Supporting Information

N-Hydroxyphthalimide Imidate Esters as Amidyl Radical Precursors in the Visible Light Photocatalyzed C-H Amidation of Heteroarenes

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(A) General Information

Melting points were determined on a digital melting point apparatus and temperatures were uncorrected. Proton nuclear magnetic resonance (^{1}H NMR) spectra, carbon nuclear magnetic resonance (^{13}C NMR) spectra and fluorine nuclear magnetic resonance (^{19}F NMR) spectra were recorded at 400, 100 MHz and 376 MHz, respectively. Data are presented as follows: chemical shift (ppm), multiplicity (s =singlet, d = doublet, t = triplet, q = quartet, sept = septet, m = multiplet), coupling constants in Hertz (Hz) and integration. Infrared spectra were recorded on a Perkin-Elmer PE-983 spectrometer with absorption in cm^{-1}. Mass and High resolution mass spectra (HRMS) spectra were recorded by ESI method. The employed solvents were dry up by standard methods when necessary. Commercially obtained reagents were used without further purification. For thin-layer chromatography (TLC), silica gel plates (Huanghai GF254) were used. Flash column chromatography was carried out using 300-400 mesh silica gel at increased pressure. Fluorescence spectra for emission and excitation were obtained on a Hitachi F-4600 FL Spectrophotometer. Cyclic voltammetry (CV) was studied with a CHI 660C electrochemical workstation using a glassy carbon working electrode, a platinum wire counter electrode and an $Ag/AgNO_3$ reference electrode.

Substrates **2a**, **2b**, **2j**, **2n**, **2p**, **2q**, **2r** and **2s** are commercially available. Substrates **2c**, **2d**, **2e**, **2f**, **2g**, **2h**, **2i**, **2k**, **2l**, **2m** and **2o** were synthesized by the procedures reported in the previous literature.[1] Amides are prepared with common methods, using acid chloride and amine as raw materials.

(B) Reaction Setup

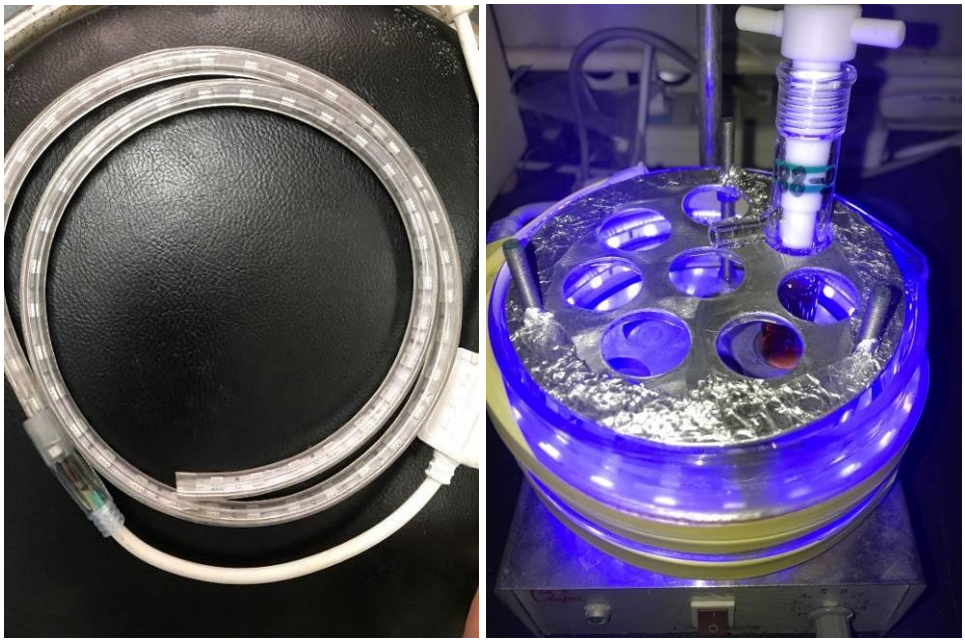


Figure S1. 8 W LEDs strip and reaction setup

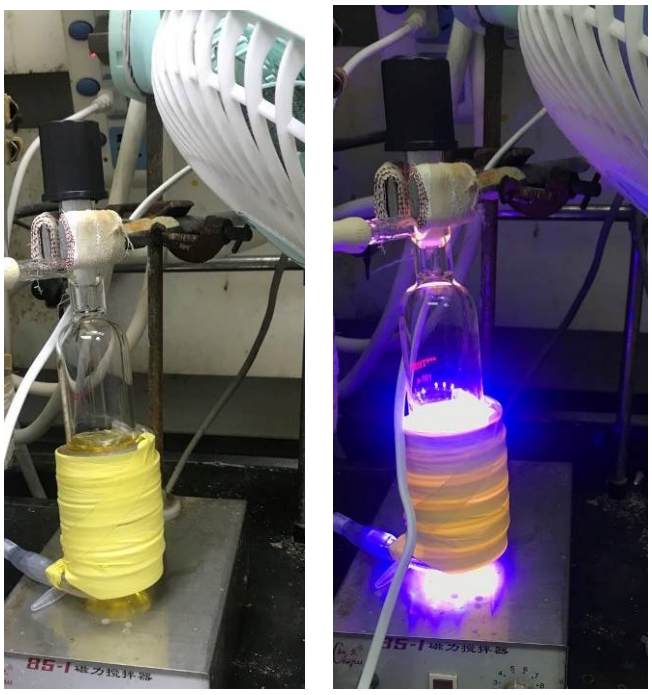
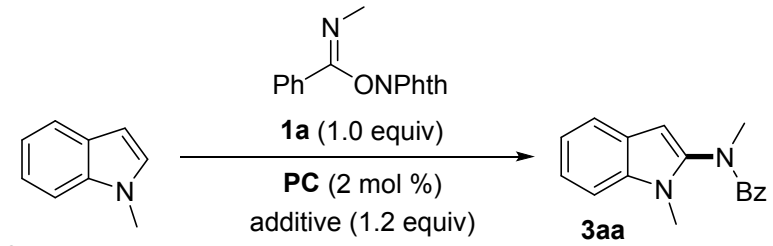


Figure S2. Reaction setup for scale-up synthesis

As depicted in the picture, reactions were carried out in oven-dried sealed tubes. Each of the reaction setup is equipped with a fan to maintain the reaction temperature.

(C) Reaction Optimization and Controls

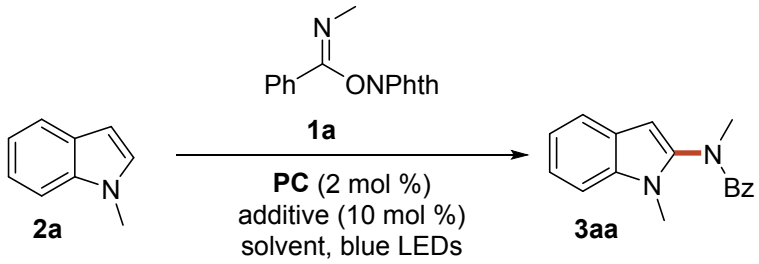
Table S1. Preliminary attempts



entry	**2a / 1a**	additive	solvent	**PC**	yield/%[a]
1	1 / 1	-	CH_3CN	*fac*-Ir(ppy)$_3$	16
2	1 / 1	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	52
3	1 / 1	-	CH_3CN	Ru(bpy)$_3$(BF_4)$_2$	n.r.
4	1 / 1	K_2CO_3	CH_3CN	*fac*-Ir(ppy)$_3$	33
5	1 / 1	K_2CO_3	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	12
6	1 / 1	K_2CO_3	CH_3CN	Ru(bpy)$_3$(BF_4)$_2$	13
7	1 / 1	-	toluene	Ir(ppy)$_2$(dtbbpy)PF_6	11
8	1 / 1	-	DCE	Ir(ppy)$_2$(dtbbpy)PF_6	8
9	1 / 1	-	DMF	Ir(ppy)$_2$(dtbbpy)PF_6	10
10	1 / 1	-	DCM	Ir(ppy)$_2$(dtbbpy)PF_6	4
11	1 / 1	-	THF	Ir(ppy)$_2$(dtbbpy)PF_6	<2
12	1 / 1	-	EA	Ir(ppy)$_2$(dtbbpy)PF_6	12
11	1 / 1	-	dioxane	Ir(ppy)$_2$(dtbbpy)PF_6	12
12	1 / 1	-	acetone	Ir(ppy)$_2$(dtbbpy)PF_6	28
13	1 / 1.2	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	73
14	1 / 1.5	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	54
15	1 / 2.0	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	36
16	1 / 2.5	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	23
17	1.2 / 1	-	CH_3CN	Ir(ppy)$_2$(dtbbpy)PF_6	25

Reaction conditions: unless otherwise noted, a solution of **1a** (0.24 mmol), **2a** (0.2 mmol), **PC** (2 mol %) and additive (1.2 eq) in solvent (2.0 mL) was irradiated by 8 W blue LEDs for 36 h. [a] Yields were determined via ^{1}H-NMR spectroscopic analysis of the crude reaction mixture relative to benzyl methyl ether as an internal standard.

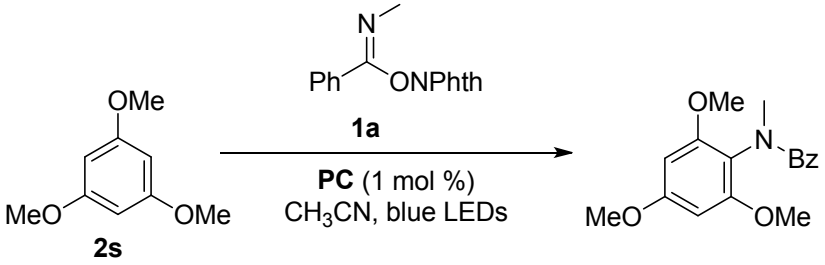
Table S2. Further optimize the reaction conditions



entry	solvent	PC	additive	time/h	yield/%[a]
1	DMSO-d_6 (*c = 0.2*)	$Ir(ppy)_3$	-	12	-
2	DMSO-d_6 (*c = 0.2*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	12	25
3	DMSO-d_6 (*c = 0.2*)	$Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$	-	12	24
4	CD_3CN (*c = 0.2*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	24	35
5	CD_3CN (*c = 0.1*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	12	50
6	CD_3CN (*c = 0.1*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	24	60
7	CD_3CN (*c = 0.1*)	$Ir(ppy)_2(dtbbpy)PF_6$	TFA	12	-
8	CD_3CN (*c = 0.1*)	$Ir(ppy)_3$	TFA	12	-
9	CD_3CN (*c = 0.1*)	Eosin Y	-	12	n.r.
10	CD_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	12	72
11	CD_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	24	75
12	CD_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$	-	36	76
13	***CH_3CN (c = 0.05)***	***$Ir(ppy)_2(dtbbpy)PF_6$***	***-***	***12***	***74/73[b]***
14	***CH_3CN (c = 0.05)***	***$Ir(ppy)_2(dtbbpy)PF_6$ (1 mol %)***		***12***	***74***
15	CH_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$ (1 mol %), **1a** (1.5 eq)		12	63
16	CH_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$ (1 mol %), **1a** (2.0 eq)		12	40
14	CH_3CN (*c = 0.05*)	-		12	0
15[c]	CH_3CN (*c = 0.05*)	$Ir(ppy)_2(dtbbpy)PF_6$		12	0

Reaction conditions: unless otherwise noted, a solution of **1a** (0.24 mmol), **2a** (0.2 mmol), **PC** (2 mol %) and additive (10 mol %) in solvent was irradiated by 8 W blue LEDs at room temperature for 12 h. [a] Yields were determined via ^{1}H-NMR spectroscopic analysis of the crude reaction mixture relative to benzyl methyl ether as an internal standard. [b] Isolated yield. [c] in dark.

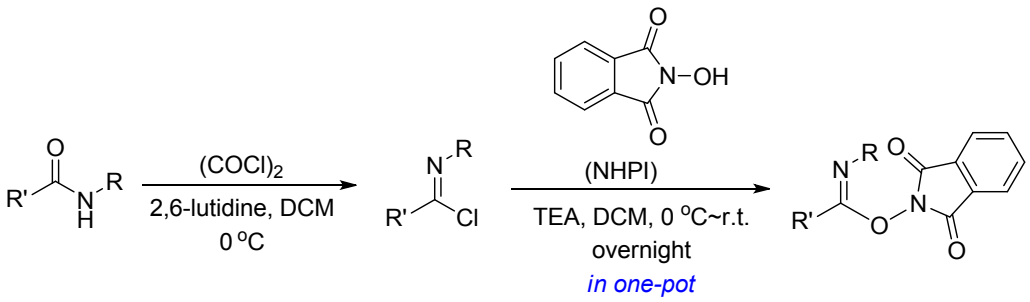
Table S3. Optimize the reaction conditions of 1,3,5-trimethoxybenzene (2s) with 1a



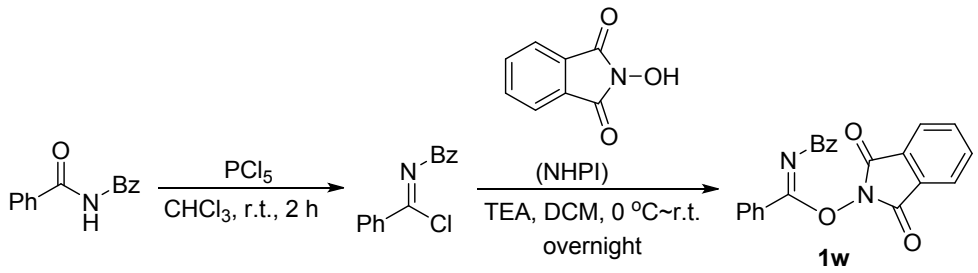
entry	PC	time/h	yield/%[a]
1	$Ir(ppy)_3$	12	13
2	$Ir(ppy)_2(dtbbpy)PF_6$	12	6
3	$Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$	12	9
4	$Ir(dFCF_3ppy)_2(bpy)PF_6$	12	22
5	4CzIPN	12	9[b]
6	$Ir(dFCF_3ppy)_2(5,5'\text{-}dCF_3bpy)PF_6$	12	0
7	$Ir(dFCF_3ppy)_2(bpy)PF_6$	24	24
8	$Ir(dFCF_3ppy)_2(bpy)PF_6$	36	26[c]

Reaction conditions: unless otherwise noted, a solution of **1a** (0.24 mmol), **2s** (0.2 mmol) and **PC** (1 mol %) in 4 mL of CH_3CN was irradiated by 8 W blue LEDs at room temperature for specific time. [a] Yields were determined via ^{1}H-NMR spectroscopic analysis of the crude reaction mixture relative to benzyl methyl ether as an internal standard. [b] with 2 mol% of **PC**. [c] Isolated yield.

(D) General Procedure for the Preparation of NHPI Imidate Esters

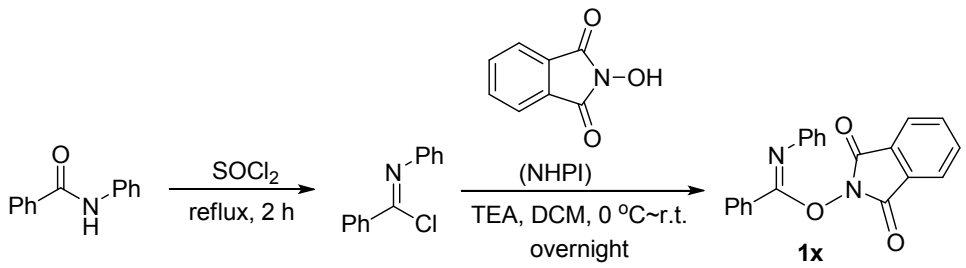


To a solution of 1.0 equiv of amide in CH_2Cl_2 (0.2 M) was added 1.5 equiv of 2,6-lutidine then 1.0 equiv of oxalyl chloride dropwise at 0 °C under an argon atmosphere resulting in the evolution of gas. After stirring for 20-45 min, 1.0 equiv of NHPI was added all at once and 2.5 equiv of triethylamine was added dropwise. Upon finishing the addition of triethylamine, the cooling bath was removed. After the reaction mixture was stirred overnight at room temperature, and CH_2Cl_2 was removed. The residue was diluted with 20 mL of ethyl acetate, filtered, and concentrated, then the corresponding residue was purified by column chromatography (SiO_2) using pure DCM or EtOAc/petroleum ether solvent mixtures as eluent as indicated.



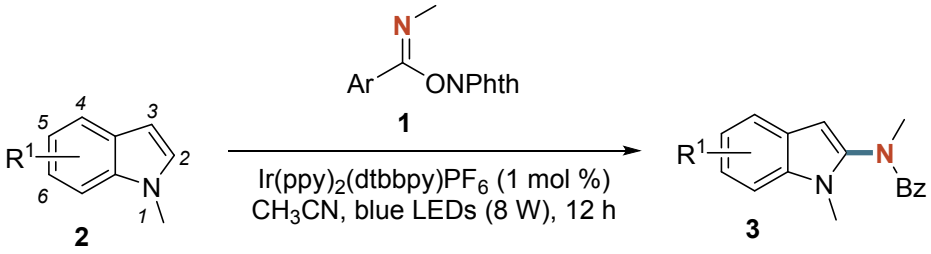
An intimate mixture of 296 mg (1.31 mmol) of dibenzamide and 328 mg (1.58 mmol) of phosphorus pentachloride was treated with about 5.0 mL of dried chloroform and was stirred for 2 h at room temperature. The resulting chloroform solution was diluted with 15 mL of ether and shaken with iced water to remove phosphoryl chloride, and the solution was dried over with anhydrous Na_2SO_4 and allowed to evaporate to give the α-chloro-*N*-benzoylbenzimide.[2] To the mixture of the obtained α-chloro-*N*-benzoylbenzimide and 1.0 equiv of NHPI was added 10 mL of DCM, and then, 1.2 equiv of triethylamine was added dropwise at room temperature. After the reaction mixture was stirred overnight at room temperature, the solvent was removed under reduced pressure and the

residue was purified by a column chromatography (SiO_2) directly using pure DCM as eluent.



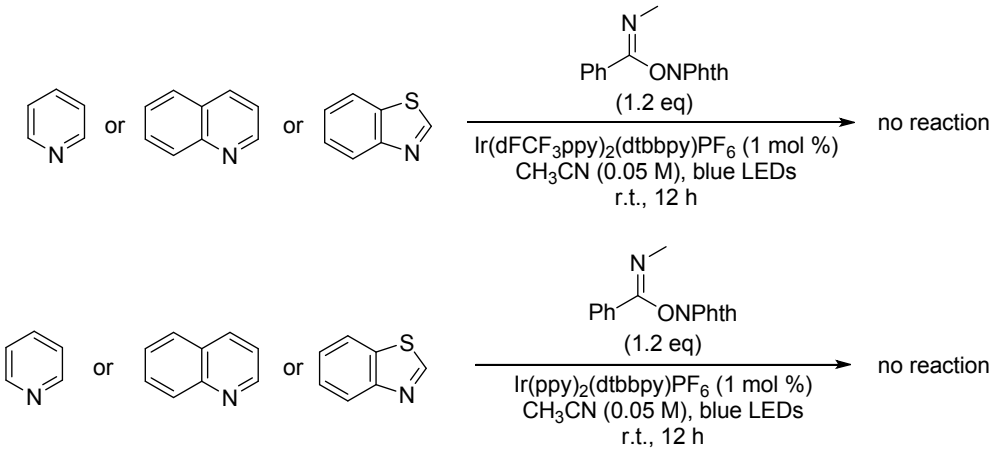
An intimate mixture of 985 mg (5.0 mmol) of benzamide and 5 mL of $SOCl_2$ was refluxed for 2 h. The resulting solution was allowed to evaporate to give 967 mg of *N*-phenyl-benzimidoyl chloride. To the mixture of 518 mg (2.4 mmol) of *N*-phenyl-benzimidoyl chloride and 402 mg (2.0 mmol) of NHPI was added 10 mL of DCM, and then, 1.2 equiv of triethylamine was added dropwise at room temperature. After the reaction mixture was stirred overnight at room temperature, the solvent was removed under reduced pressure and the residue was purified by a column chromatography (SiO_2) directly using pure DCM as eluent.

(E) General Procedure for the Visible Light Photocatalyzed C-H Amidation of Heteroarenes

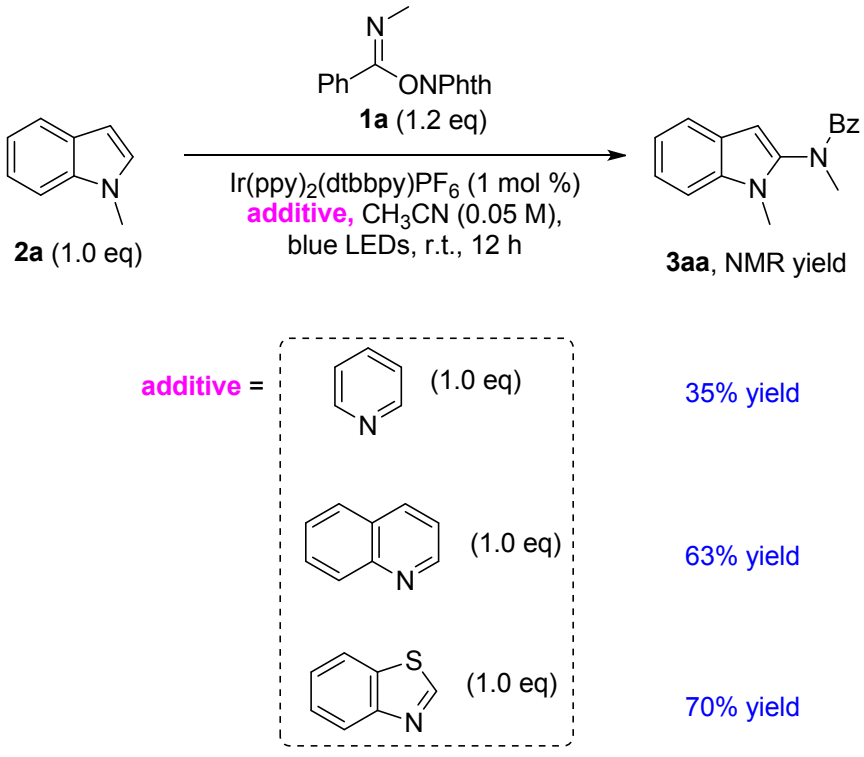


An oven-dried 5 mL sealed tube equipped with a Teflon septum and a magnetic stir bar was charged with the photocatalyst (0.002 mmol, 0.01 equiv, either $Ir(ppy)_2(dtbbpy)PF_6$ or $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$), NHPI imidate ester (0.24 mmol, 1.2 equiv) and the corresponding heteroarene (0.2 mmol, 1.0 equiv, if solid), 4.5 mL of dry CH_3CN was added via a syringe and the mixture was degassed via argon bubbling for 10 min. The tube was purged with a stream of argon and the corresponding heteroarene (0.2 mmol, 1.0 equiv, if liquid) was added via a syringe and the tube was sealed with a Teflon septum. Then the vail was placed in blue light from a 8 W blue LEDs strip at room temperature (a fan was employed to maintain this temperature). After the indicated time period, the reaction was concentrated *in vacuo* directly. Purification of the crude product by a column chromatography (SiO_2) or prep-TLC using the indicated solvent system as eluent afforded the desired amidation product.

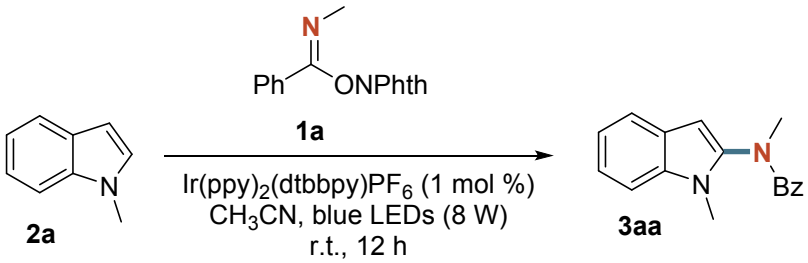
(F) Reactions of Some Other Heteroarenes



Pyridine, quinoline, benzothiazole were also not suitable for these reaction conditions (see the above Scheme), but they would not significantly suppress the reaction of **1a** and **2a** when they are used as additives (see the Scheme shown below).



(G) Scale-up Synthesis



An oven-dried 100 mL sealed tube equipped with a Teflon septum and magnetic stir bar was charged with the photocatalyst $Ir(ppy)_2(dtbbpy)PF_6$ (27.4 mg, 0.03 mmol, 0.01 equiv), NHPI imidate ester **1a** (1.0 g, 3.6 mmol, 1.2 equiv), 70 mL of dry CH_3CN was added via syringe and the mixture was degassed via argon bubbling for 30 min. The tube was purged with a stream of argon and the corresponding *N*-Me-indole **2a** (394 mg, 3.0 mmol, 1.0 equiv) was added via syringe and the mixture was degassed via argon bubbling for another 5 min. The tube was sealed with Teflon septum. Then the vail placed in blue light from a 8W blue LEDs strip at room temperature (a fan was employed to maintain this temperature). After stirring for 12 h in blue light, the reaction was concentrated *in vacuo* directly. Purification of the crude product by chromatography on silica gel using DCM as eluent afforded the desired amidation product **3aa** (484 mg, 61% yield, pala yellow solid).

(H) Cyclic Voltammetry Experiments

The cyclic voltammetry experiments were performed. The oxidation potential of *N*-Me-indole **2a** was determined as: **2a** (E_p^{ox} = 0.84 V vs $Ag/AgNO_3$, 1.14 V vs SCE) (Figure S3). The data are consistent with the reference.[3] The oxidation potential of *N*-Ph-indole **2n** was determined as: **2n** (E_p^{ox} = 1.03 V vs $Ag/AgNO_3$, 1.33 V vs SCE) (Figure S3). The reduction potential of NHPI imidate ester **1a** was determined as: **1a** (E_p^{re} = -1.61 V vs $Ag/AgNO_3$, -1.31V vs SCE) (Figure S4). The cyclic voltammetry (CV) was studied with a CHI 660C electrochemical workstation using a glassy carbon working electrode, a platinum wire counter electrode and an $Ag/AgNO_3$ reference electrode (volume 10 mL; acetonitrile as solvent, $n Bu_4N^+PF_6^-$ 0.05 M as the supporting electrolyte, **2a**, **2n** and **1a** (2 mM) as the tested compound). The scan speed was 100 $mV \cdot s^{-1}$. The potential ranges investigated for oxidations were 0 to +2 V vs $Ag/AgNO_3$ for **2a** and **2n**. The potential ranges investigated for reduction was -2 V to 0 V vs $Ag/AgNO_3$ for **1a** (Referenced to SCE by subtracting 0.3 V from the relative value).

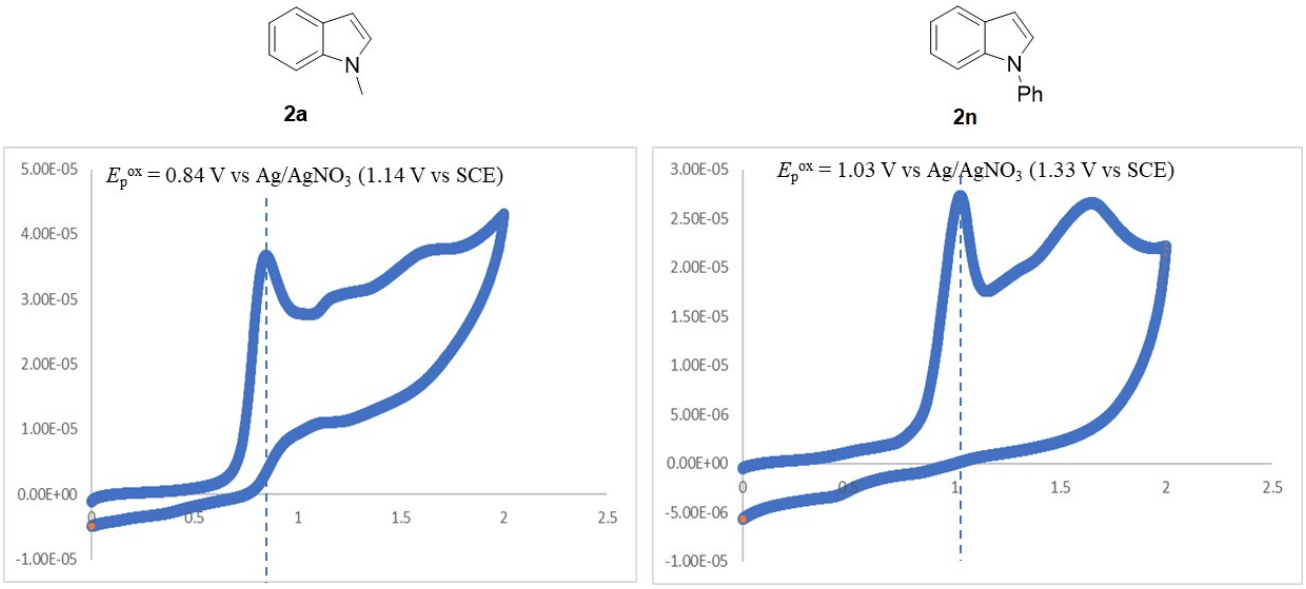


Figure S3. Cyclic voltammetry of **2a** and **2n**

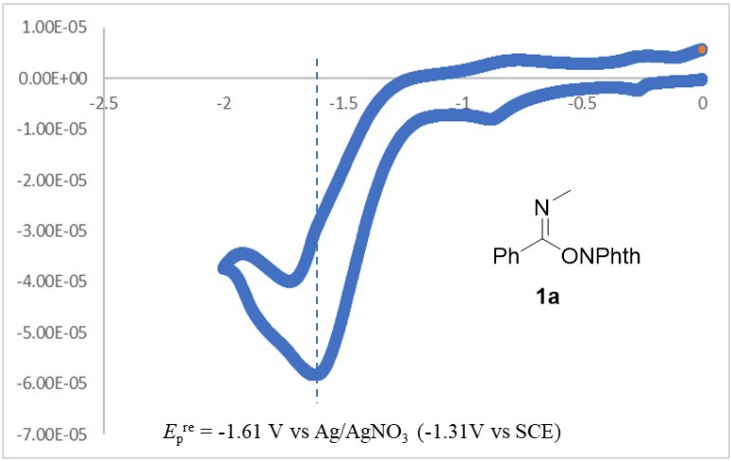


Figure S4. Cyclic voltammetry of **1a**

(I) Stern-Volmer Quenching Studies

The measurements were performed using a 0.2 mM solution of photocatalyst ($Ir(ppy)_2(dtbbpy)PF_6$ or $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$) in 2.0 mL of degassed CH_3CN with varying concentration of a quencher. The samples were excited at 400 nm and emission intensity was recorded at 563 nm for $Ir(ppy)_2(dtbbpy)PF_6$ or 487 nm for $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$.

The quenching of the excited state $Ir(III)^*$ by NHPI imidate ester **1a**, *N*-Me-indole **2a** and *N*-Ph-indole **2n** was carried out in CH_3CN separately (Figures S5-S7). The results revealed that **2a** and **2n** could significantly quench $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6^*$ (Figure S5). Indoles **2a** and **2n** also display slight quenching ability to $Ir(ppy)_2(dtbbpy)PF_6^*$, and the quenching ability of **2a** is better than that of **2n** (Figure S6). $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6^*$ and $Ir(ppy)_2(dtbbpy)PF_6^*$ could not be quenched by **1a** (Figure S7).

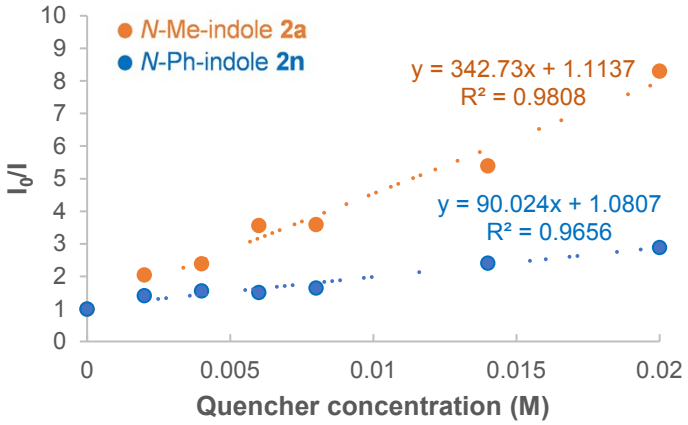


Figure S5. $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$ emission quenching with **2a** and **2n**

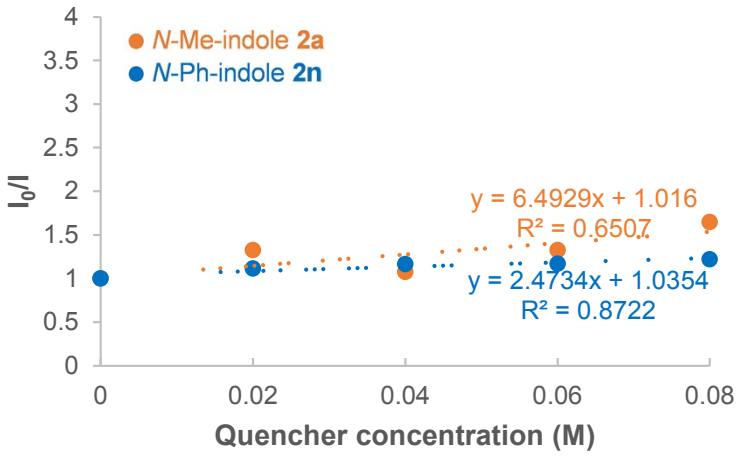


Figure S6. $Ir(ppy)_2(dtbbpy)PF_6$ emission quenching with **2a** and **2n**

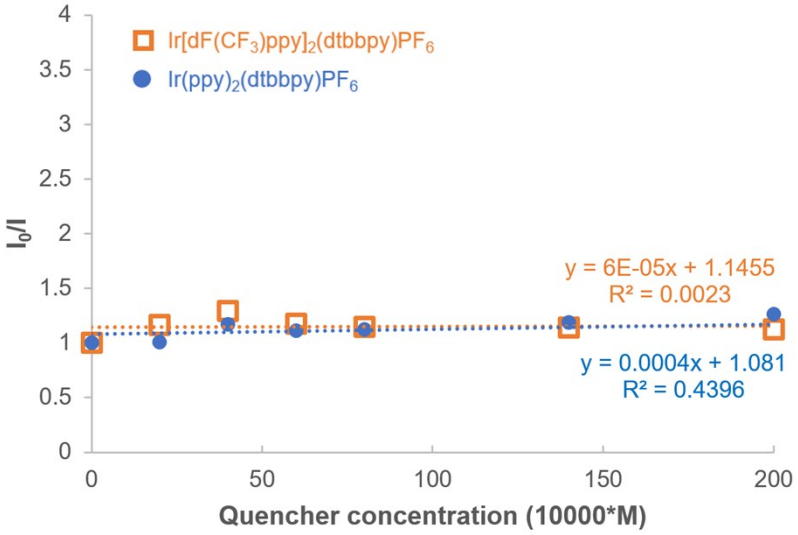
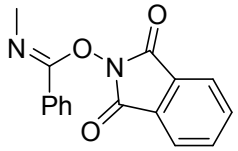
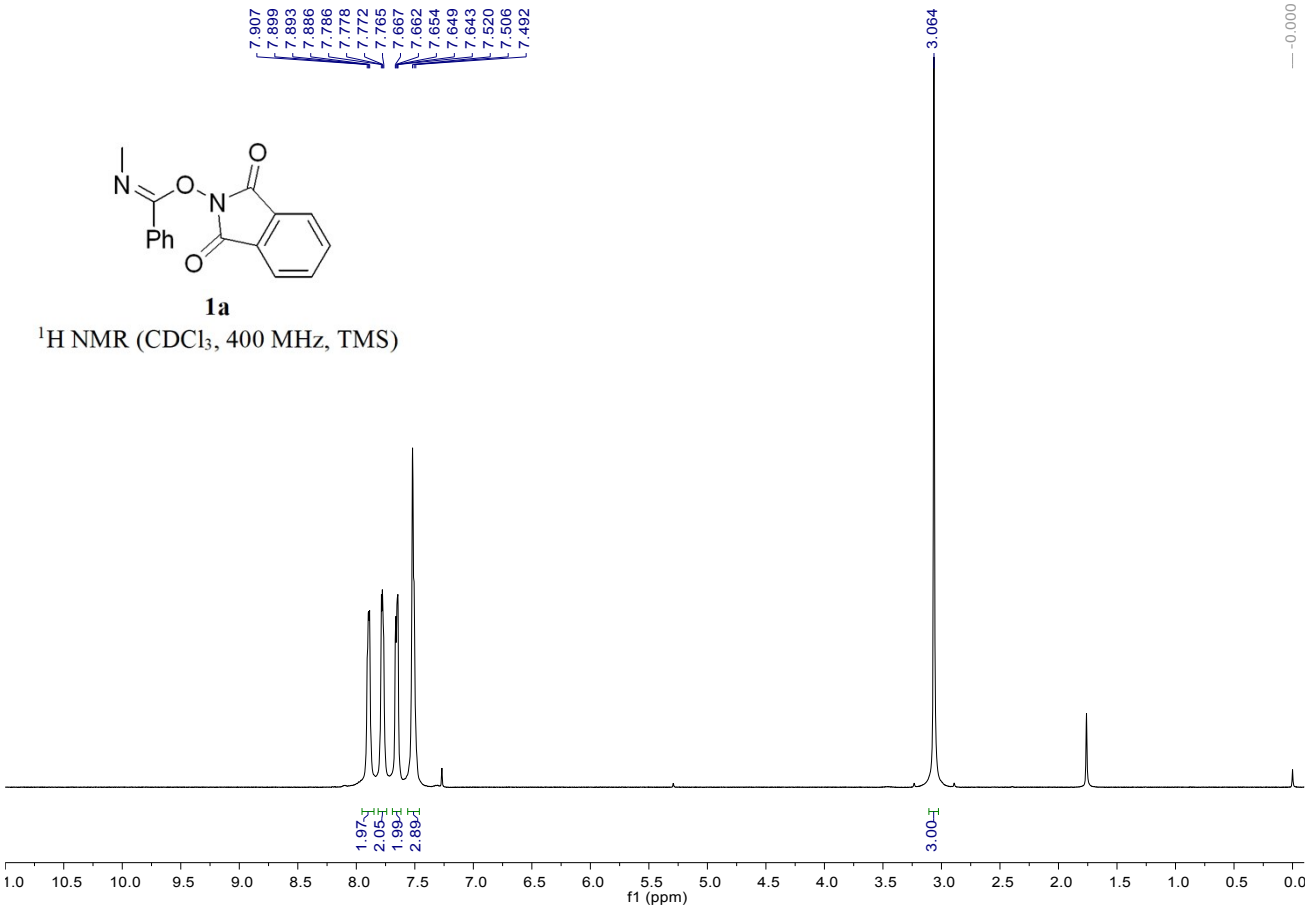


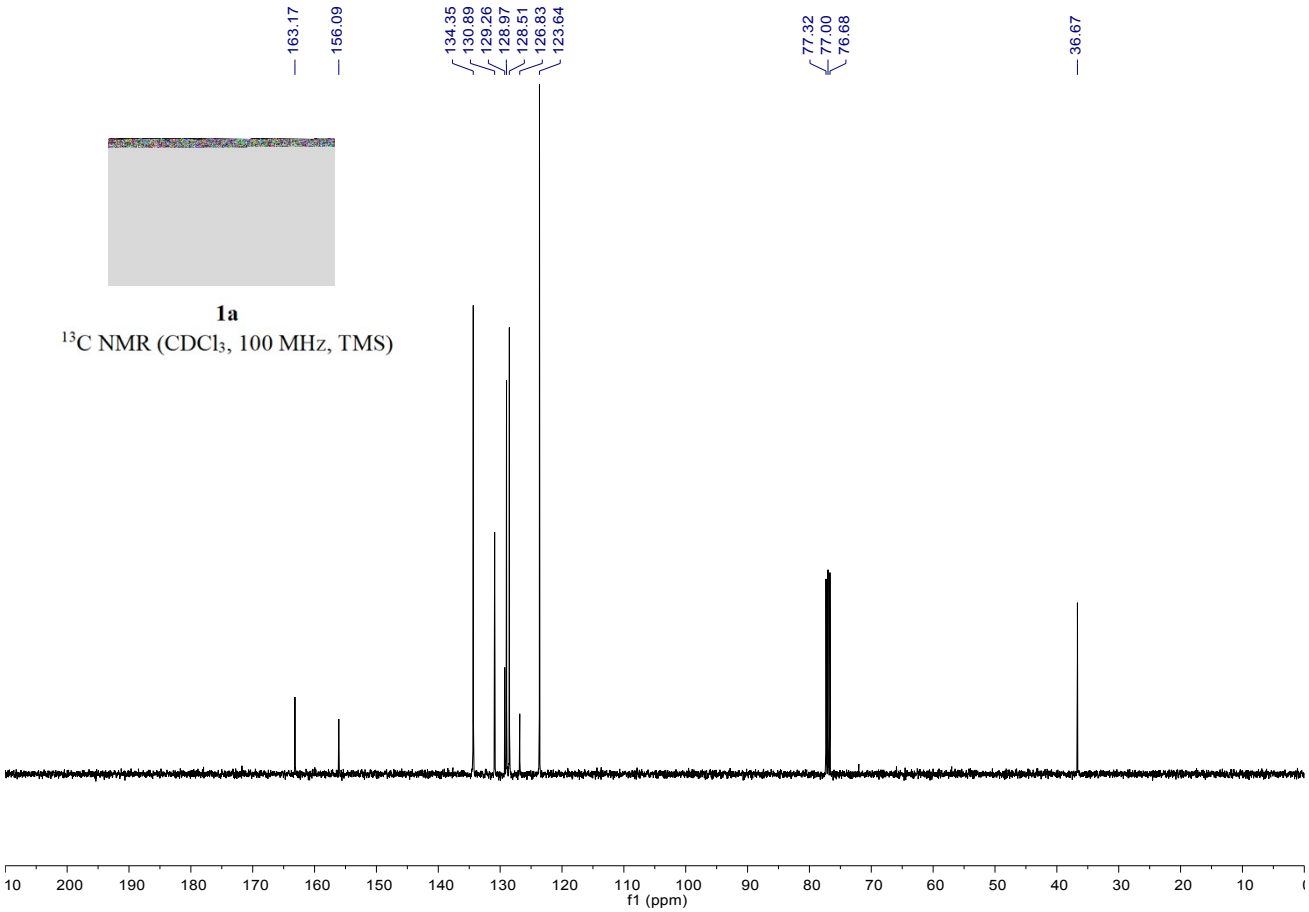
Figure S7. $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$ and $Ir(ppy)_2(dtbbpy)PF_6$ emission quenching with **1a**

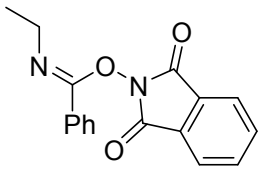
(J) Spectroscopic Data of Substrates and Products



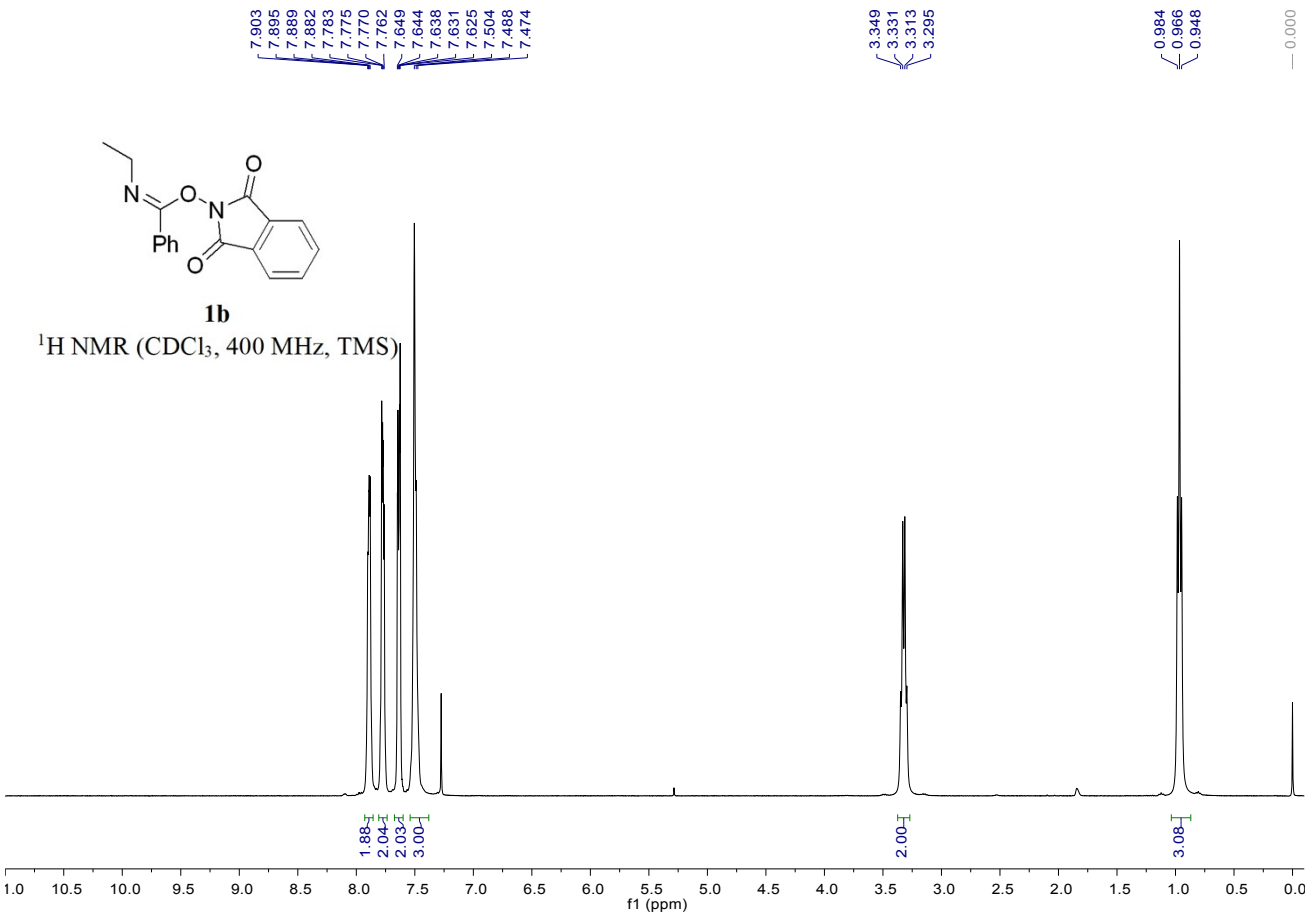
1,3-dioxoisoindolin-2-yl (*Z*)-N-methylbenzimidate (1a). The title product was obtained after purification by column chromatography (DCM). A white solid, 956 mg, 75% yield; M.p.: 176-178 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.90 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.78 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.69 - 7.62 (m, 2H), 7.56 - 7.46 (m, 3H), 3.06 (s, 3H). ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 156.1, 134.4, 130.9, 129.3, 129.0, 128.5, 126.8, 123.6, 36.7; IR (neat) ν 1790, 1720, 1465, 1372, 1241, 1184, 1138, 1006, 877, 755, 692 cm^{-1}; HRMS (ESI) Calcd. for $C_{16}H_{13}N_2O_3^+$ Requires: 281.0921, Found: 281.0923.





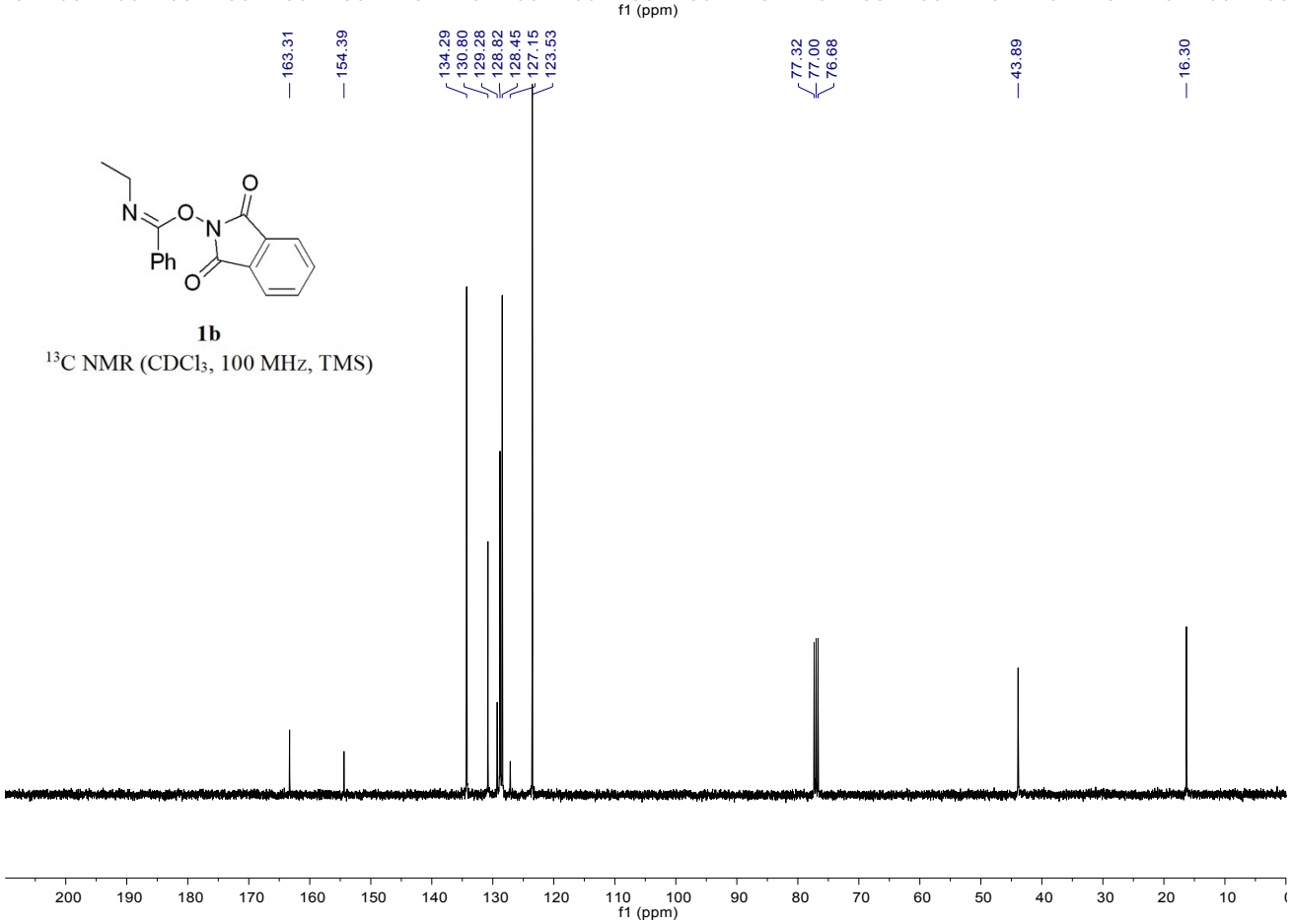


1,3-dioxoisoindolin-2-yl (*Z*)-N-ethylbenzimidate (1b). The title product was obtained after purification by column chromatography (PE:EA = 4:1). A white solid, 936 mg, 70% yield; M.p.: 125-127 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.89 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.77 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.68 - 7.60 (m, 2H), 7.54 - 7.42 (m, 3H), 3.32 (q, *J* = 7.2 Hz, 2H), 0.97 (t, *J* = 7.2 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.3, 154.4, 134.3, 130.8, 129.3, 128.8, 128.5, 127.1, 123.5, 43.9, 16.3; IR (neat) ν 2969, 1788, 1735, 1697, 1447, 1376, 1233, 1187, 1140, 1005, 878, 776, 693 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2O_3^+$Requires: 295.1077, Found: 295.1075.



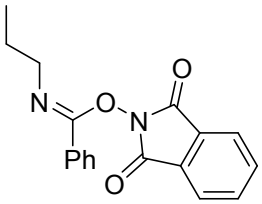
1b

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

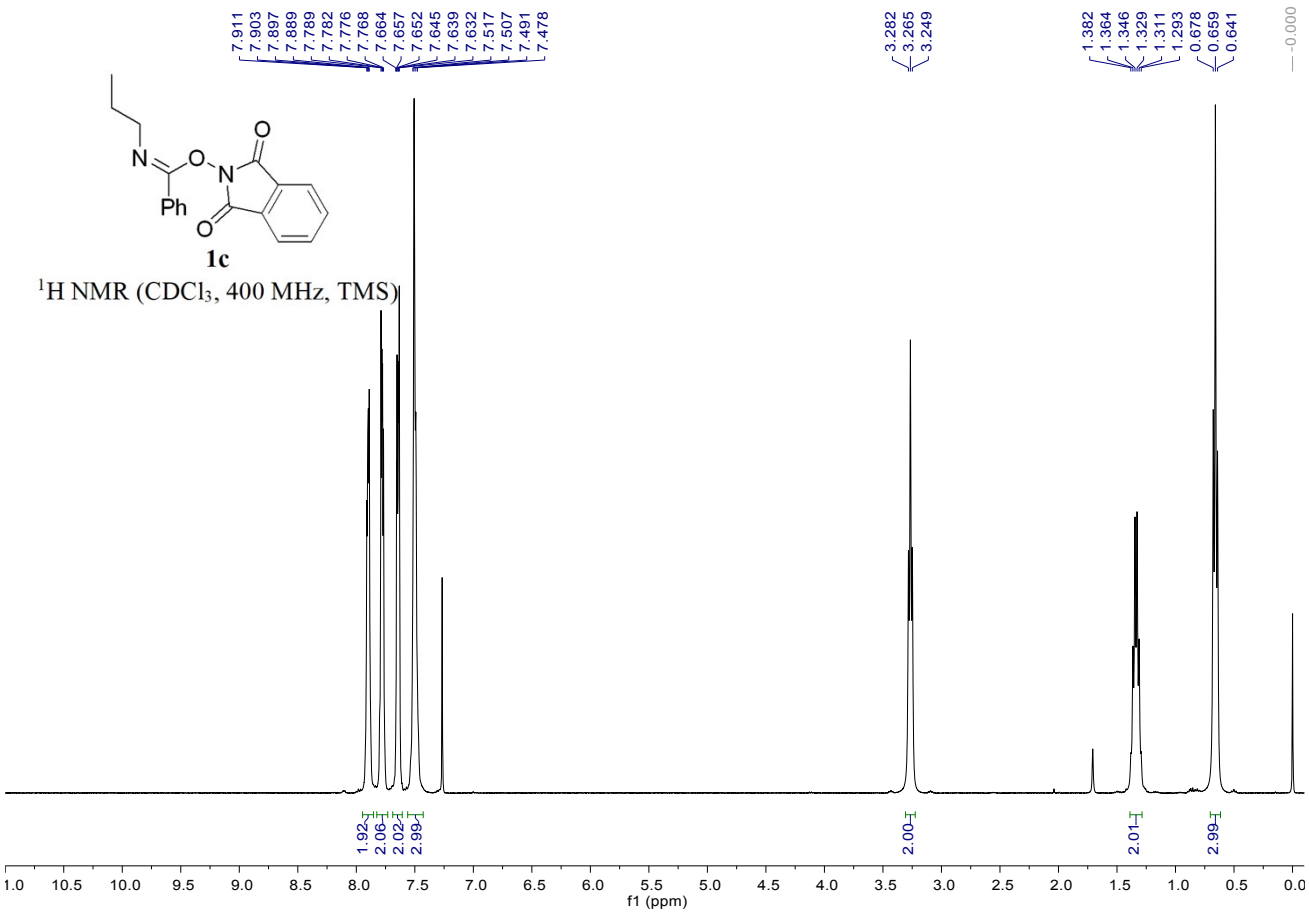


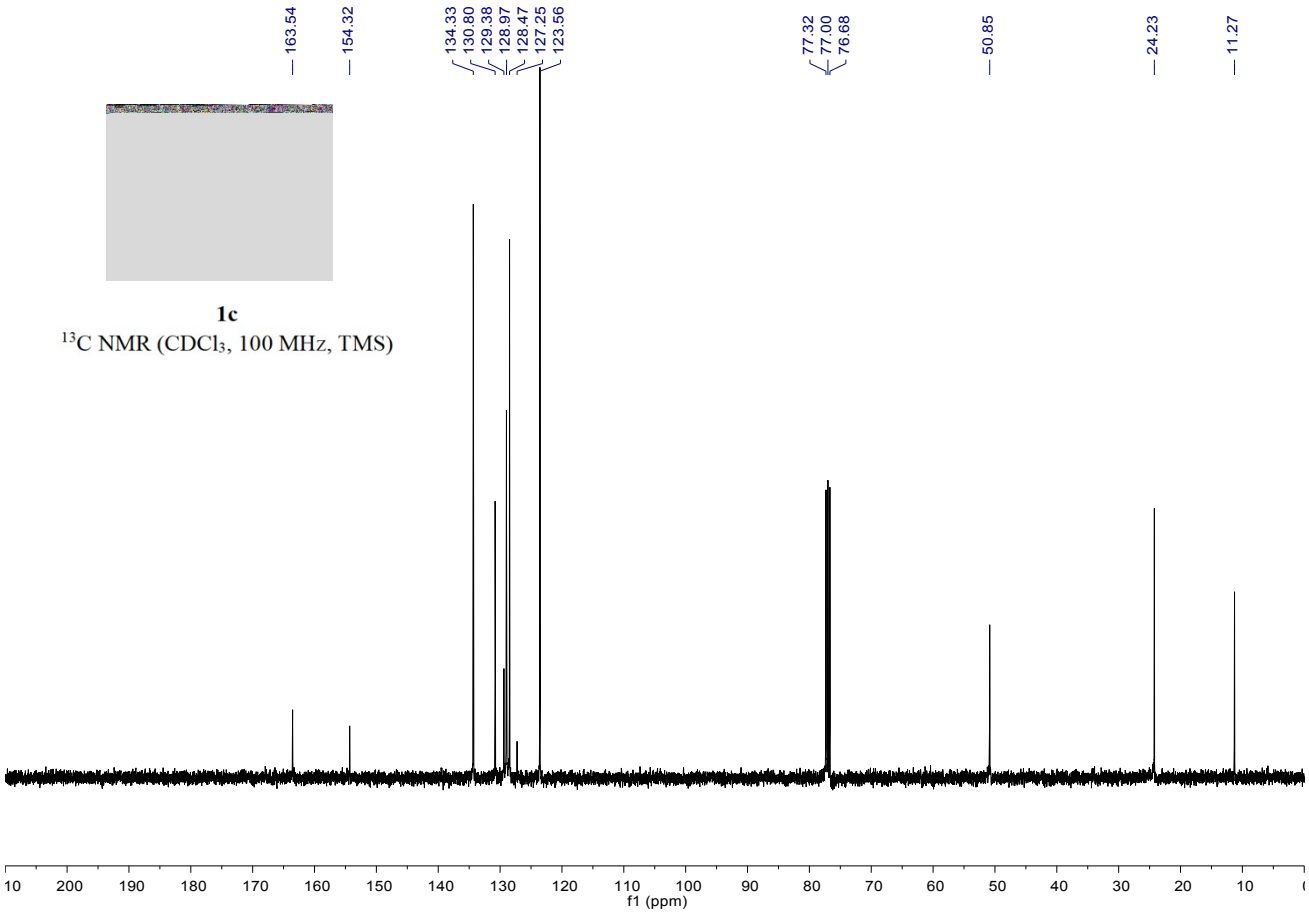
1b

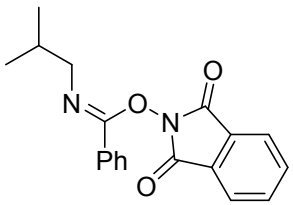
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



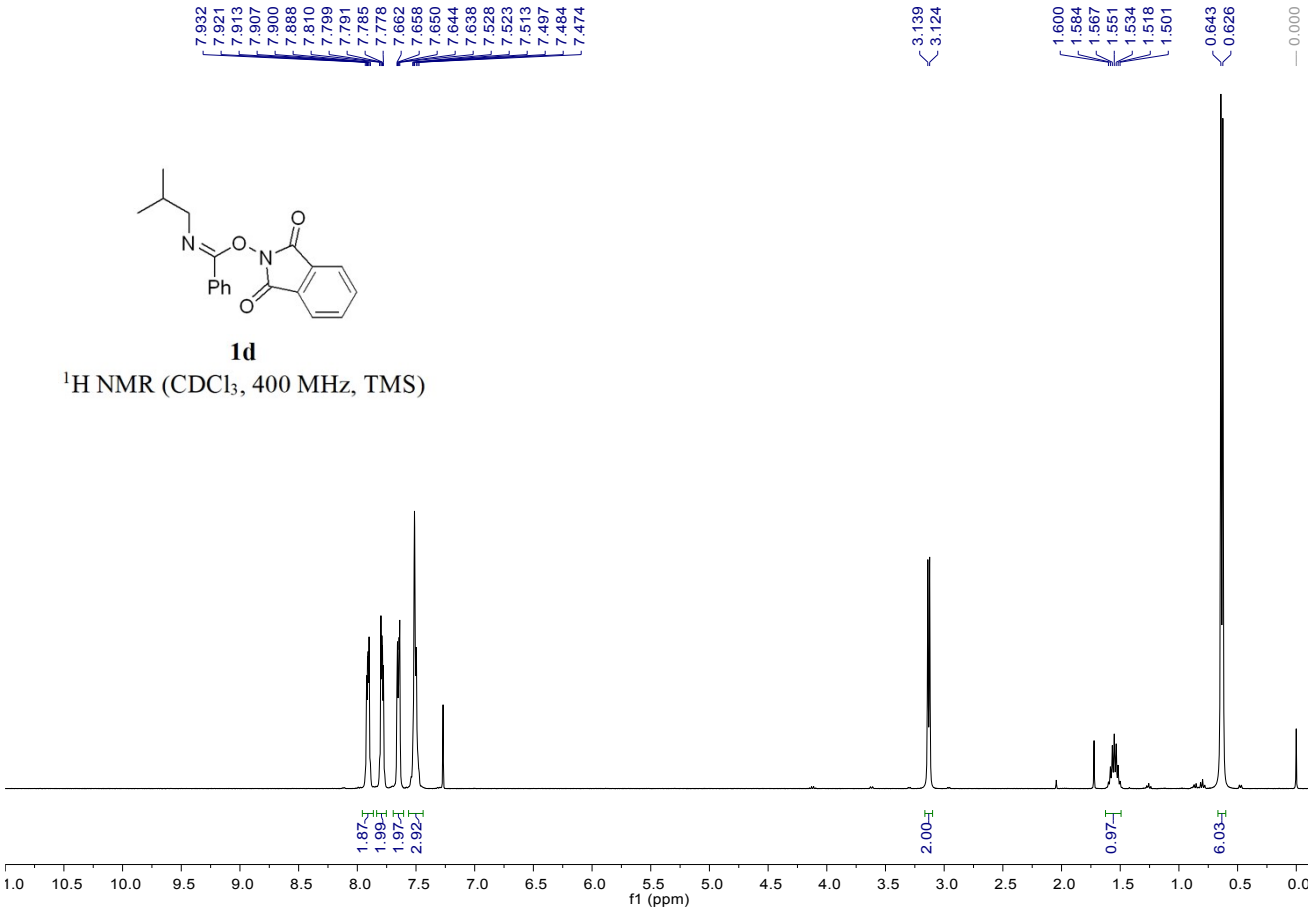
1,3-dioxoisoindolin-2-yl (Z)-N-propylbenzimidate (1c). The title product was obtained after purification by column chromatography (PE:EA = 4:1). A white solid, 715 mg, 51% yield; M.p.: 68-70 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.90 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.78 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.69 - 7.61 (m, 2H), 7.56 - 7.43 (m, 3H), 3.27 (t, *J* = 6.8 Hz, 2H), 1.40 - 1.27 (m, 2H), 0.66 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.5, 154.3, 134.3, 130.8, 129.4, 129.0, 128.5, 127.2, 123.6, 50.8, 24.2, 11.3; IR (neat) ν 2956, 2933, 1790, 1738, 1707, 1467, 1354, 1242, 1183, 1131, 1016, 879, 766, 696 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{17}N_2O_3^+$ Requires: 309.1234, Found: 309.1225.





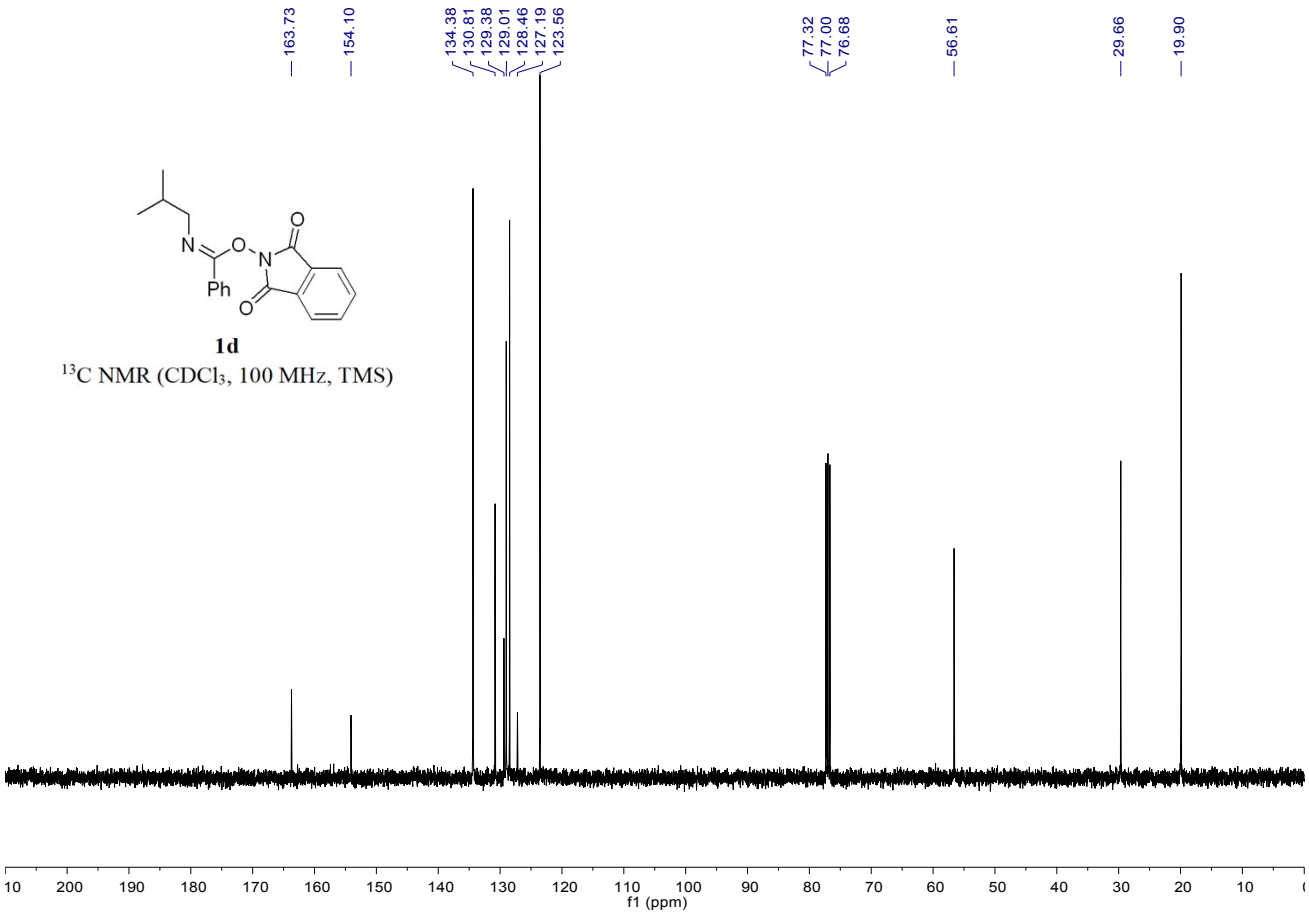


1,3-dioxoisoindolin-2-yl (*Z*)-N-isobutylbenzimidate (1d). The title product was obtained after purification by column chromatography (PE:EA = 4:1). A white solid, 1.29 g, 88% yield; M.p.: 100-102 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.96 - 7.86 (m, 2H), 7.84 - 7.75 (m, 2H), 7.69 - 7.61 (m, 2H), 7.56 - 7.44 (m, 3H), 3.13 (d, J = 6.0 Hz, 2H), 1.63 - 1.49 (m, 1H), 0.63 (d, J = 6.8 Hz, 6H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.7, 154.1, 134.4, 130.8, 129.4, 129.0, 128.5, 127.2, 123.6, 56.6, 29.7, 19.9; IR (neat) ν 2953, 2868, 1790, 1735, 1706, 1466, 1366, 1235, 1186, 1136, 1017, 879, 767, 694 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{19}N_2O_3^+$ Requires: 323.1390, Found: 323.1386.



