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Electronic Supplementary Information

Rhodium(III)-Catalyzed [5+1] Annulation of 2-Alkenylphenols with Maleimides: Access to highly functionalize Spirocyclic Skeletons

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Note – References are provided in the footnote, wherever applicable.

General Information

All chemicals were purchased from commercial suppliers and used as delivered unless otherwise specified. Reactions were carried out using distilled solvents. NMR spectra were recorded on a BRUKER-AV400 spectrometer in $CDCl_3$ and DMSO-d_6 (400 MHz, ^1H and 100 MHz, ^{13}C). Tetramethylsilane (TMS; δ = 0 ppm) or residual non-deuterated $CDCl_3$ signal (δ = 7.27 ppm); and residual non-deuterated DMSO signal (δ = 2.5 ppm) served as internal standards for 1H NMR. The corresponding residual non-deuterated solvent signals ($CDCl_3$: δ = 77.16 ppm; DMSO: δ = 39.50 ppm) were used as internal standards for ^{13}C NMR. Chemical shifts (δ) are reported in parts per million downfield from the internal reference and coupling constants in Hertz (Hz). IR spectra were measured using a Perkin-Elmer FT-IR Spectrometer. Mass spectra were measured with Micromass Q-TOF (ESI-HRMS). The melting points of the products were determined using a Buchi melting point apparatus. Flash column chromatography was carried out using commercially obtained silica gel, and thin-layer chromatography was carried out using Merck silica gel 60 F_{254} TLC plates. Visualization was accomplished with short wave UV light or $KMnO_4$ staining solutions followed by heating. Flash column chromatography was performed using silica gel (230-400 mesh) with solvents distilled prior to use.

Experimental Section

Starting Materials

No attempts were made to optimize yields for substrate preparation. All spectral data obtained was according to the previously reported.

Maleimide derivatives [**2a**, **2b**, **2e**, **2f**, **2j**, **2l**, **2m**] and Maleic anhydride [**2n**], *p*-Benzoquinone [**2o**], 1,4-Naphoquinone [**2p**], Dimethyl maleate [**2q**] was purchased from commercial suppliers and used as delivered. Maleimide derivatives [**2c**, **2d**, **2g**, **2h**, **2i**, **2k**] are known molecules and was prepared according to reported literature procedure.[1]

2-Alkenylphenol derivatives [**1a-1i**, **1l**, **1j**, **1m**] was prepared according to reported literature procedure.[2] 2-(1-Phenylvinyl)phenol derivatives **[1n-1p]**,[3] 2-(Hept-1-en-2-yl)phenol derivatives [**1q-1s**],[4] substrates [**1t**],[5] [**1w**],[6] [**1y**],[7] [**1z**],[8] [**1z'**],[9] [**1k, 6a**],[10] [**4a, 5a**],[11] are known molecules and was prepared according to reported literature procedure. 2-Allylphenol [**1u**], 2-Phenylphenol [**1v**], 1-Naphthol [**1x**] was purchased from commercial suppliers and used without further manipulation.

Catalysts $[Cp^*Co(CO)I_2]$,[12a] $[Cp^*RhCl_2]_2$,[12b] and $[Cp^*IrCl_2]_2$[12c] was prepared according to reported literature procedure.

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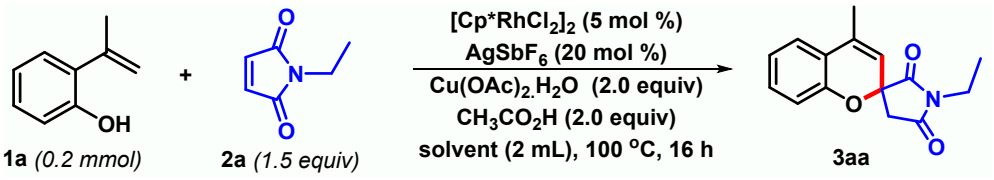
Detail Optimization Studies

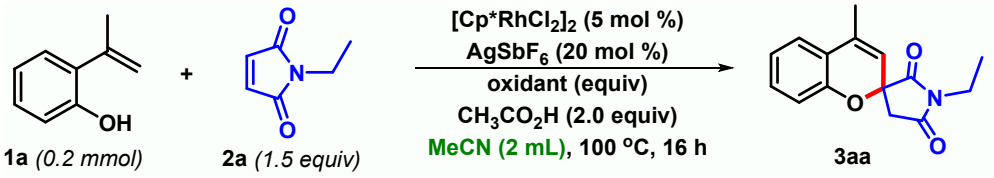
Optimization table SI 1, experimentations have been performed as shown in general experimental procedure (a). In all the cases, the crude products were submitted directly for ^{1}H-NMR analysis for calculating the yields in which 1,3,5-trimethoxybenzene (11.2 mg, 0.0667 mmol) has been used as an internal standard. nd = not detected.

1. Solvent Screening



entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	DCE	100	16	nd
2	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	TFE	100	16	nd
3	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	MeCN	100	16	53
4	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	DMF	100	16	37
5	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	THF	100	16	trace
6	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	toluene	100	16	nd
7	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	dioxane	100	16	trace
8	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	ethyl acetate	100	16	nd
9	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	EtOH	100	16	nd
10	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	MeOH	100	16	33
11	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	^{t}AmOH	100	16	trace
12	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	H_2O	100	16	24
13	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	DMSO	100	16	28
14	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2.H_2O$	HFIP	100	16	trace

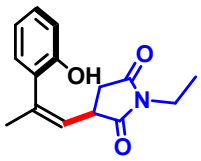
2. Oxidant Screening



1a *(0.2 mmol)* + **2a** *(1.5 equiv)* → **3aa**

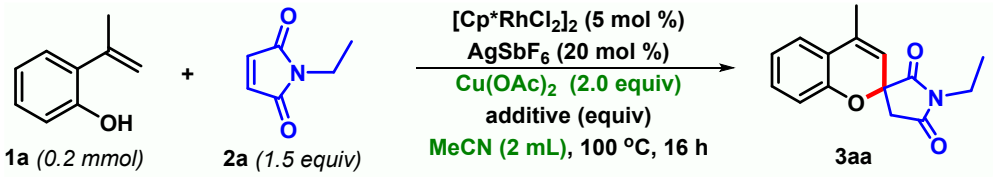
entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1[a]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	AgOAc	MeCN	100	16	14
2	$[Cp^*RhCl_2]_2$	CH_3CO_2H	Ag_2O	MeCN	100	16	trace
3	$[Cp^*RhCl_2]_2$	CH_3CO_2H	Ag_2CO_3	MeCN	100	16	nd
4	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Zn(OAc)_2$	MeCN	100	16	nd
5	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	64
6	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$CuBr_2$	MeCN	100	16	nd
7	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$CuCl_2$	MeCN	100	16	nd
8	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$CuCO_3$	MeCN	100	16	61
9	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$CuSO_4.5H_2O$	MeCN	100	16	nd
10	$[Cp^*RhCl_2]_2$	CH_3CO_2H	CuO	MeCN	100	16	trace
11[b]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2 + O_2$	MeCN	100	16	48
12[c]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	61
13	$[Cp^*RhCl_2]_2$	CH_3CO_2H	none	MeCN	100	16	nd

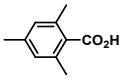
[a] 19% byproduct **3aa'** formed along with **3aa**. [b] 1.0 equiv of $Cu(OAc)_2$ was used with oxygen balloon. [c] 1.5 equiv of $Cu(OAc)_2$ was used.



3aa', 19%
(byproduct)

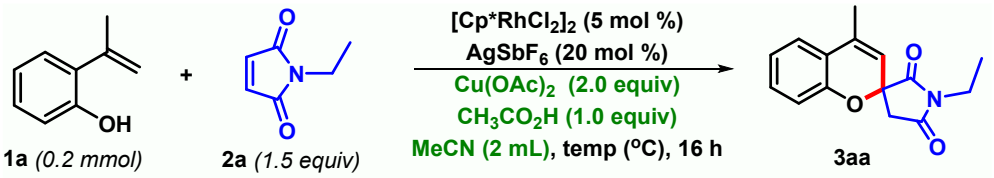
3. Additive Screening



entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1	$[Cp^*RhCl_2]_2$	none	$Cu(OAc)_2$	MeCN	100	16	51
2	$[Cp^*RhCl_2]_2$	LiOAc	$Cu(OAc)_2$	MeCN	100	16	46
3	$[Cp^*RhCl_2]_2$	NaOAc	$Cu(OAc)_2$	MeCN	100	16	49
4	$[Cp^*RhCl_2]_2$	CF_3CO_2Na	$Cu(OAc)_2$	MeCN	100	16	34
5	$[Cp^*RhCl_2]_2$	Na_2CO_3	$Cu(OAc)_2$	MeCN	100	16	40
6	$[Cp^*RhCl_2]_2$	$NaHCO_3$	$Cu(OAc)_2$	MeCN	100	16	42
7	$[Cp^*RhCl_2]_2$	NH_4Cl	$Cu(OAc)_2$	MeCN	100	16	nd
8	$[Cp^*RhCl_2]_2$	$AdCO_2H$	$Cu(OAc)_2$	MeCN	100	16	55
9	$[Cp^*RhCl_2]_2$	PivOH	$Cu(OAc)_2$	MeCN	100	16	42
10	$[Cp^*RhCl_2]_2$	CF_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	nd
11	$[Cp^*RhCl_2]_2$	$ClCH_3CO_2H$	$Cu(OAc)_2$	MeCN	100	16	39
12	$[Cp^*RhCl_2]_2$		$Cu(OAc)_2$	MeCN	100	16	63
13[a]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	55
14[b]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	69
15[c]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	64

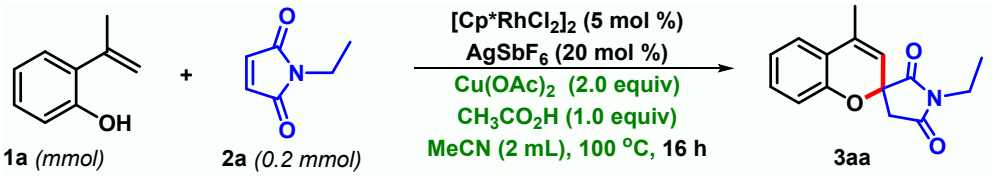
[a] 3.0 equiv CH_3CO_2H was used. [b] CH_3CO_2H (1.0 equiv). [c] CH_3CO_2H (0.5 equiv).

4. Temperature Screening



entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	120	16	57
2	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	80	16	65
3	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	60	16	14

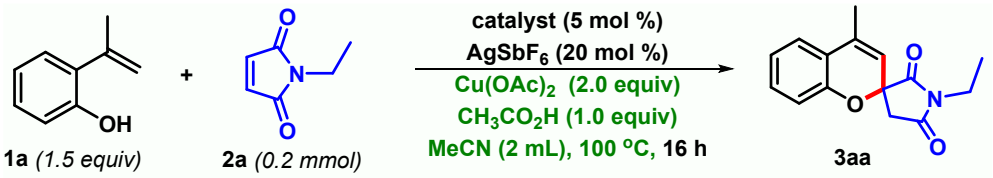
5. Substrate Equivalence Screening



entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1[a]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	84
2[b]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	79

[a] Substrates mole **1a** (0.3 mmol), **2a** (0.2 mmol) were used. [b] **1a** (0.24 mmol), **2a** (0.2 mmol).

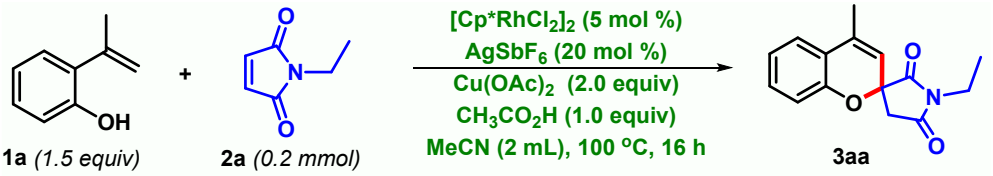
6. Catalyst Screening



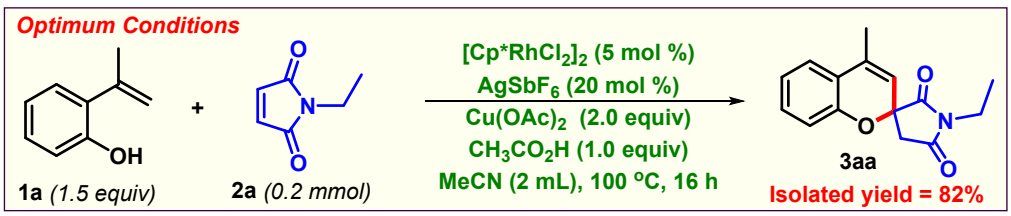
entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1	$[Cp^*Co(CO)I_2]$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	nd
2	$[Cp^*IrCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	nd
3	Ru(p-cymene)$Cl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	nd
4	none	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	nd
5[a]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	52
6[b]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	81
7	$[Cp^*Rh(CH_3CN)](SbF_6)_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	48
8[c]	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	16	64

[a] Absence of $AgSbF_6$ activator. [b] 2.5 mol % of catalyst loading. [c] 1.0 mol % of catalyst loading.

7. Time Studies



entry	catalyst (5 mol %)	additive (1.0 equiv)	oxidant (2.0 equiv)	solvent (2 mL)	temp. (°C)	time (h)	yield (%)
1	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	8	73
2	$[Cp^*RhCl_2]_2$	CH_3CO_2H	$Cu(OAc)_2$	MeCN	100	24	84



General Procedure

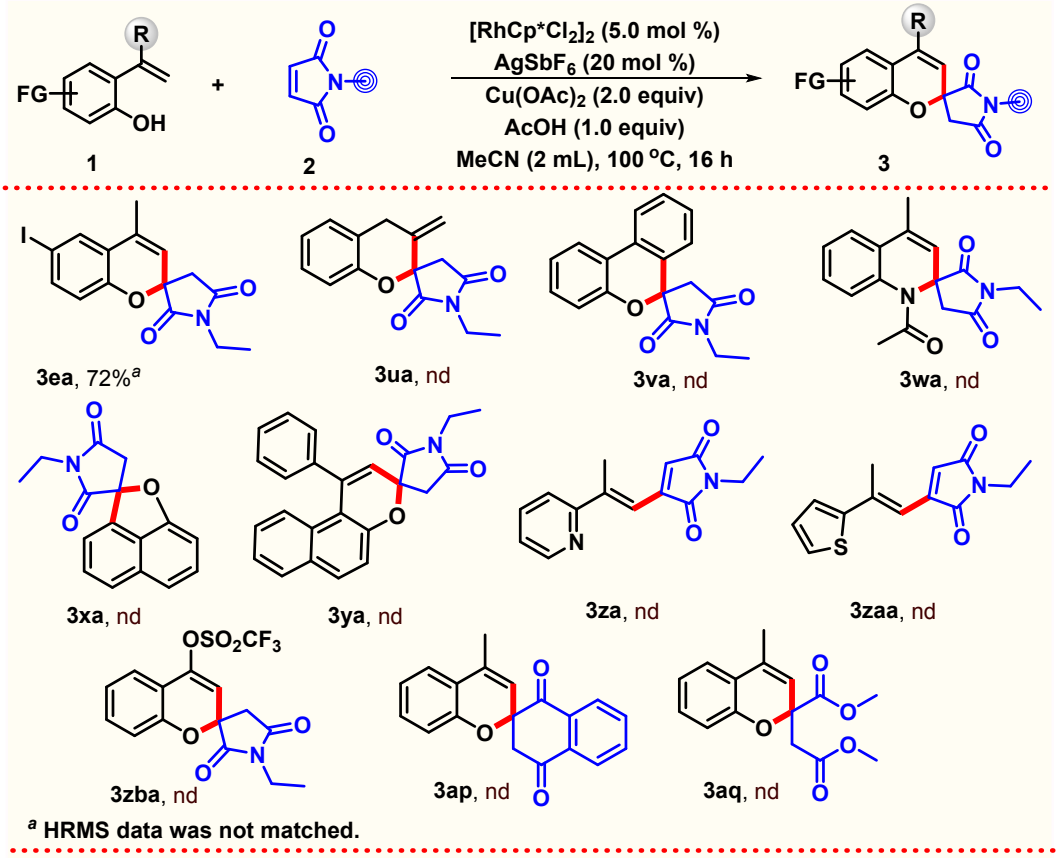
A. Experimental procedure for the spirocyclic skeletons (Scheme - 2 and 3).

To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides (0.20 mmol) and 2-Isopropenylphenol derivatives (0.3 mmol, 1.5 equiv), catalyst $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), activator $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), additive AcOH (12.0 mg, 0.2 mmol, 1.0 equiv), oxidant anhydrous $Cu(OAc)_2$ (73.0 mg, 0.4 mmol, 2.0 equiv), and acetonitrile solvent (2 mL). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred at the same temperature for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, filtered through a silica (230-400 mess size) pad using a mixture of EtOAc and petroleum ether (1:1, 100 mL), and concentrated under vacuo. In the optimization Table, the crude products were submitted directly for ^{1}H-NMR analysis for calculating the yields wherein 1,3,5-trimethoxybenzene (11.2 mg, 0.0667 mmol) has been used as an internal standard. For the substrate scope (Scheme - 2 and 3), the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product.

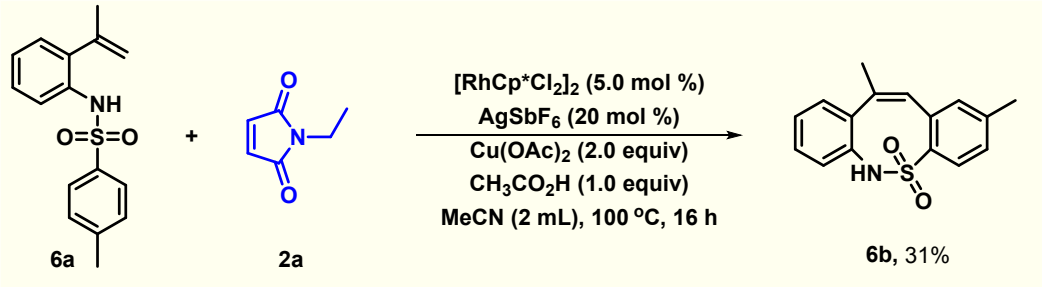
B. Experimental procedure for the scale-up reaction (Scheme - 4a).

To an oven-dried 50-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with *N*-Ethylmaleimide (1.0 g, 8.0 mmol) and 2-Isopropenylphenol (1.61 g, 12.0 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (122.5 mg, 2.5 mol%), $AgSbF_6$ (268.2 mg, 20 mol%), AcOH (480.0 mg, 8.0 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (1.45 g, 8.0 mmol, 2.0 equiv), and acetonitrile solvent (40 mL) were taken.. The vial was sealed with a screw cap and placed in a pre-heated oil bath at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature and concentrated under vacuo. The crude product was purified on a silica gel (230-400 mess size) using flash column chromatography using EtOAc/petroleum ether as eluent (3:97 to 6:94 *v/v*) to afford the desired products **3aa**, 1.6 g in 77% yield.

Scope for 2-Alkenylphenol and maleimide derivatives



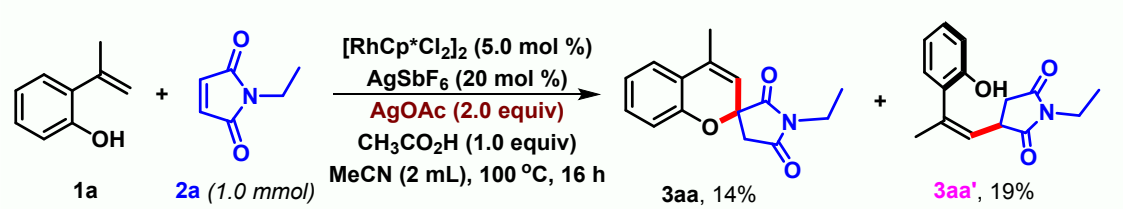
Scheme SI-1. Scope for 2-alkenylphenol and maleimide derivatives.



During the substrate scope for 2-Alkenylphenol derivative, the reaction of 2-Isopropenyl-N-tosyl-aniline **6a** with *N*-Ethylmaleimide **2a**, the desired spirocyclic product was not observed. Instead, an unusual self-coupling product **6b** was observed in 31% of the yield. While optimization, the result was published by Zhou and Yi group.[13]

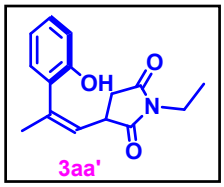
[13] L. Li, H. Gao, M. Sun, Z. Zhou, W. Yi, *Org. Lett.,* 2020, **22**, 5473.

Hydroarylated Product Detection



To an oven-dried 16-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (125.2 mg, 1.0 mmol) and 2-Isopropenylphenol **1a** (1.5 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (31.0 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (68.5 mg, 20 mol%, 0.2 equiv), AcOH (60.0 mg, 1.0 mmol, 1.0 equiv), oxidant AgOAc (333.8 mg, 2.0 mmol, 2.0 equiv), and acetonitrile solvent (10 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the mixture was cooled to room temperature and concentrated under vacuo. The crude product was purified on a silica gel (230-400 mess size) using flash column chromatography using EtOAc/petroleum ether as eluent (3:97 to 12:88 *v/v*) to afford the spirocyclic product **3aa**, 36.2 mg, and hydroarylated product **3aa′**, 49.4 mg in 14 and 19% yield, respectively.

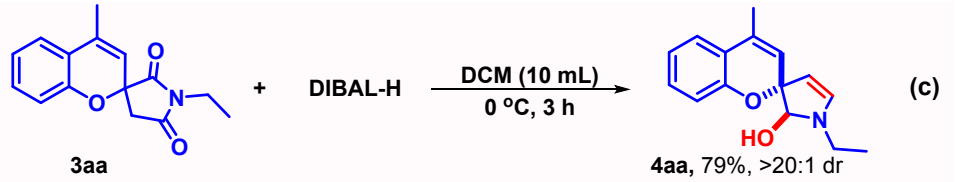
(Z)-1-Ethyl-3-(2-(2-hydroxyphenyl)prop-1-en-1-yl)pyrrolidine-2,5-dione (3aa').



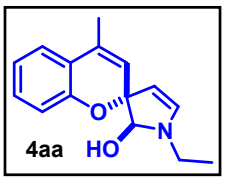
Appearance – Yellow Oil; R_f = 0.20 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.24 – 7.15 (m, 1H), 6.99 (d, *J* = 8.3 Hz, 2H), 6.93 (dd, *J* = 10.9, 3.8 Hz, 1H), 5.49 (d, *J* = 9.8 Hz, 1H), 3.55 (q, *J* = 7.2 Hz, 2H), 3.36 (td, *J* = 9.2, 5.2 Hz, 1H), 2.81 (dd, *J* = 18.4, 9.2 Hz, 1H), 2.48 (dd, *J* = 18.4, 5.2 Hz, 1H), 2.06 (s, 3H), 1.15 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 179.9, 175.9, 152.5, 140.1, 129.2, 128.6, 127.5, 124.3, 120.8, 117.7, 41.4, 35.1, 34.3, 25.7, 12.9; **FT-IR (cm^{-1})** 3438, 2978, 2939, 2850, 1771, 1693, 1604, 1447, 1406, 1348, 1224, 1128; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{15}H_{17}NO_3Na$ 282.1106; found 282.1108.

Derivatization of Spirocyclic Product

I. Reduction with DIBAL-H

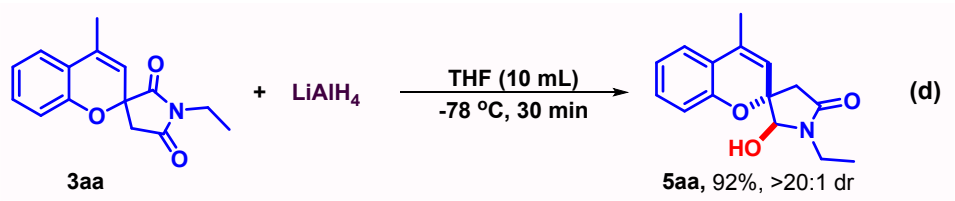


1'-Ethyl-4-methyl-1',2'-dihydrospiro[chromene-2,3'-pyrrol]-2'-ol (4aa).



Prepared using the following procedure. To an oven dried 10 ml two neck round bottle flask, equipped with a magnetic stir bar was charged with spirocyclic product **3aa** (102.6 g, 0.4 mmol) and dry DCM (10 ml, 0.4 M) under argon atmosphere. The solution was placed in ice-water and stirred for fifteen minutes under argon condition. Later, DIBAL-H (1M solution in toluene) was added (2 ml, 2.0 mmol) drop wise to the reaction mixture and stirred for 3 h at 0 °C under argon atmosphere. On completion of reaction (monitored by TLC), reaction mixture was quenched with saturated solution of NH_4Cl (5 ml). The reaction mixture was extracted with diethyl ether (10 ml x 2 times), dried over anhydrous sodium sulfate. The reaction mixture was concentrated under reduced pressure and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 6:94 *v/v*) to obtain the desired product **4aa**, 76.7 mg in 79% yield. **Appearance** – Brown Oil; $\boldsymbol{R_f}$ = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.22 (ddd, *J* = 8.1, 7.4, 1.6 Hz, 1H), 7.16 (dd, *J* = 7.4, 1.6 Hz, 1H), 6.99 – 6.92 (m, 2H), 6.55 (s, 1H), 6.42 (t, *J* = 2.5 Hz, 1H), 6.31 (s, 1H), 5.63 – 5.62 (m, 1H), 5.41 (br. s, 1H), 3.76 (q, *J* = 7.3 Hz, 2H), 2.12 (d, *J* = 1.3 Hz, 3H), 1.30 (t, *J* = 7.3 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 151.7, 129.3, 128.5, 128.5, 126.2, 123.4, 120.8, 120.8, 120.5, 119.9, 115.3, 107.6, 44.1, 26.4, 16.3; **^{13}C{^{1}H}/DEPT-135 NMR (100 MHz, $CDCl_3$)** δ 128.5, 128.5, 123.4, 120.8, 120.8, 120.5, 119.9, 115.3, 107.6, 44.1, 26.4, 16.3; **FT-IR (cm^{-1})** 3509, 3032, 2977, 2928, 2876, 2848, 1644, 1576, 1487, 1453, 1405, 1343, 1282, 1208, 1170, 1031; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{15}H_{17}NO_2Na$ 266.1157; found 266.1154.

II. Reduction with $LiAlH_4$

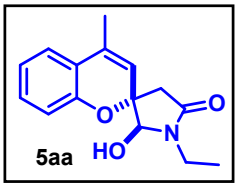


3aa **5aa,** 92%, >20:1 dr

Attempts were made to reduce both carbonyl groups:-

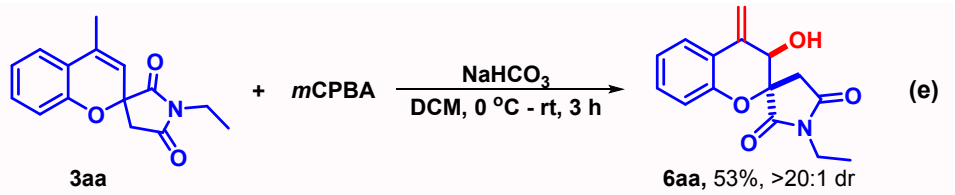
entry	initial temp (°C)	final temp (°C)	time (min)	starting material 3aa remaining	product yield (%)
1	0	0	30	decomposed	-
2	-78	0	60	decomposed	-
3	-78	-78	30	completely consumed	92
4	-78	-78	720	completely consumed	90

1'-Ethyl-2'-hydroxy-4-methylspiro[chromene-2,3'-pyrrolidin]-5'-one (5aa).

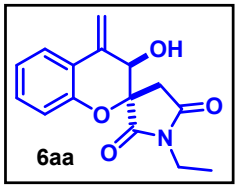


Prepared using the following procedure. To an oven dried 25 ml two neck round bottle flask, equipped with a magnetic stir bar was charged with spirocyclic product **3aa** (102.6 g, 0.4 mmol) and dry THF (10 ml, 0.4 M) under argon atmosphere. The solution was placed at suitable temperature as shown above in optimization table and stirred for fifteen minutes under argon condition. Later, $LiAlH_4$ (45.6 mg, 1.2 mmol, 3.0 equiv) was added in one batch to the reaction mixture and stirred for given time at various temperature under argon atmosphere. The reaction mixture was monitored by TLC at various interval of time. On completion of reaction, the reaction mixture was quenched with saturated solution of NH_4Cl (5 ml). The reaction mixture was extracted with ethyl acetate (10 ml x 2 times), dried over anhydrous sodium sulfate. The reaction mixture was concentrated under reduced pressure and the crude product was purified on a silica gel (100-200 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (20:80 to 60:40 *v/v*) to obtain the desired product **5aa**, 95.4 mg in 92% yield. **Appearance** –White Solid; **mp** = 131 - 133 °C; R_f = 0.30 (50% EtOAc /Hexane); **^{1}H NMR (400 MHz, DMSO-d_6)** δ 7.24 (dd, J = 7.6, 1.3 Hz, 1H), 7.16 (td, J = 8.3, 1.3 Hz, 1H), 6.95 (td, J = 7.6, 0.8 Hz, 1H), 6.78 (d, J = 8.0 Hz, 1H), 6.36 (d, J = 7.3 Hz, 1H), 5.70 (d, J = 1.1 Hz, 1H), 4.92 (d, J = 7.3 Hz, 1H), 3.48 – 3.37 (m, 1H), 3.14 – 3.05 (m, 1H), 2.62 (d, J = 17.1 Hz, 1H), 2.04 (d, J = 1.1 Hz, 1H), 1.06 (t, J = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6)** δ 171.5, 151.7, 130.4, 129.8, 124.2, 123.3, 122.0, 121.5, 116.4, 86.4, 80.8, 42.3, 34.8, 18.2, 13.5; **^{13}C{^{1}H}/DEPT-135 NMR (100 MHz, DMSO-d_6)** δ 129.8, 124.2, 122.0, 121.5, 116.4, 86.4, 42.3, 34.8, 18.2, 13.5; **FT-IR (cm^{-1})** 3416, 2958, 2921, 1666, 1489, 1445, 1404, 1352, 1228, 1054, 1028, 999; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{17}NO_3H$ 260.1287; found 260.1283.

III. Hydroxylation with *m*CPBA

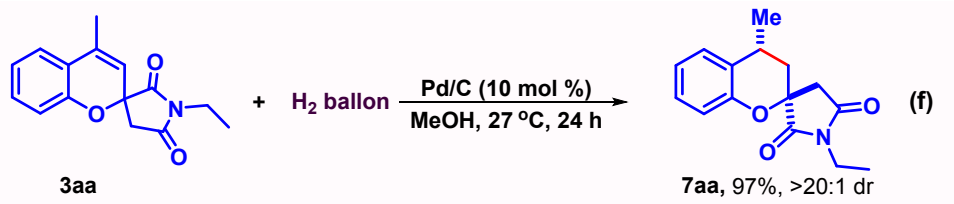


1'-Ethyl-3-hydroxy-4-methylenespiro[chromane-2,3'-pyrrolidine]-2',5'-dione (6aa).

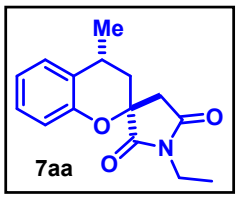


Prepared using the following procedure. To an oven dried 25 ml two neck round bottle flask, equipped with a magnetic stir bar was charged with spirocyclic product **3aa** (102.9 g, 0.4 mmol), $NaHCO_3$ (50.5 mg, 0.6 mmol, 1.5 equiv) and dry DCM (10 ml, 0.4 M) under argon atmosphere. The solution was placed at 0 °C and stirred for fifteen minutes under argon condition. Later, mCPBA (70%, 119 mg, 0.48 mmol, 3.0 equiv) was dissolved in 10 ml of dry DCM and was added in dropwise to the reaction mixture in 20 minutes. Next the reaction mixture was gradually warmed to the room temperature and stirred for 3 h. On completion of reaction (monitored by TLC), reaction mixture was quenched with saturated solution of $Na_2S_2O_4$ (10 ml). The reaction mixture was extracted with DCM (10 ml x 2 times), washed with $NaHCO_3$ solution and dried over anhydrous sodium sulfate. The reaction mixture was concentrated under reduced pressure and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 10:90 *v/v*) to obtain the desired product **6aa**, 58.1 mg in 53% yield. **Appearance** – White Solid; **mp** = 144 - 146 °C; R_f = 0.35 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.53 (d, *J* = 8.2 Hz, 1H), 7.21 (t, *J* = 7.4 Hz, 1H), 7.00 (t, *J* = 7.4 Hz, 1H), 6.87 (d, *J* = 8.2 Hz, 1H), 5.76 (d, *J* = 1.9 Hz, 1H), 5.52 (d, *J* = 1.9 Hz, 1H), 4.96 (s, 1H), 3.63 (q, *J* = 7.2 Hz, 2H), 3.26 (br. s, 1H), 2.86 (d, *J* = 18.2 Hz, 1H), 2.52 (d, *J* = 18.2 Hz, 1H), 1.22 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 175.1, 174.2, 150.8, 138.0, 130.1, 125.0, 122.3, 120.0, 117.7, 108.1, 80.9, 67.4, 36.1, 34.3, 12.9; **FT-IR (cm^{-1})** 3471, 2923, 2852, 1786, 1707, 1569, 1483, 1404, 1346, 1303, 1255, 1225, 1136, 1041, 890; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{15}NO_4H$ 274.1079; found 274.1078.

IV. Hydrogenation with H_2, Pd/C

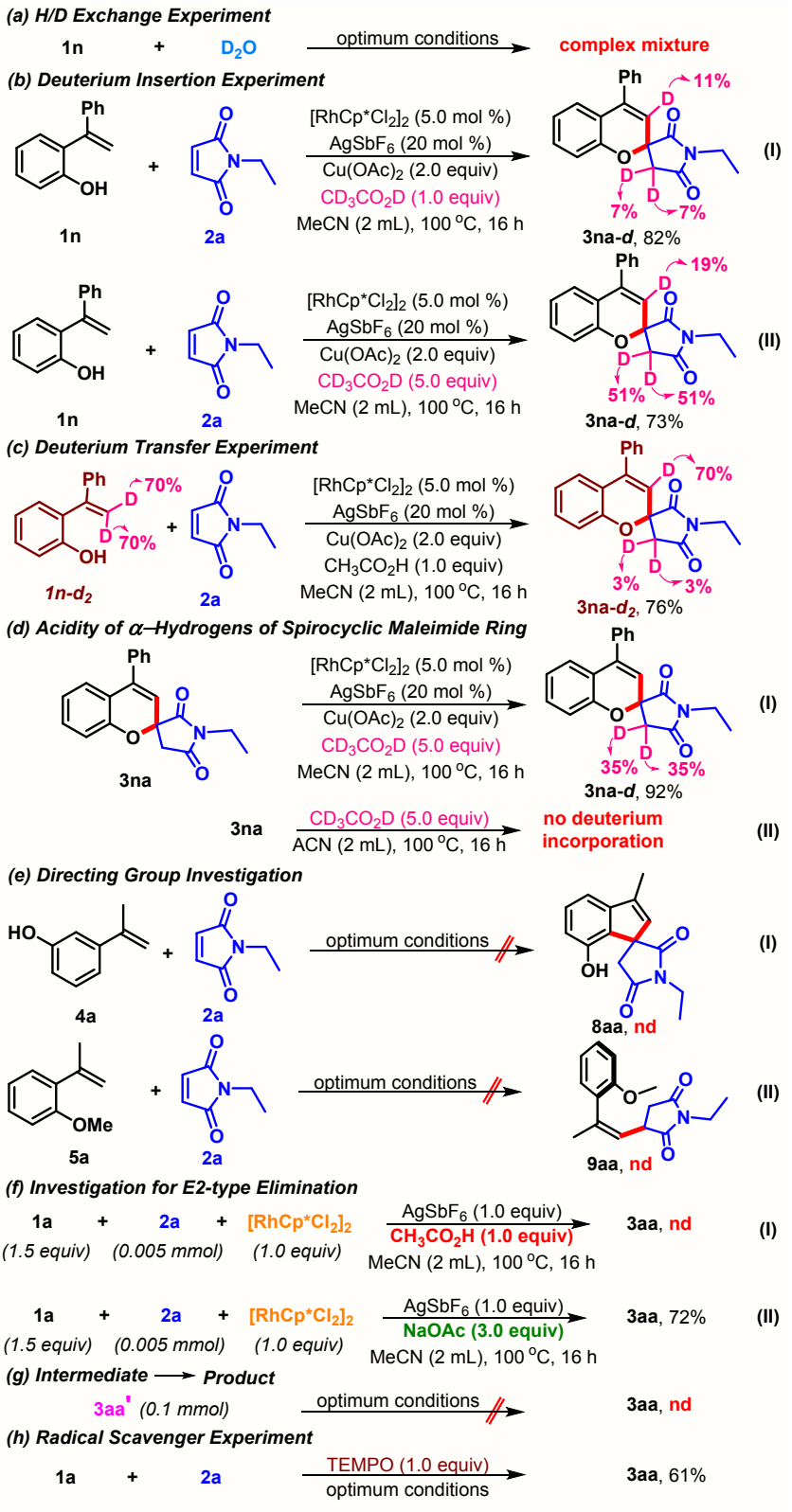


1'-Ethyl-4-methylspiro[chromane-2,3'-pyrrolidine]-2',5'-dione (7aa).

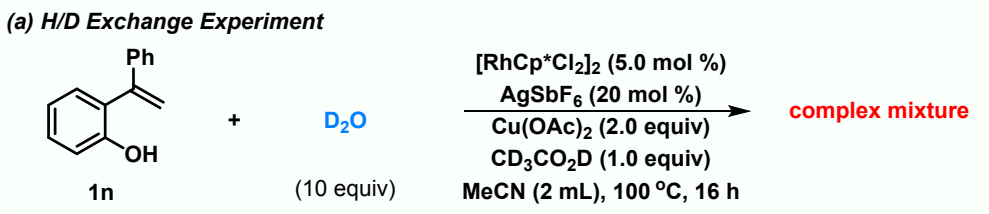


Prepared using the following procedure. To an oven dried 25 ml single neck round bottle flask, equipped with a magnetic stir bar was charged with spirocyclic product **3aa** (102.9 g, 0.4 mmol) in MeOH (8 mL, 0.5 M) was added Pd/C (32 mg, 10 mol %) at the room temperature. The flask was evacuated and refilled with argon gas. Finally, the reaction vessel was connected to a hydrogen balloon. The reaction mixture was stirred at the room temperature for 24 h. After completion of the reaction (monitored by TLC), the reaction mixture was filtered through the bed of Celite. The filtrate was concentrated in vacuo, and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product **7aa** in 97% yield. **Appearance** – White Solid; **mp** = 119 - 121 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.25 (d, *J* = 8.2 Hz, 1H), 7.12 (t, *J* = 7.5 Hz, 1H), 6.95 (t, *J* = 7.5 Hz, 1H), 6.83 (d, *J* = 8.2 Hz, 1H), 3.64 (q, *J* = 7.2 Hz, 2H), 3.04 – 2.95 (m, 1H), 2.83 (d, *J* = 18.1 Hz, 1H), 2.77 (d, *J* = 18.1 Hz, 1H), 1.15 (t, *J* = 12.8 Hz, 1H), 1.90 (dd, *J* = 12.8, 5.6 Hz, 1H), 1.41 (d, *J* = 6.7 Hz, 3H), 1.24 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 175.8, 173.5, 151.8, 127.8, 127.0, 125.9, 121.7, 117.4, 77.6, 41.4, 36.2, 34.1, 25.5, 19.8, 13.0; **FT-IR (cm^{-1})** 2964, 2926, 2873, 1786, 1714, 1579, 1488, 1448, 1404, 1349, 1303, 1234, 1115, 1035, 883; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{17}NO_3H$ 260.1287; found 260.1284.

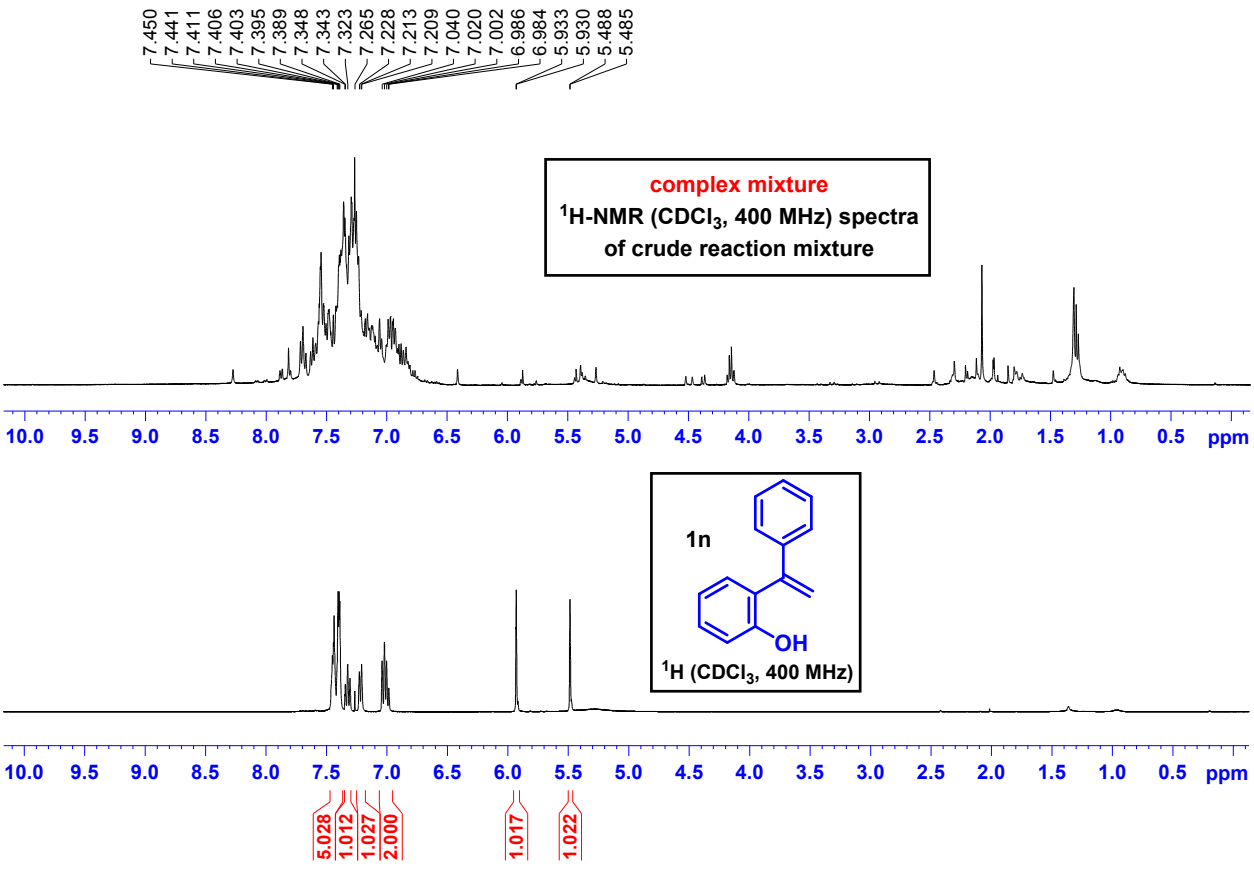
Mechanistic Studies

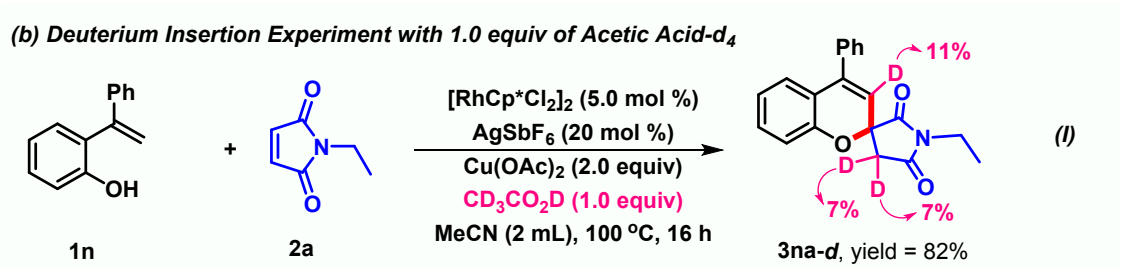


(a) H/D Exchange Experiment

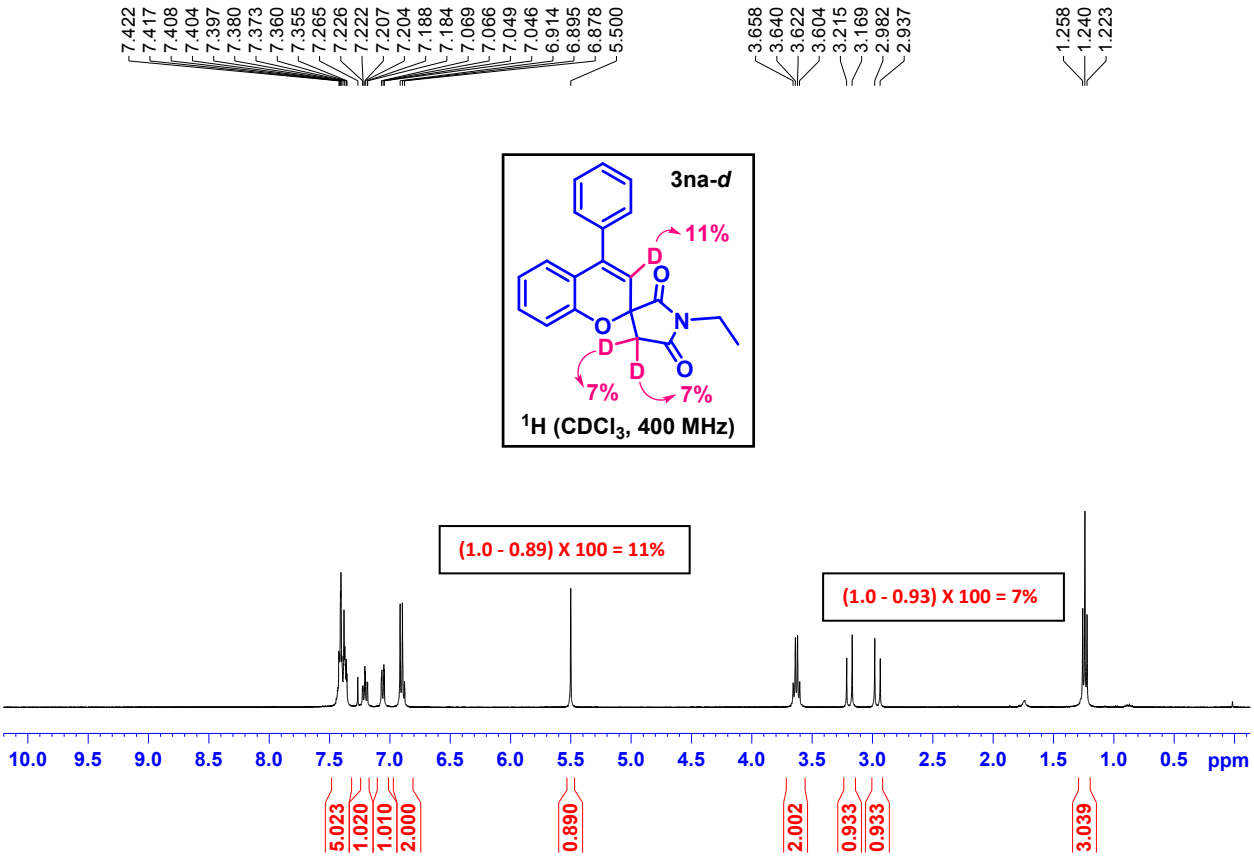


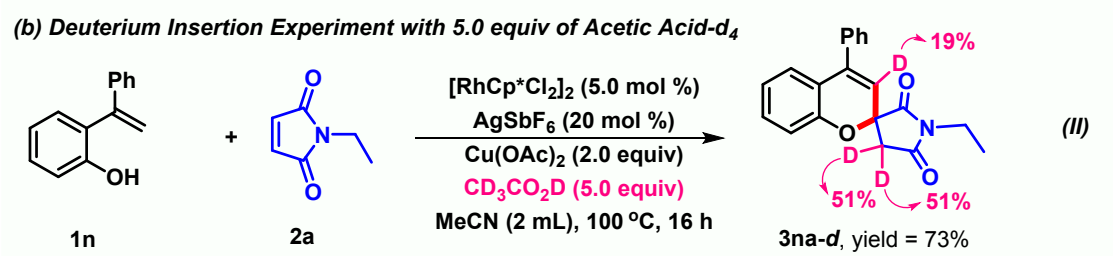
To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with 2-(1-phenylvinyl)phenol (39.3 mg, 0.20 mmol), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), CD_3CO_2D (13.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile solvent (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After the reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC. Substrate **1n** *was completely disappeared, and several fan spots were formed* on the TLC plate. Subsequently, the mixture was filtered through a silica (230-400 mess size) pad using a mixture of EtOAc and petroleum ether (1:1, 100 mL) and concentrated under vacuo. Next, the crude product was further analyzed by ^{1}H-NMR.



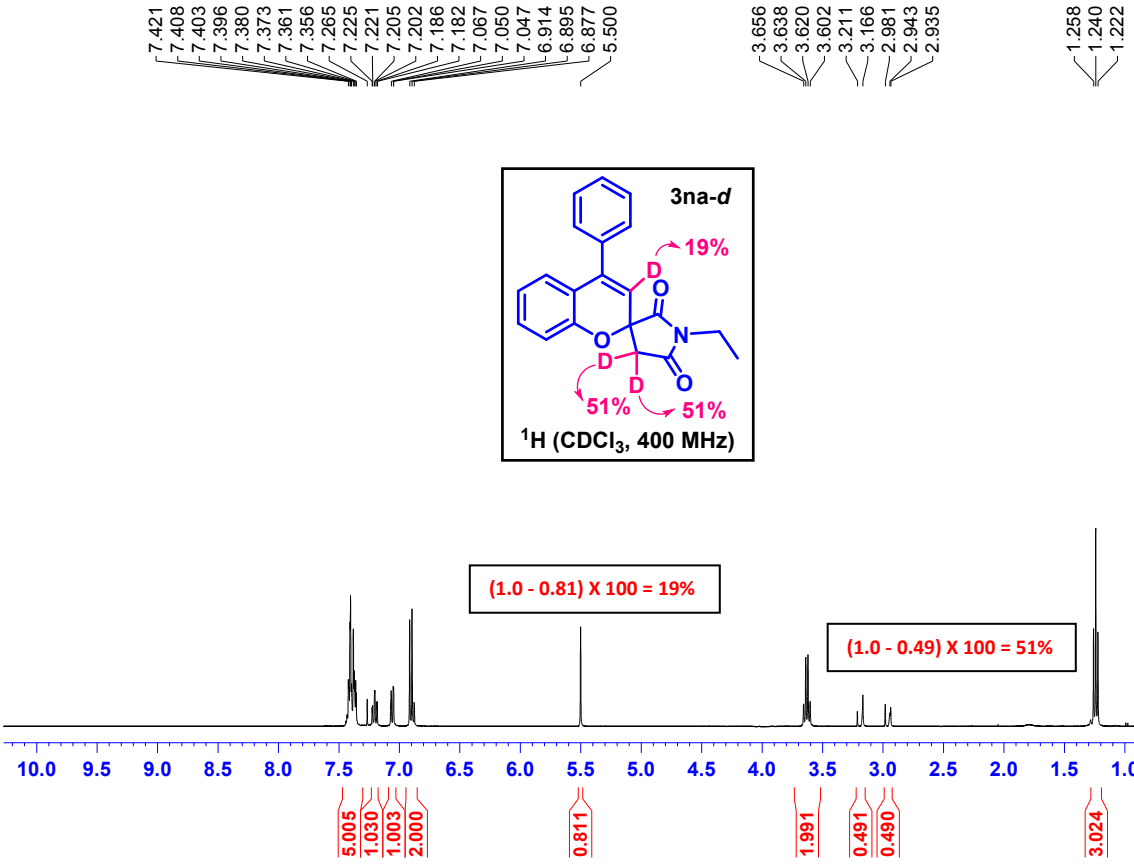


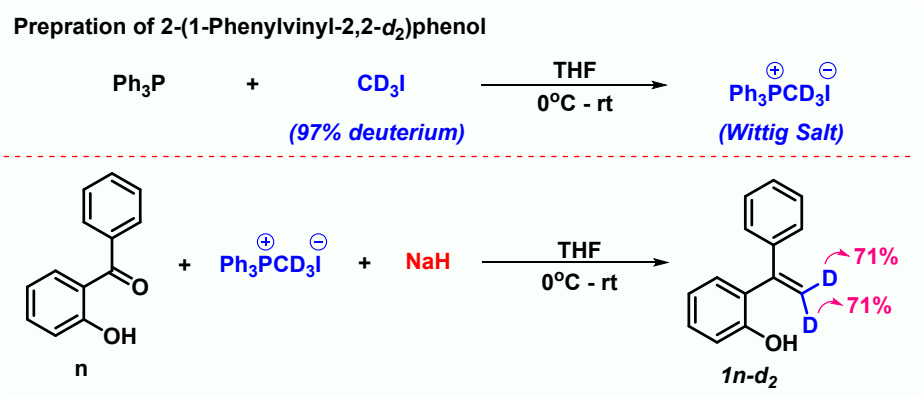
To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 2-(1-phenylvinyl)phenol **1n** (39.3 mg, 0.3 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), ***CD_3CO_2D (13.0 mg, 0.2 mmol, 1.0 equiv)***, anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product **3na-*d*** in 82% yield along with 7% deuterium insertion at alfa carbon to the carbonyl group of maleimide ring and 11% deuterium insertion at olefin carbon.



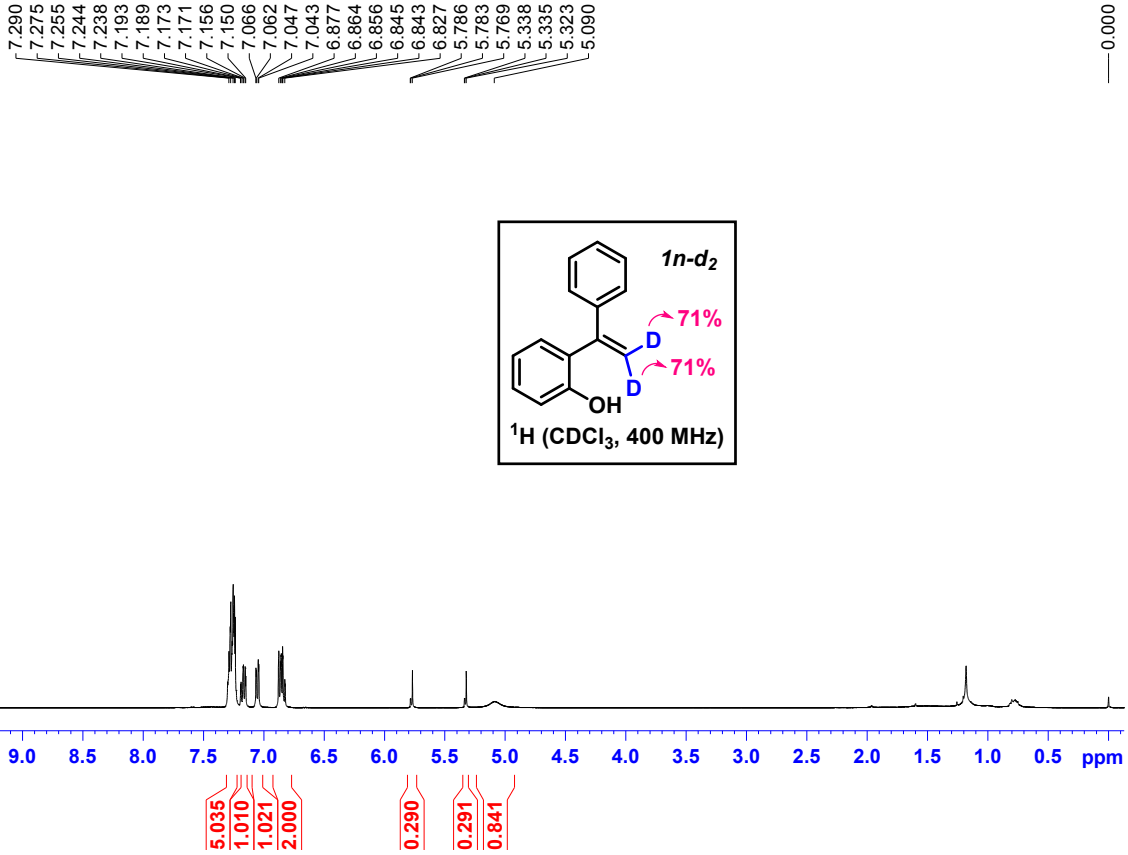


To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 2-(1-phenylvinyl)phenol **1n** (39.3 mg, 0.3 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), ***CD_3CO_2D (64.0 mg, 1.0 mmol, 5.0 equiv)***, anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product **3na-*d*** in 73% yield along with 51% deuterium insertion at alfa carbon to the carbonyl group of maleimide ring and 19% deuterium insertion at olefin carbon.



