Supporting Information

Electrochemical trifluoromethylation/cyclization for the synthesis of isoquinoline-1,3-diones and oxindoles

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Table of Contents

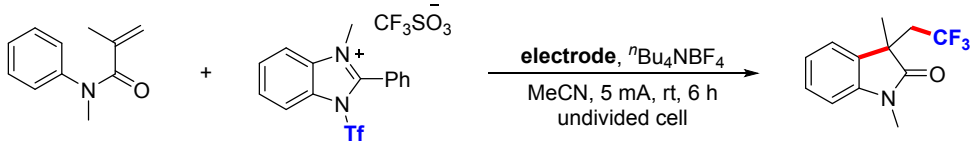
1. General information

Reagents were purchased from commercial sources and were used as received. ^{1}H and ^{13}C Nuclear Magnetic Resonance (NMR) spectra were recorded on Bruker Avance 400 Ultrashield NMR spectrometers. ^{19}F NMR spectra were recorded on a Varian 400 instrument spectrometer. Chemica shifts (δ) were given in parts per million (ppm) and were measured downfield from internal tetramethylsilane. High-resolution mass spectrometry (HRMS) data were obtained on an FTICR-MS instrument (Ionspec 7.0 T). The melting points were determined on an X-4 microscope melting point apparatus and are uncorrected. Conversion was monitored by thin layer chromatography (TLC). Flash column chromatography was performed over silica gel (100-200 mesh).

The starting material **1** was synthesized as reported by Pan[1]. The starting materials **2** were synthesized as reported also by Liu[2]. The starting materials **3** were synthesized as reported by Nevado[3].

2. Investigation of the key reaction parameters.

Table S1. Screening of electrode.[a]

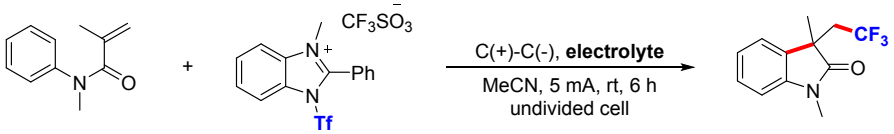


2a **1** **4a**

Entry	electrode	Yield (%)[b]
1	C(+)-C(-)	44
2	Pt(+)-C(-)	trace
3	C(+)-Pt(-)	trace

[a]Reaction conditions: **2a** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv), MeCN (6 mL), undivided cell with two **electrodes** (each 1.0 × 1.0 cm^2) 5 mA cm^{-2}, at room temperature under argon atmosphere for 6 h. [b]Yields were determined by ^{19}F NMR spectroscopy using fluorobenzene as an internal standard.

Table S2. Screening of electrolyte.[a]

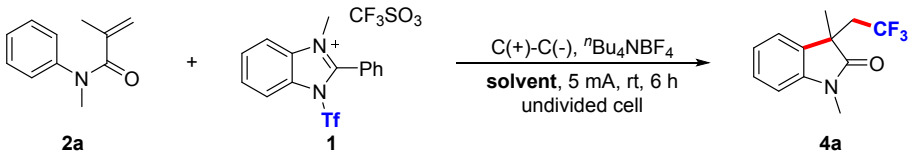


2a **1** **4a**

Entry	electrolyte	Yield (%)[b]
1	$^{n}Bu_4NBF_4$	44
2	$^{n}Bu_4NPF_6$	40
3	$^{n}Bu_4NClO_4$	36
4	$^{n}Bu_4NOAc$	12
5	$^{n}Bu_4NI$	trace
6	$^{n}Bu_4NBr$	trace

[a]Reaction conditions: **2a** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), **electrolyte** (0.3 mmol, 1 equiv), MeCN (6 mL), undivided cell with two graphite electrodes (each 1.0 × 1.0 cm^2) 5 mA cm^{-2}, at room temperature under argon atmosphere for 6 h. [b]Yields were determined by ^{19}F NMR spectroscopy using fluorobenzene as an internal standard.

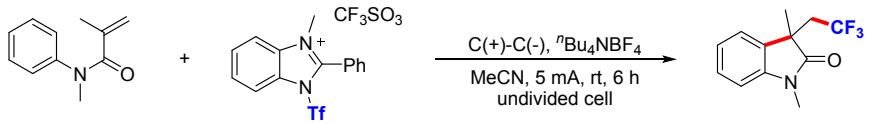
Table S3. Screening of solvent.[a]



Entry	Solvent	Yield (%)[b]
1	THF	0
2	DMF	0
3	Acetone	34
4	MeCN	44
5	DCE	trace
6	EA	trace

[a]Reaction conditions: **2a** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv), **solvent** (6 mL), undivided cell with two graphite electrodes (each 1.0 × 1.0 cm^2) 5 mA cm^{-2}, at room temperature under argon atmosphere for 6 h. [b]Yields were determined by ^{19}F NMR spectroscopy using fluorobenzene as an internal standard.

Table S4. Screening of reaction concentration.[a]

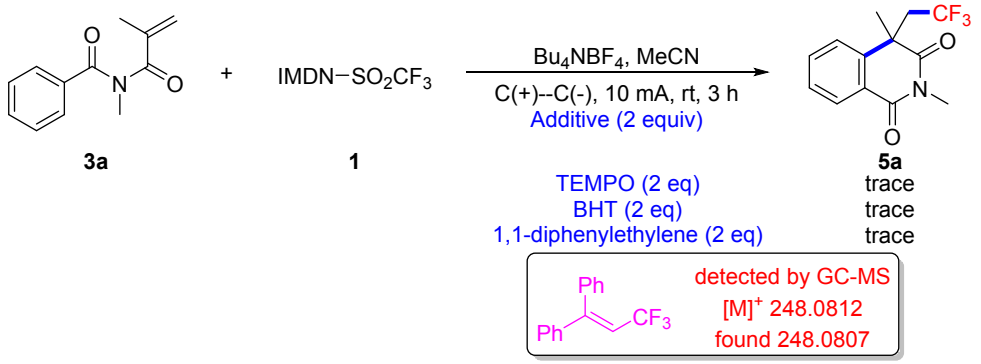


Entry	concentration	Yield (%)[b]
1	0.05 M	44
2	0.1 M	43
3	0.025	42

[a]Reaction conditions: **2a** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv), **MeCN** (6 mL or 3 mL or 12 mL), undivided cell with two graphite electrodes (each 1.0 × 1.0 cm^2) 5 mA cm^{-2}, at room temperature under argon atmosphere for 6 h. [b]Yields were determined by ^{19}F NMR spectroscopy using fluorobenzene as an internal standard.

3. Investigation of the mechamism.

3.1 Radical inhibition experiment



Scheme S1

To a 15 mL glass vial was equipped with two graphite sheet electrodes (10 mm × 10 mm × 3 mm). Then **3a** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv) and additive (TEMPO (93.8 mg, 0.6 mmol) or BHT (132 mg, 0.6 mmol) or 1,1-diphenylethylene (108 mg, 0.6 mmol)) were added to this

undivided cell. After replacing the reaction device with an argon atmosphere, 6 mL of acetonitrile was added. The reaction mixture was stirred and electrolyzed at a constant current of 10 mA under room temperature for 3 h. Then monitor the reaction and capture intermediates by thin layer chromatography (TLC) and High-resolution mass spectrometry (HRMS), respectively.

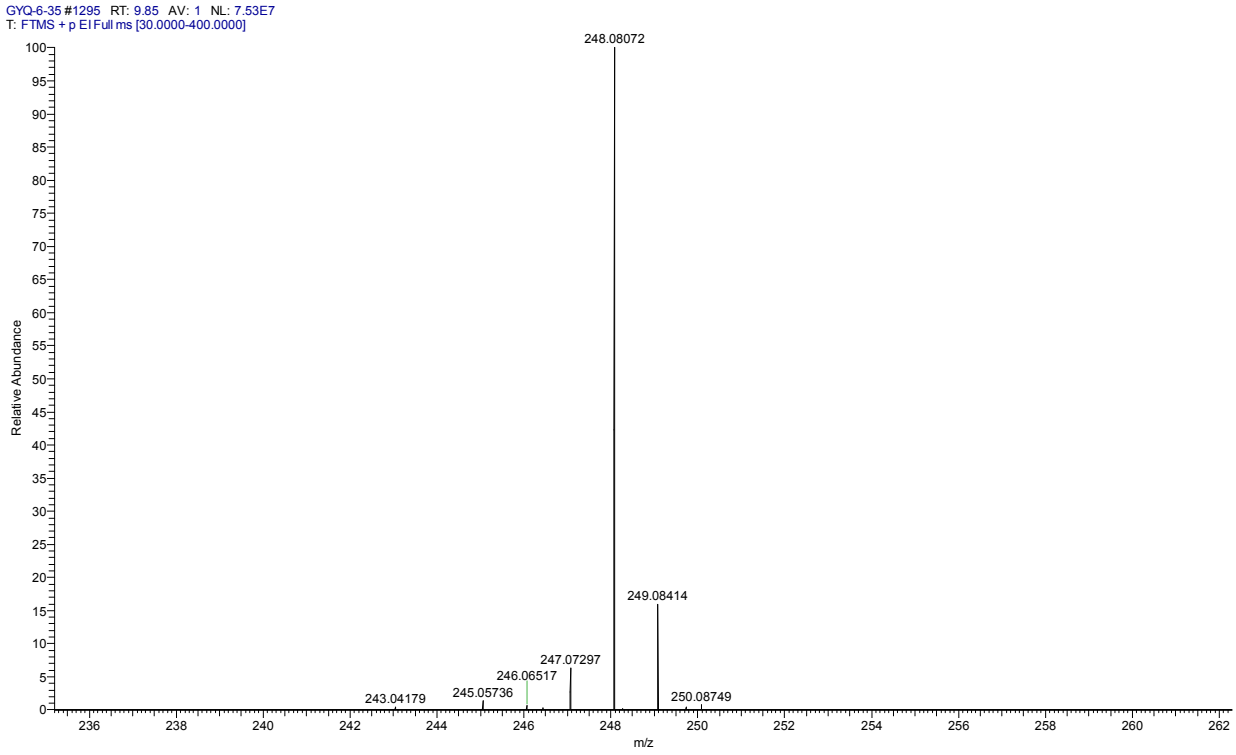
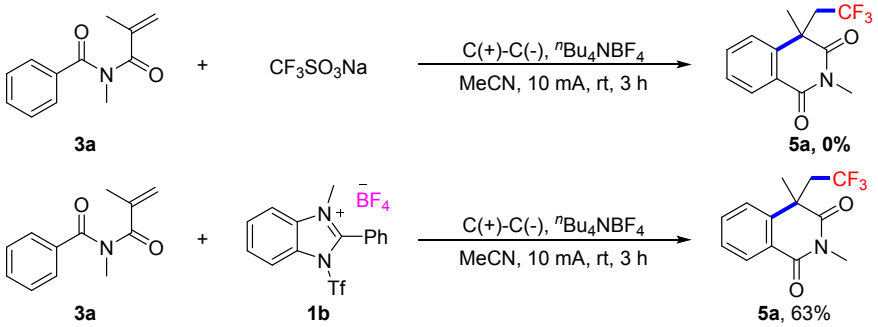


Figure S1 High resolution mass spectrum of 1,1-diphenylethylene capture product.

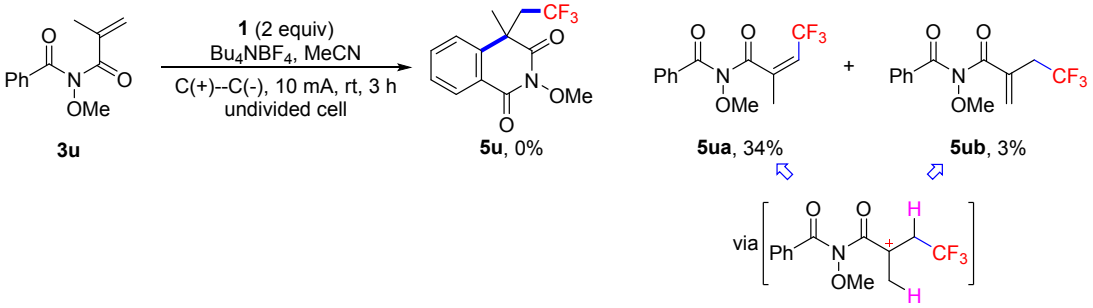
3.2 Exploration experiment on the source of trifluoromethyl radical



Scheme S2

When using CF_3SO_3Na as the trifluoromethyl reagent, **5a** was not obtained. Meanwhile, when **1b** was used as the trifluoromethyl reagent, **5a** can be obtained smoothly (Scheme S2). This result indicates that the trifluoromethyl radical is derived from the benzimidazole cation part of reagent **1**.

3.3 Anodizing verification experiment



Scheme S3

To a 15 mL glass vial was equipped with two graphite sheet electrodes (10 mm × 10 mm × 3 mm). Then **3u** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv) were added to this undivided cell. After replacing the reaction device with an argon atmosphere, 6 mL of acetonitrile was added. The reaction mixture was stirred and electrolyzed at a constant current of 10 mA under room temperature for 3 h. The isolated yield is given.

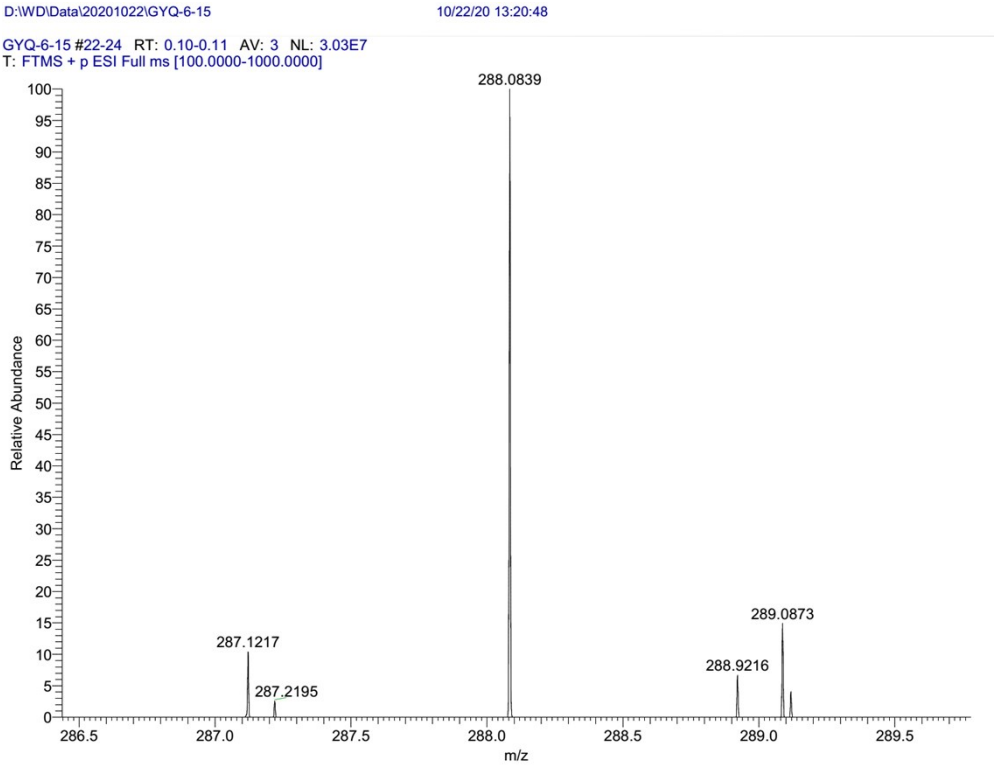
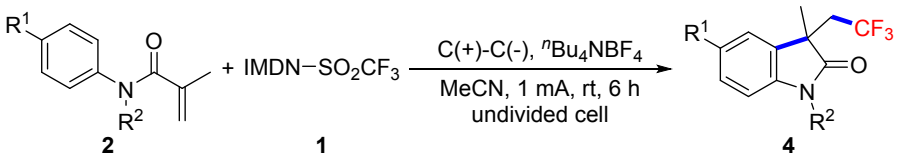


Figure S2 High resolution mass spectrum of [**5ub**+**H**]

4. Experimental procedures and product characterization.

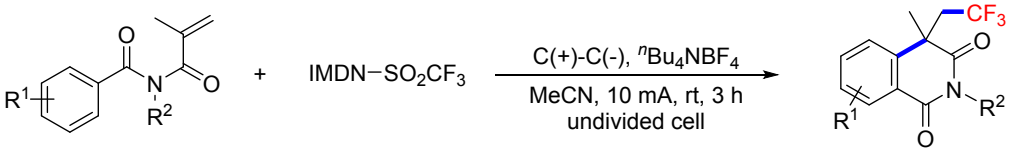
4.1 General procedure for the synthesis of oxindoles:



To a 15 mL glass vial was equipped with two graphite sheet electrodes (10 mm × 10 mm × 3 mm). Then **2** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv) were added to this undivided cell. After replacing the reaction device with an argon atmosphere, 6 mL of acetonitrile was added. The reaction

mixture was stirred and electrolyzed at a constant current of 1 mA under room temperature for 6 h. When the reaction is completed, extracted with ethyl acetate, washed with brine, dried over anhydrous sodium sulfate, concentrated in vacuo, and purified by column chromatography (hexane/ethyl acetate) to afford the corresponding target compounds.

4.2 General procedure for the synthesis of isoquinoline-1,3-diones:



To a 15 mL glass vial was equipped with two graphite sheet electrodes (10 mm × 10 mm × 3 mm). Then **3** (0.3 mmol, 1 equiv), **1** (0.6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (0.3 mmol, 1 equiv) were added to this undivided cell. After replacing the reaction device with an argon atmosphere, 6 mL of acetonitrile was added. The reaction mixture was stirred and electrolyzed at a constant current of 10 mA under room temperature for 3 h. When the reaction is completed, extracted with ethyl acetate, washed with brine, dried over anhydrous sodium sulfate, concentrated in vacuo, and purified by column chromatography (hexane/ethyl acetate) to afford the corresponding target compounds.

#Note:

Substrate containing methoxy group on the benzene ring cannot successfully obtained the target product. Only dearomatized product can be obtained, but it is inseparable from unknown impurities and no pure dearomatized product can be obtained.

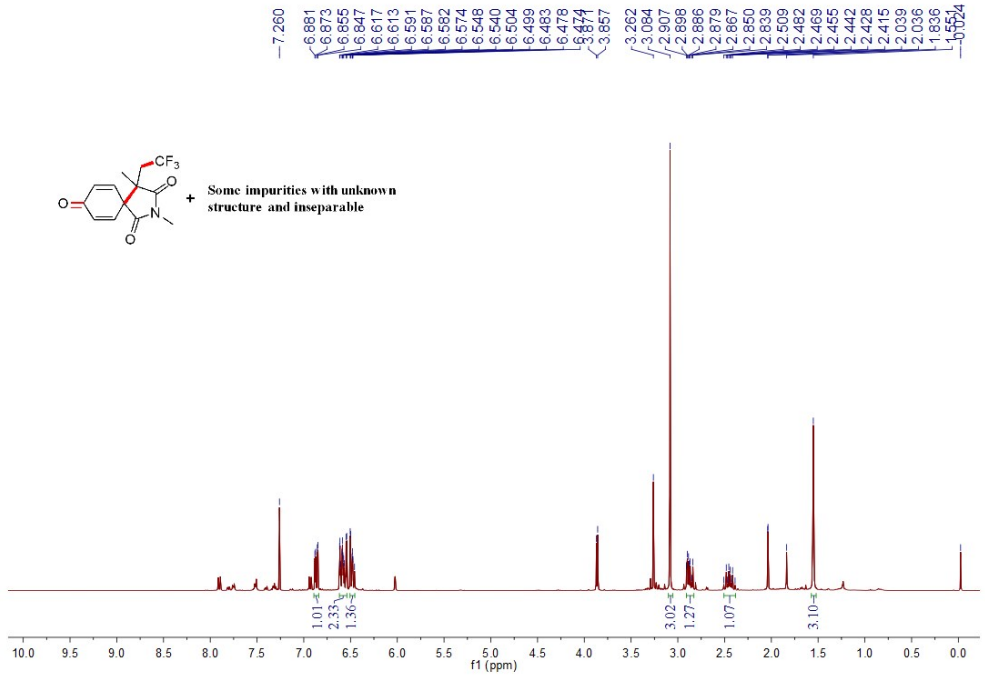


Figure S3 ^{1}H NMR spectrum (400 MHz, $CDCl_3$) of dearomatized product

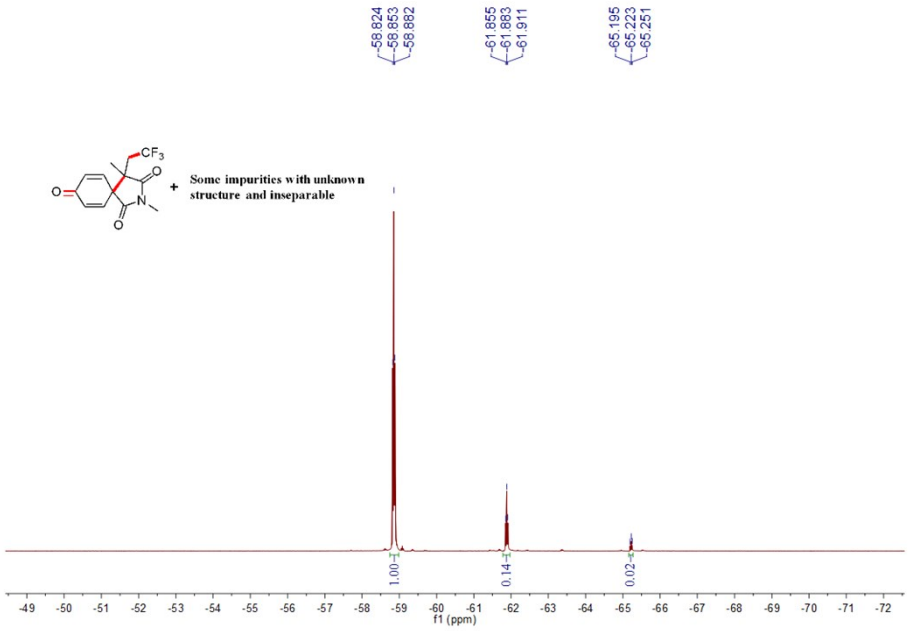
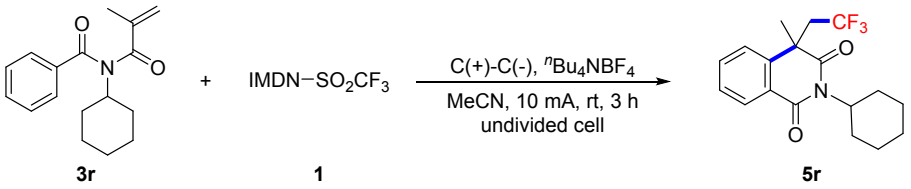


Figure S4 ^{19}F NMR spectrum (376 MHz, $CDCl_3$) of dearomatized product

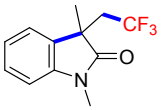
4.3 Gram scale



To a 100 mL glass vial was equipped with two graphite sheet electrodes (100 mm × 40 mm × 3 mm). Then **3r** (813 mg, 3 mmol, 1 equiv), **1** (2940 mg, 6 mmol, 2 equiv), $^{n}Bu_4NBF_4$ (988 mg, 3 mmol, 1 equiv) were added to this undivided cell. After replacing the reaction device with an argon atmosphere, 60 mL of acetonitrile was added. The reaction mixture was stirred and electrolyzed at a constant current of 20 mA under room temperature for 16 h. When the reaction is completed, extracted with ethyl acetate, washed with brine, dried over anhydrous sodium sulfate, concentrated in vacuo, and purified by column chromatography (hexane/ethyl acetate from 20/1 to 10/1) to afford **5r** (550 mg, 54%).

4.4 Product Characterization

1,3-dimethyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4a**)



Colorless oil, yield 67% (48.9 mg).

R_f 0.50 (Petroleum ether/EtOAc, 5/1).

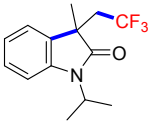
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (t, J = 7.7 Hz, 1H), 7.28 – 7.25 (m, 1H), 7.09 (t, J = 7.5 Hz, 1H), 6.88 (d, J = 7.8 Hz, 1H), 3.23 (s, 3H), 2.82 (dq, J = 15.1, 10.7 Hz, 1H), 2.65 (dq, J = 15.1, 10.5 Hz, 1H), 1.40 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 178.5, 142.9, 131.0, 128.5, 125.3 (q, J = 276.6 Hz), 123.5, 122.6, 108.4, 44.4 (q, J = 1.7 Hz), 40.6 (q, J = 28.2 Hz), 26.4, 25.0.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.94 (t, J = 10.5 Hz, 3F).

HRMS (ESI) calcd for $C_{12}H_{13}F_3NO$ $[M+H]^+$ 244.0949, found, 244.0942.

1-isopropyl-3-methyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4b**)



Colorless oil, yield 45% (36.6 mg).

R_f 0.70 (Petroleum ether/EtOAc, 5/1).

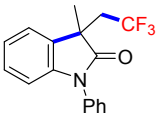
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29 – 7.24 (m, 2H), 7.10 – 7.00 (m, 2H), 4.68 – 4.58 (m, 1H), 2.90 – 2.76 (m, 1H), 2.69 – 2.54 (m, 1H), 1.52 – 1.43 (m, 6H), 1.38 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 141.6, 131.4, 128.2, 125.2 (q, J = 276.7 Hz), 123.7, 122.0, 110.1, 44.1 (q, J = 1.9 Hz), 44.0, 40.8 (q, J = 28.0 Hz), 25.3, 19.2, 19.0.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.97 (t, J = 10.5 Hz, 3F).

HRMS (ESI) calcd for $C_{14}H_{17}F_3NO$ $[M+H]^+$ 272.1262, found, 272.1254.

3-methyl-1-phenyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4c**)



Colorless oil, yield 65% (61.1 mg).

R_f 0.55 (Petroleum ether/EtOAc, 10/1).

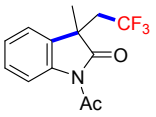
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.53 (t, J = 7.7 Hz, 2H), 7.46 – 7.36 (m, 3H), 7.32 (d, J = 7.4 Hz, 1H), 7.23 (dd, J = 7.8, 1.2 Hz, 1H), 7.13 (td, J = 7.5, 0.9 Hz, 1H), 6.84 (d, J = 7.9 Hz, 1H), 2.97 (dq, J = 15.1, 10.7 Hz, 1H), 2.73 (dq, J = 15.1, 10.4 Hz, 1H), 1.54 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 178.0, 143.0, 134.3, 130.7, 129.7, 128.5, 128.3, 126.6, 123.8, 123.1, 109.8, 125.3 (q, J = 276.5 Hz), 44.5 (q, J = 2.0 Hz), 41.1 (q, J = 28.1 Hz), 25.5.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.86 (t, J = 10.2 Hz, 3F).

HRMS (ESI) calcd for $C_{17}H_{15}F_3NO$ $[M+H]^+$ 306.1106, found, 306.1099.

1-acetyl-3-methyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4d**)



Colorless oil, yield 46% (37.4 mg).

R_f 0.65 (Petroleum ether/EtOAc, 10/1).

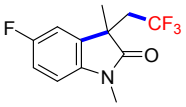
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (d, J = 8.2 Hz, 1H), 7.39 – 7.32 (m, 1H), 7.24 (dd, J = 12.3, 4.4 Hz, 2H), 2.98 – 2.85 (m, 1H), 2.75 – 2.63 (m, 4H), 1.48 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 179.5, 170.9, 139.1, 129.8, 128.9, 125.3, 124.9 (q, J = 276.4 Hz), 123.0, 116.9, 45.0 (q, J = 2.0 Hz), 41.4 (q, J = 28.0 Hz), 26.6, 26.4.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.95 (t, J = 10.2 Hz, 3F).

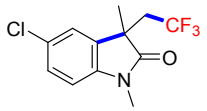
HRMS (ESI) calcd for $C_{13}H_{13}F_3NO_2$ $[M+H]^+$ 272.0898, found, 272.0889.

5-fluoro-1,3-dimethyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4e**)



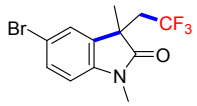
Colorless oil, yield 41% (32.4 mg).
R_f 0.45 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.04 – 7.00 (m, 2H), 6.80 (dd, J = 9.1, 4.0 Hz, 1H), 3.23 (s, 3H), 2.89 – 2.77 (m, 1H), 2.69 – 2.57 (m, 1H), 1.40 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 159.3 (d, J = 239.5 Hz), 138.8, 132.6 (d, J = 8.0 Hz), 125.1 (q, J = 276.4 Hz), 114.8 (d, J = 23.4 Hz), 111.8 (dd, J = 23.6, 1.2 Hz), 109.0 (d, J = 8.2 Hz), 44.8, 40.5 (q, J = 28.3 Hz), 26.6, 24.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.97 (t, J = 10.9 Hz, 3F), -120.32 (m, 1F).
HRMS (ESI) calcd for $C_{12}H_{12}F_4NO$ $[M+H]^+$ 262.0855, found, 262.0848.

5-chloro-1,3-dimethyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4f**)



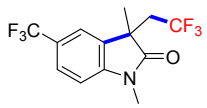
Colorless oil, yield 40% (33.1 mg).
R_f 0.55 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29 (dd, J = 8.3, 2.1 Hz, 1H), 7.24 (d, J = 1.9 Hz, 1H), 6.81 (d, J = 8.3 Hz, 1H), 3.22 (s, 3H), 2.83 (dq, J = 15.2, 10.6 Hz, 1H), 2.68 – 2.57 (m, 1H), 1.40 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 177.9, 141.5, 132.7, 128.5, 128.1, 125.1 (q, J = 276.5 Hz), 124.1, 109.4, 44.6 (q, J = 1.9 Hz), 40.5 (q, J = 28.4 Hz), 26.6, 24.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.96 (t, J = 10.5 Hz, 3F).
HRMS (ESI) calcd for $C_{12}H_{12}ClF_3NO$ $[M+H]^+$ 278.0560, found, 278.0552.

5-bromo-1,3-dimethyl-3-(2,2,2-trifluoroethyl)indolin-2-one (**4g**)



Colorless oil, yield 50% (48.2 mg).
R_f 0.50 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44 (dd, J = 8.3, 1.9 Hz, 1H), 7.37 (d, J = 1.8 Hz, 1H), 6.76 (d, J = 8.3 Hz, 1H), 3.22 (s, 3H), 2.82 (dq, J = 15.2, 10.6 Hz, 1H), 2.62 (dq, J = 15.2, 10.4 Hz, 1H), 1.40 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 177.8, 141.9, 133.1, 131.4, 126.8, 115.3, 109.9, 44.5 (q, J = 1.7 Hz), 40.6 (q, J = 28.3 Hz), 26.5, 24.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.95 (t, J = 10.5 Hz, 3F).
HRMS (ESI) calcd for $C_{12}H_{12}BrF_3NO$ $[M+H]^+$ 322.0054, found, 322.0049.

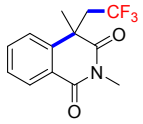
1,3-dimethyl-3-(2,2,2-trifluoroethyl)-5-(trifluoromethyl)indolin-2-one (**4h**)



White solid, yield 41% (38.4 mg). M.p. = 90 – 93 °C.
R_f 0.50 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.61 (d, J = 8.1 Hz, 1H), 7.49 (s, 1H), 6.96 (d, J = 8.2 Hz, 1H), 3.27 (s, 3H), 2.93 – 2.80 (m, 1H), 2.74 – 2.61 (m, 1H), 1.44 (s, 3H).

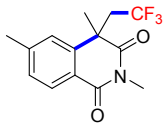
^{13}C NMR (100 MHz, $CDCl_3$) δ 177.3, 144.8, 130.5, 125.3 (q, J = 4.0 Hz), 124.0 (q, J = 32.6 Hz), 123.3 (q, J = 269.8 Hz), 122.6, 119.6 (q, J = 1.8 Hz), 107.3, 43.3 (q, J = 1.7 Hz), 39.5 (q, J = 28.3 Hz), 25.6, 23.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.51 (s, 3F), -62.07 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{12}F_6NO$ $[M+H]^+$ 312.0823, found, 312.0814.

2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5a**)



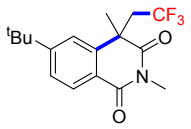
Colorless oil, yield 78% (63.4 mg).
R_f 0.40 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 (dd, J = 7.9, 0.8 Hz, 1H), 7.66 (td, J = 7.9, 1.2 Hz, 1H), 7.48 (t, J = 7.4 Hz, 1H), 7.42 (d, J = 7.9 Hz, 1H), 3.40 (s, 3H), 3.38 – 3.26 (m, 1H), 2.80 (dq, J = 15.1, 9.7 Hz, 1H), 1.65 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.5, 162.7, 139.4, 132.8, 128.3, 127.0, 124.6, 123.9 (q, J = 277.0 Hz), 123.2, 43.3 (q, J = 27.3 Hz), 42.6 (q, J = 1.8 Hz), 30.1, 26.4.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.65 (t, J = 10.0 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{13}F_3NO_2$ $[M+H]^+$ 272.0898, found, 272.0891.

2,4,6-trimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5b**)



Colorless oil, yield 67% (57.7 mg).
R_f 0.35 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 (d, J = 8.1 Hz, 1H), 7.28 (d, J = 8.4 Hz, 1H), 7.19 (s, 1H), 3.43 – 3.27 (m, 4H), 2.78 (dq, J = 15.0, 9.7 Hz, 1H), 2.46 (s, 3H), 1.64 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.7, 162.7, 143.7, 139.4, 128.3, 128.1, 125.0, 123.9 (q, J = 277.2 Hz), 120.7, 43.3 (q, J = 27.3 Hz), 42.6 (q, J = 1.7 Hz), 30.1, 26.3, 20.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.64 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{14}H_{15}F_3NO_2$ $[M+H]^+$ 286.1055, found, 286.1047.

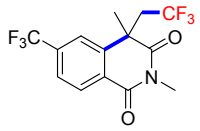
6-(tert-butyl)-2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5c**)



White solid, yield 57% (55.6 mg). M.p. = 93 – 95 °C.
R_f 0.40 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 (s, 1H), 7.68 (d, J = 8.3 Hz, 1H), 7.34 (d, J = 8.3 Hz, 1H), 3.42 – 3.26 (m, 4H), 2.84 – 2.72 (m, 1H), 1.63 (s, 3H), 1.37 (s, 9H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.8, 163.1, 150.3, 136.4, 130.2, 124.8, 124.4, 123.9 (q, J = 277.1 Hz), 122.7, 43.3 (q, J = 27.4 Hz), 42.6 (q, J = 1.7 Hz), 33.8, 30.2, 30.1, 26.4.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.60 (t, J = 10.2 Hz, 3F).

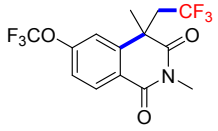
HRMS (ESI) calcd for $C_{17}H_{21}F_3NO_2$ $[M+H]^+$ 328.1524, found, 328.1516.

2,4-dimethyl-4-(2,2,2-trifluoroethyl)-6-(trifluoromethyl)isoquinoline-1,3(2H,4H)-dione (**5d**)



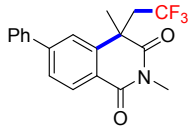
White solid, yield 52% (52.3 mg). M.p. = 87 – 89 °C.
R_f 0.35 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.55 (s, 1H), 7.87 (dd, J = 8.3, 1.6 Hz, 1H), 7.55 (d, J = 8.3 Hz, 1H), 3.46 – 3.30 (m, 4H), 2.80 (dq, J = 15.2, 9.6 Hz, 1H), 1.66 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.7, 162.5, 143.9, 130.9 (q, J = 33.5 Hz), 130.1 (q, J = 3.4 Hz), 126.70 (q, J = 3.3 Hz), 126.69, 125.0, 124.8 (q, J = 277.0 Hz), 123.3 (q, J = 270.9 Hz), 44.2 (q, J = 27.7 Hz), 43.7 (q, J = 2.2 Hz), 31.1, 27.7.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.59 (t, J = 10.2 Hz, 3F), -62.98 (s, 3F).
HRMS (ESI) calcd for $C_{14}H_{11}F_6NNaO_2$ $[M+Na]^+$ 362.0592, found, 362.0590.

2,4-dimethyl-4-(2,2,2-trifluoroethyl)-6-(trifluoromethoxy)isoquinoline-1,3(2H,4H)-dione (**5e**)



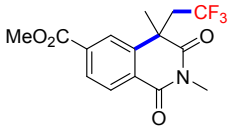
Colorless oil, yield 51% (54.5 mg).
R_f 0.45 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 (s, 1H), 7.55 – 7.43 (m, 2H), 3.40 (s, 3H), 3.39 – 3.29 (m, 1H), 2.78 (dq, J = 15.2, 9.7 Hz, 1H), 1.66 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.0, 162.5, 148.8 (q, J = 1.8 Hz), 138.7, 127.8, 126.3, 126.1, 124.8 (q, J = 276.9 Hz), 120.9, 120.3 (q, J = 257.2 Hz), 44.4 (q, J = 27.6 Hz), 43.4 (q, J = 2.1 Hz), 31.1, 27.6.
^{19}F NMR (376 MHz, $CDCl_3$) δ -57.98 (s, 3F), -61.67 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{14}H_{12}F_6NO_3$ $[M+H]^+$ 356.0721, found, 356.0714.