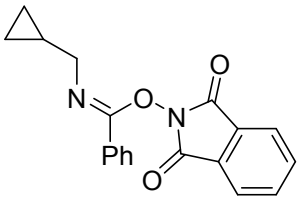
1d

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

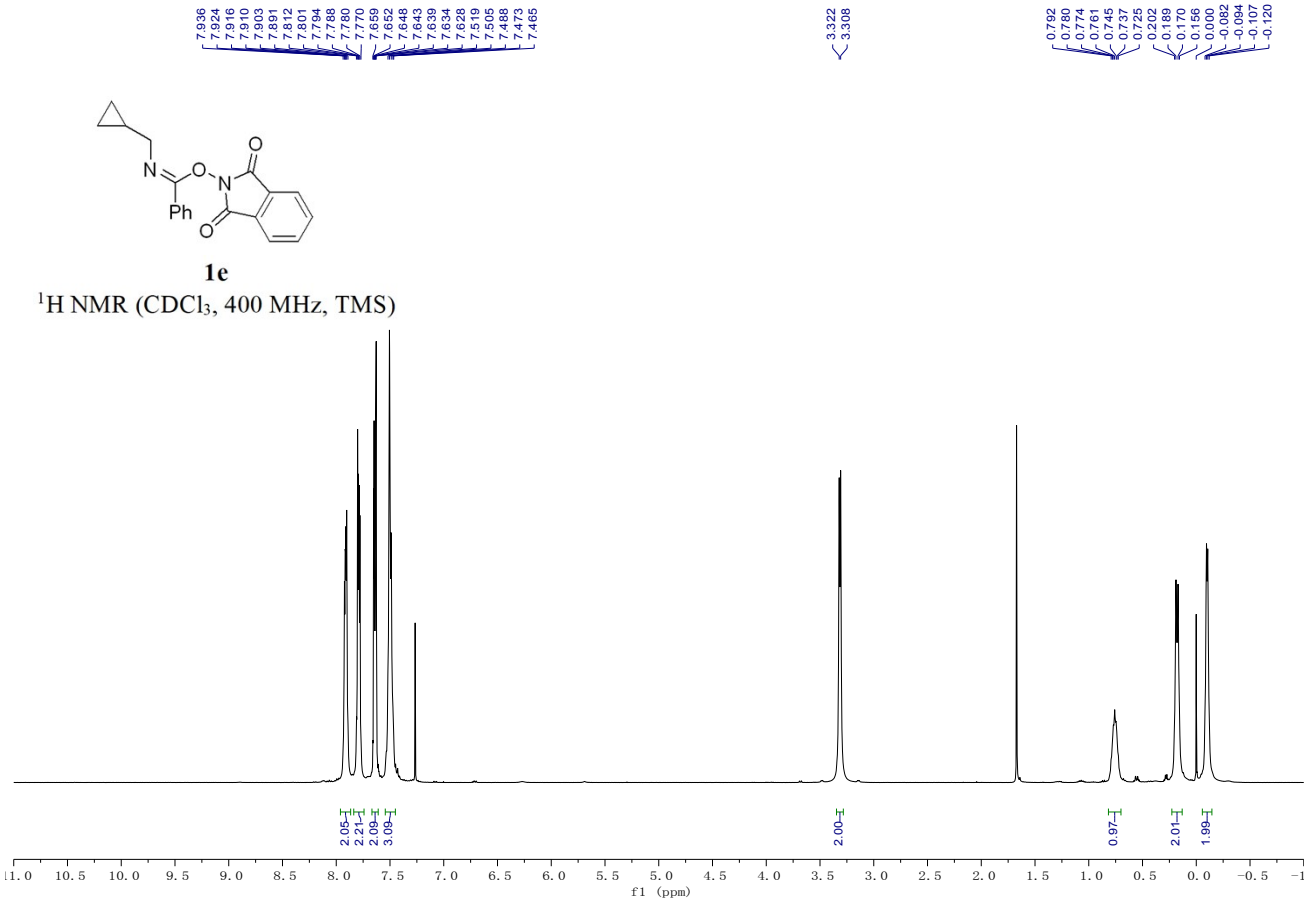


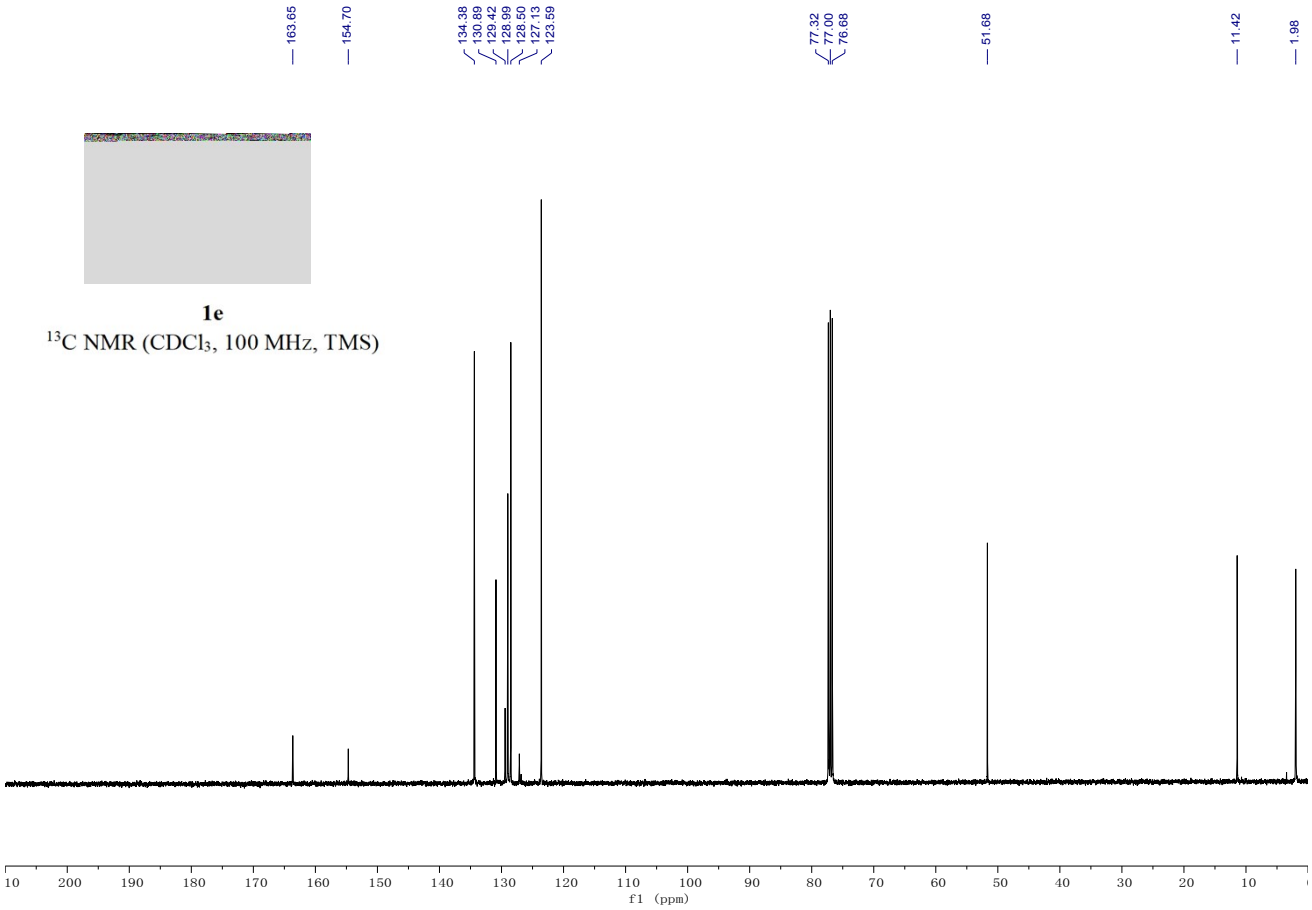
1d

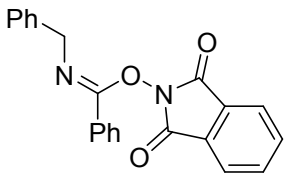
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



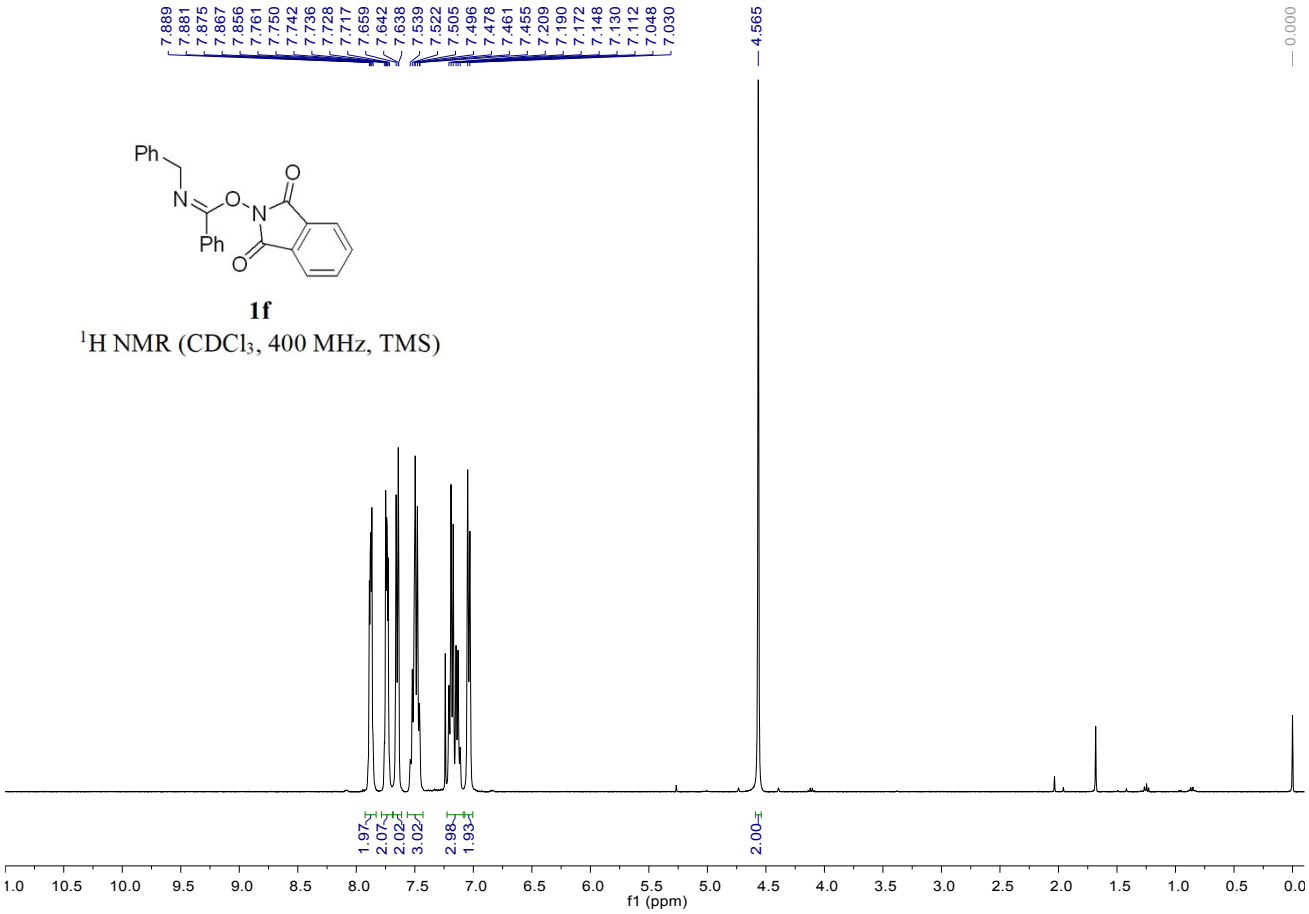
1,3-dioxoisoindolin-2-yl (*Z*)-N-(cyclopropylmethyl)benzimidate (1e). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.05 g, 72% yield; M.p.: 87-89 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.96 - 7.87 (m, 2H), 7.79 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.67 - 7.61 (m, 2H), 7.54 - 7.45 (m, 3H), 3.31 (d, *J* = 5.4 Hz, 2H), 0.82 - 0.70 (m, 1H), 0.23 - 0.13 (m, 2H), -0.06 - -0.15 (m, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.6, 154.7, 134.4, 130.9, 129.4, 129.0, 128.5, 127.1, 123.6, 51.7, 11.4, 2.0; IR (neat) ν 3070, 3001, 2898, 2852, 1797, 1742, 1711, 1447, 1372, 1239, 1186, 1136, 1017, 1006, 878, 771, 690 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{17}N_2O_3^+$ Requires: 321.1234, Found: 321.1226.





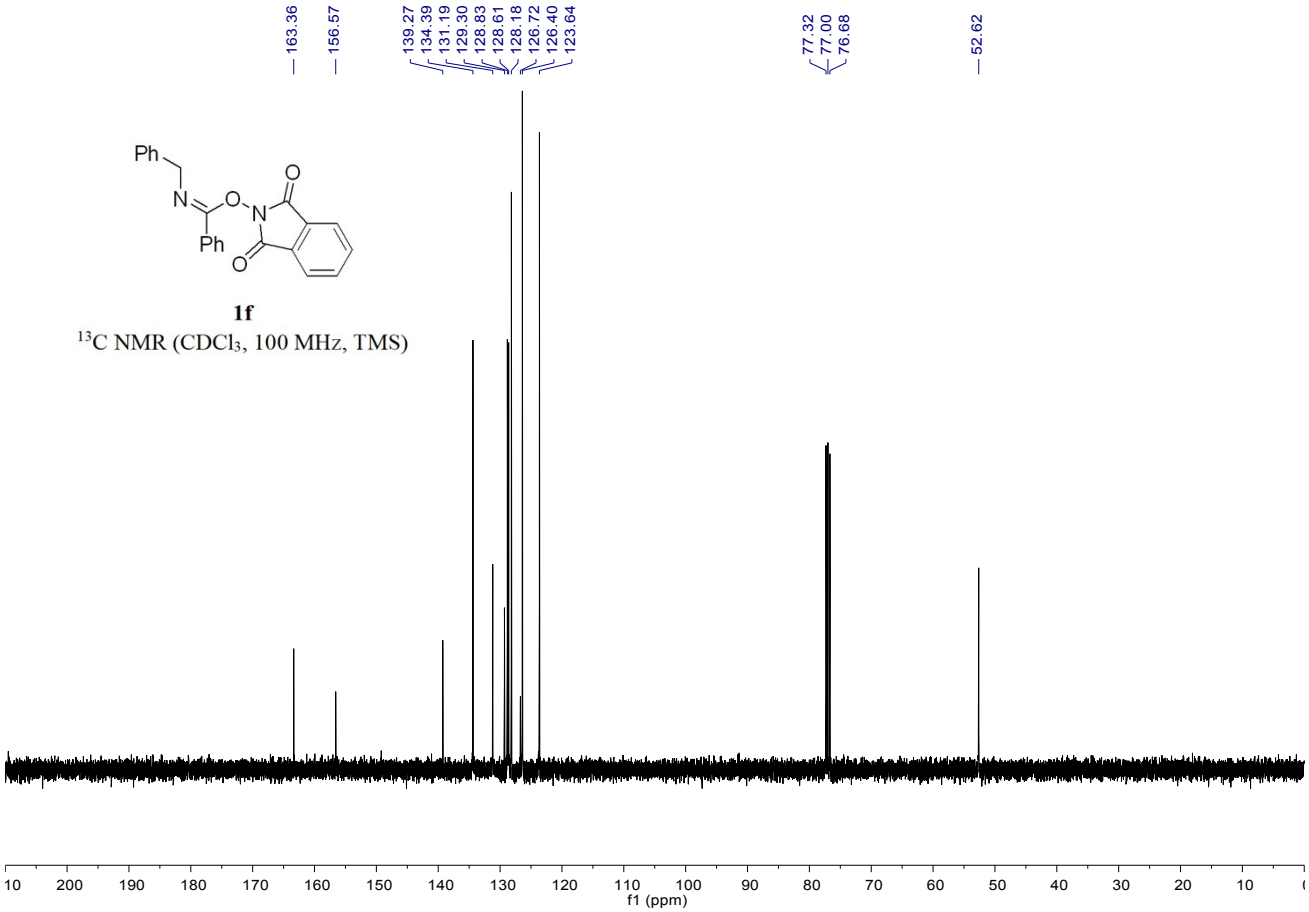


1,3-dioxoisoindolin-2-yl (Z)-N-benzylbenzimidate (1f). The title product was obtained after purification by column chromatography (PE:EA = 2:1). A white solid, 826 mg, 51% yield; M.p.: 100-102 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.92 - 7.83 (m, 2H), 7.74 (dd, J = 5.6, 3.2 Hz, 2H), 7.68 - 7.61 (m, 2H), 7.56 - 7.43 (m, 3H), 7.22 - 7.09 (m, 3H), 7.07 - 7.00 (m, 2H), 4.57 (s, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.4, 156.6, 139.3, 134.4, 131.2, 129.3, 128.8, 128.6, 128.2, 126.7, 126.4, 123.6, 52.6; IR (neat) ν 1798, 1737, 1706, 1455, 1352, 1244, 1184, 1042, 981, 876, 735, 694 cm^{-1}; HRMS (ESI) Calcd. for $C_{22}H_{17}N_2O_3^+$ Requires: 357.1234, Found: 357.1228.



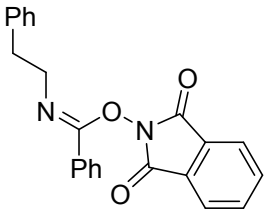
1f

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

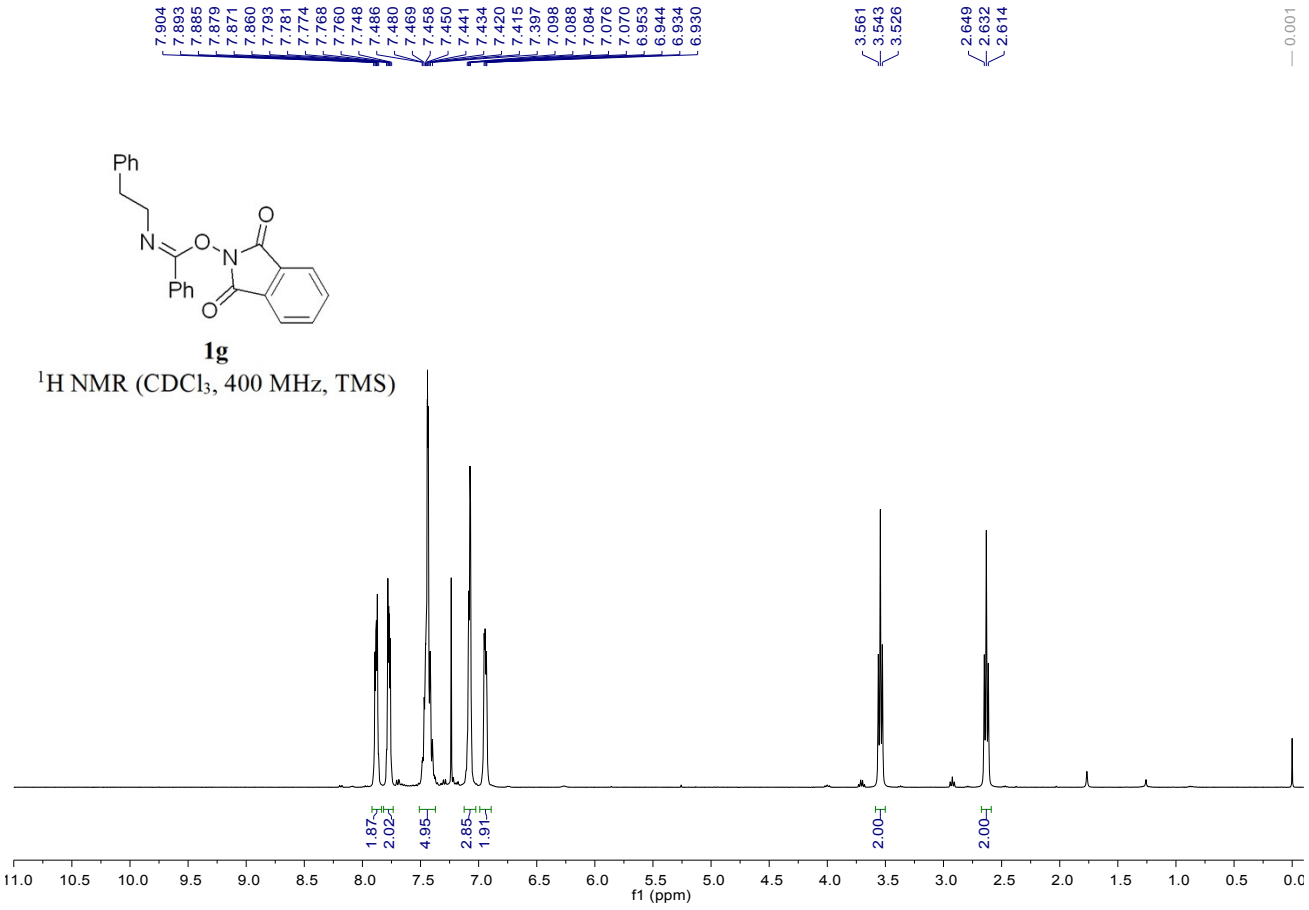


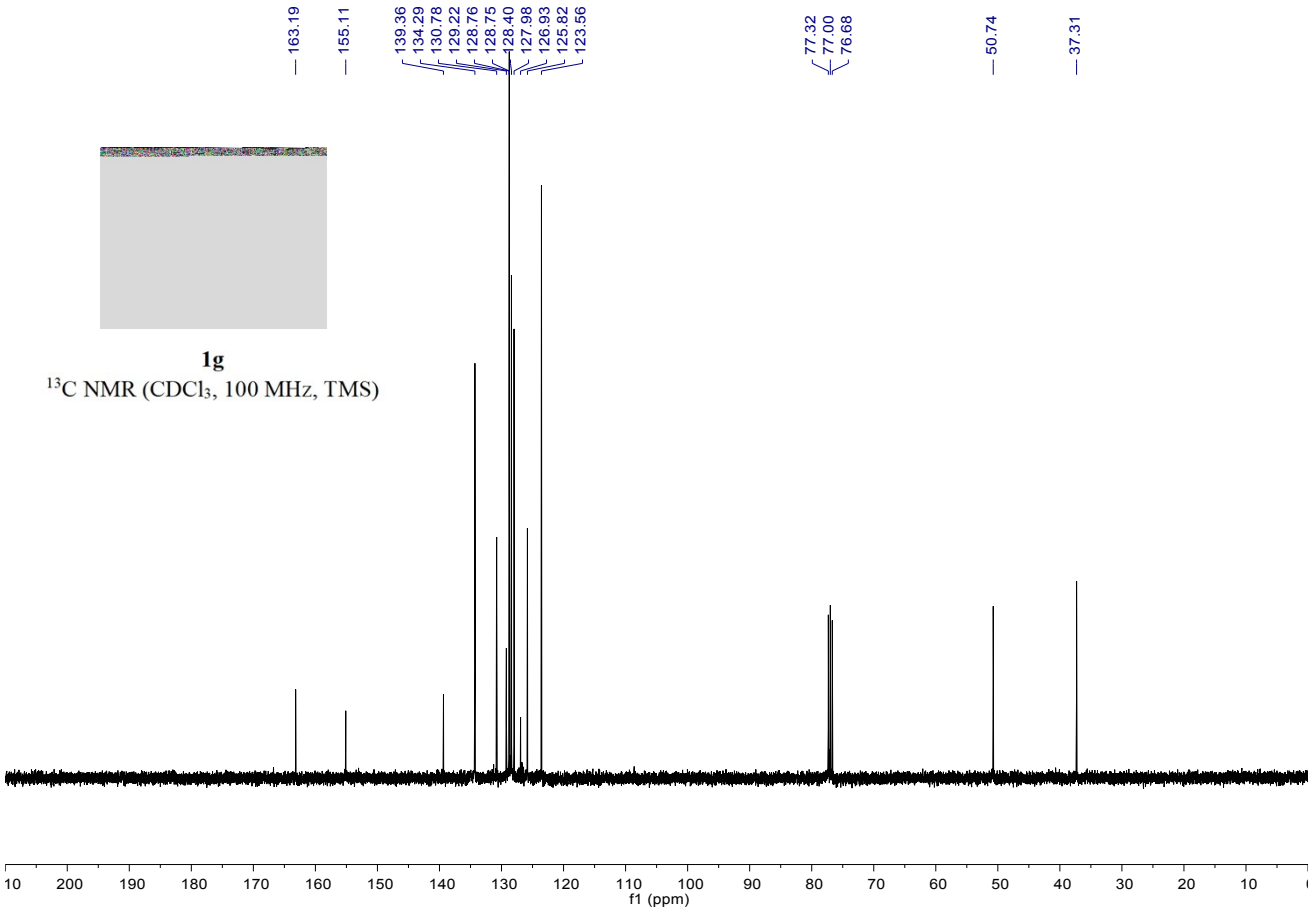
1f

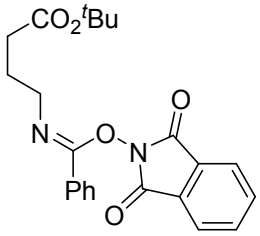
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



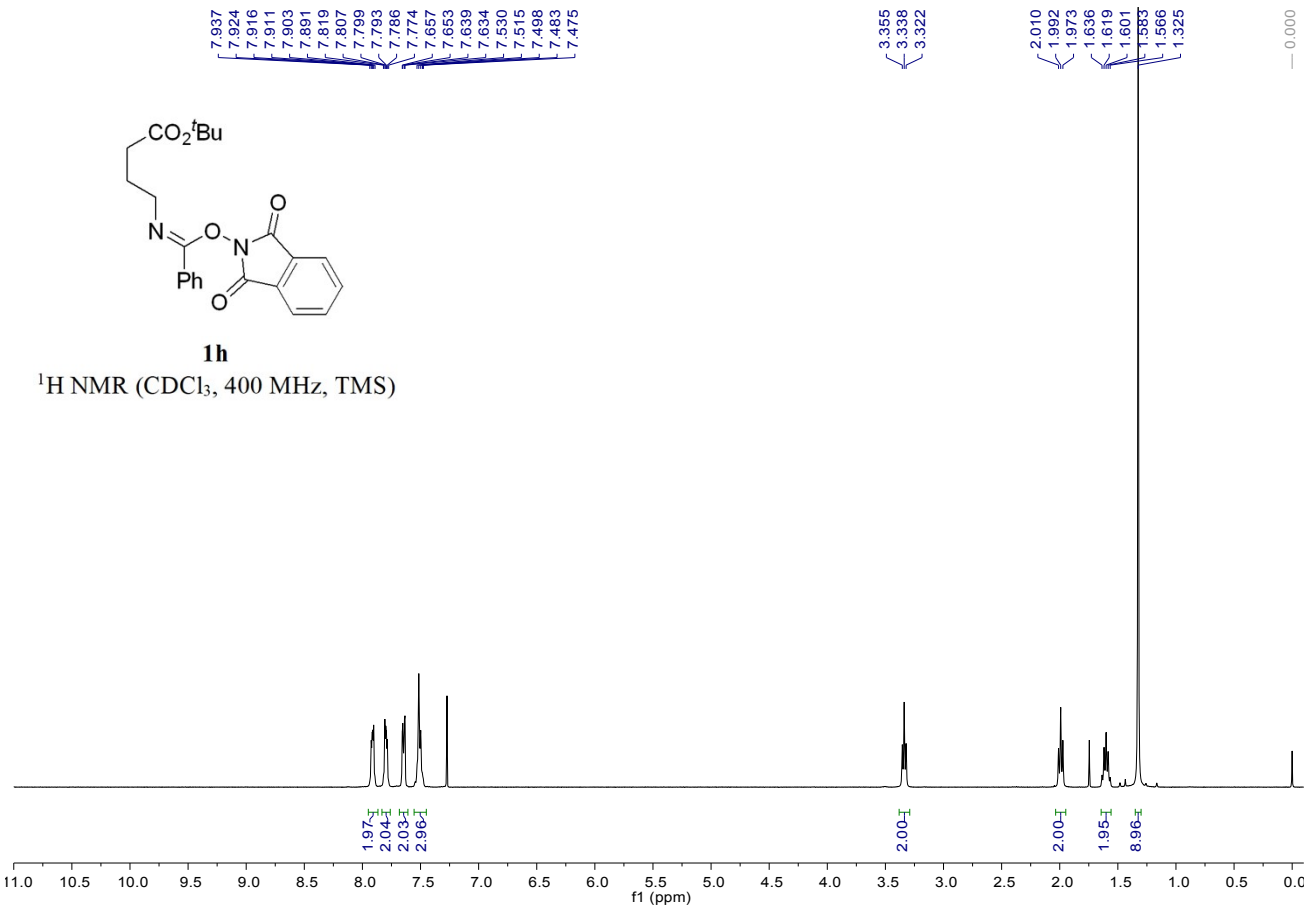
1,3-dioxoisoindolin-2-yl (*Z*)-N-phenethylbenzimidate (1g). The title product was obtained after purification by column chromatography (DCM). A white solid, 875 mg, 52% yield; M.p.: 120-121 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.92 - 7.83 (m, 2H), 7.82 - 7.74 (m, 2H), 7.51 - 7.37 (m, 5H), 7.12 - 7.02 (m, 3H), 6.99 - 6.89 (m, 2H), 3.54 (t, *J* = 6.9 Hz, 2H), 2.63 (t, *J* = 7.0 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 155.1, 139.4, 134.3, 130.8, 129.2, 128.8, 128.7, 128.4, 128.0, 126.9, 125.8, 123.6, 50.7, 37.3; IR (neat) ν 2932, 2878, 1790, 1737, 1698, 1446, 1375, 1235, 1186, 1135, 1043, 1018, 878, 772, 690 cm^{-1}; HRMS (ESI) Calcd. for $C_{23}H_{19}N_2O_3^+$ Requires: 371.1390, Found: 371.1393.





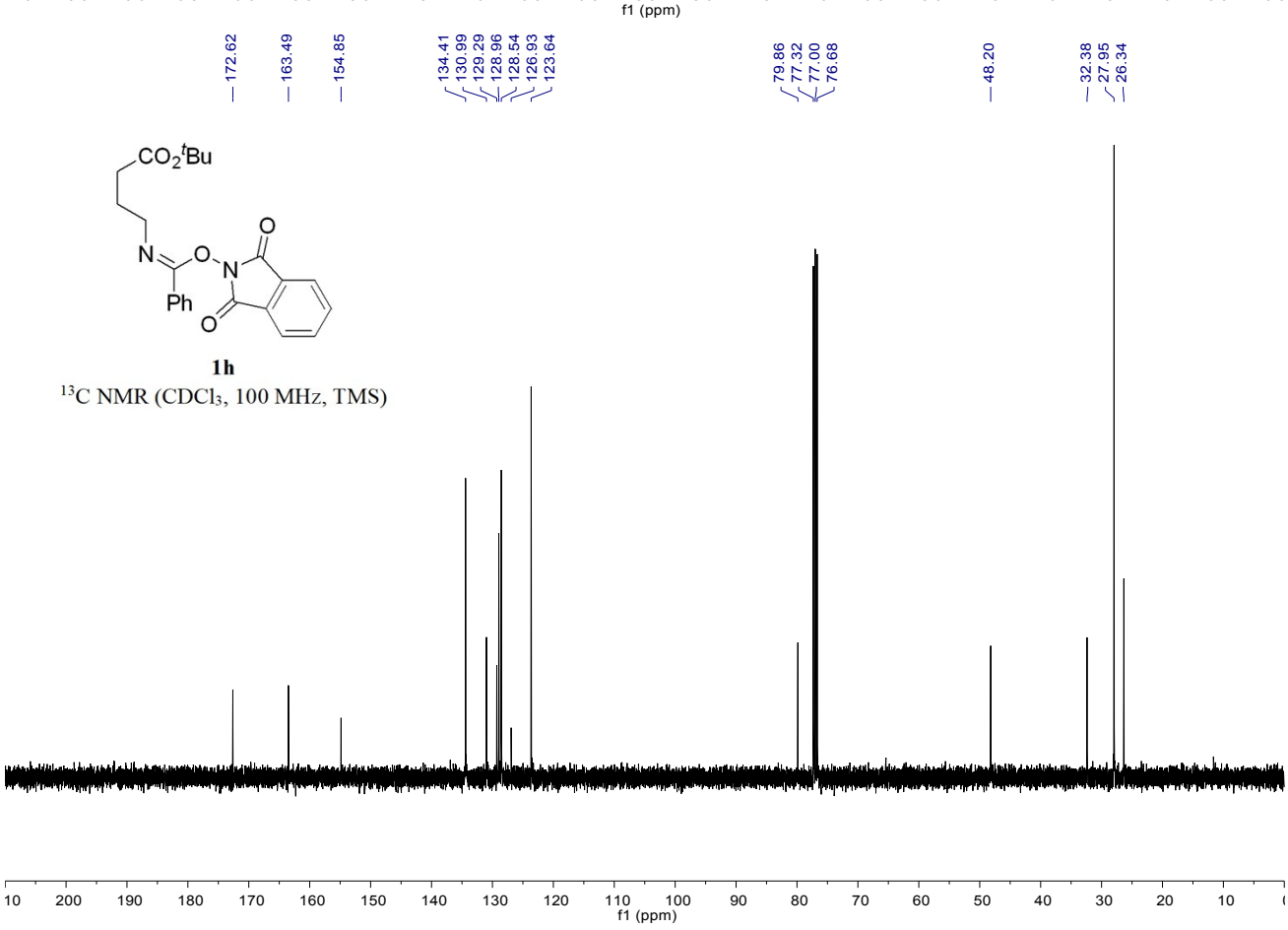


***tert*-butyl (*Z*)-4-((((1,3-dioxoisoindolin-2-yl)oxy)(phenyl)methylene)amino)butanoate (1h)**. The title product was obtained after purification by column chromatography (DCM). A white solid, 1.77 g, 95% yield; M.p.: 78-80 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.95 - 7.87 (m, 2H), 7.83 - 7.76 (m, 2H), 7.68 - 7.61 (m, 2H), 7.56 - 7.45 (m, 3H), 3.34 (t, *J* = 6.6 Hz, 2H), 1.99 (t, *J* = 7.5 Hz, 2H), 1.64 - 1.56 (m, 2H), 1.33 (s, 9H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 172.6, 163.5, 154.9, 134.4, 131.0, 129.3, 129.0, 128.5, 126.9, 123.6, 79.9, 48.2, 32.4, 27.9, 26.3; IR (neat) ν 2964, 2938, 1794, 1741, 1714, 1364, 1243, 1183, 1148, 1005, 879, 779, 697 cm^{-1}; HRMS (ESI) Calcd. for $C_{23}H_{24}N_2O_5Na^+$ Requires: 431.1577, Found: 431.1581.



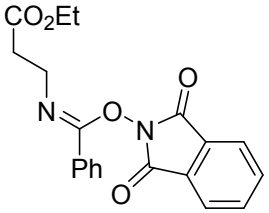
1h

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

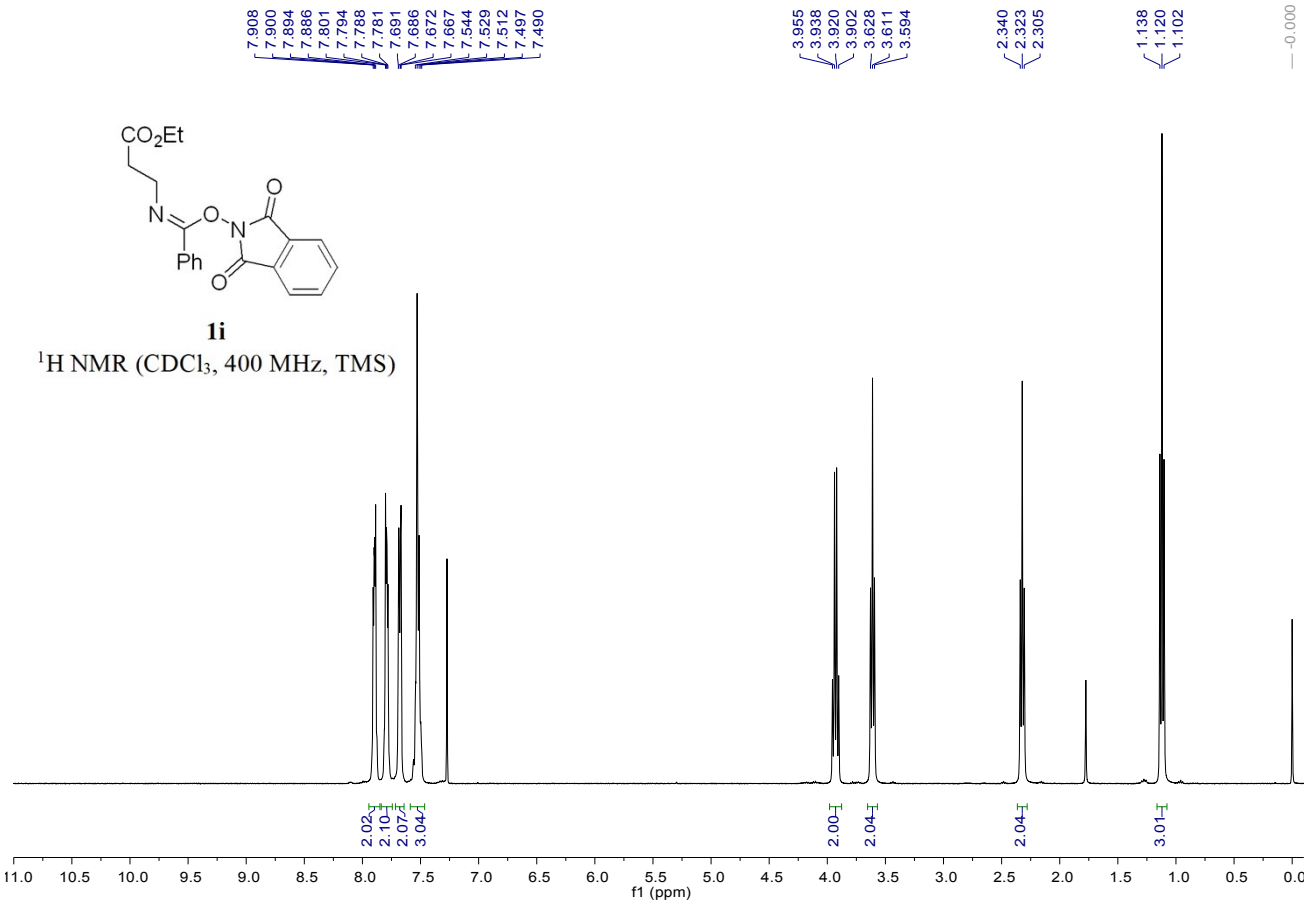


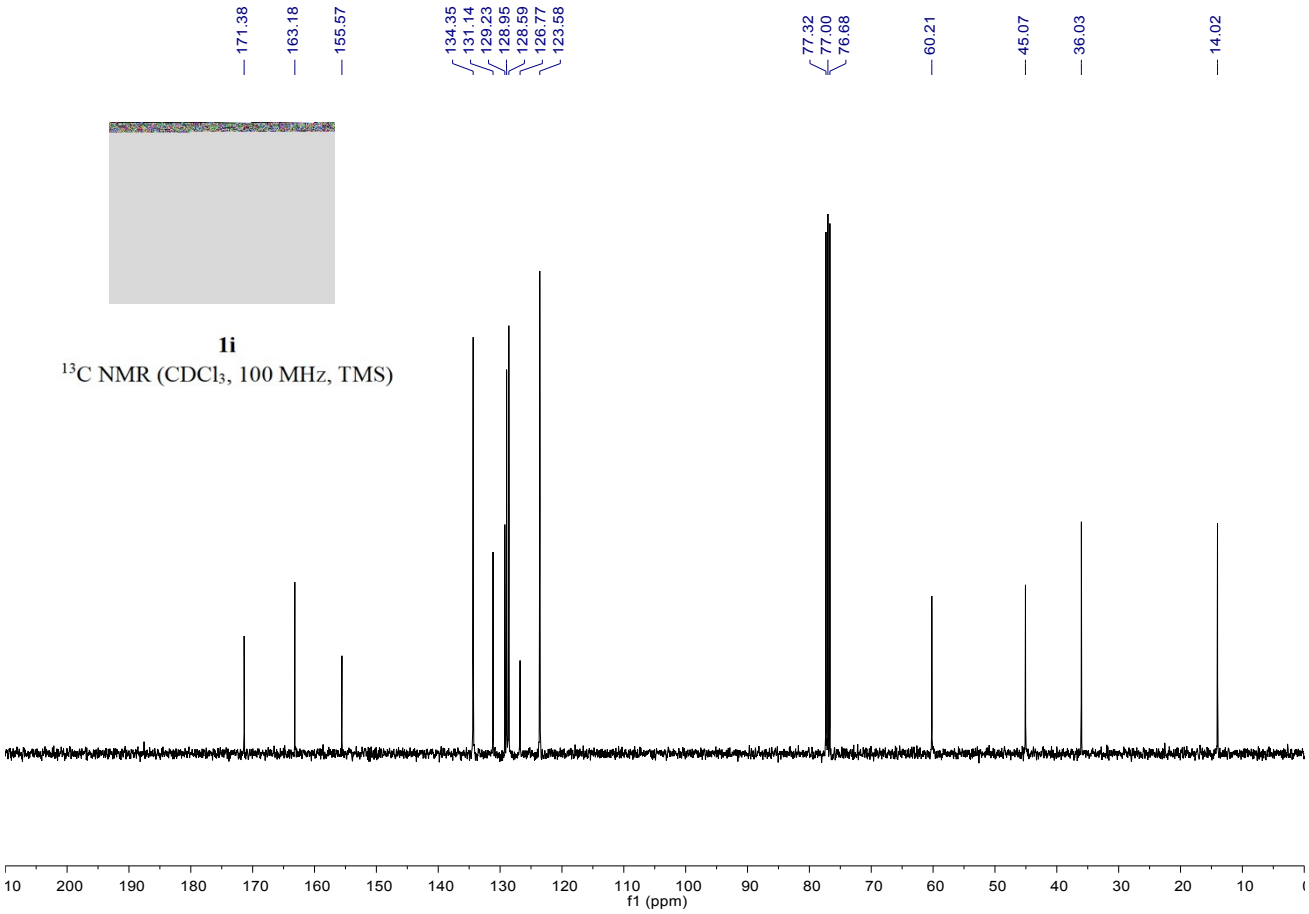
1h

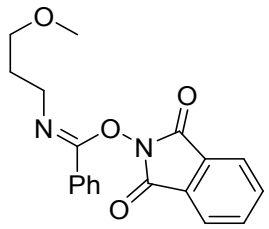
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



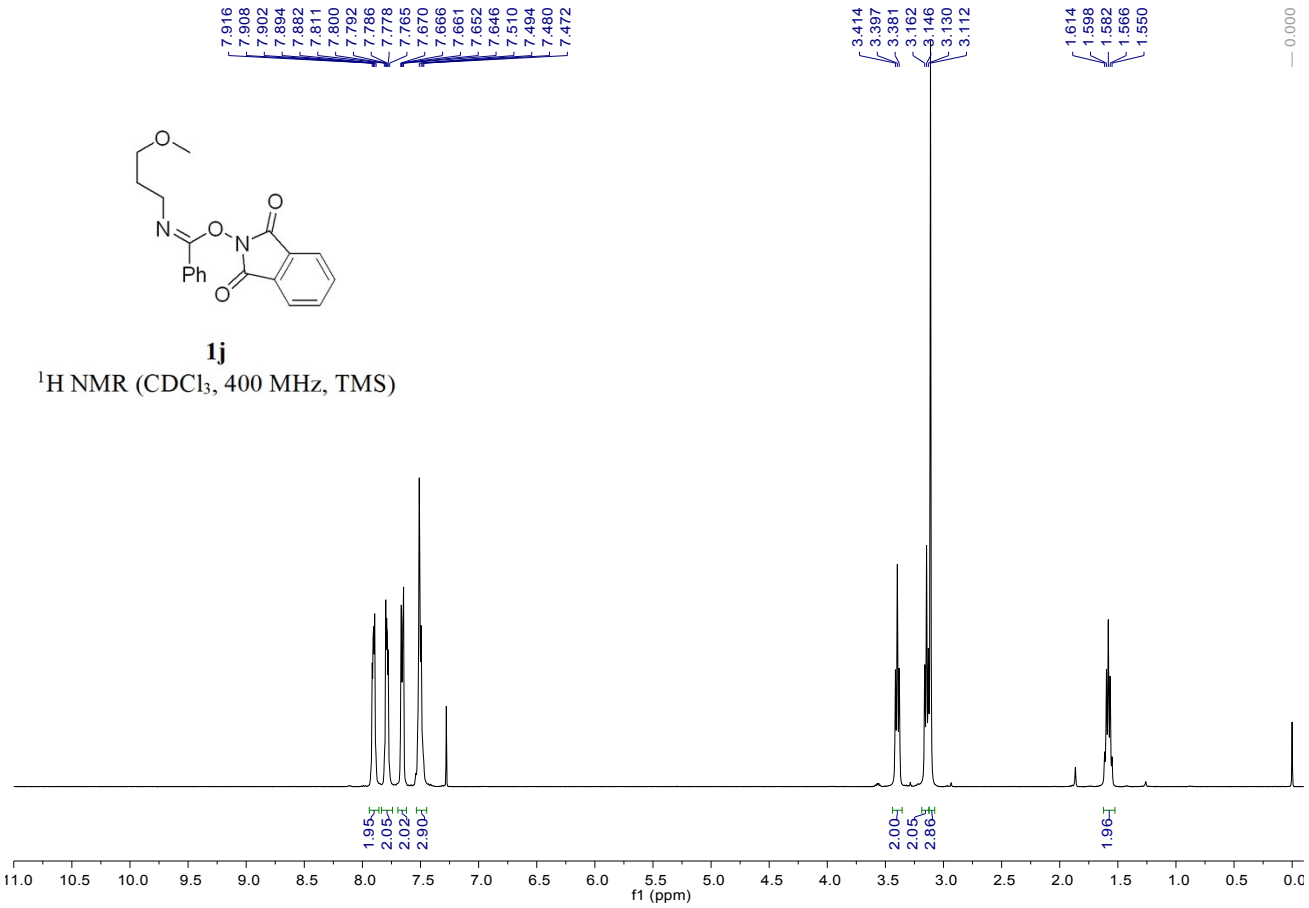
ethyl (*Z*)-3-((((1,3-dioxoisoindolin-2-yl)oxy)(phenyl)methylene)amino)propanoate (1i). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.48 g, 89% yield; M.p.: 76-78 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.90 (dd, *J* = 5.6, 3.2 Hz, 2H), 7.79 (dd, *J* = 5.2, 2.8 Hz, 2H), 7.72 - 7.64 (m, 2H), 7.59 - 7.46 (m, 3H), 3.93 (q, *J* = 7.1 Hz, 2H), 3.61 (t, *J* = 6.9 Hz, 2H), 2.32 (t, *J* = 6.9 Hz, 2H), 1.12 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.4, 163.2, 155.6, 134.4, 131.1, 129.2, 128.9, 128.6, 126.8, 123.6, 60.2, 45.1, 36.0, 14.0; IR (neat) ν 1791, 1743, 1706, 1447, 1370, 1221, 1182, 1020, 983, 879, 774, 692 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{18}N_2O_5Na^+$ Requires: 389.1108, Found: 389.1106.





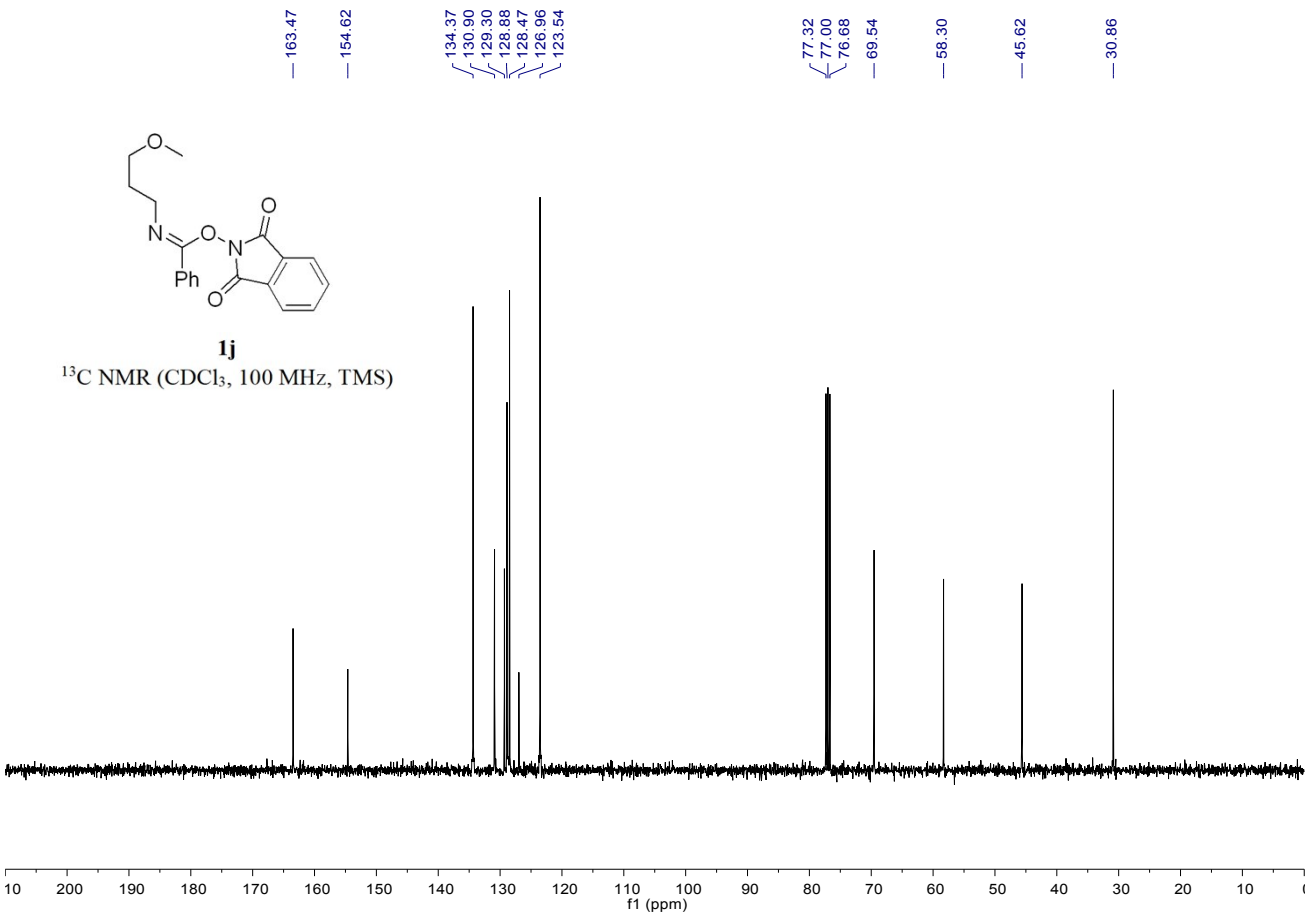


1,3-dioxoisoindolin-2-yl (*Z*)-N-(3-methoxypropyl)benzimidate (1j). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.25 g, 81% yield; M.p.: 84-86 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.94 - 7.86 (m, 2H), 7.84 - 7.74 (m, 2H),, 7.69 - 7.62 (m, 2H), 7.54 - 7.45 (m, 3H), 3.40 (t, J = 6.4 Hz, 2H), 3.15 (t, J = 6.4 Hz, 2H), 3.11 (s, 3H), 1.58 (qu, J = 6.4 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.5, 154.6, 134.4, 130.9, 129.3, 128.9, 128.5, 127.0, 123.5, 69.5, 58.3, 45.6, 30.9; IR (neat) ν 2889, 2860, 1797, 1735, 1698, 1448, 1353, 1241, 1186, 1118, 1005, 878, 778, 695 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{19}N_2O_4^+$ Requires: 339.1339, Found: 339.1338.



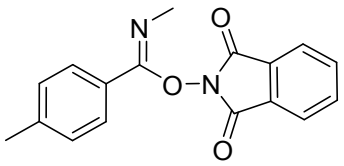
1j

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

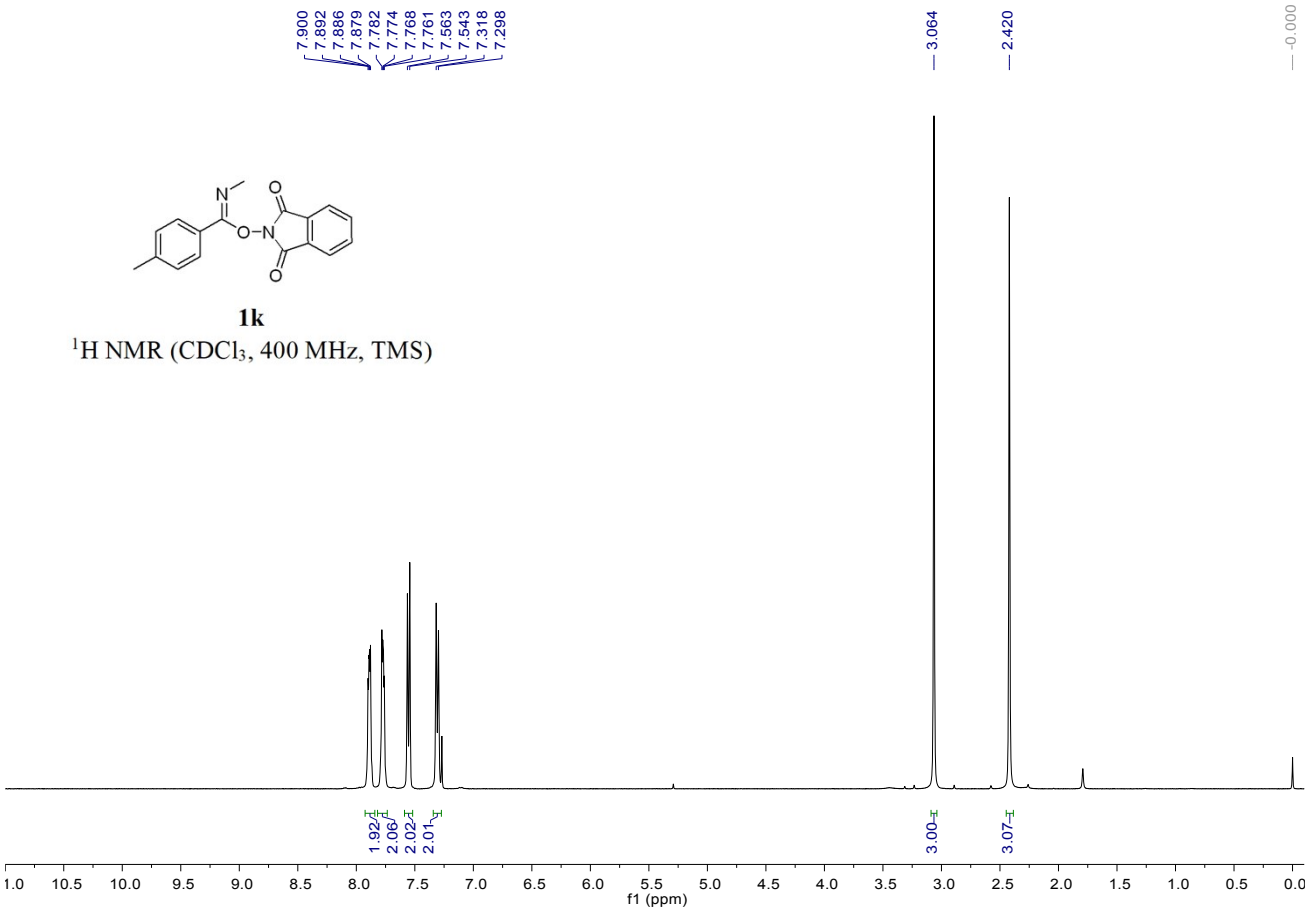


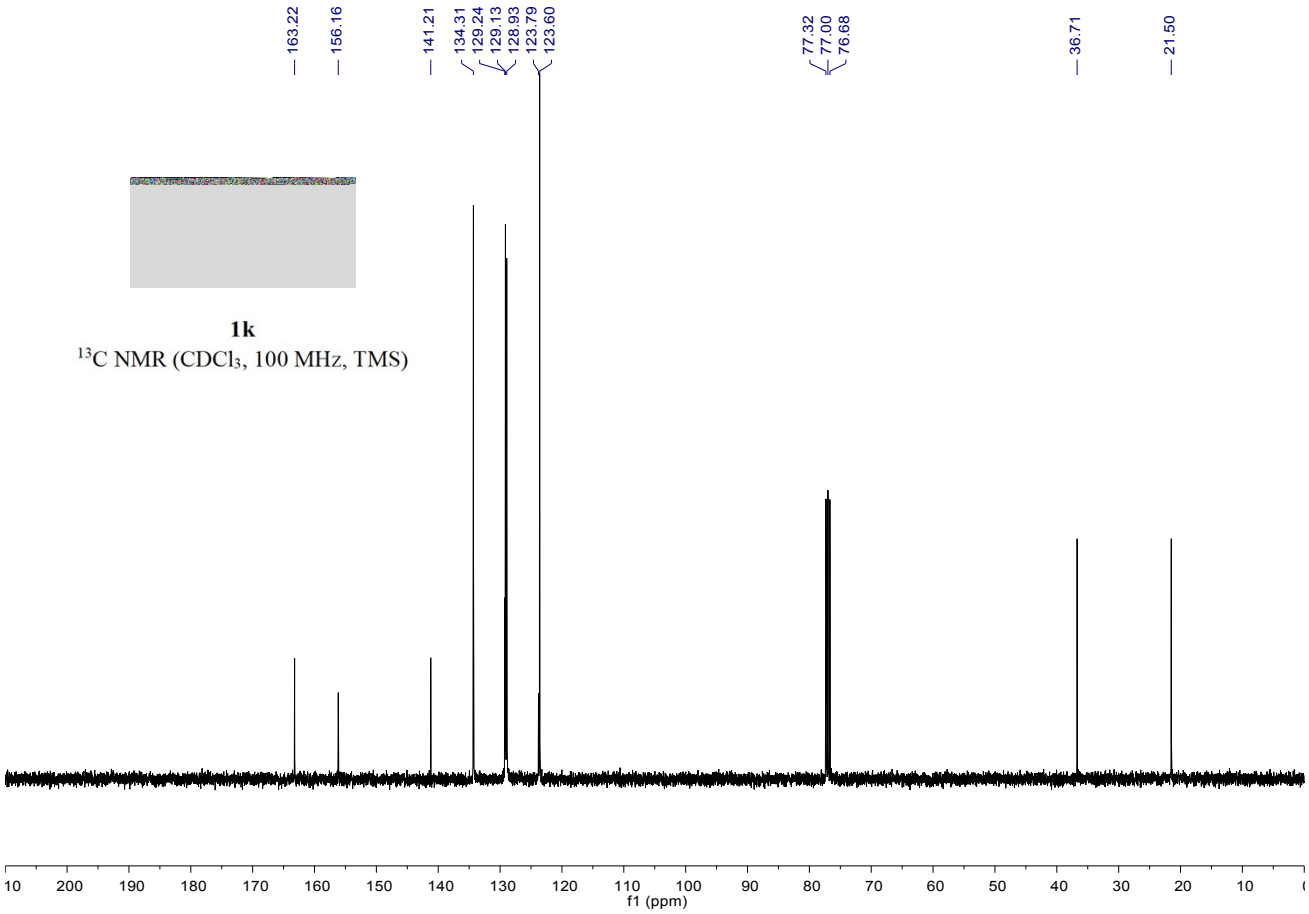
1j

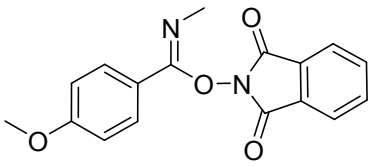
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



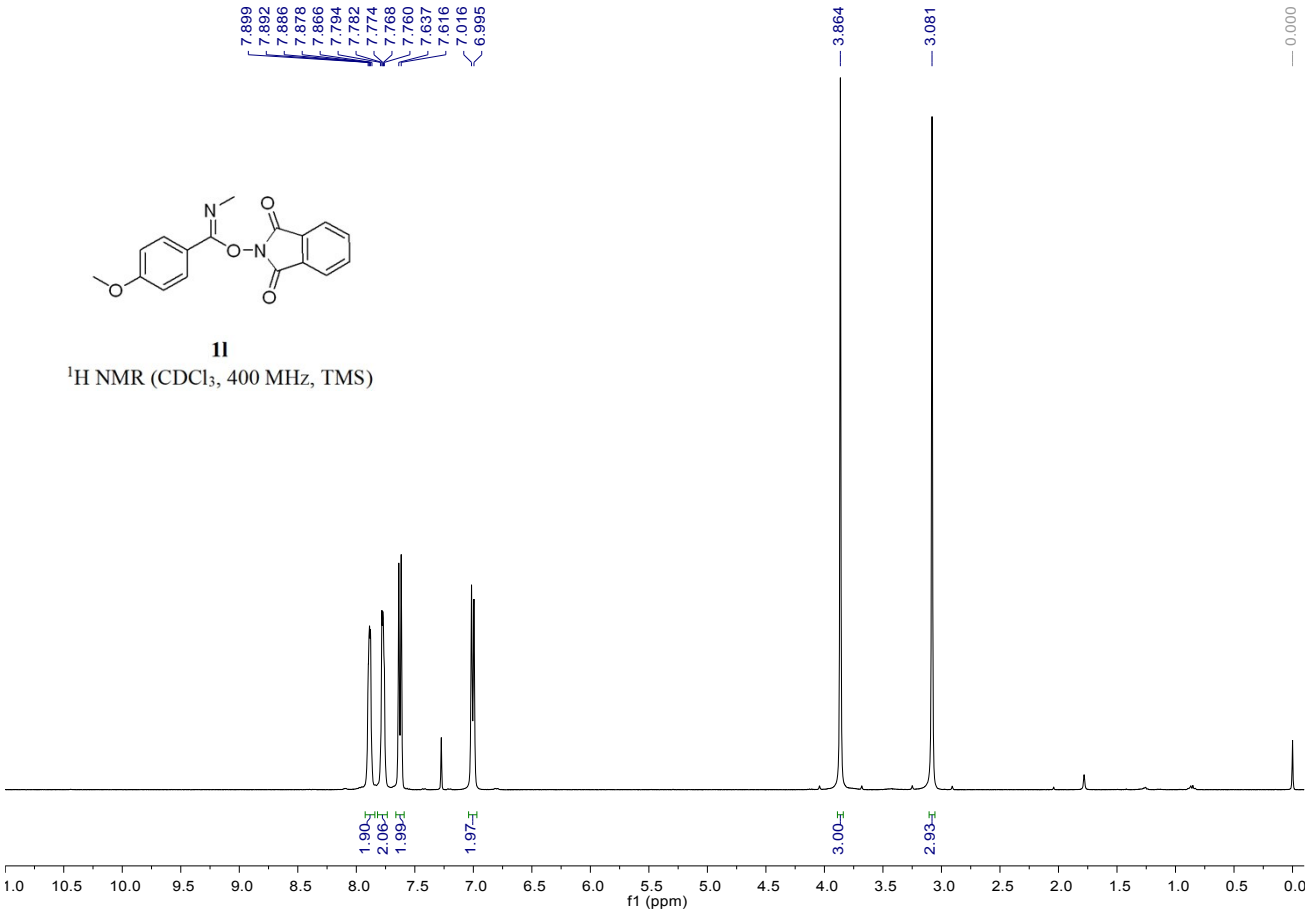
1,3-dioxoisoindolin-2-yl (*Z*)-N,4-dimethylbenzimidate (1k). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.18 g, 82% yield; M.p.: 188-190 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.89 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.77 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.55 (d, *J* = 7.9 Hz, 2H), 7.31 (d, *J* = 7.8 Hz, 2H), 3.06 (s, 3H), 2.42 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 156.2, 141.2, 134.3, 129.2, 129.1, 128.9, 123.8, 123.6, 36.7, 21.5; IR (neat) ν 1791, 1732, 1711, 1466, 1243, 1184, 1133, 1004, 877, 826, 688 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2O_3^+$ Requires: 295.1077, Found: 295.1070.





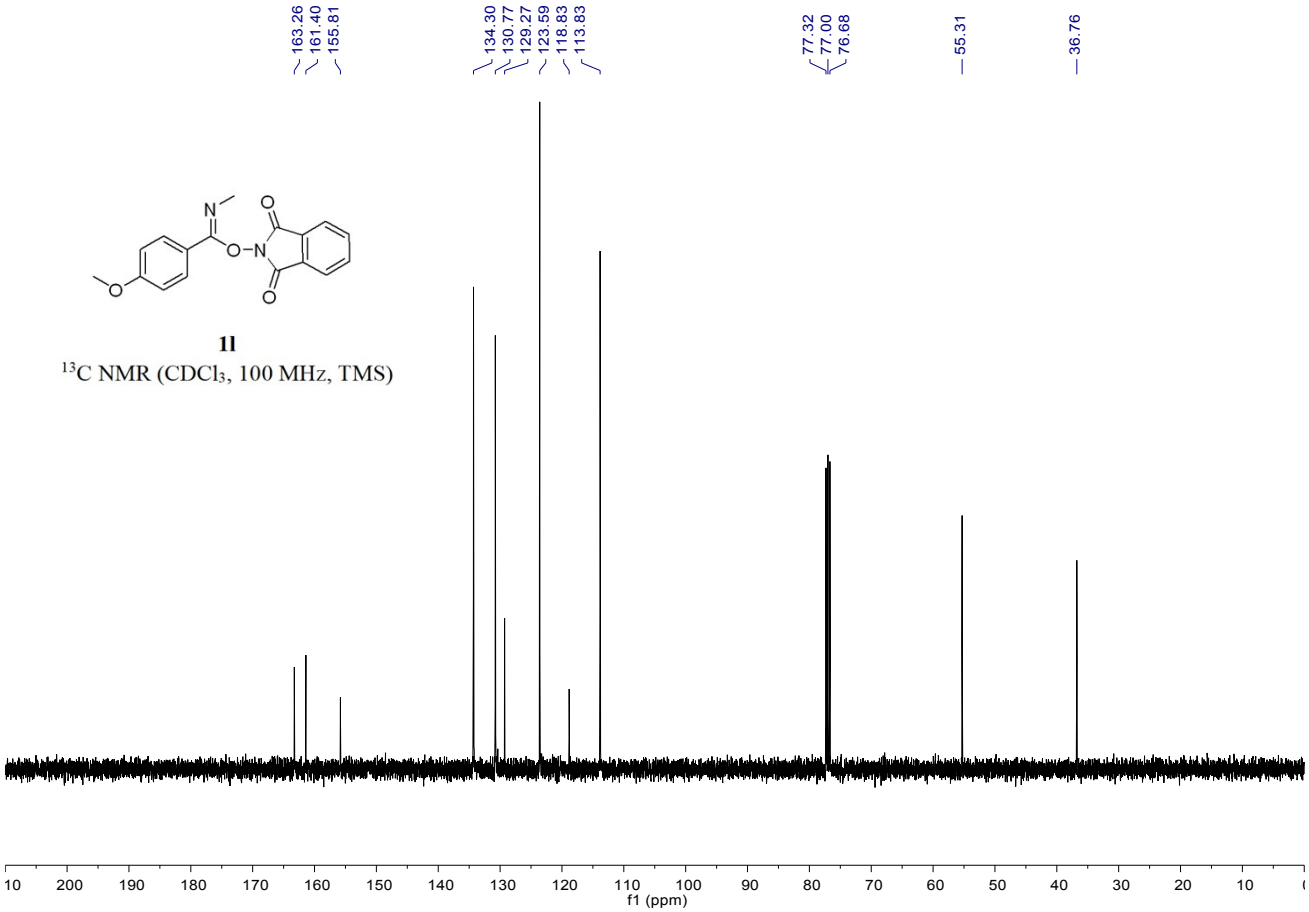


1,3-dioxoisoindolin-2-yl (*Z*)-4-methoxy-N-methylbenzimidate (1l). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.1 g, 80% yield; M.p.: 148-150 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.89 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.77 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.63 (d, *J* = 8.4 Hz, 2H), 7.01 (d, *J* = 8.3 Hz, 2H), 3.86 (s, 3H), 3.08 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.3, 161.4, 155.8, 134.3, 130.8, 129.3, 123.6, 118.8, 113.8, 55.3, 36.8; IR (neat) ν 3064, 2965, 1798, 1736, 1711, 1608, 1511, 1353, 1264, 1177, 1116, 1015, 997, 879, 841, 799, 696 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2O_4^+$ Requires: 311.1026, Found: 311.1025.



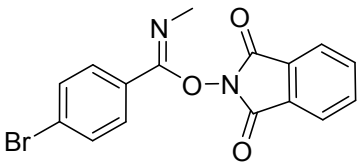
1l

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

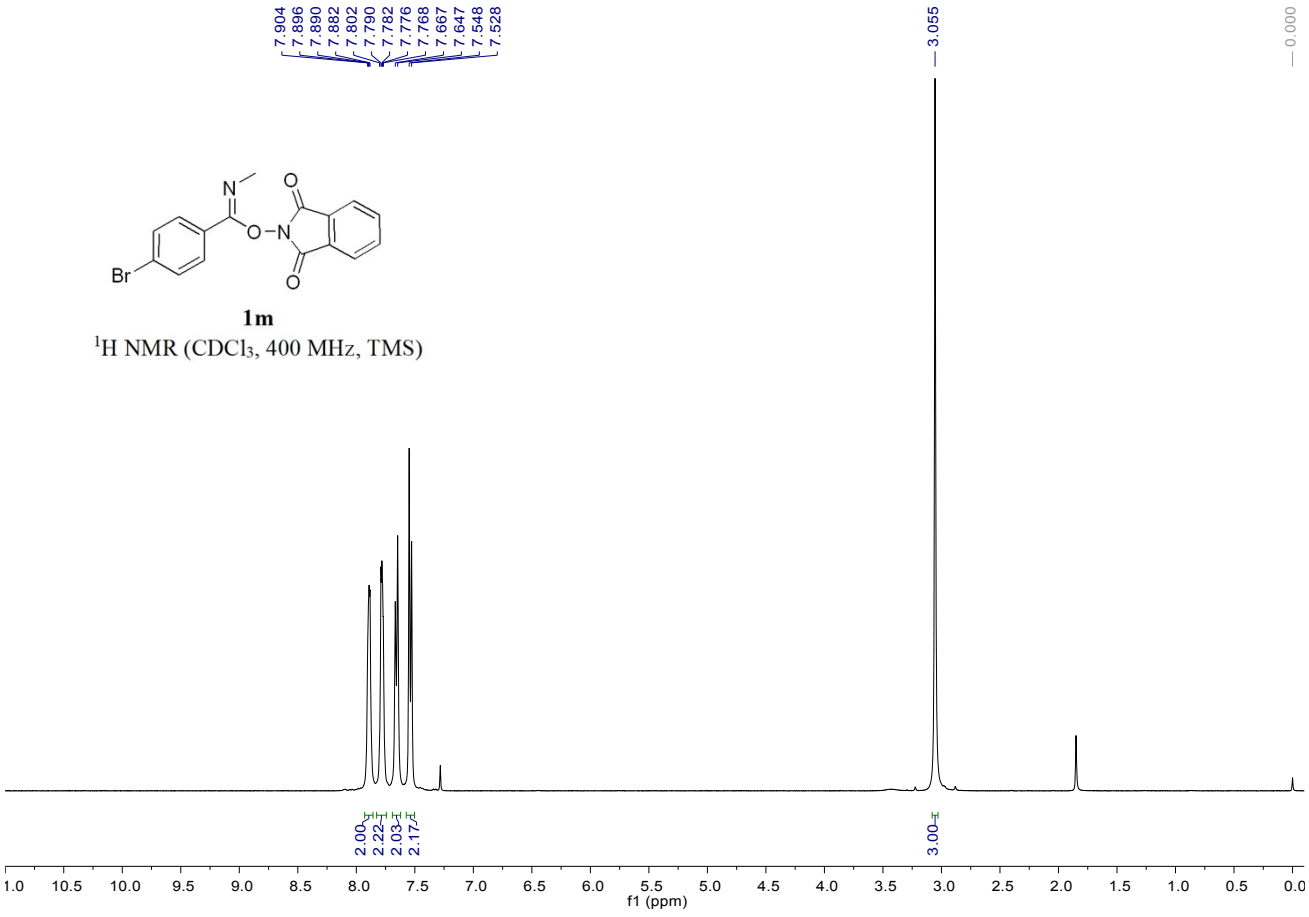


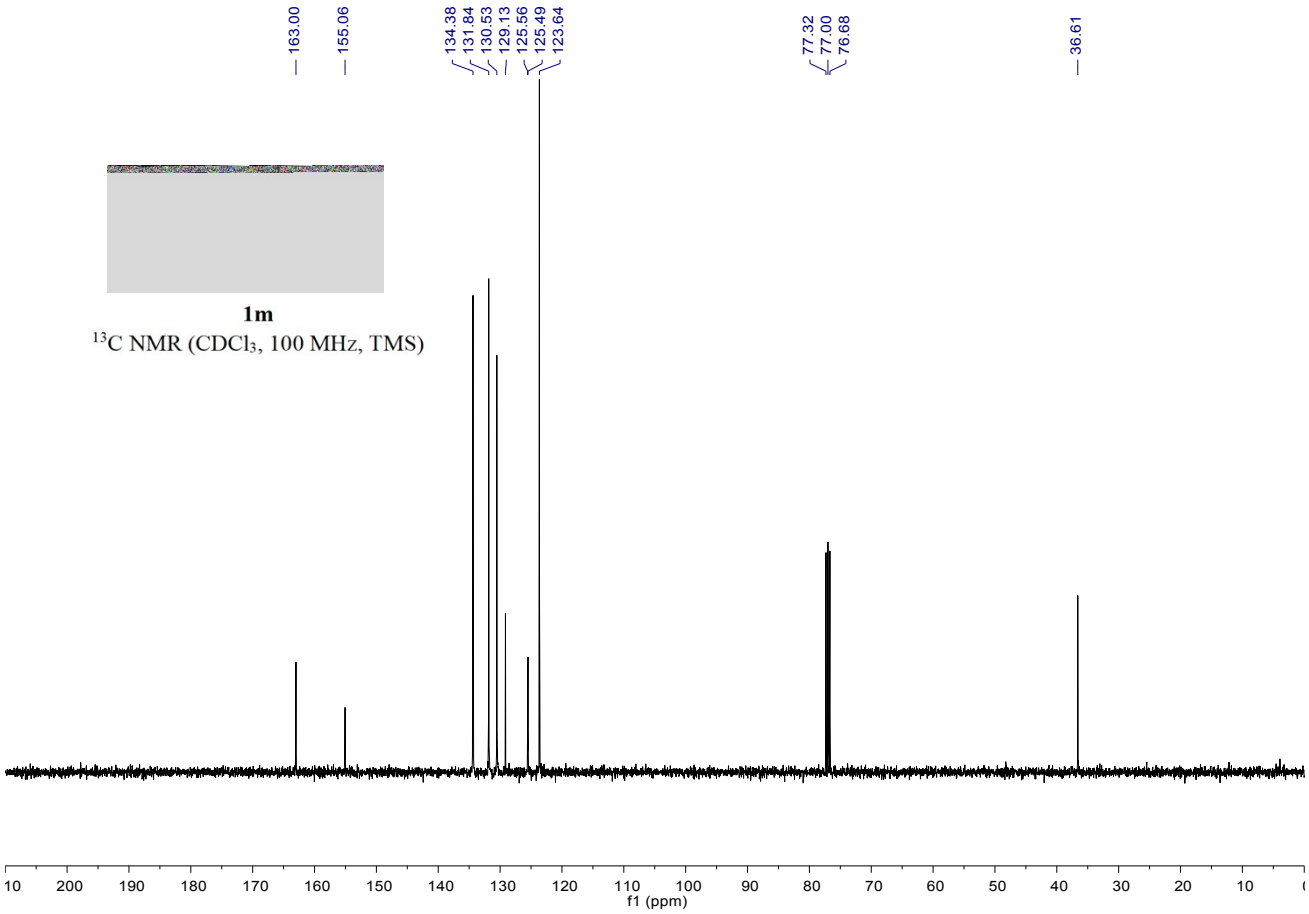
1l

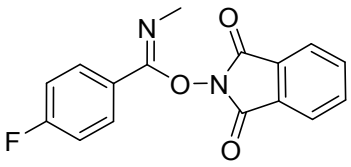
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



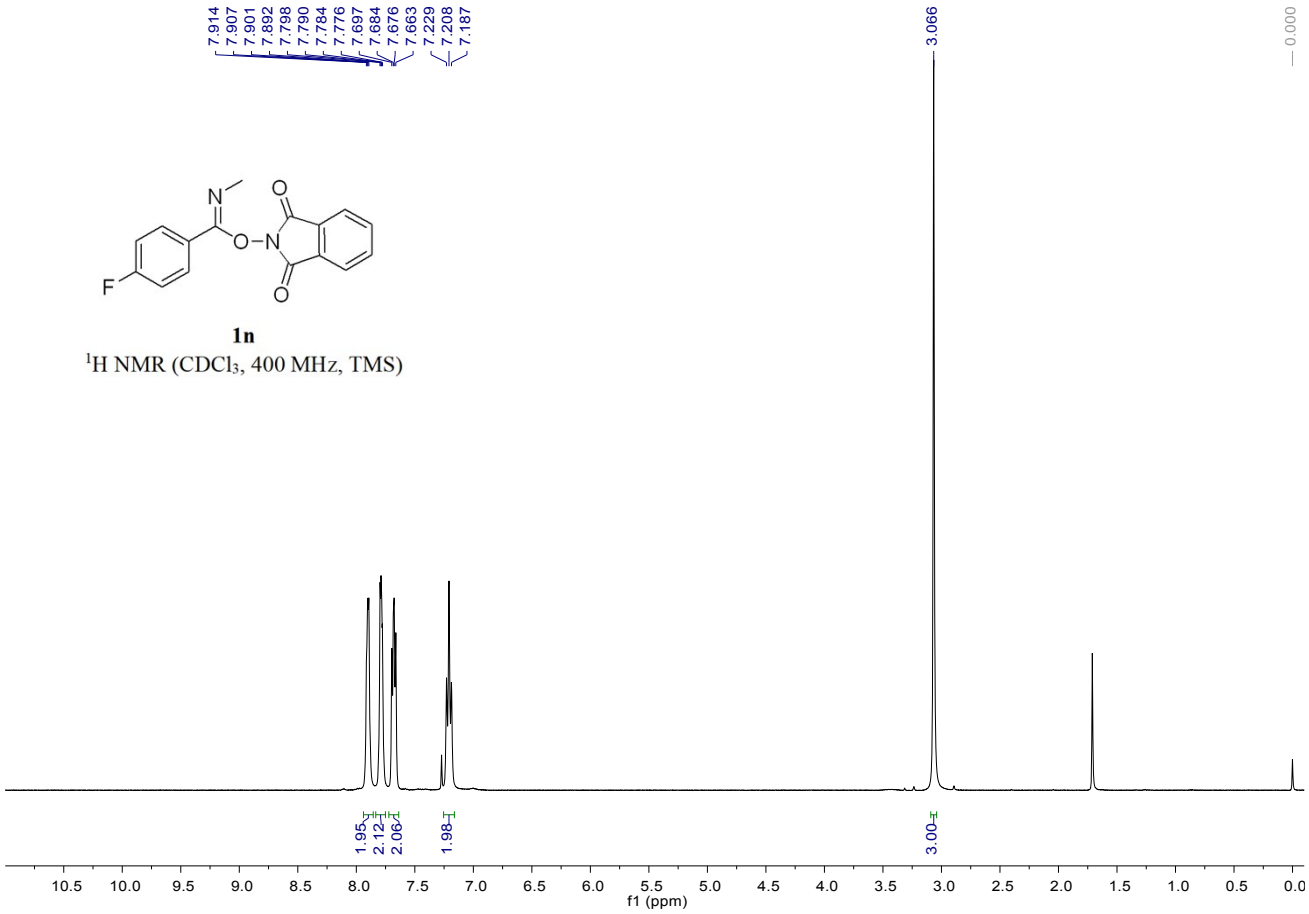
1,3-dioxoisoindolin-2-yl (*Z*)-4-bromo-N-methylbenzimidate (1m). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.58 g, 97% yield; M.p.: 178-180 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.93 - 7.86 (m, 2H), 7.78 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.66 (d, *J* = 8.1 Hz, 2H), 7.54 (d, *J* = 8.2 Hz, 2H), 3.05 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.0, 155.1, 134.4, 131.8, 130.5, 129.1, 125.6, 125.5, 123.6, 36.6; IR (neat) ν 1791, 1731, 1706, 1590, 1486, 1376, 1236, 1188, 1142, 1071, 1002, 880, 835, 781, 686 cm^{-1}; HRMS (ESI) Calcd. for $C_{16}H_{12}N_2O_3Br^+$ Requires: 359.0026, Found: 359.0018.





