Electronic Supplementary Material (ESI) for Organic & Biomolecular Chemistry.

Electronic Supplementary Information

Visible-light-induced Aerobic Epoxidation with Vitamin B2-based Photocatalyst

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1. General Information

^{1}H-NMR, ^{13}C-NMR and ^{19}F-NMR were recorded on a BRUKER AVANCE at 500, 126 and 471 MHz, respectively. Chemical shifts (δ) were reported referenced to an internal tetramethylsilane standard or the $CDCl_3$ residual peak (δ 7.26) for ^{1}H-NMR. Chemical shifts of ^{13}C-NMR were reported relative to $CDCl_3$ (δ 77.0). Data were reported in the following order: chemical shift (δ) in ppm; multiplicities were indicated s (singlet), bs (broad singlet), d (doublet), t (triplet), m (multiplet); coupling constants (J) were in Hz. MS spectra were analyzed by Agilent G6460A LC-MS (ESI) and Agilent 7890/7000D GC-MS (EI, 70eV). UV-vis spectra were collected with an Agilent Cary 8454 diode array spectrophotometer. Analytical thin-layer chromatography (TLC) was performed on silica gel and visualized with UV light. Purification of reaction products was carried out by flash chromatography on silica gel (200−300 mesh). The photocatalytic reactions were performed with a multi-station reactor purchased from Shanghai Taitan Science Co. Ltd. (China). And the temperature and irradiation power could be set and well-controlled by the reactor (model: PR-6, see pictures below). The substrates could be easily prepared with reported methods by the reaction of corresponding ketones and aldehydes in EtOH-H_2O in the presence of NaOH. In addition, most of the solid substrates could be recrystallized with EtOAc/petroleum ether system. All the substrates and products (except **b15**, **b23** and **b24**) were known compounds and the characterizations were consistent with that in the literature. $^{18}O_2$ (98 atom% ^{18}O) was brought from Guangzhou Yigas Gases Co., Ltd. (P. R. China). Unless otherwise noted, other commercially available compounds and solvents with analytical grade were purchased from Energy Chemical Co., Ltd. (P. R. China) and used without further purification.

2. Experimental Section

Typical experimental procedure of photocatalytic epoxidation. In a 10 mL of Schlenk tube equipped with a magnetic stirring bar, the substrates (**a1-a26**, 0.25 mmol), RFT (3 mol%), DBU (2.5 equiv.) and MeOH (2 mL) were added. Having been charged with an air balloon (atmospheric pressure), then the reaction tube was irradiated under a 2 W blue LED (450 nm) with continuous stirring at room temperature for 3 h. After completion, the reaction mixture was purified by chromatography column on silica gel with EtOAc/petroleum ether to get the desired products.

Procedure of sunlight-driven and gram-scale reaction. The substrates **a1** (5.0 mmol, 1.04 g), RFT (5 mol%: 3 mol% for the 1st day and 2 mol% for the 2nd day), DBU (3.5 equiv.: 2.5 equiv. for the 1st day and 1.0 equiv. for the 2nd day) and MeOH (45 mL) were added into a round bottom flask equipped with a magnetic stirring bar. Then the reaction mixture was open to air and irradiated by sunlight with continuous stirring at room temperature for ca. 2 daytimes. After completion, the reaction mixture was purified by chromatography column on silica gel with EtOAc/petroleum ether to get the desired products (0.639 g, 57% yield).

Note: In the case of large-scale experiment, the catalyst and organic base were firstly added to the reaction mixture just as the same ratio as the small-scale experiment. After one day reaction, certain amounts of substrate were still there and not converted by TLC. Therefore, another batch of catalyst and organic base were added to the reaction, and the conversion looked much better after the second day.

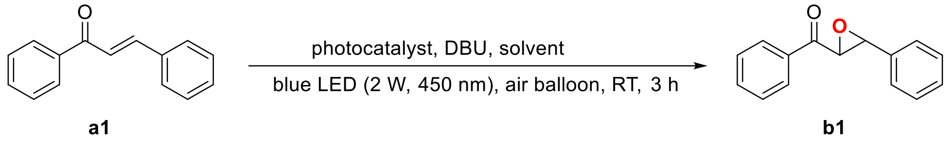
Procedure of control experiments:

(a) Blank experiments: Following the above typical procedure of photocatalytic reaction, the blank experiments were carried out in the absence of RF, base, light, or dioxygen, respectively. After the defined reaction time, the same work-up of typical reaction was conducted to detect or isolate the desired products.

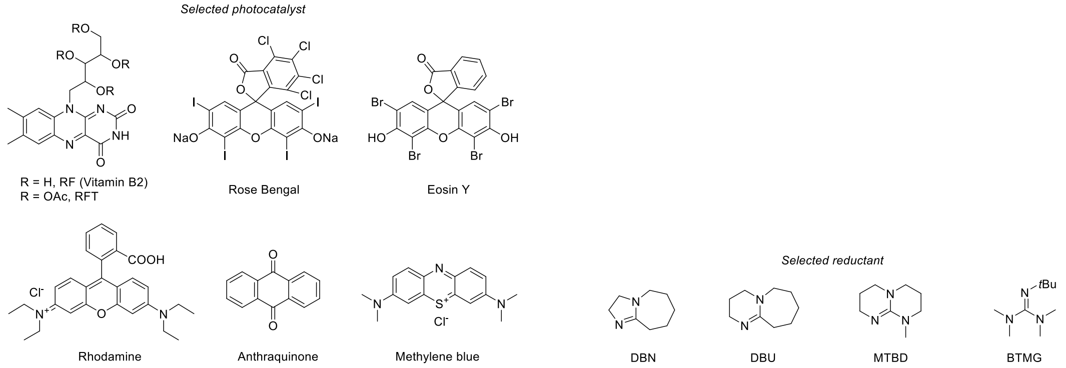
(b) With additives: Following the typical procedure of photocatalytic reaction, excess amounts of TEMPO, TEMP, DABCO or tBuBnNO were additionally added to the reaction mixture, respectively. After the defined time, the same work-up of typical reaction was conducted to detect or isolate the products.

(c) ^{18}O-isotopical labelling experiment: Following the typical procedure of photocatalytic reaction, on one hand, the Schlenk tube was charged with ^{18}O-dioxygen rather than air. After the defined time, the reaction mixture was diluted with anhydrous MeCN and injected to mass spectrometer for mass analysis of the epoxides.

3. **Table S1.** The supplementary data for the optimization of reaction conditions.

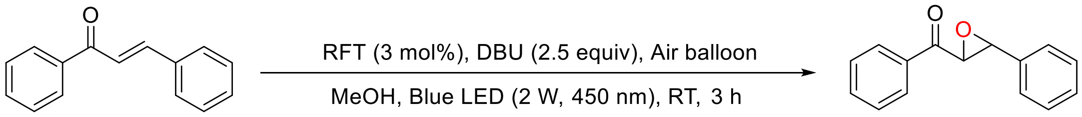


entry	photocatalysts (mol%)	DBU (equiv.)	solvent	yield (%)
1	RF (5%)	DBU (2.0)	MeOH	60
2	RF (5%)	DBU (2.5)	MeOH	74
3	RF (5%)	DBU (3.0)	MeOH	70
4	RF (3%)	DBU (2.5)	MeOH	73
5	RF (1%)	DBU (2.5)	MeOH	42
6	**RFT (3%)**	DBU **(2.5)**	**MeOH**	**75**
7	RFT (3%)	DBU (2.5)	EtOH	68
8	RFT (3%)	DBU (2.5)	TFE	10
9	RFT (3%)	DBU (2.5)	MeCN	45
10	RFT (3%)	DBU (2.5)	EtOAc	trace
11	RFT (3%)	DBU (2.5)	CH_2Cl_2	trace
12	RFT (3%)	DBU (2.5)	THF	24
13	RFT (3%)	DBU (2.5)	dioxane	12



^{a}Unless otherwise stated, the reaction conditions were: **a1** (0.25 mmol), photocatalyst (1-5 mol%), reductant/base (2.0-3.0 equiv.) and solvent (2 mL) were mixed and stirred in a glass tube equipped with an air balloon under visible-light irradiation at room temperature. The products were shown with isolated yields.

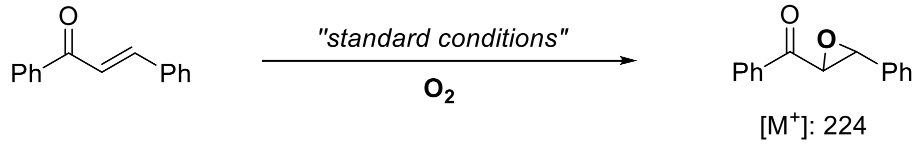
4. **Table S2.** Results of blank experiments.

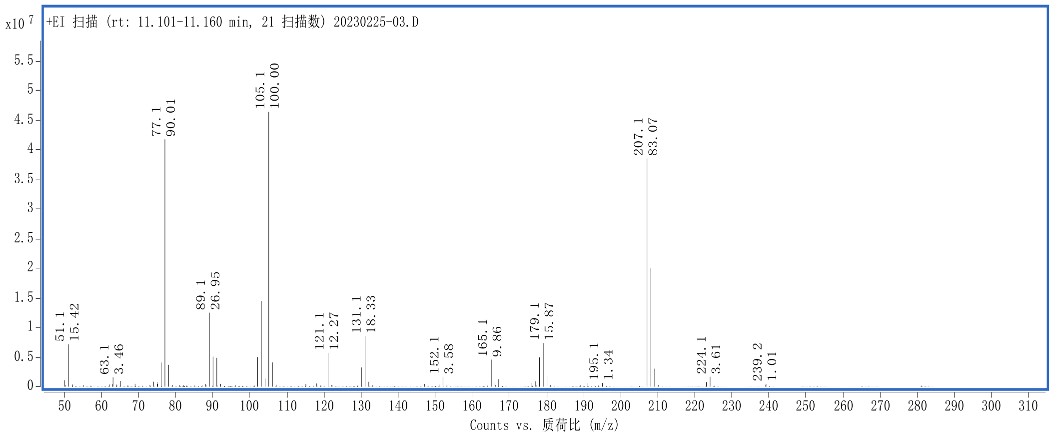


entry	RFT	DBU	air	light	remarks
1	no	yes	yes	yes	no reaction
2	yes	no	yes	yes	no reaction
3	yes	yes	no	yes	no reaction
4	yes	yes	yes	no	no reaction

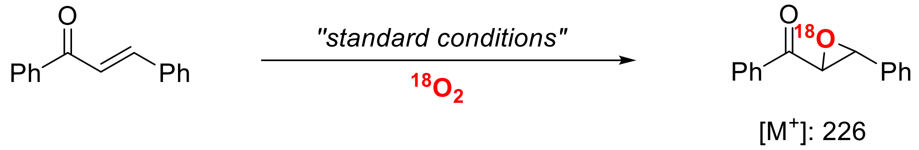
5. GC-MS Spectra of ^{18}O-Labelling Experiment

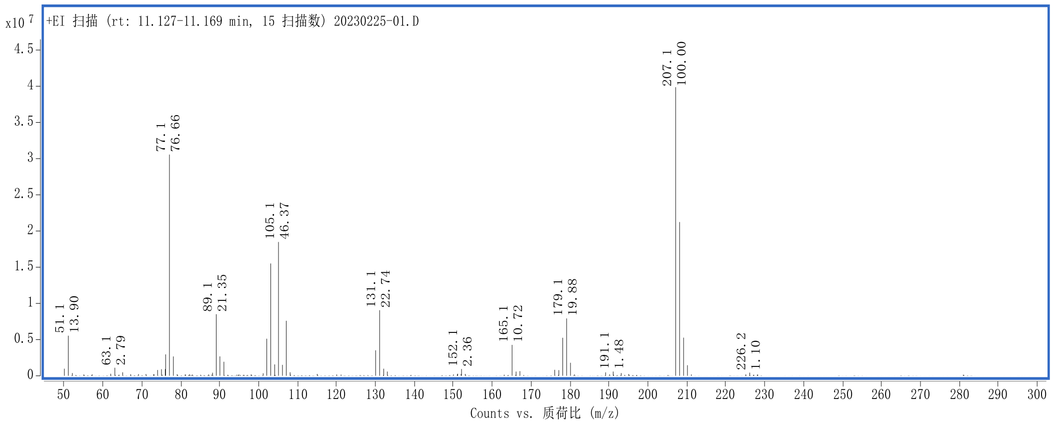
(a)





(b)





6. Characterization data of isolated products

phenyl(3-phenyloxiran-2-yl)methanone (**b1**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (42.5 mg, 76% yield). ^{1}H NMR (500 MHz, Chloroform-d) δ 8.04–7.99 (m, 2H), 7.65–7.60 (m, 1H), 7.49 (t, *J* = 7.8 Hz, 2H), 7.39 (tdd, *J* = 7.3, 4.6, 1.6 Hz, 5H), 4.30 (d, *J* = 1.8 Hz, 1H), 4.08 (d, *J* = 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-d) δ 193.1, 135.5, 134.0, 129.0, 128.9, 128.8, 128.3, 125.8, 61.0, 59.4.

(4-methoxyphenyl)(3-phenyloxiran-2-yl)methanone (**b2**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Orange solid (52.1 mg, 82% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.81 (d, *J* = 7.7 Hz, 2H), 7.47 (t, *J* = 7.4 Hz, 1H), 7.31 (t, *J* = 7.7 Hz, 2H), 7.22 (d, *J* = 8.3 Hz, 2H), 6.83 (d, *J* = 8.3 Hz, 2H), 4.58–4.53 (m, 1H), 3.84–3.80 (m, 1H), 3.78 (s, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 199.21, 158.72, 135.68, 133.59, 133.38, 128.85, 128.56, 128.52, 114.05, 55.29, 48.05, 47.81.

(3-phenyloxiran-2-yl)(p-tolyl)methanone (**b3**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (51.2 mg, 86% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.94–7.89 (m, 2H), 7.38 (tdd, *J* = 8.1, 6.3, 3.4 Hz, 5H), 7.27 (d, *J* = 8.0 Hz, 2H), 4.27 (d, *J* = 1.9 Hz, 1H), 4.06 (d, *J* = 1.9 Hz, 1H), 2.42 (s, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.59, 145.09, 135.64, 133.06, 129.59, 129.02, 128.77, 128.50, 125.82, 60.94, 59.33, 21.82.

(4-fluorophenyl)(3-phenyloxiran-2-yl)methanone (**b4**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (36.3 mg, 60% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.10–8.03 (m, 2H), 7.44–7.34 (m, 5H), 7.19–7.12 (m, 2H), 4.24 (d, J = 2.0 Hz, 1H), 4.07 (d, J = 1.9 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 191.59, 167.25 (d, C-F, $1J_{\text{C-F}}$ = 258.3 Hz), 135.35, 131.91 (d, C-F, $3J_{\text{C-F}}$ = 2.5 Hz), 131.18 (d, C-F, $3J_{\text{C-F}}$ = 10.1 Hz), 129.13, 128.81, 125.79, 116.13 (d, C-F, $2J_{\text{C-F}}$ = 22.7 Hz), 61.08, 59.30. ^{19}F NMR (471 MHz, Chloroform-*d*) δ -103.09.

(4-chlorophenyl)(3-phenyloxiran-2-yl)methanone (**b5**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (41.2 mg, 64% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.00–7.94 (m, 2H), 7.49–7.44 (m, 2H), 7.43–7.34 (m, 5H), 4.23 (d, J = 1.8 Hz, 1H), 4.07 (d, J = 1.9 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.06, 140.62, 135.26, 133.71, 129.82, 129.26, 129.17, 128.83, 125.79, 61.10, 59.40.

(3-phenyloxiran-2-yl)(4-(trifluoromethyl)phenyl)methanone (**b6**)[3]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow solid (34.3 mg, 47% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.14 (d, J = 8.1 Hz, 2H), 7.76 (d, J = 8.1 Hz, 2H), 7.45–7.35 (m, 5H), 4.26 (t, *J* = 1.6 Hz, 1H), 4.10 (t, *J* = 1.5 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.56, 137.97, 135.15 (q, C-F, 2J_{C-F} = 32.8 Hz), 129.28, 128.84 (d, C-F, 3J_{C-F} = 8.8 Hz), 125.93 (q, C-F, 3J_{C-F} = 3.8 Hz), 125.79, 123.43 (q, C-F, 1J_{C-F} = 273.4 Hz), 61.32, 59.52. ^{19}F NMR (471 MHz, Chloroform-*d*) δ -63.29.

benzo[d][1,3]dioxol-5-yl(3-phenyloxiran-2-yl)methanone (**b7**)[4]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow solid (46.9 mg, 70% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.63 (dd, *J* = 8.2, 1.8 Hz, 1H), 7.49 (d, *J* = 1.7 Hz, 1H), 7.43–7.34 (m, 5H), 6.86 (d, *J* = 8.3 Hz, 1H), 6.06 (s, 2H), 4.21 (d, *J* = 1.9 Hz, 1H), 4.06 (d, *J* = 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 190.90, 152.63, 148.49, 135.58, 130.34, 129.02, 128.77, 125.79, 125.05, 108.18, 107.98, 102.06, 60.90, 59.23.

naphthalen-2-yl(3-phenyloxiran-2-yl)methanone (**b8**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Orange solid (36.3 mg, 53% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.50–8.45 (m, 1H), 7.96 (dd, *J* = 8.7, 1.8 Hz, 1H), 7.88–7.77 (m, 3H), 7.54 (t, *J* = 7.4 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.37–7.29 (m, 5H), 4.35 (d, *J* = 1.9 Hz, 1H), 4.07 (d, *J* = 1.9 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.90, 135.88, 135.55, 132.82, 132.35, 130.40, 129.67, 129.02, 128.99, 128.82, 128.76, 127.83, 127.03, 125.82, 123.62, 61.03, 59.45.

furan-2-yl(3-phenyloxiran-2-yl)methanone (**b9**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Brown liquid (18.2 mg, 34% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.67 (d, *J* = 1.6 Hz, 1H), 7.46 (d, *J* = 3.7 Hz, 1H), 7.42–7.32 (m, 5H), 6.60 (dd, *J* = 3.7, 1.7 Hz, 1H), 4.18–4.12 (m, 2H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 182.03, 151.19, 147.60, 135.33, 129.03, 128.69, 125.79, 119.49, 112.63, 60.63, 59.56.

(3-(4-methoxyphenyl)oxiran-2-yl)(phenyl)methanone (**b10**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (33.0 mg, 52% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.92 (d, *J* = 8.5 Hz, 2H), 7.29 (q, *J* = 5.9 Hz, 5H), 6.86 (d, *J* = 8.5 Hz, 2H), 4.19–4.13 (m, 1H), 4.01–3.95 (m, 1H), 3.78 (s, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 191.34, 164.25, 135.72, 130.77, 128.97, 128.75, 128.61, 125.81, 114.12, 60.88, 59.18, 55.57.

phenyl(3-(p-tolyl)oxiran-2-yl)methanone (**b11**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (32.3 mg, 54% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.03–7.98 (m, 2H), 7.64–7.58 (m, 1H), 7.48 (t, *J* = 7.9 Hz, 2H), 7.26 (d, *J* = 8.4 Hz, 2H), 7.21 (d, *J* = 8.0 Hz, 2H), 4.29 (d, *J* = 1.9 Hz, 1H), 4.04 (d, *J* = 1.8 Hz, 1H), 2.38 (s, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 193.16, 139.02, 135.46, 133.90, 132.42, 129.42, 128.82, 128.29, 125.74, 61.03, 59.42, 21.24.

(3-(4-fluorophenyl)oxiran-2-yl)(phenyl)methanone (**b12**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow solid (28.4 mg, 47% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.01 (dd, *J* = 8.1, 1.4 Hz, 2H), 7.66–7.60 (m, 1H), 7.50 (t, *J* = 7.8 Hz, 2H), 7.38–7.32 (m, 2H), 7.09 (t, *J* = 8.6 Hz, 2H), 4.26 (d, *J* = 1.9 Hz, 1H), 4.07 (d, *J* = 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.88, 163.20 (d, C-F, $1J_{C\text{-}F}$ = 248.2 Hz), 135.43, 134.07, 131.29 (d, C-F, $3J_{C\text{-}F}$ = 3.8 Hz), 128.93, 128.36, 127.60 (d, C-F, $3J_{C\text{-}F}$ = 8.8 Hz), 115.87 (d, C-F, $2J_{C\text{-}F}$ = 22.7 Hz), 60.97, 58.79. ^{19}F NMR (471 MHz, Chloroform-*d*) δ -112.18.

(3-(4-chlorophenyl)oxiran-2-yl)(phenyl)methanone (**b13**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow solid (31.0 mg, 48% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.00 (d, *J* = 7.7 Hz, 2H), 7.63 (t, *J* = 7.4 Hz, 1H), 7.50 (t, *J* = 7.6 Hz, 2H), 7.41–7.28 (m, 4H), 4.28–4.22 (m, 1H), 4.09–4.03 (m, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.68, 135.33, 134.92, 134.07, 134.01, 129.00, 128.89, 128.32, 127.09, 60.90, 58.65.

phenyl(3-(4-(trifluoromethyl)phenyl)oxiran-2-yl)methanone (**b14**)[5]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (49.6 mg, 68% yield). ^{1}H NMR (500

MHz, Chloroform-*d*) δ 8.04–7.98 (m, 2H), 7.70–7.61 (m, 3H), 7.50 (dt, *J* = 7.9, 3.6 Hz, 4H), 4.27 (d, *J* = 1.8 Hz, 1H), 4.15 (d, *J* = 1.7 Hz, 1H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 192.44, 139.61, 135.32, 134.20, 131.20 (q, C-F, $2J_{C\text{-}F}$ = 32.8 Hz), 128.97, 128.39, 126.12, 125.80 (q, C-F, $3J_{C\text{-}F}$ = 3.8 Hz), 123.89 (q, C-F, $1J_{C\text{-}F}$ = 272.2 Hz), 60.90, 58.44. ^{19}F NMR (471 MHz, Chloroform-*d*) δ -62.68.

(3-(2,6-dimethylphenyl)oxiran-2-yl)(phenyl)methanone (**b15**): Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (30.9 mg, 49% yield). ^1H NMR (500 MHz, Chloroform-*d*) δ 8.15–8.10 (m, 2H), 7.67–7.62 (m, 1H), 7.52 (t, *J* = 7.8 Hz, 2H), 7.14 (t, *J* = 7.6 Hz, 1H), 7.01 (d, *J* = 7.5 Hz, 2H), 4.25–4.20 (m, 2H), 2.39 (s, 6H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 194.64, 137.05, 135.69, 134.04, 132.25, 128.92, 128.59, 128.22, 59.02, 57.74, 19.93. HR-MS $[M+Na]^+$ Calcd.: 275.1048, Found: 275.1036.

(3-(2,6-dichlorophenyl)oxiran-2-yl)(phenyl)methanone (**b16**)[6]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (33.6 mg, 46% yield). ^1H NMR (500 MHz, Chloroform-*d*) δ 8.17 (dd, *J* = 8.2, 1.5 Hz, 2H), 7.66 (td, *J* = 7.3, 1.3 Hz, 1H), 7.54 (t, *J* = 7.8 Hz, 2H), 7.34 (d, *J* = 8.2 Hz, 2H), 7.28–7.25 (m, 1H), 4.52 (d, *J* = 1.9 Hz, 1H), 4.29 (d, *J* = 1.9 Hz, 1H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 193.42, 135.72, 134.09, 131.50, 130.29, 128.87, 128.73, 128.56, 57.27, 57.24.

(3-(naphthalen-2-yl)oxiran-2-yl)(phenyl)methanone (**b17**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Yellow solid (47.7 mg, 66% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.05–7.99 (m, 2H), 7.86 (ddd, *J* = 14.7, 6.7, 3.5 Hz, 4H), 7.64–7.59 (m, 1H), 7.54–7.46 (m, 4H), 7.44–7.40 (m, 1H), 4.40 (d, *J* = 1.8 Hz, 1H), 4.24 (d, *J* = 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 193.03, 135.51, 134.02, 133.67, 133.10, 132.90, 128.92, 128.82, 128.38, 127.92, 127.87, 126.72, 126.63, 125.89, 122.45, 61.22, 59.68.

p-tolyl(3-(*p*-tolyl)oxiran-2-yl)methanone (**b18**)[3]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (44.1 mg, 70% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.90 (d, *J* = 8.2 Hz, 2H), 7.26 (dd, *J* = 8.2, 6.5 Hz, 4H), 7.20 (d, *J* = 7.9 Hz, 2H), 4.27 (d, *J* = 2.0 Hz, 1H), 4.02 (d, *J* = 2.0 Hz, 1H), 2.41 (s, 3H), 2.37 (s, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 192.72, 145.01, 138.99, 133.10, 132.62, 129.56, 129.45, 128.47, 125.80, 60.99, 59.39, 21.80, 21.29.

