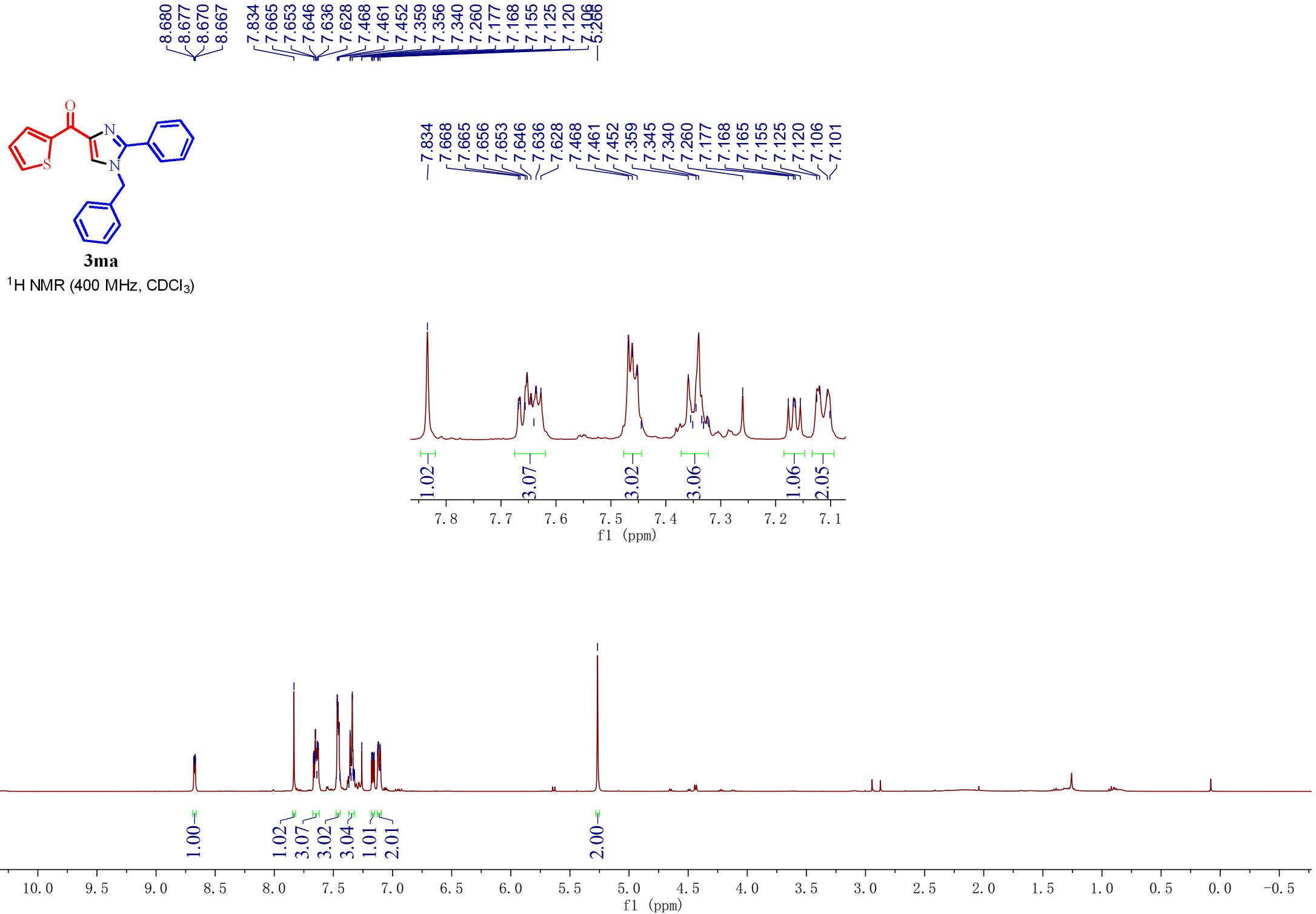


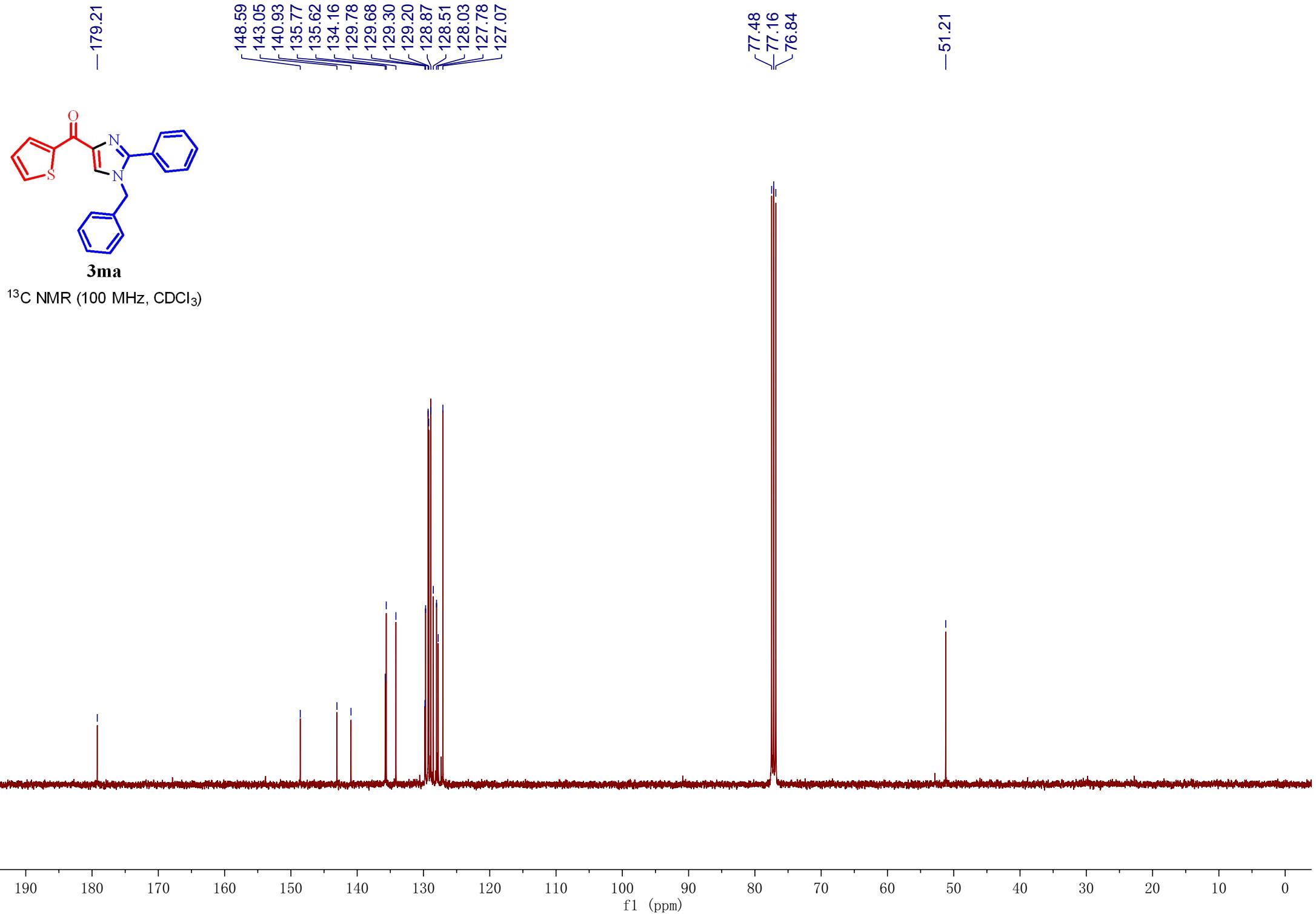
3la

