Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

For

Divergent Synthesis of α-Aryl Ketones/Esters via Rhodium-Catalyzed Selective Deesterification and Decarbonylation of the Diazo Compounds

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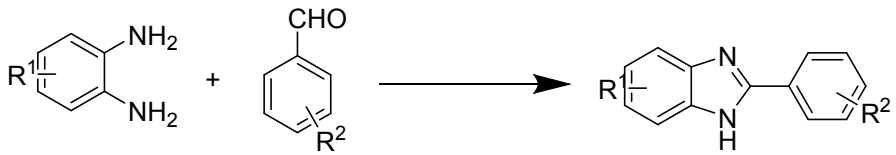
1、General information

All chemicals were purchased from Adamas Reagent, energy chemical company, J&K Scientific Ltd, Bide Pharmatech Ltd and Tansoole. The reagents and solvents were purchased from commercial suppliers and used without further purification. Reactions were monitored by TLC or GC-MS analysis. Flash column chromatography was performed over silica gel (200-300 mesh).

^{1}H-NMR and ^{13}C-NMR spectra were recorded in $CDCl_3$ on a Bruker Avance III 500MHz NMR spectrometer (500 MHz ^{1}H, 125 MHz ^{13}C) at room temperature. Chemical shifts were reported in ppm on the scale relative to $CDCl_3$ (δ = 7.26 for ^{1}H-NMR , δ = 77.00 for ^{13}C-NMR) as an internal reference. High resolution mass spectra were recorded using a Thermo Fisher Scientific LTQ FT Ultra or Waters Micromass GCT Premier instrument. Coupling constants (*J*) were reported in Hertz (Hz).

2. General procedure for synthesis of benzimidazoles

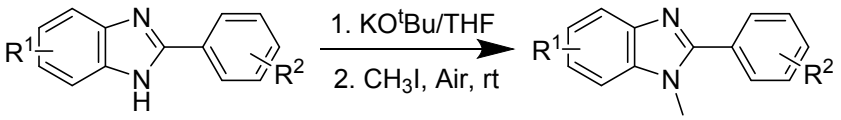
2.1. General reaction procedure for the preparation of 3



1 2 3

An aqueous solution of $Na_2S_2O_5$ (1.5 mmol), followed by the corresponding o-phenylenediamine (1.0 mmol) was added to a solution of substituted benzaldehyde (1.0 mmol) in ethanol and the mixture was stirred at 70 ^oC. After completion of the reaction, the reaction mixture was cooled, water was added, and the mixture was left to stand for 1 h at 0 ^oC to obtain a precipitate. Then, the obtained crude solid was purified by column chromatography to afford benzimidazole compounds with the required purity.

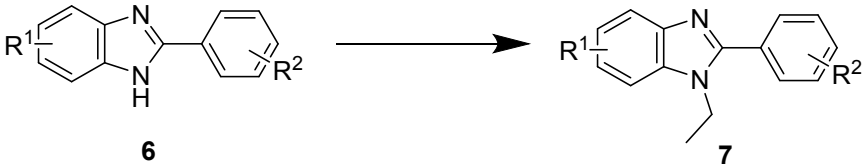
2.2. General process for the synthesis of 5



4 5

Excess methyl iodine (0.25 mL) was added to a stirred THF solution of 2-phenylbenzimidazole (0.7 g, 3.6 mmol) and KO^{t}Bu (0.42 g, 3.7 mmol) at RT under air. The resulting solution was then further stirred for a few hours. After the removal of the solvent, compound **5** were obtained quantitatively.

2.3. General process for the synthesis of 7

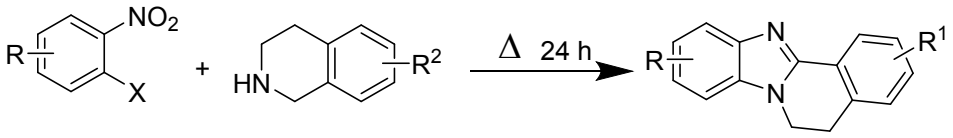


6 7

The precursor 2- phenyl-1H-Benzo[d]imidazole (PhBI) was synthesized using a previously reported procedure. Under a nitrogen atmosphere, PhBI (1.12 g, 5.8 mmol) was dissolved in acetone. An equivalent amount of KOH was added and the mixture was stirred for 30 min, followed by addition of bromoethane. After cooling to room temperature, the reactant mixture was heated to relux overnight and quenched with

ice water. The mixture was then extracted by dichloromethane (3x30 mL). The organic layer was dried with Na_2SO_4, and the solvent was removed under vacuum. The product was then obtained by column chromatography on silica gel.

2.4. General process for the synthesis of 10



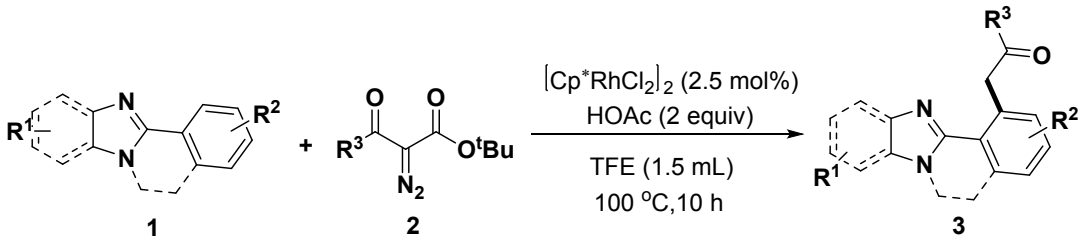
8
8a, x= F
8b, x= Cl
8c, x= Br
8d, x= I

9 (2.5 equiv)

10

A mixture of **8** (1 mmol) and **9** (2.5 mmol) was stirred at 130 °C or 150 °C, for 24 h under an argon atmosphere. The cooled reaction mixture was diluted with methanol and treated with a saturated ammonia solution in methanol (0.5 mL). The resulting solution was concentrated in vacuo. The residue was purified by silica gel column chromatography (heptane: EtOAc 4:1 to EtOAc) to afford the fused benzimidazole **10**.

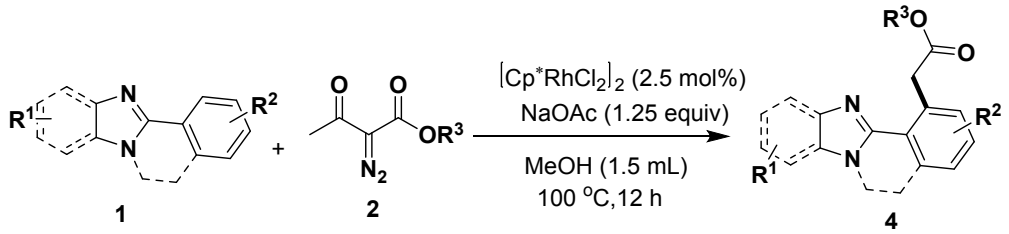
3. Experimental Procedures



1 **2** **3**

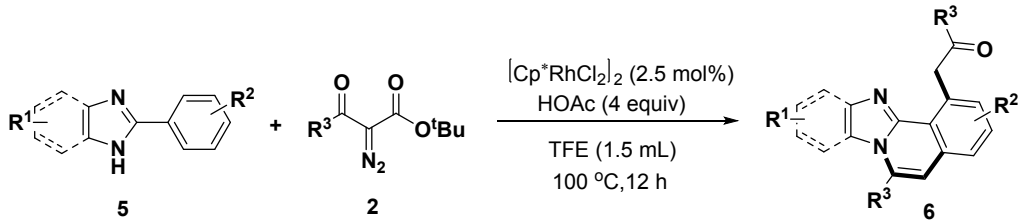
Scheme 2

A sealed tube was charged with 2-Phenylbenzimidazole **1** (0.2 mmol, 1.0 equiv), $[Cp^*RhCl_2]_2$ (3.1 mg, 2.5 mol %), HOAc (24 mg, 2.0 equiv), diazo compounds **2** (0.25 mmol, 1.25 equiv) in TFE (1.5 mL). Under an argon atmosphere, the reaction mixture was stirred at 100 °C for 10 h. Then the mixture was cooled to room temperature and saturated ammonium chloride solution was added to the reaction mixture and extracted with EtOAc. The combined organic phase was washed with saturated brine and dried over Na_2SO_4. The solvent was evaporated, and the residue was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate) to afford the corresponding products **3** (Scheme 2).



Scheme 3

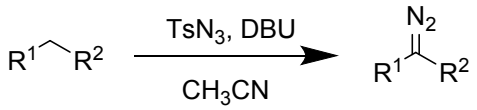
A sealed tube was charged with 2-Phenylbenzimidazole **1** (0.2 mmol, 1.0 equiv), $[Cp^*RhCl_2]_2$ (3.1 mg, 2.5 mol %), NaOAc (20.5 mg, 1.25 equiv), diazo compounds **2** (0.25 mmol, 1.25 equiv) in MeOH (1.5 mL). Under an argon atmosphere, the reaction mixture was stirred at 100 °C for 12 h. Then the mixture was cooled to room temperature and saturated ammonium chloride solution was added to the reaction mixture and extracted with EtOAc. The combined organic phase was washed with saturated brine and dried over Na_2SO_4. The solvent was evaporated, and the residue was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate) to afford the corresponding products **4** (Scheme 3).



Scheme 4

A sealed tube was charged with 2-Phenylbenzimidazole **5** (0.2 mmol, 1.0 equiv), $[Cp^*RhCl_2]_2$ (3.1 mg, 2.5 mol %), HOAc (48 mg, 4.0 equiv), diazo compounds **2** (0.25 mmol, 1.25 equiv) in TFE (1.5 mL). Under an argon atmosphere, the reaction mixture was stirred at 100 °C for 12 h. Then the mixture was cooled to room temperature and saturated ammonium chloride solution was added to the reaction mixture and extracted with EtOAc. The combined organic phase was washed with saturated brine and dried over Na_2SO_4. The solvent was evaporated, and the residue was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate) to afford the corresponding products **6** (Scheme 4).

4. General process for preparation of diazo substrates



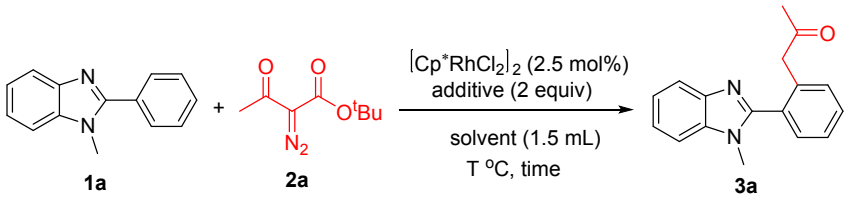
R^1 = carbonyl
R^2 = carbonyl or ester

To a solution of ketonic ester or 1,3 di-ketone (5.0 mmol, 1.0 equiv.) in CH3CN (10 mL), TsN_3 (6.0 mmol, 1.1 equiv.) was added. Then the reaction mixture was cooled to 0 oC and a solution of DBU (6.0 mmol, 1.1 equiv.) in 10 mL CH_3CN was added dropwise. Next, the reaction temperature was raised to room temperature. After

stirring for 3 hours, the residue was extracted with EA for three times. The combined organic layers were washed with water and brine sequentially, dried over anhydrous Na_2SO_4, filtered and concentrated. The crude product was purified by flash chromatography on silica gel to afford the corresponding product in 70-90% yields.

5. Optimization of experiment conditions

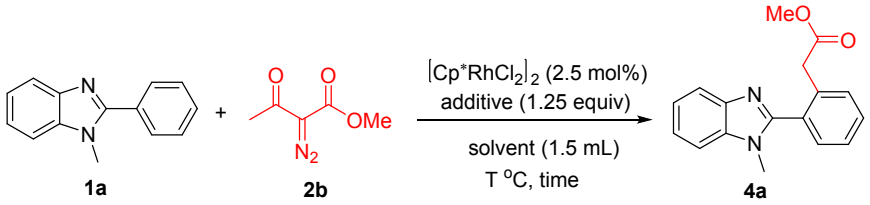
Table S1. Optimization of the reaction for Rh-Catalyzed deesterification of diazo Compounds[a]



entry	additive	solvent	T (°C)	time (h)	yield (%)[b]
1	KOAc	TFE	100	12	58
2	HOAc	TFE	100	12	93 (86)[c]
3	HCOOH	TFE	100	12	trace
4	TFA	TFE	100	12	trace
5	$C(CH_3)_3COOH$	TFE	100	12	58
6	HOAc	DCM	100	12	N.D
7	HOAc	THF	100	12	N.D
8	HOAc	Toluene	100	12	N.D
9	HOAc	CH_3CN	100	12	N.D
10	HOAc	1,4-dioxane	100	12	N.D
11	HOAc	TFE	120	12	14
12	HOAc	TFE	80	12	86 (78)[c]
13	HOAc	TFE	60	12	79
14	HOAc	TFE	100	14	93 (84)[c]
15	HOAc	TFE	100	10	95 (89)[c]
16	HOAc	TFE	100	8	77

[a] The reactions carried out with 1a (0.20 mmol) and 2a (0.25 mmol) in the presence of 2.5 mol % Rh(III) catalyst in 1.5 mL of solvent for the indicated times under a N_2 atmosphere. [b] GC yields. [c] isolated yields.

Table S2. Optimization of the reaction for Rh-catalyzed decarbonylation of diazo compounds[a]

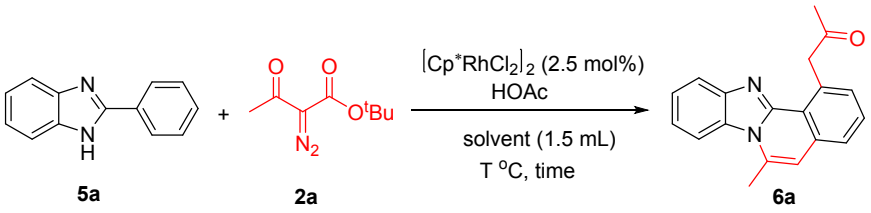


1a **2b** **4a**

entry	additive	solvent	T (°C)	time (h)	yield (%)[b]
1	KOAc	MeOH	100	12	80 (76)[c]
2	AgOAc	MeOH	100	12	90 (83)[c]
3	K_2CO_3	MeOH	100	12	trace
4	Cs_2CO_3	MeOH	100	12	n.r
5	KO^tBu	MeOH	100	12	7
6	**NaOAc**	**MeOH**	**100**	**12**	**88 (80)[c]**
7	HOAc	MeOH	100	12	55
8	$Cu(OAc)_2$	MeOH	100	12	31
9	NaOAc	H_2O	100	12	trace
10	NaOAc	THF	100	12	n.r
11	NaOAc	CH_3CN	100	12	n.r
12	NaOAc	Toluene	100	12	n.r
13	NaOAc	1,4-dioxane	100	12	trace
14	NaOAc	MeOH	80	12	52
15	NaOAc	MeOH	60	12	24
16	NaOAc	MeOH	100	10	60

[a]The reactions carried out with 1a (0.20 mmol) and 2b (0.25 mmol) in the presence of 2.5mol % Rh(III) catalyst in 1.5 mL of solvent for the indicated times Under a N_2 atmosphere. [b] GC yields. [c] isolated yield.

Table S3. Optimization of the reaction for Rh-catalyzed dual C-H activation[a]



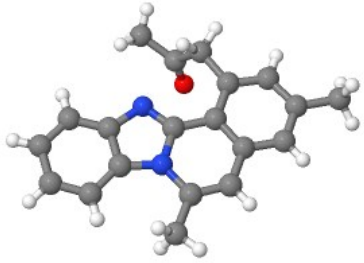
5a **2a** **6a**

entry	**2a** (equiv)	HOAc (equiv)	solvent	T (^{o}C)	time (h)	yield (%)[b]
1	2.5	4	TFE	100	16	90 (86)[c]
2	2.3	4	TFE	100	16	73
3	2.5	3	TFE	100	16	83
4	2.5	4	TFE	80	16	88
5	2.5	4	TFE	60	16	12
6	2.5	4	TFE	100	12	98 (95)[c]
7	2.5	4	TFE	100	8	75
8	2.5	4	CH_3CN	100	12	4
9	2.5	4	1,4-dioxane	100	12	N.D

[a]The reactions carried out with 5a (0.20 mmol) and 2a (0.25 mmol) in the presence of 2.5 mol% Rh(III) catalyst in 1.5 mL of solvent for the indicated times Under a N_2 atmosphere. [b] GC yields.[c] isolated yield in parentheses.

6. Crystal data of 6e

Crystallographic data for compound **6e** (CCDC-1846859) has been deposited with the Cambridge Crystallographic Data Centre, Copies of the data can be obtained, free of charge, on application to CCDC (Email:deposit@ccdc.cam.ac.uk).



Displacement ellipsoids are drawn at 30% probability level

Bond precision: C-C = 0.0027 A Wavelength=0.71073

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alpha=90 beta=99.114(10) gamma=90

Temperature: 0 K

	Calculated	Reported
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Hall group	-P 2ybc	-P 2ybc
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Sum formula	C20 H18 N2 O	C20 H18 N2 O
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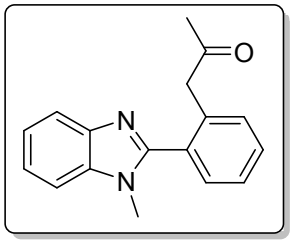
Dx,g cm-3	1.290	1.290
Z	4	4
Mu (mm-1)	0.080	0.080
F000	640.0	640.3
F000'	640.24	
h,k,lmax	12,16,12	12,16,12
Nref	2740	2733
Tmin,Tmax		0.226,1.000
Tmin'		

Correction method= # Reported T Limits: Tmin=0.226 Tmax=1.000
AbsCorr = MULTI-SCAN
Data completeness= 0.997 Theta(max)= 24.990
R(reflections)= 0.0517(1993) wR2(reflections)= 0.1519(2733)
S = 1.027 Npar= 210

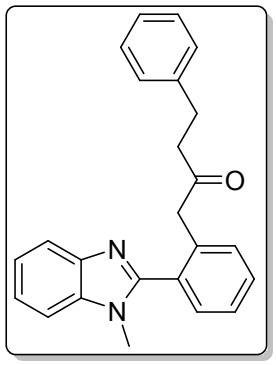
7. Characterization data for products

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3a)



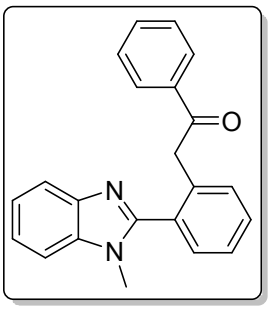
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a brown solid (40.1 mg, 76%). mp. 100 °C (lit. 100-102 °C). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (d, *J* = 7.0 Hz, 1H), 7.48 – 7.44 (m, 1H), 7.42 – 7.37 (m, 3H), 7.33 – 7.28 (m, 3H), 3.99 (s, 2H), 3.67 (s, 3H), 1.95 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 205.8, 152.9,142.3, 135.4, 135.2, 131.2, 130.1, 129.8, 126.8, 122.6, 122.2, 119.4, 109.7, 48.0, 30.8, 29.7. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 265.1335; found: 265.1337.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl) phenyl)-4-phenylbutan-2-one (3b)



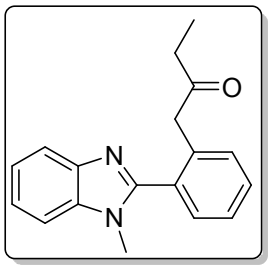
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a brown liquid (46.0 mg, 65%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (dd, *J* = 6.6, 1.8 Hz, 1H), 7.48 – 7.45 (m, 1H), 7.44 – 7.39 (m, 3H), 7.34 (ddd, *J* = 7.1, 6.1, 3.6 Hz, 2H), 7.27 (d, *J* = 7.6 Hz, 1H), 7.18 – 7.11 (m, 3H), 6.94 – 6.90 (m, 2H), 3.97 (s, 2H), 3.64 (s, 3H), 2.55 (s, 4H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.2, 153.0, 142.5, 140.7, 135.5, 135.4, 131.4, 130.1, 129.9, 128.3, 128.1, 126.9, 125.9, 122.8, 122.4, 119.6, 109.7, 47.5, 44.1, 30.9, 29.5. HRMS (EI, m/z) calcd for $C_{24}H_{22}N_2O$ $[M+H]^+$: 355.1805; found: 355.1807.

2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)-1-phenylethan-1-one (3c)



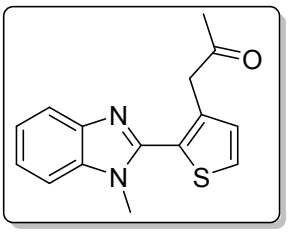
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow liquid (53.5mg, 82%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.70 – 7.65 (m, 3H), 7.39 (ddd, *J* = 10.3, 8.6, 5.1 Hz, 5H), 7.27 (ddd, *J* = 12.3, 6.3, 4.2 Hz, 5H), 4.46 (s, 2H), 3.60 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 197.7, 152.9, 142.5, 136.4, 135.7, 135.4, 132.9, 131.5, 130.1,130.0, 128.3, 128.1, 126.8, 122.6, 122.2, 119.6, 109.6, 42.8, 30.8. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 327.1492; found: 327.1493.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl) phenyl) butan-2-one (3d)



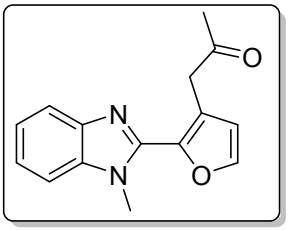
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a brown liquid (40.6 mg, 73%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.78 (dd, *J* = 6.8, 1.7 Hz, 1H), 7.47 (ddd, *J* = 7.6, 6.5, 2.5 Hz, 1H), 7.43 – 7.37 (m, 3H), 7.35 – 7.28 (m, 3H), 3.96 (s, 2H), 3.68 (s, 3H), 2.26 (d, *J* = 7.3 Hz, 2H), 0.77 (t, *J* = 7.3 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 208.6, 153.0, 142.5, 135.6, 135.5，131.3, 130.2, 130.1, 129.9, 126.8, 122.7, 122.3, 119.5, 109.6, 46.9, 35.8, 30.9, 7.6. HRMS (EI, m/z) calcd for $C_{18}H_{18}N_2O$ $[M+H]^+$: 279.1492; found: 279.1495.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)thiophen-3-yl)propan-2-one (3e)



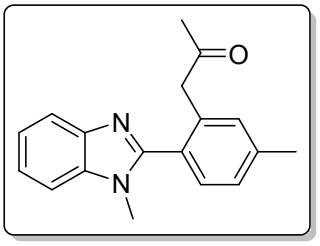
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (42.1 mg, 78%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.82 – 7.77 (m, 1H), 7.51 (d, *J* = 5.1 Hz, 1H), 7.40 – 7.29 (m, 3H), 7.07 (d, *J* = 5.1 Hz, 1H), 4.01 (s, 2H), 3.84 (s, 3H), 2.16 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.5, 147.2, 142.86 (s), 137.0, 135.9, 130.1, 127.3, 126.7, 123.1, 122.5, 119.9, 109.7, 43.9, 31.3, 29.8. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 271.0900; found: 271.0903.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)furan-3-yl)propan-2-one (3f)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a brown liquid (41.1 mg, 81%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.76 – 7.72 (m, 1H), 7.57 (d, *J* = 1.7 Hz, 1H), 7.38 – 7.36 (m, 1H), 7.32 – 7.27 (m, 2H), 6.57 (d, *J* = 1.7 Hz, 1H), 4.25 (s, 2H), 4.03 (s, 3H), 2.29 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.1, 144.4, 143.1, 142.8, 142.3, 135.7, 122.9, 122.4, 121.6, 119.6, 114.4, 109.3, 40.7, 31.6, 30.0. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 255.1128; found: 255.1129.

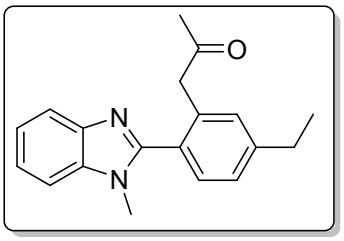
1-(5-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3g)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a brown liquid (53.4 mg, 96%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.81 – 7.76 (m, 1H), 7.37 (dt, *J* = 4.7, 3.1 Hz, 1H), 7.32 – 7.26 (m, 3H), 7.20 (dd, *J* = 7.8, 0.8 Hz, 1H), 7.12 (s, 1H), 3.95 (s, 2H), 3.66 (s, 3H), 2.40 (s, 3H), 1.94 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.0, 153.0, 142.4,

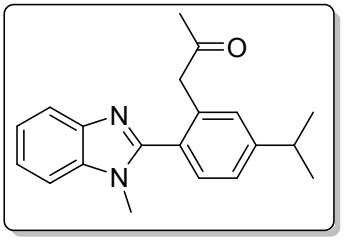
139.8, 135.4, 135.0, 131.9, 130.0, 127.6, 127.1, 122.5, 122.1, 119.3, 109.6, 47.9, 30.8, 29.7, 21.2. HRMS (EI, m/z) calcd for $C_{18}H_{18}N_2O$ $[M+H]^+$: 279.1492; found: 279.1494.

1-(5-ethyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3h)



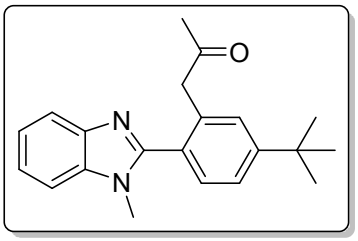
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (45.6 mg, 78%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (d, *J* = 6.7 Hz, 1H), 7.39 (d, *J* = 7.0 Hz, 1H), 7.34 – 7.29 (m, 3H), 7.24 (dd, *J* = 7.7, 1.0 Hz, 1H), 7.14 (s, 1H), 3.99 (s, 2H), 3.69 (s, 3H), 2.71 (t, *J* = 7.6 Hz, 2H), 1.96 (s, 3H), 1.29 (t, *J* = 7.6 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.1, 153.2, 146.2, 142.3, 135.5, 135.1, 130.9, 130.2, 127.3, 126.4, 122.6, 122.2, 119.4, 109.7, 48.2, 31.0, 30.0, 28.7, 15.6. HRMS (EI, m/z) calcd for $C_{19}H_{20}N_2O$ $[M+H]^+$: 293.1648; found: 293.1649.

1-(5-isopropyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3i)



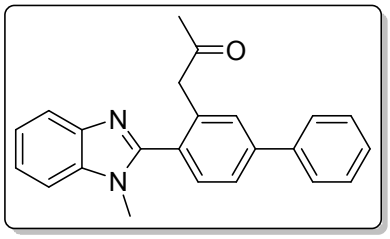
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a brown liquid (39.2 mg, 64%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.78 (d, *J* = 6.6 Hz, 1H), 7.39 (d, *J* = 7.0 Hz, 1H), 7.35 – 7.28 (m, 3H), 7.26 (dd, *J* = 7.8, 1.5 Hz, 1H), 7.15 (s, 1H), 4.00 (s, 2H), 3.70 (s, 3H), 2.96 (dt, *J* = 13.8, 6.9 Hz, 1H), 1.96 (s, 3H), 1.29 (d, *J* = 6.9 Hz, 6H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 206.1,153.3, 150.7, 142.5, 135.5, 135.1, 130.2, 129.5, 127.5, 124.9, 122.6, 122.2, 119.4, 109.7, 48.3, 33.9, 31.0, 29.8, 23.8. HRMS (EI, m/z) calcd for $C_{20}H_{22}N_2O$ $[M+H]^+$: 307.1805; found: 307.1807.

1-(5-(tert-butyl)-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3j)



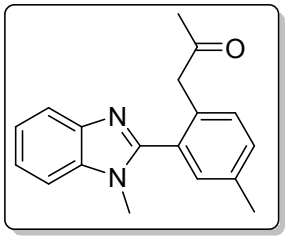
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 3:1, v/v) to give the product as a brown solid (53.1 mg, 83%). mp. 86 °C (lit. 86-87 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.77 (dd, *J* = 6.7, 1.8 Hz, 1H), 7.42 (dd, *J* = 8.0, 2.0 Hz, 1H), 7.40 – 7.37 (m, 1H), 7.34 – 7.29 (m, 4H), 4.02 (s, 2H), 3.70 (s, 3H), 1.96 (s, 3H), 1.36 (s, 9H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.2, 153.3, 152.9, 142.6, 135.5, 134.8, 129.9, 128.3, 127.3, 123.9, 122.5, 122.1, 119.5, 109.6, 48.5, 34.7, 31.2, 31.0, 29.8. HRMS (EI, m/z) calcd for $C_{21}H_{24}N_2O$ $[M+H]^+$: 321.1961; found: 321.1964.

1-(4-(1-methyl-1H-benzo[d]imidazol-2-yl)-[1,1'-biphenyl]-3-yl)propan-2-one (3k)



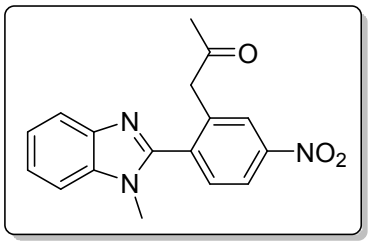
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (64.6 mg, 95%). mp. 120 °C (lit. 120-122 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.82 (dd, *J* = 6.5, 2.1 Hz, 1H), 7.67 – 7.63 (m, 3H), 7.55 (d, *J* = 1.8 Hz, 1H), 7.50 – 7.46 (m, 3H), 7.41 (ddd, *J* = 8.1, 6.2, 3.0 Hz, 2H), 7.37 – 7.32 (m, 2H), 4.11 (s, 2H), 3.75 (s, 3H), 2.00 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 205.8, 152.9, 142.6, 142.6, 140.0, 135.8, 135.6, 130.6, 130.0, 129.1, 128.8, 128.8, 127.8, 127.1, 127.1, 125.5, 122.7, 122.2, 119.6, 109.7, 48.3, 31.0, 29.8. HRMS (EI, m/z) calcd for $C_{23}H_{20}N_2O$ $[M+H]^+$: 341.1648; found: 341.1650.

1-(4-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl) propan-2-one (3l)



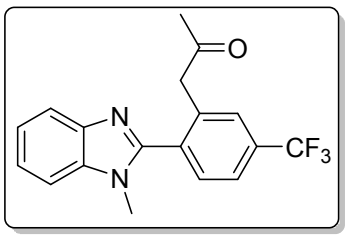
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow liquid (47.3 mg, 85%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (d, *J* = 7.3 Hz, 1H), 7.39 (d, *J* = 7.1 Hz, 1H), 7.34 – 7.27 (m, 3H), 7.23 – 7.17 (m, 2H), 3.93 (s, 2H), 3.68 (s, 3H), 2.40 (s, 3H), 1.95 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.1, 153.1, 142.3, 136.7, 135.4, 132.1, 131.1, 130.8, 130.7, 129.9, 122.7, 122.3, 119.5, 109.7, 47.7, 30.9, 29.8, 21.0. HRMS (EI, m/z) calcd for $C_{18}H_{18}N_2O$ $[M+H]^+$: 279.1492; found: 279.1493.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)-5-nitrophenyl)propan-2-one (3m)



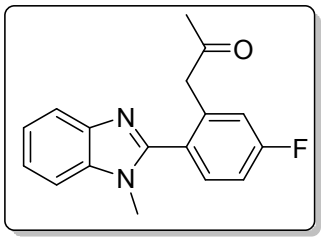
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 2:1, v/v) to give the product as a yellow solid (31.5 mg, 51%). mp. 136 °C (lit. 136-138 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.27 (dd, *J* = 8.4, 2.3 Hz, 1H), 8.18 (d, *J* = 2.2 Hz, 1H), 7.78 (d, *J* = 7.6 Hz, 1H), 7.59 (d, *J* = 8.4 Hz, 1H), 7.43 (d, *J* = 7.5 Hz, 1H), 7.35 (dtd, *J* = 16.3, 7.3, 1.2 Hz, 2H), 4.22 (s, 2H), 3.74 (s, 3H), 2.00 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 204.3,150.7, 148.2, 142.5, 137.7, 136.8, 135.6, 131.0, 126.4, 123.4, 122.8, 121.9, 119.9, 110.0, 48.0, 31.6, 29.9. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_3O_3$ $[M+H]^+$: 310.1186; found: 310.1187.

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)-5-(trifluoromethyl)phenyl)propan-2-one (3n)



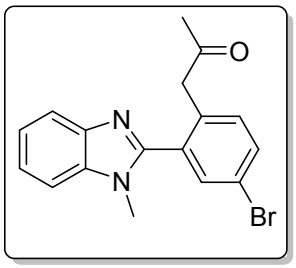
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (29.9 mg, 45%). mp. 111 °C (lit. 111-114 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (dd, *J* = 7.0, 1.4 Hz, 1H), 7.68 (dd, *J* = 8.0, 1.1 Hz, 1H), 7.57 (s, 1H), 7.52 (d, *J* = 8.0 Hz, 1H), 7.43 – 7.40 (m, 1H), 7.37 – 7.30 (m, 2H), 4.13 (s, 2H), 3.71 (s, 3H), 1.97 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 204.8, 151.6, 142.4, 136.5, 135.5, 134.0, 131.8, 130.6, 128.3 (q, *J* = 3.7 Hz), 124.8, 123.8 (q, *J* = 3.8 Hz), 123.1, 122.5, 119.7, 109.9, 47.9, 31.0, 29.9. HRMS (EI, m/z) calcd for $C_{18}H_{15}N_3OF_3$ $[M+H]^+$: 333.1209; found: 333.1210.

1-(5-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3o)



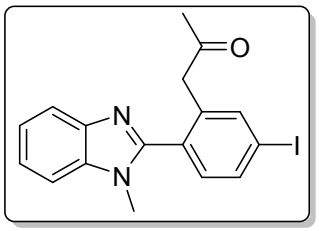
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a brown liquid (53.0 mg, 94%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.81 – 7.73 (m, 1H), 7.40 – 7.28 (m, 4H), 7.10 (td, *J* = 8.3, 2.6 Hz, 1H), 7.03 (dd, *J* = 9.3, 2.6 Hz, 1H), 3.99 (s, 2H), 3.67 (s, 3H), 1.96 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 205.0, 152.0, 142.4, 138.0, 135.5, 131.6(d, *J* = 8.8 Hz), 122.8, 122.3, 119.5, 118.5, 118.3, 114.1, 113.9, 109.7, 47.9, 30.9, 29.8. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_3OF$ $[M+H]^+$: 283.1241; found: 283.1243.

1-(4-bromo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3p)



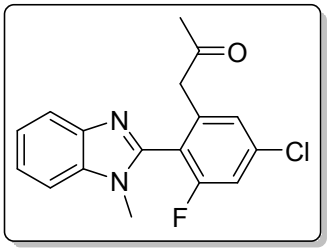
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow liquid (50.6 mg, 74%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.77 (dd, *J* = 7.0, 1.3 Hz, 1H), 7.59 (dd, *J* = 8.2, 2.1 Hz, 1H), 7.55 (d, *J* = 2.1 Hz, 1H), 7.40 (dd, *J* = 7.1, 1.4 Hz, 1H), 7.35 – 7.29 (m, 2H), 7.18 (d, *J* = 8.2 Hz, 1H), 3.97 (s, 2H), 3.71 (s, 3H), 1.96 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.1, 151.4, 142.5, 135.5, 134.4, 132.9, 123.9, 132.8, 132.3, 123.0, 122.4, 120.6, 119.7, 109.8, 47.5, 31.0, 29.8. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_2OBr$ $[M+H]^+$: 343.0441; found: 343.0442.

1-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3q)



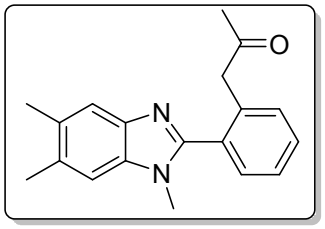
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow liquid (49.9 mg, 64%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.75 (dd, *J* = 11.1, 4.8 Hz, 2H), 7.67 (d, *J* = 1.5 Hz, 1H), 7.40 – 7.36 (m, 1H), 7.35 – 7.28 (m, 2H), 7.11 (d, *J* = 8.0 Hz, 1H), 3.98 (s, 2H), 3.68 (s, 3H), 1.95 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.0, 152.0, 142.5, 140.2, 137.4, 136.0, 135.5, 131.5, 129.8, 122.9, 122.3, 119.6, 109.7, 96.0, 47.5, 31.0, 29.9. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_2OI$ $[M+H]^+$: 391.0302; found: 391.0305.

1-(5-chloro-3-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3r)



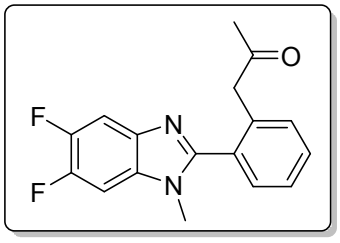
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (35.4 mg, 56%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.77 (d, *J* = 7.5 Hz, 1H), 7.41 (d, *J* = 7.5 Hz, 1H), 7.33 (dtd, *J* = 16.2, 7.2, 1.0 Hz, 2H), 7.16 (dd, *J* = 9.1, 1.9 Hz, 1H), 7.06 (s, 1H), 3.66 (d, *J* = 1.2 Hz, 3H), 1.86 (s, 3H), 0.07 (s, 2H).^{13}C NMR (126 MHz, $CDCl_3$) δ 204.3, 161.4, 159.5, 146.8, 142.6, 139.2 (d, *J* = 3.2 Hz), 136.7, 135.4, 127.3 (d, *J* = 3.1 Hz), 123.1, 122.4, 119.6 (, 115.3, 115.1, 109.8, 47.4 (d, *J* = 2.2 Hz), 29.8, 1.0. HRMS (EI, m/z) calcd for $C_{17}H_{14}ClFN_2O$ $[M+H]^+$: 317.0851; found: 317.0855.

1-(2-(1,5,6-trimethyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3s)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (44.4 mg, 76%). mp. 96 °C (lit. 96-98 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.55 (s, 1H), 7.45 (ddd, *J* = 7.6, 6.4, 2.7 Hz, 1H), 7.41 – 7.37 (m, 2H), 7.30 (d, *J* = 7.6 Hz, 1H), 7.16 (s, 1H), 3.97 (s, 2H), 3.63 (s, 3H), 2.42 (s, 3H), 2.40 (s, 3H), 1.96 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.9, 152.1, 141.0, 135.3, 134.0, 131.8, 131.2, 131.1, 130.4, 130.2, 129.7, 126.8, 119.5, 109.9, 48.0, 30.8, 29.8, 20.5, 20.2. HRMS (EI, m/z) calcd for $C_{19}H_{20}N_2O$ $[M+H]^+$: 293.1648; found: 293.1650.

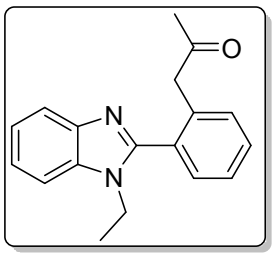
1-(2-(5,6-difluoro-1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3t)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (42.6 mg, 71%). mp. 90 °C (lit. 90-91 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ

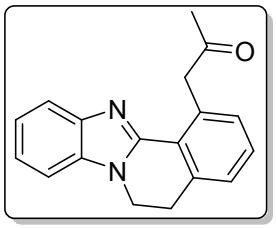
7.54 – 7.46 (m, 2H), 7.40 (dtd, *J* = 9.0, 7.5, 1.4 Hz, 2H), 7.32 – 7.29 (m, 1H), 7.16 (dd, *J* = 9.7, 6.9 Hz, 1H), 4.01 (s, 2H), 3.65 (s, 3H), 1.97 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 205.7, 154.6, 149.2 (d, *J* = 15.7 Hz), 148.8 (d, *J* = 15.0 Hz), 147.2 (d, *J* = 15.7 Hz), 146.9 (d, *J* = 15.0 Hz), 137.8 – 137.5 (m), 135.3, 131.5, 130.9 (d, *J* = 10.6 Hz), 130.1(d, *J* = 12.4 Hz), 129.8, 127.0, 106.8 (d, *J* = 19.5 Hz), 97.8, 97.6, 48.2, 31.3, 29.8. HRMS (EI, m/z) calcd for $C_{17}H_{14}N_2OF_2$ $[M+H]^+$: 301.1147; found: 301.1148.

1-(2-(1-ethyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3u)



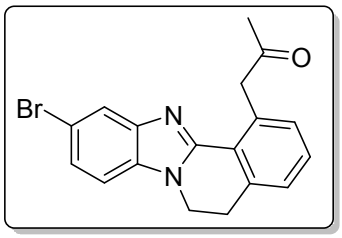
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (34.5 mg, 62%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 – 7.77 (m, 1H), 7.48 (ddd, *J* = 7.7, 5.8, 3.2 Hz, 1H), 7.45 – 7.39 (m, 3H), 7.34 – 7.29 (m, 3H), 4.10 (q, *J* = 7.3 Hz, 2H), 3.91 (s, 2H), 1.99 (s, 3H), 1.38 (t, *J* = 7.3 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.8, 152.4, 142.8, 135.2, 134.3, 131.3, 130.4, 129.9, 126.9, 122.6, 122.2, 119.8, 110.1, 47.9, 39.3, 29.9, 15.0. HRMS (EI, m/z) calcd for $C_{18}H_{18}N_2O$ $[M+H]^+$: 279.1492; found: 279.1494.

1-(5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (3v)



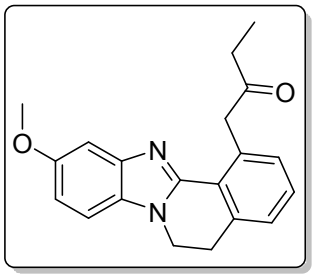
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 3:1, v/v) to give the product as a yellow solid (52.4 mg, 95%). mp. 95 °C (lit. 95-97 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.75 (dd, *J* = 6.6, 1.8 Hz, 1H), 7.34 (ddd, *J* = 7.5, 4.2, 3.0 Hz, 2H), 7.30 – 7.23 (m, 3H), 7.18 (d, *J* = 7.5 Hz, 1H), 4.55 (s, 2H), 4.28 (t, *J* = 6.8 Hz, 2H), 3.25 (t, *J* = 6.8 Hz, 2H), 2.46 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.1, 148.5, 143.6, 135.7, 134.6, 133.8, 131.5, 129.4, 127.2, 125.3, 122.6, 122.0, 119.7, 108.9, 49.9, 40.0, 30.1, 29.3. HRMS (EI, m/z) calcd for $C_{18}H_{16}N_2O$ $[M+H]^+$: 277.1335; found: 277.1337.

1-(10-bromo-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (3w)



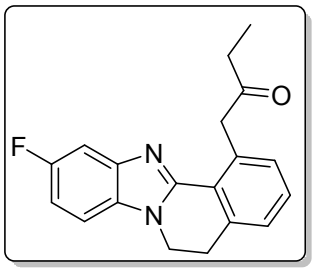
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow solid (58.8 mg, 83%). mp. 96 °C (lit. 96-98 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.86 (d, *J* = 1.6 Hz, 1H), 7.36 – 7.32 (m, 2H), 7.24 (d, *J* = 7.0 Hz, 1H), 7.19 – 7.15 (m, 2H), 4.48 (s, 2H), 4.23 (t, *J* = 6.8 Hz, 2H), 3.23 (t, *J* = 6.8 Hz, 2H), 2.44 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.8, 149.5, 144.8, 135.6, 134.8, 132.7, 131.7, 129.8, 127.2, 125.5, 124.9, 122.4, 114.8, 110.1, 49.9, 40.2, 30.1, 29.1. HRMS (EI, m/z) calcd for $C_{18}H_{15}N_2OBr$ $[M+H]^+$: 355.0441; found: 355.0442.

1-(10-methoxy-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3x)



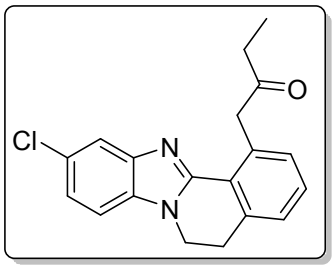
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (60.8 mg, 95%). mp. 103 °C (lit. 103-104 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 (t, *J* = 7.5 Hz, 1H), 7.21 (dd, *J* = 12.1, 6.0 Hz, 3H), 7.16 (d, *J* = 7.5 Hz, 1H), 6.92 (dd, *J* = 8.7, 2.3 Hz, 1H), 4.53 (s, 2H), 4.23 (t, *J* = 6.8 Hz, 2H), 3.88 (s, 3H), 3.22 (t, *J* = 6.8 Hz, 2H), 2.81 (q, *J* = 7.3 Hz, 2H), 1.14 (t, *J* = 7.3 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 209.6, 156.0, 148.8, 144.2, 135.3, 134.5, 131.5, 129.2, 128.5, 127.0, 125.4, 112.7, 109.2, 101.8, 55.8, 48.8, 40.1, 35.8, 29.3, 8.0. HRMS (EI, m/z) calcd for $C_{20}H_{20}N_2O_2$ $[M+H]^+$: 321.1598; found: 321.1599.

1-(10-fluoro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3y)



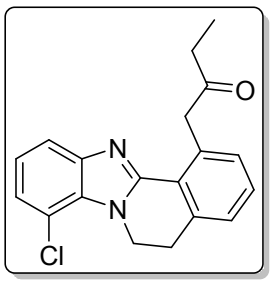
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (41.3 mg, 67%). mp. 111 °C (lit. 111-113 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.39 – 7.30 (m, 2H), 7.24 – 7.20 (m, 2H), 7.16 (d, *J* = 7.4 Hz, 1H), 7.01 (td, *J* = 9.1, 2.4 Hz, 1H), 4.49 (s, 2H), 4.23 (t, *J* = 6.8 Hz, 2H), 3.22 (t, *J* = 6.8 Hz, 2H), 2.81 (q, *J* = 7.4 Hz, 2H), 1.15 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 209.5, 160.2, 158.3, 149.9, 135.4, 134.8, 131.7, 130.3, 129.6, 127.1, 125.0, 110.9, 110.7, 109.1 (d, *J* = 10.3 Hz), 105.3, 105.1, 48.9 40.2, 35.8, 29.1, 8.0. HRMS (EI, m/z) calcd for $C_{19}H_{17}N_2OF$ $[M+H]^+$: 309.1398; found: 309.1399.

1-(10-chloro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3z)



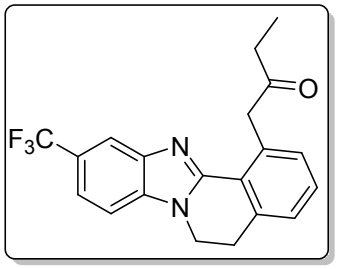
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (42.0 mg, 65%). mp. 112 °C (lit. 112-114 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.74 – 7.62 (m, 1H), 7.34 (t, *J* = 7.6 Hz, 1H), 7.26 – 7.20 (m, 3H), 7.16 (d, *J* = 7.4 Hz, 1H), 4.47 (s, 2H), 4.24 (t, *J* = 6.8 Hz, 2H), 3.24 (t, *J* = 6.8 Hz, 2H), 2.81 (q, *J* = 7.4 Hz, 2H), 1.15 (t, *J* = 7.4 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 209.4, 149.7, 144.3, 135.6, 134.9, 132.4, 131.7, 129.8, 127.4, 127.1, 124.9, 123.0, 119.3, 109.6, 48.9, 40.2, 35.9, 29.2, 8.0. HRMS (EI, m/z) calcd for $C_{19}H_{17}N_2OCl$ $[M+H]^+$: 325.1102; found: 325.1105.

1-(8-chloro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3aa)



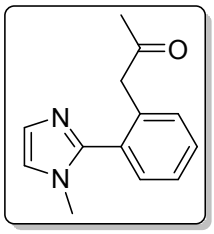
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (60.3 mg, 93%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.59 (dd, *J* = 7.9, 1.0 Hz, 1H), 7.33 (t, *J* = 7.6 Hz, 1H), 7.24 – 7.20 (m, 1H), 7.19 – 7.10 (m, 3H), 4.78 (t, *J* = 6.7 Hz, 2H), 4.45 (s, 2H), 3.21 (t, *J* = 6.7 Hz, 2H), 2.81 (q, *J* = 7.4 Hz, 2H), 1.15 (t, *J* = 7.4 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 209.3, 149.5, 145.3, 135.8, 134.9, 131.6, 129.7, 126.8, 125.8, 124.8, 123.9, 122.3, 118.3, 115.7, 48.9, 42.0, 35.7, 29.4, 7.9. HRMS (EI, m/z) calcd for $C_{19}H_{17}N_2OCl$ $[M+H]^+$: 325.1102; found: 325.1103.