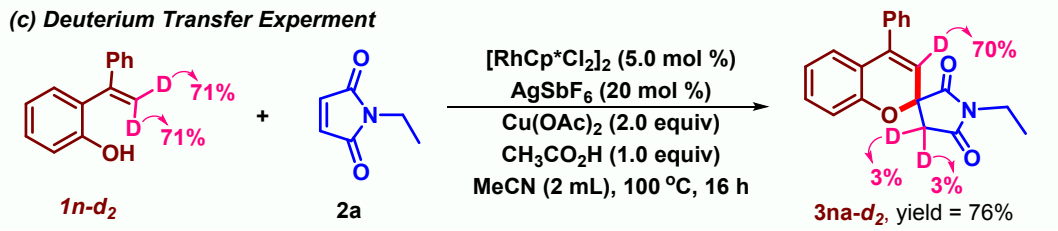


2-(1-Phenylvinyl-2,2-d2)phenol **1n-*d*$_2$** is a known molecule and prepared according to the literature procedure.[14] Methyl-*d*$_3$-triphenylphosphonium iodide was prepared according to the literature procedure.[15]

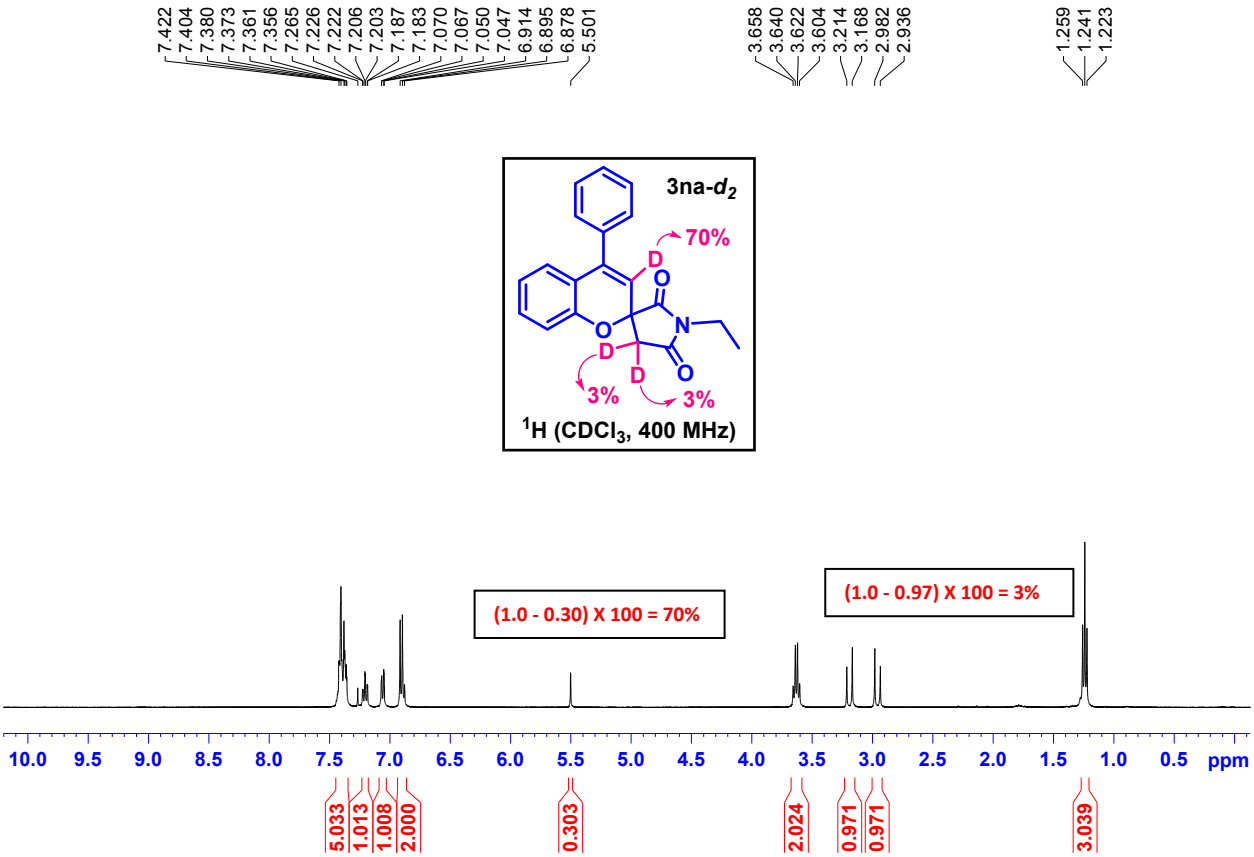


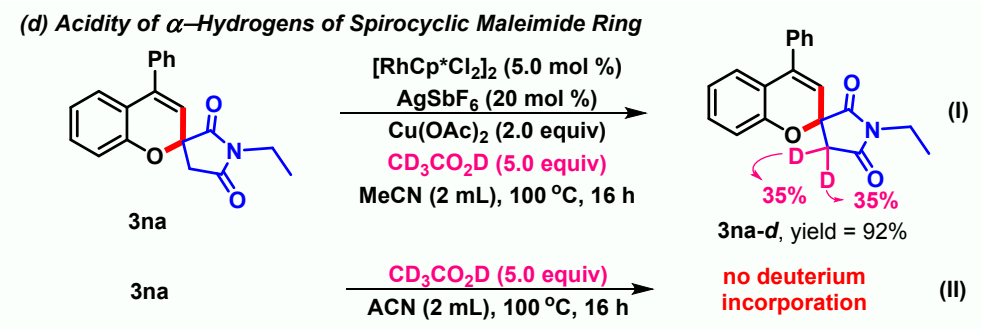
[14] A. Seoane, N. Casanova, N. Quiñones, J. L. Mascareñas, M. Gulías, *J. Am. Chem. Soc.,* 2014, **136**, 7607.

[15] J. J. Gajewski, K. B. Peterson, J. R. Kagel, Y. C. J. Huang, *J. Am. Chem. Soc.,* 1989, **111**, 9078.

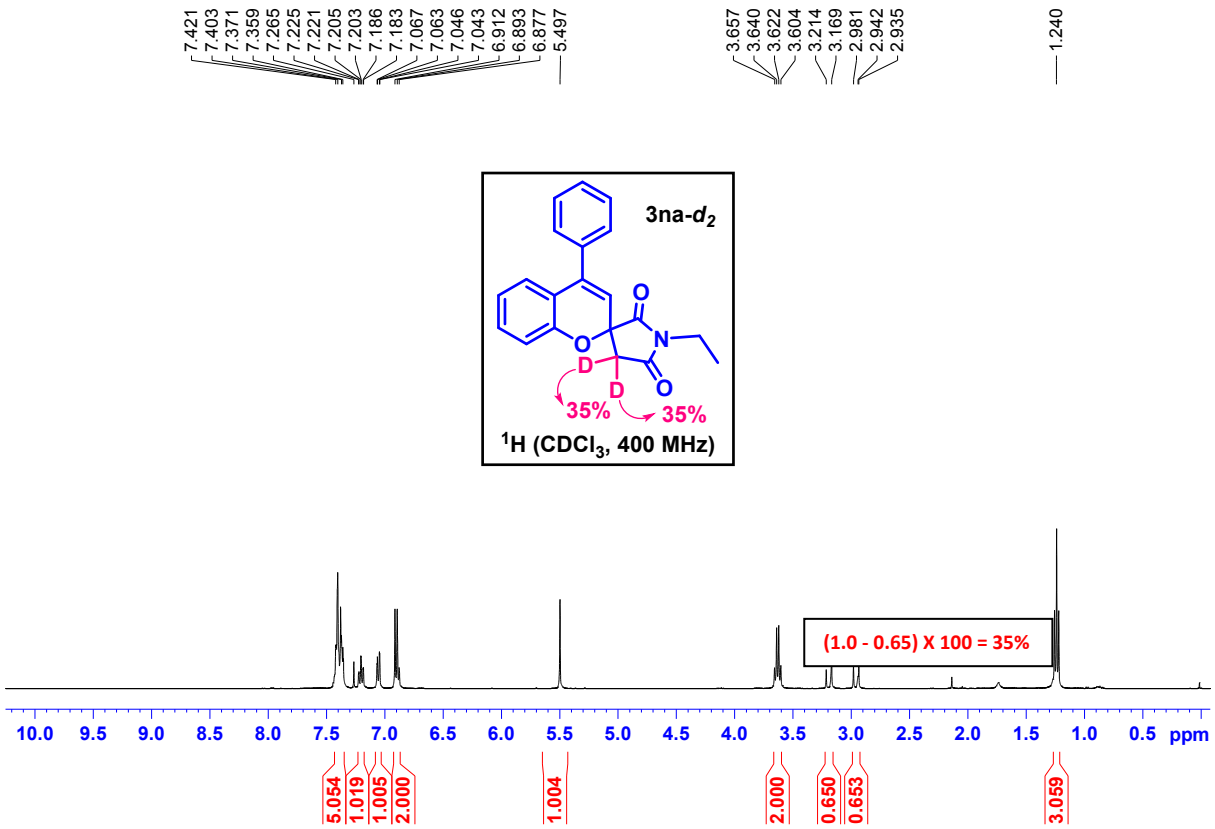


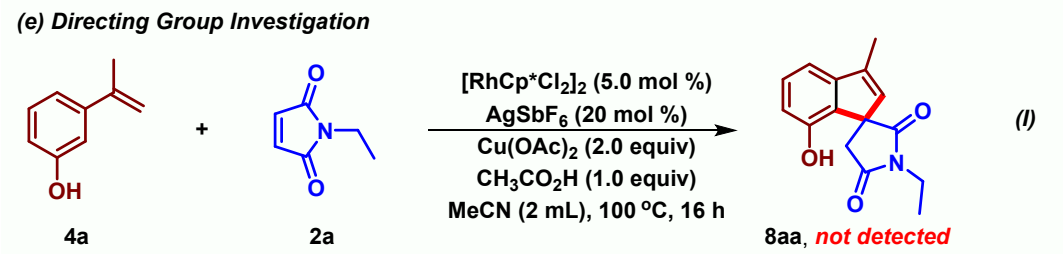
To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 2-(1-phenylvinyl-2,2-d_2)phenol ***1n-d*$_2$** (40.0 mg, 0.3 mmol, 1.5 equiv), [Cp*$RhCl_2$]$_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), CH_3CO_2H (12.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude product was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product **3na-*d*$_2$** in 76% yield along with 3% deuterium insertion at alfa carbon to the carbonyl group of maleimide ring and 70% deuterium intact at olefin carbon.



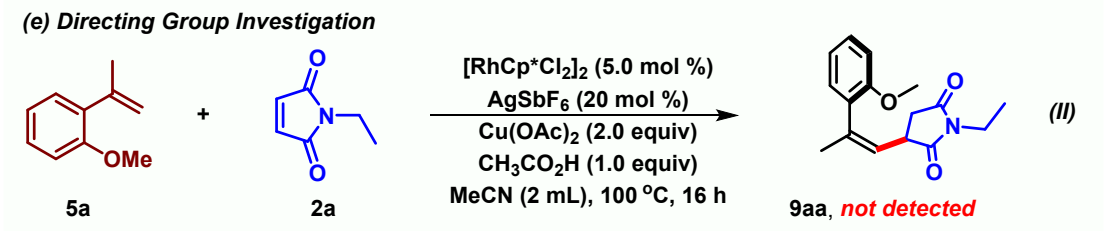


To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with 2-(1-phenylvinyl-2,2-d_2)phenol **3n*a*** (63.9 mg, 0.2 mmol), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), ***CD_3CO_2D (64.0 mg, 1.0 mmol, 5.0 equiv)***, anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the mixture was stirred for 16 h. Later, the mixture was cooled to room temperature, and the crude mixture was purified on a silica gel (230-400 mess size) flash column chromatography using EtOAc/ petroleum ether as eluent (3:97 to 7:93 *v/v*) to obtain the desired product **3na-*d*** in 92% yield along with 35% deuterium insertion at alfa carbon to the carbonyl group of maleimide ring. Interestingly, no deuterium incorporation occurred in absence of Rh(III)-catalyst.

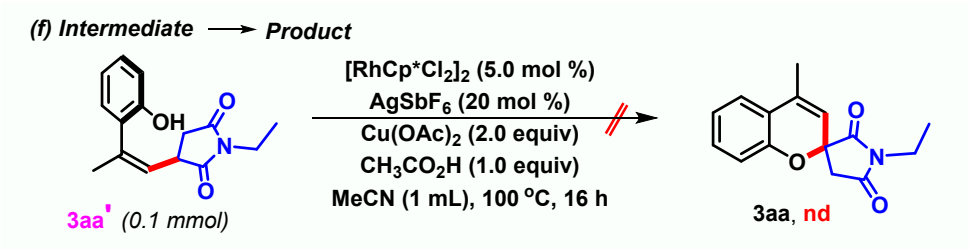




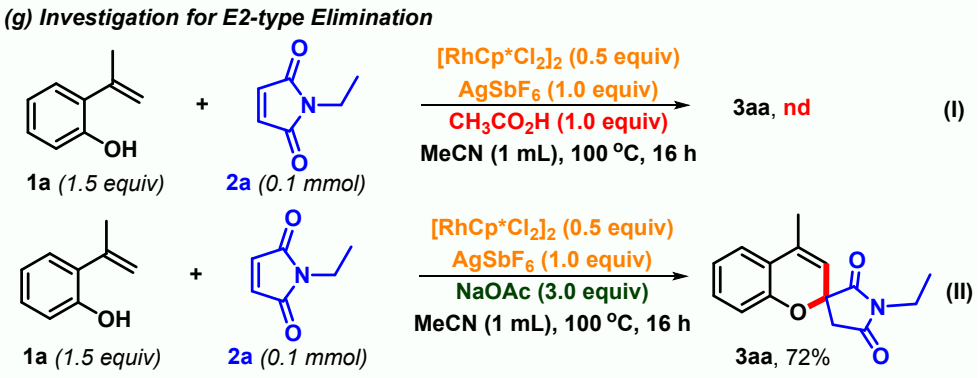
To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 3-(prop-1-en-2-yl)phenol **4a** (40.0 mg, 0.3 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), CH_3CO_2H (12.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC, which suggested both substrates *were intact* in the reaction mixture.



To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 1-methoxy-2-(prop-1-en-2-yl)benzene **5a** (44.5 mg, 0.3 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), CH_3CO_2H (12.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC, which suggested both substrates *were intact* in the reaction mixture.

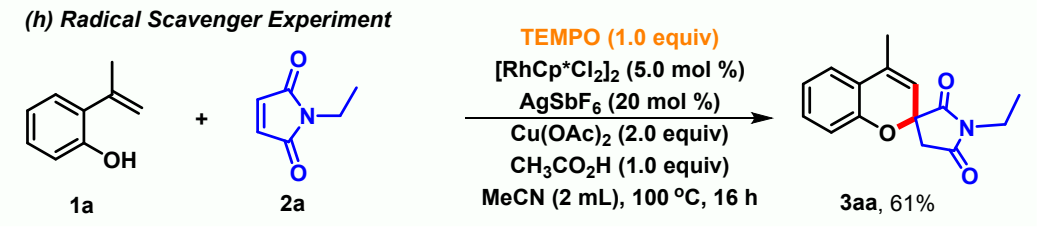


To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with **3aa'** (25.9 mg, 0.10 mmol) $[Cp^*RhCl_2]_2$ (3.1 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (6.9 mg, 20 mol%, 0.2 equiv), CH_3CO_2H (6.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (36.4 mg, 0.4 mmol, 2.0 equiv), and acetonitrile (1 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC, which suggested substrate **3aa'** was decomposed in several fan spots.

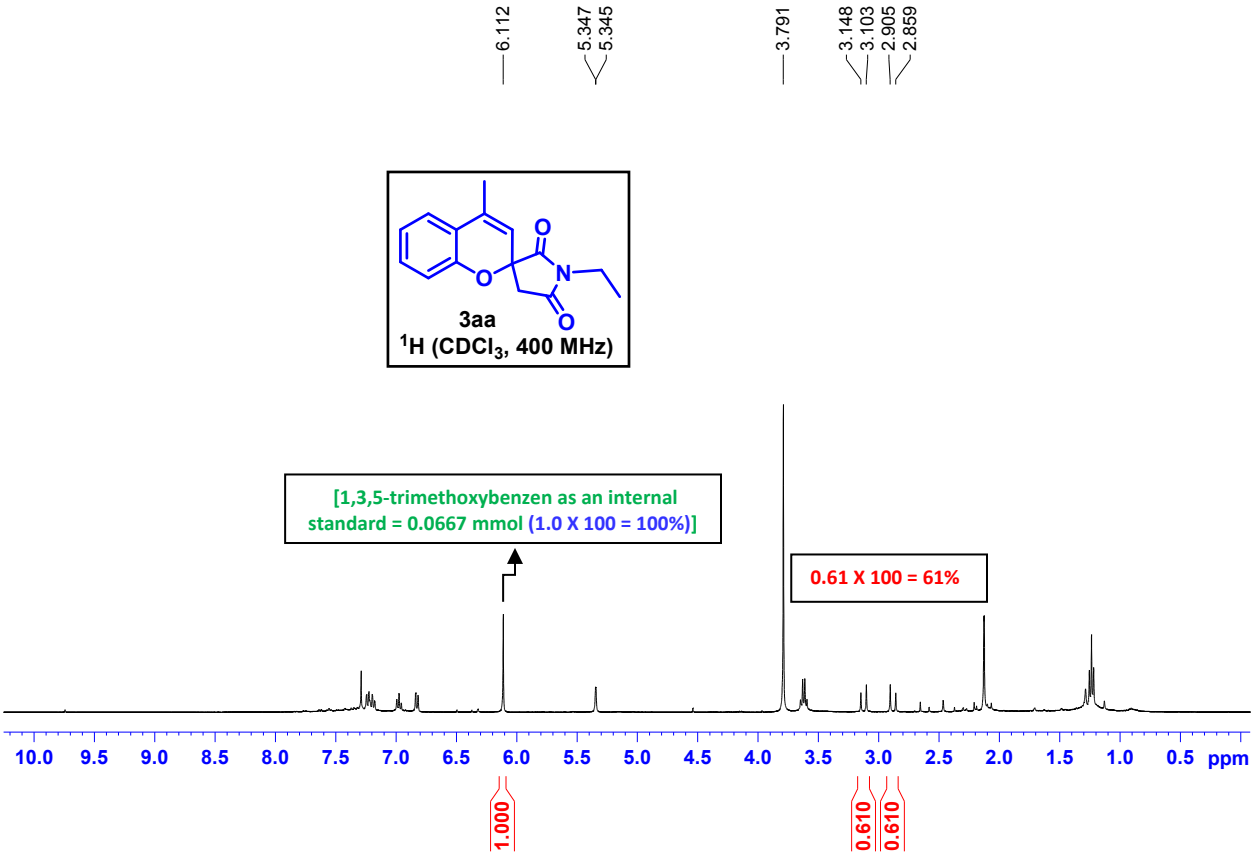


To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides (12.6 mg, 0.1 mmol) and 2-Isopropenylphenol (20.2 mg, 0.15 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (31.0 mg, 0.05 mmol, 0.5 equiv), $AgSbF_6$ (34.4 mg, 0.1 mmol, 1.0 equiv), AcOH (6.0 mg, 0.1 mmol, 1.0 equiv) in first experiment and NaOAc (24.6 mg, 0.3 mmol, 3.0 equiv) in second experiment and acetonitrile solvent (1 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred at the same temperature for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC, which suggested spirocyclic product **3aa** was not observed at all in the first experiment while 72% yield of **3aa** was observed in the second experiment.

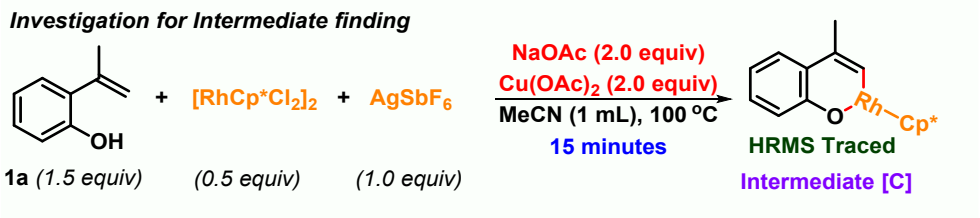
(h) Radical Scavenger Experiment



To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with maleimides **2a** (25.3 mg, 0.20 mmol), 2-Isopropenylphenol **1a** (40.0 mg, 0.3 mmol, 1.5 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 5 mol%, 0.05 equiv), $AgSbF_6$ (13.7 mg, 20 mol%, 0.2 equiv), CH_3CO_2H (12.0 mg, 0.2 mmol, 1.0 equiv), anhydrous $Cu(OAc)_2$ (72.8 mg, 0.4 mmol, 2.0 equiv), *TEMPO (1.0 equiv)* and acetonitrile (2 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, and the reaction mixture was stirred for 16 h. After completion of the reaction, the reaction mixture was cooled to room temperature. The crude mixture was filtered through a silica (230-400 mess size) pad using a mixture of EtOAc and petroleum ether (1:1, 100 mL) and concentrated under vacuo. The crude product was submitted for ^{1}H-NMR analysis to yield calculation for the desired product in 61% of NMR yield.



Attempt to synthesize six-membered Rhodacycle intermediate [c]



To an oven-dried 8-mL screw-cap reaction vial, equipped with a magnetic stir bar was charged with 2-Isopropenylphenol (13.4 mg, 0.1 mmol, 1.0 equiv), $[Cp^*RhCl_2]_2$ (31.0 mg, 0.05 mmol, 0.5 equiv), $AgSbF_6$ (34.4 mg, 0.1 mmol, 1.0 equiv), NaOAc (16.2 mg, 0.2 mmol, 2.0 equiv) and acetonitrile solvent (1 mL, 1M). The vial was sealed with a screw cap and placed in a pre-heated metal block at 100 °C, for 15 minutes. The reaction mixture was cooled to room temperature, and the crude mixture was checked by TLC, which suggested starting substrate 2-Isopropenylphenol **1a** was completely consumed. The reaction mixture was filtered through Whatman Filter Paper (solvent – ethyl acetate was used). And the filtrate was concentrated under vacuo. Next, the crude reaction mass was further analyzed by HRMS.

Intermediate [B]: HRMS (ESI-TOF) m/z $[M]^+$ Calculated for $C_{21}H_{27}O_3Rh$ 430.1032; found 430.1036.

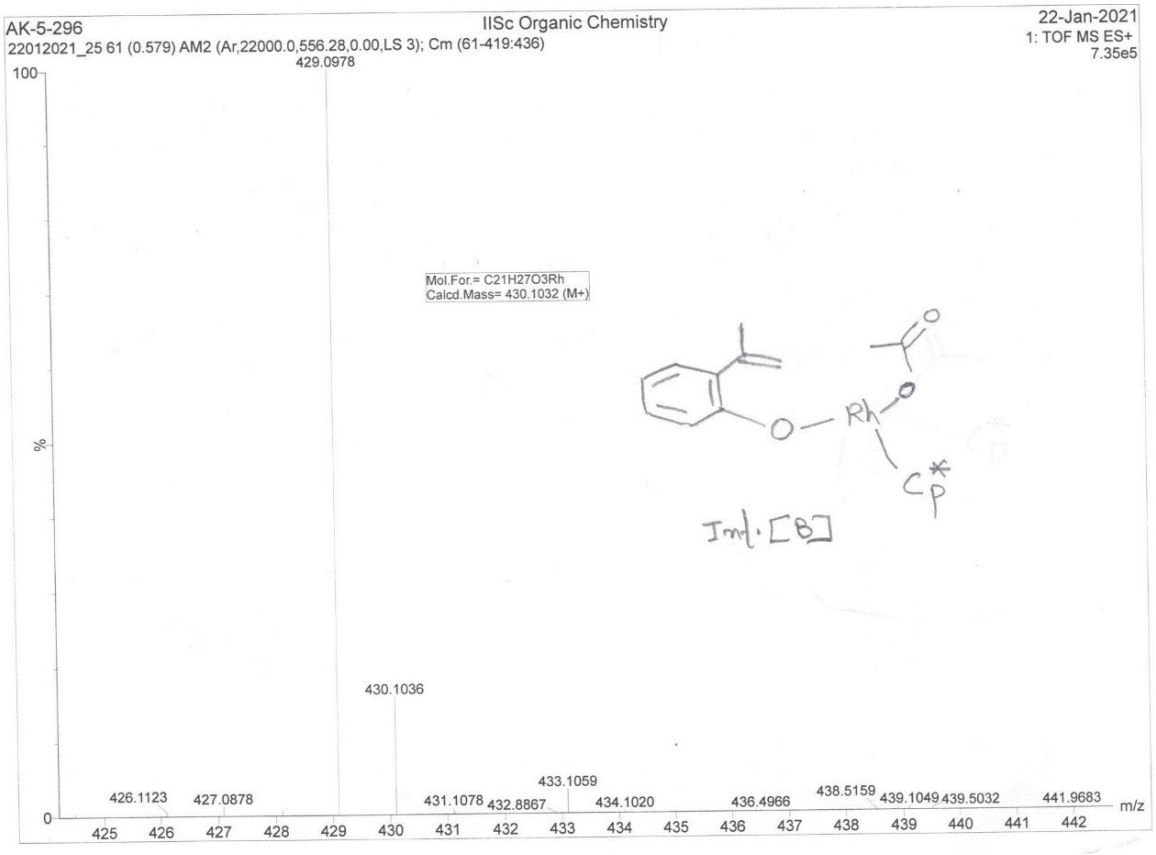


Figure: HRMS spectrum of intermediate **[B]**

Intermediate [C]: HRMS (ESI-TOF) m/z $[M + H]^+$ Calculated for $C_{19}H_{23}ORhH$ 371.0882; found 371.0885.

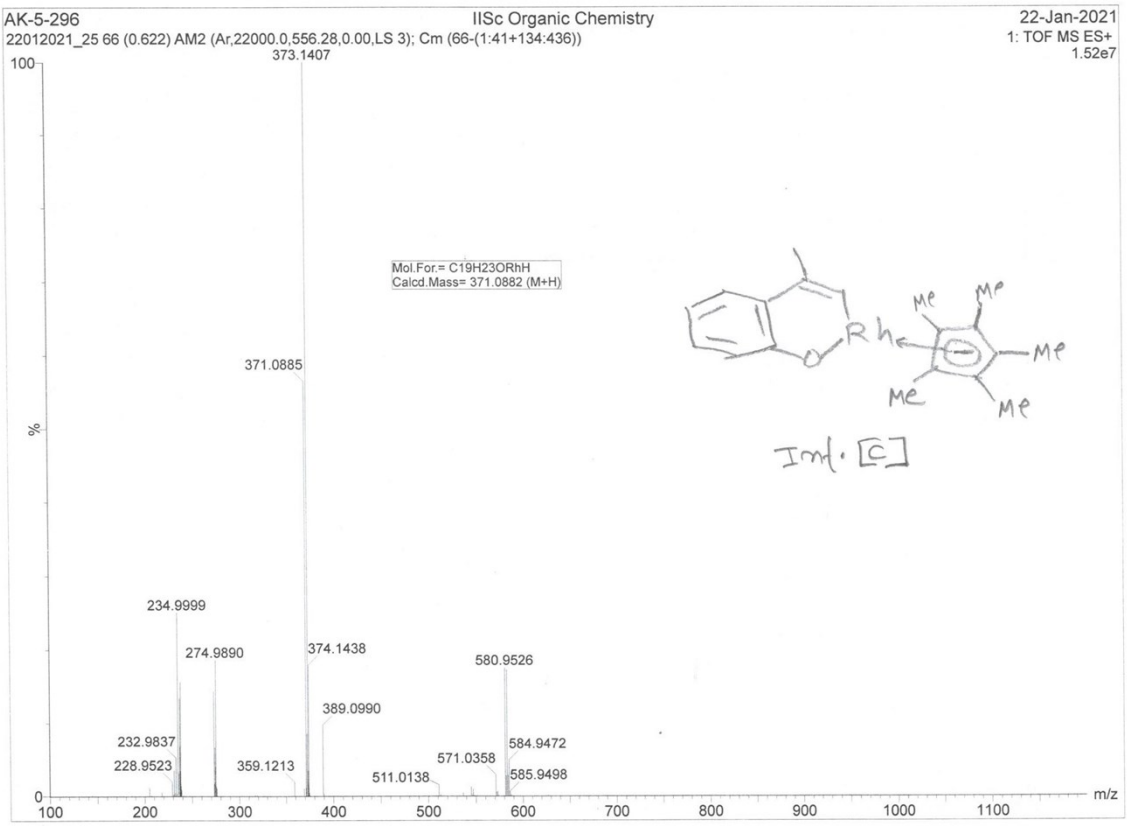
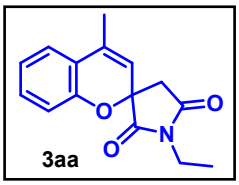


Figure: HRMS spectrum of intermediate **[C]**

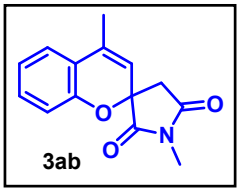
Characterization Data

(1) ***1'-Ethyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3aa).***



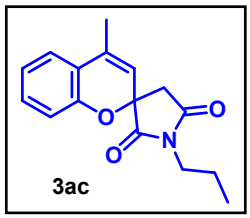
Prepared as shown in *general experimental procedure A*. **Yield** = 82% (42.2 mg); **Appearance** – White Solid; **mp** = 96 - 98 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.24 – 7.11 (m, 2H), 6.94 (td, *J* = 7.5, 1.0 Hz, 1H), 6.79 (dd, *J* = 98.0, 0.8 Hz, 1H), 5.31 (d, *J* = 1.2 Hz, 1H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.08 (d, *J* = 18.2 Hz, 1H), 2.84 (d, *J* = 18.2 Hz, 1H), 2.09 (d, *J* = 1.2 Hz, 3H), 1.20 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.2, 173.2, 151.9, 133.5, 130.0, 123.9, 121.9, 121.4, 117.1, 115.7, 78.8, 44.4, 34.0, 18.1, 12.9; **FT-IR (cm^{-1})** 2982, 2950, 2921, 1784, 1712, 1655, 1488, 1447, 1401, 1348, 1226, 1152; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{15}NO_3H$ 258.1130; found 258.1133.

(2) ***1',4-Dimethylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ab).***



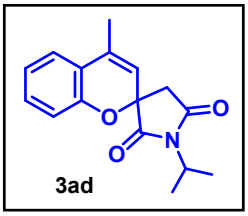
Prepared as shown in *general experimental procedure A*. **Yield** = 79% (38.5 mg); **Appearance** – Off White Solid; **mp** = 115 - 117 °C; R_f = 0.35 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.25 – 7.11 (m, 2H), 6.96 (td, *J* = 7.5, 0.9 Hz, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 5.32 (d, *J* = 1.2 Hz, 1H), 3.12 (d, *J* = 18.3 Hz, 1H), 3.04 (s, 3H), 2.88 (d, *J* = 18.3 Hz, 1H), 2.10 (d, *J* = 1.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.5, 173.4, 151.8, 133.7, 130.1, 124.0, 122.0, 121.3, 116.9, 115.7, 78.8, 44.4, 25.0, 18.1; **FT-IR (cm^{-1})** 2983, 2949, 2922, 1788, 1712, 1656, 1487, 1440, 1381, 1281, 1228, 1153; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{14}H_{13}NO_3Na$ 266.0793; found 266.0790.

(3) ***4-Methyl-1'-propylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ac).***



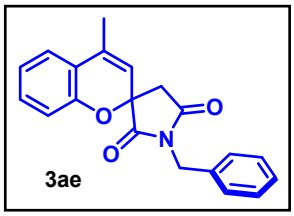
Prepared as shown in *general experimental procedure A*. **Yield** = 83% (45.1 mg); **Appearance** – Dark Yellow Oil; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.24 – 7.14 (m, 2H), 6.95 (td, *J* = 7.5, 0.9 Hz, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 5.32 (d, *J* = 1.1 Hz, 1H), 3.51 (t, *J* = 7.2 Hz, 2H), 3.11 (d, *J* = 18.2 Hz, 1H), 2.86 (d, *J* = 18.2 Hz, 1H), 2.10 (d, *J* = 1.1 Hz, 3H), 1.72 – 1.58 (m, 2H), 0.92 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.4, 173.4, 151.9, 133.6, 130.0, 123.9, 121.9, 121.4, 117.0, 115.7, 78.8, 44.3, 40.6, 20.9, 18.1, 11.2; **FT-IR (cm^{-1})** 2970, 2926, 2876, 1784, 1711, 1656, 1606, 1488, 1440, 1350, 1206, 1151, 1014, 929; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{16}H_{17}NO_3H$ 272.1287; found 272.1288.

(4) ***1'-Isopropyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ad).***



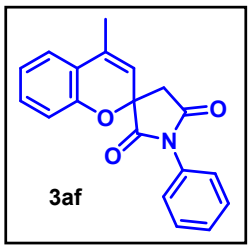
Prepared as shown in *general experimental procedure A*. **Yield** = 56% (30.4 mg); **Appearance** – Brown Solid; **mp** = 86 - 88 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.23 – 7.14 (m, 2H), 6.95 (t, *J* = 7.3 Hz, 1H), 6.81 (d, *J* = 8.0 Hz, 1H), 5.30 (s, 1H), 1.41 (sept, *J* = 6.9 Hz, 1H), 3.06 (d, *J* = 18.1 Hz, 1H), 2.82 (d, *J* = 18.1 Hz, 1H), 2.10 (d, *J* = 0.8 Hz, 3H), 1.41 (d, *J* = 6.9 Hz, 6H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.5, 173.2, 152.0, 133.6, 130.0, 123.9, 121.9, 121.4, 117.1, 115.6, 78.3, 44.3, 44.2, 19.3, 19.1, 18.2; **FT-IR (cm^{-1})** 2980, 2956, 2923, 1783, 1712, 1656, 1488, 1451, 1360, 1228, 1130; **HRMS (ESI-TOF) m/z** [M + H]$^+$ Calculated for $C_{16}H_{17}NO_3H$ 272.1287; found 272.1287.

(5) ***1'-Benzyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ae).***



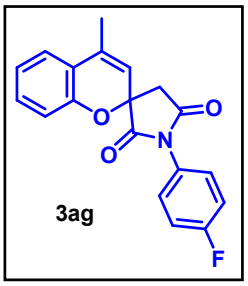
Prepared as shown in *general experimental procedure A*. **Yield** = 91% (58.2 mg); **Appearance** – White Solid; **mp** = 107 - 109 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.39 (dd, *J* = 7.7, 1.5 Hz, 2H), 7.36 – 7.29 (m, 3H), 7.23 – 7.15 (m, 2H), 6.96 (td, *J* = 7.5, 1.1 Hz, 1H), 6.79 (dd, *J* = 8.0, 0.7 Hz, 1H), 5.29 (d, *J* = 1.2 Hz, 1H), 4.69 (d, *J* = 1.2 Hz, 2H), 3.12 (d, *J* = 18.3 Hz, 1H), 2.88 (d, *J* = 18.3 Hz, 1H), 2.10 (d, *J* = 1.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.1, 173.0, 151.9, 135.3, 133.8, 130.1, 128.8, 128.8, 128.1, 123.9, 122.0, 121.4, 116.9, 115.7, 78.8, 44.4, 42.6, 18.1; **FT-IR (cm^{-1})** 2923, 2853, 1786, 1716, 1606, 1488, 1451, 1347, 1229, 1181; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{20}H_{17}NO_3Na$ 342.1106; found 342.1106.

(6) ***4-Methyl-1'-phenylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3af).***



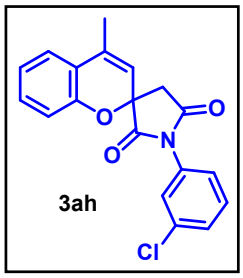
Prepared as shown in *general experimental procedure A*. **Yield** = 85% (52.1 mg); **Appearance** – Off White Solid; **mp** = 178 - 180 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.51 – 7.44 (m, 2H), 7.43 – 7.38 (m, 1H), 7.38 – 7.33 (m, 2H), 7.26 – 7.16 (m, 2H), 6.98 (td, *J* = 7.5, 1.1 Hz, 1H), 6.86 (dd, *J* = 8.0, 0.8 Hz, 1H), 5.46 (d, *J* = 1.3 Hz, 1H), 3.31 (d, *J* = 18.3 Hz, 1H), 3.06 (d, *J* = 18.3 Hz, 1H), 2.14 (d, *J* = 1.3 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.3, 172.3, 151.9, 134.1, 131.4, 130.2, 129.2, 128.8, 126.3, 124.0, 122.1, 121.4, 116.8, 115.7, 78.8, 44.4, 18.2; **FT-IR (cm^{-1})** 2922, 2852, 1791, 1716, 1653, 1494, 1381, 1198; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{19}H_{15}NO_3Na$ 328.0950; found 328.0953.

(7) ***1'-(4-Fluorophenyl)-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ag).***



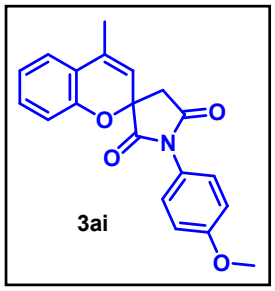
Prepared as shown in *general experimental procedure A*. **Yield** = 78% (50.5 mg); **Appearance** – Off White Solid; **mp** = 175 - 177 °C; R_f = 0.35 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 77.34 (ddd, *J* = 10.2, 5.1, 2.7 Hz, 2H), 7.26 – 7.13 (m, 4H), 6.98 (td, *J* = 7.5, 1.1 Hz, 1H), 6.85 (dd, *J* = 8.0, 0.9 Hz, 1H), 5.44 (d, *J* = 1.3 Hz, 1H), 3.30 (d, *J* = 18.4 Hz, 1H), 3.06 (d, *J* = 18.4 Hz, 1H), 2.14 (d, *J* = 1.3 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.2, 172.2, 162.3 ($J_{C\text{-}F}$ = 237.7 Hz), 151.8, 134.3, 130.2, 128.2 ($J_{C\text{-}F}$ = 8.8 Hz), 127.3, 124.0, 122.1, 121.3, 116.6, 116.2 ($J_{C\text{-}F}$ = 22.8 Hz), 115.7, 78.8, 44.3, 18.2; **FT-IR (cm^{-1})** 2962, 2925, 2847, 1793, 1722, 1653, 1610, 1513, 1448, 1385, 1252, 1030; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{19}H_{14}FNO_3Na$ 346.0855; found 346.0860.

(8) ***1'-(3-Chlorophenyl)-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ah).***



Prepared as shown in *general experimental procedure A*. **Yield** = 76% (51.8 mg); **Appearance** – Light Brown Oil; R_f = 0.30 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.44 – 7.35 (m, 3H), 7.30 – 7.27 (m, 1H), 7.26 – 7.18 (m, 2H), 6.98 (td, *J* = 7.5, 1.0 Hz, 1H), 6.86 (dd, *J* = 8.0, 0.8 Hz, 1H), 5.43 (d, *J* = 1.2 Hz, 1H), 3.30 (d, *J* = 18.4 Hz, 1H), 3.06 (d, *J* = 18.4 Hz, 1H), 2.14 (d, *J* = 1.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 172.9, 171.9, 151.8, 134.7, 134.4, 132.5, 130.2, 130.2, 129.0, 126.6, 124.5, 124.1, 122.2, 121.3, 116.5, 115.7, 78.8, 44.3, 18.2; **FT-IR (cm^{-1})** 2924, 2854, 1794, 1727, 1655, 1591, 1483, 1446, 1377, 1275, 1228, 1188; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{19}H_{14}ClNO_3Na$ 362.0560; found 362.0562.

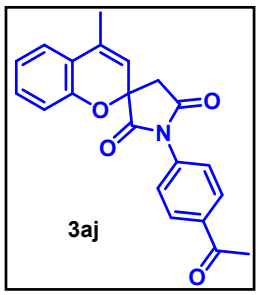
(9) ***1'-(4-Methoxyphenyl)-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ai).***



Prepared as shown in *general experimental procedure A*. **Yield** = 71% (47.8 mg); **Appearance** – Off White Solid; **mp** = 175 - 177 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.27 – 7.15 (m, 4H), 7.00 – 6.93 (m, 3H), 6.84 (d, *J* = 8.1 Hz, 1H), 5.44 (d, *J* = 1.2 Hz, 1H), 3.81 (s, 3H), 3.27 (d, *J* = 18.3 Hz, 1H), 3.03 (d, *J* = 18.3 Hz, 1H), 2.13 (d, *J* = 1.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.5, 172.6, 159.6, 151.9, 134.0, 130.1, 127.6, 124.0, 122.0, 121.4, 116.9, 115.7, 114.5, 78.8, 55.5, 44.3, 18.2; **FT-IR (cm^{-1})** 2925, 2847, 1793, 1722, 1610, 1513, 1485, 1448, 1385, 1252,

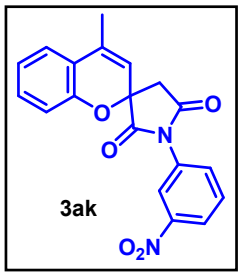
1195, 1030; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{20}H_{17}NO_4Na$ 358.1055; found 358.1052.

(10) ***1'-(4-Acetylphenyl)-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3aj).***



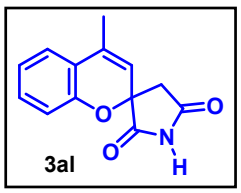
Prepared as shown in *general experimental procedure A*. **Yield** = 72% (50.1 mg); **Appearance** – Off White Solid; **mp** = 176 - 178 °C; R_f = 0.25 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.04 (d, *J* = 8.6 Hz, 2H), 7.51 (d, *J* = 8.6 Hz, 2H), 7.26 – 7.16 (m, 2H), 6.98 (td, *J* = 7.5, 0.9 Hz, 1H), 6.85 (d, *J* = 7.3 Hz, 1H), 5.45 (d, *J* = 1.2 Hz, 1H), 3.32 (d, *J* = 18.4 Hz, 1H), 3.08 (d, *J* = 18.4 Hz, 1H), 2.61 (s, 3H), 2.14 (d, *J* = 1.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 197.0, 172.9, 171.9, 151.8, 136.8, 135.5, 134.4, 130.2, 129.1, 126.2, 124.1, 122.2, 121.3, 116.4, 115.7, 78.8, 44.3, 26.7, 18.2; **FT-IR (cm^{-1})** 2923, 2846, 1794, 1726, 1684, 1604, 1485, 1378, 1267, 1190; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{21}H_{17}NO_4Na$ 370.1055; found 370.1053.

(11) ***4-Methyl-1'-(3-nitrophenyl)spiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ak).***



Prepared as shown in *general experimental procedure A*. **Yield** = 67% (46.8 mg); **Appearance** – Dark Brown Solid; **mp** = 114 - 116 °C; R_f = 0.25 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.33 (s, 1H), 8.26 (d, *J* = 8.2 Hz, 1H), 7.77 (d, *J* = 8.1 Hz, 1H), 7.66 (t, *J* = 8.2 Hz, 1H), 7.22 (dd, *J* = 13.6, 5.7 Hz, 2H), 6.99 (t, *J* = 7.5 Hz, 1H), 6.86 (d, *J* = 8.1 Hz, 1H), 5.46 (s, 1H), 3.35 (d, *J* = 18.4 Hz, 1H), 3.12 (d, *J* = 18.4 Hz, 1H), 2.16 (d, *J* = 0.8 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 172.6, 171.6, 151.7, 148.5, 134.7, 132.5, 132.1, 130.3, 123.0, 124.1, 123.4, 122.3, 121.6, 121.2, 116.1, 115.7, 78.9, 44.3, 18.2; **FT-IR (cm^{-1})** 2924, 2854, 1796, 1727, 1650, 1533, 1484, 1379, 1350, 1185; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{19}H_{14}N_2O_5Na$ 373.0800; found 373.0799.

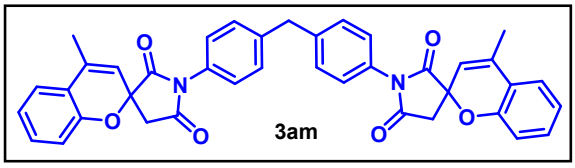
(12) ***4-Methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3al).***



Prepared as shown in *general experimental procedure A*. **Yield** = 76% (34.8 mg); **Appearance** – Off White Solid; **mp** = 126 - 128 °C; R_f = 0.30 (30% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.73 (s, 1H), 7.19 (dd, *J* = 15.1, 7.6 Hz, 2H), 6.96 (t, *J* = 7.6 Hz, 1H), 6.82 (d, *J* = 8.0 Hz, 1H), 5.36 (d, *J* = 1.3 Hz, 1H), 3.15 (d, *J* = 18.4 Hz, 1H), 2.91 (d, *J* = 18.4 Hz, 1H), 2.10 (d, *J* = 1.3 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.7, 173.5, 151.7, 133.8, 130.1, 124.0, 122.1,

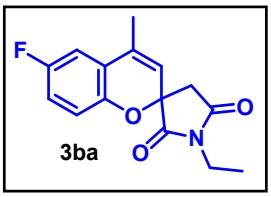
121.2, 116.5, 115.7, 80.0, 45.3, 18.1; **FT-IR (cm^{-1})** 3231, 2923, 2855, 1790, 1732, 1657, 1487, 1447, 1381, 1345, 1234, 1194, 1042; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{13}H_{11}NO_3H$ 230.0817; found 230.0828.

(13) ***1',1'''-(Methylenebis(4,1-phenylene))bis(4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione) (3am).***



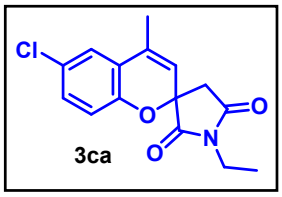
Prepared as shown in *general experimental procedure A*. **Yield** = 66% (82.3 mg); **Appearance** – Light Brown Solid; **mp** = 216 - 218 °C; R_f = 0.20 (30% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.26 (s, 8H), 7.20 (ddd, *J* = 17.0, 8.4, 1.4 Hz, 4H), 7.00 – 6.91 (m, 2H), 6.84 (dd, *J* = 8.0, 0.7 Hz, 2H), 5.43 (d, *J* = 1.2 Hz, 2H), 4.02 (s, 2H), 3.27 (d, *J* = 18.3 Hz, 2H), 3.03 (d, *J* = 18.3 Hz, 2H), 2.12 (d, *J* = 1.2 Hz, 6H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.3, 172.4, 151.8, 141.0, 134.0, 130.1, 129.7, 129.6, 126.4, 124.0, 122.0, 121.4, 116.8, 115.8, 78.8, 44.4, 41.1, 18.2; **FT-IR (cm^{-1})** 2983, 2949, 2922, 1712, 1656, 1487, 1440, 1381, 1281, 1228, 1153; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{39}H_{30}N_2O_6Na$ 645.2002; found 645.2007.

(14) ***1'-Ethyl-6-fluoro-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ba).***



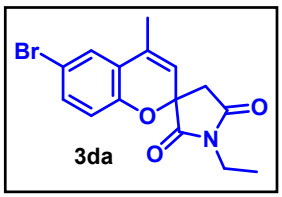
Prepared as shown in *general experimental procedure A*. **Yield** = 71% (58.8 mg); **Appearance** – White Solid; **mp** = 133 - 135 °C; R_f = 0.50 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 6.91 (dd, *J* = 9.0, 3.0 Hz, 1H), 6.85 (td, *J* = 8.4, 3.0 Hz, 1H), 6.74 (dd, *J* = 8.8, 4.7 Hz, 1H), 5.38 (s, 1H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.08 (d, *J* = 18.3 Hz, 1H), 2.85 (d, *J* = 18.3 Hz, 1H), 2.07 (d, *J* = 1.4 Hz, 3H), 1.20 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.0, 173.0, 157.8 ($J_{C\text{-}F}$ = 237.7 Hz), 147.8 ($J_{C\text{-}F}$ = 2.1 Hz), 133.10 ($J_{C\text{-}F}$ = 1.8 Hz), 122.6 ($J_{C\text{-}F}$ = 7.9 Hz), 118.3, 116.5 ($J_{C\text{-}F}$ = 8.1 Hz), 116.0 ($J_{C\text{-}F}$ = 23.2 Hz), 110.7 ($J_{C\text{-}F}$ = 24.6 Hz), 78.8, 44.1, 34.1, 18.0, 12.90; **FT-IR (cm^{-1})** 2983, 2946, 2852, 1785, 1722, 1656, 1587, 1485, 1434, 1402, 1379, 1347, 1260, 1226, 1149, 1026, 901; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{14}FNO_3H$ 276.1036; found 276.1032.

(15) ***6-Chloro-1'-ethyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ca).***



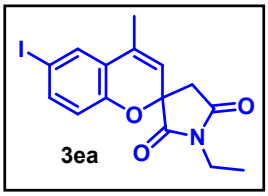
Prepared as shown in *general experimental procedure A*. **Yield** = 79% (69.4 mg); **Appearance** – White Solid; **mp** = 159 - 161 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.16 (d, *J* = 2.4 Hz, 1H), 7.11 (dd, *J* = 8.5, 2.4 Hz, 1H), 6.73 (d, *J* = 8.5 Hz, 1H), 5.36 (s, 1H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.08 (d, *J* = 18.3 Hz, 1H), 2.86 (d, *J* = 18.3 Hz, 1H), 2.07 (d, *J* = 1.1 Hz, 3H), 1.20 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.8, 172.9, 150.5, 132.9, 129.6, 126.9, 123.9, 122.8, 118.2, 116.9, 78.9, 44.2, 34.1, 18.0, 12.9; **FT-IR (cm^{-1})** 2958, 2923, 2852, 1784, 1712, 1655, 1446, 1401, 1347, 1225, 1151, 1039, 891; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{15}H_{14}ClNO_3Na$ 314.0560; found 314.0563.

(16) ***6-Bromo-1'-ethyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3da).***



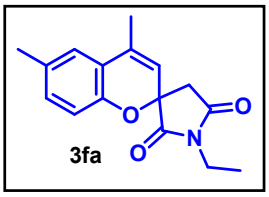
Prepared as shown in *general experimental procedure A*. **Yield** = 72% (48.2 mg); **Appearance** – White Solid; **mp** = 157 - 159 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.34 (d, *J* = 2.3 Hz, 1H), 7.34 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.69 (d, *J* = 8.5 Hz, 1H), 5.36 (d, *J* = 1.3 Hz, 1H), 3.60 (q, *J* = 7.2 Hz, 2H), 3.09 (d, *J* = 18.3 Hz, 1H), 2.87 (d, *J* = 18.3 Hz, 1H), 2.08 (d, *J* = 1.3 Hz, 3H), 1.21 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.8, 172.8, 151.0, 132.9, 132.6, 126.8, 123.3, 118.2, 117.4, 114.2, 78.9, 44.2, 34.1, 18.0, 12.9; **FT-IR (cm^{-1})** 2981, 2930, 2851, 1781, 1711, 1652, 1481, 1444, 1402, 1345, 1225, 1150, 1025; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{14}BrNO_3H$ 336.0235; found 336.0235.

(17) ***1'-Ethyl-6-iodo-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ea).***



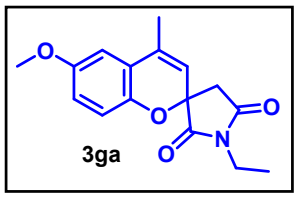
Prepared as shown in *general experimental procedure A*. **Yield** = 72% (55.2 mg); **Appearance** – White Solid; **mp** = 156 - 158 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.17 (d, *J* = 2.4 Hz, 1H), 7.13 (dd, *J* = 8.5, 2.5 Hz, 1H), 6.74 (d, *J* = 8.5 Hz, 1H), 5.37 (d, *J* = 1.3 Hz, 1H), 3.59 (q, *J* = 7.2 Hz, 2H), 3.09 (d, *J* = 18.3 Hz, 1H), 2.87 (d, *J* = 18.3 Hz, 1H), 2.09 (d, *J* = 1.3 Hz, 3H), 1.21 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.8, 172.9, 150.5, 133.0, 129.6, 126.9, 123.9, 122.8, 118.2, 116.9, 78.9, 44.2, 34.1, 18.0, 12.9; **FT-IR (cm^{-1})** 2960, 2924, 2851, 1786, 1713, 1654, 1477, 1458, 1404, 1375, 1345, 1226, 1025; **HRMS (ESI-TOF) m/z** $[M + Na - HI]^+$ Calculated for $C_{15}H_{14}NO_3Na$ 279.0871; found 279.0949.

(18) ***1'-Ethyl-4,6-dimethylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3fa).***



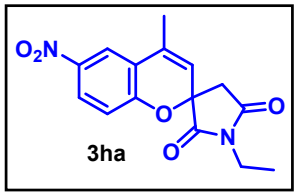
Prepared as shown in *general experimental procedure A*. **Yield** = 71% (38.6 mg); **Appearance** – White Solid; **mp** = 114 - 116 °C; R_f = 0.40 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.01 (s, 1H), 6.98 (d, *J* = 8.1 Hz, 1H), 6.70 (d, *J* = 8.1 Hz, 1H), 5.32 (d, *J* = 1.3 Hz, 1H), 3.59 (q, *J* = 7.2 Hz, 2H), 3.09 (d, *J* = 18.2 Hz, 1H), 2.84 (d, *J* = 18.2 Hz, 1H), 2.29 (s, 3H), 2.09 (d, *J* = 1.3 Hz, 3H), 1.21 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.4, 173.3, 149.7, 133.8, 131.1, 130.4, 124.5, 121.2, 117.1, 115.4, 78.6, 44.3, 34.0, 20.8, 18.2, 12.9; **FT-IR (cm^{-1})** 2981, 2921, 2855, 1785, 1711, 1654, 1490, 1445, 1403, 1375, 1344, 1220, 1150, 1026, 990, 925, 892; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{16}H_{17}NO_3H$ 272.1287; found 272.1284.

(19) ***1'-Ethyl-6-methoxy-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ga).***



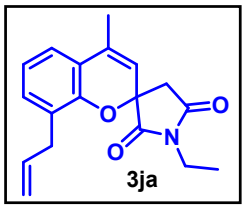
Prepared as shown in *general experimental procedure A*. **Yield** = 68% (39.2 mg); **Appearance** – Off White Solid; **mp** = 94 - 96 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 6.77 (d, *J* = 2.6 Hz, 1H), 6.77 – 6.69 (m, 2H), 5.36 (d, *J* = 1.3 Hz, 1H), 3.77 (s, 3H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.08 (d, *J* = 18.2 Hz, 1H), 2.83 (d, *J* = 18.2 Hz, 1H), 2.08 (d, *J* = 1.3 Hz, 3H), 1.20 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.3, 173.3, 154.5, 145.8, 133.7, 122.3, 118.0, 116.1, 114.4, 110.1, 78.6, 55.8, 44.1, 34.0, 18.2, 12.9; **FT-IR (cm^{-1})** 2982, 2944, 2835, 1783, 1721, 1654, 1580, 1489, 1401, 1347, 1269, 1231, 1203, 1149; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{16}H_{17}NO_4Na$ 310.1055; found 310.1051.

(20) ***1'-Ethyl-4-methyl-6-nitrospiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ha).***



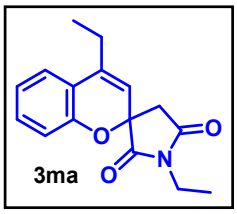
Prepared as shown in *general experimental procedure A*. **Yield** = 38% (22.9 mg); **Appearance** – Light Brown Solid; **mp** = 173 - 175 °C; R_f = 0.25 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.19 – 7.97 (m, 2H), 6.88 (d, *J* = 9.2 Hz, 1H), 5.46 (d, *J* = 1.1 Hz, 1H), 3.60 (q, *J* = 7.2 Hz, 2H), 3.13 (d, *J* = 18.3 Hz, 1H), 2.95 (d, *J* = 18.3 Hz, 1H), 2.17 (s, 3H), 1.21 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.1, 172.3, 157.2, 142.5, 132.5, 126.0, 121.7, 119.9, 118.9, 116.1, 79.8, 44.2, 34.3, 18.1, 12.9; **FT-IR (cm^{-1})** 2960, 2924, 2852, 1786, 1713, 1523, 1459, 1343, 1224; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{15}H_{14}N_2O_5H$ 303.0981; found 303.0978.

(21) ***8-Allyl-1'-ethyl-4-methylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ja).***



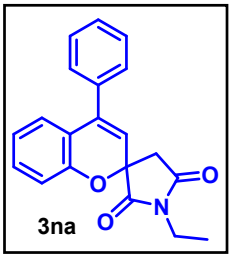
Prepared as shown in *general experimental procedure A*. **Yield** = 67% (40.1 mg); **Appearance** – Light Yellow Solid; **mp** = 66 - 68 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.11 (dd, *J* = 7.6, 1.4 Hz, 1H), 7.05 (d, *J* = 6.5 Hz, 1H), 6.90 (t, *J* = 7.6 Hz, 1H), 5.88 (ddt, *J* = 16.8, 10.5, 6.5 Hz, 1H), 5.34 (d, *J* = 1.2 Hz, 1H), 5.02 – 4.93 (m, 2H), 3.60 (q, *J* = 7.2 Hz, 2H), 3.32 – 3.25 (m, 2H), 3.08 (d, *J* = 18.2 Hz, 1H), 2.85 (d, *J* = 18.2 Hz, 1H), 2.11 (d, *J* = 1.2 Hz, 3H), 1.21 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.1, 173.3, 149.5, 136.5, 134.0, 130.9, 126.9, 122.2, 121.6, 121.4, 116.8, 115.6, 78.7, 44.4, 34.0, 33.9, 18.4, 12.9; **FT-IR (cm^{-1})** 2979, 2946, 2852, 1785, 1713, 1657, 1448, 1401, 1348, 1225; **HRMS (ESI-TOF) m/z** [M + H]$^+$ Calculated for $C_{18}H_{19}NO_3H$ 298.1443; found 298.1446.

(22) ***1',4-Diethylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ma).***



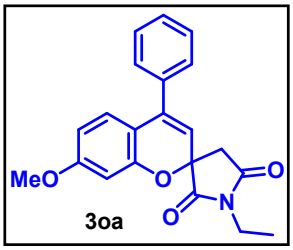
Prepared as shown in *general experimental procedure A*. **Yield** = 86% (46.8 mg); **Appearance** – Light Yellow Solid; **mp** = 74 - 76 °C; R_f = 0.55 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.24 (dd, *J* = 7.8, 1.4 Hz, 1H), 7.16 (td, *J* = 7.8, 1.4 Hz, 1H), 6.94 (ddd, *J* = 7.7, 1.1, 0.5 Hz, 1H), 6.81 (dd, *J* = 8.0, 1.1 Hz, 1H), 5.30 (s, 1H), 3.59 (q, *J* = 7.2 Hz, 2H), 3.10 (d, *J* = 18.2 Hz, 1H), 2.86 (d, *J* = 18.2 Hz, 1H), 2.49 (qd, *J* = 7.2, 1.2 Hz, 2H), 1.21 (t, *J* = 7.2 Hz, 3H), 1.18 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.3, 173.2, 152.0, 138.9, 129.8, 123.5, 121.9, 120.9, 115.9, 115.2, 78.8, 44.4, 34.0, 24.0, 12.9, 11.9; **FT-IR (cm^{-1})** 2973, 2936, 2845, 1785, 1715, 1652, 1488, 1449, 1401, 1347, 1226, 1152, 1013, 894; **HRMS (ESI-TOF) m/z** [M + H]$^+$ Calculated for $C_{16}H_{17}NO_3H$ 272.1287; found 272.1290.

(23) ***1'-Ethyl-4-phenylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3na).***



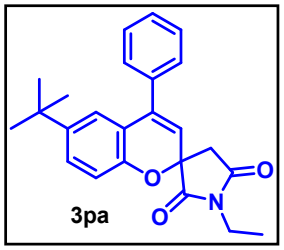
Prepared as shown in *general experimental procedure A*. **Yield** = 89% (56.9 mg); **Appearance** – Colourless Oil; R_f = 0.60 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.44 – 7.34 (m, 5H), 7.21 (td, *J* = 8.0, 1.5 Hz, 1H), 7.05 (dd, *J* = 8.0, 1.5 Hz, 1H), 6.93 – 6.86 (m, 2H), 5.50 (s, 1H), 3.63 (q, *J* = 7.2 Hz, 2H), 3.20 (d, *J* = 18.3 Hz, 1H), 2.96 (d, *J* = 18.3 Hz, 1H), 1.24 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.9, 173.1, 152.3, 140.0, 136.9, 130.3, 128.7, 128.5, 128.5, 126.4, 122.0, 121.0, 118.5, 116.2, 78.8, 44.2, 34.1, 13.0; **FT-IR (cm^{-1})** 2971, 2925, 2853, 1785, 1712, 1449, 1400, 1349, 1228, 1153; **HRMS (ESI-TOF) m/z** [M + Na]$^+$ Calculated for $C_{20}H_{17}NO_3Na$ 342.1106; found 342.1103.

(24) ***1'-Ethyl-7-methoxy-4-phenylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3oa).***



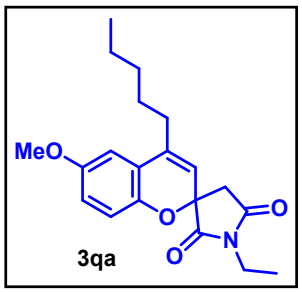
Prepared as shown in *general experimental procedure A*. **Yield** = 83% (58.1 mg); **Appearance** – Yellow Oil; R_f = 0.55 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.42 – 7.34 (m, 5H), 6.97 (d, *J* = 8.5 Hz, 1H), 6.49 (d, *J* = 2.5 Hz, 1H), 6.44 (dd, *J* = 8.5, 2.5 Hz, 1H), 5.35 (s, 1H), 3.79 (s, 3H), 3.63 (q, *J* = 7.2 Hz, 2H), 3.18 (d, *J* = 18.2 Hz, 1H), 2.95 (d, *J* = 18.2 Hz, 1H), 1.62 (s, 2H), 1.24 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.1, 173.1, 161.4, 153.7, 139.9, 137.2, 128.6, 128.5, 128.4, 127.3, 115.6, 114.2, 107.6, 102.2, 79.0, 55.4, 44.3, 34.1, 12.9; **FT-IR (cm^{-1})** 2977, 2956, 2925, 2844, 1786, 1714, 1639, 1499, 1441, 1383, 1281, 1164, 1011, 1034; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{21}H_{19}NO_4H$ 350.1392; found 350.1392.