2,4-dimethyl-6-phenyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5f**)



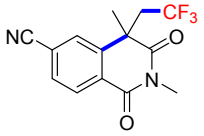
White solid, yield 56% (58.2 mg). M.p. = 133 – 135 °C.
R_f 0.25 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.52 (s, 1H), 7.89 (d, J = 8.2 Hz, 1H), 7.66 (d, J = 8.1 Hz, 2H), 7.57 – 7.45 (m, 3H), 7.41 (t, J = 7.3 Hz, 1H), 3.53 – 3.30 (m, 4H), 2.84 (dq, J = 15.2, 9.6 Hz, 1H), 1.69 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.5, 162.7, 140.0, 138.0, 131.3, 128.0, 127.1, 126.5, 126.1, 125.2, 123.9 (q, J = 277.1 Hz), 123.6, 43.3 (q, J = 27.4 Hz), 42.4 (q, J = 1.5 Hz), 30.2, 26.5.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.52 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{19}H_{17}F_3NO_2$ $[M+H]^+$ 348.1211, found, 348.1201.

methyl 2,4-dimethyl-1,3-dioxo-4-(2,2,2-trifluoroethyl)-1,2,3,4-tetrahydroisoquinoline-6-carboxylate (**5g**)



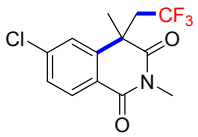
White solid, yield 48% (47.3 mg). M.p. = 91 – 93 °C.
R_f 0.50 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.92 (d, J = 1.7 Hz, 1H), 8.30 (dd, J = 8.3, 1.8 Hz, 1H), 7.52 (d, J = 8.3 Hz, 1H), 3.96 (s, 3H), 3.47 – 3.32 (m, 4H), 2.88 – 2.77 (m, 1H), 1.68 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.9, 165.6, 162.9, 144.7, 134.3, 130.8, 130.4, 126.2, 124.8 (q, J = 276.7 Hz), 124.6, 52.5, 44.3 (q, J = 27.5 Hz), 42.4 (q, J = 1.8 Hz), 31.0, 27.6.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.62 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{15}H_{15}F_3NO_4$ $[M+H]^+$ 330.0953, found, 330.0945.

2,4-dimethyl-1,3-dioxo-4-(2,2,2-trifluoroethyl)-1,2,3,4-tetrahydroisoquinoline-6-carbonitrile (**5h**)



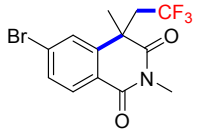
White solid, yield 48% (43.4 mg). M.p. = 107 – 109 °C.
R_f 0.35 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.59 (d, J = 1.6 Hz, 1H), 7.91 (dd, J = 8.2, 1.8 Hz, 1H), 7.57 (d, J = 8.2 Hz, 1H), 3.47 – 3.36 (m, 4H), 2.86 – 2.75 (m, 1H), 1.68 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.3, 161.9, 144.8, 136.2, 133.5, 127.0, 125.5, 123.30 (s), 117.2, 112.8, 44.2 (q, J = 27.6 Hz), 42.4 (q, J = 1.9 Hz), 30.9, 27.7.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.58 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{14}H_{12}F_3N_2O_2$ $[M+H]^+$ 297.0851, found, 297.0844.

6-chloro-2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5i**)



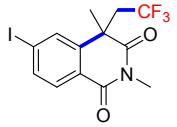
White solid, yield 46% (42.5 mg). M.p. = 99 – 101 °C.
R_f 0.45 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (s, 1H), 7.61 (d, J = 8.4 Hz, 1H), 7.36 (d, J = 8.4 Hz, 1H), 3.44 – 3.26 (m, 4H), 2.82 – 2.70 (m, 1H), 1.64 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.1, 162.6, 138.6, 134.5, 133.9, 129.0, 127.3, 125.8, 124.8 (q, J = 277.0 Hz), 44.3 (q, J = 27.3 Hz), 43.4 (q, J = 2.0 Hz), 31.1, 27.6.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.60 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{12}ClF_3NO_2$ $[M+H]^+$ 306.0509, found, 306.0502.

6-bromo-2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5j**)



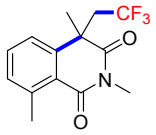
White solid, yield 55% (57.5 mg). M.p. = 119 – 121 °C.
R_f 0.45 (Petroleum ether/EtOAc, 5/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.77 (d, J = 8.5 Hz, 1H), 7.30 (d, J = 8.4 Hz, 1H), 3.40 (s, 3H), 3.38 – 3.28 (m, 1H), 2.76 (dq, J = 15.2, 9.6 Hz, 1H), 1.64 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.0, 161.5, 138.1, 135.8, 131.0, 126.5, 124.9, 123.8 (q, J = 276.9 Hz), 121.2, 43.2 (q, J = 27.6 Hz), 42.4 (q, J = 1.8 Hz), 30.0, 26.6.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.58 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{12}BrF_3NO_2$ $[M+H]^+$ 350.0004, found, 349.9995.

6-iodo-2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5k**)



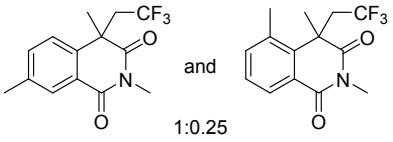
White solid, yield 54% (64.2 mg). M.p. = 109 – 111 °C.
R_f 0.35 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.60 (d, J = 1.9 Hz, 1H), 7.96 (dd, J = 8.3, 1.9 Hz, 1H), 7.15 (d, J = 8.3 Hz, 1H), 3.42 – 3.28 (m, 4H), 2.75 (dq, J = 15.1, 9.7 Hz, 1H), 1.63 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.0, 162.4, 142.6, 139.8, 138.1, 127.4, 125.9, 124.8 (q, J = 277.1 Hz), 93.2, 44.2 (q, J = 27.6 Hz), 43.5 (q, J = 1.8 Hz), 31.0, 27.6.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.54 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{12}F_3INO_2$ $[M+H]^+$ 397.9865, found, 397.9857.

2,4,8-trimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5l**)



Colorless oil, yield 57% (48.8 mg).
R_f 0.30 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (d, J = 7.7 Hz, 1H), 7.44 (s, 1H), 7.38 (t, J = 7.6 Hz, 1H), 3.52 – 3.37 (m, 4H), 3.31 – 3.19 (m, 1H), 2.61 (s, 3H), 1.75 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.2, 162.4, 136.9, 135.9, 134.1, 126.7, 126.4, 123.8, 123.7 (q, J = 276.8 Hz), 43.3 (q, J = 2.1 Hz), 39.6 (q, J = 27.1 Hz), 26.1, 25.8, 20.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -62.94 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{14}H_{15}F_3NO_2$ $[M+H]^+$ 286.1055, found, 286.1046.

2,4,7-trimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione and 2,4,5-trimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5m**)



White solid, yield 38% (32.3 mg). M.p. = 72 – 75 °C and 91 – 93 °C.

R_f 0.35 (Petroleum ether/EtOAc, 10/1).

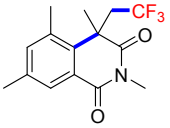
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (d, J = 7.7 Hz, 0.25H), 8.07 (s, 1H), 7.46 – 7.43 (m, 1.32H), 7.37 (t, J = 7.7 Hz, 0.27H), 7.29 (d, J = 8.0 Hz, 1H), 3.39 (s, 0.81H), 3.38 (s, 3H), 3.35 – 3.24 (m, 1.38H), 2.77 (dq, J = 15.1, 9.8 Hz, 1.27H), 2.60 (s, 0.79H), 2.42 (s, 3H), 1.73 (s, 0.77H), 1.61 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 175.8, 174.8, 163.98, 163.91, 138.5, 138.1, 137.55, 137.47, 135.7, 134.8, 129.3, 128.3, 128.0, 125.6, 125.0 (q, J = 277.0 Hz), 124.0, 44.9 (q, J = 1.4 Hz), 44.3 (q, J = 27.2 Hz), 43.3 (q, J = 1.4 Hz), 41.2 (q, J = 27.1 Hz), 31.2, 27.7, 27.4, 22.5, 21.0.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.63 (t, J = 10.2 Hz, 3F), -62.95 (t, J = 10.2 Hz, 0.75F).

HRMS (ESI) calcd for $C_{14}H_{15}F_3NO_2$ $[M+H]^+$ 286.1055, found, 286.1046.

2,4,5,7-tetramethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5n**)



Colorless oil, yield 75% (72.4 mg).

R_f 0.40 (Petroleum ether/EtOAc, 10/1).

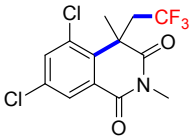
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (s, 1H), 7.27 (s, 1H), 3.50 – 3.36 (m, 4H), 3.23 (dq, J = 19.7, 9.9 Hz, 1H), 2.57 (s, 3H), 2.38 (s, 3H), 1.72 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 175.9, 164.2, 139.5, 137.8, 135.5, 134.6, 128.5, 125.3 (q, J = 276.8 Hz), 125.2, 44.7 (q, J = 2.3 Hz), 41.3 (q, J = 27.0 Hz), 27.7, 27.5, 22.4, 20.7.

^{19}F NMR (376 MHz, $CDCl_3$) δ -62.93 (t, J = 10.5 Hz, 3F).

HRMS (ESI) calcd for $C_{15}H_{17}F_3NO_2$ $[M+H]^+$ 300.1211, found, 300.1204.

5,7-dichloro-2,4-dimethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5o**)



White solid, yield 49% (50.3 mg). M.p. = 148 – 150 °C.

R_f 0.35 (Petroleum ether/EtOAc, 10/1).

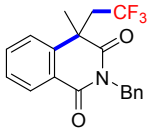
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.56 (d, J = 1.9 Hz, 1H), 7.34 (d, J = 1.9 Hz, 1H), 3.43 – 3.31 (m, 4H), 2.79 – 2.69 (m, 1H), 1.68 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 171.6, 159.7, 143.6, 138.4, 136.8, 131.2, 124.1, 123.7 (q, J = 277.1 Hz), 119.0, 43.6 (q, J = 27.6 Hz), 42.9 (q, J = 2.2 Hz), 30.2, 26.8.

^{19}F NMR (376 MHz, $CDCl_3$) δ -61.46 (t, J = 9.8 Hz, 3F).

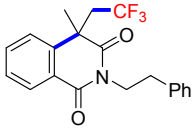
HRMS (ESI) calcd for $C_{13}H_{11}Cl_2F_3NO_2$ $[M+H]^+$ 340.0119, found, 340.0110.

2-benzyl-4-methyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5p**)



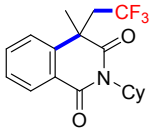
Colorless oil, yield 59% (61.5 mg).
R_f 0.30 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 (dd, J = 7.9, 1.2 Hz, 1H), 7.64 (td, J = 7.8, 1.4 Hz, 1H), 7.46 (t, J = 7.2 Hz, 1H), 7.40 (d, J = 7.7 Hz, 3H), 7.31 – 7.20 (m, 3H), 5.28 – 5.14 (m, 2H), 3.38 (dq, J = 15.1, 10.4 Hz, 1H), 2.81 (dq, J = 15.2, 9.7 Hz, 1H), 1.62 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 172.0, 161.1, 138.0, 134.4, 131.5, 127.2, 126.3, 126.1, 125.7, 125.1, 123.3, 121.9, 122.6 (q, J = 277.1 Hz), 41.6, 41.54 (q, J = 27.6 Hz), 41.52 (q, J = 1.7 Hz), 29.2.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.20 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{19}H_{17}F_3NO_2$ $[M+H]^+$ 348.1211, found, 348.1202.

4-methyl-2-phenethyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5q**)



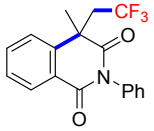
Colorless oil, yield 65% (70 mg).
R_f 0.30 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.30 (dd, J = 7.9, 1.2 Hz, 1H), 7.66 (td, J = 7.8, 1.4 Hz, 1H), 7.49 (t, J = 7.2 Hz, 1H), 7.41 (d, J = 7.9 Hz, 1H), 7.36 – 7.26 (m, 4H), 7.25 – 7.18 (m, 1H), 4.33 – 4.17 (m, 2H), 3.37 (dq, J = 15.1, 10.4 Hz, 1H), 2.92 (t, J = 8.0 Hz, 2H), 2.80 (dq, J = 15.1, 9.8 Hz, 1H), 1.59 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 173.2, 162.3, 139.4, 137.5, 132.8, 128.3, 128.0, 127.4, 127.0, 125.4, 124.6, 124.0 (q, J = 277.0 Hz), 123.2, 43.1 (q, J = 27.3 Hz), 42.6 (q, J = 1.7 Hz), 40.9, 32.6, 30.2.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.28 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{20}H_{19}F_3NO_2$ $[M+H]^+$ 362.1368, found, 362.1360.

2-cyclohexyl-4-methyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5r**)



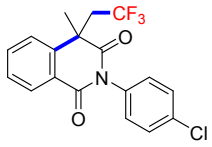
White solid, yield 76% (77.4 mg). M.p. = 85 – 87 °C.
R_f 0.35 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (dd, J = 7.9, 0.8 Hz, 1H), 7.62 (t, J = 7.6 Hz, 1H), 7.46 (t, J = 7.6 Hz, 1H), 7.38 (d, J = 7.9 Hz, 1H), 4.81 (tt, J = 12.2, 3.6 Hz, 1H), 3.32 (dq, J = 15.1, 10.4 Hz, 1H), 2.76 (dq, J = 15.1, 9.8 Hz, 1H), 2.45 – 2.31 (m, 2H), 1.84 (d, J = 12.7 Hz, 2H), 1.69 – 1.62 (m, 6H), 1.46 – 1.19 (m, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.6, 163.9, 140.3, 133.5, 129.4, 127.9, 125.5, 124.9, 125.1 (q, J = 277.3 Hz), 54.3, 44.3 (q, J = 27.3 Hz), 43.8 (q, J = 1.9 Hz), 31.2, 28.73, 28.69, 26.44, 26.42, 25.4.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.28 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{18}H_{21}F_3NO_2$ $[M+H]^+$ 340.1524, found, 340.1515.

4-methyl-2-phenyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5s**)



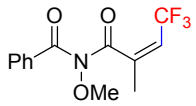
White solid, yield 46% (45.5 mg). M.p. = 189 – 190 °C.
R_f 0.30 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.32 (dd, J = 7.9, 0.9 Hz, 1H), 7.72 (td, J = 7.8, 1.3 Hz, 1H), 7.56 – 7.42 (m, 5H), 7.19 (d, J = 7.3 Hz, 2H), 3.38 (dq, J = 15.1, 10.4 Hz, 1H), 2.87 (dq, J = 15.1, 9.8 Hz, 1H), 1.77 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.5, 163.7, 140.5, 135.2, 134.2, 129.7, 129.4, 128.8, 128.2, 125.9, 125.2 (q, J = 277.1 Hz), 124.4, 44.6 (q, J = 27.3 Hz), 44.1 (q, J = 1.9 Hz), 31.2.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.33 (t, J = 9.8 Hz, 3F).
HRMS (ESI) calcd for $C_{18}H_{15}F_3NO_2$ $[M+H]^+$ 334.1055, found, 334.1044.

2-(4-chlorophenyl)-4-methyl-4-(2,2,2-trifluoroethyl)isoquinoline-1,3(2H,4H)-dione (**5t**)



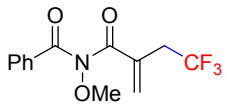
White solid, yield 39% (43.5 mg). M.p. = 189 – 191 °C.
R_f 0.25 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.31 (dd, J = 7.9, 1.0 Hz, 1H), 7.73 (td, J = 7.9, 1.4 Hz, 1H), 7.56 – 7.45 (m, 4H), 7.12 (d, J = 8.6 Hz, 2H), 3.43 – 3.31 (m, 1H), 2.87 (dq, J = 15.1, 9.8 Hz, 1H), 1.77 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 174.4, 163.5, 140.5, 134.8, 134.4, 133.6, 129.7, 129.67, 129.64, 128.3, 125.9, 125.1 (q, J = 277.2 Hz), 124.2, 44.8 (q, J = 27.4 Hz), 44.1 (q, J = 1.6 Hz), 31.1.
^{19}F NMR (376 MHz, $CDCl_3$) δ -61.39 (t, J = 10.2 Hz, 3F).
HRMS (ESI) calcd for $C_{18}H_{14}ClF_3NO_2$ $[M+H]^+$ 368.0665, found, 368.0656.

N-methoxy-N-(4,4,4-trifluoro-2-methylbut-2-enoyl)benzamide (**5ua**)



Colorless oil, yield 34% (29.5 mg).
R_f 0.70 (Petroleum ether/EtOAc, 10/1).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 7.1 Hz, 2H), 7.46 – 7.38 (m, 3H), 6.93 (q, J = 8.1 Hz, 1H), 3.98 (s, 3H), 2.23 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 161.5, 146.8, 137.8 (q, J = 5.0 Hz), 130.8, 128.9, 128.7, 128.4 (q, J = 35.0 Hz), 125.8, 122.5 (q, J = 270.3 Hz), 63.2, 13.9.
^{19}F NMR (376 MHz, $CDCl_3$) δ -59.37 (d, J = 7.1 Hz, 3F).
HRMS (ESI) calcd for $C_{13}H_{13}F_3NO_3$ $[M+H]^+$ 288.0848, found, 288.0839.

N-methoxy-N-(4,4,4-trifluoro-2-methylenebutanoyl)benzamide (**5ub**)



Colorless oil, yield 3% (2.9 mg).

R_f 0.50 (Petroleum ether/EtOAc, 10/1).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.69 (d, J = 7.2 Hz, 2H), 7.43 – 7.39 (m, 3H), 6.80 (s, 1H), 6.17 (s, 1H), 3.96 (s, 3H), 3.28 (q, J = 10.4 Hz, 2H).

^{19}F NMR (376 MHz, $CDCl_3$) δ -65.65 (t, J = 10.5 Hz, 3F).

HRMS (ESI) calcd for $C_{13}H_{13}F_3NO_3$ $[M+H]^+$ 288.0848, found, 288.0839.

5. References

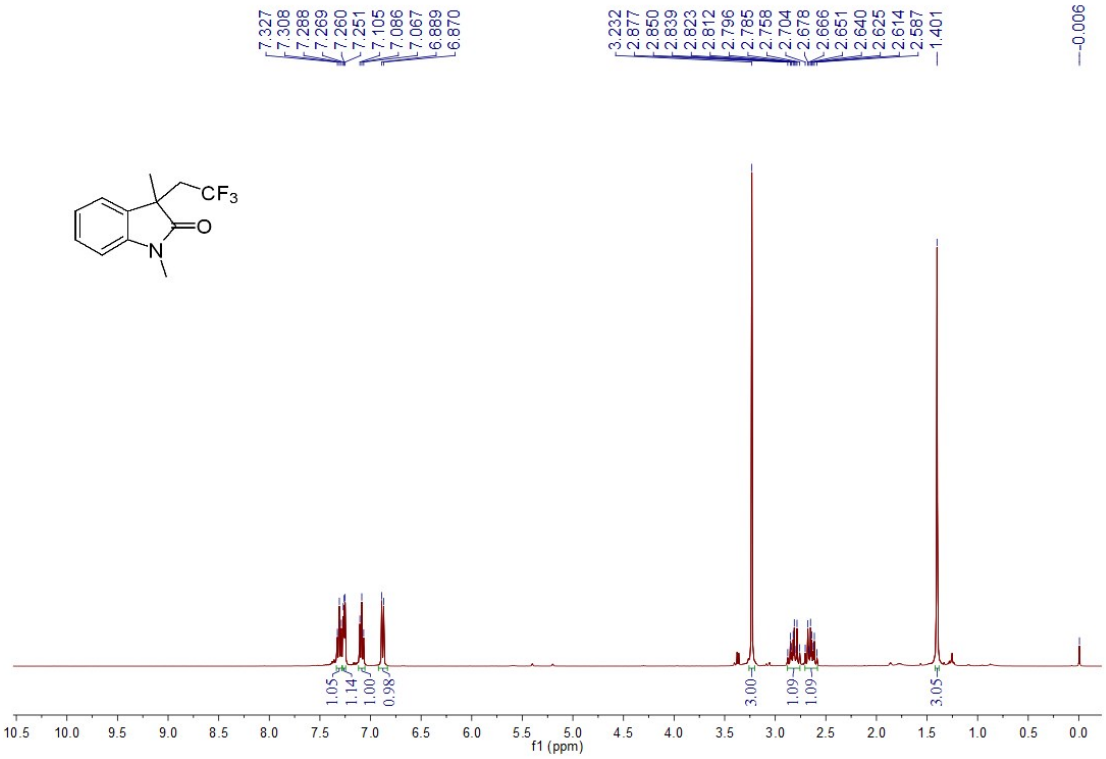
[1] W. Zhang, Z. Zou, W. Zhao, S. Lu, Z. Wu, M. Huang, X. Wang, Y. Wang, Y. Liang, Y. Zhu, Y. Zheng and Y. Pan, *Nat Commun*., 2020, **11**, 2572–2578.

[2] T. Wu, X. Mu, G. Liu, *Angew. Chem. Int. Ed.*, 2011, **50**, 12578–12581.

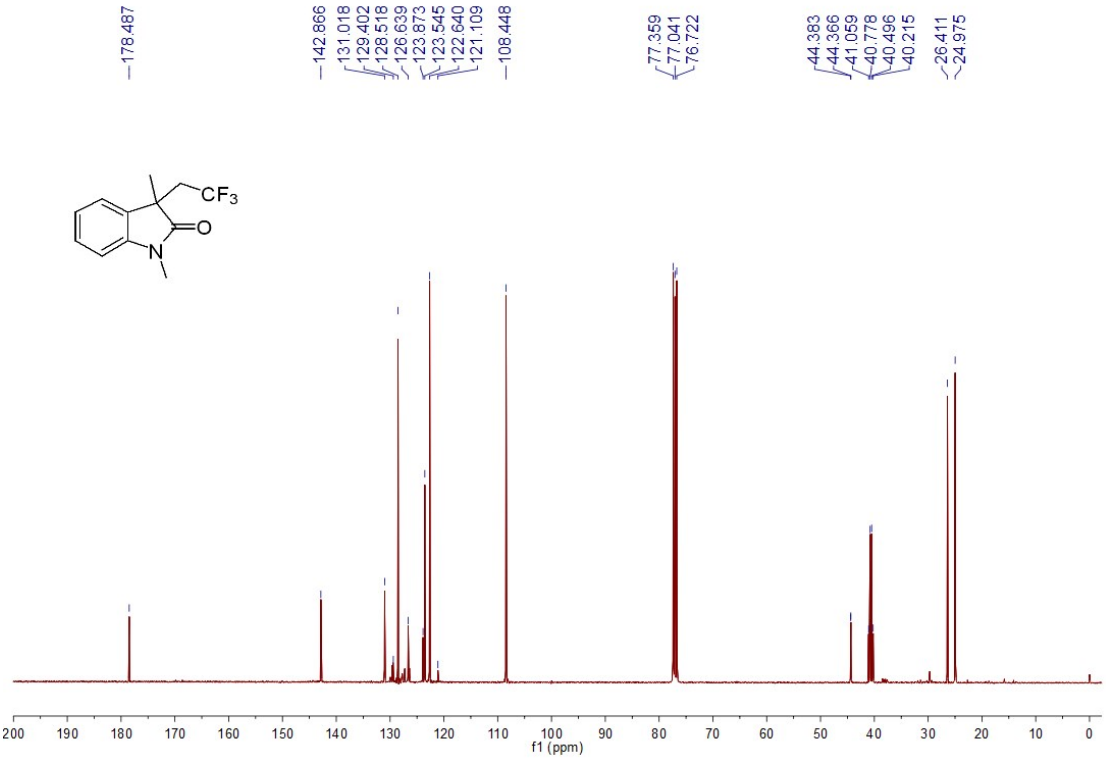
[3] W. Kong, M. Casimiro, N. Fuentes, E. Merino, C. Nevado, *Angew. Chem. Int. Ed.*, 2013, **52**, 13086–13090.

6. NMR spectra

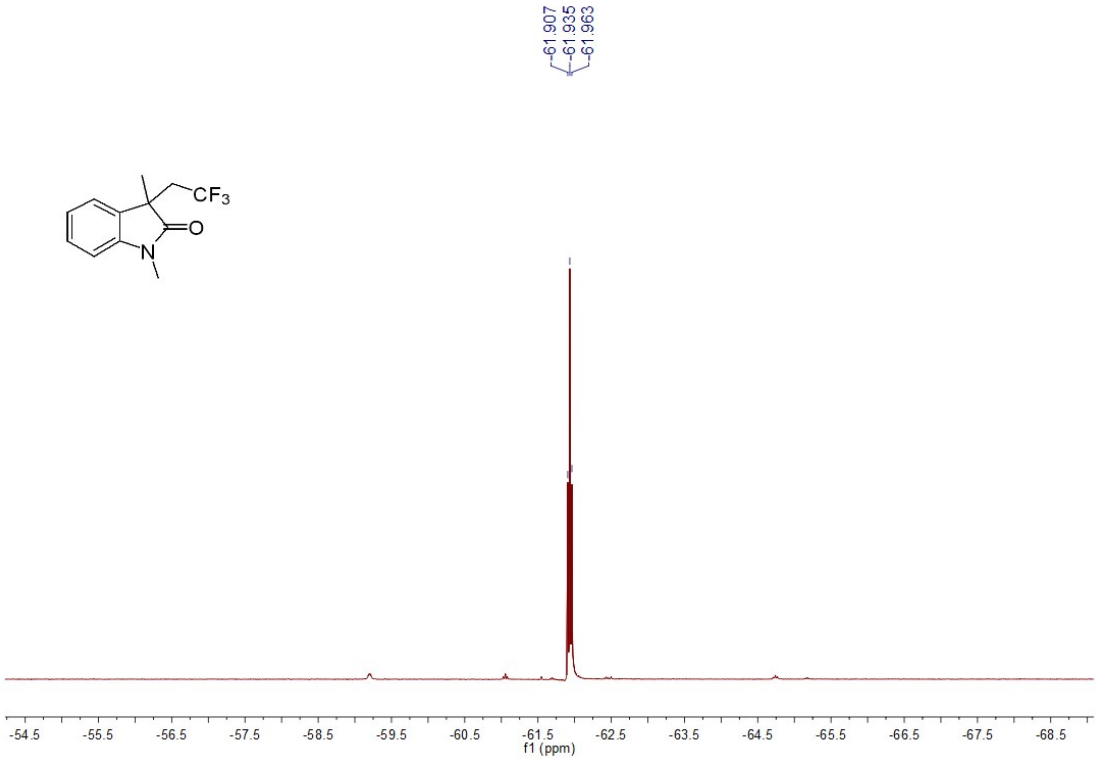
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4a**



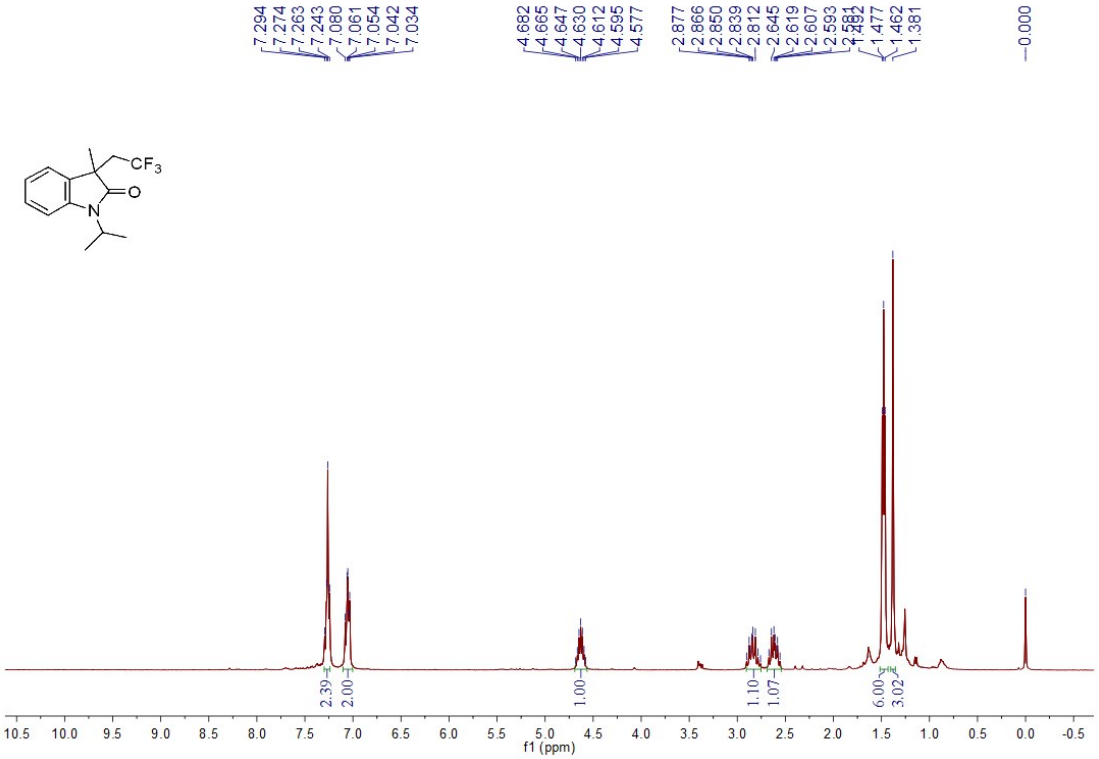
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4a**



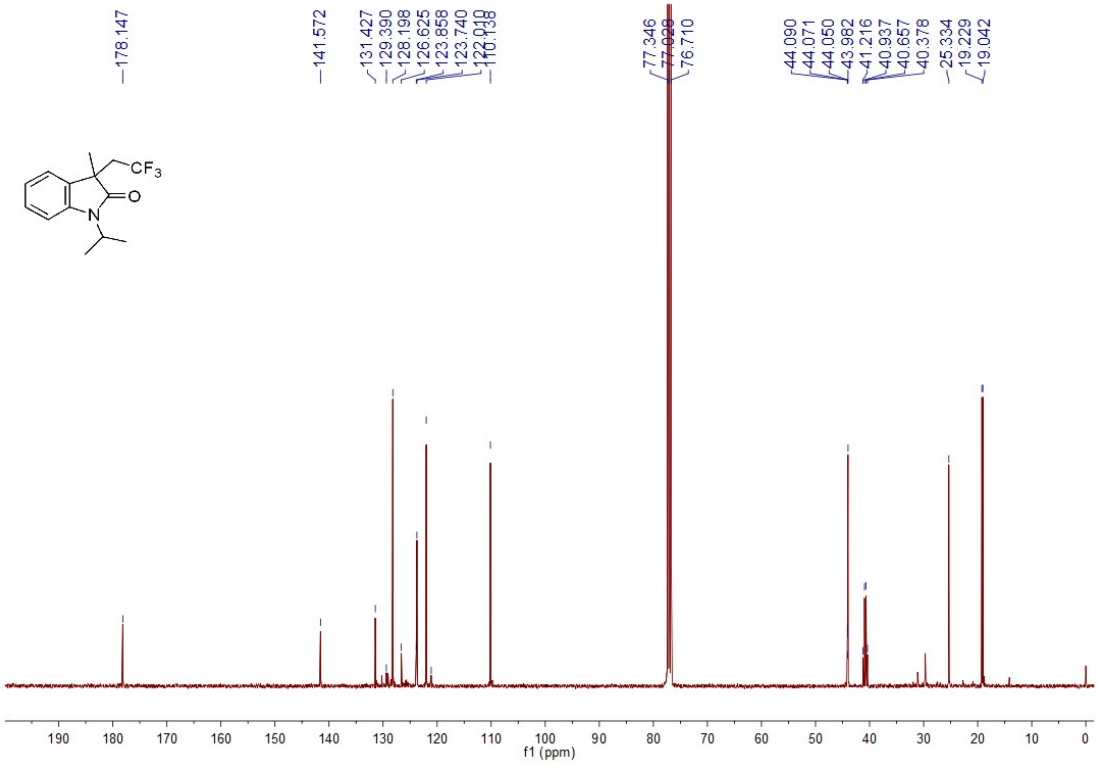
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4a**



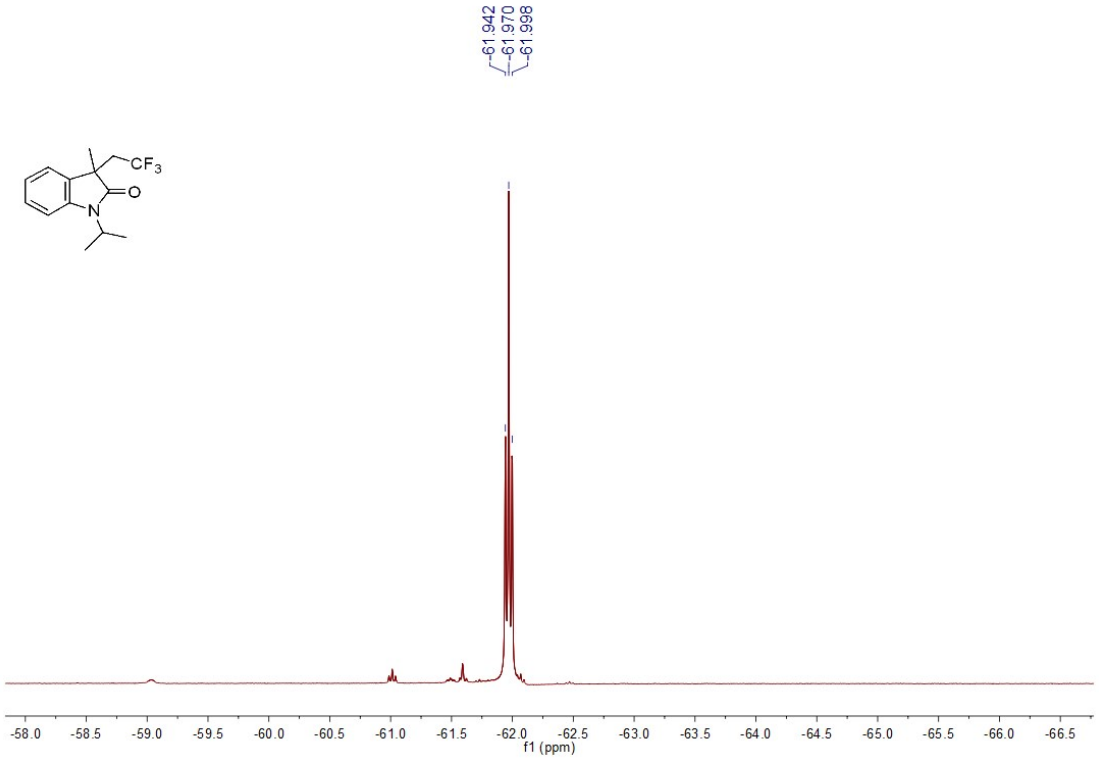
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4b**



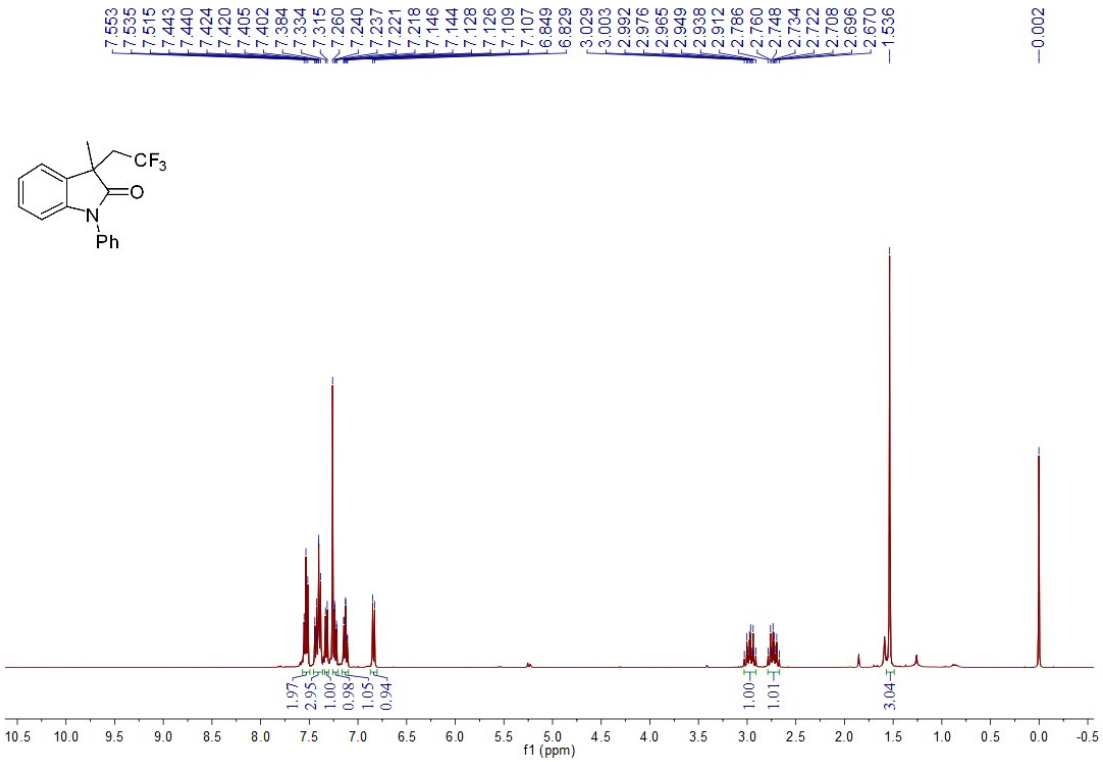
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4b**



