

Supporting Information

for

Molecular-oxygen-promoted Cu-catalyzed oxidative direct amidation of nonactivated carboxylic acids with azoles

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Experimental procedures, analytical data and NMR spectra

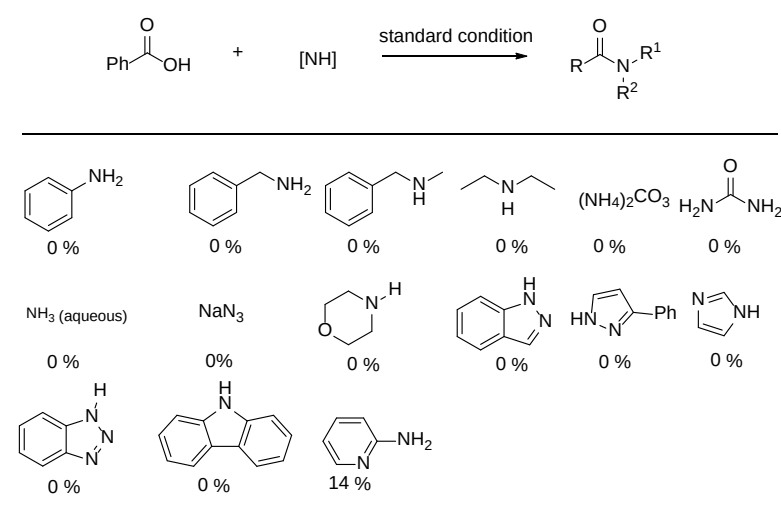
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General information

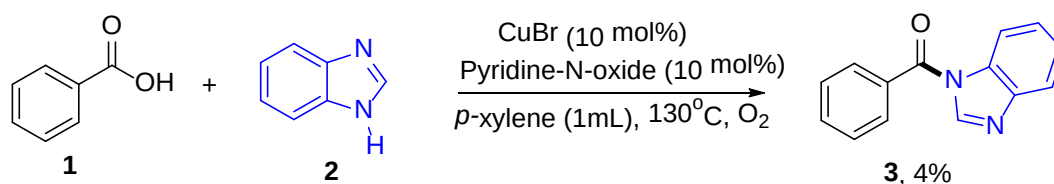
All experiments were conducted with a sealed pressure vessel. Flash column chromatography was performed over silica gel (200–300 mesh). ^1H NMR spectra were recorded on a Bruker AVIII-500M spectrometer, chemical shifts (in ppm) were referenced to CDCl_3 ($\delta = 7.26$ ppm) or $\text{DMSO}-d_6$ ($\delta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

The compatibility of reaction with other amines

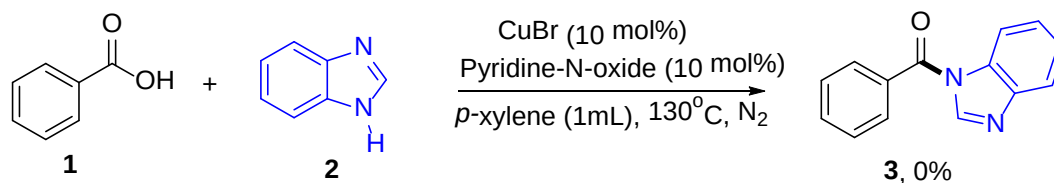


Scheme S1: The compatibility of reaction with other amines.

Experiments with pyridine *N*-oxide as possible intermediate

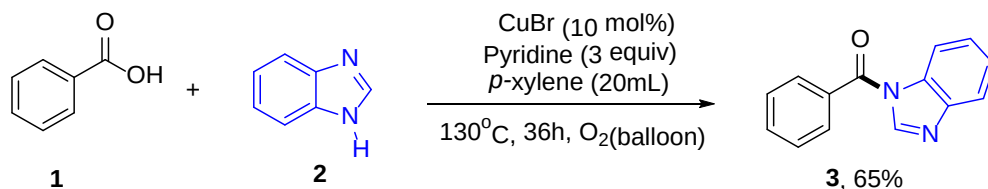


A sealed pressure vessel was charged with benzoic acid (**1**, 30.0 mg, 0.25 mmol), benzimidazole (**2**, 60 mg, 0.5 mmol), CuBr (3.6 mg, 0.025 mmol), pyridine *N*-oxide (2.3 mg, 0.025 mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O_2 for 20 hours. After cooling down to room temperature, the mixture was measured by GC without further purification. Most of starting materials remain, and only 4% yield of product **3** generated.



A sealed pressure vessel was charged with benzoic acid (**1**, 30.0 mg, 0.25 mmol), benzimidazole (**2**, 60 mg, 0.5 mmol), CuBr (3.6 mg, 0.025 mmol), pyridine *N*-oxide (2.3 mg, 0.025 mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under N₂ for 20 hours. After cooling down to room temperature, the mixture was measured by GC without further purification. Starting materials remain, and no product generated at all.

Preparative scale of the reaction



A sealed pressure vessel was charged with benzoic acid (**1**, 610.0 mg, 5 mmol), benzimidazole (**2**, 1180 mg, 10 mmol), CuBr (71.7 mg, 0.5 mmol), pyridine (1200 mg, 15 mmol), and *p*-xylene (20 mL). The resulting solution was stirred at 130 °C under O₂ (balloon) monitored by TLC and GC for 32 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 720 mg of (1*H*-benzo[*d*]imidazol-1-yl)(phenyl)methanone (**3**) in 65% isolated yield as a white solid.

Procedure and characterization data for amidation reactions

Typical procedure and characterization data for amidation reaction

A sealed pressure vessel was charged with benzoic acid (**1**, 30.0 mg, 0.25 mmol), benzimidazole (**2**, 60 mg, 0.5 mmol), CuBr (3.6 mg, 0.025 mmol), pyridine (60 mg, 0.75 mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under O₂ 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 47 mg of (1*H*-benzo[*d*]imidazol-1-yl)(phenyl)methanone (**3**) in 85% isolated yield as a white solid.

(1*H*-Benzo[*d*]imidazol-1-yl)(phenyl)methanone (**3**, CAS: 62573-86-8)¹

85% yield (47 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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).; ¹³C NMR: (125 MHz, CDCl₃, ppm) δ 167.1 (s), 144.0 (s), 143.1 (s), 133.2 (s), 132.8 (s), 132.1 (s), 129.5 (s), 129.0 (s), 125.8 (s), 125.3 (s), 120.5 (s), 115.4 (s). mp = 70.1 - 71.2 °C.

(1*H*-Benzo[*d*]imidazol-1-yl)(*p*-tolyl)methanone (**4**, CAS: 28997-00-4)¹

75% yield (44 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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).; ¹³C NMR: (125 MHz, CDCl₃, ppm) δ 167.1 (s), 144.3 (s), 144.1 (s), 143.1 (s), 132.2 (s), 130.0 (s), 129.8 (s), 129.7 (s), 125.6 (s), 125.1 (s), 120.5 (s), 115.4 (s), 21.7 (s). mp = 92.7 - 94.5 °C.

(1*H*-Benzo[*d*]imidazol-1-yl)(4-ethylphenyl)methanone (**5**, new compound)

92% yield (57 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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 (s), 150.4 (s), 144.1 (s), 143.2 (s), 132.2 (s), 130.2 (s), 129.9 (s), 128.5 (s), 125.6 (s), 125.1 (s), 120.5 (s), 115.4 (s), 29.0 (s), 15.1 (s). new compound, HRMS m/z (EI) calcd. for $\text{C}_{16}\text{H}_{14}\text{N}_2\text{O}$ M^+ 250.1106, found 250.1111. mp = 90.1 - 91.8 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-isopropylphenyl)methanone (6, CAS: 901440-49-1)

71% yield (47 mg) as a white solid. ^1H 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 (s), 154.9 (s), 144.0 (s), 143.2 (s), 132.2 (s), 130.3 (s), 129.9 (s), 127.2 (s), 125.6 (s), 125.1 (s), 120.5 (s), 115.4 (s), 34.3 (s), 23.6 (s). mp = 75.5 - 76.5 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-propylphenyl)methanone (7, new compound)²

64% yield (43 mg) as a white solid. ^1H NMR (500 MHz, CDCl_3) δ 8.27 (s, 1H), 8.23 - 8.15 (m, 1H), 7.89 - 7.80 (m, 1H), 7.77 - 7.65 (m, 2H), 7.49 - 7.41 (m, 2H), 7.39 (d, $J = 8.2$ Hz, 2H), 2.76 - 2.68 (m, 2H), 1.76 - 1.68 (m, 2H), 0.99 (t, $J = 7.3$ Hz, 3H).; ^{13}C NMR (126 MHz, CDCl_3) δ 167.1 (s), 149.0 (s), 149.0 (s), 143.2 (s), 132.2 (s), 130.2 (s), 129.8 (s), 129.1 (s), 125.7 (s), 125.2 (s), 120.5 (s), 115.4 (s), 38.0 (s), 24.2 (s), 13.8 (s). new compound, HRMS m/z (EI) calcd. for $\text{C}_{17}\text{H}_{16}\text{N}_2\text{O}$ M^+ 264.1263, found 264.1262. mp = 53.8 - 54.9 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-(tert-butyl)phenyl)methanone (8, CAS: 20208-57-5)

95% yield (66 mg) as a white solid. ^1H 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 (s), 157.2 (s), 144.1 (s), 143.2 (s), 132.2 (s), 130.0 (s), 129.7 (s), 126.0 (s), 125.7 (s), 125.1 (s), 120.5 (s), 115.5 (s), 35.3 (s), 31.1 (s). mp = 131.3 - 132.8 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-butylphenyl)methanone (9, new compound)

67% yield (46 mg) as a white solid. ^1H NMR: (500 MHz, CDCl_3 , ppm) δ 8.27 (s, 1H), 8.19 (dd, $J = 6.9$, 2.0 Hz, 1H), 7.88 - 7.82 (m, 1H), 7.73 (d, $J = 8.1$ Hz, 2H), 7.44 (qd, $J = 7.4$, 6.0 Hz, 2H), 7.39 (d, $J = 8.0$ Hz, 2H), 3.00 (t, $J = 7.5$ Hz, 2H), 1.68 - 1.65 (m, 2H), 1.42 - 1.38 (m, 2H), 0.96 (t, $J = 7.4$ Hz, 3H).; ^{13}C NMR: (125 MHz, CDCl_3 , ppm) δ 167.1 (s), 149.2 (s), 144.0 (s), 143.2 (s), 132.2 (s), 130.1 (s), 129.8 (s), 129.1 (s), 125.7 (s), 125.1 (s), 120.5 (s), 115.4 (s), 35.7 (s), 33.2 (s), 22.3 (s), 13.9 (s). new compound, HRMS m/z (EI) calcd. for $\text{C}_{18}\text{H}_{18}\text{N}_2\text{O}$ M^+ 278.1419, found 278.1411. mp = 37.1-38.9 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-fluorophenyl)methanone (10, CAS: 154786-24-0)⁴

53% yield (32 mg) as a white solid. ^1H 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 165.9 (s), 165.6(d, $J= 254.63\text{Hz}$), 144.1 (s), 142.7 (s), 132.3(d, $J= 9.13\text{Hz}$), 132.1 (s), 129.5(d, $J= 3.38\text{Hz}$), 125.8 (s), 125.4 (s), 120.6 (s), 116.5(d, $J= 22.13\text{Hz}$), 115.3 (s). mp = 74.8 - 76.1 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-chlorophenyl)methanone (11, CAS: 71589-37-2)⁵

52% yield (34 mg) as a white solid. ^1H 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 166.0 (s), 144.0 (s), 142.7 (s), 139.9 (s), 132.0 (s), 131.1 (s), 130.9 (s), 129.5 (s), 125.9 (s), 125.4 (s), 120.6 (s), 115.4 (s). mp = 149.3 - 150.9 $^\circ\text{C}$.

(1H-Benzo[d]imidazol-1-yl)(4-bromophenyl)methanone (12, CAS: 304668-33-5)¹

40% yield (30 mg) as a white solid. ^1H 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).; ¹³C NMR: (125 MHz, CDCl₃, ppm) δ 166.1 (s), 144.0 (s), 142.6 (s), 132.4 (s), 132.0 (s), 131.6 (s), 131.0 (s), 128.4 (s), 125.9 (s), 125.4 (s), 120.6 (s), 115.4 (s). mp = 108.9 - 110.3 °C.

(1H-Benzo[d]imidazol-1-yl)(4-methoxyphenyl)methanone (13, CAS: 13361-55-2)³

77% yield (48 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.27 (s, 1H), 8.13 (dd, *J* = 6.5, 2.6 Hz, 1H), 7.86 - 7.78 (m, 3H), 7.46 - 7.40 (m, 2H), 7.06 (d, *J* = 8.8 Hz, 2H), 3.92 (s, 3H).; ¹³C NMR (126 MHz, CDCl₃) δ 166.4 (s), 163.7 (s), 144.0 (s), 143.1 (s), 132.3 (s), 132.1 (s), 125.5 (s), 125.0 (s), 124.7 (s), 120.5 (s), 115.3 (s), 114.4 (s), 55.6 (s). mp = 111.9 - 113.7 °C.

(1H-Benzo[d]imidazol-1-yl)(4-vinylphenyl)methanone (14, CAS: 300396-82-1)

89% yield (54 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.24 (s, 1H), 8.21 - 8.13 (m, 1H), 7.85 - 7.80 (m, 1H), 7.81 - 7.74 (m, 2H), 7.60 (d, *J* = 8.3 Hz, 2H), 7.47 - 7.40 (m, 2H), 6.80 (dd, *J* = 17.6, 10.9 Hz, 1H), 5.94 (d, *J* = 17.6 Hz, 1H), 5.47 (d, *J* = 10.9 Hz, 1H).; ¹³C NMR (126 MHz, CDCl₃) δ 166.7 (s), 144.0 (s), 143.0 (s), 142.4 (s), 135.4 (s), 132.1 (s), 131.7 (s), 130.1 (s), 126.6 (s), 125.7 (s), 125.2 (s), 120.5 (s), 117.6 (s), 115.4 (s). mp = 95.8 - 96.6 °C.

(1H-Benzo[d]imidazol-1-yl)(4-cyclohexylphenyl)methanone (15, new compound)

78% yield (60 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.26 (s, 1H), 8.20 (dd, *J* = 6.7, 2.0 Hz, 1H), 7.83 (dd, *J* = 6.7, 2.1 Hz, 1H), 7.74 (d, *J* = 8.2 Hz, 2H), 7.47 - 7.39 (m, 4H), 2.67 - 2.58 (m, 1H), 1.94 - 1.85 (m, 4H), 1.81 - 1.76 (m, 1H), 1.51 - 1.38 (m, 4H), 1.33 - 1.25 (m, 1H).; ¹³C NMR (126 MHz, CDCl₃) δ 167.0 (s), 154.1 (s), 144.0 (s), 143.2 (s), 132.2 (s), 130.2 (s), 129.9 (s), 127.5 (s), 125.6 (s), 125.1 (s), 120.4 (s), 115.4 (s), 44.7 (s), 34.1 (s), 26.6 (s), 25.9 (s). new compound, HRMS *m/z* (EI) calcd. for C₂₀H₂₀N₂O M+ 304.1576, found 304.1570. mp = 94.0 - 96.1 °C.

(1H-Benzo[d]imidazol-1-yl)(o-tolyl)methanone (16, CAS: 200626-52-4)

76% yield (45 mg) as a yellow oil. ¹H NMR (500 MHz, CDCl₃) δ 8.18 - 8.14 (m, 1H), 7.97 (s, 1H), 7.84 - 7.81 (m, 1H), 7.51 (td, *J* = 7.6, 1.3 Hz, 1H), 7.46 - 7.42 (m, 3H), 7.37 (dd, *J* = 8.9, 8.4 Hz, 2H), 2.40 (s, 3H).; ¹³C NMR (126 MHz, CDCl₃) δ 167.6 (s), 144.3 (s), 143.0 (s), 136.7 (s), 133.0 (s), 131.7(s), 131.6(s), 131.4 (s), 127.9 (s), 126.0 (s), 125.8 (s), 125.3 (s), 120.6 (s), 115.4 (s), 19.4 (s).

(1H-Benzo[d]imidazol-1-yl)(3,4-dimethylphenyl)methanone (17, CAS: 333348-55-3)

72% yield (45 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.25 (s, 1H), 8.20 - 8.15 (m, 1H), 7.86 - 7.79 (m, 1H), 7.59 (s, 1H), 7.52 (dd, *J* = 7.8, 1.7 Hz, 1H), 7.46 - 7.39 (m, 2H), 7.32 (d, *J* = 7.8 Hz, 1H), 2.38 (s, 3H), 2.35 (s, 3H).; ¹³C NMR (126 MHz, CDCl₃) δ 167.2 (s), 144.0 (s), 143.2 (s), 143.0 (s), 137.7 (s), 132.2 (s), 130.7 (s), 130.3 (s), 130.1 (s), 127.3 (s), 125.5 (s), 125.0 (s), 120.4 (s), 115.4 (s), 20.0 (s), 19.7 (s). mp = 122.1 - 123.3 °C.

(1H-Benzo[d]imidazol-1-yl)(3,4-dichlorophenyl)methanone (18, CAS: 330215-63-9)

28% yield (21 mg) as a white solid.. ¹H NMR (500 MHz, CDCl₃) δ 8.19 (s, 1H), 8.18 - 8.14 (m, 1H), 7.93 (d, *J* = 2.0 Hz, 1H), 7.87 - 7.83 (m, 1H), 7.69 (d, *J* = 8.3 Hz, 1H), 7.64 (dd, *J* = 8.3, 2.0 Hz, 1H), 7.49 - 7.44 (m, 2H).; ¹³C NMR (126 MHz, CDCl₃) δ 164.7 (s), 144.0 (s), 142.4 (s), 138.2 (s), 134.0 (s), 132.4 (s), 131.9 (s), 131.4 (s), 131.2 (s), 128.4 (s), 126.1 (s), 125.7 (s), 120.8 (s), 115.4 (s). mp = 131.8 - 133.7 °C.

(1H-Benzo[d]imidazol-1-yl)(3-chlorophenyl)methanone (19, CAS :200626-53-5)

51% yield (26 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.23 (s, 1H), 8.21 - 8.18 (m, 1H), 7.86 - 7.83 (m, 1H), 7.82 - 7.79 (m, 2H), 7.72 - 7.68 (m, 1H), 7.61 - 7.57 (m, 2H), 7.48 - 7.41 (m, 2H).; ¹³C NMR (126 MHz, CDCl₃) δ 167.1 (s), 144.0 (s), 143.1 (s), 133.2 (s), 132.8 (s), 132.1

(s), 129.5 (s), 129.0 (s), 125.8 (s), 125.3 (s), 120.5 (s), 115.5 (s). mp = 102.3 - 104.1 °C.

(1H-Benzo[d]imidazol-1-yl)(naphthalen-2-yl)methanone (20, new compound)

59% yield (40 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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); ¹³C NMR: (125 MHz, CDCl₃, ppm). δ 167.2 (s), 144.0 (s), 143.2 (s), 135.3 (s), 132.2 (s), 131.1 (s), 129.9 (s), 129.2 (s), 129.0 (s), 128.0 (s), 127.6 (s), 125.8 (s), 125.3 (s), 125.0 (s), 120.6 (s), 115.4 (s). new compound, HRMS m/z (EI) calcd. for C₁₈H₁₂N₂O M+ 272.0950, found 272.0953. mp = 105.1 - 106.7 °C.

(1H-Benzo[d]imidazol-1-yl)(pyren-4-yl)methanone (21, new compound)

88% yield (76 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.32 - 8.20 (m, 6H), 8.17 - 8.06 (m, 4H), 8.01 (s, 1H), 7.91 - 7.86 (m, 1H), 7.51 - 7.46 (m, 2H); ¹³C NMR (126 MHz, CDCl₃) δ 167.6 (s), 144.3 (s), 143.5 (s), 133.8 (s), 132.0 (s), 131.0 (s), 130.4 (s), 130.0 (s), 129.9 (s), 129.6 (s), 126.9 (s), 126.8 (s), 126.7 (s), 126.5 (s), 126.5 (s), 126.0 (s), 125.9 (s), 125.4 (s), 124.6 (s), 124.1 (s), 124.0 (s), 123.2 (s), 120.6 (s), 115.6 (s). new compound, HRMS m/z (EI) calcd. for C₂₄H₁₄N₂O M+ 346.1106, found 346.1103. mp = 132.5 - 134.3 °C.

(1H-Benzo[d]imidazol-1-yl)(thiophen-3-yl)methanone (22, new compound)

40% yield (24 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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); ¹³C NMR: (125 MHz, CDCl₃, ppm) δ 161.5 (s), 144.0 (s), 142.5 (s), 134.7 (s), 133.2 (s), 132.1 (s), 128.2 (s), 127.6 (s), 125.7 (s), 125.3 (s), 120.6 (s), 115.4 (s). new compound), HRMS m/z (EI) calcd. for C₁₂H₈N₂OS M+ 228.0357, found 228.0361. mp = 124.8 - 126.3 °C.

(1H-Benzo[d]imidazol-1-yl)(furan-3-yl)methanone (23, new compound)

61% yield (33 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.48 (s, 1H), 8.26 - 8.22 (m, 1H), 8.15 - 8.10 (m, 1H), 7.84 (dd, *J* = 6.8, 1.8 Hz, 1H), 7.62 (t, *J* = 1.7 Hz, 1H), 7.48 - 7.42 (m, 2H), 6.91 (dd, *J* = 1.9, 0.8 Hz, 1H); ¹³C NMR (126 MHz, CDCl₃) δ 160.4 (s), 147.7 (s), 144.6 (s), 143.8 (s), 141.9 (s), 131.9 (s), 125.9 (s), 125.3 (s), 120.8 (s), 120.5 (s), 115.4 (s), 110.4 (s). new compound, HRMS m/z (EI) calcd. for C₁₂H₈N₂O₂ M+ 212.0586, found 212.0589. mp = 128.3 - 130.1 °C.

(1H-Benzo[d]imidazol-1-yl)(cyclohexyl)methanone (24, CAS: 294649-09-5)

51% yield (30 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.44 (s, 1H), 8.28 - 8.22 (m, 1H), 7.80 (dd, *J* = 7.2, 1.3 Hz, 1H), 7.45 - 7.35 (m, 2H), 3.06 (tt, *J* = 11.5, 3.4 Hz, 1H), 2.04 (dd, *J* = 14.1, 1.6 Hz, 2H), 1.96 - 1.87 (m, 2H), 1.81 - 1.70 (m, 3H), 1.48 - 1.39 (m, 2H), 1.37 - 1.30 (m, 1H); ¹³C NMR (126 MHz, CDCl₃) δ 173.8 (s), 143.7 (s), 140.8 (s), 131.6 (s), 125.9 (s), 125.0 (s), 120.4 (s), 115.8 (s), 44.1 (s), 29.4 (s), 25.5 (s), 25.4 (s). mp = 57.8 - 59.1 °C.

1-(1H-Benzo[d]imidazol-1-yl)hexan-1-one (25, CAS: 901547-84-0)

47% yield (26 mg) as a yellow oil. ¹H NMR (500 MHz, CDCl₃) δ 8.41 (s, 1H), 8.29 - 8.21 (m, 1H), 7.80 (dd, *J* = 7.1, 1.4 Hz, 1H), 7.47 - 7.35 (m, 2H), 3.00 (t, *J* = 7.4 Hz, 2H), 1.91 - 1.84 (m, 2H), 1.48 - 1.36 (m, 4H), 0.94 (t, *J* = 7.1 Hz, 3H); ¹³C NMR (126 MHz, CDCl₃) δ 170.4 (s), 143.8 (s), 140.9 (s), 131.5 (s), 125.9 (s), 125.0 (s), 120.5 (s), 115.6 (s), 35.9 (s), 31.2 (s), 24.0 (s), 22.4 (s), 13.9 (s).

1-(1H-Benzo[d]imidazol-1-yl)ethan-1-one (26, CAS: 18773-95-0)⁸

46% yield (19 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 8.36 (s, 1H), 8.27 - 8.16 (m, 1H), 7.80 (dd, *J* = 7.0, 1.6 Hz, 1H), 7.45 - 7.38 (m, 2H), 2.74 (s, 3H); ¹³C NMR (126 MHz,

CDCl₃) δ 167.2 (s), 143.9 (s), 141.3 (s), 131.4 (s), 125.9 (s), 125.1 (s), 120.5 (s), 115.5 (s), 23.8 (s). mp = 92.0 - 93.4 °C.

(5,6-Dimethyl-1H-benzo[d]imidazol-1-yl)(phenyl)methanone (27, CAS: 16109-46-9)⁷

89% yield (57 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, 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).; ¹³C NMR: (125 MHz, CDCl₃, ppm). δ 167.1 (s), 142.5 (s), 142.4 (s), 135.1 (s), 134.3 (s), 133.9 (s), 133.0 (s), 130.5 (s), 129.5 (s), 129.0 (s), 120.5 (s), 115.7 (s), 20.5 (s), 20.3 (s). mp = 91.8 - 93.7 °C.

Typical procedure and characterization data for amine exchange reaction

A sealed pressure vessel was charged with (1H-benzo[d]imidazol-1-yl)(phenyl)methanone (**3**) (55.0 mg, 0.25 mmol), pyridin-2-amine (**27**, 35.5 mg, 0.375 mmol), and *p*-xylene (1 mL). The resulting solution was stirred at 130 °C under air overnight. 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 47 mg of *N*-(pyridin-2-yl)benzamide (**29**) in 92% isolated yield as a yellow solid.