(25) ***6-(tert-Butyl)-1'-ethyl-4-phenylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3pa).***



Prepared as shown in *general experimental procedure A*. **Yield** = 82% (61.7 mg); **Appearance** – Yellow Oil; R_f = 0.55 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.42 – 7.34 (m, 5H), 6.97 (d, *J* = 8.5 Hz, 1H), 6.49 (d, *J* = 2.5 Hz, 1H), 6.44 (dd, *J* = 8.5, 2.5 Hz, 1H), 5.35 (s, 1H), 3.79 (s, 3H), 3.63 (q, *J* = 7.2 Hz, 2H), 3.18 (d, *J* = 18.2 Hz, 1H), 2.95 (d, *J* = 18.2 Hz, 1H), 1.62 (s, 2H), 1.24 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.1, 173.1, 161.4, 153.7, 139.9, 137.2, 128.6, 128.5, 128.4, 127.3, 115.6, 114.2, 107.6, 102.2, 79.0, 55.4, 44.3, 34.1, 12.9; **FT-IR (cm^{-1})** 2959, 2924, 2853, 1786, 1714, 1634, 1490, 1449, 1400, 1345, 1225; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{24}H_{25}NO_3Na$ 398.1732; found 398.1730.

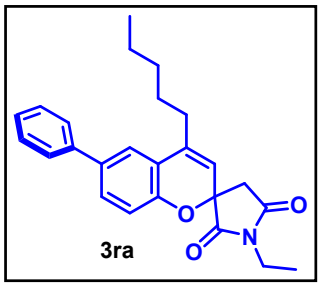
(26) ***1'-Ethyl-6-methoxy-4-pentylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3qa).***



Prepared as shown in *general experimental procedure A*. **Yield** = 71% (48.6 mg); **Appearance** – Light Brown Solid; **mp** = 76 - 78 °C; R_f = 0.60 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 6.79 (d, *J* = 2.8 Hz, 1H), 6.74 (d, *J* = 8.7 Hz, 1H), 6.70 (dd, *J* = 8.7, 2.8 Hz, 1H), 5.34 (s, 1H), 3.77 (s, 3H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.08 (d, *J* = 18.2 Hz, 1H), 2.83 (d, *J* = 18.2 Hz, 1H), 2.44 – 2.39 (m, 2H), 1.58 – 1.52 (m, 2H), 1.37 – 1.34 (m, 4H), 1.20 (t, *J* = 7.2 Hz, 3H), 0.90 (t, *J* = 7.0 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.3, 173.3, 154.5, 146.0, 137.7, 121.8, 117.0, 116.4, 114.0, 110.0, 78.6, 55.7, 44.1, 34.0, 31.6, 31.3, 27.4, 22.5, 14.0, 12.9; **FT-IR (cm^{-1})** 2958, 2929, 2862, 1784, 1714,

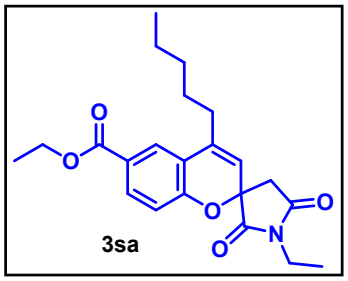
1650, 1578, 1489, 1400, 1347, 1268, 1228, 1201, 1149, 1044; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{20}H_{25}NO_4H$ 344.1862; found 344.1859.

(27) ***1'-Ethyl-4-pentyl-6-phenylspiro[chromene-2,3'-pyrrolidine]-2',5'-dione (3ra).***



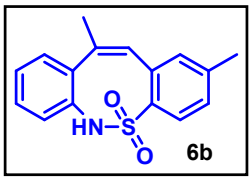
Prepared as shown in *general experimental procedure A*. **Yield** = 65% (50.5 mg); **Appearance** – White Solid; **mp** = 145 - 147 °C; R_f = 0.55 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.56 – 7.53 (m, 2H), 7.46 – 7.42 (m, 3H), 7.40 (dd, *J* = 8.3, 2.1 Hz, 1H), 7.34 (t, *J* = 7.3 Hz, 1H), 6.90 (d, *J* = 8.3 Hz, 1H), 5.35 (s, 1H), 3.62 (q, *J* = 7.2 Hz, 2H), 3.15 (d, *J* = 18.2 Hz, 1H), 2.90 (d, *J* = 18.2 Hz, 1H), 2.58 – 2.45 (m, 2H), 1.61 (dd, *J* = 15.7, 8.5 Hz, 3H), 1.43 – 1.32 (m, 4H), 1.24 (t, *J* = 7.2 Hz, 3H), 0.92 (t, *J* = 7.0 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 174.2, 173.2, 151.7, 140.9, 137.7, 135.2, 128.8, 128.6, 127.0, 126.9, 122.5, 121.0, 116.4, 116.2, 78.9, 44.5, 34.1, 31.6, 31.2, 27.3, 22.5, 14.0, 12.9; **FT-IR (cm^{-1})** 2958, 2926, 2855, 1784, 1713, 1650, 1481, 1401, 1375, 1347, 1225, 1150; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{25}H_{27}NO_3Na$ 412.1889; found 412.1886.

(28) ***Ethyl 1'-ethyl-2',5'-dioxo-4-pentylspiro[chromene-2,3'-pyrrolidine]-6-carboxylate (3sa).***



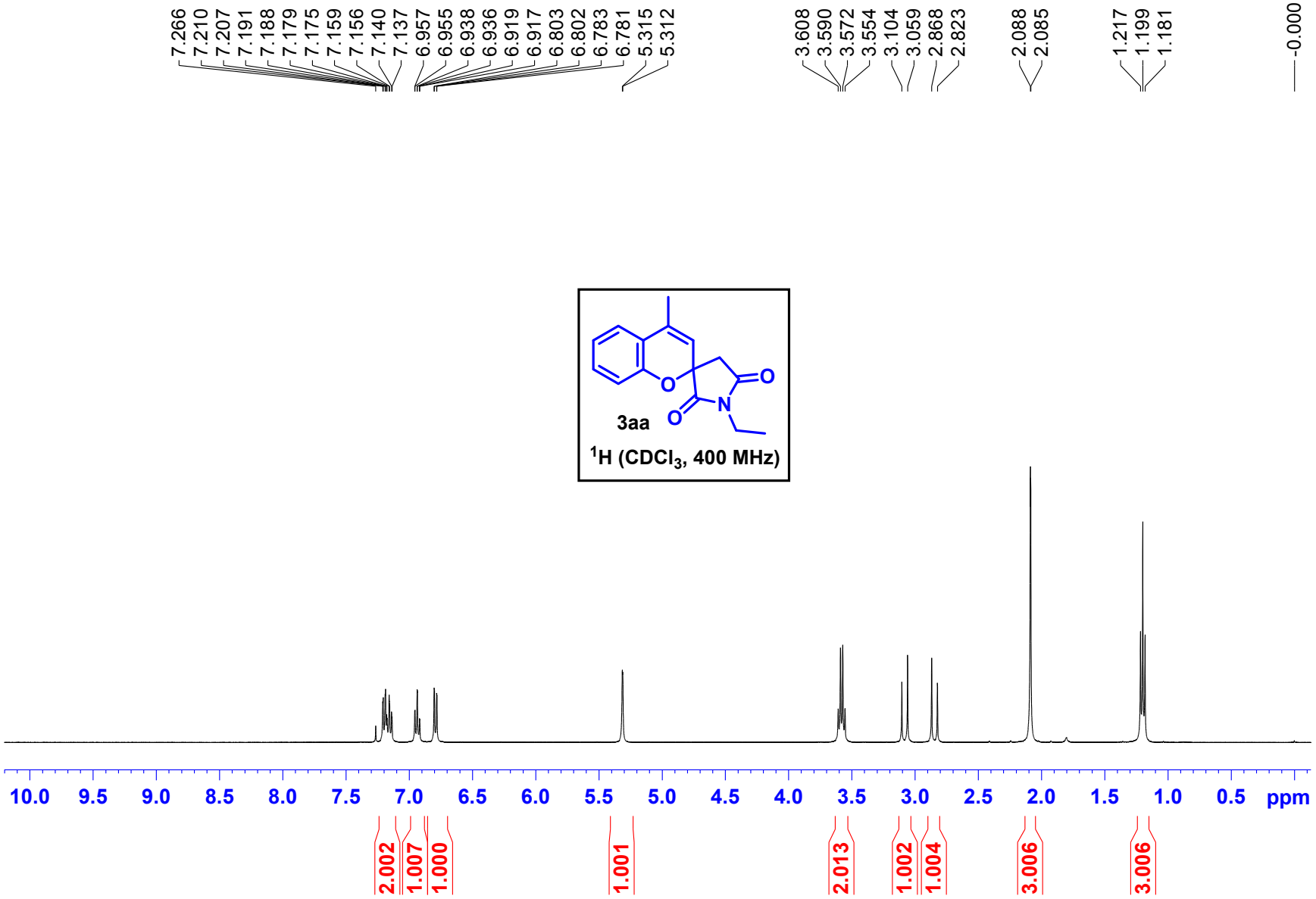
Prepared as shown in *general experimental procedure A*. **Yield** = 54% (41.5 mg); **Appearance** – Brown Solid; **mp** = 79 - 81 °C; R_f = 0.35 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.94 (d, *J* = 2.0 Hz, 1H), 7.87 (dd, *J* = 8.4, 2.0 Hz, 1H), 6.83 (d, *J* = 8.4 Hz, 1H), 5.33 (s, 1H), 4.35 (q, *J* = 7.2 Hz, 2H), 3.58 (q, *J* = 7.2 Hz, 2H), 3.11 (d, *J* = 18.2 Hz, 1H), 2.89 (d, *J* = 18.2 Hz, 1H), 2.48 (td, *J* = 6.7, 3.1 Hz, 2H), 1.59 – 1.54 (m, 2H), 1.39 – 1.35 (m, 7H), 1.20 (t, *J* = 7.2 Hz, 3H), 0.91 (t, *J* = 7.2 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 173.7, 172.8, 166.1, 156.0, 137.3, 131.6, 125.5, 124.2, 120.5, 116.4, 115.8, 79.2, 60.8, 44.4, 34.1, 31.5, 31.0, 27.1, 22.4, 14.4, 14.0, 12.9; **FT-IR (cm^{-1})** 2958, 2929, 2861, 1786, 1715, 1651, 1607, 1400, 1260, 1221, 1109, 1021; **HRMS (ESI-TOF) m/z** $[M + H]^+$ Calculated for $C_{22}H_{27}NO_5H$ 386.1967; found 386.1970.

(29) ***(Z)-2,11-Dimethyl-6H-dibenzo[c,g][1,2]thiazocine 5,5-dioxide (6b).***



Prepared as shown in *general experimental procedure A*. **Yield** = 31% (17.7 mg); **Appearance** – Yellow Solid; **mp** = 191 - 193 °C; R_f = 0.45 (20% EtOAc /Hexane); **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.83 (d, *J* = 8.2 Hz, 1H), 7.53 (d, *J* = 7.4 Hz, 1H), 7.27 – 7.23 (m, 2H), 7.20 (td, *J* = 7.5, 1.5 Hz, 1H), 7.15 (dd, *J* = 7.5, 1.5 Hz, 1H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.94 (s, 1H), 6.82 (s, 1H), 6.33 (s, 1H), 2.29 (s, 3H), 2.27 (d, *J* = 1.5 Hz, 3H); **^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$)** δ 142.6, 138.2, 137.5, 134.6, 134.1, 133.5, 131.2, 129.1, 128.9, 128.6, 127.8, 127.2, 127.0, 126.7, 25.3, 21.2; **^{13}C{^{1}H}/DEPT-135 NMR (100 MHz, $CDCl_3$)** δ 131.2, 129.1, 128.9, 128.6, 127.8, 127.2, 127.0, 126.7, 25.3, 21.2; **FT-IR (cm^{-1})** 3236, 2918, 2374, 1647, 1455, 1486, 1399, 1318, 1154, 1064; **HRMS (ESI-TOF) m/z** $[M + Na]^+$ Calculated for $C_{16}H_{15}NO_2Na$ 308.0721; found 308.0724.

^{1}H and ^{13}C NMR Spectra



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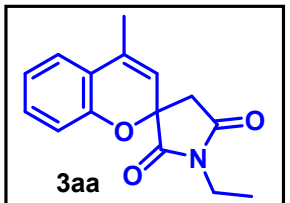
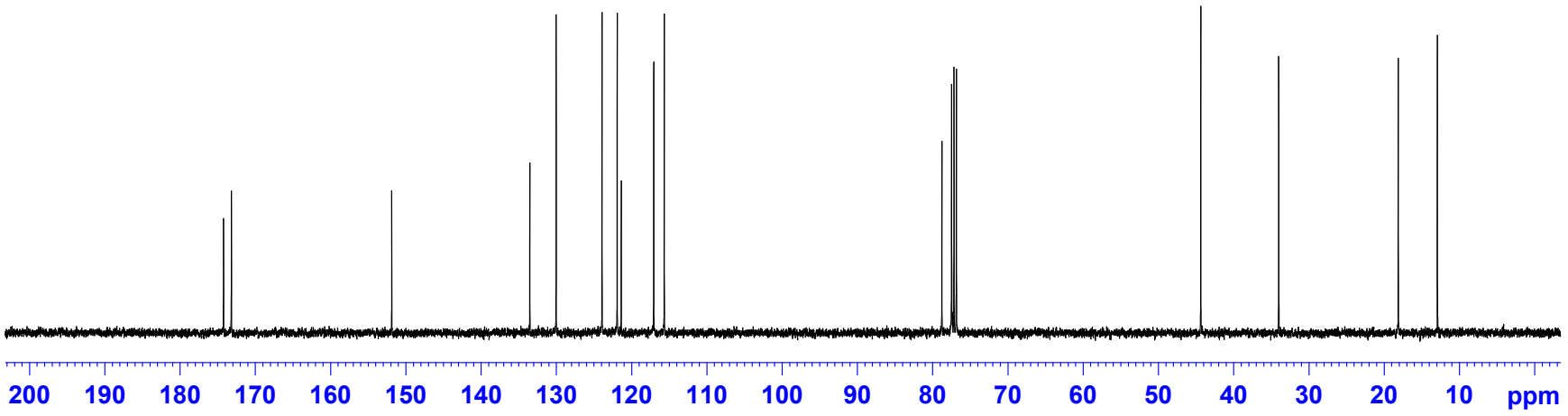
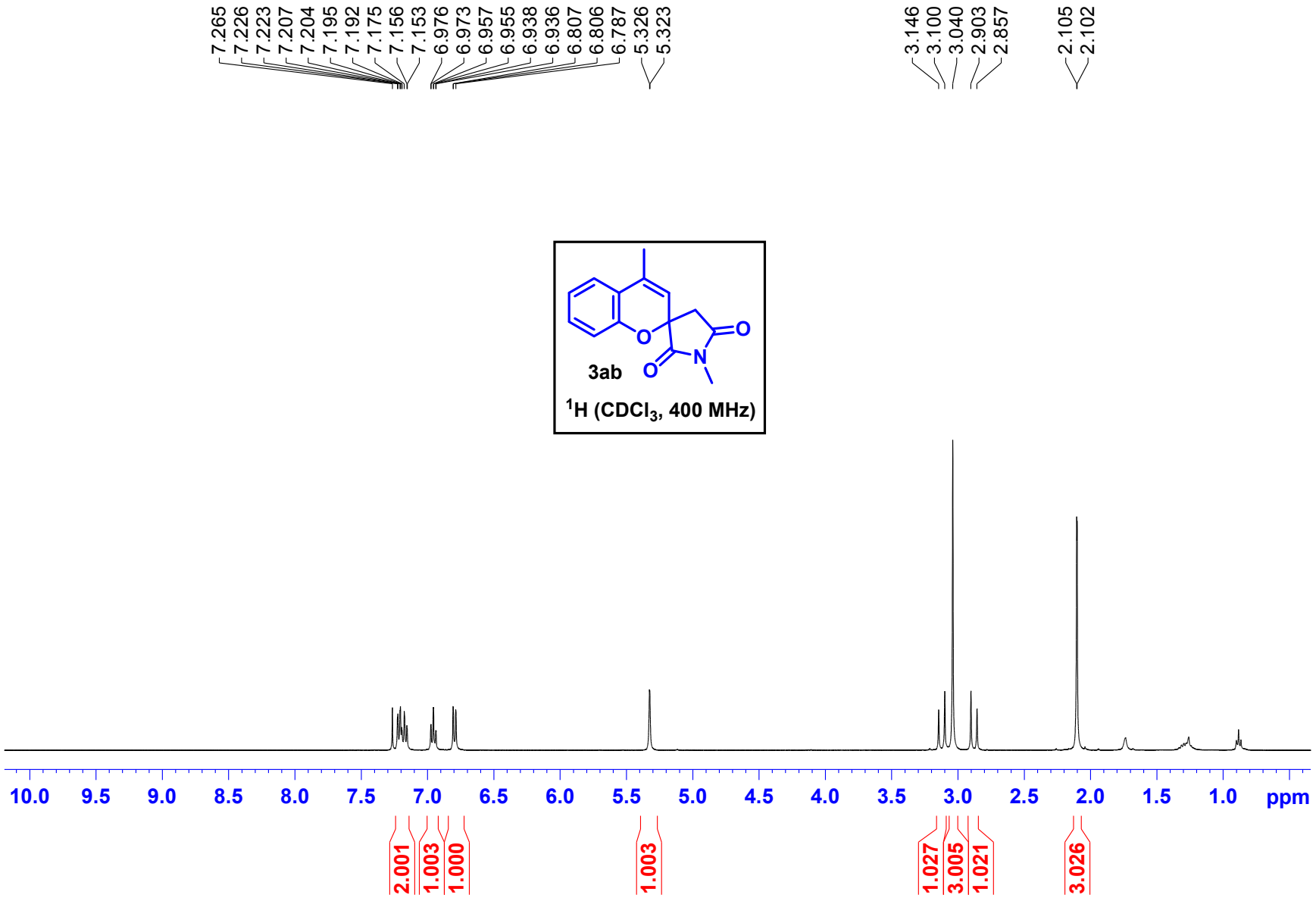


3aa

^{13}C ($CDCl_3$, 100 MHz)

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7.192
7.175
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6.957
6.955
6.938
6.936
6.807
6.806
6.787
5.326
5.323
3.146
3.100
3.040
2.903
2.857
2.105
2.102
O
O
N
O
3ab
^{1}H (CDCl$_3$, 400 MHz)
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9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
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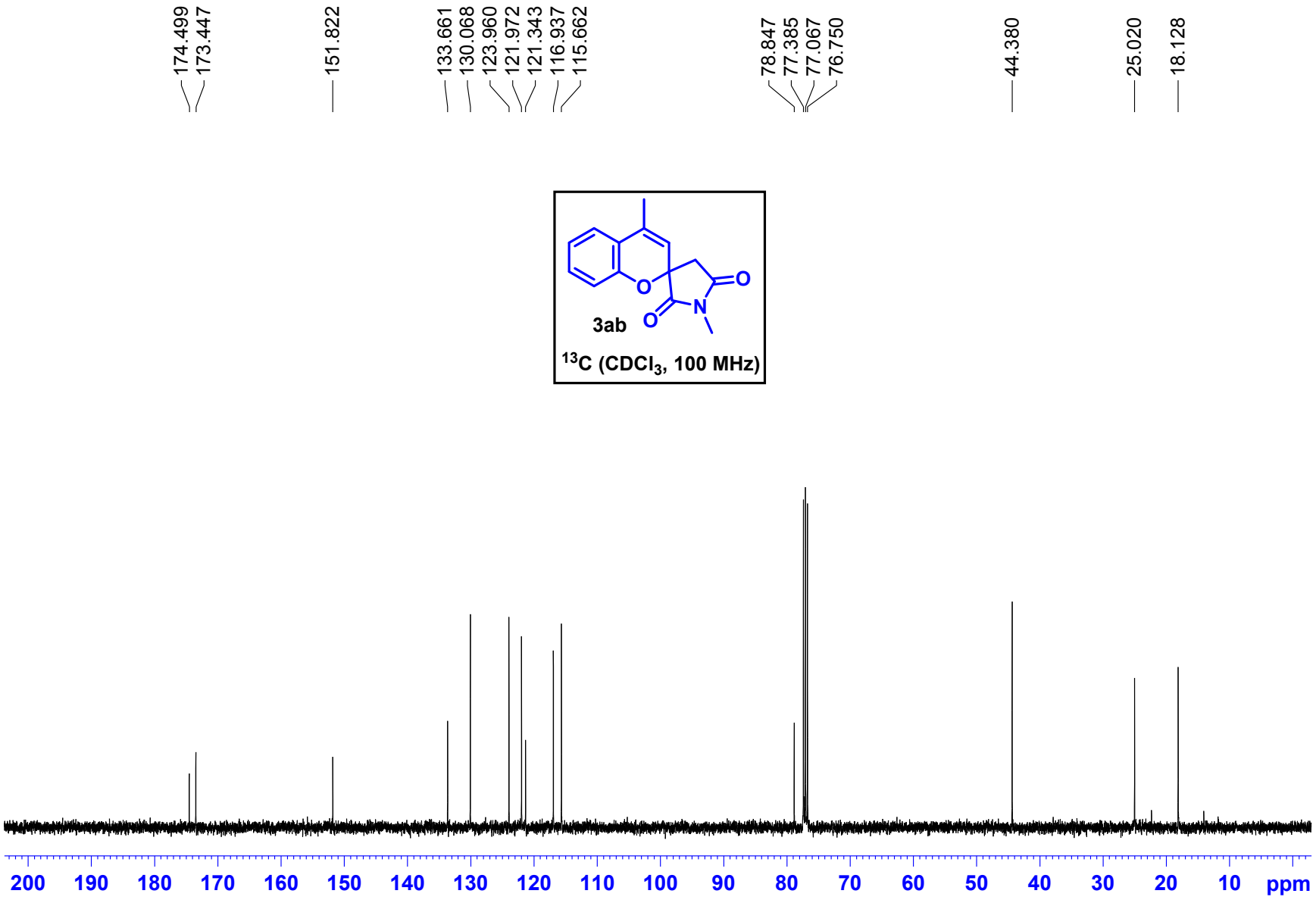
3.026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121.972
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77.067
76.750
44.380
25.020
18.128
O
O
O
N
3ab
13C (CDCl3, 100 MHz)
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70
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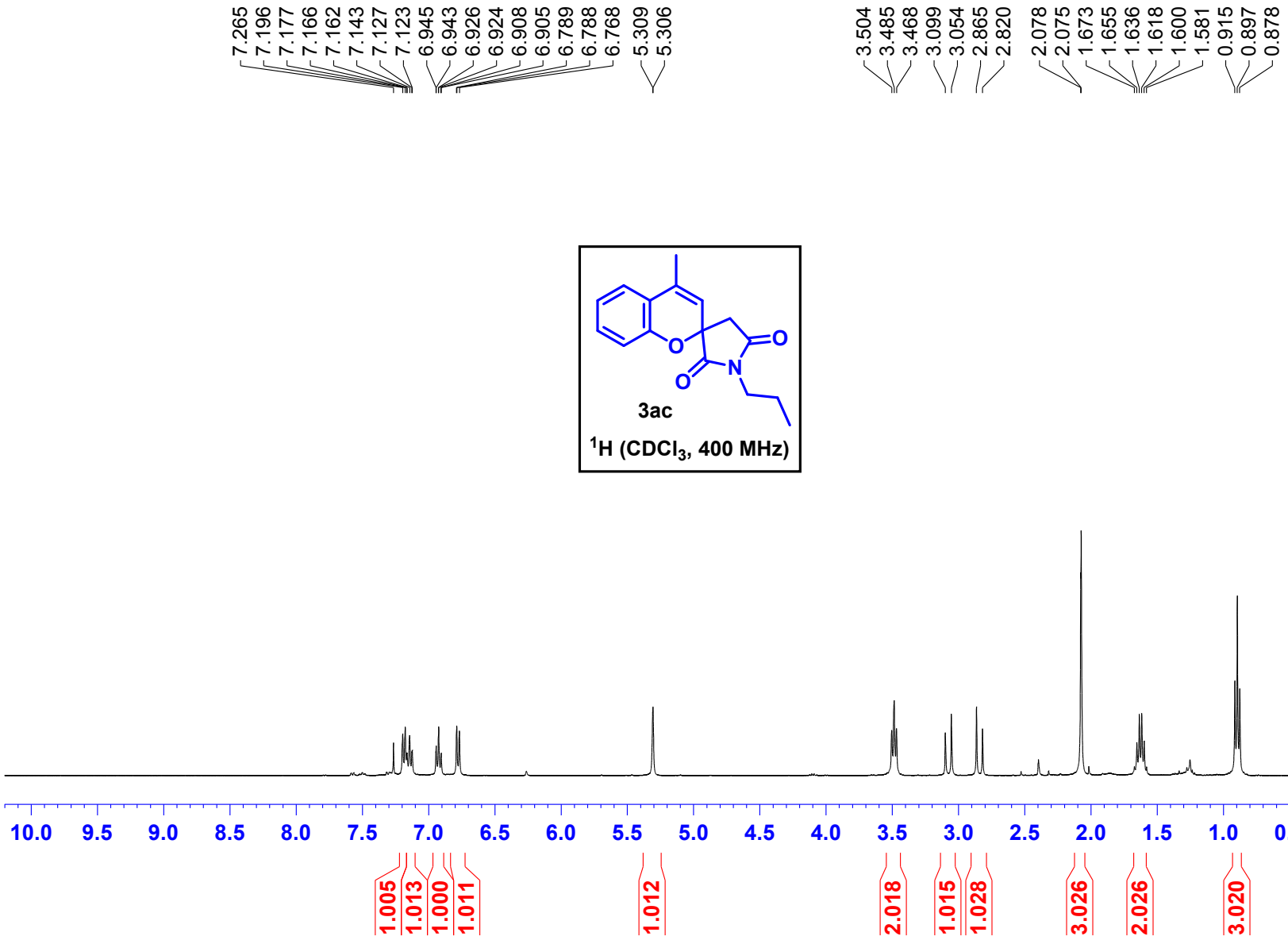
10
ppm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7.123
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6.943
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6.905
6.789
6.788
6.768
5.309
5.306
3.504
3.485
3.468
3.099
3.054
2.865
2.820
2.078
2.075
1.673
1.655
1.636
1.618
1.600
1.581
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0.878
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O
O
N
3ac
1H (CDCl3, 400 MHz)
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9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
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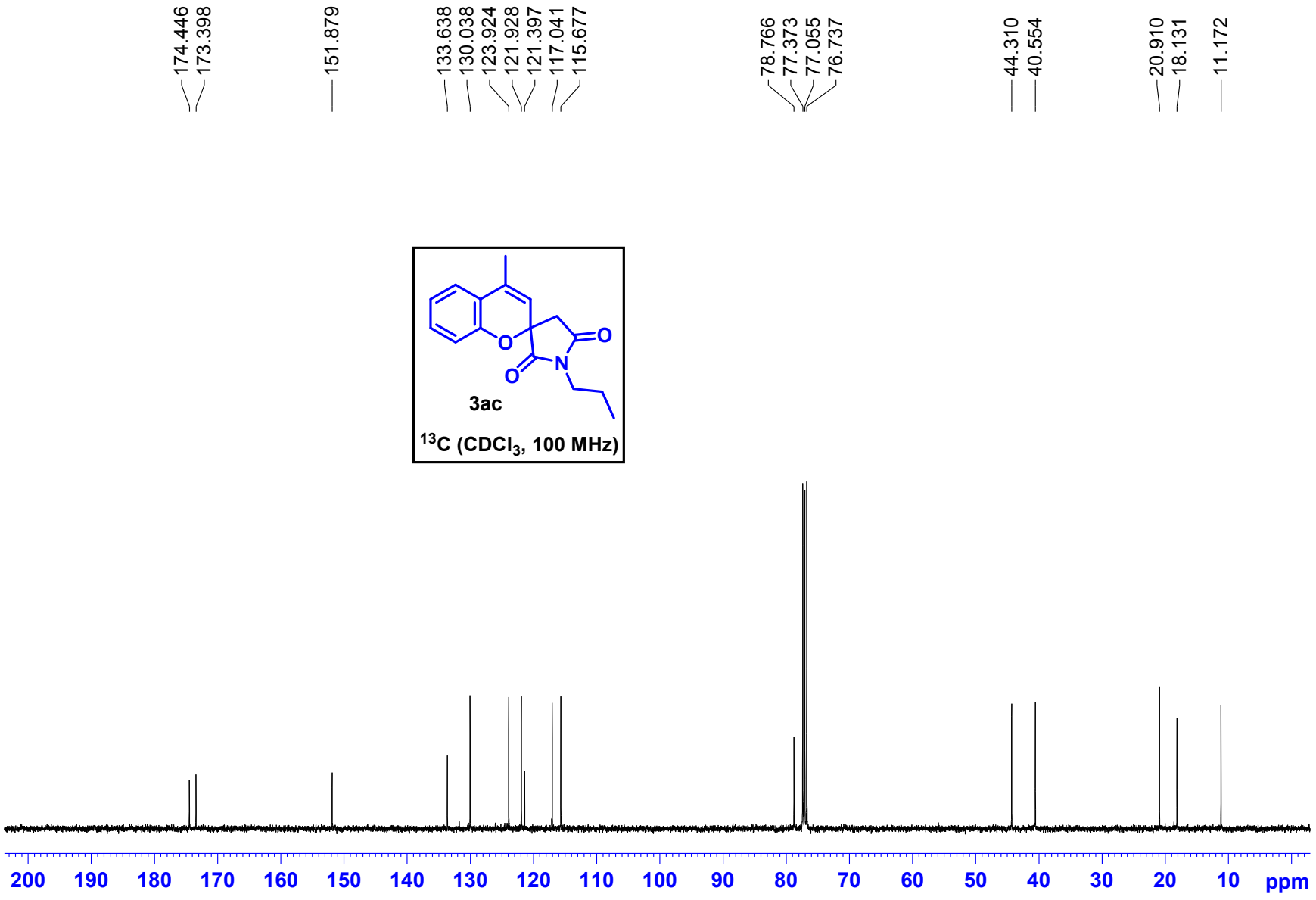
3.020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76.737
44.310
40.554
20.910
18.131
11.172
O
O
O
N
3ac
^{13}C (CDCl$_3$, 100 MHz)
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120
110
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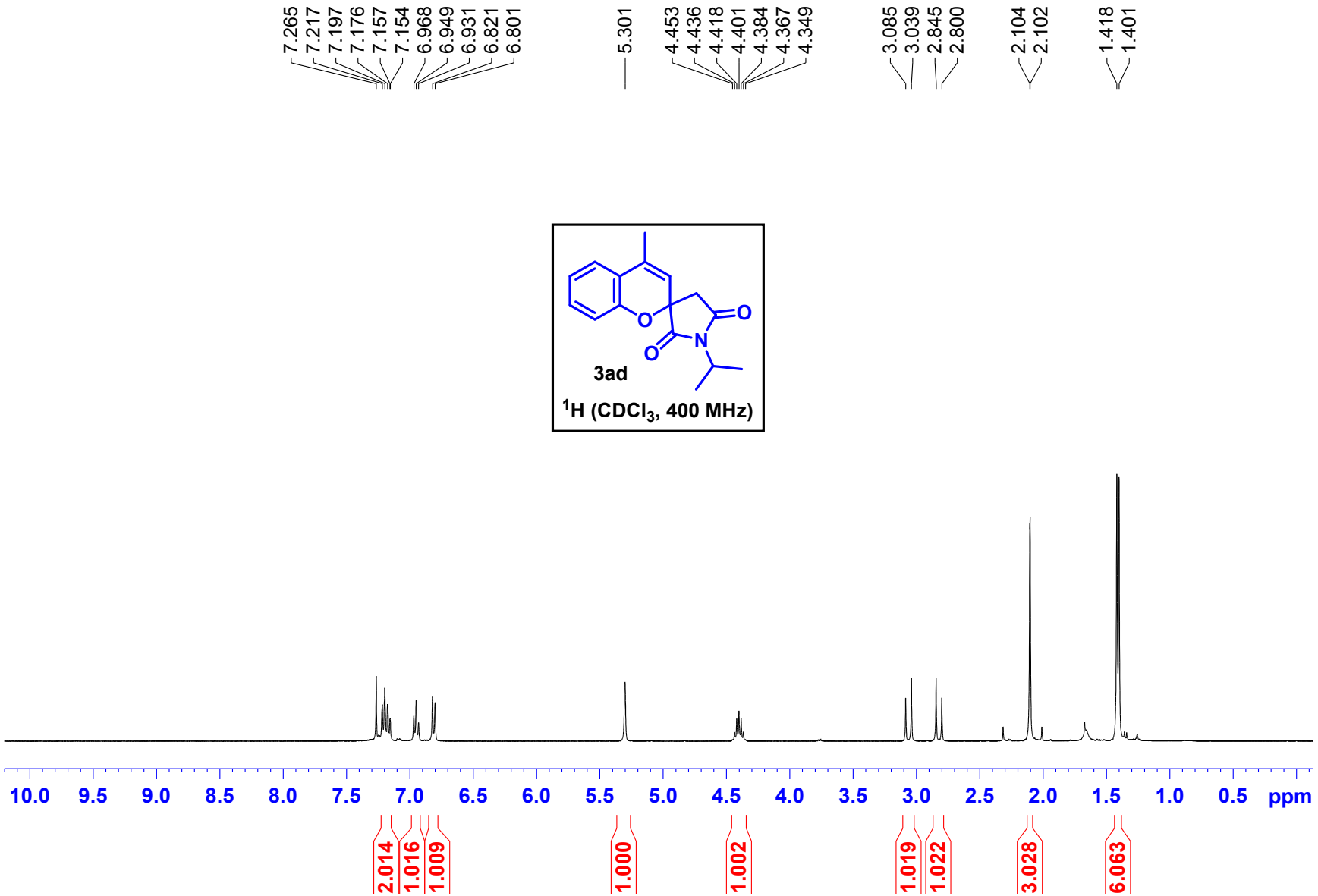
30
20
10
ppm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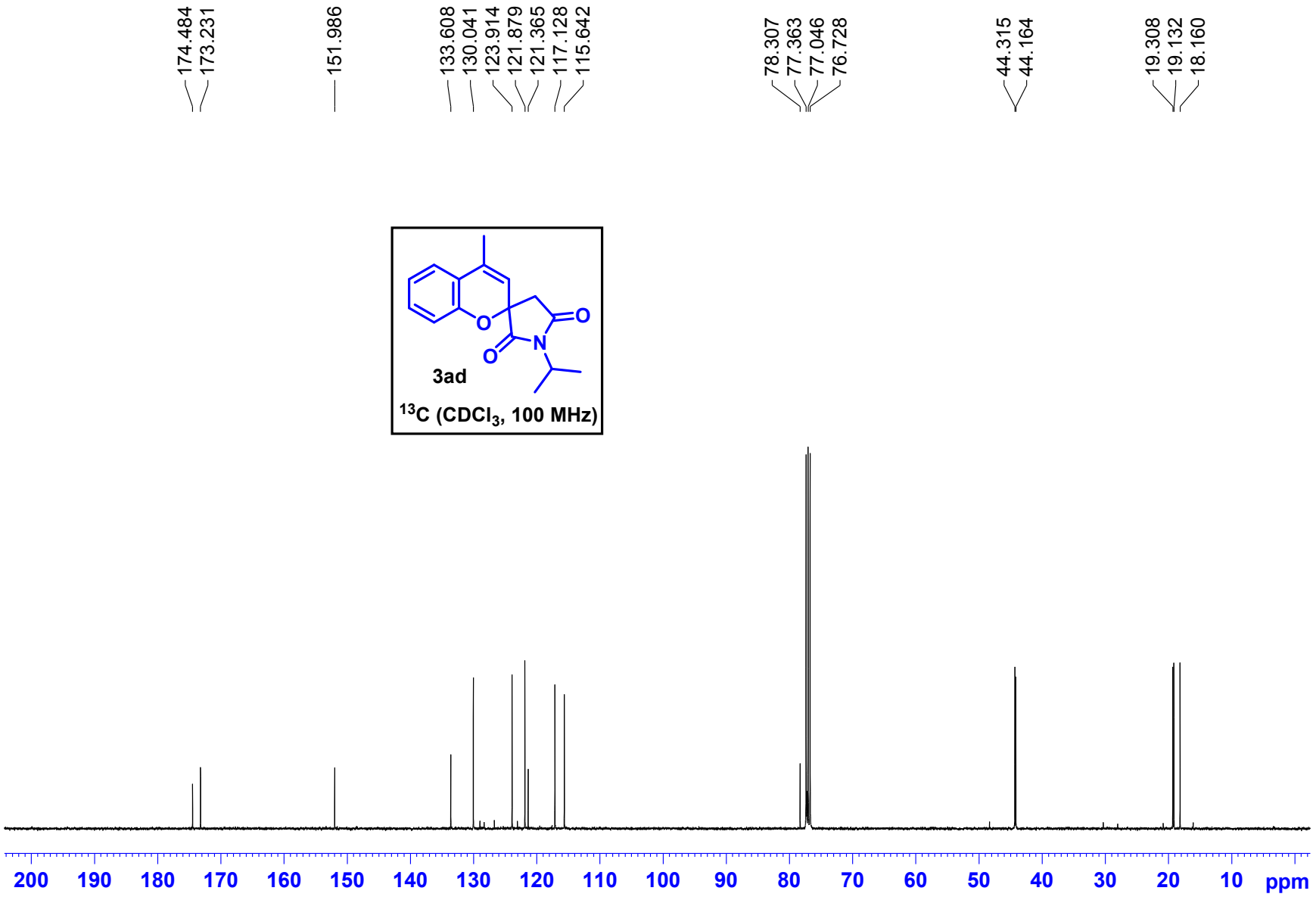
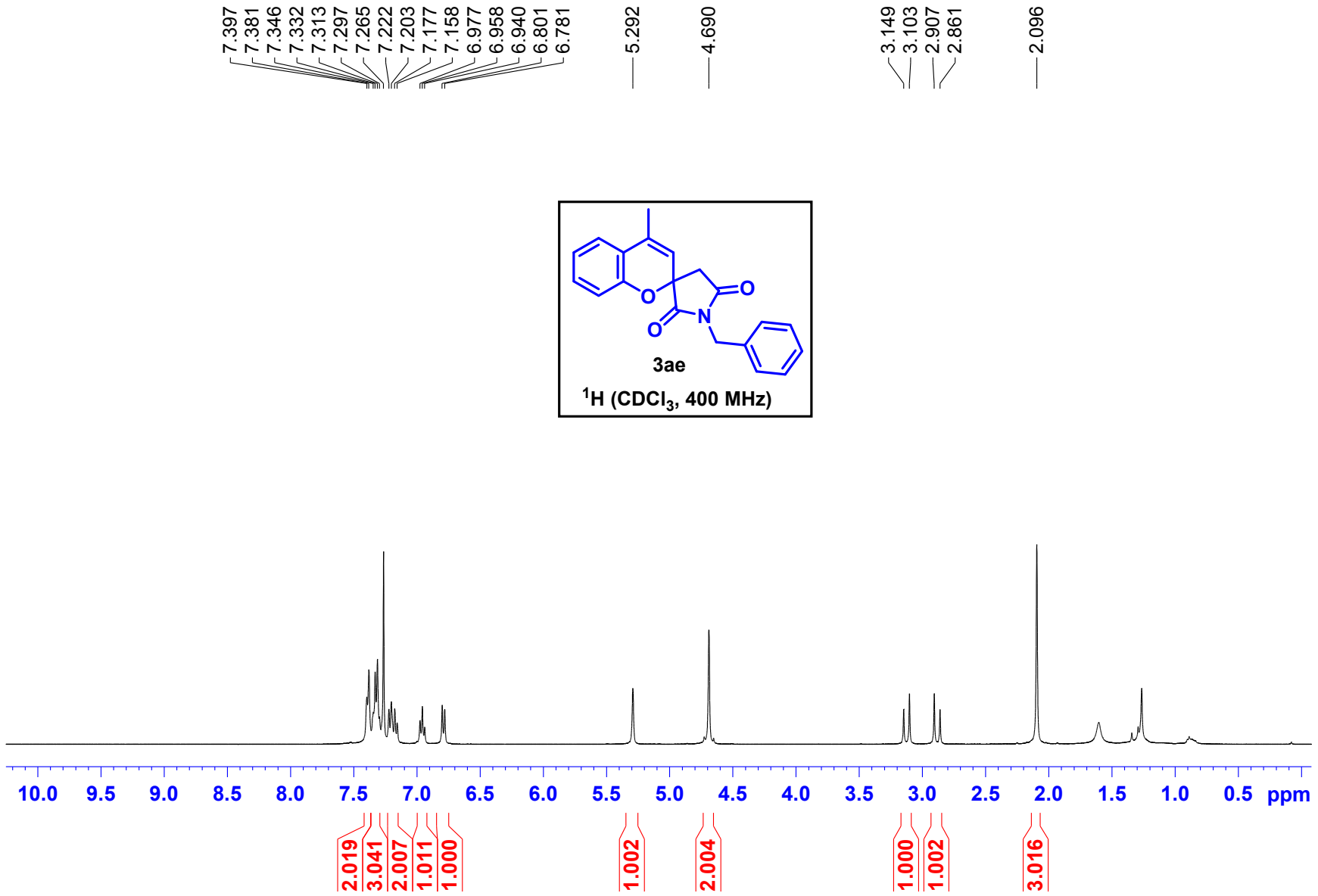
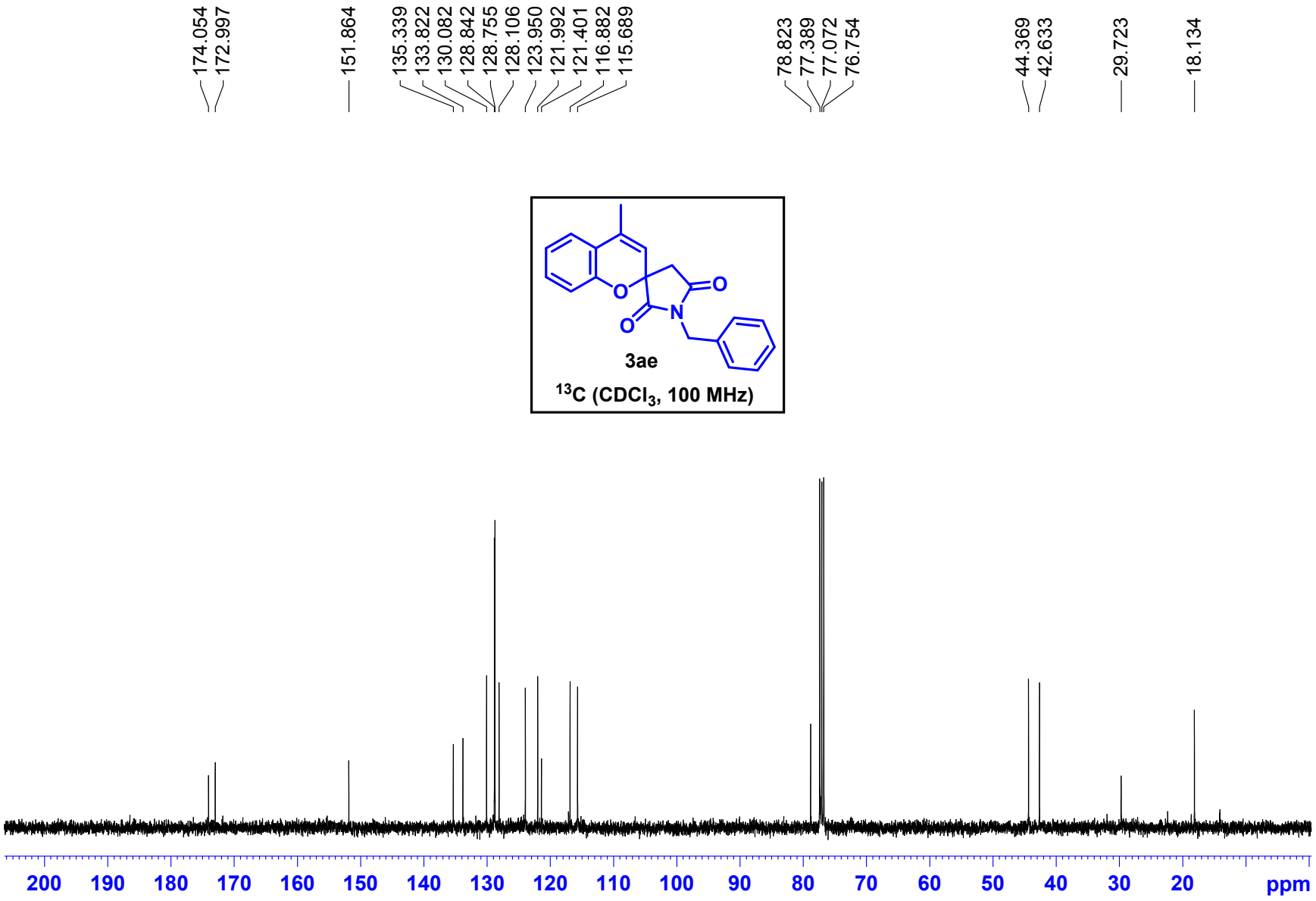
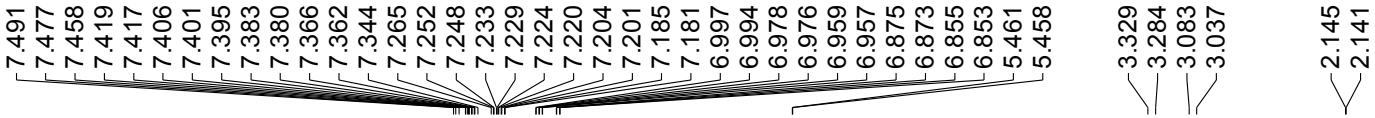
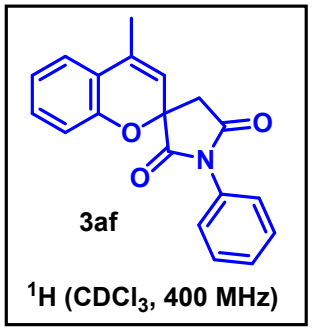
4.436
4.418
4.401
4.384
4.367
4.349
3.085
3.039
2.845
2.800
2.104
2.102
1.418
1.401
O
O
N
O
3ad
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
2.014
1.016
1.009
1.000
1.002
1.019
1.022
3.028
6.063


174.484
173.231
151.986
133.608
130.041
123.914
121.879
121.365
117.128
115.642
78.307
77.363
77.046
76.728
44.315
44.164
19.308
19.132
18.160
O
O
O
N
3ad
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.397
7.381
7.346
7.332
7.313
7.297
7.265
7.222
7.203
7.177
7.158
6.977
6.958
6.940
6.801
6.781
5.292
4.690
3.149
3.103
2.907
2.861
2.096
O
O
N
O
3ae
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
2.019
3.041
2.007
1.011
1.000
1.002
2.004
1.000
1.002
3.016

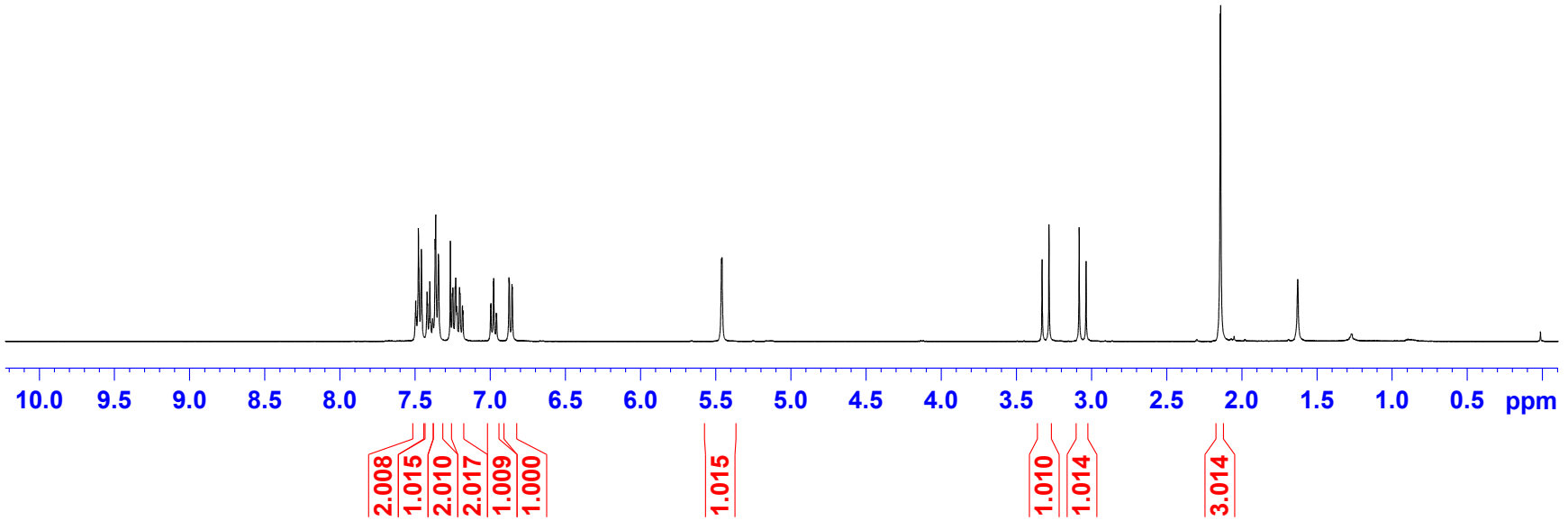

174.054
172.997
151.864
135.339
133.822
130.082
128.842
128.755
128.106
123.950
121.992
121.401
116.882
115.689
78.823
77.389
77.072
76.754
44.369
42.633
29.723
18.134
O
O
O
N
3ae
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
ppm

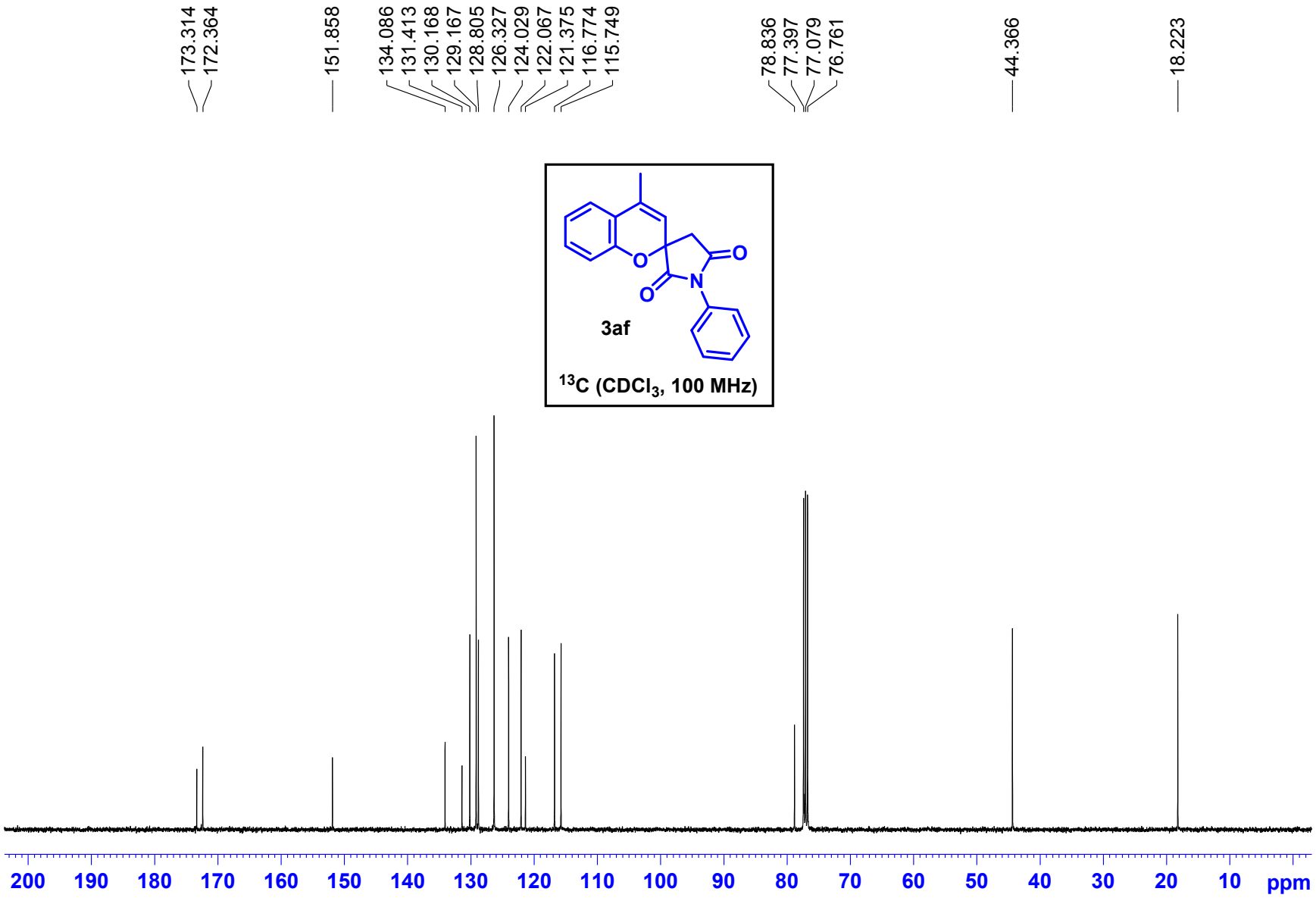


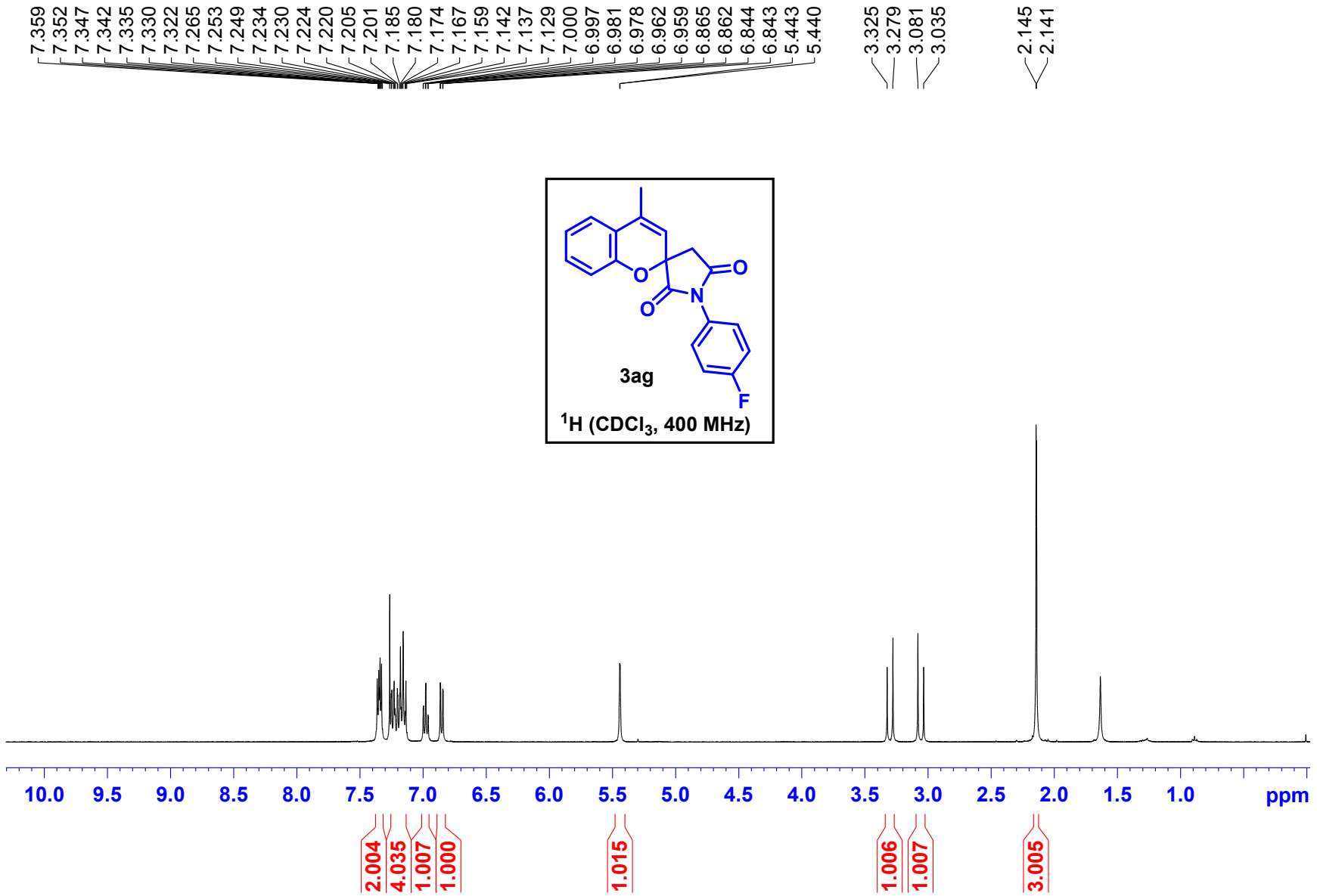


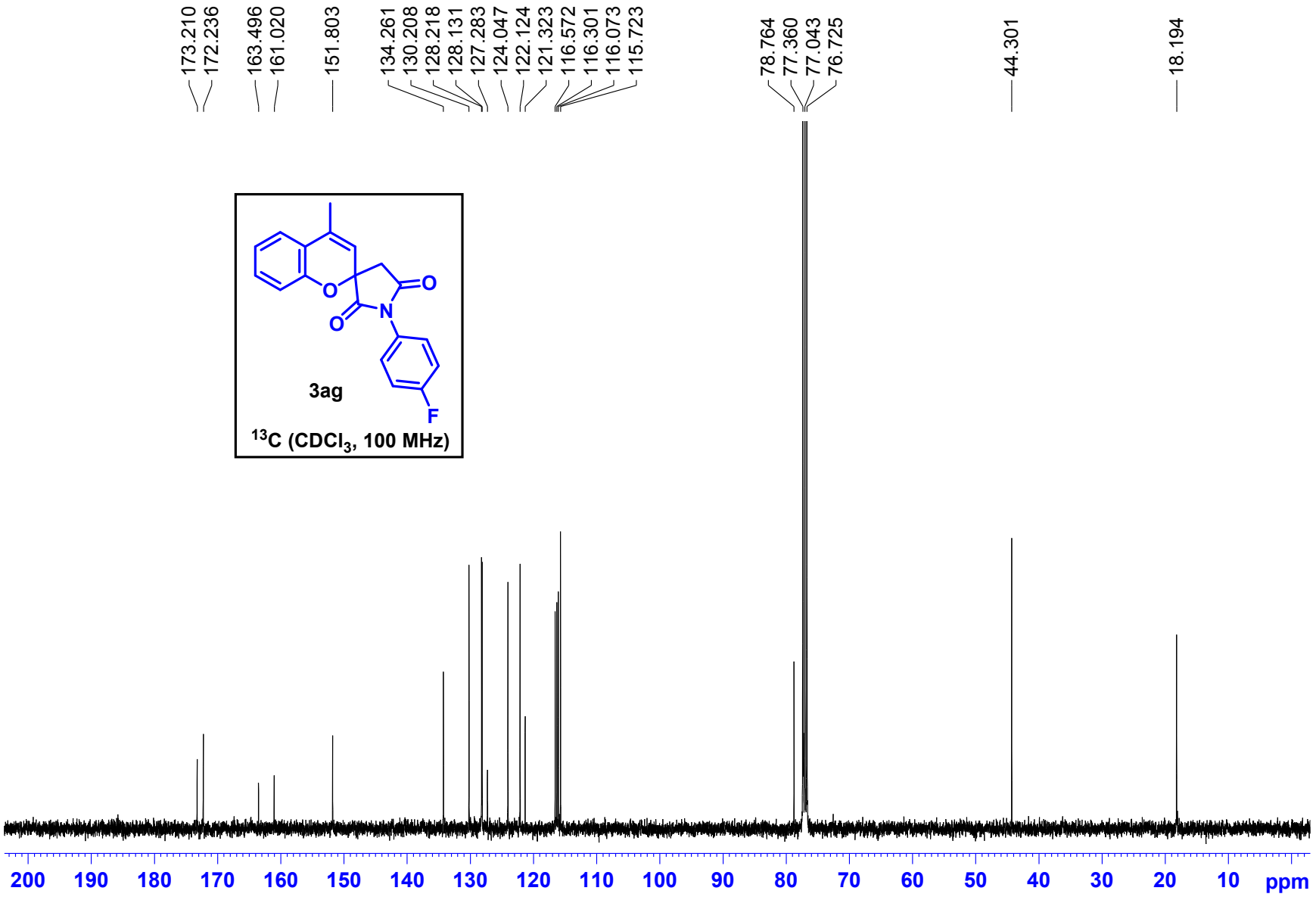
3af

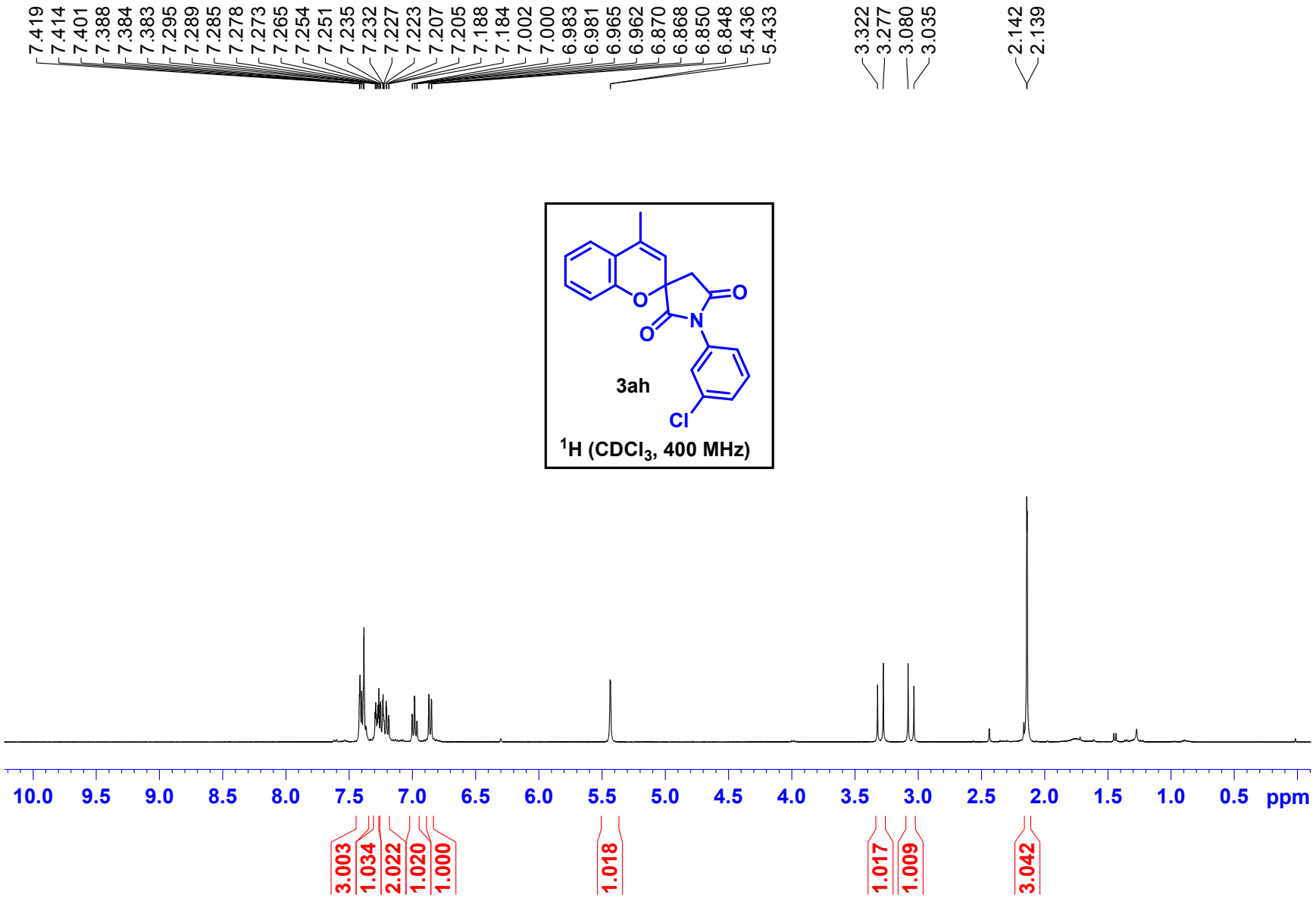
^{1}H ($CDCl_3$, 400 MHz)