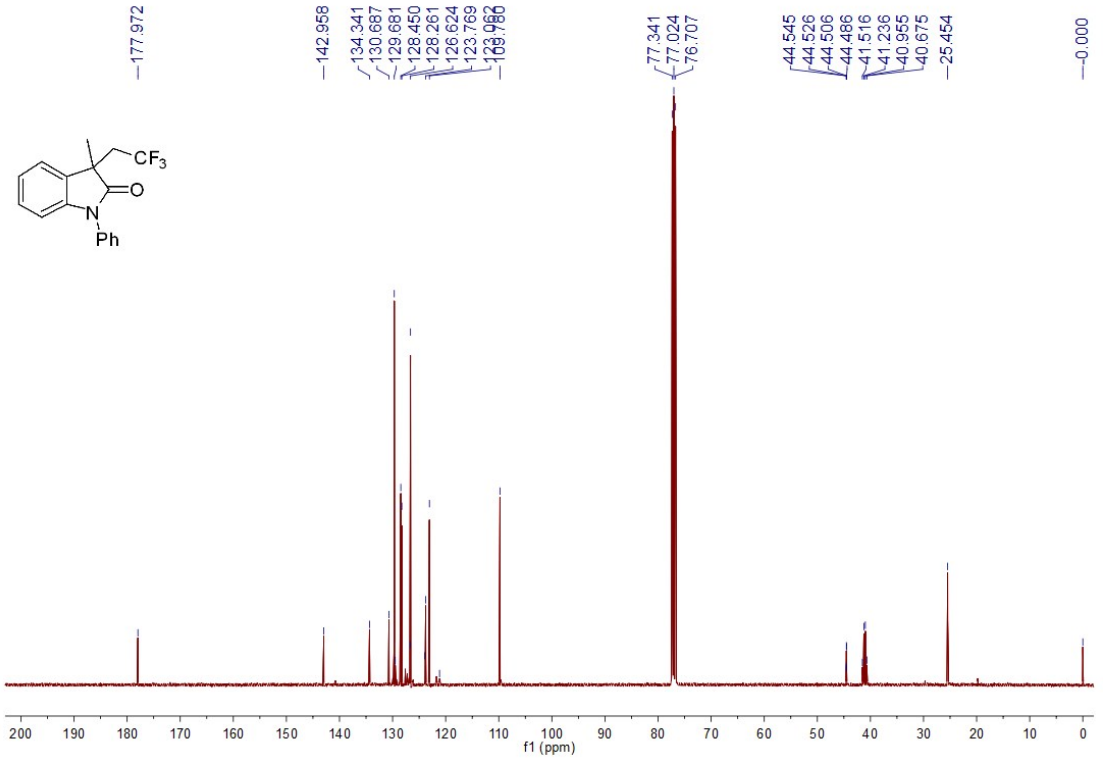
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4b**



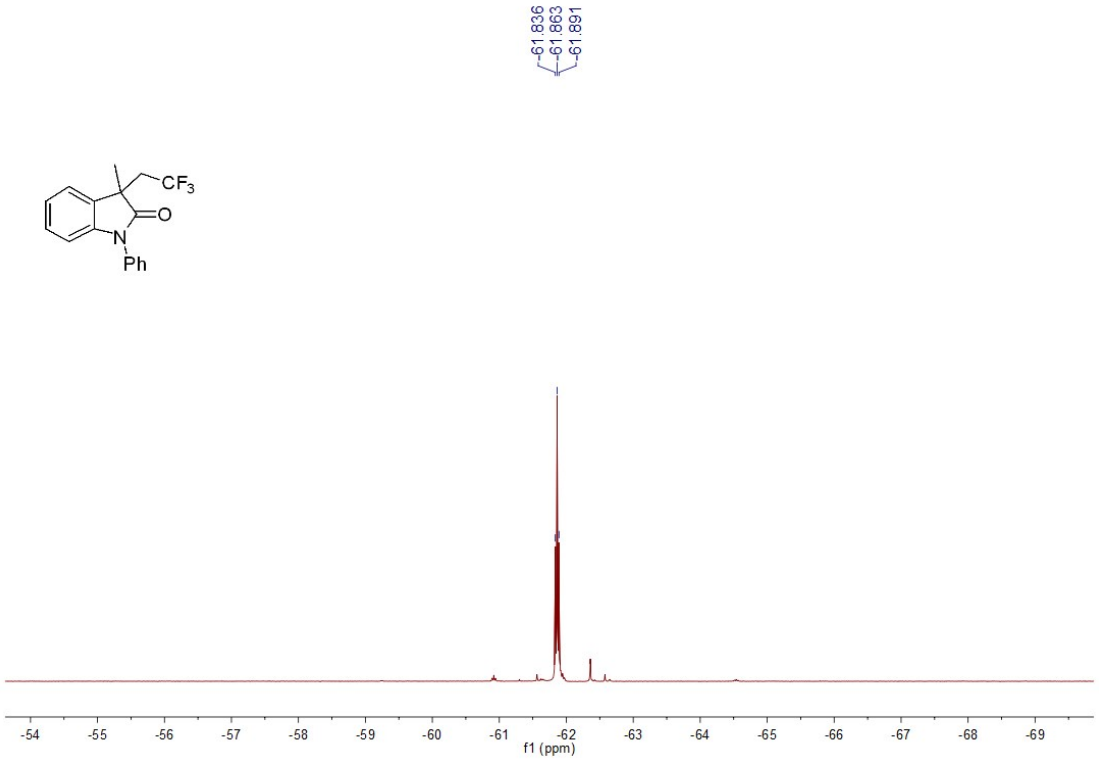
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4c**



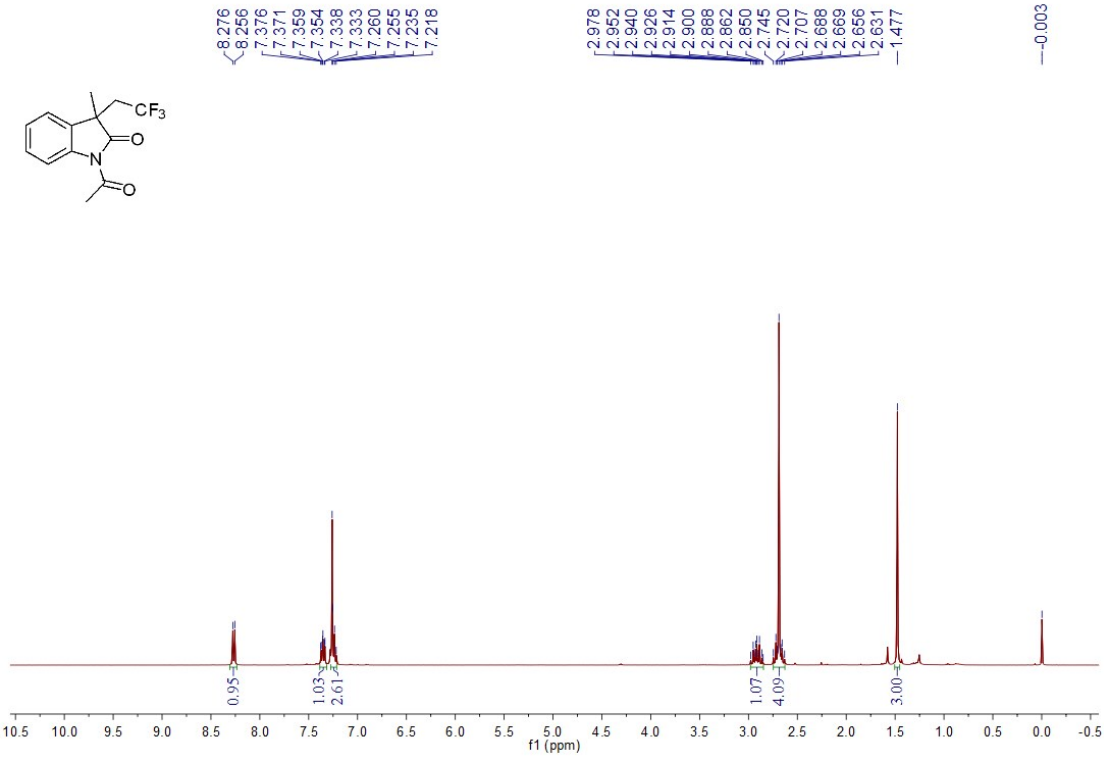
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4c**



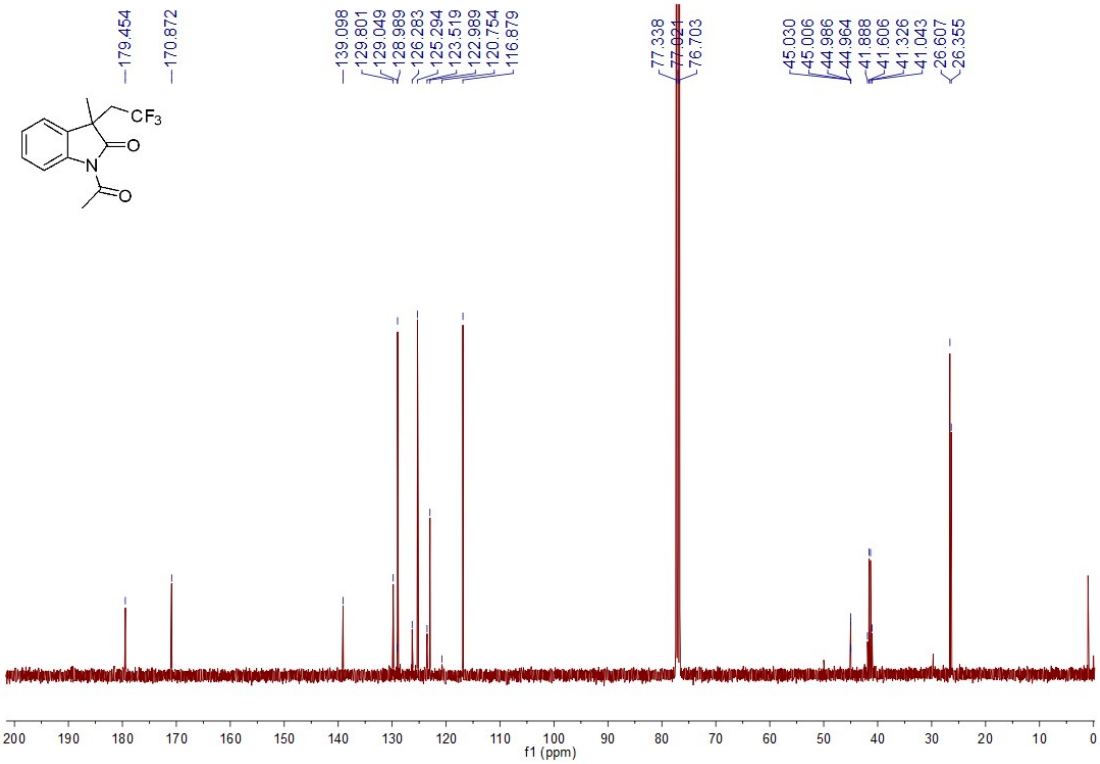
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4c**



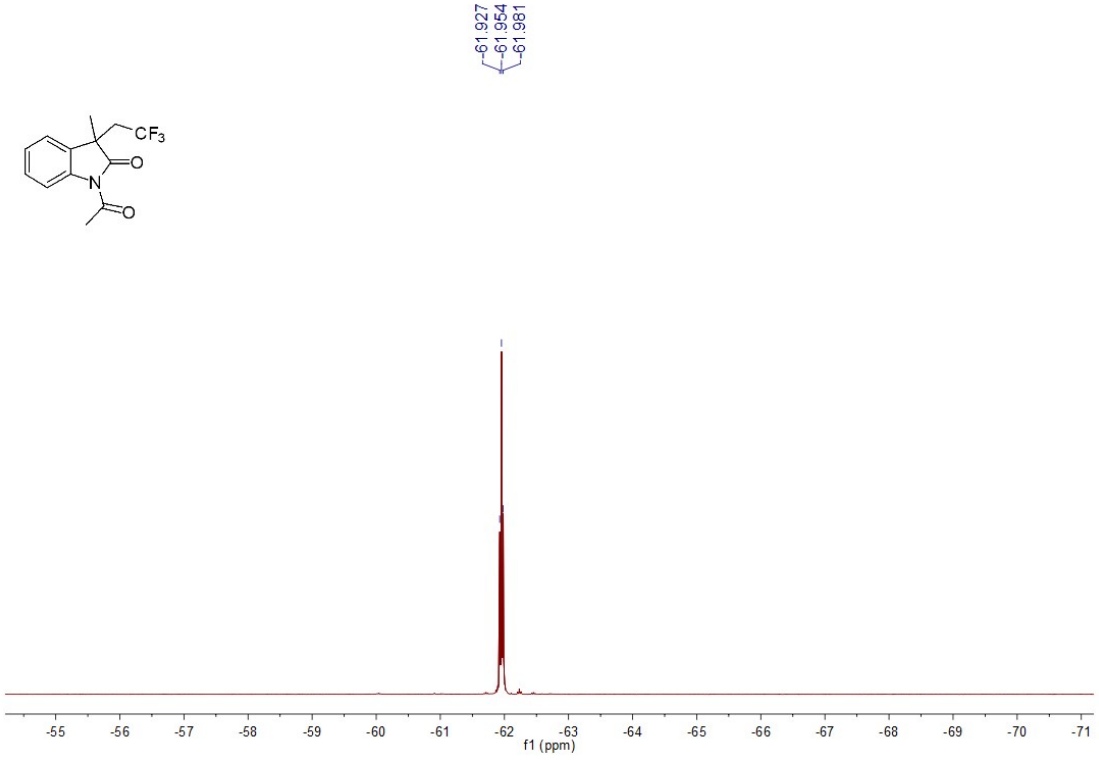
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4d**



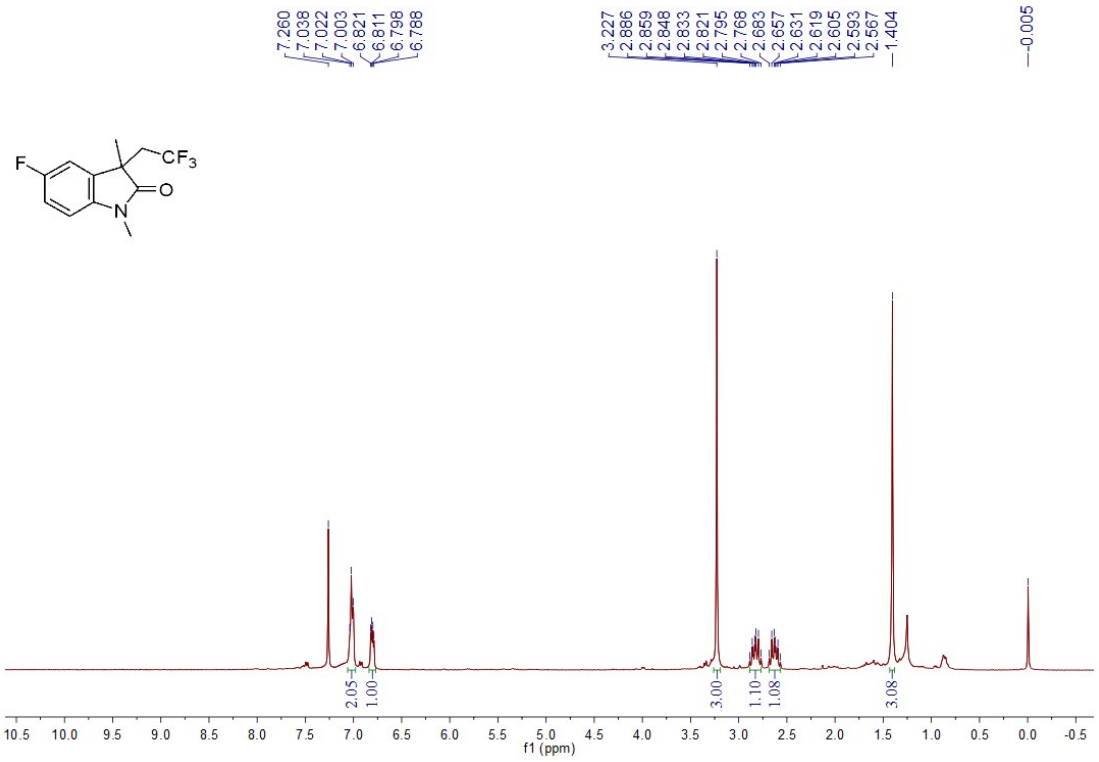
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4d**



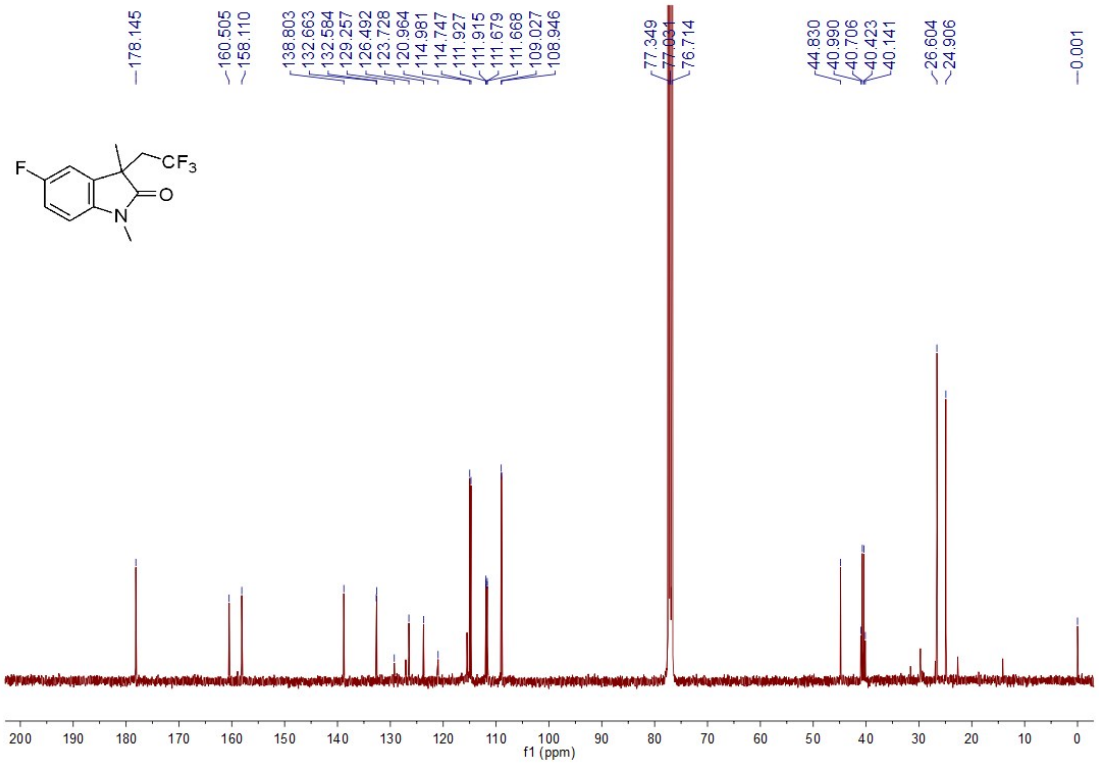
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4d**



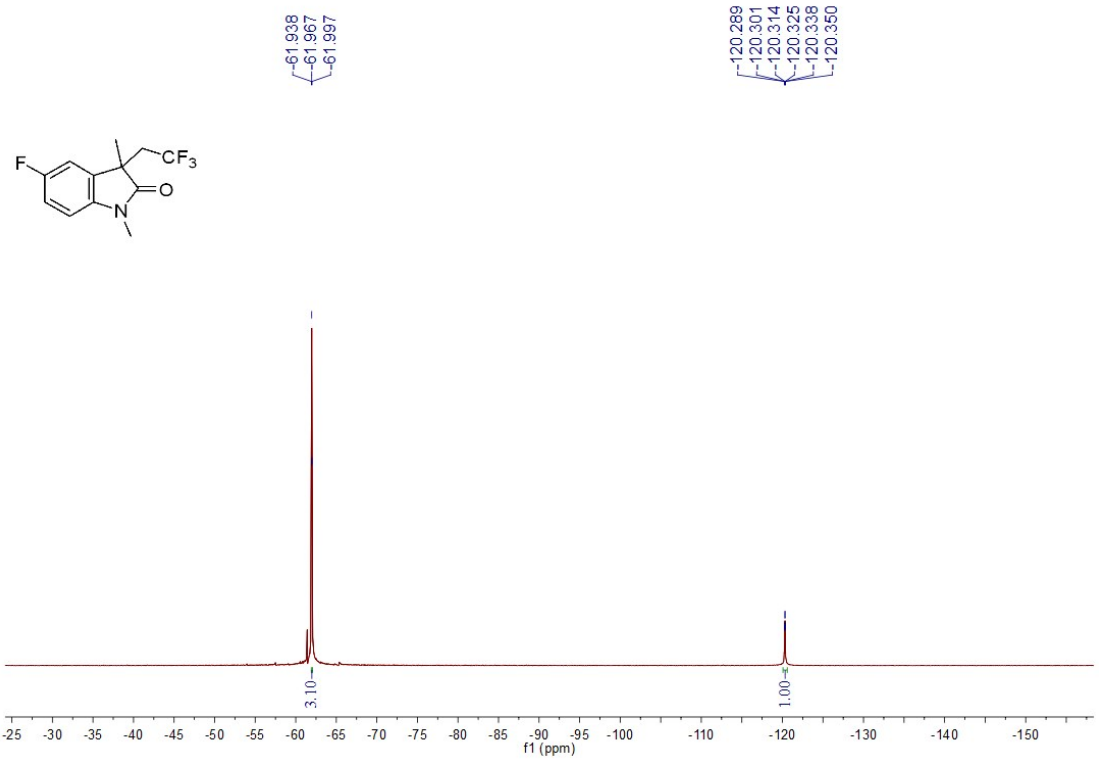
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4e**



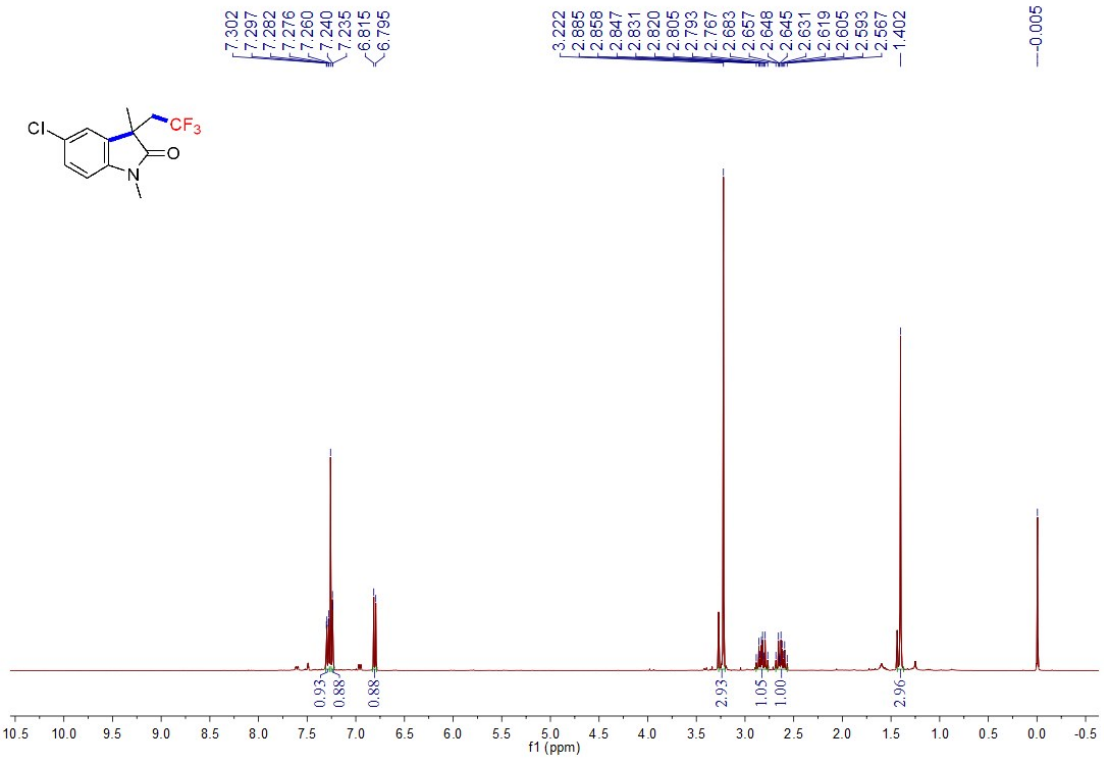
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4e**



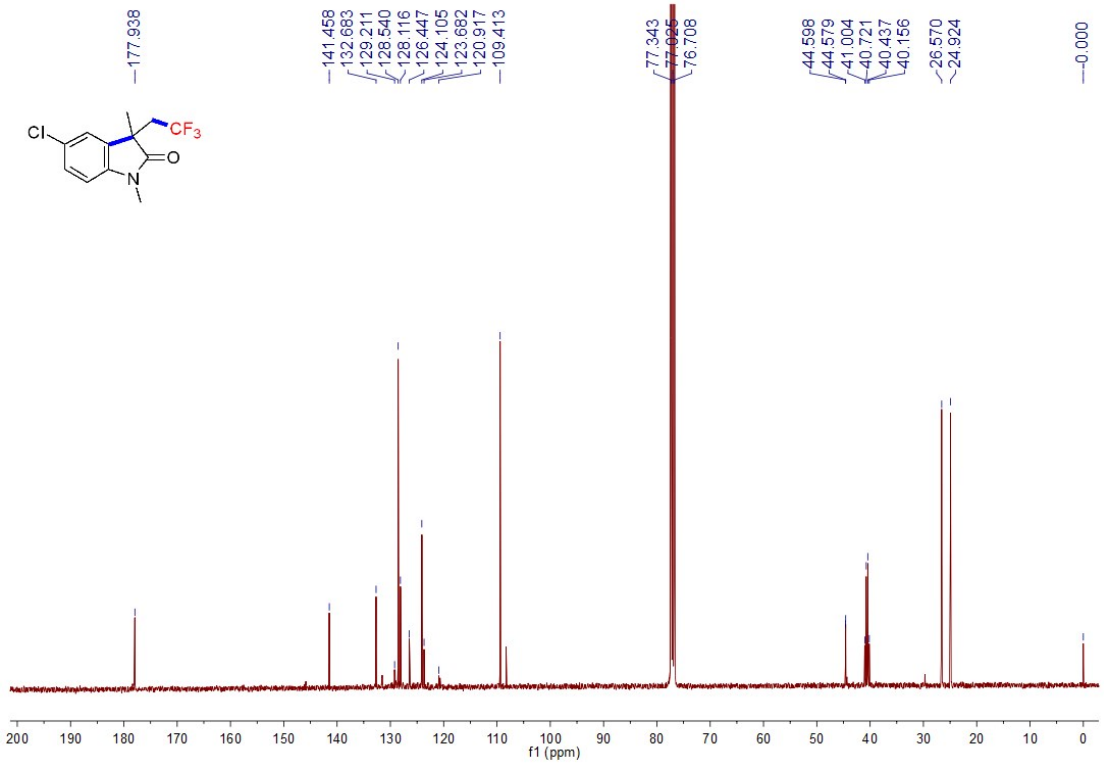
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4e**



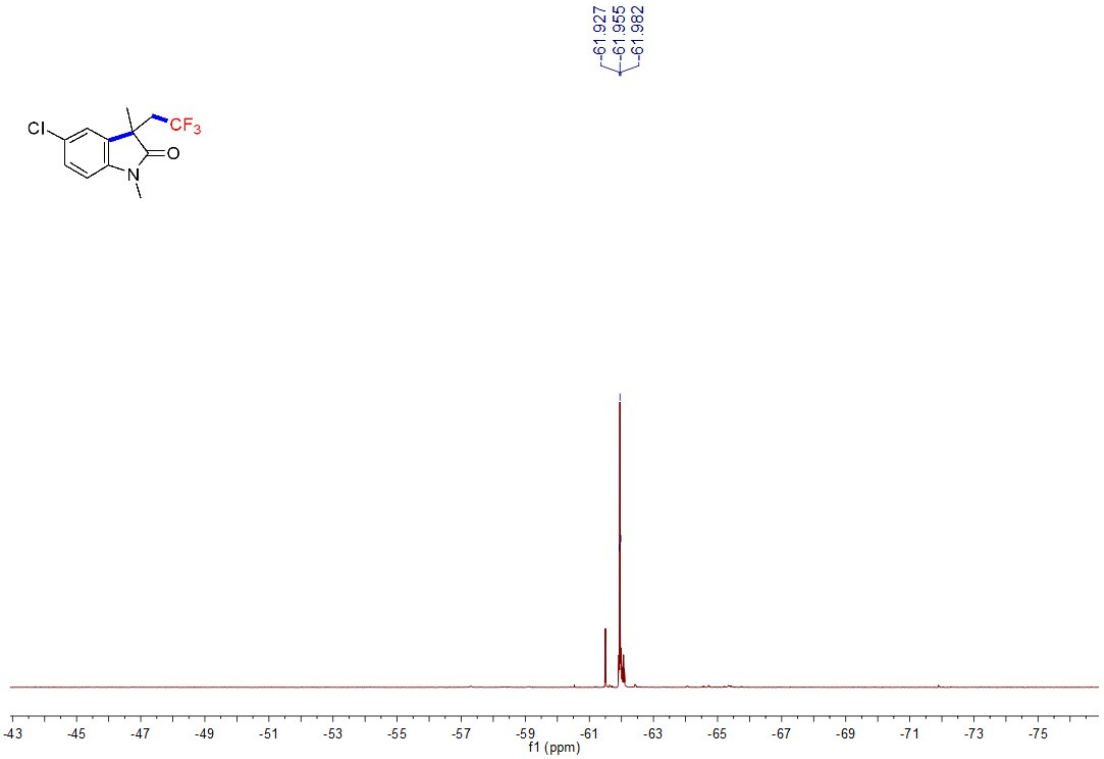
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4f**



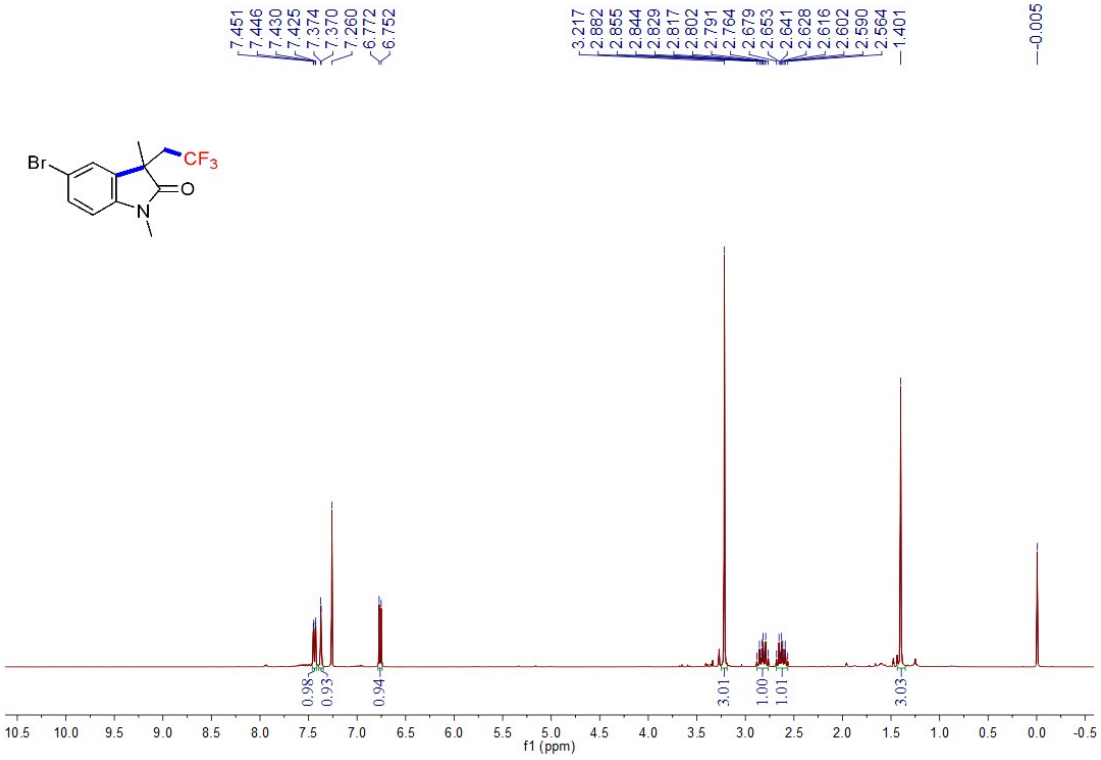
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4f**



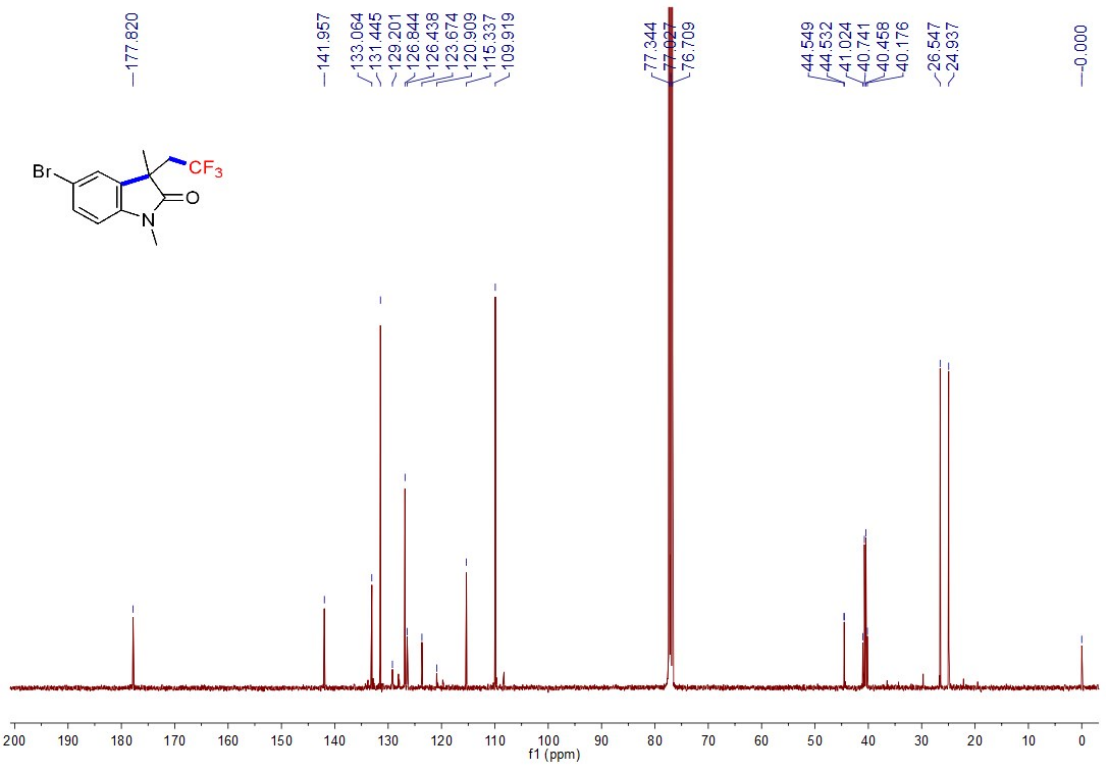
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4f**



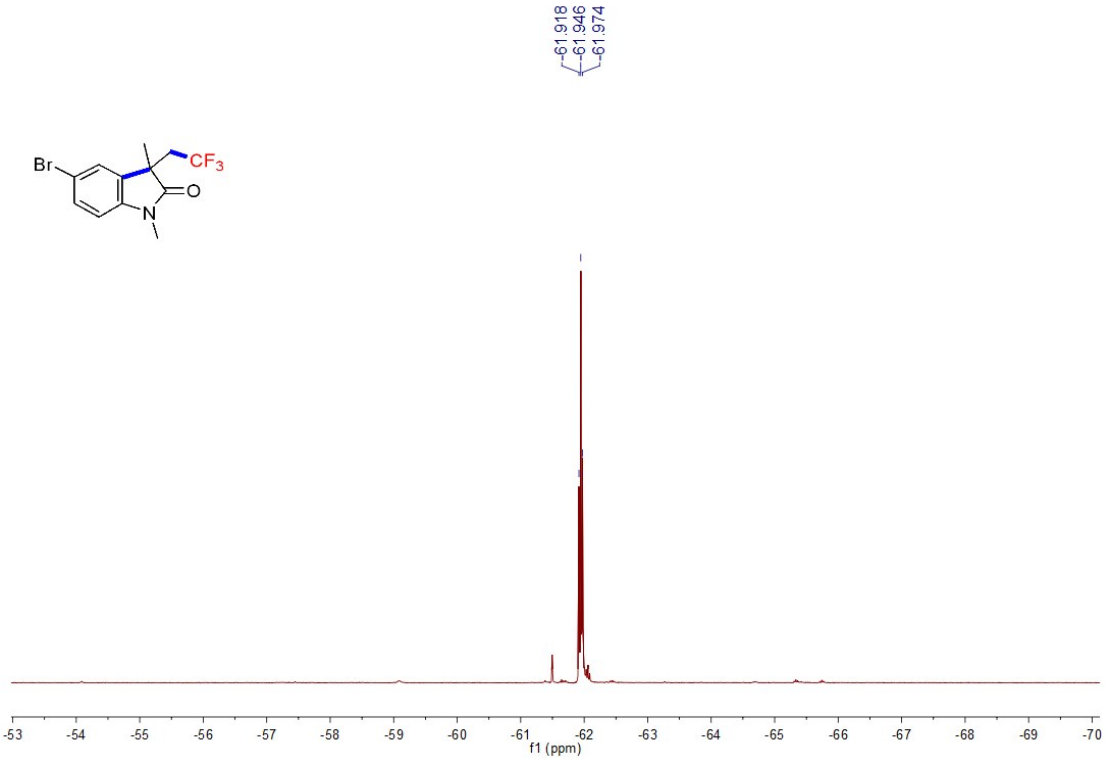
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4g**