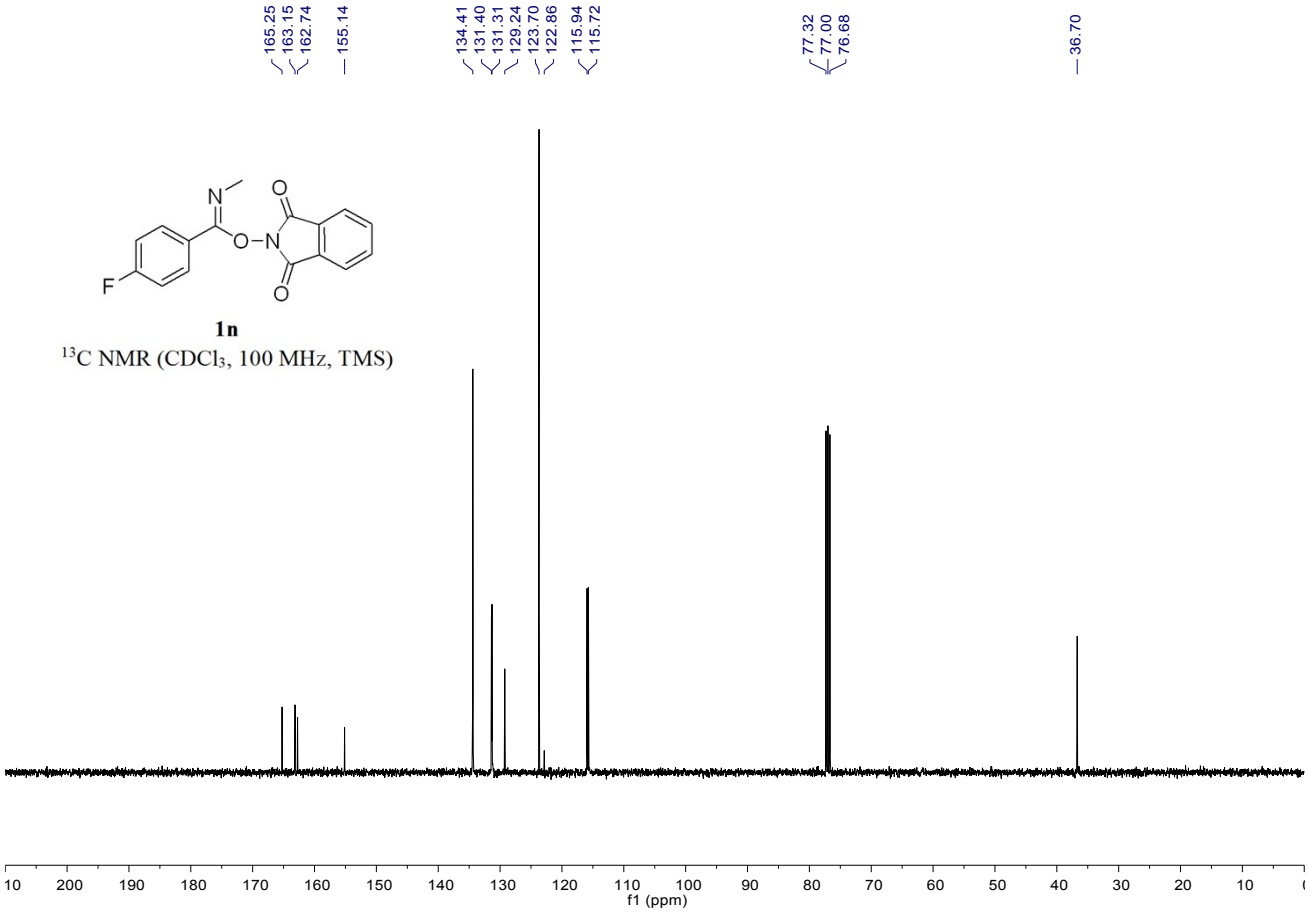


1,3-dioxoisoindolin-2-yl (*Z*)-4-fluoro-N-methylbenzimidate (1n). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.08 g, 80% yield; M.p.: 167-169 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.94 - 7.86 (m, 2H), 7.79 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.68 (dd, *J* = 8.4, 5.2 Hz, 2H), 7.21 (t, *J* = 8.4 Hz, 2H), 3.07 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 164.0 (d, *J* = 251.9 Hz), 163.1, 155.1, 134.4, 131.4 (d, *J* = 8.7 Hz), 129.2, 123.7, 122.9, 115.8 (d, *J* = 22.0 Hz), 36.7; ^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$) δ -107.9 (tt, *J* = 8.8, 5.5 Hz); IR (neat) ν 1793, 1733, 1717, 1602, 1505, 1371, 1241, 1184, 1135, 1021, 1005, 877, 845, 688 cm^{-1}; HRMS (ESI) Calcd. for $C_{16}H_{12}N_2O_3F^+$ Requires: 299.0827, Found: 299.0816.



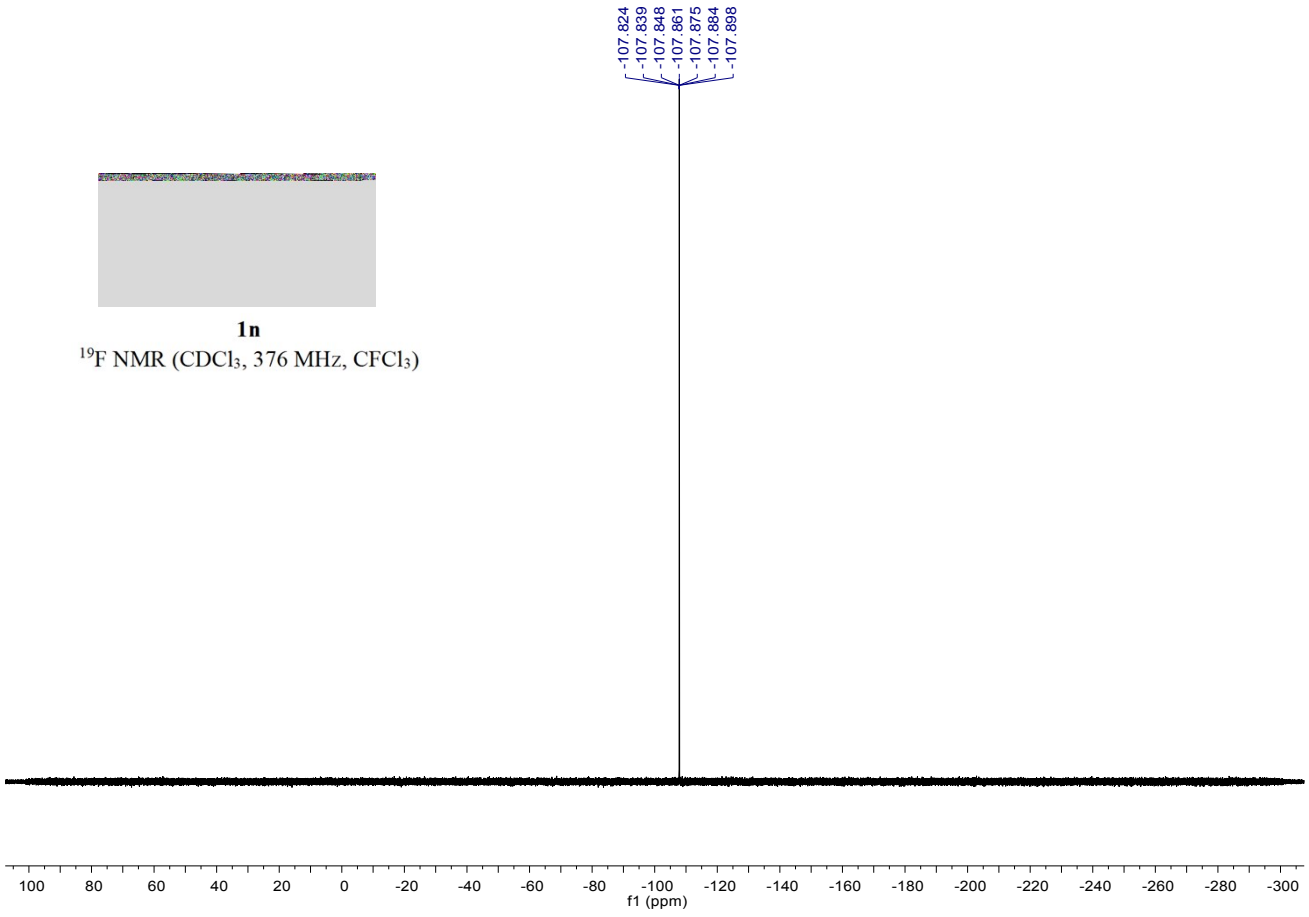
1n

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)



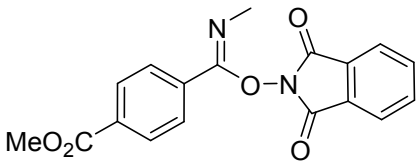
1n

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

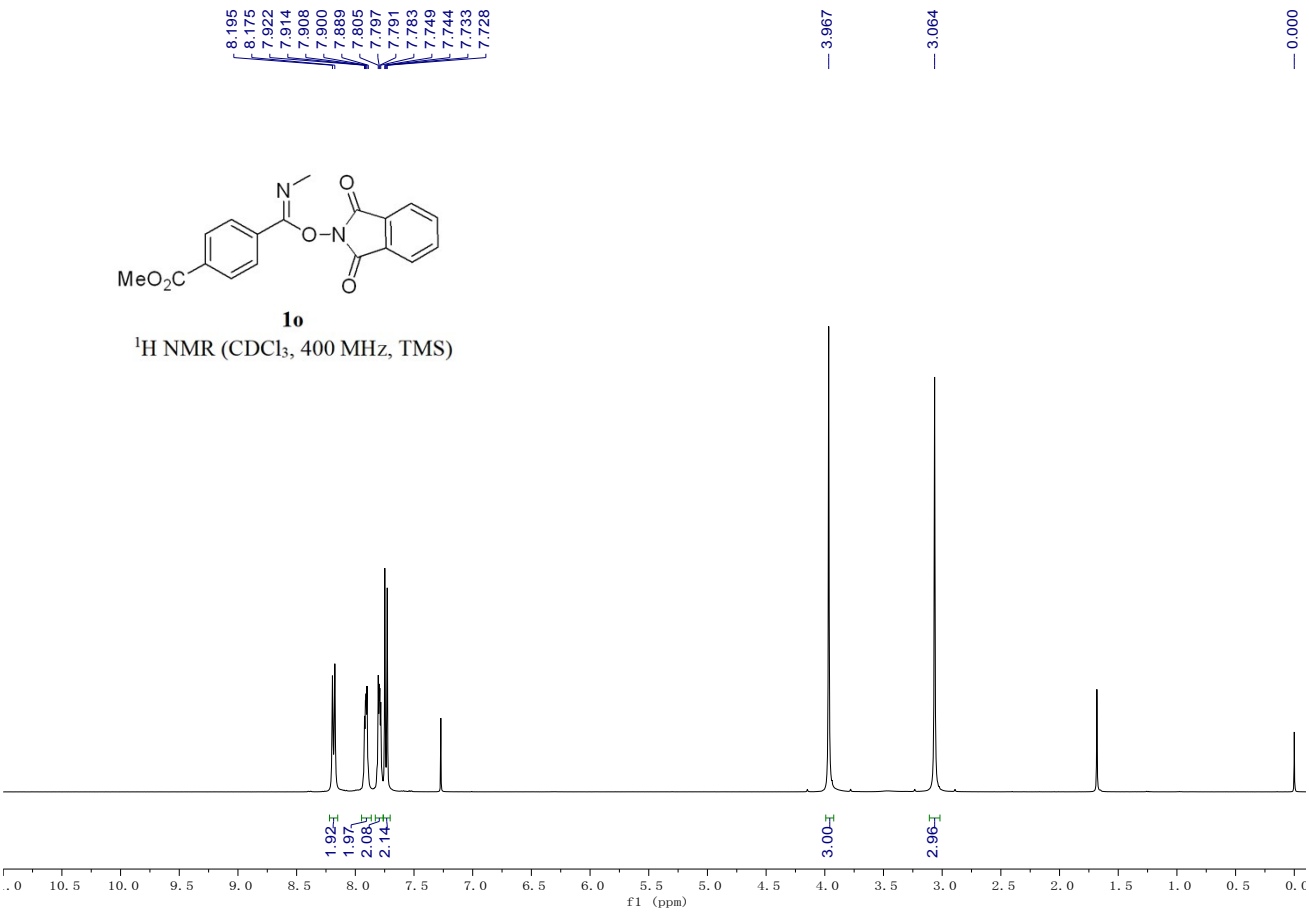


1n

^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$)

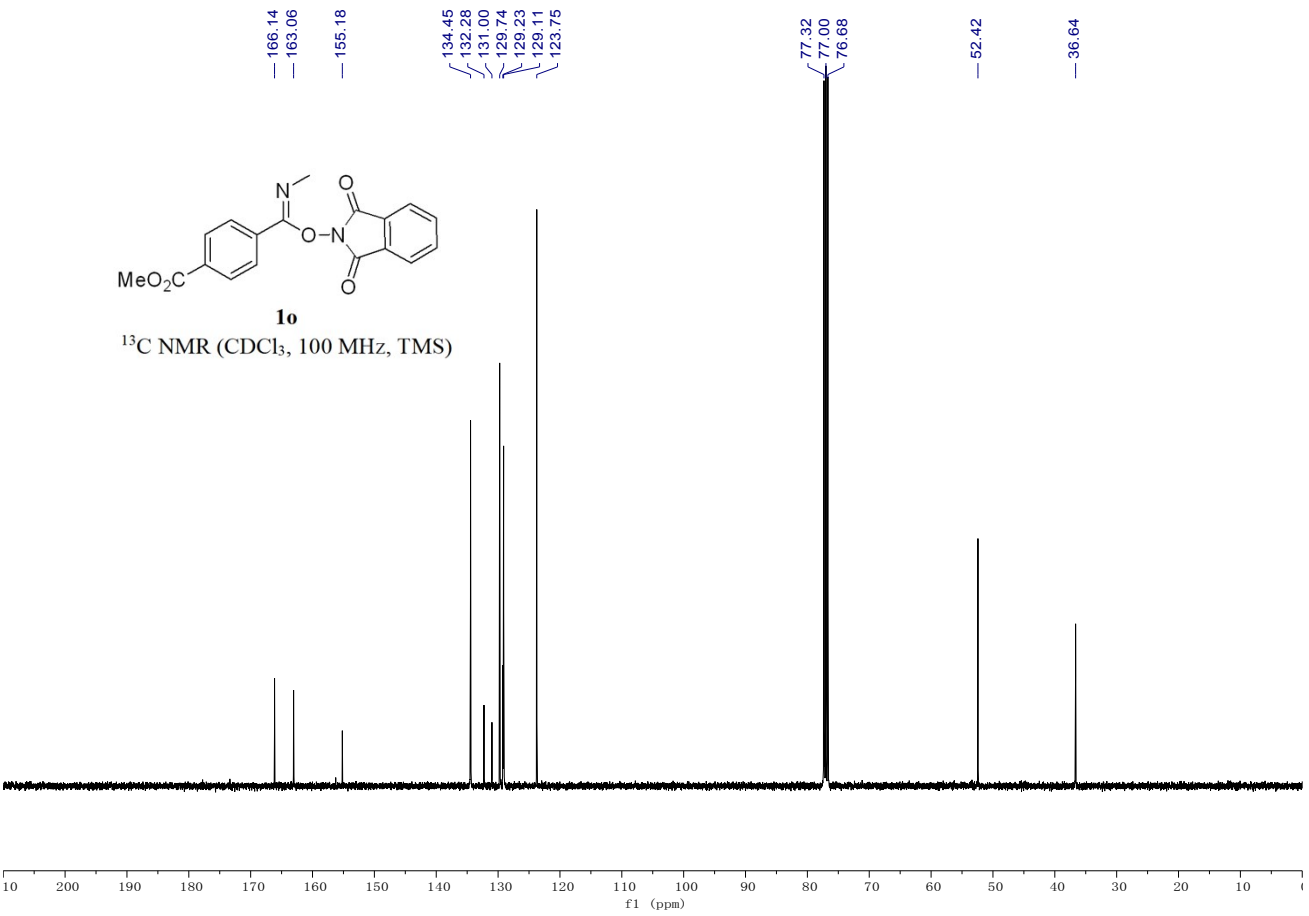


methyl (*Z*)-4-(((1,3-dioxoisoindolin-2-yl)oxy)(methylimino)methyl)benzoate (1o). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.28 g, 83% yield; M.p.: 186-188 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 8.18 (d, *J* = 8.0 Hz, 2H), 7.95 - 7.86 (m, 2H), 7.79 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.76 - 7.70 (m, 2H), 3.97 (s, 3H), 3.06 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 166.1, 163.1, 155.2, 134.5, 132.3, 131.0, 129.7, 129.2, 129.1, 123.7, 52.4, 36.6; IR (neat) ν 1797, 1737, 1718, 1435, 1355, 1283, 1240, 1187, 1116, 1006, 878, 763, 692 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{15}N_2O_5^+$ Requires: 339.0976, Found: 339.0976.



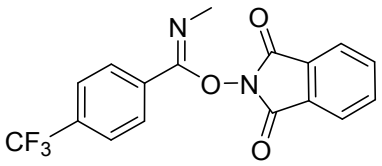
1o

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

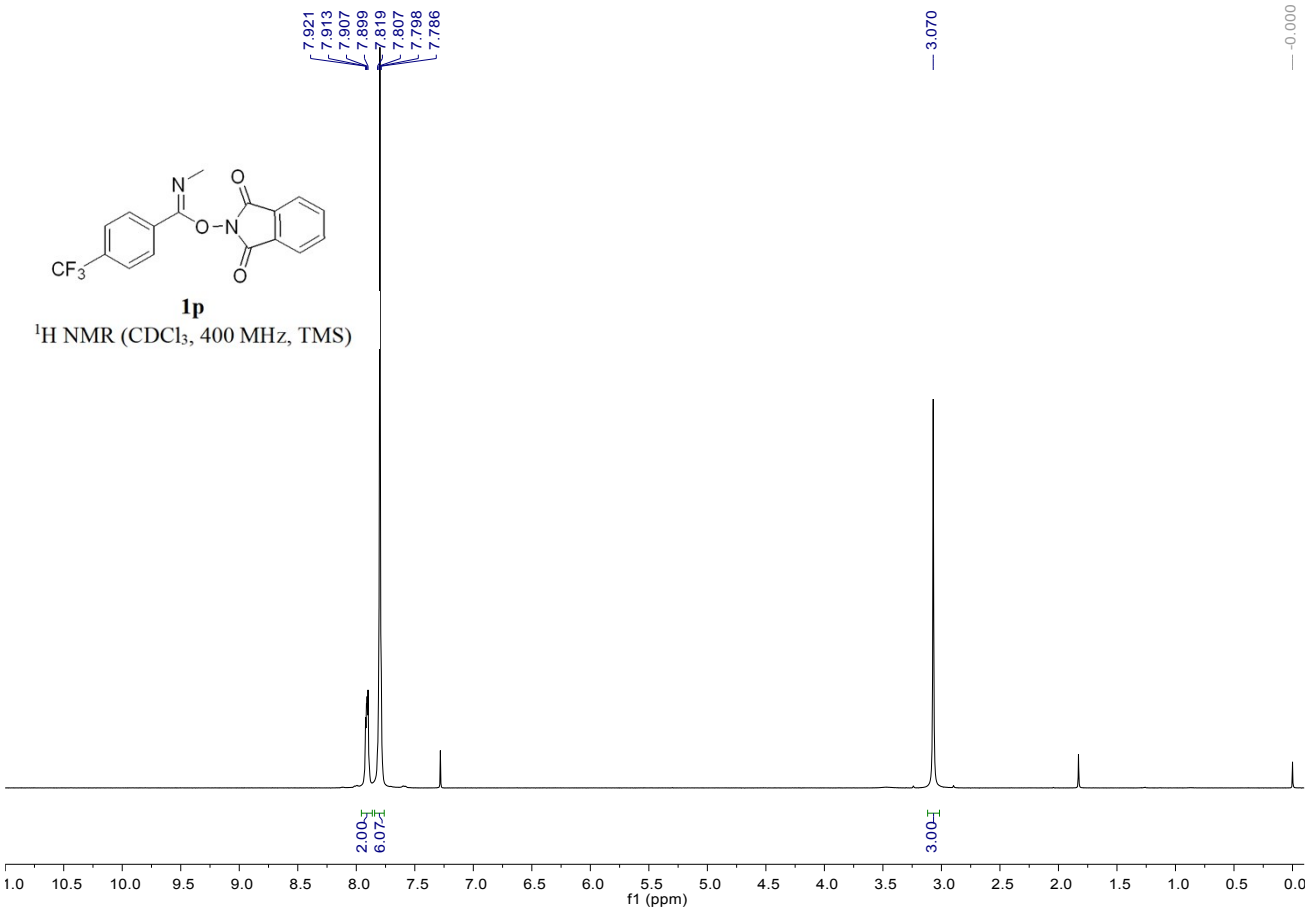


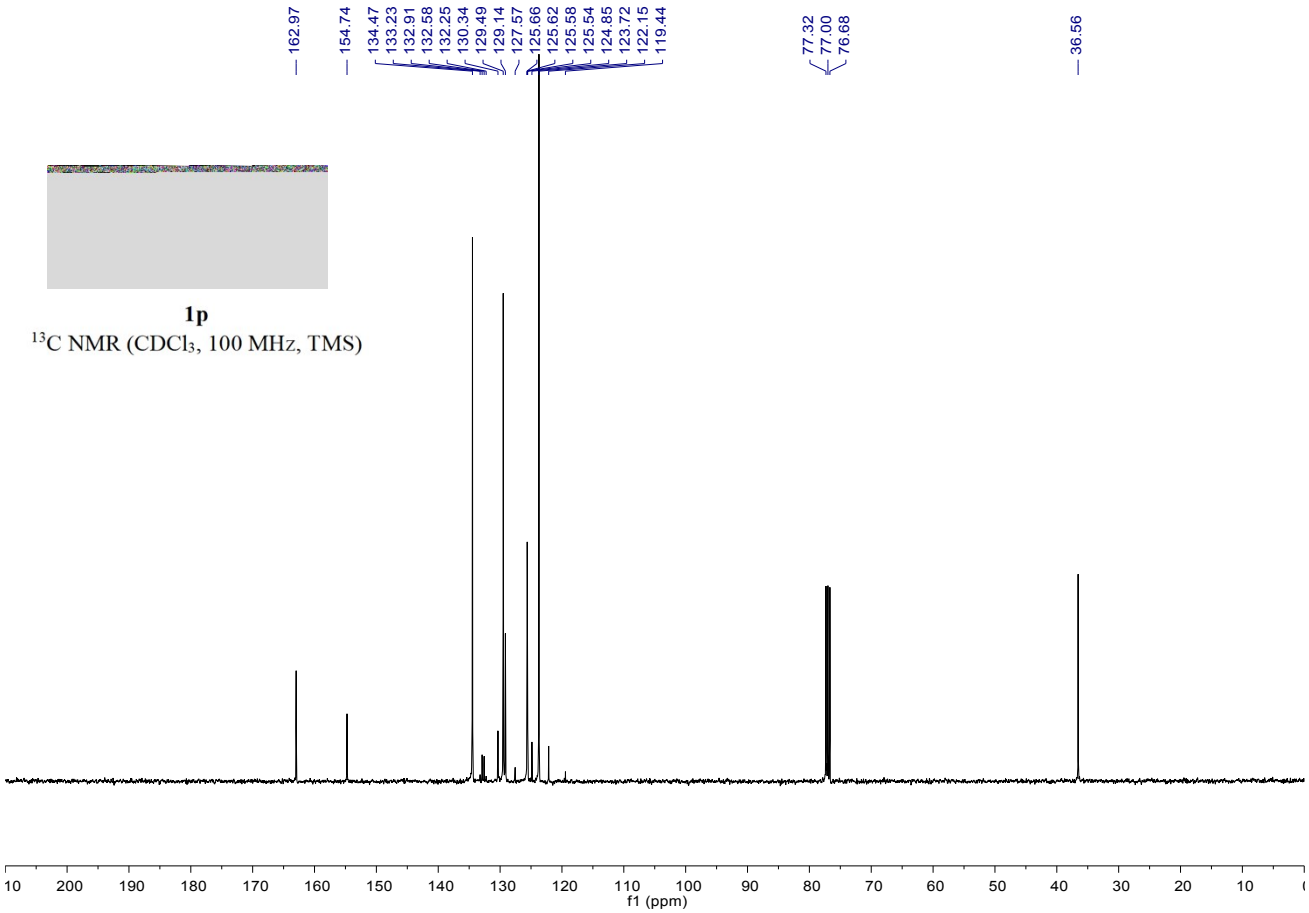
1o

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



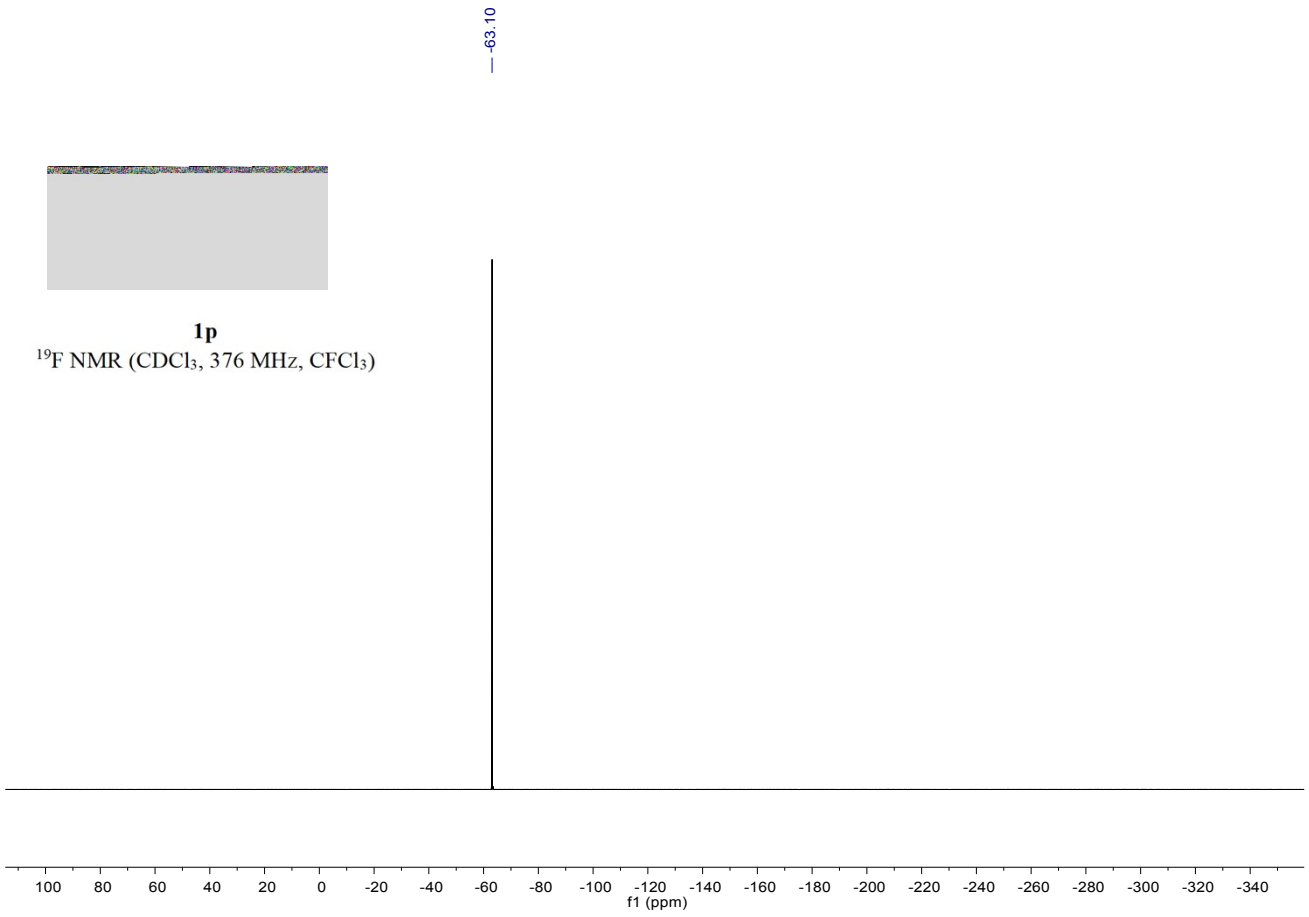
1,3-dioxoisoindolin-2-yl (*Z*)-N-methyl-4-(trifluoromethyl)benzimidate (1p). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.02 g, 65% yield; M.p.: 155-157 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.91 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.84 - 7.76 (m, 6H), 3.07 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.0, 154.7, 134.5, 132.7 (q, *J* = 32.8 Hz), 130.3, 129.5, 129.1, 125.6 (q, *J* = 3.8 Hz), 123.7, 123.5 (q, *J* = 272.6 Hz), 36.6; ^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$) δ -63.1; IR (neat) ν 1793, 1737, 1718, 1407, 1321, 1169, 1185, 1065, 1009, 879, 847, 780, 697 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{11}N_2O_3F_3Na^+$ Requires: 371.0614, Found: 371.0607.





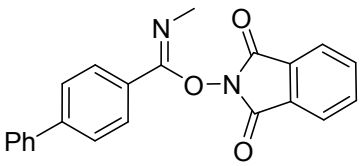
1p

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

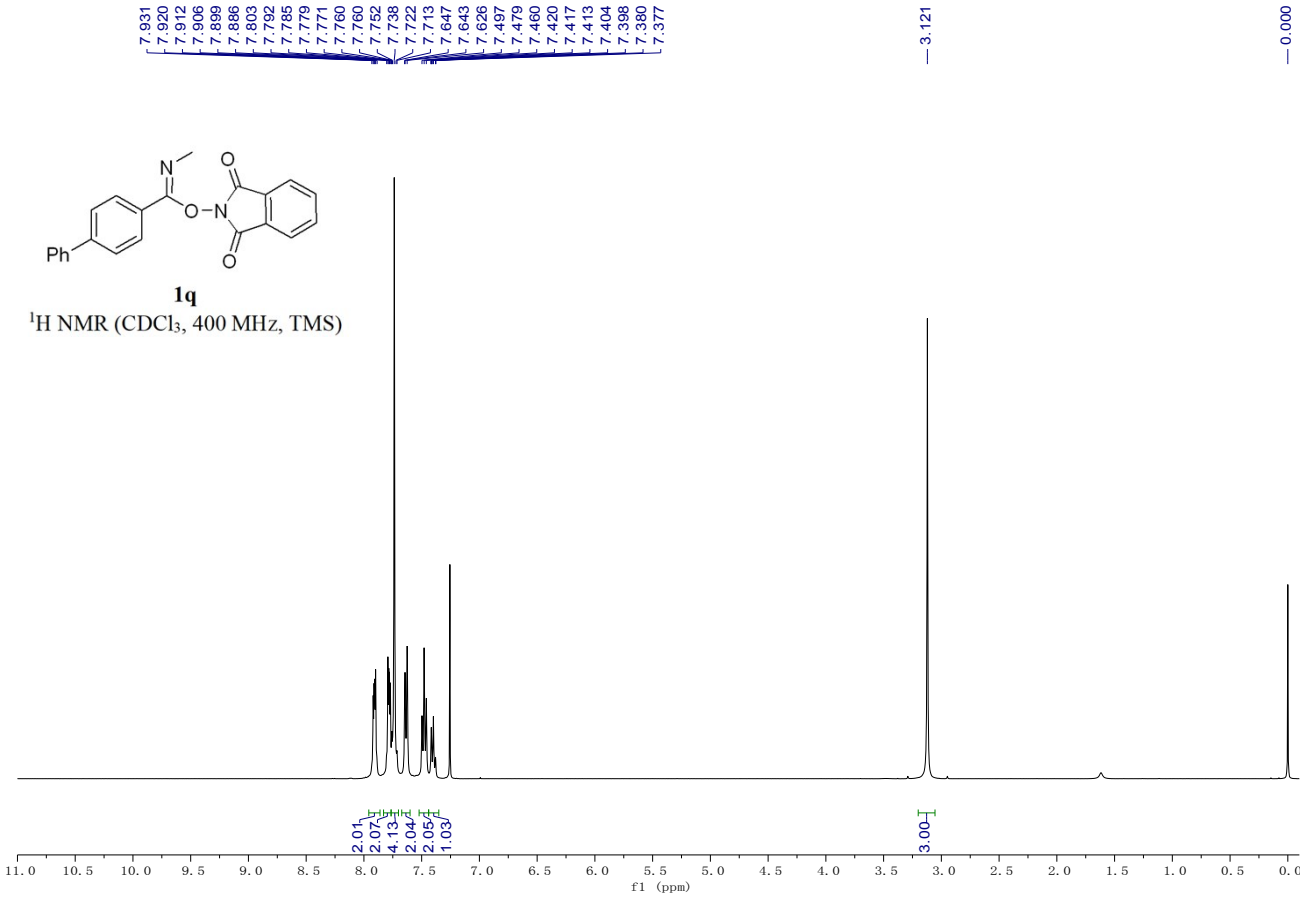


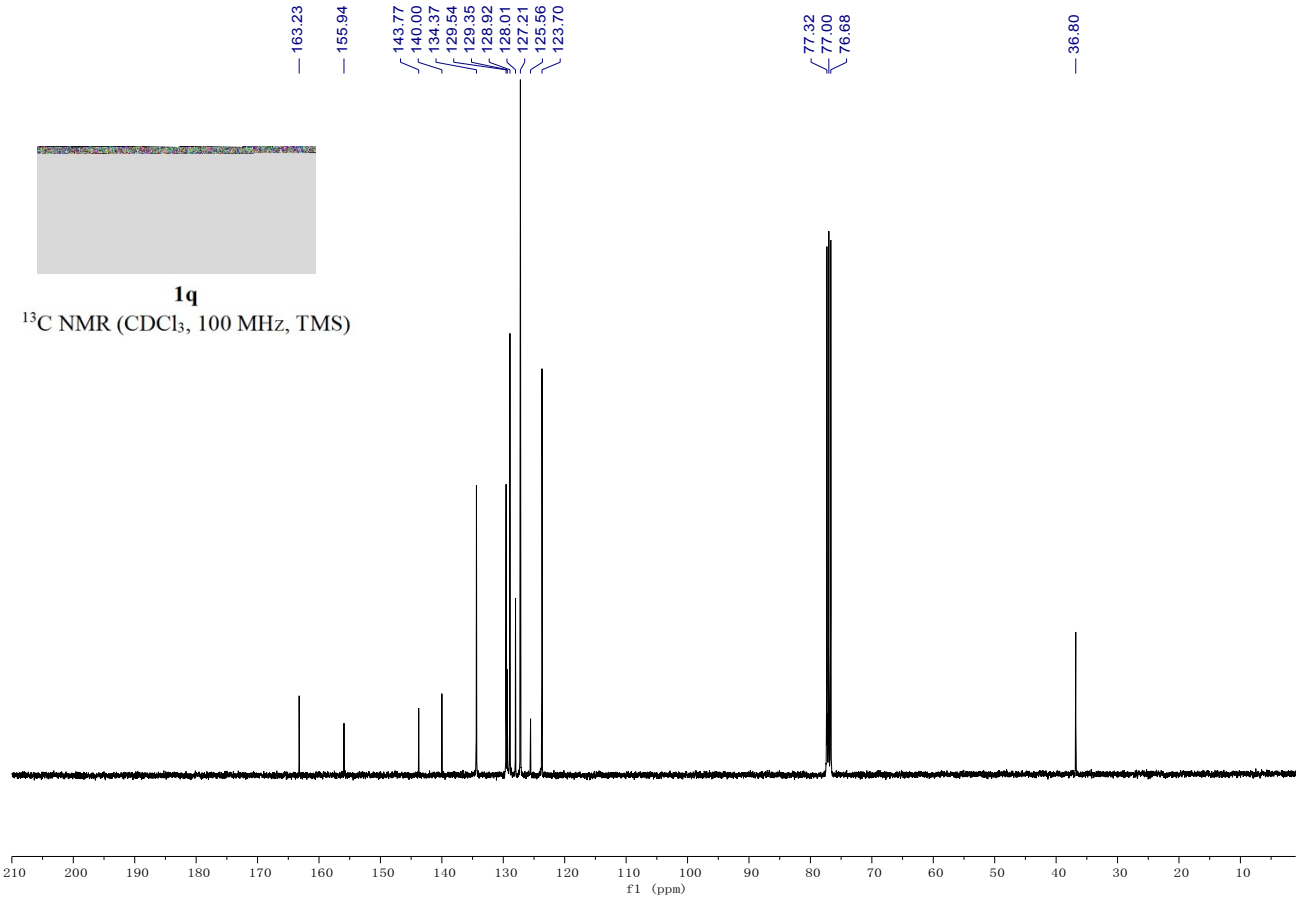
1p

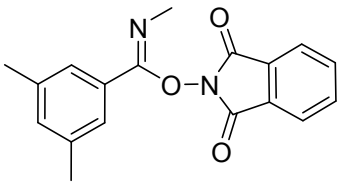
^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$)



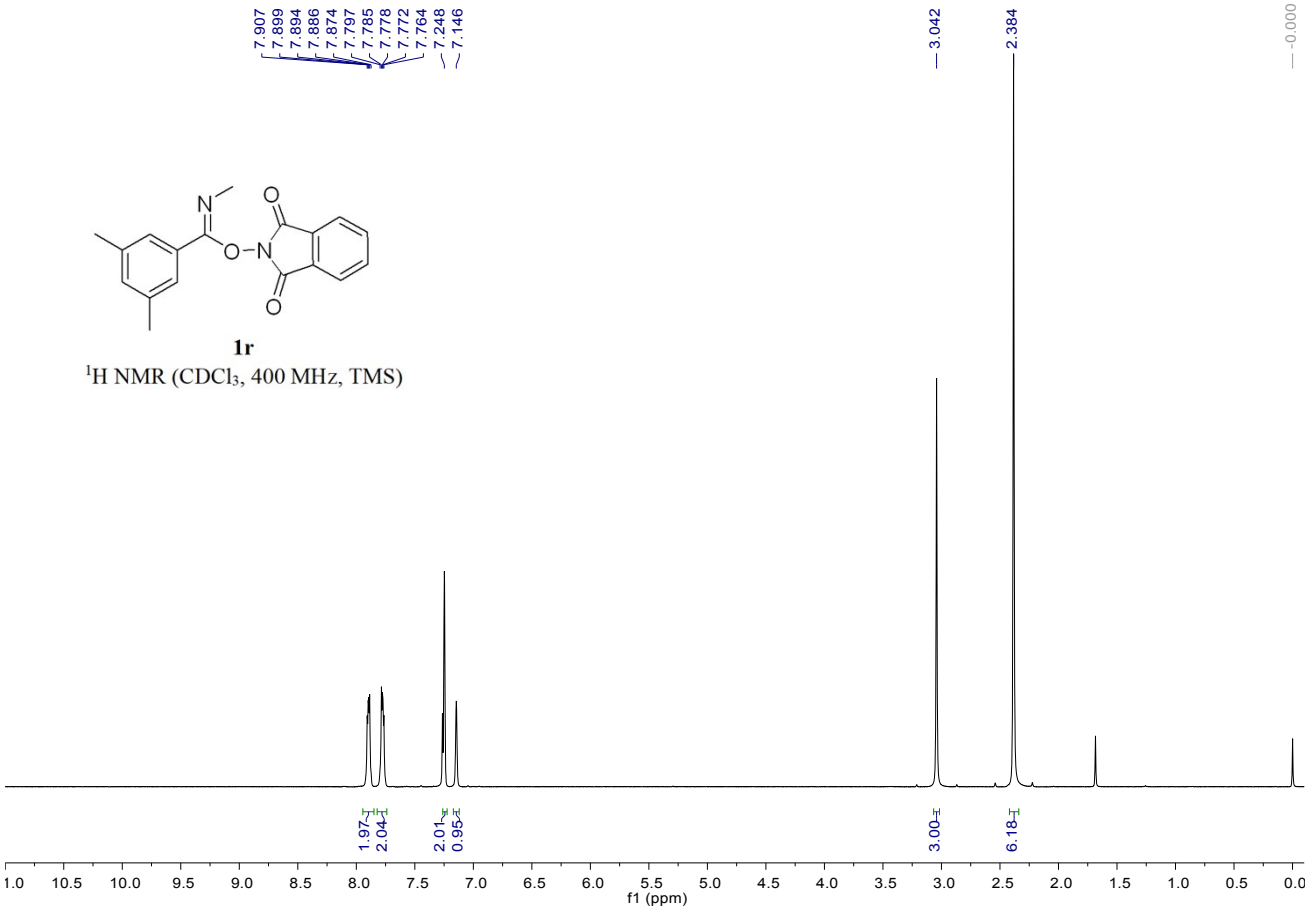
1,3-dioxoisoindolin-2-yl (*Z*)-N-methyl-[1,1'-biphenyl]-4-carbimidate (1q). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.39 g, 86% yield; M.p.: 190-192 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.96 - 7.86 (m, 2H), 7.83 - 7.76 (m, 2H), 7.76 - 7.70 (m, 4H), 7.67 - 7.60 (m, 2H), 7.48 (t, *J* = 7.5 Hz, 2H),z 7.44 - 7.35 (m, 1H), 3.12 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 155.9, 143.8, 140.0, 134.4, 129.5, 129.3, 128.9, 128.0, 127.2, 125.6, 123.7, 36.8; IR (neat) ν 1788, 1740, 1705, 1608, 1467, 1358, 1332, 1244, 1183, 1136, 1031, 1006, 878, 855, 765, 690 cm^{-1}; HRMS (ESI) Calcd. for $C_{22}H_{17}N_2O_3^+$ Requires: 357.1234, Found: 357.1129.





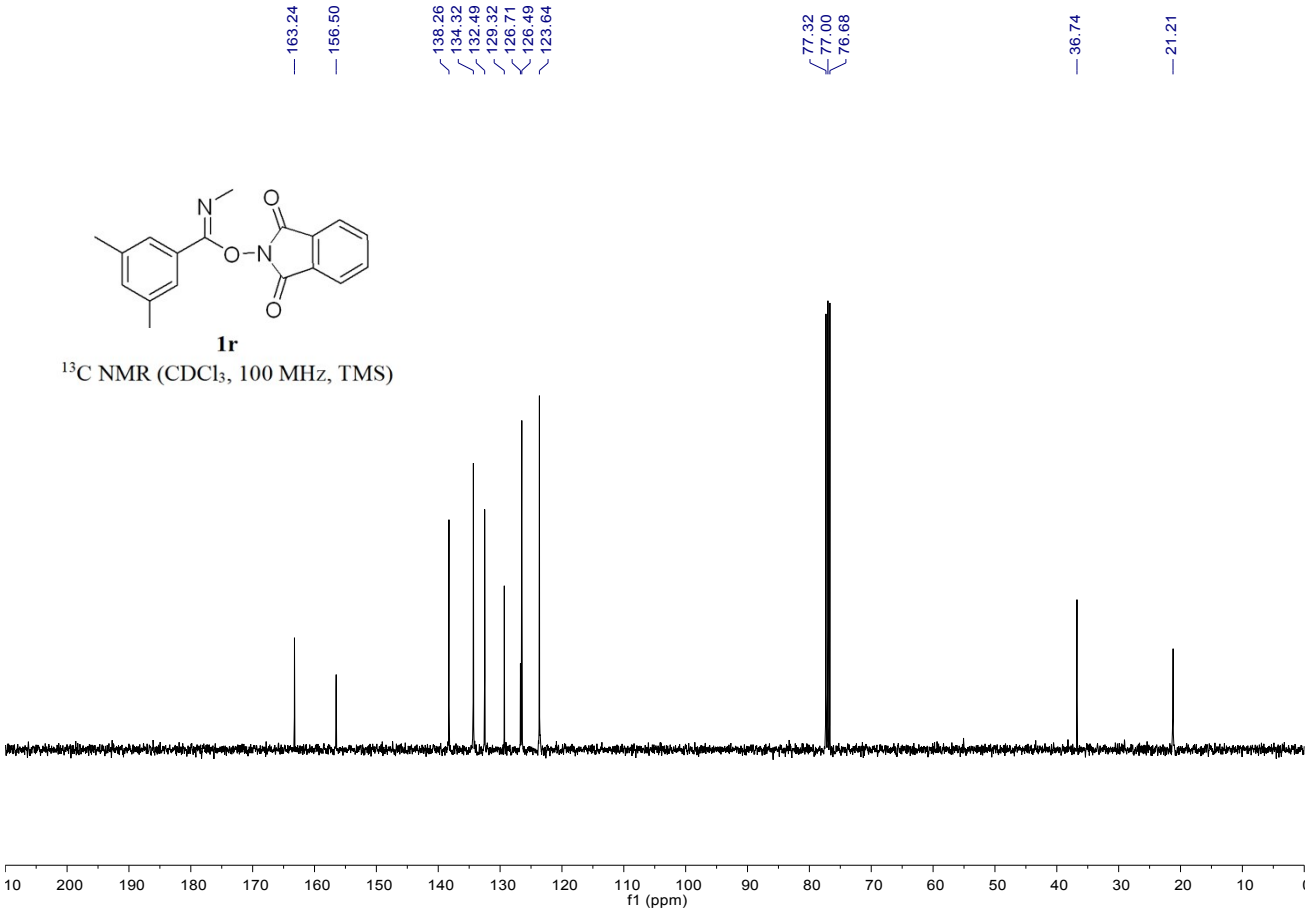


1,3-dioxoisoindolin-2-yl (*Z*)-N,3,5-trimethylbenzimidate (1r). The title product was obtained after purification by column chromatography (DCM). A white solid, 687 mg, 49% yield; M.p.: 156-158 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.90 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.77 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.25 (s, 2H), 7.15 (s, 1H), 3.04 (s, 3H), 2.38 (s, 6H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 156.5, 138.3, 134.3, 132.5, 129.3, 126.7, 126.5, 123.6, 36.7, 21.2; IR (neat) ν 1789, 1726, 1713, 1466, 1358, 1182, 1139, 1080, 990, 967, 877, 850, 787, 693 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{17}N_2O_3^+$ Requires: 309.1234, Found: 309.1222.



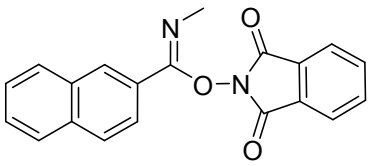
1r

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

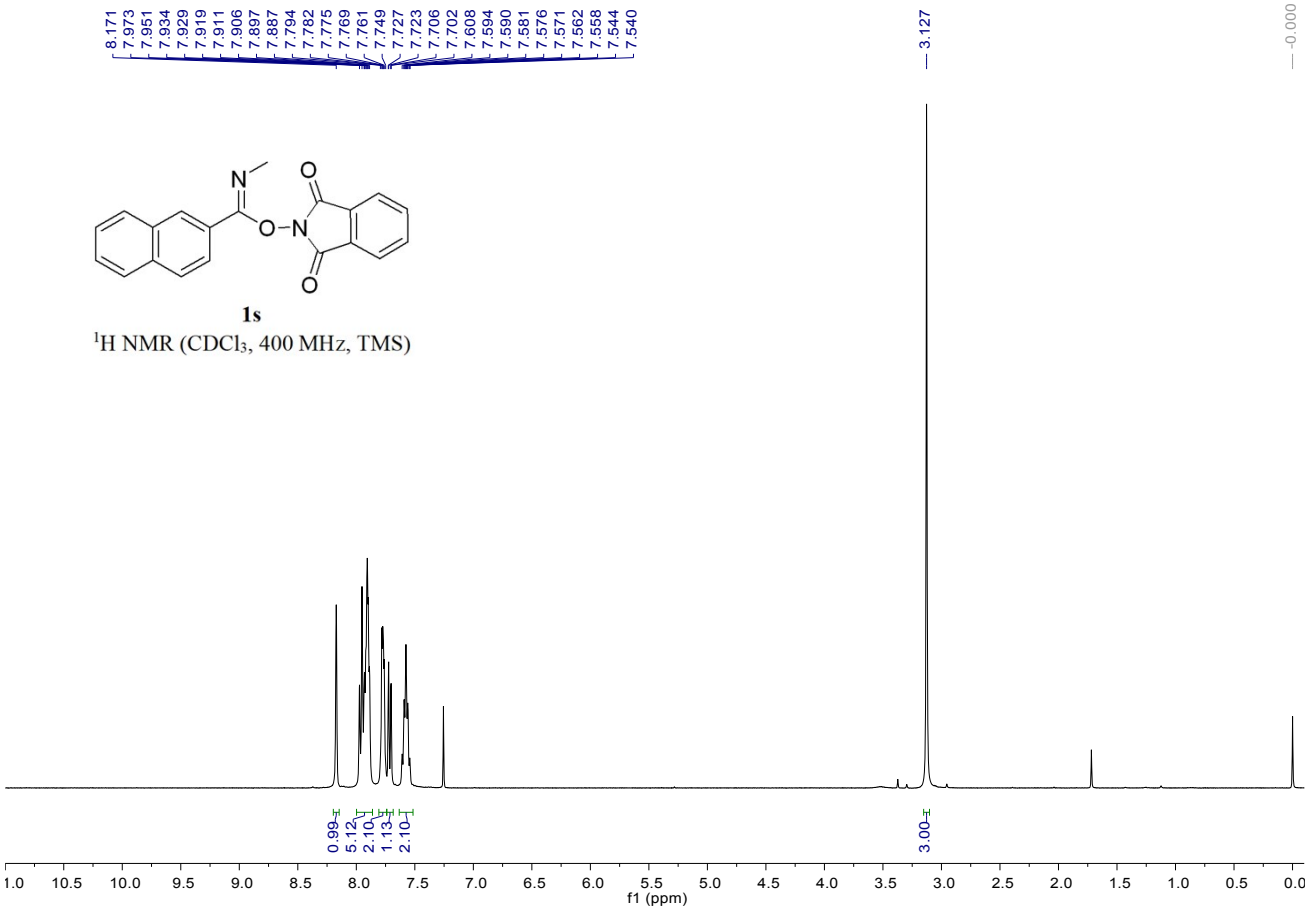


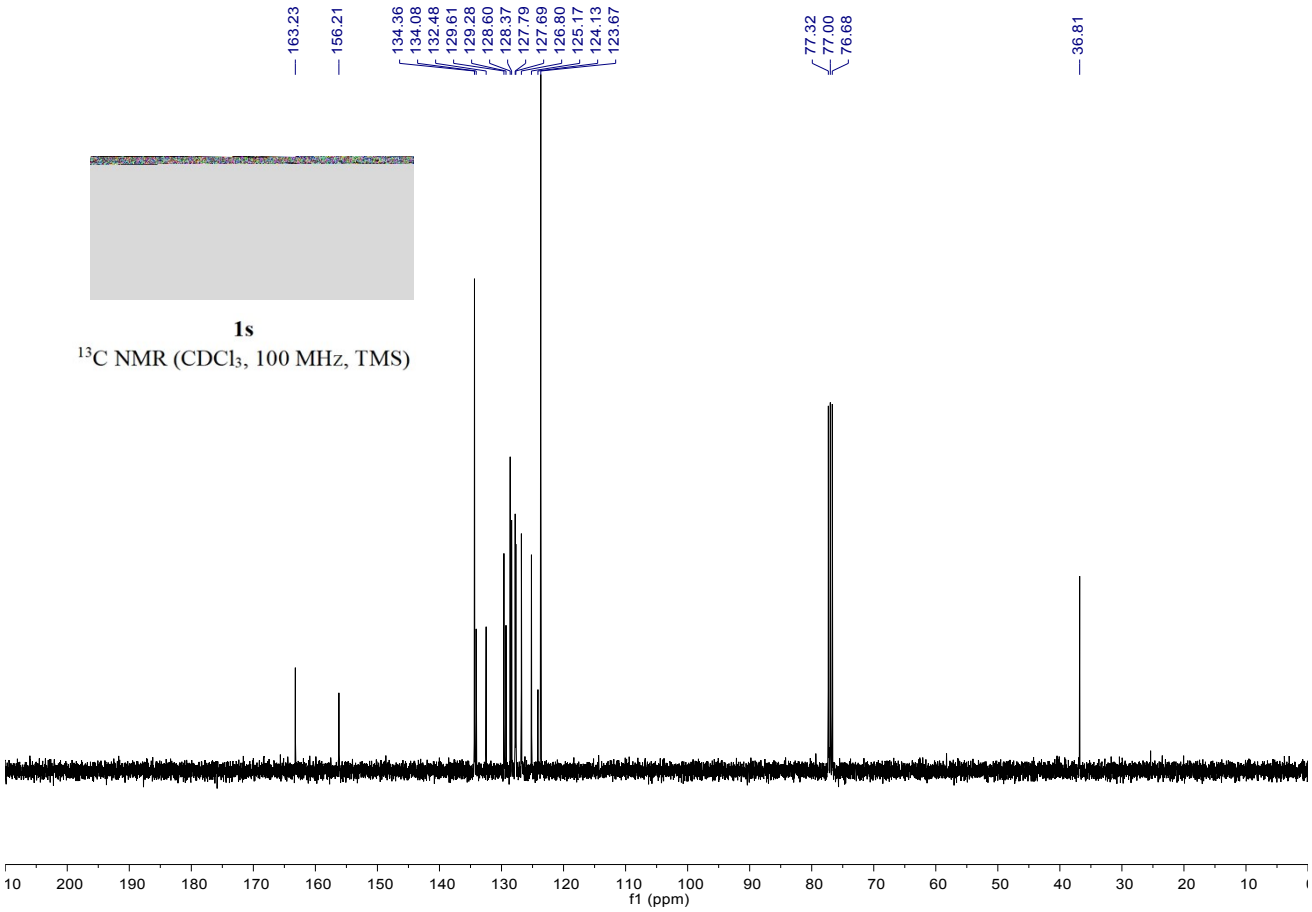
1r

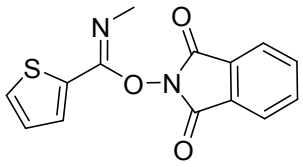
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



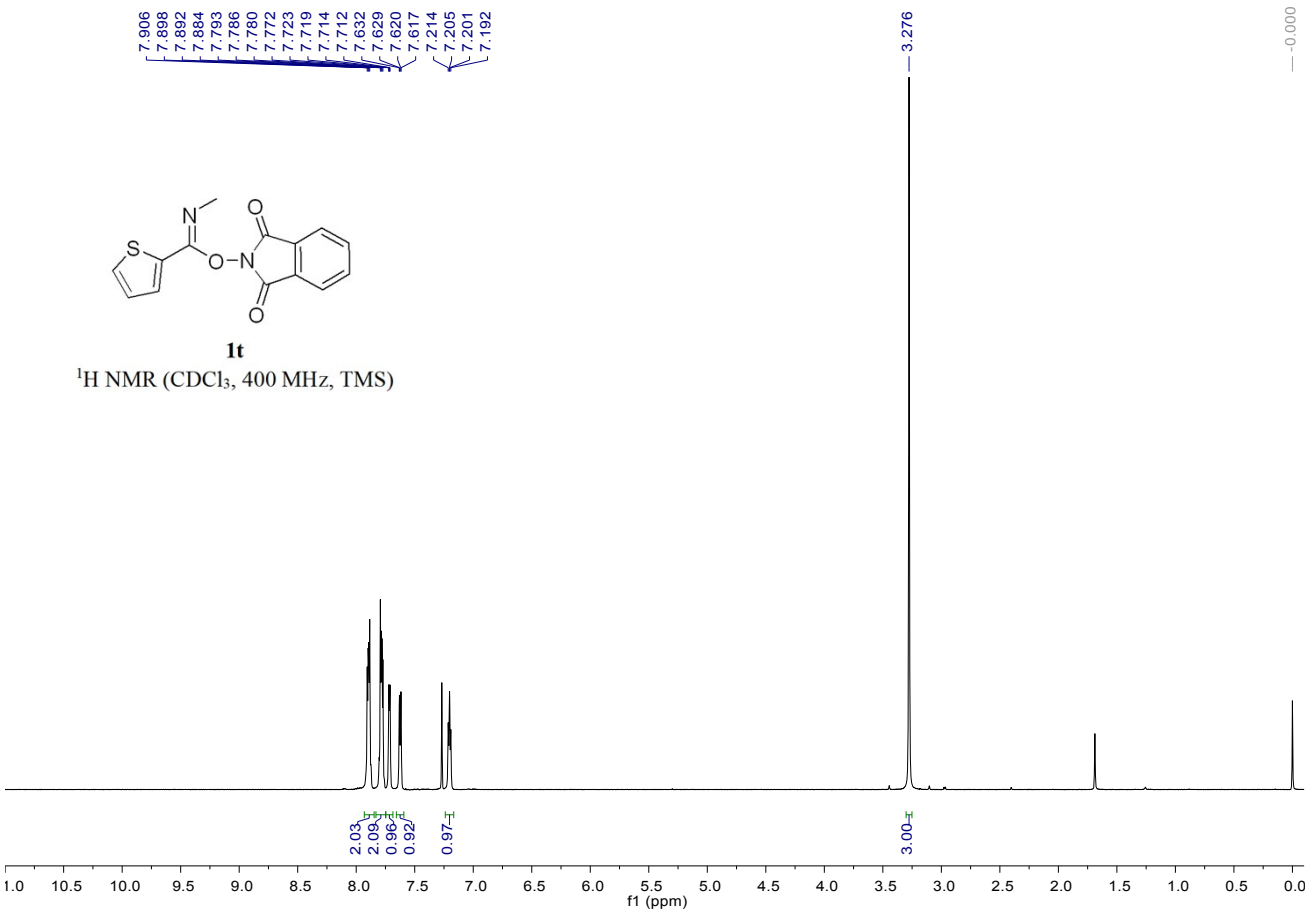
1,3-dioxoisoindolin-2-yl (*Z*)-N-methyl-2-naphthimidate (1s). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.18 g, 80% yield; M.p.: 204-206 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 8.17 (s, 1H), 8.00 - 7.86 (m, 5H), 7.77 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.71 (dd, *J* = 8.5, 1.6 Hz, 1H), 7.63 - 7.52 (m, 2H), 3.13 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 156.2, 134.4, 134.1, 132.5, 129.6, 129.3, 128.6, 128.4, 127.8, 127.7, 126.8, 125.2, 124.1, 123.7, 36.8; IR (neat) ν 1791, 1733, 1720, 1465, 1372, 1356, 1184, 1137, 1029, 965, 877, 830, 750, 689 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{15}N_2O_3^+$ Requires: 331.1077, Found: 331.1071.





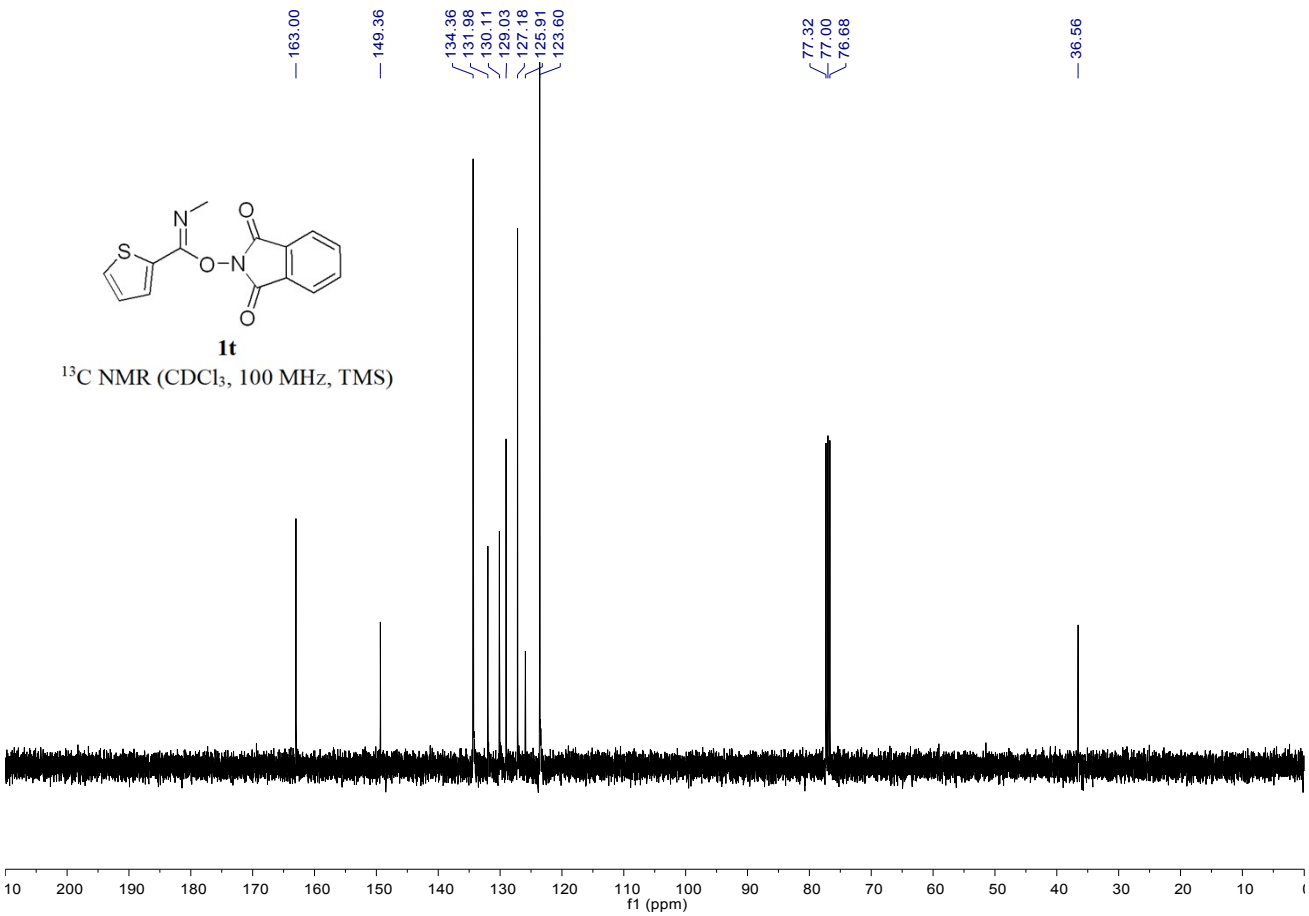


1,3-dioxoisoindolin-2-yl (*Z*)-N-methylthiophene-2-carbimidate (1t). The title product was obtained after purification by column chromatography (DCM). A white solid, 1.09 g, 84% yield; M.p.: 178-180 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.90 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.78 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.75 - 7.69 (m, 1H), 7.66 - 7.59 (m, 1H), 7.20 (dd, *J* = 5.1, 3.8 Hz, 1H), 3.28 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.0, 149.4, 134.4, 132.0, 130.1, 129.0, 127.2, 125.9, 123.6, 36.6; IR (neat) ν 1793, 1738, 1686, 1467, 1359, 1252, 1189, 1079, 985, 880, 783, 693 cm^{-1}; HRMS (ESI) Calcd. for $C_{14}H_{11}N_2O_3S^+$ Requires: 287.0485, Found: 287.0484.