(4-chlorophenyl)(3-(4-chlorophenyl)oxiran-2-yl)methanone (**b19**)[7]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (42.3 mg, 58% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.99–7.93 (m, 2H), 7.50–7.45 (m, 2H), 7.41–7.36 (m, 2H), 7.30 (d, *J* = 8.5 Hz, 2H), 4.18 (d, *J* = 1.8 Hz, 1H), 4.06 (d, *J* = 1.8 Hz, 1H). ^{13}C{^{1}H} NMR

(126 MHz, Chloroform-*d*) δ 191.71, 140.74, 135.09, 133.82, 133.62, 129.81, 129.30, 129.08, 127.11, 61.00, 58.66.

(3-methyloxiran-2-yl)(phenyl)methanone (**b20**)[8]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow liquid (14.6 mg, 36% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.04–8.00 (m, 2H), 7.65–7.60 (m, 1H), 7.51 (t, *J* = 7.8 Hz, 2H), 3.98 (d, *J* = 1.9 Hz, 1H), 3.23 (qd, *J* = 5.1, 2.0 Hz, 1H), 1.53 (d, *J* = 5.1 Hz, 3H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 194.62, 135.61, 133.80, 128.80, 128.30, 58.36, 55.92, 17.64.

cyclohexyl(3-phenyloxiran-2-yl)methanone (**b21**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Colorless liquid (19.6 mg, 34% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.39–7.32 (m, 3H), 7.28 (dd, *J* = 7.7, 2.0 Hz, 2H), 3.91 (d, *J* = 1.8 Hz, 1H), 3.59 (d, *J* = 1.7 Hz, 1H), 2.57 (ddt, *J* = 11.4, 8.4, 3.3 Hz, 1H), 1.99–1.89 (m, 1H), 1.81 (tq, *J* = 10.1, 4.0, 3.6 Hz, 3H), 1.68 (ddd, *J* = 15.6, 7.7, 5.0 Hz, 1H), 1.54–1.42 (m, 1H), 1.35–1.17 (m, 4H).^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 207.87, 135.42, 128.93, 128.69, 125.68, 62.03, 58.39, 46.82, 28.35, 27.51, 25.71, 25.66, 25.28.

2,2-dimethyl-1-(3-phenyloxiran-2-yl)propan-1-one (**b22**)[1]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light yellow solid (29.6 mg, 58% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.41–7.35 (m, 3H), 7.31 (dt, *J* = 7.5, 1.5 Hz, 2H), 3.86 (q, *J* = 1.9 Hz, 2H), 1.24 (s, 9H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 208.12, 135.62, 128.90, 128.72, 125.62, 59.35, 59.14, 43.59, 25.72.

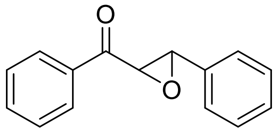
(4-(methylthio)phenyl)(3-phenyloxiran-2-yl)methanone (**b23**): Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). White solid (22.9 mg, 34% yield). ^1H NMR (500 MHz, Chloroform-*d*) δ 7.96–7.90 (m, 2H), 7.39 (dtdd, *J* = 9.6, 7.3, 5.1, 2.2 Hz, 5H), 7.30–7.25 (m, 2H), 4.24 (d, *J* = 1.8 Hz, 1H), 4.07 (d, *J* = 1.9 Hz, 1H), 2.52 (s, 3H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 191.93, 147.47, 135.58, 131.69, 129.03, 128.77, 128.74, 125.80, 125.08, 60.97, 59.29, 14.68. HR-MS $[M+Na]^+$ Calcd.: 293.0612, Found: 293.0610.

(4-(dimethylamino)phenyl)(3-phenyloxiran-2-yl)methanone (**b24**): Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Brown solid (21.4 mg, 32% yield). ^1H NMR (500 MHz, Chloroform-*d*) δ 7.97 (d, *J* = 8.3 Hz, 2H), 7.78 (d, *J* = 15.6 Hz, 1H), 7.63 (dd, *J* = 7.5, 1.8 Hz, 2H), 7.56 (d, *J* = 15.6 Hz, 1H), 7.40 (q, *J* = 6.6 Hz, 3H), 6.62 (d, *J* = 8.4 Hz, 2H), 3.48 (t, *J* = 1.5 Hz, 3H), 2.92 (s, 3H). $^{13}C\{^1H\}$ NMR (126 MHz, Chloroform-*d*) δ 187.97, 153.07, 142.77, 135.46, 131.09, 129.97, 128.85, 128.23, 127.30, 122.20, 111.40, 50.82, 30.18. HR-MS $[M+Na]^+$ Calcd.: 290.1157, Found: 290.1151.

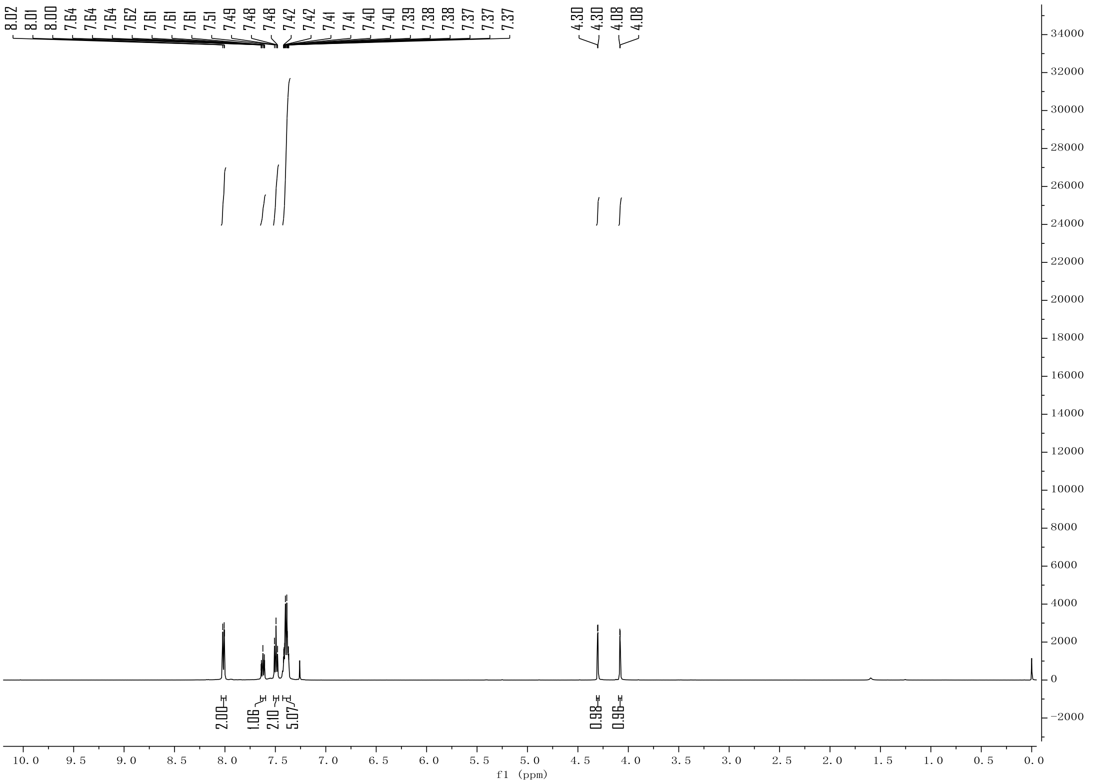
(2-hydroxyphenyl)(3-phenyloxiran-2-yl)methanone (**b25**)[9]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Light orange solid (41.4 mg, 69% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.93 (dd, *J* = 8.1, 1.7 Hz, 1H), 7.50 (dd, *J* = 13.3, 6.8 Hz, 3H), 7.43 (t, *J* = 7.5 Hz, 2H), 7.38 (t, *J* = 7.3 Hz, 1H), 7.05 (dt, *J* = 7.5, 3.1 Hz, 2H), 5.48 (dd, *J* = 13.4, 2.9 Hz, 1H), 3.08 (dd, *J* = 16.8, 13.3 Hz, 1H), 2.89 (dd, *J* = 16.8, 2.9 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 191.96, 161.57, 138.77, 136.22, 128.87, 128.80, 127.08, 126.18, 121.64, 120.96, 118.16, 79.62, 44.70.

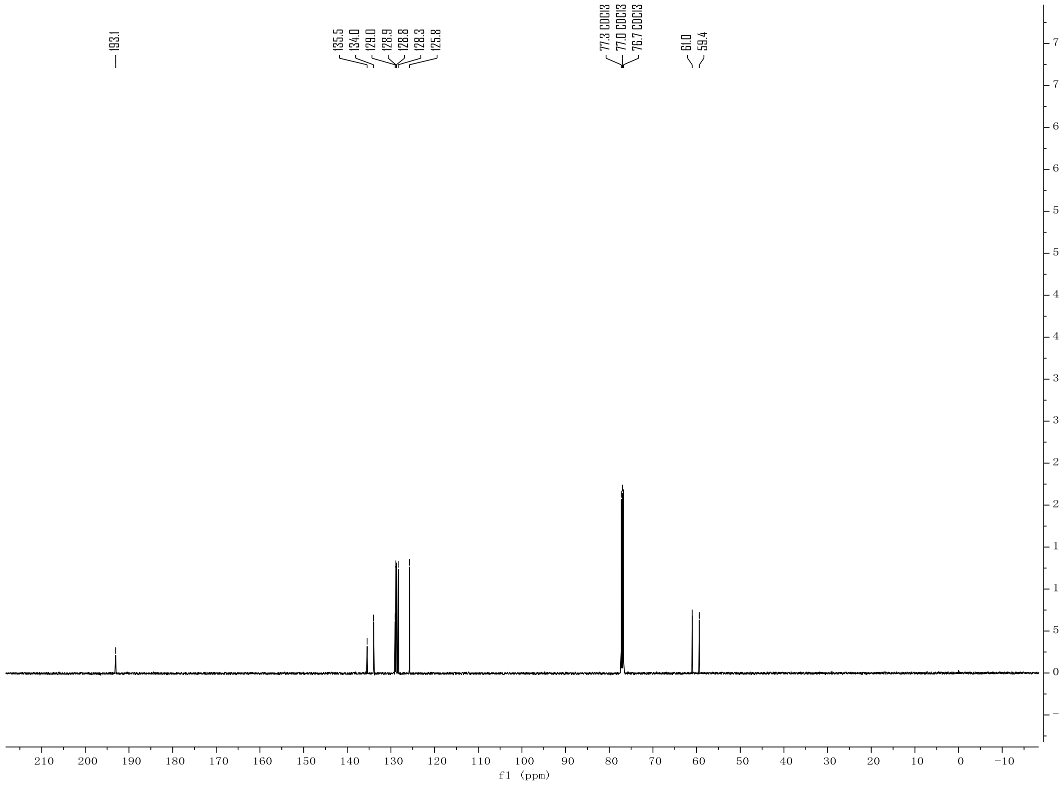
3'-phenyl-3,4-dihydro-1H-spiro[naphthalene-2,2'-oxiran]-1-one (**b26**)[2]: Following the general procedure, the title product was isolated by column chromatography (EtOAc/petroleum ether, v/v = 1: 79). Yellow solid (20.6 mg, 33% yield). ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.12 (dd, *J* = 7.9, 1.4 Hz, 1H), 7.52 (td, *J* = 7.5, 1.4 Hz, 1H), 7.42–7.33 (m, 6H), 7.22 (d, *J* = 7.6 Hz, 1H), 4.37 (s, 1H), 2.86–2.80 (m, 2H), 2.50–2.40 (m, 1H), 1.86 (dt, *J* = 13.5, 4.1 Hz, 1H). ^{13}C{^{1}H} NMR (126 MHz, Chloroform-*d*) δ 193.59, 143.33, 134.22, 134.11, 132.67, 128.72, 128.34, 128.31, 127.64, 126.98, 126.62, 64.31, 64.09, 27.33, 25.32.

7. ^{1}H, ^{13}C, and ^{19}F NMR Spectra of Isolated Products



phenyl(3-phenyloxiran-2-yl)methanone (**b1**)

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59.4
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6000
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5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
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180
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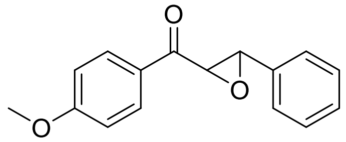
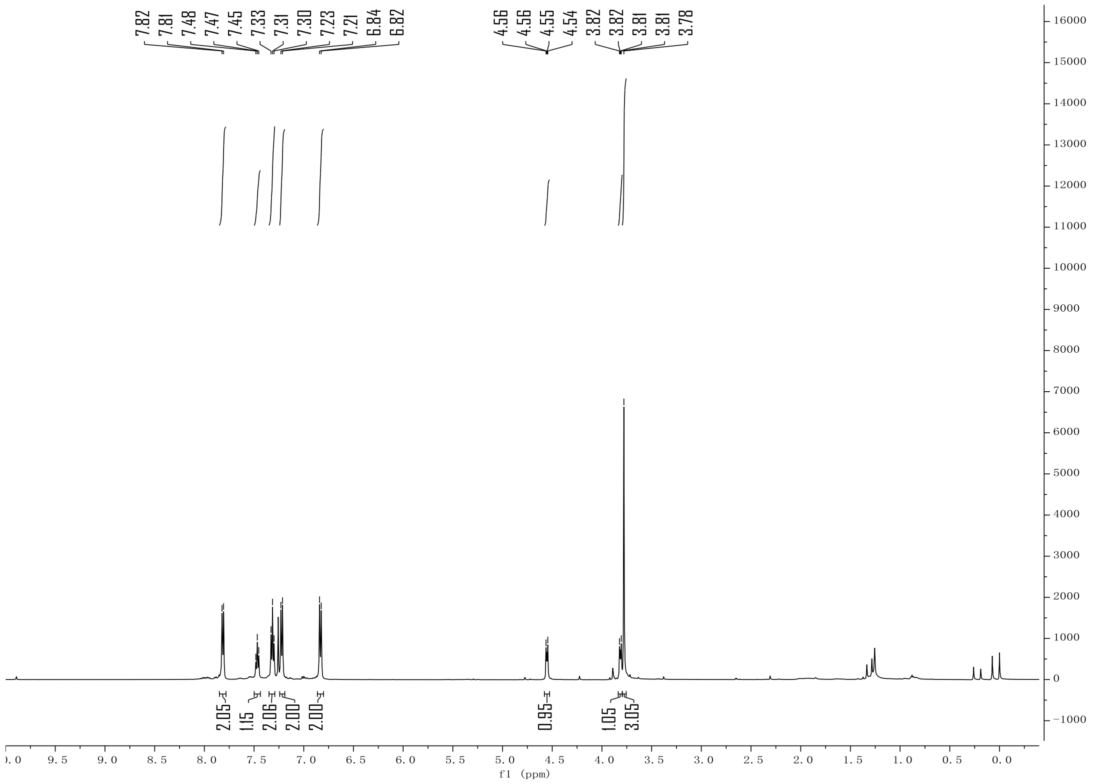
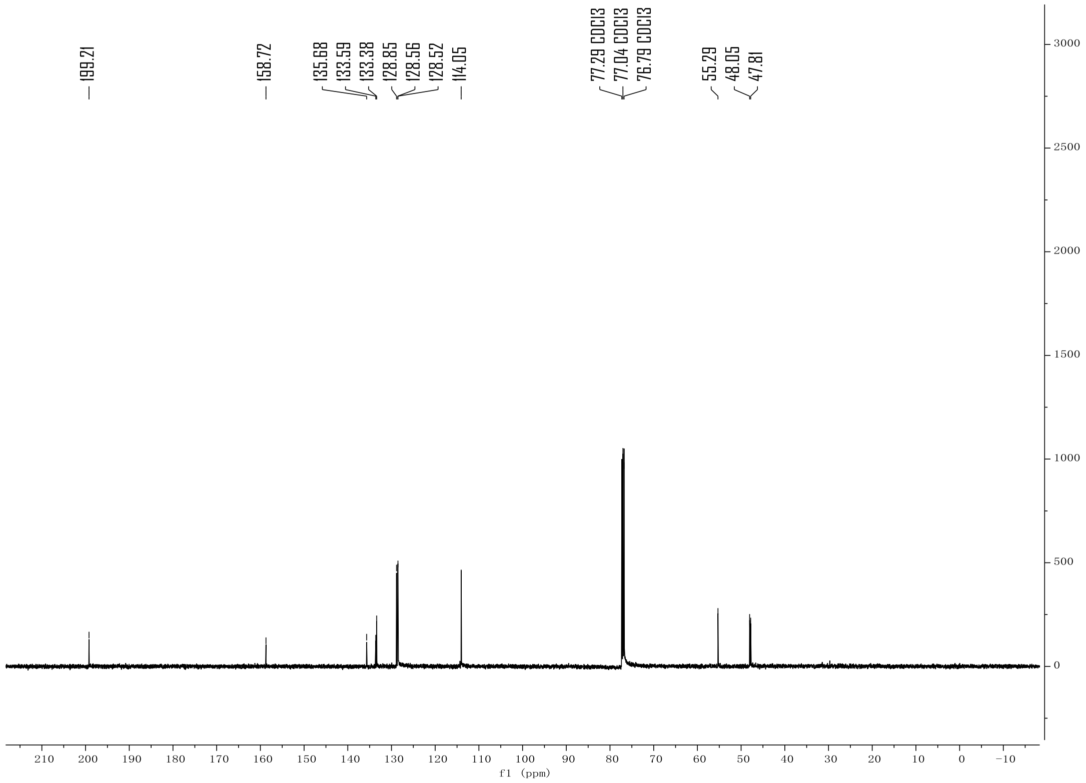
f1 (ppm)



(4-methoxyphenyl)(3-phenyloxiran-2-yl)methanone (**b2**)

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76.79 CDCl3
55.29
48.05
47.81
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1500
1000
500
0
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200
190
180
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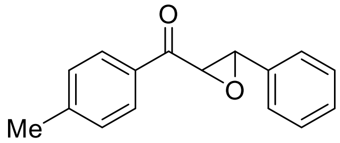
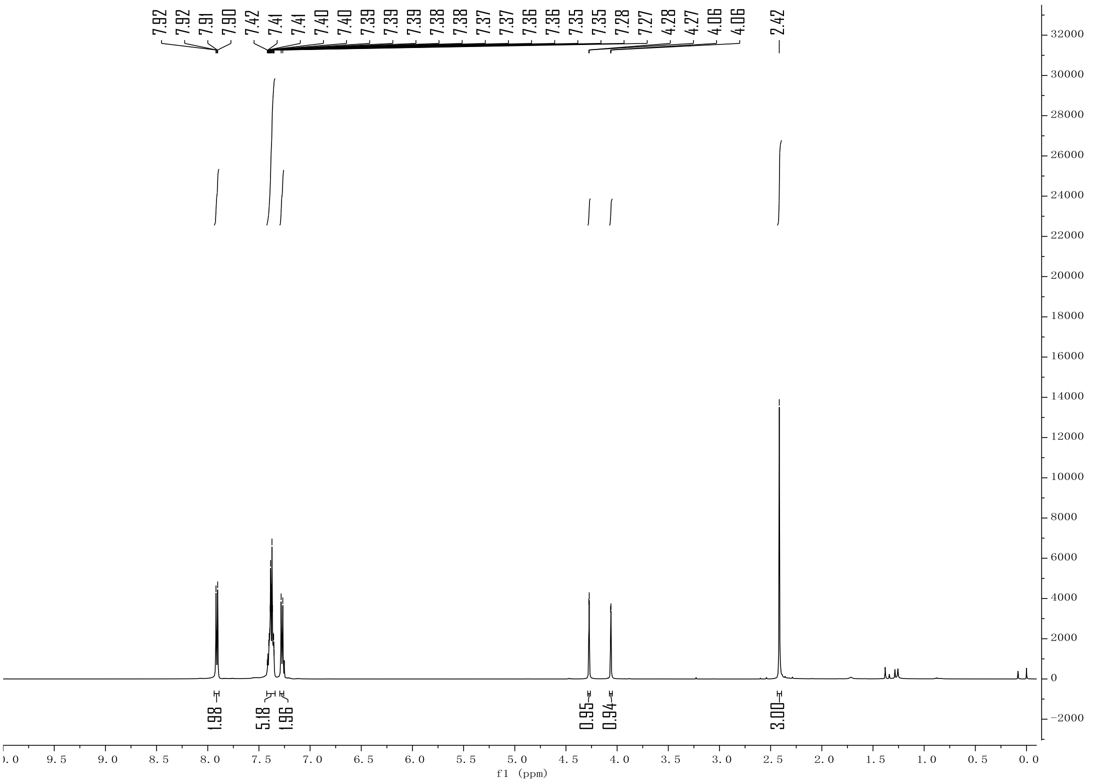
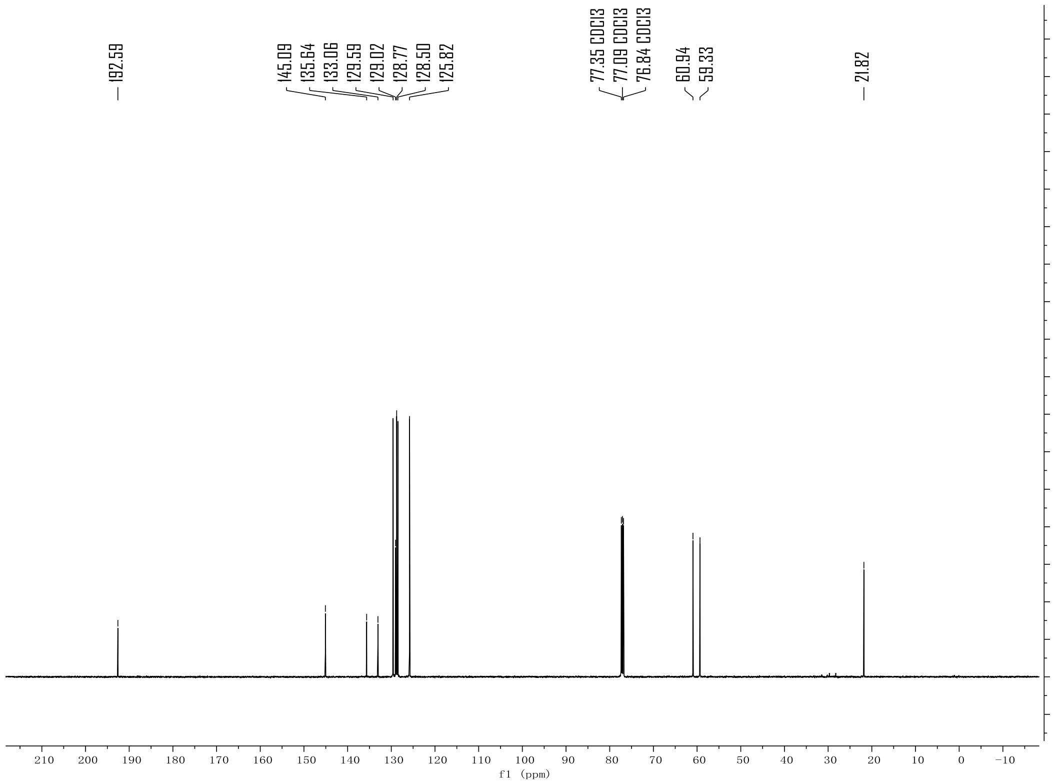
f1 (ppm)



(3-phenyloxiran-2-yl)(p-tolyl)methanone (**b3**)

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128.50
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76.84 CDCl3
60.94
59.33
21.82
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7000
6500
6000
5500
5000
4500
4000
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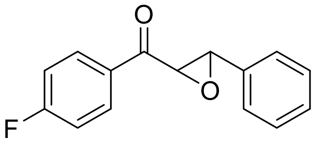
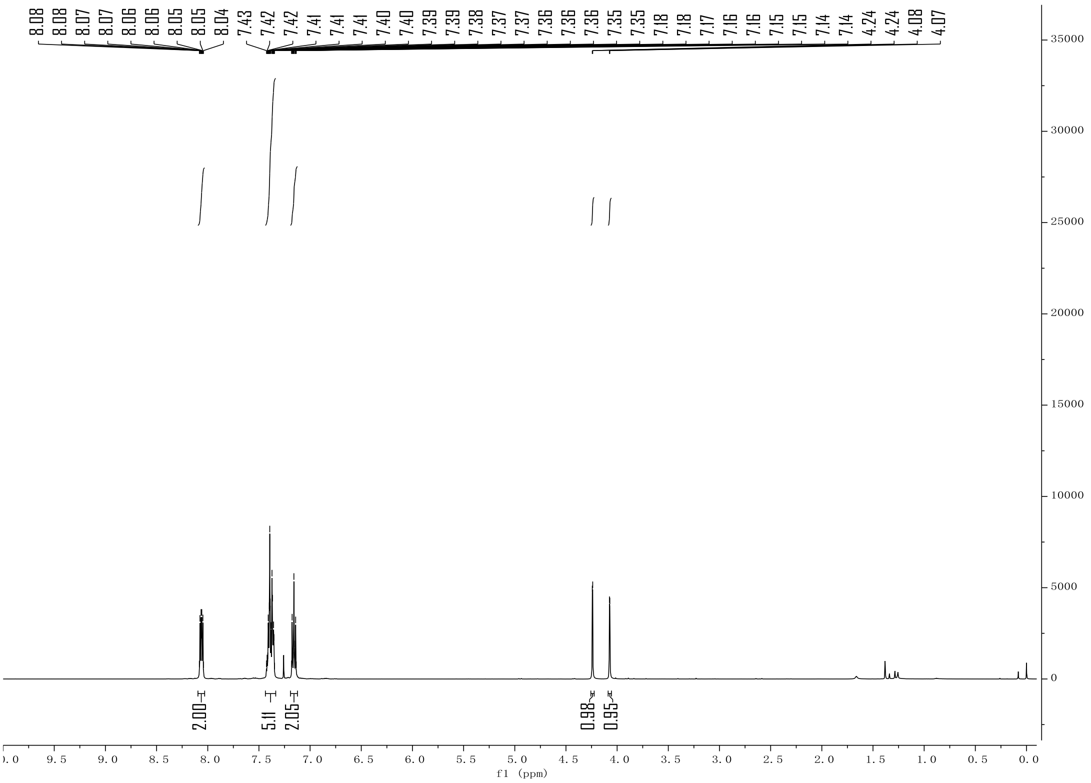
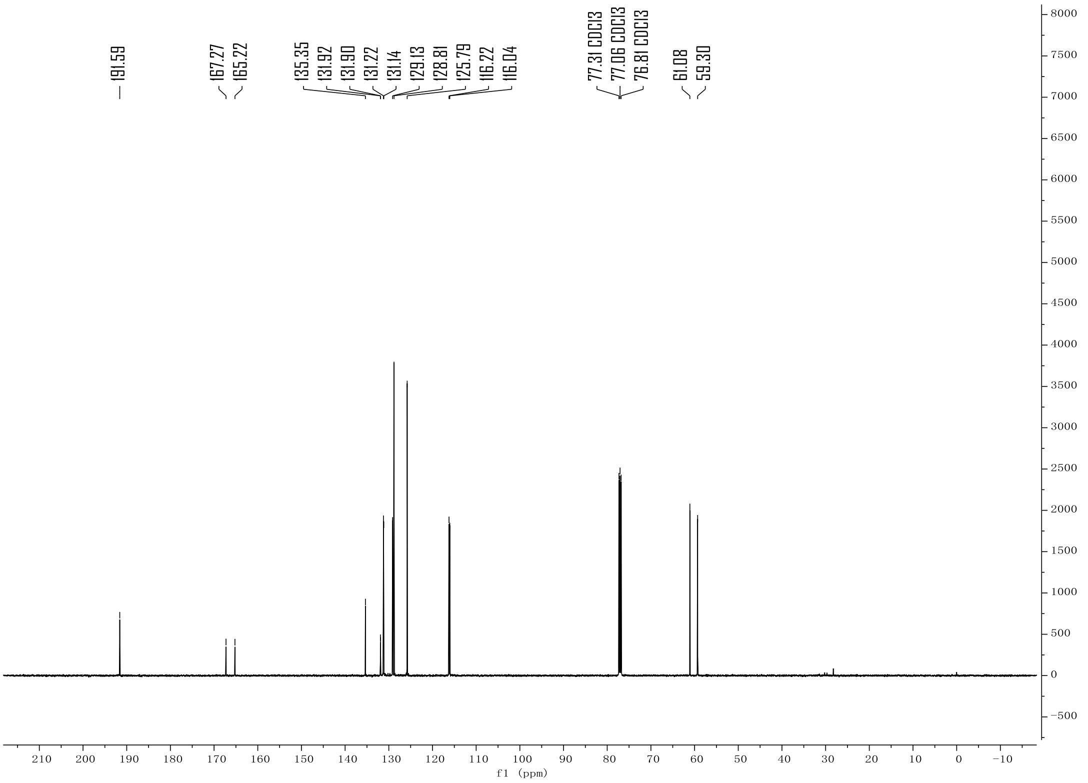
f1 (ppm)



(4-fluorophenyl)(3-phenyloxiran-2-yl)methanone (**b4**)

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77.06 CDCl3
76.81 CDCl3
61.08
59.30
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7000
6500
6000
5500
5000
4500
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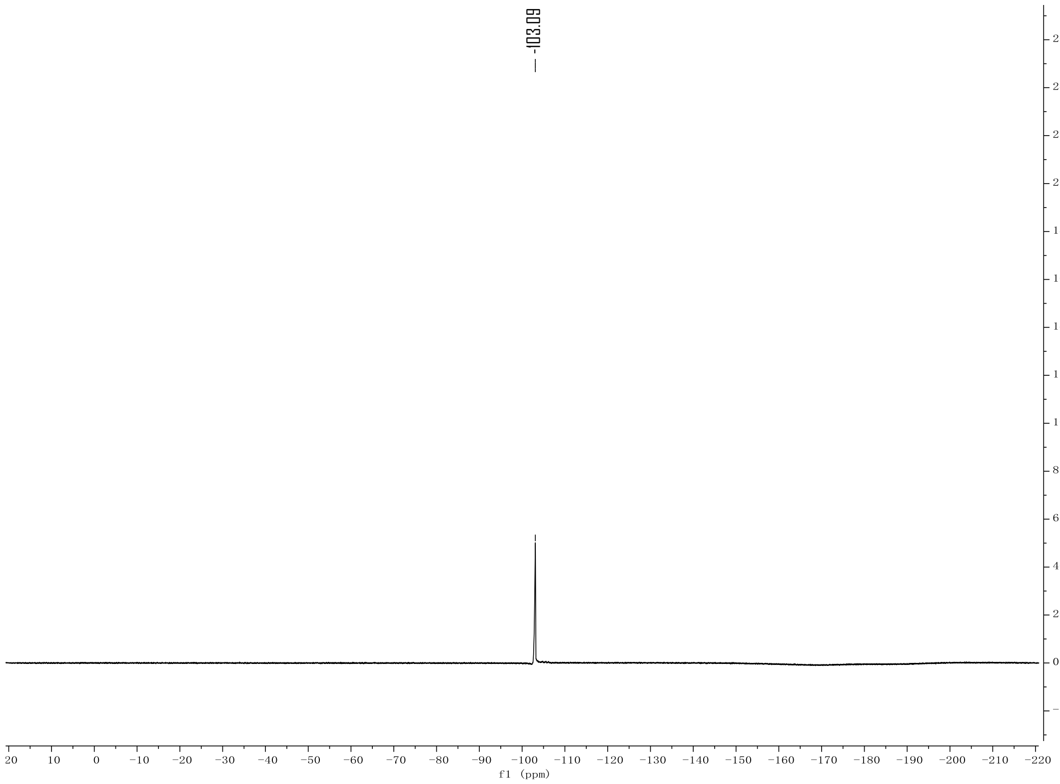
f1 (ppm)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-30
-40
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-60
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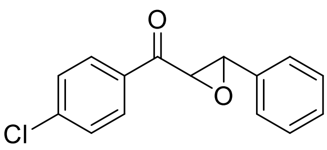
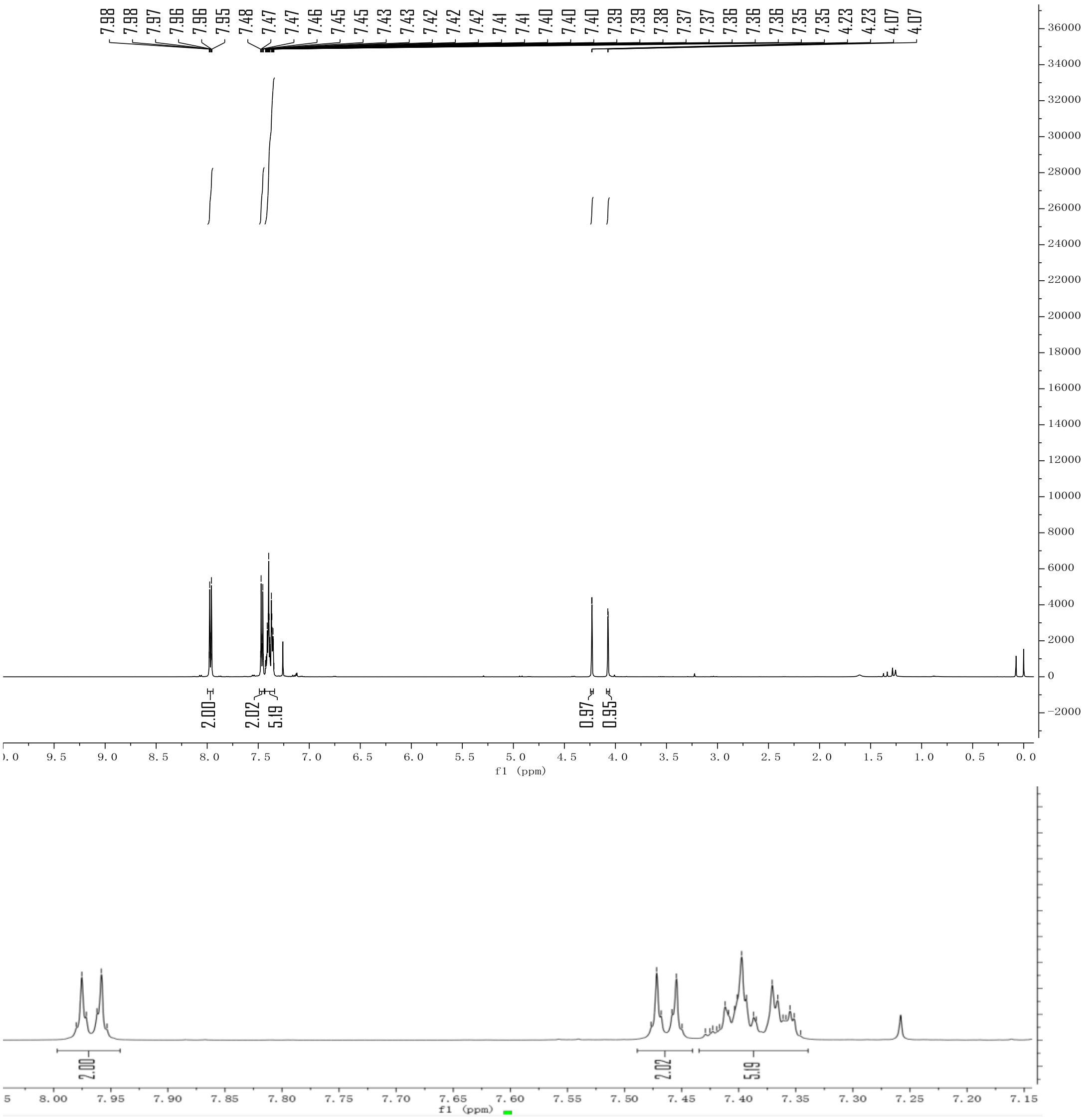
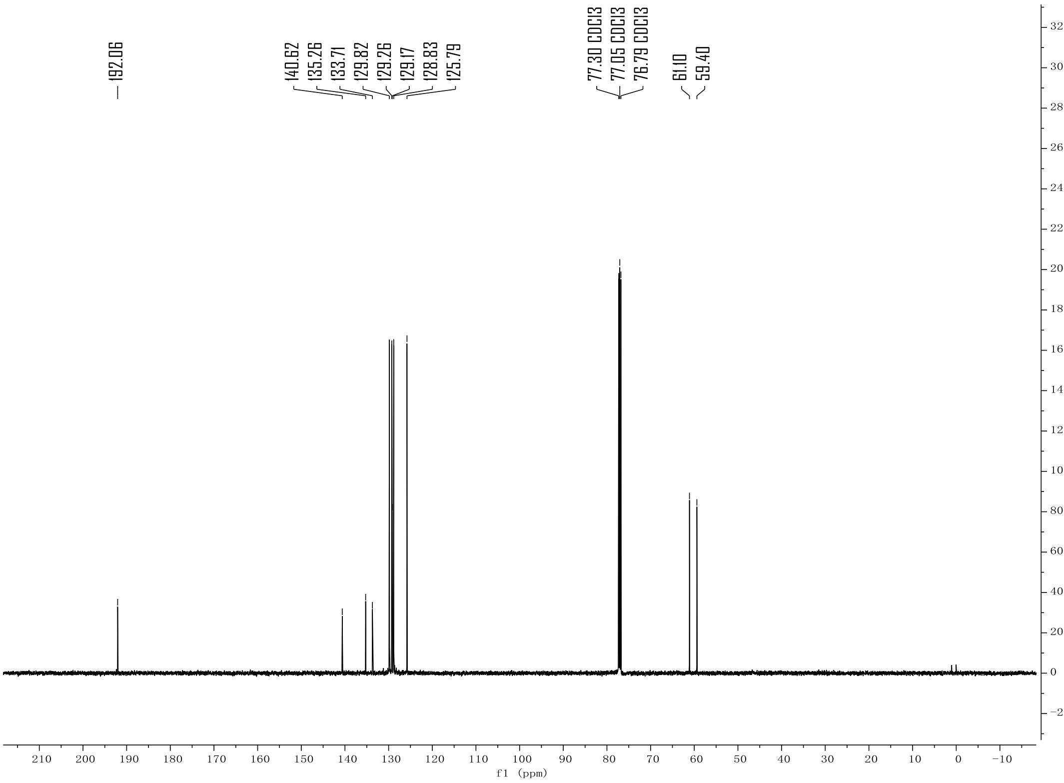
f1 (ppm)



(4-chlorophenyl)(3-phenyloxiran-2-yl)methanone (**b5**)

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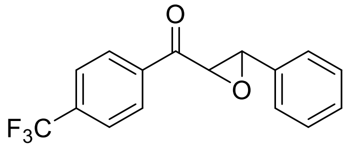
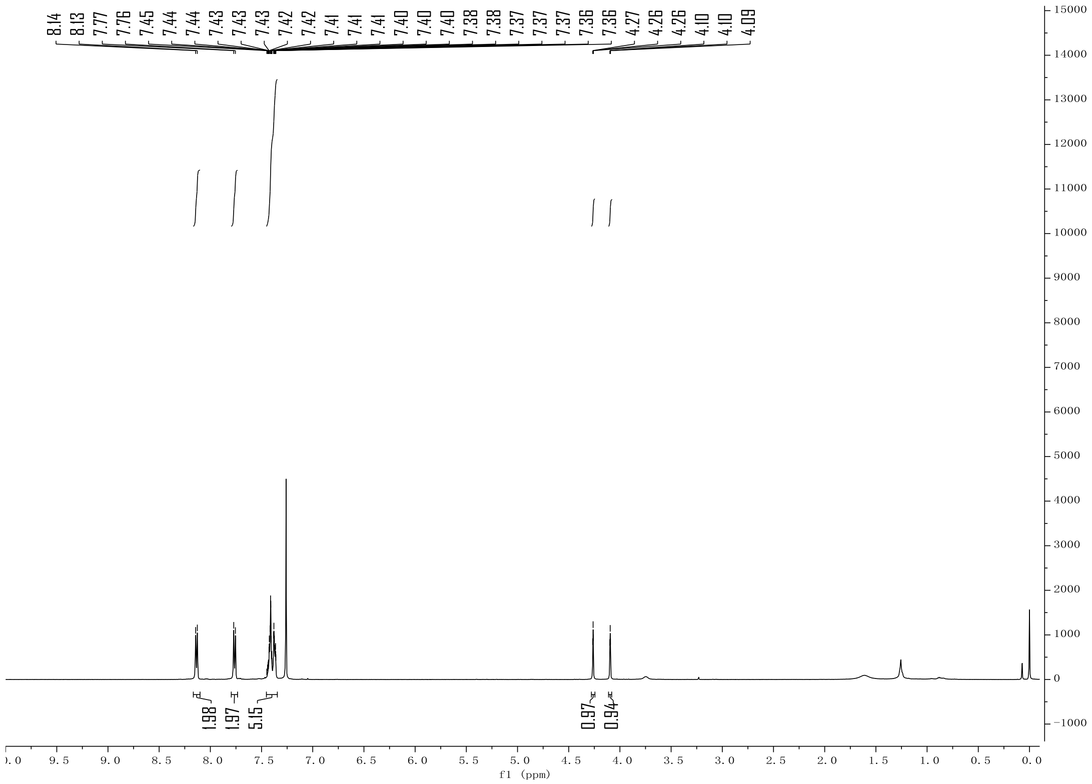
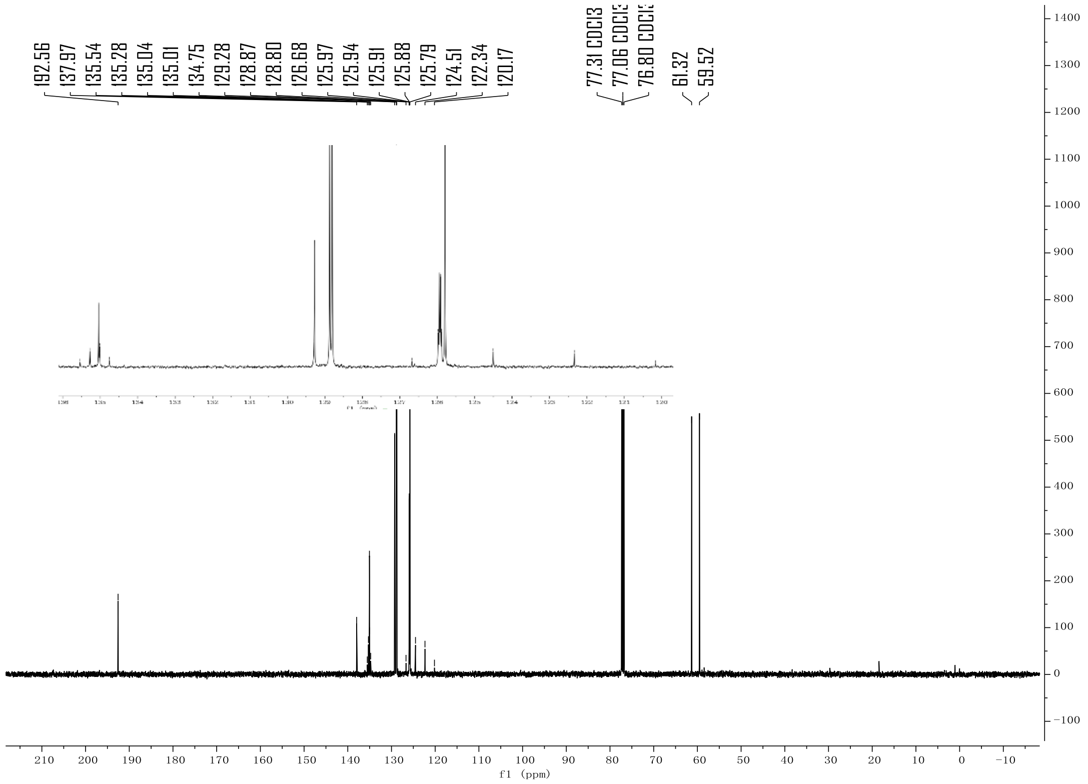
f1 (ppm)



(3-phenyloxiran-2-yl)(4-(trifluoromethyl)phenyl)methanone (**b6**)

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125.91
125.88
125.79
124.51
122.34
120.17
77.31 CDCl3
77.06 CDCl3
76.80 CDCl3
61.32
59.52
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700
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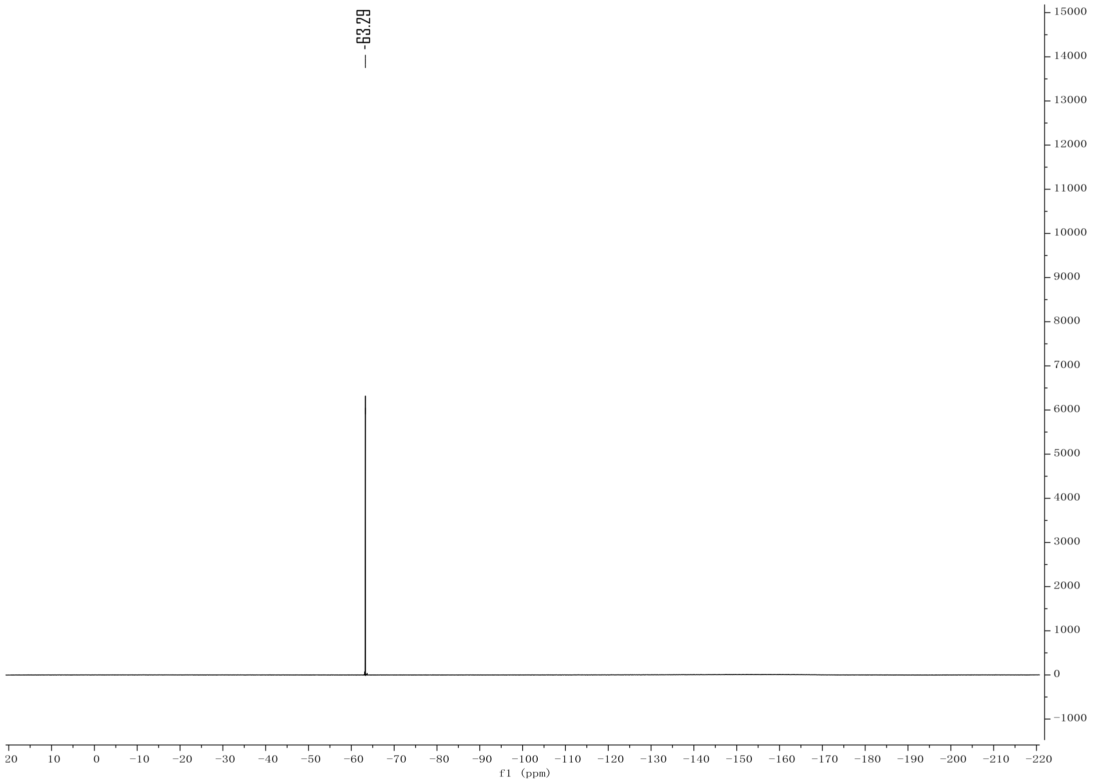
f1 (ppm)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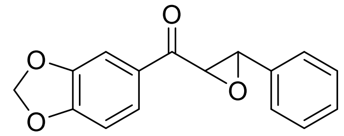
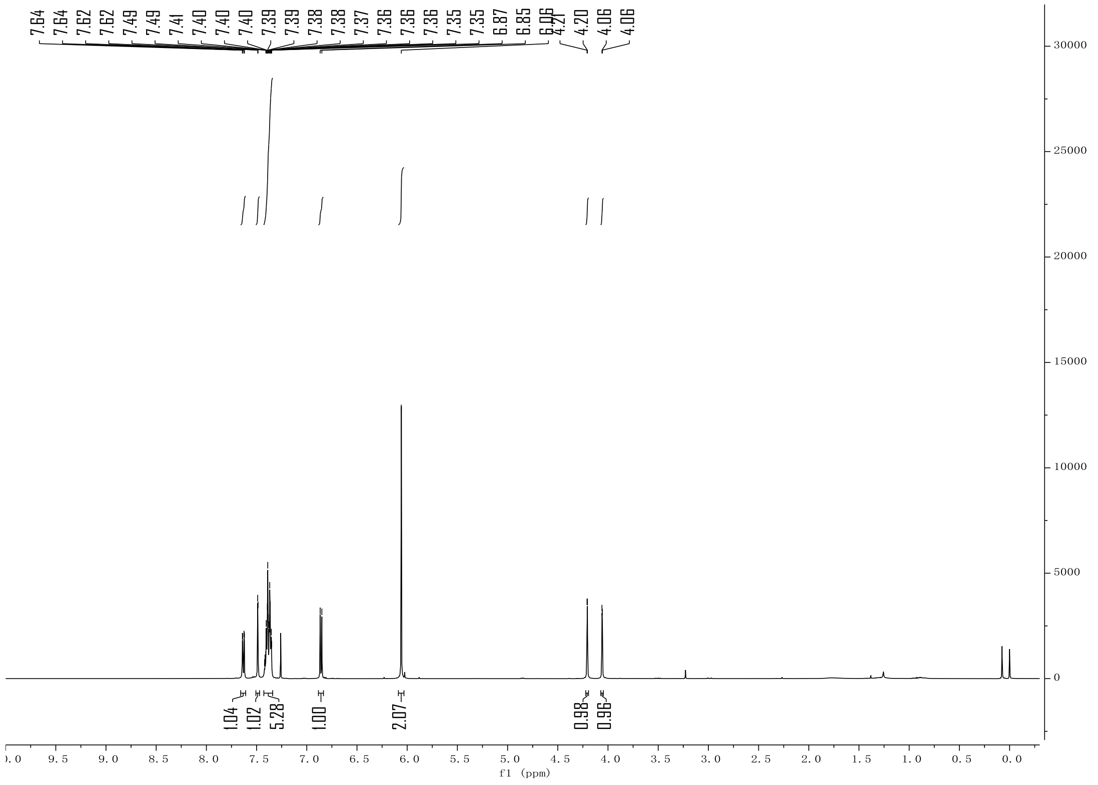
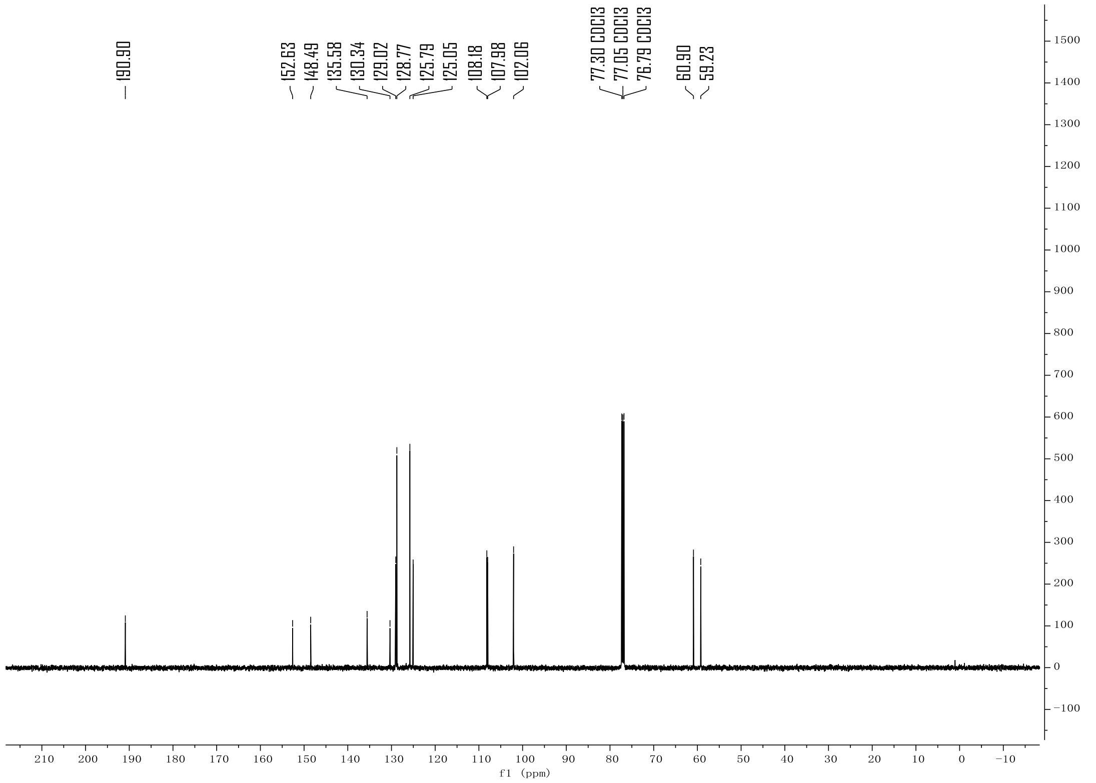
f1 (ppm)



benzo[d][1,3]dioxol-5-yl(3-phenyloxiran-2-yl)methanone (**b7**)