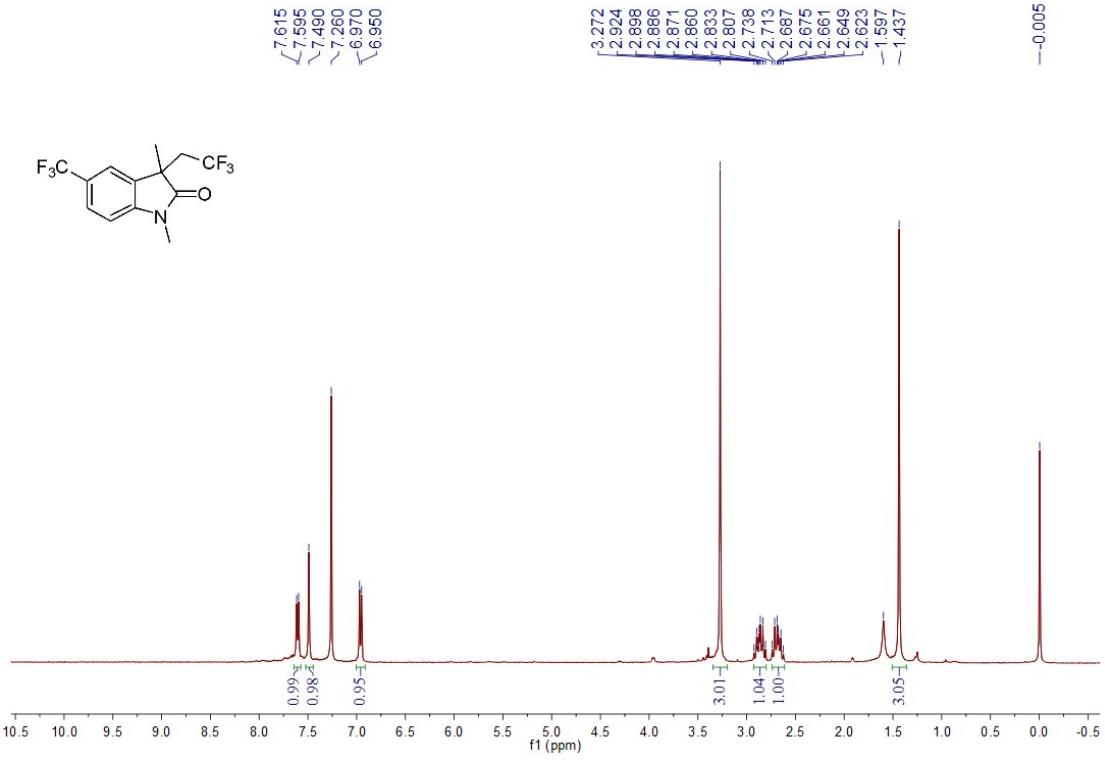
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4g**



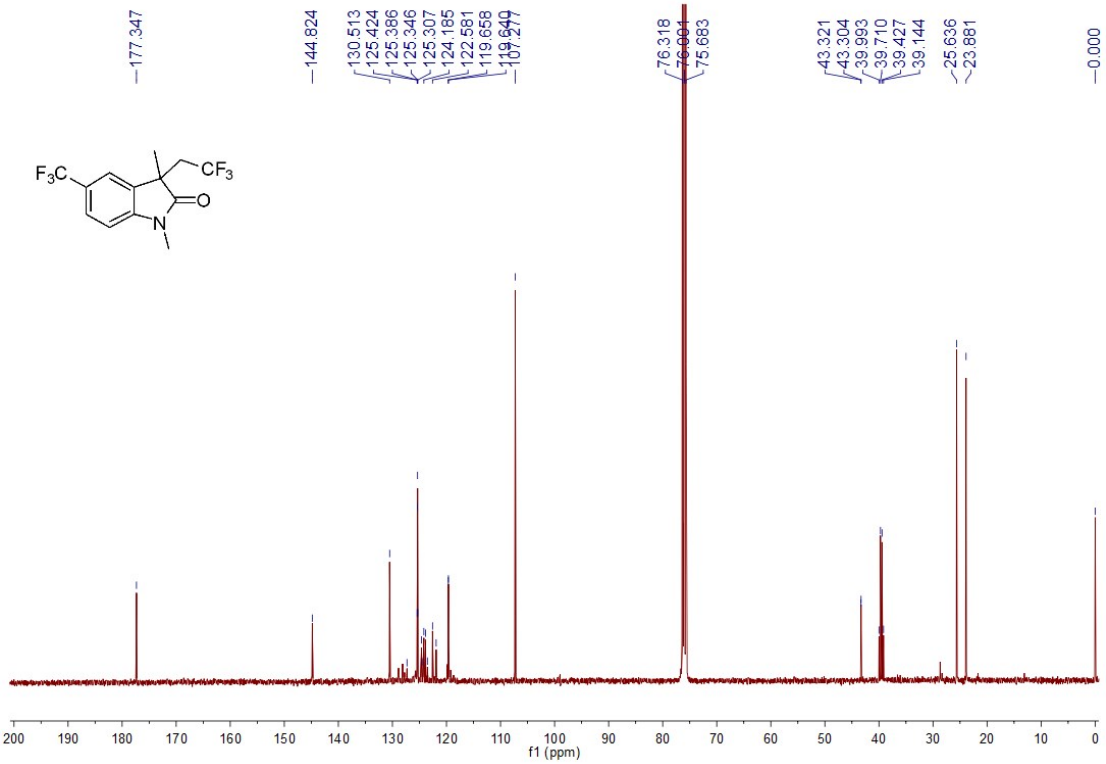
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4g**



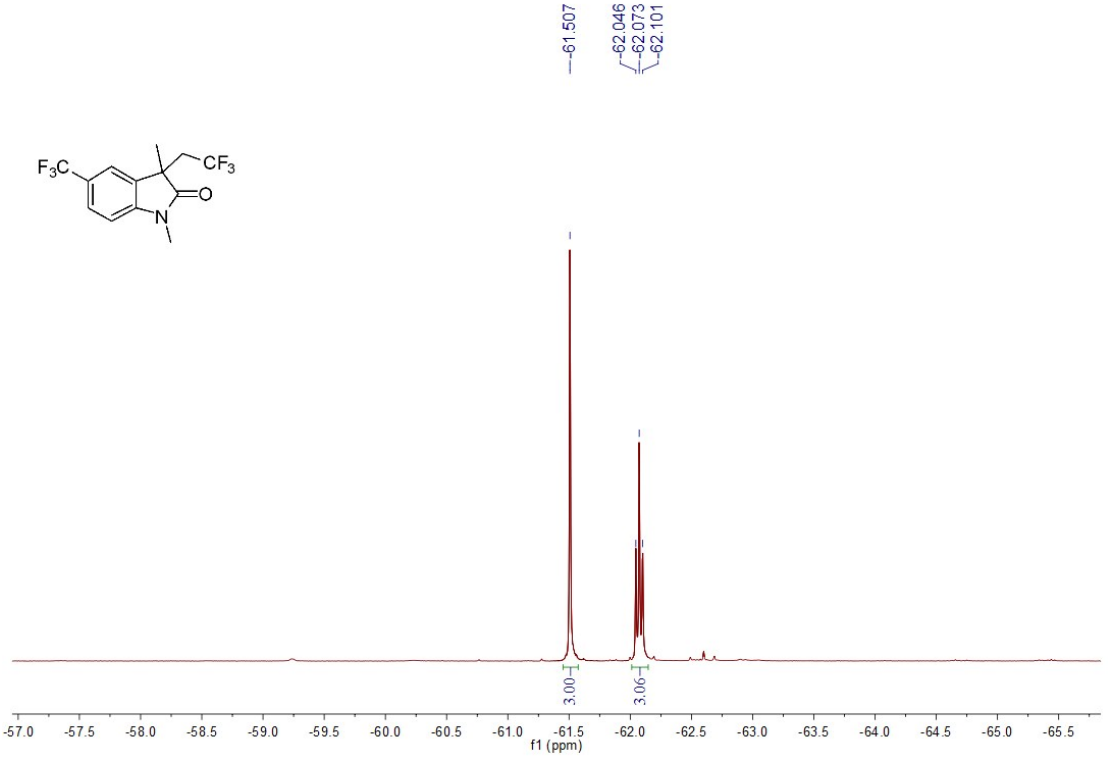
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **4h**



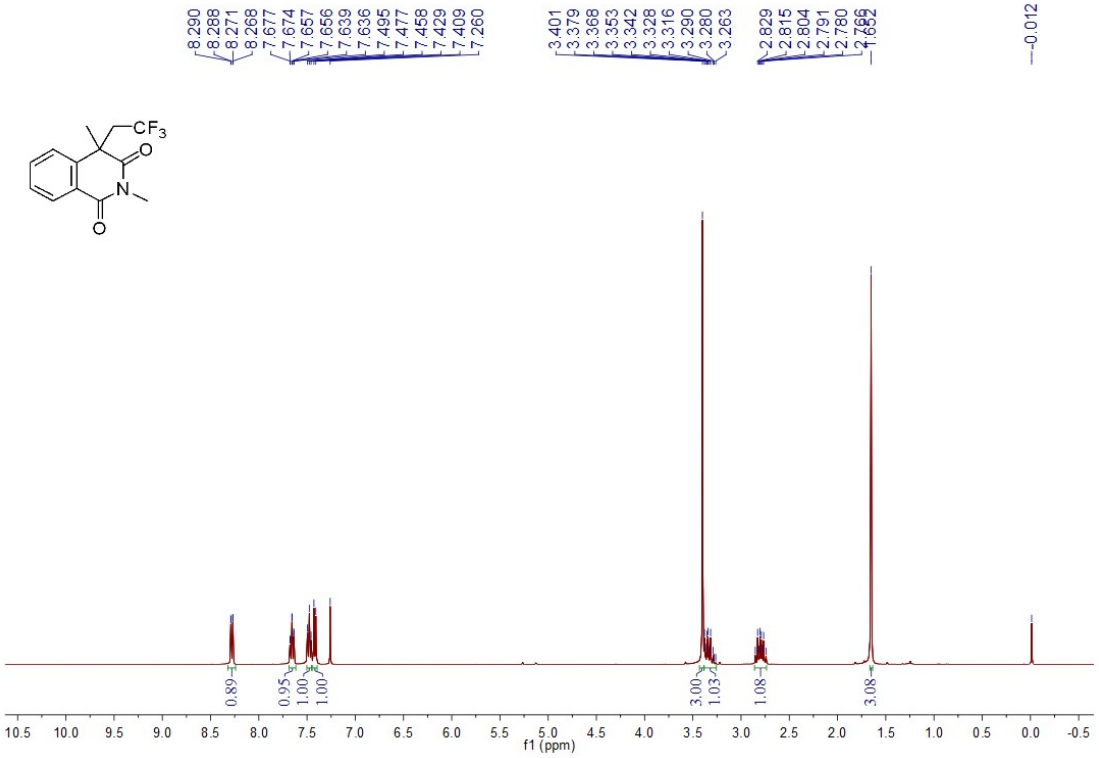
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **4h**



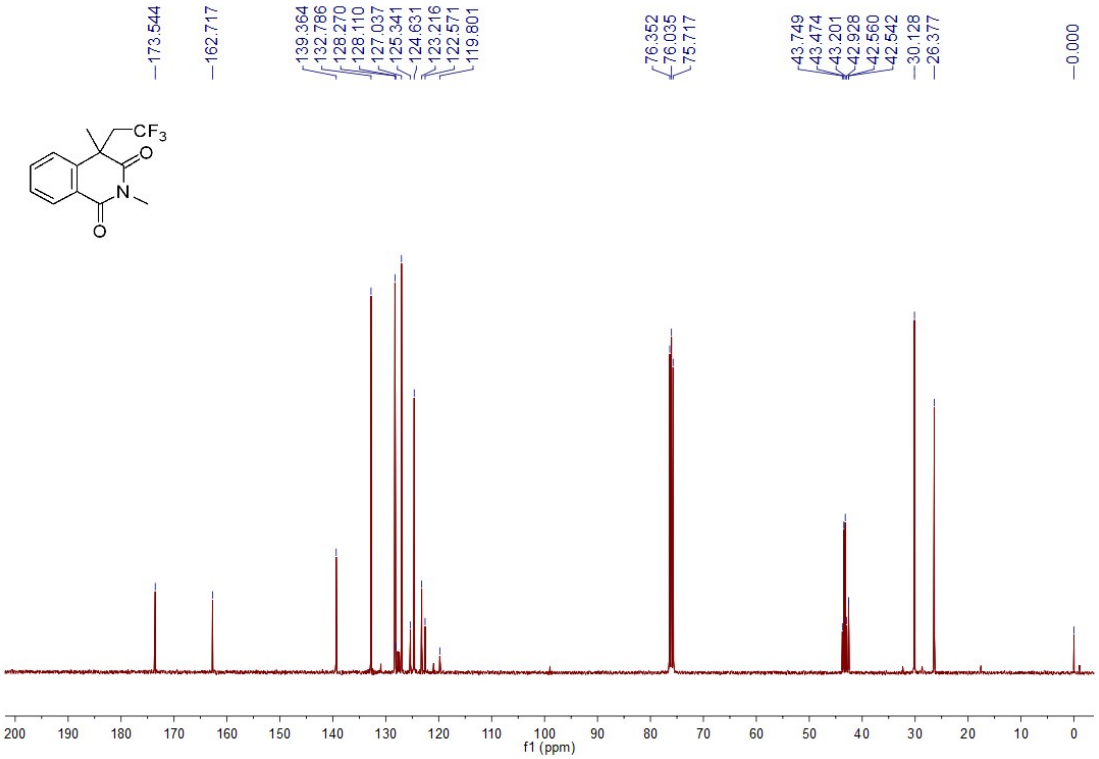
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **4h**



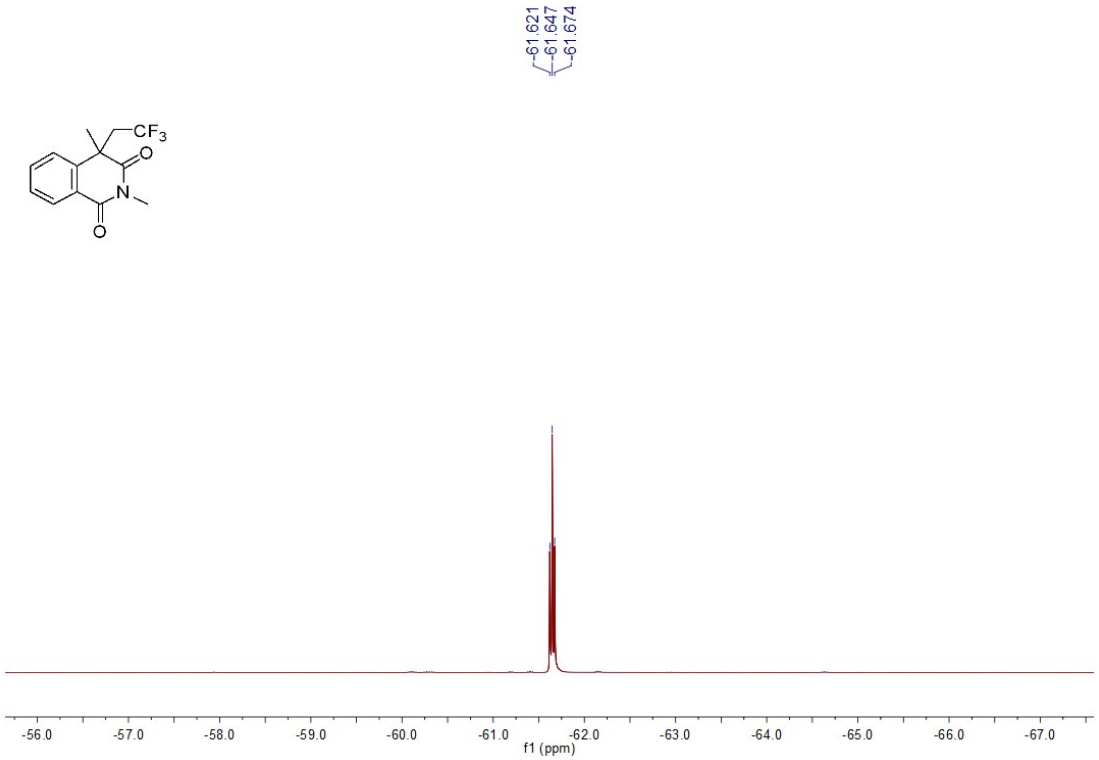
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5a**



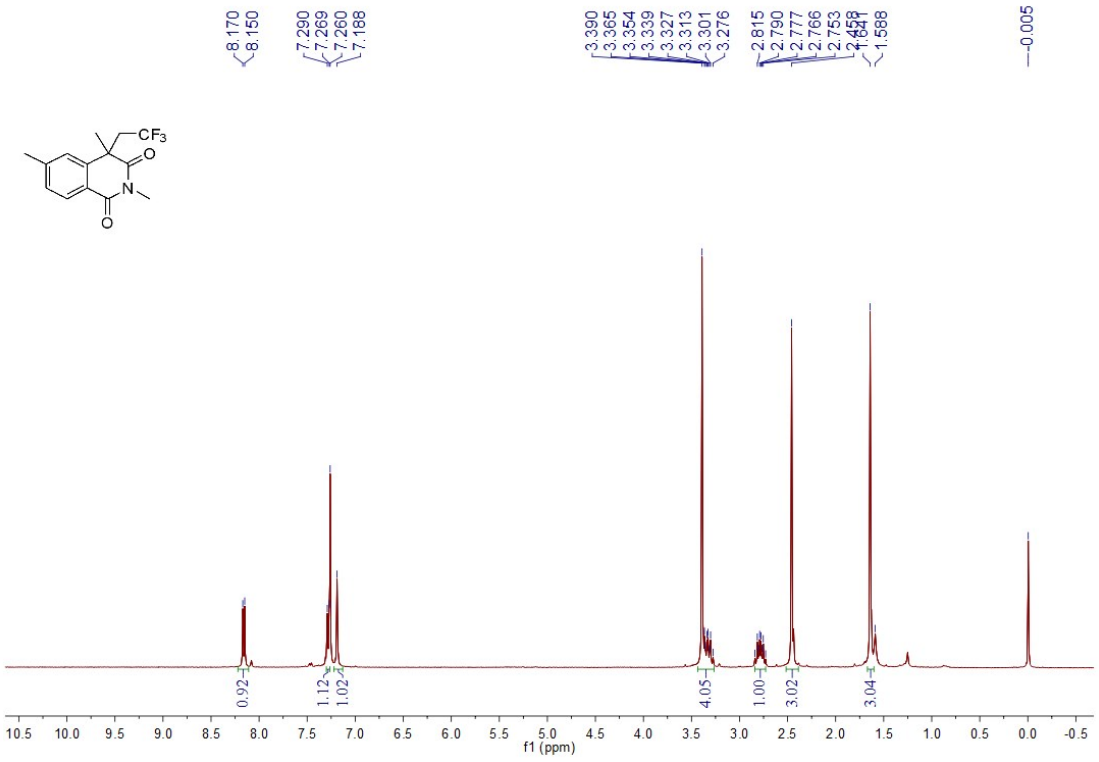
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5a**



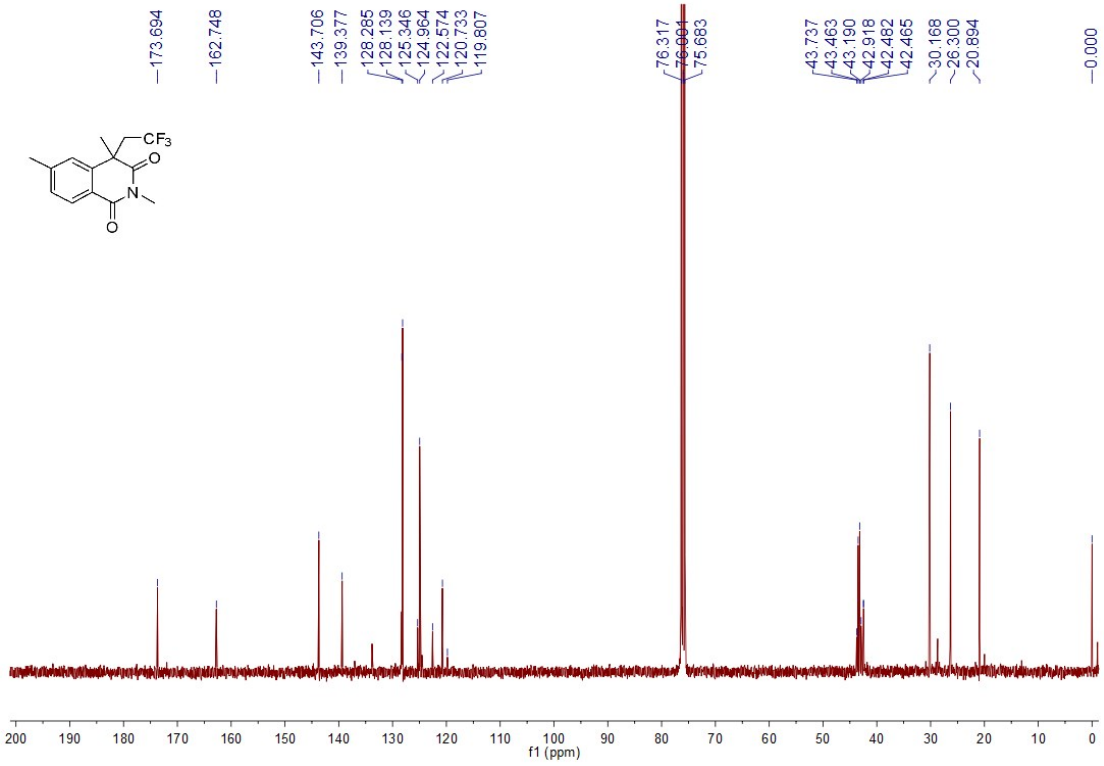
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5a**



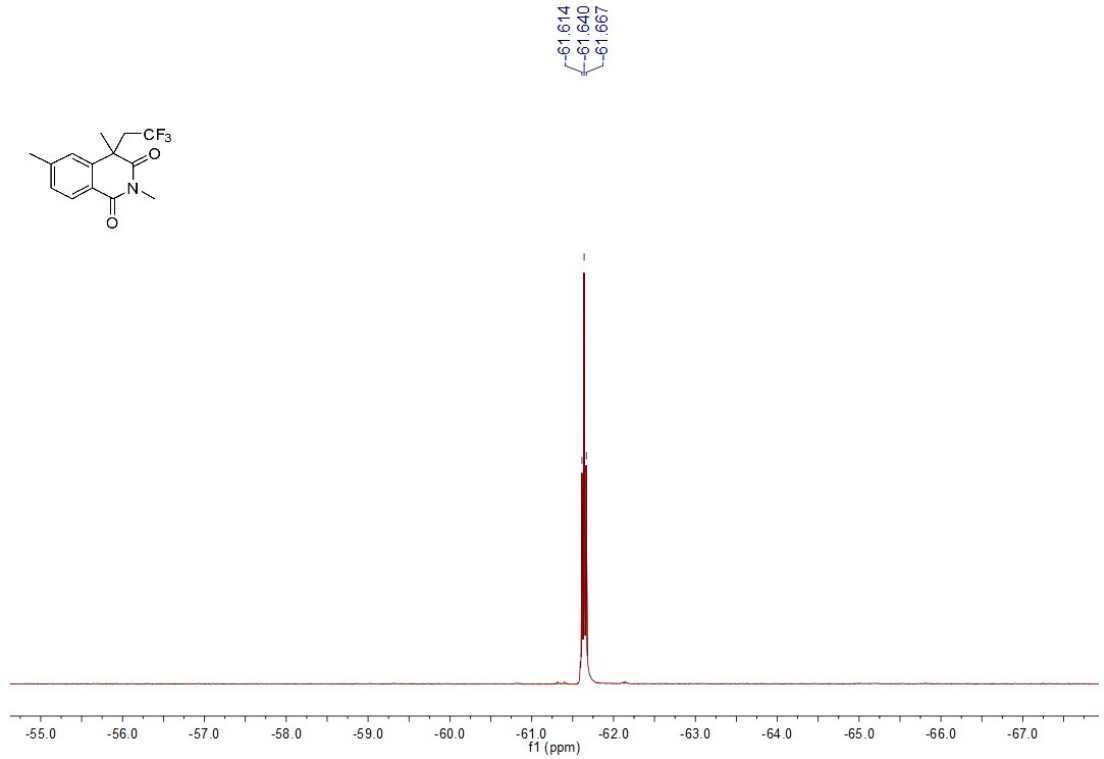
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5b**



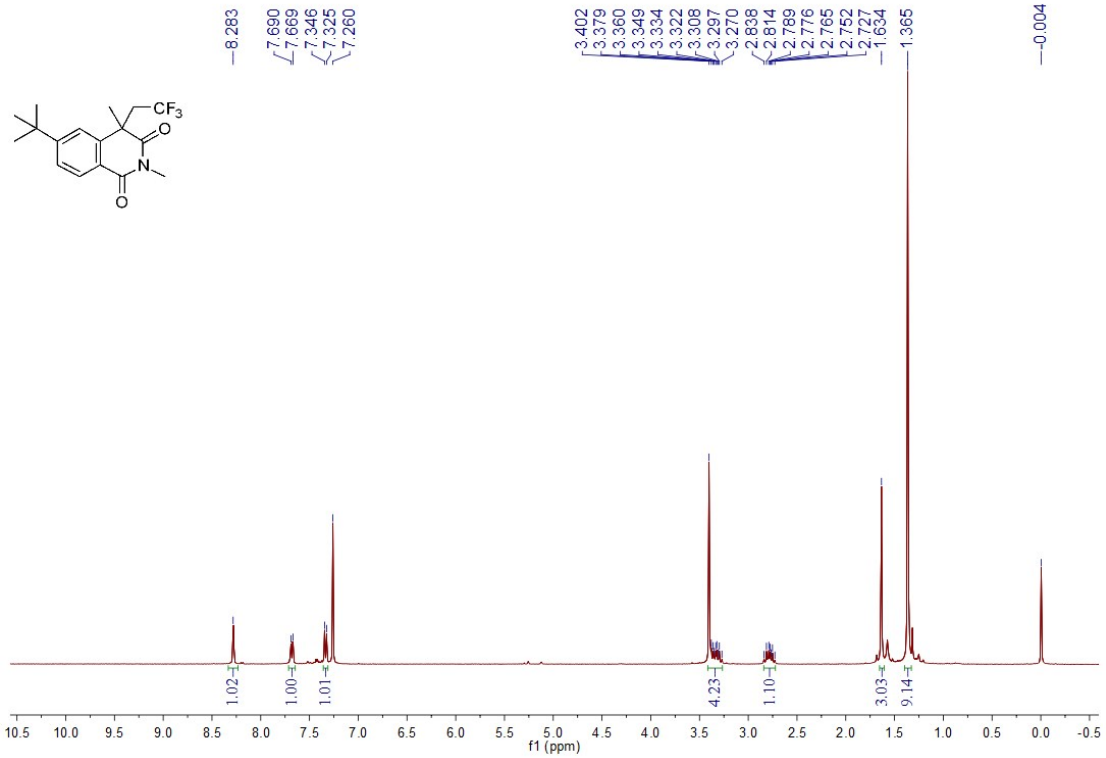
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5b**



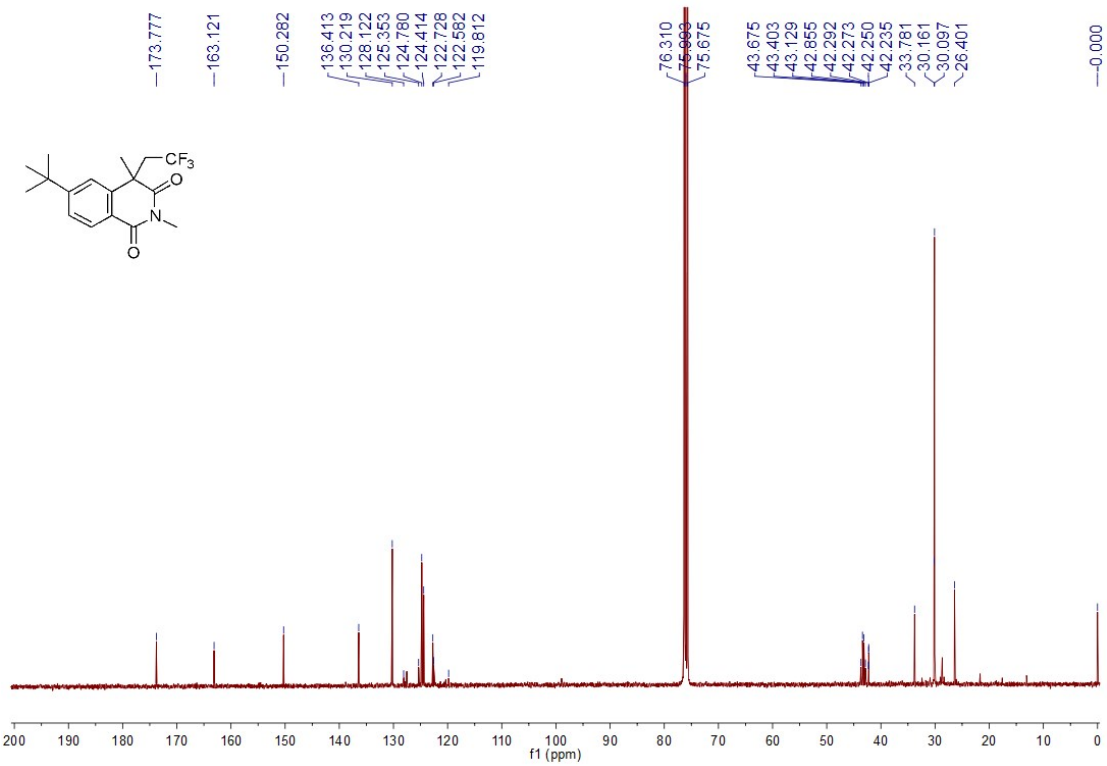
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5b**



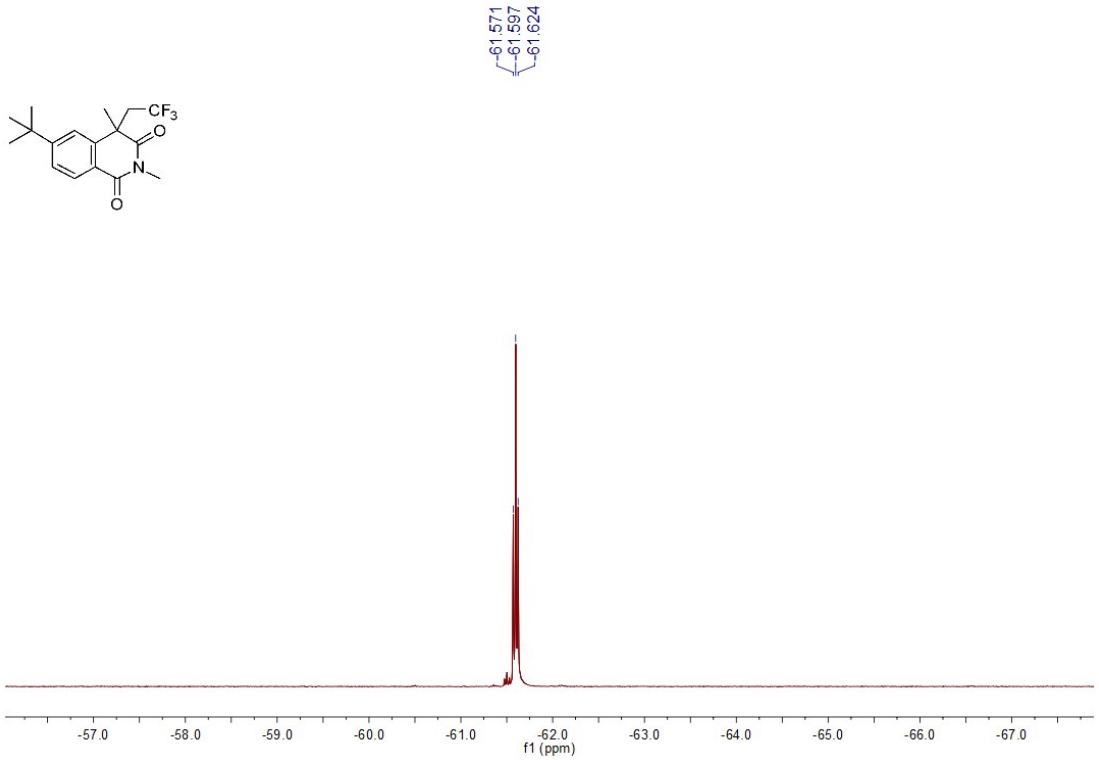
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5c**



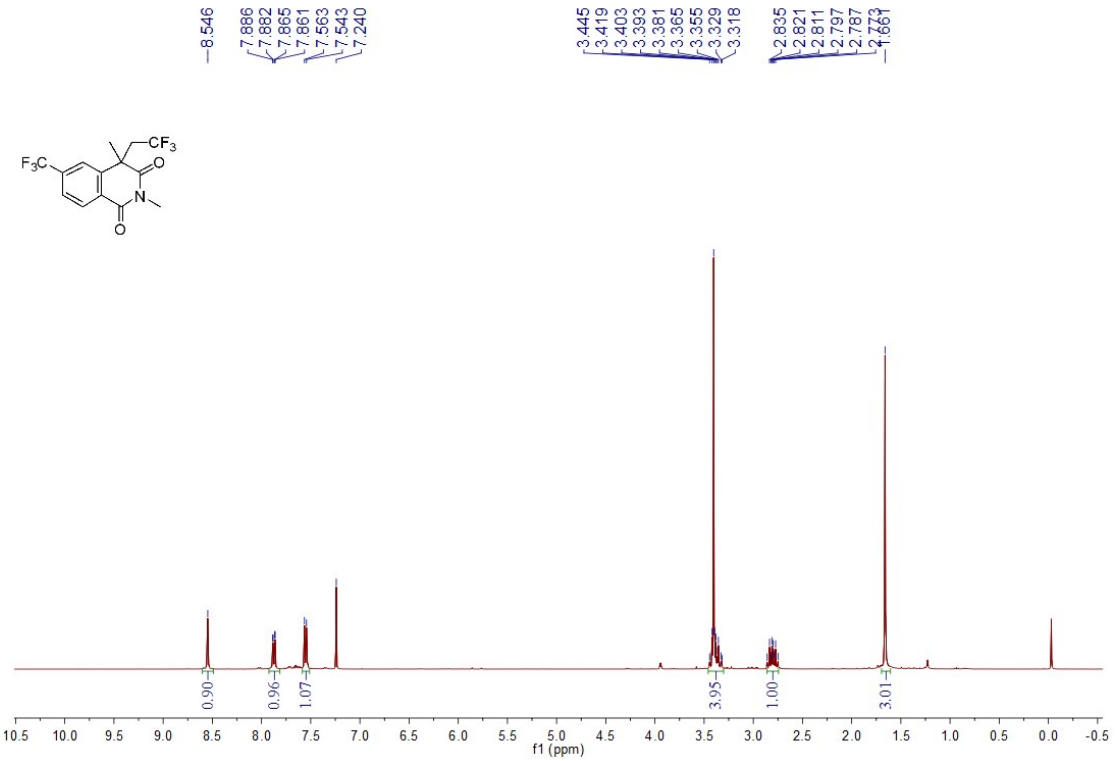
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5c**



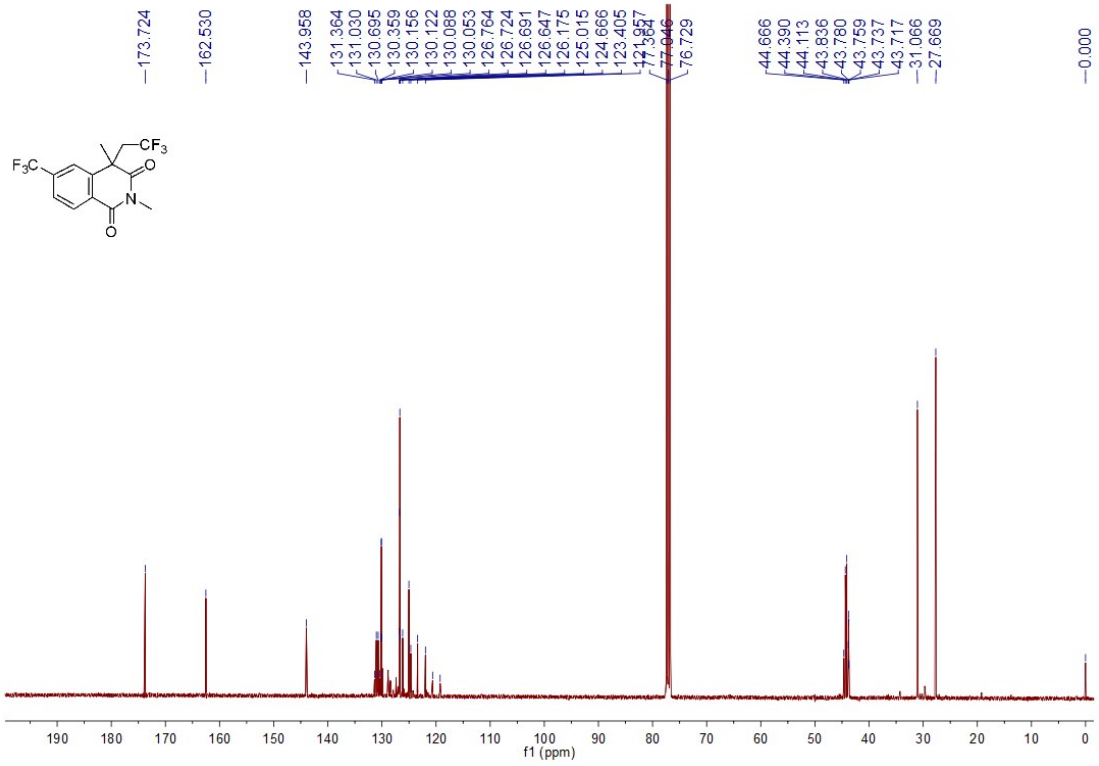
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5c**