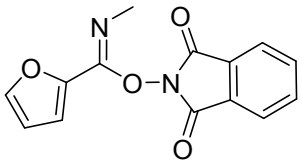
1t

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

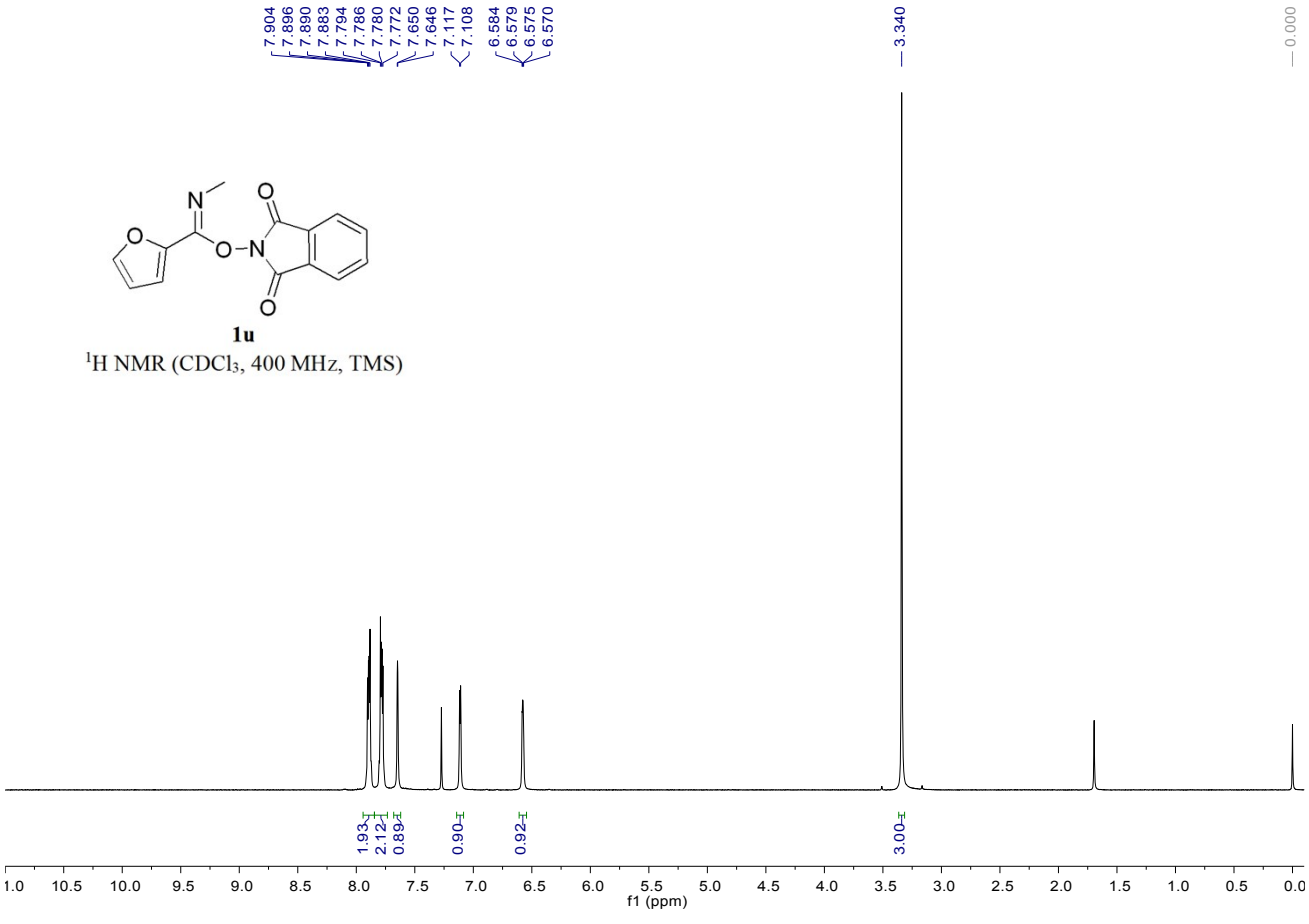


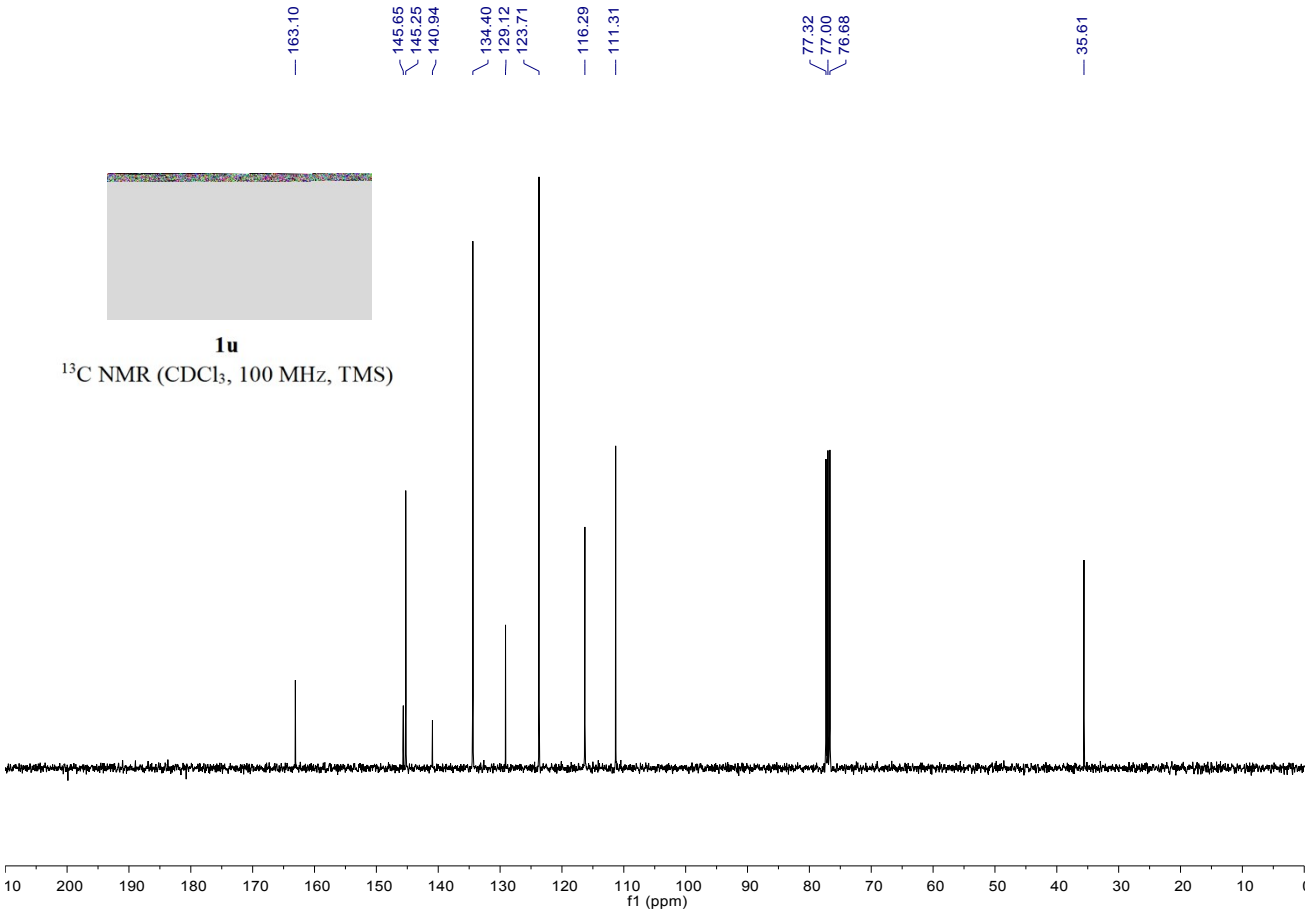
1t

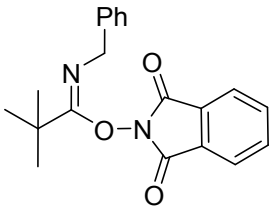
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



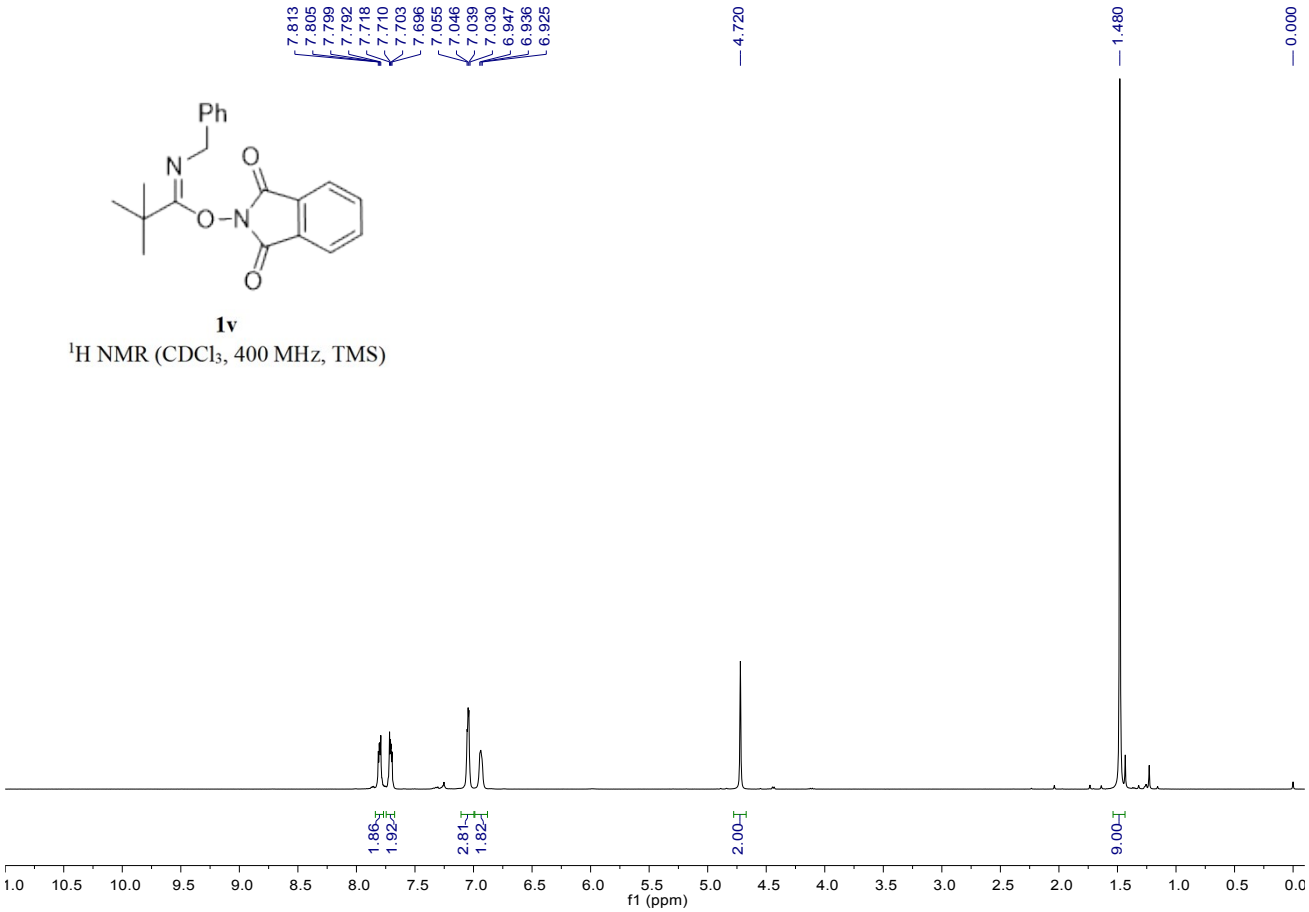
1,3-dioxoisoindolin-2-yl (*Z*)-N-methylfuran-2-carbimidate (1u). The title product was obtained after purification by column chromatography (DCM). A white solid, 823 mg, 67% yield; M.p.: 194-196 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.89 (dd, *J* = 5.4, 3.1 Hz, 2H), 7.78 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.65 (d, *J* = 1.7 Hz, 1H), 7.11 (d, *J* = 3.5 Hz, 1H), 6.58 (dd, *J* = 3.5, 1.8 Hz, 1H), 3.34 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.1, 145.6, 145.3, 140.9, 134.4, 129.1, 123.7, 116.3, 111.3, 35.6; IR (neat) ν 3145, 1792, 1729, 1702, 1466, 1381, 1258, 1190, 1158, 1032, 973, 902, 880, 774, 691 cm^{-1}; HRMS (ESI) Calcd. for $C_{14}H_{11}N_2O_4^+$ Requires: 271.0713, Found: 271.0714.





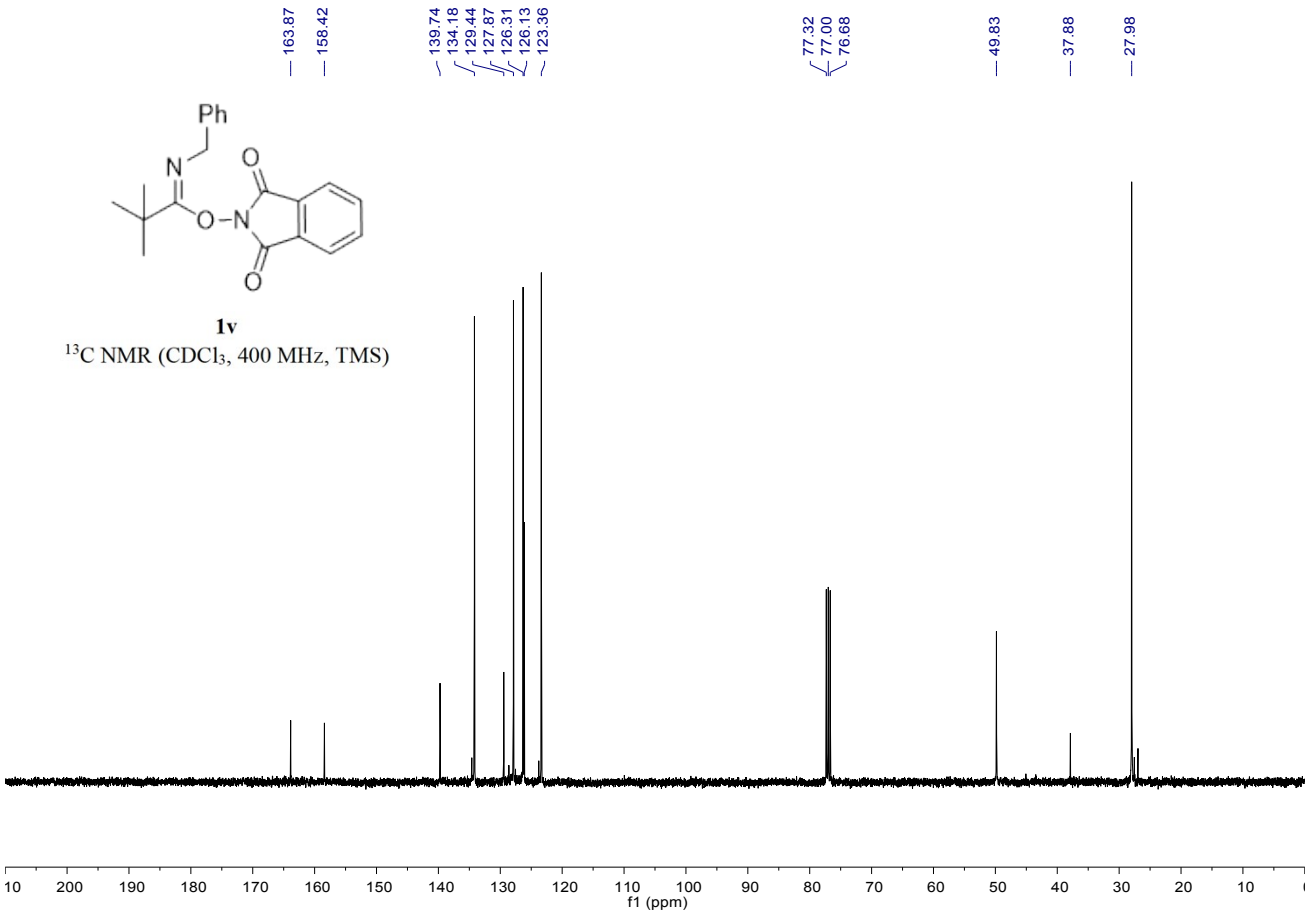


1,3-dioxoisoindolin-2-yl (Z)-N-benzylpivalimidate (1v). The title product was obtained after purification by column chromatography (PE:EA = 10:1). A white solid, 1.37 g, 50% yield; M.p.: 104 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.80 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.71 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.11 - 7.00 (m, 3H), 6.99 - 6.88 (m, 2H), 4.72 (s, 2H), 1.48 (s, 9H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.9, 158.4, 139.7, 134.2, 129.4, 127.9, 126.3, 126.1, 123.4, 49.8, 37.9, 28.0; IR (neat) ν 2972, 1797, 1736, 1714, 1451, 1367, 1347, 1187, 1104, 1079, 980, 877, 786, 738, 695 cm^{-1}; HRMS (FI) Calcd. for $C_{20}H_{20}N_2O_3^+$ Requires: 336.1468, Found: 336.1471.



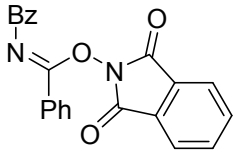
1v

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

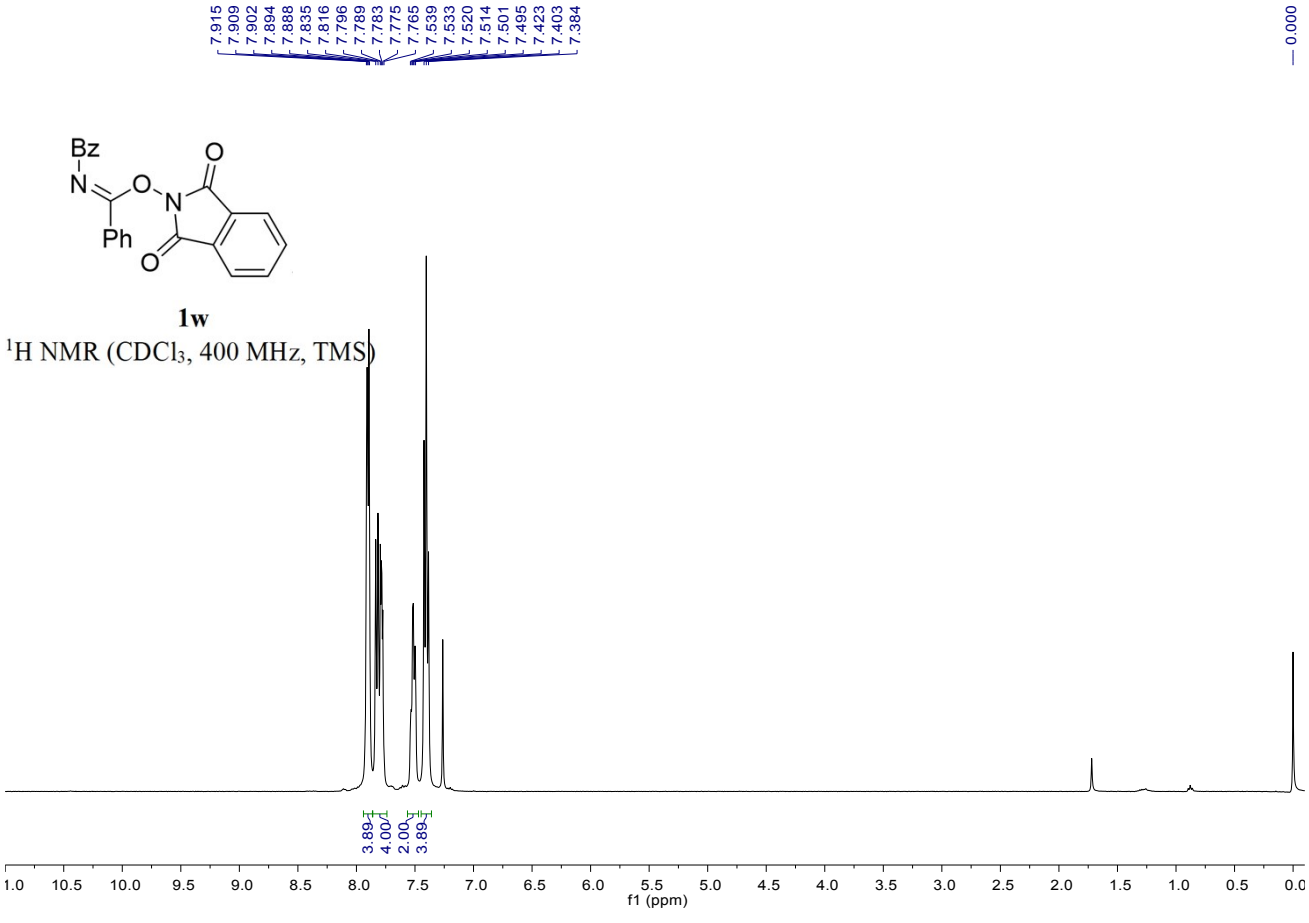


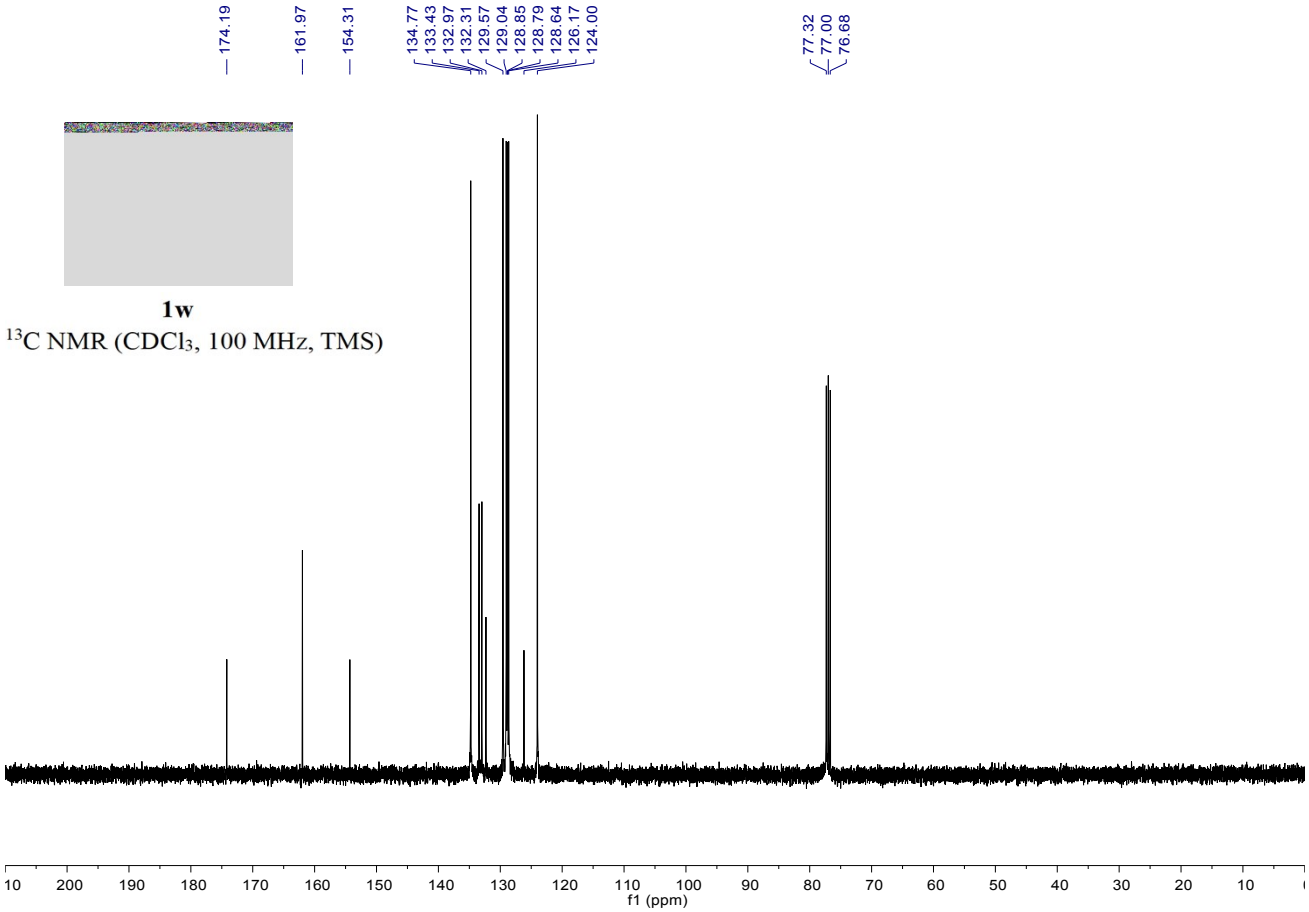
1v

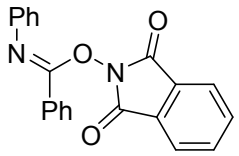
^{13}C NMR ($CDCl_3$, 400 MHz, TMS)



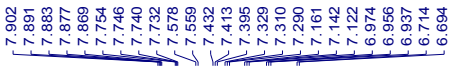
1,3-dioxoisoindolin-2-yl (Z)-N-benzoylbenzimidate (1w). The title product was obtained after purification by silica gel column chromatography (DCM). A white solid, 276 mg, 75% yield; M.p.: 182 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.94 - 7.86 (m, 4H), 7.86 - 7.74 (m, 4H), 7.51 (dt, *J* = 7.5, 3.6 Hz, 2H), 7.40 (t, *J* = 7.7 Hz, 4H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 174.2, 162.0, 154.3, 134.8, 133.4, 133.0, 132.3, 129.6, 129.0, 128.85, 128.79, 128.6, 126.2, 124.0; IR (neat) ν 1799, 1742, 1709, 1667, 1448, 1355, 1267, 1234, 1185, 1134, 1042, 1021, 877, 868, 776, 734, 688 cm^{-1}.

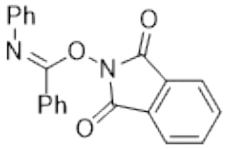






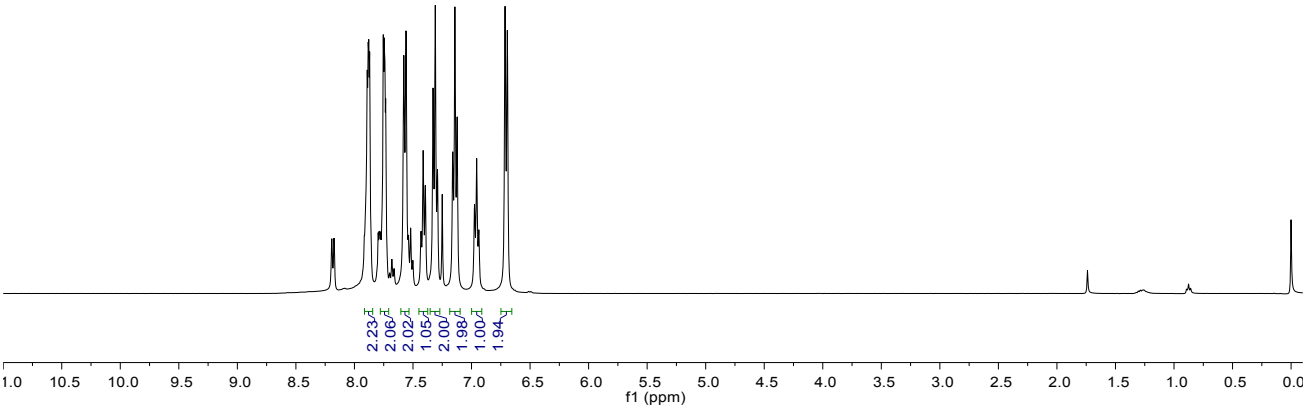
1,3-dioxoisoindolin-2-yl (Z)-N-phenylbenzimidate (1x). The title product was obtained after purification by silica gel column chromatography (DCM). A white solid, 330 mg, 48% yield (87% w/w); M.p.: 138 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.88 (dd, *J* = 5.5, 3.2 Hz, 2H), 7.74 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.57 (d, *J* = 7.7 Hz, 2H), 7.41 (t, *J* = 7.4 Hz, 1H), 7.31 (t, *J* = 7.6 Hz, 2H), 7.14 (t, *J* = 7.7 Hz, 2H), 6.96 (t, *J* = 7.4 Hz, 1H), 6.70 (d, *J* = 7.7 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 162.7, 145.4, 134.7, 134.5, 131.4, 130.5, 129.8, 129.1, 128.9, 128.3, 123.7, 123.7, 120.8; IR (neat) ν 1733, 1699, 1593, 1365, 1183, 1038, 1019, 1009, 877, 775, 692, 677 cm^{-1}.

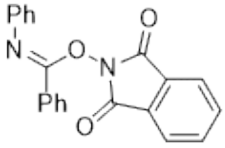




1x

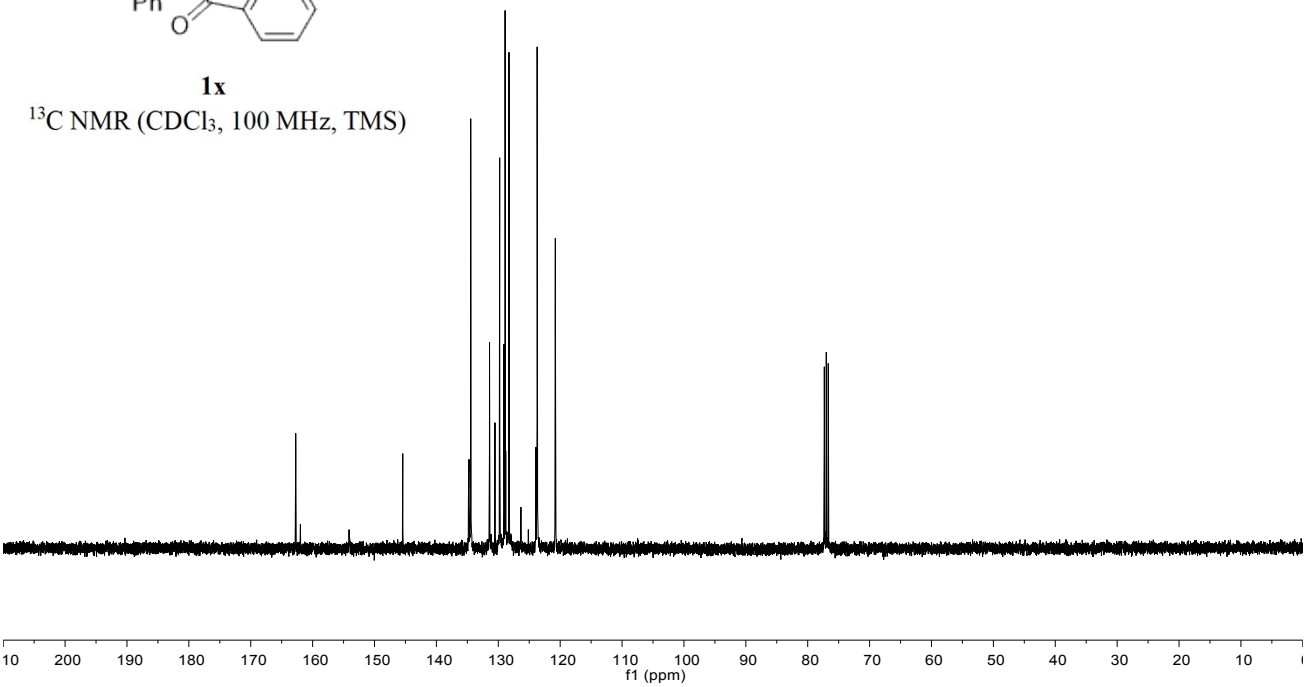
^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

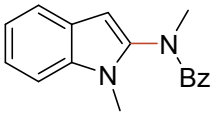




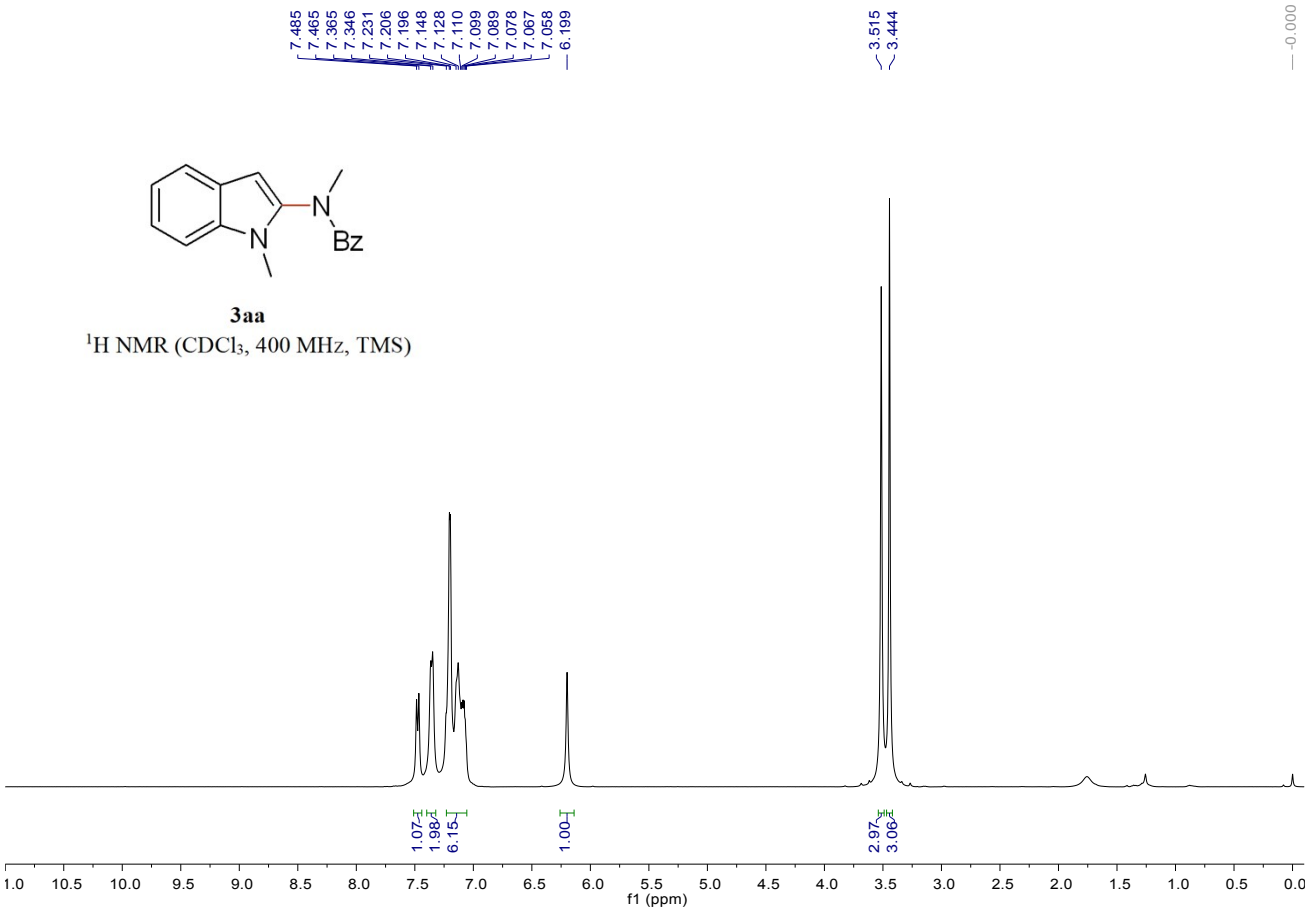
1x

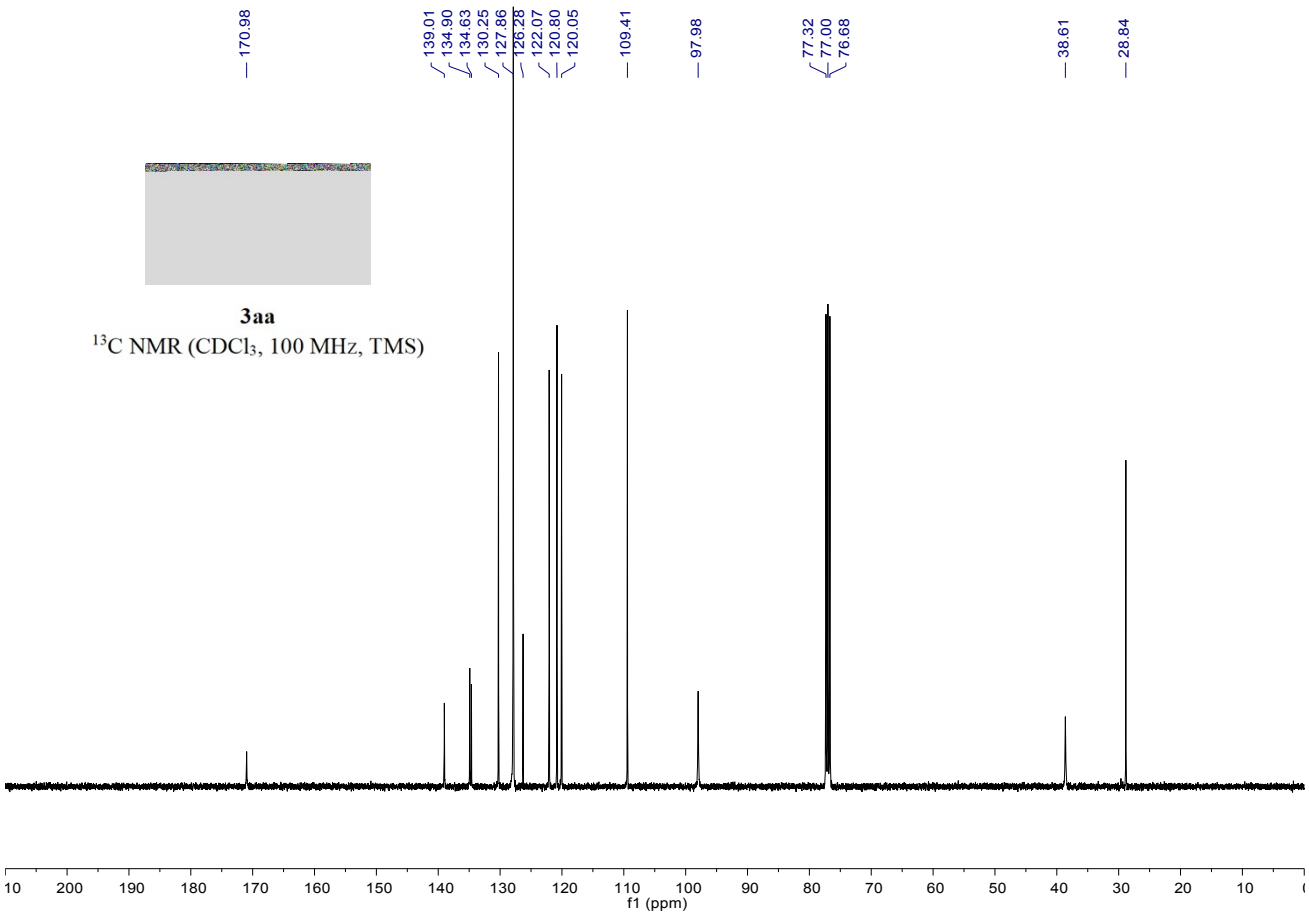
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

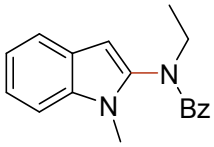




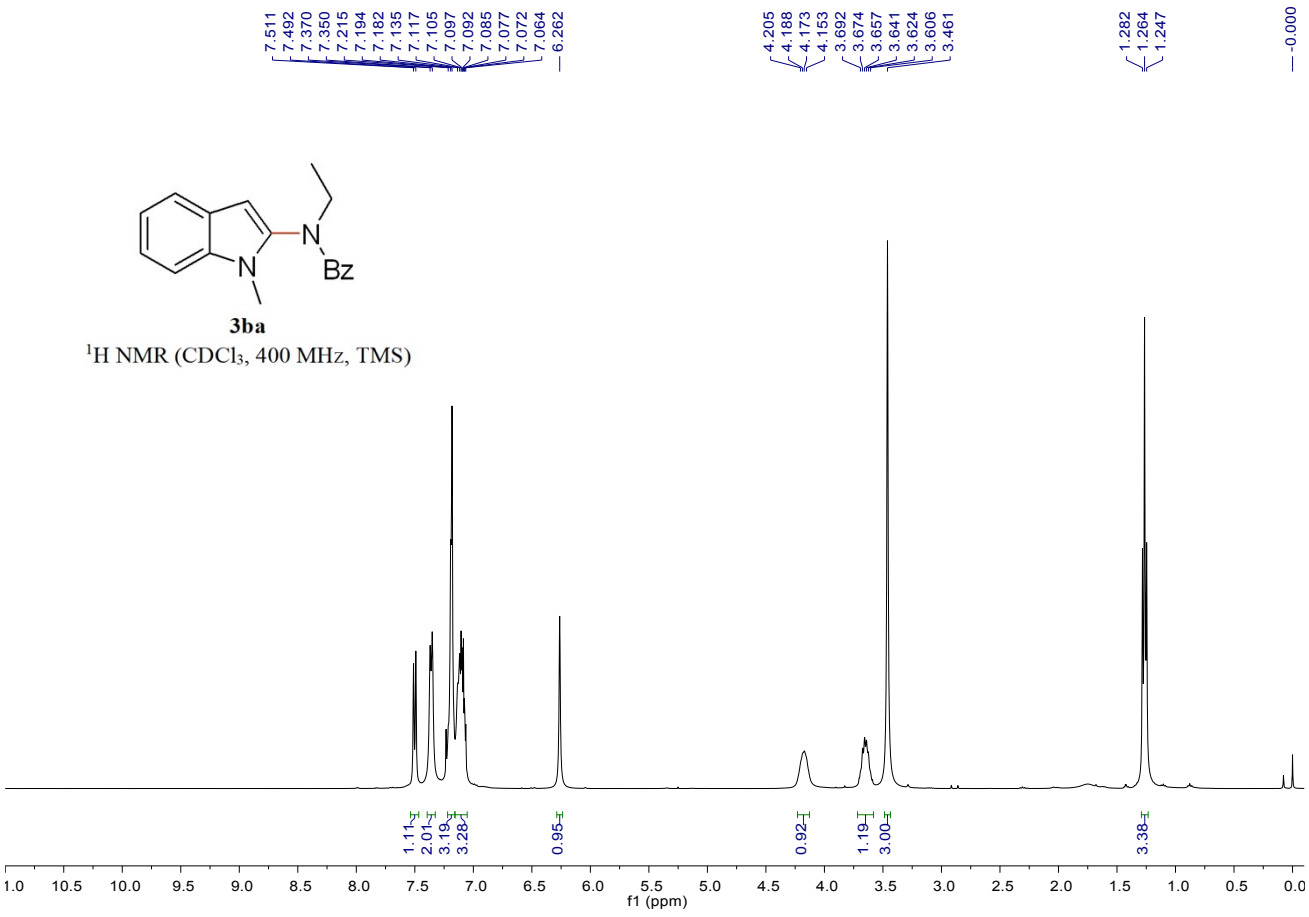
***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3aa)**.[4] The title product was obtained after purification by prep-TLC (PE:EA = 4:1). An off-white solid, 39 mg, 73% yield; M.p.: 110 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.48 (d, *J* = 8.0 Hz, 1H), 7.36 (d, *J* = 7.6 Hz, 2H), 7.23 - 7.06 (m, 6H), 6.20 (s, 1H), 3.51 (s, 3H), 3.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.0, 139.0, 134.9, 134.6, 130.3, 127.9, 126.3, 122.1, 120.8, 120.1, 109.4, 98.0, 38.6, 28.8; IR (neat) ν 1648, 1557, 1444, 1393, 1339, 1316, 1253, 1090, 1010, 775, 735, 716, 694, 670 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{16}N_2ONa^+$ Requires: 287.1155, Found: 287.1153.





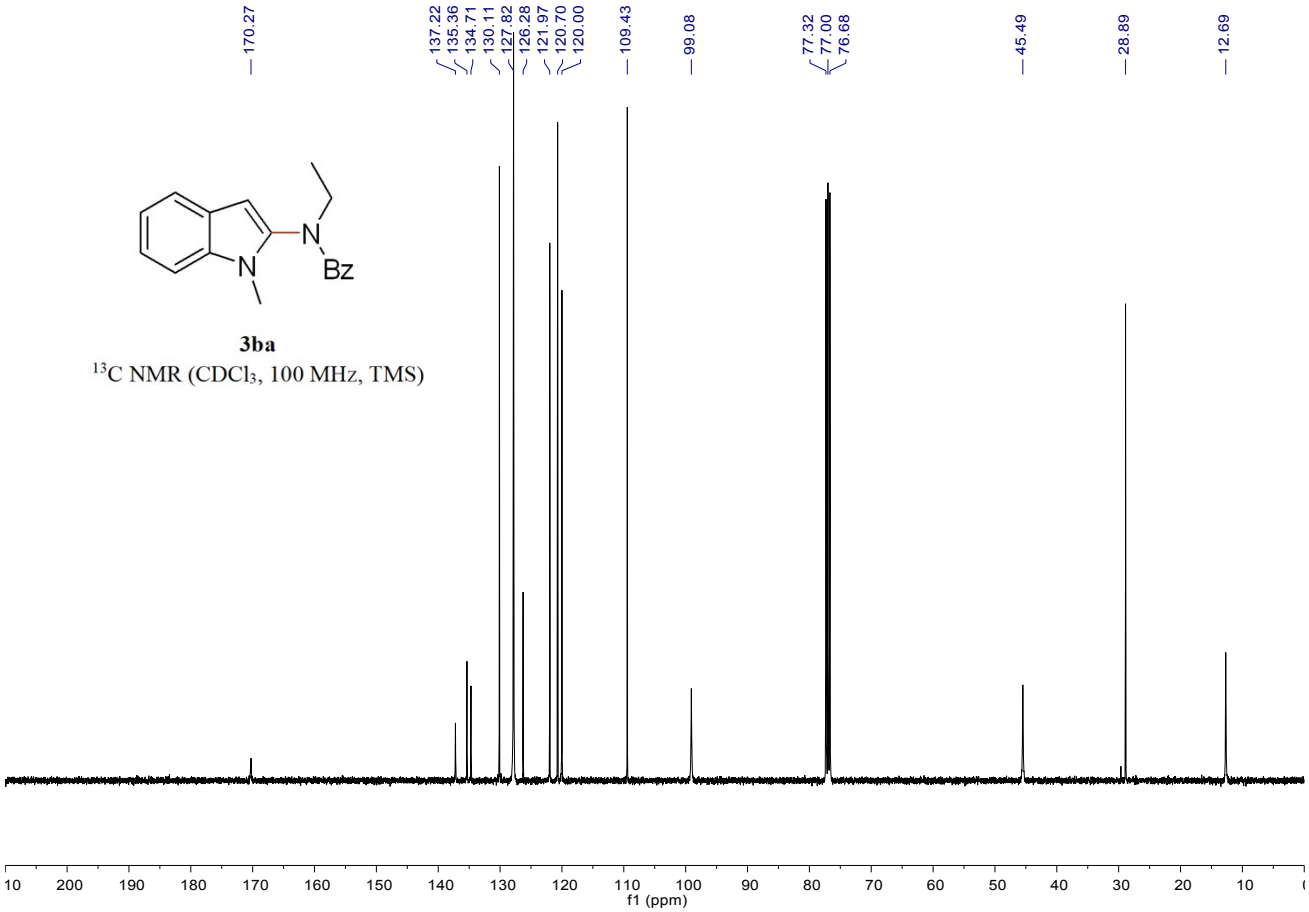


***N*-ethyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3ba)**. The title product was obtained after purification by column chromatography (PE:EA = 10:1). A brown solid, 45 mg, 81% yield; M.p.: 106 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.6 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 2H), 7.23 - 7.16 (m, 3H), 7.16 - 7.04 (m, 3H), 6.26 (s, 1H), 4.23 - 4.13 (m, 1H), 3.65 (dq, *J* = 14.1, 7.2 Hz, 1H), 3.46 (s, 3H), 1.26 (t, *J* = 7.2 Hz, 4H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.3, 137.2, 135.4, 134.7, 130.1, 127.8, 126.3, 122.0, 120.7, 120.0, 109.4, 99.1, 45.5, 28.9, 12.7; IR (neat) ν 2969, 2930, 1645, 1556, 1401, 1299, 1095, 870, 774, 750, 721, 699, 668 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1311.



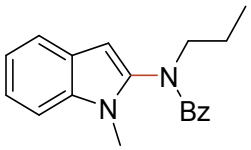
3ba

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

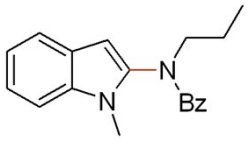


3ba

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

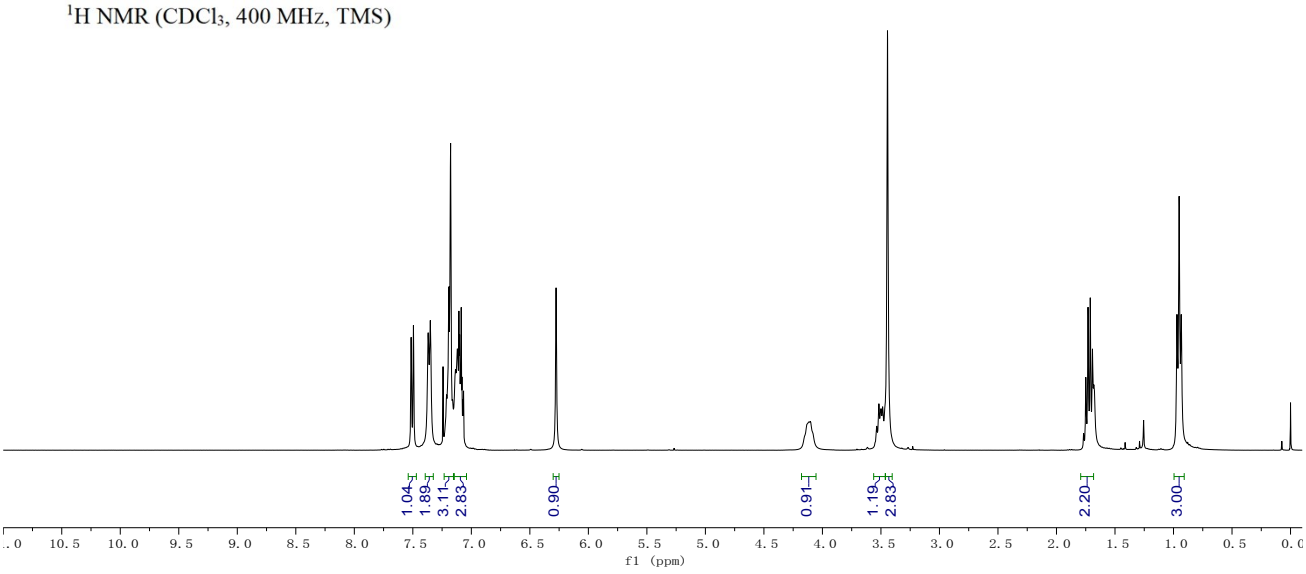


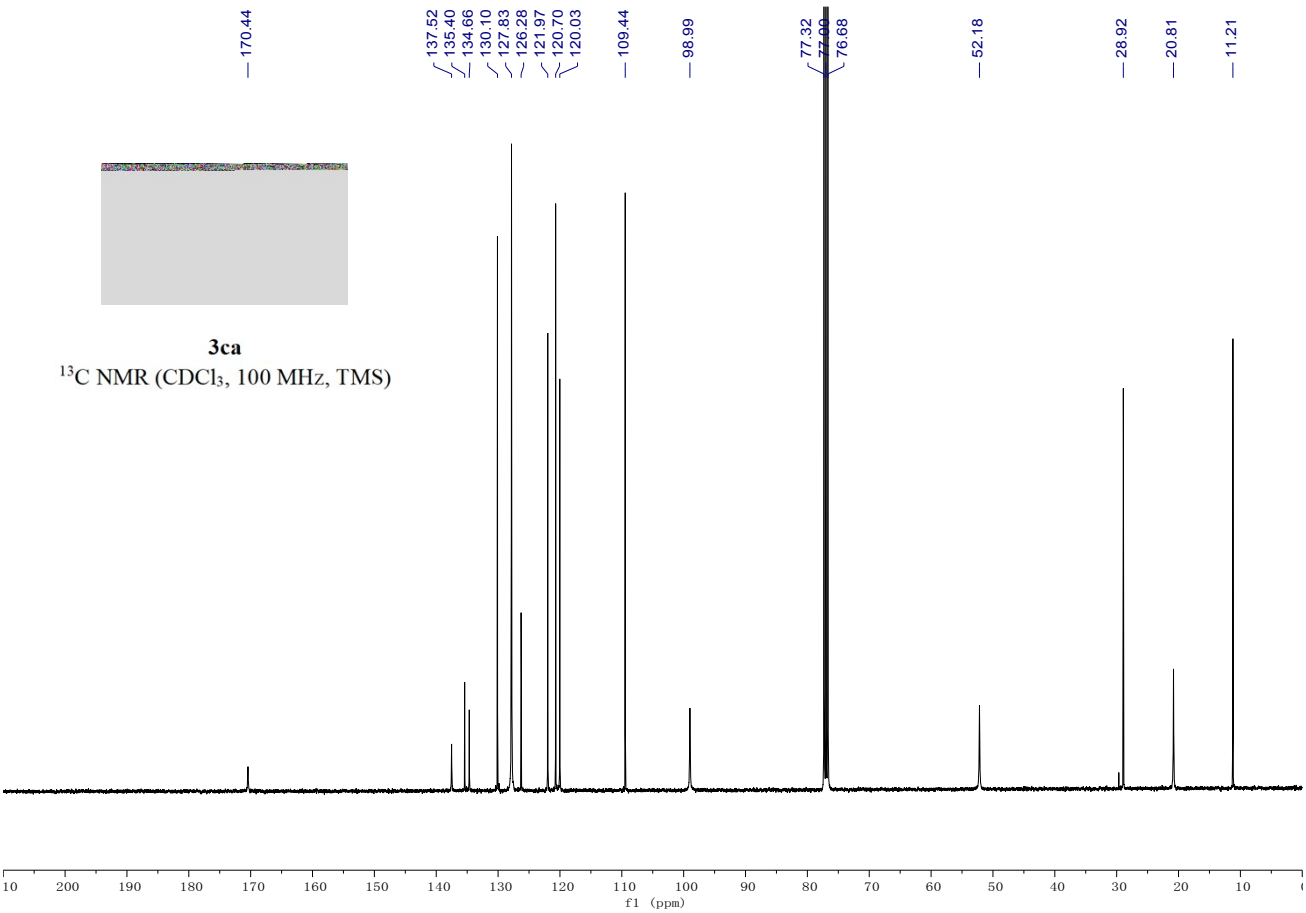
***N*-(1-methyl-1H-indol-2-yl)-*N*-propylbenzamide (3ca)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 28 mg, 48% yield; M.p.: 89 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.6 Hz, 1H), 7.36 (d, *J* = 7.6 Hz, 2H), 7.23 - 7.15 (m, 3H), 7.15 - 7.04 (m, 3H), 6.28 (s, 1H), 4.18 - 4.05 (m, 1H), 3.56 - 3.46 (m, 1H), 3.44 (s, 3H), 1.79 - 1.68 (m, 1H), 0.95 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.4, 137.5, 135.4, 134.7, 130.1, 127.8, 126.3, 122.0, 120.7, 120.0, 109.4, 99.0, 52.2, 28.9, 20.8, 11.2; IR (neat) ν 3055, 2961, 2927, 1645, 1556, 1435, 1399, 1338, 1314, 1110, 934, 780, 718, 700, 666 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{20}N_2ONa^+$ Requires: 315.1468, Found: 315.1466.

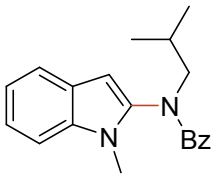


3ca

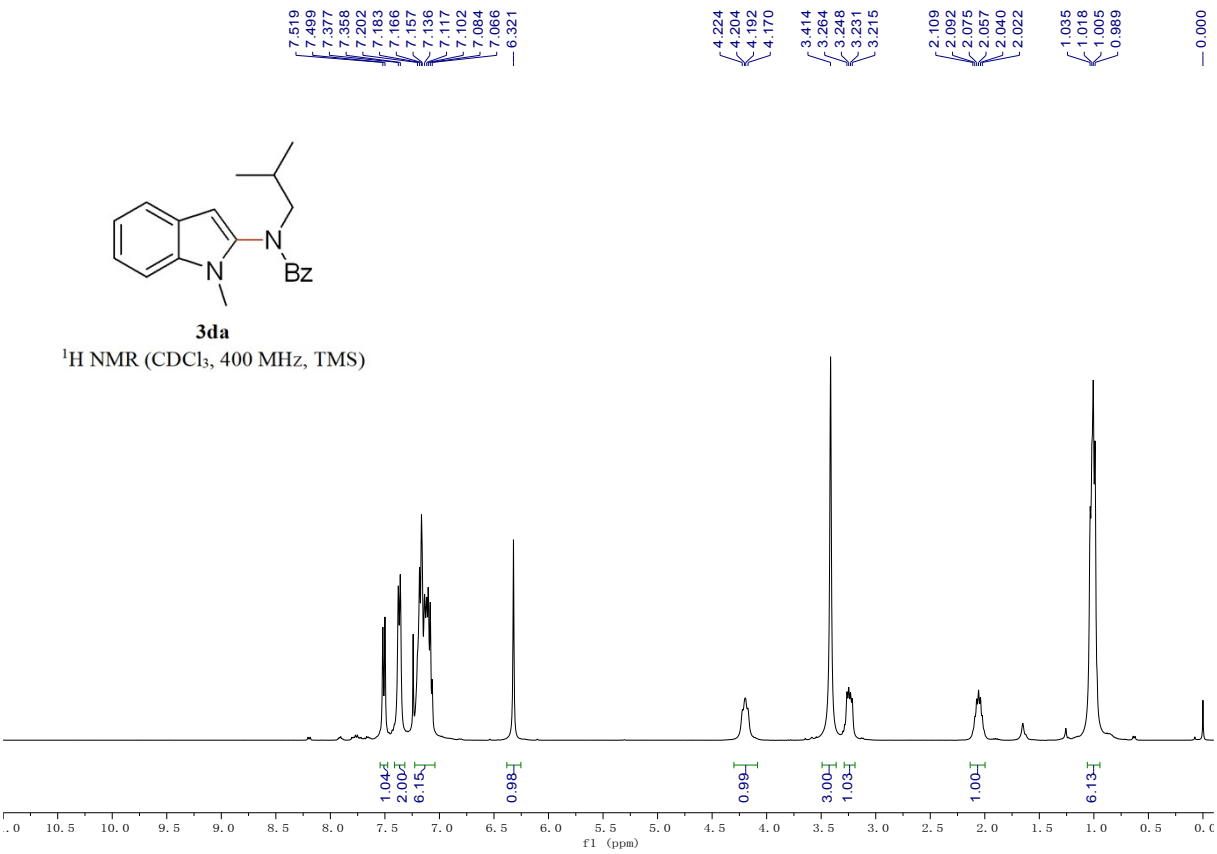
^{1}H NMR ($CDCl_3$, 400 MHz, TMS)





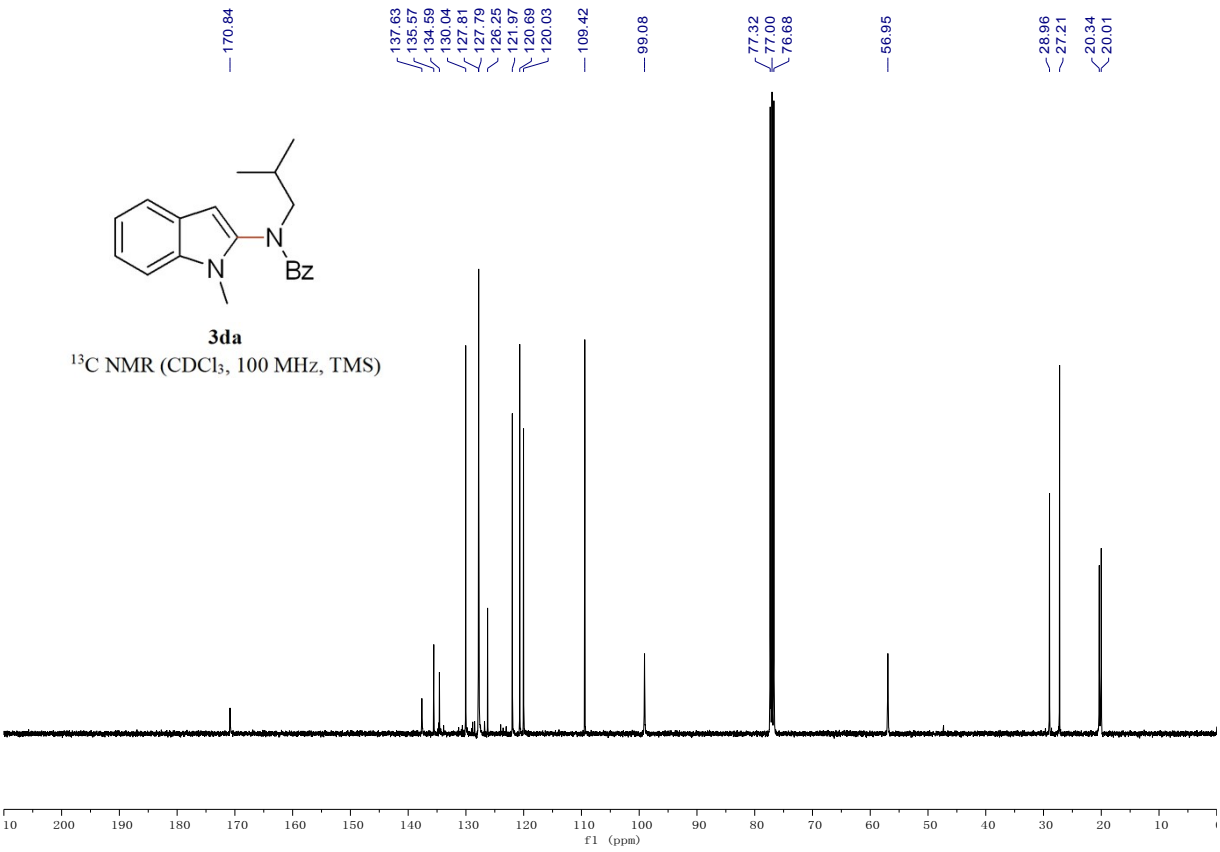


N-isobutyl-N-(1-methyl-1H-indol-2-yl)benzamide (3da). The title product was obtained after purification by prep-TLC (DCM). A yellow solid, 23 mg, 37% yield; M.p.: 85 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.51 (d, *J* = 8.0 Hz, 1H), 7.37 (d, *J* = 7.6 Hz, 2H), 7.23 - 7.04 (m, 6H), 6.32 (s, 1H), 4.20 (dd, *J* = 13.2, 8.0 Hz, 1H), 3.41 (s, 3H), 3.24 (dd, *J* = 13.2, 6.4 Hz, 1H), 2.13 - 2.00 (m, 1H), 1.01 (dd, *J* = 11.8, 6.7 Hz, 6H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.8, 137.6, 135.6, 134.6, 130.0, 127.81, 127.79, 126.3, 122.0, 120.7, 120.0, 109.4, 99.1, 56.9, 29.0, 27.2, 20.3, 20.0; IR (neat) ν 2958, 2924, 1655, 1557, 1463, 1399, 1318, 1281, 1114, 1019, 936, 777, 725, 698, 668 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{22}N_2ONa^+$ Requires: 329.1624, Found: 329.1621.



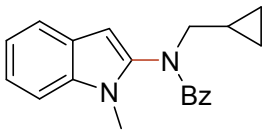
3da

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

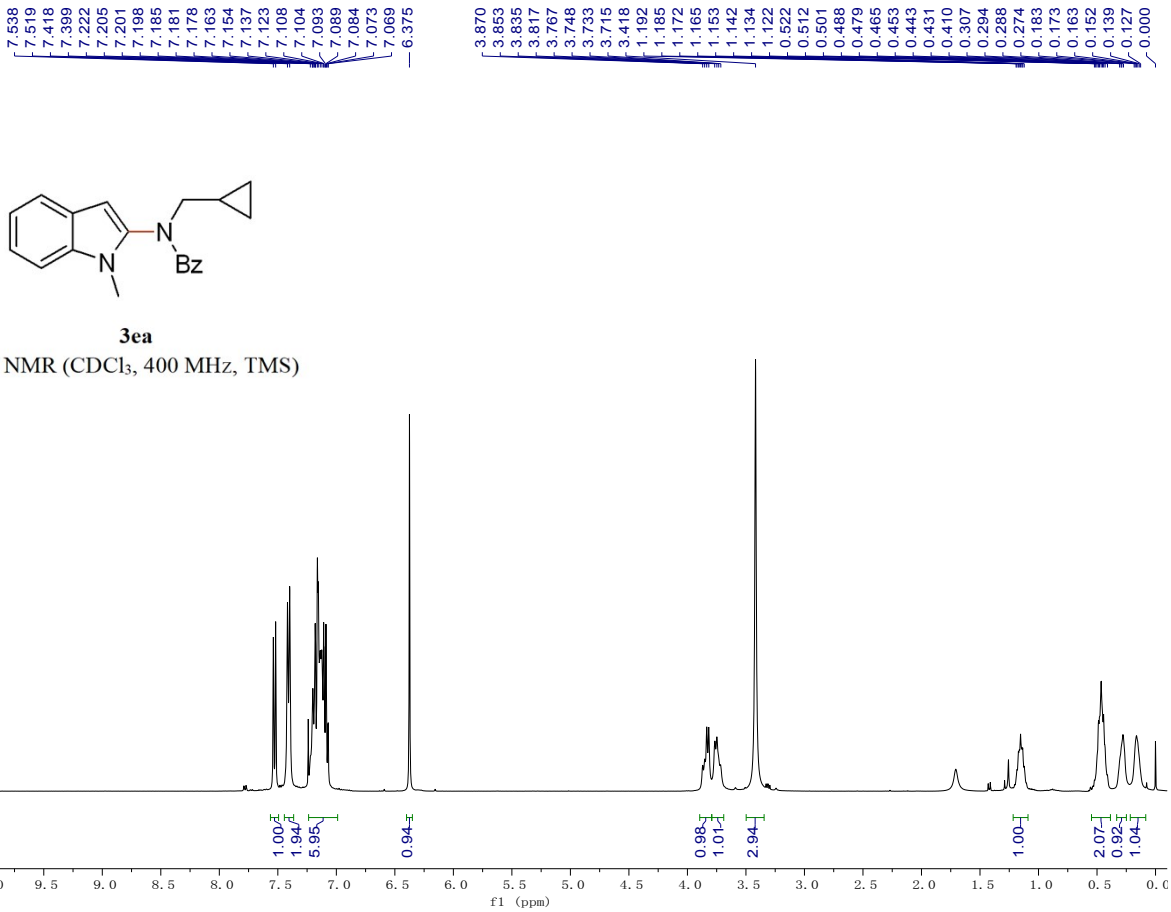


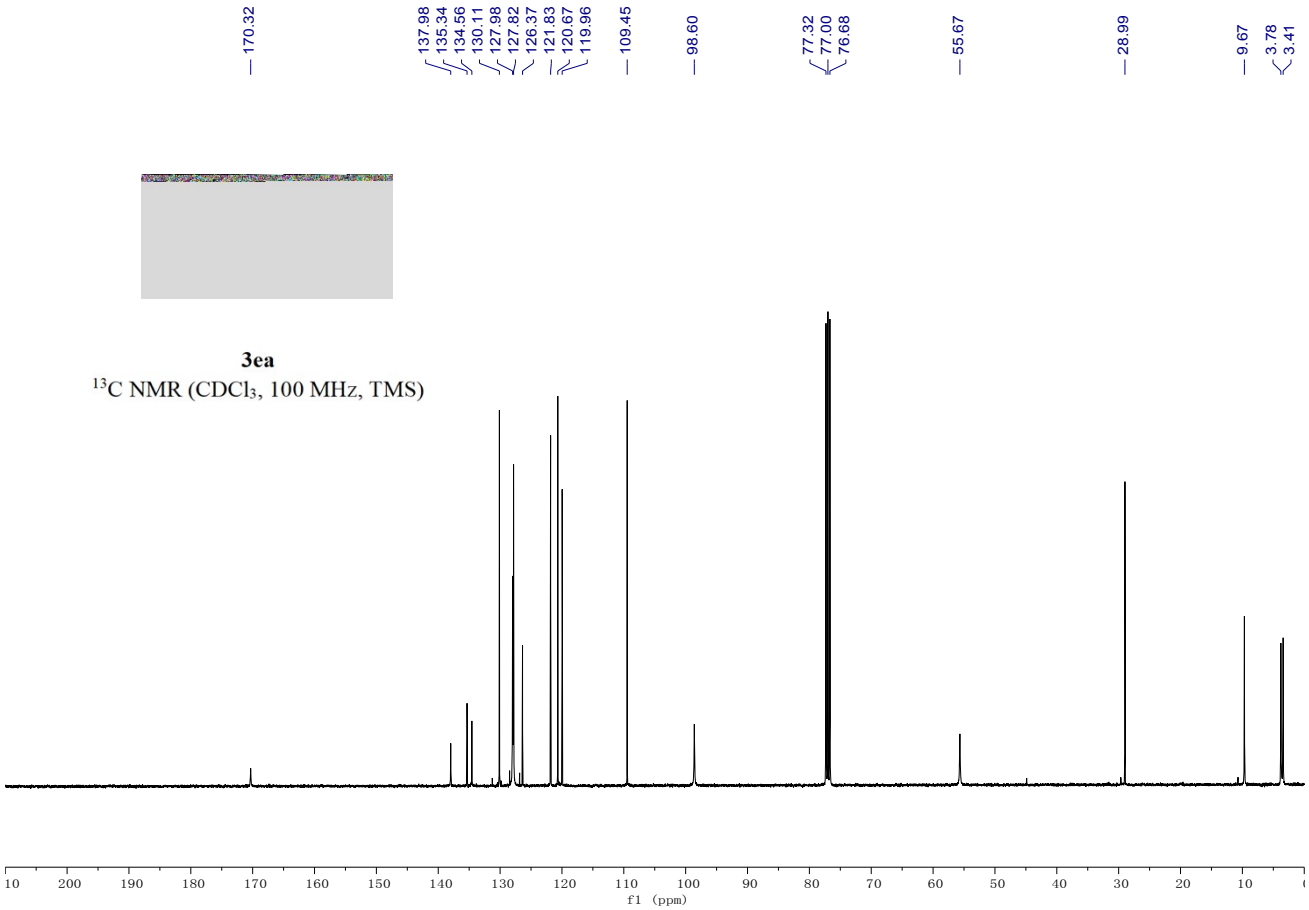
3da

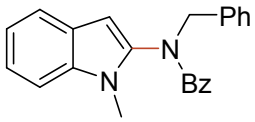
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



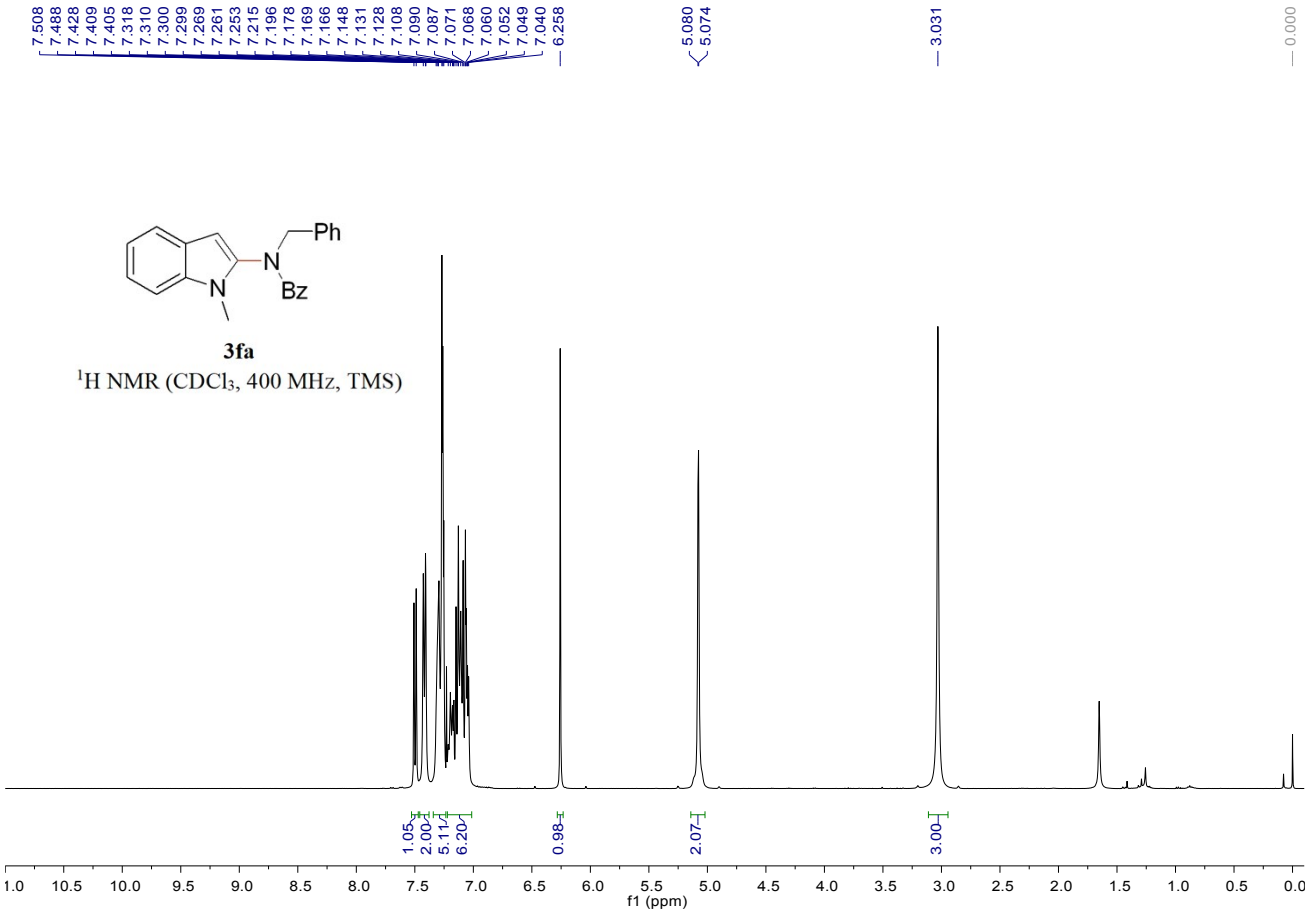
***N*-(cyclopropylmethyl)-*N*-(1-methyl-1H-indol-2-yl)benzamide (3ea)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 31 mg, 50% yield; M.p.: 92 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.53 (d, *J* = 7.8 Hz, 1H), 7.41 (d, *J* = 7.6 Hz, 2H), 7.25 - 6.99 (m, 6H), 6.38 (s, 1H), 3.89 - 3.79 (m, 1H), 3.79 - 3.69 (m, 1H), 3.42 (s, 3H), 1.23 - 1.08 (m, 1H), 0.55 - 0.39 (m, 2H), 0.33 - 0.22 (m, 1H), 0.23 - 0.10 (m, 1H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.3, 138.0, 135.3, 134.6, 130.1, 128.0, 127.8, 126.4, 121.8, 120.7, 120.0, 109.5, 98.6, 55.7, 29.0, 9.7, 3.8, 3.4; IR (neat) ν 3013, 2932, 1645, 1557, 1474, 1446, 1397, 1375, 1316, 1277, 1123, 1021, 949, 777, 744, 718, 693, 667 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{20}N_2ONa^+$ Requires: 327.1468, Found: 327.1467.