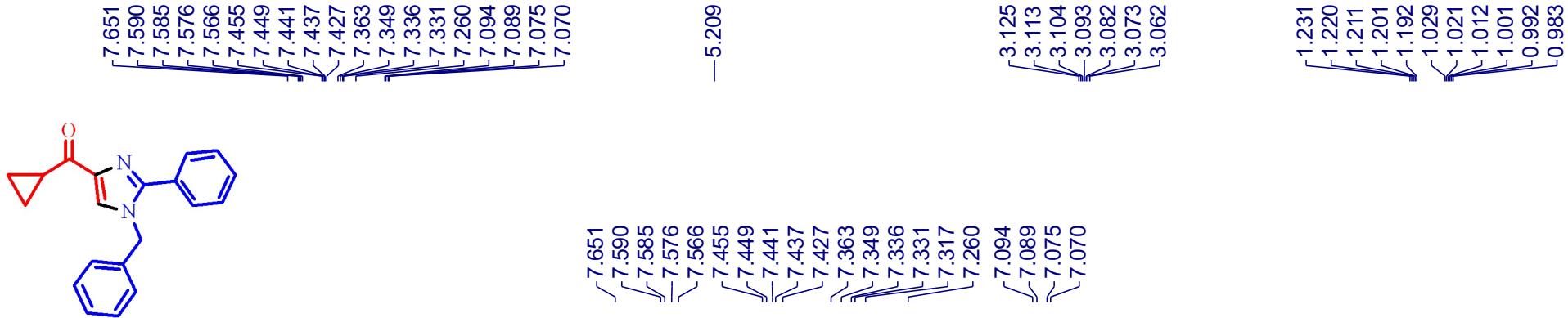
<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)