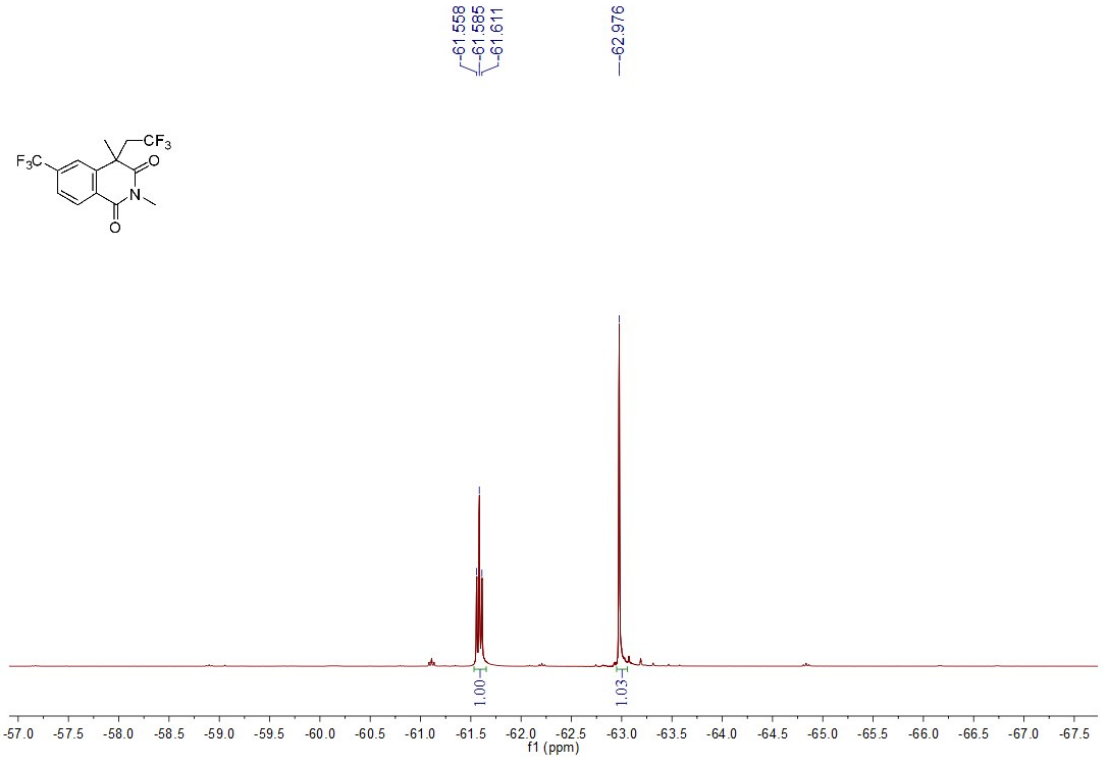
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5d**



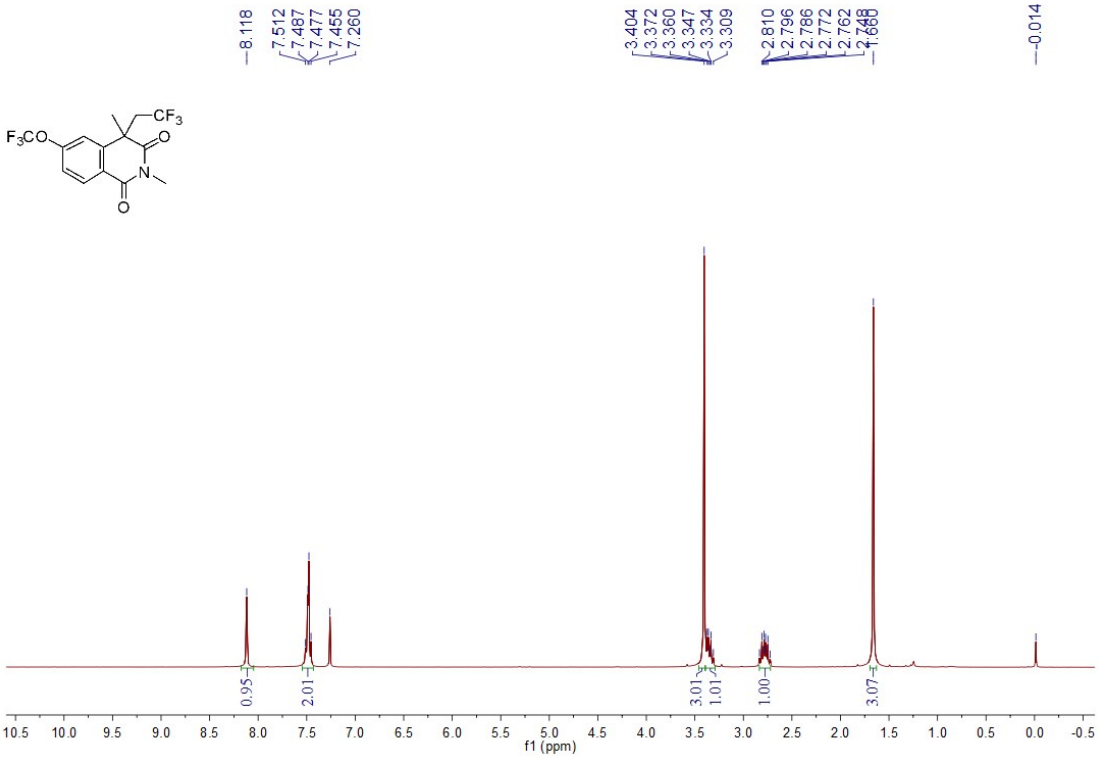
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5d**



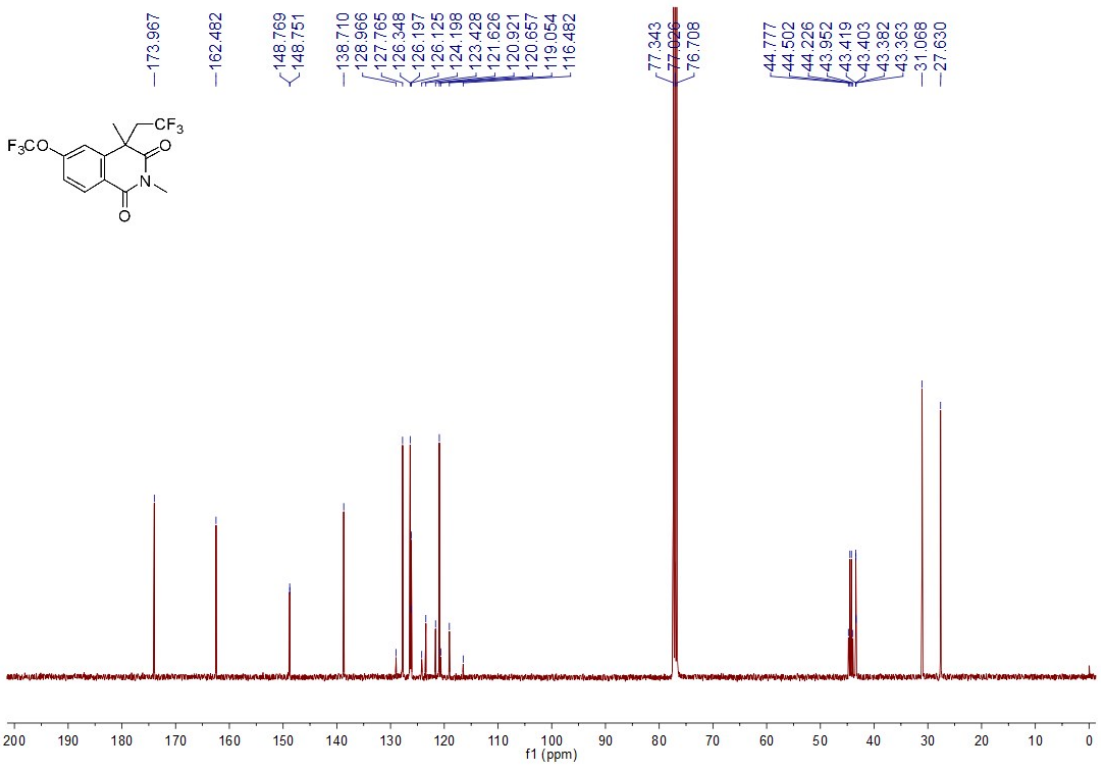
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5d**



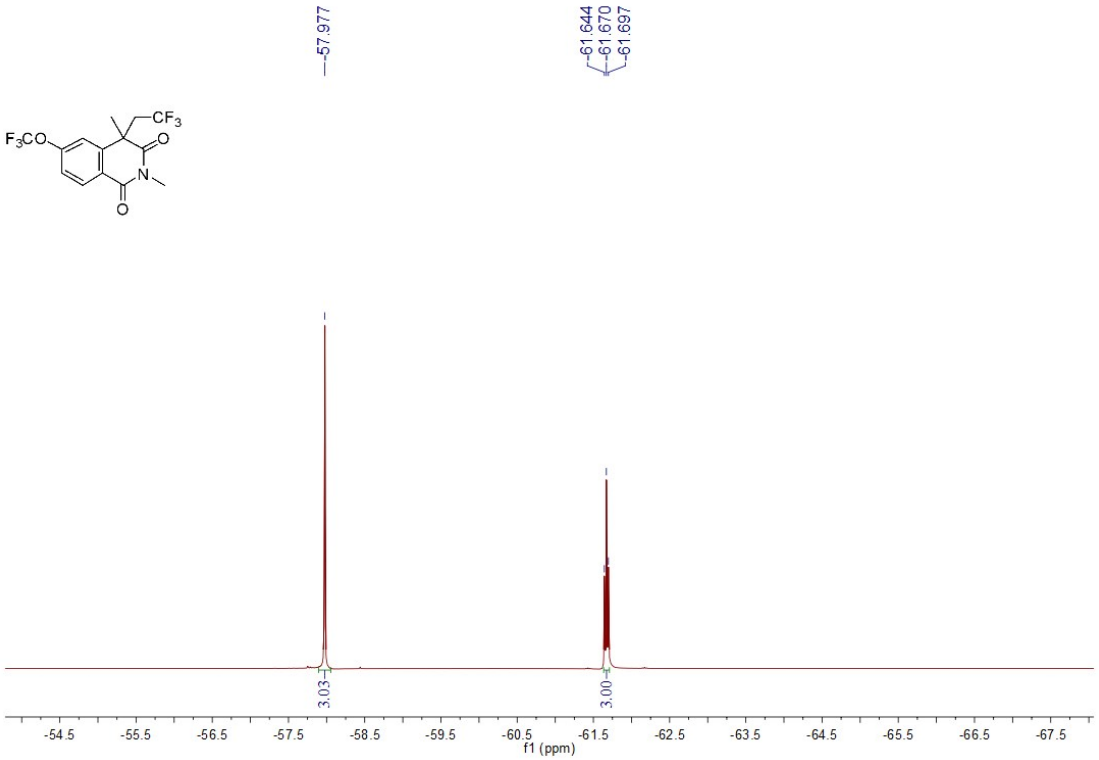
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5e**



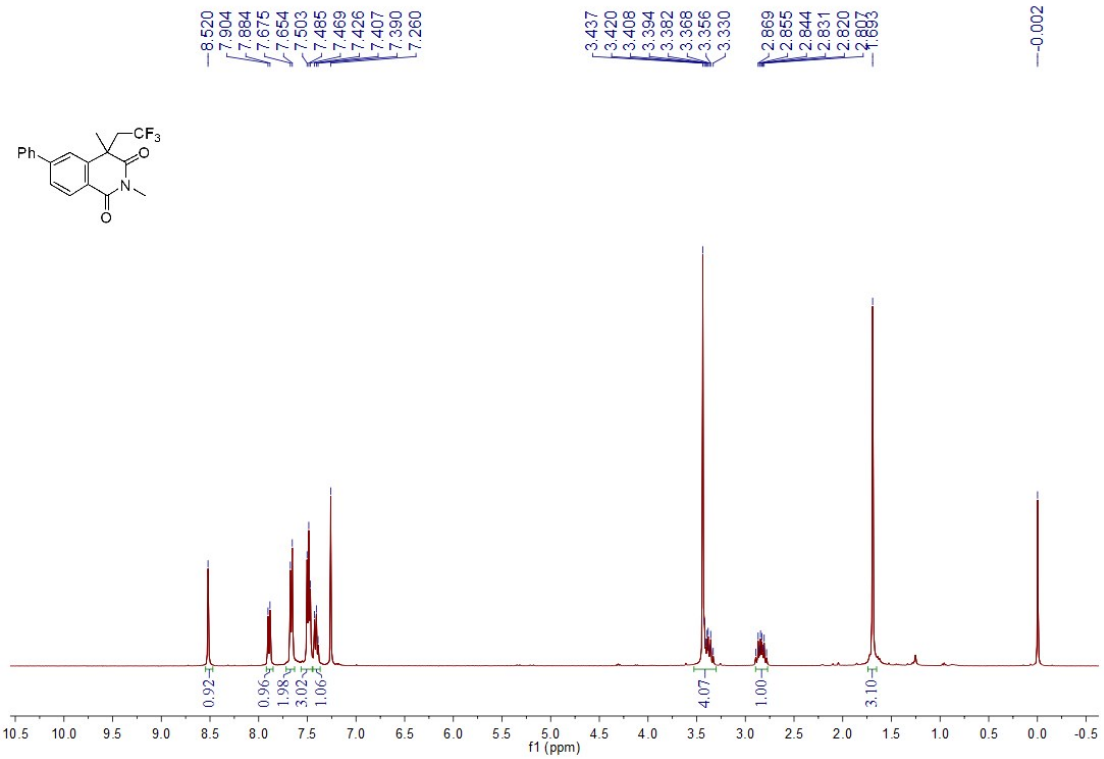
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5e**



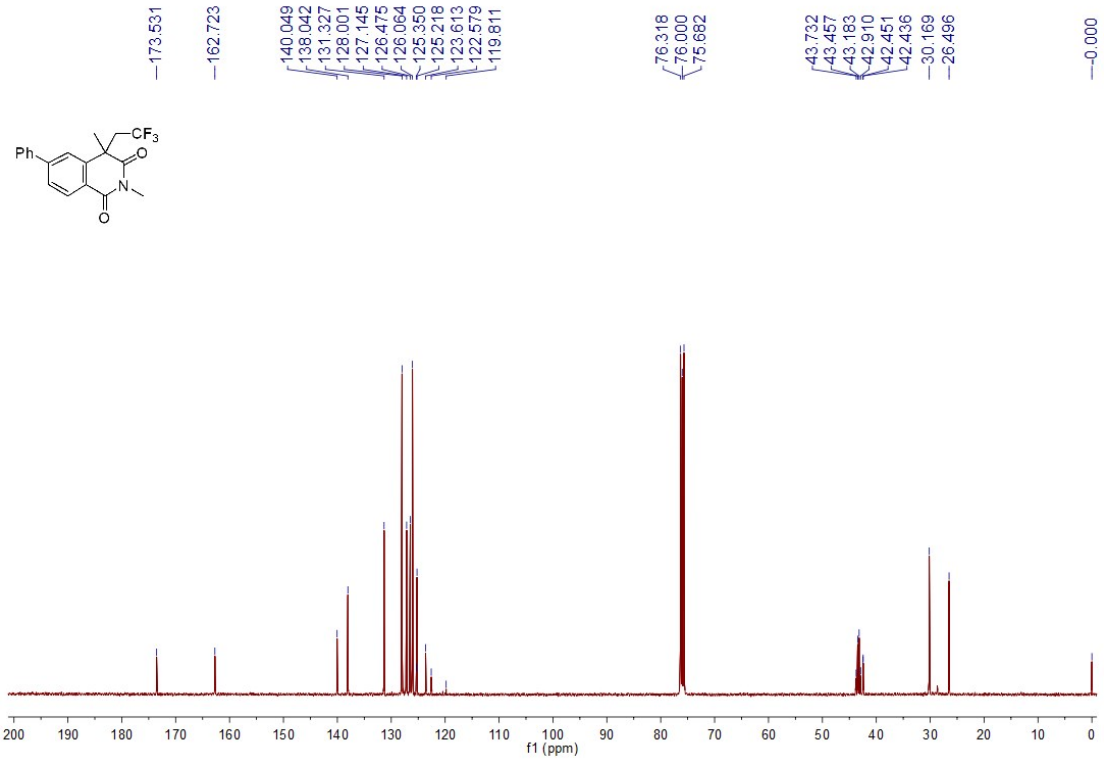
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5e**



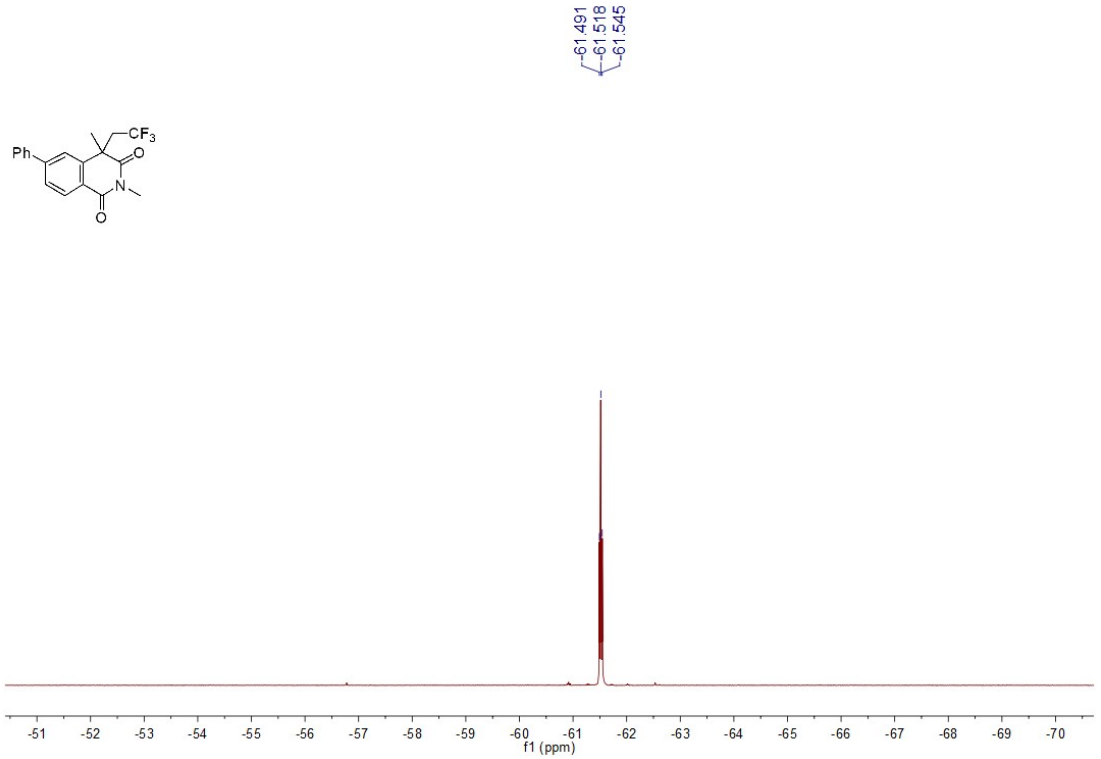
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5f**



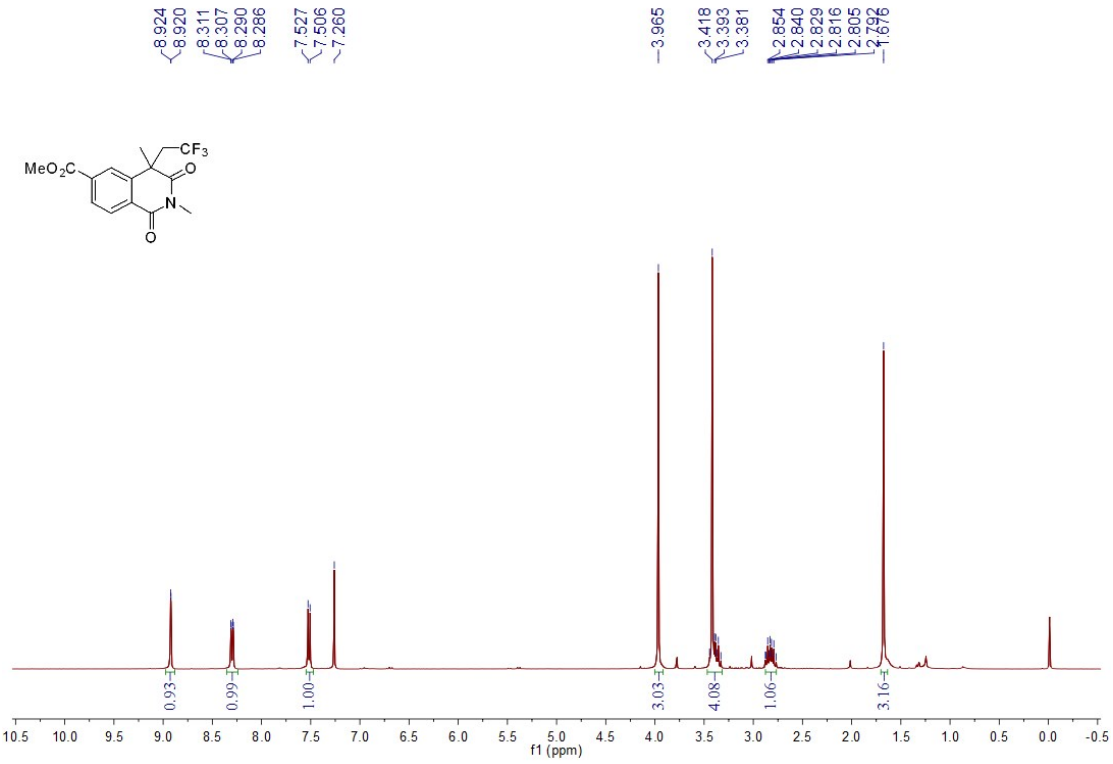
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5f**



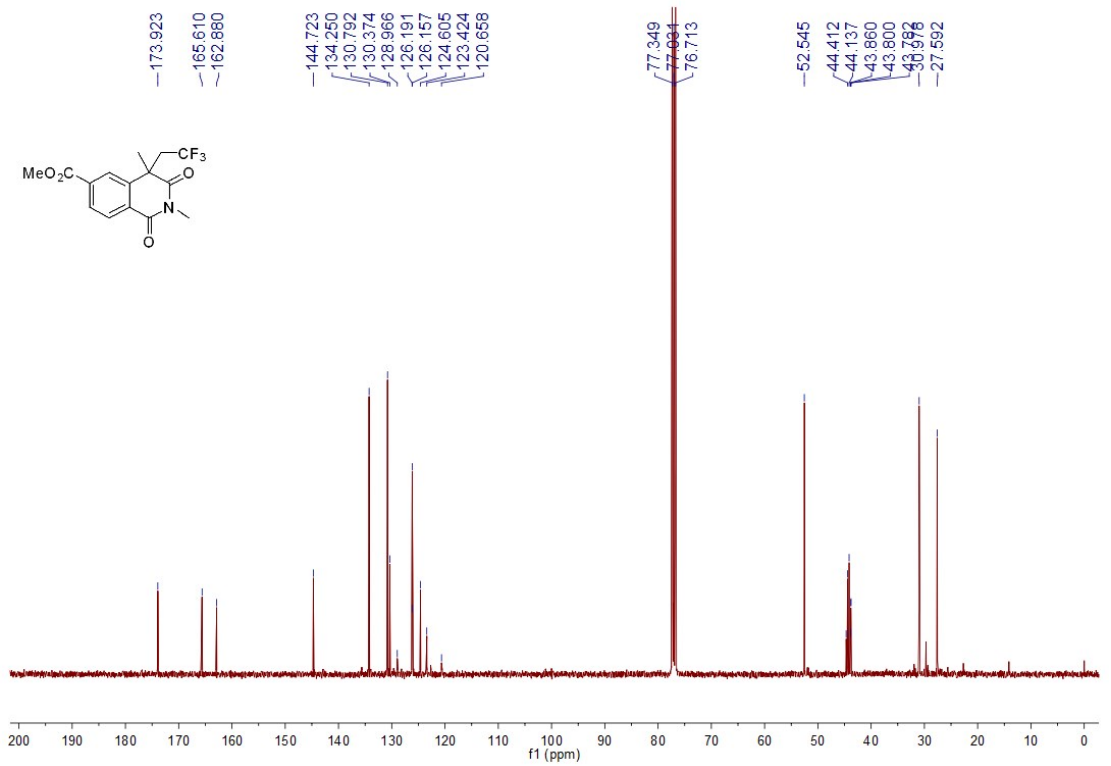
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5f**



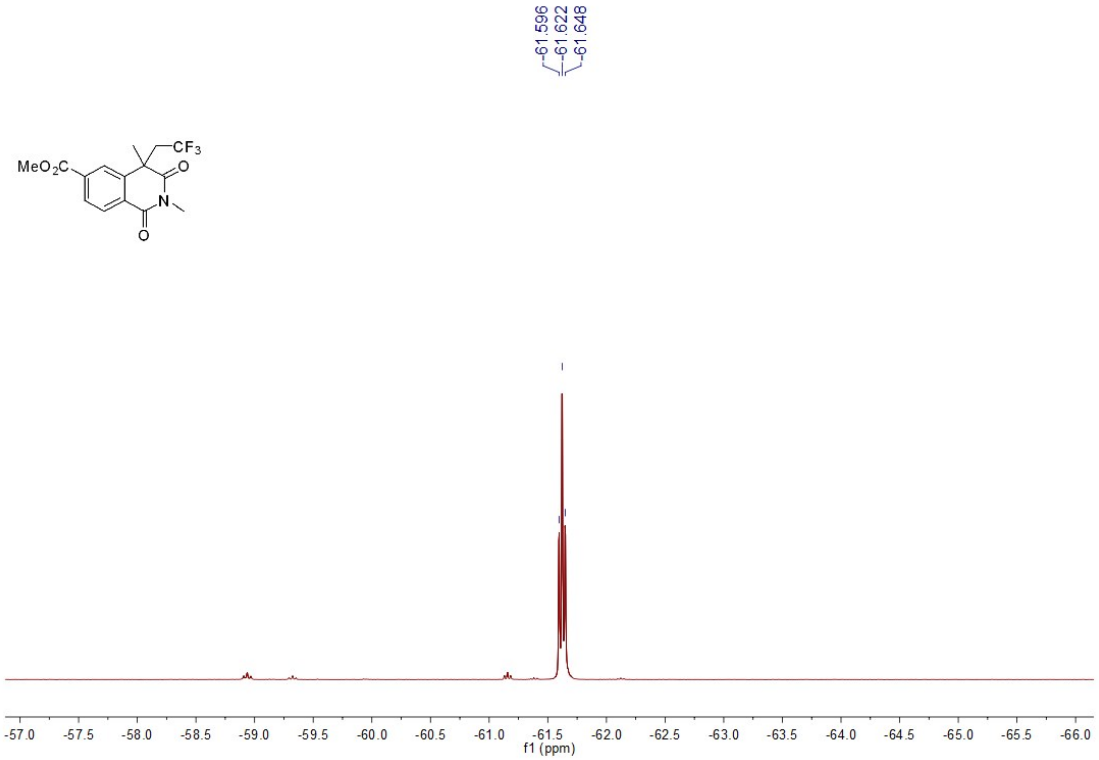
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5g**



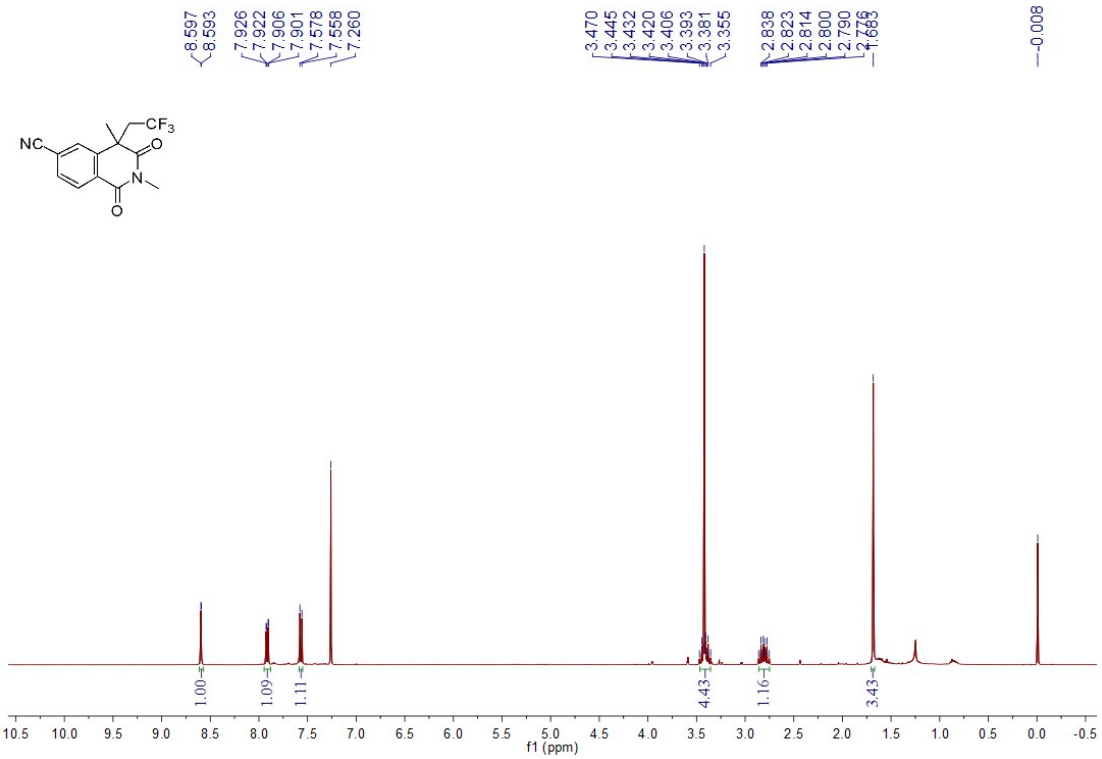
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5g**



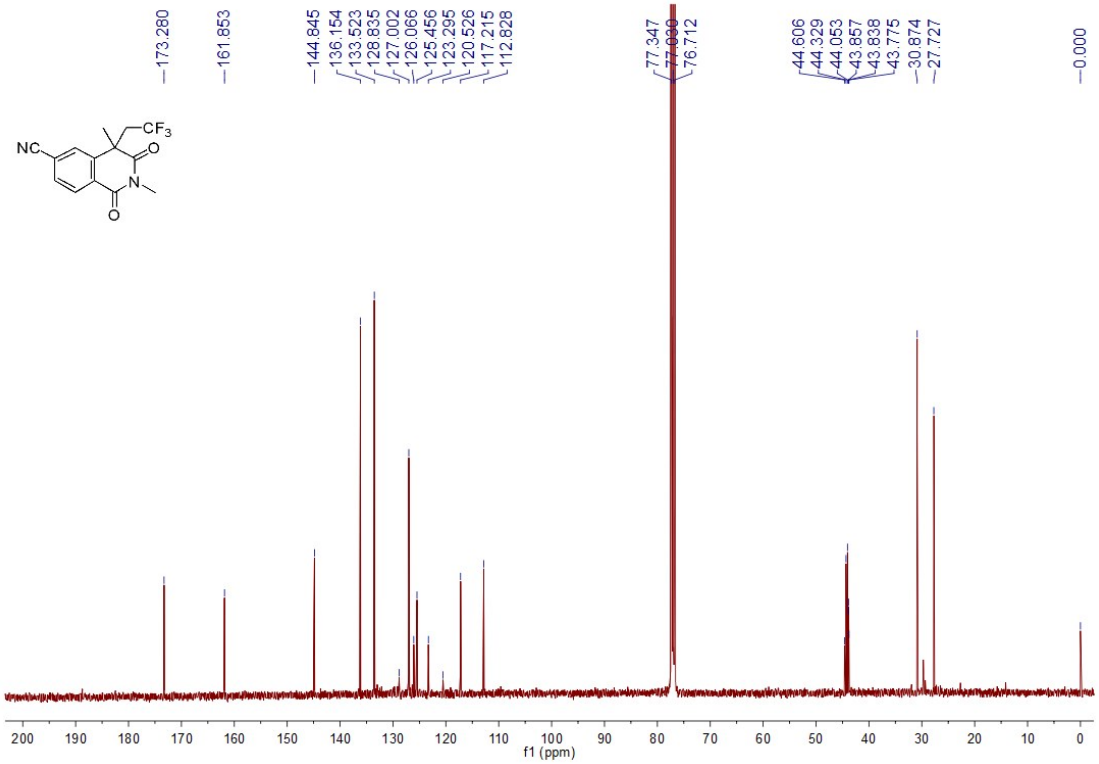
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5g**



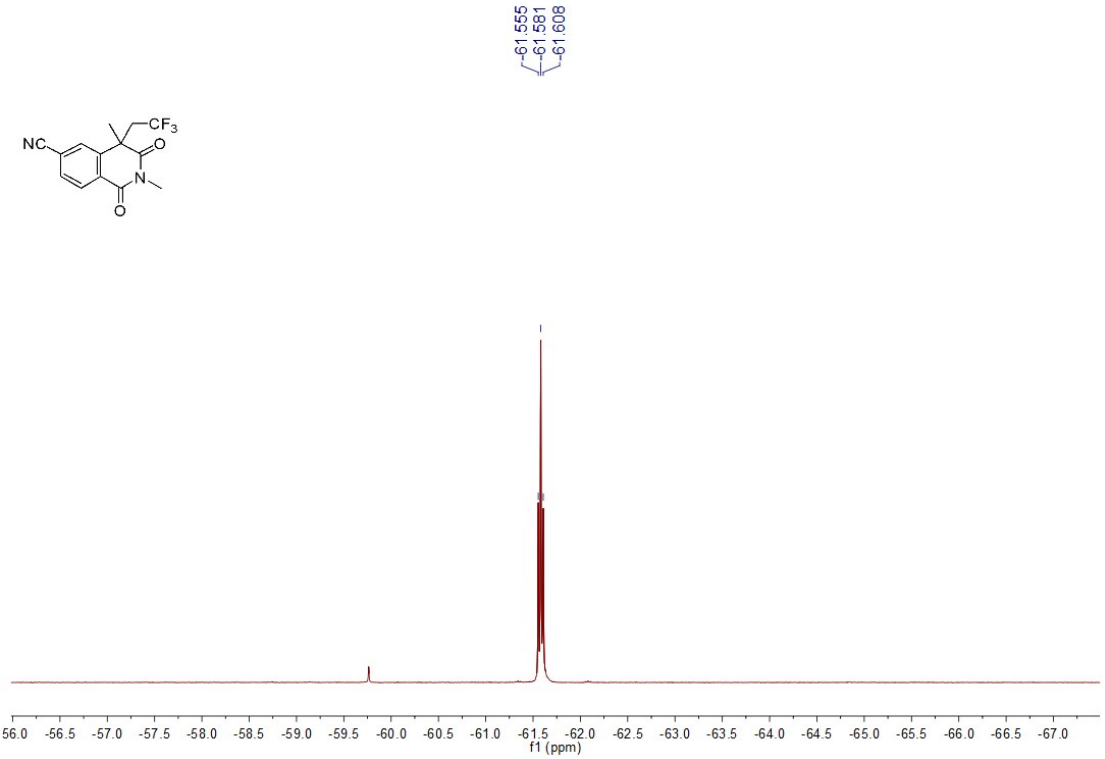
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5h**



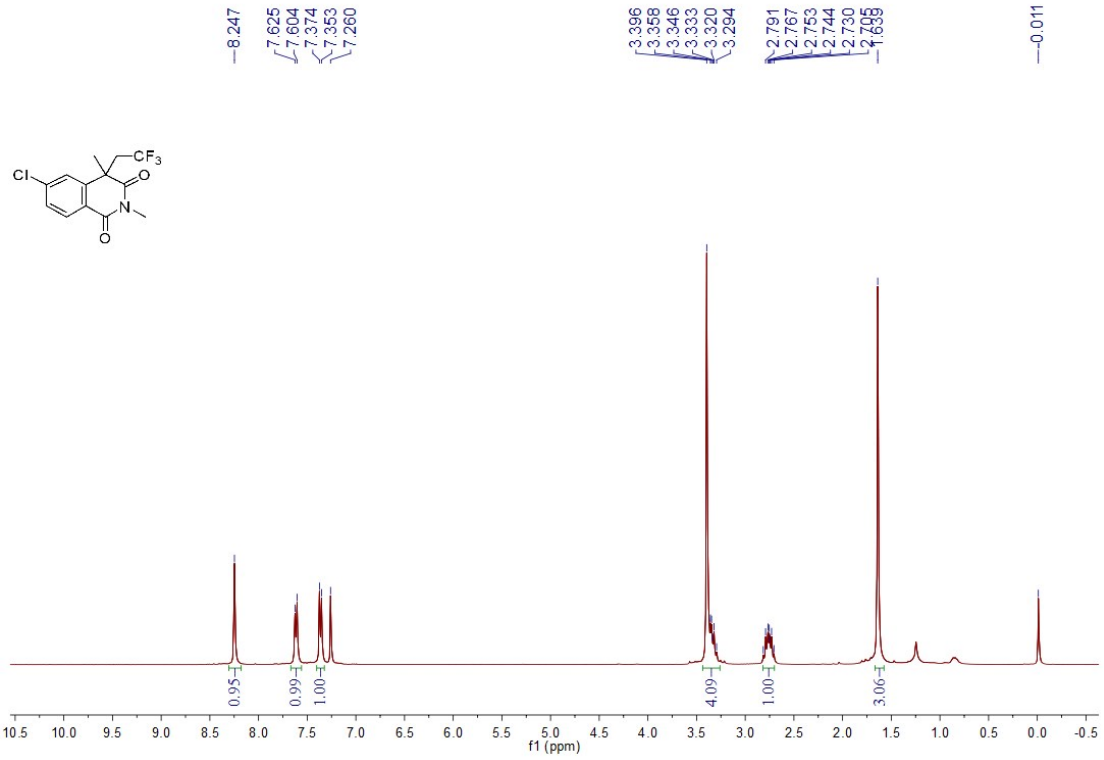
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5h**