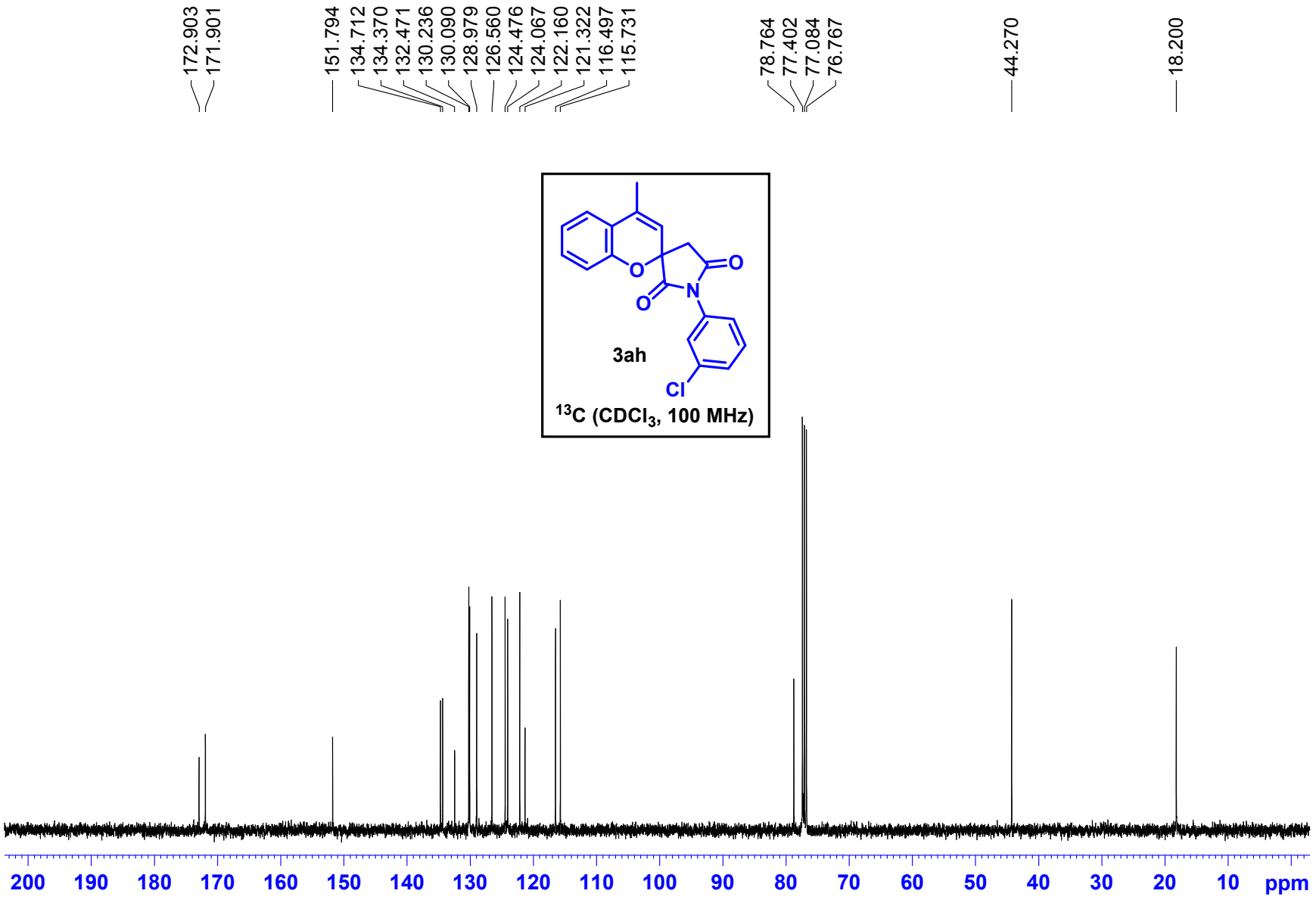


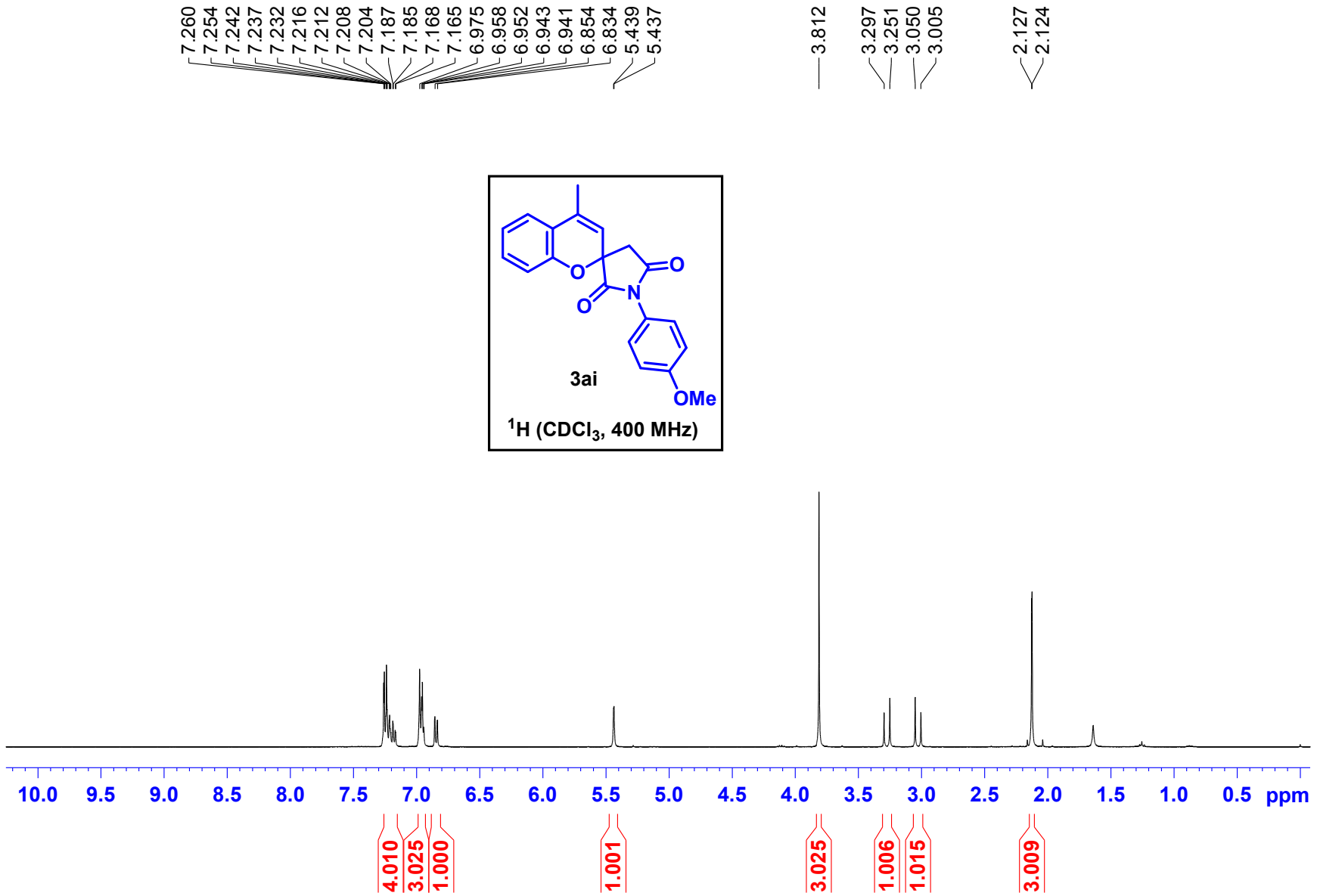

173.314
172.364
151.858
134.086
131.413
130.168
129.167
128.805
126.327
124.029
122.067
121.375
116.774
115.749
78.836
77.397
77.079
76.761
44.366
18.223
O
O
O
N
3af
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

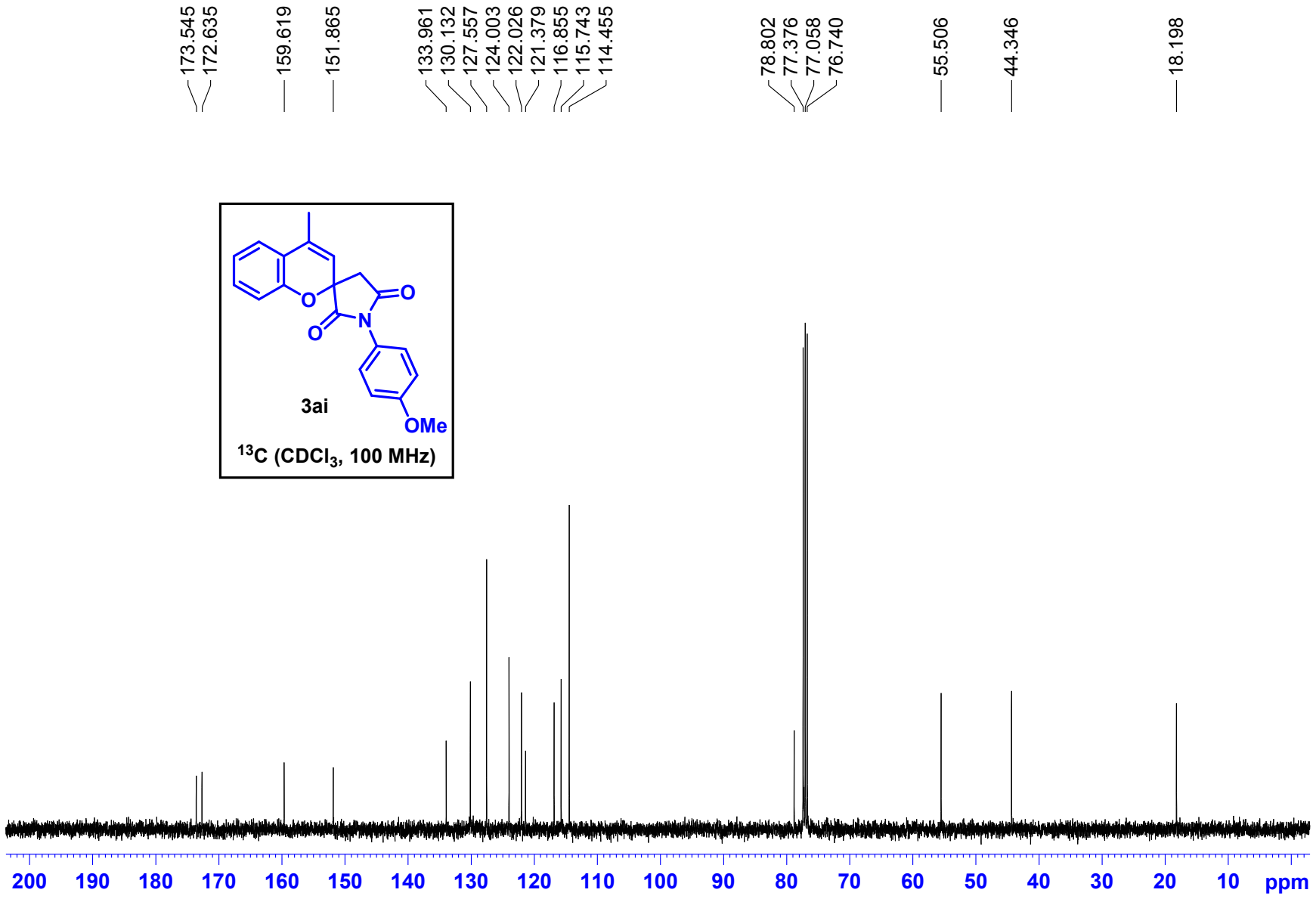

7.359
7.352
7.347
7.342
7.335
7.330
7.322
7.265
7.253
7.249
7.234
7.230
7.224
7.220
7.205
7.201
7.185
7.180
7.174
7.167
7.159
7.142
7.137
7.129
7.000
6.997
6.981
6.978
6.962
6.959
6.865
6.862
6.844
6.843
5.443
5.440
3.325
3.279
3.081
3.035
2.145
2.141
O
O
N
O
F
3ag
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
ppm
2.004
4.035
1.007
1.000
1.015
1.006
1.007
3.005

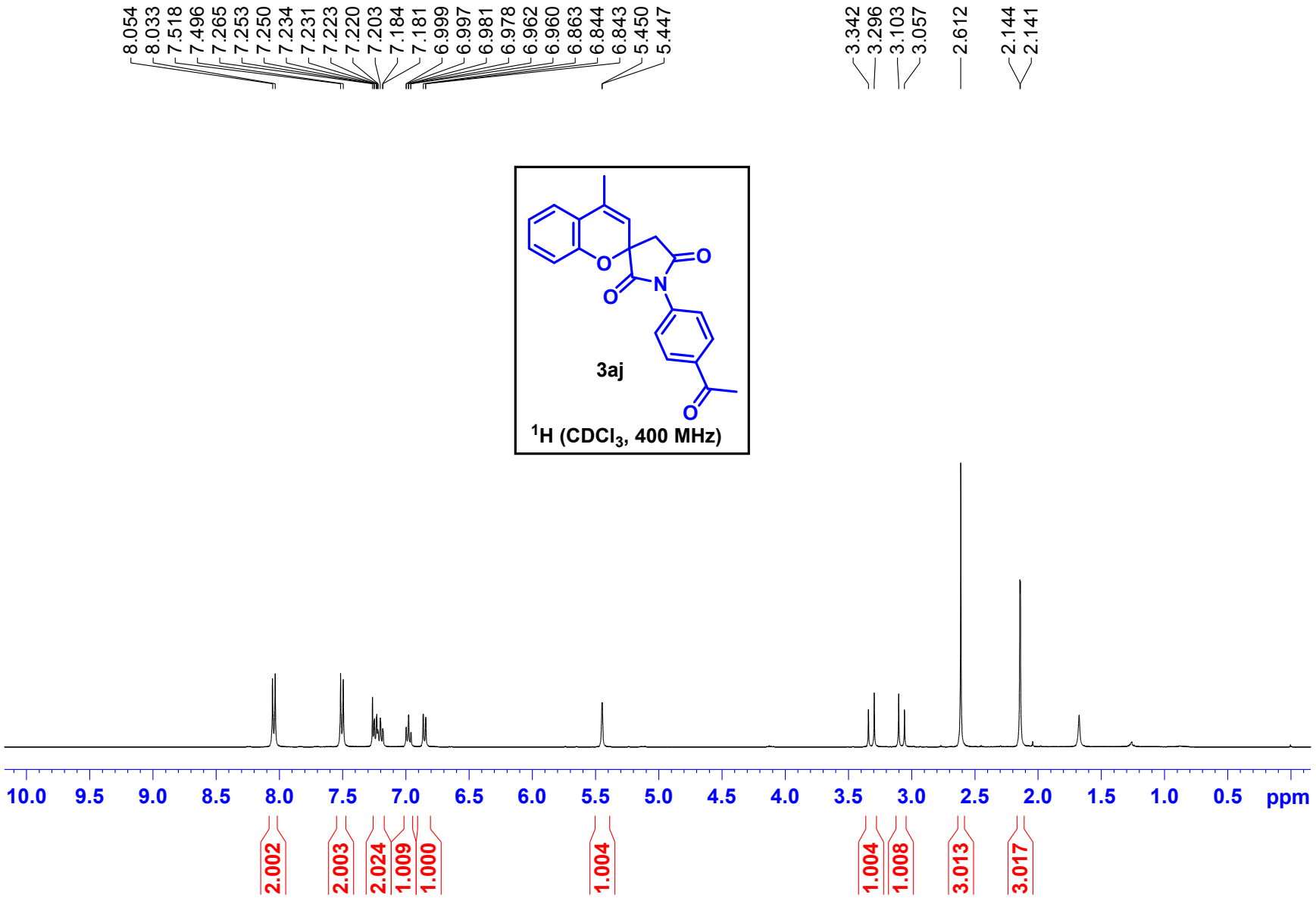

173.210
172.236
163.496
161.020
151.803
134.261
130.208
128.218
128.131
127.283
124.047
122.124
121.323
116.572
116.301
116.073
115.723
78.764
77.360
77.043
76.725
44.301
18.194
O
O
O
N
F
3ag
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

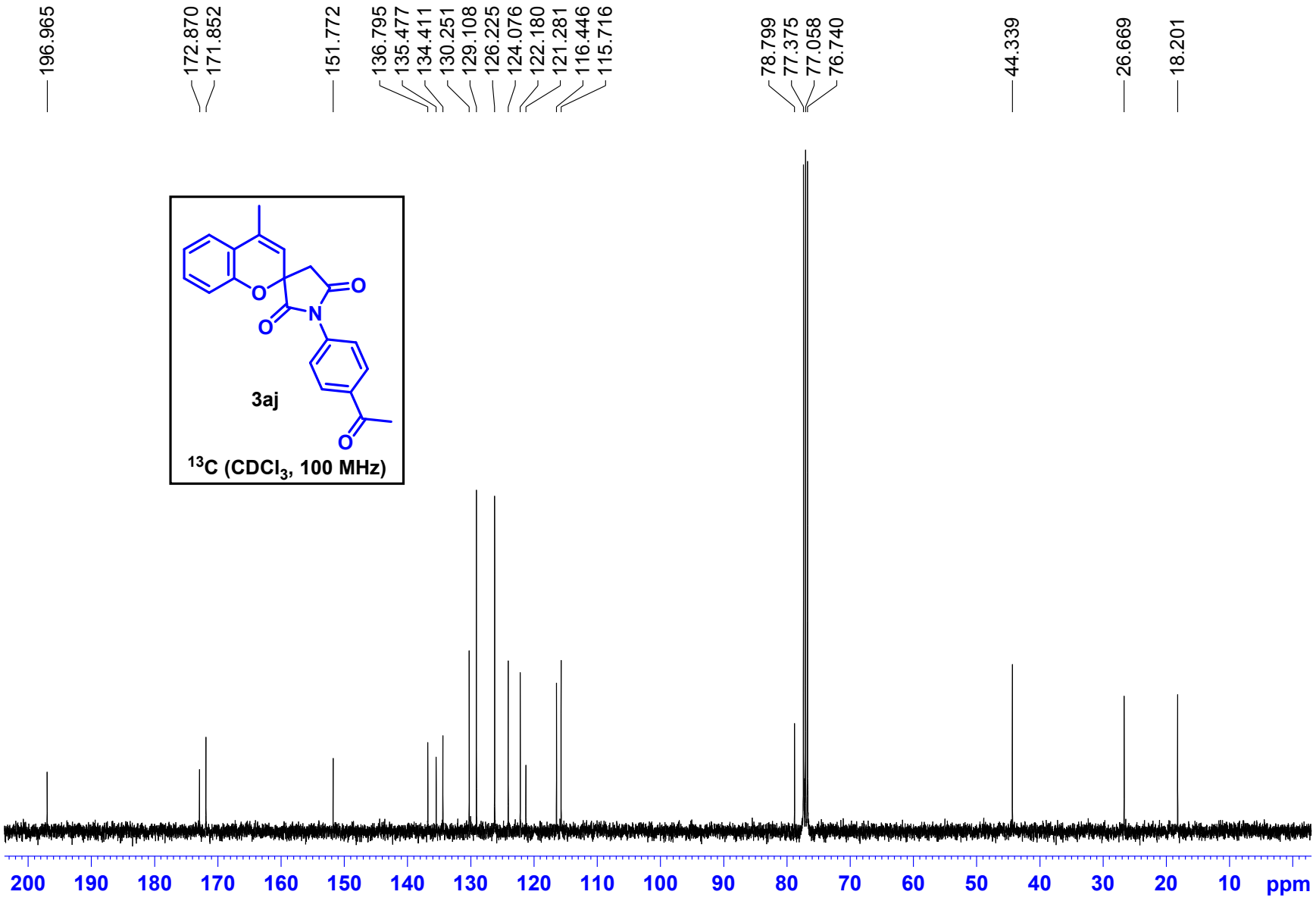

7.419
7.414
7.401
7.388
7.384
7.383
7.295
7.289
7.285
7.278
7.273
7.265
7.254
7.251
7.235
7.232
7.227
7.223
7.207
7.205
7.188
7.184
7.002
7.000
6.983
6.981
6.965
6.962
6.870
6.868
6.850
6.848
5.436
5.433
3.322
3.277
3.080
3.035
2.142
2.139
O
O
O
N
Cl
3ah
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
3.003
1.034
2.022
1.020
1.000
1.018
1.017
1.009
3.042

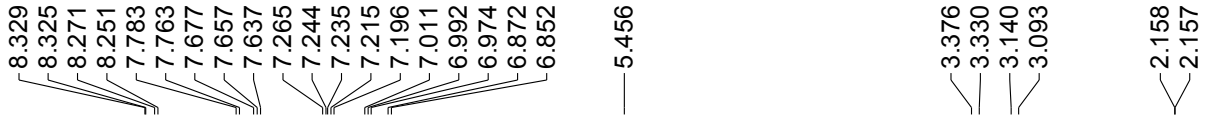

172.903
171.901
151.794
134.712
134.370
132.471
130.236
130.090
128.979
126.560
124.476
124.067
122.160
121.322
116.497
115.731
78.764
77.402
77.084
76.767
44.270
18.200
O
O
N
O
Cl
3ah
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

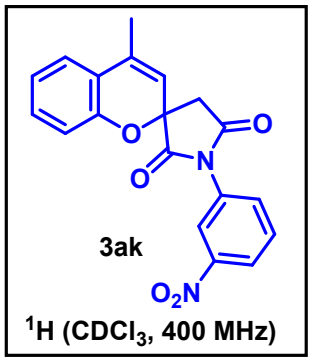

7.260
7.254
7.242
7.237
7.232
7.216
7.212
7.208
7.204
7.187
7.185
7.168
7.165
6.975
6.958
6.952
6.943
6.941
6.854
6.834
5.439
5.437
3.812
3.297
3.251
3.050
3.005
2.127
2.124
O
O
N
O
OMe
3ai
^{1}H (CDCl$_3$, 400 MHz)
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 ppm
4.010
3.025
1.000
1.001
3.025
1.006
1.015
3.009

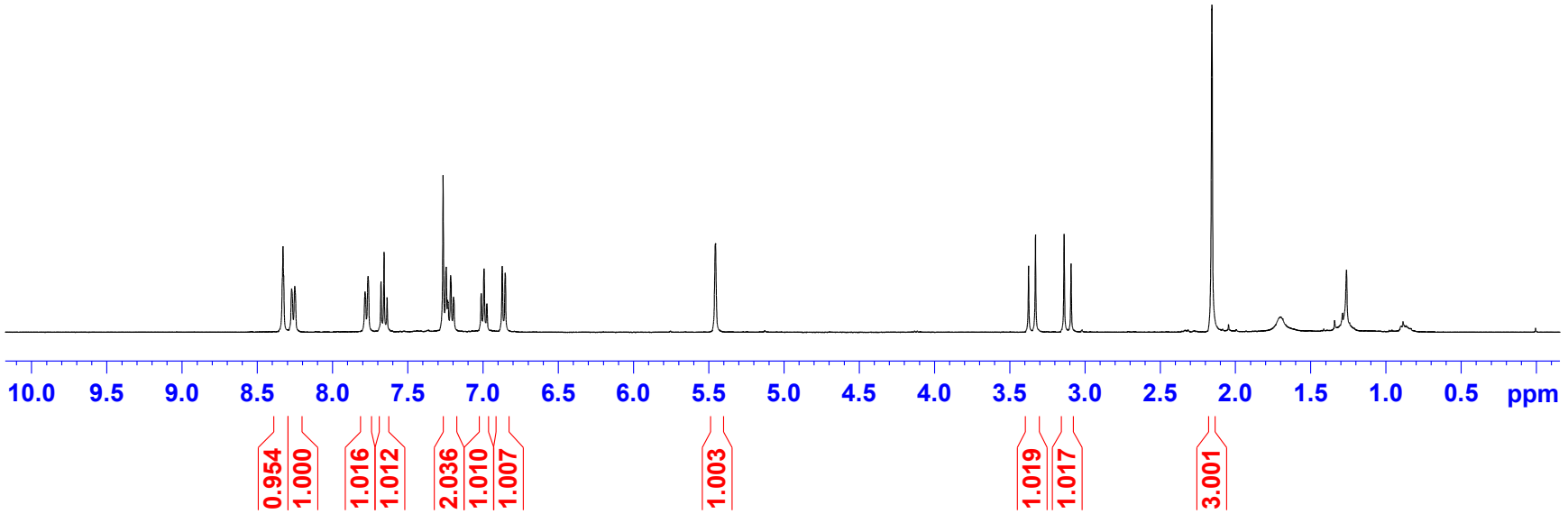

173.545
172.635
159.619
151.865
133.961
130.132
127.557
124.003
122.026
121.379
116.855
115.743
114.455
78.802
77.376
77.058
76.740
55.506
44.346
18.198
O
O
N
O
3ai
OMe
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

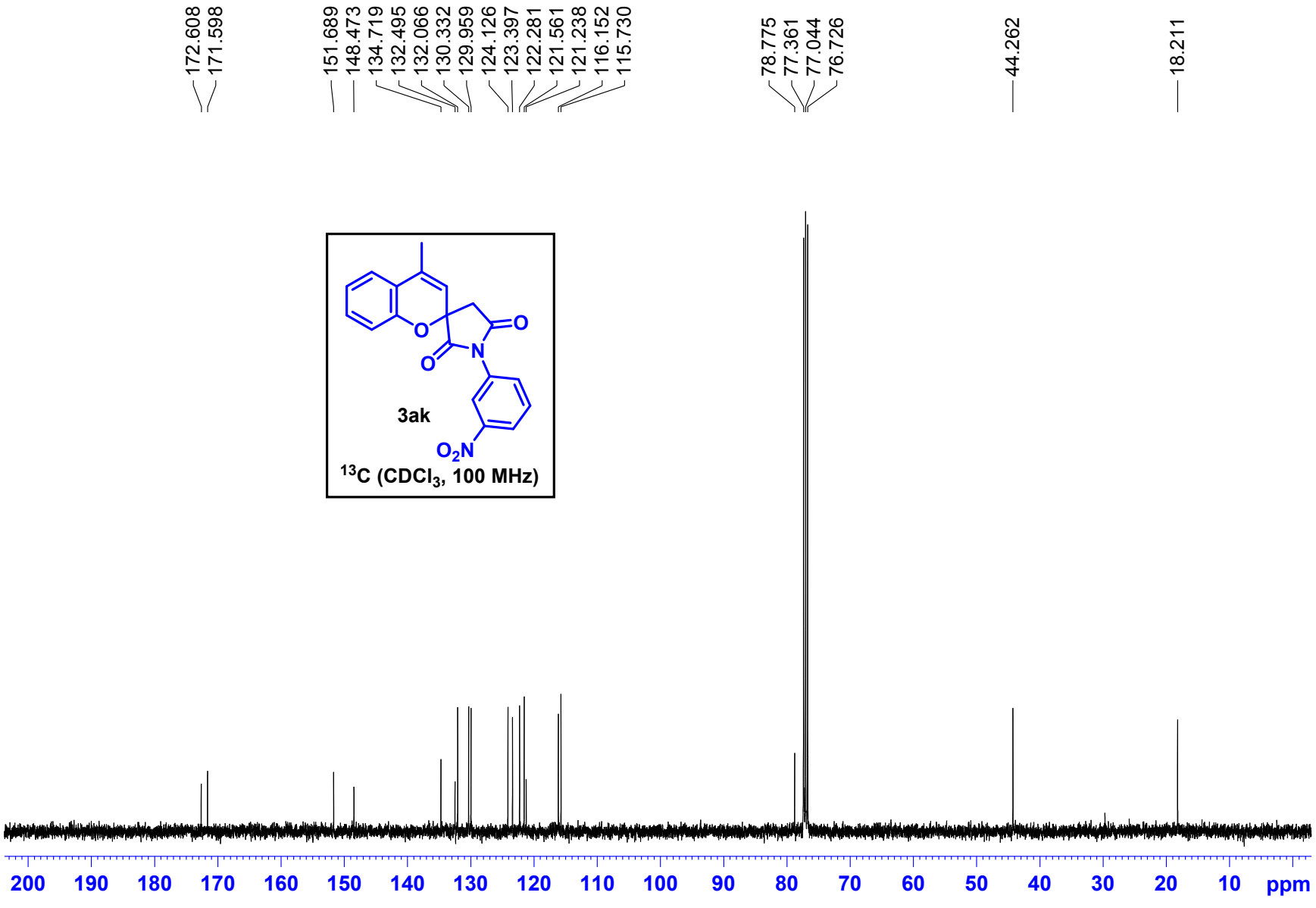

8.054
8.033
7.518
7.496
7.265
7.253
7.250
7.234
7.231
7.223
7.220
7.203
7.184
7.181
6.999
6.997
6.981
6.978
6.962
6.960
6.863
6.844
6.843
5.450
5.447
3.342
3.296
3.103
3.057
2.612
2.144
2.141
3aj
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
2.002
2.003
2.024
1.009
1.000
1.004
1.004
1.008
3.013
3.017

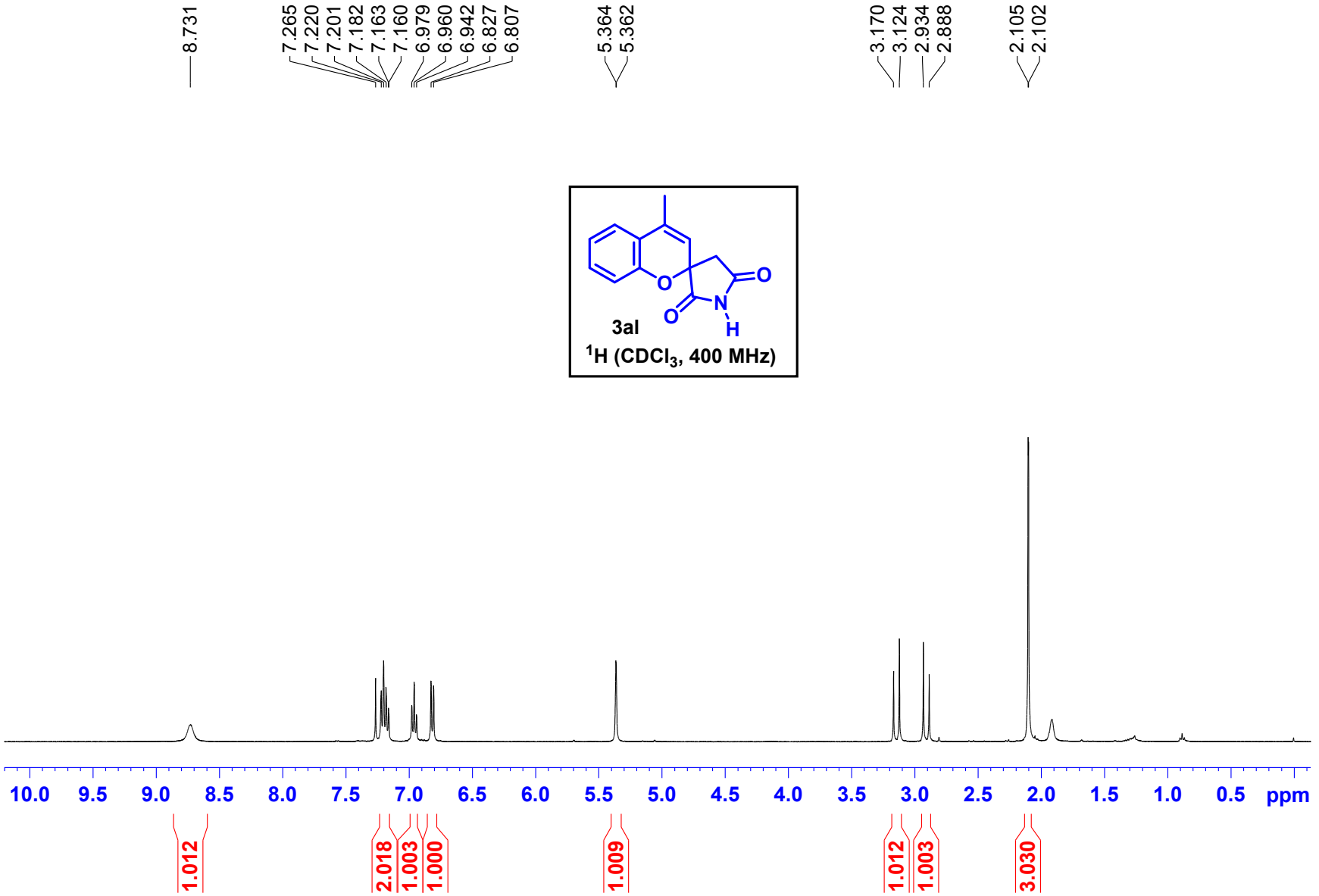

196.965
172.870
171.852
151.772
136.795
135.477
134.411
130.251
129.108
126.225
124.076
122.180
121.281
116.446
115.716
78.799
77.375
77.058
76.740
44.339
26.669
18.201
O
O
O
N
O
3aj
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

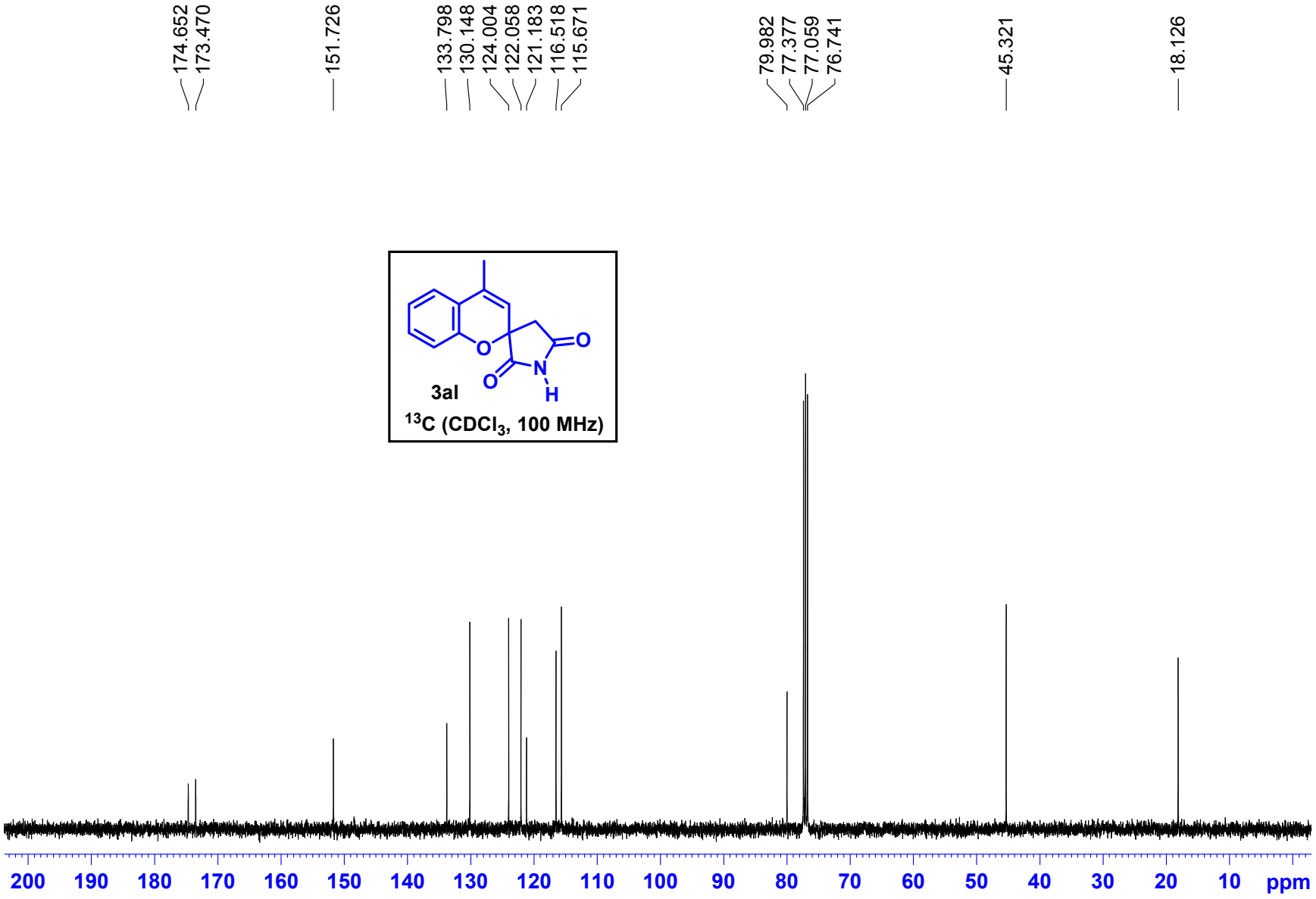

8.329
8.325
8.271
8.251
7.783
7.763
7.677
7.657
7.637
7.265
7.244
7.235
7.215
7.196
7.011
6.992
6.974
6.872
6.852
5.456
3.376
3.330
3.140
3.093
2.158
2.157

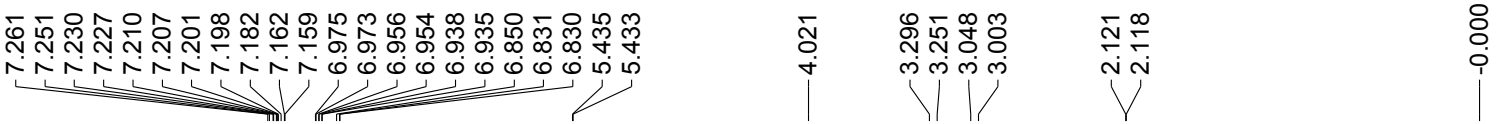

O
O
O
N
3ak
O_2N
^1H ($CDCl_3$, 400 MHz)

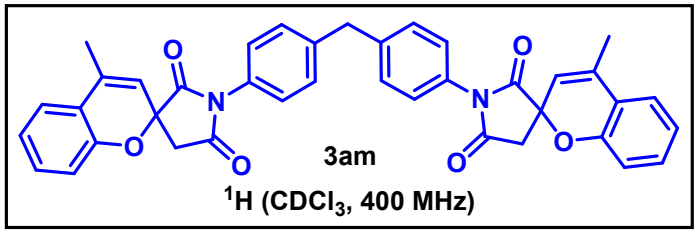

10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
0.954
1.000
1.016
1.012
2.036
1.010
1.007
1.003
1.019
1.017
3.001

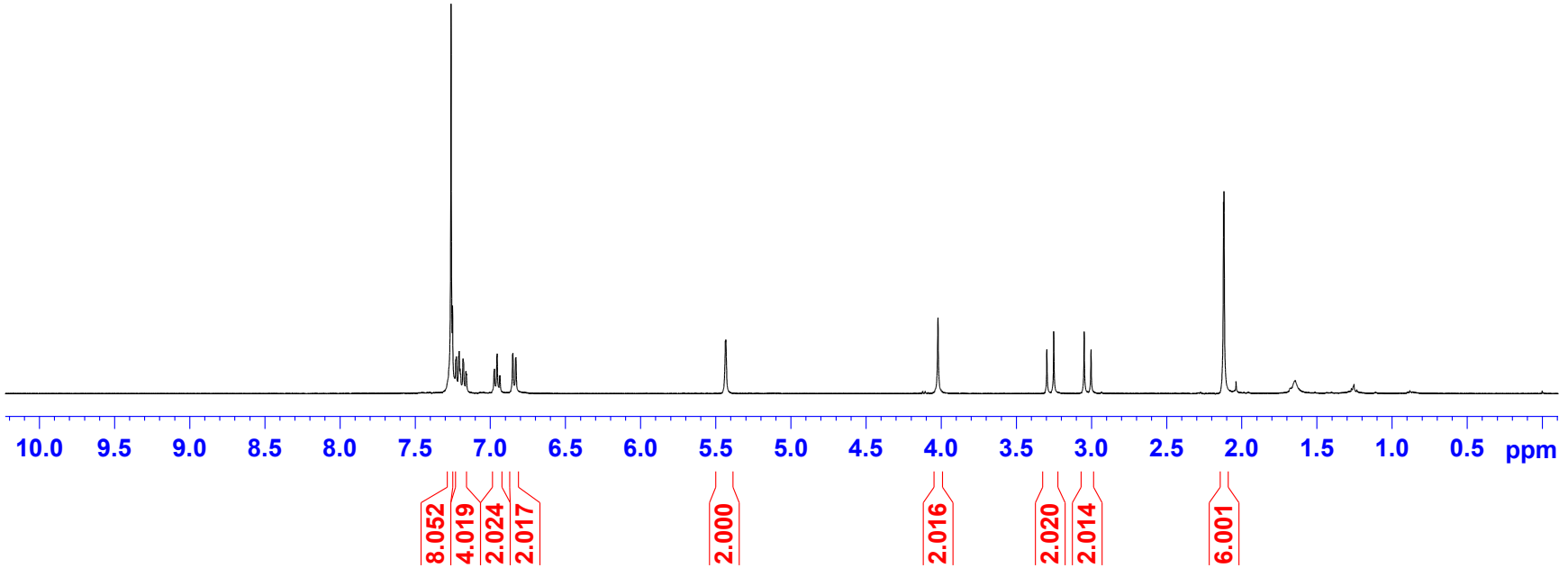

172.608
171.598
151.689
148.473
134.719
132.495
132.066
130.332
129.959
124.126
123.397
122.281
121.561
121.238
116.152
115.730
78.775
77.361
77.044
76.726
44.262
18.211
O
O
O
N
3ak
O2N
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

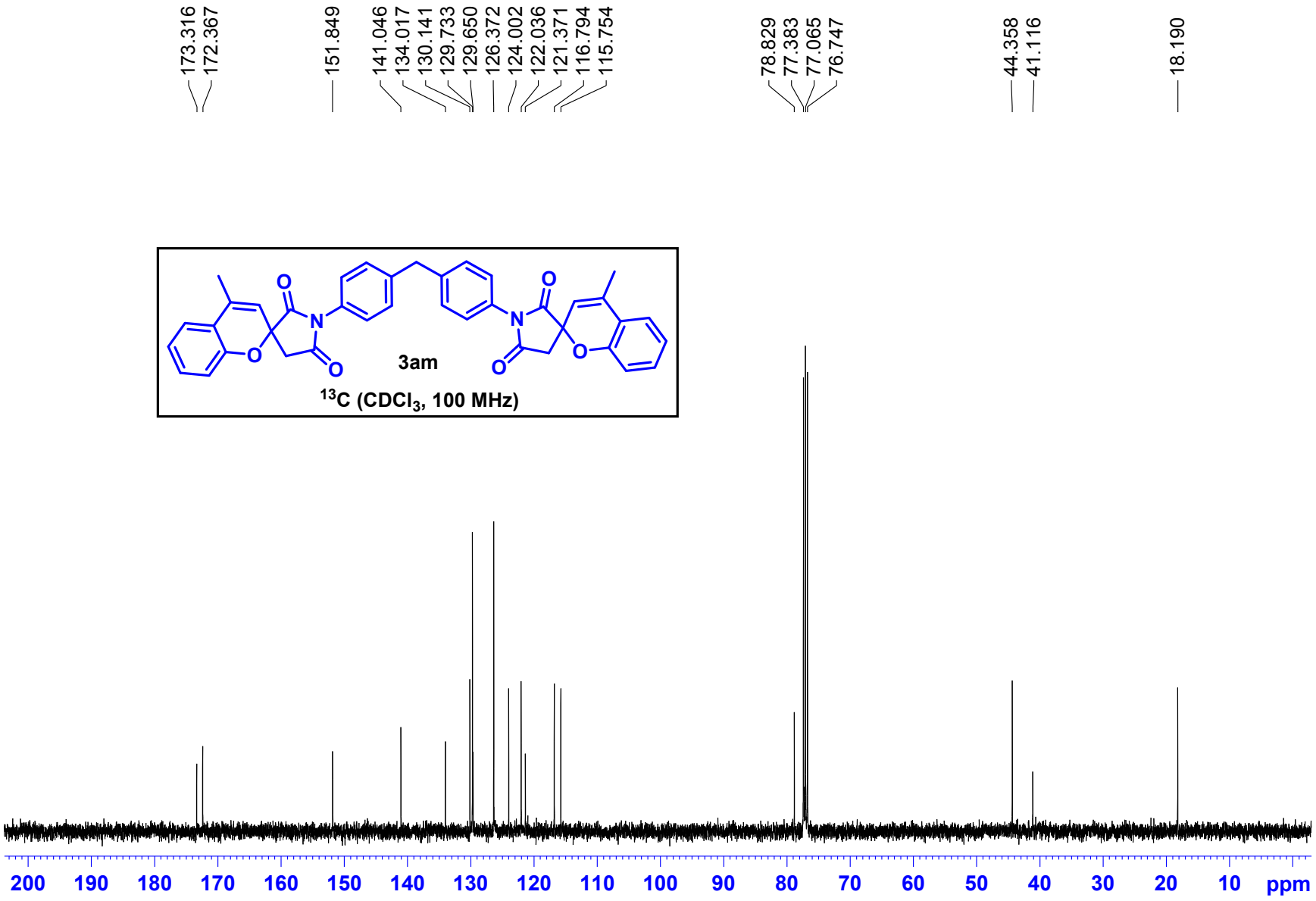

8.731
7.265
7.220
7.201
7.182
7.163
7.160
6.979
6.960
6.942
6.827
6.807
5.364
5.362
3.170
3.124
2.934
2.888
2.105
2.102
O
O
N
H
O
3al
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.012
2.018
1.003
1.000
1.009
1.012
1.003
3.030

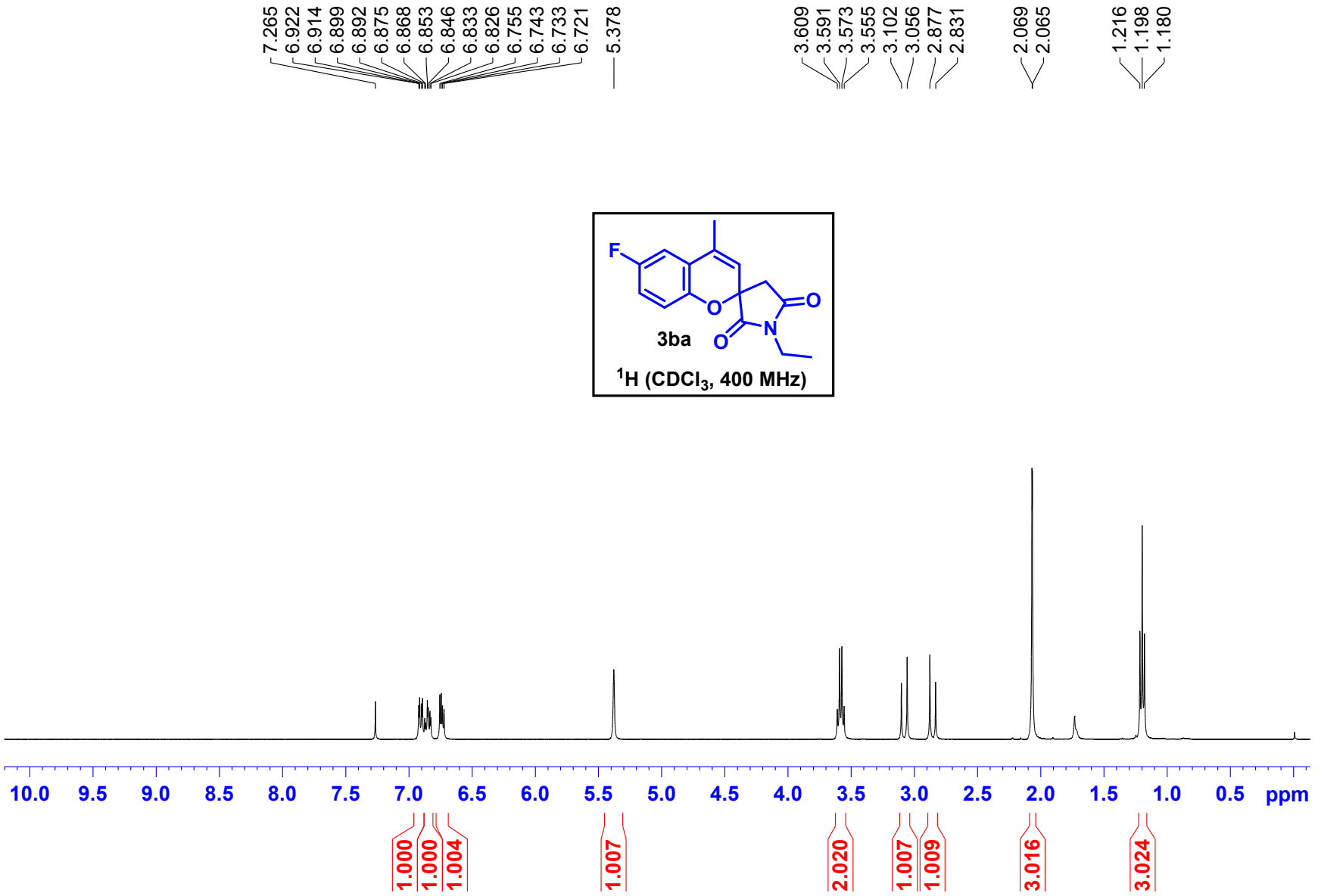

174.652
173.470
151.726
133.798
130.148
124.004
122.058
121.183
116.518
115.671
79.982
77.377
77.059
76.741
45.321
18.126
O
O
N
H
O
3al
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

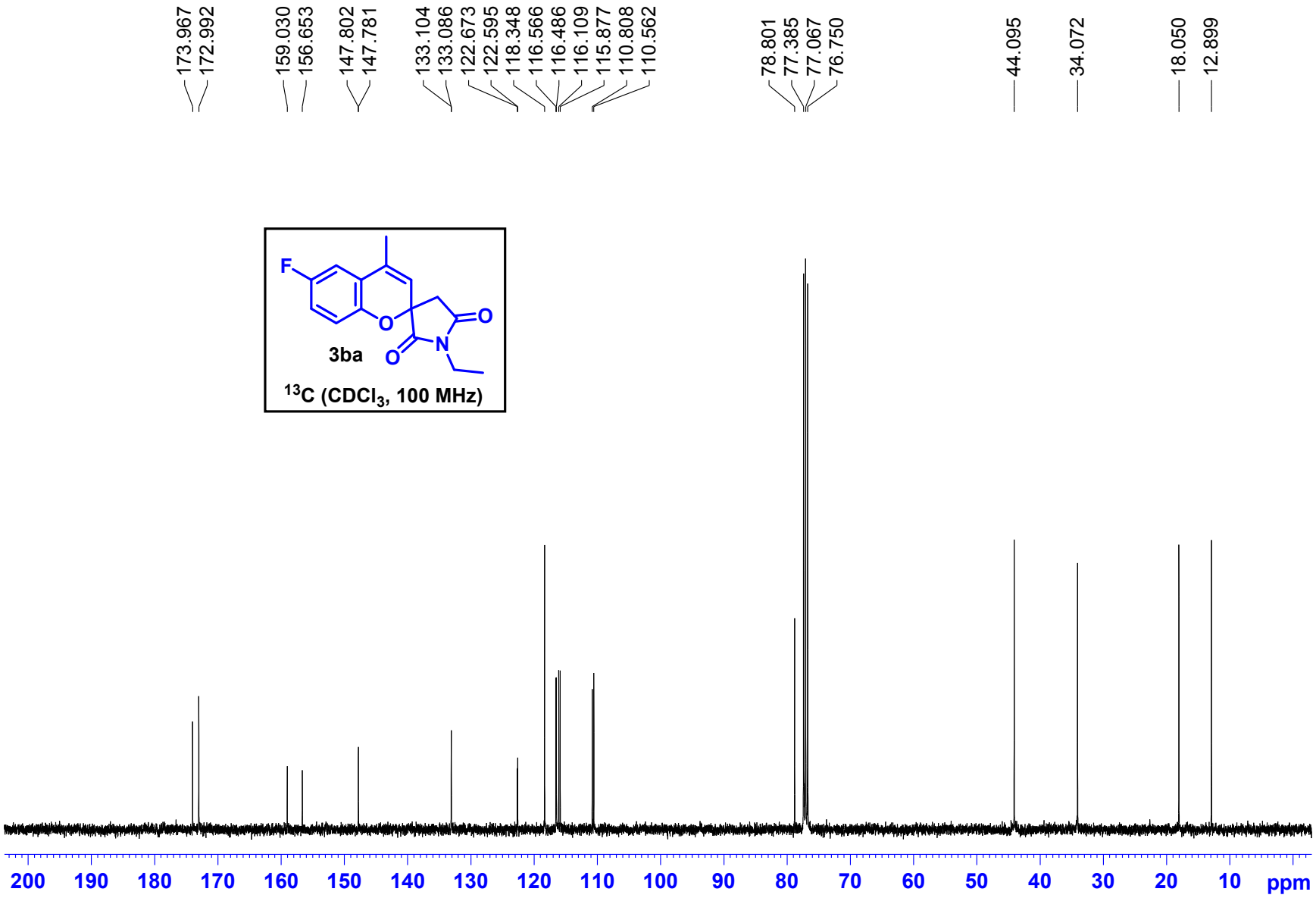

7.261
7.251
7.230
7.227
7.210
7.207
7.201
7.198
7.182
7.162
7.159
6.975
6.973
6.956
6.954
6.938
6.935
6.850
6.831
6.830
5.435
5.433
4.021
3.296
3.251
3.048
3.003
2.121
2.118
-0.000