1-(10-(trifluoromethyl)-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3ab)



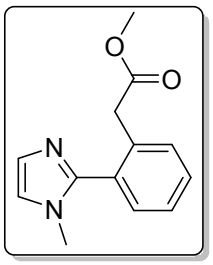
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (56.6 mg, 79%). mp. 165 °C (lit. 165-167 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.97 (s, 1H), 7.50 (dd, *J* = 8.4, 1.1 Hz, 1H), 7.37 (dd, *J* = 17.1, 8.1 Hz, 2H), 7.24 (d, *J* = 6.0 Hz, 1H), 7.18 (d, *J* = 7.5 Hz, 1H), 4.49 (s, 2H), 4.29 (t, *J* = 6.8 Hz, 2H), 3.25 (t, *J* = 6.7 Hz, 2H), 2.83 (q, *J* = 7.4 Hz, 2H), 1.16 (t, *J* = 7.4 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 209.4, 150.4, 142.9, 135.8, 135.7, 135.1, 131.8, 130.0, 129.5, 127.2, 126.3, 124.8, 119.5 (dd, *J* = 7.1, 3.6 Hz), 117.2 (q, *J* = 4.1 Hz), 109.2, 48.9, 40.3, 35.9, 29.1, 8.0. HRMS (EI, m/z) calcd for $C_{20}H_{17}N_2OF_3$ $[M+H]^+$: 359.1366; found: 359.1368.

1-(2-(1-methyl-1H-imidazol-2-yl)phenyl)propan-2-one (3ac)



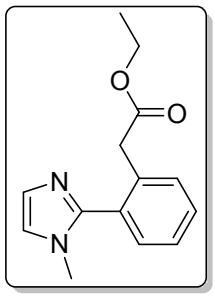
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (41.0 mg, 96%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.39 (td, *J* = 7.4, 1.7 Hz, 1H), 7.33 (dtd, *J* = 8.9, 7.5, 1.4 Hz, 2H), 7.25 (dd, *J* = 7.5, 0.7 Hz, 1H), 7.12 (s, 1H), 6.95 (s, 1H), 3.89 (s, 2H), 3.53 (s, 3H), 1.97 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 201.8, 146.2, 135.6, 131.1, 129.9, 129.5, 128.5, 126.9, 121.2, 47.8, 33.8, 29.5.
HRMS (EI, m/z) calcd for $C_{13}H_{14}N_2O$ $[M+H]^+$: 215.1179; found: 215.1181.

methyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4a)



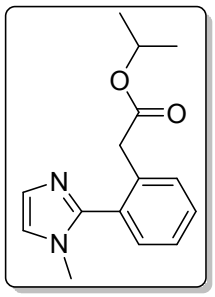
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (28.1 mg, 61%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.41 – 7.38 (m, 1H), 7.35 (dd, *J* = 7.3, 6.1 Hz, 2H), 7.30 (dd, *J* = 6.7, 2.5 Hz, 1H), 7.10 (d, *J* = 1.1 Hz, 1H), 6.95 (d, *J* = 1.1 Hz, 1H), 3.79 (s, 2H), 3.55 (s, 3H), 3.52 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 172.0, 146.6, 135.0, 130.9, 130.8, 130.3, 129.3, 128.0, 127.0, 120.9, 51.8, 38.6, 33.5. HRMS (EI, m/z) calcd for $C_{13}H_{14}N_2O_2$ $[M+H]^+$: 231.1128; found: 231.1130.

ethyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4b)



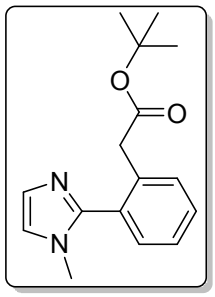
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (27.8 mg, 57%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.42 – 7.38 (m, 1H), 7.35 (ddd, *J* = 8.8, 5.3, 1.7 Hz, 2H), 7.31 – 7.29 (m, 1H), 7.10 (d, *J* = 1.2 Hz, 1H), 6.95 (d, *J* = 1.2 Hz, 1H), 3.99 (q, *J* = 7.1 Hz, 2H), 3.78 (s, 2H), 3.52 (s, 3H), 1.15 (t, *J* = 7.1 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.5, 146.7, 135.2, 130.9, 130.3, 129.3, 128.0, 126.9, 120.8, 60.6, 38.8, 33.6, 14.1. HRMS (EI, m/z) calcd for $C_{14}H_{16}N_2O_2$ $[M+H]^+$: 245.1285; found: 245.1287.

isopropyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4c)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (29.9 mg, 58%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.40 – 7.36 (m, 1H), 7.35 – 7.31 (m, 2H), 7.29 – 7.26 (m, 1H), 7.08 (d, *J* = 1.3 Hz, 1H), 6.93 (d, *J* = 1.2 Hz, 1H), 4.82 (dt, *J* = 12.5, 6.3 Hz, 1H), 3.72 (s, 2H), 3.51 (s, 3H), 1.11 (d, *J* = 6.3 Hz, 6H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.0, 146.7, 135.3, 130.8, 130.3, 129.2, 127.9, 126.8, 126.3, 120.8, 67.9, 39.1, 33.5, 21.7. HRMS (EI, m/z) calcd for $C_{15}H_{18}N_2O_2$ $[M+H]^+$: 259.1441; found: 259.1442.

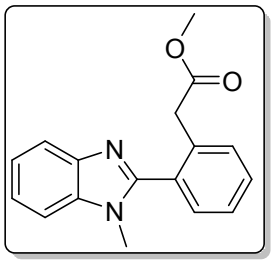
tert-butyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4d)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow liquid (23.4 mg, 43%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.40 – 7.37 (m, 1H), 7.34 (dd, *J* = 8.6, 1.0 Hz, 2H), 7.28 (t, *J* = 3.9 Hz, 1H), 7.10 (d, *J* = 1.2 Hz, 1H), 6.93 (d, *J* = 1.2 Hz, 1H), 3.68 (s, 2H), 3.51 (s, 3H), 1.32 (s, 9H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.9, 146.8, 135.6, 130.8, 130.3, 129.2, 128.0, 126.7, 120.7,

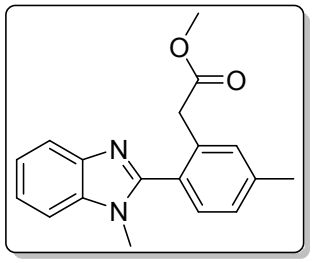
80.4, 40.0, 33.5, 27.9, 27.6. HRMS (EI, m/z) calcd for $C_{16}H_{20}N_2O_2$ $[M+H]^+$: 273.1598; found: 273.1600.

Methyl2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4e)



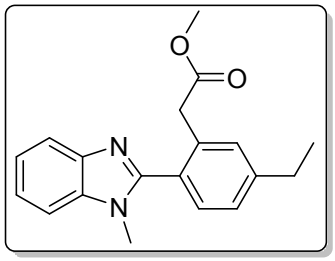
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (44.8 mg, 80%). mp. 85 °C (lit. 85-86 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.80 (d, *J* = 7.0 Hz, 1H), 7.50 – 7.45 (m, 1H), 7.44 – 7.38 (m, 4H), 7.35 – 7.29 (m, 2H), 3.86 (s, 2H), 3.67 (s, 3H), 3.44 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.7, 152.8, 142.7, 135.5, 134.7, 130.9, 130.4, 130.2, 129.9, 127.0, 122.6, 122.1, 119.7, 109.6, 51.7, 38.4, 30.8. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O_2$ $[M+H]^+$: 281.1285; found: 281.1286.

methyl 2-(5-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4f)



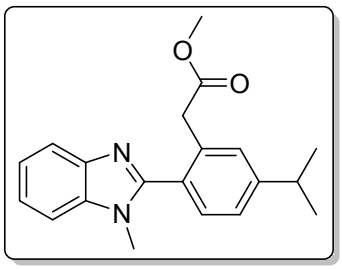
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (48.2 mg, 82%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.81 – 7.77 (m, 1H), 7.41 – 7.37 (m, 1H), 7.34 – 7.27 (m, 3H), 7.24 – 7.20 (m, 2H), 3.82 (s, 2H), 3.66 (s, 3H), 3.44 (s, 3H), 2.42 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.8, 152.9, 142.7, 139.9, 135.5, 134.5, 131.6, 130.1, 127.8, 127.4, 122.5, 122.1, 119.6, 109.5, 51.7, 38.4, 30.8, 21.3. HRMS (EI, m/z) calcd for $C_{18}H_{18}N_2O_2$ $[M+H]^+$: 295.1441; found: 295.1442.

methyl 2-(5-ethyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4g)



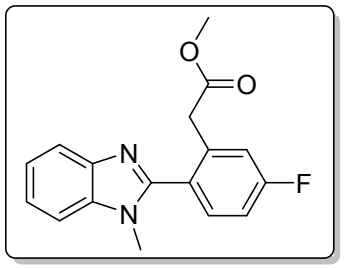
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow solid (47.4 mg, 77%). mp. 85 °C (lit. 85-87 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.82 – 7.77 (m, 1H), 7.41 – 7.38 (m, 1H), 7.34 – 7.29 (m, 3H), 7.24 (d, *J* = 6.2 Hz, 2H), 3.84 (s, 2H), 3.68 (s, 3H), 3.45 (s, 3H), 2.73 (q, *J* = 7.6 Hz, 2H), 1.28 (d, *J* = 7.6 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.9, 153.1, 146.1, 142.8, 135.6, 134.6, 130.5, 130.2, 127.7, 126.6, 122.5, 122.1, 119.7, 109.5, 51.7, 38.5, 30.9 28.6, 15.2. HRMS (EI, m/z) calcd for $C_{19}H_{20}N_2O_2$ $[M+H]^+$: 309.1598; found: 309.1599.

Methyl 2-(5-isopropyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate(4h)



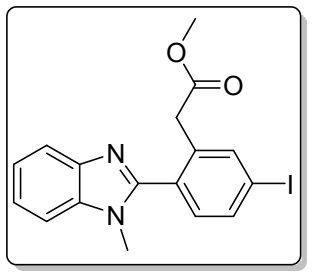
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow solid (39.3 mg, 61%). mp. 103 °C (lit. 103-104 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.81 – 7.77 (m, 1H), 7.41 – 7.38 (m, 1H), 7.32 (qd, *J* = 6.8, 1.5 Hz, 3H), 7.28 (dd, *J* = 4.5, 1.5 Hz, 2H), 3.85 (s, 2H), 3.69 (s, 3H), 3.44 (s, 3H), 2.97 (dt, *J* = 13.8, 6.9 Hz, 1H), 1.30 (d, *J* = 6.9 Hz, 6H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.9, 153.1, 150.7, 142.8, 135.5, 134.5, 130.2, 129.2, 127.8, 125.1, 122.4, 122.0, 119.7, 109.5, 51.7, 38.6, 33.9, 30.9, 23.8. HRMS (EI, m/z) calcd for $C_{20}H_{22}N_2O_2$ $[M+H]^+$: 323.1754; found: 323.11756.

methyl 2-(5-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4i)



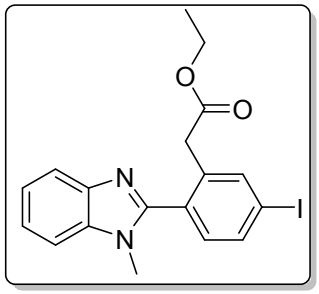
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a yellow solid (41.1 mg, 69%). mp. 109 °C (lit. 109-110 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.79 (dd, *J* = 6.8, 1.6 Hz, 1H), 7.39 (td, *J* = 8.2, 3.5 Hz, 2H), 7.35 – 7.30 (m, 2H), 7.18 – 7.10 (m, 2H), 3.83 (s, 2H), 3.66 (s, 3H), 3.47 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.1, 164.2, 162.2, 151.8, 142.6, 137.49 (d, *J* = 8.3 Hz), 135.5, 132.1 (d, *J* = 8.8 Hz), 122.8, 122.3, 119.8, 118.1, 118.0, 114.4, 114.2, 109.6, 51.9, 38.4, 30.8. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_2O_2F$ $[M+H]^+$: 299.1190; found: 299.1192.

methyl 2-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4j)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc =3:1, v/v) to give the product as a yellow solid (37.4 mg, 46%). mp. 110 °C (lit. 110-112 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.77 (ddd, *J* = 9.7, 7.3, 1.5 Hz, 3H), 7.41 – 7.38 (m, 1H), 7.33 (ddd, *J* = 8.8, 7.0, 1.3 Hz, 2H), 7.12 (d, *J* = 8.0 Hz, 1H), 3.82 (s, 2H), 3.67 (s, 3H), 3.44 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 171.1, 151.8, 142.7, 139.9, 136.9, 136.2, 135.5, 131.6, 130.1, 122.9, 122.3, 119.8, 109.6, 96.1, 51.9, 38.0, 30.9. HRMS (EI, m/z) calcd for $C_{17}H_{15}N_2O_2$ I $[M+H]^+$: 407.0251; found: 407.0252.

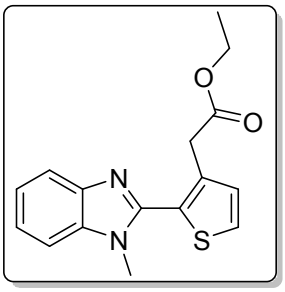
ethyl 2-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4k)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a white solid (51.2 mg, 61%).

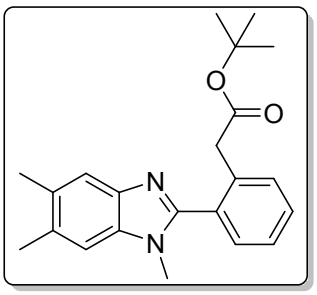
mp. 115 °C (lit. 115-117 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.80 – 7.75 (m, 3H), 7.40 (dd, *J* = 7.0, 1.5 Hz, 1H), 7.32 (tt, *J* = 8.5, 3.6 Hz, 2H), 7.13 (d, *J* = 8.0 Hz, 1H), 3.87 (q, *J* = 7.1 Hz, 2H), 3.81 (s, 2H), 3.67 (s, 3H), 0.98 (t, *J* = 7.1 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.7, 151.8, 142.7, 140.0, 137.1, 136.2, 135.5, 131.6, 130.1, 122.9, 122.3, 119.8, 109.6, 96.1, 60.8, 38.4, 30.9, 13.8. HRMS (EI, m/z) calcd for $C_{18}H_{17}N_2O_2I$ $[M+H]^+$: 421.0407; found: 421.0409.

ethyl 2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)thiophen-3-yl)acetate (4l)



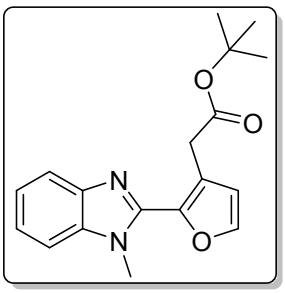
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow liquid (41.1 mg, 75%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 – 7.78 (m, 1H), 7.49 (d, *J* = 5.2 Hz, 1H), 7.41 – 7.36 (m, 1H), 7.32 (tt, *J* = 8.5, 3.6 Hz, 2H), 7.18 (d, *J* = 5.2 Hz, 1H), 4.07 (q, *J* = 7.1 Hz, 2H), 3.84 (s, 2H), 3.80 (s, 3H), 1.17 (t, *J* = 7.1 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.8, 147.0, 142.9, 136.4, 135.9, 129.7, 127.3, 126.8, 123.0, 122.5, 119.9, 109.6, 60.9, 34.6, 31.2, 14.0. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 301.1005; found: 301.1008.

tert-butyl 2-(2-(1,5,6-trimethyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4m)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a brown liquid (43.4 mg, 62%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.55 (s, 1H), 7.45 (ddd, *J* = 7.9, 6.0, 2.7 Hz, 1H), 7.41 – 7.36 (m, 3H), 7.15 (s, 1H), 3.69 (s, 2H), 3.60 (s, 3H), 2.42 (s, 3H), 2.40 (s, 3H), 1.21 (s, 9H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.7, 152.0, 141.4, 135.4, 134.2, 131.7, 130.8, 130.6, 130.2, 129.7, 126.8, 119.7, 109.7, 80.6, 39.9, 30.8, 27.8, 27.8, 20.6, 20.3. HRMS (EI, m/z) calcd for $C_{22}H_{26}N_2O_2$ $[M+H]^+$: 351.2067; found: 351.2069.

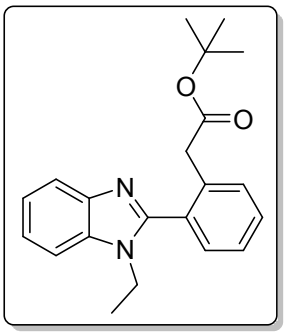
tert-butyl 2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)furan-3-yl)acetate (4n)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 4:1, v/v) to give the product as a brown liquid (44.9 mg, 72%).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.76 (dd, *J* = 6.8, 1.7 Hz, 1H), 7.55 (d, *J* = 1.8 Hz, 1H), 7.37 (dd, *J* = 6.9, 1.8 Hz, 1H), 7.32 – 7.27 (m, 2H), 6.66 (d, *J* = 1.8 Hz, 1H), 4.04 (s, 2H), 4.00 (s, 3H), 1.45 (s, 9H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.5, 144.5, 143.1, 142.7, 142.1, 135.7, 122.8, 122.3, 121.8, 119.7, 114.1, 109.3, 80.8, 32.6,

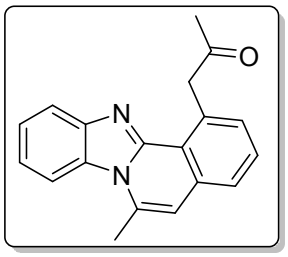
31.5, 28.1. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 313.1547; found: 313.1548.

tert-butyl 2-(2-(1-ethyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate(4o)



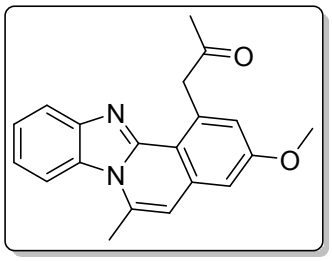
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 5:1, v/v) to give the product as a yellow solid (43.0 mg, 64%). mp. 100 °C (lit. 100-102 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.84 – 7.77 (m, 1H), 7.50 – 7.46 (m, 1H), 7.44 – 7.37 (m, 4H), 7.34 – 7.28 (m, 2H), 4.09 (q, *J* = 7.3 Hz, 2H), 3.68 (s, 2H), 1.38 (t, *J* = 7.3 Hz, 3H), 1.22 (s, 9H).^{13}C NMR (126 MHz, $CDCl_3$) δ 170.6, 152.4, 143.1, 135.3, 134.4, 130.9, 130.6, 129.9, 129.8, 126.8, 122.5, 122.0, 119.9, 109.9, 80.6, 39.9, 39.3, 27.7, 15.0. HRMS (EI, m/z) calcd for $C_{21}H_{24}N_2O_2$ $[M+H]^+$: 337.1911; found: 337.1914.

1-(6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6a)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (54.7 mg, 95%).mp. 168 °C (lit. 168-170 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.05 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.1 Hz, 1H), 7.58 – 7.53 (m, 2H), 7.46 (t, *J* = 7.5 Hz, 1H), 7.36 – 7.30 (m, 2H), 6.72 (s, 1H), 4.76 (s, 2H), 2.94 (d, *J* = 0.5 Hz, 3H), 2.59 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.1, 147.8, 143.9, 134.6, 134.4, 133.0, 130.7, 130.3, 129.1, 125.1, 123.7, 121.6, 120.9, 119.9, 113.8, 111.3, 51.1, 30.2, 21.1.HRMS (EI, m/z) calcd for $C_{19}H_{16}N_2O$ $[M+H]^+$: 289.1335; found: 289.1336.

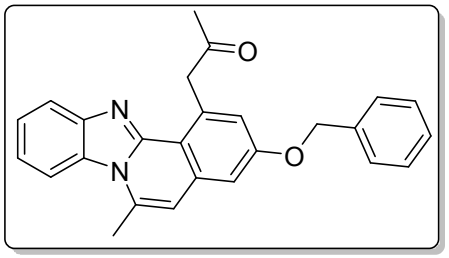
1-(3-methoxy-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6b)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (46.4 mg, 73%). mp. 205 °C (lit. 205-206 °C). 1H NMR (500 MHz, CDCl3) δ 7.95 (d, J = 8.0 Hz, 1H), 7.60 – 7.55 (m, 2H), 7.35 (dd, J = 6.4, 1.7 Hz, 1H), 7.29 – 7.24 (m, 3H), 6.79 (s, 1H), 4.86 (s, 2H), 3.01 (s, 3H), 2.81 (s, 3H), 2.51 (s, 3H).13C NMR (126 MHz, CDCl3) δ 156.7, 147.0, 134.7, 134.2, 133.1, 130.7, 130.0, 129.7, 129.1, 125.2, 124.2, 121.7, 111.6, 111.4, 51.2, 30.3, 21.3, 17.8. HRMS (EI, m/z) calcd for $C_{20}H_{18}N_2O_2$ $[M+H]^+$: 319.1441; found: 319.1443.

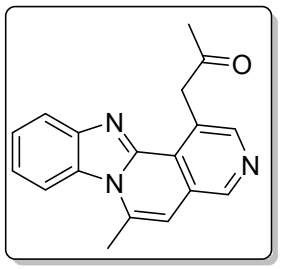
1-(3-(benzyloxy)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one

(6c)



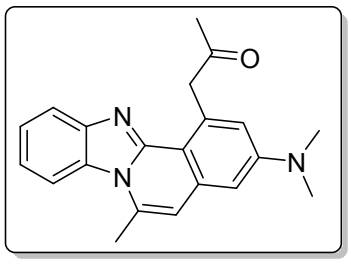
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (71.0 mg, 90%). mp. 151 °C (lit. 151-153 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.97 (d, *J* = 8.3 Hz, 1H), 7.88 (d, *J* = 7.3 Hz, 1H), 7.42 (ddd, *J* = 28.3, 18.5, 7.2 Hz, 6H), 7.27 (t, *J* = 7.3 Hz, 1H), 7.04 (s, 1H), 6.96 (d, *J* = 1.6 Hz, 1H), 6.58 (s, 1H), 5.12 (s, 2H), 4.70 (s, 2H), 2.85 (s, 3H), 2.59 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 206.8, 159.0, 148.0, 144.0, 136.4, 135.0, 134.9, 130.3, 128.6, 128.1, 127.5, 123.6, 121.1, 120.5, 119.4, 115.0, 113.7, 111.1, 107.3, 70.0, 51.1, 30.2, 21.0. HRMS (EI, m/z) calcd for $C_{26}H_{22}N_2O_2$ $[M+H]^+$: 395.1754; found: 395.1756.

1-(6-methylbenzo[4,5]imidazo[2,1-a][2,6]naphthyridin-1-yl)propan-2-one (6d)



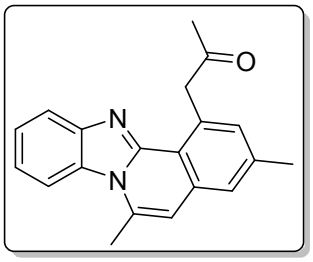
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 1:2, v/v) to give the product as a yellow solid (43.9 mg, 76%). mp. 210 °C (lit. 210-211 °C). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.94 (s, 1H), 8.49 (s, 1H), 8.10 (d, *J* = 8.5 Hz, 1H), 7.97 (d, *J* = 8.2 Hz, 1H), 7.53 – 7.49 (m, 1H), 7.40 (ddd, *J* = 8.3, 7.2, 1.1 Hz, 1H), 6.82 (s, 1H), 4.67 (s, 2H), 3.02 (s, 3H), 2.60 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.9, 148.6, 147.8, 145.7, 144.0, 136.8, 130.4, 127.0, 125.8, 124.5, 122.9, 120.6, 114.1, 47.4, 30.3, 21.4. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 290.1288; found: 290.1290.

1-(3-(dimethylamino)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6e)



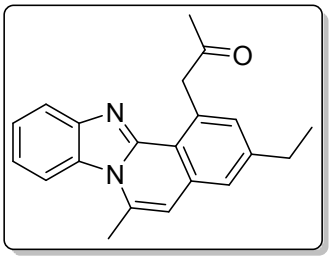
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 3:1, v/v) to give the product as a yellow solid (49.7 mg, 75%). mp. 193 °C (lit. 193-194 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.98 (d, *J* = 8.3 Hz, 1H), 7.84 (s, 1H), 7.40 (t, *J* = 7.3 Hz, 1H), 7.23 (d, *J* = 7.5 Hz, 1H), 6.77 (d, *J* = 1.8 Hz, 1H), 6.63 (d, *J* = 21.6 Hz, 2H), 4.72 (s, 2H), 3.07 (s, 6H), 2.91 (s, 3H), 2.55 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.5, 150.4, 148.7, 135.5, 134.9, 134.5, 130.5, 123.3, 120.5, 119.0, 116.8, 113.4, 111.4, 105.2, 51.4, 40.1, 30.1, 21.1. HRMS (EI, m/z) calcd for $C_{21}H_{21}N_3O$ $[M+H]^+$: 332.11757; found: 332.1758.

1-(3,6-dimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6f)



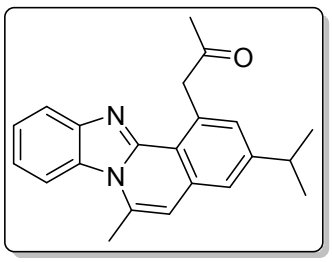
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (58.0 mg, 96%). mp. 154 °C (lit. 154-155 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.02 (d, *J* = 8.4 Hz, 1H), 7.90 (d, *J* = 7.8 Hz, 1H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.33 – 7.27 (m, 2H), 7.17 (s, 1H), 6.63 (s, 1H), 4.71 (s, 2H), 2.91 (s, 3H), 2.59 (s, 3H), 2.49 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.2, 148.0, 144.0, 139.2, 134.5, 134.2, 133.2, 132.3, 130.4, 125.0, 123.6, 121.3, 119.6, 118.6, 113.7, 111.1, 51.0, 30.2, 21.5, 21.1. HRMS (EI, m/z) calcd for $C_{20}H_{18}N_2O$ $[M+H]^+$: 303.1492; found: 303.1493.

1-(3-ethyl-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6g)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 10:1, v/v) to give the product as a white solid (51.8 mg, 82%). mp. 128 °C (lit. 128-129 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.01 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 8.1 Hz, 1H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.34 (s, 1H), 7.29 (t, *J* = 7.7 Hz, 1H), 7.20 (s, 1H), 6.65 (s, 1H), 4.73 (s, 2H), 2.90 (s, 3H), 2.81 (q, *J* = 7.6 Hz, 2H), 1.36 (t, *J* = 7.6 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.2, 148.0, 145.3, 143.9, 134.4, 134.2, 133.3, 131.2, 130.3, 123.7, 123.6, 121.3, 119.6, 118.8, 113.7, 111.2, 51.1, 30.2, 28.7, 21.2, 15.0. HRMS (EI, m/z) calcd for $C_{21}H_{20}N_2O$ $[M+H]^+$: 317.1648; found: 317.1647.

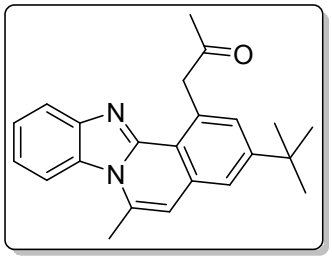
1-(3-isopropyl-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6h)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a white solid (57.4 mg, 87%). mp. 114 °C (lit. 114-116 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.04 (d, *J* = 8.3 Hz, 1H), 7.91 (d, *J* = 7.1 Hz, 1H), 7.44 (dd, *J* = 17.4, 9.9 Hz, 2H), 7.30 (t, *J* = 7.5 Hz, 1H), 7.24 (s, 1H), 6.71 (s, 1H), 4.74 (s, 2H), 3.07 (dt, *J* = 13.5, 6.7 Hz, 1H), 2.94 (s, 3H), 2.58 (s, 3H), 1.37 (d, *J* = 6.8 Hz, 6H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.2, 149.9, 148.0, 144.0, 134.4, 134.3, 133.3, 130.3, 130.0, 123.6, 122.3, 121.3, 119.7, 119.0, 113.7, 111.4, 51.2, 34.0, 30.1, 23.8, 21.1. HRMS (EI, m/z) calcd for $C_{22}H_{22}N_2O$ $[M+H]^+$: 331.1805; found: 331.1807.

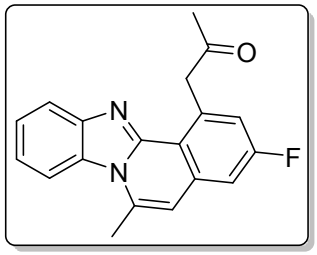
1-(3-(tert-butyl)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one

(6i)



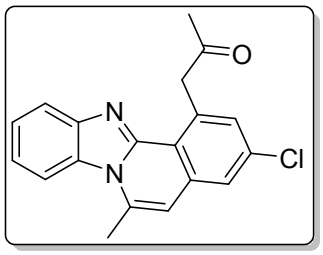
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (64.7 mg, 94%). mp. 147 °C (lit. 147-149 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.07 (d, *J* = 8.4 Hz, 1H), 7.92 (d, *J* = 8.0 Hz, 1H), 7.56 (d, *J* = 1.1 Hz, 1H), 7.44 (dd, *J* = 19.7, 12.1 Hz, 2H), 7.31 (t, *J* = 7.7 Hz, 1H), 6.76 (s, 1H), 4.77 (s, 2H), 2.98 (s, 3H), 2.57 (s, 3H), 1.44 (s, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 207.1, 152.2, 148.0, 144.0, 134.4, 134.0, 133.0, 130.4, 128.8, 123.6, 121.4， 121.3, 119.8, 118.7, 113.8, 111.7, 51.4, 34.9, 31.2, 30.1, 21.2. HRMS (EI, m/z) calcd for $C_{23}H_{24}N_2O$ $[M+H]^+$: 345.1961; found: 345.1963.

1-(3-fluoro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6j)



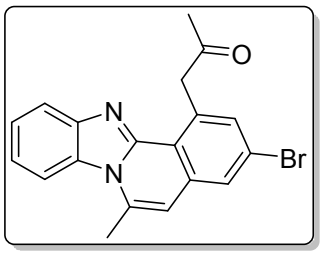
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a white solid (47.7 mg, 78%). mp. 163 °C (lit. 163-165 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.00 (d, *J* = 8.4 Hz, 1H), 7.90 (d, *J* = 7.2 Hz, 1H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.31 (t, *J* = 7.6 Hz, 1H), 7.17 (dd, *J* = 8.8, 2.2 Hz, 1H), 7.08 (d, *J* = 8.7 Hz, 1H), 6.63 (s, 1H), 4.71 (s, 2H), 2.90 (s, 3H), 2.59 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 206.2, 143.8, 137.8 (d, *J* = 9.0 Hz), 135.8, 134.8, 130.2, 124.0, 121.7, 119.8, 119.1, 118.9, 117.5, 113.8, 110.6, 110.0, 109.8, 51.0, 30.2, 21.1. HRMS (EI, m/z) calcd for $C_{19}H_{15}N_2OF$ $[M+H]^+$: 307.1241; found: 307.1242.

1-(3-chloro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6k)



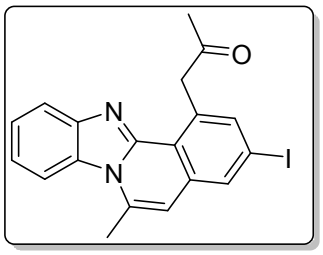
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (57.3 mg, 89%). mp. 182 °C (lit. 182-183 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.99 (d, *J* = 8.4 Hz, 1H), 7.90 (d, *J* = 6.2 Hz, 1H), 7.46 (dd, *J* = 14.1, 4.7 Hz, 2H), 7.31 (dd, *J* = 15.3, 7.5 Hz, 2H), 6.58 (s, 1H), 4.67 (s, 2H), 2.88 (s, 3H), 2.58 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.2, 147.1, 143.8, 136.4, 135.8, 134.7, 134.0, 130.7, 130.2, 124.2, 124.2, 124.0, 121.9, 119.8, 119.2, 113.8, 110.2, 50.8, 30.1, 21.1. HRMS (EI, m/z) calcd for $C_{19}H_{15}N_2OCl$ $[M+H]^+$: 323.0946; found: 323.0948.

1-(3-bromo-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6l)



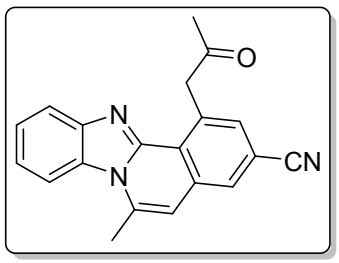
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a yellow solid (70.3 mg, 96%).mp. 183 °C (lit. 183-184 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.04 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 5.9 Hz, 1H), 7.69 (s, 1H), 7.51 – 7.42 (m, 2H), 7.34 (t, *J* = 7.6 Hz, 1H), 6.63 (s, 1H), 4.68 (s, 2H), 2.95 (s, 3H), 2.58 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.2, 147.2, 143.8, 136.4, 135.9, 134.3, 133.4, 130.2, 127.4, 124.1, 122.0, 119.9, 113.9, 110.2, 50.8, 30.1, 21.2. HRMS (EI, m/z) calcd for $C_{19}H_{15}N_2OBr$ $[M+H]^+$: 367.0441; found: 367.0442.

1-(3-iodo-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6m)



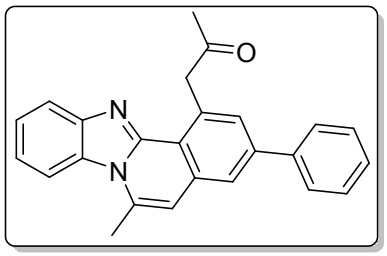
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a white solid (79.5 mg, 96%).mp. 195 °C (lit. 195-196 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.01 (d, *J* = 8.4 Hz, 1H), 7.90 (d, *J* = 8.7 Hz, 2H), 7.62 (s, 1H), 7.46 (t, *J* = 7.6 Hz, 1H), 7.32 (t, *J* = 7.7 Hz, 1H), 6.56 (s, 1H), 4.64 (s, 2H), 2.90 (s, 3H), 2.58 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.2, 147.3, 143.9, 138.9, 136.2, 135.7, 134.3, 133.7, 130.3, 124.1, 122.0, 120.2, 120.0, 113.9, 109.9, 95.6, 50.6, 30.2, 21.2. HRMS (EI, m/z) calcd for $C_{19}H_{15}N_2OI$ $[M+H]^+$: 415.0302; found: 415.0305.

6-methyl-1-(2-oxopropyl)benzo[4,5]imidazo[2,1-a]isoquinoline-3-carbonitrile (6n)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 1:1, v/v) to give the product as a yellow solid (58.2 mg, 93%). mp. 255 °C (lit. 255-256 °C). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.09 (d, *J* = 8.4 Hz, 1H), 7.95 (d, *J* = 8.1 Hz, 1H), 7.87 (d, *J* = 1.2 Hz, 1H), 7.52 (t, *J* = 7.4 Hz, 2H), 7.41 (dd, *J* = 11.5, 4.1 Hz, 1H), 6.75 (s, 1H), 4.76 (s, 2H), 3.00 (s, 3H), 2.62 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 205.7, 146.3, 143.9, 136.7, 135.9, 132.7, 132.0, 130.2, 129.4, 124.6, 123.6, 122.9, 120.4, 118.4, 114.0, 112.3, 110.1, 50.9, 30.2, 21.2. HRMS (EI, m/z) calcd for $C_{17}H_{16}N_2O$ $[M+H]^+$: 314.1288; found: 314.1290.

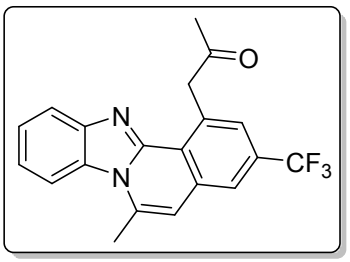
1-(6-methyl-3-phenylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6o)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 10:1, v/v) to give the product as a white solid (64.1 mg, 88%). mp. 199 °C (lit. 199-200 °C).^{1}H NMR

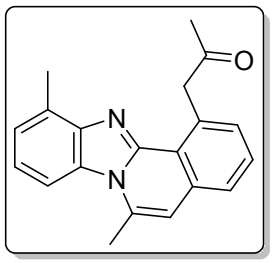
(500 MHz, $CDCl_3$) δ 8.08 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.0 Hz, 1H), 7.77 (s, 1H), 7.72 (d, *J* = 7.4 Hz, 2H), 7.59 (s, 1H), 7.51 – 7.45 (m, 3H), 7.41 (t, *J* = 7.3 Hz, 1H), 7.34 (t, *J* = 7.6 Hz, 1H), 6.81 (s, 1H), 4.82 (s, 2H), 3.00 (s, 3H), 2.61 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 207.0, 147.8, 144.1, 141.7, 140.1, 135.0, 134.9, 133.5, 130.5, 129.9, 128.9, 127.8, 127.4, 123.8, 123.3, 121.7, 119.9, 113.8, 111.5, 51.3, 30.2, 21.3. HRMS (EI, m/z) calcd for $C_{25}H_{20}N_2O$ $[M+H]^+$: 365.1648; found: 365.1650.

1-(6-methyl-3-(trifluoromethyl)benzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6p)



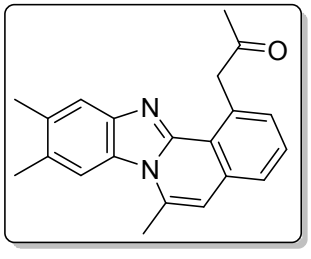
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 2:1, v/v) to give the product as a yellow solid (48.4 mg, 68%). mp. 155 °C (lit. 155-157 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.03 (d, *J* = 8.4 Hz, 1H), 7.93 (d, *J* = 8.1 Hz, 1H), 7.79 (s, 1H), 7.53 (s, 1H), 7.48 (t, *J* = 7.6 Hz, 1H), 7.34 (t, *J* = 7.7 Hz, 1H), 6.72 (s, 1H), 4.75 (s, 2H), 2.91 (s, 3H), 2.61 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 206.0, 146.7, 143.8, 136.1, 135.6, 132.7, 130.6, 130.2 (d, *J* = 16.7 Hz), 126.4 (d, *J* = 3.0 Hz), 125.0, 124.2, 123.0, 122.4, 122.2 (dd, *J* = 8.0, 4.0 Hz), 120.2, 114.0, 110.8, 51.1, 30.2, 21.1. HRMS (EI, m/z) calcd for $C_{20}H_{15}N_2OF_3$ $[M+H]^+$: 357.1209; found: 357.1210.

1-(6,11-dimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6q)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a white solid (38.7 mg, 64%). mp. 112 °C (lit. 112-114 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.89 (d, *J* = 8.2 Hz, 1H), 7.55 (d, *J* = 4.6 Hz, 2H), 7.33 (t, *J* = 4.2 Hz, 1H), 7.27 (d, *J* = 7.1 Hz, 1H), 7.25 – 7.20 (m, 1H), 6.71 (s, 1H), 4.82 (s, 2H), 2.93 (s, 3H), 2.81 (s, 3H), 2.51 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.7, 147.0, 143.2, 134.6, 134.1, 132.9, 130.6, 129.9, 129.6, 128.9, 125.1, 123.9, 121.5, 121.1, 111.3, 51.1, 30.2, 21.1, 17.6. HRMS (EI, m/z) calcd for $C_{20}H_{18}N_2O$ $[M+H]^+$: 303.1492; found: 303.1493.

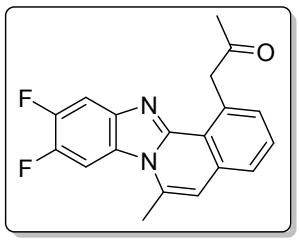
1-(6,9,10-trimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6r)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 2:1, v/v) to give the product as a white solid (60.7 mg, 96%). mp. 162 °C (lit. 162-163 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.72 (s, 1H), 7.66 (s, 1H), 7.54 – 7.48 (m, 2H), 7.32 (dd, *J* = 6.3, 1.9 Hz, 1H), 6.62 (s, 1H), 4.74 (s, 2H), 2.86 (d, *J* = 0.4 Hz, 3H), 2.58 (s, 3H), 2.42 (s, 3H), 2.39 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ

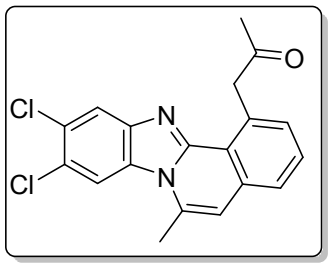
207.2, 147.2, 142.6, 134.6, 134.1, 132.8, 130.7, 130.5, 128.8, 128.6, 125.0, 120.9, 119.6, 113.8, 110.7, 51.1, 30.2, 21.0, 20.8, 20.3. HRMS (EI, m/z) calcd for $C_{21}H_{20}N_2O$ $[M+H]^+$: 317.1648; found: 317.1649.

1-(9,10-difluoro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6s)



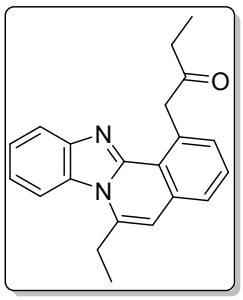
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (54.4 mg, 84%). mp. 181 °C (lit. 181-182 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 (dd, *J* = 10.9, 7.1 Hz, 1H), 7.64 – 7.55 (m, 3H), 7.35 (d, *J* = 6.3 Hz, 1H), 6.75 (s, 1H), 4.70 (s, 2H), 2.88 (s, 3H), 2.54 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.8, 134.3, 133.7, 132.8, 131.0, 129.5, 125.3, 125.3, 125.1, 125.1, 112.0, 106.5, 106.3, 102.3 – 102.1 (m), 102.1, 101.9, 51.0, 30.1, 20.7. HRMS (EI, m/z) calcd for $C_{19}H_{14}N_2OF_2$ $[M+H]^+$: 325.1147; found: 325.1147.

1-(9,10-dichloro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6t)



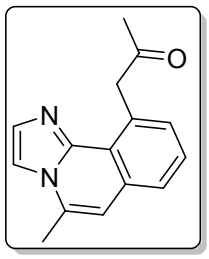
The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 8:1, v/v) to give the product as a yellow solid (69.8 mg, 98%). mp. 184 °C (lit. 184-185 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.05 (s, 1H), 7.92 (s, 1H), 7.58 (t, *J* = 7.5 Hz, 1H), 7.53 (d, *J* = 7.7 Hz, 1H), 7.35 (d, *J* = 7.0 Hz, 1H), 6.72 (s, 1H), 4.66 (s, 2H), 2.83 (s, 3H), 2.54 (s, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 206.6, 149.3, 143.1, 134.6, 133.9, 133.1, 131.2, 129.8, 129.1, 127.9, 125.3, 125.1, 120.5, 120.5, 114.9, 112.2, 51.1, 30.1, 20.8. HRMS (EI, m/z) calcd for $C_{19}H_{14}N_2OCl_2$ $[M+H]^+$: 357.0556; found: 357.0557.

1-(6-ethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (6u)



The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 6:1, v/v) to give the product as a white solid (50.6 mg, 80%). mp. 91 °C (lit. 91-92 °C).^{1}H NMR (500 MHz, $CDCl_3$) δ 8.02 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 8.1 Hz, 1H), 7.63 – 7.54 (m, 2H), 7.46 (t, *J* = 7.6 Hz, 1H), 7.34 (ddd, *J* = 9.4, 6.9, 1.7 Hz, 2H), 6.78 (s, 1H), 4.74 (s, 2H), 3.34 (q, *J* = 7.3 Hz, 2H), 2.99 (q, *J* = 7.3 Hz, 2H), 1.52 (t, *J* = 7.3 Hz, 3H), 1.20 (t, *J* = 7.4 Hz, 3H).^{13}C NMR (126 MHz, $CDCl_3$) δ 209.6, 148.0, 144.0, 140.1, 134.6, 133.0, 130.9, 129.8, 129.1, 125.3, 123.7, 121.7, 121.0, 119.8, 114.3, 109.1, 50.2, 35.9, 26.4, 12.0, 8.1. HRMS (EI, m/z) calcd for $C_{21}H_{20}N_2O$ $[M+H]^+$: 317.1648; found: 317.1649.

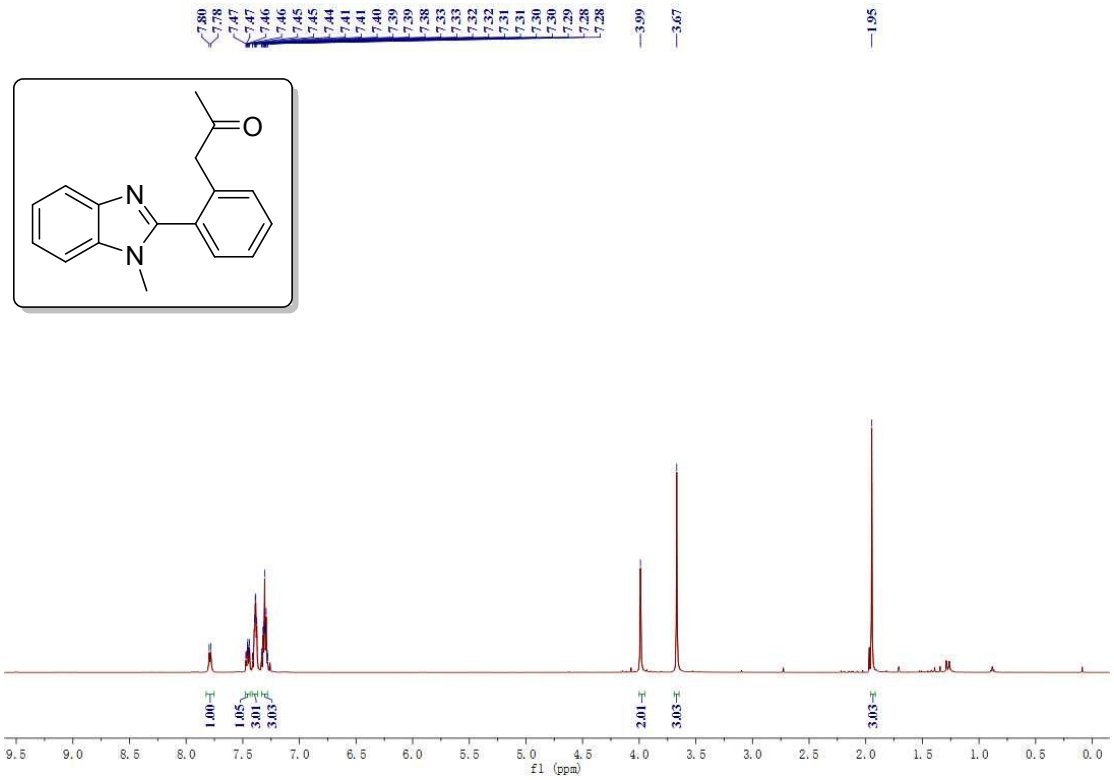
1-(5-methylimidazo[2,1-a]isoquinolin-10-yl)propan-2-one (6v)

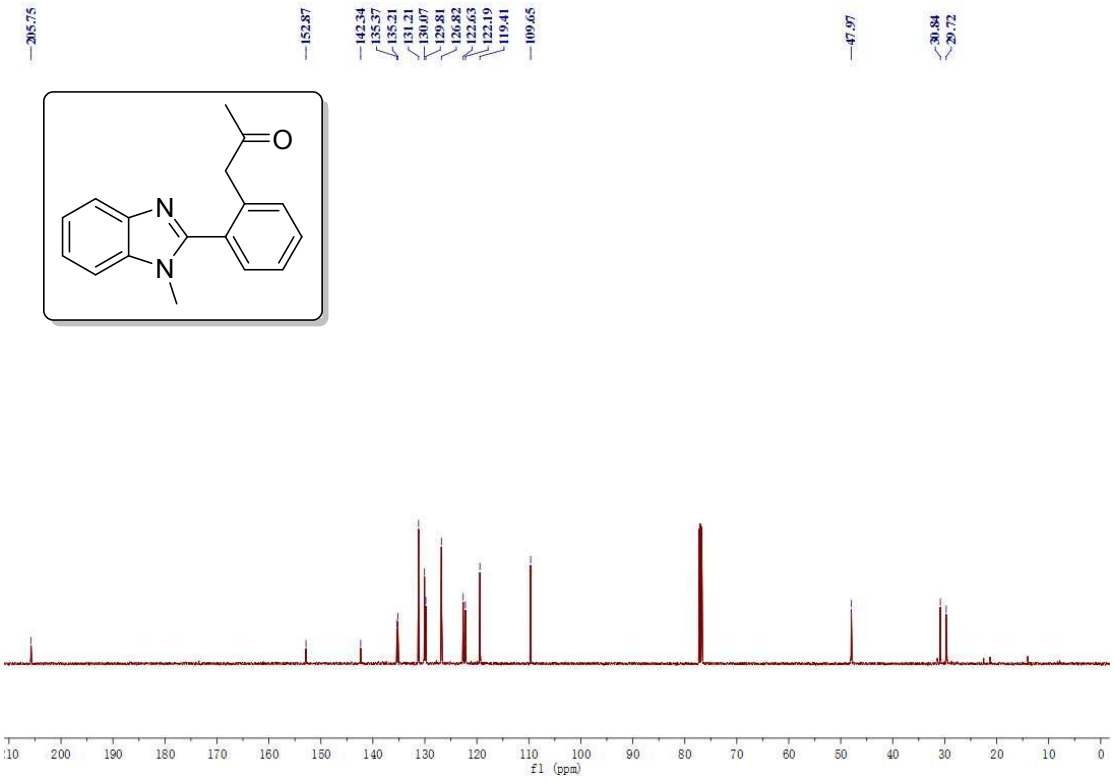


The reaction was performed by following the general procedure. The residue was purified by flash column chromatograph (silica gel, petroleum ether:EtOAc = 2:1, v/v) to give the product as a yellow solid (27.6 mg, 58%). mp. 125 °C (lit. 125-127 °C).^{1}H NMR (500 MHz, DMSO) δ 7.89 (d, *J* = 1.2 Hz, 1H), 7.70 (d, *J* = 7.4 Hz, 1H), 7.60 (d, *J* = 1.1 Hz, 1H), 7.51 (t, *J* = 7.6 Hz, 1H), 7.36 (d, *J* = 7.1 Hz, 1H), 7.13 (s, 1H), 4.65 (s, 2H), 2.62 (s, 3H), 2.31 (s, 3H).^{13}C NMR (126 MHz, DMSO) δ 205.9, 142.3, 132.4, 132.3, 130.9, 130.6, 127.3, 125.4, 120.9, 112.4, 111.5, 49.9, 30.0, 18.2. HRMS (EI, m/z) calcd for $C_{15}H_{14}N_2O$ $[M+H]^+$: 239.1179; found: 239.1180.