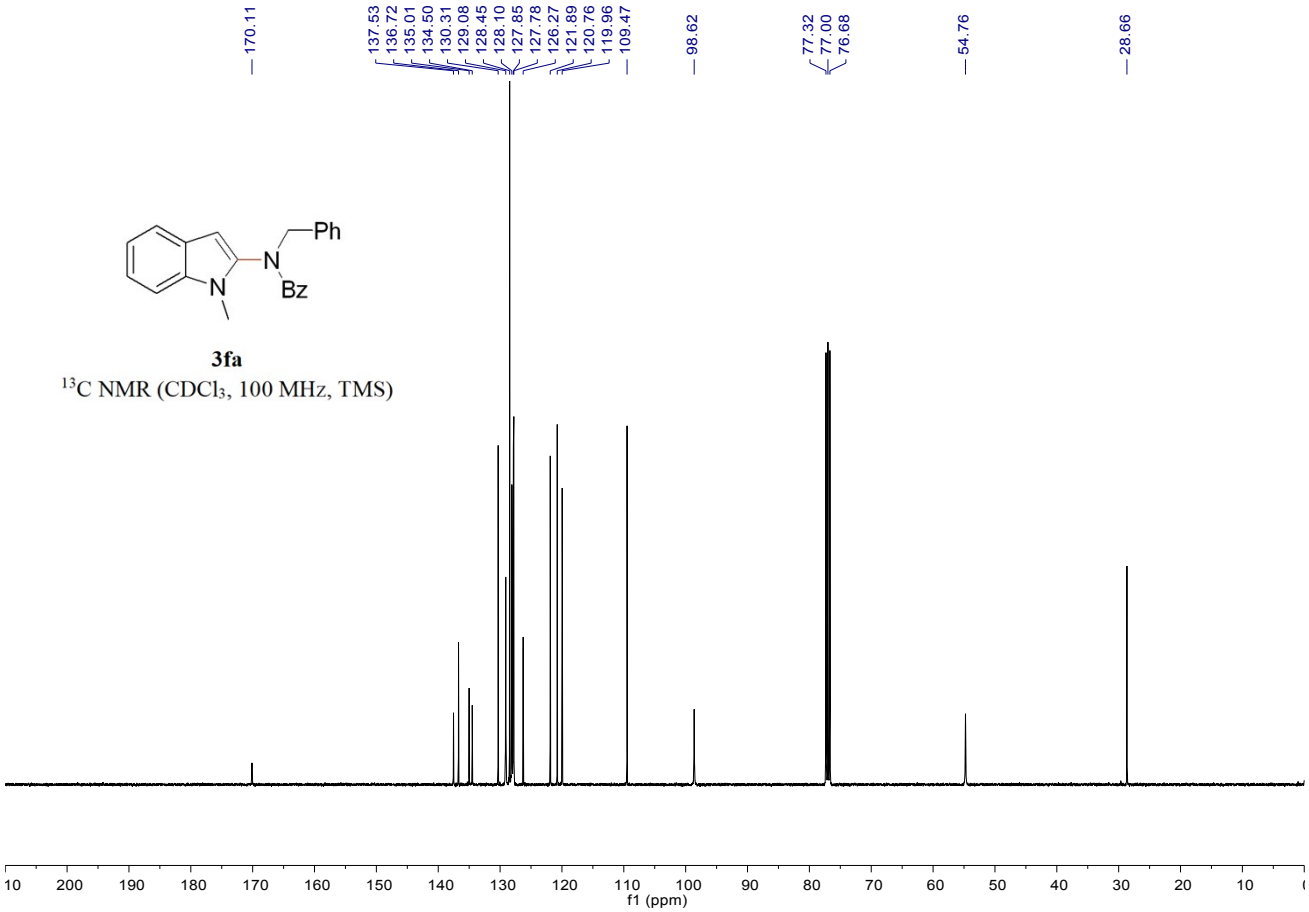


***N*-benzyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3fa)**. The title product was obtained after purification by prep-TLC (PE:EA = 4:1). A colorless oil, 46 mg, 67% yield; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.8 Hz, 1H), 7.46 - 7.38 (m, 2H), 7.34 - 7.23 (m, 5H), 7.22 - 7.01 (m, 6H), 6.26 (s, 1H), 5.08 (d, *J* = 2.4 Hz, 2H), 3.03 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.1, 137.5, 136.7, 135.0, 134.5, 130.3, 129.1, 128.5, 128.1, 127.9, 127.8, 126.3, 121.9, 120.8, 120.0, 109.5, 98.6, 54.8, 28.7; IR (neat) ν 3058, 2938, 1651, 1549, 1433, 1376, 1309, 1263, 1077, 779, 696, 666 cm^{-1}; HRMS (ESI) Calcd. for $C_{23}H_{20}N_2ONa^+$ Requires: 363.1468, Found: 363.1471.



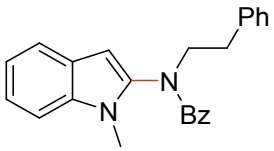
3fa

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

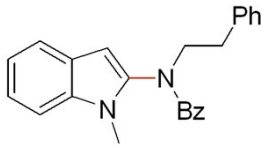


3fa

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

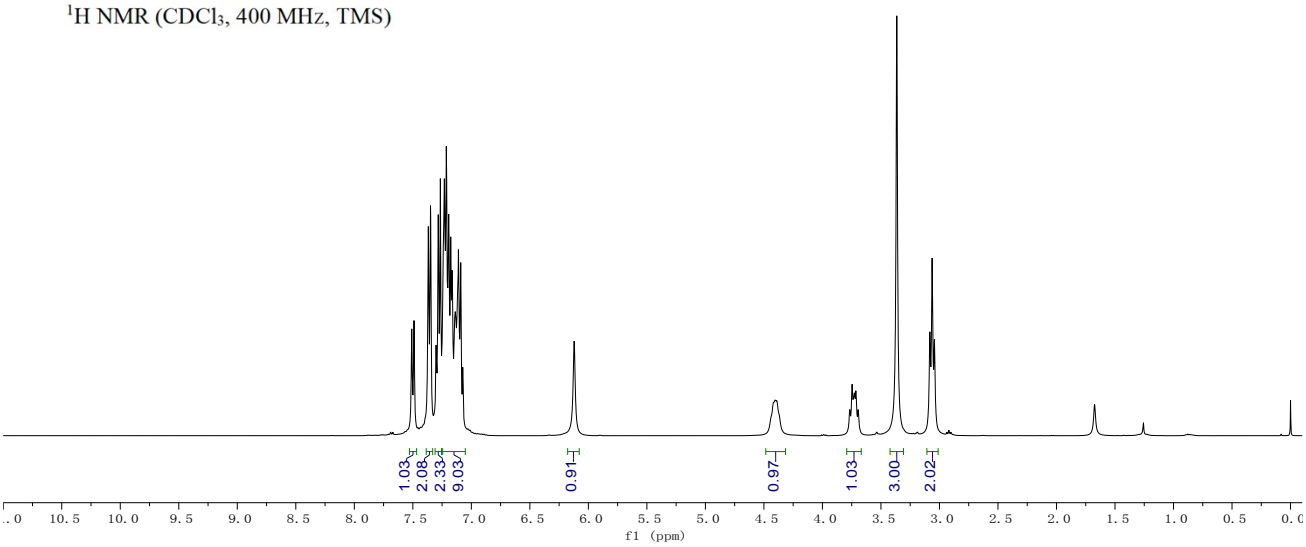


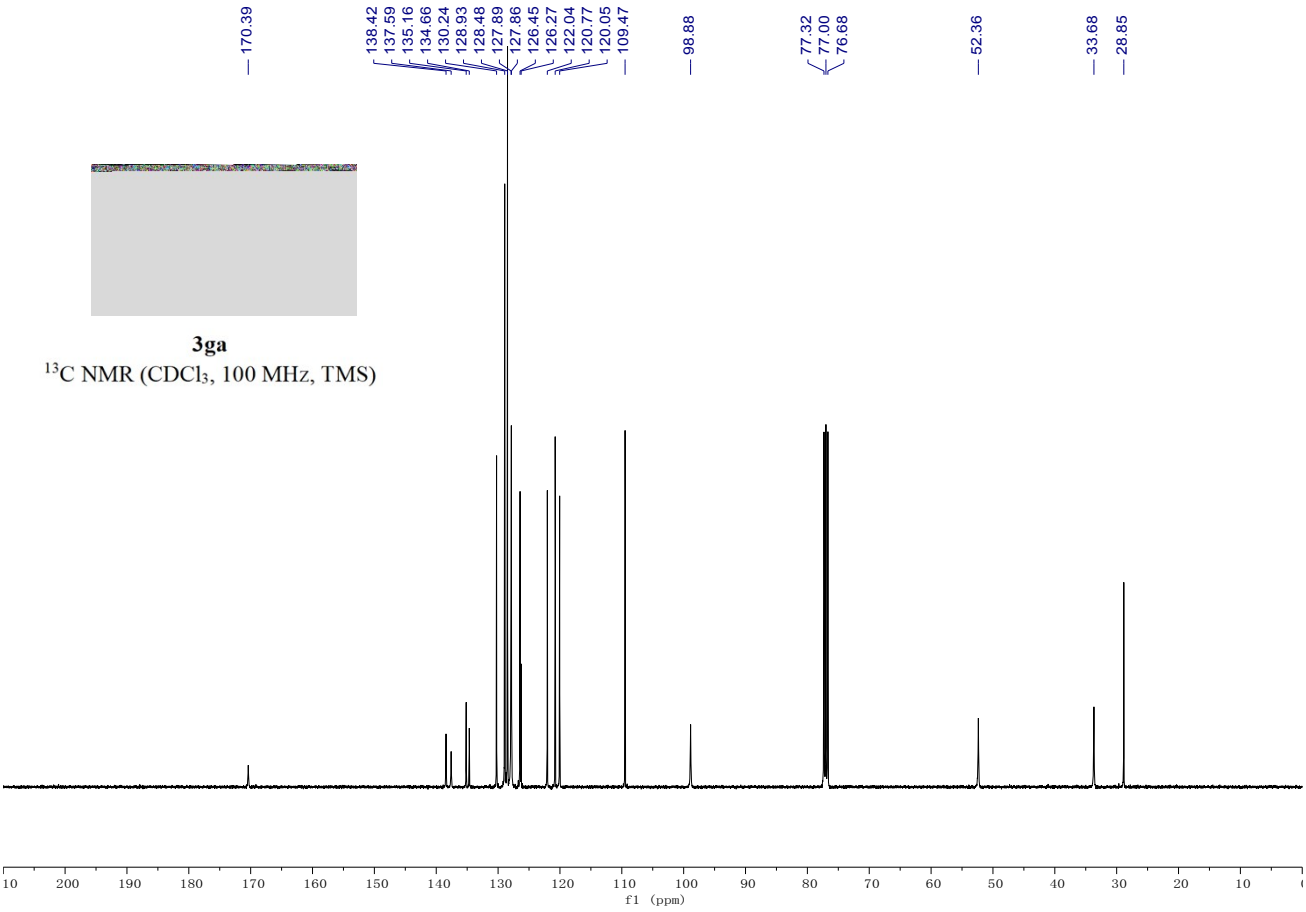
N-(1-methyl-1H-indol-2-yl)-N-phenethylbenzamide (3ga). The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 45 mg, 63% yield; M.p.: 123 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.8 Hz, 1H), 7.36 (d, *J* = 7.7 Hz, 2H), 7.31 - 7.26 (m, 2H), 7.24 - 7.05 (m, 9H), 6.12 (s, 1H), 4.48 - 4.31 (m, 1H), 3.73 (dt, *J* = 13.1, 8.1 Hz, 1H), 3.36 (s, 3H), 3.06 (t, *J* = 8.0 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.4, 138.4, 137.6, 135.2, 134.7, 130.2, 128.9, 128.5, 127.9, 127.9, 126.4, 126.3, 122.0, 120.8, 120.0, 109.5, 98.9, 52.4, 33.7, 28.8; IR (neat) ν 2936, 1647, 1553, 1448, 1395, 1333, 1295, 1140, 1005, 780, 752, 720, 699, 667 cm^{-1}; HRMS (ESI) Calcd. for $C_{24}H_{22}N_2ONa^+$ Requires: 377.1624, Found: 377.1621.

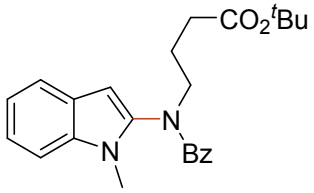


3ga

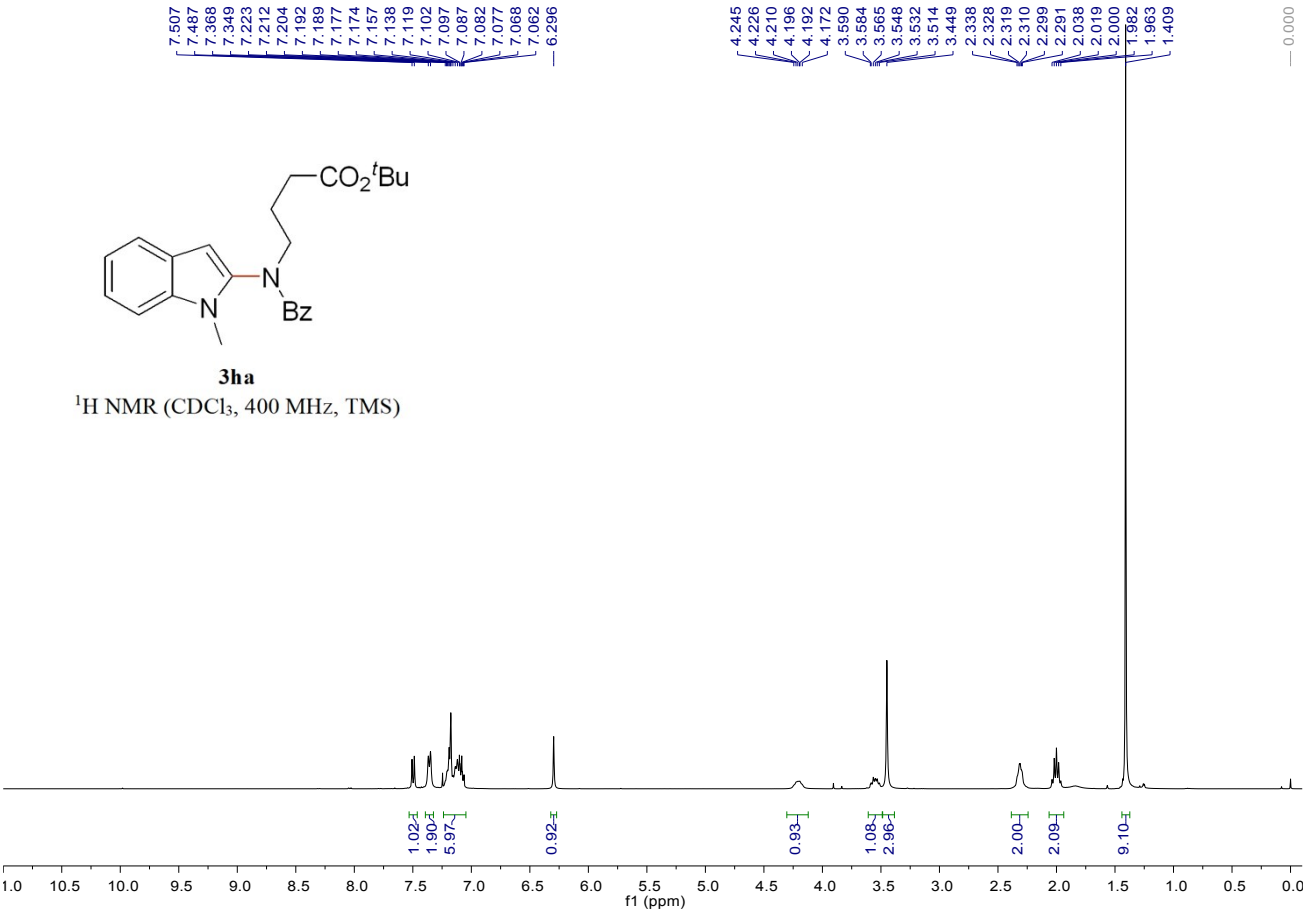
^{1}H NMR ($CDCl_3$, 400 MHz, TMS)





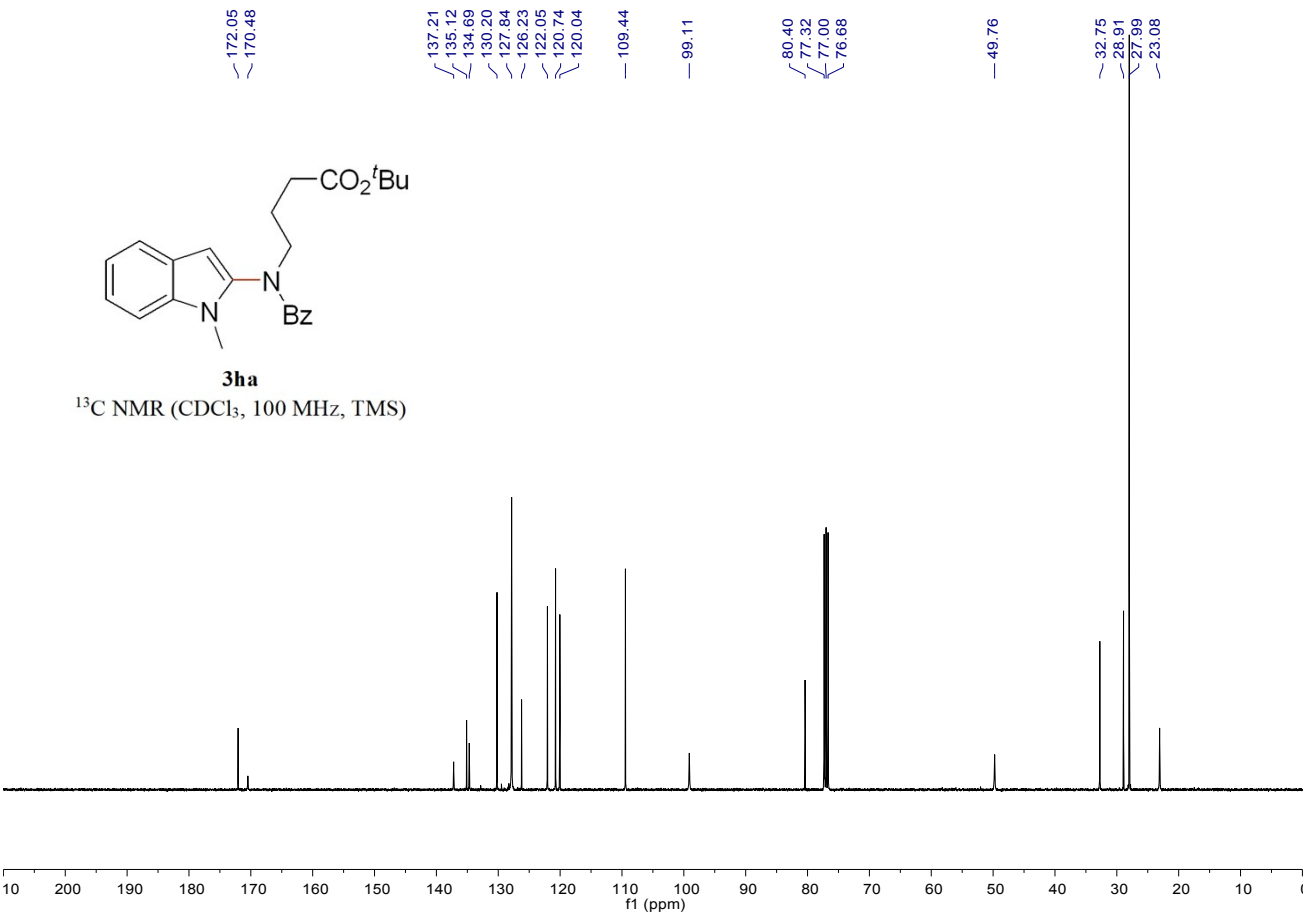


***tert*-butyl 4-(*N*-(1-methyl-1H-indol-2-yl)benzamido)butanoate (3ha)**. The title product was obtained after purification by prep-TLC (PE:EA = 4:1). A yellow solid, 64 mg, 81% yield; M.p.: 73 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.9 Hz, 1H), 7.36 (d, *J* = 7.6 Hz, 2H), 7.24 - 7.05 (m, 6H), 6.30 (s, 1H), 4.30 - 4.12 (m, 1H), 3.61 - 3.49 (m, 1H), 3.45 (s, 3H), 2.39 - 2.24 (m, 2H), 2.00 (qu, *J* = 7.5 Hz, 2H), 1.41 (s, 9H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 172.1, 170.5, 137.2, 135.1, 134.7, 130.2, 127.8, 126.2, 122.1, 120.7, 120.0, 109.4, 99.1, 80.4, 49.8, 32.7, 28.9, 28.0, 23.1; IR (neat) ν 2978, 2932, 1723, 1644, 1560, 1447, 1399, 1316, 1271, 1154, 782, 739, 719, 669 cm^{-1}; HRMS (ESI) Calcd. for $C_{24}H_{28}N_2O_3Na^+$ Requires: 415.1992, Found: 415.1992.



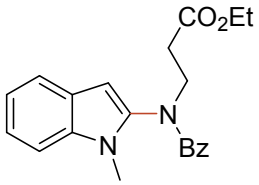
3ha

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

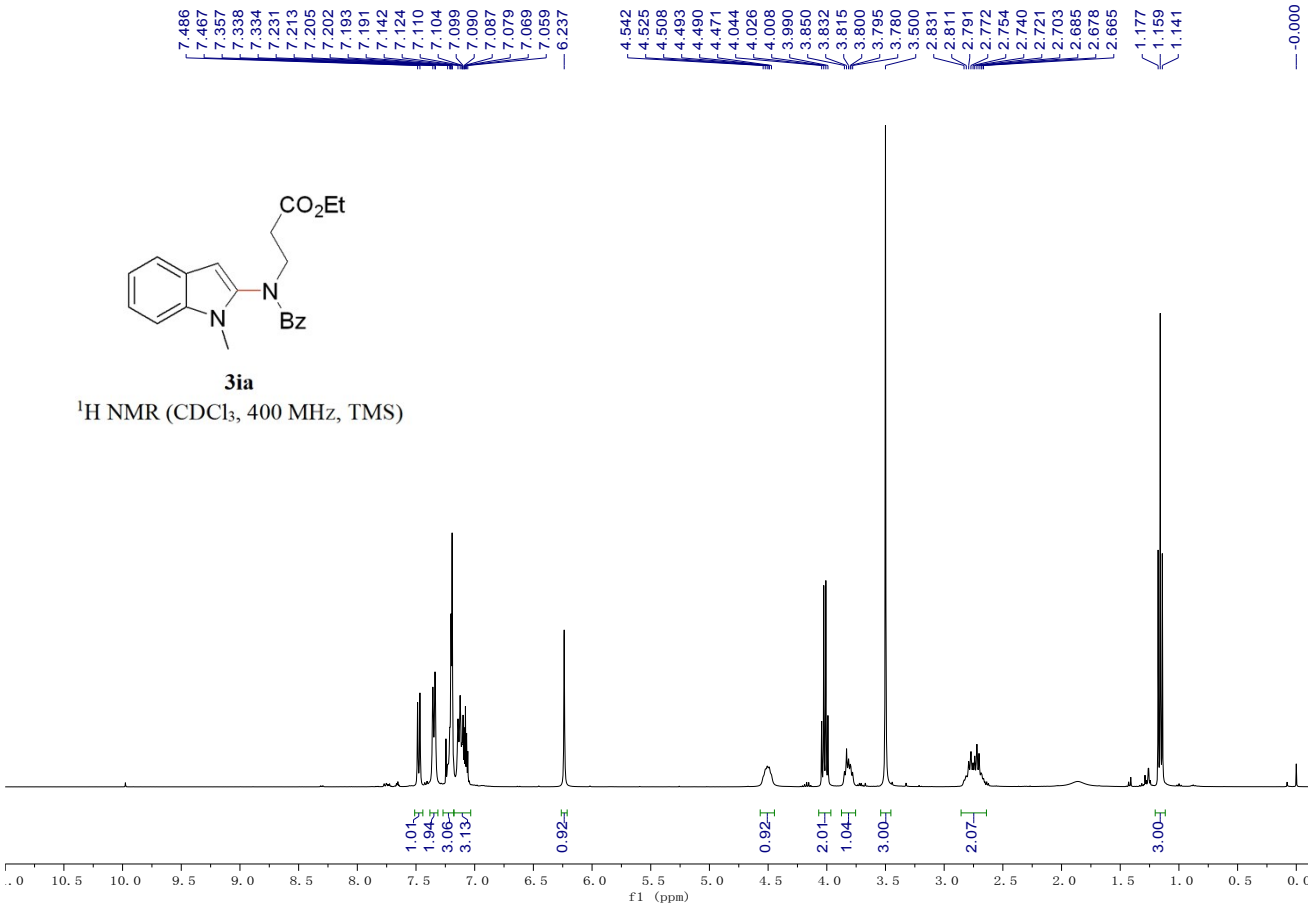


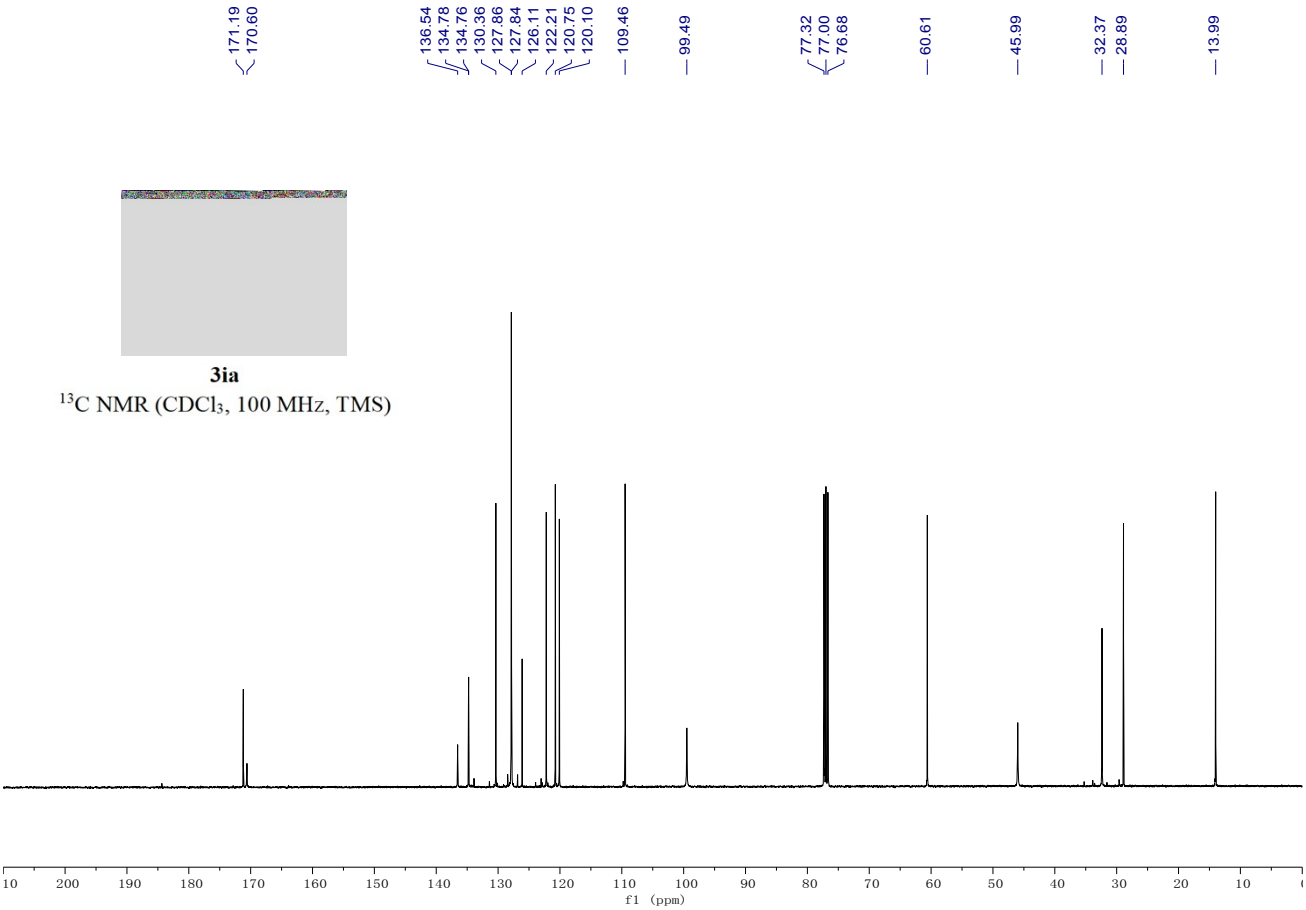
3ha

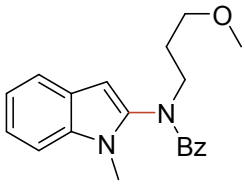
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



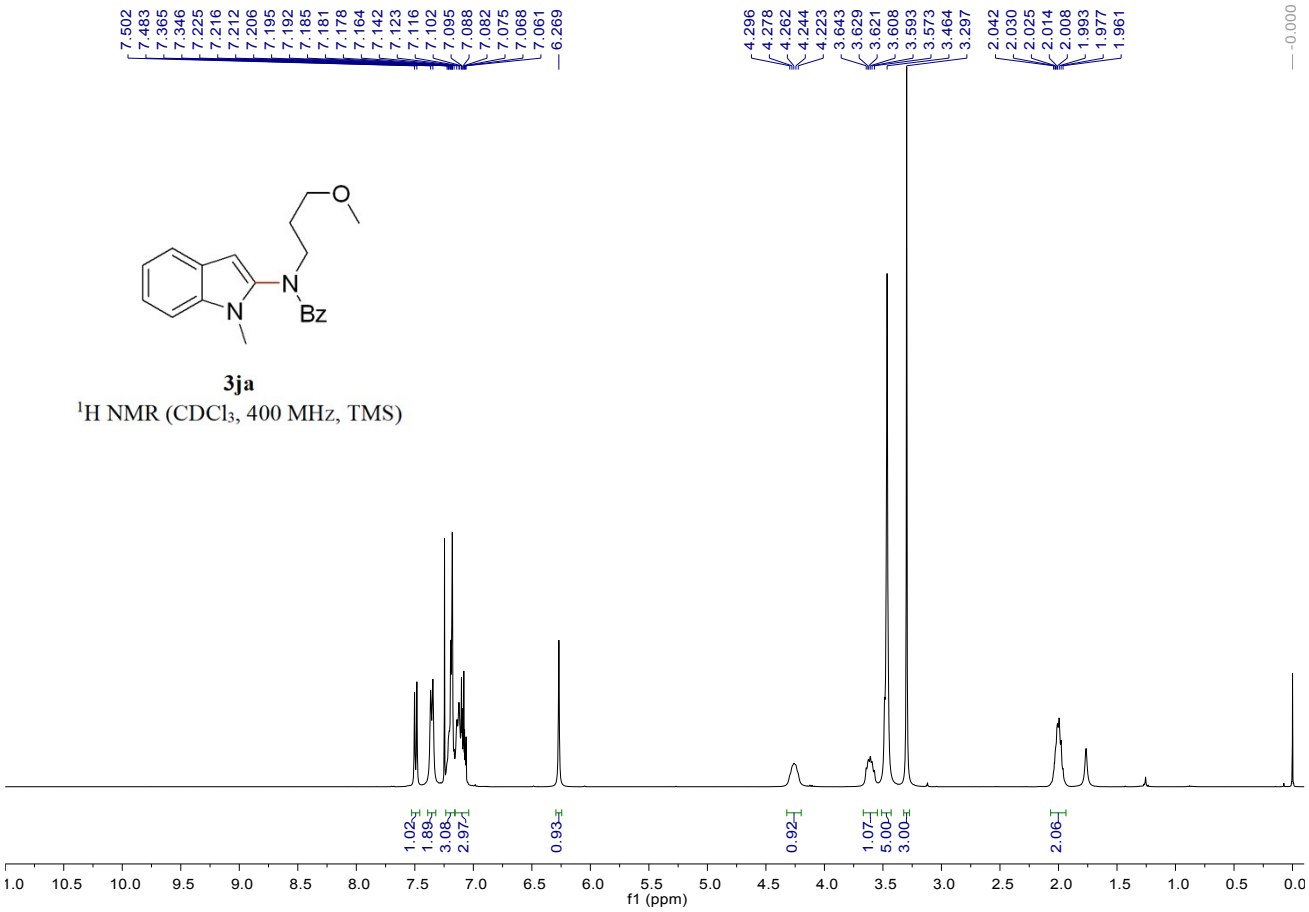
ethyl 3-(*N*-(1-methyl-1H-indol-2-yl)benzamido)propanoate (3ia). The title product was obtained after purification by prep-TLC (DCM). A yellow oil, 53 mg, 75% yield; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.48 (d, *J* = 7.9 Hz, 1H), 7.35 (d, *J* = 7.3 Hz, 2H), 7.27 - 7.18 (m, 3H), 7.18 - 7.03 (m, 3H), 6.24 (s, 1H), 4.57 - 4.45 (m, 1H), 4.02 (q, *J* = 7.1 Hz, 2H), 3.87 - 3.75 (m, 1H), 3.50 (s, 3H), 2.86 - 2.64 (m, 2H), 1.16 (t, *J* = 7.1 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.2, 170.6, 136.5, 134.78, 134.76, 130.4, 127.9, 127.8, 126.1, 122.2, 120.7, 120.1, 109.5, 99.5, 60.6, 46.0, 32.4, 28.9, 14.0; IR (neat) ν 2979, 1728, 1652, 1552, 1435, 1337, 1313, 1180, 1141, 1029, 781, 750, 715, 696, 667 cm^{-1}; HRMS (ESI) Calcd. for $C_{21}H_{22}N_2O_3Na^+$ Requires: 373.1523, Found: 373.1517.





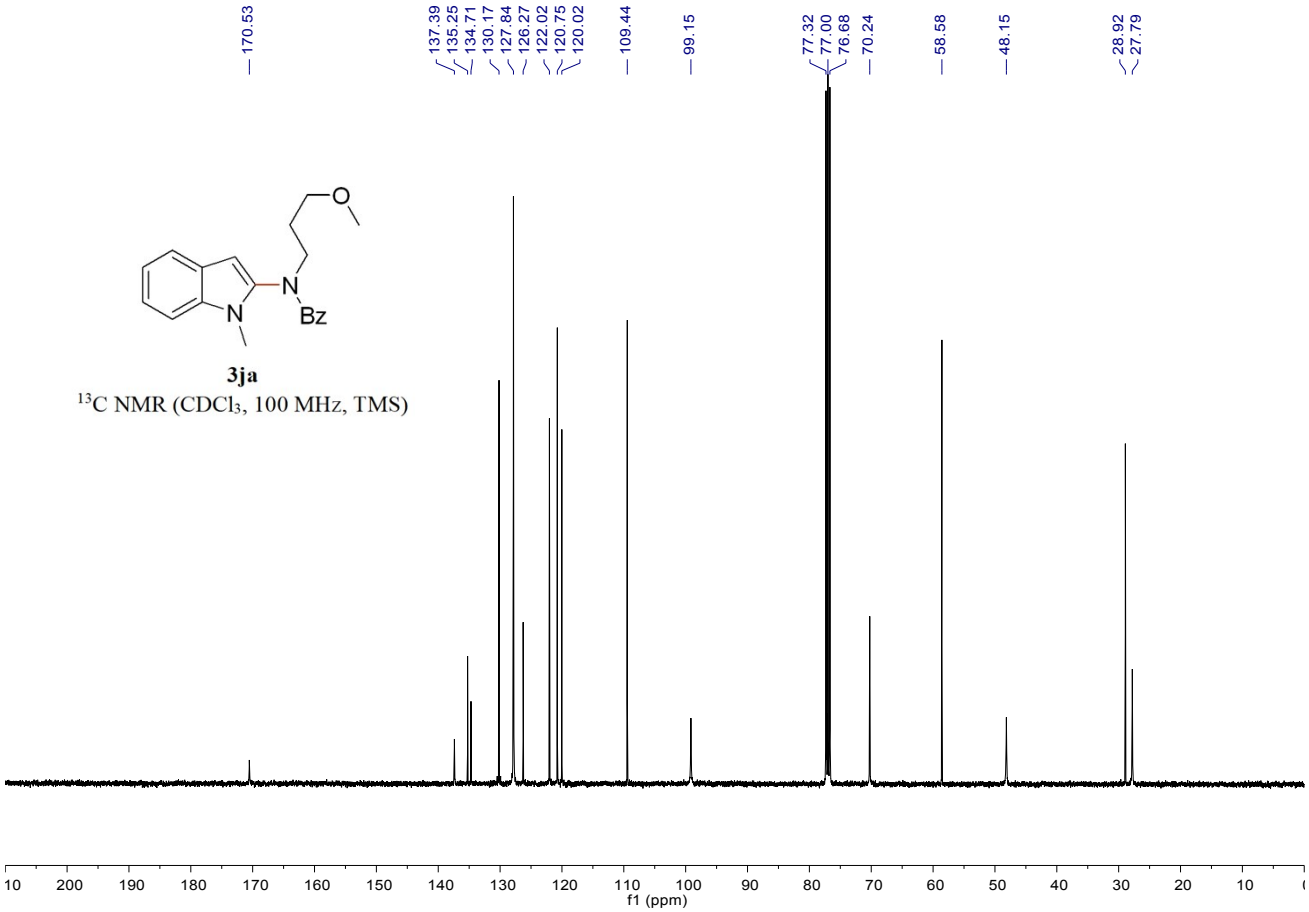


***N*-(3-methoxypropyl)-*N*-(1-methyl-1H-indol-2-yl)benzamide (3ja)**. The title product was obtained after purification by prep-TLC (PE:EA = 2:1). An off-white solid, 37 mg, 58% yield; M.p.: 106-107 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.49 (d, *J* = 7.8 Hz, 1H), 7.36 (d, *J* = 7.6 Hz, 2H), 7.24 - 7.16 (m, 3H), 7.16 - 7.04 (m, 3H), 6.27 (s, 1H), 4.32 - 4.20 (m, 1H), 3.67 - 3.55 (m, 1H), 3.46 (s, 5H), 3.30 (s, 3H), 2.07 - 1.94 (m, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.5, 137.4, 135.3, 134.7, 130.2, 127.8, 126.3, 122.0, 120.7, 120.0, 109.4, 99.2, 70.2, 58.6, 48.2, 28.9, 27.8; IR (neat) ν 2981, 2951, 2890, 1650, 1557, 1448, 1353, 1300, 1268, 1115, 925, 777, 739, 718, 695, 670 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{22}N_2O_2Na^+$ Requires: 345.1574, Found: 345.1567.



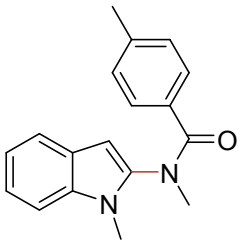
3ja

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

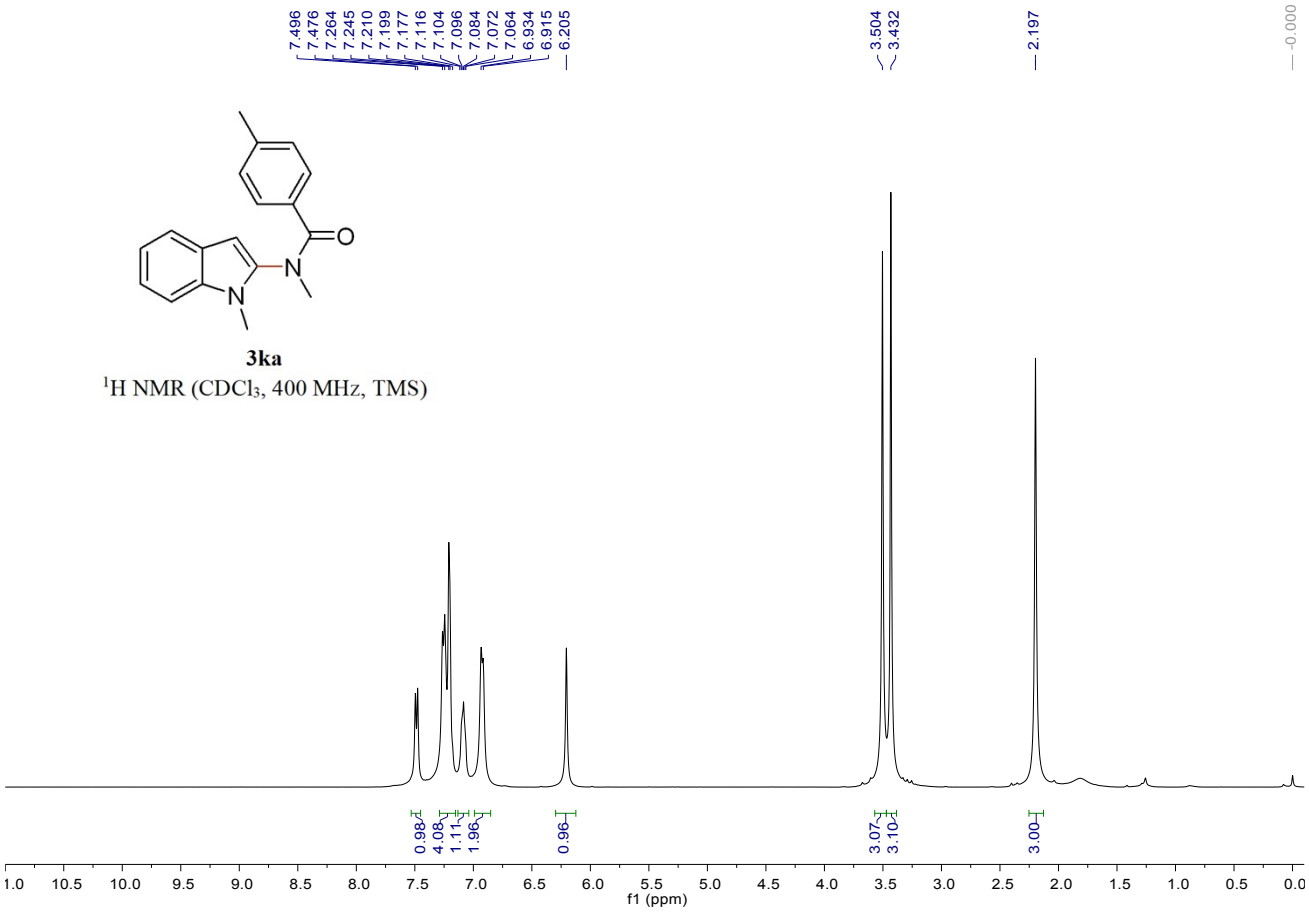


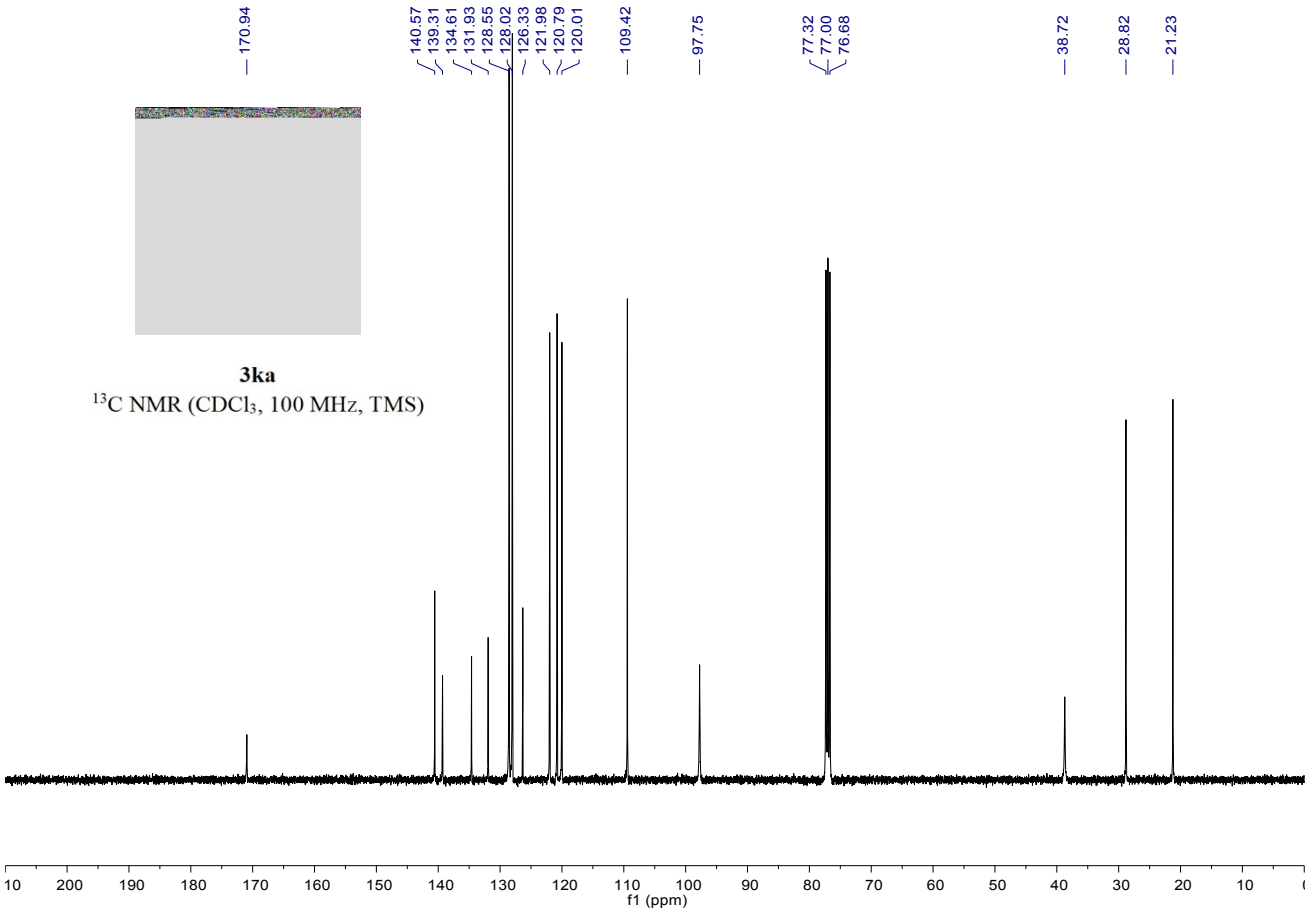
3ja

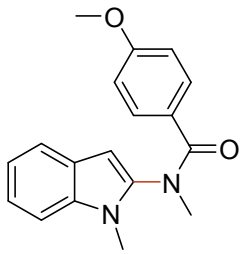
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



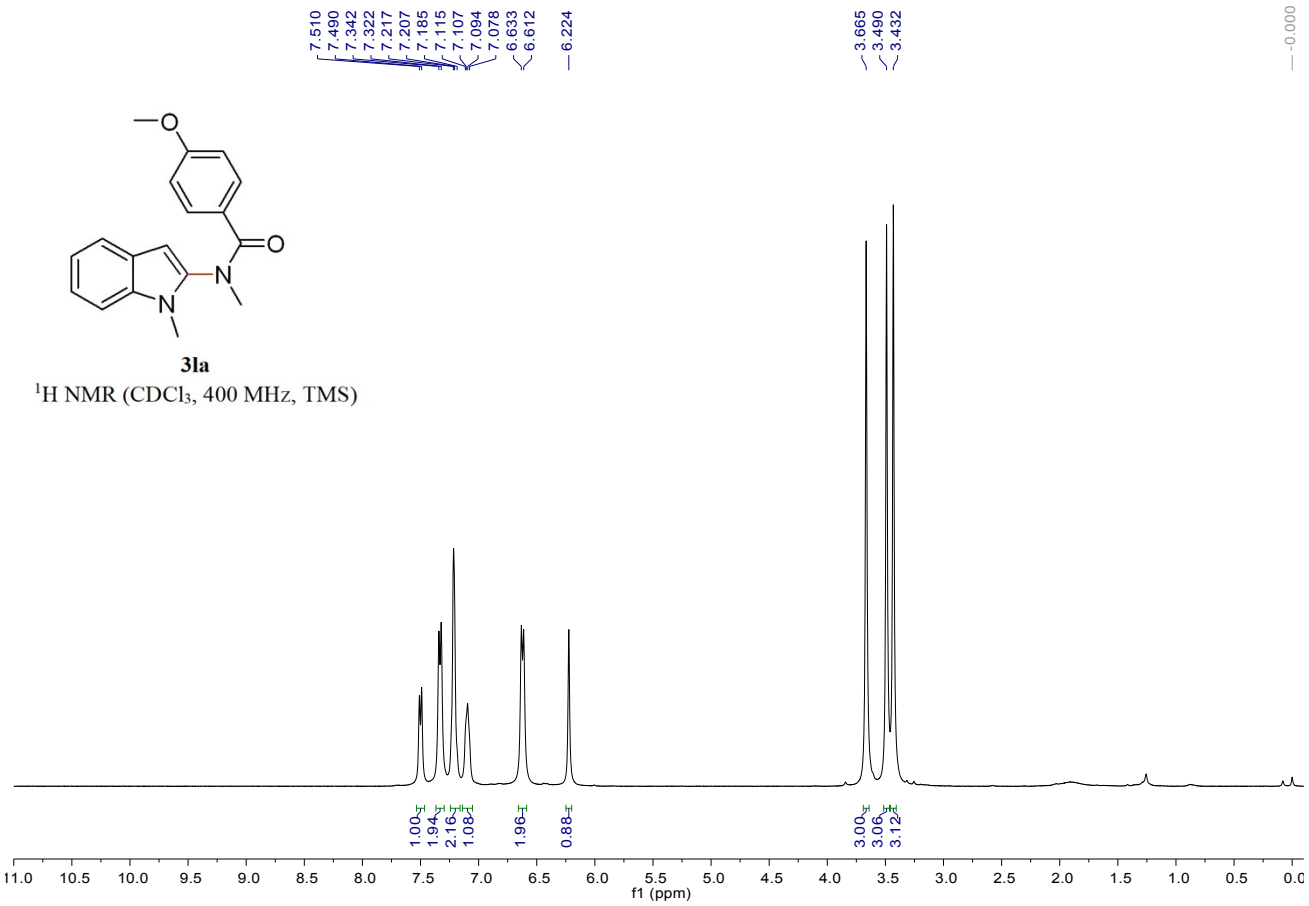
***N*,4-dimethyl-N-(1-methyl-1H-indol-2-yl)benzamide (3ka)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 35 mg, 62% yield; M.p.: 143 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.49 (d, *J* = 7.9 Hz, 1H), 7.29 - 7.15 (m, 4H), 7.13 - 7.04 (m, 1H), 6.92 (d, *J* = 7.8 Hz, 2H), 6.21 (s, 1H), 3.50 (s, 3H), 3.43 (s, 3H), 2.20 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 140.6, 139.3, 134.6, 131.9, 128.5, 128.0, 126.3, 122.0, 120.8, 120.0, 109.4, 97.7, 38.7, 28.8, 21.2; IR (neat) ν 3053, 1648, 1559, 1476, 1462, 1390, 1335, 1314, 1256, 1182, 1088, 1010, 835, 776, 736, 680 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1314.





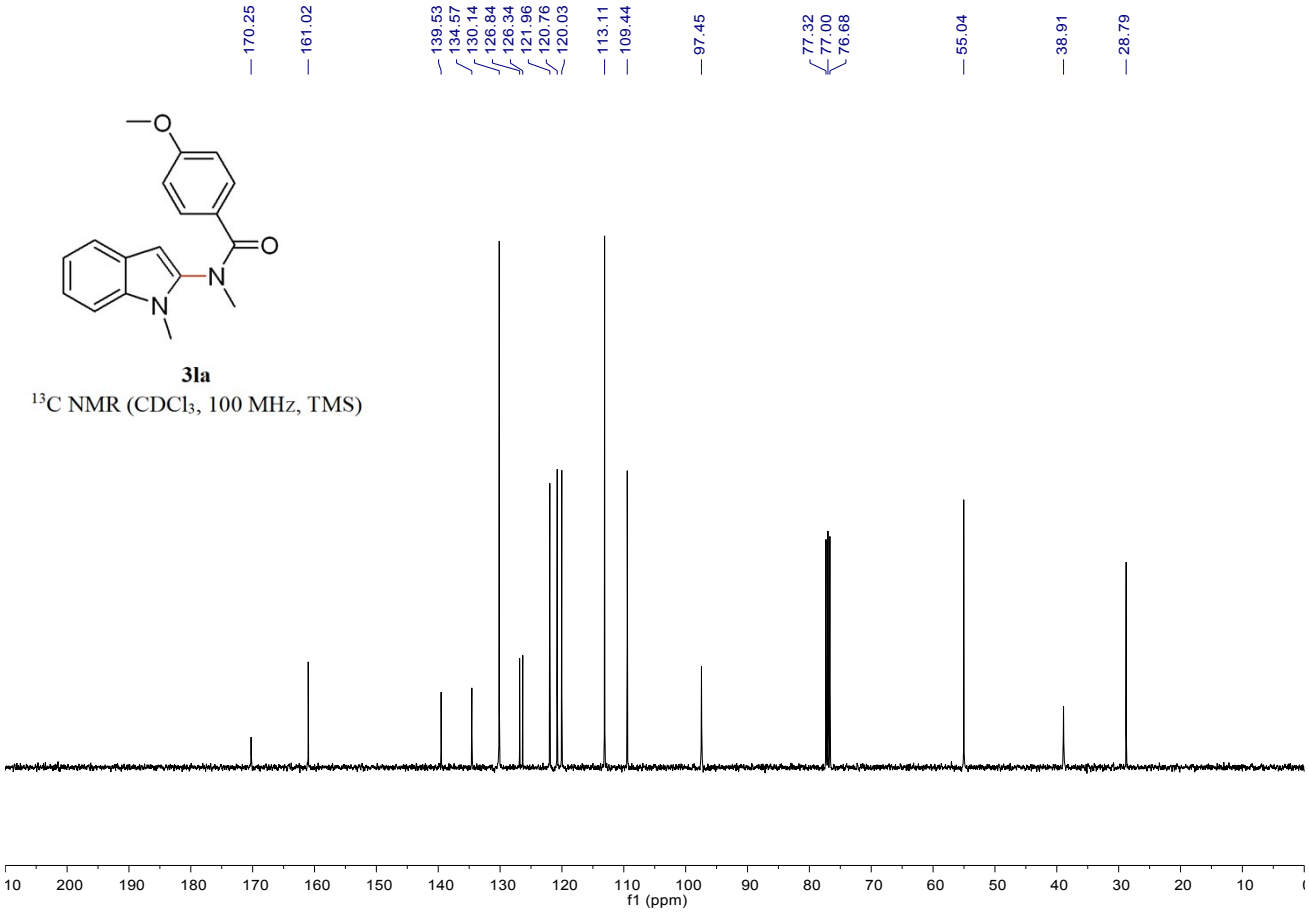


4-methoxy-*N*-methyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3la). The title product was obtained after purification by prep-TLC (PE:EA = 4:1). A brown solid, 60 mg, 76% yield; M.p.: 135 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.50 (d, *J* = 7.9 Hz, 1H), 7.33 (d, *J* = 8.3 Hz, 2H), 7.24 - 7.16 (m, 2H), 7.14 - 7.05 (m, 1H), 6.62 (d, *J* = 8.3 Hz, 2H), 6.22 (s, 1H), 3.67 (s, 3H), 3.49 (s, 3H), 3.43 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.3, 161.0, 139.5, 134.6, 130.1, 126.8, 126.3, 122.0, 120.8, 120.0, 113.1, 109.4, 97.4, 55.0, 38.9, 28.8; IR (neat) ν 2930, 1645, 1603, 1559, 1510, 1461, 1391, 1340, 1314, 1252, 1177, 1165, 1089, 1033, 847, 751, 739, 683 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2O_2Na^+$ Requires: 317.1261, Found: 317.1258.



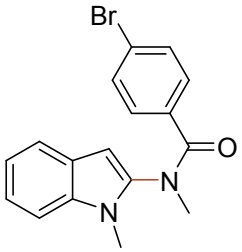
3la

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

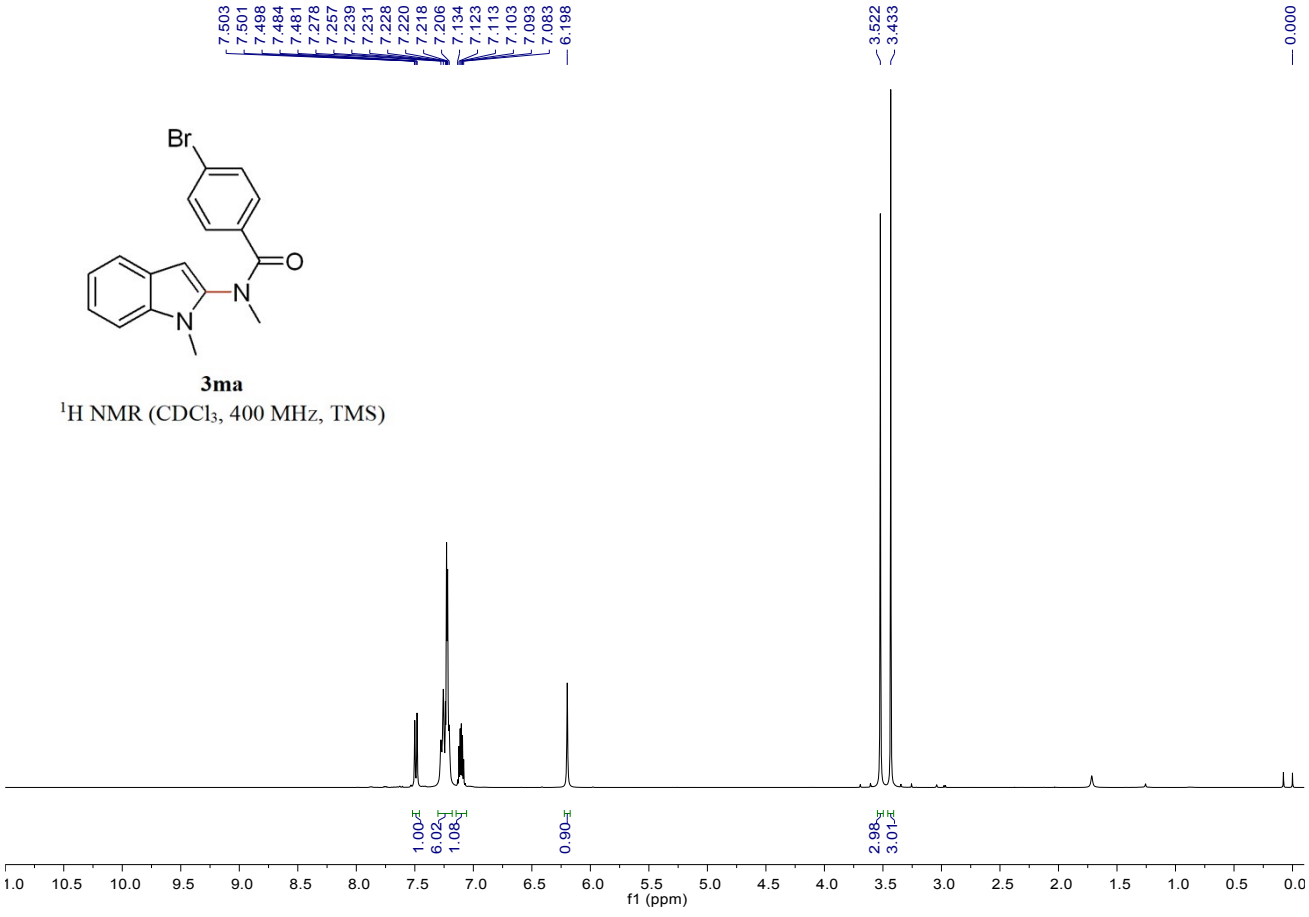


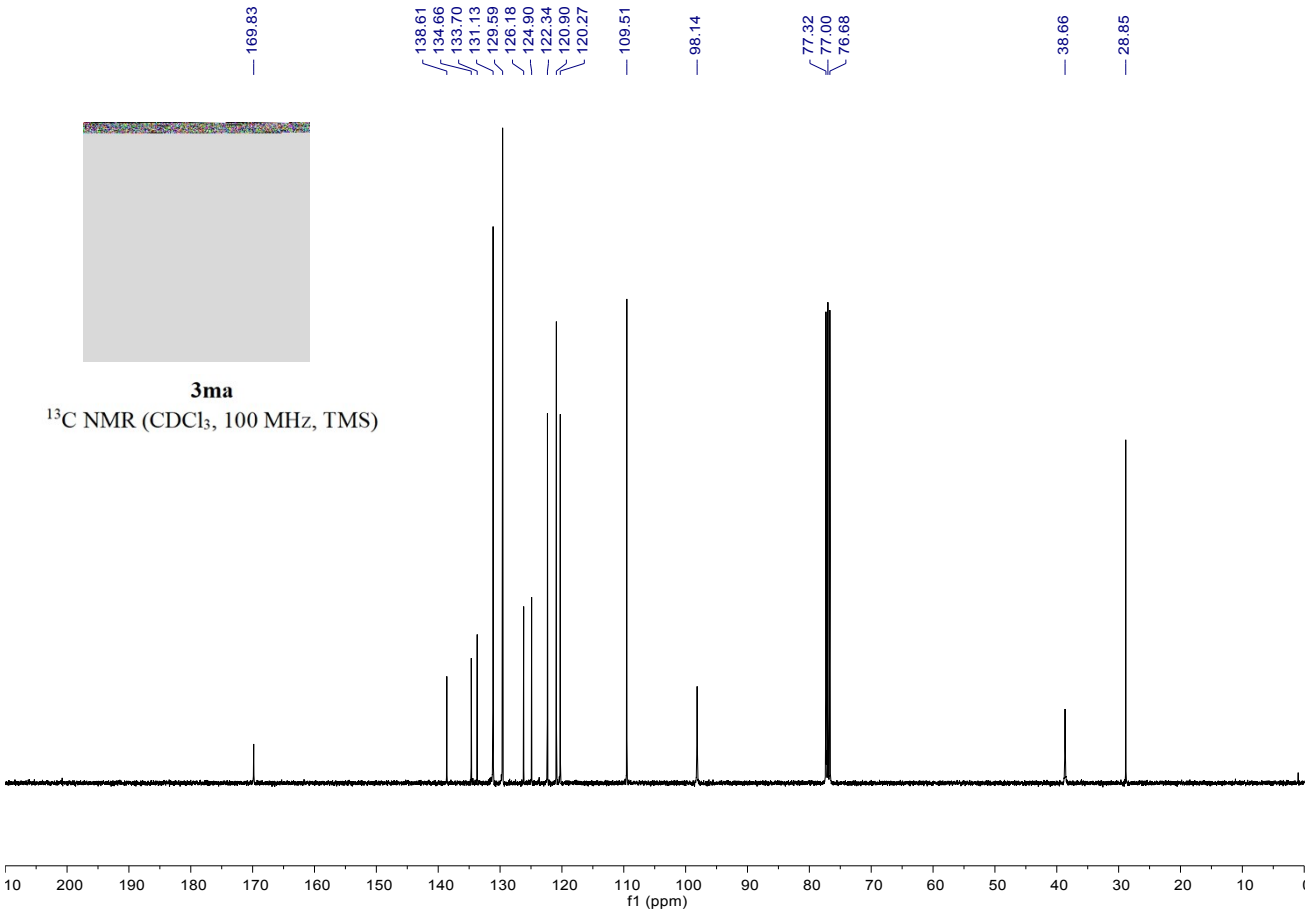
3la

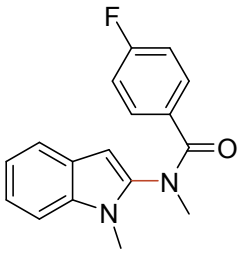
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



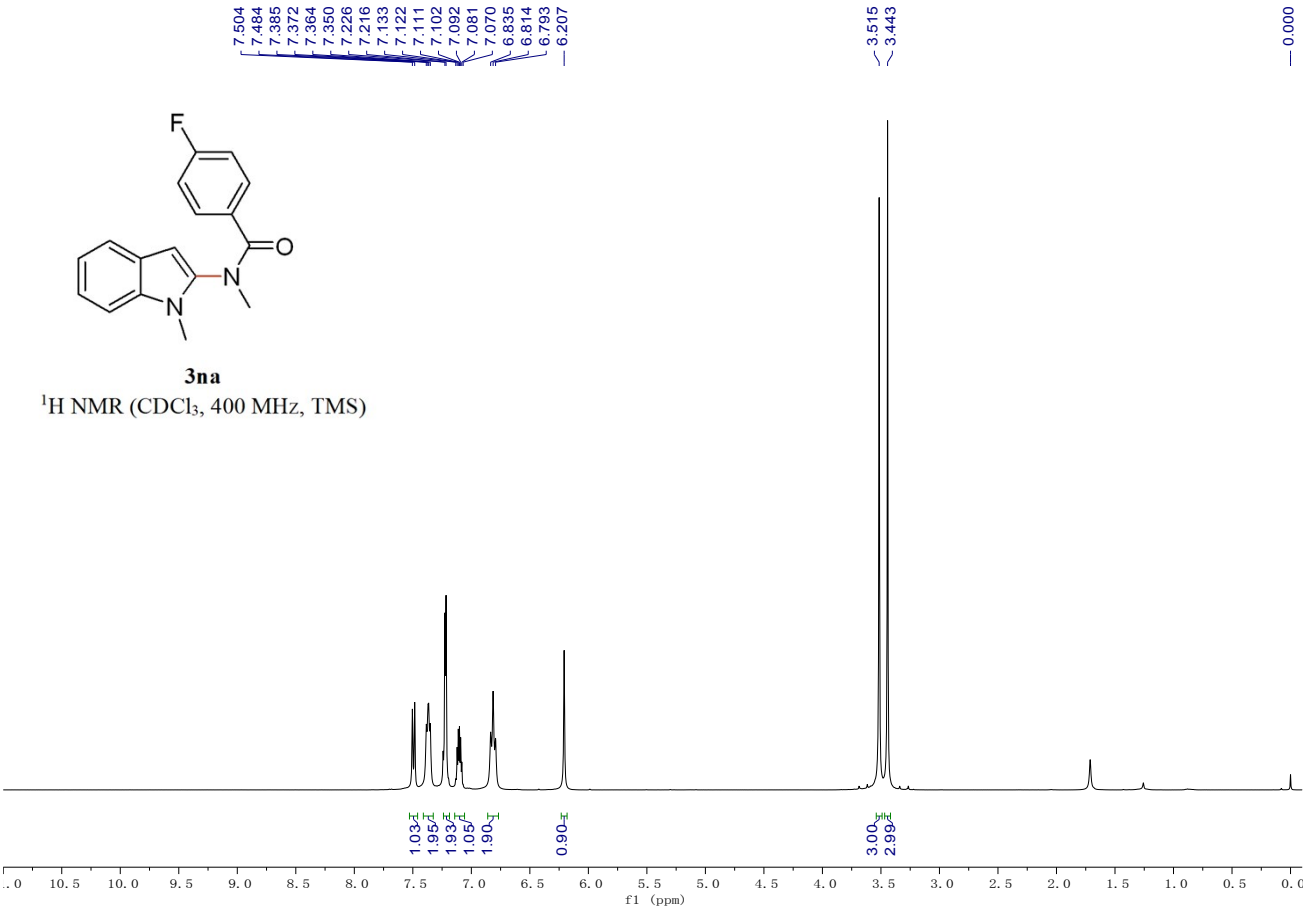
4-bromo-*N*-methyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3ma). The title product was obtained after purification by prep-TLC (PE:EA = 4:1). An off-white solid, 53 mg, 77% yield; M.p.: 184 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.53 - 7.46 (m, 1H), 7.30 - 7.18 (m, 6H), 7.16 - 7.05 (m, 1H), 6.20 (s, 1H), 3.52 (s, 3H), 3.43 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 169.8, 138.6, 134.7, 133.7, 131.1, 129.6, 126.2, 124.9, 122.3, 120.9, 120.3, 109.5, 98.1, 38.7, 28.9; IR (neat) ν 1652, 1589, 1556, 1466, 1395, 1345, 1313, 1253, 1167, 1091, 1067, 1014, 1003, 839, 782, 749, 677 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2ONaBr^+$ Requires: 365.0260, Found: 365.0253.





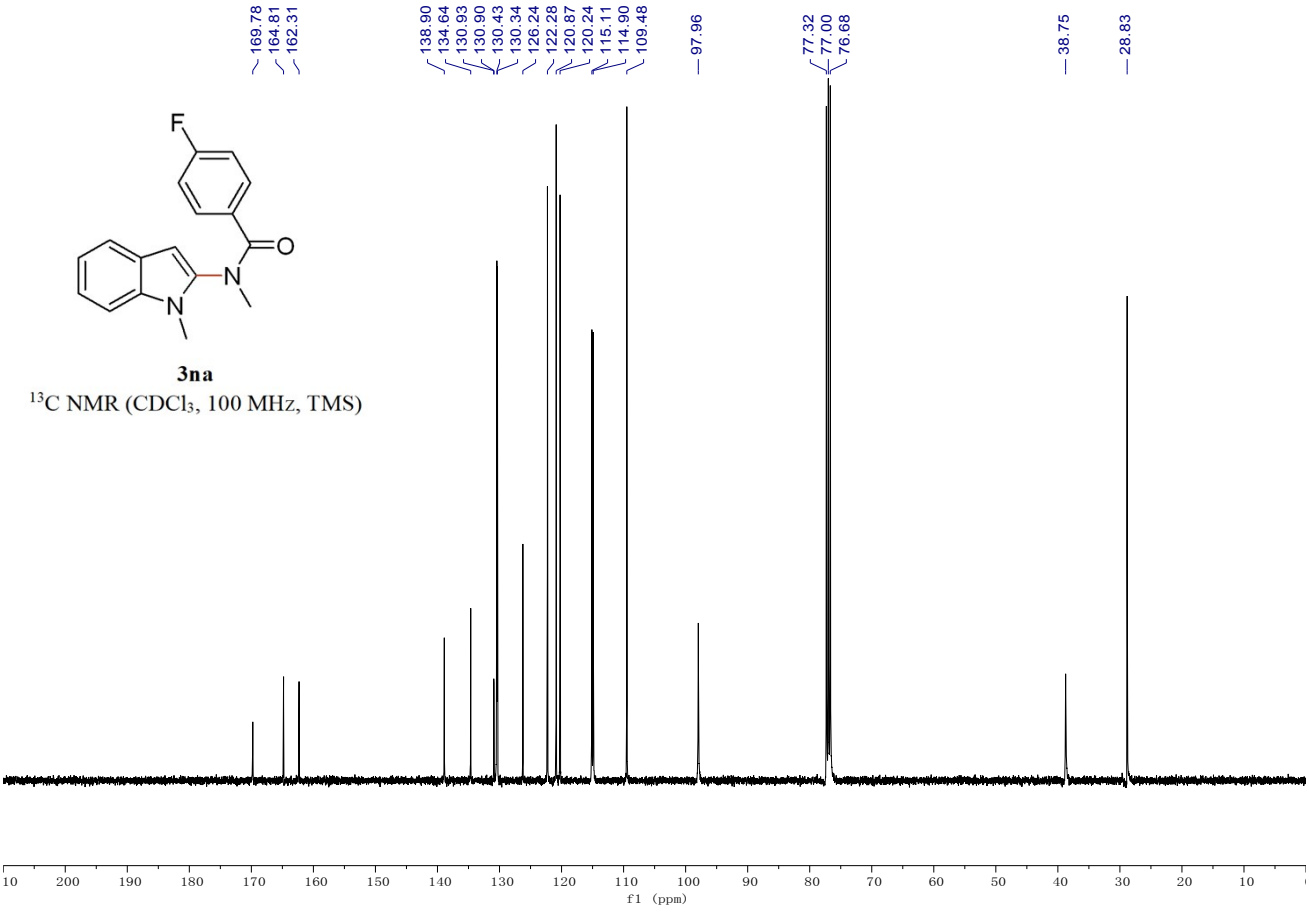


4-fluoro-*N*-methyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3na). The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 31 mg, 55% yield; M.p.: 90 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.49 (d, *J* = 7.8 Hz, 1H), 7.37 (dd, *J* = 8.5, 5.4 Hz, 2H), 7.24 - 7.19 (m, 2H), 7.14 - 7.06 (m, 1H), 6.81 (t, *J* = 8.4 Hz, 2H), 6.21 (s, 1H), 3.52 (s, 3H), 3.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 169.8, 163.6 (d, *J* = 251.5 Hz), 138.9, 134.6, 130.9 (d, *J* = 3.3 Hz), 130.4 (d, *J* = 8.8 Hz), 126.2, 122.3, 120.9, 120.2, 115.0 (d, *J* = 21.8 Hz), 109.5, 98.0, 38.7, 28.8; ^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$) δ -108.9 (td, *J* = 8.5, 4.2 Hz); IR (neat) ν 1654, 1597, 1558, 1507, 1463, 1391, 1333, 1315, 1255, 1226, 1151, 1089, 1004, 850, 780, 749, 737, 680 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2OFNa^+$ Requires: 305.1061, Found: 305.1066.



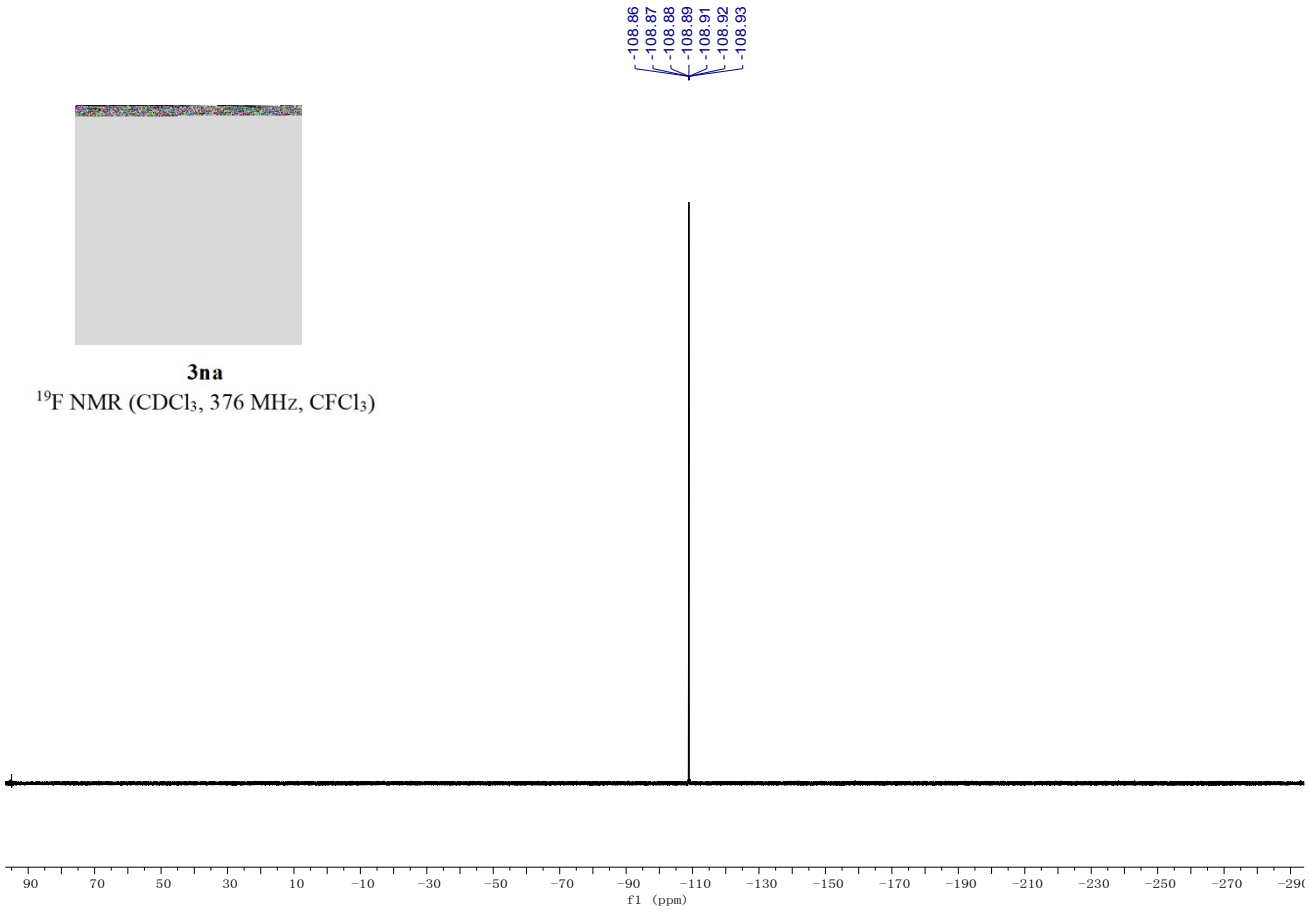
3na

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)



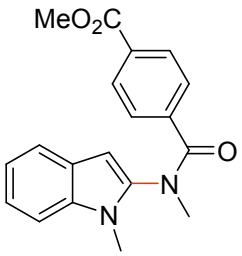
3na

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

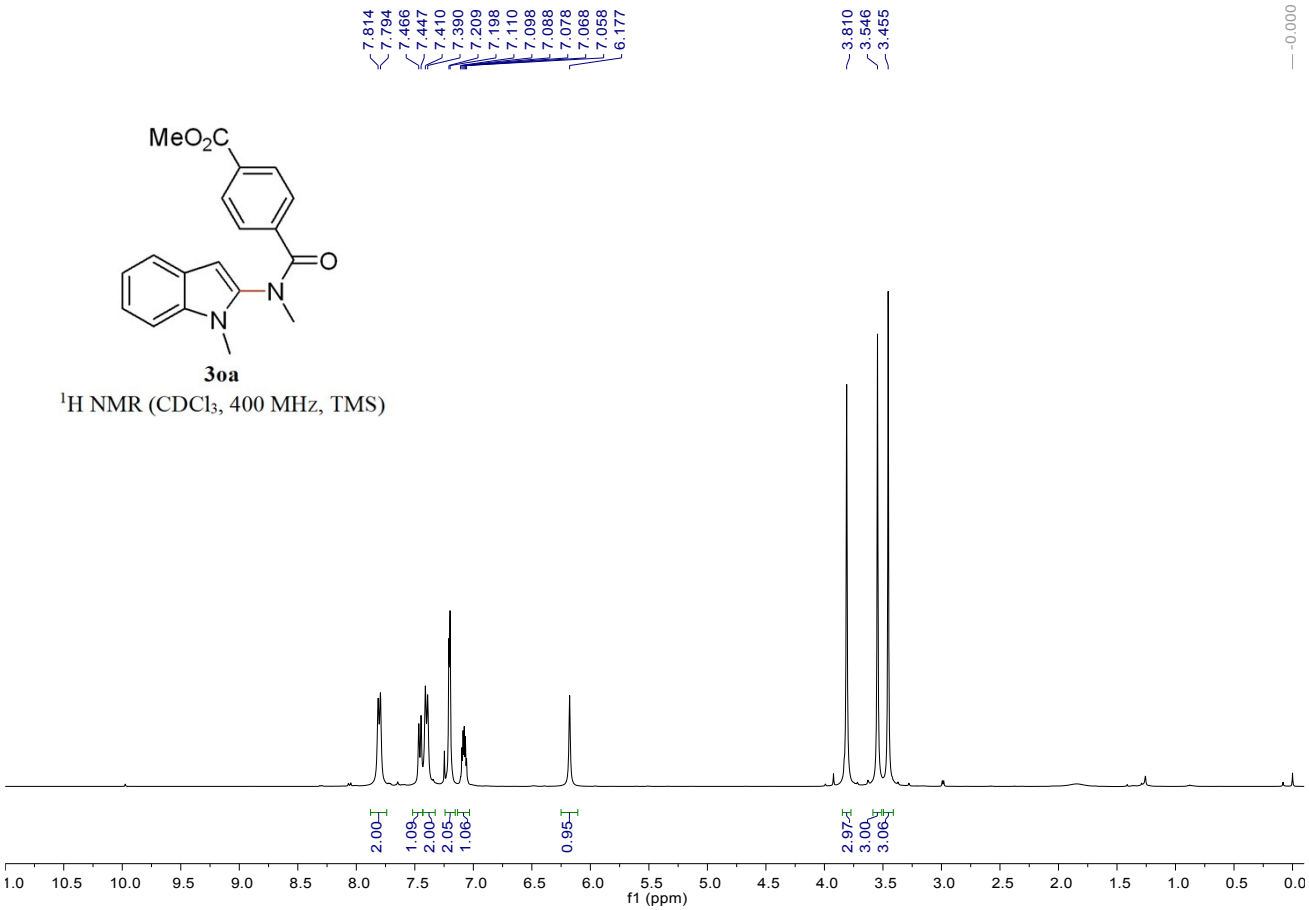


3na

^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$)

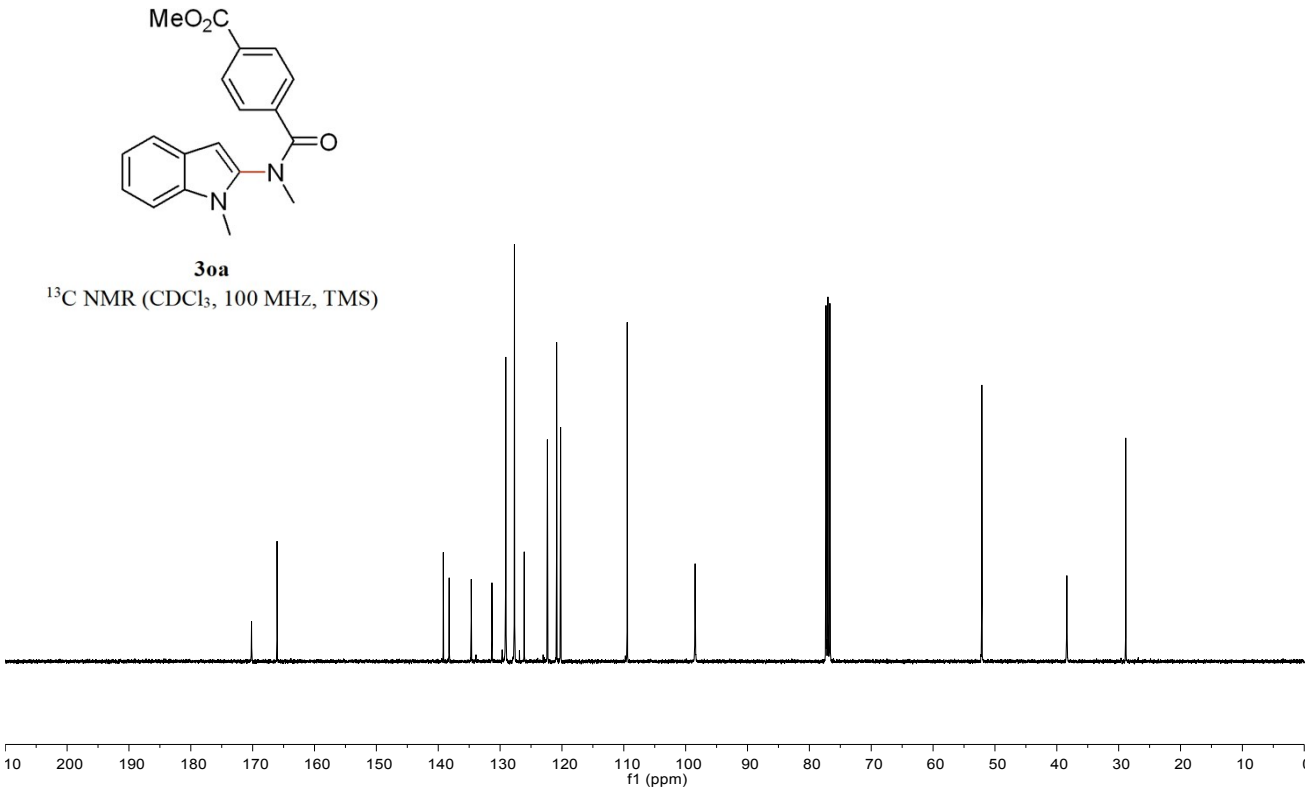


methyl 4-(methyl(1-methyl-1H-indol-2-yl)carbamoyl)benzoate (3oa). The title product was obtained after purification by prep-TLC (DCM). A yellow solid, 45 mg, 70% yield; M.p.: 147 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.80 (d, *J* = 8.1 Hz, 2H), 7.46 (d, *J* = 7.9 Hz, 1H), 7.40 (d, *J* = 8.0 Hz, 2H), 7.20 (d, *J* = 4.1 Hz, 2H), 7.13 - 7.03 (m, 1H), 6.18 (s, 1H), 3.81 (s, 3H), 3.55 (s, 3H), 3.46 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.2, 166.1, 139.2, 138.2, 134.7, 131.3, 129.1, 127.7, 126.1, 122.4, 120.9, 120.2, 109.4, 98.5, 52.1, 38.4, 28.9; IR (neat) ν 1716, 1656, 1557, 1397, 1343, 1280, 1253, 1108, 1091, 1011, 867, 782, 740, 730, 678 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{18}N_2O_3Na^+$ Requires: 345.1210, Found: 345.1208.



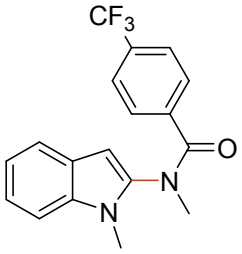
3oa

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

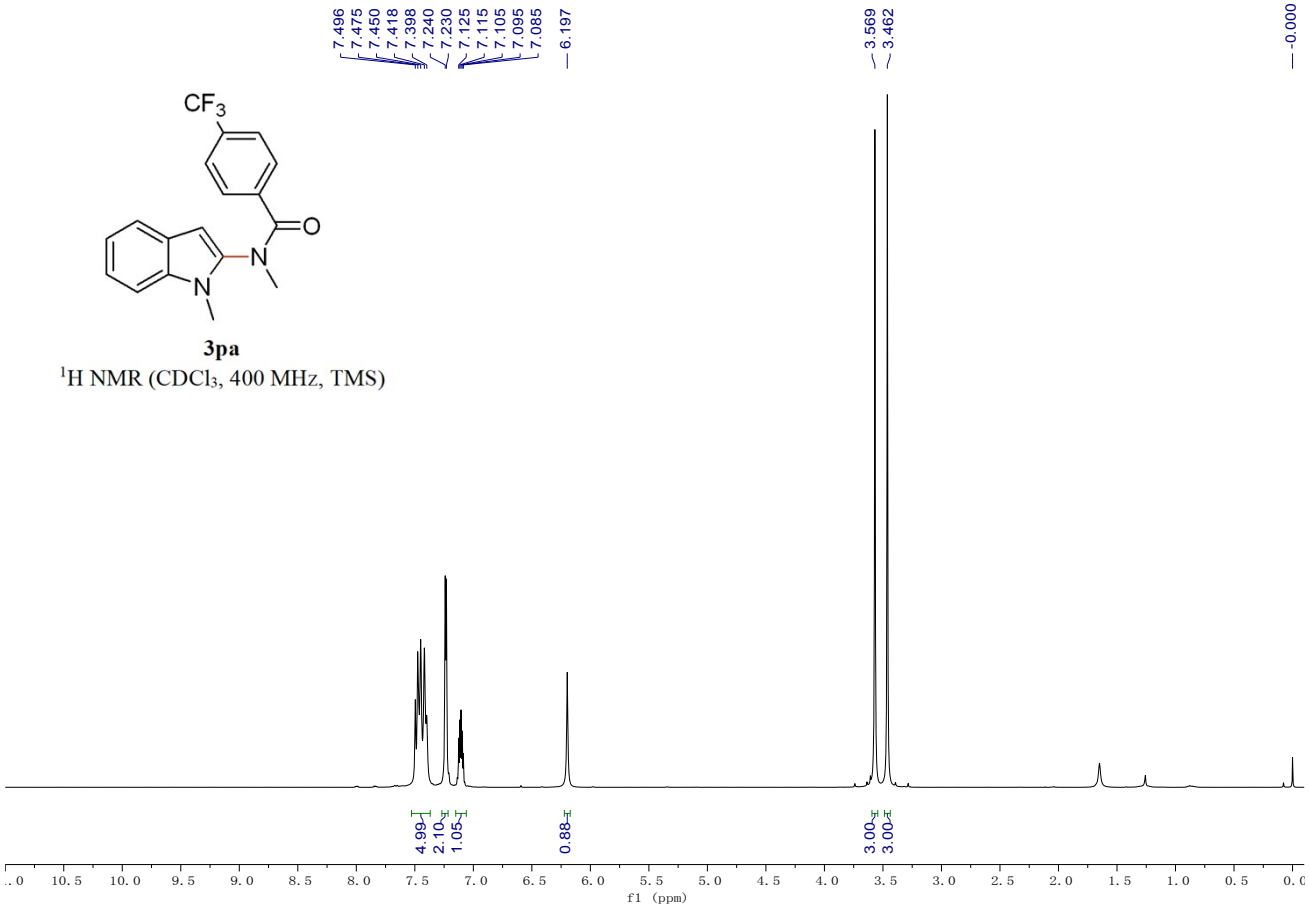


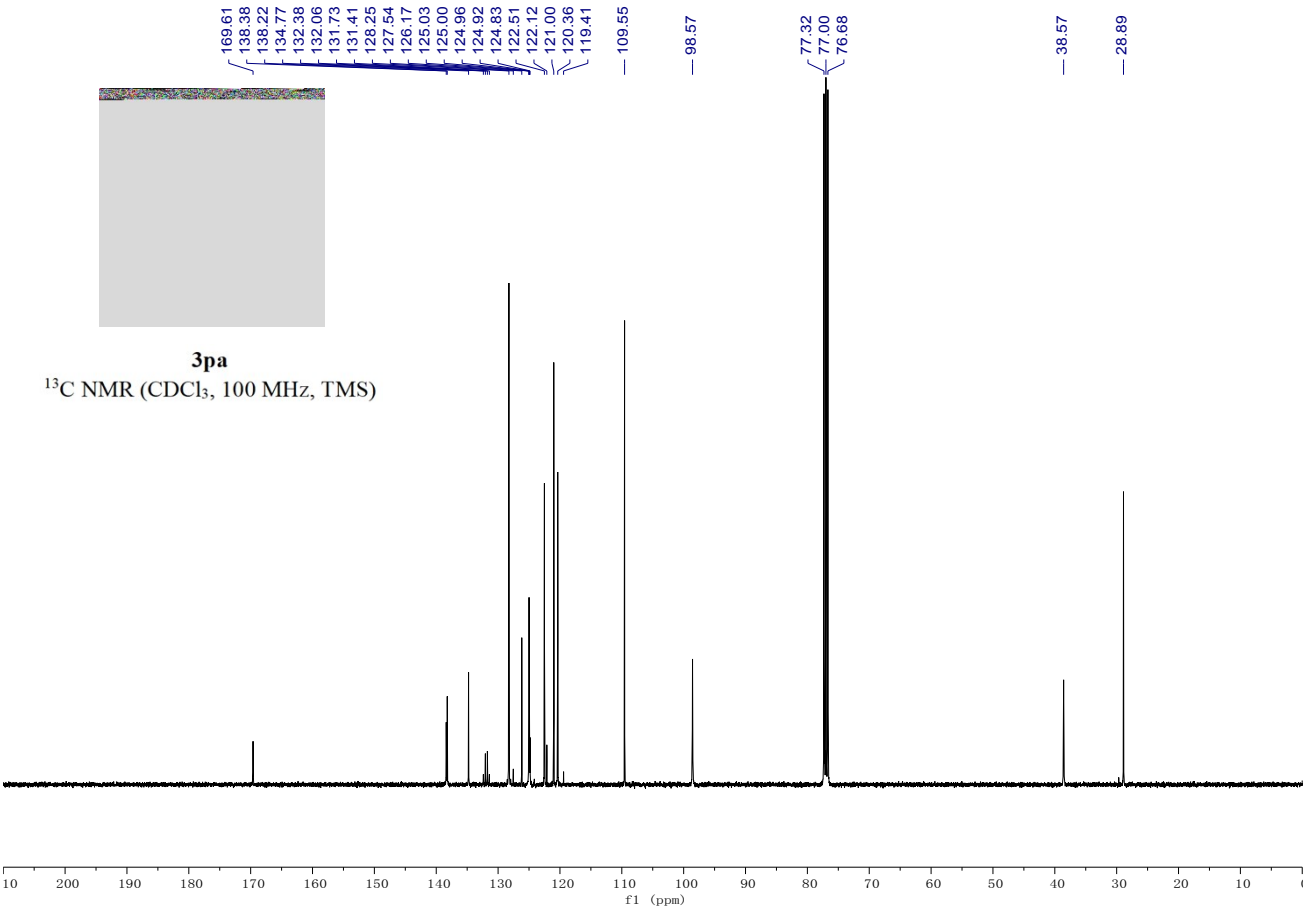
3oa

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



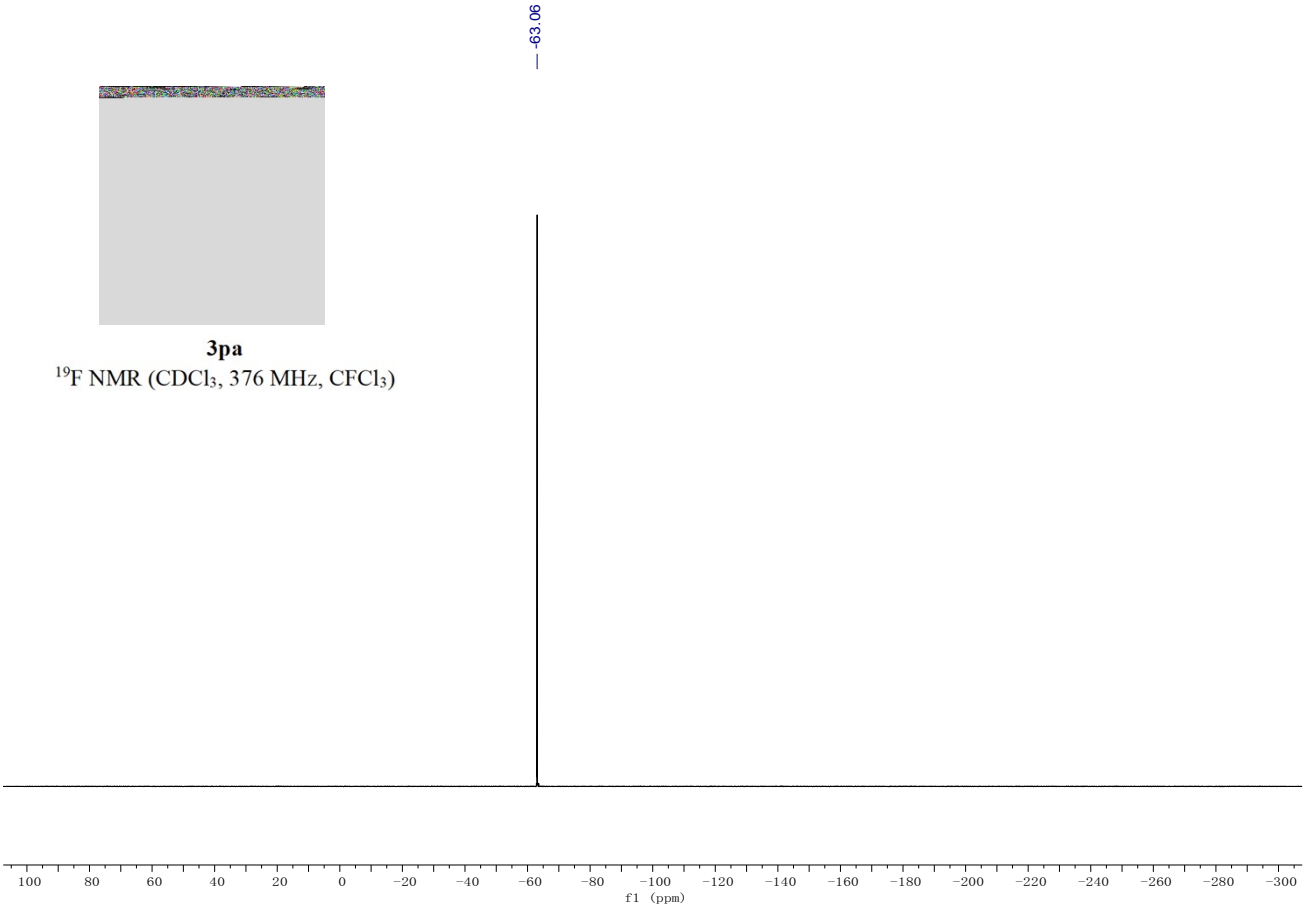
***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)-4-(trifluoromethyl)benzamide (3pa)**. The title product was obtained after purification by prep-TLC (DCM). A white solid, 33 mg, 50% yield; M.p.: 105-107 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.53 - 7.37 (m, 5H), 7.24 (d, *J* = 4.0 Hz, 2H), 7.15 - 7.06 (m, 1H), 6.20 (s, 1H), 3.57 (s, 3H), 3.46 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 169.6, 138.4, 138.2, 134.8, 131.9 (q, *J* = 32.7 Hz), 128.3, 126.2, 125.0 (q, *J* = 3.9 Hz), 123.5 (q, *J* = 272.5 Hz), 122.5, 121.0, 120.4, 109.6, 98.6, 38.6, 28.9; ^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$) δ -63.1; IR (neat) ν 1659, 1557, 1464, 1407, 1322, 1166, 1124, 1066, 1018, 852, 751, 677 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{15}N_2OF_3Na^+$ Requires: 355.1029, Found: 355.1020.





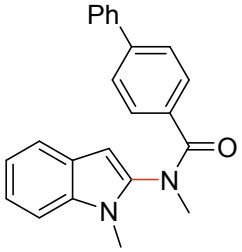
3pa

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

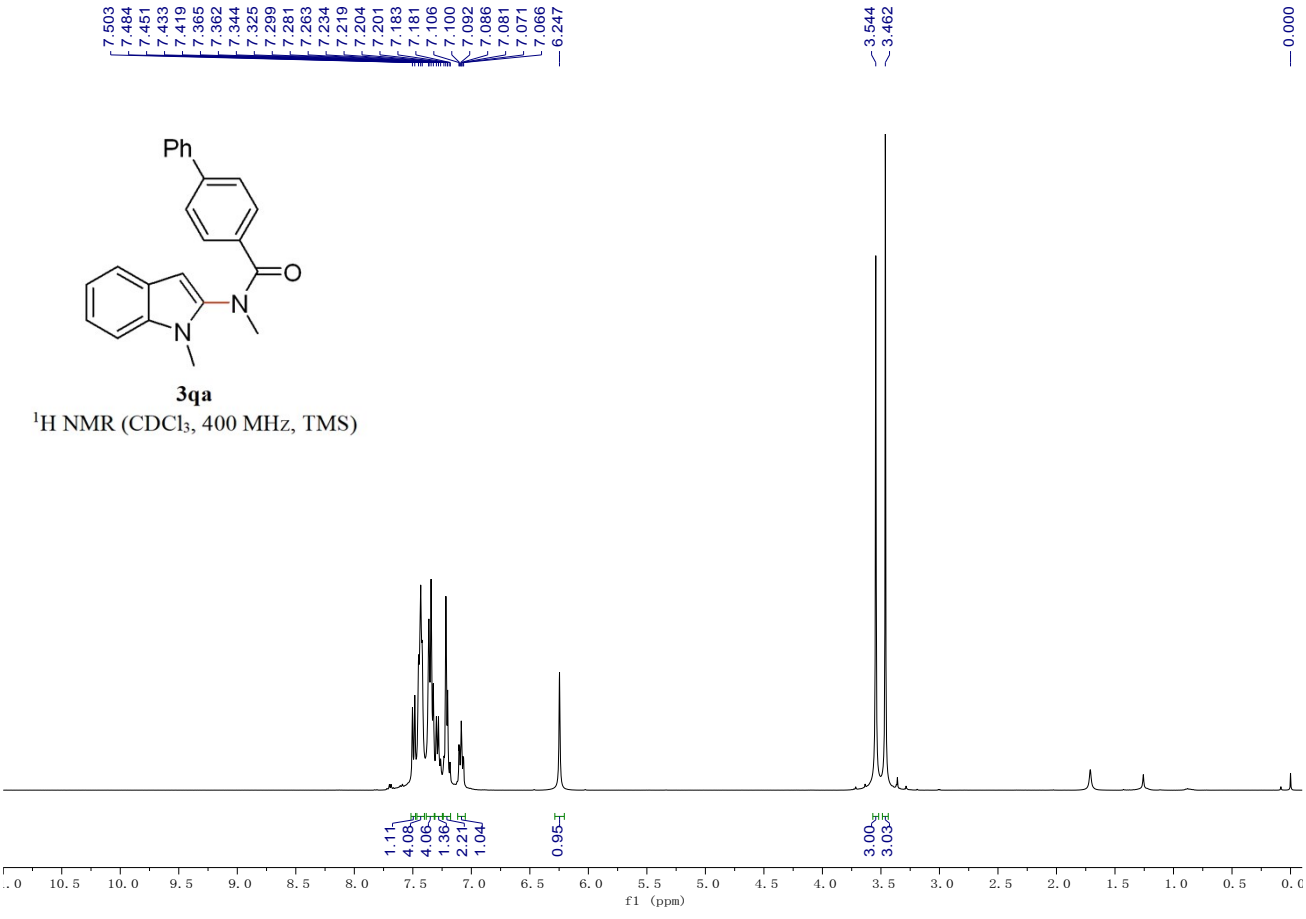


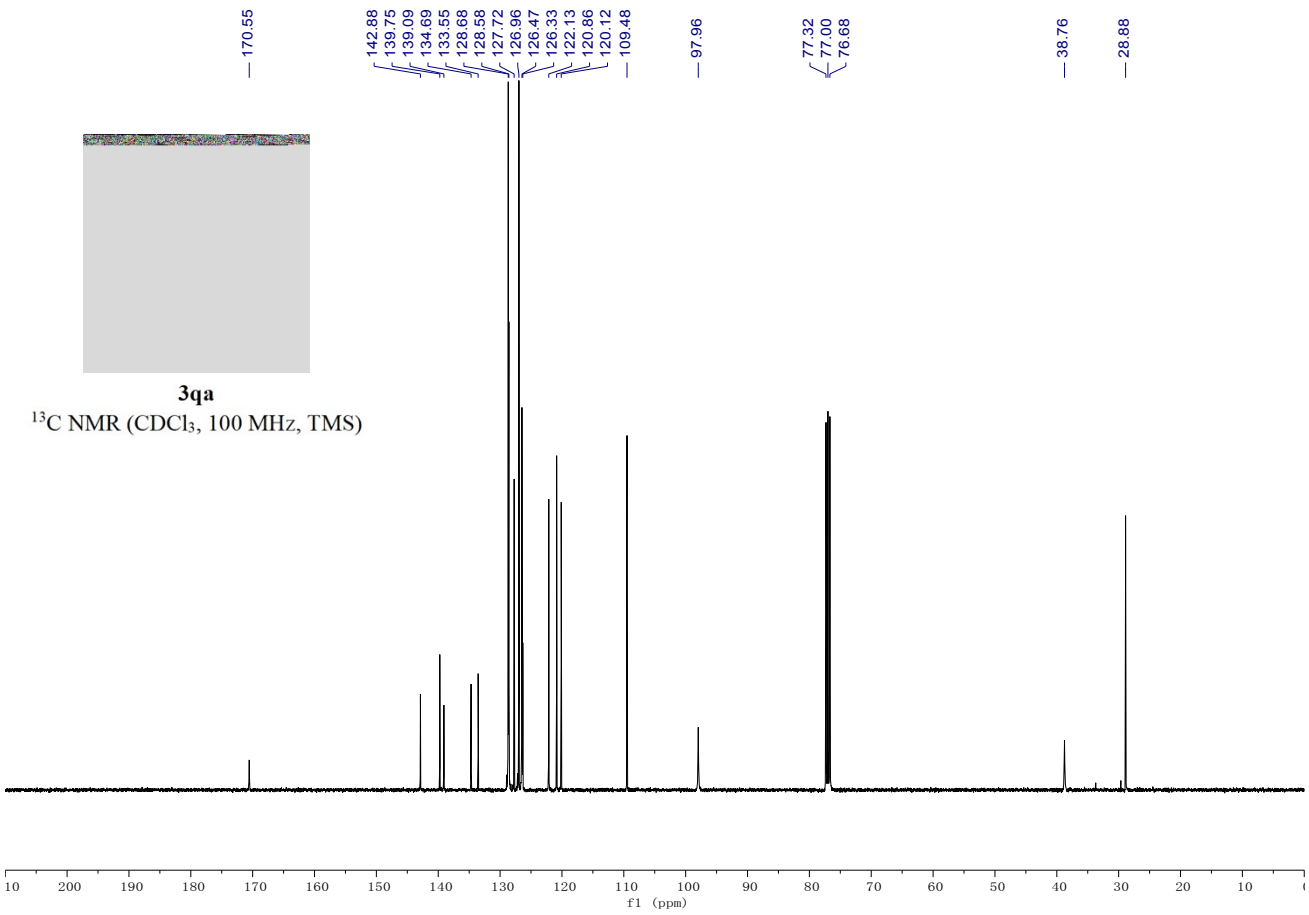
3pa

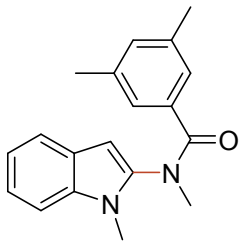
^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$)



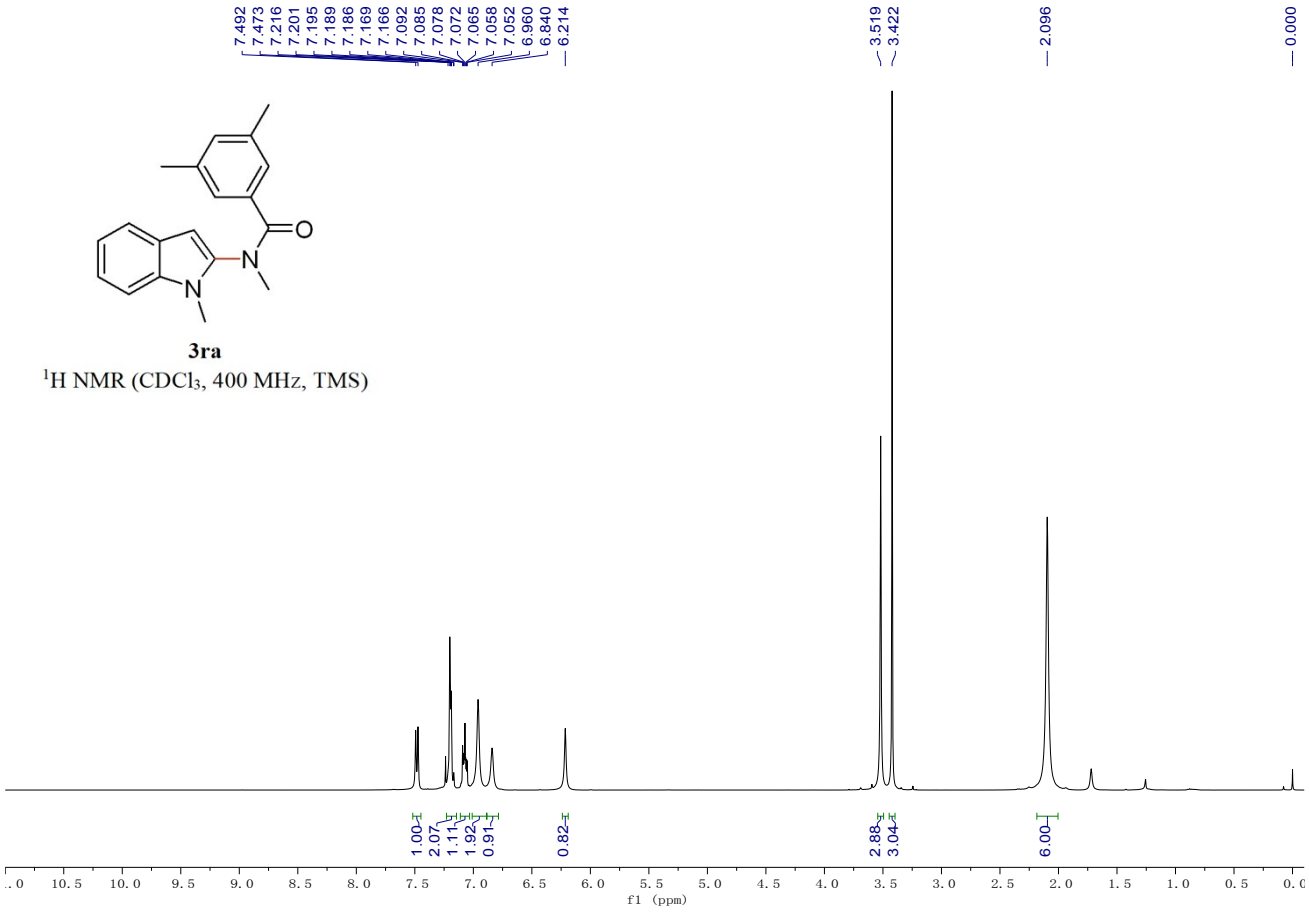
***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)-[1,1'-biphenyl]-4-carboxamide (3qa)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 42 mg, 62% yield; M.p.: 143 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.49 (d, *J* = 7.9 Hz, 1H), 7.47 - 7.40 (m, 4H), 7.39 - 7.32 (m, 4H), 7.31 - 7.25 (m, 1H), 7.24 - 7.18 (m, 2H), 7.12 - 7.05 (m, 1H), 6.25 (s, 1H), 3.54 (s, 3H), 3.46 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.6, 142.9, 139.7, 139.1, 134.7, 133.5, 128.7, 128.6, 127.7, 127.0, 126.5, 126.3, 122.1, 120.9, 120.1, 109.5, 98.0, 38.8, 28.9; IR (neat) ν 1644, 1578, 1560, 1460, 1426, 1393, 1341, 1316, 1256, 1092, 1007, 849, 774, 746, 731, 701, 682 cm^{-1}; HRMS (ESI) Calcd. for $C_{23}H_{20}N_2ONa^+$ Requires: 363.1468, Found: 363.1470.





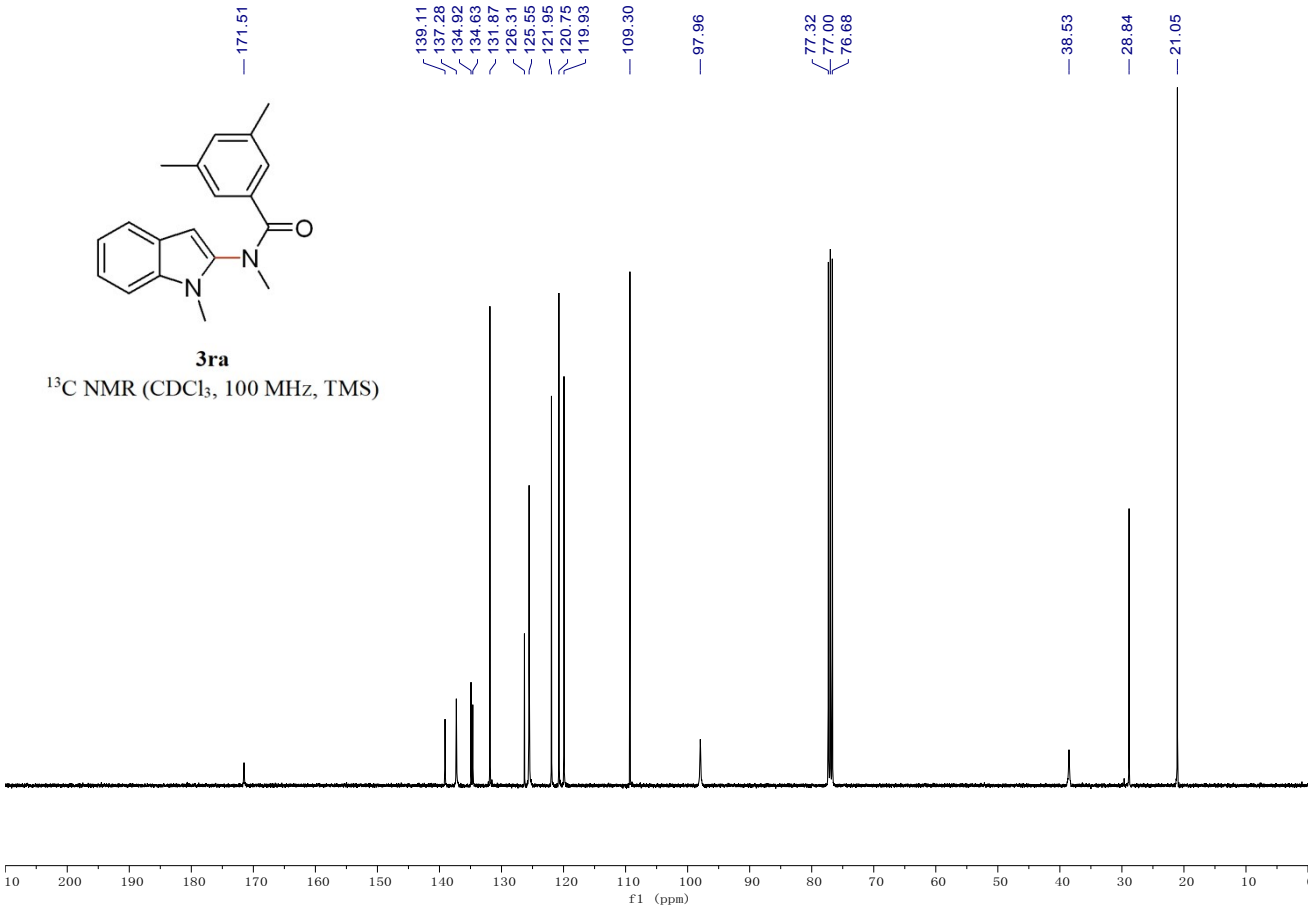


***N*,3,5-trimethyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3ra)**. The title product was obtained after purification by prep-TLC (DCM to PE:EA = 4:1). An off-white solid, 33 mg, 57% yield; M.p.: 136 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.48 (d, *J* = 7.9 Hz, 1H), 7.23 - 7.14 (m, 2H), 7.11 - 7.03 (m, 1H), 6.96 (s, 2H), 6.84 (s, 1H), 6.21 (s, 1H), 3.52 (s, 3H), 3.42 (s, 3H), 2.10 (s, 6H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.5, 139.1, 137.3, 134.9, 134.6, 131.9, 126.3, 125.6, 122.0, 120.8, 119.9, 109.3, 98.0, 38.5, 28.8, 21.1; IR (neat) ν 2918, 1649, 1604, 1557, 1459, 1394, 1341, 1315, 1245, 1151, 1093, 858, 810, 751, 732, 690 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{20}N_2ONa^+$ Requires: 315.1468, Found: 315.1467.



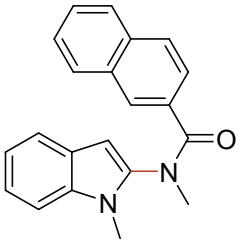
3ra

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

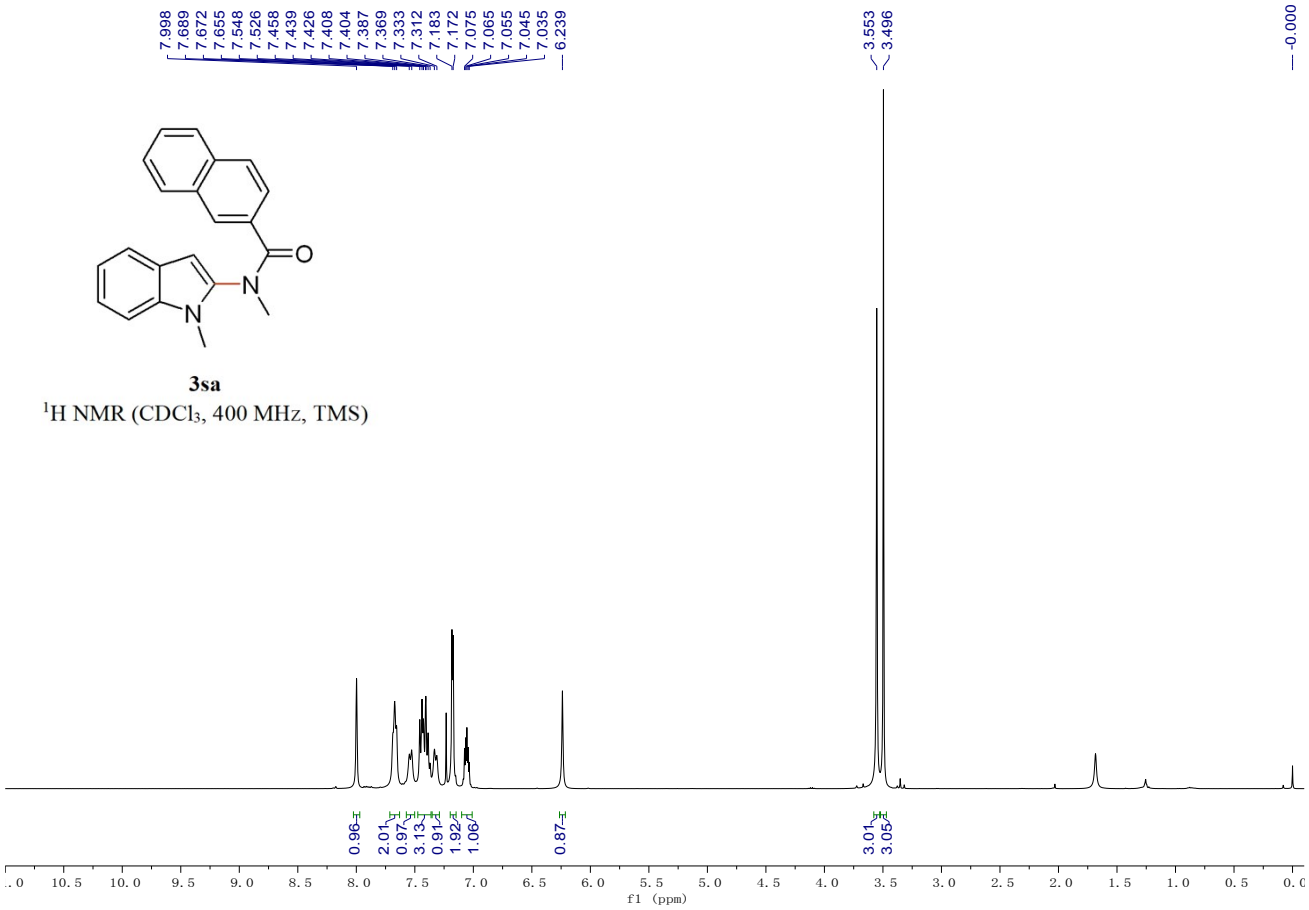


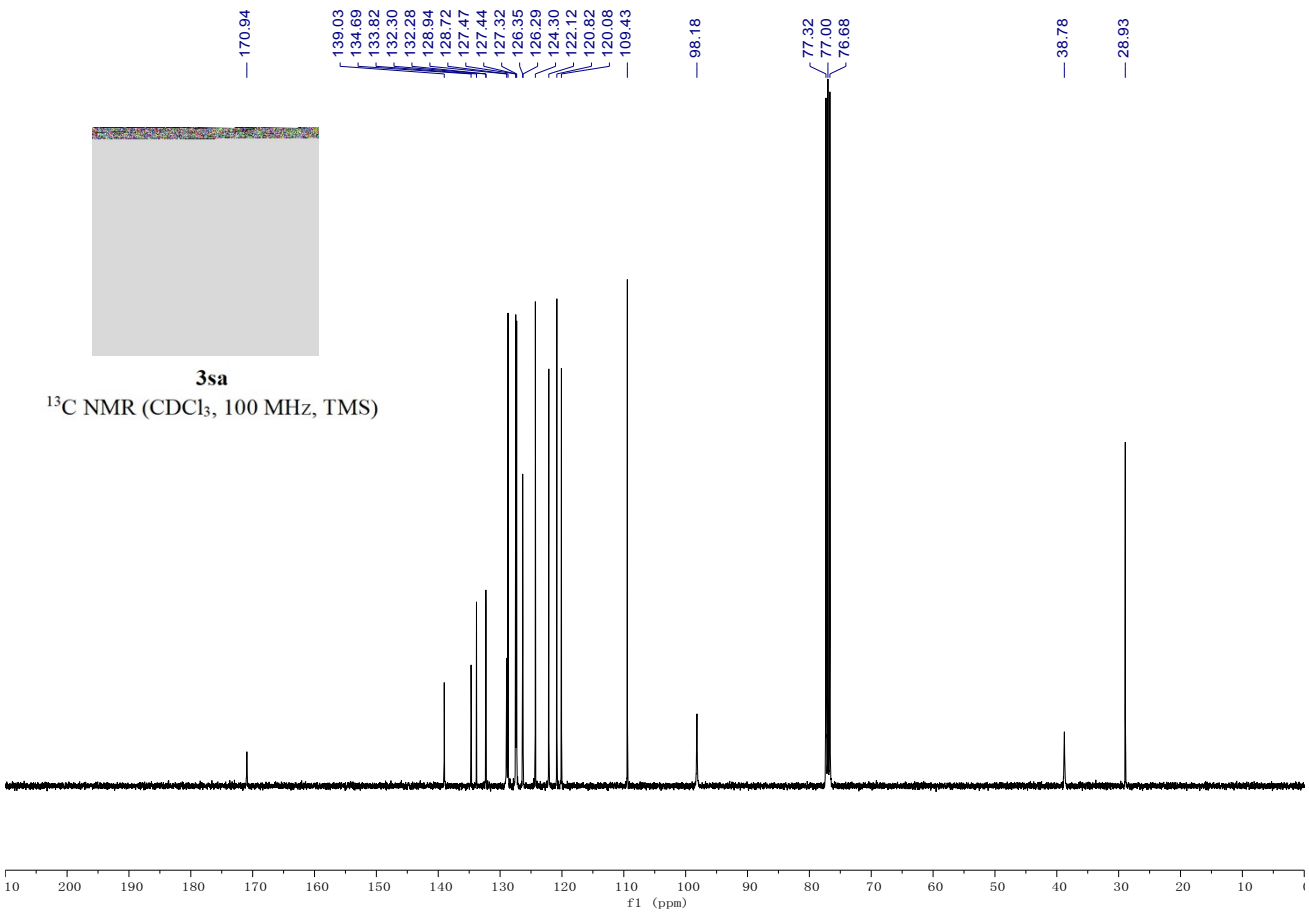
3ra

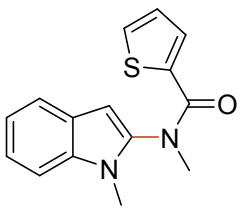
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



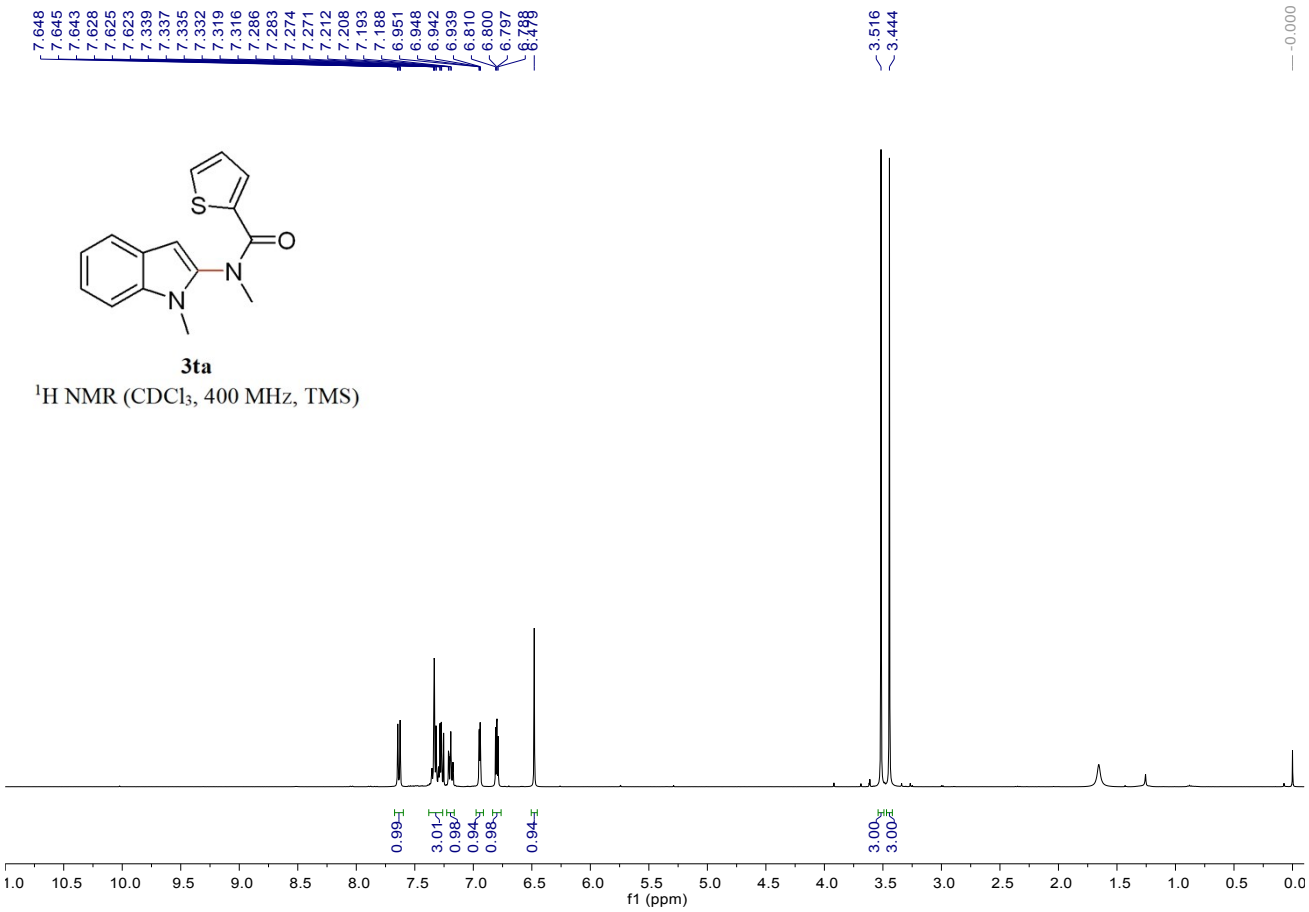
***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)-2-naphthamide (3sa)**. The title product was obtained after purification by prep-TLC (DCM). An yellow solid, 28 mg, 44% yield; M.p.: 140 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 8.00 (s, 1H), 7.67 (t, *J* = 6.6 Hz, 2H), 7.54 (d, *J* = 8.6 Hz, 1H), 7.47 - 7.36 (m, 3H), 7.32 (d, *J* = 8.6 Hz, 1H), 7.18 (d, *J* = 4.1 Hz, 2H), 7.10 - 7.01 (m, 1H), 6.24 (s, 1H), 3.55 (s, 3H), 3.50 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 139.0, 134.7, 133.8, 132.30, 132.28, 128.9, 128.7, 127.5, 127.4, 127.3, 126.3, 126.3, 124.3, 122.1, 120.8, 120.1, 109.4, 98.2, 38.8, 28.9; IR (neat) ν 3056, 1642, 1554, 1466, 1411, 1339, 1310, 1241, 1124, 1088, 865, 817, 776, 751, 739, 682 cm^{-1}; HRMS (ESI) Calcd. for $C_{21}H_{18}N_2ONa^+$ Requires: 337.1311, Found: 337.1307.





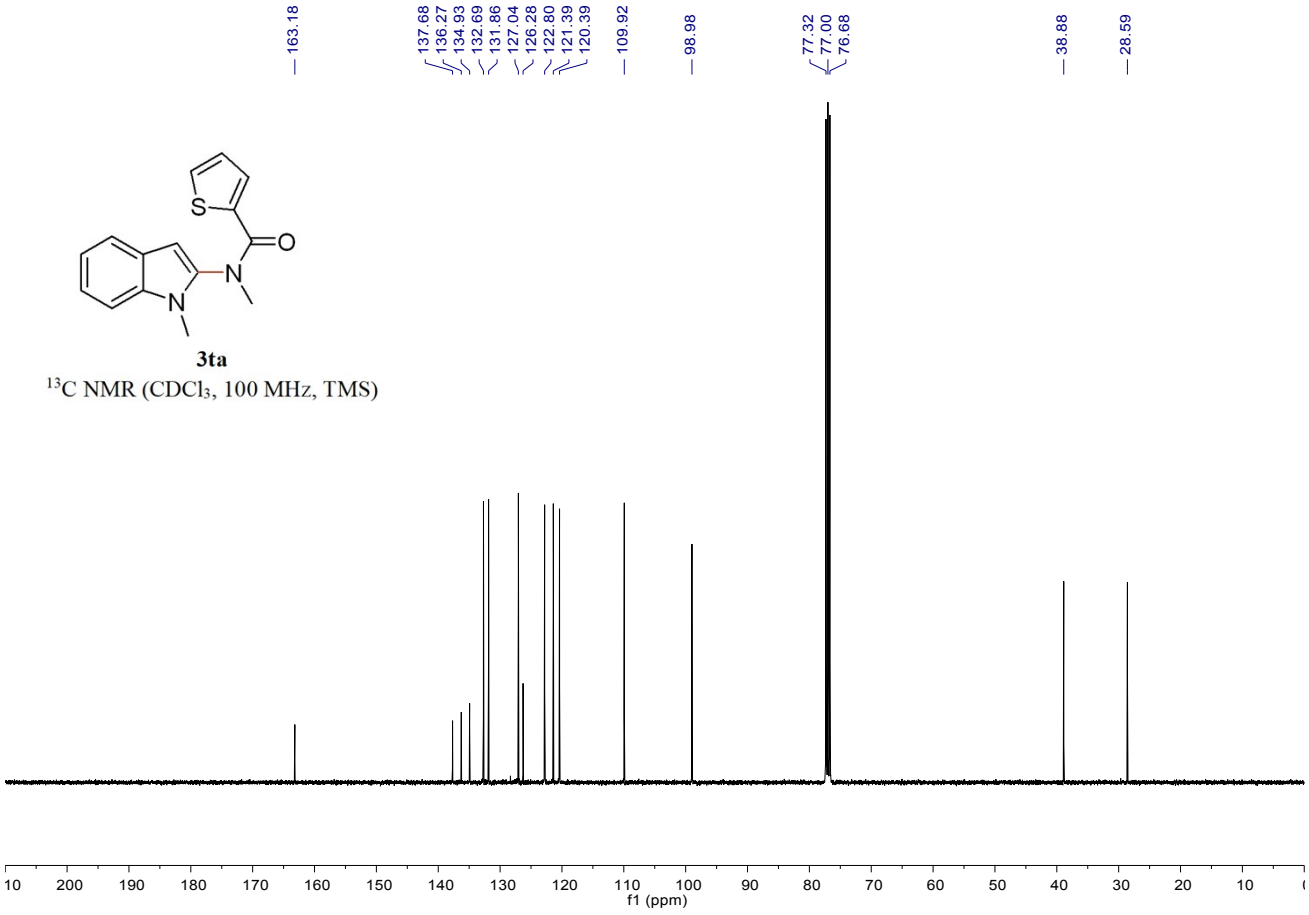


***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)thiophene-2-carboxamide (3ta)**. The title product was obtained after purification by prep-TLC (PE:EA = 4:1). A yellow solid, 21 mg, 38% yield; M.p.: 142 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.64 (dt, *J* = 8.0, 1.0 Hz, 1H), 7.38 - 7.26 (m, 3H), 7.23 - 7.16 (m, 1H), 6.94 (dd, *J* = 3.9, 1.2 Hz, 1H), 6.80 (dd, *J* = 5.0, 3.8 Hz, 1H), 6.48 (s, 1H), 3.52 (s, 3H), 3.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 163.2, 137.7, 136.3, 134.9, 132.7, 131.9, 127.0, 126.3, 122.8, 121.4, 120.4, 109.9, 99.0, 38.9, 28.6; IR (neat) ν 2924, 1624, 1557, 1510, 1466, 1420, 1359, 1339, 1312, 1253, 1224, 1072, 796, 739, 729, 682 cm^{-1}; HRMS (ESI) Calcd. for $C_{15}H_{14}N_2ONaS^+$ Requires: 293.0719, Found: 293.0707.



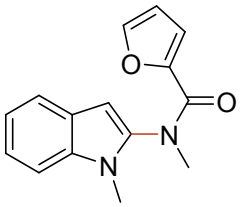
3ta

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

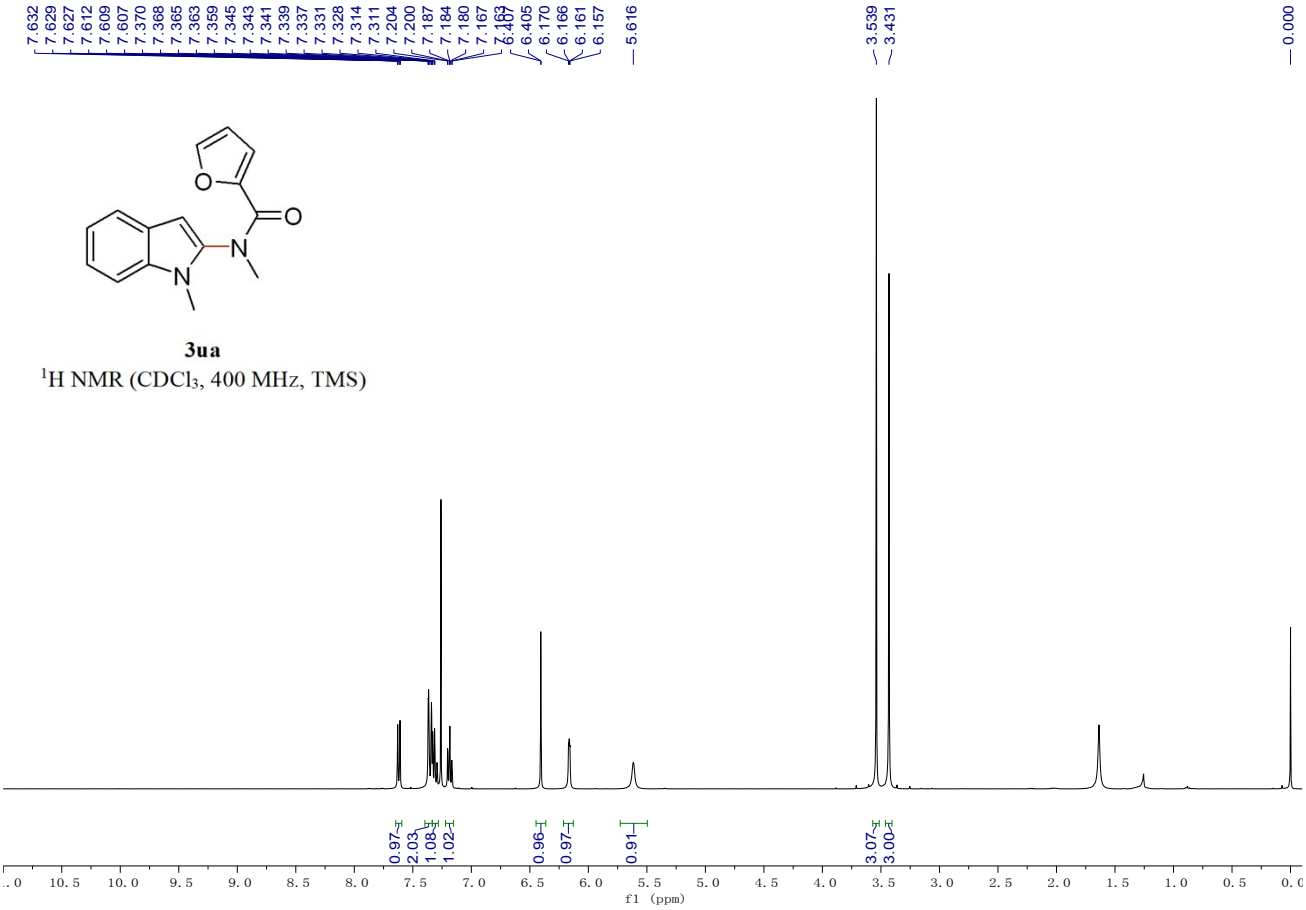


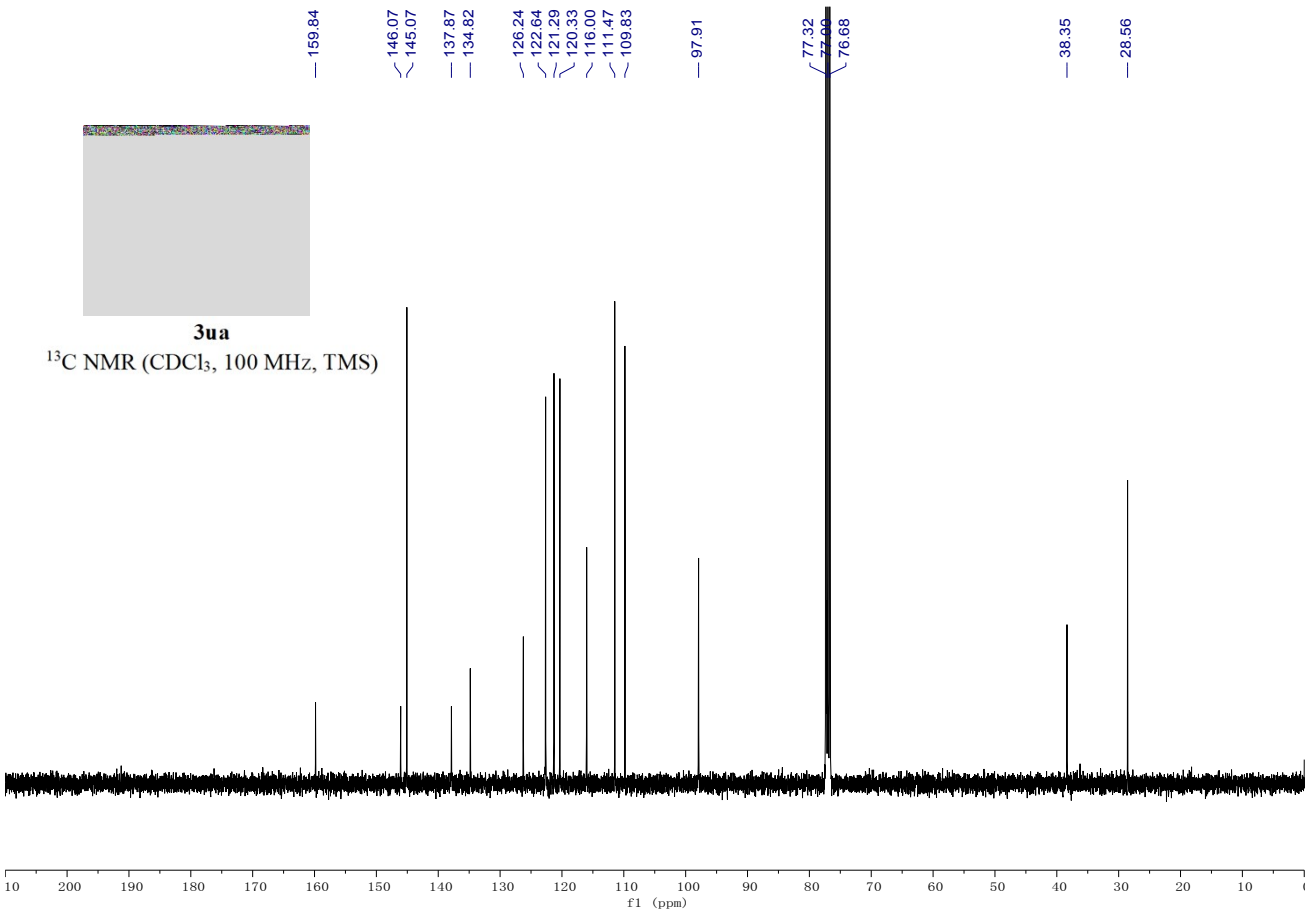
3ta

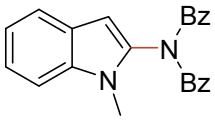
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



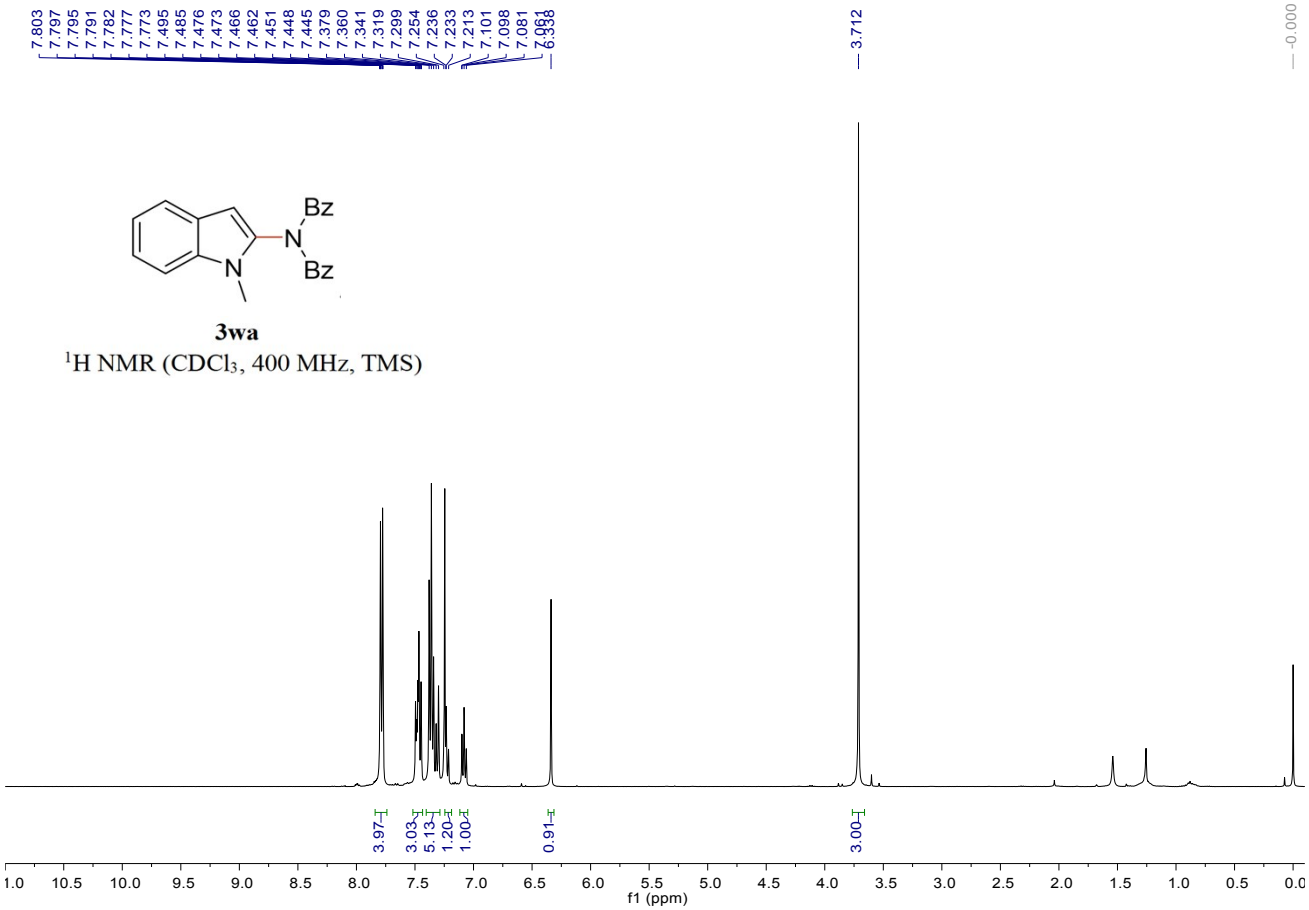
***N*-methyl-*N*-(1-methyl-1H-indol-2-yl)furan-2-carboxamide (3ua)**. The title product was obtained after purification by prep-TLC (PE:EA = 4:1). An yellow solid, 8 mg, 16% yield; M.p.: 144 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.62 (dt, *J* = 7.9, 1.0 Hz, 1H), 7.40 - 7.33 (m, 2H), 7.33 - 7.28 (m, 1H), 7.22 - 7.15 (m, 1H), 6.41 (d, *J* = 0.7 Hz, 1H), 6.16 (dd, *J* = 3.6, 1.7 Hz, 1H), 5.62 (s, 1H), 3.54 (s, 3H), 3.43 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 159.8, 146.1, 145.1, 137.9, 134.8, 126.2, 122.6, 121.3, 120.3, 116.0, 111.5, 109.8, 97.9, 38.4, 28.6; IR (neat) ν 3120, 2922, 1628, 1579, 1556, 1464, 1435, 1387, 1341, 1315, 1237, 1171, 1086, 1039, 1010, 932, 855, 808, 787, 751, 688 cm^{-1}; HRMS (ESI) Calcd. for $C_{15}H_{14}N_2O_2Na^+$ Requires: 277.0948, Found: 277.0954.





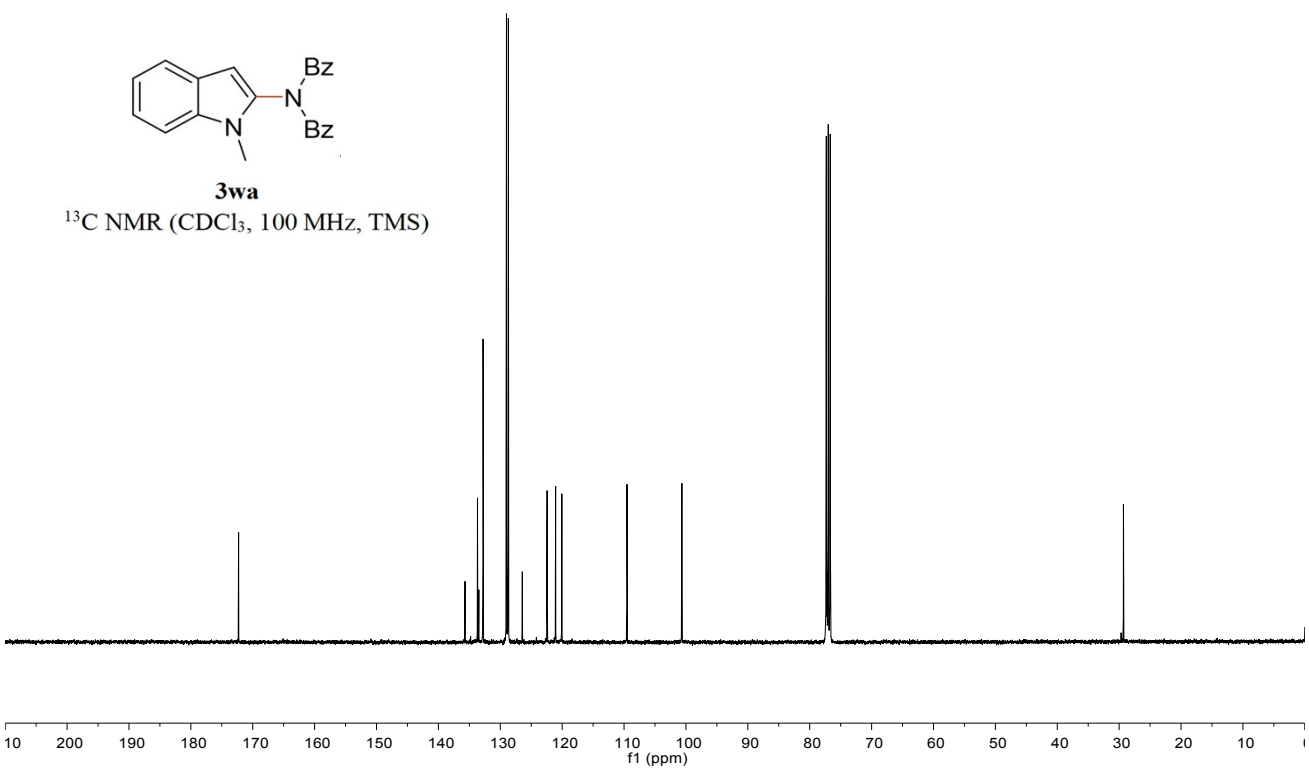


***N*-benzoyl-*N*-(1-methyl-1H-indol-2-yl)benzamide (3wa)**. The title product was obtained after purification by silica gel column chromatography (DCM:PE = 1:1 to PE:EA = 4:1). An yellow solid, 13 mg, 18% yield; M.p.: 143 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.84 - 7.74 (m, 4H), 7.52 - 7.44 (m, 3H), 7.40 - 7.29 (m, 5H), 7.24 - 7.19 (m, 1H), 7.12 - 7.05 (m, 1H), 6.34 (s, 1H), 3.71 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 172.3, 135.7, 133.7, 133.5, 132.8, 129.0, 128.7, 126.4, 122.4, 121.1, 120.1, 109.5, 100.7, 29.3; IR (neat) ν 1698, 1682, 1546, 1446, 1286, 1250, 1235, 1118, 941, 856, 765, 739, 713, 689, 670 cm^{-1}.



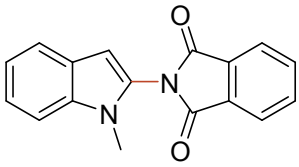
3wa

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

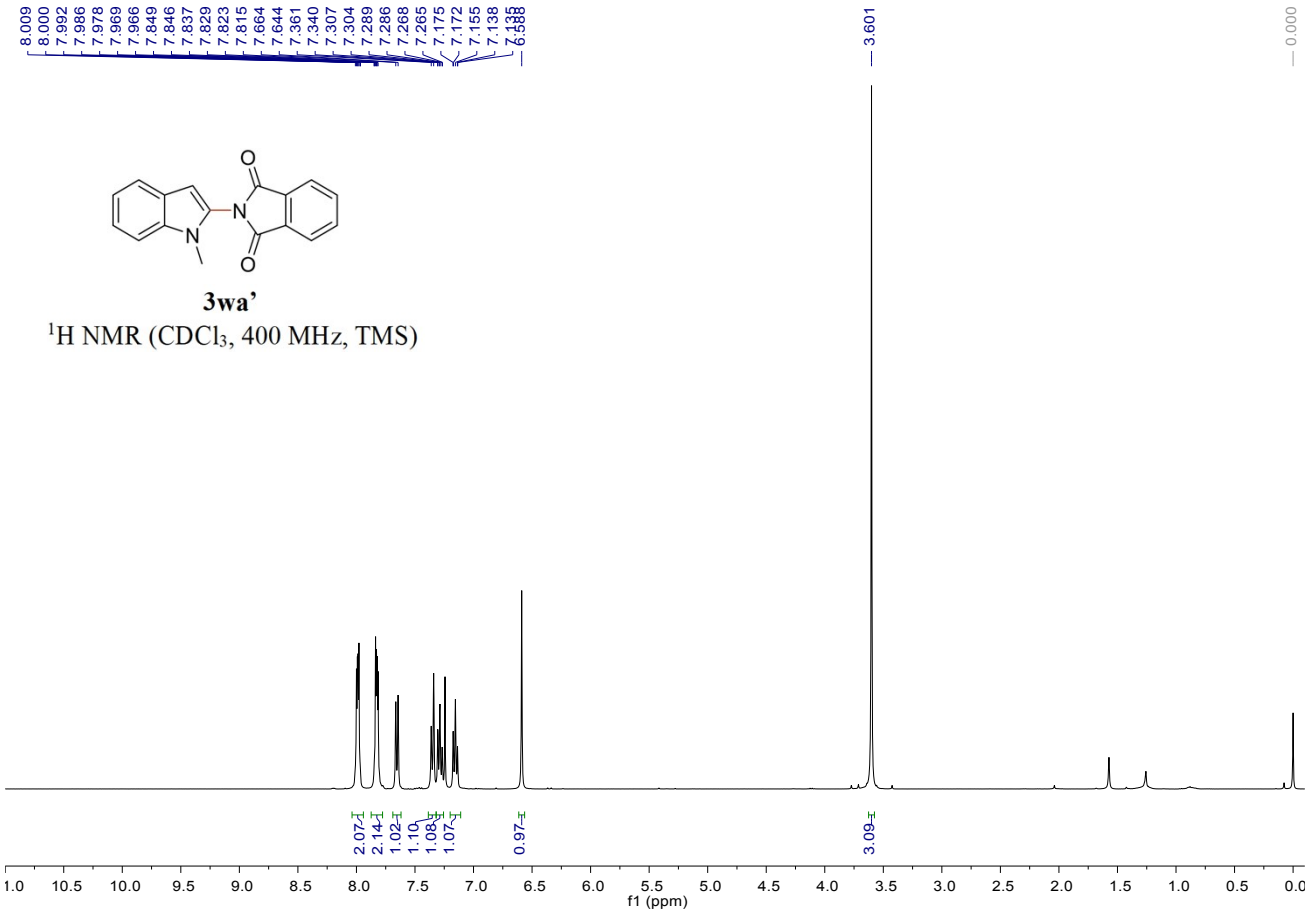


3wa

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

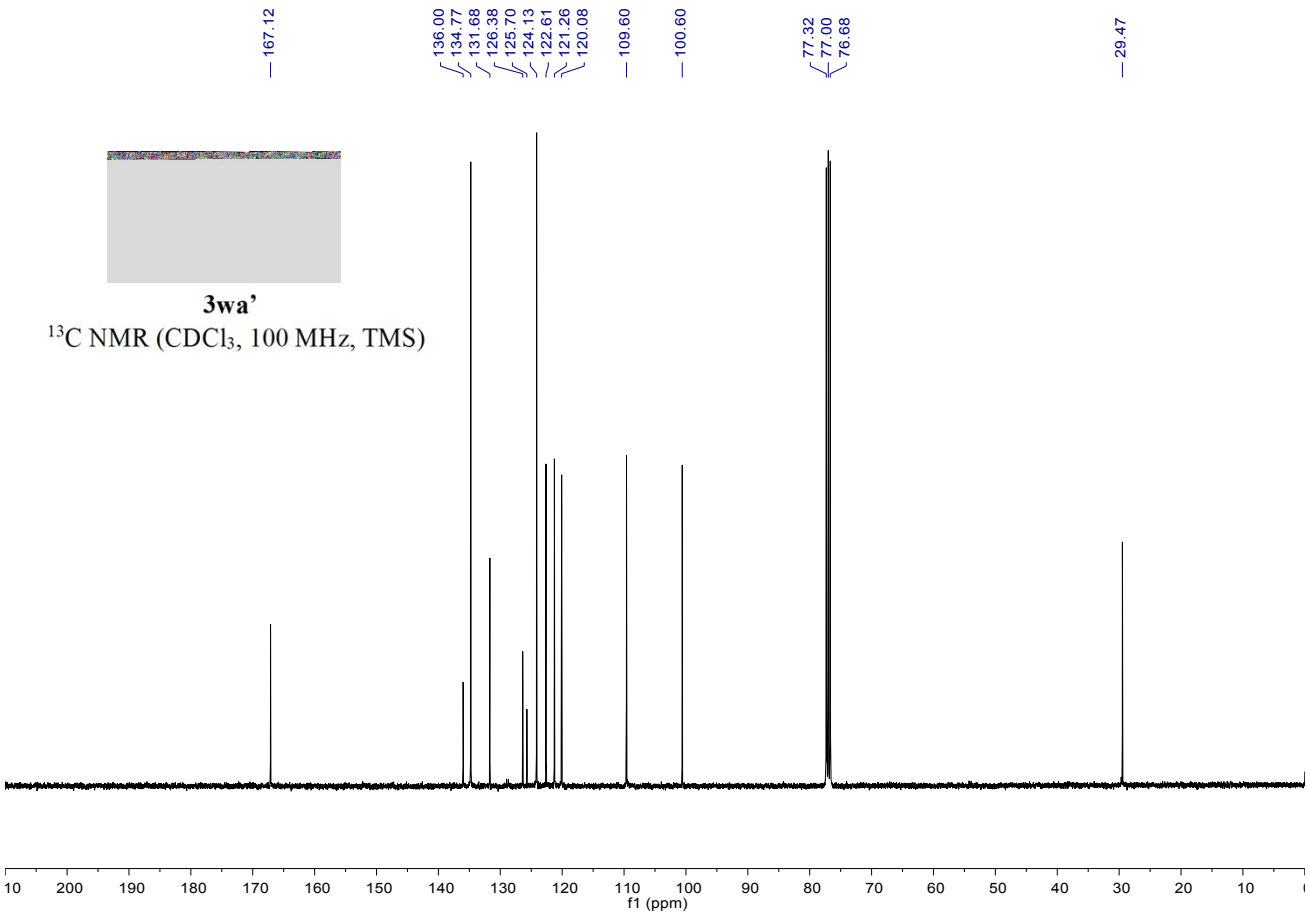


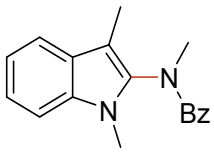
2-(1-methyl-1H-indol-2-yl)isoindoline-1,3-dione (3wa'). The title product was obtained after purification by silica gel column chromatography (DCM:PE = 1:1 to PE:EA = 4:1). An yellow solid, 28 mg, 50% yield; M.p.: 227-229 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.99 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.83 (dd, *J* = 5.5, 3.1 Hz, 2H), 7.65 (d, *J* = 7.9 Hz, 1H), 7.35 (d, *J* = 8.2 Hz, 1H), 7.32 - 7.26 (m, 1H), 7.20 - 7.11 (m, 1H), 6.59 (s, 1H), 3.60 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 167.1, 136.0, 134.8, 131.7, 126.4, 125.7, 124.1, 122.6, 121.3, 120.1, 109.6, 100.6, 29.5; IR (neat) ν 1791, 1714, 1551, 1439, 1378, 1349, 1304, 1236, 1077, 880, 796, 751, 733, 714, 673 cm^{-1}.



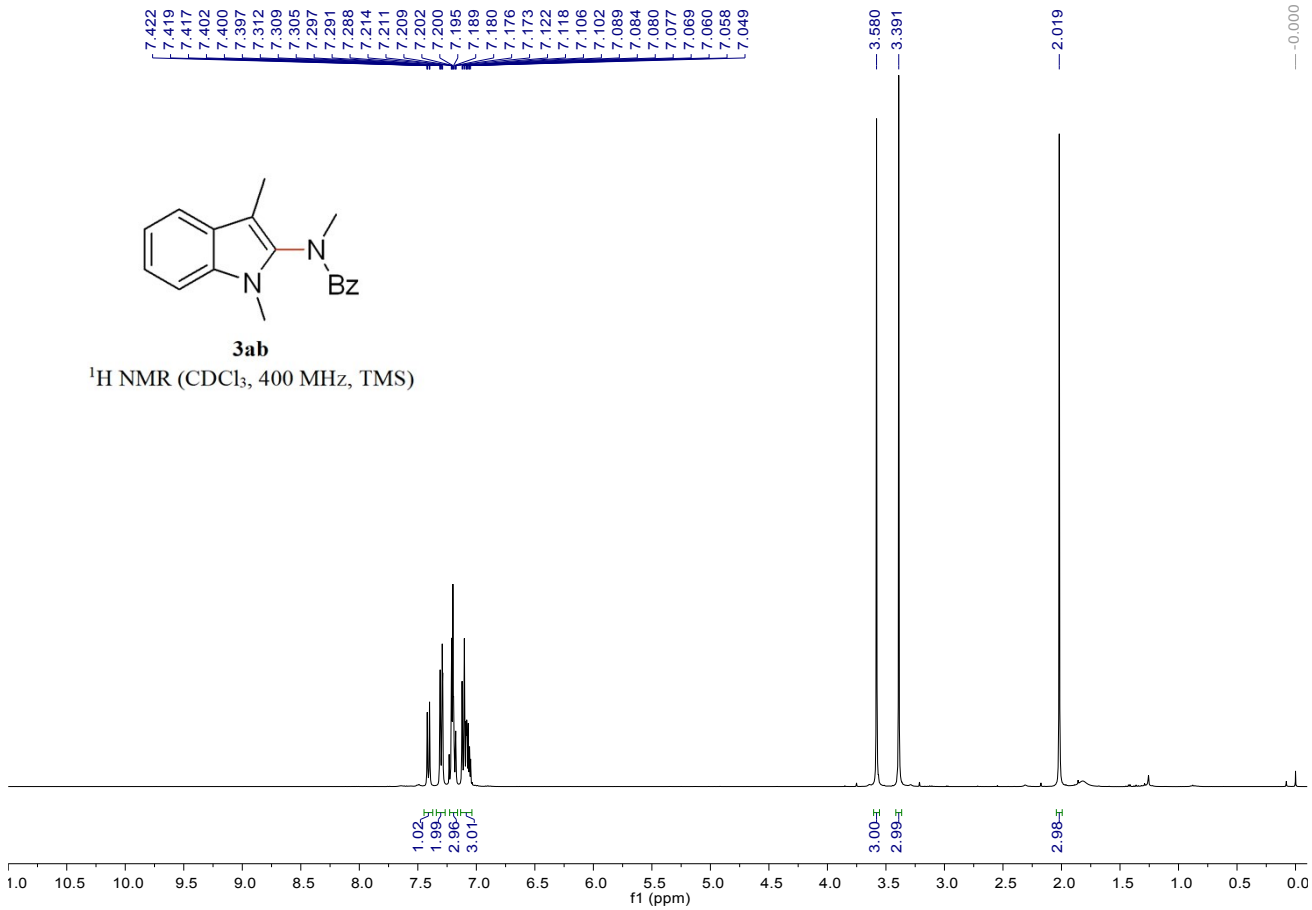
3wa'

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)



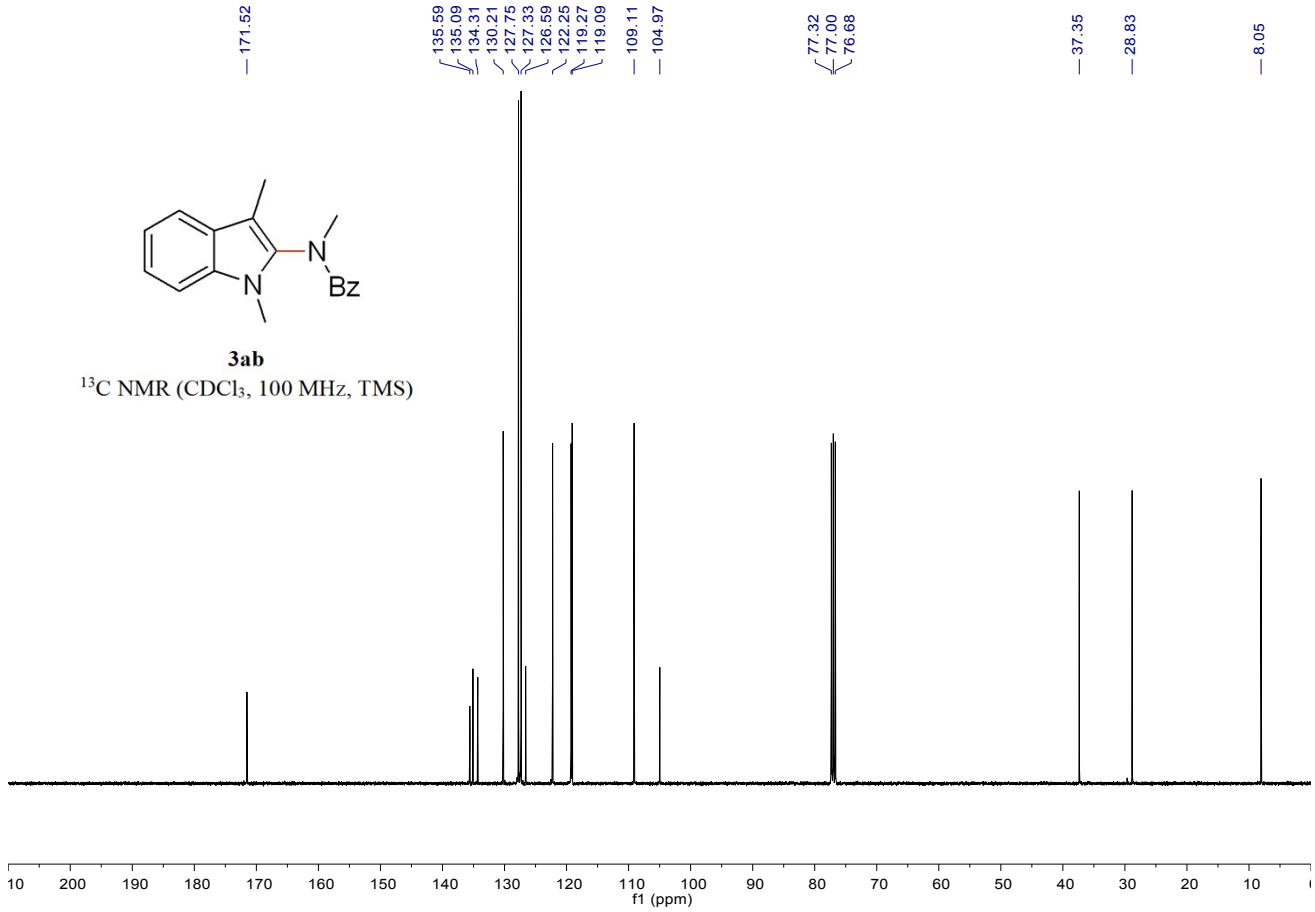


***N*-(1,3-dimethyl-1H-indol-2-yl)-*N*-methylbenzamide (3ab)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 40 mg, 72% yield; M.p.: 112 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.41 (dt, *J* = 7.9, 1.0 Hz, 1H), 7.34 - 7.27 (m, 2H), 7.23 - 7.16 (m, 3H), 7.13 - 7.04 (m, 3H), 3.58 (s, 3H), 3.39 (s, 3H), 2.02 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.5, 135.6, 135.1, 134.3, 130.2, 127.7, 127.3, 126.6, 122.3, 119.3, 119.1, 109.1, 105.0, 37.4, 28.8, 8.1; IR (neat) ν 2915, 1652, 1467, 1449, 1337, 1265, 1097, 1013, 782, 756, 712, 690, 679 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1307.



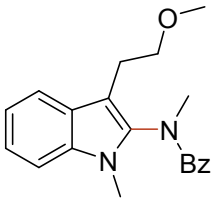
3ab

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

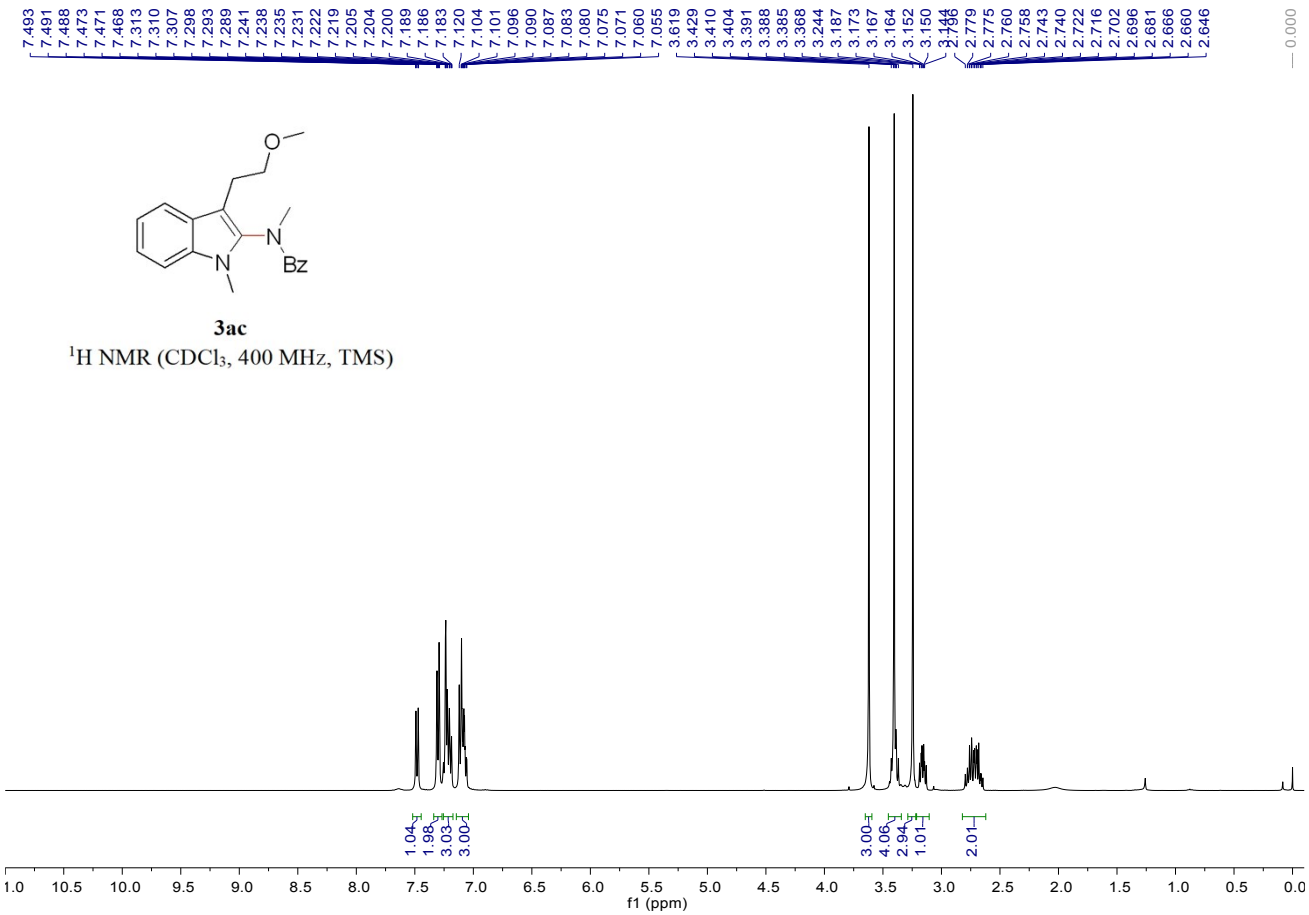


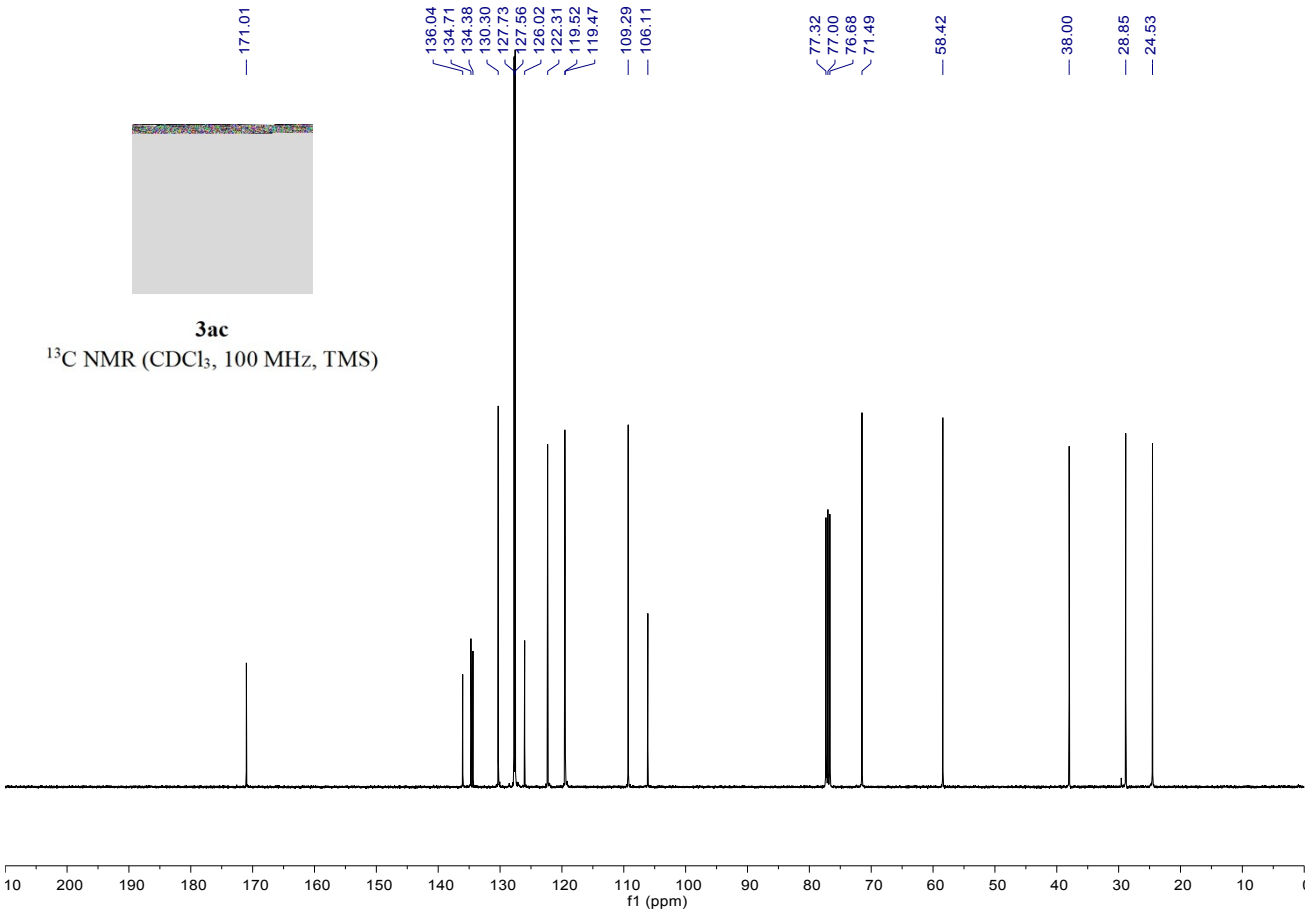
3ab

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



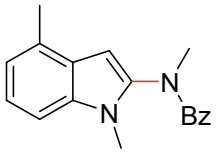
***N*-(3-(2-methoxyethyl)-1-methyl-1H-indol-2-yl)-*N*-methylbenzamide (3ac)**. The title product was obtained after purification by prep-TLC (PE:EA = 2:1). An off-white solid, 53 mg, 82% yield; M.p.: 120 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.48 (dt, *J* = 8.0, 1.0 Hz, 1H), 7.30 (dt, *J* = 7.1, 1.4 Hz, 2H), 7.26 - 7.17 (m, 3H), 7.15 - 7.04 (m, 3H), 3.62 (s, 3H), 3.45 - 3.34 (m, 4H), 3.24 (s, 3H), 3.21 - 3.10 (m, 1H), 2.82 - 2.62 (m, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.0, 136.0, 134.7, 134.4, 130.3, 127.7, 127.6, 126.0, 122.3, 119.5, 119.5, 109.3, 106.1, 71.5, 58.4, 38.0, 28.9, 24.5; IR (neat) ν 2926, 1652, 1465, 1435, 1404, 1345, 1289, 1258, 1103, 1014, 963, 925, 794, 750, 718, 695, 661 cm^{-1}; HRMS (ESI) Calcd. for $C_{20}H_{22}N_2O_2Na^+$ Requires: 345.1574, Found: 345.1572.



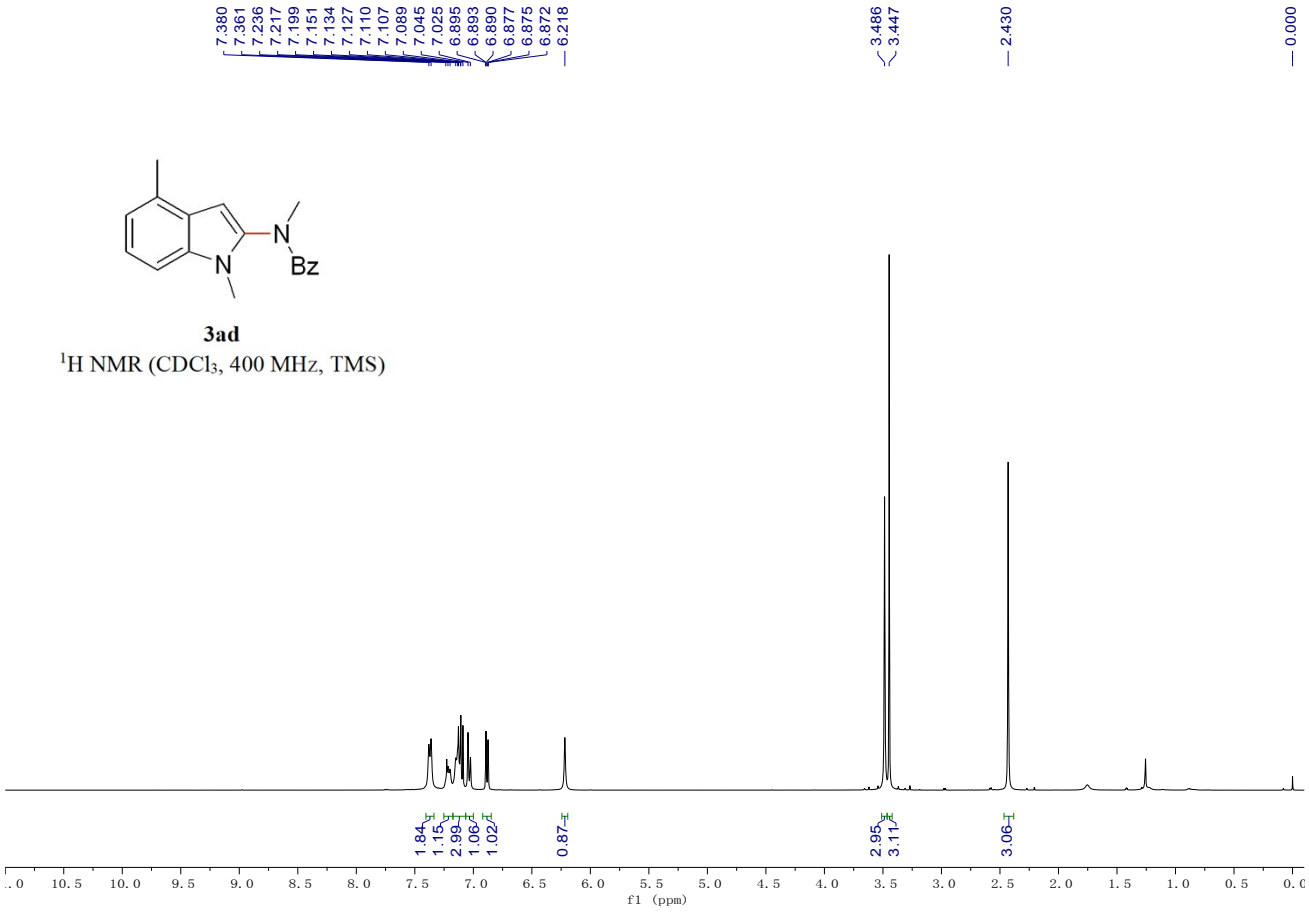


3ac

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

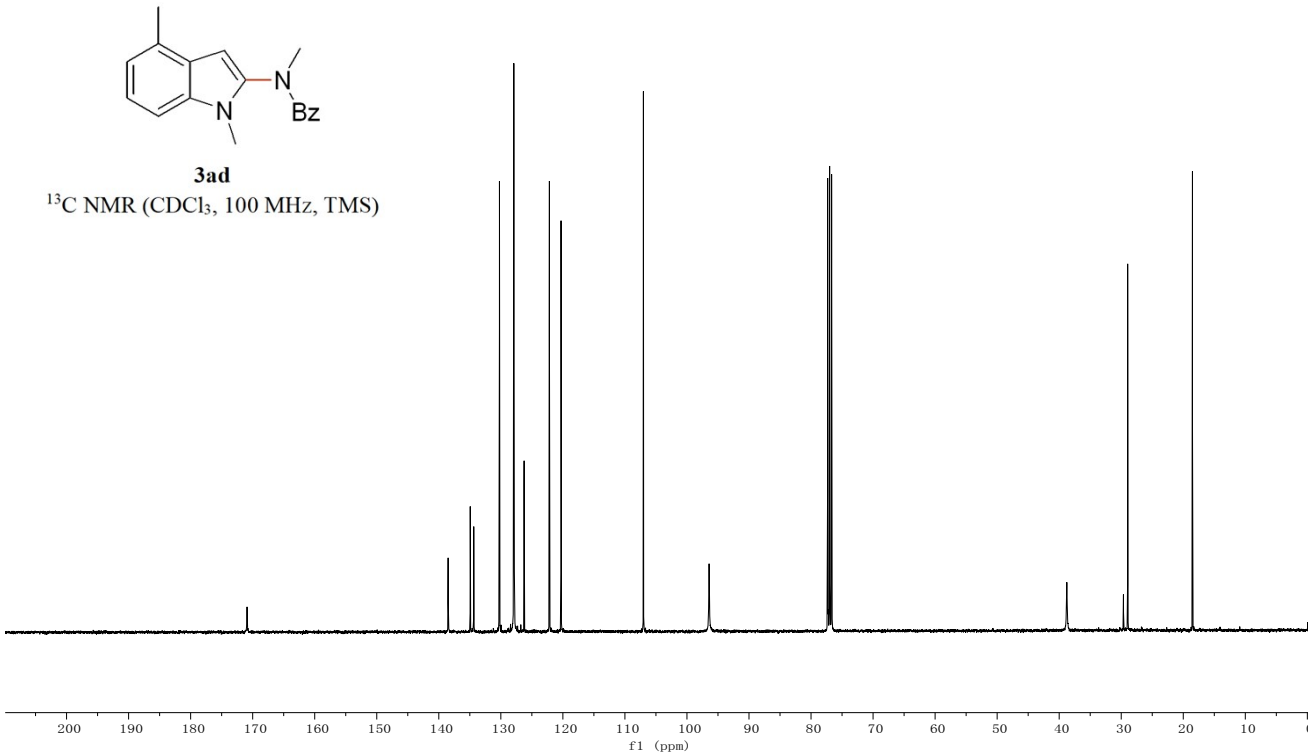


***N*-(1,4-dimethyl-1H-indol-2-yl)-*N*-methylbenzamide (3ad)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 43 mg, 77% yield; M.p.: 135 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.37 (d, J = 7.6 Hz, 2H), 7.25 - 7.17 (m, 1H), 7.17 - 7.07 (m, 3H), 7.04 (d, J = 8.2 Hz, 1H), 6.88 (dt, J = 7.1, 1.0 Hz, 1H), 6.22 (s, 1H), 3.49 (s, 3H), 3.45 (s, 3H), 2.43 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 138.5, 134.9, 134.4, 130.3, 130.2, 127.91, 127.87, 126.2, 122.2, 120.3, 107.0, 96.4, 38.8, 29.0, 18.5; IR (neat) ν 2917, 1647, 1551, 1416, 1343, 1297, 1250, 1135, 1067, 1006, 948, 794, 766, 748, 719, 707, 663 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1306.



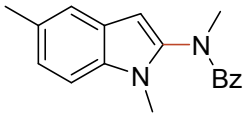
3ad

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

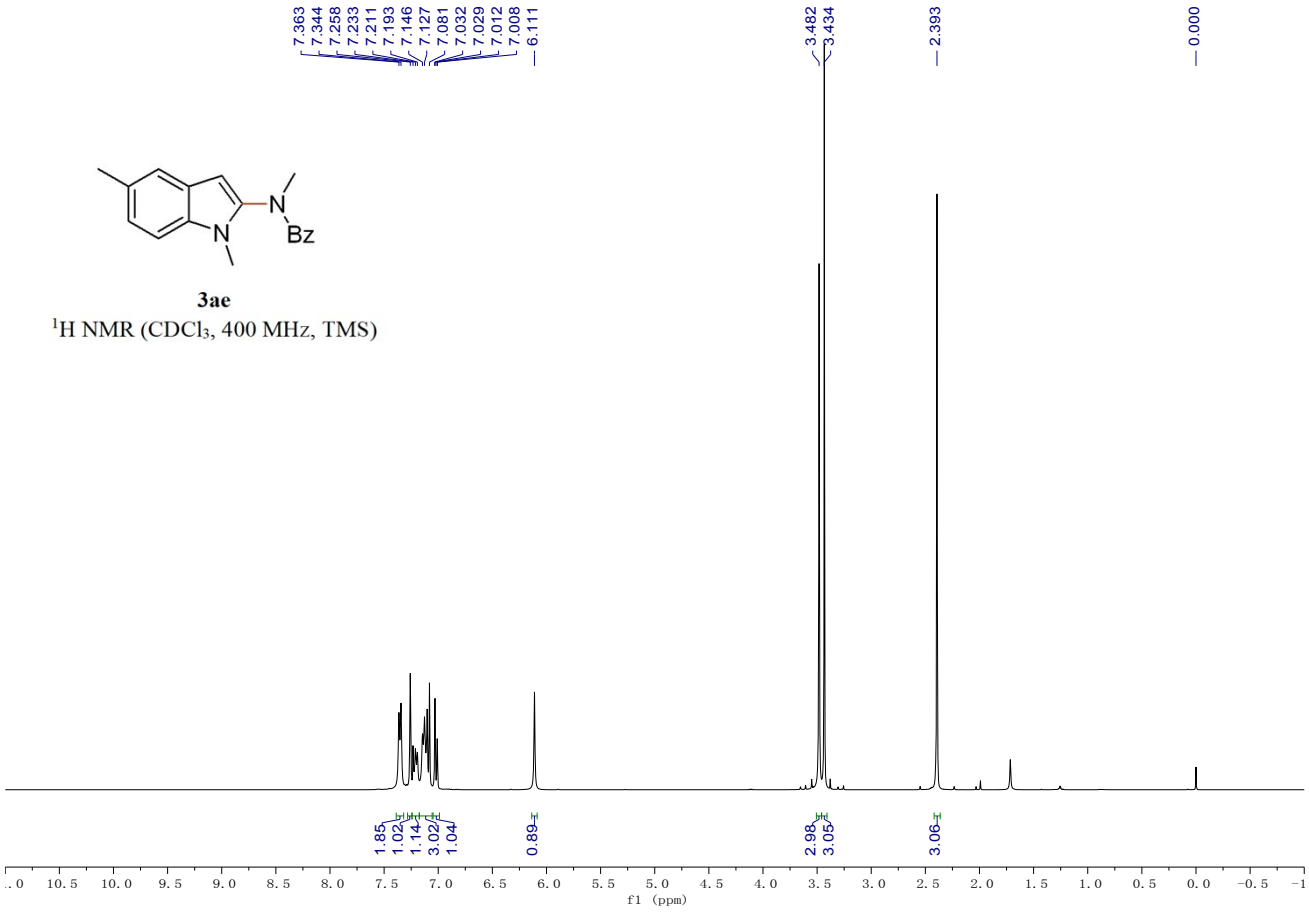


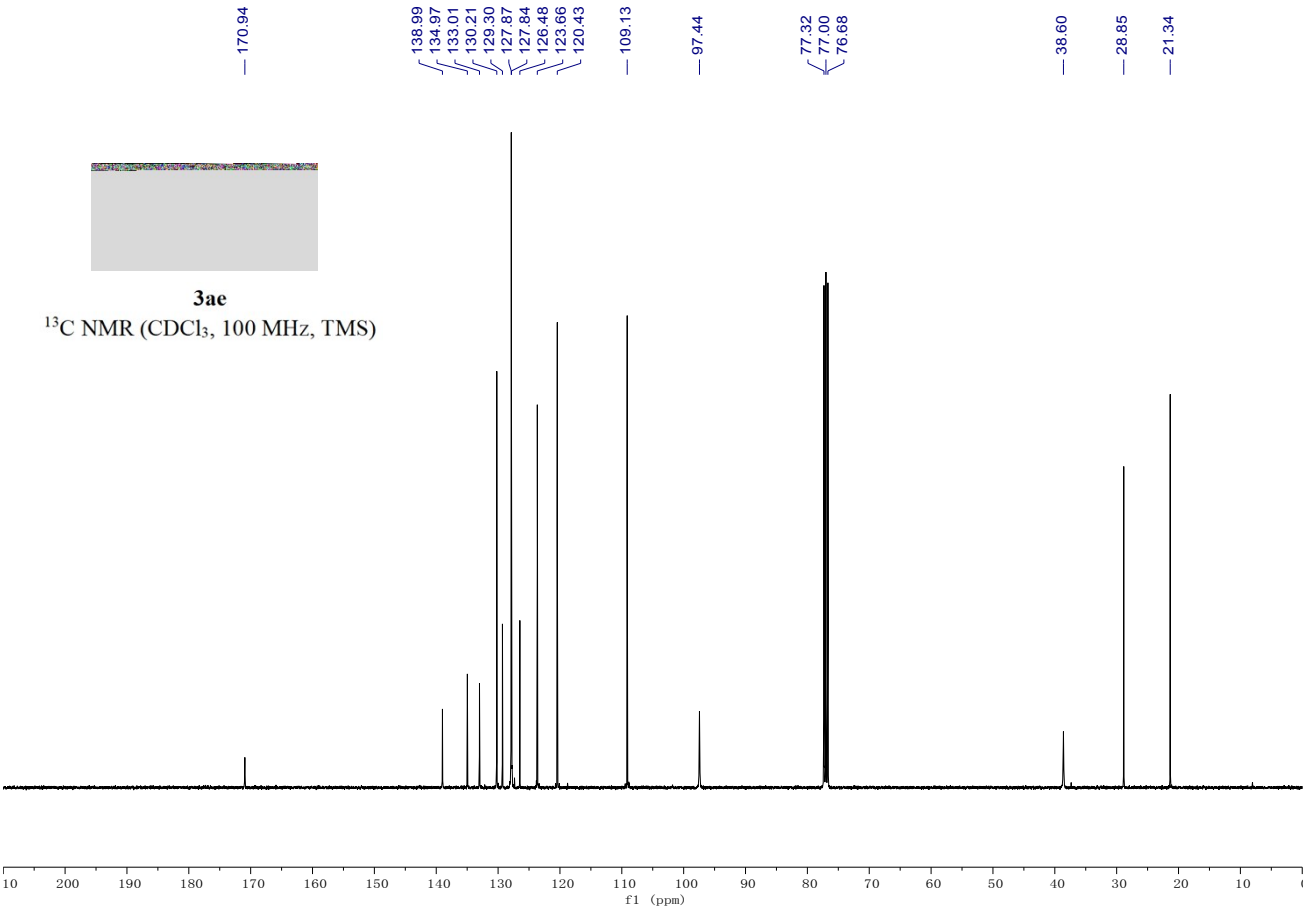
3ad

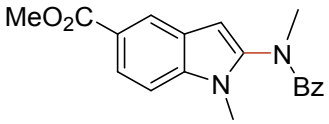
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



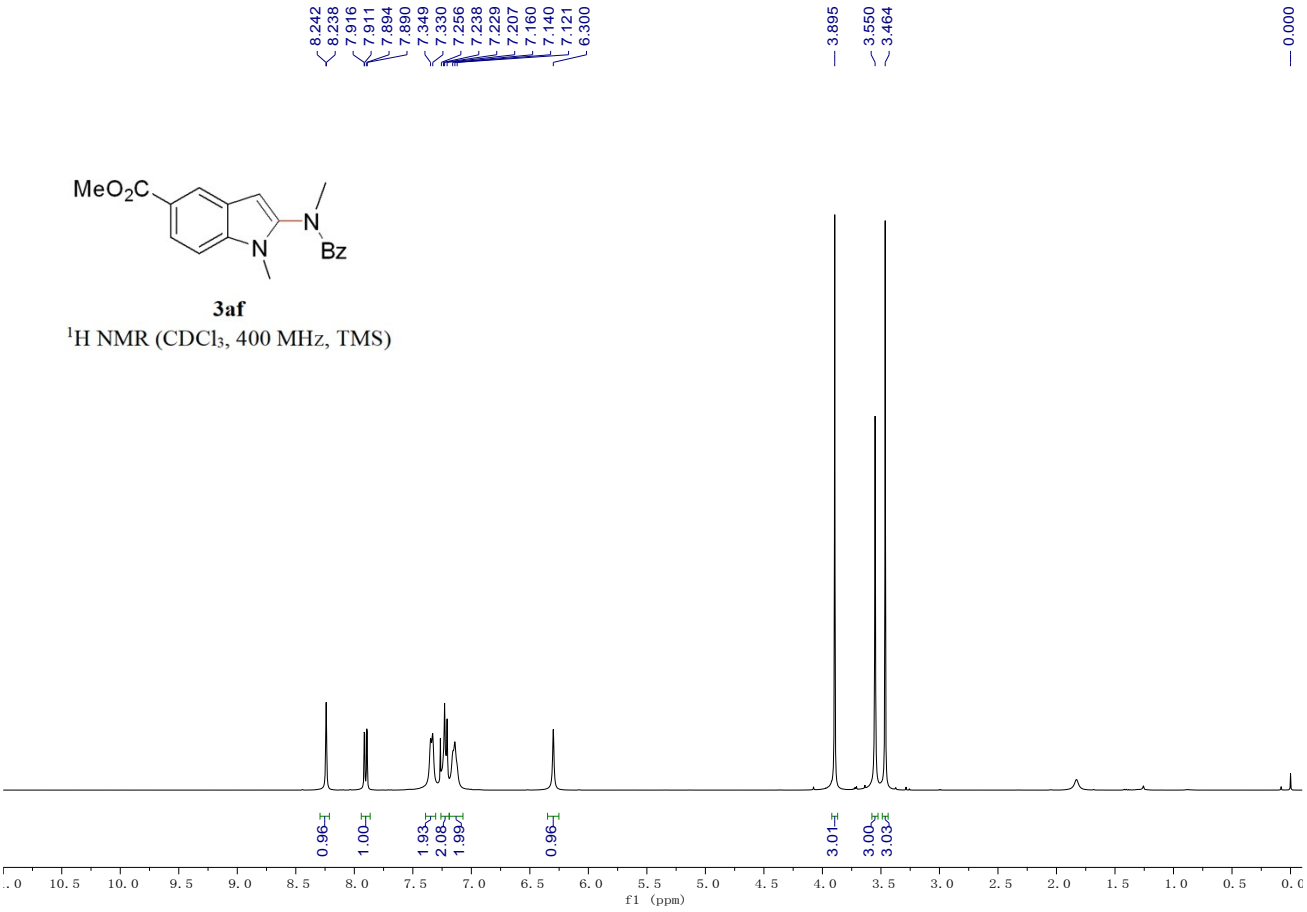
***N*-(1,5-dimethyl-1H-indol-2-yl)-*N*-methylbenzamide (3ae)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid 35 mg, 62% yield; M.p.: 114 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.35 (d, *J* = 7.6 Hz, 2H), 7.26 (s, 1H), 7.24 - 7.17 (m, 1H), 7.17 - 7.06 (m, 3H), 7.02 (dd, *J* = 8.4, 1.6 Hz, 1H), 6.11 (s, 1H), 3.48 (s, 3H), 3.43 (s, 3H), 2.39 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 139.0, 135.0, 133.0, 130.2, 129.3, 127.9, 127.8, 126.5, 123.7, 120.4, 109.1, 97.4, 38.6, 28.9, 21.3; IR (neat) ν 2922, 1666, 1652, 1558, 1486, 1446, 1391, 1334, 1252, 1086, 1006, 795, 720, 698, 668 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1312.





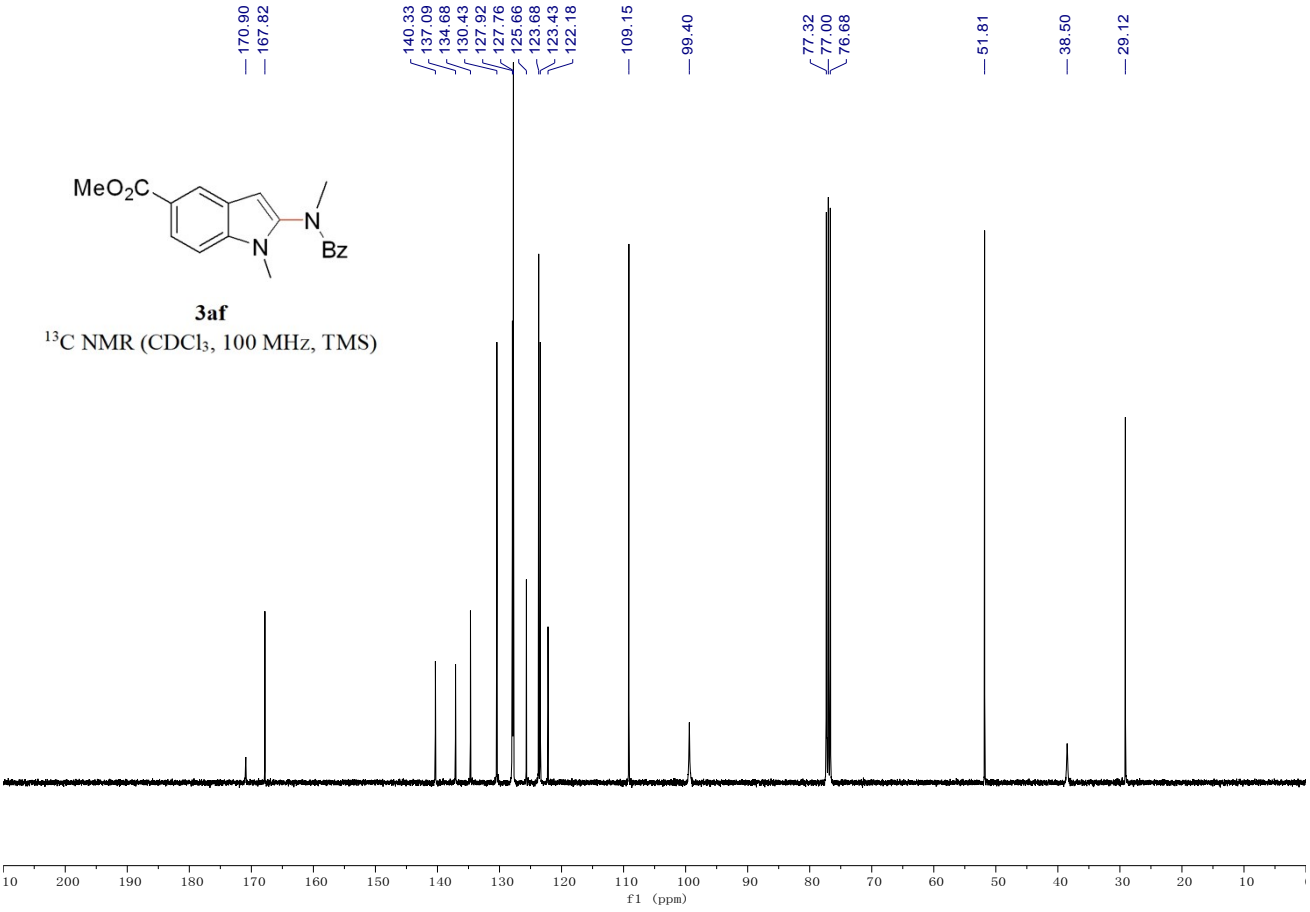


methyl 1-methyl-2-(*N*-methylbenzamido)-1H-indole-5-carboxylate (3af). The title product was obtained after purification by prep-TLC (PE:EA = 2:1). A yellow solid, 32 mg, 50% yield; M.p.: 174 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 8.24 (d, *J* = 1.6 Hz, 1H), 7.90 (dd, *J* = 8.7, 1.7 Hz, 1H), 7.34 (d, *J* = 7.6 Hz, 2H), 7.26 - 7.19 (m, 2H), 7.19 - 7.07 (m, 2H), 6.30 (s, 1H), 3.89 (s, 3H), 3.55 (s, 3H), 3.46 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 167.8, 140.3, 137.1, 134.7, 130.4, 127.9, 127.8, 125.7, 123.7, 123.4, 122.2, 109.2, 99.4, 51.8, 38.5, 29.1; IR (neat) ν 2954, 1698, 1655, 1563, 1446, 1427, 1337, 1307, 1255, 1159, 1104, 1083, 971, 922, 820, 769, 719, 696, 677 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{18}N_2O_3Na^+$ Requires: 345.1210, Found: 345.1208.



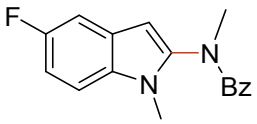
3af

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

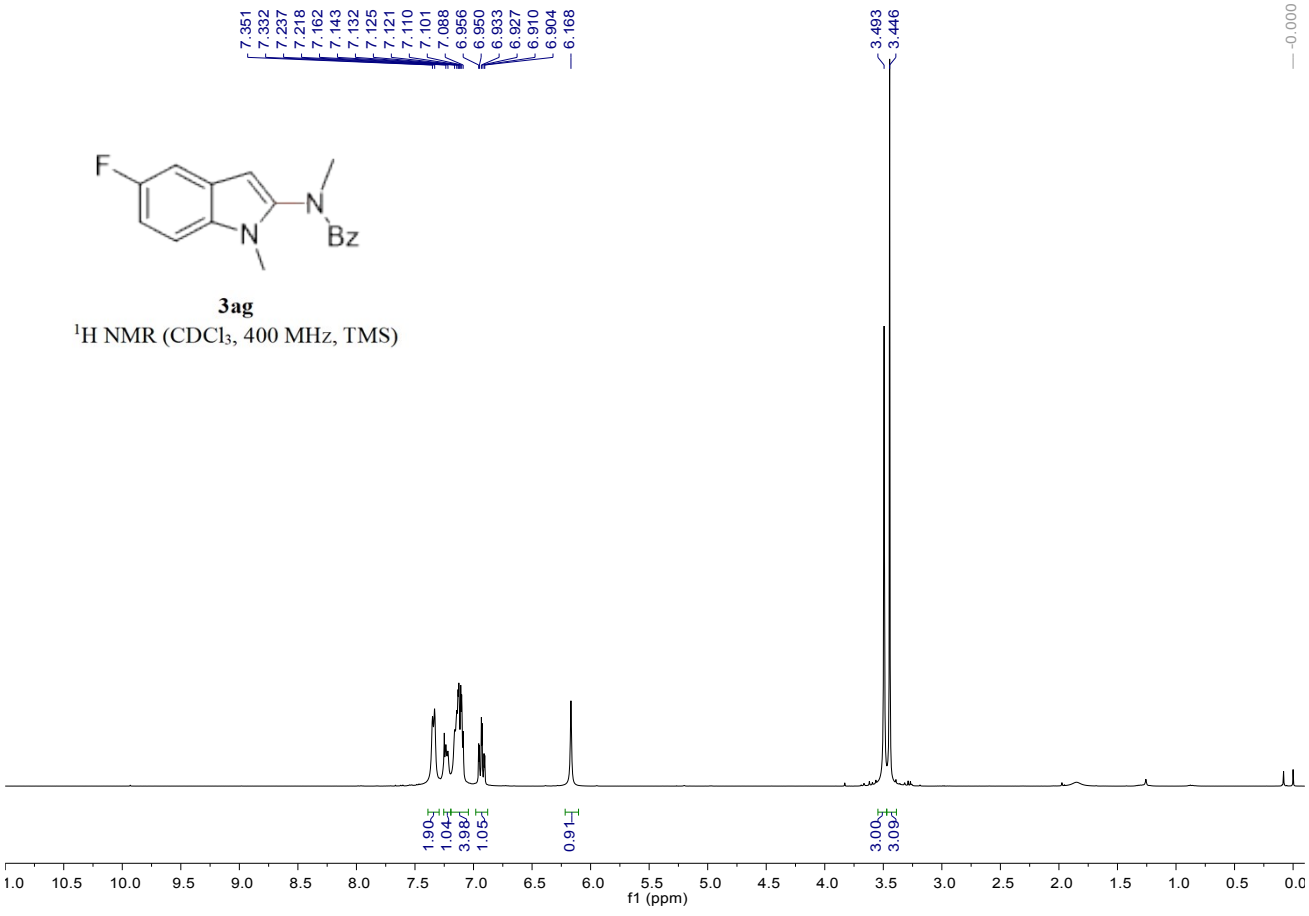


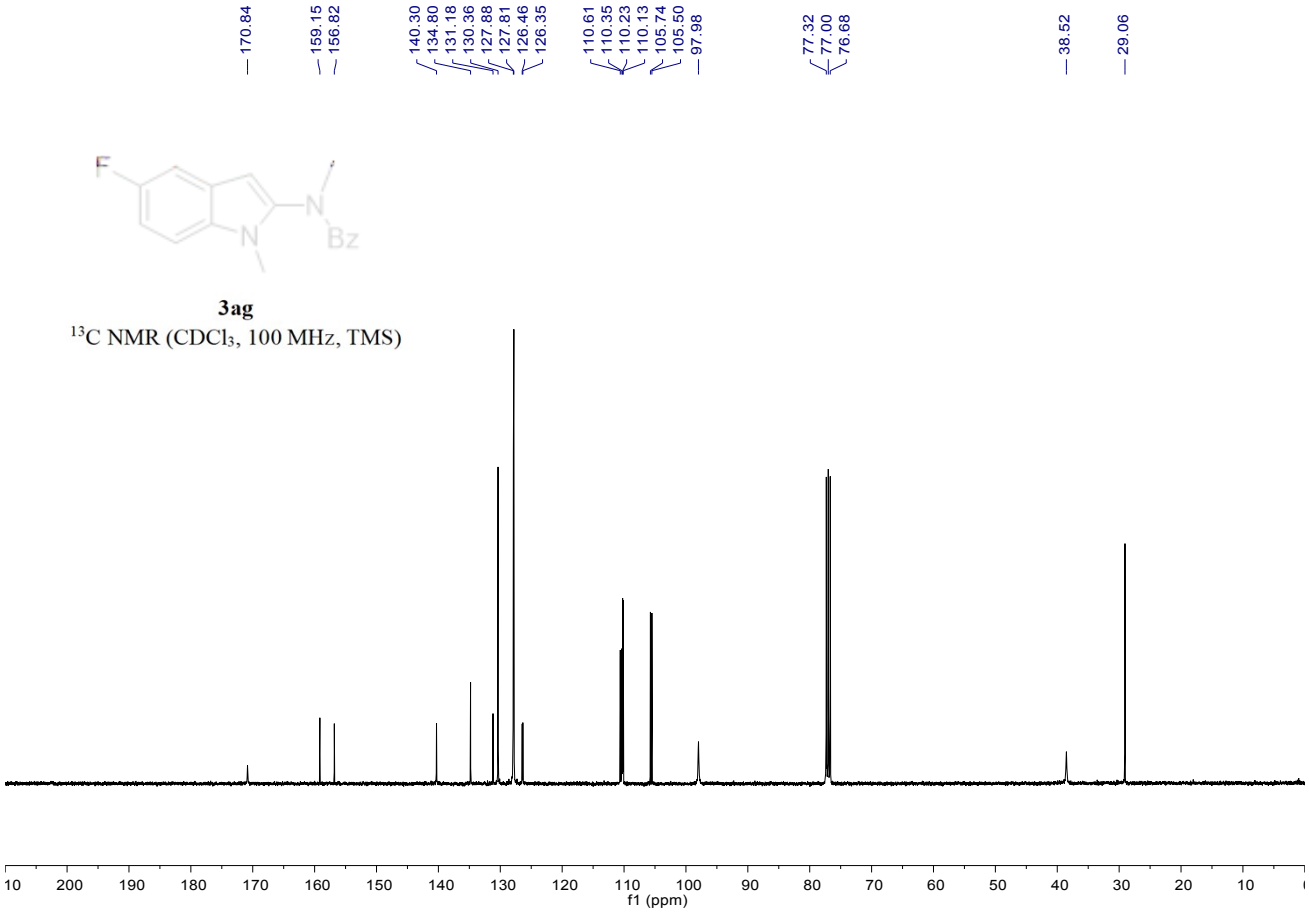
3af

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



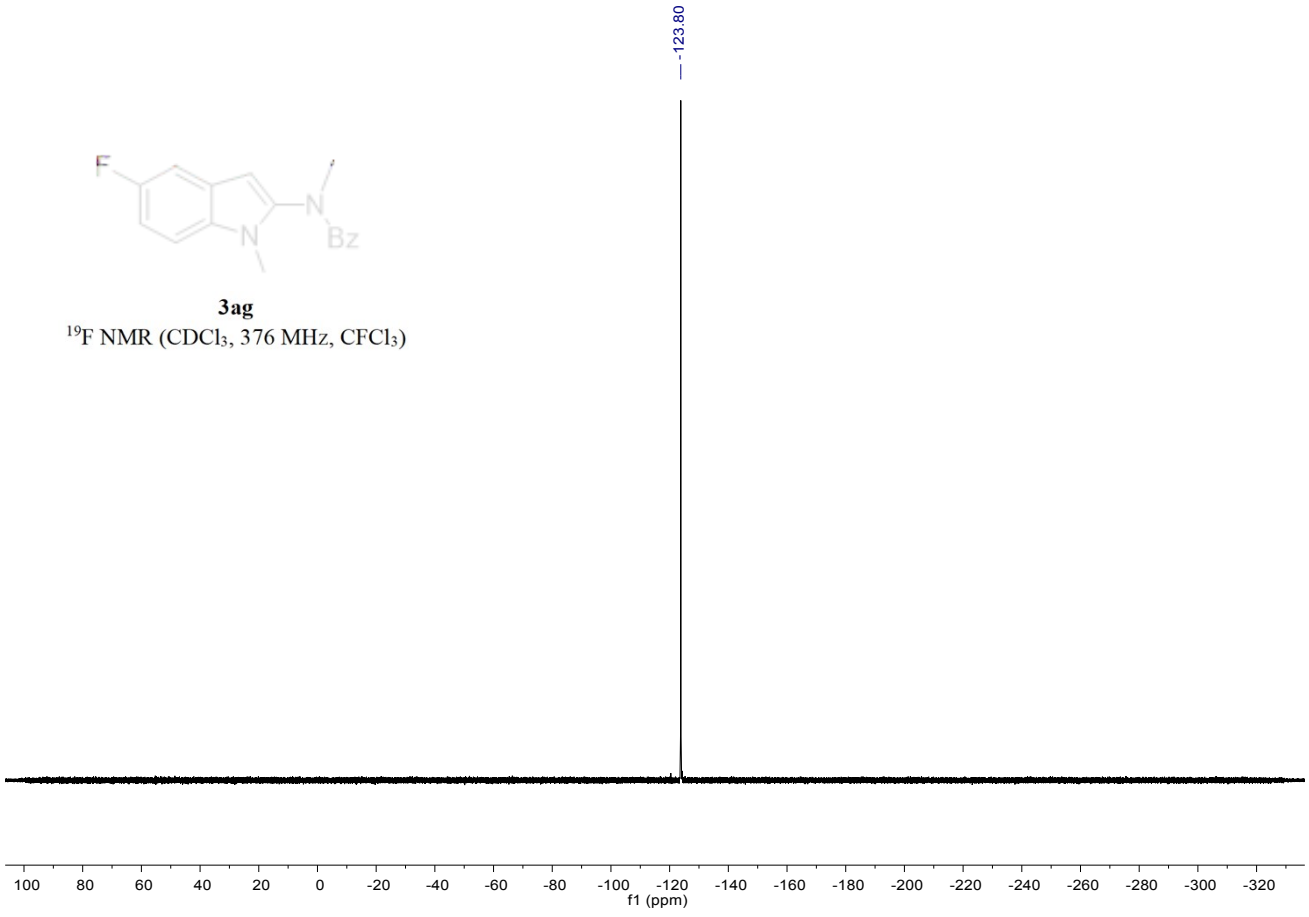
N-(5-fluoro-1-methyl-1H-indol-2-yl)-N-methylbenzamide (3ag). The title product was obtained after purification by prep-TLC (DCM). A pale yellow solid, 42 mg, 75% yield; M.p.: 104 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.34 (d, *J* = 7.6 Hz, 2H), 7.25 - 7.19 (m, 1H), 7.19 - 7.04 (m, 4H), 6.93 (td, *J* = 9.1, 2.5 Hz, 1H), 6.17 (s, 1H), 3.49 (s, 3H), 3.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.8, 158.0 (d, *J* = 235.0 Hz), 140.3, 134.8, 131.2, 130.4, 127.9, 127.8, 126.4 (d, *J* = 10.4 Hz), 110.5 (d, *J* = 26.2 Hz), 110.2 (d, *J* = 9.6 Hz), 105.6 (d, *J* = 23.7 Hz), 98.0, 38.5, 29.1; ^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$) δ -123.8; IR (neat) ν 3063, 1655, 1583, 1480, 1435, 1399, 1345, 1251, 1189, 1094, 1010, 953, 886, 804, 765, 718, 691, 672 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2OFNa^+$ Requires: 305.1061, Found: 305.1059.