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125.79
125.05
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77.30 CDCl3
77.05 CDCl3
76.79 CDCl3
60.90
59.23
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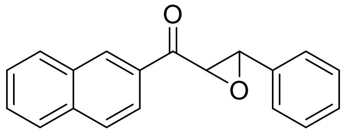
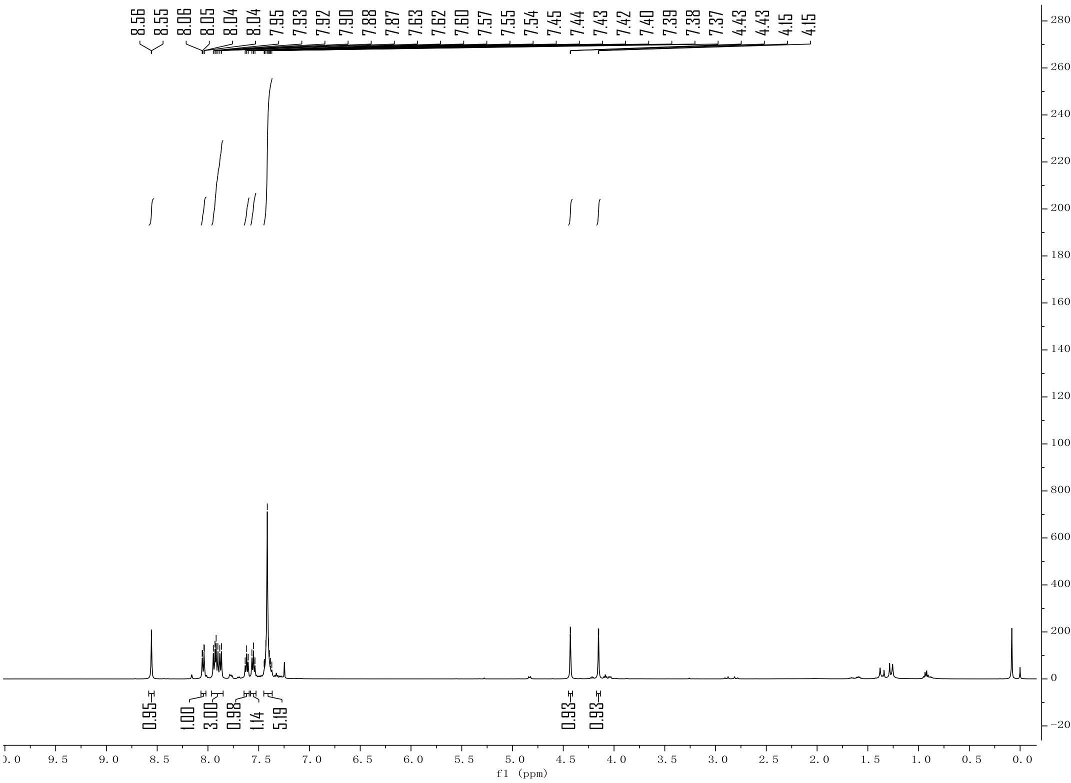
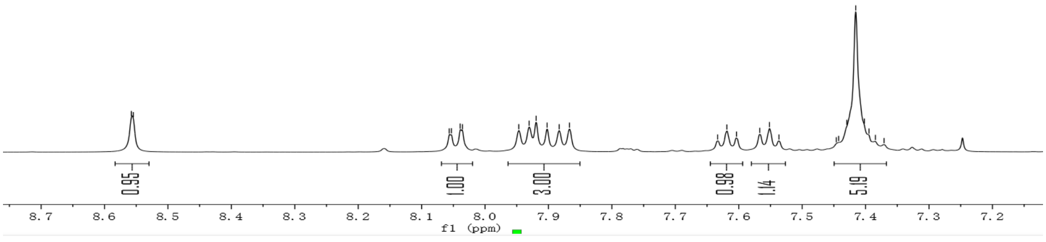
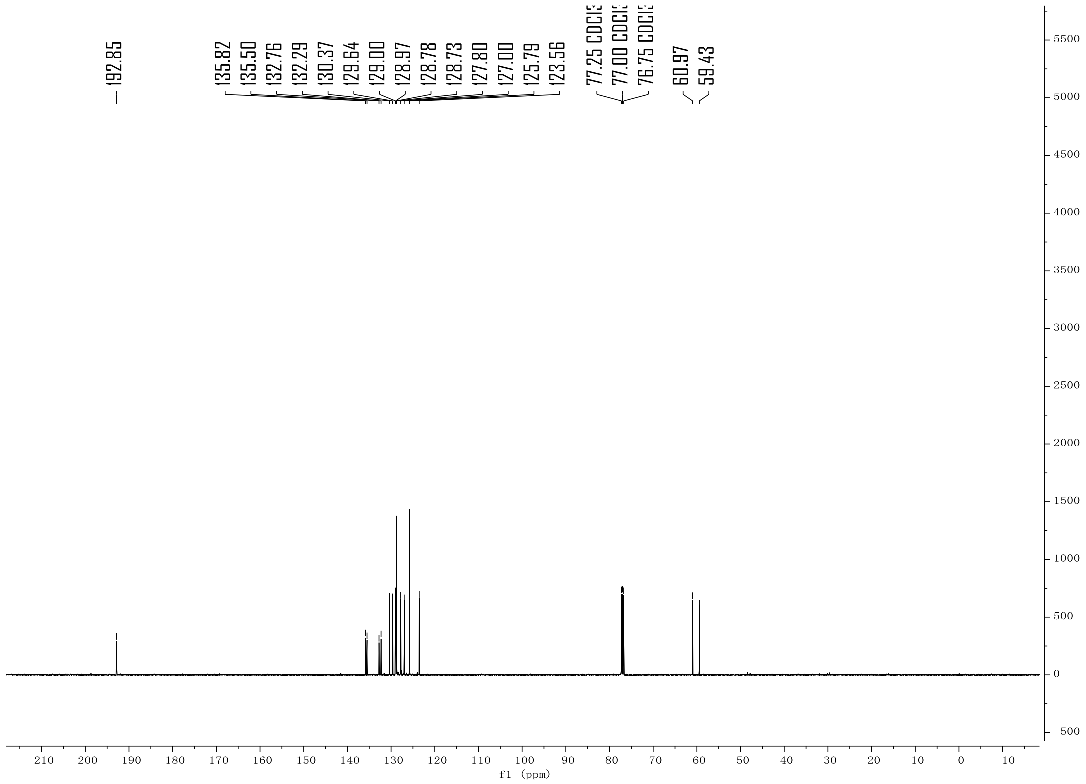
f1 (ppm)



naphthalen-2-yl(3-phenyloxiran-2-yl)methanone (**b8**)



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127.00
125.79
123.56
77.25 CDCl3
77.00 CDCl3
76.75 CDCl3
60.97
59.43
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
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200
190
180
170
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120
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100
90
80
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40
30
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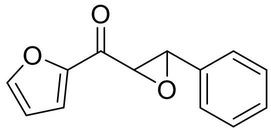
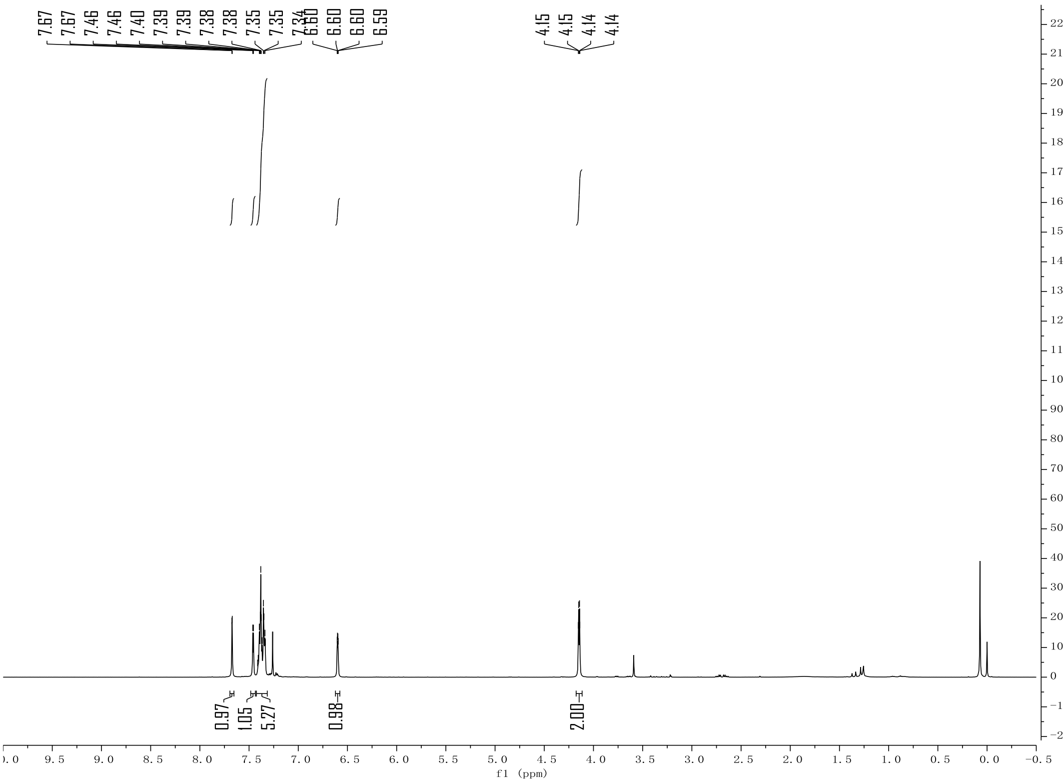
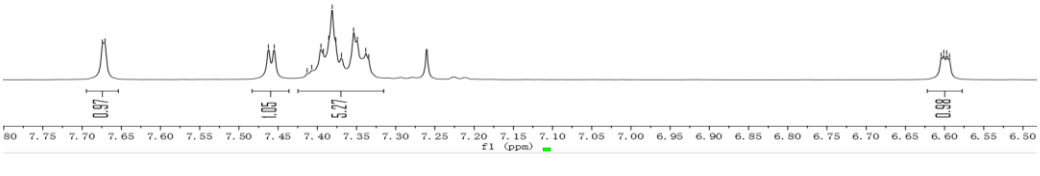
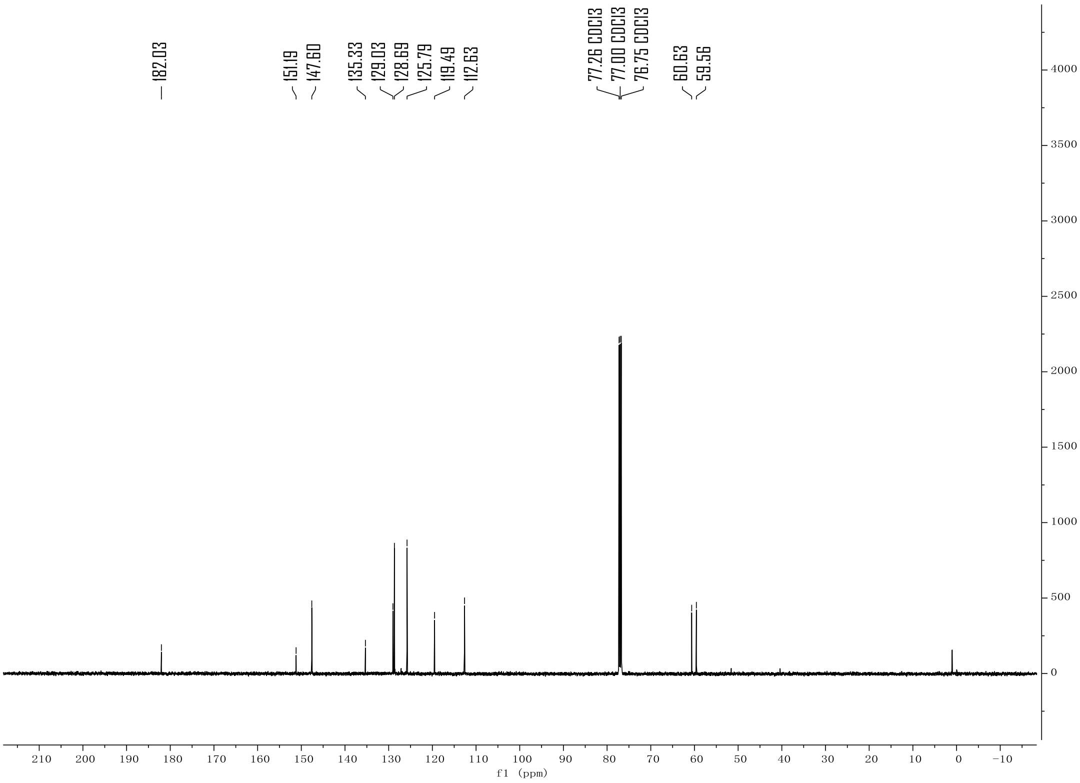
f1 (ppm)



furan-2-yl(3-phenyloxiran-2-yl)methanone (**b9**)



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76.75 CDCl3
60.63
59.56
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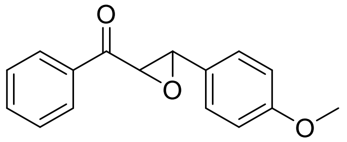
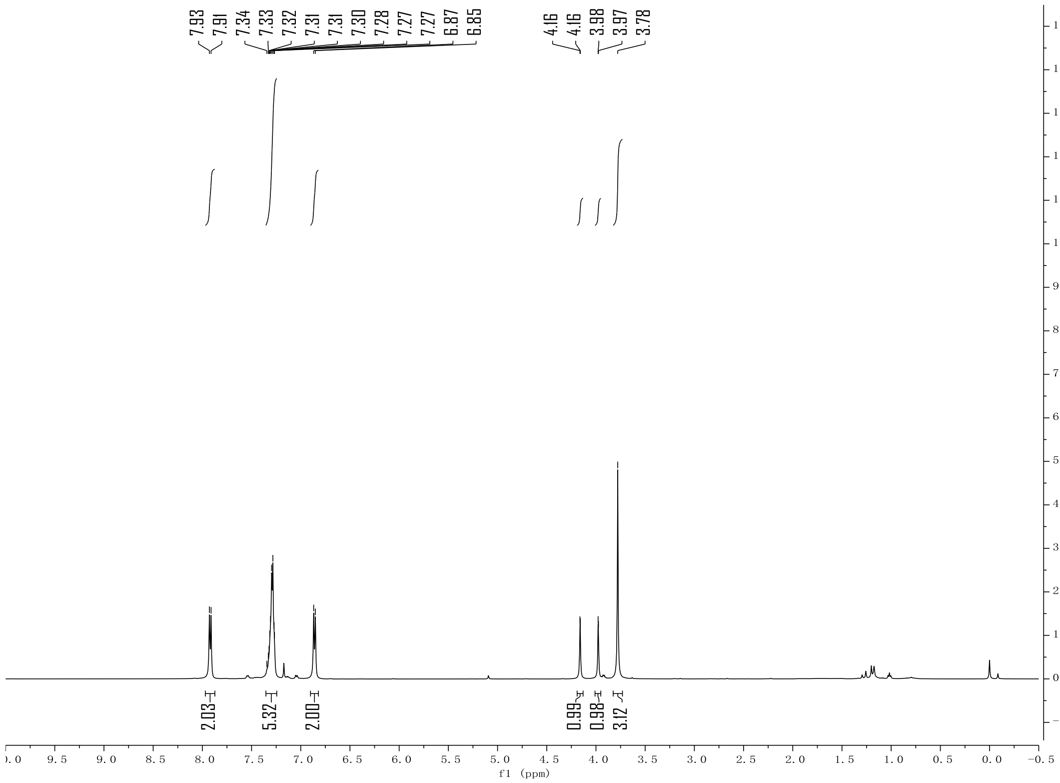
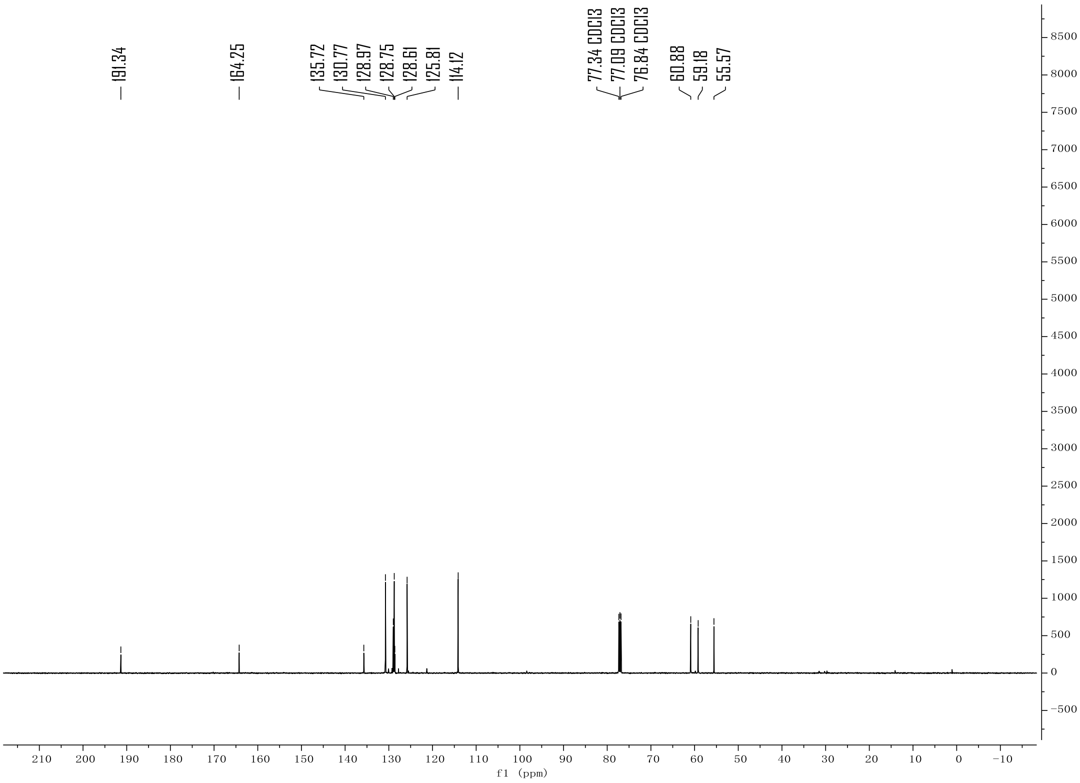
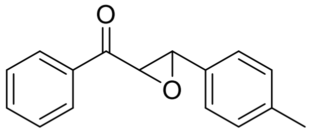
f1 (ppm)



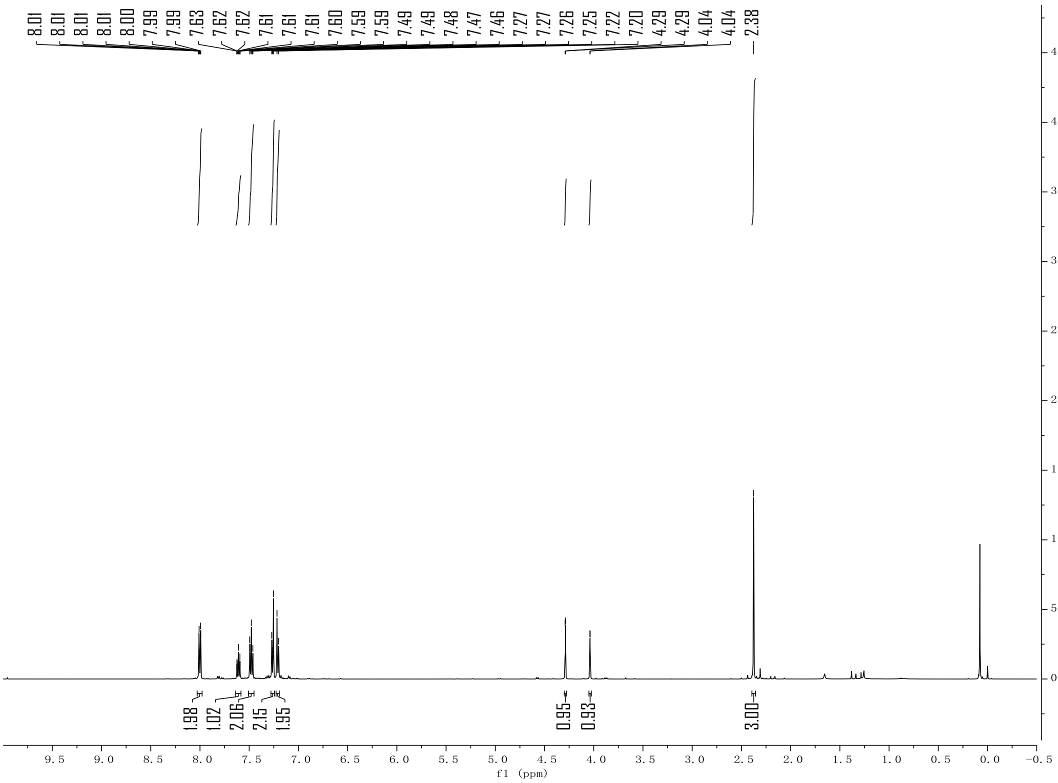
(3-(4-methoxyphenyl)oxiran-2-yl)(phenyl)methanone (**b10**)