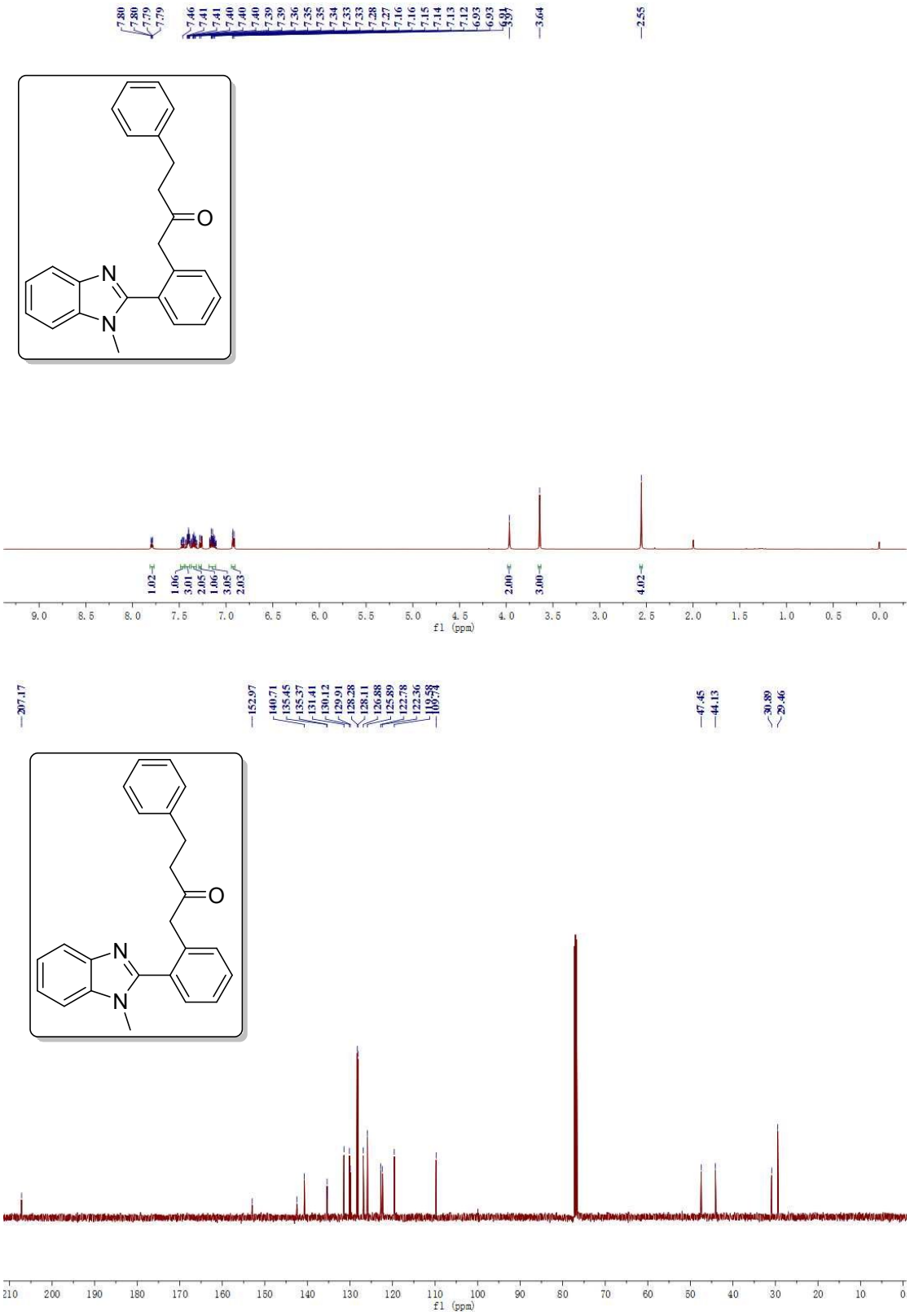
8. NMR spectroscopic data

1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3a)

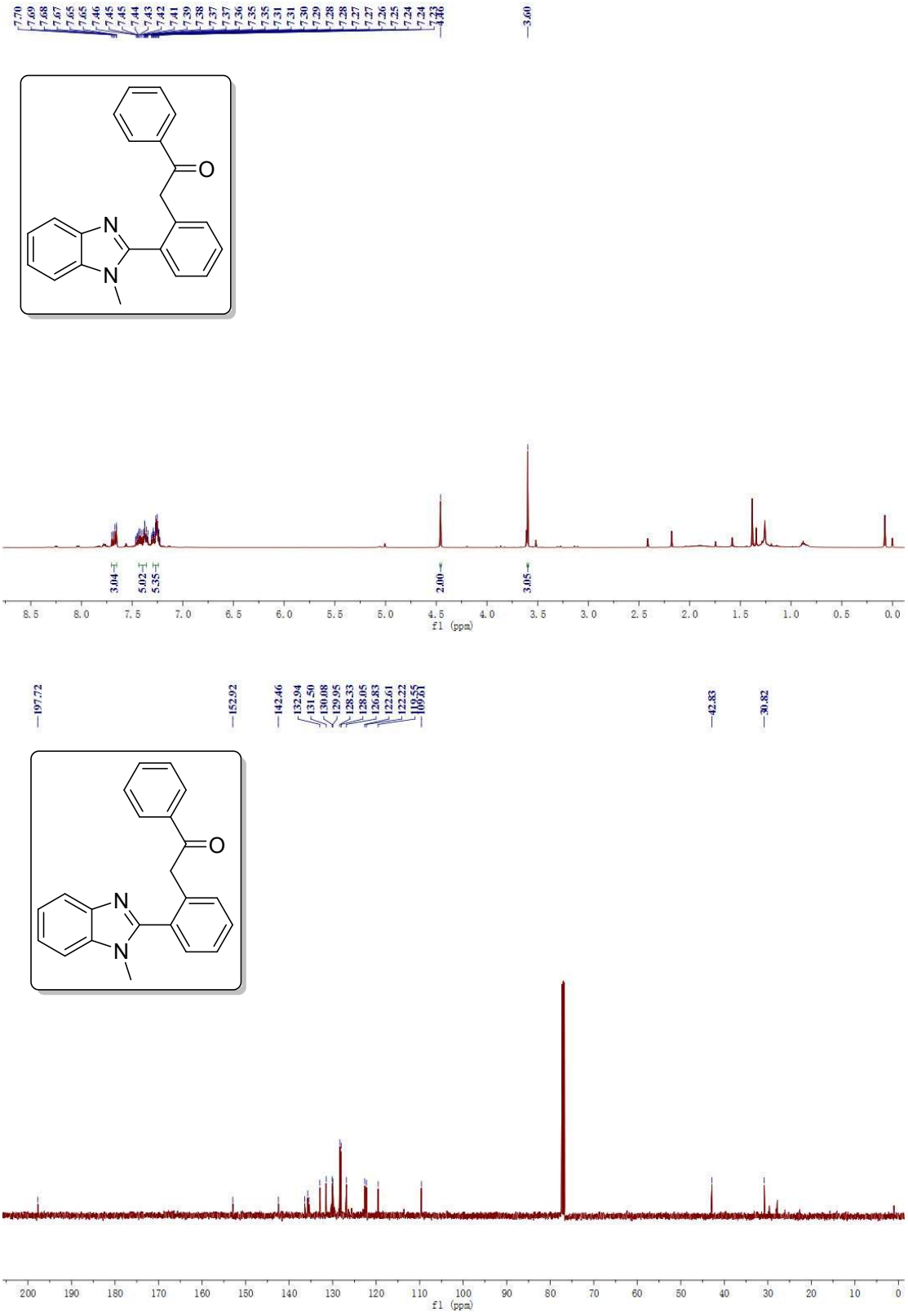




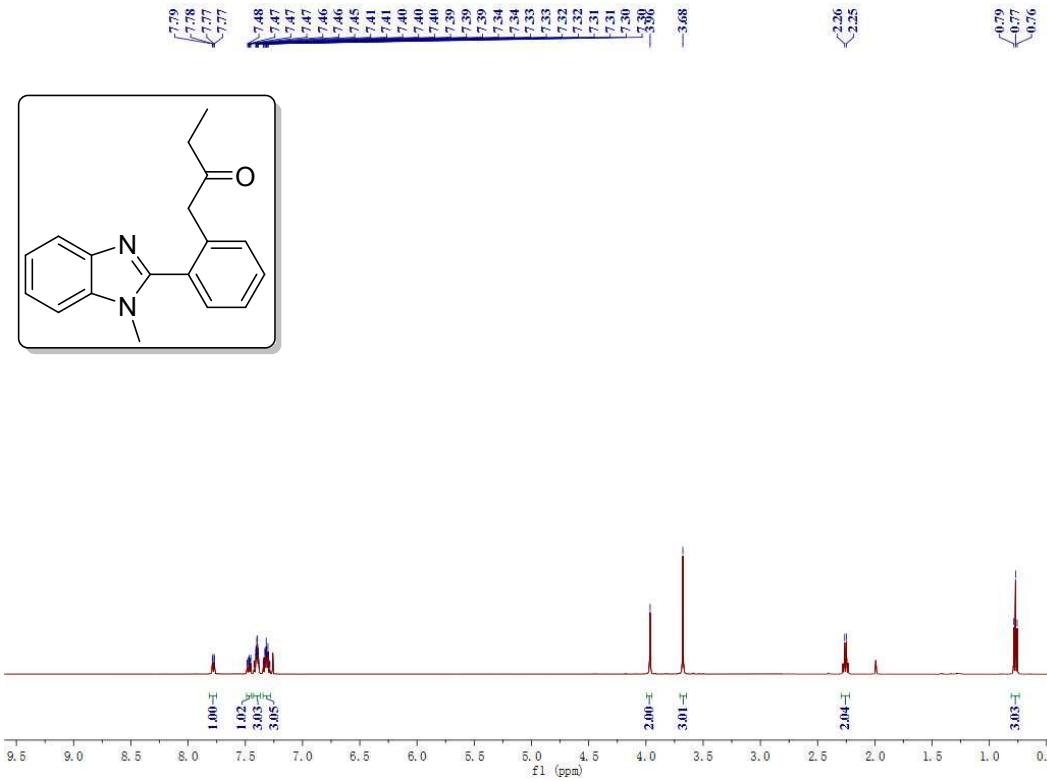
1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl) phenyl)-4-phenylbutan-2-one (3b)

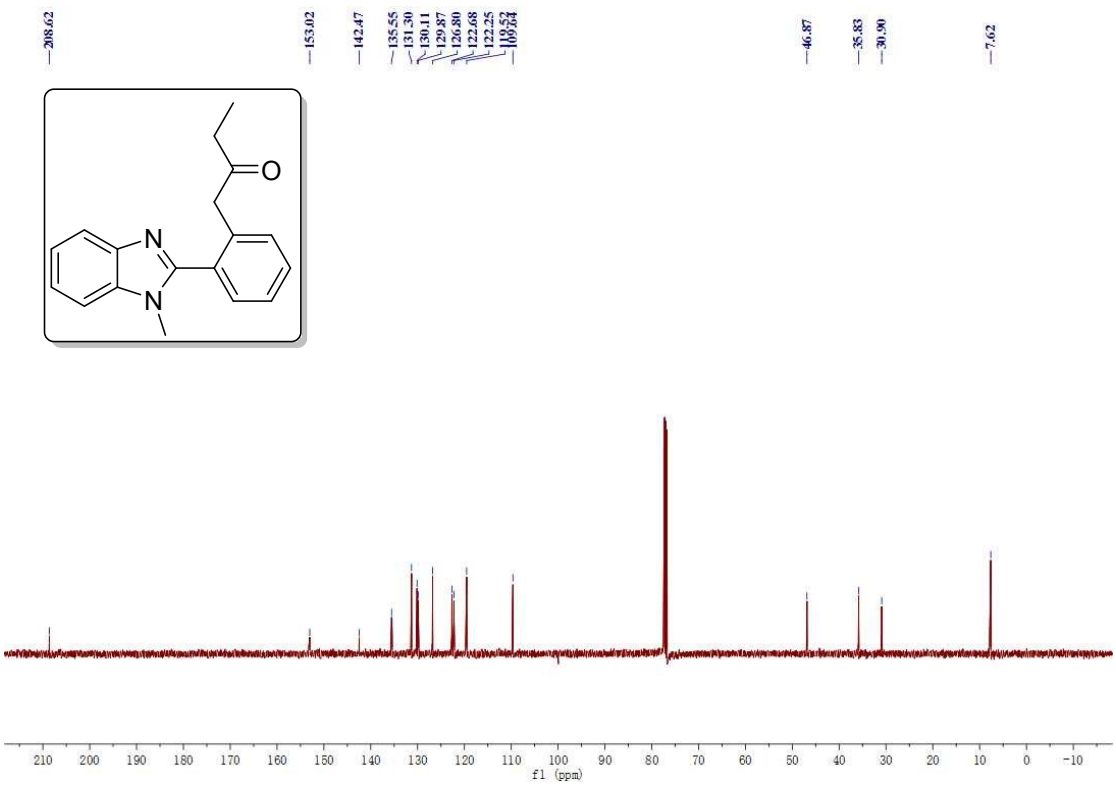


2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)-1-phenylethan-1-one (3c)

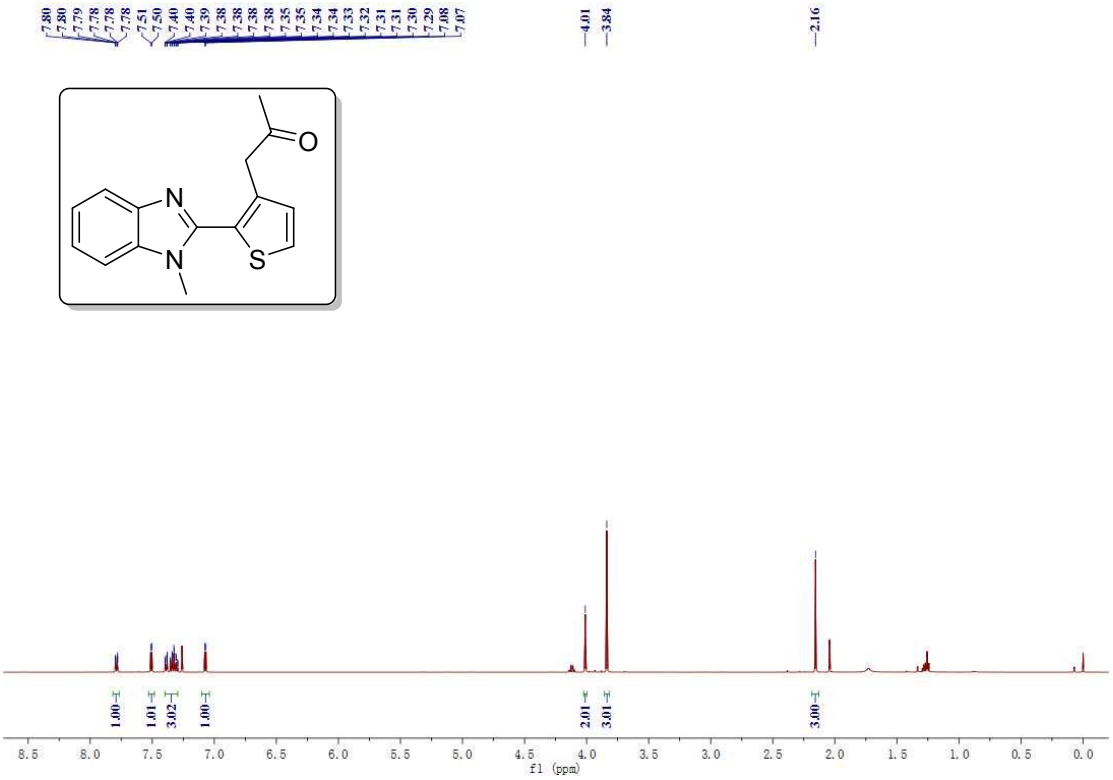


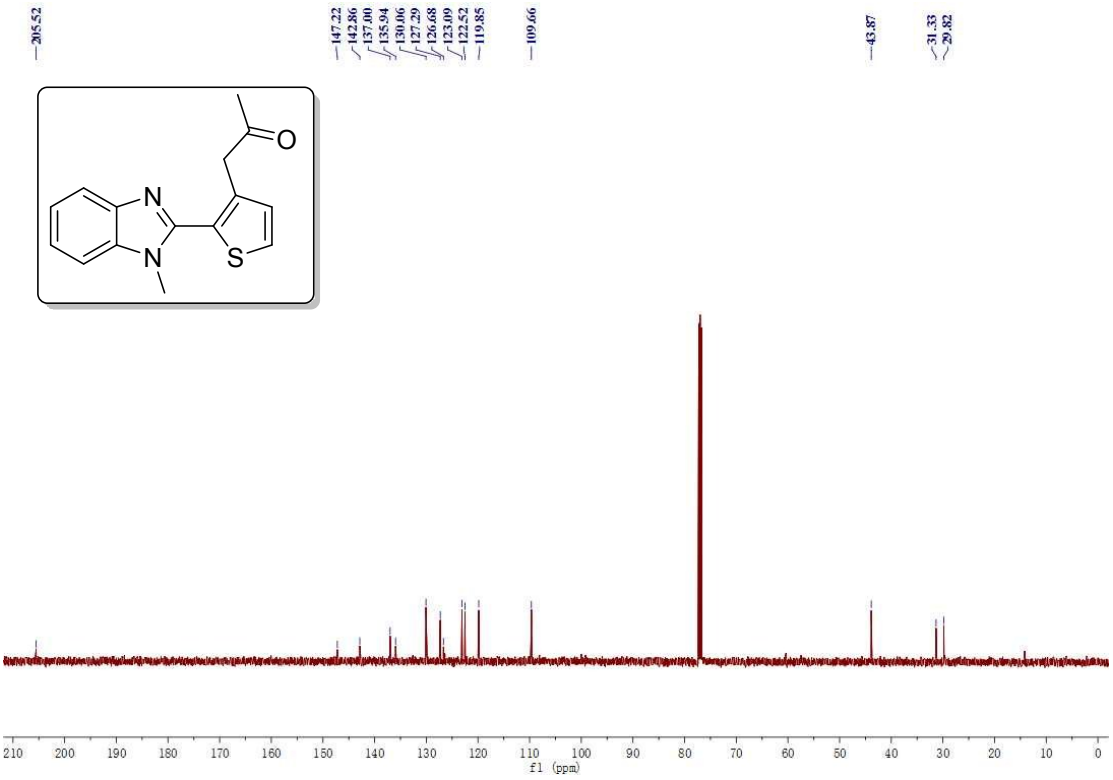
1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl) phenyl) butan-2-one (3d)



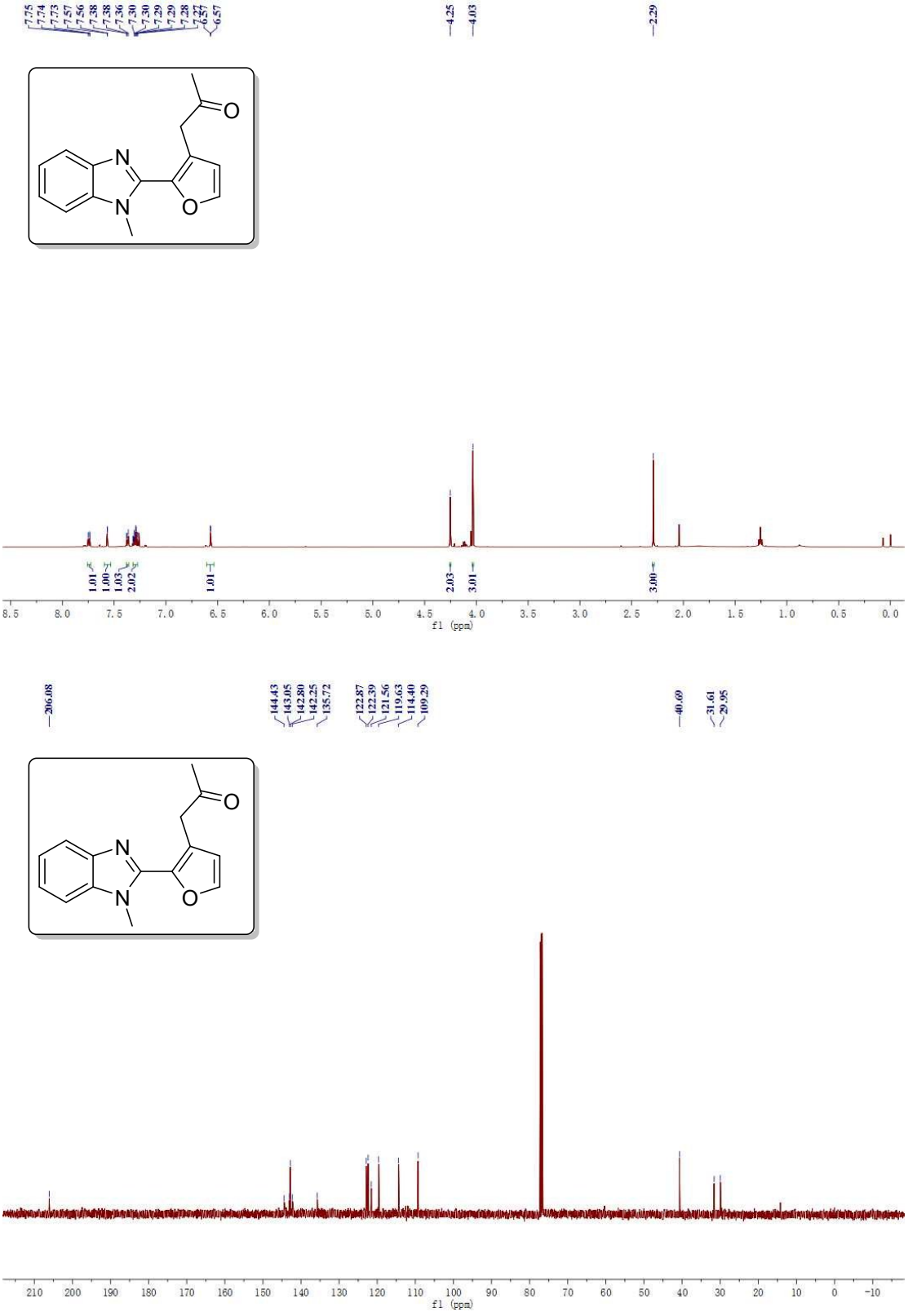


1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)thiophen-3-yl)propan-2-one (3e)

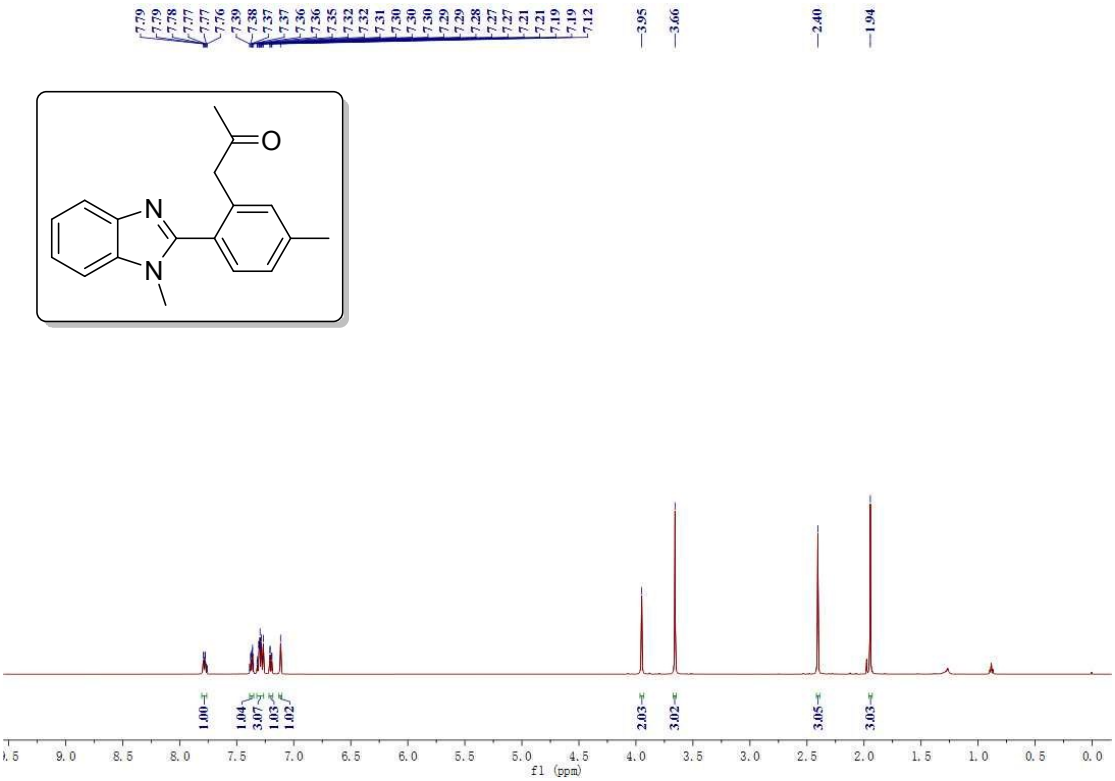




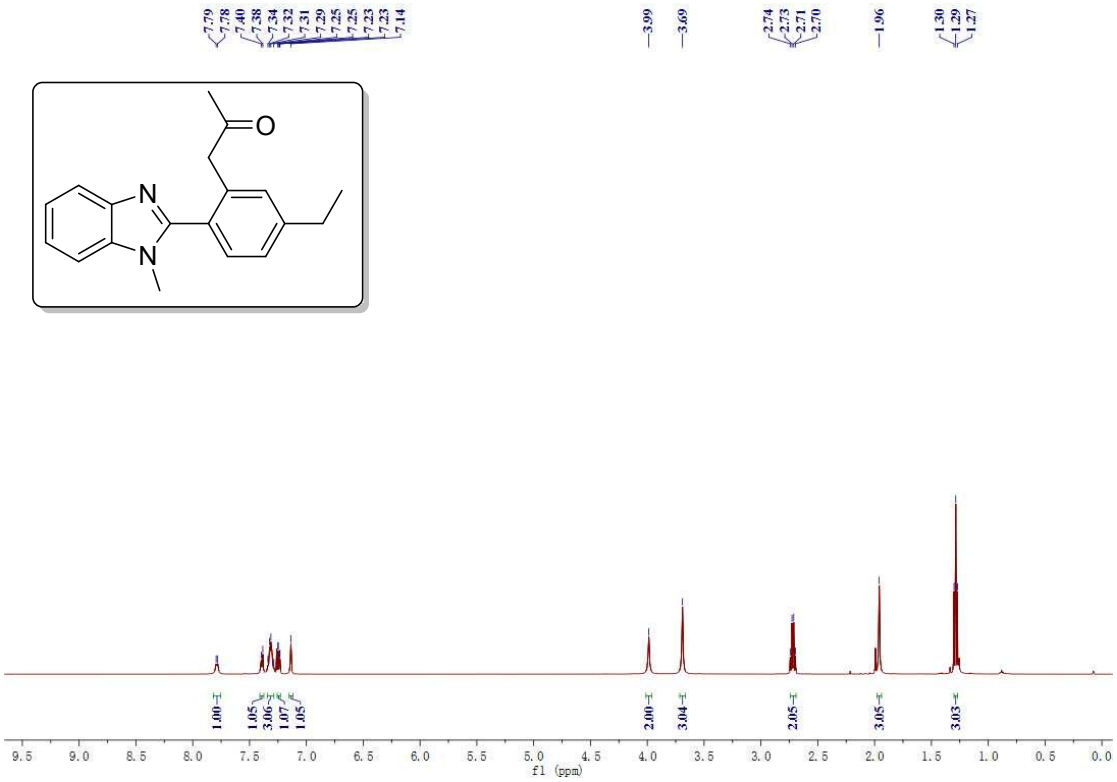
1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)furan-3-yl)propan-2-one (3f)

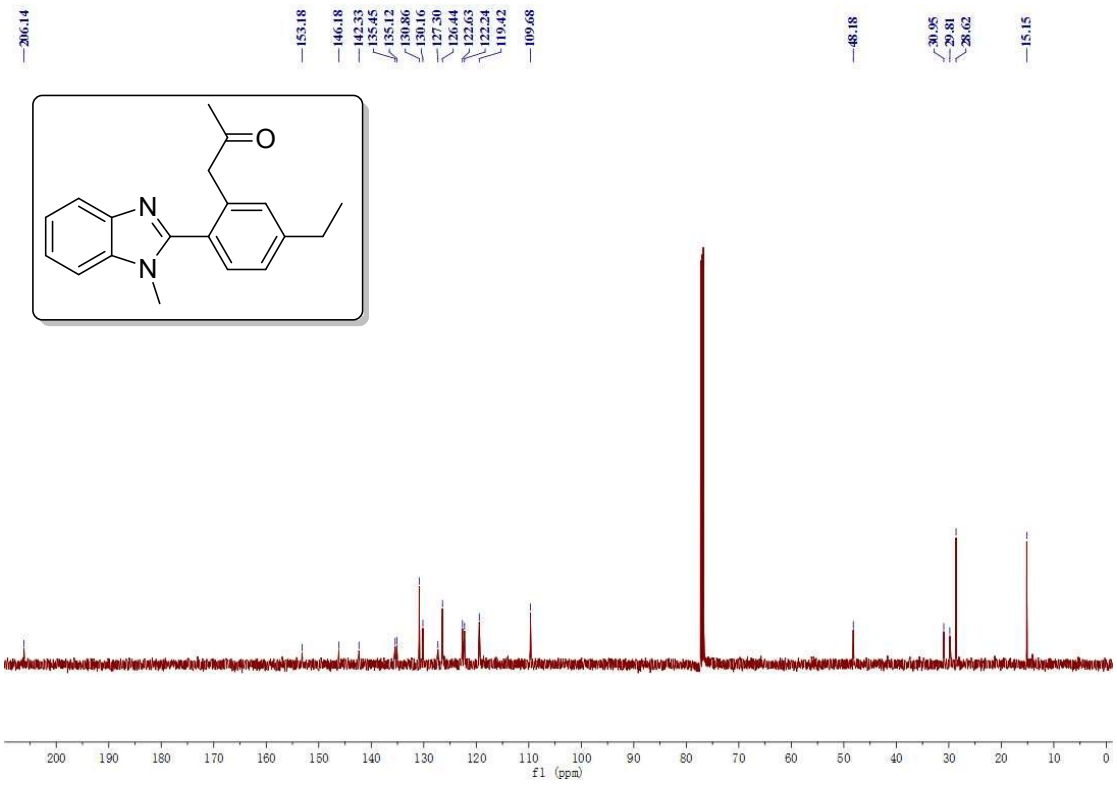


1-(5-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3g)

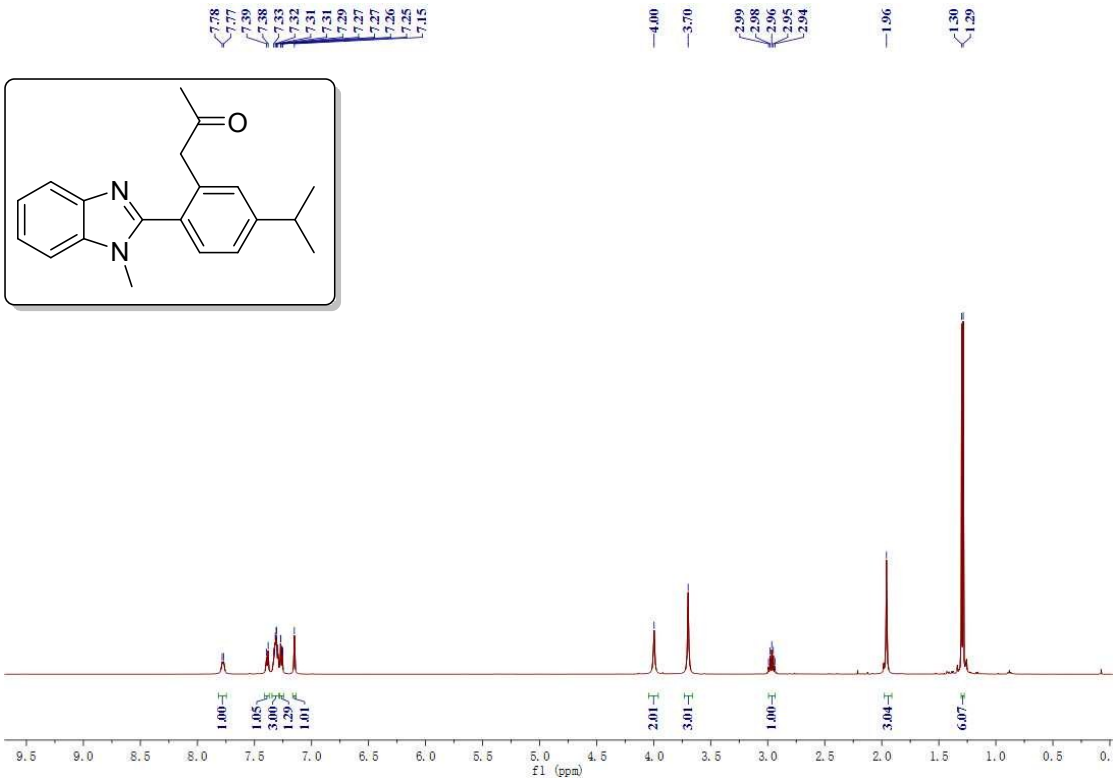


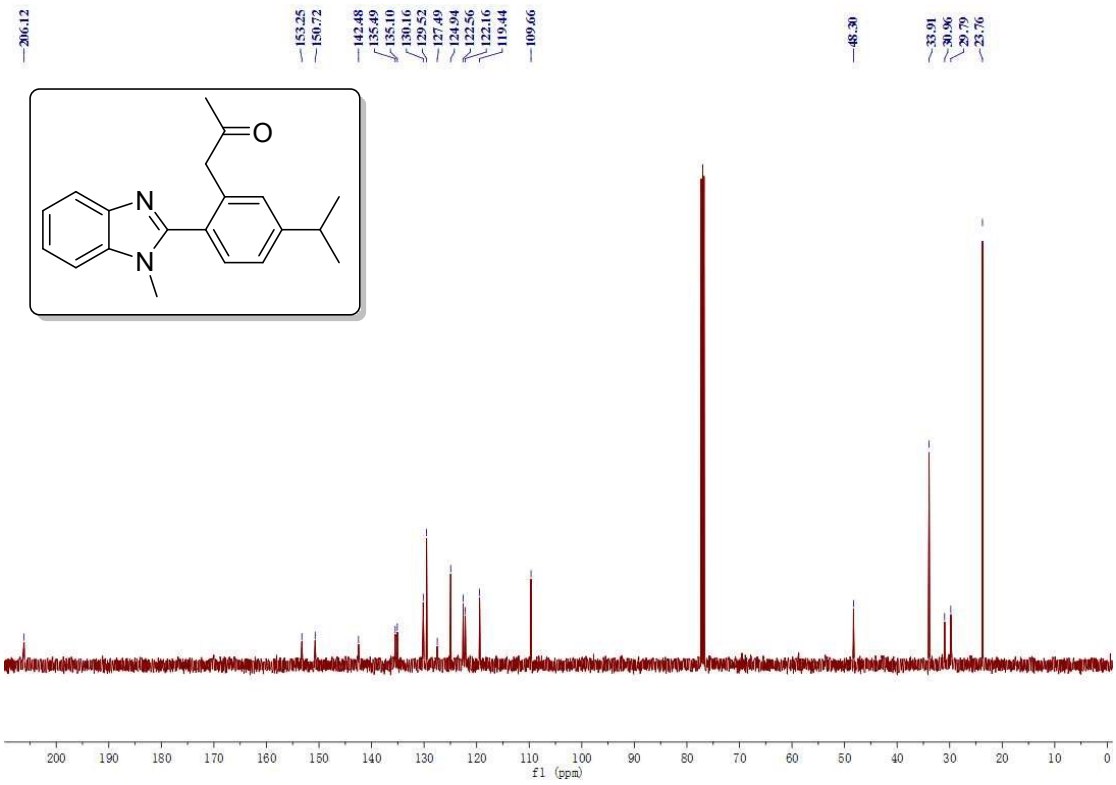
1-(5-ethyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3h)



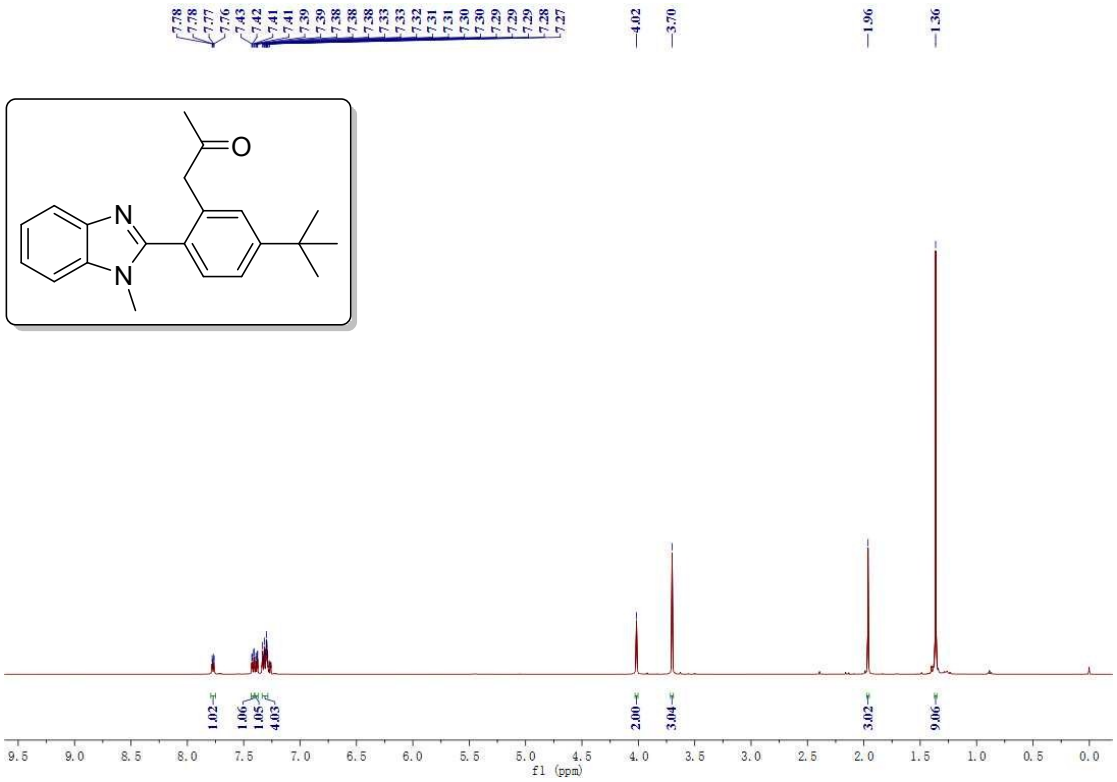


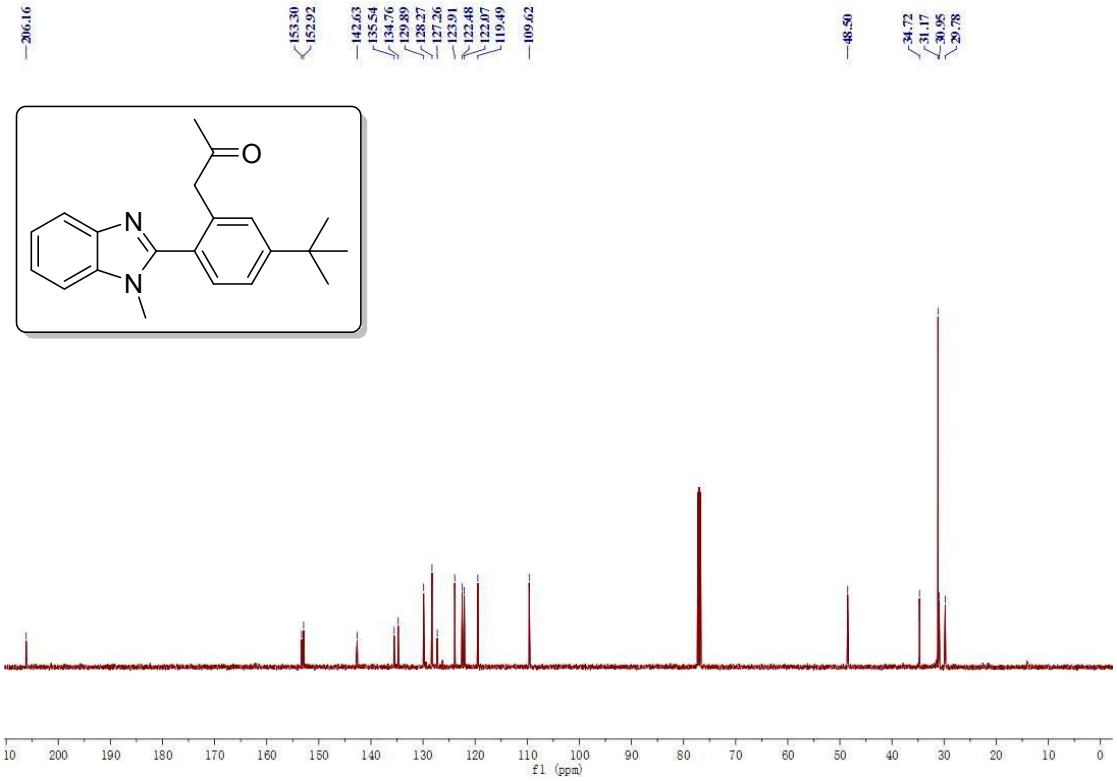
1-(5-isopropyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3i)



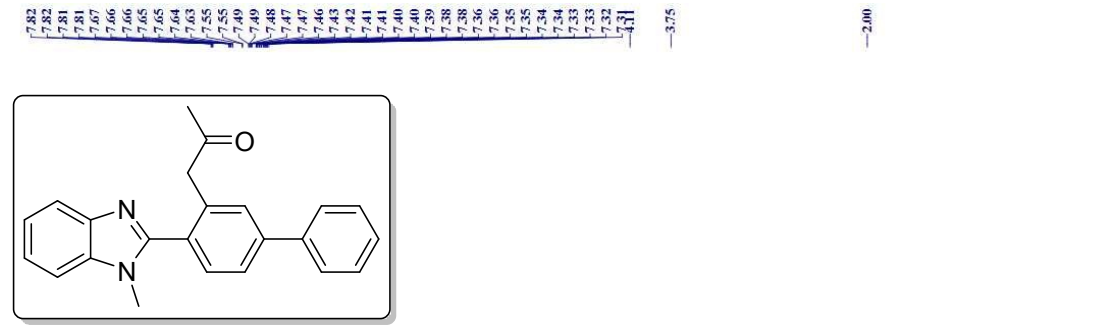


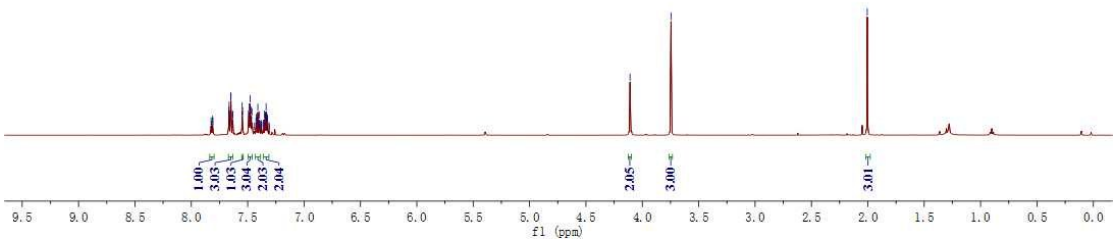
1-(5-(tert-butyl)-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3j)

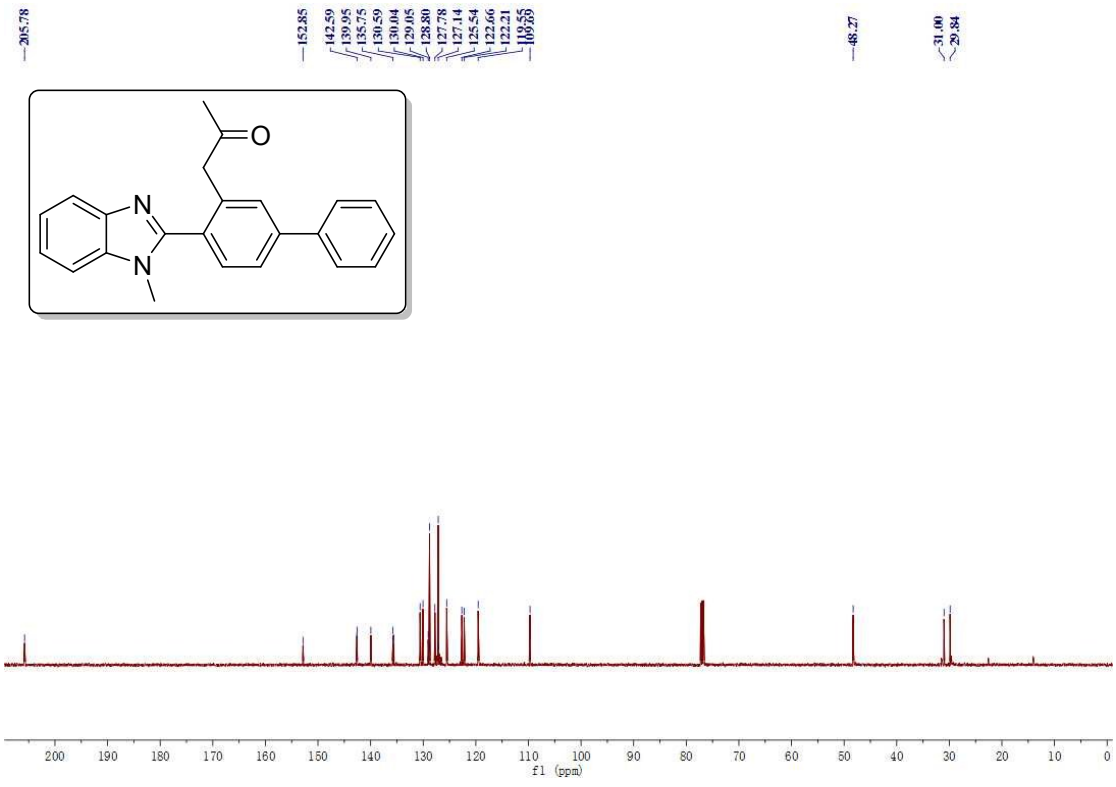




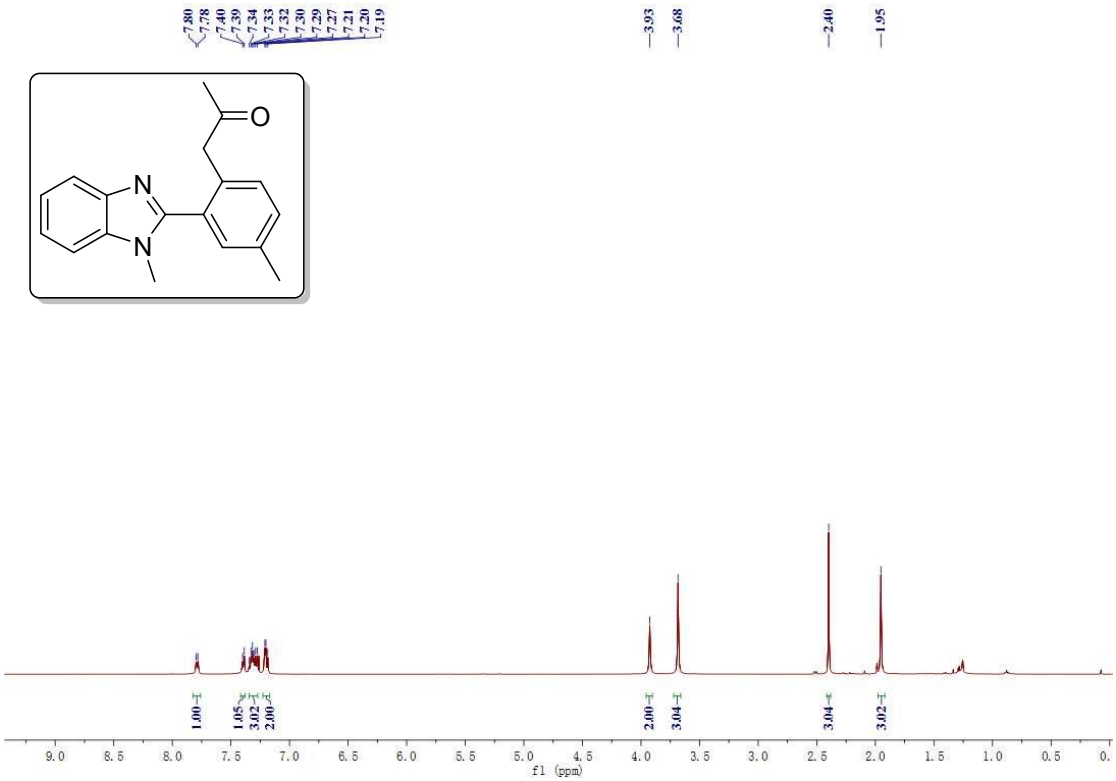
1-(4-(1-methyl-1H-benzo[d]imidazol-2-yl)-[1,1'-biphenyl]-3-yl)propan-2-one (3k)

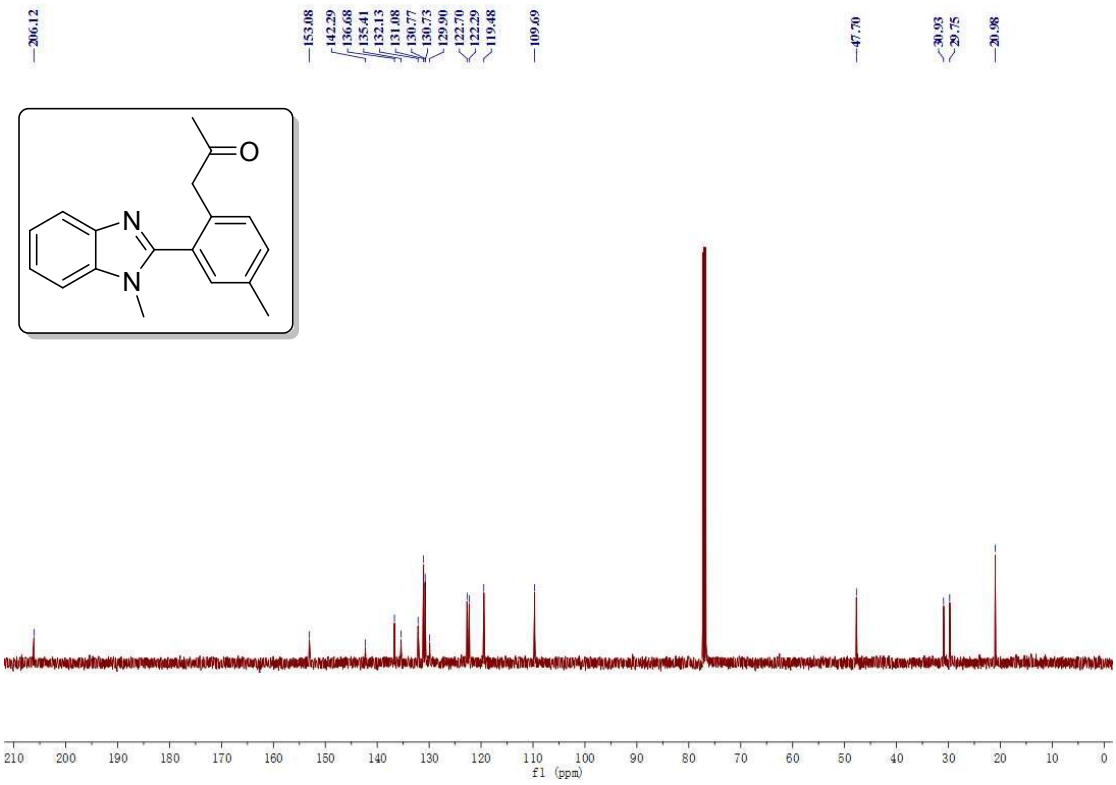




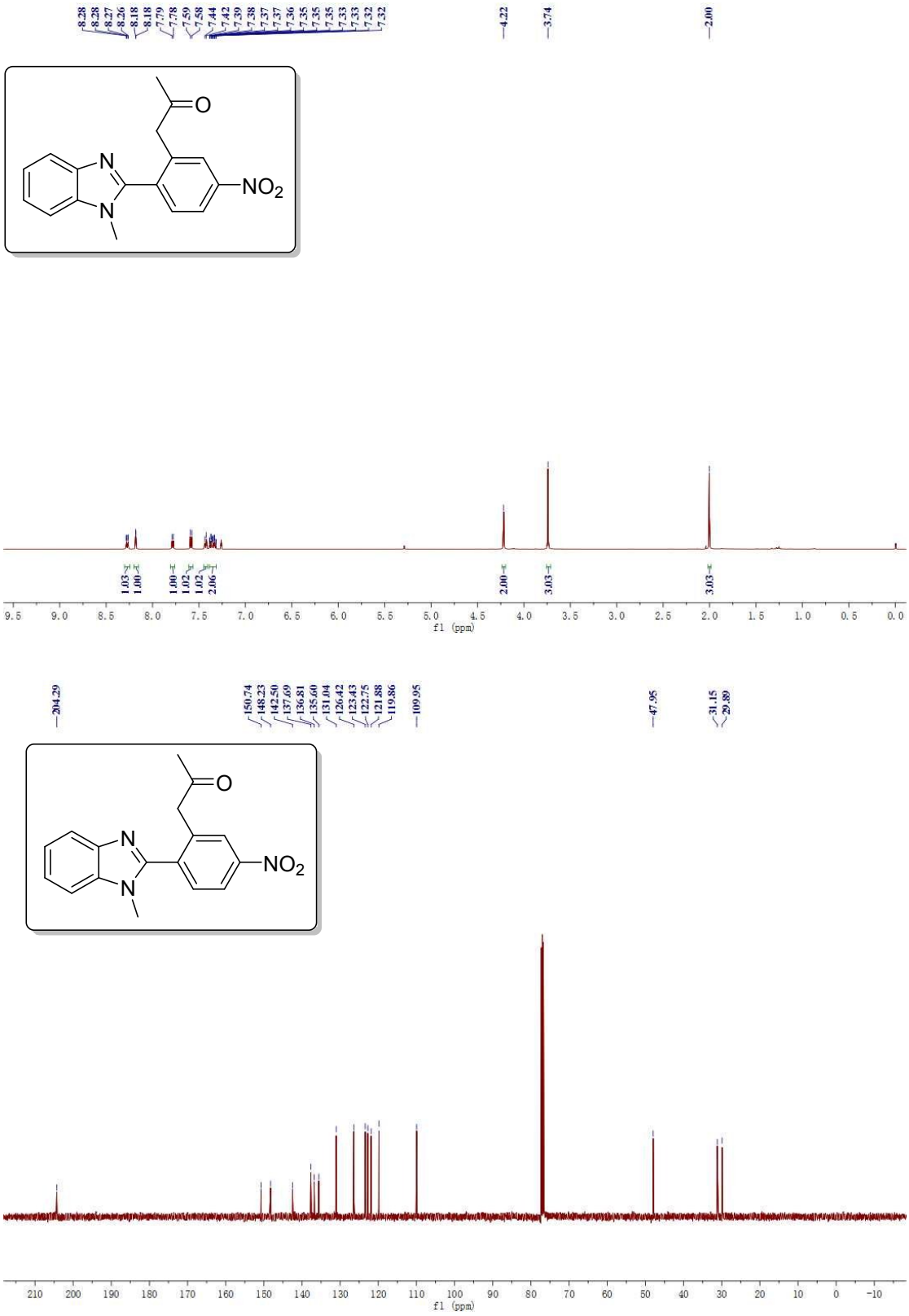


1-(4-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl) propan-2-one (3l)

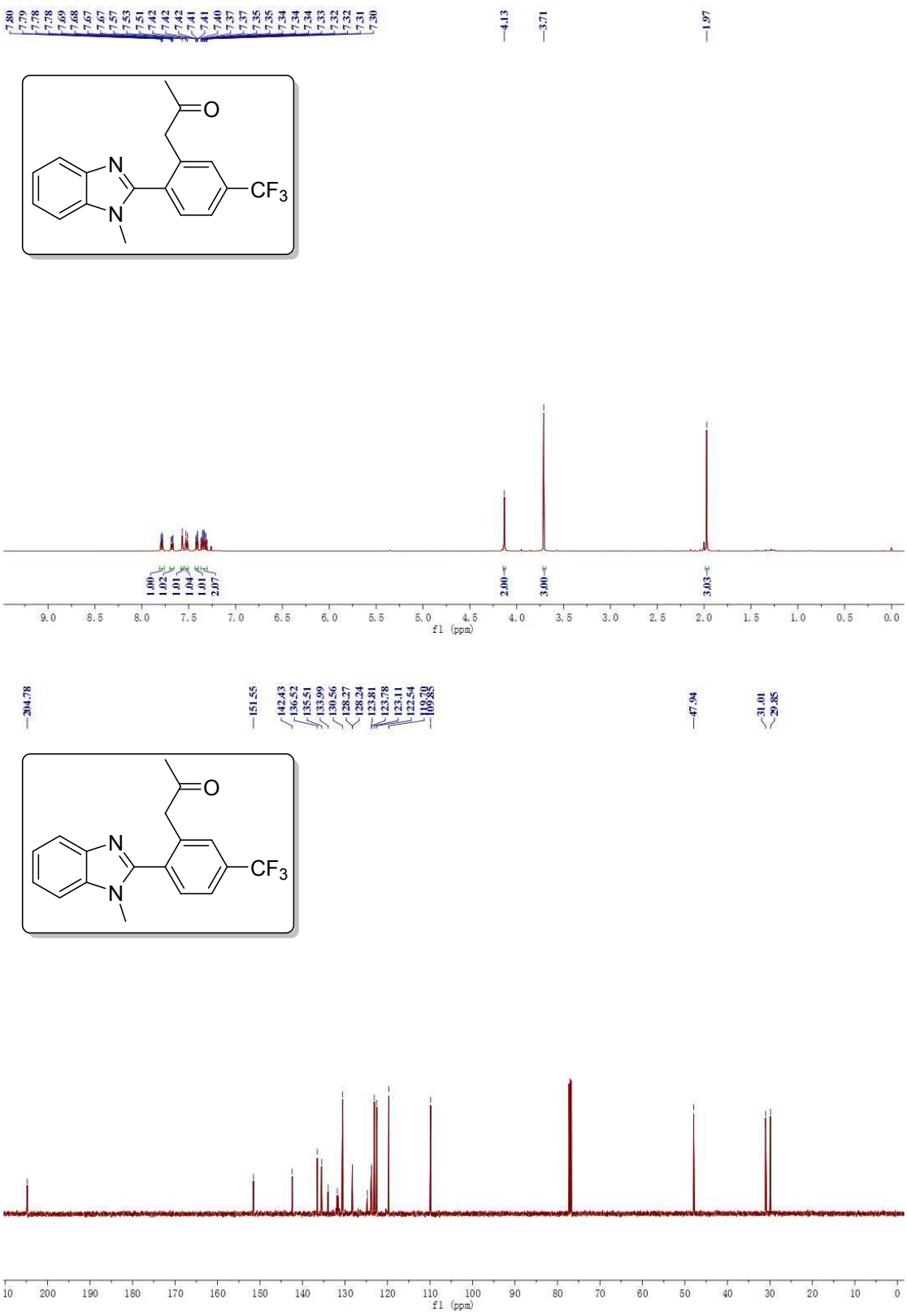




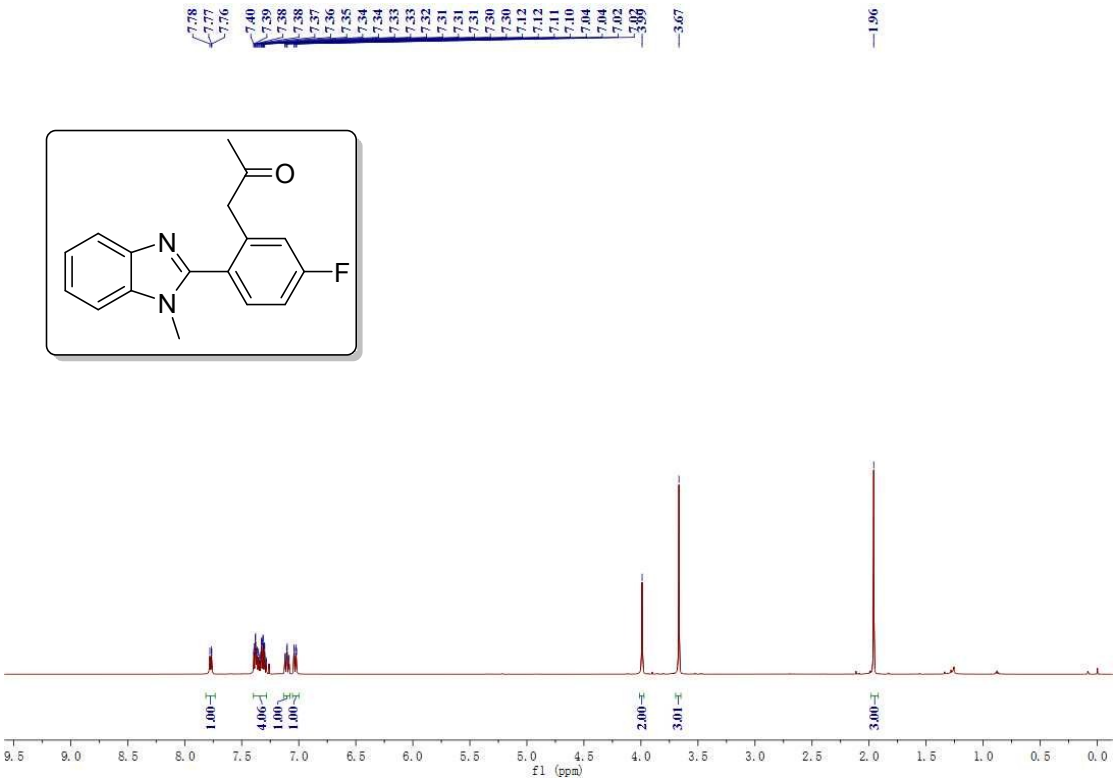
1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)-5-nitrophenyl)propan-2-one (3m)



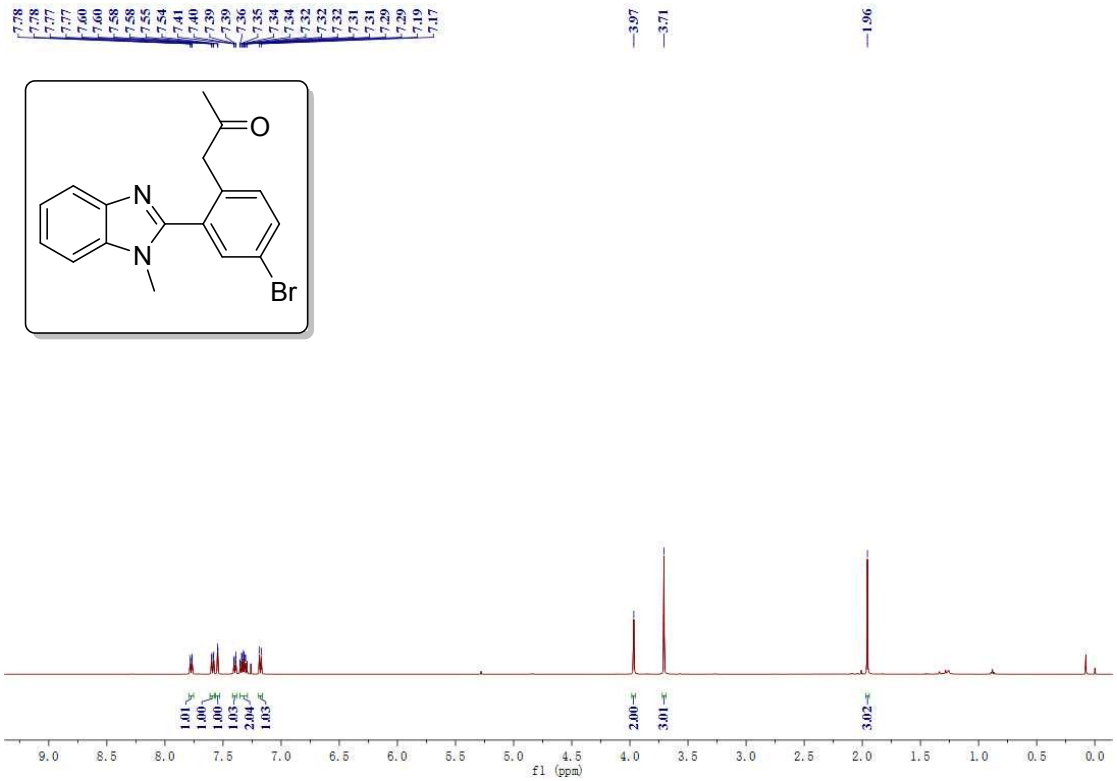
1-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)-5-(trifluoromethyl)phenyl)propan-2-one (3n)

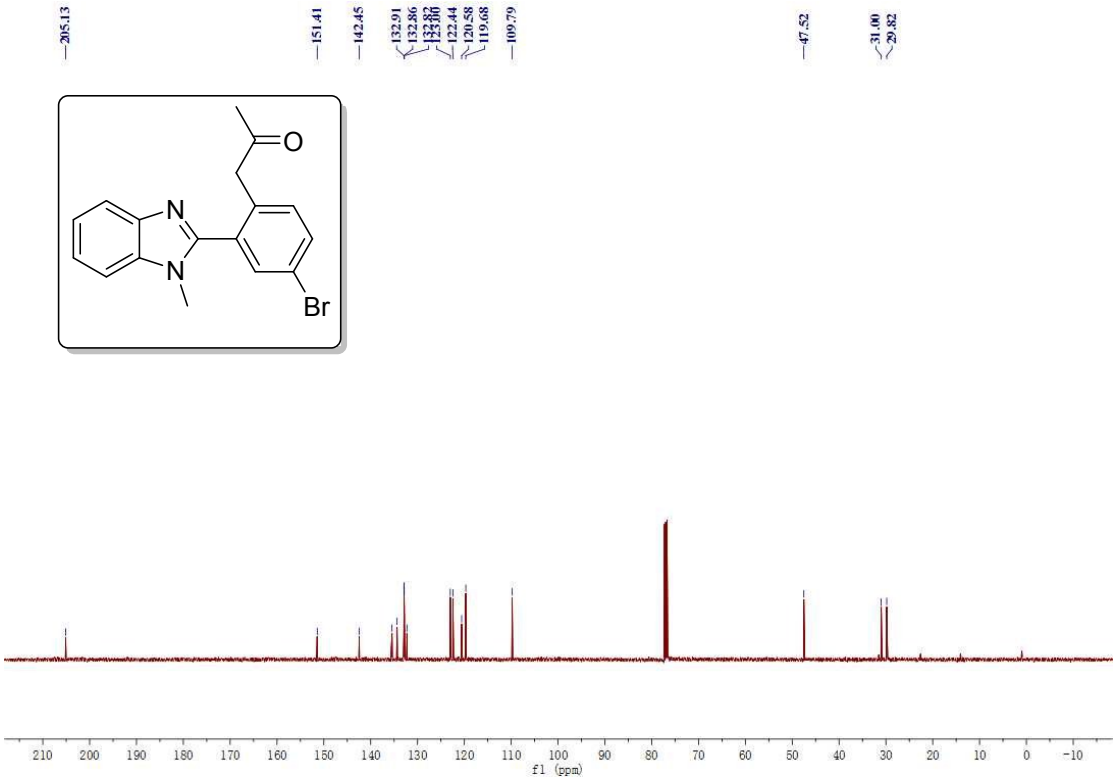


1-(5-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3o)

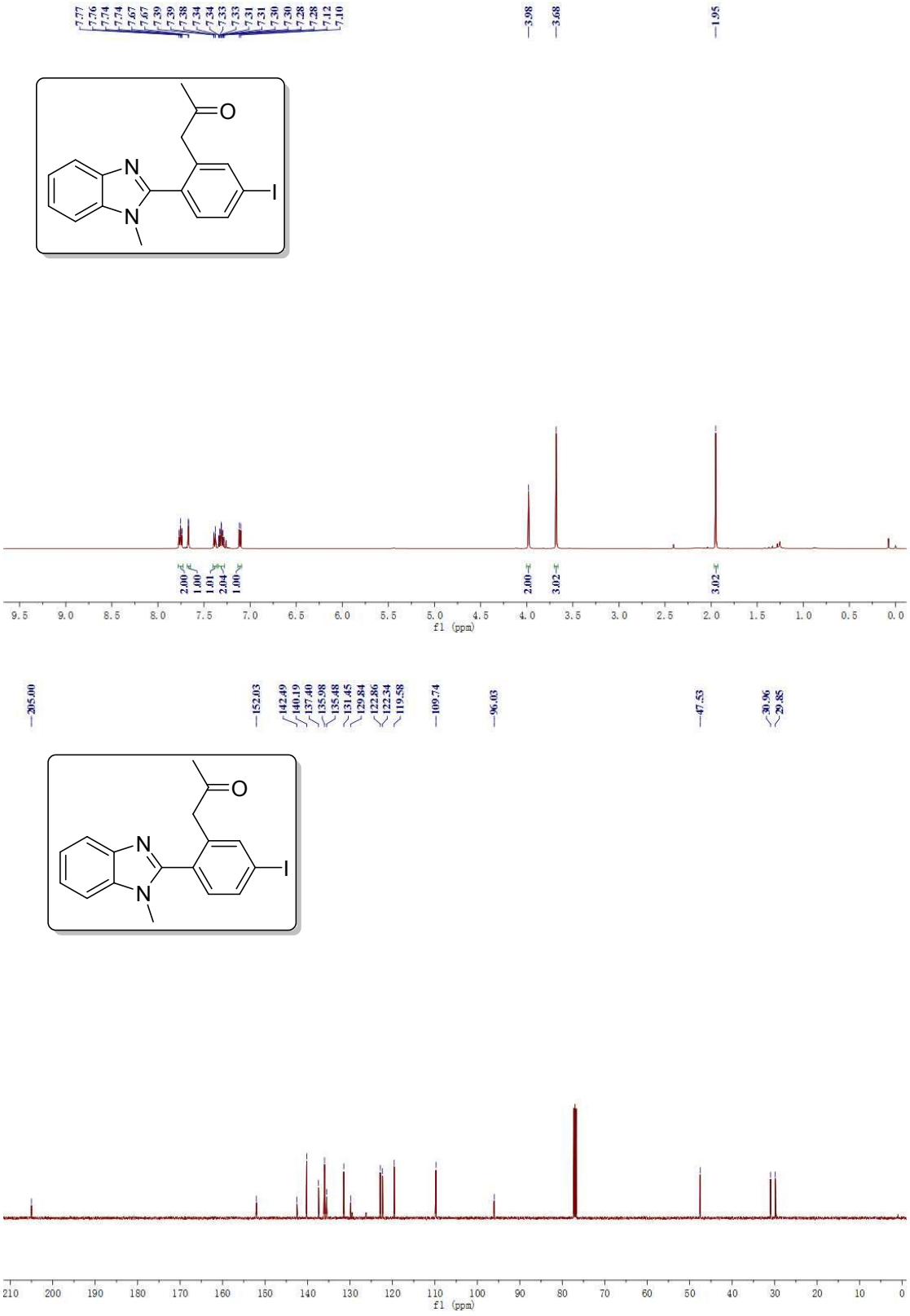


1-(4-bromo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3p)



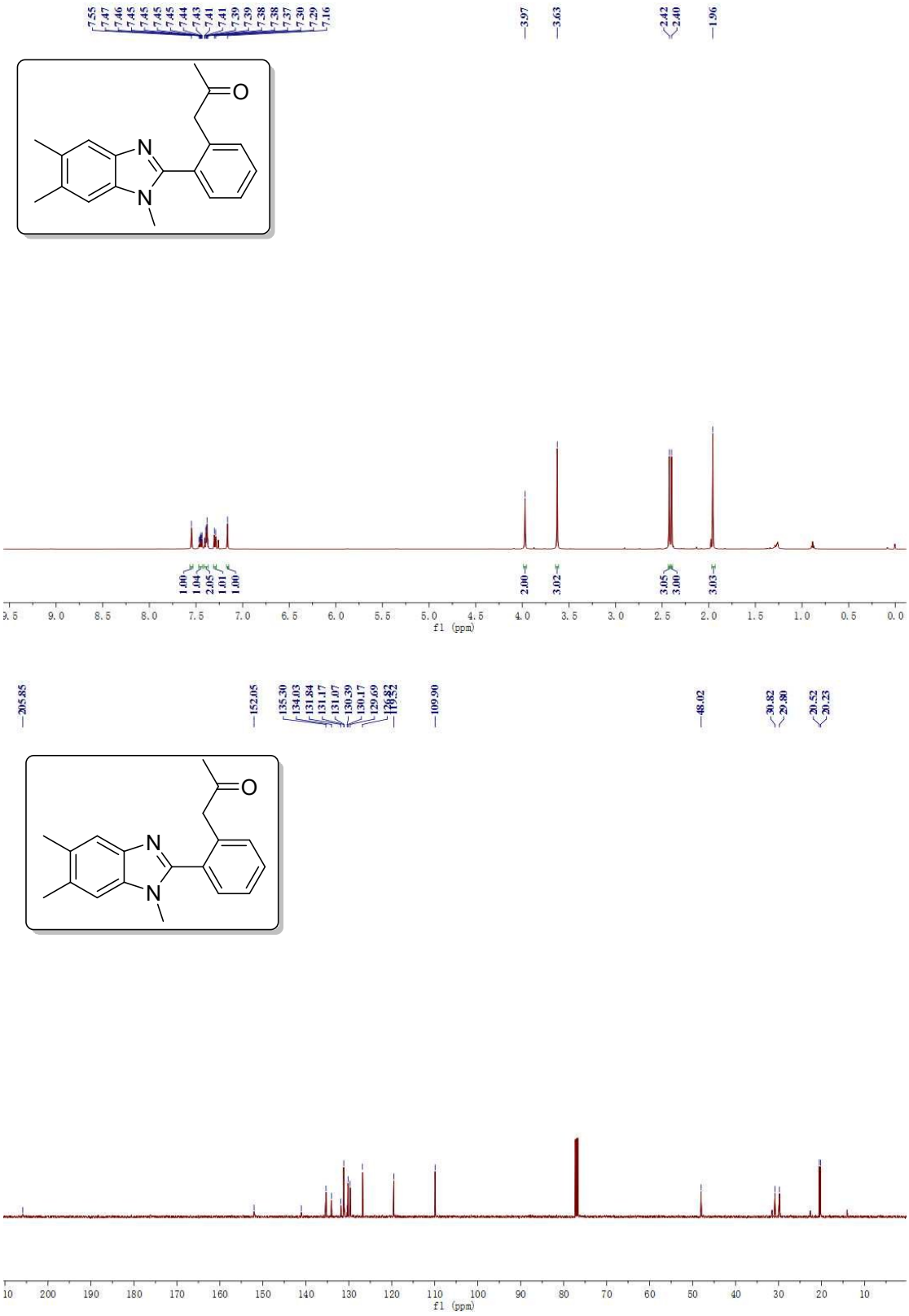


1-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3q)

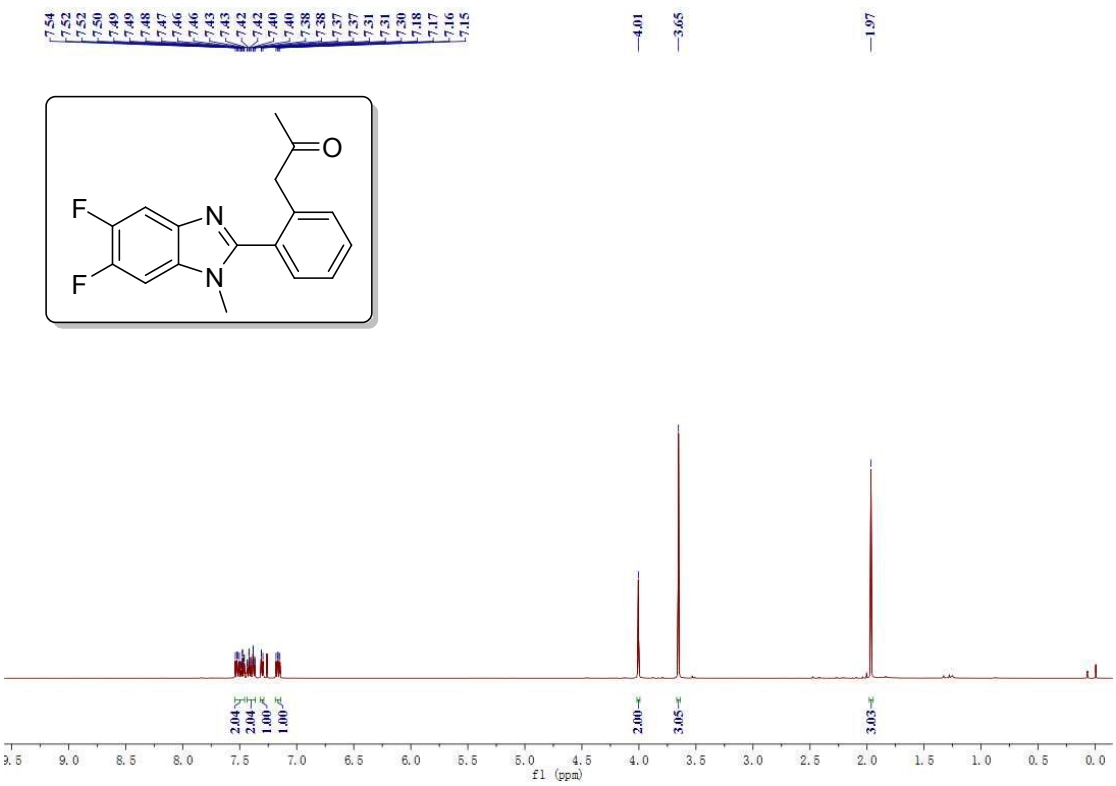


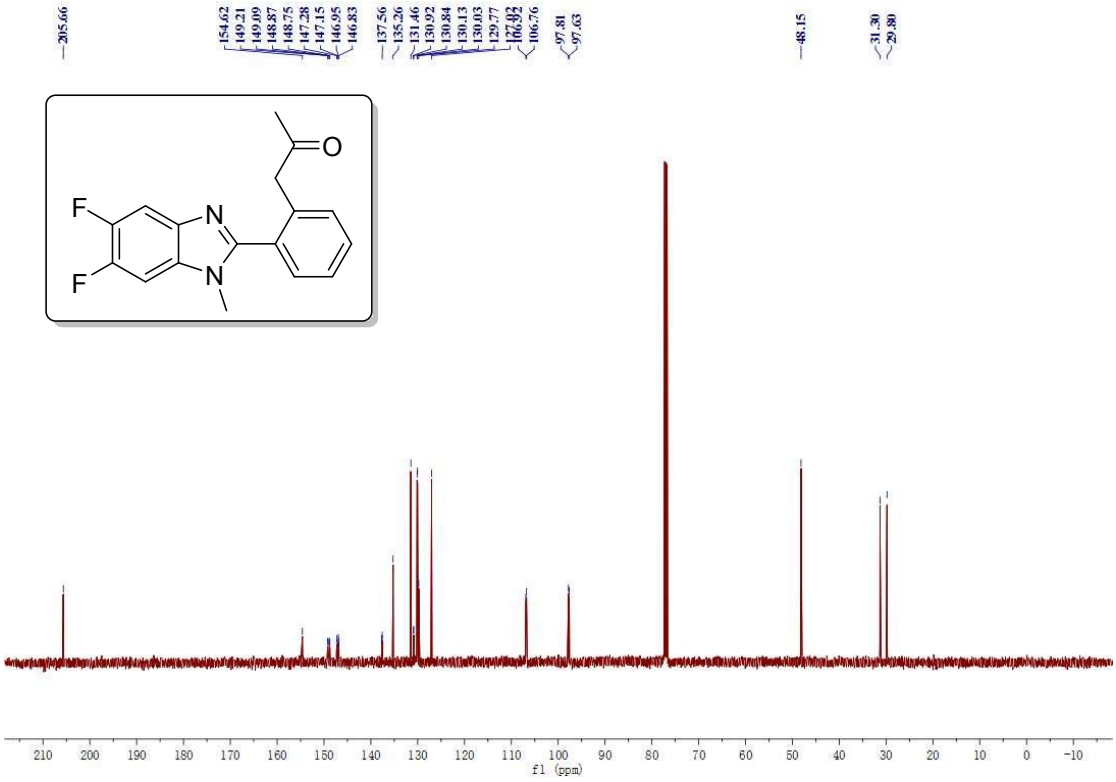
1-(5-chloro-3-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3r)

1-(2-(1,5,6-trimethyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3s)

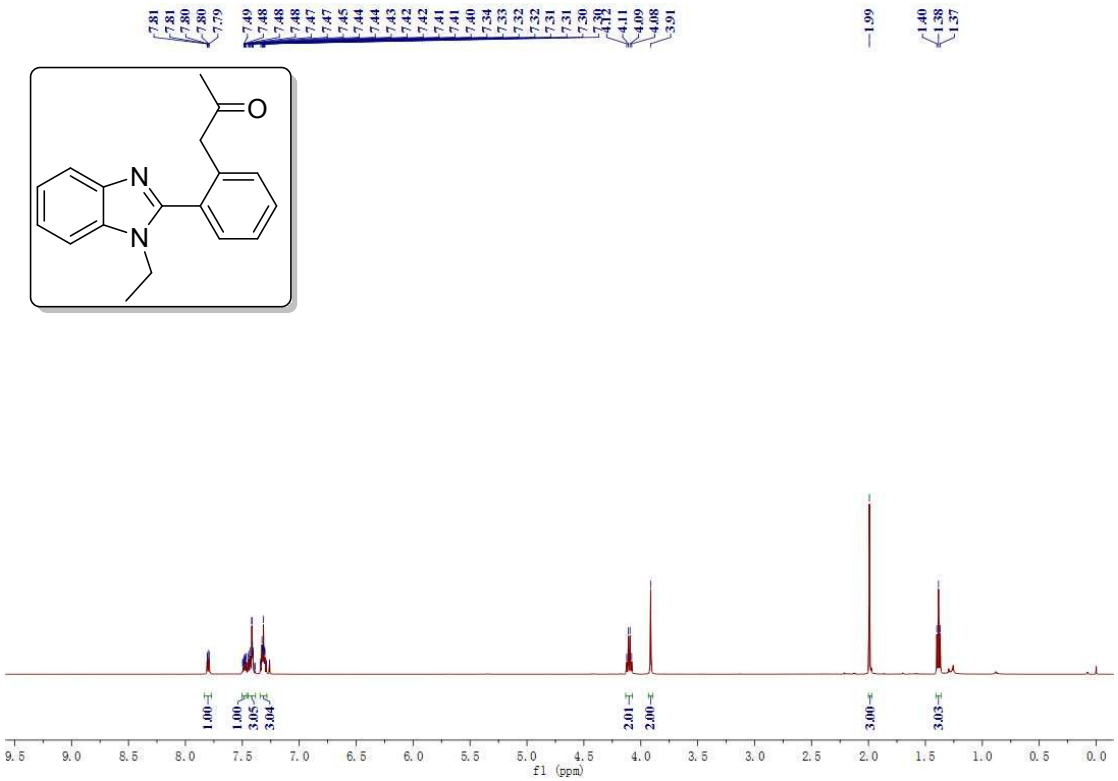


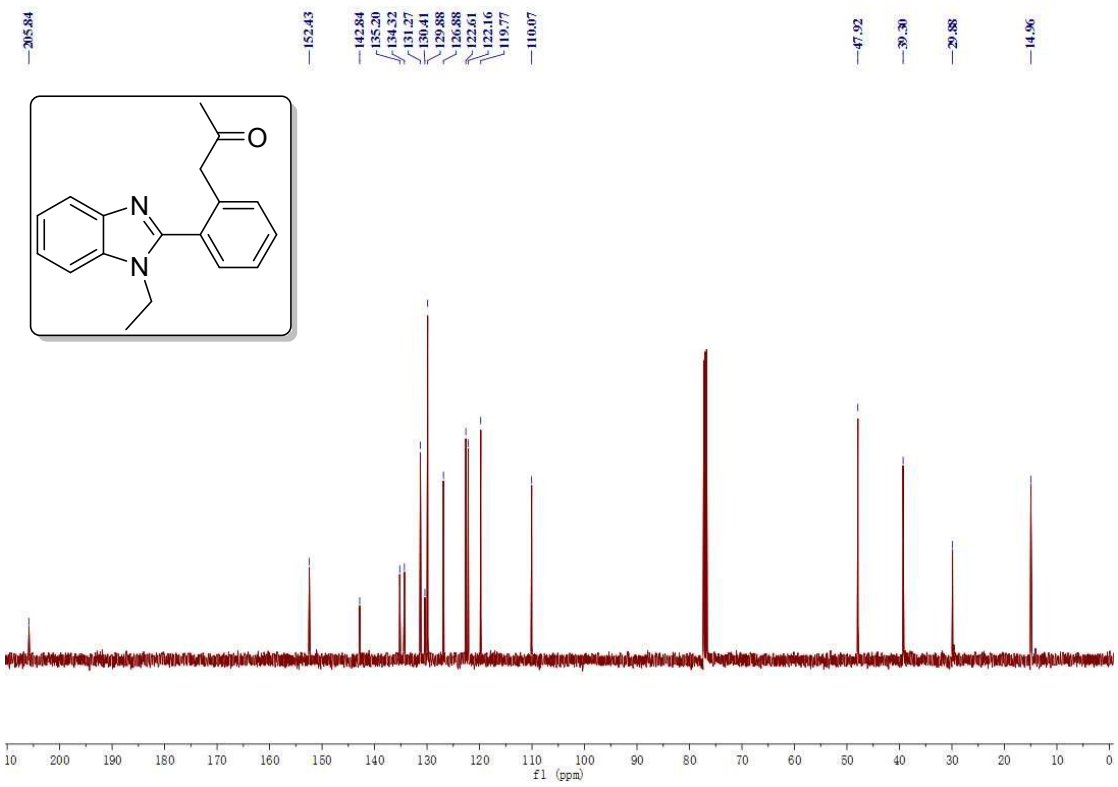
1-(2-(5,6-difluoro-1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3t)



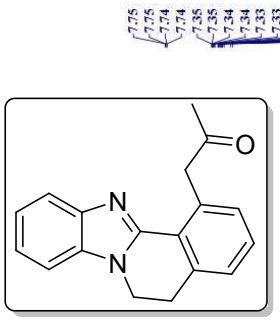


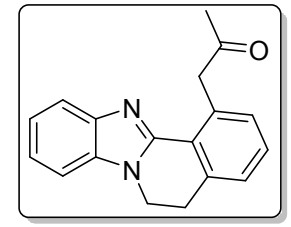
1-(2-(1-ethyl-1H-benzo[d]imidazol-2-yl)phenyl)propan-2-one (3u)



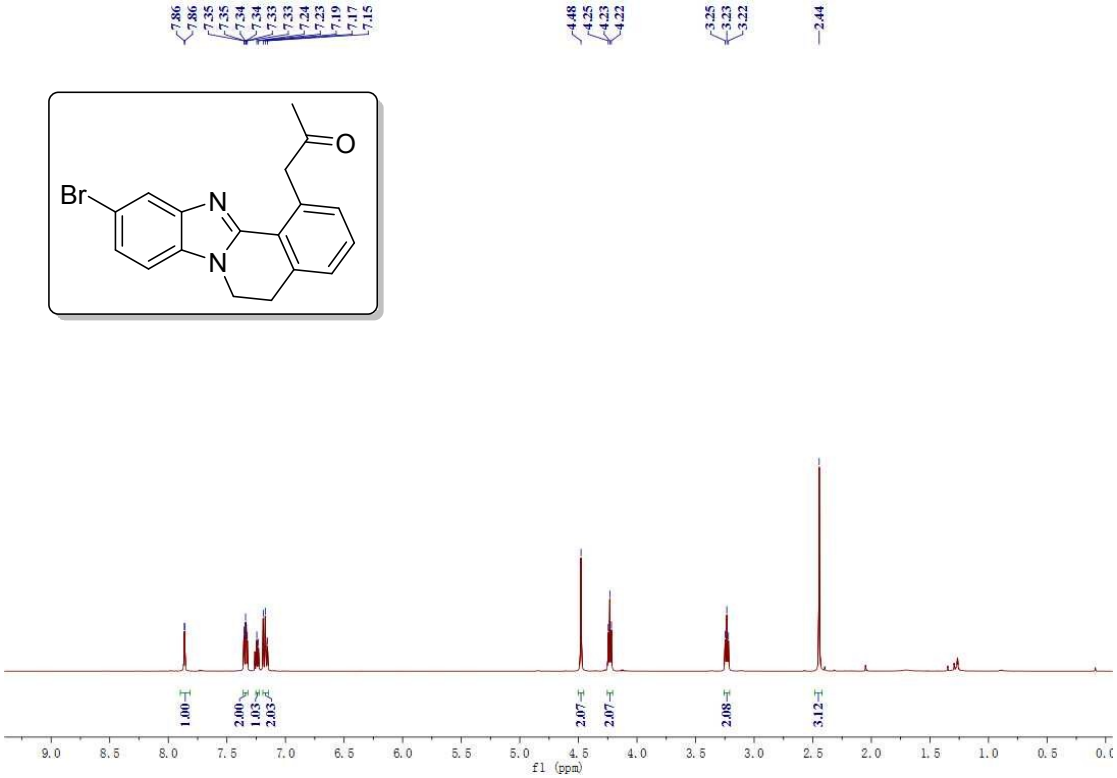


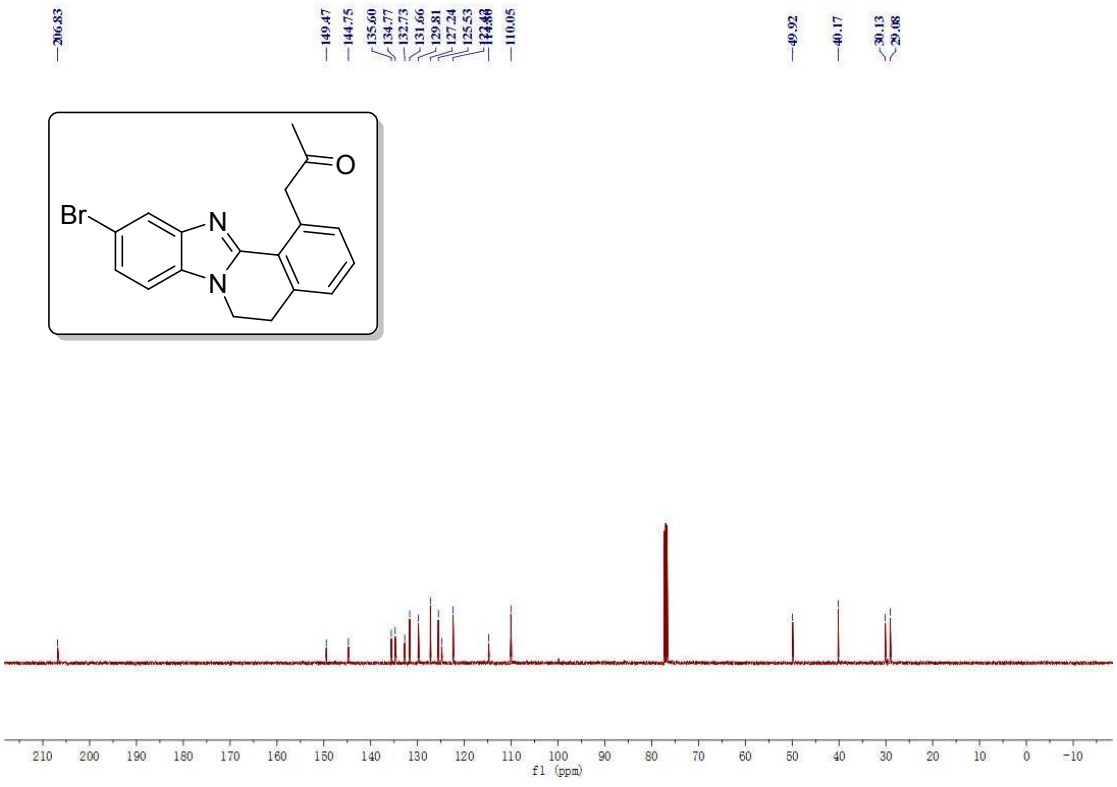
1-(5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (3v)