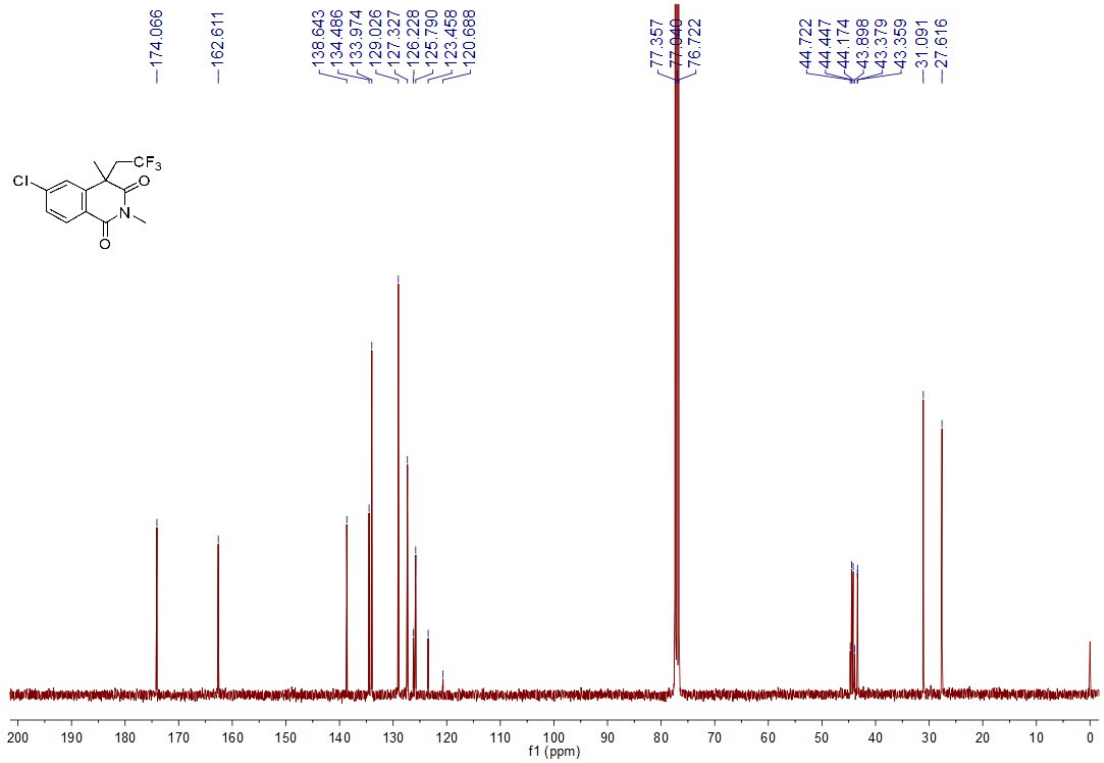
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5h**



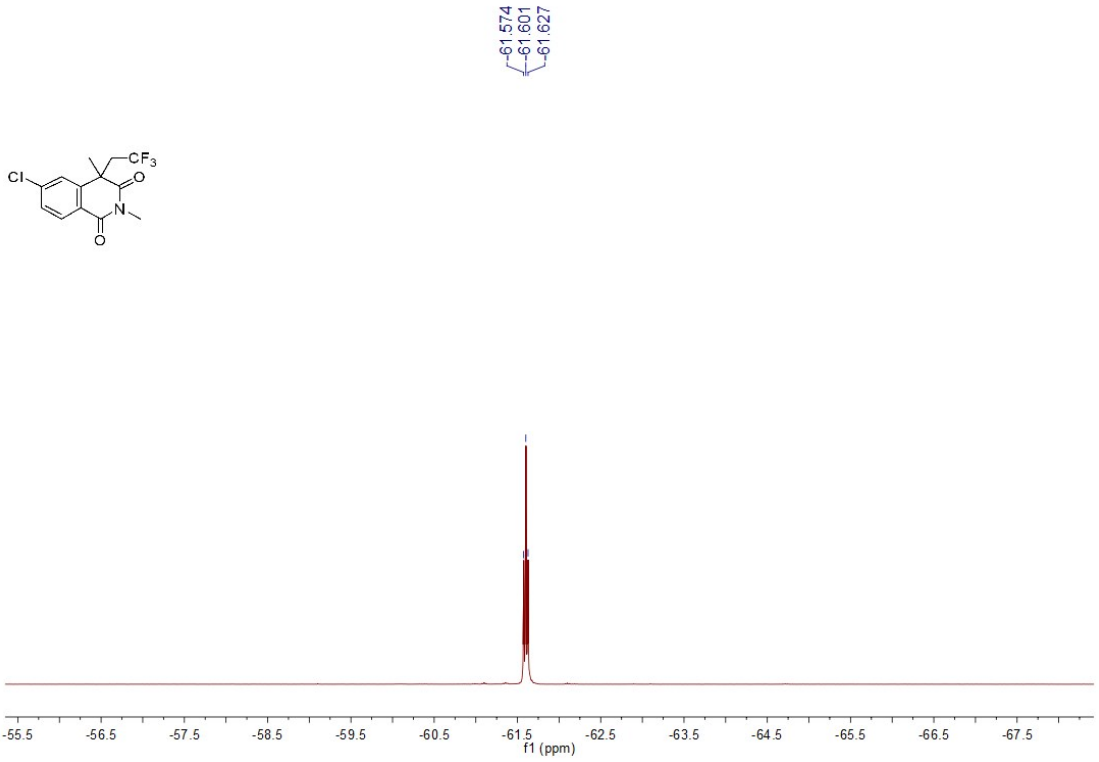
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5i**



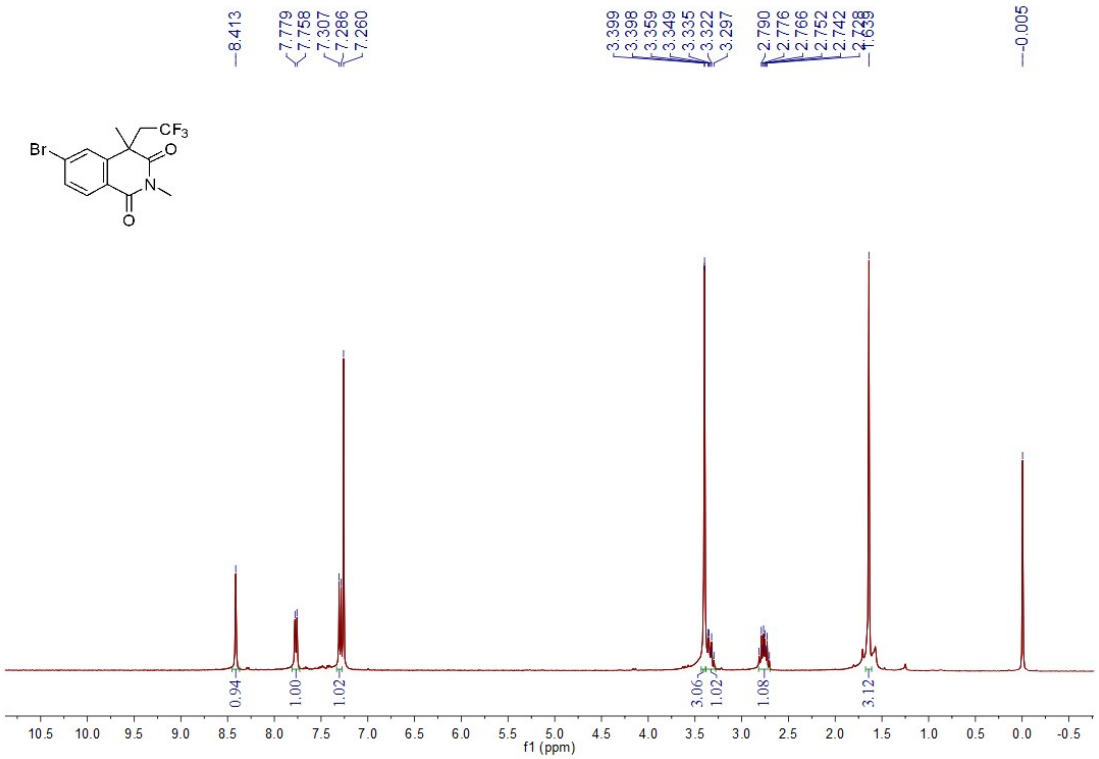
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5i**



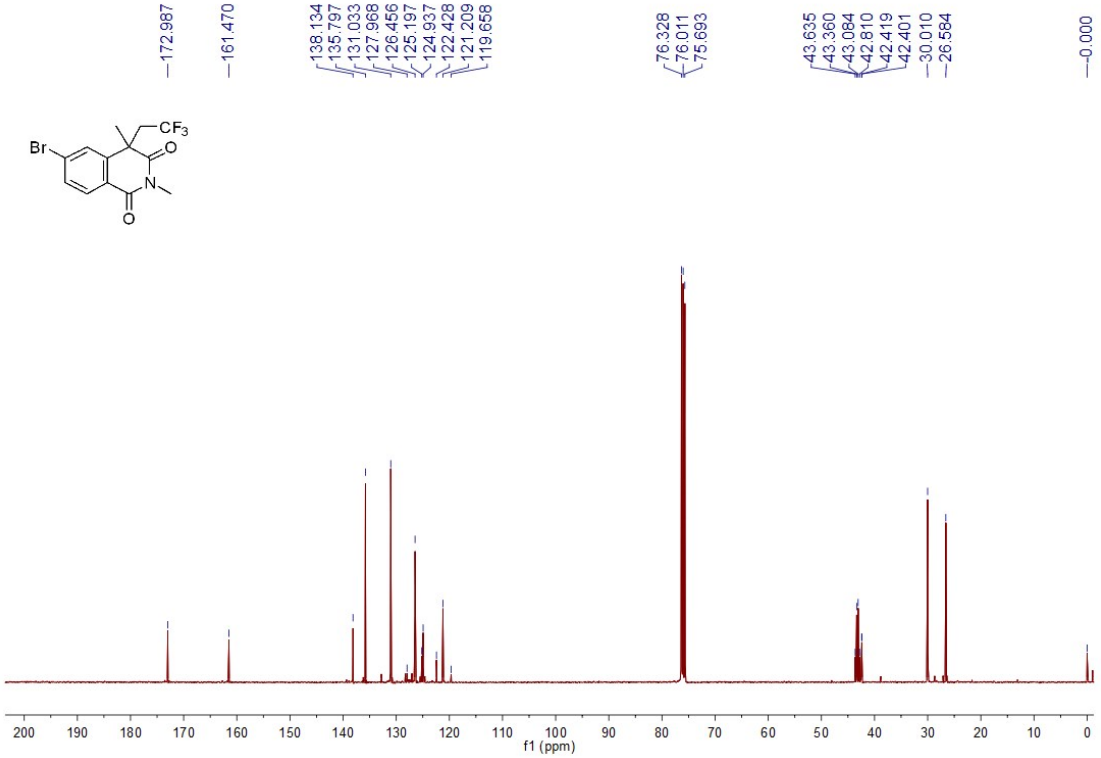
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5i**



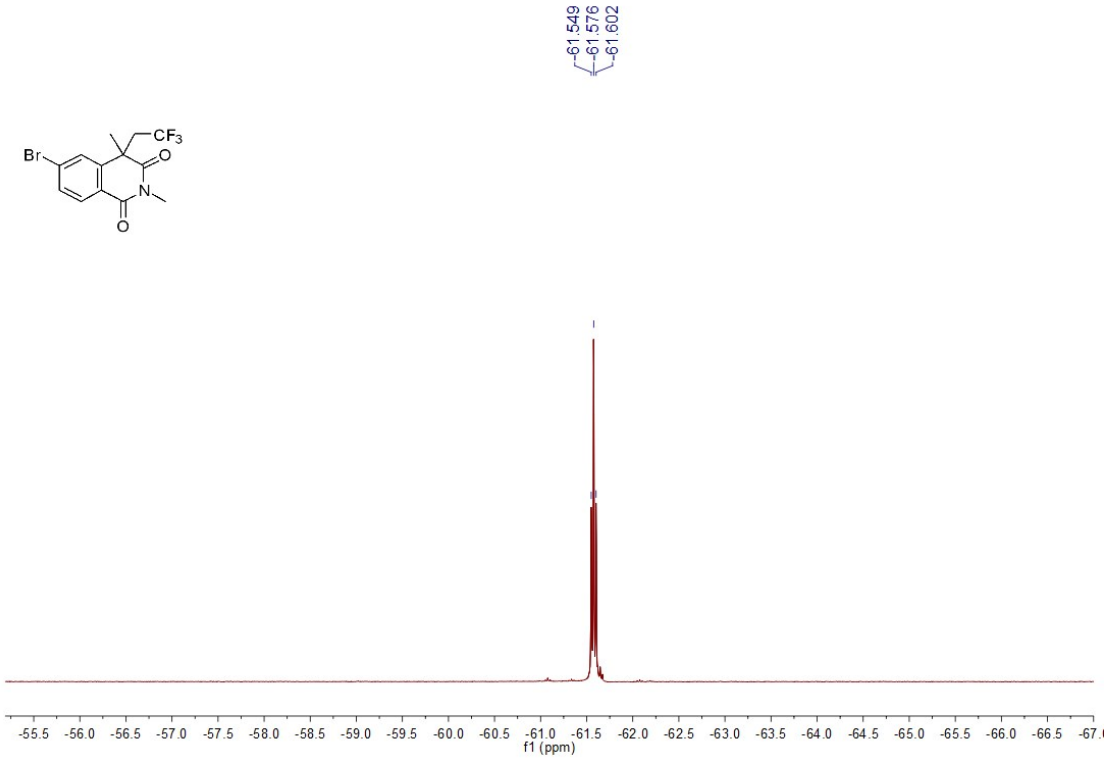
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5j**



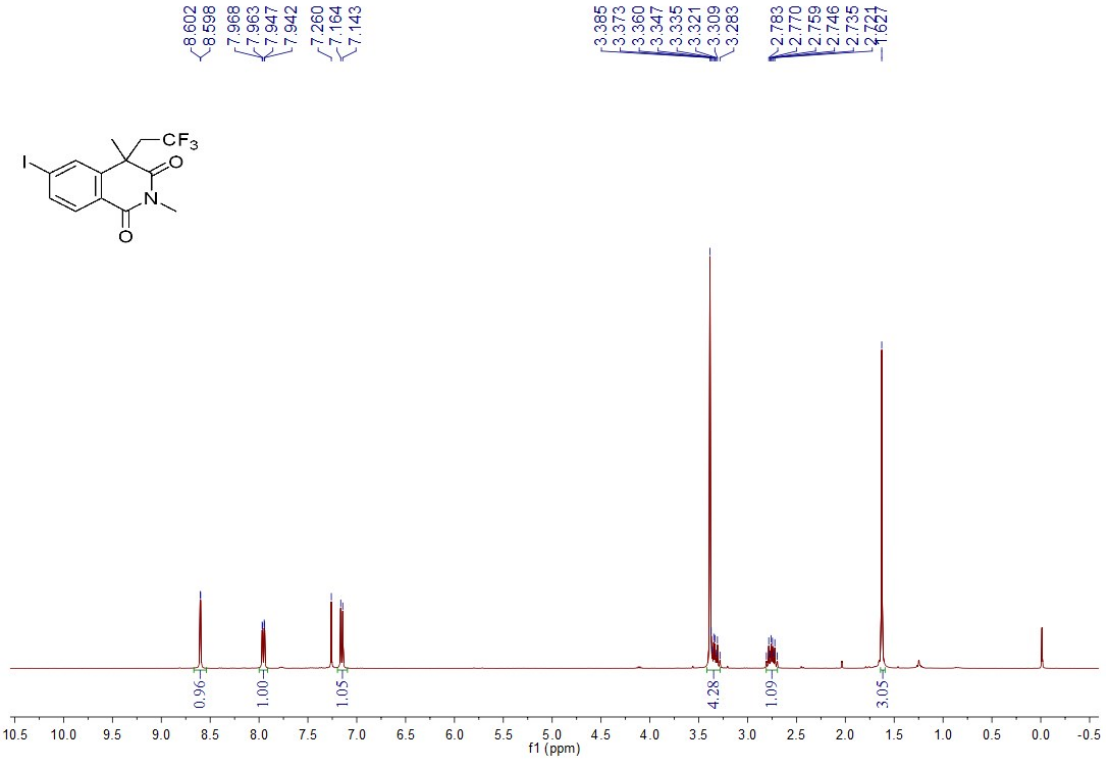
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5j**



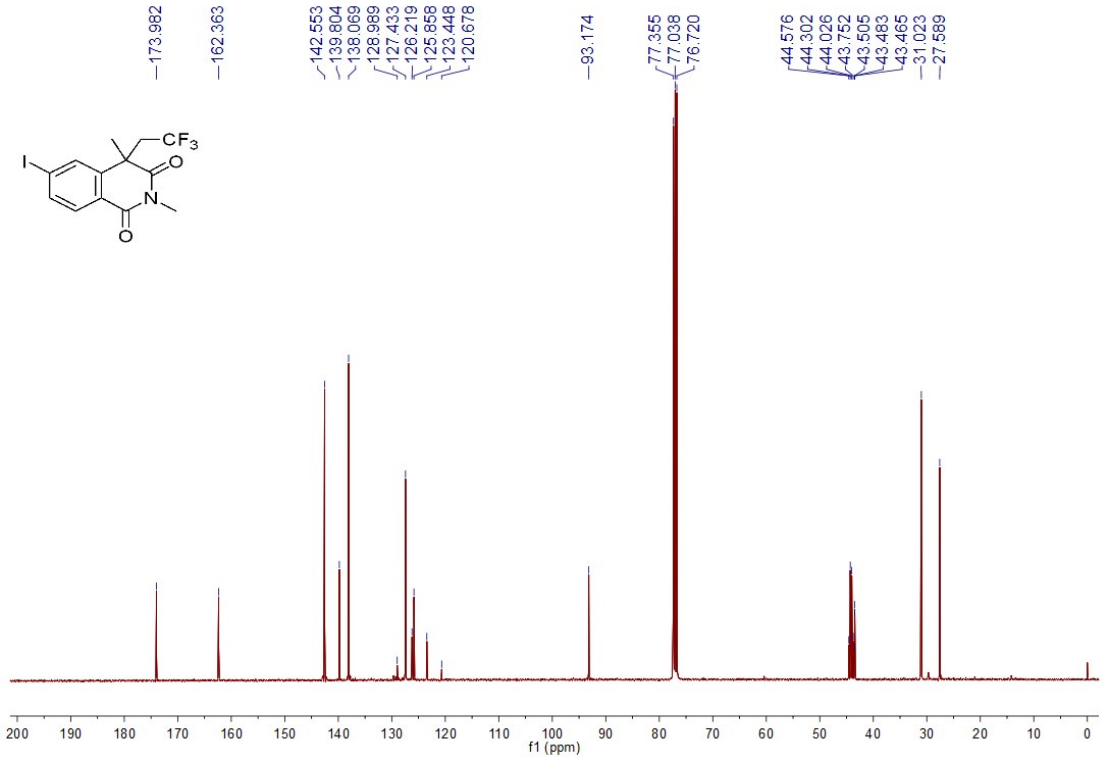
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5j**



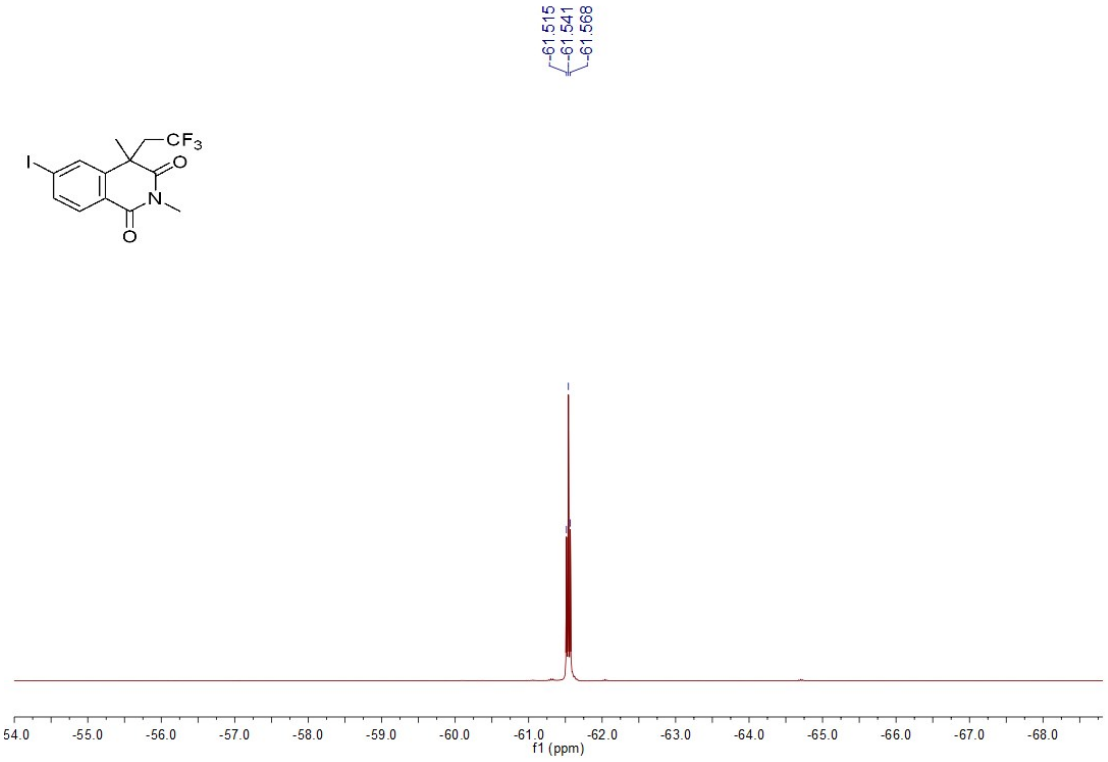
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5k**



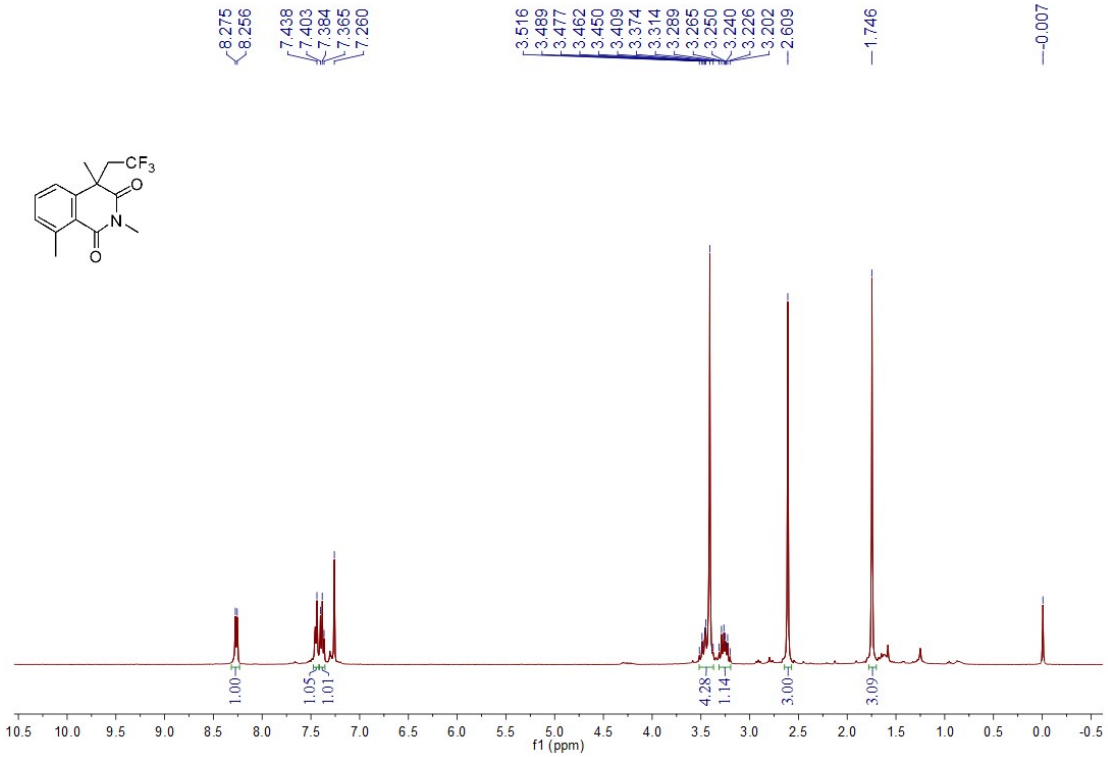
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5k**



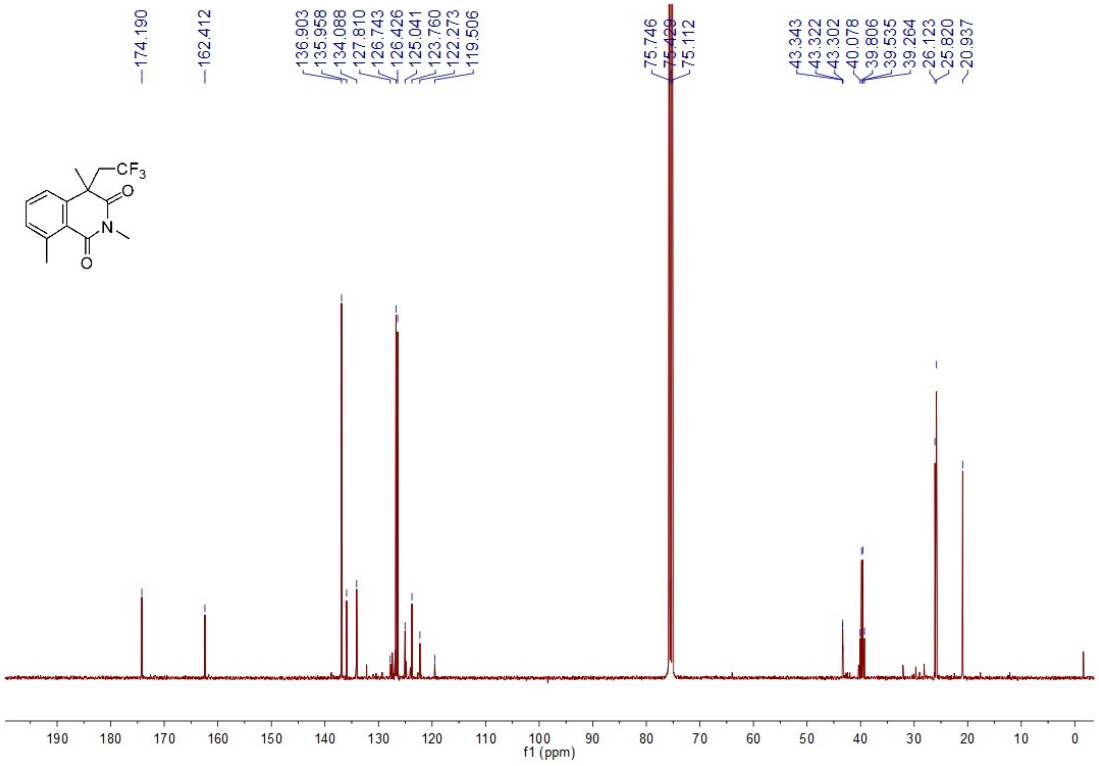
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5k**



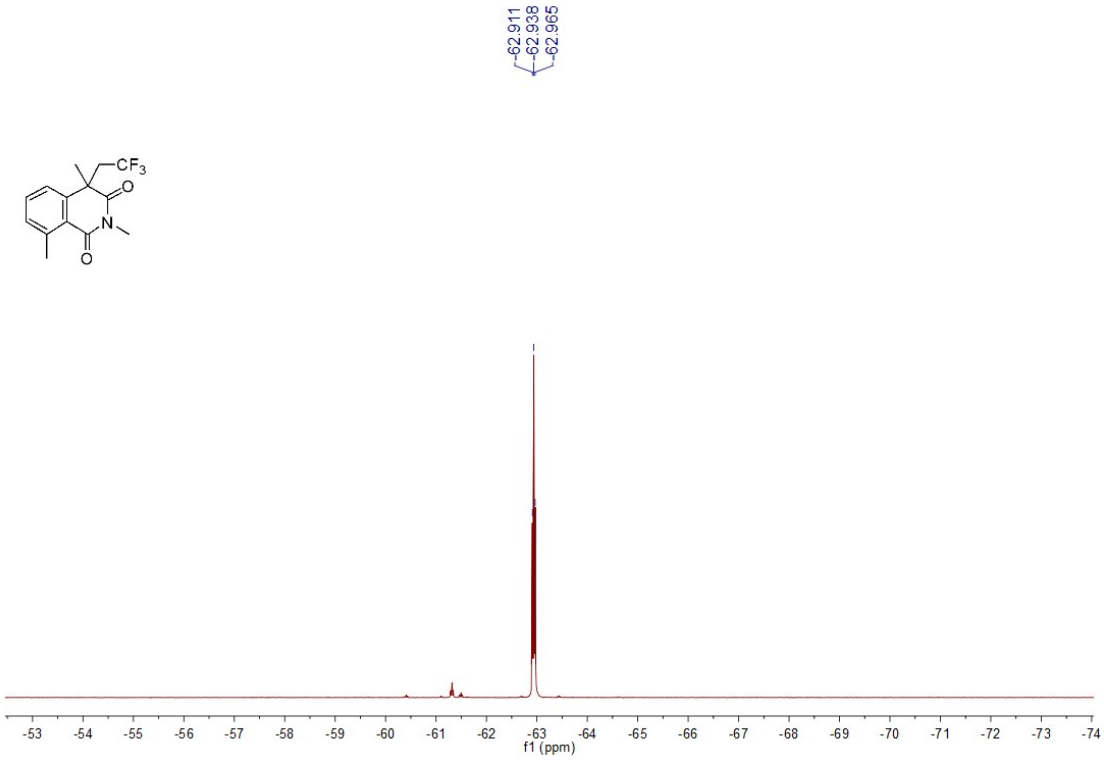
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5l**



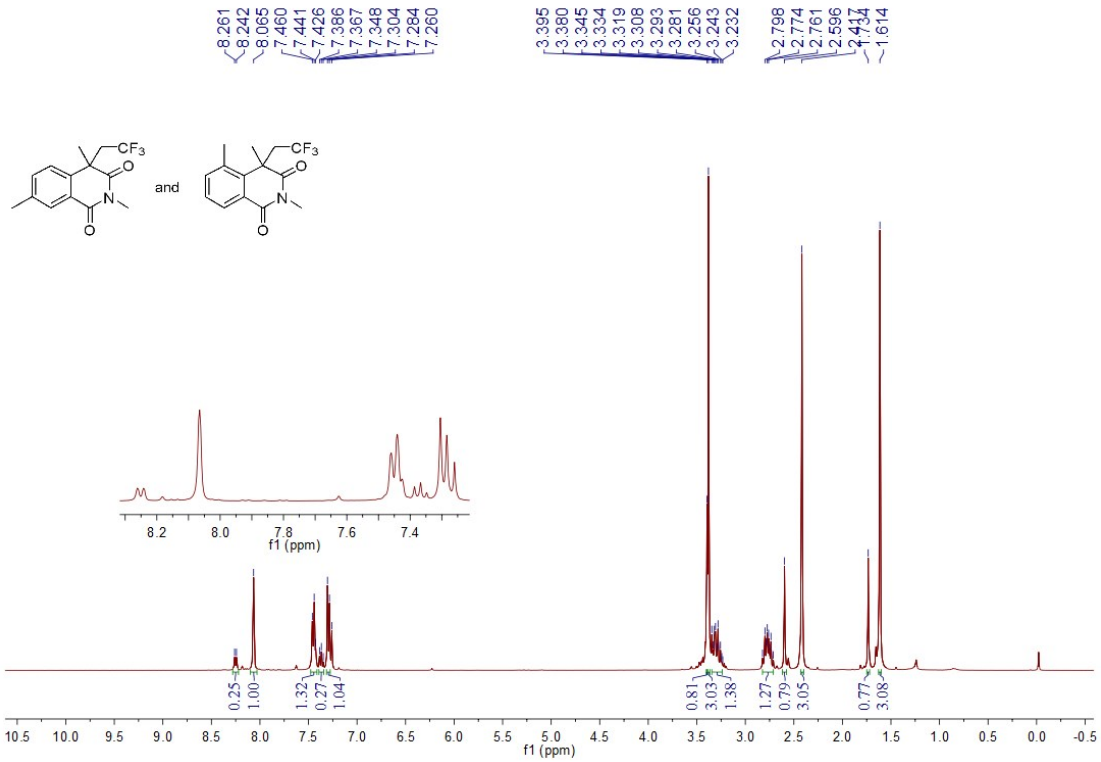
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5l**



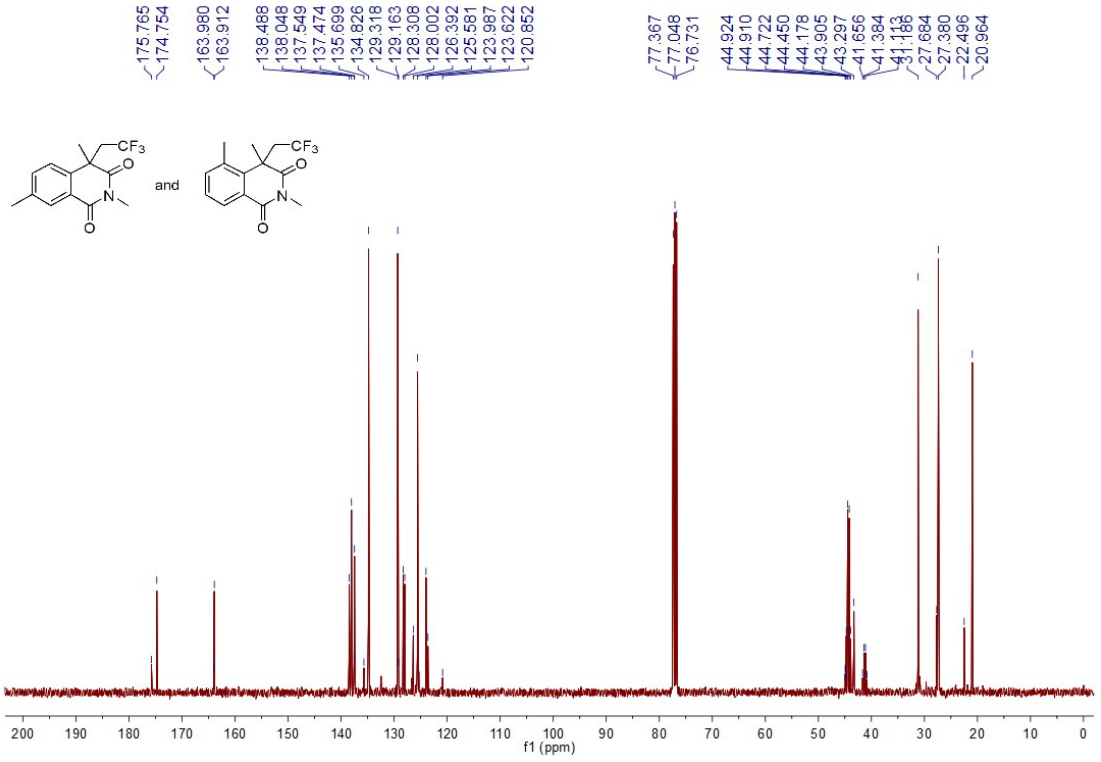
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5l**



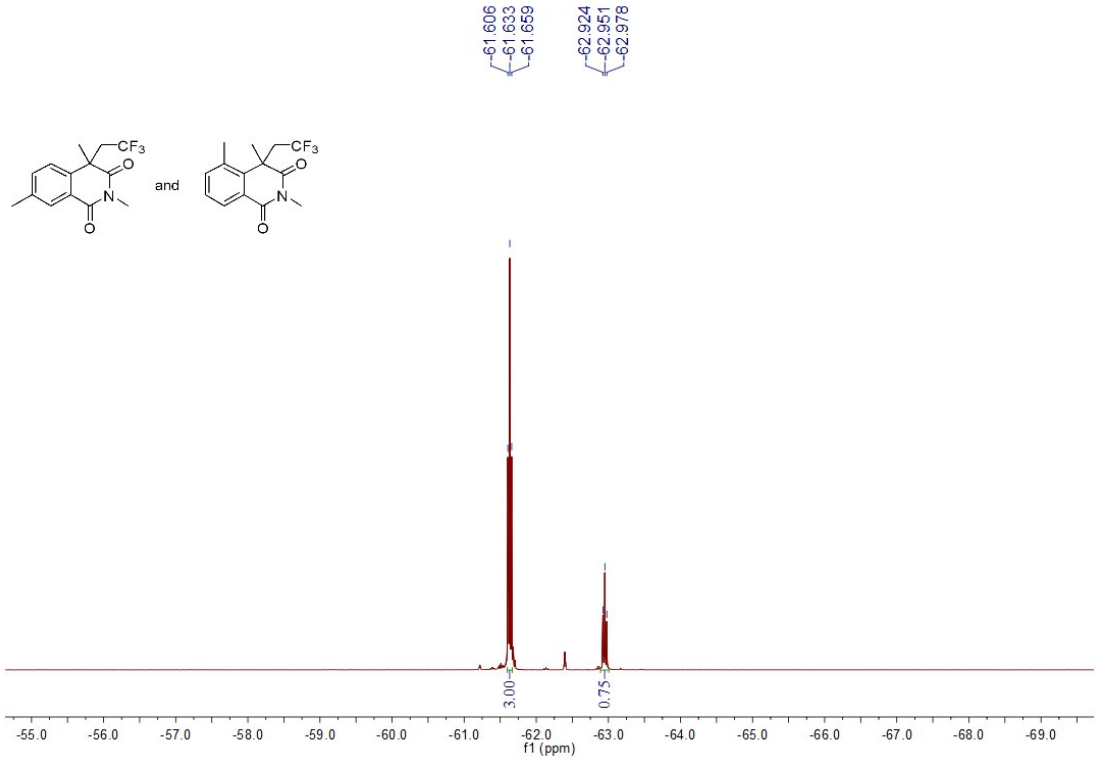
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5m**