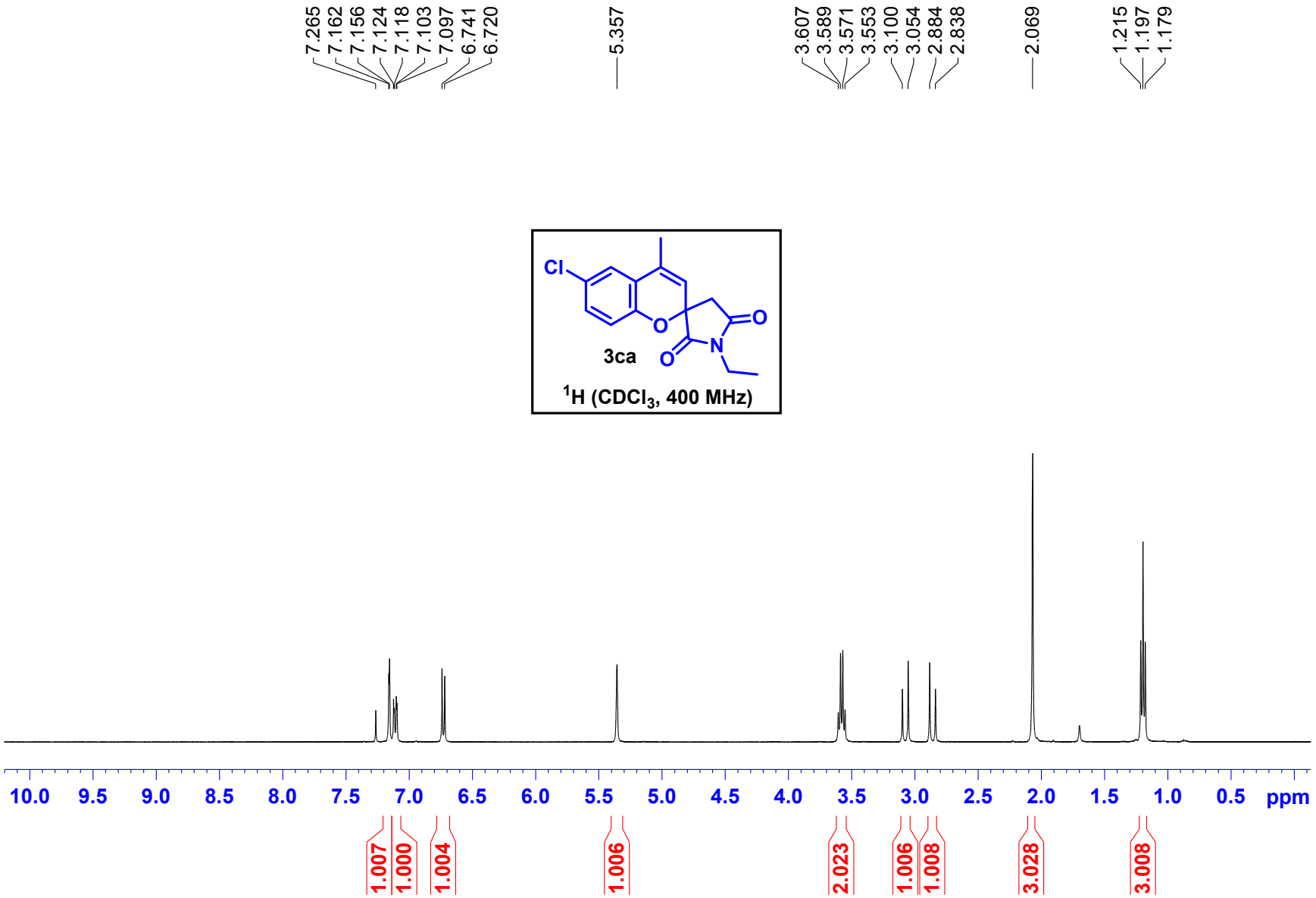

3am
^{1}H (CDCl$_3$, 400 MHz)

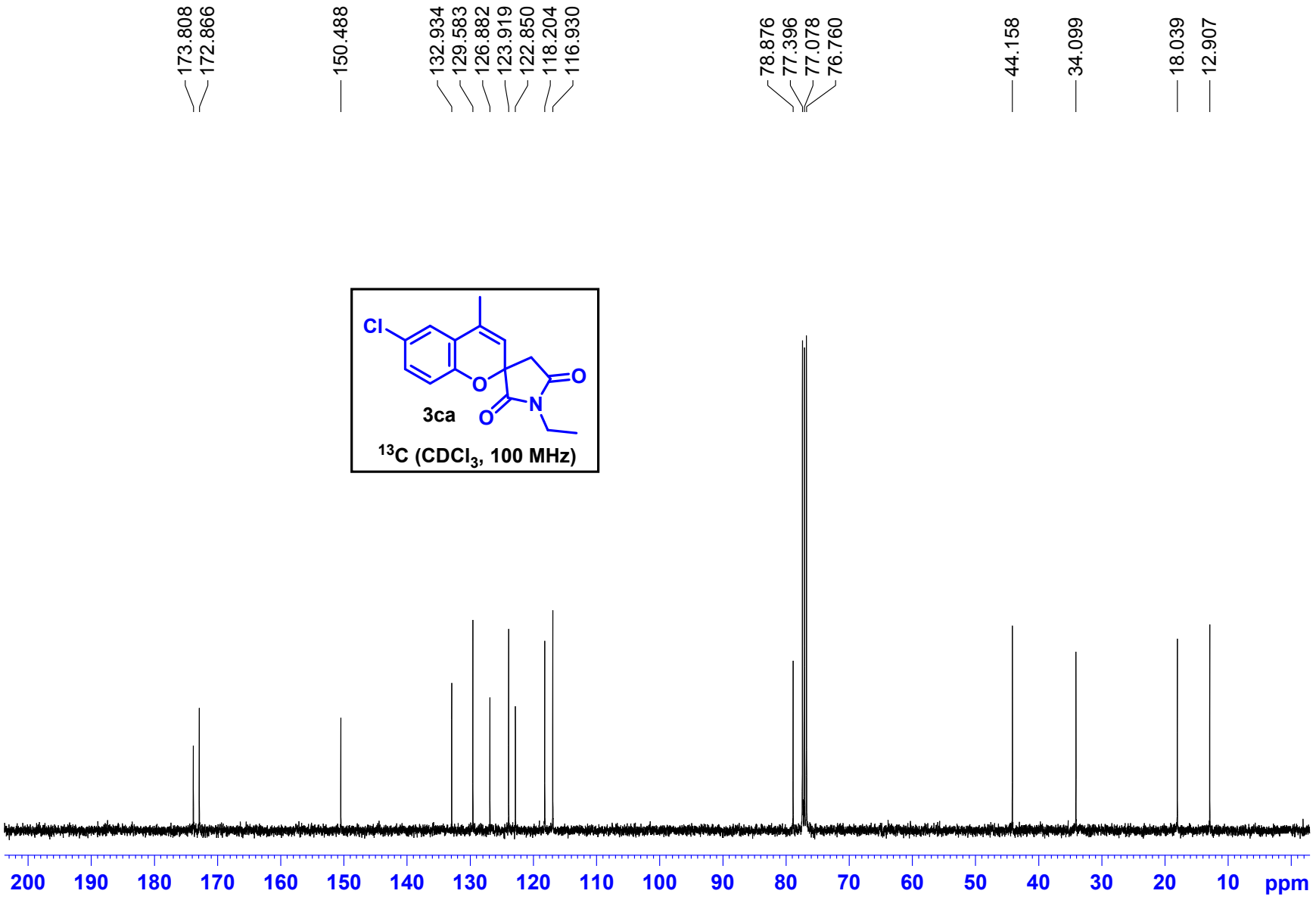

10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
8.052
4.019
2.024
2.017
2.000
2.016
2.020
2.014
6.001

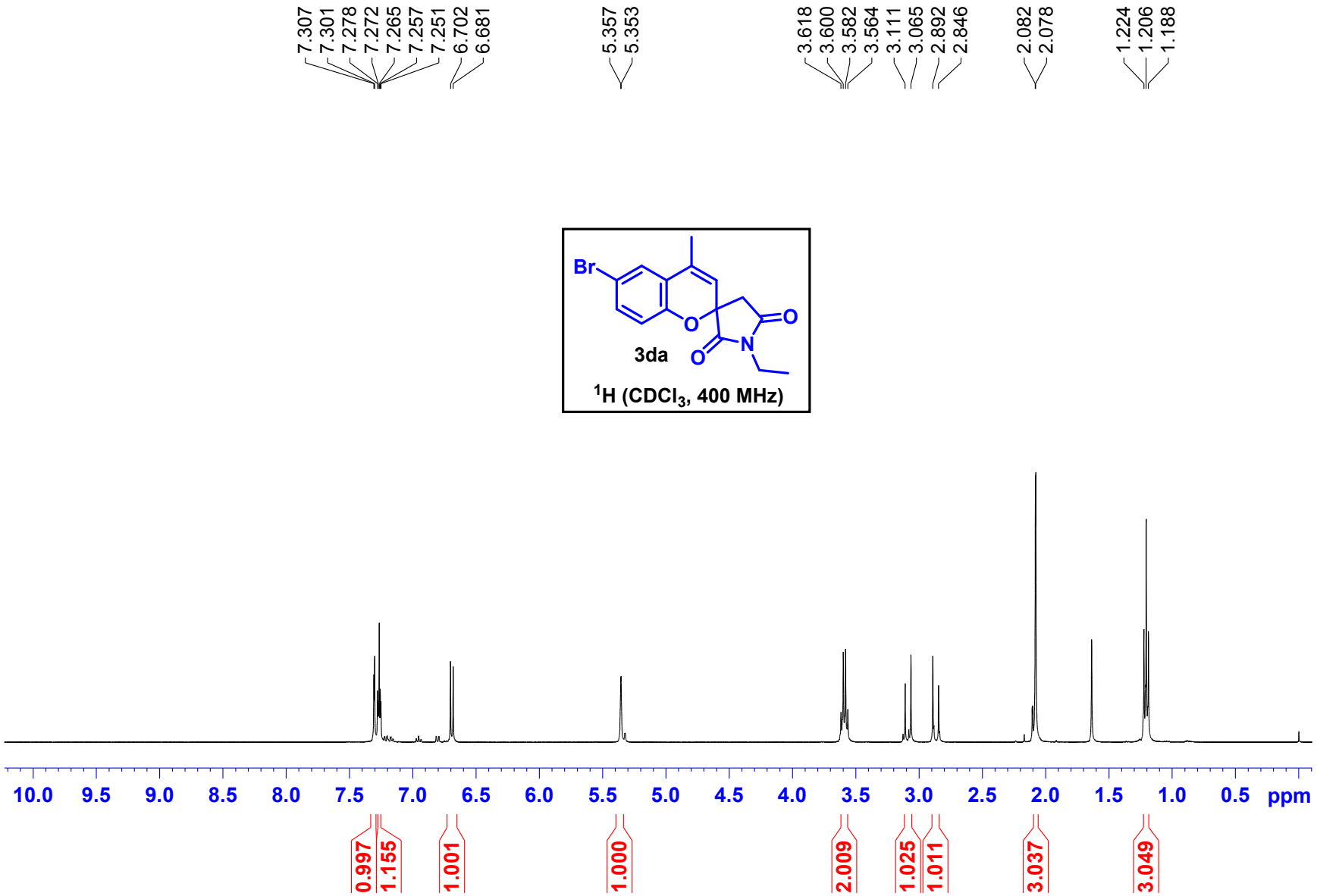

173.316
172.367
151.849
141.046
134.017
130.141
129.733
129.650
126.372
124.002
122.036
121.371
116.794
115.754
78.829
77.383
77.065
76.747
44.358
41.116
18.190
3am
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.265
6.922
6.914
6.899
6.892
6.875
6.868
6.853
6.846
6.833
6.826
6.755
6.743
6.733
6.721
5.378
3.609
3.591
3.573
3.555
3.102
3.056
2.877
2.831
2.069
2.065
1.216
1.198
1.180
F
O
O
N
O
3ba
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.000
1.000
1.004
1.007
2.020
1.007
1.009
3.016
3.024

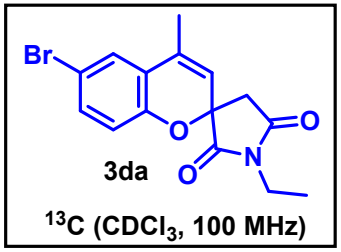

173.967
172.992
159.030
156.653
147.802
147.781
133.104
133.086
122.673
122.595
118.348
116.566
116.486
116.109
115.877
110.808
110.562
78.801
77.385
77.067
76.750
44.095
34.072
18.050
12.899
F
O
O
N
O
3ba
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.265
7.162
7.156
7.124
7.118
7.103
7.097
6.741
6.720
5.357
3.607
3.589
3.571
3.553
3.100
3.054
2.884
2.838
2.069
1.215
1.197
1.179
Cl
O
O
N
O
3ca
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.007
1.000
1.004
1.006
2.023
1.006
1.008
3.028
3.008

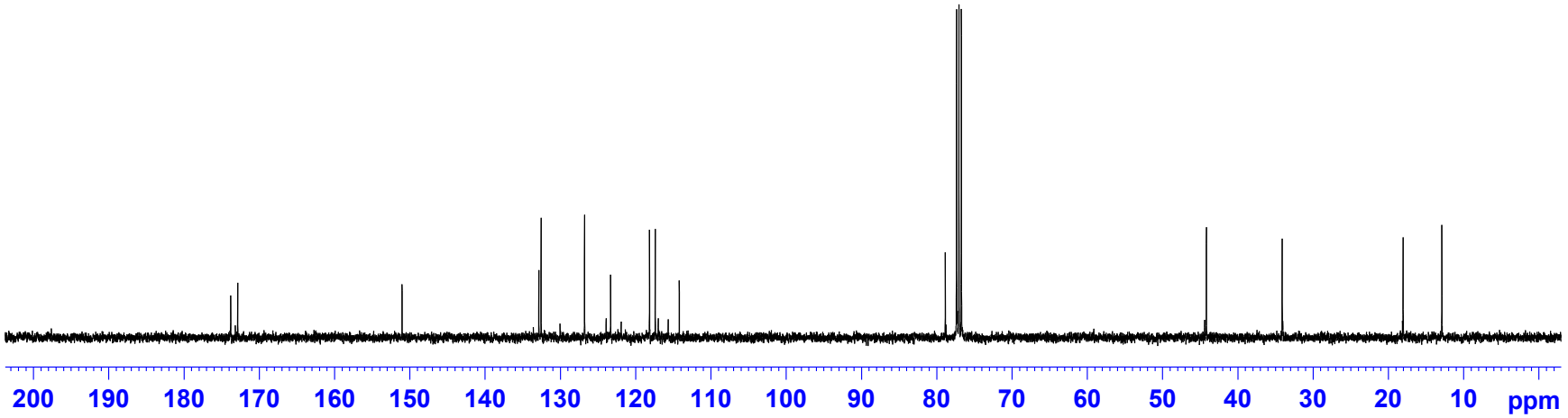

173.808
172.866
150.488
132.934
129.583
126.882
123.919
122.850
118.204
116.930
78.876
77.396
77.078
76.760
44.158
34.099
18.039
12.907
Cl
O
O
N
O
3ca
^{13}C (CDCl$_3$, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

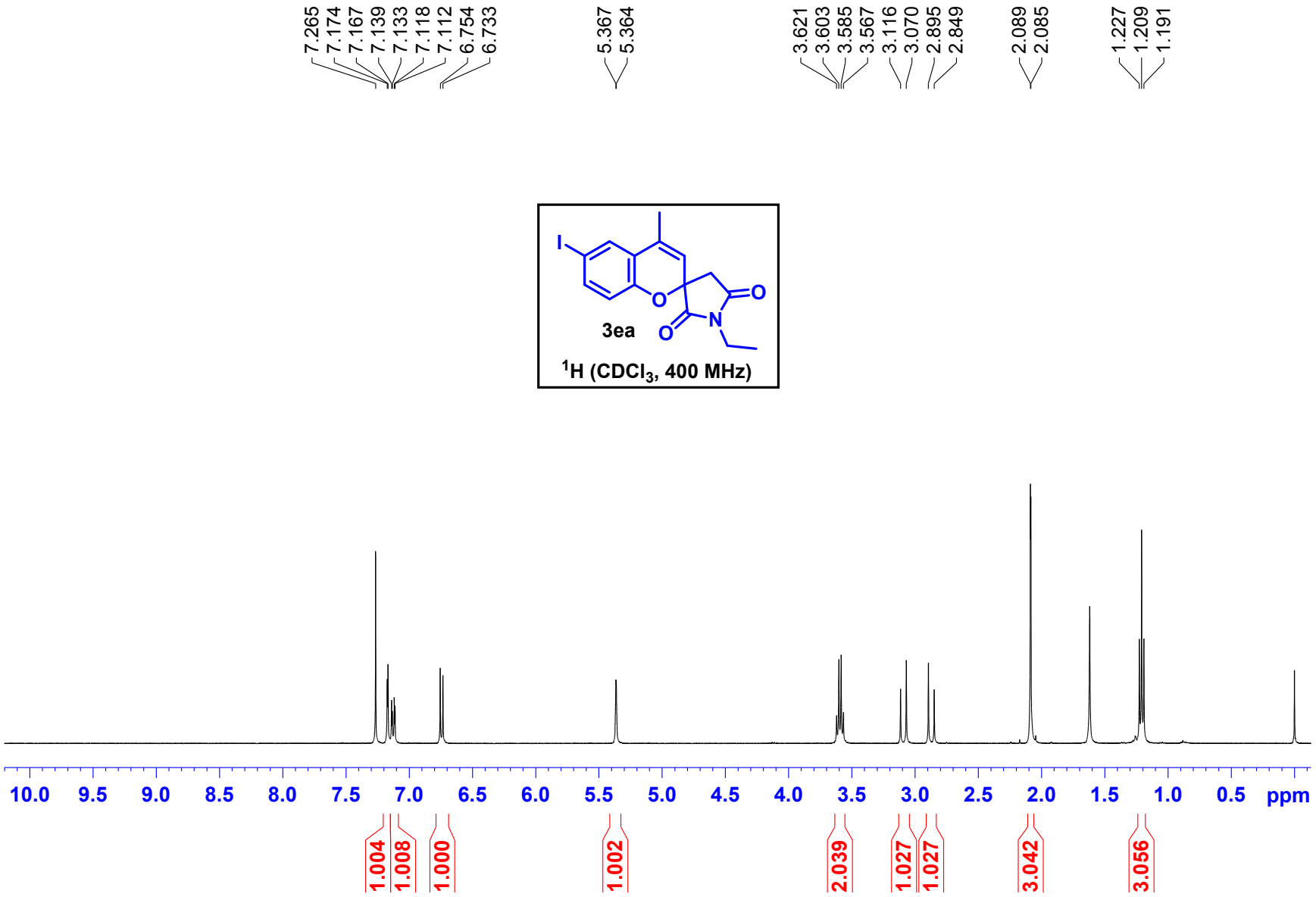

7.307
7.301
7.278
7.272
7.265
7.257
7.251
6.702
6.681
5.357
5.353
3.618
3.600
3.582
3.564
3.111
3.065
2.892
2.846
2.082
2.078
1.224
1.206
1.188
Br
O
O
N
O
3da
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
0.997
1.155
1.001
1.000
2.009
1.025
1.011
3.037
3.049

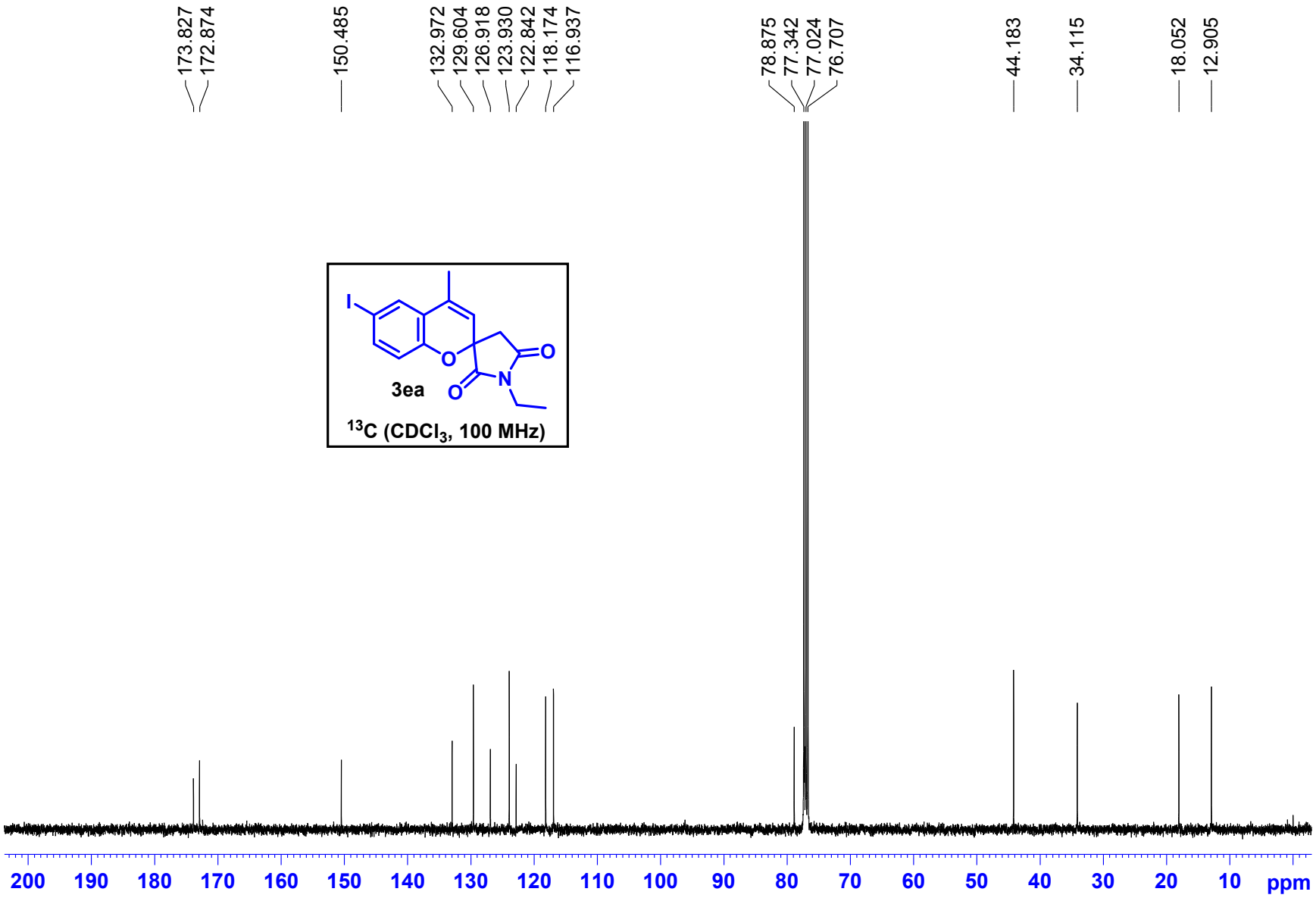
173.773
172.829
151.038
132.862
132.560
126.788
123.325
118.159
117.396
114.205
78.866
77.365
77.046
76.729
44.178
34.104
18.033
12.889

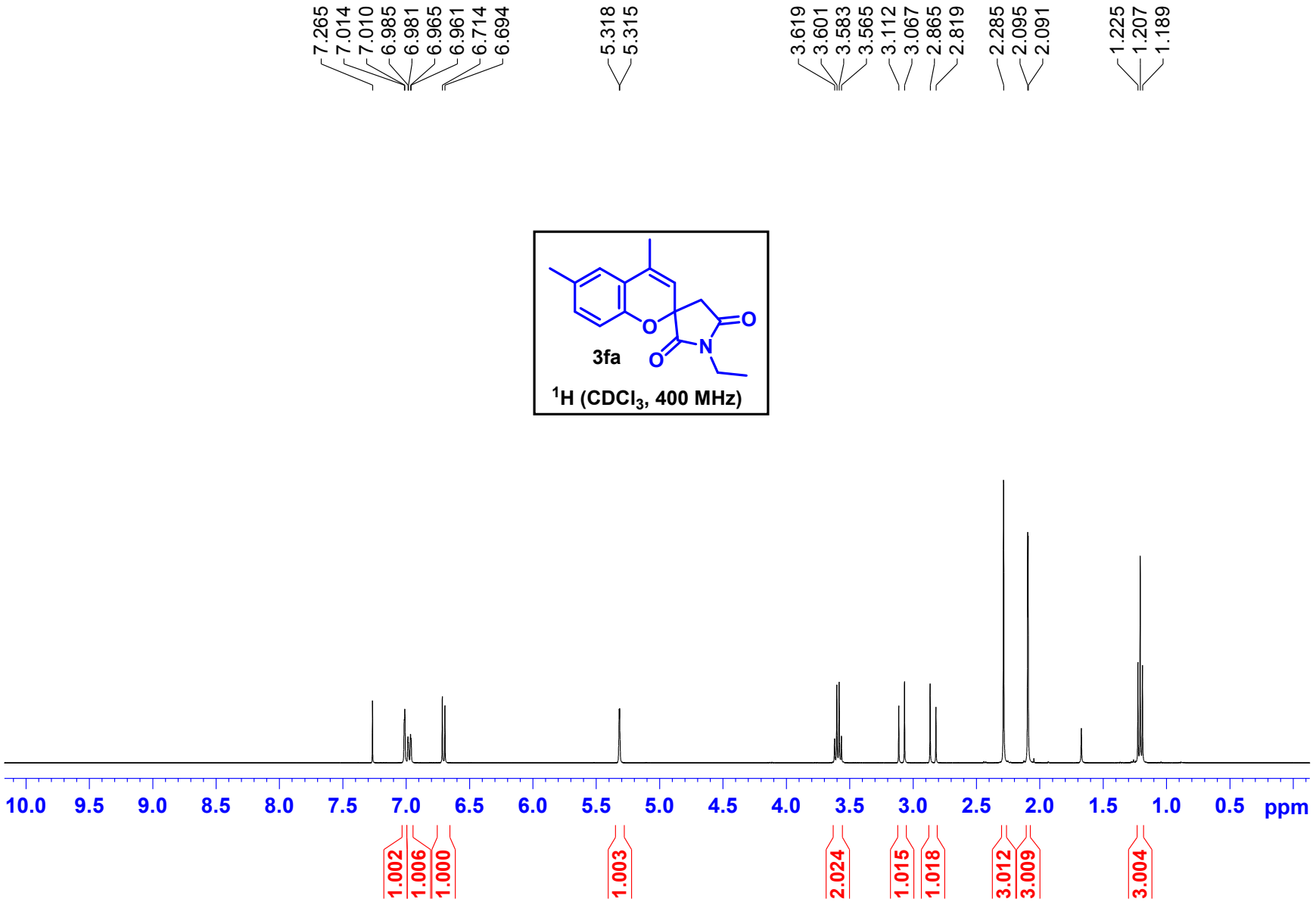


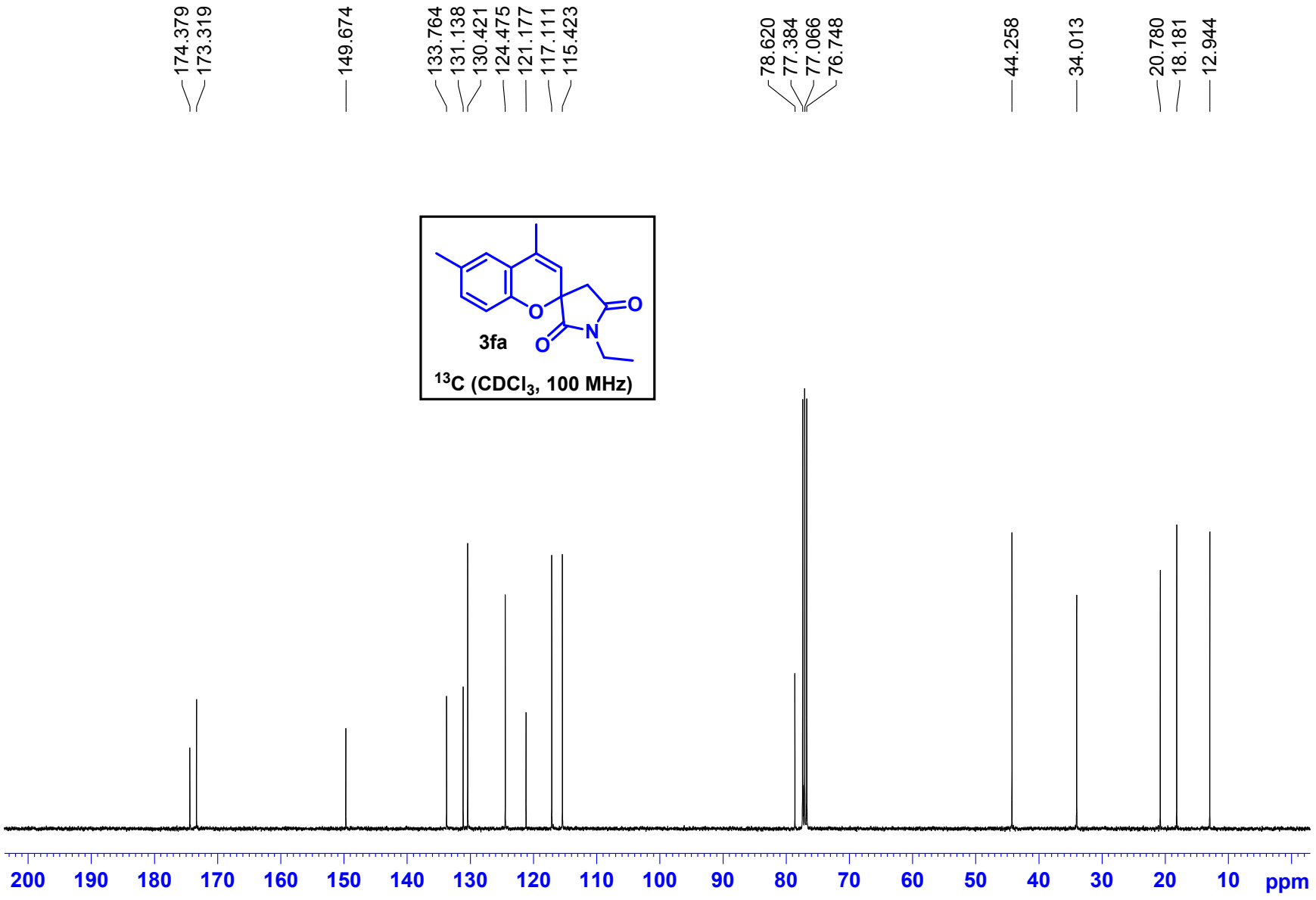
^{13}C ($CDCl_3$, 100 MHz)

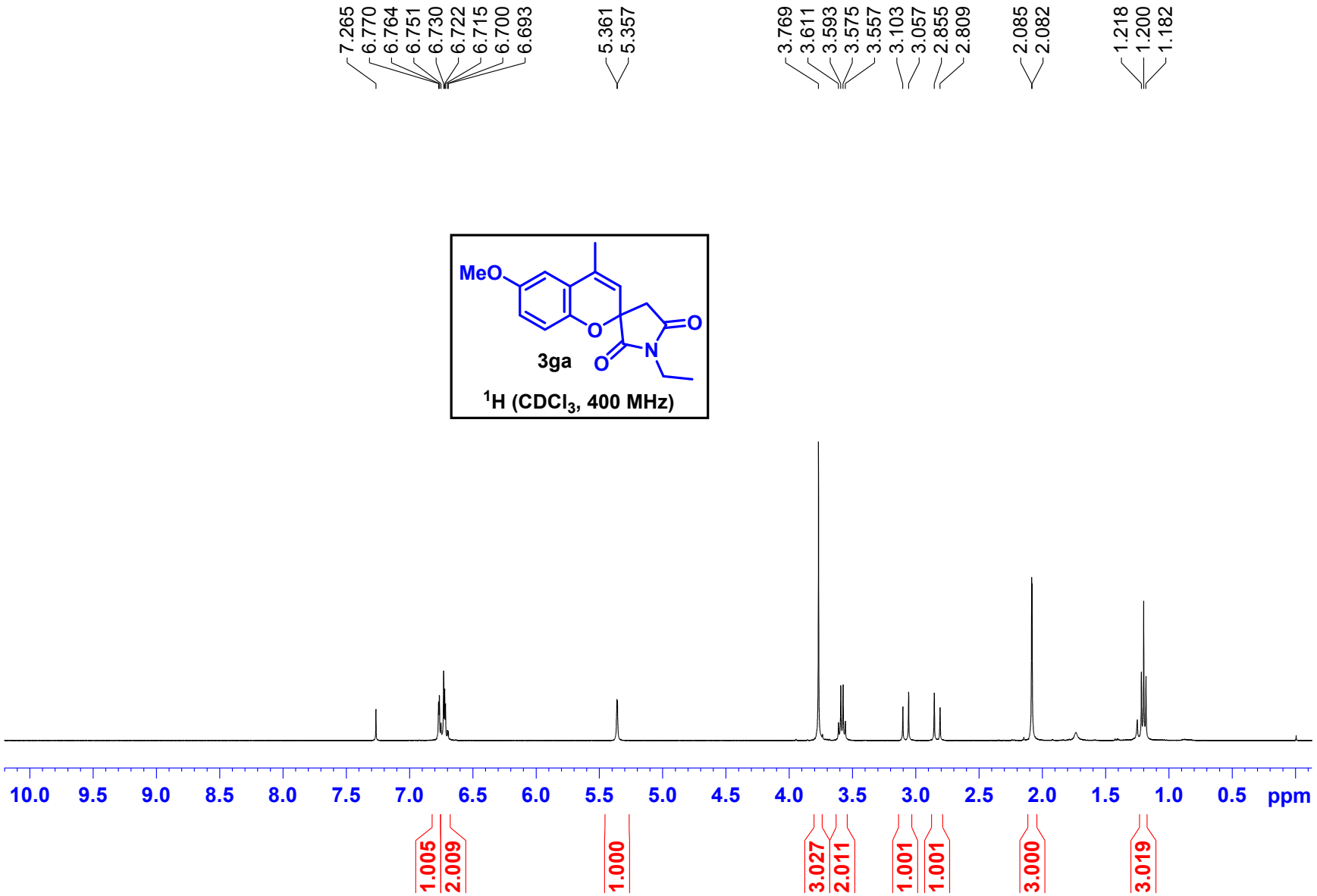


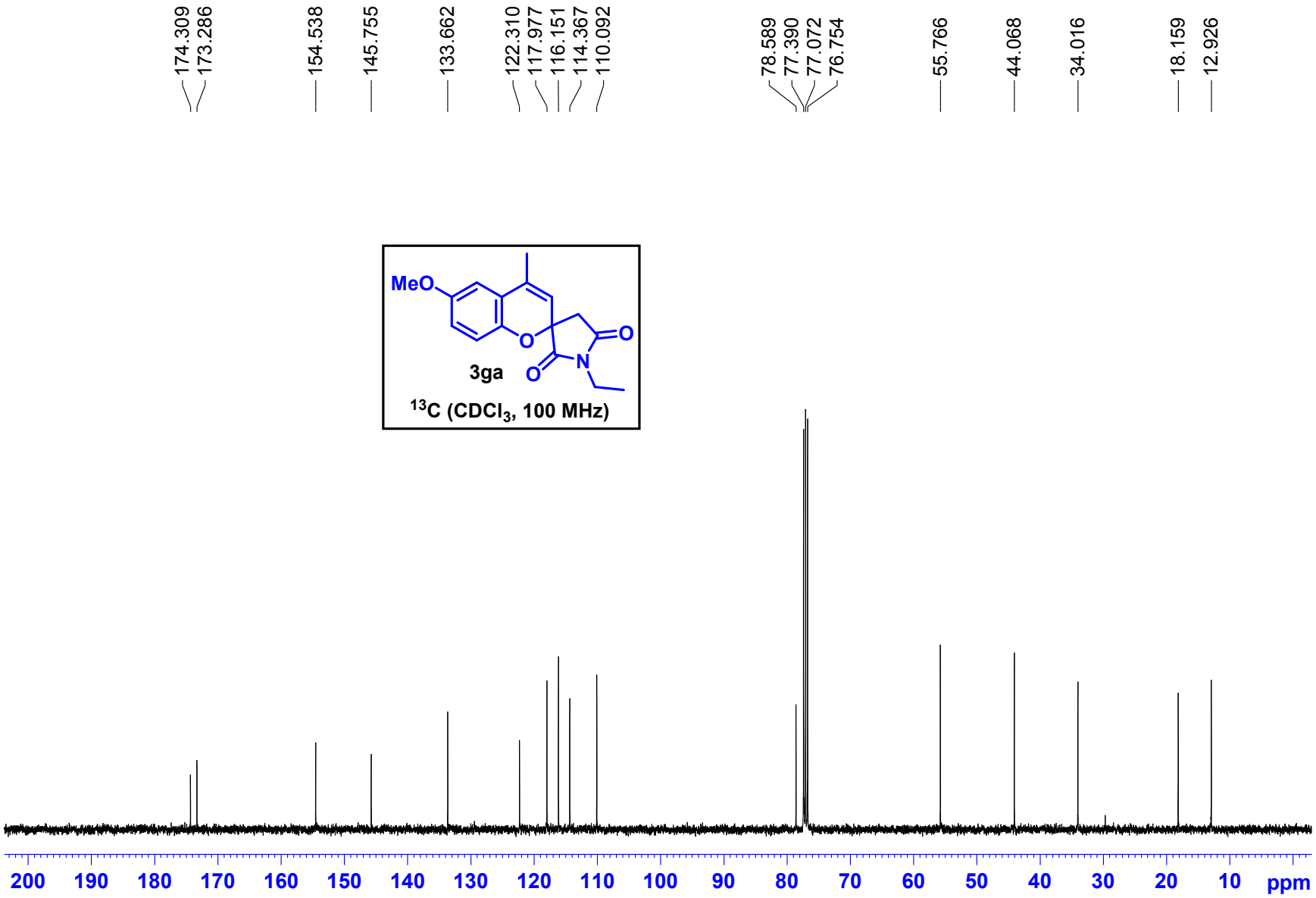

7.265
7.174
7.167
7.139
7.133
7.118
7.112
6.754
6.733
5.367
5.364
3.621
3.603
3.585
3.567
3.116
3.070
2.895
2.849
2.089
2.085
1.227
1.209
1.191
I
O
O
N
O
3ea
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.004
1.008
1.000
1.002
2.039
1.027
1.027
3.042
3.056

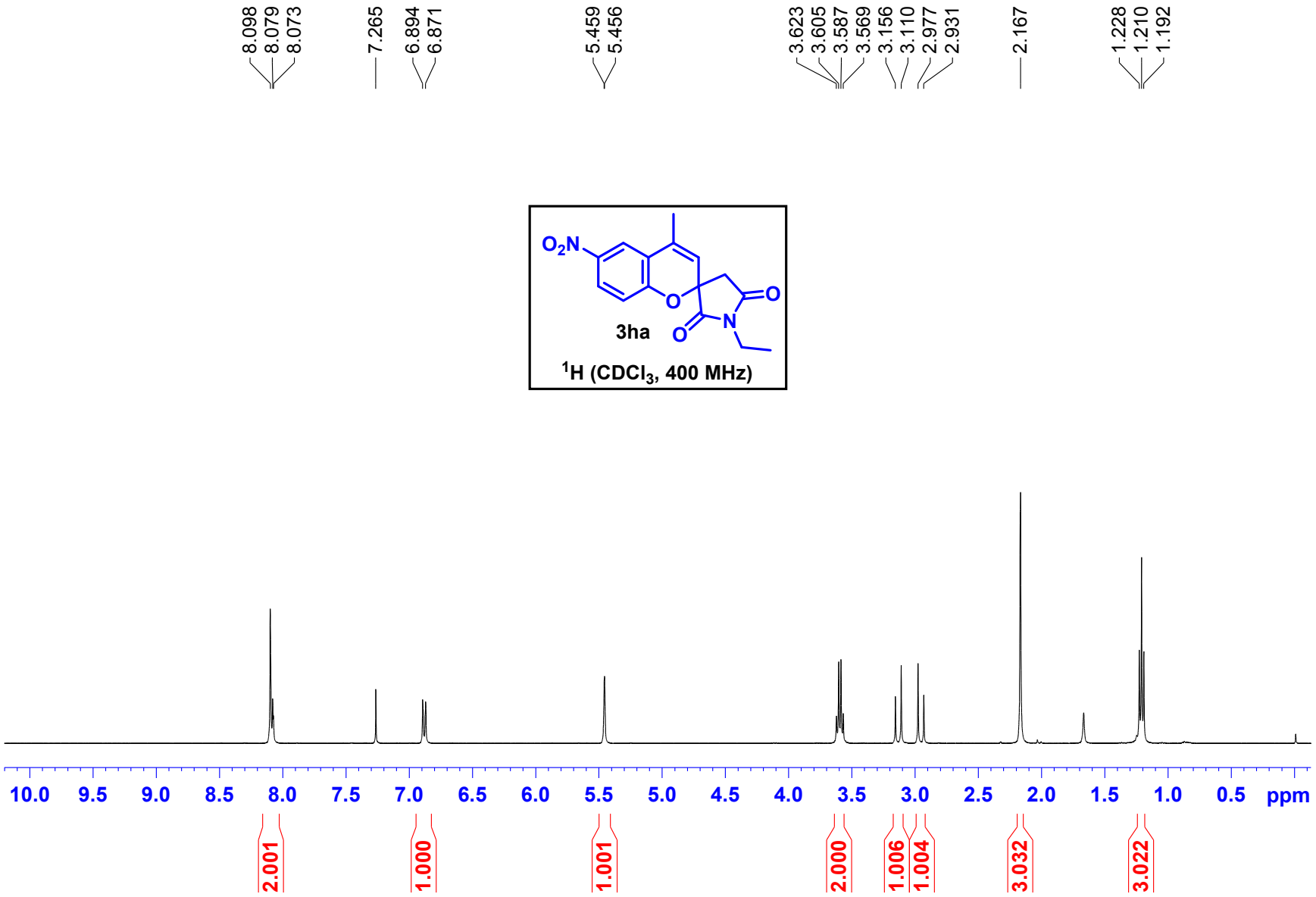

173.827
172.874
150.485
132.972
129.604
126.918
123.930
122.842
118.174
116.937
78.875
77.342
77.024
76.707
44.183
34.115
18.052
12.905
I
O
O
N
O
3ea
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

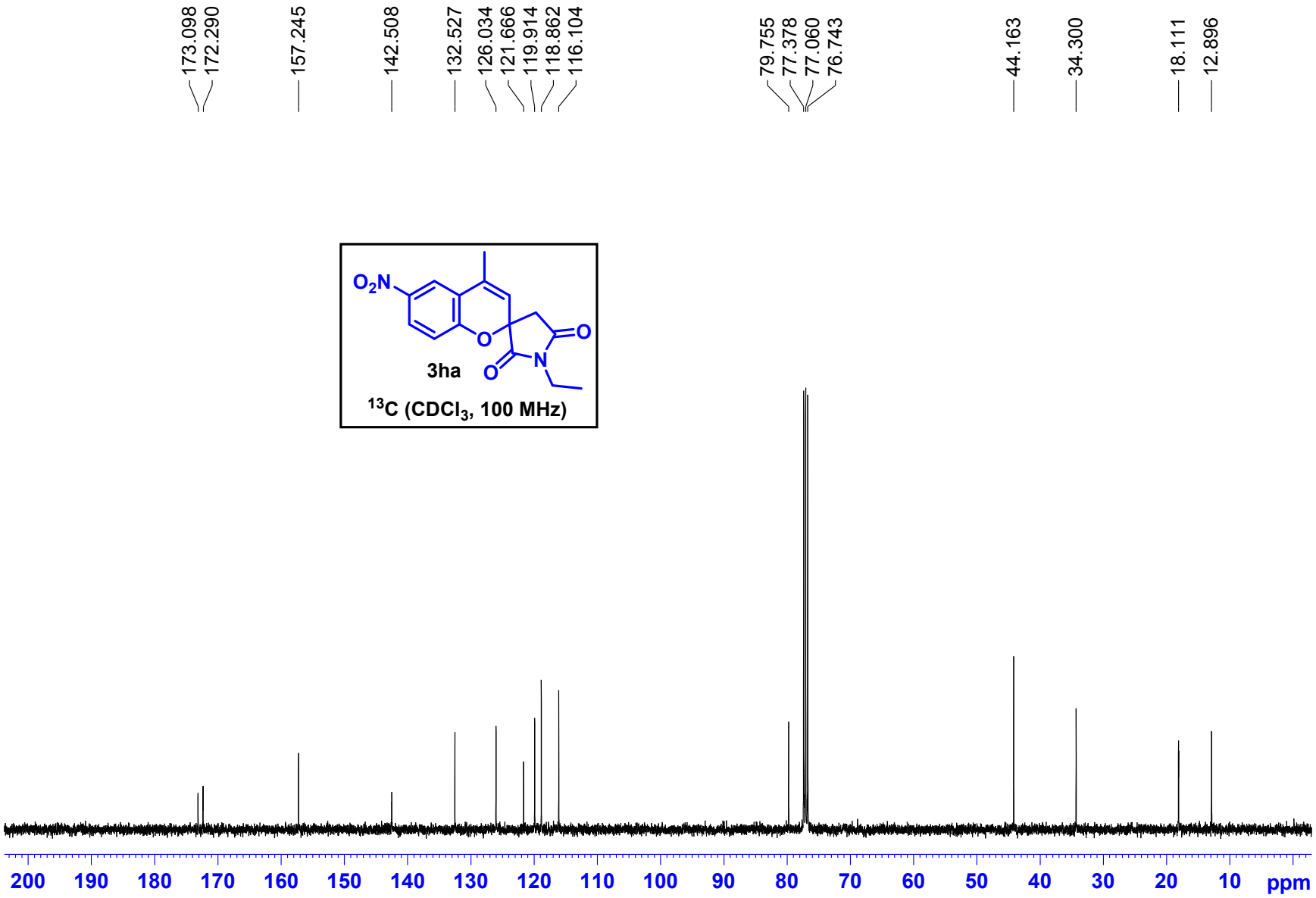

7.265
7.014
7.010
6.985
6.981
6.965
6.961
6.714
6.694
5.318
5.315
3.619
3.601
3.583
3.565
3.112
3.067
2.865
2.819
2.285
2.095
2.091
1.225
1.207
1.189
3fa
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.002
1.006
1.000
1.003
2.024
1.015
1.018
3.012
3.009
3.004

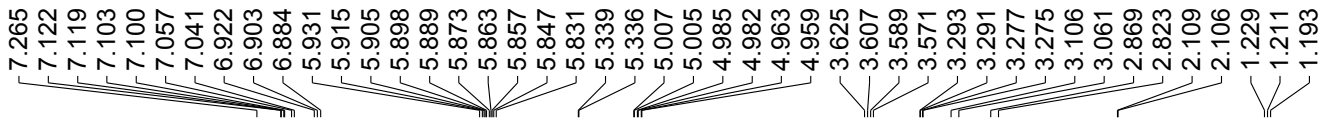

174.379
173.319
149.674
133.764
131.138
130.421
124.475
121.177
117.111
115.423
78.620
77.384
77.066
76.748
44.258
34.013
20.780
18.181
12.944
O
O
O
N
3fa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

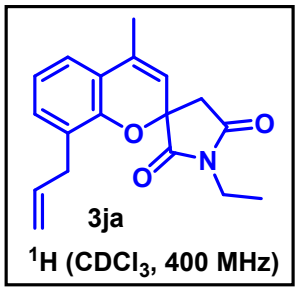

7.265
6.770
6.764
6.751
6.730
6.722
6.715
6.700
6.693
5.361
5.357
3.769
3.611
3.593
3.575
3.557
3.103
3.057
2.855
2.809
2.085
2.082
1.218
1.200
1.182
MeO
O
O
N
O
3ga
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.005
2.009
1.000
3.027
2.011
1.001
1.001
3.000
3.019

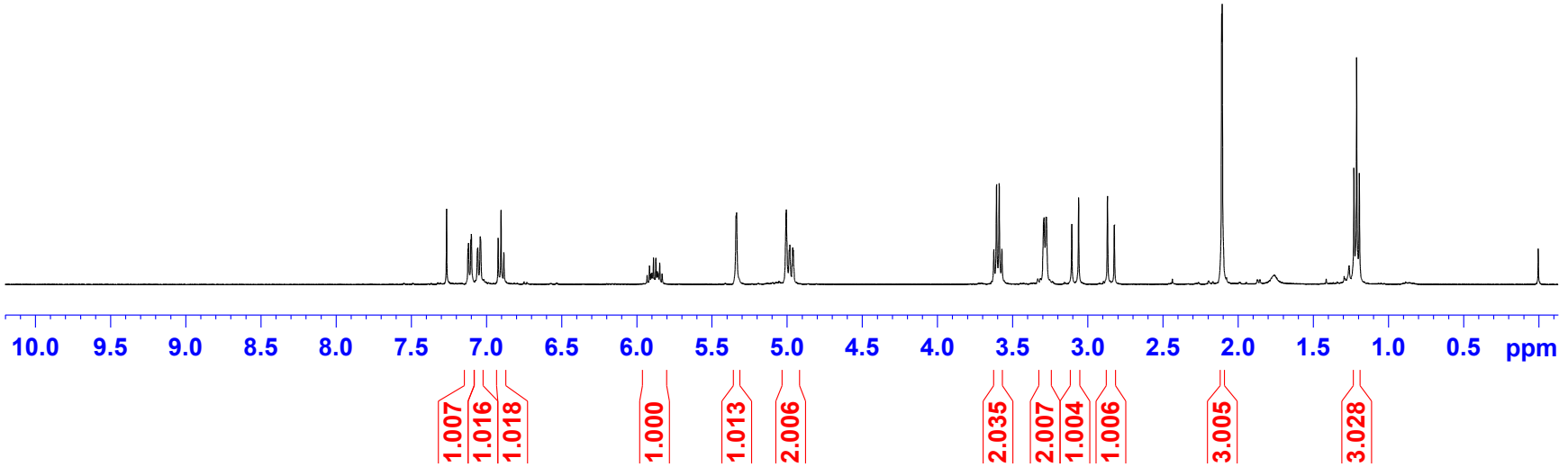

174.309
173.286
154.538
145.755
133.662
122.310
117.977
116.151
114.367
110.092
78.589
77.390
77.072
76.754
55.766
44.068
34.016
18.159
12.926
MeO
O
O
N
O
3ga
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

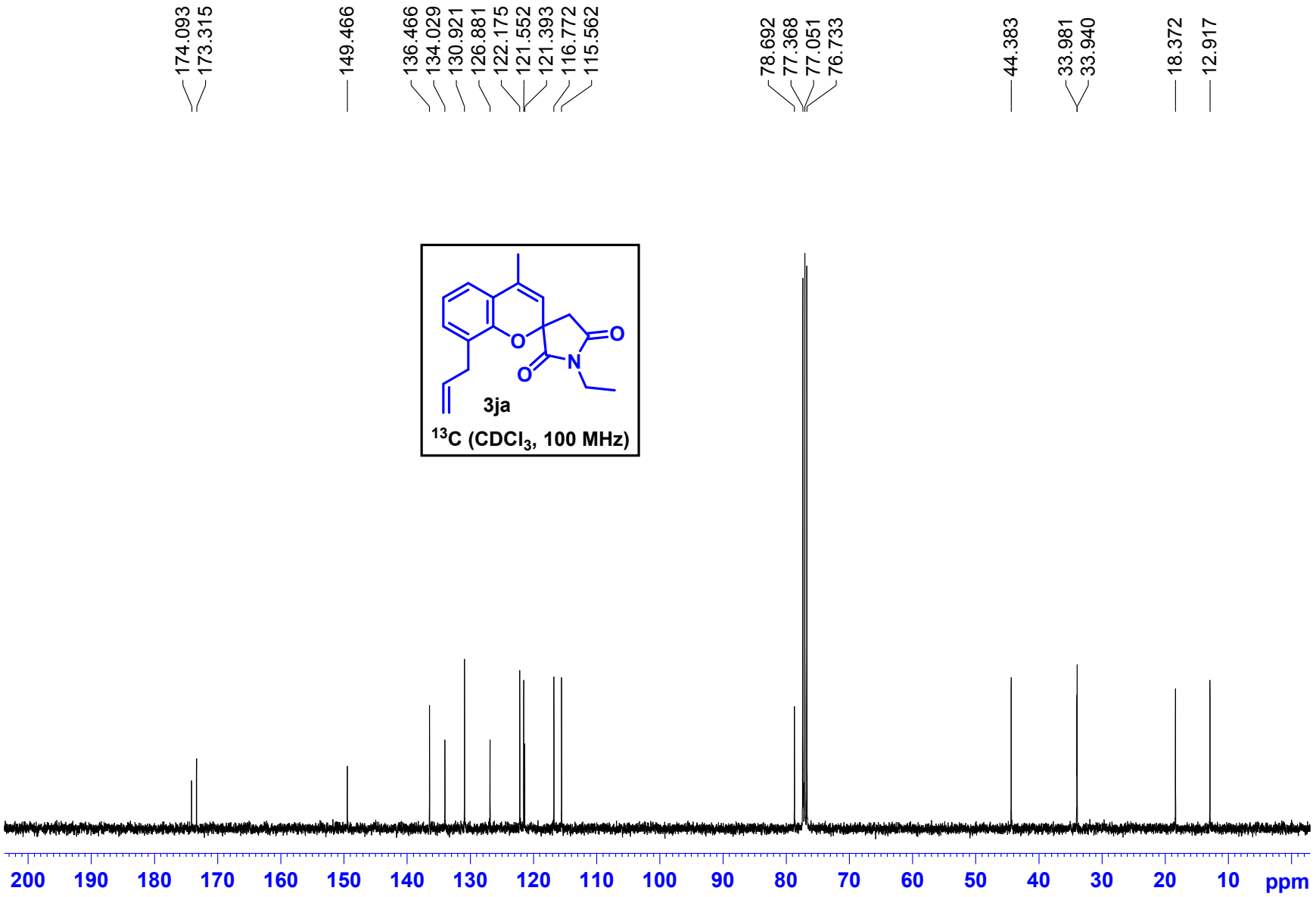

8.098
8.079
8.073
7.265
6.894
6.871
5.459
5.456
3.623
3.605
3.587
3.569
3.156
3.110
2.977
2.931
2.167
1.228
1.210
1.192
O2N
O
O
N
O
3ha
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
2.001
1.000
1.001
2.000
1.006
1.004
3.032
3.022

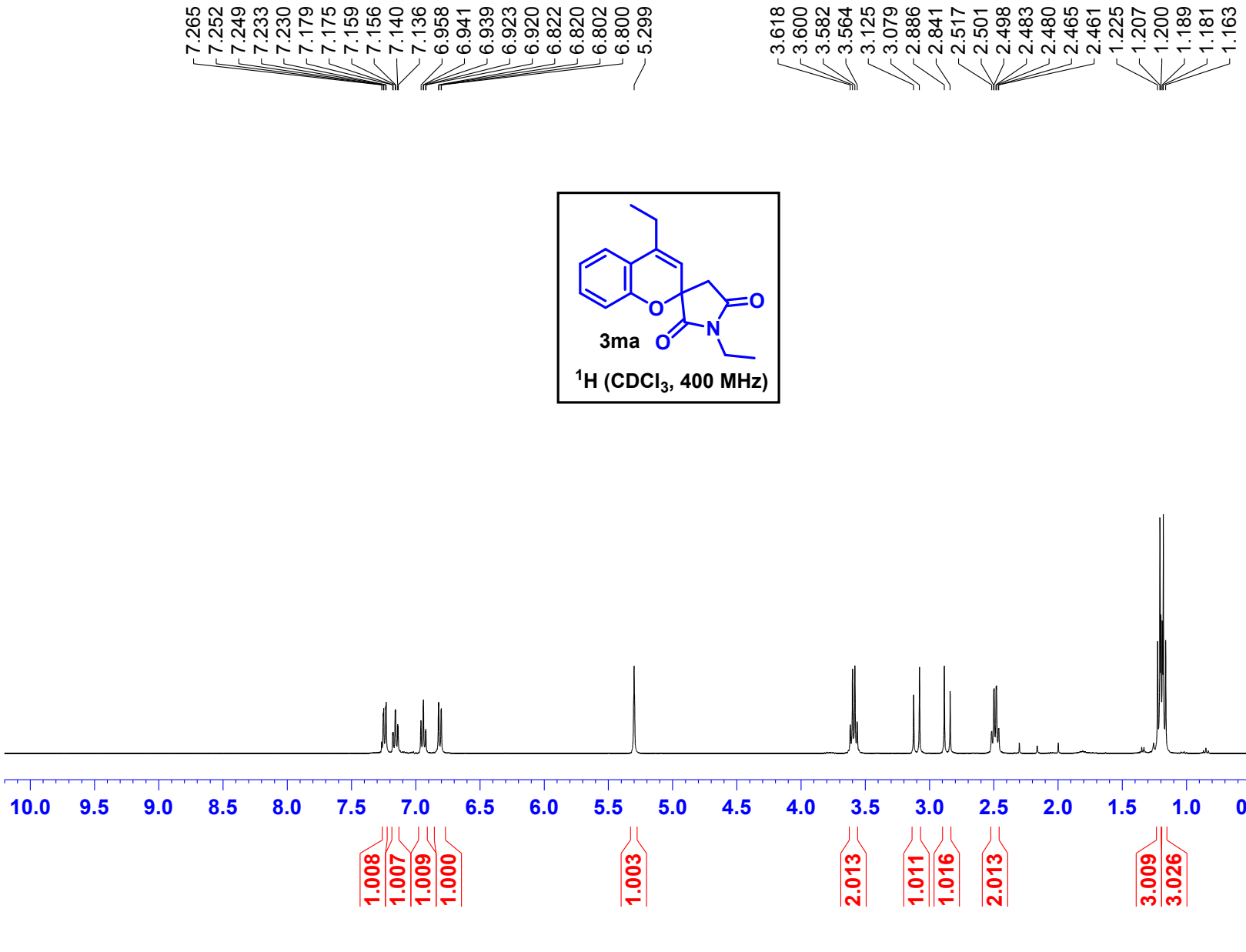

173.098
172.290
157.245
142.508
132.527
126.034
121.666
119.914
118.862
116.104
79.755
77.378
77.060
76.743
44.163
34.300
18.111
12.896
O2N
O
O
N
O
3ha
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

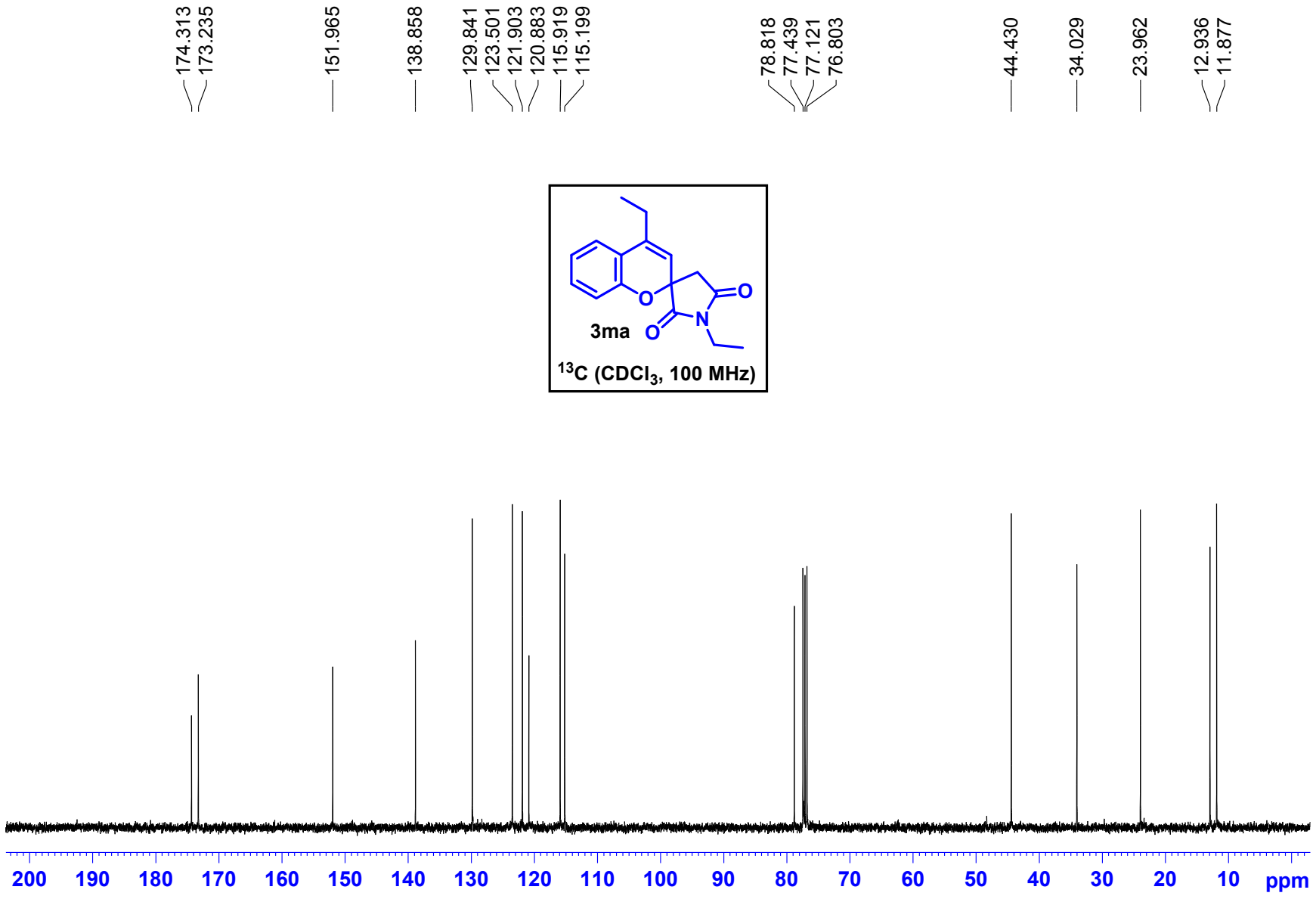

7.265
7.122
7.119
7.103
7.100
7.057
7.041
6.922
6.903
6.884
5.931
5.915
5.905
5.898
5.889
5.873
5.863
5.857
5.847
5.831
5.339
5.336
5.007
5.005
4.985
4.982
4.963
4.959
3.625
3.607
3.589
3.571
3.293
3.291
3.277
3.275
3.106
3.061
2.869
2.823
2.109
2.106
1.229
1.211
1.193