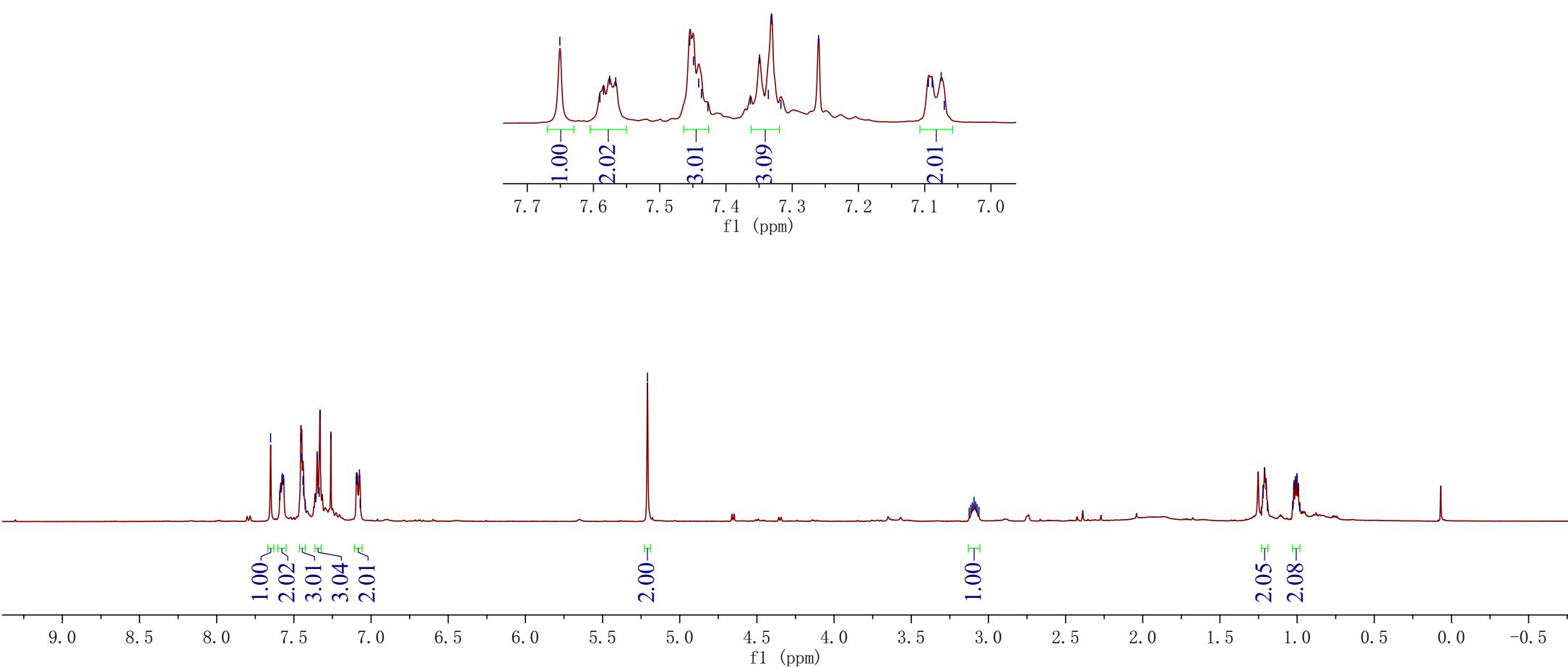


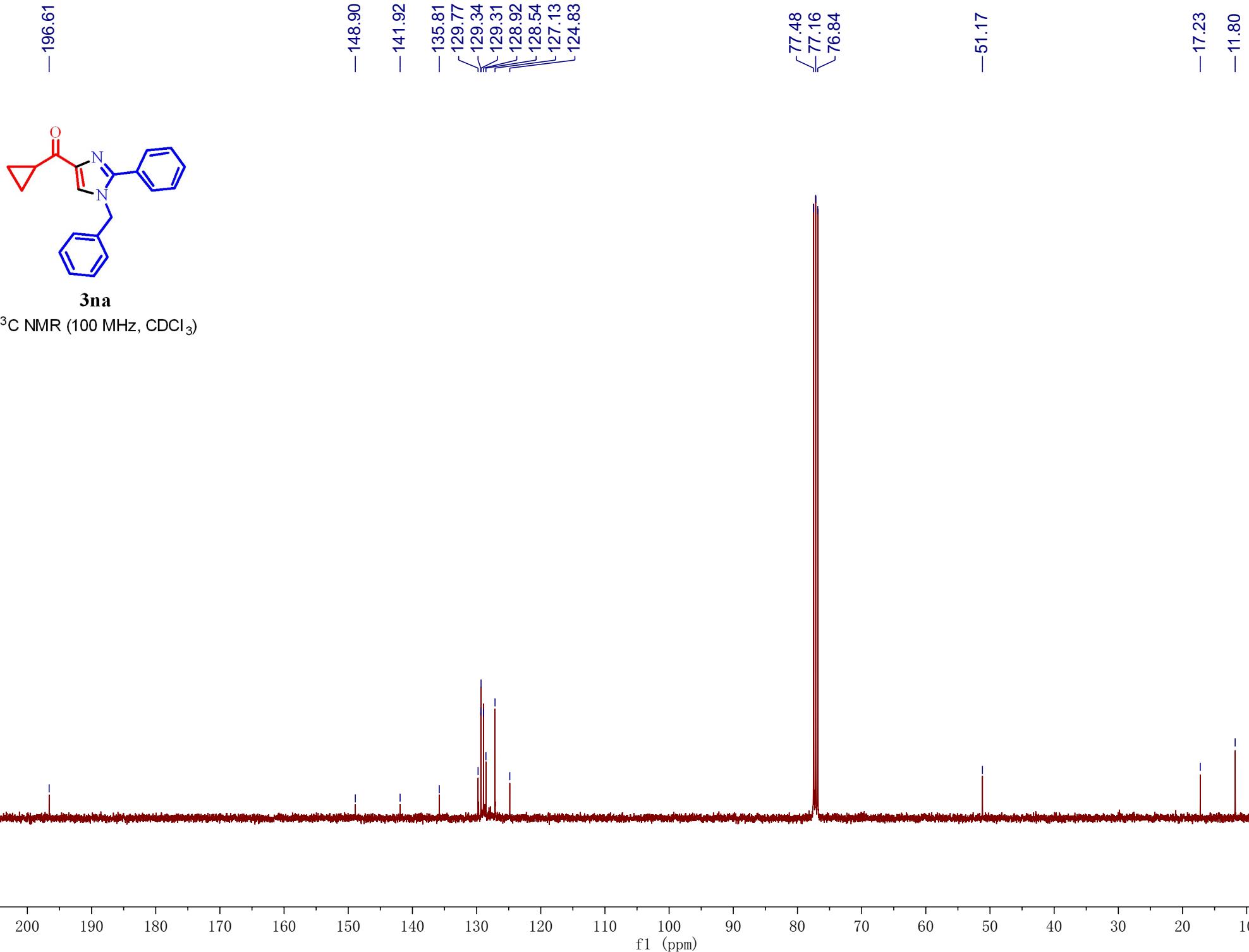




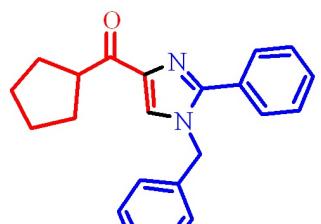


$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )



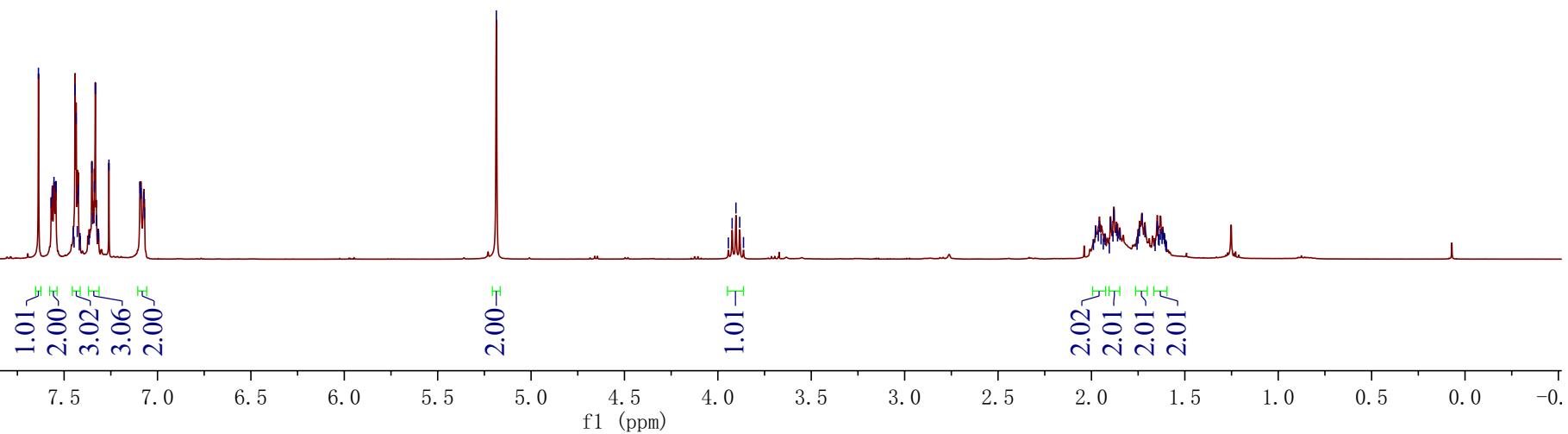
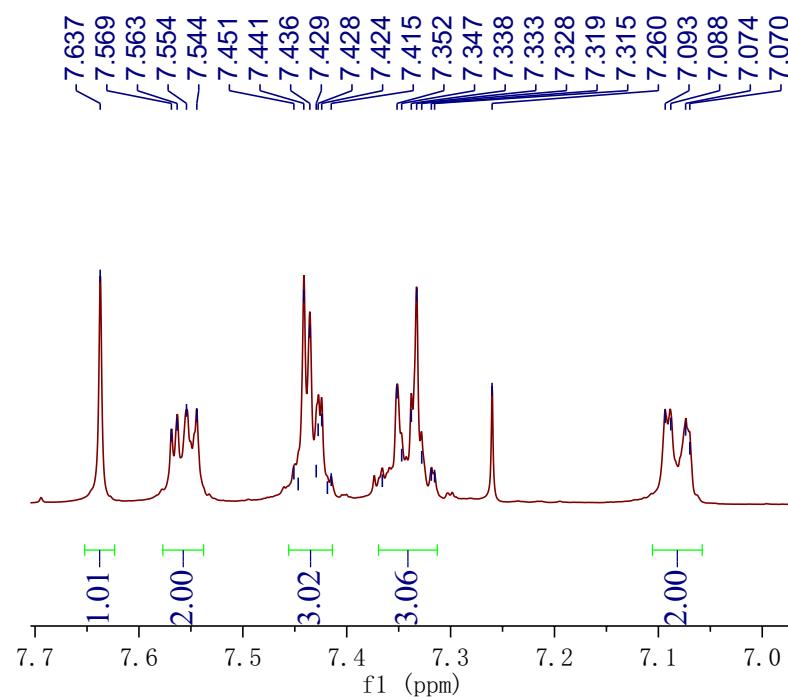


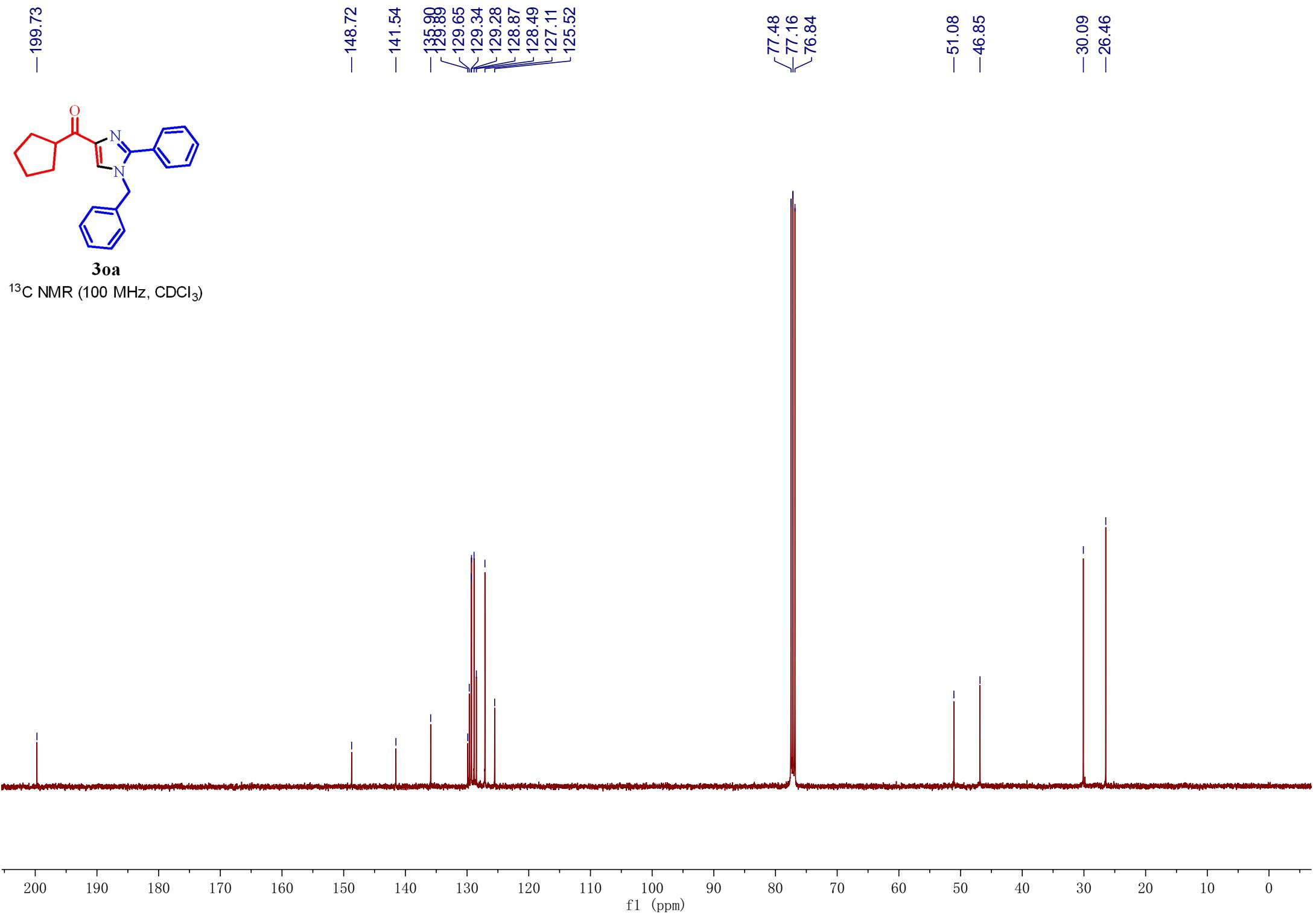
7.637  
7.569  
7.563  
7.554  
7.544  
7.451  
7.447  
7.441  
7.436  
7.429  
7.428  
7.424  
7.419  
7.415  
7.366  
7.352  
7.347  
7.338  
7.333  
7.328  
7.319  
7.315  
7.260  
7.093  
7.088  
7.074  
7.070



**3oa**

$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )



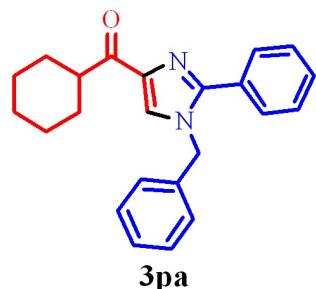


7.637  
7.570  
7.564  
7.555  
7.546  
7.542  
7.443  
7.438  
7.429  
7.426  
7.416  
7.364  
7.350  
7.332  
7.316  
7.260  
7.090  
7.084  
7.070  
7.066

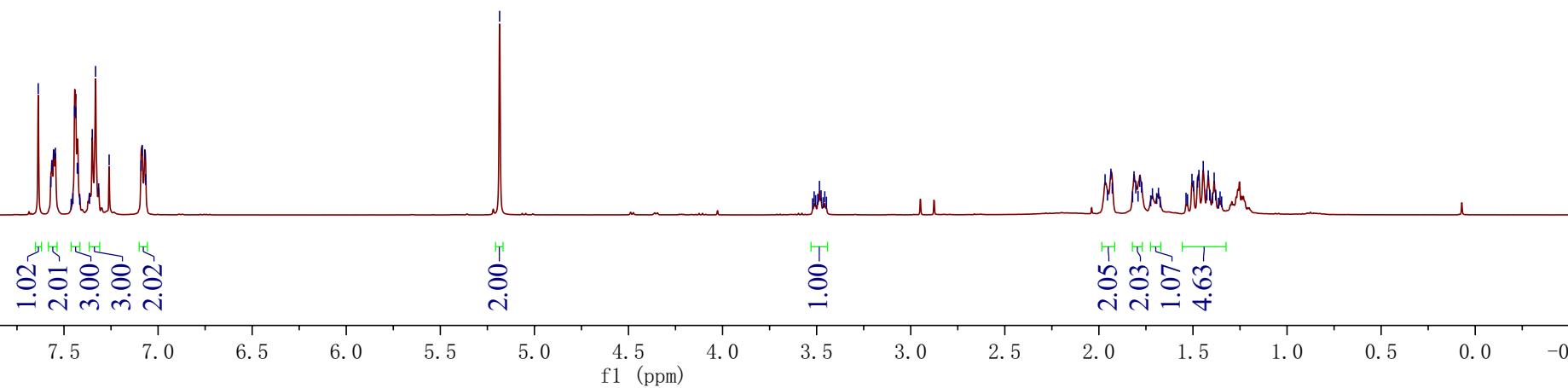
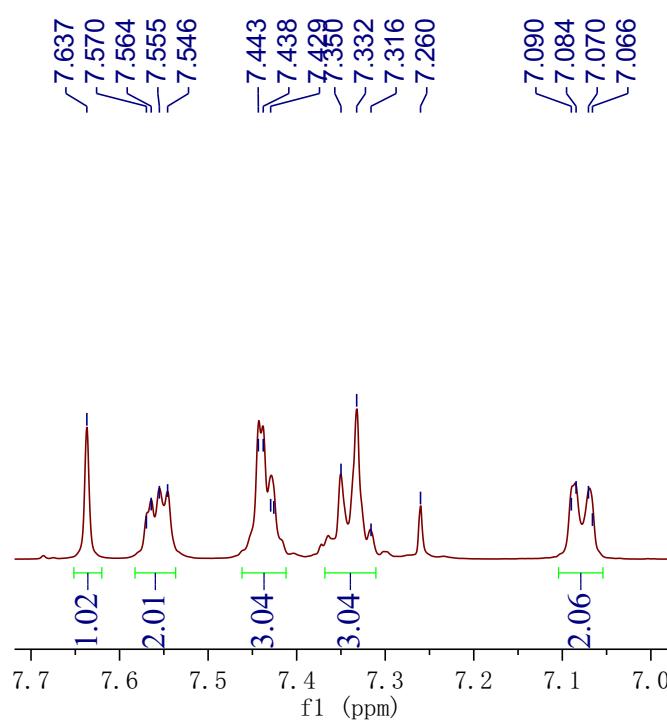
—5.185

3.522  
3.513  
3.505  
3.494  
3.485  
3.477  
3.466  
3.457  
3.448

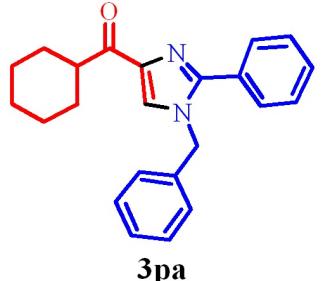
1.967  
1.954  
1.936  
1.929  
1.821  
1.813  
1.804  
1.792  
1.783  
1.773  
1.715  
1.683  
1.505  
1.497  
1.476  
1.468  
1.445  
1.426  
1.419  
1.411  
1.396  
1.387  
1.379  
1.356



$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )



-200.32



<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>)

-148.68

-140.88  
-135.86  
-129.77  
-129.70  
-129.37  
-129.29  
-128.88  
-128.49  
-127.10  
-125.72

77.48  
77.16  
76.84

-51.10  
-45.75

29.07  
26.17  
25.84

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0

f1 (ppm)