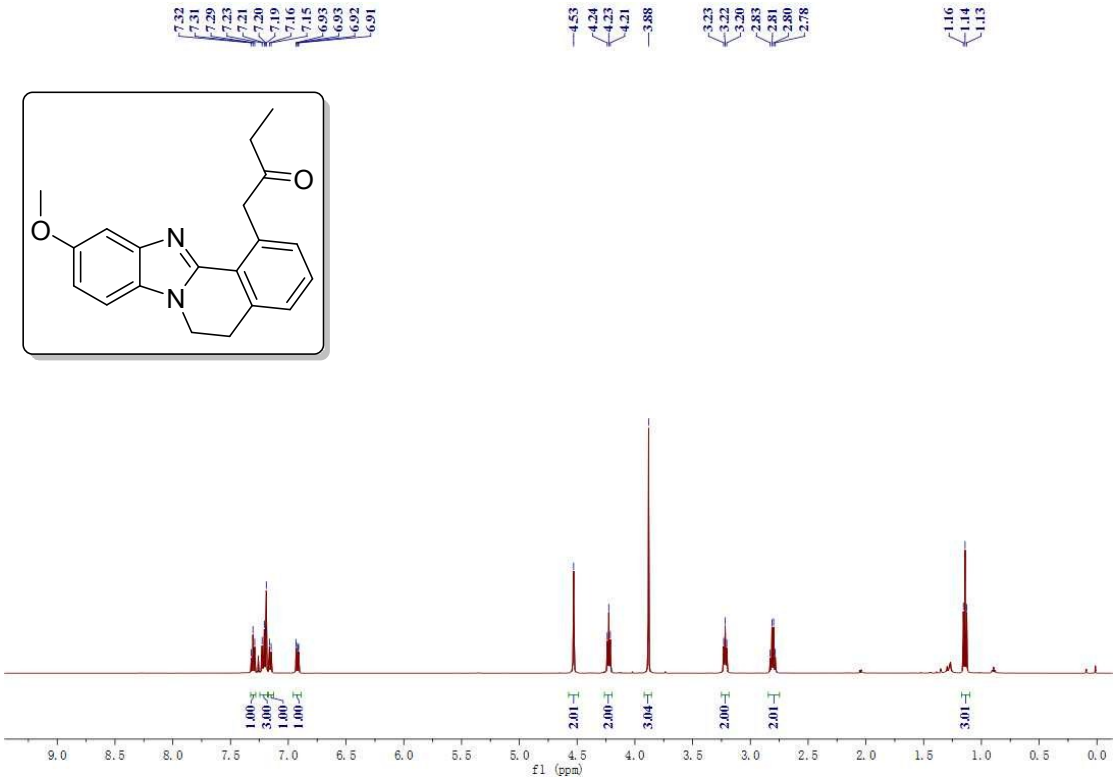


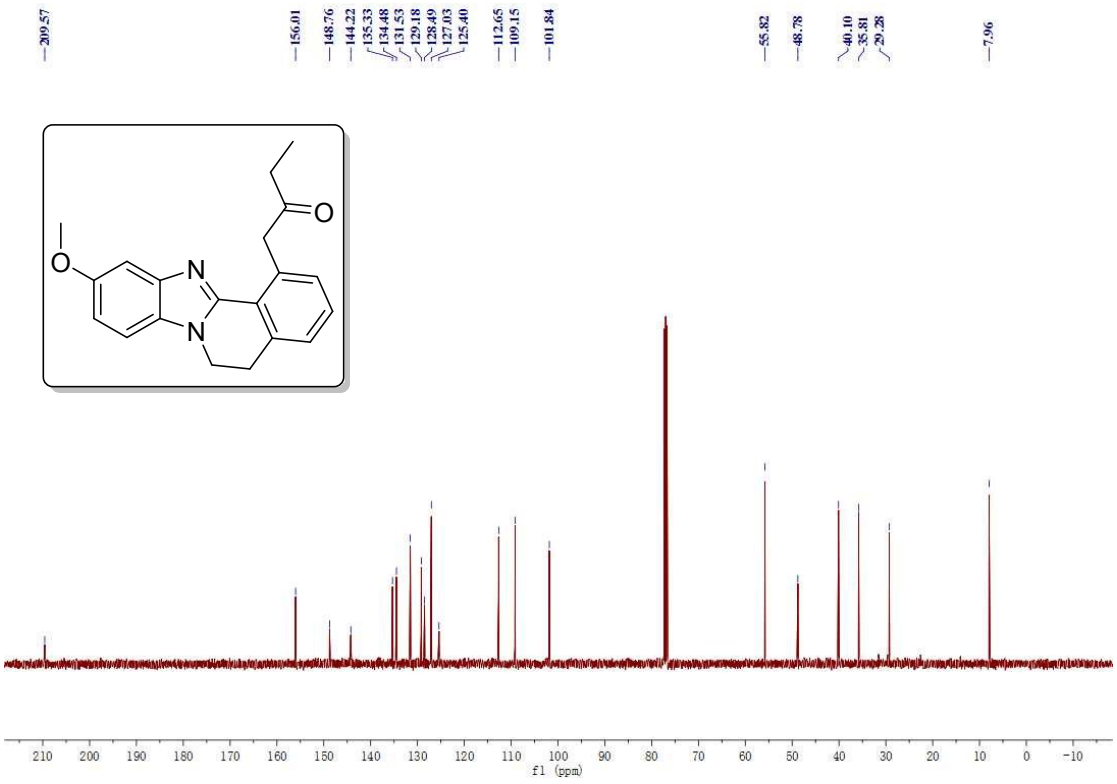
1-(10-bromo-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (3w)



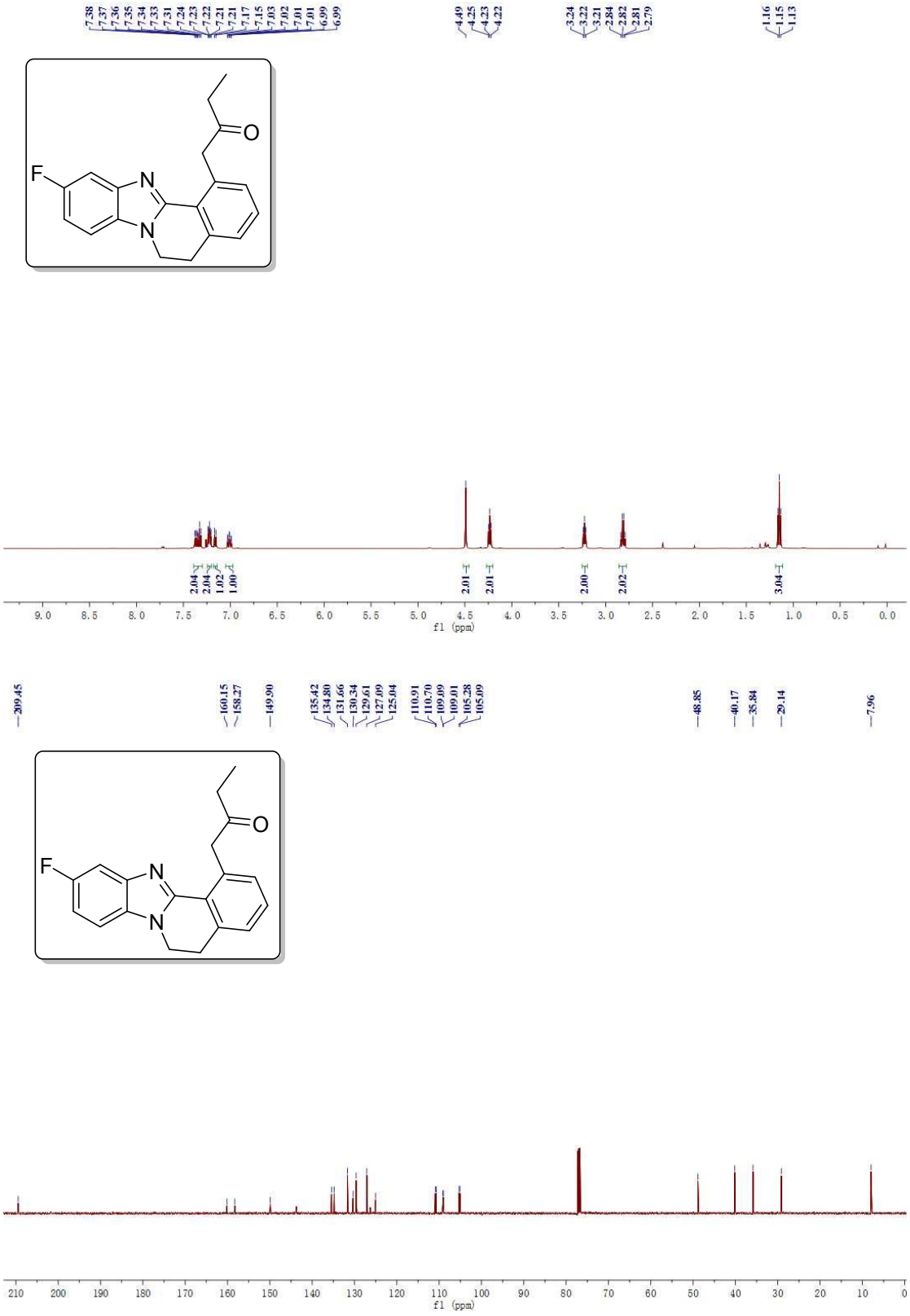


1-(10-methoxy-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3x)

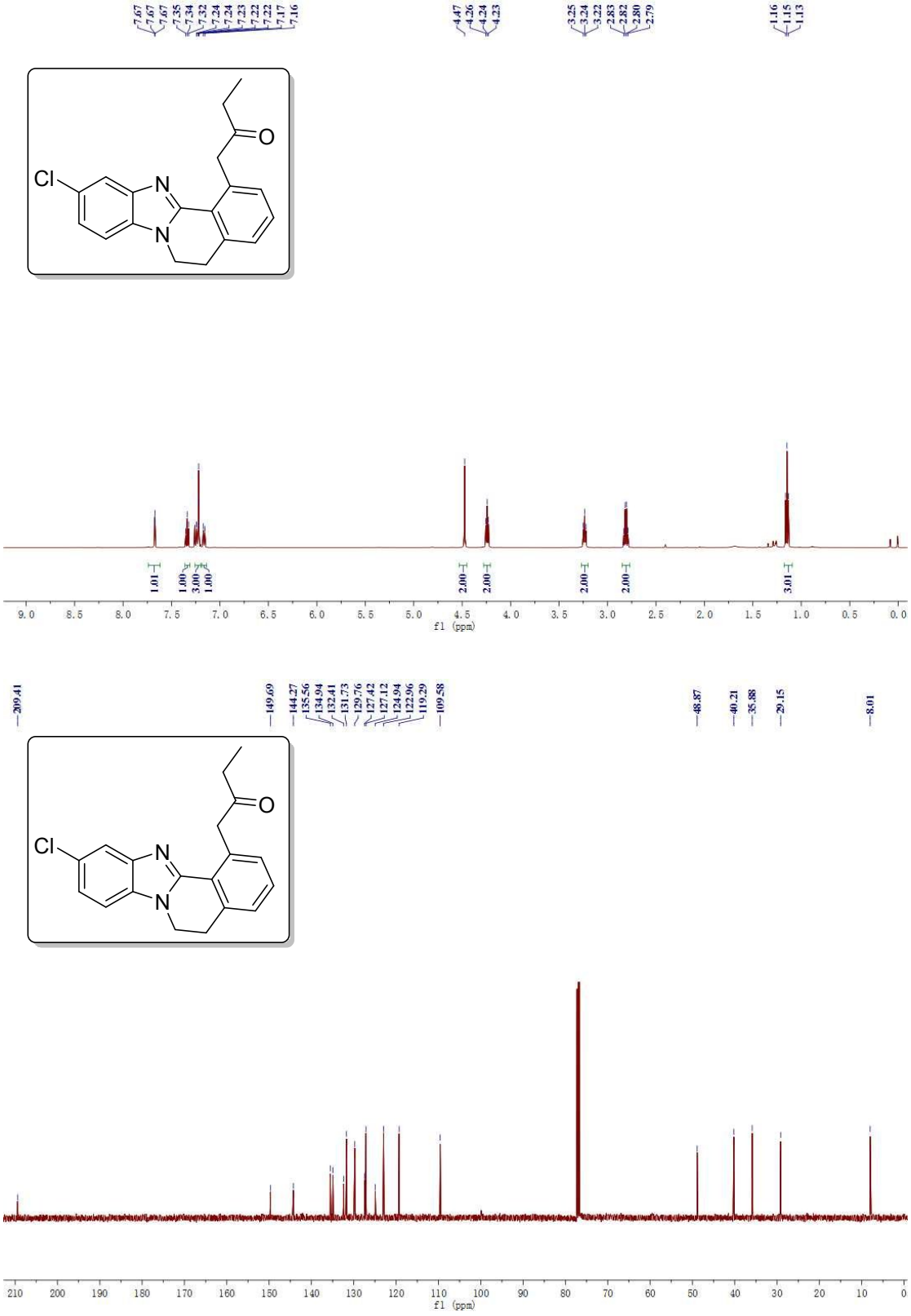




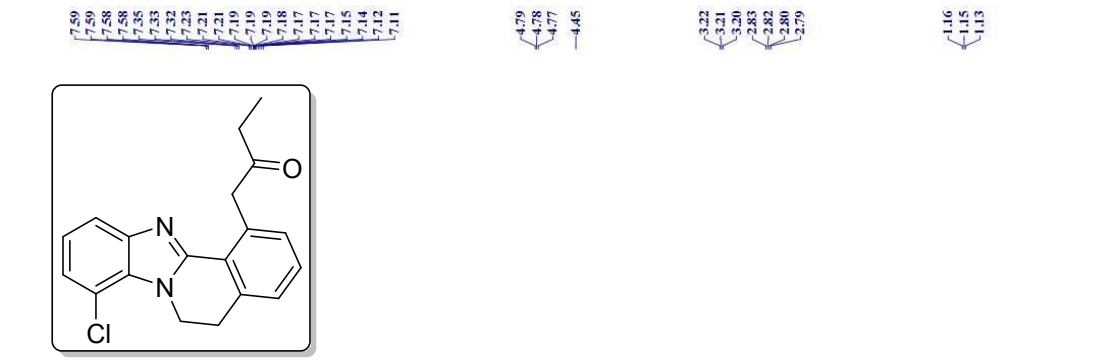
1-(10-fluoro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3y)

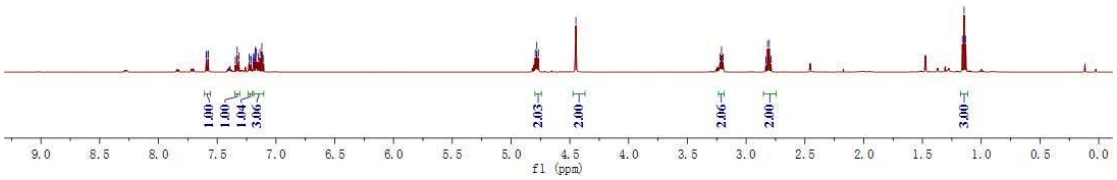


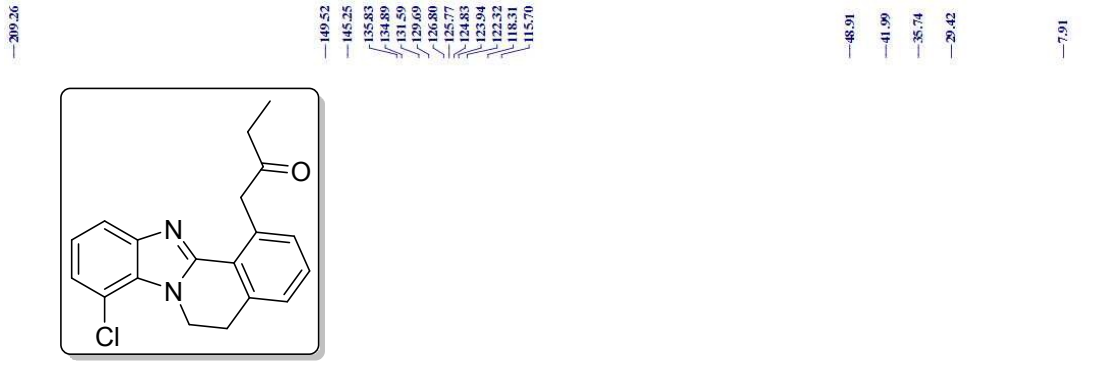
1-(10-chloro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3z)

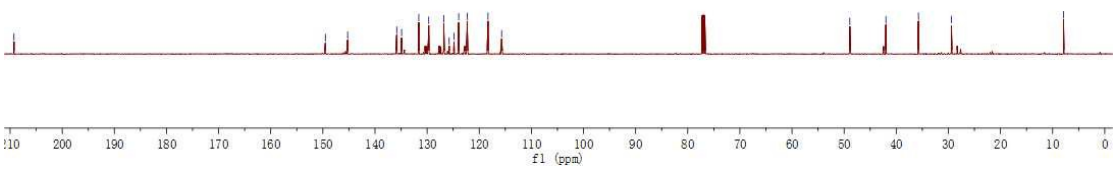


1-(8-chloro-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3aa)

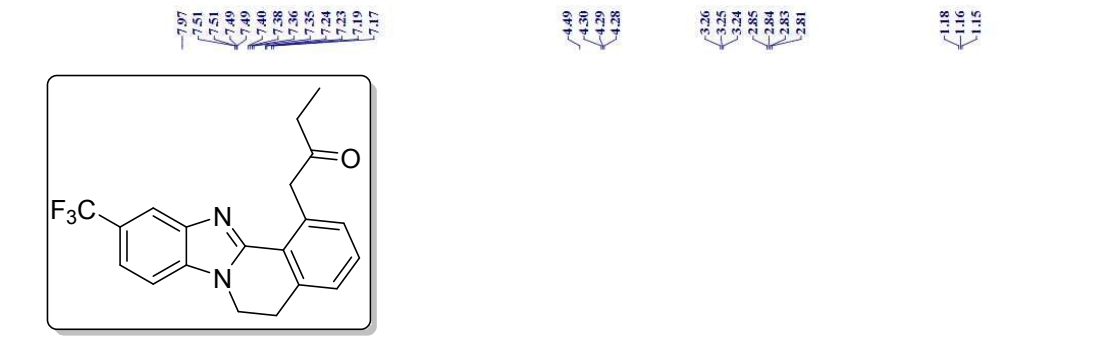


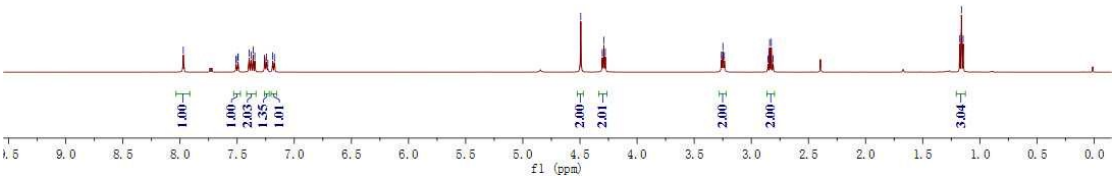


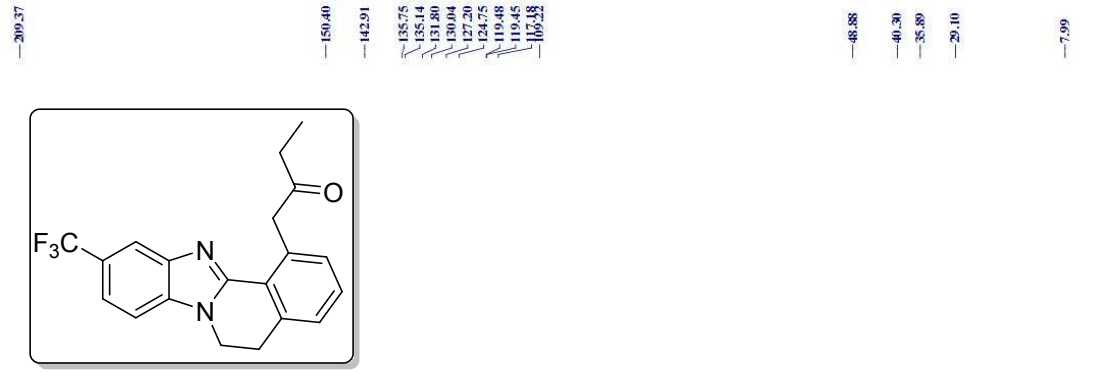




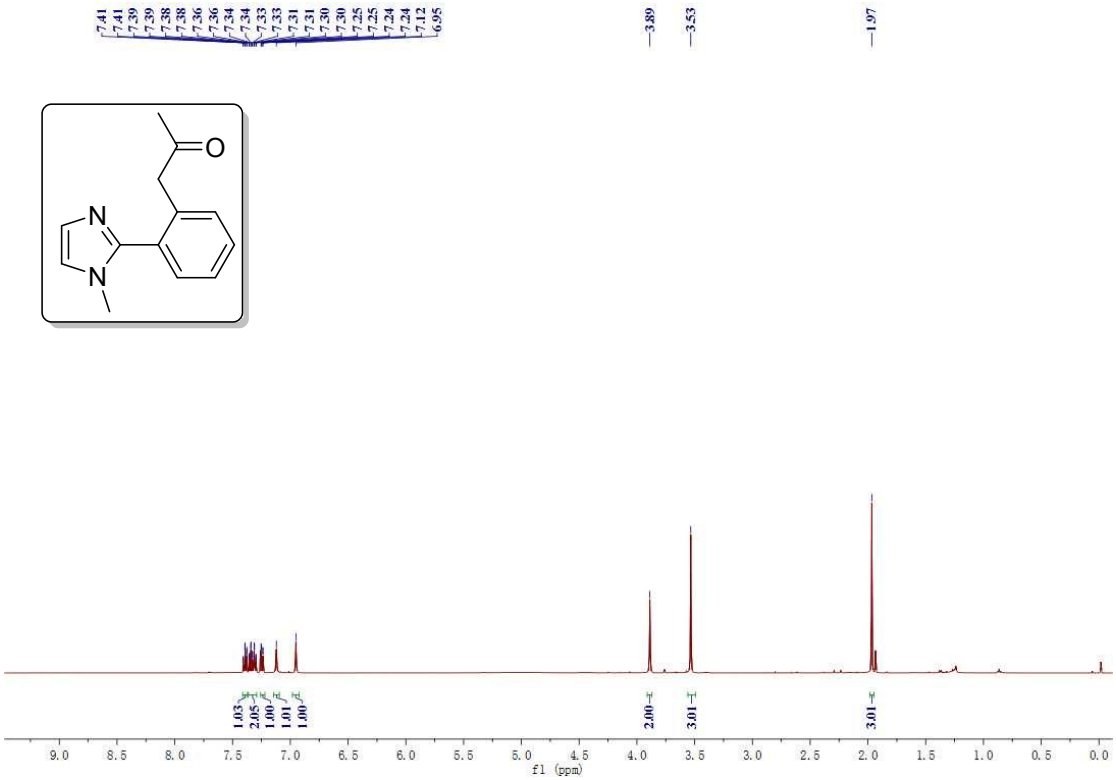
1-(10-(trifluoromethyl)-5,6-dihydrobenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (3ab)

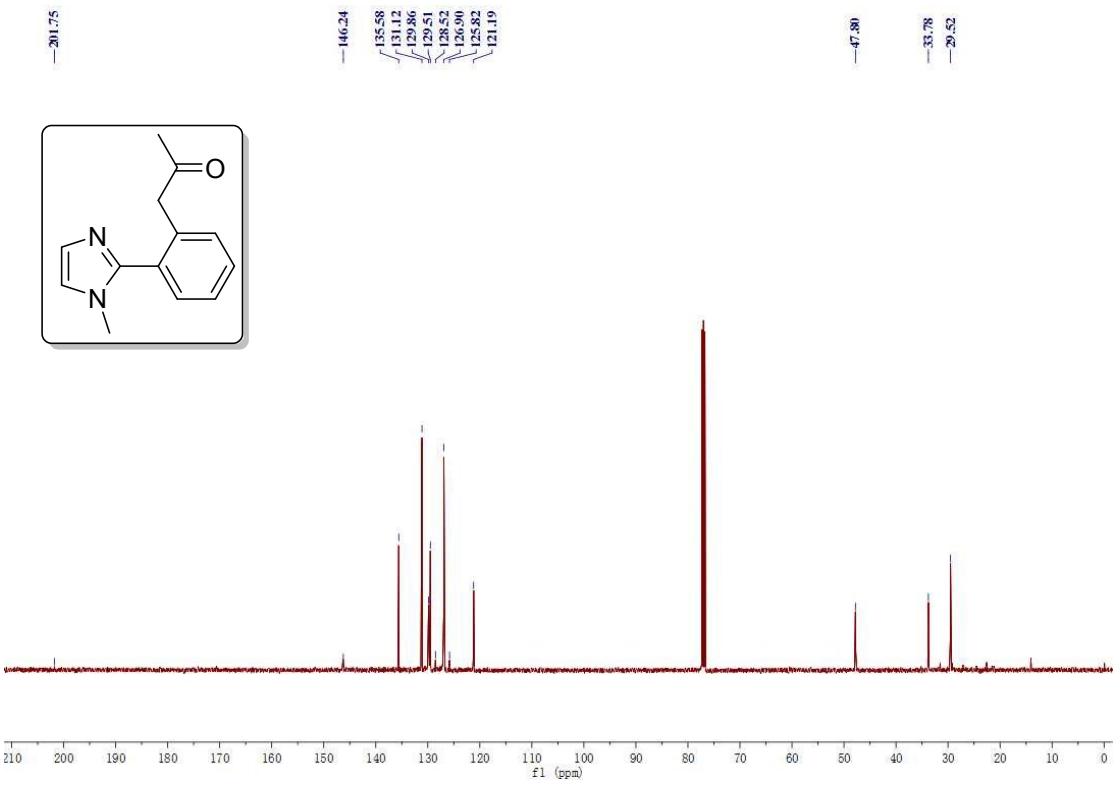




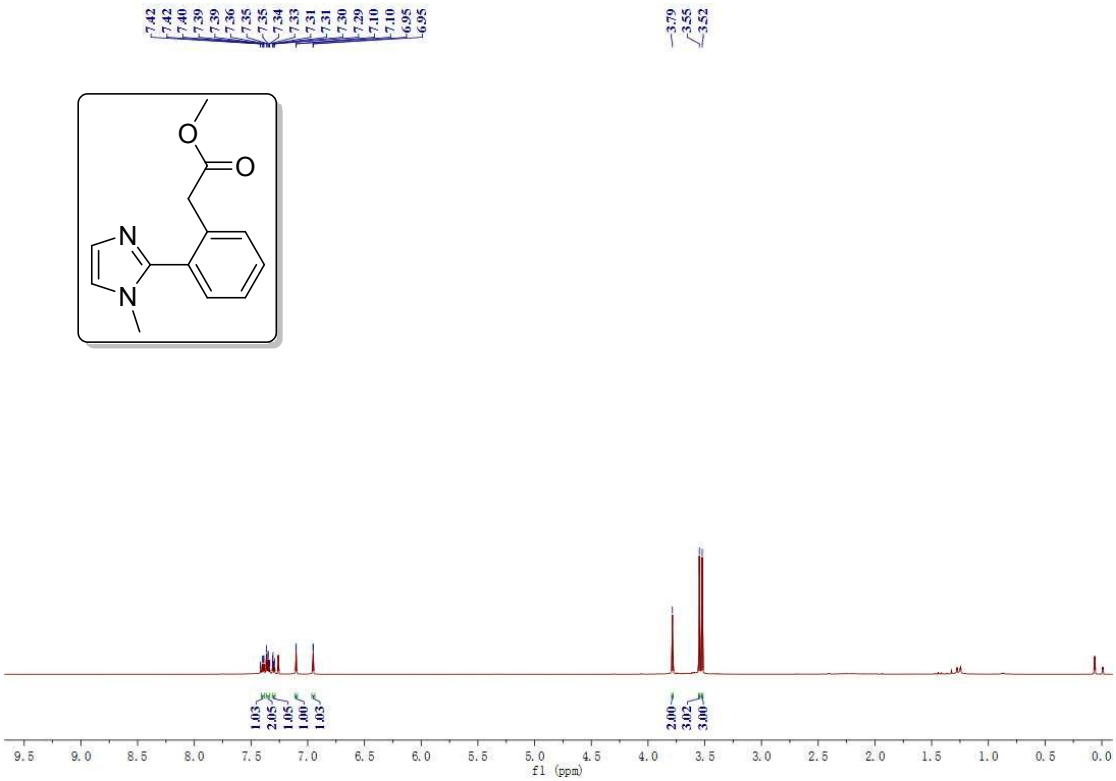


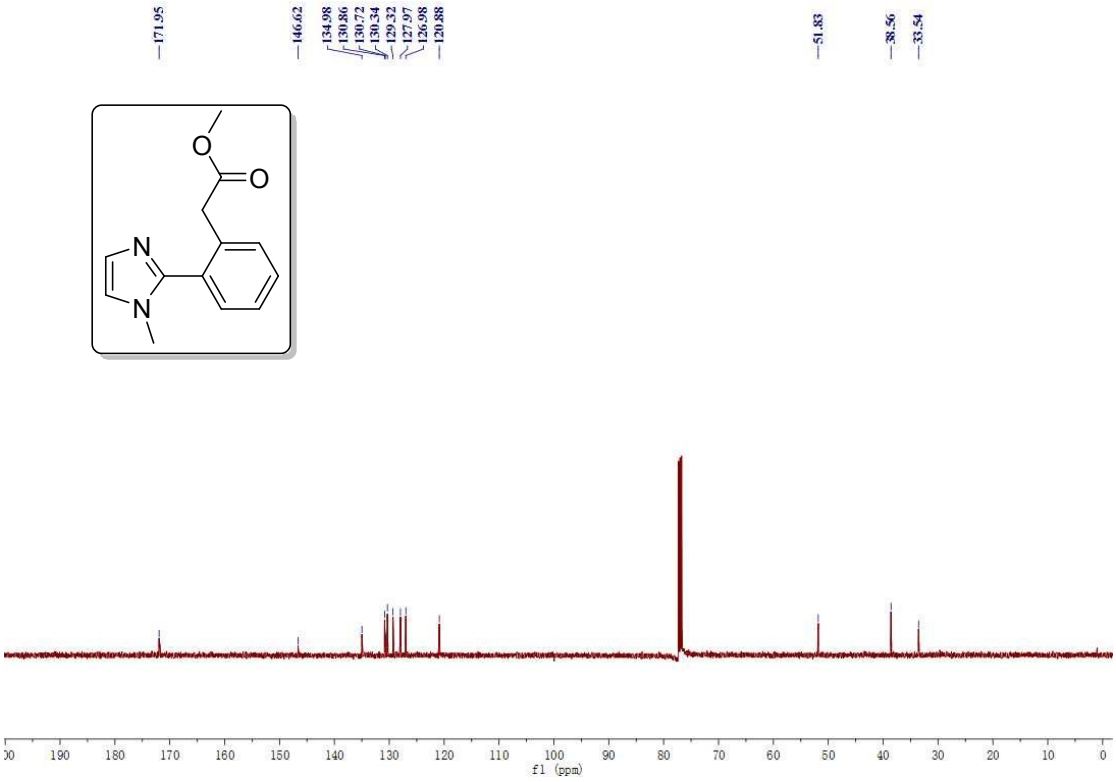
1-(2-(1-methyl-1H-imidazol-2-yl)phenyl)propan-2-one (3ac)



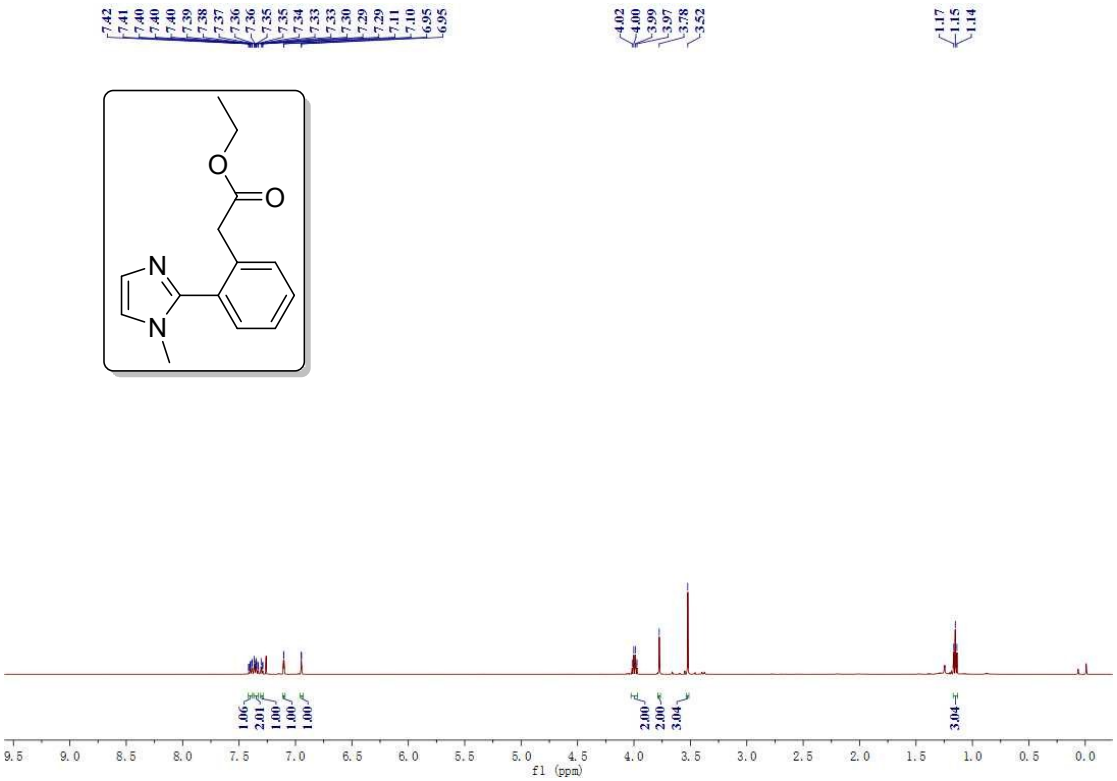


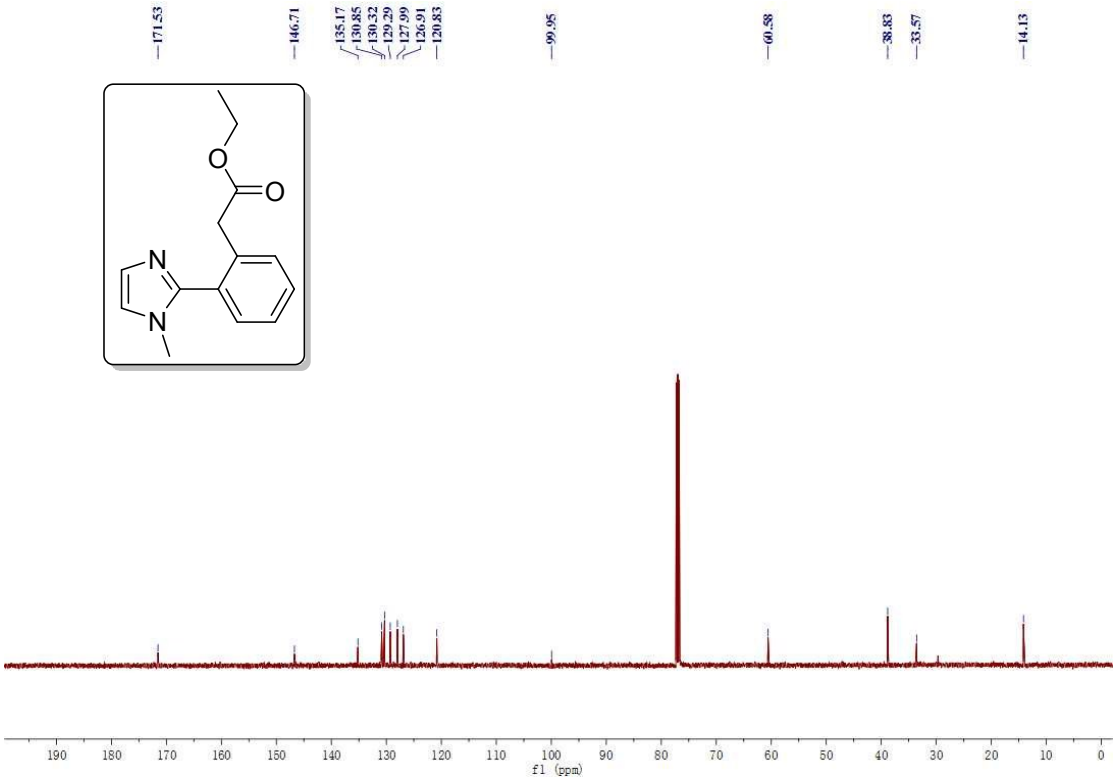
methyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4a)



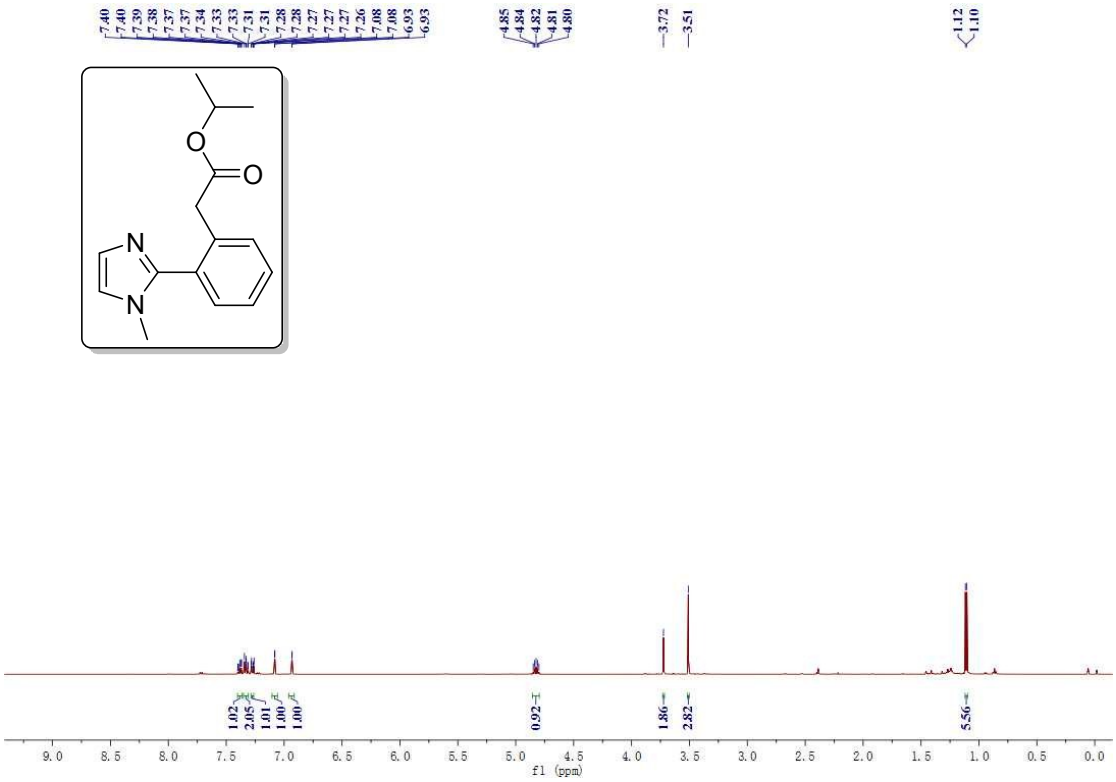


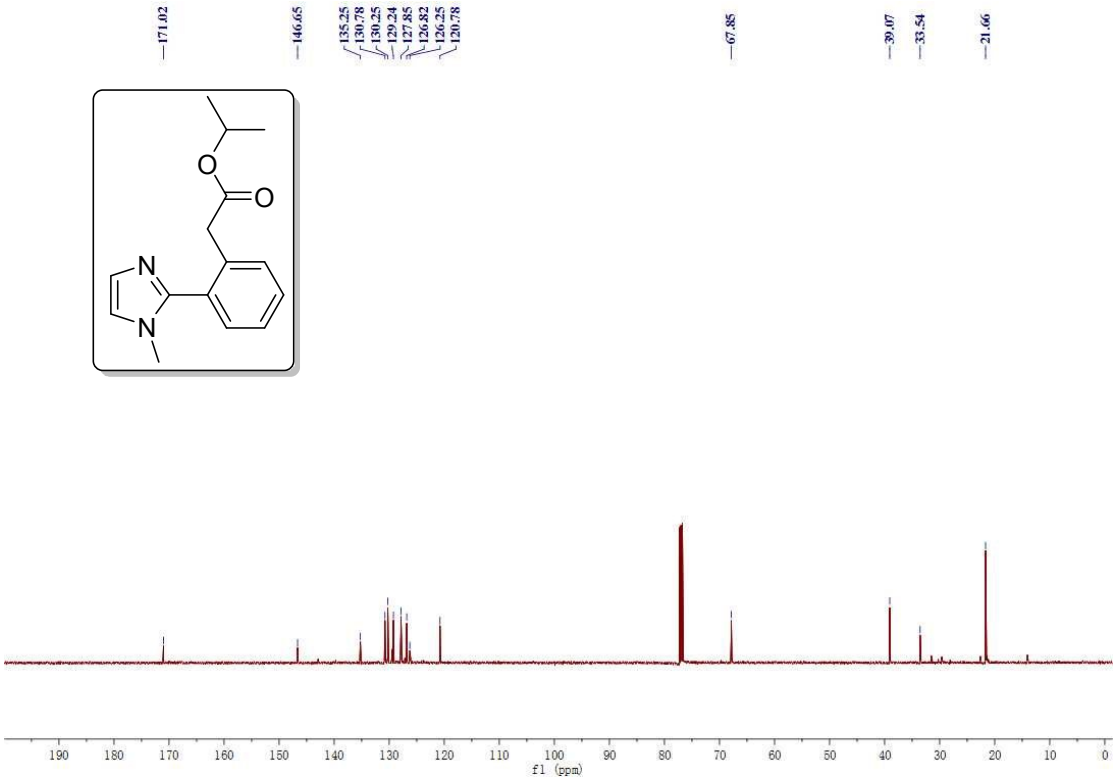
ethyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4b)



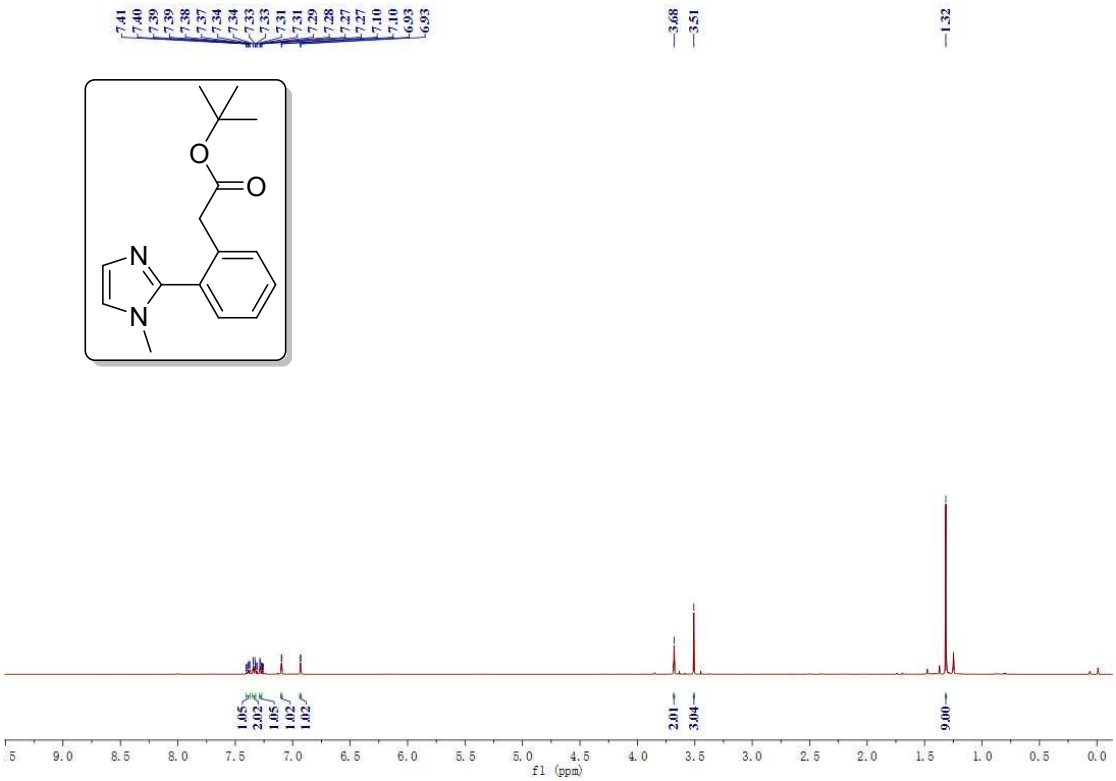


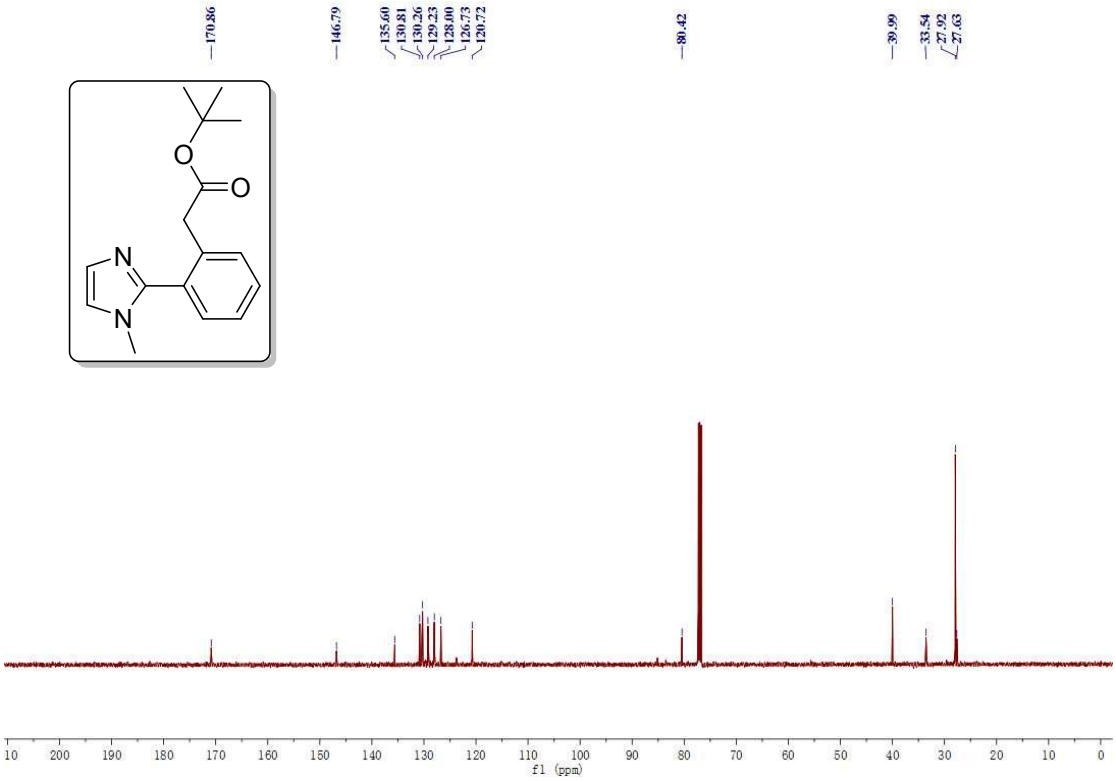
isopropyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4c)