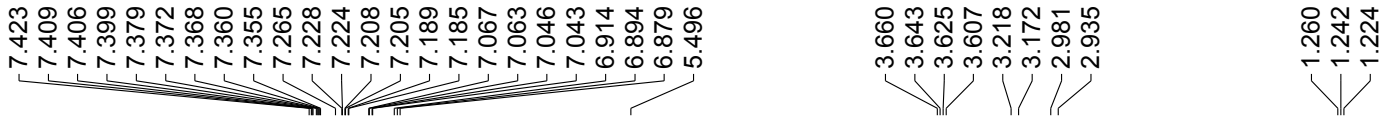

O
O
O
N
3ja
^{1}H (CDCl$_3$, 400 MHz)

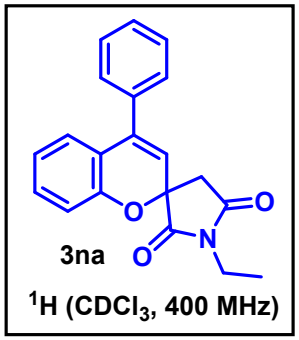

10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.007
1.016
1.018
1.000
1.013
2.006
2.035
2.007
1.004
1.006
3.005
3.028

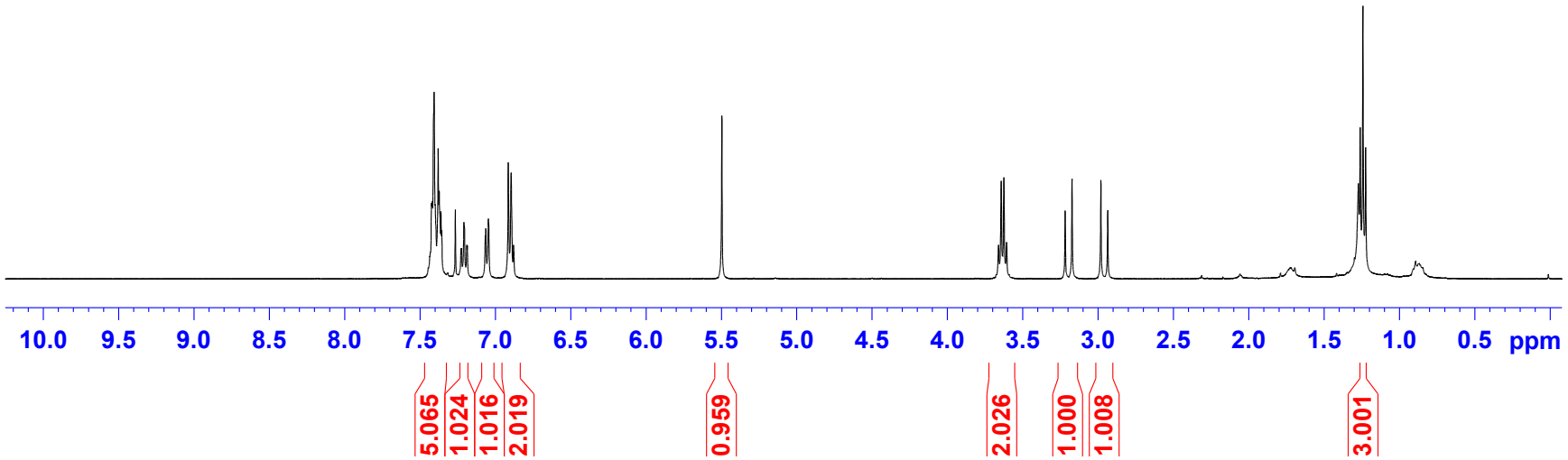

174.093
173.315
149.466
136.466
134.029
130.921
126.881
122.175
121.552
121.393
116.772
115.562
78.692
77.368
77.051
76.733
44.383
33.981
33.940
18.372
12.917
O
O
O
N
3ja
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

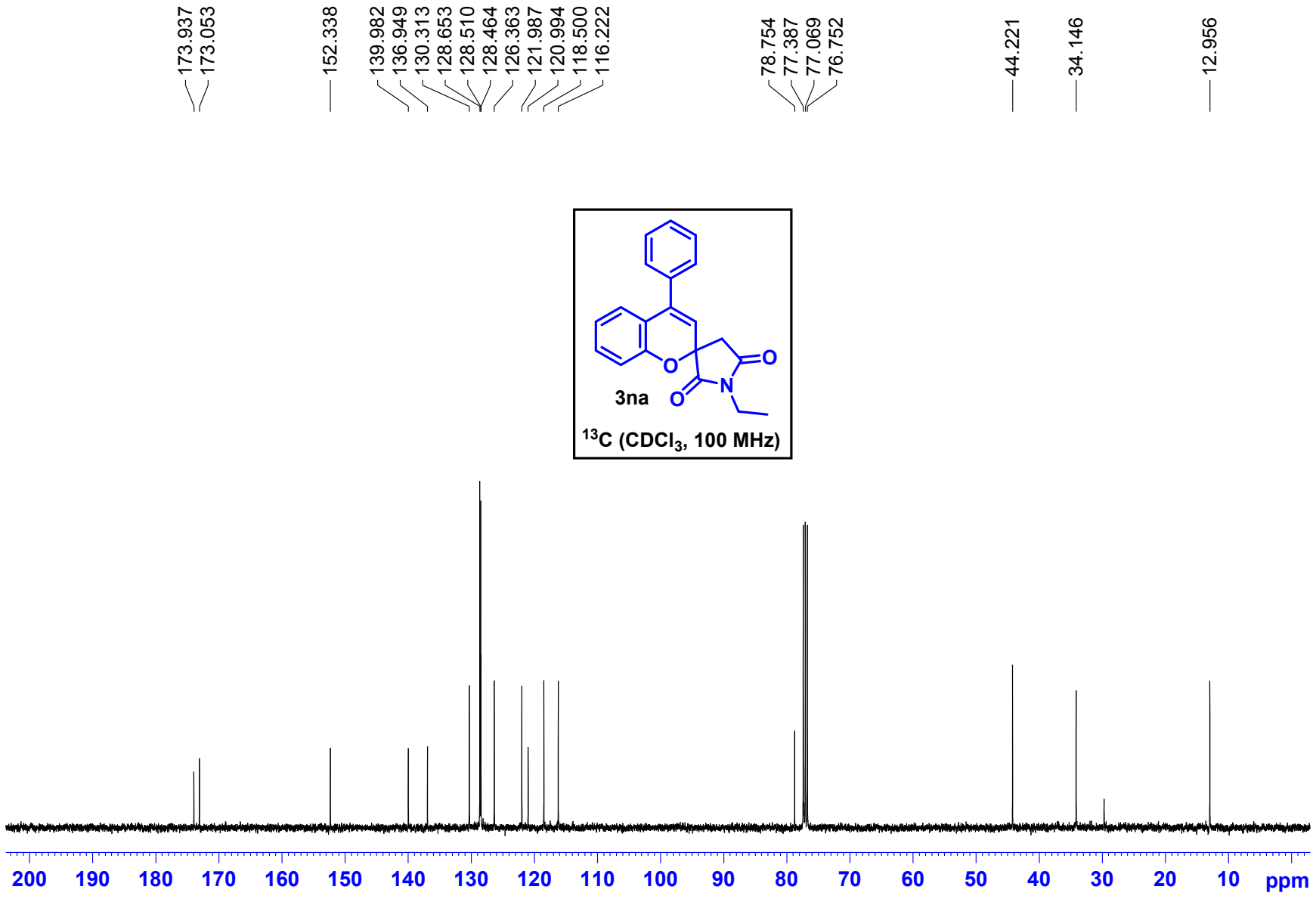

7.265
7.252
7.249
7.233
7.230
7.179
7.175
7.159
7.156
7.140
7.136
6.958
6.941
6.939
6.923
6.920
6.822
6.820
6.802
6.800
5.299
3.618
3.600
3.582
3.564
3.125
3.079
2.886
2.841
2.517
2.501
2.498
2.483
2.480
2.465
2.461
1.225
1.207
1.200
1.189
1.181
1.163
3ma
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.008
1.007
1.009
1.000
1.003
2.013
1.011
1.016
2.013
3.009
3.026

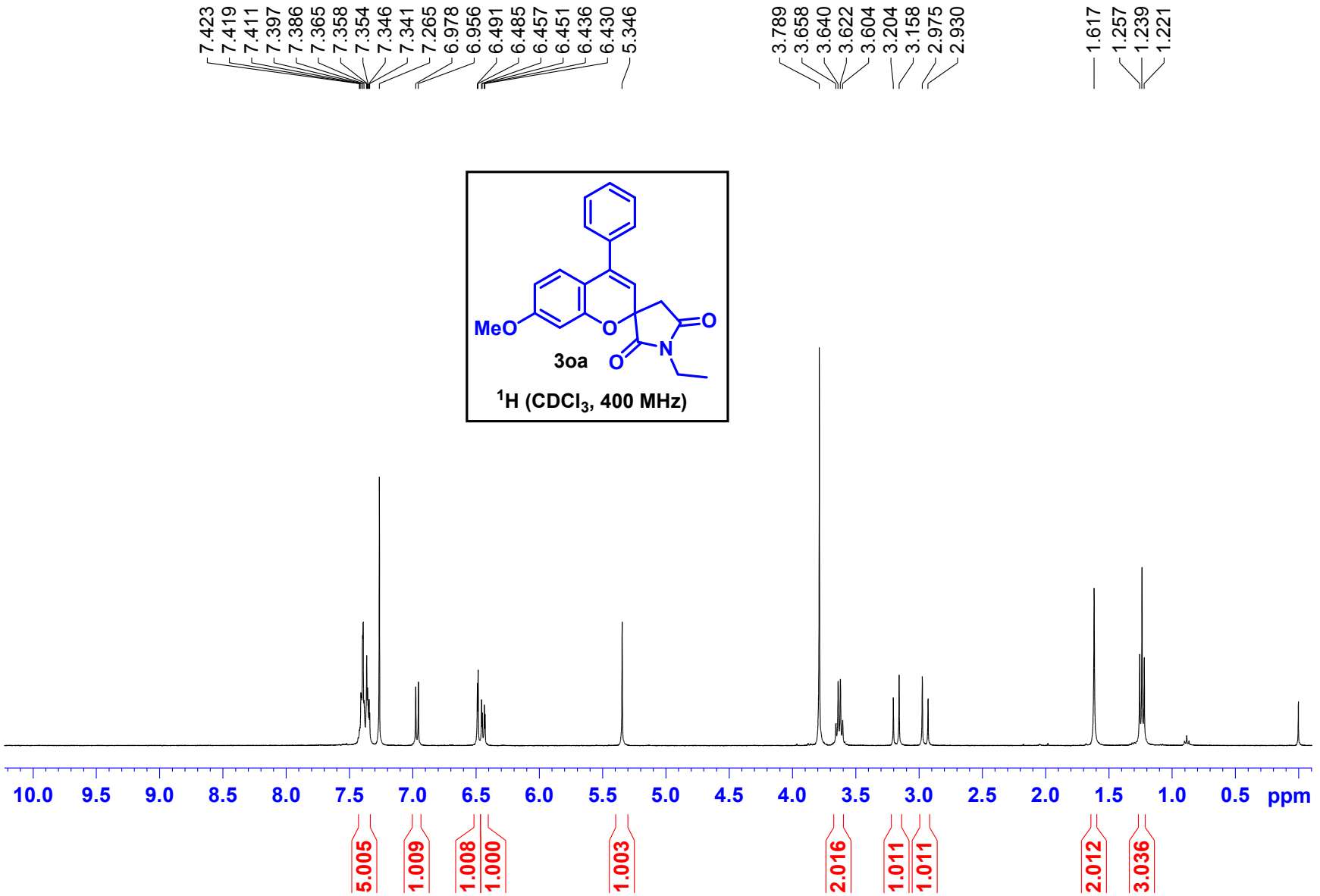

174.313
173.235
151.965
138.858
129.841
123.501
121.903
120.883
115.919
115.199
78.818
77.439
77.121
76.803
44.430
34.029
23.962
12.936
11.877
3ma
^{13}C (CDCl$_3$, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

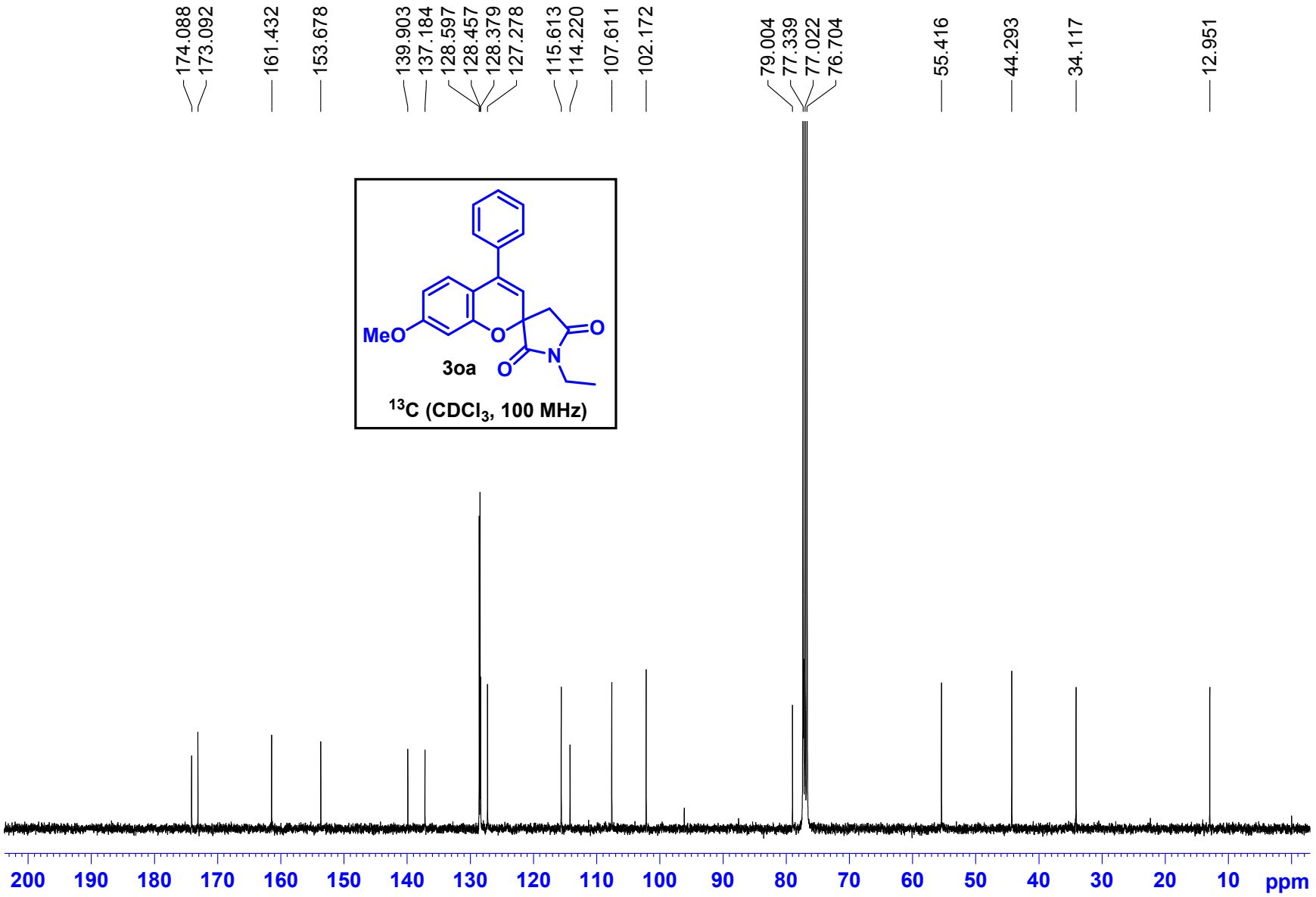

7.423
7.409
7.406
7.399
7.379
7.372
7.368
7.360
7.355
7.265
7.228
7.224
7.208
7.205
7.189
7.185
7.067
7.063
7.046
7.043
6.914
6.894
6.879
5.496
3.660
3.643
3.625
3.607
3.218
3.172
2.981
2.935
1.260
1.242
1.224

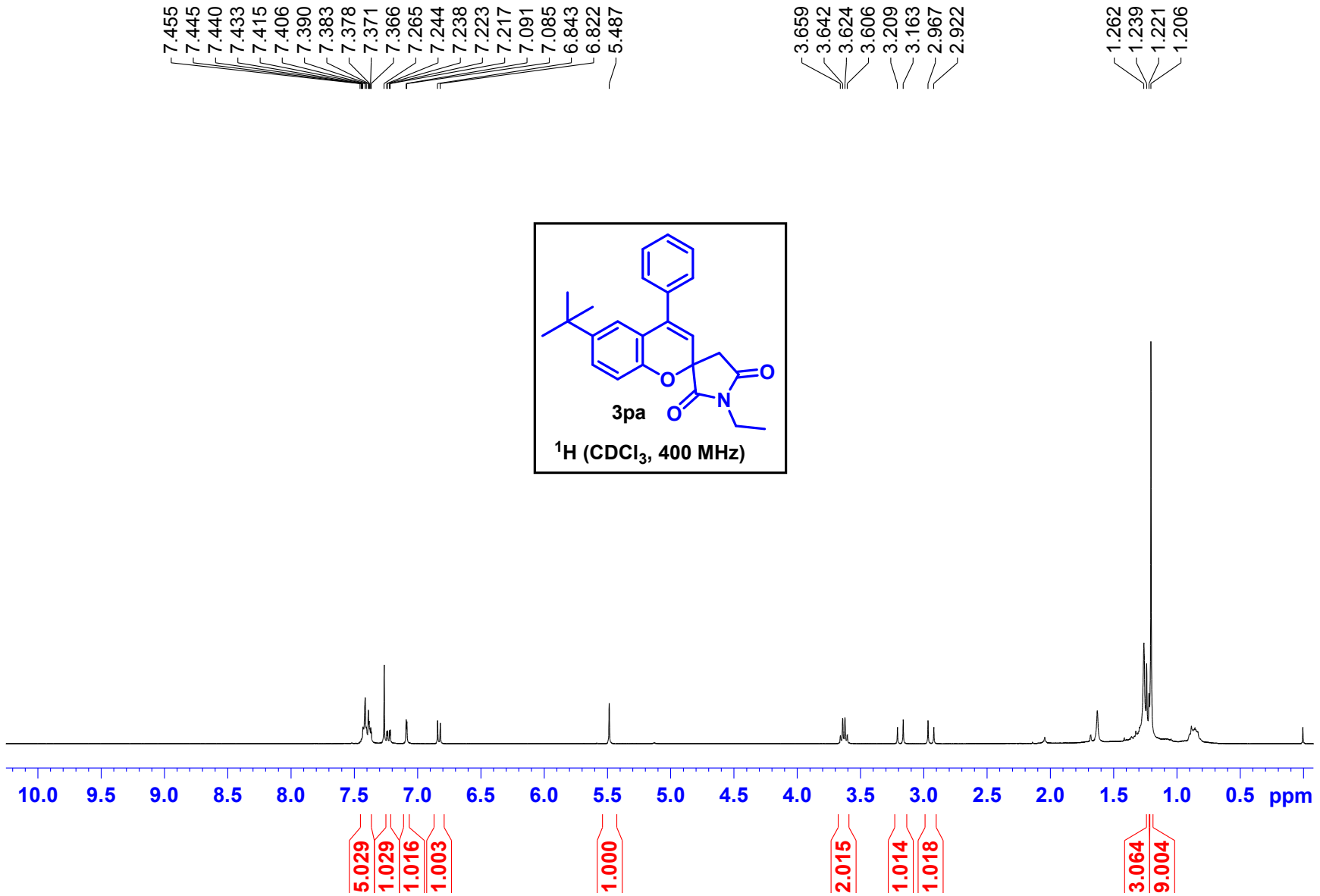

O
O
N
O
3na
^{1}H (CDCl$_3$, 400 MHz)

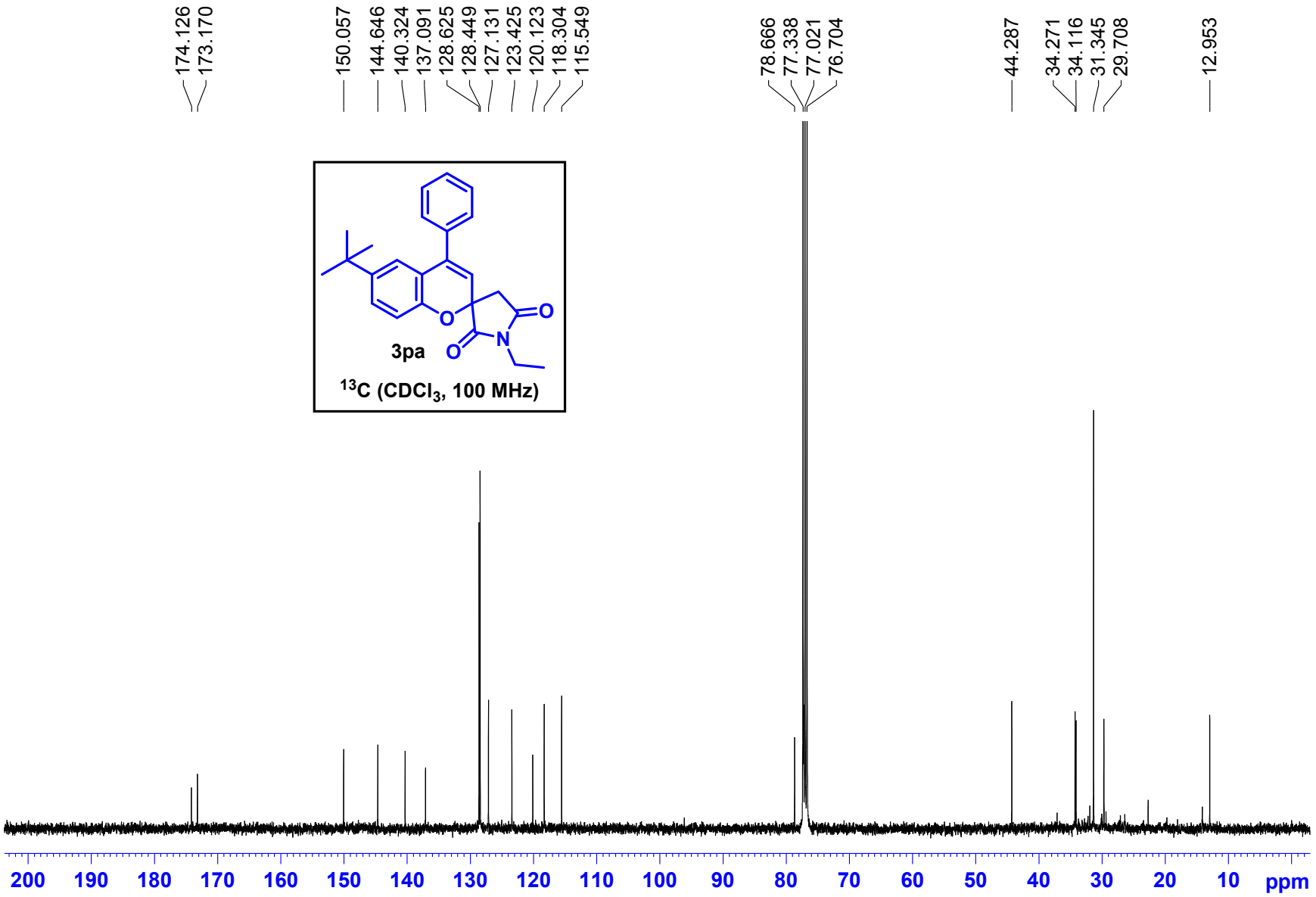

10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
5.065
1.024
1.016
2.019
0.959
2.026
1.000
1.008
3.001

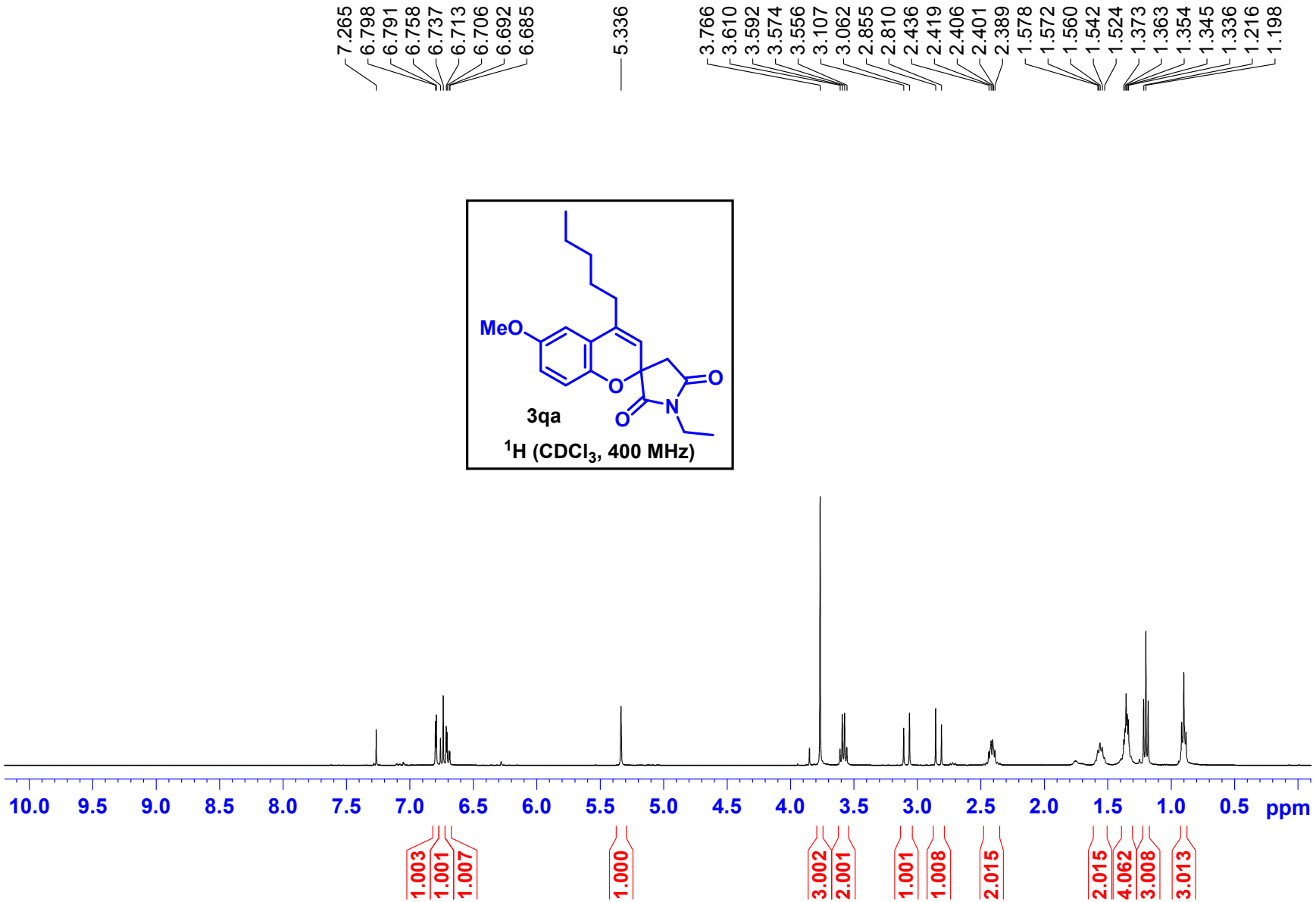

173.937
173.053
152.338
139.982
136.949
130.313
128.653
128.510
128.464
126.363
121.987
120.994
118.500
116.222
78.754
77.387
77.069
76.752
44.221
34.146
12.956
3na
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.423
7.419
7.411
7.397
7.386
7.365
7.358
7.354
7.346
7.341
7.265
6.978
6.956
6.491
6.485
6.457
6.451
6.436
6.430
5.346
3.789
3.658
3.640
3.622
3.604
3.204
3.158
2.975
2.930
1.617
1.257
1.239
1.221
MeO
O
O
N
O
3oa
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
5.005
1.009
1.008
1.000
1.003
2.016
1.011
1.011
2.012
3.036

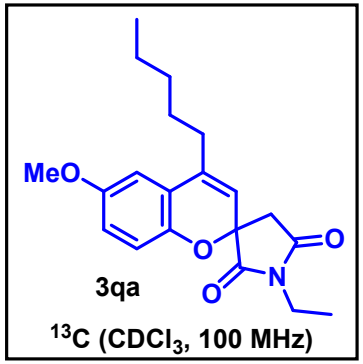

174.088
173.092
161.432
153.678
139.903
137.184
128.597
128.457
128.379
127.278
115.613
114.220
107.611
102.172
79.004
77.339
77.022
76.704
55.416
44.293
34.117
12.951
MeO
O
O
N
O
3oa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.455
7.445
7.440
7.433
7.415
7.406
7.390
7.383
7.378
7.371
7.366
7.265
7.244
7.238
7.223
7.217
7.091
7.085
6.843
6.822
5.487
3.659
3.642
3.624
3.606
3.209
3.163
2.967
2.922
1.262
1.239
1.221
1.206
3pa
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
5.029
1.029
1.016
1.003
1.000
2.015
1.014
1.018
3.064
9.004


174.126
173.170
150.057
144.646
140.324
137.091
128.625
128.449
127.131
123.425
120.123
118.304
115.549
78.666
77.338
77.021
76.704
44.287
34.271
34.116
31.345
29.708
12.953
O
O
N
O
3pa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

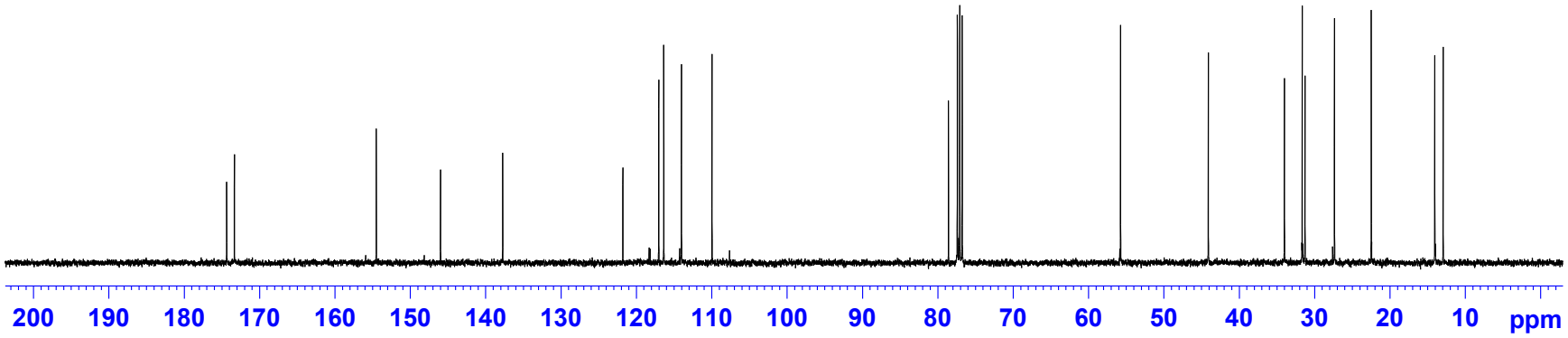

7.265
6.798
6.791
6.758
6.737
6.713
6.706
6.692
6.685
5.336
3.766
3.610
3.592
3.574
3.556
3.107
3.062
2.855
2.810
2.436
2.419
2.406
2.401
2.389
1.578
1.572
1.560
1.542
1.524
1.373
1.363
1.354
1.345
1.336
1.216
1.198
MeO
O
O
N
O
3qa
^{1}H ($CDCl_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.003
1.001
1.007
1.000
3.002
2.001
1.001
1.008
2.015
2.015
4.062
3.008
3.013

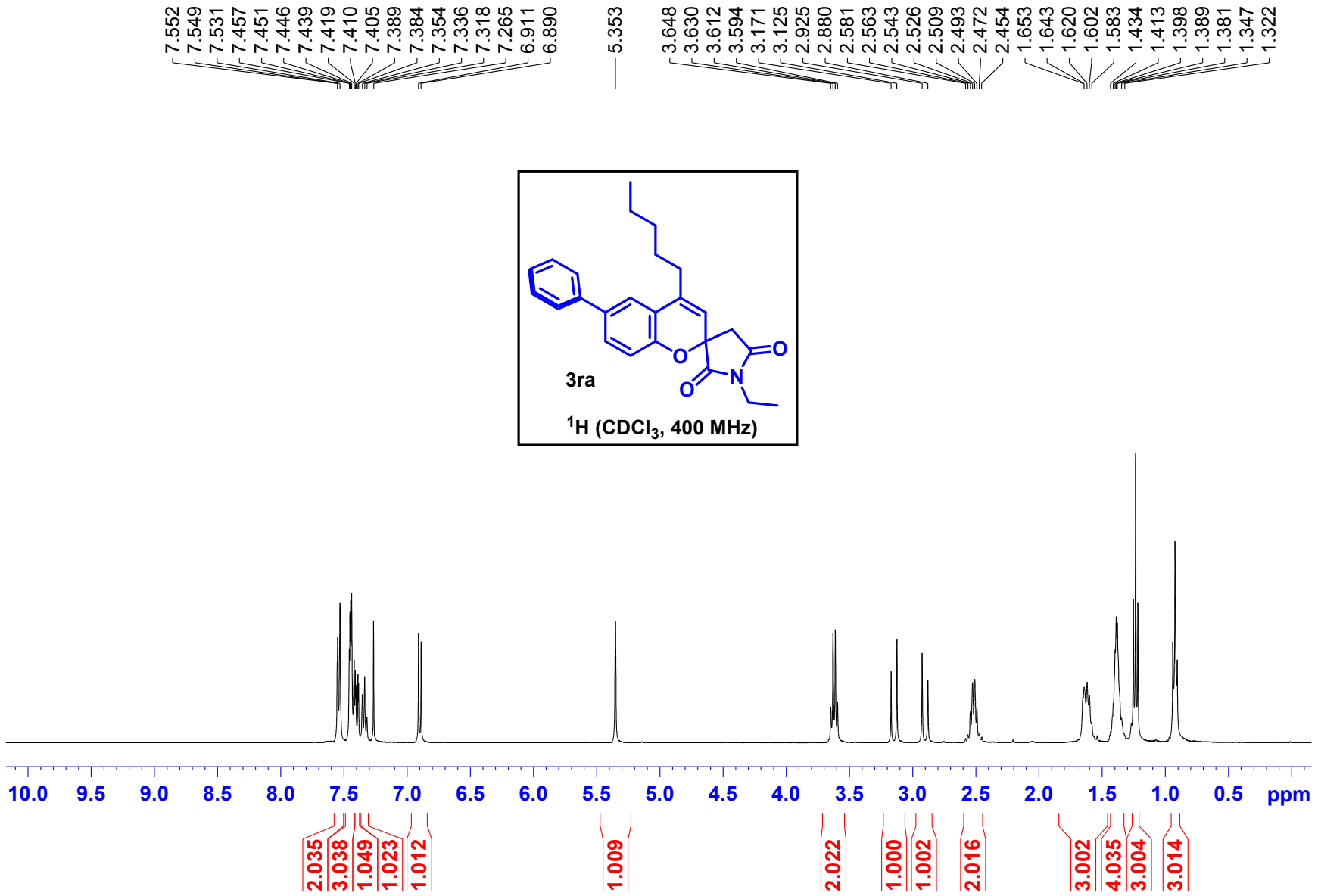
174.345
173.321
154.492
145.958
137.748
121.801
117.035
116.368
114.028
109.970
78.584
77.405
77.088
76.770
55.742
44.099
34.005
31.642
31.270
27.359
22.478
14.046
12.936

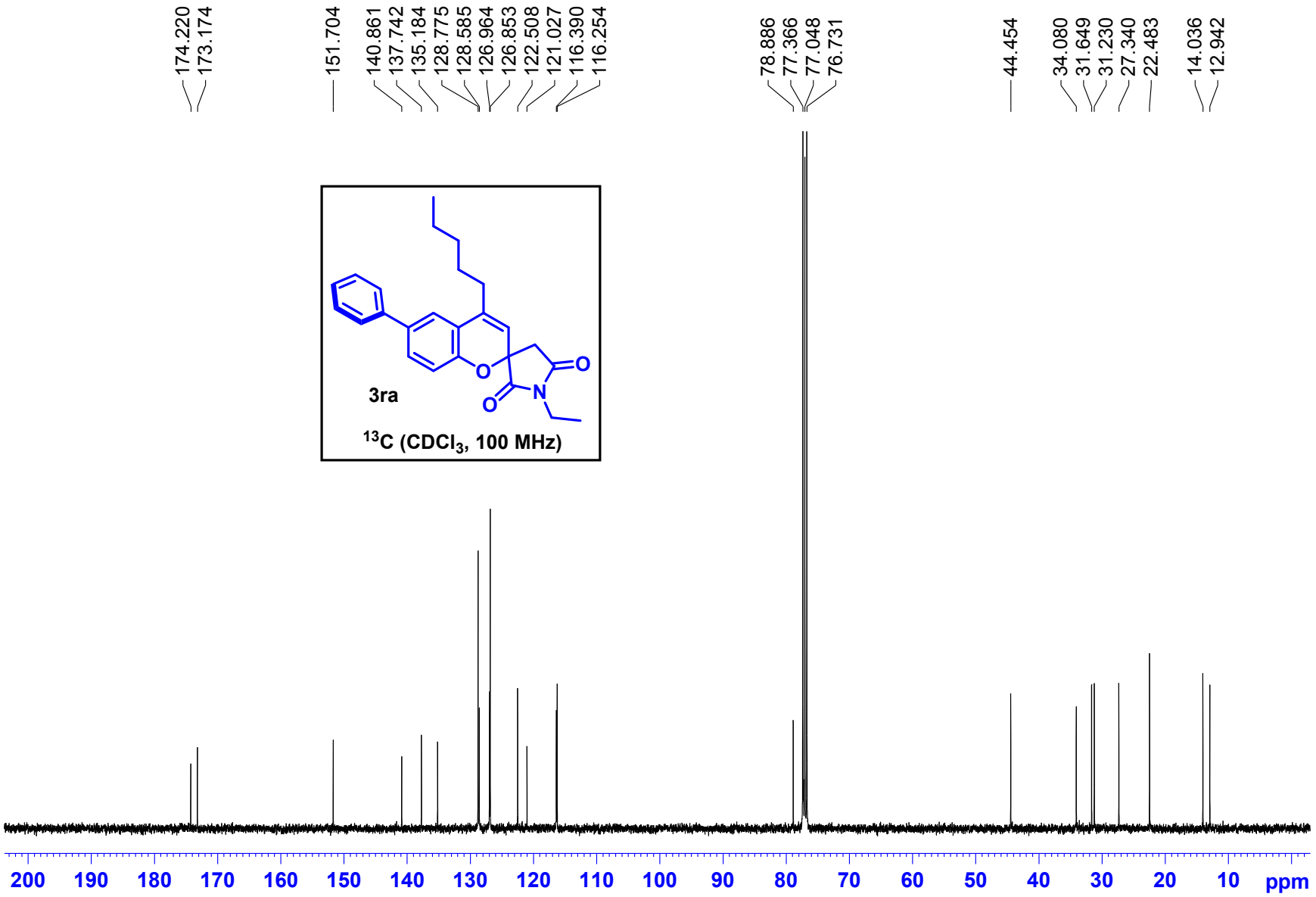


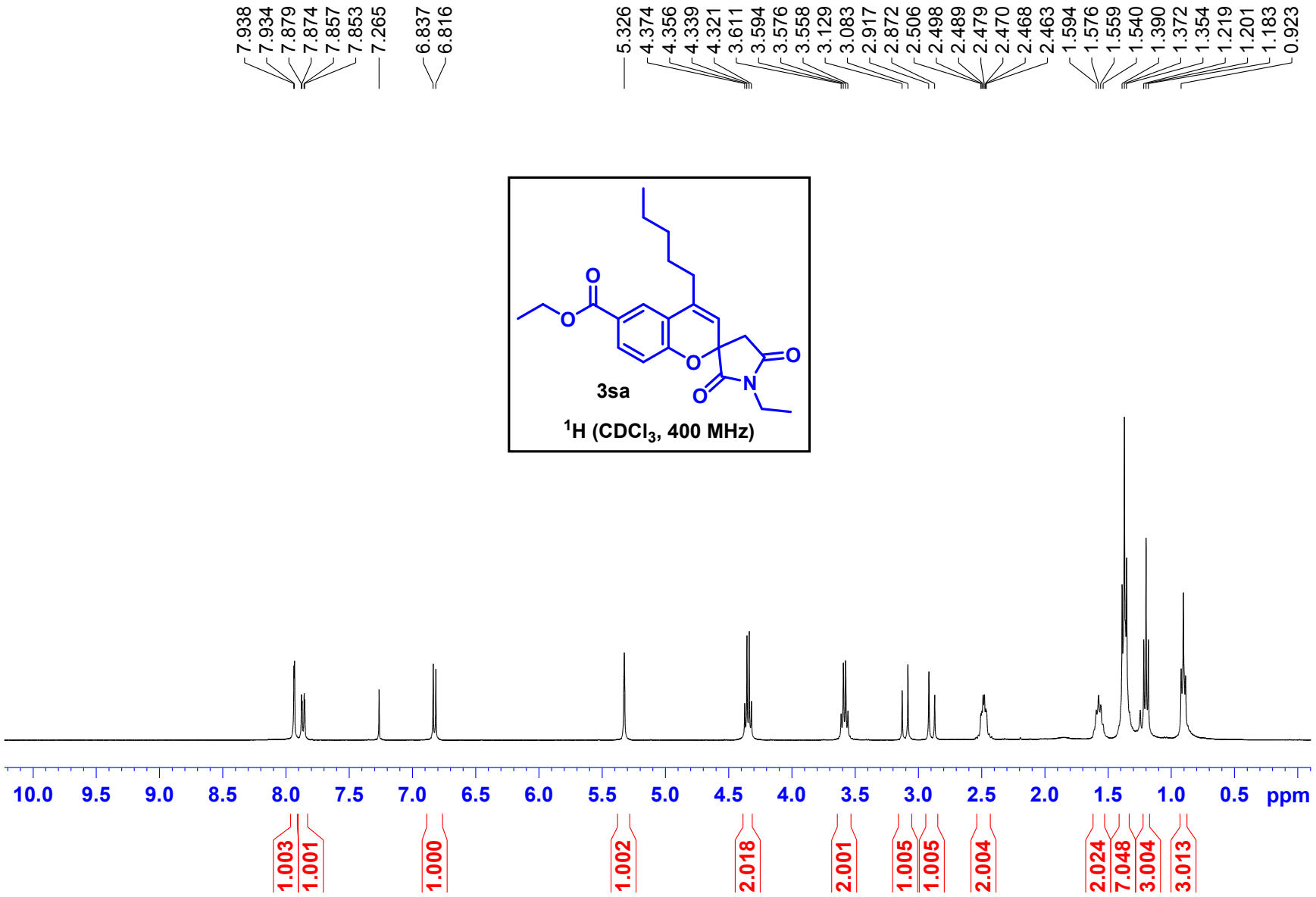
3qa

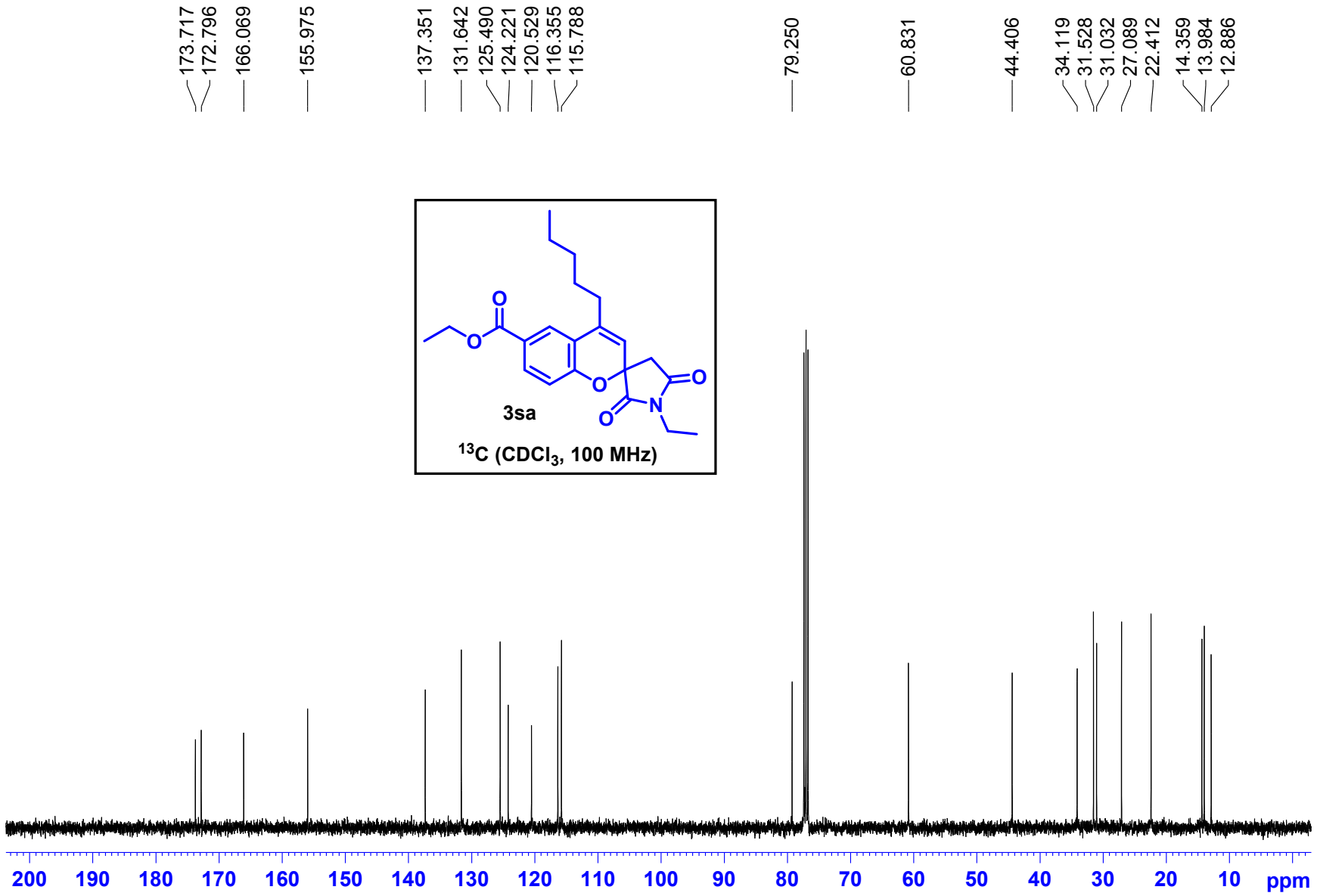
^{13}C ($CDCl_3$, 100 MHz)

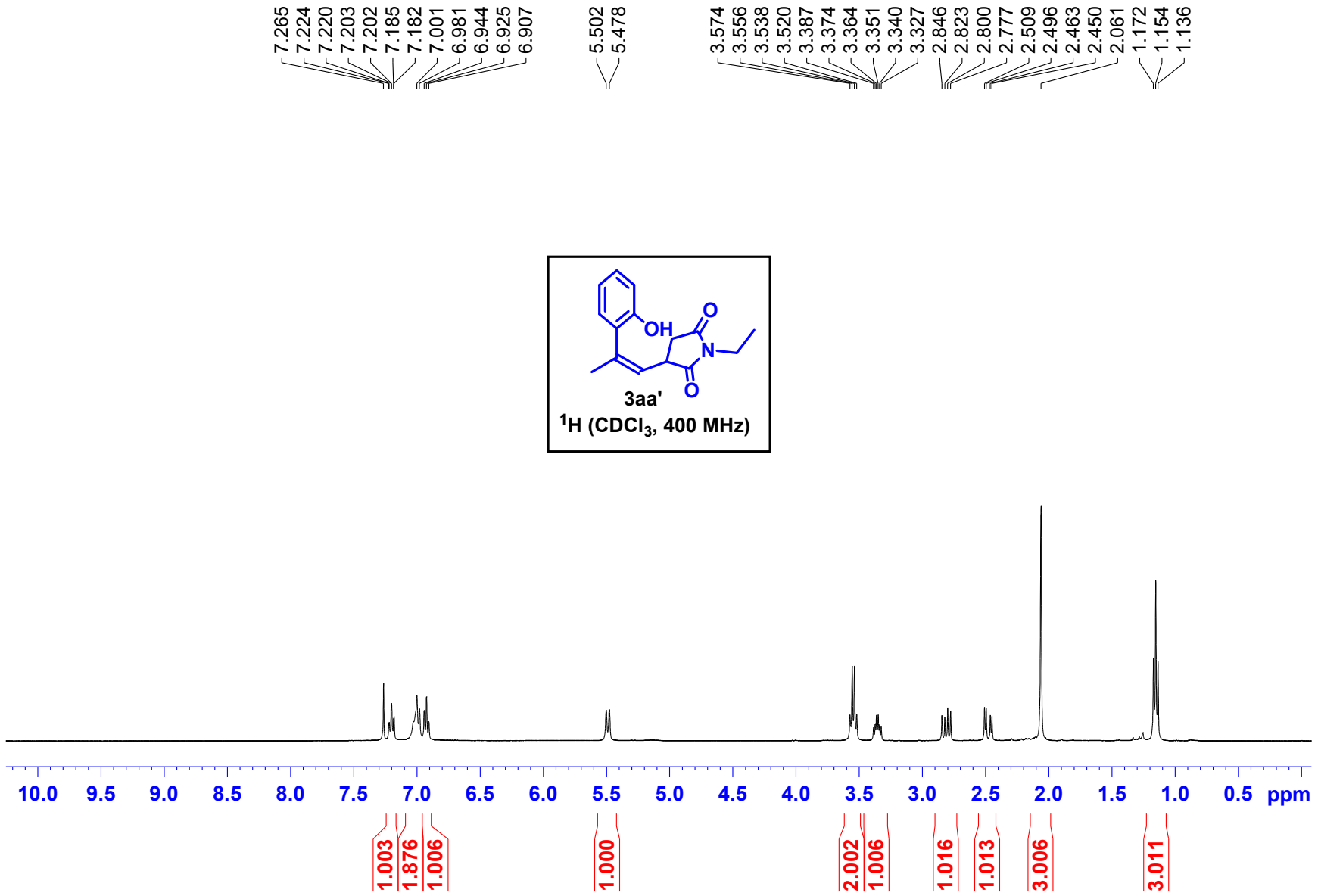


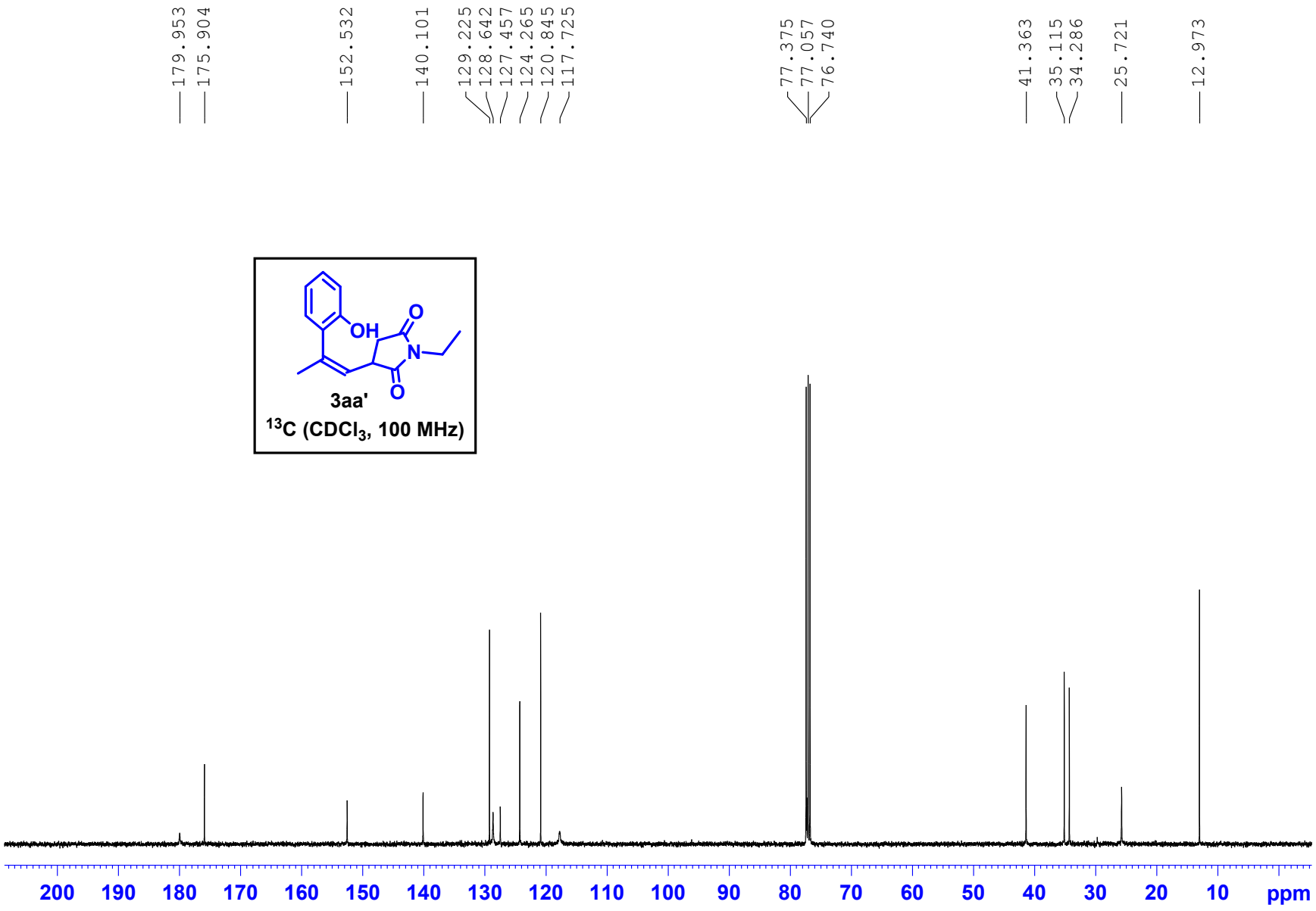

7.552
7.549
7.531
7.457
7.451
7.446
7.439
7.419
7.410
7.405
7.389
7.384
7.354
7.336
7.318
7.265
6.911
6.890
5.353
3.648
3.630
3.612
3.594
3.171
3.125
2.925
2.880
2.581
2.563
2.543
2.526
2.509
2.493
2.472
2.454
1.653
1.643
1.620
1.602
1.583
1.434
1.413
1.398
1.389
1.381
1.347
1.322
O
O
N
O
3ra
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
2.035
3.038
1.049
1.023
1.012
1.009
2.022
1.000
1.002
2.016
3.002
4.035
3.004
3.014

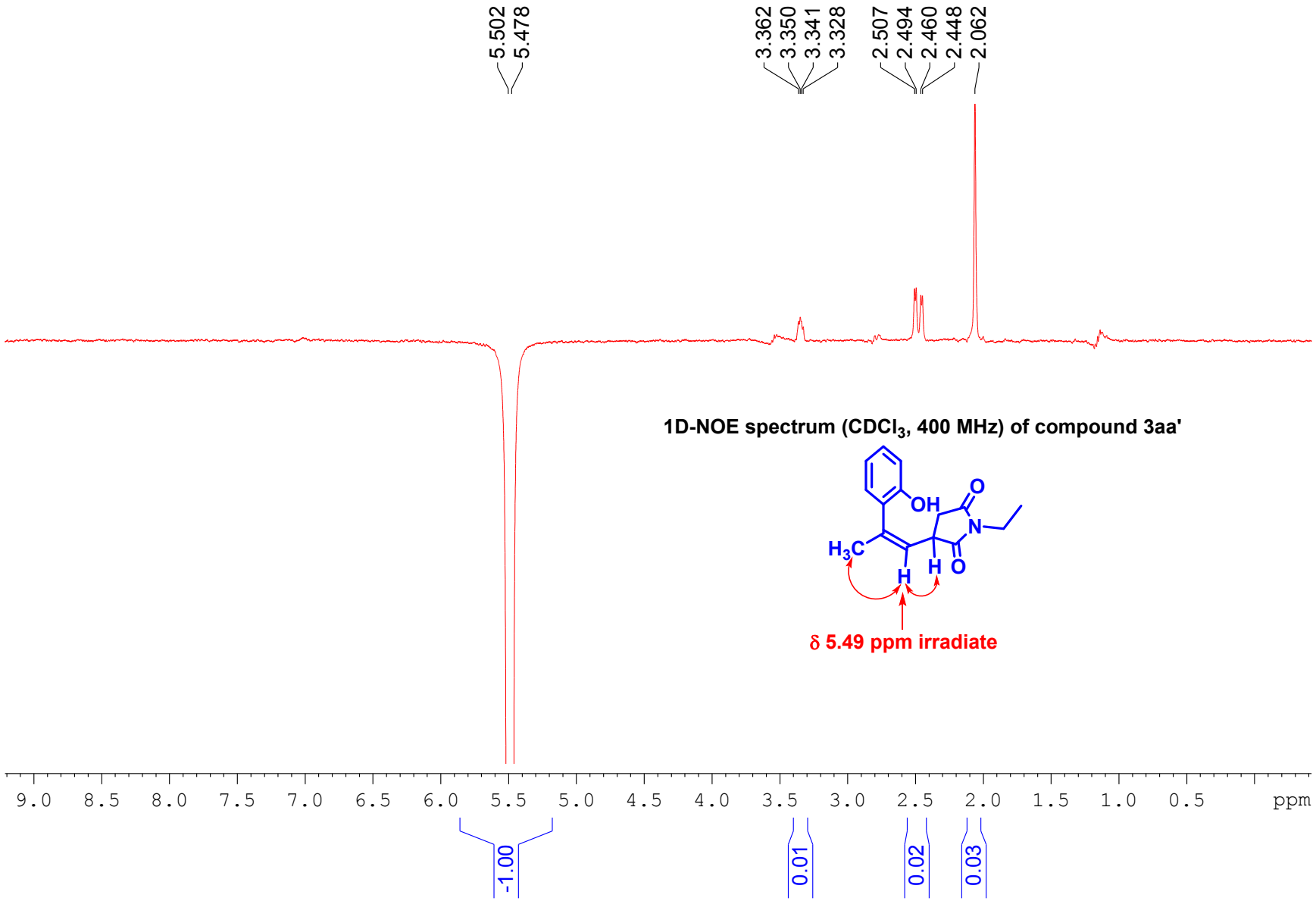

174.220
173.174
151.704
140.861
137.742
135.184
128.775
128.585
126.964
126.853
122.508
121.027
116.390
116.254
78.886
77.366
77.048
76.731
44.454
34.080
31.649
31.230
27.340
22.483
14.036
12.942
3ra
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