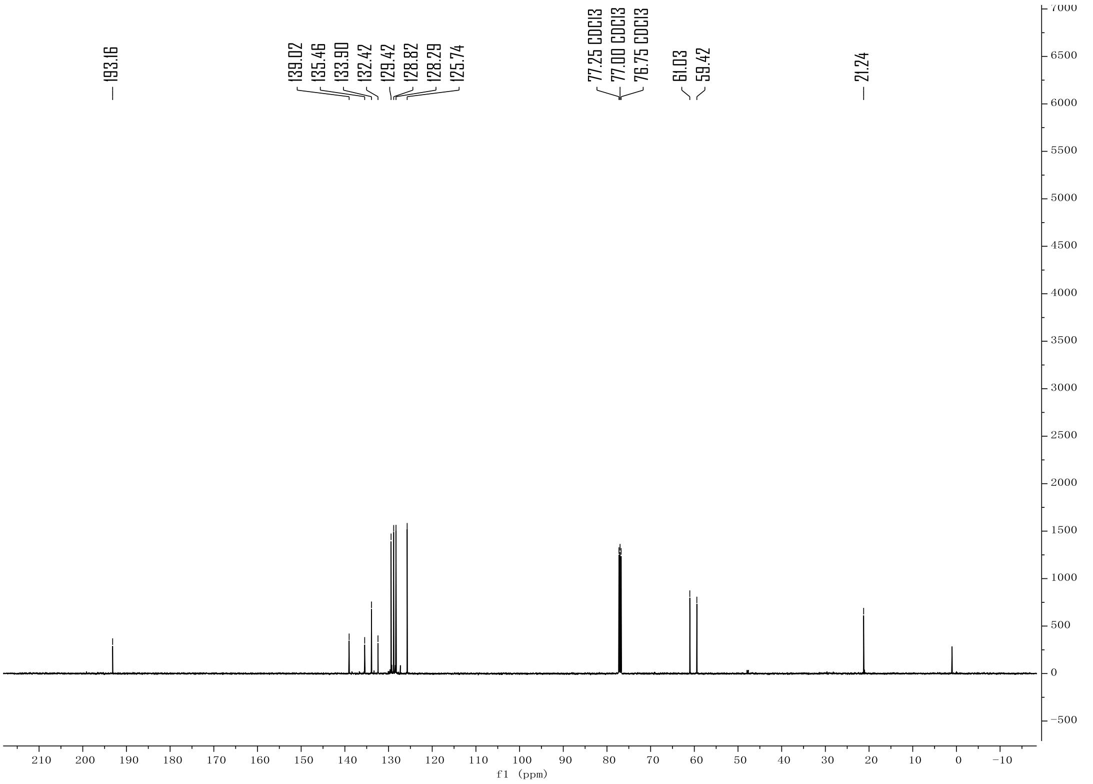



191.34
164.25
135.72
130.77
128.97
128.75
128.61
125.81
114.12
77.34 CDCl3
77.09 CDCl3
76.84 CDCl3
60.88
59.18
55.57
8500
8000
7500
7000
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

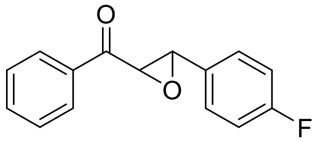


phenyl(3-(p-tolyl)oxiran-2-yl)methanone (**b11**)

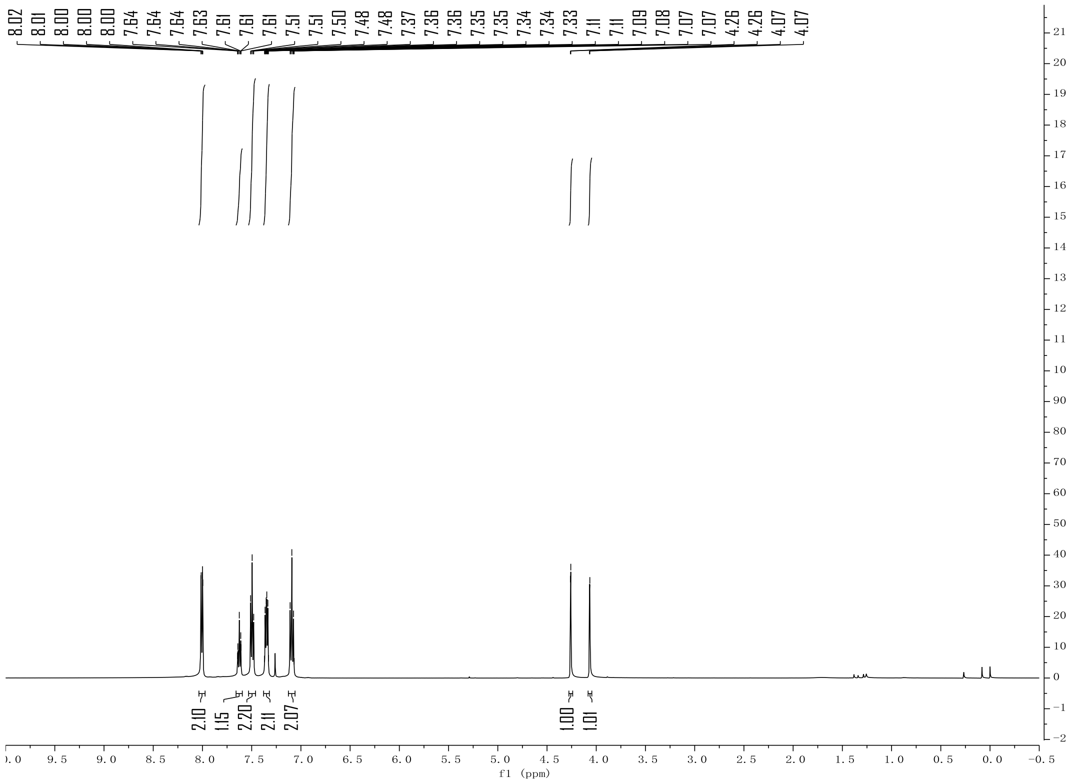


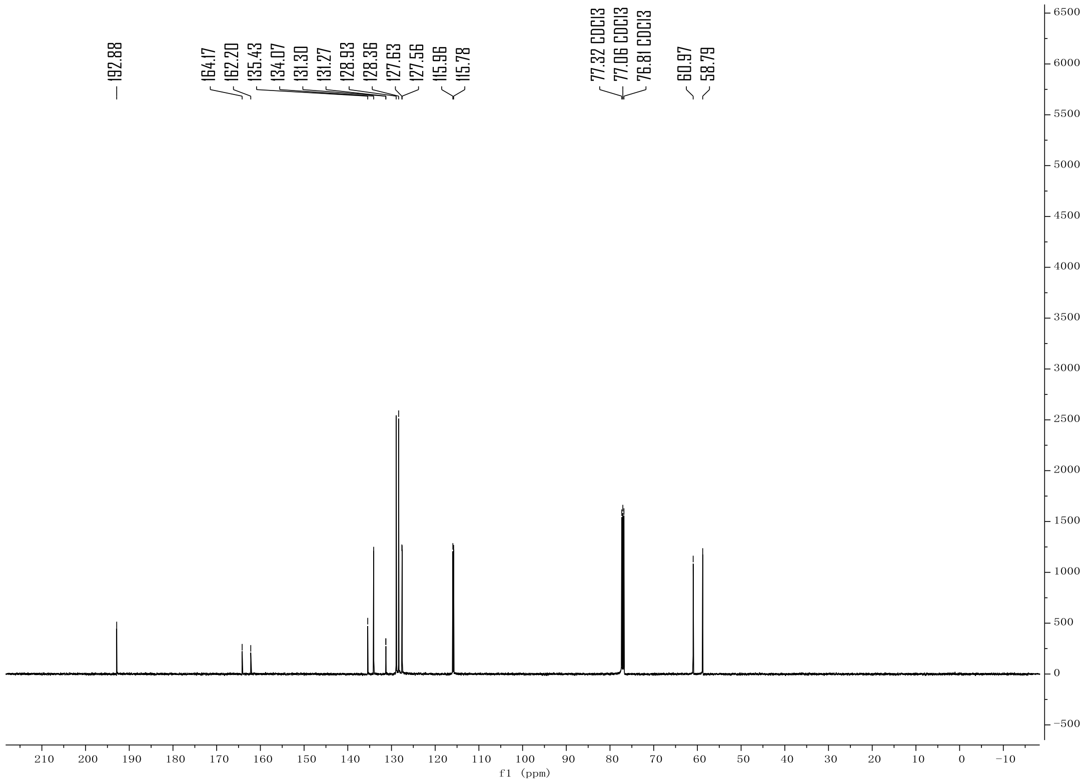


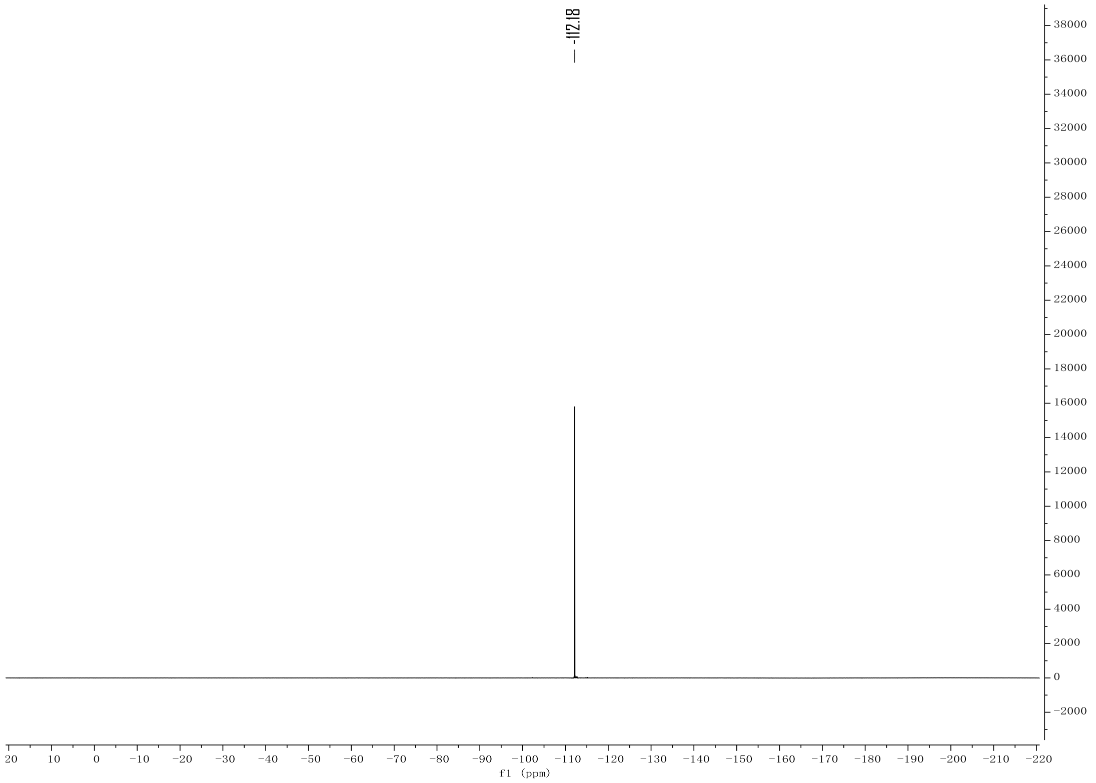
193.16
139.02
135.46
133.90
132.42
129.42
128.82
128.29
125.74
77.25 CDCl3
77.00 CDCl3
76.75 CDCl3
61.03
59.42
21.24
7000
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)



(3-(4-fluorophenyl)oxiran-2-yl)(phenyl)methanone (**b12**)




192.88
164.17
162.20
135.43
134.07
131.30
131.27
128.93
128.36
127.63
127.56
115.96
115.78
77.32 CDCl3
77.06 CDCl3
76.81 CDCl3
60.97
58.79
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
−500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)


-112.18
38000
36000
34000
32000
30000
28000
26000
24000
22000
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0
-2000
20
10
0
-10
-20
-30
-40
-50
-60
-70
-80
-90
-100
-110
-120
-130
-140
-150
-160
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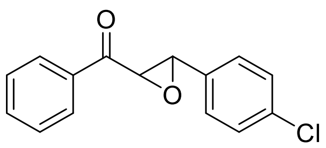
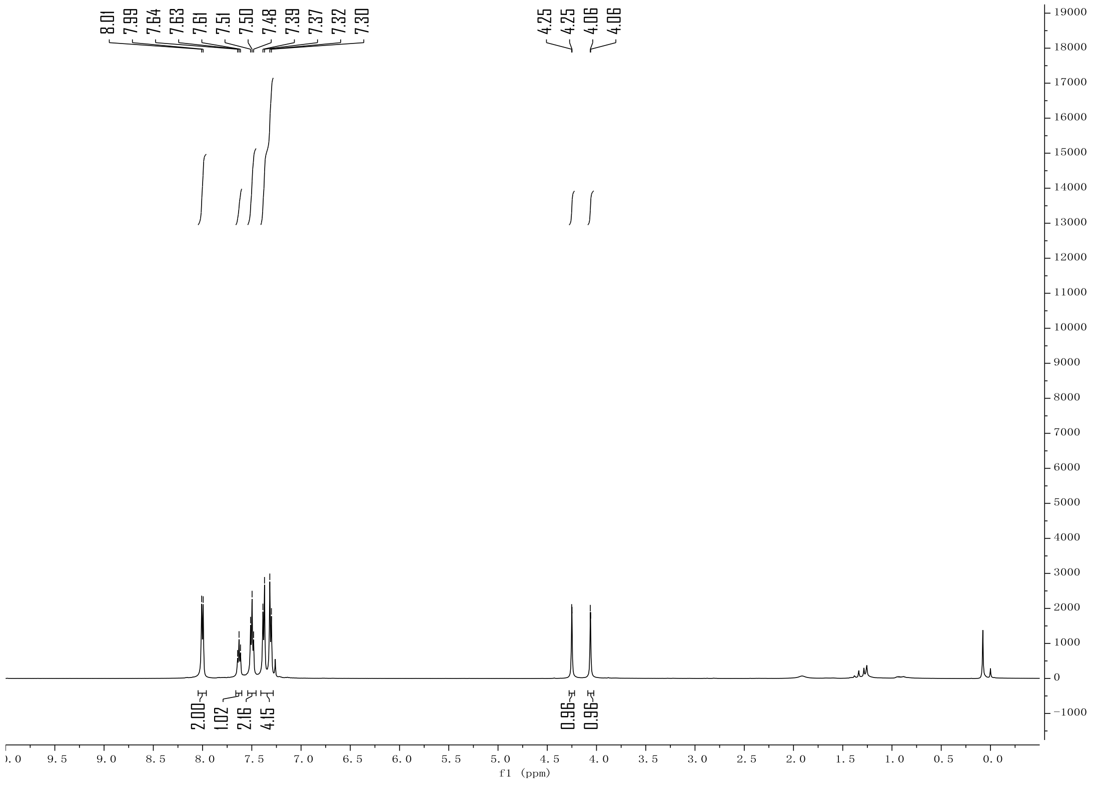
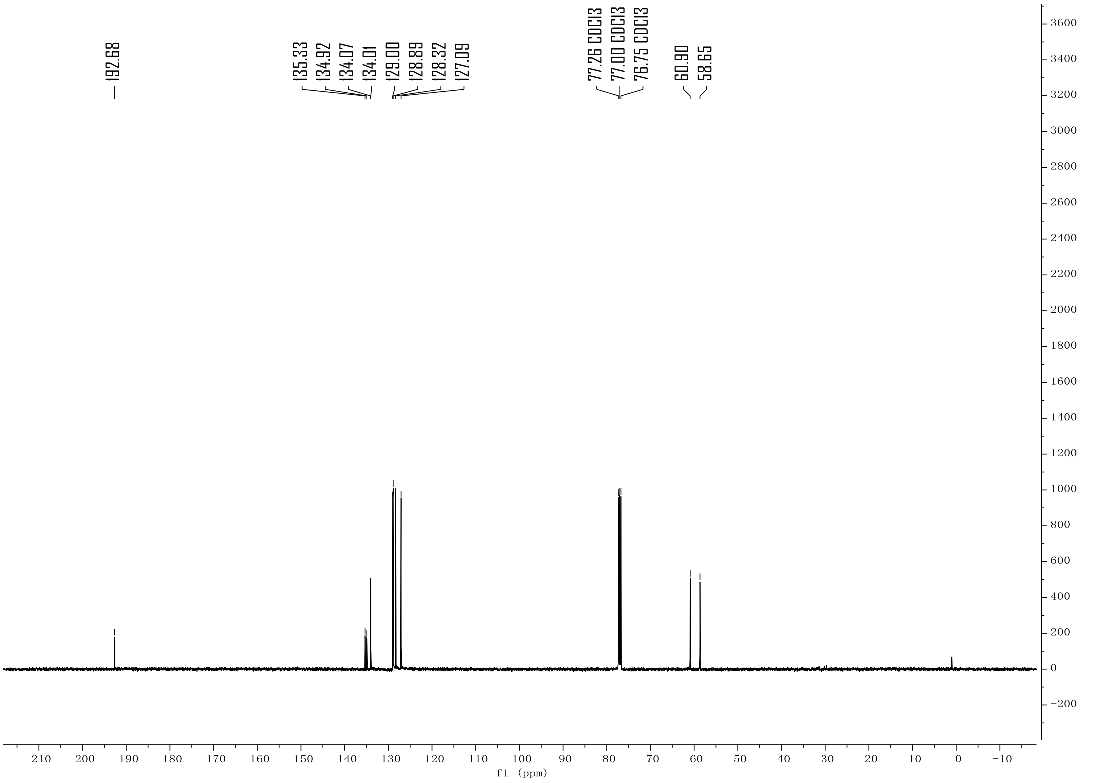
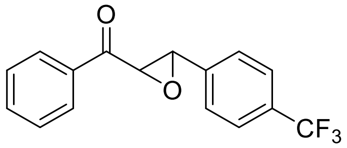
-200
-210
-220
f1 (ppm)



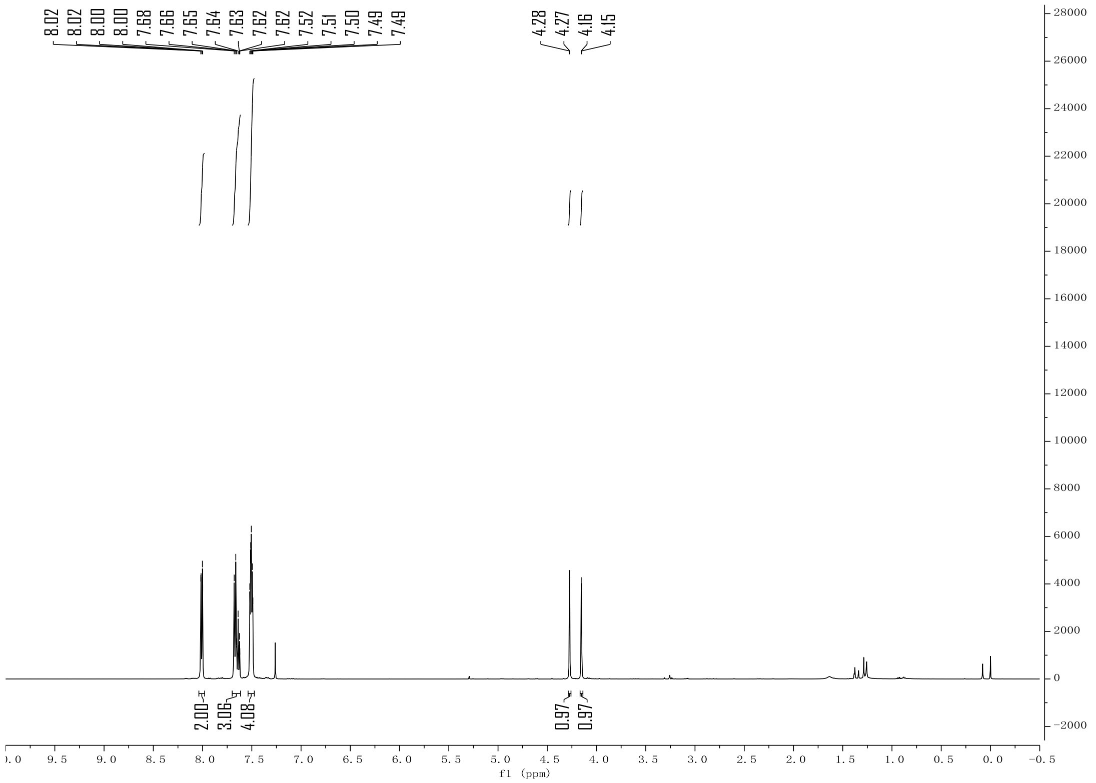
(3-(4-chlorophenyl)oxiran-2-yl)(phenyl)methanone (**b13**)

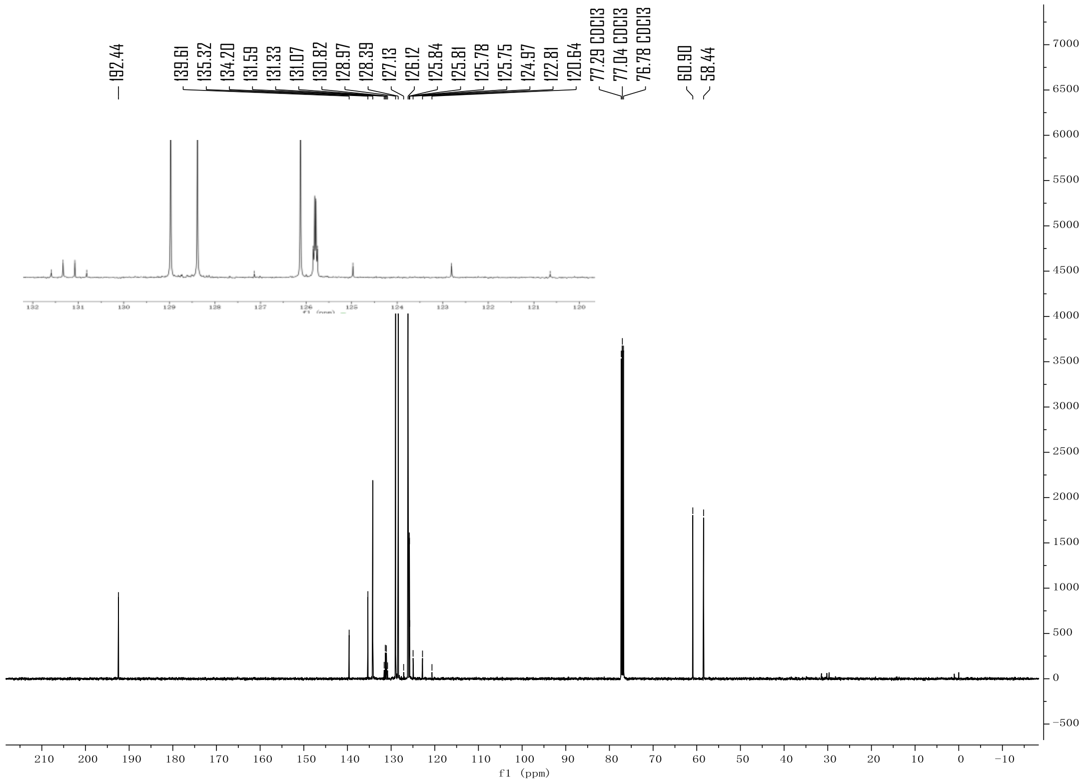


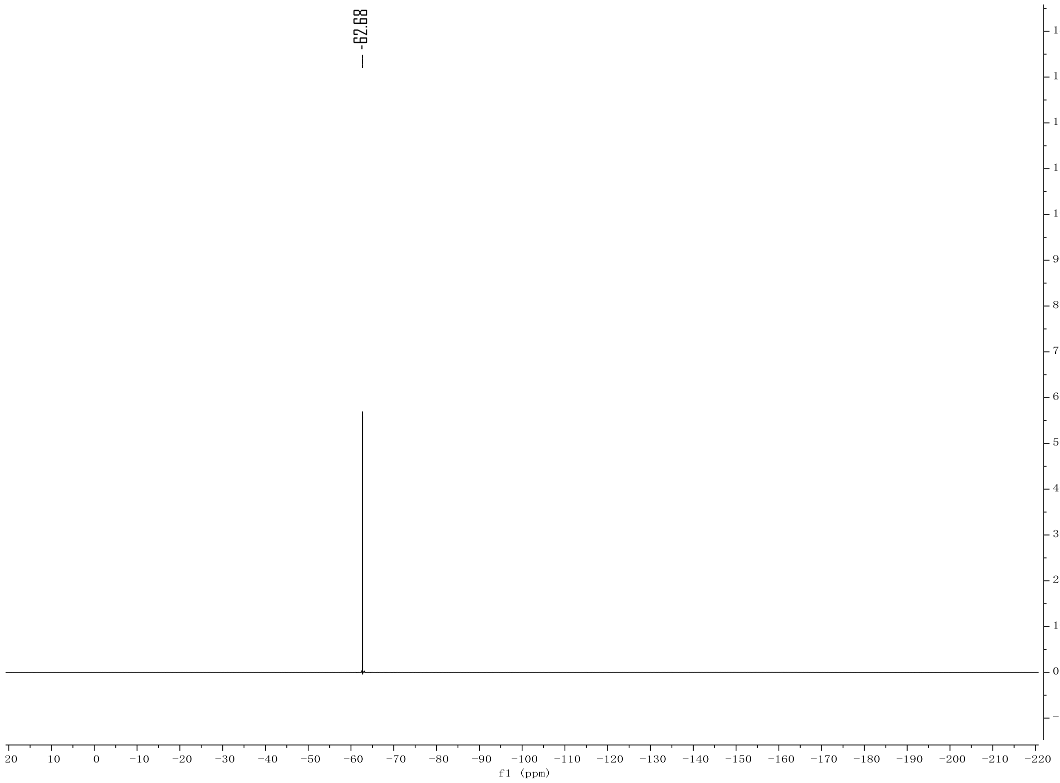

192.68
135.33
134.92
134.07
134.01
129.00
128.89
128.32
127.09
77.26 CDCl3
77.00 CDCl3
76.75 CDCl3
60.90
58.65
3600
3400
3200
3000
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
0
−200
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)



phenyl(3-(4-(trifluoromethyl)phenyl)oxiran-2-yl)methanone (**b14**)




192.44
139.61
135.32
134.20
131.59
131.33
131.07
130.82
128.97
128.39
127.13
126.12
125.84
125.81
125.78
125.75
124.97
122.81
120.64
77.29 CDCl3
77.04 CDCl3
76.78 CDCl3
60.90
58.44
7000
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
−500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)



-62.68
140000
130000
120000
110000
100000
90000
80000
70000
60000
50000
40000
30000
20000
10000
0
-10000
20
10
0
-10
-20
-30
-40
-50
-60
-70
-80
-90
-100
-110
-120
-130
-140
-150
-160
-170
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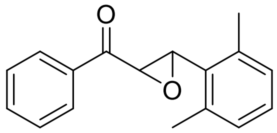
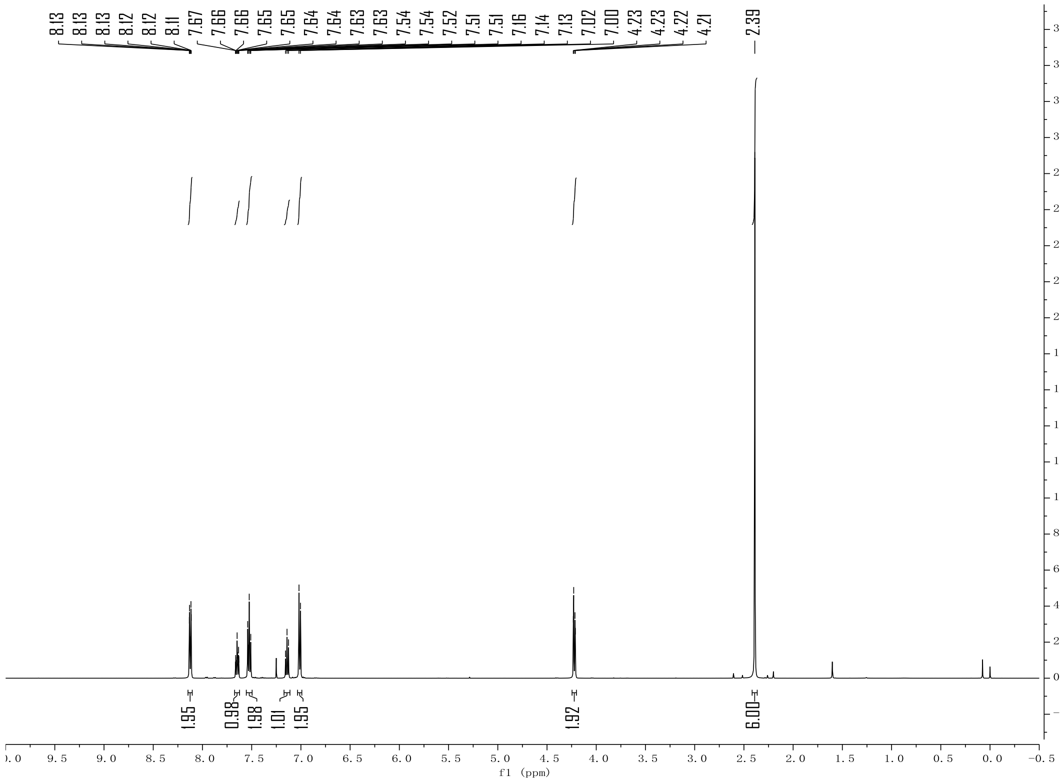
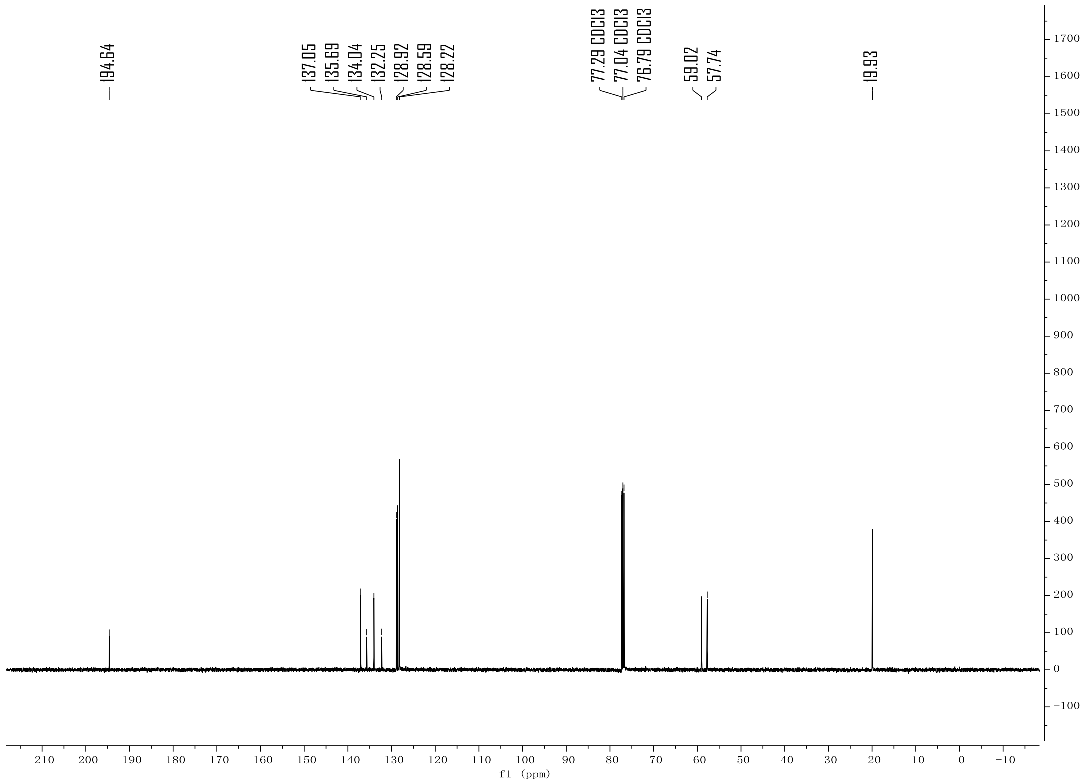
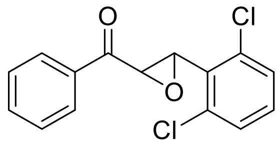
-190
-200
-210
-220
f1 (ppm)



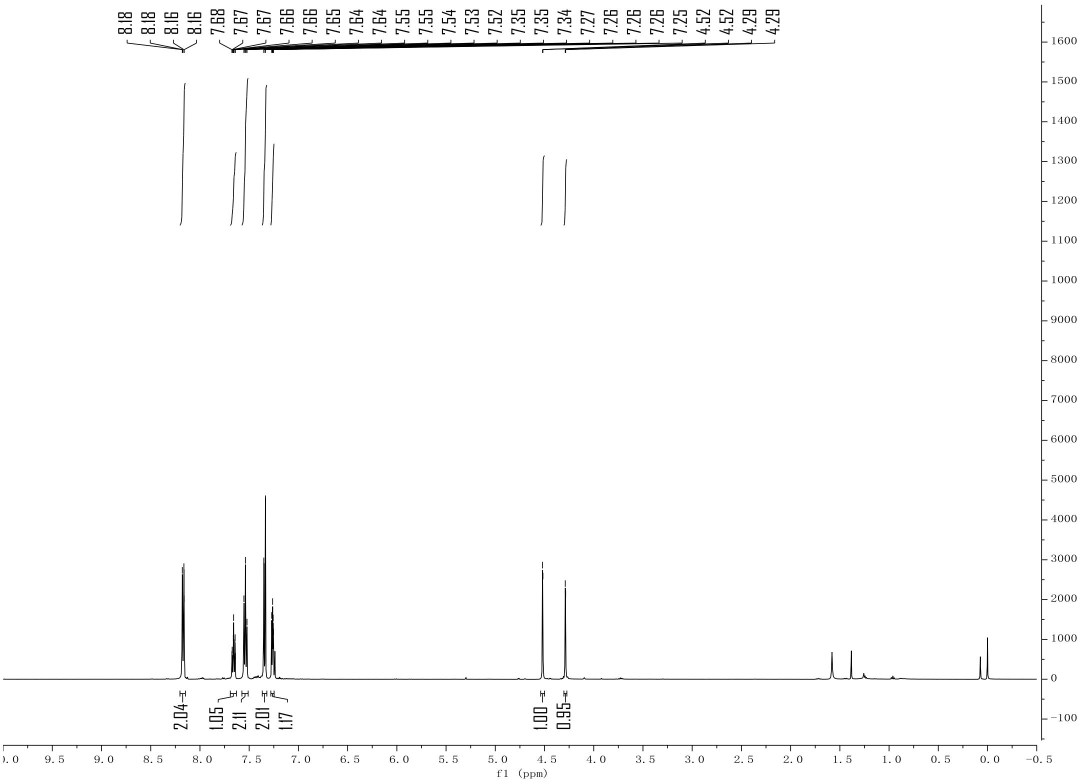
(3-(2,6-dimethylphenyl)oxiran-2-yl)(phenyl)methanone (**b15**)

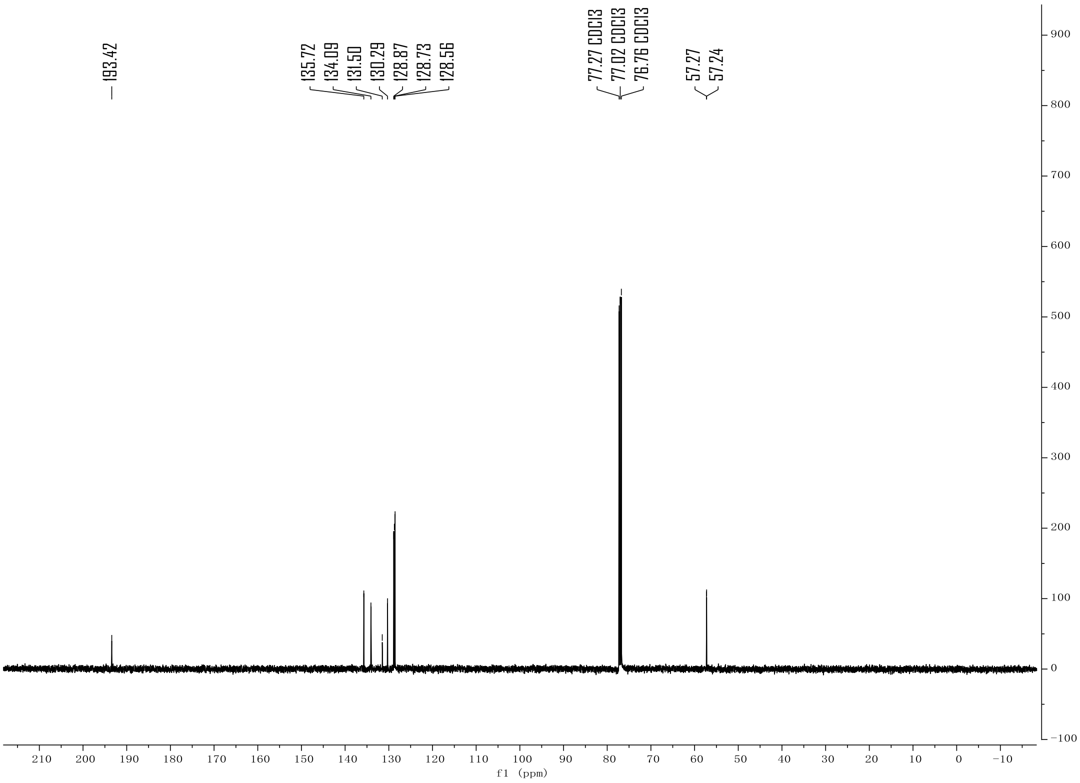


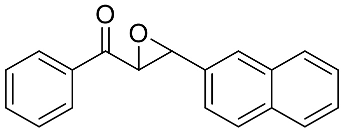

194.64
137.05
135.69
134.04
132.25
128.92
128.59
128.22
77.29 CDCl3
77.04 CDCl3
76.79 CDCl3
59.02
57.74
19.93
f1 (ppm)



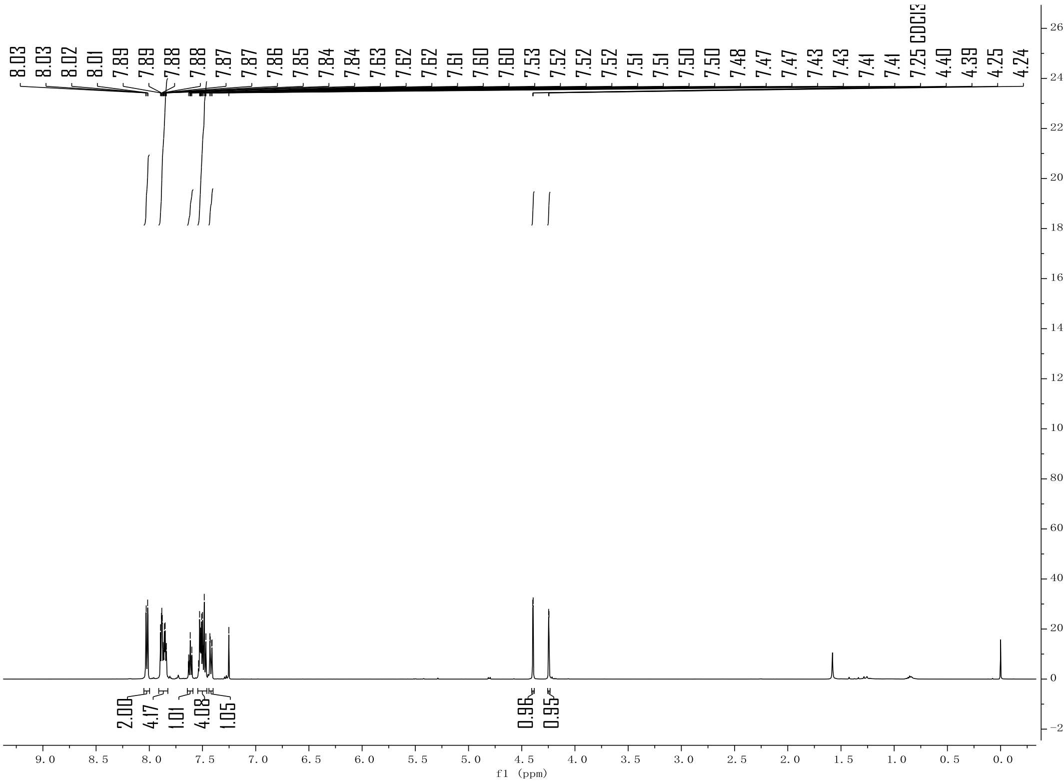
(3-(2,6-dichlorophenyl)oxiran-2-yl)(phenyl)methanone (**b16**)

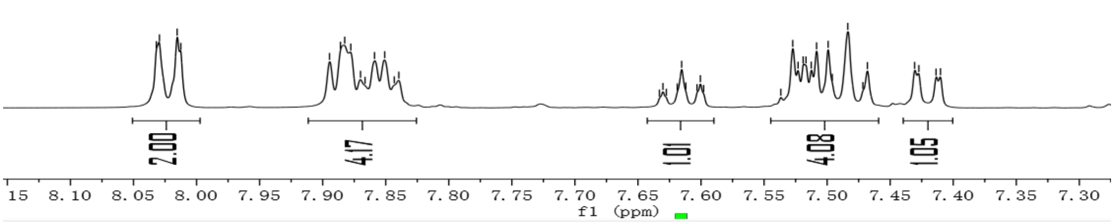


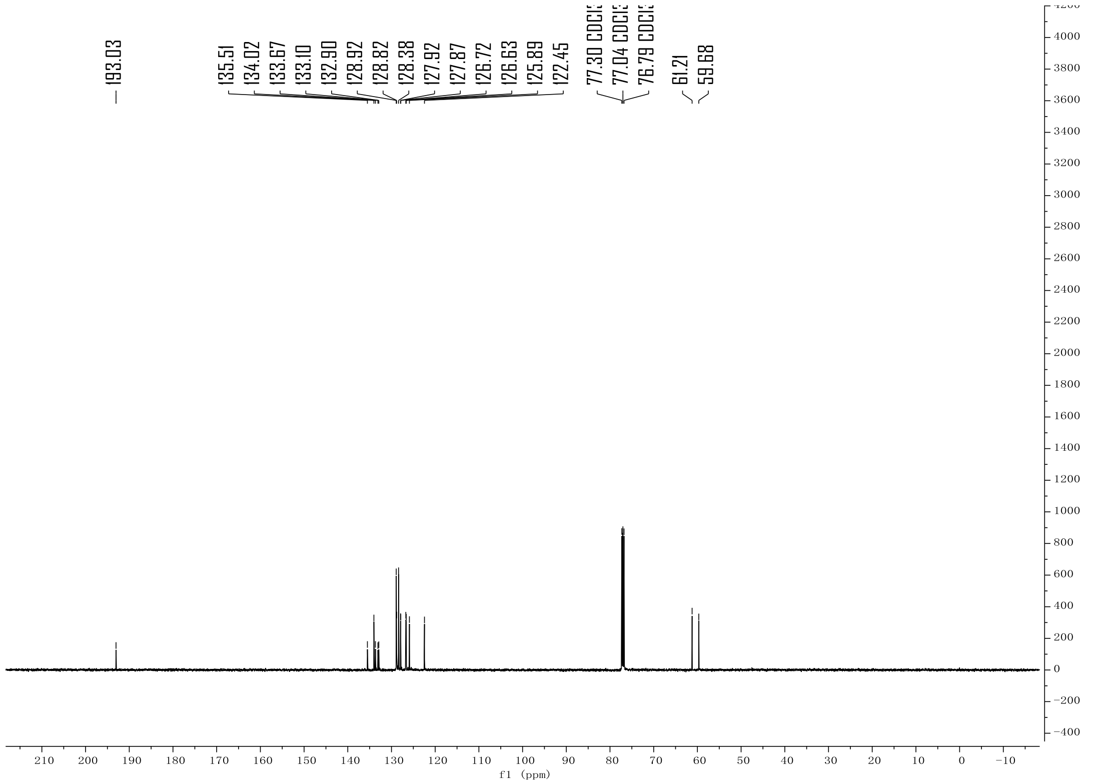

193.42
135.72
134.09
131.50
130.29
128.87
128.73
128.56
77.27 CDCl3
77.02 CDCl3
76.76 CDCl3
57.27
57.24
900
800
700
600
500
400
300
200
100
0
−100
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 −10
f1 (ppm)

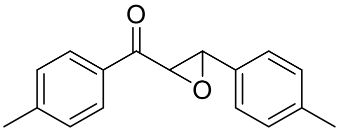


(3-(naphthalen-2-yl)oxiran-2-yl)(phenyl)methanone (**b17**)

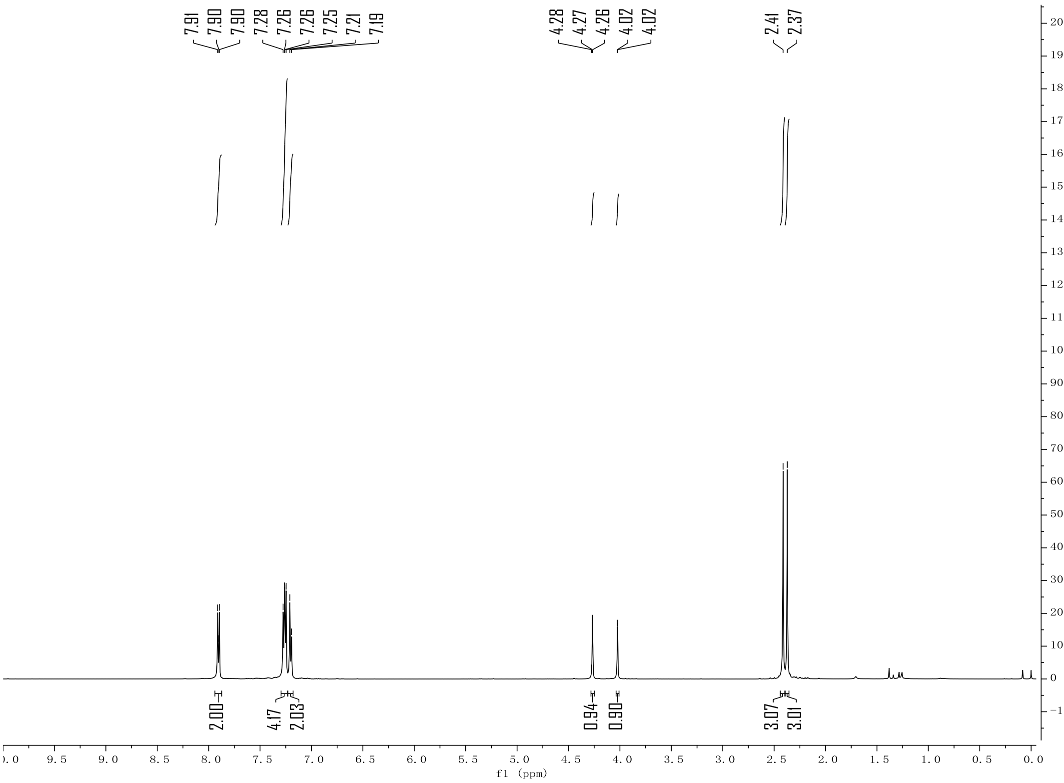


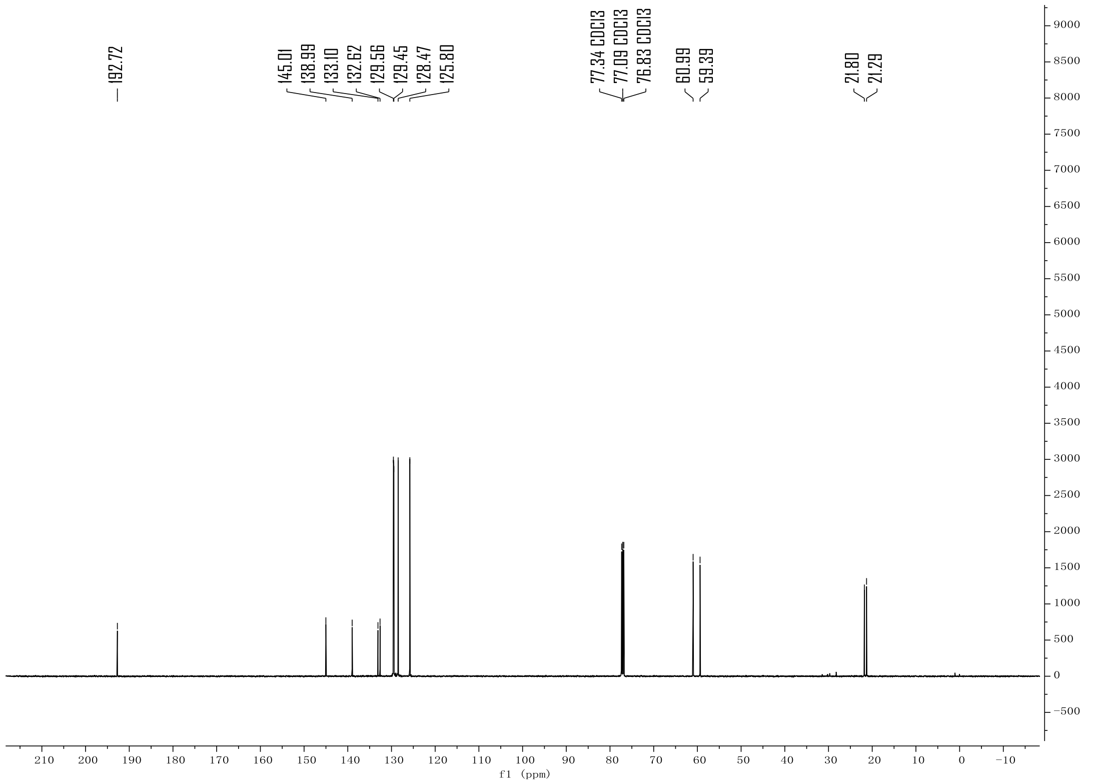


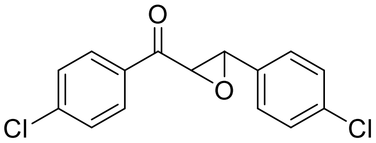

193.03
135.51
134.02
133.67
133.10
132.90
128.92
128.82
128.38
127.92
127.87
126.72
126.63
125.89
122.45
77.30 CDCl3
77.04 CDCl3
76.79 CDCl3
61.21
59.68
4000
3800
3600
3400
3200
3000
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
0
−200
−400
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)



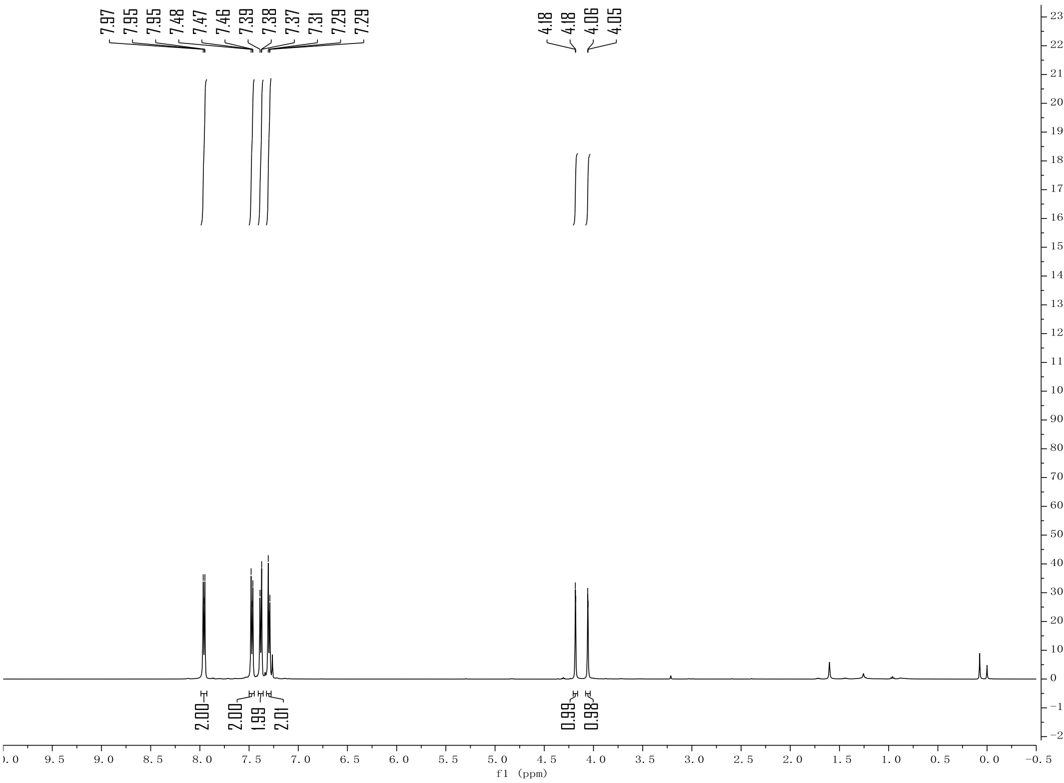
p-tolyl(3-(*p*-tolyl)oxiran-2-yl)methanone (**b18**)

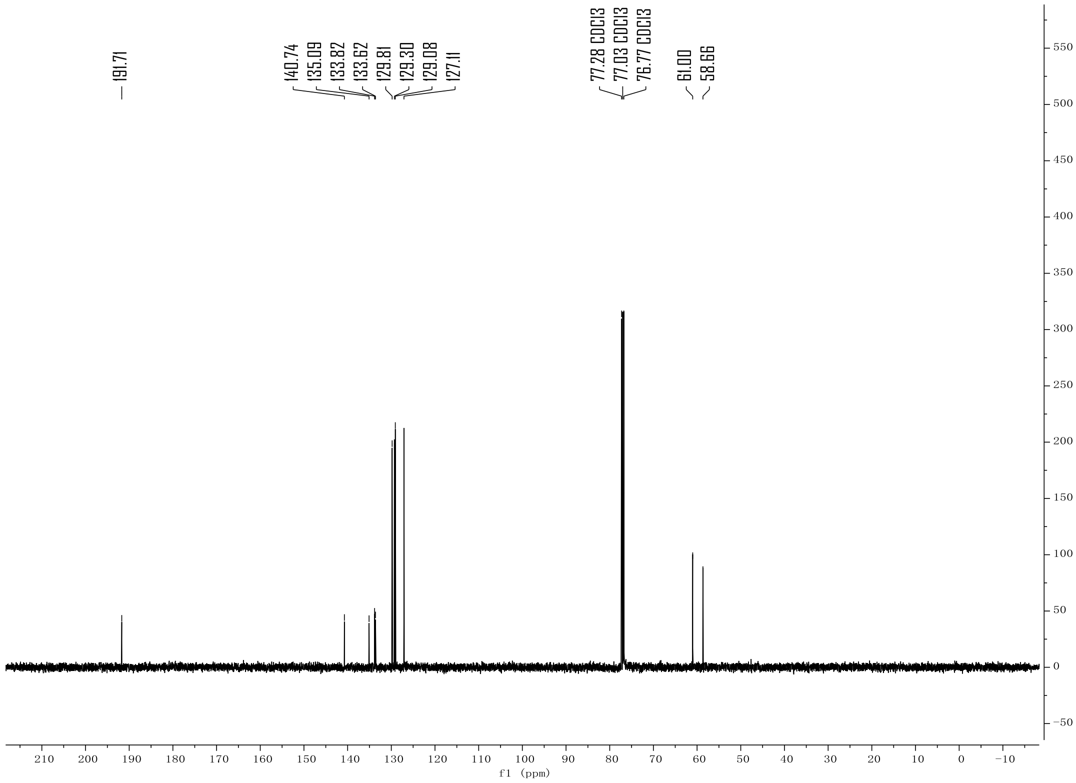


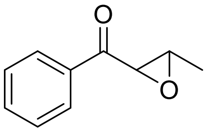

192.72
145.01
138.99
133.10
132.62
129.56
129.45
128.47
125.80
77.34 CDCl3
77.09 CDCl3
76.83 CDCl3
60.99
59.39
21.80
21.29
9000
8500
8000
7500
7000
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)



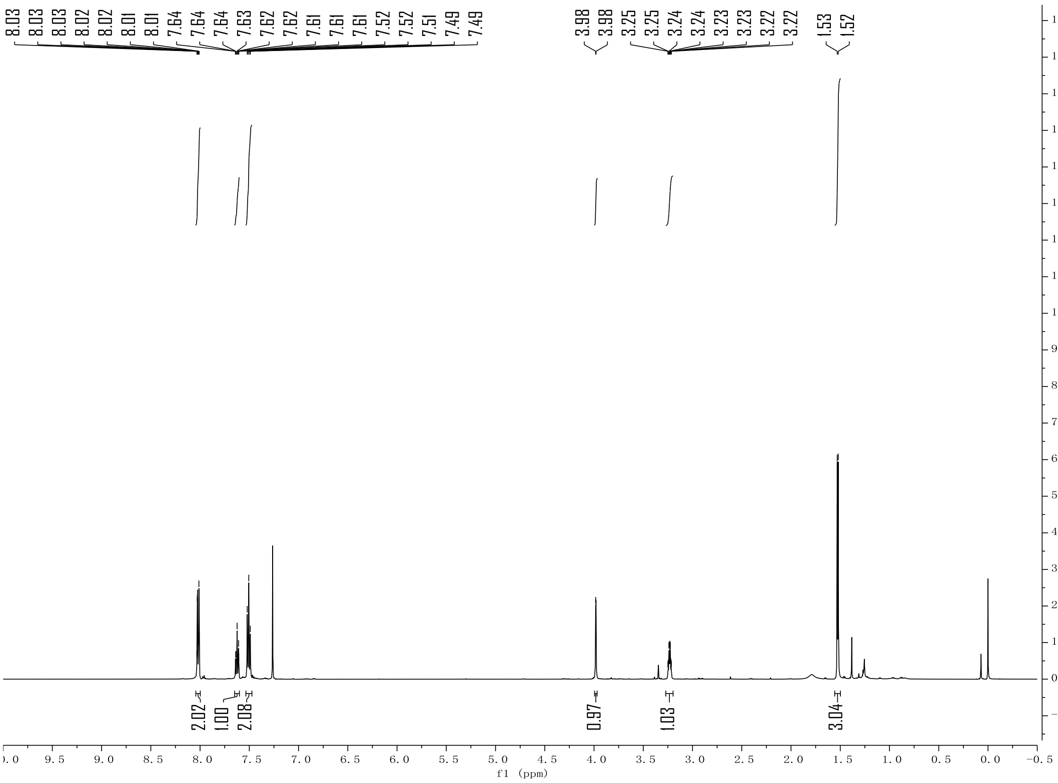
(4-chlorophenyl)(3-(4-chlorophenyl)oxiran-2-yl)methanone (**b19**)

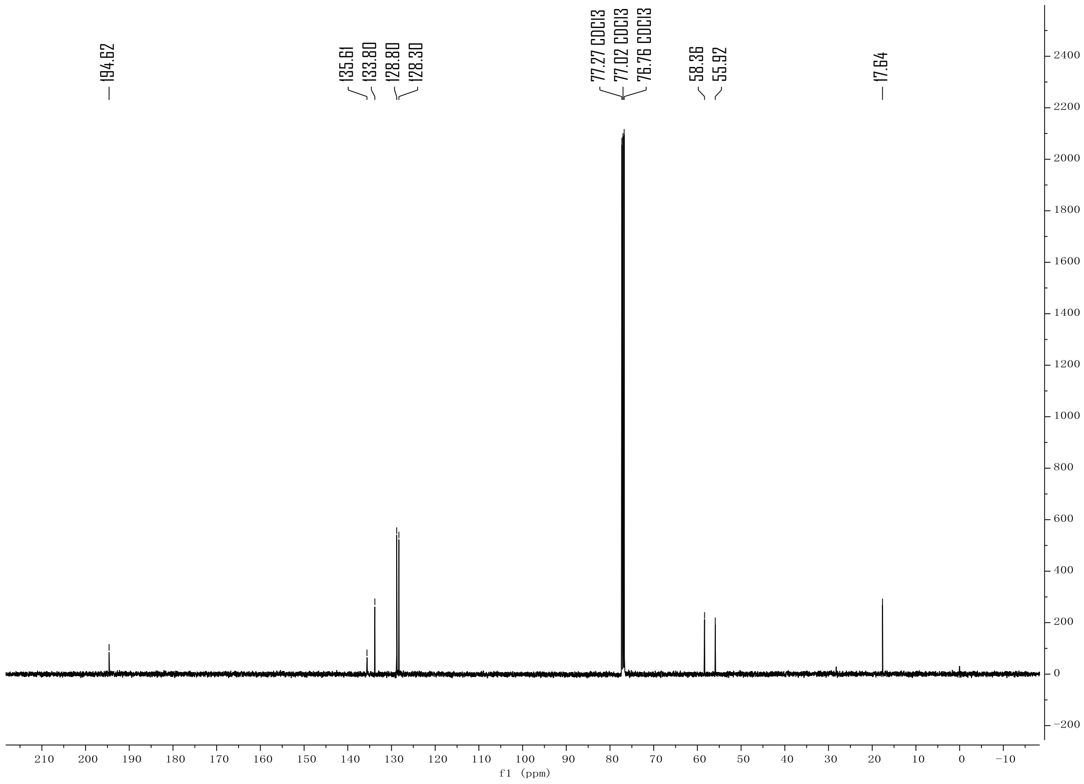


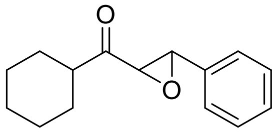

191.71
140.74
135.09
133.82
133.62
129.81
129.30
129.08
127.11
77.28 CDCl3
77.03 CDCl3
76.77 CDCl3
61.00
58.66
550
500
450
400
350
300
250
200
150
100
50
0
-50
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)



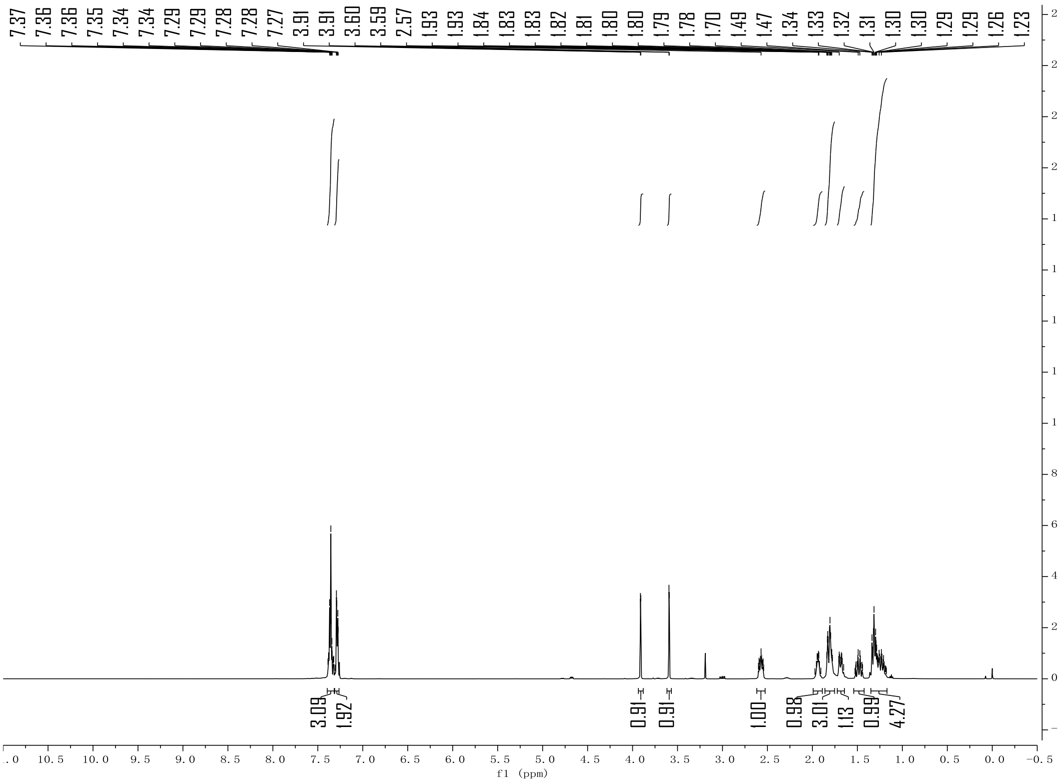
(3-methyloxiran-2-yl)(phenyl)methanone (**b20**)

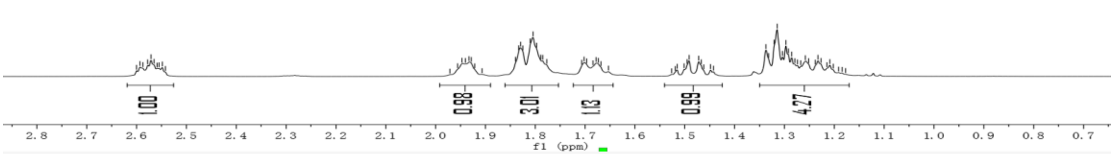


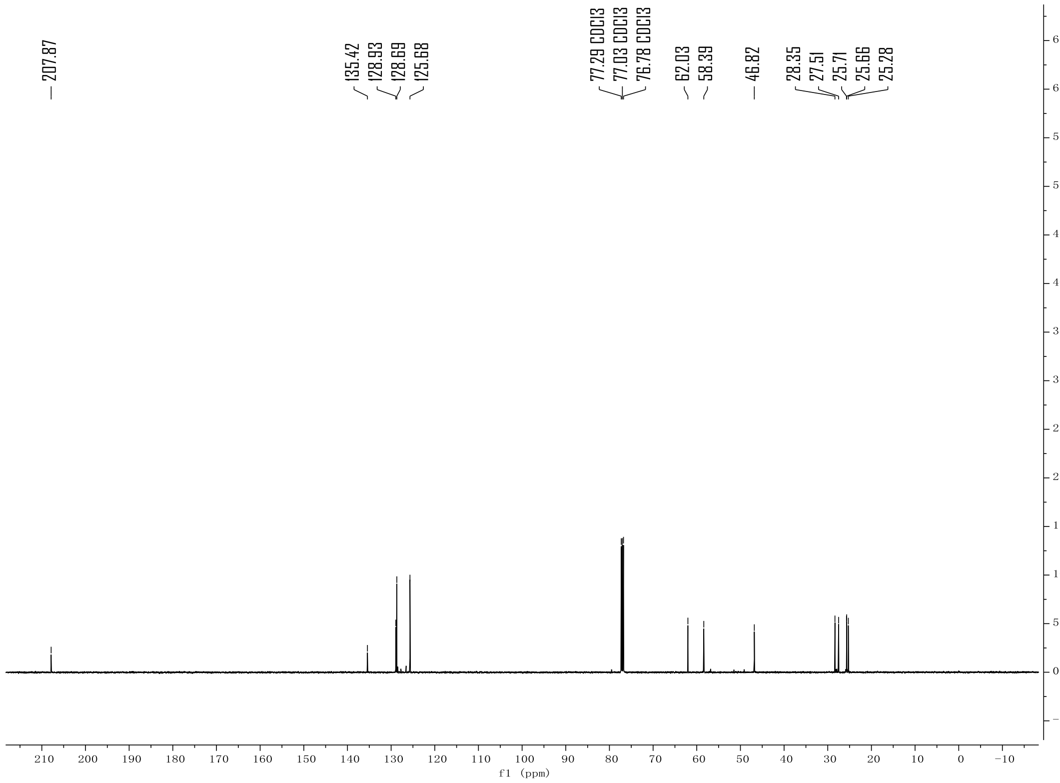

194.62
135.61
133.80
128.80
128.30
77.27 CDCl3
77.02 CDCl3
76.76 CDCl3
58.36
55.92
17.64
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
0
-200
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

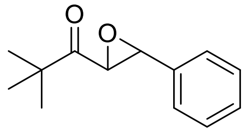


cyclohexyl(3-phenyloxiran-2-yl)methanone (**b21**)

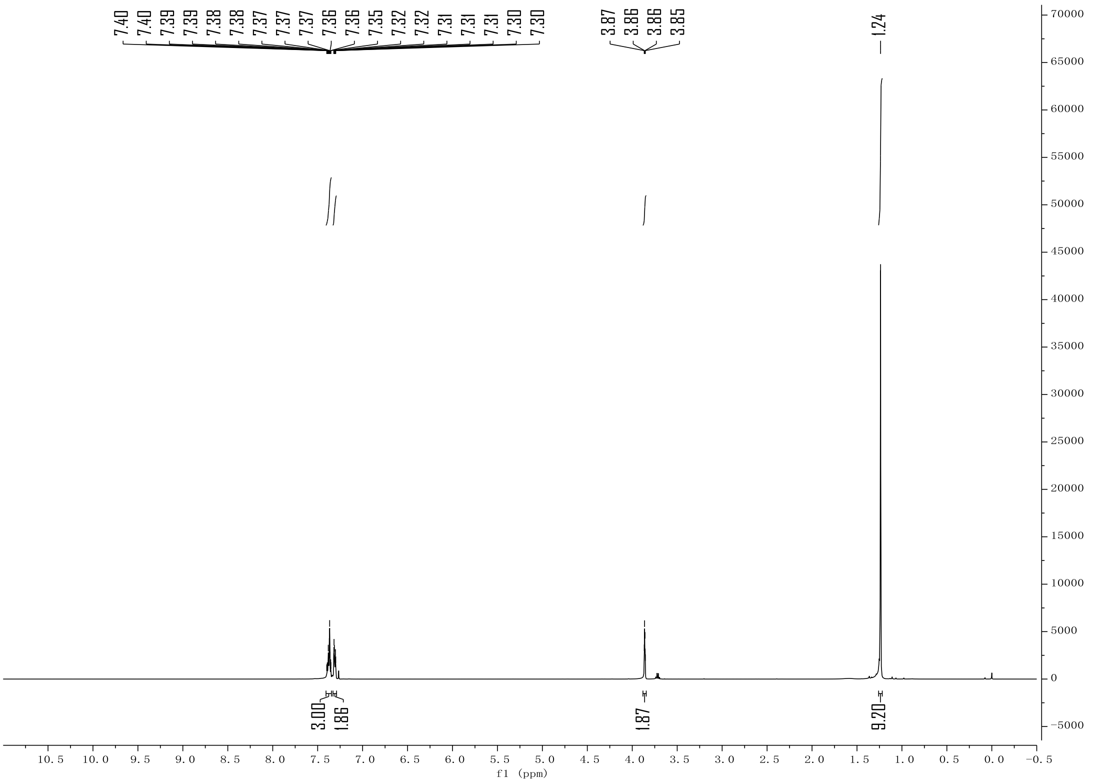


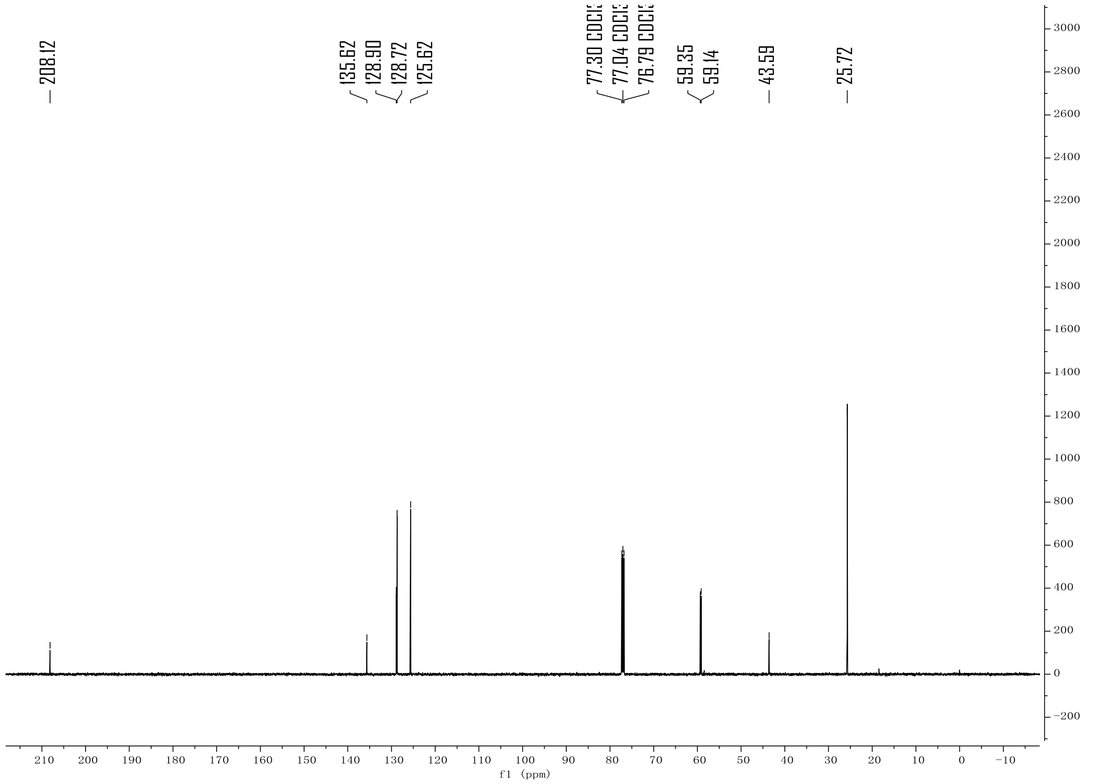



207.87
135.42
128.93
128.69
125.68
77.29 CDCl3
77.03 CDCl3
76.78 CDCl3
62.03
58.39
46.82
28.35
27.51
25.71
25.66
25.28
6500
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
−500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)

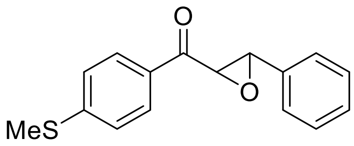


2,2-dimethyl-1-(3-phenyloxiran-2-yl)propan-1-one (**b22**)

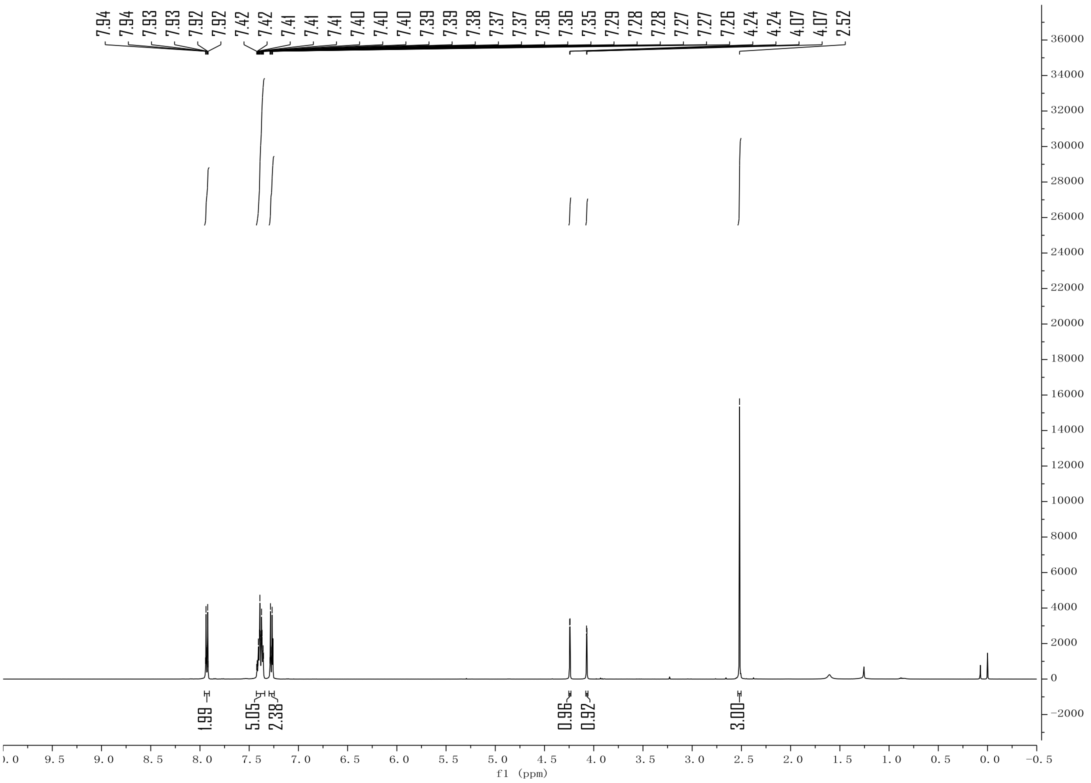


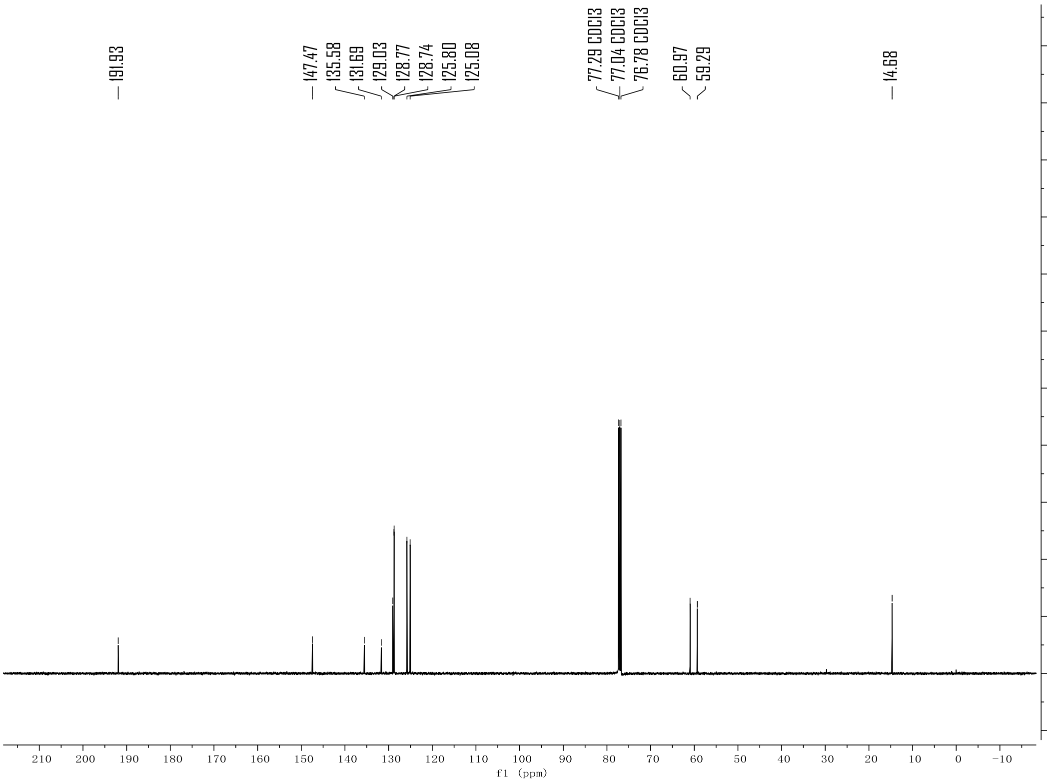


208.12
135.62
128.90
128.72
125.62
77.30 CDCl3
77.04 CDCl3
76.79 CDCl3
59.35
59.14
43.59
25.72
3000
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
0
−200
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 −10
f1 (ppm)

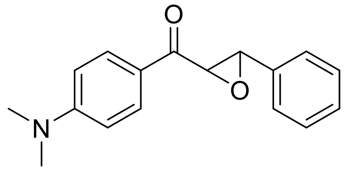


(4-(methylthio)phenyl)(3-phenyloxiran-2-yl)methanone (**b23**)

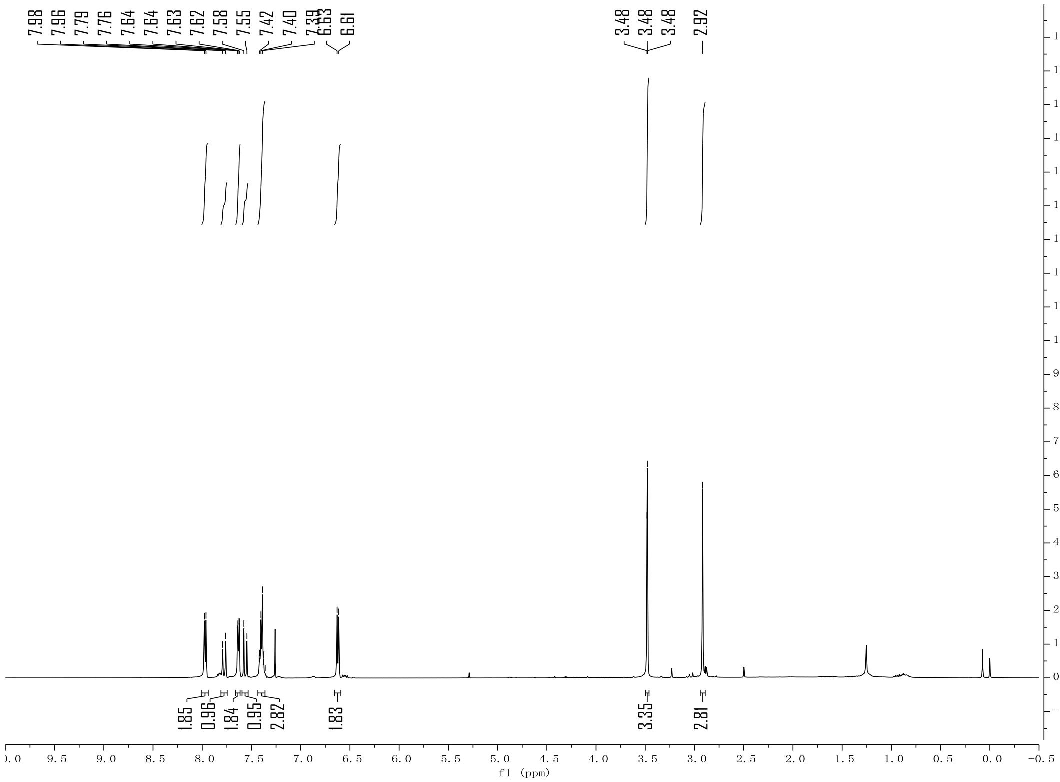


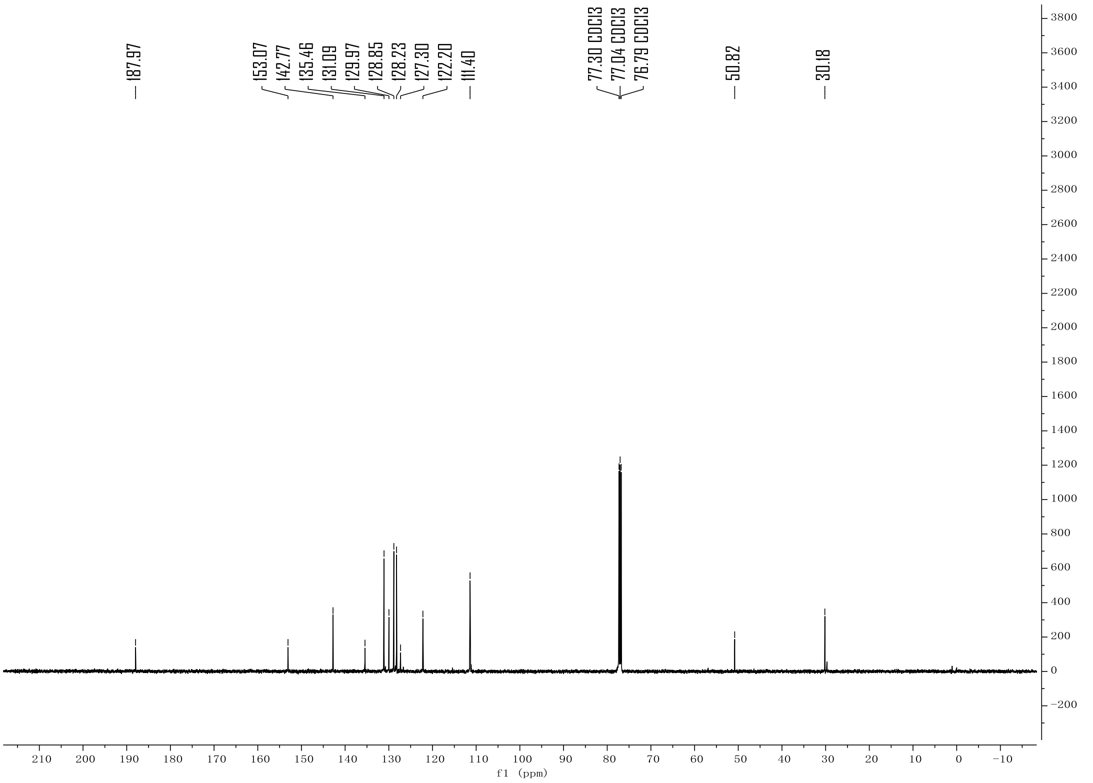


191.93
147.47
135.58
131.69
129.03
128.77
128.74
125.80
125.08
77.29 CDCl3
77.04 CDCl3
76.78 CDCl3
60.97
59.29
14.68
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

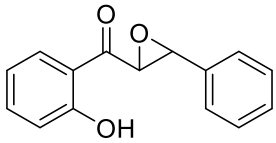


(4-(dimethylamino)phenyl)(3-phenyloxiran-2-yl)methanone (**b24**)

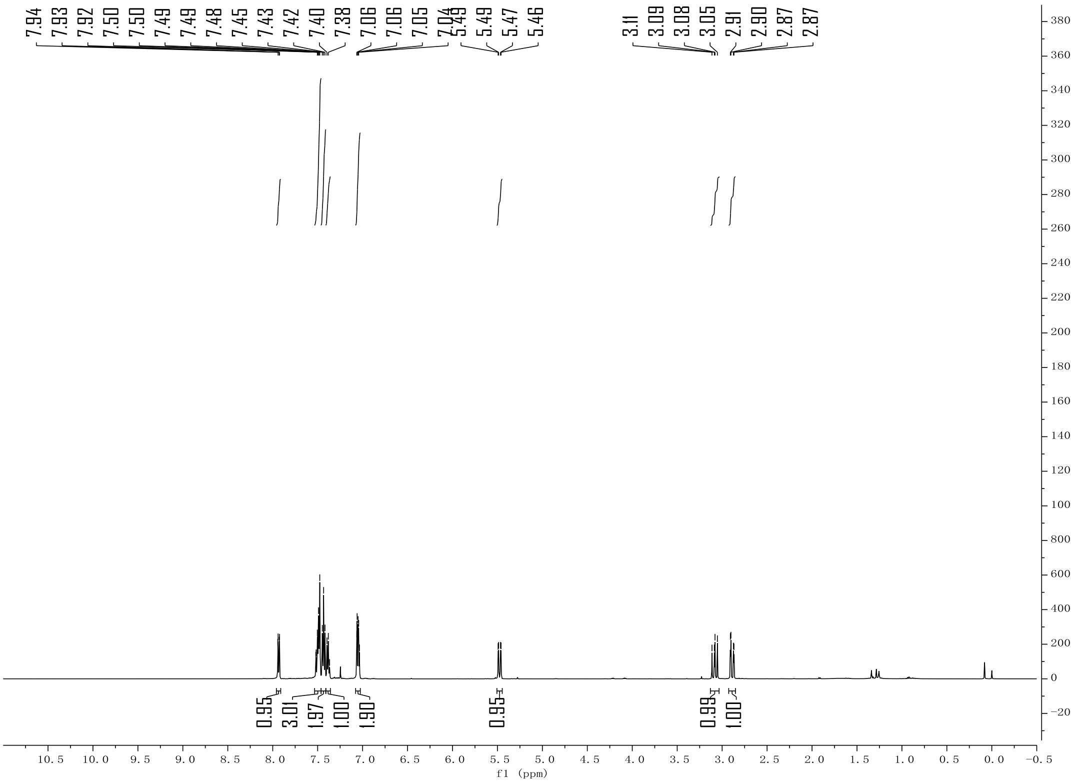


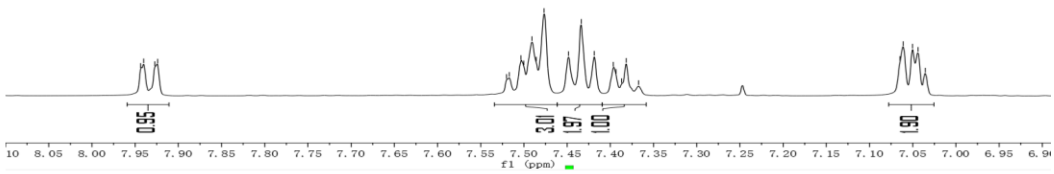


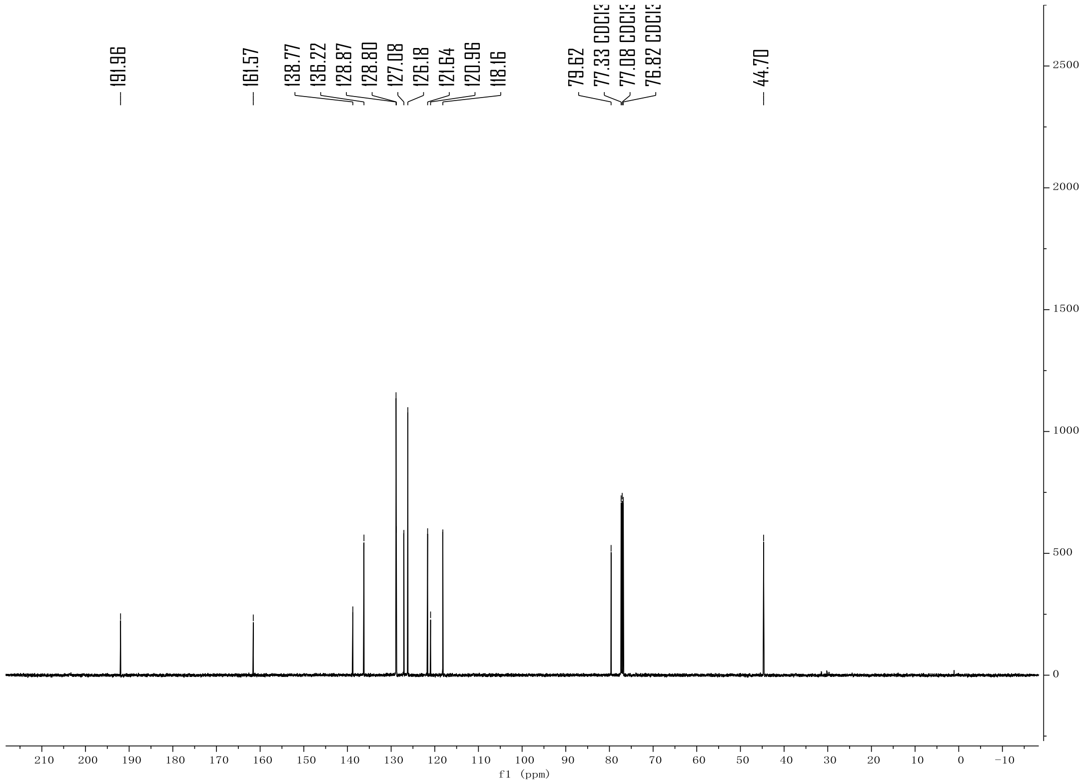
187.97
153.07
142.77
135.46
131.09
129.97
128.85
128.23
127.30
122.20
111.40
77.30 CDCl3
77.04 CDCl3
76.79 CDCl3
50.82
30.18
3800
3600
3400
3200
3000
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
400
200
0
−200
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
−10
f1 (ppm)

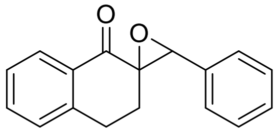


(2-hydroxyphenyl)(3-phenyloxiran-2-yl)methanone (**b25**)

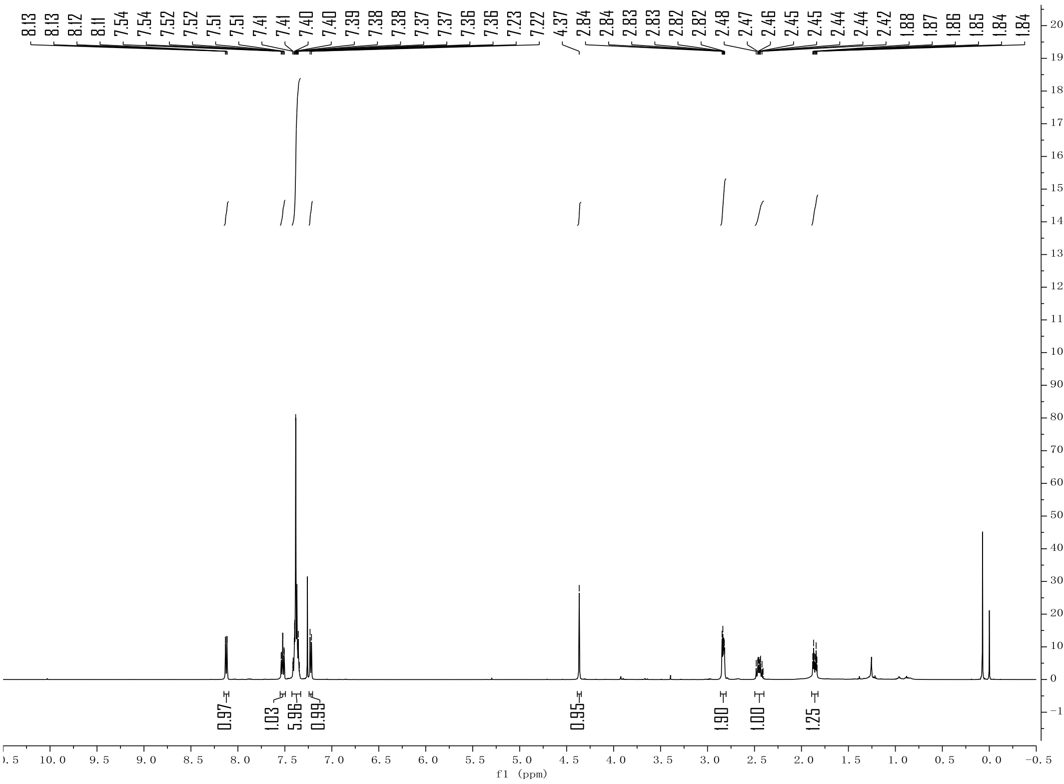


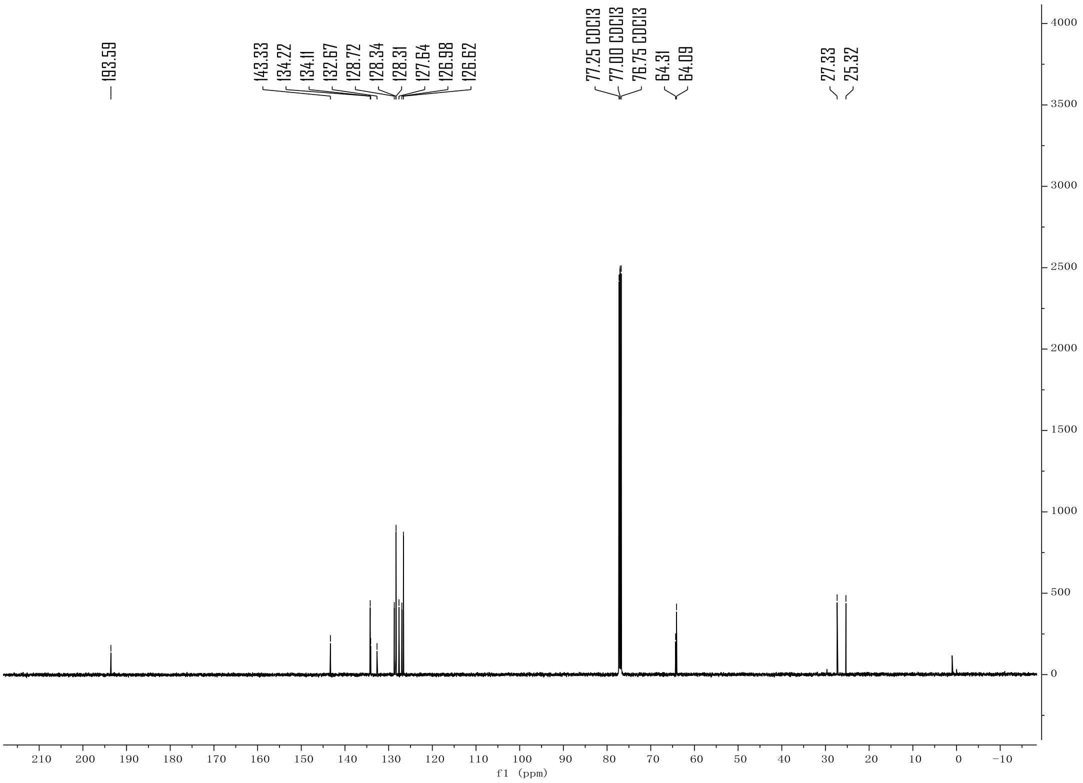



191.96
161.57
138.77
136.22
128.87
128.80
127.08
126.18
121.64
120.96
118.16
79.62
77.33 CDCl3
77.08 CDCl3
76.82 CDCl3
44.70
2500
2000
1500
1000
500
0
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)



3'-phenyl-3,4-dihydro-1H-spiro[naphthalene-2,2'-oxiran]-1-one (**b26**)

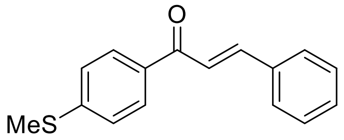



193.59
143.33
134.22
134.11
132.67
128.72
128.34
128.31
127.64
126.98
126.62
77.25 CDCl3
77.00 CDCl3
76.75 CDCl3
64.31
64.09
27.33
25.32
4000
3500
3000
2500
2000
1500
1000
500
0
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

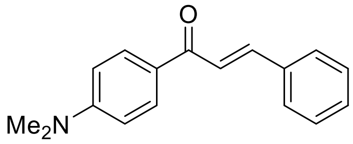
8. Supplementary Information of Reaction Scope

In the case of **a23** which contained S-group, except for epoxide **b23**, other two minor unknown compounds were also observed on TLC. However, they disappeared from TLC very soon. They were unlikely to be the products from the oxidation of S, such as sulfone or sulfoxide, because they were highly volatile. For the reaction of **a24**, the corresponding epoxide **b24** was formed as the major product on TLC, but being accompanied by a series of minor impurities which were too difficult to separate. Maybe they were the byproducts from the oxidation of N-group, however, much more efforts were needed to characterize those minor byproducts by other ways in the future.

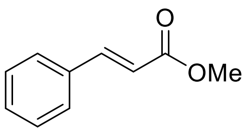
Meanwhile, other selected α,β-unsaturated substrates were also tested in this catalytic system (see the structures below). The hydrolysis of the ester **aa1** or the amide **aa2** taken place under the alkaline condition to give the corresponding acids, however, no desired epoxides were found after reaction. For the reaction of 1-phenylprop-2-en-1-one (**aa3**), the substrate was quite unstable and became to a mixture of several unknown substances by TLC. In the case of 4-phenylbut-3-en-2-one (**aa4**), no epoxidation occurred in spite of several attempts by adjusting the reaction conditions.



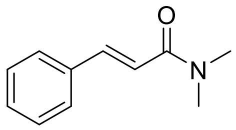
a23



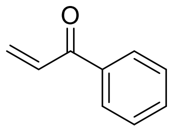
a24



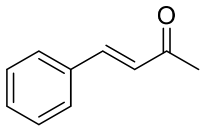
aa1



aa2



aa3



aa4

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