***N*-(Pyridin-2-yl)benzamide (29, CAS : 4589-12-2)⁹**

92% yield (47 mg) as a yellow solid. ¹H NMR (500 MHz, CDCl₃) δ 8.84 (s, 1H), 8.40 (d, *J* = 8.4 Hz, 1H), 8.23 (d, *J* = 4.1 Hz, 1H), 7.98 - 7.88 (m, 2H), 7.81 - 7.69 (m, 1H), 7.57 (t, *J* = 7.4 Hz, 1H), 7.49 (t, *J* = 7.6 Hz, 2H), 7.11 - 6.99 (m, 1H).; ¹³C NMR (126 MHz, CDCl₃) δ 165.8 (s), 151.6 (s), 147.9 (s), 138.5 (s), 134.3 (s), 132.2 (s), 128.8 (s), 127.2 (s), 119.9 (s), 114.2 (s). mp = 78.2 - 81.1 °C.

***N*-(Pyridin-2-yl)-2-naphthamide (31, CAS : 159257-88-2)⁹**

85% yield (53 mg) as a white solid. ¹H NMR (500 MHz, CDCl₃) δ 9.15 (s, 1H), 8.49 - 8.40 (m, 2H), 8.23 (ddd, *J* = 4.9, 1.7, 0.7 Hz, 1H), 7.99 (dd, *J* = 8.5, 1.8 Hz, 1H), 7.94 - 7.87 (m, 3H), 7.76 (ddd, *J* = 8.4, 7.4, 1.9 Hz, 1H), 7.57 (dtd, *J* = 14.7, 6.9, 1.3 Hz, 2H), 7.04 (ddd, *J* = 7.3, 4.9, 0.9 Hz, 1H).; ¹³C NMR (126 MHz, CDCl₃) δ 165.9 (s), 151.7 (s), 147.8 (s), 138.5 (s), 135.0 (s), 132.5 (s), 131.4 (s), 129.0 (s), 128.7 (s), 128.0 (d, *J* = 9.8 Hz), 127.7 (s), 126.9 (s), 123.6 (s), 119.9 (s), 114.3 (s). mp = 162.2 - 163.1 °C.

(1H-Indazol-1-yl)(phenyl)methanone (32, CAS: 23301-00-0)¹

95% yield (52 mg) as a white solid. ¹H NMR: (500 MHz, CDCl₃, ppm) δ 8.58 (dd, *J* = 8.5 Hz, *J* = 0.7 Hz, 1 H), 8.21 (d, *J* = 0.6 Hz, 1 H), 8.09-8.07 (m, 2 H), 7.79 -7.77(m, 1 H), 7.64– 7.59 (m, 2 H), 7.55- 7.51(m, 2H), 7.43- 7.40(m, 1H); ¹³C NMR: (125 MHz, CDCl₃, ppm.) δ 168.3 (s), 140.2 (s), 140.1 (s), 133.2 (s), 132.2 (s), 130.9 (s), 129.4 (s), 127.9 (s), 126.1 (s), 124.8 (s), 120.9 (s), 115.8 (s). mp = 83.8–84.2 °C.

(1H-Benzo[d][1,2,3]triazol-1-yl)(phenyl)methanone (33, CAS: 4231-62-3)¹

56% yield (43 mg) as a white solid. ¹H NMR (500 MHz, DMSO-d₆) δ 8.27 (d, *J* = 8.3 Hz, 1H), 8.23 (d, *J* = 8.3 Hz, 1H), 8.06 (dd, *J* = 8.3, 1.2 Hz, 2H), 7.80 (ddd, *J* = 8.2, 7.2, 0.9 Hz, 1H), 7.78 – 7.70 (m, 1H), 7.67 – 7.51 (m, 3H); ¹³C NMR (126 MHz, DMSO-d₆) δ 167.5 (s), 146.0 (s), 134.6 (s), 132.6 (s), 132.2 (d, *J* = 11.0 Hz), 131.7 (s), 129.3 (s), 127.7 (s), 120.9 (s), 115.3 (s). mp 111–112 °C.

***N*-Phenylbenzamide (34, CAS: 93-98-1)¹⁰**

68% yield (34 mg) as a white solid. ¹H NMR (500 MHz, DMSO-d₆) δ 10.26 (s, 1H), 7.99 – 7.84 (m, 2H), 7.73 (d, *J* = 7.6 Hz, 2H), 7.57 (ddd, *J* = 6.4, 3.7, 1.2 Hz, 1H), 7.50 (dd, *J* = 10.3, 4.6 Hz,

2H), 7.34 (t, $J = 7.9$ Hz, 2H), 7.10 (t, $J = 7.4$ Hz, 1H); ^{13}C NMR (126 MHz, DMSO- d_6) δ 166.8 (s), 139.8 (s), 135.7 (s), 132.6 (s), 129.6 (s), 129.4 (s), 128.5 (s), 124.8 (s), 121.5 (s). mp 161–163 °C.

***N*-Benzyl-*N*-methylbenzamide (35, CAS: 61802-83-3)¹⁰**

78% yield (57 mg) as a yellow oil. ^1H NMR (500 MHz, CDCl_3) δ 7.47–7.29 (m, 9H), 7.17 (s, 1H), 4.77 (s, 1H), 4.51 (s, 1H), 3.03 (s, 1.5H), 2.86 (s, 1.5H); ^{13}C NMR (126 MHz, CDCl_3) δ 172.3 (s), 171.6 (s), 137.0 (s), 136.5 (s), 136.1 (s), 129.6 (s), 128.8 (s), 128.7 (s), 128.3 (s), 128.1 (s), 127.5 (s), 126.9 (s), 126.7 (s), 55.1 (s), 50.7 (s), 36.9 (s), 33.1 (s).

Phenyl(3-phenyl-1*H*-pyrazol-1-yl)methanone (36, CAS: 126382-89-6)¹

71% yield (44 mg) as a white solid. ^1H NMR (500 MHz, CDCl_3) δ 8.48 (d, $J = 2.9$ Hz, 1H), 8.31 – 8.23 (m, 2H), 7.91 – 7.87 (m, 2H), 7.68 – 7.60 (m, 1H), 7.54 (dd, $J = 10.7, 4.8$ Hz, 2H), 7.46 – 7.39 (m, 3H), 6.87 (d, $J = 2.9$ Hz, 1H); ^{13}C NMR: (125 MHz, CDCl_3 , ppm.) δ 166.11 (s), 155.89 (s), 132.98 (s), 131.86 (s), 131.79 (s), 131.73 (s), 131.48 (s), 129.18 (s), 128.74 (s), 128.02 (s), 126.37 (s), 107.18 (s). mp = 63.7–64.9 °C.

***N*-(Pyridin-2-yl)cyclohexanecarboxamide (37, CAS: 68134-77-0)¹¹**

86% yield (44 mg) as a white solid. ^1H NMR (500 MHz, CDCl_3) δ 8.71 (s, 1H), 8.29 – 8.15 (m, 2H), 7.73 – 7.60 (m, 1H), 7.06 – 6.90 (m, 1H), 2.24 (tt, $J = 11.7, 3.5$ Hz, 1H), 1.95 – 1.88 (m, 2H), 1.82 – 1.76 (m, 2H), 1.66 (dd, $J = 9.0, 3.3$ Hz, 1H), 1.52 (qd, $J = 12.3, 3.2$ Hz, 2H), 1.28 – 1.20 (m, 3H); ^{13}C NMR (126 MHz, CDCl_3) δ 175.0 (s), 151.8 (s), 147.4 (s), 138.4 (s), 119.5 (s), 114.3 (s), 46.3 (s), 29.4 (s), 25.6 (s), 25.5 (s). mp = 88.1–89.3 °C.

Phenyl(piperidin-1-yl)methanone (38, CAS: 776-75-0)¹⁰

74% yield (35 mg) as a yellow oil. ^1H NMR (500 MHz, CDCl_3) δ 7.42 – 7.32 (m, 5H), 3.70 – 3.33 (m, 4H), 1.66 – 1.50 (m, 6H); ^{13}C NMR (126 MHz, CDCl_3) δ 170.3 (s), 136.5 (s), 129.3 (s), 128.3 (s), 126.7 (s), 48.7 (s), 43.1 (s), 26.5 (s), 25.6 (s), 24.5 (s).

***N,N*-Diethylbenzamide (39, CAS: 1696-17-9)¹²**

68% yield (30 mg) as a yellow oil. ^1H NMR (500 MHz, CDCl_3) δ 7.41 – 7.31 (m, 5H), 3.54 – 3.24 (m, 4H), 1.27 – 1.05 (m, 6H); ^{13}C NMR (126 MHz, CDCl_3) δ 171.2 (s), 137.2 (s), 129.0 (s), 128.3 (s), 126.2 (s), 43.2 (s), 39.2 (s), 14.1 (s), 12.9 (s).

Methyl benzoyl-L-leucinate (40, CAS: 3005-60-5)¹³

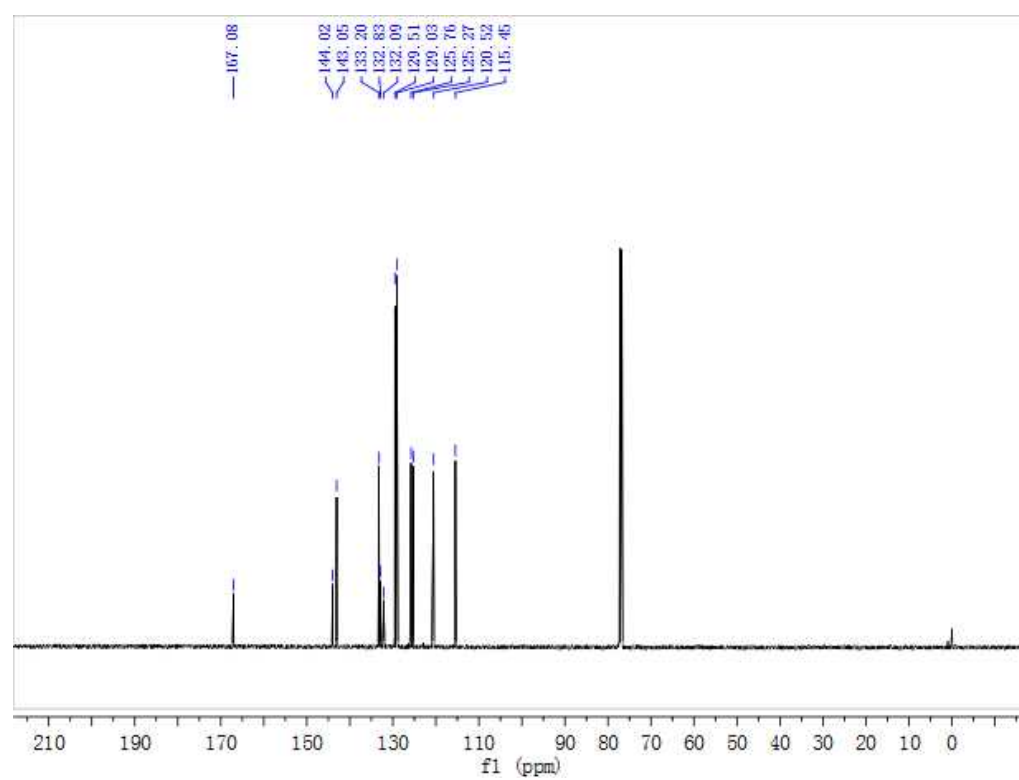
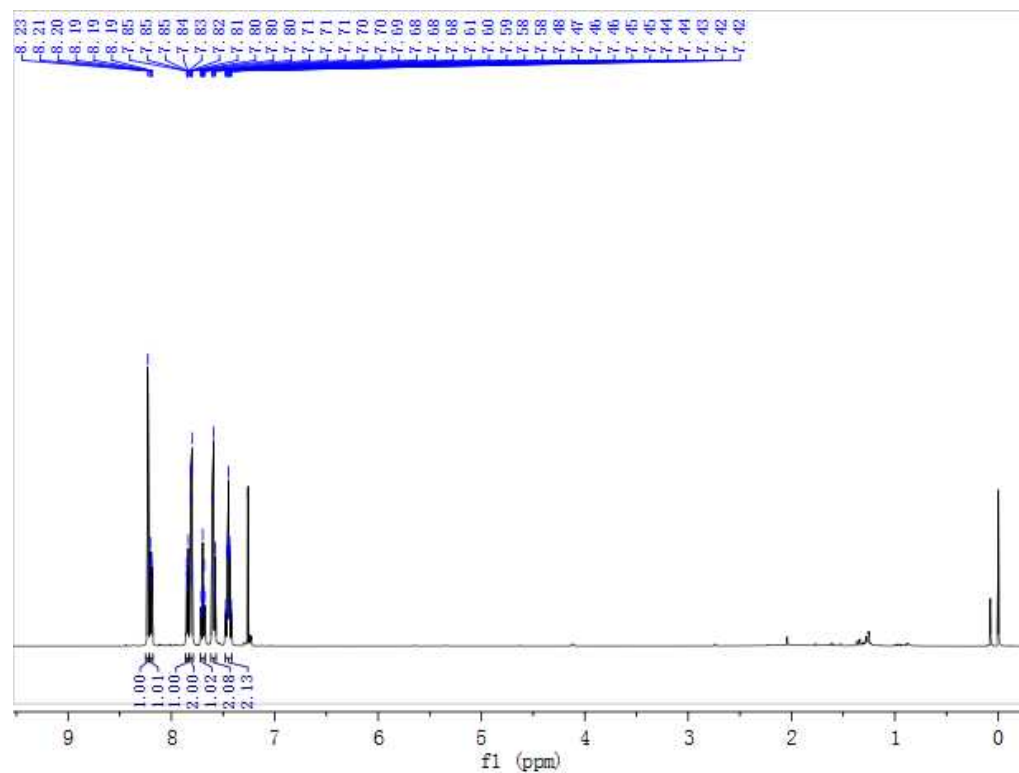
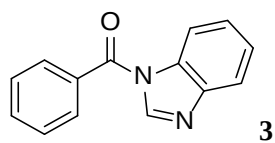
A sealed pressure vessel was charged with (1*H*-benzo[*d*]imidazol-1-yl)(phenyl)methanone (3) (55.0 mg, 0.25 mmol), L-leucine methyl ester hydrochloride (68 mg, 0.375 mmol), Et_3N (38 mg, 0.375 mmol), and *p*-xylene (1 mL). The mixture was stirred at room temperature for 3 minutes. Then the resulting solution was heated to 130 °C under air overnight. 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 1:6) to give 45 mg of methyl benzoyl-L-leucinate (40) in 71% isolated yield as a white solid. ^1H NMR (500 MHz, CDCl_3) δ 7.85 – 7.76 (m, 2H), 7.56 – 7.39 (m, 3H), 6.64 (d, $J = 8.0$ Hz, 1H), 4.93 – 4.83 (m, 1H), 3.78 (s, 3H), 1.80 – 1.65 (m, 3H), 1.00 (d, $J = 9.6, 3\text{H}$), 0.99 (d, $J = 9.6, 3\text{H}$); ^{13}C NMR (126 MHz, CDCl_3) δ 173.7 (s), 167.1 (s), 133.9 (s), 131.7 (s), 128.5 (s), 127.0 (s), 52.3 (s), 51.1 (s), 41.8 (s), 25.0 (s), 22.8 (s), 22.0 (s).

References

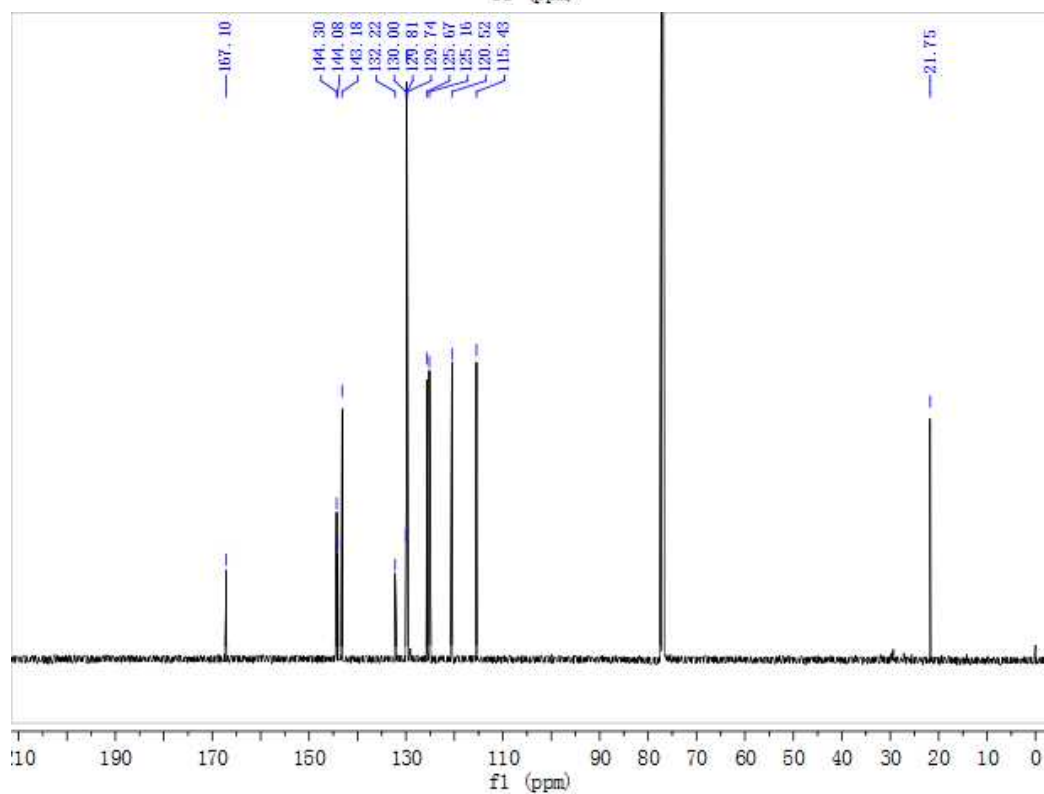
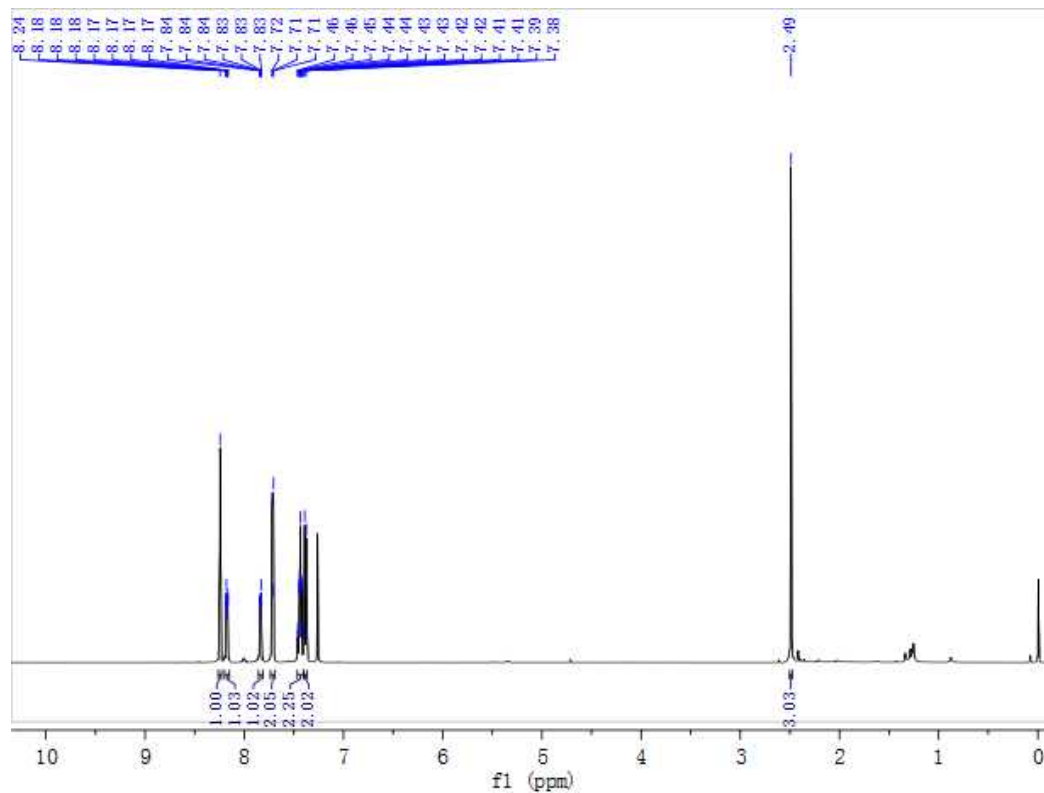
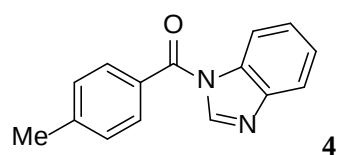
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NMR Spectra

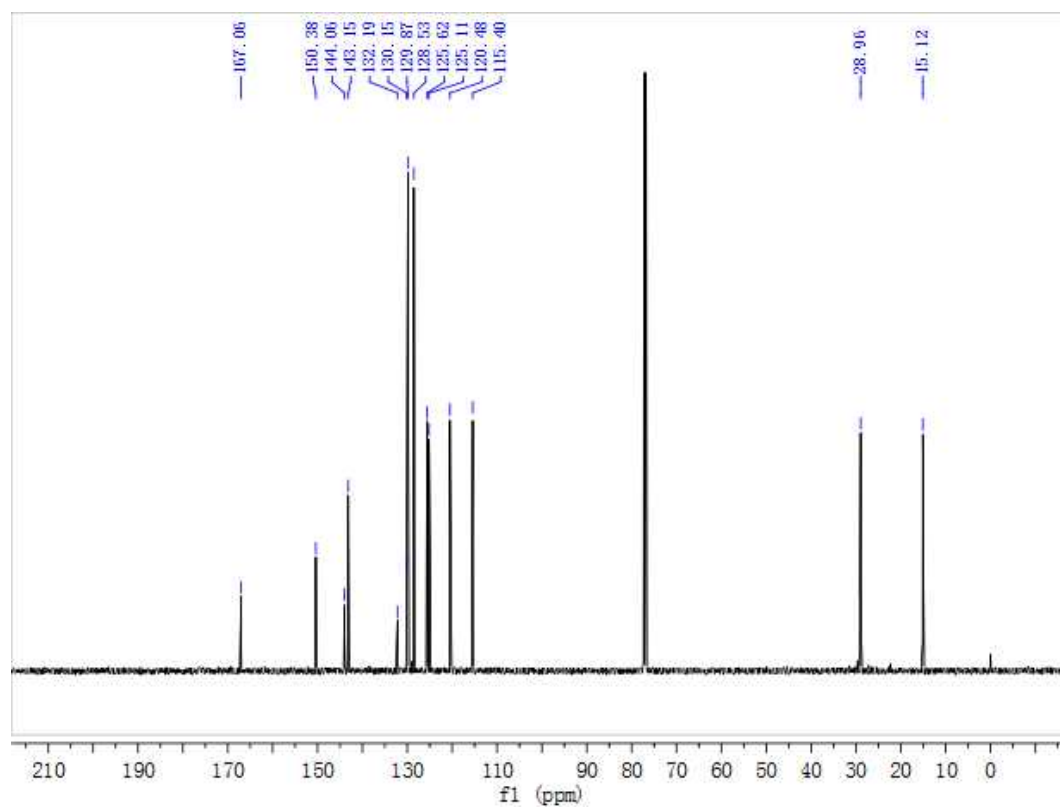
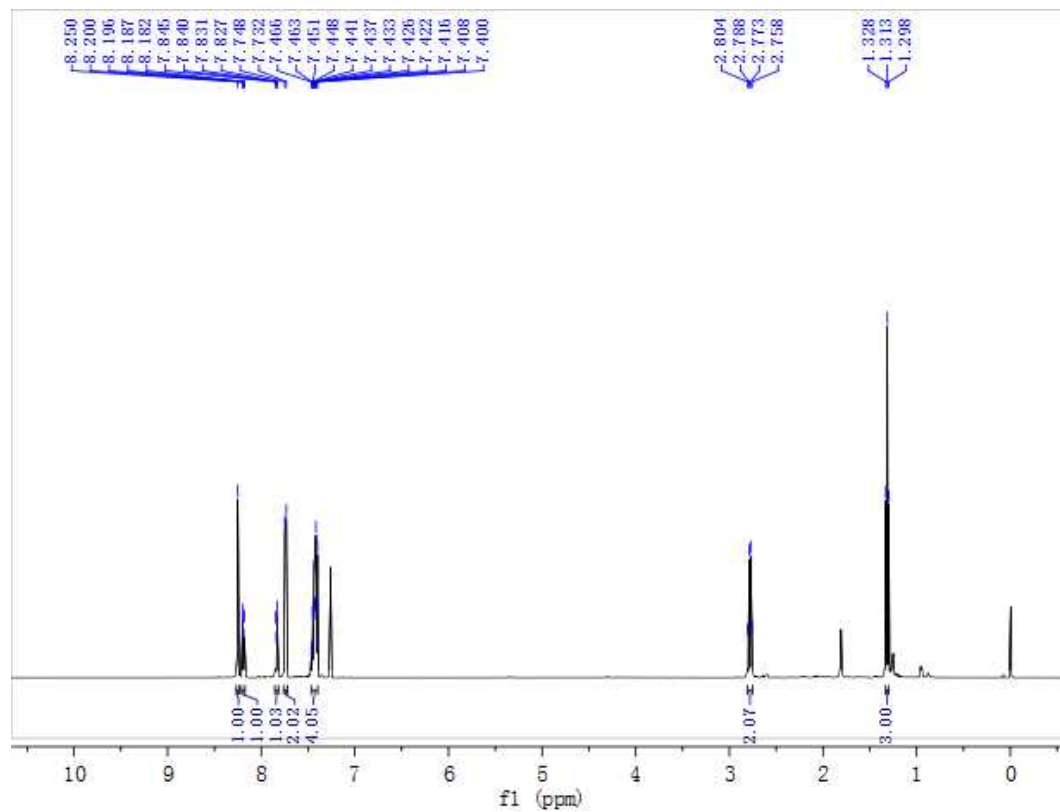
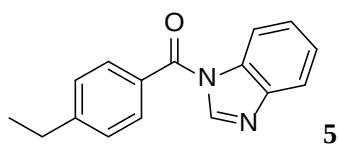
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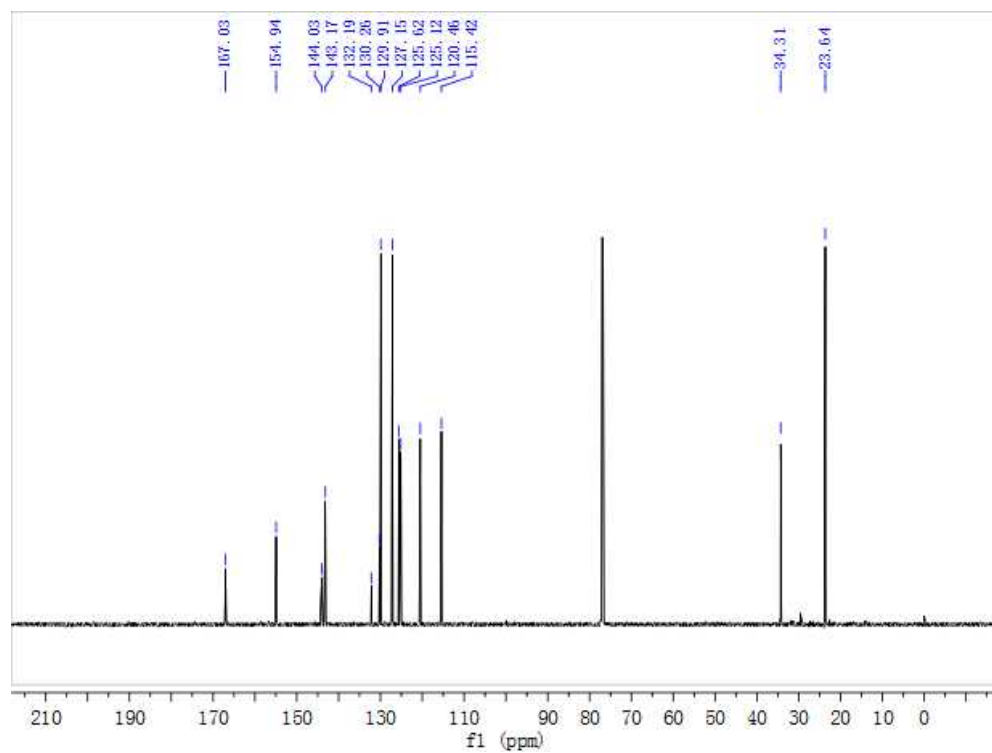
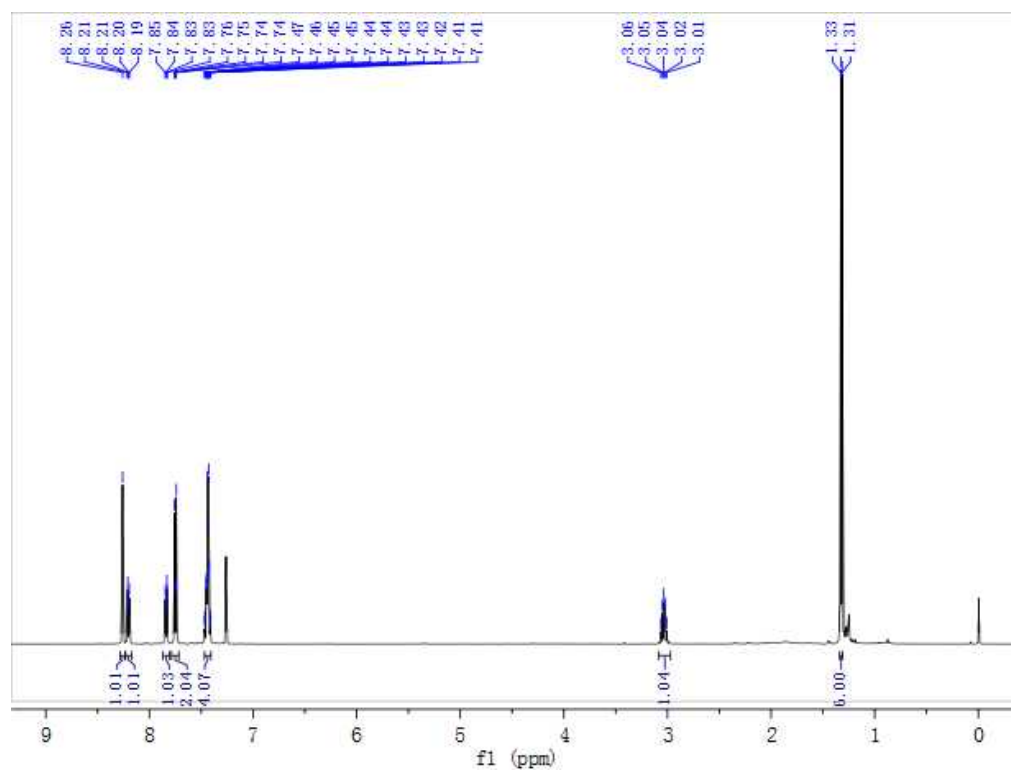
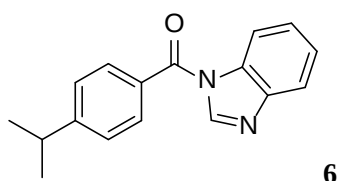
(1*H*-Benzo[d]imidazol-1-yl)(*p*-tolyl)methanone (CAS: 28997-00-4)