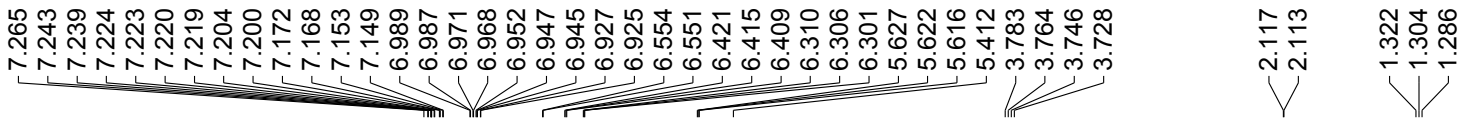

7.938
7.934
7.879
7.874
7.857
7.853
7.265
6.837
6.816
5.326
4.374
4.356
4.339
4.321
3.611
3.594
3.576
3.558
3.129
3.083
2.917
2.872
2.506
2.498
2.489
2.479
2.470
2.468
2.463
1.594
1.576
1.559
1.540
1.390
1.372
1.354
1.219
1.201
1.183
0.923
O
O
O
O
N
O
3sa
^{1}H (CDCl$_3$, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.003
1.001
1.000
1.002
2.018
2.001
1.005
1.005
2.004
2.024
7.048
3.004
3.013

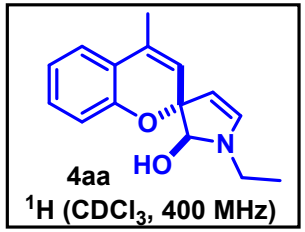

173.717
172.796
166.069
155.975
137.351
131.642
125.490
124.221
120.529
116.355
115.788
79.250
60.831
44.406
34.119
31.528
31.032
27.089
22.412
14.359
13.984
12.886
3sa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.265
7.224
7.220
7.203
7.202
7.185
7.182
7.001
6.981
6.944
6.925
6.907
5.502
5.478
3.574
3.556
3.538
3.520
3.387
3.374
3.364
3.351
3.340
3.327
2.846
2.823
2.800
2.777
2.509
2.496
2.463
2.450
2.061
1.172
1.154
1.136
OH
O
N
O
3aa'
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.003
1.876
1.006
1.000
2.002
1.006
1.016
1.013
3.006
3.011


179.953
175.904
152.532
140.101
129.225
128.642
127.457
124.265
120.845
117.725
77.375
77.057
76.740
41.363
35.115
34.286
25.721
12.973
OH
O
N
O
3aa'
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

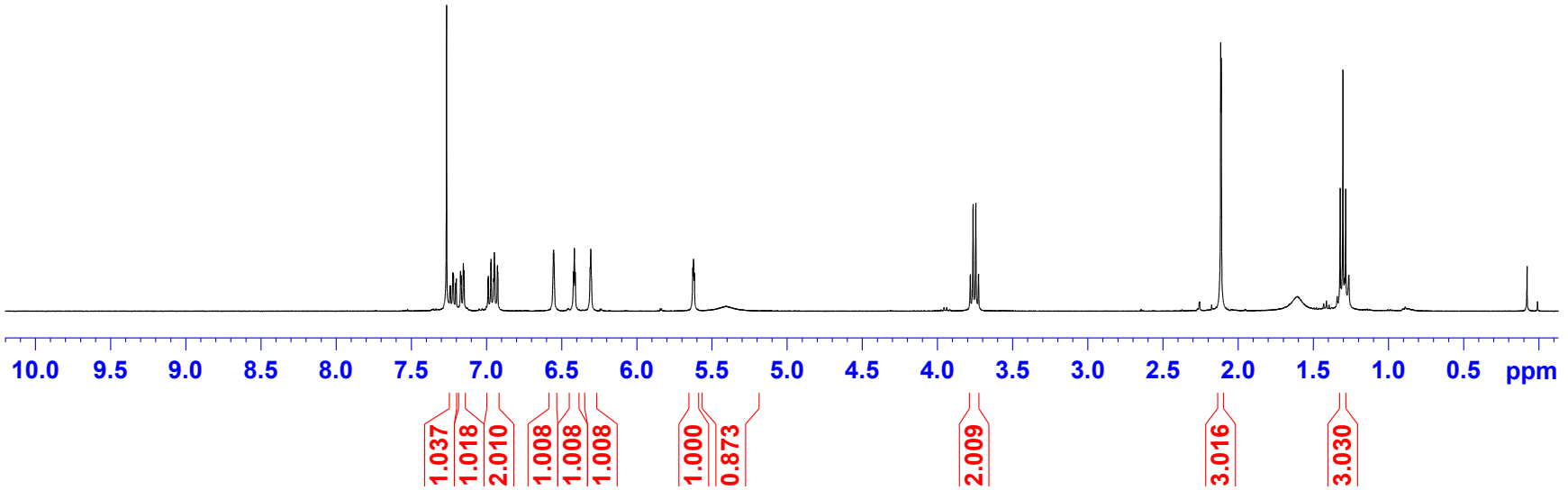

5.502
5.478
3.362
3.350
3.341
3.328
2.507
2.494
2.460
2.448
2.062
1D-NOE spectrum (CDCl3, 400 MHz) of compound 3aa'
OH
O
N
H3C
H
H
O
δ 5.49 ppm irradiate
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
-1.00
0.01
0.02
0.03

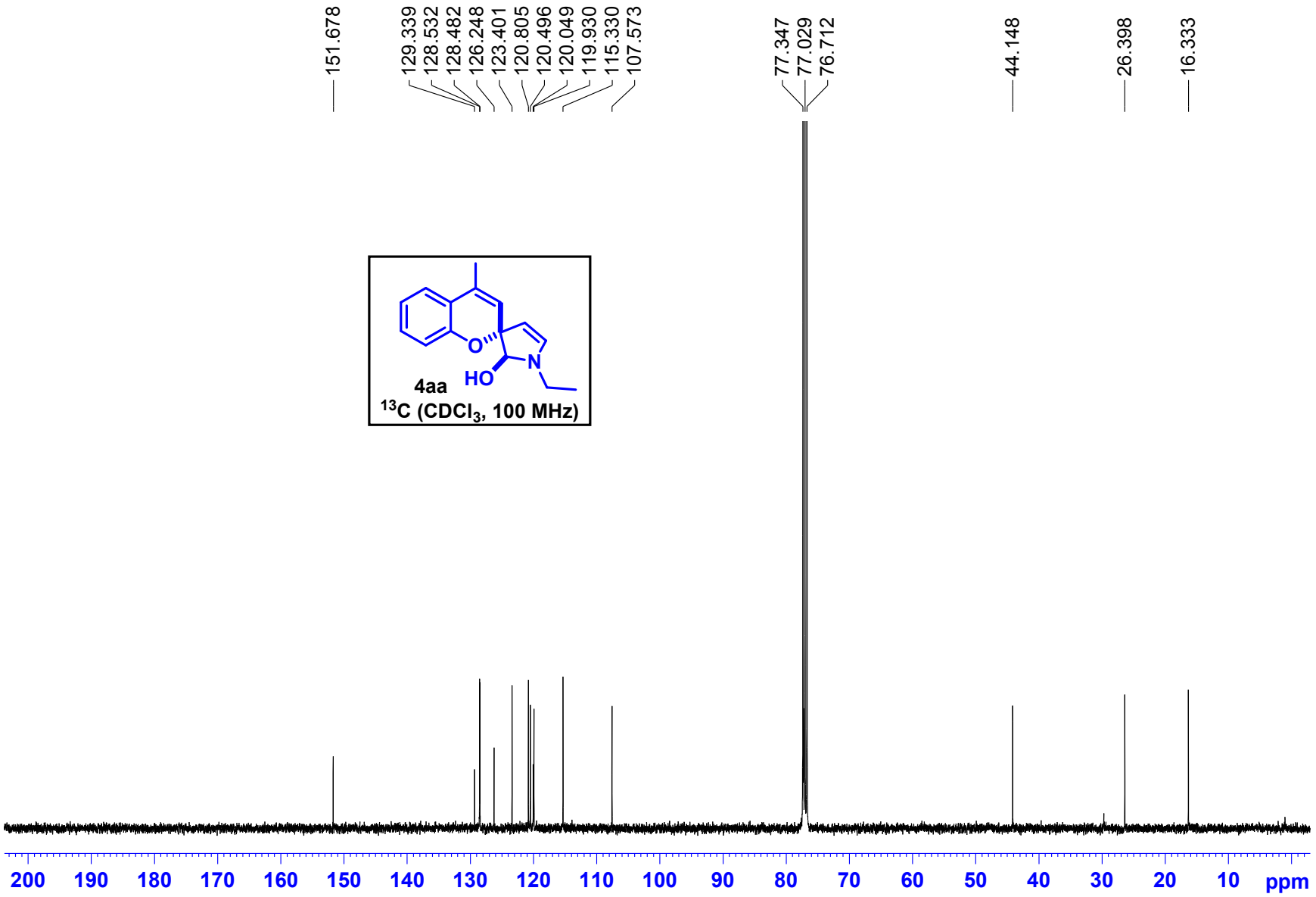


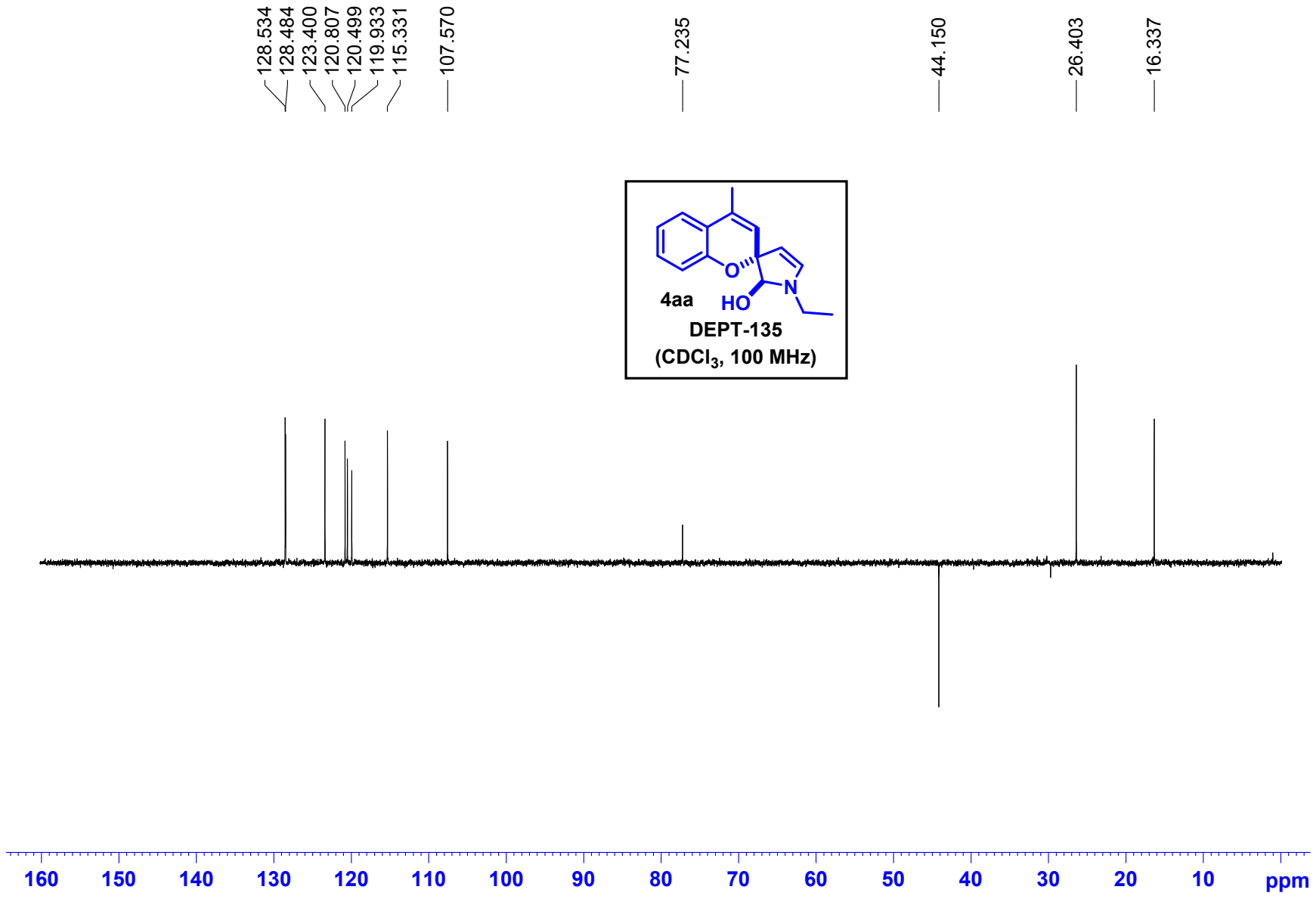


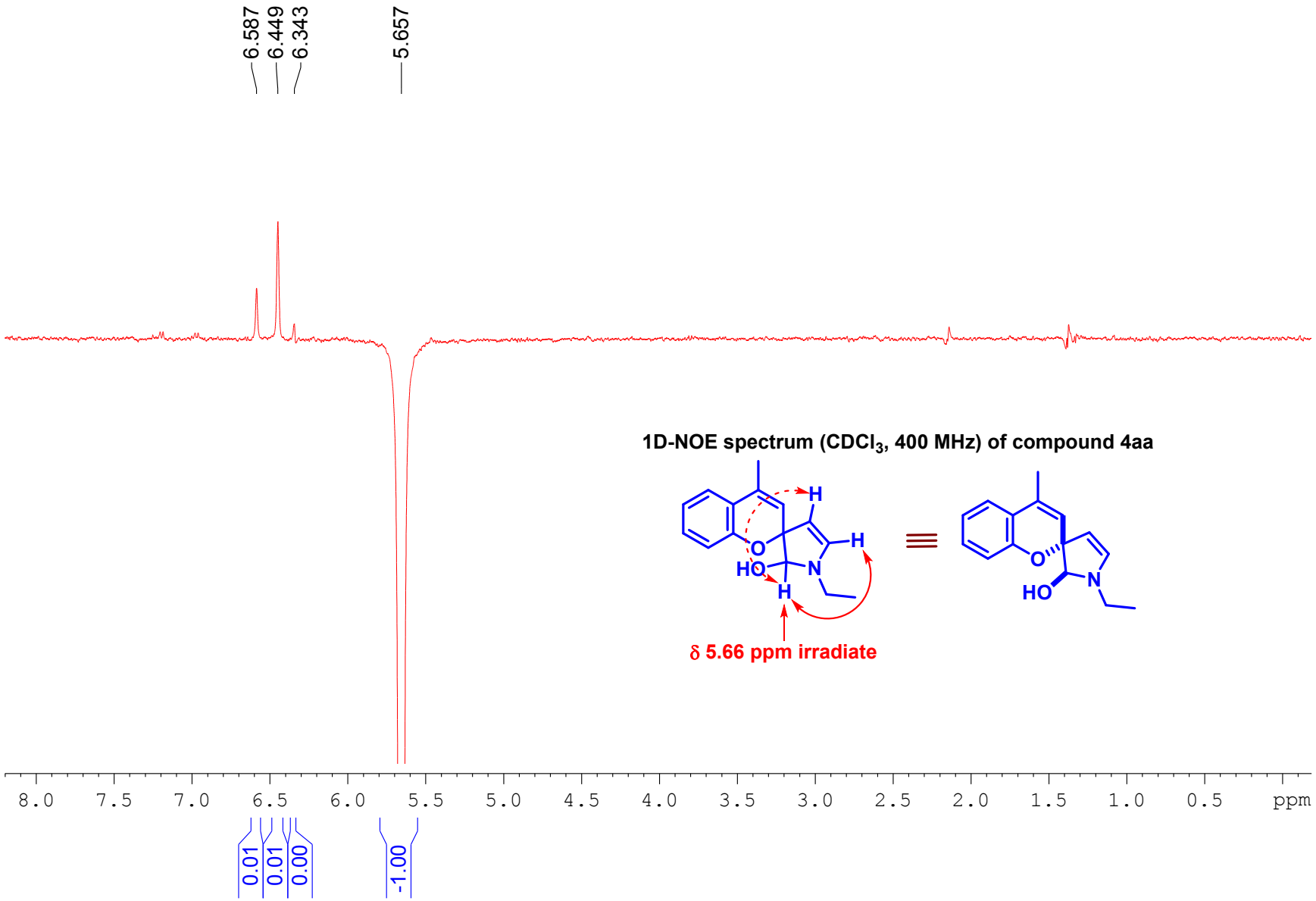
4aa

^{1}H ($CDCl_3$, 400 MHz)

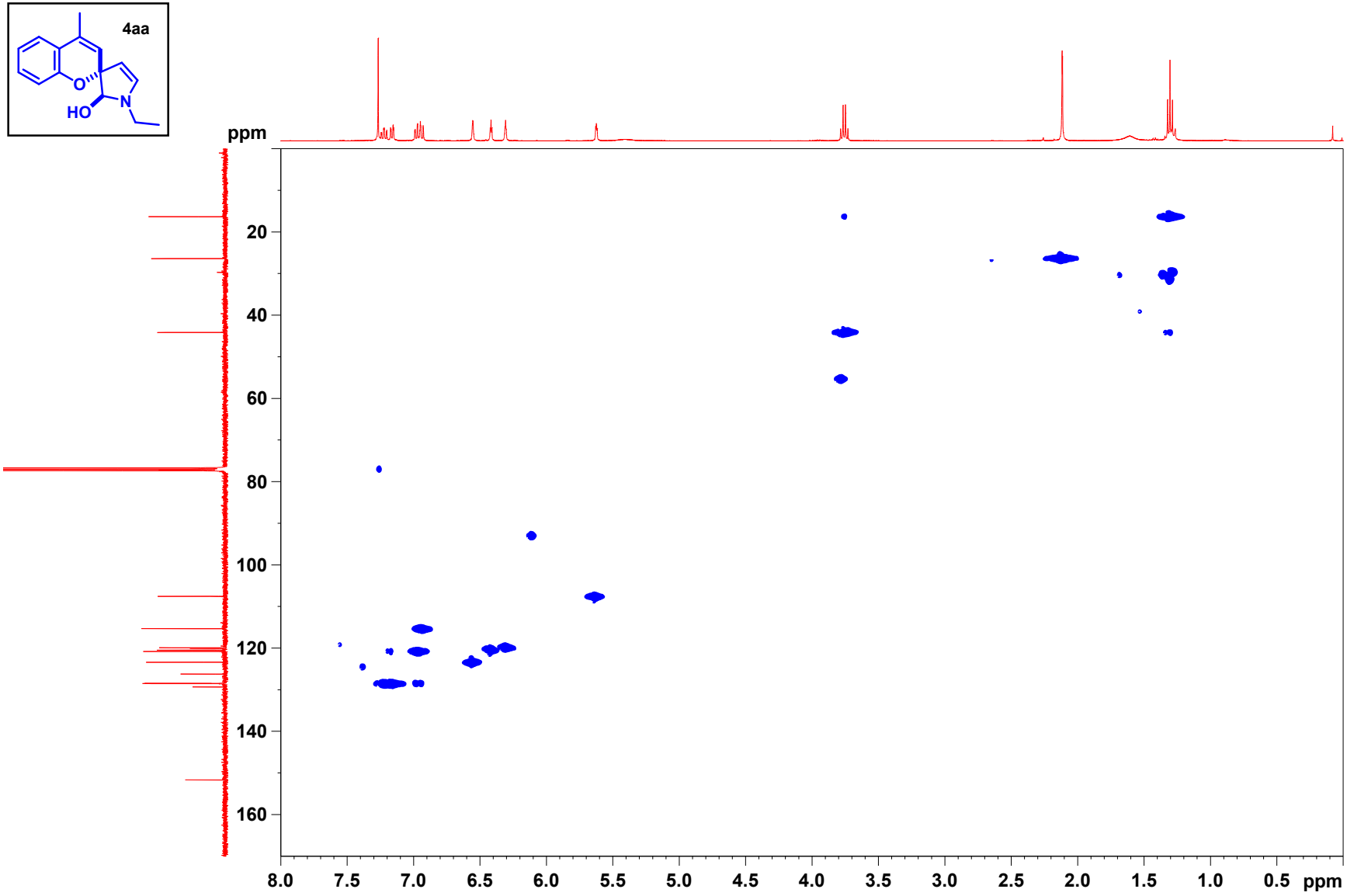



151.678
129.339
128.532
128.482
126.248
123.401
120.805
120.496
120.049
119.930
115.330
107.573
77.347
77.029
76.712
44.148
26.398
16.333
O
N
HO
4aa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

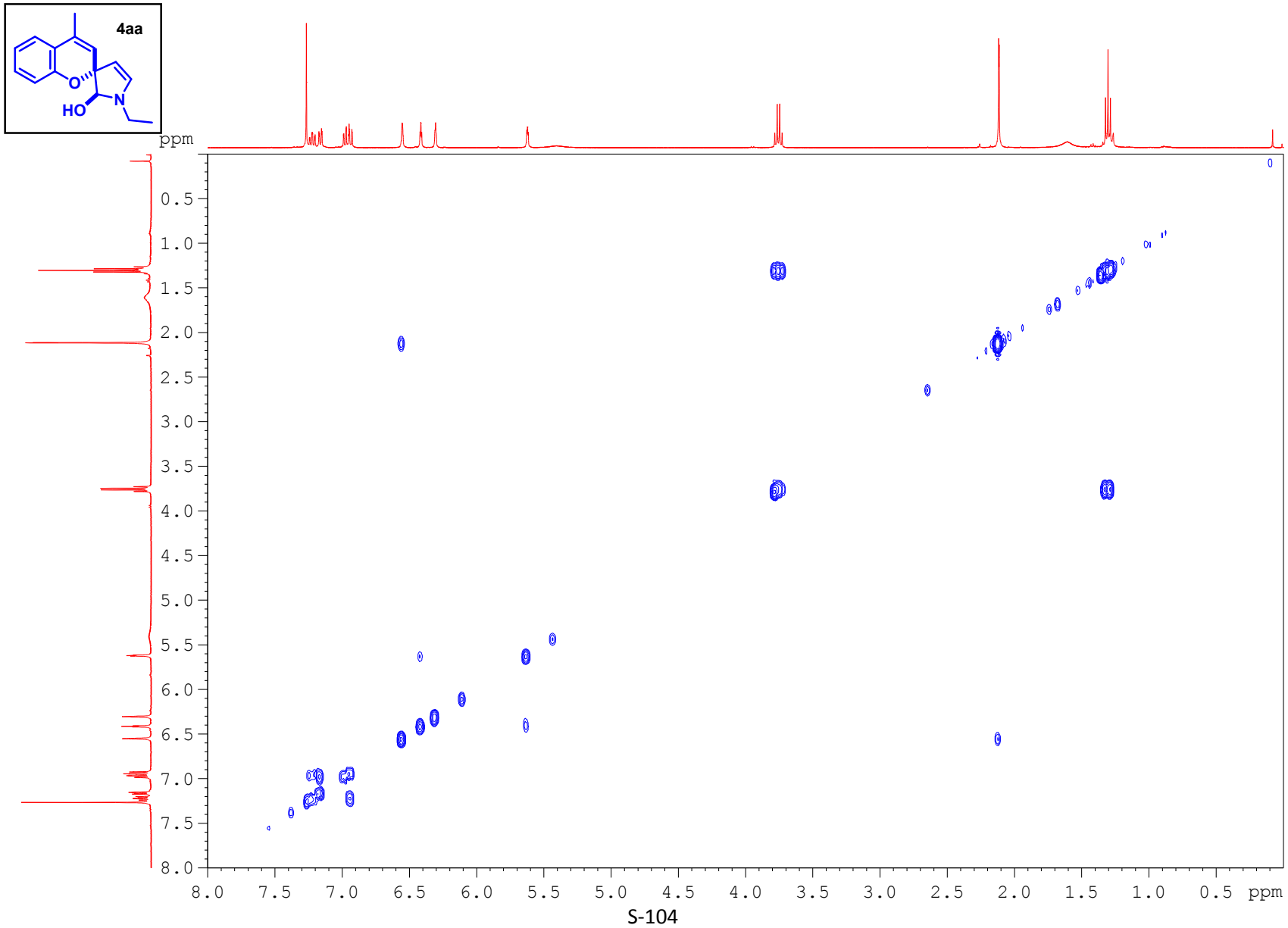

128.534
128.484
123.400
120.807
120.499
119.933
115.331
107.570
77.235
44.150
26.403
16.337
4aa
HO
O
N
DEPT-135
(CDCl3, 100 MHz)
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


6.587
6.449
6.343
5.657
1D-NOE spectrum (CDCl3, 400 MHz) of compound 4aa
H
H
O
HO
N
H
δ 5.66 ppm irradiate
HO
N
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
0.01
0.01
0.00
-1.00

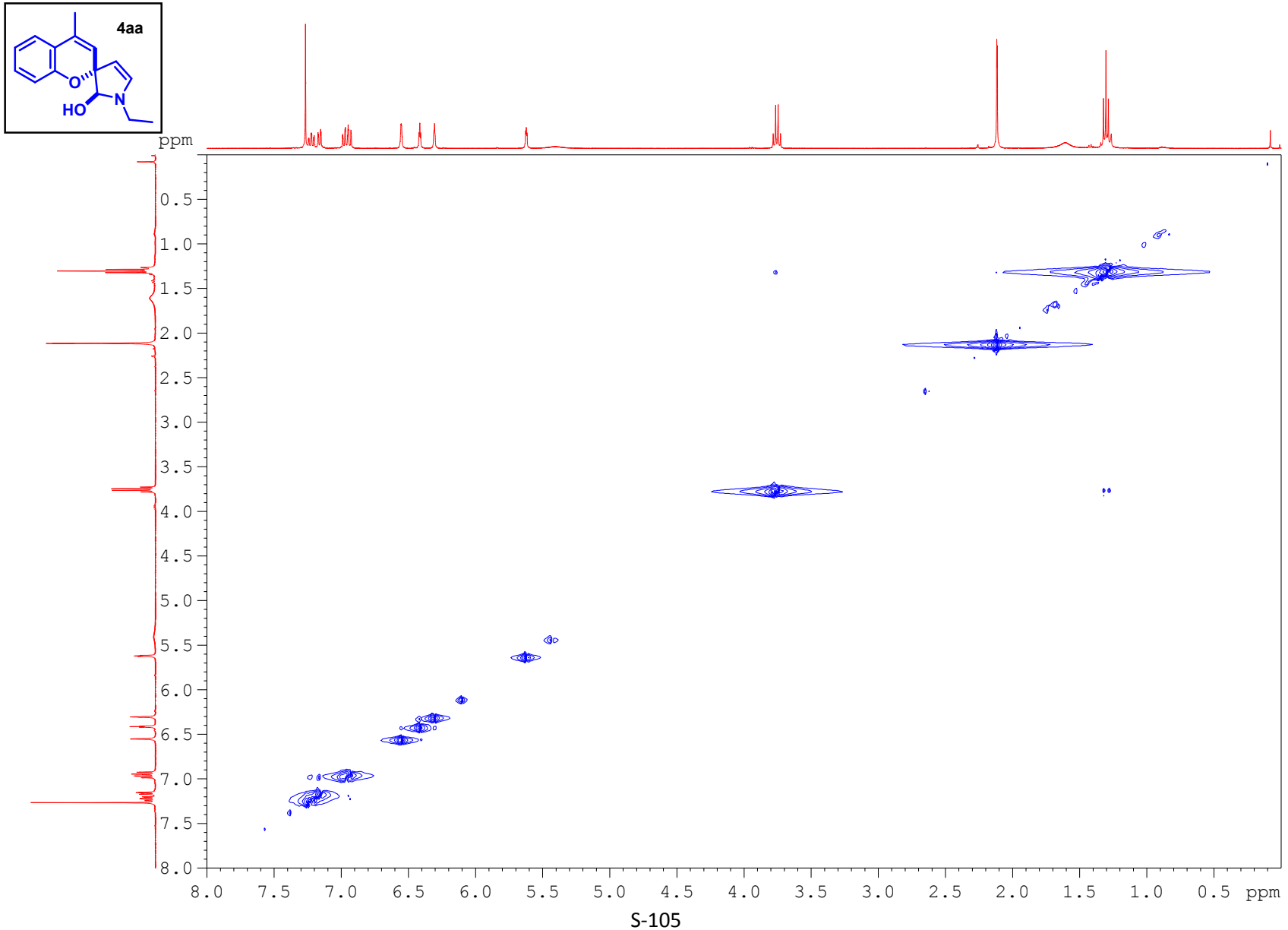
HSQC spectrum ($CDCl_3$, 400 MHz) of compound 4aa

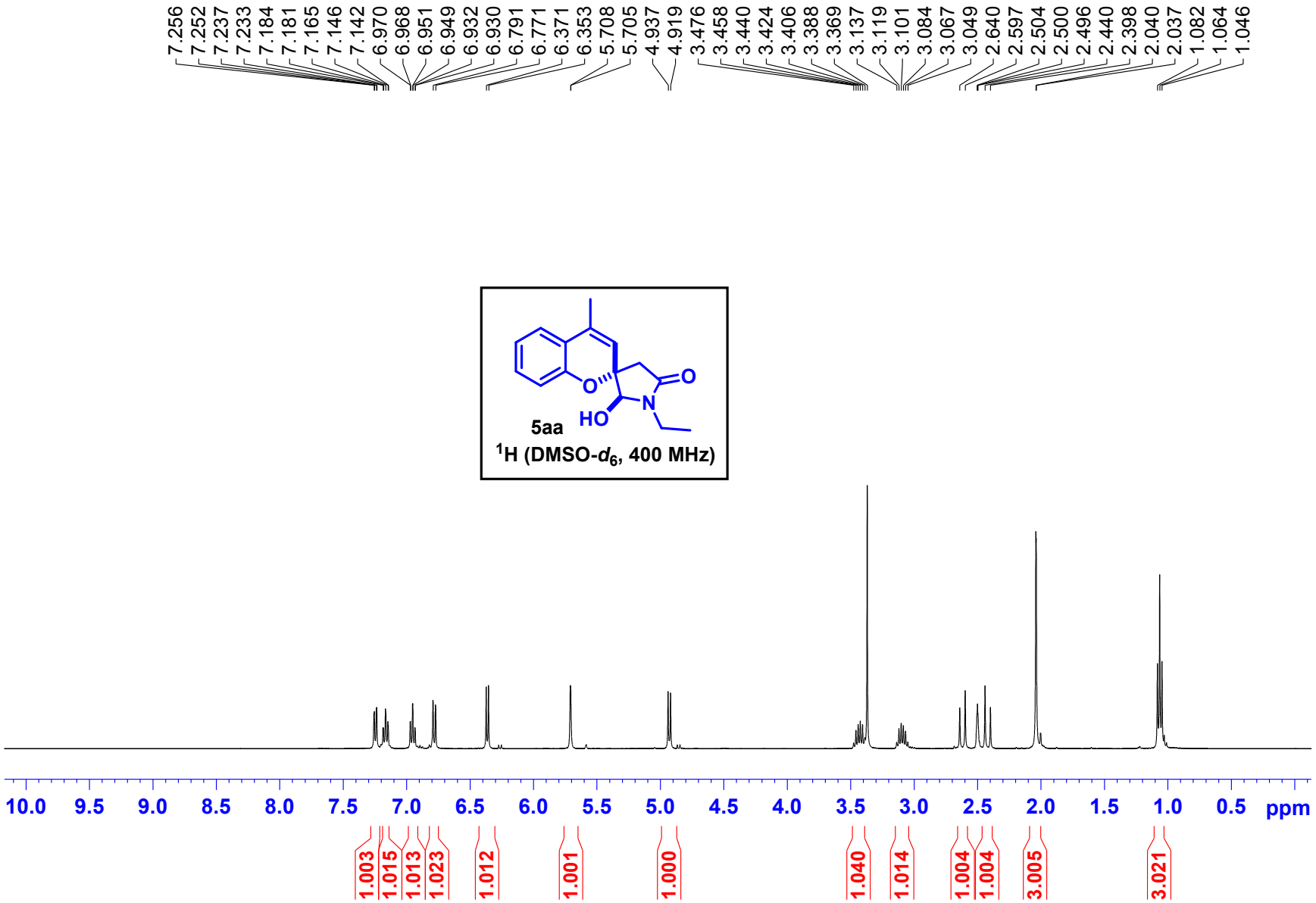


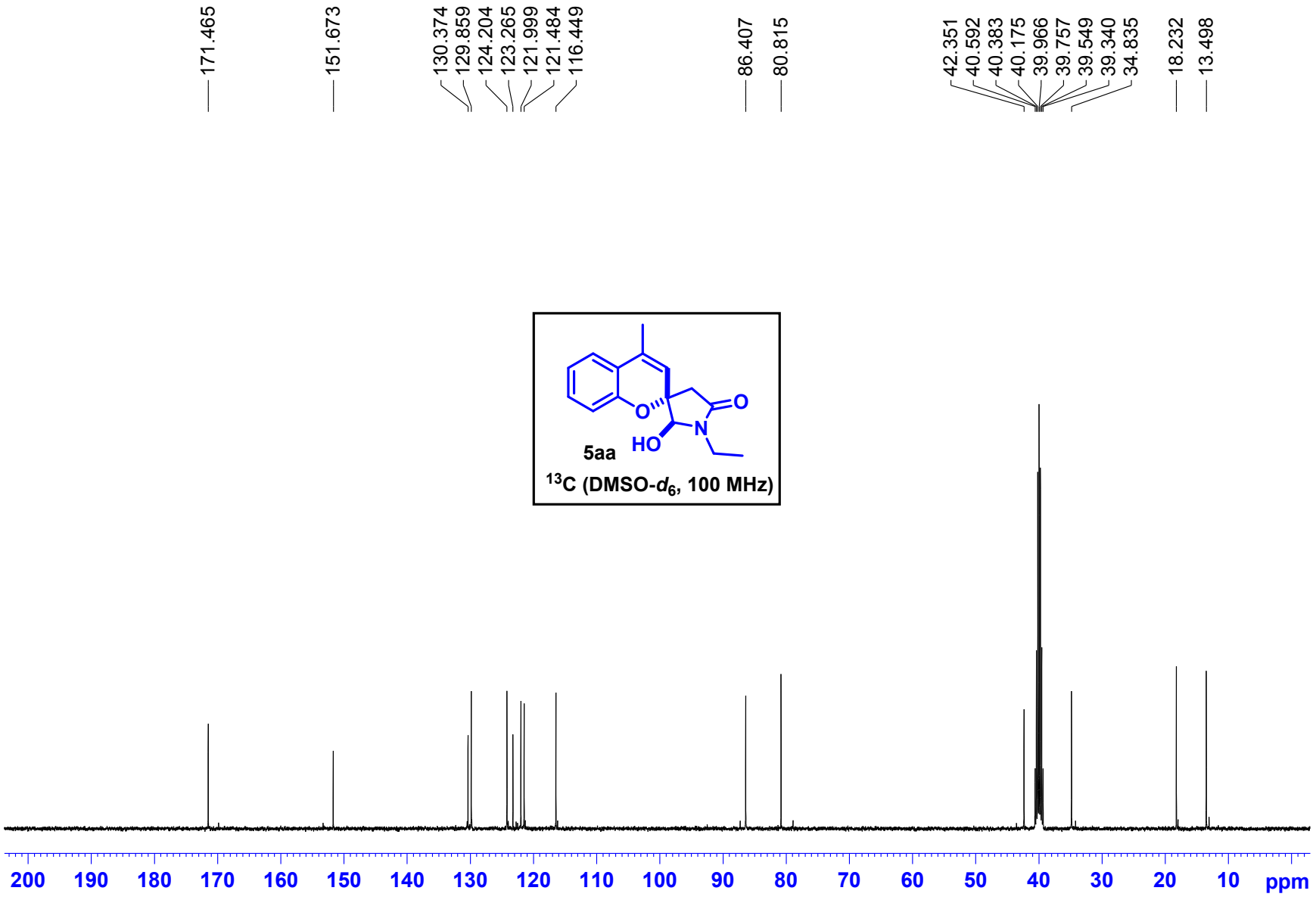
COSY spectrum ($CDCl_3$, 400 MHz) of compound 4aa

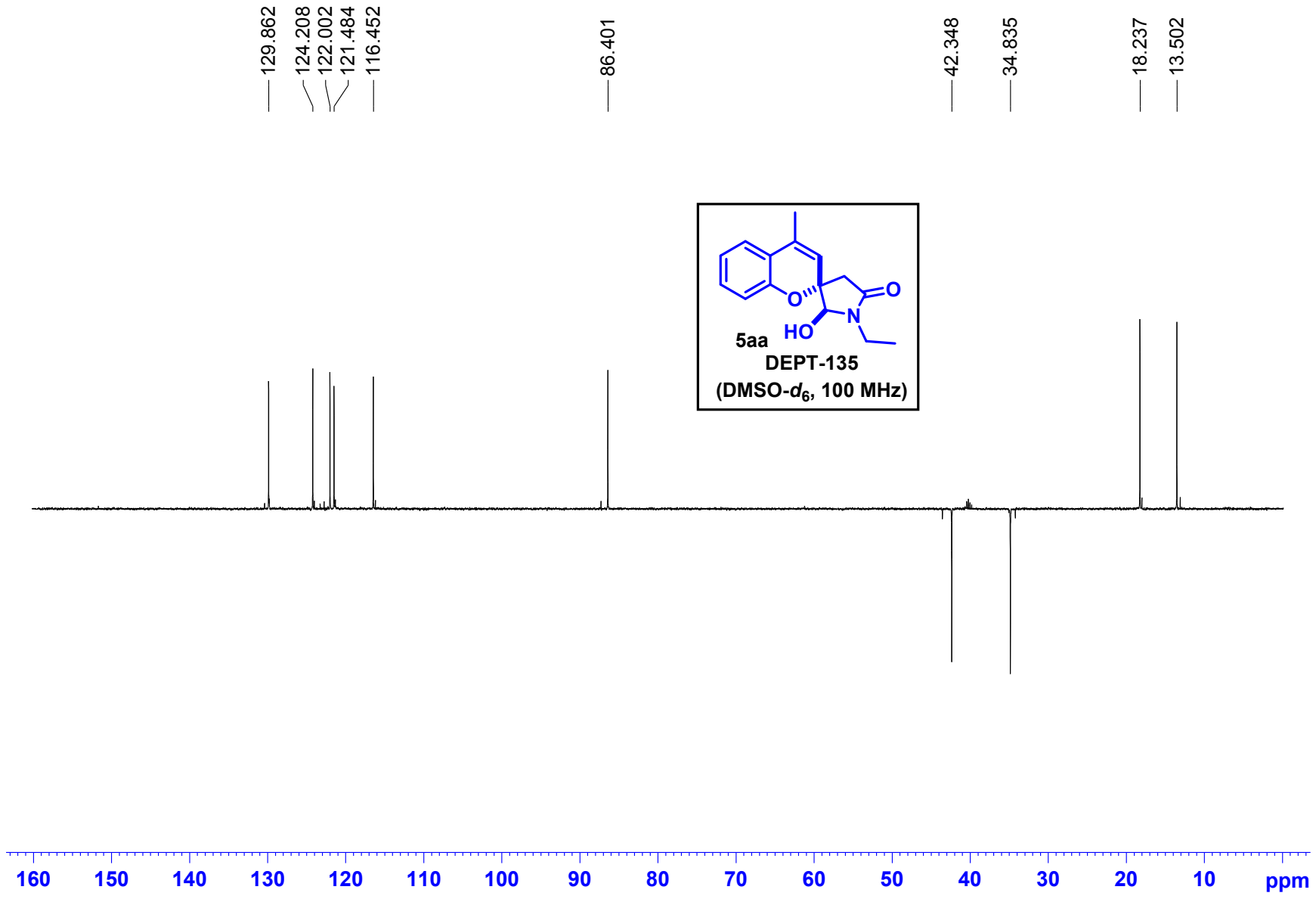


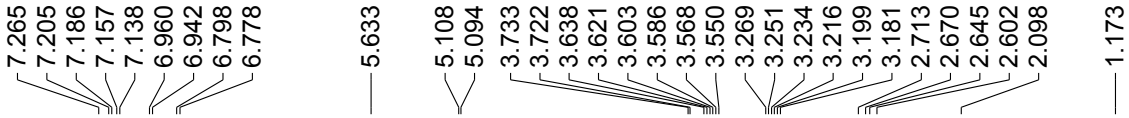
NOSY spectrum ($CDCl_3$, 400 MHz) of compound 4aa

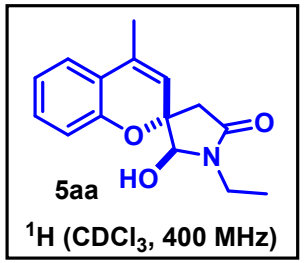



7.256
7.252
7.237
7.233
7.184
7.181
7.165
7.146
7.142
6.970
6.968
6.951
6.949
6.932
6.930
6.791
6.771
6.371
6.353
5.708
5.705
4.937
4.919
3.476
3.458
3.440
3.424
3.406
3.388
3.369
3.137
3.119
3.101
3.084
3.067
3.049
2.640
2.597
2.504
2.500
2.496
2.440
2.398
2.040
2.037
1.082
1.064
1.046
O
O
N
HO
5aa
1H (DMSO-d6, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.003
1.015
1.013
1.023
1.012
1.001
1.000
1.040
1.014
1.004
1.004
3.005
3.021


171.465
151.673
130.374
129.859
124.204
123.265
121.999
121.484
116.449
86.407
80.815
42.351
40.592
40.383
40.175
39.966
39.757
39.549
39.340
34.835
18.232
13.498
O
O
N
HO
5aa
13C (DMSO-d6, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

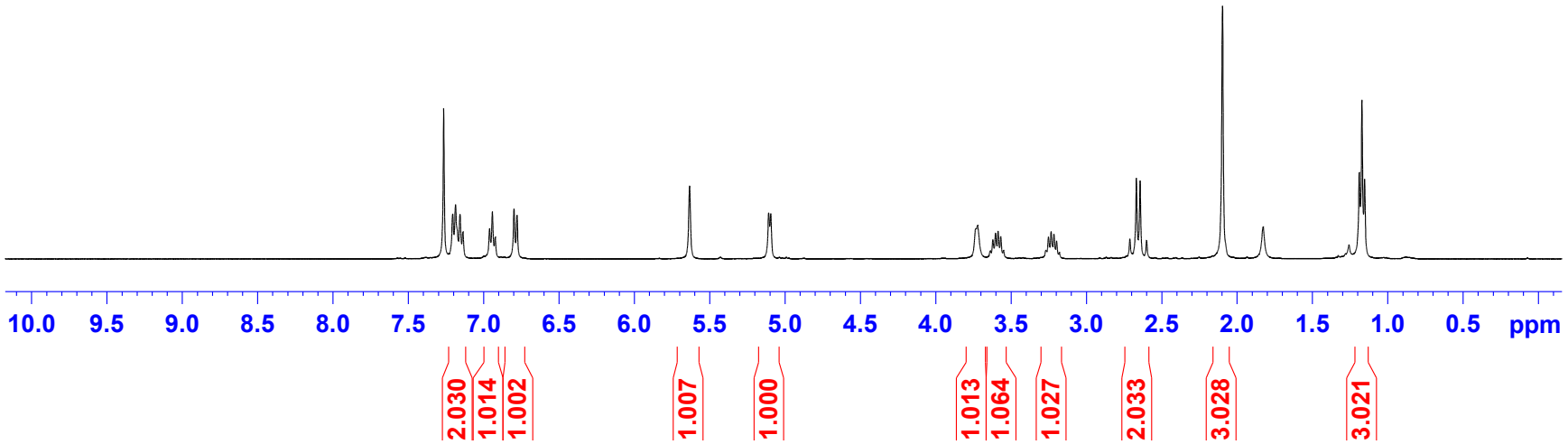

129.862
124.208
122.002
121.484
116.452
86.401
42.348
34.835
18.237
13.502
O
O
N
HO
5aa
DEPT-135
(DMSO-d_6, 100 MHz)
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

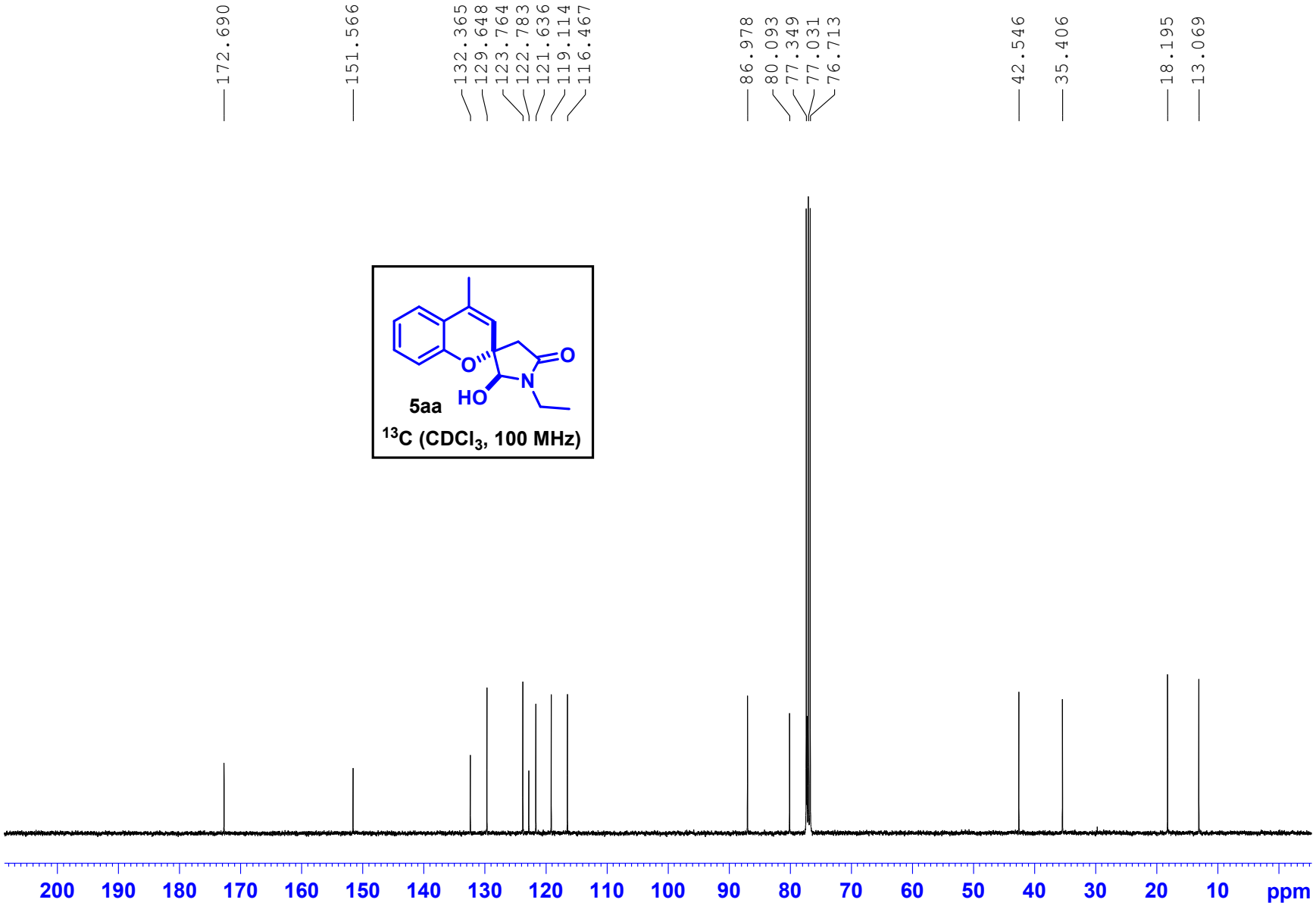


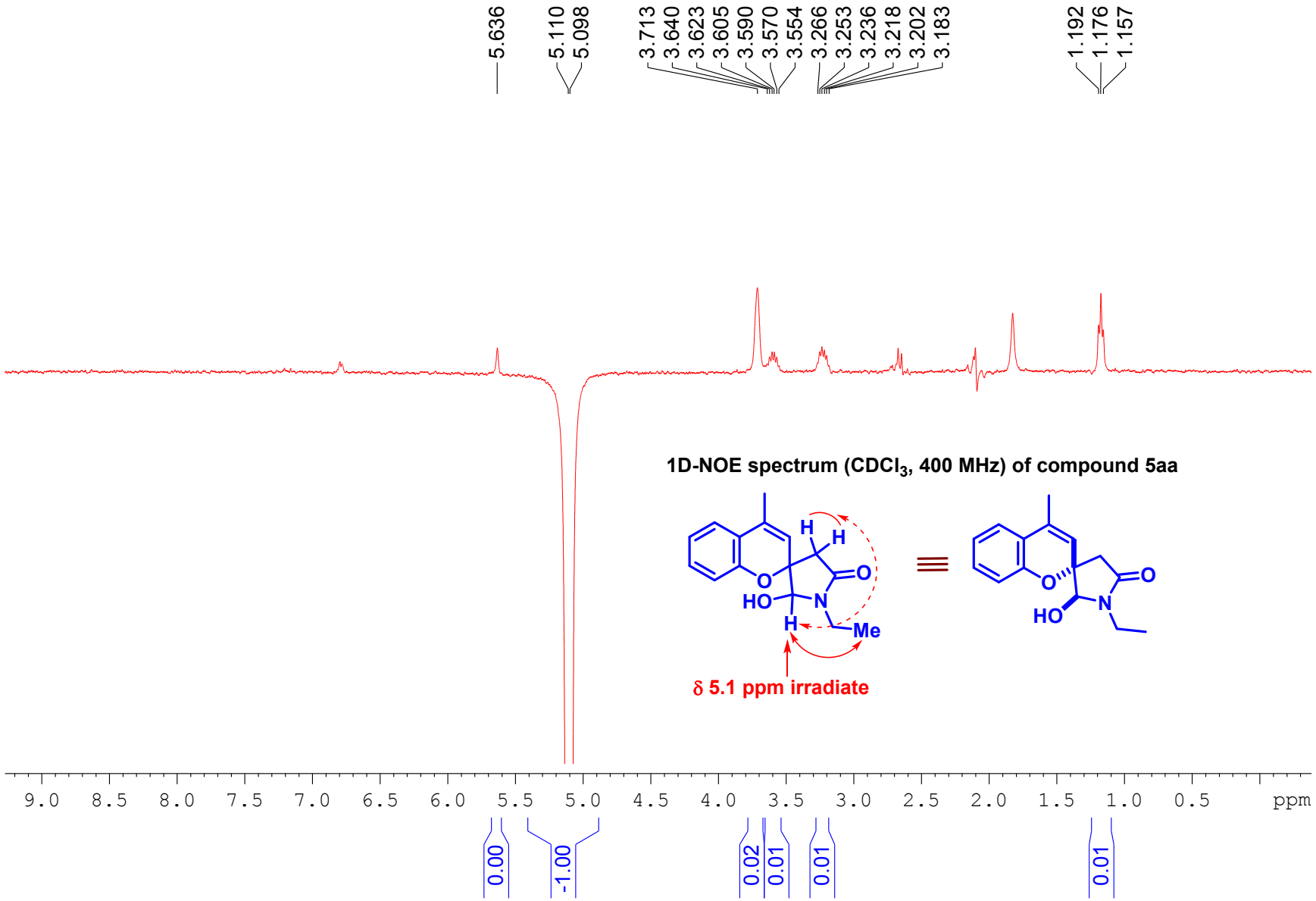


5aa

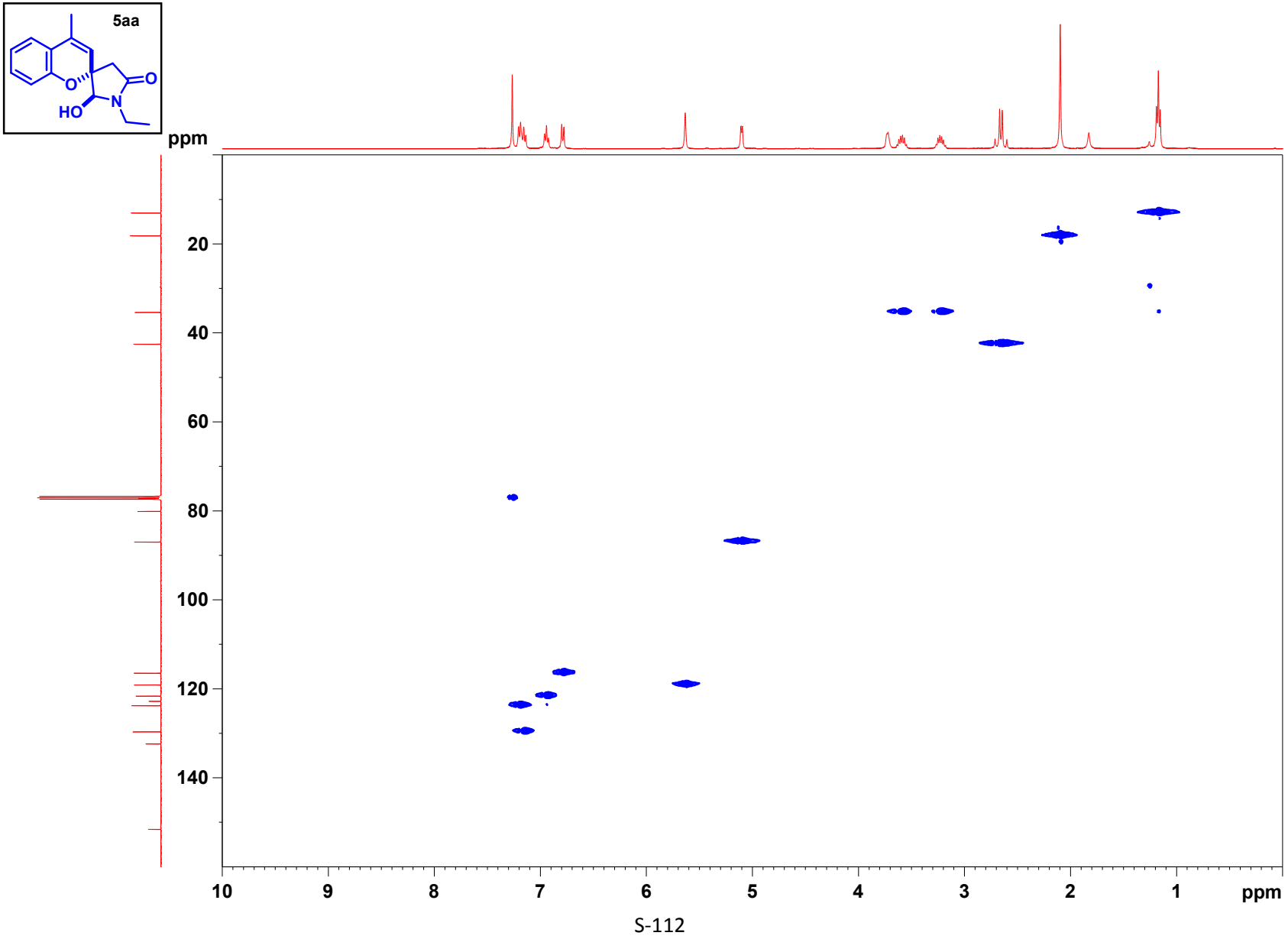
^{1}H ($CDCl_3$, 400 MHz)



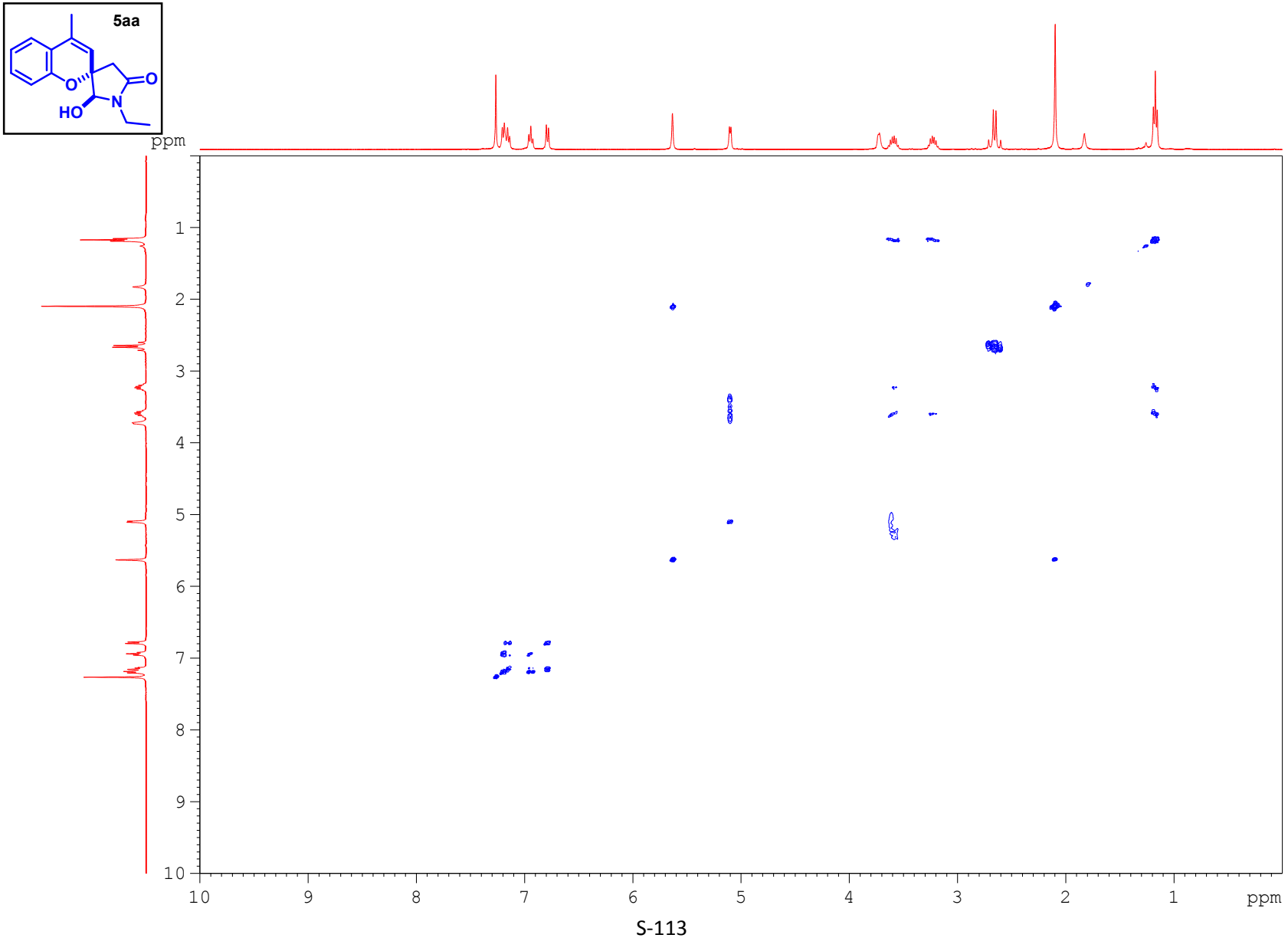

172.690
151.566
132.365
129.648
123.764
122.783
121.636
119.114
116.467
86.978
80.093
77.349
77.031
76.713
42.546
35.406
18.195
13.069
O
O
N
HO
5aa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


5.636
5.110
5.098
3.713
3.640
3.623
3.605
3.590
3.570
3.554
3.266
3.253
3.236
3.218
3.202
3.183
1.192
1.176
1.157
1D-NOE spectrum (CDCl3, 400 MHz) of compound 5aa
H
H
O
HO
N
H
Me
O
δ 5.1 ppm irradiate
O
O
HO
N
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
0.00
-1.00
0.02
0.01
0.01
0.01