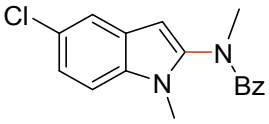
3ag

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

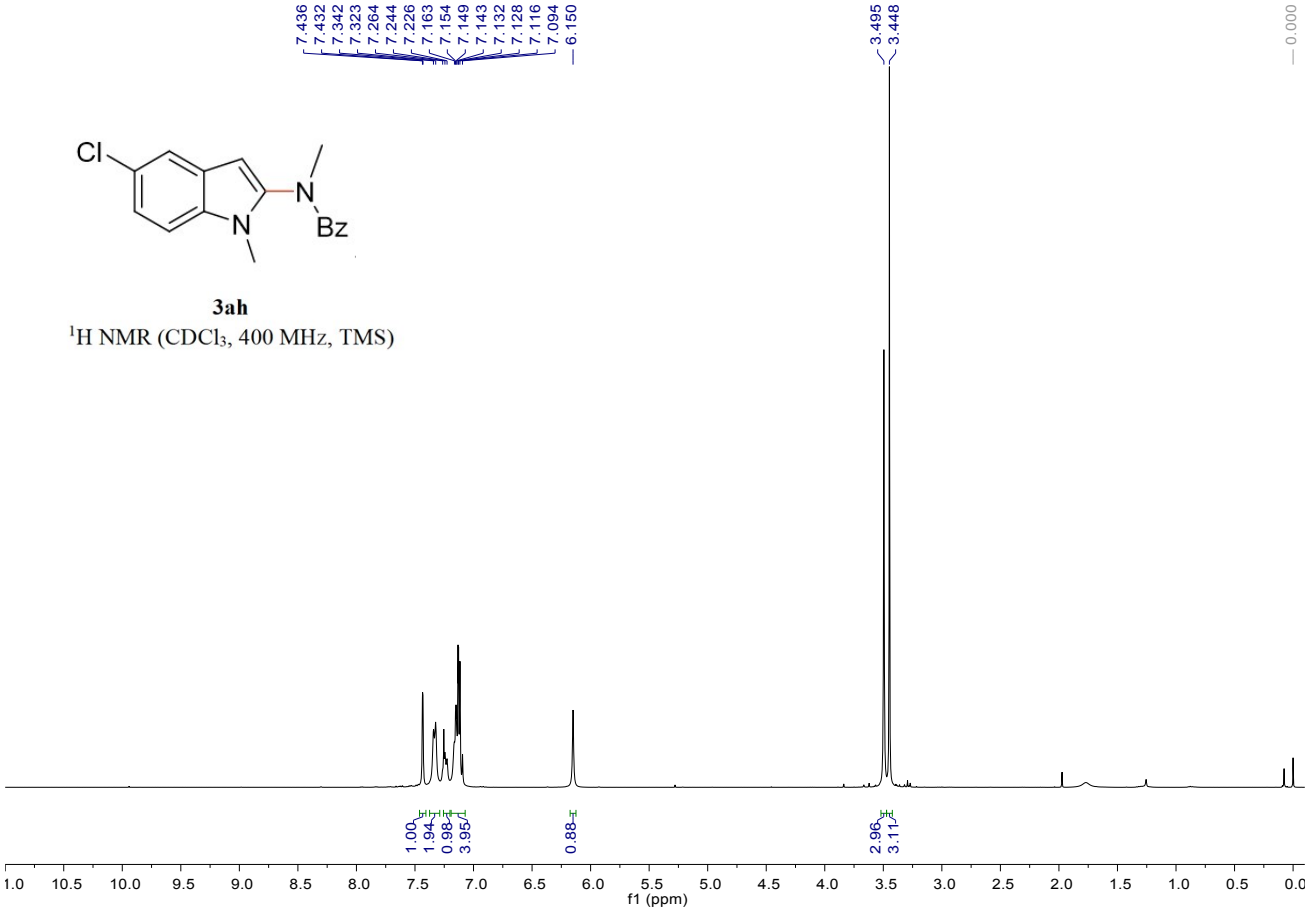


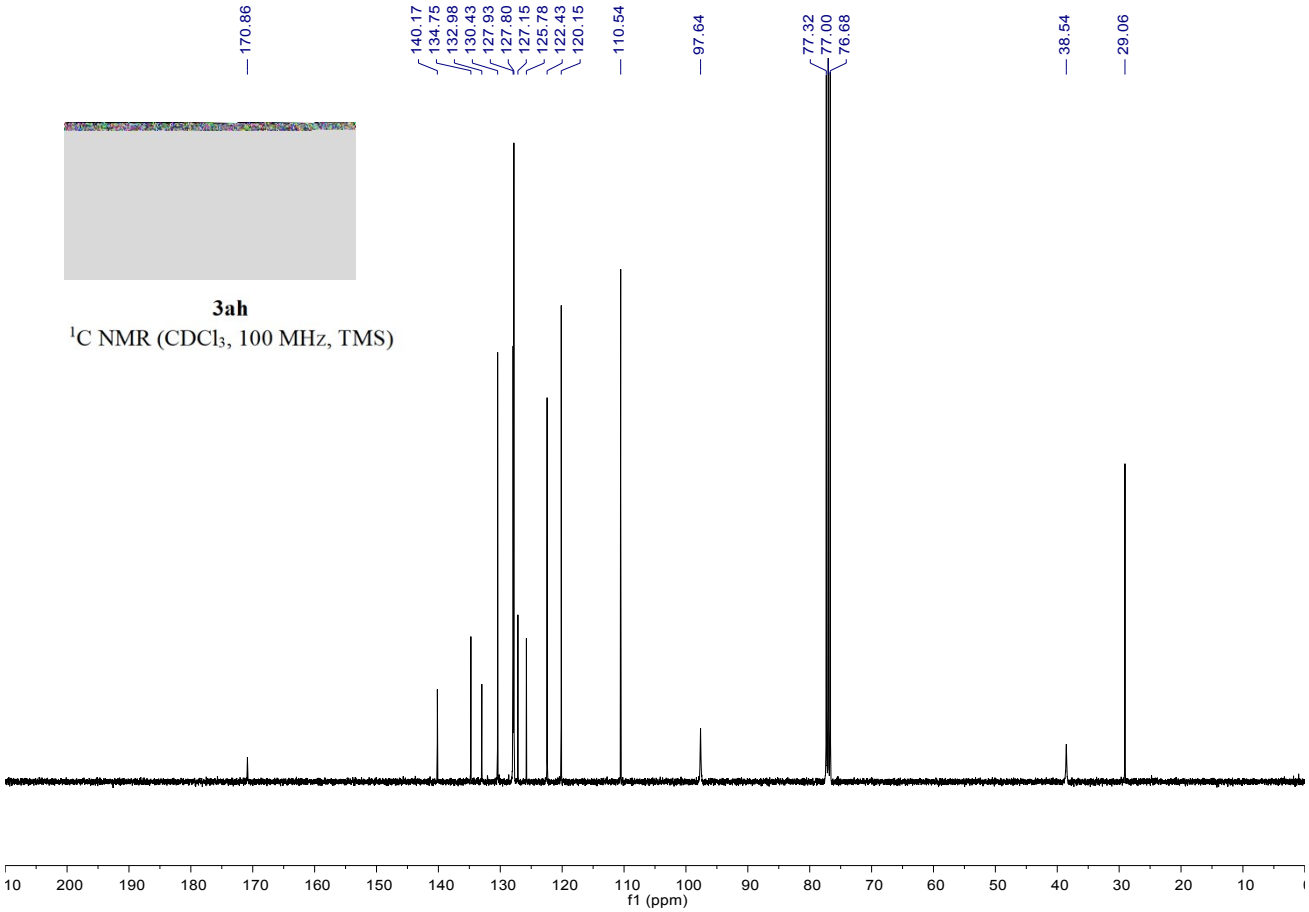
3ag

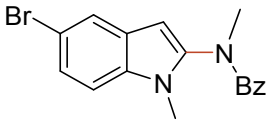
^{19}F NMR ($CDCl_3$, 376 MHz, $CFCl_3$)



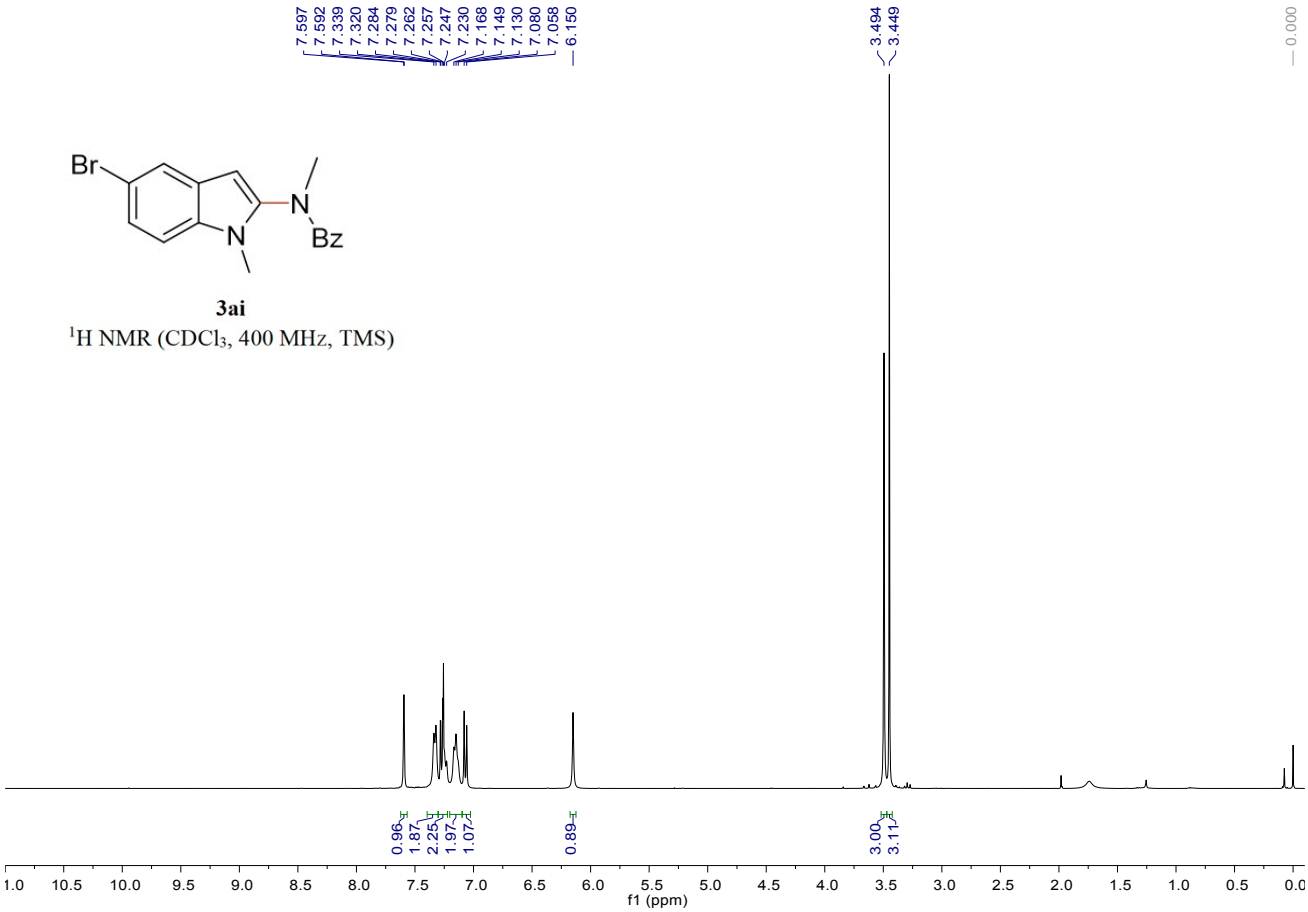
N-(5-chloro-1-methyl-1H-indol-2-yl)-N-methylbenzamide (3ah). The title product was obtained after purification by prep-TLC (DCM). A yellow solid, 38 mg, 63% yield; M.p.: 110 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.43 (d, *J* = 1.9 Hz, 1H), 7.33 (d, *J* = 7.6 Hz, 2H), 7.26 - 7.20 (m, 1H), 7.19 - 7.07 (m, 4H), 6.15 (s, 1H), 3.50 (s, 3H), 3.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 140.2, 134.8, 133.0, 130.4, 127.9, 127.8, 127.2, 125.8, 122.4, 120.1, 110.5, 97.6, 38.5, 29.1; IR (neat) ν 2920, 1651, 1572, 1473, 1344, 1249, 1344, 1328, 1249, 1093, 1063, 1009, 920, 866, 787, 713, 692 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2ONaCl^+$ Requires: 321.0765, Found: 321.0766.





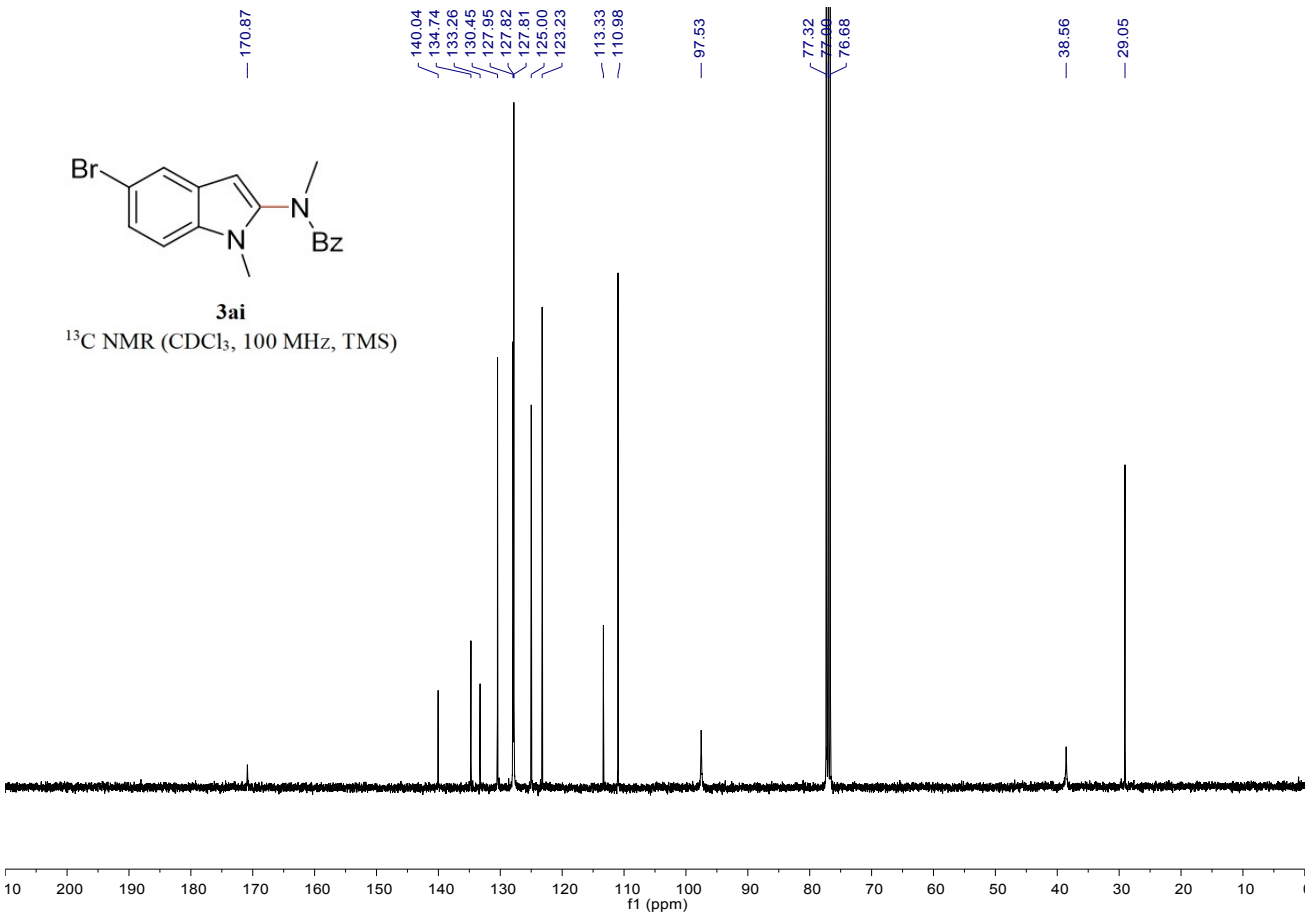


N-(5-bromo-1-methyl-1H-indol-2-yl)-N-methylbenzamide (3ai). The title product was obtained after purification by prep-TLC (DCM). A pale yellow solid, 49 mg, 72% yield; M.p.: 143 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.59 (d, *J* = 1.9 Hz, 1H), 7.33 (d, *J* = 7.6 Hz, 2H), 7.30 - 7.21 (m, 2H), 7.15 (t, *J* = 7.5 Hz, 2H), 7.07 (d, *J* = 8.7 Hz, 1H), 6.15 (s, 1H), 3.49 (s, 3H), 3.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.9, 140.0, 134.7, 133.3, 130.4, 127.9, 127.82, 127.81, 125.0, 123.2, 113.3, 111.0, 97.5, 38.6, 29.1; IR (neat) ν 2918, 1654, 1569, 1470, 1394, 1327, 1093, 1010, 867, 784, 713, 686 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{15}N_2ONaBr^+$ Requires: 365.0260, Found: 365.0262.



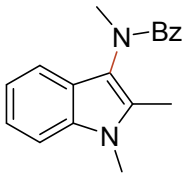
3ai

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

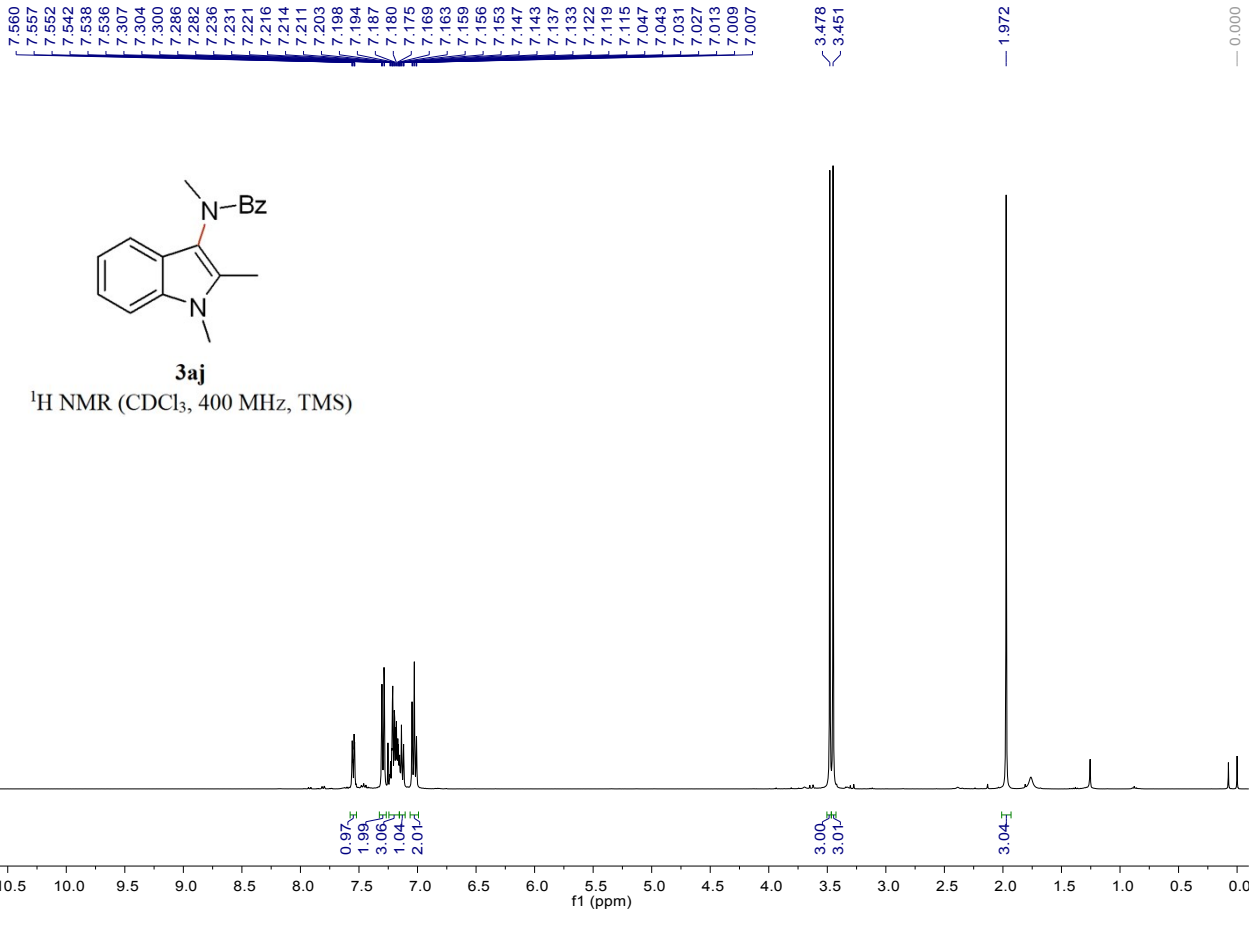


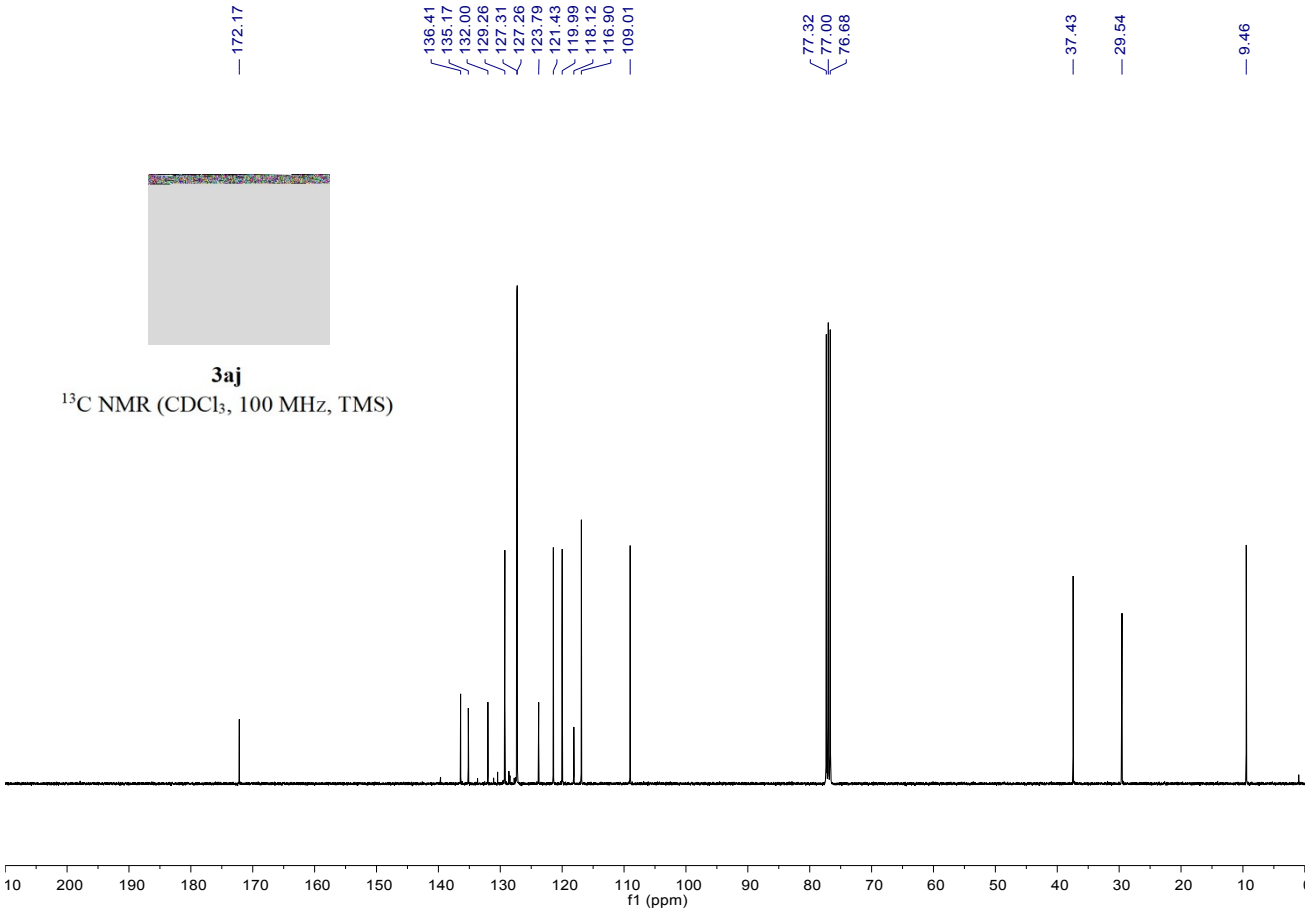
3ai

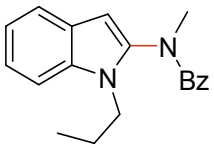
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



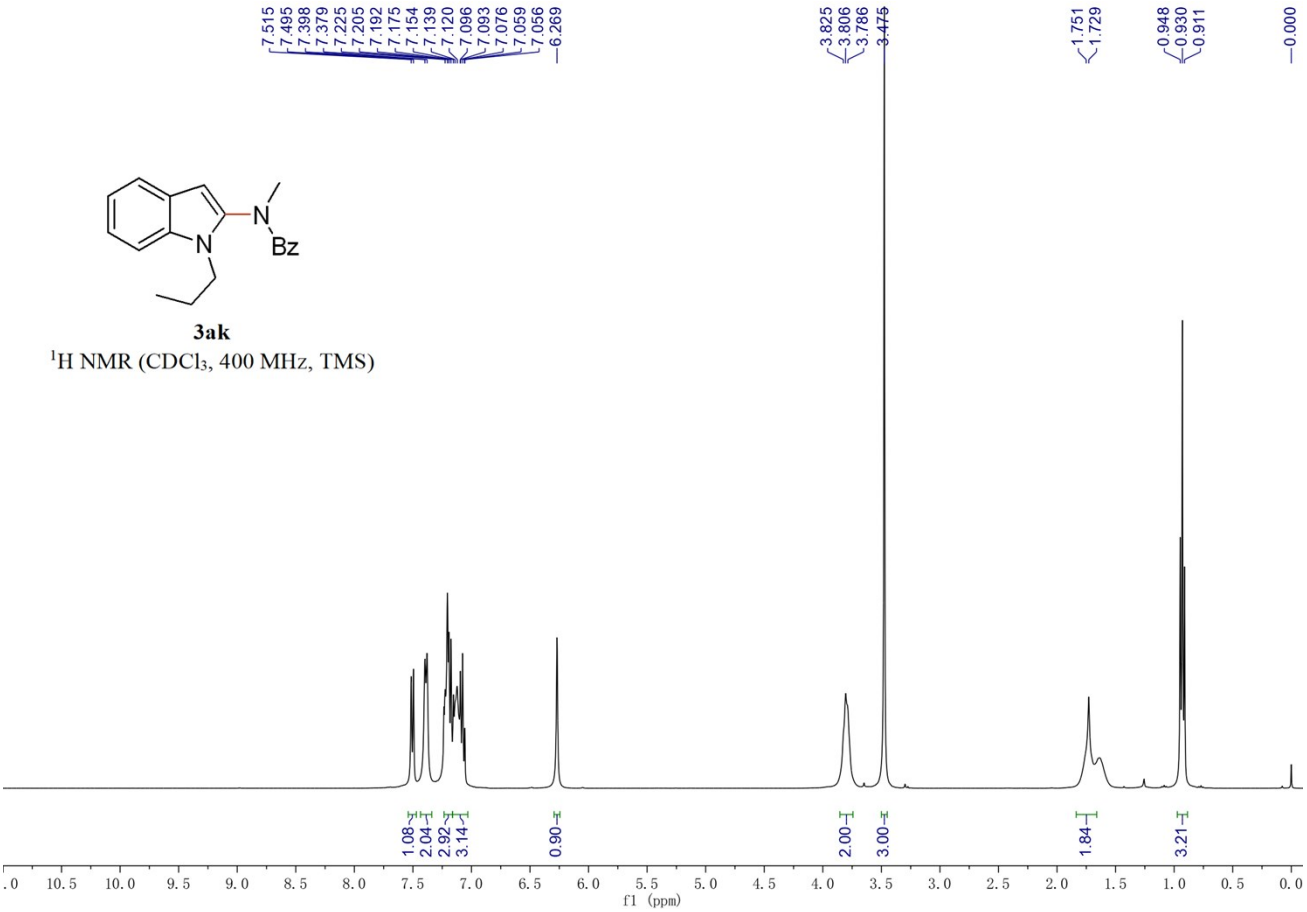
***N*-(1,2-dimethyl-1H-indol-3-yl)-*N*-methylbenzamide (3aj)**. The title product was obtained after purification by prep-TLC (PE:EA = 4:1). A yellow solid, 22 mg, 39% yield; M.p.: 164 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.59 - 7.51 (m, 1H), 7.33 - 7.26 (m, 2H), 7.26 - 7.13 (m, 4H), 7.18 - 7.09 (m, 2H), 7.07 - 6.98 (m, 2H), 3.48 (s, 3H), 3.45 (s, 3H), 1.97 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 172.2, 136.4, 135.2, 132.0, 129.3, 127.3, 127.3, 123.8, 121.4, 120.0, 118.1, 116.9, 109.0, 37.4, 29.5, 9.5; IR (neat) ν 2918, 1642, 1473, 1420, 1382, 1360, 1226, 1107, 1003, 853, 788, 749, 724, 697, 658 cm^{-1}; HRMS (ESI) Calcd. for $C_{18}H_{18}N_2ONa^+$ Requires: 301.1311, Found: 301.1311.





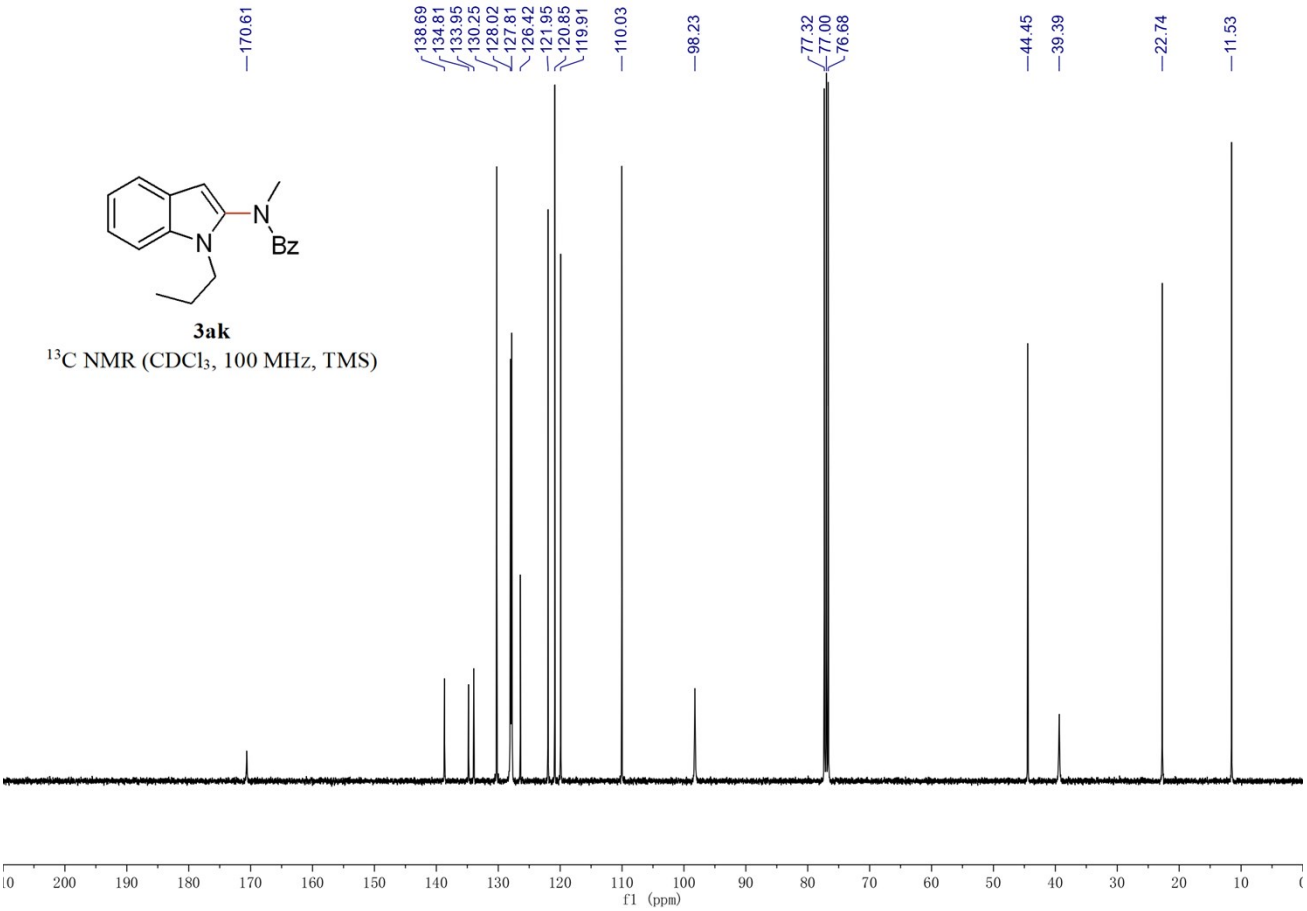


***N*-methyl-*N*-(1-propyl-1H-indol-2-yl)benzamide (3ak)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 37 mg, 64% yield; M.p.: 73 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.51 (d, *J* = 7.8 Hz, 1H), 7.39 (d, *J* = 7.6 Hz, 2H), 7.23 - 7.16 (m, 3H), 7.16 - 7.03 (m, 3H), 6.27 (s, 1H), 3.85 - 3.74 (m, 2H), 3.48 (s, 3H), 1.84 - 1.66 (m, 2H), 0.93 (t, *J* = 7.4 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.6, 138.7, 134.8, 133.9, 130.2, 128.0, 127.8, 126.4, 122.0, 120.9, 119.9, 110.0, 98.2, 44.5, 39.4, 22.7, 11.5; IR (neat) ν 2964, 1652, 1557, 1461, 1402, 1346, 1316, 1209, 1093, 1013, 919, 776, 748, 728, 698, 669 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{20}N_2ONa^+$ Requires: 315.1468, Found: 315.1469.



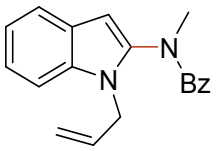
3ak

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

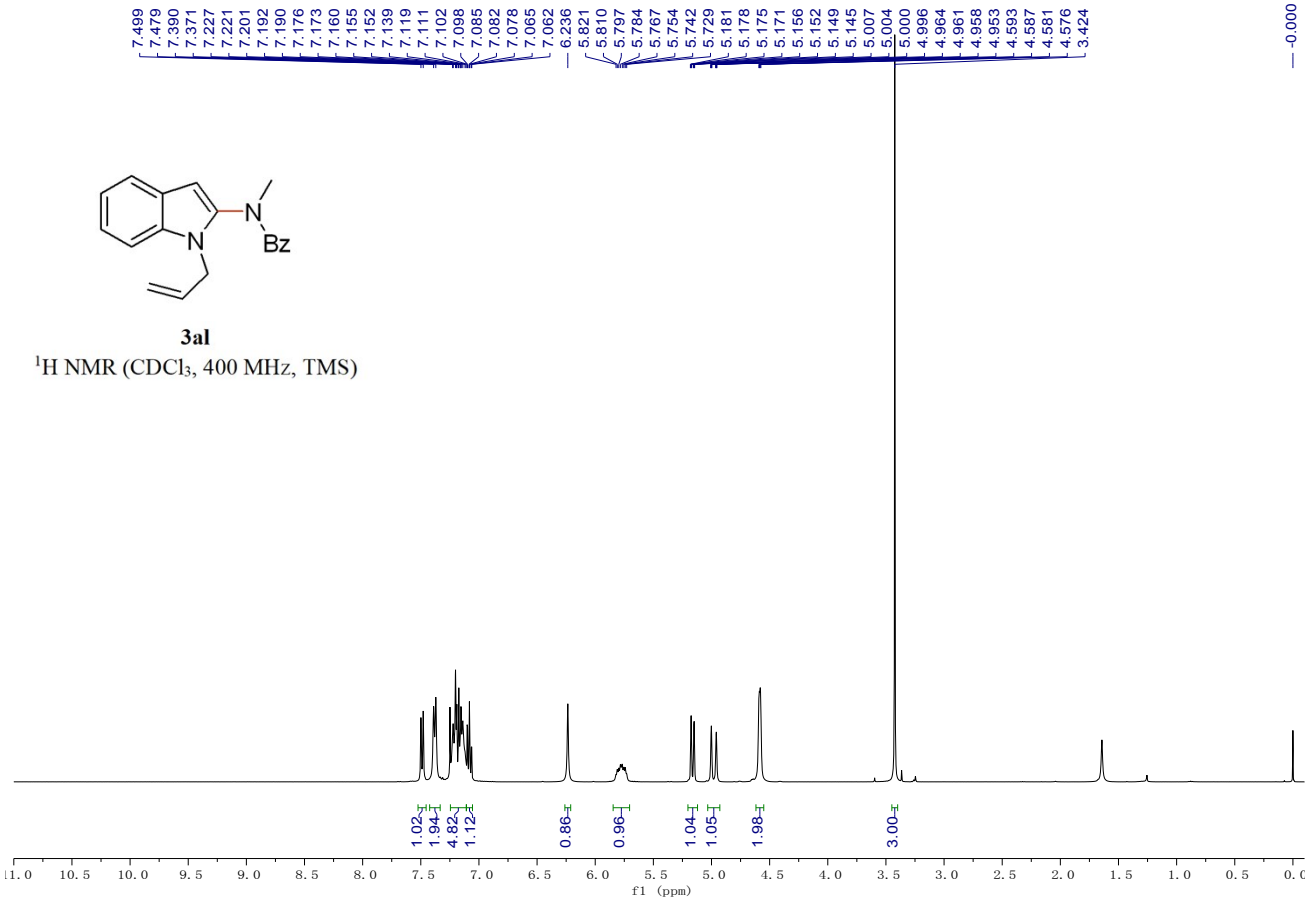


3ak

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

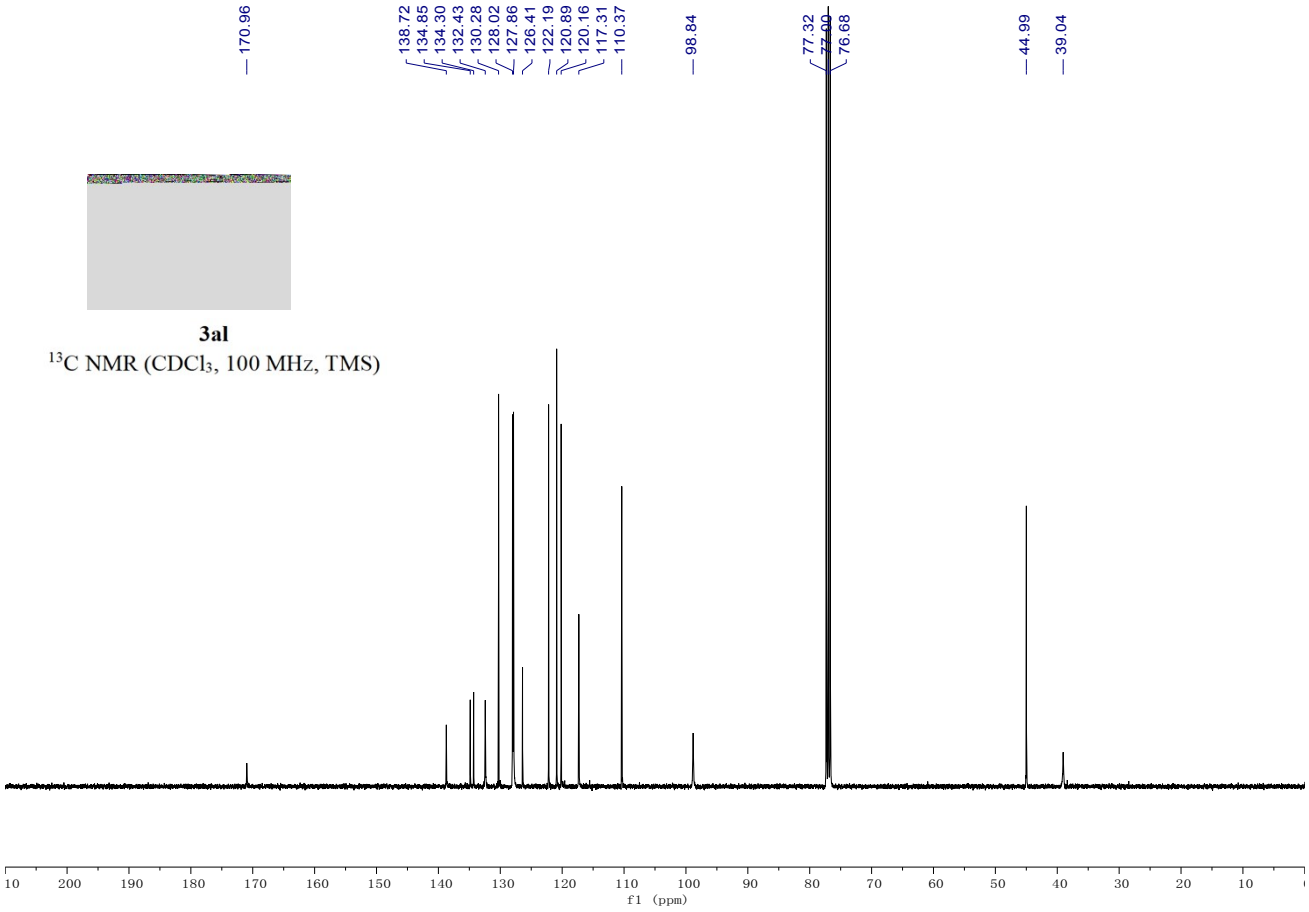


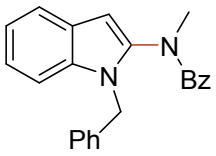
***N*-(1-allyl-1H-indol-2-yl)-*N*-methylbenzamide (3al)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 18 mg, 31% yield; M.p.: 92 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.49 (d, *J* = 7.9 Hz, 1H), 7.38 (d, *J* = 7.6 Hz, 2H), 7.25 - 7.11 (m, 5H), 7.11 - 7.06 (m, 1H), 6.24 (s, 1H), 5.85 - 5.70 (m, 1H), 5.20 - 5.12 (m, 1H), 5.03 - 4.93 (m, 1H), 4.62 - 4.55 (m, 2H), 3.42 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.0, 138.7, 134.9, 134.3, 132.4, 130.3, 128.0, 127.9, 126.4, 122.2, 120.9, 120.2, 117.3, 110.4, 98.8, 45.0, 39.0; IR (neat) ν 1651, 1578, 1560, 1461, 1400, 1346, 1315, 1199, 1014, 988, 924, 782, 748, 717, 696, 671 cm^{-1}; HRMS (ESI) Calcd. for $C_{19}H_{18}N_2ONa^+$ Requires: 313.1311, Found: 313.1314.



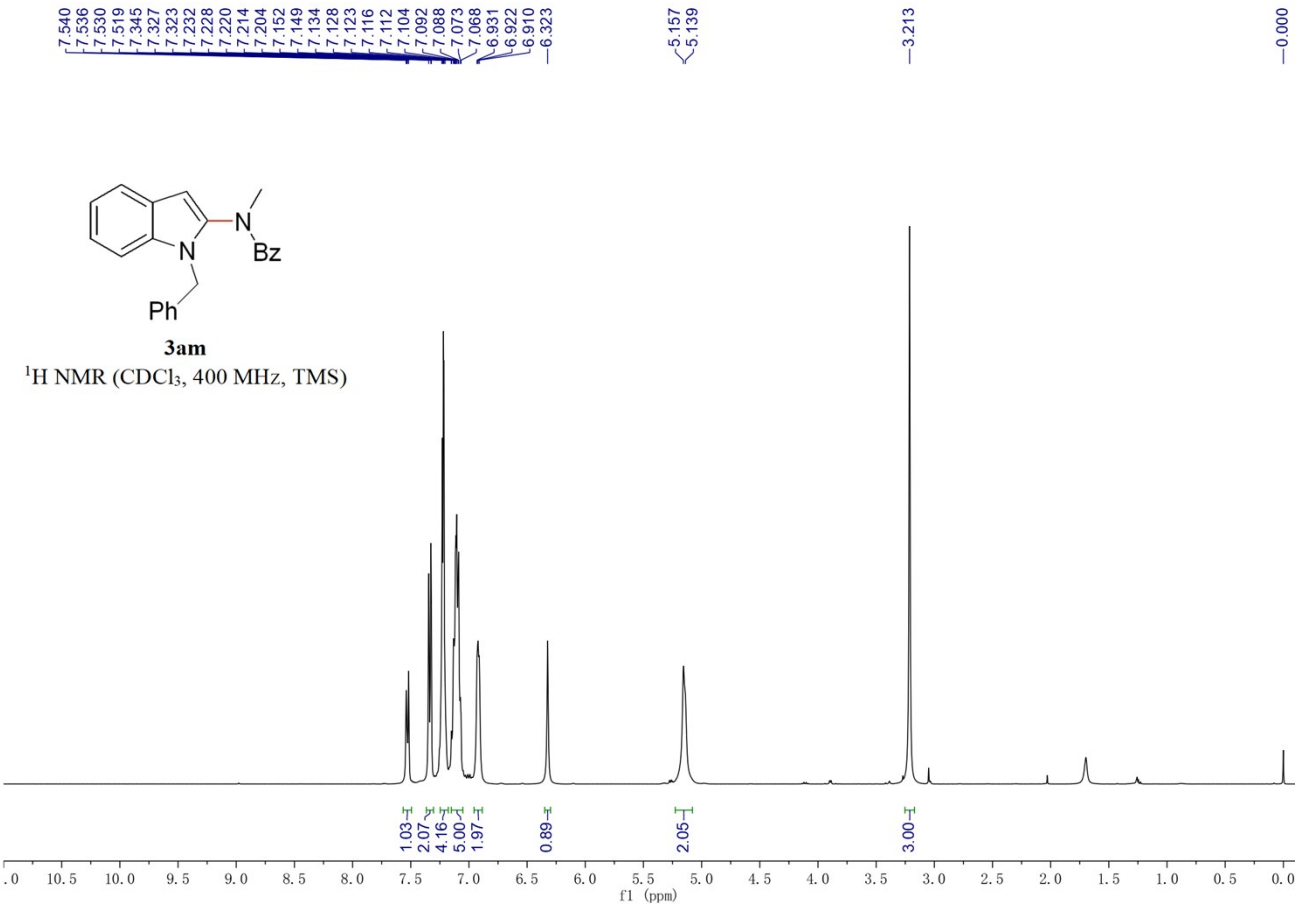
3al

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)



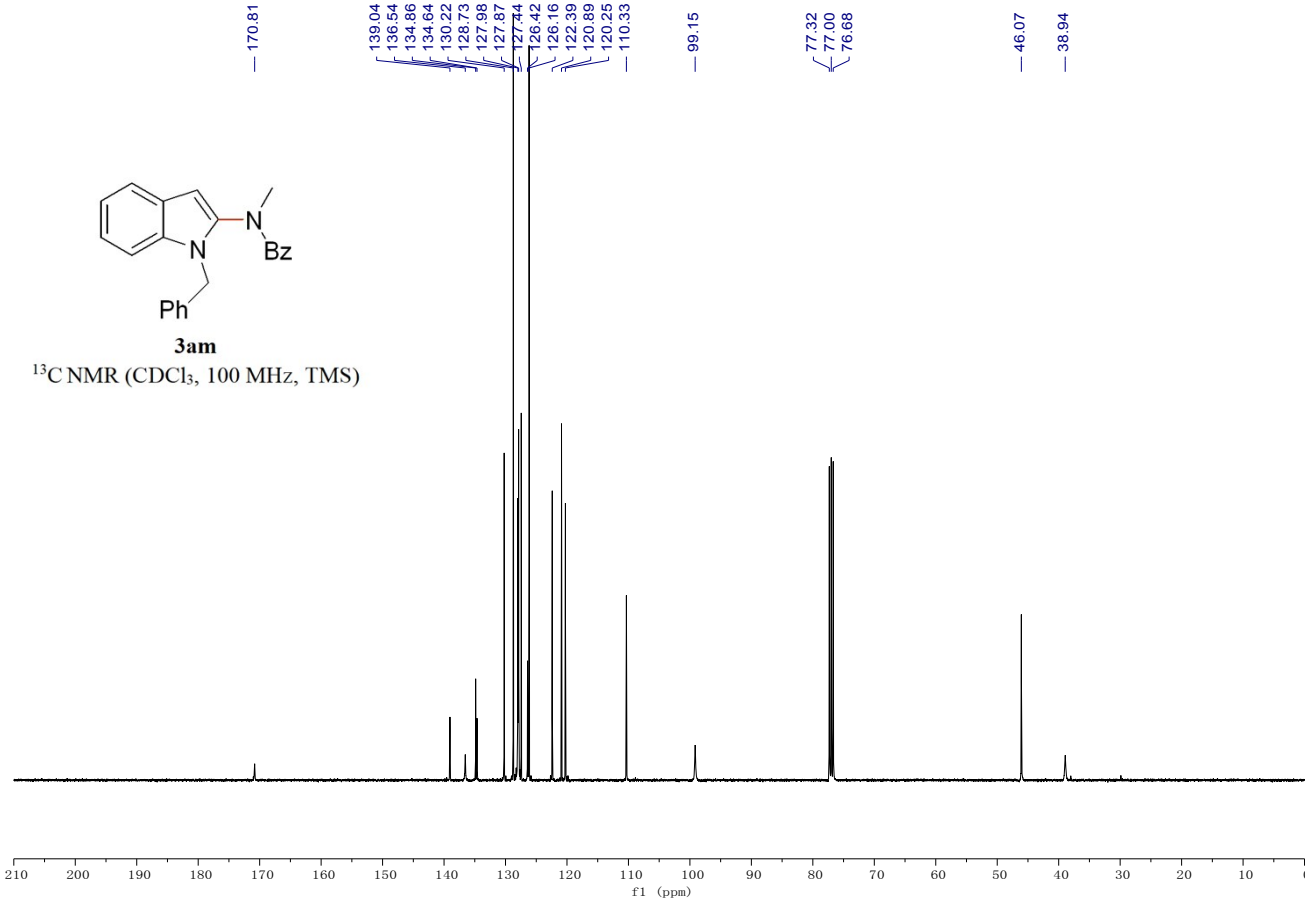


***N*-(1-benzyl-1H-indol-2-yl)-*N*-methylbenzamide (3am)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 40 mg, 59% yield; M.p.: 136 ^{o}C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.57 - 7.49 (m, 1H), 7.37 - 7.30 (m, 2H), 7.25 - 7.18 (m, 4H), 7.15 - 7.05 (m, 5H), 6.96 - 6.89 (m, 2H), 6.32 (s, 1H), 5.23 - 5.08 (m, 2H), 3.21 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 170.8, 139.0, 136.5, 134.9, 134.6, 130.2, 128.7, 128.0, 127.9, 127.4, 126.4, 126.2, 122.4, 120.9, 120.3, 110.3, 99.1, 46.1, 38.9; IR (neat) ν 1651, 1578, 1558, 1450, 1354, 1314, 1249, 1162, 1078, 1013, 795, 778, 732, 698, 674 cm^{-1}; HRMS (ESI) Calcd. for $C_{23}H_{20}N_2ONa^+$ Requires: 363.1468, Found: 363.1463.



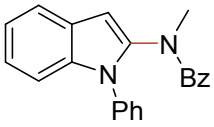
3am

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

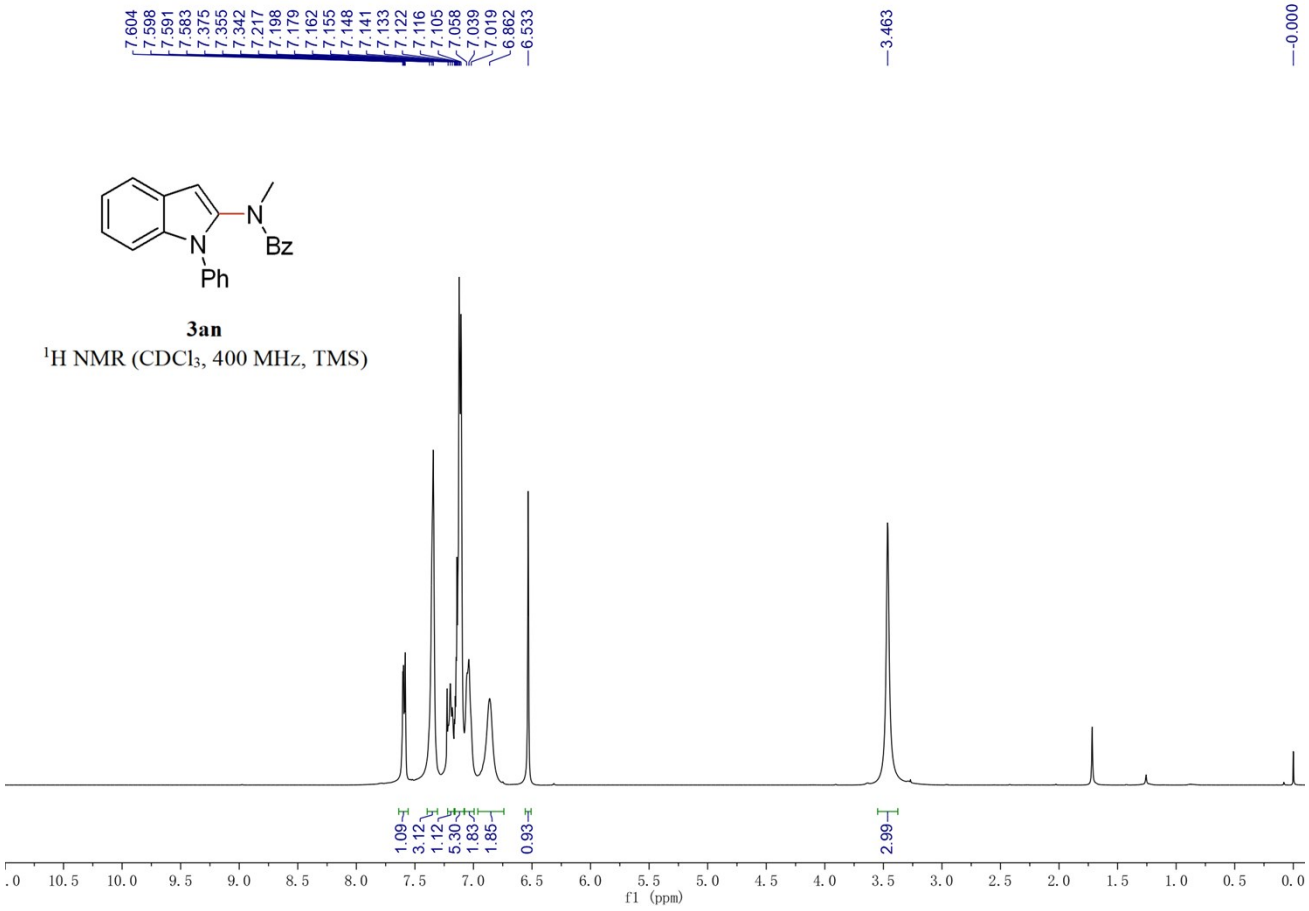


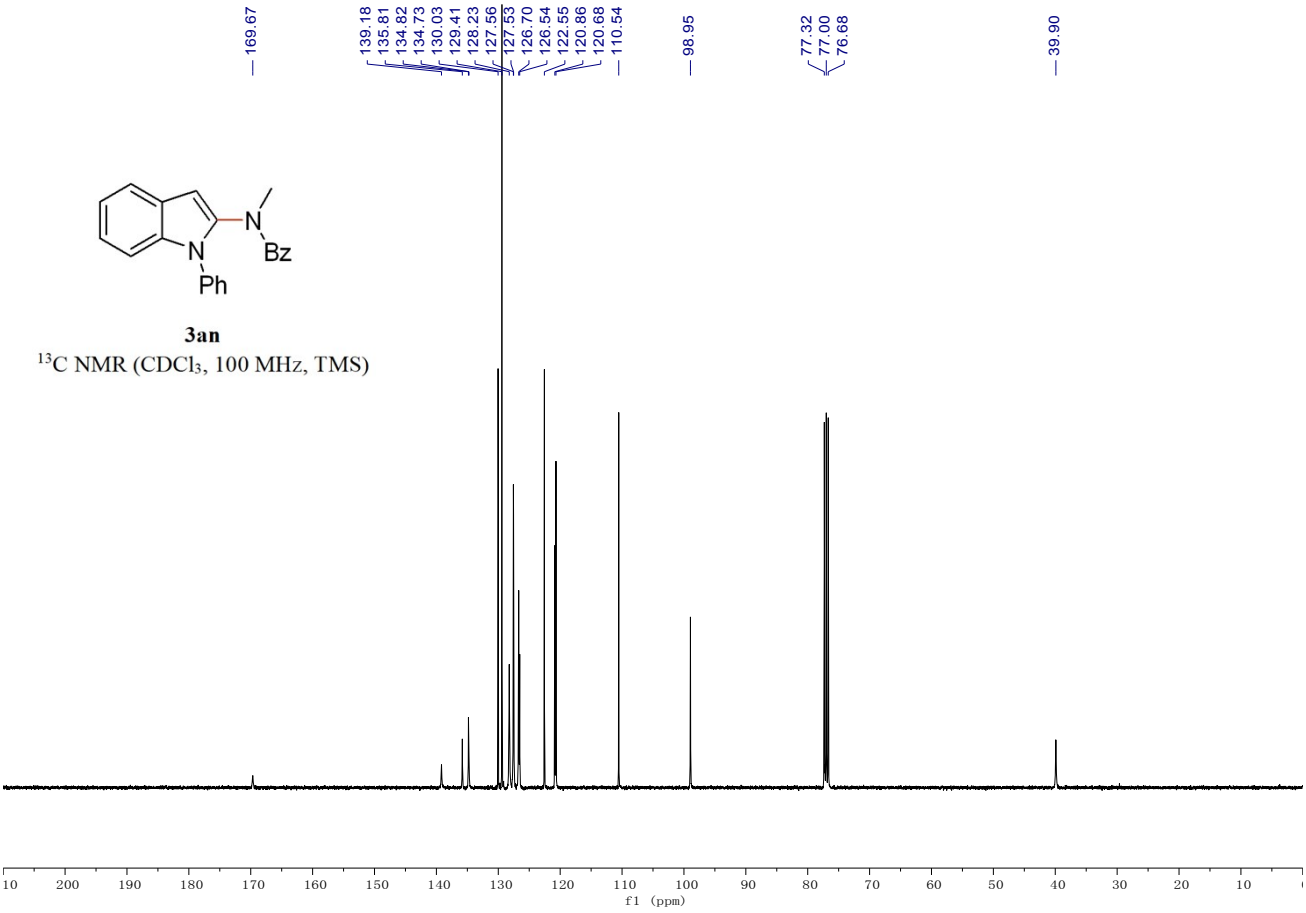
3am

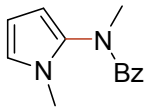
^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



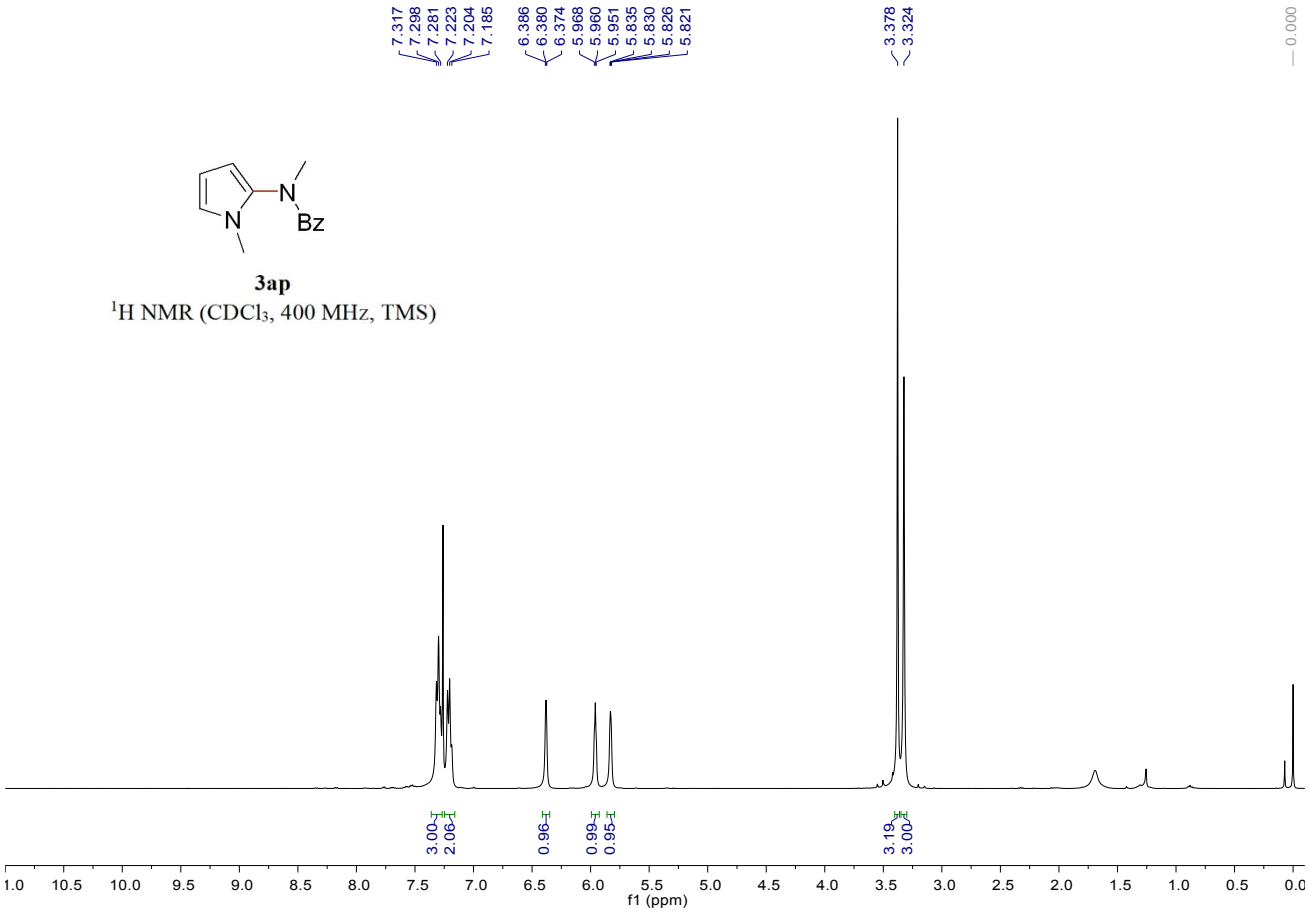
***N*-methyl-*N*-(1-phenyl-1H-indol-2-yl)benzamide (3an)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 41 mg, 62% yield; M.p.: 117 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.64 - 7.56 (m, 1H), 7.35 (d, *J* = 5.0 Hz, 3H), 7.20 (t, *J* = 7.5 Hz, 2H), 7.18 - 7.08 (m, 6H), 7.04 (t, *J* = 7.7 Hz, 2H), 6.96 - 6.72 (m, 2H), 6.53 (s, 1H), 3.46 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 169.7, 139.2, 135.8, 134.8, 134.7, 130.0, 129.4, 128.2, 127.6, 127.5, 126.7, 126.5, 122.5, 120.9, 120.7, 110.5, 98.9, 39.9; IR (neat) ν 3066, 1651, 1560, 1498, 1489, 1450, 1356, 1313, 1198, 1074, 1046, 1014, 793, 782, 750, 726, 694, 663 cm^{-1}; HRMS (ESI) Calcd. for $C_{22}H_{18}N_2ONa^+$ Requires: 349.1311, Found: 349.1309.





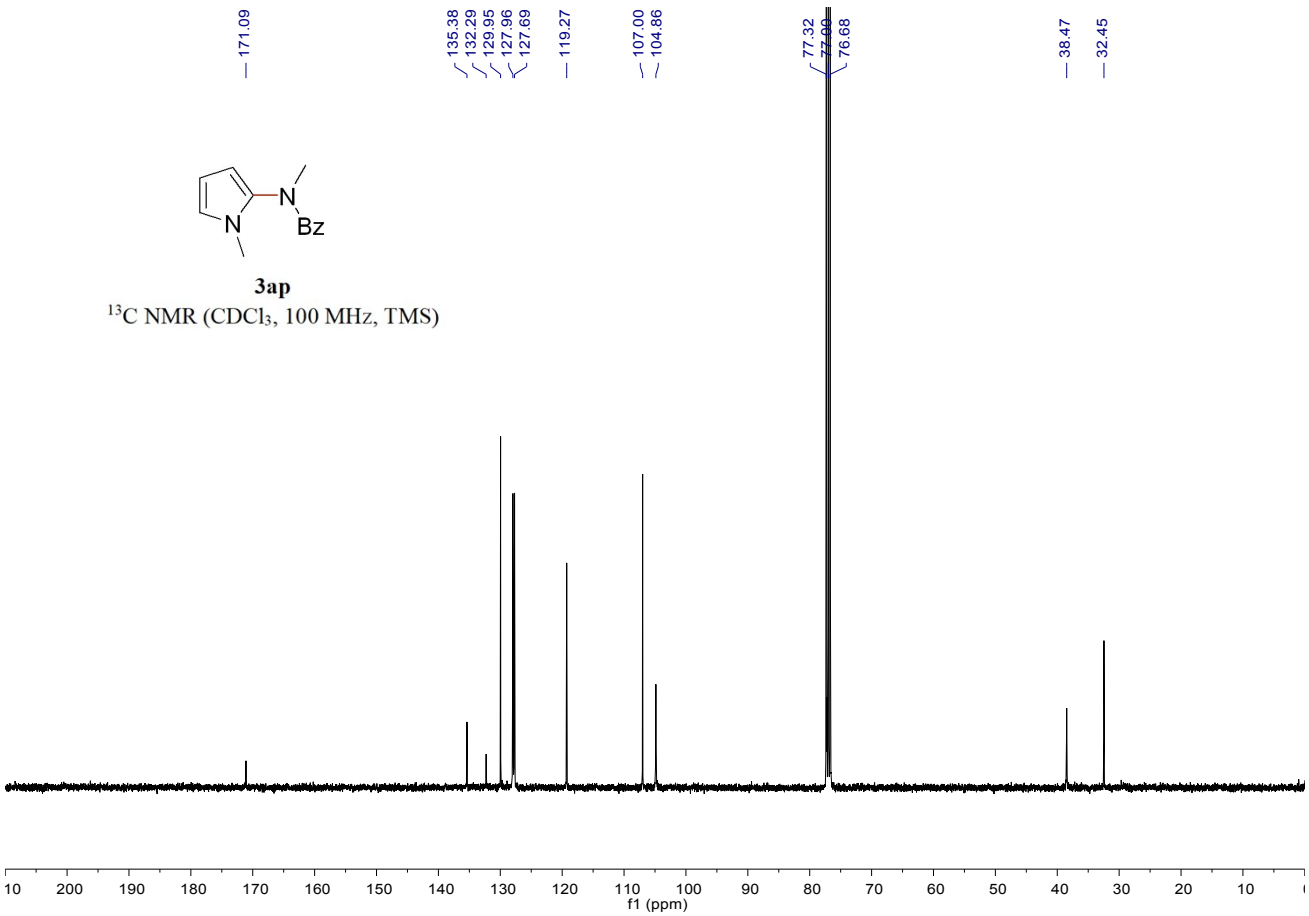


***N*-methyl-N-(1-methyl-1H-pyrrol-2-yl)benzamide (3ap)**. The title product was obtained after purification by prep-TLC (DCM). An off-white solid, 22 mg, 50% yield; M.p.: 89 °C; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.36 - 7.27 (m, 3H), 7.25 - 7.16 (m, 2H), 6.41 - 6.35 (m, 1H), 5.96 (t, J = 3.4 Hz, 1H), 5.86 - 5.80 (m, 1H), 3.38 (s, 3H), 3.32 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 171.1, 135.4, 132.3, 129.9, 128.0, 127.7, 119.3, 107.0, 104.9, 38.5, 32.5; IR (neat) ν 2923, 1637, 1555, 1487, 1419, 1355, 1314, 1257, 1105, 1085, 1024, 986, 799, 740, 716, 669 cm^{-1}; HRMS (ESI) Calcd. for $C_{13}H_{14}N_2ONa^+$ Requires: 237.0998, Found: 237.1007.



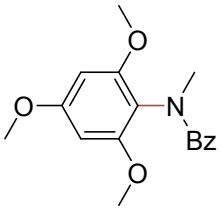
3ap

^{1}H NMR ($CDCl_3$, 400 MHz, TMS)

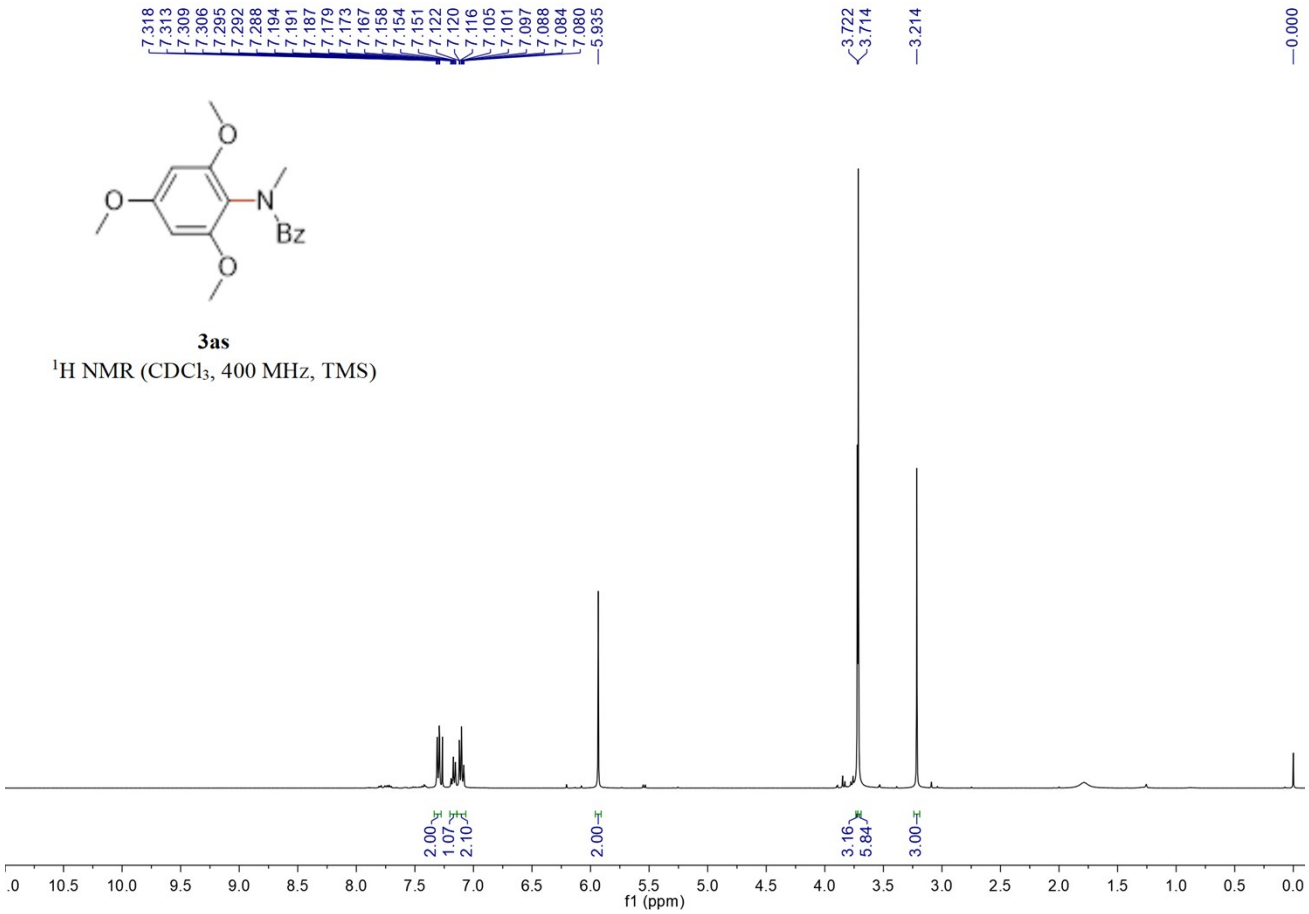


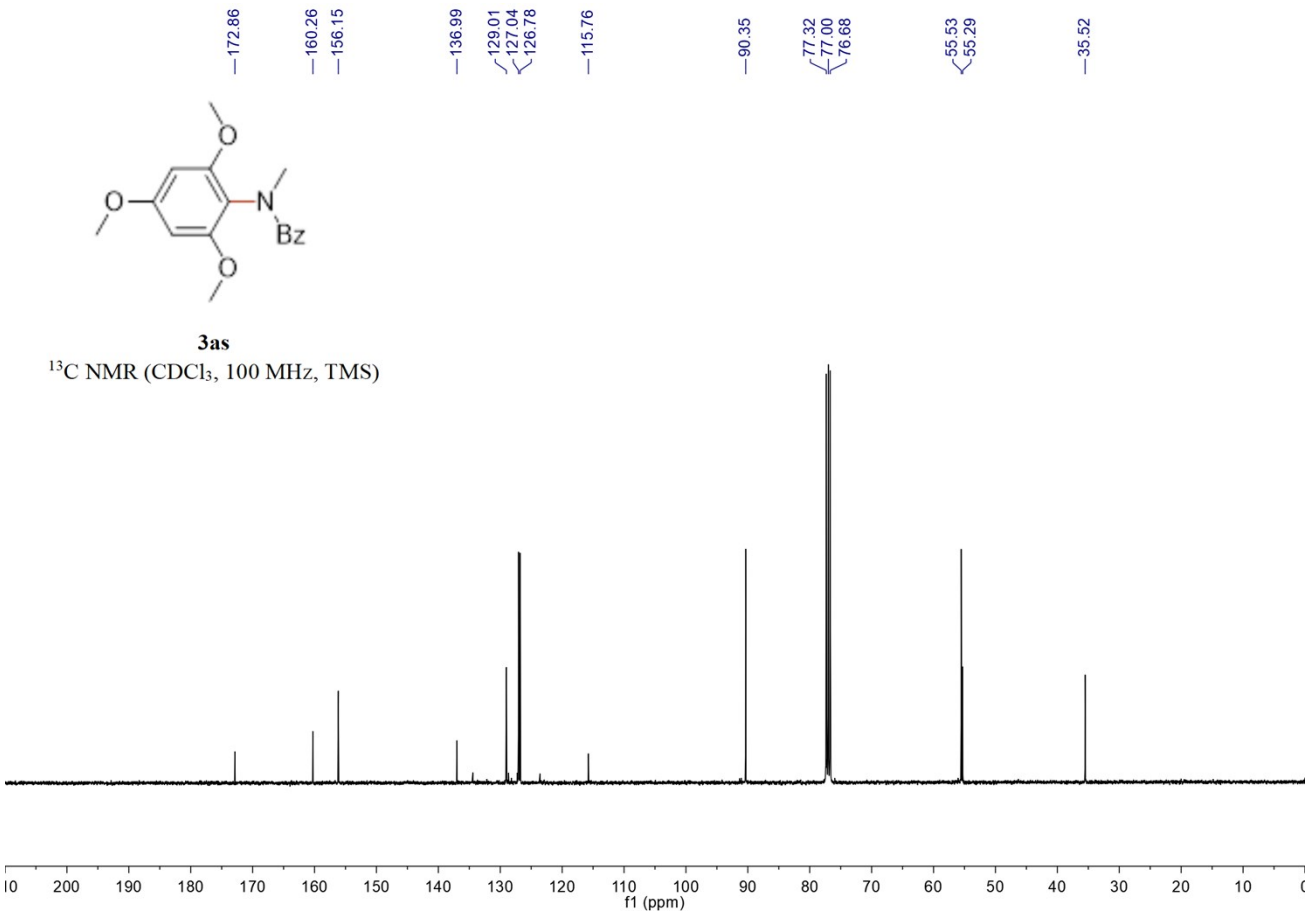
3ap

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)



N-methyl-N-(2,4,6-trimethoxyphenyl)benzamide (3as). The title product was obtained after purification by prep-TLC (PE/EA = 2/1). An pale brown oil, 16 mg, 26% yield; ^{1}H NMR ($CDCl_3$, 400 MHz, TMS) δ 7.33 - 7.28 (m, 2H), 7.20 - 7.14 (m, 1H), 7.14 - 7.07 (m, 2H), 5.93 (s, 2H), 3.72 (s, 3H), 3.71 (s, 6H), 3.21 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz, TMS) δ 172.9, 160.3, 156.2, 137.0, 129.0, 127.0, 126.8, 115.8, 90.4, 55.5, 55.3, 35.5; IR (neat) ν 2934, 1730, 1636, 1590, 1508, 1455, 1417, 1341, 1229, 1204, 1157, 1126, 1056, 813, 728, 702 cm^{-1}; HRMS (ESI) Calcd. for $C_{17}H_{20}NO_4^+$ Requires: 302.1387, Found: 302.1390.





3as

^{13}C NMR ($CDCl_3$, 100 MHz, TMS)

(K) References

(1) Greulich, T. W.; Daniliuc, C. G.; Studer, A. *Org. Lett.* **2015**, *17*, 254.

(2) Titherley, A. W.; Worrall, E. *J . Chem. Soc.* **1910**, *97*, 839.

(3) Huvaere, K.; Skibsted, L. H. *J. Am. Chem. Soc.* **2009**, *131*, 8049.

(4) Davies, J.; Svejstrup, T. D.; Fernandez Reina, D.; Sheikh, N. S.; Leonori, D. *J. Am. Chem. Soc.* **2016**, *138*, 8092.