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

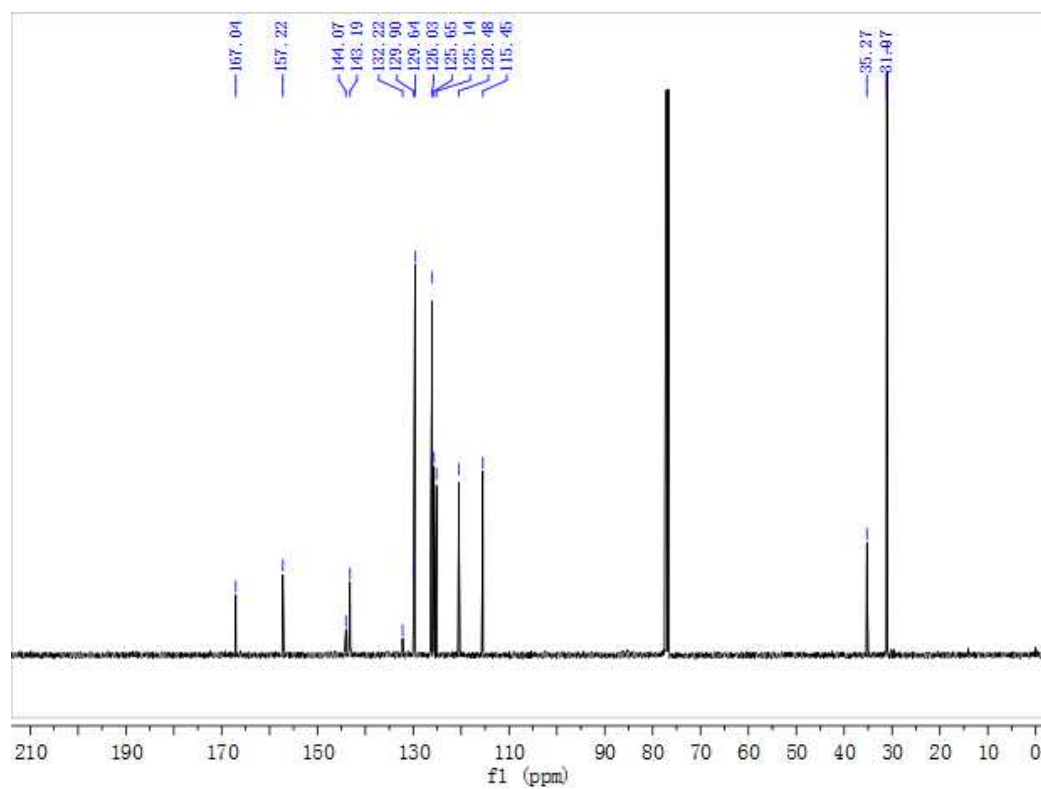
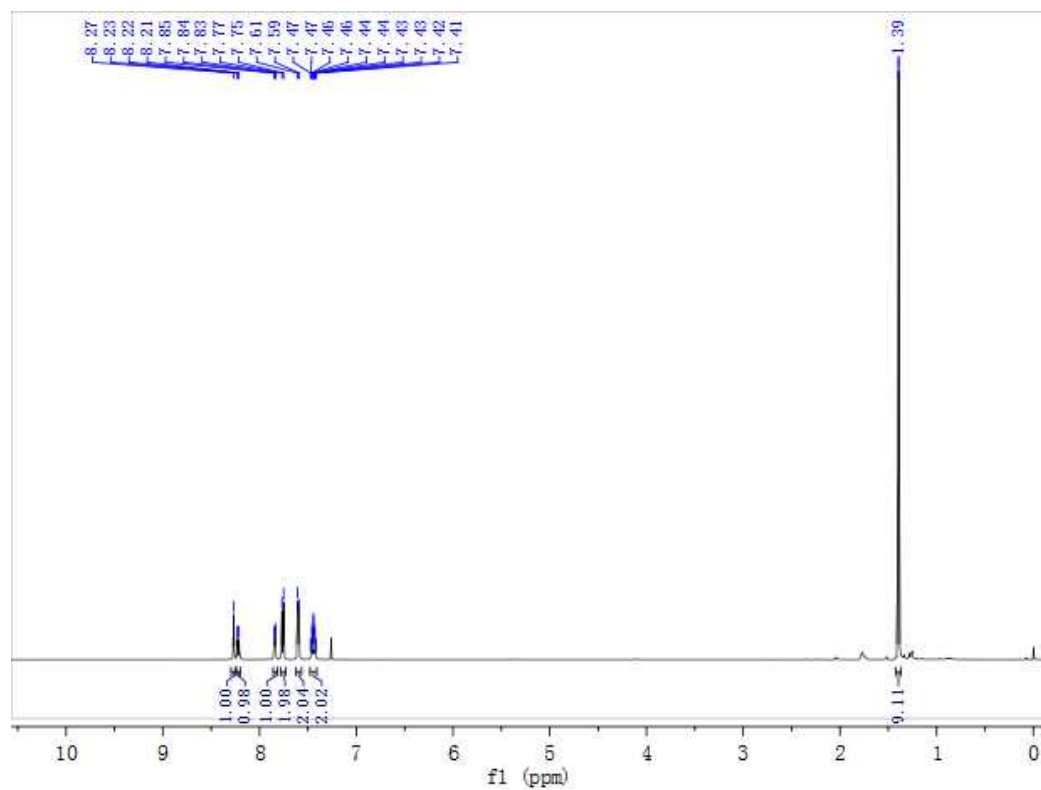
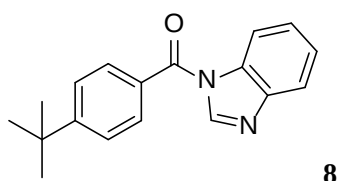


(1*H*-Benzo[*d*]imidazol-1-yl)(4-isopropylphenyl)methanone (CAS: 901440-49-1)



CCCC1=CC=C(C=C1)C(=O)N2C=NC3C=CC=CC=C23

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



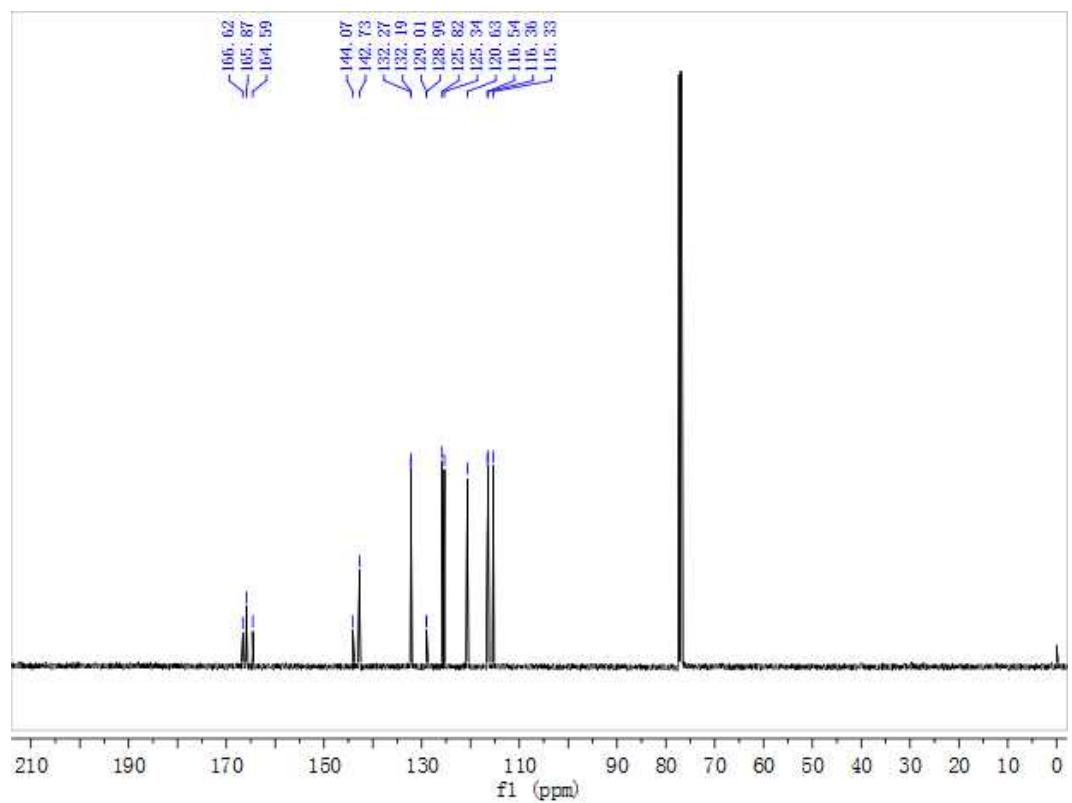
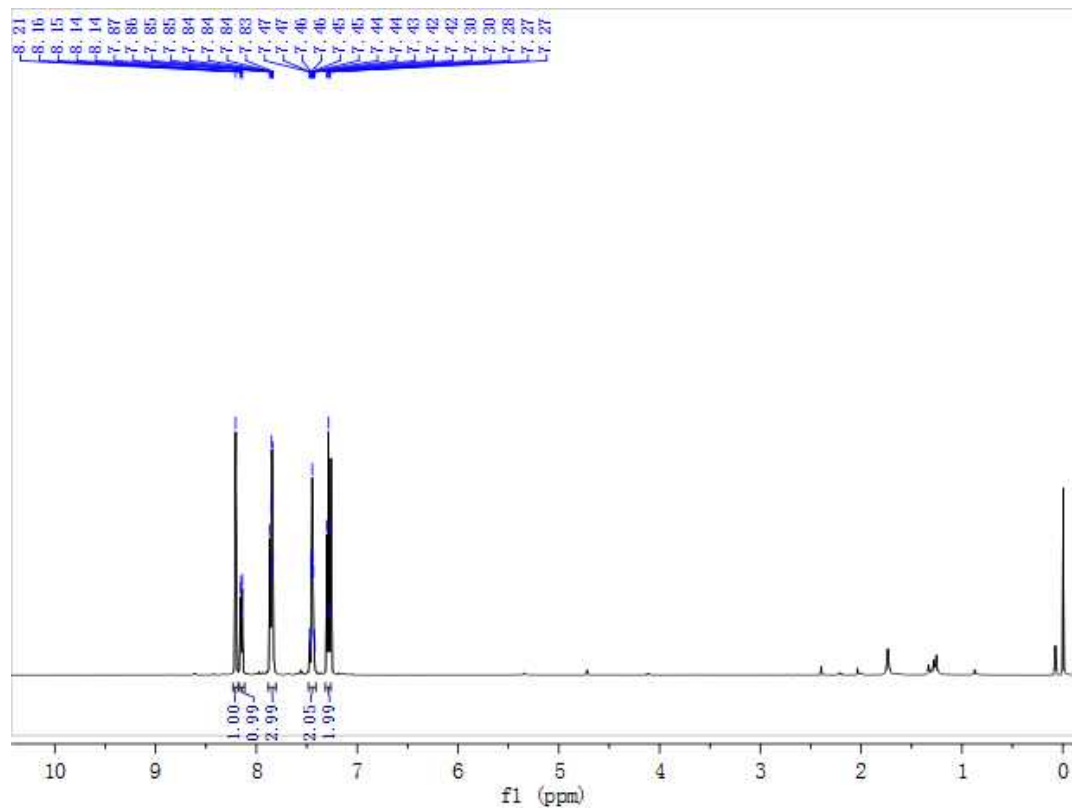
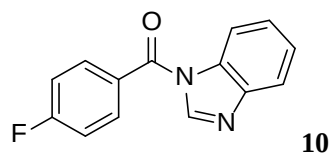
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¹H NMR spectrum of compound 10 in CDCl₃. The spectrum shows peaks in the aromatic region (7.38-8.27 ppm) and aliphatic region (0.95-2.75 ppm). Integration values are provided below the peaks.

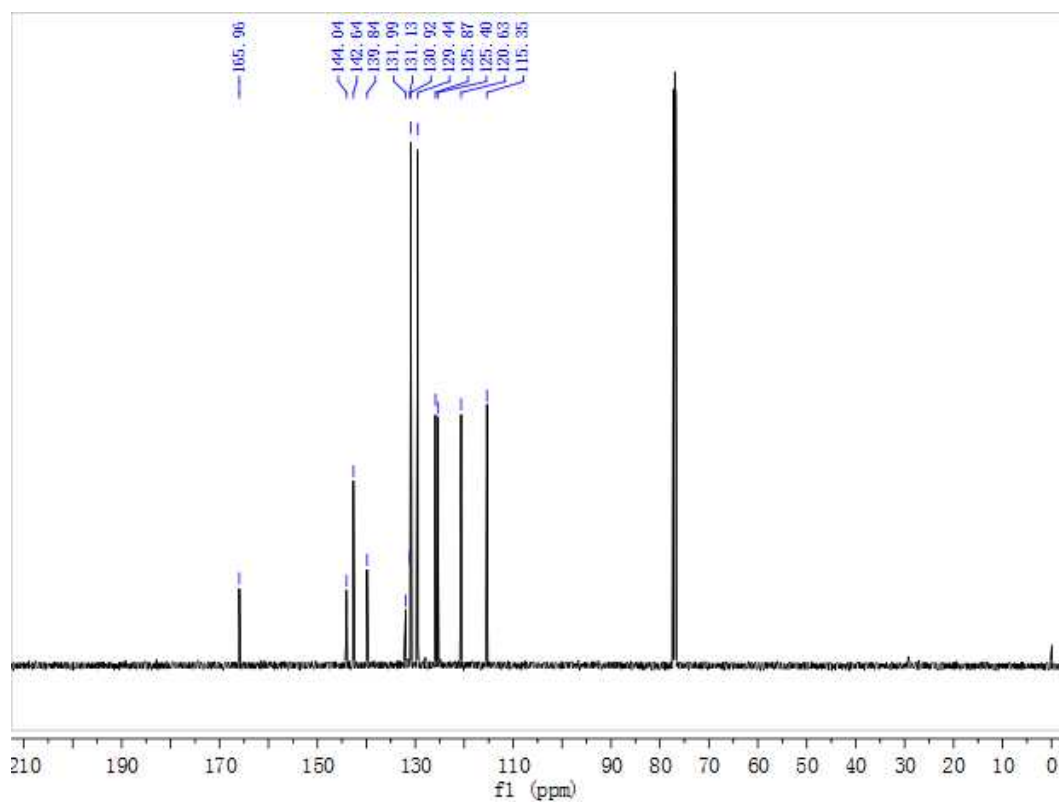
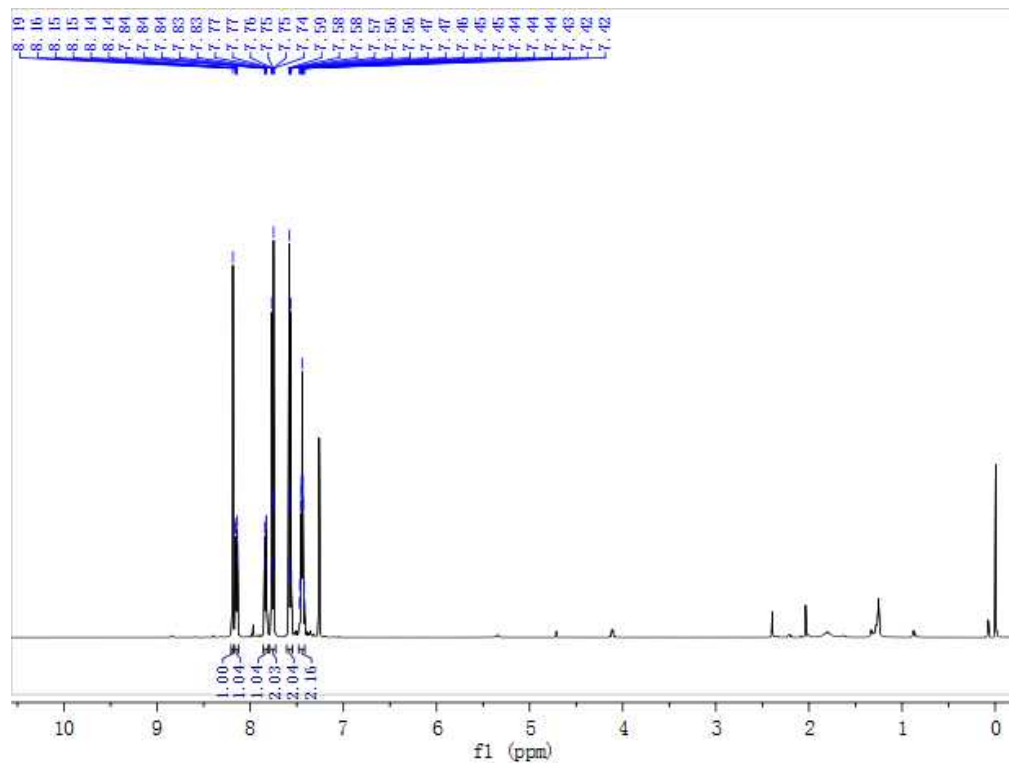
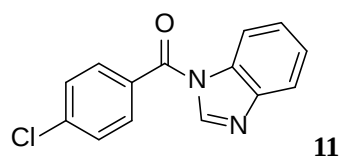
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8.10	
8.08	
8.06	
8.04	
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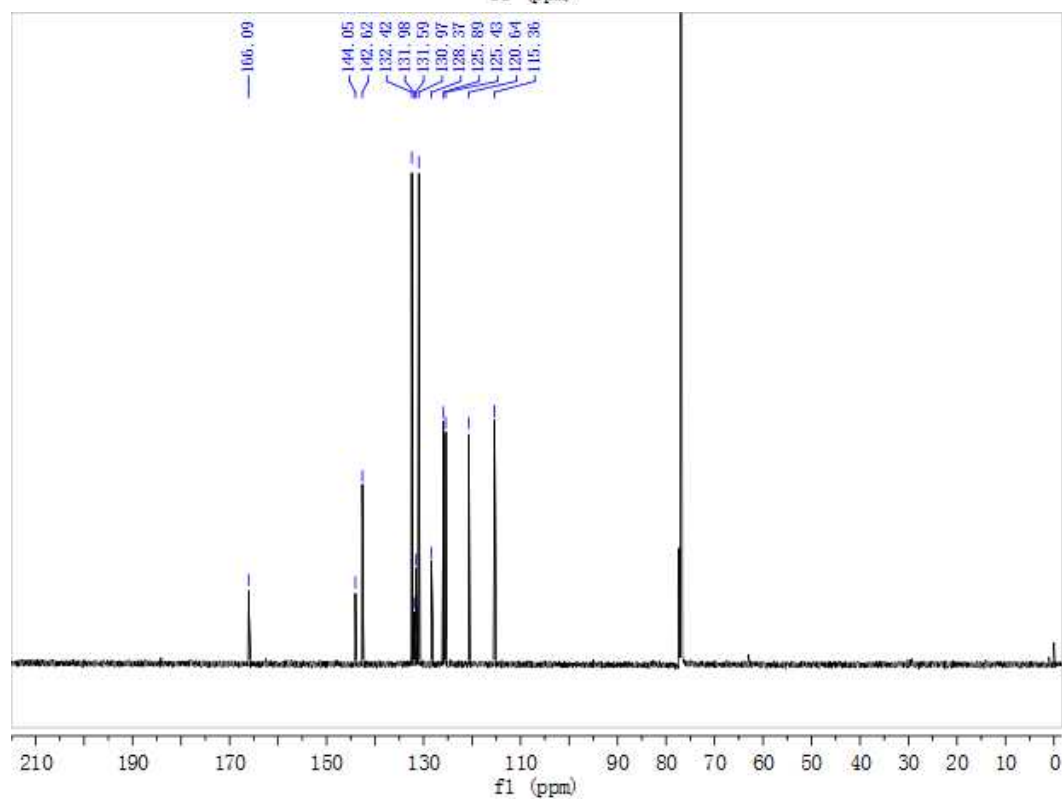
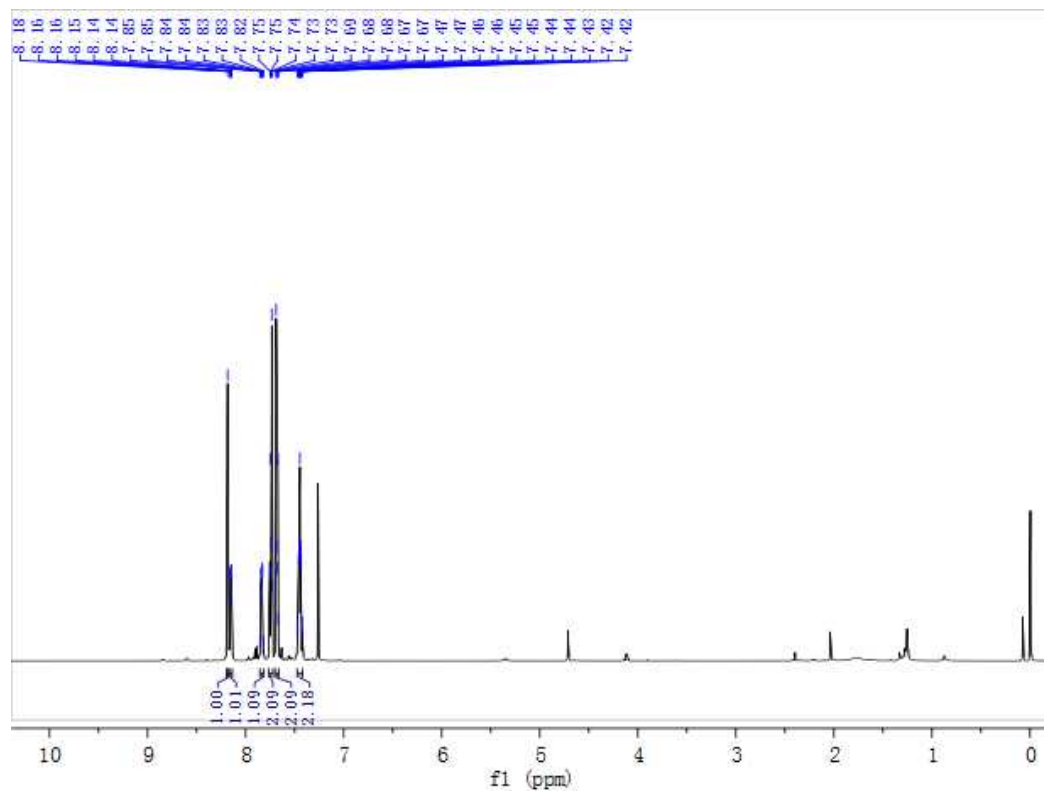
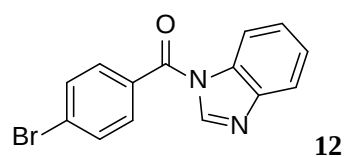
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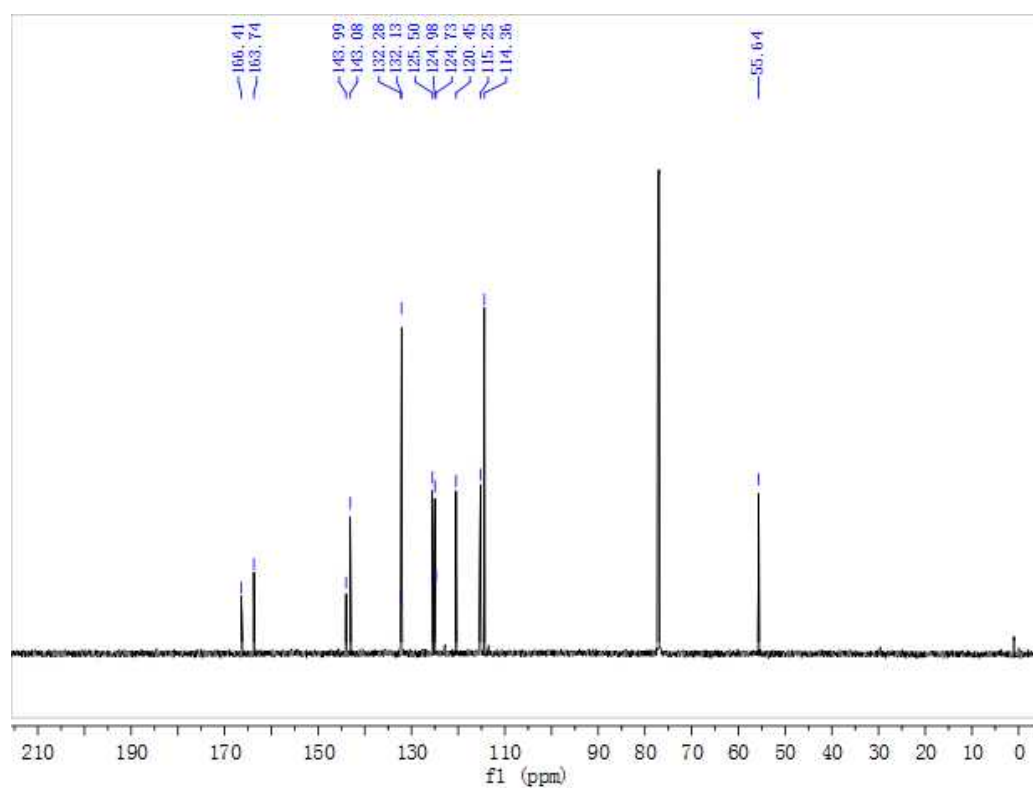
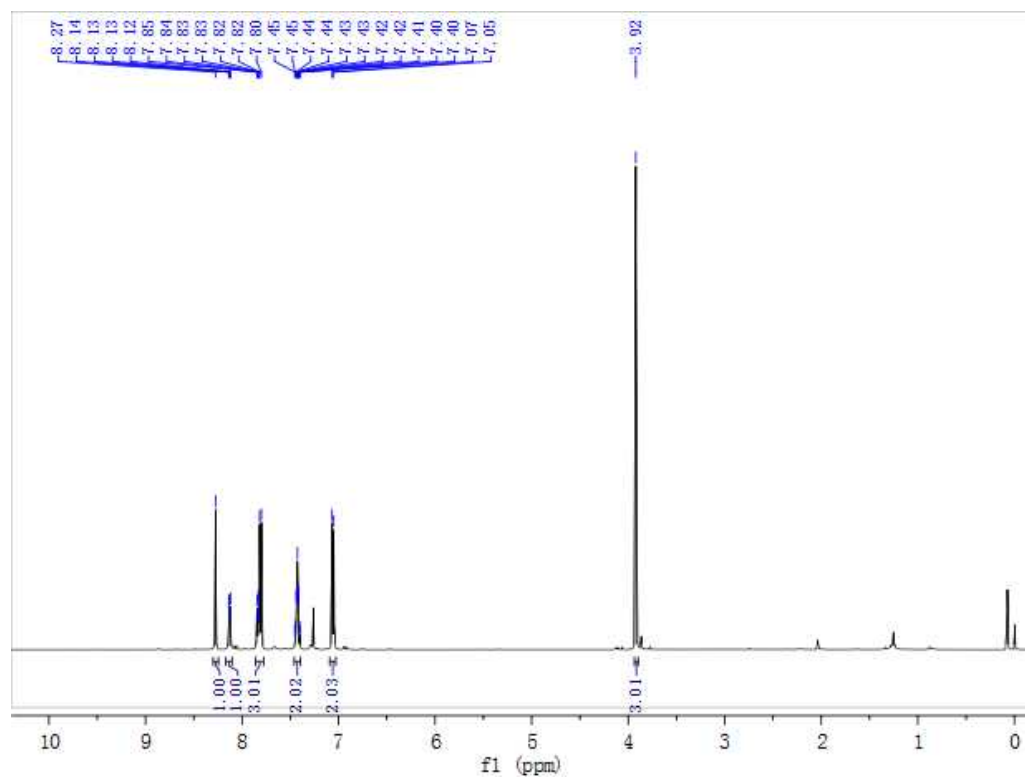
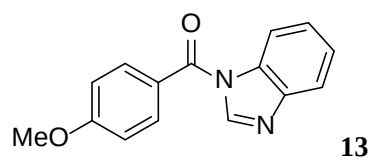
(1*H*-Benzo[d]imidazol-1-yl)(4-chlorophenyl)methanone (CAS: 71589-37-2)



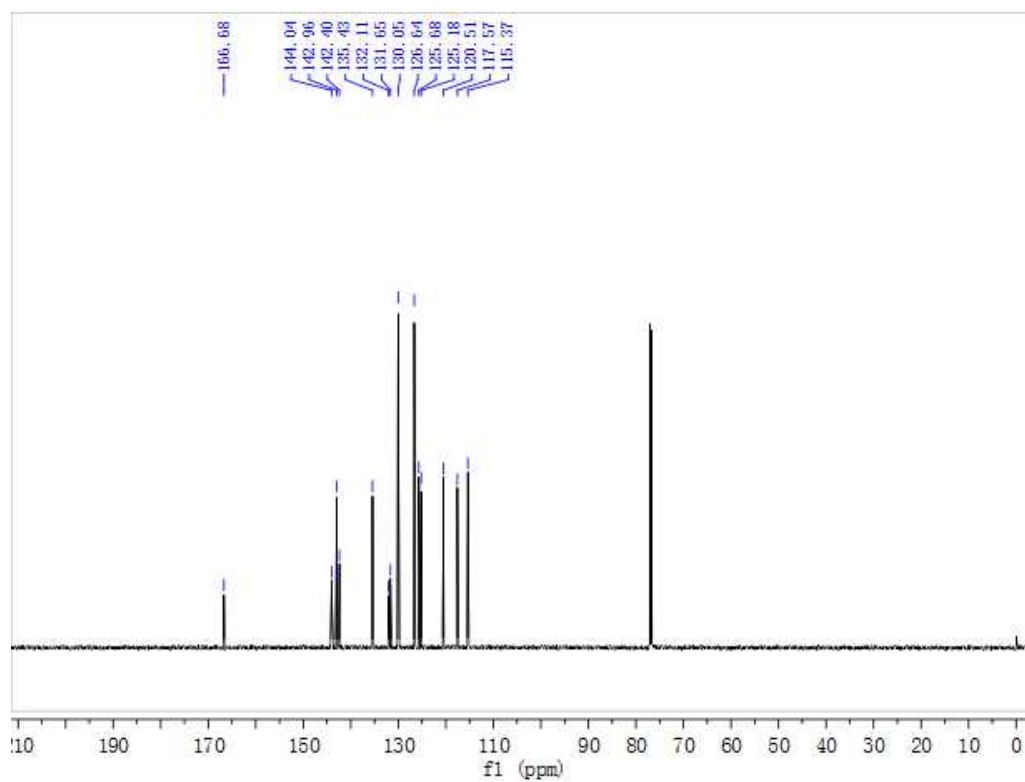
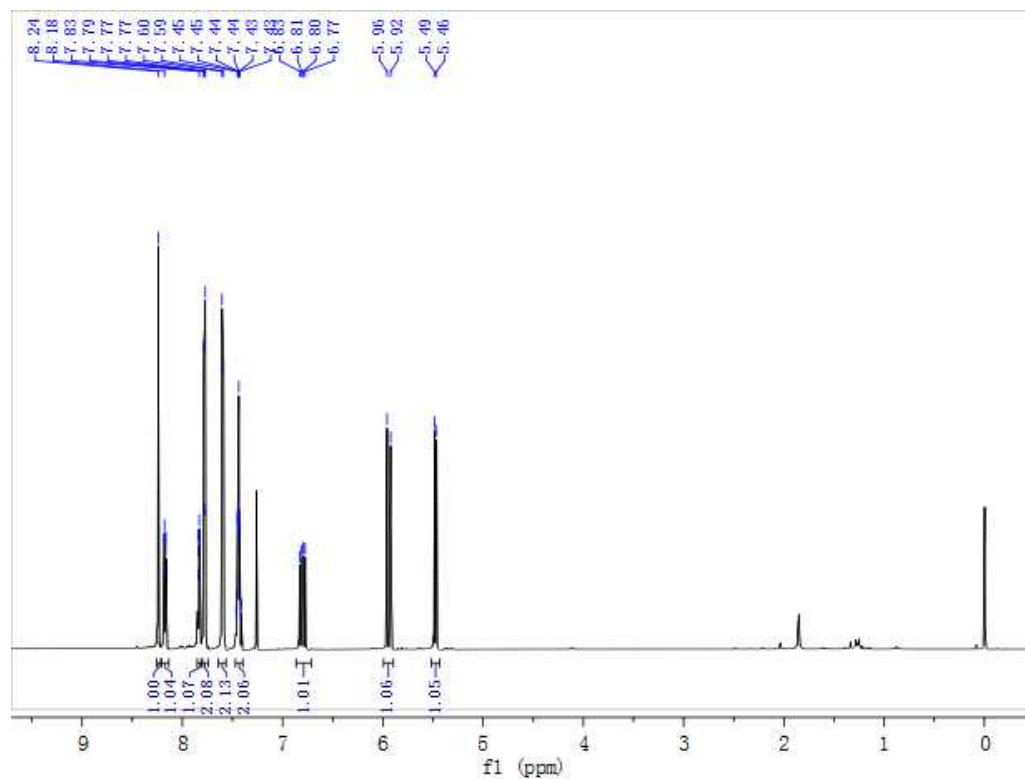
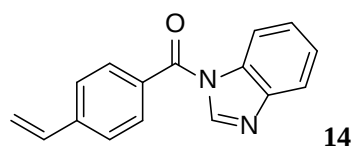
(1*H*-Benzo[d]imidazol-1-yl)(4-bromophenyl)methanone (CAS: 304668-33-5)



(1*H*-Benzo[d]imidazol-1-yl)(4-methoxyphenyl)methanone (CAS: 13361-55-2)



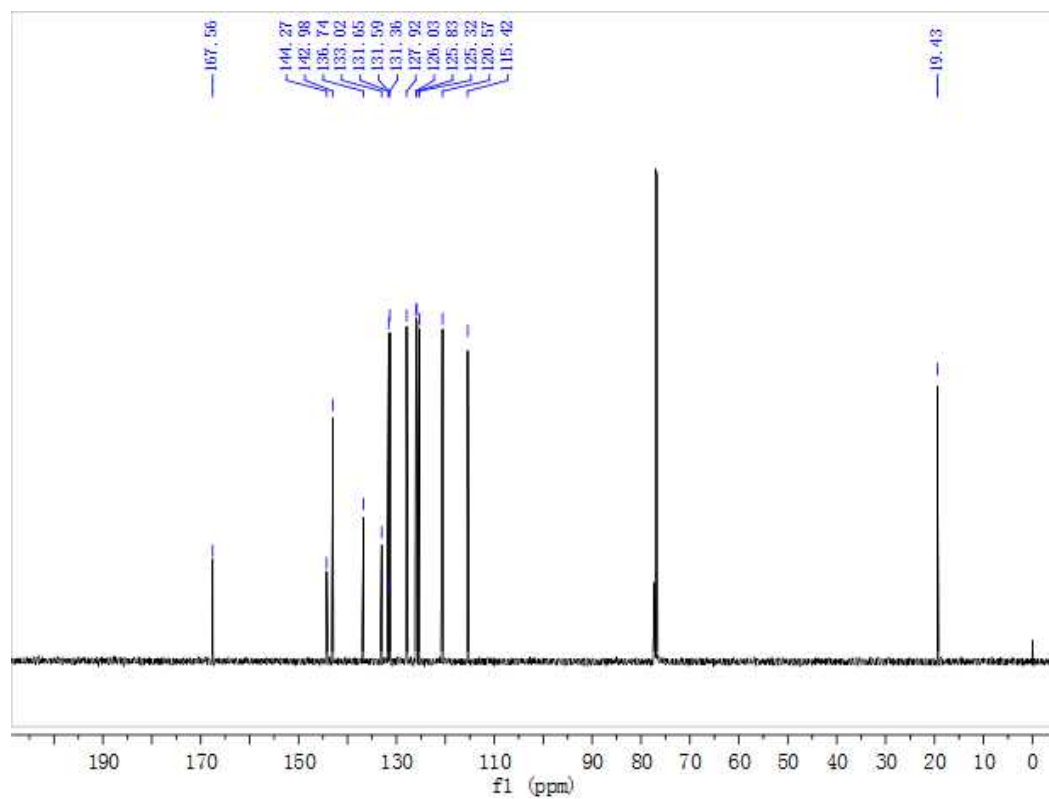
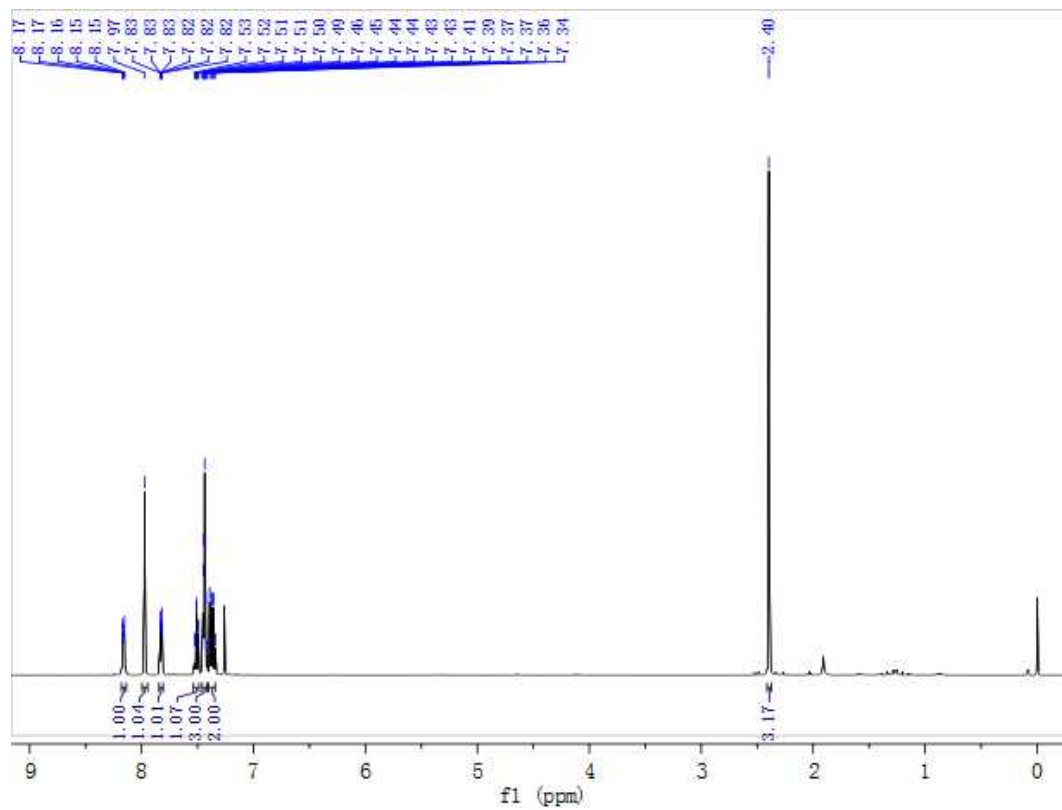
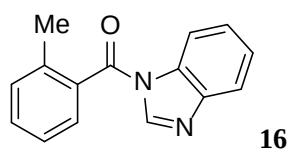
(1*H*-Benzo[*d*]imidazol-1-yl)(4-vinylphenyl)methanone (CAS: 300396-82-1)



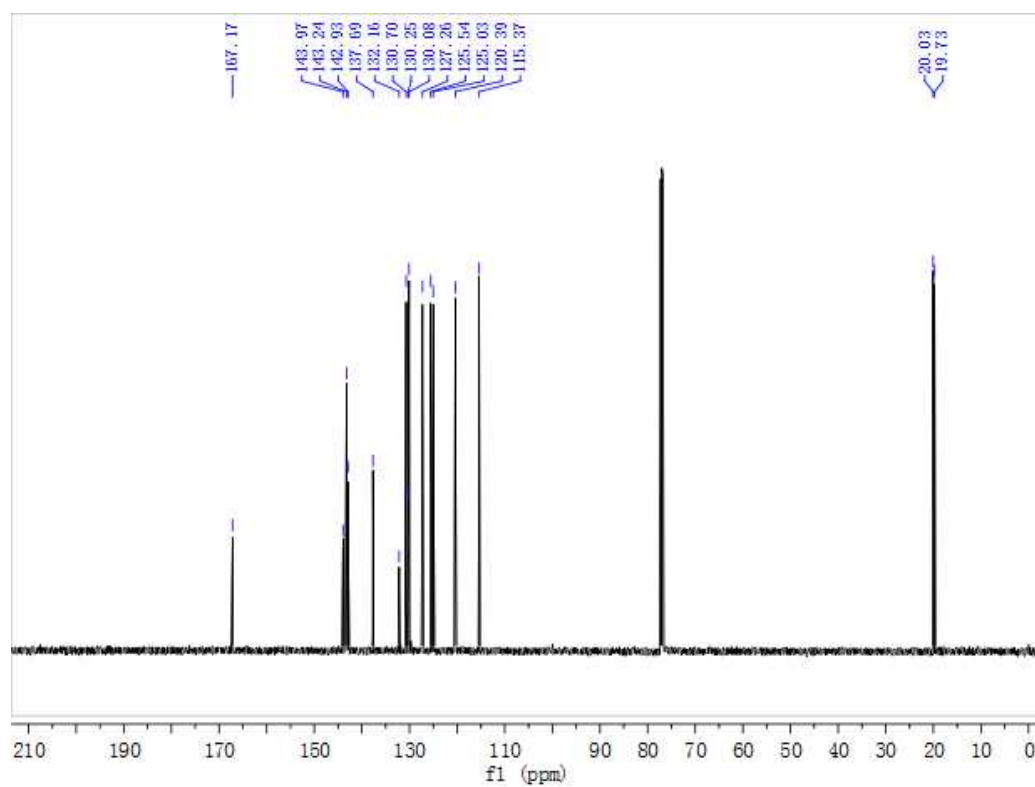
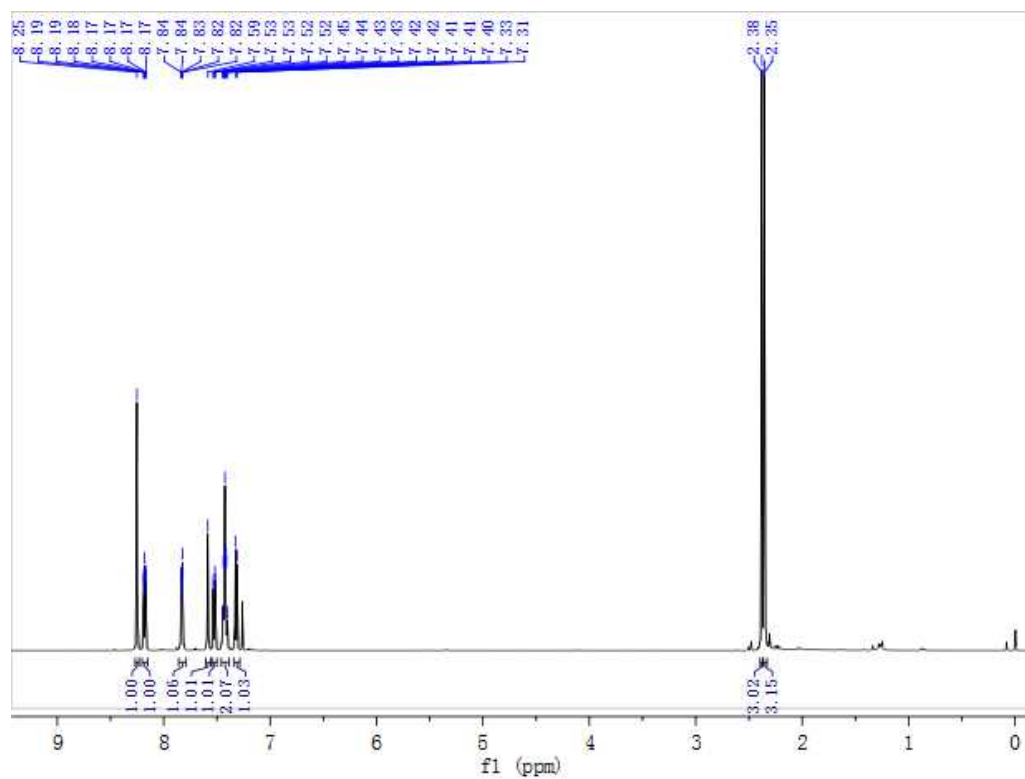
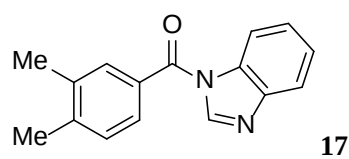
Chemical structure of 1-(4-(cyclohexyl)phenyl)pyrrolidine-2,5-dione. The structure consists of a cyclohexane ring attached to a para-substituted benzene ring. The benzene ring is further substituted with a pyrrolidine-2,5-dione ring (a five-membered ring with two carbonyl groups and two nitrogen atoms).

[illegible]

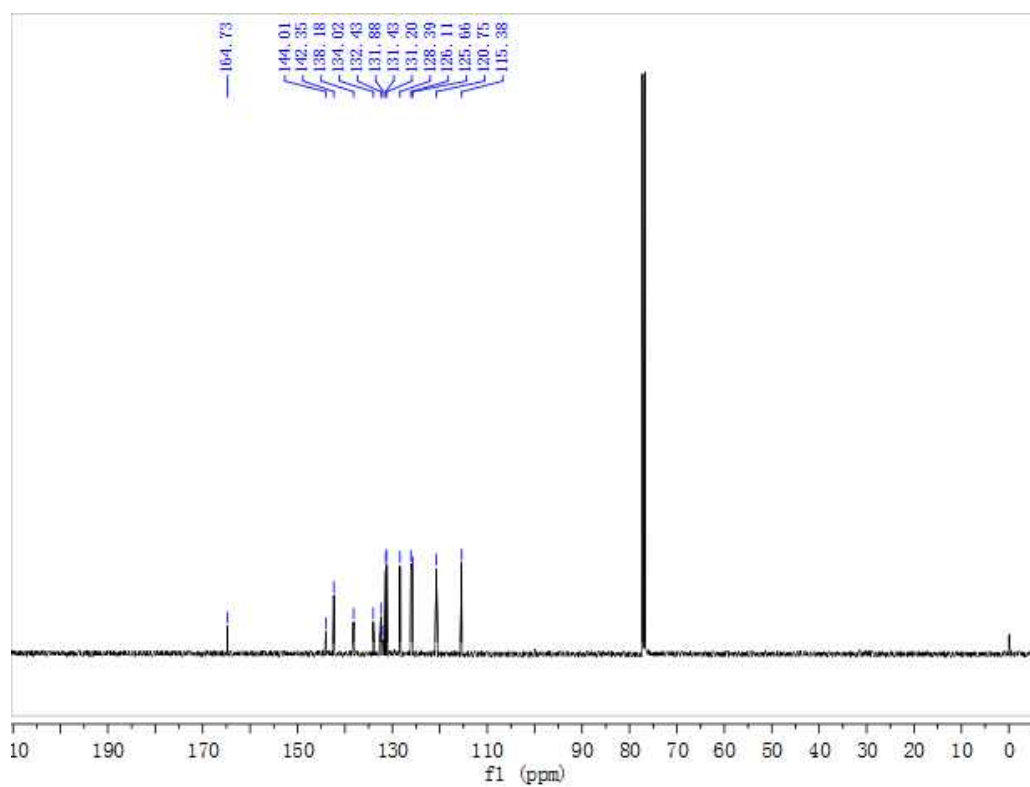
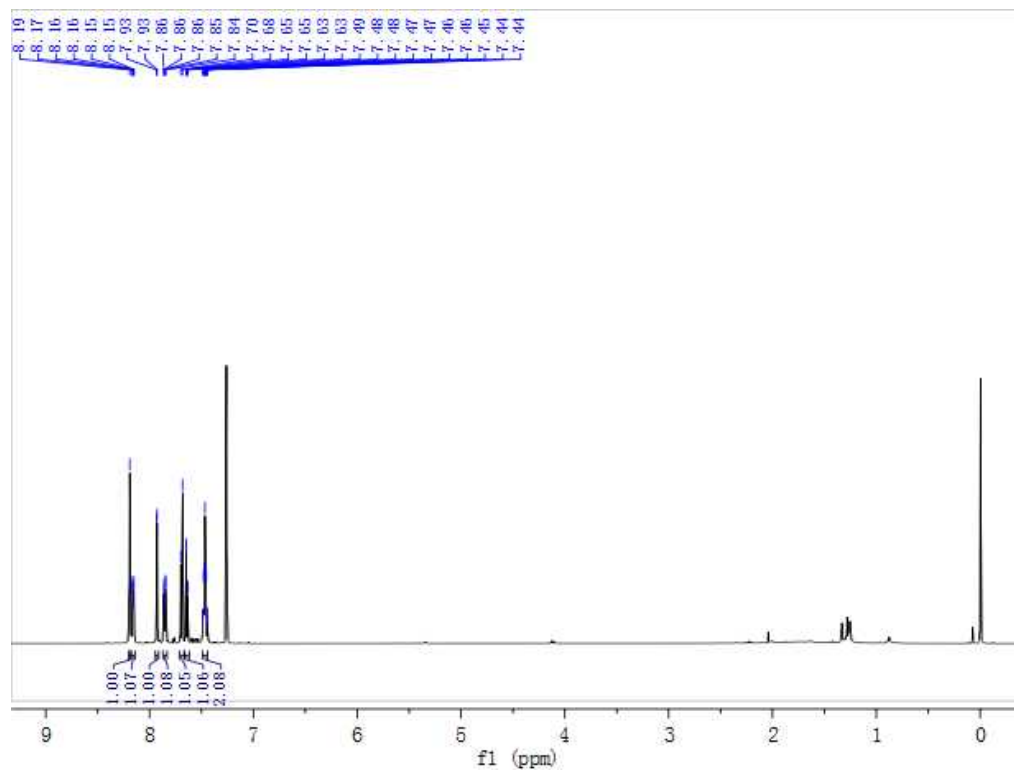
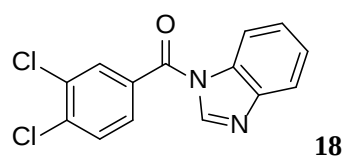
(1*H*-Benzo[d]imidazol-1-yl)(*o*-tolyl)methanone (CAS: 200626-52-4)



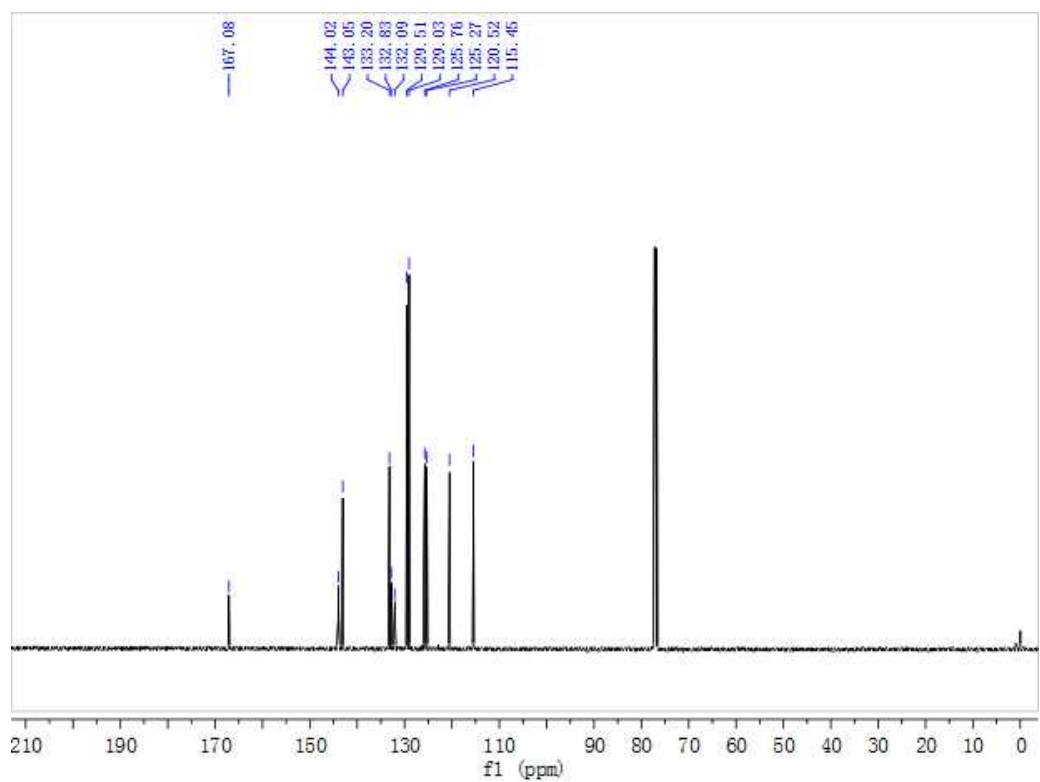
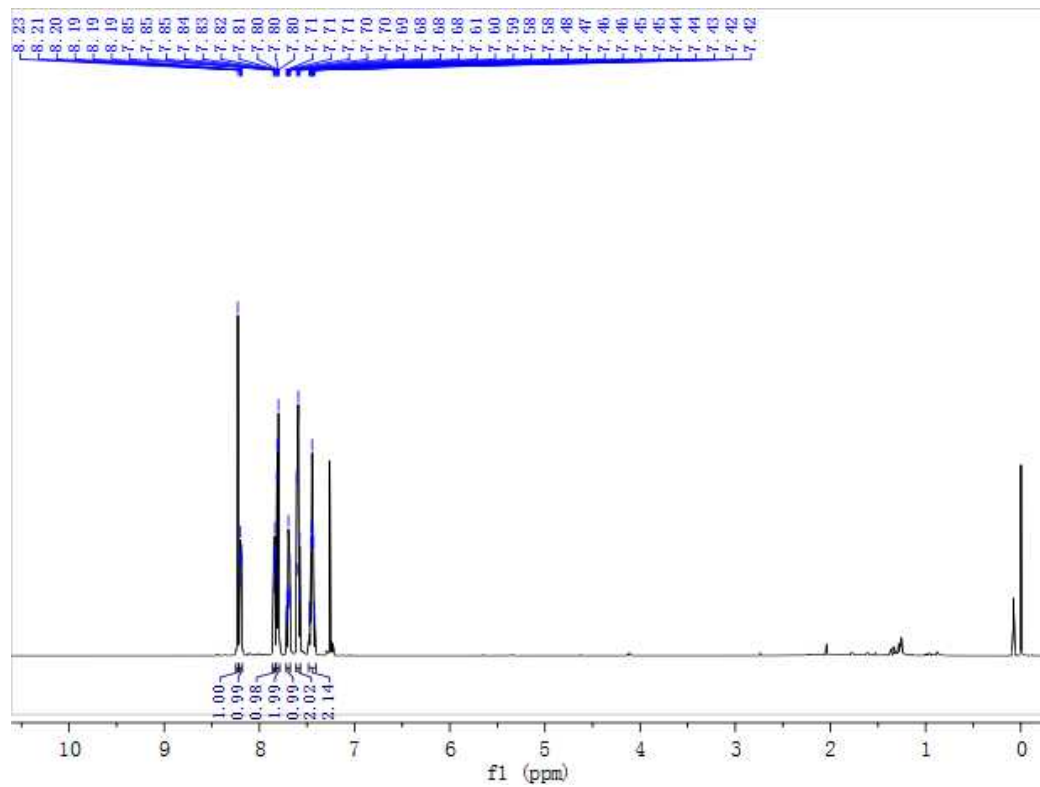
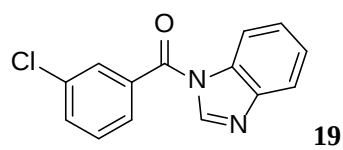
(1H-Benzo[d]imidazol-1-yl)(3,4-dimethylphenyl)methanone (CAS: 333348-55-3)



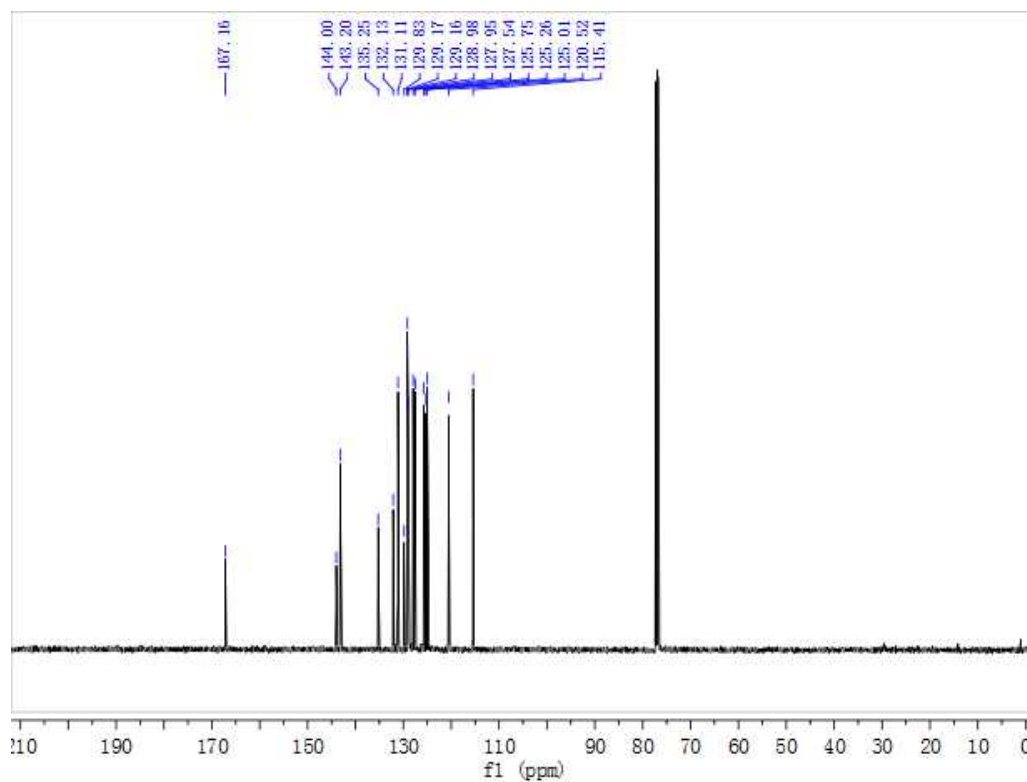
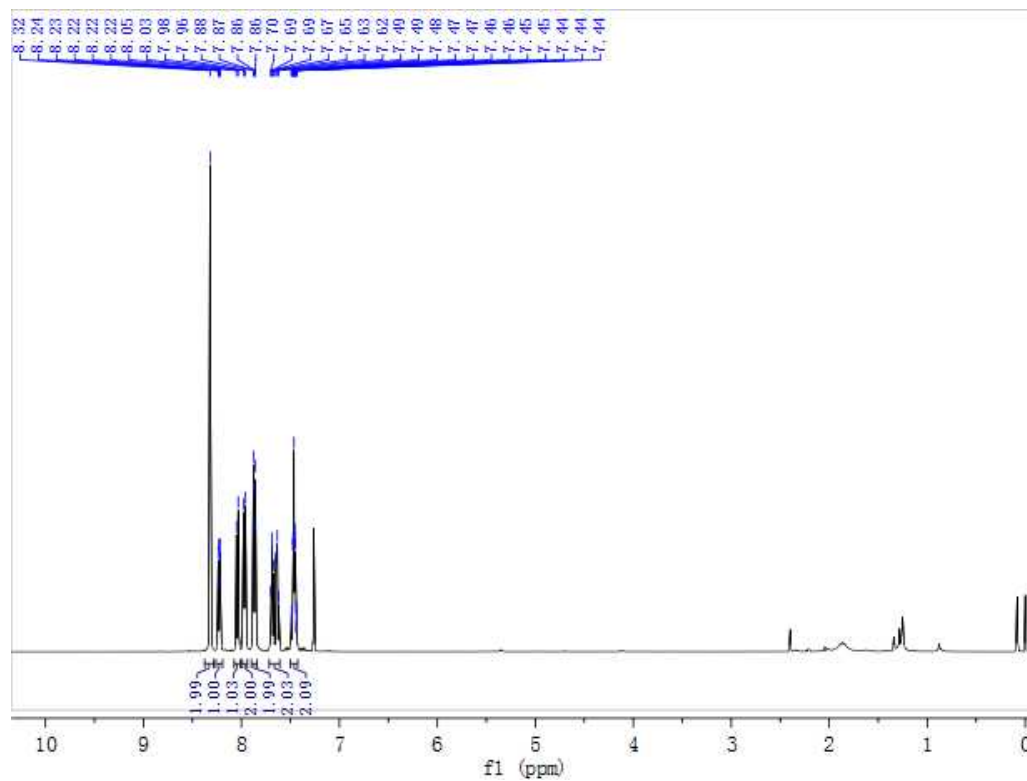
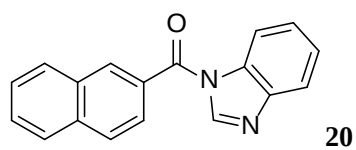
(1*H*-Benzo[d]imidazol-1-yl)(3,4-dichlorophenyl)methanone (CAS: 330215-63-9)



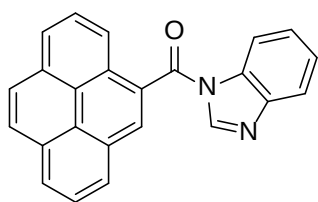
(1H-Benzo[d]imidazol-1-yl)(3-chlorophenyl)methanone (CAS :200626-53-5)



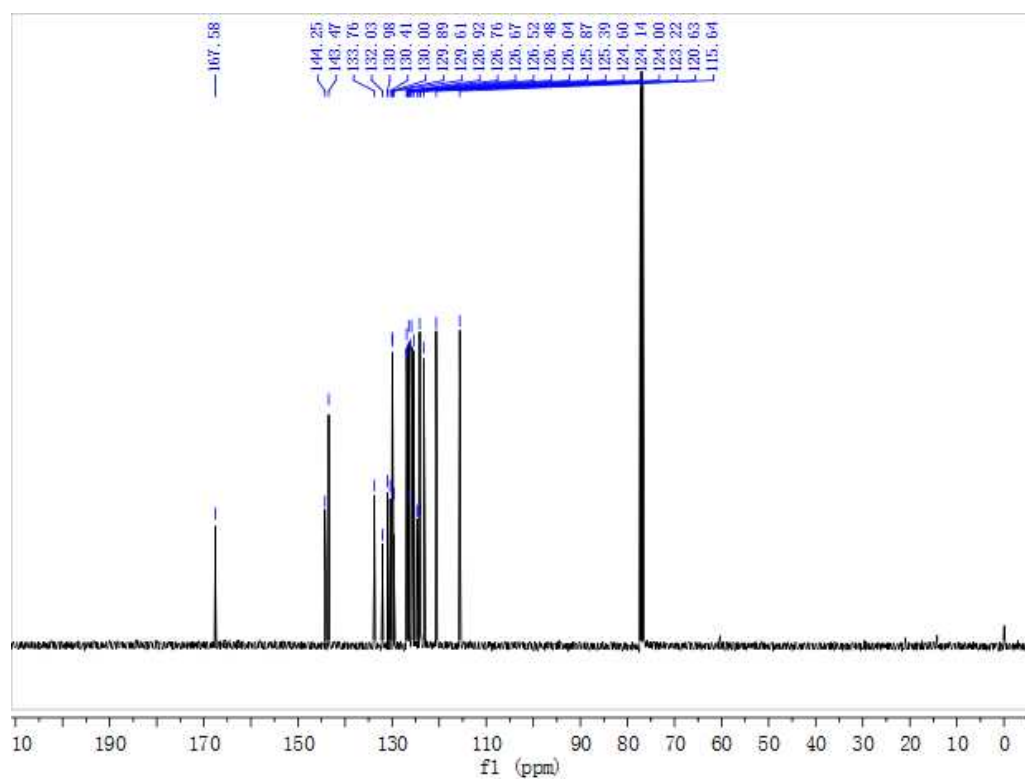
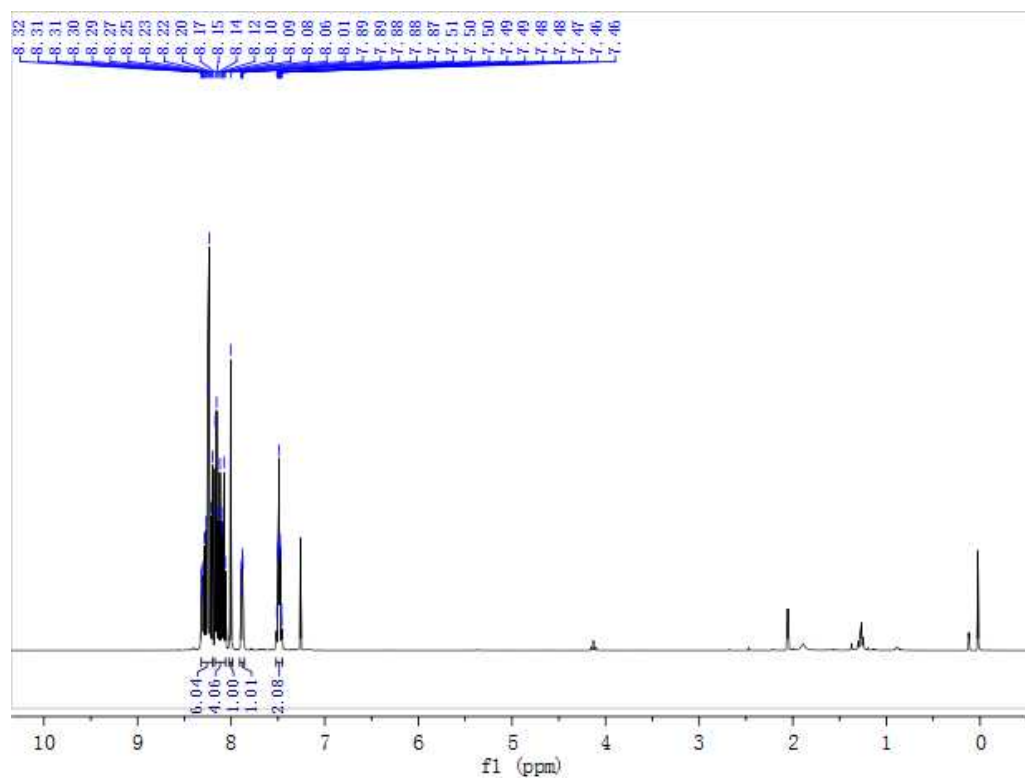
(1*H*-Benzo[*d*]imidazol-1-yl)(naphthalen-2-yl)methanone (new compound)



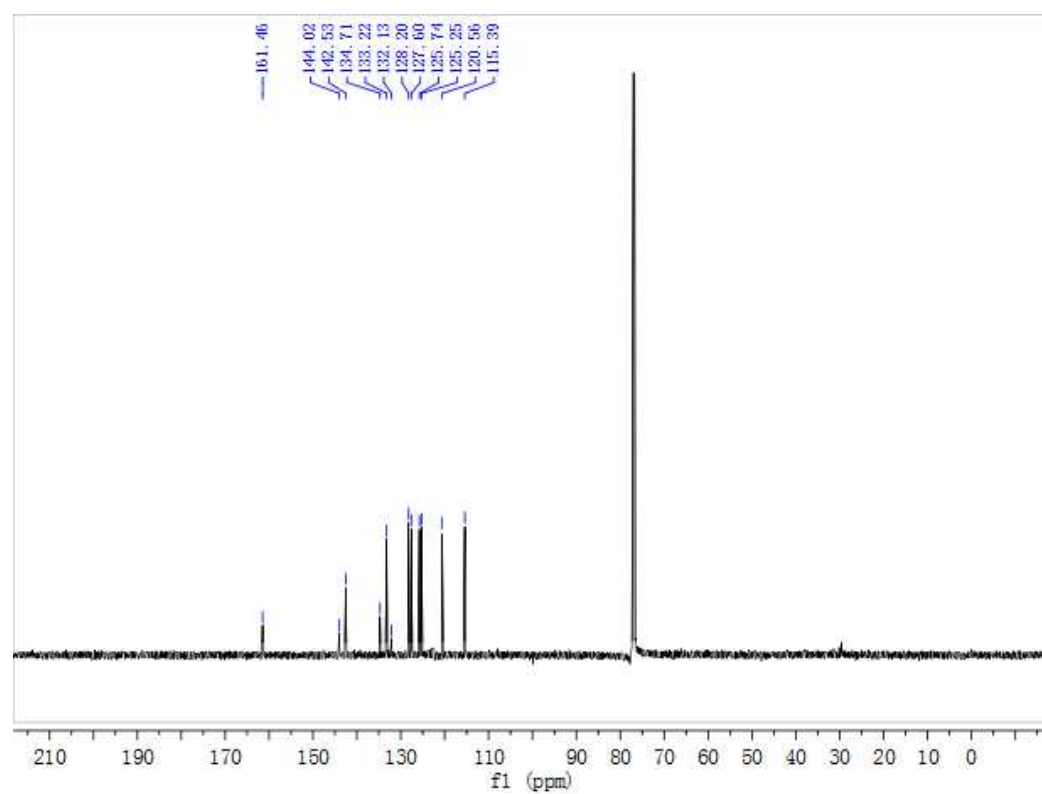
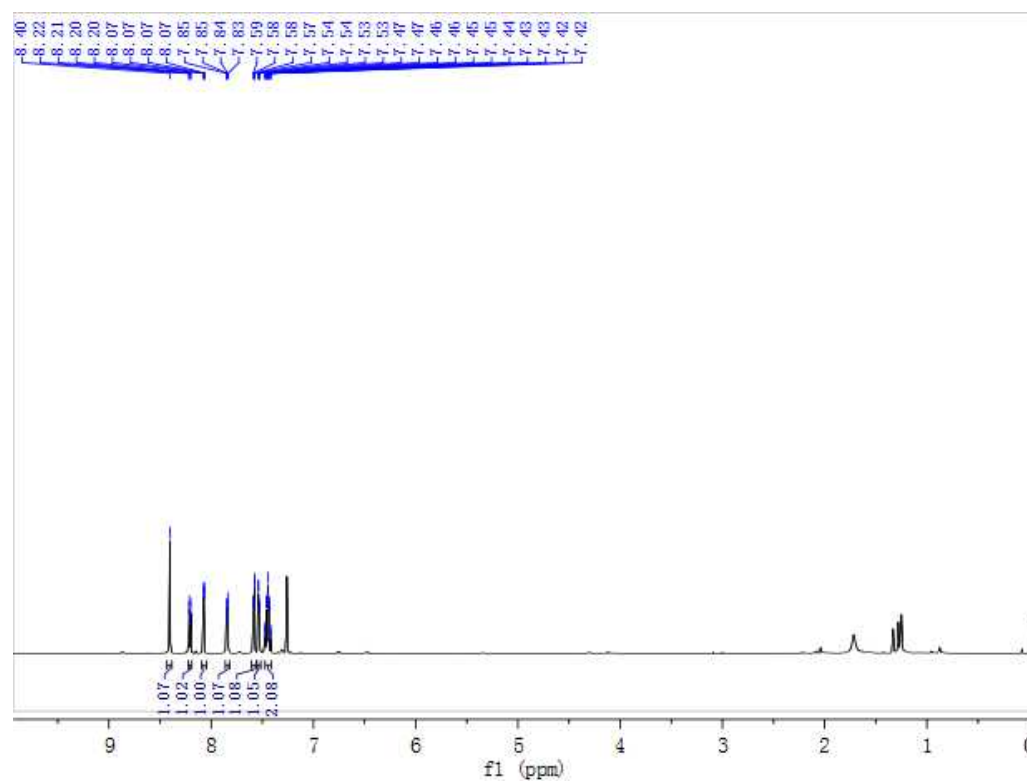
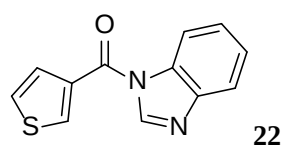
(1H-Benzo[d]imidazol-1-yl)(pyren-4-yl)methanone (new compound)



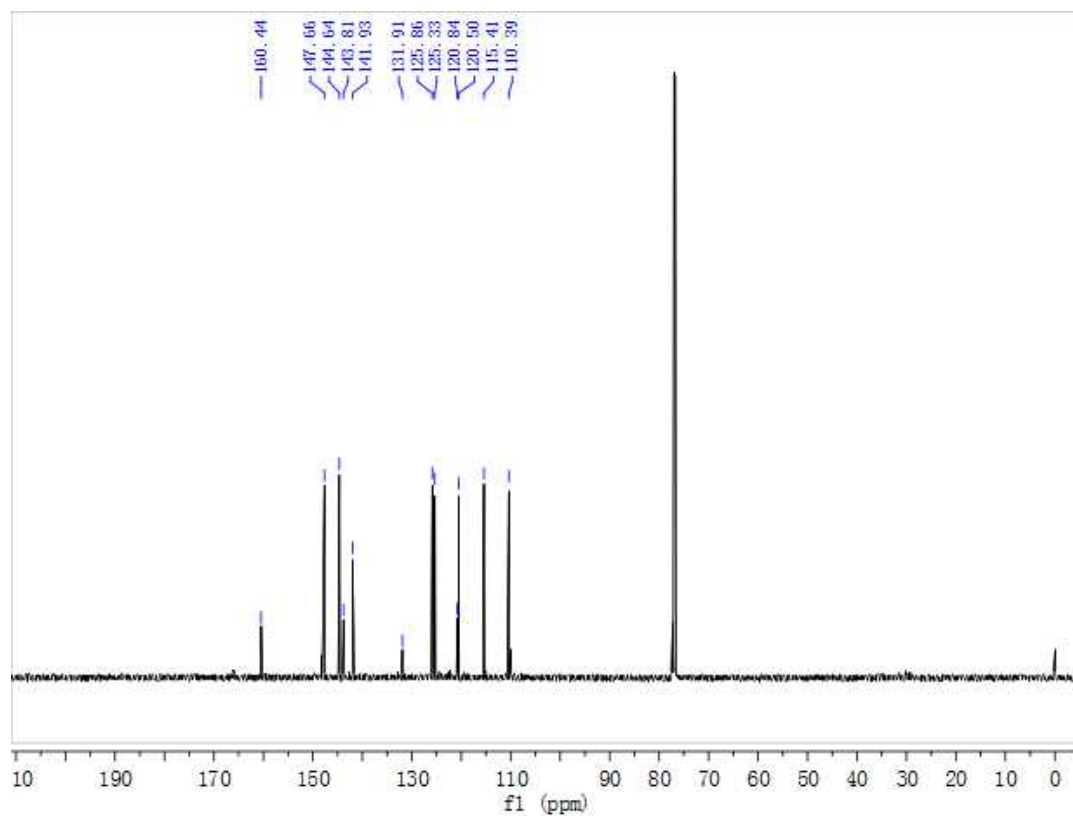
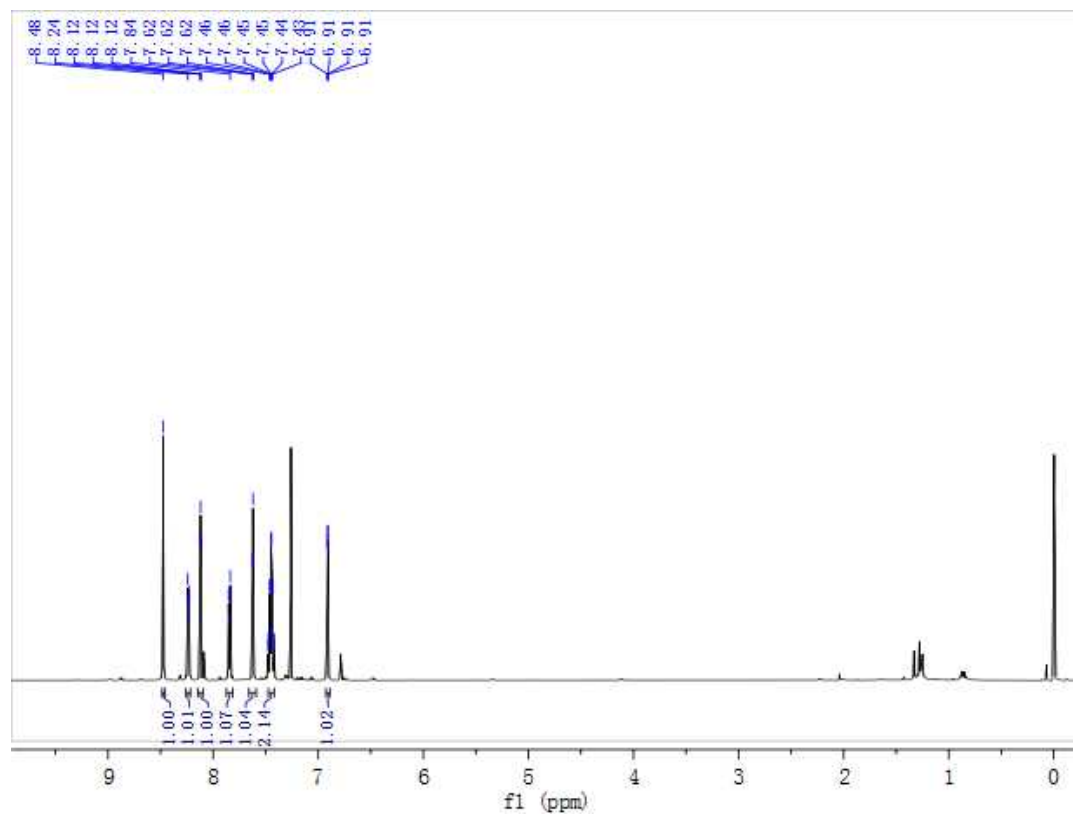
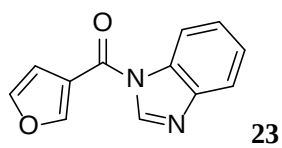
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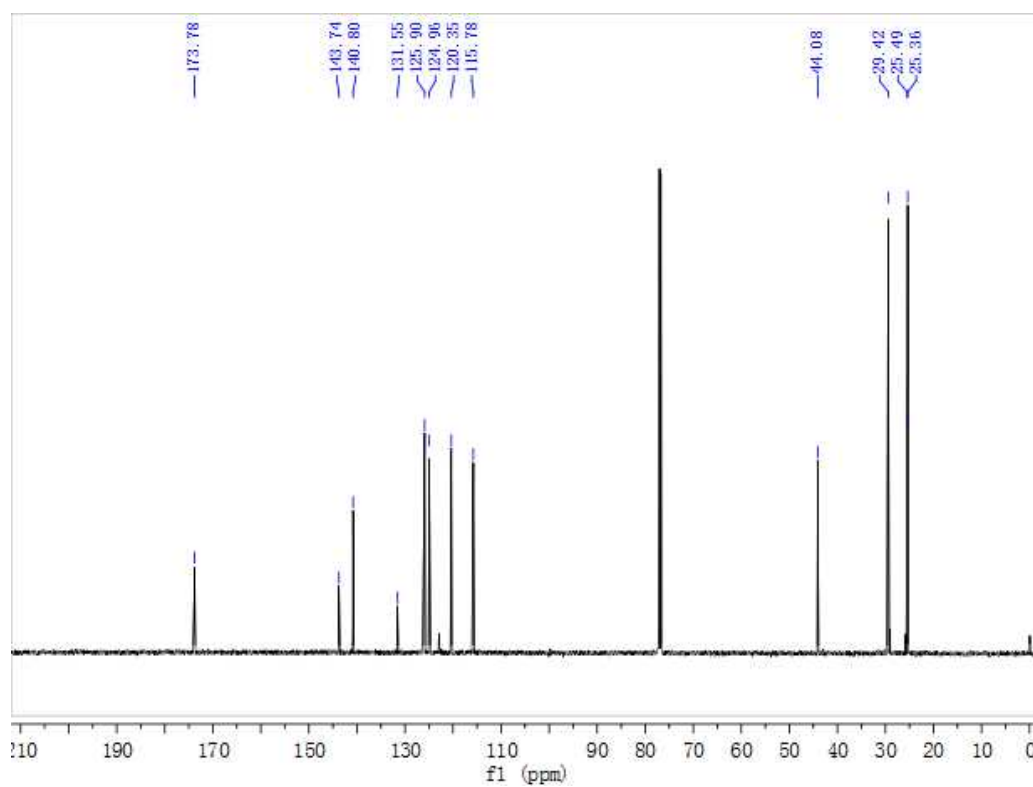
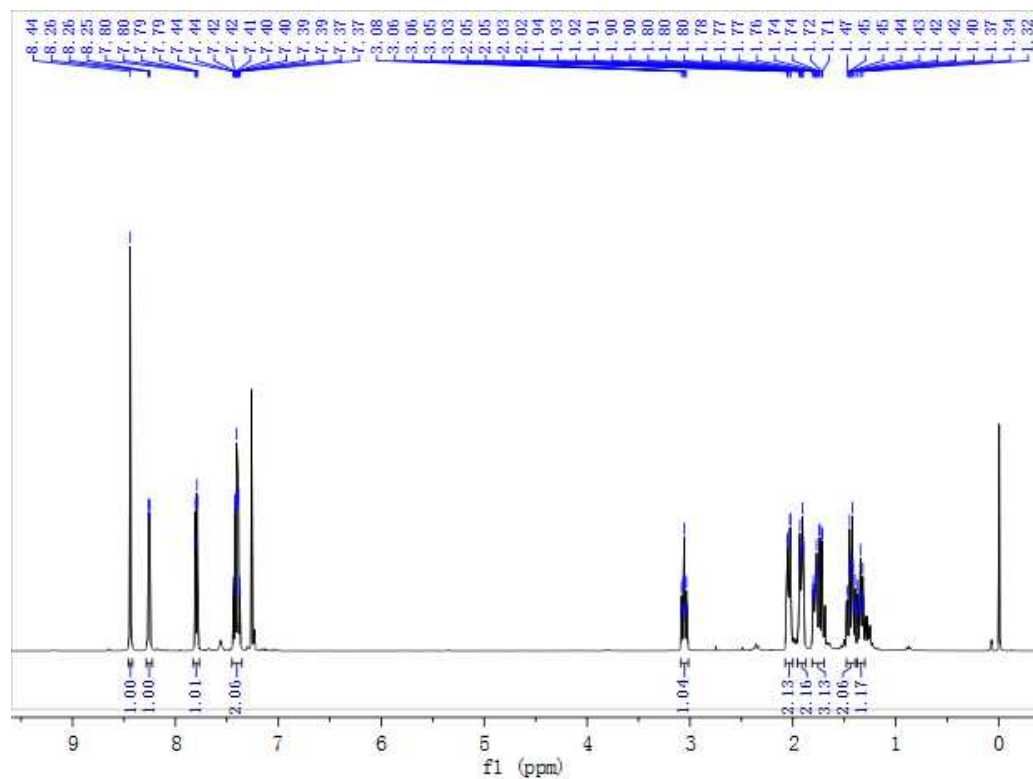
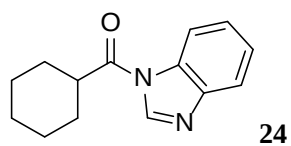
(1*H*-Benzo[*d*]imidazol-1-yl)(thiophen-3-yl)methanone (new compound)



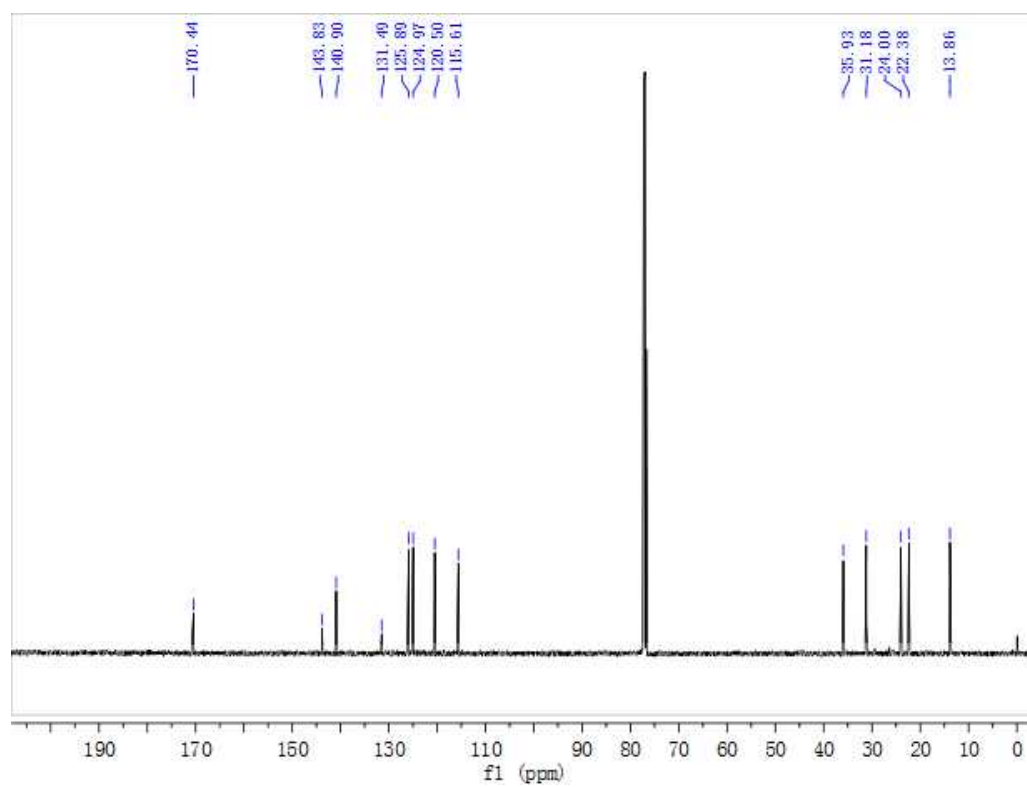
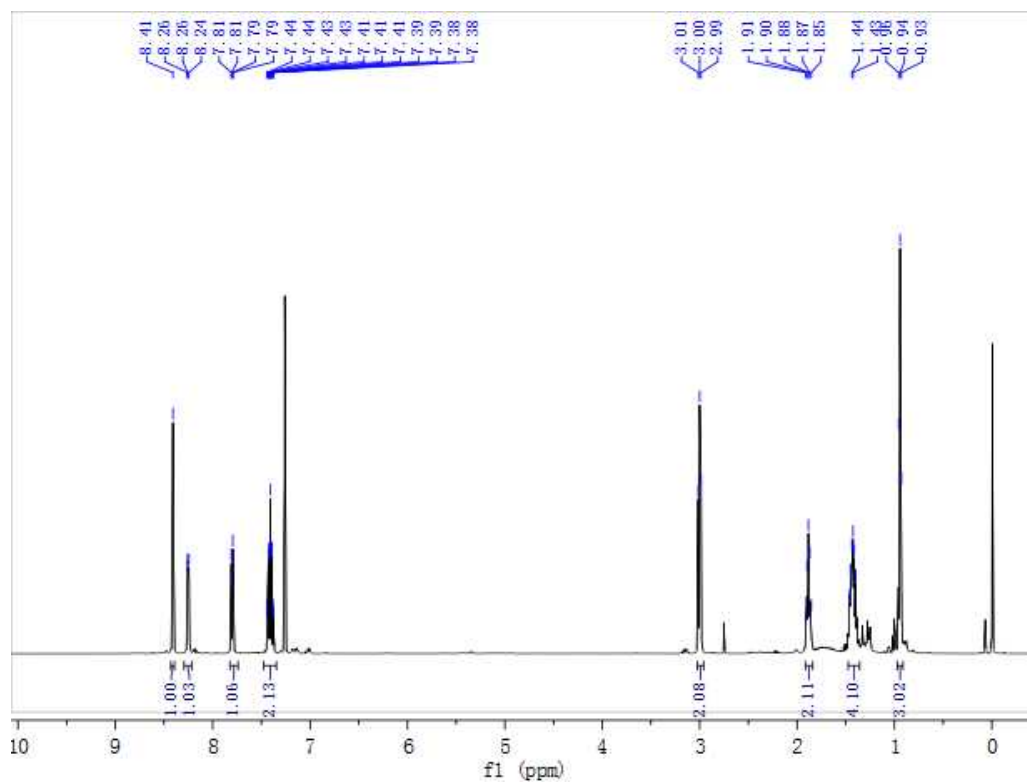
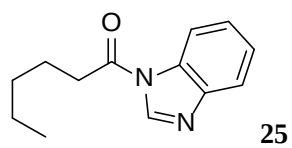
(1*H*-Benzo[d]imidazol-1-yl)(furan-3-yl)methanone (new compound)



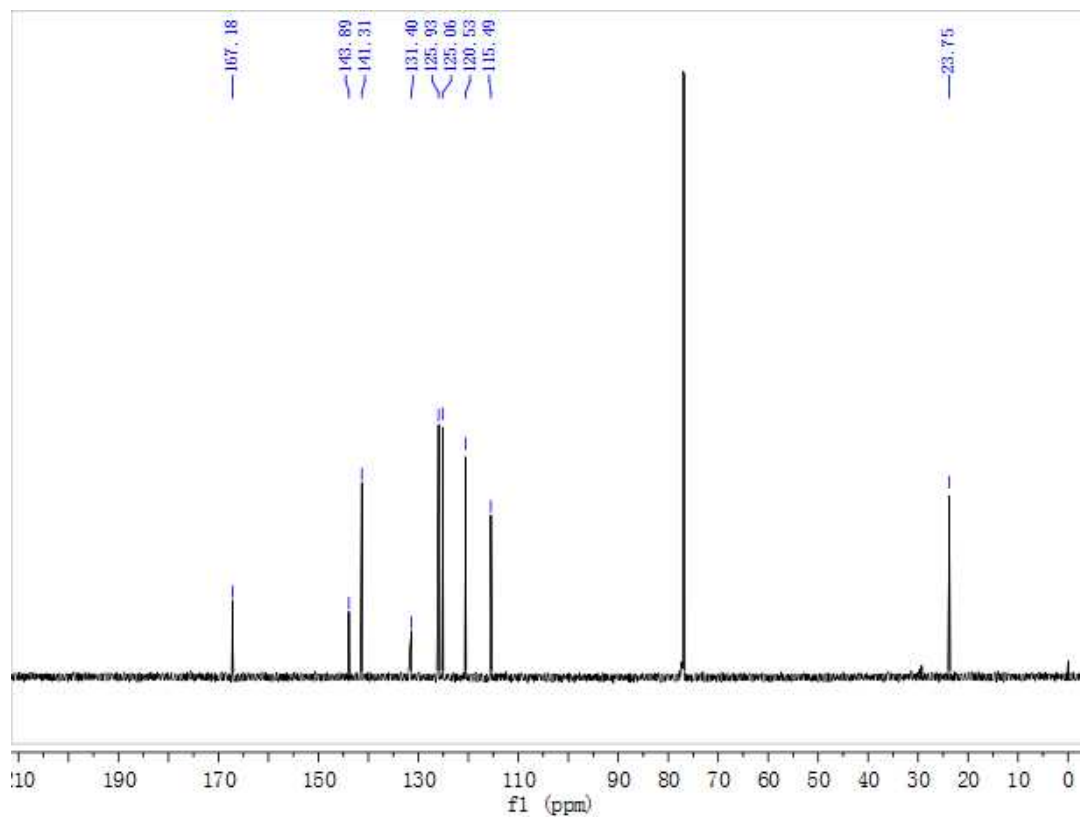
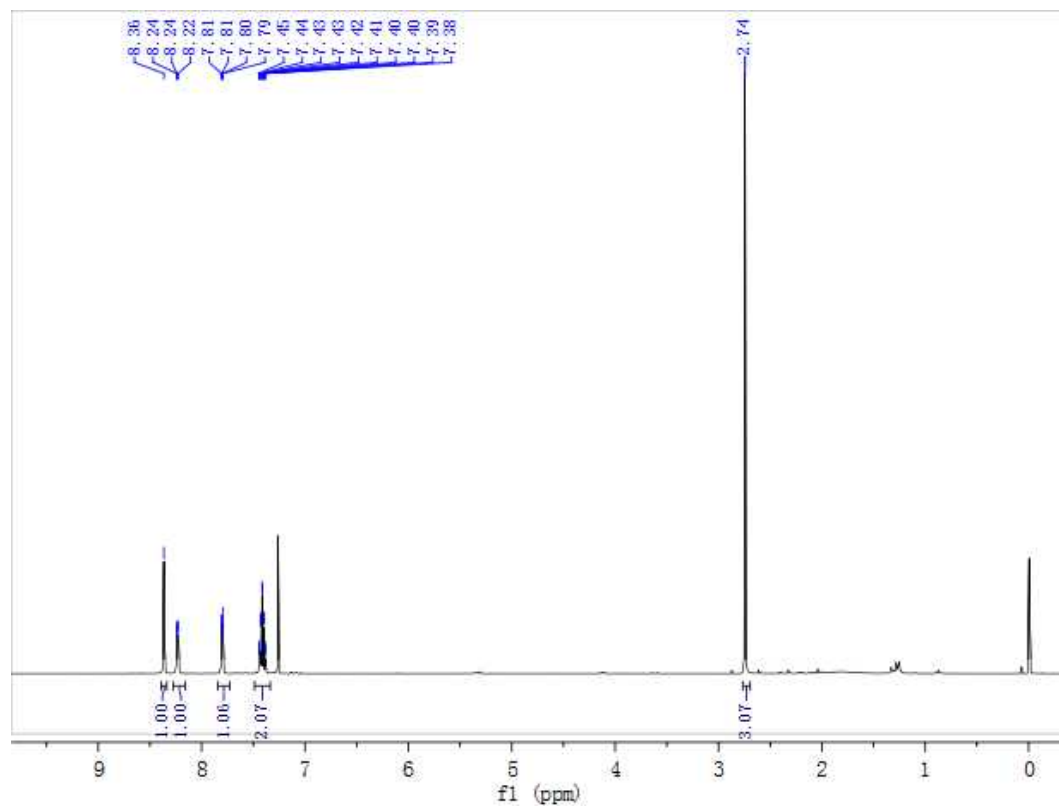
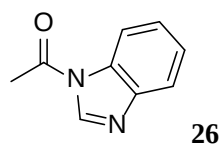
(1H-Benzo[d]imidazol-1-yl)(cyclohexyl)methanone (CAS: 294649-09-5)



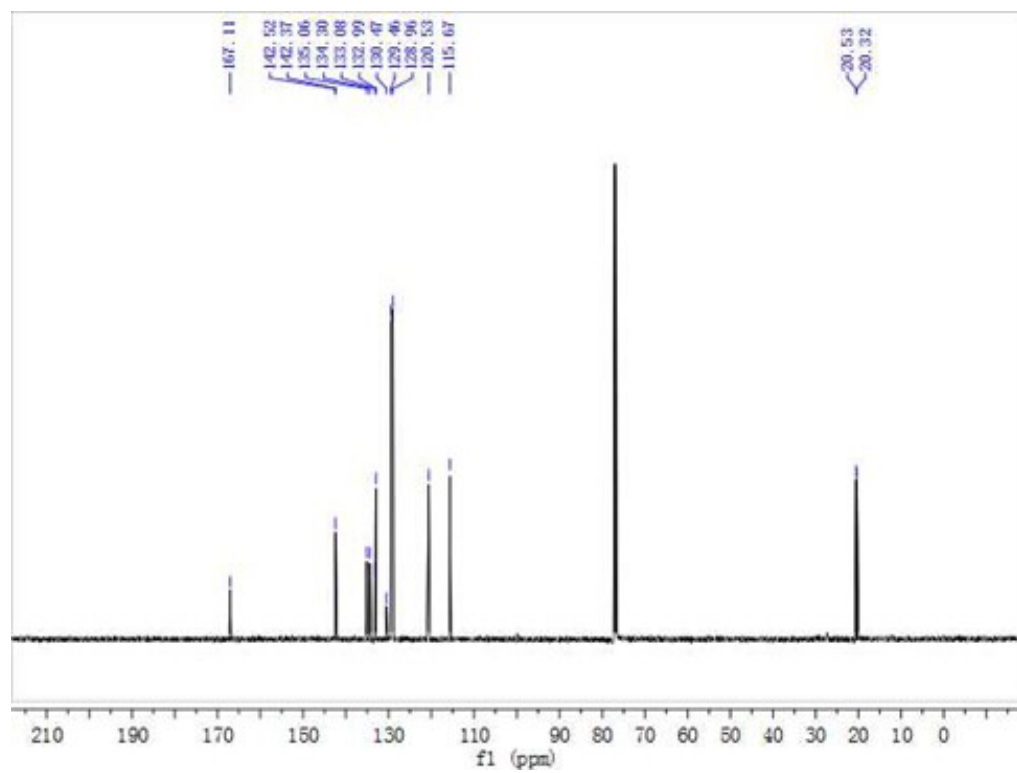
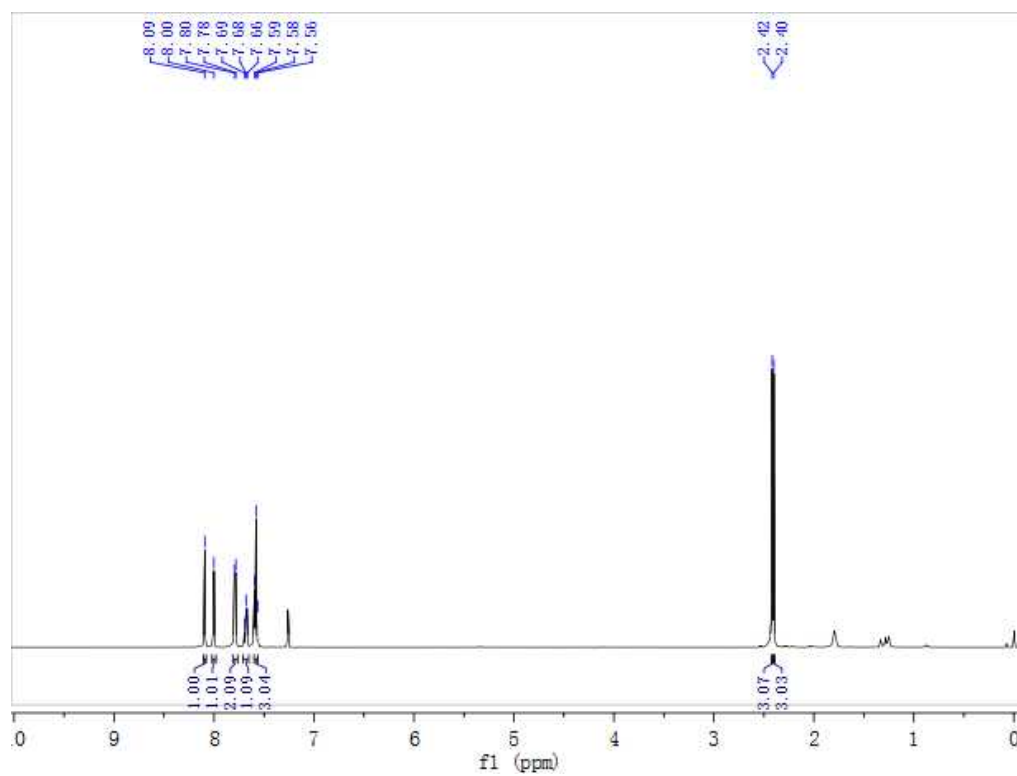
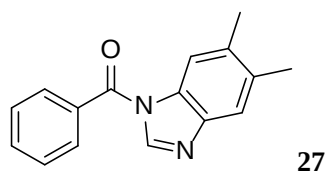
1-(1*H*-Benzo[d]imidazol-1-yl)hexan-1-one (CAS: 901547-84-0)



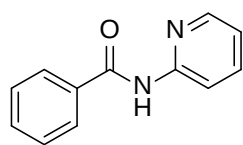
1-(1*H*-Benzo[*d*]imidazol-1-yl)ethan-1-one (CAS: 18773-95-0)



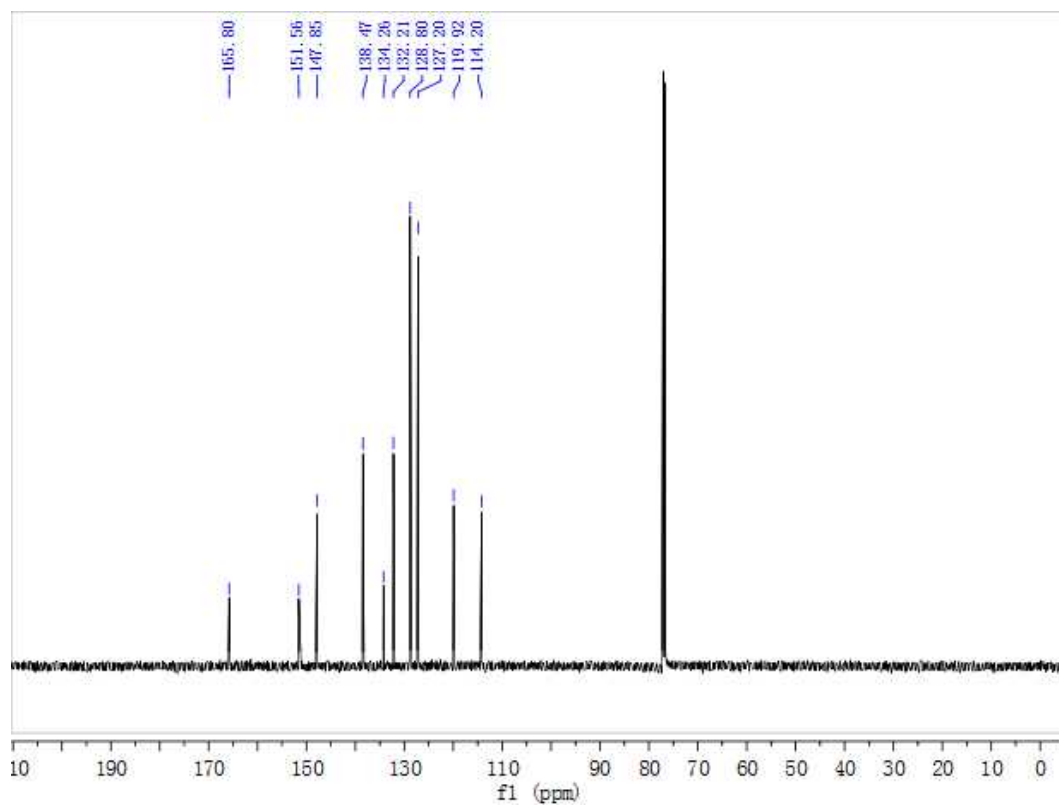
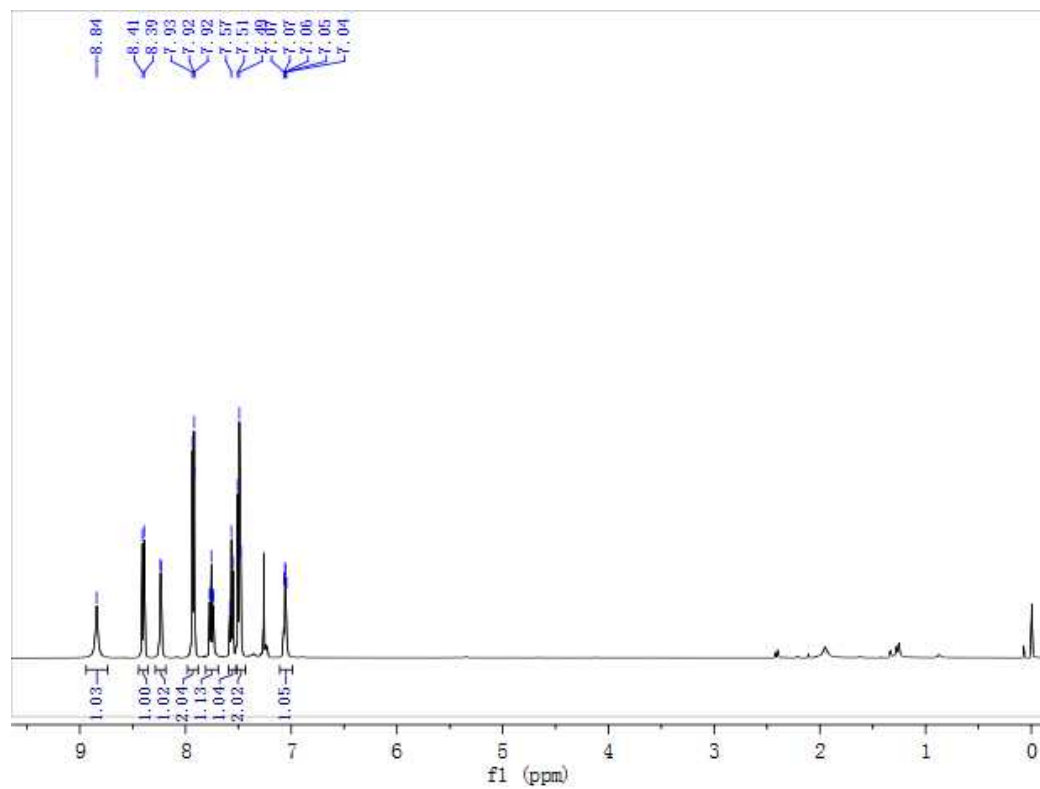
(5,6-Dimethyl-1*H*-benzo[d]imidazol-1-yl)(phenyl)methanone (CAS: 16109-46-9)



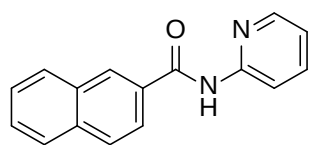
***N*-(Pyridin-2-yl)benzamide (CAS : 4589-12-2)**



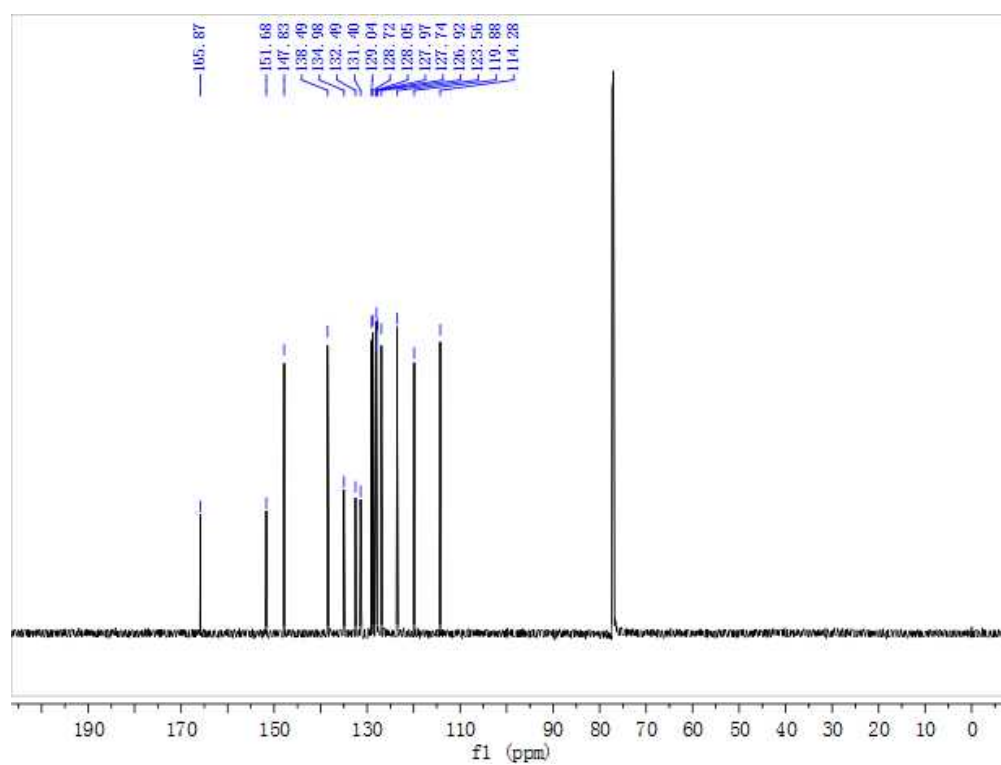
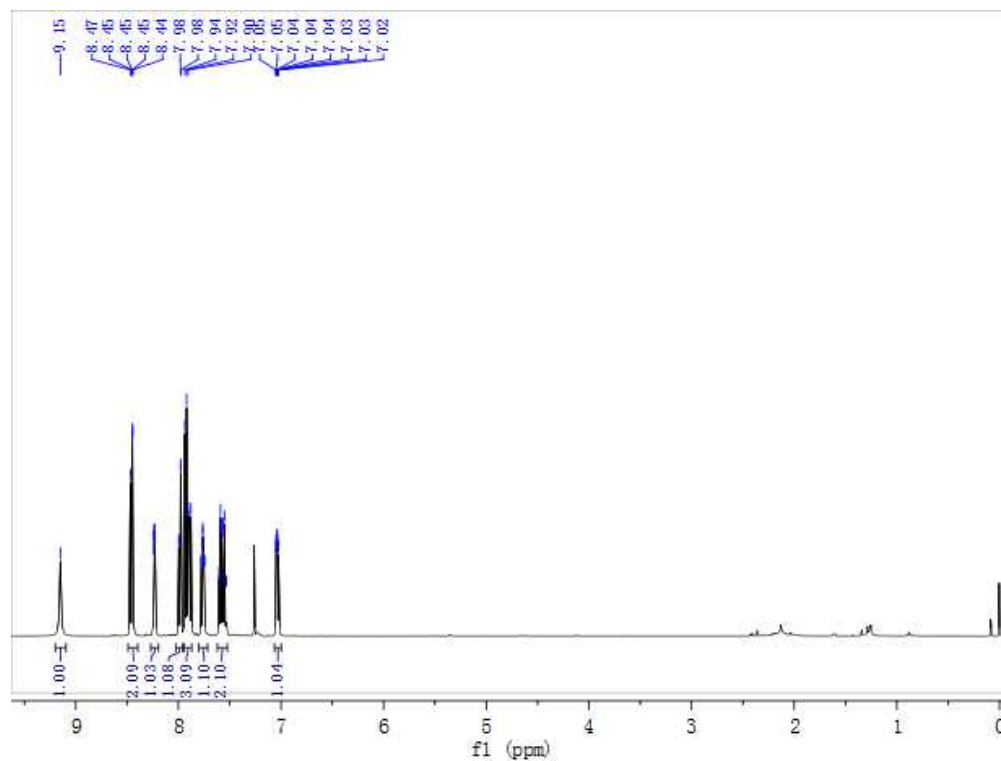
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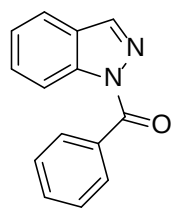
N-(Pyridin-2-yl)-2-naphthamide (CAS : 159257-88-2)



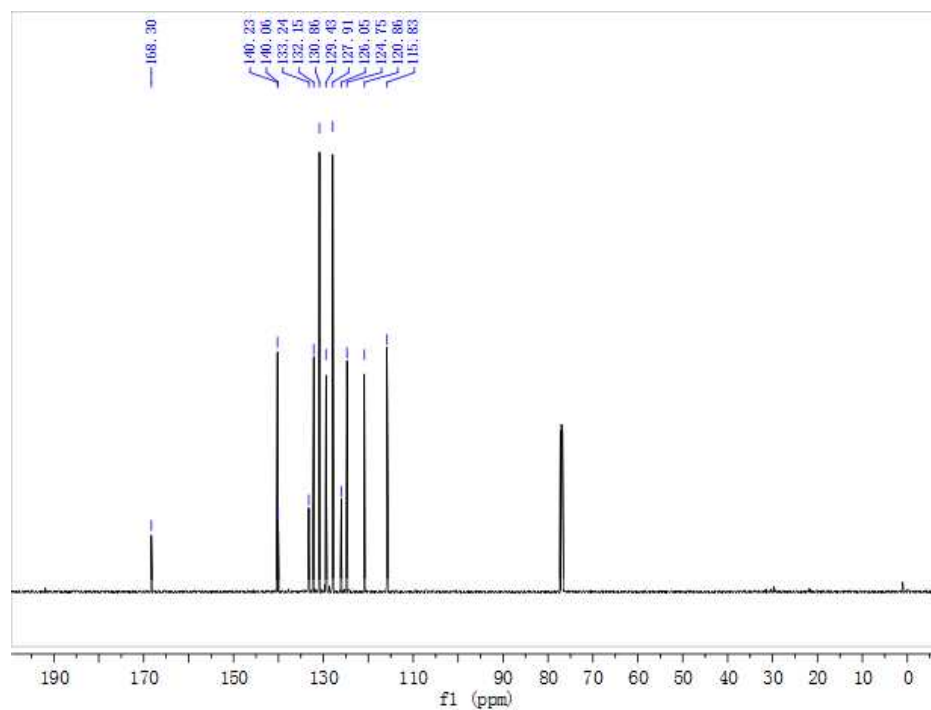
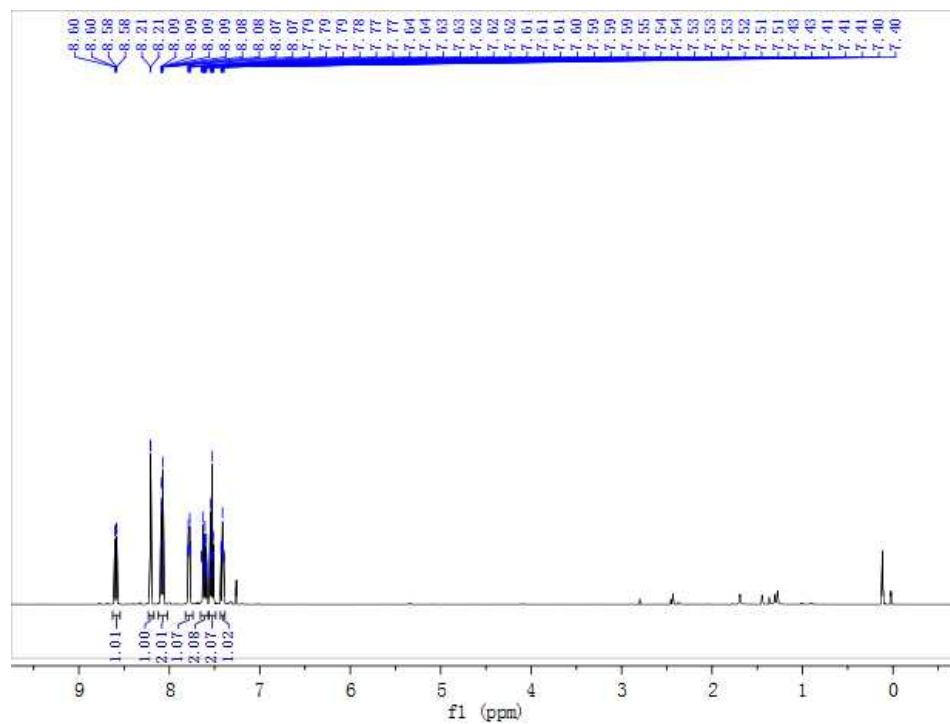
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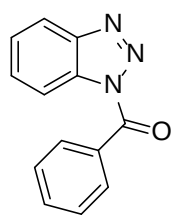
(1*H*-Indazol-1-yl)(phenyl)methanone (CAS: 23301-00-0)



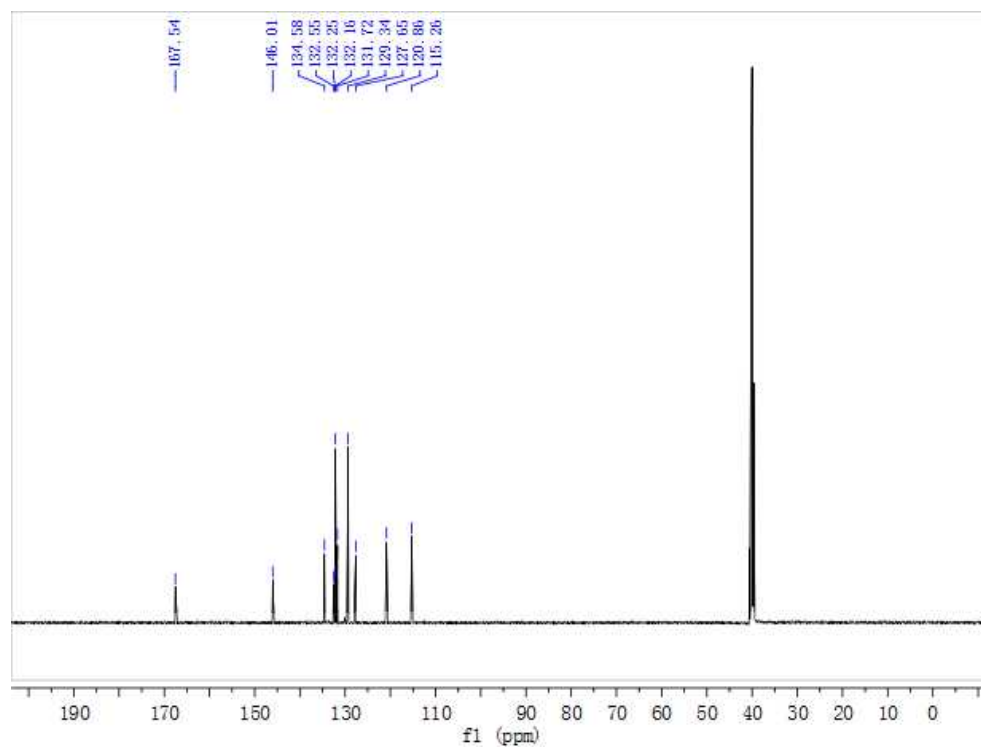
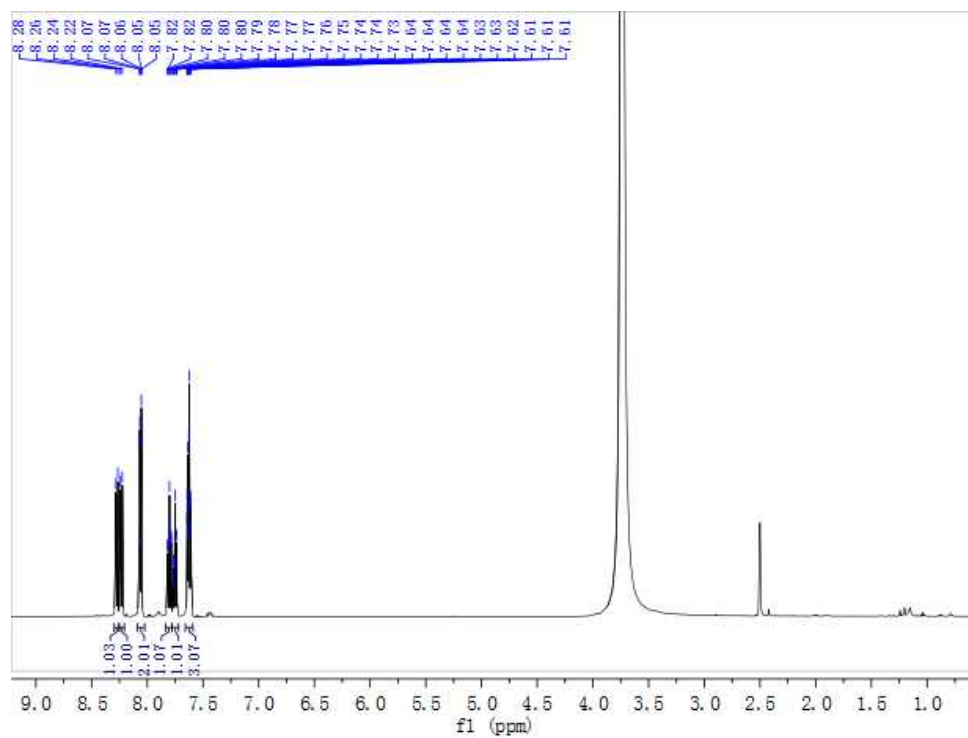
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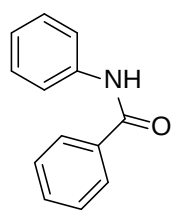
(1H-Benzo[d][1,2,3]triazol-1-yl)(phenyl)methanone (CAS: 4231-62-3)



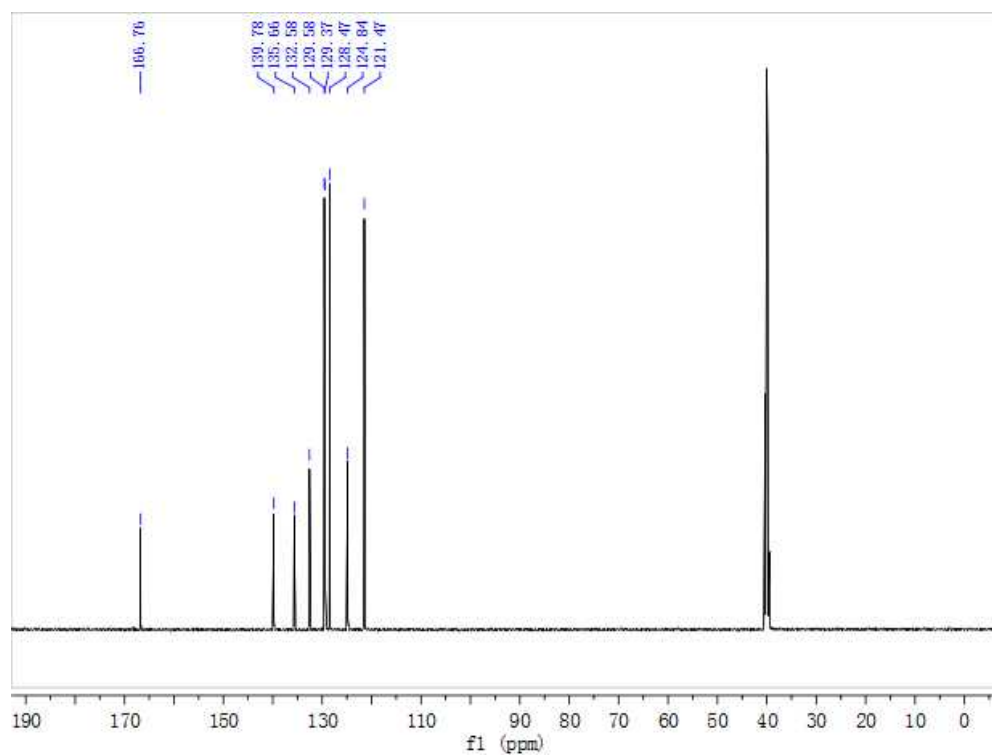
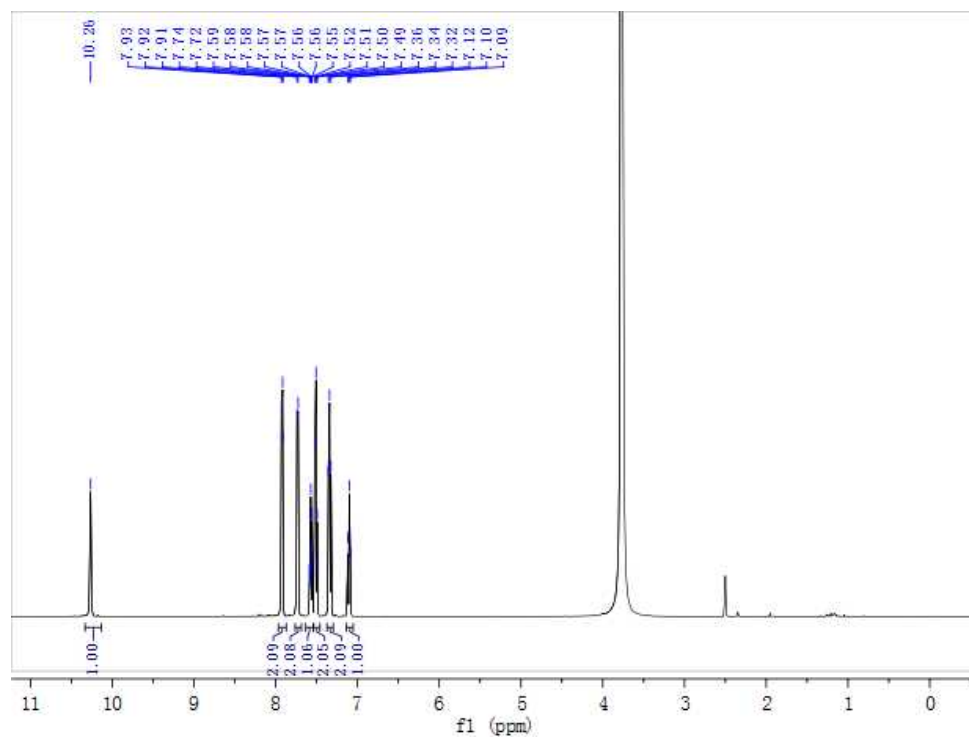
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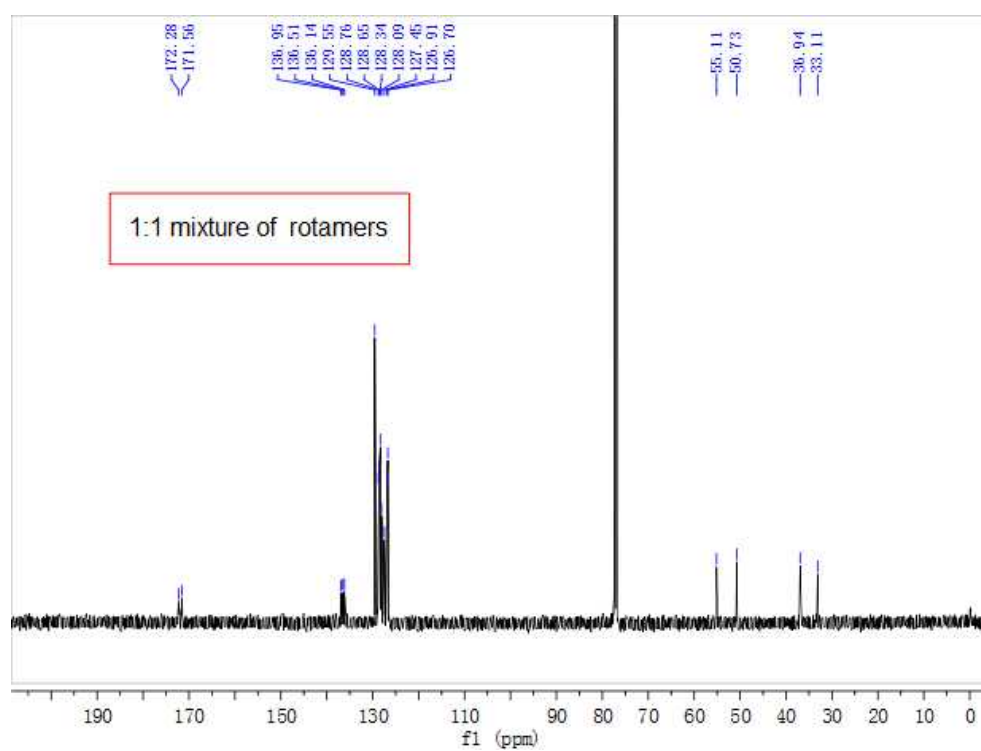
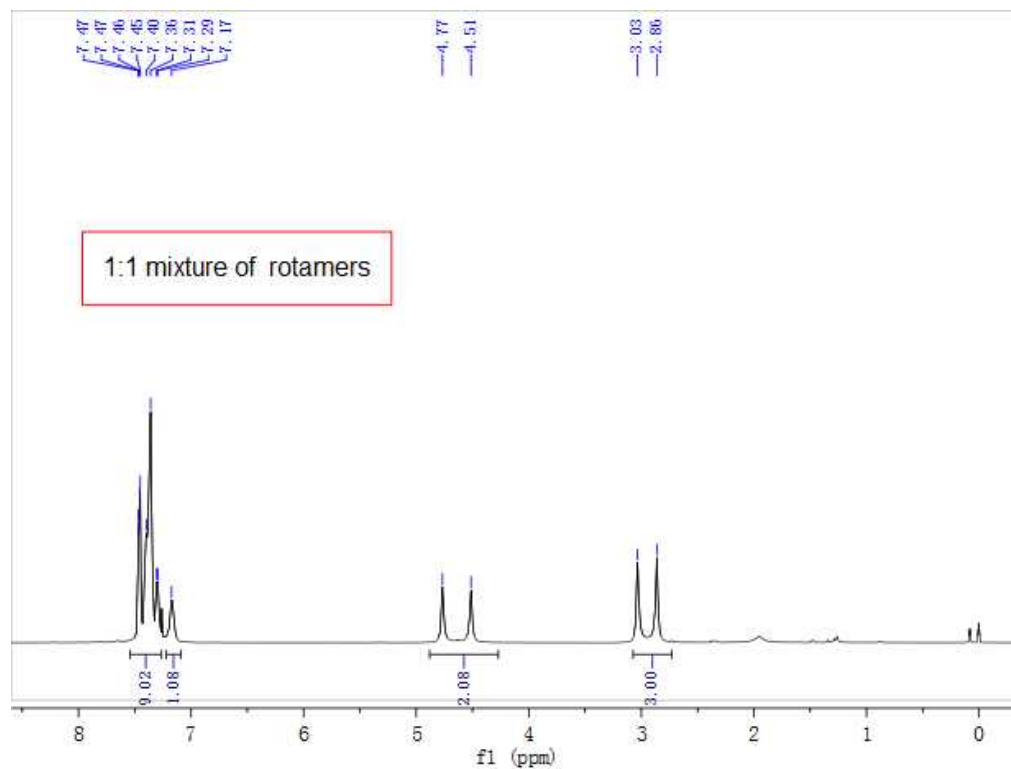
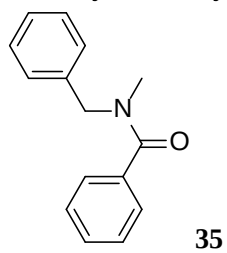
N-Phenylbenzamide (CAS: 93-98-1)



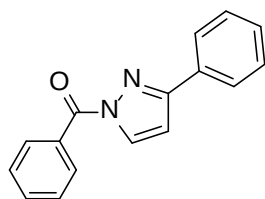
34



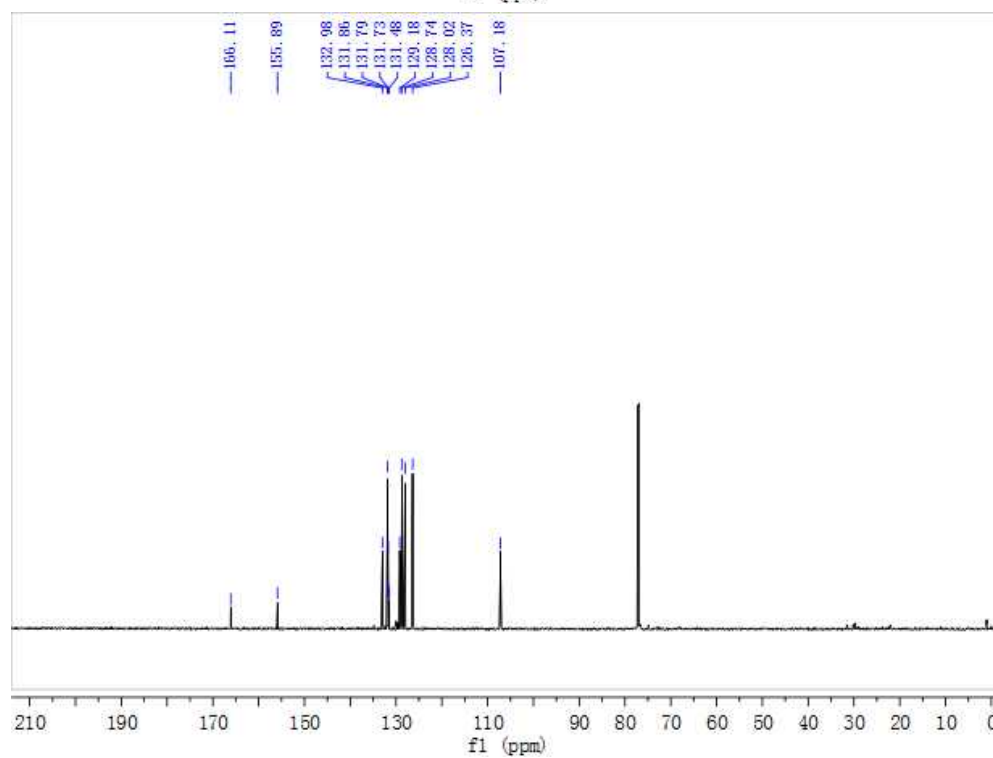
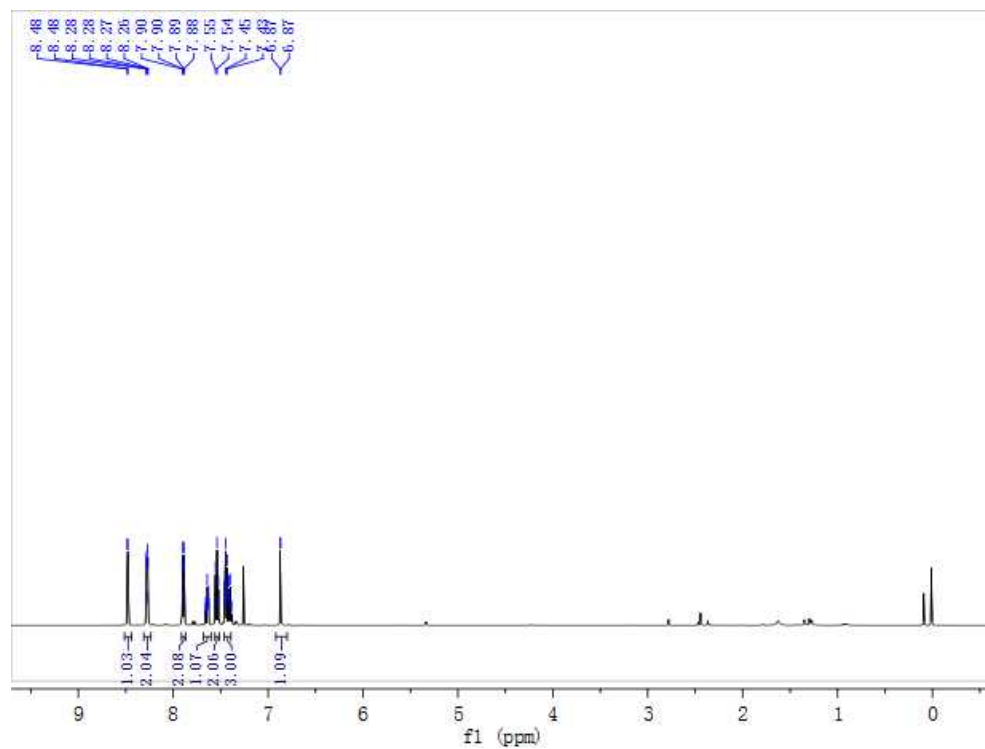
N-Benzyl-N-methylbenzamide (CAS: 61802-83-3)



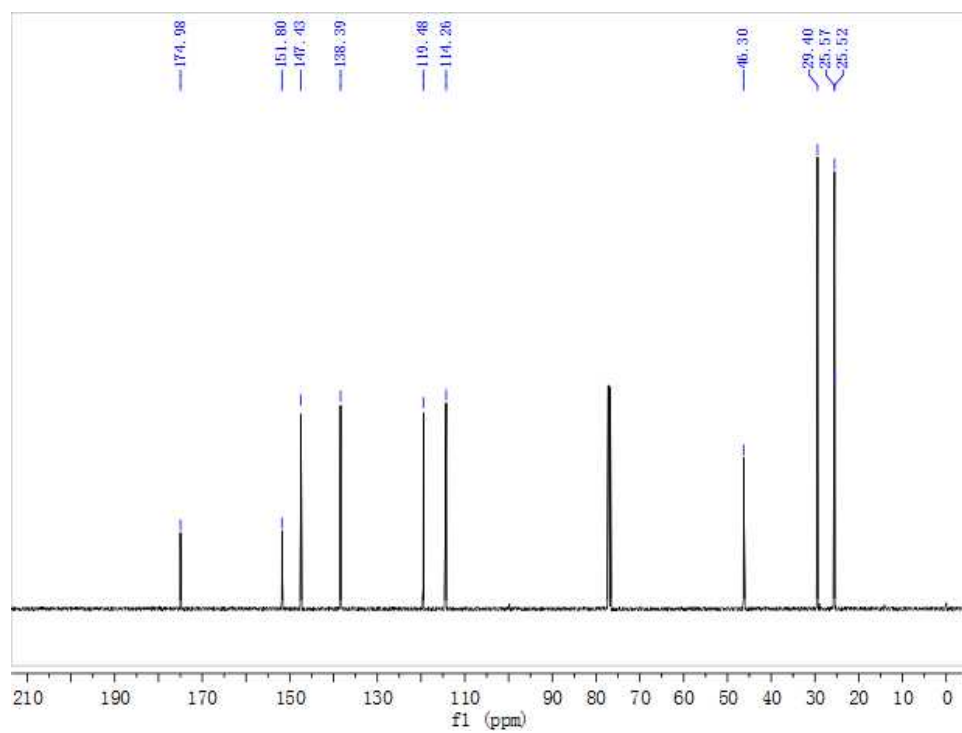
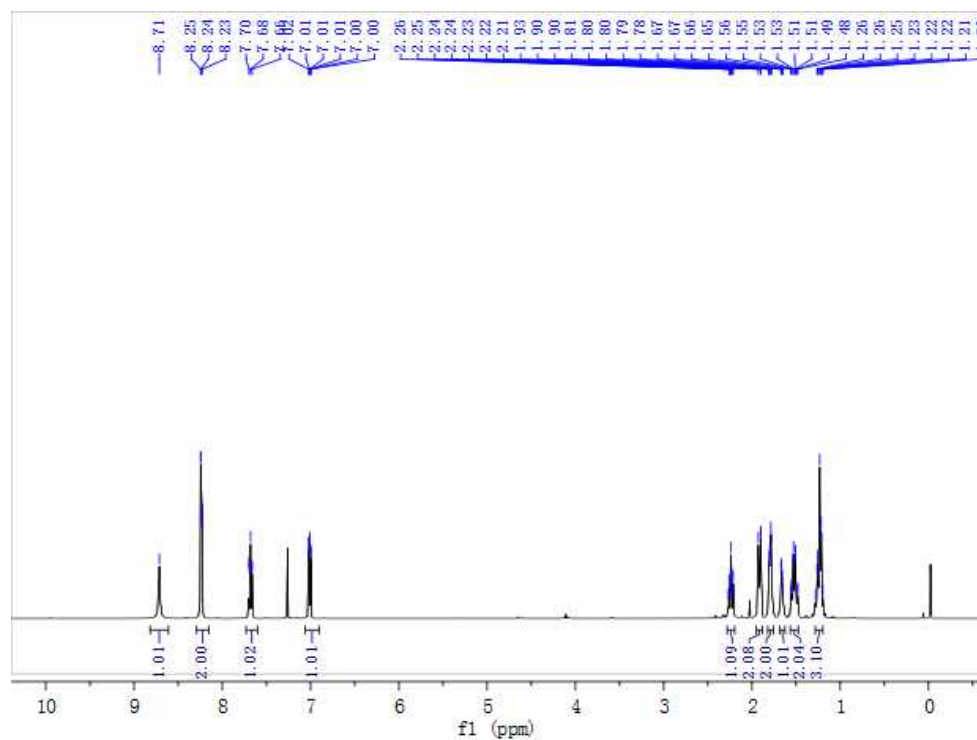
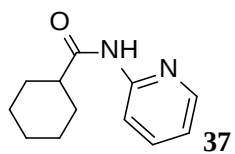
Phenyl(3-phenyl-1H-pyrazol-1-yl)methanone (CAS: 126382-89-6)



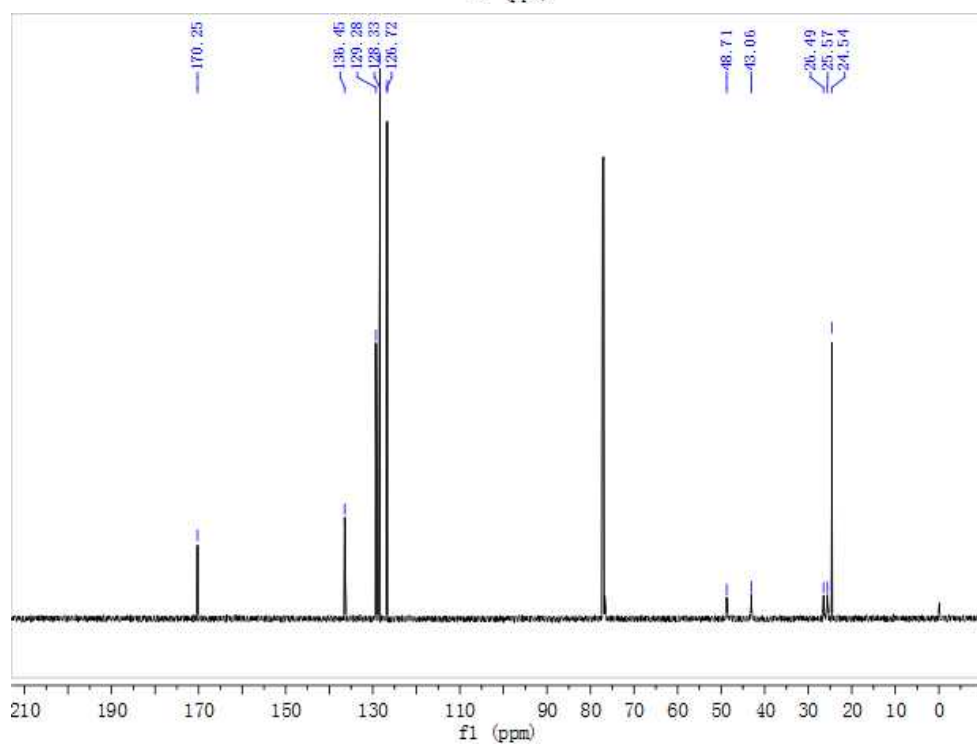
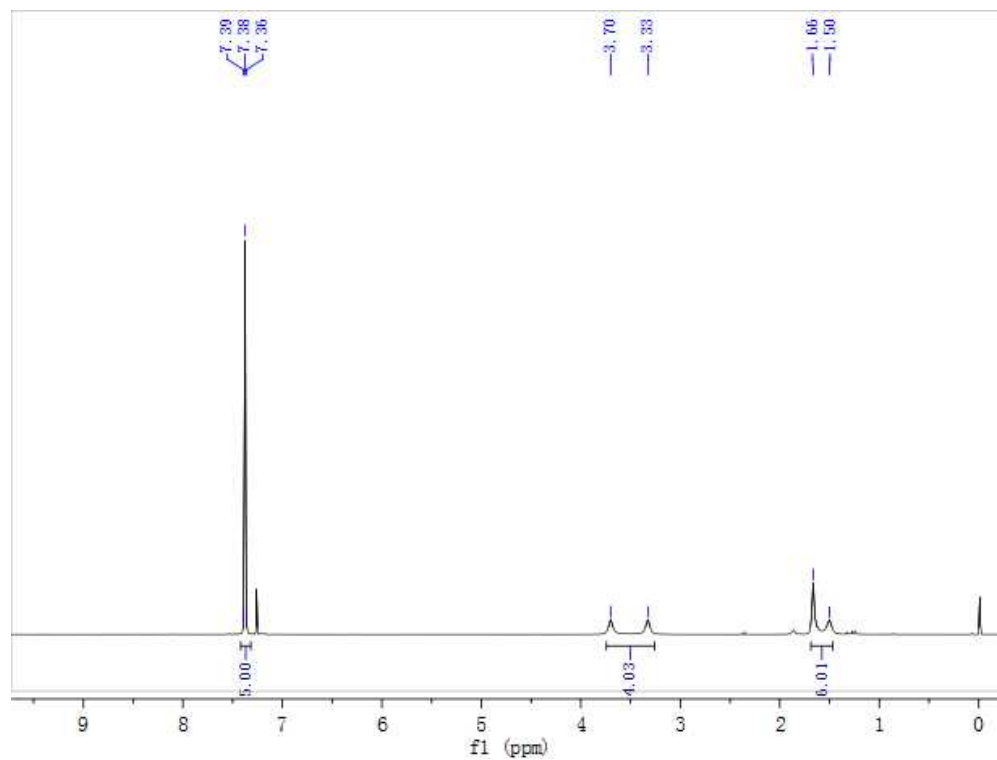
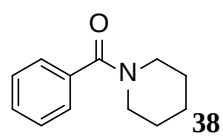
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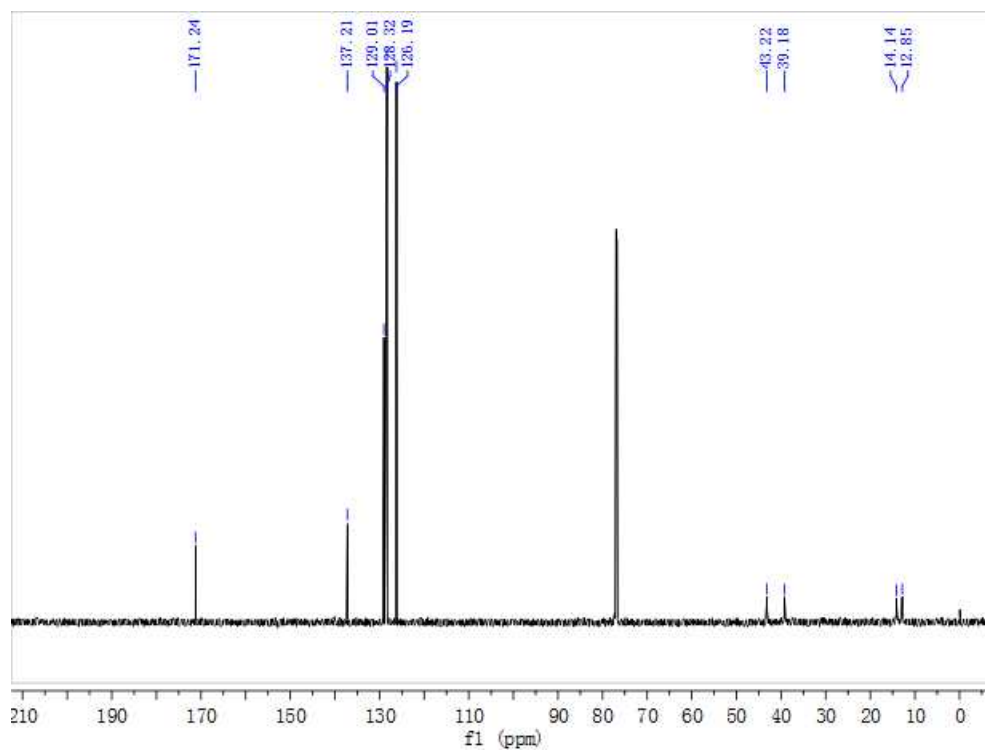
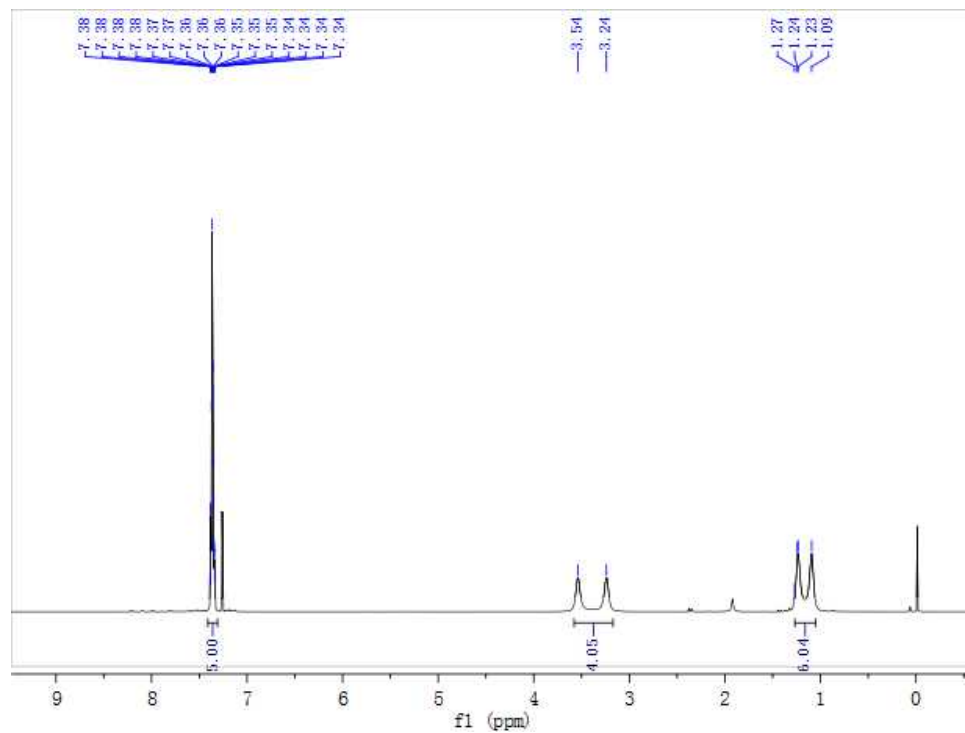
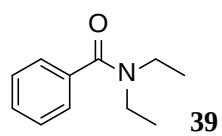
***N*-(Pyridin-2-yl)cyclohexanecarboxamide (CAS: 68134-77-0)**



Phenyl(piperidin-1-yl)methanone (CAS: 776-75-0)



***N,N*-Diethylbenzamide (CAS: 1696-17-9)**



Methyl benzoyl-L-leucinate (CAS: 3005-60-5)