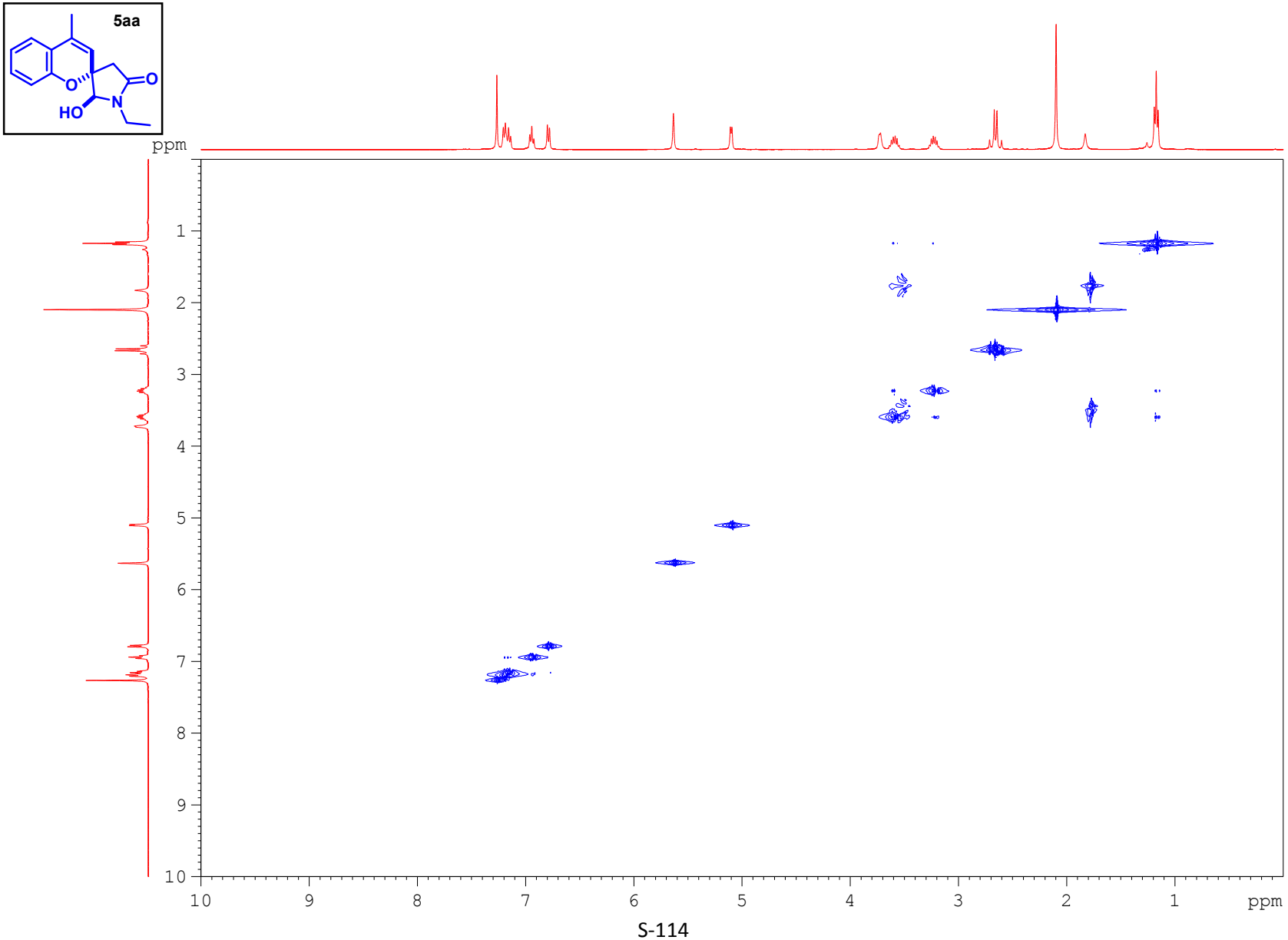
HSQC spectrum ($CDCl_3$, 400 MHz) of compound 5aa

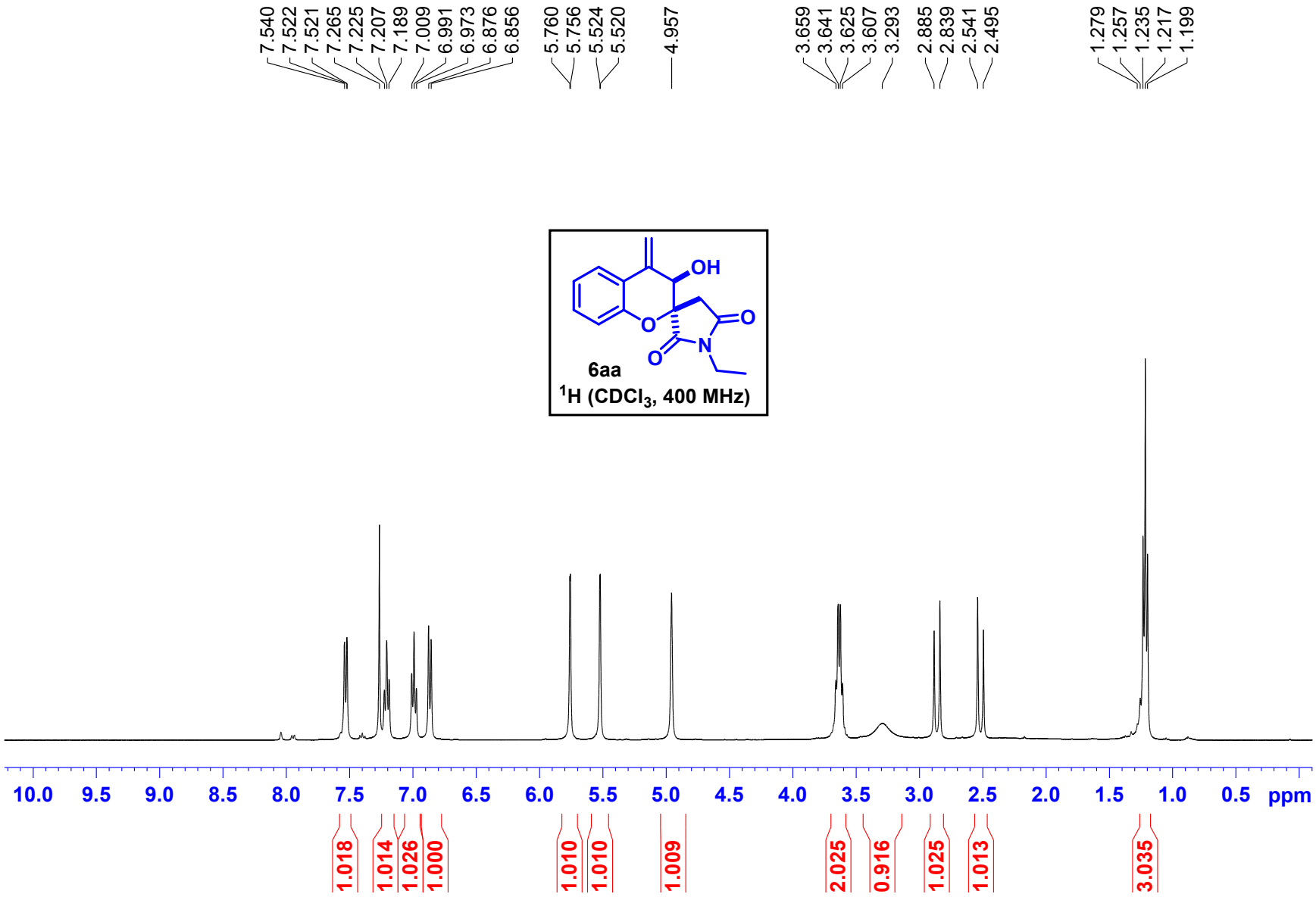


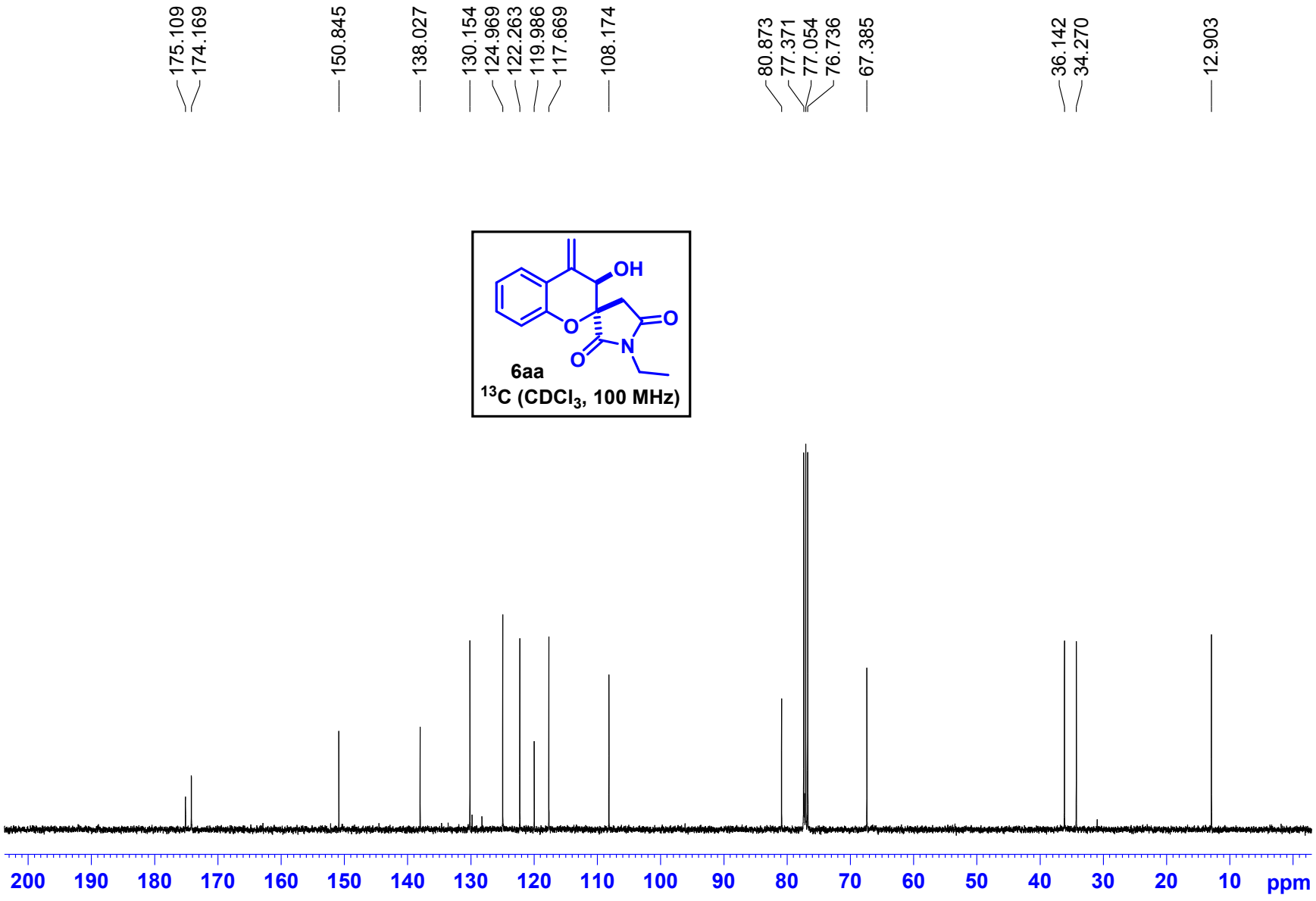
COSY spectrum ($CDCl_3$, 400 MHz) of compound 5aa

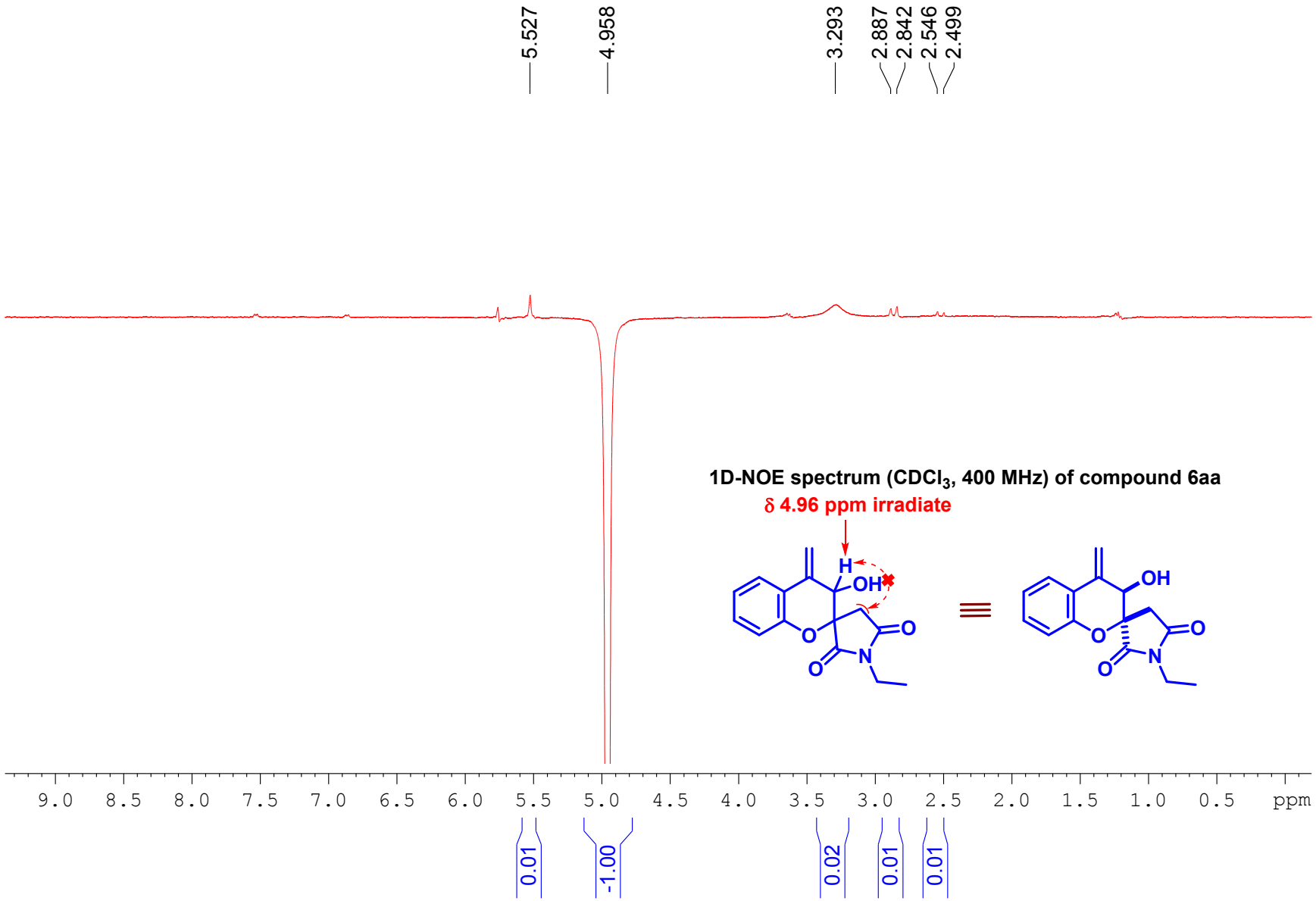


NOSY spectrum ($CDCl_3$, 400 MHz) of compound 5aa

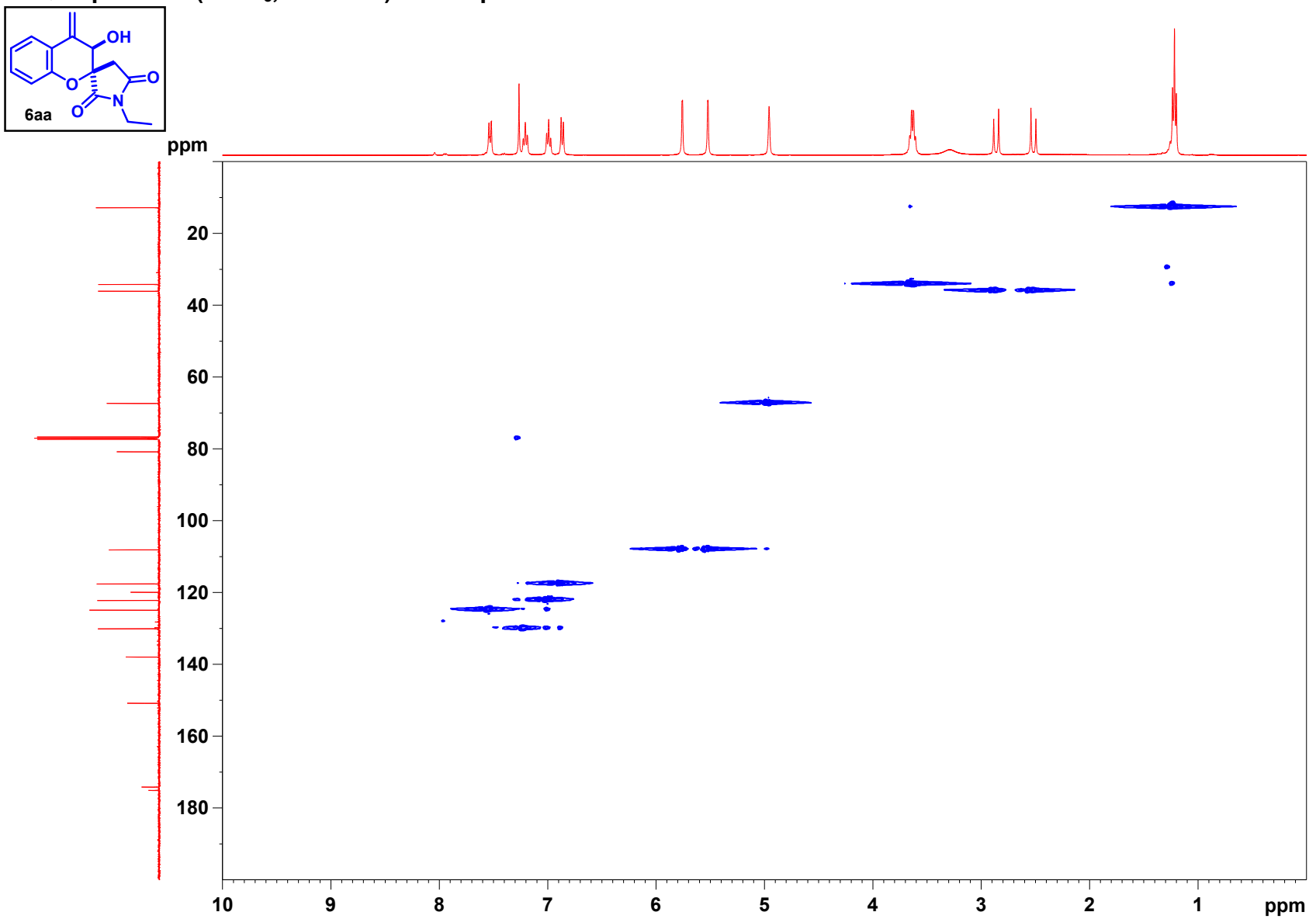



7.540
7.522
7.521
7.265
7.225
7.207
7.189
7.009
6.991
6.973
6.876
6.856
5.760
5.756
5.524
5.520
4.957
3.659
3.641
3.625
3.607
3.293
2.885
2.839
2.541
2.495
1.279
1.257
1.235
1.217
1.199
OH
O
O
O
N
6aa
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.018
1.014
1.026
1.000
1.010
1.010
1.009
2.025
0.916
1.025
1.013
3.035

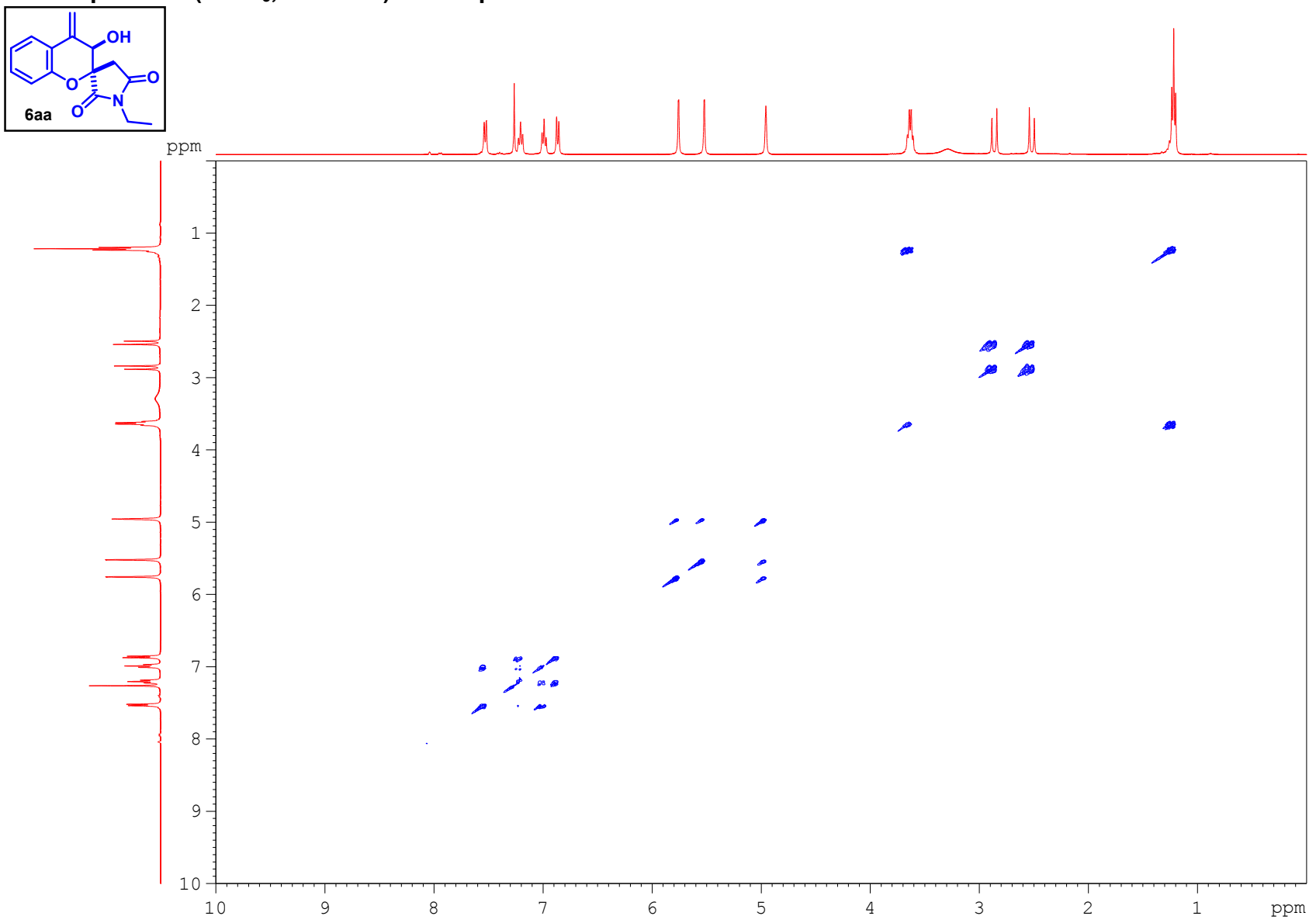

175.109
174.169
150.845
138.027
130.154
124.969
122.263
119.986
117.669
108.174
80.873
77.371
77.054
76.736
67.385
36.142
34.270
12.903
OH
O
O
N
O
6aa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


5.527
4.958
3.293
2.887
2.842
2.546
2.499
1D-NOE spectrum (CDCl3, 400 MHz) of compound 6aa
δ 4.96 ppm irradiate
H
OH
O
O
N
O
OH
O
O
N
O
9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 ppm
0.01
-1.00
0.02
0.01
0.01

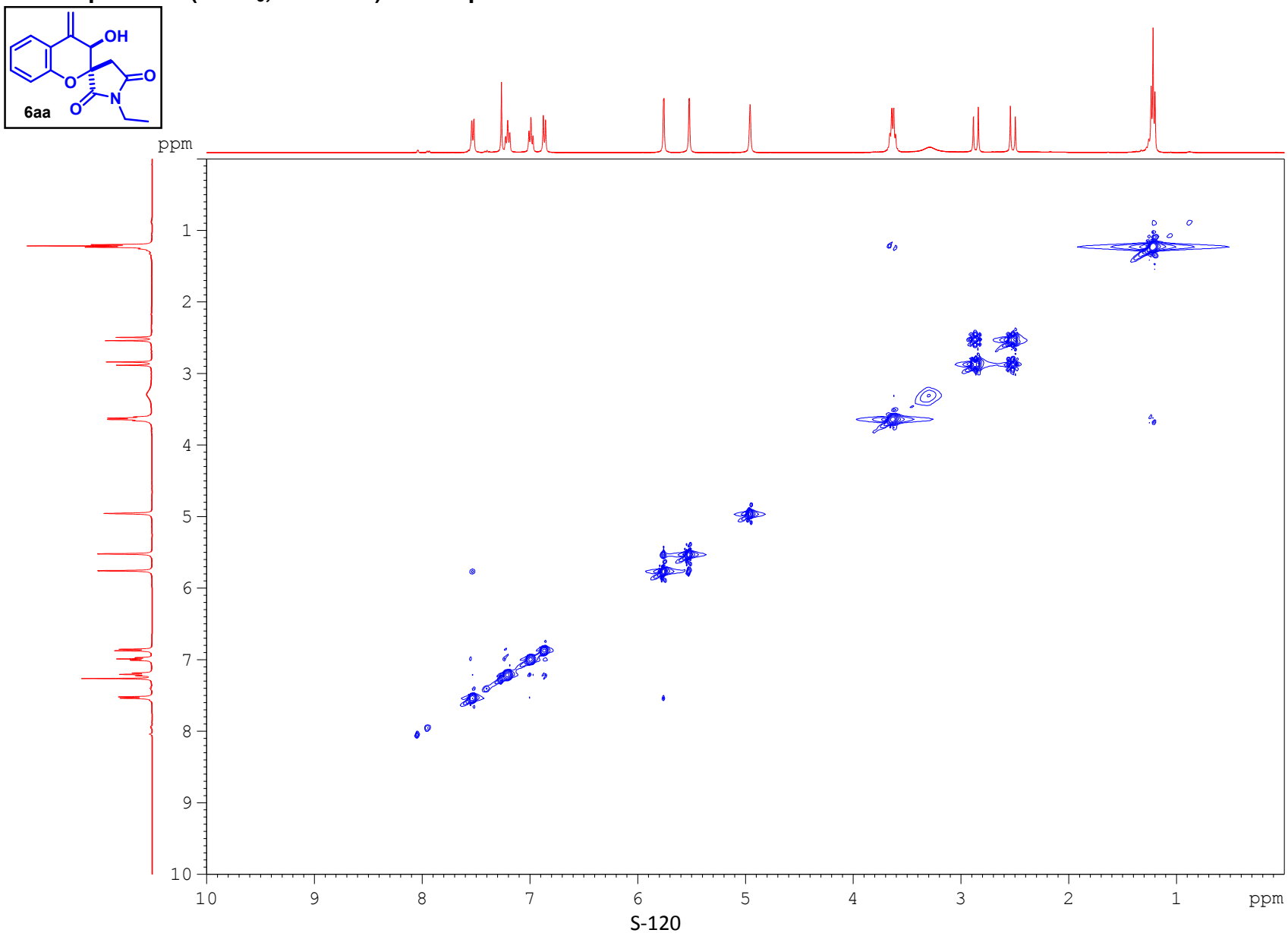
HSQC spectrum ($CDCl_3$, 400 MHz) of compound 6aa

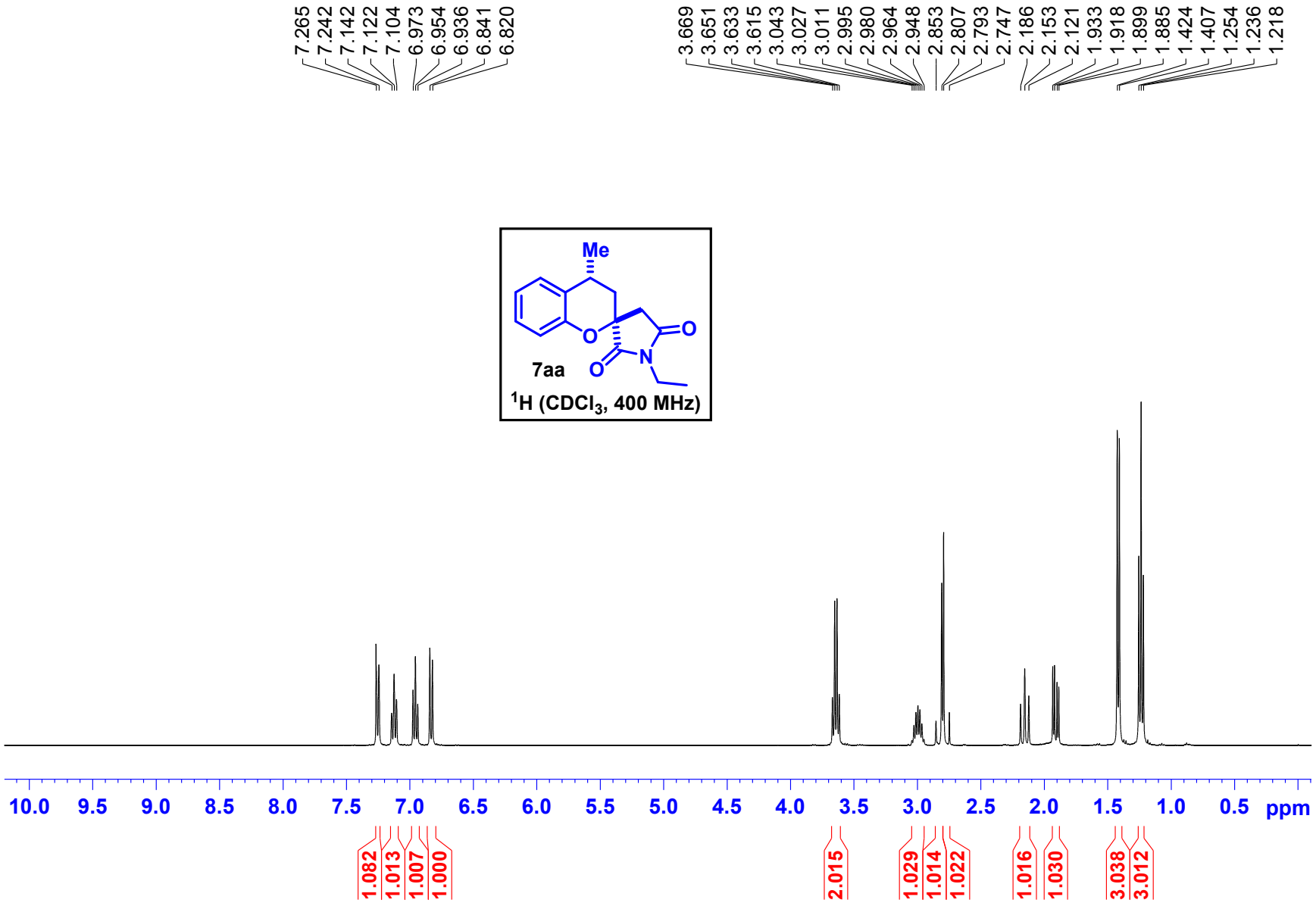


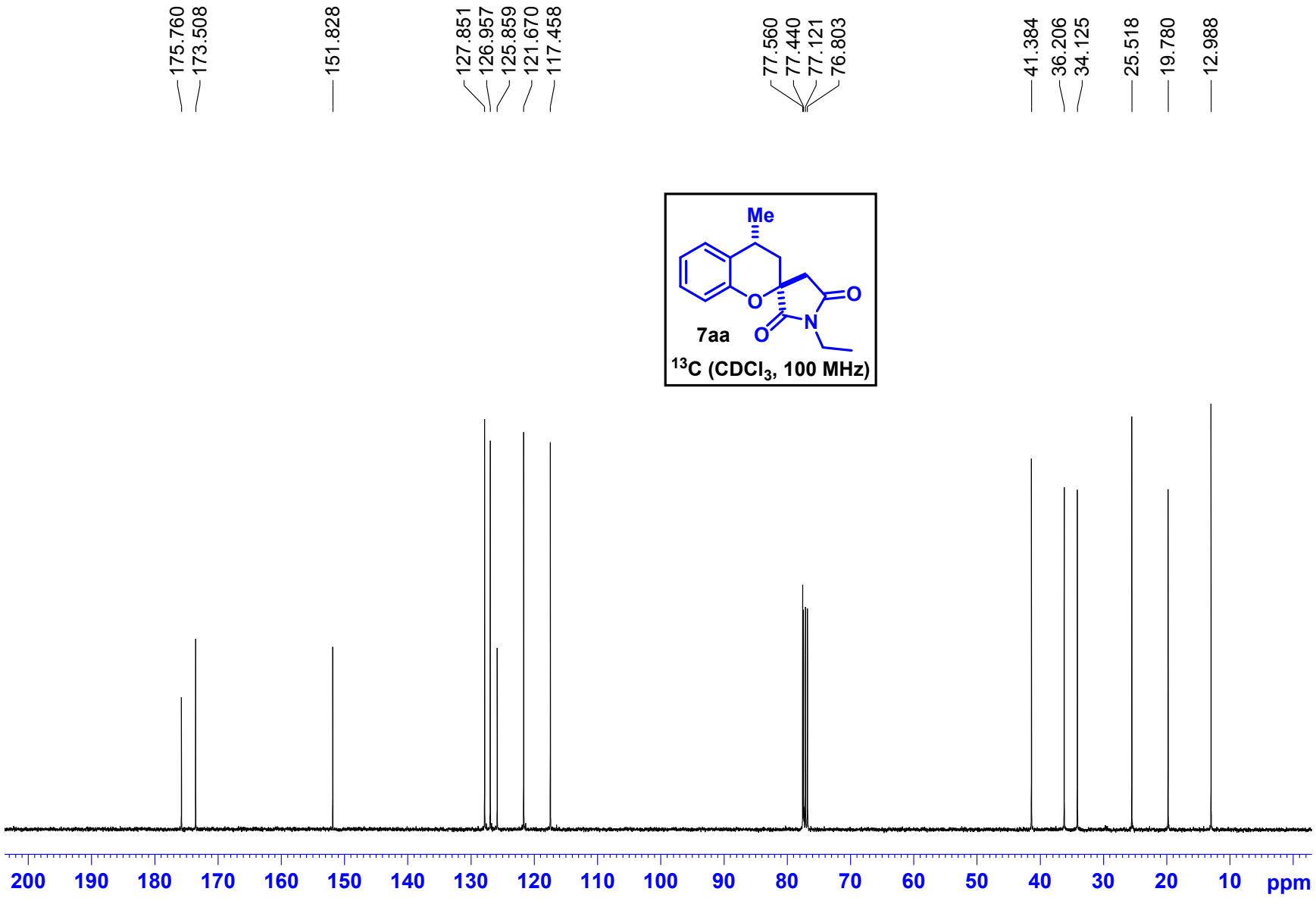
COSY spectrum ($CDCl_3$, 400 MHz) of compound 6aa

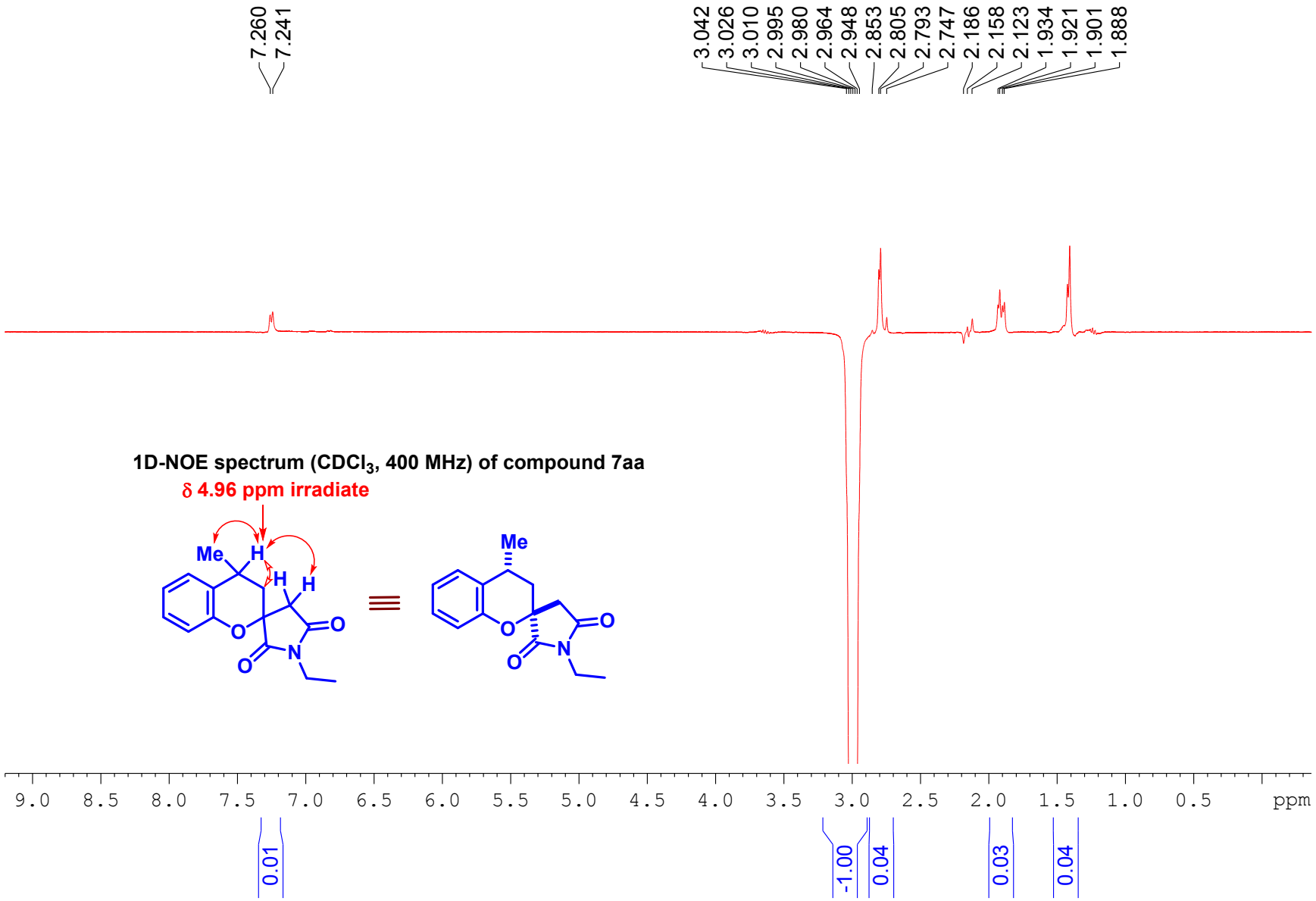


NOSY spectrum ($CDCl_3$, 400 MHz) of compound 6aa

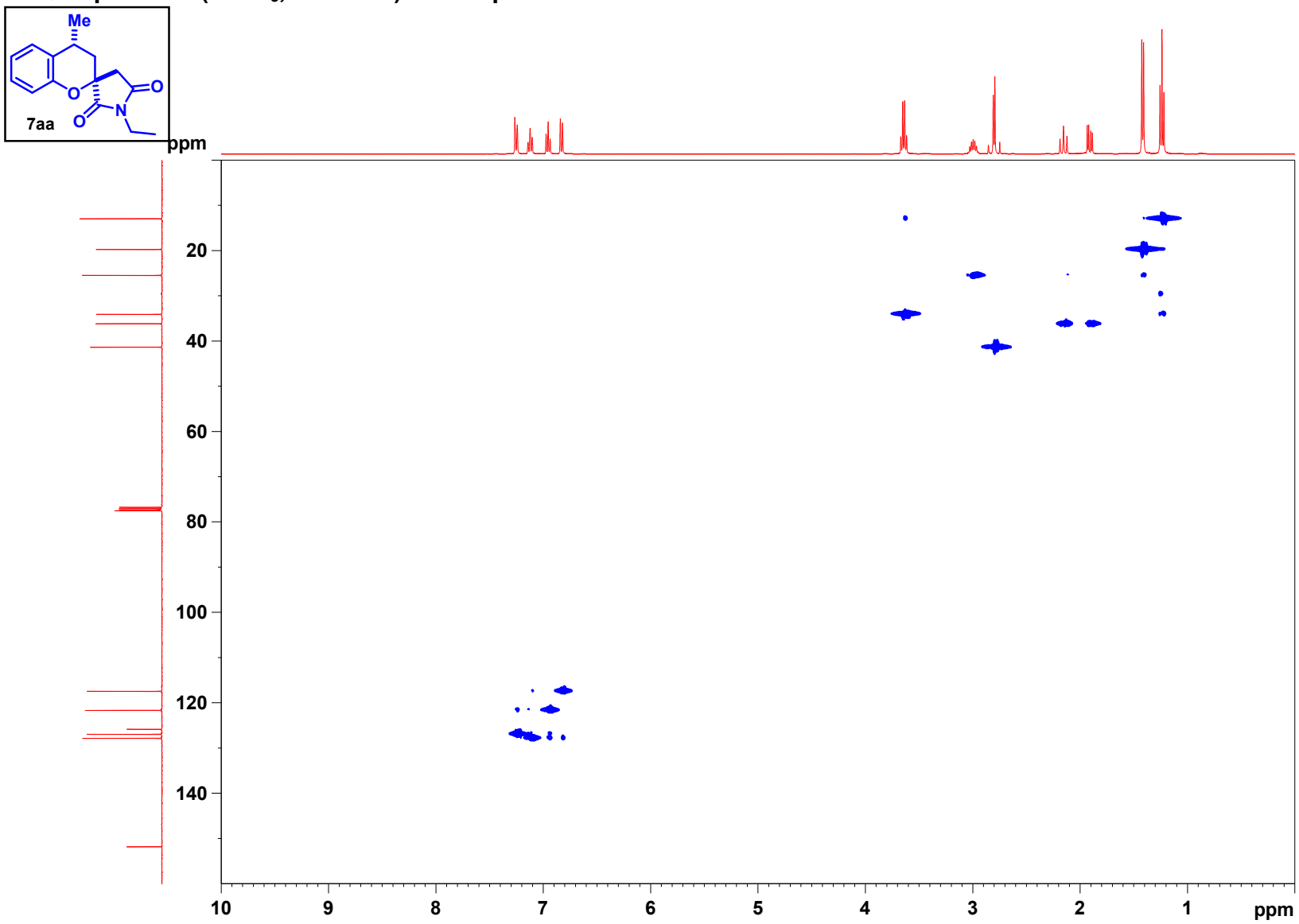



7.265
7.242
7.142
7.122
7.104
6.973
6.954
6.936
6.841
6.820
3.669
3.651
3.633
3.615
3.043
3.027
3.011
2.995
2.980
2.964
2.948
2.853
2.807
2.793
2.747
2.186
2.153
2.121
1.933
1.918
1.899
1.885
1.424
1.407
1.254
1.236
1.218
Me
O
O
N
O
7aa
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.082
1.013
1.007
1.000
2.015
1.029
1.014
1.022
1.016
1.030
3.038
3.012

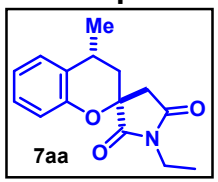

175.760
173.508
151.828
127.851
126.957
125.859
121.670
117.458
77.560
77.440
77.121
76.803
41.384
36.206
34.125
25.518
19.780
12.988
Me
O
O
N
O
7aa
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


7.260
7.241
3.042
3.026
3.010
2.995
2.980
2.964
2.948
2.853
2.805
2.793
2.747
2.186
2.158
2.123
1.934
1.921
1.901
1.888
1D-NOE spectrum (CDCl3, 400 MHz) of compound 7aa
δ 4.96 ppm irradiate
Me
H
H
H
O
O
N
O
Me
O
O
N
O
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
0.01
-1.00
0.04
0.03
0.04

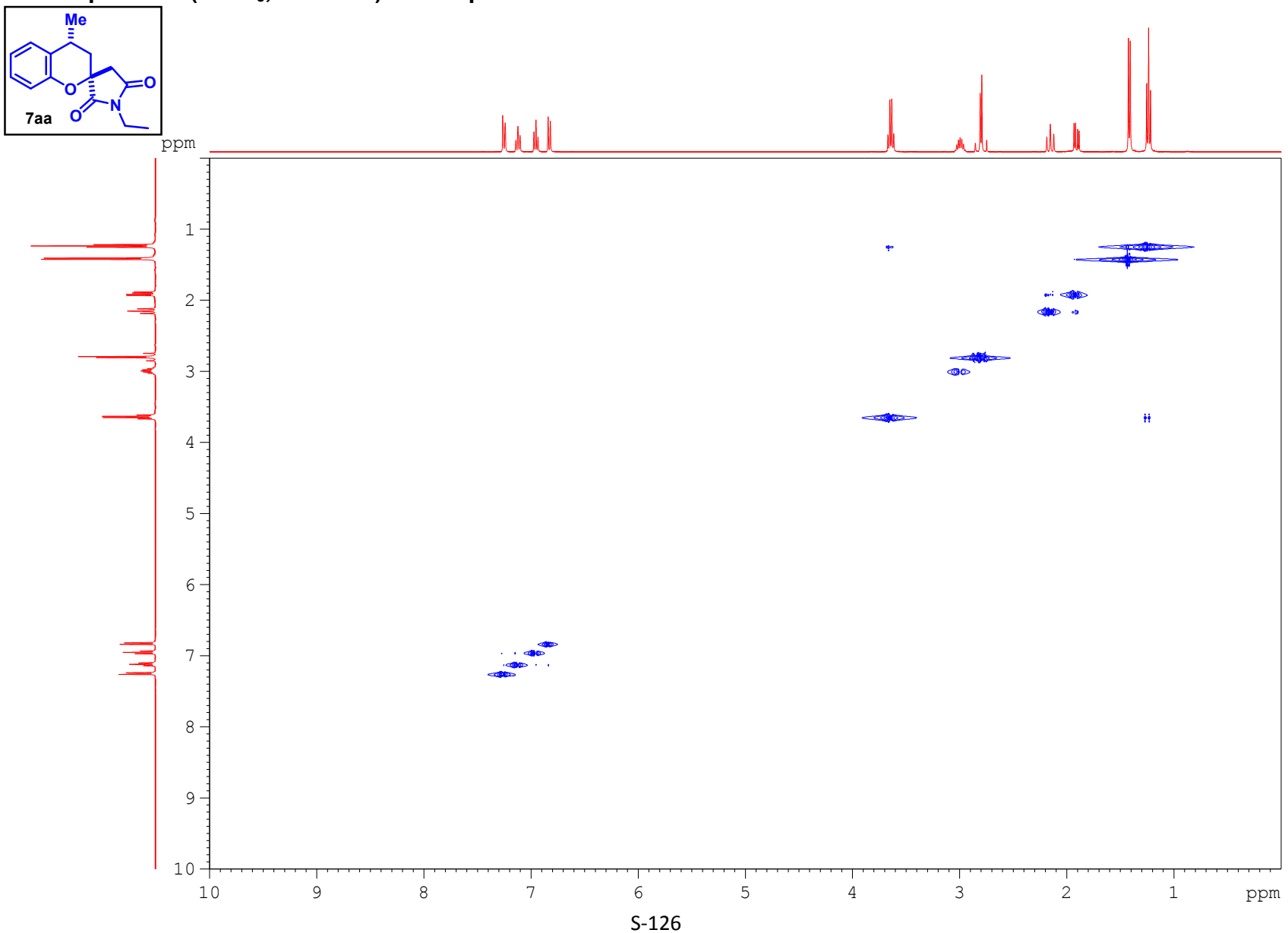
HSQC spectrum ($CDCl_3$, 400 MHz) of compound 7aa

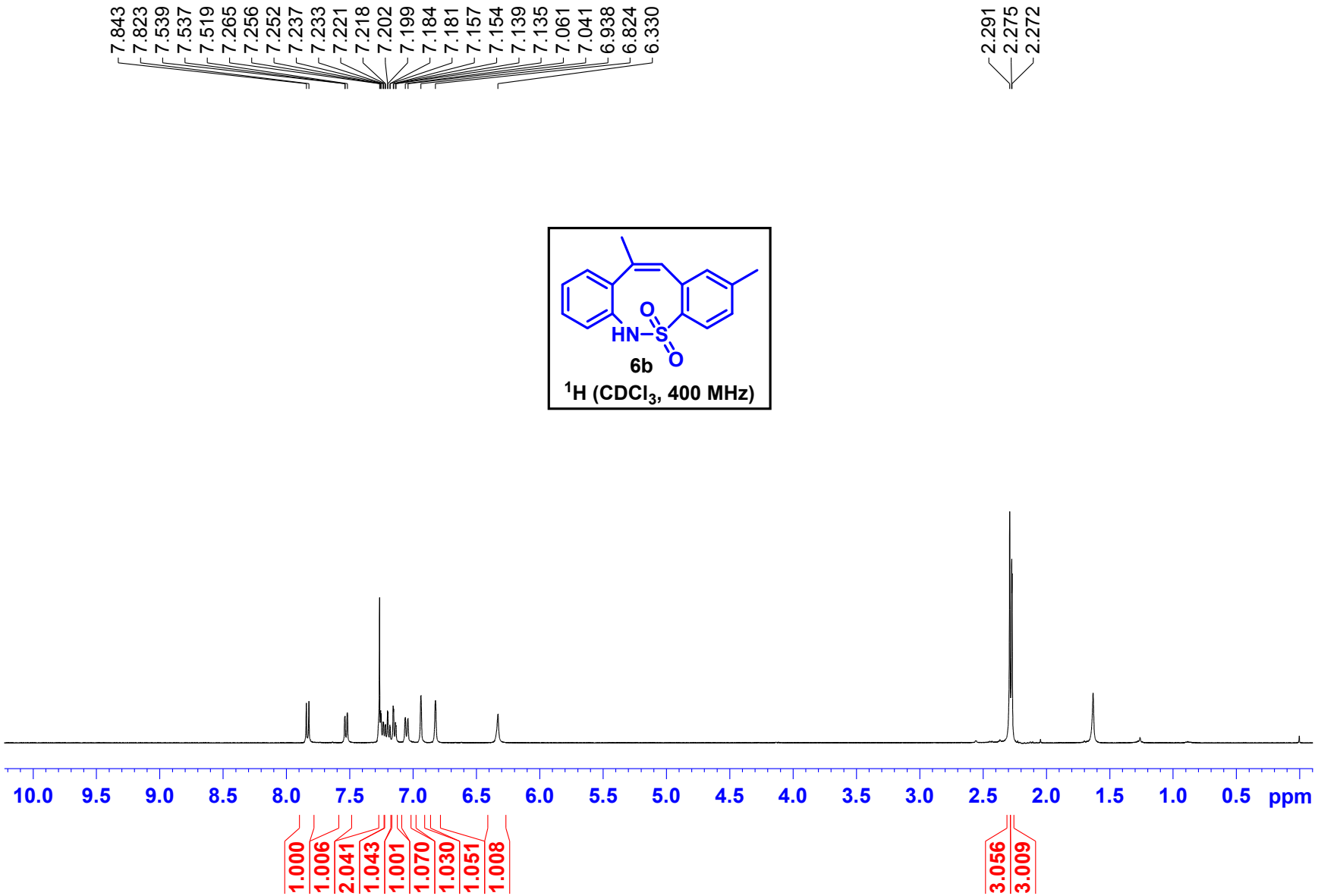


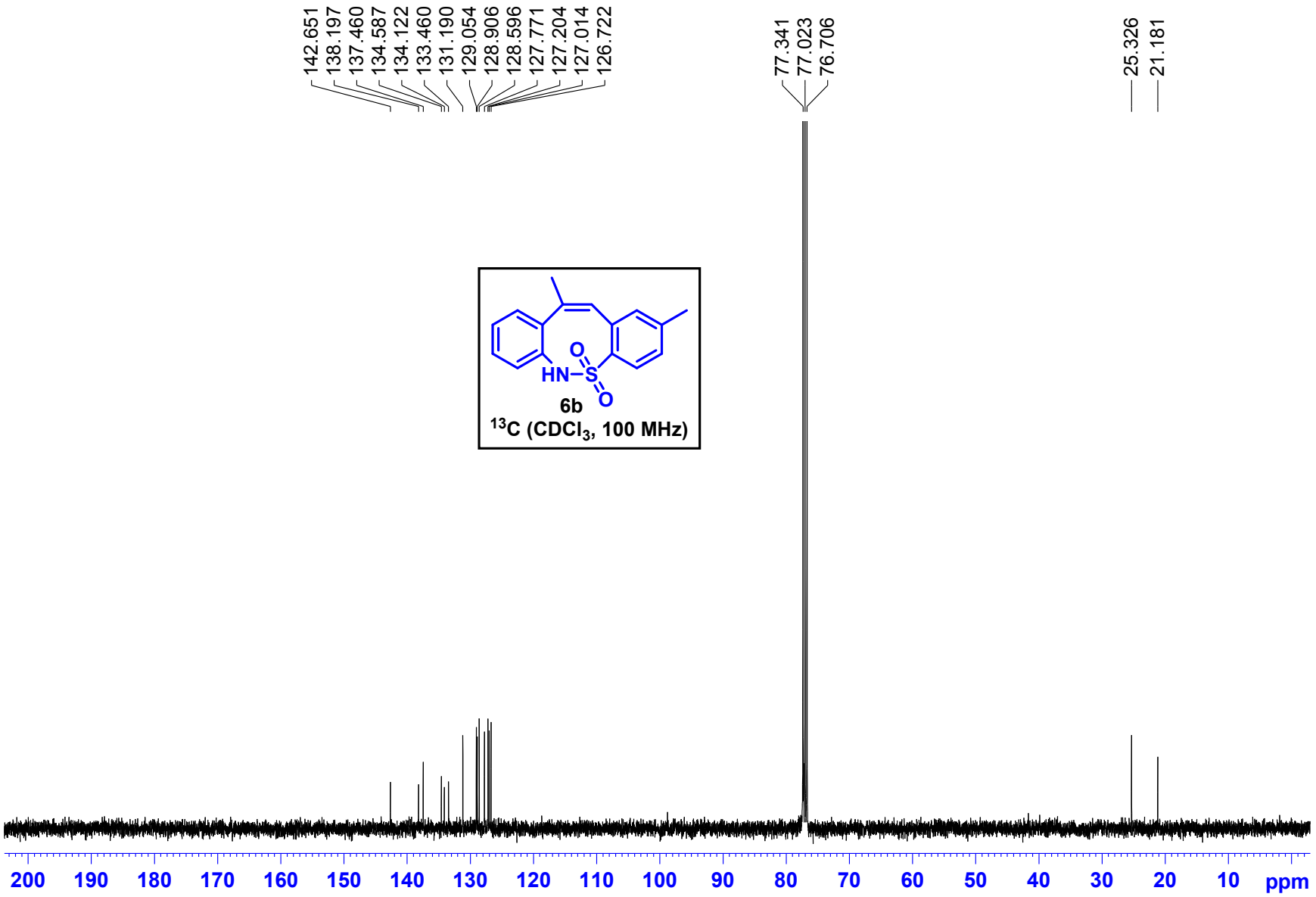
COSY spectrum ($CDCl_3$, 400 MHz) of compound 7aa

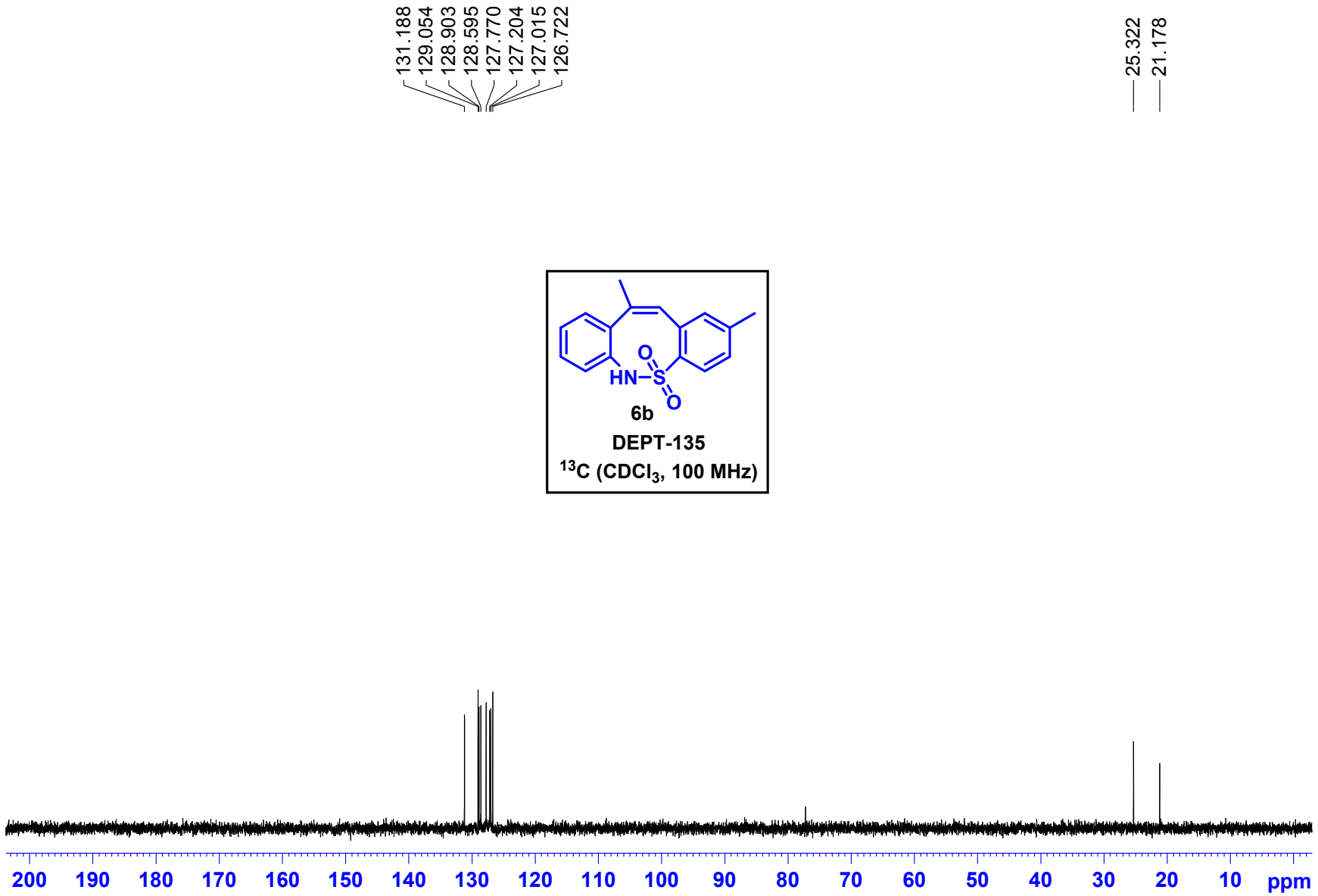


NOSY spectrum ($CDCl_3$, 400 MHz) of compound 7aa




7.843
7.823
7.539
7.537
7.519
7.265
7.256
7.252
7.237
7.233
7.221
7.218
7.202
7.199
7.184
7.181
7.157
7.154
7.139
7.135
7.061
7.041
6.938
6.824
6.330
2.291
2.275
2.272
HN
S
O
O
6b
1H (CDCl3, 400 MHz)
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
ppm
1.000
1.006
2.041
1.043
1.001
1.070
1.030
1.051
1.008
3.056
3.009


142.651
138.197
137.460
134.587
134.122
133.460
131.190
129.054
128.906
128.596
127.771
127.204
127.014
126.722
77.341
77.023
76.706
25.326
21.181
HN
S
O
O
6b
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm


131.188
129.054
128.903
128.595
127.770
127.204
127.015
126.722
25.322
21.178
O
HN–S
O
6b
DEPT-135
13C (CDCl3, 100 MHz)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm