Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

for

Cu-Catalyzed Aerobic Oxidative Amidation of Aryl Alkyl Ketones with Azoles to Afford Tertiary Amides via Selective C-C bond Cleavage

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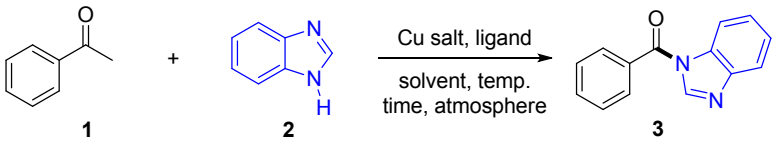
1. Optimization of the reaction conditions

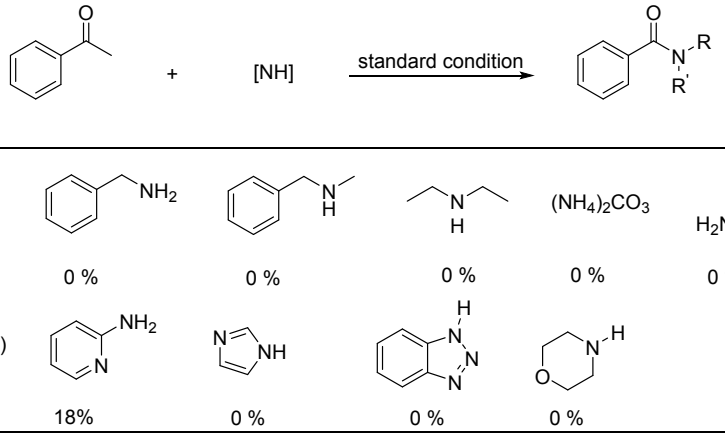


1 **2** **3**

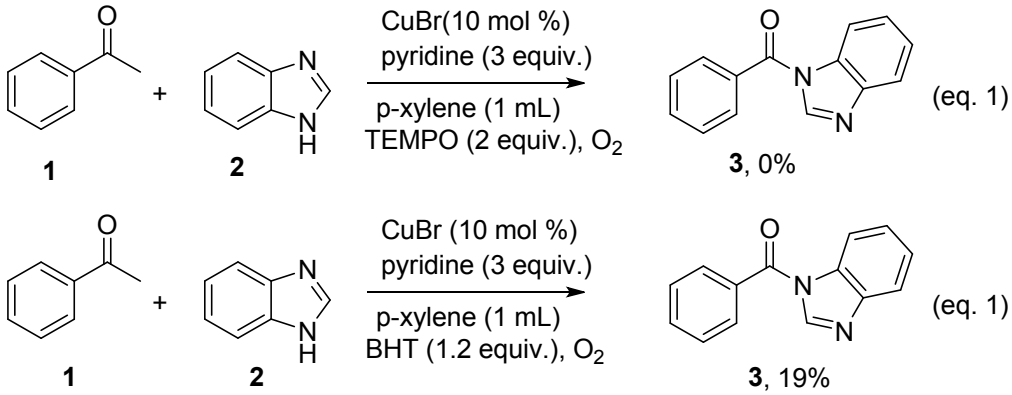
Entry	Catalyst (mol%)	atmosphere	ligand (equiv.)	temperature	solvent	yield of **3** (%)[b]
1	CuCl (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	16
2	CuI (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	64
3	$Cu(OAc)_2$ (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	7
4	CuBr (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	76[c]
5	Cu_2O (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	30
6	CuBr (10)	O_2	pyridine (3)	130 °C	DMSO	no product
7	CuBr (10)	O_2	pyridine (3)	130 °C	DMF	no produt
8	CuBr (10)	O_2	pyridine (3)	130 °C	Toluene	38
9	CuBr (10)	O_2	2-Phenyl-py (3)	130 °C	*p*-Xylene	62
10	CuBr (10)	O_2	2-Methyl-py (3)	130 °C	*p*-Xylene	33
11	CuBr (10)	O_2	2-amino-py (3)	130 °C	*p*-Xylene	no product
12	CuBr (10)	O_2	Et_3N (3)	130 °C	*p*-Xylene	no product
13	CuBr (10)	O_2	DMEDA (3)	130 °C	*p*-Xylene	no product
14	CuBr (10)	O_2	pyridine (0.5)	130 °C	*p*-Xylene	trace
15	CuBr (10)	O_2	pyridine (2)	130 °C	*p*-Xylene	74
16	CuBr (10)	O_2	pyridine (4)	130 °C	*p*-Xylene	64
17	CuBr (10)	N_2	pyridine (3)	130 °C	*p*-Xylene	no product
18	CuBr (10)	air	pyridine (3)	130 °C	*p*-Xylene	11
19	CuBr (10)	O_2	pyridine (3)	120 °C	*p*-Xylene	22
20	CuBr (10)	O_2	pyridine (3)	110 °C	*p*-Xylene	4
21	CuBr (20)	O_2	pyridine (3)	130 °C	*p*-Xylene	4
22[d]	CuBr (10)	O_2	pyridine (3)	130 °C	*p*-Xylene	39[c]
23	-	O_2	pyridine (3)	130 °C	*p*-Xylene	no product
24	CuBr (10)	O_2	-	130 °C	*p*-Xylene	33

Reaction Conditions: [a] acetophenone (**1**, 0.25 mmol), benzoimidazole (**2**, 0.5 mmol), Cu salt (10 mol%), ligand, solvent (1 mL), 16 h, temp., corresponding atmosphere. [b] GC yield. [c] Isolated yield. [d] acetophenone (**1**, 0.25 mmol), benzoimidazole (**2**, 0.25 mmol).

2. The Compatibility of Reaction with other nitrogen source



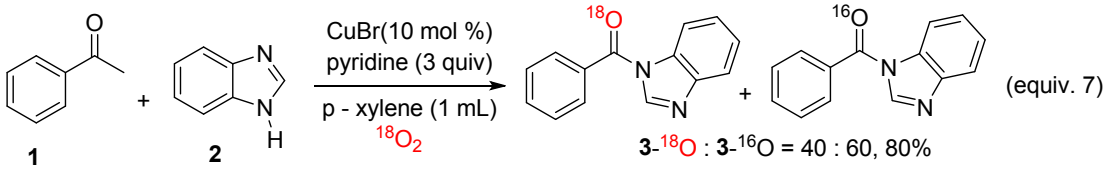
3. Radical Trapping Experiments



A sealed pressure vessel was charged with acetophenone **1** (30.0 mg, 0.25 mmol), benzimidazole **2** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), TEMPO (78.1mg, 0.5 mmol) or BHT (66.1 mg, 0.3 mmol) and p-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. After cooling down to room temperature, the mixture was measured by GC without further purification. the reaction were mostly inhibited. Which indicates that a radical pathway might be involved in this reaction.

4. Labeling Experiments

4.1. ^{18}O labeling experiment under $^{18}O_2$.



A sealed pressure vessel was charged with acetophenone **1a** (30.0 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and p-xylene (1 mL). The resulting solution was stirred at 130 °C under $^{18}O_2$ for

16 hours. Upon completion of the reaction, the solvents were removed via rotary evaporator and the residue was purified with flash chromatography (silica gel, ethyl acetate: petroleum ether=1:4) to give 45 mg of (1H-benzo[d]imidazol-1-yl)(phenyl)methanone **3** in 80% isolated yield as a white solid. then the amide **3** was determined by GC-MS and HRMS (Figure S2). In these cases, part of carbonyl oxygen atom of **3** was labeled. The ratio of **3**-^{18}O : 3-^{16}O = 40 : 60.

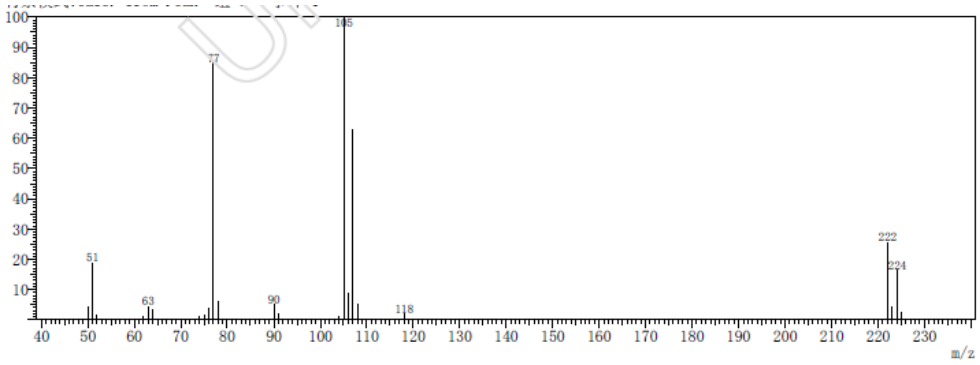
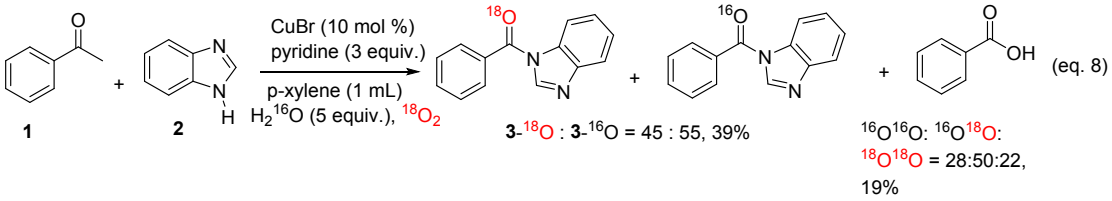


Figure S1. ^{18}O labeling experiment under $^{18}O_2$ measured by GC-MS.

4.2. ^{18}O labeling experiment in the present of $H_2{}^{18}O$ under $^{18}O_2$.



A sealed pressure vessel was charged with acetophenone **1a** (30.0 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), p-xylene (1 mL), and $H_2{}^{16}O$(22.5 mg). The resulting solution was stirred at 130 ^oC under $^{18}O_2$ for 16 hours. After cooling down to room temperature, the mixture was measured by GC-MS without further purification. In these cases, no carbonyl oxygen atom of **3** was labeled. The ratio of **3**-^{18}O : 3-^{16}O = 45 : 55.

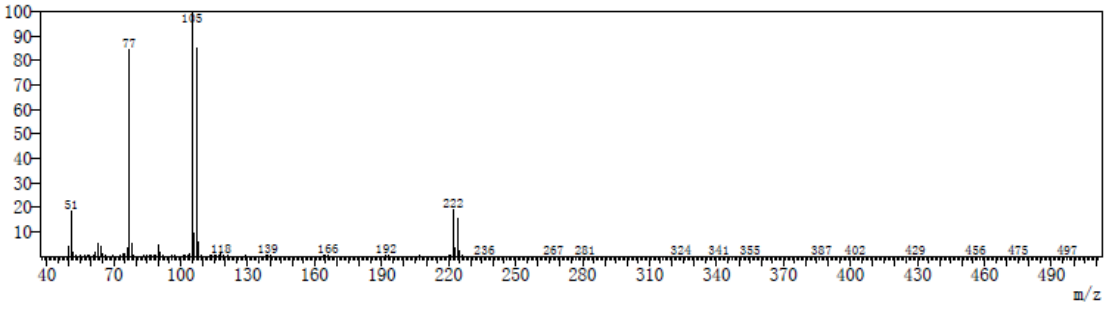
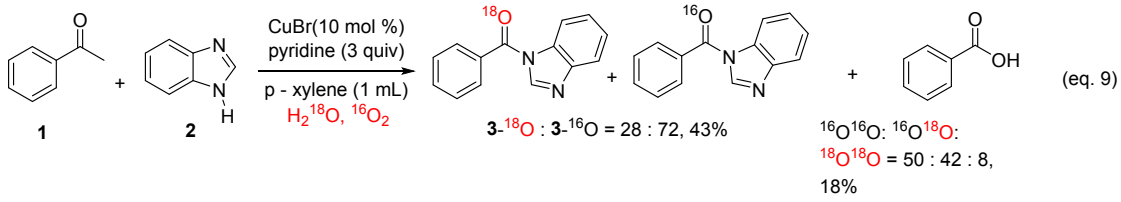


Figure S4. ^{18}O labeling experiment in the present of $H_2{}^{16}O$ under O^{18} .

4.3. ^{18}O labeling experiment in the present of $H_2{}^{18}O$ under $^{16}O_2$.



A sealed pressure vessel was charged with acetophenone **1a** (30.0 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), p-xylene (1 mL), and $H_2{}^{18}O$ (25 mg, 1.25 mmol). The resulting solution was stirred at 130 °C under $^{16}O_2$ for 16 hours. After cooling down to room temperature, the mixture was measured by GC-MS without further purification. In these cases, part of carbonyl oxygen atom of **3** was labeled. The ratio of **3**-^{18}O : **3**-^{16}O = 28 : 72.

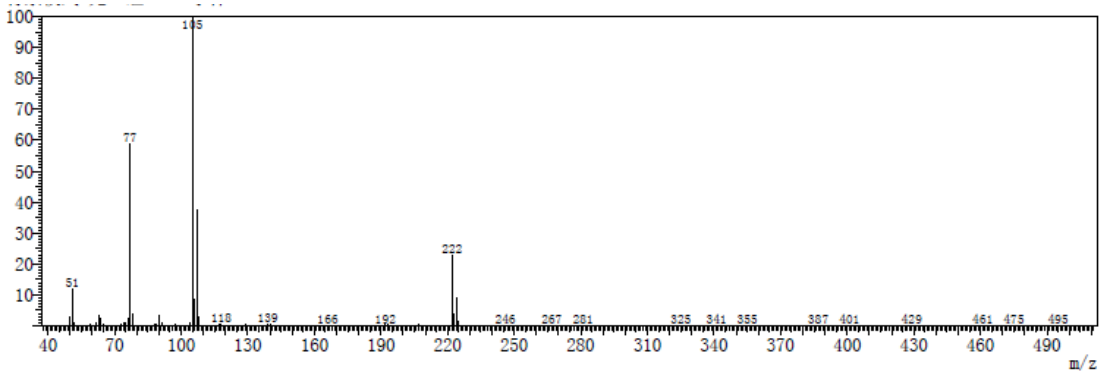
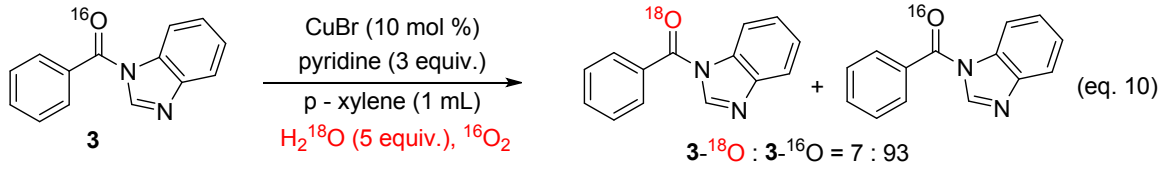


Figure S3. ^{18}O labeling experiment in the present of $H_2{}^{18}O$ under $^{16}O_2$.

4.4. ^{18}O labeling experiment of 3a in the present of $H_2{}^{18}O$



A sealed pressure vessel was charged with **3**(55mg, 0.25 mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), p-xylene (1 mL), $H_2{}^{18}O$ (5 equiv.). The resulting solution was stirred at 130 °C under $^{16}O_2$ for 16 hours. After cooling down to room temperature, the mixture was measured by GC-MS without further purification. In these cases, tiny part of carbonyl oxygen atom of **3** was labeled. The ratio of **3**-^{18}O : **3**-^{16}O = 7 : 93.

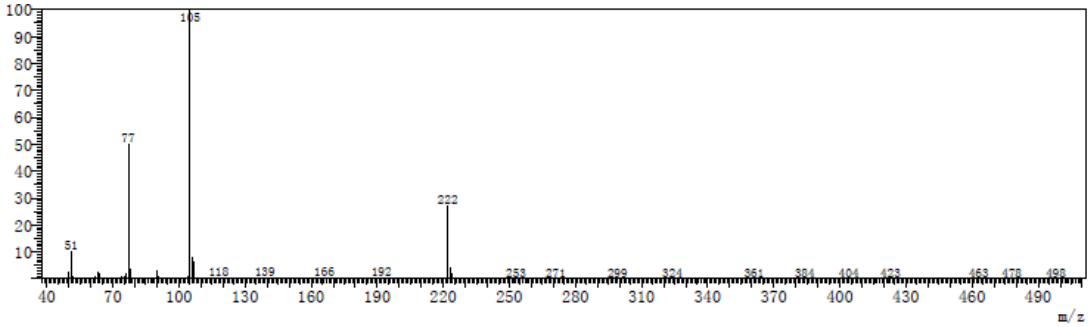


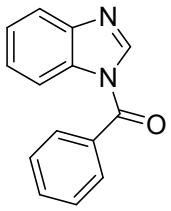
Figure S5. ^{18}O labeling experiment in the present of $H_2{}^{16}O$ under $^{18}O_2$.

5. General information

All experiments were conducted with a sealed pressure vessel. Flash column chromatography was performed over silica gel (200-300 mesh). ^{1}H NMR spectra were recorded on a Bruker AVIII-500M spectrometers, Chemical shifts (in ppm) were referenced to $CDCl_3$ (δ = 7.26 ppm) or DMSO-d_6 (δ = 2.54 ppm) as an internal standard. ^{13}C NMR spectra were obtained by using the same NMR spectrometers and were calibrated with $CDCl_3$ (δ = 77.0 ppm) or DMSO-d_6 (δ = 40.45 ppm). Unless otherwise noted, materials obtained from commercial suppliers were used without further purification.

6. Procedure and characterization data for products

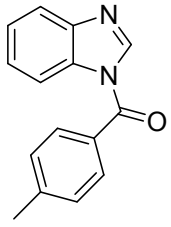
(1H-benzo[d]imidazol-1-yl)(phenyl)methanone（CAS：62573-86-8）[1]



3

A sealed pressure vessel was charged with acetophenone **1a** (30.0 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 ^{o}C under O_2 monitored by TLC and GC) for 16 hours. Upon completion of the reaction, the solvents were removed via rotary evaporator and the residue was purified with flash chromatography (silica gel, ethyl acetate: petroleum ether=1:4) to give 42 mg of (1H-benzo[d]imidazol-1-yl)(phenyl)methanone **3** in 76% isolated yield as a white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.22 (s, 1 H), 8.21–8.19 (m, 1 H), 7.85–7.83 (m, 1 H), 7.82 -7.80(m, 2 H), 7.71–7.68 (m, 1 H), 7.61–7.58 (m, 2 H), 7.47–7.42 (m, 2 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.1, 144.0, 143.1, 133.2, 132.8, 132.1, 129.5, 129.0, 125.8, 125.3, 120.5, 115.4. mp = 70.1–71.2 ^{o}C.

(1H-benzo[d]imidazol-1-yl)(p-tolyl)methanone（CAS：28997-00-4）[7]

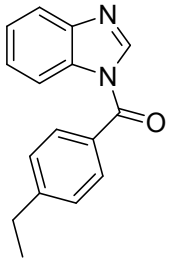


4

A sealed pressure vessel was charged with 1-(p-tolyl)ethanone **1b** (33.0 mg, 0.25 mmol), benzimidazole **2a**(60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 ^{o}C under O_2 for 16 hours. affords 29.5 mg (50%) of **4**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.24 (s, 1 H), 8.19–8.17 (m, 1 H), 7.84–7.83 (m, 1 H), 7.72 -7.71(m, 2 H), 7.46–7.41 (m, 2 H), 7.39–7.38 (m, 2 H), 2.49 (s, 1 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.1, 144.3, 144.1, 143.1,

132.2, 130.0, 129.8, 129.7, 125.6, 125.1, 120.5, 115.4, 21.7. mp = 92.7–94.5 °C.

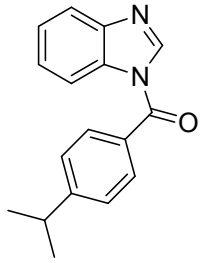
(1H-benzo[d]imidazol-1-yl)(4-ethylphenyl)methanone（new compound）



5

A sealed pressure vessel was charged with 1-(4-ethylphenyl)ethanone **1c** (37.0 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 31.9 mg (51%) of **5**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.25 (s, 1 H), 8.20–8.18 (m, 1 H), 7.85–7.83 (m, 1 H), 7.75 -7.73(m, 2 H), 7.47–7.40 (m, 4 H), 2.78 (q, *J*=8.00Hz, 2 H), 1.31 (t, *J*=7.5, 3 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.1, 150.4, 144.1, 143.2, 132.2, 130.2, 129.9, 128.5, 125.6, 125.1, 120.5, 115.4, 29.0, 15.1. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 250.1106, found 250.1111. mp = 90.1–91.8 °C.

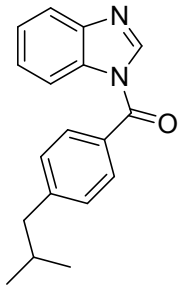
(1H-benzo[d]imidazol-1-yl)(4-isopropylphenyl)methanone（CAS：901440-49-1）[10]



6

A sealed pressure vessel was charged with 1-(4-isopropylphenyl)ethanone **1d** (41 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 46.8 mg (71%) of **6**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.26 (s, 1 H), 8.21–8.19 (m, 1 H), 7.85–7.83 (m, 1 H), 7.76 -7.74(m, 2 H), 7.47–7.41 (m, 4 H), 3.08-2.99 (m, 1 H), 1.32 (d, *J*=7.00, 6 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.0, 154.9, 144.0, 143.2, 132.2, 130.3, 129.9, 127.2, 125.6, 125.1, 120.5, 115.4, 34.3,23.6. mp = 75.5–76.5 °C.

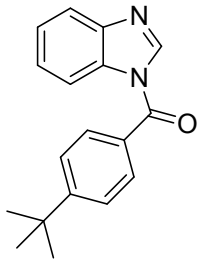
(1H-benzo[d]imidazol-1-yl)(4-isobutylphenyl)methanone（new compound）



7

A sealed pressure vessel was charged with 1-(4-isobutylphenyl)ethanone **1f** (45 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 49.3 mg (71%) of **7**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.27 (s, 1 H), 8.19–8.18 (m, 1 H), 7.85-7.83 (m, 1 H), 7.74 -7.72(m, 2 H), 7.46–7.40 (m, 2 H), 7.36–7.34 (m, 2 H), 2.59 (d, *J*=7.50, 2H), 1.95(m, 1H), 0.95(d, *J*=6.50, 6 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.0, 148.0, 143.9, 143.2, 132.2, 130.1, 129.7, 129.6, 125.6, 125.1, 120.4, 115.4, 45.3, 30.7, 22.3. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 278.1419, found 278.1415. mp = 63.4–64.8 °C.

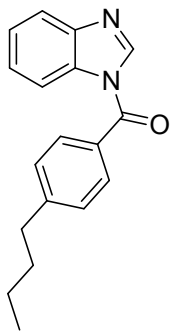
(1H-benzo[d]imidazol-1-yl)(4-(tert-butyl)phenyl)methanone（CAS：20208-57-5）[6]



8

A sealed pressure vessel was charged with 1-(4-(tert-butyl)phenyl)ethanone **1g** (45 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 28.4 mg (41%) of **8**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.26 (s, 1 H), 8.23–8.21 (m, 1 H), 7.85-7.83 (m, 1 H), 7.77 -7.75(m, 2 H), 7.61–7.59 (m, 2 H), 7.47–7.41 (m, 2 H), 1.39(s, 9 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.1, 157.2, 144.1, 143.2, 132.2, 130.0, 129.7, 126.0, 125.7, 125.1, 120.5, 115.5, 35.3, 31.1. mp = 131.3–132.8 °C.

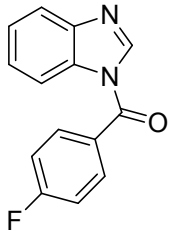
(1H-benzo[d]imidazol-1-yl)(4-butylphenyl)methanone（ new compound ）



9

A sealed pressure vessel was charged with 1-(4-butylphenyl)ethanone **1e** (45 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 43.7 mg (63%) of **9**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.27 (s, 1 H), 8.20–8.19 (m, 1 H), 7.85-7.84 (m, 1 H), 7.74 -7.73(m, 2 H), 7.47–7.38 (m, 4 H), 2.74 (t, *J*=8.00, 2H), 1.70- 1.64(m, 2H), 1.44-1.35 (m, 2 H), 0.96 (t, *J*=7.50, 3 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.1, 149.2, 144.0, 143.2, 132.2, 130.1, 129.8, 129.1, 125.7, 125.1, 120.5, 115.4, 35.7, 33.2, 22.3, 13.9. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 278.1419, found 278.1411. mp = 37.1-38.9 °C

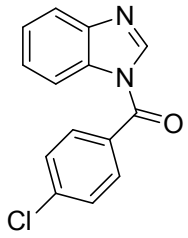
(1H-benzo[d]imidazol-1-yl)(4-fluorophenyl)methanone（CAS：154786-24-0）[8]



10

A sealed pressure vessel was charged with 1-(4-fluorophenyl)ethanone **1h** (35 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 31.2 mg (52%) of **10**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.21 (s, 1 H), 8.16–8.14 (m, 1 H), 7.87-7.83 (m, 3 H), 7.48 -7.42(m, 2 H), 7.30–7.27 (m, 2 H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 165.88, 165.61(d, *J*= 254.63Hz), 144.06, 142.73, 132.23(d, *J*= 9.13Hz), 132.09, 129.50(d, *J*= 3.38Hz), 125.83, 125.35, 120.64, 116.45(d, *J*= 22.13Hz), 115.34. mp = 74.8–76.1 °C.

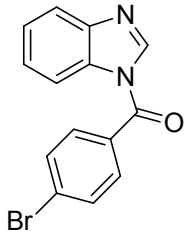
(1H-benzo[d]imidazol-1-yl)(4-chlorophenyl)methanone（CAS：71589-37-2）[9]



11

A sealed pressure vessel was charged with 1-(4-chlorophenyl)ethanone **1i** (39 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 29.3 mg (46%) of **11**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.19 (s, 1 H), 8.17–8.15 (m, 1 H), 7.85-7.83 (m, 1 H), 7.78 -7.75(m, 2 H), 7.59–7.57 (m, 2 H); 7.48-7.43 (m, 2H) ; ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.00, 144.00, 142.66, 139.86, 131.96, 131.07, 130.94, 129.46, 125.90, 125.43, 120.63, 115.36. mp = 149.3–150.9 °C.

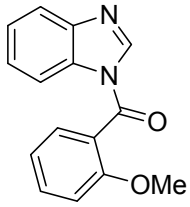
(1H-benzo[d]imidazol-1-yl)(4-bromophenyl)methanone（CAS：304668-33-5）[1]



12

A sealed pressure vessel was charged with 1-(4-bromophenyl)ethanone **1j** (50 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 43.6 mg (58%) of **12**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.18 (s, 1 H), 8.17–8.13 (m, 1 H), 7.85-7.81 (m, 1 H), 7.75 -7.73(m, 2 H), 7.69–7.66 (m, 2 H); 7.47-7.42（m, 2H）; ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.08, 144.03, 142.61, 132.41, 131.96, 131.58, 130.96, 128.35, 125.88, 125.42, 120.63, 115.35. mp = 108.9–110.3 °C.

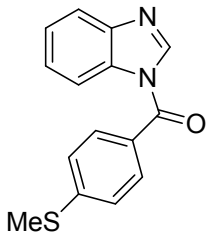
(1H-benzo[d]imidazol-1-yl)(2-methoxyphenyl)methanone（CAS：816441-32-4）[1]



13

A sealed pressure vessel was charged with 1-(2-methoxyphenyl)ethanone **1m** (37 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 37.8 mg (60%) of **13**. white oil. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.27-8.25 (m, 1 H), 7.97 (s, 1 H), 7.81-7.79 (m, 1 H), 7.60 -7.53(m, 2 H), 7.46–7.40 (m, 2 H); 7.15-7.12（m, 1H）, 7.07-7.05 (m, 1H), 3.78(s, 3H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 165.81, 156.49, 144.09, 143.59, 133.48, 131.58, 129.91, 125.68, 125.12, 122.93, 121.15, 120.32, 115.50, 111.60, 55.71.

(1H-benzo[d]imidazol-1-yl)(4-(methylthio)phenyl)methanone（new compound）

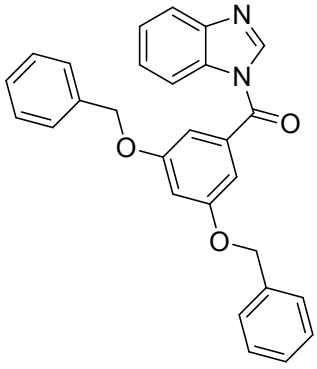


14

A sealed pressure vessel was charged with 1-(4-(methylthio)phenyl)ethanone **1n** (42 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 26.8 mg (40%) of **14**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.27 (s,

1 H), 8.16-8.14 (m, 1 H), 7.85-7.83 (m, 1 H), 7.75 -7.72(m, 2 H), 7.47–7.41 (m, 2 H); 7.39-7.37（m, 2H）, 2.57(s, 3H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.49, 146.86, 143.95, 142.94, 132.16, 130.15, 128.35, 125.67, 125.31, 125.16, 120.49, 115.33, 14.74. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 268.0670, found 268.0674. mp = 84.3–85.8 °C.

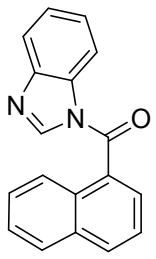
(1H-benzo[d]imidazol-1-yl)(3,5-bis(benzyloxy)phenyl)methanone（new compound）



15

A sealed pressure vessel was charged with 1-(3,5-bis(benzyloxy)phenyl)ethanone **1q** (84 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 37.9 mg (35%) of **15**. yellow oil. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.237-8.196 (m, 1 H), 8.167 (s, 1 H), 7.851-7.833 (m, 1 H), 7.477 -7.346(m, 12 H), 6.990–6.986 (m, 2 H); 6.904- 6.895（m, 1H）, 5.09 (s, 4H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.66, 160.11, 144.06, 143.03, 135.91, 134.54, 132.04, 128.72, 128.31, 127.50, 125.76, 125.29, 120.53, 115.51, 108.46, 106.97, 70.46. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 430.1630, found 434.1622.

(1H-benzo[d]imidazol-1-yl)(naphthalen-1-yl)methanone（CAS：26670-22-4）[1]

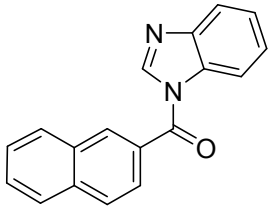


16

A sealed pressure vessel was charged with 1-(naphthalen-1-yl)ethanone **1k** (43 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 53.0 mg (78%) of **16**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.32-8.30 (m, 1 H), 8.12–8.10 (m, 1 H), 8.00-7.96 (m, 3 H), 7.86 -7.84(m, 1 H), 7.74–7.72 (m, 1 H); 7.62-7.55（m, 3H）, 7.50-7.46 (m, 2H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.06, 144.21, 143.18, 133.57, 132.42, 131.83, 130.58, 130.19, 128.67, 128.13, 127.35, 127.14,

125.90, 125.42, 124.53, 124.48, 120.60, 115.64. mp = 101.5–102.3 °C.

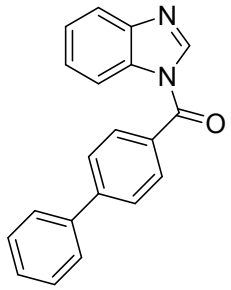
(1H-benzo[d]imidazol-1-yl)(naphthalen-2-yl)methanone（new compound）



17

A sealed pressure vessel was charged with 1-(naphthalen-2-yl)ethanone **1l** (43 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 40.8 mg (60%) of **17**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.33-8.32 (m, 2 H), 8.23–8.22 (m, 1 H), 8.05-8.04 (m, 1 H), 7.98 -7.96(m, 2 H), 7.88–7.86 (m, 2 H); 7.70-7.67（m, 1H）, 7.65-7.62 (m, 1H), 7.49-7.44 (m, 2H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.16, 144.03, 143.22, 135.31, 132.20, 131.11, 129.90, 129.19, 128.99, 127.98, 127.56, 125.78, 125.28, 125.03, 120.55, 115.44. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 272.0950, found 272.0953. mp = 105.1–106.7 °C.

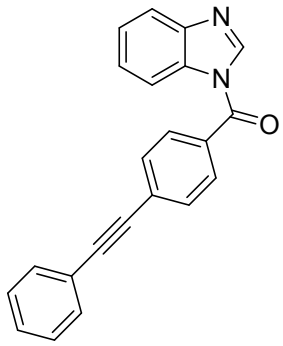
[1,1'-biphenyl]-4-yl(1H-benzo[d]imidazol-1-yl)methanone（CAS：349407-41-6）[11]



18

A sealed pressure vessel was charged with 1-([1,1'-biphenyl]-4-yl)ethanone **1o** (50 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 46.9 mg (63%) of **18**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.31 (s, 1 H), 8.23-8.21 (m, 1 H), 7.91-7.89 (m, 2 H), 7.87 -7.85(m, 1 H), 7.82–7.80 (m, 2 H); 7.68-7.67（m, 2H）, 7.53-7.43(m, 5H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.85, 146.19, 144.09, 143.04, 139.31, 132.17, 131.34, 130.24, 129.09, 128.58, 127.64, 127.30, 125.75, 125.25, 120.57, 115.44. mp = 117.8–119.5 °C.

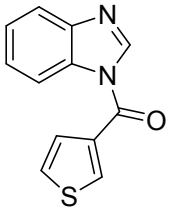
(1H-benzo[d]imidazol-1-yl)(4-(phenylethynyl)phenyl)methanone（new compound）



19

A sealed pressure vessel was charged with 1-(4-(phenylethynyl)phenyl)ethanone **1p** (55 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 40.2 mg (50%) of **19**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.24 (s, 1 H), 8.18-8.17 (m, 1 H), 7.86-7.84 (m, 1 H), 7.81 -7.79(m, 2 H), 7.73–7.72 (m, 2 H); 7.59-7.57（m, 2H）, 7.48-7.43(m, 2H), 7.40-7.38 (m, 3H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.38, 144.03, 142.80, 132.04, 132.00, 131.80, 131.77, 129.61, 129.02, 128.66, 128.46, 125.80, 125.32, 122.31, 120.58, 115.38, 93.39, 87.94. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 322.1106, found 322.1111. mp = 145.3–147.1 °C.

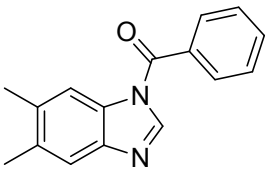
(1H-benzo[d]imidazol-1-yl)(thiophen-3-yl)methanone（new compound）



20

A sealed pressure vessel was charged with 1-(thiophen-3-yl)ethanone **1r**(32 mg, 0.25 mmol), benzimidazole **2a** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 28.5 mg (50%) of **20**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.404 (s, 1 H), 8.217- 8.199 (m, 1 H), 8.074-8.066 (m, 1 H), 7.851 -7.834(m, 1 H), 7.586– 7.574 (m, 1 H); 7.542- 7.526（m, 1H）, 7.475- 7.416(m, 2H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 161.46, 144.02, 142.53, 134.71, 133.22, 132.13, 128.20, 127.60, 125.74, 125.25, 120.56, 115.39. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 228.0357, found 228.0361. mp = 124.8–126.3 °C.

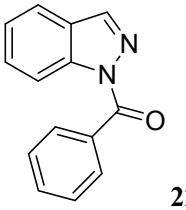
(5,6-dimethyl-1H-benzo[d]imidazol-1-yl)(phenyl)methanone（CAS：16109-46-9）[5]



21

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 5,6-dimethyl-1H-benzo[d]imidazole **2b** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (67 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 42.5 mg (68%) of **21**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.09 (s, 1 H), 8.00 (s, 1 H), 7.80-7.78 (m, 2 H), 7.69 -7.66(m, 1 H), 7.59– 7.56 (m, 3 H), 2.42(s, 3H), 2.40(s, 3H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.11, 142.52, 142.37, 135.06, 134.30, 133.08, 132.99, 130.47, 129.46, 128.96, 120.53, 115.67, 20.53, 20.32. mp = 91.8–93.7 °C.

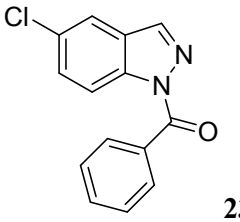
(1H-indazol-1-yl)(phenyl)methanone（CAS：23301-00-0）[3]



22

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2c** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 45.5 mg (82%) of **22**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.58 (d, *J*= 8.5Hz, 1 H), 8.211 (s, 1 H), 8.075-8.057 (m, 2 H), 7.798 -7.782(m, 1 H), 7.648– 7.592 (m, 2 H), 7.544- 7.513(s, 2H), 7.434- 7.404(m, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 168.39, 140.30, 140.12, 133.29, 132.22, 130.89, 129.51, 127.97, 126.11, 124.82, 120.92, 115.89. mp = 83.8–84.2 °C.

(5-chloro-1H-indazol-1-yl)(phenyl)methanone （new compound）

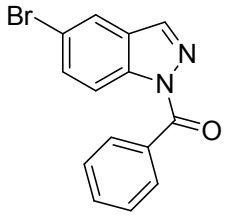


23

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2e** (78mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 46 mg (71%) of **23**. yellow solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.509 (d, *J*= 8.9Hz, 1 H), 8.15 (s, 1H), 8.09-8.04 (m, 2 H), 8.07 -8.05(m, 2 H), 7.75 (d, *J*=1.8Hz, 1 H), 7.64- 7..59(m, 1H), 7.57(dd, *J*= 8.9, 2.0 Hz, 1H), 7.54 – 7.51(m, 2H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ

168.08, 139.20, 138.62, 132.75, 132.47, 130.98, 130.36, 129.90, 128.02, 127.10, 120.31, 116.93. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 256.0403, found 256.0400. mp = 132.3–133.8 °C.

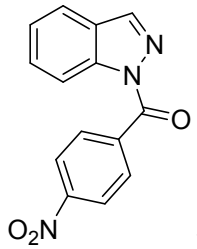
(5-bromo-1H-indazol-1-yl)(phenyl)methanone（CAS：936846-01-4）[2]



24

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2f** (100mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 67 mg (89%) of **24**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.46 (d, *J*= 8.8Hz, 1 H), 8.15 (s, 1H), 8.07-8.05 (m, 2 H), 7.95 -7.90(m, 2 H), 7.71 (dd, *J*= 8.8 Hz, 1.8Hz, 1 H), 7.65-7.59(m, 1H), 7.56 – 7.49(m, 2H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 168.13, 139.08, 138.96, 132.76, 132.51, 131.00, 128.04, 127.65, 123.52, 117.93, 117.25. mp = 128.9–129.5 °C.

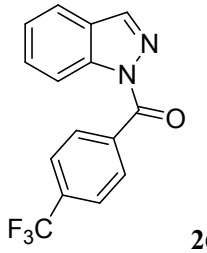
(1H-indazol-1-yl)(4-nitrophenyl)methanone（CAS：500900-65-2）[4]



25

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2c** (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 36 mg (53%) of **25**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.57 (dd, *J*= 8.4Hz, 0.7Hz, 1 H), 8.39- 8.32 (m, 2H), 8.28- 8.18 (m, 3 H), 7.81(d, *J*= 7.9Hz, 1 H), 7.69– 7.64 (m, 1 H), 7.49- 7.44(m, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.33, 149.59, 141.31, 139.89, 138.99, 131.83, 130.05, 126.27, 125.49, 123.06, 121.21, 115.89. mp = 155.4–157.3 °C.

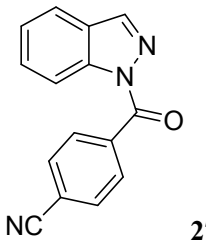
(1H-indazol-1-yl)(4-(trifluoromethyl)phenyl)methanone （new compound）



26

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2c** (93mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 52 mg (71%) of **26**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.58 (dd, *J*= 8.4Hz, 0.7Hz, 1 H), 8.22 (s , 1H), 8.17 (d, *J*= 8.1Hz, 2 H), 7.79(t, *J*= 7.5Hz, 3 H), 7.65 (ddd, *J*= 8.3Hz, 7.2Hz, 1.0Hz, 1 H), 7.49- 7.41(m, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 167.08, 140.93, 139.96, 136.65, 133.52(q, *J*= 32.7Hz), 131.14, 129.83, 126.20, 125.22,124.96(q, *J*= 3.7Hz) 123.64(q, *J*= 272.7Hz), 121.09, 115.88. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 290.0667, found 290.0661. mp = 102.5–103.8 °C.

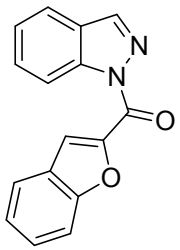
4-(1H-indazole-1-carbonyl)benzonitrile （new compound）



27

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2c** (73mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 51 mg (82%) of **27**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.56 (d, *J*= 8.4Hz, 1 H), 8.21 (s, 1H), 8.16-8.14 (m, 2 H), 7.82 -7.80(m, 3 H), 7.67– 7.64 (m, 1 H), 7.47- 7.44(m, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.49, 141.16, 139.87, 137.28, 131.66, 131.31, 129.95, 126.20, 125.38, 121.15, 118.00, 115.84, 115.43. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 247.0746, found 247.0751. mp = 148.1–149.0 °C.

benzofuran-2-yl(1H-indazol-1-yl)methanone （new compound）

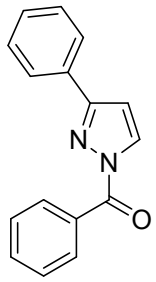


28

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole

2c (60mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 34 mg (52%) of **28**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.62 (d, *J*= 8.4Hz, 1 H), 8.40 (d, *J*= 0.6 Hz, 1H), 8.26 (s, 1H), 7.77(t, *J*= 7.8Hz, 2H), 7.67(d, *J*= 8.4Hz, 1 H), 7.65- 7.58(m, 1H), 7.53- 7.46(m, 1H), 7.40(t, *J*= 7.5Hz, 1H), 7.33(t, *J*= 7.5Hz, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 157.66, 155.64, 145.87, 140.82, 140.17, 129.75, 128.12, 127.41, 125.90, 124.96, 123.78, 123.51, 121.04, 119.06, 115.93, 112.34. HRMS m/z (EI) calcd. for C9H10F3O2 M+ 262.0742, found 262.0737. mp = 150.8–152.8 °C.

phenyl(3-phenyl-1H-pyrazol-1-yl)methanone（CAS：126382-89-6）[1]



29

A sealed pressure vessel was charged with acetophenone **1a** (30 mg, 0.25 mmol), 1H-indazole **2d** (72mg, 0.5mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 16 hours. affords 22 mg (37%) of **29**. white solid. ^{1}H NMR: (500 MHz, $CDCl_3$, ppm) δ 8.48 (d, *J*= 3.0Hz, 1 H), 8.281- 8.262 (m,2H), 8.902-7.885 (m, 2 H), 7.656 -7.627(m, 1 H), 7.554– 7.523 (m, 2 H), 7.463- 7.399(m, 3H), 6.871(d, *J*= 3.0, 1H); ^{13}C NMR: (125 MHz, $CDCl_3$, ppm).) δ 166.11, 155.89, 132.98, 131.86, 131.79, 131.73, 131.48, 129.18, 128.74, 128.02, 126.37, 107.18. mp = 63.7-64.9 °C

7. References

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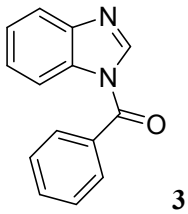
8. NMR Spectra

(1H-benzo[d]imidazol-1-yl)(phenyl)methanone（CAS：62573-86-8）[1]



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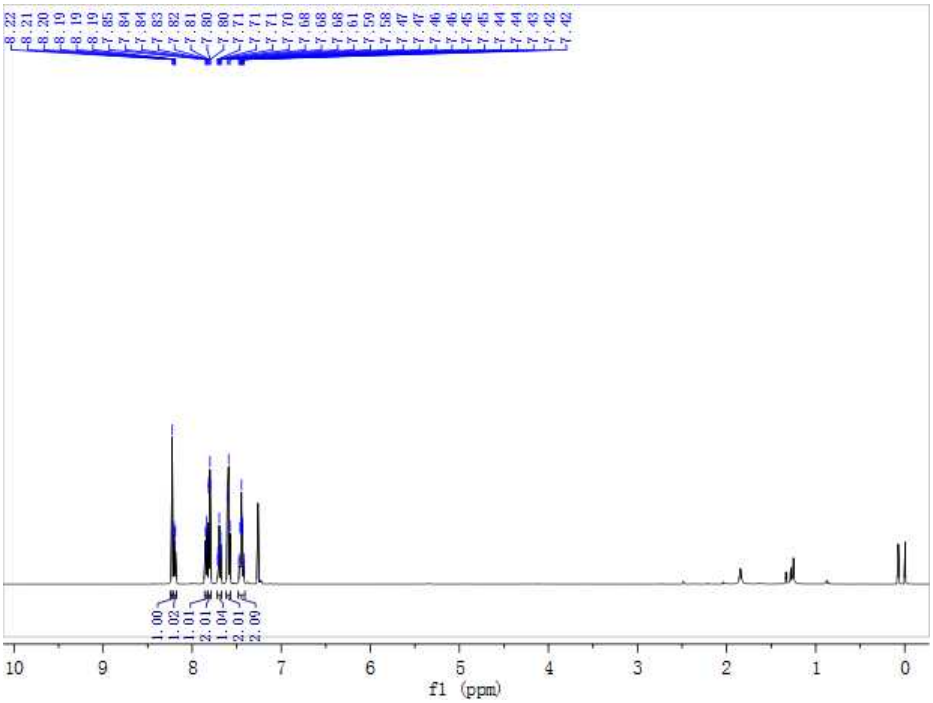
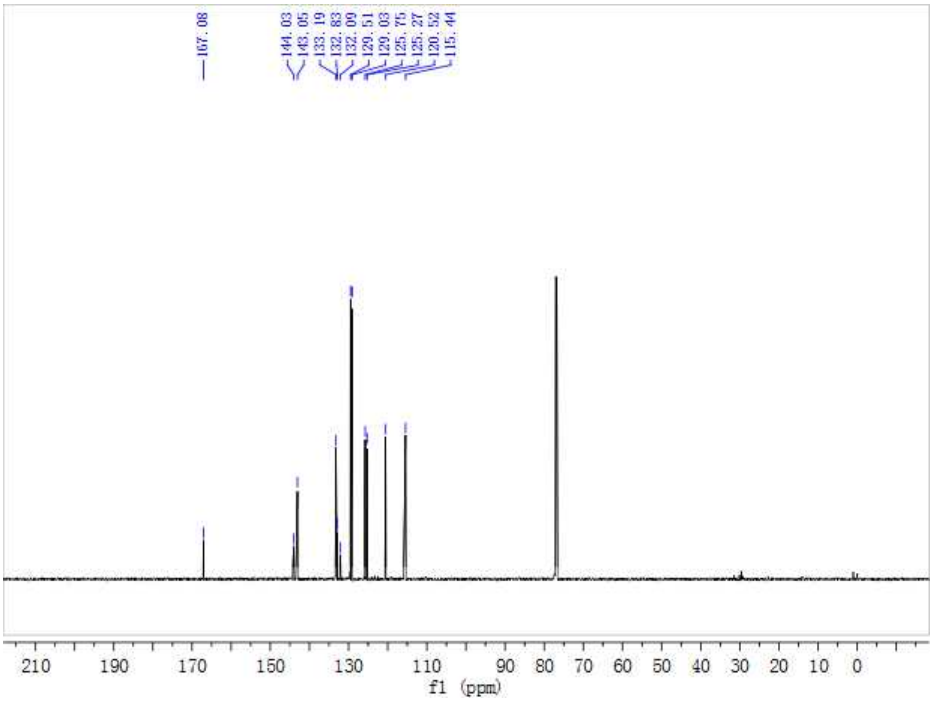
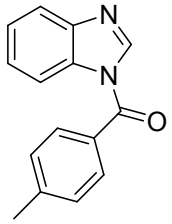




(1H-benzo[d]imidazol-1-yl)(p-tolyl)methanone（CAS：28997-00-4）[7]



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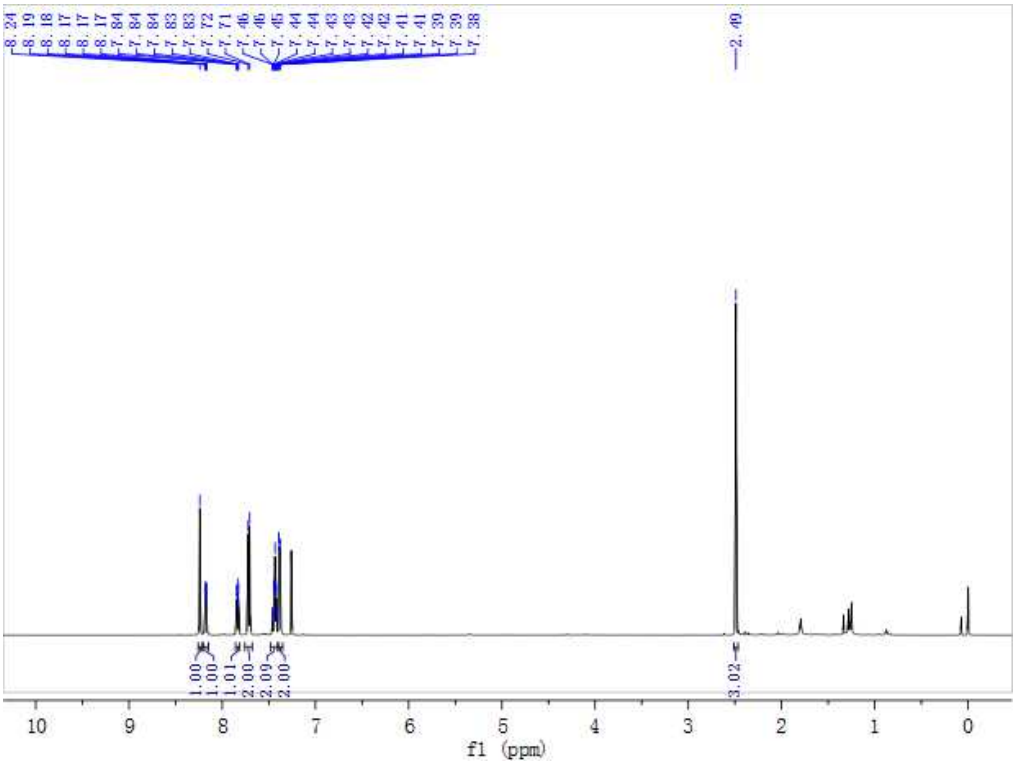
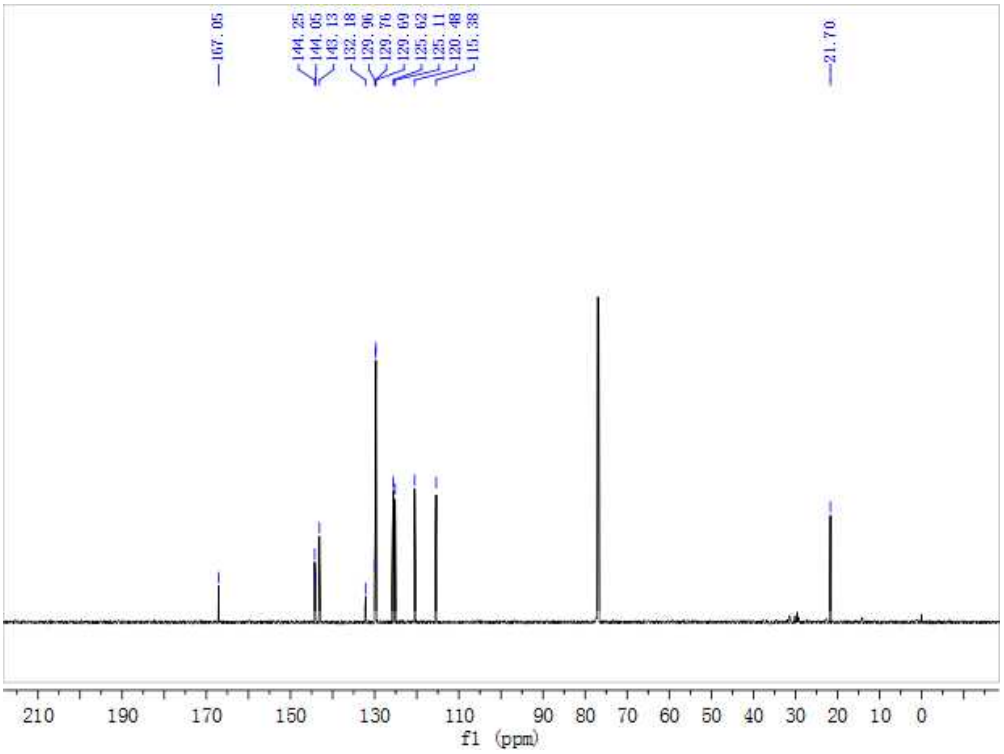
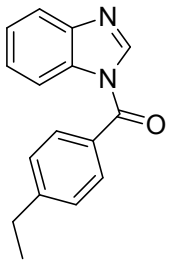




(1H-benzo[d]imidazol-1-yl)(4-ethylphenyl)methanone（new compound）



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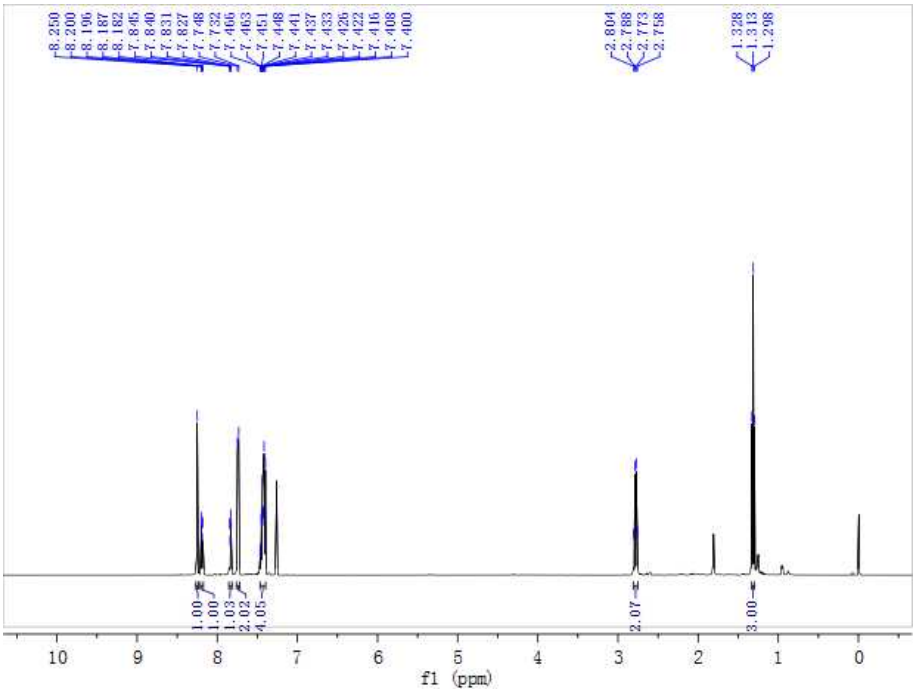
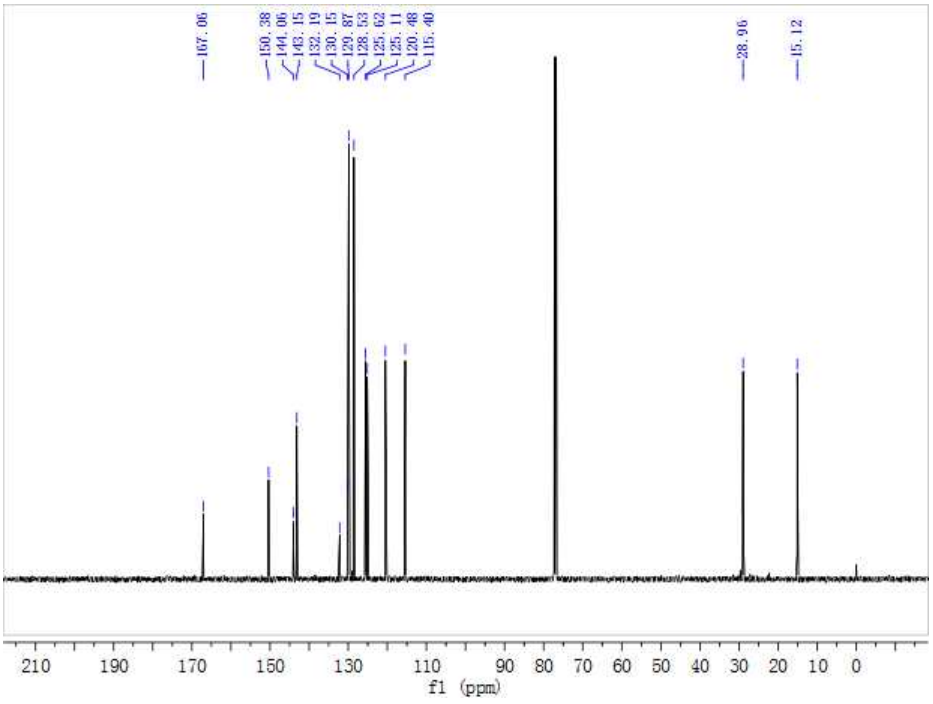
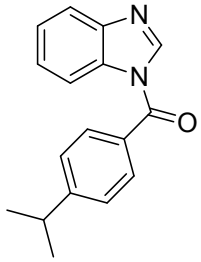




(1H-benzo[d]imidazol-1-yl)(4-isopropylphenyl)methanone（CAS：901440-49-1）[10]



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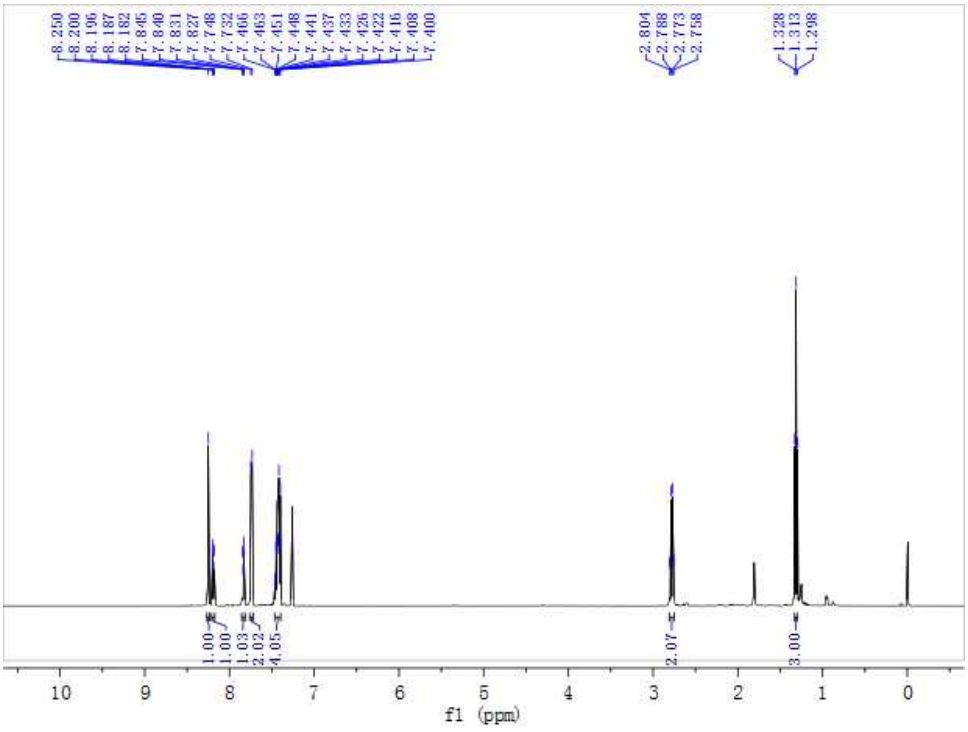
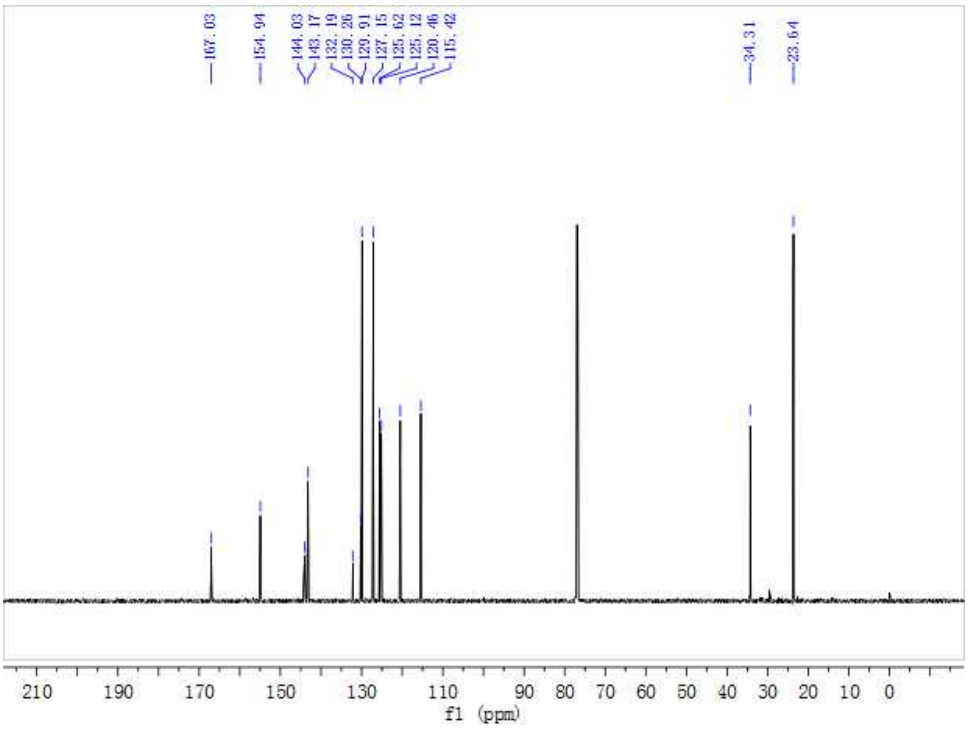
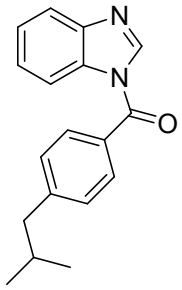




(1H-benzo[d]imidazol-1-yl)(4-isobutylphenyl)methanone（new compound）



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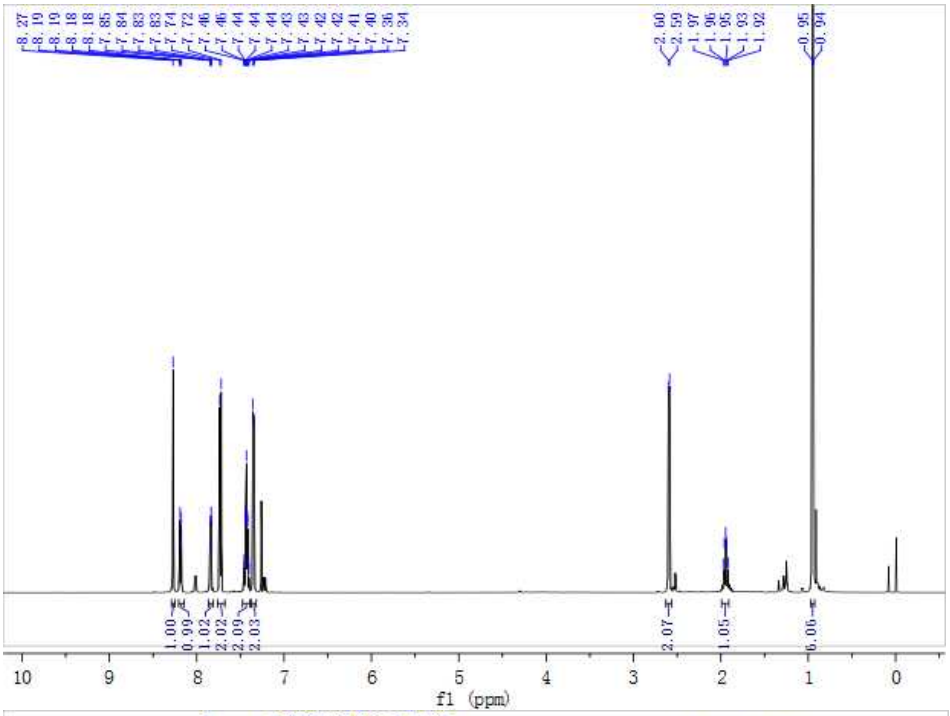
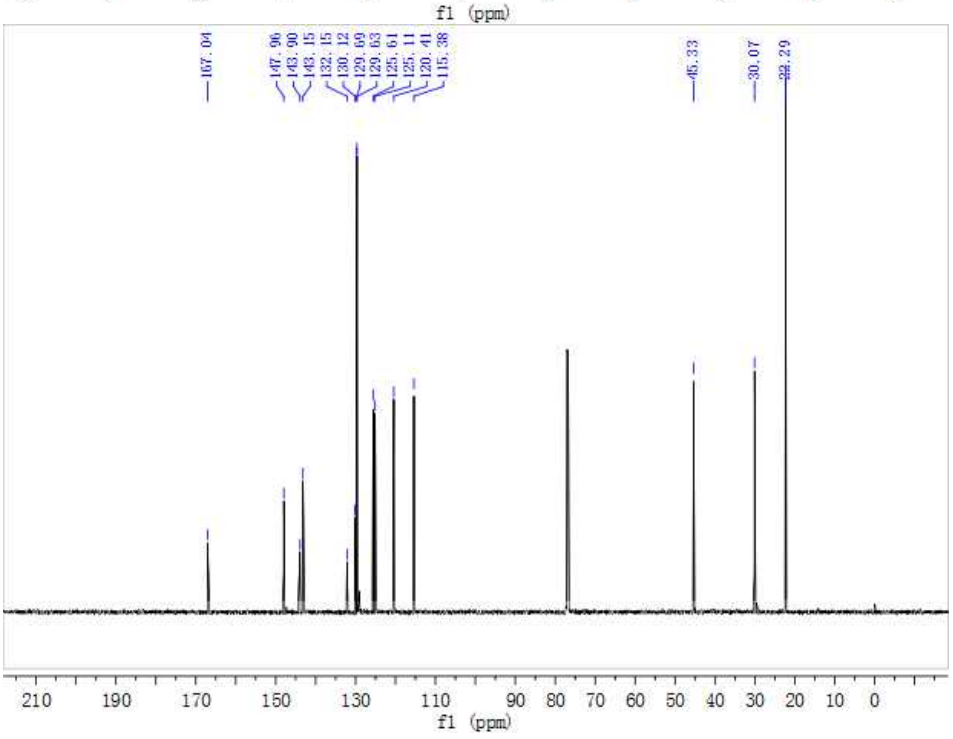
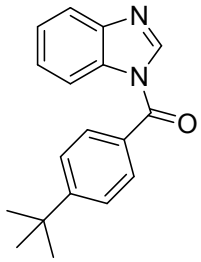




(1H-benzo[d]imidazol-1-yl)(4-(tert-butyl)phenyl)methanone（CAS：20208-57-5）[6]



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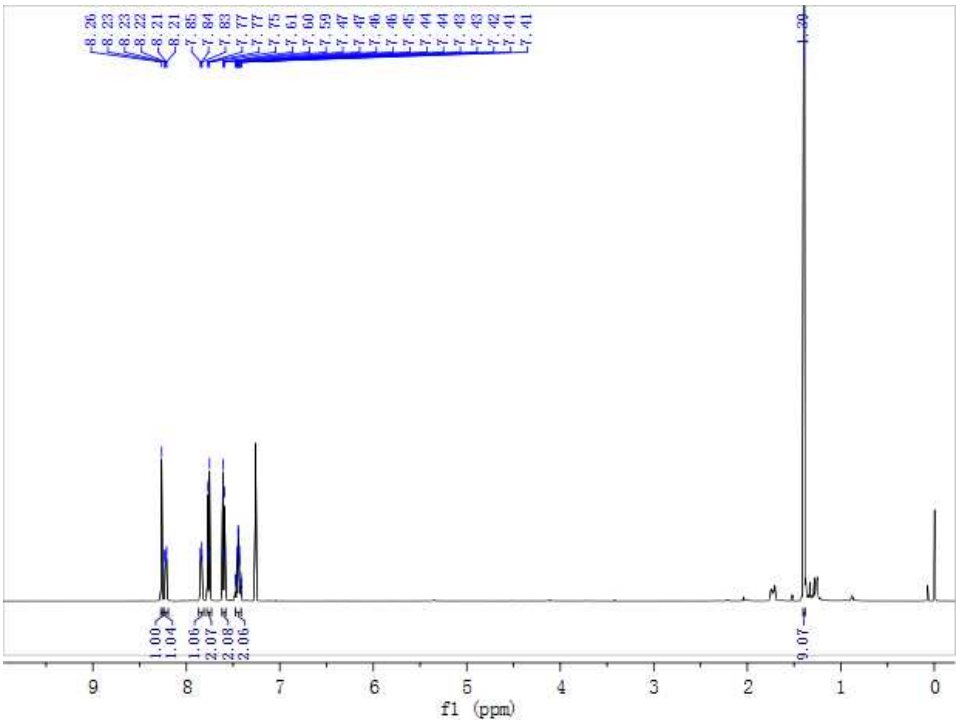
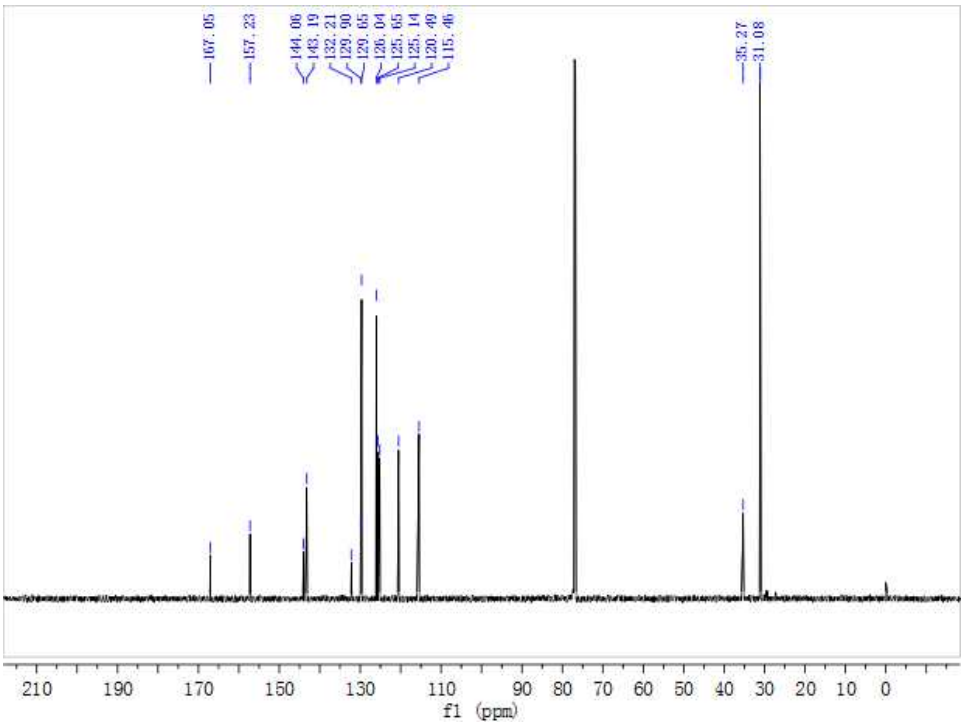
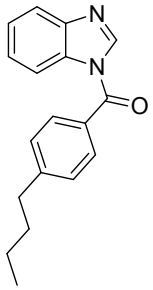




(1H-benzo[d]imidazol-1-yl)(4-butylphenyl)methanone（new compound）



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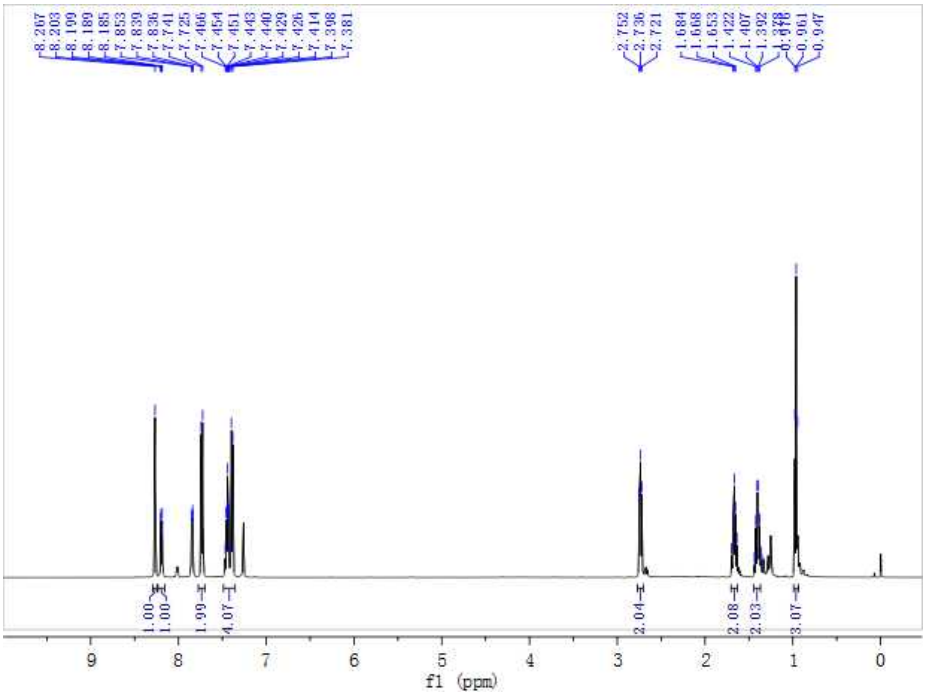
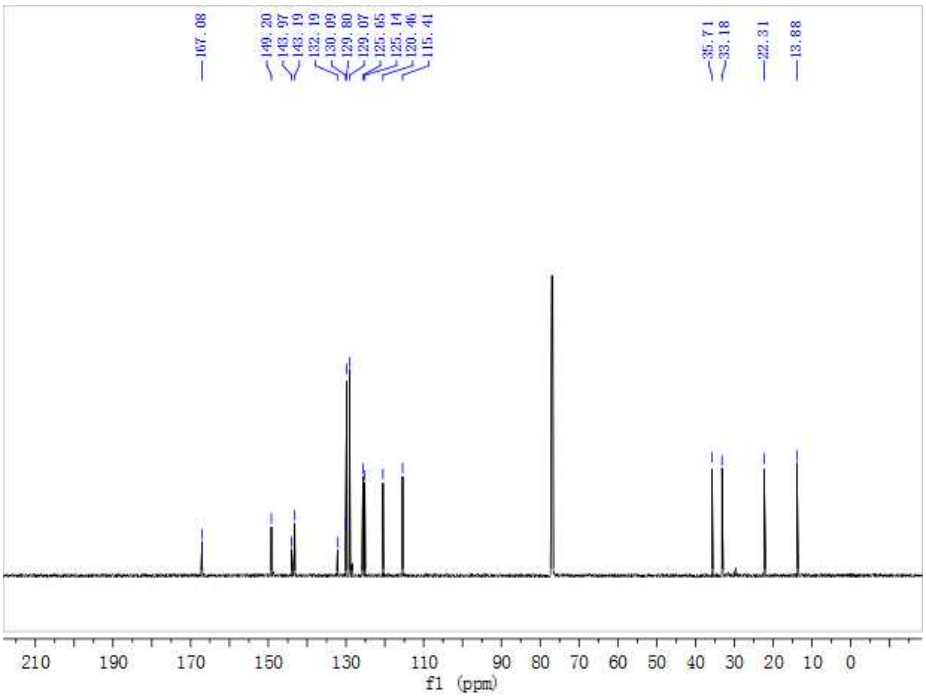
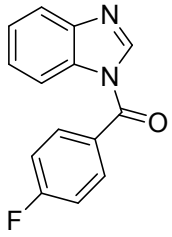




(1H-benzo[d]imidazol-1-yl)(4-fluorophenyl)methanone（CAS：154786-24-0）[8]



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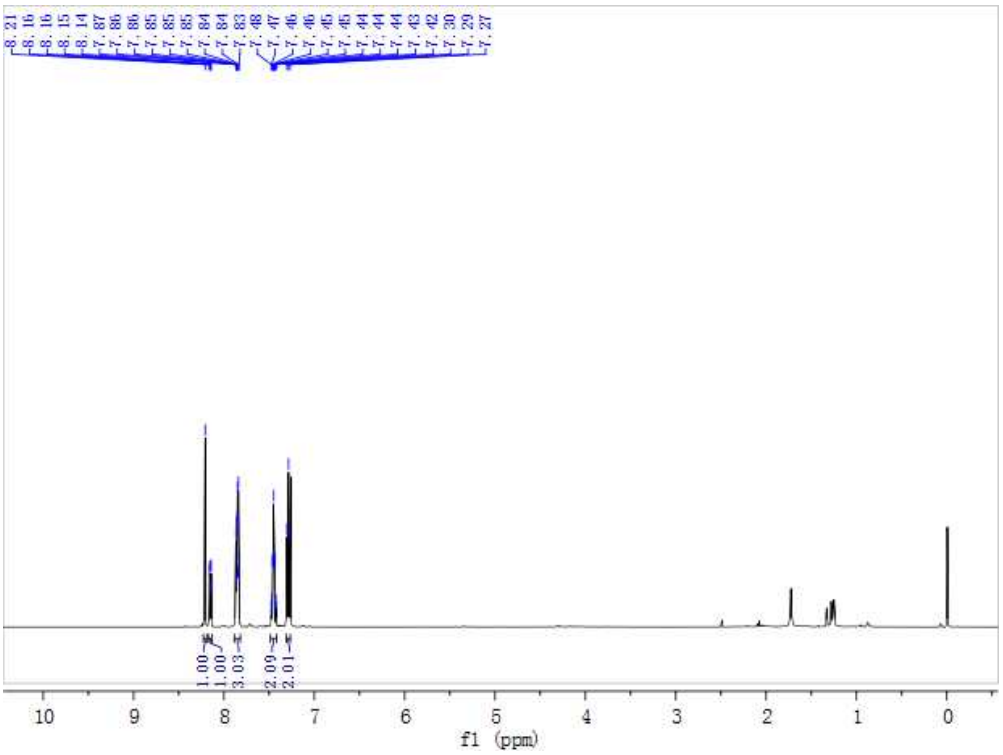
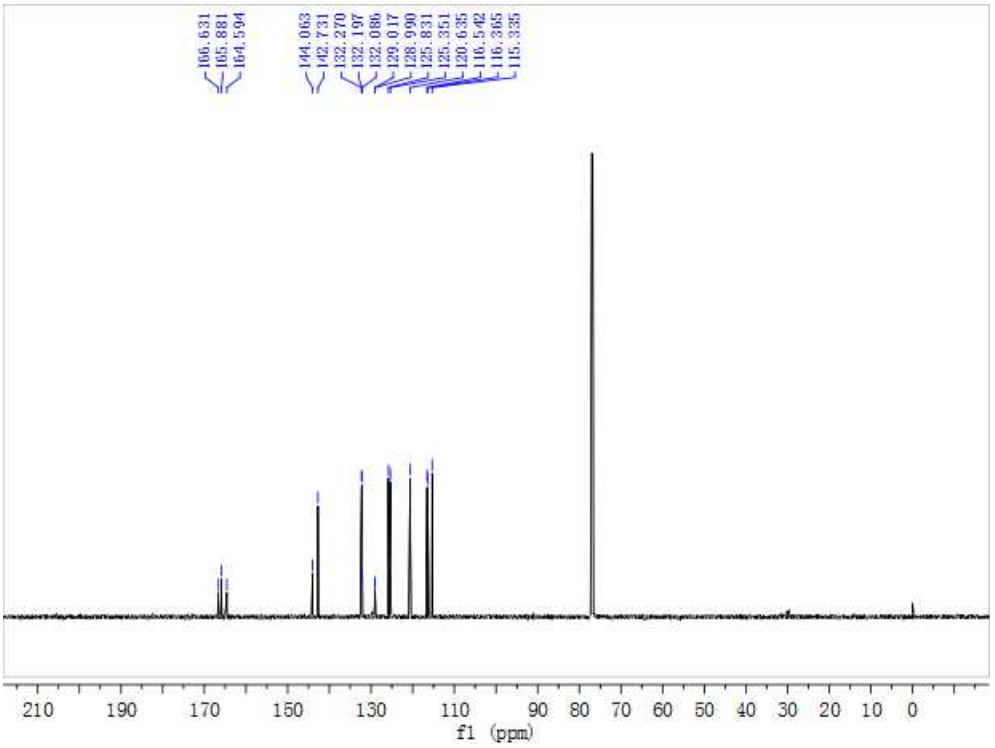
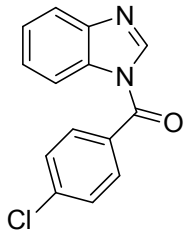




(1H-benzo[d]imidazol-1-yl)(4-chlorophenyl)methanone（CAS：71589-37-2）[9]



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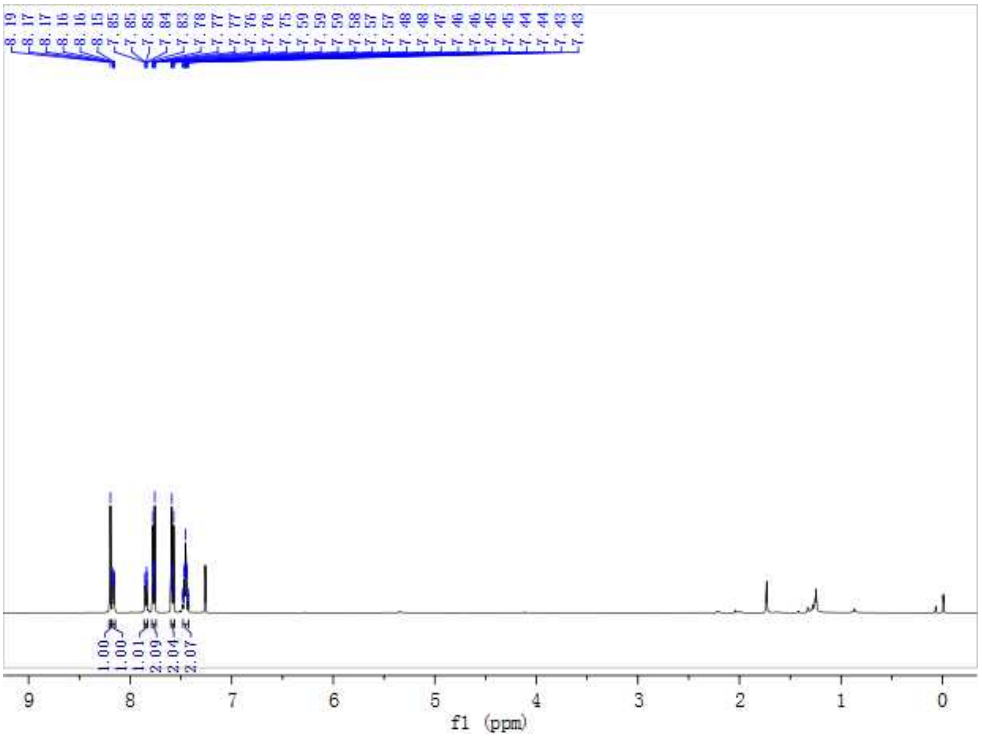
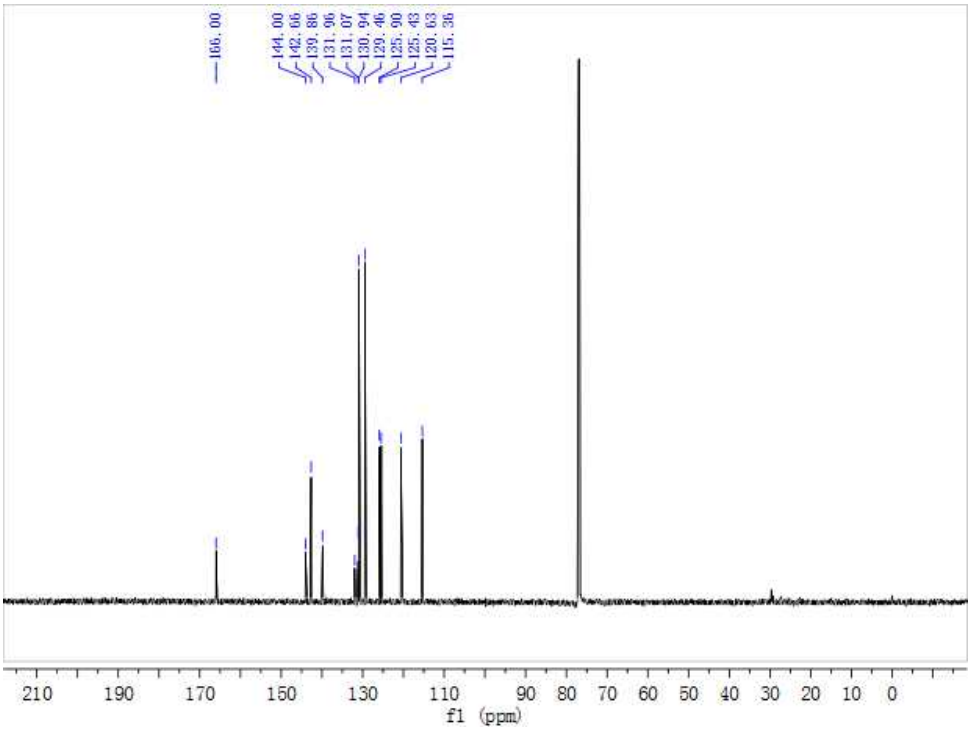
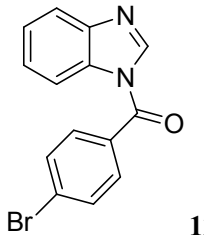




(1H-benzo[d]imidazol-1-yl)(4-bromophenyl)methanone（CAS：304668-33-5）[1]



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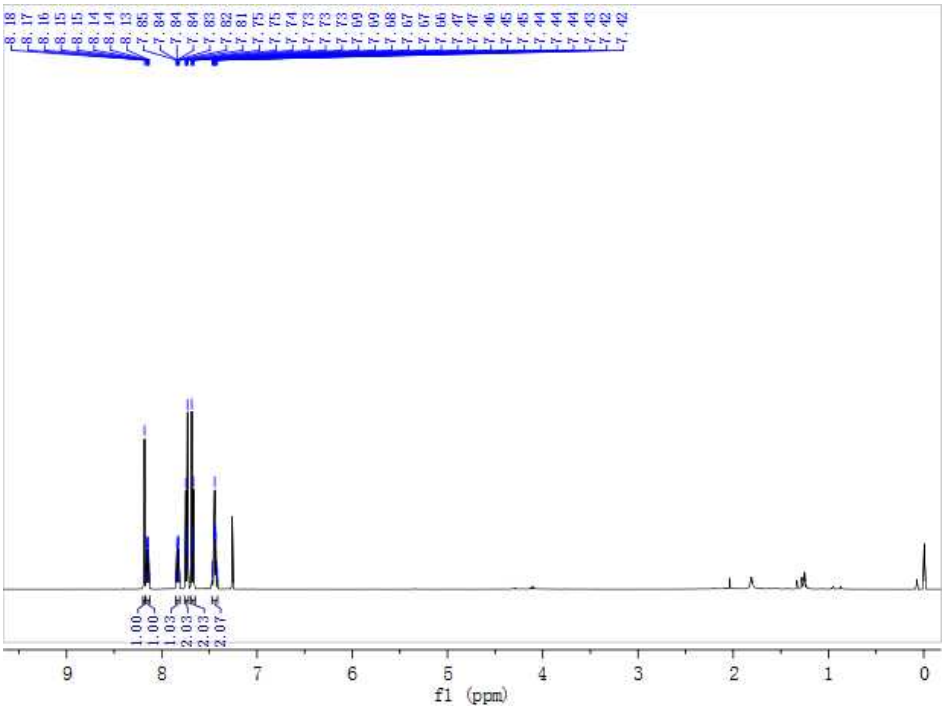
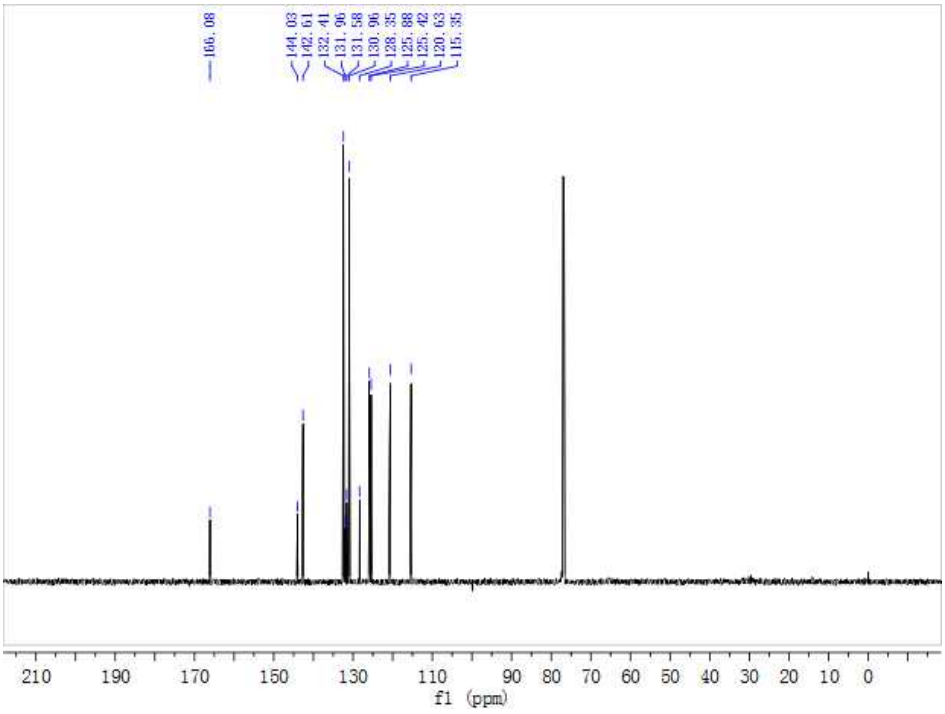
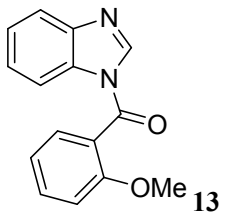




(1H-benzo[d]imidazol-1-yl)(2-methoxyphenyl)methanone（CAS：816441-32-4）[1]



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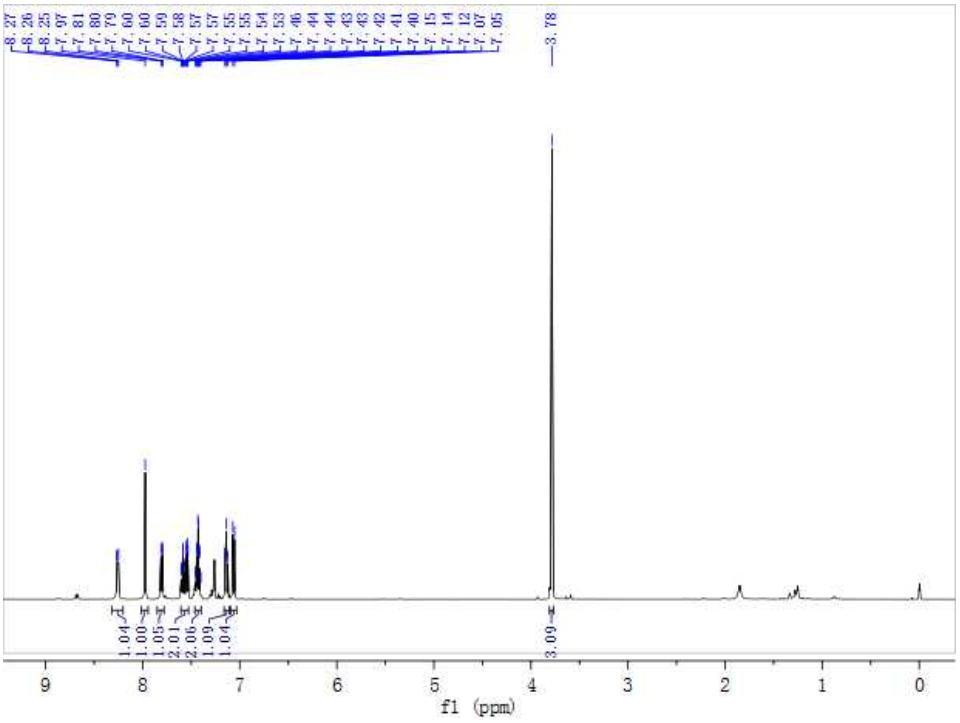
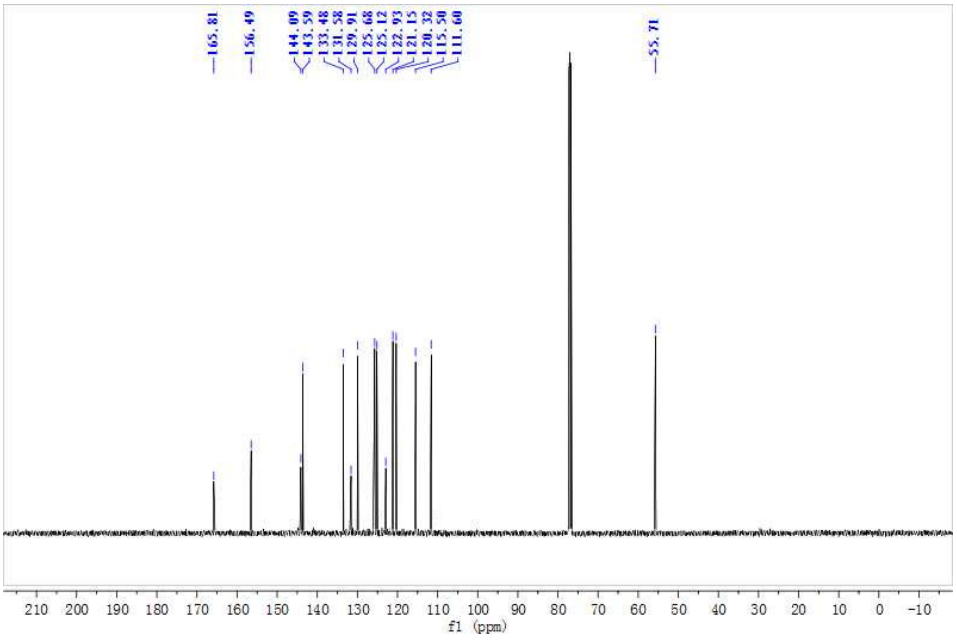
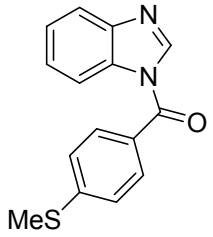




(1H-benzo[d]imidazol-1-yl)(4-(methylthio)phenyl)methanone（new compound）



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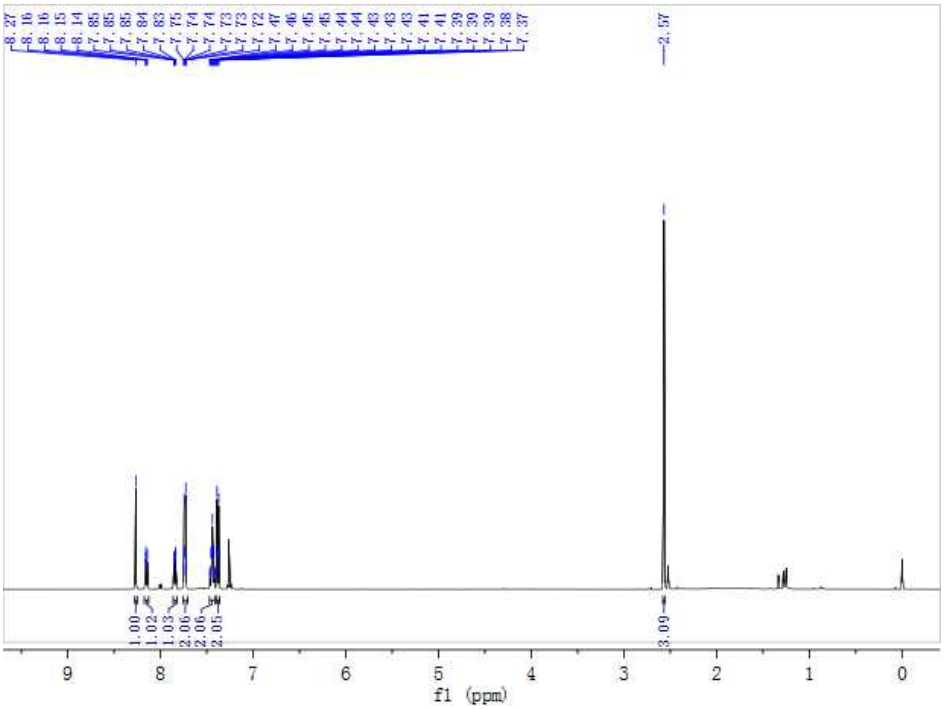
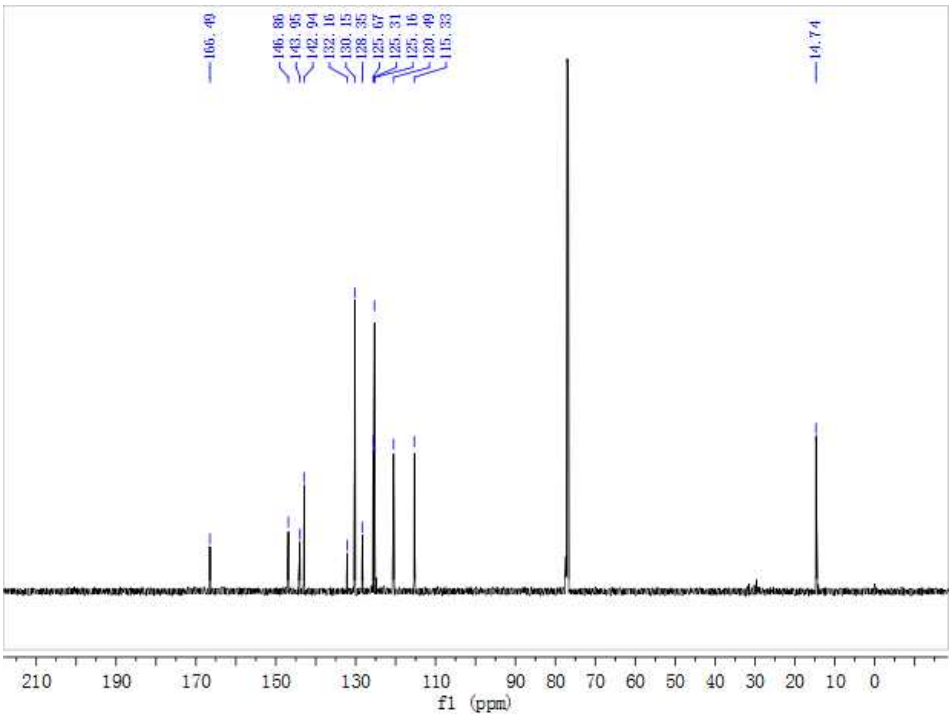
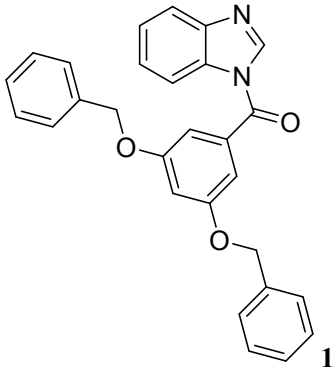




(1H-benzo[d]imidazol-1-yl)(3,5-bis(benzyloxy)phenyl)methanone（new compound）



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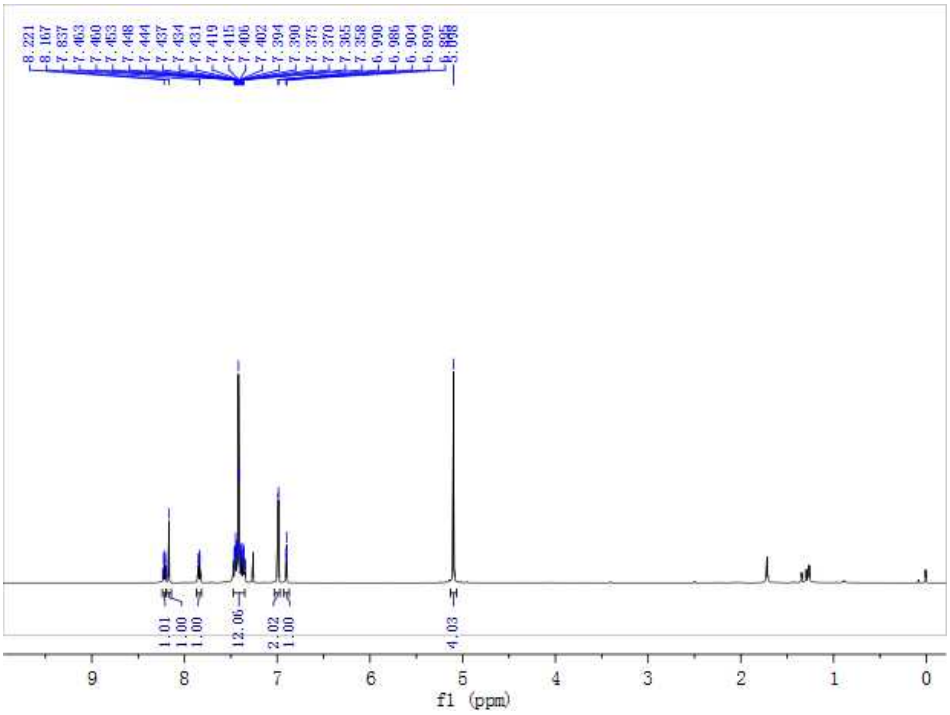
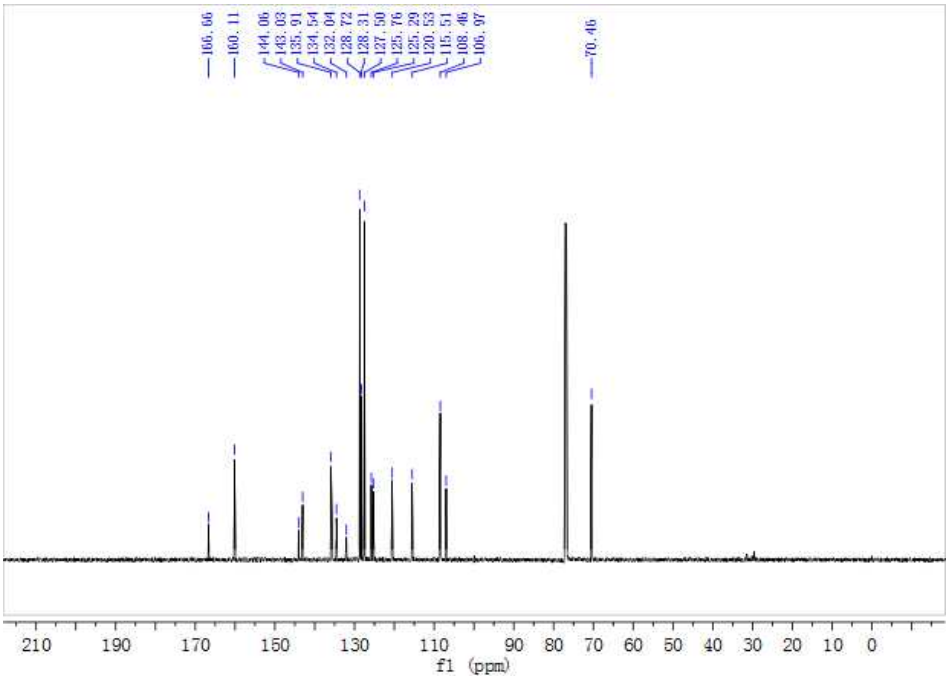
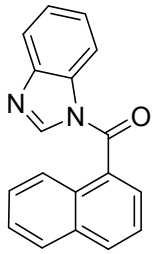




(1H-benzo[d]imidazol-1-yl)(naphthalen-1-yl)methanone（CAS：26670-22-4）[1]



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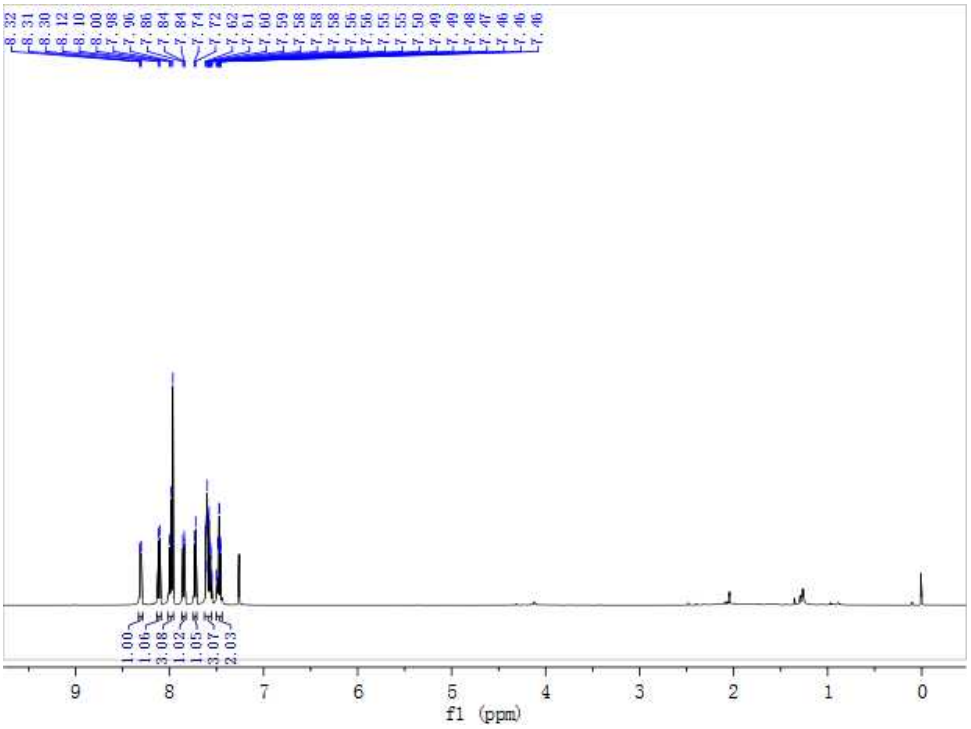
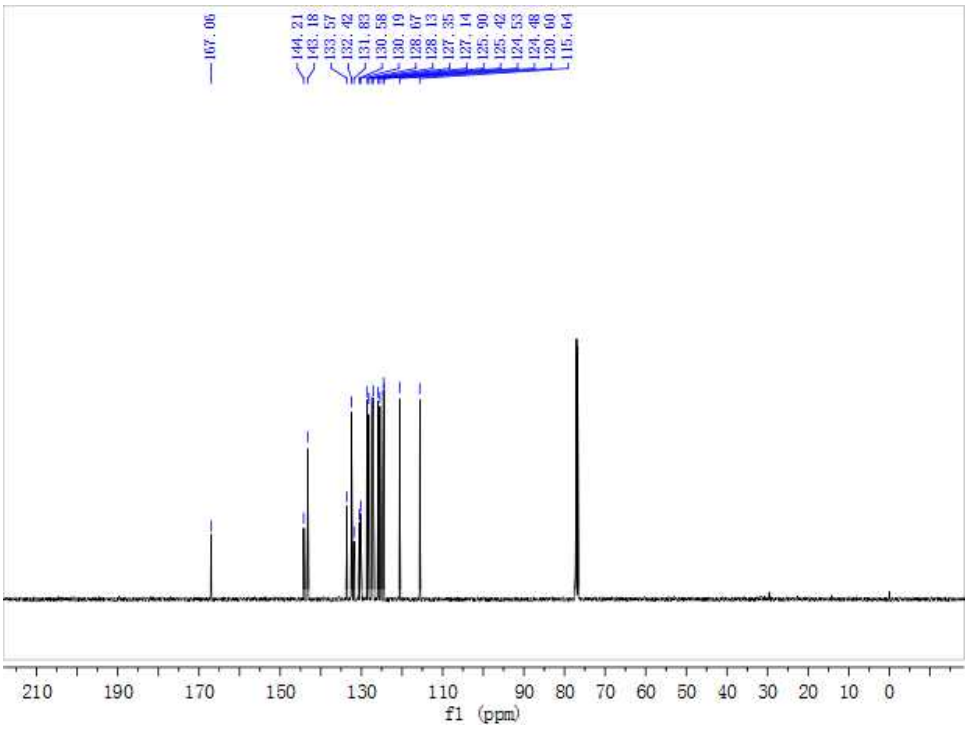
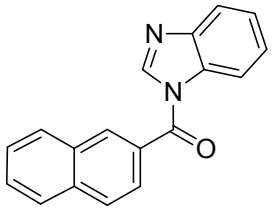




(1H-benzo[d]imidazol-1-yl)(naphthalen-2-yl)methanone（new compound）



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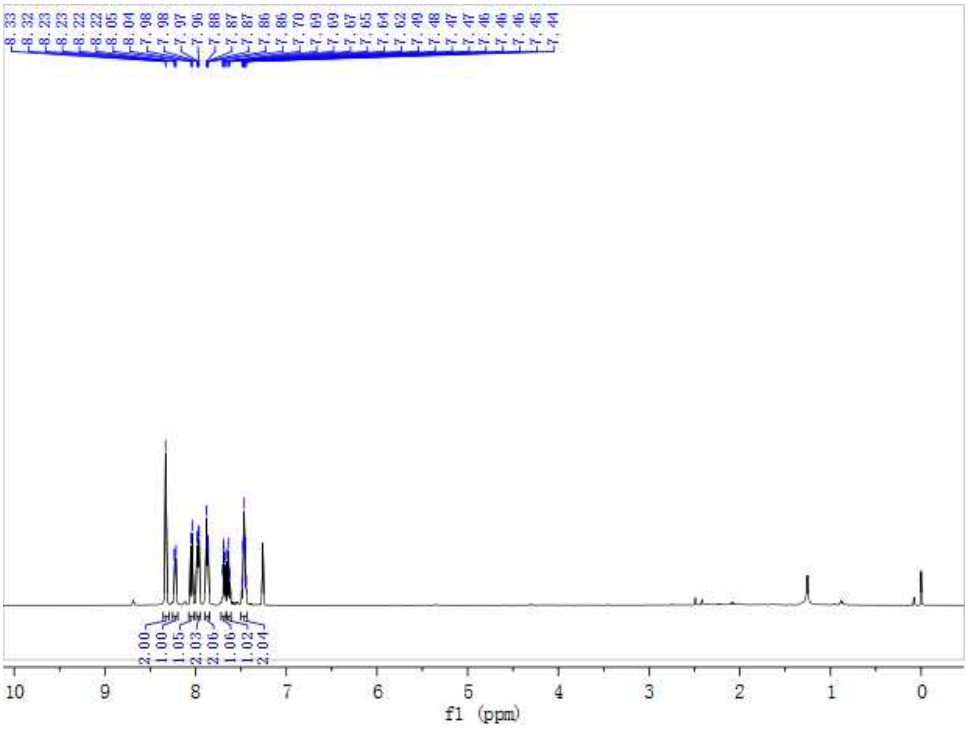
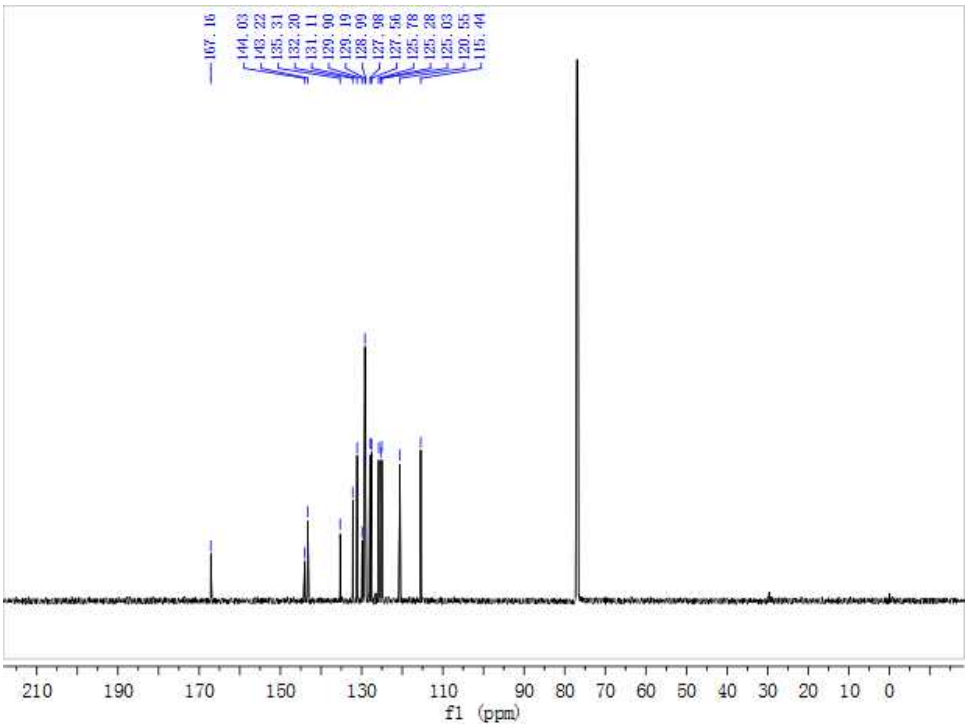
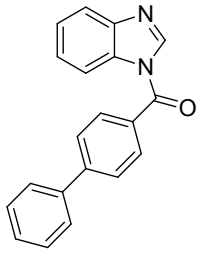




[1,1'-biphenyl]-4-yl(1H-benzo[d]imidazol-1-yl)methanone（CAS：349407-41-6）[11]



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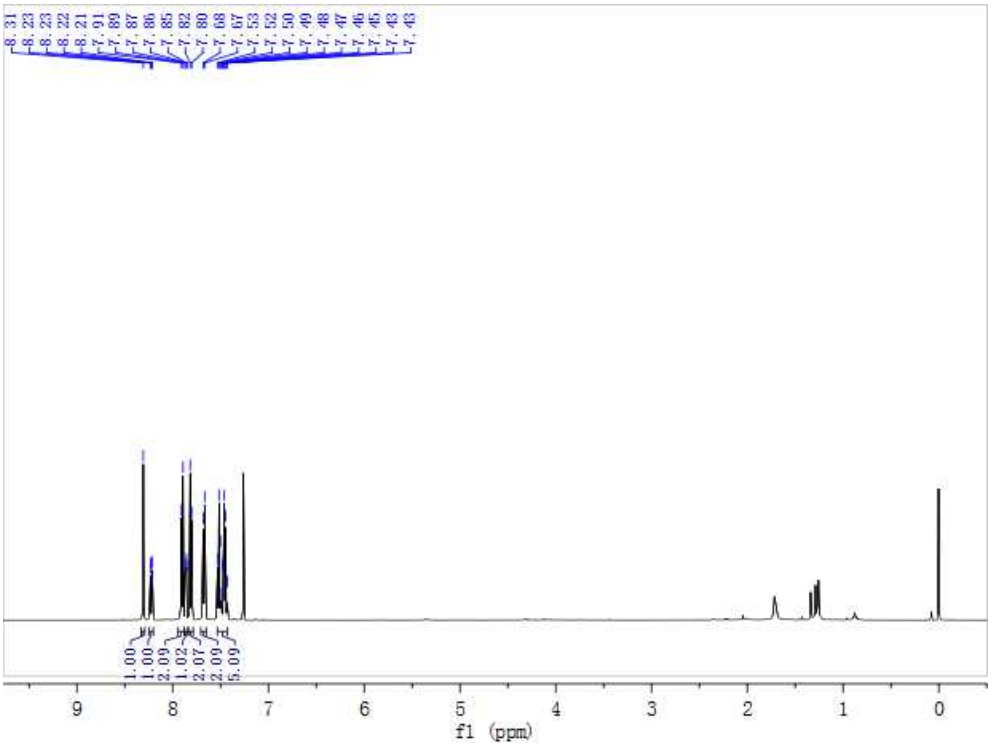
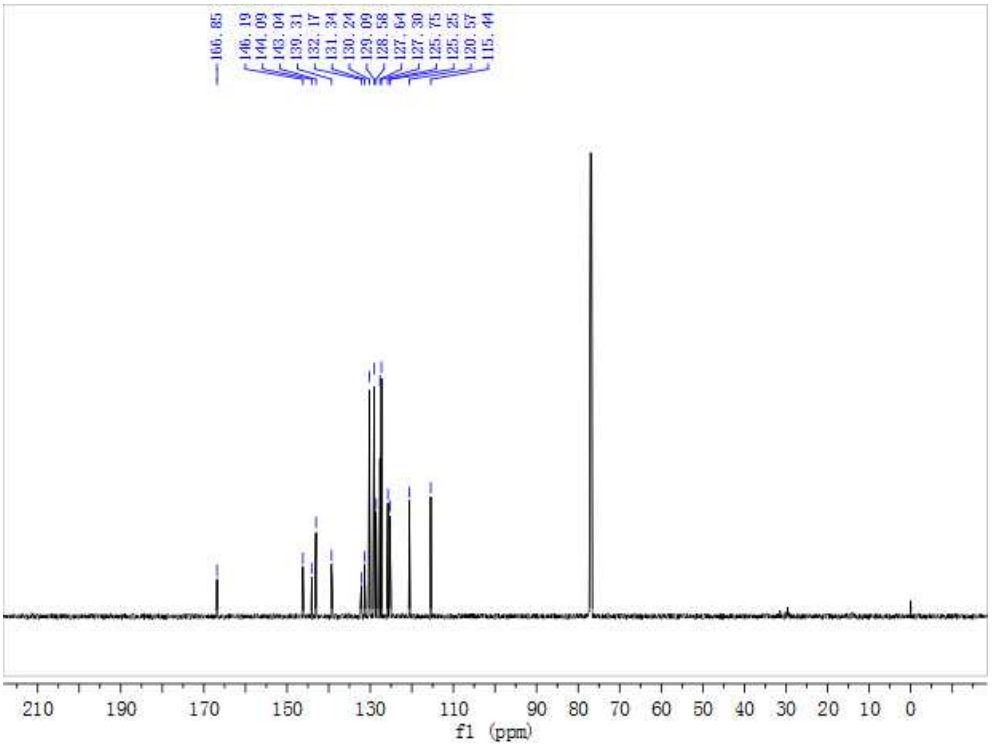
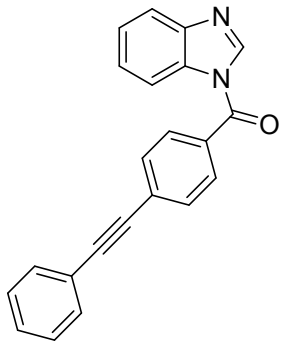




(1H-benzo[d]imidazol-1-yl)(4-(phenylethynyl)phenyl)methanone（new compound）



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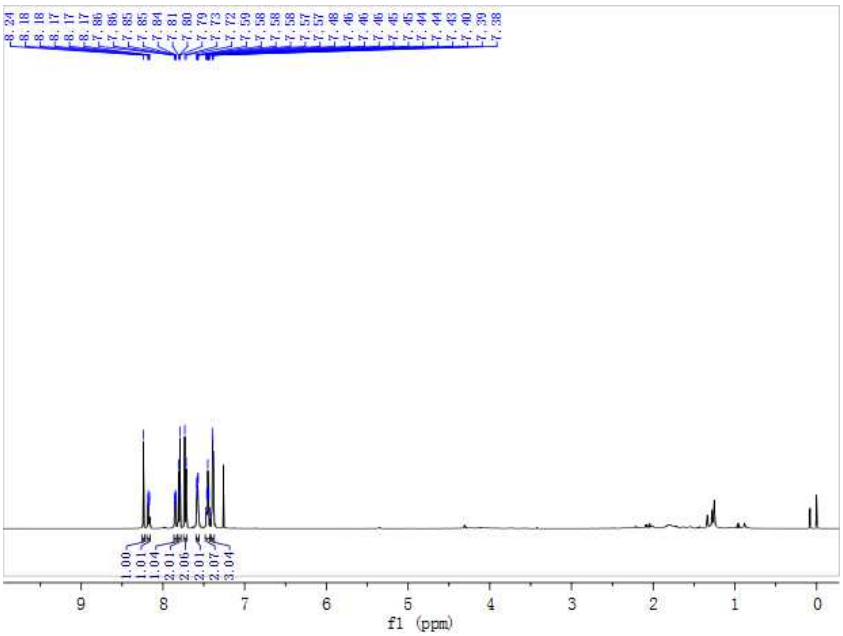
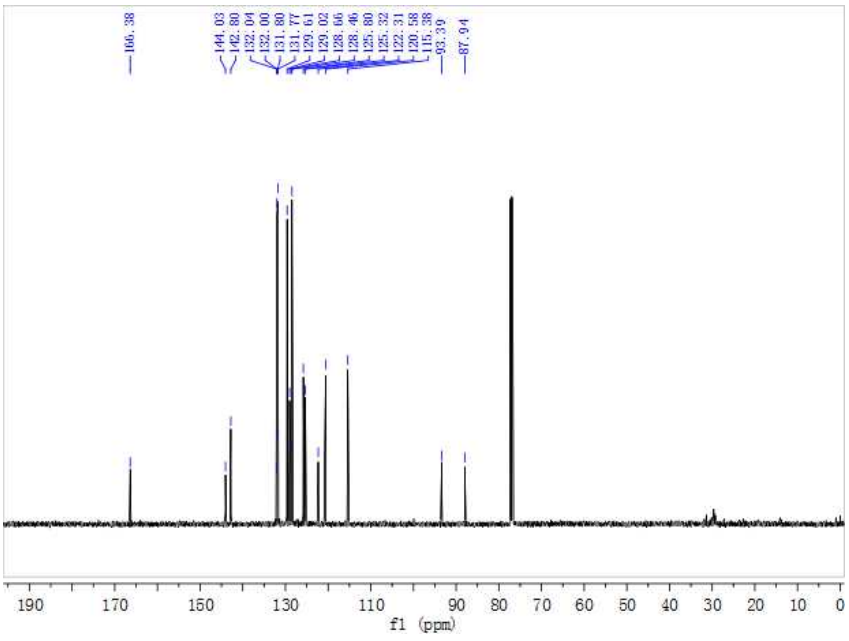
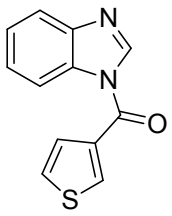




(1H-benzo[d]imidazol-1-yl)(thiophen-3-yl)methanone（new compound）



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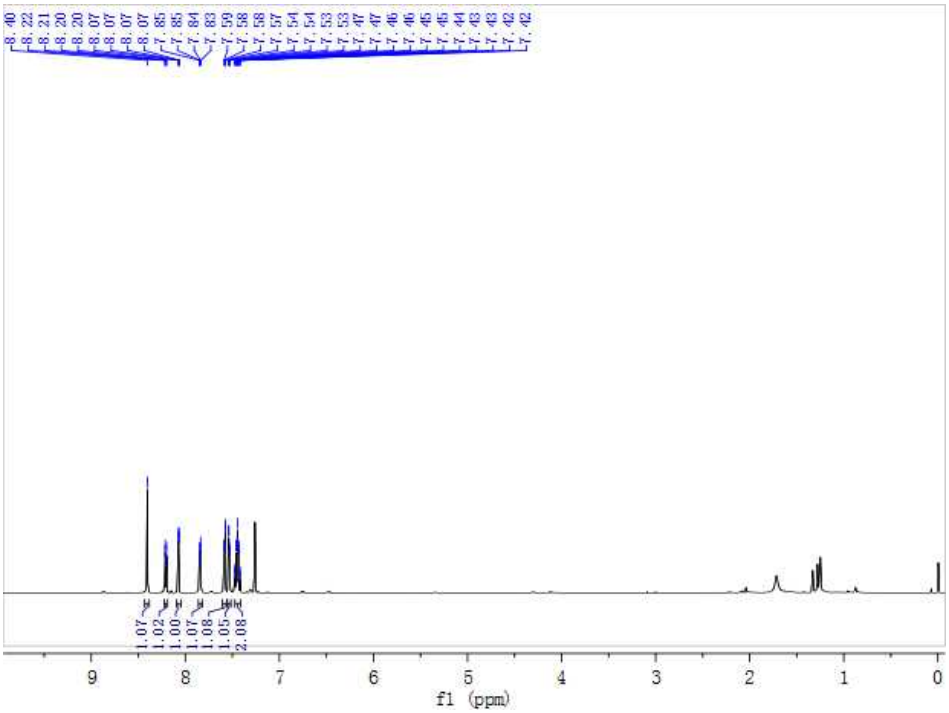
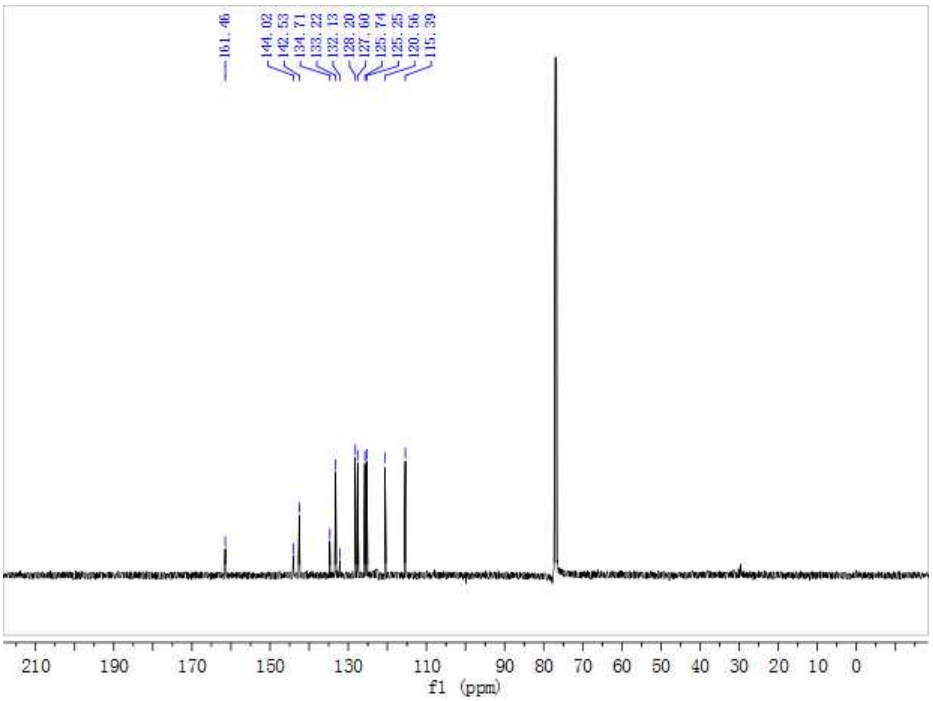
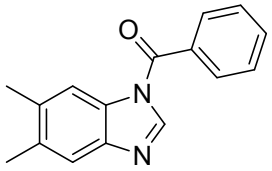




(5,6-dimethyl-1H-benzo[d]imidazol-1-yl)(phenyl)methanone（CAS：16109-46-9）[5]



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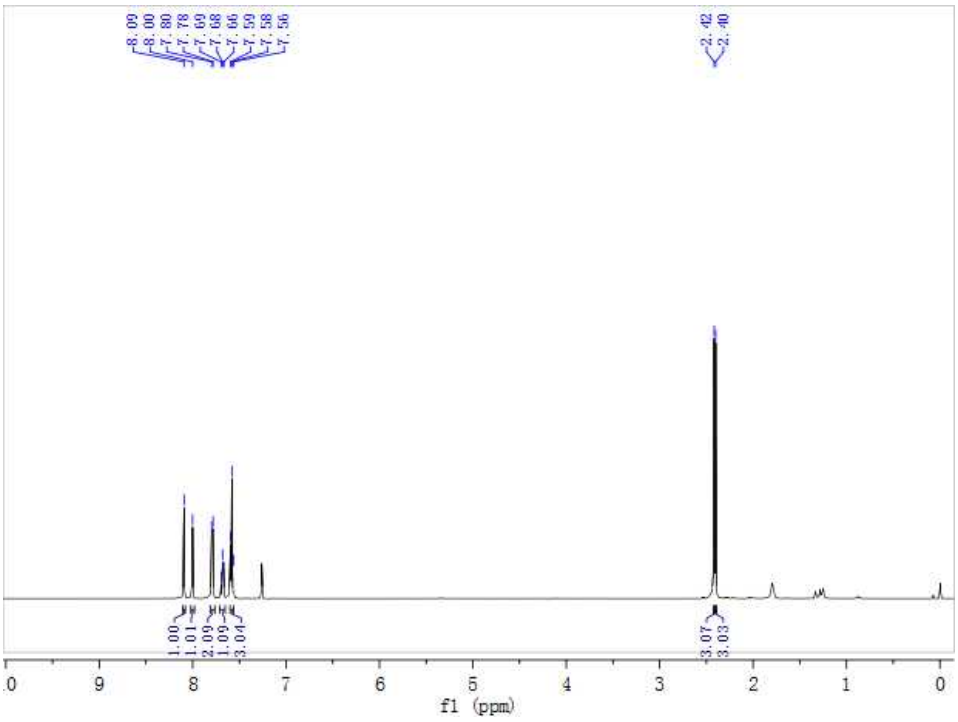
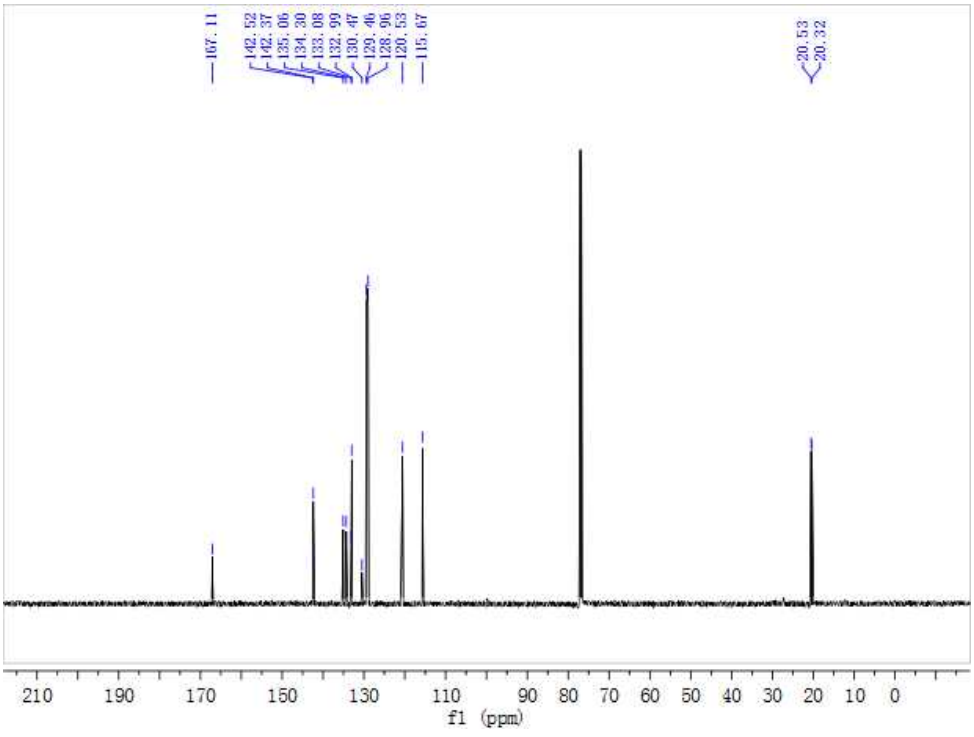
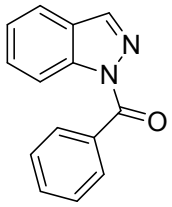




(1H-indazol-1-yl)(phenyl)methanone（CAS：23301-00-0）[3]



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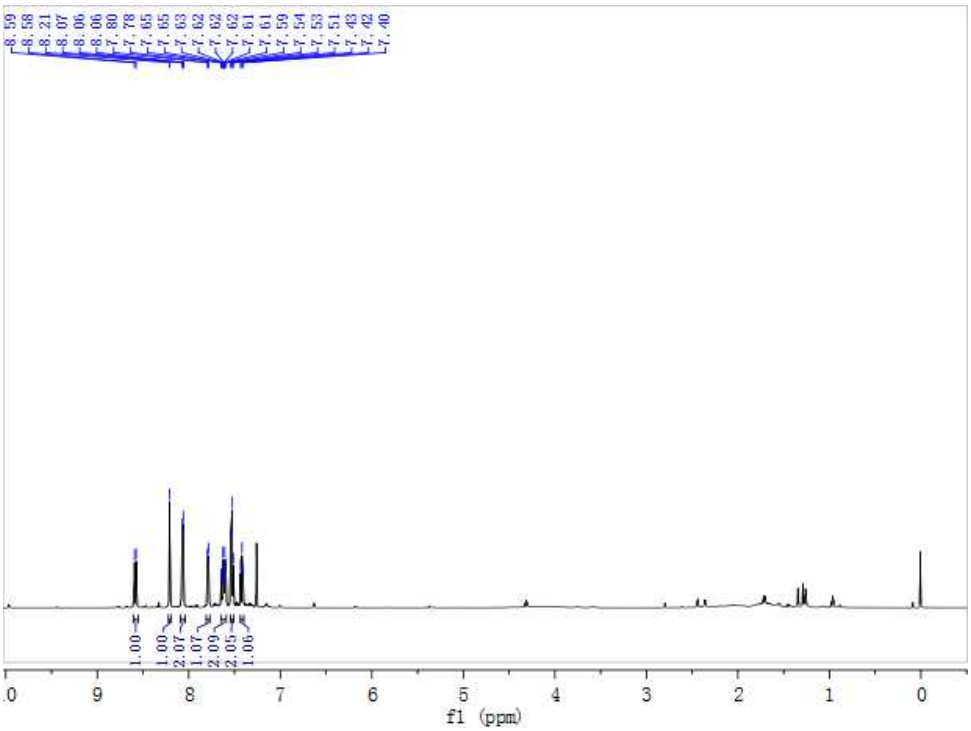
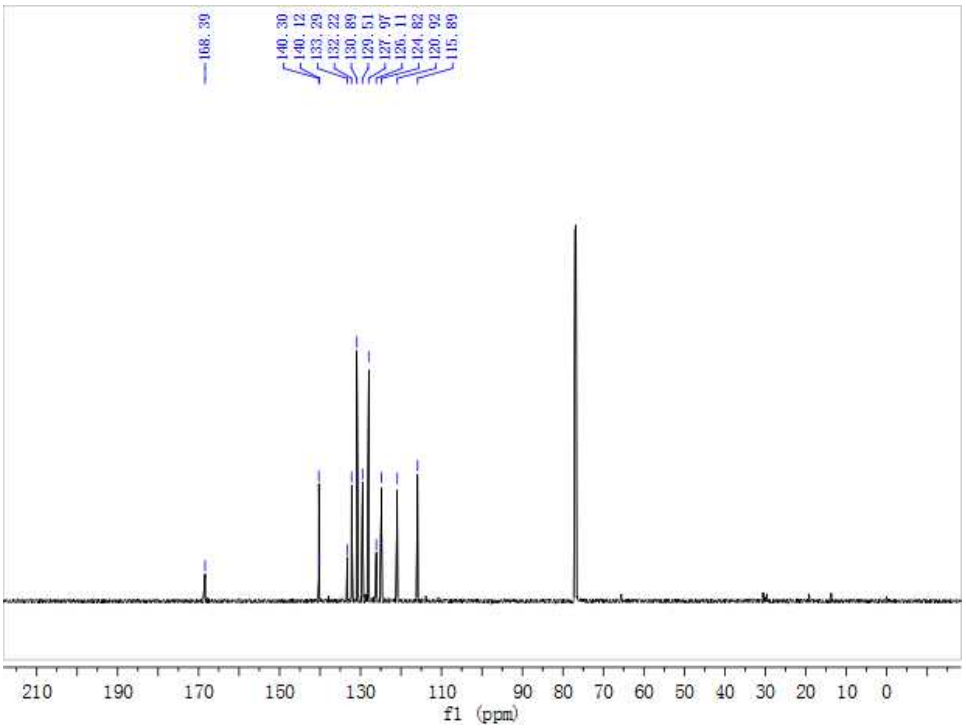
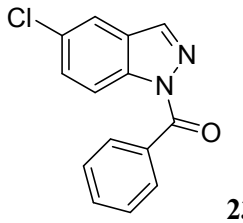




(5-chloro-1H-indazol-1-yl)(phenyl)methanone （new compound）



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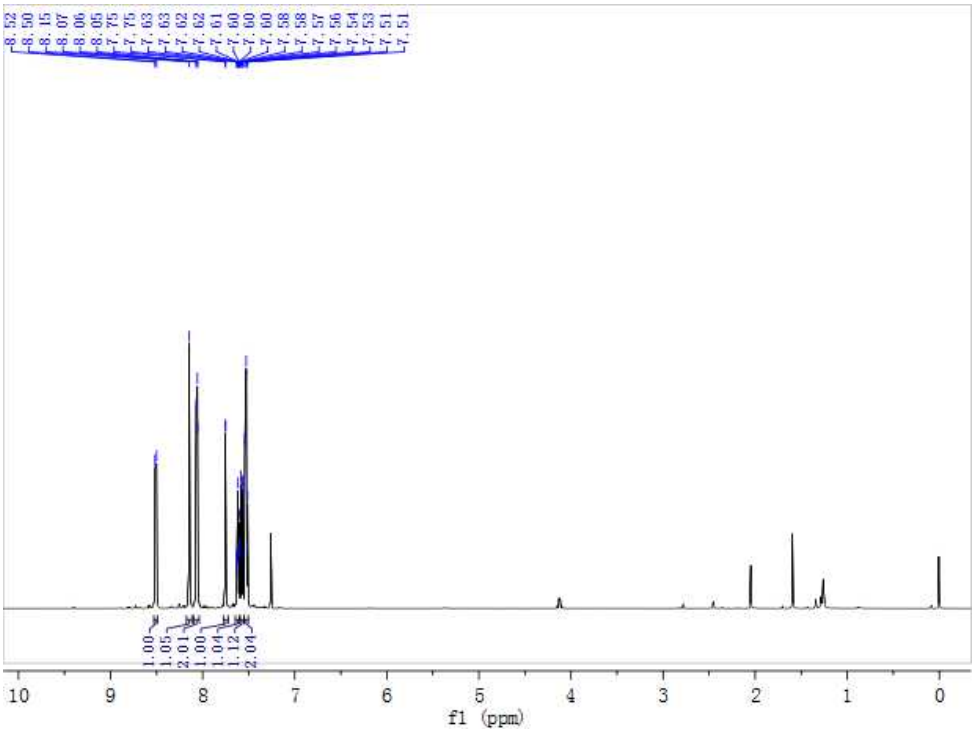
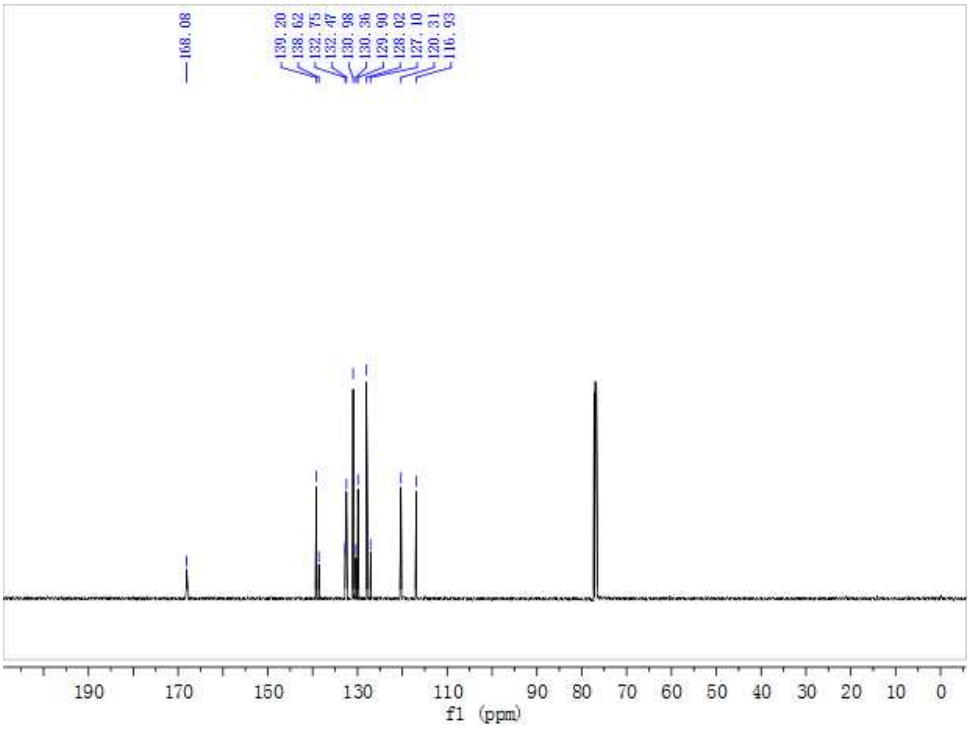
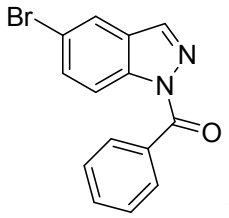




(5-bromo-1H-indazol-1-yl)(phenyl)methanone（CAS：936846-01-4）[2] 915



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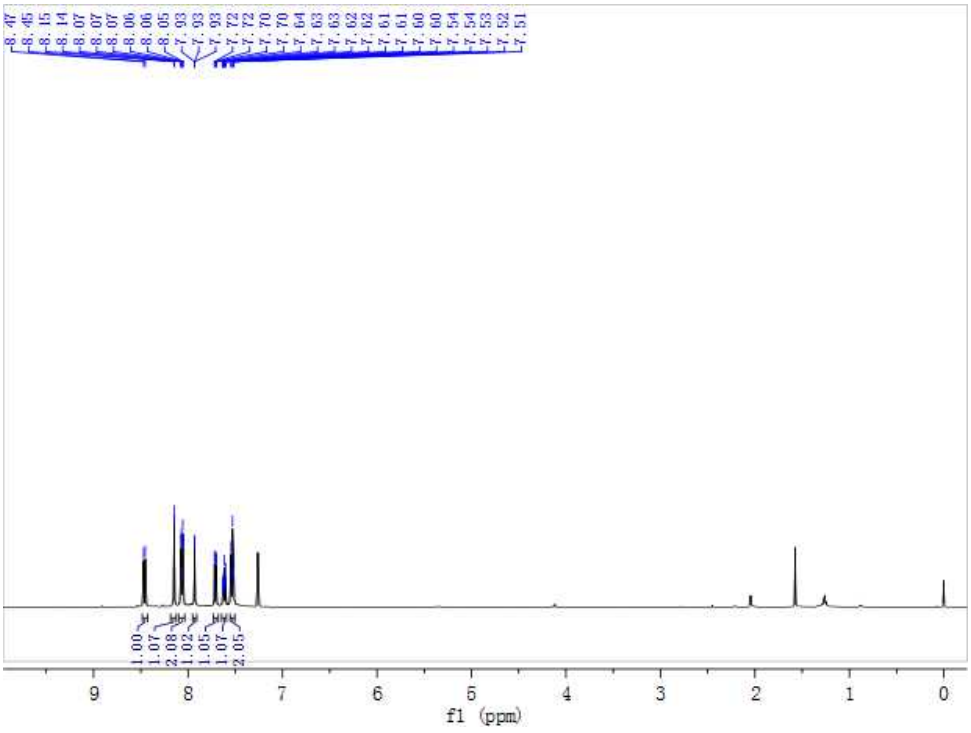
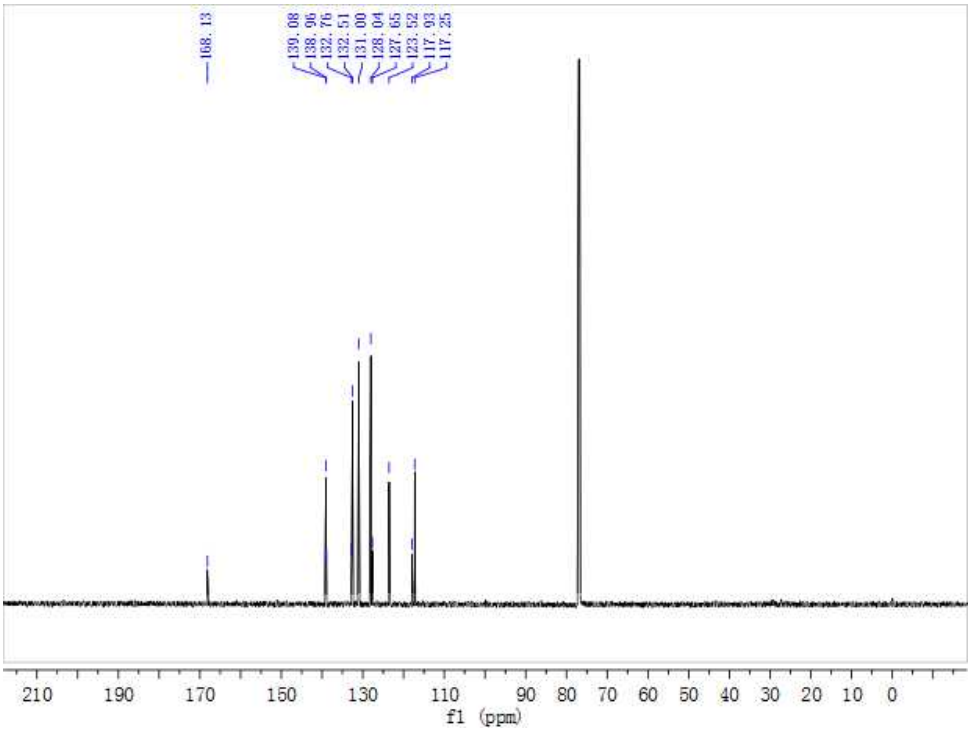
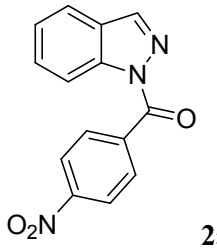




(1H-indazol-1-yl)(4-nitrophenyl)methanone（CAS：500900-65-2）[4]



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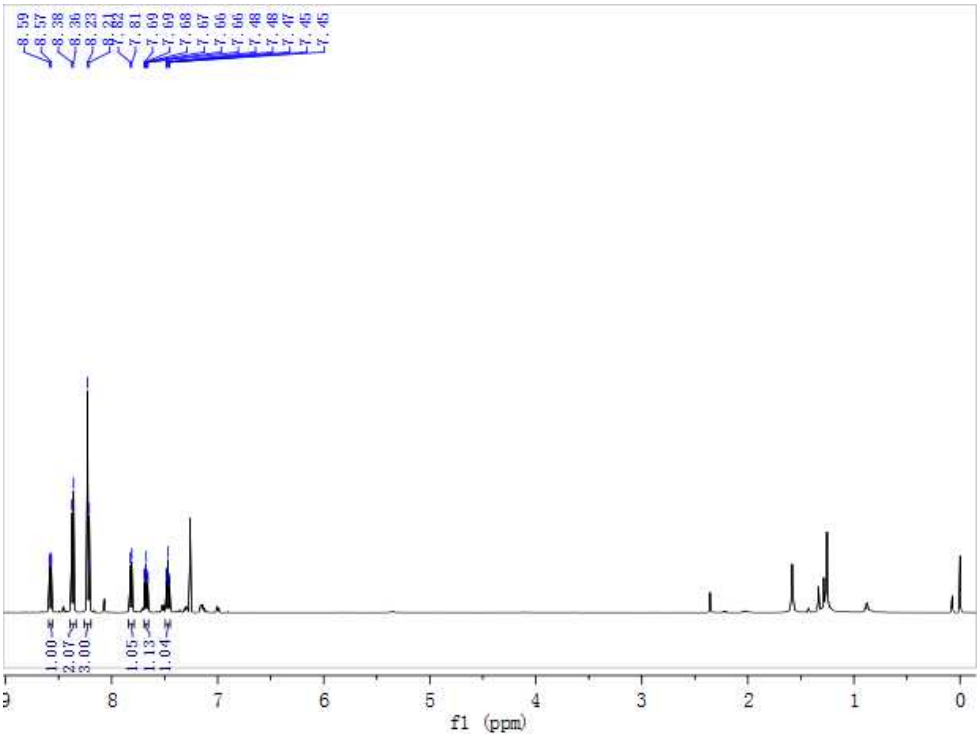
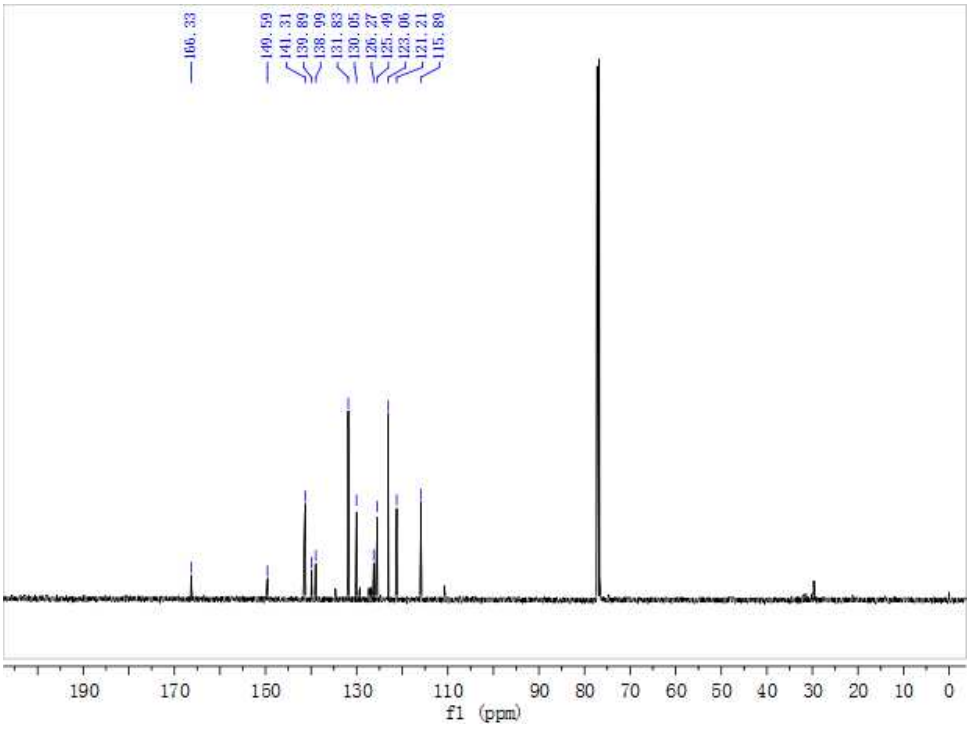
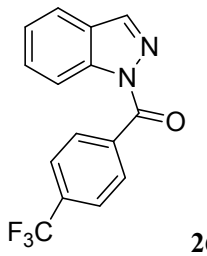




(1H-indazol-1-yl)(4-(trifluoromethyl)phenyl)methanone （new compound）



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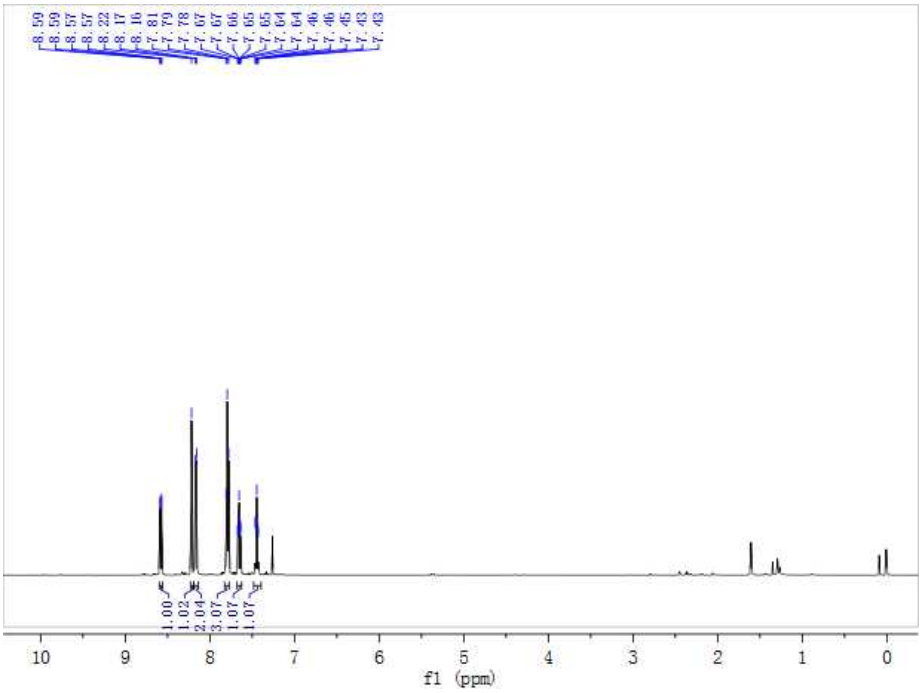
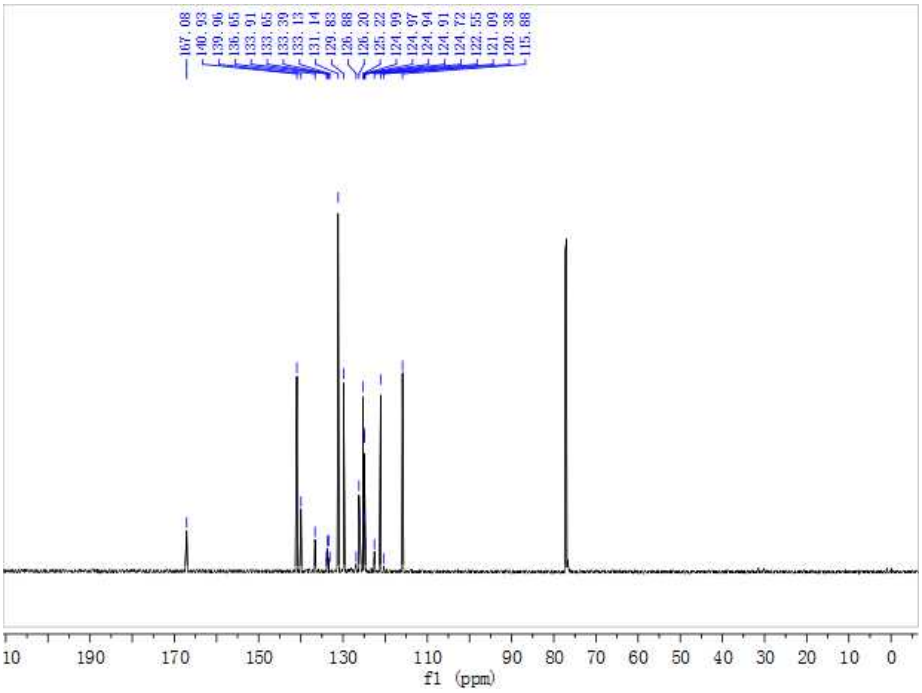
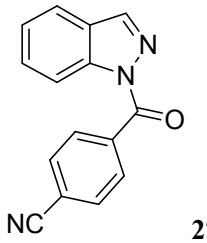




4-(1H-indazole-1-carbonyl)benzonitrile （new compound）



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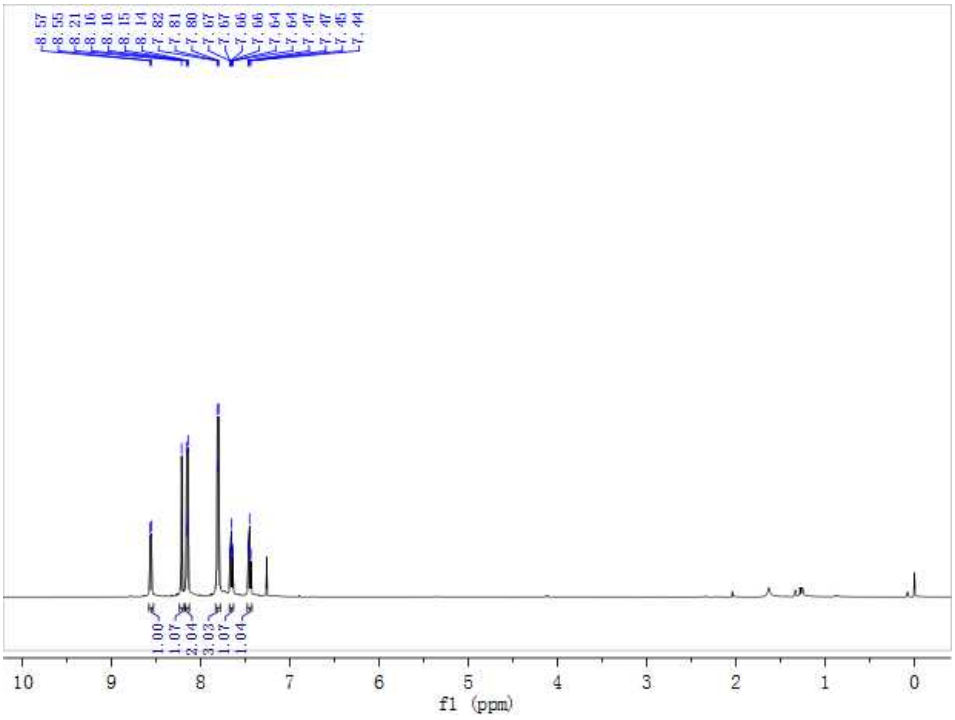
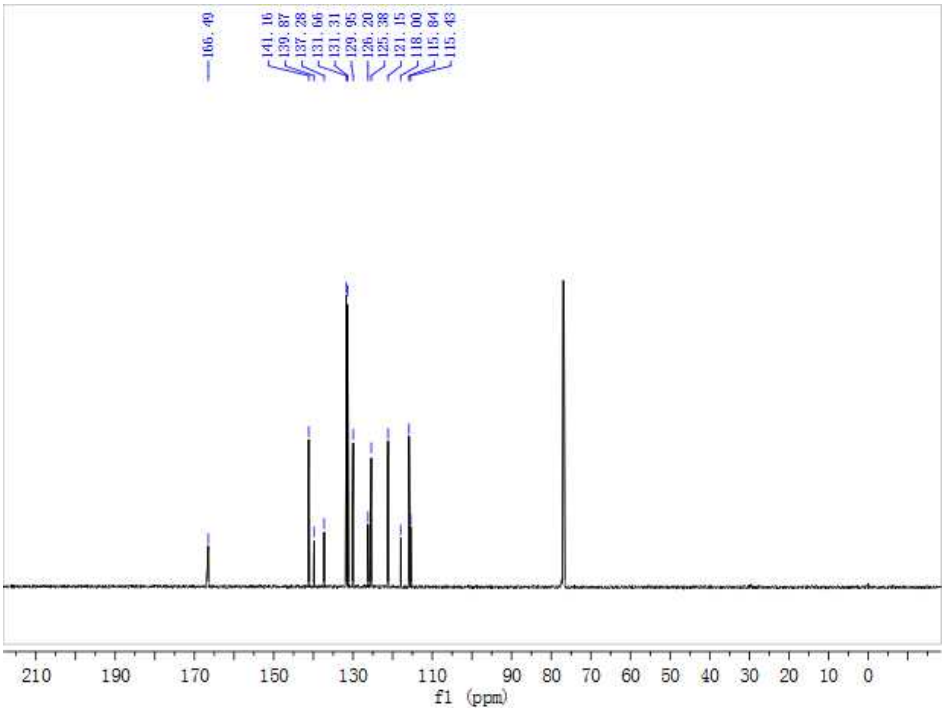
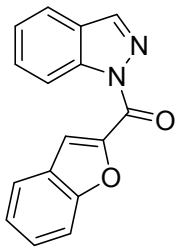




benzofuran-2-yl(1H-indazol-1-yl)methanone （new compound）



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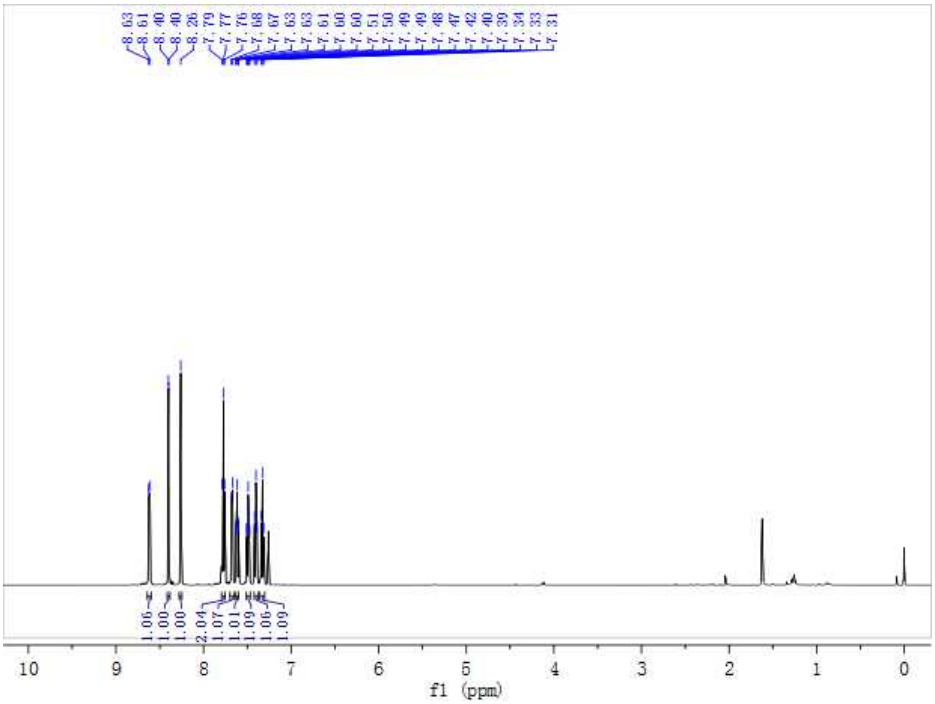
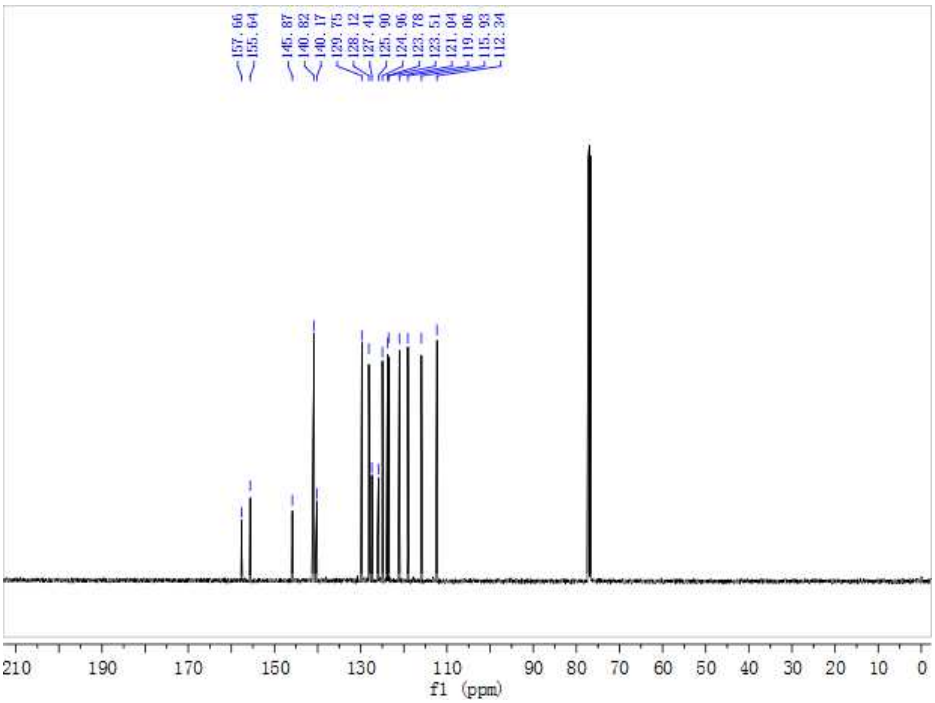
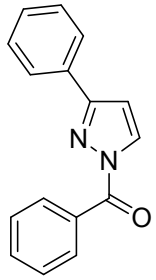




phenyl(3-phenyl-1H-pyrazol-1-yl)methanone（CAS：126382-89-6）[1]



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