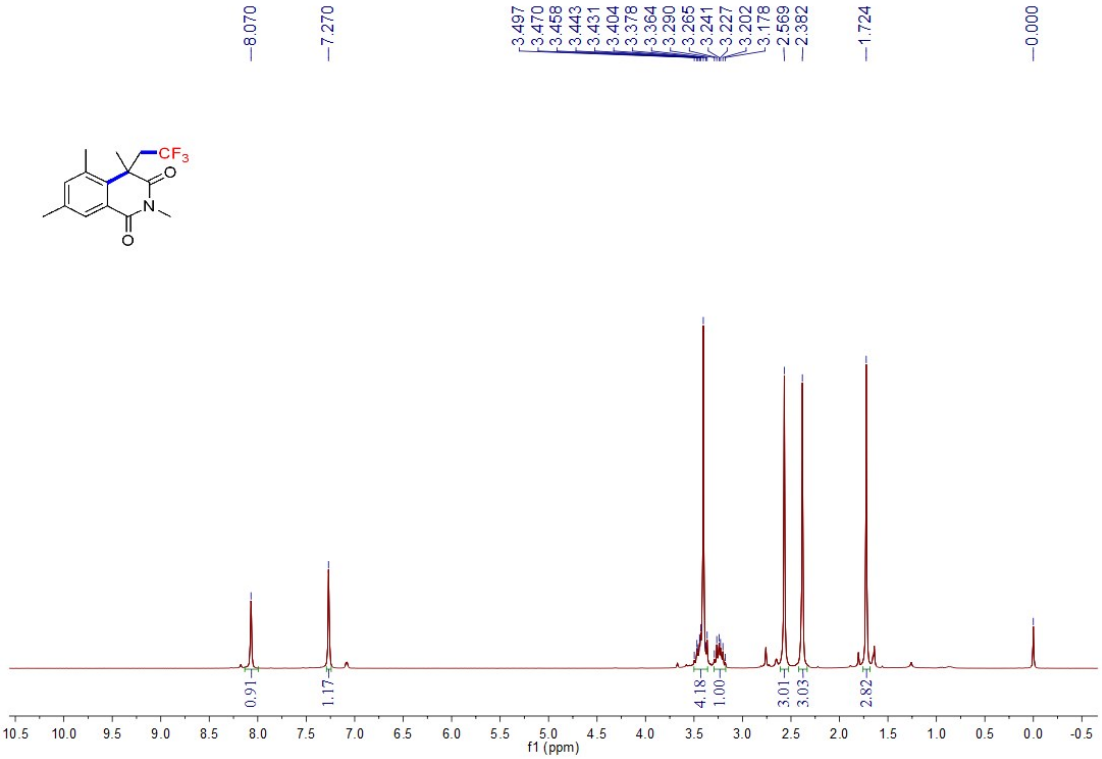
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5m**



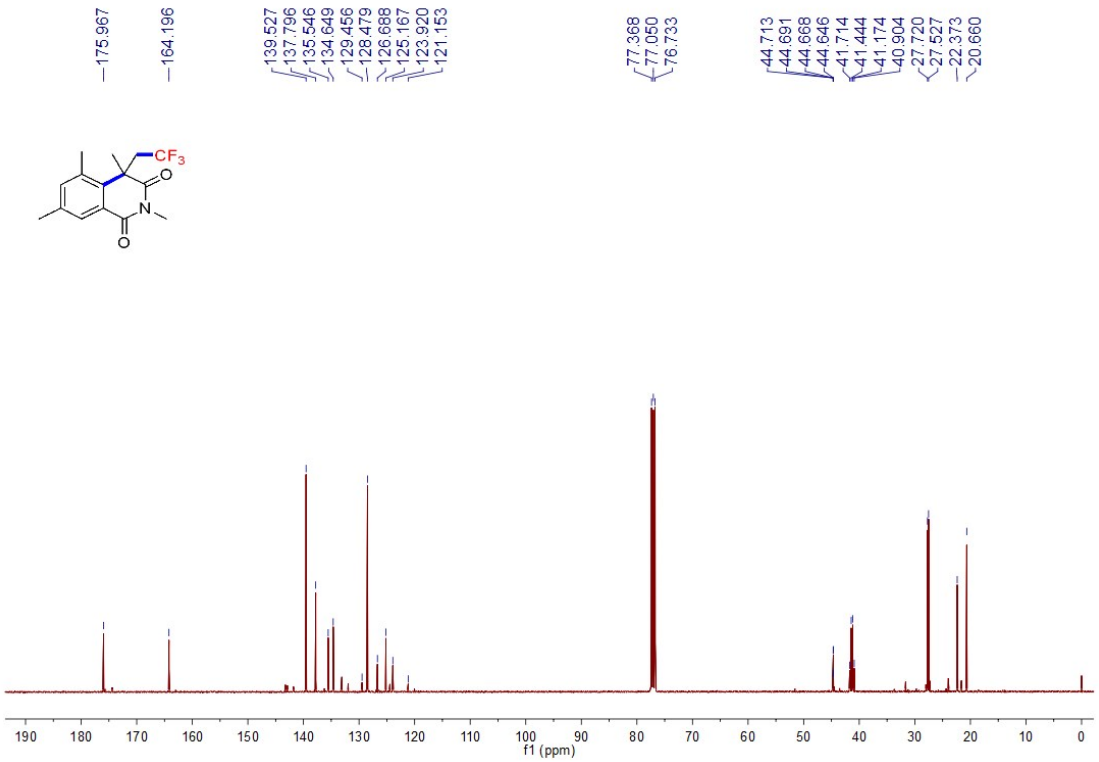
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5m**



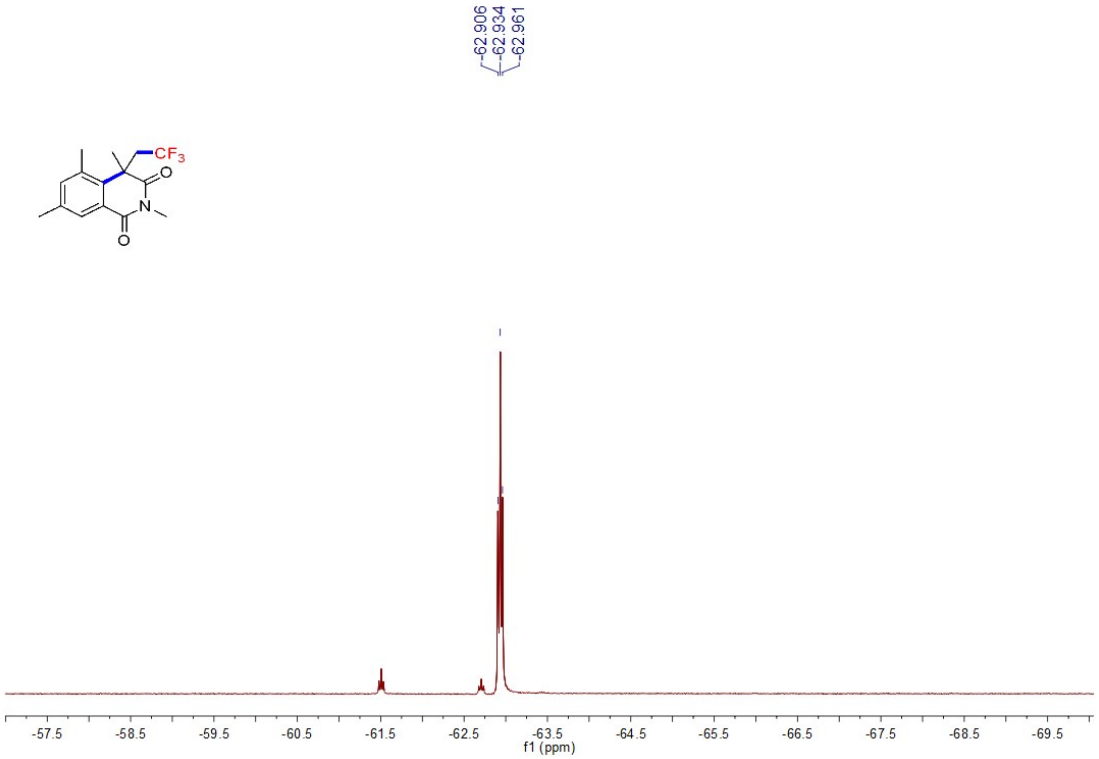
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5n**



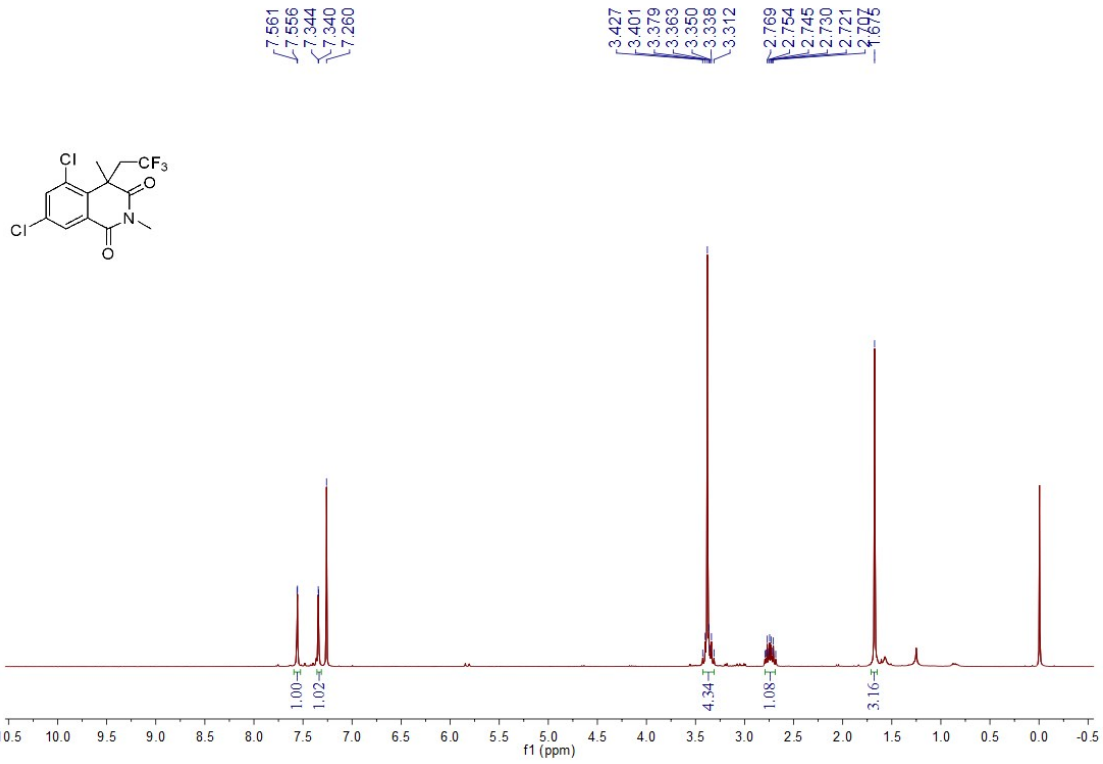
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5n**



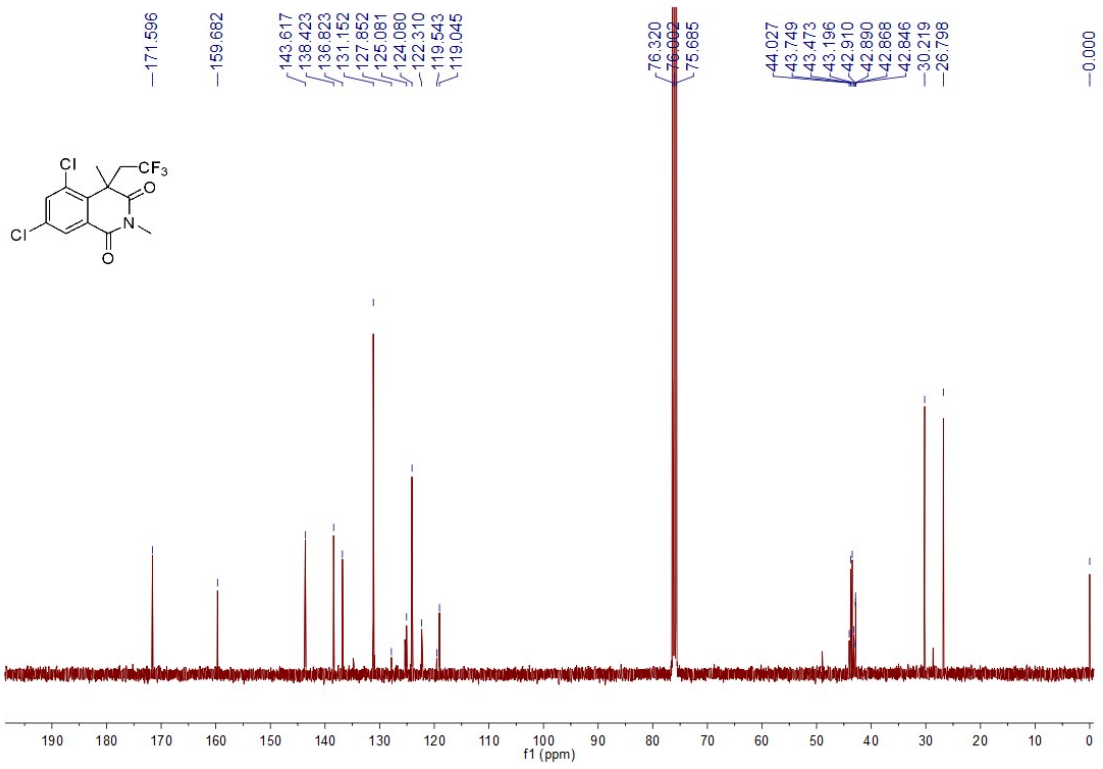
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5n**



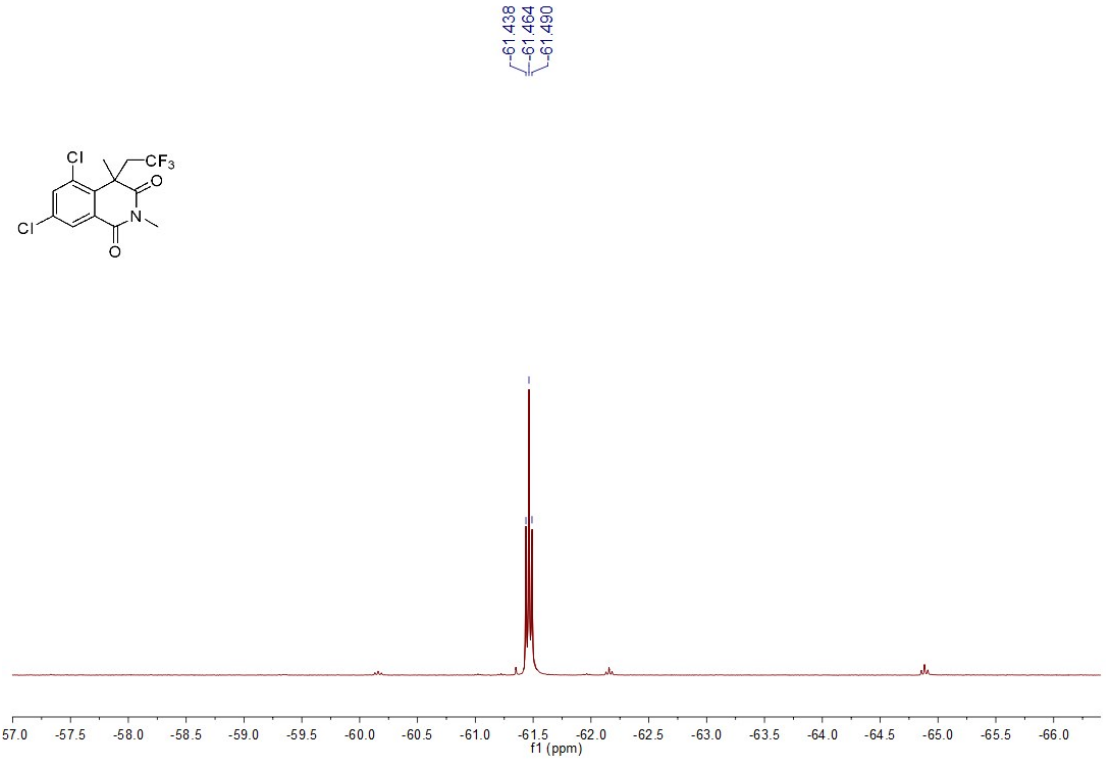
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5o**



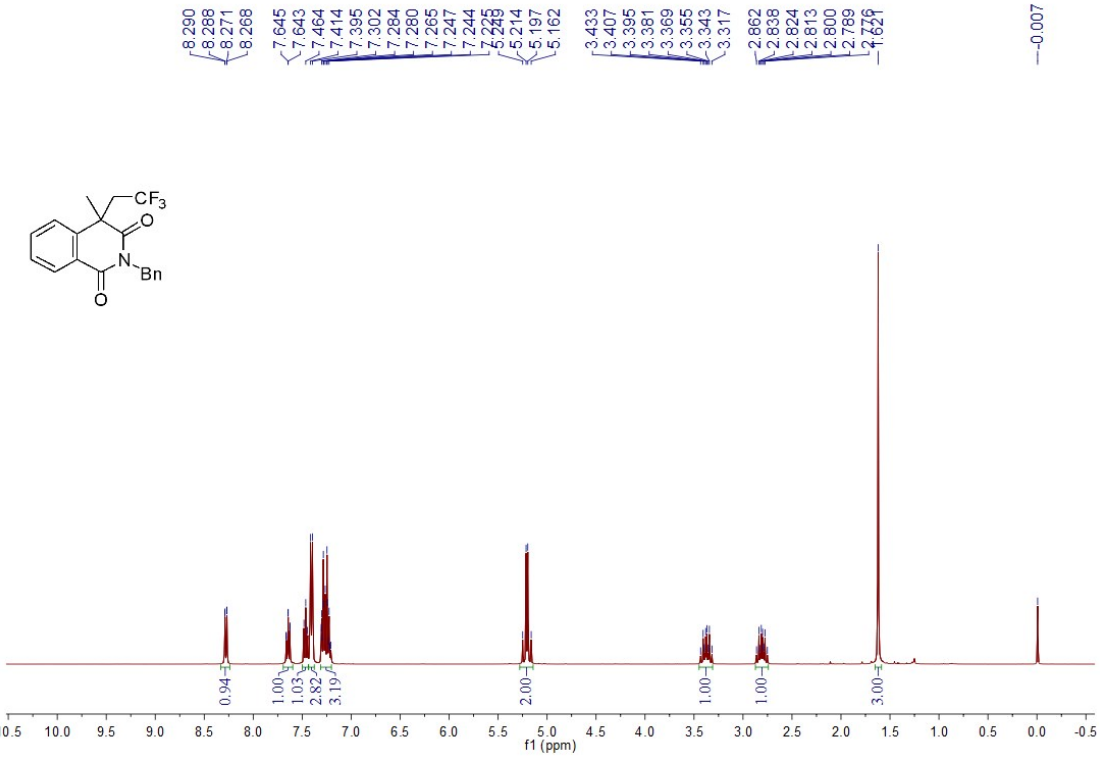
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5o**



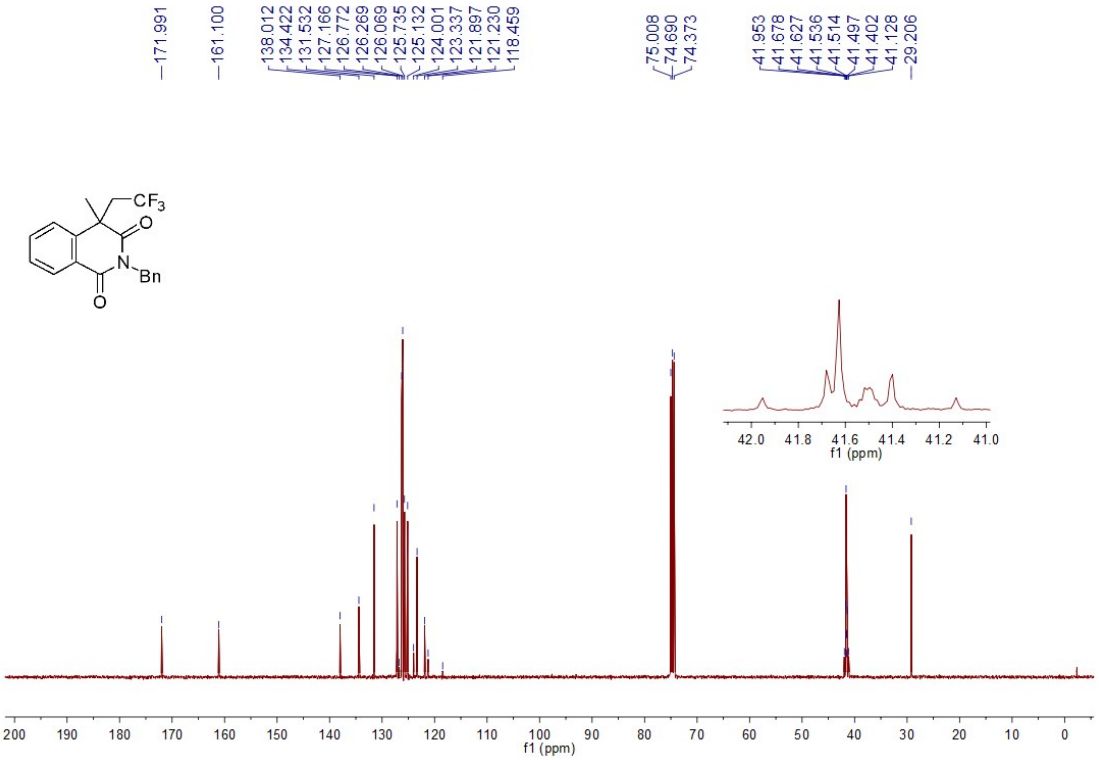
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5o**



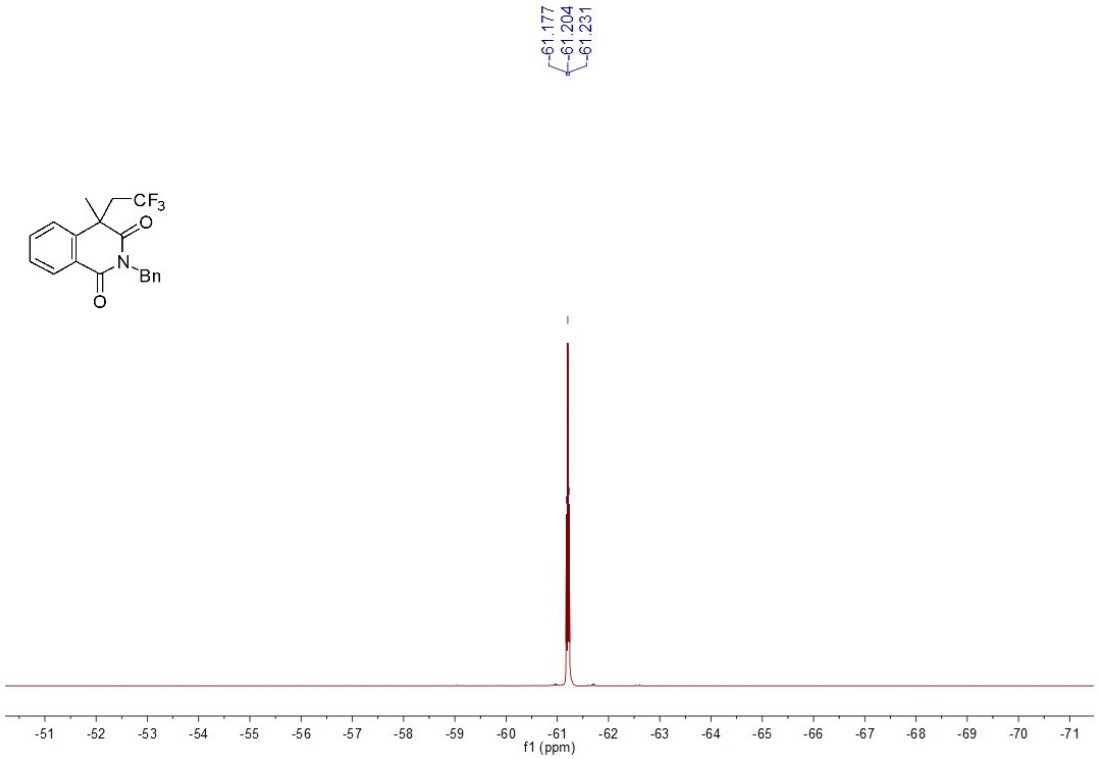
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5p**



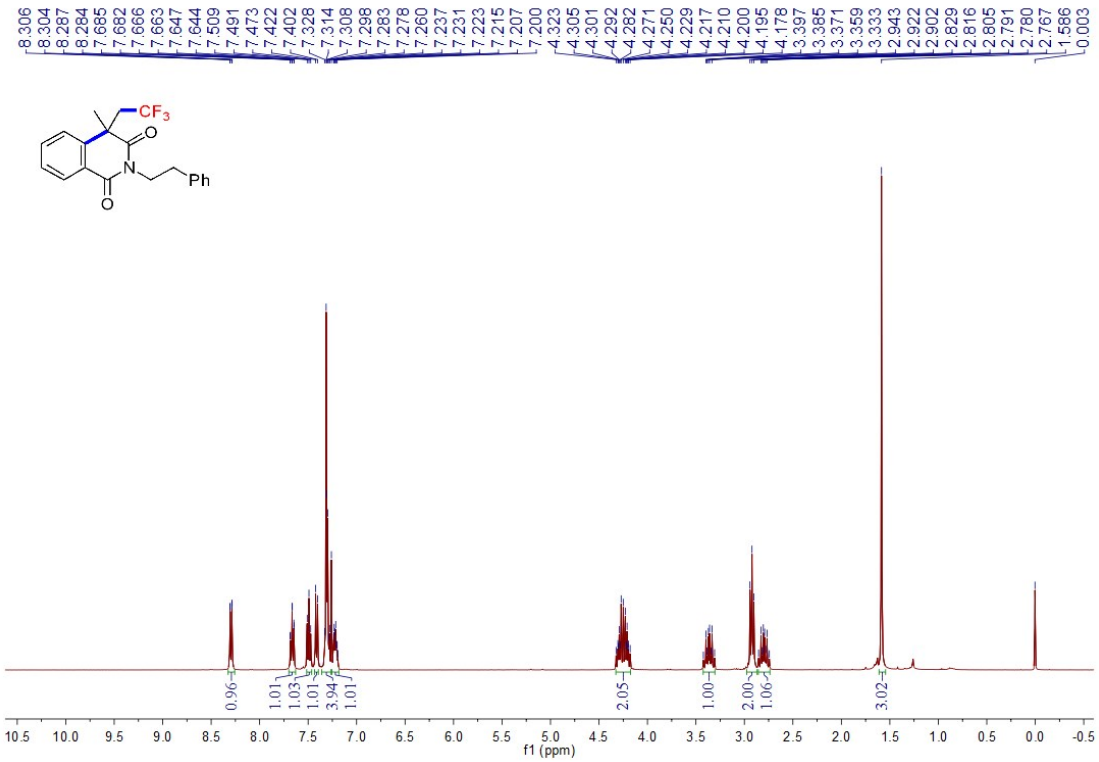
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5p**



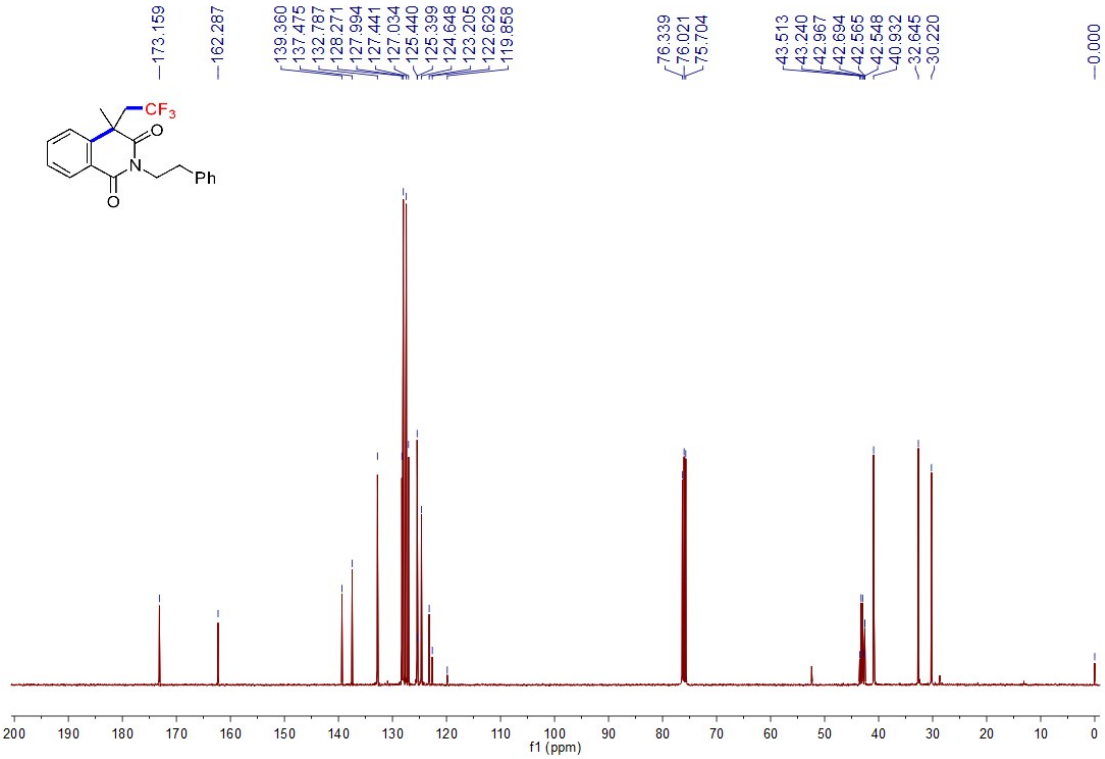
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5p**



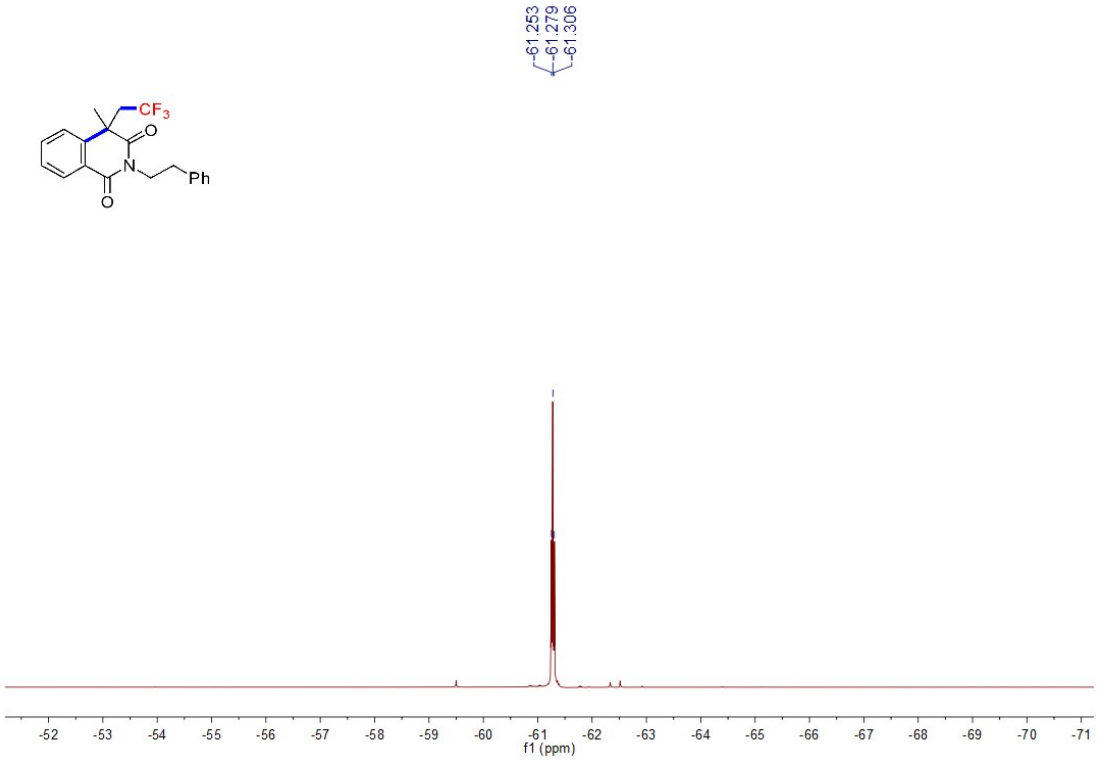
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5q**



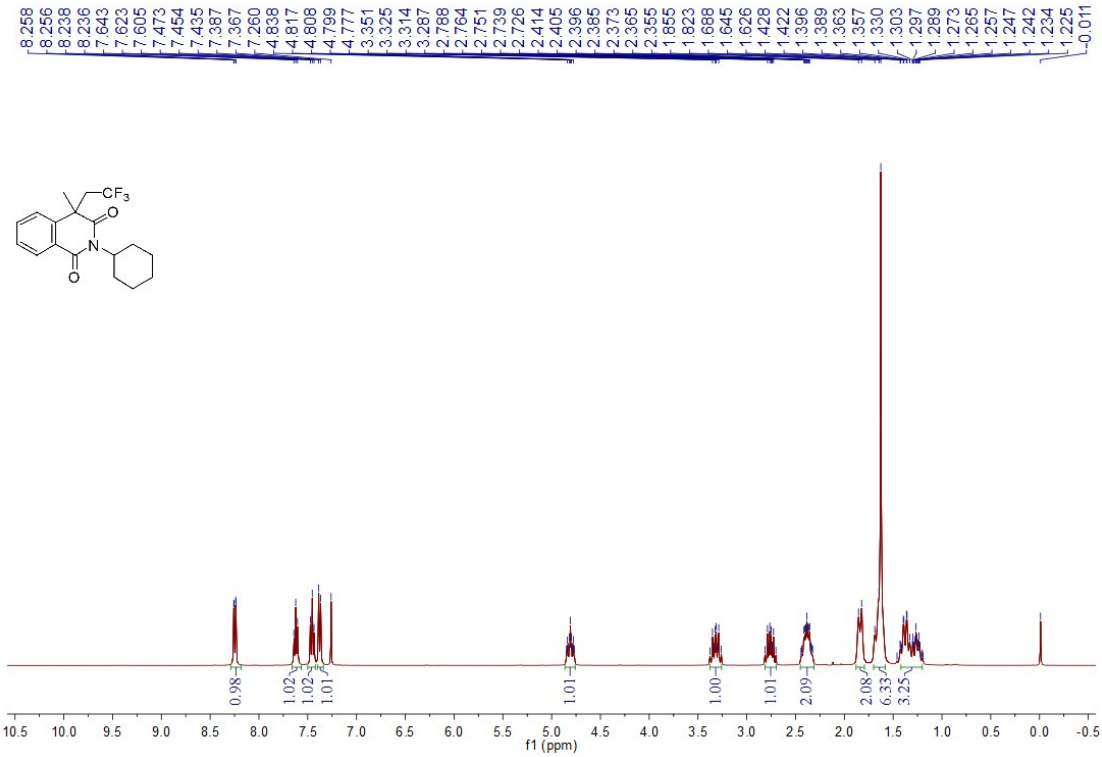
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5q**