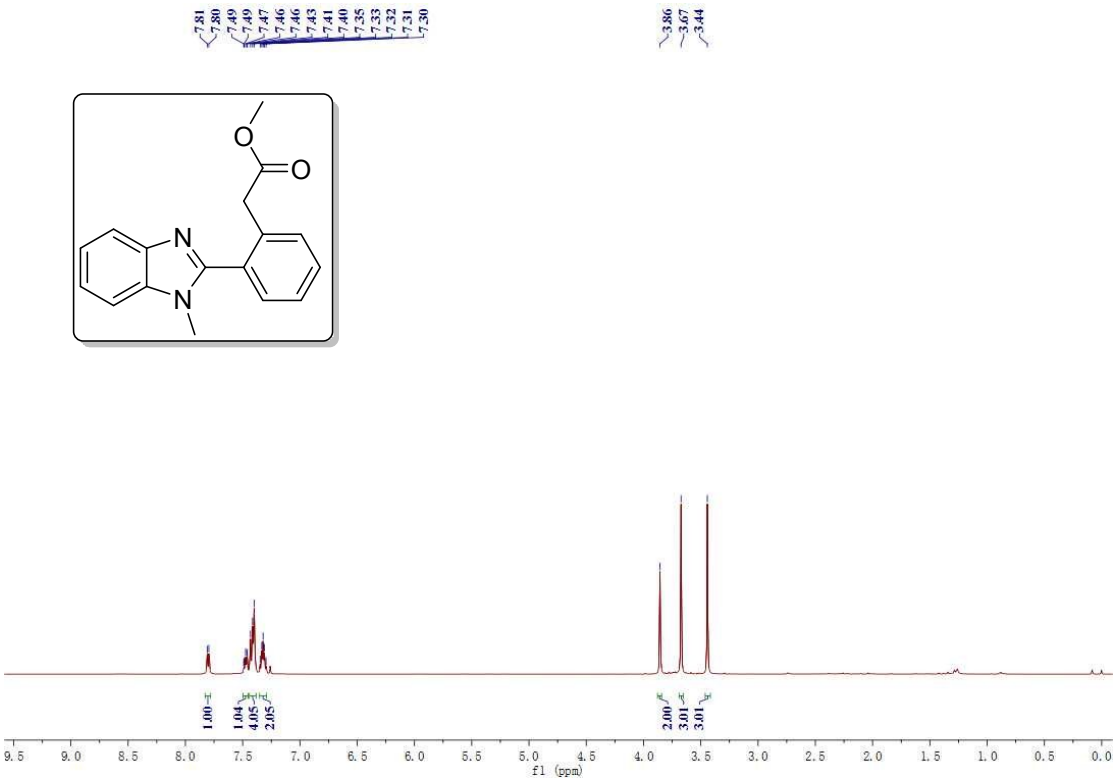


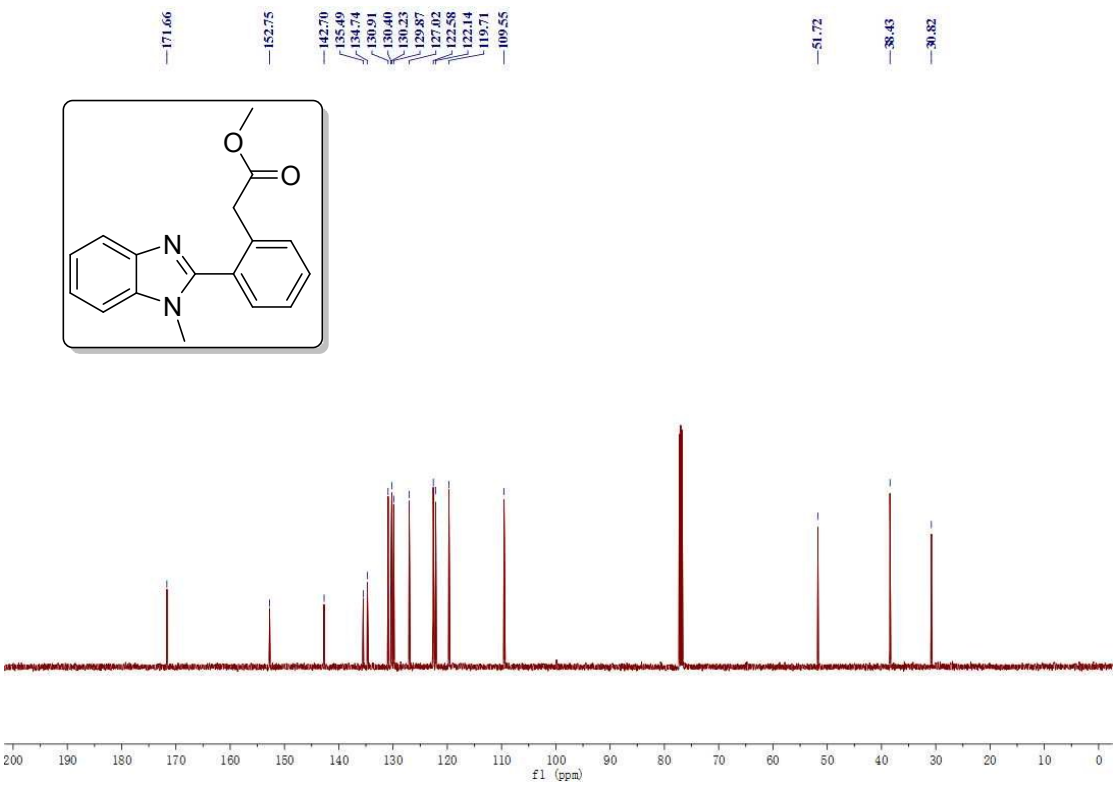
tert-butyl 2-(2-(1-methyl-1H-imidazol-2-yl)phenyl)acetate (4d)



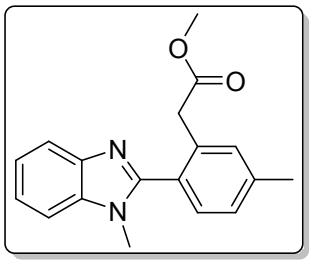


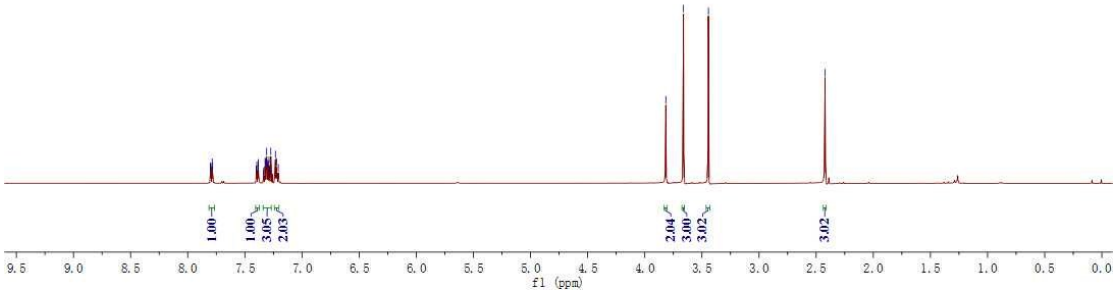
Methyl2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4e)

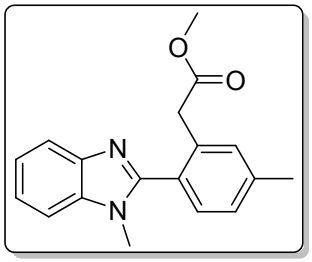


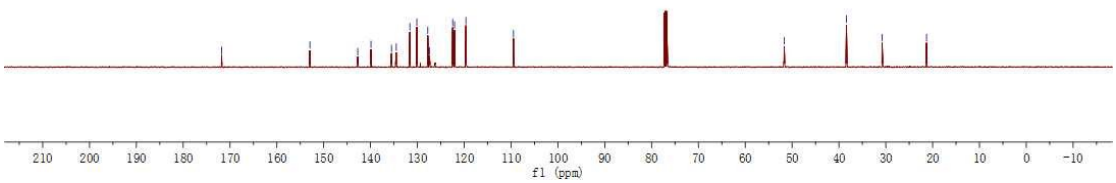


Methyl2-(5-methyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4f)

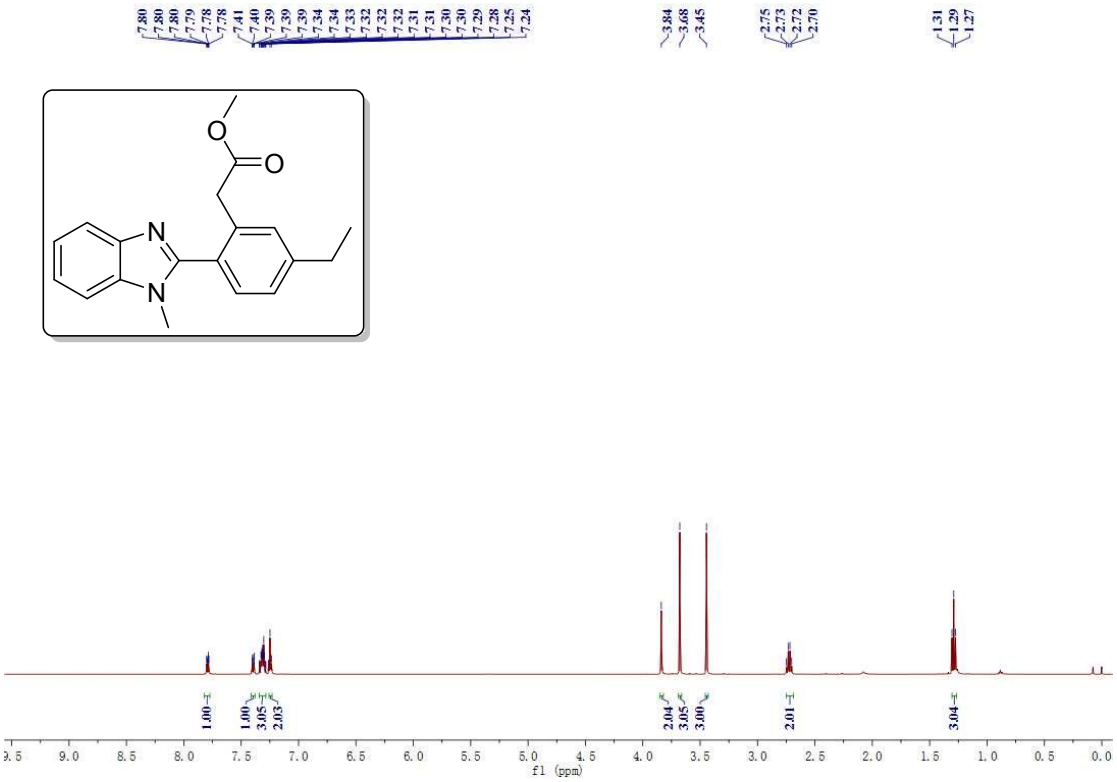


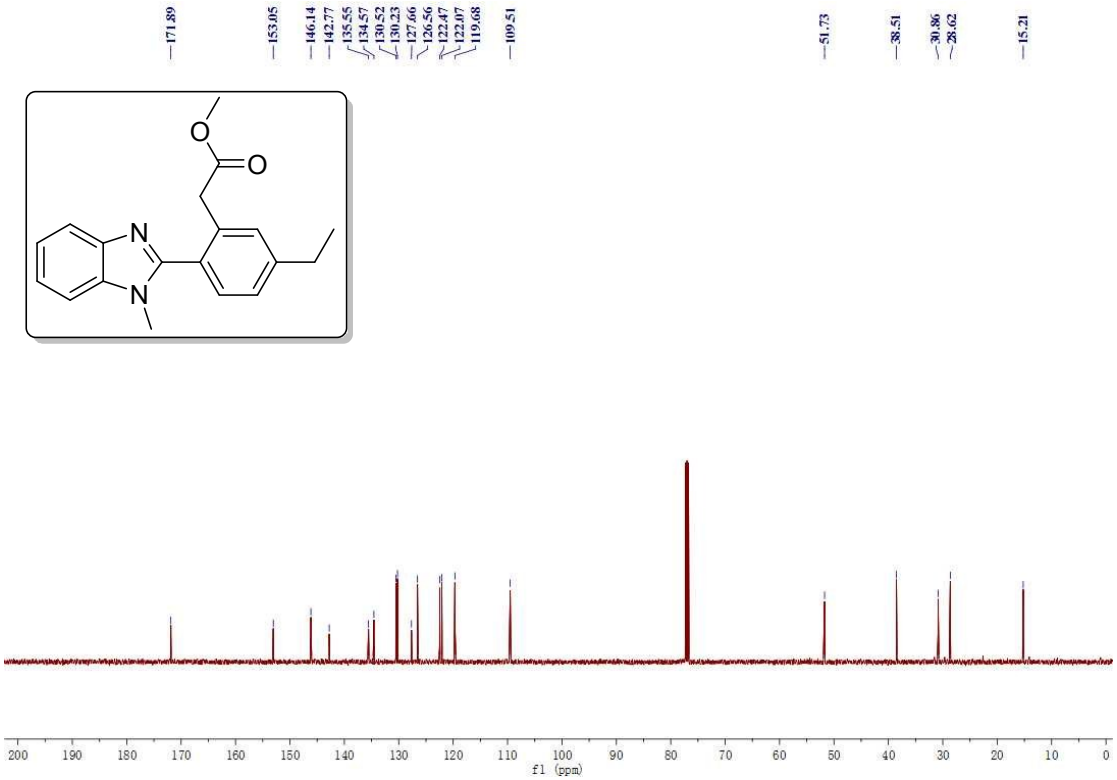




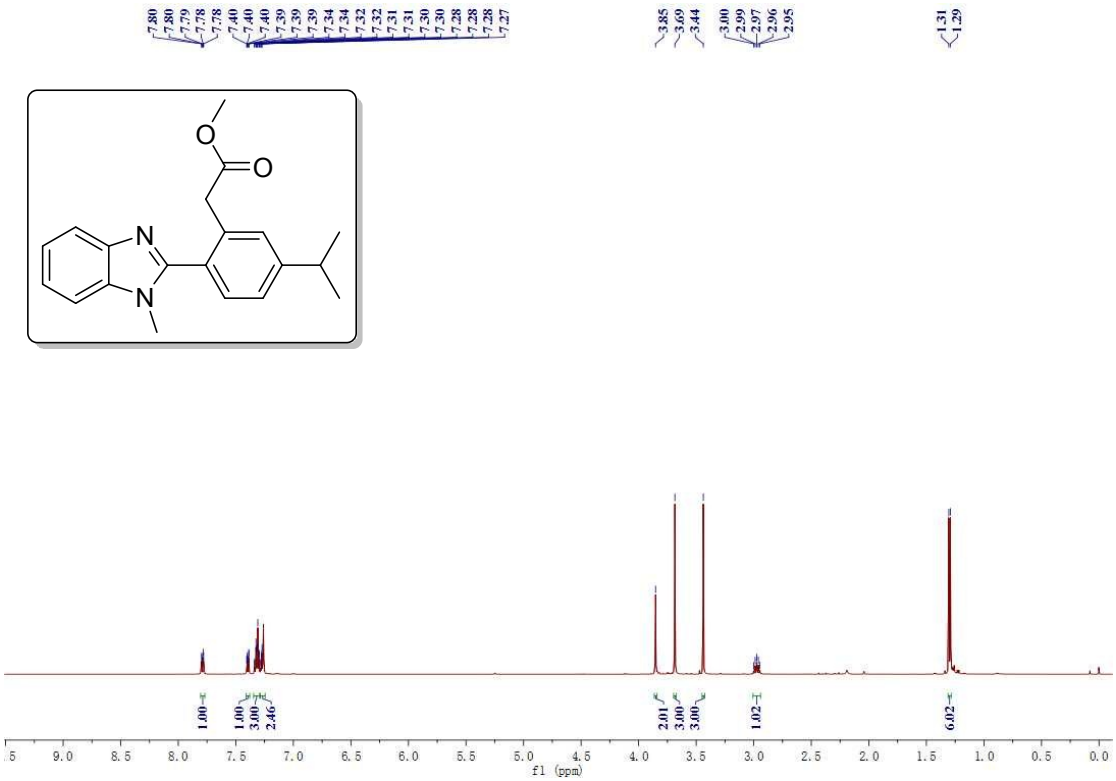


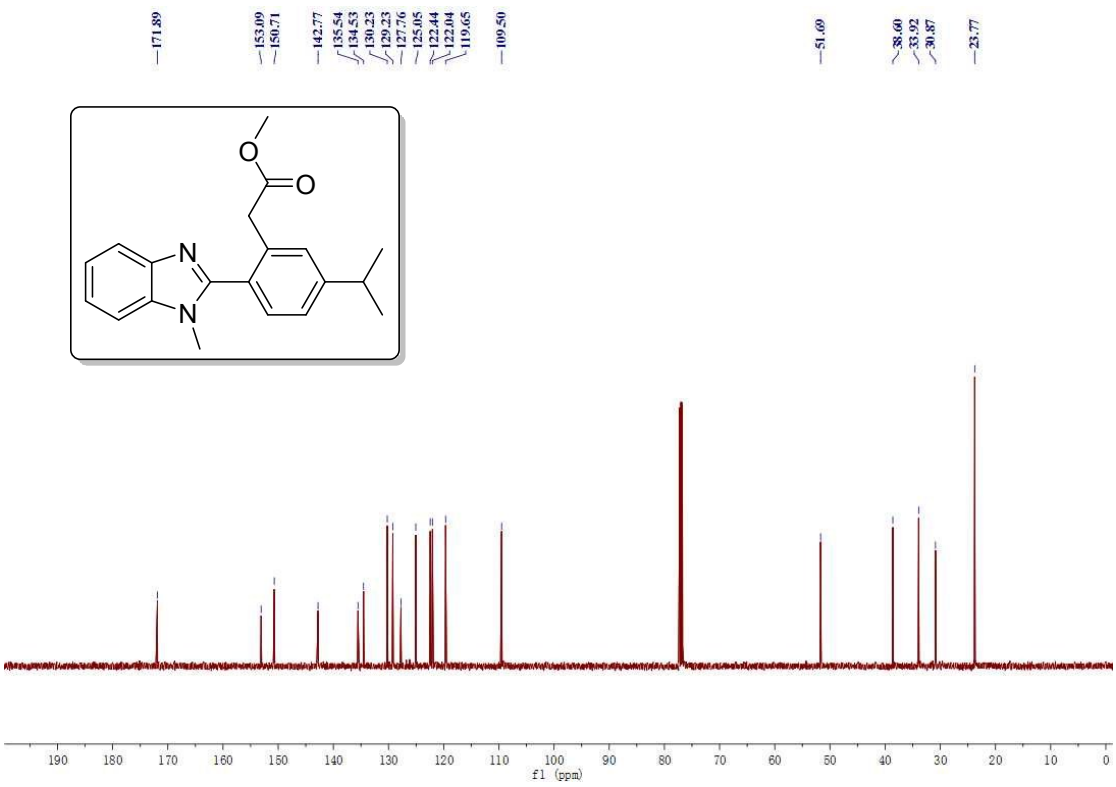
methyl 2-(5-ethyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4g)



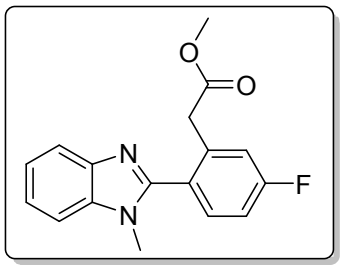


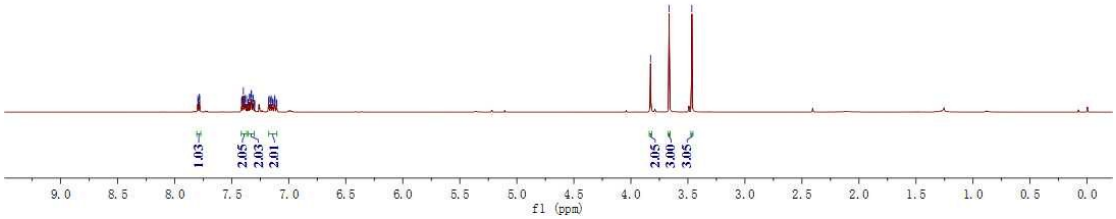
Methyl2-(5-isopropyl-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate(4h)

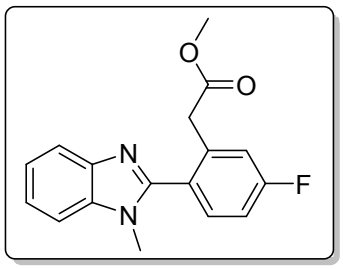


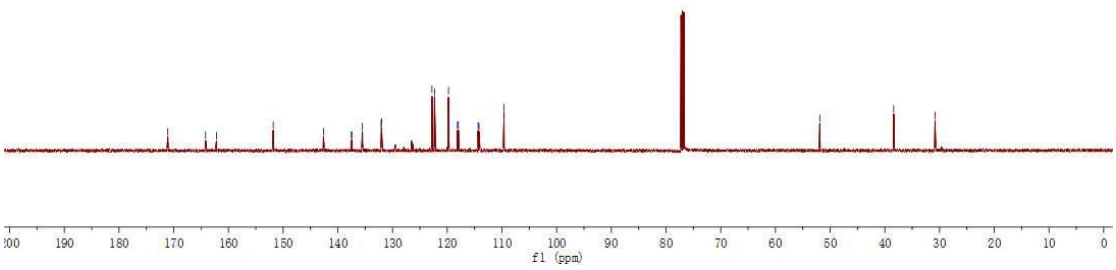


methyl2-(5-fluoro-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4i)

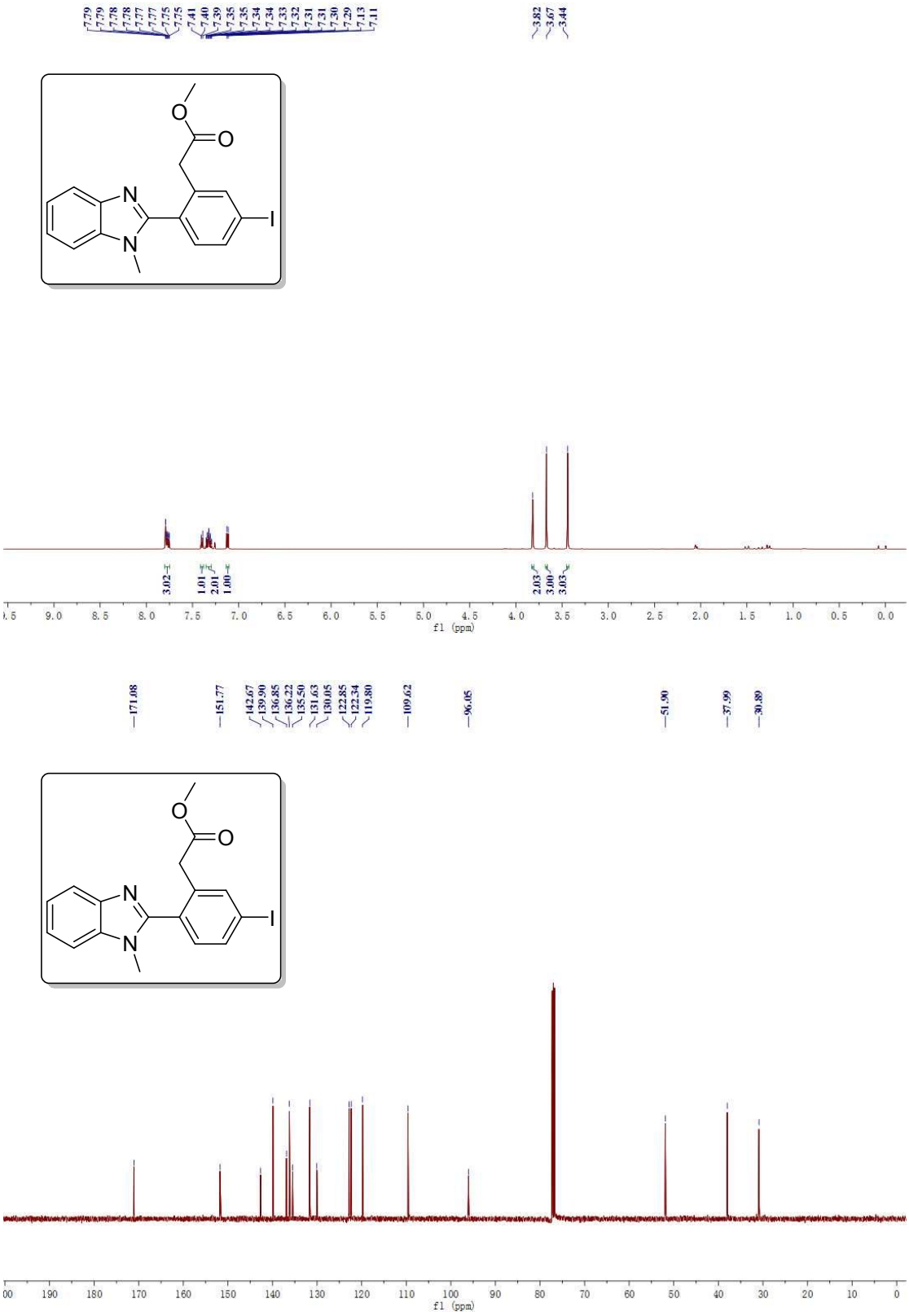




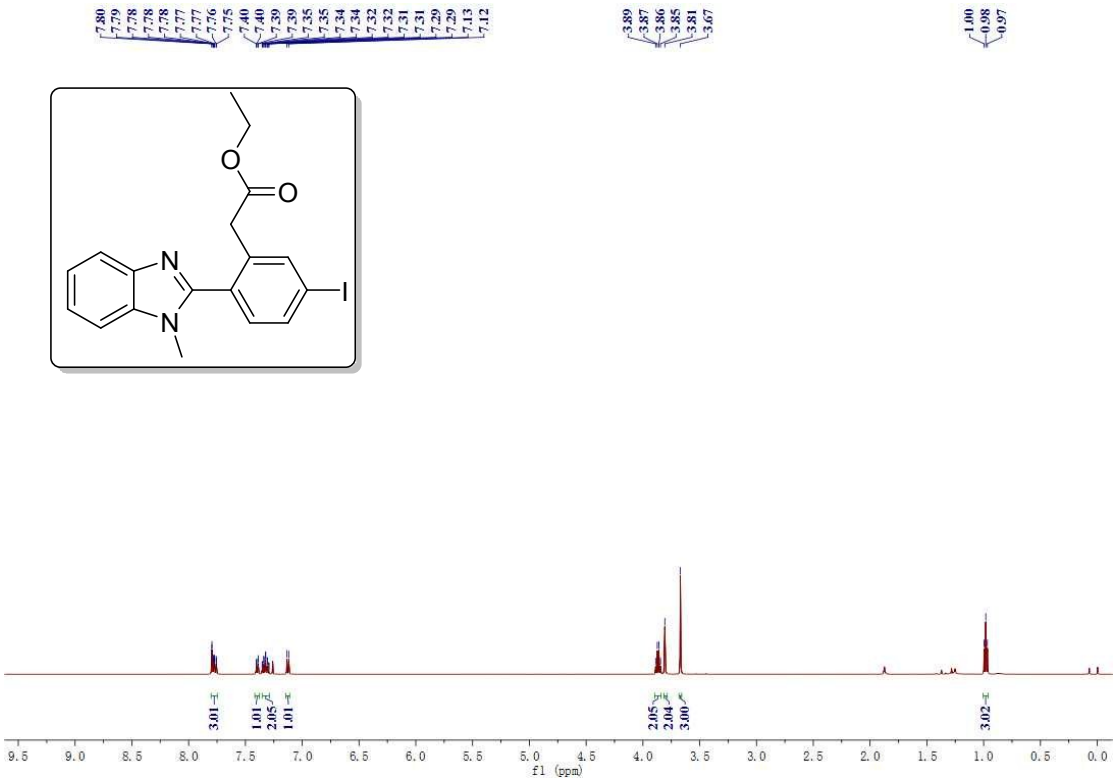


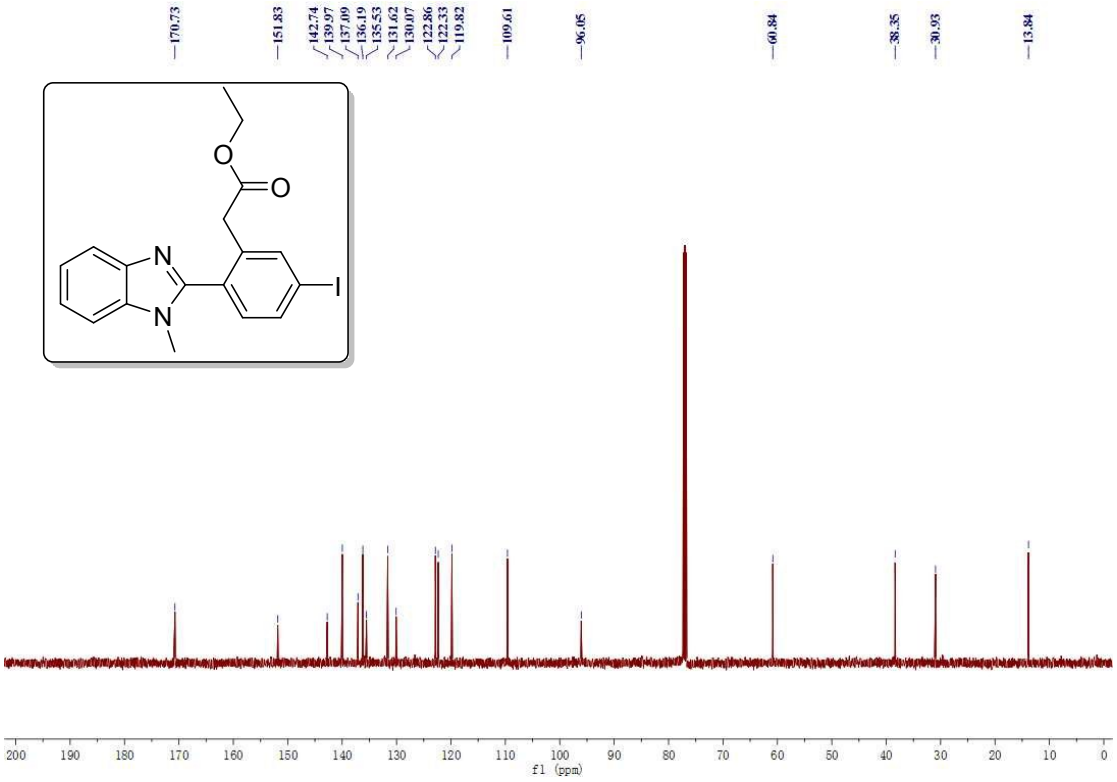


methyl 2-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4j)

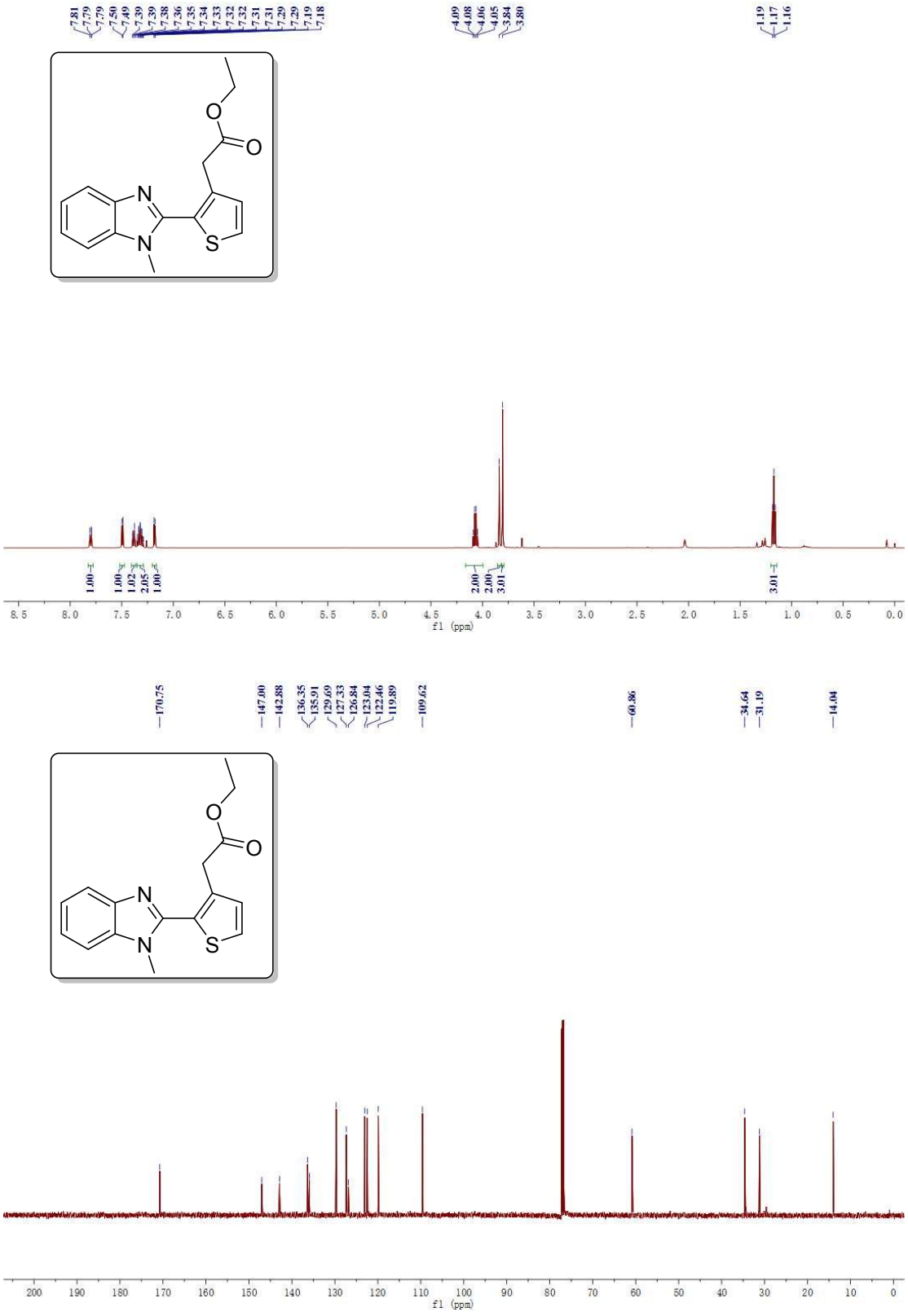


ethyl 2-(5-iodo-2-(1-methyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4k)

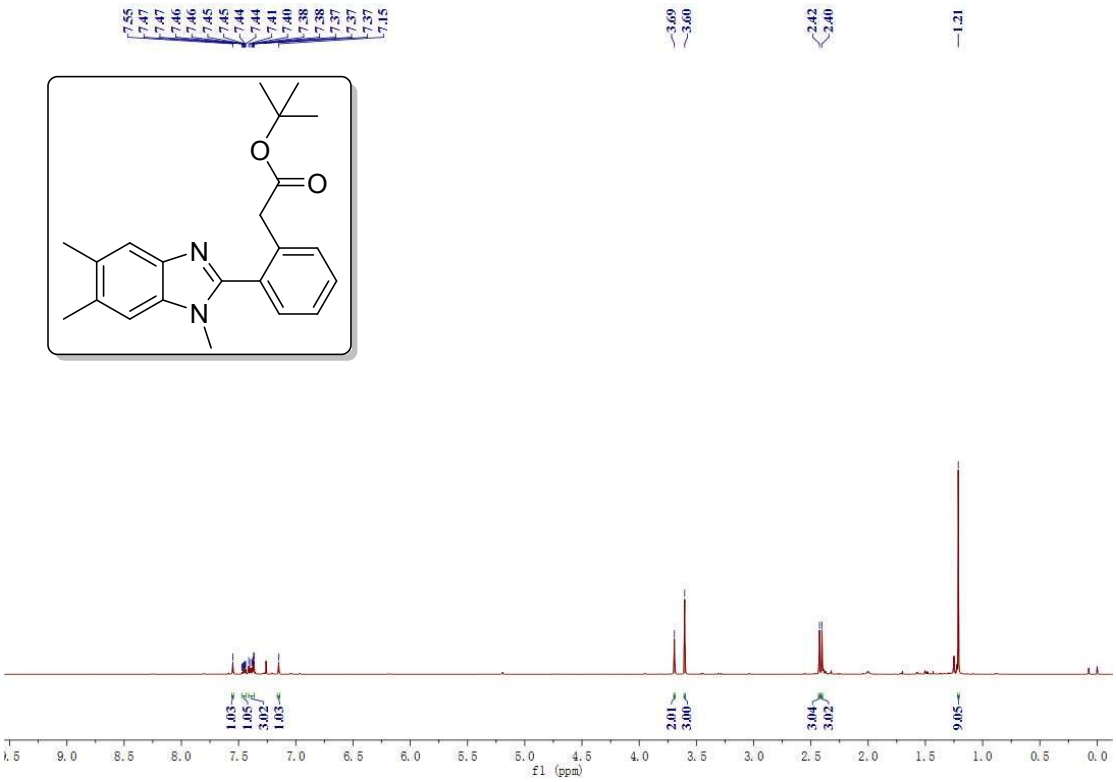


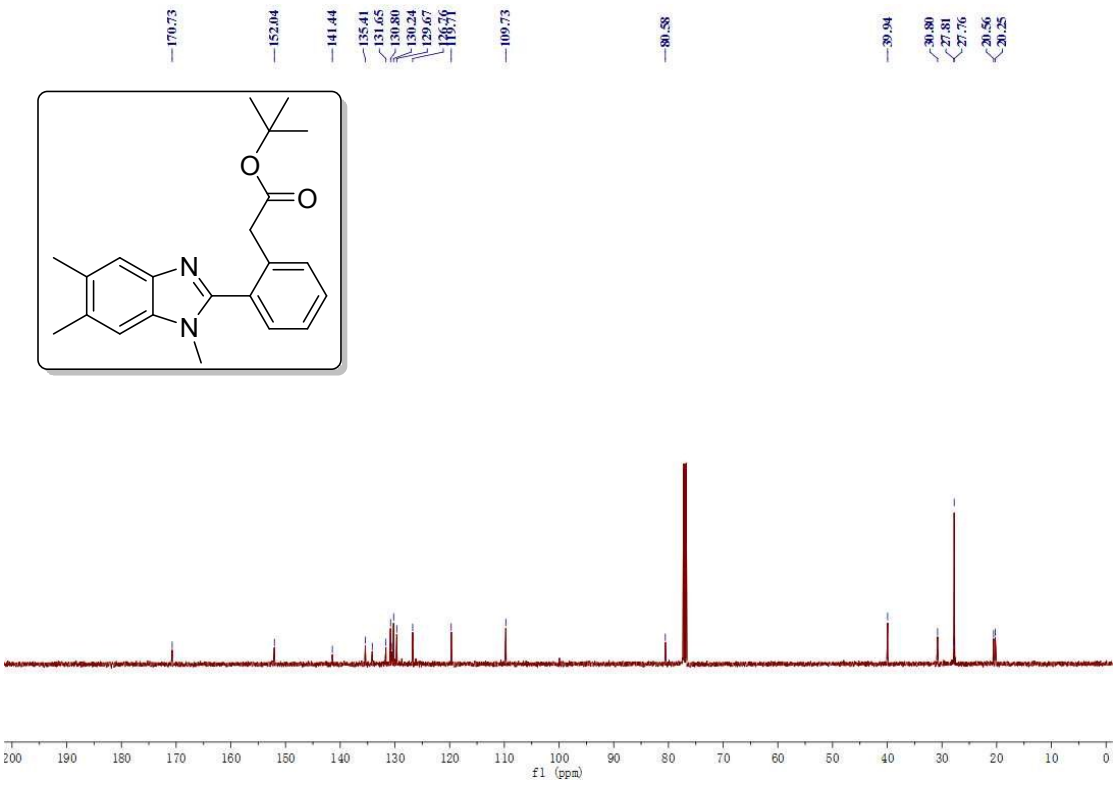


ethyl 2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)thiophen-3-yl)acetate (4l)

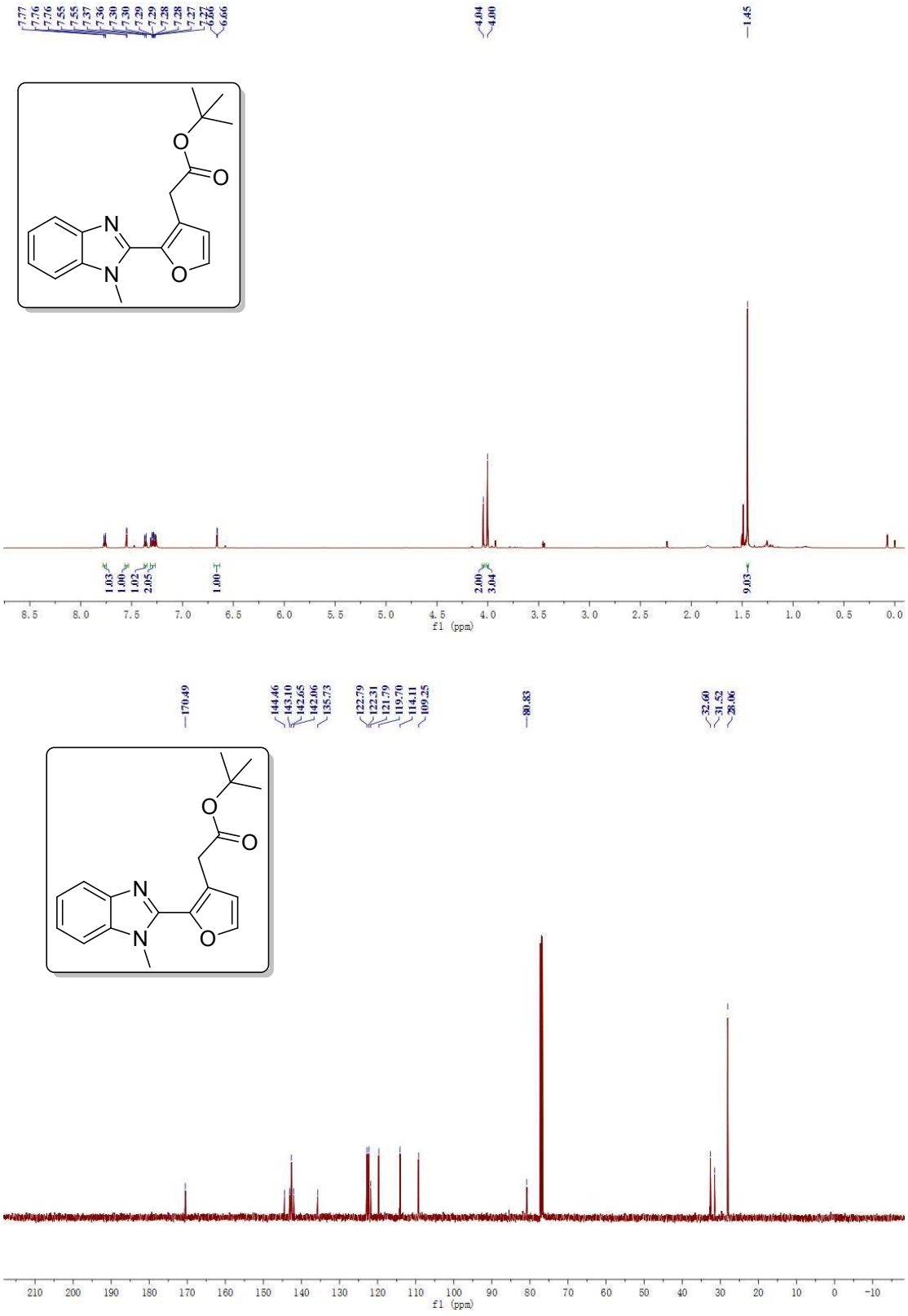


tert-butyl 2-(2-(1,5,6-trimethyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4m)

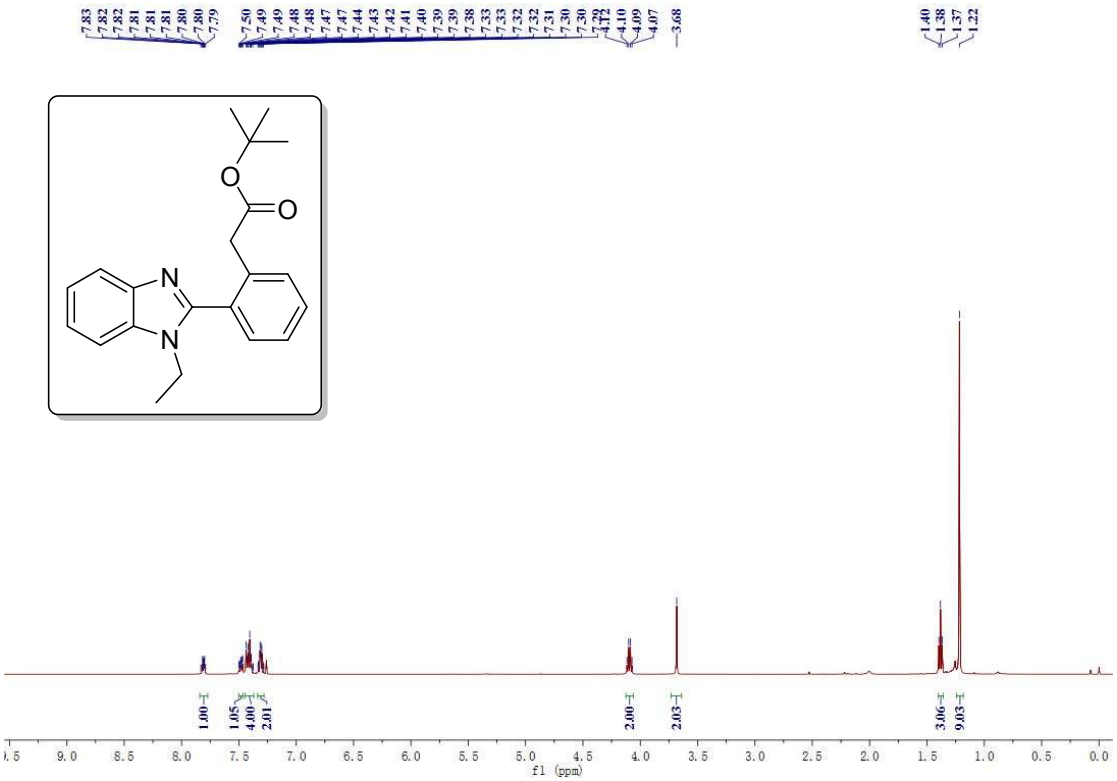


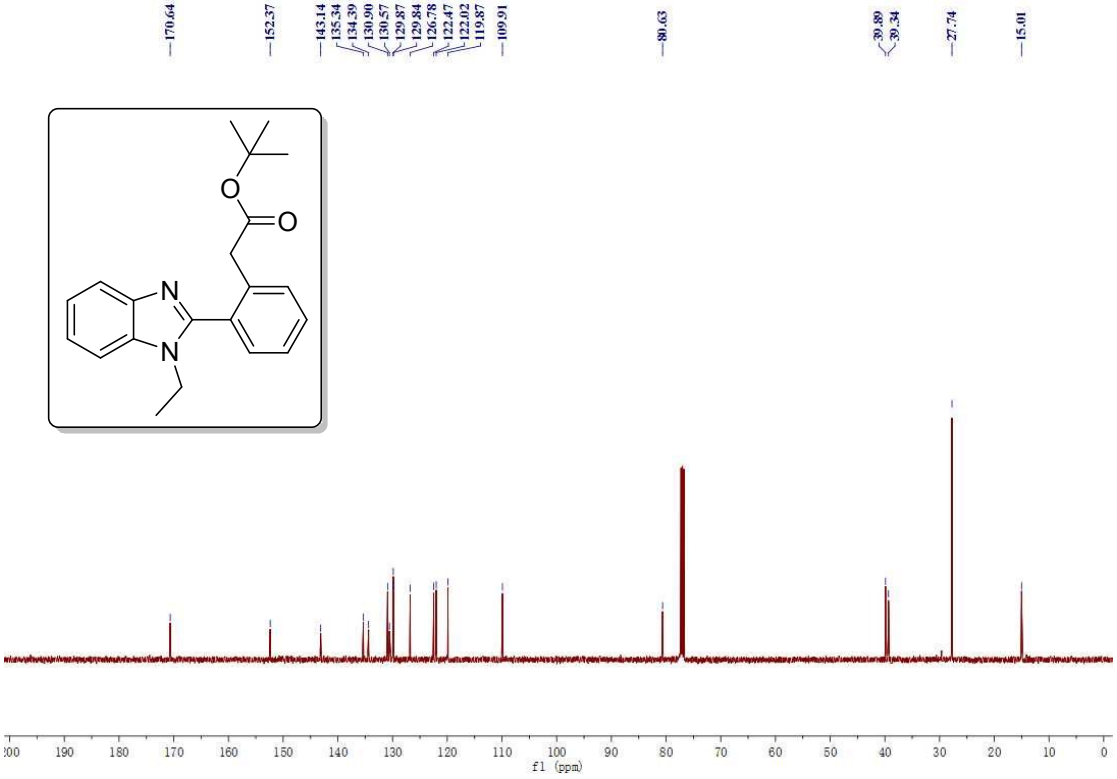


tert-butyl 2-(2-(1-methyl-1H-benzo[d]imidazol-2-yl)furan-3-yl)acetate (4n)

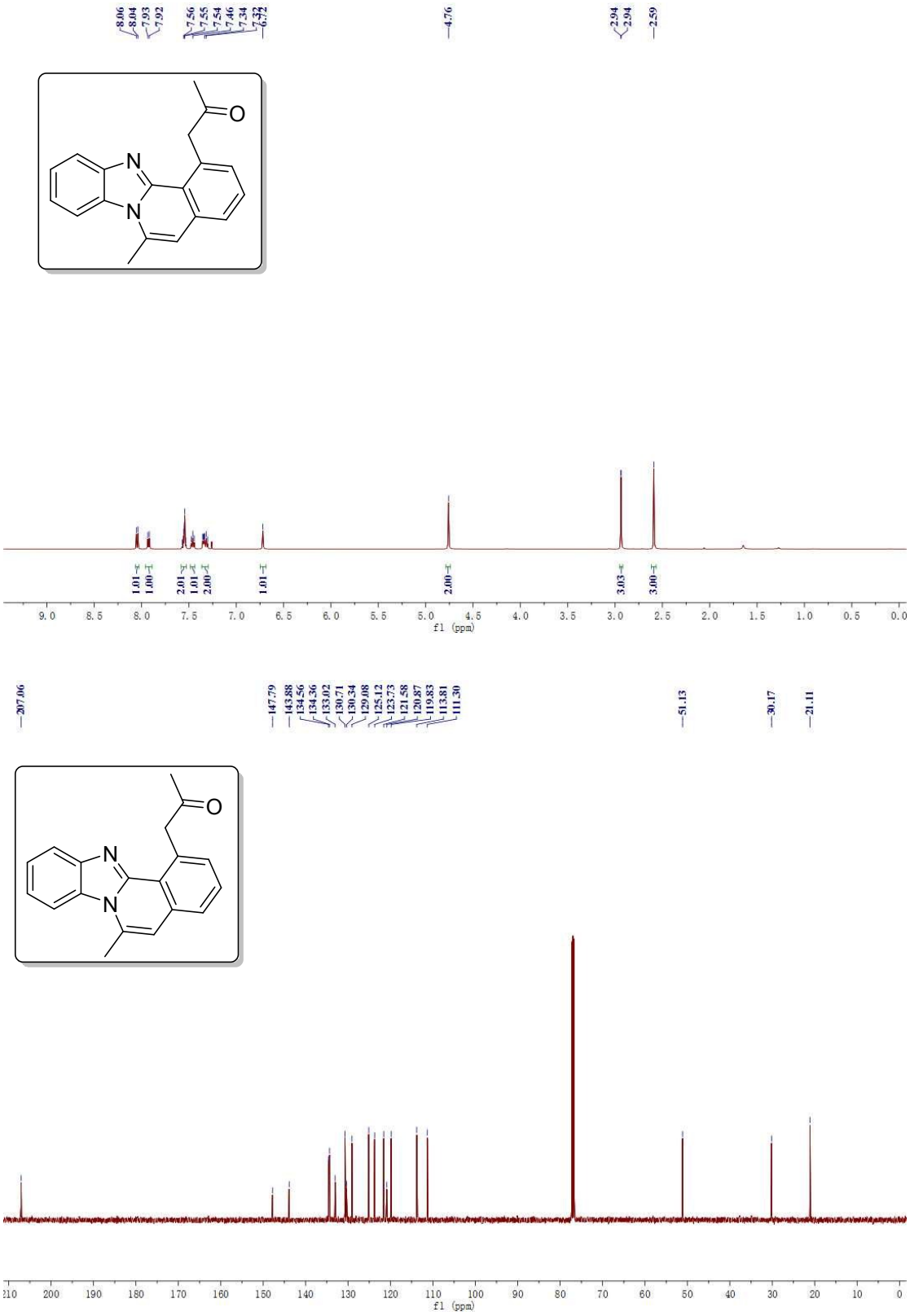


tert-butyl 2-(2-(1-ethyl-1H-benzo[d]imidazol-2-yl)phenyl)acetate (4o)

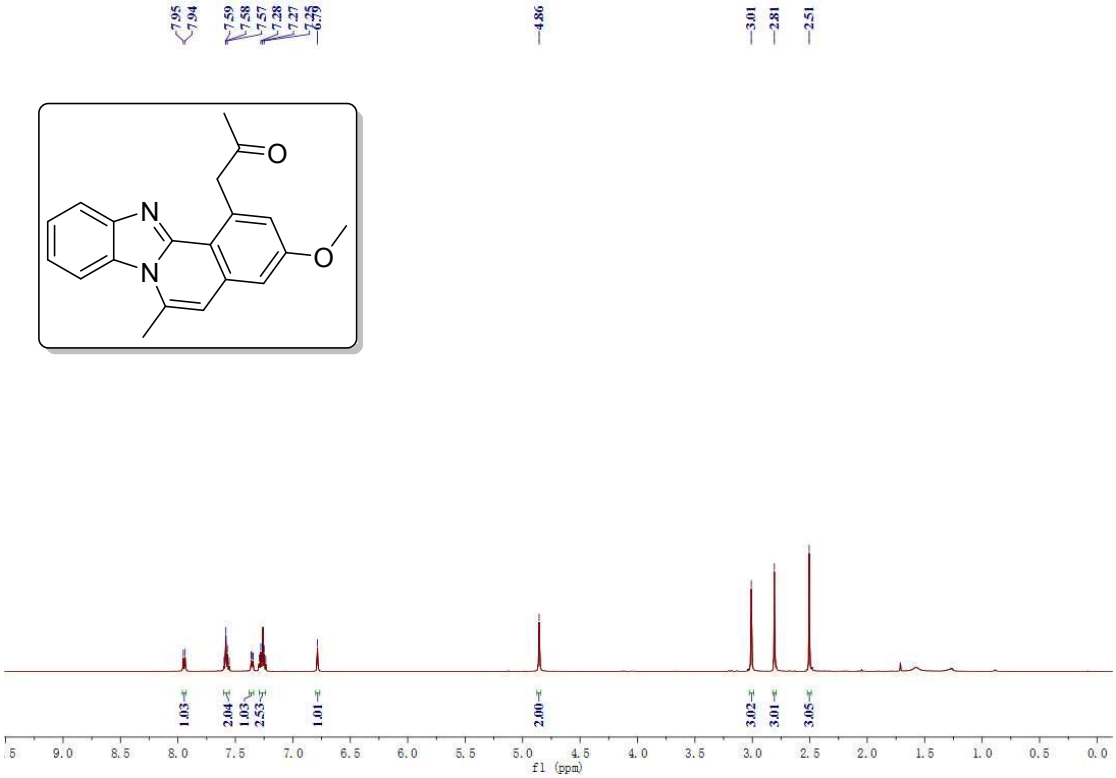


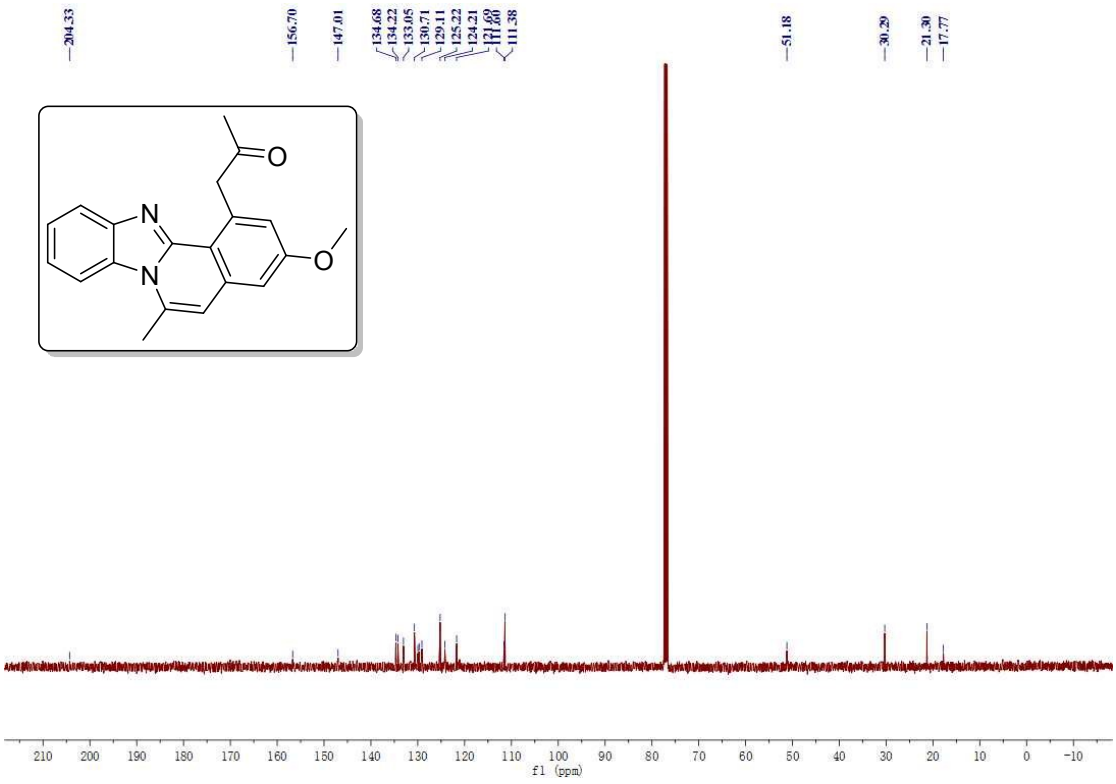


1-(6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6a)

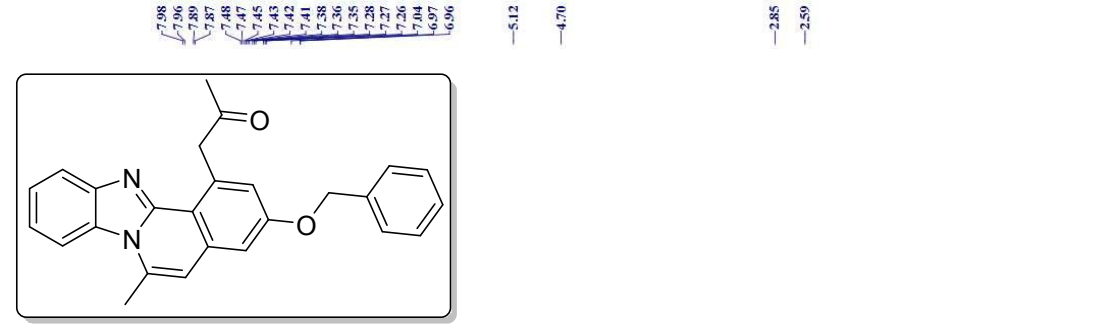


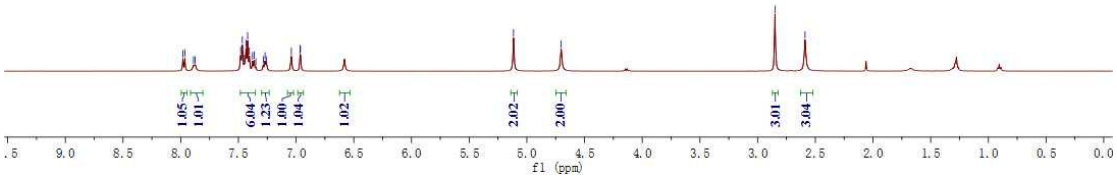
1-(3-methoxy-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6b)

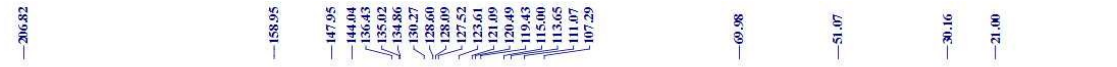


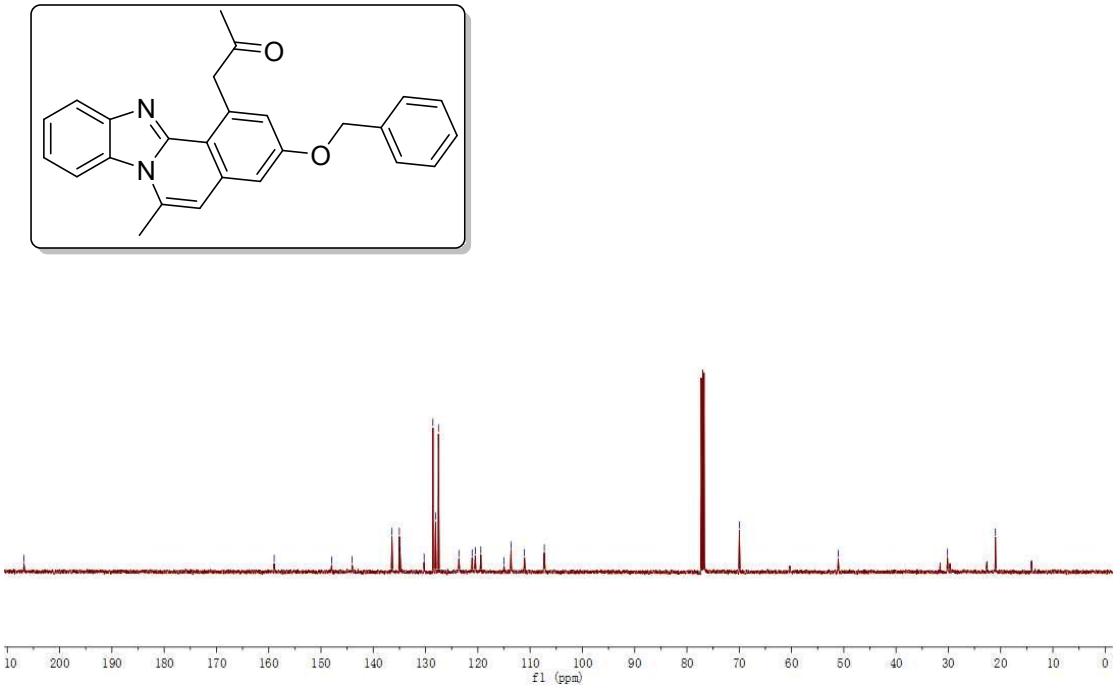


1-(3-(benzyloxy)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6c)

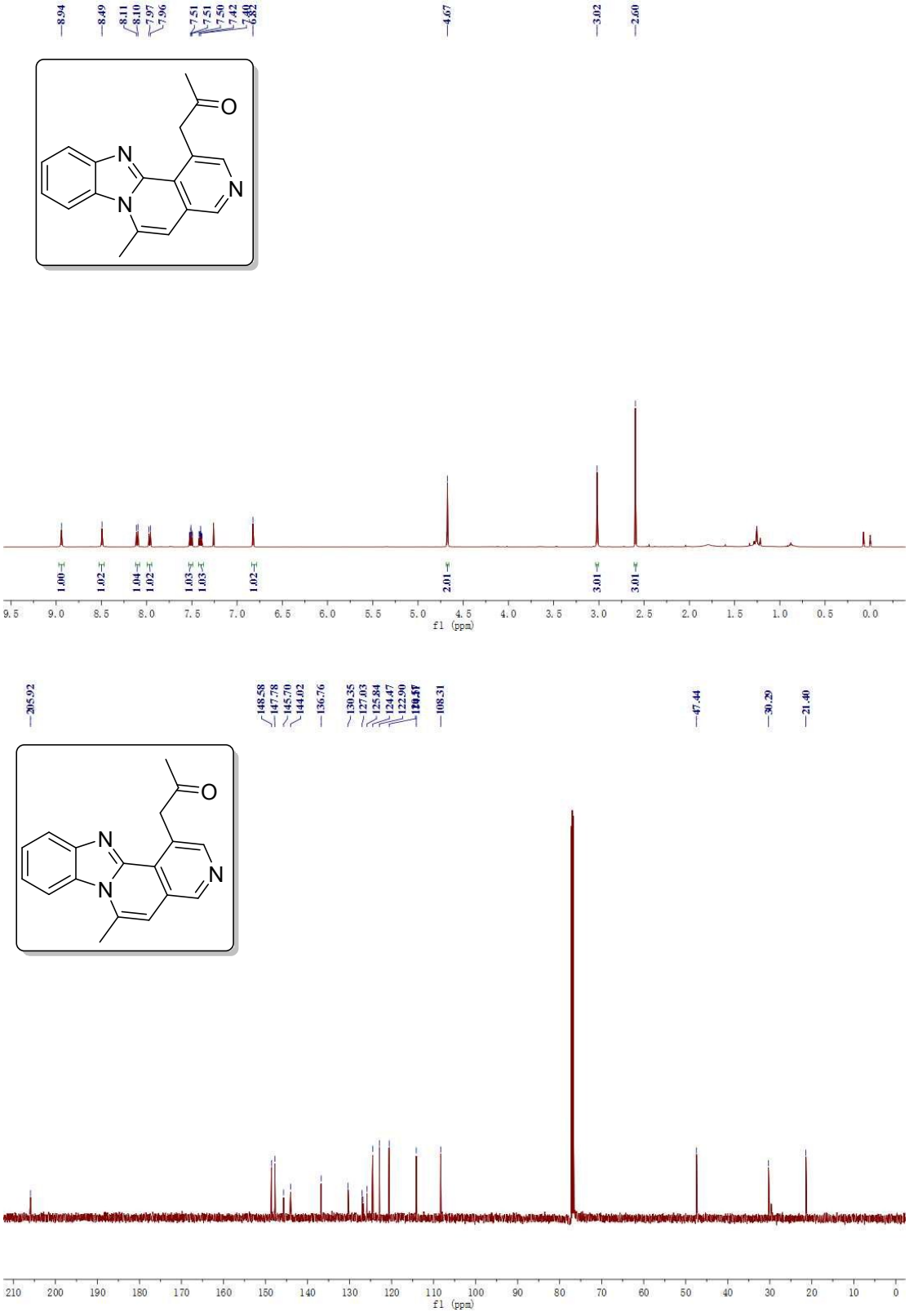




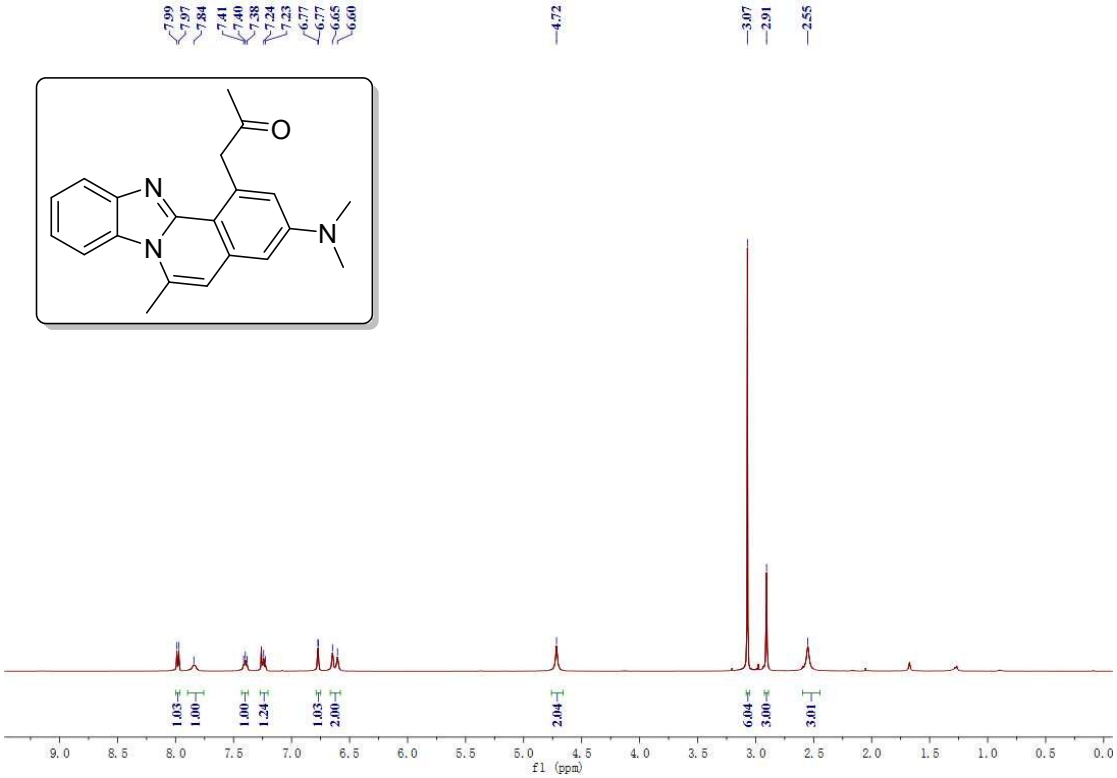


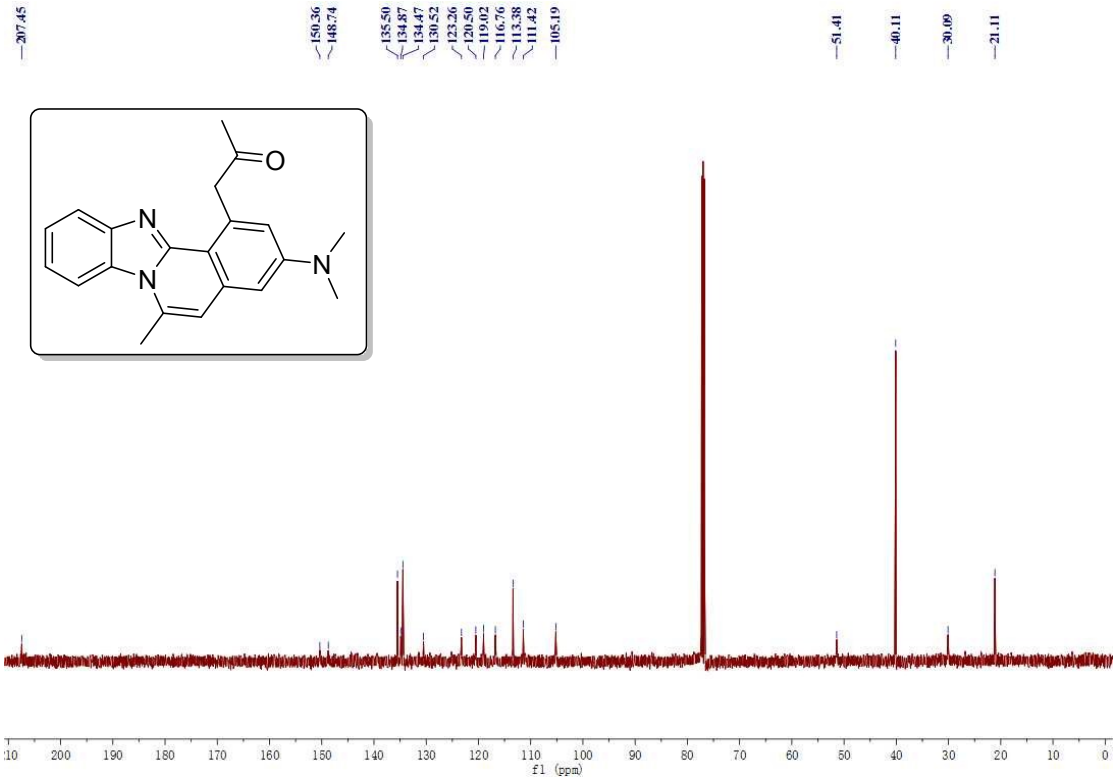


1-(6-methylbenzo[4,5]imidazo[2,1-a][2,6]naphthyridin-1-yl)propan-2-one (6d)

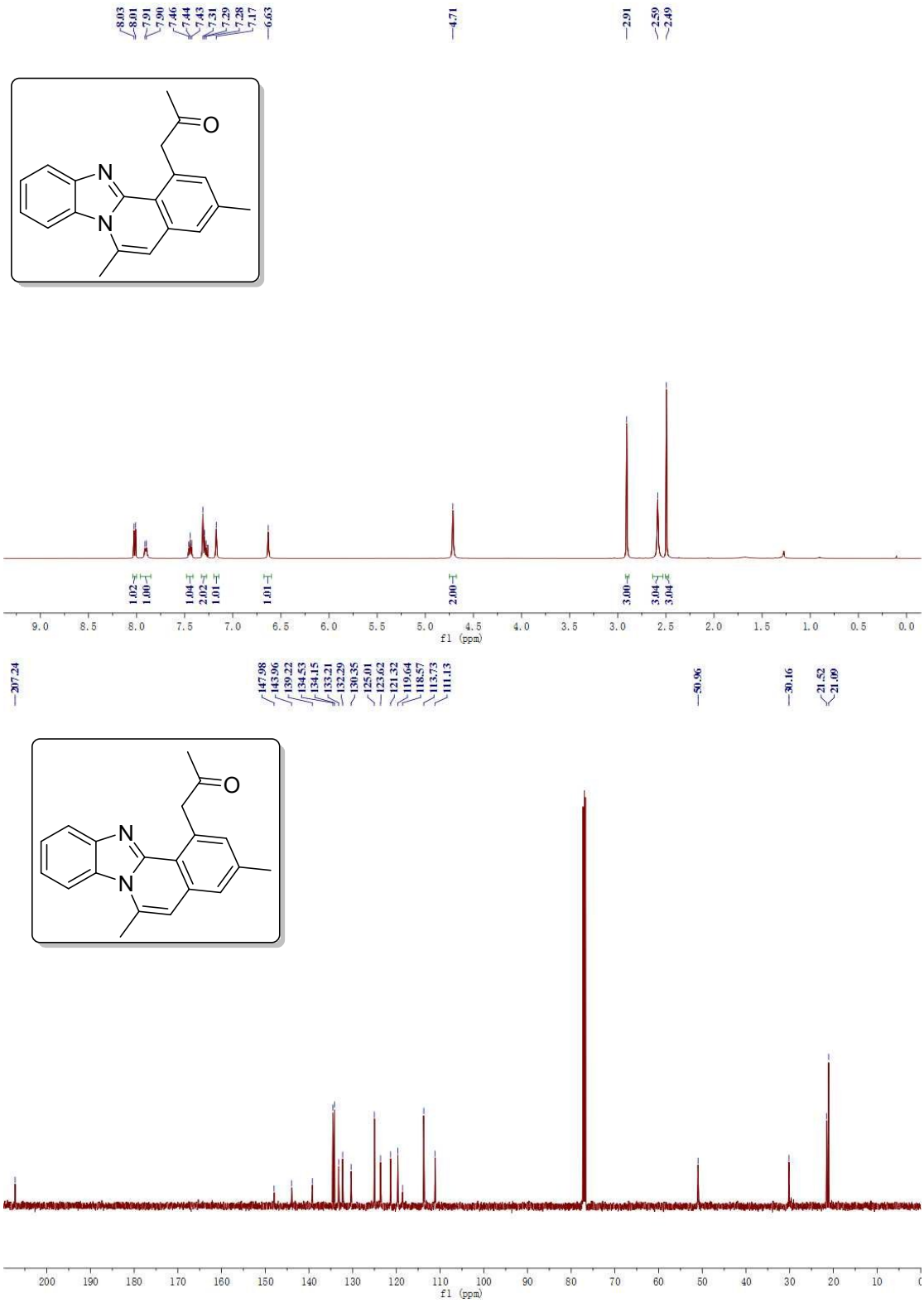


1-(3-(dimethylamino)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6e)

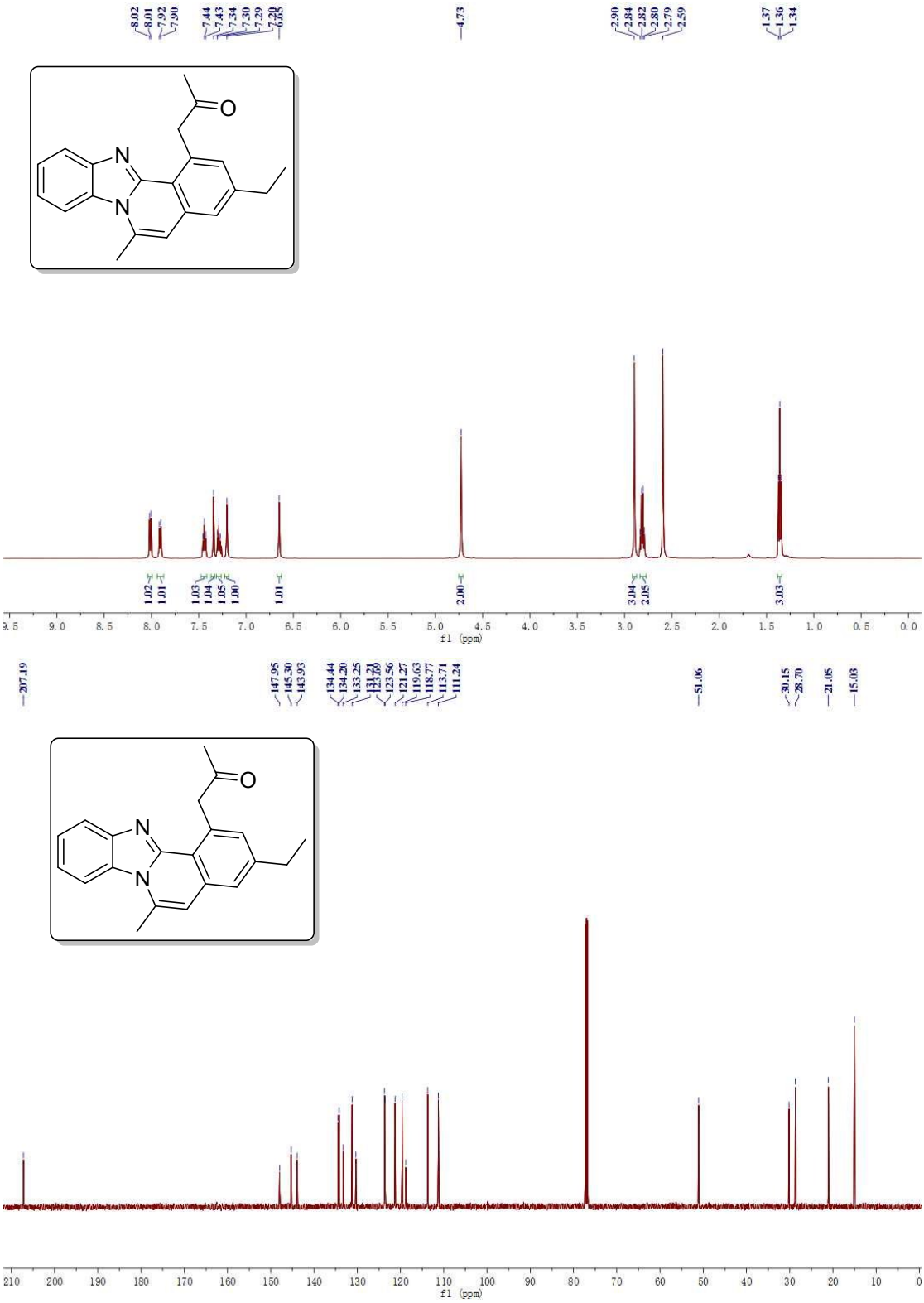




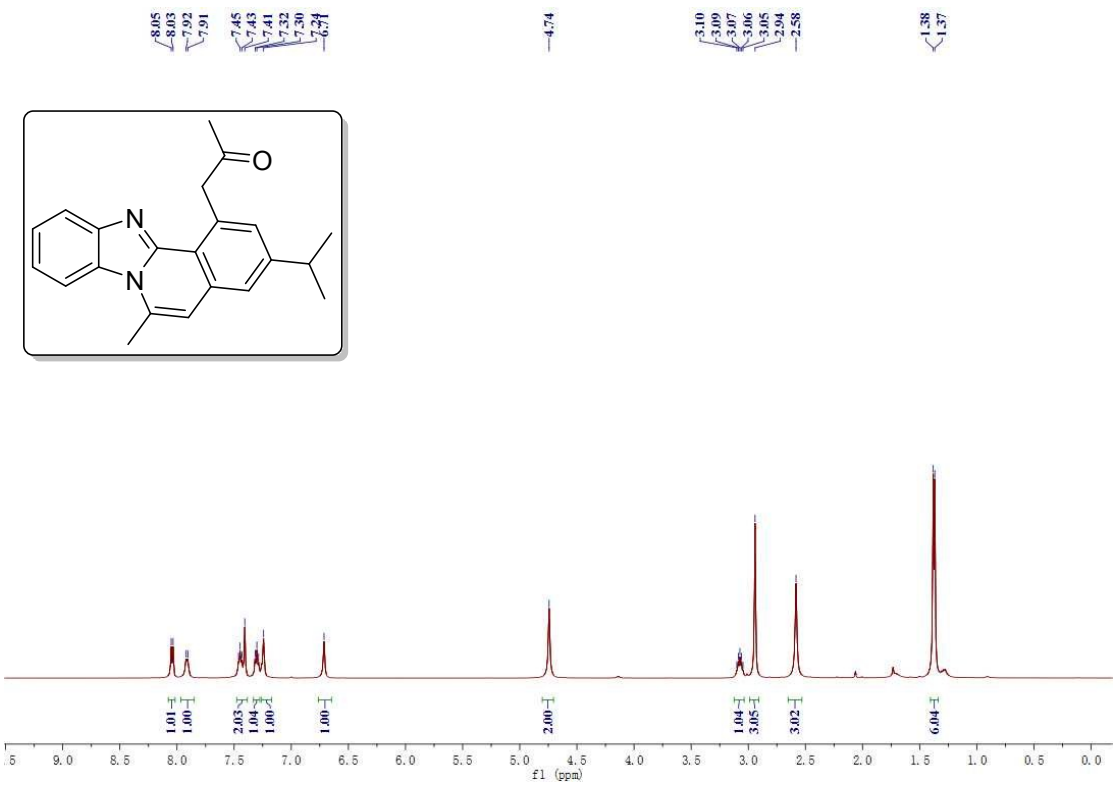
1-(3,6-dimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6f)

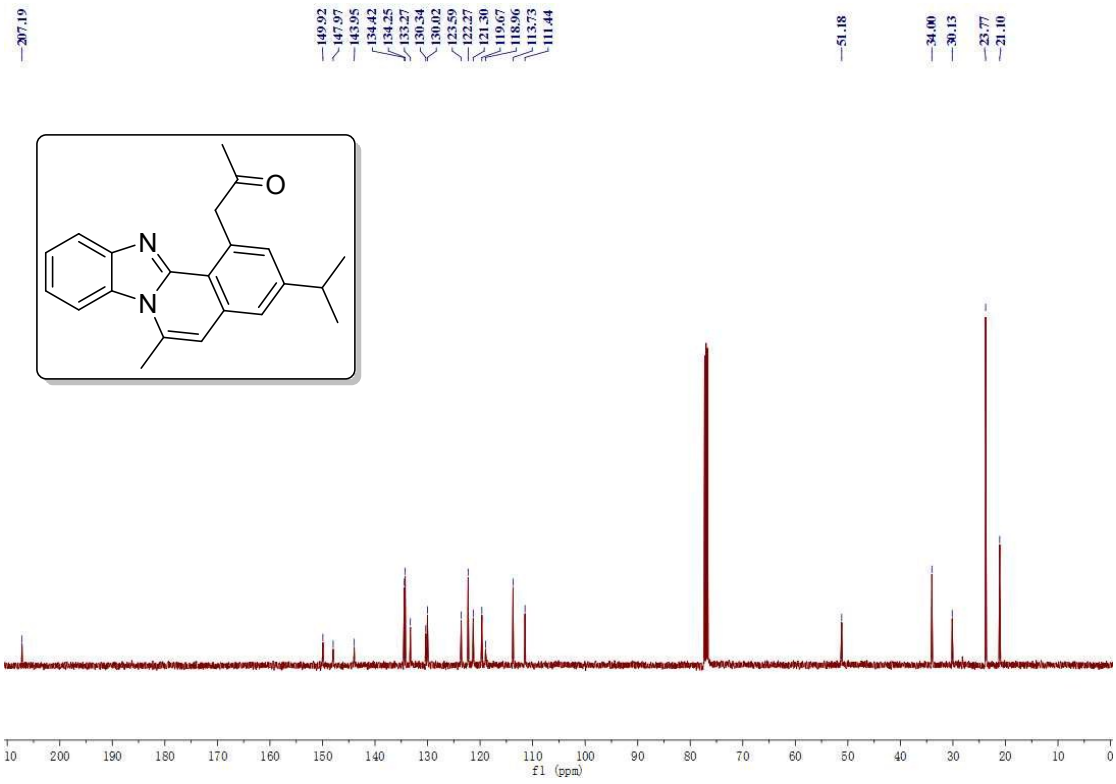


1-(3-ethyl-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6g)

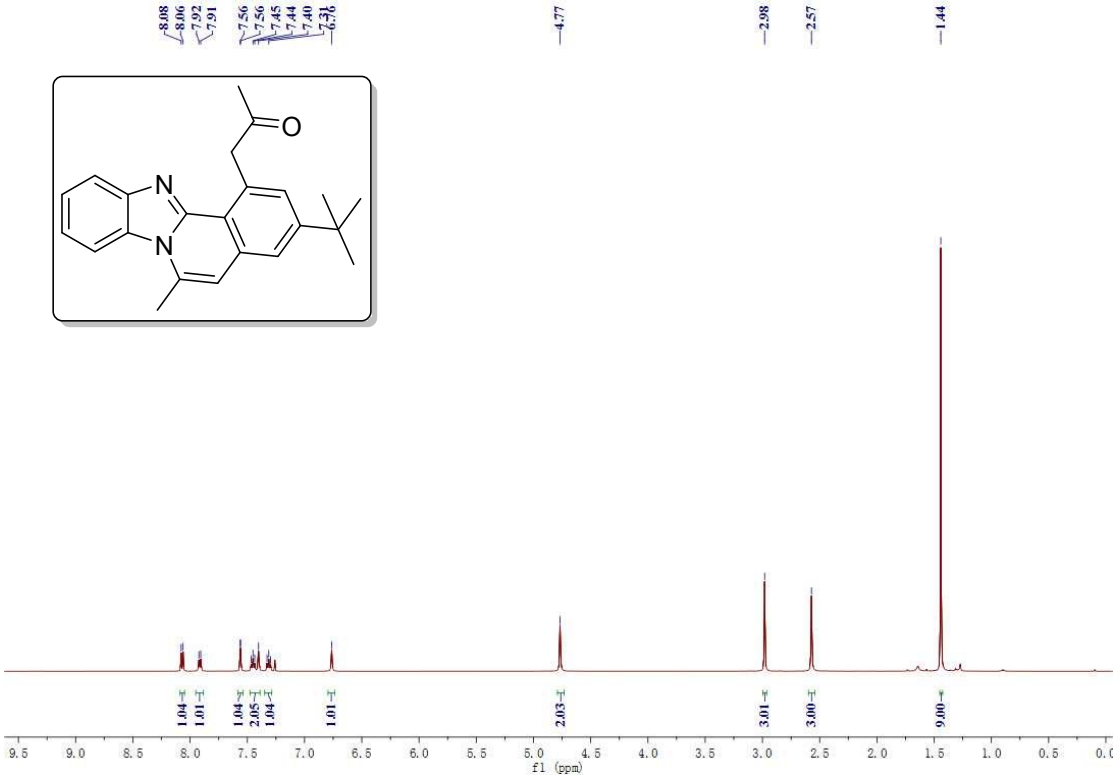


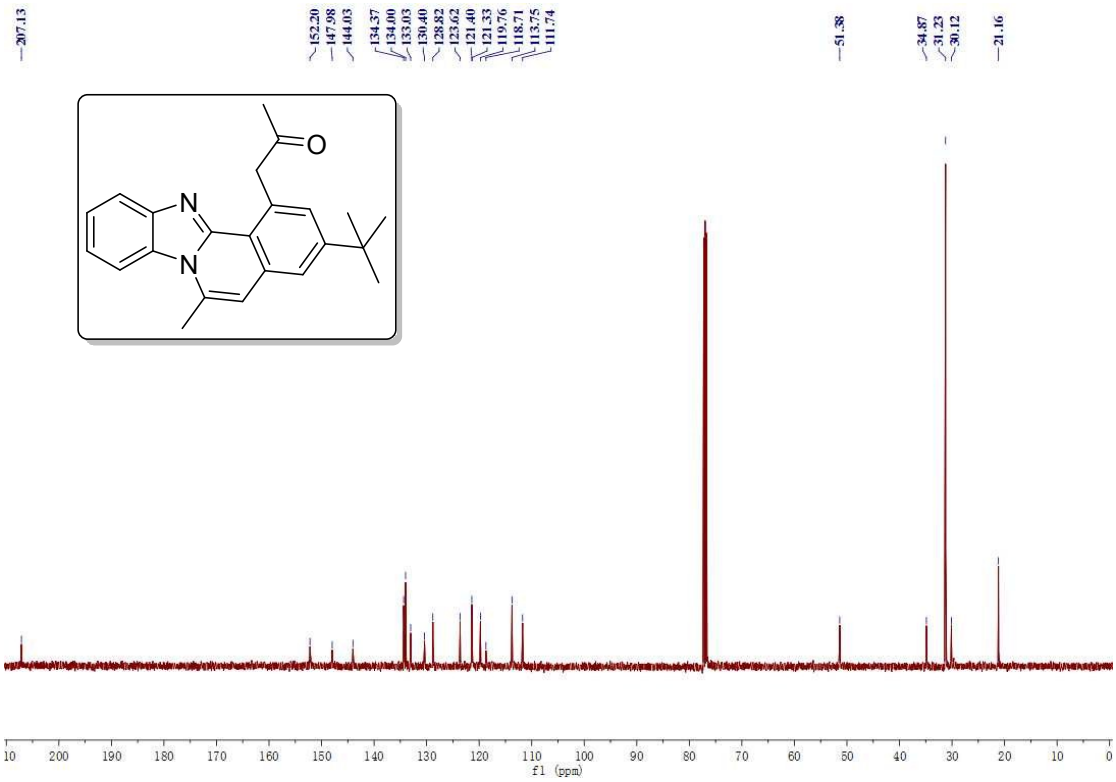
1-(3-isopropyl-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6h)





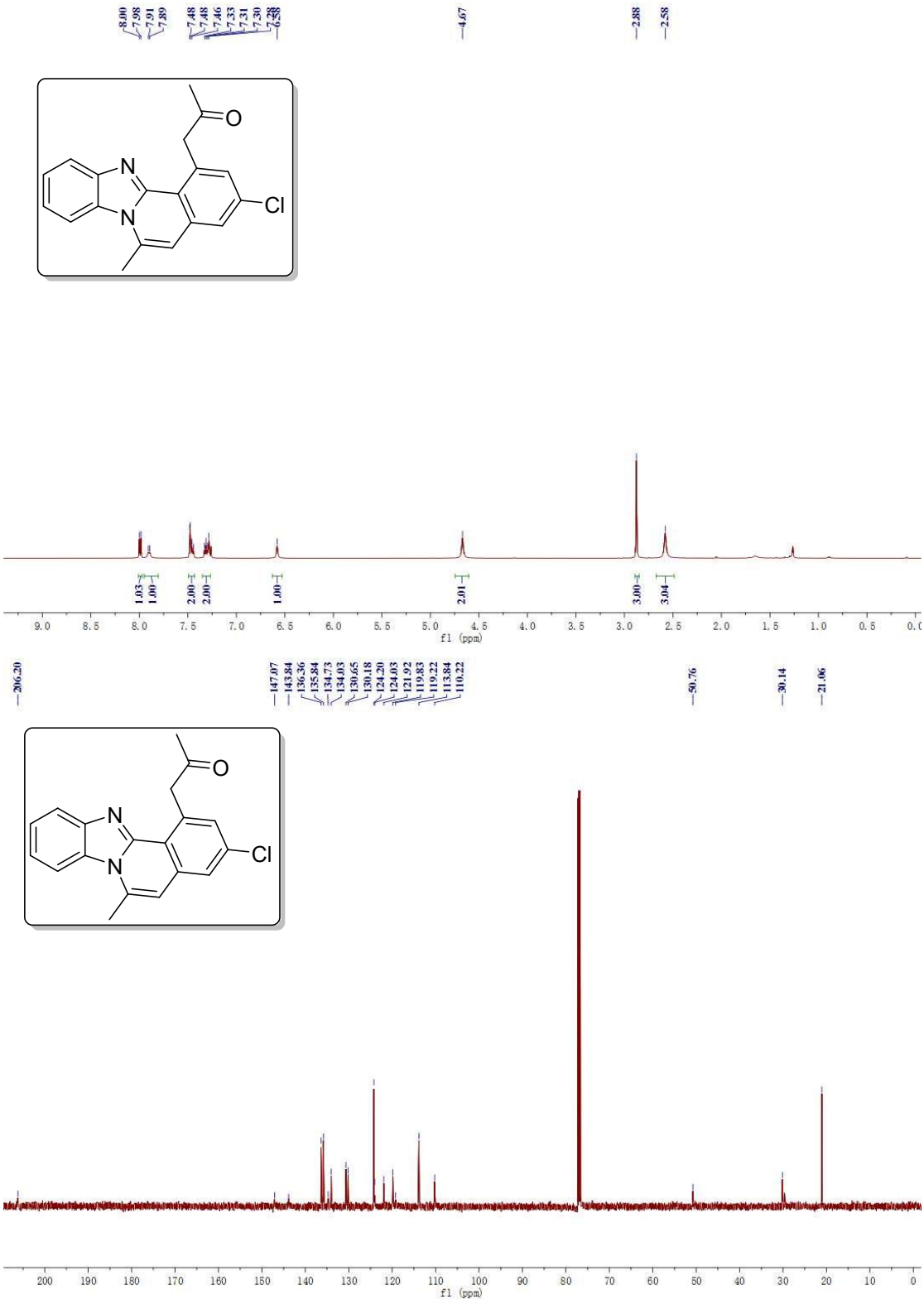
1-(3-(tert-butyl)-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6i)



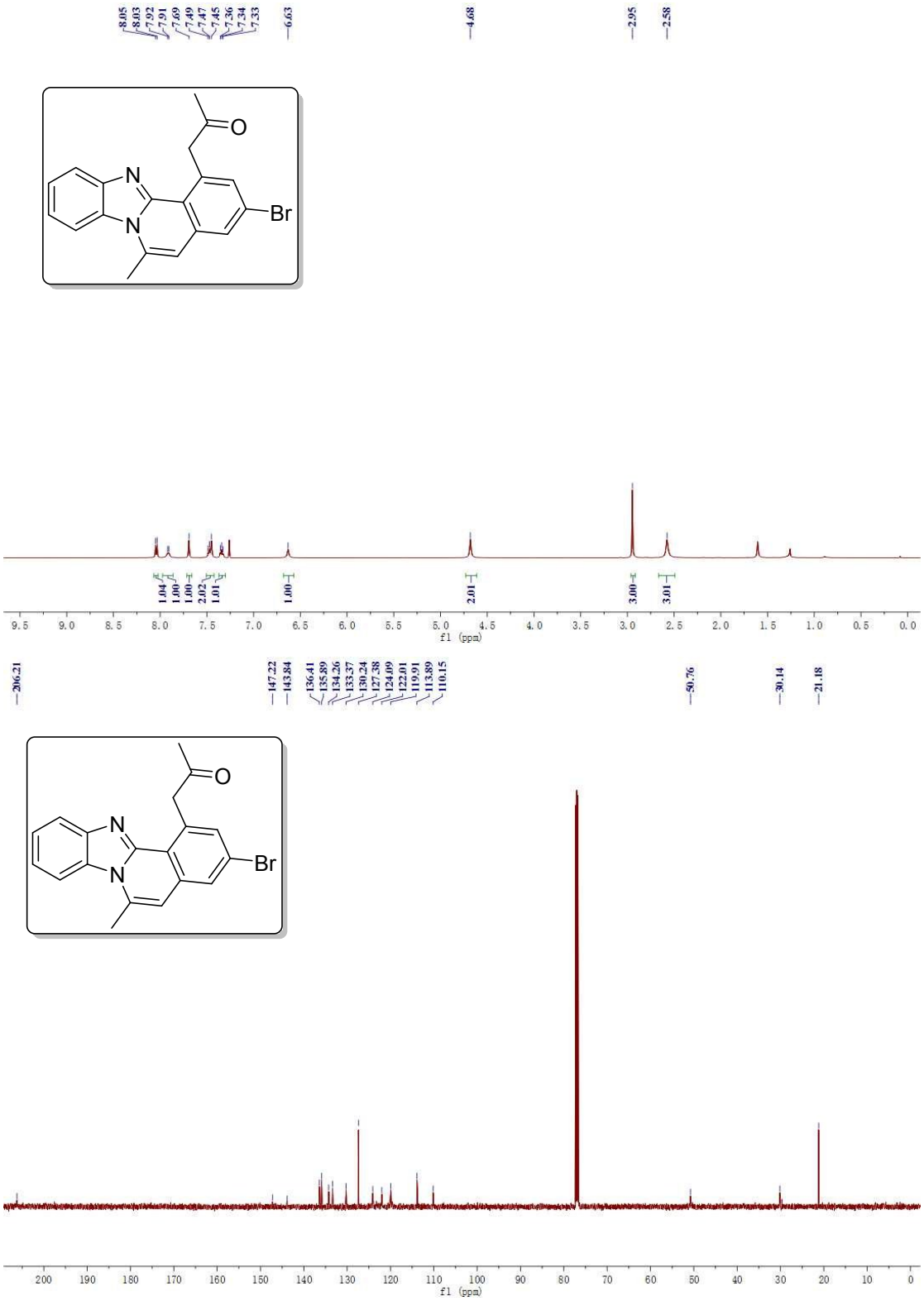


1-(3-fluoro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6j)

1-(3-chloro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6k)

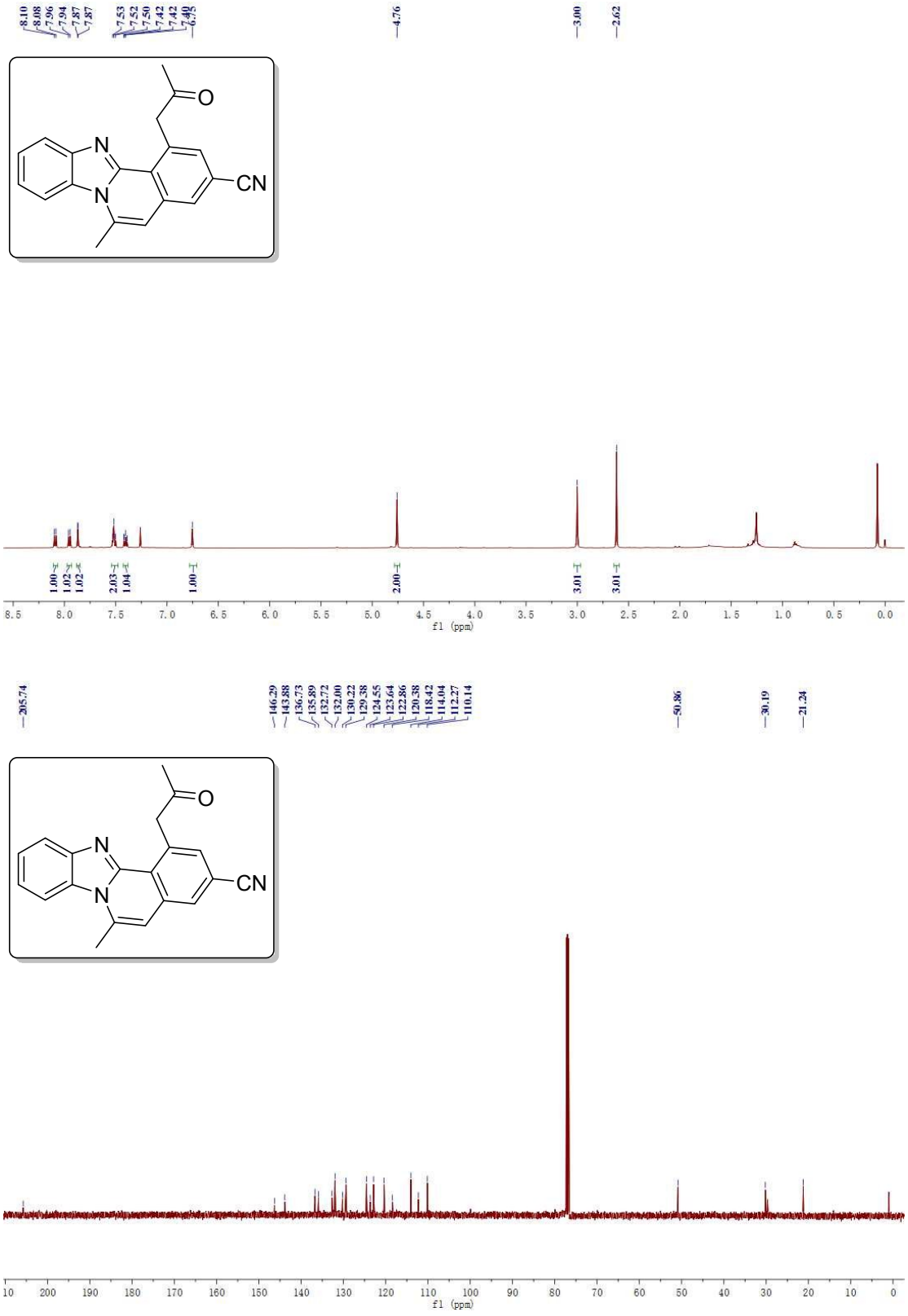


1-(3-bromo-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6l)

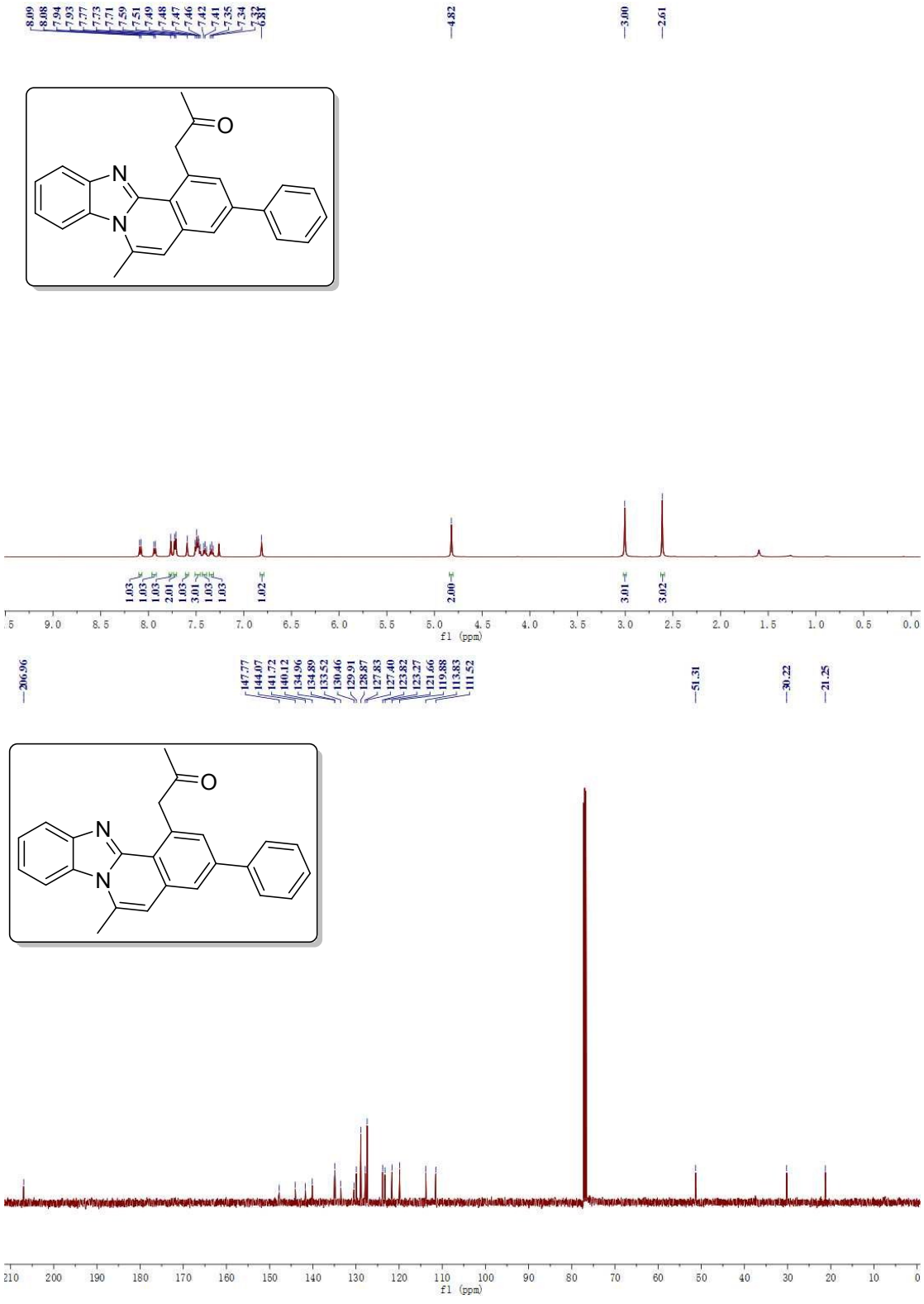


1-(3-iodo-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6m)

6-methyl-1-(2-oxopropyl)benzo[4,5]imidazo[2,1-a]isoquinoline-3-carbonitrile (6n)



1-(6-methyl-3-phenylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6o)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7.94
7.93
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7.73
7.71
7.59
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7.35
7.34
7.32
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3.00
2.61
O
N
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1.03
1.03
2.01
1.03
3.01
1.03
1.03
1.02
2.00
3.01
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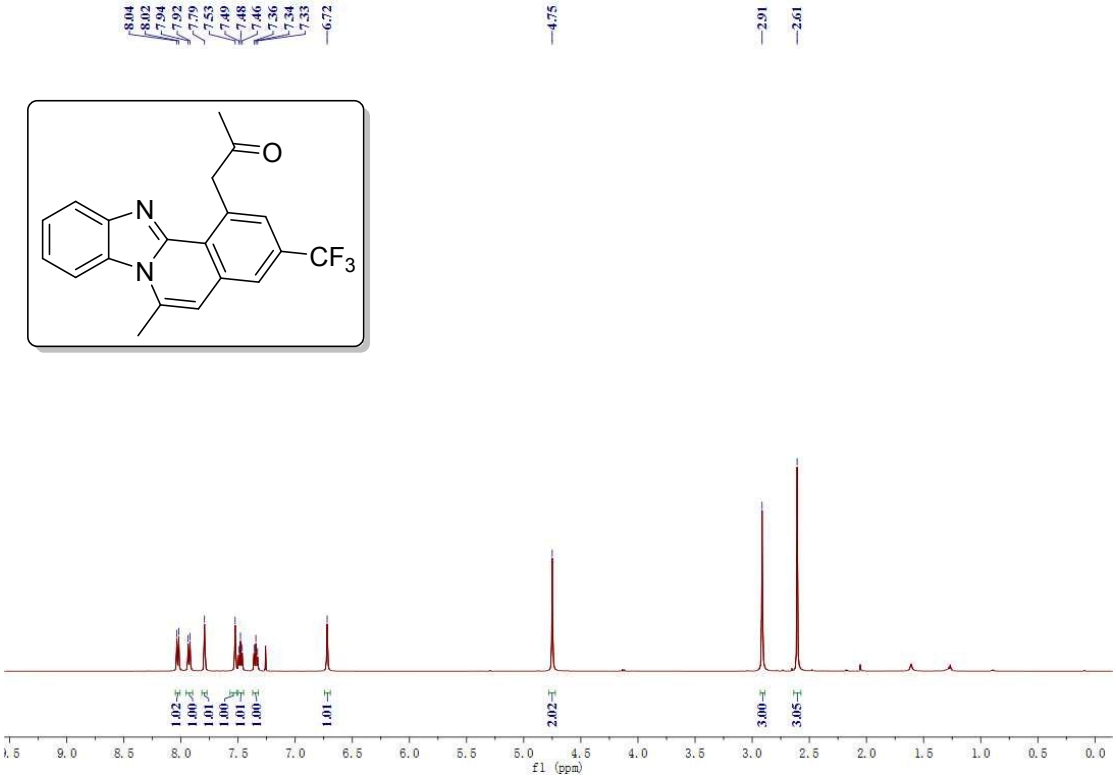
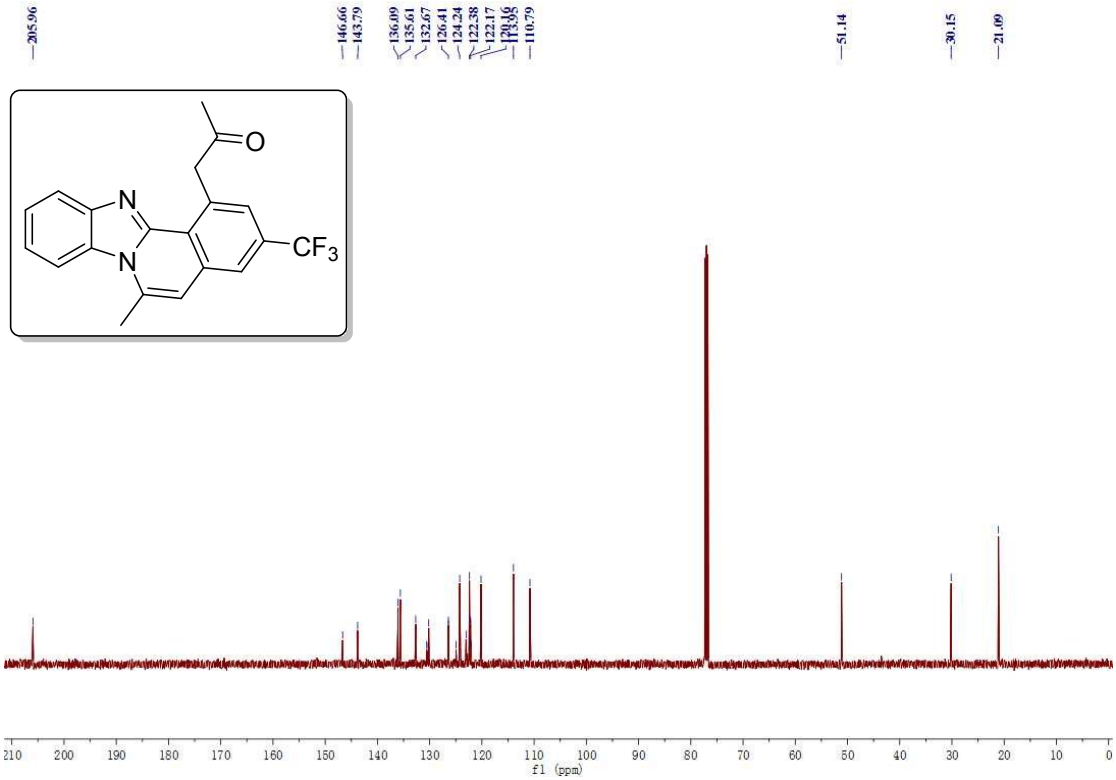
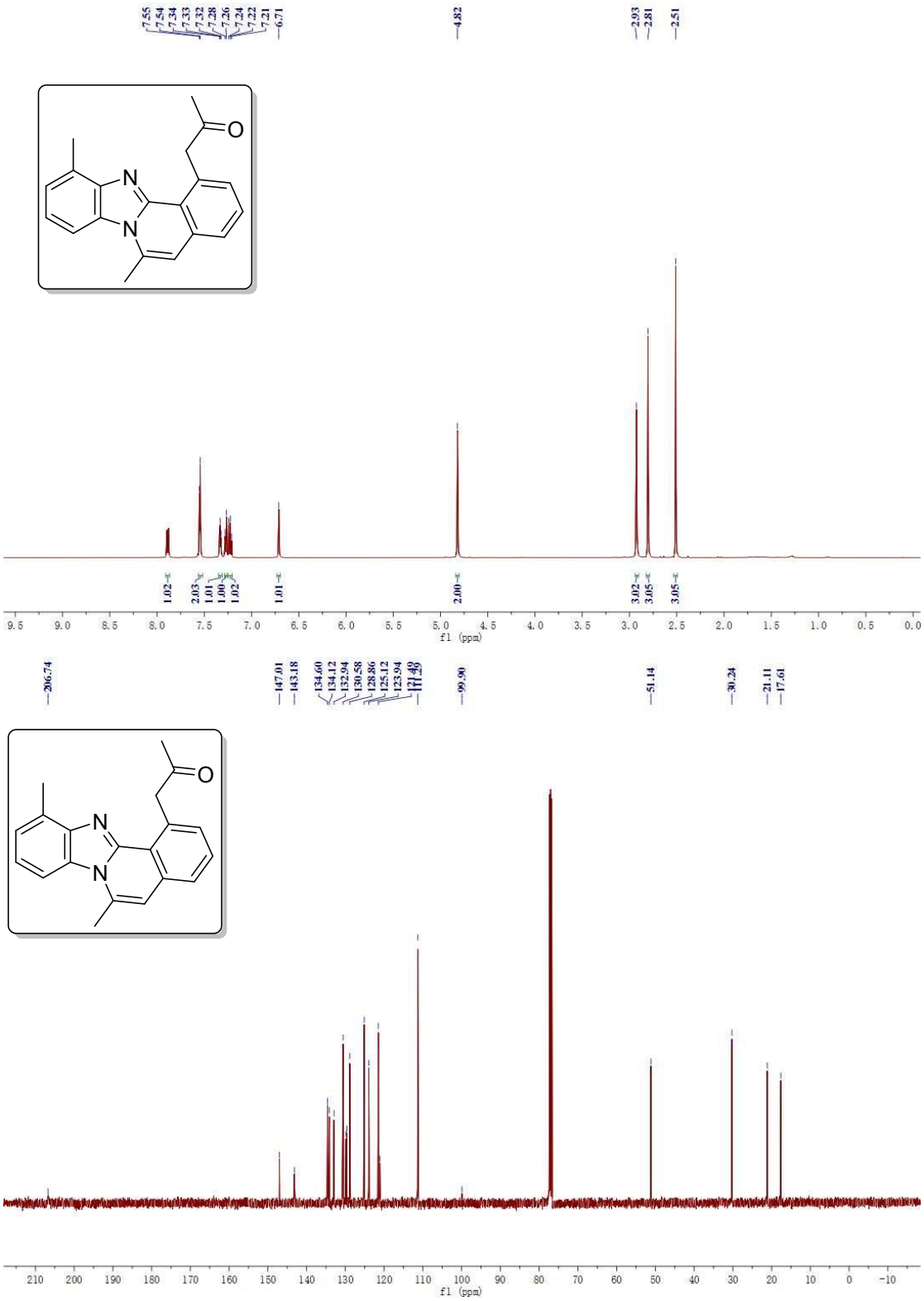
128.87
127.83
127.40
123.82
123.27
121.66
119.88
113.83
111.52
51.31
30.22
21.25
O
N
N
f1 (ppm)

1-(6-methyl-3-(trifluoromethyl)benzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6p)

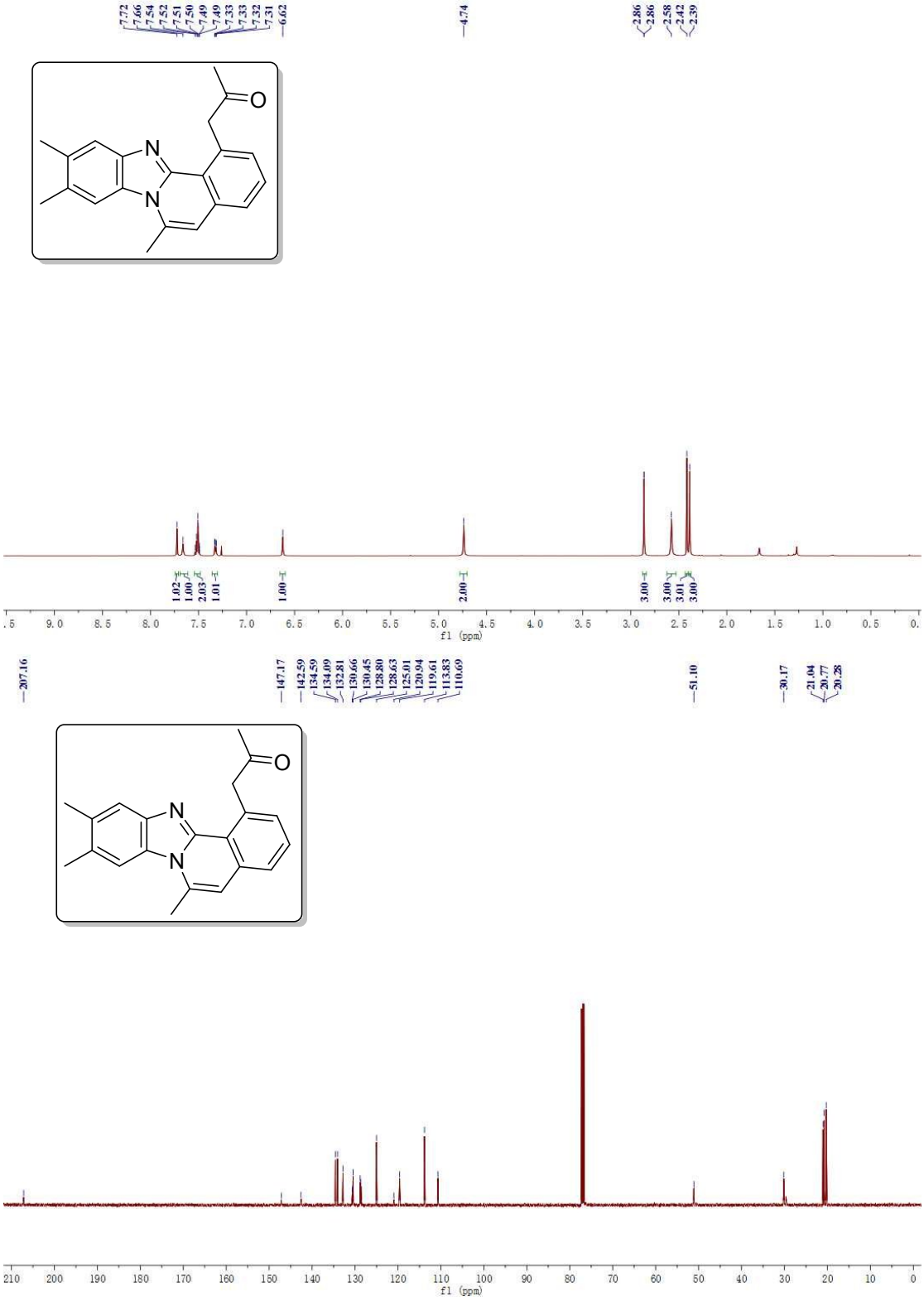




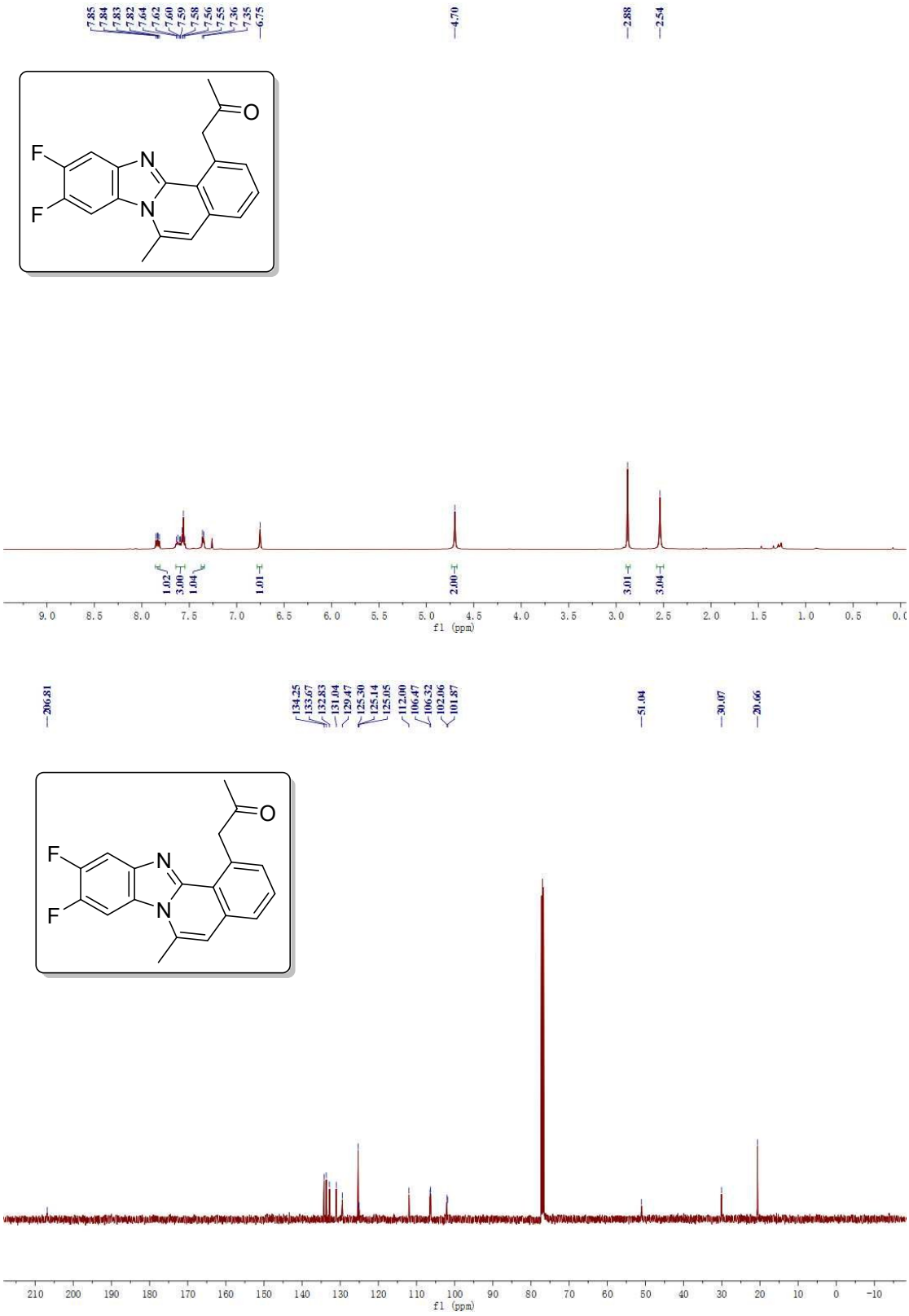
1-(6,11-dimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6q)



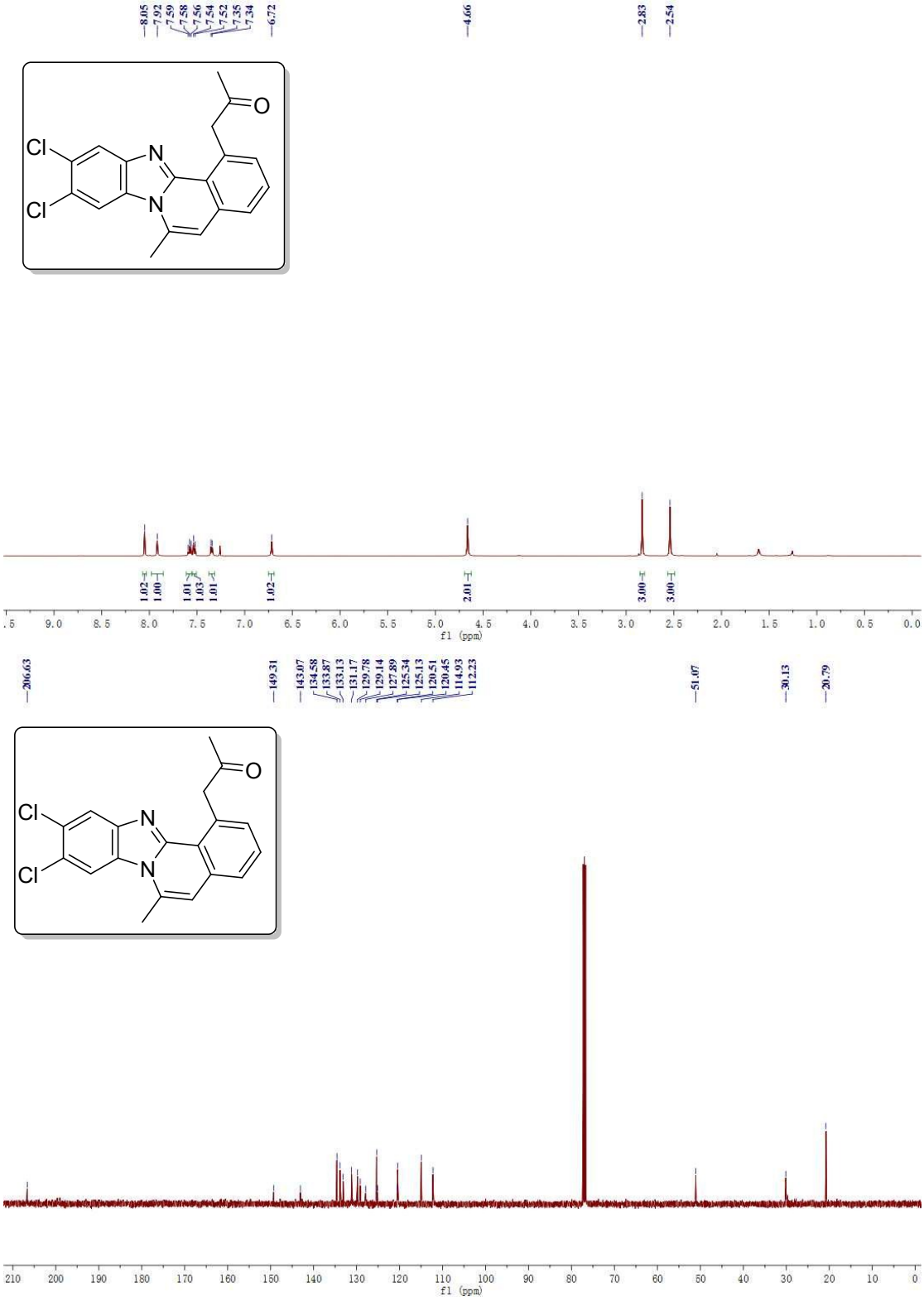
1-(6,9,10-trimethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6r)



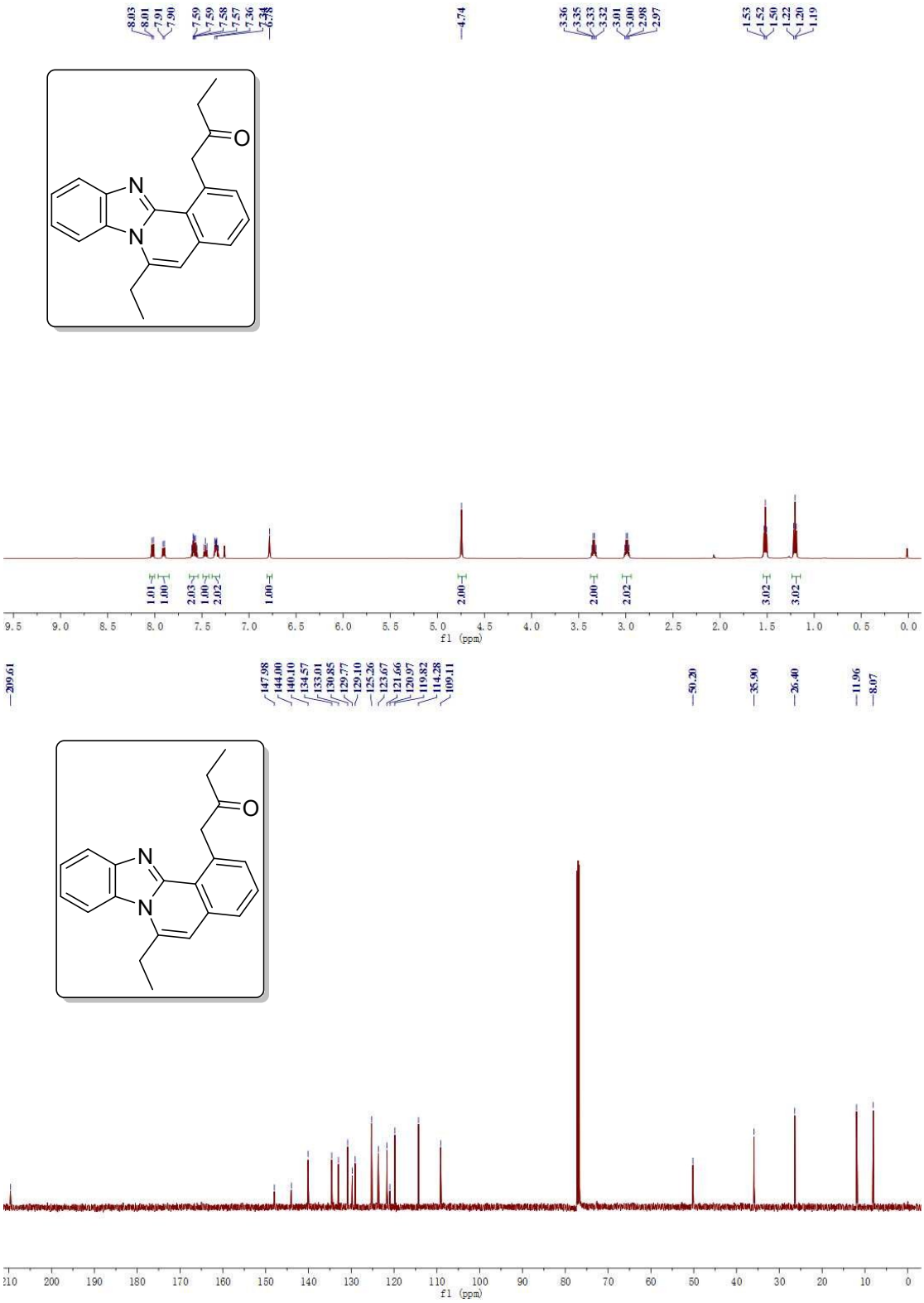
1-(9,10-difluoro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6s)



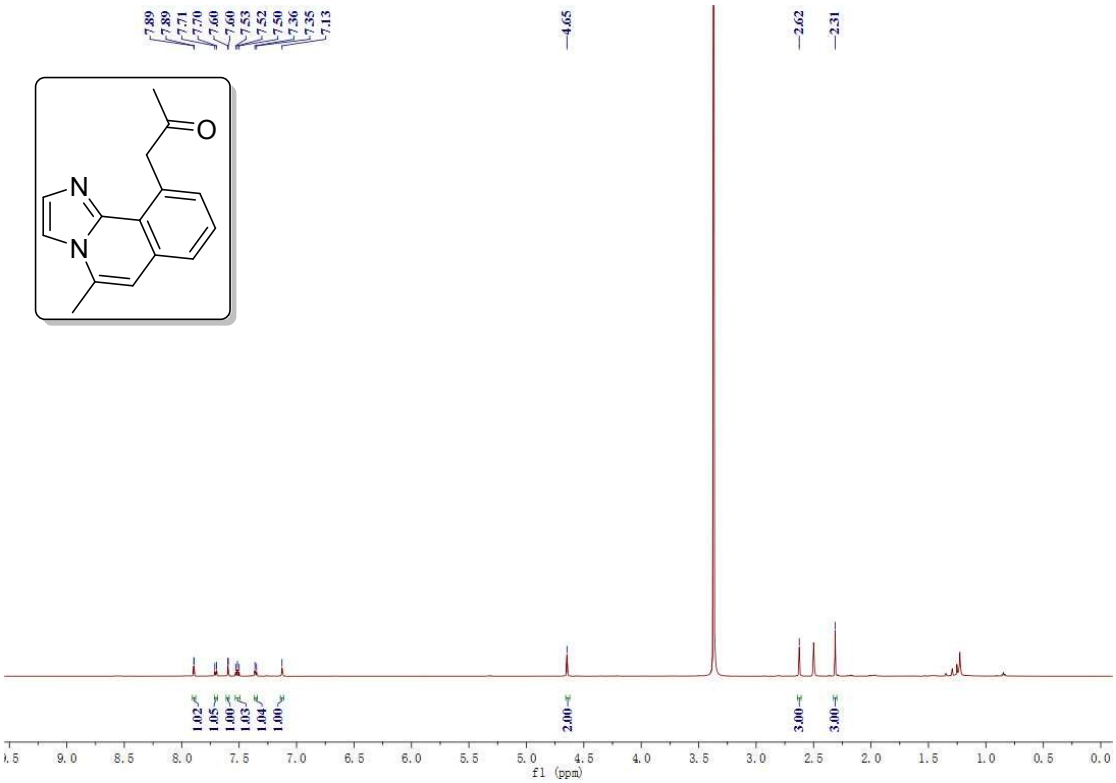
1-(9,10-dichloro-6-methylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)propan-2-one (6t)

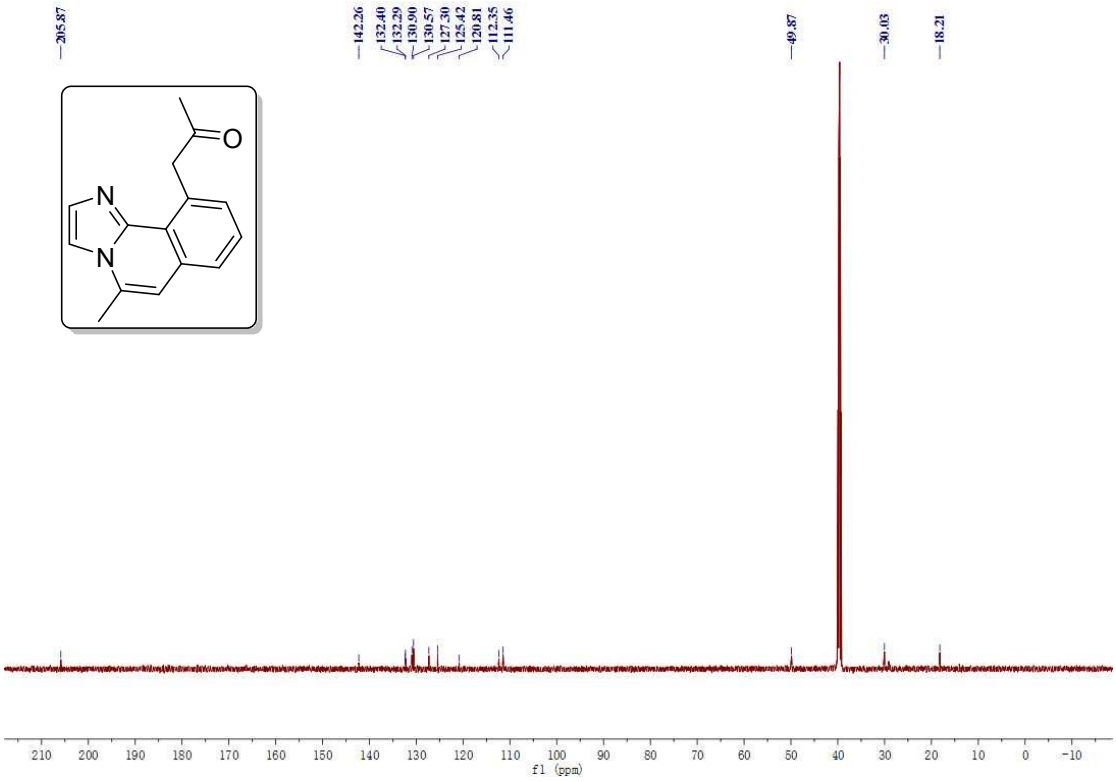


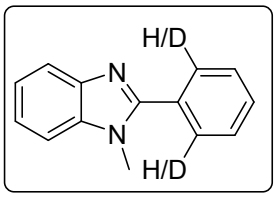
1-(6-ethylbenzo[4,5]imidazo[2,1-a]isoquinolin-1-yl)butan-2-one (6u)

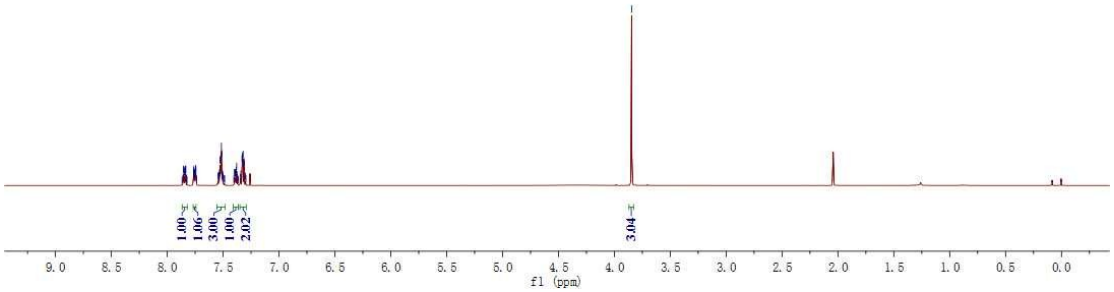


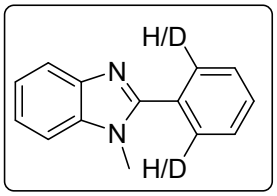
1-(5-methylimidazo[2,1-a]isoquinolin-10-yl)propan-2-one (6v)

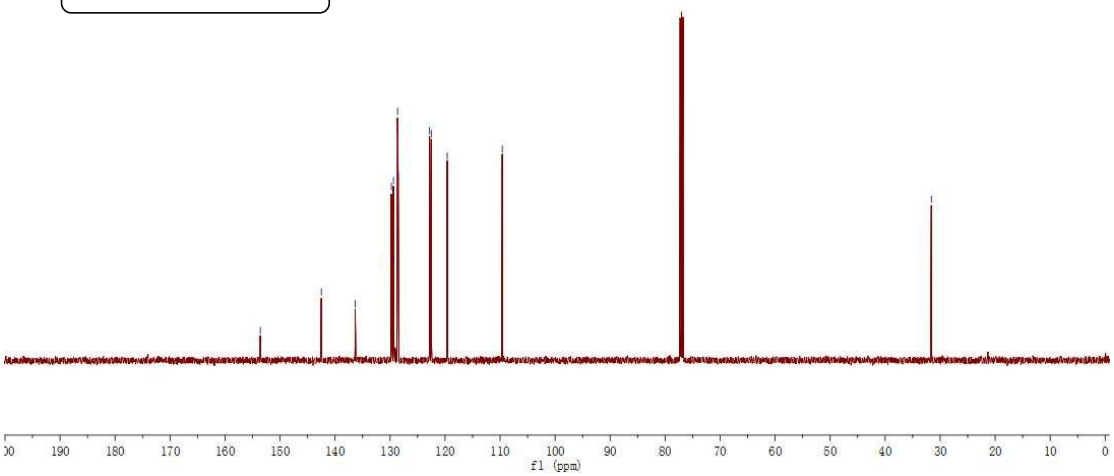





H/D
N
N
H/D


1.00
1.06
3.00
1.00
2.02
3.04
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


H/D
N
N
H/D


190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.81
7.80
7.79
7.79
7.50
7.50
7.49
7.48
7.48
7.47
7.47
7.44
7.44
7.42
7.42
7.41
7.41
7.40
7.39
7.39
7.39
7.35
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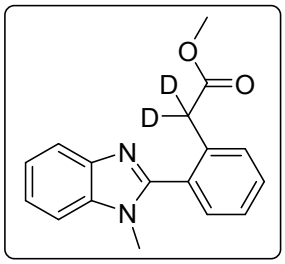
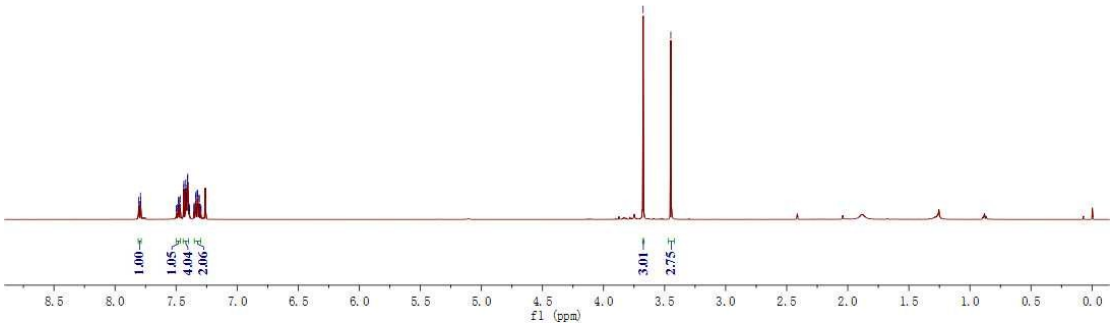
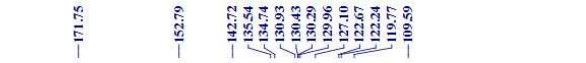
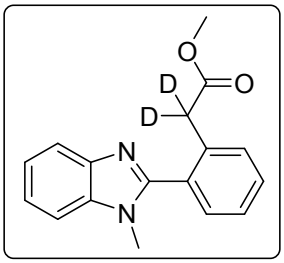
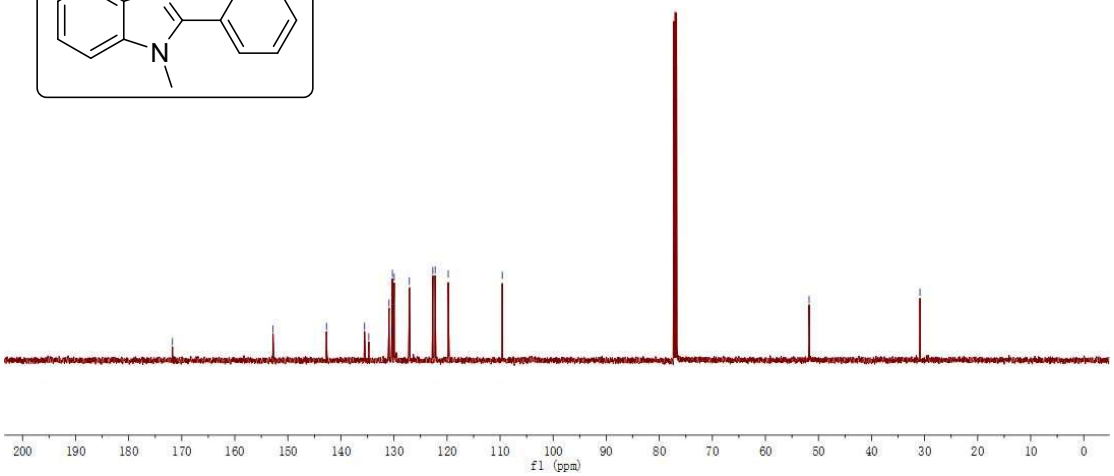
7.31
7.31
7.30
7.29
3.68
3.45


O
D
O
D
N
N


1.00
1.05
4.04
2.06
3.01
2.75
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


171.75
152.79
142.72
135.54
134.74
130.93
130.43
130.29
129.96
127.10
122.67
122.24
119.77
109.59
51.77
30.86


O
D
O
D
N
N


200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)