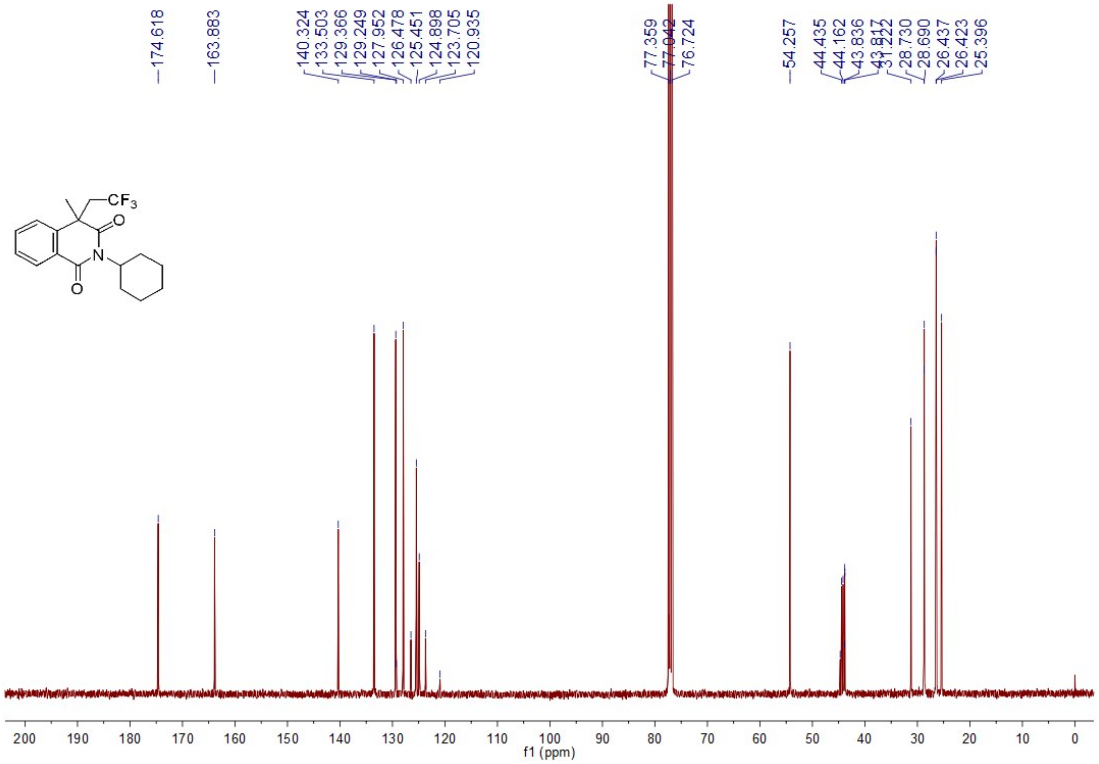
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5q**



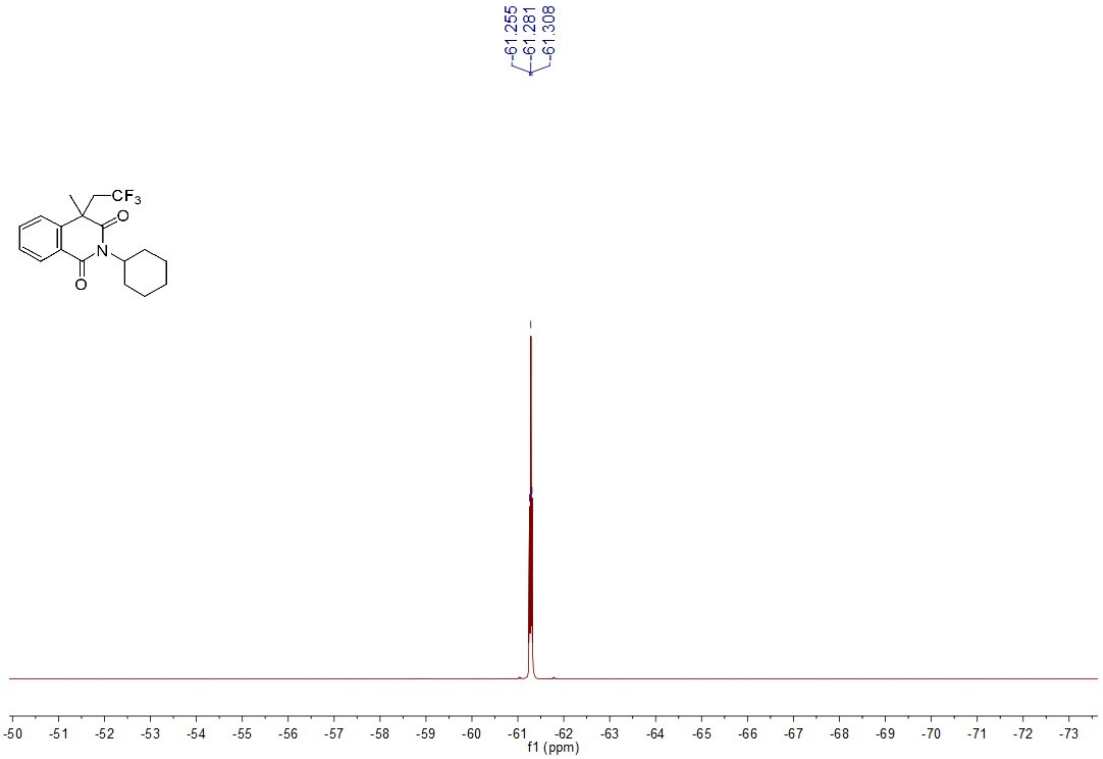
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5r**



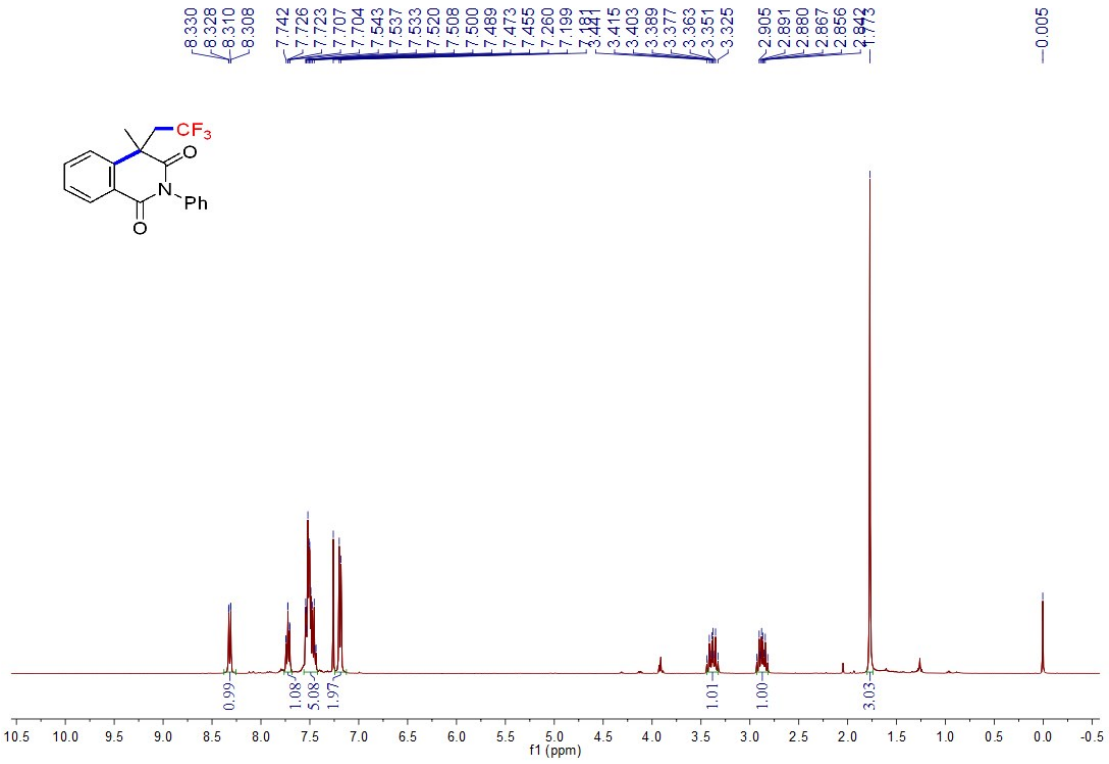
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5r**



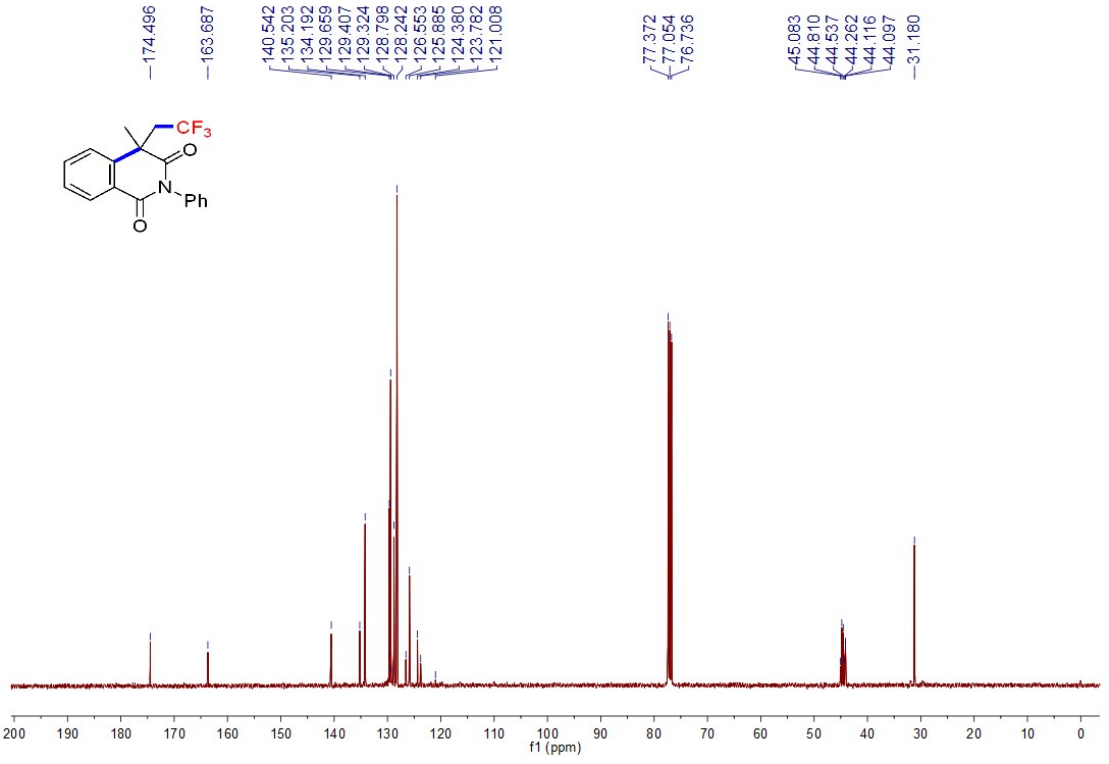
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5r**



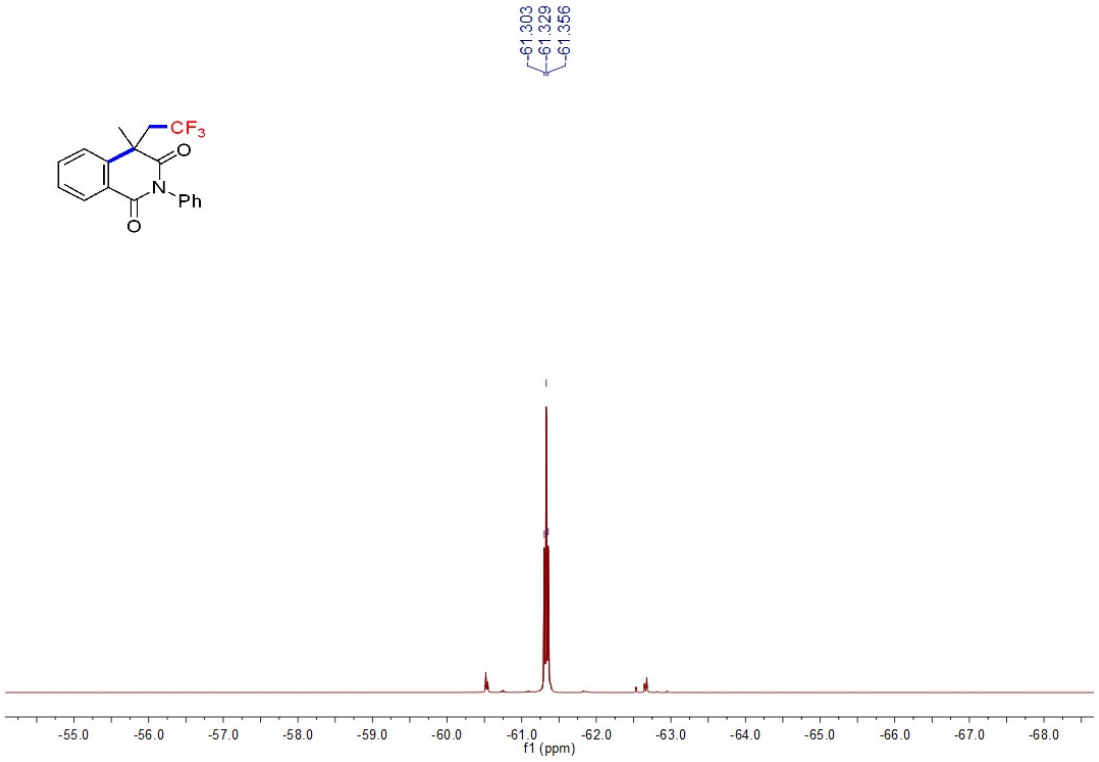
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5s**



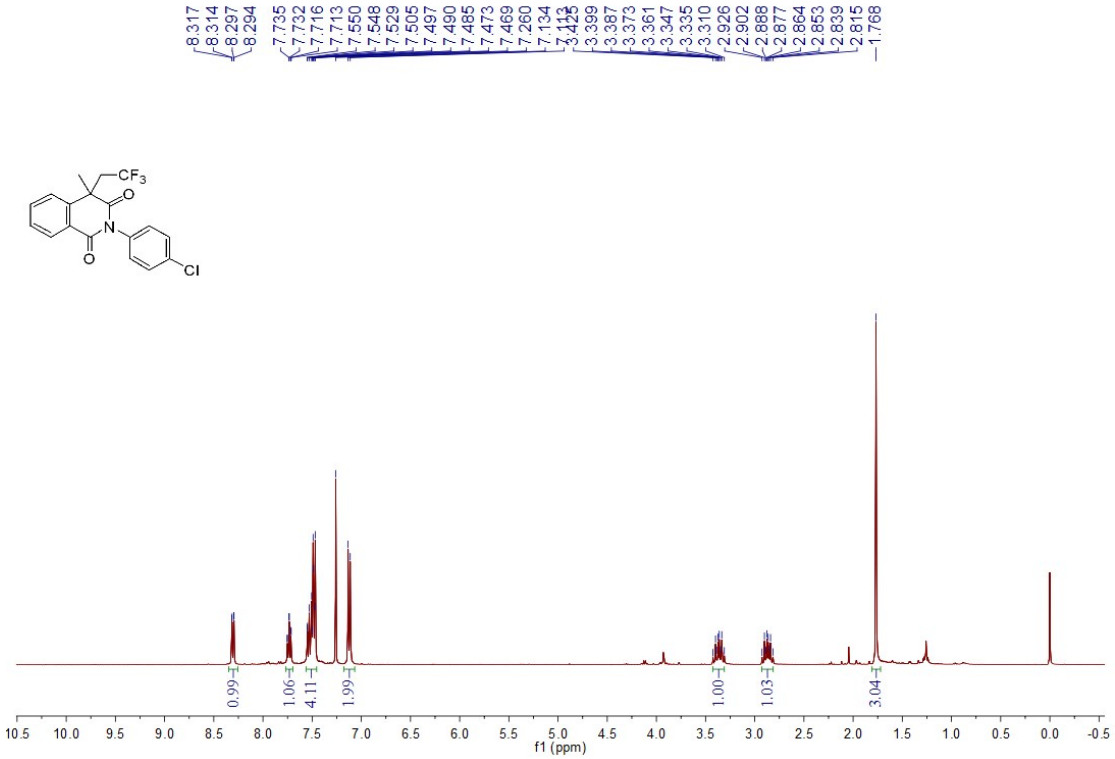
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5s**



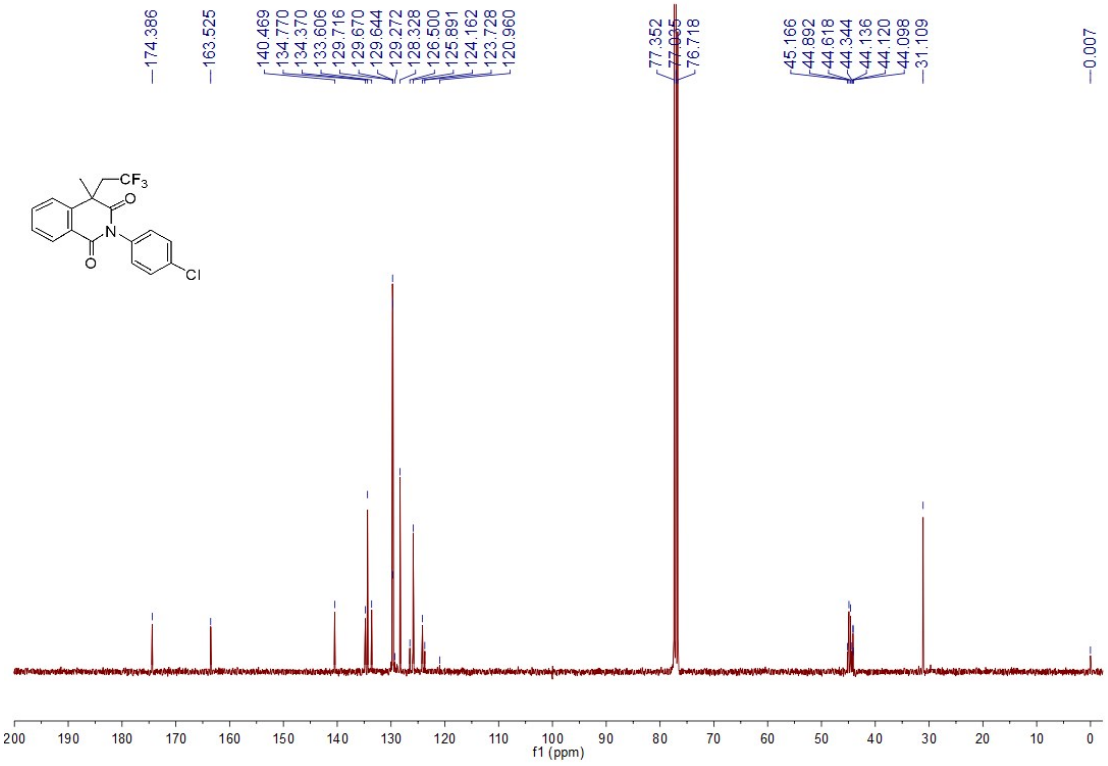
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5s**



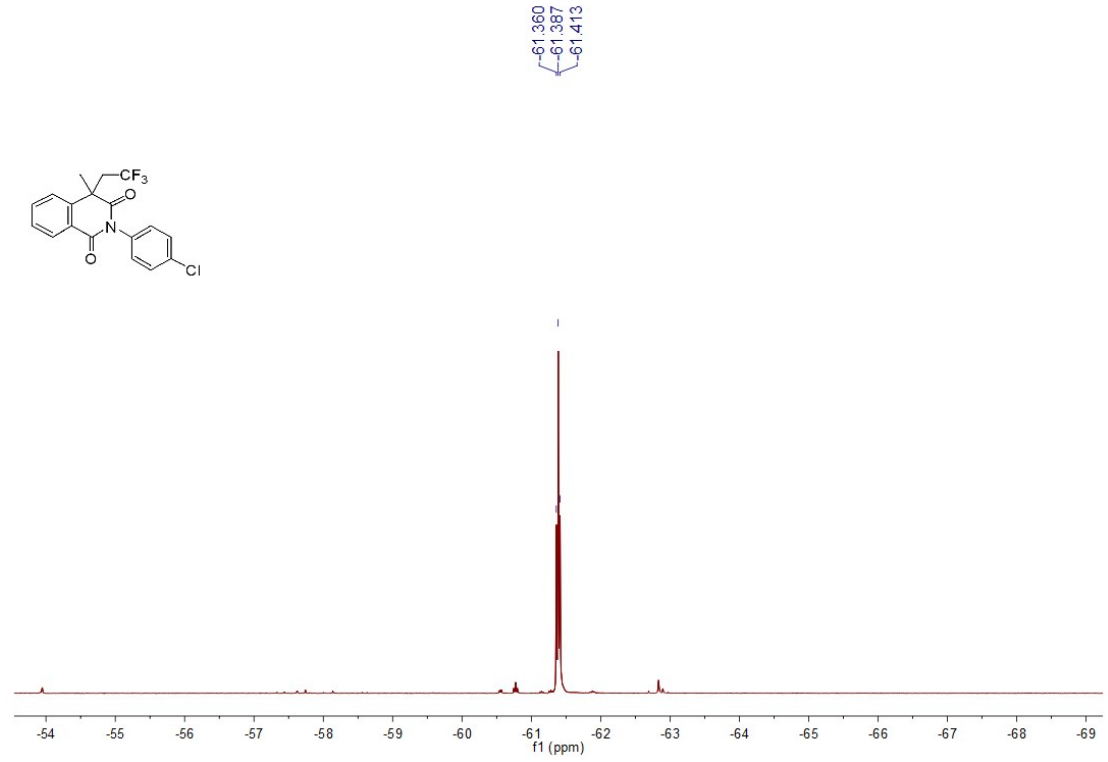
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5t**



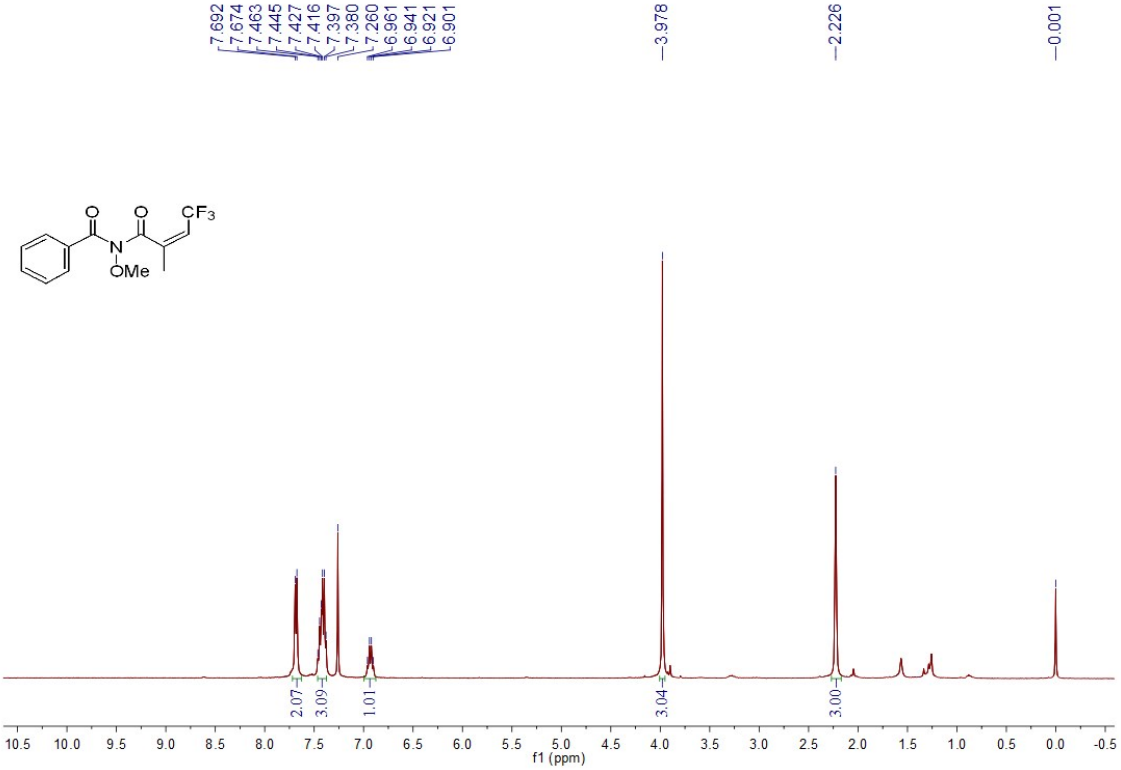
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5t**



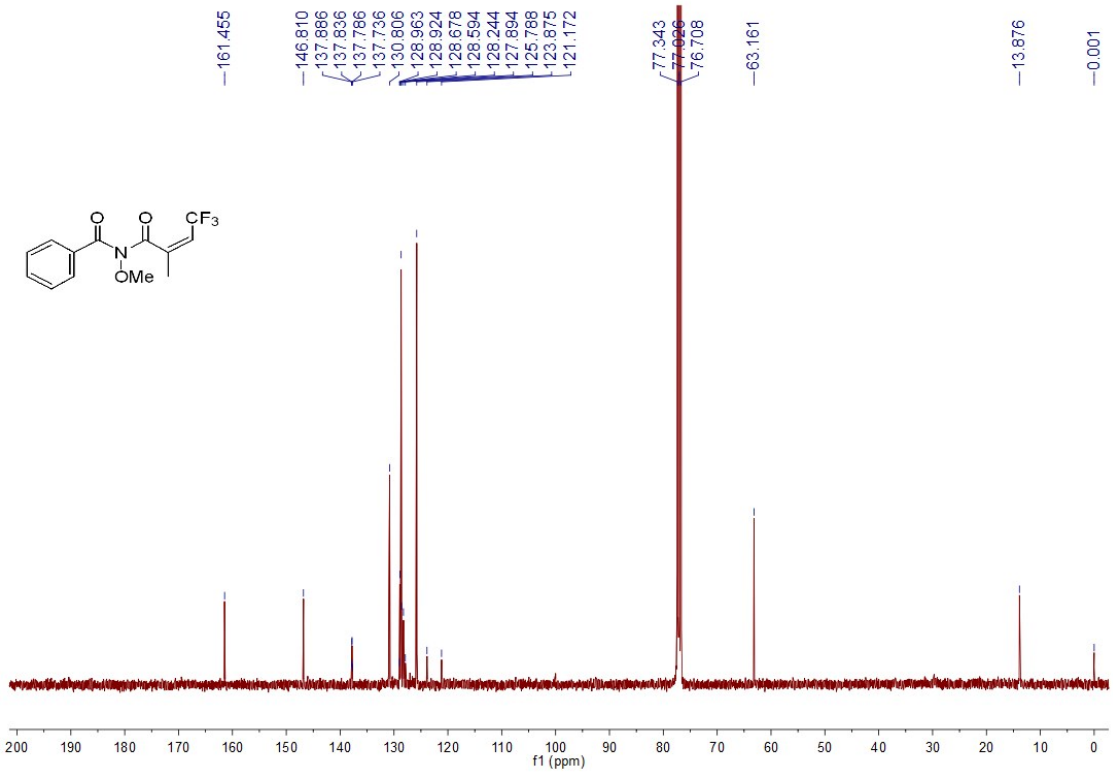
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5t**



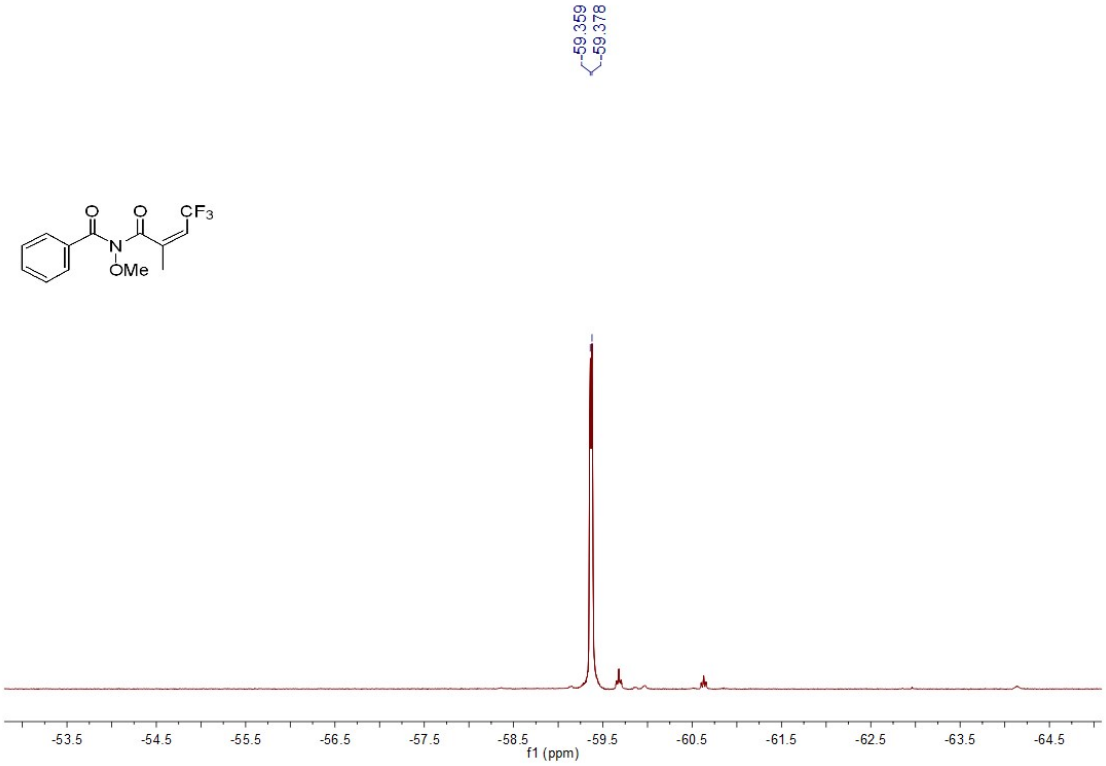
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5ua**



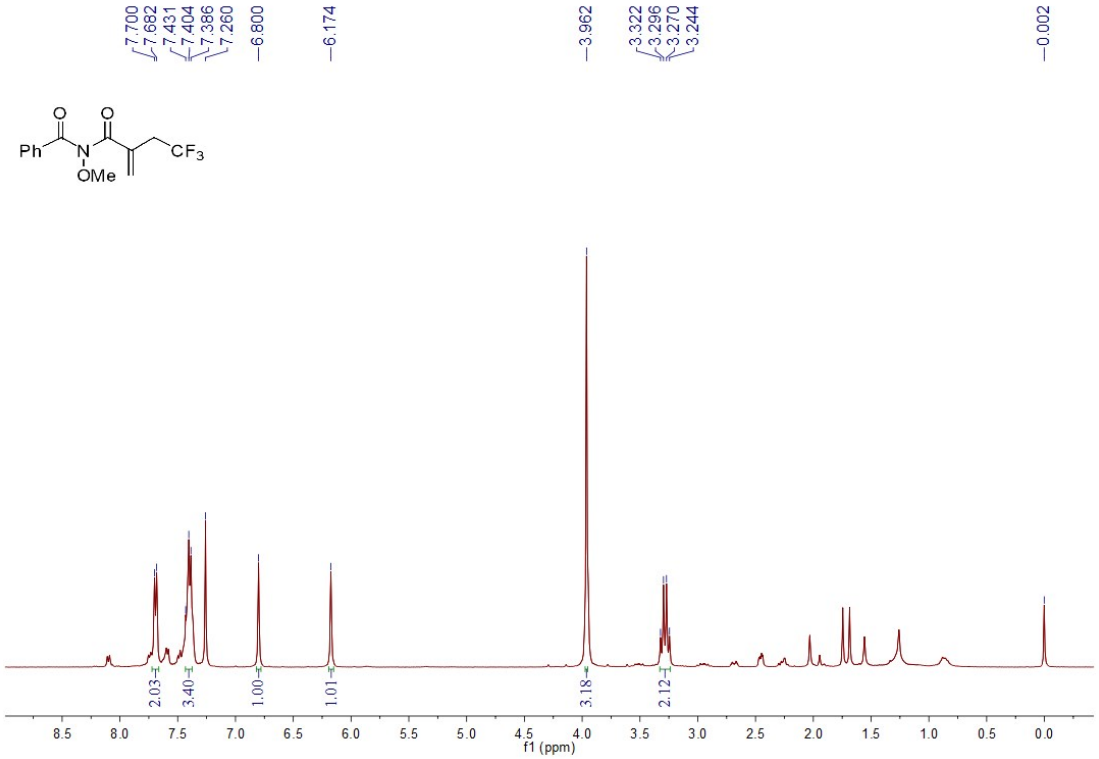
^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **5ua**



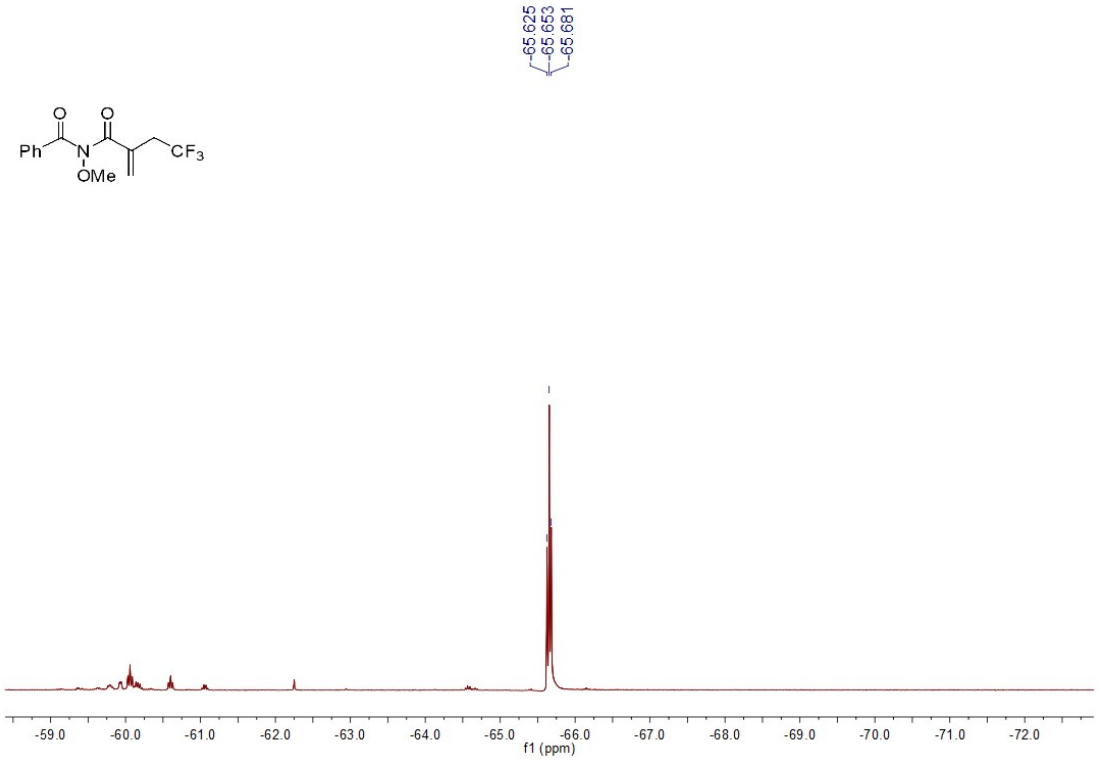
^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5ua**



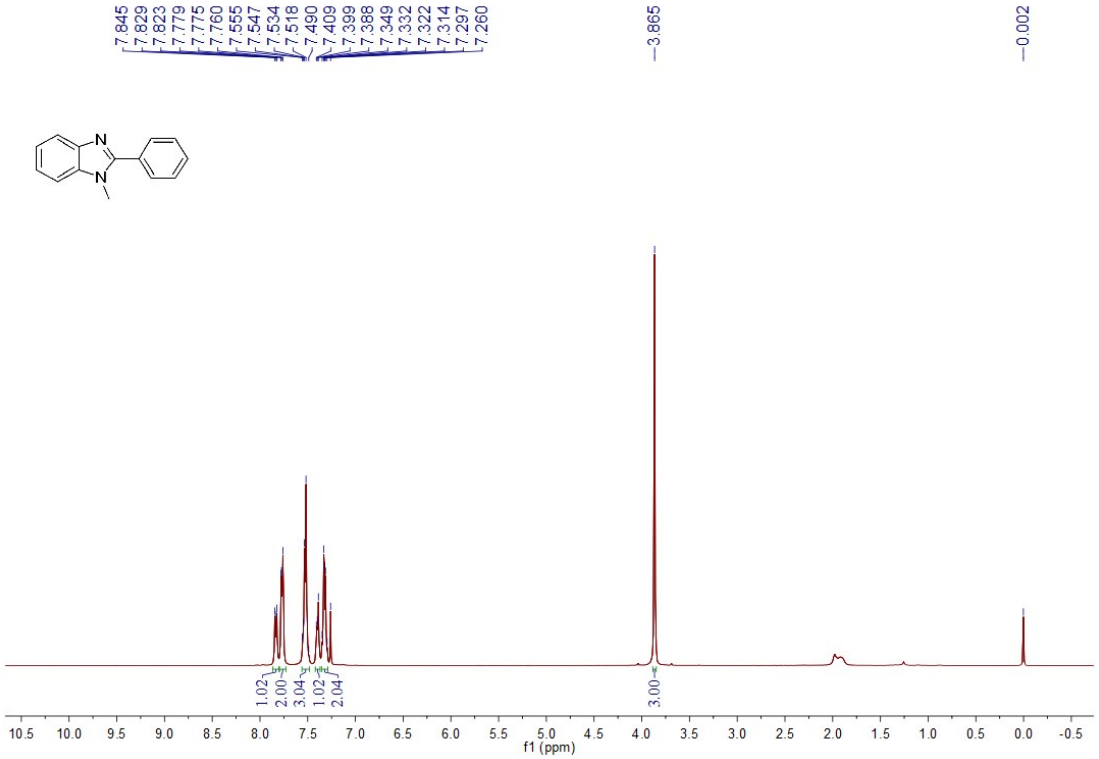
^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **5ub**



^{19}F NMR spectrum (376 MHz, $CDCl_3$) of compound **5ub**



^{1}H NMR spectrum (400 MHz, $CDCl_3$) of compound **1-methyl-2-phenyl-1*H*-benzo[*d*]imidazole**



^{13}C NMR spectrum (100 MHz, $CDCl_3$) of compound **1-methyl-2-phenyl-1*H*-benzo[*d*]imidazole**